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Published in:
Marine Chemistry

DOI:
[10.1016/j.marchem.2019.103748](https://doi.org/10.1016/j.marchem.2019.103748)

Publication date:
2020

Document version:
Peer reviewed version

Link:
[Link to publication in PEARL](#)

Citation for published version (APA):

Sedwick, P. N., Bowie, A. R., Church, T. M., Cullen, J. T., Johnson, R. J., Lohan, M. C., Marsay, C. M., McGillicuddy, D. J., Sohst, B. M., Tagliabue, A., & Ussher, S. J. (2020). Dissolved iron in the Bermuda region of the subtropical North Atlantic Ocean: Seasonal dynamics, mesoscale variability, and physicochemical speciation. *Marine Chemistry*, 219(0), 103748-103748. <https://doi.org/10.1016/j.marchem.2019.103748>

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Download date: 28. Apr. 2025

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**Dissolved iron in the Bermuda region of the subtropical North Atlantic Ocean:
Seasonal dynamics, mesoscale variability, and physicochemical speciation**

P.N. Sedwick^a, A.R. Bowie^{b,c}, T.M. Church^d, J.T. Cullen^e, R.J. Johnson^f, M.C. Lohan^g, C.M. Marsay^h, D.J. McGillicuddy, Jr.ⁱ, B.M. Sohst^a, A. Tagliabue^j, S.J. Ussher^k

^a*Department of Ocean, Earth and Atmospheric Sciences, Old Dominion University, Norfolk, Virginia 23508, USA*

^b*Institute for Marine and Antarctic Studies, University of Tasmania, Hobart, Tasmania 7001, Australia*

^c*Antarctic Climate and Ecosystems Cooperative Research Centre, University of Tasmania, Hobart, Tasmania 7001, Australia*

^d*School of Earth, Ocean and Environment, University of Delaware, Newark, Delaware 19716, USA*

^e*School of Earth and Ocean Sciences, University of Victoria, Victoria, British Columbia V8W 3V6, Canada*

^f*Bermuda Institute of Ocean Sciences, St. Georges GE01, Bermuda*

^g*Ocean and Earth Science, National Oceanography Centre, University of Southampton, Southampton SO14 3ZH, UK*

^h*Skidaway Institute of Oceanography, University of Georgia, Savannah, Georgia 31411, USA*

ⁱ*Woods Hole Oceanographic Institution, Woods Hole, Massachusetts 02543, USA*

^j*School of Environmental Sciences, University of Liverpool, Liverpool L69 3GP, UK*

^k*School of Geography, Earth and Environmental Sciences, University of Plymouth, Plymouth PL4 8AA, UK*

Keywords

Dissolved iron

Seasonal variability

Physicochemical speciation

Sargasso Sea

32 **Abstract**

33 Water-column data from seven cruises in 2007-2008 reveal pronounced temporal and spatial
34 variations in the distribution of dissolved iron (DFe, $<0.4 \mu\text{m}$) over the upper 1000 m of the
35 Sargasso Sea near Bermuda, in the western subtropical North Atlantic Ocean. In near-surface
36 waters, DFe exhibits a clear seasonal cycle, increasing from $\sim 0.1\text{-}0.3 \text{ nM}$ in spring to $\sim 0.4\text{-}1.0$
37 nM in summer-early fall. The observed seasonal ranges appear to reflect the extent of winter
38 convective mixing and of summer dust deposition, both of which are closely tied to atmospheric
39 circulation processes. Surface DFe concentrations also show significant (\sim two-fold)
40 submesoscale lateral variations during summer, perhaps as a result of lateral inhomogeneities in
41 wet deposition and wind-driven mixing. The summer vertical profiles reveal pronounced DFe
42 minima and sometimes deeper maxima in the lower euphotic zone, which likely reflect
43 biological uptake and shallow remineralization, and eddy-driven lateral gradients in these
44 processes. Significant variability is also seen in the mesopelagic zone, with a DFe concentration
45 range of $\sim 0.4\text{-}0.7 \text{ nM}$ at 1000 m depth, which may reflect mesoscale isopycnal displacements
46 and/or lateral advection of iron-rich waters in the lower thermocline. Physicochemical iron
47 speciation measurements indicate that the major fraction of DFe that accumulates in surface
48 waters of the Sargasso Sea during summer is colloidal-sized Fe(III), which appears to be
49 complexed by strong, iron-binding organic ligands. Concentrations of soluble iron (sFe, <0.02
50 μm) were considerably lower than DFe in the upper euphotic zone during summer, except over
51 the subsurface DFe minima, where sFe accounts for $\sim 50\text{-}100\%$ of the DFe pool. Labile Fe(II),
52 on average, accounted for around 20% of DFe, with maximum concentrations of around 0.1 nM
53 in near-surface waters and in the lower thermocline. The seasonal-scale DFe changes that we
54 have documented near Bermuda are of the same magnitude as basin-scale lateral gradients across
55 the North Atlantic, underscoring the importance of time-series observations in understanding the
56 behavior of trace elements in the upper ocean.

57 **1. Introduction**

58 As an essential micronutrient, the transition metal iron modulates marine primary production and
59 the oceanic cycling of carbon and the macronutrient elements (Boyd and Ellwood, 2010;
60 Tagliabue et al., 2017). As such, there is a need to understand the oceanic distribution of

61 dissolved iron (DFe), which is operationally defined by filtration through 0.2 μm or 0.4 μm -pore
62 filters and is assumed to be directly available to phytoplankton, and the full range of processes
63 that control this distribution. In this regard, new basin-scale data on the oceanic distribution of
64 trace elements that are emerging from the GEOTRACES program (e.g., Mawji et al., 2015;
65 Schlitzer et al., 2018) represent a major advance. For example, recent basin-scale transects in the
66 Atlantic have revealed pronounced lateral gradients in DFe that are thought to reflect deposition
67 of North African soil dust, hydrothermal emissions from the Mid-Atlantic Ridge, sedimentary
68 inputs from the continental margins, and the balance between biological uptake, scavenging and
69 remineralization (e.g., Saito et al., 2013; Ussher et al., 2013; Conway and John, 2014; Rijkenberg
70 et al., 2014; Hatta et al., 2015; Sedwick et al., 2015; Pham and Ito, 2018).

71 However, such data only provide quasi-synoptic ‘snapshots’ of water column distributions,
72 which limits their utility in identifying and characterizing time-varying input, removal and
73 internal cycling processes. For DFe, we are lacking the kind of temporally-resolved data sets
74 that have proved critical in understanding the oceanic cycling of macronutrients (e.g., Doney et
75 al., 1996; Karl et al., 1997; Arrigo, 2005, Moore et al., 2013). A limited number of time-series
76 observations have revealed substantial temporal variations in DFe over seasonal and shorter
77 timescales in surface waters of the subtropical (Wu and Boyle, 2002; Boyle et al., 2005; Sedwick
78 et al. 2005; Fitzsimmons et al. 2015a; Hayes et al., 2015), temperate (Bonnet and Guieu, 2006;
79 Birchill et al., 2017) and polar oceans (Sedwick et al., 2000, 2008, 2011; Coale et al., 2005). In
80 order to better define and understand such temporal changes at a mechanistic level, there is a
81 need for sustained time-series observations of iron and other trace elements, together with
82 associated physical and biogeochemical measurements.

83 Established ocean time-series programs, such as the Bermuda Atlantic Time-series Study
84 (BATS; Steinberg et al., 2001; Lomas et al., 2013) and the Hawaii Ocean Time-series (HOT;
85 Karl and Lukas, 1996; Church et al., 2013) are well suited for collecting such observations.
86 Previous work in the subtropical North Atlantic near Bermuda has documented substantial,
87 seasonal-scale increases in surface DFe concentrations, from relatively low values ($\sim 0.1\text{-}0.2$ nM)
88 in spring to higher values ($\sim 0.6\text{-}2$ nM) in summer, which were suggested to reflect seasonal
89 changes in vertical mixing, biological uptake, particle scavenging and dust deposition (Wu and
90 Boyle, 2002; Sedwick et al., 2005). To better understand this apparent seasonal variability in

91 DFe, we collected samples for iron measurements from the upper water column (≤ 1000 m) in the
92 BATS region during seven cruises, spanning nearly two full annual cycles, in calendar years
93 2007 and 2008. Our results delineate a consistent seasonal cycle for DFe in the upper ocean of
94 the BATS region, and reveal significant lateral gradients in DFe concentrations over meso- and
95 submesoscales, as well as providing insights into the physicochemical speciation of DFe during
96 the summer months.

97 **2. Methods**

98 **2.1. Study area and sample collection**

99 Located in the western North Atlantic Subtropical Gyre, the Bermuda region of the Sargasso Sea
100 is relatively well studied, as a result of ongoing time-series programs including Hydrostation S,
101 BATS and the Oceanic Flux Program (e.g., Steinberg et al., 2001; Phillips and Joyce, 2007;
102 Lomas et al., 2013; Conte and Webber, 2014). Annual mean surface circulation in the BATS
103 region is characterized by weak geostrophic flow towards the southwest, whereas the westward
104 propagation of mesoscale eddies dominates upper ocean circulation on monthly to seasonal
105 timescales (Siegel et al., 1999; Steinberg et al., 2001; McGillicuddy et al., 2007). A seasonal
106 thermocline and stable, shallow, surface mixed layer are typically present from late spring
107 through early fall, and are eroded by deep convective mixing during the late fall through early
108 spring (Steinberg et al. 2001).

109 Here we report data from seawater samples and observations that were collected during seven
110 research cruises aboard RV *Atlantic Explorer* between spring 2007 and early fall 2008 (Table 1),
111 as part of the Iron Air-Sea Transfer (FeAST) project. A total of 20 stations were sampled for
112 trace metal measurements in the Sargasso Sea surrounding the BATS region, between latitudes
113 29° - 34° N and longitudes 61° - 67° W (Fig. 1). These stations were all located in the deep ocean
114 (>2000 m water depth), and mostly within mesoscale eddies. The eddies were identified and
115 tracked using sea level altimetry, as described by McGillicuddy et al. (2007). Station locations
116 and associated mesoscale circulation information are provided in Table S1 in the Supplementary
117 Material. Corresponding hydrographic data (temperature, salinity, chlorophyll fluorescence,
118 dissolved oxygen) were collected using the standard BATS conductivity-temperature-depth
119 (CTD) rosette system (Lomas et al., 2013, and references therein). The CTD rosette was

120 deployed directly before or after each trace-metal sampling cast, typically within 1 km of the
121 trace-metal cast location.

122 The water-column samples for trace metal analysis were collected in modified 5 L Teflon-lined
123 external closure Niskin-X samplers (General Oceanics Inc.) that were deployed on a Kevlar line
124 and closed using PVC-coated messengers (Sedwick et al., 2005). Sample depths were estimated
125 from line out as measured by a metering sheave, which introduces an uncertainty of around 10%
126 in collection depths as a result of wire angles of as much as 30° from the vertical. Corresponding
127 surface seawater (0-1 m depth) samples were collected in 1 L wide-mouth low density
128 polyethylene (LDPE) bottles (Nalgene) mounted on the end of a ~5 m bamboo pole. This pole
129 was extended from the ship's stern for sample collection, whilst backing slowly into the wind.
130 Upon recovery, all seawater samples were transferred into a shipboard Class-100 clean container
131 laboratory. In this shipboard clean laboratory, the seawater samples were filtered through either
132 0.4 µm pore Supor Acropak filter capsules (Pall Corp.), which had been pre-rinsed with ~5 L of
133 ultrapure deionized water (>18 MΩ cm, Barnstead Nanopure) followed by several hundred mL
134 of each sample (Niskin-X samples), or through 0.4 µm pore Poretics polycarbonate membrane
135 filters (surface samples) mounted in a perfluoroalkoxy alkane (PFA) filtering assembly (Savillex;
136 Sedwick et al., 2005). All filtered seawater samples were acidified to pH 1.7 by adding 4 mL of
137 6 M ultrapure hydrochloric acid (Seastar Baseline) per liter of sample (i.e., + 0.024 M HCl), and
138 stored in rigorously acid-cleaned 125 mL LDPE bottles (Nalgene) prior to analysis (Sedwick et
139 al., 2005). Additional ~40 mL aliquots were taken from selected filtered water-column samples,
140 and stored at -20°C in screw-cap polypropylene tubes (Falcon) for post-cruise analyses of
141 dissolved nitrate+nitrite, phosphate and silicic acid, using standard autoanalyzer methods, at the
142 Bermuda Institute of Ocean Sciences or at the Marine Science Institute of the University of
143 California Santa Barbara (nutrient data are presented in Table S2 of the Supplementary Data).

144 In order to examine lateral gradients in the surface concentrations of dissolved trace metals, near-
145 surface seawater samples were collected along several transects between water-column sampling
146 stations during the two summer cruises (FeAST-2 and FeAST-6; Table 1). Near-surface (~2 m
147 depth) seawater was collected using a trace-metal clean underway 'towfish' system (Bruland et
148 al., 2005; Sedwick et al., 2011) and pumped directly into a shipboard Class-100 'clean bubble'.
149 During these transects, water samples were collected once every hour, thereby providing near-

150 surface water samples at spacings of ~12 km. These underway seawater samples were filtered
151 in-line through a 0.4 μm pore Supor Acropak filter capsule (Pall Corp.), and then acidified with
152 hydrochloric acid and stored in LDPE bottles, as described for the water-column trace metal
153 samples. The FeAST-6 cruise also sampled a number of stations south of 29°N, and collected
154 near-surface underway samples between these stations. The water column data for those FeAST-
155 6 stations are reported by Shelley et al. (2012) and are not discussed here, however the DFe data
156 from the FeAST-6 underway samples south of 29°N will be discussed in relation to the FeAST-2
157 underway data.

158 **2.2. Sample analysis**

159 Dissolved iron (DFe, <0.4 μm) was determined in the filtered, acidified seawater samples by
160 flow injection analysis with in-line preconcentration and spectrophotometric detection, modified
161 from the method of Measures et al. (1995) as described by Sedwick et al. (2008). The accuracy
162 of this method was assessed by analysis of reference seawater samples from the SAFe program
163 (all concentrations reported are ± 1 sd): the method used in this study yielded DFe concentrations
164 of 0.11 ± 0.01 nM ($n = 15$) and 0.97 ± 0.06 nM ($n = 14$) for SAFe reference seawater S1 and D2,
165 compared with consensus values of 0.093 ± 0.008 nM and 0.933 ± 0.023 nM respectively.
166 Robust estimates of our long-term analytical precision are based on multiple separate
167 determinations of the SAFe seawater reference materials, which yield estimates of intermediate
168 precision (see Worsfold et al., 2019) of $\pm 15\%$ ($n = 33$) at the concentration levels of SAFe S and
169 $\pm 9\%$ ($n = 16$) at the concentration levels of SAFe D2. The analytical limit of detection is
170 estimated as the DFe concentration equivalent to a peak area that is three times the standard
171 deviation on the system manifold blank, which is below 0.04 nM (Sedwick et al., 2005).

172 In addition, several types of iron speciation measurements were made using samples collected
173 during the summer cruises: soluble iron (sFe, <0.02 μm) was measured in samples collected
174 during FeAST-2 and FeAST-6; labile iron (II) was determined in samples collected during
175 FeAST-2, and iron-binding ligands were measured in samples collected during FeAST-2. For
176 sFe measurements, seawater samples were filtered as described for DFe, and the filtrate was then
177 filtered through dilute-acid-cleaned, sample-rinsed 0.02 μm Anotop syringe filters using a
178 peristaltic pump (Ussher et al., 2010). The resulting filtrate was acidified to pH 1.7 and stored in

179 acid-cleaned 60 mL LDPE bottles, for post-cruise determinations of sFe by flow injection
180 analysis using the method described for DFe. The analytical accuracy, precision and limit of
181 detection for the sFe measurements are assumed to be the same as for the DFe determinations.

182 For measurements of iron(II) (Fe(II)), subsamples of unfiltered seawater were drawn directly
183 from the Niskin-X sampler or pole-sample bottle into a 60 mL fluorinated ethylene propylene
184 bottle (Nalgene). Operationally-defined labile Fe(II) (Sarhou et al., 2011) was measured
185 immediately using flow-injection analysis with in-line preconcentration and chemiluminescence
186 detection (Bowie et al., 2002). The analytical limit of detection was estimated as the Fe(II)
187 concentration corresponding to a signal equal to three times the standard deviation of triplicate
188 analyses of the blank (Bowie et al., 2004; Sarhou et al., 2011), and averaged 5.1 pM. There is
189 currently no standard reference material for Fe(II) in seawater, thus we are unable to provide
190 rigorous estimates of the accuracy and precision of the Fe(II) determinations. However, an
191 indication of the internal instrumental precision for the Fe(II) measurements is provided by
192 repeat measurements of standards prepared in low-Fe(II) seawater, which yielded average
193 relative standard deviations of <7% for standard additions of 0.2, 0.4, 0.6, 0.8 and 1.0 nM.

194 Iron-binding ligands were determined in 0.4 μm -filtered seawater samples using competitive
195 ligand exchange adsorptive cathodic stripping voltammetry (CLE-ACSV), employing 2,3-
196 dihydroxynaphthalene (DHN) as the competing ligand (van den Berg, 2006). Aliquots of
197 seawater samples (10 mL) were dispensed into individual 15 mL perfluoroalkoxy (PFA) vials
198 (Savillex) that were previously conditioned with seawater/DHN solution. DHN in methanol
199 solution was added to a final concentration of 0.5 μM , and iron standard solution was added to
200 the titration vessels to achieve a range of +0 to +8 nM DFe. Samples were then allowed to
201 equilibrate for ~24 hours, after which they were transferred to PFA voltammetric cells and 0.5
202 mL of a potassium bromate/3-(4-(2-hydroxyethyl)-1-piperazinyl)propansulfonic acid/ammonia
203 solution was added to adjust the solution to pH 8 and to provide an oxidant for catalysis of the
204 reaction of Fe-DHN at the hanging mercury drop electrode (HMDE). Labile DFe was
205 determined using a Metrohm model 663VA HMDE connected to a $\mu\text{Autolab II}$ potentiostat
206 (Ecochemie/Metrohm) after purging the sample with nitrogen gas for 5 minutes followed by 90
207 seconds adsorption at -0.1 V, 8 seconds equilibration and a sampled direct current scan with a
208 frequency of 10 s^{-1} and a step size of 4 mV. Titration data were fitted to a Scatchard or

209 Langmuir type equation to determine relevant thermodynamic parameters and concentrations of
210 DFe complexing ligands (van den Berg, 2006; Cullen et al. 2006).

211 **3. Results and discussion**

212 Vertical concentration profiles for DFe, grouped according to each of the seven FeAST cruises,
213 are presented in Figures 2a-2g, with the corresponding data presented in Table S2 of the
214 Supplementary Data.

215 **3.1. Seasonal changes in dissolved iron**

216 The data from our seven cruises in the BATS region reveal pronounced seasonal-scale changes
217 in DFe concentrations in near surface waters, which increase from ~0.1-0.3 nM during spring to
218 ~0.4-1.0 nM during summer and early fall (Fig. 2). In addition, consistent seasonal changes in
219 the shape of the DFe concentration profiles over the upper 300 m of the water column are
220 apparent. Samples collected during spring (March and April) define a fairly homogeneous DFe
221 distribution over the upper water column (Figs. 2a, 2e), whereas DFe profiles from the summer
222 (June and July) and early fall (September) are marked by the development of a near-surface
223 concentration maximum, a sub-surface minimum centered at depths of ~75-150 m, and, in some
224 profiles, a deeper sub-surface maximum centered near depths of 90-150 m (Figs. 2b, 2c, 2f, 2g).
225 The single profile from late fall (November 2007; Fig. 2d) shows uniformly elevated DFe
226 concentrations over the euphotic zone, as might be expected to result from the seasonal cooling
227 and deepening of the summer mixed layer. At depths greater than 300 m, DFe concentrations
228 uniformly increase, reaching concentrations of ~0.4-0.7 nM at 1000 m (Fig. 2).

229 Although there are some significant differences between individual DFe profiles collected during
230 a single cruise (e.g., for FeAST-2 and FeAST-3), a general seasonal pattern remains, as is
231 evident from the cruise-averaged DFe concentration profiles presented in Figures 2h-2n.
232 Evidence to support our interpretation of these general, inter-cruise differences as reflecting
233 temporal rather than spatial variability is provided by water-column DFe profiles from stations 1-
234 3 (sampled on 26 April 2007) and 2-2 (sampled 9 July 2007), shown in Figure 3a. These profiles
235 represent repeat samplings of a single mode-water eddy that was tracked for several months
236 using sea-level altimetry (Figs. 3b, 3c). A comparison of the DFe concentration profiles from

237 these two stations (Fig. 3a) clearly indicates the development of a surface maximum, subsurface
238 minimum and subsurface maximum over the 2.5-month period between samplings, assuming
239 Lagrangian behavior for waters within the eddy. Our results thus support and expand on earlier
240 suggestions of a seasonal cycle in DFe that were based on samples collected in the Sargasso Sea
241 during cruises in the spring and summer (Wu and Boyle, 2002; Sedwick et al., 2005).

242 As noted in these previous studies, such seasonal changes in the vertical distribution of DFe may
243 be interpreted as the result of a number of forcing processes that exhibit well established
244 seasonal changes in the BATS region; namely:

245 (1) Vertical mixing of the upper water column, which is driven by seasonal changes in surface
246 heat flux and wind stress. In the BATS region, the water column is convectively mixed to ~200-
247 400 m depth during the winter months, then re-stratified by warming during the late spring and
248 into summer, when the surface mixed layer shoals to depths of 20 m or less, before convective
249 overturn again commences in the late fall (Steinberg et al., 2001; Lomas et al., 2013);

250 (2) Primary production and the associated export of particulate organic matter, which is greatest
251 during the winter-spring period, before nutrients have been exhausted from the upper euphotic
252 zone (Michaels and Knap, 1996; Steinberg et al., 2001; Lomas et al., 2013); and

253 (3) Deposition of iron-bearing aerosols, which reaches a maximum during the summer months,
254 when high pressure systems over the subtropical North Atlantic region facilitate the atmospheric
255 transport of soil dust from northern Africa to the Sargasso Sea (Moody et al., 1995; Huang et al.,
256 1999; Arimoto et al., 2003; Kadko et al., 2015).

257 Based on our understanding of the geochemical behavior of iron in the ocean (e.g., Johnson et
258 al., 1997; Boyd and Ellwood, 2010; Tagliabue et al., 2017), the known seasonal modulation of
259 these three processes provides a consistent framework to understand the observed seasonal-scale
260 changes in the vertical distribution of DFe that we have documented in the Sargasso Sea. During
261 the winter and early spring, vertical mixing homogenizes DFe over the upper water column,
262 while primary production removes DFe from the water column through biological assimilation
263 and associated export of organic matter and particle scavenging (Wu and Boyle, 2002; Sedwick
264 et al., 2005). Collectively, these processes result in the relatively low and uniform DFe
265 concentration profiles for our spring cruises (Figs. 2a, 2e). From late spring into summer, a

266 seasonal thermocline forms together with a shallow surface mixed layer from which
267 macronutrients are depleted by biological uptake, whereas DFe increases as a result of an
268 elevated deposition of mineral aerosols that partially dissolve in surface waters (Sedwick et al.,
269 2005; Fishwick et al. 2014).

270 In addition, subsurface DFe minima typically form in association with a subsurface chlorophyll
271 maximum (SCM), which develops in late spring-early summer and persists into early fall (Figs.
272 2b, 2c, 2f, 2g). These subsurface DFe minima likely reflect the scavenging removal of DFe by
273 biogenic particles formed in and exported from the lower euphotic zone, as well as the enhanced
274 cellular iron requirements of the phytoplankton that inhabit the low-light environment of the
275 SCM (Bruland et al., 1994; Sunda & Huntsman, 1997; Sedwick et al., 2005). Profiles from some
276 of the summer and early fall cruises also show DFe maxima at depths below the subsurface DFe
277 minima (Figs. 2b, 2c, 2g), perhaps reflecting remineralization or desorption of DFe from
278 particles exported from shallower depths. Finally, DFe data from the single cruise in late fall
279 (Fig. 2d) indicate the onset of convective overturning, with the downward mixing of DFe-rich
280 surface waters resulting in uniformly elevated concentrations over the upper 75 m.

281 Beyond these apparent seasonal trends, the DFe profiles from 2007 and 2008, together with
282 previously published data from 2003 and 2004 (Sedwick et al., 2005), provide hints of
283 interannual variability modulated by the aforementioned forcing processes. For example, the
284 upper water column DFe concentrations in spring 2007 (~0.1 nM, Fig. 2a), are around half the
285 values measured in spring 2008 (~0.2 nM, Fig. 2e), which may reflect the deeper surface mixing
286 and greater net primary production in winter-early spring 2007 compared to winter-early spring
287 2008 (as revealed by Figures 1A and 4A in Lomas et al., 2013). In addition, near-surface DFe
288 concentrations as high as 2 nM were measured in the BATS region during summer 2003
289 (Sedwick et al., 2005), compared with maximum concentrations less than 1 nM in summer and
290 early fall of 2007 and 2008 (Figs 2b, 2c, 2f, 2g). This perhaps reflects a greater total aerosol iron
291 deposition to the Bermuda region during the summer of 2003, which included an intense
292 ‘Saharan dust’ deposition event in late July-early August (Sedwick et al., 2005).

293 **3.2. Mesoscale variability in the vertical distribution of dissolved iron**

294 As well as the apparent temporal variations in the vertical distribution of DFe in the Sargasso
295 Sea, our field data provide evidence of significant lateral gradients in DFe concentrations at the
296 meso- and submesoscale. During the July 2007 (FeAST-2) and June 2008 (FeAST-6) cruises,
297 water-column and near-surface samples were collected over relatively broad geographic areas.
298 The FeAST-2 cruise collected water-column samples in three different types of eddies – cyclonic
299 (Fig. 1, Stations 2-1, 2-3, 2-4, 2-6), mode-water (Fig. 1, Station 2-2) and anticyclonic (Fig. 1,
300 Station 2-5) – as well as sampling near-surface waters along transits between these eddies.
301 Although water-column DFe data from the FeAST-6 cruise south of 29°N are not discussed here
302 (results are presented by Shelley et al., 2012), we will discuss data from the underway near-
303 surface samples collected from that area during FeAST-6.

304 The three types of eddies sampled during the FeAST-2 cruise are characterized by distinct
305 hydrographic differences: cyclonic eddies exhibit upward doming of the seasonal and main
306 pycnoclines; anticyclones exhibit depression of both density surfaces; and mode-water eddies
307 have an upward domed seasonal pycnocline, whereas the main pycnocline is depressed
308 (McGillicuddy et al., 2007). Figure 4 shows schematic depictions of these features, together
309 with a map of satellite-derived sea level altimetry indicating the three eddies that were sampled
310 during the FeAST-2 cruise. Figure 5 shows interpolated along-track vertical sections of raw
311 temperature and in-situ fluorescence data from CTD casts that were made at stations across these
312 three eddies during the FeAST-2 cruise. The temperature section (Fig. 5a) clearly reflects the
313 abovementioned pycnocline displacements, whereas the fluorescence section (Fig. 5b) reveals
314 inter-eddy differences in the depth and intensity of the SCM, which are likely driven by
315 differences in vertical nutrient supply.

316 Both cyclonic and mode-water eddies result in the upwelling of nutrients into the euphotic zone,
317 with eddy/wind interactions amplifying this process for mode-water eddies (McGillicuddy et al.,
318 2007). This likely explains the shallower, more intense SCM observed in these eddy types
319 during the FeAST-2 cruise, particularly in the mode-water eddy, relative to the deeper and
320 weaker SCM in the anticyclonic eddy (Fig. 5b) where the nutricline was depressed (data not
321 shown). Importantly, these inter-eddy differences in fluorescence are reflected in the
322 corresponding DFe profiles (Fig. 6), and are consistent with our interpretation of the subsurface
323 DFe minima as the result of enhanced biological uptake and scavenging within the SCM. All

324 three eddy types showed surface maxima in DFe, presumably reflecting aerosol iron input. In
325 the cyclonic and mode-water eddies, DFe concentrations decrease to minima at a depth of 75 m,
326 close to the depth of the SCM (Fig. 6, left and middle panels). In the case of the anticyclonic
327 eddy, with its deeper and weaker SCM, a corresponding DFe minimum was absent and the
328 lowest DFe concentration was measured in the sample from 300 m depth (Fig. 6, right panels).

329 There are other inter-eddy differences in the FeAST-2 DFe profiles that appear to reflect
330 differences in the hydrographic structures and histories of the eddies. The cyclonic eddy, with its
331 up-doming of the main pycnocline, displays a gradual increase in DFe of ~ 0.1 nM between 150
332 m and 500 m depth in three of the four casts. This contrasts with the mode-water eddy, in which
333 there is little change in temperature between these depths, and the corresponding DFe
334 concentrations are indistinguishable (Fig. 6). In addition, the anticyclonic eddy had significantly
335 higher surface DFe concentrations (~ 1 nM) than the cyclonic and mode-water eddies (~ 0.5 - 0.7
336 nM). This difference likely reflects the back trajectory of the anticyclone, which approached the
337 BATS region from much further south than the cyclonic and mode-water eddies, based on sea
338 level altimetry analyses over the period before the cruise (data not shown). As such, surface
339 waters carried within the anticyclonic eddy are likely to have had relatively high initial DFe
340 concentrations, based on the north-to-south increase in near-surface DFe concentrations revealed
341 by samples collected during the FeAST-6 cruise (as discussed in Section 3.3, below, and by
342 Shelley et al., 2012).

343 The impact of mesoscale eddies on the depths of isopycnal surfaces extends well below the
344 surface mixed layer and seasonal thermocline. In the CTD profiles obtained nearest to our trace
345 metal sampling casts, temperatures at 1000 m depth varied by as much as several degrees celsius
346 (e.g., compare lower panels in Fig. 6). Our data reveal strong vertical gradients in DFe
347 concentration within the main thermocline between depths of 400 m and 1000 m (Figs. 2, 6).
348 Still higher DFe concentrations are expected at depths below our deepest samples; for example,
349 DFe concentrations above 0.8 nM were measured in the 1200-1500 m depth range at the BATS
350 station in November 2011 (Hatta et al., 2015; Sedwick et al., 2015). Given these vertical
351 concentration gradients, it seems likely that meso- and submesoscale differences in the depth of
352 isopycnal surfaces contributed to the relatively wide range of DFe concentrations (0.39-0.75 nM)
353 that were measured in samples from 1000 m depth during our two-year study. Unfortunately, we

354 cannot conduct a rigorous examination of DFe as a function of density in our deepest samples,
355 because the Kevlar line samples for DFe were collected as much as ~1 km from the location of
356 the nearest CTD rosette casts (i.e., we lack co-located measurements of DFe concentration and
357 density), and there is also an uncertainty of as much as 10% in our DFe sample depths. An
358 additional factor that may influence the observed variability in the DFe concentrations of our
359 deepest samples is the lateral advection of iron-rich waters from the Bermuda platform or the
360 North American continental margins (Hatta et al., 2015; Conway et al., 2018), a process which
361 has been proposed to explain elevated concentrations of dissolved manganese and particulate
362 lithogenic elements at mesopelagic depths in the Sargasso Sea (Sholkovitz et al., 1994; Conte et
363 al., 2019)

364 **3.3. Lateral variability in near-surface dissolved iron concentrations**

365 Data from underway samples collected during our two summer cruises, FeAST-2 (3 towfish
366 transects, Fig. 7a) and FeAST-6 (4 towfish transects, Fig. 7b), reveal analytically significant
367 variations (>0.1 nM) in the DFe concentrations of near-surface (~ 2 m depth) samples over
368 lateral distances of as little as ~10-15 km (samples were collected hourly while underway at ~6-8
369 knots). For near-surface seawater collected during the FeAST-2 cruise, DFe concentrations
370 display variations of around 0.3-0.4 nM along each towfish transect (Table 2), with DFe
371 differing by more than 0.2 nM in some consecutive samples (Fig. 7a). For these samples, there
372 are no discernable trends in near-surface DFe concentrations in relation to sea level anomaly (see
373 Fig. 4), sea-surface temperature, or sea-surface salinity (data not shown), and the mean DFe
374 concentrations along each FeAST-2 towfish transect are similar at around 0.6 ± 0.1 nM (Table
375 2). It should be noted, however, that the FeAST-2 towfish transects did not cross the
376 anticyclonic eddy, where an elevated surface DFe concentration (Fig. 6) is thought to reflect the
377 south-to-north trajectory of this circulation feature. We suggest that the submesoscale variations
378 in near-surface DFe concentrations along the FeAST 2 towfish transects may reflect lateral
379 inhomogeneities in wet deposition and wind-driven vertical mixing, as a result of the localized
380 rain events and squalls that are often observed during the summer months. Given that DFe
381 concentrations measured in rainwater near Bermuda are as much as two orders of magnitude
382 higher than surface seawater in this region (Sedwick et al., 2007), it is conceivable that spatially

383 patchy rainwater inputs could produce substantial lateral variations in surface DFe
384 concentrations while having little impact on the salinity of surface waters.

385 Larger lateral differences are apparent from towfish samples collected during the FeAST-6
386 cruise, with DFe concentrations varying by ~0.3-0.7 nM along each transect (Table 2), and by as
387 much as 0.7 nM between consecutive towfish samples (Fig. 7b). Moreover, the data from the
388 FeAST-6 towfish transects, which span a larger meridional range (~24°-31°N) than the FeAST-2
389 transects (~29°-32°N), suggest that there is a north-to-south gradient in near-surface DFe
390 concentrations to the south of Bermuda. Although the mean DFe concentration along the
391 northernmost FeAST-6 transect (~0.6 nM, Table 2, tow 1) is not significantly different from the
392 mean values along the FeAST-2 transects, there is an apparent southward increase in DFe
393 concentrations to values above 0.8 nM south of 30°N (Fig. 7b). In addition, the mean near-
394 surface DFe concentrations are significantly higher (0.97-1.13 nM) for the three southernmost
395 towfish transects sampled during the FeAST-6 cruise (Table 2, tows 2, 3 and 4). These
396 observations suggest a regional-scale trend in near surface DFe concentrations, which increase
397 from ~0.6 nM in the BATS region to around 1 nM or more to the south of 29°N. There is no
398 obvious relationship between near-surface DFe concentrations and the mesoscale circulation
399 features that were traversed by the FeAST-6 towfish transects (see Shelley et al., 2012).

400 The observed lateral variations in near-surface DFe concentrations are most likely driven by
401 differences in mineral aerosol deposition and vertical mixing, and their impact on the DFe
402 inventory of surface waters in our study region, as discussed in Section 3.1. For the individual
403 towfish transects in the BATS region, there were no significant relationships between near-
404 surface DFe concentrations and underway surface salinity or temperature, with the exception of
405 FeAST-6 Tow 1 (Fig. 7), for which there are strong positive correlations between DFe and
406 salinity ($r^2 = 0.56$) and temperature ($r^2 = 0.81$). These correlations appear to reflect regional-
407 scale, meridional increases in near-surface DFe, salinity and temperature to the south of the
408 BATS region, rather than patchiness in wet deposition or wind mixing, which would instead be
409 expected to result in negative correlations of DFe with salinity and temperature. As discussed by
410 Shelley et al. (2012), southward increases in the near-surface concentrations of dissolved iron
411 and aluminum likely reflect higher annual dust deposition south of ~30°N (Albani et al., 2014) as
412 well as lessened vertical mixing and biological removal, noting that the region between 25°N and

413 32°N represents a transition from seasonally-stratified waters in the north to permanently-
414 stratified, oligotrophic waters in the south (Steinberg et al., 2001). Similar increases of ~0.5 nM
415 in surface DFe concentrations between ~30°N and ~20°N in the western subtropical North
416 Atlantic have been reported previously (Wu & Boyle, 2002; Bergquist et al., 2007; Rijkenberg et
417 al., 2014).

418 **3.4. Physicochemical speciation of dissolved iron during summer**

419 Knowledge of the physicochemical speciation of DFe, and its variability in space and time, is
420 key to a mechanistic understanding of numerous processes that impact the ocean iron cycle.
421 These processes include biological uptake, aerosol dissolution, aggregation and disaggregation,
422 sorption and desorption, and interaction with organic matter (Tagliabue et al., 2017). Although
423 the logistical constraints of our cruise program allowed for DFe speciation measurements during
424 only the FeAST-2 (July 2007) and FeAST-6 (June 2008) cruises, our results complement other
425 iron speciation data from the Sargasso Sea, including analyses of samples collected from the
426 BATS station during GEOTRACES cruises GA02 (June 2010) and GA03 (November 2011).

427 **3.4.1. Vertical distribution of soluble iron**

428 During the FeAST-2 and FeAST-6 cruises, soluble Fe (<0.02 μm) concentrations were generally
429 low (<0.2 nM) throughout the upper water column, and increased to ~0.2-0.3 nM at 1000 m
430 depth (Fig. 8; data provided in Table S2 in the Supplementary Material). The sFe profiles reveal
431 some vertical structure in the euphotic zone, including sFe concentration maxima near the depth
432 of the DFe minima (Figs. 8a, 8b, 8c, 8e, 8g, 8h). However, in discussing these data it should be
433 noted that analytical uncertainties are considerable ($\pm 15\%$ or more) for the sFe concentrations
434 below 0.1 nM. Several samples for which sFe is greater than DFe are considered contaminated
435 (Fig. 8d, 1000 m sample; Fig. 8e, 300 m sample; Fig. 8f, 75 m sample; Fig. 8g, 10 m sample),
436 while a number of other samples with conspicuously high sFe concentrations might reflect
437 contamination during processing (Fig. 8c, surface sample; Fig. 8f, 10 m and 90 m samples).
438 Consistent with previously reported data from the Sargasso Sea (Wu et al., 2001; Fitzsimmons et
439 al., 2015b), sFe concentrations were considerably lower than DFe in the upper euphotic zone, but
440 account for a substantial proportion (~50-100%) of DFe in the lower euphotic zone, where DFe
441 minima are typically associated with maxima in chlorophyll fluorescence (see Fig. 6).

442 Concentrations of both sFe and DFe increase below the euphotic zone, with sFe accounting for
443 31-56% of DFe concentrations at 1000 m depth.

444 Our results indicate that the DFe pool is dominated by colloidal iron (cFe, calculated as the
445 difference between DFe and sFe) in near-surface waters; by sFe in the subsurface euphotic zone,
446 where DFe concentrations are low; and by significant proportions of both sFe and cFe at
447 mesopelagic depths. These observations imply that dust-derived DFe is dominated by colloidal-
448 sized organic and/or inorganic species in near-surface waters, consistent with the results of
449 dissolution experiments using Bermuda aerosols (Fishwick et al., 2014), whereas sFe accounts
450 for a major proportion of DFe in the lower euphotic zone, perhaps due to preferential biological
451 uptake or scavenging removal of cFe, and/or sustained production and biological uptake of sFe
452 from the cFe pool (Bergquist et al., 2007; Fitzsimmons et al., 2015b, 2015c; Birchill et al., 2017).
453 In our deeper samples, sFe and cFe concentrations tend to be more similar, which Fitzsimmons
454 et al. (2015b) ascribe to conditions that approach a steady state, where DFe is reversibly
455 exchanged between soluble- and colloidal-size fractions following the remineralization of iron in
456 the mesopelagic zone.

457 **3.4.2. Vertical distribution of Fe(II)**

458 Water column concentrations of labile Fe(II) were generally low during the FeAST-2 cruise
459 (data provided in Table S2 in the Supplementary Material), with maximum values around 0.1 nM
460 measured in samples collected near the surface and in the 750-1000 m depth range (Figs. 8a-8f).
461 In general, the low Fe(II) and sFe concentrations are analytically indistinguishable over the upper
462 water column, except in the subsurface DFe minima, where Fe(II) does not exhibit the
463 concentration maxima observed for sFe (Figs. 8a, 8b, 8c, 8e). The elevated Fe(II) concentrations
464 near the sea surface may reflect recent dry or wet deposition of aerosols, which are known to
465 contain readily soluble Fe(II) species (Kieber et al., 2001, 2005; Schroth et al., 2009), or the
466 photochemical reduction of dissolved or particulate Fe(III) species (Barbeau et al., 2001;
467 Hansard et al, 2009). The somewhat elevated Fe(II) concentrations at 750-1000 m depth may
468 reflect remineralization of organic matter, given that apparent oxygen utilization is greatest at
469 ~800 m depth in our study region, as well as slower rates of Fe(II) oxidation in the cooler waters
470 of the lower thermocline (Millero et al., 1987; Sedwick et al., 2015). Importantly, the similar

471 concentrations of Fe(II) and sFe above 300 m depth imply that reduced iron species may
472 constitute a major proportion of the sFe pool within the euphotic zone.

473 Dissolved Fe(II) was also measured in 0.2 μm filtered water column samples collected from the
474 BATS station during GEOTRACES cruise GA03, in November 2011 (Sedwick et al., 2015). In
475 this case the concentration profile again showed an elevated Fe(II) concentration (0.25 nM) near
476 the sea surface, with the higher Fe(II) concentration (compared to the FeAST-2 samples) perhaps
477 related to the larger inventory of DFe in the surface mixed layer in November 2011, when DFe
478 concentrations were \sim 0.7 nM over the upper 100 m of the water column (Sedwick et al., 2015).
479 Below the euphotic zone, the Fe(II) concentrations in the GEOTRACES samples were somewhat
480 higher (0.15-0.24 nM) than the values measured in the FeAST-2 samples. This difference might
481 reflect interannual variability, given that data from the GEOTRACES GA03 section suggests an
482 influence of iron sources on the Bermuda platform and/or North American continental margins
483 on the water column distributions of cFe and DFe at the BATS station (Fitzsimmons et al.,
484 2015b; Hatta et al., 2015; Sedwick et al., 2015; Conway et al., 2018). However, there were also
485 methodological differences in the Fe(II) measurements during FeAST-2 and GEOTRACES
486 GA03, in that the FeAST-2 samples were not filtered, which could conceivably influence the
487 measured Fe(II) concentrations.

488 **3.4.3. Iron-binding ligands**

489 Measurements of dissolved, iron-binding ligands were restricted to a single water column profile
490 from the FeAST-2 cruise in July 2007 (data provided in Table S2 in the Supplementary
491 Material). These results are presented in Figure 9, which shows vertical concentration profiles of
492 DFe as well as the two iron-binding ligands that were analytically resolved: L₁, a strong iron-
493 binding ligand (logarithm of conditional stability constant = 11.8-12.9), and L₂, a weaker iron-
494 binding ligand (logarithm of conditional stability constant = 10.4-11.0). The vertical profile of
495 L₁ concentrations is quite similar to and exceeds DFe concentrations to a depth of 300 m, below
496 which L₁ was not detected. The weaker ligand, L₂, was substantially higher than DFe, with
497 highest concentrations in the deeper samples. These data indicate that DFe was largely
498 complexed by an excess of strong, iron-binding ligands in the upper water column.

499 These results are consistent with other electrochemical measurements of iron-binding ligands in

500 samples collected from the BATS region in June 2010 (Gerringa et al., 2015; Buck et al., 2016)
501 and November 2011 (Buck et al., 2015, Buck et al., 2016; Fitzsimmons et al., 2015c), which
502 used 2-(2-Thiazolylazo)-p-cresol and salicylaldoxime, respectively, as competing ligands.
503 However, there are apparent differences between these different data sets in the number and
504 strengths of ligands detected, and in the excess concentrations of these ligands relative to DFe.
505 Reasons for these differences, both analytical and environmental, are discussed by Buck et al.
506 (2016), and likely extend to our data as well. In addition, Fitzsimmons et al. (2015c) have
507 examined iron-binding ligands in both soluble and colloidal size fractions in the upper water
508 column at the BATS station, and use thermodynamic calculations to argue that much of the
509 strong ligand-bound iron in the euphotic zone may in fact be present as inorganic iron
510 oxyhydroxide species, rather than iron complexed by colloidal-sized organic ligands. Further
511 research will be required to reconcile these observations.

512 **3.5. Implications for biogeochemical modeling of iron in the ocean**

513 Towards the goal of achieving a mechanistic understanding of oceanic iron cycling, time-series
514 observations provide an important metric against which to assess the veracity of ocean
515 biogeochemical models. In the case of dissolved iron, contemporary models are lacking in their
516 ability to reproduce seasonal scale variability in the upper ocean. For example, Figure 10 shows
517 the seasonal (April, July, November) multi-model minimum, average and maximum solutions for
518 DFe concentrations over the upper 1000 m at 31°N, 65°W, from the oceanic iron model
519 intercomparison project (FeMIP) that is reported by Tagliabue et al. (2016). Comparing these
520 FeMIP model solutions to our average DFe profiles for those months in 2007 (Figs. 2h, 2i, 2k;
521 data also shown as crosses in Fig. 10) shows the observed DFe concentrations to be substantially
522 less than the average model solutions in subsurface waters, as well as in surface waters during
523 the spring. These discrepancies likely reflect limitations in the model parameterizations of
524 remineralization release and scavenging loss of DFe in subsurface waters, as well as aerosol
525 input and biological removal of DFe in the surface mixed layer.

526 **4. Concluding remarks**

527 Our multi-cruise, seasonal-scale measurements of dissolved iron in the Bermuda region
528 demonstrate substantial temporal and spatial variations in the distribution of this biologically

529 important trace element in the upper ocean. Most striking are the pronounced seasonal-scale
530 changes in the distribution of DFe over the euphotic zone and upper thermocline. Here vertical
531 profiles evolve from the relatively low and homogeneous DFe concentrations ($\sim 0.1\text{--}0.3$ nM) in
532 early spring, to the elevated near-surface concentrations ($\sim 0.4\text{--}1$ nM) and underlying
533 concentration minima in the SCM, which reflect the impact of thermal stratification and elevated
534 mineral aerosol deposition during the summer and early fall. The potential importance of time-
535 series measurements of iron and other trace elements is underscored by recognizing that the
536 seasonal-scale of DFe changes that we observed in the surface ocean near Bermuda are of the
537 same magnitude as the basin-scale lateral gradients in DFe concentrations across the North
538 Atlantic (e.g., Hatta et al., 2015; Rijkenberg et al. 2014), and that such temporal variability may
539 exist in many different oceanic settings (e.g., Bonnet and Guieu, 2006; Birchill et al., 2017).

540 Our data also reveal significant lateral variations in the distribution of DFe in the Sargasso Sea,
541 which appear to reflect mesoscale physical circulation features and their impact on biological
542 production, as well as submesoscale to regional-scale gradients in the dry and wet deposition of
543 aerosol iron to the ocean surface. Finally, our time-series data highlight the limitations of
544 contemporary ocean biogeochemical models in simulating the vertical distribution and temporal
545 variability of DFe. In this regard, research that links time-series measurements of iron speciation
546 and major physical and biogeochemical variables with the development of mechanistic
547 numerical models offers the possibility to significantly advance our understanding of the ocean
548 iron cycle and its sensitivity to future environmental changes.

549 **Acknowledgements**

550 We thank the officers and crew of *RV Atlantic Explorer*, the numerous scientific participants in
551 the FeAST cruises, and chief scientists Maureen Conte, John Dacey and Carl Lamborg, who
552 allowed us to piggyback our field sampling on their shipboard programs. This research was
553 primarily funded by the US National Science Foundation through awards OCE-0222053 and
554 OCE-0138352 (to PNS), and OCE-0222046 (to TMC); PNS and ARB also acknowledge support
555 from the Institute of Marine and Antarctic Studies, the Antarctic Climate and Ecosystems CRC,
556 and the University of Tasmania through a Visiting Scholar award, and SJU acknowledges
557 support provided by a European Commission Marie Curie Fellowship (PIOF-GA-2009-235418

558 SOLAIROS). Valery Kosnyrev is thanked for preparation of the altimetric analyses, and DJM
559 gratefully acknowledges support from NSF and NASA. Altimeter products were produced and
560 distributed by AVISO (www.aviso.oceanobs.com/) as part of the Ssalto ground processing
561 segment.

562

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837

838 **Table 1**839 Details of cruises conducted for the FeAST project aboard RV *Atlantic Explorer*

840	841	842	843
Cruise	Cruise period	Water-column DFe stations in area around BATS region	
844	FeAST-1	23-27 April, 2007	1-1, 1-2, 1-3
845	FeAST-2	5-15 July, 2007	2-1, 2-2, 2-3, 2-4, 2-5, 2-6
846	FeAST-3	24-27 September, 2007	3-1, 3-2
847	FeAST-4	5-9 November, 2007	4-1, 4-2, 4-3
848	FeAST-5	28-30 March, 2008	5-1
849	FeAST-6	5-18 June, 2008	6-1, 6-6
850	FeAST-7	21-26 September, 2008	7-1, 7-2, 7-3

852

853

854 **Table 2**

855 Range and mean of DFe concentrations in FeAST near-surface seawater towfish samples

856	857	858	859
Towfish transect	DFe range	DFe mean \pm standard deviation	
860	FeAST-2 tow 1	0.44-0.82 nM	0.63 \pm 0.12 nM
861	FeAST-2 tow 2	0.47-0.83 nM	0.60 \pm 0.10 nM
862	FeAST-2 tow 3	0.47-0.75 nM	0.60 \pm 0.07 nM
863	FeAST-6 tow 1	0.31-0.90 nM	0.59 \pm 0.23 nM
864	FeAST-6 tow 2	0.81-1.13 nM	0.97 \pm 0.09 nM
865	FeAST-6 tow 3	0.84-1.35 nM	1.03 \pm 0.16 nM
866	FeAST-6 tow 4	0.88-1.58 nM	1.13 \pm 0.19 nM

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868

869

870 **Figure Captions**

871

872 **Figure 1.** Map of the BATS region of the Sargasso Sea showing stations where water-column
873 samples were collected for DFe measurements during the FeAST program. Point labelled 1-1
874 corresponds to cruise FeAST-1, station 1, and so on. Also shown is the island of Bermuda, and
875 locations of Hydrostation S (HS) the Tudor Hill atmospheric sampling tower (TH).

876

877 **Figure 2.** Top panels: Vertical concentration profiles of DFe from the seven FeAST cruises in
878 2007-2008 (see Table 1 for cruise dates). Bottom panels: Corresponding profiles showing the
879 average value of DFe concentration versus depth for each of the FeAST cruises. Points in
880 parentheses represent samples for which contamination is suspected.

881

882 **Figure 3. a.** Vertical concentration profiles of DFe in the same mode-water eddy, sampled in
883 April 2007 and July 2007. Location of mode-water eddy (white circle) on **b.** 13 April 2007 (just
884 before FeAST-1) and **c.** 6 July 2007 (during FeAST-2), overlain on corresponding sea level
885 anomaly maps with color scale in mm (analysis performed using altimeter products produced
886 and distributed by AVISO (www.aviso.oceanobs.com/) as part of the Ssalto ground processing
887 segment).

888

889 **Figure 4.** Schematic cross-section of subsurface isopycnals across **a.** cyclonic eddy, **b.**
890 anticyclonic eddy, and **c.** mode-water eddy (adapted from McGillicuddy et al., 2007). Bottom
891 panel shows locations of each type of eddy (white circles) that was sampled during FeAST-2
892 cruise in July 2007, overlain on a map of sea level anomaly from 6 July 2007, with color scale in
893 mm (analysis performed using altimeter products produced and distributed by AVISO
894 (www.aviso.oceanobs.com/) as part of the Ssalto ground processing segment).

895

896 **Figure 5.** Vertical quasi-sections of temperature (top; raw data) and in-situ fluorescence
897 (bottom; raw data) from conventional CTD casts across the cyclonic eddy (CE, 3 crossings),
898 mode-water eddy (MWE) and anticyclonic eddy (ACE) during FeAST-2 cruise in July 2007.

899

900 **Figure 6.** Water column profiles of DFe concentration (top) and temperature and in-situ
901 fluorescence (bottom) for the three types of eddies sampled during FeAST-2 cruise in July 2007:
902 cyclonic (left panels), mode-water (center panels) and anticyclonic (right panels).

903

904 **Figure 7.** Map of DFe concentrations in near-surface towfish samples collected during **a.**
905 FeAST-2 (top) and **b.** FeAST-6 (bottom) cruises.

906

907 **Figure 8.** Vertical concentration profiles of DFe, soluble Fe (sFe) and labile Fe(II) from stations
908 sampled during FeAST-2 and FeAST-6 cruises. Data points shown in parentheses or offscale
909 had measured sFe > DFe, so these sFe samples are considered to have been contaminated.

910

911 **Figure 9.** Vertical concentration profiles of DFe, strong iron-binding ligand (L_1), and weaker
912 iron-binding ligand (L_2) at trace metal station 2 sampled during FeAST-2 cruise.

913

914 **Figure 10.** Multi-model minimum, average and maximum solutions for DFe concentrations over
915 the upper 1000 m at 31°N, 65°W, for April (top panel), July (middle panel) and November
916 (bottom panel) from the ocean iron model intercomparison project (Tagliabue et al., 2016). Our
917 average observed DFe concentrations from April, July and November 2007 are shown as crosses.

918