*** MANUSCRIPT IN PREP FOR GLOBAL BIOGEOCHEMICAL CYCLES ***

2 3 How well do global ocean biogeochemistry models simulate dissolved iron 4 distributions?

- Alessandro Tagliabue¹, Olivier Aumont², Ros DeAth³, John P. Dunne⁴, Stephanie Dutkiewicz⁵,
- 7 Eric Galbraith⁶, Kazuhiro Misumi⁷, J. Keith Moore⁸, Andy Ridgwell^{3,9}, Elliot Sherman⁸, Charles
- 8 Stock⁴, Marcello Vichi^{10,11}, Christoph Völker¹² and Andrew Yool¹³
- 9

1

- 10 1. School of Environmental Sciences, University of Liverpool, Liverpool, UK
- 11 2. IRD-LOCEAN, Institut Pierre Simon LaPlace, 4 Place Jussieu, 75252 Paris Cedex 05, France.
- 12 3. School of Geographical Sciences, University of Bristol, Bristol, UK.
- 4. NOAA/Geophysical Fluid Dynamics Laboratory, 201 Forrestal Road, Princeton NJ 08540,
 USA
- 15 5. Center for Global Change Science, Massachusetts Institute of Technology, Cambridge, MA20139. USA.
- 17 6. ICREA-ICTA, Universitat Autonoma de Barcelona, Bellaterra, Barcelona, Spain
- 7. Environmental Science Research Laboratory, Central Research Institute of Electric Power
 Industry, Abiko, Chiba, Japan.
- 20 8. Department of Earth System Science, University of California, Irvine, CA. USA.
- 21 9. Department of Earth Sciences, University of California, Riverside, CA 92521, USA
- 22 10. Department of Oceanography, University of Cape Town, Cape Town, South Africa
- 23 11. Nansen-Tutu Centre for Marine Environmental Research, Cape Town, South Africa
- 24 12. Alfred-Wegener-Institut, Helmholtz-Zentrum für Polar- und Meeresforschung,
- 25 Bremerhaven, Germany
- 26 13. National Oceanography Centre, University of Southampton Waterfront Campus, European
- 27 Way, Southampton, SO14 3ZH, UK
- 28
- Main Point 1: First intercomparison of 13 global iron models highlights key challenges in
 reproducing new iron data
- 31 *Main Point 2*: Wide uncertainty in iron input fluxes, which results in poorly constrained
- 32 residence times
- 33 *Main Point 3*: Reducing uncertainty in scavenging and biological cycling is a priority

35 Abstract

36

34

- Numerical models of ocean biogeochemistry are relied upon to make projections about the
 impact of climate change on marine resources and test hypotheses regarding the drivers of
- 38 impact of climate change on marine resources and test hypotheses regarding the drivers of 39 past changes in climate and ecosystems. In large areas of the ocean, iron availability regulates
- 40 the functioning of marine ecosystems and hence the ocean carbon cycle. Accordingly, our
- 41 ability to quantify the drivers and impacts of fluctuations in ocean ecosystems and carbon
- 42 cycling in space and time relies on first achieving an appropriate representation of the
- 43 modern marine iron cycle in models. When the iron distributions from thirteen global ocean
- 44 biogeochemistry models are compared against the latest oceanic sections from the
- 45 GEOTRACES programme we find that all models struggle to reproduce many aspects of the
- 46 observed spatial patterns. Models that reflect the emerging evidence for multiple iron sources
- 47 or subtleties of its internal cycling perform much better in capturing observed features than
- 48 their simpler contemporaries, particularly in the ocean interior. We show that the substantial
- uncertainty in the input fluxes of iron results in a very wide range of residence times across
 models, which has implications for the response of ecosystems and global carbon cycling to
- 51 perturbations. Given this large uncertainty, iron-fertilisation experiments based on any single

- 52 current generation model should be interpreted with caution. Improvements to how such
- 53 models represent iron scavenging and also biological cycling are needed to raise confidence in
- 54 their projections of global biogeochemical change in the ocean.
- 55

56 **1. Introduction**

57

58 With the important role played by dissolved iron (DFe) in regulating ocean biogeochemical 59 cycles well established [Boyd and Ellwood, 2010], most three dimensional global 60 biogeochemistry models now include a prognostic DFe tracer as standard. These models explicitly represent the DFe limitation of primary production that is prevalent across large 61 62 areas of the ocean [C M Moore et al., 2013]. This has allowed quantitative projections 63 regarding the impacts of environmental change in Fe-limited regions [Bopp et al., 2013], how 64 DFe may regulate glacial-interglacial changes to the global carbon cycle [*Tagliabue et al.*, 2009] and the wider role played by different nutrients as drivers of planktonic diversity 65 [Ward et al., 2013]. However, the robustness of these results is reliant on how a given model 66 represents the ocean DFe cycle. For example, a model that accounted for hydrothermal 67 68 sources of Fe was shown to be less sensitive to changes in aeolian iron supply than the same model without a hydrothermal input [*Tagliabue et al.*, 2010]. Equally, there is a six-fold 69 70 difference in the estimated impact of dust variations on glacial and interglacial changes in 71 atmospheric CO₂ (5-28 ppm) [Kohfeld and Ridgwell, 2009] that is largely driven by details of 72 the modeled DFe cvcle.

73

74 In brief, the ocean iron cycle is regulated by a complex array of different processes [Boyd and 75 *Ellwood*, 2010]. DFe is thought to be supplied to the ocean from atmospheric deposition 76 [*lickells et al.*, 2005], continental margins [*Elrod et al.*, 2004] and hydrothermal vents 77 [*Tagliabue et al.*, 2010], with potential emerging roles for input from rivers [*Rijkenberg et al.*, 78 2014], icebergs [Raiswell et al., 2008] and glaciers [Gerringa et al., 2012]. DFe is relatively 79 insoluble in oxygenated seawater and DFe levels are maintained to a large part due to 80 complexation with organic ligands that bind Fe [Gledhill and Buck, 2012]. Unbound, or free Fe 81 can then precipitate as solid forms or be scavenged by particles [Bruland et al., 2014]. DFe is 82 operationally defined by the filter size (usually 0.2µm) and over half of the DFe pool can be 83 colloidal [Boye et al., 2010; Fitzsimmons and Boyle, 2014; Wu et al., 2001]. This implies that the 84 aggregation and coagulation of colloidal Fe, termed 'colloidal pumping' [Honeyman and 85 *Santschi*, 1989], may also be an important loss of DFe. As a divalent metal, Fe also undergoes rapid redox transformations between Fe(II) and Fe(III) species mediated by oxidation. 86 reduction and photochemical processes [Wells et al., 1995]. The biological cycling of Fe is also 87 88 complex with varying cellular requirements for Fe [Raven, 1988; Raven et al., 1999] and the 89 role of luxury uptake [Marchetti et al., 2009] driving a wide range in phytoplankton Fe quotas 90 [Sunda and Huntsman, 1997; Twining and Baines, 2013]. Equally, the recycling of DFe by 91 bacteria, viruses and zooplankton is emerging as a key component in governing the Fe supply 92 to phytoplankton [Barbeau et al., 1996; Boyd et al., 2012; Hutchins and Bruland, 1994; Strzepek et al., 2005]. Lastly, process studies and basin scale data syntheses have highlighted 93 94 important specificities to the remineralisation lengths scale and vertical profile of DFe, 95 relative to other nutrients [*Frew et al.*, 2006; *Tagliabue et al.*, 2014c; *Twining et al.*, 2014]. 96 97 The earliest global iron models were informed by the first efforts to synthesise the emerging

97 The earliest global from models were informed by the first enorts to synthesise the emerging
 98 datasets on DFe in the late 1990s [Johnson et al., 1997]. These models only considered a dust
 99 source, applied constant phytoplankton Fe demands and inferred that the seemingly constant

- 100 deep ocean DFe concentrations indicated a threshold stabilisation of DFe by organic ligands
- 101 [*Archer and Johnson*, 2000; *Lefèvre and Watson*, 1999]. As available DFe datasets expanded, it
- 102 became clear that deep ocean concentrations were more regionally and temporally varied

103 than accounted for by these models and that explicitly computing un-complexed DFe led to a better model-data agreement [*Parekh et al.*, 2004]. At the same time, assumptions regarding 104 fixed iron solubility in dust and constant C:Fe ratios in exported organic matter were being 105 106 questioned and alternatives tested [*Ridgwell*, 2001; *Watson et al.*, 2000]. Towards the end of 107 the Joint Global Ocean Flux Study (JGOFS) era more complicated treatments of the demand for 108 DFe from different phytoplankton groups also emerged and when coupled to realistic models 109 of ocean circulation, provided the first estimates of the areal extent of DFe limitation [Aumont 110 et al., 2003; Moore et al., 2002]. In more recent years, and particularly with the advent of the 111 GEOTRACES programme (www.geotraces.org), observations of DFe have expanded rapidly 112 [*Mawji et al.*, 2015; *Tagliabue et al.*, 2012]. This has driven the representation of DFe sources 113 associated with margin sediments [Moore and Braucher, 2008] and hydrothermal vents 114 [*Tagliabue et al.*, 2010] in models. At the same time efforts to account for redox speciation 115 [Tagliabue and Völker, 2011] and variability in Fe binding ligands [Misumi et al., 2013; Völker and Tagliabue, 2015] in global models have also been undertaken. 116 117 118 Until now there has been no comprehensive effort to evaluate how different global models 119 represent DFe, apart from the one off model-data comparisons typical of individual publications [Moore and Braucher, 2008; Tagliabue et al., 2008]. Our maturing vision of the 120 121 oceanic distribution of DFe and our deeper understanding of how it interacts with broader 122 biogeochemical cycles presently allows a more widespread intercomparison of global iron 123 models. In conducting the first 'iron model intercomparison project' (FeMIP) we aim to 124 intercompare as broad a suite as possible of global ocean biogeochemistry models with a

focus on the reproduction of features present in the full depth ocean sections emerging from
the GEOTRACES programme. In doing so we highlight the challenges present for global ocean
biogeochemistry models in simulating the distribution of DFe, which emerges as unique to
that of other nutrients.

128 that of ot 129

130 2. Methodology

131

132 **2.1 Intercomparison process**

133 The goal of this study was to include as many global iron models as possible in order to 134 135 ensure a 'state of the art' view on their representation of Fe cycling. In that regard, our 136 thirteen models (Table 1) range from those used in the recent IPCC report for coupled climate-carbon studies, to those focused on global patterns of Fe cycling and effects on ocean 137 138 biogeochemical cycles and phytoplankton diversity, to those concerned with geological 139 timescales. This inclusive design thus did not impose a rigid set of guidelines regarding the 140 model forcings, as done for the ocean carbon-cycle model intercomparison (OCMIP) and 141 climate model intercomparison (CMIP) projects. While imposing identical ocean circulation 142 or external forcing scenarios would have permitted a more direct cross comparison of the 143 different iron models, the extra constraints would have drastically reduced the number of Fe 144 models able to participate and hinder our aim to account for the full diversity of Fe models. 145 Groups submitted their best representation of the dissolved iron distribution in netCDF 146 format at monthly frequency for a canonical year on their standard model grid, alongside 147 additional requested information (temperature, salinity, nitrate, phosphate and silicic acid 148 concentrations, where available). We compiled model data from thirteen model 149 configurations: BEC [*J K Moore et al.*, 2013], BFM [*Vichi et al.*, 2007], BLING [*Galbraith et al.*, 150 2010], COBALT [Stock et al., 2014], GENIE (Fe scheme as summarised by [Matsumoto et al., 151 2013]), MEDUSA1 [Yool et al., 2011], MEDUSA2 [Yool et al., 2013], MITecco [Dutkiewicz et al., 152 2015], MITigsm [Dutkiewicz et al., 2014], PISCES1 [Aumont et al., 2015], PISCES2 [Resing et al., 2015; Völker and Tagliabue, 2015], REcoM [Hauck et al., 2013] and TOPAZ [Dunne et al., 153

- 154 2013], all implemented at the global scale. All models were then regridded onto a 1° x 1°
- 155 horizontal grid with 33 vertical levels (bounded by 0, 10, 20, 30, 40, 50, 75, 100, 125, 150,
- 156 200, 250, 300, 400, 500, 600, 700, 800, 900, 1000, 1100, 1200, 1300, 1400, 1500, 1750, 2000,
- 157 2500, 3000, 3500, 4000, 4500, 5000 and 5500m) as a common FeMIP grid.
- 158

159 2.2 Observational datasets

160

161 Observations of dissolved iron are taken from two sources. Firstly, we use an updated version 162 of a global DFe database [*Tagliabue et al.*, 2012] with approximately 20,000 individual 163 observations. This database was gridded at monthly resolution on the FeMIP grid to compare models and observations grid cell by grid cell and month by month, with no volume 164 weighting. Secondly, we extracted DFe data from recent GEOTRACES sections from the 2014 165 166 intermediate data product [*Mawji et al.*, 2015]. For comparison purposes (Sec 3.2) the modeled DFe from the longitude, latitude and month of each sampling station was then 167 168 extracted and the observed data was regridded on the same 33 vertical levels as the models 169 (averaging where more than one observation was present in a particular depth bin). We use 170 datasets collected on the GA-02 West Atlantic cruise [*Rijkenberg et al.*, 2014], the GA-03 North Atlantic zonal transect [Hatta et al., 2014], the CoFeMUG south Atlantic zonal cruise [Saito et 171 *al.*, 2013], the GIPY-6 Atlantic sector of the Southern Ocean cruise [*Chever et al.*, 2010; *Klunder* 172 et al.. 2011] and the recently completed GP-16 Equatorial Pacific zonal section [Resing et al., 173 174 2015] that is not vet in the GEOTRACES data product. We note that all IDP2014 GEOTRACES 175 data [Mawji et al., 2015] is also included in the global dataset.

176

177 **2.3 Brief introduction of the different iron models**

178 179 The goal here is not to exhaustively describe the FeMIP models for which we refer to the 180 original publications. Rather we seek to summarise how the models treat important components of the Fe cycle and to highlight important differences (Table 1). In our summary 181 182 we focused on how each model treated the sources of Fe, the chemistry of Fe (including the 183 representation of Fe binding ligands, how free Fe is computed and whether scavenging is a 184 first order rate or a second order function of particle concentrations), biological cycling of Fe 185 (if Fe/C ratios were variable and if zooplankton excretion of Fe depends on the Fe content of 186 prey) and particle Fe dynamics (how many particle pools were simulated and whether the Fe 187 regeneration efficiency was unique or coupled to organic matter). 188

189 All models considered a dust source of Fe and only BFM, GENIE and MEDUSA1 did not 190 consider sedimentary Fe supply, only BEC, BFM, PISCES1 and PISCES1 include river input of 191 Fe, while BEC and PISCES1 and PISCES2 are the only models that represent hydrothermal Fe 192 input. All models except BEC compute the free Fe concentration that can be scavenged based 193 on Parekh et al. [2004] and all except BFM, COBALT, MEDUSA1 and MEDUSA2 have a second 194 order scavenging rate, i.e. a dependency on particle concentrations. Only PISCES1 and 195 PISCES2 include a representation of colloidal losses of dFe, based on aggregation of dissolved 196 organic material [Aumont et al., 2015]. It is notable that despite a maturing understanding of 197 the variations in the concentrations of Fe binding ligands [Gledhill and Buck, 2012], most 198 FeMIP models still assume a constant ligand concentration (as per the earliest Fe models) that 199 is 1 nM for all models except BFM and PISCES1 who use 0.6 nM. Two exceptions in this regard 200 are PISCES2 and TOPAZ. TOPAZ applies an empirical relationship to dissolved organic carbon 201 (DOC) to derive ligand concentrations (5x10⁻⁵ mol ligand per mol DOC). PISCES2 is the only 202 FeMIP model to represent a dynamic ligand pool with explicit sources and sinks [Völker and 203 *Tagliabue*, 2015] and a variable computation of the colloidal Fe fraction [*Liu and Millero*, 204 1999], modified to account for hydrothermal ligand supply [Resing et al., 2015]. BLING

switches off Fe scavenging when oxygen drops below 1 mmol m⁻³ [*Galbraith et al.,* 2010] and both BLING and COBALT reduce the stability of Fe-ligand complexes in the presence of light

- 207 [*Galbraith et al.*, 2010; *Stock et al.*, 2014]. Both the MITecco and MITigsm models cap DFe to a
- 208 maximum value of 1.3 nM with any excess Fe being numerically deleted. Due to the noted
- flexibility in planktonic demands for Fe [Sunda and Huntsman, 1997; Twining and Baines,
- 210 2013], almost all FeMIP models have variable Fe/C ratios, with only MEDUSA1, MEDUSA2,
- 211 MITecco and MITigsm retaining fixed Fe/C ratios. Recycling by zooplankton is variable in
- some FeMIP models and thus dependent on an assumed zooplankton Fe quota, except for
- BEC, BLING, MEDUSA1, MEDUSA2 and REcoM where there is a fixed rate of recycling. Lastly,
- all models include one particulate Fe pool, except PISCES1 and PISCES2 that consider 2 and
- BEC, which represents sinking implicitly (accounting for ballasting). Only COBALT invokes
 reduced regeneration efficiency relative to organic material that elongates the regeneration
- 216 reduced regeneration efficiency relative to organic material that elongates the reg 217 depth-scale beyond that that for sinking organic material [*Stock et al.*, 2014].

3.1 Inter-model differences in dissolved iron distributions and cycling

218

Finally, it is notable several models were only run for a few decades or centuries (BEC, BFM,COBALT, MEDUSA1, MEDUSA2, MITecco and MITigsm), a time comparable to the respective

- residence time of Fe in the model in some cases, making them potentially more sensitive to
- their initial conditions. This issue is discussed in more detail in Sec. 3.1.1.

224 **3. Results**

225 226

227

228 **3.1.1 Iron fluxes and residence times**

229 230 Beginning with an integrated view, there is substantial variability in the modeled Fe residence 231 times across the FeMIP models with two broad groupings of a few years and a few hundred vears (Table 2). Across the thirteen models, all include dust sources, ten include sediment 232 233 sources, but only three include hydrothermal and riverine Fe sources, respectively (Table 2). 234 Even for a given source, there is substantial inter-model difference in its strength. For 235 example, dust fluxes of dissolved iron range from ~ 1 to >30 Gmol Fe yr⁻¹ between models (Table 2, accounting for any inter-model variations in solubility and mineral fraction). These 236 237 inter-model differences across all input fluxes result in a wide range of total iron inputs to the ocean (66.9±67.1 Gmol Fe yr⁻¹, Table 1). In contrast we find a surprising degree of agreement 238 239 in the mean ocean iron concentration (0.58±0.14 nM, Table 2) from the models, with slightly 240 greater inter-model differences in the total integrated inventory of Fe reflecting different 241 model grid sizes (e.g. some models do not include the Arctic Ocean or the Mediterranean). 242 Ultimately this results in a wide range of residence times of dissolved iron in the models (~5 243 to > 500 years, Table 2) that reflects different assumptions regarding the strength of the 244 sources of DFe to the ocean, compensated by variable scavenging rates in order to reproduce 245 the observed DFe concentration. 246

247 The derivation of the residence time for Fe from each model allows us to evaluate the impact 248 of the shorter runs performed for some models. Taken at face value, even the relatively short 249 runs performed by almost all the models (except BFM, MEDUSA1 and perhaps also MEDUSA2) 250 are more than twice the residence time for Fe in that particular model. Nevertheless, it should 251 be noted that many of these residence times for the global ocean are likely skewed towards 252 lower values due to strong local sources that have a muted wider influence. For example, 253 much of the interior Fe distribution in the PISCES1 model has been shown to be linked to a 254 subducted preformed component [*Tagliabue et al.*, 2014b], suggesting that the deep ocean equilibration timescale in this model, at least, must be much longer than the 11 years of its 255

256average residence time. This is likely to be the case for models that employ a formulation for257the rate of DFe scavenging that depends on particulate fluxes, as biogenic fluxes in the ocean258interior are considerably slower than near the surface where sedimentary and dust sources259are dominant. Feedbacks will also exist between DFe inventory and biological fluxes, meaning260that a ~1000 yr time-scale component to the overall equilibrium adjustment will exist that261involves the redistribution of major nutrients globally. As such, this raises questions

- regarding the distributions of Fe in the ocean interior for models that are only run for a few
- 263 decades, even if that is longer than the average residence time.
- 264

265 **3.1.2 Statistical assessment of FeMIP models**

266 267 In order to provide a general picture of variability amongst the models, we examine 268 correlations between observed and simulated DFe at the same locations (Table 3). When 269 viewed globally throughout the entire water column, correlations between observations and the models can be as high as 0.51, while some are even anti-correlated. The mean biases 270 271 against observations are between -0.02 and -0.48 nM. In the 0-100m depth stratum, where Fe 272 is likely be to playing a role in regulating phytoplankton growth rates, all but one of the model 273 correlations fall between 0.33 and 0.48, implying no clear link between model complexity and 274 strength of correlation. On the other hand, the mean biases range from -0.29 to 0.67 nM, 275 which is suggests less overall agreement in the absolute DFe levels. The 100-500 m depth 276 slice has the overall highest correlations, and all but three models reach their highest 277 correlations in this depth range. In the abyssal layers only the three models that consider 278 hydrothermal iron input (BEC, PISCES1 and PISCES2) show a reasonable correlation with 279 observations (R=0.20 to 0.35, other models are < 0.15), highlighting the importance of this 280 source in the deep ocean. However, the inclusion of hydrothermal iron input does not obviously lead to a significant improvement in the surface ocean. Similarly, including (or not) 281 sedimentary Fe input does not seem closely linked to reproducing observations in the surface 282 or intermediate layers. For example the two versions of MEDUSA with and without 283 284 sedimentary iron input do not show much difference in their correlation coefficients. It is also 285 important to note that we lack substantial coastal DFe datasets where sediments and/or river 286 supply results in high DFe levels in a number of models (see Sec 3.1.3). Section 3.2 will more closely examine the different models using recent large-scale GEOTRACES sections as case 287 288 studies in different ocean regions. 289

290 **3.1.3 Inter-model differences in dissolved iron**

291 292 To examine the inter-model differences in dissolved iron in more detail, we compare the 293 model mean DFe over the 0-100m, 100-500m, 500-1000m and 2000-5000m depth slices, 294 repeating the analysis for the boreal (30-90N), tropical (30N-30S) and austral latitudes (90S-295 30S). This enables us to group the models into 'high', 'moderate' and 'low' in terms of their 296 DFe distribution, relative to the full model suite (Figure 1). Comparing Figure 1 with the 297 statistical summary (Table 3) suggests that the inter-model trend in the average DFe 298 concentration for the different depth slices does not always reflect good statistical agreement 299 with the observations. However, it should be noted that while the inter-model trends in 300 average DFe reflect full spatial and temporal averages, the statistics determined from 301 observations only concern locations with available DFe observations (which is not spatially 302 and temporally complete).

303

Beginning with the surface ocean (0-100m) that is heavily influenced by surface sources and biological uptake. MEDUSA1, MEDUSA2 and TOPAZ are consistently relatively high in iron for all three latitudinal zones, including the Fe limited Southern latitudes. BEC is also relatively

- 307 rich in Fe, but only in the northern and tropical latitudes. The lowest DFe concentrations in all
- 308 three geographic zones are simulated by the BLING, COBALT and MITigsm models, with the
- 309 remaining models intermediate throughout.
- 310
- 311 The relative tendencies between the different FeMIP models are generally conserved in the
- 312 100-500m and 500-1000m depth slices that are more heavily influenced by remineralisation
- 313 processes. Notable departures from this general trend are PISCES2 displaying relatively
- higher DFe levels in both depth bins. While both BFM and REcoM become more DFe rich in the 500 1000m doubt him TODAT standard him FDDAT
- the 500-1000m depth bin, TOPAZ stands out less as a high DFe model. In terms of
- hemispheric contrasts, BEC becomes lower in DFe in the Southern region; otherwise the inter-model trends are preserved.
- 318
- In the deepest depth bin deep ocean sources such as hydrothermal vents, as well as sediments
 are important. Unsurprisingly, the models that include hydrothermal vent DFe sources (BEC,
 PISCES1 and PISCES2) show high DFe levels. In contrast, the high DFe levels for BFM,
- MITecco, MITigsm and REcoM cannot be ascribed to hydrothermal DFe input and may be
- 322 MITecco, MITigsm and REcoM cannot be ascribed to hydrothermal DFe input and may 323 related to initial conditions (e.g. for BFM) or deep ocean transport of high DFe levels.
- However, it is notable that BFM, MITecco, MITigsm and REcoM do not perform well
- 524 nowever, it is notable that BFM, MITECCO, MITIgsm and KECOM do not perform well 325 statistically in this depth range (Table 3). The RUNG and MEDUSA1 models simulate th
- 325 statistically in this depth range (Table 3). The BLING and MEDUSA1 models simulate the
- lowest concentrations in this depth bin. For a large number of models (BLING, GENIE,
 MEDUSA1 MEDUSA2 TOPA7) DEconcentrations decline in the 2000 E000m bin relations.
- MEDUSA1, MEDUSA2, TOPAZ), DFe concentrations decline in the 2000-5000m bin, relative to
 the 500-1000m bin.
- 329

3.1.4 Surface DFe distributions in the models

330 331

332 Due to its role as a limiting nutrient, we explore the simulated annual mean surface DFe 333 concentrations from the FeMIP models in more detail (Figure 2, upper 50m average). Here we 334 see that, as suggested by the range in the model biases (Table 3), there is a substantial degree 335 of inter-model discord in the surface Fe distributions. Most models agree that the highest DFe 336 concentrations are found underneath the Saharan dust plume in the tropical Atlantic, but 337 others also emphasise dust supply into the Arabian Sea and enhanced DFe along the 338 continental margins. A large number of the models suggest the lowest DFe concentrations are 339 found across the Pacific Ocean. Exceptions are GENIE and MEDUSA1, who have much higher 340 DFe concentrations therein and BEC, MEDUSA2 and TOPAZ, who restrict low DFe to the south 341 Pacific only. BFM, MITecco and MITigsm have a very DFe deplete sub-Arctic Pacific that is not 342 as extreme in the other FeMIP models. When the seasonality in DFe (presented as the maximum minus minimum DFe concentration over the year, Figure 3) is compared, strong 343 344 inter-model differences also emerge. For example, some models show remarkably little 345 seasonality (BFM, GENIE, MEDUSA1, MEDUSA2 and MITigsm), whereas others have large seasonal cycles over wide areas (>0.5nM, BEC, MITecco, PISCES1, PISCES2 and TOPAZ). This 346 illustrates where high annual mean concentrations in these regions are masking strong 347 seasonal minima. For this reason it is not straightforward to compare the models against 348 349 observed Fe that might have been collected during different seasons. At this stage, incomplete sampling over the seasonal cycle is prevalent for virtually all locations with DFe 350 351 measurements [*Tagliabue et al.*, 2012], which precludes the mapping of DFe seasonality from observations. Table 3 is therefore more suited for a statistical assessment of the surface DFe 352 353 for a given model against all available observations (where seasonal variations are accounted 354 for by comparing model and data DFe at identical longitudes, latitudes, depths and months). 355

356 **3.2 Comparison to recent GEOTRACES ocean sections**

357

358 To more closely examine how the different DFe models represent the observed distribution of

- 359 DFe we focus on a range of recent GEOTRACES sections. As described above (Sec 2.2) each
- 360 model is extracted at the exact location of the sampling locations, with the observations
- regridded onto the same vertical grid. We refer the readers to the below cited papers for a
- 362 more complete discussion of each observational section and additional interpretation. In this
- 363 assessment we emphasise the key features observed on each section and how different
- 364 models are able to reproduce them. Because of this goal and because a given model may do a 365 good job of reproducing one feature, but not another, we did not perform statistical
- 366 assessments of the individual models for each section.
- 367

368 3.2.1 West Atlantic

369

The GA-02 West Atlantic meridional section provides unprecedented coverage of DFe concentrations along the Atlantic Ocean, as well as insights into different mechanisms that control the cycling, regeneration and supply of DFe [*Rijkenberg et al.*, 2014]. The key features of this section are (i) low surface DFe in both the northern and southern end member surface

- waters, (ii) a surface DFe enrichment around 20°N in the tropics and associated with a subsurface DFe minima (iii) a strong DFe regeneration maying at 5, 10°N centered around 20°N in the tropics and associated with a
- subsurface DFe minima, (iii) a strong DFe regeneration maxima at 5-10°N centered around (iv) a hydrothermal signal at around 5°S and between 2000 2000m death and
- 500-1000m, (iv) a hydrothermal signal at around 5°S and between 2000-3000m depth and
 (v) a hotspot of DFe that is present over much of the water column associated with the
- 378 confluence of the Brazil and Falklands current at around 35-40°S.
- 379

380 *Model representation of key features* (Figure 4): (i) Almost all models capture low DFe in the 381 Southern end member surface waters, except MEDUSA1 and MEDUSA2 and perhaps also 382 REcoM and TOPAZ. However it is only in BFM and COBALT, and to a lesser degree BEC, BLING, MITigsm, PISCES1 and PISCES2 that reproduce the observed low DFE concentrations 383 384 associated with the northern endmember surface waters. (ii) A surface DFe enrichment 385 (presumably from dust) around 20°N is clearly present in BEC, MEDUSA1, MEDUSA2, 386 MITecco, MITigsm, PISCES1, PISCES2, REcoM and TOPAZ, but is less apparent in other models 387 (BFM, BLING, COBALT, GENIE). Nevertheless, in MEDUSA2, REcoM and TOPAZ the influence 388 of surface dust deposition appears to be much greater than is observed. Only PISCES1, PISCES2 and COBALT, show the observed subsurface minima in DFe below the dust signal. 389 390 (iii) With respect to the strong DFe regeneration maxima at 5-10°N centered around 500-391 1000m, COBALT displays a regeneration maximum at around the right depth level, while in 392 BEC high concentrations appear to be smeared from surface to the sea floor. In all other 393 models the regeneration signal in DFe is generally too small or absent and where it is present 394 (e.g. BFM, BLING, GENIE) it is generally too shallow in the water column. (iv) Concerning the 395 hydrothermal signal at around 5°S and between 2000-3000m depth, of the three models that 396 include hydrothermal DFe input, only PISCES2, with a greater longevity of hydrothermal Fe 397 [Resing et al., 2015], shows a hint of DFe enrichment in the right location. MEDUSA2 398 underestimates DFe in the ocean interior along the entire Atlantic section. (v) No models 399 capture the elevated DFe over almost the entire water column around 35-40°S. In the 400 observations, this is ascribed to the offshore export of Brazilian shelf waters or DFe input 401 from the dissolution of particulate Fe associated with the Rio de la Plata river [Rijkenberg et 402 al., 2014].

402 403

404 **3.2.2 Subtropical North Atlantic**

405

The GA-03 North Atlantic zonal section crossed the subtropical North Atlantic between Cape
Verde and Woods Hole (USA) via Bermuda. Key signals in the dataset [*Hatta et al.*, 2014] are

408 (i) strong enhancements in DFe associated with DFe regeneration and also coastal input along

the eastern and western margins, (ii) a surface enrichment along with a subsurface minimumin DFe and (iii) a strong hydrothermal anomaly over the mid Atlantic ridge.

411

412 *Model representation of key features* (Figure 5): (i) Enhanced DFe in the subsurface along the

- 413 margins is represented to different degrees by the FeMIP models. BLING, COBALT, MITecco
- and PISCES1 have hints of subsurface maxima in DFe along the eastern margin. It is
- 415 encouraging that the addition of ligand production during remineralisation in PISCES2 clearly
- 416 improves the intensity of the remineralised DFe signal. However, none of these models have a
- 417 broad homogenous signal (down to > 2000m) of elevated DFe that is observed on the eastern 418 margin, except perhaps BEC, which has a strong subsurface maximum that spreads over all
- 419 depth levels. (ii) The subsurface minima in DFe underlying a surface (presumably dust)
- 420 enrichment is captured clearly by COBALT, PISCES1 and PISCES2 and slightly less clearly by
- 421 BEC, BFM and BLING. (iii) A hydrothermal anomaly is present in PISCES1, but closer in
- 422 magnitude to the observations in PISCES2, while BEC also displays a strong hydrothermal
- 423 signal. COBALT displays a sediment signal at depth that is not reproduced by the
- 424 observations. It also notable that many of the models present an 'inverted' DFe profile, with
- 425 decreasing DFe concentrations towards the ocean interior (GENIE, MEDUSA1, MEDUSA2,
- 426 REcoM and TOPAZ), which could be indicative of too great a residence time for DFe at the
- 427 ocean surface. Also, BLING, COBALT, MEDUSA2 and TOPAZ seem to be systematically too low428 in terms of their interior ocean DFe levels across this section.
- 429

430 3.2.3 Subtropical South Atlantic

431

The CoFeMUG section traversed the south Atlantic between Namibia and Brazil and had the
following notable signatures [*Noble et al.*, 2012; *Saito et al.*, 2013]: (i) a remineralisation
signal and/or sediment input on the eastern margin, (ii) low overall surface concentrations
and (iii) a strong hydrothermal signal at depth.

436

437 *Model representation of key features* (Figure 6): (i) Interestingly, more models are able to 438 simulate a remineralisation signal on the eastern side of the basin (COBALT, MEDUSA1, 439 MEDUSA2, MITecco, PISCES1, PISCES2, REcoM and TOPAZ) for this section than for the GA03 440 section. Although for some models this feature is too weak or spread over too many depth 441 levels. (ii) All models, except MEDUSA1, MEDUSA2, REcoM and TOPAZ, are able to reproduce the overall low DFe conditions in the surface waters. (iii) BEC and PISCES1 represent a DFe 442 443 anomaly over the ridge as observed, but this is underestimated. PISCES2 represents a 444 stronger hydrothermal signal, but it appears to spread too far off-axis relative to that 445 observed. Again, COBALT displays a strong sediment signal in the deep ocean that is not 446 observed. BFM, BLING, MEDUSA2 and to some extent TOPAZ underestimate interior ocean 447 DFe levels.

448

449 **3.2.4 Southern Tropical Pacific**

450

The GP-16 cruise ran from Ecuador to Tahiti [*Resing et al.*, 2015] and displays the following
key features: (i) DFe enrichment along the eastern margin over almost the entire water
column, (ii) low surface concentrations and (iii) a remarkable hydrothermal plume
propagating westward for > 4000km from the East Pacific Rise to at least 150°W.

455

456 *Model representation of key features:* (Figure 7), (i) BEC, COBALT, PISCES2 and TOPAZ are the
457 only models able to produce the broad signal of elevated DFe throughout the entire water
458 column on the eastern margin. BLING, MEDUSA1, MEDUSA2 and REcoM display an

459 enrichment in DFe but this remains more tightly localised than observed. (ii) All models

- 460 capture the low DFe levels typical of Pacific surface waters, but for some models (BFM, BLING,
- 461 COBALT, GENIE, MEDUSA2 and TOPAZ), low DFe is also too prevalent in the ocean interior.
- 462 (iii) BEC and PISCES1 capture a local hydrothermal signal above the East Pacific Rise, but only
- 463 PISCES2 goes any way towards reproducing the degree of off axis transport. As seen
- 464 previously, MITigsm and COBALT show DFe increases near the sea floor, but these are more
- widespread than seen in the observations. As noted previously, BFM, BLING COBALT,
- 466 MEDUSA2 and TOPAZ show too little DFe in the ocean interior (<0.3nM), relative to the
- 467 observations (>0.6 nM away from the hydrothermal plume).468

69 **3.2.5 Southern Ocean – Atlantic Sector**

469 470

Both the GIPY-4 and GIPY-5 cruises ran from Cape Town (South Africa) to the Antarctic
continent along the so-called 'GoodHope' line during the International Polar Year [*Chever et al.*, 2010; *Klunder et al.*, 2011]. These cruises sampled at different resolutions north and south
of the Polar Front and have been blended to form one section. Notable features in this dataset
include (i) low but non zero concentrations at the surface that propagate into the subsurface,

- 476 (ii) a strong remineralisation signal at around 500m near 60S and (iii) a strongly local
- 477 hydrothermal signal over the Bouvet region ridge crest at around 54°S and more widespread
 478 elevated DFe in the abyssal ocean north of the ridge (i.e. between ~54°S and the northern end
- 479 of the transect.480
- 481 Model representation of key features: (Figure 8), (i) Most models display low overall DFe concentrations at the surface. GENIE, MEDUSA1, MEDUSA2 and to a lesser degree REcoM and 482 483 TOPAZ over estimate surface DFe concentrations. But even the models that have low surface 484 DFe show rapid increases with depth, indicating that the ferricline is too shallow in all models. (ii) No FeMIP model captures the remineralisation signal seen in the subsurface just south of 485 the Polar Front. (iii) Despite including a hydrothermal source, BEC is unable to represent the 486 487 local hydrothermal enrichment. While PISCES1 represents a slight hydrothermal anomaly 488 that appears to be from an adjacent source, the longer lifetime of hydrothermal Fe in PISCES2 489 leads to the anomaly being too widespread in the abyssal ocean. On the other hand, both BEC 490 and PISCES2 do show elevated DFe in the abyssal ocean north of the main ridge at 54°S that 491 compares well with the data. COBALT, MITecco and MITigsm again show a sediment signal in
- DFe at depth, while COBALT and TOPAZ show very high values near the Antarctic coast. None
 of these features are observed in the dataset. The BFM stands out from the other models with
 the large underestimation of DFe in the Southern Ocean interior as already seen for the GA-02
 section.
- 496497 **4. Discussion**

498499 **4.1 Examining inter-model differences in Fe distributions relative to other nutrients**

500 501 In short, we find a wide range of simulated DFe distributions from current global ocean 502 biogeochemical models that reflects an apparent lack of inter-model agreement in the 503 processes that control the oceanic distribution of DFe. When assessed against the best DFe 504 datasets, most models perform modestly both quantitatively in terms of magnitudes and 505 patterns, and qualitatively in representing the inferred mechanisms. This has important 506 implications for how models are used to understand biogeochemical cycles [Galbraith et al., 507 2010; Moore et al., 2002; Tagliabue et al., 2014a], planktonic diversity and resource 508 competition [Dutkiewicz et al., 2012; Ward et al., 2013], as well as the ocean response to 509 fluctuations in the environment in general [Bopp et al., 2013; Dutkiewicz et al., 2013; Tagliabue et al., 2009]. It is noteworthy that this inter-model disagreement appears to be 510

solely driven by the particular way in which different models represent the Fe cycle. If we
examine the models in terms of macronutrients (nitrate and phosphate) then, taking the long

513 meridional GA02 section as example, we see a much stronger inter-model and model-data

agreement (Figures 9 and 10). Although inter-model differences due to specific physical
models are visible in the Atlantic water mass structure, the mechanisms driving the N and P

516

539

cycles are similar.

517 518 We further contextualise the inter-model Fe differences by examining how they represent the 519 relative inventories of Fe and NO₃ in the ocean interior by plotting the Fe* tracer (Fe – 520 $NO_3 * r_{Fe/N}$). Defining $r_{Fe/N}$ in the same way as for the GA02 section [*Rijkenberg et al.*, 2014] 521 (based on the observed Fe:apparent oxygen utilisation relationship, which results in a Fe/N 522 ratio of 0.47 mmol/mol) and using PO₄ (and a NO_3/PO_4 ratio of 16/1) for GENIE and BLING, 523 which do not simulate NO₃, allows us to examine DFe concentrations relative to NO₃, (Figure 11). The data shows relatively replete waters originating from the northern hemisphere 524 linked to North Atlantic Deep Water (NADW), which becomes flanked above and below by 525 relatively Fe poor water from the southern hemisphere linked to Antarctic Intermediate 526 527 Water (AAIW) and Antarctic Bottom Water (AABW). There is also a zone of relatively 528 depleted Fe in the subsurface overlying the NADW signal in the northern hemisphere likely 529 linked to northern subtropical mode water. In these sections we can see that NADW is relatively impoverished in DFe in MEDUSA1, MEDUSA2 and TOPAZ, despite these models 530 531 generally overestimating surface DFe. This may indicate an overly short lifetime for Fe away 532 from the surface and subsequent lack of permanence in the NADW signal. Looking at 533 southern sourced waters, all models except BFM perform well (notwithstanding the northern 534 sourced water biases). Obviously, this comparison should only be taken as indicative since 535 different models are underpinned by different relationships between NO₃ and Fe and the 536 actual planktonic Fe:N ratio can vary from the value chosen in the *Rijkenberg et al.* [2014] 537 study [*Twining and Baines*, 2013]. Nevertheless, it does provide an additional means to assess 538 the relative transport of Fe and NO₃ through the ocean interior.

540 **4.2 Identifying the key processes at different depth strata**

541 One important inter-model difference that clearly impacts the agreement with observations 542 543 and the role of Fe on biota is the strong surface enrichments evident in some models 544 (MEDUSA1, MEDUSA2, REcoM and TOPAZ). In the observations, any Fe enrichments due to dust deposition are far more localised and apparently short lived in space (e.g. Figures 4 and 545 546 5). For the models surface overestimation of iron implies either too large an iron source or 547 that the residence time for Fe at the surface is too long. The latter possibility highlights the 548 importance of how models treat the scavenging process and could also be linked to constant 549 Fe/C ratios that do not permit 'luxury uptake' of Fe at high DFe concentrations (specifically 550 MEDUSA1, MEDUSA2, MITecco and MITigsm). MEDUSA1, MEDUSA2 and REcoM are three of 551 the four models with the longest residence times (decades to centuries, Table 2), relative to 552 the other FeMIP models, and produce high surface enrichment despite having some of the 553 lowest dust inputs (Table 2). For MEDUSA1 and MEDUSA2 the first order fixed scavenging 554 rate may be too low or have not enough variability to remove Fe rapidly when concentrations 555 are high. The constant Fe/C ratios used in these two models may also contribute to this 556 anomalous feature. In REcoM, Fe/C ratios are variable and the scavenging is second order, but 557 may simply be too low. DFe in TOPAZ has one of the shortest residence times (~8 years, Table 2), which implies that the surface accumulation of DFe may instead be linked to relatively 558 559 large sources or the variable ligand concentration. Since the ligand concentration in TOPAZ 560 depends on DOC, which typically decays from surface to deep, there may be too much DFe stabilisation occurring in the surface ocean. 561

562 563 At intermediate depths, the inclusion of a prognostic Fe binding ligand pool with a particle degradation source [Völker and Tagliabue, 2015] clearly improves the reproduction of 564 565 subsurface maxima in DFe associated with remineralisation (compare PISCES2 with PISCES1) 566 for many of the transects. Other models (COBALT and to a lesser degree BEC and BLING) are 567 able to reproduce these features but evidently do so for different reasons. These may be 568 related to the implicit formulation of particle flux (BEC) that ignores lateral transport of 569 particulate Fe or the shutdown of Fe scavenging in low oxygen conditions (BLING). It is 570 interesting that there appears to be two groups of subsurface DFe maxima seen in the 571 observations. Sometimes these features are tightly constrained to a small depth stratum (e.g. 572 equatorial ocean for GA-02, western margin on GA-03 and eastern margin on CoFeMUG), 573 while in other locations the DFe enrichments span almost the entire water column (eastern 574 margins on GA-03 and GP-16). Most models represent one or the other. For example, 575 subsurface maxima are always tightly bounded in depth for some models (e.g. COBALT and PISCES2) or spread over depth in others (BEC) with no regional variations. Future work 576 577 should explore the potential mechanisms involved, which might be linked to subsurface 578 dissolution of dust, nutrient trapping or impacts of low oxygen. Emerging Fe isotope work 579 highlights the potential for non-reductive Fe release from margins [Conway and John, 2014; 580 *Homoky et al.*, 2013] in addition to the role of reducing sediments represented in models.

581

582 In the ocean interior the best models (in terms of their linear correlation coefficients) are 583 those that include hydrothermal input (Table 3). While including such a source is clearly 584 important, it is possible that this is overemphasised in the correlations at the expense of other 585 deep ocean structure that is evident in many of the sections. For example, many of the ocean 586 sections do not show any 'watermass' related structure for DFe that is seen in macronutrients 587 (e.g. Figures 9 and 10). Although adding a hydrothermal ligand seems to improve the ability of 588 PISCES2 to reproduce the GP-16 data (Figure 7) and perhaps also the GA-02 hydrothermal 589 signal (Figure 4), it results in too widespread a hydrothermal anomaly in the Southern Ocean 590 (Figure 8) indicating too long a lifetime for this pool and the need for further refinement of 591 the processes governing hydrothermal Fe input [*Tagliabue*, 2014].

592

593 **4.2 Inter-Model differences in DFe inputs and cycling: the importance of scavenging**

594

595 It is notable that there is a great deal of variability in both the total Fe input flux (66.9 ± 67.1) 596 Gmol Fe yr⁻¹) and the strength of a given source across the models, yet the mean ocean DFe is 597 strikingly similar (0.58±0.14 nM). To some extent, this agreement reflects the calibration of 598 scavenging rates to represent global average iron concentrations in agreement with 599 observations. While this relative homogeneity in modeled mean DFe would be consistent with 600 an earlier view of the oceanic Fe inventory [Johnson et al., 1997], if anything, the emerging 601 oceanic sections of DFe as part of the GEOTRACES programme have highlighted an 602 unexpected variability in DFe distributions in the ocean interior [Mawji et al., 2015]. This is in 603 stark contrast to the other main limiting nutrients, which more closely reflect large-scale 604 ocean circulation patterns and watermass related features (e.g. Figures 9 and 10). Thus the 605 apparent small differences in the mean ocean DFe between models more likely arises from a 606 modeling community that reflects an earlier parsimonious view of the system. The relative 607 constancy in the mean ocean DFe concentrations in the models may reflect homogenous 608 ligand concentrations of either 0.6 or 1.0 nM, but we note that even models with varying 609 ligand concentrations (PISCES2 and TOPAZ) show too much interior ocean uniformity. 610

In contrast to the mean DFe, there is a substantial degree of inter-model disagreement in thestrength of different sources. For instance, BFM, BLING, GENIE, MEDUSA1, MEDUSA2,

613 MITecco, MITigsm and REcoM all have atmospheric input fluxes of < 5 Gmol Fe vr⁻¹, whereas as in BEC, COBALT, PISCES1, PISCES2 and TOPAZ dust supply is much higher (> 20 Gmol Fe 614 yr⁻¹). Yet this does not drive a similar trend in mean ocean DFe (with MITecco, MITigsm and 615 616 REcoM showing amongst the highest DFe concentrations. Table 2). We note that these 617 represent the total DFe flux from dust, accounting for model specific Fe mineralogy and 618 solubility. Equally, for those models that include sedimentary Fe input, this flux term can 619 range from very small (e.g. < 5 Gmol Fe yr⁻¹ in MEDUSA2 or REcoM) to very large (> 70 Gmol 620 Fe yr⁻¹ in BEC, COBALT, MITecco, MITigsm and TOPAZ). Again this does not map onto mean DFe trends. We note that the closer agreement for hydrothermal Fe input is more likely to 621 622 reflect the fact that only two models actually include this term, rather than greater confidence regarding the actual flux. Overall, the total input of DFe does not explain the inter-model 623 624 variations found in mean DFe (R^2 =0.06). This implies that there must be a great deal of 625 variability in how each model treats the scavenging of Fe in order to ultimately arrive at a 626 relatively similar mean ocean DFe concentration.

627

628 Most early Fe models that explicitly computed free Fe and sought to represent its scavenging 629 by sinking particles, treated the scavenging rate constant as a tunable parameter [Archer and Johnson, 2000; Johnson et al., 1997; Parekh et al., 2004; Watson et al., 2000]. This was viable in 630 these relatively simple box models against few observations, but is a less straightforward 631 632 solution for the multi tracer/process 3D biogeochemical models used presently where 633 scavenging itself maybe a function of other model parameters (e.g. particle concentrations) 634 and hence can vary considerably in space. Despite the long acknowledged influence of the particle concentration on the scavenging rate [Honeyman et al., 1988], a subset of the FeMIP 635 636 models persist with a globally uniform scavenging rate (Table 1). However, even for those 637 models that have implemented a second order scavenging rate, there is a question of how this 638 should operate. For example, should the model rely only on organic carbon or also include 639 biogenic silica and calcium carbonate? Non biogenic particles, such as dust, as well as Fe and manganese oxides, may also be important as Fe scavengers [*Haves et al.*, 2015; *Wagener et al.*, 640 641 2008; Ye et al., 2011]. There is also the important question of the specific affinity for free Fe for these various carrier phases. Once Fe is scavenged onto particles, desorption of Fe will be 642 643 important in resupplying the DFe pool. Some models consider constant desorption rates [Moore and Braucher, 2008], while others explicitly account for disaggregation dynamics and 644 645 the impact of bacterial activity [Aumont et al., 2015]. Finally, there is the question of regional and temporal variability in colloidal dynamics. Only one group of FeMIP models attempt to 646 account for this process (Table 1), yet given the apparent importance of colloidal Fe within 647 648 the DFe fraction [Boye et al., 2010; Fitzsimmons and Boyle, 2014; Wu et al., 2001], colloidal 649 pumping losses might be as large as those from the scavenging of free Fe. Some progress may 650 be made by exploiting the legacy from the field of Thorium (Th) cycling, for which a number of 651 different theories have been developed to describe its scavenging, including colloidal 652 components [Anderson, 2003; Burd et al., 2000; Lam and Marchal, 2015; Marchal and Lam, 653 2012; Savoye et al., 2006]. With an expanding database of paired Fe and Th observations, 654 including the particulate phase, as part of GEOTRACES [Mawji et al., 2015] it may be possible 655 to refine this crucial component of the Fe cycle in the coming years.

656 657

4.3 Impact of Fe on wider biogeochemical cycles: the importance of biological Fe cycling

658
659 The biological cycling of DFe in a given model will dictate the net influence of a model's DFe
660 cycling on wider biogeochemical cycling and air-sea CO₂ exchange. In that regard, the large
661 oceanic sections, focused process studies and laboratory experiments all provide essential
662 and complementary information. For example, early laboratory studies demonstrated a large
663 degree of flexibility in the phytoplankton Fe/C ratios as a function of DFe levels and cell size,

as well as enhanced Fe/C ratios at lower light levels [Sunda and Huntsman, 1997]. Similar 664 ranges in Fe/C ratios are also seen in single cell analyses of phytoplankton from the ocean 665 [Twining and Baines, 2013]. The enhanced Fe/C ratio seen at low light is thought to reflect so-666 667 called 'biodilution', where Fe uptake continues when phytoplankton carbon fixation is light 668 limited, and/or a greater absolute demand for Fe at low light [Sunda and Huntsman, 1997; 669 Sunda and Huntsman, 1998]. Almost all FeMIP models permit flexibility in the Fe/C ratio of phytoplankton (Table 1), with those that consider Fe uptake independent of C fixation able to 670 671 account for any biodilution and the BLING model considers a direct impact of Fe on 672 photosynthesis. Emerging recent work has suggested that there are important inter-specific 673 differences in how phytoplankton Fe demands respond to light [Strzepek et al., 2012]. In their 674 laboratory study, Strzepek et al. [2012] found that while temperate diatom species indeed 675 showed elevated Fe/C ratios at low light, the opposite was true for Antarctic diatom species. 676 This raises questions about how models that generally do not consider different species 677 specifically (but rather represent broader 'functional types') can account for these potentially important regional distinctions in how environmental variations impact biological Fe cycling. 678 679 680 Detailed process studies, mostly from the Southern Ocean, have sought to quantify Fe cycling at the ecosystem level. In doing so, the importance of regenerated Fe in the fuelling of 681 biological productivity via the so-called 'ferrous wheel' has emerged as potentially important 682 [Bowie et al., 2009; Bowie et al., 2015; Boyd et al., 2012; Boyd et al., 2005; Sarthou et al., 2008; 683

- 684 Strzepek et al., 2005]. This has been demonstrated via the development of the 'fe-ratio', which 685 represents the proportion of Fe uptake from 'new' Fe sources. It has been determined for sites across the Southern Ocean by assembling Fe budgets that combine measurements of Fe pools 686 and fluxes alongside laboratory estimates. The *f*e-ratio is generally around 0.1 (i.e. strongly 687 688 reliant on recycled Fe) in the low productivity regions of the Southern Ocean [Bowie et al., 689 2009; Boyd et al., 2005] and reaches around 0.5 and greater (i.e. less reliant on recycled Fe) in 690 the naturally fertilised Kerguelen Island phytoplankton bloom [Bowie et al., 2015; Sarthou et 691 al., 2008]. Langrangian process studies have demonstrated a strong seasonal decline in the fe-692 ratio as the spring phytoplankton bloom declines [Boyd et al., 2012], which are consistent 693 with low rates of Fe input during summer [Tagliabue et al., 2014c]. In agreement, direct 694 measurements of Fe fluxes between various components of the food web have highlighted 695 that only regenerative fluxes can support the measured Fe demand [Boyd et al., 2012;
- 696 Strzepek et al., 2005; Tagliabue et al., 2014c].
- 697

698 The sensitivity of a given model's biological productivity to new or regenerated forms of Fe is 699 crucial, as this will underpin its sensitivity to change. At present we do not know if the FeMIP 700 models place the correct emphases on new and recycled Fe in different ocean regions. Many 701 models rely on fixed rates of Fe regenerated by zooplankton and the remineralisation of 702 organic material, while others allow this to be vary (Table 1). A key parameter in driving the 703 turnover of Fe by the zooplankton and bacterial communities in such models is an estimate of 704 the heterotroph demand for Fe, which is then balanced against the Fe/C provided as nutrition. 705 New measurements of stocks and turnover of Fe from specific ocean regions are also 706 beginning to emerge [Boyd et al., 2015], which will be invaluable in assessing the magnitude 707 and variability of the modelled rates. 708

709 **5. Future Work**

710

A weakness of the current intercomparison is that we did not truly intercompare the Fe

- 712 models, but instead compared the models' coupled physical-biogeochemical framework
- 713 (including Fe). This was necessary to retain as broad a suite of models as possible for this first
- intercomparison. In future work, it would be useful to intercompare different Fe models

- vithin the same physical model framework (e.g. as possible in the NEMO or MITgcm
- 716 modelling frameworks). Additionally, a set of planned model perturbations could be
- performed where each individual model is subjected to a modification to its Fe supply (either
- as a direct fertilisation event or by an alteration to one of the input fields). Much could be
- real from the way the Fe cycle responds to such perturbations across the different models.
- 720

721 Reducing uncertainty in the input fluxes of Fe is clearly important, but has proved difficult to 722 achieve over recent years (even for long standing Fe sources such as dust). Some progress 723 could be made by implementing 'source specific' tracers (such as aluminium or manganese) 724 alongside Fe to constrain individual sources. Constraining scavenging rates has emerged as a 725 key priority and parallel simulation of Th may help constrain rates of Fe loss and the particle 726 pools. Moreover, many of the models used specifically for ecological questions are only run for 727 a few decades at most. This makes this subset of models more sensitive to their initial 728 conditions. A priority for such 'resource intensive' models would be the availability of input 729 fields based on data climatologies (such as those available for macronutrients as part of the 730 World Ocean Atlas datasets) or consensus distributions that may emerge from improved models.

731 732

As described in Sec. 4.3 an assessment of the different biological Fe models is also a priority,

as this will underpin the carbon cycle response and has not been compared against the

paradigms recently emerging from experimental work. In a follow up Phase of FeMIP we

could more closely compare the models against the detailed process study measurements

made (for example) as part of the FeCycle set of experiments [*Boyd et al.*, 2012; *Boyd et al.*,
2005]. A range of the Fe models could be set up in a one dimensional lagrangian framework
and forced by observed physics to be compared rigorously against the measured Fe stocks

- 740 and cycling rates.
- 741

742 6. Conclusions

743

744 We have compared the projected DFe distributions from thirteen global ocean 745 biogeochemistry models against each other and with available datasets. Newly available full 746 depth sections of DFe for different oceanic regions as part of the GEOTRACES programme 747 have greatly facilitated this task. All models do relatively poorly in reproducing a global DFe dataset of around 20,000 observations, which highlights the need for greater understanding 748 749 of how the ocean Fe cycle functions and how Fe should be represented in global ocean 750 models. We find a large degree of inter-model variability in the input fluxes of DFe, which 751 leads to great variability in the modeled residence times. The stronger inter-model agreement 752 in the mean ocean DFe most likely reflects earlier views of constant deep ocean DFe levels 753 maintained by a homogenous ligand pool and requires calibration via poorly constrained 754 scavenging rates. The way different models treat DFe scavenging has emerged as a key 755 uncertainty that would benefit from stronger observational constraints. More detailed inter-756 model tests, particularly linked to process study data, are needed to assess the models' 757 biological components.

758

In closing, we re-emphasise the importance of the iron cycle in global ocean biogeochemistry
models, given its role, alongside NO₃, as one of the two most important limiting nutrients.
Although the models analysed here struggle to capture the detailed distribution of this highly
dynamic element, it is very likely that biogeochemical models that include an iron cycle can
produce a more realistic simulation than models that do not. Improving the quantitative
understanding of iron cycling should be a major priority for ocean biogeochemistry research.

766

767 **7. Acknowledgements**

- 768
- We thank everyone that has been involved in the development of the FeMIP ocean models
- over many years. A.T especially thanks Eric Achterberg, Andrew Bowie, Maarten Klunder, Joe
- 771 Resing, Micha Rijkenberg, Mak Saito, Christian Schlosser and Peter Sedwick for sharing DFe
- 772 datasets ahead of their publication and Edward Mawji, Reiner Schlitzer, Elena Masferrer-
- 773 Dodas and the wider GEOTRACES community for their efforts in producing the Intermediate
- Data Product (2014) that facilitated this inter comparison. PISCES1 and PISCES2 simulations
- 775 made use of the N8 HPC facilities, funded by the N8 consortium and EPSRC grant
- EP/K000225/1. MV acknowledges the BFM system team (http://bfm-community.eu) for the
- public availability of the BFM model. We thank Bob Anderson and Ric Williams for comments
 on the draft manuscript and those of two anonymous reviewers that improved the final
- 779 version

780781 8. References

- 782
- Anderson, R. F. (2003), Chemical Tracers of Particle Transport, *Treatise on Geochemistry*, 247273. doi:10.1016/b0-08-043751-6/06111-9
- Archer, D. E., and K. Johnson (2000), A model of the iron cycle in the ocean, *Global Biogeochemical Cycles*, *14*(1), 269-279. doi:10.1029/1999gb900053
- Aumont, O., E. Maier-Reimer, S. Blain, and P. Monfray (2003), An ecosystem model of the
 global ocean including Fe, Si, P colimitations, *Global Biogeochemical Cycles*, *17*(2).
- 789 doi:10.1029/2001gb001745
- Aumont, O., C. Ethé, A. Tagliabue, L. Bopp, and M. Gehlen (2015), PISCES-v2: an ocean
- biogeochemical model for carbon and ecosystem studies, *Geoscientific Model Development*,
 8(8), 2465-2513. doi:10.5194/gmd-8-2465-2015
- Barbeau, K., J. W. Moffett, D. A. Caron, P. L. Croot, and D. L. Erdner (1996), Role of protozoan
 grazing in relieving iron limitation of phytoplankton, *Nature*, *380*(6569), 61-64.
 doi:10.1038/380061a0
- Bopp, L., et al. (2013), Multiple stressors of ocean ecosystems in the 21st century: projections
 with CMIP5 models, *Biogeosciences*, *10*(10), 6225-6245. doi:10.5194/bg-10-6225-2013
- Bowie, A. R., D. Lannuzel, T. A. Remenyi, T. Wagener, P. J. Lam, P. W. Boyd, C. Guieu, A. T.
- 799 Townsend, and T. W. Trull (2009), Biogeochemical iron budgets of the Southern Ocean south
- of Australia: Decoupling of iron and nutrient cycles in the subantarctic zone by the
 summertime supply, *Global Biogeochemical Cycles*, 23(4). doi:10.1029/2009gb003500
- Bowie, A. R., et al. (2015), Iron budgets for three distinct biogeochemical sites around the
 Kerguelen Archipelago (Southern Ocean) during the natural fertilisation study, KEOPS-2,
- 804 *Biogeosciences*, 12(14), 4421-4445. doi:10.5194/bg-12-4421-2015
- Boyd, P. W., and M. J. Ellwood (2010), The biogeochemical cycle of iron in the ocean, *Nature Geoscience*, *3*(10), 675-682. doi:10.1038/ngeo964
- 807 Boyd, P. W., R. F. Strzepek, M. J. Ellwood, D. A. Hutchins, S. D. Nodder, B. S. Twining, and S. W.
- 808 Wilhelm (2015), Why are biotic iron pools uniform across high- and low-iron pelagic
- 809 ecosystems?, *Global Biogeochemical Cycles*, 29(7), 1028-1043. doi:10.1002/2014gb005014

- 810 Boyd, P. W., et al. (2012), Microbial control of diatom bloom dynamics in the open ocean,
- 811 *Geophysical Research Letters*, 39(18). doi:10.1029/2012gl053448
- 812 Boyd, P. W., et al. (2005), FeCycle: Attempting an iron biogeochemical budget from a
- 813 mesoscale SF6 tracer experiment in unperturbed low iron waters, *Global Biogeochemical*814 *Cycles*, 19(4). doi:10.1029/2005gb002494
- Boye, M., J. Nishioka, P. Croot, P. Laan, K. R. Timmermans, V. H. Strass, S. Takeda, and H. J. W.
- de Baar (2010), Significant portion of dissolved organic Fe complexes in fact is Fe colloids, *Marine Chemistry*, *122*(1-4), 20-27. doi:10.1016/j.marchem.2010.09.001
- Bruland, K. W., R. Middag, and M. C. Lohan (2014), Controls of Trace Metals in Seawater, *Treatise on Geochemistry, 2nd edition*, 19-51. doi:10.1016/b978-0-08-095975-7.00602-1
- Burd, A. B., S. B. Moran, and G. A. Jackson (2000), A coupled adsorption–aggregation model of
 the POC/ ratio of marine particles, *Deep Sea Research Part I: Oceanographic Research Papers*,
 47(1), 103-120. doi:10.1016/s0967-0637(99)00047-3
- 823 Chever, F., E. Bucciarelli, G. Sarthou, S. Speich, M. Arhan, P. Penven, and A. Tagliabue (2010),
- 824 Physical speciation of iron in the Atlantic sector of the Southern Ocean along a transect from

the subtropical domain to the Weddell Sea Gyre, *J Geophys Res-Oceans*, *115*(C10).

826 doi:10.1029/2009jc005880

- Conway, T. M., and S. G. John (2014), Quantification of dissolved iron sources to the North
 Atlantic Ocean, *Nature*, *511*(7508), 212-215. doi:10.1038/nature13482
- Dunne, J. P., et al. (2013), GFDL's ESM2 Global Coupled Climate–Carbon Earth System Models.
- Part II: Carbon System Formulation and Baseline Simulation Characteristics*, *Journal of Climate*, *26*(7), 2247-2267. doi:10.1175/jcli-d-12-00150.1
- B32 Dutkiewicz, S., J. R. Scott, and M. J. Follows (2013), Winners and losers: Ecological and
 B33 biogeochemical changes in a warming ocean, *Global Biogeochemical Cycles*, *27*(2), 463-477.
- 834 doi:10.1002/gbc.20042
- B35 Dutkiewicz, S., B. A. Ward, F. Monteiro, and M. J. Follows (2012), Interconnection of nitrogen
- fixers and iron in the Pacific Ocean: Theory and numerical simulations, *Global Biogeochemical Cycles*, *26*(1). doi:10.1029/2011gb004039
- Dutkiewicz, S., B. A. Ward, J. R. Scott, and M. J. Follows (2014), Understanding predicted shifts
- in diazotroph biogeography using resource competition theory, *Biogeosciences*, *11*(19), 54455461. doi:10.5194/bg-11-5445-2014
- 841 Dutkiewicz, S., A. E. Hickman, O. Jahn, W. W. Gregg, C. B. Mouw, and M. J. Follows (2015),
- 842 Capturing optically important constituents and properties in a marine biogeochemical and
- 843 ecosystem model, *Biogeosciences*, *12*(14), 4447-4481. doi:10.5194/bg-12-4447-2015
- Elrod, V. A., W. M. Berelson, K. H. Coale, and K. S. Johnson (2004), The flux of iron from
 continental shelf sediments: A missing source for global budgets, *Geophysical Research Letters*,
- 846 *31*(12). doi:10.1029/2004gl020216
- Fitzsimmons, J. N., and E. A. Boyle (2014), Both soluble and colloidal iron phases control
 dissolved iron variability in the tropical North Atlantic Ocean, *Geochimica et Cosmochimica*Acta 125 529 550 doi:10.1016/j.gca.2013.10.022
- 849 *Acta*, *125*, 539-550. doi:10.1016/j.gca.2013.10.032

- 850 Frew, R. D., D. A. Hutchins, S. Nodder, S. Sanudo-Wilhelmy, A. Tovar-Sanchez, K. Leblanc, C. E.
- Hare, and P. W. Boyd (2006), Particulate iron dynamics during FeCycle in subantarctic waters
- southeast of New Zealand, *Global Biogeochemical Cycles*, *20*(1). doi:10.1029/2005gb002558
- Galbraith, E. D., A. Gnanadesikan, J. P. Dunne, and M. R. Hiscock (2010), Regional impacts of iron-light colimitation in a global biogeochemical model, *Biogeosciences*, 7(3), 1043-1064.
- 855 doi:10.5194/bg-7-1043-2010
- Gerringa, L. J. A., A. C. Alderkamp, P. Laan, C. E. Thuroczy, H. J. W. De Baar, M. M. Mills, G. L. van
- Dijken, H. van Haren, and K. R. Arrigo (2012), Iron from melting glaciers fuels the
- phytoplankton blooms in Amundsen Sea (Southern Ocean): Iron biogeochemistry, *Deep-Sea Res Pt Ii*, 71-76, 16-31. doi:Doi 10.1016/J.Dsr2.2012.03.007
- Gledhill, M., and K. N. Buck (2012), The organic complexation of iron in the marine
 environment: a review, *Frontiers in microbiology*, *3*, 69. doi:10.3389/fmicb.2012.00069
- Hatta, M., C. I. Measures, J. Wu, S. Roshan, J. N. Fitzsimmons, P. Sedwick, and P. Morton (2014),
- An overview of dissolved Fe and Mn Distributions during the 2010–2011 U.S. GEOTRACES
- 864 north Atlantic Cruises: GEOTRACES GA03, Deep Sea Research Part II: Topical Studies in
- 865 *Oceanography*. doi:10.1016/j.dsr2.2014.07.005
- Hauck, J., C. Völker, T. Wang, M. Hoppema, M. Losch, and D. A. Wolf-Gladrow (2013),
- Seasonally different carbon flux changes in the Southern Ocean in response to the southern
 annular mode, *Global Biogeochemical Cycles*, *27*(4), 1236-1245. doi:10.1002/2013gb004600
- 869 Hayes, C. T., et al. (2015), Intensity of Th and Pa scavenging partitioned by particle chemistry
- in the North Atlantic Ocean, *Marine Chemistry*, *170*, 49-60.
- 871 doi:10.1016/j.marchem.2015.01.006
- Homoky, W. B., S. G. John, T. M. Conway, and R. A. Mills (2013), Distinct iron isotopic
- signatures and supply from marine sediment dissolution, *Nature communications*, *4*, 2143.
 doi:10.1038/ncomms3143
- Honeyman, B. D., and P. H. Santschi (1989), A Brownian-pumping model for oceanic trace
 metal scavenging: Evidence from Th isotopes, *Journal of Marine Research*, 47(4), 951-992.
 doi:10.1357/002224089785076091
- Honeyman, B. D., L. S. Balistrieri, and J. W. Murray (1988), Oceanic trace metal scavenging: the
 importance of particle concentration, *Deep Sea Research Part A. Oceanographic Research Papers*, 35(2), 227-246. doi:10.1016/0198-0149(88)90038-6
- Hutchins, D. A., and K. W. Bruland (1994), Grazer-Mediated Regeneration and Assimilation of
 Fe, Zn and Mn from Planktonic Prey, *Marine Ecology Progress Series*, *110*(2-3), 259-269.
 doi:Doi 10.3354/Meps110259
- Jickells, T. D., et al. (2005), Global iron connections between desert dust, ocean
 biogeochemistry, and climate, *Science*, *308*(5718), 67-71. doi:10.1126/Science.1105959
- Johnson, K. S., R. M. Gordon, and K. H. Coale (1997), What controls dissolved iron
- concentrations in the world ocean?, *Marine Chemistry*, 57(3-4), 137-161. doi:10.1016/s03044203(97)00043-1

- Klunder, M. B., P. Laan, R. Middag, H. J. W. De Baar, and J. C. van Ooijen (2011), Dissolved iron
- in the Southern Ocean (Atlantic sector), Deep Sea Research Part II: Topical Studies in
- 891 *Oceanography*, *58*(25-26), 2678-2694. doi:10.1016/j.dsr2.2010.10.042
- Kohfeld, K. E., and A. Ridgwell (2009), Glacial-Interglacial Variability in Atmospheric CO2, *Surface Ocean--Lower Atmosphere Processes*, *187*, 251-286. doi:10.1029/2008gm000845
- Lam, P. J., and O. Marchal (2015), Insights into particle cycling from thorium and particle data, *Annual review of marine science*, *7*, 159-184. doi:10.1146/annurev-marine-010814-015623
- Lefèvre, N., and A. J. Watson (1999), Modeling the geochemical cycle of iron in the oceans and
 its impact on atmospheric CO2concentrations, *Global Biogeochemical Cycles*, *13*(3), 727-736.
 doi:10.1029/1999gb900034
- Liu, X., and F. J. Millero (1999), The solubility of iron hydroxide in sodium chloride solutions, *Geochimica et Cosmochimica Acta*, 63(19-20), 3487-3497. doi:10.1016/s0016-
- 901 7037(99)00270-7
- Marchal, O., and P. J. Lam (2012), What can paired measurements of Th isotope activity and
 particle concentration tell us about particle cycling in the ocean?, *Geochimica et Cosmochimica Acta*, 90, 126-148. doi:10.1016/j.gca.2012.05.009
- 905 Marchetti, A., M. S. Parker, L. P. Moccia, E. O. Lin, A. L. Arrieta, F. Ribalet, M. E. Murphy, M. T.
- Maldonado, and E. V. Armbrust (2009), Ferritin is used for iron storage in bloom-forming
 marine pennate diatoms, *Nature*, 457(7228), 467-470. doi:10.1038/nature07539
- 908 Matsumoto, K., K. Tokos, A. Huston, and H. Joy-Warren (2013), MESMO 2: a mechanistic
- 909 marine silica cycle and coupling to a simple terrestrial scheme, *Geoscientific Model*
- 910 Development, 6(2), 477-494. doi:10.5194/gmd-6-477-2013
- Mawji, E., R. Schlitzer, E. Masferrer-Dodas, and GEOTRACES-group (2015), The GEOTRACES
 Intermediate Data Product 2014, *Marine Chemistry*. doi:10.1016/j.marchem.2015.04.005
- 913 Misumi, K., K. Lindsay, J. K. Moore, S. C. Doney, D. Tsumune, and Y. Yoshida (2013), Humic
- 914 substances may control dissolved iron distributions in the global ocean: Implications from
- numerical simulations, *Global Biogeochemical Cycles*, *27*(2), 450-462. doi:10.1002/gbc.20039
- Moore, C. M., et al. (2013), Processes and patterns of oceanic nutrient limitation, *Nature Geoscience*. doi:10.1038/ngeo1765
- Moore, J. K., and O. Braucher (2008), Sedimentary and mineral dust sources of dissolved iron
 to the world ocean, *Biogeosciences*, 5(3), 631-656. doi:10.5194/bg-5-631-2008
- Moore, J. K., S. C. Doney, D. M. Glover, and I. Y. Fung (2002), Iron cycling and nutrient-
- 921 limitation patterns in surface waters of the World Ocean, *Deep-Sea Res Pt Ii*, 49(1-3), 463-507.
 922 doi:10.1016/S0967-0645(01)00109-6
- 923 Moore, J. K., K. Lindsay, S. C. Doney, M. C. Long, and K. Misumi (2013), Marine Ecosystem
- 924 Dynamics and Biogeochemical Cycling in the Community Earth System Model [CESM1(BGC)]:
- 925 Comparison of the 1990s with the 2090s under the RCP4.5 and RCP8.5 Scenarios, *Journal of*
- 926 *Climate*, *26*(23), 9291-9312. doi:10.1175/jcli-d-12-00566.1

- 927 Noble, A. E., et al. (2012), Basin-scale inputs of cobalt, iron, and manganese from the
- 928 Benguela-Angola front to the South Atlantic Ocean, *Limnology and Oceanography*, 57(4), 989-
- 929 1010. doi:10.4319/lo.2012.57.4.0989
- Parekh, P., M. J. Follows, and E. Boyle (2004), Modeling the global ocean iron cycle, *Global Biogeochemical Cycles*, *18*(1). doi:10.1029/2003gb002061
- Raiswell, R., L. G. Benning, M. Tranter, and S. Tulaczyk (2008), Bioavailable iron in the
- Southern Ocean: the significance of the iceberg conveyor belt, *Geochemical transactions*, *9*, 7.
 doi:10.1186/1467-4866-9-7
- Raven, J. A. (1988), The iron and molybdenum use efficiencies of plant growth with different
 energy, carbon and nitrogen sources, *New Phytologist*, *109*(3), 279-287. doi:10.1111/j.14698137.1988.tb04196.x
- Raven, J. A., M. C. W. Evans, and R. E. Korb (1999), The role of trace metals in photosynthetic
 electron transport in O2-evolving organisms, *Photosynthesis research*, 60(2/3), 111-150.
 doi:10.1023/a:1006282714942
- 941 Resing, J. A., P. N. Sedwick, C. R. German, W. Jenkins, J. W. Moffett, B. Sohst, and A. Tagliabue
- 942 (2015), Basin-scale transport of hydrothermal dissolved metals across the South Pacific
- 943 Ocean, *Nature*. doi:10.1038/nature14577
- Ridgwell, A. J. (2001), Glacial-interglacial perturbations in the global carbon cycle, Universityof East Anglia.
- Rijkenberg, M. J., R. Middag, P. Laan, L. J. Gerringa, H. M. van Aken, V. Schoemann, J. T. de Jong,
- and H. J. de Baar (2014), The distribution of dissolved iron in the west atlantic ocean, *Plos One*,
 9(6), e101323. doi:10.1371/journal.pone.0101323
- Saito, M. A., A. E. Noble, A. Tagliabue, T. J. Goepfert, C. H. Lamborg, and W. J. Jenkins (2013),
 Slow-spreading submarine ridges in the South Atlantic as a significant oceanic iron source, *Nature Geoscience*, 6(9), 775-779. doi:10.1038/Ngeo1893
- 952 Sarthou, G., D. Vincent, U. Christaki, I. Obernosterer, K. R. Timmermans, and C. P. D. Brussaard
- 953 (2008), The fate of biogenic iron during a phytoplankton bloom induced by natural
- 954 fertilisation: Impact of copepod grazing, *Deep Sea Research Part II: Topical Studies in*
- 955 *Oceanography*, *55*(5-7), 734-751. doi:10.1016/j.dsr2.2007.12.033
- 956 Savoye, N., C. Benitez-Nelson, A. B. Burd, J. K. Cochran, M. Charette, K. O. Buesseler, G. A.
- Jackson, M. Roy-Barman, S. Schmidt, and M. Elskens (2006), 234Th sorption and export
- models in the water column: A review, *Marine Chemistry*, *100*(3-4), 234-249.
- 959 doi:10.1016/j.marchem.2005.10.014
- Stock, C. A., J. P. Dunne, and J. G. John (2014), Global-scale carbon and energy flows through
 the marine planktonic food web: An analysis with a coupled physical-biological model, *Progress in Oceanography*, *120*, 1-28. doi:10.1016/j.pocean.2013.07.001
- 963 Strzepek, R. F., K. A. Hunter, R. D. Frew, P. J. Harrison, and P. W. Boyd (2012), Iron-light
- 964 interactions differ in Southern Ocean phytoplankton, *Limnology and Oceanography*, 57(4),
 965 1182 1200 doi:10.4210/lo.2012.57.4.1182
- 965 1182-1200. doi:10.4319/lo.2012.57.4.1182

- 966 Strzepek, R. F., M. T. Maldonado, J. L. Higgins, J. Hall, K. Safi, S. W. Wilhelm, and P. W. Boyd
- 967 (2005), Spinning the "Ferrous Wheel": The importance of the microbial community in an iron
- 968 budget during the FeCycle experiment, *Global Biogeochemical Cycles*, *19*(4), GB4S26.
- 969 doi:10.1029/2005gb002490
- Sunda, W. G., and S. A. Huntsman (1997), Interrelated influence of iron, light and cell size on
 marine phytoplankton growth, *Nature*, *390*(6658), 389-392. doi:10.1038/37093
- 972 Sunda, W. G., and S. A. Huntsman (1998), Processes regulating cellular metal accumulation
- and physiological effects: Phytoplankton as model systems, *Science of The Total Environment*,
 219(2-3), 165-181. doi:10.1016/s0048-9697(98)00226-5
- Tagliabue, A. (2014), More to hydrothermal iron input than meets the eye, *Proceedings of the National Academy of Sciences of the United States of America*, *111*(47), 16641-16642.
- 977 doi:10.1073/pnas.1419829111
- Tagliabue, A., and C. Völker (2011), Towards accounting for dissolved iron speciation in global
 ocean models, *Biogeosciences*, 8(10), 3025-3039. doi:10.5194/bg-8-3025-2011
- Tagliabue, A., L. Bopp, and O. Aumont (2008), Ocean biogeochemistry exhibits contrasting
 responses to a large scale reduction in dust deposition, *Biogeosciences*, 5(1), 11-24.
- Tagliabue, A., O. Aumont, and L. Bopp (2014a), The impact of different external sources of iron
 on the global carbon cycle, *Geophysical Research Letters*, 41(3), 920-926.
 doi:10.1002/2012gl050050
- 984 doi:10.1002/2013gl059059
- 785 Tagliabue, A., R. G. Williams, N. Rogan, E. P. Achterberg, and P. W. Boyd (2014b), A ventilation-
- based framework to explain the regeneration-scavenging balance of iron in the ocean, *Geophysical Research Letters*, 41(20), 7227-7236. doi:10.1002/2014gl061066
- Tagliabue, A., J.-B. Sallée, A. R. Bowie, M. Lévy, S. Swart, and P. W. Boyd (2014c), Surface-water
 iron supplies in the Southern Ocean sustained by deep winter mixing, *Nature Geoscience*, 7(4),
 314-320. doi:10.1038/ngeo2101
- 791 Tagliabue, A., T. Mtshali, O. Aumont, A. R. Bowie, M. B. Klunder, A. N. Roychoudhury, and S.
- Swart (2012), A global compilation of dissolved iron measurements: focus on distributions
 and processes in the Southern Ocean, *Biogeosciences*, 9(6), 2333-2349. doi:10.5194/bg-92333-2012
- Tagliabue, A., L. Bopp, D. M. Roche, N. Bouttes, J. C. Dutay, R. Alkama, M. Kageyama, E. Michel,
 and D. Paillard (2009), Quantifying the roles of ocean circulation and biogeochemistry in
 governing ocean carbon-13 and atmospheric carbon dioxide at the last glacial maximum, *Climate of the Past*, 5(4), 695-706. doi:10.5194/cp-5-695-2009
- 799 Tagliabue, A., et al. (2010), Hydrothermal contribution to the oceanic dissolved iron
 inventory, *Nature Geoscience*, *3*(4), 252-256. doi:10.1038/ngeo818
- Twining, B. S., and S. B. Baines (2013), The trace metal composition of marine phytoplankton,
 Annual review of marine science, *5*, 191-215. doi:10.1146/annurev-marine-121211-172322
- Twining, B. S., S. D. Nodder, A. L. King, D. A. Hutchins, G. R. LeCleir, J. M. DeBruyn, E. W. Maas, S.
 Vogt, S. W. Wilhelm, and P. W. Boyd (2014), Differential remineralization of major and trace

- 1005 elements in sinking diatoms, *Limnol. Oceanogr*, *59*(3), 689-704.
- 1006 doi:10.4319/lo.2014.59.3.0689
- 1007 Vichi, M., N. Pinardi, and S. Masina (2007), A generalized model of pelagic biogeochemistry for
- the global ocean ecosystem. Part I: Theory, *Journal of Marine Systems*, 64(1-4), 89-109.
- 1009 doi:10.1016/j.jmarsys.2006.03.006
- 1010 Völker, C., and A. Tagliabue (2015), Modeling organic iron-binding ligands in a three-
- 1011 dimensional biogeochemical ocean model, *Marine Chemistry*, 173, 67-77.
- 1012 doi:10.1016/j.marchem.2014.11.008
- Wagener, T., E. Pulido-Villena, and C. Guieu (2008), Dust iron dissolution in seawater: Results
 from a one-year time-series in the Mediterranean Sea, *Geophysical Research Letters*, *35*(16).
- 1015 doi:10.1029/2008gl034581
- 1016 Ward, B. A., S. Dutkiewicz, C. M. Moore, and M. J. Follows (2013), Iron, phosphorus, and 1017 nitrogen supply ratios define the biogeography of nitrogen fixation. *Limnology and*
- 1018 Oceanography, 58(6), 2059-2075. doi:10.4319/lo.2013.58.6.2059
- Watson, A. J., D. C. E. Bakker, A. J. Ridgwell, P. W. Boyd, and C. S. Law (2000), Effect of iron
 supply on Southern Ocean CO2 uptake and implications for glacial atmospheric CO2, *Nature*,
 407(6805), 730-733. doi:10.1038/35037561
- Wells, M. L., N. M. Price, and K. W. Bruland (1995), Iron chemistry in seawater and its
 relationship to phytoplankton: a workshop report, *Marine Chemistry*, *48*(2), 157-182.
 doi:10.1016/0304-4203(94)00055-i
- Wu, J., E. Boyle, W. Sunda, and L. S. Wen (2001), Soluble and colloidal iron in the oligotrophic
 North Atlantic and North Pacific, *Science*, *293*(5531), 847-849. doi:10.1126/science.1059251
- Ye, Y., T. Wagener, C. Völker, C. Guieu, and D. A. Wolf-Gladrow (2011), Dust deposition: iron
 source or sink? A case study, *Biogeosciences*, 8(8), 2107-2124. doi:10.5194/bg-8-2107-2011
- Yool, A., E. E. Popova, and T. R. Anderson (2011), Medusa-1.0: a new intermediate complexity
 plankton ecosystem model for the global domain, *Geoscientific Model Development*, 4(2), 381417. doi:10.5194/gmd-4-381-2011
- Yool, A., E. E. Popova, and T. R. Anderson (2013), MEDUSA-2.0: an intermediate complexity
 biogeochemical model of the marine carbon cycle for climate change and ocean acidification
 studies, *Geoscientific Model Development*, 6(5), 1767-1811. doi:10.5194/gmd-6-1767-2013
- 1035
- 1036 Figure Legends
- 1037
- Figure 1. Histograms of the average DFe concentration (nM) simulated by the FeMIP models
 across four different depth bins for three regions. The Northern Hemisphere is 30°N-90°N,
 Tropics are 30°S-30°N and the Southern Hemisphere is 30°S-90°S.
- 1041
- **Figure 2.** Annual mean DFe concentrations (nM) averaged over the upper 50m from the
- 1043 FeMIP models. Data averaged over the period January to June and July to December is taken
- 1044 from the expanded *Tagliabue et al.* [2012] dataset and has been averaged over 5° bins in 1045 latitude and longitude to improve visibility.
- 1046

- 1047 **Figure 3.** Annual maximum minus annual minimum DFe concentrations (nM) averaged over the upper 50m from the FeMIP models. 1048 1049 1050 **Figure 4.** DFe concentrations (nM) from the GA-02 [*Rijkenberg et al.*, 2014] cruise and 1051 extracted from the FeMIP models. 1052 1053 Figure 5. DFe concentrations (nM) from the GA-03 cruise [Hatta et al., 2014] and extracted 1054 from the FeMIP models. 1055 1056 Figure 6. DFe concentrations (nM) from the CoFeMUG cruise [Noble et al., 2012] and extracted from the FeMIP models. 1057 1058 1059 Figure 7. DFe concentrations (nM) from the GP-16 cruise [Resing et al., 2015] and extracted from the FeMIP models. 1060 1061 1062 Figure 8. DFe concentrations (nM) from the GIPY-4 and 5 cruises [Chever et al., 2010; Klunder 1063 et al., 2011] and extracted from the FeMIP models 1064 1065 **Figure 9.** NO₃ concentrations (µM) from the GA-02 cruise [*Rijkenberg et al.*, 2014] and extracted from the FeMIP models (NO₃ data not provided for GENIE). 1066 1067 1068 **Figure 10.** PO₄ concentrations (µM) from the GA-02 cruise [*Rijkenberg et al.*, 2014] and extracted from the FeMIP models (PO₄ not provided for MEDUSA-1, MEDUSA-2, RECOM and 1069 1070 TOPAZ). 1071 1072 **Figure 11.** Fe* (Fe – NO₃*r_{Fe/N}, nM) from the GA-02 cruise cruise [*Rijkenberg et al.*, 2014] and 1073 extracted from the FeMIP models. For models that do not provide NO3, PO4 is used and
- 1074 converted to NO3 assuming a ratio of 16:1.