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Research Document 2010/106

Document de recherche 2010/106

**Information in support of Science
Advisory Report (SAR) 2010/012:
“Ocean Fertilization: Mitigating
Environmental Impacts of Future
Scientific Research”**

**Information à l'appui de l'avis
scientifique (AS) 2010/012 :
« Fertilisation des océans : atténuation
des impacts environnementaux de la
recherche scientifique »**

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This document is available on the Internet at:

<http://www.dfo-mpo.gc.ca/csas/>

Ce document est disponible sur l'Internet à:

ISSN 1499-3848 (Printed / Imprimé)

ISSN 1919-5044 (Online / En ligne)

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Correct citation for this publication:

Christian, J., Peña, A., Denman, K., Gilbert, D. and Lyon, P. 2010. Information in support of Science Advisory Report (SAR) 2010/012: “Ocean Fertilization: Mitigating Environmental Impacts of Future Scientific Research”. DFO Can. Sci. Advis. Sec. Res. Doc. 2010/106. iv + 17 p.

ABSTRACT

Various geoengineering schemes are being proposed to decrease the rate of global warming associated with the buildup of greenhouse gases from human activities. Ocean fertilization attempts to sequester more carbon dioxide from the atmosphere in the ocean interior by adding nutrients to the ocean to stimulate growth of phytoplankton. Phytoplankton convert carbon dioxide to organic matter, which can settle into the subsurface ocean to remain sequestered for decades or centuries. The London Convention/London Protocol (LC/LP) is the global body with authority to regulate ocean fertilization. In 2008, the Scientific and Legal Working Groups of the LC/LP recommended proceeding toward regulation of the activity, and proposed that Parties “agree to the concept of regulation such that commercially driven activities are prohibited”. This document reviews the state of the science of ocean fertilization and its impacts in support of Canadian Science Advisory Secretariat [Science Advisory Report 2010/012](#). The Report addresses four questions: (1) What are the most significant deleterious intended and unintended consequences of ocean fertilization and what is the level of scientific confidence regarding their impacts? (2) Is there sufficient knowledge to determine at what scale a project would likely not cause irreversible and unacceptable harm to an ecosystem? If so, what are the criteria that would define the upper limit of such a project? (3) Is the Convention’s Draft Assessment Framework adequate for assessing scientific research proposals involving ocean fertilization? (4) What are the most pressing or most important research areas on ocean fertilization? The report considers both the scientific basis for regulation of ocean fertilization and the impact of regulation on scientific research. This supporting Research Document presents a more extensive and technical review of the supporting scientific literature.

RÉSUMÉ

Des options de géo-ingénierie ont été proposées pour réduire le taux de réchauffement global résultant de l'accumulation des gaz à effet de serre produits par les activités humaines. La fertilisation des océans a été proposée comme méthode de géo-ingénierie qui permettrait d'augmenter, dans certaines régions de l'océan mondial, l'absorption par les océans du dioxyde de carbone de l'atmosphère. Le phytoplancton convertit le dioxyde de carbone en matière organique, qui peut couler dans l'océan profond et rester là pour des décennies ou siècles. La Convention de Londres / Protocole de Londres (CL/PL) est l'organisme international avec l'autorité appropriée pour réglementer la fertilisation des océans. En 2008, les groupes de travail scientifique et légal de la CL/PL ont recommandé de cheminer vers une réglementation de cette activité, et ont proposé que les Partis "s'entendent sur le concept d'une réglementation telle que des activités commerciales seraient interdites. Ce document passe en revue l'état de la science de la fertilisation des océans et ses impacts en appui à l'[Avis scientifique 2010/012](#) du Secrétariat canadien de consultation scientifique. L'avis répond à quatre questions: (1) Quelles sont les conséquences négatives les plus importantes, prévues et imprévues, de la fertilisation des océans, et quel est le degré de certitude scientifique vis-à-vis des impacts d'une telle pratique? (2) Dispose-t-on de suffisamment de connaissances pour déterminer l'échelle à laquelle un projet ne sera pas susceptible de causer des dommages irréversibles et inacceptables à l'écosystème? Le cas échéant, quels sont les critères qui pourraient nous permettre de définir les limites supérieures associées à un tel projet? (3) Est-ce que le cadre d'évaluation provisoire de la CL/PL convient pour évaluer les propositions de recherche scientifique sur la fertilisation des océans? (4) Quels domaines de la recherche sur la fertilisation des océans affichent le caractère le plus urgent ou important? L'avis considère la base scientifique pour la réglementation de la fertilisation des océans ainsi que l'impact de cette réglementation sur la recherche scientifique. Ce Document de Recherche en appui à l'avis présente un examen plus approfondi et plus technique basé sur la littérature scientifique.

CONTEXT

We are now releasing to the atmosphere over 8 Pg (8 billion metric tons) of carbon from fossil fuel emissions annually. Of that, nearly one third is taken up by the oceans. The oceans contain roughly 50 times more carbon (mostly as dissolved inorganic carbon) than the atmosphere. By taking up roughly one third of the carbon from burning of fossil fuels, the oceans are reducing both the rate of increase of carbon dioxide in the atmosphere and the associated rate of global warming associated with the enhanced greenhouse effect.

Various geoengineering schemes are being proposed to decrease the rate of global warming associated with the build up of greenhouse gases from human activities (Royal Society, 2009). One such technique, ocean fertilization, attempts to sequester more carbon dioxide from the atmosphere in the ocean interior by adding a limiting nutrient to particular regions of the ocean to stimulate the growth of phytoplankton, converting carbon dioxide to organic matter through photosynthesis. The organic carbon settles into the subsurface ocean where carbon may remain sequestered for decades or centuries.

Obtaining carbon credits or “offsets” for enhancing natural carbon sinks is potentially a means by which a nation or organization can offset its own carbon dioxide emissions in order to comply with assigned emissions quotas. Understandably, ocean fertilization is envisioned as a means to generate carbon credits. While research has increased our level of understanding of advantages and disadvantages of this practice, the degree of uncertainty associated with the current state of knowledge of the effects of ocean fertilization has driven international organizations to take steps to ensure that appropriate, precautionary measures are put in place.

Internationally, the London Convention/London Protocol (LC/LP) is regarded as the global body with authority to manage ocean fertilization. The LC/LP are international treaties designed to govern marine pollution and regulate dumping of wastes at sea. Canada is a leader in these forums, providing expertise to both the legal/policy and scientific bodies. In May 2008, the Scientific and Legal Working Groups of the LC/LP were tasked to evaluate the issue of ocean fertilization. Subsequently, they recommended proceeding toward regulation of the activity. The legal group proposed that Parties “agree to the concept of regulation such that commercially driven activities are prohibited”.

At the November 2008 Meeting of Contracting Parties to the LC/LP, Governing Bodies adopted a non-binding resolution indicating that ocean fertilization should be considered as dumping and that, given the current state of knowledge, ocean fertilization activities other than “legitimate scientific research” should not be allowed. Canada supports the LC/LP resolution to not allow ocean fertilization, with the exception of legitimate scientific research. The application of the precautionary approach at this point in time is appropriate, given that ocean fertilization will continue to be a science and policy issue as long as there is potential to generate carbon credits for trading via this practice, that the activity may pose a risk of serious or irreversible harm, and that there is a lack of scientific confidence as to whether it will work.

In February 2009, the Intersessional Technical Working Group of the LC/LP developed a Draft “Assessment Framework for Scientific Research Involving Ocean Fertilization”. The framework provides a tool for assessing scientific research proposals on a case-by-case basis to determine if a proposed activity is consistent with the aims and objectives of the London Convention or Protocol and meets the requirements, as appropriate.

This document examines the current state of scientific knowledge and understanding of ocean fertilization, and in this context, it addresses known issues, uncertainties, and fundamental questions required for informed decision making. As part of this evaluation, this document reviews the Draft Assessment Framework created by the LC/LP Intersessional Technical Working Group. Finally, this document formed the basis for a discussion by experts from the Government of Canada and academia at a workshop held in Ottawa, Ontario on September 29 and 30, 2009. The Canadian Science Advisory Secretariat has released Science Advisory Report 2010/012, and Proceedings documents resulting from the workshop. This supporting Research Document covers the same issues in more detail than can be accommodated within the Advisory Report, and contains more extensive scientific literature references for those who wish to pursue the issue further. The reasoning behind evaluations of impact likelihood and confidence in these conclusions that are summarized in the Advisory Report is explained here in much greater detail.

1. INTRODUCTION

Ocean fertilization has been proposed both as a geoengineering scheme, with the objective of removing CO₂ from the atmosphere and storing it in the subsurface ocean (sequestration), and as a means of enhancing production of fish or other seafood for human harvesting. Neither has actually been demonstrated to date, except for limited sequestration of carbon in mesoscale open ocean iron fertilization experiments.

Fertilization proposals are loosely divided into 'micronutrient' and 'macronutrient' fertilization. Macronutrients are the elements N, P, and Si (required by phytoplankton in molar ratios of about 1 in 10-100 relative to carbon), while micronutrients include Fe, Zn, Mo and about a dozen other principally metallic elements (required in ratios less than 1 to 10000). Fe is the only micronutrient element that has been seriously considered for ocean fertilization; macronutrient fertilization proposals normally involve N, although it is far from clear that this could accomplish the desired goal without adding P as well (Matear and Elliott, 2004). Another possibility involves simultaneous fertilization with P and Fe (hybrid micronutrient/macronutrient, Karl and Letelier, 2008) but this has received little serious attention from fertilization proponents or opponents to date. Another approach that has been suggested is to induce upwelling of natural subsurface nutrient pools, i.e. not to add nutrients to the ocean but by human intervention to increase the flux of nutrients into the surface layer from the subsurface ocean (Karl and Letelier, 2008; Dutreuil et al., 2009).

The ocean has a mosaic of nutrient limitations, with some regions being Fe limited, while others are principally limited by N, P, or Si (Moore et al., 2002). In principle, only a single element is limiting for phytoplankton production at any given time; this is a venerable concept in oceanography and phycology (e.g., Droop, 1983). However, it is not strictly true (e.g., Bruland et al., 1991; Armstrong, 1999; Flynn, 2001), and regions may undergo limitation by different elements at different times, or possibly a mosaic of different limitations at the mesoscale. In any case, all of the four elements cited above play a role in creating the global mosaic. For example, while the proximate limiting nutrient in one region may be silica, iron limitation elsewhere may set up the conditions for Si limitation by determining the N/Si ratio in thermocline waters (Takeda, 1998; Sarmiento et al., 2004).

Continental shelf waters are normally N-limited, due to denitrification in shelf sediments (Fennel et al., 2006) and extensive terrestrial inputs of Fe and Si (Whitney et al., 2005), although they may become Fe-limited under strong upwelling conditions (Hutchins and Bruland, 1998). Open

ocean waters may be limited by any of the four elements cited, or possibly others such as Zn (Brand et al., 1983), or even certain organic compounds such as cyanocobalamin (vitamin B₁₂) (Fiala and Oriol, 1984), although no compelling evidence for limitation in situ by these other elements or compounds exists. Iron limitation is considered to be a critical factor in maintaining high-nutrient, low-chlorophyll (HNLC) regions (up to 30% of the world ocean) where surface macronutrient pools are never entirely depleted, although this should not be taken to mean that complete drawdown would necessarily occur in the absence of Fe limitation (Mitchell et al., 1991; Zahariev et al., 2008). However, at least some drawdown would be expected, with associated uptake of atmospheric CO₂. The idea that this drawdown could be catalyzed by a fairly small amount of iron given the large (approximately 100000 to 1) stoichiometric ratio of C to Fe in oceanic phytoplankton is the basis for the hypothesis that iron-stimulated phytoplankton production was a major factor in glacial-interglacial change in atmospheric CO₂ (Martin, 1990), and subsequently of geoengineering proposals. However, mesoscale iron fertilization experiments suggest that actual net sequestration of carbon per unit of iron added is much lower (de Baar et al., 2005; 2008).

2. HISTORICAL AND SCIENTIFIC BACKGROUND

2.1 THE GLOBAL OCEAN CARBON CYCLE AND ATMOSPHERIC CO₂

The ocean is by far the largest 'exchangeable' reservoir of carbon on Earth, i.e., exchangeable with the atmosphere on time scales of thousands of years or less. The ocean is therefore considered to be the reservoir that expands and contracts over the glacial-interglacial cycle, which generates an oscillation in atmospheric CO₂ of approximately 100 ppm (e.g., Raynaud et al., 1993). The ocean has also absorbed approximately half of the cumulative anthropogenic emissions of CO₂ to date (total uptake ~118 Pg C; Denman et al., 2007).

Ocean carbon chemistry is complex and somewhat counterintuitive. CO₂ combines with water to form carbonic acid, H₂CO₃, which then dissociates to form bicarbonate (HCO₃⁻) and carbonate (CO₃²⁻) ions. Approximately 90% of the total CO₂ (TCO₂ = CO₂+HCO₃⁻+CO₃²⁻, also known as dissolved inorganic carbon (DIC)) in seawater is present as bicarbonate, 10% as carbonate and 1% as CO₂ gas. Only the fraction of DIC present as dissolved CO₂ contributes to the ocean surface pCO₂ (i.e. partial pressure of CO₂), which opposes the invasion of CO₂ from the atmosphere into the ocean. For this reason, uptake of CO₂ does not saturate as with other gases such as oxygen and nitrogen, and the time scale for equilibration is much longer (~1 year vs 1 month for oxygen). This chemistry is in large part responsible for Earth's present climate, because most of the exchangeable carbon resides in the ocean.

The near surface concentration of CO₂ is controlled in large part by two sets of processes that are known as the 'solubility pump' and the 'biological pump'. These together maintain a strong gradient of DIC (TCO₂) between the surface ocean and the deep ocean. The solubility pump results from the greater solubility of CO₂ gas in cold water, and the fact that the deep ocean is largely ventilated in the high latitudes (deep water in the tropics and subtropics sinks in the high latitudes and flows horizontally at depth, and there is very little exchange between surface and deep waters). The biological pump results from net production of particulate and dissolved organic carbon (POC, DOC), and particulate inorganic carbon (carbonate minerals) by plankton and fish in the surface layer, and its remineralization and dissolution in the intermediate and deep ocean. The biological pump includes sedimentation of both organic and inorganic particulate carbon, as well as downward mixing and advection of DOC. Globally, the biological

pump accounts for about two thirds of the cross-thermocline gradient of DIC and the solubility pump for the rest (Murnane et al., 1999).

At steady state the magnitude of downward transport is equivalent to upward transport of DIC by mixing and upwelling. Departures from this steady state, as over a glacial-interglacial cycle, result in fluctuations of atmospheric CO₂. Nutrients such as N and P follow a similar cycle: upward mixing and upwelling of dissolved inorganic N and P is approximately equivalent to downward transport of the organic forms. Ocean fertilization for geoengineering purposes is intended to enhance the biological pump, resulting in net ocean uptake of CO₂.

2.1.1 Export of carbon, remineralization, and sequestration time

Deep ocean renewal time is about 500 years on average, but some subsurface waters are much older (Stuiver et al., 1983). The oldest waters are those of the North Pacific oxygen minimum zone (~1700 years, Stuiver et al., 1983). Thermocline and intermediate waters are renewed on a much shorter time scale of a few decades or less (e.g., Sonnerup et al., 1999). In the North Pacific Ocean the ventilated thermocline reaches depths of around 500 m and densities of 1026.8 kg m⁻³. This is the upper limit to surface density in the North Pacific, so it is assumed that isopycnal layers deeper than this are not ventilated (since their outcrop at the surface would be observed). It is necessary for carbon to be remineralized at depths greater than this density surface to remain sequestered for a significant period. The minimum depth for long-term sequestration is the maximum depth of the winter mixed layer, but in some regions it is substantially deeper.

Remineralization of organic carbon declines with depth, and the particle flux attenuates approximately exponentially (Martin et al., 1987; Christian et al., 1997): remineralization is both continuous and biased towards shallower depth strata. The sequestration timescale is therefore likely to be highly variable even among carbon atoms sinking out of the euphotic zone in the same time and place, and fairly short on average. Furthermore, because both the remineralization length scale and the circulation of the subsurface layers in which remineralization occurs are variable and poorly known, any estimate of the sequestration timescale will be highly uncertain. Phytoplankton blooms can generate rapidly sinking pulses of organic matter that are transported to the deep ocean with little remineralization (e.g., Lampitt, 1985; Scharek et al., 1999), and if such blooms could be assumed to arise reliably from fertilization, the sequestration timescale would be several hundred years. However, in open ocean fertilization experiments to date this has not occurred, and even detecting enhanced export across the bottom of the euphotic zone has proven difficult (de Baar et al., 2005; Boyd et al., 2007).

2.2 MICRONUTRIENT FERTILIZATION

Iron fertilization has been the most intensively studied form of ocean fertilization to date. The basis for iron fertilization as a form of geoengineering originated largely in the work of John Martin (1935-1993) and his collaborators in the 1970s and 1980s. Martin is widely known as the father of the “iron hypothesis” regarding the formation of HNLC conditions, but it is less well known that Martin and his collaborators played a pivotal role in quantifying elemental ratios in plankton for a much larger suite of elements than had been considered in the earlier work of Redfield and others (Martin and Knauer, 1973). Martin observed that iron concentrations in plankton and particulate matter were very low (10⁻⁴-10⁻⁵ mol/mol) relative to carbon, which eventually gave rise to the idea that substantial ocean uptake of CO₂ could be catalyzed by fairly small additions of iron. Cellular iron quotas in oceanic phytoplankton are much lower than

in coastal species (e.g., Sunda and Huntsman, 1995). This difference in iron requirements between coastal and oceanic phytoplankton is a resilient feature of oceanic ecosystems (Brand et al., 1983; Sunda and Huntsman, 1995; Strzepek and Harrison, 2004), which is believed to result from an evolutionary trade off between the ability to grow in iron-impoverished conditions and the ability to acclimate rapidly to changing irradiance (Strzepek and Harrison, 2004).

Martin (1990) also made the connection between glacial-interglacial changes in atmospheric CO₂ and deposition of aeolian iron at the ocean surface, by examining the record of aeolian dust in Antarctic ice cores. The flux of terrestrial mineral dust to Antarctica and therefore presumably to the iron-poor waters of the Southern Ocean was much higher in the glacial climate, so it was reasonable to hypothesize that iron enhanced ocean uptake of CO₂ in the glacial climate. However it is important to note that subsequent investigations of the palaeoceanography of the Southern Ocean do not entirely support this hypothesis. In particular, nitrogen isotopic ratios in marine sediments do not indicate that HNLC conditions disappeared (Francois et al., 1992), although in some regions there is evidence of a significant increase in organic sedimentation (Kumar et al., 1995). Although iron fertilization is among the best hypotheses available to explain what *initiates* the sequence of events that lead to glacial CO₂ drawdown (Broecker, 2000), it is now believed to account for a modest amount of CO₂ uptake (~50 PgC, Watson and Naveira Garabato, 2006).

In the 1980s Martin initiated a series of cruises to HNLC regions to test the hypothesis that iron was the principal limiting nutrient for phytoplankton production (Martin et al., 1991). The primary approach employed was bottle experiments (adding iron to seawater in bottles and measuring the growth of phytoplankton relative to unamended controls). The results were always controversial, in part because the relatively small volume of water sampled invariably excludes some zooplankton and therefore alleviates grazing pressure on the phytoplankton, and in part because enhanced phytoplankton growth frequently occurred in the unamended controls, as it is extremely difficult to conduct these experiments aboard ship without contaminating the bottles with iron (Dugdale and Wilkerson, 1990; Banse, 1991; Cullen, 1991). Eventually it was concluded that further bottle experiments would do nothing to alleviate the controversy, and that it would be necessary to conduct an open-water fertilization experiment. The first such experiment was conducted in the eastern equatorial Pacific in 1993 with funding primarily from the US National Science Foundation (Martin et al., 1994). This same year Martin passed away; he did not live to see the results of the experiment but would have felt vindicated. The experiment silenced the naysayers: it was now clear that iron was the primary limiting nutrient in at least some HNLC waters. But in other ways the experiment was a failure: no net sequestration of carbon was observed, and the iron precipitated rapidly. In subsequent experiments both the timing and the chemical form of the iron addition were altered to maximize the fraction remaining in solution or at least in the upper layer. There have now been about a dozen open water iron fertilization experiments, in all of the major HNLC regions (de Baar et al., 2005; Boyd et al., 2007).

2.2.1 Iron solution chemistry

The aqueous chemistry of iron is complex. The forms thermodynamically favoured under ocean surface conditions are highly particle-reactive, that is, they will bind to almost any surface they come in contact with, so that iron is constantly being removed from solution and transported downwards by sinking particles (scavenging). As a result the whole-ocean residence time for iron is estimated as only around 100 years, compared to 3000 years for N and 30000 for P. The residence time of added iron in the surface ocean (where both oxygen and particulate matter

concentrations are high) is much shorter, and in deliberate fertilization experiments a large fraction of the added iron precipitates from solution on a time scale of hours to days.

2.2.2 Past iron fertilization experiments

The first in situ fertilization experiment in the equatorial Pacific was deemed to be a success because it unequivocally demonstrated that iron limited phytoplankton growth and macronutrient uptake (Martin et al., 1994). Yet it was not successful from a geoengineering perspective because it did not demonstrate enhanced net export of carbon from the surface ocean or enhanced ocean uptake of atmospheric CO₂. Subsequently there have been about a dozen additional experiments, as well as a few others conducted by commercial organizations that did not report any results in the open literature. Most of these experiments induced some enhanced export of carbon, although again as a prototype demonstration of geoengineering the results are not particularly encouraging because the sequestration of carbon per unit of iron added is quite low (de Baar et al., 2005; 2008). Measured as net uptake of CO₂ from the atmosphere it is substantially lower (de Baar et al., 2008).

There have also been "natural" fertilization experiments, i.e. focused field campaigns around groups of islands known to be significant sources of iron to adjacent open ocean waters, and "virtual" experiments with a variety of types of ocean models. The net result of all of these is to cast significant doubt on the viability of iron fertilization as a geoengineering strategy. Some of the natural fertilization experiments appeared to suggest that net sequestration of carbon per unit of iron input was much larger than in the deliberate addition experiments (Blain et al., 2007). However, in these experiments the magnitude of the iron source is not known and must be inferred from field data, so the ratio of carbon sequestered to iron added is a ratio of two not very well constrained quantities. Subsequent investigations have cast considerable doubt on these high sequestration ratio estimates (Pollard et al., 2009). Model experiments show clearly that successful geoengineering depends strongly on this ratio: it must be several orders of magnitude larger than observed in the field experiments for geoengineering to be viable (Zeebe and Archer, 2005). Even if the most optimistic ratios turned out to be correct, fertilization would still make a very modest contribution to slowing atmospheric CO₂ growth (Sarmiento and Orr, 1991; Aumont and Bopp, 2006; Zahariev et al., 2008). Model studies have also shown that surface waters depleted of nutrients and subducted will eventually be upwelled again and the downstream effects on primary production, ecosystems and fisheries are potentially large (Gnanadesikan and Marinov, 2008; Zahariev et al., 2008).

2.3 MACRONUTRIENT FERTILIZATION

Macronutrient fertilization has a somewhat less distinguished pedigree than micronutrient fertilization. In contrast to the hundreds of scientific papers written on iron fertilization (largely due to the extensive funding of mesoscale iron fertilization experiments by national science agencies), very few papers have been written on macronutrient fertilization. In fact, a landmark evaluation of the global potential using an ocean model (Matear and Elliott, 2004) had been cited only four times as of early 2010 (ISI Web of Science), and none of the citing articles deals directly with further evaluation of this strategy. It can reasonably be said that experimental verification of the viability of macronutrient fertilization is in its infancy.

An important difference between micronutrient and macronutrient fertilization is that micronutrient fertilization seeks to reduce atmospheric CO₂ by redistributing macronutrients downward in the ocean, while macronutrient fertilization seeks to increase the total ocean nutrient pool. Partitioning of carbon between the ocean and atmosphere is, at least in the

equilibrium state, closely related to the whole-ocean inventory of N and P (although there is some variation superimposed on this by the variable reserve of unused nutrients in the surface ocean). Macronutrient fertilization is potentially more permanent than micronutrient fertilization, i.e., continuous fertilization into the indefinite future would not necessarily be required to prevent sequestered carbon from being lost. It can also reasonably be assumed that the 'downstream' ecological effects of micronutrient and macronutrient fertilization will be quite different, although they are highly uncertain in each case. Macronutrient fertilization will tend to increase (macro)nutrient concentrations in newly upwelled thermocline water in the decades following fertilization, while iron fertilization can either increase or decrease them depending on the depth of remineralization.

3. QUESTIONS FOR ASSESSMENT

In the following discussion we assess the four questions that were central to the CSAS assessment. We estimate both the likelihood of particular impacts occurring and the level of scientific confidence in those assessments. Ocean fertilization is defined here as deliberate addition of inorganic nutrients such as iron, nitrogen, or phosphorus to the ocean with the objective of stimulating phytoplankton production and subsequent ecosystem-level changes including, but not limited to, enhanced uptake of CO₂ from the atmosphere. We refer to "successful" fertilization as a means of assessing its possible impacts: *successful fertilization in this context means net sequestration of atmospheric carbon*. None of these comments should be construed as commentary on the likelihood of such successful sequestration occurring.

3.1 WHAT ARE THE MOST SIGNIFICANT DELETERIOUS INTENDED AND UNINTENDED CONSEQUENCES OF OCEAN FERTILIZATION AND WHAT IS THE LEVEL OF SCIENTIFIC CONFIDENCE REGARDING THEIR IMPACTS?

3.1.1 Oxygen depletion

Remineralization of organic matter under oxic conditions consumes oxygen. Thus, reduction of subsurface oxygen concentrations is not an inadvertent consequence of ocean fertilization, but an intended consequence. If carbon is sequestered, oxygen will be reduced and might be depleted locally. The stoichiometry is clear and inflexible.

Oxygen in the thermocline has already been observed to be declining in some regions (e.g., Whitney et al., 2007), and will inevitably decline globally as the ventilation temperature increases.

We rate the probability of subsurface oxygen reduction as a result of successful fertilization **High** and the level of confidence in this conclusion **High**. However, the probability of decrease in subsurface oxygen concentrations leading to deleterious consequences (hypoxic/anoxic conditions) is **Moderate** and the level of confidence in this conclusion is **Low**.

3.1.2 Ocean acidification

There have been claims floated in the lay press and the blogosphere by both proponents and opponents of fertilization that it will generally alleviate or exacerbate ocean acidification, respectively. Neither has any evidentiary basis.

Ocean acidification is occurring on a massive scale and can not be stopped except by a complete halt to CO₂ emissions. It can be slowed by restraining and ultimately reducing emissions. Any additional effect of ocean fertilization is likely to be small compared to even small variations in future emissions.

A shift in community composition toward or away from calcareous phytoplankton does not result in a net gain or loss of alkalinity from the ocean, except for the small net loss associated with seafloor burial of carbonates. Calcification and carbonate dissolution redistribute alkalinity within the water column, so that a change of species composition may simultaneously exacerbate and alleviate ocean acidification in different depth strata.

Increased remineralization will make the naturally acidic waters below eastern boundary currents (EBCs) more so. Upwelling of water bearing an anthropogenic acidification signature onto the continental shelves has begun to be observed and will almost certainly increase (Feely et al., 2008). This would be particularly acute with macronutrient fertilization along the shelf break in EBC regions.

Iron fertilization of HNLC regions should favour diatoms, reducing calcification and therefore alleviating acidification of surface waters in the short term. Southern Ocean and Subarctic Pacific surface waters are among those where aragonite undersaturation is projected to occur this century (Orr et al., 2005). It would also be a slight negative feedback on atmospheric CO₂ growth (Zondervan et al., 2001).

Macronutrient fertilization with N only will almost certainly alter phytoplankton species composition as the ecosystem is driven towards phosphorus limitation. Some calcifying species are favoured by high N/P ratios, so this could result in enhanced calcification, slightly exacerbating acidification of the surface ocean and alleviating it in the deep ocean.

We rate the likelihood of significant exacerbation of ocean acidification as a result of ocean fertilization to be **Moderate** and the level of confidence in scientific understanding of this area of impact to be **Low**.

3.1.3 Change in phytoplankton community structure and food web

Change in phytoplankton community structure is also an *intended* consequence of ocean fertilization. If it does not occur, fertilization will not be successful as a geoengineering strategy.

Glibert et al. (2008) note that both the viability of macronutrient fertilization as a geoengineering strategy, and the assumption that it will not have extreme adverse ecological consequences, depend on very tenuous assumptions about the evolution of community structure in response to fertilization. These assumptions are untested, and are quite likely to be incorrect.

There is no a priori reason to assume that harmful algal blooms (HABs) will occur as a result of ocean fertilization, but the risk that they will is real and the potential for predicting where and when is low. A recent publication showed that in the subarctic Pacific HNLC region, iron addition selected for neurotoxin (domoic acid) producing *Pseudonitzschia* species and increased cellular concentrations of the toxin (Trick et al., 2010).

We rate the likelihood of significant changes in phytoplankton community structure as a result of ocean fertilization to be **High** but the consequences of these changes for the rest of the food

web to be unknown and the level of confidence in scientific understanding of this area of impact to be **Low**.

3.1.4 Production of climate active gases

Nitrous oxide (N₂O)

N₂O is a potent greenhouse gas with a radiative efficiency (the amount of greenhouse warming per unit concentration in the atmosphere) about 200 times greater than CO₂ and a mean atmospheric lifetime of about 100 years (Forster et al., 2007). It is also implicated in stratospheric ozone depletion.

N₂O is produced by two distinct biological processes: nitrification (oxidation of ammonium (NH₄⁺) to nitrate (NO₃⁻)), and denitrification (reduction of NO₃⁻ to N₂). Denitrification occurs only under suboxic and anoxic conditions and so has long been considered as a potential side effect of ocean fertilization (e.g., Fuhrman and Capone, 1991). Nitrification may be more important as a source of *atmospheric* N₂O (Dore et al., 1998; Lueker et al., 2003), even if denitrification is the larger source of N₂O overall, because N₂O can be reduced to N₂ without ever escaping to the atmosphere, and because nitrification mostly occurs at quite shallow depths.

Nitrification occurs in direct proportion to remineralization of organic N (which has the same oxidation state as NH₄⁺), and so is generally greatest immediately below the euphotic zone. Any increase in export production will inevitably increase nitrification and associated N₂O production. This will occur predominantly at shallow depths that are ventilated on time scales of a few decades at most unless the remineralization length scale increases substantially. An increase in export production in the vicinity of EBCs will intensify the suboxic conditions naturally found there and will increase N₂O production by denitrification. It is likely that at least some of this N₂O will escape to the atmosphere but the proportion is not well constrained.

Macronutrient fertilization implies injection of reduced N (ammonium or urea), which could stimulate a significant increase in nitrification and associated N₂O production, whether or not the excess N is actually utilized by phytoplankton.

We rate the likelihood of significant increase in ocean efflux of N₂O locally as a result of ocean fertilization to be **High** and the level of confidence in this conclusion to be **Moderate**. We rate the likelihood of additional production remote in space and time ('downstream effects') to be **Moderate** and the level of confidence in predicting these to be **Low**.

Methane (CH₄)

CH₄ has a radiative efficiency approximately 25 times that of CO₂ and an atmospheric lifetime of about 12 years (Forster et al., 2007). As well as radiative forcing, oxidation of methane affects climate by injecting water vapour into the stratosphere. Atmospheric oxidation of methane also produces CO₂, so the global warming potential is equal to that of CO₂ *plus* the additional radiative efficiency prior to oxidation plus the water vapour effect.

The ocean plays a small role in the global source of atmospheric methane. Methanogenesis occurs almost exclusively in the sediments, whereas denitrification occurs in the water column because nitrate is the most thermodynamically favourable electron acceptor after oxygen. Methanogenesis is the least efficient form of heterotrophic metabolism and so only occurs after all other electron acceptors (e.g., oxygen, nitrate, sulfate) are consumed. Methanogenesis in

marine sediments is much less per unit of organic matter consumed than in freshwater sediments because of the large concentration of sulfate in seawater.

An enhanced ocean methane source is among the least likely consequences (at least, unlikely to be of sufficient magnitude to be of interest) of ocean fertilization.

We rate the likelihood of significant increase in ocean efflux of CH₄ as a result of ocean fertilization to be **Low** and the level of confidence in this conclusion to be **Moderate to High**.

Dimethylsulfide (DMS)

Dimethylsulfide is a volatile sulfur compound produced by oceanic microbial communities and is the largest natural source of atmospheric sulfate aerosol. It is produced by enzymatic cleavage of dimethylsulfopropionate (DMSP), a nonvolatile compound produced in large quantities by marine phytoplankton. Many different types of phytoplankton produce DMSP, although intracellular concentrations vary greatly among species. The exact function or functions of DMSP in phytoplankton are still not known.

Enhanced production of DMS has been observed in several iron fertilization experiments. The Canadian SERIES experiment had by far the highest concentrations of DMS but these were also present outside the fertilized patch; the in patch concentration was little different or even at times lower (Levasseur et al., 2006). In one Southern Ocean fertilization experiment, the in patch DMS concentration exceeded the out of patch concentration by almost a factor of five (Wingenter et al., 2004).

It has recently been suggested that the climatic effect of enhanced DMS production due to ocean fertilization could be as large or larger than the effect of CO₂ drawdown, and that this could allow fertilization to succeed as a geoengineering strategy even if fails on CO₂ alone (Wingenter et al., 2007).

We rate the likelihood of significant increase in ocean efflux of DMS as a result of ocean fertilization to be **Moderate** and the level of confidence in this conclusion to be **Low**. Scientific understanding of the climatic effects of increased DMS flux is also **Low**.

Methyl halides (halocarbons)

Methyl halides such as methyl bromide (CH₃Br), methyl chloride (CH₃Cl), and methyl iodide (CH₃I) are also produced by marine phytoplankton although their exact biological function is not known (e.g., Scarratt and Moore, 1998). They are both greenhouse gases and catalysts for stratospheric ozone loss. Their atmospheric lifetime is short (~ 1 year). Methyl bromide has a radiative efficiency about 7000 times larger than CO₂, so despite its short atmospheric lifetime it has a global warming potential larger than CO₂ even over a 100 year time horizon (Forster et al., 2007).

Enhanced production of methyl bromide was observed in an iron fertilization experiment in the Southern Ocean, whereas methyl iodide concentration went down, likely due to increased microbial oxidation in fertilized seawater (Wingenter et al., 2004).

We rate the likelihood of significant increase in ocean efflux of halides to be **Moderate** and the level of confidence in this conclusion to be **Low**.

Sulfur hexafluoride (SF₆)

Sulfur hexafluoride is an industrially produced compound that has found use as a purposefully added tracer in oceanography because it is nonreactive and measurable at extremely low concentrations. It has been added in mesoscale iron addition experiments to help to track the patch of fertilized water and to distinguish fertilized from unfertilized waters and estimate mixing of the two.

SF₆ is a volatile gas and injection into the near surface ocean will increase the atmospheric inventory. It is an extremely potent greenhouse gas with a very long atmospheric lifetime (Forster et al., 2007).

Further use of SF₆ will likely be required because past fertilization experiments have not produced estimates of sequestration efficiency that are sufficiently consistent and well constrained to be extrapolated with confidence. The climatic impact of SF₆ needs to be counted among the impacts of fertilization.

We rate the likelihood of significant increase in ocean efflux of SF₆ to be **High** and the level of confidence in this conclusion to be **Moderate** (because less destructive alternative technologies could emerge).

3.2 IS THERE SUFFICIENT KNOWLEDGE TO DETERMINE AT WHAT SCALE A PROJECT WOULD LIKELY NOT CAUSE IRREVERSIBLE AND UNACCEPTABLE HARM TO AN ECOSYSTEM? IF SO, WHAT ARE THE CRITERIA THAT WOULD DEFINE THE UPPER LIMIT OF SUCH A PROJECT?

There is probably sufficient knowledge at present to define a scale at which fertilization experiments are unlikely to have such effects. Fertilization experiments to date have been of quite limited scale and have largely occurred in the open ocean. Lasting impacts are negligible. These experiments set a precedent for defining a research or pilot scale (e.g., <1000 square kilometres) below which no significant impacts should be expected. The scale at which ocean fertilization will result in persistent alterations of the ecosystem, however, can only be accurately determined by exceeding it.

Biogeochemical and radiative impacts of the type described in 3.1 above have likely occurred on a small scale as result of past mesoscale fertilizations. Evaluation of such impacts depends on both the *scale* and the *sign* of the effect. For example, if the effect on subsurface oxygen levels is invariably in the direction of oxygen depletion, there is an ethical argument that it is not acceptable even if its scale is many of orders of magnitude below other natural and anthropogenic effects (this argument has been made against sewage disposal in the Strait of Juan de Fuca, for example). If the sign is unknown, or the net effect is unlikely to be nonzero, this argument does not hold.

For ocean acidification, CH₄, DMS and halocarbons, it is not known whether the net effect is positive, negative, or zero. For oxygen depletion and N₂O production there is likely a net effect (reduction in subsurface O₂ and increased efflux of N₂O), but its scale is unimaginably small compared to other human perturbations such as transport of nitrogen fertilizer to the ocean in runoff or change in ventilation temperature due to greenhouse gases.

For broader ecosystem impacts (community structure, food webs) it is impossible to state with confidence that such impacts will not occur, and there have been theoretical studies that

suggest that small perturbations can propagate to larger scales through the intrinsic dynamics of the ecosystem (Neufeld et al., 2002). However, such scenarios are highly speculative. By comparison to the impact of human harvesting of marine organisms, fertilization would have to occur on a very large scale to have an impact comparable to even a tiny fraction of the global fishing fleet.

3.3 IS THE LC/LP DRAFT ASSESSMENT FRAMEWORK ADEQUATE FOR ASSESSING SCIENTIFIC RESEARCH PROPOSAL INVOLVING OCEAN FERTILIZATION?

The Draft Assessment Framework provides a comprehensive listing of potential impacts, and a systematic framework for assessing these.

The Draft Assessment Framework provides a mechanism for verifying that legitimate scientific research is being conducted.

The Draft Assessment Framework provides a framework for risk assessment and risk management. It seeks to "ensure that a precautionary approach is followed" (Section 8.1) but does not embrace extreme interpretations of the precautionary principle that could be used to block virtually any proposed experiment. It seeks to ensure that "environmental risks are minimized and the benefits maximized", i.e., there is an explicit recognition that there are potential benefits and that some level of risk is acceptable.

The Draft Assessment Framework places a significant burden of impact assessment and reporting on scientific institutions that was not previously present. For experiments below a certain scale this may not be consistent with the risk assessment framework, i.e., the burden of assessment and reporting is disproportionate to the risks involved.

3.4 WHAT ARE THE MOST PRESSING OR MOST IMPORTANT RESEARCH AREAS ON OCEAN FERTILIZATION?

The effectiveness of ocean fertilization as a geoengineering strategy remains unproven. Therefore, research into potential impacts can not be entirely separated from the question of effectiveness. Unless its effectiveness is demonstrated, ocean fertilization will not proceed on a scale likely to produce significant impacts.

The impact of new nutrient inputs on phytoplankton community structure, sedimentation, and biogeochemical cycles remains a lively area of scientific research with important repercussions for humanity regardless of whether ocean fertilization as a geoengineering strategy proceeds. For example, the climate models used in the IPCC assessment reports now increasingly include an ocean carbon cycle with (mostly rudimentary) representations of plankton communities and the biological pump (Friedlingstein et al., 2006; Denman et al., 2007). What regulates the fraction of primary production exported from the euphotic zone, the ratio of organic to inorganic carbon in the sinking flux, and the remineralization of organic matter below the euphotic zone are among the most vexing and intractable issues in biological oceanography and have important implications for future atmospheric CO₂. Ocean acidification gives these issues a new urgency as all of these processes will be affected by ocean acidification and will in turn affect the regional manifestations of acidification.

The scientific community has learned a great deal from open ocean fertilization experiments that could not have been learned any other way. Further experimentation with different combinations

of nutrients and in different regions could advance the sciences of oceanography and biogeochemistry significantly, with negligible environmental impact.

Literature Cited

- Armstrong, R. A., An optimization-based model of iron-light-ammonium colimitation of nitrate uptake and phytoplankton growth, *Limnol. Oceanogr.*, 44, 1436-1446, 1999.
- Aumont, O., and L. Bopp, Globalizing results from ocean in situ iron fertilization studies, *Global Biogeochem. Cycles*, 20, 10.1029/2005GB002591, 2006.
- Banse, K., Rates of phytoplankton cell division in the field and in iron enrichment experiments, *Limnol. Oceanogr.*, 36, 1886-1898, 1991.
- Blain, S., and others, Effect of natural iron fertilization on carbon sequestration in the Southern Ocean, *Nature*, 446, 1070-1074, 2007.
- Boyd, P. W., and others, Mesoscale iron enrichment experiments 1993-2005: Synthesis and future directions, *Science* 315, 612-617, 2007.
- Brand, L. E., W. G. Sunda, and R. R. L. Guillard, Limitation of marine phytoplankton reproductive rates by zinc, manganese, and iron, *Limnol. Oceanogr.*, 28, 1182-1198, 1983.
- Broecker, W. S., Abrupt climate change: causal constraints provided by the paleoclimate record, *Earth-Sci. Rev.*, 51, 137-154, 2000.
- Bruland, K. W., J. R. Donat, and D. A. Hutchins, Interactive influences of bioactive trace metals on biological production in oceanic waters, *Limnol. Oceanogr.*, 36, 1555-1577, 1991.
- Christian, J. R., M. R. Lewis, and D. M. Karl, Vertical fluxes of carbon, nitrogen, and phosphorus in the North Pacific Subtropical Gyre near Hawaii, *J. Geophys. Res.*, 102, 15667-15677, 1997.
- Cullen, J. J., Hypotheses to explain high-nutrient conditions in the open sea, *Limnol. Oceanogr.*, 36, 1578-1599, 1991.
- de Baar, H. J. W., and others, Synthesis of iron fertilization experiments: from the iron age in the age of enlightenment, *J. Geophys. Res.*, 110, 10.1029/2004JC002601, 2005.
- de Baar, H. J. W., L. J. A. Gerringa, P. Laan, and K. R. Timmermans, Efficiency of carbon removal per added iron in ocean iron fertilization, *Mar. Ecol. Prog. Ser.*, 364, 269-282, 2008.
- Denman, K. L., G. Brasseur, A. Chidthaisong, P. Ciais, P. M. Cox, R. E. Dickinson, D. Hauglustaine, C. Heinze, E. Holland, D. Jacob, U. Lohmann, S. Ramachandran, P. Leite da Silva Dias, S. C. Wofsy, and X. Zhang, Couplings between changes in the climate system and biogeochemistry, in *Climate Change 2007: The physical science basis. Contribution of Working Group I to the Fourth Assessment Report of the Intergovernmental Panel on Climate Change*, edited by S. Solomon, D. Qin, M. Manning,

-
- Z. Chen, M. Marquis, K. Averyt, M. Tignor, and H. Miller, pp. 499-587, Cambridge University Press, Cambridge, 2007.
- Dore, J. E., B. N. Popp, D. M. Karl, and F. J. Sansone, A large source of atmospheric nitrous oxide from subtropical North Pacific surface waters, *Nature*, 396, 63-66, 1998.
- Droop, M. R., 25 years of algal growth kinetics, *Bot. Mar.*, 26, 99-112, 1983.
- Dugdale, R., and F. Wilkerson, Iron addition experiments in the Antarctic: a reanalysis, *Global Biogeochem. Cycles*, 4, 13-19, 1990.
- Dutreuil, S., L. Bopp, and A. Tagliabue, Impact of enhanced vertical mixing on marine biogeochemistry: lessons for geo-engineering and natural variability, *Biogeosciences*, 6, 901-912, 2009.
- Feely, R. A., C. L. Sabine, J. M. Hernandez-Ayon, D. Janson, and B. Hales, Evidence for upwelling of corrosive "acidified" water onto the continental shelf, *Science*, 320, 1490-1492, 2008.
- Fennel, K., J. Wilkin, J. Levin, J. Moisan, J. O'Reilly, and D. Haidvogel, Nitrogen cycling in the Middle Atlantic Bight: Results from a three-dimensional model and implications for the North Atlantic nitrogen budget, *Global Biogeochem. Cycles*, 20, 10.1029/2005GB002456, 2006.
- Fiala, M., and L. Oriol, Vitamine B₁₂ et phytoplancton dans l'océan Antarctique. Distribution et approche expérimentale, *Mar. Biol.*, 79, 325-332, 1984.
- Flynn, K. J., A mechanistic model for describing dynamic multi-nutrient, light, temperature interactions in phytoplankton, *J. Plankton Res.*, 23, 977-997, 2001.
- Forster, P., V. Ramaswamy, P. Artaxo, T. Berntsen, R. Bettes, D. W. Fahey, J. Haywood, J. Lean, D. C. Lowe, G. Myhre, J. Nganga, R. Prinn, G. Raga, M. Schulz, and R. Van Dorland, Changes in atmospheric constituents and radiative forcing, in *Climate Change 2007: The Physical Science Basis. Contribution of Working Group I to the Fourth Assessment Report of the Intergovernmental Panel on Climate Change*, edited by S. Solomon, D. Qin, M. Manning, Z. Chen, M. Marquis, K. B. Averyt, M. Tignor, and H. L. Miller, pp. 129-234, Cambridge University Press, Cambridge, 2007.
- Francois, R., M. A. Altabet, and L. H. Burckle, Glacial to interglacial changes in surface nitrate utilization in the Indian sector of the Southern Ocean as recorded by sediment $\delta^{15}\text{N}$, *Paleoceanography*, 7, 589-606, 1992.
- Friedlingstein, P., and others, Climate - carbon cycle feedback analysis: results from the C⁴MIP model intercomparison, *J. Climate*, 19, 3337-3353, 2006.
- Fuhrman, J. A., and D. G. Capone, Possible biogeochemical consequences of ocean fertilization, *Limnol. Oceanogr.*, 36, 1951-1959, 1991.
- Glibert, P. M., and others, Ocean urea fertilization for carbon credits poses high ecological risks, *Mar. Poll. Bull.*, 56, 1049-1056, 2008.

-
- Gnanadesikan, A., and I. Marinov, Export is not enough: nutrient cycling and carbon sequestration, *Mar. Ecol. Prog. Ser.*, 364, 289-294, 2008.
- Hutchins, D. A., and K. W. Bruland, Iron-limited diatom growth and Si:N uptake ratios in a coastal upwelling regime, *Nature*, 393, 561-564, 1998.
- Karl, D. M., and R. M. Letelier, Nitrogen fixation-enhanced carbon sequestration in low nitrate, low chlorophyll seascapes, *Mar. Ecol. Prog. Ser.*, 364, 257-268, 2008.
- Kumar, N., R. F. Anderson, R. A. Mortlock, P. N. Froelich, P. Kubik, B. Dittrich-Hannen, and M. Suter, Increased biological productivity and export production in the glacial Southern Ocean, *Nature*, 378, 675-680, 1995.
- Lampitt, R. S., Evidence for seasonal deposition of detritus and its subsequent resuspension, *Deep-Sea Res.*, 32, 885-897, 1985.
- Levasseur, M., M. G. Scarratt, S. Michaud, A. Merzouk, C. S. Wong, M. Arychuk, W. Richardson, R. B. Rivkin, M. Hale, E. Wong, A. Marchetti, and H. Kiyosawa, DMSP and DMS dynamics during a mesoscale iron fertilization experiment in the Northeast Pacific - Part I: Temporal and vertical distributions, *Deep-Sea Res. II*, 53, 2353-2369, 2006.
- Lueker, T. J., S. J. Walker, M. K. Vollmer, R. F. Keeling, C. D. Nevison, and R. F. Weiss, Coastal upwelling air-sea fluxes revealed in atmospheric observations of O₂/N₂, CO₂ and N₂O, *Geophys. Res. Lett.*, 30, 10.1029/2002GL016615, 2003.
- Martin, J. H., Glacial-interglacial CO₂ change: the iron hypothesis, *Paleoceanography*, 5, 1-13, 1990.
- Martin, J. H., and G. A. Knauer, The elemental composition of plankton, *Geochim. Cosmochim. Acta*, 37, 1639-1653, 1973.
- Martin, J. H., and others, Testing the iron hypothesis in ecosystems of the equatorial Pacific Ocean, *Nature*, 371, 123-129, 1994.
- Martin, J. H., G. A. Knauer, D. M. Karl, and W. W. Broenkow, VERTEX: carbon cycling in the northeast Pacific, *Deep-Sea Res.*, 34, 267-285, 1987.
- Martin, J. H., R. M. Gordon, and S. E. Fitzwater, The case for iron, *Limnol. Oceanogr.*, 36, 1793-1802, 1991.
- Matear, R. J., and B. Elliott, Enhancement of ocean uptake of anthropogenic CO₂ by macronutrient fertilization, *J. Geophys. Res.*, 109, 10.1029/2000JC000321, 2004.
- Mitchell, B. G., E. A. Brody, O. Holm-Hansen, and C. R. McClain, Light limitation of phytoplankton biomass and macronutrient utilization in the Southern Ocean, *Limnol. Oceanogr.*, 36, 1662-1677, 1991.
- Moore, J. K., S. C. Doney, J. A. Kleypas, D. M. Glover, and I. Y. Fung, An intermediate complexity marine ecosystem model for the global domain, *Deep-Sea Res. II*, 49, 403-462, 2002.

-
- Murnane, R. J., J. L. Sarmiento, and C. Le Quéré, Spatial distribution of air-sea CO₂ fluxes and the interhemispheric transport of carbon by the oceans, *Global Biogeochem. Cycles*, 13, 287-305, 1999.
- Neufeld, Z., P. H. Haynes, V. Garçon, and J. Sudre, Ocean fertilization experiments may initiate a large scale phytoplankton bloom, *Geophys. Res. Lett.*, 29, 10.1029/2001GL013677, 2002.
- Orr, J. C., and others, Anthropogenic ocean acidification over the twenty-first century and its impact on calcifying organisms, *Nature*, 437, 681-686, 2005.
- Pollard, R. T., and others, Southern Ocean deep-water carbon export enhanced by natural iron fertilization, *Nature*, 457, 577-580, 2009.
- Raynaud, D., J. Jouzel, J. M. Barnola, J. Chappellaz, R. J. Delmas, and C. Lorius, The ice core record of greenhouse gases, *Science*, 259, 926-934, 1993.
- Royal Society, Geoengineering the climate: science, governance and uncertainty, RS Policy document 10/09, 2009. (available at <http://royalsociety.org/Geoengineering-the-climate/>)
- Sarmiento, J. L., and J. C. Orr, Three-dimensional simulations of the impact of Southern Ocean nutrient depletion on atmospheric CO₂ and ocean chemistry, *Limnol. Oceanogr.*, 36, 1928-1950, 1991.
- Sarmiento, J. L., N. Gruber, M. A. Brzezinski, and J. P. Dunne, High-latitude controls of thermocline nutrients and low latitude biological productivity, *Nature*, 427, 56-60, 2004.
- Scarratt, M. G., and R. M. Moore, Production of methyl bromide and methyl chloride in laboratory cultures of marine phytoplankton, *Mar. Chem.*, 59, 311-320, 1998.
- Scharek, R., M. Latasa, D. M. Karl, and R. R. Bidigare, Temporal variations in diatom abundance and downward vertical flux in the oligotrophic North Pacific gyre, *Deep-Sea Res.*, 46, 1051-1075, 1999.
- Sonnerup, R. E., P. D. Quay, and J. L. Bullister, Thermocline ventilation and oxygen utilization rates in the subtropical North Pacific based on CFC distributions during WOCE, *Deep-Sea Res.*, 46, 777-805, 1999.
- Strzepek, R. F., and P. J. Harrison, Photosynthetic architecture differs in coastal and oceanic diatoms, *Nature*, 431, 689-692, 2004.
- Stuiver, M., P. D. Quay, and H. G. Ostlund, Abyssal water carbon-14 distribution and the age of the world oceans, *Science*, 219, 849-851, 1983.
- Sunda, W. G., and S. A. Huntsman, Iron uptake and growth limitation in oceanic and coastal phytoplankton, *Mar. Chem.*, 50, 189-206, 1995.
- Takeda, S., Influence of iron availability on nutrient consumption ratio of diatoms in oceanic waters, *Nature*, 393, 774-777, 1998.

-
- Trick, C. G., B. D. Bill, W. P. Cochlan, M. L. Wells, V. L. Trainer, and L. D. Pickell, Iron enrichment stimulates toxic diatom production in high-nitrate, low-chlorophyll areas, *Proc. Natl. Acad. Sci. USA*, 107, 5887-5892, 2010.
- Watson, A. J., and A. C. Naveira Garabato, The role of Southern Ocean mixing and upwelling in glacial-interglacial atmospheric CO₂ change, *Tellus*, 58B, 73-87, 2006.
- Whitney, F. A., W. R. Crawford, and P. J. Harrison, Physical processes that enhance nutrient transport and primary productivity in the coastal and open ocean of the subarctic NE Pacific, *Deep-Sea Res. II*, 52, 681-706, 2005.
- Whitney, F. A., H. J. Freeland, and M. Robert, Persistently declining oxygen levels in the interior waters of the eastern subarctic Pacific, *Prog. Oceanogr.*, 75, 179-199, 2007.
- Wingenter, O. W., K. B. Haase, P. Strutton, G. Friedrich, S. Meinardi, D. R. Blake, and F. S. Rowland, Changing concentrations of CO, CH₄, C₅H₈, CH₃Br, CH₃I, and dimethyl sulfide during the Southern Ocean iron enrichment experiments, *Proc. Nat. Acad. Sci. USA*, 101, 8537-8541, 2004.
- Wingenter, O. W., S. M. Elliot, and D. R. Blake, New directions: enhancing the natural sulfur cycle to slow global warming, *Atmos. Environ.*, 41, 7373-7375, 2007.
- Zahariev, K., J. R. Christian, and K. L. Denman, Preindustrial, historical, and fertilization simulations using a global ocean carbon model with new parameterizations of iron limitation, calcification, and N₂ fixation, *Prog. Oceanogr.*, 77, 56-82, 2008.
- Zeebe, R. E., and D. Archer, Feasibility of ocean fertilization and its impact on future atmospheric CO₂ levels, *Geophys. Res. Lett.*, 32, 10.1029/2005GL022449, 2005.
- Zondervan, I., R. E. Zeebe, B. Rost, and U. Riebesell, Decreasing marine biogenic calcification: a negative feedback on rising atmospheric pCO₂, *Global Biogeochem. Cycles*, 15, 507-516, 2001.