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Efficiency of carbon removal per added iron in ocean iron fertilization

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ABSTRACT: The major response to ocean iron fertilization is by large diatoms, which at Fe-replete ambient seawater show an optimum C:Fe elemental ratio of ~23 000 and a higher ratio of ~160 000 or more under Fe-limited conditions. The efficiency of CO_2 drawdown during the several weeks of artificial fertilization experiments with concomitant observations is in the range of $100 < (CO_2{:}Fe) < 1000$ and is unknown in direction (positive or negative) and magnitude in the period after observations. The efficiency of biogenic carbon export into deeper water layers ranges from $\sim 650 < (C{:}Fe)_{export} < \sim 25\,000$ for reported export depths in the 100 to 250 m range. Variations in ocean initial conditions and variable weather during an experiment cause this range of ~2 orders of magnitude. Approximately 75% of Fe added in fertilization experiments is lost very rapidly. Hence the above efficiencies can be multiplied 4-fold, to $\sim 2600 < (C{:}Fe)_{export} < \sim 100\,000$, for the sake of comparison with natural fertilization with Fe-organic complexes, which stabilize Fe in solution. Quantification of the Fe source of natural fertilization is difficult, leading to an export efficiency in the $\sim 2400 < (C{:}Fe)_{export} < \sim 800\,000$ range. Due to severe under-sampling, the existing datasets of artificial experiments and natural fertilizations may allow a wider range of alternative assessments than reported here.

KEY WORDS: Iron · Fertilization · Ocean · Efficiency · Carbon · Export · Diatoms

INTRODUCTION

This study assessed the efficiency (or efficacy) of carbon removal from the upper ocean and atmosphere by natural or artificial Fe fertilization of ocean surface waters. The high-nutrient, low-chlorophyl regions (HNLC) of the oceans have been shown to suffer from the lack of essential trace nutrient iron, but other factors, notably light limitation (Smith & Nelson 1985, Mitchell et al. 1991, de Baar et al. 2005) also play a key role. Among the 3 major HNLC regions of the World ocean (the Antarctic Ocean, the equatorial Pacific upwelling region and the subarctic North Pacific Ocean), it is only in the Antarctic Ocean that Fe stimulation of CO_2 fixation would, in principle, yield long-term C storage in deeper waters (Sarmiento & Orr 1991). Fertilization of the Antarctic Ocean with extra Fe has been suggested both as an explanation for the past glacial periods (Martin 1990) and as an option for solving the present day fossil-fuel CO_2 problem. The latter intentional, large-scale, ocean iron fertilization (OIF) concept is the subject of this article, which focuses on the efficiency (effectiveness, efficacy) of carbon removal, with emphasis on the most relevant lines of evidence, i.e. relevance for the Antarctic Ocean.

PARAMETERIZATION

Universal response of large diatoms. Briefly, from all Fe-limitation and Fe-fertilization observations and experiments in bottles (reviewed by de Baar & Boyd

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2000), in natural fertilization systems (de Baar et al. 1995, Blain et al. 2007, Pollard et al. 2007), and in artificial *in situ* fertilizations (de Baar et al. 2005, Hoffmann et al. 2006, Tsuda et al. 2007), it is known that the supply of Fe strongly favors and stimulates the larger size-class (typically >10 µm) diatoms (de Baar & Boyd 2000). One key parameter is the cellular requirement $(C:Fe)_{large\text{-}diatoms\text{-}optimal}$ of C versus Fe for healthy well-growing large diatoms in optimal Fe-replete conditions (Table 1).

Efficiency of net dissolved inorganic carbon (DIC) removal. Overall, the DIC decrease presently observed from *in situ* Fe fertilization experiments thus far tends to be ~51% (±26%, SD) of total primary production, ranging between 16 and 87% across 7 *in situ* fertilizations (de Baar et al. 2005). For these experiments, the ratio

$$(\Delta DIC:Fe)_{NCP} = [(\Delta DIC_{in\text{-}patch} - \Delta DIC_{control\text{-}station}):Fe] \quad (1)$$

of observed DIC decrease versus added Fe serves as one of the efficiency parameters. The DIC loss is taken over the complete patch (i.e. lateral patch dilution has been taken into account). The DIC loss represents net community production (NCP) and is useful well beyond the plankton ecosystem, i.e. in larger ocean or global biosphere contexts.

Efficiency of atmosphere to ocean CO_2 flux. Net uptake of DIC leads to a decrease in the equilibrium value of pCO_2 in surface waters, and as result an influx of CO_2 into the ocean can be compared with the gas flux at a control site, leading to an efficiency for CO_2 drawdown:

$$(C:Fe)_{gas\text{-}flux\ efficiency} = CO_2:Fe =$$
$$(Flux_{fertilized\text{-}patch} - Flux_{control\text{-}station}) \,/\, Fe_{added} \quad (2)$$

as another efficiency parameter to be compared between experiments (Table 1). This gas flux tends to be slow, partly due to slow chemical equilibration of the CO_2 system in seawater, such that after the period of observation there may or may not be considerably more gas exchange taking place in subsequent weeks or even months. This CO_2 gas exchange may continue to be from the atmosphere into the ocean, but may also become reversed due to the common sequence of net community production being succeeded by collapse of the phytoplankton bloom which is followed in turn by a stage of net community respiration which tends to drive excess CO_2 back into the atmosphere. However, we only assess the observed gas exchange flux during the actual period of shipboard observation of an experiment.

Efficiency of particle export into deeper waters. The export of settling organic matter (i.e. organic carbon) into deeper water layers has been assessed using the ^{234}Th deficiency method (Buesseler et al. 2004, 2005, Blain et al. 2007, Morris et al. 2007) and sediment traps (Boyd et al. 2004, Salter et al. 2007). This export is often defined just below the euphotic zone at a depth of 100 m, leading to an export efficiency

$$(C:Fe)_{export\text{-}efficiency\text{-}100m} =$$
$$[(C_{export\text{-}in\text{-}patch} - C_{export\text{-}control\text{-}site})_{100m}:Fe] \quad (3)$$

However, Buesseler et al. (2004) reckon that on average >50% of the particles sinking at 100 m depth are remineralized before they reached 250 m depth and thus an extrapolated efficiency is defined at 250 m (Table 1). For the Kerguelen Ocean and Plateau Compared Study (KEOPS), an observed export is reported for 200 m depth (Blain et al. 2007).

Fate in deep ocean waters. Once exported below the 100 to 250 m depth horizon, the fate of this carbon is not further quantified in this paper, but a brief qualitative prediction of its fate is as follows. From settling fluxes into sediment traps, it is known that the downward flux of particulate matter (i.e. biogenic C, N and P; often accompanied by Si opal and $CaCO_3$ skeletons) decreases exponentially with increasing depth due to its utilization as substrate by bacteria where most is respired into CO_2, nitrate and phosphate. For the soft tissue components (C, N, P) some differentiation exists of the apparent first-order rate of remineralization (i.e. dissolution) as function of depth (Wakeham et al. 1980, de Baar et al. 1983), but we take the simplified view that decomposition delivers DIC, nitrate and phosphate in standard Redfield proportions (C:N:P = ~106:~16:1) into the intermediate and deep waters. Exponential function fits

Table 1. Parameters for comparison between fertilization experiments. Export at 250 m is by extrapolation from export at 100 m depth. Not listed are the actual observed export at 200 m in the Kerguelen Ocean and Plateau Compared Study (KEOPS) and 150 m in the European Iron Fertilization Experiment (EIFEX)

True element ratio values	Complete equation	Eq. no.
$(C:Fe)_{large\text{-}diatoms\text{-}optimal}$		
Efficiency ratio values		
$(\Delta DIC:Fe)_{NCP}$	$(\Delta DIC_{in\text{-}patch} - \Delta DIC_{control\text{-}station}):Fe$	(1)
$(C:Fe)_{gas\text{-}flux\ efficiency}$	$(Flux_{fertilized\text{-}patch} - Flux_{control\text{-}station}) \,/\, Fe_{added}$	(2)
$(C:Fe)_{export\text{-}efficiency\text{-}100m}$	$(C_{export\text{-}in\text{-}patch} - C_{export\text{-}control\text{-}site})_{100m}:Fe$	(3)
$(C:Fe)_{export\text{-}efficiency\text{-}250m}$	$(C_{export\text{-}in\text{-}patch} - C_{export\text{-}control\text{-}site})_{250m}:Fe$	

to sediment trap fluxes versus depth (Knauer & Martin 1981, de Baar et al. 1983, Martin et al. 1987) typically show that of the export across a ~100 m depth horizon, some 90% or more is remineralized in the 100 to 1000 m depth range, and that between 1000 m and ~4000 m depth, half or more of the C particle flux is lost once again. Combining such exponential loss function with ocean circulation modeling provides a prediction of how long the DIC, nitrate and phosphate will remain in the ocean interior before eventually (e.g. due to upwelling) returning to the surface ocean. Excess CO_2 then will be outgassed into the atmosphere, while the parallel excess nitrate and phosphate create high nutrient regions, where the recycled nutrients are suitable for once again supporting phytoplankton growth, provided there is adequate Fe and favorable light conditions. The problem is that trace-nutrient Fe does not follow Redfield stoichiometry during re-mineralization; Fe is very particle-reactive and due to adsorptive scavenging is lost from deep ocean waters (de Baar & de Jong 2001). This is shown by modeling of the ocean Fe cycle versus the regular nutrient phosphate cycle (Parekh et al. 2005, Bergquist & Boyle 2006). Thus, each time new upwelled water rich in DIC and major nutrients reaches the surface, it has lost most or virtually all of its remineralized Fe content and only by new Fe addition (e.g. dust, anoxic margin sediments, artificial OIF) can the plankton bloom and subsequently start the biological cycle all over again.

Combining these concepts of particle flux and remineralization with ocean circulation modeling provides an assessment of the fate of carbon sequestered below 100 m depth (chosen as the standard depth for export in this paper). From these modeling approaches we know that among the major HNLC regions, only the Antarctic Ocean would yield significant long term (>100 yr) carbon storage (Sarmiento & Orr 1991). Moreover, the deeper the defined horizon of particle flux, the lower the $(C:Fe)_{export\text{-}depth(m)\text{-}efficiency}$. At ~1000 m the efficiency will be an order of magnitude lower than at 100 m depth, at ~4000 m the efficiency may well be ~40-fold lower than at 100 m. In general, deep ocean waters tend to have a longer deep residence time than intermediate depth waters, hence a longer storage time of sequestered carbon before the water mass reaches the surface again and exchanges its excess CO_2 with the atmosphere. However, with increasing depth the C:Fe efficiency is less. Thus, with increasing depth the expected longer storage time is countered by the expected lower C:Fe efficiency. Quantification by deep ocean simulation modeling of this fate and storage time in the deep oceans of the here quantified OIF-driven export at ~100 m depth is recommended.

RESULTS

When aiming to resolve an issue in biological oceanography, Lalli (1991) and Lalli & Parsons (1993) have suggested a combination of 3 lines of approach: (1) observations in the field; (2) mesoscale size experiments (e.g. mesocosms); and (3) controlled experiments in the laboratory; and these 3 approaches interlinked by modeling and interpretation. No single approach in itself can provide the final answer, but the combination may. For the role of Fe in ocean ecosystems, Fe fertilization experiments and CO_2 transfer in the global oceans, this has been further developed by de Baar & Boyd (2000). Accordingly, to assess the effectiveness of OIF, the several lines of evidence (Approaches 1 to 3 above) as well as modeling (e.g. Gnanadesikan & Marinov 2008) need consideration, as does the spectrum of response to added Fe from a single species to a complete plankton community.

Iron requirement of large diatoms

Determining the Fe content of plankton cells is complicated by the fact that Fe is extremely reactive towards particles and tends to adsorb on the outside of the cell. Analyses of whole plankton would yield a severe overestimation. Attempts have been made to avoid this by gentle washing of the external Fe fraction with, for example, Ti-citrate solution (Hudson & Morel 1989, Tovar-Sanchez et al. 2003, Tang & Morel 2006). Unfortunately, it is not certain that washing removes all adsorbed Fe; and the process can rupture the cells, leading to an overall underestimation of cellular Fe content. Recently, these problems have been overcome by the application of synchrotron X-ray radiation and detection, which allows quantification of the truly intracellular Fe, distinct from the surface-bound Fe (Twining et al. 2004). Moreover, the method examines individual cells, thus permitting taxonomic and size classification. For large diatoms in the Fe-enriched patch of the Southern Ocean Iron Experiment (SOFeX)-South experiment, an intracellular C:Fe ratio of ~23 000 was reported and, thus, we consider ~23 000 representative of $(C:Fe)_{large\text{-}diatoms\text{-}optimal}$ as well (Table 2). The optimal Fe conditions for optimal growth rate were confirmed independently by the very high (virtually theoretical maximum) value F_v/F_m = 0.65, confirming very healthy phytoplankton cells (Coale et al. 2004). In the natural Fe-depleted region outside the fertilized SOFeX patch, i.e. at the control station, the cellular C:Fe ratio of ~160 000 for diatoms is considered sub-optimal, due to ambient Fe depletion, also confirmed by the low F_v/F_m = 0.25 before the Fe addition.

Table 2. The thus-far best estimate of $(C:Fe)_{large\text{-}diatoms\text{-}optimal}$, ~23 000, as well as various other estimates from the literature for C:Fe of diatoms and whole plankton assemblages (Bruland et al. 1991, based on Martin & Knauer 1973, Martin et al. 1976, Collier & Edmond 1983) and their deep ocean debris and mineralization ratios (Martin et al. 1989). Literature values of diatoms for the 84% (n = 21) lower percentile of all published values, after Sarthou et al. (2005). Using all sources (n = 25), including 4 more outlier ratio values, the average increases to C:Fe = 68 863 ± 112 167 (mean ± SD). GFAAS: graphite furnace atomic absorption spectrometry. See 'Results' for definitions of study area acronyms

Description	C:Fe ratio value	Method	Source
$(C:Fe)_{large\text{-}diatoms\text{-}optimal}$	~23 000	SOFeX IN patch; synchrotron X-ray	Twining et al. (2004)
$(C:Fe)_{large\text{-}diatoms\text{-}Fe\text{-}limited}$	~100 000–160 000	SOFeX OUT patch	Twining et al. (2004)
$(C:Fe)_{diatoms\text{-}literature}$	28 571 ± 24 440	84% percentile (n = 21)	Sarthou et al. (2005)
$(C:Fe)_{diatoms\text{-}literature}$	68 863 ± 112 167	100% percentile (n = 25)	Sarthou et al. (2005)
$(C:Fe)_{whole\text{-}plankton\text{-}tows}$	~21 000	GFAAS	Bruland et al. (1991)
$(C:Fe)_{excess\text{-}deep\text{-}particles}$	~33 000	Debris by GFAAS	Martin et al. (1989)
$(C:Fe)_{\textit{Thalassiosira oceanica}}$	~30 000–100 000	Optimal growth rate	Sunda et al. (1991)
$(C:Fe)_{\textit{Thalassiosira oceanica}}$	~500 000	Suboptimal 90% growth	Sunda et al. (1991)
$(C:Fe)_{nitricline}$	~483 000	Linear regression	Martin et al. (1989)
$(C:Fe)_{oxycline}$	~384 000	Linear regression	Martin et al. (1989)
$(C:Fe)_{plankton\text{-}content}$	227000 ± 5000 (n = 10) 175 000–333 000 range	KEOPS shipboard; ^{55}Fe radiotracer, with oxalate washing off the extracellular Fe	Sarthou et al. (2008)

Another approach to assess iron requirement is by assessing growth rate as a function of the ambient concentration of dissolved trace-nutrient Fe. As there is no good understanding yet on the effect of Fe chemical speciation (notably Fe-organic complexes) on the growth rate of a given phytoplankton species, it is best to simply take the dissolved Fe concentration as the growth-rate-controlling parameter in otherwise natural, unperturbed, Antarctic seawater (Timmermans et al. 2004). Timmermans et al. (2004) demonstrated that the K_m values for growth of large Antarctic oceanic diatoms *Fragilariopsis kerguelensis*, *Chaetoceros dichaeta*, *Thalassiosira sp.*, *Corethron pennatum* and *Actinocyclus* sp. range from 0.19 nM to 1.14 nM, i.e. generally well above the average concentration of dissolved Fe in Antarctic surface waters. Moreover, these K_m values have been shown to be inversely related to the surface:volume ratio of these diatoms (de Baar et al. 2005). These results are consistent with the observation that large diatoms bloom only occasionally in the open Antarctic Ocean, presumably upon an event of Fe supply either from below (de Baar et al. 1995) or from above by wet deposition of Fe-rich dust. Notably in the Southern Ocean Iron Enrichment Experiment (SOIREE) fertilization, the (inside-patch) dissolved Fe was always above ~0.2 nM, allowing *Fragilariopsis kerguelensis* to maintain at least its half maximum growth rate (de Baar et al. 2005) and eventually become the dominant species in response to the Fe fertilization.

The above independent estimates of intracellular Fe concentration (Twining et al. 2004) and large diatoms growth rates (Timmermans et al. 2004) were shown to be in perfect agreement with the diffusion limitation of growth rate of these diatoms (de Baar et al. unpubl. data). At increasing size the less favorable surface: volume ratio prescribes a necessary higher ambient Fe concentration for growth continuation. Conversely, small cells with the same intracellular Fe concentration, i.e. spheres with a typical diameter less than ~8 µm, are never Fe-limited in an ocean where dissolved Fe always exceeds an 0.01 nM concentration (de Baar et al. unpubl. data). This is consistent with the observation that small *Chaetoceros brevis* is never Fe-limited (Timmermans et al. 2001).

While Fe enrichments thus far have shown a response from the larger size class of diatoms, from decades of independent field observations we know that major blooms in the Antarctic Ocean are due to either such large diatoms, or the colony-forming *Phaeocystis antarctica*. For example, in the Ross Sea there are blooms of either diatoms or *P. antarctica* in distinct regions and seasons, and the cause for this distinction is of great interest (Arrigo et al. 2003, Tagliabue & Arrigo 2005). Recently, Sedwick et al. (2007) have shown from *P. antarctica* cultures in natural Antarctic seawater the K_m value for growth of ~0.45 nM dissolved Fe. This is in the same range as for the above large diatoms. Previous estimates for incubations of Ross Sea Prymnesiophytes were about one order of magnitude lower (Coale et al. 2003), the latter presumably solitary *P. antarctica* cells (diameter ~4 to 6 µm; Rousseau et al. 2007) with a more favorable surface:volume ratio for diffusive Fe assimilation.

Other literature estimates of iron requirement of diatoms

In context of the Iron Resources and Oceanic Nutrients: Advancement of Global Ecosystem Simulations (IRONAGES) project, reviews based on previous studies have focused on the limitations by light, major nutrients and iron, of major bloom-forming groups, the diatoms (Sarthou et al. 2005), *Phaeocystis* spp. (Schoemann et al. 2005), diazotrophs (La Roche & Breitbarth 2005), and picoplankton (Timmermans et al. 2005), while fine reviews already existed for the coccolithophorids (Iglesias-Rodríguez et al. 2002, Paasche 2002). Briefly, the cell quota for diatoms have an average (mean ± SD) C:Fe = 28 571 ± 24 440 based on 21 publications of laboratory incubation studies (Sarthou et al. 2005).

Several of the previous studies, while novel at the time, may in retrospect require caveats for the following reasons. Firstly, the problem of measuring true intracellular Fe content as mentioned above renders many old C:Fe values questionable. Secondly, in the pioneering days of metal–phytoplankton interaction experiments, it was not yet possible to achieve the ultraclean dissolved metal concentrations we know to exist in the real ocean. In order to overcome this, the culture media were overdosed with 10^{-4}M EDTA, which binds all dissolved metal ions; by then overdosing with the metal of interest, a 'free metal' concentration similar to the total metal in the real ocean was achieved (Provasoli et al. 1957). This method has led to many breakthroughs (e.g. Sunda & Guillard 1976). For Fe limitation, the inherent paradigm has become (de Baar 1994) that the calculated free Fe′ is the controlling agent for phytoplankton growth. However, these experiments were conducted in EDTA media. Since the 1994 discovery of >99% organic complexation of dissolved Fe in the real ocean (Gledhill & van den Berg 1994), EDTA media now are known to be not representative of the real ocean (Gerringa et al. 2000). Moreover, with the ultraclean methods of today, it is not necessary anymore to add EDTA to circumvent inadvertent contamination.

From a comparison of the cellular Fe requirements of the coastal diatom *Thalassiosira pseudonana* and the oceanic diatom *T. oceanica* (Sunda et al. 1991), one can determine from the reported growth curve that at the optimum growth rate of ~1.2 d^{-1} of *T. oceanica*, the corresponding suggested cell ratio is in the range of C:Fe = ~30 000 to 100 000. At suboptimal (~90% of maximum) rate of growth the corresponding cell ratio presumably is in the order of C:Fe = ~500 000. However, these experiments were in EDTA media and not relevant for the real ocean. Otherwise, (Sunda et al. 1991) reported that it was not possible to drive *T. oceanica* into Fe limitation, at most a suppression to 90% of maximum growth was realized. This is consistent with the fact that *T. oceanica* is a very small diatom and therefore will never be Fe diffusion limited in real ocean waters (de Baar et al. 2008), akin to the similarly small oceanic diatom *Chaetoceros brevis* which could not be driven into Fe limitation in natural Antarctic seawater either (Timmermans et al. 2001), except after the addition of very strong siderophore DFOB (desferrioxamine-B) making Fe unavailable for uptake. Quite remarkably, addition of DFOB to natural Antarctic seawater cannot even drive *T. oceanica* into Fe limitation (K. R. Timmermans et al. unpubl. data). Somehow, DFOB-bound Fe appears to be available to *T. oceanica*, but not at all available to *C. brevis* (Timmermans et al. 2001). Similarly, Hutchins et al. (1999) reported that different phytoplankton species can access different Fe-organic species, i.e. something more than just inorganic Fe′(III) is affecting growth. Sedwick et al. (2007) also report that DFOB bound Fe somehow is partly available for *Phaeocystis antarctica*. Researchers are only beginning to unravel the intriguing interactions between several organic ligands and several different phytoplankton species (Rijkenberg et al. 2006, 2008), as natural iron–plankton interactions can only be assessed in natural seawater.

Efforts to derive C:Fe ratio values from the deep ocean

One of the first reliable vertical profiles of dissolved Fe in the deep ocean was in the north Pacific Ocean at 50° N, 145° W (Ocean Station Papa) which appeared to exhibit similarity to the vertical distribution of nitrate (Martin et al. 1989) but, linear correlation, akin to e.g. phosphate versus nitrate (Redfield et al. 1963), between Fe and nitrate was not successful. However, by combining data only in the nitricline of 50° N, 145° W (Ocean Station Papa) and adjacent stations at 40° N, 140° W (Vertex T-5) and 45° N, 143° W (Vertex T-6) a linear relation was achieved (Martin et al. 1989). Conversion of N to C by classical Redfield (C:N = 106:16 = ~6.6) yielded C:Fe = ~483 000 (de Baar & de Jong 2001, p. 164). Similarly, versus the oxycline, C:Fe = ~384 000 was obtained. On the other hand, excess Fe on deep particles (excess over lithogenic Fe) presumably represents biogenic Fe and led to a C:Fe = ~33 000 (Martin et al. 1989). This is similar to the C:Fe = ~21 000 of whole plankton tows (Bruland et al. 1991).

With some more recent vertical profiles of dissolved Fe in other ocean basins (de Baar & de Jong 2001, Parekh et al. 2005, Bergquist & Boyle 2006), it now appears that dissolved Fe in the deep oceans is within a narrow band of concentrations and does not show the general in-

crease from young deep Atlantic to old deep Pacific waters as do the micronutrients N, P and Si. This is likely due to net removal of dissolved Fe from deep waters by adsorptive scavenging, while there is no net removal of micronutrients (Parekh et al. 2005). Hence, it appears that each ocean basin has its own Fe cycle and that there is little exchange of Fe between basins (de Baar & de Jong 2001). It seems there is no straightforward relationship between deep Fe and deep micronutrients. Most likely the deep ocean ratio of dissolved Fe versus major nutrient (e.g. nitrate or phosphate) tends to decrease with the age of deep water; hopefully, the ongoing Geotraces global mapping project will provide new insight into this area in the near future.

Values of C:Fe ratio derived from the 1993–2002 *in situ* fertilization experiments

By comparison of the *in situ* fertilization experiments, a compilation of efficiency values $(\Delta DIC:Fe)_{NCP}$ has been reported before (de Baar et al. 2005, their Table 3) and is given in Table 3. Moreover, we have calculated the other efficiency ratio values for gas influx during the observation period of each experiment, as well as the export efficiency at the 100 m depth horizon. Note that in most ocean regions, the export efficiency at the 250 m depth horizon would be about half that at 100 m, as observed and applied by Buesseler et al. (2004) in the context of the SOFeX-South experiment in the Antarctic Ocean.

The $(C:Fe)_{gas\text{-}flux\text{-}efficiency}$ varies between 100 and 1000 mol CO_2 gas influx into the ocean mol^{-1} Fe added. This estimate may, on the one hand, be considered a lower limit, as gas influx would presumably continue when the bloom continued after the period of observation, i.e. after the ships had left. On the other hand, it may just as well be considered an upper limit. For example, upon the collapse of a bloom (e.g. at end of SERIES), the ecosystem would most likely tend to shift to net community respiration, eventually accompanied by outgassing of CO_2 in this post-bloom period. Such a major shift has been reported for the final observation period at the natural Kerguelen Plateau study (Lefèvre et al. 2008).

The estimates of export flux efficiency $(C:Fe)_{export\text{-}efficiency\text{-}100m}$ or $(C:Fe)_{export\text{-}efficiency\text{-}250m}$ are very modest compared to those that have been suggested in geoengineering proposals (C:Fe > 100 000) for reducing the greenhouse effect by removing CO_2 from the atmosphere (Buesseler et al. 2004). These efficiency values are in the range $650 < (C:Fe)_{export\text{-}efficiency} < 6600$; much lower than initial suggestions derived directly from assumed cellular composition ratio of plankton of C:Fe = ~100 000 or more (Table 2). The low export efficiencies may partly be ascribed to the fact that ~75% of the added fresh Fe(II) is removed rapidly into fine colloids (Nishioka et al. 2005) within 24 to 48 h after each fertilization:

$$[\text{Colloidal Fe}] = 0.76\,[\text{Dissolved Fe}] + 0.052 \quad (R^2 = 0.93) \qquad (4)$$

This removal is likely the first step towards the next stage of aggregation into larger particles, which then are lost due to settling into deeper water layers. This would appear consistent with ~63% of added Fe (1100 out of 1740 kg Fe) that cannot be accounted for in an Fe budget of SOIREE (Bowie et al. 2001). Such major loss does not only require repeated fertilizations, but also causes a ~4-fold decrease of efficiency. During

Table 3. Calculated values of efficiency ratio of C versus total added Fe in each experiment for total estimates over complete period of each experiment for net community production, CO_2 gas influx from the air, carbon export across 100 m depth horizon into deeper waters, and by extrapolation, export across 250 m depth horizon (Buesseler et al. 2004). Each value has large uncertainty. Compilation of best estimates following de Baar et al. (2005) with many incompatibilities between experiments and inherent uncertainties (de Baar et al. 2005, their Fig. 22 & Table 3). For SOFeX-South the $(\Delta DIC:Fe)_{NCP}$ estimate is integrated over a 225 km^2 patch area, while the $(\Delta DIC:Fe)_{NCP}$ Option 2 is integrated over a 1000 km^2 patch area, following Buesseler et al. (2004). For some other experiments, other $(\Delta DIC:Fe)_{NCP}$ options have also been published (de Baar et al. 2005, their Table 3, italics), but these were considered not the best estimates. If nothing else, the variations between presumed best estimates and other-option estimates for any one experiment underlines the intrinsic difficulty of quantification. Moreover, in some experiments it is not clear if the published changes of terms in the carbon budget are truly by difference between the fertilized in-patch and the control site (as here defined in Table 1, 'Complete equation'), or only for the fertilized in-patch, i.e. without correcting for simultaneous changes of terms in the the carbon budget at the unfertilized control site

Efficiency ratio	IronEx II	SOIREE	EisenEx	SEEDS I	SOFeX-South	SOFeX-North	SERIES
$(\Delta DIC:Fe)_{NCP}$	9672	3701	2847	15 000	8028	4110	16 887
$(\Delta DIC:Fe)_{NCP}$ Option 2	–	–	–	–	35 680	–	–
$(C:Fe)_{gas\text{-}flux\text{-}efficiency}$	630.5	100.8	219.0	853.7	1003.5	548.0	–
$(C:Fe)_{export\text{-}efficiency\text{-}100m}$	3150	Negligible	–	766	6648	–	650
$(C:Fe)_{export\text{-}efficiency\text{-}250m}$	–	–	–	–	3300	–	–

the Subarctic Pacific Iron Experiment for Ecosystem Dynamics Study (SEEDS) I experiment, the over 75% colloid Fe fraction tended to remain in the surface waters (Nishioka et al. 2003), at least during the 13 d of observations, nevertheless being colloids this fraction would remain inaccessible for direct uptake by phytoplankton.

Fine colloidal Fe is itself deemed unavailable for uptake by phytoplankton, but reduction by daytime photochemistry, or perhaps also in the vicinity of the cell wall due to membrane-bound reductases (Salmon et al. 2006), may bring colloidal Fe back into a soluble state suitable for plankton uptake. The diurnal physical chemistry of Fe colloids (Nishioka et al. 2005, Croot et al. 2005) requires further investigation.

Perhaps in future experiments, when adding dissolved Fe in a dissolved organic-complexed form (e.g. bound with excess EDTA), this could be overcome and 4-fold higher export efficiencies (ranging from 2600 < $(C:Fe)_{export\text{-}efficiency}$ < 27000) would not be inconceivable. As a matter of fact, this strategy has been used in 2 experiments (GreenSea 1 and 2) in the Gulf of Mexico, where researchers added extra lignopolysulfonate (cheap by-product of paper industry); however, the brief report does not have information on carbon export (Markels & Barber 2001).

C:Fe ratio derived from SEEDS II and the European Iron Fertilization Experiment (EIFEX)

The SEEDS II fertilization in 2004 was at the same location as SEEDS I, but had very different initial conditions and responses (Tsuda et al. 2007). This, in itself, illustrates the lack of predictability of ocean iron fertilization. Moreover, the fertilized patch was surrounded by a non-uniform water mass of different characteristics and this makes comparison between the fertilized patch and the control site less reliable (A. Tsuda pers. comm.). Upon addition of 322 kg Fe (5760 mol) in a 64 km^2 patch at Day T = 0 and an additional 160 kg (2842 mol) at Day T = 6 in a 200 km^2 patch, the particulate organic carbon (POC) export flux collected in a sediment trap at 100 m depth was variable, increasing from ~26 to ~35 mmol m^{-2} d^{-1}. At the control site the export flux at 100 m depth was lower at ~17 mmol m^{-2} d^{-1} (A. Tsuda & H. Saito unpubl. results). During the first 25 d, the total POC export at 100 m depth was 640 mmol m^{-2} in the fertilized patch and 423 mmol m^{-2} at the control site. By difference the excess POC export at 100 m depth was 217 mmol m^{-2}. Meanwhile, the detectable patch size increased to 1000 km^2 by Day 15 and then decreased to 830 km^2 by Day 22. This lateral patch dynamics is a major challenge for deriving carbon budgets and excess carbon export. Nevertheless, the excess POC export of 217 mmol m^{-2} would yield lower and upper limits for the export efficiency at 100 m depth in the range of ~1600 < $(C:Fe)_{export\text{-}efficiency\text{-}100m}$ < ~25 000 for an assumed 64 or 1000 km^2 patch size, respectively. This efficiency at 100 m depth in SEEDS II is more than the $(C:Fe)_{export\text{-}efficiency\text{-}100m}$ = 766 (Table 3) at 100 m depth in SEEDS I. An exciting finding of SEEDS II is the importance of mesozooplankton. The initial copepod biomass in SEEDS II was 3-fold higher than in SEEDS I and SERIES; the copepod biomass also increased strongly, some days earlier inside the patch than outside. On the other hand, an observed relationship between copepod biomass and export flux collected at 40 m depth in sediment traps shows no significant difference between in-patch and outside patch (out-patch) of SEEDS II (Tsuda et al. 2007). These observations suggest that the POC export flux collected in sediment traps at 40 m depth was a function of the copepod biomass and the iron-enhanced growth of phytoplankton had a minor effect on the export flux collected in 40 m depth sediment traps in SEEDS II (Tsuda et al. 2007). Moreover, vertical migration of mesozooplankton causes an extra carbon export flux which is not collected in sediment traps. In SEEDS II this vertical migration increases the excess carbon export at 40 m depth with ~70% (H. Saito unpubl. results) leading to an ~70% increase of the $(C:Fe)_{export\text{-}efficiency\text{-}40m}$ at 40 m depth. Similarly the $(C:Fe)_{export\text{-}efficiency\text{-}100m}$ at 100 m depth may increase due to vertical migration.

The 2004 EIFEX in the Southern Ocean (Hoffmann et al. 2006) was twice fertilized with 25 180 mol Fe, once in a 150 km^2 patch and once in a 400 km^2 patch. Currently, we have no information on whether or not and to what extent the second fertilization overlapped with the water mass of the first fertilization. In between the first and the second fertilization, the size of the patch was reportedly observed to be ~600 km^2. Initial export fluxes at 150 m depth were high at ~21 ± 5 mmol C m^{-2} d^{-1} and remained high and constant at both in-patch and out-patch stations during the first 21 d (Jacquet et al. 2008, their Fig. 3). Export production increased slightly during Day 17 to Day 34 at out-patch stations, while at in-patch stations it decreased to the lowest values at Day 20 to Day 24. From Day 25 to Day 32, a massive increase of carbon export to as high as ~54 ± 14 mmol C m^{-2} d^{-1} occurred at the in-patch stations (Jacquet et al. 2008, after their Fig. 3), coinciding with massive sinking of large diatoms. During that final period, the excess total flux appears to have been on the order of ~350 mmol C m^{-2}, which, with an assumed average patch size of ~400km^2 and a total Fe addition of 50 360 mol would lead to an estimate of $(C:Fe)_{export\text{-}efficiency\text{-}150m}$ = ~2780 for EIFEX.

Values of C:Fe ratio in natural fertilization

The Fe supply in natural Fe fertilizations at both the Kerguelen plateau and the Crozet Island plateau comes from suboxic or anoxic sediments. These sediments can be just below the surface waters exhibiting enhanced phytoplankton, i.e. there may be an upward transport of dissolved Fe from the sediments to the surface waters, as suggested for the Kerguelen Plateau (Blain et al. 2007). Alternatively, dissolved Fe from sediments at an adjacent island plateau might be transported laterally by horizontal currents and water mass mixing to a site with observed enhanced phytoplankton, as suggested for the Crozet Island region (Planquette et al. 2007). Due to the >99% organic complexation (Gerringa et al. 2008), this Fe is maintained in solution far better than the artificial fresh Fe(II) additions of the *in situ* fertilization experiments, where within 12 to 48 h some 75% of the added Fe (II) is lost very rapidly due to oxidation into colloids (Eq. 4) which in turn may aggregate into large particles. Hence, for natural fertilization the efficiency ratio values may well be on the order of 4-fold higher (or more) than for the experimental fertilizations.

Values of C:Fe in natural fertilization at the Crozet Plateau

At the Crozet Plateau, high carbon export rates have been reported from sediment trap collection. Normalized to 100 m depth these export rates range from ~1 to ~36 mmol C m^{-2} d^{-1} in the productive region north of the plateau and ~2 to 4 mmol C m^{-2} d^{-1} in the HNLC control region south of the plateau (Salter et al. 2007). These values are in good agreement with independent estimates by the ^{234}Th deficiency method, ranging from initially 15 mmol C m^{-2} d^{-1} in the high-chlorophyll bloom region, compared with 5 mmol C m^{-2} d^{-1} in the low-chlorophyll, non-bloom region (Morris et al. 2007). After a moderately small bloom and export event at the southern control stations, the POC export became more uniform, resulting in equally high levels of POC export (~20 mmol C m^{-2} d^{-1}) throughout the study region.

The magnitude of Fe influx from various sources is very difficult to quantify. For the blooming area of the Crozet Natural Iron Bloom and Export Experiment (CROZEX) the soluble Fe influx from aerosols has been assessed at 100 nmol m^{-2} d^{-1}, the vertical influx from below sediments at 61 nmol m^{-2} d^{-1}, and the horizontal influx from the nearby islands at 390 nmol m^{-2} d^{-1} (Planquette et al. 2007). Each of these values is based on a suite of estimates and inherent assumptions (see Planquette et al. 2007); assuming that the atmospheric and vertical fluxes are also present at the HNLC control site, and that the horizontal flux continues for 100 d of winter, then the estimated stock of Fe available at the start of spring is equivalent to 0.039 mmol m^{-2} yr^{-1} (Pollard et al. unpubl. data).

Next, by comparing the ^{234}Th-deficiency-derived export (mmol m^{-2} d^{-1}) of biogenic Si (i.e. diatoms) with the seasonal deficit (mmol m^{-2}) of dissolved silicate in the upper water layer, Pollard et al. (unpubl. data) arrive at an estimated bloom duration of 61 d. This yields an excess C export of the productive bloom region compared to the low production control region of 670 mmol m^{-2} per growing season, which, divided by the excess Fe supply of 0.039 mmol m^{-2} yr^{-1} yields their reported best estimate efficiency $(C:Fe)_{export\text{-}efficiency\text{-}100m}$ = ~17 200 for export across the 100 m depth horizon (Pollard et al. unpubl. data). Furthermore, by using upper and lower limits of C export and Fe supply, upper and lower limits of efficiency of $(C:Fe)_{export\text{-}efficiency\text{-}100m}$ = ~60 400 and ~5400, respectively, are achieved (Pollard et al. unpubl. data).

The vertical POC flux collected in a deep sediment trap at 3000 m depth was found to have only 4% of the above-mentioned downward-POC flux across the 100 m depth horizon, i.e. 96% of the downward-settling POC flux was lost in the 100 to 3000 m depth interval. This is consistent with the expectations mentioned in the above subsection 'Parameterization — Fate in deep ocean waters' for the fate of POC in the deep ocean and yields an efficiency ratio at 3000 m of $(C:Fe)_{export\text{-}efficiency\text{-}3000m}$ = ~730. For intermediate depths in the 100 to 3000 m range the efficiency ratio would be between ~17 200 and ~730. When combining this with an ocean circulation model, the *a priori* expectation is that the deepest regenerated CO_2 (with lowest C:Fe efficiency) will have the longest ocean storage time, while the major regeneration in the 100 to 1000 m depth 'ocean twilight zone' (Buesseler et al. 2007), with its relatively high C:Fe efficiency, will have shorter deep ocean storage time. Thus for a range of depth intervals the more favorable C:Fe efficiency tends to be coupled with less favorable storage time. However, regional circulation and mixing — as well as rapid deep sedimentation events with or without enhancement-by-ballast effects of Si opal and $CaCO_3$ skeletons — will cause deviations from this simple *a priori* expectation.

Natural fertilization at the Kerguelen plateau

The Kerguelen plateau consistently shows elevated chlorophyll *a* (chl *a*) levels (as observed by satellite) and this 'ocean island effect' has been investigated in KEOPS (Blain et al. 2007). The diffusive upward flux of Fe from sediments below was 31 nM m^{-2} d^{-1} (partly due to

Table 4. Ratio values of plankton uptake rate, plankton content, and various efficiency values derived for the KEOPS field study of natural fertilization processes (Blain et al. 2007). The reported 200 000 ± 118 000 mol mol^{-1} value (Blain et al. 2007) apparently slightly deviates from the actual value of 274 000 ± 142 000 mol mol^{-1} (range is 131 000 to 416 000) in the more recently published article of the shipboard experiments (Sarthou et al. 2008). Export efficiencies at 100 and 200 m reported by Blain et al. (2007); alternative export efficiency values (Invoked 2) calculated in this study. Ratio values C:Fe of plankton uptake rate and plankton content after inversion of ratio values Fe:C reported by Sarthou et al. (2008). Also shown is an Invoked 3 scenario for sake of comparison with diatom C:Fe composition under optimal Fe-replete conditions, see 'Discussion—Concordia'

Ratio values	KEOPS	KEOPS Invoked 1 (Blain et al. 2007)	KEOPS Invoked 2 (present study)	KEOPS Invoked 3 (present study)
Plankton uptake rate				
$(C:Fe)_{plankton\text{-}uptake\text{-}rate}$	–	200 000 ± 118 000[a]	–	–
	–	131 000 to 416 000 (n = 6)[b]	–	–
Plankton content				
$(C:Fe)_{plankton\text{-}content}$	–	227 000 ± 5000 (n = 10)[b]	–	–
		175 000 to 333 000		
$(C:Fe)_{large\text{-}diatoms\text{-}Fe\text{-}limited}$	–	–	~160 000[c]	–
$(C:Fe)_{large\text{-}diatoms\text{-}optimal}$	–	–	–	~23 000[c]
Efficiency ratios				
Excess Fe supply rate (nM m^{-2} d^{-1})	27	204 ± 77 (Invoked 1)	~289 (Invoked 2)	~2013 (Invoked 3)
$(\Delta DIC:Fe)_{NCP}$	668 000	–	–	–
	Seasonal budget	–	–	–
$(\Delta DIC:Fe)_{NCP}$	–	149 000 Seasonal budget including lithogenic Fe supply	–	–
$(C:Fe)_{export\text{-}efficiency\text{-}100m}$	400 000 ± 180 000	21 000 to 123 600	16 800 to 98 880	2415 to 14 214
$(C:Fe)_{export\text{-}efficiency\text{-}200m}$	526 000 ± 290 000	70 000 ± 46 000	56 000 ± 37 000	8050 ± 5290
		22 400 to 174 000	17 920 to 193 200	2576 to 20 010

[a]Blain et al. (2007); [b]Sarthou et al. (2008); [c]Twining et al. (2004)

enhanced vertical diffusivity) as compared to 4 nM m^{-2} d^{-1} at the off-plateau control site. The mean excess of ^{234}Th-derived export production on the plateau versus at the control site was 10.8 ± 4.9 mmol C m^{-2} d^{-1} at 100 m depth and 14.2 ± 7.9 mmol C m^{-2} d^{-1} at 200 m depth. Dividing by the excess upward diffusive Fe flux of 27 nM m^{-2} d^{-1} leads to $(C:Fe)_{export\text{-}efficiency\text{-}100m}$ = 400 000 and $(C:Fe)_{export\text{-}efficiency\text{-}200m}$ = 526 000. Presumably these record high export efficiencies were deemed non-realistic by the authors. Therefore, an additional Fe supply term was invoked. Shipboard ^{55}Fe uptake and regeneration experiments would lead to a hypothesized excess Fe input of 204 ± 77 nM m^{-2} d^{-1} over the plateau, but the source of this excess Fe is unaccounted for. With the same above carbon export at 200 m depth, we calculate a range of 22 400 < $(C:Fe)_{export\text{-}efficiency\text{-}200m}$ < 174 000 — not dissimilar to the reported $(C:Fe)_{export\text{-}efficiency\text{-}200m}$ = 70 000 ± 46 000 (Blain et al. 2007).

In general, the reported efficiency values of the KEOPS (Table 4) natural environment exceed those reported from the *in situ* experiments. Taking into account the 75% loss term in the latter experiments, we simply (if not simplistically) multiply the $(\Delta DIC:Fe)_{NCP}$ values (Table 3) of *in situ* experiments by a factor of 4 and arrive at a range of 11 388 to 67 548 from low EisenEx to high SERIES. This range is still well below the range 149 000 to 668 000 for the reported $(\Delta DIC:Fe)_{NCP}$ seasonal budget estimates of KEOPS (Table 4). Similarly, multiplying the export efficiency values 4-fold, we obtain a range of 2600 (SERIES at 100 m) to 26 592 (SOFeX-South at 100 m), which just overlaps with the wide range from 21 000 to 916 000 for various export efficiency estimates of KEOPS (Table 4). Apart from this somewhat arbitrary 4-fold adjustment, there remains the challenge to

Table 5. Export efficiency at 100 m depth during SEEDS II, estimated after A. Tsuda et al. (unpubl. data), and the export efficiency at 150 m during EIFEX following Smetacek (2005) and Jacquet et al. (2008) for the two 7 t iron-sulphate ($FeSO_4.7H_2O$) enrichments (in ~150 km^2 and ~400 km^2 patches). One mol $FeSO_4.7H_2O$ equals 278 g, 1 mol Fe equals 55.847 g. Upon publication of findings for dissolved inorganic carbon (DIC) and pCO_2, differences between in-patch and out-patch stations the $(\Delta DIC:Fe)_{NCP}$ and $(C:Fe)_{gas\text{-}flux\text{-}efficiency}$ may also be assessed

Efficiency ratio	SEEDS II	EIFEX
$(C:Fe)_{export\text{-}efficiency\text{-}100m}$	~1600–25000	–
$(C:Fe)_{export\text{-}efficiency\text{-}150m}$	–	~2780

bridge the gap between the very high efficiency ratio values reported for KEOPS (Blain et al. 2007) and lower efficiency ratio values for the artificial fertilizations (Tables 3 & 5). Below, we will show how these, at first glance inconsistent, findings can be reconciled.

DISCUSSION

Concordia

The excess Fe input of 204 ± 77 nM m^{-2} d^{-1} over the Kerguelen plateau invoked on the basis of shipboard ^{55}Fe uptake experiments, inevitably yields high C:Fe efficiencies which are consistent with the ratio values of these shipboard ^{55}Fe-based experiments (Blain et al. 2007). In other words, the reported (Blain et al. 2007) consistency of high efficiencies of short and seasonal budgets with shipboard observations of C:Fe uptake rate ratios (200 000 ± 118 000 mol mol^{-1}) is a circular argument. Otherwise, the latter 200 000 ± 118 000 mol mol^{-1} value apparently slightly deviates from the actual value of 274 000 ± 142 000 mol mol^{-1} (range is 131 000 to 416 000) in the more recently published article of the shipboard experiments (Sarthou et al. 2008). Nevertheless, the plankton content ratio range 175 000 < $(C:Fe)_{plankton\ content}$ < 333 000 reported for the Kerguelen plateau is not dissimilar to the $(C:Fe)_{large\text{-}diatoms\text{-}Fe\text{-}limited}$ = ~160 000 for large diatoms of the unfertilized control site of SOFeX-South (Table 2) as determined by the reliable synchrotron X-ray method (Twining et al. 2004). This value of ~160 000 now may be used as a second approach for another invoked Fe supply scenario leading to efficiency ratio values (Table 4, KEOPS Invoked 2) similar (but somewhat lower) to the first invoked scenario (Table 4, KEOPS Invoked 1). As the result of this second invoked scenario one finds similar efficiency ratio values for the export, conversely suggesting the large diatoms at Kerguelen Plateau were as Fe-limited as at the SOFeX oligotrophic control site. Finally, when assuming truly Fe-replete large diatoms, one may invoke the best estimate, $(C:Fe)_{large\text{-}diatoms\text{-}optimal}$ = ~23 000 (Table 2), and thus arrive at much lower C:Fe efficiency values for the Invoked 3 scenario (Table 4). The values obtained in this fashion, ranging from 2415 to 20 010, are closer to the 650 to 6648 export efficiency of the artificial *in situ* experiments (Table 3). Moreover, when taking into account the ~75% immediate loss of added Fe in the *in situ* fertilization experiments, multiplying the latter export efficiencies by 4 would yield a range of 2600 to 25 600, which overlaps with the Invoked 3 scenario of a 2415 to 20 010 export range at KEOPS for truly Fe-replete diatoms driving this export.

Indeed, the mean dissolved Fe concentration in the surface waters over the Kerguelen plateau is low (0.090 ± 0.034 nM) and only slightly higher than at the oligotrophic control site (0.073 ± 0.014 nM) (Blain et al. 2008). These dissolved Fe concentrations are similar to those found elsewhere in the oligotrophic Antarctic Ocean; for example, 0.04 to 0.06 nM at the beginning of EisenEx (Nishioka et al. 2005). At the out-patch stations of SOFeX-South, values were somewhat higher, at ~0.1 nM (and perhaps occasionally higher, at ~0.5 nM), and at these out-patch stations, $(C:Fe)_{large\text{-}diatoms\text{-}Fe\text{-}limited}$ values = ~100 000 to 160 000 were found. In contrast, the fertilized patch of SOFeX-South had a typical dissolved Fe of ~1.2 nM, an order of magnitude higher than at Kerguelen Plateau, thus stimulating more rapid growth of large diatoms. Indeed, the cell division rates of key large diatom species in seawater of the Kerguelen Plateau are ~0.2 d^{-1} (Timmermans et al. 2008) which is about half of the optimal rate of growth (Timmermans et al. 2004).

Nevertheless, at the Kerguelen Plateau, the slight difference of ~0.02 nM extra dissolved Fe compared to the SOFeX-South out-patch station, as well as the likely more steady supply from below, would be beneficial for long term maintenance of an elevated abundance and elevated rate of growth (but by no means optimal in the sense of Monod growth). The F_v/F_m values, indicative of more or less adequate supply of Fe for healthy state of growth, were reported somewhat higher, at 0.55 over the Plateau, as compared to ~0.3 at the KEOPS control site (Blain et al. 2007, their supplement). Over the complete spring and summer season, a modest increase of cell division rate (say from 0.15 d^{-1} to 0.2 d^{-1}) may yield the 'cumulative interest' effect for the significant difference between steady blooms at Kerguelen versus the generally low chlorophyll abundance (HNLC) of open Antarctic Ocean waters. In other words, it appears that the large diatoms in surface waters of the Kerguelen plateau are only somewhat less Fe-limited than elsewhere in the open Antarctic Ocean, yet nevertheless, by steady moderate rate of growth, do make an impact on budgets of C and Si. This might explain the reported very high, 175 000 < $(C:Fe)_{plankton\text{-}content}$ < 333 000, values for algae growing in Fe = ~0.09 nM surface waters of Kerguelen Plateau, as compared to the $(C:Fe)_{large\text{-}diatoms\text{-}optimal}$ = ~23 000 in the Fe-replete ~1.2 nM waters of the in-patch of SOFeX-South. Indeed, in the Fe-enriched patch of SOFeX-South, the F_v/F_m was at its virtual theoretical maximum ratio, ~0.65 (Coale et al. 2004).

Uncertainties

For both the *in situ* fertilization experiments and the natural fertilizations the approaches or methodologies

for quantification of the carbon C stocks and turnover rates are the same or similar, hence the inherent uncertainties likely are similar. On the other hand, the quantifications of the Fe supply, stocks and turnover rates are vastly different. For the *in situ* experiments, the total amount of added Fe is accurately known, but the fate of this Fe is very poorly quantified due to some 60 to 75% being lost, somehow, somewhere. Budget estimations of the fate of added Fe are difficult and only one budget assessment has been published (Bowie et al. 2001). For the natural fertilization experiments, the quantification of Fe supply from below sediments is difficult and in KEOPS was shown to be far lower than desirable.

Ranges of estimates

The oceans are dynamic regions, with much variability in time and place, where day-to-day variability of the weather (wind, insolation) is a major driver of variability of any one upper ocean plankton ecosystem. In every single fertilization experiment or natural fertilization study, the complete 3D mapping every day of all key variables cannot be achieved with merely 1 (or at most 3) ships and their shipboard observers. For each experiment the ensuing dataset, therefore, is very limited in space, in time, and in a very restricted number of key variables. Thus, we are only scratching the surface. Nevertheless, for each study, the overarching synthesis article (e.g. Tsuda et al. 2003, 2007, Boyd et al. 2004, Coale et al. 2004, Blain et al. 2007) very cleverly combines these limited observations into an overall coherent interpretation, based on the underlying more specific reports (e.g. Buesseler et al. 2004, Nishioka et al. 2005, Gerringa et al. 2008) of that 1 study.

Previously (de Baar et al. 2005, Boyd et al. 2007) and in this article, an effort has been made to combine these individual field projects (IronEx II through CROZEX, KEOPS) into a synthesis, where inevitably its conclusions carry the sum of all uncertainties of the single studies. Moreover, the various estimates as cited here from work by others, as well as in this article, are all based on many inevitable assumptions, as required due to lack of data or knowledge, where a large number of these assumptions are highly debatable, if not questionable. In other words, due to the limited datasets, there is much room for alternative interpretations and conclusions.

CONCLUSIONS

The most reliable estimate of $(C:Fe)_{large\text{-}diatoms\text{-}optimal}$ is to date ~23 000 for *in situ* large Antarctic diatoms within the Fe-replete SOFeX-South patch. This is in good agreement with the diffusion limitation (de Baar et al. unpubl. data) of the rate of growth of large Antarctic diatoms in shipboard and laboratory experiments of incubations of single species diatoms in filtered ultraclean natural Antarctic seawater (Timmermans et al. 2004). In the Fe-limited, suboptimal growth conditions of the open Antarctic Ocean, large diatoms survive at very low, but likely persistent, rates of growth and may have lower suboptimal intracellular C:Fe ratio values, from ~160 000 at out-patch stations of SOFeX-South to ~227 000 in the ambient 0.09 nM Fe surface waters of Kerguelen Plateau.

In the artificial Fe fertilization experiments, the 4 estimates thus far of $(C:Fe)_{gas\text{-}flux\text{-}efficiency}$ range from 100 to 1000 (Table 3). In the period after observation ships have left, this efficiency may either increase or decrease, due to either continued net community production or a shift to final net community respiration, respectively.

The estimates of carbon export efficiency at 100 m depth range from 650 (SERIES) to 6648 (SOFeX-South) with an extrapolated value at 250 m of ~3300 for SOFeX-South (Tables 3 & 5). For EIFEX the export efficiency at 150 m depth is estimated at ~2780 (Table 5).

For these artificial fertilizations the ~75% immediate loss of added Fe may be corrected for by a factor of ~4 when assuming future or natural fertilization as a more stable Fe-organic complex. This would increase the CO_2 drawdown efficiency in the 400 to 4000 range and the various export efficiency estimates to a range from ~2600 (SERIES) to ~26 600 (SOFeX-South), or even ~100 000 when including the high estimate of SEEDS II based on an assumed very large 1000 km^2 patch size.

For the artificial *in situ* Fe fertilization experiments, at least the amount of added Fe is accurately known, such that the uncertainty in the above estimations of C:Fe efficiency is due solely to uncertainty in the assessment of the C budgets. For natural Fe fertilizations the supply of Fe also must be derived from field observations, such that the derived supply of Fe has an uncertainty equal or larger than the uncertainty in the parallel estimation of the C budget. Therefore, the $(C:Fe)_{export\text{-}efficiency}$ of natural fertilizations has an inherently larger uncertainty than that of the artificial fertilization experiments.

In the natural Fe fertilization at the Crozet Plateau the $(C:Fe)_{export\text{-}efficiency}$ at 100 m depth was reported to be ~17 200, within reported upper and lower estimates of ~60 400 and ~5400, respectively (Pollard et al. 2008). In the natural Fe fertilization at the Kerguelen Plateau, the derived $(C:Fe)_{export\text{-}efficiency}$ at either 100 m or 200 m depth ranged from ~8050 to ~526 000, largely depending on the assumed or invoked Fe supply term.

RECOMMENDATIONS

When delivering the iron fertilizer in a more stable organic-complexed dissolved form, the efficiency of carbon export into deeper ocean waters may range from ~2600 to ~26 600 per added amount of iron. Laboratory research is needed to unravel which specific Fe-organic complexing chemical molecule would both stabilize Fe in solution and leave it available for uptake by the target group of phytoplankton, e.g. large diatoms. This will be influenced by photochemistry (notably Fe photoreduction) which should be part of such studies.

The wide variability of export efficiency is due to wide variations of initial ocean conditions before iron addition and wide variation of weather conditions after iron addition. Initial conditions of major influence are the initial abundance of phytoplankton and its major taxonomic groups and size classes, micro- and mesozooplankton, water column stratification, and temperature. For nutrient-rich HNLC regions, the initial concentrations of major nutrients are deemed in ample supply, yet the weather strongly controls the light climate for photosynthesis, not only by the variations of cloud cover controlling incoming solar irradiance at the ocean surface, but perhaps even more by wind forcing algae to be mixed deeper down, where light conditions are unfavorable. This strong weather-dependence renders the outcome of commercial-scale OIF unpredictable, its statistics more akin to gambling than prudent financial investment. Moreover, even the very highest efficiency value of ~25 000 is far less than the first-hypothesized values in the ~500 000 range and OIF business plans should at least use a 20-fold higher amount of Fe than initially hypothesized.

The iron fertilization experiments done thus far were by no means designed for assessing commercial-scale OIF. When pursuing commercial-scale OIF, far more extensive quantification of carbon export (e.g. by far more intensive ^{234}Th mapping, many more sediment trap deployments, or other methods yet to be designed and validated) is required for accurate accountability of the carbon storage, which in turn is required for appropriate accountability of intended financial carbon credits.

Beyond the efficiency of CO_2 influx from the atmosphere into the ocean and efficiency of export below a defined 100 m or 250 m depth horizon, there are additional factors beyond the scope of this paper (some even beyond the scope of oceanography). These have been considered elsewhere and in companion articles of this Theme Section and include (1) longevity of deep ocean storage in relation to the ventilation time of underlying waters (after how many years, decades, or perhaps centuries, will the temporarily stored deep ocean CO_2 surface again and ventilate back into the atmosphere); (2) negative feedbacks (i.e. enhanced production of stronger greenhouse gases methane and nitrous oxide); (3) the financial and energy expenditure and CO_2 emission (e.g. by ships) of OIF; (4) environmental protection; (5) international treaties (the London 'Dumping' Convention, Antarctic Treaty, etc.); and last, but not least, (6) the ethics of OIF in, for example, the precautionary principle and the transgenerational principle. Briefly, even when ignoring the latter two factors (international treaties and ethics) perhaps the question to ask a proponent of OIF is whether or not one would invest his or her own personal savings in an OIF enterprise, or advise his or her children to invest their money as a means of securing their personal future financial security.

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