Iron in the Sargasso Sea: Implications for the processes controlling dissolved Fe distribution in the ocean

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[1] In the surface water of the winter Sargasso Sea, “dissolved” Fe concentrations (defined as iron that passes through a filter with an 0.4 micrometer diameter pore size) decrease with increasing latitude from 0.8 nM at 26°N to 0.2 nM at 31°N. It does not appear that this pattern can be explained solely by spatial variations of eolian Fe deposition. Vertical mixing, phytoplankton growth and particle scavenging all appear to play important roles in controlling dissolved Fe distributions in the surface water. Concentrations of dissolved Fe in the subsurface waters increase with increasing depth from ~0.2 nM at ~200 m to ~0.6 nM at depths below 1000 m, reflecting an imbalance between Fe removal by particle scavenging and Fe regeneration from microbial decomposition of organic matter sinking from the euphotic zone. Simple model calculations suggest that scavenging loss of Fe and ventilation of low preformed Fe high P water from source region may create deep ocean Fe:P concentration ratios (5 × 10⁻⁴ for the Atlantic and 1.2 × 10⁻⁴ for the Pacific) that are much lower than the ratios of Fe to P regenerated from the organic matter sinking from the euphotic zone (24–74 × 10⁻⁴ for the Atlantic and 11–32 × 10⁻⁴ for the Pacific). We hypothesize that this low Fe:P ratio may be the driving force for Fe limitation of algal growth observed in high nutrient low chlorophyll oceans and that regional changes of Fe:C ratios in organic matter sinking from the euphotic zone may play an important role in regulating dissolved Fe concentrations in the deep waters of the world ocean.

INDEX TERMS: 1030 Geochemistry: Geochemical cycles (0330); 1050 Geochemistry: Marine geochemistry (4835, 4850); 1065 Geochemistry: Trace elements (3670); 3670 Mineralogy and Petrology: Minor and trace element composition; KEYWORDS: Fe deposition, Sargasso Sea, mineralogy, world ocean


1. Introduction

[2] Iron plays an important role in the ocean biological carbon pump. Iron limits algal growth in many ocean regions [Coale et al., 1996; Behrenfeld et al., 1997; Boyd et al., 2000], and may also control the rate of N₂ fixation in the present-day low-latitude oceans [Falkowski, 1997; Wu et al., 2000]. Glacial-interglacial variations of atmospheric CO₂ [Petit et al., 1999] have been attributed to biological utilization of macronutrients in the Southern Ocean [Falkowski, 1997; Martin, 1990; Watson et al., 2000] which may result from the changes in eolian Fe flux to the ocean and hypothesized Fe-driven changes in ocean nitrogen inventory [Falkowski, 1997; Broecker and Henderson, 1998]. Before we can include Fe properly in global biogeochemical models [Archer and Johnson, 2000; Lefevre and Watson, 1999], we must understand the mechanisms controlling oceanic iron concentrations and biological availability in the ocean. [3] Potential Fe sources to the ocean are river discharge and eolian deposition. The later is thought to be the dominant Fe input to the remote ocean [Duce, 1986; Wells et al., 1995]. Fluvial Fe input is substantially attenuated within the coastal zone [Wu and Luther, 1996] because of colloidal agglomeration in the estuarine zone [Boyle et al., 1977] and because of the low Fe solubility in seawater [Wu et al., 2001], dust-borne Fe particles can be transported over long distances through the atmosphere to remote regions of the ocean [Duce and Tindale, 1991]. As eolian particles are deposited into sea surface waters, Fe is partially released into the ambient seawater. The released Fe is then taken up by algae or scavenged by sinking particles, and subsequently exported to subsurface waters. Microbial degradation of sinking organic particles releases the Fe into the deep
water as “dissolved” forms where it resides for \( \sim \)100 years before being removed to deep sea sediments by particle scavenging [Bruland et al., 1994].

[4] In the surface waters of oligotrophic gyres, “dissolved” Fe concentrations (operationally defined as that passing through a polycarbonate filter with a 0.4 micrometer diameter pore size) show temporal and spatial variations in response to stratification conditions [Bruland et al., 1994; Wu and Luther, 1994]. Beneath the surface euphotic zone, dissolved Fe concentrations increase with increasing depth, resembling major nutrients such as nitrate and phosphate [Martin and Gordon, 1988; Wu and Luther, 1994; Martin et al., 1993]. In the deep ocean, dissolved Fe concentrations are higher in the North Atlantic [Wu and Luther, 1994; Wu et al., 2001] but are lower in the North Pacific and the Southern Ocean [Bruland et al., 1994; Johnson et al., 1997; Sohrin et al., 2000; Boye et al., 2001; Measures and Vink, 2001; Wu et al., 2001]. The processes responsible for these distributions are not well understood.

[5] Here we present new data on dissolved Fe in the Sargasso Sea to illuminate the relative importance of processes controlling dissolved Fe distribution. Based on published data from the North Pacific, we propose a simple conceptual model for controls on the Fe distribution in deep waters of the global ocean.

2. Sampling and Analysis
[6] Seawater samples were collected from \( \sim \)0.5 m depth using an underway sampler [Vink et al., 2000] along a transect from 31.67°N, 64.17°W to 26.10°N, 70.00°W in the Sargasso Sea (Oceanus 318 cruise; J. Moffett, Chief Scientist). Vertical profile samples were collected in March 1998 (at 31.7°N, 64.2°W, Oceanus 318), July 1998 (at 34.8°N, 57.8°W, Oceanus 326, L. Keigwin, Chief Scientist), and April 2001 (at 22.8°N, 158.8°W, Wecoma MP2, M. Neumann, Chief Scientist) using our MITESS sampler [Dickey et al., 1998; Bell et al., 2002]. Briefly, this trace-metal clean device is lowered beneath a hydrowire and under program control opens and closes sealed trace-metal clean polyethylene bottles initially filled with distilled water. Less dense distilled water floats out of the bottle and is replaced by denser seawater. Bottles are opened and closed in-situ, so they are immune to contamination from surface operations. Upon returning to surface, samples were handled inside a class-100 clean flow bench on board the ship within a few hours after sample collection. The samples were passed through 0.4 μm Nuclepore filters inside a class-100 clean bench on board the ship. The filtrates were acidified to pH 2.2 with 2 mL of triple-distilled 6N Vycor HCl (blank \( \sim \)1.8 nmol Fe/1L HCl) per 1000 mL sample. Fe in the sample was measured using an isotope dilution-ICPMS method [Wu and Boyle, 1998]. This method uses triplicate 1.3 mL samples and has a detection limit of \( \sim \)0.03 nM. The procedural blank is 0.08 ± 0.01 nM; this blank is mainly due to Fe blanks arising from the ICPMS hardware (nebulizer, spray chamber, sampling and skimming cones, and ion beam focusing hardware), not the reagents or Fe contamination due to sample handling. On a nanomolar basis, this procedural blank can be lowered by 1 order of magnitude by using large sample size (such as 10–50 mL, instead of 1.3 mL). The precision of the method is \( \sim \)1–2% at 1.0 nM. \(^{57}\)Fe spike recovery is >95%. This method detects total Fe in the sample because in addition to Fe forms that equilibrate with the isotope spike, Fe in colloidal particles and organic materials that can be coprecipitated with Mg(OH)\(_2\) can be ionized in the high temperature (~8000 °C) zone of the inductively coupled plasma, and detected by the magnetic sector mass spectrometer. As discussed by Wu et al. [2001], we have evidence that Fe that passes through a 0.4 μm filter contains a significant component of colloidal Fe (0.02–0.4 μm). Thus, the “dissolved” Fe (<0.4 μm) that we report here includes both soluble and colloidal Fe.

[7] Low-level surface water dissolved inorganic phosphorus concentrations were determined using a modified “MAGIC” method [Wu et al., 2000].

3. Results and Discussion
3.1. Surface Water
[8] Dissolved Fe concentrations in the surface waters of the Sargasso Sea increase from \( \sim \)0.2 nM at 31°N to \( \sim \)0.8 nM at 26°N (Figure 1a). This distribution can be attributed to a combined effect of eolian Fe deposition, water mixing, phytoplankton uptake and removal, and passive particle scavenging. Measures et al. [1984] show that there is a 20% decline in surface water Al concentrations going from Barbados to Bermuda. Based on a simple model of aluminum in the mixed layer, they suggest that this decrease is due to a proportional decrease in the eolian Al flux. If the eolian Fe flux scales with the eolian Al flux, a comparable proportional decrease (~20% of 0.8 nM or ~0.16 nM) in Fe would be attributable to the reduced eolian flux. However, the Fe residence time is probably shorter than that of Al, which complicates matters because it may then show seasonal variations of surface water Fe in response to the fluctuation of eolian Fe deposition that are damped for longer residence time Al.

[9] The hydrocast CTD data along the transect indicate that the mixed layer depth increases from \( \sim \)80 m at 26°N to 180 m at 31°N. Because of increased convective mixing to the north, sea surface temperature decreases from ~25 °C at 26°N to ~19.5 °C at 31°N (Figure 1a) and surface water DIP concentrations increase from ~0.4 nM at 26°N to ~3 nM at 31°N (Figure 1b). Thus the observed northward decrease in surface water dissolved Fe concentration (Figure 1a) may be attributed, at least in part, to the greater volume through which the eolian input is mixed northward, because concentrations of dissolved Fe at the base of mixed layer are very low (0.2 nM, Figure 2). If we assume that, before winter time mixing occurs, the water column at 31°N had a homogeneous 80 m mixed layer with dissolved Fe concentrations of 0.64 nM (Fe concentration of 0.8 nM at 26°N corrected for ~20% decline in eolian Fe input along the transect) and if we assume a linear decrease in dissolved Fe concentrations from 0.64 nM at 80 m to 0.2 nM at 180 m, a mixing from the sea surface to 180 m depth will result in a mixed wintertime Fe concentration of 0.52 nM in the
homogenized 180 m water column, accounting for only 
\( \frac{15}{100} \) of the latitudinal dissolved Fe gradient shown in 
Figure 1a. Because this calculated 0.52 nM dissolved Fe 
concentration is 
\( \frac{0.29}{100} \) nM higher than observed at 31°N, we suggest that biological uptake and particle 
scavenging also must play an important role in controlling 
latitudinal changes in dissolved Fe concentrations. 

As the mixed layer depth increases from 80 m at 
26°N to 180 m at 31°N, mixing between low PO4 (0.5 nM) 
surface water and high PO4 (70 nM) water in the upper 
thermocline would increase the wintertime mixed layer PO4 
concentration to \(~38\) nM. We actually observe \(~3\) nM at 
31°N (Figure 1b) and attribute that difference to biological 
uptake followed by subsequent particulate P export into 
water beneath the euphotic zone. The northward increase of 
Chl a (Figure 1b) is consistent with macronutrient concentra-
tion increases to the north. Assuming a Fe:P ratio of 
\( \frac{5.0 \times 10^{-3}}{10^{-3}} \) (as calculated by simple model presented 
below), this biological uptake should decrease surface water 
Fe concentration only by \(~0.19 \pm 0.09\) nM. This decrease in 
combination with the effects of eolian Fe deposition and vertical mixing, should lead to a Fe concentration of 
\(~0.33\) nM at 31°N which is \(~0.13\) nM higher than observed 
(0.2 nM, Figure 1a). The remaining difference between 
calculated and observed Fe could be due to removal of Fe by absorption on sinking particles, although the difference 
is small and also could be due to uncertainties in our 
assumptions.

Our results suggest that eolian deposition is not the 
only factor controlling the dissolved Fe distribution in the 
surface mixed layer. Convective mixing, biological Fe 
uptake and particle scavenging may also play an important 
role in controlling dissolved Fe concentrations in the surface 
mixed layer. Surface stratification conditions have been 
found to strongly affect concentrations of dissolved Fe in 
oligotrophic surface waters [Wu and Luther, 1994; Bruland et al., 1994]. A dissolved Fe maximum that we observed in 
the surface water near Bermuda in summer (Figure 2) may 
result from enhanced accumulation of eolian Fe in shallowed 
mixed layer (although only the single surface water point 
observed at the deep-cast station in July 1998 is shown here, 
12 underway surface samples collected both before and after 
the station site showed an average value of \(0.58 \pm 0.07\) nM, 
confirming the regional and analytical significance of the 
surface datum plotted here). The Fe maximum is not 
observed in late fall through early spring, because the eolian

\[\text{Figure 1. (a) Surface water (~0.5 m) dissolved (<0.4 \ \mu m) iron (crosses) and sea surface temperature (circles) and (b) inorganic phosphate (pluses) and chlorophyll a (squares) versus latitude along a transect from 26.1°N, 70.0°W to 31.7°N, 64.2°W in the Sargasso sea in March 1998. Chl a was measured by K. Cavender-Bares at MIT [Cavenders-Bares, 1999].}\]

\[\text{Figure 2. Vertical profiles of dissolved iron in the Sargasso Sea near Bermuda in March 1998 (at 31.7°N, 64.2°W, crosses) and in July 1998 (at 34.8°N, 57.8°W, squares).}\]
signal is redistributed vertically by enhanced convective mixing and algal uptake following nutrient injections from subsurface waters [Wu and Luther, 1994].

[12] From these observations, one may expect a seasonal cycle of Fe limitation to algal growth in the Sargasso Sea. In summer, high concentrations of dissolved Fe in the surface mixed layer combined with a limited supply of macronutrients from below should lead to a condition where biological growth is not limited by Fe. In contrast, during enhanced mixing events in winter, as macronutrients are injected into the surface, an initial algal growth in the spring can decrease surface water dissolved Fe concentrations to the point where subsequent algal growth is Fe limited. For Fe limitation to occur, however, Fe concentrations in the subsurface waters must be lower than biological Fe requirements of phytoplankton to completely consume upwelled nitrate and phosphate. If the iron: nitrate ratio in the upwelling subsurface water is higher than that required by phytoplankton, Fe would not be depleted before macronutrients N and P are completely consumed and surface water stratification conditions will not affect the Fe status of algal growth. The low Fe to nitrate ratio in upwelling waters is also the root cause of Fe limitation of algal growth in high nutrient low chlorophyll (HNLC) regions.

[13] In HNLC upwelling regions, eolian Fe deposition is exceedingly low [Duce and Tindale, 1991] and the strong upwelling provides ample nitrate and phosphate to the surface water. Because Fe concentrations in upwelled waters are lower than required by phytoplankton if they are to completely consume upwelled nitrate and phosphate, and because eolian Fe deposition in this region is low, the upwelled Fe tends to be depleted before macronutrients N and P are completely consumed [Watson et al., 2000]. Fe limitation thus becomes a persistent feature in these surface waters [Boyd et al., 2000]. In the following section, we will discuss the processes controlling a low ratio of Fe to N and P in oceanic deep waters.

3.2. Deep Water

[14] In the water column of the Sargasso Sea below the base of the wintertime surface mixed layer, dissolved Fe concentrations increase with increasing depth from \( \sim 0.2 \text{nM} \) at \( \sim 200 \text{ m} \) to \( \sim 0.6 \text{nM} \) at depths \( \geq 1000 \text{ m} \) (Figure 2), a value that is about a factor of 2 higher than in the Ross Sea [Sohrin et al., 2000, Boye et al., 2001] and in the central North Pacific [Bruland et al., 1994; Wu et al., 2001].

[15] The downward increase of dissolved Fe concentrations at 200–1000 m is consistent with a nutrient-type behavior for Fe in the ocean. As an essential element for algal growth, labile Fe in the surface water is effectively incorporated into biological tissues. Some of this organic matter sinks through water column and is decomposed by bacteria. Fe from these organic particles is then released into the water column in dissolved forms. If this process exceeds Fe removal by particle scavenging (a process by which dissolved Fe binds to particle surfaces and is removed from the water column as particles sink to the seafloor), dissolved Fe concentrations will increase with increasing depth because deeper water is generally older and accumulates more Fe. In this aspect, the behavior of Fe is similar to that of the macronutrients nitrate and phosphate.

[16] The nutrient type distribution of dissolved Fe in the upper 1000 m suggests that, although eolian deposition is the primary Fe source to the remote ocean [Duce and Tindale, 1991], the microbial decomposition of sinking organic particles transporting that iron may be the major route by which eolian Fe is transported into the deep sea [Bruland et al., 1994]. Laboratory experiments on the dissolution of eolian Fe in seawater indicate that while labile eolian Fe can be released into ambient solution within 1–2 hours after aerosols contact surface water [Zhuang et al., 1990], Fe inside large aerosol particles is not readily soluble and may sink through water column without participating in the biogeochemical cycle. Assuming that the Fe released from microbial decomposition of sinking organic matter is the primary source for dissolved Fe in the deep sea and that particle scavenging is the major Fe removal pathway, we can describe the dissolved Fe distribution in deep ocean using a simple “pipe” model as illustrated in Figure 3. We emphasize that this model is intended to serve as a semiquantitative back-of-the-envelope illustration of the processes involved; a complete model of the oceanic iron cycle would require a more sophisticated approach.

[17] In this “pipe” model, deep water in the North Atlantic is considered to “ventilate” from cold high-latitude North Atlantic on timescales of a century to a millennium. During transit at depth, the deep water receives Fe regenerated from microbial decomposition of organic particles sinking from the euphotic zone. At the same time, Fe is actively scavenged by sinking particles and removed to deep sea sediments. In the Western North Atlantic at 31°N, the effect of Antarctic Intermediate Water and Antarctic Bottom Water (Southern Component Water [Broecker and Peng, 1982]) is relatively weak as compared to the water sinking from high-latitude North Atlantic. Neglecting the effect of Southern Component Water (which represents a small percentage of deep North Atlantic waters), the ventilation process can be quantified by following equation:

\[
\frac{d[Fe]}{dt} = F - k[Fe],
\]

where \( F \) is the rate of Fe regeneration at depth, and \( k \) is the rate constant for \( < 0.4 \text{ µM} \) Fe scavenged by particles in the deep water (0.01/year, which is based on Bruland et al.’s [1994] estimate on the scavenging residence time of 70–140 years for dissolved Fe). The solution of this equation is:

\[
F = \frac{(k[Fe] - k[Fe]_0 e^{-kt})/(1 - e^{-kt})}{},
\]

where \([Fe]_0\) is taken as the concentrations of dissolved Fe below 1000 m at 59°N (0.55 nM [Martin et al., 1993]), \( t \) is the time the water takes to flow from at 59°N to 30°N (\( \sim 50–150 \text{ years} \) as estimated from deep water C\(^{14}\) content [Stuiver et al., 1983]), and \([Fe]_0\) is the concentration of dissolved Fe below 1000 m at 31°N (0.65 nM, Figure 2).

[18] From equation (2), we obtain a value of 6.8–8.0 nM/1000 yr for \( F \). As a model derived parameter, \( F \) represents the rate of dissolved Fe regeneration below 1000 m in the North Atlantic. This value is 2 orders of magnitude lower than total eolian Fe deposition reported for the North Atlantic [Duce and Tindale, 1991], and if biological iron is quantitatively released during regeneration, is consistent
with seawater eolian Fe solubility of 1% reported by Jickells et al. [1998].

To estimate the ratio of Fe vs. C in organic matter exported to deep sea, we divide the Fe regeneration flux $F_{Fe}$ by an oxygen utilization rate (150–460 µM/1000 yr) (In this calculation, a Redfield C:O ratio of 106:165 is assumed for organic matter sinking from the euphotic zone [Takahashi et al., 1985]). Oxygen utilization rate is estimated from the difference of deep water O$_2$ concentrations between 59°N and 30°N (~23 µM) and deep water ventilation time between the two latitudes (~50–150 years)). The calculated Fe:C regeneration ratio of 23–70 × 10$^{-6}$ is within the range of Fe:C ratio for laboratory cultured oceanic diatom T. oceanica growing at 1.5 d$^{-1}$ (Sunda and Huntsman, 1995). For this algal species, the cellular Fe:C ratio increases with increasing growth rate to ~10 × 10$^{-6}$ Fe:C [Sunda and Huntsman, 1995]. Above this value, luxury iron uptake increases cellular Fe:C ratio from 10 × 10$^{-6}$ to 50 × 10$^{-6}$ with a constant maximum growth rate of 1.5 d$^{-1}$. The similarity between the Fe:C regeneration ratio and the Fe:C ratio of oceanic diatom growing at 1.5 d$^{-1}$ can be taken as a hint that Fe export to the deep water “dissolved” Fe pool may be dominated by organic matter produced by phytoplankton growing at their maximum rates, assuming that Fe fractionation from C during regeneration is small.

The Fe:C regeneration ratio that we obtain from above “model” (23–70 × 10$^{-6}$) is 2 orders of magnitude lower than the ratio of total Fe to C flux to deep sea sediment traps deployed in the western North Atlantic (~0.028 [Jickells and Spokes, 2001; Jickells et al., 1998]) and is 1 order of magnitude higher than the deep water Fe:C concentration ratio (0.5 mmol Fe/mol P, equivalent to Fe:C ratios of 4.7 × 10$^{-6}$) as calculated from deep water Fe and P concentrations by assuming a C:P Redfield ratio of 106:1 [Sunda and Huntsman, 1995; Fung et al., 2000]. The difference between the Fe:C concentration ratio and the deep water Fe:C concentration ratio may result mainly from the removal of regenerated Fe from deep sea by particle scavenging [Bruland et al., 1994], and from a low preformed Fe:P ratio (5 × 10$^{-5}$) in source waters [Martin et al., 1993, Broecker and Peng, 1982]. If the model derived Fe:C regeneration ratio (23–70 × 10$^{-6}$) represents the Fe:C ratio in organic matter sinking from the euphotic zone and thus perhaps also the algal Fe:C uptake ratio in the surface water, the 1 order of magnitude lower Fe:C concentration ratios in the thermocline (4.7 × 10$^{-6}$ [Martin and Gordon, 1988]) and deep waters (as compared to the deep water Fe:C regeneration ratio) imply that the recycled Fe from upwelling subsurface waters can support only ~10% of the Fe needed for phytoplankton to deplete upwelled macronutrient. Because the Fe:P ratio in upwelling waters is much lower than the Fe:P algal uptake ratio, when subsurface water mixes to sea surface, algal consumption of macronutrients will require additional Fe. In remote oceans, this additional Fe presumably derives from fresh eolian deposition. However, eolian Fe deposition is episodic and sporadic. Mixing events do not always accompany pulses of dust input. The residence time of eolian Fe in the surface water (days-weeks [Coale et al., 1996; Measures and Vink, 1999]) is too short to maintain high concentrations of dissolved Fe in the surface water for a long time. In this situation, available Fe may limit algal growth and food web structure during intense mixing events. Because highly productive algal blooms during these events are major routes of nutrient transport to the deep sea [Michaelis et al., 1996; Anderson and Sarmento, 1994], Fe availability may limit the export of nutrient and carbon to the deep sea. It has been shown that Fe availability controls diatom shell thickness and hence the export of organic carbon to the deep sea [Hutchins and Bruland, 1998]. By controlling the extent of algal blooms during mixing events, Fe limitation may play an important role in determining the relative recycling of major nutrients between thermocline and deep water and the vertical distribution of these nutrients in the ocean.

3.3. Pacific Ocean

As a particle-reactive element with a residence time of ~100 years in the deep sea [Bruland et al., 1994], “dissolved” Fe should occur at extremely low concentrations in waters older than a few hundred years. If particle scavenging dominates regeneration, “dissolved” Fe concentration in the deep Pacific should be much lower than those of the deep Atlantic. The deep water “dissolved” Fe concentration (~0.3 nM) that we observed at station Aloha in the North Pacific near Hawaii is similar to that reported by Bruland et al. [1994] at the VERTEX-IV site (200 km north of Hawaii) (Figure 4). This value is much lower than those in the North Atlantic (Figure 2) but is larger than the Fe solubility (0.08 nM [Wu et al., 2001]). This value is similar to the “dissolved” Fe observed in the Pacific sector of the Southern Ocean (~0.3 nM [Sohrin et al., 2000; Measures and Vink, 2001]). Although we accept that Fe-organic complexation [Rue and Bruland, 1995; van den Berg, 1995; Wu and Luther, 1995; Boye et al., 2001] plays a key role in maintaining dissolved Fe concentrations in the old deep waters of the North Pacific Ocean [Johnson et al.,
[23] We consider a section from 70°S to 28°N in the Pacific. The deep-water ventilation time in this section is assumed to be 300–700 years [Stuiver et al., 1983]. Dissolved Fe concentrations are ~0.30 nM in deep waters of the Pacific at 70°S and 27°N (Figure 4 [Bruland et al., 1994; Sohrin et al., 2000]). The Fe scavenging rate constant is set at 0.01/year [Bruland et al., 1994]. Using these values in the equation 2, we estimate the rate of dissolved Fe regeneration in the deep Pacific to be 3.0 nM/1000 yr. The ratio of Fe vs. C in organic matter exported to deep waters (10–30 × 10⁻⁶) can then be calculated from the Fe regeneration flux by using the oxygen utilization rate of 150–460 nM/1000 yr [Broecker and Peng, 1982] and the Redfield ratio of 106C:165O [Takahashi et al., 1985].

[24] The Fe:C regeneration ratio of 10–30 × 10⁻⁶ is 1 order of magnitude higher than the Fe:C concentration ratio calculated from Fe and P concentrations in Pacific deep waters between 70°S and 27°N (1.1 × 10⁻⁶), implying that the water upwelled from the subsurface may contain only about 5% of the iron needed for biological depletion of upwelled macronutrients. The relative Fe depletion and the lower eolian Fe deposition in the Pacific readily explains Fe limitation observed in the three HNLC upwelling regions of the Pacific Ocean [Martin and Fitzwater, 1988; Coale et al., 1996; Boyd et al., 2000]. Similar Fe limitation has not been observed in the North Atlantic [Martin et al., 1993], probably because of higher eolian Fe deposition and a higher Fe:P concentration ratio in the upwelling subsurface waters.

[25] These estimates seem to imply that Fe:C regeneration ratio in the Pacific (~10–30 × 10⁻⁶) is lower than in the North Atlantic (~23–70 × 10⁻⁶). The lower ratio in the Pacific is consistent with lower eolian Fe deposition to the South Pacific [Duce and Tindale, 1991], and with the results of laboratory experiments showing that the cellular Fe:C ratio (Fe cell quota) increases with increasing Fe concentration in the culture media [Sunda and Huntsman, 1995]. It appears that changes in the Fe:C ratio of sinking organic matter in response to regional variations of eolian deposition may play an important role in controlling deep water Fe concentrations. This hypothesis remains to be verified in the future by direct measurements of the biological Fe:C ratio in sinking particles collected from areas of diverse eolian Fe flux.

[26] If indeed the Fe:C ratio in sinking particles varies with eolian Fe deposition, it will have important implications for deep water dissolved Fe concentration in the glacial ocean. It has been estimated that during glacial time, eolian Fe deposition to the ocean increases two- to five-fold in low latitudes and 10- to 50-fold in high latitudes such as in the Southern Ocean [Mahowald et al., 1999]. If the Fe:C ratio in sinking particles is linearly linked to eolian Fe input, and assuming everything else remaining the same, a five-fold increase in eolian Fe flux to the low-latitude Atlantic would increase the Fe export to the deep sea by five fold, which should increase the deep water dissolved Fe concentration. According to equation (2), a five-fold increase in Fe export to the deep waters of low-latitude Atlantic would increase the dissolved Fe concentration in deep waters of 70°S Atlantic by a factor of 5 if the scavenging residence

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**Figure 4.** Vertical profile of dissolved iron in the North Pacific near Hawaii in April 2001 (at 28.8°N, 158.8°W, crosses) as compared to that reported by Bruland et al. [1994] (circles) for a station at 200 km North of Hawaii, and that by Rue and Bruland [1995] (squares) for station Aloha near Hawaii.

1997], particle scavenging should cause deep water dissolved Fe concentrations to fall below total concentrations of Fe-binding organic ligands, if Fe scavenging rate exceeds the rate of Fe regeneration from sinking organic matter in the deep sea. Although excess concentrations of Fe-binding organic ligands (relative to total dissolved Fe) have been documented by field data from deep waters of the North Pacific [Rue and Bruland, 1995; Wu et al., 2001], previous ocean Fe modeling [Johnson et al., 1997; Archer and Johnson, 2000] have equated deep water dissolved Fe concentrations to organic ligand concentrations. Here we propose an alternative concept to describe dissolved Fe distribution in the deep Pacific which does not require a stoichiometric relationship between deep water concentrations of dissolved Fe and Fe-binding organic ligands.

[25] In this model, it is assumed that there is a competition between Fe input and Fe removal in deep sea and that Fe input from the microbial decomposition of sinking organic matter and Fe loss by particle scavenging are the primary driving forces. The imbalance of these two processes along deepwater flow paths would thus lead to a variation of dissolved Fe concentrations with distance along the deep-water flow path. When Fe input exceeds Fe loss, Fe concentrations would increase, whereas when the removal rate is larger than the input rate, the Fe concentrations would decrease. In the previous section, we have used this concept to explain the dissolved Fe distribution in the Sargasso Sea. Here we extend this conceptual model to the Pacific Ocean.
time does not change. This five-fold increase in deep water dissolved Fe concentration should result in an enhanced biological utilization of upwelled macronutrients in HNLC regions. However, complete utilization of nutrients would still require an additional supply of available Fe because even after the five-fold increases in dissolved Fe export, the resulting Fe:P concentration ratio in the deep water is about two-fold lower than that found in organic matter sinking from the euphotic zone. This additional Fe may be derived from eolian Fe deposition as a result of 10–50 fold higher eolian Fe deposition to the Southern Ocean during glacial time. If, in the present-day Southern Ocean, ~10% of available Fe input to the Southern Ocean surface water is via eolian Fe deposition [Watson et al., 2000], a 50-fold increase in eolian Fe input to this region during glacial time would increase the total Fe supply to the surface water by about a factor of 5, which, in combination with a factor of 5 increase in North Atlantic deep water dissolved Fe concentrations, would be adequate to meet the demand for a complete biological utilization of upwelled macronutrients. Because there are significant uncertainties associated with these calculations, we suggest that eolian Fe inputs in both the low-latitude Atlantic and the high-latitude Southern Ocean may be comparably important in controlling biological productivity in the high-latitude upwelling Southern Ocean on glacial/interglacial timescales. This hypothesis remains to be tested by future field measurements of Fe:C ratios in living organisms and sinking organic matter.

4. Conclusions

[27] The iron distribution in the Sargasso Sea can be reconciled with a view that dissolved (<0.4 μm) Fe concentrations in ocean surface waters are controlled not only by eolian Fe deposition, but also by convective mixing, phytoplankton uptake and particle scavenging. Our estimates imply that the dissolved Fe distribution in the deep waters of the world ocean can be described by an imbalance between Fe regeneration and particle scavenging. Maintenance of this imbalance requires a deep water Fe:P regeneration ratio much higher than the deep water Fe:P concentration ratio. This difference may be the root cause of Fe limitation of algal growth in regions of high upwelling and low eolian Fe deposition. Deep water dissolved Fe concentrations in the ocean appear to be regulated by regional changes of Fe:C ratio in the organic matter sinking from the euphotic zone.


References

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