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# **Gulf Stream rings may rival atmospheric iron supply to the North Atlantic subtropical gyre**

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1 **The North Atlantic subtropical gyre is a source region of biologically-available nitrogen**  
2 **to the global ocean, owing to nitrogen fixation by diazotrophic cyanobacteria<sup>1</sup>. These**  
3 **organisms have a high iron (Fe) requirement<sup>2</sup>, which is typically assumed to be satisfied**  
4 **in the North Atlantic by deposition of Fe-bearing Saharan dust<sup>3</sup>. However, dust supply**  
5 **is episodic, resulting in highly-variable Fe concentrations at the ocean's surface<sup>4,5</sup>,**  
6 **whilst Fe-depleted subsurface waters<sup>4,5</sup> limit supply from below. Here, we report the**  
7 **first observation of Fe in a Gulf Stream cold-core ring, where Fe concentrations were**  
8 **elevated compared to subtropical waters, reflecting the ring's origin in the Fe-rich Slope**  
9 **Sea. Using surrounding data from the GEOTRACES GA03 section and recent advances**  
10 **in satellite tracking of rings, we calculate that cold-core rings provide a net flux of**  
11  **$0.3 \pm 0.17 \times 10^8$  mol Fe yr<sup>-1</sup> across the north-western gyre edge, within the wide range of**  
12 **estimates of atmospheric Fe supply over the entire gyre area,  $0.4$ - $8.6 \times 10^8$  mol Fe yr<sup>-1</sup>**  
13 **(median  $2 \times 10^8$  mol Fe yr<sup>-1</sup>). By linking mesoscale ocean circulation to Fe supply, our**  
14 **results provide a new view of subtropical Fe dynamics that challenges the paradigm**  
15 **that atmospheric deposition persistently dominates subtropical North Atlantic iron**  
16 **supply.**

17

18 *Iron sources to the North Atlantic subtropical gyre.* Primary productivity in much of the  
19 Southern Ocean as well as the equatorial and sub-Arctic Pacific has been shown to be limited  
20 by iron (Fe)<sup>6,7</sup>. In contrast, in oligotrophic regions of the ocean such as the North Atlantic  
21 subtropical gyre (NASG), productivity is thought to be limited instead by macronutrients, and  
22 thus much less sensitive to Fe addition<sup>7,8</sup>. Owing to its proximity to the Sahara Desert, the  
23 North Atlantic receives the largest atmospheric dust fluxes globally<sup>3</sup>, resulting in dissolved  
24 Fe concentrations of up to 2 nmol kg<sup>-1</sup> in surface waters<sup>5</sup>. It has thus long been assumed that  
25 dust provides ample Fe for phytoplankton to utilise available macronutrients in this region.

26 However, the degree to which Fe from dust dissolves in seawater and is stabilised therein by  
27 organic ligands is widely debated<sup>9</sup>. Moreover, dust deposition is highly localised and  
28 episodic, varying dramatically with storm activity, location and season<sup>3,5,10</sup>, and may quickly  
29 overwhelm the capacity of seawater and organic ligands to maintain the supplied Fe in  
30 solution, leading to precipitation. In fact, dissolved Fe concentrations at the surface in the  
31 western gyre can be as low as 0.09 nmol kg<sup>-1</sup> during winter, with a potentially growth-  
32 limiting dissolved Fe minimum (0.02-0.20 nmol kg<sup>-1</sup>) present in subsurface waters (50-150  
33 m) year-round<sup>4,5</sup>. Background Fe concentrations of the gyre in the absence of dust deposition  
34 are also reproducibly low (Suppl. Info.). In addition to limiting nitrogen uptake in the North  
35 Atlantic<sup>11</sup>, a recent study has found that Fe may also limit phosphate acquisition by the  
36 microbial community in the North Atlantic in areas distal from the Saharan dust plume<sup>12</sup>.  
37 Therefore, all sources of Fe must be considered for their potential to fuel primary  
38 productivity within the gyre.

39

40 ***Gulf Stream rings.*** As early as the 1930s, scientists discovered boluses of anomalously cold  
41 water in the NASG and inferred that eddies must transport water from the Slope Sea, which  
42 lies between the continental shelf and the Gulf Stream, into the gyre<sup>13</sup>. These ‘cold-core  
43 rings’ are formed when a Gulf Stream meander becomes so large that it folds back onto itself,  
44 forming a loop that pinches off from the main current. These rings trap Slope Sea water, and  
45 are easily identifiable in satellite observations as they propagate into the subtropical gyre,  
46 characterised by a circular depression of sea-surface height. Although it has been proposed  
47 that eddy-driven, cross-Gulf Stream transport constitutes an important supply of phosphorus  
48 (P) to the subtropical gyre<sup>14</sup>, and it is well known that lateral processes in general play a role  
49 in supplying macronutrients to the gyres<sup>15-18</sup>, the effect of Gulf Stream rings on Fe transport  
50 has not been considered, largely due to a paucity of high-quality Fe data. Here, by combining

51 a recent satellite-derived dataset of mesoscale eddy activity<sup>19</sup> with a new dissolved Fe  
52 dataset<sup>4</sup> from a North Atlantic GEOTRACES section<sup>20,21</sup> (GA03; Fig. 1), we quantify the  
53 potential of Gulf Stream rings to supply Fe to the NASG. The GA03 dataset is ideal for this  
54 purpose, with two stations located in the Slope Sea, several within the NASG, and one station  
55 (USGT11-6) serendipitously situated at the edge of a Gulf Stream cold-core ring (Fig. 1).

56

57 ***Cross Gulf Stream iron transport.*** Slope Water is identifiable in the GA03 section by its low  
58 temperature and salinity (Figs 2-3), and is characterised by high concentrations of  
59 macronutrients<sup>21</sup> and CFCs<sup>20</sup>, reflecting upwelling of nutrient-rich water and contributions  
60 from shelf and Labrador Sea Water sources<sup>21,22</sup>. Slope Water is also greatly enriched in  
61 dissolved Fe compared to waters of equivalent density in the subtropical gyre<sup>4</sup> (Fig. 3; 0.64  
62 vs. 0.30 nmol kg<sup>-1</sup> above the 1026.5 isopycnal; Methods). These higher Fe concentrations  
63 have been attributed to a margin sediment source, which is important throughout the mid-  
64 depth (~600-2000 m) subtropical North Atlantic<sup>4</sup>. Within the subtropical gyre, however, a  
65 wedge of Fe-depleted Subtropical Mode Water (STMW)<sup>23</sup> separates this enriched mid-depth  
66 layer from the surface (Fig. 2). The cold-core ring sampled along GA03 transports Slope Sea  
67 water into the Fe-depleted gyre, with dissolved Fe concentrations 25% higher above the  
68 1026.5 isopycnal, and 60% higher above 500 m, than the open gyre (Figs 2-3).

69 The observation of a ring of Fe-rich Slope Water within the Fe-poor subtropical gyre  
70 suggests that rings could represent a significant source of Fe to the NASG, assuming the  
71 GA03 observations of the difference between gyre and Slope Sea Fe concentrations are  
72 characteristic for the region. Such an assumption is justified for a number of reasons, as  
73 discussed here and in more detail in the Supplementary Information. Firstly, the Fe-poor  
74 nature of the NASG waters is well documented; not only do the US GEOTRACES GA03  
75 zonal section<sup>4</sup> (2011) and the separate Dutch GEOTRACES GA02 meridional transect<sup>24</sup>

76 (2010) both show consistently low Fe concentrations through waters of the subtropical gyre,  
77 but three station reoccupations near Bermuda also provide evidence for the temporal stability  
78 of low Fe concentrations in the NASG over a period of three years<sup>25,26</sup>. The temporal  
79 variability of Slope Sea Fe concentration cannot be similarly directly assessed since the Fe  
80 concentration of its subsurface waters has not been previously reported. However, the  
81 propagation of characteristic sediment-derived Fe stable isotope compositions into the ocean  
82 interior<sup>4</sup> suggests that the Slope Sea Fe is relatively long-lived, consistent with the  
83 observation here of elevated Fe within a ring that was shed a number of weeks before  
84 sampling (Fig. 2). Furthermore, earlier work describes a qualitatively similar gradient of high  
85 Fe in very surface North American shelf waters decreasing into the open gyre<sup>27</sup>. At the basin  
86 scale, water column Fe datasets from reoccupied ocean stations in three other regions of the  
87 ocean, both with and without proximal sediment sources, also indicate the relative temporal  
88 stability of Fe profiles, similar to the macronutrients, at least on a sub-decadal  
89 timescale<sup>20,26,28</sup>. We thus feel confident in using the GA03 Fe data as sufficiently  
90 representative of the system in order to calculate the contribution of ring-driven transport to  
91 the Fe budget of the gyre.

92         This ring-driven Fe transport flux depends on: 1) the number of rings that cross the  
93 Gulf Stream per year, 2) the amount of dissolved Fe each ring carries, and 3) the fraction of  
94 this transported Fe that remains within the gyre rather than being re-entrained into the Gulf  
95 Stream. Recent progress in detecting and tracking eddies<sup>19,20</sup> makes the quantification of the  
96 ring-driven flux timely. We identified all Gulf-Stream-crossing cold-core rings in a  
97 database<sup>19</sup> of eddies detected in satellite altimetry data for 1993-2014 (Methods), and  
98 calculate that an average of  $7.7 \pm 2.5$  cyclonic rings cross the Gulf Stream each year (Suppl.  
99 Fig. 1), with an average surface area of  $3.9 \pm 1.5 \times 10^4$  km<sup>2</sup> (the equivalent of a circular vortex,  
100 radius  $111 \pm 70$  km). As the ring identified at station USGT11-6 was shed many weeks before

101 sampling (Methods) and its Fe inventory may thus have been affected by uptake, scavenging,  
102 or physical mixing of Fe, we chose not to use this ring as the endmember in our calculations.  
103 Instead, since cold-core rings enclose Slope Sea water pinched off by the Gulf Stream, we  
104 assumed that the average Fe concentration in Slope Water above the 1026.5 isopycnal  
105 ( $0.64 \pm 0.12$  nmol kg<sup>-1</sup>; stations USGT11-1 and 11-2) is representative of the initial dissolved  
106 Fe concentration in the core of the average cold core ring. We consider the water column  
107 above the 1026.5 isopycnal since we are interested in Fe that is accessible to NASG surface  
108 ecosystems over the annual cycle, and this isopycnal represents the density of the maximum  
109 NASG winter mixed layer.

110         If all rings dissipate entirely within the gyre, as assumed in an early study<sup>29</sup>, their  
111 near-surface Fe burden would ultimately enter the subtropical mixed layer. In this view, the  
112 volume flux due to rings entering the gyre is balanced by an equivalent volume transport out  
113 of the gyre, largely due to warm-core rings, with the average Fe concentration of the interior  
114 gyre above the 1026.5 isopycnal ( $0.30 \pm 0.10$  nmol kg<sup>-1</sup>). Given the  $0.34$  nmol kg<sup>-1</sup> Fe  
115 concentration difference and the ring statistics from satellite altimetry, the assumption of  
116 complete dissipation leads to a ring-driven Fe supply to the subtropical gyre of  $0.3 \pm 0.17 \times 10^8$   
117 mol dissolved Fe yr<sup>-1</sup>, accompanied by a dissolved phosphate supply of  $1.3 \pm 0.8 \times 10^{10}$  mol  
118 year<sup>-1</sup>; Methods).

119         Our estimate of the ring-driven Fe and phosphate supply may be considered an upper  
120 limit, given that rings can be re-entrained into the Gulf Stream after only partial dissipation<sup>30</sup>.  
121 In these cases, only a fraction of the nutrients within the ring may be biologically consumed  
122 within the ring and/or mixed laterally with surrounding subtropical waters, before being re-  
123 entrained. A conservative estimate for the ring-driven nutrient supply is provided by  
124 assuming that nutrients become available only after physical dissipation of the rings, which  
125 assumes no biological consumption in the ring before re-entrainment. Assuming a lateral



126 diffusivity of  $300 \text{ m}^2 \text{ s}^{-1}$ , as diagnosed from float observations of analogous cold-core rings  
127 shed from the Kuroshio Extension<sup>30</sup>, half of the volume-integrated Fe anomaly in the ring  
128 would be mixed with surrounding waters within two months, and  $\leq 15\%$  of the integrated Fe  
129 would remain at the end of one year (Methods; ED Fig. 2). With ring lifetimes thought to  
130 average more than a year<sup>31</sup>, we estimate that accounting for re-entrainment of rings into the  
131 Gulf Stream after only partial dissipation would reduce our estimate by 15% at most.  
132 Additionally, rings are not the only processes that can transport Fe and nutrients across the  
133 Gulf Stream. Rather, a suite of mesoscale processes, including but not limited to rings, can  
134 mix nutrients down-gradient from the relatively high concentrations of the Slope Water into  
135 the NASG. We estimate that the total Fe supply due to down-gradient mixing across the Gulf  
136 Stream may be up to seven times larger than our ring-derived estimates (Methods).

137

138 ***Comparison of ring-derived fluxes with atmospheric supply.*** How does the ring-driven flux  
139 of soluble Fe to the NASG compare to that delivered by atmospheric deposition? Answering  
140 this question requires a robust quantification of atmospheric Fe fluxes, which are highly  
141 uncertain. Drawing upon a wide variety of independent observation- and model-based  
142 estimates, we have established a representative range of gyre-wide atmospheric Fe deposition  
143 fluxes to the NASG (Methods; Suppl. Info). A key uncertainty for the biogeochemical  
144 relevance of atmospheric Fe deposition is the solubility of the Fe delivered to the surface  
145 ocean. Different studies approach this question differently. For example, some models  
146 included in our compilation include Fe as a prognostic variable and directly simulated its  
147 solubility; in these, Fe solubility in the dust deposited to the NASG ranged between  $\sim 1\text{-}2\%$   
148 (Table S1). In other studies, where only total Fe deposition was reported, we calculated  
149 soluble Fe delivery by assuming an appropriate upper bound of 5% solubility (see a more  
150 detailed discussion in Supplementary Information). The resulting range of atmospheric

151 soluble Fe fluxes over the entire area of the subtropical gyre spans a factor of more than 20,  
152 from  $0.4\text{-}8.6\times 10^8$  mol Fe yr<sup>-1</sup> (Fig. 4), with a median of  $2\times 10^8$  mol Fe yr<sup>-1</sup>. Our estimate of  
153 ring-driven soluble Fe transport across one boundary of the NASG thus represents between  
154 3% and 75% of the range of estimated atmospheric deposition fluxes over the entire gyre  
155 surface, or 15% of the median deposition flux (Fig 4). In the face of the uncertainty of the  
156 magnitude of atmospheric soluble Fe supply, this result indicates that ring-driven transport of  
157 Fe across the Gulf Stream is quantitatively important for Fe supply to the gyre. This is  
158 especially true for the north-western gyre, since the westward propagation of the rings means  
159 that they are most likely to dissipate within the north-western gyre. A scale analysis suggests  
160 that this ring-driven supply of Fe may in fact be the dominant mechanism of Fe supply to this  
161 region if the rings dissipate within ~1000 km of the Gulf Stream (Fig. 4). Such a significant  
162 role for ocean circulation in Fe supply via lateral transport represents a paradigm shift in our  
163 understanding of sources of Fe to the subtropics, and parallels recognition of the importance  
164 of lateral transport for macronutrient budgets of subtropical gyres<sup>15,18</sup>. Our quantification of  
165 the ring-driven Fe flux suggests that such transport processes must be included in Fe budgets  
166 and models of ocean biogeochemistry.

167

168 ***Implications for gyre biogeochemistry.*** Two other factors combine to reinforce the  
169 importance of cross-Gulf Stream Fe transport for the subtropical Fe budget and the  
170 biogeochemistry of the NASG. First, Slope Water contains a higher excess of Fe-binding  
171 ligands than the open surface gyre<sup>32</sup>. Not only does this mean that a higher percentage of the  
172 dissolved Fe coming from the Slope Sea may be bioavailable compared to dust-derived Fe,  
173 but a supply of excess ligands could also enhance *in situ* Fe dissolution from dust as well as  
174 stabilising the Fe thus released<sup>33</sup>. Second, ring-driven Fe supply is accompanied by a supply  
175 of the macronutrients nitrate and phosphate to the gyre, and, importantly, as previously

176 noted<sup>14</sup>, by an excess supply of phosphate relative to nitrate (Fig. 2d; Methods). If rings also  
177 carry an excess of Fe over nitrate relative to the physiological requirements of non-  
178 diazotrophic phytoplankton, this could create an ecological niche for diazotrophs<sup>34</sup>.  
179 Resource-competition theory<sup>35</sup> suggests that in order for ring-driven transport to support  
180 diazotrophy in the NASG, the supply of Fe and P relative to N should exceed their relative  
181 requirement by non-diazotrophic phytoplankton<sup>31</sup>.

182 Here, our calculations yield a PO<sub>4</sub>:NO<sub>3</sub> ratio of ring-driven transport below 1:15, i.e.  
183 an excess of PO<sub>4</sub> relative to the Redfield ratio of 1:16, consistent with previous results  
184 suggesting excess PO<sub>4</sub> supply to the NASG<sup>14,36</sup>. Therefore, the surplus PO<sub>4</sub> in rings should  
185 become available to diazotrophs following exhaustion of the NO<sub>3</sub> supply by non-  
186 diazotrophs. The corresponding Fe:NO<sub>3</sub> ratio in the rings is ~1:8800, or 0.11 mmol mol<sup>-1</sup>.  
187 Cell quota studies<sup>37</sup> suggests that non-diazotrophic phytoplankton cells have Fe:N ratios of  
188 0.06-0.31 mmol mol<sup>-1</sup> N (reported Fe:PO<sub>4</sub> ratios of 1-5 mmol mol<sup>-1</sup>, and converted assuming  
189 a Redfield N:P ratio). If the non-diazotrophic assemblage is comprised of cells at the low-  
190 end of this range, then the ring-driven nutrient supply could support diazotrophy. Otherwise,  
191 rings would be expected to leave surplus PO<sub>4</sub>, but little Fe, behind after non-diazotrophic  
192 exhaustion of NO<sub>3</sub>, priming the system for N<sub>2</sub> fixation in response to atmospheric Fe  
193 deposition events.

194 We thus speculate that, dependent on the microbial and phytoplankton assemblage,  
195 and together with the stabilizing and solubilizing effect discussed above, ring-driven Fe  
196 supply may contribute to the support of diazotrophy in the NASG, whilst also potentially  
197 influencing P acquisition<sup>12</sup>. Even if ring transport of Fe does not support diazotrophy directly,  
198 ring transport of excess phosphate would still be expected to support diazotrophy in response  
199 to atmospheric Fe deposition events. More broadly, eddy-driven transport of Fe may be  
200 important in other similarly dynamic regions of the oceans, including the South Atlantic,

201 where Agulhas rings carry elevated Fe concentrations into the subtropics<sup>26</sup>, and the North  
202 Pacific, where Haida eddies carry Fe from Alaskan shelf waters to the Fe-limited open  
203 ocean<sup>38</sup>. Patchy supply of Fe and macronutrients by eddies may thus have a significant effect  
204 on local and regional biogeochemistry, playing an important and often-overlooked role in  
205 primary productivity, nitrogen fixation and carbon cycling within oligotrophic gyres.

206

## 207 **Methods**

208

209 **Data availability.** GA03 dissolved Fe concentration data shown in Figs. 2-3 and used in  
210 calculations are taken from Conway and John<sup>4</sup>. Supporting data for macronutrients, salinity  
211 and temperature along GA03 are reproduced from the Ocean Data Facility<sup>21</sup> and all GA03  
212 data are freely available<sup>20</sup> in the GEOTRACES Intermediate Data Product 2014.

213

214 **Satellite-derived eddy database.** Faghmous *et al.*<sup>19</sup> provide a database of eddies detected in  
215 satellite altimetry, compiled for the years 1993-2014. In this database, a cyclonic eddy is  
216 defined as the outermost closed contour of altimetric sea level anomaly (SLA) containing a  
217 single minimum in sea level. This minimum is defined as a grid cell whose SLA is less than  
218 its surrounding 24 neighbouring grid points (on a 5×5 grid), where each side of a grid box is  
219 0.25° in latitude or longitude. To track the eddies over time, all eddies in the next day SLA  
220 image within a geographical boundary are checked to see whether there is an eddy that  
221 qualifies to be stitched to the current day's eddy to form a track. This geographical boundary  
222 accounts for westward propagation at the Rossby phase speed.

223

224 **Cross-Gulf Stream ring identification.** Using the Faghmous *et al.*<sup>19</sup> database, we identified  
225 and recorded the size of cold-core rings that cross the climatological Gulf Stream position

226 into the subtropical gyre as follows: 1) Construct a monthly climatology of sea surface height  
227 (SSH) over the satellite record; 2) Interpolate the monthly mean climatological SSH along  
228 each cyclonic eddy track in the vicinity of the Gulf Stream; 3) Tag any eddies that cross from  
229 north of the climatological Gulf Stream position (regions with SSH below 0.52 m from the  
230 AVISO absolute dynamic topography) to the subtropical side of the Gulf Stream (SSH  
231 greater than 0.55 m) and persist as a coherent track for at least three weeks.

232 We compared the performance of this objective identification against an ad-hoc visual  
233 assessment for a random subset of the satellite record (comprising 8 years and 66 eddies –  
234 more than a third of all cold-core rings from the final record). For two examples of this ad-  
235 hoc visual assessment, see Suppl. Animation 1 for a clear example of a cold core ring  
236 penetrating into the subtropical gyre from 1993, and Suppl. Animation 2 for the ring sampled  
237 at station USGT11-6 from 2011. The eddies identified by the algorithm generally have the  
238 expected characteristics of a cold-core ring: they are formed from a steep meander of the Gulf  
239 Stream, shed to the south, and propagate westward. Moreover, the eddy amplitude (i.e. the  
240 absolute value of the sea level anomaly along the track) is 46 cm, further verifying that these  
241 rings travel across the position of the climatological Gulf Stream and are, thus, large negative  
242 anomalies relative to the average subtropical sea-surface height. Furthermore, our estimate of  
243 ring number,  $7.7 \pm 2.5$  rings  $\text{yr}^{-1}$  (Suppl. Fig. 1), is within the uncertainty bounds of earlier  
244 work that counted the number of rings identified by cold anomalies in satellite sea surface  
245 temperature snapshots and divided by an estimate of their lifetime<sup>39</sup>.

246

247 ***Estimation of dissolved Fe anomaly in a ring.*** We estimated the dissolved Fe concentration  
248 within a cold-core ring based on the Slope Sea stations from the GA03 Section dataset<sup>4</sup>. To  
249 do this, we calculated the depth integrated average Fe concentration observed in near-surface  
250 layers (i.e. in and above the density of the STMW, isopycnal 1026.5). The 1026.5 isopycnal

251 was at depths of ~100 m in the Slope Sea, deepening to 350 m in the open gyre (Fig. 2), and  
252 is taken as the deepest relevant depth of Fe that may become available for productivity in the  
253 gyre, since this isopycnal represents the maximum winter mixed layer density in the gyre<sup>40</sup>.  
254 Taking data from 0-100 m at stations USGT11-1 and -2 within the Slope Sea gave an average  
255 of 0.64 nmol kg<sup>-1</sup> for the Slope Sea, with a standard deviation of 0.12 nmol kg<sup>-1</sup>. Similarly,  
256 taking the mean of data from 0 to 350 m at GA03 stations USGT11-8 and USGT11-10, to  
257 represent the interior of the gyre, gave 0.30 nmol kg<sup>-1</sup>, with a standard deviation of 0.10 nmol  
258 kg<sup>-1</sup>. We excluded the outlier top data point at Station 8 (1.2 nmol kg<sup>-1</sup>) from the average to  
259 preclude any effect from recent dust deposition or contamination biasing the gyre average.  
260 The calculated Fe anomaly ( $\Delta\text{Fe}$ ) within an average cold-core ring is thus assumed to be the  
261 difference between these numbers (0.34 nmol kg<sup>-1</sup>). We chose not to use the average  
262 observed ring concentrations from above the 1026.5 isopycnal or above 500 m at USGT11-6  
263 (0.37 and 0.47 nmol kg<sup>-1</sup> respectively) as the Slope Water end member, because it is clear  
264 from SSH data that (a) station USGT11-6 was at the edge of the ring and (b) the ring was  
265 shed about 5–6 weeks before sampling, and thus has presumably lost significant Fe due to  
266 scavenging, biological uptake, and mixing with subtropical waters, as suggested by the strong  
267 interleaving seen in Fig. 3 (Fe dissipation estimate below).

268

269 ***Calculation of ring-driven Fe transport.*** We calculated the total ring-driven supply of Fe to  
270 the NASG ( $\varphi_{\text{Fe}}$ ) as the product of the Fe anomaly in a ring relative to subtropical  
271 concentrations ( $\Delta\text{Fe}$ ) times the number of rings ( $n$ ), their surface area ( $A$ ), and a characteristic  
272 depth scale of the Fe anomaly accessible to the surface ocean over a seasonal cycle ( $D$ ):

273 
$$\varphi_{\text{Fe}} = \Delta\text{Fe} \times n \times A \times D \quad (1)$$

274 where  $\Delta\text{Fe} = 0.34 \mu\text{mol m}^{-3}$  (equivalent to 0.34 nmol kg<sup>-1</sup>),  $n = 7.7 \text{ rings yr}^{-1}$ ,  $A = 3.9 \times 10^4$   
275 km<sup>2</sup>, and  $D = 225$ , which is the average depth of the 1026.5 isopycnal across the Slope Sea

276 and subtropical gyre stations. Under these assumptions, the total ring-driven supply of Fe is  
 277 equal to  $0.3 \pm 0.17 \times 10^8$  mol dissolved Fe year<sup>-1</sup>. The uncertainty is propagated from the  
 278 standard deviations of the ring number ( $7.7 \pm 2.5$  per year), size ( $3.9 \pm 1.5 \times 10^4$  km<sup>2</sup>), and Fe  
 279 concentrations ( $0.2 \mu\text{mol m}^{-3}$ ), and includes an estimate of uncertainty on  $D$  of 50 m. This  
 280 calculation makes the implicit assumption that the volume transport by cold core rings into  
 281 the subtropical gyre (represented by  $n \times A \times D$ ) is balanced by an equal volume transport  
 282 out of the subtropical gyre with Fe concentrations equal to those observed in the GA03  
 283 subtropical stations.

284

285 ***Estimation of the Fe dissipation from the ring.*** To estimate the amount of Fe that would  
 286 dissipate from a cold-core ring during its time in the gyre, we use the same diffusivity as was  
 287 observed for cold-core rings shed from the Kuroshio Extension<sup>30</sup>. We assume a circular  
 288 vortex<sup>30</sup> with initial tracer concentration:

$$289 \quad \text{Fe}(r, 0) = \text{Fe}_o + \text{Fe}_1 \exp\left(\frac{-r^2}{a^2}\right) \quad (2)$$

290 where  $\text{Fe}_o$  is the subtropical Fe concentration, taken to be  $0.30 \mu\text{mol m}^{-3}$ ; (equivalent to  $0.30$   
 291  $\text{nmol kg}^{-1}$ );  $\text{Fe}_1$  is the Fe anomaly at the centre of the ring relative to the background  
 292 concentration, set at  $0.34 \mu\text{mol m}^{-3}$  (equivalent to  $0.34 \text{nmol kg}^{-1}$ );  $r$  is the distance from the  
 293 ring's centre; and  $a^2$  sets the exponential decay length scale, taken to be 48 km, as was found  
 294 to be appropriate for a ring of about 100 km radius by Qiu and colleagues<sup>30</sup>. The isopycnal  
 295 diffusion of Fe out of the ring proceeds according to:

$$296 \quad \frac{\partial \text{Fe}(r,t)}{\partial t} = A_h \frac{1}{r} \frac{\partial}{\partial r} \left[ r \frac{\partial \text{Fe}(r,t)}{\partial r} \right] \quad (3)$$

297 where  $A_h$  is the diffusivity, taken to be equal to  $300 \text{ m}^2 \text{ s}^{-1}$  as found for the cold-core rings  
 298 shed from the Kuroshio<sup>30</sup>. Suppl. Fig. 2a. shows the solution to this equation at various  
 299 times, and illustrates the rapid loss of Fe from the ring core. Suppl. Fig. 2b quantifies the

300 integrated Fe anomaly in the central 50 km of the ring (relative to the background subtropical  
301 concentration) as a function of time. These results show that after only 2 months, the ring  
302 would lose half of its integrated Fe anomaly, with less than 15% remaining after 1 year.  
303 These timescale calculations are consistent with the observation of strong interleaving within  
304 the ring in the GA03 section (Fig. 3), which satellite data suggest was shed from the Gulf  
305 Stream 5–6 weeks before it was sampled.

306

307 ***Comparison of ring-driven flux to scaling for total down-gradient mixing of Fe and other***

308 ***terms in the Fe conservation equation.*** A basic scale analysis of the down-gradient transport

309 of Fe provides a check on the order of magnitude of the ring-based estimate. Such along-

310 isopycnal diffusion scales as  $\frac{A_h D \Delta \text{Fe}}{L^2}$ , where  $A_h$  is the isopycnal diffusivity, with estimates of

311  $A_h$  ranging from 500 to 1500 m<sup>2</sup> s<sup>-1</sup>;  $D$  is the thickness of the vertical layer of interest

312 (nominally the 225 m average depth of the 26.5 isopycnal<sup>40</sup>);  $\Delta \text{Fe}$  is the difference in Fe

313 concentrations across the Gulf Stream of 0.34 nmol kg<sup>-1</sup>; and  $L$  is the horizontal length scale

314 over which the Fe change is observed (i.e. about 50 km). We note that these diffusivity

315 estimates for  $A_h$  may be higher than that used to estimate the mixing out of the rings above, as

316 turbulent diffusivity scales with the length over which the turbulent motions are averaged<sup>41</sup>.

317 With these parameter choices, the down-gradient diffusion of Fe into the subtropical gyre is

318 estimated as supplying 480-1450 μmol m<sup>-2</sup> year<sup>-1</sup>, with the range coming from the range of  $A_h$

319 values (500 to 1500 m<sup>2</sup> s<sup>-1</sup>). For a Gulf Stream length along the Slope Sea of 2000 km and

320 over a 50 km width, the total supply of Fe due to down-gradient diffusion is estimated at

321 between 0.3×10<sup>8</sup> and 2×10<sup>8</sup> mol year<sup>-1</sup>. This calculation yields a flux that is up to 7 times

322 larger than the ring-derived estimate. We take the agreement in order of magnitude between

323 the low end of this estimate with that derived from the ring statistics as an indication that the

324 ring-based estimate of Fe supply is within reasonable limits. That the diffusion-based



325 estimate may be substantially larger than the ring-based estimate is consistent with the idea  
326 that rings are one of many mesoscale processes moving Fe from the Slope Sea across the  
327 Gulf Stream.

328 This isopycnal mixing term is one of several physical mechanisms that can potentially  
329 transport Fe into or out of the subtropical gyre, in the layer above the annual maximum mixed  
330 layer. Following the approach in Williams and Follows<sup>15</sup>, we provide a scale analysis of the  
331 following additional transport terms for comparison to the ring-driven transport. On a  
332 seasonal basis, the vertical entrainment term may dominate phosphate and nitrate budgets<sup>15</sup>,  
333 since there is an accumulation of these macronutrients in the seasonal pycnocline. However,  
334 iron is depleted to depths below even the permanent pycnocline (i.e. below the 26.5  
335 isopycnal). Thus, we expect vertical entrainment to be a small term, and may cause dilution  
336 of the iron deposited on the ocean surface through a deepening mixed layer. In any case, for  
337 an iron budget integrated to the base of the deepest annual mixed layer, the vertical  
338 entrainment term is approximately offset by the biological export term<sup>15</sup>, and we expect the  
339 budget to be dominated by Ekman advection, diapycnal mixing, and vertical advection, each  
340 of which is scaled in the following analysis.

341 Ekman advection is estimated from the average down-front winds along the Gulf  
342 Stream create, which create Ekman transports of  $U = 2 \text{ m}^2 \text{ s}^{-1}$  (equivalent to  $4 \text{ cm s}^{-1}$  over an  
343 Ekman layer of 50 m depth<sup>14</sup>) acting across surface Fe concentrations that decrease from 0.6  
344 to  $0.3 \text{ } \mu\text{mol m}^{-3}$  going southward across the Gulf Stream. Therefore, over a length scale of  
345 order 50 km, we estimate an Ekman transport convergence,  $\frac{\Delta U Fe}{\Delta y}$ , of  $O(10^{-4}) \text{ } \mu\text{mol m}^{-2} \text{ year}^{-1}$ .  
346 This is many orders of magnitude smaller than that due to ring-driven transport (Fig. 4), even  
347 assuming convergence over a broad area. The dominance of eddies in the lateral transport  
348 term agrees with the results from a recently-submitted manuscript based on the results of a  
349  $1/10^\circ$  ocean model (Yamamoto A. *et al.*, unpublished data). Diapycnal mixing at the base of

350 the maximum wintertime mixed layer appears to be an extraordinarily small term, since there  
351 is a homogenous, low-Fe layer extending all the way to the 1026.75 isopycnal, well beneath  
352 the densest subtropical mixed layer (<1026.5). This term scales as  $A_v \frac{\Delta Fe}{D}$ , where  $A_v$  is a  
353 turbulent diapycnal diffusivity, typically  $O(10^{-5})$ , and  $\Delta Fe$  is the vertical iron difference over  
354 some depth,  $D$ , at the base of the annual maximum mixed layer. Since the vertical Fe  
355 gradient is essentially zero near the base of this layer, so too will the turbulent mixing supply  
356 be close to zero. Finally, vertical advection is also a small Fe removal term at the base of the  
357 annual maximum mixed layer, given the slow downwelling velocities,  $O(25 \text{ m year}^{-1})$ , acting  
358 on the low Fe concentrations  $0.3 \text{ umol m}^{-3}$  to yield an estimate for this term of  $wFe|_{z=D}$  of  $-7$   
359  $\text{umol m}^{-2} \text{ year}^{-1}$ . Hence, the mesoscale eddy-driven supply of Fe and atmospheric deposition  
360 appear to far exceed any other physical transport mechanisms. In steady state, the sum of Fe  
361 export in organic molecules and other sinking particles should balance these supply terms.

362

363 *Calculation of cross-Gulf Stream phosphate and nitrate fluxes.* Analogous calculations to  
364 those for Fe can be made for the ring-driven supply of dissolved phosphate and nitrate, with  
365 these calculations advantaged by much greater data coverage. Using GA03 dissolved  
366 nutrient data, as well as 7 much higher-resolution sections across the Gulf Stream (6 from the  
367 CLIMODE program in January 2006 and February/March 2007<sup>14,42</sup>, and 1 from cruise  
368 EN596 in April 2017 (J. B. Palter, unpublished data), we find that phosphate concentrations  
369 just north of the Gulf Stream and above the 1026.5 isopycnal are, on average,  $0.2 \text{ mmol m}^{-3}$   
370 higher than for the same layer on the subtropical side of the Gulf Stream, whilst for nitrate  
371 the corresponding difference is  $2.25 \text{ mmol m}^{-3}$ . Given the ring characteristics from the  
372 altimetry detection and tracking as above, we estimate a ring-driven phosphate supply of  
373  $1.3 \times 10^{10} \pm 0.8 \times 10^{10} \text{ mol year}^{-1}$  and a nitrate supply of  $14.6 \times 10^{10} \pm 9 \times 10^{10} \text{ mol year}^{-1}$ , with  
374 uncertainties based on ring statistics as for Fe, and also including variability in phosphate and

375 nitrate concentrations. For comparison, a study using a data-constrained ocean model<sup>18</sup>  
376 estimated a lateral supply of phosphate plus dissolved organic phosphorus to the North  
377 Atlantic subtropical gyre of approximately  $3.2 \times 10^{10}$  mol P year, most of which occurs on the  
378 southern fringe of the Gulf Stream. In that coarse-resolution model study, the supply due to  
379 lateral mean flow and parameterized eddy mixing was combined into one term that was  
380 approximately a factor of 2.5 greater than our estimate for the rings alone, corroborating that  
381 rings are likely to provide an important fraction of the total cross-Gulf Stream nutrient  
382 supply.

383

384 *Calculation of atmospheric Fe deposition fluxes to the subtropical gyre.* Accurate  
385 estimation of soluble Fe flux to the oceans requires knowledge of three parameters: a) aerosol  
386 deposition flux; b) Fe content of the deposited aerosols; and c) the fraction of deposited Fe  
387 that dissolves in seawater. Whilst the Fe content of mineral aerosol, and Saharan dust in  
388 particular, has been shown to be very similar to that of average upper continental crust<sup>43–46</sup>,  
389 uncertainties on the other two parameters are large, even leading to disagreement by several  
390 orders of magnitude on dust deposition fluxes to the same region<sup>47</sup>. We represent this range  
391 in uncertainty by estimating soluble Fe deposition to the NASG using a wide variety of  
392 methods, including both extrapolation from observational estimates in the eastern Atlantic  
393 close to the Saharan source<sup>48–50</sup> and at Bermuda in the western Atlantic<sup>51</sup>, as well as  
394 integration of simulated atmospheric deposition fluxes from numerous modelling studies<sup>3,52–</sup>  
395 <sup>57</sup> (Suppl. Info.). The fractional solubility of Fe in atmospheric aerosols ranges from <1% to  
396 >95% with a median value of ~3% in the Atlantic Ocean<sup>51</sup>. Data compilations as well as  
397 studies within the subtropical North Atlantic near Bermuda show that remote marine aerosols  
398 have fractional Fe solubility higher than the values of <1% observed for fresh mineral dust,  
399 perhaps the result of source composition, natural physical or chemical processing during

400 atmospheric transport, or the increased importance of aerosols from anthropogenic  
401 combustion sources<sup>9,33,51,58,59</sup>. We choose an upper-bound estimate for the solubility of Fe in  
402 atmospheric aerosols deposited in the subtropical North Atlantic of 5%, which is among the  
403 highest seen in the literature for this region (Suppl. Info.). Thus, our estimate of ring-  
404 mediated transport of Fe is compared to what is likely to be a maximum estimate of the  
405 atmospheric source. See Suppl. Info. for an extended discussion of all the models and details  
406 of the calculations carried out to generate the range described in the main text.

407

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554

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556

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565

## 566 **Author Contributions**

567

568 TMC and GFdS conceived the study; JBP carried out the ring-driven Fe transport  
569 calculations; GFdS carried out the atmospheric deposition calculations; all authors  
570 contributed to the writing of the manuscript.

571

## 572 **Additional Information**

573

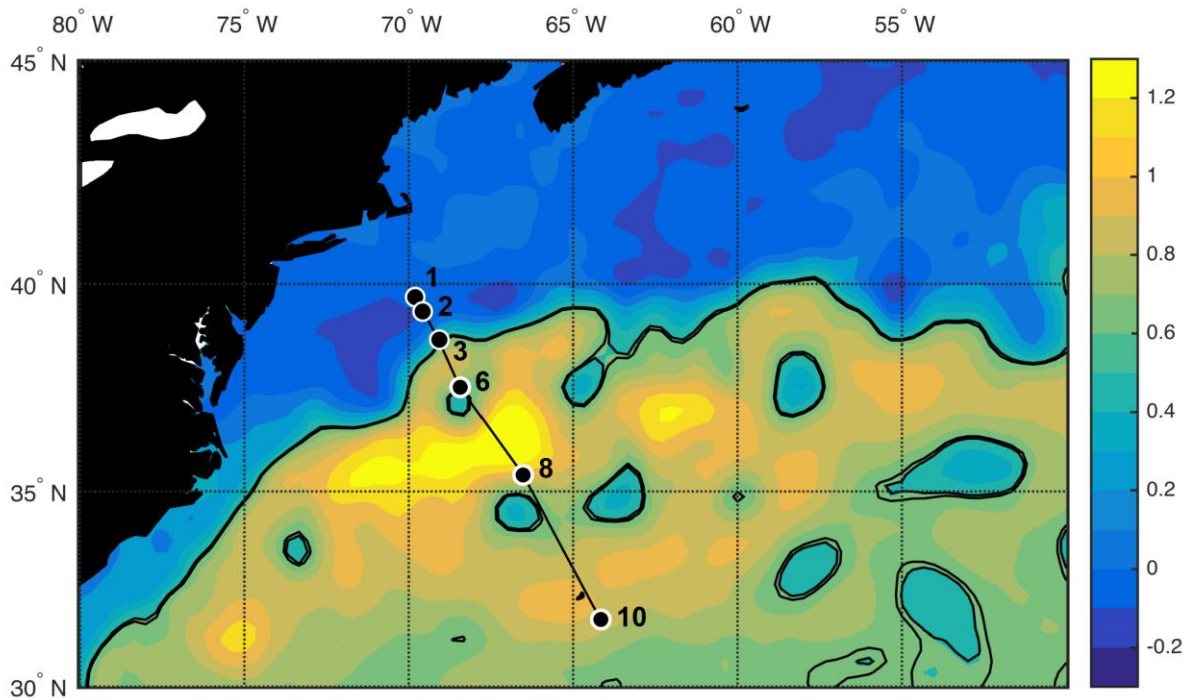
574 Supplementary Information is available in the online version of the paper. Reprints and  
575 permissions information is available online at [www.nature.com/reprints](http://www.nature.com/reprints). Correspondence and  
576 requests for materials should be addressed to TMC.

577

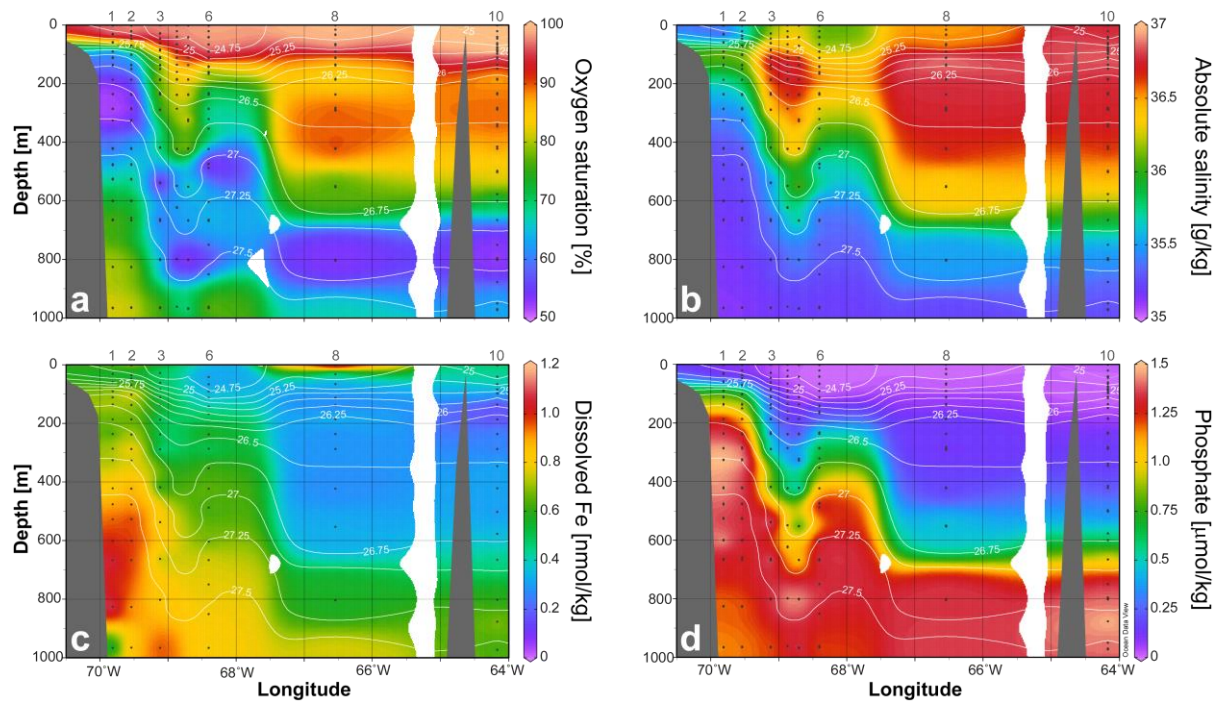
578 **Competing Financial Interests**

579 The authors declare no competing financial interests.

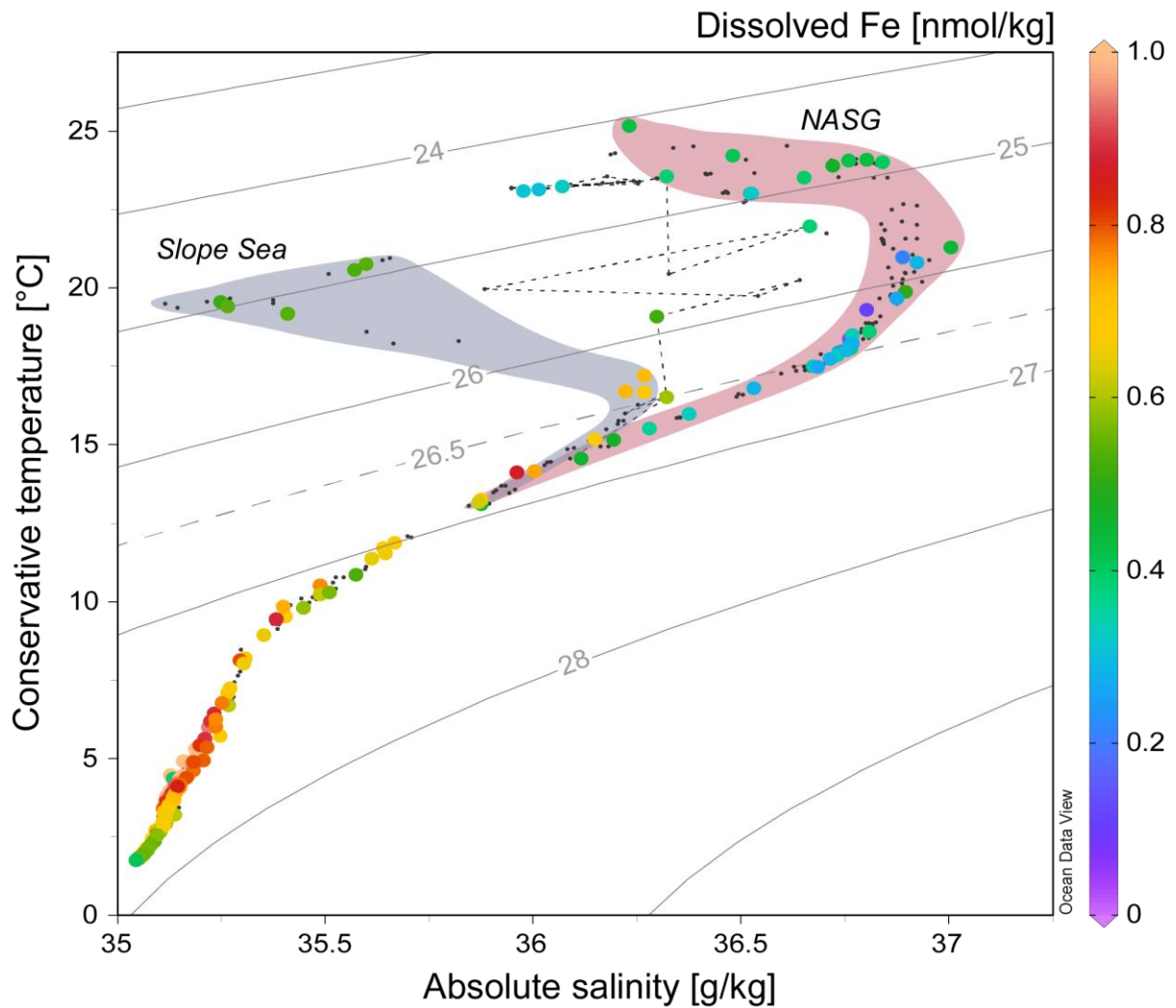




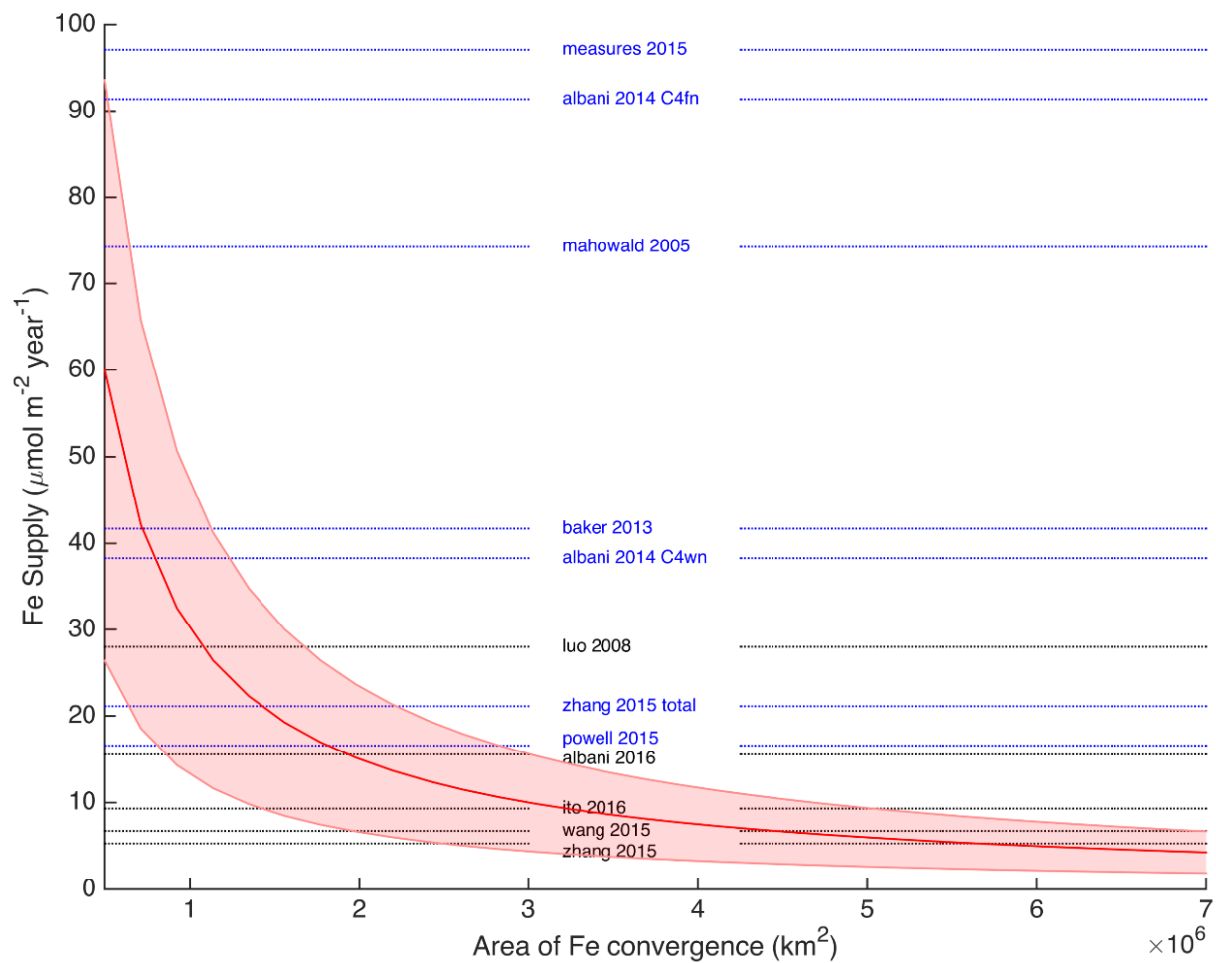
**Figure 1. GA03 Fe station sampling locations<sup>20</sup>, Gulf Stream and cold-core ring in satellite altimetry.** Satellite observations of sea-surface height (metres, black contours at 0.52 and 0.55 m) from the AVISO merged mean absolute dynamic topography product (November 9<sup>th</sup>–16<sup>th</sup> 2011) show that Station 6, sampled along the GEOTRACES GA03 (USGT11) section (black line) on November 14, 2011, was at the edge of a cold-core ring. Weekly AVISO data shows that the ring was shed from the Gulf Stream between September 28 and October 8, 2011 (see Suppl. Animation 2).



**Figure 2. A cold-core ring observed in the GA03 section.** The transition from (a) poorly-oxygenated, fresh, Fe- and P-rich waters of the Slope Sea in the west to well-oxygenated, salty, Fe- and P-depleted waters of the gyre<sup>4,21</sup>. Stations are numbered, and sampling depths marked by dots. To the east of the Gulf Stream (centred around Station 3), the upward doming of isopycnals mark the cold-core ring (Station 6). Two additional stations within the Gulf Stream improve the spatial resolution of oxygen, salinity and phosphate data, which resolve the transition between the gyre and the ring more clearly than the Fe distribution.



580 **Figure 3. GA03 Temperature-salinity diagram, with Fe concentrations<sup>4</sup> in colour.** The  
 581 Slope Sea (grey) has low temperature, low salinity and elevated Fe near the  $\sigma_\theta = 26.5$   
 582 isopycnal (dashed grey) relative to the NASG (pink). The dashed black line represents  
 583 Station 6 at the edge of the ring, with temperature-salinity showing strong interleaving  
 584 between the Slope Sea and NASG. Ring Fe concentrations are also elevated, especially  
 585 relative to Fe-depleted STMW around the  $\sigma_\theta = 26.5$  isopycnal that dominates the subsurface  
 586 NASG (Fig. 2). Grey contours show potential density anomaly  $\sigma_\theta$  in units of  $\text{kg m}^{-3}$ .



587 **Figure 4. Ring-driven dissolved Fe supply compared to atmospheric dissolved Fe**  
 588 **deposition.** Based on our estimate of total ring-driven Fe supply to the NASG, we calculate  
 589 the supply per unit area as a function of the area over which the ring-driven supply converges  
 590 (red line; shading denotes uncertainty), ranging from a small region very near the Gulf  
 591 Stream to the entire gyre. Horizontal lines show atmospheric deposition of soluble Fe to the  
 592 entire NASG from modelling and observational studies. Black lines represent studies that  
 593 modelled Fe solubility explicitly, while blue lines represent those to which we applied 5% Fe  
 594 solubility (Suppl. Info.).