Dear The Editor and Anonymous Reviewers,

First of all, we greatly appreciate the time and effort spent in reviewing of our manuscript. Unfortunately, we recently heard that the Korean Iron Fertilization Experiment in the Southern Ocean (KIFES) project, which was the inspiration for this review, has lost its funding. However, we here at Korea Polar Research Institute, as well as our domestic and international collaborators, fully intend to seek other sources of funding to revitalize the KIFES project. We apologize in advance that our responses to reviewer questions on KIFES are necessarily vague due to the current state of uncertainty surrounding the future of the project. Nevertheless, we believe that this manuscript is still worthy of publication as a detailed review of the history of ocean iron fertilization experiment designs and results. We have changed the title as follows: **"Ocean Iron Fertilization Experiment: Past-Present-Future looking to a future Korean Iron Fertilization Experiment in the Southern Ocean (KIFES) Project"** and still provide design guidelines for KIFES even though it is not presently funded.

We provide our responses (plain text with blue and red colors) to all comments (*italic text*) below.

- Response to Reviewer Comments -

Reviewer #1

-Response to General Comments:

1 ***Downward carbon fluxes can be quantified using diverse metrics. The really important one from the point of view of geoengineering would be the amount of carbon sequestered below the depth of deepest winter mixing in the study region, which most previous OIF did not measure. The article frequently uses terms such as "efficiency of OIF at reducing atmospheric CO2", but the authors never define clearly what they mean by this. The efficiency of the biological carbon pump can be quantified using several approaches, but from a geoengineering point of view the efficiency is less important that the absolute amount. The article would be more helpful if the authors defined clearly which metrics really matter. Moreover, it would be useful if the authors more explicitly assessed which of the experiments conducted to date were actually capable of detecting an enhancement of export if it had occurred (based on duration of the experiment relative to the phases of the bloom and the type of measurements that were taken), which of these did find a response in particle flux (e.g. EIFEX, SERIES), and how to what depth the carbon flux was followed.***

→ Thank you for pointing this out. To reflect the Reviewer's suggestion, we added the following to defining the "efficiency of OIF" in the Introduction:

"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 OIF efficiency (Buesseler and Boyd, 2003)."

Based on the Reviewer's comment, we have created a new Table (Table 5, page 32 of this response) that includes where available: absolute magnitude of export carbon flux; measurement depth; and the methods applied to detect an enhancement of export. Using Table 5, we also have created new Section "2.5 Assessment of export carbon flux" to more explicitly assess the enhancement of carbon flux in the manuscript as follows:

- 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. The biomass increased with minimum delay of 1 week (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, 2003). 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), 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 effect 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 ***The previous OIF differed significantly in experimental design, especially in terms of patch size, duration, location, and also in terms of which measurements were taken. I found the discussion of these aspects in Section 3.2 rather unsatisfying: especially since the authors are in advanced stages of planning a new experiment, what have they concluded from this literature about how best to design an OIF? What are their recommendations in terms of best patch size, minimum duration, and which measurements are required to quantify the effect on carbon sequestration? I think that discussion of these points is important, especially since the authors are clearly interested in persuading the scientific (and, presumably, wider) community that their proposed experiment will provide answers about the scope for geoengineering via OIF. The clear conclusion that they do appear to have drawn is that the experiment should be located inside an eddy. However, to accurately measure downward carbon flux out of the patch at the depth of maximum winter mixing will require a large patch to ensure that sediment traps potentially several hundred metres below the surface are not at too high a risk to actually miss possible particle fluxes. What is their conclusion about the minimum duration that is needed? Given the results of SERIES, SEEDS-2, EIFEX, and LOHAFEX, it would seem to me that one should aim at between 35 and 40 days post-fertilisation. Further, what recommendations can be made about measurement approaches to quantify carbon fluxes? An important point to me is that having multiple redundant methods is very important, e.g. thorium profiles, frequent deployments of sediment traps at multiple depths (ideally neutrally buoyant traps), and high-frequency measurements of properties such as pCO2 and O2:Ar ratios. It also strikes me that autonomous platforms should play a much greater role in future OIF than they have in the past, e.g. a combination of gliders and Lagrangian floats equipped with biogeochemical sensors. Especially bio-optical sensors such as fluorescence and backscatter can be extremely useful to help constrain downward particle fluxes and their vertical and horizontal variations.***

→ We agree. More discussion is necessary. As suggested by Reviewer, we have moved this Section to "4 Future:

Considerations for designing future OIF experiments" to supplement discussion for designing a future OIF experiment that would maximize the effectiveness of OIF. We have revised Section 4 to consider the methods for iron addition (i.e., 'How'), tracking methods and measurement parameters (i.e., 'What'), location (i.e., 'Where'), timing (i.e., 'When'), and duration (i.e., 'How long') to build on the results of OIF experiments as follows:

- 4 Future: Considerations for designing future OIF experiments

"Scientific research on OIF has focused on improving our understanding of the efficiency, 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, 2015). It is therefore of paramount importance that future OIF experiments continue to focus on the efficiency 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 maintaining 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 oxidation times would suggest. 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 efficiency 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 during previous OIF experiments (Table 5). Because of their high vertical, water column 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 and 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 efficient 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 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-sea floor over time (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. 9a) 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. 9b) (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 9a) (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 efficiency (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."

3 ***The discussion of possible unintended side-effects could be similarly improved by trying to draw clearer conclusions rather than just summarising results from the previous literature. For example, it seems to me that the main conclusion about domoic acid is that it is very variable regardless of fertilisation, with the cited Smith et al. paper actually reporting higher per-cell quotas from natural than from artificially fertilised waters (the cited Trick et al. paper relied on bottle incubations and extrapolations based on claims about likely bloom size made by geoengineering companies on an internet site). Moreover, while a degree of oxygen consumption would certainly result from OIF, the sentence that "Box model solutions have further suggested that anoxic conditions may develop after OIF" is quite misleading: the cited reference is actually a much more realistic 3-dimensional model that only found anoxia developing in part of the western Indian Ocean, and only after many years of sustained complete nutrient utilisation in the Southern Ocean. This is probably a significantly more extreme scenario than could be achieved in practice, suggesting that anoxic conditions are actually quite unlikely. Conversely, increased production of other relevant gases, such as N2O, is clearly an important concern (though the discussion of DMS could do with some reference to the fact that its role in climate seems to be rather more complex than originally thought).***

→ We apologize for the confusion. Based on the Reviewer's comments, we have provided conclusions about side effects based on recent modeling results and have revised Section "3.1 Environmental side effects" as follows:

- 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 effects 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 suggested that iron fertilization of 2 % of the Southern Ocean 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). Whereas in the SERIES experiment, DMS production decreased 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 the origin of regional variation (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 with the DMS response further study in 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, OIF-induced reductions in oxygen concentration based on more sophisticated and realistic models have been smaller resulting in well-oxygenated end-conditions rather than oceanic anoxia (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*' (10^6 and 10^5 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 have been shown cause harm to marine communities in coastal waters (Silver et al., 2010). However, no DA was found during EisenEx, even though '*Pseudo-nitzschia*' were the dominant diatom species (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 have yet to reached 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 efficiency 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 effect 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."

-Specific Comments:

4 ***Abstract Line 10, and page 10 final paragraph Line 1: make it clear that these side- effects are possible side effects, and that changes in community composition may have unintended consequences.***

→ We have made it clear by changing "including side effects" to "including possible side effects" in the Abstract Line 10 and "The changes of phytoplankton community after iron addition discussed in Section 2.4 also has unintended consequences" to "The changes of phytoplankton community composition after iron addition discussed in Section 2.4 may also have unintended consequences" in page 10 final paragraph Line 1.

5 ***Page 4 Paragraph 3: > and < signs for latitude are the wrong way round.***

→ We corrected > and < signs for latitude.

6 ***Page 12 final paragraph: given the large number and large scale of natural mesoscale blooms in HNLC regions (e.g. due to iceberg-derived iron), I think it is fair to say that the risks to the environment from small-scale OIF experiments is very small indeed, and I think that the authors should be prepared to make that case. The risks of large-scale OIF for geoengineering purposes are the risks that are not understood, and small-scale studies are what we therefore need to undertake at this point to assess these risks better.***

→ The reviewer is correct. We have revised Section "3.2 International law of the sea to OIF" by rephrasing sentences as follows:

- 3.2 International law of the sea to OIF

"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."

7 ***Page 14 Paragraph 2: Sentence starting "To data . . ." should read "To date, the only OIF experiment . . ."***

→ Done.

8 ***Page 14 Section 4.2.3: What do the authors mean by "rehearsal"? Will they add only a tracer, such as SF6, or will iron be added as well?***

→ We apologize for the confusion. By "rehearsal', we intended to indicate that we would conduct hydrographic surveys outside/inside the eddy structure in the eastern Bransfield Basin and employ drifting-buoys prior to the actual (eddy structure) OIF experiment without iron addition. Basically, we would not plan to add SF_6 as a chemical tracer in the KIFES project. We deleted the word and revised the KIFES section. Please, find it below (page 25–28 of this response).

9 ***Page 14 Section 4.2.4: As I indicated in one of my general comments, I think that future OIF could benefit greatly from using autonomous platforms, such as gliders, equipped with biogeochemical sensors. If this is not planned at present, I would urge the project leaders to consider their use.***

→ Thank you. Yes. We would plan to use autonomous platforms (page 27 of this response).

10 ***Page 15 Section 4.2.5: What is the second stage of KIFES?***

→ We meant the second 5-years of the KIFES project (2021–2025). Given the current status of KIFES funding, we deleted mention of the second stage of KIFES.

11 ***In Figure 4, the authors could consider marking the study region proposed for KIFES.***

→ We have included the number "14" to indicate KIFES in the Antarctic Peninsula in Figure 4 (page 35 of this response).

12 ***Figure 8 provides a summary of carbon flux related data for two experiments, EIFEX and SOIREE (though Fig 8b is referred to in the context of IronEx-2 in the text). Several other experiments did report comparable***

data, either with sediment traps, thorium deficits, or both. Comparison of these data is obviously complicated by the fact that different experiments measured flux at different depths, but trying to summarise the results of all of the studies that reported particle fluxes might be helpful. Moreover, when the authors state on Page 9 Paragraph 2 that "That being said, EIFEX was the exception. Significant changes in export production were not found in any of the other OIF experiments", it should be made clear that only a subset of all OIF experiments was actually designed in such a way that an enhancement of downward particle flux could be detected (especially given the short duration of several experiments).

→ We now compare carbon flux from all the studies in Table 5. Please refer our Response (1).

Reviewer #2

-General Comments:

13 **(1)** ***The presentation of results from previous experiments seems too much like a catalog of data, but there is no thorough discussion on why the outcomes of the experiments were so different, and what has been learned from these experiments. (2) Further, given that KIFES is planned to take place in the Southern Ocean, it is not obvious to the reader how the detailed presentation of results from experiments carried out in other oceanic basin is relevant here.***

→ (1) We have created a new "Table 2 Summary of OIF experiments; objective, significant results, and limitation" summarizing Section 2 including OIF design, biogeochemical response, and limitations (page 29–30 of this response). We have also modified manuscript with new Section "2.6 Significant results and limitations in previous OIF experiments" to discuss why the outcomes of the experiments were so different, and what has been learned from these experiments as follows:

- 2.6 Significant results and limitations in previous OIF experiments

"To understand how various physical and biogeochemical properties response in HNLC regions by artificially adding iron, previous OIF experiments have been conducted with various objectives (Table 2). These various objectives have contributed to having idea to find out optimal conditions that have potential capacity to efficiently sequester carbon by studying whether organic carbon produced by iron enrichment is retained into deep ocean export flux or recycled in the water column by grazing pressure and remineralization in each HNLC regions (Smetacek et al., 2012). To test iron hypothesis, initial artificial OIF experiments have focused on whether iron supply limits phytoplankton growth in HNLC regions based on "bottom up" approach and confirmed increased phytoplankton biomass by showing maximum drawdown of $p\mathrm{CO_2}$ by 130 ppm in SEEDS-1 and increase in primary production by 1800 mg C m^{-2} d^{-1} in IronEx-2 (Figure 7e and f) (de Baar et al., 2005; Boyd et al., 2007). Massive phytoplankton bloom was due to abrupt rise in diatom (Coale et al., 1996; Boyd et al., 2000). Especially, in "bottom up" approach, there were multiple efforts to detect export fluxed to deep layer of iron-induced massive phytoplankton bloom to confirm the second condition 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). In the case of OIF experiments with low amounts of carbon fluxed below mixed layer in spite of highly increased phytoplankton production in the water column, OIF experiments were focused on recycled carbon which is determined by grazing pressure (SEEDS-2) and bacterial remineralization (SERIES) based "top down" approach (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 during OIF experiments such as SAGE, LOHAFEX, designed to confirm biogeochemical response by added iron in very low silicate concentrations (<~2 nM) (Table 2) (Coale et al., 2004; Harvey et al., 2010; Martin et al., 2013)."

→ (2) The reason we are producing a comprehensive review of previous OIF experiments is to lay an efficient groundwork for new projects by determining the advantages and disadvantages, successes, and failures of earlier efforts. We hope that understanding the history of OIF experimentation will lead to more efficient experimental strategies and designs, which will in turn produce successful OIF experiments through the selection/adoption of useful approaches and tools that have already had verifiable success in the field.

Contrast to the previous experiments, the idea that the KIFES project was developed was initiated from deep-sediment core information obtained in the eastern Bransfield Basin. 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. Please refer Section "5.1 Background for future KIFES suggestion" (page 25–26 of this response).

14 ***In the same line of thought, the rationale for artificial vs. natural iron experiments could also be discussed.***

→ Good point. We have added the rationale for artificial vs. natural iron experiment in 3.2 Section as follows:

- 3.2 International law of the sea to OIF

"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 effect of long-term and large-scale fertilization (Aumont and Bopp, 2006; Blain et al., 2007; Denman, 2008; Pollard et al., 2009)."

"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)."

→ We also have added the rationale for artificial vs. natural iron experiment in 4 Section. Please refer our Response (2).

15 ***Overall, model studies are poorly represented in this review. Given that C sequestration estimates, as well as large scale and long term impacts of OIF are mostly determined through model studies, it might be relevant to mention them and how additional experiments might help constrain such models (see also comments below).***

→ Thank you for pointing out this deficiency. We have added the model results for carbon sequestration estimates in Section 3.2 as follows:

- 3.2 International law of the sea to OIF

"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)."

We have also added modelling results in our discussion of side effects in Section 3.1. Please refer our Response (3).

-Specific Comments:

16 ***p. 7, line 28: the authors explain Fv/Fm but the term is used much earlier in the text. I suggest shifting the explanation to the first time Fv/Fm is used. I am also not sure that the description as written is very useful for people outside the field.***

→ Done.

17 ***p.8, lines 16-28: Given the large differences in mixed layer depth between experiments, I would suggest the authors also discuss mixed layer integrated chlorophyll stocks as these better reflect the real biomass built up (i.e. standing stocks accumulated during EIFEX were similar to those for SEEDS even though concentrations were an order of magnitude lower).***

→ Thank you for suggestion. We have added some sentences about integrated chlorophyll stocks in Section 2.4 as follows:

- 2.4 Biogeochemical responses

"However, added iron influences the growth of phytoplankton from surface to euphotic depth because added iron is mixed within the mixed layer by physical processes (Coale et al., 1998). Although maximum chlorophyll-a concentration during SEEDS-1 (22 mg m^{-3}) was much higher than EIFEX (3.16 mg m^{-3}), mixed layer integrated chlorophyll-a concentrations was similar with ~250 mg m^{-2}. There were distinct differences between mixed layer integrated chlorophyll-a concentration and surface chlorophyll-a concentration. Therefore, during previous OIF experiment, to quantify the exact changes in phytoplankton biomass to iron addition, it would be important to detect the changes in integrated primary production within water column by iron added within mixed layer."

18 ***p.8, line 26: there is a mistake in the sentence ("were appeared at"?), and the message is not clear.***

→ We apologize for this confusion. We have revised the sentences explaining satellite chlorophyll-a concentration images to clearly deliver the message as follows:

- 2.4 Biogeochemical responses

"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 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 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)."

19 ***p. 9, line 1: add "the" before "surface".***

→ Done.

20 ***p. 10 lines 17-21: More recent model studies do not show development of anoxic conditions for large-scale iron fertilization in the Southern Ocean (see for instance Oschlies et al. Biogeosciences, 7, 4017–4035, 2010; Keller et al. Nature Communications, doi: 10.1038/ncomms4304, 2013).***

→ We apologize for confusion. We have added more recent model studies in Section 3.1. Please refer our Response (3).

21 ***p.10, line 36: Change "also has" to "could also have"***

→ Done.

22 ***p. 11, line 3: Change "even though generally…" to "even though diatom species of the genus Pseudo-nitzschia were dominant numerically".***

→ Done.

23 ***p. 11, lines 20-28: I feel that the question how is somehow too easily brushed aside. This review could be used to discuss protocols and relevant parameters that should be measured, applied or developed. Not all experiments followed similar protocols, or measured all parameters.***

→ We have modified manuscript. Please refer our Response (13).

24 ***p.13 lines 23-31: (1) Can the authors give a reference for the mentioned studies. (2) Further, the rationale for doing the experiment in the Bransfield Basin is not clear.***

→ (1) Thanks. We included references.

→ (2) Please find the revised KIFES parts below (page 25–26 of this response).

25 ***Table 2: It would be more useful if the authors provided with initial nutrient and DIC and the delta values (rather than the final concentrations).***

→ Done (page 31 of this response).

26 ***Figure 3. I do not understand how oxygen is part of the settling component.***

→ We have removed this path from Figure 3 (page 34 of this response).

27 ***Legend Figure 4, line 3: Change "nitrate and silicate were presented" to "nitrate and silicate were plotted"***

→ Thank you. Done.

28 ***Figure 5 legend: Change to "Picture for iron addition procedure" to "Illustration" or "Photographs of iron the addition procedure. Panels a-e taken during EIFEX and LOHAFEX***

→ Done.

29 ***Figure 5a legend: Change legend: a) Iron (II) sulfate bags***

→ Done.

30 ***Figure 5b legend: The photograph shows the funnel where iron and HCl was poured, not the HCl.***

→ Done.

31 ***Figure 5f: I am not sure were this picture was taken (the corresponding web page gives no information) but I find it misleading as the iron mixture is released in much lower quantities than depicted here (compare with the size of the hose in panel d taken during EIFEX) and has a different appearance too. I would recommend removing this panel, unless reliable information of its provenance can be provided.***

→ The figure has been removed (page 36 of this response).

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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 efficiency of artificial OIF in terms of atmospheric carbon sequestration (i.e., export production efficiency) 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.

Objectives: (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 R/V *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 O_2/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 effects 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 measuring of the efficiency 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.

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 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 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)		

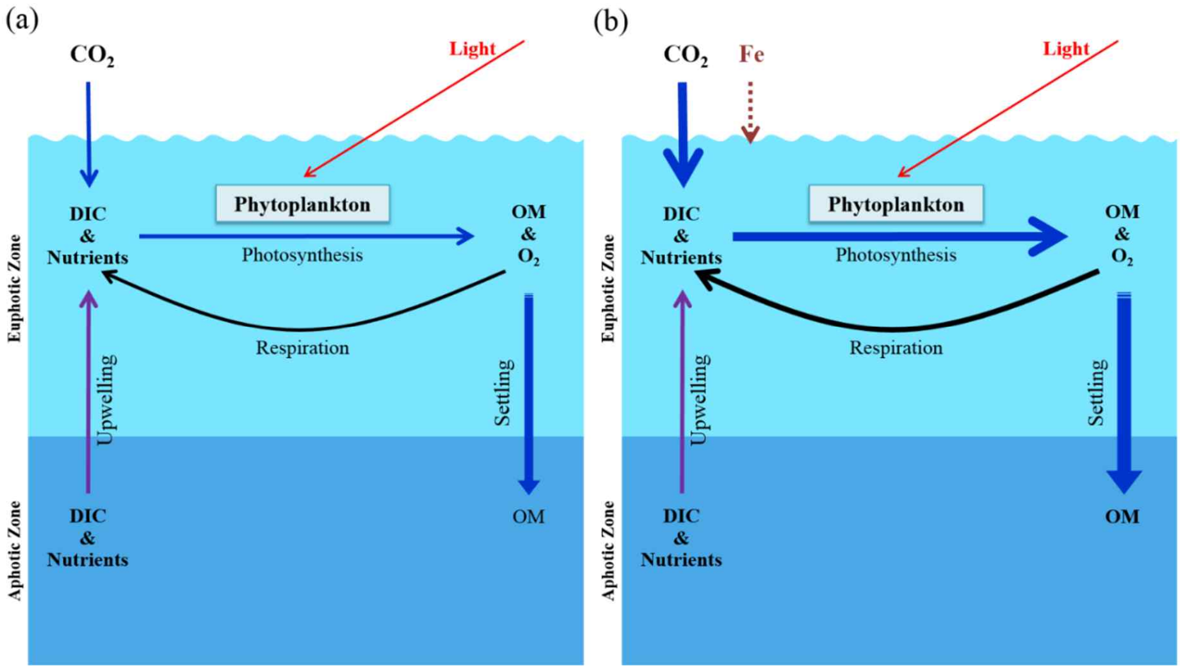
Sources are as follow: Martin et al., 1994; Steinberg et al., 1998; Boyd et al., 2000; Bakker et al., 2001; Frew et al., 2001; Bakker et al., 2005; Hiscock and Millero, 2005; Smetacek et al., 2005; Takeda and Tsuda, 2005; Wong et al., 2006; Boyd et al., 2007; Tsumune et al., 2009; Harvey et al., 2010;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

Table 5. Export flux (mg C m^{-2} d^{-1}).

	Experiment	In patch Initial (day)	In patch Maximum (day)	Outside Initial (day)	Outside Maximum (day)	Depth (m)	Method	Measurement
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, BSi, PON, PIC) *Transmissometer
4	EisenEx							*Water column (^{234}Th) *Sediment trap at 20, 500, 3500 m (POC)
5	SEEDS-1	374/166 (2)	1000/140 (13)			50/200	^{234}Th, trap	*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, ^{13}C, ^{15}N) *Free-drifting sediment traps at 100 and 300 m (POC, BSi, ^{13}C, ^{15}N, ^{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, $^{13}C_{org}$, ^{15}N, PON)
10	FeeP							
11	SAGE							*Free-drifting sediment traps at 80, 300 m (POC, PN, PSi) *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; 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.

Figure 3



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

Figure 4

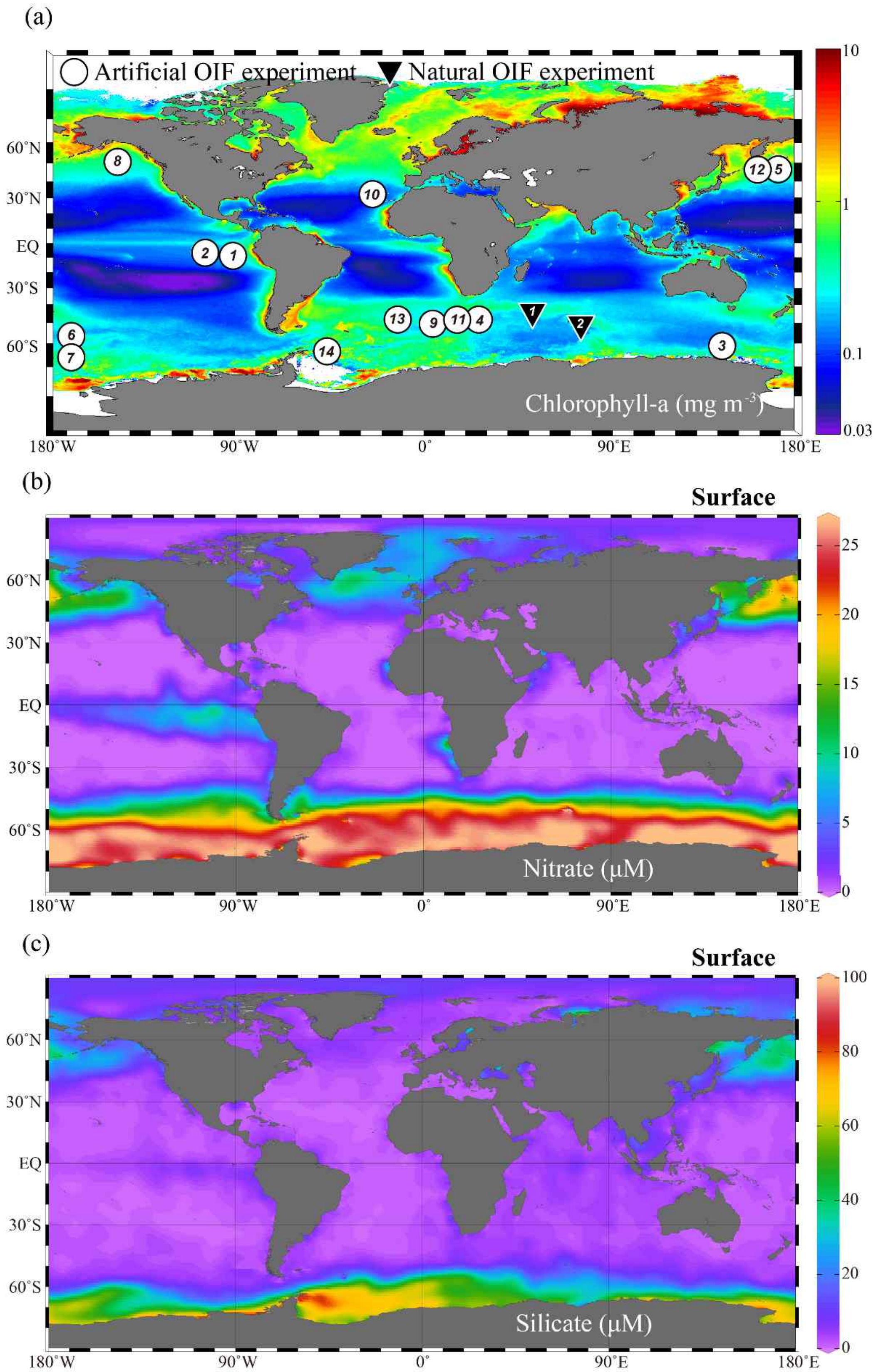

(a)
Artificial OIF experiment
Natural OIF experiment
1 2 3 4 5 6 7 8 9 10 11 12 13 14
1 2
Chlorophyll-a (mg m-3)
10
1
0.1
0.03
60°N
30°N
EQ
30°S
60°S
180°W
90°W
0°
90°E
180°E
(b)
Surface
Nitrate (μM)
25
20
15
10
5
0
(c)
Surface
Silicate (μM)
100
80
60
40
20
0

Figure 5