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# Storage of Carbon Dioxide by Greening the Oceans?

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The widespread use of fossil energy results in the release of the greenhouse gas carbon dioxide ($CO_2$) to the atmosphere at an average rate of 5.9 petagrams of carbon per year (Pg C $yr^{-1}$; 1 Pg = $10^{15}$ g) (Sabine et al., Chapter 2, this volume). The resulting rapid increase of atmospheric $CO_2$ is a major factor in global warming (Houghton et al. 2001). Because of the potential risks of climate change, it is crucial to consider how to curb fossil-fuel $CO_2$ emissions and whether techniques are available to absorb atmospheric $CO_2$. Such a consideration requires weighing the economic, societal, and environmental costs of climate change relative to those of reducing $CO_2$ emissions and implementing $CO_2$ sequestration.

The oceans absorb roughly 30 percent of contemporary $CO_2$ emissions caused by human activity (Sabine et al., Chapter 2, this volume). Sluggish exchange between surface waters and the deep ocean sets the maximum rate for net oceanic $CO_2$ uptake and makes it a slow process relative to the rapid anthropogenic $CO_2$ release. If anthropogenic $CO_2$ emissions were stopped now, the oceans would eventually take up 70–80 percent of the $CO_2$ emissions, over a period of several centuries (Maier-Reimer and Hasselmann 1987; Archer et al. 1997).

Past and current oceanic $CO_2$ uptake reduces the capacity for future oceanic $CO_2$ absorption (Matear and Hirst 1999). It also influences oceanic pH (Archer et al. 1998), calcification (Gattuso et al. 1998; Riebesell et al. 2000), and the marine ecosystem (Houghton et al. 2001). Warming of surface waters and possible changes in oceanic circulation caused by climate change will have indirect effects on the physical and biological carbon pumps in the oceans (Sarmiento et al. 1998; Matear and Hirst 1999). These changes will provide positive, as well as negative, feedbacks on oceanic $CO_2$ uptake, the atmospheric $CO_2$ content, and climate change.

Because the oceans absorb so much of the $CO_2$ emitted by human activity, it is not surprising that scientists, entrepreneurs, and politicians are investigating whether it is possible to accelerate net oceanic $CO_2$ absorption. One possible technique is fertilization of ocean regions where low nutrient availability currently limits phytoplankton growth. Addition of the macronutrients nitrogen and phosphorus and the trace element iron has been suggested. In this chapter I consider the principle, efficiency, duration, costs, and environmental consequences of $CO_2$ storage by fertilization.

## Iron Limitation of Algal Growth

The idea of iron fertilization has received wide attention since John Martin (1990a: 11) commented, "With half a shipload of iron, I could give you an ice age." Large areas of the oceans, notably the Southern Ocean, the equatorial Pacific Ocean, and the subarctic Pacific Ocean, have low algal growth, despite high year-round nutrient concentrations in surface water (Figure 26.1). Hart (1934) suggested that low iron levels might limit phytoplankton growth in Antarctic waters.

Dust is a major iron source for the open oceans. Dust input to the oceans strongly depends on the proximity of dust sources on land and prevailing wind patterns. Ocean regions with high nutrient concentrations and low algal growth receive relatively little atmospheric dust (Duce and Tindale 1991; Watson 2001). An increase of atmospheric $CO_2$ upon a reduction in dust deposition at the end of glacial periods suggests a feedback between dust-derived iron, algal growth in the oceans, atmospheric $CO_2$, and climate (Martin 1990b; Watson et al. 2000).

The arrival of accurate methods for iron analysis allowed tests and confirmation of the iron hypothesis (De Baar et al. 1990; Hudson and Morel 1990; Martin et al. 1990). In situ iron enrichment experiments have been conducted in the equatorial Pacific Ocean (IronEx I, II), the Southern Ocean (SOIREE, EisenEx, SOFeX), and the subarctic Pacific Ocean (SEEDS, SERIES) (Table 26.1). These mesoscale experiments were started only after extensive site surveys. The rapid subduction of the fertilized waters in IronEx I demonstrates the critical importance of site selection for these experiments. Iron was added once in some experiments and repeatedly in others. Inert tracer, drifting buoys, and ADCP (Acoustic Doppler Current Profiler) measurements were used to track the iron-enriched waters.

The in situ iron additions promoted development of an algal bloom (Figure 26.2a–d) (Martin et al. 1994; Coale et al. 1996; Boyd et al. 2000; Smetacek 2001; Johnson et al. 2002; Tsuda et al. 2003). Diatoms, large algae with siliceous shells, thrived upon iron addition (Coale et al. 1996; Boyd et al. 2000), whereas no changes in calcifying algae have been recorded. Algal growth decreased the concentrations of nutrients and $CO_2$ in the mixed layer (Table 26.1, Figure 26.2e-f). The algal $CO_2$ uptake either reduced $CO_2$ release to the atmosphere or increased the uptake of atmospheric $CO_2$ by surface waters (Cooper et al. 1996; Watson et al. 2000), depending on their $CO_2$ saturation level (Figure 26.2e).

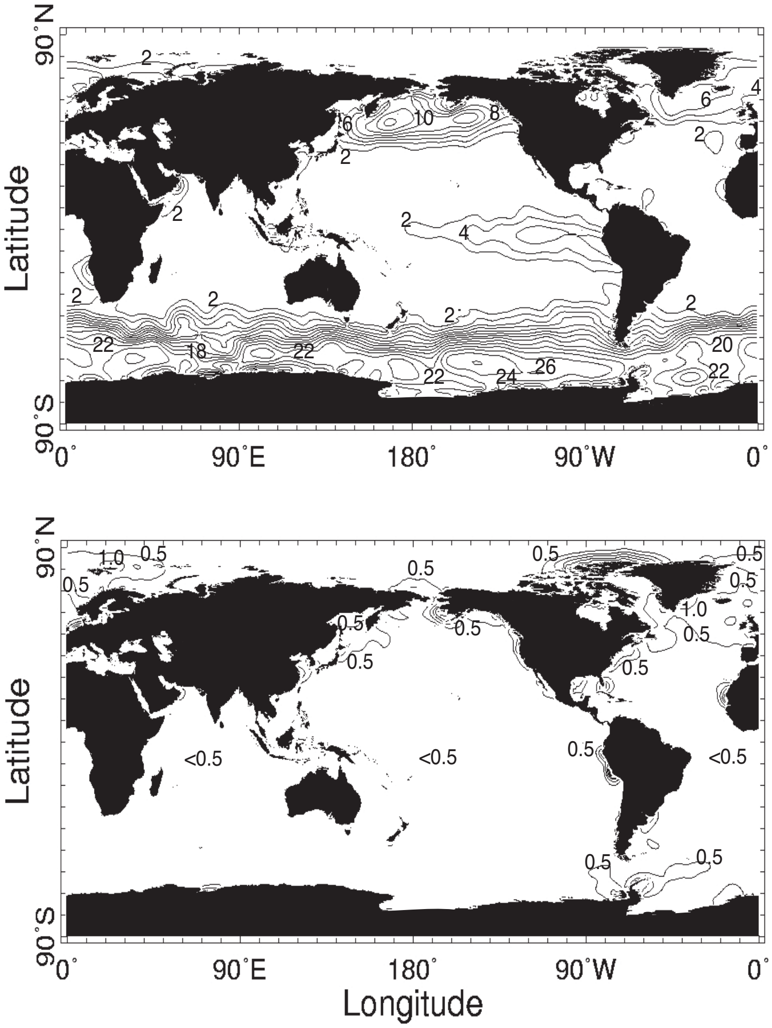

**Figure 26.1.** Seasonally averaged concentrations of nitrate and chlorophyll in oceanic surface waters with contours at 2 µmol $l^{-1}$ and 0.5 µmol $l^{-1}$ intervals, respectively. Data are from Conkright et al. (1998a, b, c).

Iron-induced algal growth resulted in the production of particulate organic carbon (POC) (Bidigare et al. 1999; Nodder and Waite 2001) and presumably also of dissolved organic carbon (DOC). A carbon budget for the SOIREE bloom suggests significant production of DOC (Bakker et al. 2003). The quantity and quality of DOC production following iron fertilization remain uncertain.

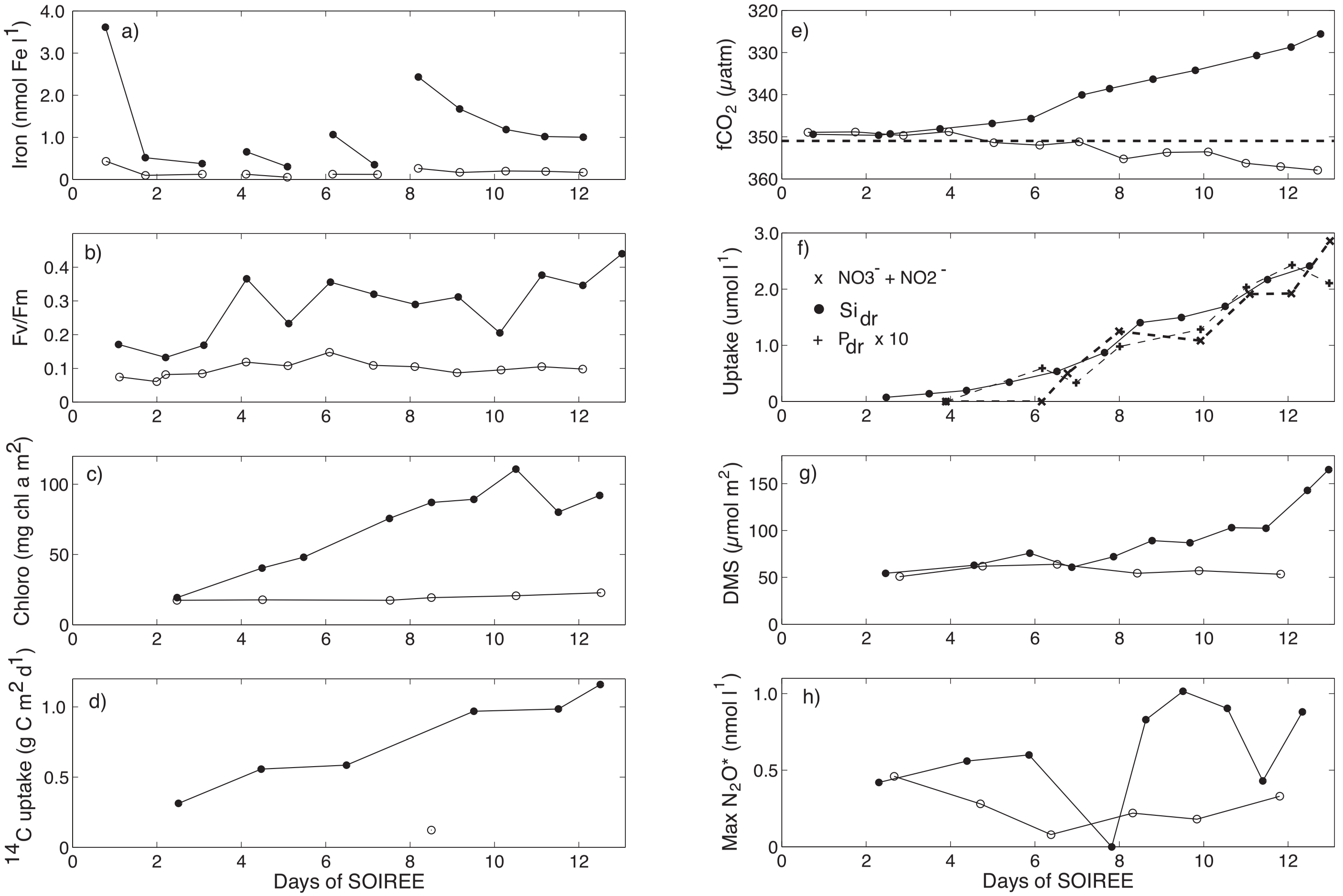
a)
Iron (nmol Fe l-1)
4.0
3.0
2.0
1.0
0
b)
Fv/Fm
0.4
0.3
0.2
0.1
0
c)
Chloro (mg chl a m2)
100
50
0
d)
14C uptake (g C m2 d-1)
1.0
0.5
0
0
2
4
6
8
10
12
Days of SOIREE
e)
fCO2 (μatm)
320
330
340
350
360
f)
Uptake (umol l-1)
x NO3- + NO2-
● Sidr
+ Pdr x 10
3.0
2.0
1.0
0
g)
DMS (μmol m2)
150
100
50
0
h)
Max N2O* (nmol l-1)
1.0
0.5
0
Days of SOIREE

**Figure 26.2.** Parameters inside (closed symbols) and outside (open symbols) the iron-fertilized waters (or "patch") in the Southern Ocean Iron RElease Experiment (after Boyd et al. 2000; Watson et al. 2000; Law and Ling 2001). Shown are (a) dissolved iron in surface water, (b) photosynthetic competency (Fv/Fm) in surface water, (c) depth-integrated chlorophyll, (d) depth-integrated $^{14}C$ uptake, a measure for primary production, (e) the fugacity of $CO_2$ ($fCO_2$) in surface water and air (dashed line, mean value of $fCO_2$ in air for SOIREE), (f) macronutrient uptake in surface water, (g) depth-integrated dissolved DMS (dimethyl sulphide) and (h) maximum $N_2O^*$ (nitrous oxide) in the pycnocline. Samples for dissolved iron were taken at roughly 3 m depth, samples for Fv/Fm, $fCO_2$ and nutrients at ~5 m depth. Underway data for dissolved iron, photosynthetic competency, and nutrients are expressed as the daily mean inside the patch (defined at>50 percent of peak levels of the tracer sulphur hexafluoride [$SF_6$] on that day) and outside the patch (defined at an $SF_6$ concentration <10 fmol $l^{-1}$). The previously reported data for $^{14}C$ uptake have been corrected by multiplication with a factor 12/14. Vertically integrated data have been integrated for the upper 65 m, the depth of the mixed layer. $Si_{dr}$ and $P_{dr}$ are dissolved reactive silica and phosphorus. Nitrate and phosphate depletion were obtained by subtraction of levels in the patch from values in ambient waters, which remained constant during SOIREE. Underway data for $fCO_2$ are indicated as a daily mean of $fCO_2$ measurements inside and outside the patch (obtained from $fCO_2$ at the 10 percent highest and 10 percent lowest $SF_6$ values, respectively). Note the reversed *y* axis for $fCO_2$. Surface water $fCO_2$ outside the patch increased during SOIREE, partly as the result of surface water warming. Maximum $N_2O^*$ is the difference between the measured $N_2O$ concentration (inside or outside the patch) and that predicted from the $N_2O$-density relationship outside the patch (Law and Ling 2001).

**Table 26.1.** The amount of added iron (Fe) and the observed drawdown of dissolved inorganic carbon (DIC) in the mixed layer during three iron fertilization experiments

| *Experiment* | *IronEx I* | *IronEx II* | *SOIREE* |
|---|---|---|---|
| Area | Equatorial Pacific | Equatorial Pacific | Southern Ocean |
| Time | 10/1993 | 05–06/1995 | 02/1999 |
| Fe added ($10^3$ mol) | 7.8[a] | 8.0[c] | 31[g] |
| –ΔDIC (µmol $kg^{-1}$) | 6–7[b] | 27[d] | 18[h] |
| –ΔDIC patch ($10^8$ mol) | n.d. | 2.4[e] (13–16 d) | 0.3–1.1[h, i] (13 days),<br>0.5–2.5[j] (42 days) |
| –ΔC:ΔFe ($10^4$) | n.d. | 3.0 | 0.11–0.35 (13 days),<br>0.16–0.81 (42 days) |
| Downward C transport | Subduction after 2–3 days | 7-fold increase in export[f] | Constant export[k, l] |

*Note:* Note that DIC drawdown in the mixed layer strongly overestimates the short-term (<1 year) storage of atmospheric $CO_2$, as significant remineralization of organic carbon is likely to occur in the mixed layer before $CO_2$ has been taken up from the atmosphere by slow air-sea exchange. –ΔC:ΔFe is the ratio of observed DIC drawdown to added iron. N.d. indicates parameters, which were not determined.

*Sources:* [a] Martin et al. (1994), [b] Watson et al. (1994), [c] Coale et al. (1996), [d] Steinberg et al. (1998), [e] Cooper et al. (1996), [f] Bidigare et al. (1999), [g] Bowie et al. (2001), [h] Bakker et al. (2001), [i] Boyd et al. (2000), [j] Abraham et al. (2000), [k] Charette and Buesseler (2000), [l] Nodder and Waite (2001).

## Carbon Storage by Iron Addition

Iron-induced algal growth, the composition of organic matter, downward carbon transport, remineralization, and future ventilation of the water jointly determine the efficiency and duration of carbon storage following iron addition. Unfortunately, the last three terms and the interactions between them are difficult to quantify.

Downward carbon transport occurs by sinking of particles, deep mixing, and subduction of carbon-rich surface water. Export of organic particles increased in the IronEx II and SOFeX experiments (Bidigare et al. 1999; Buesseler et al. 2003) but remained constant in SOIREE (Table 26.1) (Charette and Buesseler 2000; Nodder and Waite 2001). Downward carbon transport by deep mixing was not observed in any of the iron enrichment experiments. Subduction of the young IronEx I bloom transported some organic carbon downward (Watson et al. 1994), presumably both as POC and DOC.

Remineralization, the breakdown of organic matter, strongly decreases the downward flux of organic carbon. Most organic carbon that leaves the mixed layer is remineralized at shallow and intermediate depths, from which the $CO_2$, released by remineralization, may be ventilated to the atmosphere by deep mixing and upwelling. Only a fraction of the organic matter reaches the deep ocean and ocean sediments, where the carbon can be stored away from the atmosphere for centuries or more.

Carbon storage by subduction in IronEx I was expected to be short-lived because the subsurface waters of the equatorial Pacific Ocean are part of a confined circulation system (Broecker and Peng 1982; Reverdin 1995) and resurface roughly every 15 years (for a thermocline depth of 250 meters [m] and an upwelling rate of 16–40 m $y^{-1}$). Thus, any organic carbon stored by subduction would be rapidly remineralized and released as $CO_2$ back into the atmosphere. Continuous iron fertilization of the equatorial Pacific Ocean between 18°N and 18°S would therefore provide little carbon storage (Sarmiento and Orr 1991).

By contrast, some Southern Ocean waters leave the surface for decades to centuries by deep mixing and subduction. Near the Subantarctic Front, intermediate water sinks below more saline water. Any extra carbon in the "new" intermediate water would be stored away from the atmosphere for long periods. The production of slowly degradable DOC in algal blooms at or south of the Subantarctic Front could favor long-term carbon storage. Sarmiento and Orr (1991) modeled continuous iron fertilization of surface waters south of 31°S. Iron addition for a 100-year period would reduce the atmospheric $CO_2$ increase by 17 percent in an IPCC business-as-usual scenario (This scenario is discussed in Chapter 4 of this volume). Breakdown of organic matter would rapidly lower the initial carbon storage (12 PgC $yr^{-1}$) to 2 PgC $yr^{-1}$ after 20 years and 1 PgC $yr^{-1}$ after 100 years of continuous iron fertilization.It is important to note that much of the stored carbon would be rapidly returned to the atmosphere as soon as the iron addition was stopped, highlighting the limited duration of carbon storage. This study probably overestimates the potential for carbon storage by its assumption of complete nutrient depletion of the fertilized waters, which has not been observed in the iron fertilization experiments. Other modeling studies vary in the amount of carbon stored but present a similar overall pattern (Broecker and Peng 1991; Joos et al. 1991a, 1991b). These calculations probably represent an upper limit for the potential of $CO_2$ sequestration by iron fertilization.

Local, rather than basinwide, iron fertilization could sequester small amounts of $CO_2$. The controls on and consequences of basinwide iron fertilization equally apply to small-scale programs, albeit at a more limited scale. Site selection and monitoring of the amount of carbon stored over a range of timescales will be major challenges to iron fertilization schemes.

## Drawbacks of Iron Fertilization

In the iron enrichment experiments, the biomass of bacteria and small algae remained fairly constant (Coale et al. 1996; Hall and Safi 2001; Gervais et al. 2002), probably as a consequence of increases in grazing by small zooplankton (Coale et al. 1996). The biomass of larger phytoplankton, notably diatoms, increased. By promoting a shift toward larger algal species, iron addition thoroughly changes the structure of the marine food web. It is conceivable that continuous iron fertilization would decrease biodiversity by consequently favoring a small number of species. Long-term, widespread iron fertilization could potentially allow the spread of harmful algal blooms, as has been observed in coastal waters, which are "fertilized" by high nutrient inputs from rivers (eutrophication).

Iron fertilization and iron-induced algal growth reduce nutrient availability to downstream ecosystems (Sarmiento and Orr 1991), which decreases the downstream productivity and changes the structure of the marine food web. One may speculate that the biomass of large algae will be affected more than that of small algae in downstream waters.

The increases in algal growth, carbon export, and remineralization upon iron addition reduce oxygen levels in subsurface waters. Long-term or large-scale fertilization could create conditions with zero oxygen concentrations ("anoxic conditions") at intermediate depths (Fuhrman and Capone 1991; Sarmiento and Orr 1991). In the modern ocean the greenhouse gases nitrous oxide ($N_2O$) and methane ($CH_4$), which are 275 and 62 times more potent than $CO_2$ over a 20-year time scale (Houghton et al. 2001), are produced in anoxic conditions. For example, nitrous oxide is produced below seasonal algal blooms in the northwestern Indian Ocean (Law and Owens 1990). As some nitrous oxide escapes to the atmosphere, these waters are important natural sources for this potent greenhouse gas. By analogy, low oxygen levels and anoxic conditions upon iron fertilization are likely to promote production of nitrous oxide and methane. In SOIREE, nitrous oxide concentrations increased at the bottom of the mixed layer (Figure 26.2h), but this did not result in extra release of $N_2O$ to the atmosphere (Law and Ling 2001). Methane concentrations did not change in SOIREE (C. S. Law, pers. comm.) nor have changes been reported for other experiments. The negative effects of $N_2O$ and $CH_4$ release to the atmosphere would counteract and possibly outweigh the benefits of the atmospheric $CO_2$ sequestration from iron addition (Jin and Gruber 2002; Jin et al. 2002). Low oxygen levels and anoxic conditions following iron fertilization would also negatively affect marine biota that depend on oxygen, such as fish.

Iron fertilization may change the marine production of important gases, such as dimethylsulphide (DMS), halocarbons, and alkyl nitrates. Dimethylsulphide is a major source of atmospheric sulphate and is thought to indirectly affect aerosol formation, albedo, and climate (Charlson et al. 1987). Volatile halocarbons and alkyl nitrates strongly influence atmospheric chemistry. Marine biological processes promote the production of DMS (Turner et al. 1996) and certain halocarbons (Chuck 2002). They may also play an important role in the production of alkyl nitrates (Chuck 2002; Chuck et al. 2002). The production and release of DMS to the atmosphere increased in the IronEx II and SOIREE iron enrichment experiments (Figure 26.2g; Turner et al. 1996; Turner et al. 2003). The concentration of some halocarbons and an alkyl nitrate changed in the EisenEx experiment (Chuck 2002). Changes in the marine source of these gases following iron fertilization could open a Pandora's box with feedbacks on atmospheric chemistry and global climate (Fuhrman and Capone 1991; Lawrence 2002). Iron fertilization has a wide range of observed and potential side effects. These require further study before any large-scale scheme is undertaken.

## Iron Fertilizer and Costs

Ratios of algal carbon uptake to added iron were $0.1 \times 10^4 : 1$ to $3.0 \times 10^4 : 1$ in the IronEx II and SOIREE experiments (Table 26.1). A ratio of $0.7 \times 10^4 : 1$ was calculated between the POC export at 100 m depth and the added iron in SOFeX (Buesseler and Boyd 2003; Buesseler et al. 2003). These ratios are one to two orders of magnitude lower than the carbon-to-iron ratio of $3 \times 10^5$:1 in severely iron-stressed diatoms (Sunda and Huntsman 1995). Iron losses (Bowie et al. 2001) and a higher carbon-to-iron content in iron-replete than in iron-stressed algae (Sunda and Huntsman 1995) may explain the difference in the ratios. Continuous iron fertilization of waters south of 31°S would require $6 \times 10^6$ tons Fe $y^{-1}$ (1 ton = $10^6$ g) for the maximum possible algal growth at a carbon-to-iron ratio of $1 \times 10^4$:1 (after Sarmiento and Orr 1991), which is a 10-fold lower ratio than Sarmiento and Orr used in their calculations.

This iron demand corresponds to 1 percent of the global crude steel output of $700 \times 10^6$ tons Fe $y^{-1}$ (Nagasawa 1995). Purposeful iron mining to meet such a large demand would strongly raise the economic and energy cost of iron fertilization. Iron for small-scale additions might be acquired at relatively low cost as a by-product from industry. Any iron fertilizer used for ocean fertilization should be of good quality, with only traces of other constituents. Use of high-quality ferrous scrap could compete with iron recycling for other purposes (Nagasawa 1995).

The cost of $CO_2$ storage by iron fertilization has been estimated as US\$5–US\$100 $ton^{-1}$ of carbon stored (Ritschard 1992). It is unclear how Ritschard accounted for the efficiency and duration of carbon storage and whether these estimates include the cost of iron mineral. The values do not include the cost of monitoring the efficiency, duration, and side effects of iron fertilization. The $CO_2$ production by acquisition of iron fertilizer and by iron fertilization itself should also be taken into account in calculating the net amount of carbon stored.

## Carbon Storage by Adding Nitrogen and Phosphorus Fertilizer

Enhancement of algal growth by fertilization with nitrogen and phosphorus has also been suggested as an option for carbon storage (Ritschard 1992). In a modeling study, nitrogen and phosphorus were added to the equatorial oceans between 18°N and 18°S (Orr and Sarmiento 1992). Continuous nutrient addition for 100 years would increase oceanic $CO_2$ uptake by 0.7 PgC $yr^{-1}$ on average, if the algal remains were removed and stored away from the atmosphere. This scenario would require a nutrient input higher than the current global nitrogen consumption for fertilizer and close to that for phosphorus. This fertilizer demand would be very costly, both economically and in energy terms. If the algae were not harvested, continuous, widespread fertilization could store 0.4 PgC $yr^{-1}$. Degradation of algal material, however, would result in large-scale anoxic conditions in subsurface waters.

Stimulation of macro-algal growth by the addition of macronutrients seems to be an inefficient tactic for carbon storage (Orr and Sarmiento 1992). Any such scheme would drastically change the local marine ecosystem in ways that might resemble the effects of high nutrient inputs in coastal seas (eutrophication), including damage to coral reefs, toxic algal blooms, and fish kills.

## Legal Issues

The 1972 London Convention bans "dumping," which it defines as " any deliberate disposal at sea of wastes or other matter from vessels, aircraft, platforms, or other man-made structures at sea" (Article 3.1.a (i) in IMO 1972). A 1991 amendment explicitly prohibits dumping of industrial waste. The 1996 Protocol to the Convention, which awaits ratification, removes some of the exemptions with respect to dumping platforms in the 1972 Convention (IMO 1972, 1996). The London Convention, however, does allow the placement of matter for a purpose other than its mere disposal, provided that such placement does not cause pollution or harm marine life. Because fertilizer, originating from recycling or industrial processes, and even anthropogenic $CO_2$ could be regarded as industrial waste (Johnston et al. 1999), it is uncertain whether these provisions would allow $CO_2$ storage by ocean fertilization.

The Protocol on Environmental Protection to the Antarctic Treaty (Anonymous 1991) states that activities in the treaty area shall limit changes of and adverse impacts on the Antarctic environment and associated ecosystems. This provision appears to rule out widespread fertilization of waters south of 60°S, once the protocol has been ratified.

Thus, it is unclear whether fertilization of international waters for the purpose of $CO_2$ storage would be allowed under international law. Of course, laws are designed to promote public policy goals. If iron fertilization were considered an appropriate public policy, then international agreements could be modified.

## Conclusion

Major uncertainties are attached to the efficiency and duration of $CO_2$ storage from ocean fertilization. Modeling studies indicate largely short-lived carbon storage upon basinwide, continuous iron fertilization of the Southern Ocean, the oceanic region with the highest potential for carbon storage from iron addition. It is unknown how climate change, changes in oceanic circulation, and increases in the atmospheric $CO_2$ content will affect future oceanic carbon storage.

The addition of nitrogen and phosphorus fertilizer can be discarded as a serious option for carbon storage based on the small amount of carbon stored and the large supply of fertilizer required. The advantages of carbon storage by iron addition (mostly temporary carbon storage with some durable carbon storage) may not justify the negative aspects of the technique, which would include a reduction of subsurface oxygen concentrations, other major changes of marine chemistry, a transformation of the marine ecosystem, possible losses in marine biodiversity, and substantial financial and energy costs. The production of dimethylsulphide, halocarbons, and alkyl nitrates could adversely affect atmospheric chemistry and global climate. Increased emission of the greenhouse gases nitrous oxide and methane would counteract, or even outweigh, the benefits of atmospheric $CO_2$ reduction by iron addition. The potential negative side effects of iron fertilization require serious investigation before a balanced judgment can be made on the applicability of the technique as a strategy for sequestering $CO_2$ and reducing climate change.

Oceanic fertilization does not provide a full solution to the impacts of the fossil-fuel-driven economy on global climate. At best, it might delay the atmospheric $CO_2$ increase and buy time. The international community should carefully weigh the expected benefits and adverse effects of temporary $CO_2$ storage, reduction of $CO_2$ emissions, and climate change and act accordingly.

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