Abstract. Since the start of the industrial revolution, human activities have caused a rapid increase in atmospheric CO_2 concentrations, which have in turn been cited as the cause of a variety climate changes such as global warming and ocean acidification. Various approaches have been proposed to reduce atmospheric CO_2 concentrations. The 'Martin (or Iron) Hypothesis' suggests that ocean iron fertilization (OIF) should be an efficient method for stimulating the biological pump in iron-limited high nutrient-low chlorophyll regions. To test the Martin hypothesis, a total 13 OIF experiments have been performed since 1990 in the Southern Ocean (7 times), in the subarctic Pacific (3 times), in the equatorial Pacific (twice), and in the subtropical Atlantic (once). These OIF field experiments demonstrated that primary production could be significantly increased after artificial iron addition. However, effectiveness in export production revealed by the OIF experiments was unexpectedly low compared to production from natural processes in all, except one of the experiments (i.e., the Southern Ocean European Iron Fertilization Experiment, EIFEX). These results, including possible side effects such as N_2O production and hypoxia development, have been scientifically debated amongst those who support and oppose OIF experimentation. In the context of increasing global and political concerns associated with climate change, it is valuable to examine the validity and usefulness of the OIF. We provide a general overview of the OIF experiments conducted over the last 25 years (past), a discussion of OIF debates including possible side effects and international law (present), a suggestion of considerations for designing future OIF experiments to maximize the effectiveness of OIF (future), and an introduction to the OIF experiment design guidelines for a future Korean Iron Fertilization Experiment in the Southern Ocean.

Keywords: Ocean Iron Fertilization; High-Nutrient and Low-Chlorophyll regions; Biological Pump; Phytoplankton; Iron

1 Introduction

Since the start of the industrial revolution, human activities have caused a rapid increase in atmospheric CO_2 from ~280 ppm (pre-industry) to ~400 ppm (present) (http://www.esrl.noaa.gov/), which has in turn led a variety of climate changes such as global warming and ocean acidification (IPCC, 2013) (Fig. 1). As the Anthropocene-climate system has rapidly changed toward the more unpredictable, scientific consensus is that the negative outcomes are a globally urgent issue that should be resolved in a timely manner for the sake of all lives on Earth (IPCC, 1990, 1992, 1995, 2001, 2007, 2013). Various ideas/approaches have been proposed to relieve/resolve the problem of global warming (Matthews, 1996; Lenton and Vaughan, 2009; Vaughan and Lenton, 2011; IPCC, 2014; Leung et al., 2014; Ming et al., 2014), largely based on two categories: (1) reduction of atmospheric CO_2 – ocean fertilization to enhance biological CO_2 uptake and/or direct capture or storage of atmospheric CO_2 through chemically engineered processes, and (2) control of solar radiation – artificial aerosol injection into the atmosphere to augment cloud formation and cloud brightening to elevate albedo (Fig. 2). One of the most attractive methods among the proposed approaches is *ocean fertilization* which targets the drawdown of atmospheric CO_2 by nutrient addition (e.g., iron, nitrogen or phosphorus compounds) to stimulate the phytoplankton growth via the ocean biological pump (ACE CRC, 2015).

The ocean biological pump (*a.k.a.* 'export production') is frequently depicted as a process whereby organic matter produced by phytoplankton during photosynthesis in surface waters is quickly transported to intermediate and/or deep waters (Fig. 3a) (Volk and Hoffert, 1985; De La Rocha, 2007). Although efficiency of the biological pump is mainly controlled by the supply of macro-nutrients (i.e., nitrate, phosphate, and silicate) into the euphotic zone leading to new production (Sarmiento and Gruber, 2006), iron acts as an essential micro-nutrient to stimulate the uptake of macro-nutrients for phytoplankton growth (Fig. 3b) (Martin and Fitzwater, 1988; Martin, 1990; Morel and Price, 2003). In the subarctic Pacific, equatorial Pacific, and Southern Ocean, which are well known as high-nutrient and low-chlorophyll (HNLC) regions (Fig. 4a and b), phytoplankton cannot completely utilize the available macro-nutrients (particularly nitrate) during photosynthesis due to a lack of iron. For this reason, primary production in these HNLC regions is relatively low in spite of the availability of nutrients (Fig. 4a and b).

It is thought, based on Arctic/Antarctic ice core analyses, that atmospheric CO_2 (~180 ppm) during the Last Glacial Maximum (LGM; ~20,000 years ago) was much lower than during pre-industrial times (~280 ppm) (Neftel et al., 1982; Barnola et al., 1987; Petit et al., 1999). Over the last 25 years, several hypotheses have been proposed to explain the mechanisms that lowered atmospheric CO_2 level during the LGM (Broecker, 1982; McElroy, 1983; Falkowski, 1997; Broecker and Henderson, 1998; Sigman and Boyle, 2000). One is particularly relevant to modern nutrient cycling in the Southern Ocean. In 1990, Martin hypothesized an LGM mechanism whereby the biological pump was substantially enhanced due to the relief of iron-limitation in HNLC regions, in particular the Southern Ocean, via high dust inputs (Fig. 3b). These dust inputs are generally regarded as one of major natural iron sources fertilizing oceans. He concluded with the now famous and often cited words "*Give me half a tanker of iron, and I will give you the next ice age*" (Martin, 1990). Since Martin's hypothesis was first published, there has been enormous interest in ocean iron fertilization (OIF) because only a small amount of iron (C:Fe ratios = 100,000:1, Anderson and Morel, 1982) is needed to stimulate a strong phytoplankton response. Therefore, much of the investigative focus has been centered on artificially adding iron to HNLC regions as a means of accelerating the ocean biological pump (ACE CRC, 2008).

To test the Martin's hypothesis, 2 natural and 13 artificial OIF experiments for scientific study have been performed to date in the subtropical Atlantic, equatorial Pacific, subarctic Pacific, and Southern Ocean (Blain et al., 2007; Pollard et al., 2009; Strong et al., 2009) (Fig. 4a and Table 1). These OIF experiments demonstrated that primary production could be significantly increased after iron addition (de Baar et al., 2005; Boyd et al., 2007). To evaluate whether OIF has potential as a geoengineering strategy for carbon sequestration, not only the amount of carbon fixed by phytoplankton at the ocean surface but also the

amount of carbon sequestered to the deep ocean must be considered in determining the effectiveness of OIF (Buesseler and Boyd, 2003). High export production/carbon sequestration effectiveness were observed from natural OIF experiments in the Southern Ocean near the Kerguelen Plateau and Crozet Islands (Blain et al., 2007; Pollard et al., 2009). However, the artificial OIF experiments showed unexpectedly weak responses compared to natural production in all the experiments (de Baar et al., 2005; Boyd et al., 2007), except one; the Southern Ocean European Iron Fertilization Experiment, EIFEX (Smetacek et al., 2012). These results, which include side effects such as N_2O production and hypoxia development (Fuhrman and Capone, 1991), have been scientifically debated amongst those who support and oppose OIF experimentation (Chisholm et al., 2001; Johnson and Karl, 2002; Lawrence, 2002; Buesseler and Boyd, 2003; Williamson et al., 2012).

In the context of increasing global and political concerns associated with rapid climate change, it is still valuable to examine the validity and usefulness of artificial OIF experimentation as a climate change strategy. Therefore, the purpose of this paper is (1) to provide an overview of the OIF experiments conducted over the last 25 years, (2) to discuss the pros and cons of OIF, including possible side effects and international law, (3) to suggest considerations for designing future OIF experiments with maximum effectiveness, and (4) to introduce design guidelines for future Korean Iron Fertilization Experiment in the Southern Ocean (KIFES) project.

2 Past: Overview of artificial OIF experiments

This overview of past OIF experimentation begins in Section 2.1 with a presentation of how each of the experiments was designed and why each was performed (Table 2). The unique prior ocean conditions for the various experiments are described in Section 2.2. Tracing the effects of iron addition is described in Section 2.3 and biogeochemical responses to the OIF experiments are presented in Section 2.4. Enhancement of carbon flux was assessed in Section 2.5 and why the outcomes of the experiments were so different and what has been learned from these experiments were discussed in Section 2.6 (Table 2).

2.1 Design/Objective of artificial OIF experiments

A total of 13 artificial OIF experiments have been conducted in the following areas: HNLC (i.e., nitrate >10 μM) regions such as the equatorial Pacific (twice), subarctic Pacific (3 times), and Southern Ocean (7 times), and one low-nutrient and low-chlorophyll (LNLC) (i.e., nitrate <10 μM) region, i.e., the subtropical Atlantic (once) (Table 1, Fig. 4a and b).

2.1.1 OIF in the equatorial Pacific

The first OIF experiment, IronEx-1 (Table 1), was carried out over 10 days in October 1993 in the equatorial Pacific (Martin et al., 1994; Coale et al., 1998). This region, located to the south of the Galapagos Islands, was proposed as an optimal place to perform an OIF experiment because (1) the warm temperatures, high light intensity, and low cloud cover allowed for rapid phytoplankton growth, (2) the relatively large number of research cruises conducted in the region provided sufficient physical and biogeochemical property information, (3) it was easily accessible, and (4) it provided an opportunity to examine the natural relationship between primary production and iron addition (via iron inputs into open-ocean waters via the plumes off the western coast of Galapagos Islands) before artificial OIF experiment (Martin and Chisholm, 1992; Martin et al., 1994).

However, the magnitude of biogeochemical responses in IronEx-1 was not as dramatic as expected (Martin et al., 1994). Three hypotheses were advanced to explain the unexpectedly weak results: (1) the possibility of other unforeseen micro-nutrient (e.g., cadmium and manganese) limitations, (2) the short residence time of bioavailable iron in the experimental surface

patch due to unstable water-column structure, and (3) the extremely high grazing stress placed on the patch by zooplankton (Cullen, 1995).

To investigate the unexpected responses revealed in IronEx-1, a second OIF experiment, IronEx-2, was conducted in May 1995 (Coale et al., 1996). The IronEx-2 research cruise occupied generally the same area for a longer period (17 days) providing more time to collect integrated information about the biogeochemical, physiological, and ecological responses to the OIF experiment. IronEx-2 demonstrated that massive phytoplankton blooming associated with OIF in the equatorial Pacific was possible, and it rekindled interest in and stimulated OIF experiments in other HNLC regions (Coale et al., 1996; Bidigare et al., 1999).

2.1.2 OIF in the Southern Ocean

The Southern Ocean, the largest HNLC region in the World Ocean, became the next region selected for OIF experimentation (Frost, 1996) because of its important role in intermediate and deep water formation, which suggested great potential for affecting the carbon sequestration associated with iron addition (Martin, 1990; Sarmiento and Orr, 1991; Cooper et al., 1996; Marshall and Speer, 2012). The Southern Ocean iron release experiment (SOIREE) (Table 1 and Fig. 4a), the first *in situ* OIF experiment performed in the Southern Ocean, took place in February 1999 (13 days) in the Australasian-Pacific sector of the Southern Ocean (Boyd et al., 2000). Iron-induced phytoplankton blooms confirmed that the supply of iron controls primary production in the Southern Ocean. It has also been shown that a model can produce LGM atmospheric CO_2 levels (~200 ppm) using SOIREE results with atmospheric dust flux obtained from the Vostok ice core analysis (Watson et al., 2000). The following year, a second Southern Ocean OIF experiment, EisenEx ('Eisen' is iron in German), was performed in November (23 days) in a closed cyclonic eddy of the Atlantic sector of the Southern Ocean (Smetacek, 2001).

The Southern Ocean exhibits markedly varied silicate concentrations on either side of the Antarctic Polar Front (PF): low silicate concentrations (<5 μM) to the north of the PF (<61° S) and high silicate concentrations (>60 μM) to the south of the PF (>61° S) (Fig. 4c). Silicate-requiring diatoms, which are one of the large-sized phytoplankton groups and have an important role in the biological pump, are responsible for ~75 % of the annual primary production in the Southern Ocean (Tréguer et al., 1995). Therefore, silicate availability is an important factor when considering the enhancement of export production via OIF experimentation. As SOIREE and EisenEx were performed to the south of the PF under intermediate silicate concentration (~5–25 μM) conditions (Boyd et al., 2000; Gervais et al., 2002) (Fig. 4c; Fig. 4a for experiment locations), the interaction between silicate availability and iron addition was not clearly verified. To elucidate this issue, two OIF experiments were conducted during January–February of 2002 in two distinct regions: The Southern Ocean iron experiment-north (SOFeX-N) and -south (SOFeX-S) of the PF (Coale et al., 2004; Hiscock and Millero, 2005) (Table 1).

To measure biologically driven gas fluxes (e.g., CO_2, dimethylsulfide, CO, N_2O, N_2, and O_2), the Surface Ocean Lower Atmosphere Study (SOLAS) Air–Sea Gas Exchange (SAGE) experiment was conducted during March–April 2004 (15 days) in HNLC sub-Antarctic waters (under low silicate concentration) between subtropical region and the PF (Harvey et al., 2010; Law et al., 2011) (Fig. 4c).

Early OIF experiments had not clearly shown whether artificial OIF could effectively reduce atmospheric CO_2 levels through enhancement of the biological pump, i.e., rapid transport of surface organic matter to intermediate/deep waters (Boyd et al., 2007), but the results were interesting enough to spur continued efforts. With the aim of confirming that OIF could increase export production, an experiment known as EIFEX was carried out during February–March 2004 in a PF cyclonic eddy core (Fig. 5). With the intention of finding deep export production, EIFEX was a much longer experiment (39 days), compared to earlier attempts (~21 days) (Smetacek et al., 2012). The Indian and German Atlantic sector iron fertilization experiment (LOHAFEX; 'Loha' is iron in Hindi) was conducted during January–March 2009 (40 days) also in a PF cyclonic

eddy at the same latitude with EIFEX, but under low silicate concentration (Fig. 4c) again with the aim of investigating an iron fertilized bloom in the surface layer, deep export carbon production, and biomass converted back into CO_2 by bacteria and/or zooplankton (Smetacek and Naqvi, 2010; Martin et al., 2013).

2.1.3 OIF in the subarctic North Pacific

A strong longitudinal gradient in aeolian dust deposition (i.e., high dust deposition in the west to low in the east), known as natural OIF, has been found in the subarctic North Pacific (Duce and Tindale, 1991). However, there was little information about differences in phytoplankton biomass and communities along the longitudinal dust gradient (Duce and Tindale, 1991; Moore et al., 2002). To investigate the relationship between phytoplankton biomass/community and this dust gradient, the Subarctic Pacific iron Experiment for Ecosystem Dynamics Study-1 (SEEDS-1) was conducted in July–August 2001 (13 days) in the western subarctic gyre using the RV *Kaiyo-Maru* (Tsuda et al., 2003, 2005), and the Subarctic Ecosystem Response to Iron Enrichment Study (SERIES) was performed in July–August 2002 (25 days) in the in the Gulf of Alaska using the RV *John P. Tully*, *El Puma*, and *Kaiyo-Maru* (Boyd et al., 2004, 2005). The main objective of SERIES was to investigate the duration of phytoplankton blooming (i.e., start to finish) after iron addition. Two years later, SEEDS-2 repeated the experiment in almost same location and season with SEEDS-1 using the RV *Hakuho-maru* and *Kilo-Moana* (Tsuda et al., 2007).

2.1.4 OIF in the subtropical North Atlantic

To investigate influence of co-limited iron and phosphate on primary production, the *in situ* PO_4^{3-} and Fe^{2+} addition experiment (FeeP) was conducted by adding both phosphate and iron in LNLC region of the subtropical North Atlantic during April–May 2004 (21 days) using two RV *Charles Darwin* and *Poseidon* (Rees et al., 2007).

2.2 Environmental conditions prior to iron addition

To investigate initial environmental conditions (~1–7 days before iron addition), physical and biogeochemical properties were determined at the sites of the OIF experiments (Steinberg et al., 1998; Coale et al., 1998; Bakker et al., 2001; Boyd and Law, 2001; Gervais et al., 2002; Coale et al., 2004; Boyd et al., 2005; Takeda and Tsuda, 2005; Tsuda et al., 2007; Cisewski et al., 2008; Harvey et al., 2010; Cavagna et al., 2011) (Fig. 6, Table 3 and 4). The OIF experiments were conducted under a wide range of physical conditions in terms of mixed layer depth (MLD) and sea surface temperature (SST).

The MLDs ranged from 10 m (SEEDS-1) to 97.6 m (EIFEX) (Fig. 6c), and were shallower in the equatorial Pacific (mean ± SD = 27.5 ± 2.5 m; SD represents standard deviation) and subarctic Pacific (mean ± SD = 22.7 ± 9.0 m) than in the Atlantic Ocean (FeeP: 40 m) and Southern Ocean (mean ± SD = 56.8 ± 18.9 m). Variation in MLD was highest in the OIF experiments conducted in the Southern Ocean and lowest in those conducted in the equatorial Pacific. MLDs in the experiments performed in the western subarctic Pacific were much shallower in SEEDS-1 (10 m) than in SEEDS-2 (28 m), even though the two experiments were carried out in nearly in the same location and season (Tsuda et al., 2007).

SST at the OIF sites ranged from -0.5 °C (SOFeX-S) to 25.2 °C (IronEx-2) (Fig. 6d). SST was much higher in the OIF experiments conducted in the equatorial Pacific (mean ± SD = 24.1 ± 1.15 °C) and Atlantic Ocean (FeeP: 20.7 °C) than those conducted in the Southern Ocean (mean ± SD = 9.4 ± 2.2 °C) and subarctic Pacific (mean ± SD = 4.9 ± 3.7 °C). Although the two OIF experiments carried out in the equatorial Pacific occurred in different seasons (i.e., IronEx-1: October, IronEx-2: May), the surface physical conditions were quite similar (Steinberg et al., 1998). SOFeX-N/S which were conducted along the same line of longitude in the Southern Ocean exhibited distinct differences in SST; 7.1 °C in SOFeX-N and -0.5 °C in SOFeX-

S (Coale et al., 2004). Among the OIF experiments conducted in the Southern Ocean, SAGE carried out in the late summer (late March – early April) had the highest SST (11.5 °C) (Harvey et al., 2010).

Regions for OIF experimentation have usually been selected using preliminary surveys to confirm that the sites were subject to HNLC conditions: high nitrate concentration (>~10 μM) and low chlorophyll-a concentration (<1 mg m^{-3}). Nitrate concentrations ranged from 7.9 μM (SAGE) to 26.3 μM (SOFeX-S) (Fig. 6e and Table 3). Among the various OIF HNLC experiment sites, the equatorial Pacific (i.e., IronEx-1 and IronEx-2) had the lowest initial nitrate concentrations (mean ± SD = 10.6 ± 0.2 μM), while the Southern Ocean had the highest (mean ± SD = 21.2 ± 5.8 μM). One exception to the focus on HNLC study sites was the FeeP experiment which took place in the subtropical North Atlantic, a typically LNLC region (nitrate <0.01 μM and chlorophyll-a <1 mg m^{-3}) (Fig. 6e and h, Table 3 and 4).

The full range of initial silicate concentrations for all the OIF experiments is expressed in the Southern Ocean where values ranged widely ranged from 0.83 μM (SAGE) to 62.8 μM (SOFeX-S) (Fig. 6f and Table 3). Generally speaking, however, initial silicate concentrations were lower in the equatorial Pacific (mean ± SD = 4.5 ± 0.6 μM) than in the Southern Ocean (mean ± SD = 15.1 ± 20.4 μM) and subarctic Pacific (mean ± SD = 27.3 ± 9.6 μM). Nevertheless, SOFeX-N, SAGE, and LOHAFEX all conducted in the Southern Ocean were representative of very low-silicate HNLC (HNLCLSi) regions with initial silicate concentrations less than 2.5 μM (Coale et al., 2004; Harvey et al., 2010; Martin et al., 2013).

Phosphate concentrations ranged from 0.01 μM (FeeP) to 1.9 μM (SOFeX-S) (Table 3). Consistent with the World Ocean Circulation Experiment sections and maps (Talley, 2007; Koltermann et al., 2011) which suggest increasing surface and near surface nitrate values from Antarctica equatorward, initial Southern Ocean phosphate concentrations were higher to the south 50° S (mean ± SD = 1.6 ± 0.2 μM) than to the north (mean ± SD = 1.1 ± 0.4 μM). They were also higher in the Atlantic sector (mean ± SD = 1.6 ± 0.2 μM) than in the Pacific sector (mean ± SD = 1.0 ± 0.5 μM). Consistent with both the meridional gradient and the basin differences, IronEx in the equatorial Pacific found generally lower initial phosphate values (<1 μM) similar to those seen by SAGE in the southwest Pacific.

Using continuous shipboard measurement systems, OIF experiments have also observed initial surface partial pressure of CO_2 (pCO_2) conditions (Wanninkhof and Thoning, 1993; Steinberg et al., 1998; Bakker et al., 2001; Bakker et al., 2005; Hiscock and Millero, 2005; Takeda and Tsuda, 2005; Smetacek et al., 2005; Wong et al., 2006; Tsumune et al., 2009; Currie et al., 2011). Initial pCO_2 values ranged from 330 ppm (SAGE) to 538 ppm (IronEx-2) (Table 3). Initial pCO_2 values were much higher in the equatorial Pacific (mean ± SD = 504.5 ± 33.5 ppm) than those in the Southern Ocean (mean ± SD = 355.3 ± 12.5 ppm) and subarctic Pacific (mean ± SD = 370 ± 16.3 ppm).

As previously mentioned, photosynthetic quantum efficiency (Fv/Fm, where Fm is the maximum chlorophyll fluorescence yield and Fv is the difference between Fm and the minimum chlorophyll fluorescence yield), is widely used to determine the degree to which iron is the limiting nutrient for phytoplankton growth (Fv/Fm ranges from 0 to 1 where conditions are less iron limited condition as Fv/Fm approaches 1) (Boyd et al., 2005). Initial Fv/Fm ratios were less than ~0.3 (Fig. 6g and Table 4) suggesting a tendency for iron limitation. Prior to iron addition, initial chlorophyll-a concentration, measured by fluorometer, ranged from 0.04 mg m^{-3} (FeeP) to 0.9 mg m^{-3} (SEEDS-1) (Fig. 6h and Table 4). However, as was the case for nitrate, compared to all the other OIF experiment sites, FeeP showed unusually low initial chlorophyll-a. The average initial OIF chlorophyll-a concentration was 0.43 ± 0.27 mg m^{-3}. Prior to the OIF experiments, except in SEEDS-1, SOFeX-S, and EIFEX where the diatoms were dominated by micro-plankton (20–200 μm), phytoplankton communities were dominated by pico-plankton (0.2–2.0 μm) and nano-plankton (2.0–20 μm) (Coale et al., 1998; Landry et al., 2000; Boyd and Law, 2001; Gervais et al., 2002; Tsuda et al., 2005; Coale et al., 2004; Boyd et al., 2005; Hoffmann et al., 2006; Harvey et al., 2010; Tsuda et al., 2007; Martin et al., 2013).

2.3 Tracing the effects of iron addition

Iron(II) and sulfate aerosols are ubiquitous in the atmosphere, and therefore iron-sulfate ($FeSO_4 \cdot H_2O$), a common form of combined iron that enters the ocean environment via dust deposition, has been frequently regarded as a bioavailable iron source for glacial periods (Zhuang et al., 1992; Zhuang and Duce, 1993; Spolaor et al., 2013). In addition, iron-sulfate is a common inexpensive agricultural fertilizer that is relatively soluble in acidified seawater (Coale et al., 1998). Therefore, OIF experiments have been carried out by releasing commercial iron-sulfate dissolved in acidified seawaters into moving ship propeller wash (Fig. 5).

The patch size fertilized by the first iron addition varied from 25 km^2 (e.g., FeeP; iron addition of 1840 kg) to 300 km^2 (e.g., LOHAFEX; iron addition of 2000 kg) (Boyd et al., 2007; Martin et al., 2013) (Table 1 and Fig. 6a and b). In general, background dissolved iron concentrations in the ocean environment are <0.2 nM. During the OIF experiments, dissolved iron concentrations increased to ~1.0–4.0 nM and decreased to background concentrations within days (Table 1). The fast decrease in dissolved iron concentrations indicates that iron-sulfate was transformed chemically into a solid form that readily sticks to other substances. This process occurs more rapidly in warmer waters (ACE CRC, 2015). Therefore, except for the single iron addition experiments of IronEx I, SEEDS-1, and FeeP (Martin et al., 1994; Tsuda et al., 2005; Rees et al., 2007), to maintain an iron-fertilized patch, most of the OIF experiments conducted multiple iron additions at the patch centre. These multiple addition experiments included: (2 additions) EIFEX, SERIES, SEEDS-2, LOHAFEX (Boyd et al., 2005; Smetacek et al., 2012; Martin et al., 2013), (3 additions) IronEx-2, EisenEx, SOFeX-N (Coale et al., 1996; Gervais et al., 2002; Coale et al., 2004; Nishioka et al., 2005), and (4 additions) SOIREE, SOFeX-S, SAGE (Boyd and Law, 2001; Coale et al., 2004; Harvey et al., 2010) (Table 1).

To trace the iron-fertilized patch, OIF experiments used a combination of biogeochemical-based and physical-based approaches. In biogeochemical approaches, the OIF experiments (except EIFEX) used sulfur hexafluoride (SF_6) artificially added as a chemical tracer (Martin et al., 1994; de Baar et al., 2005). SF_6 is useful for investigating physical mixing and advection-diffusion processes in the ocean environment due to its low solubility, nontoxicity, and biogeochemically inert characteristics (Law et al., 1998). Injected SF_6 concentrations were continuously monitored using gas chromatography with an electron capture detector system (Law et al., 1998; Tsumune et al., 2005). Usually only one SF_6 injection was necessary as background levels are generally extremely low in the ocean (<1.2 fM; f: femto-, 10^{-15}) (Law et al., 1998; Law et al., 2006; Martin et al., 2013), however, in the SAGE experiment with higher mixing and lateral dilution, there were three injections (Harvey et al., 2010). Underway sampling systems, measuring biogeochemical parameters such as Fv/Fm ratio, pCO_2, and chlorophyll fluorescence, were also used in the fertilized patch as alternative means of following the patch (Gervais et al., 2002; Boyd et al., 2005; Tsuda et al., 2007; Harvey et al., 2010; Smetacek et al., 2012).

In physically based approaches, surface-drifting buoys equipped with Array for Real-time Geostrophic Oceanography (ARGO) and Global Positioning System (GPS) sensors have been used to map moving fertilized patches in space and time (Coale et al., 1998; Boyd and Law, 2001; Law et al., 2006; Martin et al., 2013). Buoy position can be transmitted to the ship every 5–10 min. The NASA-airborne oceanographic lidar aircraft have also been employed to assess the large-scale effects of iron addition on surface chlorophyll in the fertilized patch compared to surrounding regions (Martin et al., 1994).

2.4 Biogeochemical responses

The biogeochemical responses to a wide range of iron addition (350–4000 kg) via OIF experiments in the HNLC regions were surveyed over periods ranging from 10–40 days (Table 1 and Fig. 6b). The initial response was a rapid increase of Fv/Fm ratio generally observed within the first 24 hours after iron addition. This was not the case in SEEDS-1 and SEEDS-2 where a detectable increase was observed 3–5 days later. The maximum post-iron-addition Fv/Fm ranged from 0.31 (SEEDS-1) to 0.65 (SOIREE and SOFeX-S) and Fv/Fm generally reached values of 0.5 or greater (Table 4 and Fig. 7a). The increase in Fv/Fm ratio after iron addition suggests that phytoplankton response to iron enrichment is prompt, and results support the hypothesis that natural phytoplankton growth in these HNLC regions is iron-limited (Boyd and Abraham, 2001; Gervais et al., 2002; Tsuda et al., 2003; Boyd et al., 2005; Barber and Hiscock, 2006; Tsuda et al., 2007; Peloquin et al., 2011; Croot et al., 2008; Martin et al., 2013).

Depletion of macro-nutrients in fertilized patches provides indirect evidence that phytoplankton growth in surface waters is driven by iron fertilization (Boyd and Law, 2001). Significant nitrate uptake (i.e., $\Delta\text{Nitrate} = [NO_3^-]_{\text{post-fertilization}} - [NO_3^-]_{\text{pre-fertilization}} < 0$) occurred in all the OIF experiments, except SAGE (Table 3 and Fig. 7b) (Martin et al., 1994; Boyd et al., 2000; Hiscock and Millero, 2005; Law et al., 2011). Negative ΔNitrate ranged from -0.7 μM in the equatorial IronEx1 experiment to -15.8 μM in SEEDS-1. However, in SAGE, concentrations of macro-nutrients in the iron fertilized patch exceeded the initial concentrations (i.e., ΔNitrate >0) due to the physical processes such as deepened MLD and lateral advection of high nutrient waters (Table 3 and Fig. 7b) (Law et al., 2011).

Changes in surface water chlorophyll-a concentrations are a direct indication of the effect of iron addition on phytoplankton growth (Fig. 7c). Generally, chlorophyll-a concentrations increased substantially 2- to 20-fold with max values of ~0.1 mg m^{-3} (FeeP) to 22 mg m^{-3} (SEEDS-1) (Fig. 7c and Table 4) when nitrate concentrations sharply decreased from 3–5 days after iron addition (Tsuda et al., 2003; Coale et al., 2004; Boyd et al., 2004; Tsuda et al., 2007; Peloquin et al., 2011; Smetacek et al., 2012). SEEDS-1 and SEEDS-2, performed under similar conditions, presented similar initial chlorophyll-a concentrations (0.8 mg m^{-3}), but their responses to iron addition were different. Iron-stimulated max chlorophyll-a concentration in SEEDS-2 (~2.5 mg m^{-3}) was much lower than those of SEEDS-1 (~22 mg m^{-3}) (Tsuda et al., 2007). Satellite observations were also used to spatially and temporally map OIF phytoplankton response (Boyd et al., 2000; Coale et al., 2004; Boyd et al., 2005; Westberry et al., 2013). Spatial changes in chlorophyll-a concentration as a result of iron addition were detected in SOFeX-N/S using Sea-viewing Wide Field-of-view Sensor (SeaWiFS) and MODerate resolution Imaging Spectrometer (MODIS) Terra Level-2 chlorophyll-a images. The chlorophyll-a image at ~28 days after iron addition in the SOFeX-N showed a phytoplankton bloom distribution resembling a long thread shape (~1.0 mg m^{-3}), while chlorophyll-a image at ~20 days in the SOFeX-S suggested a somewhat broader bloom pattern (~0.4 mg m^{-3}) (Fig. 7d) (Westberry et al., 2013).

However, influence of iron addition on the phytoplankton growth covers from surface to euphotic depth as added iron is mixed within the mixed layer by physical processes (Coale et al., 1998). Although maximum chlorophyll-a concentrations during SEEDS-1 (~22 mg m^{-3}) were much higher than EIFEX (~3.2 mg m^{-3}), mixed layer integrated chlorophyll-a concentrations were similar to ~250 mg m^{-2}. There were distinct differences between mixed layer integrated chlorophyll-a concentration and surface chlorophyll-a concentration. Therefore, during previous OIF experiments, to quantify the exact changes in phytoplankton biomass to iron addition, it would be important to detect the change in integrated primary productions within MLDs. Associated with the OIF-induced phytoplankton blooms, the magnitude of primary productivity integrated from the surface to euphotic depth in iron fertilized patches also became significantly elevated compared to initial levels (i.e., $\Delta PP = PP_{\text{post-fertilization}} - PP_{\text{pre-fertilization}}$, where PP is primary productivity). Increases in ΔPP ranged from 360 mg C m^{-2} d^{-1} (SAGE) to 1800 mg C m^{-2} d^{-1} (IronEx-2) with maximum values of 790 mg C m^{-2} d^{-1} (EisenEx) to 2430 mg C m^{-2} d^{-1} (IronEx-2) (Fig. 7e

and Table 4). As a result of increased ΔPP, drawdown of pCO_2 (negative ΔpCO_2: air→sea) was enhanced during the all OIF experiments except SAGE (Fig. 7f). In SAGE, physical mixing caused an increase in macro-nutrients (positive ΔNitrate, Fig. 7b), which resulted in a reversed pCO_2 pattern (positive ΔpCO_2: sea→air) (Currie et al., 2011). The largest ΔpCO_2 change occurred in SEEDS-1 conducted in the subarctic North Pacific (Fig. 7f). It also produced the largest ΔNitrate and the greatest chlorophyll increase (Fig. 7b and c) (Tsuda et al., 2003; de Baar et al., 2005). Overall, OIF ΔpCO_2 reductions ranged from -6 ppm (SEEDS-2) to -130 ppm (SEEDS-1) (Table 3 and Fig. 7f), and were associated with DIC decreases of 6 μM (IronEx-1) to 58 μM (SEEDS-1) (Steinberg et al., 1998; Bakker et al., 2001; Bakker et al., 2005; Boyd et al., 2007; Berg et al., 2011; Currie et al., 2011).

Using both microscopes and high performance liquid chromatography pigment analysis, changes in phytoplankton community effected by iron addition were also investigated. During IronEx-2, SOIREE, EisenEx, SEEDS-1, SOFeX-S, SERIES, and EIFEX, the dominant phytoplankton community tended to shift from pico- and nano-plankton to micro-plankton, resulting in diatom-dominated phytoplankton blooming, a key component for biological pump enhancement (Landry et al., 2000; Boyd and Law, 2001; Gervais et al., 2002; Tsuda et al., 2005; Coale et al., 2004; Boyd et al., 2005; Hoffmann et al., 2006; Harvey et al., 2010). However, there were no observations on taxonomic shift toward diatom-dominated phytoplankton communities in other OIF experiments (Coale et al., 1998; Coale et al., 2004; Rees et al., 2007; Tsuda et al., 2007; Peloquin et al., 2011; Martin et al., 2013). As noted above, the SEEDS-1 and SEEDS-2 experiments were carried out under similar ocean conditions. Nevertheless, SEEDS-2, which resulted in a minimal increase in chlorophyll-a (<3 mg m^{-3}), was also subject to extensive copepod (meso-zooplankton; 200–2000 μm) grazing (~5 times greater than in SEEDS-1) and therefore did not produce a prominent diatom bloom (Tsuda et al., 2007).

2.5 Assessment of export carbon flux

Early OIF experiments showed that iron addition stimulates the first step of the biological pump, promotion of phytoplankton growth. To determine whether the second step of the biological pump, export of carbon to the deep sea (i.e., increased export production), is enhanced after iron addition, the export flux of particulate organic carbon (POC) has been estimated using, either together and/or individually, chemical tracers such as natural radiotracer thorium-234 (^{234}Th; half-life = 24.1 days) and the stable carbon isotope of particulate organic matter ($^{13}C_{org}$), free-drifting sediment traps, beam-transmissometers, and underwater video profilers (UVP) (Table 5) (Bidigare et al., 1999; Nodder et al., 2001; Boyd et al., 2004; Buesseler et al., 2004; Coale et al., 2004; Aono et al., 2005; Tsuda et al., 2007; Smetacek et al., 2012; Martin et al., 2013).

The ^{234}Th isotope has a strong affinity for suspended particles, and the extent of ^{234}Th removal in the water column is indicative of the export production below the euphotic zone associated with surface primary productivity (Buesseler, 1998). In IronEx-2, which was the first OIF experiment in which POC flux was estimated, surface values were calculated from the so-called ^{234}Th activity balance method (Bidigare et al., 1999). The ^{234}Th deficiency of the surface ocean (25 m) during IronEx-2 was evident in the iron-fertilized patch, indicating iron-stimulated export production (Table 5). However, there were no ^{234}Th observations conducted in the unfertilized patch for comparison and nor were there observations to estimate downward POC export to the deep ocean (Bidigare et al., 1999).

SOIREE was first study to quantify downward export processes to the deep Southern Ocean using a comprehensive suite of methods such as ^{234}Th and $^{13}C_{org}$ estimates derived from high volume pump sampling, free-drifting sediment trap deployments, and beam transmissometer (Nodder and Waite, 2001). However, no measurable change in carbon export was

observed in response to iron-stimulated primary production during the 13-day occupation of the SOIREE (Fig. 8b and Table 5) (Charette and Buesseler, 2000; Nodder and Waite, 2001; Trull and Armand, 2001; Waite and Nodder, 2001). For EisenEx, ^{234}Th observations showed no differences between in-patch and outside-patch export rates (U. Riebesell et al., unpublished manuscript). Although POC export fluxes in the surface layer (50 m) changed from 374 to 1000 mg C m^{-2} d^{-1} with the formation of an iron-induced phytoplankton bloom during SEEDS-1, there was no significant increase in POC export flux measured from the drifting sediment trap deployments at 200 m during the observation period (Aono et al., 2005). These results suggest that most of the POC stayed in the surface mixed layer, that is, did not extend down to 200 m (Takeda and Tsuda, 2005).

For SOFeX-N/S, enhanced POC fluxes out of the mixed layer after iron enrichment were obtained from ^{234}Th observations (SOFeX-S) and free-profiling robotic Lagrangian carbon explorers with transmissometers (SOFeX-N) (Bishop et al., 2004; Buesseler et al., 2005). However, the absolute magnitude of these flux increases was similar to those for natural blooms in the Southern Ocean. During SERIES and SEEDS-2, which allowed comprehensive time-series measurements of the development and decline of an iron-stimulated bloom, POC fluxes defined by the sediment trap deployment showed temporal variation with development and decline phases in the fertilized patch (Boyd et al., 2004; Aramaki et al., 2009). These results suggested that only small part of the decrease in mixed layer POC was subsequently captured by the trap and losses of POC flux were mainly governed by bacterial remineralization and mesozooplankton grazing (Boyd et al., 2004; Tsuda et al., 2007). For SAGE and LOHAFEX under Si limitation in the Southern Ocean (Fig. 4c and 6f), there was no detection for fertilization-induced export by any method (Table 5) (Peloquin et al., 2011; Martin et al., 2013).

In contrast to other previous experiments, EIFEX provided clear evidence that the carbon export was stimulated by artificial iron addition (Jacquet et al., 2008). During EIFEX, initial the export flux, estimated using ^{234}Th in the upper 100 m of the fertilized patch, was 340 mg C m^{-2} d^{-1} (Fig. 8a and Table 5) (Smetacek et al., 2012). This value remained constant for 25 days after iron addition. Then, between 30 and 36 days after iron addition, a massive increase in export flux as high as 1692 mg C m^{-2} d^{-1} was observed in the fertilized patch, while the initial value remained constant in the unfertilized patch (Fig. 8a and Table 5). The profiling transmissometer with high-resolution coverage also showed that there was an increase in exported POC below 200 m after 28 days. At least half of iron-induced biomass sank far below to a depth of 1000 m with tenfold higher sinking rate (500 m d^{-1}), comparable to the initial conditions, via aggregate formations of diatom species, '*Chaetoceros dichaeta'* (Smetacek et al., 2012). That being said, EIFEX was the exception. Significant changes in export production were not found in any of the other OIF experiments, suggesting that the effectiveness of iron addition on this component of the biological pump remains a question that needs to be resolved in future OIF experiments (Bidigare et al., 1999; Nodder et al., 2001; Boyd et al., 2004; Buesseler et al., 2004; Coale et al., 2004; Aono et al., 2005; Tsuda et al., 2007; Smetacek et al., 2012; Martin et al., 2013).

2.6 Significant results and limitations in previous OIF experiments

To understand how various physical and biogeochemical properties response to artificial iron addition in HNLC regions, previous OIF experiments have been conducted with various objectives (Table 2). These various objectives have contributed to develop ideas/approaches to find optimal conditions that have potential capacity to efficiently sequester carbon (Smetacek et al., 2012). To test iron hypothesis, initial artificial OIF experiments (e.g., SEEDS-1 and IronEx-2) have focused on whether iron supply limits phytoplankton growth in HNLC regions and have confirmed increases in phytoplankton biomass by showing maximum drawdown of pCO_2 by 130 ppm in SEEDS-1 and in primary production by 1800 mg C m^{-2} d^{-1} in IronEx-2 (Fig. 7e and f) (de Baar et al., 2005; Boyd et al., 2007). Massive phytoplankton bloom was due to rapid increase in diatom production

(Coale et al., 1996; Boyd et al., 2000). There were multiple efforts to detect deep export production from surface iron-induced massive phytoplankton bloom, as the second step of iron hypothesis (Bidigare et al., 1999; Charette and Buesseler, 2000; Coale et al., 2004; Smetacek et al., 2012). EIFEX only showed significant export carbon to deep layer of 3000 m by aggregate formation with highly fast sinking rates (Table 5) (Smetacek et al., 2012). Despite highly increased phytoplankton production in the mixed layer by OIF experiments (e.g., SEEDS-1, SOFeX-N/S, SERIES, and SEEDS-2), export production was relatively low. Thus, the study focus was on high bacterial remineralization (SERIES) and/or grazing pressure (SEEDS-2) in the upper water columns (Boyd et al., 2004; Tsuda et al., 2007). Relatively slight increase in primary production to iron addition (~500 mg C m^{-2} d^{-1}) occurred in SAGE and LOHAFEX experiments, which were designed to investigate biogeochemical response to iron addition in very low silicate concentrations (<~2 nM) (Table 2) (Coale et al., 2004; Harvey et al., 2010; Martin et al., 2013).

3 Present: OIF debates and considerations including possible side effects and international law

3.1 Environmental side effects

OIF has been proposed as one potential way (*a.k.a.* 'Carbon Capture Storage') of rapidly and efficiently reducing atmospheric CO_2 levels at relatively minimal cost (Buesseler and Boyd, 2003). Over the past 25 years, controlled OIF experiments have illustrated that substantial increases in phytoplankton biomass can be instigated in HNLC regions through iron addition that results in the drawdown of DIC and macronutrients (de Baar et al., 2005; Boyd et al., 2007; Smetacek et al., 2012; Martin et al., 2013). However, the effectiveness of enhancement in this export production, which results in a net transfer of CO_2 from the atmosphere to the ocean intermediate/deep layer (i.e., 'biological pump'), is not yet fully understood or quantified as it appears to vary with region, season, and as yet unknown factors (Smetacek et al., 2012). Therefore, it is uncertain whether OIF has the potential to sequester CO_2 at a significant rate (~1 Gt of CO_2 per year). In the meantime, there are possible environmental side effects in response to iron addition, such as production of greenhouse gases (e.g., N_2O and CH_4) (Lawrence, 2002; Liss et al., 2005; Law, 2008), development of hypoxia/anoxia in water column (Sarmiento and Orr, 1991), and toxic algal blooms (e.g., *Pseudo-nitzschia*) (Silver et al., 2010; Trick et al., 2010), that have been seen and should be addressed before artificial OIF is conducted. These OIF experiment side-effects may themselves effect climate and ecosystem changes that have unexpected negative outcomes (Fuhrman and Capone, 1991). Therefore, it is not surprising that the OIF validation and usefulness has been a subject of debate (Williamson et al., 2012).

OIF experiments have measured climate-relevant gases (i.e., N_2O, CH_4, dimethylsulfide, and halogenated volatile organic compounds) that are produced by biological activity and/or photochemical reaction (Liss et al., 2005) to investigate change before and after iron addition. CH_4 has been considered to be relatively low risk as most of the CH_4 formed in the ocean is used as energy source for microorganisms and is converted to CO_2 before reaching to the sea surface (Smetacek and Naqvi, 2008; Williamson et al., 2012). Measurements of dissolved CH_4 during the SOFeX-N showed slightly elevated concentrations at less than 1 % (Wingenter et al., 2004). Simulated Southern Ocean large-scale iron fertilization has suggested that enhancement of CH_4 emission would offset only <1 % of the resulting carbon sequestration (Oschlies et al., 2010). On the other hand, the ocean is already a significant source for atmospheric N_2O, which has relatively the long lifetime (~110 years) in the atmosphere and has a global warming potential about 300 times greater than CO_2 (Forster et al., 2007). Therefore, any enhancement of biological production that might enhance N_2O emission could work to increase atmospheric greenhouse gas levels rather than decrease them (Bange, 2006). During the SOIREE experiment, a significant increase (~7 %) in mean N_2O saturation in the pycnocline of the fertilized patch was associated with increased phytoplankton biomass (Law and Ling, 2001).

Measurements of N_2O saturation during SERIES also showed increases of 8 % at 30–50 m, which were coincident with the accumulation of ammonium and nitrite attributable to bacterial remineralization (Boyd et al., 2004; Law, 2008). Model estimates suggested that potential N_2O production on longer timescales (6 weeks) would subsequently offset by 6–12 % increased carbon reduction benefits resulting from remineralization of additional carbon fixed during SOIREE (Law and Ling, 2001). This estimate is in the same range as the N_2O offset of 6–18 % suggested by an earlier modeling study (Jin and Gruber, 2003) and the 5–9 % suggested by a more recent modeling study investigating the effectiveness of long-term and large-scale Southern Ocean OIF (Oschlies et al., 2010). Complicating the story, however, excess N_2O was not found after iron addition during EIFEX, which showed significant vertical export with formation of rapidly sinking aggregate (Walter et al., 2005; Law, 2008). An explanation for the absence of N_2O accumulation below EIFEX patch might be limited bacterial remineralization by rapid export to the seafloor (Walter et al., 2005).

Unlike N_2O emissions which have the potential to offset the effectiveness of OIF, dimethylsulfide (DMS), hypothesized to be a precursor of sulfate aerosols that cause cloud formation and so climate cooling, may contribute to the homeostasis of the earth's climate by countering warming from increasing CO_2 (Charlson et al., 1987). The DMS response to iron addition was measured during all OIF experiments. In equatorial Pacific and Southern Ocean, DMS increased, but in the subarctic Pacific, it remained constant or decreased (Lawrence, 2002; Boyd et al., 2007). Significant short-term increases in DMS production were found in IronEx-2, SOIREE, EisenEx, and SOFeX-N (Turner et al., 1996; Turner et al., 2004; Wingenter et al., 2004; Liss et al., 2005). The maximum DMS production observed was a 6.5-fold increase after iron addition during SOIREE (Turner et al., 2004). Similarly, a 5-fold enhancement of DMS was observed during SOFeX-N. Estimates derived by extrapolation of SOFeX-N DMS results to the Southern Ocean OIF experiment with 2 % areas suggested that iron fertilization would enhance DMS production by 20 %, which would lead to a 2 °C decrease in air temperature over the Southern Ocean (Wingenter et al., 2007). Interestingly, there were no significant changes in DMS production after iron addition in SEEDS-1 and SEEDS-2, despite increases in primary production (Turner et al., 1996; Takeda and Tsuda, 2005; Nagao et al., 2009). Contrast to SEEDS-1 and SEEDS-2, DMS production decreased in SERIES experiment due to the relatively high bacterial dimethylsulfoniopropionate (DMSP) metabolism (Levasseur et al., 2006), which is precursor of DMS production. It is therefore clear that there are yet unknown factors affecting iron-induced DMS response, as it appears that OIF could be a significant source of DMS production in Southern Ocean and yet induce a DMS sink in subarctic Pacific. These results indicate that further observation-based and modeling studies are required to determine different features from place to place (Law, 2008).

Halogenated volatile organic compounds (HVOCs, such as CH_3Cl, CH_3Br, and CH_3I), well known for their ability to destroy ozone in the lower stratospheric ozone and marine boundary layer (Solomon et al., 1994), were also measured during the OIF experiments (Wingenter et al., 2004; Liss et al., 2005). During SOFeX-N experimentation, iron addition results for HVOC were complicated: CH_3Cl concentrations remained unchanged; CH_3Br concentrations increased by ~14 %; and while generally CH_3I concentrations decreased by ~23 % (Wingenter et al., 2004). CH_3I concentrations increased 2-fold in EisenEx (Liss et al., 2005). Therefore, as the DMS response above, further study is needed to understand the complexity of the HVOC response.

Decomposition of iron addition-enhanced biomass may cause decreased oxygen concentrations in the subsurface waters (Williamson et al., 2012). Although mid-water oxygen depletion has not been reported during the OIF experiments to date, it has been suggested that OIF-induced oxygen depletion may occur as increased downward carbon exports elevate microbial respiration (Fuhrman and Capone, 1991). Early studies using box model solutions have further suggested that anoxic conditions may develop after OIF (Sarmiento and Orr, 1991). However, more sophisticated and realistic models associated with OIF-

induced oxygen changes in water columns showed well-oxygenated conditions without developing anoxic conditions (Oschlies et al., 2010; Keller et al., 2014).

The changes of phytoplankton community composition after iron addition discussed in Section 2.4 may also have unintended consequences, in particular, toxin production (Silver et al., 2010; Trick et al., 2010). Some OIF experiments (including IronEx-2, SOIREE, EisenEx, and SOFeX-N/S) generated large blooms of diatoms dominated by pennate diatoms belonging to the genus '*Pseudo-nitzschia*' (de Baar et al., 2005; Trick et al., 2010). Some species of the genus '*Pseudo-nitzschia*' have the capacity to produce the neurotoxin domoic acid (DA) that is known to detrimentally affect marine ecosystems. For example, during IronEx-2 and SOFeX-S, high cell abundances of '*Pseudo-nitzschia*' (106 and 105 cells l^{-1}, respectively) combined with moderate DA quotas (0.05 and 1 pg DA $cell^{-1}$, respectively) produced toxin levels as high as 45 ng DA l^{-1} in IronEx-2 and 220 ng DA l^{-1} in SOFeX-S; i.e., toxin levels that is able to cause certain damages to marine communities in coastal waters (Silver et al., 2010). However, no DA was found during EisenEx, even though diatom species of the genus '*Pseudo-nitzschia*' were dominant numerically (Gervais et al., 2002; Assmy et al., 2007).

The direct and indirect environmental consequences of OIF remain unresolved due to inconsistent, highly uncertain outcomes (Williamson et al., 2012; Johnson and Karl, 2002; Chisholm et al., 2001), suggesting that we haven't yet reached to a conclusion as to whether OIF is a feasible carbon removal strategy (Boyd et al., 2007). Therefore, evaluation and prediction are paramount. It continues to be a valuable exercise to seek answers to scientific questions about the effectiveness of OIF as a means of reducing atmospheric CO_2 as well as to quantify possible OIF side effects. In particular, potential trace gas emissions such as N_2O and DMS, which are influenced by the remineralization of sinking particles that follows OIF-induced blooms, are important to understand. They can directly and indirectly modify the desired carbon sequestration effectiveness and they can do so both positively and negatively. Therefore, monitoring of N_2O and DMS to evaluate the effectiveness of OIF as a geoengineering approach is essential.

3.2 International law of the sea to OIF

To date, assessment of the effectiveness of OIF has been limited by the small area of the fertilized patches (25–300 km^2) used in the experiments (Fig. 6a). Patch sizes have been limited in part due to the time and expense of comparing fertilized and unfertilized areas (ACE CRC, 2008). Nevertheless, as these small-scale OIF experiments have demonstrated considerable potential for easily and efficiently reducing atmospheric CO_2 levels, physical/biogeochemical/ecological models and natural (long-term) iron fertilization experiments have been studied in an effort to overcome some of the limitations of short-term iron-addition experiments and to predict the effectiveness of long-term and large-scale fertilization (Aumont and Bopp, 2006; Blain et al., 2007; Denman, 2008; Pollard et al., 2009). Earlier simplistic global biogeochemical models suggested that massive fertilization could draw down atmospheric CO_2 by as much as 107 ppm in 100 years (Joos et al., 1991; Peng and Broecker, 1991; Sarmiento and Orr, 1991; Kurz and Maier-Reimer, 1993). Recent global models with a more realistic ecosystem and biogeochemical cycles predict values closer to 33 ppm drawdown in atmospheric CO_2. These results suggest that the amount of carbon sequestration resulting from OIF would represent only a modest offset, a contribution less than 10 % for the range of IPCC future emissions scenarios (Aumont and Bopp, 2006; Denman, 2008). Natural OIF experiments also showed much higher carbon sequestration rates than the small-scale OIF experiments (Morris and Charette, 2013), suggesting that there may be scaling or timing issues in the smaller experiments that preclude simple scaling-up as a prediction tool (see discussion in Section 4). For this reason, several commercial companies (e.g., GreenSea Venture and Climos, http://www.greenseaventure.com; http://www.climos.com) have been promoting large-scale commercial OIF experiments as

a climate mitigation strategy and a means to gain carbon credits (Chisholm et al., 2001; Buesseler and Boyd, 2003). However, this effort has not been able to move forward because we have little knowledge about the potential magnitude of possible side effects related to large-scale geoengineering OIF. It remains difficult to extrapolate findings from the small-scale OIF experiments because the environmental/ecosystem side effects from these miniature studies are themselves quite variable and not yet clearly understood. However, presently available studies do indicate that the known side effects from small-scale studies are themselves small-scale. It therefore seems reasonable that we should continue to undertake small-scale studies to better assess these risks and so lay the groundwork for evaluating the potential efficacy and impacts of large-scale OIF as a geoengineering solution to anthropogenic change.

With potential risks and benefits of OIF, there have been legal issues surrounding OIF raised to support the further study and increase understanding of OIF (Williamson et al., 2012). At present, large-scale and/or commercial OIF experiments are banned by international regulation. The international Convention on the Prevention of Marine Pollution by Dumping of Wastes and Other Matter (London Convention, 1972) and Protocol to the London Convention (London Protocol, 1996) placed legal restrictions on dumping of wastes and other matter that cause hazard, harm, and damage in the ocean and/or interfere with the marine environment. In 2007, the London Convention & Protocol (LC/LP) scientific groups released a statement of concern about ocean fertilization and recommended that ocean fertilization activities be evaluated carefully to ensure that such operations were not contrary to the aims of the LC/LP. Under the LC/LP, commercial activities are prohibited, and only 'small-scale' legitimate scientific research in 'coastal waters' is allowed (Resolution LC-LP.1 (2008), 2008). LC/LP also developed an assessment framework for scientific ocean fertilization research to be applied on a case-by-case basis founded on the agreed definition and compliance with the aims and objectives of Resolution LC-LP.1 (2008) (Fig. 9) (Assessment Framework for Scientific Research Involving Ocean Fertilization, 2010). This framework demands preliminary scientific research to get a permission for OIF experimentation as transparent/reasonable scientific rationale/purpose and risk analysis undertaken using parameters such as problem formulation, site selection, exposure assessment, effects assessment, risk characterization, and risk management must be provided (Assessment Framework for Scientific Research Involving Ocean Fertilization, 2010). Monitoring is also required an integral component of all approved (i.e., legitimate) scientific research activity to assess ecological impacts and to review actual versus intended geoengineering benefits (ACE CRC, 2015). In October 2013, LC/LP categorized artificial ocean fertilization as marine geoengineering, thereby prohibiting operational OIF activities, but enabling OIF scientific research that meets the permit conditions through the environmental assessment framework (Resolution LP.4 (8), 2013).

4 Future: Considerations for designing future OIF experiments

Scientific research on OIF has focused on improving our understanding of the effectiveness, capacity, and risks of OIF as an atmospheric CO_2 removal strategy. Although the first OIF experiments took place more than twenty years ago, the legal and economic aspects of such a strategy in terms of international laws of the sea and carbon offset markets are not yet clear (ACE CRC, 2015). It is therefore of paramount importance that future OIF experiments continue to focus on the effectiveness and capacity of OIF as a means of reducing of atmospheric CO_2, but in doing so should carefully consider iron addition method (i.e., 'How'), tracking methods and measurement parameters (i.e., 'What'), locations (i.e., 'Where'), timing (i.e., 'When'), and duration (i.e., 'How long') to build on the results of OIF experiments, develop our understanding of the magnitude and sources of uncertainties, and in so doing build confidence in our ability to reproduce results.

How: The first consideration for a successful OIF experiment lies in the strategy/approach to maintain added iron within the upper mixed layer. During the first OIF experiment, IronEx-1, the patch was fertilized with acidified iron(II) sulfate

according to the target concentrations of 3.6 nM because iron-enrichment bottle incubation experiments performed in deck-board incubators using ocean water suggested maximum phytoplankton growth rates in response to iron additions of 1–2 nM (Fitzwater et al., 1996). However, subject to horizontal dispersion, concentrations of iron added in the open ocean rapidly decreased from 3.6 nM to 0.25 nM in just four days. Further, the magnitude of the open ocean biogeochemical response was less than bottle enrichment experiments suggested (Coale et al., 1998). Seeking to sustain enhanced iron concentrations in patches, since IronEx-2, the technique of applying repeated (2 to 4) iron infusions has been used in all OIF experiments except SEEDS-1 and FeeP (de Baar et al., 2005; Boyd et al., 2007). Like IronEx-1, SOIREE showed that losses in dissolved iron after the first iron infusion rapidly increased due to horizontal dispersion, and also noted loss due to oxidation of the additional iron(II) to iron(III) (Bowie et al., 2001). However, SOIREE demonstrated that four additions of iron with intervals of about 3 days led to a persistent elevation of both dissolved and particulate iron within the mixed layer at the end of the experiment through fast reduction combined with an increase in the concentration of iron-binding ligands after multiple infusions. Both EIFEX and SOFeX-S also found that multiple iron(II) infusions allowed iron to persist in the mixed layer longer than its expected oxidation times. They determined that the relatively low oxidation rates were related to a combination of photochemical production, slow oxidation, and possibly organic complexation (Croot et al., 2008). Blain et al. (2007) explained that the higher carbon sequestration effectiveness of natural OIF experiments compared to artificial OIF experiments partly resulted from the slow and continuous iron addition that occurs in the natural environment. Short-term infusions of large amounts of iron tend to lead the substantial loss of artificially added iron. Therefore, to increase ration of the amount of carbon flux exported to the amount of iron supplied, multiple additions of iron are more efficient.

What: The second consideration for a successful OIF experiment is effective tracing of fertilized patch including detection of carbon sequestration (Buesseler and Boyd, 2003) and monitoring of possible side effects. OIF side effects include emission of climate-relevant gases such as N_2O and DMS that directly contribute to warming and cooling of the environment, respectively (Law, 2008). During IronEx-1, the fertilized patch was subsequently traced with large variety of physical-biogeochemical techniques and parameters such as GPS and ARGO equipped drifting buoys, SF_6, Fv/Fm ratio, pCO_2, and chlorophyll fluorescence using underway sampling systems, and satellite images (Martin et al., 1994; Coale et al., 1998). As IronEx-1 provided potential evidence to support Martin's iron hypothesis by showing an increase in phytoplankton bloom with iron enrichment, many subsequent OIF experiments adopted the tracing methods introduced by IronEx-1, and were similarly able to detect environmental changes through the observation of both physical and biogeochemical parameters before and after iron addition (Martin et al., 1994; Coale et al., 1996; Boyd et al., 2000; Tsuda et al., 2005; Coale et al., 2004; Boyd et al., 2004; Smetacek et al., 2012). Carbon export fluxes can be detected using ^{234}Th, $^{13}C_{org}$, free-drifting sediment traps, beam-transmissometers, and UVPs (Table 5) (Bidigare et al., 1999; Nodder et al., 2001; Boyd et al., 2004; Buesseler et al., 2004; Coale et al., 2004; Aono et al., 2005; Tsuda et al., 2007; Smetacek et al., 2012; Martin et al., 2013). In particular, it is possible to evaluate the temporal evolution of iron-induced export carbon fluxes into deeper waters by applying the thorium deficiency method and sediment trap fluxes that were used during previous OIF experiments (Table 5). Because of their high vertical resolution, the profiling transmissometer, the UVP with its camera that photographs particles, and transmissometers riding on profiling autonomous floats could provide a record of temporal evolution in POC stocks through successive depth layers once calibrated using POC measurements (Smetacek et al., 2012; Martin et al., 2013). Future OIF experiments could benefit from these technological advances so as to more efficiently trace carbon export flux at higher vertical and temporal resolution than has been done in the past. Nevertheless, the application of multiple methods including trap fluxes and ^{234}Th deficiency to provide relatively direct flux estimates combined with autonomous profilers with their higher resolution would produce the best results.

Where: The third consideration for a successful OIF experiment is the location selection. The dominance of diatoms in

phytoplankton communities plays major role in biological pump efficiency because some species of diatom rapidly sink in aggregate formations and have high accumulation rates of heavily silicified frustules (Tréguer et al., 1995). On the other hand, mesozooplankton (i.e., copepods) graze on large diatoms and so are a major limiting factor in diatom production (Coale et al., 2004; Tsuda et al., 2007). Therefore, to obtain the greatest possible carbon export flux in response to iron addition, OIF experiments should be designed in regions with high silicate concentrations and low copepod abundances. In selecting sites for iron fertilization, it is also important to isolate the iron-fertilized patch from the surrounding unfertilized waters to easily and efficiently observe iron-induced changes (Coale et al., 1996). Ocean eddies provide an excellent setting for OIF experimentation as they have physically rotating water column structures, that naturally tend to isolate interior waters from the surrounding waters. Mesoscale eddies range from 25–250 km in diameter and maintain their characteristics for 10–100 days after formation (Morrow and Le Traon, 2012). Eddy centers, in which fertilization is performed, tend to be subject to relatively slow current speeds compared to the surrounding environment and have high vertical coherence, providing ideal conditions for tracing the same water column from the surface to the deep during the experiment (Smetacek and Naqvi, 2008). Iron additions were carried out at the center of eddies in EisenEx, EIFEX, and LOHAFEX conducted in the Southern Ocean (Smetacek, 2001; Smetacek and Naqvi, 2008; Smetacek and Naqvi, 2010; Smetacek et al., 2012). Observations were also made outside the eddy core well away from the iron-fertilized patch to provide similar information about environmental conditions to compare with patch observations. EIFEX showed a clear difference in export carbon flux between waters within the patch and external to the patch (Smetacek et al., 2012). Therefore, finding of an appropriate eddy setting in a study area should be one of the high priority considerations in conducting an OIF experiment (Smetacek and Naqvi, 2008).

When: The fourth consideration for successful OIF experiment is timing including when an experiment starts. Primary production in ocean environment is generally limited by nutrient availability and/or by light availability, often referred to as single- or co-limitation. Primary production in the Southern Ocean, a representative HNLC region, is subject to co-limitation by micro-nutrients (i.e., iron) and light availability (Mitchell et al., 1991). Previous Southern Ocean OIF experiments have been conducted from spring to late summer, and revealed that during this time of year primary production is limited by iron supply rather than light availability (de Baar et al., 2005; Smetacek and Naqvi, 2008; Peloquin et al., 2011). However, the most opportune time, to distinguish phytoplankton blooms increased by iron addition from natural blooms, is during the month of March when natural phytoplankton blooms decline in the Southern Ocean.

How long: The fifth consideration for successful OIF experiment is how long it lasts. Although it has been reported that the periods that phytoplankton blooms have been maintained by OIF have lasted from ~10 to 40 days (Martin et al., 1994; Coale et al., 1996; Boyd et al., 2000; Tsuda et al., 2005; Coale et al., 2004; Boyd et al., 2004; Smetacek et al., 2012), it has also been suggested that most OIF experiments did not cover the full response times from onset to termination (Boyd et al., 2005). For example, SOIREE and SEEDS-1, had relatively short observation periods (~13 days) and saw increasing trends in primary production throughout the experiments (Fig. 10a), suggesting that the observation period should have been extended. Furthermore, after the end of SOIREE, ocean color satellite images showed continued high chlorophyll-a concentrations (~1 mg m^{-3}) in the iron fertilized patch, which was seen as a long ribbon shape that extended some ~150 km for ~46 days; (~7 weeks) after the initial iron addition (Fig. 10b) (Abraham et al., 2000). This result indicates that short experiment periods may not be sufficient for detecting the full influence of artificial iron addition on primary production (Fig. 8b) (Boyd et al., 2000; Tsuda et al., 2003; de Baar et al., 2005). However, SERIES, SEEDS-2, EIFEX, and LOHAFEX did fully monitor all the phases of the phytoplankton bloom from onset to termination. Among OIF experiments, EIFEX, the second-longest at ~39 days, alone observed iron-induced deep export production between 30 and 36 days after iron addition (Fig. 8a and 10a) (Assmy et al., 2013; Smetacek et al., 2012). Furthermore, long-term observation period covering the later stage of bloom development during natural OIF experiments has made it possible to obtain high carbon sequestration effectiveness (Blain et al., 2007; Pollard et

al., 2009). It is therefore important to predict both the necessary time for onset and the time required for the response to run its full course, otherwise it is not possible to quantify the net effect. In addition, to detect the enhancement of the carbon export flux to iron addition, the observation period should last at least 35 to 40 days after iron addition.

In conclusion, to maximize the effectiveness of OIF experiments in the future, we suggest a design that incorporates: ('How') multiple iron additions to 1–2 nM concentration; ('What') multiple means of tracing the fertilized patch including both trap fluxes and/or ^{234}Th deficiency to obtain direct flux estimates and autonomous platforms such as gliders, equipped with biogeochemical sensors, to obtain high vertical resolution, and monitoring side effect such as N_2O and DMS; ('Where') in an eddy structure with high silicate concentration and low copepod abundance; ('When') e.g., March in the Southern Ocean; ('How long') at least >~35 days.

5. Design for a Korean Iron Fertilization Experiment in the Southern Ocean (KIFES)

5.1 Background - Bransfield Basin

The last artificial OIF experiment, LOHAFEX was led by scientists from CSIR-National Institute of Oceanography in Goa, Alfred-Wegener Institute for Polar and Marine Research (AWI) in Bremerhaven, and 5 other nations. The German government suddenly halted LOHAFEX just before the departure of RV Polarstern from the port following protests by NGOs and environmentalists against OIF experimentation due to concern about direct and long-term side effects of artificial iron fertilization on marine ecosystem. To date, the only OIF experiment with scientific and legal review processes was 'LOHAFEX' conducted in the Southern Ocean. Although people are still worried about side effects of OIF and scientists are still curious about the measurable effects of OIF on the ocean environment, there have been no further intensive investigations to fill the gap between supporters and opponents of OIF as a geoengineering approach since LOHAFEX. There are still many unknowns to be investigated about OIF experiments.

The paleoclimate team at Korea Polar Research Institute (KOPRI) found geological evidence of intensive organic carbon burial in the sediments (Yoo et al., 2016), which removes atmospheric CO_2, in the eastern Bransfield Basin on the Antarctic Peninsula. The diatomaceous ooze layer was well preserved in the buried sediments of the Bransfield Basin (Bahk et al., 2003; Kang et al., 2003; Bak et al., 2015), and represents the fast sinking of diatoms within a short time. Scientists at KOPRI suspect that enhancement of the diatom flux may be related to input of bioavailable iron that controls phytoplankton population by allowing efficient use of surface nutrients. In addition, this unique increase in diatom production, the fast sinking rate of the organic matter, and the remarkably well-preserved organic carbon sediments in this area, suggest the existence of a strong 'biological pump (i.e., significant export production)'. This type of 'bottom-up' approach (see potential for a surface source by looking at the sedimentary evidence) has not been considered in the location selection for previous experiments. Therefore, it is expected that OIF in diatom-dominated eastern Bransfield Basin will be effective for carbon export.

A science-oriented iron fertilization project, KIFES (Fig. 11), was launched in 2016 with the research funding supported by the Korean Ministry of Oceans and Fisheries. This project was planned mainly by KOPRI with domestic collaborators (i.e., Incheon National University, Inha University, Pusan National University, Hanyang University, and Yeonsei University) and strengthened by international collaborators (i.e., AWI, Institute of Geological and Nuclear Sciences, MIT-WHOI, University of Otago, University of California at Irvine, McMaster University, University of South Florida, Royal Netherlands Institute for Sea Research, and Dalhousie University). The main purpose of KIFES was (1) to evaluate the effectiveness of artificial OIF in terms of atmospheric carbon sequestration (i.e., effectiveness in export production) in the Southern Ocean, (2) to

determine the environmental conditions that would maximize effectiveness of artificial OIF, and (3) to reveal short- and long-term side effects derived from a small-scale artificial OIF experiment. Unfortunately, KIFES has lost its present funding source. Nevertheless, optimism prevails that alternative funding will be found at a future date and the following section (5.2) is intended to provide a basic set of design guidelines with expectation that an opportunity to move forward with KIFES will occur in near future.

5.2 A plan for the future: KIFES

The KIFES design entails a 5-year project plan. It would model the 'EIFEX' program that found deep carbon by conducting an OIF experiment in an eddy structure. The KIFES project would include a preliminary environmental survey in the eastern Bransfield Basin, a preliminary environmental survey both outside and inside an eddy structure, an OIF experiment, and an assessment of the full KIFES project. In this section, we introduce the major goal, objective, and main tasks of KIFES.

5.2.1 Year one plan

Goal: To gather information about oceanographic conditions in the eastern Bransfield Basin (eBB) including both eddy development and distribution.

Objective: To understand as best we can the physical and biogeochemical oceanography if relevance to the eBB through analysis of earlier data sets and review of published papers.

Main tasks: (1) Database of physical and biogeochemical parameters from previous surveys conducted in the eastern Bransfield Basin; (2) Review of eBB oceanographic conditions using data analysis and references; (3) Design of oceanographic cruise map for the first preliminary eBB survey, based on results from tasks (1) and (2); (4) Analysis of eddy development and distribution using satellite data in the eBB; (5) Preparation of scientific instruments for ocean physical and biogeochemical monitoring; (6) Establishment of an international collaborative OIF network; and (7) KIFES field program proposal preparation for approval of LC/LP.

5.2.2 Year two plan

Goal: First preliminary hydrographic survey to provide a foundational understanding of eBB oceanographic conditions.

Objective: (1) To gain information about oceanographic conditions from in-situ measurements in the eBB; and (2) To provide background information before KIFES experiment.

Main tasks: (1) Using ice breaker RV *ARAON*, field investigation in the eBB of physical and biogeochemical parameters associated with both carbon sequestration as well as OIF side effect (e.g., N_2O), based on the first year task results; and (2) Continued preparation of LC/LP proposal.

5.2.3 Year three plan

Goal: Preliminary hydrographic survey outside/inside eddy structure prior to the KIFES experiment.

Objective: To compare oceanographic conditions outside and inside an eBB eddy structure prior to the KIFES experiment.

Main tasks: (1) Detection of an eBB eddy using observations from acoustic Doppler current profilers and satellites; (2) Intensive physical and biogeochemical field investigation both inside and outside an eddy structure; (3) Assessment of physical and biogeochemical properties outside vs. inside an eddy structure prior to KIFES experiment; and (4) Submission of the LC/LP proposal to obtain approval for the KIFES experiment from International Maritime Organization.

5.2.4 Year four plan

Goal: KIFES – OIF experiment in an eddy structure (Fig. 11).

Objective: To conduct the eBB artificial OIF experiment.

Main tasks: (1) Execution of the KIFES field campaign, a scientific OIF experiment that will survey the region both inside and outside an eddy structure in the eBB employing underway sampling systems (e.g., such as high frequent pCO_2 and Oxygen/Argon ratios), gliders equipped with biogeochemical sensors, sediment traps deployed at multiple depths, sub-bottom profilers, and satellite observations; and (2) Assessment of KIFES carbon sequestration effectiveness and environmental (ocean and atmosphere) side effects.

5.2.5 Year five plan

Goal: Integrated assessment of the KIFES project.

Objective: To evaluate whether small-scale OIF experimentation can be an effective tool for detecting the effectiveness of artificially induced export production.

Main tasks: (1) Submission of the KIFES assessment report; (2) Writing and submission of scientific results to international journals; (3) Collection of feedback about the KIFES project from international scientific/oceanographic communities; and (4) Production of a final artificial OIF experiment summary (including Main tasks 1–3).

5.3 Final Remark

None of the KIFES scientists has any interest in selling carbon credits by conducting OIF experiments. Rather, KIFES interest lies in the detailed investigation of the biogeochemical effects of artificial iron addition in the Southern Ocean and in the OIF evaluation as one possible geo-engineering method that might be used to mitigate the realities of the climate change effects we face. We look to a future where the KIFES project or one like it becomes a reality so that we may work towards providing a clear answer as to whether or not OIF is promising as a geo-engineering solution. The KIFES project would provide fundamental information and guidelines for future OIF experiments in HNLC regions. In particular, the aforementioned risks and side effects of OIF will be thoroughly investigated so as to belay international concern. And lastly, we emphasize that international cooperation is essential for a project as organizationally and scientifically complex as KIFES, which seeks to improve of our outlook for the Earth's future.

6 Summary

To test the Martin's hypothesis, a total 13 artificial OIF experiments for scientific study were conducted in the HNLC

regions during the last 25 years. The biogeochemical responses to OIF experiments were observed in the increases of primary production as a result of drawdowns of macro-nutrients and DIC. In most experiments, dominance of phytoplankton group tended to be shifted from small-sized groups to large-sized groups, resulting in diatom-dominated phytoplankton community. However, the effectiveness in export production enhancing ocean biological pump was not clearly confirmed by the OIF experiments, except in one, EIFEX. Likewise, the possible environmental side effects in response to iron addition, such as production of climate-relevant gases, development of hypoxia/anoxia in water column, and toxic algal blooms, were not fully evaluated due to inconsistent outcomes with large uncertainty depending on OIF experiment conditions and settings. In particular, monitoring of N_2O and DMS must be considered in determining effectiveness of OIF as a geoengineering approach because these potential trace gas emissions can directly and indirectly modify carbon reduction benefits resulting from OIF. Therefore, validation and suitability of artificial OIF for mitigation of rapidly increasing atmospheric CO_2 levels have been debated for three decades. At present, large-scale or commercial OIF experiments are prohibited by international regulation, so small-scale OIF experimentation with scientific purpose is permitted to understand the effectiveness, capacity, and risks of artificial OIF. To maximize effectiveness of OIF, future OIF experiments should be conducted by carefully considering the major factors such as the methods for iron addition, tracking methods, measurement parameters, location, timing, and experiment duration, under international OIF regulations. Finally, we look to a future the KIFES project or one like it becomes a reality so that we may work towards providing a clear answer as to whether or not OIF is promising as a geo-engineering solution.

Acknowledgements: This research was a part of the project titled 'Korean Iron Fertilization Experiment in the Southern Ocean (KOPRI, PM 16060)' funded by the Ministry of Oceans and Fisheries, Korea. Funding for this work was partly provided by Korea Polar Research Institute (KOPRI) project (PE17030). This work was partly supported by the National Research Foundation of Korea (NRF) grant funded by the Korea government (MSIP) (No. 2015R1C1A1A01052051). A. M. Macdonald was supported by NOAA grant #NA11OAR4310063 and internal WHOI funding. K.-T Park was partly supported by MSIP (NRF-2016M1A5A1901769) (KOPRI-PN16081).

References

Abraham, E. R., Law, C. S., Boyd, P. W., Lavender, S. J., Maldonado, M. T., and Bowie, A. R.: Importance of stirring in the development of an iron-fertilized phytoplankton bloom, Nature, 407, 727-730, 2000.

ACE CRC: Position Analysis: Ocean Fertilisation: Science and Policy Issues, ACE CRC, Hobart, 2008.

ACE CRC: Position Analysis: Ocean Fertilisation, ACE CRC, Hobart, 2015.

Anderson, M. A. and Morel, F. M. M.: The influence of aqueous iron chemistry on the uptake of iron by the coastal diatom *Thalassiosira weissflogii*, Limnol. Oceanogr., 27, 789-813, 1982.

Assessment Framework for Scientific Research Involving Ocean Fertilization, LC 32/15, Annex 6, 2010.

Aono, T., Yamada, M., Kudo, I., Imai, K., Nojiri, Y., and Tsuda, A.: Export fluxes of particulate organic carbon estimated from $^{234}Th/^{238}U$ disequilibrium during the Subarctic Pacific Iron Experiment for Ecosystem Dynamics Study (SEEDS 2001), Prog. Oceanogr., 64, 263-282, 2005.

Aramaki, T., Nojiri, Y., and Imai, K.: Behavior of particulate materials during iron fertilization experiments in the Western Subarctic Pacific (SEEDS and SEEDS II), Deep-Sea Res. Pt. II, 56, 2875-2888, 2009.

Assmy, P., Henjes, J., Klaas, C., and Smetacek, V.: Mechanisms determining species dominance in a phytoplankton bloom induced by the iron fertilization experiment EisenEx in the Southern Ocean, Deep-Sea Res. Pt. I, 54, 340-362, 2007.

Assmy, P., Smetacek, V., Montresor, M., Klaas, C., Henjes, J., Strass, V. H., Arrieta, J. M., Bathmann, U., Berg, G. M., Breitbarth, E., Cisewski, B., Friedrichs, L., Fuchs, N., Herndl, G. J., Jansen, S., Krägefsky, S., Latasa, M., Peeken, I., Röttgers, R., Scharek, R., Schüller, S. E., Steigenberger, S., Webb, A., and Wolf-Gladrow, D.: Thick-shelled, grazer-protected diatoms decouple ocean carbon and silicon cycles in the iron-limited Antarctic Circumpolar Current, Proc. Natl. Acad. Sci. USA., 110, 20633-20638, 2013.

Aumont, O. and Bopp, L.: Globalizing results from ocean *in situ* iron fertilization studies, Global Biogeochem. Cycles, 20, GB2017, 2006.

Bahk, J. J., Yoon, H. I., Kim, Y., Kang, C. Y., and Bae, S. H.: Microfabric analysis of laminated diatom ooze (Holocene) from the eastern Bransfield Strait, Antarctic Peninsula, Geosci. J., 7, 135-142, 2003.

Bak, Y.-S., Yoon, H. I., Yoo, K.-C., and Lee, Y.-U.: Diatom succession representing the paleoclimatic change from laminated sediments around Antarctica, J. Korean Earth Sci. Soc., 36, 190-197, 2015.

Bakker, D. C. E., Watson, A. J., and Law, C. S.: Southern Ocean iron enrichment promotes inorganic carbon drawdown, Deep-Sea Res. Pt. II, 48, 2483-2507, 2001.

Bakker, D. C. E., Bozec, Y., Nightingale, P. D., Goldson, L., Messias, M.-J., de Baar, H. J. W., Liddicoat, M., Skjelvan, I., Strass, V., and Watson, A. J.: Iron and mixing affect biological carbon uptake in SOIREE and EisenEx, two Southern Ocean iron fertilisation experiments, Deep-Sea Res. Pt. I, 52, 1001-1019, 2005.

Bange, H. W.: New Directions: The importance of oceanic nitrous oxide emissions, Atmos. Environ., 40, 198-199, 2006.

Barber, R. T. and Hiscock, M. R.: A rising tide lifts all phytoplankton: Growth response of other phytoplankton taxa in diatom-dominated blooms, Global Biogeochem. Cycles, 20, GB4S03, 2006.

Barnola, J. M., Raynaud, D., Korotkevich, Y. S., and Lorius, C.: Vostok ice core provides 160,000-year record of atmospheric CO_2, Nature, 329, 408-414, 1987.

Behrenfeld, M. J., Bale, A. J., Kolber, Z. S., Aiken, J., and Falkowski, P. G.: Confirmation of Iron Limitation of Phytoplankton Photosynthesis in the Equatorial Pacific-Ocean, Nature, 383, 508-511, 1996.

Berg, G. M., Mills, M. M., Long, M. C., Bellerby, R., Strass, V., Savoye, N., Röttgers, R., Croot, P. L., Webb, A., and Arrigo, K. R.: Variation in particulate C and N isotope composition following iron fertilization in two successive phytoplankton communities in the Southern Ocean, Global Biogeochem. Cycles, 25, GB3013, 2011.

Bidigare, R. R., Hanson, K. L., Buesseler, K. O., Wakeham, S. G., Freeman, K. H., Pancost, R. D., Millero, F. J., Steinberg, P., Popp, B. N., Latasa, M., Landry, M. R., and Laws, E. A.: Iron-stimulated changes in ^{13}C fractionation and export by equatorial Pacific phytoplankton: Toward a paleogrowth rate proxy, Paleoceanography, 14, 589-595, 1999.

Bishop, J. K. B., Wood, T. J., Davis, R. E., and Sherman, J. T.: Robotic Observations of Enhanced Carbon Biomass and Export at 55°S During SOFeX, Science, 304, 417-420, 2004.

Blain, S., Queguiner, B., Armand, L., Belviso, S., Bombled, B., Bopp, L., Bowie, A., Brunet, C., Brussaard, C., Carlotti, F., Christaki, U., Corbiere, A., Durand, I., Ebersbach, F., Fuda, J.-L., Garcia, N., Gerringa, L., Griffiths, B., Guigue, C., Guillerm, C., Jacquet, S., Jeandel, C., Laan, P., Lefevre, D., Lo Monaco, C., Malits, A., Mosseri, J., Obernosterer, I., Park, Y.-H., Picheral, M., Pondaven, P., Remenyi, T., Sandroni, V., Sarthou, G., Savoye, N., Scouarnec, L., Souhaut, M., Thuiller, D., Timmermans, K., Trull, T., Uitz, J., van Beek, P., Veldhuis, M., Vincent, D., Viollier, E., Vong, L., and Wagener, T.: Effect of natural iron fertilization on carbon sequestration in the Southern Ocean, Nature, 446, 1070-1074, 2007.

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., and Boyd, P. W.: The fate of added iron during a mesoscale fertilisation experiment in the Southern Ocean, Deep-Sea Res. Pt. II, 48, 2703-2743, 2001.

Boyd, P. W., Watson, A. J., Law, C. S., Abraham, E. R., Trull, T., Murdoch, R., Bakker, D. C. E., Bowie, A. R., Buesseler, K. O., Chang, H., Charette, M., Croot, P., Downing, K., Frew, R., Gall, M., Hadfield, M., Hall, J., Harvey, M., Jameson, G.,

LaRoche, J., Liddicoat, M., Ling, R., Maldonado, M. T., McKay, R. M., Nodder, S., Pickmere, S., Pridmore, R., Rintoul, S., Safi, K., Sutton, P., Strzepek, R., Tanneberger, K., Turner, S., Waite, A., and Zeldis, J.: A mesoscale phytoplankton bloom in the polar Southern Ocean stimulated by iron fertilization, Nature, 407, 695-702, 2000.

Boyd, P. W. and Abraham, E. R.: Iron-mediated changes in phytoplankton photosynthetic competence during SOIREE, Deep-Sea Res. Pt. II, 48, 2529-2550, 2001.

Boyd, P. W. and Law, C. S.: The Southern Ocean Iron RElease Experiment (SOIREE)—introduction and summary, Deep-Sea Res. Pt. II, 48, 2425-2438, 2001.

Boyd, P. W., Law, C. S., Wong, C. S., Nojiri, Y., Tsuda, A., Levasseur, M., Takeda, S., Rivkin, R., Harrison, P. J., Strzepek, R., Gower, J., McKay, R. M., Abraham, E., Arychuk, M., Barwell-Clarke, J., Crawford, W., Crawford, D., Hale, M., Harada, K., Johnson, K., Kiyosawa, H., Kudo, I., Marchetti, A., Miller, W., Needoba, J., Nishioka, J., Ogawa, H., Page, J., Robert, M., Saito, H., Sastri, A., Sherry, N., Soutar, T., Sutherland, N., Taira, Y., Whitney, F., Wong, S.-K. E., and Yoshimura, T.: The decline and fate of an iron-induced subarctic phytoplankton bloom, Nature, 428, 549-553, 2004.

Boyd, P. W., Strzepek, R., Takeda, S., Jackson, G., Wong, C. S., McKay, R. M., Law, C., Kiyosawa, H., Saito, H., Sherry, N., Johnson, K., Gower, J., and Ramaiah, N.: The evolution and termination of an iron-induced mesoscale bloom in the northeast subarctic Pacific, Limnol. Oceanogr., 50, 1872-1886, 2005.

Boyd, P. W., Jickells, T., Law, C. S., Blain, S., Boyle, E. A., Buesseler, K. O., Coale, K. H., Cullen, J. J., de Baar, H. J. W., Follows, M., Harvey, M., Lancelot, C., Levasseur, M., Owens, N. P. J., Pollard, R., Rivkin, R. B., Sarmiento, J., Schoemann, V., Smetacek, V., Takeda, S., Tsuda, A., Turner, S., and Watson, A. J.: Mesoscale Iron Enrichment Experiments 1993-2005: Synthesis and Future Directions, Science, 315, 612-617, 2007.

Broecker, W. S.: Ocean chemistry during glacial time, Geochim. Cosmochim. Acta, 46, 1689-1705, 1982.

Broecker, W. S. and Henderson, G. M.: The sequence of events surrounding Termination II and their implications for the cause of glacial-interglacial CO_2 changes, Paleoceanography, 13, 352-364, 1998.

Buesseler, K. O.: The decoupling of production and particulate export in the surface ocean, Global Biogeochem. Cycles, 12, 297-310, 1998.

Buesseler, K. O. and Boyd, P. W.: Will Ocean Fertilization Work?, Science, 300, 67-68, 2003.

Buesseler, K. O., Andrews, J. E., Pike, S. M., and Charette, M. A.: The Effects of Iron Fertilization on Carbon Sequestration in the Southern Ocean, Science, 304, 414-417, 2004.

Buesseler, K. O., Andrews, J. E., Pike, S. M., Charette, M. A., Goldson, L. E., Brzezinski, M. A., Lance, V. P.: Particle export during the Southern Ocean Iron Experiment (SOFeX), Limnol. Oceanogr., 1, 311-327, 2005.

Cavagna, A. J., Fripiat, F., Dehairs, F., Wolf-Gladrow, D., Cisewski, B., Savoye, N., Andrè, L., and Cardinal, D.: Silicon uptake and supply during a Southern Ocean iron fertilization experiment (EIFEX) tracked by Si isotopes, Limnol. Oceanogr., 56, 147-160, 2011.

Charette, M. A. and Buesseler, K. O.: Does iron fertilization lead to rapid carbon export in the Southern Ocean?, Geochem. Geophys. Geosyst., 1, 2000GC000069, 2000.

Charlson, R. J., Lovelock, J. E., Andreae, M. O., and Warren, S. G.: Oceanic phytoplankton, atmospheric sulphur, cloud albedo and climate, Nature, 326, 655-661, 1987.

Chisholm, S. W., Falkowski, P. G., and Cullen, J. J.: Dis-Crediting Ocean Fertilization, Science, 294, 309-310, 2001.

Cisewski, B., Strass, V. H., Losch, M., and Prandke, H.: Mixed layer analysis of a mesoscale eddy in the Antarctic Polar Front Zone, J. Geophys. Res., 113, C05017, 2008.

Coale, K. H., Johnson, K. S., Fitzwater, S. E., Gordon, R. M., Tanner, S., Chavez, F. P., Ferioli, L., Sakamoto, C., Rogers, P., Millero, F., Steinberg, P., Nightingale, P., Cooper, D., Cochlan, W. P., Landry, M. R., Constantinou, J., Rollwagen, G., Trasvina, A., and Kudela, R.: A massive phytoplankton bloom induced by an ecosystem-scale iron fertilization experiment

in the equatorial Pacific Ocean, Nature, 383, 495-501, 1996.

Coale, K. H., Johnson, K. S., Fitzwater, S. E., Blain, S. P. G., Stanton, T. P., and Coley, T. L.: IronEx-I, an *in situ* iron-enrichment experiment: Experimental design, implementation and results, Deep-Sea Res. Pt. II, 45, 919-945, 1998.

Coale, K. H., Johnson, K. S., Chavez, F. P., Buesseler, K. O., Barber, R. T., Brzezinski, M. A., Cochlan, W. P., Millero, F. J., Falkowski, P. G., Bauer, J. E., Wanninkhof, R. H., Kudela, R. M., Altabet, M. A., Hales, B. E., Takahashi, T., Landry, M. R., Bidigare, R. R., Wang, X., Chase, Z., Strutton, P. G., Friederich, G. E., Gorbunov, M. Y., Lance, V. P., Hilting, A. K., Hiscock, M. R., Demarest, M., Hiscock, W. T., Sullivan, K. F., Tanner, S. J., Gordon, R. M., Hunter, C. N., Elrod, V. A., Fitzwater, S. E., Jones, J. L., Tozzi, S., Koblizek, M., Roberts, A. E., Herndon, J., Brewster, J., Ladizinsky, N., Smith, G., Cooper, D., Timothy, D., Brown, S. L., Selph, K. E., Sheridan, C. C., Twining, B. S., and Johnson, Z. I.: Southern Ocean Iron Enrichment Experiment: Carbon Cycling in High- and Low-Si Waters, Science, 304, 408-414, 2004.

Cooper, D. J., Watson, A. J., and Nightingale, P. D.: Large decrease in ocean-surface CO_2 fugacity in response to *in situ* iron fertilization, Nature, 383, 511-513, 1996.

Croot, P. L., Bluhm, K., Schlosser, C., Streu, P., Breitbarth, E., Frew, R., and Van Ardelan, M.: Regeneration of Fe(II) during EIFeX and SOFeX, Geophys. Res. Lett., 35, L19606, 2008.

Cullen, J. J.: Status of the iron hypothesis after the Open-Ocean Enrichment Experiment, Limnol. Oceanogr., 40, 1336-1343, 1995.

Currie, K. I., Macaskill, B., Reid, M. R., and Law, C. S.: Processes governing the carbon chemistry during the SAGE experiment, Deep-Sea Res. Pt. II, 58, 851-860, 2011.

de Baar, H. J. W., Boyd, P. W., Coale, K. H., Landry, M. R., Tsuda, A., Assmy, P., Bakker, D. C. E., Bozec, Y., Barber, R. T., Brzezinski, M. A., Buesseler, K. O., Boyé, M., Croot, P. L., Gervais, F., Gorbunov, M. Y., Harrison, P. J., Hiscock, W. T., Laan, P., Lancelot, C., Law, C. S., Levasseur, M., Marchetti, A., Millero, F. J., Nishioka, J., Nojiri, Y., van Oijen, T., Riebesell, U., Rijkenberg, M. J. A., Saito, H., Takeda, S., Timmermans, K. R., Veldhuis, M. J. W., Waite, A. M., and Wong, C.-S.: Synthesis of iron fertilization experiments: From the Iron Age in the Age of Enlightenment, J. Geophys. Res., 110, C09S16, 2005.

De La Rocha, C. L.: The Biological Pump, in: Treatise on Geochemistry update, edited by: Holland, H. D. and Turekian, K. K., Elsevier Pergamon, Oxford, 1-29, 2007.

Denman, K. L.: Climate change, ocean processes and ocean iron fertilization, Mar. Ecol.-Prog. Ser., 364, 219–225, 2008.

Duce, R. A. and Tindale, N. W.: Atmospheric transport of iron and its deposition in the ocean, Limnol. Oceanogr., 36, 1715-1726, 1991.

Falkowski, P. G.: Evolution of the nitrogen cycle and its influence on the biological sequestration of CO_2 in the ocean, Nature, 387, 272-275, 1997.

Fitzwater, S. E., Coale, K. H., Gordon, R. M., Johnson, K. S., and Ondrusek, M. E.: Iron deficiency and phytoplankton growth in the equatorial Pacific, Deep-Sea Res. Pt. II, 43, 995-1015, 1996.

Forster, P., Ramaswamy, V., Artaxo, P., Berntsen, T., Betts, R., Fahey, D. W., Haywood, J., Lean, J., Lowe, D. C., Myhre, G., Nganga, J., Prinn, R., Raga, G., Schultz, M., and Van Dorland, R.: Changes in atmospheric constituents and in radiative forcing. Cambridge, United Kingdom, Cambridge University Press, 129-234, 2007.

Frew, R. D., Bowie, A. R., Croot, P. L., and Pickmere S.: Macronutrient and trace-metal geochemistry of an *in situ* iron-induced Southern Ocean bloom, Deep-Sea Res. Pt. II, 48, 2467-2481, 2001.

Frost, B. W.: Phytoplankton bloom on iron rations, Nature, 383, 475-476, 1996.

Fuhrman, J. A. and Capone, D. G.: Possible biogeochemical consequences of ocean fertilization, Limnol. Oceanogr., 36, 1951-1959, 1991.

Gervais, F., Riebesell, U., and Gorbunov, M. Y.: Changes in primary productivity and chlorophyll *a* in response to iron

fertilization in the Southern Polar Frontal Zone, Limnol. Oceanogr., 47, 1324-1335, 2002.

Harvey, M. J., Law, C. S., Smith, M. J., Hall, J. A., Abraham, E. R., Stevens, C. L., Hadfield, M. G., Ho, D. T., Ward, B., Archer, S. D., Cainey, J. M., Currie, K. I., Devries, D., Ellwood, M. J., Hill, P., Jones, G. B., Katz, D., Kuparinen, J., Macaskill, B., Main, W., Marriner, A., McGregor, J., McNeil, C., Minnett, P. J., Nodder, S. D., Peloquin, J., Pickmere, S., Pinkerton, M. H., Safi, K. A., Thompson, R., Walkington, M., Wright, S. W., and Ziolkowski, L. A.: The SOLAS air–sea gas exchange experiment (SAGE) 2004, Deep-Sea Res. Pt. II, 58, 753-763, 2010.

Hiscock, W. T. and Millero, F. J.: Nutrient and carbon parameters during the Southern Ocean iron experiment (SOFeX), Deep-Sea Res. Pt. I, 52, 2086-2108, 2005.

Hoffmann, L. J., Peeken, I., Lochte, K., Assmy, P., and Veldhuis, M.: Different reactions of Southern Ocean phytoplankton size classes to iron fertilization, Limnol. Oceanogr., 51, 1217-1229, 2006.

IPCC: Climate Change: The IPCC Scientific Assessment of Climate Change (Eds. J. T. Houghton, G. J. Jenkins, and J. J. Ephraums), Cambridge Univ. Press, Cambridge, 1990.

IPCC: Climate Change 1992: The Supplementary Report to the IPCC Scientific Assessment (Eds. J. T. Houghton, B. A. Callander, and S. K. Varney), Cambridge Univ. Press, 1992.

IPCC: Climate Change 1995: The Science of Climate Change, Contribution of Working Group 1 to the Second Assessment Report of the Intergovernmental Panel on Climate Change (Eds. J. T. Houghton, L. G. Meira Filho, B. A. Callander, N. Harris, A. Kattenberg, and K. Maskell), Cambridge Univ. Press, Cambridge, 1995.

IPCC: Climate Change 2001: The Scientific Basis, Contribution of Working Group 1 to the Third Assessment Report of the Intergovernmental Panel on Climate Change (Eds. D. J. Griggs, M. Noguer, P. J. van der Linden, X. Dai, K. Maskell, and C. A. Johnson), Cambridge Univ. Press, Cambridge, 2001.

IPCC: Climate Change 2007: The Physical Science Basis, Contribution of Working Group I to the Fourth Assessment Report of the Intergovernmental Panel on Climate Change (Eds. S. Solomon, D. Qin, M. Manning, Z. Chen, M. Marquis, K. B. Averyt, M. Tignor, and H. L. Miller), Cambridge Univ. Press, Cambridge, 2007.

IPCC: Climate Change 2013: The Physical Science Basis, Contribution of Working Group I to the Fifth Assessment Report of the Intergovernmental Panel on Climate Change (Eds. T. F. Stocker, D. Qin, G.-K. Plattner, M. Tignor, S. K. Allen, J. Boschung, A. Nauels, Y. Xia, V. Bex, and P. M. Midgley), Cambridge Univ. Press, Cambridge, 2013.

IPCC: Climate Change 2014: Mitigation of Climate Change, Contribution of Working Group III to the Fifth Assessment Report of the Intergovernmental Panel on Climate Change (Eds. O. Edenhofer, R. Pichs-Madruga, Y. Sokona, E. Farahani, S. Kadner, K. Seyboth, A. Adler, I. Baum, S. Brunner, P. Eickemeier, B. Kriemann, J. Savolainen, S. Schlömer, C. von Stechow, T. Zwickel, and J. C. Minx), Cambridge Univ. Press, Cambridge, 2014.

Jacquet, S. H. M., Savoye, N., Dehairs, F., Strass, V. H., and Cardinal, D.: Mesopelagic carbon remineralization during the European Iron Fertilization Experiment, Global Biogeochem. Cycles, 22, GB1023, 2008.

Jin, X. and Gruber, N.: Offsetting the radiative benefit of ocean iron fertilization by enhancing N_2O emissions, Geophys. Res. Lett., 30, 2249, 2003.

Johnson, K. S. and Karl, D. M.: Is Ocean Fertilization Credible and Creditable?, Science, 296, 467-468, 2002.

Joos, F., Sarmiento, J. L., and Siegenthaler, U.: Estimates of the effect of Southern Ocean iron fertilization on atmospheric CO_2 concentrations, Nature, 349, 772-775, 1991.

Kang, J.-S., Kang, S.-H., Kim, D., and Kim, D.-Y.: Planktonic centric diatom Minidiscus chilensis dominated sediment trap material in eastern Bransfield Strait, Antarctica, Mar. Ecol. Prog. Ser., 255, 93–99, 2003.

Keller, D. P., Feng, E. Y., and Oschlies, A.: Potential climate engineering effectiveness and side effects during a high carbon dioxide-emission scenario, Nature Communications, 5, 3304, 2014.

Kolber, Z. S., Barber, R. T., Coale, K. H., Fitzwateri, S. E., Greene, R. M., Johnson, K. S., Lindley, S., and Falkowski, P. G.:

Iron limitation of phytoplankton photosynthesis in the equatorial Pacific Ocean, Nature, 371, 145-149, 1994.

Koltermann, K. P., Gouretski, V. V., and Jancke, K.: Hydrographic Atlas of the World Ocean Circulation Experiment (WOCE), vol. 3, Atlantic Ocean, edited by M. Sparrow, P. Chapman, and J. Gould, Int. WOCE Proj. Off., Southampton, U. K., 2011.

Kudo, I., Noiri, Y., Cochlan, W. P., Suzuki, K., Aramaki, T., Ono, T., and Nojiri, Y.: Primary productivity, bacterial productivity and nitrogen uptake in response to iron enrichment during the SEEDS II, Deep-Sea Res. Pt. II, 56, 2755-2766, 2009.

Kurz, K. D., Maier-Reimer, E.: Iron fertilization of the austral ocean—the Hamburg model assessment, Global Biogeochem. Cycles, 7, 229–244, 1993.

Landry, M. R., Constantinou, J., Latasa, M., Brown, S. L., Bidigare, R. R., and Ondrusek, M. E.: Biological response to iron fertilization in the eastern equatorial Pacific (IronEx II). III. Dynamics of phytoplankton growth and microzooplankton grazing, Mar. Ecol. Prog. Ser., 201, 57-72, 2000.

Latasa, M., Henjes, J., Scharek, R., Assmy, P., Röttgers, R., and Smetacek, V.: Progressive decoupling between phytoplankton growth and microzooplankton grazing during an iron-induced phytoplankton bloom in the Southern Ocean (EIFEX), Mar. Ecol. Prog. Ser., 513, 39-50, 2014.

Law, C. S., Watson, A. J., Liddicoat, M. I., and Stanton, T.: Sulphur hexafluoride as a tracer of biogeochemical and physical processes in an open-ocean iron fertilisation experiment, Deep-Sea Res. Pt. II, 45, 977-994, 1998.

Law, C. S. and Ling, R. D.: Nitrous oxide flux and response to increased iron availability in the Antarctic Circumpolar Current, Deep-Sea Res. Pt. II, 48, 2509-2527, 2001.

Law, C. S., Crawford, W. R., Smith, M. J., Boyd, P. W., Wong, C. S., Nojiri, Y., Robert, M., Abraham, E. R., Johnson, W. K., Forsland, V., and Arychuk, M.: Patch evolution and the biogeochemical impact of entrainment during an iron fertilisation experiment in the sub-Arctic Pacific, Deep-Sea Res. Pt. II, 53, 2012-2033, 2006.

Law, C. S.: Predicting and monitoring the effects of large-scale ocean iron fertilization on marine trace gas emissions, Mar. Ecol. Prog. Ser., 364, 283-288, 2008.

Law, C. S., Smith, M. J., Stevens, C. L., Abraham, E. R., Ellwood, M. J., Hill, P., Nodder, S., Peloquin, J., Pickmere, S., Safi, K., and Walkington, C. M.: Did dilution limit the phytoplankton response to iron addition in HNLCLSi sub-Antarctic waters during the SAGE experiment?, Deep-Sea Res. Pt. II, 58, 786-799, 2011.

Lawrence, M. G.: Side Effects of Oceanic Iron Fertilization, Science, 297, 1993, 2002.

Lenton, T. M. and Vaughan, N. E.: The radiative forcing potential of different climate geoengineering options, Atmos. Chem. Phys., 9, 5539-5561, 2009.

Leung, D. Y. C., Caramanna, G., and Maroto-Valer, M. M.: An overview of current status of carbon dioxide capture and storage technologies, Renew. Sust. Energ. Rev., 39, 426-443, 2014.

Levasseur, M., Scarratt, M. G., Michaud, S., Merzouk, A., Wong, C. S., Arychuk, M., Richardson, W., Rivkin, R. B., Hale, M., Wong, E., Marchetti, A., and Kiyosawa, H.: DMSP and DMS dynamics during a mesoscale iron fertilization experiment in the Northeast Pacific—Part I: Temporal and vertical distributions, Deep-Sea Res. Pt. II, 53, 2353-2369, 2006.

Liss, P., Chuck, A., Bakker, D., and Turner, S.: Ocean fertilization with iron: effects on climate and air quality, Tellus B, 57, 269-271, 2005.

London Convention: Convention on the Prevention of Marine Pollution by Dumping of Wastes and Other Matter 1972, 1972.

London Protocol: 1996 Protocol to the Convention on the Prevention of Marine Pollution by Dumping of Wastes and Other Matter, 1972, 1996.

Marshall, J. and Speer, K.: Closure of the meridional overturning circulation through Southern Ocean upwelling, Nat. Geosci., 5, 171-180, 2012.

Martin, J. H. and Fitzwater, S. E.: Iron deficiency limits phytoplankton growth in the north-east Pacific subarctic, Nature, 331, 341-343, 1988.

Martin, J. H.: Glacial-interglacial CO_2 change: The Iron Hypothesis, Paleoceanography, 5, 1-13, 1990.

Martin, J. H. and Chisholm, P.: Design for a mesoscale iron enrichment experiment. Woods Hole Oceanographic Institution, U.S JGOFS Planning Report, 15, 1992.

Martin, J. H., Coale, K. H., Johnson, K. S., Fitzwater, S. E., Gordon, R. M., Tanner, S. J., Hunter, C. N., Elrod, V. A., Nowicki, J. L., Coley, T. L., Barber, R. T., Lindley, S., Watson, A. J., Van Scoy, K., Law, C. S., Liddicoat, M. I., Ling, R., Stanton, T., Stockel, J., Collins, C., Anderson, A., Bidigare, R., Ondrusek, M., Latasa, M., Millero, F. J., Lee, K., Yao, W., Zhang, J. Z., Friederich, G., Sakamoto, C., Chavez, F., Buck, K., Kolber, Z., Greene, R., Falkowski, P., Chisholm, S. W., Hoge, F., Swift, R., Yungel, J., Turner, S., Nightingale, P., Hatton, A., Liss, P., and Tindale, N. W.: Testing the iron hypothesis in ecosystems of the equatorial Pacific Ocean, Nature, 371, 123-129, 1994.

Martin, P., van der Loeff, M. R., Cassar, N., Vandromme, P., d'Ovidio, F., Stemmann, L., Rengarajan, R., Soares, M., González, H. E., Ebersbach, F., Lampitt, R. S., Sanders, R., Barnett, B. A., Smetacek, V., and Naqvi, S. W. A.: Iron fertilization enhanced net community production but not downward particle flux during the Southern Ocean iron fertilization experiment LOHAFEX, Global Biogeochem. Cycles, 27, 871-881, 2013.

Matthews, B.: Climate engineering: a critical review of proposals, their scientific and political context, and possible impacts, Compiled for scientists for global responsibility, 1996. ⟨http://records.viu.ca/earles/geol312o/assignments/mitigation.htm⟩.

McElroy, M. B.: Marine biological controls on atmospheric CO_2 and climate, Nature, 302, 328-329, 1983.

Ming, T., de_Richter, R., Liu, W., and Caillol, S.: Fighting global warming by climate engineering: Is the Earth radiation management and the solar radiation management any option for fighting climate change?, Renew. Sust. Energ. Rev., 31, 792-834, 2014.

Mitchell, B. G., Brody, E. A., Holm-Hansen, O., McClain, C., and Bishop, J.: Light limitation of phytoplankton biomass and macronutrient utilization in the Southern Ocean, Limnol. Oceanogr., 36, 1662-1677, 1991.

Moore, J. K., Doney, S. C., Glover, D. M., and Fung, I. Y.: Iron cycling and nutrient-limitation patterns in surface waters of the world ocean, Deep-Sea Res. Pt. II, 49, 463-507, 2002.

Morel, F. M. M. and Price, N. M.: The Biogeochemical Cycles of Trace Metals in the Oceans, Science, 300, 944-947, 2003.

Morris, P. J. and Charette, M. A.: A synthesis of upper ocean carbon and dissolved iron budgets for Southern Ocean natural iron fertilisation studies, Deep-Sea Res. Pt. II, 90, 147-157, 2013.

Morrow, R. and Le Traon, P.-Y.: Recent advances in observing mesoscale ocean dynamics with satellite altimetry, Adv. Space Res., 50, 1062-1076, 2012.

Nagao, I., Hashimoto, S., Suzuki, K., Toda, S., Narita, Y., Tsuda, A., Saito, H., Kudo, I., Kato, S., Kajii, Y., and Uematsu, M.: Responses of DMS in the seawater and atmosphere to iron enrichment in the subarctic western North Pacific (SEEDS-II), Deep-Sea Res. Pt. II, 56, 2899-2917, 2009.

Neftel, A., Oeschger, H., Schwander, J., Stauffer, B., and Zumbrunn, R.: Ice core sample measurements give atmospheric CO_2 content during the past 40,000 yr, Nature, 295, 220-223, 1982.

Nishioka, J., Takeda, S., Baar, H. J. W. D., Croot, P. L., Boye, M., Laan, P., and Timmermans, K. R.: Changes in the concentration of iron in different size fractions during an iron enrichment experiment in the open Southern Ocean, Mar. Chem., 95, 51-63, 2005.

Nodder, S. D., Charette, M. A., Waite, A. M., Trull, T. W., Boyd, P. W., Zeldis, J., and Buesseler, K. O.: Particle transformations and export flux during an *in situ* iron-stimulated algal bloom in the Southern Ocean, Geophys. Res. Lett., 28, 2409-2412, 2001.

Nodder, S. D. and Waite, A. M.: Is Southern Ocean organic carbon and biogenic silica export enhanced by iron-stimulated increases in biological production? Sediment trap results from SOIREE, Deep-Sea Res. Pt. II, 48, 2681-2701, 2001.

Oschlies, A., Koeve, W., Rickels, W., and Rehdanz, K.: Side effects and accounting aspects of hypothetical large-scale Southern

Ocean iron fertilization, Biogeosciences, 7, 4017-4035, 2010.

Peloquin, J., Hall, J., Safi, K., Ellwood, M., Law, C. S., Thompson, K., Kuparinen, J., Harvey, M., and Pickmere, S.: Control of the phytoplankton response during the SAGE experiment: A synthesis, Deep-Sea Res. Pt. II, 58, 824-838, 2011.

Peng, T.-H. and Broecker, W. S.: Factors limiting the reduction of atmospheric CO_2 by iron fertilization, Limnol. Oceanogr., 36, 1919-1927, 1991.

Petit, J. R., Jouzel, J., Raynaud, D., Barkov, N. I., Barnola, J. M., Basile, I., Bender, M., Chappellaz, J., Davis, M., Delaygue, G., Delmotte, M., Kotlyakov, V. M., Legrand, M., Lipenkov, V. Y., Lorius, C., Pepin, L., Ritz, C., Saltzman, E., and Stievenard, M.: Climate and atmospheric history of the past 420,000 years from the Vostok ice core, Antarctica, Nature, 399, 429-436, 1999.

Pollard, R. T., Salter, I., Sanders, R. J., Lucas, M. I., Moore, C. M., Mills, R. A., Statham, P. J., Allen, J. T., Baker, A. R., Bakker, D. C. E., Charette, M. A., Fielding, S., Fones, G. R., French, M., Hickman, A. E., Holland, R. J., Hughes, J. A., Jickells, T. D., Lampitt, R. S., Morris, P. J., Nedelec, F. H., Nielsdottir, M., Planquette, H., Popova, E. E., Poulton, A. J., Read, J. F., Seeyave, S., Smith, T., Stinchcombe, M., Taylor, S., Thomalla, S., Venables, H. J., Williamson, R., and Zubkov, M. V.: Southern Ocean deep-water carbon export enhanced by natural iron fertilization, Nature, 457, 577-580, 2009.

Rees, A. P., Nightingale, P. D., Owens, N. J. P., and Team, P. F.: FeeP—An *in situ* PO_4^{3-} and Fe^{2+} addition experiment to waters of the sub-tropical north-east Atlantic, Geophys. Res. Abstr., 9, 2007.

Resolution LC-LP.1 (2008) on the Regulation of Ocean Fertilization, LC 30/16, Annex 6, 2008.

Resolution LP.4 (8) on the Amendment to the London Protocol to Regulate the Placement of Matter for Ocean Fertilization and Other Marine Geoengineering Activities, LP.8, LC 35/15, Annex 4, Annex 5, 2013.

Sarmiento, J. L. and Orr, J. C.: Three-dimensional simulations of the impact of Southern Ocean nutrient depletion on atmospheric CO_2 and ocean chemistry, Limnol. Oceanogr., 36, 1928-1950, 1991.

Sarmiento, J. L. and Gruber, N.: Ocean Biogeochemical Dynamics, Princeton University Press, 2006.

Sigman, D. M. and Boyle, E. A.: Glacial/interglacial variations in atmospheric carbon dioxide, Nature, 407, 859-869, 2000.

Silver, M. W., Bargu, S., Coale, S. L., Benitez-Nelson, C. R., Garcia, A. C., Roberts, K. J., Sekula-Wood, E., Bruland, K. W., and Coale, K. H.: Toxic diatoms and domoic acid in natural and iron enriched waters of the oceanic Pacific, Proc. Natl. Acad. Sci. USA., 107, 20762-20767, 2010.

Smetacek, V.: EisenEx: International Team Conducts Iron Experiment In Southern Ocean, U.S. JGOFS News, 11, 11–14, 2001.

Smetacek, V., Bathmann, U., and Helmke, E.: The expeditions Antarktis XXI/3-4-5 of the Research Vessel "Polarstern" in 2004. Reports on Polar and Marine Research 500, 2005.

Smetacek, V. and Naqvi, S. W. A.: The next generation of iron fertilization experiments in the Southern Ocean, Philos. Trans. A Math. Phys. Eng. Sci., 366, 3947-3967, 2008.

Smetacek, V. and Naqvi, S. W. A.: The expedition of the research vessel "Polarstern" to the Antarctic in 2009 (ANT-XXV/3 - LOHAFEX), Berichte zur Polar- und Meeresforschung (Reports on Polar and Marine Research). Bremerhaven, Alfred Wegener Institute for Polar and Marine Research, 613, 2010.

Smetacek, V., Klaas, C., Strass, V. H., Assmy, P., Montresor, M., Cisewski, B., Savoye, N., Webb, A., d/'Ovidio, F., Arrieta, J. M., Bathmann, U., Bellerby, R., Berg, G. M., Croot, P., Gonzalez, S., Henjes, J., Herndl, G. J., Hoffmann, L. J., Leach, H., Losch, M., Mills, M. M., Neill, C., Peeken, I., Rottgers, R., Sachs, O., Sauter, E., Schmidt, M. M., Schwarz, J., Terbruggen, A., and Wolf-Gladrow, D.: Deep carbon export from a Southern Ocean iron-fertilized diatom bloom, Nature, 487, 313-319, 2012.

Smetacek, V.: Ocean iron fertilization experiments: The dawn of a new era in applied ocean sciences?, KOPRI, Korea, 2015.

Solomon, S., Garcia, R. R., and Ravishankara, A. R.: On the role of iodine in ozone depletion, J. Geophys. Res., 99, 20491-20499, 1994.

Spolaor, A., Vallelonga, P., Cozzi, G., Gabrieli, J., Varin, C., Kehrwald, N., Zennaro, P., Boutron, C., and Barbante, C.: Iron speciation in aerosol dust influences iron bioavailability over glacial-interglacial timescales, Geophys. Res. Lett., 40, 1618-1623, 2013.

Steinberg, P. A., Millero, F. J., and Zhu, X.: Carbonate system response to iron enrichment, Mar. Chem., 62, 31-43, 1998.

Strong, A.L., Cullen, J. J., and Chisholm, S. W.: Ocean fertilization: Science, policy, and commerce. Oceanography, 22, 236–261, 2009.

Takeda, S. and Tsuda, A.: An *in situ* iron-enrichment experiment in the western subarctic Pacific (SEEDS): Introduction and summary, Prog. Oceanogr., 64, 95-109, 2005.

Talley, L. D.: Hydrographic Atlas of the World Ocean Circulation Experiment (WOCE), Volume 2: Pacific Ocean (eds. Sparrow, M., Chapman, P., and Gould, J.), International WOCE Project Office, Southampton, U.K., ISBN 0-904175-54-5, 2007.

Thiele, S., Fuchs, B. M., Ramaiah, N., and Amann, R.: Microbial Community Response during the Iron Fertilization Experiment LOHAFEX, Appl. Environ. Microbiol., 78, 8803-8812, 2012.

Tréguer, P., Nelson, D. M., Van Bennekom, A. J., DeMaster, D. J., Leynaert, A., and Quéguiner, B.: The Silica Balance in the World Ocean: A Reestimate, Science, 268, 375-379, 1995.

Trick, C. G., Bill, B. D., Cochlan, W. P., Wells, M. L., Trainer, V. L., and Pickell, L. D.: Iron enrichment stimulates toxic diatom production in high-nitrate, low-chlorophyll areas, Proc. Natl. Acad. Sci. USA., 107, 5887-5892, 2010.

Trull, T., and Armand, L. K.: Insights into Southern Ocean carbon export from the $\delta^{13}C$ of particles and dissolved inorganic carbon during the SOIREE iron release experiment, Deep-Sea Res. Pt. II, 48, 2655-2680, 2001.

Tsuda, A., Takeda, S., Saito, H., Nishioka, J., Nojiri, Y., Kudo, I., Kiyosawa, H., Shiomoto, A., Imai, K., Ono, T., Shimamoto, A., Tsumune, D., Yoshimura, T., Aono, T., Hinuma, A., Kinugasa, M., Suzuki, K., Sohrin, Y., Noiri, Y., Tani, H., Deguchi, Y., Tsurushima, N., Ogawa, H., Fukami, K., Kuma, K., and Saino, T.: A Mesoscale Iron Enrichment in the Western Subarctic Pacific Induces a Large Centric Diatom Bloom, Science, 300, 958-961, 2003.

Tsuda, A., Kiyosawa, H., Kuwata, A., Mochizuki, M., Shiga, N., Saito, H., Chiba, S., Imai, K., Nishioka, J., and Ono, T.: Responses of diatoms to iron-enrichment (SEEDS) in the western subarctic Pacific, temporal and spatial comparisons, Prog. Oceanogr., 64, 189-205, 2005.

Tsuda, A., Takeda, S., Saito, H., Nishioka, J., Kudo, I., Nojiri, Y., Suzuki, K., Uematsu, M., Wells, M. L., Tsumune, D., Yoshimura, T., Aono, T., Aramaki, T., Cochlan, W. P., Hayakawa, M., Imai, K., Isada, T., Iwamoto, Y., Johnson, W. K., Kameyama, S., Kato, S., Kiyosawa, H., Kondo, Y., Levasseur, M., Machida, R. J., Nagao, I., Nakagawa, F., Nakanishi, T., Nakatsuka, S., Narita, A., Noiri, Y., Obata, H., Ogawa, H., Oguma, K., Ono, T., Sakuragi, T., Sasakawa, M., Sato, M., Shimamoto, A., Takata, H., Trick, C. G., Watanabe, Y. W., Wong, C. S., and Yoshie, N.: Evidence for the grazing hypothesis: Grazing reduces phytoplankton responses of the HNLC ecosystem to iron enrichment in the western subarctic pacific (SEEDS II), J. Oceanogr., 63, 983-994, 2007.

Tsumune, D., Nishioka, J., Shimamoto, A., Takeda, S., and Tsuda, A.: Physical behavior of the SEEDS iron-fertilized patch by sulphur hexafluoride tracer release, Prog. Oceanogr., 64, 111-127, 2005.

Tsumune, D., Nishioka, J., Shimamoto, A., Watanabe, Y. W., Aramaki, T., Nojiri, Y., Takeda, S., Tsuda, A., and Tsubono, T.: Physical behaviors of the iron-fertilized patch in SEEDS II, Deep-Sea Res. Pt. II, 56, 2948-2957, 2009.

Turner, S. M., Nightingale, P. D., Spokes, L. J., Liddicoat, M. I., and Liss, P. S.: Increased dimethyl sulphide concentrations in sea water from *in situ* iron enrichment, Nature, 383, 513-517, 1996.

Turner, S. M., Harvey, M. J., Law, C. S., Nightingale, P. D., and Liss, P. S.: Iron-induced changes in oceanic sulfur biogeochemistry, Geophys. Res. Lett., 31, L14307, 2004.

Vaughan, N. E. and Lenton, T. M.: A review of climate geoengineering proposals, Clim. Change, 109, 745-790, 2011.

Volk, T. and Hoffert, M. I.: Ocean Carbon Pumps: Analysis of Relative Strengths and Efficiencies in Ocean-Driven

Atmospheric CO_2 Changes, in: The Carbon Cycle and Atmospheric CO2: Natural Variations Archean to Present, edited, Geophys. Monogr. Ser, 99-110, 1985.

Waite, A. M. and Nodder, S. D.: The effect of *in situ* iron addition on the sinking rates and export flux of Southern Ocean diatoms, Deep-Sea Res. Pt. II, 48, 2635-2654, 2001.

Walter, S., Peeken, I., Lochte, K., Webb, A., and Bange, H. W.: Nitrous oxide measurements during EIFEX, the European Iron Fertilization Experiment in the subpolar South Atlantic Ocean, Geophys. Res. Lett., 32, L23613, 2005.

Wanninkhof, R. and Thoning, K.: Measurement of fugacity of CO_2 in surface water using continuous and discrete sampling methods, Mar. Chem., 44, 189-204, 1993.

Watson, A. J., Bakker, D. C. E., Ridgwell, A. J., Boyd, P. W., and Law, C. S.: Effect of iron supply on Southern Ocean CO_2 uptake and implications for glacial atmospheric CO_2, Nature, 407, 730-733, 2000.

Westberry, T. K., Behrenfeld, M. J., Milligan, A. J., and Doney, S. C.: Retrospective satellite ocean color analysis of purposeful and natural ocean iron fertilization, Deep-Sea Res. Pt. I, 73, 1-16, 2013.

Williamson, P., Wallace, D. W. R., Law, C. S., Boyd, P. W., Collos, Y., Croot, P., Denman, K., Riebesell, U., Takeda, S., and Vivian, C.: Ocean fertilization for geoengineering: A review of effectiveness, environmental impacts and emerging governance, Process Saf. Environ. Protect., 90, 475-488, 2012.

Wingenter, O. W., Haase, K. B., Strutton, P., Friederich, G., Meinardi, S., Blake, D. R., and Rowland, F. S.: Changing concentrations of CO, CH_4, C_5H_8, CH_3Br, CH_3I, and dimethyl sulfide during the Southern Ocean Iron Enrichment Experiments, Proc. Nat. Acad. Sci. U.S.A., 101, 8537-8541, 2004.

Wingenter, O. W., Elliot, S. M., and Blake, D. R.: New Directions: Enhancing the natural sulfur cycle to slow global warming, Atmos. Environ., 41, 7373-7375, 2007.

Wong, C. S., Timothy, D. A., Law, C. S., Nojiri, Y., Xie, L., Wong, S.-K. E., and Page, J. S.: Carbon distribution and fluxes during the SERIES iron fertilization experiment with special reference to the fugacity of carbon dioxide (fCO_2), Deep-Sea Res. Pt. II, 53, 2053-2074, 2006.

Yoo, K.-C., Yoon, H. I., Lee, J. I., Lee, M. K., and Kim, K.: Spectral analysis of time series in marine laminated sediments of the eastern Basin of Bransfield Strait (Antarctic Peninsula), 2016 SCAR Meeting, Kuala Lumpur, p984, 2016.

Zhuang, G., Yi, Z., Duce, R. A., and Brown, P. R.: Link between iron and sulphur cycles suggested by detection of Fe(II) in remote marine aerosols, Nature, 355, 537-539, 1992.

Zhuang, G. and Duce, R. A.: The adsorption of dissolved iron on marine aerosol particles in surface waters of the open ocean, Deep-Sea Res. Pt. I, 40, 1413-1429, 1993.

Table 1. Summary of OIF experiments; time, location, research vessel, amounts of Fe addition (day of Fe addition from the beginning of OIF experiment), background Fe concentrations, Fe concentrations after Fe additions, tracer, patch size fertilized by first Fe addition, experiments periods, and characteristics of study regions.

	Experiment	Time	Location	Research Vessel	Fe (kg) (day)	Initial Fe (nM)	After Fe (nM)	Tracer	Patch size (km^2)	Period (days)	Region
1	IronEx-1	Oct 1993	5° S, 90° W Equatorial Pacific	RV *Columbus Iselin*	①450 (0)	0.06	3.60	SF_6	64	10	HNLC
2	IronEx-2	May 1995	3.5° S, 104° W Equatorial Pacific	RV *Melville*	①225 (0) ②112 (3) ③112 (7)	0.02	2.00 1.00 1.00	SF_6	72	17	HNLC
3	SOIREE	Feb 1999	61° S 141° E Southern Ocean- Australasian-Pacific sector	RV *Astrolab*	①768 (0) ②312 (3) ③312 (5) ④353 (7)	0.08	3.80 2.60 2.60 2.50	SF_6	50	13	HNLC
4	EisenEx	Nov 2000	48° S, 21° E Southern Ocean- Atlantic sector	RV *Polarstern*	①780 (0) ②780 (7) ③780 (16)	0.06	2.00	SF_6	50	23	HNLC
5	SEEDS-1	Jul–Aug 2001	48.5° N, 165° E North Pacific- Western subarctic gyre	RV *Kaiyo-Maru*	①350 (0)	0.05	2.90	SF_6	80	13	HNLC
6	SOFeX-N	Jan–Feb 2002	56.23° S, 172° W Southern Ocean- Atlantic sector	RV *Revelle* RV *Melville*	①631 (0) ②631 (5) ③450 (30)		1.20	SF_6	225	40	HNLCLSi
7	SOFeX-S	Jan–Feb 2002	66.45° S, 171.8° W Southern Ocean- Atlantic sector	RV *Revelle* RV *Melville* RV *Polar star*	①315 (0) ②315 (4) ③315 (7) ④315 (11)		0.70	SF_6	225	28	HNLC
8	SERIES	Jul-Aug 2002	50.14° N, 144.75° W North Pacific- Eastern subarctic Pacific	RV *John P. Tully* RV *El Puma* RV *Kaiyo Maru*	①315 (0) ②315 (6)	<0.10	2.00 0.60	SF_6	77	25	HNLC

To be continued

	Experiment	Time	Location	Research Vessel	Fe (kg) (day)	Initial Fe (nM)	After Fe (nM)	Tracer	Patch size (km^2)	Period (days)	Region
9	EIFEX	Feb–Mar 2004	50° S, 2° E Southern Ocean-Atlantic sector	RV *Polarstern*	①1406 (0) ②1406 (13)	0.20	1.50 0.34		167	39	HNLC
10	FeeP	Apr–May 2004	27.5° N 22.5° W North Atlantic-Subtropical north-east Atlantic	RV *Charles Darwin* RV *Poseidon*	①1840 (0)	0.20	3.00	SF_6	25	21	LNLC
11	SAGE	Mar–Apr 2004	46.7° S 172.5° E Southern Ocean-Southeast of New Zealand	RV *Tangaroa*	①265 (0) ②265 (6) ③265 (9) ④265 (12)	0.09	3.03 1.59 0.55 1.01	SF_6	36	15	HNLCLSi
12	SEEDS-2	Jul–Aug 2004	48° N, 166° E North Pacific-Western subarctic gyre	RV *Hakuho-Maru* RV *Kilo-Moana*	①332 (0) ②159 (6)	0.17	1.38	SF_6	64	26	HNLC
13	LOHAFEX	Jan–Mar 2009	48° S, 15° W Southern Ocean-Atlantic sector	RV *Polarstern*	①2000 (0) ②2000 (21)		2.00	SF_6	300	40	HNLCLSi
1	CROZEX*	Nov 2004–Jan 2005	44° S, 50° E Southern Ocean-South of sub-Antarctic Front	RV *Discovery*		0.04	0.55				HNLC
2	KEOPS*	Jan–Feb 2005	50° S, 73° E Southern Ocean-South of Polar Front	RV *Marion Dufresne*							HNLC

* Natural OIF experiments

Sources are as follows: Martin et al., 1994; Coale et al., 1996; Coale et al., 1998; Boyd et al., 2000; Gervais et al., 2002; Tsuda et al., 2003; Boyd et al., 2004; Bakker et al., 2005; Boyd et al., 2005; Coale et al., 2004; de Baar et al., 2005; Hiscock and Millero, 2005; Nishioka et al., 2005; Tsuda et al., 2005; Tsumune et al., 2005; Boyd et al., 2007; Rees et al., 2007; Tsuda et al., 2007; Harvey et al., 2010; Law et al., 2011; Smetacek et al., 2012; Martin et al., 2013.

Table 2. Summary of OIF experiments; objective, significant results, and limitation

	Experiment	Objective	Significant results	Limitation
1	IronEx-1	* To test the iron hypothesis	* Increased phytoplankton production in response to added iron	* Relatively small increase in primary production * Rapid loss of iron within mixed layer
2	IronEx-2	* To test four hypotheses which were raised by lack of biogeochemical response upon iron addition during IronEx-1	* A massive phytoplankton bloom response to iron addition * Significant drawdown of pCO_2 * Diatom-dominated community	* Limited information on export flux * No observation of the fate of the bloom
3	SOIREE	* To confirm of iron limitation in phytoplankton growth in the Southern Ocean * To understand downward fluxes	* Iron-induced decreases in pCO_2 * Diatom-dominated bloom with low mesozooplankton grazing	* No increase in export flux * Unknown remineralization
4	EisenEx	* To artificially stimulate an airborne 'dust' episode in the Southern Ocean with OIF * To study the response of phytoplankton bloom under limited light condition	* Maintenance of iron-induced phytoplankton bloom in austral spring	* No difference in POC flux between inside patch and outside patch in the eddy
5	SEEDS-1	* To test the iron hypothesis in the subarctic North Pacific Ocean * To examine the changes in the species composition and the specific growth responses of key diatom species	* Significant drawdown in pCO_2 * Shifting from oceanic diatoms to neritic centric diatom	* No increase in export flux * Unknown trophic interactions
6	SOFeX-N	* To investigate the effects of iron enrichment in regions with low silicate concentrations	* Enhanced growth of diatom groups * pCO_2 depressed by increased primary production	* Entrainment of dissolved silicate into the patch by physical mixing
7	SOFeX-S	* To investigate the effects of iron enrichment in regions with high silicate concentrations	*Increased POC flux out of the mixed layer	*Small POC flux relative to natural blooms
8	SERIES	* To detect the decline and fate of an iron-fertilized diatom bloom * To measure the response of trophic interactions to iron addition * To measure carbon flux out of the surface layer	* Decline and fate of iron-added bloom * Bacterial remineralization and mesozooplankton grazing accounting as main process of POC decrease	* Inefficient transfer of iron-increased POC below the permanent thermocline

To be continued

	Experiment	Objective	Significant results	Limitation
9	EIFEX	* To find out the growth and demise phase of phytoplankton bloom in fertilized patch * To confirm the second condition of the iron hypothesis	* Significant enhancement of deep carbon export (>3000 m) * The occurrence of rapidly sinking large aggregate	
10	FeeP	* To understand phosphate and iron limitation in biological activity	*Increased *Trichodesmium*	* No overall shift in phytoplankton community
11	SAGE	* To understand gas transfer processes and influence of OIF on biologically driven gas exchange	*Successful measurement of gas exchange at strong wind speeds	* No phytoplankton bloom * Increase in pCO_2 due to physical mixing
12	SEEDS-2	* To determine the fate of an iron-stimulated diatom bloom * To verify the vertical export carbon flux	* Higher abundance of mesozooplankton (copepod) during bloom-development phase	* No extensive diatom bloom
13	LOHAFEX	* To investigate the fate of iron fertilized bloom biomass related to heterotrophs and export flux under silicate-limiting conditions	* Recycled carbon by grazing and microbial food web in low silicate waters	* Lack of fertilization-induced export due to silicon limitation and bacterial remineralization

Sources are as follows: Martin et al., 1994; Coale et al., 1996; Coale et al., 1998; Bidigare et al., 1999; Boyd et al., 2000; Charette and Buesseler, 2000; Gervais et al., 2002; Tsuda et al., 2003; Boyd et al., 2004; Coale et al., 2004; Bakker et al., 2005; Boyd et al., 2005; de Baar et al., 2005; Hiscock and Millero, 2005; Nishioka et al., 2005; Tsuda et al., 2005; Tsumune et al., 2005; Boyd et al., 2007; Rees et al., 2007; Tsuda et al., 2007; Harvey et al., 2010; Law et al., 2011; Smetacek et al., 2012; Martin et al., 2013

Table 3. Changes of chemical parameters from initial to after concentrations by OIF experiments. Note that *Δ[X] represents changes in concentrations (i.e., $[X]_{post\text{-}fertilization} - [X]_{pre\text{-}fertilization}$).

	Experiment	Initial NO_3 (μM)	*ΔNO_3 (μM)	Initial PO_4 (μM)	*ΔPO_4 (μM)	Initial Si (μM)	*ΔSi (μM)	Initial pCO_2 (ppm)	*ΔpCO_2 (ppm)	Initial DIC (μM)	*ΔDIC (μM)
1	IronEx-1	10.8	-0.70	0.92	-0.02	3.90	-0.02	471	-13.0	2044	-6.00
2	IronEx-2	10.4	-4.00	0.80	-0.25	5.10	-4.00	538	-73.0	2051	-27.0
3	SOIREE	25.0	-3.00	1.50	-0.24	10.0	-3.00	350	-(38.0–32.0)		-(18.0–15.0)
4	EisenEx	22.0	-1.00	1.60	-0.10	10.0	0	360	-(20–18)		-(15.0–12.0)
5	SEEDS-1	18.5	-15.8			31.8	-26.8	390	-130		-58.0
6	SOFeX-N	21.9	-1.40	1.40	-0.09	2.50	-1.10	367	-26	2109	-14.0
7	SOFeX-S	26.3	-3.50	1.87	-0.21	62.8	-4.00	365	-36	2176	-21.0
8	SERIES	10.0–12.0	-(9.00–7.00)	>1.00	-0.50	14.0–16.0	-(14.0–12.0)	350	-85	2030	-37.0
9	EIFEX	25.0	-1.50	1.80	-0.30	19.0	-11.0	360	-30	2135	-13.5
10	FeeP	<0.01		0.01							<-1.00
11	SAGE	7.90–10.3	1.50–3.90	0.62–0.85		0.83–0.97		330	8.00	2057	25.0
12	SEEDS-2	18.4	-5.72			36.1		370	-6.00		
13	LOHAFEX	20.0	-2.50	1.30	-0.20	0.50–1.40		357.5	-(15–7)		

Sources are as follow: Martin et al., 1994; Steinberg et al., 1998; Boyd et al., 2000; Bakker et al., 2001; Frew et al., 2001; Bakker et al., 2005; Hiscock and Millero, 2005; Smetacek et al., 2005; Takeda and Tsuda, 2005; Wong et al., 2006; Boyd et al. 2007; Tsumune et al., 2009; Harvey et al., 2010; Smetacek and Naqvi, 2010; Berg et al., 2011; Currie et al., 2011; Law et al., 2011.

Table 4. Changes of biological parameters from initial to after (maximum) concentrations by OIF experiments. Note that *PP (mg C m^{-2} d^{-1}) was estimated by multiplying PP (mg C m^{-3} d^{-1}) with mixed layer depth (m).

	Experiment	Initial Fv/Fm	Maximum Fv/Fm	Initial Chlorophyll (mg m^{-3})	Maximum Chlorophyll (mg m^{-3})	Initial PP (mg C m^{-2} d^{-1})	Maximum PP (mg C m^{-2} d^{-1})
1	IronEx-1	0.30	0.60	0.24	0.65	300–450*	805–1330*
2	IronEx-2	0.25	0.50	0.15–0.20	4.00	630	2430
3	SOIREE	0.22	0.65	0.25	2.00	120	1300
4	EisenEx	0.30	0.56	0.50	2.50	130–220	790
5	SEEDS-1	0.19	0.31	0.80–0.90	21.8	420	1670
6	SOFeX-N	0.20	0.5	0.15	2.60	144	1500
7	SOFeX-S	0.25	0.65	0.30	3.80	216	972
8	SERIES	0.24	0.55	0.35	5.00	300	2000
9	EIFEX	0.28	0.6	0.70	3.16	750	1500
10	FeeP			0.04	0.07		
11	SAGE	0.27	0.61	0.63	1.33	540	900
12	SEEDS-2	0.29	0.40	0.80	2.48	390	1000
13	LOHAFEX	0.33	0.40–0.50	0.50	1.25	960	1560

Sources are as follow: Kolber et al., 1994; Martin et al., 1994; Behrenfeld et al., 1996; Steinberg et al., 1998; Boyd et al., 2000; Boyd and Law, 2001; Gervais et al., 2002; Coale et al., 2004; Boyd et al., 2005; de Baar et al., 2005; Takeda and Tsuda, 2005; Tsuda et al., 2005; Assmy et al., 2007; Boyd et al., 2007; Tsuda et al., 2007; Kudo et al., 2009; Harvey et al., 2010; Berg et al., 2011; Currie et al., 2011; Peloquin et al., 2011; Smetacek et al., 2012; Thiele et al., 2012; Martin et al., 2013; Latasa et al., 2014.

Table 5. Export flux (mg C m^{-2} d^{-1}); initial and maximum values in patch at each depth (measurement day from the beginning of OIF experiment), initial and maximum values outside patch at each depth (measurement day from the beginning of OIF experiment), measurement depth, measurement method, and method description to detect enhanced export production.

	Experiment	In patch Initial (day)	In patch Maximum (day)	Outside Initial (day)	Outside Maximum (day)	Depth (m)	Method	Method description
1	IronEx-1							
2	IronEx-2	84 (0)	600 (7)			25	^{234}Th	*Water column at 0, 25 m (^{234}Th, $^{13}C_{org}$)
3	SOIREE	146/73 (0)	193/74 (11)	146/73 (0)	78/38 (11)	110/310	^{234}Th, trap	*Water column in the upper 100 m (^{234}Th, $^{13}C_{org}$) *Free-drifting sediment traps at 110, 310 m (^{234}Th, $^{13}C_{org}$, total mass, POC, biogenic silica (BSi), particulate organic nitrogen (PON))
4	EisenEx							*Transmissometer *Water column (^{234}Th)
5	SEEDS-1	374/166 (2)	1000/140 (13)			50/200	^{234}Th, trap	*Sediment trap at 20, 500, 3500 m (POC) *Water column in the upper 200 m (POC, PON, BSi, ^{234}Th) *Drifting sediment trap at 40, 60, 100, 200 m (POC, PON, BSi, ^{234}Th)
6	SOFeX-N							*Autonomous floats profiling transmissometer *Water column (^{234}Th, POC)
7	SOFeX-S	36/19 (6)	112/142 (27)	48/38 (7)	49/56 (27)	50/100	^{234}Th	*Water column at 25, 50, 75, 100, 125 m (^{234}Th, POC, PON, BSi)
8	SERIES	120/48 (3)	480/192 (24)	192 (3)	139 (15)	50/100	^{234}Th, trap	*Water column in the upper 110 m (POC, BSi) *Free-drifting sediment traps at 100 and 300 m (POC, BSi, ^{234}Th)
9	EIFEX	340 (0)	1692 (32)	396 (0)	516 (32)	100	^{234}Th	*Transmissometer *Sediment trap in the 1000 m upper layer (BSi, POC, PON, ^{234}Th, $^{13}C_{org}$, PON, stable nitrogen isotope of PON ($^{15}N_{PON}$))
10	FeeP							
11	SAGE							*Free-drifting sediment traps at 80, 300 m (POC, PON, BSi) *Particle camera deployments
12	SEEDS-2	290/316 (1-4)	580/336 (19-22)	299/212 (1-8)	509/204 (18-31)	40/100	Trap	*Water column in the upper 100 m (POC, PON, BSi, $^{13}C_{org}$) *Drifting sediment trap at 40, 70, 100 m (POC, PON, BSi, $^{13}C_{org}$)
13	LOHAFEX	60 (0)	94 (23)	78 (4)	97 (25)	100	^{234}Th	*Neutrally buoyant sediment traps at 200, 450 m (POC, PON, ^{234}Th) *Underwater video profiler *Water column in the upper 100 m (POC, PON, ^{234}Th)

Sources are as follow: Bidigare et al., 1999; Nodder et al., 2001; Nodder and Waite, 2001; Trull and Armand, 2001; Waite and Nodder, 2001; Bishop et al., 2004; Boyd et al., 2004; Buesseler et al., 2004; Coale et al., 2004; Aono et al., 2005; Takeda and Tsuda, 2005; Tsuda et al., 2007; Jacquet et al., 2008; Aramaki et al., 2009; Berg et al., 2011; Peloquin et al., 2011; Smetacek et al., 2012; Martin et al., 2013.

Figure Captions

Fig. 1. Diagram showing the monthly atmospheric CO_2 concentrations (ppm) (blue) according to the Mauna Loa Observatory, Hawaii (http://www.esrl.noaa.gov/gmd/ccgg/trends/data.html), global monthly land surface air and sea surface temperature anomalies (°C) (red) (http://data.giss.nasa.gov/gistemp/), and pH (green) measured at station ALOHA in the central Pacific (http://hahana.soest.hawaii.edu/hot/products/HOT_surface_CO2.txt). The data values represent moving average values for 12 months and shading indicates the standard deviation of 12 months.

Fig. 2. Schematic representation of several proposed climate-engineering methods (modified from Matthews, 1996).

Fig. 3. The iron hypothesis as suggested by Martin (1990). (a) Efficiency of the biological pump under normal conditions and (b) efficiency of the biological pump as a result of Fe enrichment. DIC is dissolved inorganic carbon and OM is organic matter (modified from Sarmiento and Gruber, 2006).

Fig. 4. Global annual distribution of surface (a) chlorophyll concentrations (mg m^{-3}), (b) nitrate concentrations (μM), and (c) silicate concentrations (μM). Chlorophyll-a concentration distribution represents the Aqua MODIS chlorophyll-a composite from July 2002 to February 2016 (http://oceancolor.gsfc.nasa.gov/cgi/l3), while the nitrate and silicate were plotted by Ocean Data View program (https://odv.awi.de) using the World Ocean Atlas 2013 dataset (https://odv.awi.de/en/data/ocean/world_ocean_atlas_2013). White circles indicate the locations of 13 artificial OIF experiments and black triangles indicate the locations of natural OIF experiments. Note that the numbers indicate the order of experiments (see Table 1).

Fig. 5. Photographs of iron the addition procedure. Panels a-e taken during EIFEX and LOHAFEX: (a) Iron (II) sulfate bags, (b) the funnel where iron and hydrochloric acid were poured, (c) tank system for mixing with Iron(II) sulfate, hydrochloric acid, and seawater, (d) outlet pipe connected with tank system, (e) pumping iron into prop wash during EIFEX (Smetacek, 2015).

Fig. 6. (a) Patch size (km^2) for first Fe addition (blue bar) and maximum patch size (sky blue bar) during OIF experiments. (b) Amounts (kg) of first Fe addition (blue bar) and total Fe addition (sky blue bar). (c) Minimum (blue bar) and maximum (sky blue bar) mixed layer depth (m). (d) Average sea surface temperature (°C). Initial (e) nitrate concentrations (μM), (f) silicate concentrations (μM), (g) Fv/Fm ratio, and (h) chlorophyll-a concentrations (mg m^{-3}) before iron addition. Note that the numbers indicate the order of experiments (see Table 1). Sources are as follows: Kolber et al., (1994); Martin et al., (1994); Behrenfeld et al., (1996); Coale et al., (1996); Steinberg et al., (1998); Boyd et al., (2000); Boyd and Law, (2001); Gervais et al., (2002); Coale et al., (2004); Boyd et al., (2004); Boyd et al., (2005); de Baar et al., (2005); Hiscock and Millero, (2005); Takeda and Tsuda, (2005); Tsuda et al., (2005); Assmy et al., (2007); Boyd et al., (2007); Tsuda et al., (2007); Harvey et al., (2010); Berg et al., (2011); Law et al., (2011); Peloquin et al., (2011); Smetacek et al., (2012); Thiele et al., (2012); Martin et al., (2013); Latasa et al., (2014).

Fig. 7. (a) Initial (coral bar) and maximum (light coral bar) Fv/Fm ratio during OIF experiments. (b) Changes in nitrate concentrations (ΔNitrate = $[NO_3^-]_{post\text{-}fertilization} - [NO_3^-]_{pre\text{-}fertilization}$; μM). (c) Initial (coral bar) and maximum (light coral bar) chlorophyll-a concentrations (mg m^{-3}). (d) Distributions of chlorophyll-a concentrations (mg m^{-3}) at ~28 days after iron addition in the SOFeX-N and at ~20 days in the SOFeX-S. White dotted box indicates phytoplankton bloom during OIF experiments. Changes in (e) primary productivity ($\Delta PP = [PP]_{post\text{-}fertilization} - [PP]_{pre\text{-}fertilization}$; mg C m^{-2} d^{-1}) and in (f) pCO_2 ($\Delta pCO_2 = [pCO_2]_{post\text{-}fertilization} - [pCO_2]_{pre\text{-}fertilization}$; ppm). Color bar indicates changes in DIC ($\Delta DIC = [DIC]_{post\text{-}fertilization} - [DIC]_{pre\text{-}fertilization}$; μM). Note that PP (mg C m^{-2} d^{-1}) of OIF experiment number 1 (IronEx-1) was estimated by multiplying PP (mg C m^{-3} d^{-1}) with mixed layer depth and the numbers indicate the order of experiments (see Table 1). Sources are as follows: Kolber et al., (1994); Martin et al., (1994); Behrenfeld et al., (1996); Coale et al., (1996); Steinberg et al., (1998); Boyd et al., (2000); Bakker et al., (2001); Boyd and Law, (2001); Gervais et al., (2002); Coale et al., (2004); Boyd et al., (2004); Bakker et al., (2005); Boyd et al., (2005); de Baar et al., (2005); Hiscock and Millero, (2005); Smetacek et al., (2005); Takeda and Tsuda, (2005); Tsuda et al., (2005); Wong et al., (2006); Assmy et al., (2007); Boyd et al., (2007); Tsuda et al., (2007); Kudo et al., (2009); Tsumune et al., (2009); Harvey et al., (2010); Smetacek and Naqvi, (2010); Berg et al., (2011); Currie et al., (2011); Law et al., (2011); Peloquin et al., (2011); Smetacek et al., (2012); Thiele et al., (2012); Martin et al., (2013); Latasa et al., (2014).

Fig. 8. Time-series of (a) ^{234}Th-derived particulate organic carbon (POC) fluxes (mg m^{-2} d^{-1}) of the upper 100 m layer in patch (coral bar) and outside patch (blue bar) during EIFEX (modified from Smetacek et al., 2012). Time-series of (b) vertically integrated ^{234}Th (dpm l^{-1}) in patch (coral circle) and outside patch (blue diamond) relative to parent ^{238}U (dpm l^{-1}; dotted black

line) during SOIREE (modified from Nodder et al., 2001).

Fig. 9. Assessment framework for scientific research involving ocean fertilization (OF) (modified from Assessment Framework for Scientific Research Involving Ocean Fertilization, 2010).

Fig. 10. (a) Time-series of mixed layer depth-integrated chlorophyll-a concentrations (mg m^{-2}) during SOIREE (pink line), SEEDS-1 (brown line), SERIES (cyan line), SEEDS-2 (blue line), and EIFEX (teal line). Sources are as follows: Boyd and Abraham, (2001); Tsuda et al., (2007); Assmy et al., (2013). (b) The distributions of chlorophyll-a concentrations (mg m^{-3}) in ~5 days and ~46 days during SOIREE from SeaWiFS Level-2 daily images.

Fig. 11. Schematic diagram of KIFES representing experiment target site (eddy structure) and survey methods (underway sampling systems, multiple sediment traps, sub-bottom profilers, sediment coring systems, and satellite observations).

Fig. 1

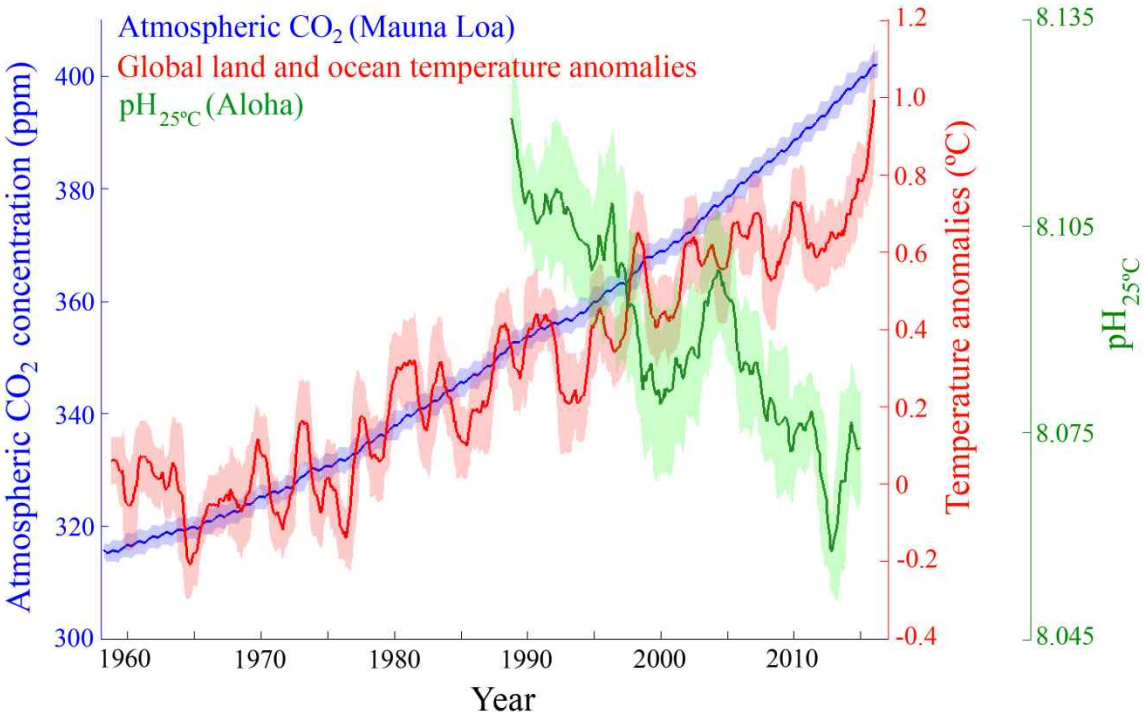
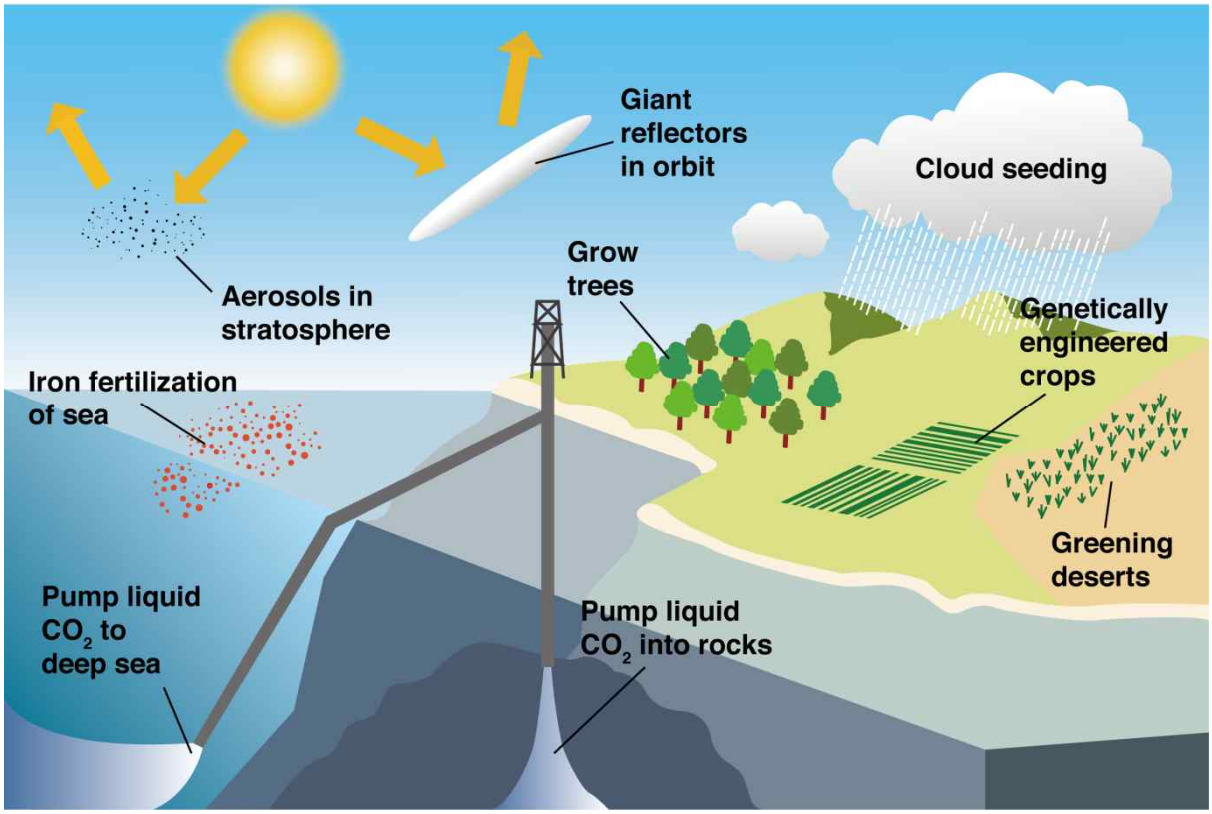
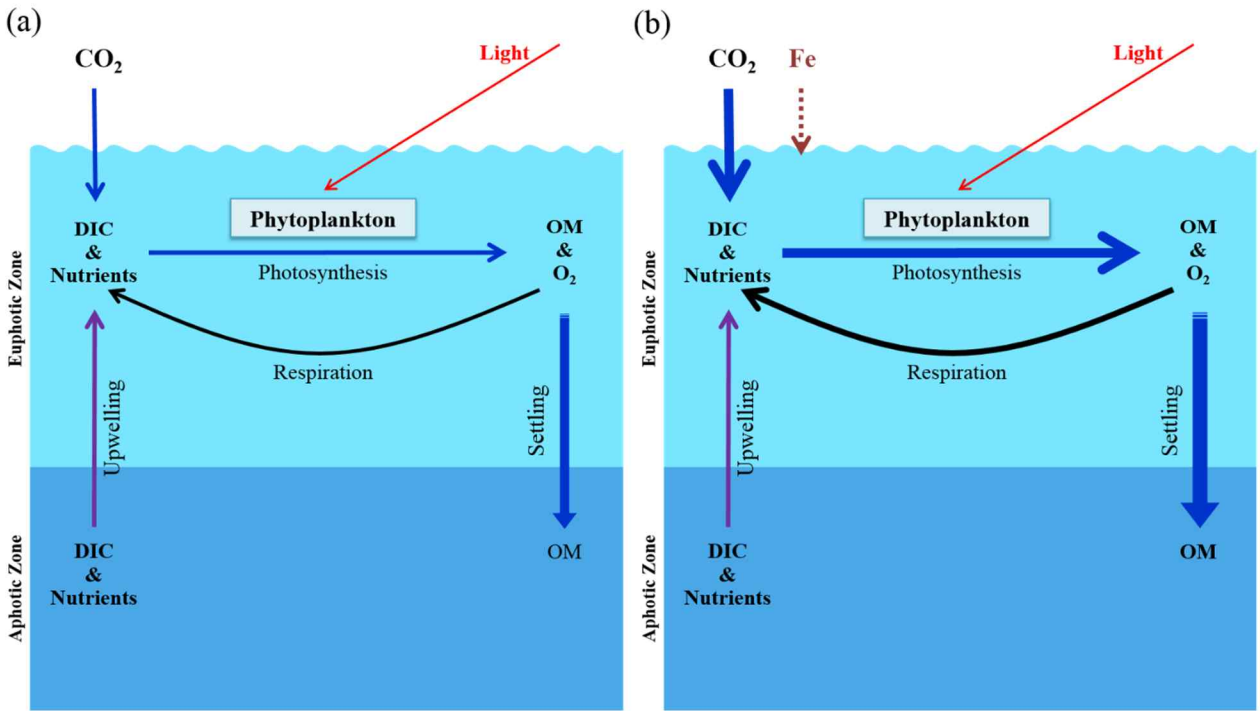

Atmospheric CO_2 (Mauna Loa)
Global land and ocean temperature anomalies
$pH_{25°C}$ (Aloha)
Atmospheric CO_2 concentration (ppm)
Temperature anomalies (°C)
$pH_{25°C}$
Year
300
320
340
360
380
400
-0.4
-0.2
0
0.2
0.4
0.6
0.8
1.0
1.2
8.045
8.075
8.105
8.135
1960
1970
1980
1990
2000
2010

Fig. 2


Giant reflectors in orbit
Cloud seeding
Grow trees
Aerosols in stratosphere
Genetically engineered crops
Iron fertilization of sea
Greening deserts
Pump liquid CO_2 to deep sea
Pump liquid CO_2 into rocks


(a)
CO_2
Light
Phytoplankton
DIC & Nutrients
Photosynthesis
OM & O_2
Euphotic Zone
Respiration
Upwelling
Settling
DIC & Nutrients
OM
Aphotic Zone
(b)
CO_2
Fe
Light
Phytoplankton
DIC & Nutrients
Photosynthesis
OM & O_2
Euphotic Zone
Respiration
Upwelling
Settling
DIC & Nutrients
OM
Aphotic Zone

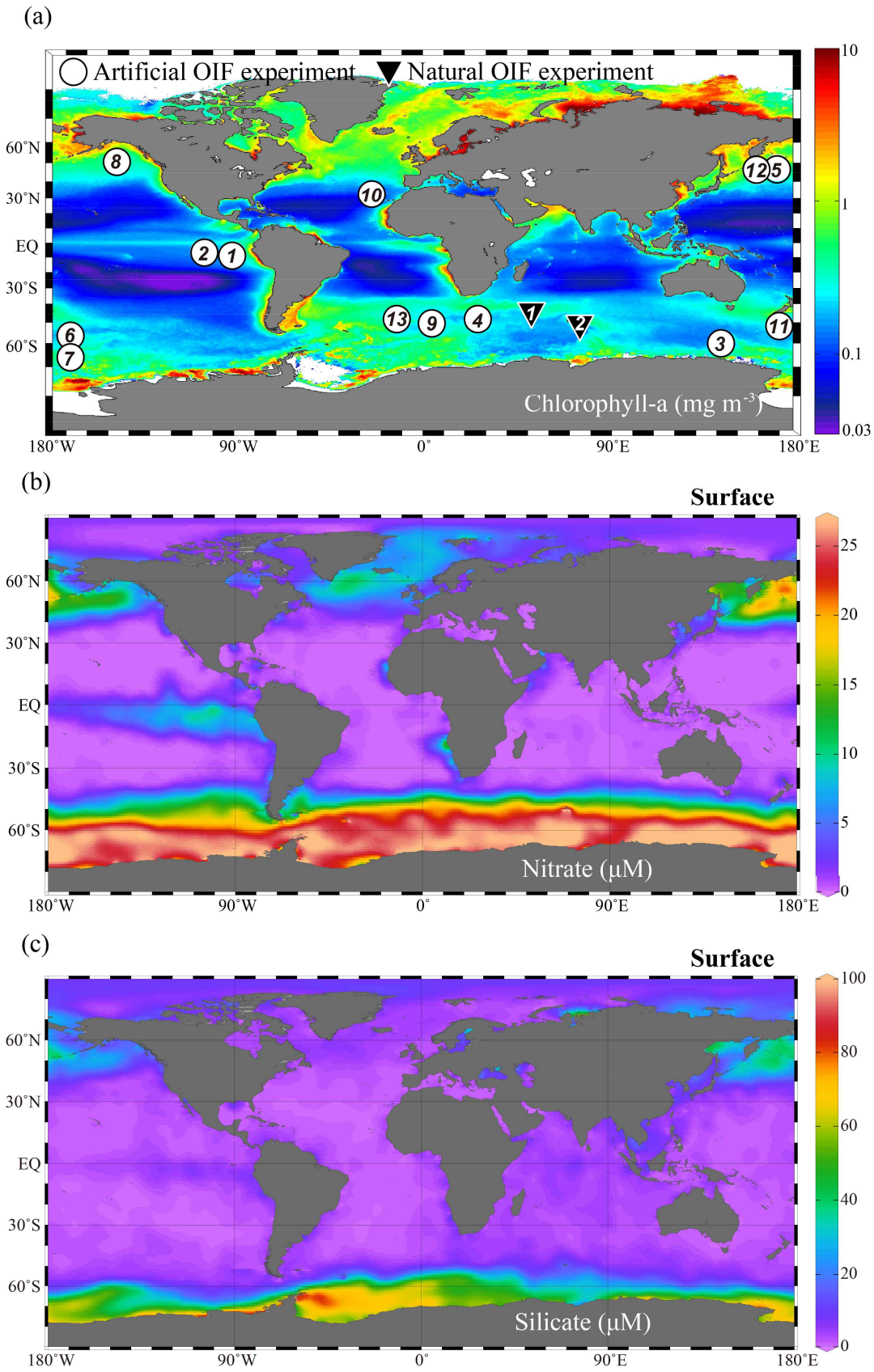
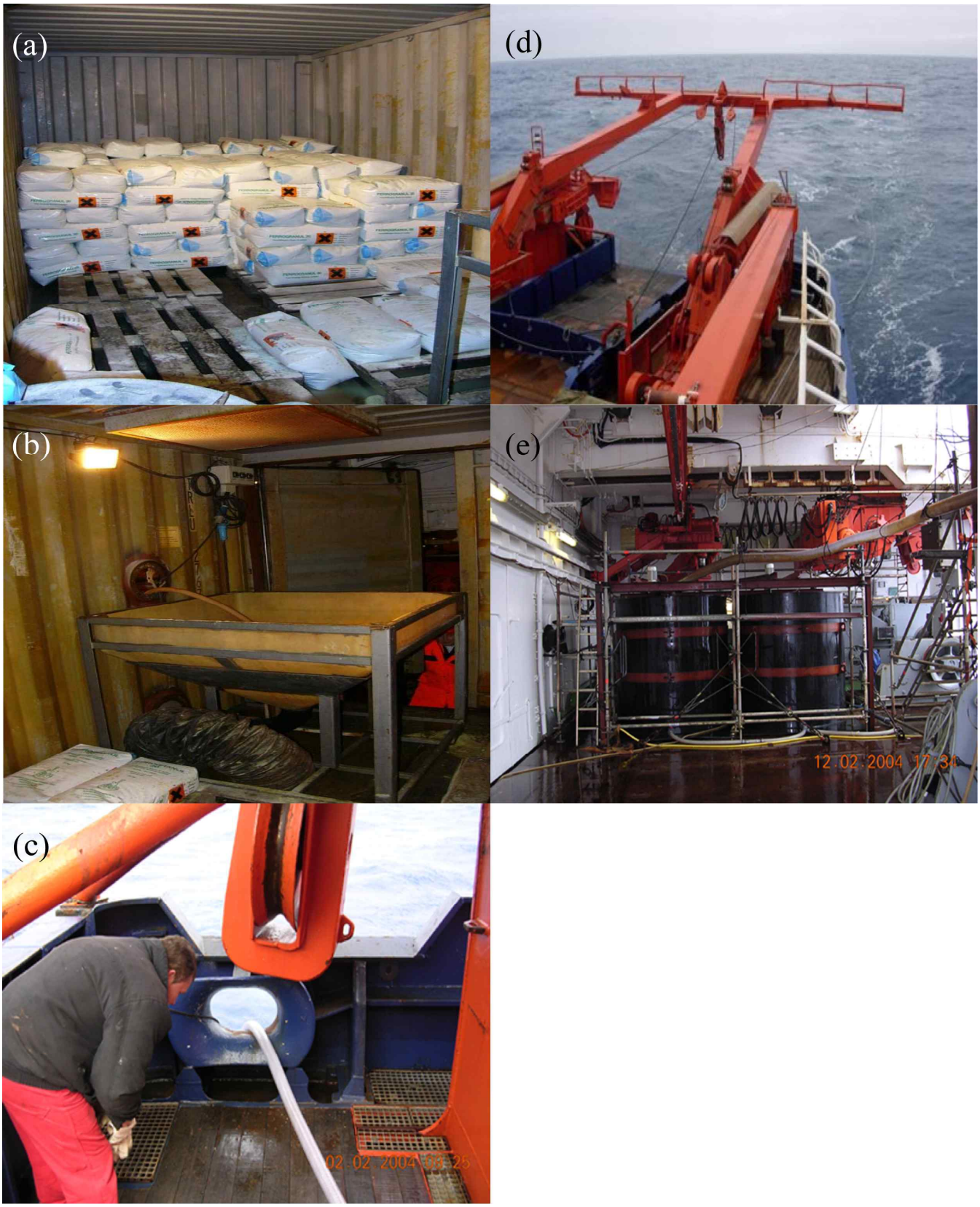
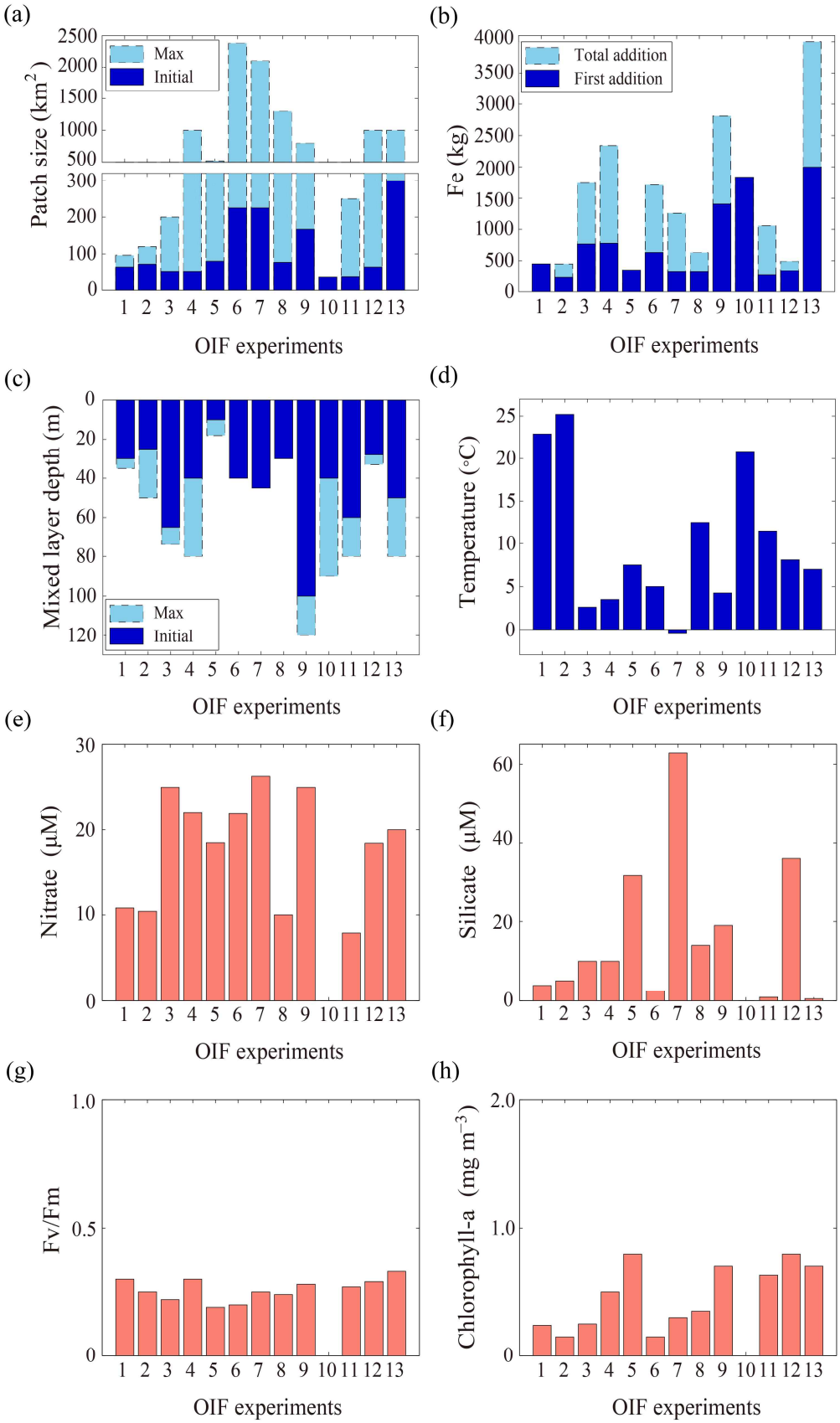
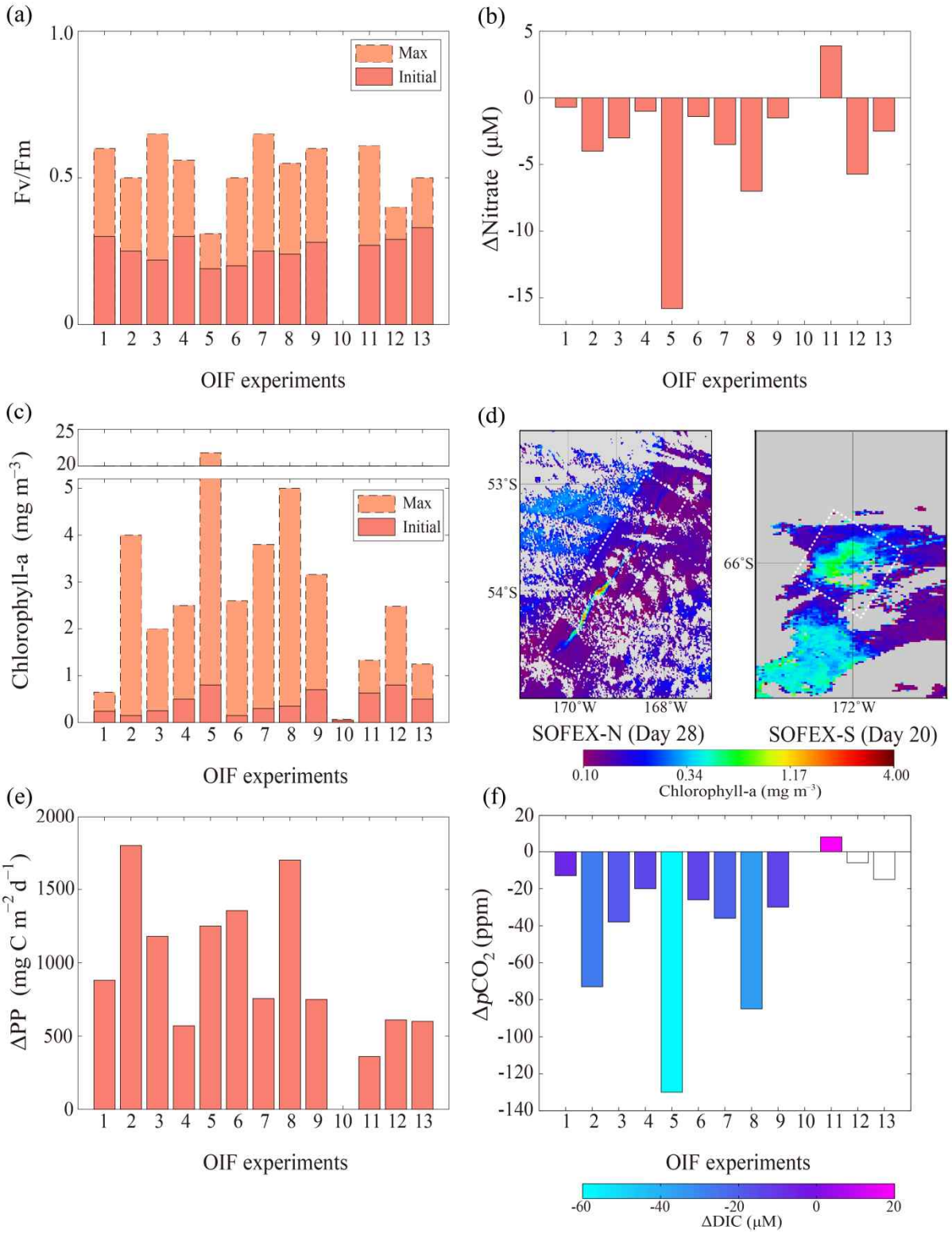
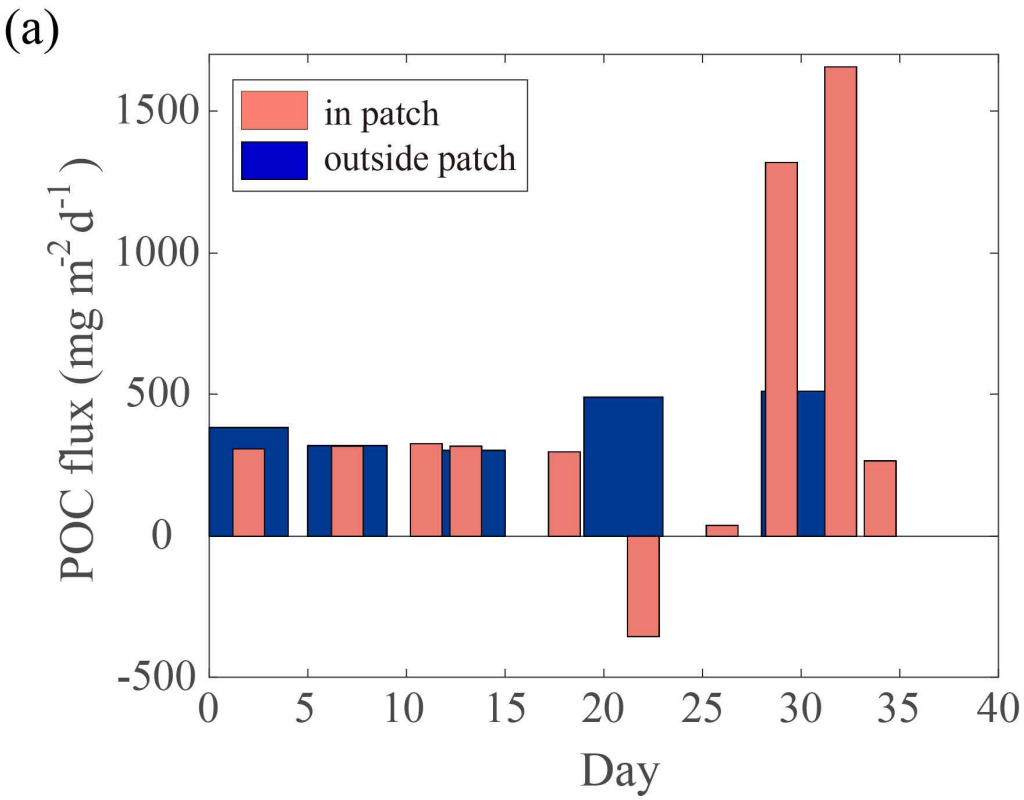

(a)
Artificial OIF experiment
Natural OIF experiment
Chlorophyll-a (mg m-3)
10
1
0.1
0.03
60°N
30°N
EQ
30°S
60°S
180°W
90°W
0°
90°E
180°E
(b)
Surface
Nitrate (μM)
25
20
15
10
5
0
(c)
Surface
Silicate (μM)
100
80
60
40
20
0

Fig. 5




(a)
Patch size (km^2)
Max
Initial
OIF experiments
(b)
Fe (kg)
Total addition
First addition
OIF experiments
(c)
Mixed layer depth (m)
Max
Initial
OIF experiments
(d)
Temperature (°C)
OIF experiments
(e)
Nitrate (μM)
OIF experiments
(f)
Silicate (μM)
OIF experiments
(g)
Fv/Fm
OIF experiments
(h)
Chlorophyll-a (mg m^{-3})
OIF experiments


(a)
Fv/Fm
Max
Initial
OIF experiments
(b)
ΔNitrate (μM)
OIF experiments
(c)
Chlorophyll-a (mg m^{-3})
Max
Initial
OIF experiments
(d)
53°S
54°S
66°S
170°W
168°W
172°W
SOFEX-N (Day 28)
SOFEX-S (Day 20)
0.10
0.34
1.17
4.00
Chlorophyll-a (mg m^{-3})
(e)
ΔPP (mg C m^{-2} d^{-1})
OIF experiments
(f)
ΔpCO_2 (ppm)
OIF experiments
-60
-40
-20
0
20
ΔDIC (μM)


(a)
in patch
outside patch
POC flux (mg m-2 d-1)
1500
1000
500
0
-500
0
5
10
15
20
25
30
35
40
Day

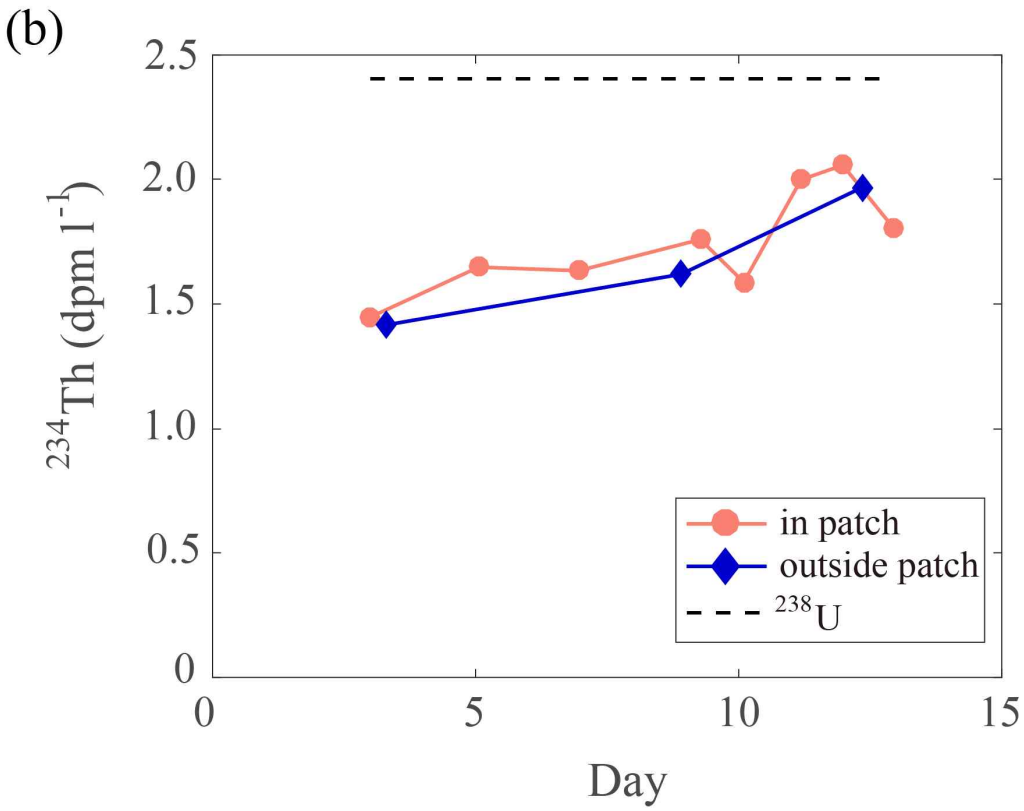
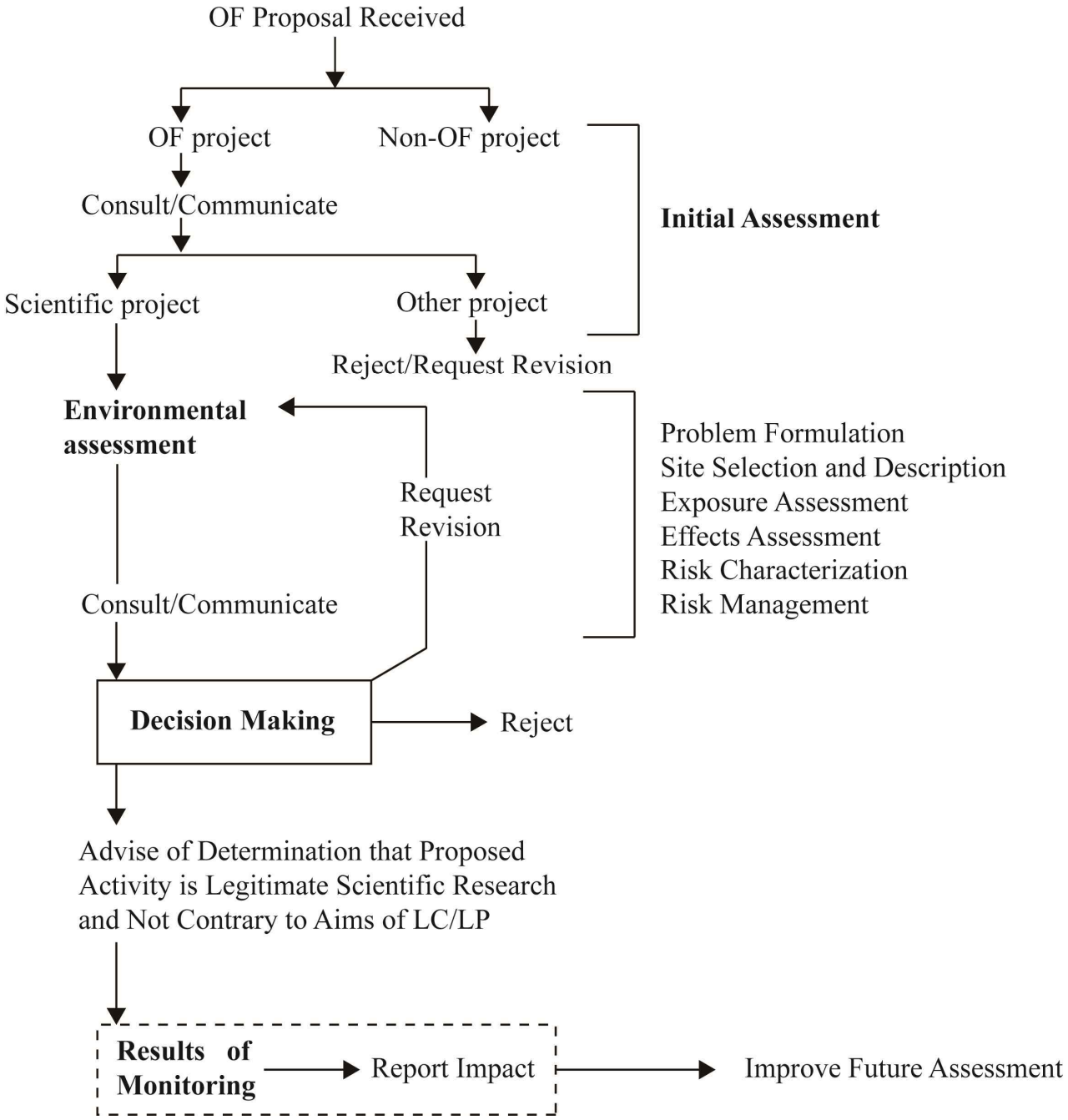

(b)
234Th (dpm l-1)
2.5
2.0
1.5
1.0
0.5
0
0
5
10
15
Day
in patch
outside patch
238U

Fig. 9


OF Proposal Received
OF project
Non-OF project
Consult/Communicate
Initial Assessment
Scientific project
Other project
Reject/Request Revision
Environmental assessment
Problem Formulation
Site Selection and Description
Exposure Assessment
Effects Assessment
Risk Characterization
Risk Management
Request Revision
Consult/Communicate
Decision Making
Reject
Advise of Determination that Proposed Activity is Legitimate Scientific Research and Not Contrary to Aims of LC/LP
Results of Monitoring
Report Impact
Improve Future Assessment

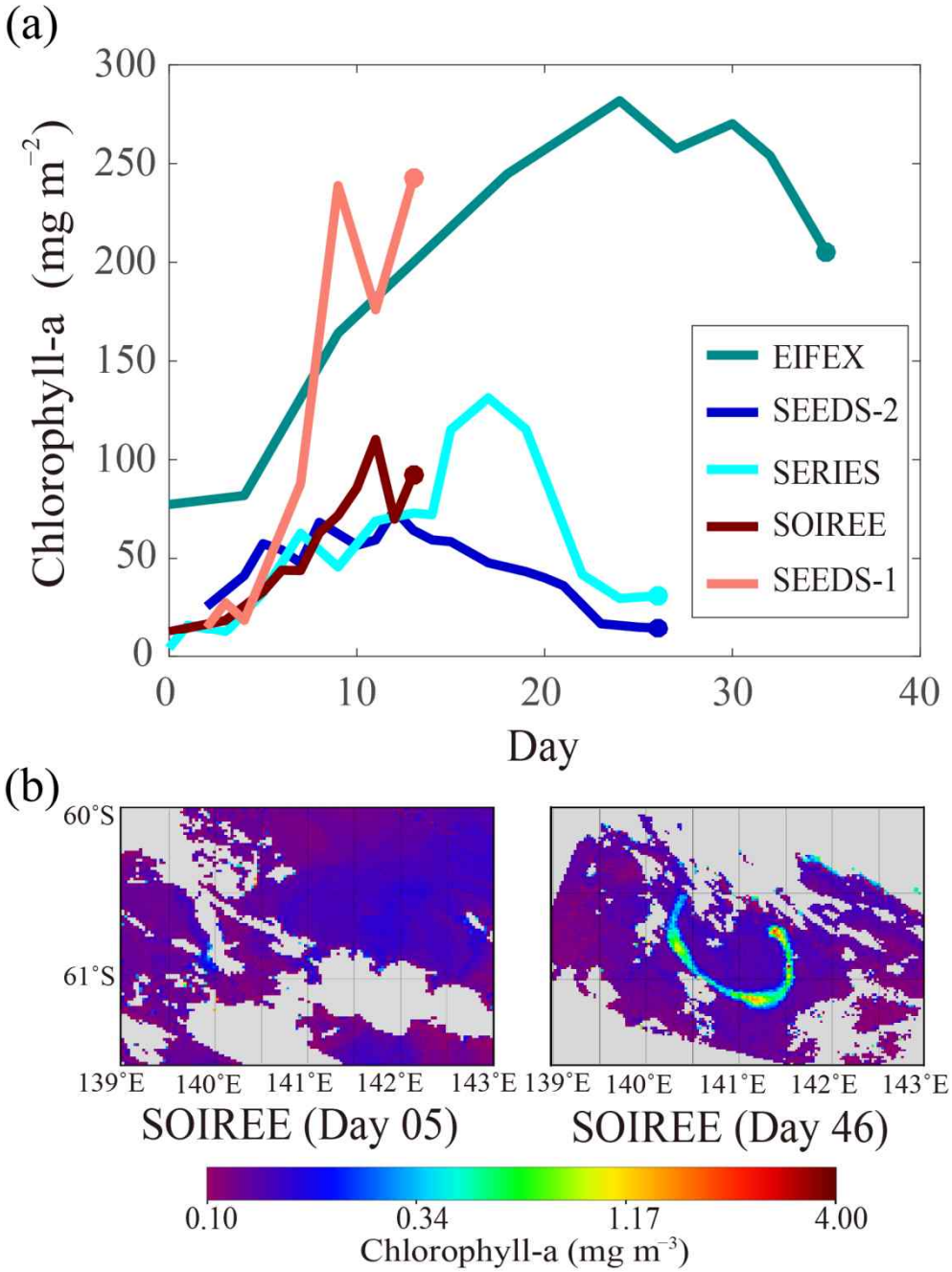
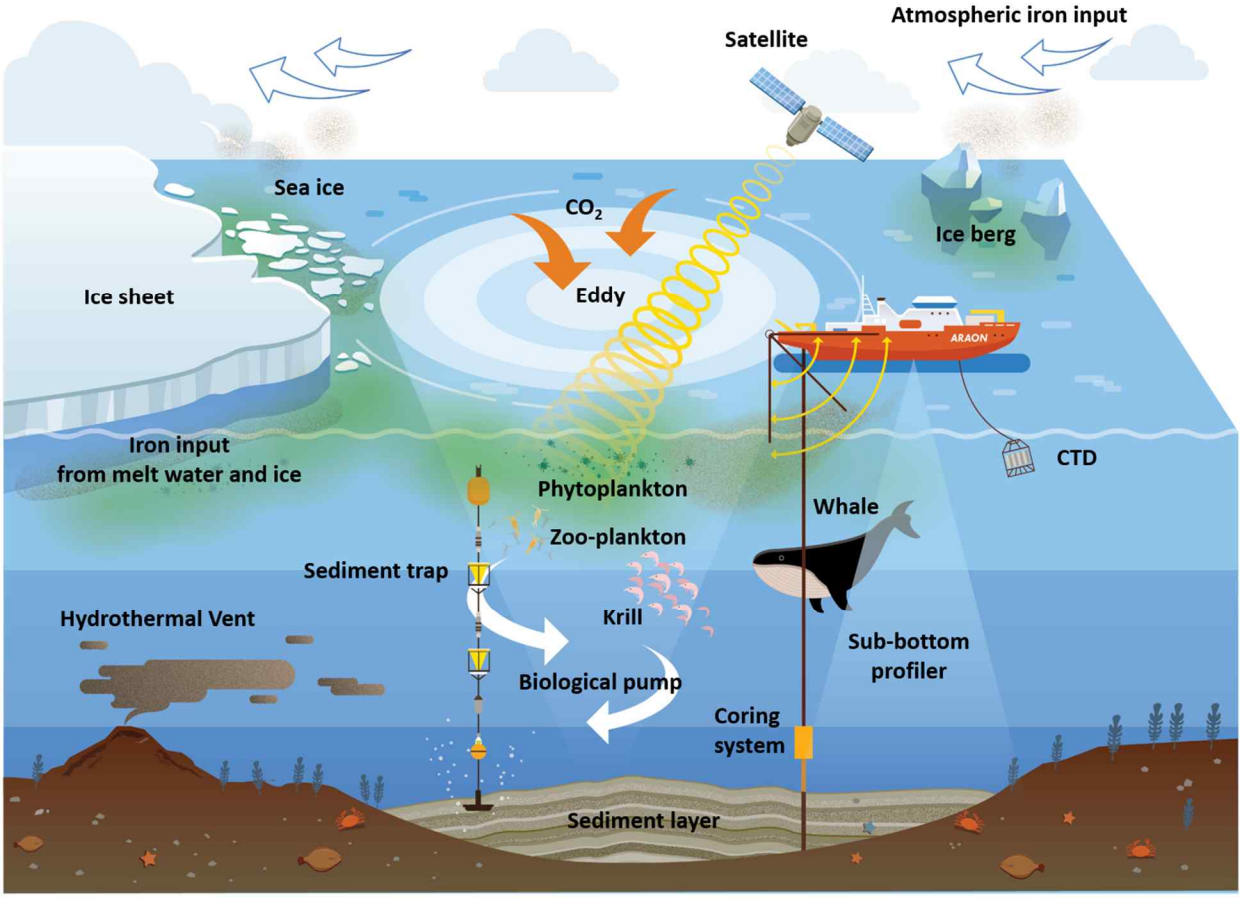

(a)
Chlorophyll-a (mg m^{-2})
300
250
200
150
100
50
0
0
10
20
30
40
Day
EIFEX
SEEDS-2
SERIES
SOIREE
SEEDS-1
(b)
60°S
61°S
139°E
140°E
141°E
142°E
143°E
SOIREE (Day 05)
SOIREE (Day 46)
0.10
0.34
1.17
4.00
Chlorophyll-a (mg m^{-3})

Fig. 11


Atmospheric iron input
Satellite
Sea ice
CO_2
Ice berg
Ice sheet
Eddy
ARAON
CTD
Iron input
from melt water and ice
Phytoplankton
Whale
Zoo-plankton
Sediment trap
Krill
Hydrothermal Vent
Sub-bottom
profiler
Biological pump
Coring
system
Sediment layer