glacial-scale enrichment would result in an air-to-sea flux of about 4.6 mol C m^{-2} $year^{-1}$ or about 2 Pg C $year^{-1}$ over an oceanic area of 3.6×10^7 km^2 (50°S to 65°S). Seasonal ice cover may reduce this estimate by a factor of 2. These estimates are several times larger than the present net annual Southern Ocean (south of 50°S) uptake of CO_2 of 0.4 Pg C $year^{-1}$ (*1*) and comparable to the current global ocean net uptake of atmospheric CO_2. Dilution-corrected estimates of POC export (Table 1) extrapolated to an annual basis suggests a similar flux on the order of 8 mol C m^{-2} $year^{-1}$.

These results demonstrate that iron addition to the Southern Ocean increases primary productivity and decreases pCO_2. It remains difficult to extrapolate these findings with confidence to their impact on atmospheric composition because the large-scale impacts of iron enrichment on midwater processes and the length scales of POC remineralization are not yet known. The results strongly suggest, however, that the Southern Ocean was more productive and exported more carbon during periods of higher atmospheric iron input, which occurred during the last glacial maximum.

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Supporting Online Material

www.sciencemag.org/cgi/content/full/304/5669/408/DC1
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REPORTS

The Effects of Iron Fertilization on Carbon Sequestration in the Southern Ocean

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An unresolved issue in ocean and climate sciences is whether changes to the surface ocean input of the micronutrient iron can alter the flux of carbon to the deep ocean. During the Southern Ocean Iron Experiment, we measured an increase in the flux of particulate carbon from the surface mixed layer, as well as changes in particle cycling below the iron-fertilized patch. The flux of carbon was similar in magnitude to that of natural blooms in the Southern Ocean and thus small relative to global carbon budgets and proposed geoengineering plans to sequester atmospheric carbon dioxide in the deep sea.

As the largest high nutrient–low chlorophyll region, the Southern Ocean was chosen for a purposeful iron fertilization experiment, SOFeX (Southern Ocean Iron Experiment). The experiment was conducted at two sites both north and south of the Antarctic Polar Front, in low- and high-silicate waters, respectively. We focus here on the "southern patch" where the inert tracer SF_6 and four enrichments of iron were added to a 15 km by 15 km patch (66°S, 172°W), which was tracked and monitored by three ships in a Lagrangian fashion for 1 month in January to February 2002. As in previous experiments (*1–3*), the addition of the essential micronutrient iron led to measurable decreases in dissolved inorganic carbon and nutrients within the surface mixed layer (upper 40 to 50 m) associated with enhanced growth of marine phytoplankton, the details of which are described in the accompanying article by Coale *et al.* (*4*).

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As part of SOFeX, we used naturally occurring radionuclide ^{234}Th (half-life = 24.1 days) as an in situ tracer of the sinking flux of particles. ^{234}Th has a high affinity for particle surfaces, and its removal on sinking particles results in a ^{234}Th "deficit," i.e., measured ^{234}Th activities that are less than those of its long-lived and soluble parent, ^{238}U, which is the primary source of ^{234}Th in seawater. "Excess" ^{234}Th may be observed at depth due to remineralization of sinking particles, such that the rate of regeneration of ^{234}Th from sinking particles exceeds the local production and decay balance. Measurements of ^{234}Th can be used to calculate the net flux of ^{234}Th on sinking particles at any given depth, and its ratio to particulate carbon and other particle-associated elements can be used to determine the sinking flux of particulate organic carbon (POC) and other elements from the surface ocean (*5*, *6*).

During SOFeX, we observed at the earliest station "IN" the patch (day 5.8), and throughout the experiment at "OUT" control stations, a ^{234}Th deficit within the mixed layer (depths <40 to 50 m) and excess ^{234}Th below these depths (Fig. 1, A and C) (the presence of SF_6 tracer was used during SOFeX to determine IN versus OUT locations). This ^{234}Th profile is typical of this site late in the growth season (*7*) and in many other oceanic settings. Within the iron-fertilized patch we observed over time both a deepening and increasing ^{234}Th deficit, and a decrease in excess ^{234}Th at depth (see time-sequence Movie S1 of ^{234}Th activity distributions). Because hydrographic conditions between IN and OUT stations were indistinguishable, we attribute these changes to the impact of iron fertilization on particle export.

By the end of the 28-day observation period, fluxes of particulate ^{234}Th out of the upper mixed layer increased within the patch (fluxes at 50 m are calculated from the 0- to 50-m total ^{234}Th activity) (Fig. 1, B and D). Of particular interest are enhanced ^{234}Th fluxes below 50 m, deeper than where the iron was introduced and the primary growth response was observed [details of the vertical structure and biomass and growth responses can be found in Coale *et al*. (*4*)]. Overall, the lower average ^{234}Th activities in the patch between 0 and 100 m result in higher calculated particulate ^{234}Th fluxes.

As with particulate ^{234}Th fluxes, the sinking flux of POC IN was greater than POC flux OUT at the end of 28 days (Fig. 2). These differences were largest deeper in the water column at 100 m versus 50 m. The 100-m POC fluxes of 12.4 mM C m^{-2} day^{-1} were ~20% the rates of biological carbon uptake as measured by ^{14}C primary-production methods near the end of SOFeX (*4*). This POC export flux is consistent with an estimate of the balance between the drawdown of inorganic C and increase in stocks of POC in the SOFeX southern patch (*4*).

Our major findings are as follows: As the experiment progressed, more particulate ^{234}Th escaped below 50 m at the base of the iron-fertilized patch; also, the impact on particle flux extended into the deeper waters. This implies changes in the quantity and quality of sinking particles such that more particles were removed from the upper 50 m, and they were less prone to remineralization. Little is known about remineralization controls, but changes in sinking rate, density, or ballasting biominerals likely play a role (*8*, *9*). It is also possible that subsurface communities feeding on detrital particles responded to the change in food supply. These heterotrophic processes can enhance flux by converting smaller suspended particles into those that sink rapidly. However, there were no obvious subsurface changes in particle composition or heterotrophic activity during SOFeX, and therefore we cannot resolve the cause of this subsurface change in particle cycling.

In the context of previous blooms in this region, the POC flux during SOFeX was relatively small. During the same month in 1998 at this site, the ^{234}Th-derived POC flux at 100 m was 20 mM C m^{-2} day^{-1} (*7*). The export of POC and associated nutrients in 1998 was in close agreement with independent estimates of export determined by the seasonal balance of nutrient supply and export (*10*). POC fluxes at the OUT control stations in 2002 were only 3 to 5 mM C m^{-2} day^{-1} at 100 m, indicating considerable interannual variability in the magnitude and/or seasonality of POC flux. Measurements of much lower biogenic silica concentrations at the start of SOFeX (*11*) suggest that the lower POC flux OUT was associated with decreased diatom abundances, a siliceous plankton group that is thought to be a major vector of vertical carbon transport out of the surface ocean (*6*, *12*).

Although the POC flux IN at 100 m at the end of SOFeX was slightly lower than in 1998, and thus not large relative to the natural cycle, it was considerably enhanced relative

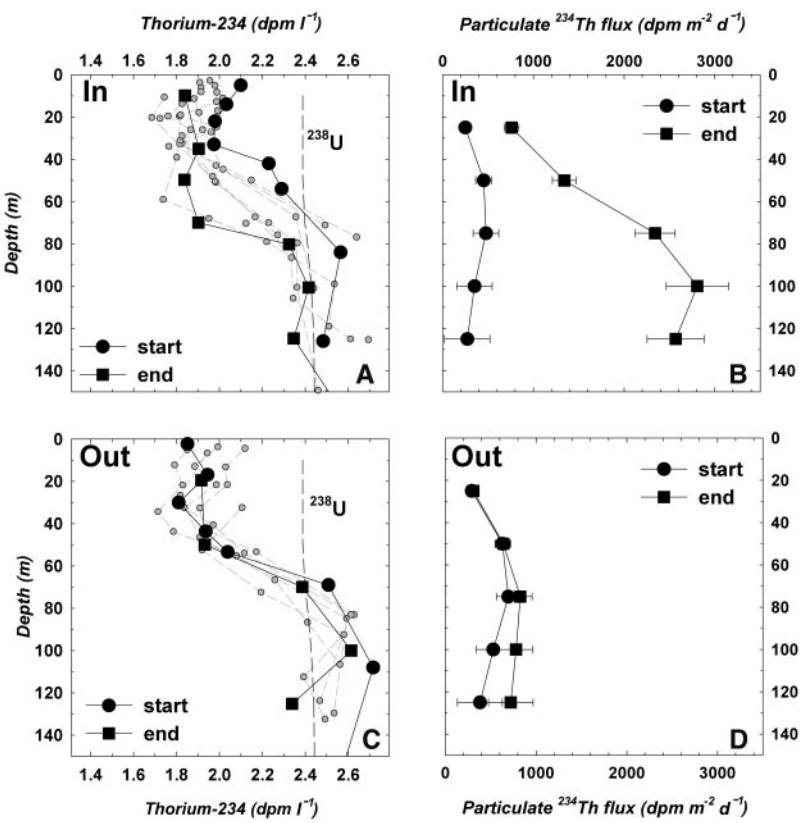

Fig. 1. Depth profiles of ^{234}Th activity (units: disintegrations per minute per liter = dpm l^{-1}) are shown for SOFeX "IN" (**A**) and "OUT" (**C**) stations. ^{238}U activity (dpm l^{-1}; dashed line) is determined by salinity (*29*). We used a newly developed ^{234}Th method (*22*) to quantify total ^{234}Th activities from 4-liter seawater samples collected by standard ocean water samplers (error on ^{234}Th = ±0.05 dpm l^{-1}). Data have been merged here between two ships, the RV *Melville* and USCG *Polar Star*, from 18 separate sampling casts; the first (start = 5.8 days after start of the first iron addition) and last (end = 27.4 days) ^{234}Th profiles are emphasized in bold to show temporal changes within this larger data set. A time sequence movie of these data can be found at *Science* Online. Also shown are the start and end particulate ^{234}Th flux profiles (dpm m^{-2} day^{-1}) for "IN" (**B**) and "OUT" (**D**) stations. ^{234}Th flux was calculated from a non–steady state model (*30*) and does not include physical transport, which, based on SF_6 data and observed activity gradients, would introduce <10 to 20% error.

to that of the OUT stations. That the POC fluxes were not higher IN may indicate that we did not reach the end of the SOFeX bloom. Observations and modeling suggest a 20- to 30-day lag between the onset of phytoplankton growth and export in these cold Southern Ocean waters (*7*, *13*). As in all previous iron experiments, SOFeX used multiple additions of iron (Fe was added starting on days 0, 4.3, 8.1, and 11.2), and thus our final observations were only ~17 days after the last iron addition.

Observations of enhanced iron concentrations, high photosynthetic efficiency, and sustained elevated biomass concentrations suggest strongly that the phytoplankton community was not iron stressed even at the end of our 28-day observation period (*4*). Without higher grazing pressure by zooplankton or limitation by other nutrients or physical controls, there was no obvious "trigger" for the crash of this iron-induced bloom. As such, we do not feel we reached the end of this SOFeX bloom.

It has been difficult in previous iron fertilization experiments in the Southern Ocean to demonstrate an obvious POC flux response, although in every case enhanced phytoplankton growth has been seen. SOIREE (Southern Ocean IRon Enrichment Experiment) took place in February 1999 at 61°S 140°E (*2*), and within a shorter, 13-day observation period, no measurable changes in POC flux were observed either by using the ^{234}Th method (*14*) or by comparing IN versus OUT differences during 2- to 3-day shallow sediment trap deployments (*15*). EisenEx ("Iron" Experiment) took place in November 2000 at 48°S 21°E (*16*), and 22 days of ^{234}Th observations showed no differences between activities IN versus OUT of the fertilized patch (*17*). EisenEx did result in a decrease over time in the sum of dissolved and particulate C stocks, and this was used to calculate a POC flux that was higher IN than OUT, in contrast to their ^{234}Th results (*17*).

SOFeX also conducted an experiment in the "northern patch" where silicate concentrations were much lower (<2 μM versus 60 μM in south). Peak primary production rates were roughly twice as high in the north, whereas chlorophyll a levels were somewhat lower (*4*). Our comparison of ^{234}Th early versus late in the northern patch did not show an obvious difference (*18*), but the data were only from one pair of ^{234}Th profiles and were hampered by the limited and more difficult sampling of the elongated and narrower northern patch (*4*). However, evidence for enhanced POC flux in the northern patch was obtained using an autonomous float to measure particle abundances. This approach provides a lower limit of 120 to 190 mM C m^{-2} for 100-m POC flux in the northern patch, and an upper limit of 760 to 1170 mM C m^{-2} after 50 days (*19*). In these same units our southern patch POC flux was 225 mM C m^{-2}, closer to the lower estimate. One difference is that the increase in POC flux in the north took place between patch days 25 and 45, demonstrating a lag between phytoplankton growth and POC flux that is longer than our 28-day observation period of bloom conditions in the south.

These few studies of the ocean's POC export response to iron have thus shown limited or no export response, or perhaps variability in POC export, such as found here between the southern and northern patch using different methods. Fundamentally, we expect particulate export to vary as a function of plankton community structure and its physiological status (*20*, *21*). Each iron fertilization experiment was conducted at a different time of the year in different biogeochemical settings in the Southern Ocean, so differences in the biological response and community structure might be expected.

Some of this apparent variability between studies may simply be related to the duration of observations, and/or data coverage and methods. Our new small-volume ^{234}Th method (*22*) allowed for high spatial and temporal coverage during SOFeX. Our conclusion here of enhanced particle export relies on subtle changes in the evolving ^{234}Th profiles that would not have been observed with the low-resolution ^{234}Th sampling techniques used during SOIREE and EisenEx.

Finally, physical differences in the experimental setting for each iron fertilization experiment should be considered when looking at variability in POC flux. Modeling suggests that the formation of sinking aggregates is sensitive to the rates of growth (slower in colder waters) and advective losses (*23*). The SOIREE patch experienced considerable stretching and dilution as the experiment progressed (*24*), whereas the southern SOFeX site maintained a generally coherent patch structure (*4*) more conducive to aggregation. The observation of a biomass decrease and surface POC flux increase after 1 to 2 weeks during the Equatorial Pacific iron fertilization experiment, IronEx II (*25*), was attributed to the four times higher ratio of growth to lateral advection seen in IronEx II relative to SOIREE.

In light of these SOFeX C-flux estimates, are geoengineering proposals for reducing the greenhouse effect through iron-enhanced carbon sequestration realistic? Using a patch size of 1000 km^2 (approximate area on 12 February with chlorophyll >0.4 mg m^{-3}), a time scale of 21 days (^{234}Th deficits increased during weeks 2 to 4), and the average difference in the IN versus OUT POC flux of 7 mM C m^{-2} day^{-1}, we estimate that SOFeX resulted in an enhanced flux at 100 m of 1800 tons of C in response to the addition of 1.26 tons of iron (*4*). However, because on average, >50% of the C carried on sinking particles at 100 m is remineralized by 250 m (*26*), the true estimate of C sequestration to depths below the more rapidly ventilated Antarctic Surface Waters (*27*) would be even lower, or only about 900 tons of C. When compared to the rates of carbon released globally due to human activities (6.5×10^9 tons/year), this C flux is indeed a small number. Also, with a $C_{sequestered}$:Fe_{added} ratio of 3.3×10^3 (molar ratio; export at 250 m; fluxes through day 28), it is difficult to see how ocean iron fertilization with such a low $C_{sequestered}$:Fe_{added} export efficiency would easily scale up to solve our larger global C imbalance problems (*28*).

One could argue that C flux increased after we left the site, or that there are other pathways of C export in addition to C flux on sinking particles measured here. For example, there are pathways of C loss due to accumulation and removal of dissolved organic C or suspended POC stocks. However, these processes involve physical mixing with

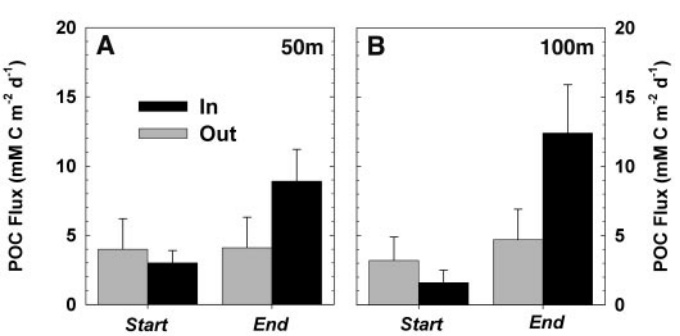

Fig. 2. POC flux data (mM C m^{-2} day^{-1}) from SOFeX determined at 50 m (**A**) and 100 m (**B**). Fluxes are shown for "IN" and "OUT" stations as indicated for the start and end of the experiment as defined in Fig. 1. POC flux is derived from the ^{234}Th flux shown in Fig. 1, multiplied by the POC/^{234}Th ratio determined on particles collected on 16 occasions during SOFeX at depths between 60 and 100 m using large-volume in situ filtration. Particles were separated in situ into two size classes, 1 to 54 μm and >54 μm. Here, we use the larger size class to calculate POC flux, where we measured a POC/^{234}Th ratio of 4.4 ± 1.0 "IN" and 6.0 ± 2.4 "OUT" (μM/dpm). This ratio at depth did not change significantly over the course of the experiment. Had we used the 1- to 54-μm size class, the POC fluxes would have been 15 to 20% higher "IN" and 15 to 20% lower "OUT."

surrounding shallow-water masses and are not efficient relative to direct sequestration of C on sinking particles to the deep ocean. We do not know whether the bloom eventually led to substantially higher C export after we left, or whether organic matter was remineralized within the surface ocean, resulting in no additional impact on C sequestration.

We have shown that iron addition to the Southern Ocean had a measurable impact on POC export, with flux increases observed not only within the iron-fertilized waters but also below the mixed layer in the "shadow" of the SOFeX patch. However, the magnitude of this flux increase was small relative to that of natural blooms in this region, or as needed to reduce current increases in atmospheric CO_2. To more accurately scale this flux response to iron as delivered in past climate cycles via dust or in proposed greenhouse mitigation schemes would require longer observations, comparisons between single versus multiple iron additions, deeper flux measurements, and larger patch scales to minimize artifacts due to dilution effects.

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Supporting Online Material

www.sciencemag.org/cgi/content/full/304/5669/414/DC1
Movie S1

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Robotic Observations of Enhanced Carbon Biomass and Export at 55°S During SOFeX

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Autonomous floats profiling in high-nitrate low-silicate waters of the Southern Ocean observed carbon biomass variability and carbon exported to depths of 100 m during the 2002 Southern Ocean Iron Experiment (SOFeX) to detect the effects of iron fertilization of surface water there. Control and "in-patch" measurements documented a greater than fourfold enhancement of carbon biomass in the iron-amended waters. Carbon export through 100 m increased two- to sixfold as the patch subducted below a front. The molar ratio of iron added to carbon exported ranged between 10^4 and 10^5. The biomass buildup and export were much higher than expected for iron-amended low-silicate waters.

The Southern Ocean plays a critical role in the global carbon cycle and in the regulation of levels of atmospheric CO_2 (*1*–*3*), yet the biological and physical processes that sequester carbon remain poorly understood largely because of the difficulties in making observations in waters surrounding Antarctica. The January to February 2002 SOFeX (*4*) was designed to examine the biological and carbon system response to the effects of purposeful addition of iron to nitrate-rich waters north and south of the Polar Front Zone (PFZ). Iron was added to two ~225 km^2 regions near 55°S 172°W and 66°S 172°W to test the hypothesis that the lack of dissolved silicate would limit both biomass growth and carbon export. Waters north and south of the PFZ had dissolved nitrate:silicate concentrations of 22:2.5 μM and 27:60 μM, respectively. The "North Patch" at 55°S was considered to be silicate limiting.

Three free-profiling robotic Lagrangian Carbon Explorers (*5*) were deployed at 55°S from the research vessel (RV) *Roger Revelle* to provide high-frequency (three times per day) profile observations of particulate organic carbon (POC) (*6*), Temperature (T) and Salinity (S) in the upper 1000 m; a fourth was deployed at 66°S. Between profiles, the SOFeX Explorers were "parked" at 100-m depth to investigate the systematics of carbon export at 100 m with an optically derived carbon flux index (CFI) (*7*).

Our "control" Carbon Explorer 1177 (*8*) was deployed ~20 km north of the planned North Patch location on 11 January 2002; it drifted northeast parallel to bathymetry as expected for the Antarctic Circumpolar Current. Two Explorers were deployed "in the patch" (Fig. 1A). Explorer 2054 was deployed on 12 January in the planned center of the North Patch; the first infusion of iron immediately followed. Explorer 2054 immediately tracked to the northwest, reflecting a current shear that soon split the initial 15 km by 15 km iron-fertilized patch in two. The northwest segment of the iron-fertilized water was sampled by Explorer 2054, and enhanced biomass levels were traced for 14 days; subsequent biomass profiles were typical of those observed by the control float. This segment was never sampled by SOFeX ships again. Explorer 2104 was deployed on 19 January (patch day 7) just prior to a second Fe infusion to the other "half" of the North Patch (*9*). It advected to the northeast on a course that closely paralleled that of the control (Fig. 1A). We refer to Explorer 2104 as the "in-patch" Explorer.

Over the following three weeks, the control (1177) and in-patch (2104) Explorers tracked each other closely, while relaying their surfacing positions, hydrography, and carbon mea-

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