

# 1 **Importance of multiple sources of iron for the upper ocean** 2 **biogeochemistry over the northern Indian Ocean**

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6

## 7 **Abstract**

8 Although the northern Indian Ocean (IO) is globally one of the most productive regions and receives dissolved  
9 iron (DFe) from multiple sources, there is no comprehensive understanding of how these different sources of DFe  
10 can impact upper ocean biogeochemical dynamics. Using an Earth system model with an ocean biogeochemistry  
11 component this study shows that atmospheric deposition is the most important source of DFe to the upper 100 m  
12 of the northern IO, contributing more than 50% of the annual DFe concentration. Sedimentary sources are locally  
13 important in the vicinity of the continental shelves and over the southern tropical IO, away from high atmospheric  
14 depositions. While atmospheric depositions contribute to more than 10% (35%) to 0-100 m (surface level)  
15 chlorophyll concentrations over large parts of the northern IO, sedimentary sources have similar contribution to  
16 chlorophyll concentrations over the southern tropical IO. Such increases in chlorophyll are primarily driven by an  
17 increase in diatom population over most of the northern IO. The regions that are susceptible to chlorophyll  
18 enhancement following external DFe additions are where low levels of background DFe and high background  
19 nitrate-to-iron values are observed. Analysis of DFe budget over selected biophysical regimes over the northern  
20 IO points to vertical mixing as the most important mechanism for DFe supply, while the importance of advection  
21 (horizontal and vertical) varies seasonally. Apart from removal of surface DFe by phytoplankton uptake,  
22 subsurface balance between DFe scavenging and regeneration is crucial in replenishing DFe pool to be made  
23 available to surface layer by physical processes.

## 24 **1 Introduction**

25 Iron is an essential micronutrient for primary producers in the ocean due to the catalytic role of iron in  
26 photosynthesis, respiration, and nitrogen fixation (Geider & La Roche, 1994; Raven, 1988). Although iron is one  
27 of the most abundant elements in the Earth's crust (McLennan, 2001), its low solubility (Sholkovitz et al., 2012)  
28 coupled with an intricate balance between complexation by ligands and high scavenging tendency does not make  
29 it readily bioavailable (Boyd & Ellwood, 2010). It has been estimated that iron availability limits primary  
30 productivity in as much as ~30% of the global oceans, which results in accumulation of unutilized macronutrients  
31 like nitrate and phosphate (Moore et al., 2013a). Even in regions experiencing nitrate limitation of productivity,  
32 nitrogen fixation is controlled by the supply of iron (e.g., Mills et al., 2004; Moore et al., 2009; Schlosser et al.,  
33 2014). Several artificial iron addition experiments performed in the open oceans have demonstrated its  
34 significance in regulating phytoplankton growth (Yoon et al., 2018), while natural iron fertilizations have also  
35 shown high levels of carbon export from the upper ocean following increased productivity (e.g., Blain et al., 2007;  
36 Pollard et al., 2009).

37 The main external sources of dissolved iron (DFe) to the world oceans are atmospheric depositions (e.g., Conway  
38 et al., 2014; Jickells et al., 2005), continental sediments (Elrod et al., 2004; Johnson et al., 1999), river inputs (e.g.,  
39 Buck et al., 2007; Canfield, 1997), sea ice (Sedwick & DiTullio, 1997; Wang et al., 2014) and iron seeping from  
40 hydrothermal vents (e.g., Nishioka et al., 2013; Tagliabue et al., 2010). Most ocean biogeochemistry models  
41 simulating the iron cycle estimate dust ( $1.4\text{--}32.7 \text{ Gmol yr}^{-1}$ ) or sedimentary sources ( $0.6\text{--}194 \text{ Gmol yr}^{-1}$ ) to have  
42 the highest contribution to ocean DFe inventory (Tagliabue et al., 2016). However, many of these models do not  
43 include hydrothermal sources of DFe. Numerical modelling using dust, sedimentary and hydrothermal sources of  
44 DFe have shown that while ocean column DFe inventory is most sensitive to sedimentary and hydrothermal DFe,  
45 atmospheric and sedimentary sources of DFe have the largest impact on atmospheric carbon dioxide (Tagliabue  
46 et al., 2014). This is because while atmospheric and sedimentary DFe can impact productivity over both the open  
47 and coastal oceans, iron from hydrothermal vents reaching the surface water depends on deepwater ventilation  
48 and stabilizing impact of organic ligands (Tagliabue et al., 2010; Sander and Koschinsky, 2011). However, with  
49 availability of more *in situ* DFe measurements, the relative importance of different sources of DFe are being re-  
50 examined at global as well as regional scales.

51 The northern Indian Ocean (IO) is one of the most productive regions of the global oceans, contributing high  
52 levels of organic carbon fluxes to the deeper ocean (e.g., Barber et al., 2001; Madhupratap et al., 2003; Rixen et  
53 al., 2019). The monsoonal winds drive phytoplankton blooms over different regions of the northern IO, arising  
54 from distinct physical mechanisms in different seasons. These mechanisms include blooms due to coastal and  
55 open ocean upwelling, advection of nutrients by ocean currents, and mixed layer deepening by winter convection.  
56 Episodic blooms are also triggered by passage of cyclones (Kuttippurath et al., 2021) and mesoscale eddies  
57 (Prasanna Kumar et al., 2004; Vidya & Prasanna Kumar, 2013). The region hosts one of the most intense oxygen  
58 minimum zones of the world oceans (Schmidtko et al., 2017) and is globally one of the major denitrification sites  
59 (e.g., Morrison et al., 1999; Bianchi et al., 2012). Several water column measurements have shown that the primary  
60 limiting nutrient over the northern IO is reactive nitrogen with possible colimitation by silicate (Košić et al., 2009;  
61 Moore et al., 2013a; Morrison et al., 1998). In recent years, a few studies using ocean biogeochemistry models  
62 have also pointed to possible iron limitation of phytoplankton blooms during southwest monsoon months (June-  
63 September), especially over upwelling regions of the western Arabian Sea (AS), which is the north-western part  
64 of the IO (Košić et al., 2009; Wiggert et al., 2007). These findings on the role of iron limitation have also been  
65 supported by incubation experiments over the AS during the late southwest monsoon, which have noted  
66 chlorophyll enhancements following iron enrichments (Moffett et al., 2015). Furthermore, *in situ* measurements  
67 during the late southwest monsoon have revealed complete drawdowns of silicate, owing to its high utilization  
68 under iron limitation, as well as high nitrate-to-iron ratios over the western AS (Naqvi et al., 2010). Nutrient  
69 enrichment experiments over the central AS during northeast monsoon months (December-March) have also  
70 revealed signatures of iron and nitrate colimitation, with addition of these two nutrients supporting increases in  
71 diatoms and coccolithophores (Takeda et al., 1995). Colimitation by nitrogen, phosphorus and iron has been  
72 identified over the southern Bay of Bengal (BoB, the north-eastern part of the IO) and the eastern equatorial IO  
73 (Twining et al., 2019). Thus, availability of iron can have major impacts on availability of other macronutrients  
74 and productivity, which can in turn impact denitrification and mid-depth oxygen levels in this region by  
75 modulating fluxes of sinking organic matters.

76 In general, there is a reduction in surface DFe concentrations over the northern IO from north to south. Systematic  
77 DFe measurements, encompassing all seasons over the AS, conducted during the Joint Global Ocean Flux Study  
78 (JGOFS) of the 1990s showed DFe concentrations often exceeding 1 nM, especially during the southwest  
79 monsoon (Measures & Vink, 1999). Subsequent measurements revealed lower levels of DFe with surface values  
80 ranging between 0.2-1.2 nM over the AS and between 0.2-0.5 nM over the BoB (Chinni et al., 2019; Chinni &  
81 Singh, 2022; Grand et al., 2015; Moffett et al., 2015; Vu & Sohrin, 2013). These values are generally higher than  
82 most of the open ocean regions. In contrast, southwards of the equatorial IO have surface DFe values generally  
83 less than 0.2 nM (e.g., Chinni et al., 2019; Grand et al., 2015; Twining et al 2019; Vu & Sohrin, 2013). The oxygen  
84 minimum zone, located to the north of the equator between depths of 150-1000 m, has elevated levels of DFe (>1  
85 nM), possibly due to DFe transport from reducing shelf sediments and remineralization of sinking organic matter  
86 (Moffett et al., 2007).

87 The overall high values of DFe over the northern IO can stem from multiple external sources of DFe identified  
88 within this region: atmospheric aerosol inputs (dust and black carbon) from South and Southwest Asia (Banerjee  
89 et al., 2019; Srinivas et al., 2012), continental shelf sediments, high river discharge, especially, over the BoB (e.g.,  
90 Chinni et al., 2019; Grand et al., 2015) and hydrothermal vents from the Central Indian Ridge that mainly impact  
91 DFe levels at depths of around 3000 m (Nishioka et al., 2013). The importance of episodic dust depositions in  
92 alleviating iron limitations of primary productivity over the central AS has been identified, during the northeast  
93 monsoon when a deeper ferricline compared to the nitracline yields a high nitrate-to-iron ratio (Banerjee and  
94 Kumar, 2014). Additionally, modelling studies over the AS have demonstrated that DFe derived from dust  
95 deposition can support about half of the observed primary productivity and a large fraction of nitrogen fixation  
96 (Guieu et al., 2019). Centennial-scale model simulations over the IO have revealed that changes in phytoplankton  
97 community structure have resulted in increased (reduced) carbon uptake over the eastern (western) IO in response  
98 to increased anthropogenic DFe deposition in the present day compared to pre-industrial levels (Pham & Ito,  
99 2021). Yet another challenge is that, away from regions with high aerosol loading, other sources of DFe can  
100 become important in supporting ocean productivity and controlling patterns of nutrient limitations. Such  
101 understanding of relative roles of different sources of DFe in controlling the biogeochemical dynamics of the  
102 northern IO remains unexplored. This is important considering the multiple sources of DFe over the northern IO.  
103 To this end, the present study uses a suite of simulations from a state-of-the art Earth system model with an iron  
104 cycle in its ocean biogeochemistry component to explore the relative contribution of different sources of DFe to  
105 phytoplankton blooms and impacts on nutrient availability over the upper 100 m of the northern IO. Furthermore,  
106 DFe budget has been analysed over the upper ocean for varied biophysical regimes in this region to identify how  
107 different sources of DFe can impact the total DFe budget.

108

## 109 **2 Data and model**

110 The study uses satellite and reanalysis products, ocean observation data, and an Earth system model to assess  
111 contributions of different sources of DFe to phytoplankton blooms over the northern IO. For the present study,  
112 the northern IO is considered to encompass 30°N–20°S latitude, 40°–105°E longitude. Thus, the tropical part of  
113 the southern IO is also included. Only the open ocean regions, having bottom depth greater than 1000 m, are  
114 studied here. The four seasons referred to in this study are defined as: the northeast monsoon: December-March;

115 spring intermonsoon: April-May; southwest monsoon: June-September; and fall intermonsoon: October-  
116 November.

## 117 **2.1 Model**

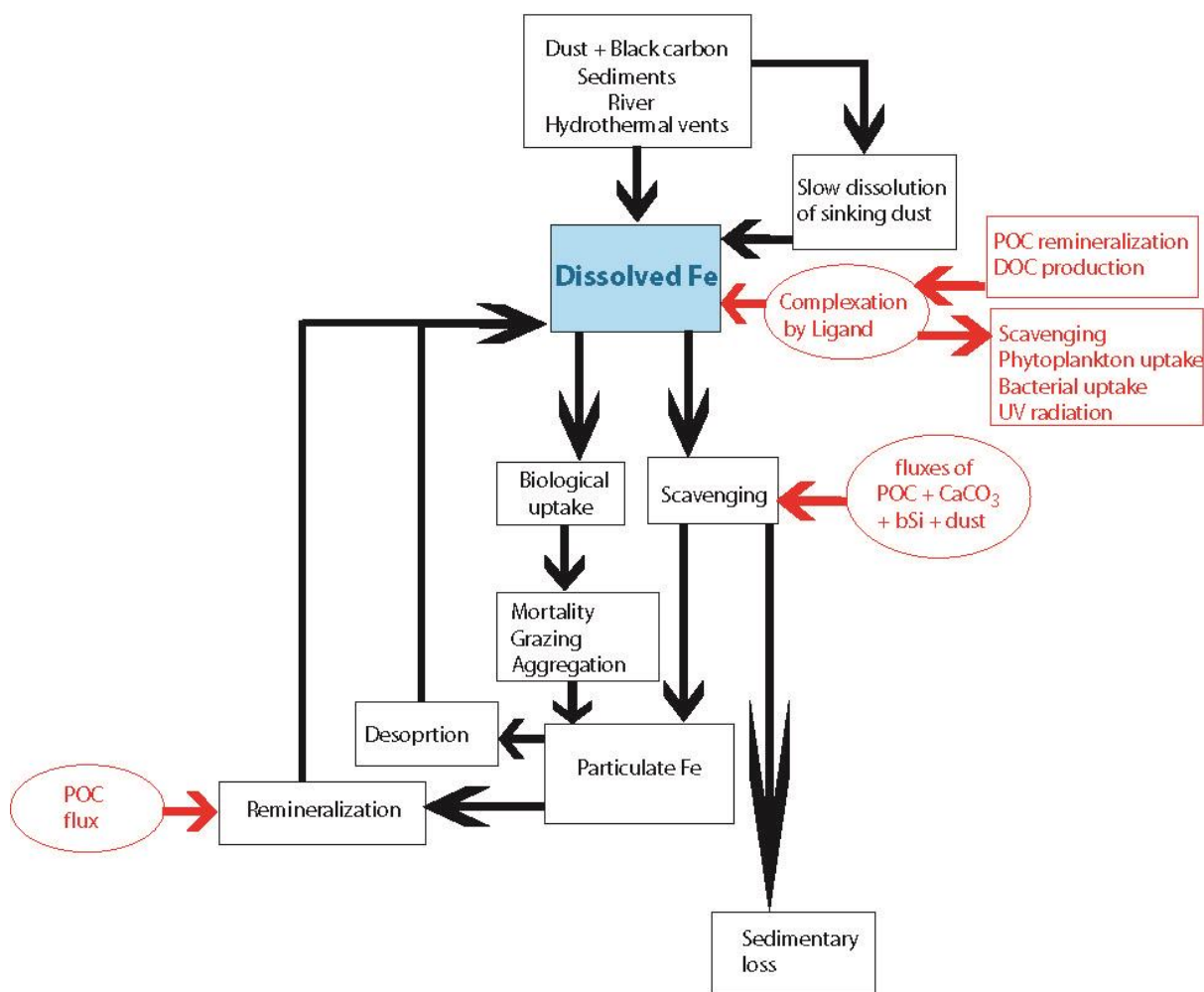
118 This study uses the ocean component Parallel Ocean Program version 2 (POP2) (Smith et al., 2010) embedded in  
119 the Community Earth System Model (CESM) version 2.1. This version of CESM incorporates several  
120 improvements over previous versions of the model (Danabasoglu et al., 2020). The POP2 model is a level-  
121 coordinate model having Arakawa B-grid in the horizontal with North Pole displaced over Greenland. The vertical  
122 resolution is 10 m for the upper 160 m and decreases with depth to 250 m in the bottom. The horizontal resolution  
123 is nominally 1° with meridional resolution increasing to 0.27° near the equator (Danabasoglu et al., 2012),  
124 implying that mesoscale eddies are not resolved. Momentum advection is based on a second-order central  
125 advection scheme while tracer advection relies on a third-order upwind advection scheme. Vertical ocean mixing  
126 is parameterized using the non-local K-Profile parameterization (Large et al., 1994), which is incorporated into  
127 CESM2.1 via the Community Ocean Vertical Mixing (CVMix) framework. Horizontal mixing is parameterized  
128 using the Gent and Williams (1990) scheme, which includes eddy-induced velocity in addition to diffusion of  
129 tracers along isopycnals. Macronutrients and oxygen are initialized from World Ocean Atlas 2013 version 2  
130 dataset (Garcia et al., 2014a, b) and alkalinity is initialized using GLObal Ocean Data Analysis Project  
131 (GLODAPv2; Olsen et al., 2016). Temperature and salinity are initialized from January-mean values from the  
132 Polar Science Center Hydrographic Climatology, which is based on data from Levitus et al. (1998). Ecosystem  
133 tracers, including iron, chlorophyll, dissolved organic and inorganic carbon are initialized from a previous CESM1  
134 simulation.

135 The biogeochemistry component of POP2 is implemented using Marine Biogeochemistry Library (MARBL),  
136 which is the most updated version of the previously implemented Biogeochemistry Elemental Cycle (BEC) model  
137 (Long et al., 2021). The model includes key limiting nutrients (N, P, Si, Fe), three types of explicit phytoplankton  
138 functional groups (diatoms, diazotrophs and nano/picophytoplankton), one implicit calcifier group, and one  
139 zooplankton type. The C:N ratio for nutrient assimilation is fixed at 117:16 (Anderson and Sarmiento, 1994),  
140 whereas P:C, Fe:C, Si:C and chlorophyll:C ratios are allowed to vary based on ambient nutrient concentrations.  
141 The Fe:C ratio is allowed to change within a fixed range based on phytoplankton growth terms, loss terms, and  
142 the iron uptake half-saturation constant for different phytoplankton groups (Moore et al., 2004). For each of the 3  
143 phytoplankton groups the minimum allowed Fe:C ratio is 2.5  $\mu\text{mol mol}^{-1}$ . The maximum allowed Fe:C ratio is 30  
144  $\mu\text{mol mol}^{-1}$  for diatoms and small phytoplankton, and 60  $\mu\text{mol mol}^{-1}$  for diazotrophs due to their higher demand  
145 for iron. The zooplankton Fe:C ratio is fixed at 3.0  $\mu\text{mol mol}^{-1}$ . Individual nutrient limitation for phytoplankton is  
146 assessed based on Michaelis-Menten nutrient uptake kinetics, which is a function of the specific nutrient  
147 concentration and nutrient uptake half-saturation coefficient. The half-saturation coefficient is nutrient-specific  
148 and phytoplankton-group specific. Nutrient limitation terms vary from 0 to 1, with 0 being the most limiting  
149 nutrient. Multiple nutrient limitation follows Liebig's law of minimum, so that the nutrient limitation term with  
150 minimum value limits phytoplankton growth rate (Long et al., 2021). Loss of phytoplankton in MARBL is  
151 accounted for by grazing, mortality, and aggregation of sinking flocculants.

152 The main DFe sources considered in MARBL are atmospheric depositions, shelf sediments, riverine inputs, and  
153 hydrothermal vents (Fig. S1). Globally, these sources of DFe account for 13.62 Gmol yr<sup>-1</sup>, 19.68 Gmol yr<sup>-1</sup>, 0.37  
154 Gmol yr<sup>-1</sup>, and 4.91 Gmol yr<sup>-1</sup>, respectively (Long et al., 2021). Atmospheric sources of DFe are from dust and  
155 black carbon depositions obtained from a fully coupled CESM2 simulation in hindcast mode at nominal 1° spatial  
156 resolution as a part of the Coupled Model Intercomparison Phase 6 (CMIP6) contribution. Dust emissions and  
157 transport/deposition are calculated, respectively, using the Community Land Model version 5 (CLM5) and  
158 Community Atmosphere model version 6 (CAM6) in Whole Atmosphere Community Climate Model (WACCM)  
159 configuration. The newly included Modal Aerosol Module version 4 (MAM4) in CAM6 includes dust in the  
160 accumulation and coarse modes. Black carbon is emitted in the primary mode and transferred to accumulation  
161 mode via aging (Liu et al., 2016). Monthly climatology of dust and black carbon for the year 2000 is used in  
162 repeating mode. About 3.5% of dust is assumed to be iron with the solubility of iron depending on the ratio  
163 between coarse and fine dust fluxes. This accounts for increasing iron solubility with increasing distance from  
164 dust source regions. A constant solubility of 6% is assigned to iron derived from black carbon aerosols. In addition  
165 to surface iron release, there is slow dissolution of sinking “hard” dust fraction (~98% of total dust) with depth  
166 such that ~0.3% of dust will dissolve over 4000 m (Armstrong et al., 2002; Moore et al., 2004). For the rest of the  
167 2% “soft” dust, remineralization takes place with a length-scale of 200 m. Sedimentary iron supply is based on  
168 sub-grid scale bathymetry that depends on two factors: firstly, for reducing sediments, it is proportional to  
169 particulate organic carbon fluxes in regions where these fluxes are larger than 3 g C m<sup>-2</sup> yr<sup>-1</sup>; secondly, in oxic  
170 sediments, it depends on constant low background fluxes and bottom current velocity, which accounts for  
171 sediment resuspension. As a result, the main sources of sedimentary DFe are along continental shelves and  
172 productive margins, with little contribution coming from the deep ocean. For the river source of DFe, discharge  
173 data for the year 2000 from Global Nutrient Export from WaterSheds (GlobalNEWS, Mayorga et al., 2010) is  
174 combined with constant DFe concentration of 10 nM. For hydrothermal vents, a constant flux of iron from the  
175 grid boxes containing vents is applied so that the total hydrothermal vent iron flux is equal to approximately 5.0  
176 Gmol yr<sup>-1</sup>.

177 Iron input to the ocean is balanced by losses from biological uptake and scavenging. The biological uptake of iron  
178 is based on the species-specific Fe:C ratio, which varies based on ambient DFe concentration, as discussed  
179 previously. The biological uptake term also includes routing of phytoplankton iron to zooplankton based on its  
180 feeding preference. Losses of iron from the biological pools are through mortality, aggregation, grazing upon  
181 phytoplankton by zooplankton, as well as higher trophic grazing on zooplankton (Long et al., 2021). The  
182 scavenging loss of DFe is expressed as a two-step process similar to the thorium scavenging model: involving the  
183 calculation of the net adsorption rate to sinking particles and modification of this rate by the ambient iron  
184 concentration (Moore and Braucher, 2008). The total sinking particles consist of particulate organic carbon,  
185 biogenic silica, calcium carbonate, and dust, which strongly influence DFe scavenging in excess of ligand  
186 concentrations. The particulate organic carbon is multiplied by 6 to account for the non-carbon portion of the  
187 organic matter that can take part in scavenging. In CESM, scavenging increases non-linearly with DFe  
188 concentration. About 90% of the scavenged iron enters the sinking particulate pool, while the rest is lost to  
189 sediments. Along with the scavenging contribution, iron released from grazing and mortality of autotrophs and  
190 zooplankton also enters the particulate iron pool. Remineralization of this sinking particulate iron replenishes DFe  
191 and is parameterized as a function of sinking particulate organic carbon flux. This results in maximum

192 remineralization taking place within the upper 100 m where particulate organic carbon flux is the highest.  
 193 Additionally, slow desorption of sinking particulate iron also releases DFe at depths and is calculated using a  
 194 constant desorption rate of  $1.0 \times 10^{-6} \text{ cm}^{-1}$  for particulate iron. The model also includes an explicit ligand tracer  
 195 for complexing Fe, with ligand sources being from particulate organic carbon remineralization and dissolved  
 196 organic matter production. Ligand sinks involve scavenging, uptake by phytoplankton, ultraviolet radiation, and  
 197 bacterial uptake or degradation (Long et al., 2021). An overview of the different sources and sinks of DFe used  
 198 in CESM-MARL is given in Figure 1.



199  
 200 **Figure 1: Schematic representation of iron cycle in the ocean component of the CESM model. The texts/boxes/arrows**  
 201 **in black show the main processes affecting the dissolved iron pool, while those in red further show what controls the**  
 202 **processes impacting the dissolved iron pool. POC (DOC): particulate (dissolved) organic carbon, bSi: biogenic silica.**

203  
 204 This study is based on 5 sets of simulations for identifying contributions from different sources of DFe: control  
 205 simulation (CTRL); and simulations that individually remove DFe supply from atmospheric depositions (NATM),  
 206 sediments (NSED), rivers (NRIV) and hydrothermal vents (NVNT). Differences between CTRL and NATM  
 207 simulations indicate the biogeochemical impacts solely due to atmospheric deposition of DFe and is referred to  
 208 as ATM. Similarly, biogeochemical impacts solely from sedimentary, river and hydrothermal DFe sources are,  
 209 respectively, referred to as SED, RIV and VNT cases. Simulations have been conducted in hindcast mode for 60  
 210 years using forcing from the Coordinated Ocean-ice Reference Experiments version 2 (CORE-II) dataset for the

211 years 1948-2007 (Large & Year, 2009). The CORE-II data includes interannual variability and consists of 6-  
212 hourly temperature, air density, specific humidity, 10 m wind-speeds, and sea-level pressure from National  
213 Centers for Environmental Prediction/ National Center for Atmospheric Research (NCEP/NCAR) Reanalysis  
214 (Kalnay et al., 1996). Daily shortwave and longwave radiation are taken from Goddard Institute for Space Studies-  
215 International Satellite Cloud Climatology Project radiative flux profile data (GISS-ISCCP-FD) (Zhang et al.,  
216 2004). Monthly precipitation is combined Global Precipitation Climatology Project (GPCP, Huffman et al., 1997)  
217 and Climate Prediction Center Merged Analysis of Precipitation (CMAP, Xie & Arkin, 1997) data. Monthly  
218 streamflow since 1948 used in this study has been previously derived from gauge data, where linear regression  
219 was also employed using CLM3 model streamflow to fill-in missing data (Dai et al., 2009). The present study  
220 uses the last 10 years of simulations, given its focus on impacts of DFe sources on biogeochemistry of the upper  
221 100 m of the oceans at seasonal scale.

222

## 223 **2.2 Observation data**

224 Monthly climatology for ocean temperature, salinity and nutrients have been obtained from World Ocean Atlas  
225 2018 (WOA18) at  $1^{\circ}\times 1^{\circ}$  spatial resolution (Garcia et al., 2019). Monthly surface chlorophyll concentrations have  
226 been obtained from the European Space Agency Ocean Color Climate Change Initiative (OC-CCI) version 5 at 4  
227 km spatial resolution for the period 2003-2020 (Satyendranath et al., 2019). OC-CCI merges ocean color  
228 information from multiple sensors: Moderate Resolution Imaging Spectroradiometer (MODIS, 2002-present),  
229 Sea-Viewing Wide Field-of-View Sensor (SeaWiFS, 1997-2010), MEidium Resolution Imaging Spectrometer  
230 (MERIS, 2002-2012) and Visible Infrared Imaging Radiometer (VIIRS, 2012-present). The product is bias-  
231 corrected and quality-controlled, yielding much lower data gaps compared to individual sensors. Monthly  
232 climatology of mixed layer depth (MLD) gridded at  $1^{\circ}\times 1^{\circ}$  spatial resolution has been obtained from Argo profiles  
233 based on a hybrid algorithm that calculates a suite of MLDs using several criteria, such as gradient/threshold  
234 method, maxima or minima of a particular property, intersection with seasonal thermocline (Holte et al., 2017).  
235 The resulting patterns are analysed to yield final MLD estimates. To explore ocean surface circulation, Ocean  
236 Surface Current Analysis Real-time (OSCAR) data at  $0.33^{\circ}\times 0.33^{\circ}$  spatial resolution and 5-day temporal resolution  
237 has been used. Horizontal velocities are measured using sea surface heights, ocean surface winds, and sea surface  
238 temperatures, thereby accounting for flows due to geostrophic balance, Ekman dynamics, and thermal wind  
239 (Dohan & Maximenko, 2010).

240

241 To examine the ability of CESM to realistically simulate the variation in DFe concentrations in the upper 100 m  
242 over the northern IO, this study uses DFe profile compilations by Tagliabue et al. (2012) and the GEOTRACES  
243 Intermediate Data Product 2021 (Schlitzer et al., 2021). To these, published data from Moffett et al. (2015) has  
244 also been added, comprising DFe data collected in the AS during September 2007. The DFe estimated in these  
245 data are based on filtration of seawater through filter sizes between 0.2-0.45  $\mu\text{m}$ .

246

## 247 **3 Results and discussions**

248 First, the performance of CESM-POP2 simulations with respect to observations over the northern IO is examined.  
249 Next, the contributions of different DFe sources to upper ocean DFe concentrations, phytoplankton blooms and

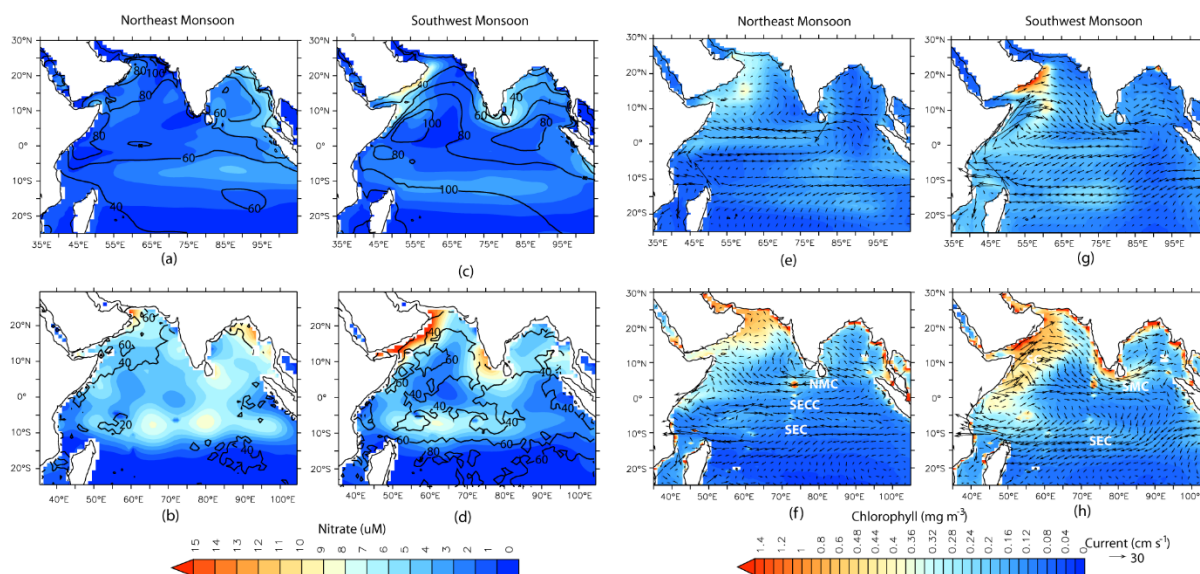
250 patterns of nutrient limitations is discussed. Finally, the paper explores how different sources of DFe can influence  
251 the total DFe budget across selected biophysical regimes over the northern IO.

### 252 **3.1 Model evaluation**

253 In this section CESM simulation (for CTRL case) of physical parameters as well as nitrate and chlorophyll  
254 concentrations over the upper 100 m of the northern IO is evaluated. Except for MLD, ocean currents, and  
255 chlorophyll, all modeled parameters have been compared with WOA18 observations. Simulated MLDs are  
256 compared with Argo-based values of Holte et al. (2017), ocean currents are compared with OSCAR data, and  
257 chlorophyll concentrations are compared with OC-CCI observations. In general, CESM shows good  
258 correspondence with observations of seasonal cycle of temperature, salinity and MLD. However, there is a  
259 positive temperature and salinity bias over IO (Figs. S2 and S3 in the Supplement). This warm bias over IO differs  
260 from the previous version of CESM, which has a cold bias in this region (Danabasoglu et al., 2020). Figure 2  
261 shows seasonal climatology in CESM simulations and observations, for MLD, nitrate concentrations, surface  
262 ocean currents, and chlorophyll concentrations. Overall, CESM simulates the main features of surface ocean  
263 circulation and spatio-temporal variations in MLD well. There are some deviations, such as a much stronger  
264 simulated Somali Current along the northeast coast of Africa, especially during the southwest monsoon season,  
265 which can lead to strong advection of upwelled nutrients away from this region. CESM also simulates a stronger  
266 South Equatorial Current during southwest monsoon, which occupies a broader region compared to observations  
267 and leads to a stronger westward flow in the model between 0-5°S latitude. The net result of the warm and positive  
268 salinity bias is that CESM simulates much deeper MLD than observations throughout the year across the study  
269 domain. Averaged annually, the largest overestimation (of ~40 m) is over the equatorial IO particularly during the  
270 spring and fall intermonsoon months, when the Wyrki Jet is prevalent over the region (Figs. S3 e-f). Additionally,  
271 MLD overestimation of ~45 m is also seen over the AS during February-March and the southern tropical IO  
272 during September-October, both associated with winter-convection.

273 With respect to the seasonal cycle of nitrate, CESM has the least bias over AS followed by BoB (Figs. 2a-d and  
274 S4), but its performance is comparatively lower over the equatorial IO and southern tropical IO. For example,  
275 WOA18 data shows the highest value of nitrate over southern tropical IO in January, whereas in CESM simulation  
276 the highest nitrate concentration is shifted to April-June associated with mixed layer deepening. On the other  
277 hand, CESM simulates a much weaker seasonal cycle of nitrate over the equatorial IO compared to WOA18  
278 observations. These regions, over southern tropical IO, and the equatorial IO, where CESM fares poorly also have  
279 fewer nutrient profile observations compared to AS and BoB. For example, no more than 10 nitrate observations  
280 are available in a grid-point over the southern tropical IO and equatorial IO, whereas there are several grid-points  
281 over the AS where more than 30 observations are available. Overall, CESM simulations underestimate nitrate  
282 with respect to WOA18 data for the upper 100 m of the water column.





283

284 **Figure 2: Comparison of CESM-CTRL simulated variables (upper panels) with observations (lower panels) for**  
 285 **northeast monsoon (a,b,e,f) and southwest monsoon (c,d,g,h). Shading in (a-d) are nitrate concentrations averaged for**  
 286 **upper 100 m and the black contours are the mixed layer depth (m). Shading in (e-h) are surface chlorophyll**  
 287 **concentrations and the vectors are the surface currents. SEC: South Equatorial current, SECC: South Equatorial**  
 288 **Counter Current, NMC: Northeast Monsoon Current, SMC: Southwest Monsoon Current, SC: Somali Current.**

289

290 Turning to chlorophyll concentrations, CESM simulations capture the main characteristics of the seasonal cycle  
 291 and its spatial distribution over the northern IO (Figs. 2e-h and S4), with certain biases and shifts in the timing of  
 292 the peak blooms. For example, over the BoB, the model has difficulty in capturing the temporal evolution of  
 293 chlorophyll concentrations. Over the AS and the equatorial IO, peak bloom in the simulations occurs in September,  
 294 in contrast to July in the observations. Similarly, over the southern tropical IO, the peak bloom is delayed in the  
 295 model to October as compared to its appearance in July in observations. Most of the AS and the BoB show  
 296 underestimation ( $\sim -60\%$ ) in simulated chlorophyll concentration with respect to OC-CCI values. Such  
 297 underestimation of major nutrients and chlorophyll over most of the northern IO are common to many modelling  
 298 studies where coastal regimes and mesoscale processes are not adequately captured without finer spatial resolution  
 299 (e.g., Dutkiewicz et al., 2012; Ilyina et al., 2013; Long et al., 2021; Moore et al., 2013b; Pham & Ito, 2021). For  
 300 example, a modelling study by Resplandy et al. (2011) has shown that eddy-induced vertical transport is  
 301 responsible for  $\sim 40\%$  of nitrate fluxes in the winter convection regions of the AS during the late northeast  
 302 monsoon. The study also showed that mesoscale eddies can account for 65-91% of vertical and lateral advection  
 303 of nitrate in the upwelling regions of the AS during the southwest monsoon. Additionally, the positive MLD bias  
 304 simulated by CEM can trigger light limitation of phytoplankton growth, leading to underestimation of  
 305 chlorophyll. If the threshold depth for photosynthesis is considered as the depth of the isolume given by  $0.415$   
 306  $\text{mol quanta m}^{-2} \text{d}^{-1}$  ( $Z_{0.415}$ , Boss & Behrenfeld, 2010; Letelier et al., 2004), then the CEM simulated MLD is  
 307 deeper than the  $Z_{0.415}$ , leading to light limitation of phytoplankton growth over the entire AS and large parts of  
 308 BoB throughout the year (Fig. S5). During the southwest monsoon, almost the entire domain experiences light  
 309 limitation, especially off the coast of Somalia and the southern tropical IO.

310 CEM simulations of DFe are evaluated next, using all available *in situ* DFe concentration data for upper 20 m  
 311 of the ocean, for different seasons. In addition, distribution of DFe along selected transects for the upper 100 m

312 are studied: (1) CLIVAR cruise 109N along the eastern IO during April 2007; and (2) GEOTRACES cruises GI-  
313 01, GI-02, GI-03, GI-04 and GI-05. While CESM simulates the general pattern of DFe distribution over the  
314 northern IO reasonably well, DFe variation with depth and with increasing distance from the coast is stronger in  
315 simulations than in observations. For upper 20 m, Pearson product-moment correlation coefficient calculated  
316 between observed and simulated DFe concentrations is 0.62 (Figs. 3a-d). The coefficients for correlation between  
317 observed and simulated DFe for GEOTRACES and CLIVAR transects vary between 0.64 and 0.38 (Fig. 3e). All  
318 these correlation coefficients are significant at 95% confidence level based on Student's t-test with n-2 degrees of  
319 freedom, where n is the sample size. This indicates that CESM is able to reproduce the north-to-south gradient in  
320 DFe concentrations, the comparatively low DFe concentration west of 65°E over the AS, as well as increases in  
321 DFe with depth over both the eastern and western IO reasonably well. Overall, CESM simulates positive bias in  
322 DFe concentration over the study domain (see Table S1). A closer look at the pattern of bias in simulated DFe  
323 reveals several features: (1) the magnitude of the positive bias is much lower to the south of 5°S latitude compared  
324 to the north, (2) CESM simulated DFe has low magnitude of negative bias to the west of 60°E longitude over the  
325 AS near the dust sources and (3) Coastal and open oceans experience similar magnitudes of positive DFe bias  
326 throughout the domain, implying that DFe bias might be stemming from multiple sources.

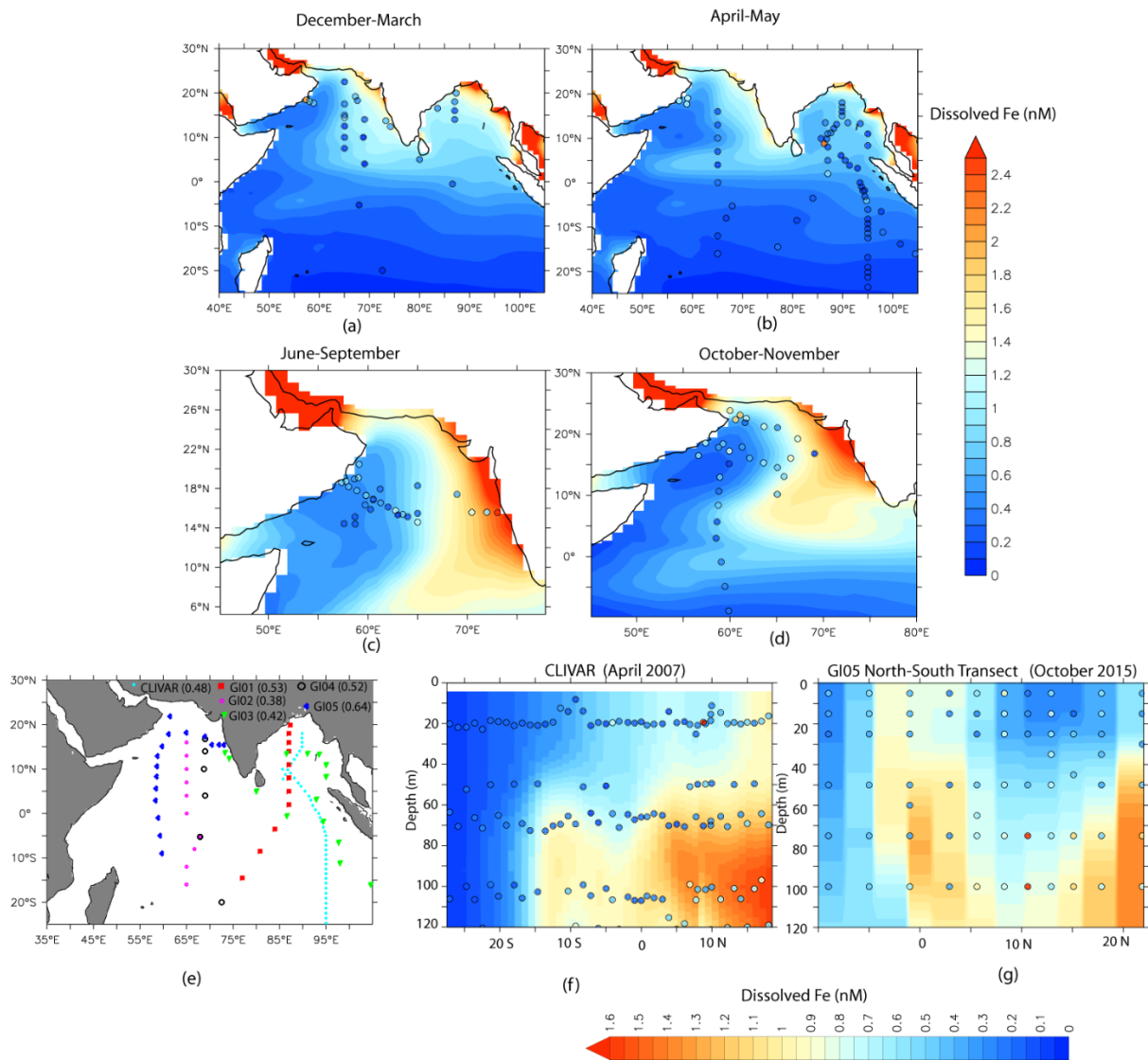
327 Figures 3 f and g show two examples of variation of DFe distribution with latitude and depth along the eastern  
328 and western IO, respectively. The model overestimates DFe values, especially to the north of the equator and at  
329 depths greater than 60 m. Such overestimation of DFe over the northern IO in CESM could result from a variety  
330 of factors, like source strength, assumed solubility of iron, and uncertainties in the removal of DFe by biological  
331 uptake as well as scavenging. With respect to source strength, dust deposition is one possible factor that can lead  
332 to overestimation of simulated DFe. Using Dust Indicators and Records of Terrestrial and MARine  
333 Palaeoenvironments (DIRTMAP) version 2 database of modern day dust deposition (Kohfeld & Harrison, 2001)  
334 an attempt has been made here to understand CESM bias in dust deposition over AS. Median dust deposition  
335 values from DIRTMAP ranges between  $\sim 14 \text{ g m}^{-2}\text{yr}^{-1}$  over the western AS (40°-60°E),  $\sim 7 \text{ g m}^{-2}\text{yr}^{-1}$  over the central  
336 AS (60°-70°E) and  $\sim 20 \text{ g m}^{-2}\text{yr}^{-1}$  over the eastern AS (70°-80°E) (Kohfeld & Harrison, 2001). Corresponding  
337 median values of dust deposition over these locations from CESM model are  $5 \text{ g m}^{-2}\text{yr}^{-1}$ ,  $9 \text{ g m}^{-2}\text{yr}^{-1}$  and  $14 \text{ g m}^{-2}\text{yr}^{-1}$   
338 respectively, indicating a general underestimation of dust deposition by CESM, especially to the west of 60°E  
339 longitude. Over the eastern IO, using mixed layer dissolved Al concentrations dust depositions have been  
340 estimated to be  $0.2\text{-}3.0 \text{ g m}^{-2}\text{yr}^{-1}$  between 20°S to 10°N latitude (Grand et al., 2015). In a separate study, based on  
341 Al concentrations in the aerosol, Srinivas and Sarin (2013) have estimated dust dry-deposition flux of  $0.3\text{-}3.0 \text{ g m}^{-2}\text{yr}^{-1}$   
342 over BoB. Dust deposition from CESM is on the lower end of this range varying from  $1.1 \text{ g m}^{-2}\text{yr}^{-1}$  over  
343 the northern BoB to  $0.2 \text{ g m}^{-2}\text{yr}^{-1}$  near the equator. Sediment traps deployed at shallow depths over the BoB have  
344 recorded annual lithogenic fluxes varying from the northern to the southern bay as  $\sim 15 \text{ g m}^{-2}\text{yr}^{-1}$  ( $\sim 89.5^\circ\text{E}$ ,  $17.5^\circ\text{N}$ )  
345 to  $\sim 4 \text{ g m}^{-2}\text{yr}^{-1}$  ( $87^\circ\text{E}$ ,  $5^\circ\text{N}$ ) (Unger et al., 2003). The corresponding variations in CESM dust deposition are  $\sim 9 \text{ g m}^{-2}\text{yr}^{-1}$ ,  
346 to  $\sim 2 \text{ g m}^{-2}\text{yr}^{-1}$ . Thus, overall, there is some underestimation of dust deposition over the northern IO, which  
347 might not explain positive DFe bias in CESM simulations. However, there is a possibility of fractional solubility  
348 of Fe from dust having an impact on DFe derived from atmospheric sources. Over the AS, percentage solubility  
349 of aerosol has been reported to vary between 0.02 and 0.43% (Srinivas et al., 2012). Considering that Fe constitutes  
350 3.5% of dust by weight and using 0.02% and 0.5% as the lower and upper bounds to Fe solubility, the total fluxes  
351 of soluble Fe based on CESM dust deposition are calculated. The calculated iron flux ranges from 0.002 (0.04)

352  $\mu\text{mol m}^{-2} \text{d}^{-1}$  over the western AS to 0.01 (0.35)  $\mu\text{mol m}^{-2} \text{d}^{-1}$  over the eastern AS for 0.02% (0.5%) solubility. The  
353 corresponding ranges of soluble Fe flux from CESM is 0.05  $\mu\text{mol m}^{-2} \text{d}^{-1}$  in the west to 0.8  $\mu\text{mol m}^{-2} \text{d}^{-1}$  in the  
354 eastern AS. Again, using median dust deposition values from DIRTMAP data and assuming 0.5% iron solubility,  
355 soluble Fe fluxes vary from 0.12 to 0.17  $\mu\text{mol m}^{-2} \text{d}^{-1}$  from west to east AS. It is therefore clear that CESM model  
356 input of soluble Fe from atmosphere is overestimated compared to observations. This inference does not change  
357 even after adding the contribution of black carbon (after assuming 6% solubility of Fe) to the atmospheric iron  
358 flux. This is because fractional solubility of Fe in CESM varies from 1.2% over northwestern AS to ~5% over the  
359 southern AS. Ship-based measurements, on the other hand, have observed that high levels of  $\text{CaCO}_3$  in the dust  
360 over the AS acts as a neutralizing agent, leading to much lower aerosol solubility (Srinivas et al., 2012).  
361 Additionally, for the GI05 transect (Fig. 3g), DFe concentration reduces drastically in the NATM case (Fig. S6 a-  
362 c), indicating that dust deposition and its solubility is the major factor contributing to the simulated levels of DFe  
363 and its biases.

364 The impact of dust solubility on DFe concentration, however, does not explain the positive biases in simulated  
365 DFe over the BoB. The percentage solubility of aerosol iron measured over the BoB is high, varying between  
366 2.3% and 24%, due to presence of acid species from anthropogenic activities (Srinivas et al., 2012). This leads to  
367 much higher soluble iron deposition than that is obtained from CESM. For example, in CESM the soluble Fe flux  
368 over BoB varies from ~0.05 to 0.35  $\mu\text{mol m}^{-2} \text{d}^{-1}$ , whereas, calculated soluble Fe flux varies from 0.06 to above 1  
369  $\mu\text{mol m}^{-2} \text{d}^{-1}$ . Thus, atmospheric supply of iron is possibly underestimated over the BoB. It is, therefore, quite  
370 possible that this positive bias in DFe stems from either sedimentary or river sources. In fact, comparing CTRL  
371 simulation with NATM and NSED along the CLIVAR transect in Figure 3f, reveals considerable contribution of  
372 sedimentary sources of DFe, especially at depth greater than 60 m (Fig. S6 d-f). Furthermore, the latitudinal  
373 change in salinity along this transect closely follows the latitudinal pattern of change in DFe from NATM case,  
374 but not DFe from NSED case. To examine this, DFe from NATM and NSED cases and salinity from CTRL case  
375 have been taken along the CLIVAR transect from depths greater than 60 m and have been detrended. The  
376 correlation between DFe from NATM and salinity is -0.75 indicating that non-atmospheric sources of DFe is  
377 associated with fresher water transported from the coastal regions. The corresponding correlation between DFe  
378 from NSED and salinity is -0.16 indicating that non-sedimentary sources of DFe has no salinity dependence. The  
379 underestimation of atmospheric iron deposition along with salinity-dependence of DFe from the NATM case  
380 together indicates that enhanced transport of sediments from continental margins is likely to be the source of DFe  
381 bias along the CLIVAR transect. One possible explanation is that the low resolution of the model is unable to  
382 capture the high velocity of the coastal currents that may limit the spreading of sediments from the coastal regions  
383 to the open oceans. The simulated coastal current is weaker than OSCAR observations during April, when the  
384 CLIVAR measurements were undertaken (Fig. S6 g-h). This can lead to greater diffusive spreading of iron from  
385 the coast into the open ocean. Such an effect of model resolution has been previously shown to result in a higher  
386 sedimentary contribution to DFe off the northwest Pacific and southwest Atlantic ocean (Harrison et al., 2018).

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390 **Figure 3: Comparison of CESM-CTRL simulated DFe (shading) with the observations (filled circles) compiled from**  
 391 **various cruises. The spatial distribution maps in (a-d) consider season-wise DFe distribution averaged over the upper**  
 392 **20 m. (e) The different cruise tracks from which DFe measurements have been used are marked. The numbers within**  
 393 **the parentheses are the correlation coefficients between observed and simulated DFe for each cruise. The vertical**  
 394 **transects in (f-g) show DFe gradients in the water column over (f) the eastern Indian Ocean and (g) the western Indian**  
 395 **Ocean.**

396 With respect to loss terms, biases in Fe uptake and scavenging can impact simulated DFe concentrations,  
 397 especially in the surface waters. To account for Fe uptake by phytoplankton, particulate organic carbon export  
 398 fluxes at 100 m calculated from  $^{234}\text{Th}$  fluxes have been used in conjunction with Fe:C ratios. Since the cellular  
 399 Fe:C ratio varies widely depending on external DFe availability and phytoplankton species composition, a lower  
 400 bound of  $6 \mu\text{mol mol}^{-1}$  and an upper bound of  $50 \mu\text{mol mol}^{-1}$  have been considered. The lower bound is based on  
 401 measurements over the eastern IO (Twining et al., 2019) where oligotrophic conditions are encountered. The  
 402 upper bound is based on measurements over the tropical North Atlantic where high dust deposition leading to  
 403 high surface DFe concentration prevails (Twining et al., 2015). Combining Fe:C values with particulate organic  
 404 carbon export fluxes from JGOFS cruises (Buesseler et al., 1998) yields Fe uptake by phytoplankton varying  
 405 between  $\sim 0.0004$  and  $\sim 0.0035 \mu\text{mol m}^{-3} \text{d}^{-1}$  for all seasons over the AS. Phytoplankton Fe uptake from CESM  
 406 over the AS varies between  $\sim 0.0001$  and  $\sim 0.002 \mu\text{mol m}^{-3} \text{d}^{-1}$ , which are on the lower end of observation-based

407 values. Over the BoB, phytoplankton Fe uptake varies between  $\sim 0.00002$  and  $\sim 0.004 \mu\text{mol m}^{-3} \text{d}^{-1}$  based on  
408 available POC measurements (Anand et al., 2017; 2018). The corresponding ranges of CESM simulated DFe  
409 uptake are  $\sim 0.0002$  to  $\sim 0.001 \mu\text{mol m}^{-3} \text{d}^{-1}$ , which is within the range of values calculated from observations. With  
410 respect to scavenging losses, based on particulate Fe value from the eastern tropical South Pacific and  $^{234}\text{Th}$  fluxes  
411 over the AS, Chinni and Singh (2022) estimated abiotic removal of  $0.001\text{-}0.005 \mu\text{mol m}^{-3} \text{d}^{-1}$  for the upper 100 m.  
412 In the present simulations, average scavenging removal is  $\sim 0.003 \mu\text{mol m}^{-3} \text{d}^{-1}$  over both the AS and BoB (range:  
413  $0.002$  to  $0.026 \mu\text{mol m}^{-3} \text{d}^{-1}$ ) and reduces to less than  $0.001 \mu\text{mol m}^{-3} \text{d}^{-1}$  to the south of the equator. Overall, Fe  
414 uptake by phytoplankton is possibly underestimated over the AS, which can contribute to some overestimation of  
415 DFe in the surface waters over this region. Over BoB, Fe uptake is within the range of observation-based values.  
416 Scavenging removal simulated by CESM is also within the range of observation-based values and is possibly not  
417 contributing to DFe bias in CESM.

418 To summarize, the ocean component of CESM has deeper MLD than observations, underestimates nitrate and  
419 chlorophyll, and overestimates DFe concentrations. Together, this can result in weaker iron-limitation in the  
420 simulations compared to observations. Over the AS, the positive bias in simulated DFe is present mostly to the  
421 east of  $60^\circ\text{E}$  longitude and can be related to the higher solubility of atmospheric iron in CESM compared to the  
422 observations. Over the BoB, DFe bias likely originates from enhanced transport of sedimentary iron from  
423 continental shelf margins. To the west of  $60^\circ\text{E}$ , simulated DFe has negative bias of low magnitude, possibly  
424 because underestimation of dust deposition is counterbalanced by overestimation of iron-solubility. Over the  
425 southern tropical IO, the magnitude of bias is also low compared to the rest of the study domain. Still, the model  
426 simulates spatial and temporal patterns of ocean physical features, as well as variations in chlorophyll  
427 concentrations, nitrate, and DFe concentrations over the northern IO reasonably well. This gives confidence in  
428 using the model to study the iron cycle over the region. Taking the above understanding of strengths and  
429 shortcomings of the model into account, the importance of different DFe sources with respect to biogeochemistry  
430 of the upper 100 m of the northern IO is explored next.

431

### 432 **3.2 Contribution of multiple iron sources**

433

434 Figure 4 summarizes the contributions of different sources to annually averaged DFe concentration. Source-wise  
435 DFe contributions for northeast and southwest monsoons are shown in Figs. S7 and S8, respectively. Overall, the  
436 relative contribution from different sources to DFe is nearly the same across different seasons, except for the  
437 somewhat higher contribution of atmospheric DFe during southwest monsoon compared to northeast monsoon.  
438 This is because the arid and semi-arid regions surrounding the northern IO experiences maximum dust activity  
439 from late spring to early southwest monsoon months (e.g., Banerjee et al., 2019; Léon and Legrand, 2003). In the  
440 annual average, atmospheric deposition is the most important source of DFe over the northern IO and contributes  
441 well above 50% of the total DFe concentrations (ATM case in Fig. 4b). Furthermore, atmospheric deposition  
442 contributes more than 70% of DFe supply over most of the AS, southern BoB, and the equatorial IO. The location  
443 of the intertropical convergence zone during northeast monsoon ( $\sim 10^\circ\text{S}$  latitude) determines the southern limit of  
444 the influence of atmospheric deposition because southwards of the intertropical convergence zone there is a rapid  
445 reduction in DFe concentrations. Dust is the predominant contributor to the atmospheric deposition flux of iron.

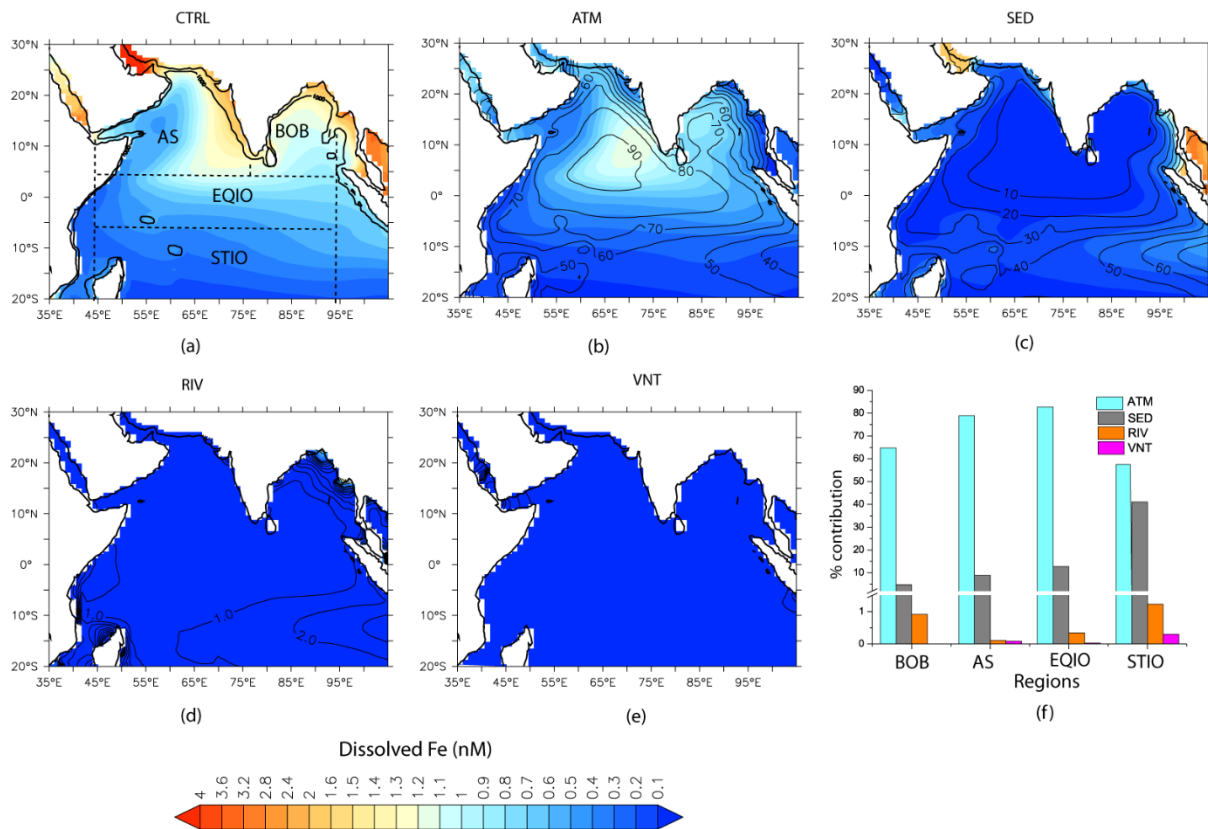
446 Over the northern AS, dust is mostly transported from Iran, Pakistan, Afghanistan, and the Arabian Peninsula,  
447 whereas over southern AS dust from north-eastern Africa also becomes important (Jin et al., 2018; Kumar et al.,  
448 2020). Over northern and southern BoB, the major sources of dust are the Indo-Gangetic Plain and northeast  
449 Africa, respectively (Banerjee et al., 2019). Eastwards of 90°E, black carbon contributes ~50% to atmospheric  
450 DFe flux during the northeast monsoon (not shown). The source of black carbon in this region is biomass burning  
451 and fossil fuel combustion transported from the Indo-Gangetic Plain and Southeast Asia (Gustafsson et al., 2009;  
452 Moorthy & Babu, 2006).

453 The second largest source of DFe is from continental shelf sediments (Fig. 4c), which become dominant in the  
454 vicinity of the shelves. High sedimentary sources of DFe are characteristic of the Andaman Sea where incoming  
455 rivers can contribute  $\sim 600 \times 10^6 \text{ T yr}^{-1}$  of sediments (Robinson et al., 2007). It has been estimated that terrestrial  
456 sources contribute more than 80% to total organic carbon in the inner shelf region of the Gulf of Martaban,  
457 adjacent to the Andaman Sea (Ramaswamy et al., 2008). Elsewhere, sedimentary contributions of ~20% to overall  
458 DFe are found in CESM runs along the northern part of west coast of India and the eastern BoB. Within Ganga-  
459 Brahmaputra system, which is responsible for discharge of  $\sim 11 \times 10^8 \text{ T yr}^{-1}$  of sediments, only 10% of sediments  
460 is estimated to be transported longshore, with most of the sediments accumulating within the shelf and  
461 subterranean canyon (Liu et al., 2009). Over the open ocean, sedimentary sources are most important within 10°-  
462 15°S latitude where the South Equatorial Current is responsible for ~50% of DFe supply via advection from the  
463 Indonesian shelf. During southwest monsoon, sedimentary contribution by the South Equatorial Current extends  
464 farther westward (~70°E longitude, Fig. S8c) compared to the northeast monsoon (~80°E longitude, Fig. S7c).  
465 Signatures of elevated Al due to sedimentary contribution is seen in ship-borne measurements (Grand et al., 2015;  
466 Singh et al., 2020). In fact, such measurements have shown that the South Equatorial Current separates DFe-rich  
467 oxygen-poor water of the northern IO from the DFe-poor oxygen-rich water of the southern tropical IO (Grand et  
468 al., 2015).

469 River sources contribute negligibly to total DFe concentrations (Fig. 4d), except in the immediate vicinity of the  
470 mouths of large river systems in the northeast BoB: the Ganges-Brahmaputra and the Irrawady-Sittang-Salween.  
471 This can arise from the fact that DFe from river is mostly concentrated within the fresher upper 30 m of the water  
472 column to the north of 21°N over the BoB and also due to high scavenging losses of iron at the river mouth.  
473 Hydrothermal vents also contribute negligibly to DFe concentrations in the upper 100 m (Fig. 4e). The  
474 hydrothermal vents supplying DFe (often excess of 1.5 nM) in the northern IO are located in the Central Indian  
475 Ridge and the Carlsberg Ridge (Chinni & Singh, 2022; Nishioka et al., 2013; Vu & Sohrin, 2013), and largely  
476 influence DFe concentrations below 1000 m depths. The shallowest hydrothermal plumes enriched with Fe are  
477 located between ~650-900 m in the Gulf of Aden (Gamo et al., 2015), overlapping with the depth range at which  
478 the Red Sea watermass spreads along the western IO (Beal et al., 2000). Since this watermass occupies  
479 progressively deeper depths with distance, sliding underneath Persian Gulf waters, surface DFe values are not  
480 impacted by these shallower vents. This is in concordance with simulations of Tagliabue et al. (2010) where,  
481 following 500 years of model integration, hydrothermal vents increase globally averaged DFe concentrations by  
482 only ~3% in the depth range of 0-100 m.

483 The average contribution of different sources of iron to the upper 100 m is summarized for different open ocean  
484 regions over the northern IO in Fig. 4f. Annually averaged atmospheric deposition is clearly the most important  
485 source of DFe throughout the northern IO. The exception to the dominant role of atmospheric deposition is the

486 southern tropical IO, where sedimentary sources of iron contribute ~40% to the upper ocean iron budget. Based  
 487 on the analysis of origin of bias in simulated DFe concentrations in Section 3.1, it is likely that contribution of  
 488 atmospheric sources to upper 100 m DFe concentration is overestimated over the eastern AS and the contribution  
 489 of sedimentary sources to upper 100 m DFe concentration is overestimated over the BoB. Averaging over the  
 490 entire domain, atmospheric source contributes ~67% to the upper 100 m DFe concentration. On masking out the  
 491 region to the east of 65°E longitude over the AS, where the highest positive bias of DFe from dust has been noted,  
 492 it is seen that atmospheric source contributes ~65% to the upper 100 m DFe concentration. Again, averaging over  
 493 the study domain, sedimentary source contributes ~30% to the upper 100 m DFe concentration. On masking out  
 494 BoB, where positive bias of DFe from sedimentary sources has been identified in Section 3.1, it is seen that  
 495 sedimentary source contributes ~33% to the upper 100 m DFe concentration. Thus, while biases in the source  
 496 strength might regionally impact the percentage contribution of DFe from various sources to the northern IO, the  
 497 overall conclusion of atmospheric source being the most important for upper ocean DFe over the northern IO,  
 498 followed by sedimentary sources, does not change. River contribution is generally ~1%, with slightly higher  
 499 contributions in BoB and the southern tropical IO. Hydrothermal vents make negligible contributions throughout  
 500 the northern IO. Adding these four sources of DFe estimated from CESM experiments does not yield the full  
 501 100% of the DFe source, owing to non-linear effects associated with iron removal processes as well as  
 502 complexation by organic ligands.



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**Figure 4: Contribution of different sources of DFe averaged over the year to the total DFe concentrations over the upper 100 m. Shading in (a) shows total DFe concentration with all sources included and shadings in (b-e) shows DFe concentrations arising from individual source. Contours in (b-e) show the percentage contribution of each source to total DFe concentrations. (f) Bar chart depicting source-specific DFe contribution (in %) over Bay of Bengal (BOB), Arabian Sea (AS), equatorial IO (EQIO), and the southern tropical IO (STIO). These regions are marked by the dashed boxes in (a). The thick black contour in (a) traces the 1000 m bathymetry.**

### 512 3.3 Phytoplankton responses to multiple iron sources

513 In this section, the impact of different sources of DFe on phytoplankton growth is examined. Since river and  
514 hydrothermal sources make negligible contributions to the upper ocean iron concentrations, as shown above, these  
515 are not considered further.

#### 516 3.3.1 Responses to atmospheric depositions

517 During the northeast and southwest monsoons, atmospheric DFe brings about increases in column-integrated  
518 chlorophyll concentrations over most of the northern IO (Figs. 5 a and c). The largest column-integrated positive  
519 response is seen in the western AS (west of ~65°E longitude) throughout the year, where atmospheric DFe  
520 accounts for more than ~20% of the column-integrated chlorophyll concentration and more than 50% of surface  
521 chlorophyll concentration (Fig. S9). This region comes under the influence of upwelling during the southwest  
522 monsoon and mixed layer deepening due to winter convection during the northeast monsoon, which can supply  
523 macronutrients required for phytoplankton growths (Madhupratap et al., 1996; Morrison et al., 1998). The other  
524 region displaying a strong positive response is the southern tropical IO during June-September, where atmospheric  
525 DFe contributes ~20% (~35%) of the column (surface) chlorophyll concentration. This is the time of the year  
526 when deep mixed layer leads to entrainment of nutrients into the surface layers (Košić et al., 2009; Lévy et al.,  
527 2007). In contrast, there are some regions, like the northern and western AS, the west coast of India and large  
528 parts of the BoB and the eastern IO, which in spite of receiving high atmospheric DFe hardly experience any  
529 chlorophyll response. These regions show <1% increase in column chlorophyll concentrations and generally  
530 coincide with high sedimentary iron input. This is discussed further in Section 3.3.3

531 Species-wise decomposition shows that the increases in chlorophyll during both northeast and southwest  
532 monsoons are driven by increases in diatoms and declines in small phytoplankton (Fig. 6). For example, over the  
533 western AS and southern tropical IO, diatoms increase by at least 40% and small phytoplankton populations  
534 decline by at least 50%. An exception is the equatorial IO, where the positive response of chlorophyll arises from  
535 growth of small phytoplankton. In general, this region has very low levels of macronutrients and is dominated by  
536 picoplankton (Vidya et al., 2013). Those regions exhibiting <1% increase in phytoplankton in response to  
537 atmospheric DFe, in contrast, are characterized by proliferation of small phytoplankton and reductions of diatoms.  
538 Although diazotrophs show positive response to atmospheric DFe addition throughout the region, this group  
539 constitutes only ~1% of total phytoplankton biomass.

540 Such differences in species response to external iron addition arise from differences in nutrient uptake between  
541 different phytoplankton functional groups in CESM. Phytoplankton growth rate ( $\mu_i$ ) is parameterized as a product  
542 of resource-unlimited growth rate ( $\mu_{ref}$  in  $d^{-1}$ ) at a reference temperature of 30°C, and three terms that describe  
543 nutrient limitation ( $V_i$ ), temperature dependence ( $T_f$ ) and light availability ( $L_i$ ). This is expressed as:

$$544 \mu_i = \mu_{ref} V_i T_f L_i \quad (1)$$

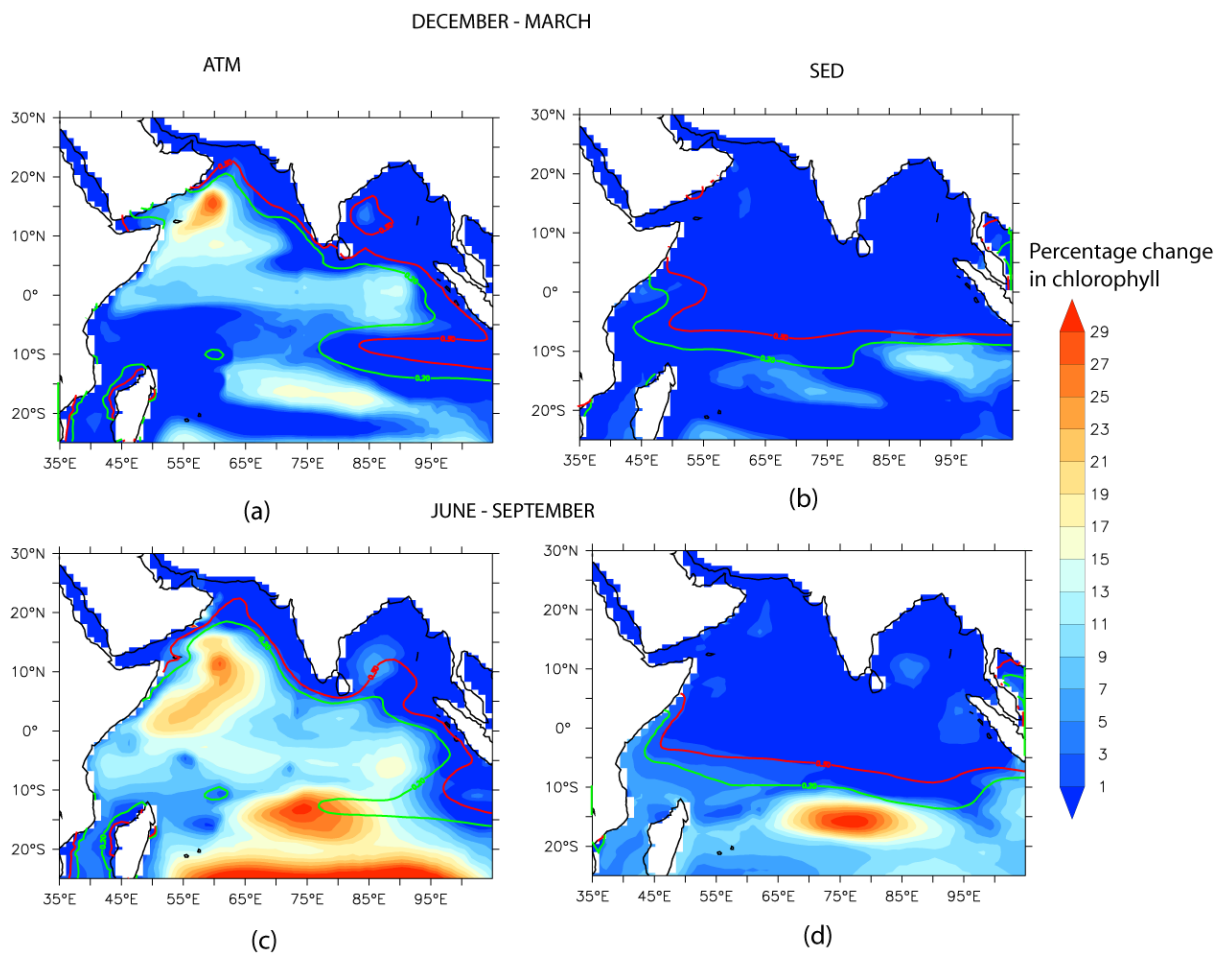
545 The nutrient limitation term for iron,  $V_i$ , for a specific phytoplankton group  $i$  is expressed as:

$$546 V_i^{Fe} = \frac{Fe}{Fe + K_i^{Fe}} \quad (2)$$

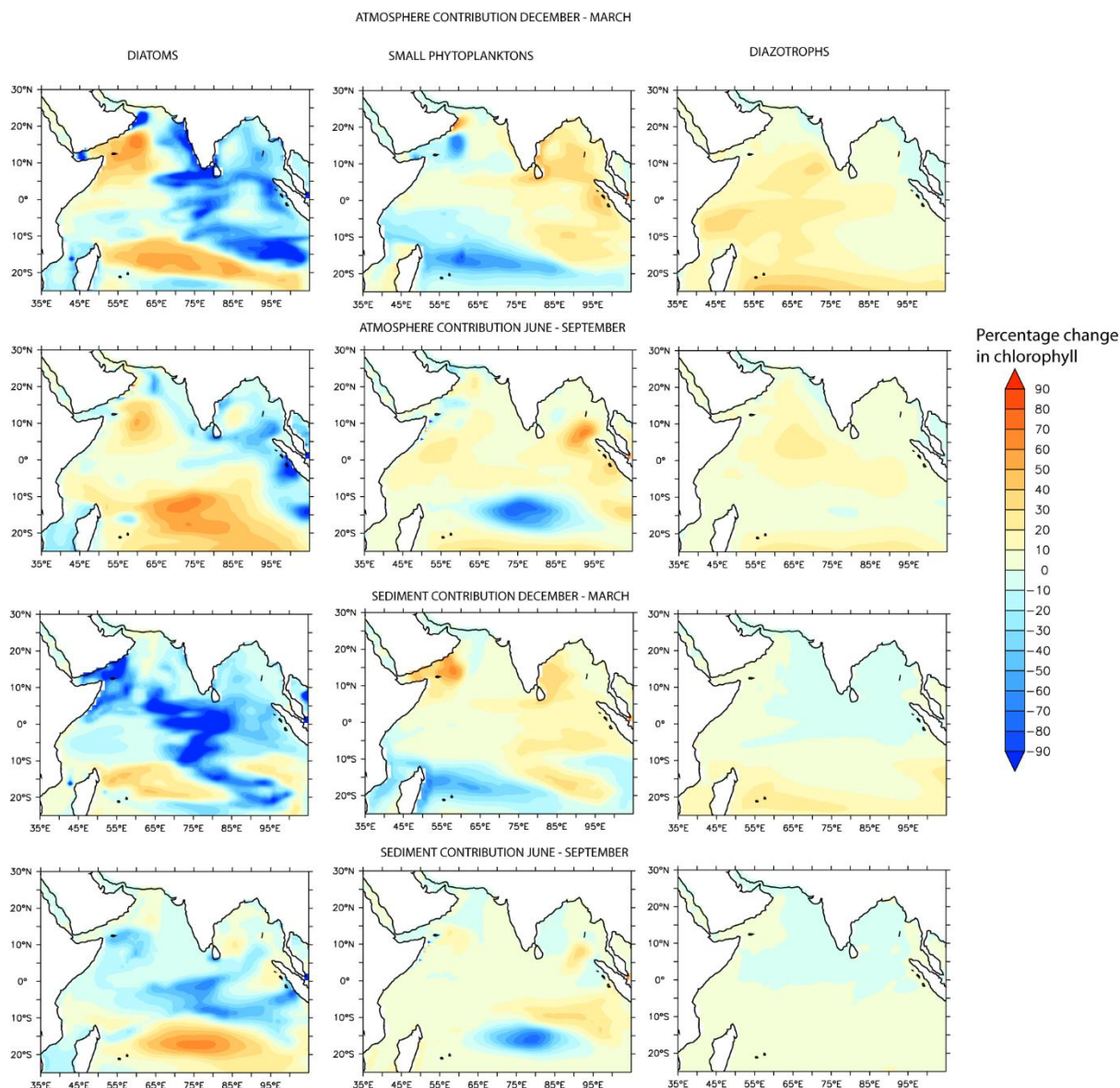
547 where  $Fe$  is the concentration of iron and  $K_i^{Fe}$  is the Fe uptake half-saturation constant for a phytoplankton group.  
548 While small phytoplankton have been assigned a value of  $3.0 \times 10^{-5} \text{ mmol m}^{-3}$  for  $K_i^{Fe}$ , diatoms have been  
549 assigned a higher value of  $7.0 \times 10^{-5} \text{ mmol m}^{-3}$ . This leads to the small phytoplankton outcompeting diatoms when



550 nutrient levels are low. Additionally, small phytoplankton are subjected to higher grazing pressure than diatoms.  
 551 The maximum grazing rate assigned in CESM is  $3.3 \text{ d}^{-1}$  for small phytoplankton versus  $3.15 \text{ d}^{-1}$  for diatoms.  
 552 Together, the differences in nutrient uptake half-saturation constant and grazing pressure between different  
 553 phytoplankton species results in diatom dominating blooms under nutrient-replete conditions.  
 554 Diatoms outperforming other phytoplankton species has been previously witnessed in *in situ* iron fertilization  
 555 experiments along with the existence of a linear relationship between diatom size and iron requirement for growth  
 556 (de Baar et al., 2005). Such shifts in phytoplankton community structure in response to DFe additions are also  
 557 corroborated by *in situ* experiments over the northern IO. For example, a nutrient addition experiment over the  
 558 northern AS during northeast monsoon period has shown that the maximum positive phytoplankton response takes  
 559 place due to nitrate+DFe addition (instead of only DFe addition), accompanied by around four-fold increases in  
 560 coccolithophores, pennate and large centric diatoms (Takeda et al., 1995). Ship-board iron addition experiments  
 561 over the AS during the southwest monsoon resulted in proliferation of visible colonies of haptophyte *Phaeocystis*  
 562 *sp.* due to silicate-limitation (Moffett et al., 2015). Over the eastern IO, where both macronutrients and  
 563 micronutrients are low, nutrient spiking with nitrogen, phosphorus, and iron resulted in increase of  
 564 Prochlorococcus, Synechococcus, as well as Eukaryotes (Twining et al., 2019).  
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 567 **Figure 5: Percentage contribution of (a and c) atmospheric and (b and d) sedimentary sources of iron during (a and b)**  
 568 **the northeast monsoon and (c and d) the southwest monsoon to upper 100 m chlorophyll concentrations. Green and**  
 569 **red contours show background DFe concentrations of 0.2 nM and 0.3 nM respectively. For the ATM (SED) case,**  
 570 **background DFe is obtained from NATM (NSED) simulation.**  
 571



572  
 573 **Figure 6: Species-wise percentage contribution to column chlorophyll (0-100 m) response associated with atmospheric**  
 574 **and sedimentary sources of DFe.**

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 577 **3.3.2 Responses to sedimentary sources of iron**

578 As shown in Fig. 4, sedimentary sources supply less than ~20% of DFe north of ~10°S latitude, whereas between  
 579 10°-15°S latitude sedimentary iron can contribute to almost half of the total DFe concentrations. Unlike  
 580 atmospheric sources, sedimentary supply of DFe is mostly confined to regions adjoining continental shelves and  
 581 islands from where they are introduced to the open ocean by seasonally varying currents. In general, sedimentary  
 582 sources make modest contribution to column productivity (<1% of chlorophyll anomalies) to the north of ~10°S  
 583 latitude as described above. This is because high dust deposition to the north of the intertropical convergence zone  
 584 results in high background DFe concentrations and controls productivity (see also Section 3.3.3). Sedimentary  
 585 sources trigger the strongest positive phytoplankton response over the southern tropical IO region during June-  
 586 September, where sedimentary DFe advected by the South Equatorial Current can facilitate more than 20%  
 587 increase of the upper 100 m chlorophyll concentrations and ~40% increase at the surface. As noted in Section 3.2,

588 although atmospheric deposition contributes nearly half of the total DFe addition to this region, the total iron  
589 deposition here is low ( $<0.2$  nM). The phytoplankton response over the southern tropical IO is dominated by an  
590 increase in diatoms, which contribute to more than 60% of total phytoplankton biomass (Fig. 6). In contrast, over  
591 the regions experiencing  $<1\%$  chlorophyll increase, there is a shift from diatoms towards small phytoplankton  
592 species (Fig. 6). For example, there is more than 80% reduction in diatoms and 50% increase in small  
593 phytoplankton over the western AS. Other current systems such as the poleward flowing Somali current, the  
594 eastward flowing Southwest Monsoon Current and its southward extension along the west coast of Indonesia also  
595 transport sedimentary DFe to the open ocean, but such advection supports only  $\sim 5\%$  phytoplankton biomass.

596 It is important to mention here that DFe bias arising from source strength has low impact on phytoplankton  
597 response to a particular source of DFe. This is because the strongest phytoplankton response to a specific DFe  
598 source is over the western AS and subtropical southern IO. As noted in Section 3.1, these regions have the least  
599 magnitude of DFe bias. For example, averaging over the upper 100 m over the northern IO, atmospheric source  
600 contributes  $\sim 13\%$  to total chlorophyll concentration. Even after masking out the region to the east of  $65^\circ\text{E}$   
601 longitude over the AS, where the highest positive DFe bias arising from atmospheric Fe has been noted, it is seen  
602 that atmospheric source contributes  $\sim 13\%$  to the upper 100 m chlorophyll concentration. Similarly, sedimentary  
603 sources contribute  $\sim 9\%$  to the upper 100 m chlorophyll concentration over the entire northern IO domain. Masking  
604 out BoB, where DFe bias is due to enhanced sediment transport, results in sedimentary source contributing  $\sim 8\%$   
605 to the upper ocean chlorophyll concentration.

606

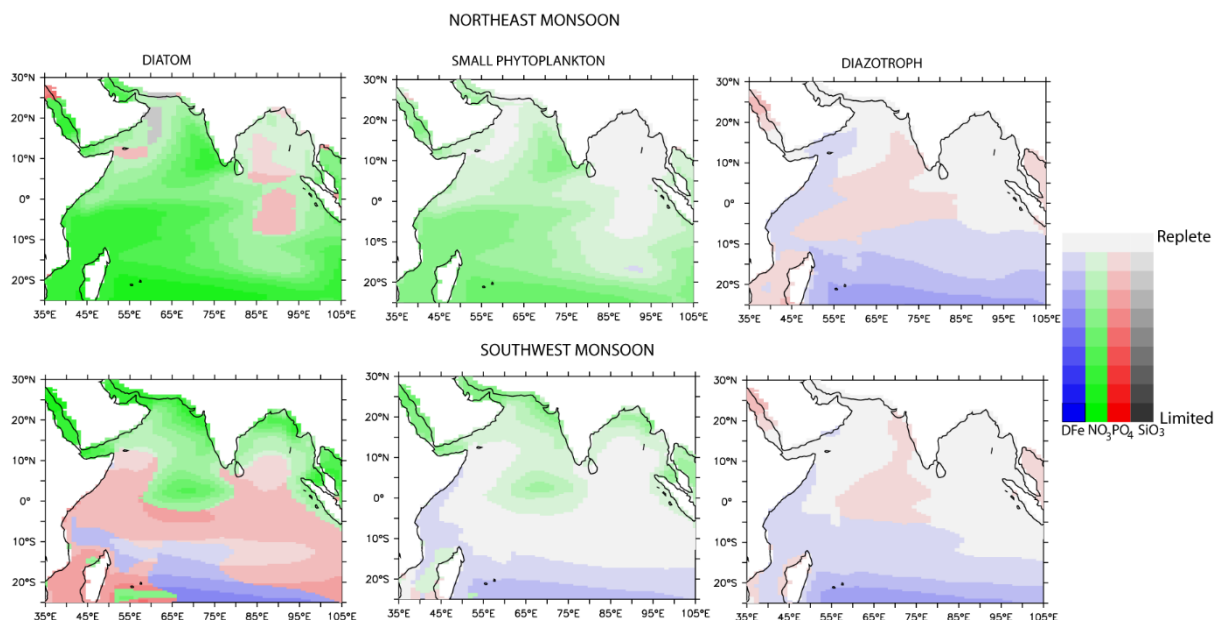
### 607 **3.3.3 Role of background nutrients in phytoplankton responses to external iron**

608

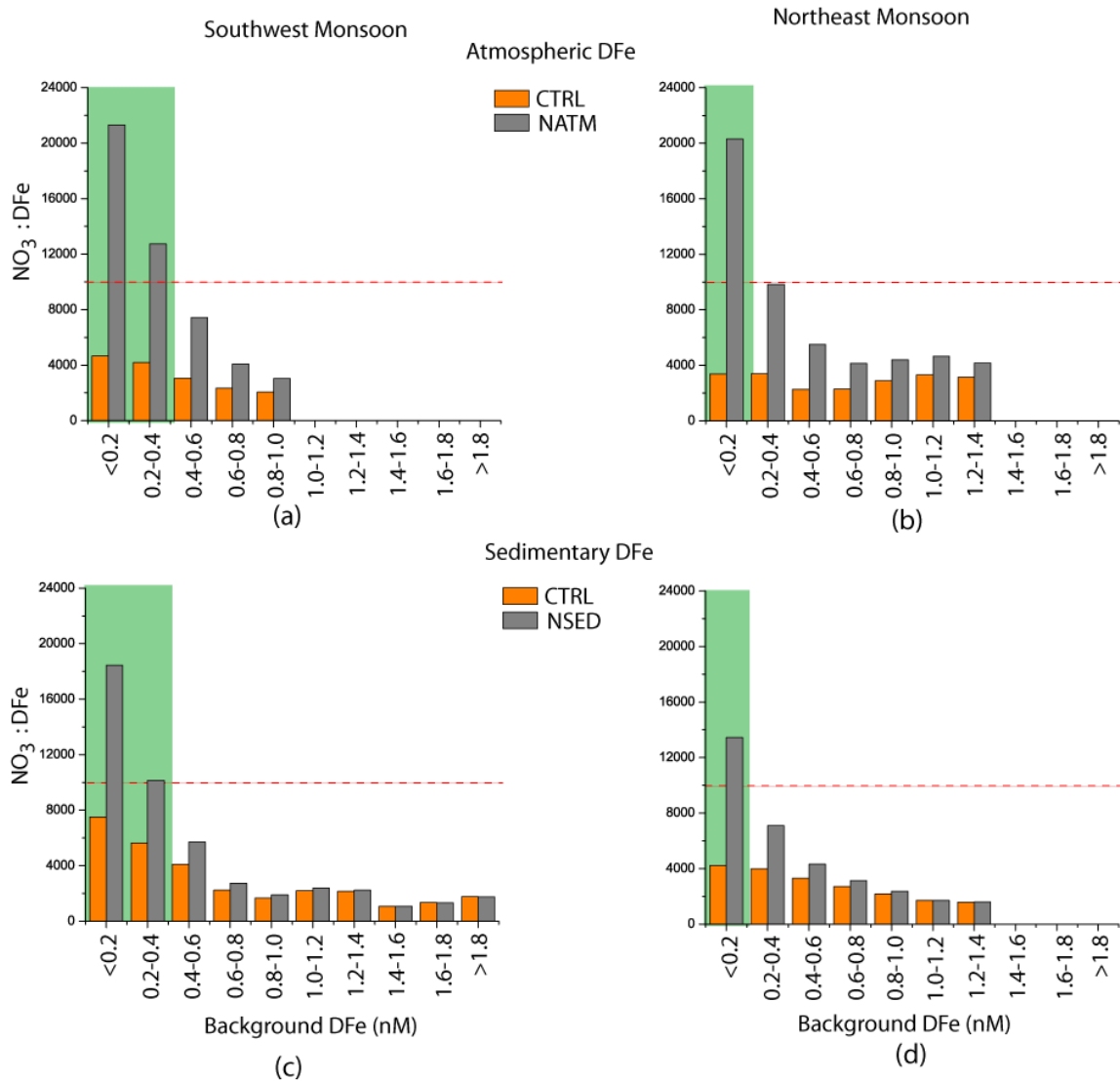
609 It emerges from the previous sections that there is heterogeneity in the phytoplankton response to atmospheric  
610 and sedimentary sources of DFe. The regions of highest DFe input from a specific source are not always the  
611 regions where strongest phytoplankton responses are evoked. What explains these differing patterns of  
612 phytoplankton response? To examine this, patterns of nutrient limitations and iron supply from an external source  
613 with respect to background DFe and nitrate ( $\text{NO}_3$ ) concentrations are examined. In considering the phytoplankton  
614 response to atmospheric sources (ATM case), background DFe is taken from the simulation without any  
615 atmospheric source (NATM). Since river and hydrothermal sources make negligible contributions to DFe over  
616 this domain, high levels of DFe in NATM mainly arise in regions where sedimentary sources are important.  
617 Similarly, for estimating phytoplankton response to sedimentary sources (SED case), background DFe is taken  
618 from simulation without any sedimentary source (NSED).

619 Generally, those regions experiencing greater than 1% increase in chlorophyll in response to atmospheric  
620 (sedimentary) sources coincide with background DFe concentration  $<0.2$ - $0.3$  nM and high background  $\text{NO}_3$ :DFe  
621 ratio from the NATM (NSED) simulation. For example, in NATM simulation, iron serves as the dominant nutrient  
622 that limits productivity over the entire northern IO, with diatoms experiencing stronger iron limitation compared  
623 to other phytoplankton groups (Fig. S10). Iron limitation is particularly severe over central and southern AS,  
624 equatorial IO and the southern tropical IO. In NSED case, there is a switch from nitrate limitation to the north of  
625 the intertropical convergence zone to iron limitation to the south of the intertropical convergence zone (Fig. S11).  
626 While iron stress is alleviated with addition of external DFe, there is a shift towards macronutrient, especially

627 nitrate, limitation (Fig. 7). South of  $\sim 15^{\circ}\text{S}$  latitude continues to experience iron limitation during June-September  
 628 due to very low dust deposition. In contrast, regions where chlorophyll increase is  $<1\%$  following DFe addition  
 629 are characterized by nitrate limitation in NATM/NSED simulations and external DFe cannot alleviate this primary  
 630 nutrient limitation. This is further illustrated in Fig. 8 where upper ocean  $\text{NO}_3:\text{DFe}$  ratio is plotted against  
 631 background DFe concentrations. Positive chlorophyll response is elicited in regions of lowest background DFe  
 632 and highest background  $\text{NO}_3:\text{DFe}$  ratio. Over the world oceans, a wide range of cellular Fe:C ratios has been  
 633 observed for diatoms, ranging from  $100 \mu\text{mol mol}^{-1}$  for DFe-replete conditions (Twining et al., 2015; 2021) to  $2$   
 634  $\mu\text{mol mol}^{-1}$  for DFe-deplete conditions (de Baar et al., 2008). Assuming a C:N ratio of 117:16 (Anderson and  
 635 Sarmiento, 1994), the range of N:Fe ratios obtained are  $\sim 1000$  and  $\sim 68000$ , respectively, for DFe-replete and DFe-  
 636 deplete conditions. Similarly, by considering iron limitation taking place for Fe:C ratio of  $10 \mu\text{mol mol}^{-1}$  for open  
 637 ocean species based on laboratory experiments (Sunda & Huntsman, 1995) and C:N ratio of 106:16, Measures  
 638 and Vink (1999) have estimated that iron limitation over the AS water takes place at  $\text{NO}_3:\text{DFe}$  ratio greater than  
 639  $\sim 15000$ . In CESM simulations  $>1\%$  increase in chlorophyll takes place when initial upper ocean  $\text{NO}_3:\text{DFe}$  ratio  
 640 is more than 10,000 corresponding to Fe-limitation scenario (Fig. 8). With the addition of DFe from atmospheric  
 641 or sedimentary sources, the upper ocean  $\text{NO}_3:\text{DFe}$  ratio reduces to less than 4000 in some cases, thereby leading  
 642 to N-limitation. Previously, iron addition experiments in AS during the southwest monsoon have shown that the  
 643 positive chlorophyll response depends on initial nitrate concentrations, with this response increasing in magnitude  
 644 with higher initial nitrate concentrations (Moffett et al., 2015). In summary, the initial upper ocean  $\text{NO}_3:\text{DFe}$  ratio  
 645 sets the ultimate limit to the magnitude and distribution of phytoplankton response following external DFe  
 646 additions.  
 647



648  
 649  
 650 **Figure 7: Patterns of surface nutrient limitations for different phytoplankton functional types from CTRL simulation.**  
 651 **Green: nitrate; blue: iron; red: phosphate; grey: silicate limitations.**  
 652



653

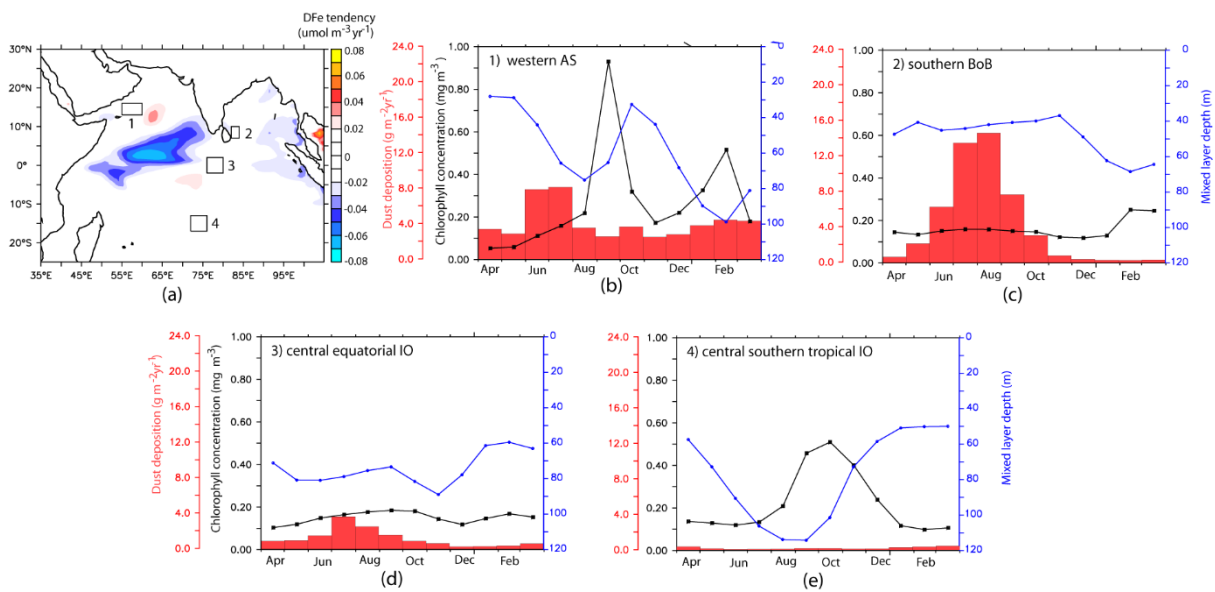
654 **Figure 8: Relation between background nutrients and phytoplankton response for atmospheric (a and b) and**  
 655 **sedimentary (c and d) sources of DFe during (a and c) southwest monsoon and (b and d) northeast monsoon. The**  
 656 **horizontal axis shows background DFe concentrations. The orange columns show upper ocean  $\text{NO}_3 : \text{DFe}$  ratio for**  
 657 **CTRL case and grey columns show  $\text{NO}_3 : \text{DFe}$  ratio for (a-b) NATM and (c-d) NSED cases. The red dashed lines show**  
 658 **the location where  $\text{NO}_3 : \text{DFe}$  ratio is 10,000: below this value N-limitation prevails in CESM. Green shades highlight**  
 659 **the regions where  $>1\%$  increase in chlorophyll following DFe addition from a specific source is induced.**  
 660

661 To sum up, atmospheric deposition is the most important source of DFe to the upper 100 m over the entire northern  
 662 IO, followed by sedimentary sources. While atmospheric DFe is deposited over wide areas of the open ocean,  
 663 sedimentary DFe fluxes arise only from continental shelves and are transported to open oceans through advection  
 664 by currents. River and hydrothermal sources make negligible contributions to the total iron budget in the upper  
 665 100 m. The primary response to atmospheric DFe is an increase in column-integrated phytoplankton biomass over  
 666 most of the northern IO. In contrast, sedimentary source of iron is responsible for increases in column-integrated  
 667 phytoplankton biomass mainly to the south of the intertropical convergence zone, where dust depositions are low.  
 668 In general, significant positive responses of phytoplankton to addition of DFe are simulated only where low levels  
 669 of background DFe concentrations and high values of background  $\text{NO}_3 : \text{DFe}$  ratio are present. Otherwise, nitrate  
 670 becomes the limiting nutrient once DFe is added. The simulations also show that positive chlorophyll response

671 to addition of DFe generally involves proliferation of diatoms, except over the equatorial IO where small  
 672 phytoplankton increase is seen.

673  
 674 **3.4 Iron budgets across different bio-physical regimes**

675 This section explores the main processes controlling DFe budget with respect to the role of atmospheric and  
 676 sedimentary sources over different bio-physical regimes of the northern IO: (1) the western AS, (2) the southern  
 677 BoB, (3) the central equatorial IO and (4) the central southern tropical IO. These regions encompass a wide range  
 678 of productivity, with the first region being highly productive with OC-CCI chlorophyll exceeding  $1.5 \text{ mg m}^{-3}$ . The  
 679 southern BoB and central southern tropical IO are moderately productive. Lastly, the central equatorial IO is  
 680 oligotrophic with surface chlorophyll concentration being  $\sim 0.1 \text{ mg m}^{-3}$ . The locations of these regions along with  
 681 CESM simulated seasonal cycles of mixed layer depths, chlorophyll and dust depositions are shown in Fig. 9.



682  
 683 **Figure 9: (a) Net DFe tendency averaged over the upper 100 m for the study period. The boxes indicate the regions**  
 684 **chosen for further studying DFe budget in Section 3.4. (b-e) Seasonal cycle of dust deposition (red columns), mixed**  
 685 **layer depth (blue curves) and chlorophyll concentrations (black curves) from CESM-CTRL case for the four regions**  
 686 **marked in (a).**

687  
 688 The net dissolved iron tendency ( $TEND_{DFe}$ ) is calculated as:

$$TEND_{DFe} = EXT + ADV + MIX + BIO \quad (3)$$

689 where the source terms on the right describe dust/sediments/rivers/vents (EXT), horizontal and vertical advection  
 690 (ADV), horizontal and vertical mixing (MIX) and biological sources/sinks (BIO). Advection includes explicitly  
 691 resolved velocity as well as an additional “bolus” velocity from parameterization of mesoscale eddies (Gent &  
 692 McWilliams, 1990). Vertical mixing includes a tracer gradient dependent term for cross-isopycnal mixing and a  
 693 non-local mixing term, which accounts for mixing due to convective and shear instabilities (Large et al., 1994).  
 694 Lateral mixing involves parameterization of mesoscale eddy-induced horizontal diffusion along isopycnal  
 695 surfaces (Redi, 1982). The BIO term includes DFe losses due to biological iron uptake and scavenging, recycling  
 696

697 of iron back to the pool via remineralization, and iron released from phytoplankton and zooplankton losses and  
698 grazing.

### 699 3.4.1 Western Arabian Sea

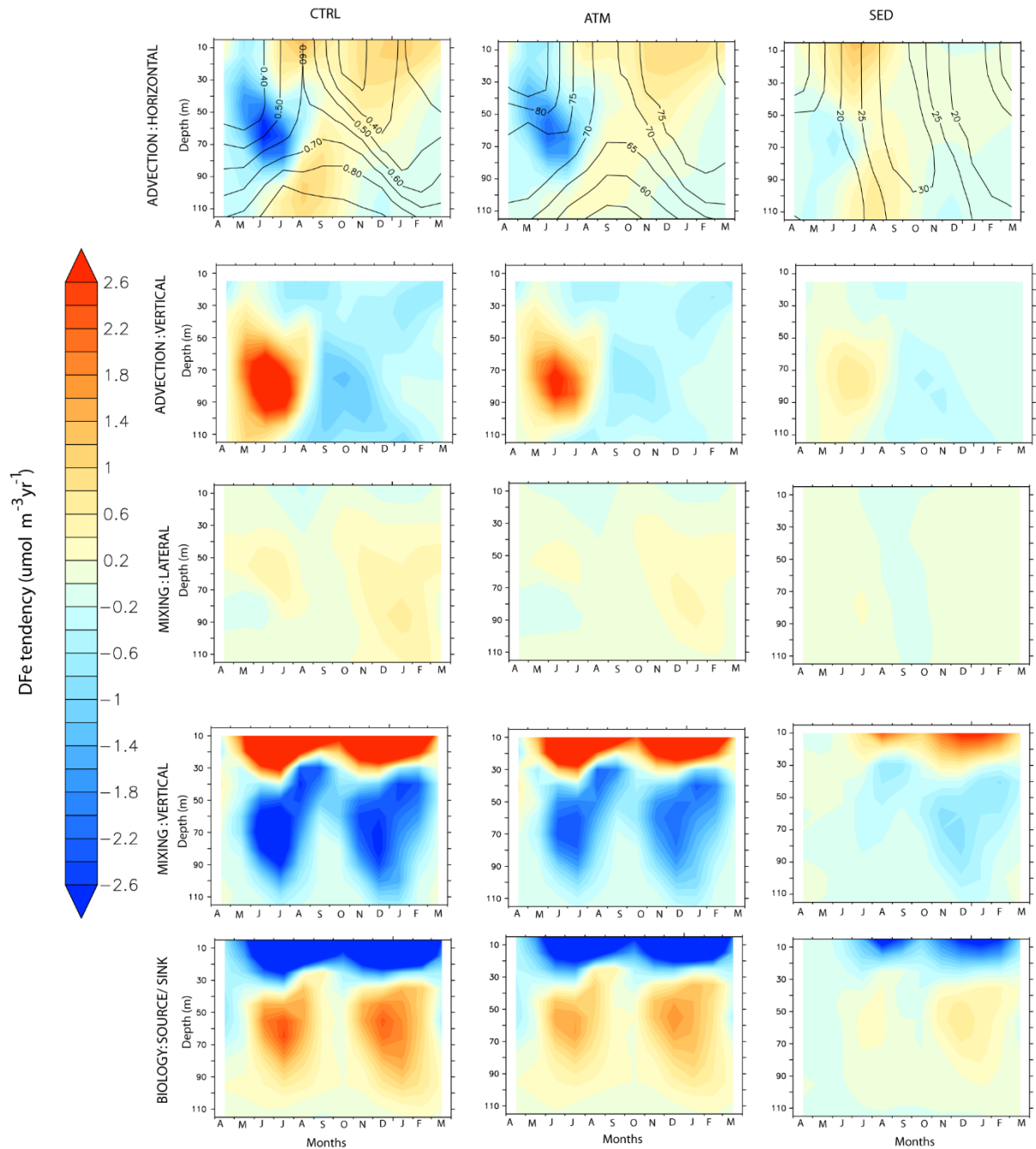
700 The western AS, off Oman and Yemen coastlines (considered here as 13°-16°N and 55°-60°E), is the most  
701 productive region in the northern IO. Primary productivity in the western AS is highest during southwest monsoon  
702 (Fig. 9b), during which alongshore southwesterly winds lead to upwelling and bring subsurface nutrients from  
703 depths of ~150-200 m (Morrison et al., 1998). Some of this upwelled water advects eastwards, transporting  
704 nutrients that enhance productivity in the central AS (Prasanna Kumar et al., 2001). The region also experiences  
705 a secondary bloom during northeast monsoon due to winter convection that deepens the mixed layer. Integrated  
706 over depths of the euphotic zone, average primary productivity over the western AS during mid and late southwest  
707 monsoon is estimated at  $135 \pm 10 \text{ mmol C m}^{-2} \text{ d}^{-1}$  and  $110 \pm 11 \text{ mmol C m}^{-2} \text{ d}^{-1}$  respectively (Barber et al., 2001). In  
708 comparison, primary productivity over the western AS during mid and late northeast monsoon is  $137 \pm 13 \text{ mmol}$   
709  $\text{C m}^{-2} \text{ d}^{-1}$  and  $88 \pm 4 \text{ mmol C m}^{-2} \text{ d}^{-1}$  (Barber et al., 2001). Although this region encounters high dust deposition  
710 (Haake et al., 1993; Mahowald et al., 2009), *in situ* measurements have hypothesized possible iron limitation  
711 during late southwest monsoon because upwelled water is drawn from above the iron-rich sub-oxic zone (Naqvi  
712 et al., 2010).

713 The largest peak in dust deposition is during southwest monsoon, followed by a second peak during northeast  
714 monsoon (Fig. 9b). Accordingly, the upper ocean DFe concentration is highest during southwest monsoon and is  
715 dominated by atmospheric sources (Fig. 10). Sedimentary contribution, although much lower, peaks during late  
716 southwest monsoon and fall intermonsoon months. Throughout the year DFe concentration increases with depth,  
717 thus pointing to consumption by phytoplankton at the surface. Vertical advection and vertical mixing are the most  
718 important physical mechanisms governing DFe supply within this region during southwest monsoon (Fig. 10).  
719 These processes begin to strengthen from May onwards to reach their peak during June-July and decrease  
720 thereafter. Decomposing DFe advection tendency into tendencies arising from gradients in tracer distribution  
721 ( $D\text{Fe}'$ ) and velocity convergence ( $U'$ ) respectively, it is seen that vertical advection of DFe arises from  $D\text{Fe}'$  and  
722  $U'$  in equal magnitude. However, the former process is dominant in June and the latter process dominates during  
723 July (Fig. S12). The maximum vertical advection of DFe is centered around 80 m depth and progressively reduces  
724 at shallower depths, as the vertical velocity reduces towards the surface. Vertical mixing prevailing in the upper  
725 40 m brings this vertically advected DFe from subsurface to the surface. Furthermore, horizontal advection plays  
726 an important role in redistributing this DFe supplied by vertical processes, with contributions from horizontal  $U'$   
727 being at least twice as large as  $D\text{Fe}'$ . During spring and early southwest monsoon, northeastward horizontal  
728 advection removes atmospheric deposited DFe throughout the upper 100 m, while aiding the supply of  
729 sedimentary DFe from Somalia and Omani continental shelves to the western AS. Later in the year as the  
730 southwest monsoon current circulation is established, and meridional currents along the western AS become  
731 stronger, its effect is first evident in the south along the Somali coast and progresses northward with time. The  
732 result is convergence of both atmospheric and sedimentary DFe in the western AS during July-September. During  
733 northeast monsoon, vertical mixing driven by winter convection, with the mixed layer deepening to 100 m, is the  
734 most important means of DFe supply, from both atmospheric and sedimentary sources, into the surface layer.

735 Additionally, horizontal advection by westward currents transports DFe from atmospheric deposition in the central  
736 AS into the western AS.

737 Removal of DFe from the water column is mainly through biological uptake in the upper 40 m. Uptake of DFe by  
738 small phytoplankton dominate biological uptake throughout the year, except during September-October when  
739 diatoms uptake of DFe becomes significant (not shown). This signature of diatoms is also observed in opal fluxes  
740 measured by sedimentary traps deployed near the western AS and has been attributed to lowering of zooplankton  
741 grazing pressures during late southwest monsoon (Smith, 2001) as well as to silicate limitation of diatoms in  
742 initially upwelled waters (Haake et al., 1993). In the subsurface layer, remineralization of sinking fluxes of  
743 particulate iron peaking at ~50 m replenishes the DFe pool during the latter part of the productive months (Fig.  
744 S16a). Iron so released is made available to the surface layer via mixing or advection, thereby playing an important  
745 role in maintaining surface DFe pool. Some of the remineralized DFe is further removed by scavenging, which  
746 peaks at ~80 m during the productive months due to large fluxes of sinking particulate organic carbon, biogenic  
747 silica, calcium carbonate and dust (Fig. S16a). Atmospheric deposition dominates biological source/sink of DFe  
748 throughout the year, while sedimentary DFe is more important for biology during northeast monsoon months.





749  
 750 **Figure 10: Evolution of the various terms of DFe budget, expressed as  $\mu\text{mol m}^{-3} \text{yr}^{-1}$ , by month and depth over the**  
 751 **western Arabian Sea. Left panels: CTRL, Middle panels: ATM and, Right panels: SED case. The contours in the upper**  
 752 **panel for CTRL show evolution of DFe concentrations (nM), while the contours in the upper panels for ATM and SED**  
 753 **cases show the percentage contribution of each of these cases to total DFe concentrations in CTRL case.**

754

### 755 3.4.2 Southern Bay of Bengal

756 The region corresponding to the southern BoB ( $7^{\circ}$ - $10^{\circ}$ N and  $82^{\circ}$ - $84^{\circ}$ E) is located to the east of Sri Lanka.  
 757 Compared to the rest of the BoB, freshwater flux from South Asian rivers reduces markedly in this region due to  
 758 advection of high salinity water from AS by the eastward flowing Southwest Monsoon Current (see Fig. 2h) as  
 759 well as upward pumping of saltier water by thermocline doming during the southwest monsoon season  
 760 (Vinayachandran et al., 2013). This leads to stronger biophysical coupling in the southern BoB, compared to the  
 761 rest of the bay, through erosion of the upper stable layer of freshwater capping. During southwest monsoon, the

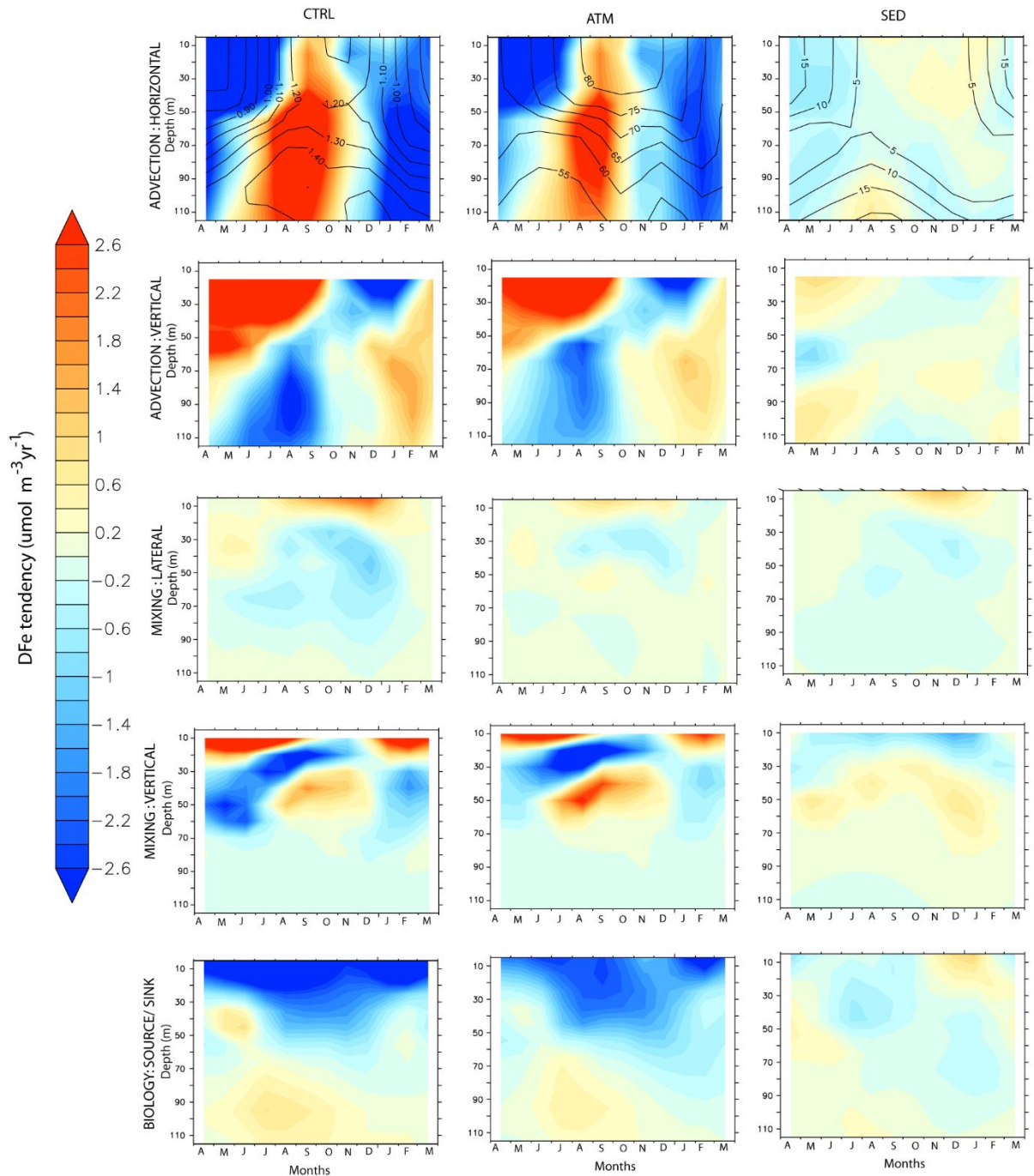
762 Southwest Monsoon Current advects nutrients and chlorophyll from the upwelling regions along the southern tip  
763 of India and Sri Lanka into the southern BoB (Vinayachandran et al., 2004). Over the open southern BoB, to the  
764 east of Sri Lanka, cyclonic wind stress curl drives open ocean upwelling leading to shoaling of the thermocline  
765 that forms the Sri Lankan dome. This results in surface chlorophyll concentration between 0.3-0.7 mg m<sup>-3</sup> and  
766 strong subsurface chlorophyll maxima between 20-50 m where chlorophyll concentration can exceed 1 mg m<sup>-3</sup>  
767 (Thushara et al., 2019). A much lower magnitude of surface chlorophyll concentration (~0.18 mg m<sup>-3</sup>, Fig. 9c)  
768 and subsurface chlorophyll maxima (~0.2 mg m<sup>-3</sup>) at 40-60 m depth is simulated by CESM. During the northeast  
769 monsoon, CESM simulates a second bloom over this region associated with winter cooling and mixed layer  
770 deepening to ~60 m (Fig. 9c). This bloom has slightly higher magnitude, peaking at ~0.25 mg m<sup>-3</sup>, compared to  
771 the southwest monsoon bloom. Surface chlorophyll data from OC-CCI also reveals the presence of northeast  
772 monsoon blooms (peak at ~0.25 mg m<sup>-3</sup>), which during some years are of higher magnitude than southwest  
773 monsoon blooms. Argo data in this region also show signatures of mixed layer deepening during winter (not  
774 shown).

775 Overall, the highest DFe over this region is encountered during the late southwest monsoon and is dominated by  
776 atmospheric deposition (Fig. 11). Vertical advection is the most important process supplying DFe to the surface  
777 layers during spring and southwest monsoon months (Fig. 11). This is aided by a positive wind stress curl  
778 established over the region from March onwards. While vertical velocity is positive during the southwest monsoon  
779 over the entire depth considered, DFe supply by vertical advection is positive only for depths less than 50 m (Fig.  
780 S13). This is because the magnitude of upward velocity gradually reduces with depth, resulting in positive values  
781 of  $U'$  upwards from 40 m depths. (Fig. S13). With the arrival of westward propagating Rossby waves to the  
782 western boundary of the BoB during October, upwelling favorable vertical motion collapses (Webber et al., 2018).

783 With respect to horizontal advection, it is seen that the magnitude and sign of convergence by the meridional  
784 component of the current mainly controls DFe supply over the southern BoB. This arises from the southward  
785 flowing current to the western flank of the Sri Lankan dome that supplies atmospheric DFe to this region. This  
786 DFe supplied by the southwards current, as well as DFe derived from upwelling, is removed by the energetic  
787 eastward currents during late spring to early fall intermonsoon months. During the rest of the year, the westward  
788 flowing currents supplies some sedimentary DFe from the Andaman Sea to the southern BoB. However, the much  
789 larger magnitude of dust deposition in the north-western BoB leads to overall negative tracer gradients and, thus,  
790 dilution of DFe by horizontal advection. The most important DFe supply mechanism during northeast monsoon  
791 is enhanced vertical mixing in the upper 20 m associated with deepening of mixed layer. Additionally,  
792 downwelling due to weakly negative wind stress curl during this time of the year removes DFe from the surface  
793 and favors its accumulation in the subsurface ocean. Lateral mixing complements DFe supply to the upper 20 m  
794 during fall and early northeast monsoon, especially from sedimentary sources.

795 Biological uptake removes DFe throughout the year from the upper 40 m especially during the southwest and  
796 northeast monsoon blooms (Fig. 11). DFe uptake in the upper 40 m is dominated by small phytoplankton during  
797 most of the year, except during northeast monsoon (not shown). Diatom DFe uptake, on the other hand, dominates  
798 the deep chlorophyll maxima present between 40-70 m throughout the year as well as within the surface layer  
799 during northeast monsoon months. Several studies have pointed to substantial nutrient uptake by diatoms in the  
800 central, coastal, and northern BoB due to riverine supply of silicates (Madhu et al., 2006; Madhupratap et al.,

801 2003). Remineralization of particulate iron as well as iron release from grazing and mortality of phytoplankton  
 802 and zooplankton have a primary peak between 50 m-80 m during July-August and secondary peak during  
 803 February-March. On the contrary, scavenging removes DFe, with its effect peaking during July-August during  
 804 blooms (Fig. S16b).



805  
 806 **Figure 11: Same as Figure 10, except over the southern Bay of Bengal.**

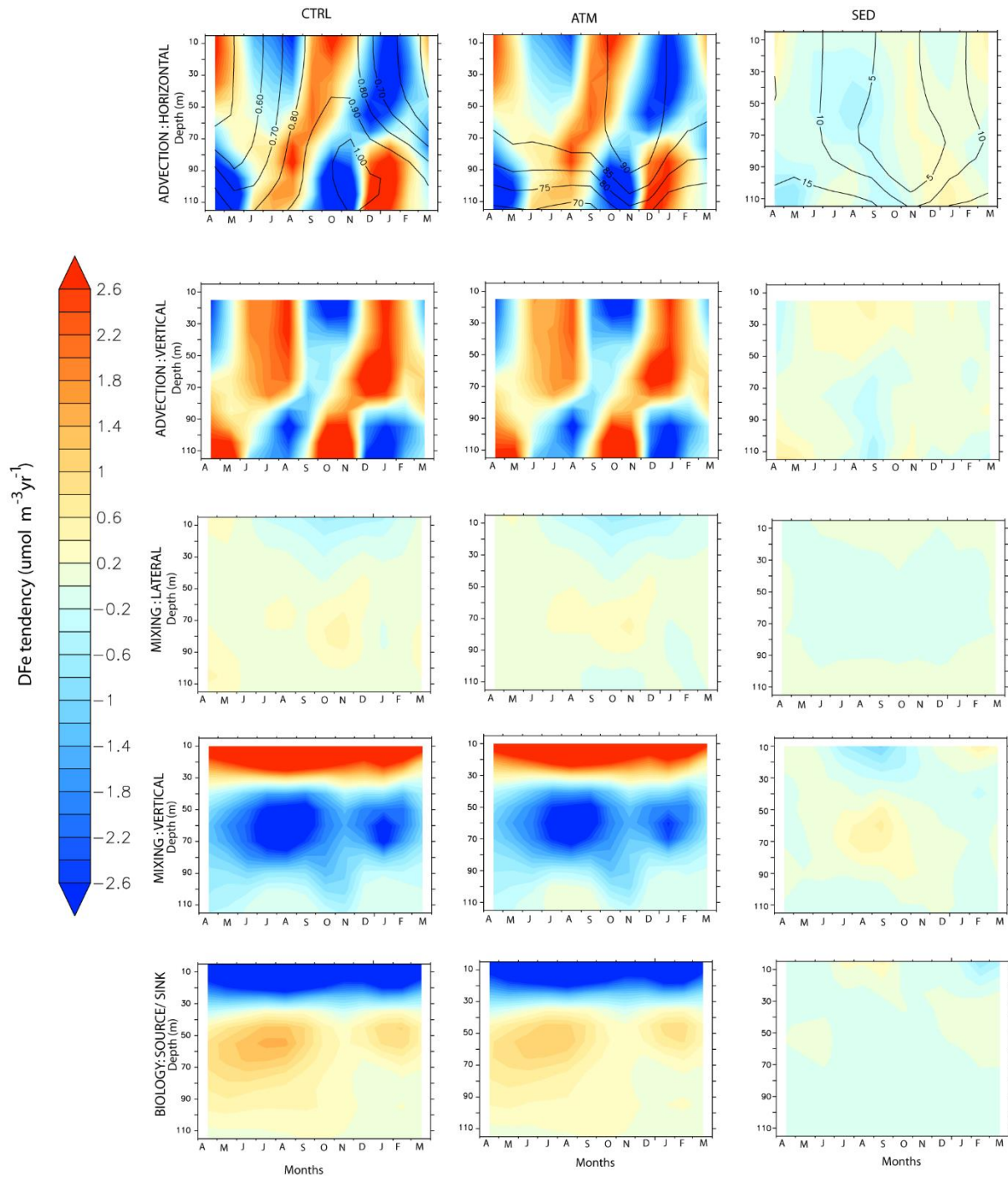
807  
 808 **3.4.3 Central Equatorial IO**

809 With chlorophyll concentrations around  $0.1 \text{ mg m}^{-3}$  for most part of the year, the central equatorial IO ( $2^{\circ}\text{S}$ - $2^{\circ}\text{N}$   
 810 and  $76^{\circ}$ - $80^{\circ}\text{E}$ ) is the least productive of all the regions considered (Fig. 9d). Unlike its counterparts in the Pacific

811 and the Atlantic Oceans, the equatorial IO experiences only transient upwelling due to changes in wind direction  
812 associated with migration of the intertropical convergence zone. This also leads to surface currents reversing their  
813 direction four times a year. Thus, the region experiences westward surface currents of weak magnitude during the  
814 southwest and northeast monsoon months and much stronger eastwards current during the spring and fall  
815 intermonsoon months (Han et al., 1999). These narrow eastwards surface currents during the intermonsoon  
816 months, known as Wyrтки jets, are in response to westerly winds (Wyrтки, 1973). The biogeochemical  
817 characteristics of the region have only been recently explored with the help of satellite and *in situ* data (e.g.,  
818 Prasanna Kumar et al., 2012; Strutton et al., 2015). Deepening of the surface layer associated with the eastward  
819 transport of water during the intermonsoon months lowers productivity (Prasanna Kumar et al., 2012).  
820 Chlorophyll concentrations, although much lower compared to the rest of the IO, peaks during October-December  
821 possibly due to wind stirring or shear instability at the base of the eastward moving Wyrтки Jet (Strutton et al.,  
822 2015). Additionally, *in situ* measurements in the central equatorial IO have revealed deep chlorophyll maxima  
823 located ~60 m depth contributing to more than 30% of the total chlorophyll biomass (Vidya et al., 2013). The  
824 peak ocean DFe concentration is encountered during August-November. Overall, comparison between CTRL,  
825 ATM and SED cases show that atmospheric deposition, peaking during July (Fig. 9d), dominates DFe contribution  
826 to the central equatorial IO, whereas sedimentary DFe plays a distant secondary role (Fig. 12).

827 Horizontal advection is the most important process of DFe supply within the mixed layer during March-May and  
828 September-November (Fig. 12). During the intervening months, vertical advection plays the predominant role in  
829 DFe supply. Decomposing the horizontal advection further into  $D\text{Fe}'$  and  $U'$  reveals that the meridional velocity  
830 convergence is the main contributor to the central equatorial IO DFe budget during March-May and September-  
831 November (Fig. S14). This originates from the westerly wind directing equatorward Ekman flow in both the  
832 hemispheres, which leads to convergence and drives eastward propagating downwelling Kelvin wave (McPhaden  
833 et al., 2015). Averaged over the upper 100 m, zonal velocity convergence, although somewhat of lower magnitude,  
834 opposes meridional velocity convergence throughout the year. When the Wyrтки jet weakens, upwelling induced  
835 by easterly wind drives upward vertical supply of DFe, whereas there is downward vertical removal of DFe during  
836 the intervening periods. This alternating between upwelling and downwelling control on DFe has an upward phase  
837 propagation. An important feature of the central equatorial IO, in contrast to other equatorial regions, is the  
838 presence of transient Equatorial Undercurrent between 60 m-200 m depth with core generally centered on the  
839 depth of the 20°C isotherm (Chen et al., 2015). The Equatorial Undercurrent appears most strongly during winter-  
840 spring months and with much weaker magnitude during summer-fall months (Chen et al., 2015; Schott &  
841 McCreary, 2001). CESM simulation reveals the signature of the upper part of the Equatorial Undercurrent in  
842 influencing DFe budget. This is characterized by the zonal velocity underneath the mixed layer (~80 m depth)  
843 showing strong eastward transport during January-April and a much weaker eastward transport during September-  
844 November. The horizontal convergence of DFe is prominent during the developing phase of the Equatorial  
845 Undercurrent (December-February and June-August), probably, associated with progressive eastward extension  
846 and strengthening of Equatorial Undercurrent from the western IO. These periods of horizontal DFe convergence  
847 are interspersed with vertical DFe convergence. Superimposed on advection, vertical mixing plays an important  
848 role in bringing subsurface DFe to the surface levels in the upper 30 m, peaking during July-August.

849 Biological removal of DFe, almost entirely by small phytoplankton, is conspicuous in the upper 40 m and peaks  
850 during September. This is in line with sediment trap studies over the central equatorial IO where peak biogenic  
851 fluxes are detected during the southwest and fall intermonsoon months and are dominated by coccolithophorids  
852 and foraminifera carbonate (Ramaswamy and Gaye, 2006). Furthermore, *in situ* water samples have shown that  
853 picoplankton, having size less than 10  $\mu\text{m}$ , consists of more than 90% of the phytoplankton biomass in central  
854 equatorial IO (Vidya et al., 2013). The period of peak biogenic flux is also characterized by peak in DFe removal  
855 by scavenging and remineralization of particulate iron released from mortality and grazing at deeper layers (Fig.  
856 S16c). A secondary increase in biological removal of DFe is noticed during January-March associated with a  
857 secondary peak in chlorophyll, although its impact is not evident in sediment trap biogenic flux data (Vidya et al.,  
858 2013). This might arise from remineralization of particulate iron being almost twice the magnitude of scavenging  
859 losses during this time of the year.



860

861 **Figure 12: Same as Figure 10, except over the central equatorial Indian Ocean.**

862

863 **3.4.4 Central Southern Tropical IO**

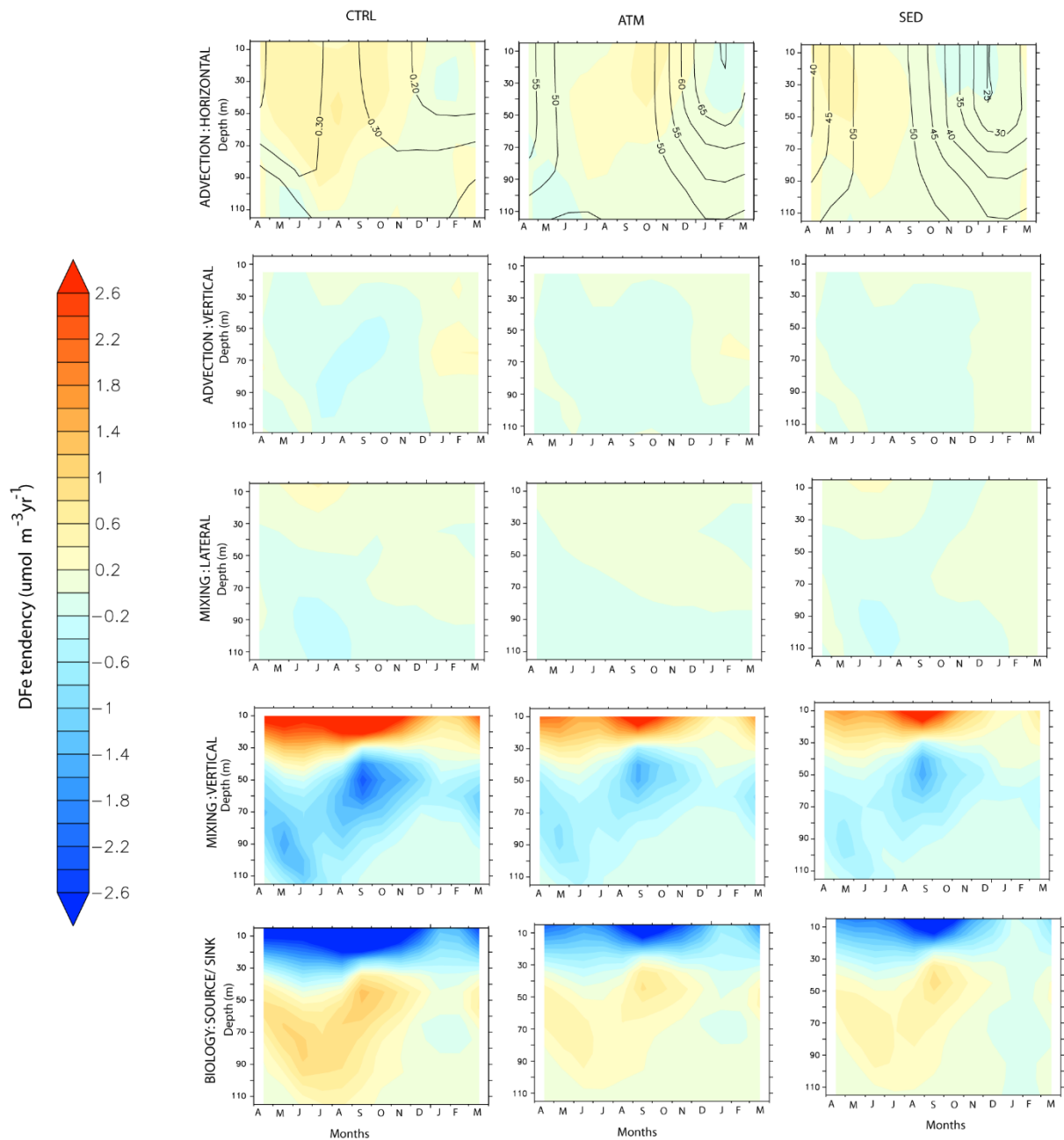
864 The central southern tropical IO (13°-17°S and 72°-76°E) is located in the transition zone between DFe-poor region  
 865 of the subtropical IO gyre and DFe-enriched northern IO. Of all the regions considered, this receives the lowest  
 866 atmospheric DFe (Fig. 9e), resulting in DFe limitation of phytoplankton growth particularly during the boreal  
 867 summer (Fig. 7). Steady southeasterly winds, prevailing throughout the year, transport dust from Australian  
 868 sources into this region. Peak in dust deposition is during austral spring and summer associated with strong source

869 activity (Kok et al., 2021; Yang et al., 2021). A secondary peak in dust deposition during austral winter is possibly  
870 associated with enhanced transport. Northern part of the central southern tropical IO lies on the Seychelles-Chagos  
871 thermocline ridge, which is characterized by doming up of the thermocline due to negative wind stress curl  
872 resulting in Ekman divergence (Vialard et al., 2009). The thermocline progressively deepens towards the sub-  
873 tropical southern IO gyre to the south as wind stress curl changes sign to positive. The westward flowing South  
874 Equatorial Current brings low salinity water and nutrients from the Indonesian region. Satellite observed enhanced  
875 chlorophyll concentration during the boreal (austral) summer (winter) months have been attributed to vertical  
876 diffusion (Košić et al., 2009; Lévy et al., 2007). Additionally, westward propagating upwelling/downwelling  
877 Rossby waves arrive in this region following La Nina/El Nino event and play a key role in modulating sea surface  
878 height and the depth of thermocline (Masumoto & Meyers, 1998; Périgaud & Delecluse, 1992). This perturbs the  
879 depth of nitracline, which has significant impact on column productivity (Kawamiya & Oschlies, 2001).

880 Both ATM and SED sources are important in this region for DFe supply, with the SED (ATM) source having  
881 higher contribution during austral winter (summer) months (Fig. 13). Analysis of CESM-simulated DFe budget  
882 reveals that vertical mixing in the upper 30 m is the most important process of DFe supply, which peaks during  
883 September. This is the time of the year when CESM records the lowest sea surface temperature resulting in mixed  
884 layer deepening. Such winter mixing leads to erosion of vertical gradient in DFe observed during the rest of the  
885 year in the upper 120 m. Horizontal advection is the next most important supplier of DFe in this region. The  
886 westward flowing South Equatorial Current is strongest during austral winter and during winter-to-summer  
887 transition months. This results in meridional velocity convergence and zonal velocity divergence resulting in a  
888 quasi-balance between DFe supply and removal (Fig. S15). Overall, horizontal advection leads to predominantly  
889 sedimentary DFe convergence during March-June and predominantly atmospheric DFe convergence during  
890 September-November.

891 The wind stress curl is mostly negative, that is upwelling favorable, throughout the year. Between April-October  
892 (austral winter), when winter convection-driven blooms are prominent, wind stress curl becomes weakly negative  
893 to slightly positive. Following this, during January-March, the wind stress curl becomes strongly negative  
894 resulting in upward velocity and favors vertical advection of both atmospheric and sedimentary DFe in equal  
895 magnitude. While vertical  $U'$  is responsible for supplying DFe in the upper 50 m, vertical  $DFe'$  is important at  
896 deeper depths (Fig. S15).

897 The biological sink of DFe peaks during the month of maximum vertical mixing, that is, during September.  
898 During this time, uptake of DFe is dominated by diatoms, which accounts for more than 80% of the total DFe  
899 uptake. Small phytoplankton dominate the rest of the year. Scavenging removal of DFe and particulate iron  
900 remineralization peaks one month later during October between 50-90 m depth range (Fig. S16d). Overall, the  
901 central southern tropical IO is the only region where atmospheric deposition and sedimentary sources of iron are  
902 equally important in driving the DFe budget.



903

904 **Figure 13: Same as Figure 10, except over the central southern tropical Indian Ocean.**

905

906 **4 Conclusions**

907

908 Using the ocean component of the Earth system model CESM version 2.1, this study elucidates the impacts of  
 909 various sources of DFe on upper ocean productivity, nutrient limitations and DFe budgets over the northern IO.  
 910 The iron cycle in CESM represents the complex interplay between several processes including DFe supply,  
 911 removal by scavenging and biological uptake, particulate iron remineralization, and organic ligand complexation.  
 912 The major sources of DFe for this region are included in this model: atmospheric deposition, sediments,  
 913 hydrothermal vents, and rivers. Although there are model biases in representing physical and biogeochemical  
 914 variables, the overall patterns of spatial and temporal variation of DFe are simulated reasonably well in CESM.



915 The study finds that atmospheric deposition is the most important source of DFe to the northern IO. Atmospheric  
916 deposition contributes well over 50% of the total DFe concentration and more than 10% (35%) to upper 100 m  
917 (surface level) chlorophyll concentrations, especially over the AS, equatorial IO, and southern tropical IO.  
918 Sedimentary sources become important along continental shelves, where they can contribute to more than 20% of  
919 total DFe. The sedimentary source has the largest impact in fueling phytoplankton blooms over the southern  
920 tropical IO during June-September. In contrast, hydrothermal and river sources have negligible impacts on upper  
921 ocean DFe pools in this region. Almost all regions that experience significant positive chlorophyll responses to  
922 atmospheric as well as sedimentary sources of DFe show a preponderance of diatoms over other phytoplankton  
923 groups. The increases in phytoplankton following external DFe addition are evoked in regions with low  
924 background DFe levels (<0.3 nM) and high initial NO<sub>3</sub>:DFe, indicating the importance of high levels of  
925 macronutrients. Following, external DFe addition, a shift to nitrate limitation of phytoplankton is observed.

926 Analysis of DFe budget across different biophysical regimes in the northern IO shows that this budget is generally  
927 dominated by atmospheric deposition, with sedimentary sources of DFe being a distant second contributor. The  
928 exception to this occurs over the southern tropical IO region, where both atmospheric and sedimentary sources  
929 become equally important. In all the regions considered, vertical mixing is the most important physical mechanism  
930 through which DFe is supplied, and furthermore this mechanism is active almost throughout the year. In contrast,  
931 the importance of horizontal and vertical advection is highly seasonal. DFe uptake by small phytoplankton in the  
932 upper ocean is the most important route through which DFe removal takes place, except in the productive waters  
933 where diatoms also participate in the removal process. At subsurface levels, competition between the removal of  
934 DFe by scavenging and remineralization of particulate iron determines the DFe pool available to the surface ocean  
935 via these aforementioned physical processes.

936 Of all DFe sources, atmospheric deposition is most likely vulnerable to future global warming, and changes to it  
937 will perhaps exert strong influence on upper ocean productivity and nutrient limitation. Additionally, 59% of the  
938 continental shelves and bathyal sea floor over the northern IO experiences hypoxic conditions (Helly and Levin,  
939 2004) and there are several lines of evidence pointing to reductions in oxygen content over this region during the  
940 last few decades due to enhanced upper ocean stratification (Schmidtko et al., 2017). This will possibly impact  
941 the flux of iron from reduced sediments. The present study thus provides foundations to explore how different  
942 future scenarios of atmospheric deposition and the extent of reducing sediments can impact biogeochemistry over  
943 the northern IO.

944

#### 945 **Code and data availability**

946 Climatology of ocean temperature, salinity and nutrients are from World Ocean Atlas 2018 available at  
947 <https://www.ncei.noaa.gov/access/world-ocean-atlas-2018/> . Monthly surface chlorophyll data from OC-CCI is  
948 obtained from <https://www.oceancolour.org/>. Monthly climatology of ocean mixed layer depth based on Holte at  
949 al. (2017) is downloaded from <http://mixedlayer.ucsd.edu/>. Surface ocean current data from OSCAR can be  
950 downloaded from: [https://podaac.jpl.nasa.gov/dataset/OSCAR\\_L4\\_OC\\_third-](https://podaac.jpl.nasa.gov/dataset/OSCAR_L4_OC_third-deg?ids=Keywords:Keywords:Projects&values=Oceans::Solid%20Earth::OSCAR&provider=PODAAC)  
951 [deg?ids=Keywords:Keywords:Projects&values=Oceans::Solid%20Earth::OSCAR&provider=PODAAC](https://podaac.jpl.nasa.gov/dataset/OSCAR_L4_OC_third-deg?ids=Keywords:Keywords:Projects&values=Oceans::Solid%20Earth::OSCAR&provider=PODAAC).

952 Dissolved iron from GEOTRACES Intermediate Data Product 2021 is available at  
953 <https://www.geotraces.org/geotraces-intermediate-data-product-2021/>. Additionally, dissolved iron profile data

954 are also obtained from Tagliabue et al. (2012) available at <https://www.bodc.ac.uk/geotraces/data/historical/>. The  
955 code for CESM2.1 can be downloaded from [https://www.cesm.ucar.edu/models/cesm2/release\\_download.html](https://www.cesm.ucar.edu/models/cesm2/release_download.html)  
956 (last access: 01 December 2020).

#### 957 **Author contributions**

958 PB conceived the study, carried out model simulations, analysed the data and wrote the manuscript.

#### 959 **Competing interests**

960 The author declares that there is no conflict of interest.

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