GEOPHYSICAL RESEARCH LETTERS, VOL. 38, L18602, doi:10.1029/2011GL048972, 2011

Luxury uptake of aerosol iron by *Trichodesmium* in the western tropical North Atlantic

Ying Chen,[1] Antonio Tovar-Sanchez,[2] Ronald L. Siefert,[3] Sergio A. Sañudo-Wilhelmy,[4] and Guoshun Zhuang[1]

Received 20 July 2011; revised 25 August 2011; accepted 26 August 2011; published 17 September 2011.

[1] Dust transported from North Africa carries micronutrient iron (Fe) to the western tropical North Atlantic (WTNA) which may significantly influence the metabolism of the N_2-fixing cyanobacteria, *Trichodesmium*. For the first time, we conducted shipboard incubation experiments using freshly collected aerosol, seawater, and *Trichodesmium* colonies. *Trichodesmium* assimilated significant amount of aerosol Fe up to 14 times higher than the control. The uptake amount increased proportionally to the P: Fe ratio that *Trichodesmium* initially contained and to the aerosol Fe added and leached to the incubation solution. *Trichodesmium* assimilated more aerosol Fe than needed for its maximum growth (0.14 d^{-1}) demonstrating a high capacity of luxury uptake of Fe from the dust. **Citation:** Chen, Y., A. Tovar-Sanchez, R. L. Siefert, S. A. Sañudo-Wilhelmy, and G. Zhuang (2011), Luxury uptake of aerosol iron by *Trichodesmium* in the western tropical North Atlantic, *Geophys. Res. Lett.*, *38*, L18602, doi:10.1029/2011GL048972.

1. Introduction

[2] *Trichodesmium* is the most prominent nitrogen (N_2)-fixing cyanobacteira that occurs throughout the oligotrophic tropical and subtropical oceans [*Capone et al.*, 1997]. With its capacity to form extensive surface blooms, this diazotroph is likely a major contributor to "new" nitrogen of the marine ecosystem [*Capone et al.*, 1998; *Carpenter et al.*, 1999]. Evidence shows that N_2 fixation by *Trichodesmium* may contribute up to half of the nitrogen (N) required to sustain total annual production in the oligotrophic North Pacific, and therefore plays a critical role in the biogeochemical cycling of N and carbon [*Karl et al.*, 1997]. *Trichodesmium* has a high Fe requirement due to additional Fe demand of nitrogenase enzyme complex, and its growth under diazotrophy (N_2) requires 5 fold more Fe than needed for equivalent growth on ammonium [*Berman-Frank et al.*, 2001; *Kustka et al.*, 2003].

[3] Fe concentration is generally low in surface water of the open ocean but relatively high in the western tropical North Atlantic (WTNA) due to the impact of African dust. Atmospheric transport of continentally derived minerals, as a dominant source of Fe to the central ocean basins [*Duce and Tindale*, 1991], may ultimately control *Trichodemium* growth and oceanic N_2 fixation [*Falkowski*, 1997]. Enhanced *Trichodesmium* blooms have been observed in the tropical Atlantic, western Pacific and tropical Indian oceans that are downwind of the major dust production areas, the Saharan Desert/Sahel, the Gobi Desert and the deserts bounding the Arabian Sea, respectively [*Karl et al.*, 2002]. However, no correlation was found between the Fe concentration (total or dissolved) in seawater and *Trichodesmium* abundance in the Arabian Sea [*Capone et al.*, 1998], the central North Atlantic [*Sañudo-Wilhelmy et al.*, 2001], and along the Atlantic Meridional Transect (AMT) [*Tyrrell et al.*, 2003]. Accordingly, extremely short residence times (6-62 d) for total Fe were found in the upper water column of the equatorial Atlantic under the path of African dust plumes, suggesting that dust supply of Fe in surface water may be rapidly removed by aggregation and biological uptake [*Croot et al.*, 2004]. *Trichodesmium* uptake of aerosol Fe could be a rapid and dynamic process where the uptake amount may increase as the increasing input of atmospheric Fe to the ocean. *Kustka et al.* [2003] observed a "luxury" uptake of 13 fold greater Fe than needed for moderately Fe-limited growth (0.1 d^{-1}) in *Trichodesmium* cultures. Nonetheless, no *in situ* investigation has been conducted on *Trichodemium* uptake of aerosol Fe over oligotrophic oceans.

[4] For the first time we conducted shipboard incubation experiments using concurrent aerosol, seawater, and *Trichodesmium* samples collected over the WTNA between 18 April and 20 May 2003. The uptake of aerosol Fe by *Trichodesmium* was investigated in the area of heavy dust loading and *Trichodesmium* bloom.

2. Shipboard Incubation Experiments

[5] Aerosol, seawater and *Trichodesmium* samples were collected at three different locations where the incubation experiments (E1, E2 and E3) were performed. Latitudes and longitudes and associated seawater salinities are provided in Table 1. These sites are under the impact of Amazon River plume considering slightly low salinity. Nonetheless African dust is still important in terms of labile/bioavailable Fe supply [*Chen and Siefert*, 2004]. Aerosols were sampled on Teflon filters using a high-volume dichotomous virtual impactor (HVDVI) and sub-sampled (cut to 1/8) by a polycarbonate die and a ceramic knife. Surface seawater was pumped from a "fish" and filtered (<0.22 μm) through an acid-cleaned polypropylene cartridge filter. *Trichodesmium* colonies were collected using an acid-cleaned all-plastic 100 μm mesh plankton net. Each experiment had four treatments with duplicates including seawater blank (T1), seawater plus aerosol (T2), seawater plus *Trichodesmium* (T3), and sea-

[1]Department of Environmental Science and Engineering, Fudan University, Shanghai, China.
[2]IMEDEA, UIB, CSIC, Esporles, Spain.
[3]Chemistry Department, U.S. Naval Academy, Annapolis, Maryland, USA.
[4]Department of Biological Sciences, University of Southern California, Los Angeles, California, USA.

0094-8276/11/2011GL048972

Table 1. Latitude (LAT) and Longitude (LON), Salinity (SAL), Dissolved Fe (DFe) and P (DP) of Seawater, Fe Concentration and Labile Fe (LFe) to Phosphate (PO_4) Ratio in Aerosols Sampled at Three Experimental Sites (E1, E2, E3); Amount of Aerosol Fe Initially Added ($[Fe]_{ini}$), and the Fe Leached in Seawater ($[Fe]_{sw}$), Assimilated by *Trichodesmium* ($[Fe]_{intra}$) and Remained on the Filter Sub-sample ($[Fe]_r$) at the End of Incubation; Relative Percent Difference (RPD) Between the $[Fe]_{ini}$ and the Summation of $[Fe]_{sw}$ and $[Fe]_r$

	Location		Seawater			Aerosol							
	LAT	LON	SAL	DFe (nM)	DP (nM)	Fe (ng m^{-3})	LFe:PO_4 (mol:mol)	Treatment	$[Fe]_{ini}$ (nmol)	$[Fe]_{sw}$ (nmol)	$[Fe]_{intra}$ (nmol)	$[Fe]_r$ (nmol)	RPD
E1	10.8	52.3	32.2	0.6	27	50	0.14	T1	—	0.4	—	—	—
								T2	54	0.5	—	61	13%
								T3	—	0.1	0.9	—	—
								T4	102	1	1.1	122	19%
E2	8.3	51.7	34.3	4.8	27	490	0.67	T1	—	1.9	—	—	—
								T2	241	29	—	188	10%
								T3	—	1.5	1.1	—	—
								T4	241	20	2.4	231	4%
E3	11.6	55.7	34.6	2.4	—	446	0.68	T1	—	2.4	—	—	—
								T2	376	142	—	261	7%
								T3	—	0.1	0.6	—	—
								T4	376	53	8.5	297	7%

water plus aerosol and *Trichodesmium* (T4). Aerosol filter was added into each 300 mL polycarbonate (PC) bottle of T2 and T4. About 100 *Trichodesmium* colonies were transferred individually into each bottle of T3 and T4 resulting a ratio of 1 colony per 3 mL seawater comparable to *Trichodesmium* bloom situation in oligotrophic oceans [*Carpenter et al.*, 2004]. All bottles were placed in a flowing seawater incubator with a transparent PC cover for 24 hours, which blocks UV light and minimizes the photochemical reduction of aerosol Fe. *Trichodesmium* colonies were removed individually from each bottle of the E1 and E2 at the end, and placed on an acid-cleaned PC membrane soaked in 5 mL of ultra-clean oxalate reagent for 5 minutes and then passed by 10 mL of Nanopure water. For E3, the colonies were deposited in a 3 mL Teflon vial for total fraction analysis. The remaining seawater and aerosol filter sub-samples were also saved for analysis. Duplicates of each treatment were combined due to extremely low concentrations of Fe and P in seawater and *Trichodesmium*. All experimental operations were performed following trace-metal clean techniques.

3. Fe and P Analysis

[6] Labile aerosol Fe was measured onboard following a sequential aqueous extraction procedure (SAEP) with the detection limit of 1.0 nM; total aerosol Fe was determined by Inductively Coupled Plasma Mass Spectrometry (HP 4500) with the analytical uncertainty of 30 nmol; soluble phosphate was determined by Ion Chromatography (Dionex DX-600. Details are given by *Chen and Siefert* [2003, 2004]. Seawater samples were acidified by HCl to pH <1.5 and stored for over 1 month prior to analysis. Fe concentrations were determined by ICP-MS (ThermoFinigan, Element 2) after pre-concentration with APDC/DDDC organic extraction [*Bruland et al.*, 1985]. P concentration was determined by MAGIC method [*Karl and Tien*, 1992]. *Trichodesmium* collected on membrane or vial were acid digested and analyzed for Fe and P using ICP-MS [*Kustka et al.*, 2003] and spectrophotometry [*Gieskes et al.*, 1991] respectively.

4. Results and Discussion

[7] Aerosol Fe initially added to the T2 and T4 ($[Fe]_{ini}$) would be distributed to the seawater ($[Fe]_{sw}$), *Trichodesmium* colonies (if had), bottle wall, or remained on the filter sub-samples ($[Fe]_r$) at the end of incubation. Fe on the bottle wall was not measured but calculated using the maximum wall capacity (~30 nmol m^{-2}) and the surface area (~0.025 m^{-2}) of bottle [*Fischer et al.*, 2007]. $[Fe]_r$ and $[Fe]_{sw}$ are found to be major components accounting for over 95% of Fe distributed in the incubation bottle (Table 1). To check for Fe contamination, we compare the sum of $[Fe]_r$ and $[Fe]_{sw}$ with the $[Fe]_{ini}$ of each treatment and calculate relative percent difference (RPD) that varies between 4–10% for E2 and E3. Relatively high RPDs (13% and 19%) are found in E1 as a result of slightly high $[Fe]_r$, suggesting possible contamination to Fe residue on filter sub-sample. Overall, these RPDs are reasonable considering inhomogeneous distribution of aerosols on each filter sub-sampled suggesting a minimal Fe contamination in our experiment. T1 is also designed to check for Fe contamination during incubation process, and the values of 0.4–2.4 nmol per 0.6 L seawater are comparable to the dissolved Fe (0.6–4.8 nM) in the WTNA indicating a good application of trace-metal clean techniques (Table 1).

4.1. Fe Uptake by *Trichodesmium*

[8] At the end of incubation, *Trichodesmium* colonies in E1 and E2 were washed with oxalate reagent and analyzed for intracellular Fe ($[Fe]_{intra}$) following the same procedure used for the natural *Trichodesmium* sampled in the WTNA. In E3 total Fe associated with *Trichodesmium* was measured, and the $[Fe]_{intra}$ in T3 without aerosol addition can be calculated by multiplying the measured Fe by the ratio (0.65 ± 0.22) between intracellular and total Fe observed at 15 stations of the WTNA (A. Tover-Sanchez, unpublished data, 2004). The average $[Fe]_{intra}$ in T3 of the E3 are significantly lower than those of the E1 and E2 (Figure 1), which is consistent with the Fe contents in *Trichodesmium* sampled from the three experimental locations (0.8 nmol per 100 colonies for E1 and E2, and 0.6 nmol per 100 colonies for E3). *Trichodesmium* at the three locations also contain different levels of phosphorus with the minimum of 98 nmol per 100 colonies at E1 to the maximum of 234 nmol at E3, resulting in a sharply decrease of Fe: P ratio from 8.0, 5.2, to 2.5 mmol mol^{-1} that may influence the uptake capacity of *Trichodesmium* for aerosol Fe. Wash with oxalate reagent may not effectively remove the particulate aerosol Fe

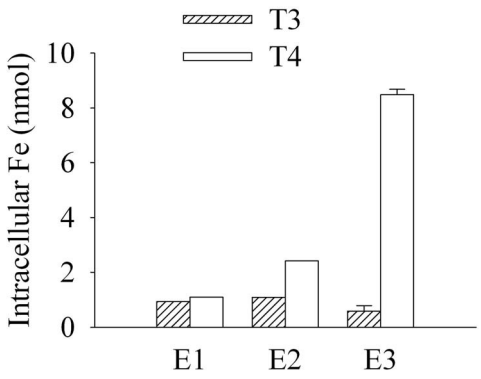

Figure 1. The intracellular Fe (nmol) in T3 (without aerosol addition) and T4 (with aerosol addition) of the incubation experiment E1, E2 and E3. The [Fe]intra in E3 was derived from the measured [Fe]total multiplying the ratio of [Fe]intra: [Fe]total in natural *Trichodesmium* sampled in the WTNA.

adsorbed on the *Trichodesmium* surface particularly in the incubation with high loadings of aerosol. The lithogenic Fe may be calculated according to the measured Al concentration that is a major and invariant component of crust [*Taylor*, 1964]. Taking into account an average Fe: Al crustal ratio of 0.04 [*Wedepohl*, 1995], it was demonstrated that lithogenic Fe was successfully distinguished from intracellular Fe for phytoplankton collected in the Southern Ocean after an oxalate wash [*Tovar-Sanchez et al.*, 2003; *Hassler and Schoemann*, 2009]. The $[Fe]_{intra}$ in T3 of the three experiments are then recalculated based on Al contents, and the correction of ~0.07 nmol of $[Fe]_{intra}$ is negligible.

[9] The measured Al concentrations on *Trichodesmium* are 34.7, 58.5 and 95.2 nmol for the T4 of E1, E2 and E3 respectively, which are used for the correction of lithogenic Fe. Al is non-bioavailable and its uptake would be very limited. Assuming the ratios of lithogenic Fe to Al were constant on *Trichodesmium* surface with and without oxalate wash, Al correction could be applied to total Fe to calculate the $[Fe]_{intra}$ for the T4 of E3. It is found that average $[Fe]_{intra}$ in T4 increased approximately a factor of 2 and 14 compared to those of T3 for the E2 and E3 respectively, suggesting significant uptakes of aerosol Fe by *Trichodesmium* in these two experiments (Figure 1). Correspondingly, $[Fe]_{sw}$ in T2 are significantly higher than those in T4 of the E2 and E3 implying that part of aerosol Fe leached in seawater had been assimilated by *Trichodesmium* (Table 1). It has been reported that *Trichodesmium* can use dust as an iron source [*Rueter*, 1988] and the uptake may happen quickly after the dissolution of particulate Fe. The increase of uptake amount of Fe is also proportional to the increase of the $[Fe]_{sw}$ in T2 for the three experiments, which further suggests that aerosol Fe freshly dissolved in seawater may be an Fe source preferred by *Trichodesmium*. The dissolution and followed uptake of aerosol Fe may happen simultaneously and therefore, previous studies couldn't find correlation between *Trichodesmium* abundance and dissolved Fe in the ocean [*Capone et al.*, 1998]. Soluble fraction of aerosol Fe freshly deposited to the ocean seems more important to the growth of *Trichodesmium*. Small change (18% increase) of $[Fe]_{intra}$ is found between T4 and T3 of the E1, which is probably due to the lower $[Fe]_{ini}$ used in E1 relative to E2 and E3. *Trichodesmium* sampled at E1 site contains the highest Fe: P ratio of 8.0 mmol mol^{-1}, indicating these colonies may have stored enough Fe for physiological metabolism. The $[Fe]_{intra}$ in T4 increases significantly as more $[Fe]_{ini}$ is added to the incubation solution from E1 to E3 (Figure 2). This is consistent with the laboratory observations where both cellular Fe content and Fe uptake rate of *Trichodesmium* increased proportionally (log/log) to the total Fe concentration in the culture media [*Kustka et al.*, 2003]. *Trichodesmium* sampled at three experimental sites contain different P: Fe ratios which increase proportionally with the uptake amounts of Fe (Figure 2), suggesting that P: Fe ratio may be one of critical factors controlling uptake capacity for aerosol Fe.

4.2. Luxury Uptake of Aerosol Fe

[10] *Sañudo-Wilhelmy et al.* [2001] suggested that the *Trichodesmium* in the central North Atlantic is not Fe-limited because the dissolved Fe: P ratios in seawater (11–15 mmol mol^{-1}) are 3–4 times higher than that measured in *Trichodesmium* (3.7 mmol mol^{-1}). Similarly, the dissolved Fe: P ratios of seawater at the experimental sites range between 22 and 400 mmol mol^{-1} (Table 1), about 3–167 times higher than the average ratio of 5.2 mmol mol^{-1} in *Trichodesmium* (Figure 2). The colonies used for experiments are not expected to be historically Fe-limited. Nonetheless they show a strong uptake of aerosol Fe during the incubation. Soluble phosphate in aerosols sampled over the WTNA range between 0.08 and 0.6 nmol m^{-3}, the lower end of which is comparable to the measurements from other Atlantic cruise [*Baker et al.*, 2006]. The molar ratio of Labile Fe to phosphate range between 0.14 and 0.68 in aerosols initially added (Table 1), which is about an order of magnitude higher than that of *Trichodesmium*. Accordingly, higher amount of aerosol Fe is assimilated compared to phosphate, causing the increases of intracellular Fe: P ratios by a factor of 1.2 and 4 at the end of incubation for E1 and E2 respectively. Therefore, *Trichodesmium* in the WTNA may be adapted to a luxury uptake of aerosol Fe as a consequence of episodic dust event transported from North Africa which contains a high ratio of labile Fe to phosphate.

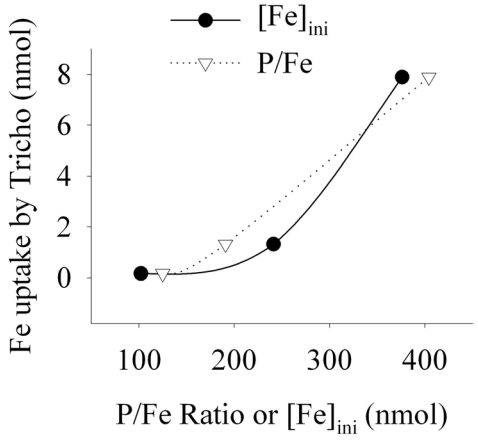

Figure 2. Amount of aerosol Fe assimilated by *Trichodesmium* (nmol) versus the P/Fe ratio (mol mol-1) of natural *Trichodesmium* sampled at experimental locations and the aerosol Fe (nmol) initially added to the incubation solutions.

[11] The C: N molar ratios of natural *Trichodesmium* range between 4.7 and 7.3 with a mean of 6.3 close to Redfield stoichiometry [*LaRoche and Breitbarth*, 2005]. The N: P ratio of 16 was reported for *Trichodesmium* grown under optimal laboratory conditions [*Berman-Frank et al.*, 2001]. Assuming that the C: P molar ratio of *Trichodesmium* was 106, the calculated Fe: C ratios would be 247 and 567 μmol: mol^{-1} in T4 of the E1 and E2 respectively. These ratios are higher compared to the Fe: C ratio of 230 μmol mol^{-1} required for the maximum growth of *Trichodesmium* cultures (IMS101) under diazotrophy (0.14 d^{-1} [*Kustka et al.*, 2003]). High Fe: C ratios (from 20 to >500 μmol: mol^{-1}) in natural *Trichodesmium* were also reported in several oceanic regions (e.g., Australia, Caribbean), which may be due to the low growth rate combined with Fe uptake in excess of growth requirements if limited by factors other than Fe [*Kustka et al.*, 2002]. *Trichodesmium* shows a rapid uptake of aerosol Fe in our experiments. The maximum uptake rate of 3.3×10^{-12} mol Fe colony^{-1} h^{-1} (based on E3) falls into the range of lab-based estimates between 2.2×10^{-12} and 14×10^{-12} mol Fe colony^{-1} h^{-1} reported by *Achilles et al.* [2003]. Our experiments with the addition of >100 nM aerosol Fe to the incubation solution for 1 day are much higher than the natural dust supply of Fe to the sea surface (10 nM d^{-1}) assuming the surface microlayer of 1 m and neglecting any losses of Fe (e.g., biological uptake). Nonetheless, the results still clearly show the large capacity of luxury uptake of aerosol Fe by the *Trichodesmium* in the WTNA.

[12] **Acknowledgments.** This work is sponsored by China Shanghai Pujiang Program (09PJ1401200), China National Natural Science Foundation (41005075), and Program for New Century Excellent Talents in University (NCET-09-0308).

[13] The Editor thanks an anonymous reviewer for their assistance in evaluating this paper.

References

Achilles, K. M., T. M. Church, S. W. Wilhelm, G. W. Luther III, and D. A. Hutchins (2003), Bioavailability of iron to *Trichodesmium* colonies in the western subtropical Atlantic Ocean, *Limnol. Oceanogr.*, *48*, 2250–2255, doi:10.4319/lo.2003.48.6.2250.

Baker, A. R., T. D. Jickells, M. Witt, and K. L. Linge (2006), Trends in the solubility of iron, aluminium, manganese and phosphorus in aerosol collected over the Atlantic Ocean, *Mar. Chem.*, *98*, 43–58, doi:10.1016/j.marchem.2005.06.004.

Berman-Frank, I., J. T. Cullen, Y. Shaked, R. M. Sherrell, and P. G. Falkowski (2001), Fe availability, cellular Fe quotas, and nitrogen fixation in *Trichodesmium*, *Limnol. Oceanogr.*, *46*, 1249–1260, doi:10.4319/lo.2001.46.6.1249.

Bruland, K. W., K. H. Coale, and L. Mart (1985), Analysis of seawater for dissolved cadmium, copper, and lead: An intercomparison of voltametric and atomic adsorption methods, *Mar. Chem.*, *17*, 285–300, doi:10.1016/0304-4203(85)90002-7.

Capone, D. G., J. P. Zehr, H. W. Paerl, B. Bergman, and E. J. Carpenter (1997), Trichodesmium a globally significant marine cyanobacterium, *Science*, *276*, 1221–1229, doi:10.1126/science.276.5316.1221.

Capone, D. G., A. Subramaniam, J. Montoya, M. Voss, C. Humborg, A. Johansen, R. Siefert, and E. J. Carpenter (1998), An extensive bloom of the N_2-fixing cyanobacterium, *Trichodesmium erythraeum*, in the central Arabian Sea, *Mar. Ecol. Prog. Ser.*, *172*, 281–292, doi:10.3354/meps172281.

Carpenter, E. J., J. P. Montoya, J. Burns, M. Mulholland, A. Subramaniam, and D. G. Capone (1999), Extensive bloom of a N_2 fixing symbiotic association in the tropical Atlantic Ocean, *Mar. Ecol. Prog. Ser.*, *185*, 273–283, doi:10.3354/meps185273.

Carpenter, E. J., A. Subramaniam, and D. G. Capone (2004), Biomass and primary productivity of the cyanobacterium *Trichodesmium* spp. in the tropical N Atlantic Ocean, *Deep Sea Res., Part I*, *51*, 173–203, doi:10.1016/j.dsr.2003.10.006.

Chen, Y., and R. L. Siefert (2003), Determination of various types of labile atmospheric Fe over remote oceans, *J. Geophys. Res.*, *108*, 4774, doi:10.1029/2003JD003515.

Chen, Y., and R. L. Siefert (2004), Seasonal and spatial distributions and dry deposition fluxes of atmospheric total and labile Fe over the tropical and sub-tropical North Atlantic Ocean, *J. Geophys. Res.*, *109*, D09305, doi:10.1029/2003JD003958.

Croot, P. L., P. Streu, and A. R. Baker (2004), Short residence time for iron in surface seawater impacted by atmospheric dry deposition from Saharan dust events, *Geophys. Res. Lett.*, *31*, L23S08, doi:10.1029/2004GL020153.

Duce, R. A., and N. W. Tindale (1991), Atmospheric transport of Fe and its deposition in the ocean, *Limnol. Oceanogr.*, *36*, 1715–1726, doi:10.4319/lo.1991.36.8.1715.

Falkowski, P. G. (1997), Evolution of the nitrogen cycle and its influence on the biological sequestration of CO_2 in the ocean, *Nature*, *387*, 272–275, doi:10.1038/387272a0.

Fischer, A. C., J. J. Kroon, T. G. Verburg, T. Teunissen, and H. T. Wolterbeek (2007), On the relevance of iron adsorption to container materials in small-volume experiments on iron marine chemistry ^{55}Fe-aided, *Mar. Chem.*, *107*, 533–546, doi:10.1016/j.marchem.2007.08.004.

Gieskes, J. M., T. Gamo, and H. Brumsack (1991), Chemical Methods for Interstitial Water Analysis aboard Joides Resolution, in *Ocean Drilling Program*, *Tech. Rep. 15.*, pp. 46–47, Tex. A&M Univ., College Station.

Hassler, C. S., and V. Schoemann (2009), Discriminating between intra- and extracellular metals using chemical extractions: An update on the case of iron, *Limnol. Oceanogr. Methods*, *7*, 479–489, doi:10.4319/lom.2009.7.479.

Karl, D. M., and G. Tien (1992), MAGIC: A sensitive and precise method for measuring dissolved phosphorus in aquatic environments, *Limnol. Oceanogr.*, *37*, 105–116, doi:10.4319/lo.1992.37.1.0105.

Karl, D., R. Letelier, L. Tupas, J. Dore, J. Christian, and D. Hebel (1997), The role of nitrogen fixation in biogeochemical cycling in the subtropical North Pacific Ocean, *Nature*, *388*, 533–538, doi:10.1038/41474.

Karl, D., A. Michaels, B. Bergman, D. Capone, E. Carpenter, R. Letelier, F. Lipachultz, H. Paerl, D. Sigman, and L. Stal (2002), Dinitrogen fixation in the world's oceans, *Biogeochemistry*, *57–58*, 47–98, doi:10.1023/A:1015798105851.

Kustka, A., E. J. Carpenter, and S. A. Sañudo-Wilhelmy (2002), Iron and marine nitrogen fixation: Progress and future directions, *Res. Microbiol.*, *153*, 255–262, doi:10.1016/S0923-2508(02)01325-6.

Kustka, A. B., S. A. Sañudo-Wilhelmy, E. J. Carpenter, D. Capone, J. Burns, and W. G. Sunda (2003), Fe requirements for dinitrogen- and ammonium-supported growth in cultures of *Trichodesmium* (IMS 101): Comparison with nitrogen fixation rates and Fe: Carbon ratios of field populations, *Limnol. Oceanogr.*, *48*, 1869–1884, doi:10.4319/lo.2003.48.5.1869.

LaRoche, J., and E. Breitbarth (2005), Importance of the diazotrophs as a source of new nitrogen in the ocean, *J. Sea Res.*, *53*, 67–91, doi:10.1016/j.seares.2004.05.005.

Rueter, J. G. (1988), Iron stimulation of photosynthesis and nitrogen fixation in Anabaena 7120 and *Trichodesmium* (Cyanophyceae), *J. Phycol.*, *24*, 249–254.

Sañudo-Wilhelmy, S. A., A. B. Kustka, C. J. Gobler, D. A. Hutchins, M. Yang, K. Lwiza, J. Burns, D. G. Capone, J. A. Raven, and E. J. Carpenter (2001), Phosphorus limitation of nitrogen fixation by *Trichodesmium* in the central Atlantic Ocean, *Nature*, *411*, 66–69, doi:10.1038/35075041.

Taylor, S. R. (1964), Abundance of chemical elements in the continental crust: A new table, *Geochim. Cosmochim. Acta*, *28*, 1273–1285, doi:10.1016/0016-7037(64)90129-2.

Tovar-Sanchez, A., S. A. Sañudo-Wilhelmy, M. Garcia-Vargas, R. S. Weaver, L. C. Popels, and D. A. Hutchins (2003), A trace metal clean reagent to remove surface-bound Fe from marine phytoplankton, *Mar. Chem.*, *82*, 91–99, doi:10.1016/S0304-4203(03)00054-9.

Tyrrell, T., E. Marañon, A. J. Poulton, A. R. Bowie, D. S. Harbour, and E. M. S. Woodward (2003), Large-scale latitudinal distribution of *Trichodesmium spp.* in the Atlantic Ocean, *J. Plankton Res.*, *25*, 405–416.

Wedepohl, H. K. (1995), The composition of the continental crust, *Geochim. Cosmochim. Acta*, *59*, 1217–1232, doi:10.1016/0016-7037(95)00038-2.

Y. Chen and G. Zhuang, Department of Environmental Science and Engineering, Fudan University, 220 Handan Rd., Shanghai 200433, China. (yingchen@fudan.edu.cn)

A. Sañudo-Wilhelmy, Department of Biological Sciences, University of Southern California, 3616 Trousdale Pkwy., Los Angeles, CA 90089-0371, USA.

R. L. Siefert, Chemistry Department, U.S. Naval Academy, 572M Holloway Rd., Annapolis, MD 21402-5026, USA.

A. Tovar-Sanchez, IMEDEA, UIB, CSIC, Miquel Marques 21, Esporles, E-07190 Mallorca, Spain.