

Contents lists available at [SciVerse ScienceDirect](http://SciVerse.Sciencedirect.com)

Process Safety and Environmental Protection

journal homepage: www.elsevier.com/locate/psep

IChemE

Ocean fertilization for geoengineering: A review of effectiveness, environmental impacts and emerging governance

Phillip Williamson^{a,*}, Douglas W.R. Wallace^b, Cliff S. Law^c, Philip W. Boyd^d, Yves Collos^e, Peter Croot^f, Ken Denman^g, Ulf Riebesell^h, Shigenobu Takedaⁱ, Chris Vivian^j

^a School of Environmental Sciences, University of East Anglia, Norwich NR4 7TJ, UK

^b Department of Oceanography, Dalhousie University, Halifax NS, B3H 4R2 Canada

^c NIWA, Greta Point, PO Box 14-901, Wellington, New Zealand

^d Department of Chemistry, University of Otago, PO Box 56, Dunedin 9054, New Zealand

^e Laboratoire ECOSYM, UMR 5119, Université Montpellier 2, CNRS, Montpellier Cedex 5, France

^f Department of Earth and Ocean Sciences, National University of Ireland, Galway, Ireland

^g School of Earth and Ocean Sciences, University of Victoria, PO Box 3065, STN CSC Victoria, BC, Canada

^h IFM-GEOMAR, Düsternbrooker Weg 20, 24105 Kiel, Germany

ⁱ Faculty of Fisheries, Nagasaki University, Bunkyo-machi 1-14, Nagasaki, Japan

^j Centre for Environment, Fisheries and Aquaculture Science, Lowestoft NR33 0HT, UK

A B S T R A C T

Dangerous climate change is best avoided by drastically and rapidly reducing greenhouse gas emissions. Nevertheless, geoengineering options are receiving attention on the basis that additional approaches may also be necessary. Here we review the state of knowledge on large-scale ocean fertilization by adding iron or other nutrients, either from external sources or via enhanced ocean mixing. On the basis of small-scale field experiments carried out to date and associated modelling, the maximum benefits of ocean fertilization as a negative emissions technique are likely to be modest in relation to anthropogenic climate forcing. Furthermore, it would be extremely challenging to quantify with acceptable accuracy the carbon removed from circulation on a long term basis, and to adequately monitor unintended impacts over large space and time-scales. These and other technical issues are particularly problematic for the region with greatest theoretical potential for the application of ocean fertilization, the Southern Ocean. Arrangements for the international governance of further field-based research on ocean fertilization are currently being developed, primarily under the London Convention/London Protocol.

© 2012 The Institution of Chemical Engineers. Published by Elsevier B.V. All rights reserved.

Keywords: Geoengineering; Negative emission technologies; Ocean fertilization; Iron; Southern Ocean; Governance

1. Introduction

Concern over human-driven climate change and the lack of success in constraining greenhouse gas emissions have increased scientific and policy interest in geoengineering – deliberate intervention in the planetary environment of a nature and scale intended to counteract anthropogenic

climate change and its impacts. Most proposed approaches involve either removing carbon dioxide (CO₂) from the atmosphere by biological or chemical means (Carbon Dioxide Removal, CDR; more generally, Negative Emissions Technologies, NET), or reflecting part of the sun's energy back into space (Solar Radiation Management, SRM). These groups of approaches are intended to reduce future global warming

* Corresponding author. Tel.: +44 1603 593111.

E-mail address: p.williamson@uea.ac.uk (P. Williamson).

Received 8 May 2012; Received in revised form 10 September 2012; Accepted 3 October 2012

by altering the Earth's energy budget, either by increasing the outgoing, long-wave radiation or decreasing the incoming, short-wave component (Royal Society, 2009; Lenton and Vaughan, 2009). Without geoengineering, it is becoming highly unlikely that 'dangerous' climate change can still be avoided (Anderson and Bows, 2011; Myhrvold and Caldeira, 2012).

Here we assess the potential benefits and risks associated with one of the earliest proposed carbon-removal techniques: large-scale ocean fertilization, achieved by increasing the supply of one or more limiting nutrients to surface waters, directly (Martin et al., 1990; Matear and Elliott, 2004) or indirectly (Lovelock and Rapley, 2007). The intention is to enhance the growth of microscopic marine plants on a scale large enough to significantly increase the uptake of atmospheric CO₂ by the ocean, removing it for long enough to provide climatic benefit. Analyses of natural, long-term climate changes (ice age cycles; Sigman and Boyle, 2000) and ship-based *in vitro* experiments (Martin et al., 1990) suggest that the global ocean uptake of carbon is sensitive to nutrient availability, and that for many (but not all) ocean regions, iron would be particularly effective as the fertilizing nutrient for geoengineering purposes.

Proposals to explore the feasibility of such approaches by scaling-up experimental studies have been controversial, attracting scientific and public criticism. As discussed in Section 9 below, the Convention on Biological Diversity (CBD), the London Convention and London Protocol (LC/LP), and UNESCO's Intergovernmental Oceanographic Commission (IOC) are amongst the international bodies with interests in this area. To assist policy and governance development, a summary of scientific understanding of ocean fertilization was prepared jointly by IOC and the non-governmental Surface Ocean-Lower Atmosphere Study (SOLAS)¹ (Wallace et al., 2010). This review extends and updates that assessment.

In addition to its potential for climate geoengineering, ocean fertilization might also be used for the aim of increasing fishery yields (Jones, 2011) and such ideas have commercial proponents². Closely similar regulatory constraints regarding research and operational deployment would apply as for geoengineering; however, the evidence base for fishery enhancement is currently much less-developed, and that potential application is not considered in any detail here.

2. Nutrient supply, marine production and carbon export

Physico-chemical processes in the ocean and ocean sediments will, over many tens of thousands of years, take up most of the CO₂ that will be released through the burning of fossil fuels (Archer, 2005). Ocean fertilization for the purpose of geoengineering aims to increase CO₂ uptake by marine biological processes (the 'biological carbon pump'), in sufficient quantity to achieve a climatically significant reduction in atmospheric levels (Martin et al., 1990; Lampitt et al., 2008).

Photosynthesis by marine phytoplankton (free-living microscopic marine plants) not only requires light and CO₂,

but also relatively large amounts of bio-available macronutrients such as nitrogen (N) and phosphorus (P), together with trace quantities of essential micro-nutrients such as iron (Fe), e.g. for use as enzyme co-factors. These nutrients are naturally available in the ocean, mostly as a result of the death and decomposition of previous generations of marine plants and other organisms (animals and microbes). Whilst such recycling mostly occurs on a timescale of days to weeks in the upper ocean, about a quarter of the nutrient release takes place on a longer timescale in mid- and deep-water, as a result of sinking downward of biological material. Only a very small proportion (usually <1%) of upper ocean production reaches the deep sea floor.

In most of the global ocean, the rate of photosynthesis and hence marine production is limited by the availability of either N or P, either seasonally (e.g. in regions subject to winter mixing and summer stratification) or on a year-round basis (e.g. in permanently stratified regions, where stratification limits nutrient re-supply from deeper water). But in around a third of the global ocean, near-surface N and P levels remain high all year; these nutrients are unused since the concentration of Fe is instead limiting.

Increasing the supply of limiting nutrient(s) to the sunlit surface ocean can be expected to have a fertilizing effect, stimulating phytoplankton growth and potentially enhancing marine production at all trophic levels. Many external processes are also important in supplying nutrients, including iron, to the upper ocean. Sources include rivers and precipitation; atmospheric dust from arid areas and volcanoes; seafloor sediments, vents and seeps; and glacial ice (Boyd and Ellwood, 2010; Breitbarth et al., 2010; Gabric et al., 2010; Duggen et al., 2010; Hamme et al., 2010, Shaw et al., 2011). 'Ocean fertilization' is here considered to be a purposeful human action (at experimental or operational scale), achieved either by directly adding nutrients, or by accelerating nutrient re-supply from deep water. However, ocean fertilization has also been used to describe the naturally increased supply of iron (and other nutrients) around islands in the Southern Ocean (Blain et al., 2007; Pollard et al., 2009; Wolff et al., 2011).

Most ocean fertilization studies – by experiments and models – have to date focused on increasing the external supply of iron as a micro-nutrient. However, fertilization studies have also involved phosphorus additions, and using artificial upwelling to bring naturally nutrient-rich deeper waters to the surface (Lovelock and Rapley, 2007; Karl and Letelier, 2008). The latter technique not only pumps nutrients upwards, but also the CO₂ released from previous cycles of production/export and sinking/decomposition (Shepherd et al., 2007). Enhanced upwelling therefore has limited potential to achieve net drawdown of CO₂ from the atmosphere, and subsequent sequestration of carbon. Nevertheless, there may be some enhancement of ocean carbon sequestration if nitrogen-fixation can be stimulated by increasing the abundance of specialised microbes that can directly use gaseous nitrogen dissolved in seawater (Karl and Letelier, 2008).

For carbon accounting³, the anticipated benefits from large-scale ocean fertilization need to be quantified, within pre-agreed confidence limits (Bertram, 2010). Reliable

¹ The Surface Ocean-Lower Atmosphere Study (SOLAS) is an international research programme co-sponsored by the International Geosphere Biosphere Programme (IGBP), the Scientific Committee on Oceanographic Research (SCOR), the World Climate Research Programme (WCRP) and the International Commission on Atmospheric Chemistry and Pollution (ICACP).

² Ocean Nourishment Corporation Pty Ltd. www.oceannourishment.com.

³ Carbon accounting can be achieved either by commercial carbon trading, or governmental/intergovernmental schemes similar to the one developed under the Kyoto Protocol for reduced emissions from deforestation and degradation (REDD and REDD+).

estimates will be needed not only of the additional amount of carbon initially exported (downward) from the upper ocean, but also the average time period for which it will be sequestered, i.e. removed from the atmosphere. Long-term monitoring will also be needed to determine whether deleterious side effects are within acceptable limits. Natural ocean circulation processes will cause impacts to spread; thus, if fertilization is repeatedly applied to a relatively constrained area, such treatment will subsequently affect very large regions of the ocean on a decadal to century scale, as discussed further in Section 5 below. Trials to properly assess the effectiveness of large-scale ocean fertilization would themselves need to be at the scale of thousands of square kilometres, with measurements over several months, if not years (Smetacek and Naqvi, 2008; Watson et al., 2008; Strong et al., 2009). Such research has yet to be carried out and presents a considerable logistical challenge.

3. Overview of ocean fertilization experiments

3.1. Iron addition

Iron in seawater is mostly in an insoluble form, limiting its biological availability. For field experiments on ocean fertilization, iron has been added as ferrous sulphate ($\text{FeSO}_4 \cdot 7\text{H}_2\text{O}$) which is a common agricultural fertilizer and relatively soluble. The ferrous sulphate is dissolved in acidified seawater, and pumped into the ocean behind a moving vessel. The acidic solution is neutralised rapidly upon mixing with ambient seawater and the iron is transformed chemically into its insoluble (ferric) form. Operational ocean fertilization might be expected to add chemical complexing agents to keep iron in solution for longer; specific techniques have been patented for this purpose (e.g. Lambert, 2011). It is a misconception that iron filings might be used directly for ocean fertilization – their solubility is far too low.

Between 1993 and 2009, 13 relatively small-scale iron fertilization field experiments were performed in the tropical Pacific, Southern Ocean, North Pacific and tropical Atlantic (Fig. 1). For comparative summary information, see Boyd et al. (2007), Strong et al. (2009), CBD (2009). The primary aim of these studies was to determine whether iron was limiting phytoplankton growth in the surface ocean; i.e. they were not designed as geoengineering trials. Each experiment affected a few hundred square kilometres for a few weeks, on a similar scale (and with similar consequences) to a natural bloom of phytoplankton. Although the scale of response has varied, together they have conclusively confirmed that shortage of iron – one of the most abundant elements on land, and constituting around 32% of the mass of the Earth as a whole – limits biological production in many regions of the ocean (Martin et al., 1994; Boyd et al., 2000; Tsuda et al., 2003).

In these experiments, the effects of the added iron on upper ocean carbon cycling (on a short-term basis) have been relatively well-documented. The observed increases in phytoplankton biomass were, as expected, accompanied by reductions in CO_2 levels in surface water (Bakker et al., 2005; Watson et al., 1994), promoting CO_2 drawdown from the atmosphere by gas exchange. The amount of CO_2 drawdown across the air–sea interface has varied greatly between studies (de Baar et al., 2005), even between two experiments carried out

three years apart at the same site in the north west Pacific Ocean (Tsuda et al., 2003; Kudo et al., 2006, 2009).

Explanations for such variability have included differences in the amount of nutrient added; the status of the phytoplankton before the fertilization; the depth of the surface mixed layer; grazing by zooplankton; and the time that fertilized waters remained in direct contact with the atmosphere. Dilution and mixing of the patch seem particularly important: whilst moderate mixing might maintain the supply of other limiting nutrients, such as silicate, into the fertilized patch (Abraham et al., 2000), high dilution might prevent phytoplankton accumulation in iron-fertilized water (Law et al., 2011). Most experiments did not continue for a sufficiently long time period to follow the decline of the stimulated phytoplankton bloom and associated carbon export. Three studies did report increased carbon export, but of different proportions (Boyd et al., 2004; Bishop et al., 2004; Smetacek et al., 2012). The highest of these values (Smetacek et al., 2012) was for relatively unusual hydrographic and biological conditions: the experiment was carried out in the central core of a mesoscale eddy in the Antarctic polar front, and the stimulated bloom of large diatom species was subsequently subject to mass mortality and rapid sinking.

Taken together, the main ecosystem changes observed in these experiments were as follows:

- Levels of the plant pigment chlorophyll-*a* increased in all experiments, by 2–25 times, with associated increases in carbon fixation (Boyd et al., 2007). Several of the artificially induced blooms of phytoplankton were visible to satellite-based ocean colour sensors⁴.
- Phytoplankton responded to the iron addition by altered rates of nutrient uptake and an increase in photosynthetic efficiency (Behrenfeld et al., 1996).
- Effects on phytoplankton production and biomass were greater in shallower surface mixed layers due to the more confined depth range and, consequently, higher average light intensity experienced by the fertilized plankton. Response was more rapid in warmer waters (Boyd, 2002; Boyd et al., 2007).
- In most experiments, the dominant phytoplankton group changed, with a shift in community composition from smaller groups (cyanobacteria) to medium-sized phytoplankton, e.g. haptophytes and larger diatoms (Boyd et al., 2007).
- Although diatoms usually increased, the most abundant diatom species varied between locations and experiments (Tsuda et al., 2005; Marchetti et al., 2006). This may reflect regional species differences of initial ‘seed’ populations as well as competitive effects (Kudo et al., 2009).
- Bacterial production and biomass increased during most experiments, by 2–15 times (Hale et al., 2006; Agawin et al., 2006; Kudo et al., 2009). A transient increase in the stocks of small grazers, microzooplankton, was also reported from some experiments (Saito et al., 2006).
- The duration of the experiments was usually too short to allow larger zooplankton to respond. However, grazing increased in two experiments with high pre-existing stocks of medium-sized zooplankton (copepods), and seems to play a major role in controlling bloom development (Sastri

⁴ <http://disc.sci.gsfc.nasa.gov/oceancolour/additional/science-focus/ocean-colour/science.focus.shtm/iron.limits.shtml>.

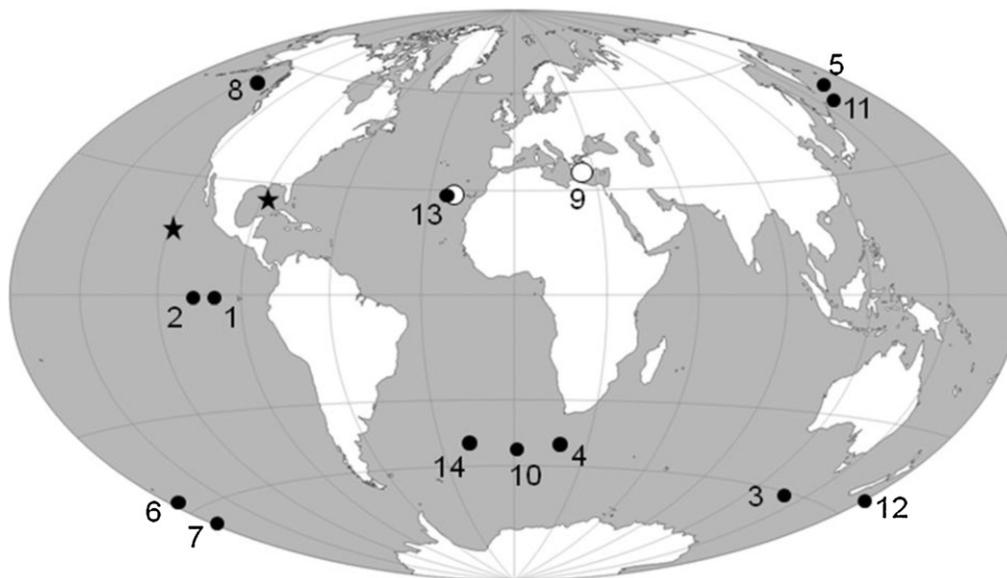


Fig. 1 – Sites of ocean fertilization studies involving patch-scale experimental nutrient additions, 1993–2009. ● Fe-addition experiments; ○ P-addition experiments; ★ commercial trials (two, both using Fe). 1. IronEx I (1993); 2. IronEx II (1995); 3. SOIREE (Southern Ocean Iron Enrichment Experiment, 1999); 4. EisenEx (2000); 5. SEEDS I (Subarctic Pacific Iron Experiment for Ecosystem Dynamics Studies, 2001); 6 and 7. SOFeX North and SOFeX South (Southern Ocean Iron Experiment, 2002); 8. SERIES (Subarctic Ecosystem Response to Iron Enrichment Study, 2002); 9. CYCLOPS (Cycling of Phosphorus in Eastern Mediterranean, 2002); 10. EIFEX, European Iron Fertilization Study, 2004); 11. SEEDS II (2004); 12. SAGE (SOLAS Air–Sea Gas Exchange experiment, 2004); 13. FEED (2004) and 14. LOHAFEX (2009). An additional, unauthorized and commercially-based ocean fertilization study was carried out in July 2012 near to site 8 by the Haida Salmon Restoration Corporation, www.hsrl.com.

and Dower, 2006; Tsuda et al., 2006, 2009; Peloquin et al., 2011).

- Information on responses further up the food chain is limited to fish trawl samples in a single study (Takeda and Tsuda, 2005). Because of the limited duration of sampling, the observed difference (including an increased catch of northern mackerel within the experimental patch) must have been pre-existing or behavioural.

3.2. Macro-nutrient additions

There have been two field experiments involving phosphorus additions, both in low nutrient waters. One used concentrated phosphoric acid mixed with sodium bicarbonate, the other direct addition of anhydrous monosodium phosphate. The solutions were pumped into surface waters behind a moving vessel.

In the Eastern Mediterranean, the experiment resulted in rapid increases in bacterial production and zooplankton biomass, and a moderate increase in rates of nitrogen-fixation (Thingstad et al., 2005). However, there was a slight decrease in phytoplankton biomass and chlorophyll, in contrast to the expected increase (Psarra et al., 2005; Krom et al., 2005). Similar effects on bacteria and phytoplankton were observed off north-west Africa when phosphate was added alone and with iron (Rees et al., 2006). These results are not yet fully explained; they suggest alternative food-chain pathways and/or additional complex limitations operating in low nutrient systems subject to P limitation (Dixon, 2008).

No ocean fertilization experiments have been carried out to date by adding nitrogen in a biologically available form. However, large-scale addition of synthetic urea ($(\text{NH}_2)_2\text{CO}$) has been proposed for fishery enhancement (Jones, 2011), either as a liquid mixed with phosphate solution and seawater, or

as spherical grains spread over the ocean surface. Modelling of the effects of large-scale macro-nutrient fertilization has indicated that the efficiency of this process for carbon sequestration is likely to be affected by changes in calcium carbonate production and other factors (Matear and Elliott, 2004).

3.3. Nutrient co-limitation and fate

The continued addition of an initially limiting nutrient will necessarily result in another factor becoming limiting. Most iron additions to high nutrient regions caused marked depletions of silicate (required by diatoms; Brzezinski et al., 2011) and nitrate (required by all phytoplankton); lack of the former is considered responsible for constraining or ending diatom bloom development in several experiments (Boyd et al., 2004; Harvey et al., 2011). Increasing patch size – for geoengineering-scale deployment – is likely to accentuate such effects, reducing potential carbon sequestration by as much as an order of magnitude (Ianson et al., 2012). For phosphate addition experiments in low nutrient regions, the biological response was probably limited by nitrogen availability (Thingstad et al., 2005). Light can also be an additional limiting factor, especially in polar regions (Cassar et al., 2011), due to season, cloud cover, deep mixing and self-shading caused by phytoplankton themselves.

The fate of externally added nutrients depends on their chemical nature. Several experiments with iron required re-fertilization because the added iron rapidly ‘disappeared’, either through formation of organic complexes (Rue and Bruland, 1997; Kondo et al., 2008) or through adsorption onto particles which sank (Bowie et al., 2001; Gordon et al., 1998; Nishioka et al., 2009). Thus added iron can be lost from surface waters before it is used by plankton, and much may be removed from the ocean permanently through burial of

particles in sediments. In the case of fertilization with phosphorus or nitrogen-containing compounds, the added nutrients are expected to be incorporated rapidly into biomass, to be subsequently recycled and released through decomposition in surface or subsurface waters, along with carbon, with relatively little being lost to sediments.

3.4. Artificial upwelling

A range of devices have been proposed to bring deeper, nutrient-rich water to the sunlit, upper ocean, potentially self-powered by wave energy and one-way valves, or by temperature and salinity gradients. For example, the ‘ocean pipes’ concept (Lovelock and Rapley, 2007) envisages a network of pipes, each ~10 m diameter and 100–300 m long, distributed across regions with low surface nutrient concentrations. Pipes could either be free-floating or tethered to the seafloor. Such a system is under commercial development by Atmocean Inc.⁵

The design of artificial upwelling devices needs to be technologically robust for structural longevity in the variable and complex hydrodynamics of the upper ocean. Enhanced primary production (without CO₂ measurements) has been demonstrated over ~30 days (Maruyama et al., 2011); however, other devices developed to date (Tsubaki et al., 2007; White et al., 2010) have not been deployed for long enough for the expected biological and biogeochemical responses to be observed. Modelling studies (Dutreuil et al., 2009; Yool et al., 2009; Oschlies et al., 2010b), indicate major uncertainties concerning ecosystem response and whether net CO₂ drawdown is achievable, primarily because dissolved inorganic carbon, as well as nutrients, will be brought to the surface in the upwelled water (Shepherd et al., 2007). Surface temperature and salinity will also be affected, and if enhanced upwelling is carried out on a sufficiently large scale, the former could have significant climatic implications (Oschlies et al., 2010b).

At some localities, fertilization effects may be partly indirect, as a result of high phosphate levels in upwelled water stimulating nitrogen-fixation (Karl and Letelier, 2008). Overall, it seems more likely that artificial upwelling will become a tool to study marine ecosystem responses to nutrient perturbations and changes in mixing regimes (Masuda et al., 2010), rather than a cost-effective measure to significantly counteract climate change.

4. Efficiency of large-scale ocean fertilization for climate geoengineering

For externally added nutrients, the overall efficiency of ocean fertilization as a means to sequester atmospheric CO₂ can be expressed as the ratio between the amount of carbon removed from global circulation on a long-term basis (e.g. for at least 100 years) and the amount of added nutrient (de Baar et al., 2008). Although this ratio need not be directly equivalent to cost-effectiveness, the economic attractiveness of ocean fertilization as a geoengineering approach will be self-evidently greater if large amounts of carbon are sequestered by relatively small quantities of added nutrient.

Twenty years ago, iron-based ocean fertilization looked like a highly efficient process. Based on the C:Fe ratio in phytoplankton (Sunda et al., 1991), it was calculated that 1 tonne of added iron might sequester more than 100,000 tonnes of

carbon. Current estimates are that even the natural (re-)supply of dissolved iron to iron-depleted waters is at least an order of magnitude less efficient (Blain et al., 2007; Pollard et al., 2009), whilst experimental treatments – and natural atmospheric inputs, via dust – seem to be several orders of magnitude less efficient (Boyd et al., 2004; Buesseler et al., 2004; Gabric et al., 2010).

Fig. 2 depicts key processes affecting the overall efficiency for ocean fertilization using added nutrients, such as iron. Solid arrows indicate the intended effects, i.e. increased carbon sequestration; open arrows indicate other relevant processes that may be counter-active, decreasing carbon uptake efficiencies. The thicker open arrows are relatively rapid processes (days to months), the thinner arrows are slower (years to decades). The *carbon export ratio* relates the amount of added nutrient to the amount of carbon leaving the upper ocean: it is controlled by a range of factors not yet well-represented in models (Boyd and Trull, 2007) including nutrient loss processes (e.g. iron removal by precipitation and scavenging onto particles before it can be used by phytoplankton); the nutrient-to-carbon ratio in fertilized biomass; variable sinking rates, and the rate of grazing of phytoplankton by zooplankton (and vertical migration by the latter). Improved delivery mechanisms for iron have been patented (e.g. Lambert, 2011) and these might improve the carbon export ratio, but with significant financial implications.

Carbon export from the upper ocean is not the bottom line for calculating overall efficiency: another crucial consideration is the *atmospheric uptake efficiency*, the proportion of the additional carbon export that is re-supplied by carbon from the atmosphere, rather than by marine mixing processes. This efficiency component depends on factors which determine the rate of air–sea gas exchange, such as wind and waves, and the rate at which exported particulate organic carbon is re-mineralised to CO₂ by bacteria or zooplankton before it reaches the deep ocean, at around 1000 m. Such ‘flux attenuation’ processes are spatially and temporally variable, and poorly understood (Buesseler and Boyd, 2009).

Model-based estimates of atmospheric uptake efficiency suggest values of up to 70–90% are possible, at least for tropical waters (Aumont and Bopp, 2006; Zahariev et al., 2008; Pan 2011; Jin et al., 2008). Field-based estimates of atmospheric uptake efficiency based on CO₂ drawdown in iron addition experiments were much lower, at only 2–20% (Watson et al., 1994; Bakker et al., 2005). However, uptake of CO₂ may have continued after measurements ended.

Using the highest estimates for both carbon export ratios and atmospheric uptake efficiencies, the overall potential for ocean fertilization to remove CO₂ from the atmosphere, based on global-scale fertilization effort over 100 years, has been calculated as 25–75 Gt (gigatonnes) of carbon (Aumont and Bopp, 2006; Zahariev et al., 2008). Most of this potential uptake would occur in the Southern Ocean. This figure compares to cumulative emissions of 900–2000 Gt carbon from fossil fuel burning for a range of IPCC future emission scenarios (Denman, 2008); i.e. geoengineering via ocean fertilization could only make a relatively modest contribution (<10%) to offsetting future emissions and hence counter-acting anthropogenic climate change.

As already noted, the proposed enhancement of the biological pump by artificial upwelling is inherently less efficient for CO₂ sequestration. Whilst initial modelling indicates that global deployment of pipes could significantly alter biological production and export of carbon, net changes to air–sea

⁵ <http://www.atmocean.com>.

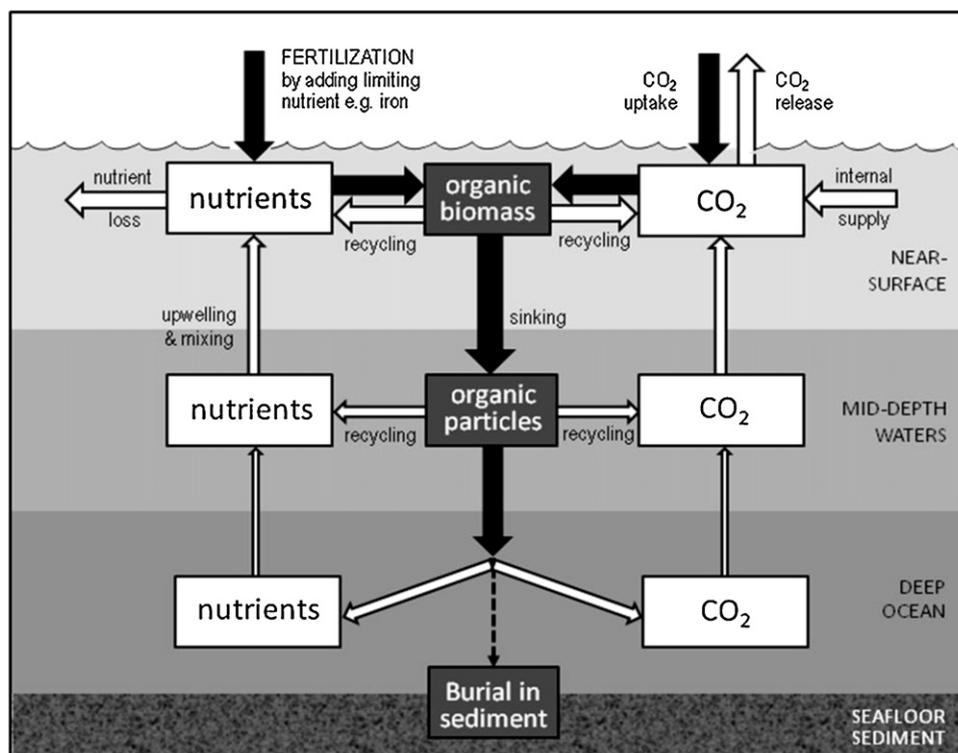


Fig. 2 – Processes affecting the sequestration efficiency of large-scale ocean fertilization based on the addition of iron or other limiting nutrients. The near-surface ocean is the sunlit, mixed layer (usually 50–200 m) that is able to rapidly exchange gases with the atmosphere, and where biological carbon fixation occurs. Processes in mid-depth waters (also known as the mesopelagic layer or twilight zone) affect the decomposition of exported organic carbon, and its return to the atmosphere on a timescale of years to decades. The deep ocean (below ~1 km) is characterized by stable temperatures and very slow circulation and mixing; carbon reaching that part of the ocean is isolated from the climate system on a timescale of centuries to millennia.

CO₂ uptake are expected to be small (Dutreuil et al., 2009; Yool et al., 2009; Oschlies et al., 2010b). This is because most carbon exported from the surface layer is decomposed and recycled at depths <500 m, and the ocean pipes would enhance its return to the surface. Alternative scenarios, yet to be investigated, could involve more complex manipulations of the nutrient supply rate (Masuda et al., 2010), or the targeted stimulation of nitrogen-fixing organisms, or of organisms that can sink deep into the ocean.

Most model simulations for large-scale ocean fertilization are for periods of 10–100 years (Denman, 2008) and such treatments would need to be maintained on such time-scales for their maximum climatic benefits to be realised. The CO₂ sequestration potential on a multi-century timescale depends on what happens when artificially CO₂ enriched deep waters are eventually returned to the ocean surface. Such waters will increase the surface ocean partial pressure of CO₂, and thereby either slow down or reverse the transfer of atmospheric CO₂ to the ocean. The fate of this CO₂-enriched water also depends on the nature of the nutrient used for fertilization. If the nutrient is re-released to deep waters via decomposition in the same proportion to carbon as used for growth, then the added nutrient can be considered to be recycled. When such recycled nutrient is upwelled, it can fuel another cycle of growth, carbon uptake and sinking so that the original extra carbon remains in the ocean. However, if the fertilizing nutrient is removed permanently from the ocean by burial in sediments (the likely fate of added iron), then the nutrient is

unavailable when the CO₂-enriched deep water is brought to the surface again by upwelling processes – and much of the extra CO₂ drawdown resulting from the initial fertilization will be returned to the atmosphere.

5. Unintended impacts of large-scale ocean fertilization

A range of unintended and mostly undesirable impacts of large-scale fertilization are considered by Wallace et al. (2010). As discussed below, these include production of climate-relevant gases, that might either reinforce or offset the benefits of CO₂ sequestration; far-field effects on productivity; mid-water oxygen decrease; changes in spatial patterns of ocean acidification; and effects on seafloor ecosystem. Whilst changes in upper ocean ecosystems can be considered intended rather than unintended, aspects of such changes may also be considered undesirable.

5.1. Production of climate-relevant gases

Iron fertilization has been observed to increase upper ocean concentrations of a range of climate-relevant gases associated with phytoplankton growth (Law, 2008). Of these, the best studied is dimethylsulphide (DMS) which, after emission to the atmosphere, might enhance the formation of particles that promote cloud formation and so influence climate. Most iron fertilization experiments have shown increased DMS

production (Turner et al., 1996, 2004; Wingenter et al., 2004), and such results have been extrapolated to suggest that fertilization of 2% of the Southern Ocean could decrease temperatures by $\sim 2^\circ\text{C}$ in that region. However, fertilization studies in the sub-Arctic Pacific have shown DMS decrease (Levasseur et al., 2006) or no effect (Nagao et al., 2009); furthermore, recent studies indicate that the linkage between DMS and climate is weaker than previously thought (Woodhouse et al., 2010; Quinn and Bates, 2011). Several other volatile trace gases have been observed to have altered concentrations in the upper ocean after fertilization (Wingenter et al., 2004; Walter et al., 2005; Law and Ling, 2001; Hashimoto et al., 2009; Kato et al., 2009; Liss et al., 2005), with potential climatic implications, e.g. via effects on tropospheric ozone concentrations. The overall significance of such upper ocean effects is currently unclear (Law, 2008).

There are also uncertainties regarding the scale of mid- and deep-water production of nitrous oxide (N_2O) and methane (CH_4), due to decomposition of additional sinking biomass under low oxygen conditions. These greenhouse gases have global warming potentials 320 times and 20 times greater than CO_2 respectively; thus any increase in their production that subsequently reaches the atmosphere would be of serious concern, potentially offsetting the desired effects of CO_2 sequestration (Fuhrman and Capone, 1991; Law, 2008; Jin and Gruber, 2003).

Methane is considered the lower risk (Smetacek and Naqvi, 2008), since most of this gas naturally produced within the ocean is used as an energy source by other marine microbes and converted to CO_2 before reaching the sea surface (Naqvi et al., 2010). The ocean is, however, a globally significant source of N_2O and any enhanced production is likely to be emitted to the atmosphere. In particular, if fertilization takes place over waters that are already low in oxygen (e.g. the tropics), models indicate that the N_2O yield could be large, offsetting 40–70% of the benefits of CO_2 reduction after 100 years. The offsetting would be much lower ($\sim 10\%$) for fertilization of waters underlain with higher oxygen concentrations, such as in the Southern Ocean (Jin and Gruber, 2003). Assessments of overall climate forcing depend critically on the accuracy of ocean circulation models, the representation of oxygen in these models, and our limited knowledge of N_2O yield during biomass decomposition. Only minor increases in N_2O production have been observed during iron addition experiments (Law and Ling, 2001; Walter et al., 2005); at this scale only transient and highly dispersed effects are likely, without ecological or climatic significance.

5.2. Far-field effects

Far-field effects, hundreds or thousands of kilometres from a (large-scale) fertilization site and occurring months, years or decades afterwards, include potential impacts on subsurface waters and sediments into which the fertilized biomass sinks (Fuhrman and Capone, 1991; Denman, 2008). Prediction and assessment of far-field impacts requires information on biomass production and sinking, as well as on the circulation and mixing of both the fertilized surface waters and the subsurface waters beneath the fertilized location. Such information can then be used in complex models which simulate ocean circulation, biology and chemistry (Aumont and Bopp, 2006; Jin et al., 2008; Oschlies et al., 2010a). However, model predictions

of far-field effects will be difficult to verify with direct observations because of the large spatial and time-scales involved.

An important far-field consequence of large-scale fertilization with limiting nutrients (e.g. with iron in a high nutrient region) involves the depletion of other non-limiting nutrients, such as nitrate or phosphate. This depletion can, in turn, reduce the productivity of remote regions downstream of the fertilization location, particularly where natural sources of the fertilizing nutrient are available, e.g. iron from shelf sea sediments or atmospheric dust (Aumont and Bopp, 2006; Jin et al., 2008; Gnanadesikan and Marinov, 2008). Thus it is possible that fertilization of an open ocean location in international waters could reduce productivity around islands and countries not involved with the fertilization activity ('nutrient robbing'; Royal Society, 2009). Models have examined the scale of such effects and, for scenarios involving large-scale fertilization over long periods, large reductions in far-field productivity are indicated (Gnanadesikan et al., 2003; Aumont and Bopp, 2006). Such models have their limitations, since not all relevant biogeochemical and physical parameters are well parameterised. Nevertheless, any such reductions could have significant consequences, including a re-distribution or overall decrease in fish production.

Far-field effects are not necessarily negative: increased nutrient levels in deep ocean waters (due to decomposition of the biomass that was increased by fertilization) may enhance the productivity of ecosystems in other remote regions, where these waters are eventually returned to the surface ocean by upwelling or mixing.

5.3. Mid-water oxygen decrease

Decomposition of fertilization-enhanced biomass will necessarily decrease oxygen levels in the ocean interior. Such impacts may be local or remote, depending on circulation patterns, and could lead to critical thresholds or tipping points being crossed. Mid-water oxygen depletion has not been reported for fertilization experiments conducted to date due to their limited scale and duration, but additional oxygen demand is an inevitable consequence of enhanced downward carbon export. Decreased oxygen levels close to the site of fertilization might precondition mid-depth waters so that they cross a critical threshold during subsequent transport through the ocean interior (e.g. towards oxygen minimum zones).

Early studies using highly simplified 'box models' predicted that large volumes of the ocean interior would become anoxic as a consequence of large-scale and continuous fertilization (Fuhrman and Capone, 1991; Sarmiento and Orr, 1991). More sophisticated models, based on more likely fertilization scenarios, predict a less dramatic scenario involving growth of the extent of low-oxygen regions rather than oceanic anoxia (Oschlies et al., 2010a). Fertilization-induced oxygen depletion of mid-depth waters that supply certain upwelling systems and oxygen minimum zones could, however, cause increased frequency and intensity of near-shore hypoxia and, as a consequence, significant mortality of marine organisms (Chan et al., 2008). Important within-ocean nutrient recycling processes might also be altered. Current models have inherent limitations in their ability to represent existing oxygen distributions, hence predictions of change in oxygen levels must be considered uncertain.

5.4. Effects on seafloor ecosystems

The effect of large-scale ocean fertilization on seafloor ecosystems depends critically on the water depth where the fertilization takes place and the sinking speeds of the particulate biomass produced. In deep waters, a large proportion of any enhanced carbon flux will be decomposed before reaching the sea floor. The enhanced carbon flux to the seafloor is likely to increase the amount of seafloor biomass, as long as oxygen is not depleted; this might have either a positive or negative effect on seafloor biodiversity, depending on its background state (Lampitt et al., 2008; Levin et al., 2001). In those parts of the Southern Ocean where upper ocean iron supply is locally enhanced through natural processes, benthic invertebrate biomass at ~4200 m water depth can be three times higher, and abundances six times higher, than in comparable adjacent regions with lower iron and lower primary production (Wolff et al., 2011).

5.5. Ocean acidification

The potential climatic benefits of ocean fertilization as a geoengineering technique are due to reductions in atmospheric CO₂. Such reductions would also reduce the rate of acidification of the upper ocean, since the two processes are very closely linked (Williamson & Turley, 2012). However, the problem has been moved, not eliminated: large-scale fertilization would lead to substantive additional CO₂ sequestration at depth, hence increasing the rate of acidification of ocean interior waters (Cao and Caldeira, 2010; Oschlies et al., 2010a). Such pH changes would alter the depth at which carbonate biominerals start to dissolve, potentially restricting the habitat of deep-ocean organisms that build shells and other structures out of these biominerals, e.g. deep-sea corals, molluscs and crustacea.

5.6. Changes in upper ocean ecosystems

In addition to changes in total primary production and biomass, it is near-inevitable that large-scale ocean fertilization will change the relative abundance of different species in the treated areas, over a wide taxonomic range. The frequently observed increase in diatom biomass in iron experiments to date might be considered beneficial, although any increase in species responsible for 'harmful algal blooms' would be a cause for concern. Shipboard experiments suggest that phytoplankton that produce the toxin domoic acid might increase in abundance in response to iron fertilization, and their rate of toxin production might also be raised (Trick et al., 2010). Re-examination of phytoplankton samples from equatorial Pacific and Southern Ocean iron fertilization studies provides supporting evidence for this unintended impact (Silver et al., 2010), although the effect was not found in a subarctic Pacific fertilization study. 'Non-deliberate' ocean fertilization with nitrogen-containing urea, through sewage, is known to favour the growth of cyanobacteria and dinoflagellates, including toxic species (Glibert et al., 2008).

As already indicated, fertilization experiments carried out to date have been of insufficient duration and spatial scale to reveal biodiversity changes at higher levels within the food chain. Nevertheless, a sustained switch to predominantly diatoms caused by purposeful fertilization (and required for significant CO₂ sequestration) would be expected also to cause a shift in the dominant zooplankton feeding on the

phytoplankton. The implications for fish stocks remain speculative. Other issues relating to ecosystem changes are discussed below.

6. Reversibility

Reversibility is an important consideration affecting the acceptability of potential geoengineering approaches (Royal Society, 2009). There is consensus within the scientific community that none of the iron fertilization field experiments conducted to date could have caused longterm alteration of ocean ecosystems. Thus the individual fertilizations of several hundred square kilometres of ocean surface, each with ~10 tonnes of iron sulphate, represent a scale comparable to natural bloom events, having effects limited to a few weeks or months (Buesseler et al., 2004; de Baar et al., 2005; Boyd et al., 2007).

However, the findings from ocean fertilization experiments to date cannot be directly scaled up to the much larger scales envisioned for operational geoengineering. Purposeful fertilization on a scale large enough to cause a measurable change in atmospheric CO₂ will necessarily also cause major alterations to the structure of regional ocean ecosystems, affecting the seafloor and ocean interior (e.g. via pH decreases and de-oxygenation) as well as the upper ocean. Large-scale regime shifts can occur naturally within marine ecosystems; for example, the relatively abrupt changes observed across a range of trophic levels in the subarctic North Pacific ecosystem in 1977, with a return to more or less the initial state observed in 1989 (Hare and Mantua, 2000). The biological indicators of the regime shift were more clearly obvious than the physical factors, which were presumed to have been the causative factors. If a similar change were to occur at the same time as large-scale ocean fertilization, it could be near-impossible to distinguish cause and effect, or to restore the system to its previous condition.

7. Verification and monitoring

Large-scale ocean fertilization for geoengineering must necessarily include measurement-based estimates of the amount of carbon sequestered (Rickels et al., 2010; Leinen, 2008; Powell, 2008). Such verification requires:

- In situ monitoring of changes in the downward carbon export (Cullen and Boyd, 2008), in both the fertilized areas and adjacent areas that were not fertilized but were otherwise similar.
- Long-term (months to years) and far-field monitoring to determine if there are subsequent rebound effects that might offset some of the initial change, or might have other negative impacts. Measured parameters should include full-depth profiles of oxygen and nitrous oxide (N₂O), as well as far-field reductions in surface nutrient levels that might decrease carbon sequestration and productivity elsewhere (Cullen and Boyd, 2008; Law, 2008).

The necessary monitoring of large scale ocean fertilization to quantify its intended effects will greatly increase its basic deployment costs. Estimates of the latter over the past 20 years have covered a wide range (5–330 US\$ per tonne C sequestered, using iron; Boyd, 2008); the most recent (Markels et al., 2011) is \$71 per tonne C, although not taking account

of atmospheric uptake efficiency nor monitoring/verification costs. In our opinion (with several of us having been involved in past ocean fertilization experiments), adequate information on both intended and unintended impacts would be extremely challenging, if not impossible, to obtain with currently available observing capabilities. The difficulties of assessing ‘control’ conditions will become greater as the scale of operational ocean fertilization is increased to a climatically significant level; furthermore, if the rate of climate change does continue to accelerate over the next 50–100 years (despite mitigation and/or geoengineering efforts) then hydrodynamic changes and other climate-driven ecosystem perturbations may add to the problem of distinguishing geoengineering impacts from those that would have happened anyway.

8. Focus on the Southern Ocean

There are a number of reasons why the Southern Ocean is of particular interest for ocean fertilization in a geoengineering context. The most basic is that modelling studies show that adding iron to that region has the greatest potential to remove CO₂ from the atmosphere (Aumont and Bopp, 2006; Oschlies et al., 2010a). That outcome is consistent with early ideas (Martin et al., 1990) based on the abundance of unused plant nutrients, and evidence that increases in iron supply to waters around Antarctica, mainly via dust, could account for up to 30% of the decrease in atmospheric CO₂ during past glacial cycles (Sigman and Boyle, 2000). The Southern Ocean has therefore received particular scientific attention, with seven ocean fertilization experiments, both polar and sub-polar.

Although the polar iron additions resulted in blooms of heavily silicified diatoms (such as *Fragilariopsis kerguelens*), the drawdown of other nutrients was incomplete (e.g. <10% for NO₃; Boyd et al., 2007). Algal growth rates were much slower than those induced by iron in sub-polar or tropical waters; as a result, bloom peak and decline were not directly observed over the typical duration (weeks) of each study. Furthermore, the relatively strong circulation and mixing in Southern Ocean polar waters enlarged the area of iron-enrichment, diluting its influence on phytoplankton growth. Studies of natural enhancement of iron availability around Kerguelen (Blain et al., 2007) and the Crozet Islands (Pollard et al., 2009) provide insights into larger-scale effects over longer time periods; however, the bathymetry of those locations slows the lateral water flow, whilst also causing both iron and macronutrients to be upwelled. Thus caution is needed in extrapolating from island-influenced studies to geoengineering scenarios for the wider Southern Ocean.

Three further features of waters around Antarctica are relevant in this context. Firstly, that they are currently characterised by >10 different iron sources, several of which are associated with the cryosphere (Boyd and Ellwood, 2010). Major uncertainties exist on how most of these supply mechanisms function; how they might naturally vary on annual to decadal timescales; and how they might alter in a changing climate. Such uncertainties would affect verification and impact monitoring for large-scale fertilization, particularly since satellite imagery may be unable to reliably distinguish iron-induced blooms from natural ones. Secondly, the open ocean areas are logistically challenging for most of the year, with implications for commercial-scale geoengineering deployments, as well as associated verification and monitoring. Thirdly, and perhaps most crucially, the relatively pristine

nature of Antarctica and its surrounding waters has resulted in special conservation protection for the Southern Ocean south of 60°S, via the Protocol on Environmental Protection to the Antarctic Treaty, also known as the Madrid Protocol⁶. Such regulations are in addition to those developed by the London Convention/London Protocol (see below), and have required additional permissions and reporting for the experimental studies carried out to date in such waters.

9. Acceptability and governance

Ocean fertilization proposals have been highly controversial in recent years, with a wide diversity of views about potential risks and benefits. Such proposals raise issues beyond technical scientific assessments due to their potential social, economic and ethical impacts (Gardiner, 2011; Galaz, 2012; CBD, 2012). There is a significant literature developing on the ethical, cultural and legal issues surrounding geoengineering in general and ocean fertilization in particular (e.g. Hale and Dilling, 2011; Orbach, 2008; Freestone and Rafuse, 2008).

The United Nations General Assembly has encouraged States to support the further study and enhance understanding of ocean fertilization (Resolution 62/215; December 2007). Four UN bodies and associated secretariats have major interests in this topic: the Intergovernmental Oceanographic Commission of UNESCO (IOC), the Convention on Biological Diversity (CBD), the London Convention/London Protocol (LC/LP) and the UN Convention on Law of the Sea (UNCLOS). Together they cover the spectrum of marine science, marine conservation and pollution regulation.

In June 2007, LC/LP Scientific Groups issued a Statement of Concern⁷ about ocean fertilization. This was subsequently endorsed by the LC/LP Governing Bodies, who agreed that the scope of work of the LC/LP included ocean fertilization; the LC/LP was competent to address this issue; and that it would further study the associated scientific and legal perspectives⁸.

In October 2008, LC/LP Parties decided that: given the present state of knowledge, ocean fertilization activities other than legitimate scientific research should not be allowed; they would further consider a potentially legally binding resolution or an amendment to the London Protocol on ocean fertilization; and they would also develop a framework for assessing the compatibility of ocean fertilization experiments with the London Convention and Protocol. The definition of ocean fertilization excluded “conventional aquaculture, or mariculture, and the creation of artificial reefs”.

In October 2010, the Governing Bodies of the London Convention and Protocol adopted the Ocean Fertilisation Assessment Framework⁹. Subsequently, a group of countries who wanted to develop a legally binding mechanism for ocean fertilization agreed to work together informally to develop an

⁶ Protection of the Antarctic environment as a wilderness with aesthetic and scientific value shall be a fundamental consideration of activities in the area.

⁷ LC/LP (2007) Statement of concern regarding iron fertilization of the oceans to sequester CO₂, LC-LP.1 Circ 14.

⁸ LC/LP (2008) Main results of the 29th Consultative Meeting and the 2nd Meeting of Contracting Parties. LC-LP.1 Circ 18. Also IMO News Release November 2007 ‘Large-scale fertilization operations not currently justified, say Parties to international treaties’ online at: http://www.imo.org/blast/mainframe.asp?topic_id=1472&doc_id=8706.

⁹ http://www.imo.org/blast/mainframemenu.asp?topic_id=1969.

amendment proposal, to potentially include other types of marine geoengineering as required. This work is on-going.

In response to concerns that large-scale ocean fertilization might be attempted before its consequences were fully understood, the 2008 CBD Conference of Parties (CoP) requested Parties, and urged other governments, to ensure that ocean fertilization activities do not take place until there is an adequate scientific basis on which to justify such activities. This justification should include an assessment of associated risks, and a global, transparent and effective control and regulatory mechanism is in place for these activities, with the exception of small scale scientific research studies within coastal waters. The 'coastal waters' exception was intended to recognise that territorial seas and other maritime jurisdiction zones already gave states the responsibility for conserving and managing their own marine resources.

The CBD Secretariat subsequently published a review (CBD, 2009) of the impacts of ocean fertilization on marine biodiversity, with its main conclusion being that sound and objectively verifiable scientific data of such impacts are scarce. To provide such information, the CBD review considered that more extensive and targeted field work, and better models of marine processes, were needed. At the 2010 CBD CoP, its previous guidance on ocean fertilization was re-affirmed and a much broader decision on geoengineering was agreed. This invited Parties and other Governments not to allow climate-related geoengineering activities that might affect biodiversity to take place until there is an adequate scientific basis on which to justify them, with appropriate consideration of the associated risks for the environment and biodiversity, and associated social, economic and cultural impacts. This was qualified by an exception for small scale scientific research studies conducted in a controlled setting, if justified by the need to gather specific scientific data and subject to a thorough prior assessment of the potential impacts on the environment.

The IOC considered issues relating to ocean fertilization at its 25th Assembly (June 2009) and its 43rd Executive Council (June 2010). The IOC has been closely involved in the CBD and LC/LP discussions. IOC Member States have agreed that the precautionary principle is fundamental to the regulation of ocean fertilization, and reasserted that IOC's main role is to respond to requests for scientific or technical information and advice from relevant bodies or Member States.

It should be noted that the precautionary principle exists in many versions, and particular care is needed in its application to geoengineering (Elliott, 2010). The LC/LP does not consider that the precautionary principle prohibits all activities with uncertain consequences; instead, it requires that the balance of potential risks and benefits is carefully assessed. Thus regulatory arrangements for ocean fertilization research are based on an appraisal of expected impacts, with the requirement that only slight, if any, environmental harm would result. The LC/LP regulatory mechanisms could therefore provide a model for wider governance of geoengineering techniques that might have environmental impacts at the transnational scale (CBD, 2012).

10. Conclusions

The scientific study of ocean fertilization since the early 1990s has greatly improved our understanding of the biological and physico-chemical processes involved, providing information

relevant to the potential application of the technique for large-scale carbon sequestration, as a geoengineering approach.

Not all uncertainties have been resolved, and the high variability of oceanic environments (i.e. low reproducibility over time at the same site) means that a unified description of marine-based geoengineering cannot yet be achieved. Nevertheless, research results to date, taken together, do not support the idea that ocean fertilization would provide a particularly effective approach to counteract the increasing atmospheric levels of CO₂, even within a wider portfolio of measures. The avoidance of undesirable climate change therefore requires more direct and urgent policy action.

Acknowledgements

We are grateful for the guidance and advice provided by the Surface Ocean Lower Atmosphere Study (SOLAS) research community, and by secretariat staff of the Intergovernmental Oceanographic Commission of UNESCO (IOC) and the London Convention/London Protocol (LC/LP) in the preparation of this synthesis. We also thank Phoebe Lam for her significant input to early discussions, and reviewers for their constructive comments. PW's contribution to this study was supported by the UK Natural Environment Research Council.

References

- Abraham, E.R., Law, C.S., Boyd, P.W., Lavender, S.J., Maldonado, M.T., Bowie, A.R., 2000. Importance of stirring in the development of an iron-fertilized plankton bloom. *Nature* 407, 727–729, <http://dx.doi.org/10.1038/35037555>.
- Agawin, N.S.R., Hale, M.S., Rivkin, R.B., Matthews, P., Li, W.K.W., 2006. Microbial response to a mesoscale iron enrichment in the NE subarctic Pacific: bacterial community composition. *Deep-Sea Res. Part II: Topical Stud. Oceanogr.* 53, 2248–2267, <http://dx.doi.org/10.1016/j.dsr2.2006.05.040>.
- Anderson, K., Bows, A., 2011. Beyond 'dangerous' climate change: emission scenarios for a new world. *Phil. Trans. R. Soc.* 369, 20–44, <http://dx.doi.org/10.1098/rsta.2010.0290>.
- Archer, D., 2005. Fate of fossil fuel CO₂ in geologic time. *J. Geophys. Res.* 110, <http://dx.doi.org/10.1029/2004JC002625>, C09S05.
- Aumont, O., Bopp, L., 2006. Globalizing results from ocean in situ iron fertilization studies. *Global Biogeochem. Cycles* 20, <http://dx.doi.org/10.1029/20905GB002591>, GB2017.
- Bakker, D.C.E., Bozec, Y., Nightingale, P.D., Goldson, L., Messias, M.-J., de Baar, H.J.W., Liddicoat, M., Skelvan, I., et al., 2005. Iron and mixing affect biological carbon uptake in SOIREE and EisenEx, two Southern Ocean iron fertilisation experiments. *Deep-Sea Res. Part I: Oceanogr. Res. Papers* 52, 1001–1019, <http://dx.doi.org/10.1016/j.dsr.2004.11.015>.
- Behrenfeld, M.J., Bale, A.J., Kolber, Z.S., Aiken, J., Falkowski, P.G., 1996. Confirmation of iron limitation of phytoplankton photosynthesis in the equatorial Pacific Ocean. *Nature* 383, 508–511, <http://dx.doi.org/10.1038/383508a0>.
- Bertram, C., 2010. Ocean iron fertilization in the context of the Kyoto protocol and the post-Kyoto process. *Energy Policy* 38, 1130–1139, <http://dx.doi.org/10.1016/j.enpol.2009.10.065>.
- Bishop, J., Wood, T.J., Davis, R.E., Sherman, J.T., 2004. Robotic observations of enhanced carbon biomass and export at 55°S during SOFeX. *Science* 304, 417–420, <http://dx.doi.org/10.1126/science.1087717>.
- Blain, S., Quéguiner, B., Armand, L., Belviso, S., Bombled, B., Bopp, L., Bowie, A., Brunet, C., et al., 2007. Effect of natural iron fertilization on carbon sequestration in the Southern Ocean. *Nature* 446, 1070–1074, <http://dx.doi.org/10.1038/nature05700>.
- Bowie, A.R., Maldonado, M.T., Frew, R.D., Croot, P.L., Achterberg, E.P., Mantoura, R.F.C., Worsfold, P.J., Law, C.S., et al., 2001. The fate of added iron during a mesoscale fertilisation experiment

- in the Southern Ocean. *Deep-Sea Res. Part II: Topical Stud. Oceanogr.* 48, 2703–2743.
- Boyd, P.W., 2002. The role of iron in the biogeochemistry of the Southern Ocean and equatorial Pacific: a comparison. *Deep-Sea Res. Part II: Topical Stud. Oceanogr.* 49, 1803–1821, [http://dx.doi.org/10.1016/S0967-0645\(02\)00013-9](http://dx.doi.org/10.1016/S0967-0645(02)00013-9).
- Boyd, P.W., 2008. Implications of large-scale iron fertilization of the oceans – introduction and synthesis. *Mar. Ecol. Prog. Ser.* 364, 213–218, <http://dx.doi.org/10.3354/meps07541>.
- Boyd, P.W., Ellwood, M.J., 2010. The biogeochemical cycle of iron in the ocean. *Nat. Geosci.* 3, 675–682, <http://dx.doi.org/10.1038/ngeo964>.
- Boyd, P.W., Jickells, T., Law, C.S., Blain, S., Boyle, E.A., Buesseler, K.O., Coale, K.H., Cullen, J.J., et al., 2007. Mesoscale iron enrichment experiments 1993–2005: synthesis and future directions. *Science* 315, 612–617, <http://dx.doi.org/10.1126/science.1131669>.
- Boyd, P.W., Law, C.S., Wong, C.S., Nojiri, Y., Tsuda, A., Levasseur, M., Takeda, S., Rivkin, R., et al., 2004. The decline and fate of an iron-induced subarctic phytoplankton bloom. *Nature* 428, 549–553, <http://dx.doi.org/10.1038/nature02437>.
- Boyd, P.W., Trull, T.W., 2007. Understanding the export of biogenic particles in oceanic waters: is there consensus? *Prog. Oceanogr.* 72, 276–312.
- Boyd, P.W., Watson, A.J., Law, C.S., Abraham, E.R., Trull, T., Murdoch, R., Bakker, D.C.E., Bowie, A.R., et al., 2000. A mesoscale phytoplankton bloom in the polar Southern Ocean stimulated by iron fertilization. *Nature* 407, 695–702, <http://dx.doi.org/10.1038/35037500>.
- Breitbarth, E., Achterberg, E.P., Ardelan, M.V., Baker, A.R., Bucciarelli, E., Chever, F., Croot, P.L., Duggen, S., et al., 2010. Iron biogeochemistry across marine systems – progress from the past decade. *Biogeosciences* 7, 1075–1097, <http://dx.doi.org/10.5194/bg-7-1075-2010>.
- Brzezinski, M.A., Baines, S.B., Balch, W.M., Beucher, C.P.O., Chai, F., Dugdale, R.C., Landry, M.R., Marchi, A., et al., 2011. Co-limitation of diatoms by iron and silicic acid in the equatorial Pacific. *Deep-Sea Res. Part II: Topical Stud. Oceanogr.* 58, 493–511, <http://dx.doi.org/10.1016/j.dsr2.2010.08.005>.
- Buesseler, K.O., Andrews, J.E., Pike, S.M., Charette, M.A., 2004. The effects of iron fertilization on carbon sequestration in the Southern Ocean. *Science* 304, 414–417.
- Buesseler, K.O., Boyd, P.W., 2009. Shedding light on processes that control particle export and flux attenuation in the twilight zone of the upper ocean. *Limnol. Oceanogr.* 54, 1210–1232.
- Cao, L., Caldeira, K., 2010. Can ocean iron fertilization mitigate ocean acidification? *Clim. Change* 99, 303–311, <http://dx.doi.org/10.1007/s10584-010-9799-4>.
- CBD (Secretariat of Convention on Biological Diversity), 2009. Scientific Synthesis of the Impacts of Ocean Fertilization on Marine Biodiversity. CBD Technical Series No. 45, Montreal. 53pp.
- CBD (Secretariat of the Convention on Biological Diversity), 2012. Geoenvironment in Relation to the Convention on Biological Diversity: Technical and Regulatory Matters. CBD Technical Series No. 66, Montreal, 152pp.
- Cassar, N., DiFiore, P.J., Barnett, B.A., Bender, M.L., Bowie, A.R., Tilbrook, B., Petrou, K., Westwood, K.J., Wright, S.W., Lefevre, D., 2011. The influence of light and iron on net community production in the Subantarctic and Polar Frontal Zones. *Biogeosciences* 8, 227–237, <http://dx.doi.org/10.5194/bg-8-227-2011>.
- Chan, F., Barth, J.A., Lubchenko, J., Kirincich, A., Weeks, H., Peterson, W.T., Menge, B.A., 2008. Emergence of anoxia in the California current large marine ecosystem. *Science* 319, 920, <http://dx.doi.org/10.1126/science.1149016>.
- Cullen, J.J., Boyd, P.W., 2008. Predicting and verifying the intended and unintended consequences of large-scale ocean iron fertilization. *Mar. Ecol. Prog. Ser.* 364, 295–301, <http://dx.doi.org/10.3354/meps07551>.
- de Baar, H.J.W., Boyd, P.W., Coale, K.H., Landry, M.R., Tsuda, A., Assmy, P., Bakker, D.C.E., Bozec, Y., et al., 2005. Synthesis of iron fertilization experiments: from the iron age in the age of enlightenment. *J. Geophys. Res.* 110, <http://dx.doi.org/10.1029/2004JC002601>, C09S16.
- de Baar, H.J.W., Gerringa, L.J.A., Laan, P., Timmermans, K.R., 2008. Efficiency of carbon removal per added iron in ocean iron fertilization. *Mar. Ecol. Prog. Ser.* 364, 269–282, <http://dx.doi.org/10.3354/meps07548>.
- Denman, K.L., 2008. Climate change, ocean processes and ocean iron fertilization. *Mar. Ecol. Prog. Ser.* 364, 219–225, <http://dx.doi.org/10.3354/meps07542>.
- Dixon, J.L., 2008. Macro and micro nutrient limitation of microbial productivity in oligotrophic subtropical Atlantic waters. *Environ. Chem.* 5, 135–142, <http://dx.doi.org/10.1071/EN07081>.
- Duggen, S., Olgun, N., Croot, P., Hoffmann, L., Dietze, H., Delmelle, P., Teschner, C., 2010. The role of volcanic ash for the surface ocean biogeochemical iron-cycle: a review. *Biogeosciences* 7, 827–844, <http://dx.doi.org/10.5194/bg-7-827-2010>.
- Dutreuil, S., Bopp, L., Tagliabue, A., 2009. Impact of enhanced vertical mixing on marine biogeochemistry; lessons for geo-engineering and natural variability. *Biogeosciences* 6, 901–912, <http://dx.doi.org/10.5194/bg-6-901-2009>.
- Elliott, K., 2010. Geoenvironment and the precautionary principle. *Int. J. Appl. Philos.* 24, 237–253.
- Freestone, D., Rafuse, R., 2008. Ocean iron fertilization and international law. *Mar. Ecol. Prog. Ser.* 364, 227–233, <http://dx.doi.org/10.3354/meps07543>.
- Fuhrman, J.A., Capone, D.G., 1991. Possible biogeochemical consequences of ocean fertilization. *Limnol. Oceanogr.* 36, 1951–1959.
- Gabric, A.J., Cropp, R.A., McTainsh, G.H., Johnston, B.M., Butler, H., Tilbrook, B., Heywood, M., 2010. Australian dust storms in 2002–2003 and their impact on Southern Ocean biogeochemistry. *Global Biogeochem. Cycles* 24, <http://dx.doi.org/10.1029/2009GB0034541>, GB2005.
- Galaz, V., 2012. Geo-engineering, governance and socio-ecological systems: critical issues and joint research needs. *Ecol. Soc.* 17, 24, <http://dx.doi.org/10.5751/ES-04677-170124>.
- Gardiner, S.M., 2011. Some early ethics of geoenvironment the climate: a commentary on the values of the Royal Society report. *Environ. Values* 20, 163–188.
- Glibert, P.M., Azanza, R., Burford, M., Furuya, K., Abal, E., Al-Azri, A., Al-Yamani, F., Andersen, P., et al., 2008. Ocean urea fertilization for carbon credits poses high ecological risks. *Mar. Pollut. Bull.* 56, 1049–1056, <http://dx.doi.org/10.1016/j.marpolbul.2008.03.010>.
- Gnanadesikan, A., Sarmiento, J.L., Slater, R.D., 2003. Effects of patchy ocean fertilization on atmospheric carbon dioxide and biological production. *Global Biogeochem. Cycles* 17, 1050, <http://dx.doi.org/10.1029/2002GB001940>.
- Gnanadesikan, A., Marinov, I., 2008. Export is not enough: nutrient cycling and carbon sequestration. *Mar. Ecol. Prog. Ser.* 364, 289–294, <http://dx.doi.org/10.3354/meps07550>.
- Gordon, R.M., Johnson, K.S., Coale, K.H., 1998. The behaviour of iron and other trace elements during the IronEx-I and PlumEx experiments in the Equatorial Pacific. *Deep-Sea Res. Part II: Topical Stud. Oceanogr.* 45, 995–1041.
- Hale, B., Dilling, L., 2011. Geoenvironment, ocean fertilization and the problem of permissible pollution. *Sci. Technol. Human Values* 36, 190–212, <http://dx.doi.org/10.1177/0162243910366150>.
- Hale, M.S., Rivkin, R.B., Matthews, P., Agawin, N.S.R., Li, W.K.W., 2006. Microbial response to a mesoscale iron enrichment in the NE subarctic Pacific: heterotrophic bacterial processes. *Deep-Sea Res. Part II: Topical Stud. Oceanogr.* 53, 2231–2247, <http://dx.doi.org/10.1016/j.dsr2.2006.05.039>.
- Hamme, R.C., et al., 2010. Volcanic ash fuels anomalous plankton bloom in subarctic northeast Pacific. *Geophys. Res. Lett.* 37, <http://dx.doi.org/10.1029/2010GL044629>, L19604.
- Hare, S., Mantua, N.J., 2000. Empirical evidence for North Pacific regime shifts in 1977 and 1989. *Prog. Oceanogr.* 47, 103–145, [http://dx.doi.org/10.1016/S0079-6611\(00\)00033-1](http://dx.doi.org/10.1016/S0079-6611(00)00033-1).

- Harvey, M.J., Law, C.S., Smith, M.J., Hall, J.A., Abraham, E.R., Stevens, C.L., Hadfield, M.G., Ho, D.T., et al., 2011. The SOLAS air–sea gas exchange experiment (SAGE) 2004. *Deep-Sea Res. Part II: Topical Stud. Oceanogr.* 58, 753–763, <http://dx.doi.org/10.1016/j.dsr2.2010.10.015>.
- Hashimoto, S., Toda, S., Suzuki, K., Kato, S., Narita, Y., Kurihara, M.K., Akatsuka, Y., Oda, H., et al., 2009. Production and air–sea flux of halomethanes in the western subarctic Pacific in relation to phytoplankton pigment concentrations during the iron fertilization experiment (SEEDS II). *Deep-Sea Res. Part II: Topical Stud. Oceanogr.* 56, 2928–2935, <http://dx.doi.org/10.1016/j.dsr2.2009.07.003>.
- Ianson, D., Völker, C., Denman, K., Kunze, K., Steiner, N., 2012. The effect of vertical and horizontal dilution on fertilized patch experiments. *Global Biogeochem. Cycles* 26, <http://dx.doi.org/10.1029/2010GB004008>, GB3002.
- Jin, X., Gruber, N., 2003. Offsetting the radiative benefit of ocean iron fertilization by enhancing N₂O emissions. *Geophys. Res. Lett.* 30, 2249–2252, <http://dx.doi.org/10.1029/2003GL018458>.
- Jin, X., Gruber, N., Frenzel, H., Doney, S.C., McWilliams, J.C., 2008. The impact on atmospheric CO₂ of iron fertilization induced changes in the ocean's biological pump. *Biogeosciences* 5, 385–406.
- Jones, I.S.F., 2011. Contrasting micro- and macro-nutrient nourishment of the ocean. *Mar. Ecol. Progr. Ser.* 425, 281–296, <http://dx.doi.org/10.3354/meps08882>.
- Karl, D.M., Letelier, R.M., 2008. Nitrogen fixation-enhanced carbon sequestration in low nitrate, low chlorophyll seascapes. *Mar. Ecol. Progr. Ser.* 364, 257–268, <http://dx.doi.org/10.3354/meps07547>.
- Kato, S., Watiri, M., Nagao, I., Uematsu, M., Kajii, Y., 2009. Atmospheric trace gas measurements during SEEDS-II over the northwestern Pacific. *Deep-Sea Res. Part II: Topical Stud. Oceanogr.* 56, 2918–2927, <http://dx.doi.org/10.1016/j.dsr.2009.07.002>.
- Kondo, Y., Takeda, S., Nishioka, J., Obata, H., Furuya, K., Johnson, W.K., Wong, C.S., 2008. Organic iron (III) complexing ligands during an iron enrichment experiment in the western subarctic North Pacific. *Geophys. Res. Lett.* 35, <http://dx.doi.org/10.1029/2008GL033354>, L12601.
- Krom, M.D., Thingstad, T.F., Brenner, S., Carbo, P., Drakopoulos, P., Fileman, T.W., Flaten, G.A.F., Groom, S., et al., 2005. Summary and overview of the CYCLOPS P addition Lagrangian experiment in the Eastern Mediterranean. *Deep-Sea Res. Part II: Topical Stud. Oceanogr.* 52, 3090–3108, <http://dx.doi.org/10.1016/j.dsr.2005.08.018>.
- Kudo, I., Noiri, Y., Cochlan, W.P., Suzuki, K., Aramaki, T., Ono, T., Nojiri, Y., 2009. Primary productivity, bacterial productivity and nitrogen uptake in response to iron enrichment during the SEEDS II. *Deep-Sea Res. Part II: Topical Stud. Oceanogr.* 56, 2755–2766, <http://dx.doi.org/10.1016/j.dsr2.2009.06.003>.
- Kudo, I., Noiri, Y., Nishioka, J., Taira, Y., Kiyosawa, H., Tsuda, A., 2006. Phytoplankton community response to Fe and temperature gradients in the NE (SERIES) and NW (SEEDS) subarctic Pacific Ocean. *Deep-Sea Res. Part II: Topical Stud. Oceanogr.* 53, 2201–2213, <http://dx.doi.org/10.1016/j.dsr2.2006.05.033>.
- Lambert, K.K., 2011. Biophysical Geoengineering Compositions and Methods. Issued patent US8033879. (Application number 12/463,409, U.S. Patent Application Publication, number US 2009/0227161 A1).
- Lampitt, R.S., Achterberg, E.P., Anderson, T.R., Hughes, J.A., Iglesias-Rodriguez, M.D., Kelly-Gerrey, B.A., Lucas, M., Popova, E.E., et al., 2008. Ocean fertilization: a potential means of geoengineering? *Phil. Trans. R. Soc.* 366, 3919–3945, <http://dx.doi.org/10.1098/rsta.2008.0139>.
- Law, C.S., 2008. Predicting and monitoring the effects of large-scale ocean iron fertilization on marine trace gas emissions. *Mar. Ecol. Progr. Ser.* 364, 283–288, <http://dx.doi.org/10.3354/meps07549>.
- Law, C.S., Ling, R.D., 2001. Nitrous oxide flux and response to increased iron availability in the Antarctic circumpolar current. *Deep-Sea Res. Part II: Topical Stud. Oceanogr.* 48, 2509–2527.
- Law, C.S., Smith, M., Stevens, C., Abraham, E.R., Ellwood, M., Hill, P., Nodder, S., Peloquin, J., et al., 2011. Did dilution limit the phytoplankton response to iron addition in HNLC sub-Antarctic waters during SAGE? *Deep-Sea Res. Part II: Topical Stud. Oceanogr.* 58, 786–799, <http://dx.doi.org/10.1016/j.dsr2.2010.10.018>.
- Leinen, M., 2008. Building relationships between scientists and business in ocean iron fertilization. *Mar. Ecol. Progr. Ser.* 364, 251–256, <http://dx.doi.org/10.3354/meps07546>.
- Lenton, T., Vaughan, N., 2009. The radiative forcing potential of different climate geoengineering options. *Atmos. Chem. Phys.* 9, 5539–5561, <http://dx.doi.org/10.5194/acp-9-5539-2009>.
- Levasseur, M., Scarratt, M.G., Michaud, S., Merzouk, A., Wong, C.S., Arychuk, M., Richardson, W., Rivkin, R.B., et al., 2006. DMSP and DMS dynamics during a mesoscale iron fertilization experiment in the Northeast Pacific – Part 1: temporal and vertical distributions. *Deep-Sea Res. Part II: Topical Stud. Oceanogr.* 53, 2353–2369, <http://dx.doi.org/10.1016/j.dsr2.2006.05.023>.
- Levin, L.A., Etter, R.J., Rex, M.A., Gooday, A.J., Smith, C.R., Pineda, J., Stuart, C.T., Hessler, R.R., Pawson, D., 2001. Environmental influences on regional deep-sea species diversity. *Ann. Rev. Ecol. Syst.* 32, 51–93.
- Liss, P., Church, A., Bakker, D., Turner, S., 2005. Ocean fertilization with iron: effects on climate and air quality. *Tellus Ser. B* 57, 269–271.
- Lovelock, J.E., Rapsley, C.G., 2007. Ocean pipes could help the Earth cure itself. *Nature* 449, 781, <http://dx.doi.org/10.1038/449403a>.
- Markels, M., Sato, T., Chen, L., Jones, I.S.F., 2011. Enhanced carbon storage in the ocean. Report of Engineering Committee for Oceanic Resources (ECOR), version 4.6. Online at www.oceanicresources.org/wp-content/uploads/2012/03/ECOR-WG-Report-4.6-clean.pdf
- Marchetti, A., Sherry, N.D., Kiyosawa, H., Tsuda, A., Harrison, P.J., 2006. Phytoplankton processes during a mesoscale iron enrichment in the NE subarctic Pacific. Part I – biomass and assemblage. *Deep-Sea Res. Part II: Topical Stud. Oceanogr.* 53, 2095–2113, <http://dx.doi.org/10.1016/j.dsr2.2006.05.038>.
- Martin, J.H., Coale, K.H., Johnson, K.S., Fitzwater, S.E., Gordon, R.M., Tanner, S.J., Hunter, C.N., Elrod, V.A., et al., 1994. Testing the iron hypothesis in ecosystems of the equatorial Pacific Ocean. *Nature* 371, 123–129, <http://dx.doi.org/10.1038/371123a0>.
- Martin, J.H., Fitzwater, S.E., Gordon, R.M., 1990. Iron deficiency limits phytoplankton growth in Antarctic waters. *Global Biogeochem. Cycles* 4, 5–12.
- Masuda, T., Furuya, K., Kohashi, N., Sato, M., Takeda, S., Uchiyama, M., Horimoto, N., Ishimaru, T., 2010. Lagrangian observation of phytoplankton dynamics at an artificially enriched subsurface water in Sagami Bay. *Jpn. J. Oceanogr.* 66, 801–813, <http://dx.doi.org/10.1007/s10872-010-0065-1>.
- Matear, R.J., Elliott, B., 2004. Enhancement of oceanic uptake of anthropogenic CO₂ by macronutrient fertilization. *J. Geophys. Res.* 109, doi:10.1029/2000JC000321, C04001.
- Maruyama, S., Yabuki, T., Sato, T., Tsubaki, K., Komiya, A., Watanabe, M., Kawamura, H., Tsukamoto, K., 2011. Evidences of increasing primary production in the ocean by Stommel's perpetual salt fountain. *Deep-Sea Res. Part I: Oceanogr. Res. Papers* 58, 567–574, <http://dx.doi.org/10.1016/j.dsr.2011.02.012>.
- Mylrold, N.P., Caldeira, K., 2012. Greenhouse gases, climate change and the transition from coal to low-carbon electricity. *Environ. Res. Lett.* 7, 014019, <http://dx.doi.org/10.1088/1748-9326/7/1/014019>.
- Nagao, I., Hashimoto, S., Suzuki, K., Toda, S., Narita, Y., Tsuda, A., Saito, H., Kudo, I., et al., 2009. *Deep-Sea Res. Part II: Topical Stud. Oceanogr.* 56, 2899–2917, <http://dx.doi.org/10.1016/j.dsr2.2009.07.001>.
- Naqvi, S.W.A., Bange, H.W., Farias, L., Monteiro, P.M.S., Scranton, M.I., Zhang, J., 2010. Marine hypoxia/anoxia as a source of CH₄ and N₂O. *Biogeosciences* 7, 2159–2190, <http://dx.doi.org/10.5194/bg-7-2159-2010>.

- Nishioka, J., Takeda, S., Kondo, Y., Obata, H., Doi, T., Tsumune, D., Wong, C.S., Johnson, W.K., et al., 2009. Changes in iron concentrations and bio-availability during an open mesoscale iron enrichment in the western subarctic Pacific, SEEDS II. *Deep-Sea Res. Part II: Topical Stud. Oceanogr.* 56, 2796–2809, <http://dx.doi.org/10.1016/j.dsr2.2009.06.006>.
- Orbach, M.K., 2008. Cultural context of ocean fertilization. *Mar. Ecol. Progr. Ser.* 364, 235–242, <http://dx.doi.org/10.3354/meps07544>.
- Oschlies, A., Koeve, W., Rickels, W., Rehdanz, K., 2010a. Side effects and accounting aspects of hypothetical large-scale Southern Ocean iron fertilization. *Biogeosciences* 7, 4017–4035, <http://dx.doi.org/10.5194/bg-7-4017-2010>.
- Oschlies, A., Pahlow, M., Yool, A., Matear, R.M., 2010b. Climate engineering by artificial ocean upwelling: channelling the sorcerer's apprentice. *Geophys. Res. Lett.* 37, <http://dx.doi.org/10.1029/2009GL041961>, L04701.
- Pan, A., Pourziaei, B., Huang, H.X., 2011. Effect of ocean iron fertilization on the phytoplankton biological carbon pump. *Adv. Appl. Maths. Mech.* 3, 52–64, <http://dx.doi.org/10.4208/aamm.10-m1023>.
- Peloquin, J., Hall, J., Safi, K., Ellwood, M., Law, C.S., Thompson, K., Kuparinen, J., Harvey, M., et al., 2011. Control of the phytoplankton response during the SAGE experiment: a synthesis. *Deep-Sea Res. Part II: Topical Stud. Oceanogr.* 58, 824–838, <http://dx.doi.org/10.1016/j.dsr2.2010.10.019>.
- Pollard, R.T., Salter, I., Sanders, R.J., Lucas, M.I., Moore, C.M., Mills, R.A., Statham, P.J., Allen, J.T., et al., 2009. Southern Ocean deep-water carbon export enhanced by natural iron fertilization. *Nature* 457, 577–580, <http://dx.doi.org/10.1038/nature07716>.
- Powell, H., 2008. Dumping iron and trading carbon. *Oceanus* 46 (1), 22–25.
- Quinn, P.K., Bates, T.S., 2011. The case against climate regulation via oceanic phytoplankton sulphur emissions. *Nature* 480, 51–56, <http://dx.doi.org/10.1038/nature10580>.
- Psarra, S., Zohary, T., Krom, M.D., Mantoura, R.F.C., Polychronaki, T., Stambler, N., Tanaka, T., Tselepidis, A., et al., 2005. Phytoplankton response to a Lagrangian phosphate addition in the Levantine Sea (Eastern Mediterranean). *Deep-Sea Res. Part II: Topical Stud. Oceanogr.* 52, 2944–2960, <http://dx.doi.org/10.1016/j.dsr2.2005.08.015>.
- Rees, A.P., Law, C.S., Woodward, E.M.S., 2006. High rates of N₂ fixation during an *in-situ* phosphate release experiment in the eastern Mediterranean Sea. *Geophys. Res. Lett.* 33, <http://dx.doi.org/10.1029/2006GL025791>, L10607.
- Rickels, W., Rehdanz, K., Oschlies, A., 2010. Methods for greenhouse gas offset accounting: a case study of ocean iron fertilization. *Ecol. Econ.* 69, 2495–2509, <http://dx.doi.org/10.1016/j.ecolecon.2010.07.026>.
- Royal Society, 2009. *Geoengineering the Climate; Science, Governance and Uncertainty*. RS Policy Document 10/09, London, 82 pp.
- Rue, E.L., Bruland, K.W., 1997. The role of organic complexation on ambient iron chemistry in the equatorial Pacific Ocean and the response of a mesoscale iron addition experiment. *Limnol. Oceanogr.* 42, 901–910.
- Saito, H., Ota, T., Suzuki, K., Nishioka, J., Tsuda, A., 2006. Role of heterotrophic dinoflagellate *Gyrodinium* sp. in the fate of an iron induced bloom. *Geophys. Res. Lett.* 33, <http://dx.doi.org/10.1029/2005GL025366>, L09602.
- Sarmiento, J.L., Orr, J.C., 1991. 3-dimensional simulations of the impact of Southern-Ocean nutrient depletion on atmospheric CO₂ and ocean chemistry. *Limnol. Oceanogr.* 36, 1928–1950.
- Sastri, A.R., Dower, J.F., 2006. Mesozooplankton community response during the SERIES iron enrichment experiment in the subarctic Pacific. *Deep-Sea Res. Part II: Topical Stud. Oceanogr.* 53, 2268–2280, <http://dx.doi.org/10.1016/j.dsr2.2006.05.034>.
- Shaw, T.J., Raiswell, R., Hexel, C.R., Vu, H.P., Moore, W.S., Dudgeon, R., Smith, K.L., 2011. Input, composition, and potential impact of terrigenous material from free-drifting icebergs in the Weddell Sea. *Deep-Sea Res. Part II: Topical Stud. Oceanogr.* 58, 1376–1383, <http://dx.doi.org/10.1016/j.dsr2.2010.11.012>.
- Shepherd, J., Iglesias-Rodriguez, D., Yool, A., 2007. Geo-engineering might cause, not cure, problem. *Nature* 449, 781, <http://dx.doi.org/10.1038/449781a>.
- Sigman, D.M., Boyle, E.A., 2000. Glacial/interglacial variations in atmospheric carbon dioxide. *Nature* 407, 859–869.
- Silver, M.W., Bargo, S., Coale, S.L., Benitez-Nelson, C.R., Garcia, A.C., Roberts, K.J., Sekula-Wood, E., Bruland, K.W., et al., 2010. Toxic diatoms and domoic acid in natural and iron enriched waters of the oceanic Pacific. *Proc. Nat. Acad. Sci. U. S. A.* 107, 20762–20767, <http://dx.doi.org/10.1073/pnas.1006968107>.
- Smetacek, V., Naqvi, S.W.A., 2008. The next generation of iron fertilization experiments in the Southern Ocean. *Phil. Trans. Roy. Soc. A* 366, 3947–3967, <http://dx.doi.org/10.1098/rsta.2008.0144>.
- Smetacek, V., 2012. Deep carbon export from a Southern Ocean iron-fertilized diatom bloom. *Nature* 487, 313–319, <http://dx.doi.org/10.1038/nature11229>.
- Strong, A.L., Cullen, J.J., Chisholm, S.W., 2009. Ocean fertilization. *Science, policy and commerce. Oceanogr.* 22, 236–261.
- Sunda, W.G., Swift, D.G., Huntsman, S.A., 1991. Low iron requirement for growth in oceanic phytoplankton. *Nature* 351, 55–57, <http://dx.doi.org/10.1038/351055a0>.
- Takeda, S., Tsuda, A., 2005. An *in situ* iron-enrichment experiment in the western subarctic Pacific (SEEDS): introduction and summary. *Progr. Oceanogr.* 64, 95–109, <http://dx.doi.org/10.1016/j.pocean.2005.02.004>.
- Thingstad, T.F., Krom, M.D., Mantoura, R.F.C., Flaten, G.F., Groom, S., Herut, B., Kress, N., Law, C.S., et al., 2005. Nature of phosphorus limitation in the ultra-oligotrophic Eastern Mediterranean. *Science* 309, 1068–1071, <http://dx.doi.org/10.1126/science.1112632>.
- Trick, C.G., Bill, B.D., Cochlan, W.P., Wells, M.L., Trainer, V.L., Pickell, L.D., 2010. Iron enrichment stimulates toxic diatom production in high-nitrate, low-chlorophyll areas. *Proc. Nat. Acad. Sci. U. S. A.* 107, 5887–5892, <http://dx.doi.org/10.1073/pnas.0910579107>.
- Tsubaki, K., Maruyama, S., Komiya, A., Mitsugashira, H., 2007. Continuous measurement of an artificial upwelling of deep sea water induced by the perpetual salt fountain. *Deep-Sea Res. Part I: Oceanogr. Res. Papers* 54, 75–84, <http://dx.doi.org/10.1016/j.dsr.2006.10.002>.
- Tsuda, A., Kiyosawa, H., Kuwata, A., Mochizuki, M., Shiga, N., Saito, H., Chiba, S., Imai, K., et al., 2005. Responses of diatoms to iron-enrichment (SEEDS) in the western subarctic Pacific, temporal and spatial comparisons. *Progr. Oceanogr.* 64, 189–205, <http://dx.doi.org/10.1016/j.pocean.2005.02.008>.
- Tsuda, A., Saito, H., Machida, R.J., Shimode, S., 2009. Meso- and microzooplankton responses to an *in situ* iron fertilization experiment (SEEDS II) in the northwest subarctic Pacific. *Deep-Sea Res. Part II: Topical Stud. Oceanogr.* 56, 2767–2778, <http://dx.doi.org/10.1016/j.dsr2.2009.06.004>.
- Tsuda, A., Saito, H., Nishioka, J., Ono, T., Noiri, Y., Kudo, I., 2006. Mesozooplankton response to iron enrichment during the diatom bloom and bloom decline in SERIES (NE Pacific). *Deep-Sea Res. Part II: Topical Stud. Oceanogr.* 53, 2281–2296, <http://dx.doi.org/10.1016/j.dsr2.2006.05.041>.
- Tsuda, A., Takeda, S., Saito, H., Nishioka, J., Nojiri, Y., Kudo, I., Kiyosawa, H., Shiimoto, A., et al., 2003. A mesoscale iron enrichment in the western Subarctic Pacific induces a large centric diatom bloom. *Science* 300, 958–961, <http://dx.doi.org/10.1126/science.1082000>.
- Turner, S.M., Harvey, M.J., Law, C.S., Nightingale, P.D., Liss, P.S., 2004. Iron-induced changes in oceanic sulfur biogeochemistry. *Geophys. Res. Lett.* 31, <http://dx.doi.org/10.1029/2004GL020296>.
- Turner, S.M., Nightingale, P.D., Spokes, L.J., Liddicoat, M.I., Liss, P.S., 1996. Increased dimethyl sulphide concentrations in sea water from *in situ* iron enrichment. *Nature* 383, 513–517, <http://dx.doi.org/10.1038/383513a0>.
- Wallace, D.W.R., Law, C.S., Boyd, P.W., Collos, Y., Croot, P., Denman, K., Lam, P.J., Riebesell, U., Takeda, S., Williamson, P.,

2010. Ocean Fertilization: a Scientific Summary for Policy Makers. IOC/UNESCO, Paris (IOC/BRO/2010/2), 18 pp.
- Walter, S., Peeken, I., Lochte, K., Webb, A., Bange, H.W., 2005. Nitrous oxide measurements during EIFEX, the European Iron Fertilization Experiment in the subpolar South Atlantic Ocean. *Geophys. Res. Lett.* 32, <http://dx.doi.org/10.1029/2005GL024619>, L23613.
- Watson, A.J., Law, C.S., van Scoy, K.A., Millero, F.J., Yao, W., Friederich, G.E., Liddicoat, M.I., Wanninkhof, R.H., et al., 1994. Minimal effect of iron fertilization on sea-surface carbon dioxide concentrations. *Nature* 371, 143–145, <http://dx.doi.org/10.1038/371143a0>.
- Watson, A.J., Boyd, P.W., Turner, S.M., Jickells, T.D., Liss, P.S., 2008. Designing the next generation of ocean iron fertilization experiments. *Mar. Ecol. Progr. Ser.* 364, 303–309, <http://dx.doi.org/10.3354/meps07552>.
- White, A., Bjorkman, K., Grabowski, E., Letelier, R., Poulos, S., Watkins, B., Karl, D., 2010. An open ocean trial of controlled upwelling using wave pump technology. *J. Atmos. Ocean. Technol.* 27, 385–396, <http://dx.doi.org/10.1175/2009JTECHO679.1>.
- Williamson, P., Turley, C., 2012. Ocean acidification in a geoengineering context. *Phil. Trans. Roy. Soc. A* 370, 4317–4342, <http://dx.doi.org/10.1098/rsta.2012.0167>.
- Wingenter, O.W., Haase, K.B., Strutton, P., Friederich, G., Meinardi, S., Blake, D.R., Rowland, F.S., 2004. Changing concentrations of CO, CH₄, C₂H₆, CH₃Br, CH₃I, and dimethyl sulfide during the southern ocean iron enrichment experiments. *Proc. Natl. Acad. Sci. U. S. A.* 101, 8537–8541, <http://dx.doi.org/10.1073/pnas.0402744101>.
- Wolff, G.A., Billett, D.S.M., Bett, B.J., Holtvoeth, J., FitzGeorge-Balfour, T., Fisher, E.H., Cross, I., Shannon, R., et al., 2011. The effects of natural iron fertilization on deep-sea ecology: the Crozet Plateau, Southern Indian Ocean. *PLoS One* 6, <http://dx.doi.org/10.1371/journal.pone.0020697>, e20697.
- Woodhouse, M.T., Carslaw, K.S., Mann, G.W., Vallina, S.M., Vogt, M., Halloran, P.R., Boucher, O., 2010. Low sensitivity of cloud condensation nuclei to changes in the sea–air flux of dimethyl-sulphide. *Atmos. Chem. Phys.* 10, 7545–7559, <http://dx.doi.org/10.5194/acp-10-7545-2010>.
- Yool, A., Shepherd, J.G., Bryden, H.L., Oschlies, A., 2009. Low efficiency of nutrient translocation for enhancing oceanic uptake of carbon dioxide. *J. Geophys. Res. – Oceans* 114, <http://dx.doi.org/10.1029/2008JC004792>, C08009.
- Zahariev, K., Christian, J., Denman, K., 2008. Preindustrial, historical and fertilization simulations using a global ocean carbon model with new parameterizations of iron limitation, calcification and N₂ fixation. *Progr. Oceanogr.* 77, 56–82.