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RESEARCH ARTICLE

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Examining the Interaction Between Free-Living Bacteria and Iron in the Global Ocean

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Key Points:

- Iron emerges as a proximal limiting factor for the growth of free-living (FL) bacteria in low iron regions, with dissolved organic carbon being the ultimate limiting resource
- While FL bacteria consume a significant amount of iron in the surface ocean and affect phytoplankton dynamics, their impact on the subsurface iron cycling is small
- A more complete understanding of the role of bacteria requires holistic consideration of the role of different bacterial groups and their specific iron requirements via expanded observations of these key groups

Supporting Information:

Supporting Information may be found in the online version of this article.

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Abstract Marine free-living (FL) bacteria play a key role in the cycling of essential biogeochemical elements, including iron (Fe), during their uptake, transformation and release of organic matter throughout the water column. Similar to phytoplankton, the growth of FL bacteria is regulated by nutritive resources such as Fe, and the low availability of these resources may influence bacterial interactions with phytoplankton, causing knock-on effects for biogeochemical cycling. Yet, knowledge of the factors limiting the growth of FL bacteria and their role within the Fe cycle is poorly constrained. Here, we explicitly represent FL, carbon-oxidizing bacteria in a three-dimensional global ocean biogeochemistry model to address these questions. We find that although Fe can emerge as proximally limiting in the tropical Pacific and in high-latitude regions during summer, the growth of FL bacteria is ultimately controlled by the availability of labile dissolved organic carbon over most of the world's oceans. In Fe-limited regions, FL bacterial biomass is sensitive to their Fe uptake capability in seasonally Fe-limitation regions and to their minimum Fe requirements in regions perennially low in Fe. Fe consumption by FL bacteria is significant in the upper ocean in our model, and their competition with phytoplankton for Fe affects phytoplankton growth dynamics and can make bacteria become more carbon limited. The impact of FL bacteria on the Fe distribution in the ocean interior is small due to a tight coupling between Fe uptake and release. Moving forward, future work that considers other bacteria groups and different bacterial metabolisms is needed to explore the broader role of bacteria in ocean Fe cycling. In this context, the global growing^o omics data from ocean observing programs can play a crucial role.

Plain Language Summary Marine heterotrophic bacteria, a ubiquitous group of microorganisms, can control the cycling of essential biogeochemical elements in the ocean, including the micronutrient iron. However, factors limiting the growth of bacteria and the impact of bacteria on ocean iron distributions are still poorly understood. In this paper, we represent the free-living (FL) bacteria in an ocean biogeochemistry model to show that the availability of labile dissolved organic carbon is the ultimate limiting factor for the growth of FL bacteria, while iron can be a co-limiting factor in iron-limited regions. We also suggest that the competition between phytoplankton and FL bacteria for iron can significantly alter both phytoplankton blooms and bacterial growth. Future studies should exploit new genomics information to further explore the roles of other bacteria groups.

1. Introduction

Heterotrophic bacteria (hereafter bacteria) account for a significant portion of the ocean carbon biomass (Buitenhuis et al., 2012) and play various roles in oceanic nutrient cycles. Despite the wide variety of bacteria that live under various conditions (Sogin et al., 2006; Zakem et al., 2020), free-living (FL), carbon-oxidizing bacteria have been the focus of previous studies due to their control on the flow of carbon and bio-essential elements through the marine food web (Azam & Malfatti, 2007; del Giorgio & Duarte, 2002; Jiao et al., 2010). Carbon-oxidizing bacteria either transforms a fraction of the organic matter fixed by phytoplankton into a highly refractory form, thus keeping carbon away from the atmosphere for several millennia or respire the fixed organic matter to release inorganic carbon and nutrients back to the seawater, thus sustaining oceanic primary production (Jiao et al., 2010, 2014). Other bacterial groups, such as particle-attached, nitrogen-oxidizing, and sulfur-oxidizing bacteria and archaea, carry out various chemical transformations, which modulate the ocean nitrogen and sulfur cycles and produce potent greenhouse gas (nitrous oxide and methane) (Bianchi et al., 2018; Kallmeyer et al., 2012; Saito et al., 2020; Zakem et al., 2020). Given the importance of bacteria in various biogeochemical processes, quantifying the constraints of bacterial growth and production and their implications on oceanic

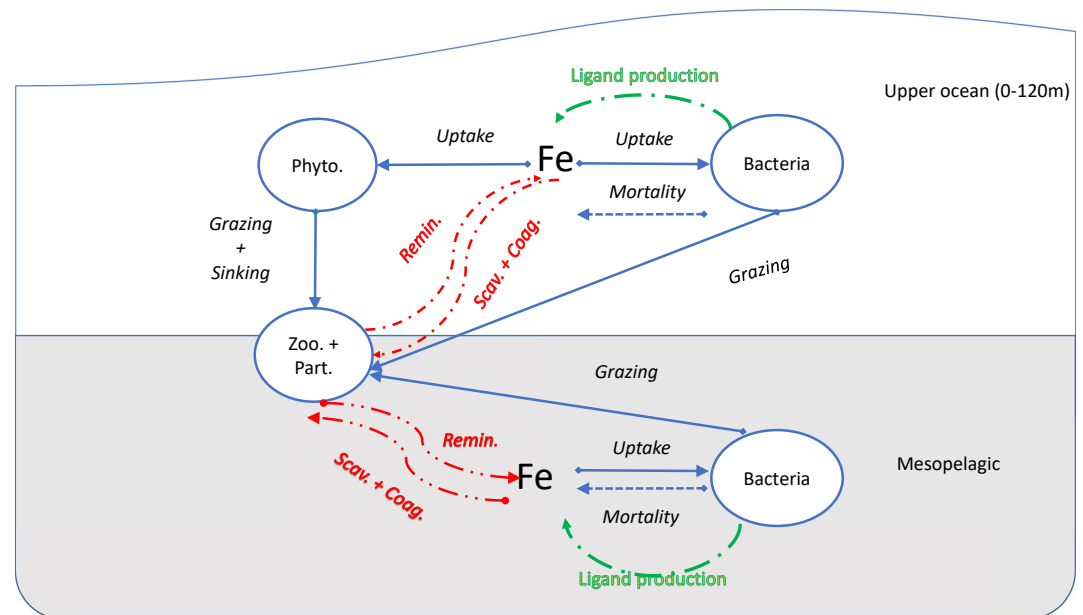


Figure 1. A simple schematic representation of the role of free-living (FL) bacteria in the oceanic Fe cycle in the upper ocean (0–120 m) and mesopelagic zones, as represented in NEMO-PISCES. FL bacteria consume and release Fe throughout the water column through their growth and mortality. In addition, FL bacteria are grazed by zooplankton, thereby potentially changing the Fe content of zooplankton and organic particles. Finally, FL bacteria produce siderophores, which can form ligand complexes with Fe and protect Fe from being removed by scavenging. It should be noted that in this current version of PISCES, the production of siderophores by FL bacteria is not yet included since it requires the representation of ligands as a prognostic tracer. However, this PISCES version assumes a constant concentration for ligands of about 1 nM, which is the mean dissolved Fe concentration in the deep ocean. The impact of siderophores production by FL bacteria on ocean Fe cycling will be the goal of a future study. Many processes induced by other bacteria groups are also not yet considered.

productivity is crucial. In this study, we will focus on the FL, carbon-oxidizing bacteria as they are the most abundant type of bacteria in the open ocean (Giovannoni, 2017).

In the ocean, FL bacteria require a suite of resources for their growth, and similar to phytoplankton, their growth can be limited by the resource that is in shortest supply (Church et al., 2000; Obernosterer et al., 2015). For example, the growth of FL bacteria in several oceanic regions is limited by labile dissolved organic carbon (LDOC), which is supplied to the ocean via various processes such as phytoplankton exudation, zooplankton excretion, viral releases, and releases from sediments, sea ice, and ice sheets (Fourquez et al., 2014; Lawson et al., 2014; Pakulski et al., 1996). In other regions such as the California upwelling and the Southern Ocean, where the micronutrient iron (Fe) has been shown to limit phytoplankton growth (Boyd et al., 2000; Bundy et al., 2016; King & Barbeau, 2007), the growth of FL bacteria may also be limited by Fe (Church et al., 2000; Ducklow & Kirchman, 2000; Obernosterer et al., 2015). Despite low dissolved Fe (dFe) concentrations, several field studies observed greater cellular Fe content and a greater Fe demand in FL bacteria compared to phytoplankton (Fourquez et al., 2015, 2020; Tortell et al., 1996, 1999). This suggests that FL bacteria may be a significant competitor with phytoplankton for Fe in the upper ocean, especially where dFe concentrations are potentially limiting. Field-based evidence of resource competition between bacteria and phytoplankton comes from the subtropical waters east of New Zealand, where the magnitude and longevity of the diatom blooms were controlled by the competition for Fe between diatoms and bacterial communities (Boyd et al., 2012). A recent theoretical modeling study also argued that FL bacteria can be a significant consumer of dFe, and that when enough labile DOC is present, FL bacteria can outcompete phytoplankton for Fe, leading to a decrease in phytoplankton biomass (Ratnarajah et al., 2021).

As FL bacteria are not light dependent, they can influence the distribution of dFe throughout the water column (Ratnarajah et al., 2021). First, FL bacteria consume and release Fe through their growth and mortality (Figure 1). Under Fe-limited conditions, FL bacteria are known to produce siderophores to keep Fe in dissolved form and to assist their Fe uptake, thereby impacting the retention and removal of dFe (Amin et al., 2009; Boiteau et al., 2016;

Bundy et al., 2018; Vraspir & Butler, 2009). As FL bacteria can be grazed upon by microzooplankton, their Fe content can modify the zooplankton recycling efficiency of Fe and the Fe remineralization length scale (e.g., Richon et al., 2020). Taken together, FL bacteria can play a significant role in modulating oceanic Fe cycling and by consequence, phytoplankton growth dynamics (Figure 1).

Despite increasing evidence on the importance of FL bacteria in modulating oceanic Fe cycling, our understanding of the interactions between FL bacteria and Fe is still very limited. For instance, it is still unclear if Fe is the ultimate factor limiting the growth of FL bacteria in the ocean, especially when both labile DOC and Fe concentrations are low (Church et al., 2000; Obernosterer et al., 2015; Ratnarajah et al., 2021). In addition, the limited existing ocean measurements have reported a wide range of values for the Fe contents of FL bacteria, resulting in a large uncertainty in our knowledge of the FL bacterial Fe demand, storage, and uptake capability (Fourquez et al., 2015; Mazzotta et al., 2020; Tortell et al., 1999). In this context, the ocean biogeochemistry models that integrate marine ecosystem dynamics into the global ocean circulation can be useful tools to fill these knowledge gaps. However, only very few model studies performed at the global scale include an explicit representation of FL bacteria. Of all the ocean biogeochemistry models that are ocean components of the Earth System Models participating in the CMIP5 and CMIP6 projects (Séférián et al., 2020; Taylor et al., 2011), only two models that explicitly represent FL bacteria as a prognostic tracer (Dunne et al., 2020; Epicoco et al., 2016; Stock et al., 2020). Even in models that represent FL bacteria, the impact of FL bacteria-Fe interaction on the global ocean Fe cycle and biogeochemistry has not yet been systematically examined (Epicoco et al., 2016; Tagliabue et al., 2016; Vichi & Masina, 2009).

In this study, we explicitly represent the biomass and Fe quotas of FL bacteria in a global ocean biogeochemistry model to examine the interaction between FL bacteria and Fe, and how this shapes phytoplankton bloom dynamics. Specifically, we explore (a) how the availability of resources (Fe and DOC) regulates the growth of FL bacteria and (b) how FL bacteria influences Fe cycling, phytoplankton dynamics, and carbon export within the upper ocean (0–500 m). It should be noted that we do not consider the impact of bacterial ligand production, nor the role of other bacterial types (e.g., particle-attached, or nitrifying bacteria) (Bressac et al., 2019; Saito et al., 2020), yet, however these topics can be the focus of future studies.

2. Model Configuration and Experimental Design

2.1. The Biogeochemical Global Ocean Model

In this study, we use the biogeochemical model PISCESv2 (Aumont et al., 2015), which is a component of the ocean modeling platform Nucleus for European Modeling of the Ocean (NEMO) (Madec, 2008). The dynamic state is simulated using the ORCA2-LIM configuration of NEMO in version 8.2 (Madec, 2008). The ORCA2 domain configuration has a horizontal resolution of approximately 2° in the extratropics, with the meridional resolution increasing to 0.5° at the equator. It includes 30 vertical levels, with an increased vertical thickness from 10 m at the surface to 500 m at 5,000 m. Representation of the topography is based on the partial-step formalism (Barnier et al., 2006). The dynamic fields used to drive the ocean are identical to those used in Aumont et al. (2015). Lateral mixing is oriented along isopycnal surfaces, and the parameterization of Gent and McWilliams (1990) for sub-grid-scale processes is used poleward of 10° latitude in both hemispheres. The turbulent kinetic energy scheme of Gaspar et al. (1990) as modified by Madec (2008) is used to represent vertical mixing.

The marine biogeochemical model PISCES-v2 (Aumont et al., 2015) has been employed in many other studies exploring ocean Fe cycling (Richon et al., 2020; Tagliabue et al., 2020), as well as large-scale ocean biogeochemistry (Aumont et al., 2018). In its standard configuration, PISCES-v2 simulates the sources and sinks of 24 prognostic variables (Figure 2). These variables represent the lower trophic levels of the marine ecosystem, including four limiting nutrients (Fe, phosphate: PO_4^{3-} , silica: $\text{Si}(\text{OH})_4$, and nitrogen [nitrate: NO_3 and ammonium: NH_4]), as well as two phytoplankton groups (nanophytoplankton and diatoms) and two zooplankton size classes (micro- and meso-zooplankton). Phytoplankton growth is controlled by light and temperature in addition to the limiting nutrients. There are three nonliving compartments in PISCES corresponding to semi-labile dissolved organic matter and two size classes of particulate organic carbon (POC), which differ by their size and their sinking speed. It should be noted that PO_4 , NO_3 , and NH_4 are linked to each other by constant and identical Redfield ratios proposed by Takahashi et al. (1985) (C/N/P: 122:16:1) in all modeled organic compartments. In contrast,

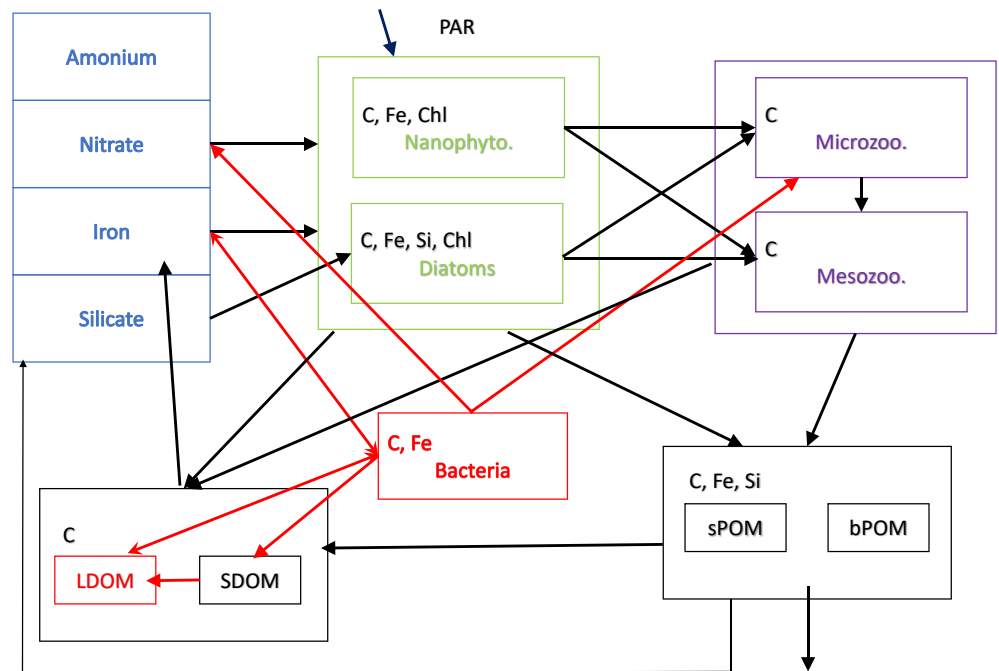


Figure 2. Architecture of PISCES v-2. This figure is adapted from Aumont et al. (2015), showing only the ecosystem model and omitting the oxygen and carbonate systems. The elements that are explicitly modeled are indicated in the left corner of each box. PO_4 , NO_3 , and NH_4 are linked by constant and identical Redfield ratios (C/N/P: 122:16:1) in all modeled organic compartments. The only processes that decouple phosphate distribution from inorganic N distribution are nitrogen fixation, denitrification and the external supplies of nutrients (atmospheric deposition, river discharge). New prognostic variables and processes added in this study are highlighted in red.

the Fe and Chlorophyll (Chl) contents in both phytoplankton groups and Si content in diatoms are prognostically simulated in the model. For both zooplankton groups, only the total carbon biomass is a prognostic variable.

In PISCES v2 (Aumont et al., 2015), dFe is consumed (uptake) by phytoplankton (diatoms and nanophytoplankton) and bacteria, recycled by zooplankton, released from the remineralization of sinking particles, and removed from water column by scavenging and colloidal pumping. Once inside the phytoplankton and bacteria cells, Fe can be transferred up the food chain via zooplankton grazing or routed to sinking particles via phytoplankton and bacteria mortality. Fe is released back to sea water by bacteria egestion, zooplankton excretion, and remineralization/solubilization of particles. External sources of dFe includes: atmospheric dust deposition, rivers, sea ice, sediment mobilization and hydrothermal vents. Fe chemistry is represented using the simple scheme based on one uniformly constant ligand and one inorganic species. Thus, it should be noted that in this current version of PISCES, the production of siderophores by FL bacteria is not yet include since it requires the representation of ligands as a prognostic tracer. The impact of siderophores production by FL bacteria on ocean Fe cycling will be the goal of a future study. Further details on ocean Fe cycling in PISCES-v2 and its ocean distribution are described in Aumont et al. (2015).

In this study, we have added to PISCES v2 three more prognostic tracers, which correspond to the bacteria carbon biomass, their Fe content, and labile dissolved organic carbon (Figure 2). These new tracers are described in detail below.

2.2. Modeling the Biomass and Iron Content of FL Bacteria

2.2.1. FL Bacteria Biomass

Building on the standard configuration of PISCES-v2 (Aumont et al., 2015), here we explicitly model the biomass of FL bacteria. The FL bacterial biomass (mol C/L) at each model grid point and at each time step depends on FL bacterial growth, mortality, aggregation, and grazing (g^Z) by microzooplankton (Z).

$$\frac{\delta B}{\delta t} = (1 - \delta^B - \sigma^B) \mu^B B - m^B \frac{B}{K_m + B} B - sh \times w^B B^2 - g^Z(B)Z \quad (1)$$

In Equation 1, the first term on the right-hand side represents FL bacterial growth, which is a function of the achieved growth rate μ^B . In turn, μ^B is a function of temperature and the concentrations of LDOC and inorganic nutrients:

$$\mu^B = \mu_{\max}^0 f_B(T) L_{\lim}^{Bact} \quad (2)$$

$$L_{\lim}^{Bact} = \min(L_N^{Bact}, L_{PO4}^{Bact}, L_{Fe}^{Bact}, L_{LDOC}^{Bact}) \quad (3)$$

While the limitation term of NO_3^- , PO_4^{3-} , and LDOC for FL bacteria is modeled using the Monod formalism ($L_i = \frac{I}{K+I}$), the Fe limitation term follows the quota formalism ($\text{Min}\left(1, \frac{\theta^{Fe,B} - \theta_{\min}^{Fe,B}}{\theta_{opt}^{Fe,B}}\right)$). In PISCES-v2, DON, DOC and DOP are not separately modeled but are linked by a fixed (Redfield) stoichiometry. Thus, when FL bacteria consume DOC, they also consume DON and DOP at the exact ratio they require to grow. According to that assumption, they do not need to supplement their needs with dissolved inorganic N and P. However, in the ocean, stoichiometric ratios in DOM are highly variable and in oligotrophic areas, C/P and C/N ratios are observed to be much higher than those required by FL bacteria (Aminot & K erouel, 2004; C. S. Hopkinson & Vallino, 2005). As a consequence, FL bacteria should take up inorganic N and P from seawater and their growth can be limited by the N and P availability. This potential limitation of our model is accounted for in Equation 3. Indeed, this formulation is similar to the one that is used in PISCES-v2 to model the degradation of semi-labile DOM (Aumont et al., 2015). We do not explicitly represent DOfFe. In PISCES, it is included in the dFe pool, which represents all the dissolved forms of Fe (truly dissolved Fe, colloidal organic and inorganic Fe, etc.)

FL bacteria are supposed to perform luxury uptake of Fe as in Buitenhuis and Geider (2010). The rate of synthesis by the cell of new FL bacterial biomass is controlled by the difference between the actual Fe quota ($\theta^{Fe,B}$) in FL bacteria and their minimum Fe quota requirement, ($\theta_{\min}^{Fe,B}$) divided by their optimum Fe quota value ($\theta_{opt}^{Fe,B}$). We set this minimum Fe quota to a low value of $1e - 6$ mol Fe/mol C since previous studies measured very low Fe/C ratios of that magnitude in FL bacteria when they are strongly limited by Fe (Fourquez et al., 2020; Tortell et al., 1996, 1999). The optimal Fe quota for FL bacteria is set to $36e - 6$ mol Fe/mol C (Fourquez et al., 2020). During the growth process, a fraction of the assimilated elements is egested (δ^B) and respired/excreted (σ^B). The temperature dependence of FL bacterial growth (Q_{10}) is set equal to that of phytoplankton (1.9) (Eppley, 1972).

The second and third terms on the right-hand side of Equation 1 represent FL bacterial mortality and aggregation, respectively. As for phytoplankton, FL bacterial mortality is modulated by a hyperbolic function of the biomass B to avoid extinction, and the FL bacterial aggregation rate (w^B) is multiplied by the turbulent shear rate (sh), following the approach in Aumont et al. (2015), as the main driver of aggregation is the local turbulence. The last term in Equation 1 represents bacteria consumption by microzooplankton, which is a function of bacteria (B) and microzooplankton biomasses (Z) and the microzooplankton grazing rate (g^Z). As described in Aumont et al. (2015), g^Z depends on temperature according to a typical exponential relationship similar to what is used for phytoplankton (Equation 25 in Aumont et al. [2015]).

2.2.2. Iron Quota in FL Bacteria

In PISCES, Fe in FL bacteria, which sets the bacterial Fe/C quota, is modeled according to the classical quota approach. Similar to the formulation adopted by Aumont et al. (2015) for the Fe quota in phytoplankton, the Fe biomass of FL bacteria (B^{Fe}) is considered rather than the FL bacterial Fe quota ($\theta^{Fe,B}$) directly. The temporal evolution of the Fe biomass of FL bacteria is modeled as follows:

$$\frac{\delta B^{Fe}}{\delta t} = (1 - \delta^B - \sigma^B) \mu^{B^{Fe}} B - m^B \frac{B}{K_m + B} B^{Fe} - sh \times w^B B * B^{Fe} - \theta^{Fe,B} g^Z(B)Z \quad (4)$$

where the FL bacterial Fe uptake ($\mu^{B^{Fe}}$) is defined as:

$$\mu^{BFe} = \theta_{\max}^{Fe,B} \mu_{\max}^0 f_B(T) L_{\lim,1}^{BFe} L_{\lim,2}^{BFe} \frac{1 - \frac{\theta_{\max}^{Fe,B}}{\theta_{\max}^{Fe,B}}}{1.05 - \frac{\theta_{\max}^{Fe,B}}{\theta_{\max}^{Fe,B}}} \quad (5)$$

Following the approach used for phytoplankton, FL bacterial Fe uptake is down-regulated at high quotas via a feedback using a normalized inverse hyperbolic function with a small shape factor set to 0.05 (Flynn & Hipkin, 1999). $L_{\lim,1}^{BFe}$ is the Fe limitation term and is parameterized as follows:

$$L_{\lim,1}^{BFe} = \frac{bFe}{bFe + K_{Fe}^{BFe}} \quad (6)$$

where bFe is bioavailable Fe, which is assumed to be equal to the dFe concentration, and K_{Fe}^{BFe} is the half-saturation constant for Fe uptake. Also as in phytoplankton, Fe surge uptake in FL bacteria can occur at very low Fe concentrations ($L_{\lim,2}^{BFe}$) as described in details in Equation 19 in Aumont et al. (2015). The modeled FL bacterial Fe quota in PISCES varies between the minimum quota and a maximum Fe quota $\theta_{\max}^{Fe,B}$, which is set to $80e - 6$ molFe/molC (Mazzotta et al., 2020).

2.3. DOC

An additional DOC pool is introduced in this version of PISCES, which corresponds LDOC. While LDOC has a very short lifetime (<few days) in the ocean because it is very efficiently consumed by FL bacteria, semi-labile DOC (SDOC) has to be hydrolyzed before being taken up by FL bacteria. It therefore has a longer lifetime (weeks to years). Both SDOC and LDOC are produced by the release of carbon from zooplankton (M for mesozooplankton and Z for microzooplankton), phytoplankton (P for nanophytoplankton and D for diatoms), and FL bacteria (B) and by the solubilization of POC. LDOC is also produced by the hydrolysis of semi-labile DOC and FL bacterial mortality and is consumed by FL bacteria. The full system of equations describing the sources and sinks of LDOC and SDOC are provided in Supporting Information S1.

Values of the parameters newly introduced in this version of PISCES are listed in Table 1. Since FL bacterial biomass and Fe quota are now explicitly represented, some other model parameters in Aumont et al. (2015) have been adjusted to keep the model biogeochemistry fields realistic and consistent with available observations. These parameters are listed in Table S1 of Supporting Information S1.

2.4. Model Experiments

The main objective of this study is to explore the factors that control FL bacterial growth and the role played by FL bacteria in the Fe cycle, phytoplankton dynamics, and carbon export. To this end, we designed a suite of sensitivity experiments in which the values of some bacteria-specific parameters are altered:

1. “Control” run: the standard model simulation
2. “10Femin” run: the minimum Fe requirement of bacteria ($\theta_{\min}^{Fe,B}$) is increased ten-fold to 10^{-5} molFe/molC
3. “3HalfK” run: The half-saturation constant for bacterial Fe uptake K_{Fe}^{BFe} is increased from 0.7 to 2.0 nmolFe/L
4. “LOWHALFK” run: The half-saturation constant for bacterial Fe uptake K_{Fe}^{BFe} is decreased three times from 0.7 to 0.2 nmolFe/L
5. “3DOC” run: The half-saturation constant for limitation of bacterial growth by LDOC $KLDOC$ is decreased from 2.0 to 0.6 $\mu\text{molC/L}$, which makes bacterial growth less sensitive to LDOC
6. “80FeC” run: the Fe quota of bacteria is set to a constant value of $80e - 6$ molFe/molC
7. “10FeC” run: the Fe quota of bacteria is set to a constant value of $10e - 6$ molFe/molC

It should be noted that in the constant quota experiments (80FeC and 10FeC), we still use the quota formalism as in the Control run to determine the Fe limitation of the growth of FL bacteria (Equations 1–4). The difference between these two simulations and the Control run is that all the Fe fluxes into and out of FL bacteria (uptake, mortality, and grazing by microzooplankton) are calculated by multiplying the carbon fluxes by a constant ratio ($10e - 6$ or $80e - 6$ molFe/molC). Through these model sensitivity experiments, we first explore how FL bacterial growth is affected by the availability in LDOC and Fe. This is achieved by changing the values of the

Table 1
Model Parameters for Bacteria With Their Values

Parameter	Unit	Value	Description
δ^B	–	0.1	Exudation of DOC
σ^B	–	0.6	Respired fraction
μ_{\max}^0	d^{-1}	0.6	Growth rate at 0°C
m^B	d^{-1}	0.01	phytoplankton mortality rate
w^B	$d^{-1} \text{ molC}^{-1}$	0.01	Minimum quadratic mortality of bacteria
$K_{Fe}^{B^{Fe}}$	nmolFe/L	0.7	Half-saturation for bacterial Fe uptake
$KLDOC$	$\mu\text{molC/L}$	2	Half-saturation constant for LDOC on bacterial growth
$\theta_{\min}^{Fe,B}$	$\mu\text{molFe/molC}$	1	Minimum Fe quota in bacteria
$\theta_{\max}^{Fe,B}$	$\mu\text{molFe/molC}$	80	Maximum Fe quota in bacteria
$\theta_{opt}^{Fe,B}$	$\mu\text{molFe/molC}$	36	Optimum Fe quota in bacteria
λ_Z		1	Fraction of DOC released as LDOC from microzooplankton
λ_M		1	Fraction of DOC released as LDOC from mesozooplankton
λ_D		0.9	Fraction of DOC released as LDOC from diatoms
λ_P		0.9	Fraction of DOC released as LDOC from nanophytoplankton
λ_B		0	Fraction of DOC released as LDOC from bacteria
λ_{PO}		0.9	Fraction of DOC released as LDOC from particulate organic carbon

half-saturation constants of LDOC and Fe uptake, as well as the minimum Fe quota of FL bacteria. Second, we examine the role of FL bacteria in the upper ocean Fe cycle by assessing how the Fe uptake and release fluxes induced by the FL bacterial activity change as a function of FL bacterial Fe quotas and its consequences on phytoplankton growth dynamics.

Each model experiment is run separately for 100 years using the same physical forcing as in Aumont et al. (2015) and is initialized from the quasi-steady state of the model simulation presented in Aumont et al. (2015). Results from the final year (annual average) of the model sensitivity runs are analyzed by comparing differences in the biogeochemistry fields in relation to the final-year (annual average) of the *Control* run.

3. Results and Discussion

3.1. Model Evaluation

A thorough evaluation of PISCES v2 is not the main objective of this study since it has been done before against observations of biogeochemical fields and features, including dFe measurements from GEOTRACES program (Aumont et al., 2015). In addition, PISCES v2 has been used to study the large-scale ocean Fe cycling and biogeochemistry in many modeling works (Aumont et al., 2015, 2017, 2018; Richon et al., 2020; Tagliabue et al., 2020). Fe distributions in this version of PISCES are also similar to those simulated in PISCES-v2. Thus, we briefly compare the model dFe concentrations against available observations and restrict the evaluation here to newly added biogeochemical fields that are most relevant to FL bacteria (DOC, FL bacteria biomass, FL bacterial Fe content).

3.1.1. Fe, DOC, and FL Bacteria Biomass and Production

First we compare the model dFe distributions at the surface, 100, and 200 m against available observations compiled by Tagliabue et al. (2012) and from GEOTRACES program in Figure 4c and 4d (Schlitzer et al., 2018). The comparison shows that this version of PISCES v2 captures the large-scale patterns and magnitudes of the observed ocean dFe concentrations. At the surface, while dFe concentration is high (~1 nM) in the tropical Atlantic, Indian Ocean and in coastal regions, it is low (~0.2 nM) in the open ocean of the tropical and North Pacific Ocean (Figures 4c and 4d). At the subsurface ocean (100 and 200 m), both model and observed dFe concentrations are higher than at the surface except for some regions in the tropical Atlantic and northern Indian

Oceans where high dust deposition leads to a higher surface dFe concentrations (Figure S2 in Supporting Information S1). Modeled subsurface dFe concentrations appear to be more uniform than that observed, due to the use of a constant ligand concentration of 0.7 nM in PISCES. Further statistical evaluation of model dFe distribution in PISCES v2 against available observations is described in Aumont et al. (2015) and Tagliabue et al. (2016).

Second, we compare the modeled DOC distribution against available observations (Hansell, 2013; Letscher et al., 2015). In PISCES, the refractory DOC component is not explicitly modeled, and should therefore be added to LDOC and SDOC pools to form the total DOC component of the model. Observations showed that this refractory DOC (RDOC) pool has an approximately uniform concentration of about 40 mmol C/m³ in the ocean (Hansell, 2013; Letscher et al., 2015). The LDOC component has a short lifetime (<a few days) in the ocean and represents a very small fraction of the total surface DOC pool (Carlson et al., 2009). Our model underestimates DOC (LDOC + SDOC + RDOC) concentrations in the tropical and subtropical regions by 10–20 mmol C/m³ (Figure 4). Our model underestimation of DOC concentrations in these regions is most likely caused by model biases in the DOC production in DOC production and/or DOC consumption. In particular, it has been shown that the N and/or P limitation in the subtropical gyres can enhance DOC production by phytoplankton (Moreno & Martiny, 2018; Zakem & Levine, 2019). In the Arctic Ocean, observations (Bussmann & Kattner, 2000; Engel et al., 2019; Fu et al., 2020; Hansell et al., 2004) also suggest that our model underestimates the surface Arctic DOC concentrations by around 30 mmol C/m (Figure 4). This is not surprising as previous studies (Connolly et al., 2020) have shown a strong influence of the relatively refractory terrestrial DOC supplied to the Arctic Ocean by rivers, a component of DOC that is not represented in our model.

We then evaluate the model (*Control* run) ability to reproduce major features of the observed ocean FL bacteria biomass (Buitenhuis et al., 2012; Xie et al., 2021) (Figure 3). Observations of FL bacteria biomass are still too sparse and limited to allow a thorough comparison of the large-scale distribution of FL bacteria biomass, even on an annual-mean basis (Figures 3a and 3b). However, observations point toward high surface FL bacteria biomass in coastal regions, in the equatorial Pacific, and in the North Atlantic and low biomass levels in the subtropical gyres, which are captured by our model (Figure 3). In general, the modeled surface distribution of FL bacteria tends to follow the large-scale distribution of chlorophyll (as a broad proxy for autotrophic biomass), since a large fraction of LDOC (Figure S1 in Supporting Information S1), which is required by FL bacteria to grow, is produced by phytoplankton and microzooplankton. When the modeled vertical distributions of FL bacteria are averaged over the global ocean and over the tropical domain (20°N–20°S) is compared with the observed vertical profiles (Figures 3c and 3d), we see that our model reproduces the vertical pattern of FL bacteria biomass within the range suggested by observations. In the upper ocean, observations indicate that our model underestimates the observed bacteria biomass. However, it should be noted that bacteria data from Buitenhuis et al. (2012) and Xie et al. (2021) include all heterotrophic bacteria types that could be the host for viruses, which could be one of the potential reasons for our model underestimation.

The modeled annual mean FL bacterial production (BP) is compared to the net primary production (NPP) in (Figure 5). At the global scale, the globally integrated BP is 8.2 PgC/year, which is about 30% of the global value of 32.2 PgC/year for NPP. The general spatial patterns of surface BP are broadly similar to those of NPP, with high values in productive coastal areas and in the eastern tropical Pacific and low values in the oligotrophic gyres and in the Southern Ocean. In general, the surface modeled BP is ~10%–20% of the surface modeled NPP, which is within the range suggested by several field observations (Ducklow & Kirchman, 2000; Ducklow et al., 2002; Kirchman et al., 1995, 2009; Lochte et al., 1997) (Figure 5b). In the low-latitude, oligotrophic areas, the surface modeled BP/NPP ratio can be higher and close to ~30%, especially in the coastal regions, which have been suggested by some studies in the Indian Ocean (Kirchman et al., 2009; Visser et al., 2015) (Figure 5b). In contrast, the BP/NPP ratio can be lower than 0.1 in the Southern Ocean, where water is colder and FL bacteria is limited by the availability of DOC (Kirchman et al., 2009; Teira et al., 2012).

3.2. Resources Limiting the Growth of FL Bacteria

Figure 6 shows the spatial patterns of the limitation terms for the two major resources limiting FL bacterial growth, that is, Fe and LDOC, on a seasonal basis (cf., Equations 2 and 3). We define the most limiting nutrient as the nutrient that produces the lowest value of the FL bacterial limitation factor (see Equation 3). Co-limitation can occur when Fe and LDOC have similar low values for this metric. Thus, we see that over most of the ocean (>90%) and for the two seasons, LDOC limits FL BP and as such, is the main limiting resource (Figure 6).

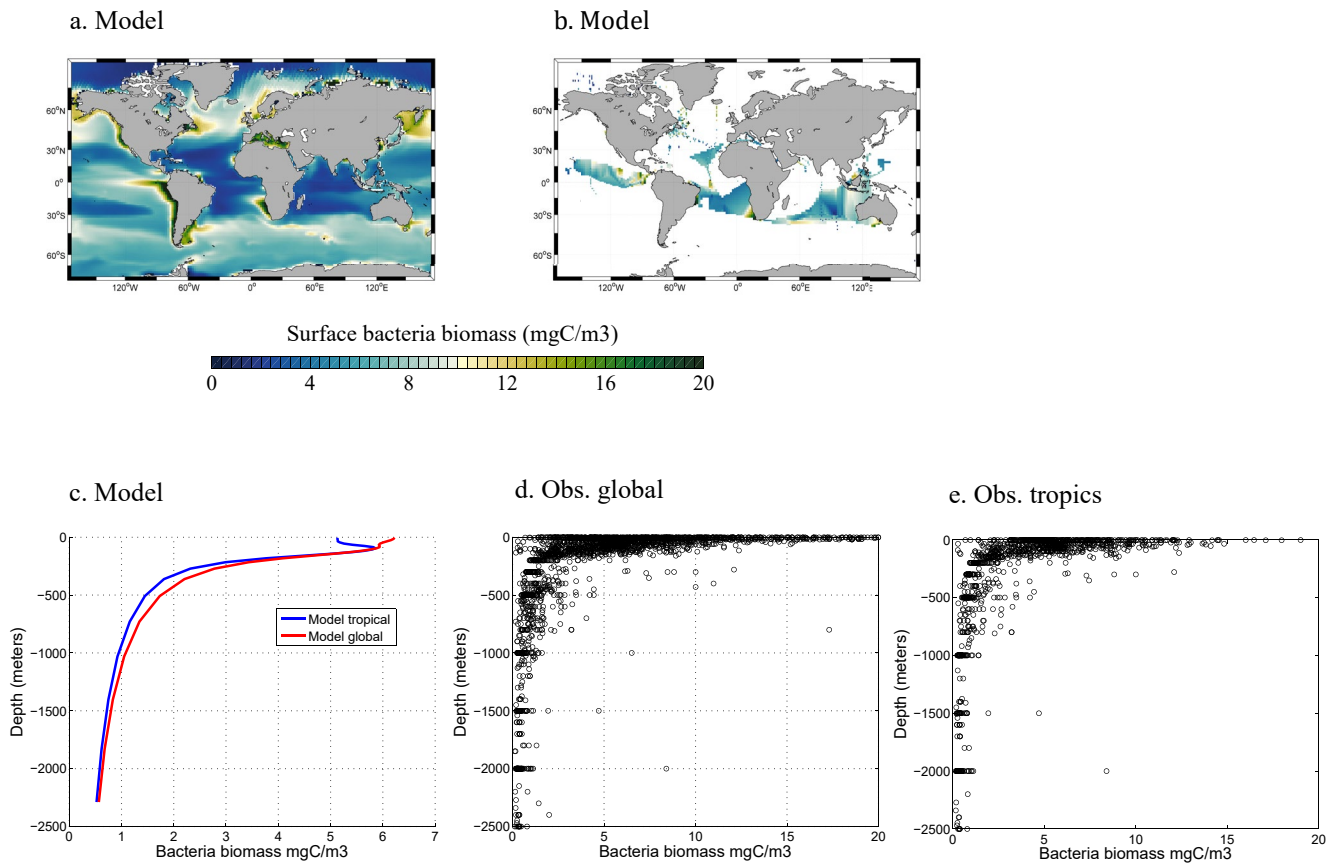


Figure 3. Annual-mean results from the standard *Control* model run. (a) Modeled surface FL bacteria biomass, (b) observations of surface heterotrophic bacteria biomass compiled by Buitenhuis et al. (2012) and Xie et al. (2021) (data are interpolated onto the model grid), (c) modeled vertical distribution of FL bacteria biomass averaged over the global ocean (red full line) and tropical ocean (20°S–20°N) (blue full line), (d) observed vertical distribution of heterotrophic bacteria biomass over the whole world's ocean; data compiled by Buitenhuis et al. (2012) and Xie et al. (2021), and (e) Same as in (d) but only for the tropics (20°S–20°N).

Exceptions are in the eastern equatorial Pacific, where FL bacteria can be limited by Fe or co-limited by both Fe and LDOC throughout the year. In addition, FL bacteria are predicted to be limited by Fe in the eastern North Pacific and in the North Atlantic during the boreal summer and are co-limited by Fe and LDOC over a large fraction of the Southern Ocean (~60°S) during the austral summer. Fe limitation emerges because these regions have high LDOC concentrations, either seasonally or throughout the year (Figure S1 in Supporting Information S1). Observational studies investigating the nutrient limitation status of FL bacteria remain limited, making it difficult to constrain our model results. However, a few field and experimental studies have shown that LDOC limits FL bacterial growth in various ocean regions and that Fe availability may control FL bacterial activity in the Southern Ocean, supporting our model results (Church et al., 2000; Fourquez et al., 2014; Obernosterer et al., 2015; Pakulski et al., 1996). Moreover, a recent incubation experiment by Fourquez et al. (2020) also suggested that in the Southern Ocean, FL bacteria could be co-limited by the availability of LDOC and Fe. These results indicate that assumptions regarding bacterial demands for Fe and LDOC will play an important role in structuring BP. We will explore this result further through model experiments in Section 4.1.

When comparing modeled Fe quotas in FL bacteria in the surface ocean with those in phytoplankton (Figure 7), we find that FL bacterial Fe quotas are almost always higher than those of phytoplankton (Figure 7). Not surprisingly, FL bacterial quotas are close to the maximal value prescribed in the model in areas where dFe concentrations are high, that is, near the coasts, in coastal upwelling systems and downstream of coastal areas or islands (such as in some areas of the Southern Ocean). In regions where Fe levels are low throughout the year, such as the central and eastern tropical Pacific and the eastern subarctic Pacific, Fe quotas in FL bacteria (and in phytoplankton) remain low at values below $20e - 6$ mol Fe/mol C. In the North Atlantic and in the Southern Oceans, these quotas are moderate to high ($30 - 50e - 6$ mol Fe/mol C). These results are consistent with the few available

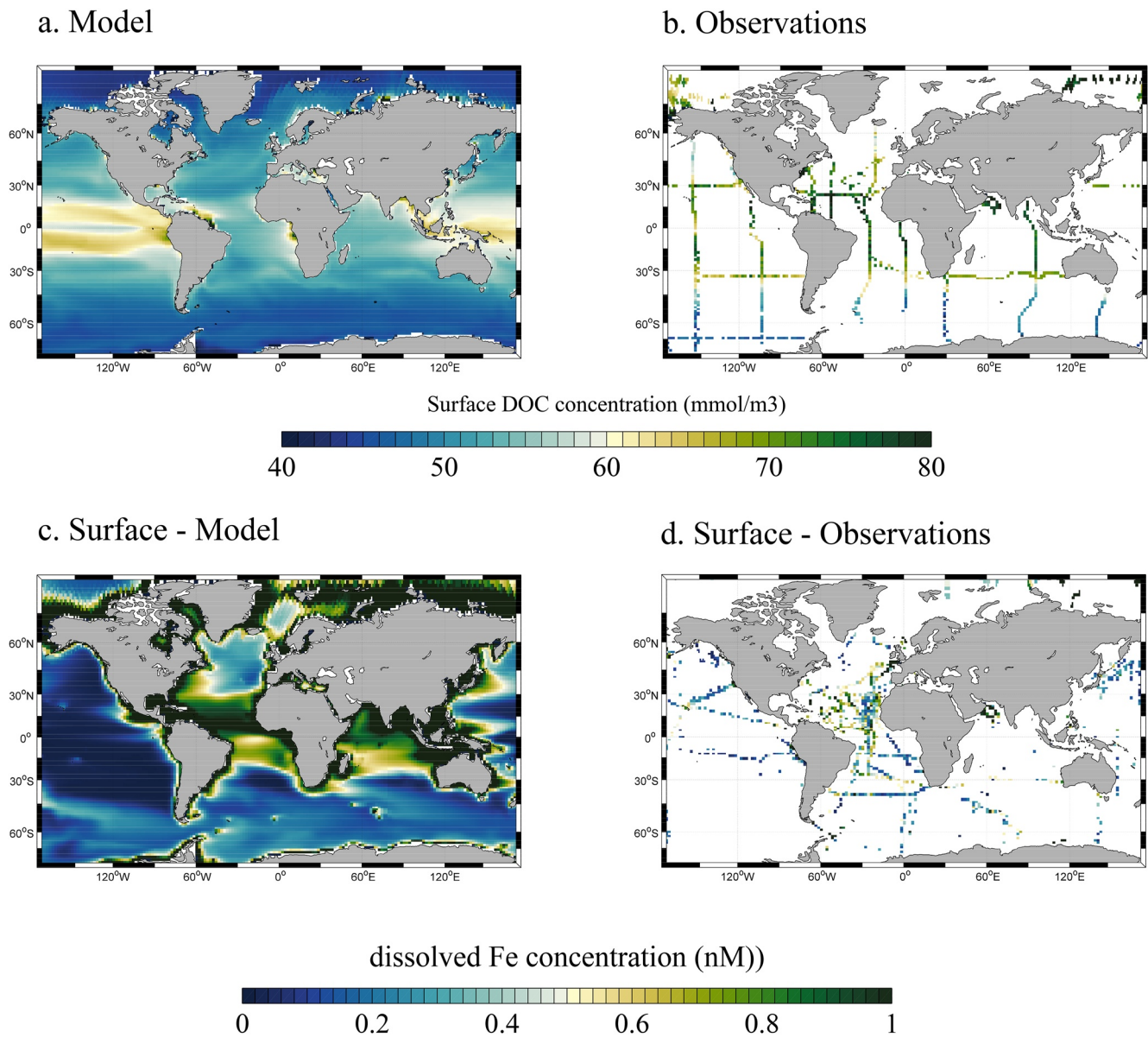


Figure 4. (a) Total DOC (LDOC + SDOC + RDOC) concentration at the surface ocean; Results from the *Control* model run, (b) Available measurements of the total DOC concentration at the surface ocean from Hansell (2013), Letscher et al. (2015), and Hansell et al. (2021). (c) Surface model dissolved Fe concentration; Results from the *Control* model run, and (d) Available measurements of the surface dissolved Fe concentrations from data set compiled by Tagliabue et al. (2012) and from GEOTRACES program (Schlitzer et al., 2018).

measurements of FL bacterial Fe quotas in the subarctic Pacific (PAPA station) (Tortell et al., 1999) and Southern Ocean (Fourquez et al., 2015), which showed values of $\sim 10e-6$ and $20-50e-6$, respectively (Figure 8).

3.3. The Role of FL Bacteria in the Fe Cycle

FL bacteria require Fe to grow and thus compete with phytoplankton for this resource in the upper ocean (0–120 m). On average over the global ocean, FL bacterial dFe uptake in the upper ocean is $0.52 \text{ nmol/m}^3/\text{year}$ (Figure 9). However, about one-third of this uptake ($0.18 \text{ nmol/m}^3/\text{year}$) is directly released to the water column by local FL bacterial mortality. This dFe flux due to FL bacterial mortality is small compared to the amount of dFe released from particle remineralization and zooplankton excretion (0.18 vs. $1.22 \text{ nmol dFe/m}^3/\text{year}$, respectively). Nevertheless, it is of the same order as the amount of dFe removed by scavenging and colloidal pumping

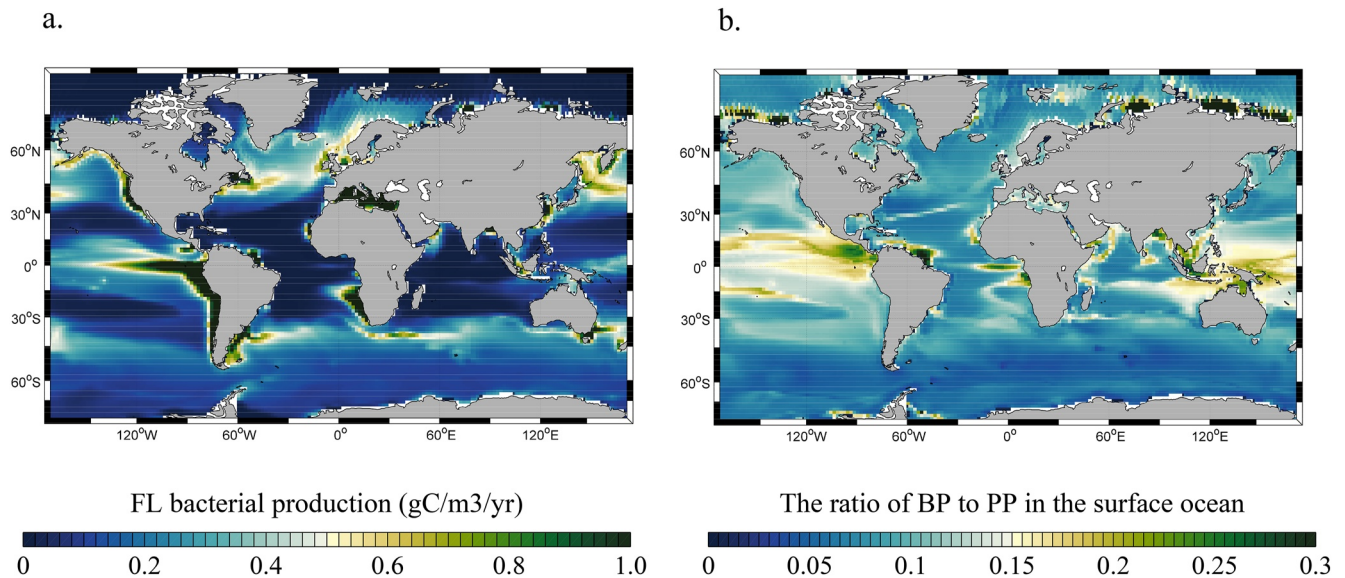


Figure 5. Results from the standard *Control* model run. (a) Annual mean surface free-living bacterial production (BP) (gC/m³/yr) and (b) Ratio of the annual-mean of BP over the annual-mean primary production in the surface ocean.

(0.28 nmol dFe/m³/year) (Figure 9). Regionally, FL bacterial Fe uptake averaged over the upper ocean is elevated along the coasts and in the mid and low latitude regions of the Indian and Atlantic Oceans (Figure 10a), where FL bacterial production is high and where both dFe concentrations and FL bacteria Fe quotas are high. Fe released by FL bacteria mortality follows the same spatial patterns as FL bacterial Fe uptake, but with a lower magnitude (Figure 10b), which is in line with the global estimate. Thus, FL bacterial activity acts as a significant removal process of dFe in the upper ocean, comparable in magnitude to scavenging and colloidal pumping processes when averaging over the upper ocean (Figure 9). However, while the latter are restricted to near-shore regions and beneath dust plumes (Figure 10c), FL bacterial removal is active throughout the open ocean.

The amount of Fe taken up by FL bacteria in the upper 120 m, 0.52 nmol Fe/m³/year, represents about 55% of the amount of Fe that is taken up phytoplankton (0.93 nmol Fe/m³/year); i.e., about one-third of the total biological Fe uptake (Figure 9). In HNLC regions such as the North Pacific, eastern equatorial Pacific, and some areas of the Southern Ocean, Fe uptake by FL bacteria can be up to ~50% of the total biological uptake (Figure 10c). In the Arctic Ocean and coastal regions, the Fe uptake by FL bacteria can even be close ~60% of the total biological Fe uptake. This is because LDOC is more available near the coast and phytoplankton is more limited by light in the Arctic Ocean. Our results are thus consistent with previous measurements in the subarctic Pacific, which reported that FL bacteria account for ~20%–40% of the total biological Fe uptake (Tortell et al., 1996). In addition, various field and incubation and modeling studies in the Southern Ocean (Boyd et al., 2012; Fourquez et al., 2020; Kuparinen et al., 2011; Ratnarajah et al., 2021) showed that FL bacteria are a significant competitor with phytoplankton for Fe if LDOC is available.

While phytoplankton activity only directly affects ocean Fe distribution in the euphotic zone, FL bacteria consume and release Fe via their growth and mortality throughout the water column. In the mesopelagic domain (defined here as 200–500 m), Fe fluxes induced by FL bacterial uptake and release are roughly equal (Figure 9: ~0.04–0.05 nmol Fe/m³/year) when averaged over the global ocean. In this vertical domain, the FL bacterial Fe release flux is always >70% of the FL bacterial Fe uptake flux and reaches ~90% over most of the world ocean (Figure S5b in Supporting Information S1). Thus, although the net impact of FL bacteria on mesopelagic dFe is a removal process, it does not have a significant impact on the dFe distribution as most of the Fe that FL bacteria take up is released back to seawater at the same location. Comparing with the Fe released by particle solubilization and the Fe removal by scavenging and colloidal pumping (Figure 9; 0.05 and 0.06 nmol Fe/m³/year), which are the most significant Fe sources and sinks in the mesopelagic zone, the net FL bacterial uptake minus mortality of Fe is an order of magnitude smaller (0.007 nmol Fe/m³/year). The regional distribution of this net uptake closely follows that of the export production via sinking particles since FL bacteria are strongly limited

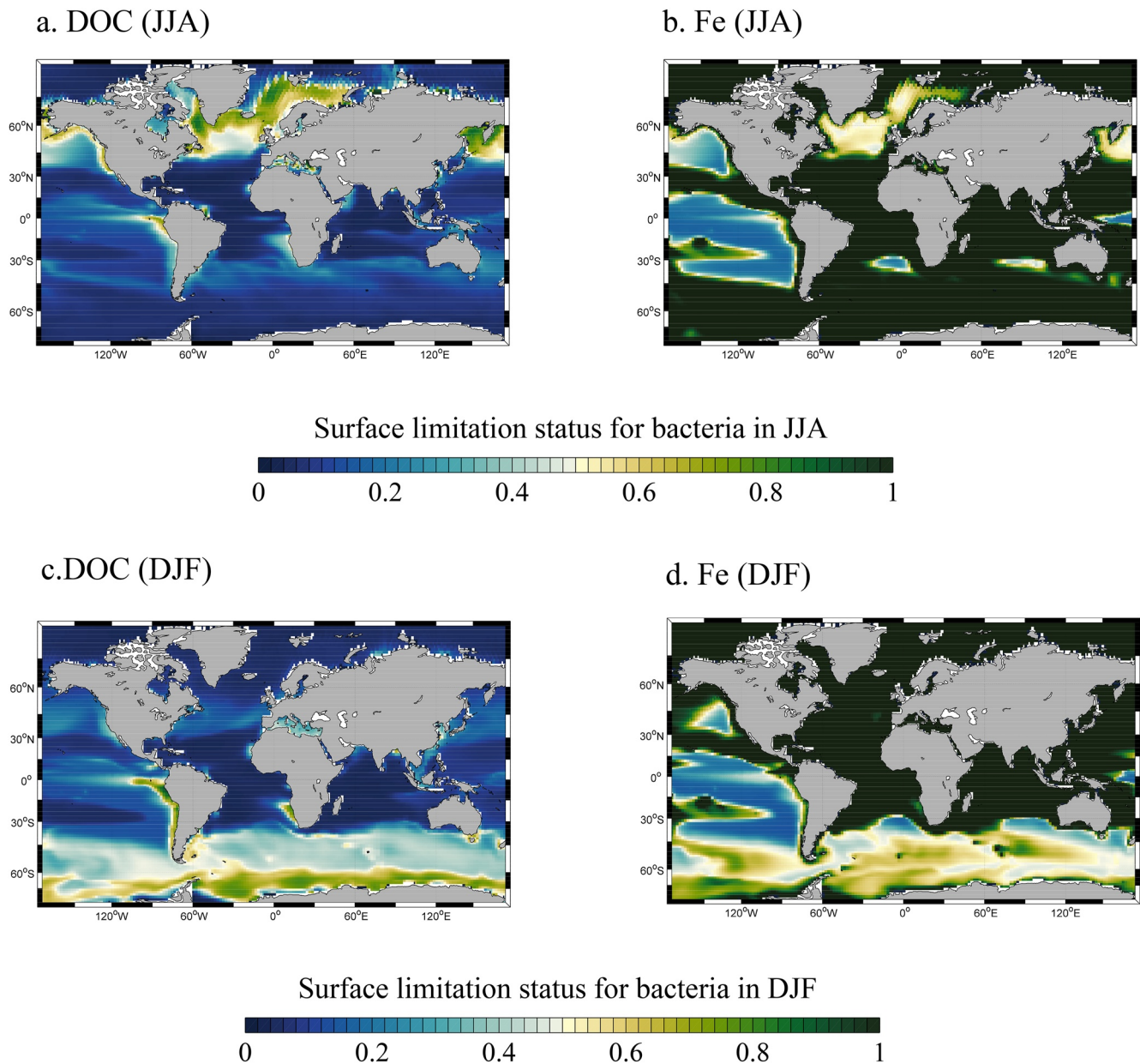


Figure 6. Results from the standard *Control* model run. Upper panels: surface nutrient limitation status of free-living (FL) bacteria averaged in JJA (June, July, and August). Lower panels: surface nutrient limitation status of FL bacteria averaged in DJF (December, January, and February). (a and c) Labile DOC (LDOC); (b and d) Fe. The most limiting nutrient is the nutrient that has lower value for this metric.

by LDOC, which is supplied to the mesopelagic domain via particle solubilization (Figure 11). In contrast to the upper 120 m, scavenging and colloidal pumping play a more significant role in the mesopelagic domain of the open ocean and are not restricted to regions affected by high levels of dust deposition (Figure 11).

Interestingly, microzooplankton satisfy a large portion of their Fe demand from FL bacteria. Averaged over the upper 120 m, Fe supplied by predation on FL bacteria amounts to 46% of the Fe supplied to microzooplankton by phytoplankton (Figure 9: 0.34 vs. 0.74 nmol Fe/m³/year). This contribution is significantly higher than the relative amount of carbon that is acquired by microzooplankton from FL bacteria, 32%. This is primarily due to the higher Fe/C ratio in FL bacteria than in phytoplankton (Figure 7). The preference of zooplankton on bacteria over phytoplankton and the resulting higher Fe/C ratios of zooplankton food might lead to changes in the flow and recycling fluxes of Fe and carbon within the food-web.

a. Bacteria

b. Phytoplankton

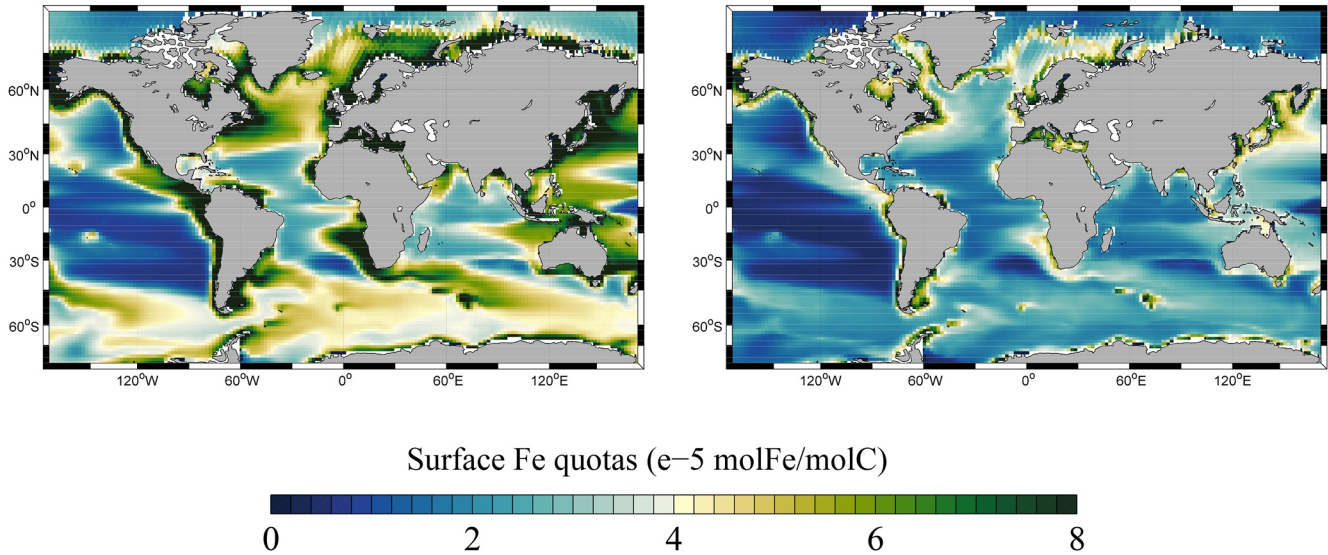


Figure 7. Results from the standard *Control* model run. (a) Annual mean surface Fe quota of free-living bacteria (biomass weighted) and (b) annual mean surface Fe quota of phytoplankton (biomass weighted).

3.4. Sensitivities of FL Bacterial Growth and Activities to Their Fe and DOC Requirements

In this section, we explore the sensitivities of FL bacterial growth and their role in the ocean Fe cycle to assumptions regarding the demands of FL bacteria for Fe and LDOC. As mentioned in the previous sections, these demands are poorly constrained due to the paucity of in situ and laboratory observations. The few available observations suggest a wide range of values for the parameters used in our model, especially for the minimum, optimal, and maximum FL bacterial Fe quotas (Fourquez et al., 2015, 2020; Mazzotta et al., 2020; Tortell et al., 1999) Using results of this sensitivity analysis, we then discuss consequences of the resource competition between FL bacteria and phytoplankton on phytoplankton blooms and carbon export.

a. 10Femin

b. 3HalfK

c. 3DOC

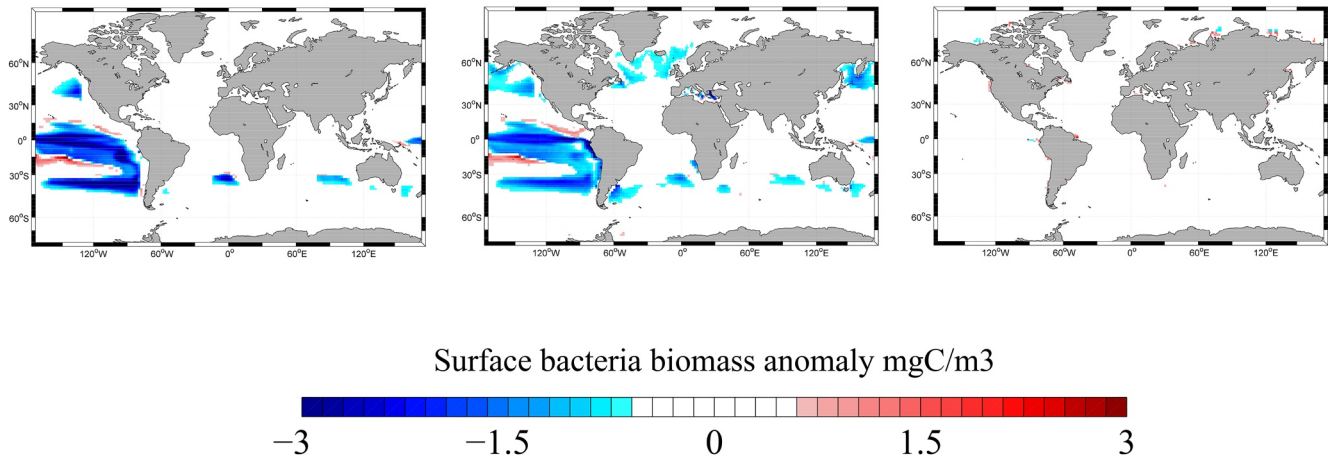


Figure 8. Surface free-living bacteria biomass anomalies (annual means) in (a) *10Femin*, (b) *3HalfK*, and (c) *3DOC*. Anomalies are computed with respect to the *Control* run.

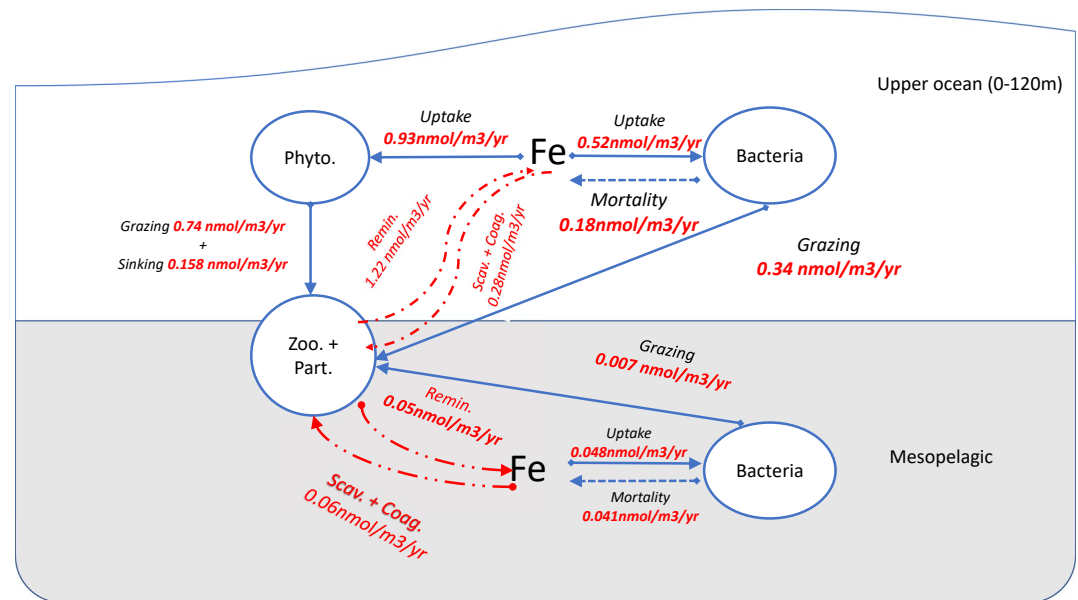


Figure 9. Schematic representation of the role of free-living bacteria on ocean Fe cycling in the upper ocean (0–120 m) and mesopelagic zones (200–500 m). Biological uptake, mortality release, remineralization, grazing, and sinking fluxes of Fe, averaged over the upper ocean or mesopelagic zone, are shown in red. Removal Fe fluxes by scavenging and colloidal pumping are also shown. Results are from the *Control* model run.

3.4.1. Sensitivities of the Growth of FL Bacteria to Their Fe and LDOC Demands

Despite a large increase in the model parameters associated with the FL bacteria Fe quotas and Fe uptake capacities (*10Femin*, *3HalfK*, *80FeC*, and *LOWHALFK*), the globally integrated FL bacteria biomass is only changed in a minor way (Table 2). This suggests that at the global scale, the FL bacteria biomass is rather insensitive to these model parameters, which can be explained by analyzing changes occurring regionally. Surface FL bacteria biomass decreased sharply (by up to 50%) in the HNLC regions (Figure 8 and Figure S4 in Supporting Information S1). In the North Atlantic and part of the North Pacific, the more modest biomass decline is due to a more severe seasonal Fe limitation that emerges in summer, after the phytoplankton spring bloom depletes surface water dFe. FL bacteria biomass appears to be more sensitive to the value of their minimum Fe requirement (*10Femin*) in the perennially Fe-limited, HNLC regions, whereas in regions where Fe limitation is strongly seasonal, changes in the half-saturation constant (*3HalfK*) have the strongest impact. These results indicate that the Fe uptake capability of FL bacteria is crucial for their Fe acquisition in the seasonal Fe-limitation regions, where high Fe supplies from the subsurface water is highly seasonal. On the other hand, in regions where Fe levels are low all year round, their Fe requirements appear to be more important. Although observations have shown that diatoms in the Southern Ocean have evolved to exceptionally low Fe/C minimum ratios, we are not aware of any study focusing on the adaptation of FL bacteria to continuous low Fe availability (Strzepek et al., 2012, 2019). Downstream of the HNLC regions, a significant increase in the surface FL bacterial biomass is simulated. This is because in HNLC regions, a stronger Fe limitation leads to an increased build up of LDOC and SDOC at the surface, which is then transported to support the growth of FL bacteria in LDOC limited areas (Figure S3 in Supporting Information S1). The compensation between regions of increased and decreased FL bacterial growth explains the weak sensitivity of the global bacterial biomass to changes in the Fe-related parameter values.

Quite surprisingly, an increase in the half-saturation constant of LDOC uptake (*3DOC*) results in very small changes in the surface bacteria biomass. This arises because, as FL bacteria are the sole consumers of LDOC in our model, a decreased efficiency in their LDOC uptake results in higher surface LDOC concentrations. These increased LDOC concentrations then compensate for the decreased efficiency of FL bacterial growth on LDOC, and thus their biomass is globally insensitive to even very large changes in the LDOC half-saturation constant. FL bacteria are more sensitive in our model to Fe uptake because there is a competition for Fe between FL bacteria and phytoplankton. However, numerous studies have evidenced that many phytoplankton species are capable of utilizing dissolved organic carbon to some extent (in case of light limitation, for instance) (Flynn et al., 2012;

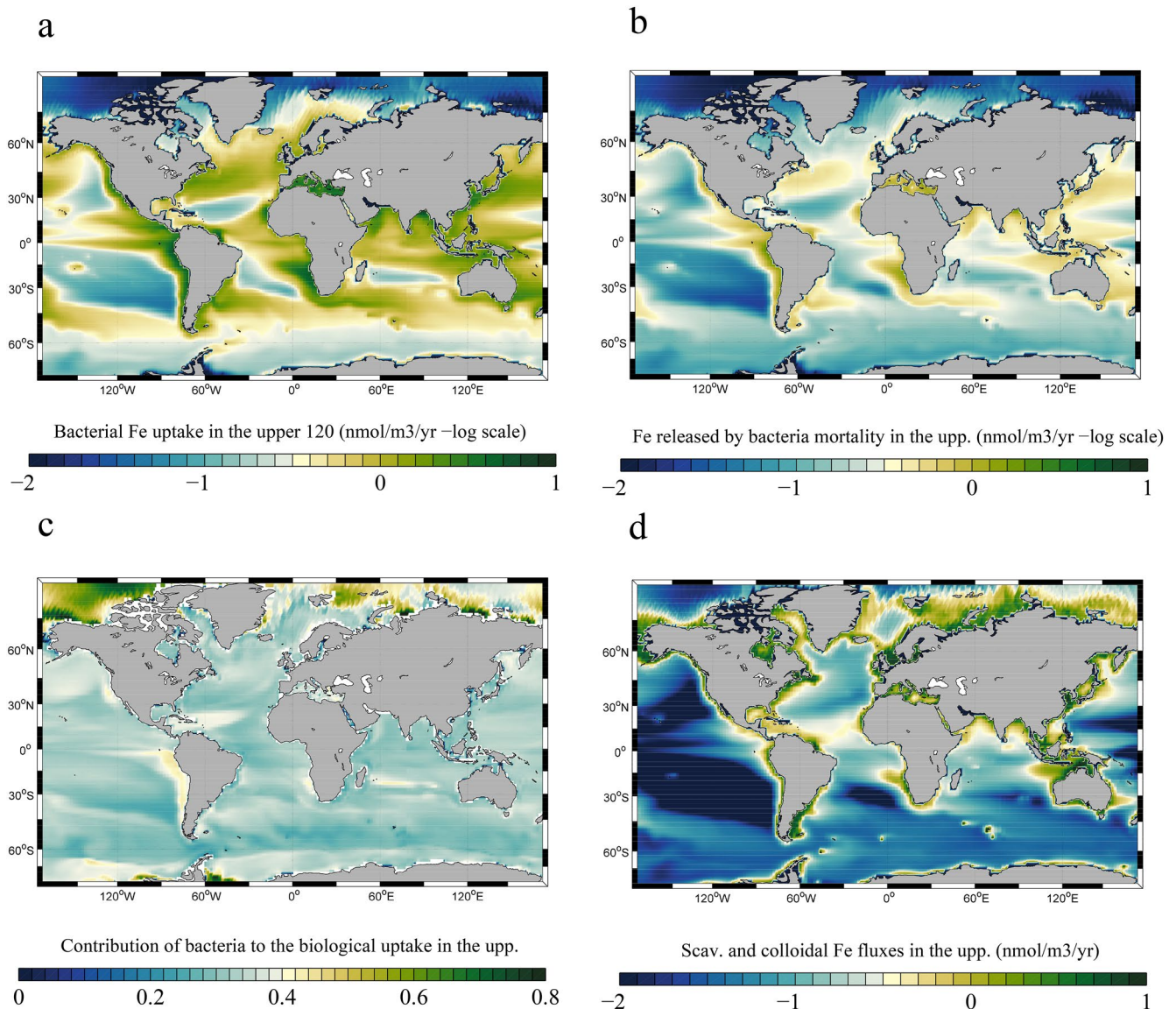


Figure 10. Results (annual average) from the standard *Control* model run. (a) free-living (FL) bacterial Fe uptake in the upper ocean (0–120 m), (b) Fe released in the upper ocean by FL bacteria mortality, (c) the relative contribution of FL bacteria to the biological Fe uptake in the upper ocean (0–120 m), and (d) scavenging and colloidal Fe fluxes in the upper ocean (0–120 m).

Stoecker et al., 2017). This suggests that incorporating the competition between phytoplankton and FL bacteria for LDOC into our model may increase the sensitivity of FL bacteria biomass to FL bacteria LDOC uptake capabilities (Kamjunke et al., 2008).

When FL bacterial Fe storage increases either by an increase in the maximum storage capability of FL bacteria (*80FeC* or by an increase in their Fe uptake capability) (*LOWHALFK*), FL bacteria biomass surprisingly still decreases in the HNLC regions of the tropical and North Pacific. This is because when FL bacteria are able to store more Fe in their cells or uptake Fe more efficiently, phytoplankton become more Fe-limited in HNLC regions (not shown). Phytoplankton thus produce less LDOC, which then limits for FL bacteria. This result suggests that the LDOC limitation of FL bacteria biomass can originate from their competition with phytoplankton for Fe.

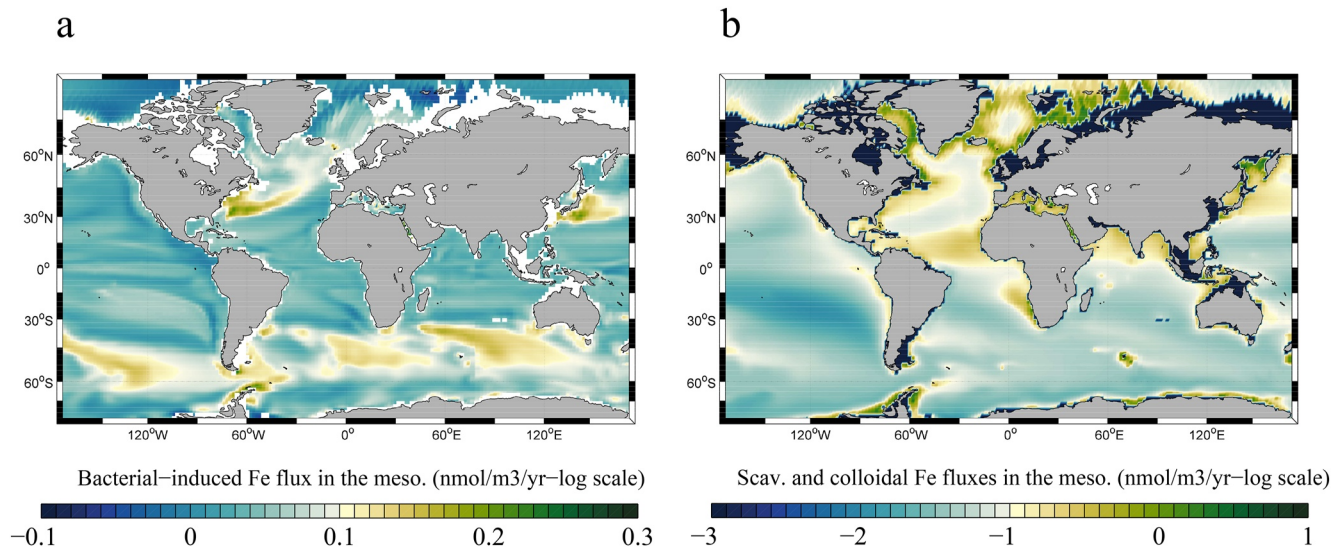


Figure 11. Results (annual average) from the standard *Control* model run. (a) free-living bacterial-induced Fe flux (uptake—release) in the mesopelagic zone (200–500 m) and (b) Fe fluxes due to scavenging and colloidal pumping in the mesopelagic zone.

3.4.2. Sensitivity of Fe Fluxes Induced by FL Bacteria to Their Fe Demand and Storage

As expected, changes in the FL bacteria quotas result in significant modifications of the Fe fluxes due to FL bacterial activities (Figure 12a). In the upper 120 m of the ocean, the globally averaged Fe uptake by FL bacteria increases to almost the same value as the Fe uptake by phytoplankton in *80FeC*, while it is reduced by a factor of about 5 in *10FeC*. At the regional scale, setting the Fe quota everywhere to its maximum value produces the largest changes in the FL bacterial contribution to the biological Fe uptake in the HNLC regions (~60%), that is, where quotas are the lowest in the control experiment (Figure S6a in Supporting Information S1). Conversely, imposing a relatively modest constant Fe/C ratio in FL bacteria greatly reduces the contribution of FL bacteria, in particular in regions where Fe is not limiting (Figure S6b in Supporting Information S1). The relative amount of Fe that is released to sea water by FL bacteria mortality is globally unchanged, at about 30% of the uptake in all sensitivity experiments (Figure S7 in Supporting Information S1).

Globally averaged Fe uptake by phytoplankton remains almost unchanged in all model sensitivity experiments, remaining close to 1 mmol/m³/year (Figure 12a). This result suggests that FL bacterial activity does not significantly alter the amount of bioavailable Fe for phytoplankton when globally integrated. This is because when FL bacterial Fe uptake is strongly increased/decreased, the release of Fe from remineralization is also increased/decreased. Thus, Fe that is taken by FL bacteria in the upper ocean is largely recycled there, and only a small fraction of it is lost to the interior of the ocean in our model.

In the mesopelagic domain, changes in the FL bacterial Fe demand and quotas do not have a strong impact on the FL bacterial Fe fluxes, except in *10FeC* (Figure 12b), and the net Fe flux by FL bacterial activities in the mesopelagic domain remains very small relative to the Fe losses due to scavenging throughout all model experiments (Figure 11). In the *10FeC* experiment, Fe fluxes through the FL bacterial pool are strongly reduced, and the Fe fluxes induced by scavenging and remineralization are decreased due to a decrease in the particulate Fe

export (Figure S8 in Supporting Information S1). In the other experiments, all fluxes appear virtually identical. Below the euphotic zone, Fe is generally not a limiting factor for FL bacteria, and the FL bacterial Fe uptake rates and quotas are thus at their maximum prescribed values. As a result, changing the half-saturation constant, the minimum Fe demand, or setting Fe quotas at their maximum values do not change the Fe fluxes driven by FL bacteria in the mesopelagic domain. Moreover, as mentioned in the previous section, most of the Fe that is assimilated by FL bacteria in this domain is released back to seawater at the same location by mortality. In our model, the FL bacteria biomass decreases to very low levels below the euphotic zone due to

Table 2
Global Integrated Free-Living Bacteria Biomass (BB PgC), Primary Production (PP PgC/Year), and Export Carbon Production at 120 m (EPC PgC/Year) for Each Model Sensitivity Experiment

	Control	10Femin	3HalfK	3DOC	80FeC	10FeC
BB	1.409	1.411	1.421	1.406	1.39	1.409
PP	32.19	31.53	31.75	32.31	30.87	32.18
EPC	6.434	6.395	6.432	6.443	6.298	6.434

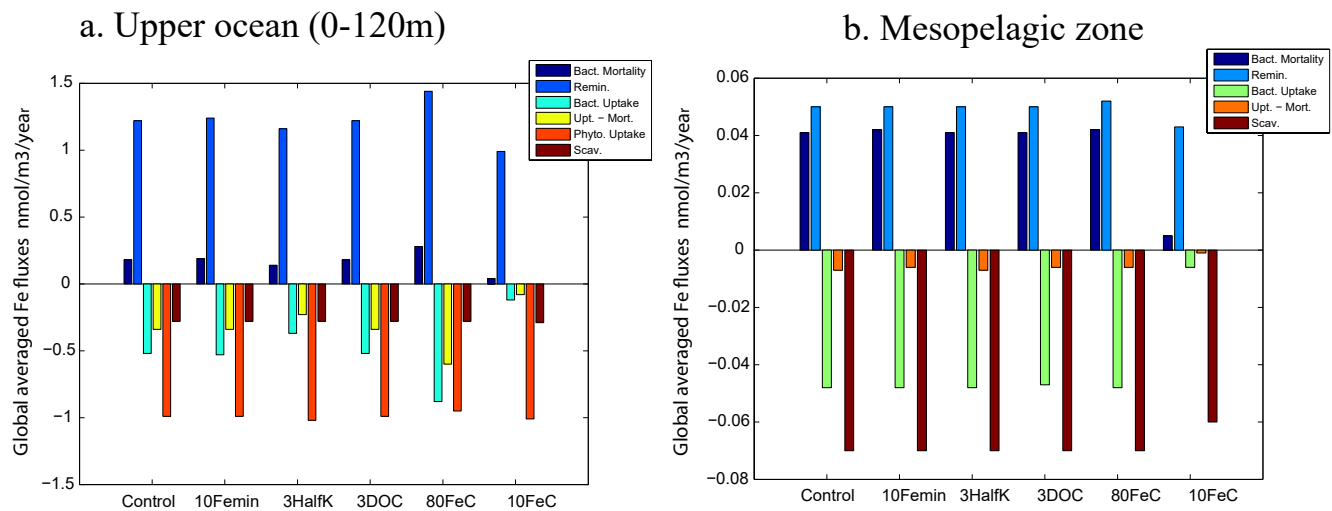


Figure 12. The global averaged Fe fluxes for major processes in the upper ocean (0–120 m) (a) and mesopelagic zone (b) from different model sensitivity experiments.

a strong LDOC limitation (Figure 3c). These low bacteria concentrations are insufficient to ensure the survival of microzooplankton, which become virtually extinct in the intermediate and deep ocean. This in turn leads to a low production of particulate biogenic Fe, which would decouple the FL bacterial Fe release flux from Fe uptake flux. This result reflects a clear deficiency of our model as observations showed a low, but non zero, microzooplankton community feeding on FL bacteria (Anderson & Tang, 2010; Giering et al., 2014; Mazuecos et al., 2015; Steinberg & Landry, 2017; Steinberg et al., 2008).

3.4.3. Consequences of Resource Competition Between FL Bacteria and Phytoplankton on Ocean Primary Production and Carbon Export

A strong increase in the FL bacterial Fe demand (*80FeC*) can significantly alter the seasonal cycle of phytoplankton biomass and chlorophyll (Figure 14). In the North Atlantic, western North Pacific, and part of the Southern Ocean, the amplitude of the seasonal variations of surface chlorophyll is reduced by up to 0.3 mg Chl/m³. During the productive season, the release of LDOC by phytoplankton stimulates FL bacterial growth. The high Fe demand of the latter, which is greatly enhanced in *80FeC*, leads to a rapid and efficient uptake of dFe from seawater. As a result, phytoplankton becomes more quickly Fe limited. Hence, the phytoplankton bloom is significantly shortened and reduced in magnitude. We also note an increase in the phytoplankton seasonal cycle at the boundaries of Fe limited regions, which further illustrates the increased lateral transport of macronutrients due to a stronger Fe limitation. In the other sensitivity experiments, very small changes in the amplitude of the chlorophyll seasonal cycle are predicted. These changes are mainly restricted to oligotrophic regions downstream of Fe-limited areas resulting, as in the *80FeC* experiment, from an enhanced seasonal lateral transport of macronutrients.

These findings are in agreement with the results of Ratnarajah et al. (2021), who used a mechanistic model of FL bacteria-phytoplankton competition in an idealized setting: Luxury Fe uptake by FL bacteria exacerbates the competition between FL bacteria and phytoplankton. Furthermore, in highly seasonal regimes, such as at mid- and high-latitudes, competition for Fe increases over the course of the productive season as the accumulation of LDOC produced by phytoplankton exudation, sloppy feeding, and zooplankton excretion stimulates the FL bacterial activity (Sturluson et al., 2008). Hence, an increased capability of FL bacteria to take up and accumulate Fe results in minimal changes during the onset and development of the phytoplankton bloom and in maximum impacts in its later stages (not shown). Thus, even though impacts can be significant in some regions, the effect of FL bacterial Fe uptake on the amplitude of the chlorophyll seasonal cycle remains at most moderate and is hampered by a strong limitation of FL bacteria by LDOC. An increased competition for Fe with phytoplankton would be possible if there were a stronger production of LDOC during the onset of the bloom than that predicted by our model or in the presence of external sources of labile DOC such as sea ice (Kähler et al., 1997; Underwood et al., 2010), ice sheets (Lawson et al., 2014) or sediments (Davis & Benner, 2007).

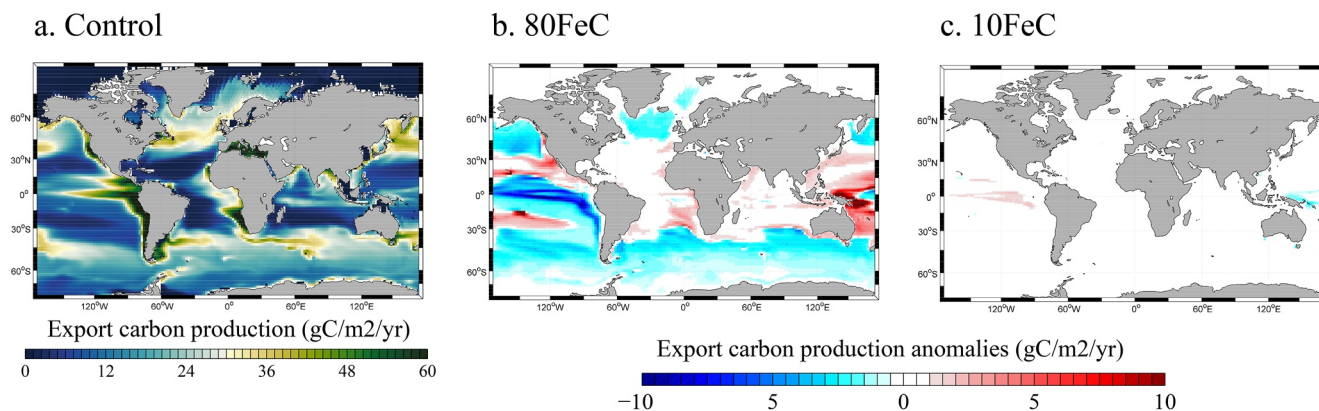


Figure 13. Export carbon production at 100 m (annual average). Results from the *Control* run (a), anomalies between the *80FeC* and *Control* run (b), and anomalies between the *10FeC* and *Control* run (c).

At the global scale, the sensitivity of carbon export (export carbon production, EPC) and primary production (PP) to the Fe bacterial uptake and demand and to the half-saturation constant of LDOC uptake is weak (Table 2). EPC and PP are changed by at most 2% and 3.5% relative to their absolute annual means, respectively. In the *80FeC* experiment, large absolute changes are predicted in the eastern equatorial Pacific Ocean where EPC is reduced by 5–10 gC/m²/yr, (i.e., by about one-third of its annual mean value) (Figure 13), and in the mid to high latitudes of both hemispheres where EPC decreases by 1–5 gC/m²/yr, (i.e., by about 10%–20% of its annual mean value). In contrast, in the subtropical gyres and in the Warm Pool of the Pacific Ocean, EPC is significantly increased, by up to 10 gC/m²/yr in the Warm Pool, due to a stronger lateral export of nutrients from the Fe limited areas (Figure S10 in Supporting Information S1). The increase in export production in the oligotrophic areas compensates for the decrease in Fe limited areas, explaining the very low sensitivity of PP and EPC globally in *80FeC*. In the other sensitivity experiments, regional changes in EPC are negligible or very small. The low sensitivity of the carbon cycle in our model at the global scale to changes related to FL bacterial processes is not surprising since the ocean carbon cycle in PISCES has been thoroughly tuned and structured to match available observations. To a large extent, global EPC is constrained by the ocean dynamics and the vertical gradients in nutrients. Providing that both are not severely altered, global EPC should remain relatively invariant. Yet, changes in EPC can be significant at the regional scale, especially in Fe-limited regions, in response to the greater Fe uptake by FL bacteria.

4. Summary and Broader Implications

4.1. Summary

In the ocean, FL bacteria require a suite of resources for their growth, especially Fe and LDOC (Church et al., 2000; Obernosterer et al., 2015; Pakulski et al., 1996). When both the labile DOC and Fe concentrations are low, it is unclear which resource is the ultimate limiting factor for FL bacterial growth or whether they can be co-limiting. We showed through different model experiments that over most of the world's ocean and over most of the year, LDOC is the ultimate limiting resource for the production of FL bacteria. Fe can emerge as the (co-) limiting factor for FL bacterial growth during the growth season or throughout the year in Fe-limited regions, that is, the equatorial Pacific, the eastern subarctic Pacific, in part of the Southern Ocean, and in some sectors of the North Atlantic Ocean. In those regions, the biomass of FL bacteria is then highly sensitive to the FL bacterial Fe demand and uptake capability. Specifically, in the seasonally Fe-limited regions such as the North Atlantic, the Fe uptake capability of FL bacteria is crucial for their Fe acquisition, whereas in regions where Fe levels are low all year round, changes in the Fe requirements of FL bacteria (i.e., their minimum and optimal Fe quotas) appear to be a more efficient strategy. Studies on the adaptation of FL bacteria to low Fe conditions are needed to confirm these results.

Through different model experiments, we also showed that FL bacteria can play an important role in the ocean Fe cycle, at least in the upper 120 m of the ocean. Fe uptake by FL bacteria is found to contribute to between 30% and 50% of the total biological uptake in the upper ocean, making it comparable to that by phytoplankton. Consequently, FL bacteria may compete with phytoplankton for Fe in Fe-limited areas, such as the equatorial Pacific,

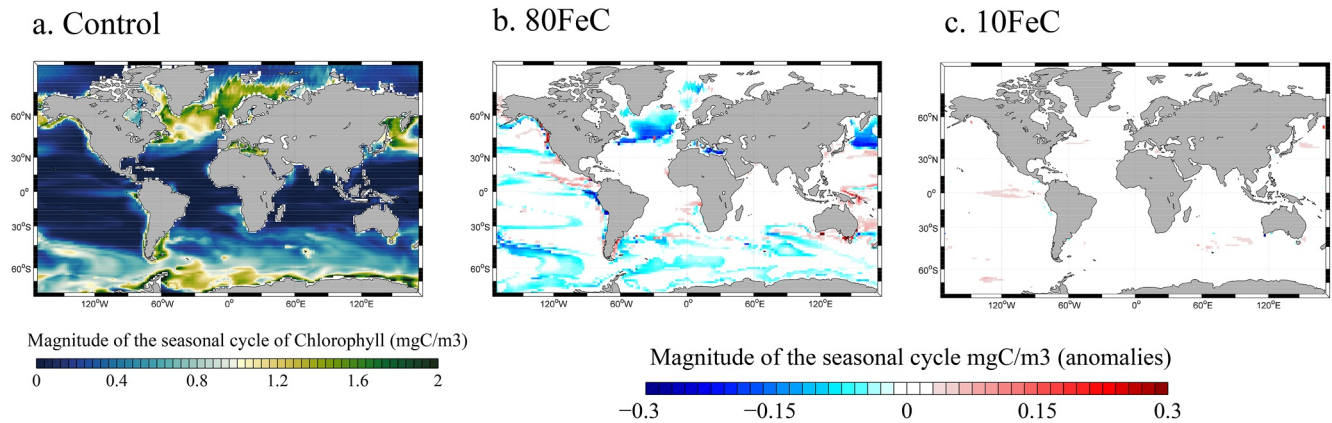


Figure 14. Amplitude of the chlorophyll seasonal cycle. (a) Results from the *Control* run, (b) anomalies between the *80FeC* run and *Control* run, and (c) anomalies between the *10FeC* run and *Control* run.

the subarctic Pacific, Southern Ocean, and the North Atlantic Ocean in summer. At high latitudes, this competition reduces the magnitude and duration of the spring phytoplankton bloom, which decreases the production of LDOC needed for FL bacteria growth. However, the impact of FL bacteria on phytoplankton remains moderate, even when a large bacterial Fe content is prescribed. This arises due to a strong limitation of FL bacteria by LDOC, especially during the onset of the phytoplankton bloom. Evidence for a strong limitation of FL bacterial growth by LDOC has been evidenced during the early and mid-stages of the spring phytoplankton blooms by numerous observations (e.g., Bird & Karl, 1999; Ducklow, 2003; Pomeroy et al., 1991).

The degree of competition between bacteria-phytoplankton is very sensitive to the modeled FL bacterial Fe demand and quotas, which are still poorly constrained at between $1e - 6$ and more than $80e - 6$ mol Fe/mol C by observations (Fourquez et al., 2015; Mazzotta et al., 2020; Tortell et al., 1996, 1999). An elevated FL bacterial demand for Fe, either to meet their metabolic requirements or to build a large Fe internal storage through luxury uptake, enhances their competition with phytoplankton (Bowie et al., 2001; Ratnarajah et al., 2021) and the amount of Fe that is cycled through FL bacteria (Figure 12). However, our model results suggest that most of the Fe used by FL bacteria is recycled in the upper ocean, with very little exported to the ocean interior. Unfortunately, quantitative observational evidence on the efficiency of Fe export due to the bacterial loop is lacking at this stage. Nevertheless, rapid and efficient Fe recycling by bacterial activity in the upper ocean is consistent with a range of field-based estimates (Boyd et al., 2005, 2012, 2015; Obernosterer et al., 2008; Tagliabue et al., 2014).

In our model, FL bacteria activities in the mesopelagic Fe cycle have a negligible impact on Fe cycling due to a tight local coupling between FL bacterial Fe uptake and release fluxes, with $\sim 90\%$ of the Fe that is taken up by FL bacteria is released back to seawater via mortality. Changes in the FL bacterial demand and storage capacity do not significantly alter this proportion, as shown by our sensitivity experiments. The tight local coupling between FL bacterial Fe uptake and release fluxes in our model is due to reduced grazing loss of bacteria by low microzooplankton populations in the subsurface ocean domain. Greater grazing of FL bacteria would transfer Fe in their cells to the sinking detrital pool. Thus, although the flux from FL bacteria to zooplankton remains largely unknown, our model most likely underestimates the role of FL bacteria in the mesopelagic Fe cycle.

4.2. Broader Implications

Our model study on the interactions between FL bacteria and Fe in the ocean is a first step and greater effort is needed to develop a holistic consideration of the role of bacteria in the ocean iron cycle. Key gaps associated with the FL bacteria physiology include, for instance, their Fe acquisition strategies and the role of Fe in their metabolism, as well as the interactions between FL bacteria, phytoplankton, and zooplankton. In addition, we did not consider in our model the bacterial production of specific ligands, like siderophores (Amin et al., 2009; Boiteau et al., 2016; Bundy et al., 2018), which could strengthen the interactions between Fe, bacteria, and phytoplankton (Guieu et al., 2019; Ito et al., 2016; Krishnamurthy et al., 2009; Pham & Ito, 2021).

More broadly, our model, as for other ocean biogeochemistry models representing bacteria (Stock et al., 2020), only considered carbon-oxidizing free-living bacteria. The omission of other key groups of bacteria, such as particle-attached, nitrifying-, or sulfur-oxidizing bacteria, and archaea leaves a large gap in our understanding of how bacteria affect the Fe cycle. For instance, particle-attached, carbon-oxidizing bacteria have been hypothesized to contribute to the decoupling between the release of Fe and macro-nutrients in sinking particles (Boyd et al., 2017; Tagliabue et al., 2019). Ammonia- and nitrite-oxidizing bacteria and archaea may have relatively high Fe contents that could result in their limitation by Fe in the upper ocean (Kim et al., 2021; Shafiee et al., 2019) and also lead to significant levels of Fe storage in low oxygen mesopelagic regions (Saito et al., 2020). Therefore, there is an urgent need for future modeling studies that explicitly represent different types of bacteria in order to have a more integrative understanding of the role of bacteria in changing ocean ecosystems. To do so requires new linkages between different types of observations and modeling. The increasing amount of information from genomic and metagenomic studies provides a new opportunity to fill these gaps (Caputi et al., 2019; Debeljak et al., 2019; Hogle et al., 2014; B. M. Hopkinson & Barbeau, 2012). The transformation in understanding would also require the combination of omics data with insights into biogeochemical processes and the cycling of resources from the GEOTRACES program (Schlitzer et al., 2018), as has been done in studies for phytoplankton (Caputi et al., 2019; Marchetti, 2019).

Data Availability Statement

The model outputs, relevant data, and MATLAB scripts to reproduce the figures shown in this manuscript are stored at Zenodo (<https://doi.org/10.5281/zenodo.6476611>).

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