



# Designing the next generation of ocean iron fertilization experiments

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**ABSTRACT:** The first generation of open-ocean iron enrichments (1993 to 2005) have all had broadly the same design. Enrichment of patches of ocean was typically on a 10 km length-scale, and experiments were of a duration of weeks. These scales were dictated by what could conveniently be achieved from research vessels, using tracers to track Lagrangian patches. The extrapolation of experimental findings to the larger scales required for carbon sequestration by ocean iron fertilization (OIF) leaves many uncertainties, to answer which, longer duration (i.e. months) and larger scale observations (100 to 200 km length-scale) are required. However, to extrapolate to a timescale of decades and to the scale of ocean basins, such observations must be conducted in parallel (and where possible assimilated into) detailed models of the physics and biogeochemistry of the fertilized waters. Our present understanding suggests that any carbon sequestration will occur as the net result of changes in the air–sea flux integrated over millions km<sup>2</sup> and many years, and can only realistically be assessed by modelling. A central role of the observational studies will be to make such models as accurate as possible in their simulations and predictions. We present a scheme for the design of a second generation of ocean iron-enrichments and discuss the challenges that are evident in linking the modelling and observational components of such studies.

**KEY WORDS:** Carbon · Sequestration · Climate · Mitigation · Southern Ocean · HNLC · Nutrient · Chlorophyll

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## UNANSWERED QUESTIONS FROM PREVIOUS RELEASES

Open-ocean iron addition experiments (FeAXs) have been performed in each of the high-nutrient, low-chlorophyll (HNLC) regions of the World Ocean—the equatorial Pacific Ocean, Southern Ocean and north Pacific Ocean—and have established the fundamental role of iron limitation in all these regions (de Baar et al. 2005, Boyd et al. 2007). These experiments have all followed a similar design (Watson et al. 1991), in which patches on the order of 10 km long are initially seeded with both inorganic iron and the inert tracer sulphur hexafluoride, and the chemical and biological consequences of enrichment are followed over an ensuing period of several weeks. The time scale and conse-

quently the spatial dimension of the experiments have been set by very practical concerns and logistical constraints; a campaign from even the largest research vessels can rarely last more than 2 mo. The endurance of the ships and the scale of funding available (not to mention the reluctance of scientists and seagoers to spend many months away from home) set this time scale. In turn, this means that patches reach only a few tens of km in length-scale before the studies end. Using 2 or more research cruises, recent experiments have increased the duration somewhat (e.g. Coale et al. 2004), but exercises such as the Southern Ocean Iron Experiments (SOFeX) and the European Iron Fertilization Experiment (EIFEX) (Hoffmann et al. 2006) are at the upper limit of what can be achieved by conventional oceanography.

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Though much has been learned from these experiments about the effects of iron addition at the ecosystem level, if we are interested in the effects of deliberate ocean iron fertilization (OIF) as a method for sequestration of carbon, or for that matter, in the climatic effects of increased iron availability on glacial–interglacial time scales, then we need to upscale our results to longer times and larger space scales. Difficulties quickly become apparent when this is attempted. Recent reviews (de Baar et al. 2005, Boyd et al. 2007) have summarized where our knowledge is still inadequate for this extrapolation and the major remaining questions are summarized in Table 1.

### HOW IS CO<sub>2</sub> SEQUESTERED FROM THE ATMOSPHERE BY OIF?

In order to understand the likely influences on the efficiency of sequestration, it is helpful to describe the carbon balance of a parcel of water that upwells into the surface layer of the ocean. Suppose we take some typical deep ocean water and bring it to the surface in an HNLC region—consider first the polar Southern Ocean or sub-antarctic. In the absence of iron fertilization, biological uptake of nutrients and carbon takes place, fixing carbon and macronutrients to the extent allowed by the iron (and light) availability. After some

Table 1. Major uncertainties in the effects of deliberate ocean iron fertilization

Process	Issue	Finding	Comments
Carbon sequestration efficiency	Carbon fixation in surface water	Variable <sup>a</sup>	Latitude, mixed layer depths + light co-limitation
	Carbon export	None/little/significant <sup>b</sup>	Limited duration of studies
	Depth of carbon export	Poorly constrained	
	Duration of carbon sequestration	Unknown	Unanswerable by observations alone
	Fraction fixed from atmosphere	Poorly known	Likely unanswerable by observations alone
Influence on dissolved oxygen	Formation of subsurface O <sub>2</sub> minima	Poorly known <sup>c</sup>	Potentially harmful, depth dependent
Production of other climate-active gases	Methane & nitrous oxide	No effect/possible enhancement <sup>d,e,f</sup>	Significant warming potential
	Dimethylsulphide	No change/ increase <sup>b</sup>	Some evidence that enhancement is transient
	Biogenic halocarbons	Reduction/no change/ increase <sup>g</sup>	Pertinent to atmospheric oxidation chemistry & particle formation
	Biogenic hydrocarbons, including alkyl nitrates	No change/increase <sup>e,g</sup>	Pertinent to atmospheric oxidation chemistry & particle formation
Effects on ecosystems and biogeochemistry	Phytoplankton species shifts	Mainly towards diatoms <sup>b</sup>	Are shifts transient?
	Mesozooplankton stocks	No change/increase <sup>b</sup>	Localised increases within Fe patch due to arrested vertical migration: duration of study & longer reproductive cycles
	Higher trophic levels	Unknown	Limited duration of studies. Possibility of enhanced secondary and higher-level production <sup>j</sup>
	Macronutrient uptake	Small to significant <sup>b</sup>	Mixed layer depths + light co-limitation
	Reduction of nutrient transport	Important in upwelling regions <sup>h</sup>	Supply flows to other areas cut off, e.g. sub-tropical gyres
	Nutrient remineralization	May affect global distributions	At present only evident from modelling studies <sup>i</sup>

<sup>a</sup>de Baar et al. (2005); <sup>b</sup>Boyd et al. (2007); <sup>c</sup>Natural O<sub>2</sub> minima: e.g. Arabian Sea & east subtropical Pacific, anthropogenic minima: e.g. Gulf of Mexico eutrophication; <sup>d</sup>Law & Ling (2001); <sup>e</sup>Wingenter et al. (2004); <sup>f</sup>Walter et al. (2005); <sup>g</sup>Liss et al. (2005); <sup>h</sup>Cooper et al. (1996); <sup>i</sup>Gnanadesikan et al. (2003); <sup>j</sup>Tsuda et al. 2006

period at the surface, which is typically of the order of 1 or 2 yr and probably depends very much on proximity to frontal regions, the water is subducted or transported by deep winter mixing out of the surface layer. As relatively cold, dense water, it can mix into the deep or intermediate waters and may not surface again for 100s of yr. If we now imagine that this water is fertilized with iron while it is at the surface and that its  $p\text{CO}_2$  decreases as the iron-stimulated bloom develops, it will begin to take up atmospheric  $\text{CO}_2$ , or to decrease the outgassing flux if the region is a net source of  $\text{CO}_2$  to the atmosphere. When this water is subducted, any  $\text{CO}_2$  it has removed from the atmosphere, or that it retains when, without fertilization, it would have been lost to the atmosphere, will be transported into the ocean's interior ocean and sequestered until that water is again brought to the surface.

It is notable that in this description the sequestration of carbon from the atmosphere is not dependent on having an export flux to any particular depth. Provided the biologically fixed carbon is not quickly respired back to  $\text{CO}_2$  while the water is still at the surface, sequestration could occur even if there were no export flux. The export flux is only important in that, since remineralization of the phytoplankton carbon is often quite fast (days, Boyd et al. 2004), export ensures that a proportion of this remineralization would occur at depths where the water is out of contact with the atmosphere. The important point is that the fertilized water has a limited residence time at the surface before subduction and that during this time fertilization induces enhanced lowering of its  $p\text{CO}_2$ . Efficient sequestration will be dependent on this time scale. In particular, if the cumulative natural iron supply to this water (from the atmosphere, for example) is sufficient to cause the carbon fixation to occur in any case during the time it is at the surface, the deliberate iron fertilization is largely redundant; its only effect is to speed up by 1 or 2 yr a sequestration that would have occurred even without the artificial fertilization, focusing enhanced productivity into a bloom at the expense of reduced production in the waters downstream. The condition for efficient sequestration is therefore that the residence time at the surface,  $\tau_r$ , must satisfy:

$$\tau_e < \tau_r < \tau_{\text{Fe}} \quad (1)$$

where  $\tau_e$  is the air–sea equilibration time for  $\text{CO}_2$ , which is typically 1 yr (Broecker & Peng 1982) and  $\tau_{\text{Fe}}$  is the time that would be required for the natural iron flux to supply iron equivalent to that needed for the fertilization. This second time scale depends on poorly known variables, such as the C:Fe ratio assumed necessary for open ocean phytoplankton and (if the iron source is atmospheric dust) the solubility of the iron in aerosol. As an example, using figures from Jickells et

al. (2005) for the iron flux to the Southern Ocean, a solubility of iron in dust of 3% (Baker & Jickells 2006), a mixed layer depth of 50 m and a molar C:Fe ratio of  $10^5$ , we calculate a  $\tau_{\text{Fe}}$  of about 10 yr.

This discussion suggests that sequestration efficiency due to iron fertilization may be less in the equatorial Pacific, as models tend to show (e.g. Sarmiento & Orr 1991, Cooper et al. 1996). Water that upwells into the surface layer in this region is strongly heated and therefore much less dense than most subsurface water, and it is unlikely to be permanently subducted below the thermocline until it has been transported to cooler regions, requiring decadal or longer residence time at or near the surface. This residence time will usually exceed  $\tau_{\text{Fe}}$  (which in most regions is shorter than that calculated for the Southern Ocean, since the atmospheric iron flux is at its lowest in polar waters). The net effect of OIF in such warm water regions would then be simply to replace the slow uptake of macronutrients and carbon that would normally take place over a period of years after the water has upwelled with an artificially induced bloom. Alternatively, recent models suggest some subduction to comparatively shallow depths may occur from the equatorial Pacific (F. Chai pers. comm.). In that case, fertilization here would provide some sequestration, though of a less permanent nature than in the sub-antarctic. In the sub-arctic Pacific some intermediate water (North Pacific Intermediate Water) is formed and efficient sequestration might be possible if waters feeding into this water mass were fertilized.

This description should make clear why realistic physical-biogeochemical modelling of a given OIF event must be an intrinsic part of any strategy to investigate the sequestration of carbon. The C sequestered cannot be measured by any simple means. It is certainly not equal to the export flux deriving from a bloom triggered by the fertilization, for example. Rather, it is the difference between the net air–sea flux of carbon dioxide due to the iron fertilization and that which would have occurred in its absence. Calculation of this flux involves integration over periods of years and an area of ocean that rapidly grows to cover millions of  $\text{km}^2$ . Clearly this can only be determined by modelling. The problem is that the model must correctly simulate the physics, chemistry and biology, and their interplay in such HNLC open-ocean provinces. But our models are not currently this good. The best we can do is likely to be to guide, improve and make more accurate the models by well-chosen measurements, not only of inorganic carbon but of all the relevant variables that we can measure. Thus, we see the program as being a joint effort of modellers and field experimentalists, with the observational program improving the models by validating any variable that

can be both modelled and measured. Observations alone cannot determine the carbon sequestration and models should not be believed unless they consistently reproduce observations.

### NEED FOR A JOINT MODELLING-OBSERVATIONAL STRATEGY

It is clear that to reduce the uncertainties about the effects of iron fertilization, observations on scales larger than used those up to now are needed, and we assume here that the initial fertilization in Generation 2 experiments would be on the order of  $200 \times 200$  km. However, as discussed in the previous section, it is evident that observations alone are insufficient. Rather, we need a joint modelling-observational strategy, because there are several crucial questions that direct observation alone cannot answer, as follows: (1) The outcome of the release, including its eventual efficiency at removing carbon from the atmosphere, will be strongly dependent on the initial physical, chemical and biological conditions at the potential site. These will vary with location and also with factors such as mixed-layer depth, proximity to oceanic fronts and degree of eddy activity. Assessment of all these will require models resolving eddies and fronts (typically the model resolution should be less than 10 km). (2) The assessment of the lateral trajectory of the iron patch also requires assimilating remote sensing and *in situ* observations into eddy resolving models, which will constrain the initial conditions of physical, biological and chemical state, as well as keep the modelled physics realistic. (3) Assessment of these outcomes will require integration up to scales that are too large and long for any ship-based observation programme. (4) Finally, this assessment requires evaluation of the 'control' case — what would have happened in the absence of a release — which can be provided by observations of a site within the surrounding HNLC waters for small experiments, but only by a model for larger experiments.

Model studies would need therefore to be carried out before, during and after a given FeAX. Adequately integrating such detailed model simulations with an observational program is likely to be as challenging as the practical logistics of the experiment at sea.

### SITE SELECTION

In the first generation of experiments, the criteria for site selection were appropriate biogeochemical conditions (HNLC, low iron, iron-limited phytoplankton, seasonal mean mixed layer depth) and relatively quiescent physical conditions (to permit a coherent

labelled patch of ocean to persist). For larger experiments that rely less on tracking a tracer, this basis for site selection will no longer be so relevant. Modelling should be used to help select the site, considering not just the large-scale dynamics (e.g. what part of the world ocean to do the release in) but also the mesoscale: proximity to fronts, eddy scales and kinetic energy. For this purpose, high resolution models and observations (from satellite altimetry, for example) would be useful and could help determine the best strategy for the iron release. The 'confined patch' strategy used up until now may well not be the best way to begin a larger and longer-scale experiment (see later), which might be better initiated by an elongated streak.

### DESIGN OF THE OBSERVATIONAL PHASE OF THE EXPERIMENT

Following the selection of a suitable site based on an ensemble of model simulations, the next step is to design a comprehensive survey of the variability exhibited by the properties that may be altered by the iron release. This survey should cover both the waters upstream (i.e. into which the iron will eventually be released) and downstream (i.e. the waters that will interact with the labelled iron patch as it evolves). The variables to be measured would include biogenic gases, downward export flux, biological productivity and nutrients. The areal extent of this survey will be dictated by the expected final areal extent of the iron release. Modelling will also be essential to provide some constraints on the probable trajectory (e.g. Coale et al. 1996) and evolution (dilution rate) of the iron-labelled patch over the subsequent 6 to 12 mo.

During the first generation of experiments, around 12 to 14 h was required to add the dissolved iron so that it formed a coherent patch of 10 km length-scale. However, logistics dictate that for iron enrichment of a  $200 \times 200$  km patch, multiple vessels would be required if a coherent enrichment patch is to be accomplished within a few days. Such a challenge would require making the iron addition in a carefully co-ordinated manner, which would involve monitoring a suite of Lagrangian (i.e. moving with the net flow of the upper ocean currents, etc.) drifters both in surface and subsurface waters (the latter ensuring the water at depth below the patch is moving in concert with the iron-enriched surface layer). A possible alternative to iron addition using multiple ships might be to use aircraft to spread the iron, though this would inevitably mean the iron would be added at the very surface, rather than homogenized into the mixed layer. We also foresee some operational difficulties in spreading  $>10$  t of material from the air in remote regions of the ocean.

Previous experiments have used the conservative chemical tracer sulphur hexafluoride as a proxy for the concurrently added iron (Watson et al. 1991); however, the use of such a tracer, which is a powerful approach, is impractical on larger spatial scales because the lifetime of the tracer in the surface water is too short and mapping a large patch would take too much ship time. It might be possible to label the centre of this 200 km long patch (and if multiple vessels were adding the iron, this might be very useful), but mapping a patch this large as it evolves would require other approaches including aircraft or helicopters with bio-optical sensors to monitor chlorophyll fluorescence or CO<sub>2</sub> (Boyd et al. 2007). As the patch evolved onto larger scales, satellite remote-sensing could be used to assess its areal extent, as was done on some of the first generation iron enrichments at scales of 500 to 1000 km<sup>2</sup> (Boyd et al. 2007).

The first set of iron enrichments made very wide-ranging ancillary measurements over and above the core physical, chemical and biological approaches. Generation 2 experiments will no doubt do likewise, but discussion of the full suite of measurements is beyond the scope of this short commentary. In Table 2 we present examples of approaches that provide data streams of different resolution, from remotely sensed to discrete measurements. As data assimilation into models is advocated as part of this multi-stranded experimental design, resolving how best to assimilate a range of datasets with different characteristics (frequency of observations, data quality) requires considerable thought and development, as do issues of

free parameter space versus computation needs for conducting eddy resolving physical-biogeochemical model simulations.

## EVOLUTION OF THE IRON-STIMULATED BLOOM

Previous experience during earlier FeAXs suggests that monitoring the latter stages of the bloom (i.e. after it terminates due to resource limitation) and the fate of the algal carbon are problematic. The main issue here is that much of the export, transformation and recycling of this algal carbon takes place in the waters underlying the surface layer that was initially iron-enriched. Hence the need for Lagrangian drifters in the surface and subsurface layers and some means to ensure that there is no lateral slippage between these layers, as is sometimes observed (Savidge et al. 1992), that would uncouple the processes occurring in each layer. Other important issues include defining the depth of carbon sequestration and the areal extent of the source region for particles settling out of the surface ocean (the so-called ‘statistical funnel’) and how it compares areally with that of the iron-enriched patch (Siegel et al. 2008). A further challenge at this time will be tracking the fate of the dissolved constituents of the bloom signature (i.e. after recycling by microbes and zooplankton). Some of these are climate-reactive gases with very high greenhouse warming potential (see Law 2008, this Theme Section), which could potentially offset the effects of C storage resulting from the bloom. This challenge becomes greater with a longer

Table 2. Sampling resolution of techniques used to obtain measurements of properties prior to, during, and after an FeAX. Range of datasets will pose problems for assimilating observations into a model that will be used to investigate the longer-term (decades) fate of the FeAX. Due to the large number of techniques used on the first generation of experiments, here we provide a small subset of techniques and properties. T: temperature; S: salinity; LIDAR: light detection and ranging; DMS: dimethylsulphide; DMSP: dimethylsulphoniopropionate. Note, in some cases analysis of samples—e.g. thorium for high-resolution estimates of export fluxes—would take months

Approach	Technique	Property	Temporal resolution
Remote-sensing	Glider	T, S, O <sub>2</sub> , nutrients, particle optics	Hours
	Instrumented buoy	T, S, O <sub>2</sub> , bio-optics, particle optics	10s of min
	Satellite	Chlorophyll, eddies (altimetry), temperature	Daily images
	Airborne LIDAR	Chlorophyll, photosynthetic competence	Min
Vessel—underway survey	Undulating tow-body	Chlorophyll, nutrients	Min
	Pumped seawater supply	Biogenic gases (e.g. DMS, CO <sub>2</sub> , CH <sub>4</sub> & DMSP), dissolved iron	Min
Vessel—discrete measurements	CTD vertical profiles	T, S, chlorophyll	Hours
	Water bottles/net tow	Water samples for N <sub>2</sub> O, phytoplankton (microscopy), grazing (experiments), thorium (export)	6 d <sup>-1</sup>
Moored instrumentation	Sediment traps	C export and C sequestration	1 measurement 2–3 d <sup>-1</sup>



experiment because of the impact of ocean currents/physical transports in diluting these signals as the patch gets larger. There is also the added difficulty of detecting such by-products of the decaying algal bloom over the background variability in these downstream waters (hence the need for some downstream survey before commencing the initial iron release).

The logistical challenges in an experiment such as we describe are formidable. They include the need for multiple ships (for the pre-release survey, experiment and post-bloom monitoring). This will necessitate the frequent replacement of vessels (occupation of a site for longer than 2 mo is not possible, as mentioned earlier) and the associated standardization and intercalibration exercises required to ensure uniformity during multiple vessel occupations. Besides using multiple ships for surveying the iron patch and adjacent areas, the emerging autonomous ocean sampling techniques should be used for monitoring and surveying processes inside the iron patch. For example, a set of sophisticated robotics vehicles (autonomous underwater vehicles, gliders and floats) equipped with optical and chemical sensors (Riser & Johnson 2008) will be useful not only to provide 3-dimensional distribution of physical and biogeochemical properties during initial release of iron and onset of phytoplankton bloom, but also for following the iron patch and detecting the decreasing signature of enhanced carbon production for months after the initial addition (Bishop et al. 2002). Finally, there will be a time lag in getting samples and data analysed and in ensuring quality control, prior to assimilation into the model.

A high-resolution eddy-resolving model, preferably guided by assimilation of real-time data, would be used to guide and simulate the release and the first phase of observations. It could be used to interpret the data being returned and to assess how representative the measurements taken at any one point are, which will be particularly important given that it will not be possible to obtain comprehensive observational coverage of the large area to be monitored using *in situ* measurements. Through near real-time iterations between observations and the model, it may be possible to command robotic vehicles to locales where their data will be most useful for assessing water and carbon movement. Finally, the model could be run forward after the main observation phase has ended, to predict the probable long term effects.

## CONCLUSIONS

It is evident that to conduct a second generation of experiments on the scales needed to address the issue of the utility of carbon sequestration and monitoring of

unintended side effects, a step increase in the scale of developmental work, costs, lead-in time, co-ordination and logistics will be necessary. We advocate a joint modeling-observational approach as the only means to conduct these second-generation studies. However, even at this early stage of development, it is clear that linking a wide range of measurements, many made on disparate time and/or spatial scales with models (via data assimilation) will be problematic. It may be necessary to use some of the observational results from the first generation of iron fertilization experiments to develop and improve physical-biogeochemical models (Fujii et al. 2005, Chai et al. 2007).

Modelling studies based on results from the first generation of studies point to the complex interplay of physics, photochemistry and biology in setting the concentrations of biogenic gases such as DMS (Le Clainche et al. 2006). It is not possible to have this level of detail in the regional or basin-scale circulation models needed to assimilate observations from the experiments, so a variety of different types of model studies will be necessary to fully interpret the experiments.

Modellers may be horrified by the faith that we are apparently placing in their codes. They will point out that the state of our knowledge is currently insufficient to place much confidence in their predictions and that different models presently give substantially different pictures of the effect of iron fertilization. Nevertheless, this is the state of our knowledge at the present: in this complex area the models represent our best synthesis of current understanding and the outcomes that they agree on are the only ones for which we can claim any certainty.

The program we advocate is a formidable challenge. However, it does not necessarily all have to be achieved in a single, massive effort. Many of the steps on the road to such a project are happening already—e.g. improvements in models, observational techniques and data assimilation methods. Implementation of second-generation iron fertilization experiments will become progressively more tractable in the coming years as we make progress in these related fields. The major challenge will then be to integrate all of these advances into a coherent program at the large scales necessary for the next generation of experiments.

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