

Processes and patterns of oceanic nutrient limitation

Many of the essential biological elements exist in multiple physico-chemical forms within the marine environment. However, the different nutrient species of a required element may not be equally available for biological uptake, either to the whole community/assemblage or for specific members of the biota. For example, sulphate is unavailable to some major clades of marine bacteria¹ and the use of N₂ gas to satisfy cellular N demand is restricted to certain prokaryotic groups², while some of the minerals contributing to atmospheric deposition of Fe to the ocean likely contribute little to pelagic biological uptake³⁻⁵. Although the term bioavailability is sometimes assumed to correspond to a measure of the absolute availability of a given nutrient to the biota, under many circumstances the bioavailability of a chemical species or complexed element may be better described in terms of the community uptake rate⁶. Consequently bioavailability reflects a combined characteristic of both the chemical form of a nutrient and the capabilities of the extant biological community.

An example of the importance of bioavailability concerns the dissolved organic nutrient pools in the upper ocean. The terms dissolved organic nitrogen and phosphorous (DON and DOP) are operationally used to describe the whole range of organic N or P containing compounds^{7,8}. However, the DON and DOP pools are neither completely available nor unavailable. Rather both pools constitute a complex mixture of compounds whose biological rates of uptake will vary both for individual organisms and between organisms⁸⁻¹⁴. Consequently, over timescales comparable to the residence time of the upper (inorganic nutrient depleted) ocean, only a proportion of the DON and DOP pools can be accessed. Hence, although there is a large DON pool in the N limited low latitude systems, only a small proportion of this is available for uptake over biologically relevant timescales. Indeed, the observed accumulation of DON in systems exhibiting good evidence of N limitation (Fig. 3) is itself suggestive of low bioavailability for much of the DON pool^{7,15-17}. Overall the integrated bioavailability and/or chemical lability of the DOP pool appears to be higher than that for the DON pool^{15,16,18,19}.

Treatment of nutrient limitation within large scale models.

As reviewed in detail elsewhere^{20,21}, within the majority of large-scale numerical ocean biogeochemical general circulation models, phytoplankton growth has traditionally been assumed to be proportional to the external nutrient concentration assuming balanced growth within a Monod type model²², with nutrient uptake parameterised as a saturating function of external nutrient concentrations using a Michaelis-Menten functional form²⁰⁻²². Within multi-element models, constant stoichiometry is also generally assumed²¹. Co-limitation by different nutrients then typically conforms to the simple minimum-type concept where growth is dictated by the smallest of all the individual Michaelis-Menten terms for different nutrients. Although such models are simple to implement, microbial growth rates are actually more likely to be related to the intracellular rather than extracellular nutrient concentrations^{21,23,24}. Moreover, intracellular stoichiometric variability is intimately linked to nutrient limitation^{21,25-28}.

Although a wide range of more physiologically realistic growth models exist²⁹⁻³³, there is arguably a lack of data for rigorously selecting between individual parameterisations and implementation of many of these models may currently be computationally prohibitive^{20,21}. Some large-scale biogeochemical models have employed a degree of stoichiometric plasticity, particularly with regards to the micronutrients³⁴⁻³⁸. However these implementations often employ simplified parameterisations rather than fully mechanistic physiological models. The non-mechanistic physiological parameterisations used in the majority of large-scale models can potentially have a significant influence on overall dynamics²¹. For example, the typical fixed-stoichiometry external nutrient growth models may amplify regional differences in productivity and limiting nutrients, while cell quota type models can decouple nutrient uptake and growth, leading to differences in response to limiting nutrients³⁹.

Past changes in upper ocean nutrient cycling.

In addition to the major redox transitions which occurred in ancient oceans⁴⁰⁻⁴⁴, the potential for significant changes in nutrient limitation in an ocean closely resembling the modern condition is illustrated by the paleo-oceanographic record of recent glacial-interglacial cycles. Variations in both the total oceanic inventory and absolute and relative fluxes of nutrients to the euphotic zone likely played a central role in modulating atmospheric CO₂ over these cycles^{45,46}. Proxies of lowered export production⁴⁷⁻⁴⁹ concurrent with N isotope records indicating more complete NO₃⁻ utilization^{47,50} suggest that both polar high-nitrate low-chlorophyll regions moved towards macronutrient limitation during peak glacial periods. Restricted nutrient supply to surface waters in these regions may have resulted from reduced upward transport of macronutrient-rich subsurface waters, increased stratification⁴⁵ and weakening of the Atlantic meridional overturning⁵¹. Additionally, aerosol Fe deposition also increased 2-4 fold during cold climate intervals⁵²⁻⁵⁵, potentially alleviating Fe limitation. The latter process may have resulted in a direct increase in export production in subantarctic waters of the glacial Southern Ocean⁴⁹, while a combination of reduced vertical fluxes and increased Fe inputs could have driven enhanced macronutrient depletion further south^{47,55}. Given the Southern Ocean's role in ventilating the deep ocean and hence the partitioning of nutrients between the remineralised and preformed pools (see main text and below), enhanced surface water macronutrient depletion likely had a significant influence on the carbon cycle, irrespective of any changes in absolute productivity or export^{45,55}.

Changes in the total nutrient inventory are more equivocal. On both glacial-interglacial and sub-millennial timescales^{51,56,57} the volume of oxygen-minimum zones responded to variations in remineralisation rates of sinking organic matter⁵⁸ and thermocline ventilation⁵⁹. Water-column denitrification appears to decrease during cold intervals of greater thermocline oxygenation, reducing the global rate of fixed N loss, potentially allowing the N inventory to grow. However, the ecological competition between diazotrophs and other plankton^{60,61} may have buffered large changes in the N inventory^{59,62}, a supposition supported by N isotope measurements from tropical Atlantic sediments⁶³. Indeed models of sedimentary N isotope records suggest at most a 30% increase in the fixed N inventory during glacial periods⁶⁴. In parallel, changes in water column oxygenation and organic matter sedimentation may have decreased sedimentary P burial, potentially increasing the P inventory^{60,65,66}.

Biological carbon storage: preformed, remineralised and ultimate limiting nutrients

Primary production in the upper ocean consumes both nutrients and dissolved inorganic carbon (DIC) to produce organic material which can subsequently sink⁶⁷ and be remineralised back to DIC at depth. The resulting downward flux of C is termed the ‘biological (soft-tissue) pump’⁶⁸. The biological pump removes phototrophically derived organic matter from the upper ocean and stores the associated C (DIC_{soft}) out of contact from the atmosphere in the deep ocean. The removal of this C from the upper ocean decreases surface water pCO_2 and draws CO_2 from the atmosphere into the ocean, thereby lowering atmospheric pCO_2 . However, at steady state, the downward flux of organic carbon and nutrients must be balanced by a physical return flux of both DIC_{soft} and inorganic nutrients back to the surface (Supplementary Fig. S4).

The efficiency of the biological pump can be conceptually understood in terms of the relative fraction of nutrients within the ocean interior which result from either remineralisation of organic material or which are transported to depth through physical processes⁶⁹⁻⁷¹. The former, termed ‘remineralised nutrients’ ($\text{Nut}_{\text{remin}}$) will be associated with a flux of carbon through the biological pump (Supplementary Fig. S4), while the latter, termed ‘preformed nutrients’⁷² (Nut_{pre}) are not associated with any biological carbon flux. The existence of preformed (physically transported) nutrients within the ocean interior represents an inefficiency in the biological pump^{45,69-71}. i.e. if these nutrients had instead arrived in the interior via a biological pathway they would be associated with biological C and hence the storage of DIC_{soft} in the deep ocean would be increased.

Vertical physical transports of water, nutrients and carbon within the ocean are controlled by the large scale ‘overturning’ circulation. The interior of the modern ocean is filled with dense so called ‘deep’ waters, formed through buoyancy loss either in the high latitude North Atlantic, or the Southern Ocean (Supplementary Fig. S4). Within the modern ocean, (macro-)nutrient drawdown in surface waters is near complete throughout the majority of the low latitudes, whereas surface macro-nutrients remain high in the Southern Ocean (Fig. 3). Consequently, the deep waters formed in the North Atlantic (NADW), which are fed by waters passing through the low latitudes, have a lower concentration of (macro-) Nut_{pre} than those formed in the Southern Ocean. Although a simplification, the nutrient biogeochemistry of the modern ocean can be conceptually separated into two loops, a low-latitude ‘northern’ pathway, with a high biological pump efficiency, and a southern pathway with low efficiency^{69,71} (Supplementary Fig. S4). Perturbations to surface nutrient cycling and their impact on ocean C storage and atmospheric pCO_2 can then be interpreted in terms of their influence on these two pathways^{71,73}. For example, an increase in nutrient utilization in the (low biological efficiency) Antarctic will drawdown atmospheric pCO_2 more effectively than equivalent surface nutrient changes in the Subantarctic, at low latitudes or the North Atlantic^{74,75}.

The strength of the biological pump is directly related to the stoichiometry with which C is coupled to the nutrients in sinking organic material (Supplementary Fig. S4). The conceptual model described above is easily understood in an ocean with only one nutrient, or equivalently where all nutrients are cycled through the entire system in a strictly conserved stoichiometry (i.e. in this scenario ‘N’ in Supplementary Fig. S4, might be considered to be ‘any Nutrient’⁴⁶). However the reality is clearly more complex, with phytoplankton capable of taking up nutrients and

hence forming organic matter with variable stoichiometries. The cellular ratio of Fe:C for example varies over 2 orders of magnitude (Fig. 1a, Supplementary Fig. S1). The remineralisation process may also alter the stoichiometry of organic material^{15,16,18,19}. Additionally, deep ocean cycling and transport of many of the trace metal nutrients, including Fe, is highly influenced by processes other than those associated with organic matter production and remineralisation, including binding to organic ligands and scavenging onto colloids and larger particles^{6,76}. Indeed the short residence times of the scavenged elements (e.g. Fe, Mn, Co) (Fig. 1a, Supplementary Table 1) result in the total oceanic pool being lower than what might be maintained from remineralisation of 'typical' organic material produced in the surface at steady state (Supplementary Table 1), seriously complicating the preformed/remineralised concept for these nutrients.

Partitioning into preformed and remineralised pools is also only a useful indicator of the efficiency of the biological pump when considering those elements for which biological activity has a significant role to play in their distribution and whose availability potentially limits the production of organic material in the surface ocean. For example, the distribution of S, K, Mg and Ca is influenced little by biological activity, with <<0.1% of the total inventory of these nutrients being in the remineralised pool (Fig. 1a, Supplementary Table 1). Consequently the concept of preformed and remineralised nutrients is primarily useful when considering the macronutrients N and P.

Due to the longer residence time (Fig. 1a) and lack of major biological source/sinks to the ocean, the total P inventory is often considered to set an upper bound on oceanic productivity^{60,61} which, over suitable timescales⁴⁶, might best be described in terms of the oceanic storage of carbon by the biological pump. However there are several reasons why this upper bound may not be reached. As discussed above, incomplete consumption of macronutrients in high-nitrate low-chlorophyll regions (Fig. 3) and in particular the Southern Ocean, currently results in biological carbon storage falling well short (by ~half) of what could be supported by the total P inventory, i.e. the preformed P pool (P_{pre}) is ~50% of the total (P_{tot})^{77,78}. Additionally, N is currently more fully depleted⁷⁹ and more limiting than P (Fig. 3) in the majority of the low latitude regions constituting the high efficiency 'loop' (Supplementary Fig. S4). Subsequently we might expect that the preformed N inventory would be lower than the stoichiometrically equivalent preformed P inventory (Supplementary Table 1). Although estimated ratios of P_{pre}/P_{total} and N_{pre}/N_{total} support this suggestion (Supplementary Table 1), it should be noted that the calculations are themselves dependent on: the ratios of O₂ consumption during N and P remineralisation being stoichiometrically conserved^{80,81}, the amount of remineralisation which occurs anaerobically, and an assumption of complete oxygen saturation at the sea surface. The potential for differences between P_{pre}/P_{total} and N_{pre}/N_{total} will reflect factors influencing the efficiency of the feedbacks which are thought to couple the N and P cycles^{46,82,83}, including, for example, the potential for Fe limitation of diazotrophy^{46,83-85}. Finally the C:N ratio appears to be less flexible than the C:P ratio²⁸ (Fig. 1, Supplementary Fig. S1), with the consequence that biological carbon storage would be expected to be more tightly coupled to the biological cycling of N than P.

A number of implications follow from the preceding arguments. Firstly, if the C:N ratio of organic matter production remains constant, the remineralised N inventory should represent both the principal control on, and best indicator of, the capacity for oceanic biological carbon storage. Secondly, carbon storage is likely to be most sensitive to changes in the C:Nutrient ratio of organic matter for the most

limiting nutrient(s), which for the modern ocean appear to be N in the efficient low latitude loop and Fe in the inefficient high latitude loop (Fig. 3). Consequently, potential changes in the average C:N ratio of low latitude organic matter production and/or subsequent processing can have a significant influence on air-sea CO₂ partitioning^{86,87}. Finally, rather than discussing the identity of an ultimate limiting nutrient^{61,82,83,88}, it may be more useful to consider how processes interact to shift control of the oceanic carbon storage from one element to another over variable time and space scales. For example, no single elemental inventory appears to set a realised upper bound on biological carbon storage in the modern ocean. Rather the influence of oceanic biota on the carbon cycle is dependent on interactions and feedbacks between the cycles of different nutrients in the context of large-scale circulation patterns, which will themselves be variable over geological time.

Derivation of Equation 1, Box 2.

Diffusive flux is governed by Ficks law:

$$J = -D\nabla S, \quad (1)$$

where J is flux of a substance per unit area down the concentration gradient ∇S , and D is the diffusion constant for the substance in seawater.

Assuming a spherical cell of radius r_0 , the flux F integrated over any concentric sphere at distance $r \geq r_0$ will be

$$F = 4\pi r^2 D \frac{\partial S}{\partial r} \quad (2).$$

At steady state, F will be independent of r and integration from $r = r_0$ to $r = \infty$ gives:

$$F = 4\pi D r_0 (S_\infty - S_0), \quad (3)$$

where S_0 is the concentration at infinite distance from the cell (= bulk concentration). In the limiting case when the diffusive transport is slow relative to the biological uptake into the cell, the cell can capture all molecules reaching the cell wall, and $S_0 = 0$. The diffusive transport to the cell under diffusion limitation is therefore:

$$F = 4\pi D r_0 S_\infty. \quad (4)$$

Since we are considering nutrient uptake to be the growth-limiting process, the specific growth rate μ of the organism will be the uptake (= F) divided by the minimum requirement of the limiting element to form a new cell Q_{\min} :

$$\mu = \frac{4\pi D r_0}{Q_{\min}} S_\infty. \quad (5)$$

The specific affinity α is defined as the slope of the specific uptake rate as a function of external nutrient at low external nutrient concentrations. Under steady state growth specific uptake rate = specific growth rate so we also have:

$$\mu = \alpha S_\infty. \quad (6)$$

Comparing (5) and (6) gives:

$$\alpha = \frac{4\pi Dr}{Q_{\min}}. \quad (7)$$

Defining σ as the content of limiting element per unit cell volume we have:

$Q_{\min} = \frac{4}{3}\pi r_o^3 \sigma$. Insertion into (7) gives:

$$\alpha = \frac{3D}{\sigma r_o^2} \quad (8).$$

Which is the form often used to argue that efficient osmotrophs should be small. This is because Eqn. 8 suggests that competitive ability under permanently low external nutrient concentration scales with size as r_o^{-2} .

Note that α is conceptually equivalent to a clearance rate: It is the volume of fluid cleared of the limiting nutrient per unit consumer per unit time. Note also that α is equivalent to biomass-specific turnover rate for the free nutrient. This can be derived as follows:

Turnover-time T for the free nutrient is the ratio between concentration S_∞ and uptake rate $V = \alpha S_\infty B$ where B is the biomass of consumers. This gives:

$$T = \frac{S_\infty}{\alpha S_\infty B} \text{ or } \alpha = \frac{T^{-1}}{B}. \quad (9)$$

For limiting elements with a convenient radioactive isotope (e.g. ^{33}P), turnover-time is often easily determined and has been used as an indicator of limitation. As shown by Eqn. 9, α can be used instead as an indicator normalized to consumer biomass and therefore has advantages when comparing nutrient limitation in samples of different biomass.

Calculation of altered external nutrient inputs

Estimates for the magnitudes of altered external fluxes of iron, nitrogen and phosphorus, as shown in Table 1 are described here. Many of these are driven by changes in dust deposition, and therefore these are presented first. Additional input or inventory changes, including the potential influence of expanding oxygen minimum zones⁸⁹ on Fe mobilisation from sediments⁹⁰, remain difficult to estimate.

Desert dust

Desert dust deposition for the historical period (1870-2000) is estimated⁹¹, where the world is divided into 7 different source areas: North Africa, Middle East/Central Asia, East Asia, North America, Australia, South America and South Africa. We use paleo-observations to deduce the time series for each of the source areas, assigning each paleo-observation to one or more source areas, based on a combination of model and observational data for each of the paleo-observations⁹². The only change is to add the observational data for the North African source from a recent study⁹³, which allows us to better constrain the most important source (North Africa), and extend this source strength back to 1870. Thus, based on the paleo-data, we have estimates for desert dust deposition at 1870 (preindustrial) and current climate. These are normalized, by the average over the 1980-2000 time period, so that these are a ratio of the source strength relative to the recent climate. The uncertainty in these estimates is estimated to be 40%⁹¹.

For climate in the future, there is obviously less ability to project accurately, partly because it is unclear what is driving the observed increase across the 20th century. Only a few studies have looked at climate change impacts on dust sources between preindustrial to current, and these suggest either an increase or decrease in dust, depending on the relative roles of land use, carbon dioxide fertilization of terrestrial vegetation and climate change^{94,95}. For the future, estimates based on climate change only indicate large possible increases⁹⁶ to small increases⁹⁷ to decreases⁹⁴. A study looking at desert area changes predicted from the BIOME4 equilibrium vegetation model⁹⁸ driven by multiple models from the Climate Model Intercomparison project (CMIP3) archive suggests that models show a wide range of estimates of future projections, and that the mean of the models for future desert area depends on the size of the carbon dioxide fertilization effect. If carbon dioxide fertilization does indeed allow desert plants to be more able to deal with water stress (e.g. ref 99), then the future will have less desert area (-1.6% in 2080-2100). However, if not, the future will have more desert area by about 8% in 2080-2100. In addition, the impact of land use could be adding 0-50% more dust (e.g. refs 97,100-102). Because of the uncertainties in future estimates described above, we currently assume a constant dust deposition into the future.

Iron deposition estimates

For this study, we assume that iron comprises 3.5% of dust. Assuming heterogeneous amounts of iron in soils, and looking at the impact on deposition suggests that variable iron content is only important near Australia, and can be neglected¹⁰³. Because direct emissions of iron from combustion is much smaller than dust, we can ignore this fraction in calculation of the total iron flux¹⁰⁴.

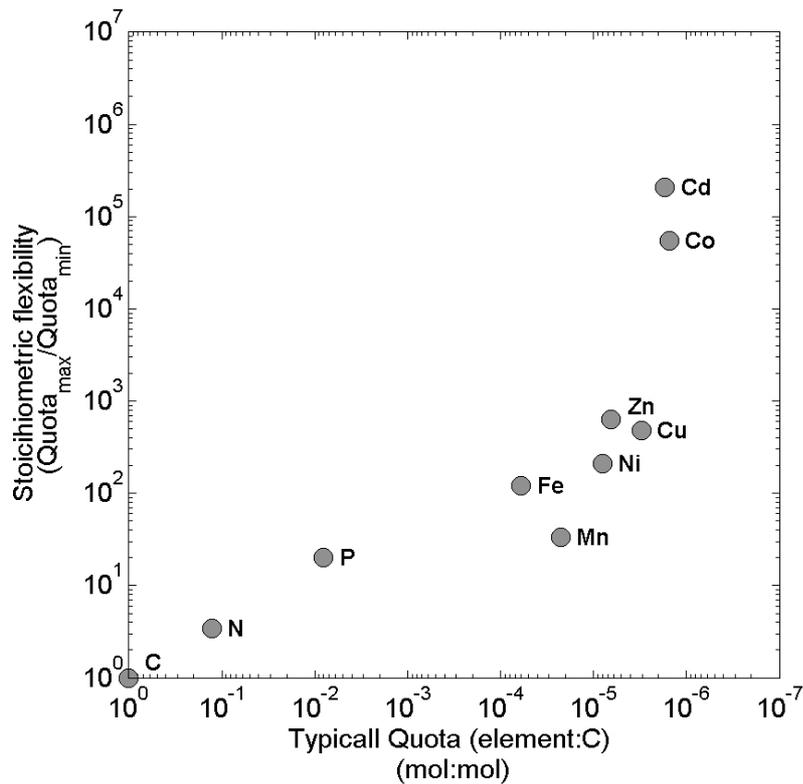
Estimates of soluble iron are difficult to make, because of the uncertainties in exactly what is defined as soluble^{105,106}. It is thought that iron can become more soluble in the atmosphere due to atmospheric processing, especially under acidic environments^{105,107}. There is also mounting evidence for higher solubility of anthropogenic combustion sources of iron¹⁰⁸⁻¹¹². We assume here that soluble iron is Fe(II), and use the model simulations from¹⁰⁴ and¹¹³ to estimate the soluble iron fraction. Since these studies include only simulations of preindustrial and current climate estimates of both atmospheric processing and combustion sources of soluble iron, we propose to use the global average rate of change of sulfur dioxide and black carbon to scale these changes to 1870. Soluble iron for preindustrial and current climates are estimated using a model^{113,104}. In addition, combustion iron sources¹⁰⁴, are scaled using historical estimates of black carbon¹¹⁴.

The uncertainties in the soluble iron deposition changes are larger than in the dust changes, due to unknowns including the sources of soluble iron and the relative roles of atmospheric processing and emissions. The mineralogy of iron sources is likely to be important¹¹⁵, but is neglected here, because there are not yet simulations including these processes. The degree of uncertainty in the temporal evolution of iron deposition should be similar to the uncertainties in dust (40% for past changes, and 100% for future changes).

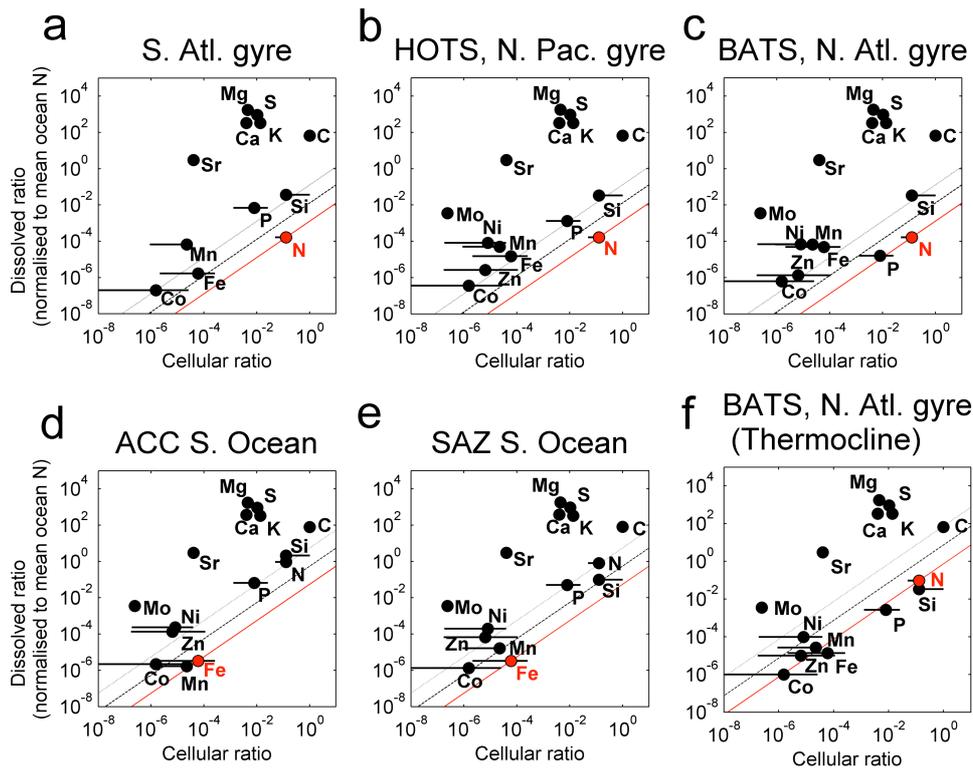
Phosphorus deposition estimates

Phosphorus deposition is also dominated by dust, with other natural sources (primary biogenic particles, sea-salts, volcanoes)¹¹⁶ included in the calculation. In addition emissions of combustion sources of phosphorus are likely to have increased with anthropogenic activity. We estimate the changes from preindustrial to current of

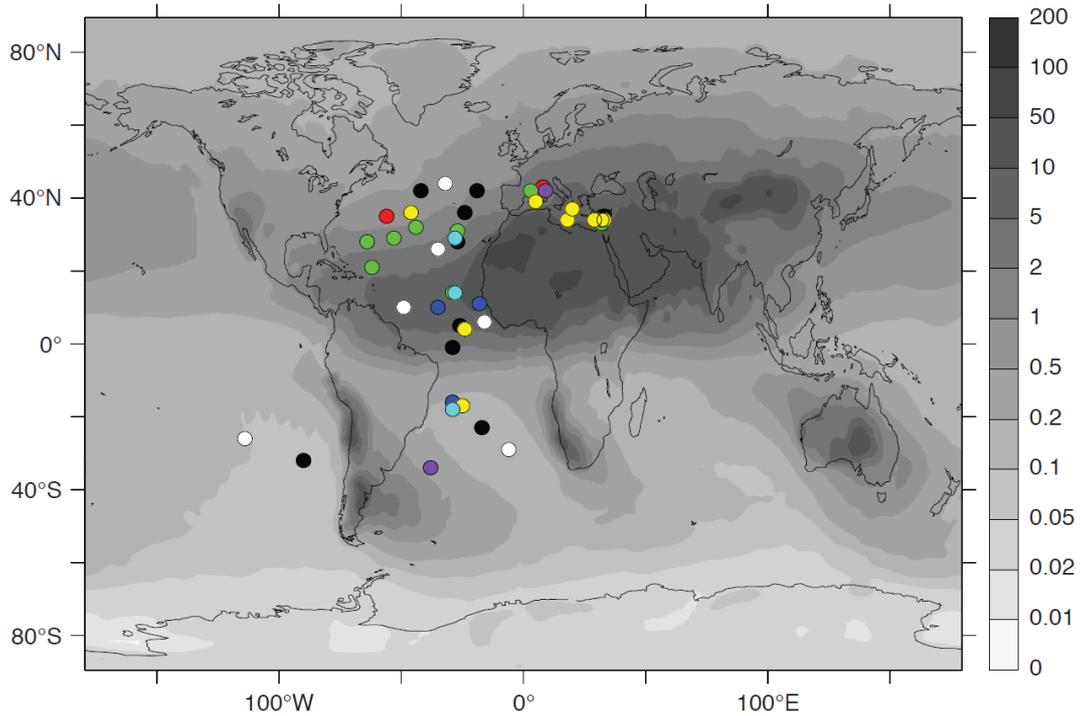
phosphorus deposition from combustion sources scaled by black carbon emissions in preindustrial compared to current¹¹⁶.



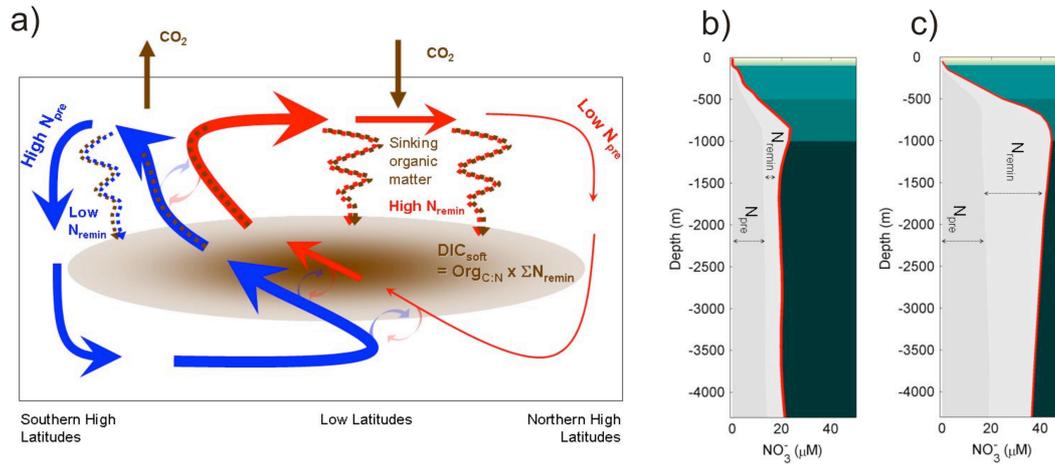
Supplementary Figure S1. Observed range of variability in cellular elemental quotas (normalised to C) as a function of a representative ('typical') quota^{117,118} (see Table S1 for data and references). Quota variability will not only depend on the availability of the nutrient in question, but in many cases the availability of other nutrients and a range of further environmental conditions including irradiance and temperature. Currently available data also necessitates compilation across multiple taxa. Despite such caveats, the broad pattern of increasing observable variability in quota with decreasing cellular requirement currently appears robust. For example, the relationship is similar if the 'typical' quota^{117,118} is replaced by the average of the observed upper and lower values (Table S1).



Supplementary Figure S2. Intercellular nutrient quotas (normalised to C) versus dissolved seawater concentrations (normalised to mean ocean NO_3^-) for five oceanic regions, with data for all available elements indicated and horizontal bars indicating the observed range of quotas (see Fig. 1 and Supplementary Table 1). Data for dissolved nutrients are collated from multiple sources for the South Atlantic gyre^{84,90,119} (~20°S, 25°W), the vicinity of the Hawaii Ocean Time Series¹¹⁹⁻¹²² (HOTS, ~22°N, 158°W), the Bermuda Atlantic Time Series^{15,84,119,122-126} (BATS, ~32°N, 64°W), the Antarctic Circumpolar Current region of the Southern Ocean^{119,127-132} (ACC, S. Ocean, ~66°S) and the Subantarctic Zone of the Southern Ocean^{119,127-132} (SAZ, S. Ocean ~54°S). Data in panels a-e are for the surface layer. For each region the nutrient typically found to promote the primary increase in phytoplankton biomass and/or productivity is indicated in red, with the solid red, dashed and dotted diagonal lines delineating elements which are equally depleted and 10 and 100 fold more replete respectively. For comparison, data are also included from the upper thermocline (~200m) at BATS indicating decoupling between the most deficient nutrients according to dissolved ratios in sub-surface pools (f) and the most deficient and proximal limiting nutrients in surface waters (c), likely due to a combination of additional surface inputs^{126,133,134} and biogeochemical stoichiometric variability^{15,124}. The indicated dissolved seawater concentrations for N and P also do not include significant dissolved organic pools, which will be variably bioavailable⁸⁻¹⁵.



Supplementary Figure S3. In situ responses of microbial groups to dust additions. Symbols indicate locations of enrichment bioassay experiments that displayed positive biological responses to dust amendment superimposed on global total soil dust deposition (mg m^{-2})¹³⁵, with colour indicating the biological group(s) that responded: phytoplankton (black), bacteria (red), diazotrophs, phytoplankton and bacteria (green), phytoplankton and diazotrophs (yellow), bacteria and diazotrophs (cyan), phytoplankton, bacteria, and diazotrophs (purple), no response (white). Not all groups were tested in all experiments.



Supplementary Figure S4. Influence of large scale circulation on the biological pump. a) Schematic circulation diagram showing nutrient transports in the high efficiency (northern) loop (red) and the low efficiency (southern) loop (blue). Nutrients and biologically derived dissolved inorganic carbon (DIC_{soft}) are transported to the surface in the Southern ocean, releasing biologically sequestered CO_2 back to the atmosphere. Surface waters within the Southern Ocean then follow one of two paths⁷³. Waters within the low efficiency loop (blue) sink back into the interior near the Antarctic continent following little (macro-)nutrient drawdown. Waters within the high efficiency loop are transported northwards through low latitude systems supporting significant downward organic carbon and nutrient fluxes, before sinking into the interior within the high latitude Atlantic. The imprint of these large scale circulation patterns can be observed in depth profiles of macronutrients which can be decomposed into the remineralised (N_{remin}) and preformed (N_{pre}) pools using the apparent degree of oxygen utilisation (**b and c**)⁷². Lower concentrations of total and in particular remineralised nutrients are observed close to where sinking occurs in the low latitude loop (profile **b**, North Atlantic) than in the regions nearer the end of this conceptual circulation pathway. Perhaps counter-intuitively, the 3 dimensional circulation of the ocean results in the end of the high efficiency loop corresponding to the North Pacific (**c**)⁴⁵.

Supplementary Table S1. Elemental quotas and oceanic concentrations

Element	Phytoplankton quota ^a (mol:mol C)	Observed maximum phytoplankton quota ^b (mol:mol C)	Observed minimum phytoplankton quota ^b (mol:mol C)	Mean ocean concentration ^c ($\mu\text{mol kg}^{-1}$)	Residence time ^c (years)	Estimated % in preformed pool
C	1	1	1	2247	130000	
H	2.12					
N	0.129	0.169	5×10^{-2}	30	3300	52
O	0.339					
P	8.07×10^{-3}	2.58×10^{-2}	1.29×10^{-3}	2.0	53000	55
S	1.05×10^{-2}			28010	8700000	>99.99
K	1.37×10^{-2}			10205	12000000	>99.98
Mg	4.52×10^{-3}			52664	13000000	>99.99
Ca	4.03×10^{-3}			10279	1000000	>99.99
Sr	4.03×10^{-5}			89.0	5100000	>99.99
Fe	6.05×10^{-5}	2.58×10^{-4}	2.13×10^{-6}	5.37×10^{-4}	350	<0*
Mn	2.26×10^{-5}	3×10^{-5}	9×10^{-7}	3.64×10^{-4}	60	<0*
Ni	8.06×10^{-6}	3.05×10^{-5}	1.86×10^{-7}	8.17×10^{-3}	6000	99
Zn	6.45×10^{-6}	1.1×10^{-4}	1.74×10^{-7}	5.35×10^{-3}	51000	87
Cu	3.06×10^{-6}	1.04×10^{-4}	2.18×10^{-7}	2.36×10^{-3}	5000	86
Cd	1.69×10^{-6}	3.35×10^{-4}	1.6×10^{-9}	6.23×10^{-4}	50000	70
Co	1.53×10^{-6}	2.56×10^{-5}	4.7×10^{-10}	2.04×10^{-5}	340	<0*
Mo	2.42×10^{-7}			0.104	800000	>99.97
Si	0.129	1.01	0.08	99.7	20000	

Notes:

a) Phytoplankton quotas representative of means across a range of taxa^{41,117,118}. The observed composition will depend on the characteristics of the culture media, particularly for trace metals. Further nutrient elements include: Li, B, F, Na, Cl, V, Cr, Se and I.

b) Maximum and minimum observed elemental quotas across different growth conditions and taxa, the latter representing a more restricted set than (a). Maximum and minimum values will typically correspond to nutrient replete or limited cultures respectively and ranges could potentially be extended through observations of other taxa and growth conditions. References are: N ref 28, P refs 25,136, Fe refs 137,138, Mn ref 139, Ni ref 140, Zn refs 141,142, Cu ref 143, Cd refs 141,142, Co ref 144, Si refs 145,146 (diatoms only).

c) Mean ocean concentrations and residence times^{119,147-149}. Global surveys of trace elements (e.g. GEOTRACES¹⁵⁰) should considerably improve these estimates.

d) Values for preformed P were calculated from the difference between the mean concentration and the oceanic mean Apparent Oxygen Utilisation scaled by a Redfield ratio for remineralisation⁸⁰. For the other elements, the remineralised pool is calculated by scaling the value for remineralised P to quotas (a) for the organic material in comparison to mean oceanic concentrations. For N and P a consistent alternative calculation can be performed by assuming the deep ocean is composed of an approximate 50:50 mix of NADW and AABW⁷⁸, then comparing end-member preformed N and P concentrations calculated for these water masses with the mean deep ocean concentration. *calculated values for scavenged elements are negative as the pool size which would result from remineralisation of organic matter would be greater than the total ocean inventory.

Supplementary Table S2. Nutrient addition studies (both in situ and bottle experiments) used to produce Fig. 3. Shown are the location and nutrients primarily and secondarily limiting chlorophyll *a* accumulation and/or carbon fixation increases in the indicated studies.

Study	°N	°E	Primary Limiting Nutrient	Secondary Limiting Nutrient
Al-Qutob et al. 2002 ¹⁵¹	29.0	-34.0	N	P
Berg et al. 2011 ¹⁵² , Hoffmann et al. 2006 ¹⁵³	-50.0	-2.0	Fe	
Bertrand et al. 2007 ¹⁵⁴	-74.3	-179.2	Fe	B ₁₂
Bertrand et al. 2007 ¹⁵⁴	-74.6	-173.2	Fe	
Bertrand et al. 2007 ¹⁵⁴	-76.4	-168.6	Fe	B ₁₂
Blain et al. 2004 ¹⁵⁵	40.0	-18.0	Fe	
Blain et al. 2004 ¹⁵⁵	41.8	-19.4	Fe	
Bonnet et al. 2008 ¹⁵⁶	-9.0	-136.0	Fe	
Bonnet et al. 2008 ¹⁵⁶	-26.0	-114.0	N	
Bonnet et al. 2008 ¹⁵⁶	-32.0	-91.0	Fe,N	
Boyd et al. 2000 ¹⁵⁷	-61.0	-140.0	Fe	
Boyd et al. 2001 ¹⁵⁸	-47.0	38.0	Fe	
Boyd et al. 2005 ¹⁵⁹	50.0	-145.0	Fe	
Buessler et al. 2004 ¹⁶⁰	-66.0	172.0	Fe	
Coale et al. 1996 ¹⁶¹	-5.0	-107.0	Fe	
Coale et al. 2004 ¹⁶²	-56.0	172.0	Fe	
Crawford et al. 2003 ¹⁶³	50.0	165.0	Fe,Zn	Fe
Davey et al. 2008 ¹⁶⁴	10.0	-49.0	N	P
Davey et al. 2008 ¹⁶⁴	5.0	-26.0	N	P
Davey et al. 2008 ¹⁶⁴	6.0	-16.0	N	P
Davey et al. 2008 ¹⁶⁴ , Mills et al. 2004 ¹⁶⁵	10.0	-34.0	N	P
Davey et al. 2008 ¹⁶⁴ , Mills et al. 2004 ¹⁶⁵	4.0	-23.0	N	Fe,P
Davey et al. 2008 ¹⁶⁴ , Mills et al. 2004 ¹⁶⁵	11.0	-17.0	N	Fe,P
Gervais et al. 2002 ¹⁶⁶	-48.0	-21.0	Fe	
Hutchins et al. 1998 ¹⁶⁷	36.0	-122.0	Fe	
Hutchins et al. 1998 ¹⁶⁷	35.9	-122.6	Fe	
Hutchins et al. 2001 ¹⁶⁸	-46.7	-142.0	Fe	Si
Hutchins et al. 2002 ¹⁶⁹	-2.3	-87.5	Fe	
Hutchins et al. 2002 ¹⁶⁹	-9.7	-81.4	Fe	
Martin et al. 1989 ¹⁷⁰	45.0	-142.9	Fe	Co
Martin et al. 1989 ¹⁷⁰	50	-145.0	Fe	Co
Martin et al. 1989 ¹⁷⁰	55.5	-147.5	Fe	Co
Martin et al. 1993 ¹⁷¹	47.0	-20.0	Fe	
Martin et al. 1993 ¹⁷¹	59.5	-20.8	Fe	
Martin et al. 1994 ¹⁷²	-5.0	-90.0	Fe	
Moisander et al. 2011 ¹⁷³	149.8	-17.7	N	
Moisander et al. 2011 ¹⁷³	155.8	-17.5	N	
Moisander et al. 2011 ¹⁷³	158.3	-25.0	N	
Moisander et al. 2011 ¹⁷³	166.2	-23.1	N	P,Fe
Moisander et al. 2011 ¹⁷³	170.0	-15.0	N,Fe	

Moore et al 2007 ¹⁷⁴	-50.0	44.0	Fe	
Moore et al 2007 ¹⁷⁴	-52.0	46.0	Fe	
Moore et al 2007 ¹⁷⁴	-50.5	44.5	Fe	
Moore et al 2007 ¹⁷⁴	-53.0	48.0	Fe	
Moore et al 2007 ¹⁷⁴	-52.0	46.0	Fe	
Moore et al. 2006 ¹⁷⁵	42.0	-42.0	Fe	P,Fe
Moore et al. 2006 ¹⁷⁵	31.0	-27.0	N	P
Moore et al. 2008 ¹⁷⁶	21.0	-62.0	N	P
Moore et al. 2008 ¹⁷⁶	28.0	-64.0	N	P
Moore et al. 2008 ¹⁷⁶	29.0	-53.0	N	P
Moore et al. 2008 ¹⁷⁶	32.0	-44.0	N	P
Moore et al. 2008 ¹⁷⁶	1.0	-27.0	N	P
Moore et al. 2009 ⁸⁴	44.0	-32.0	N	
Moore et al. 2009 ⁸⁴	36.0	-46.0	N	P
Moore et al. 2009 ⁸⁴	28.0	-27.0	N	P
Moore et al. 2009 ⁸⁴	-17.4	-25.0	N	
Moore et al. 2009 ⁸⁴	-23.0	-17.1	N	
Moore et al. 2009 ⁸⁴	-28.5	-5.5	N	
Nielsdottir et al. 2009 ¹⁷⁷	59.2	-19.9	Fe	
Nielsdottir et al. 2009 ¹⁷⁷	58.9	-20.4	Fe	
Nielsdottir et al. 2009 ¹⁷⁷	62.9	-19.6	Fe	
Nielsdottir et al. 2009 ¹⁷⁷	59.7	-18.8	Fe	
Saito et al. 2005 ¹⁷⁸	8.7	-90.6	Fe,Co	
Saito et al. unpubl	-13.3	0.0	N	Co
Saito et al. unpubl	-13.3	0.0	N,Fe	
Sedwick et al. 2002 ¹⁷⁹	-46.0	65.0	Fe	
Sedwick et al. 2002 ¹⁷⁹	-44.2	63.3	Fe	
Sedwick et al. 2002 ¹⁷⁹	-43.3	62.5	Fe	N,P
Suggett et al. 2009 ¹⁸⁰	29.0	34.0	N,P	
Suzuki et al. 2002 ¹⁸¹	50.0	145.0	Fe	
Tsuda et al. 2003 ¹⁸²	49.0	165.0	Fe	
Tsuda et al. 2007 ¹⁸³	48.0	166.0	Fe	
Tanaka et al. 2011 ¹⁸⁴	39.1	5.4	N	P
Tanaka et al. 2011 ¹⁸⁴	34.1	18.5	N	P
Tanaka et al. 2011 ¹⁸⁴	33.6	32.7	N	P

Supplementary Table S3. Dust bioassay studies used to produce Fig. S3. Shown are the locations and microbial groups that responded to nutrient additions. +, -, and 0 indicate positive, negative, or no responses were detected respectively. Where +/- are listed together both positive and negative responses to that microbial group were detected.

Study	°N	°E	Phytoplankton	Bacteria	Diazotroph
Blain et al. 2004 ¹⁵⁵	42	-19	+		
Bonnet et al. 2005 ¹⁸⁵	43	8	+	+	
Bonnet et al. 2008 ¹⁵⁶	-26	-114	0		
Bonnet et al. 2008 ¹⁵⁶	-32	-90	+/-		
Davey et al. 2008 ¹⁶⁴	10	-49	0		
Davey et al. 2008 ¹⁶⁴	5	-26	+		
Davey et al. 2008 ¹⁶⁴	6	-16	0		
Herut et al. 2005 ¹⁸⁶	33	32	+/-	+	
Laghdass et al. 2011 ¹⁸⁷ , Guieu et al. 2010 ¹⁸⁸	42	9	+		
Marañón et al. 2010 ¹⁸⁹	26	-35	-	0	0
Marañón et al. 2010 ¹⁸⁹	14	-28	+/-	+	0
Marañón et al. 2010 ¹⁸⁹	-18	-29	-	+	+
Marañón et al. 2010 ¹⁸⁹	-34	-38	+/-	-	+
Marañón et al. 2010 ¹⁸⁹	-16	-29	-	+	0
Marañón et al. 2010 ¹⁸⁹	-1	-29	+	-	0
Marañón et al. 2010 ¹⁸⁹	14	-29	+/-	+	0
Marañón et al. 2010 ¹⁸⁹	29	-28	-	+	+
Mills and Moore, unpubl	44	-32	0		
Mills and Moore, unpubl	36	-46	+		+
Mills and Moore, unpubl	28	-27	+		0
Mills and Moore, unpubl	-17	-25	+		+
Mills and Moore, unpubl	-23	-17	+		0
Mills and Moore, unpubl	-29	-6	0		0
Mills and Moore, unpubl	21	-62	+	+	
Mills and Moore, unpubl	28	-64	+	+	
Mills and Moore, unpubl	29	-53	+	+	
Mills and Moore, unpubl	31	-27	+	+	
Mills and Moore, unpubl	32	-44	+	+	
Mills and Moore, unpubl	36	-24	+	0	
Mills and Moore, unpubl	35	-56	0	+	
Mills and Moore, unpubl	42	-42	+	0	
Mills et al. 2004 ¹⁶⁵	10	-35	0		+
Mills et al. 2004 ¹⁶⁵	4	-24	+		+
Mills et al. 2004 ¹⁶⁵	11	-18	0		+
Pulido-Villena et al. 2008 ¹⁹⁰	43	8	+	+	
Ternon et al. 2011 ¹⁹¹	34	18	+		+
Ternon et al. 2011 ¹⁹¹	34	33	+		+
Ternon et al. 2011 ¹⁹¹	37	20	+		+
Ternon et al. 2011 ¹⁹¹	39	5	+		+

References

- 1 Tripp, H.J. *et al.*, SAR11 marine bacteria require exogenous reduced sulphur
for growth. *Nature* **452**, 741-744 (2008).
- 2 Canfield, D.E., Glazer, A.N., & Falkowski, P.G., The Evolution and Future of
Earth's Nitrogen Cycle. *Science* **330**, 192-196 (2010).
- 3 Jickells, T.D. *et al.*, Global iron connections between desert dust, ocean
biogeochemistry, and climate. *Science* **308**, 67-71 (2005).
- 4 Mahowald, N.M. *et al.*, Atmospheric Iron Deposition: Global Distribution,
Variability, and Human Perturbations in *Annual Review of Marine Science*
(Annual Reviews, Palo Alto, 2009), Vol. 1, pp. 245-278.
- 5 Schroth, A.W., Crusius, J., Sholkovitz, E.R., & Bostick, B.C., Iron solubility
driven by speciation in dust sources to the ocean. *Nat. Geosci.* **2**, 337-340
(2009).
- 6 Morel, F.M.M., Milligan, A.J., & Saito, M.A., Marine Bioinorganic
Chemistry: The Role of Trace Metals in the Ocean Cycles of Major Nutrients.
Treatise on Geochemistry **6**, 113-143 (2003).
- 7 Jackson, G.A. & Williams, P.M., Importance of dissolved organic nitrogen
and phosphorus to biological nutrient cycling. *Deep-Sea Research Part a-
Oceanographic Research Papers* **32**, 223-235 (1985).
- 8 Hansell, D.A. & Carlson, C.A., Biogeochemistry of Marine Dissolved Organic
Matter. *Academic Press, San Diego* (2002).
- 9 Dyhrman, S.T. *et al.*, Phosphonate utilization by the globally important marine
diazotroph *Trichodesmium*. *Nature* **439**, 68-71 (2006).
- 10 Dyhrman, S.T., Webb, E.A., Anderson, D.M., Moffett, J.W., & Waterbury,
J.B., Cell-specific detection of phosphorus stress in *Trichodesmium* from the
western north Atlantic. *Limnology and Oceanography* **47**, 1832-1836 (2002).
- 11 Zubkov, M.V., Fuchs, B.M., Tarran, G.A., Burkill, P.H., & Amann, R., High
rate of uptake of organic nitrogen compounds by *Prochlorococcus*
cyanobacteria as a key to their dominance in oligotrophic oceanic waters.
Applied and Environmental Microbiology **69**, 1299-1304 (2003).
- 12 Zubkov, M.V. *et al.*, Light enhanced amino acid uptake by dominant
bacterioplankton groups in surface waters of the Atlantic Ocean. *Fems
Microbiology Ecology* **63**, 36-45 (2008).
- 13 Michelou, V.K., Lomas, M.W., & Kirchman, D.L., Phosphate and adenosine-5
'-triphosphate uptake by cyanobacteria and heterotrophic bacteria in the
Sargasso Sea. *Limnology and Oceanography* **56**, 323-332.
- 14 Casey, J.R. *et al.*, Phytoplankton taxon-specific orthophosphate (Pi) and ATP
utilization in the western subtropical North Atlantic. *Aquatic Microbial
Ecology* **58**, 31-44 (2009).
- 15 Wu, J., Sunda, W., Boyle, E.A., & Karl, D.M., Phosphate depletion in the
Western North Atlantic Ocean. *Science* **289**, 759-762 (2000).
- 16 Smith, S.V., Kimmerer, W.J., & Walsh, T.W., Vertical flux and
biogeochemical turnover regulate nutrient limitation of net organic production
in the North Pacific gyre. *Limnology and Oceanography* **31**, 161-167 (1986).
- 17 Bronk, D.A., See, J.H., Bradley, P., & Killberg, L., DON as a source of
bioavailable nitrogen for phytoplankton. *Biogeosciences* **4**, 283-296 (2007).
- 18 Clark, L.L., Ingall, E.D., & Benner, R., Marine phosphorus is selectively
remobilized. *Nature* **393**, 426-426 (1998).

- 19 Hopkinson, C.S. & Vallino, J.J., Efficient export of carbon to the deep ocean
20 through dissolved organic matter. *Nature* **433**, 142-145 (2005).
- 21 Follows, M.J. & Dutkiewicz, S., Modeling Diverse Communities of Marine
22 Microbes in *Annual Review of Marine Science, Vol 3*, edited by C. A. Carlson
& S. J. Giovannoni (Annual Reviews, Palo Alto, 2011), Vol. 3, pp. 427-451.
- 23 Flynn, K.J., Ecological modelling in a sea of variable stoichiometry:
24 Dysfunctionality and the legacy of Redfield and Monod. *Progress in
Oceanography* **84**, 52-65 (2010).
- 25 Monod, J., The growth of bacterial cultures. *Annual Reviews of Microbiology*
26 **3**, 371-394 (1949).
- 27 Droop, M.R., Some thoughts on nutrient limitation in algae. *Journal of
Phycology* **9**, 264-272 (1973).
- 28 Droop, M.R., Vitamin B12 and marine ecology. IV The kinetics of uptake,
29 growth and inhibition in *Monochrysis lutheri*. *Journal of the Marine Biological
Association of the UK* **48**, 689-733 (1968).
- 30 Rhee, G.Y., Effects of N-P atomic ratios and nitrate limitation on algal growth,
31 cell composition and nitrate uptake. *Limnology and Oceanography* **23**, 10-25
(1978).
- 32 Goldman, J.C., McCarthy, J.J., & Peavey, D.G., Growth rate influence on the
33 chemical composition of phytoplankton in oceanic waters. *Nature* **279**, 210-
215 (1979).
- 34 Sterner, R.W. & Elser, J.J., *Ecological Stoichiometry: The Biology of Elements
from Molecules to the Biosphere*. (Princeton Univ. Press, Princeton, 2002).
- 35 Geider, R.J. & La Roche, J., Redfield revisited: variability of C : N : P in
36 marine microalgae and its biochemical basis. *Eur. J. Phycol.* **37**, 1-17 (2002).
- 37 Droop, M.R., Vitamin B₁₂ and marine ecology. IV. The kinetics of uptake,
growth and inhibition in *Monochrysis lutheri*. *J. mar. biol. Ass. U.K.* **48**, 689-
733 (1968).
- Droop, M.R., Some thoughts on nutrient limitation in algae. *J. Phycol.* **9**, 264-
272 (1973).
- Geider, R.J., MacIntyre, H.L., & Kana, T.M., A dynamic regulatory model of
phytoplankton acclimation to light, nutrients, and temperature. *Limnology and
Oceanography* **43**, 679-694 (1998).
- Pahlow, M. & Oschlies, A., Chain model of phytoplankton P, N and light
colimitation. *Marine Ecology-Progress Series* **376**, 69-83 (2009).
- Flynn, K.J., A mechanistic model for describing dynamic multi-nutrient, light,
temperature interactions in phytoplankton. *Journal of Plankton Research* **23**,
977-997 (2001).
- Moore, J.K., Doney, S.C., Glover, D.M., & Fung, I.Y., Iron cycling and
nutrient-limitation patterns in surface waters of the World Ocean. *Deep-Sea
Research Part II-Topical Studies in Oceanography* **49**, 463-507 (2002).
- Moore, J.K., Doney, S.C., & Lindsay, K., Upper ocean ecosystem dynamics
and iron cycling in a global three-dimensional model. *Global Biogeochemical
Cycles* **18**, - (2004).
- Aumont, O. & Bopp, L., Globalizing results from ocean in situ iron
fertilization studies. *Global Biogeochemical Cycles* **20**, GB2017,
doi:2010.1029/2005GB002591. (2006).
- Aumont, O., Maier-Reimer, E., Blain, S., & Monfray, P., An ecosystem model
of the global ocean including Fe, Si, P colimitations. *Global Biogeochemical
Cycles* **17**, 26 (2003).

- 38 Sarmiento, J.L., Slater, R.D., Dunne, J., Gnanadesikan, A., & Hiscock, M.R.,
Efficiency of small scale carbon mitigation by patch iron fertilization.
Biogeosciences **7**, 3593-3624 (2010).
- 39 Mourino-Carballido, B., Pahlow, M., & Oschlies, A., High sensitivity of ultra-
oligotrophic marine ecosystems to atmospheric nitrogen deposition.
Geophysical Research Letters **39**, 6 (2012).
- 40 Canfield, D.E., A new model for Proterozoic ocean chemistry. *Nature* **396**,
450-453 (1998).
- 41 Quigg, A. *et al.*, The evolutionary inheritance of elemental stoichiometry in
marine phytoplankton. *Nature* **425**, 291-294 (2003).
- 42 Dupont, C.L., Butcher, A., Valas, R.E., Bourne, P.E., & Caetano-Anolles, G.,
History of biological metal utilization inferred through phylogenomic analysis
of protein structures. *Proc. Natl. Acad. Sci. U. S. A.* **107**, 10567-10572 (2010).
- 43 Dupont, C.L., Yang, S., Palenik, B., & Bourne, P.E., Modern proteomes
contain putative imprints of ancient shifts in trace metal geochemistry. *Proc.*
Natl. Acad. Sci. U. S. A. **103**, 17822-17827 (2006).
- 44 Saito, M.A., Sigman, D.M., & Morel, F.M.M., The bioinorganic chemistry of
the ancient ocean: the co-evolution of cyanobacterial metal requirements and
biogeochemical cycles at the Archean-Proterozoic boundary? *Inorg. Chim.*
Acta **356**, 308-318 (2003).
- 45 Sigman, D.M., Hain, M.P., & Haug, G.H., The polar ocean and glacial cycles
in atmospheric CO₂ concentration. *Nature* **466**, 47-55 (2010).
- 46 Sigman, D.M. & Haug, G.H., Biological pump in the past in *Treatise On*
Geochemistry, edited by H. D. Holland, K. K. Turekian, & H. Elderfield
(Elsevier Sci., New York, 2003).
- 47 Francois, R. *et al.*, Contribution of Southern Ocean surface-water stratification
to low atmospheric CO₂ concentrations during the last glacial period. *Nature*
389, 929-935 (1997).
- 48 Jaccard, S.L. *et al.*, Glacial/interglacial changes in subarctic North Pacific
stratification. *Science* **308**, 1003-1006 (2005).
- 49 Kohfeld, K.E., Le Quere, C., Harrison, S.P., & Anderson, R.F., Role of marine
biology in glacial-interglacial CO₂ cycles. *Science* **308**, 74-78 (2005).
- 50 Galbraith, E.D. *et al.*, Consistent relationship between global climate and
surface nitrate utilization in the western subarctic Pacific throughout the last
500 ka. *Paleoceanography* **23**, 11 (2008).
- 51 Schmittner, A. & Galbraith, E.D., Glacial greenhouse-gas fluctuations
controlled by ocean circulation changes. *Nature* **456**, 373-376 (2008).
- 52 Mahowald, N. *et al.*, Dust sources and deposition during the last glacial
maximum and current climate: A comparison of model results with paleodata
from ice cores and marine sediments. *Journal of Geophysical Research-*
Atmospheres **104**, 15895-15916 (1999).
- 53 Lambert, F. *et al.*, Dust-climate couplings over the past 800,000 years from the
EPICA Dome C ice core. *Nature* **452**, 616-619 (2008).
- 54 Winckler, G., Anderson, R.F., Fleisher, M.Q., Mcgee, D., & Mahowald, N.,
Covariant glacial-interglacial dust fluxes in the equatorial Pacific and
Antarctica. *Science* **320**, 93-96 (2008).
- 55 Martinez-Garcia, A. *et al.*, Southern Ocean dust-climate coupling over the past
four million years. *Nature* **476**, 312-315 (2011).
- 56 Suthhof, A., Ittekkot, V., & Gaye-Haake, B., Millennial-scale oscillation of
denitrification intensity in the Arabian Sea during the late Quaternary and its

- potential influence on atmospheric N₂O and global climate. *Global Biogeochemical Cycles* **15**, 637-649 (2001).
- 57 Thunell, R.C. & Kepple, A.B., Glacial-holocene delta N-15 record from the Gulf of Tehuantepec, Mexico: Implications for denitrification in the eastern equatorial Pacific and changes in atmospheric N₂O. *Global Biogeochemical Cycles* **18**, (2004).
- 58 Matsumoto, K., Biology-mediated temperature control on atmospheric pCO₂ and ocean biogeochemistry. *Geophysical Research Letters* **34**, (2007).
- 59 Galbraith, E.D., Kienast, M., Pedersen, T.F., & Calvert, S.E., Glacial-interglacial modulation of the marine nitrogen cycle by high-latitude O-2 supply to the global thermocline. *Paleoceanography* **19**, (2004).
- 60 Lenton, T.M. & Klausmeier, C.A., Biotic stoichiometric controls on the deep ocean N : P ratio. *Biogeosciences* **4**, 353-367 (2007).
- 61 Tyrrell, T., The relative influences of nitrogen and phosphorus on oceanic primary production. *Nature* **400**, 525-531 (1999).
- 62 Schmittner, A., Galbraith, E.D., Hostetler, S.W., Pedersen, T.F., & Zhang, R., Large fluctuations of dissolved oxygen in the Indian and Pacific oceans during Dansgaard-Oeschger oscillations caused by variations of North Atlantic Deep Water subduction. *Paleoceanography* **22**, (2007).
- 63 Ren, H. *et al.*, Foraminiferal Isotope Evidence of Reduced Nitrogen Fixation in the Ice Age Atlantic Ocean. *Science* **323**, 244-248 (2009).
- 64 Deutsch, C., Sigman, D.M., Thunell, R.C., Meckler, A.N., & Haug, G.H., Isotopic constraints on glacial/interglacial changes in the oceanic nitrogen budget. *Global Biogeochemical Cycles* **18**, (2004).
- 65 Lenton, T.M. & Watson, A.J., Redfield revisited 1. Regulation of nitrate, phosphate, and oxygen in the ocean. *Global Biogeochemical Cycles* **14**, 225-248 (2000).
- 66 Tamburini, F. & Follmi, K.B., Phosphorus burial in the ocean over glacial-interglacial time scales. *Biogeosciences* **6**, 501-513 (2009).
- 67 Eppley, R.W. & Peterson, B.J., Particulate organic matter flux and planktonic new production in the deep ocean. *Nature* **282**, 677-680 (1979).
- 68 Volk, T. & Hoffert, M.I., Ocean carbon pumps: Analysis of relative strengths and efficiencies in ocean driven atmospheric CO₂ changes, in *The Carbon Cycle and Atmospheric CO₂: Natural Variations Archaean to Present*, Geophys. Monogr. Ser., vol. 32, edited by E. T. Sundquist and W. S. Broecker, 99-110, AGU, Washington, D. C. (1985).
- 69 Toggweiler, J.R., Murnane, R., Carson, S., Gnanadesikan, A., & Sarmiento, J.L., Representation of the carbon cycle in box models and GCMs - 2. Organic pump. *Global Biogeochemical Cycles* **17**, 13 (2003).
- 70 Ito, T. & Follows, M.J., Preformed phosphate, soft tissue pump and atmospheric CO₂. *J. Mar. Res.* **63**, 813-839 (2005).
- 71 Marinov, I. *et al.*, Impact of oceanic circulation on biological carbon storage in the ocean and atmospheric pCO₂. *Global Biogeochemical Cycles* **22**, 15 (2008).
- 72 Broecker, W.S., NO a conservative water-mass tracer. *Earth Planet. Sci. Lett.* **23**, 100-107 (1974).
- 73 Marinov, I., Gnanadesikan, A., Toggweiler, J.R., & Sarmiento, J.L., The Southern Ocean biogeochemical divide. *Nature* **441**, 964-967 (2006).

- 74 Sarmiento, J.L. & Orr, J.C., 3-Dimensional simulations of the impact of
Southern-Ocean nutrient depletion on atmospheric CO₂ and ocean chemistry.
Limnology and Oceanography **36**, 1928-1950 (1991).
- 75 Marinov, I., Follows, M., Gnanadesikan, A., Sarmiento, J.L., & Slater, R.D.,
How does ocean biology affect atmospheric pCO₂? Theory and models.
Journal of Geophysical Research-Oceans **113**, 20 (2008).
- 76 Boyd, P.W. & Ellwood, M.J., The biogeochemical cycle of iron in the ocean.
Nat. Geosci. **3**, 675-682 (2010).
- 77 Gnanadesikan, A. & Marinov, I., Export is not enough: nutrient cycling and
carbon sequestration. *Marine Ecology-Progress Series* **364**, 289-294 (2008).
- 78 Broecker, W.S. *et al.*, How much deep water is formed in the Southern Ocean?
Journal of Geophysical Research-Oceans **103**, 15833-15843 (1998).
- 79 Deutsch, C., Sarmiento, J.L., Sigman, D.M., Gruber, N., & Dunne, J.P.,
Spatial coupling of nitrogen inputs and losses in the ocean. *Nature* **445**, 163-
167 (2007).
- 80 Anderson, L.A. & Sarmiento, J.L., Redfield Ratios of Remineralization
Determined by Nutrient Data-Analysis. *Global Biogeochemical Cycles* **8**, 65-
80 (1994).
- 81 Schneider, B., Karstensen, J., Oschlies, A., & Schlitzer, R., Model-based
evaluation of methods to determine C : N and N : P regeneration ratios from
dissolved nutrients. *Global Biogeochemical Cycles* **19**, 18 (2005).
- 82 Redfield, A.C., The biological control of chemical factors in the environment.
American Scientist **46**, 205-221 (1958).
- 83 Falkowski, P.G., Evolution of the nitrogen cycle and its influence on the
biological sequestration of CO₂ in the ocean. *Nature* **387**, 272-275 (1997).
- 84 Moore, C.M. *et al.*, Large-scale distribution of Atlantic nitrogen fixation
controlled by iron availability. *Nat. Geosci.* **2**, 867-871 (2009).
- 85 Moore, J.K. & Doney, S.C., Iron availability limits the ocean nitrogen
inventory stabilizing feedbacks between marine denitrification and nitrogen
fixation. *Global Biogeochemical Cycles* **21**, (2007).
- 86 Oschlies, A., Schulz, K.G., Riebesell, U., & Schmittner, A., Simulated 21st
century's increase in oceanic suboxia by CO₂-enhanced biotic carbon export.
Global Biogeochemical Cycles **22**, 10 (2008).
- 87 Riebesell, U. *et al.*, Enhanced biological carbon consumption in a high CO₂
ocean. *Nature* **450**, 545-U510 (2007).
- 88 Toggweiler, J.R., An ultimate limiting nutrient. *Nature* **400**, 511-512 (1999).
- 89 Stramma, L., Johnson, G.C., Sprintall, J., & Mohrholz, V., Expanding oxygen-
minimum zones in the tropical oceans. *Science* **320**, 655-658 (2008).
- 90 Noble, A.E. *et al.*, Basin-scale inputs of cobalt, iron, and manganese from the
Benguela-Angola front to the South Atlantic Ocean. *Limnology and
Oceanography* **57**, 989-1010 (2012).
- 91 Mahowald, N. *et al.*, Observed 20th century desert dust variability: impact on
climate and biogeochemistry. *Atmospheric Chemistry and Physics* **10**, 10875-
10893 (2010).
- 92 Mahowald, N. Dimethyl Sulfide Oxidation in the Atmosphere.
- 93 Mulitza, S. *et al.*, Increase in African dust flux at the onset of commercial
agriculture in the Sahel region. *Nature* **466**, doi:10.1038/nature09213, 09226-
09228 (2010).
- 94 Mahowald, N.M. & Luo, C., A less dusty future? *Geophysical Research
Letters* **30**, 1903, doi: 1910.1029/2003GRL017880 (2003).

- 95 Mahowald, N. *et al.*, Change in atmospheric mineral aerosols in response to
climate: last glacial period, pre-industrial, modern and doubled-carbon dioxide
climates *Journal of Geophysical Research* **111**, D10202,
doi:10.1029/12005JD006653 (2006).
- 96 Woodward, S., Roberts, D., & Betts, R., A simulation of the effect of climate
changed-induced desertification on mineral dust aerosol. *Geophysical
Research Letters* **32**, L18810, doi:18810.11029/12005GL023482 (2005).
- 97 Tegen, I., Werner, M., Harrison, S.P., & Kohfeld, K.E., Relative importance of
climate and land use in determining present and future global soil dust
emission. *Geophysical Research Letters* **31**, L05105,
doi:05110.01029/02003GL019216, 012004 (2004).
- 98 Kaplan, J. *et al.*, Climate change and Arctic ecosystems: 2: Modeling,
paleodata-model comparisons and future projections. *Journal of Geophysical
Research* **108**, doi:10.1029/2002JD002559 (2003).
- 99 Smith, S. *et al.*, Elevated CO₂ increases productivity and invasive species
success in an arid ecosystem. *Nature* **408**, 79-82 (2000).
- 100 Prospero, J., Ginoux, P., Torres, O., Nicholson, S., & Gill, T., Environmental
Characterization of Global sources of atmospheric soil dust derived from the
NIMBUS-7 TOMS absorbing aerosol product. *Reviews of Geophysics* **40**,
1002, doi:10.1029/2000RG000095 (2002).
- 101 Luo, C., Mahowald, N., & Corral, J.d., Sensitivity study of meteorological
parameters on mineral aerosol mobilization, transport and idistribution. *Journal
of Geophysical Research* **108**, 4447, 4410.1029/2003JD0003483 (2003).
- 102 Mahowald, N., Rivera, G., & Luo, C., Comment on "Relative importance of
climate and land use in determining present and future global soil dust
emission". *Geophysical Research Letters* **31**, L24105,
24110.21029/22004GL021272 (2004).
- 103 Hand, J. *et al.*, Estimates of soluble iron from observations and a global
mineral aerosol model: Biogeochemical implications. *Journal of Geophysical
Research* **109**, D17205, 17210.11029/12004JD004574 (2004).
- 104 Luo, C. *et al.*, Combustion iron distribution and deposition. *Global
Biogeochemical Cycles* **22**, doi:10.1029/2007GB002964 (2008).
- 105 Jickells, T. & Spokes, L., Atmospheric iron inputs to the oceans in
Biogeochemistry of iron in seawater, edited by D. R. Turner & K. Huntger
(John Wiley and Sons, Ltd, Chichester, 2001), Vol. 7, pp. 85-121.
- 106 Raiswell, R. & Canfield, D.E., The Iron Biogeochemical Cycle Past and
Present Geochemical Perspectives *Geochemical Perspectives* **1**, 1-220 (2012).
- 107 Meskhidze, N., Chameides, W., & Nenes, A., Dust and pollution: A recipe for
enhanced ocean fertilization? *Journal of Geophysical Research* **110**,
doi:10.1029/2004JD005082 (2005).
- 108 Guieu, C., Bonnet, S., Wagener, T., & Loye-Pilot, M.-D., Biomass Burning as
a source of dissolved iron to the open ocean? *Geophysical Research Letters*
22, L19608; doi:19610.11029/12005GL022962 (2005).
- 109 Chuang, P., Duvall, R., Shafer, M., & Schauer, J., The origin of water soluble
particulate iron in the Asian atmospheric outflow. *Geophysical Research
Letters* **32**, doi:10.1029/2004GL021946 (2005).
- 110 Sedwick, P., Sholkovitz, E., & Church, T., Impact of anthropogenic
combustion emissions on the fractional solubility of aerosol iron: Evidence
from the Sargasso Sea. *Geochemistry, Geophysics and Geosystems* **8**,
doi:10.1029/2007GC001586 (2007).

- 111 Sholkovitz, E.R., Sedwick, P.N., Church, T.M., Baker, A.R., & Powell, C.F.,
Fractional solubility of aerosol iron: Synthesis of a global-scale data set.
Geochimica Et Cosmochimica Acta **89**, 173-189 (2012).
- 112 Sholkovitz, E.R., Sedwick, P.N., & Church, T.M., Influence of anthropogenic
combustion emissions on the deposition of soluble aerosol iron to the ocean:
Empirical estimates for island sites in the North Atlantic. *Geochimica Et
Cosmochimica Acta* **73**, 3981-4003 (2009).
- 113 Mahowald, N. *et al.*, Atmospheric Iron deposition: Global distribution,
variability and human perturbations. *Annual Reviews of Marine Sciences* **1**,
245-278, doi:210.1146/annurev/marine.010908.163727 (2009).
- 114 Lamarque, J.-F. *et al.*, Historical (1850-200) gridded anthropogenic and
biomass burning emissions of reactive gases and aerosols: methodology and
application. *Atmospheric Chemistry and Physics* **10**, 7017-7039 (2010).
- 115 Journet, E., Desbouefs, K., Caquineau, S., & Colin, J.-L., Mineralogy as a
critical factor of dust iron solubility. *Geophysical Research Letters* **35**,
doi:10.1029/2007GL031589 (2008).
- 116 Mahowald, N., Jickells, T.D., & Alex R. Baker, P.A., Claudia R. Benitez-
Nelson, Gilles Bergametti, Tami C. Bond, Ying Chen, David D. Cohen, Barak
Herut, Nilgun Kubilay, Remi Losno, Chao Luo, Willy Maenhaut, Kenneth A.
McGee, Gregory S. Okin, Ronald L. Siefert, Seigen Tsukuda, The global
distribution of atmospheric phosphorus deposition and anthropogenic impacts.
Global Biogeochemical Cycles **22**, doi:10.1029/2008GB003240 (2008).
- 117 Ho, T.Y. *et al.*, The elemental composition of some marine phytoplankton.
Journal of Phycology **39**, 1145-1159 (2003).
- 118 Quigg, A., Irwin, A.J., & Finkel, Z.V., Evolutionary inheritance of elemental
stoichiometry in phytoplankton. *Proceedings of the Royal Society B* **278**, 526-
534 (2011).
- 119 Whitfield, M., Interactions between phytoplankton and trace metals in the
ocean in *Advances in Marine Biology, Vol 41* (2001), Vol. 41, pp. 1-128.
- 120 Noble, A.E., Saito, M.A., Maiti, K., & Benitez-Nelson, C.R., Cobalt,
manganese, and iron near the Hawaiian Islands: A potential concentrating
mechanism for cobalt within a cyclonic eddy and implications for the hybrid-
type trace metals. *Deep-Sea Research Part II-Topical Studies in
Oceanography* **55**, 1473-1490 (2008).
- 121 Boyle, E.A., Bergquist, B.A., Kayser, R.A., & Mahowald, N., Iron,
manganese, and lead at Hawaii Ocean Time-series station ALOHA: Temporal
variability and an intermediate water hydrothermal plume (vol 69, pg 933,
2005). *Geochimica Et Cosmochimica Acta* **69**, 5165-5166 (2005).
- 122 Bruland, K.W. & Franks, R.P., Mn, Ni, Cu, Zn and Cd in the western North
Atlantic. In, Trace Metals in Seawater, C. S. Wong, E. Boyle, K. W. Bruland,
J. D. Burton and E. D. Goldberg, eds., Plenum, 395-14. (1983).
- 123 Saito, M.A. & Moffett, J.W., Temporal and spatial variability of cobalt in the
Atlantic Ocean. *Geochimica Et Cosmochimica Acta* **66**, 1943-1953 (2002).
- 124 Moore, C.M. *et al.*, Relative influence of nitrogen and phosphorus availability
on phytoplankton physiology and productivity in the oligotrophic sub-tropical
North Atlantic Ocean *Limnology and Oceanography* **53**, 291-305 (2008).
- 125 Shiller, A.M., Manganese in surface waters of the Atlantic Ocean.
Geophysical Research Letters **24**, 1495-1498 (1997).

- 126 Sedwick, P.N. *et al.*, Iron in the Sargasso Sea (Bermuda Atlantic Time-series
Study region) during summer: Eolian imprint, spatiotemporal variability, and
ecological implications. *Global Biogeochemical Cycles* **19**, 13 (2005).
- 127 Middag, R., de Baar, H.J.W., Laan, P., Cai, P.H., & van Ooijen, J.C.,
Dissolved manganese in the Atlantic sector of the Southern Ocean. *Deep-Sea
Research Part II-Topical Studies in Oceanography* **58**, 2661-2677 (2011).
- 128 Coale, K.H. *et al.*, Southern ocean iron enrichment experiment: Carbon
cycling in high- and low-Si waters. *Science* **304**, 408-414 (2004).
- 129 Loscher, B.M., Relationships among Ni, Cu, Zn, and major nutrients in the
Southern Ocean. *Marine Chemistry* **67**, 67-102 (1999).
- 130 Bown, J. *et al.*, The biogeochemical cycle of dissolved cobalt in the Atlantic
and the Southern Ocean south off the coast of South Africa. *Marine Chemistry*
126, 193-206 (2011).
- 131 Saito, M.A. *et al.*, A seasonal study of dissolved cobalt in the Ross Sea,
Antarctica: micronutrient behavior, absence of scavenging, and relationships
with Zn, Cd, and P. *Biogeosciences* **7**, 4059-4082 (2011).
- 132 Klunder, M.B., Laan, P., Middag, R., De Baar, H.J.W., & van Ooijen, J.C.,
Dissolved iron in the Southern Ocean (Atlantic sector). *Deep-Sea Research
Part II-Topical Studies in Oceanography* **58**, 2678-2694 (2011).
- 133 Wu, J.F. & Boyle, E., Iron in the Sargasso Sea: Implications for the processes
controlling dissolved Fe distribution in the ocean. *Global Biogeochemical
Cycles* **16**, (2002).
- 134 Moore, C.M. *et al.*, Iron limits primary productivity during spring bloom
development in the central North Atlantic. *Global Change Biology* **12**, 626-
634 (2006).
- 135 Mahowald, N.M. *et al.*, Atmospheric global dust cycle and iron inputs to the
ocean. *Global Biogeochemical Cycles* **19**, - (2005).
- 136 Bertilsson, S., Berglund, O., Karl, D.M., & Chisholm, S.W., Elemental
composition of marine Prochlorococcus and Synechococcus: Implications for
the ecological stoichiometry of the sea. *Limnology and Oceanography* **48**,
1721-1731 (2003).
- 137 Sunda, W.G. & Huntsman, S.A., Iron Uptake and Growth Limitation in
Oceanic and Coastal Phytoplankton. *Marine Chemistry* **50**, 189-206 (1995).
- 138 Sunda, W.G. & Huntsman, S.A., Interrelated influence of iron, light and cell
size on marine phytoplankton growth. *Nature* **390**, 389-392 (1997).
- 139 Sunda, W.G. & Huntsman, S.A., Antagonisms between cadmium and zinc
toxicity and manganese limitation in a coastal diatom. *Limnology and
Oceanography* **41**, 373-387 (1996).
- 140 Egleston, E.S. & Morel, F.M.M., Nickel limitation and zinc toxicity in a urea-
grown diatom. *Limnology and Oceanography* **53**, 2462-2471 (2008).
- 141 Sunda, W.G. & Huntsman, S.A., Control of Cd concentrations in a coastal
diatom by interactions among free ionic Cd, Zn, and Mn in seawater.
Environmental Science & Technology **32**, 2961-2968 (1998).
- 142 Sunda, W.G. & Huntsman, S.A., Effect of Zn, Mn, and Fe on Cd accumulation
in phytoplankton: Implications for oceanic Cd cycling. *Limnology and
Oceanography* **45**, 1501-1516 (2000).
- 143 Sunda, W.G. & Huntsman, S.A., Regulation of Copper Concentration in the
Oceanic Nutricline by Phytoplankton Uptake and Regeneration Cycles.
Limnology and Oceanography **40**, 132-137 (1995).

- 144 Sunda, W.G. & Huntsman, S.A., Cobalt and zinc interreplacement in marine
phytoplankton: Biological and geochemical implications. *Limnology and*
145 *Oceanography* **40**, 1404-1417 (1995).
- 146 Timmermans, K.R., van der Wagt, B., & de Baar, H.J.W., Growth rates, half-
saturation constants, and silicate, nitrate, and phosphate depletion in relation to
iron availability of four large, open-ocean diatoms from the Southern Ocean.
Limnology and Oceanography **49**, 2141-2151 (2004).
- 147 Brzezinski, M.A., The Si-C-N ratio of marine diatoms - Interspecific
variability and the effect of some environmental variables. *Journal of*
Phycology **21**, 347-357 (1985).
- 148 Johnson, K.S., The Periodic Table of Elements in the Ocean
<http://www.mbari.org/chemsensor/about.html>.
- 149 Sarmiento, J.L. & Gruber, N., Ocean Biogeochemical Dynamics. *Princeton*
University Press (2006).
- 150 Nozaki, Y., A fresh look at element distribution in the North Pacific. **EOS**,
221 (1997).
- 151 Henderson, G.M. *et al.*, GEOTRACES - An international study of the global
marine biogeochemical cycles of trace elements and their isotopes. *Chem*
Erde-Geochem. **67**, 85-131 (2007).
- 152 Al-Qutob, M., Hase, C., Tilzer, M., and Lazar, B. Phytoplankton drives nitrite
dynamics in the Gulf of Aqaba, Red Sea. *Mar. Ecol. Prog. Ser.* **239**, 233-239
(2002).
- 153 Berg, G.M., *et al.* Variable C and N assimilation strategies in successive, iron-
induced phytoplankton populations in the Southern Ocean determined from C
and n isotopes. *Global Biogeochemical Cycles* **25**, GB3013,
doi:10.1029/2010GB003824 (2011).
- 154 Hoffmann, L. J., Peeken, I., Lochte, K., Assmy, P., and Veldhuis, M. Different
reactions of Southern Ocean phytoplankton size classes to iron fertilization,
Limnology and Oceanography **51**, 1217-1229 (2006).
- 155 Bertrand, E.M., *et al.* Vitamin B12 and iron co-limitation of phytoplankton
growth in the Ross Sea. *Limnology and Oceanography* **52**, 1079-1093 (2007).
- 156 Blain, S., *et al.* Availability of iron and major nutrients for phytoplankton in
the northeast Atlantic Ocean, *Limnology and Oceanography* **49**, 2095-2104
(2004).
- 157 Bonnet, S., *et al.* Nutrient limitation of primary productivity in the Southeast
Pacific (BIOSCOPE cruise). *Biogeosciences* **5**, 215-225 (2008).
- 158 Boyd, P. W. *et al.* A mesoscale phytoplankton bloom in the polar Southern
Ocean stimulated by iron fertilization. *Nature* **407**, 695-702 (2000).
- 159 Boyd, P. W. & Abraham, E. R. Iron-mediated changes in phytoplankton
photosynthetic competence during SOIREE. *Deep-Sea Res. II* **48**, 2529-50
(2001).
- 160 Boyd, P. W. *et al.* The evolution and termination of an iron-induced mesoscale
bloom in the northeast subarctic Pacific. *Limnology and Oceanography* **50**,
1872-1886 (2005).
- 161 Buesseler, K., Andrews, J. E., Pike, S. M. and Charette, M. A. The effect of
iron fertilization on carbon sequestration in the Southern Ocean, *Science* **304**,
414- 417 (2004).
- 162 Coale, K. H. *et al.* A massive phytoplankton bloom induced by an ecosystem-
scale iron fertilization experiment in the Equatorial Pacific Ocean. *Nature* **383**,
495-501 (1996).

- 162 Coale, K. H. *et al.* Southern Ocean iron enrichment experiment: Carbon
cycling in high- and low-Si waters. *Science* **304**, 408–414 (2004).
- 163 Crawford, D. W., *et al.* Influence of zinc and iron enrichments on
phytoplankton growth in the northeastern subarctic Pacific. *Limnology and
Oceanography* **48**, 1583–1600 (2003).
- 164 Davey M., *et al.* Nutrient limitation of picophytoplankton photosynthesis and
growth in the tropical North Atlantic. *Limnology and Oceanography* **53**, 1722–
1733 (2008).
- 165 Mills M. M., *et al.* Iron and phosphorus co-limit nitrogen fixation in the
Eastern Tropical North Atlantic. *Nature* **429**, 292–294 (2004).
- 166 Gervais, F., Riebesell, U., and Gorbunov, M. Changes in primary productivity
and chlorophyll a in response to iron fertilization in the Southern Polar Frontal
Zone. *Limnology and Oceanography* **47**, 1324–1335 (2002).
- 167 Hutchins, D. A., and Bruland, K. W. Iron-limited diatom growth and Si :N
uptake ratios in a coastal upwelling regime. *Nature* **393**, 561–564 (1998).
- 168 Hutchins, D. A., Campbell, B.J., Cottrell, M.T., Takeda, S. Response of
marine bacterial community composition to iron additions in three iron-limited
regimes. *Limnology and Oceanography* **46**, 1535–1545 (2001).
- 169 Hutchins, D.A., *et al.* Phytoplankton iron limitation in the Humboldt Current
and Peru Upwelling. *Limnology and Oceanography* **47**, 997–1011 (2002).
- 170 Martin, J. H., Gordon, R. M., Fitzwater, S. & Broenkow, W. W. VERTEX:
Phytoplankton/iron studies in the Gulf of Alaska. *Deep Sea Research A.
Oceanographic Research Papers* **36**, 649–680 (1989).
- 171 Martin, J. H., Fitzwater, S. E., Gordon, R.M., Hunter, C.N., Tanner, S.J. Iron,
primary production and carbon-nitrogen flux studies during the JGOFS North
Atlantic Bloom Experiment *Deep-Sea Research II* **40**, 115–134 (1993).
- 172 Martin, J. H., *et al.* Testing the Iron Hypothesis in Ecosystems of the
Equatorial Pacific Ocean. *Nature* **371**, 123–129 (1994).
- 173 Moisander PH, *et al.* Analogous nutrient limitations in unicellular diazotrophs
and Prochlorococcus in the South Pacific Ocean. *ISME J*
doi:10.1038/ismej.2011.152 (2011).
- 174 Moore C. M., *et al.* Iron supply influences spring bloom development in the
central North Atlantic. *Global Change Biology* **12**, 626–634 (2006).
- 175 Moore, C. M., *et al.* Iron–light interactions during the CROZet natural iron
bloom and EXport experiment (CROZEX) I: Phytoplankton growth and
photophysiology. *Deep-Sea Research II* **54**, (18–20), 2045–2065 (2007).
- 176 Moore C. M., *et al.* Relative influence of nitrogen and phosphorus availability
on phytoplankton physiology and productivity in the oligotrophic sub-tropical
North Atlantic Ocean. *Limnology and Oceanography* **53**, 291–305 (2008).
- 177 Nielsdóttir, M. C., *et al.* Iron limitation of the postbloom phytoplankton
communities in the Iceland Basin. *Global Biogeochemical Cycles* **23**, GB3001
(2009).
- 178 Saito, M. A., Rocap, C., and Moffett, J. W. Production of cobalt binding
ligands in a Synechococcus feature at the Costa Rica Upwelling Dome.
Limnology and Oceanography **50**, 279–290 (2005).
- 179 Sedwick, P. N., *et al.* Chemical limitations on phytoplankton production in the
Crozet Basin, Subantarctic Southern Ocean, *Deep-Sea Research II* **49**, 3327–
3349 (2002).
- 180 Suggett D. J. *et al.* Nitrogen and phosphorus limitation of oceanic microbial
growth during spring in the Gulf of Aqaba. *Aquatic Microbial Ecology* **56**,

- 227–239 (2009).
- 181 Suzuki, K., *et al.* East-west gradients in the photosynthetic potential of
phytoplankton and iron concentration in the subarctic Pacific Ocean during
early summer. *Limnology and Oceanography* **47**, 1581-1594 (2002).
- 182 Tsuda, A., *et al.* A mesoscale iron enrichment in the western subarctic Pacific
induces a large centric diatom bloom, *Science* **300**, 958–961 (2003).
- 183 Tsuda, A., *et al.* Evidence for the grazing hypothesis: Grazing reduces
phytoplankton responses of the HNLC ecosystem to iron enrichment in the
western Subarctic Pacific (SEEDS II). *Journal of Oceanography* **63**, 983-994
(2007).
- 184 Tanaka, T., *et al.* Lack of P- limitation of phytoplankton and heterotrophic
prokaryotes in surface waters of three anticyclonic eddies in the stratified
Mediterranean Sea. *Biogeosciences* **8**, 525-539 (2011).
- 185 Bonnet, S., Guieu, C., Chiaverini, J., Ras, J., Stock, A. Impact of atmospheric
inputs on the autotrophic communities in a low nutrient low chlorophyll
system, *Limnology and Oceanography* **50**, 1810–1819 (2005).
- 186 Herut, B., *et al.* Response of East Mediterranean surface water to Sahara dust:
on-board microcosm experiment and field observations. *Deep-Sea Research II*
52, 3024-3040 (2005).
- 187 Laghdass, M., *et al.* Effects of Saharan dust on the microbial community
during a large clean mesocosm experiment in the NW Mediterranean Sea,
Aquatic Microbial Ecology **62**, 201-213 (2011).
- 188 Guieu C, *et al.* Large clean mesocosms and simulated dust deposition: a new
methodology to investigate responses of marine oligotrophic ecosystems to
atmospheric inputs. *Biogeosciences* **7**, 2765–2784 (2010)
- 189 Marañón, E., *et al.* Degree of oligotrophy controls the response of microbial
plankton to Saharan dust. *Limnology and Oceanography* **55**, 2339-2352
(2010).
- 190 Pulido-Villena, E., Rerolle, V. Guieu, C. Transient fertilizing effect of dust in
P-deficient LNLC surface ocean. *Geophysical Research Letters*, **37**, L01603,
doi: 10.1029/2009GL041415 (2010).
- 191 Ternon, E., Guieu, C., L’Helguen, S., Catala, P. Longitudinal variability of the
biogeochemical role of Mediterranean aerosols in the Mediterranean Sea.
Biogeosciences, **8**, 1067-1080 (2011).