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Matching carbon pools and fluxes for the Southern Ocean Iron Release Experiment (SOIREE)

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Abstract

The Lagrangian Southern Ocean Iron Release Experiment (SOIREE) allowed study of a gradually evolving iron-mediated phytoplankton bloom in water labelled with the inert tracer sulfur hexafluoride, SF_6. This article describes a pelagic carbon budget for the mixed layer in SOIREE and assesses the extent to which closure of the budget is achieved. Net community production (NCP) converted 837 mmol m^{-2} of inorganic carbon to organic carbon in 12.0 d after the first iron addition. A large fraction (41%) of NCP remained as particulate organic carbon in the mixed layer of the iron-enriched patch, while 23% was lost by horizontal dispersion and 0–29% was exported. The closure of the carbon budget is hampered by the lack of measurements of dissolved organic carbon (DOC), by a major uncertainty in carbon export, and by use of empirical conversion factors in estimates of carbon biomass and metabolic rates. Lagrangian carbon-budget studies may be improved by direct measurement of all major carbon parameters and conversion factors. Carbon cycling in the SOIREE bloom resembled that in 'natural' algal blooms in the open Southern Ocean in some respects, but not in all. Daily NCP in the SOIREE bloom (70 mmol m^{-2} d^{-1}) was higher than in natural blooms, partly because other studies did not account for horizontal dispersion, were for longer periods or included less productive areas. The build-up of POC stock and carbon export as a fraction of NCP in SOIREE were in the lower range of observations elsewhere.

Keywords: Iron enrichment; SOIREE; Carbon budget; Lagrangian

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1. Introduction

Net community production (NCP) determines the potential for carbon export from oceanic surface waters (Hansell and Carlson, 1998). Carbon export may occur in sinking particles, by respiration of vertically migrating zooplankton, or as dissolved organic carbon (DOC) transported away from the sea surface (Martin et al., 1987; Murray et al., 1996; Hansell and Carlson, 1998). A carbon budget of the mixed layer allows assessment of individual carbon pools and fluxes in the productive surface waters and of carbon exchanges between the surface ocean, the atmosphere, and the deep ocean. Closure of such carbon budgets is particularly challenging (Michaels et al., 1994; Fasham et al., 1999).

Carbon export, primary production, respiration, grazing, and horizontal dispersion of carbon stocks are often difficult to quantify. For example, measurements of ^{14}C uptake and ^{18}O production do not directly correspond to either gross primary production, net primary production (NPP) or NCP (Bender et al., 1999). Estimates for heterotrophic respiration and zooplankton grazing require sample incubation and empirical conversions, which introduce considerable uncertainty in the final values.

Lagrangian experiments, in which water is labelled with an inert tracer, provide a rare opportunity for assessing the effect of horizontal dispersion on carbon dynamics. The Lagrangian Southern Ocean Iron Release Experiment (SOIREE) was carried out in February 1999 south of the Antarctic Polar Front (APF). SOIREE had the objective to test whether low iron concentrations limit phytoplankton growth in these HNLC (high nutrients–low chlorophyll) waters. Four iron additions promoted the development of an algal bloom and resulted in major biogeochemical changes during the 13-d experiment (Boyd et al., 2000; Watson et al., 2000).

In this article, we present a carbon budget for the mixed layer, which highlights the changes in carbon cycling upon the SOIREE iron additions. NCP, the balance between primary production and respiration in a system (Williams, LeB., 1993), is a key component of the carbon budget, as it allows a comparison between changes in the organic and inorganic carbon pools (Bakker et al., 1997; Sweeney et al., 2000). The carbon budget provides the basis for a discussion of the uncertainties in the carbon parameters and how these might be reduced in future. We address the extent to which carbon cycling in the SOIREE bloom mimicked that in 'natural' blooms in the Southern Ocean.

2. Methods

2.1. Site and experimental design

Iron (768 kg Fe) and inert tracer SF_6 (165 g) were added to the mixed layer at 61°S 140°E, south of the APF (Boyd et al., 2000; Boyd and Law, 2001). The Lagrangian release was done across an expanding hexagon over an area of ~50 km^2. Additional iron was added after 3, 5 and 7 d (312, 312 and 353 kg) (Boyd and Law, 2001). Times (in days) are adjusted relative to the midpoint of the first iron addition on 9 February at 12:00 UTC. The surface water SF_6 concentration was used as a tracer for the iron-enriched waters (Law et al., 2003). Daily CTD profiling was carried out inside and outside the iron-enriched patch for 12 d. Samples for most biogeochemical variables were taken from the CTD rosette, which had 12 Niskin bottles of 12 l each. Generally three CTD casts, closely spaced in time and position, were needed to meet the water requirements at each station.

Carbon stocks and fluxes were measured (directly) or estimated (indirectly). Many parameters have been reported before, although not necessarily for exactly the same times or in the same detail. A summary of the sampling and analysis of these parameters is provided here and in Appendix A. These data are available on the CD-ROM Appendix of Law et al. (2001). A more extensive description is given for data that have not been reported before, and these are marked in the text by (*new data*). The compilation of the data is presented for the first time. The relative error (RE) of each parameter was determined, either from the accuracy of the measurements or from uncertainty in the estimation procedure, as described in Appendix A.

2.2. Inorganic carbon parameters

Continuous measurements of the fugacity of CO_2 (fCO_2) in surface water were used for the construction of daily fCO_2 maps on a 0.5 km × 0.5 km grid (Bakker et al., 2001). Fits for surface water fCO_2 corresponding to the 10% lowest and highest SF_6 values for each day were taken from Watson et al. (2000) (Fig. 1). Data points in or above the interval of the upper fit are outside the iron-enriched patch (OUT), data below it inside the patch (IN). Data

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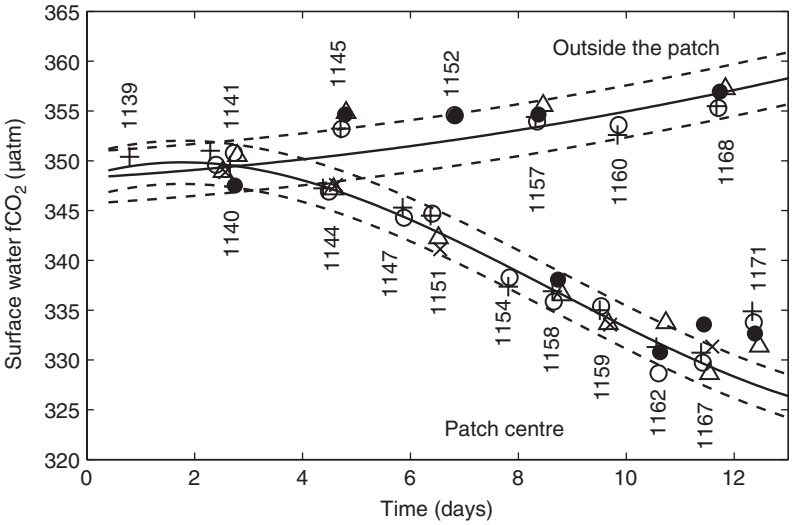

Fig. 1. Surface water fCO_2 during closure of the upper Niskin bottle for CTD casts. The bold lines, which indicate fits of fCO_2 at the 10% lowest (upper line) and highest (lower line) SF_6 values, have been taken from Watson et al. (2000). The dashed lines correspond to 84% and 74% of the root mean square error of the fits (Bakker et al., 2005). Data points in or above the interval around the upper bold line are outside the patch. Data in or below the lower interval are in the patch centre. Station numbers range from 1139 to 1171. Symbols indicate cast numbers: 1 (+), 2 (o), 3 (●), 4 (Δ), and 5 (x).

points in or below the interval around the lower fit are in the patch centre (IC). Surface-water fCO_2 during the closure of the upper Niskin bottle on the CTD helps one to evaluate the position of CTD casts relative to the biogeochemical signature of the iron-enriched patch (Fig. 1). Several IN casts were outside the patch centre towards the end of the experiment.

The concentration of dissolved inorganic carbon (DIC) was determined by coulometry on samples from the CTD and the surface-water supply (Bakker et al., 2001). The net DIC change in the mixed layer of the patch centre was calculated by two methods: directly by using the DIC data from the CTD (ΔDIC_{DIC}) and indirectly by using the fCO_2 maps (ΔDIC_{fCO_2}), following the method in Bakker et al. (2001, 2005).

For calculation of ΔDIC_{fCO_2} a fit between fCO_2 and DIC from the ship's surface supply was made ($DIC = 0.43\ fCO_2 + 1984.1$, $r = 0.89$, root mean square error $= 2.6\,\mu mol\,kg^{-1}$, 83 data points) (Fig. 6 in Bakker et al., 2001). The DIC change at each grid point on an fCO_2 map was taken as the difference between DIC calculated from the fCO_2 at the grid point and DIC at fCO_2 corresponding to the 10% lowest SF_6 values for that map (Bakker et al., 2001). Daily vertical profiles of DIC indicate that changes in DIC uniformly occurred in the upper 50 m, while DIC did not change significantly between 50 and 65 m depth (Bakker et al., 2001). Thus, the average DIC change across the mixed layer (ΔDIC_{fCO_2}) was calculated for the grid points in the patch centre by multiplication of the surface water DIC change with a 50 m depth interval (Bakker et al., 2001).

The effect of CO_2 air–sea exchange on DIC (ΔDIC_{AirSea}) was determined for grid points on the fCO_2 maps in the patch centre (Bakker et al., 2001). The quadratic relationship for short-term, ship-board wind speed (Wanninkhof, 1992) was used for the calculation of the gas transfer velocity from average wind speed for each mapping period. Vertical diffusion of DIC across the pycnocline (ΔDIC_{VDif}) was calculated from the DIC gradient between 50 and 90 m depth and a vertical diffusivity of $0.11\,cm^2\,s^{-1}$ ($\pm 0.2\,cm^2\,s^{-1}$) (Law et al., 2003; Bakker et al., 2005).

2.3. Organic carbon pools and fluxes

2.3.1. Particulate organic carbon

The stock of particulate organic carbon (POC) was determined by three methods. A moving pump attached to the CTD-frame collected 400 l of water through the upper 100 m, which was passed through two filters (70 µm, 1.2 µm) for the analysis of POC (POC_M) (Charette and Buesseler, 2000). A second, submersible pump collected water at 10, 30, 60, and

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100 m depth during and between CTD casts. The pump returned several hundreds of litres of water via a hose to filtration devices on deck for analysis of POC (POC_H) (Trull and Armand, 2001). The water was filtered through a 300-μm screen and a pre-combusted GF/F filter (~0.7 μm). The filters from both pumps were fumed overnight with concentrated hydrochloric acid (HCl). The POC content on the filters was determined by CHN analysis.

The particulate carbon concentration was determined in water collected from several depths by Niskin bottles on the CTD rosette. Two litres of sample were filtered through a pre-combusted GF/F filter. The filter was stored frozen. The carbon content on the filters was determined by CHN analysis. The filters were not fumed with HCl prior to analysis and the filter blank was not determined. The absence of calcifying algae in microscopic observations (Gall et al., 2001a) and of their remains in size-fractionated particle samples (Trull and Armand, 2001) indicates that particulate inorganic carbon (PIC) was not present in significant amounts. Particulate carbon stock from the Niskin bottles is taken equal to particulate organic carbon (POC_{Nis}) (*new data*).

Water-column transmissivity was determined with a SeaTech beam transmissometer (0.25 m path length, $\lambda = 660$ nm) attached to the CTD frame. Percent transmission was converted to the particle beam attenuation coefficient (c_p) after Gardner et al. (1993). Linear regression (POC_{Nis} (mmol m^{-3}) = 34.8 c_p (m^{-1}), $r^2 = 0.72$, 101 values) was used for the conversion of c_p into POC_{cp} (*new data*).

2.3.2. Particulate carbon export

The export of particulate carbon was determined by thorium-234 depletion over the upper 100 m (Charette and Buesseler, 2000) and by floating sediment traps at 110 m depth (Nodder and Waite, 2001) (Appendix A).

2.3.3. Carbon biomass

Carbon biomass is defined here as the carbon content of living biota. Samples for bacteria and small plankton were taken from several depths by the CTD rosette. Mesozooplankton were collected by net tows from an average depth of 91 m to the surface (Zeldis, 2001). The abundance of bacteria and plankton species was determined by appropriate techniques (Gall et al., 2001a; Hall and Safi, 2001; Zeldis, 2001) (Appendix A). The carbon biomass of the biota was estimated from species abundance, the dimensions of individuals (e.g. shape, diameter, length) and algorithms for the conversion of these parameters to carbon biomass. The mesozooplankton stock in the upper 65 m was determined by applying a depth correction to the carbon biomass from the tow samples. Total carbon biomass in the upper 65 m was calculated from the carbon biomass of all biota. The plankton were assigned to size classes: pico (0.2–2 μm), nano (2–20 μm), micro (20–200 μm), and meso (>200 μm) (Sieburth et al., 1978).

2.3.4. Bacterial and plankton growth parameters

Bacterial production (BP) was determined by methyl-^{3}H thymidine incorporation (Hall and Safi, 2001). Bacterial growth efficiency (BGE) is the ratio of BP and bacterial carbon demand (BCD):

$$BGE = BP/BCD = BP/(BP + BR) \quad (1)$$

(Carlson et al., 1999). Bacterial respiration (BR) was calculated from BP and BGE. A value of 30% was taken for BGE, similar to observations of 26% and 30% south of the APF at 6°W (Kähler et al., 1997).

Carbon 14-uptake was determined in 24-h deck incubations at light levels matching the sampling depths (Gall et al., 2001b). A dark correction was applied. The ^{14}C data of Gall et al. (2001b) were corrected with a factor 12/14.

Nano- and microzooplankton grazing was determined by the dilution technique after 12 d in the patch centre (Hall and Safi, 2001). Nano- and microzooplankton respiration was estimated after 0 d by assuming that net DIC production from NPP, heterotrophic respiration, vertical diffusion and air-sea exchange was zero, and that ^{14}C uptake was a proxy for NPP. Grazing and respiration of nano- and microzooplankton throughout SOIREE was estimated by scaling the values for grazing (12.0 d) and respiration (0.0 d) to the carbon biomass of these zooplankton.

Mesozooplankton grazing and egestion were determined by 24-hour incubation of large copepods with prey (Zeldis, 2001). Respiration of large copepods was calculated as the difference between grazing and egestion, under the assumption that excretion and changes in mesozooplankton biomass were negligible. Grazing and respiration of all mesozooplankton were estimated by multiplying the values for large copepods by a factor 2 (after Zeldis, 2001).

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3. Results

3.1. A carbon budget for SOIREE

The SOIREE iron additions promoted an algal bloom, which resulted in the conversion of dissolved inorganic carbon into organic carbon (Figs. 2a, c, e, 3a and c). The mixed-layer depth was roughly 60 m throughout SOIREE with occasional deepening to 74 m (Law et al., 2003). No changes were observed in carbon parameters below 65 m or outside the patch (Figs. 2b, d, f, 3b and d). Here we quantify the changes in carbon cycling upon iron addition and prepare a carbon budget for the upper 65 m of the patch centre (Fig. 4). Linear interpolation between values at different sampling depths is used to

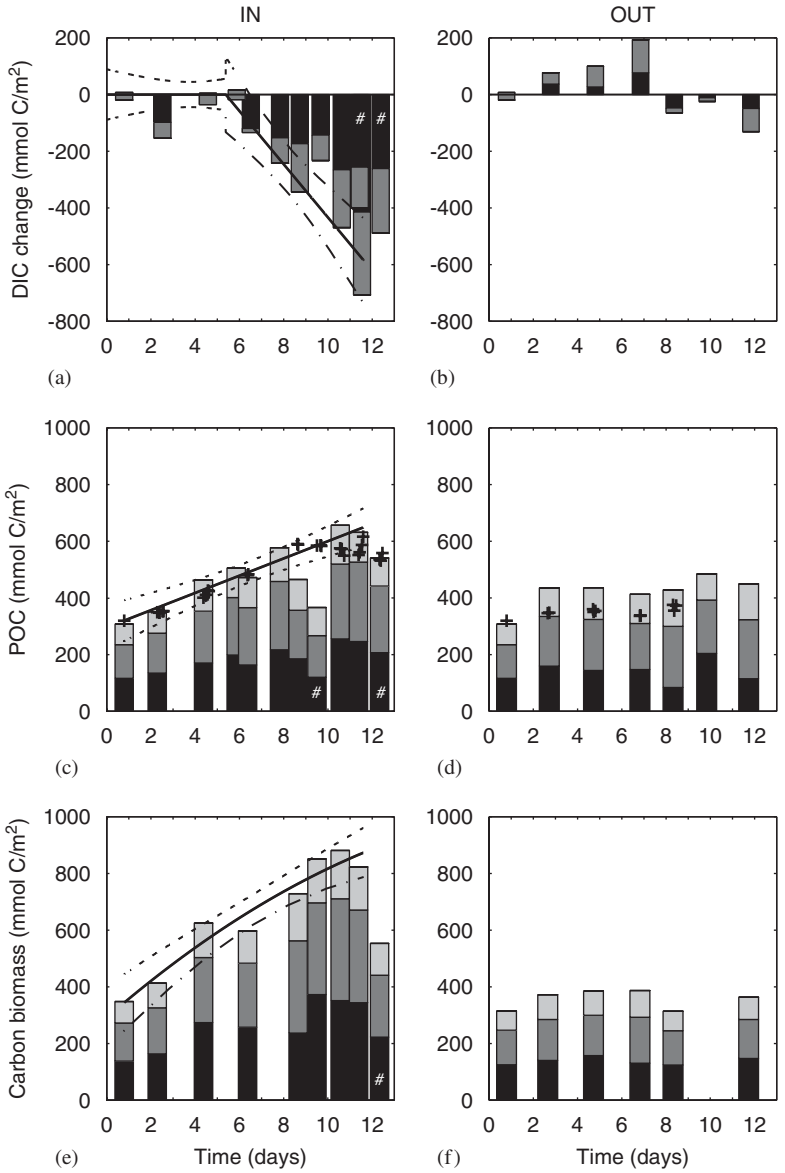

Fig. 2. (a, b) Changes in dissolved inorganic carbon (DIC), (c, d) particulate organic carbon (POC_{Nis}), and (e, f) carbon biomass for 0–65 m depth at CTD stations inside (IN) and outside (OUT) the iron-enriched waters in SOIREE. The values have been integrated for the upper 25 m (black bars), 25–50 m (dark grey) and 50–65 m (light grey). Changes in DIC are shown for the upper 50 m only, relative to an arbitrary level of 109736 mmol C m^{-2}. Fits to the data (bold lines) are shown with 95% confidence intervals (dashed lines) (Table 1). Casts with a hash symbol (#) have been excluded from the fits. The fits for DIC and POC_{Nis} are used in the carbon budget. (c, d) Plus symbols (+) indicate POC_{cp}.

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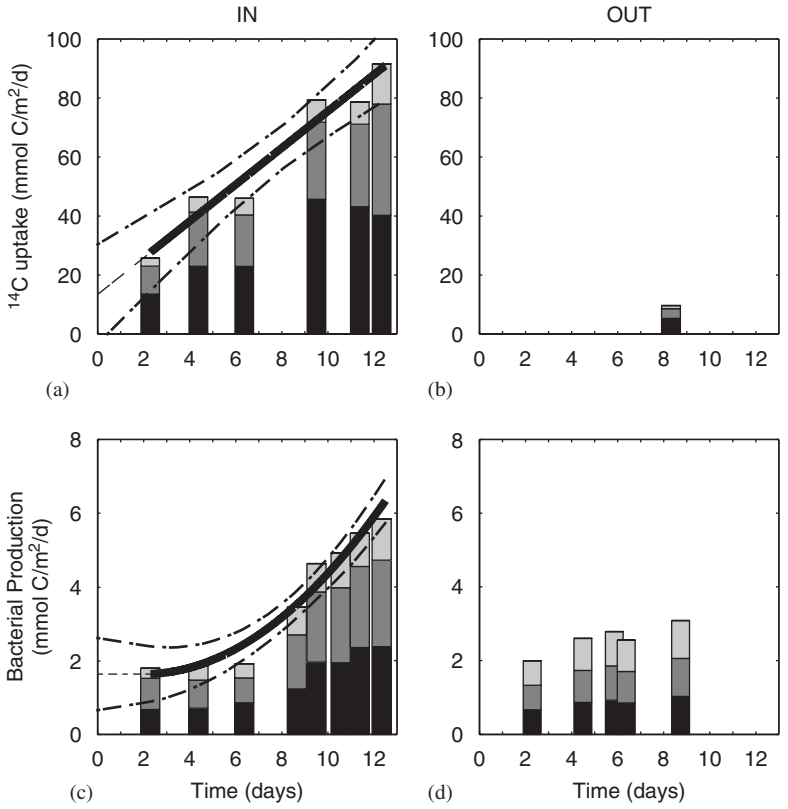

Fig. 3. Evolution of (a, b) ^{14}C uptake and (c, d) bacterial production for the upper 65 m at CTD stations inside and outside the patch. The rates have been integrated for the upper 25 m (black bars), 25–50 m (dark grey) and 50–65 m (light grey). Fits to the data (bold lines) are shown with 95% confidence intervals (dashed lines) (Table 1).

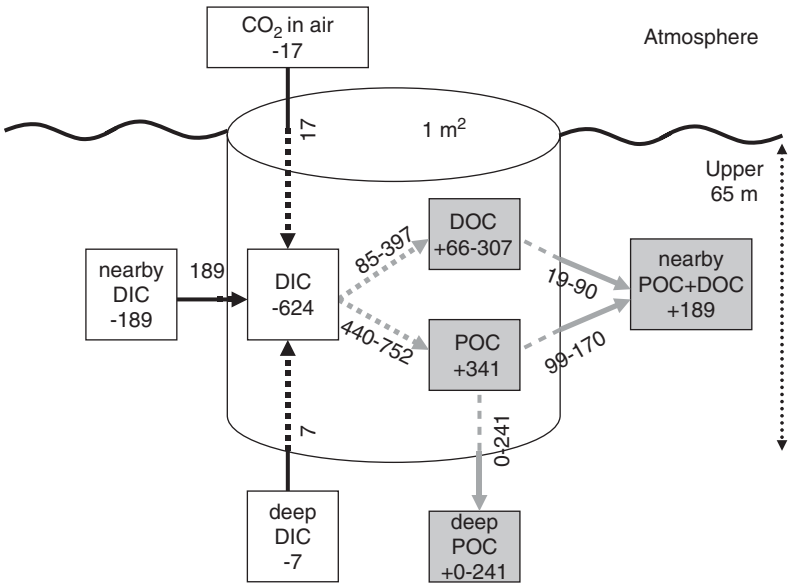

Fig. 4. Carbon transfers between the inorganic and organic carbon pools in the upper 65 m of the iron-enriched patch in SOIREE, as well as carbon exchanges with adjacent reservoirs. The values correspond to the carbon flows and changes in carbon pools after 12.0 d (in mmol m^{-2}, Eqs. (2) and (3)). The abbreviations are as in the text.

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integrate the carbon parameters across the upper 65 m. Fits to the CTD data over time describe the evolution of carbon stocks and fluxes during the experiment (Table 1). First order fits are used, unless the temporal change is clearly non-linear.

3.2. Inorganic carbon pools and fluxes

Two independent estimates for DIC change, from CTD samples and from fCO_2 maps ($\Delta DIC_{fCO_{2w}}$), show a DIC reduction in the patch centre within 4–6 d of the first iron addition (Figs. 2a and 5a). The DIC change was relatively small for the CTD casts in the iron-enriched waters after 11.5 and 12.3 d (casts 1167/3 and 1171/3) (Fig. 2a), primarily because these IN casts were outside the patch centre, as seen in fCO_2 (Fig. 1). A fit has been made to the DIC data from the CTD in the patch centre (Fig. 2a; Table 1). Such fits allow comparison of variables with different sampling times, reflect the overall trend in the parameters, and reduce the impact of outliers. There is good correspondence between DIC changes from the fit to the CTD data (ΔDIC_{DIC}) and those based on the fCO_2 maps (Fig. 5a).

The total DIC change by NCP ($\Delta DIC_{NCP\_fCO_2}$) has been estimated for the upper 65 m of the patch centre from the net DIC change based on the fCO_2 maps (ΔDIC_{fCO_2}) and from changes in DIC by vertical diffusion (ΔDIC_{VDif}), by air-sea exchange (ΔDIC_{AirSea}), and by horizontal dispersion (ΔDIC_{HDis}):

$$\begin{aligned}\Delta DIC_{NCP\_fCO_2} &= \Delta DIC_{fCO_2} - \Delta DIC_{VDif} \\ &\quad - \Delta DIC_{AirSea} - \Delta DIC_{HDis} \\ &= -NCP_{DIC} \end{aligned} \quad (2)$$

(Bakker et al., 2005). Multiplication of $\Delta DIC_{NCP\_fCO_2}$ by minus one gives NCP (NCP_{DIC}). The error analysis by Bakker et al. (2005) is adopted (Appendix A).

After 12.0 d, DIC based on the fCO_2 maps (ΔDIC_{fCO_2}) had decreased by 624 mmol m^{-2} ($\pm$4%) in the upper 65 m (Figs. 4 and 5b). Air–sea exchange and vertical diffusion had little effect on DIC, while horizontal dispersion had added 189 mmol m^{-2} ($\pm$74%) of DIC to the patch centre. The total reduction of DIC by NCP ($\Delta DIC_{NCP_fCO_2}$) was 837 mmol m^{-2} ($\pm$21%).

3.3. Organic carbon pools and fluxes

3.3.1. Particulate carbon export

The two techniques for measuring carbon export from the SOIREE patch gave different results: zero carbon export with an upper limit of 7.2 mmol m^{-2} d^{-1} from ^{234}Th depletion in the upper 100 m (Charette and Buesseler, 2000) and an average carbon export of 12.8 mmol m^{-2} d^{-1} from the sediment traps at 110 m depth (Nodder and Waite, 2001). The latter value is the average of three trap deployments in the patch centre: 12.2 mmol m^{-2} d^{-1} (between 0 and 2 d or T0-2), 10.5 mmol m^{-2} d^{-1}

Table 1
Coefficients of fits with the equation $y = a_2t^2 + a_1t + a_0$ to CTD data (y) in the upper 65 m at IN stations for time t (in days) in SOIREE (Figs. 2 and 3)

Parameter	ΔDIC_I (mmol m^{-2})	ΔDIC_{II} (mmol m^{-2})	POC_{Nis} (mmol m^{-2})	Biomass (mmol m^{-2})	^{14}C uptake (mmol m^{-2} d^{-1})	BP (mmol m^{-2} d^{-1})
a_2	0	0	0	−1.51	0	0.0426
a_1	0	−93.9	30.4	67.7	13.4	−0.170
a_0	0	505.4	296	291	6.2	1.8
RMS	17.9	96.6	46.8	58.2	6.4	0.40
#	3	8	9	8	6	8
r	—	−0.93	0.93	0.97	0.97	0.95
Period (d)	0.8–5.4	5.4–11.6	0.8–11.6	0.8–11.6	2.3–12.3	2.3–12.3
In fit	1139–1147	1144–1167/4	1139–1167	1139–1167	1140–1171	1140–1171
Not in fit	1140	1167/3, 1171	1159, 1171	1171	—	—

The parameters are dissolved inorganic carbon (DIC), particulate organic carbon (POC_{Nis}), carbon biomass, ^{14}C uptake and bacterial production (BP). The root mean square error (RMS), the number of data points (#) and the correlation coefficient (r) are shown. Values at some stations are excluded from the fits, partly because they were outside the patch centre, as seen in fCO_2 (Fig. 1). We assume that no significant changes in the carbon parameters occurred within the first 0.8 d.

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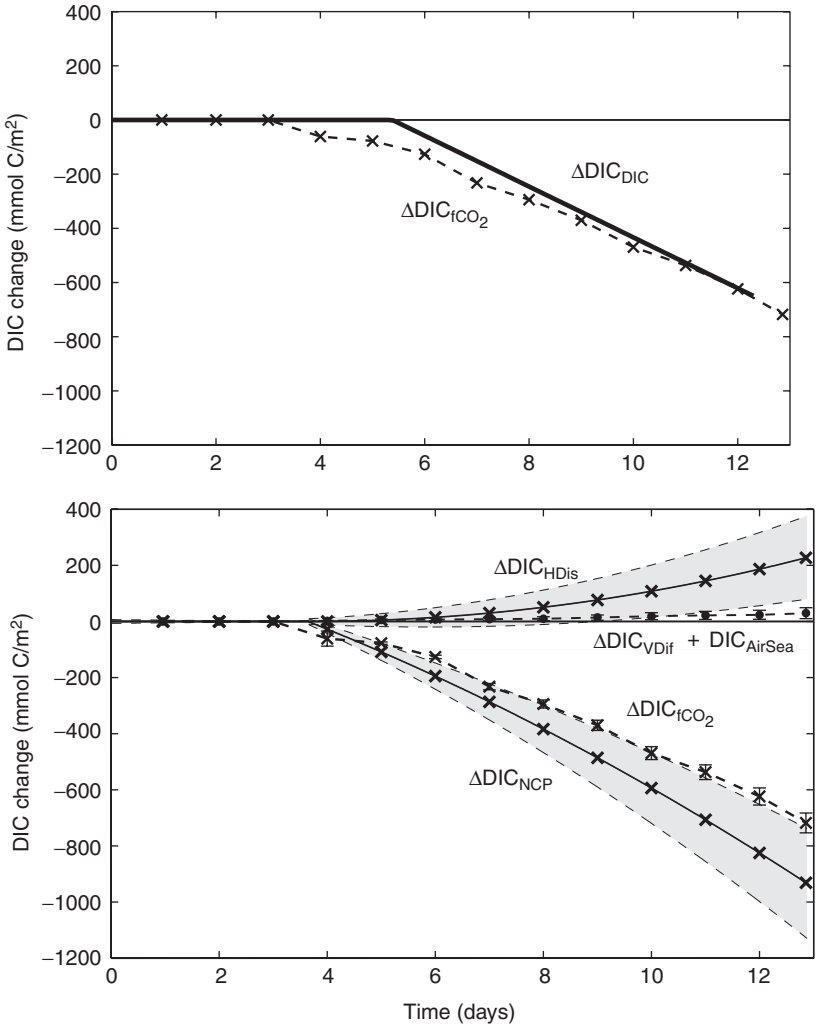

Fig. 5. (a, b) Changes in dissolved inorganic carbon (DIC) in the upper 65 m of the patch centre during SOIREE. (a) The net DIC change has been determined from surface water fCO_2 maps (ΔDIC_{fCO_2}) (- - x- -) and from the fit to the DIC change over time in CTD samples (ΔDIC_{DIC}) (solid line) (as in Fig. 2a, Table 1). (b) Comparison of the net DIC change from surface water fCO_2 maps (ΔDIC_{fCO_2}) (- - x- -) with changes in DIC by horizontal dispersion (ΔDIC_{HDis}) (upper line with crosses), vertical diffusion (ΔDIC_{VDif}) and CO_2 air-sea exchange (ΔDIC_{AirSea}) (shown together as dashed line with dots) allows calculation of the DIC change by net community production (ΔDIC_{NCP}) (lower line with crosses) (Eq. (2)). The error bars and error intervals (grey shading) indicate the uncertainty in the data and the fits.

(T7–9), and 15.7 mmol m^{-2} d^{-1} (T11-13) (Nodder and Waite, 2001). The thorium values convert to a carbon export of 0–10.4 mmol m^{-2} d^{-1} from 65 m depth with the relationship of Martin et al. (1987). Average carbon export from the sediment traps corresponds to 20.1 mmol m^{-2} d^{-1} at 65 m depth with the same equation.

3.3.2. Particulate organic carbon

The POC_{Nis} stock had increased by 330 mmol m^{-2} in the upper 65 m of the patch centre after 12.0 d (Fig. 2c). The fit to the POC_{Nis} data is used as the best estimate for POC stock in the carbon budget (Fig. 2c; Table 1). The POC values based on water-column transmissivity (POC_{cp}) reflect the general trend better than POC_{Nis} (Fig. 2c). The POC stock from the Niskin bottles exceeded that from the pumps by an average 13–17% in the upper 100 m (average POC_{Nis}–POC_H = 82 mmol m^{-2}, σ_{n-1} = 110 mmol m^{-2}, 7 values; average POC_{Nis}–POC_M = 106 mmol m^{-2}, σ_{n-1} = 106 mmol m^{-2}, 10 values).

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Table 2
Carbon biomass for bacteria and plankton classes in the upper 65 m at CTD stations inside the iron-enriched patch in SOIREE

Time (d)	Bacteria ($mmol\,m^{-2}$)	Pico Phyto ($mmol\,m^{-2}$)	Nano Phyto ($mmol\,m^{-2}$)	Micro Phyto ($mmol\,m^{-2}$)	Nano Zoo ($mmol\,m^{-2}$)	Micro Zoo ($mmol\,m^{-2}$)	Meso Zoo ($mmol\,m^{-2}$)	Total Biomass ($mmol\,m^{-2}$)
0.8	23	25	41	42	46	4	167	348
2.3	24	49	56	57	55	4	167	413
4.4	23	48	185	118	81	3	167	625
6.4	24	78	216	61	48	3	167	597
8.7	17	17	167	279	80	3	167	729
9.5	20	76	252	217	114	4	167	851
10.6	24	46	(295)	225	(119)	5	167	881
11.4	21	37	336	137	122	3	167	823
12.3	23	18	95	160	84	6	167	553

Brackets indicate interpolated values.

3.3.3. Carbon biomass

Carbon biomass reached $881\,mmol\,C\,m^{-2}$ in the upper 65 m after 10.6 d (Fig. 2e; Table 2). The response to the iron enrichment differed between trophic levels. Algal carbon biomass increased 6-fold, mainly by a higher biomass of autotrophic nanoflagellates and diatoms.

By definition, POC stock, which includes both living and dead material, should exceed carbon biomass in the particulate size range, i.e. larger than ~1 μm. 'Particulate' carbon biomass was calculated by assuming that bacteria were smaller and pico-phytoplankton were larger than 1 μm in size. Particulate carbon biomass and POC_{Nis} stock were similar in size outside the patch. However, biomass exceeded POC_{Nis} by $94\,mmol\,m^{-2}$ ($\sigma_{n-1} = 81\,mmol\,m^{-2}$, seven values) inside the patch, even with two stations with low POC_{Nis} excluded. The difference is within the uncertainty of both parameters (REs of 17% in POC_{Nis} and of 50% in carbon biomass) (Appendix A). Size fractionation suggests that mesozooplankton biomass has been overestimated or that mesozooplankton are underrepresented in the POC stock.

3.3.4. The fate of NCP

Net community production (NCP_{DIC}) resulted in the production of POC and DOC. Some of the organic particles were exported from the upper 65 m, while horizontal dispersion also removed organic carbon from the patch centre. Eq. (3) relates the changes in organic carbon stocks in upper 65 m of the patch centre (ΔPOC_{Nis}, ΔDOC) to NCP:

$$NCP_{DIC} = \Delta POC_{Nis} + \Delta DOC + Export + \Delta DIC_{HDis} \quad (3)$$

(Fig. 6). We estimate carbon export from the upper 65 m for three scenarios (Table 3). These correspond to the lower (A) and upper (B) limits of $0–10.4\,mmol\,m^{-2}\,d^{-1}$ from the thorium method and to (C) carbon export of $20.1\,mmol\,m^{-2}\,d^{-1}$ from the sediment traps. The removal of newly produced organic carbon from the patch centre by horizontal dispersion is taken equal to the replenishment of DIC from neighbouring waters (ΔDIC_{HDis}). This assumption neglects the gradual loss of organic carbon from the mixed layer by carbon export. The change in DOC stock is calculated as the residual of the other terms in Eq. (3).

Fig. 6 illustrates the fate of NCP for scenario A with zero carbon export. After 12.0 d 41% of NCP remained as POC ($341\,mmol\,C\,m^{-2}$) in the upper 65 m of the patch centre, while 0–29% had been exported ($0–241\,mmol\,C\,m^{-2}$), and 23% ($189\,mmol\,C\,m^{-2}$) had been removed by horizontal dispersion (Fig. 4; Table 3). The scenarios suggest that 8–37% of NCP was transferred into DOC.

3.3.5. Organic carbon fluxes

Carbon-14 uptake rapidly increased upon iron addition, while BP increased from 7 d onwards (Fig. 3a and c). Grazing by nano- and microzooplankton was $91\,mmol\,C\,m^{-2}\,d^{-1}$ after 12 d (Hall and Safi, 2001). Respiration by this plankton has been estimated as $14\,mmol\,C\,m^{-2}\,d^{-1}$ for the same day. Mesozooplankton grazing and respiration were constant at 3.0 and $2.4\,mmol\,C\,m^{-2}\,d^{-1}$, respectively (Zeldis, 2001).

The DIC change by NCP (ΔDIC_{NCP_bio}) is calculated from the DIC change by ^{14}C uptake ($\Delta DIC_{^{14}C}$), bacterial respiration (ΔDIC_{BR}), zooplankton respiration (ΔDIC_{ZR}), and horizontal

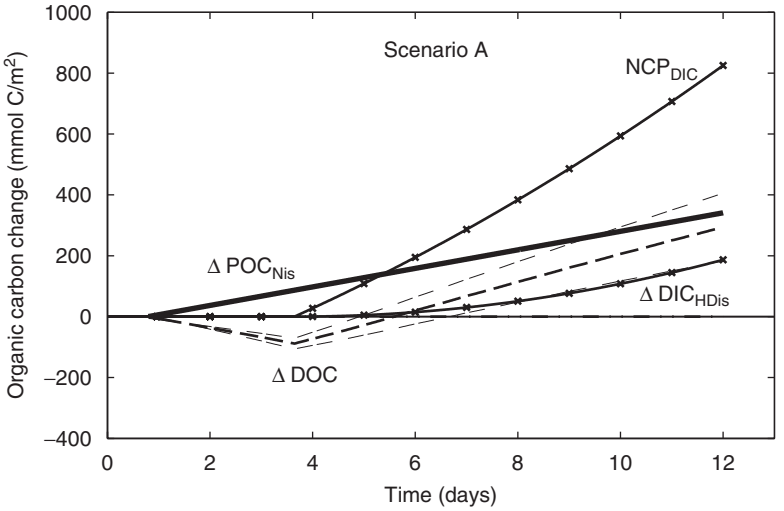

Fig. 6. The fate of net community production (NCP_{DIC}, upper line with crosses) in the upper 65 m of the patch centre during SOIREE for scenario A with zero carbon export. The bold line indicates changes in POC_{Nis}, as given by the fit to the CTD data in Fig. 2c (Table 1). Horizontal dispersion (ΔDIC_{HDis}, lower line with crosses, as in Fig. 5b) and carbon export (here taken as zero) remove organic carbon from the upper 65 m of the patch centre. Eq. (3) allows calculation of the change in DOC (dashed line, error intervals as thin dashed lines).

Table 3
The fate of net community production (NCP_{DIC}) in the upper 65 m of the SOIREE patch is calculated with three scenarios for carbon export after 12.0 d (Eq. (3))

Scenario	NCP_{DIC} (mmol m^{-2})	ΔDIC_{HDis} (mmol m^{-2})	Export (mmol m^{-2})	ΔPOC_{Nis} (mmol m^{-2})	ΔDOC (mmol m^{-2})
A	837 ± 176	189 ± 140	0	341 ± 61	307 ± 111
B	837 ± 176	189 ± 140	125	341 ± 61	182 ± 111
C	837 ± 176	189 ± 140	241 ± 49	341 ± 61	66 ± 160

The scenarios correspond to the lower (A) and higher (B) estimates from the thorium method and (C) average carbon export from the floating sediment traps after correction to the upper 65 m. Fig. 6 depicts the results for scenario A.

dispersion of biota (ΔDIC_{HDis_bio}), with ^{14}C uptake taken as a proxy for NPP:

$$\Delta DIC_{NCP\_bio} = \Delta DIC_{^{14}C} + \Delta DIC_{BR} + \Delta DIC_{ZR} + \Delta DIC_{HDis\_bio}. \quad (4)$$

The effect of horizontal dispersion is determined for the sum of the first three terms in Eq. (4). Alternatively the DIC change by NCP ($\Delta DIC_{NCP\_^{14}C}$) may be calculated with ^{14}C uptake as a proxy for NCP and horizontal dispersion ($\Delta DIC_{HDis\_^{14}C}$):

$$\Delta DIC_{NCP\_^{14}C} = \Delta DIC_{^{14}C} + \Delta DIC_{HDis\_^{14}C}. \quad (5)$$

The calculations give ΔDIC_{NCP_bio} and $\Delta DIC_{NCP\_^{14}C}$ as 64% and 99% of $\Delta DIC_{NCP\_fCO_2}$, respectively, after 12.0 d. This suggests that ^{14}C uptake was a better proxy for NCP than for NPP during SOIREE.

4. Discussion

4.1. Optimisation of carbon budgets

4.1.1. Lagrangian experiments

The SOIREE carbon budget contains ample uncertainties, despite the textbook-like evolution of the algal bloom. Here we discuss the strong and weak points in the budget and suggest how uncertainties may be reduced in future studies. The carbon budget has strongly benefited from the Lagrangian nature of the experiment. The addition of SF_6 tracer allowed accurate quantification of horizontal dispersion. The pre-site survey and frequent OUT stations demonstrated little spatial or temporal variation in the biogeochemical properties of the surrounding waters. The initiation of the bloom by iron addition meant that biogeochemical changes in the seawater, labelled with an inert

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tracer, occurred relative to a near steady-state situation. The bloom's gradual evolution and the continuous measurements of surface water SF_6 and fCO_2 greatly facilitated the interpretation of the experimental results.

4.1.2. Sampling frequency, depth resolution, and missing parameters

Regular sampling for biogeochemical parameters at daily IN and OUT stations resulted in a coherent data set. Three successive CTD casts were needed at each station to collect sufficient water for the analysis of all parameters. Simultaneous values are a major advantage for budget studies. Use of a CTD rosette with 24 or 36 large (~20 l) Niskin bottles improves the accuracy of budget studies but is subject to logistic and technical constraints. Additional instruments, e.g., nets and pumps, should be deployed shortly before, between, or after corresponding CTD casts.

The CTD samples were taken at slightly variable depths. Ideally sampling for the construction of a carbon budget is done at constant depths, with at least three samples in the mixed layer, two samples below it, and additional samples directly above and below the base of the mixed layer. Time constraints, large water requirements for some parameters, and the number and size of the Niskin bottles limited the number of sampling depths. Time constraints also limited the number of floating sediment traps that could be deployed and retrieved.

The depth correction of carbon export and mesozooplankton stock has added uncertainty to the SOIREE carbon budget. Extension of the budget to 100 m depth would have decreased the accuracy of other parameters, which were sparsely sampled or had strong gradients below the mixed layer. Additional measurements of carbon export and mesozooplankton stock at the bottom of the mixed layer should be made in future studies.

Lack of DOC measurements prevents closure of the SOIREE carbon budget. Accurate DOC data are vital to a proper understanding of pelagic carbon cycling. Stock of PIC should also be determined. Extra net tows are needed for an assessment of diurnal migration of mesozooplankton.

4.1.3. Accuracy of carbon parameters

The accuracy of the largest carbon pools and fluxes strongly impacts the overall quality of the carbon budget. The RE, and hence the absolute error, are low for carbon data, which were determined by accurate measurements at high spatial and temporal resolution, e.g., $\Delta DIC_{fCO_{2w}}$ with a 4% RE (Appendix A). Carbon-14 uptake has a somewhat higher uncertainty (20%), as a result of sample incubation. The RE is sizeable for parameters that require an empirical conversion, cell counts, or visual measurements, e.g., carbon biomass (35–60%). The uncertainty is high for parameters that are based on few data points, require multiple empirical conversions, or are the residual of several terms, e.g., grazing and respiration rates, BP, and ΔDOC.

The observations in SOIREE confirm findings that export fluxes from shallow drifting traps (<150 m depth) often exceed ^{234}Th export by a factor 1.8–3.5, possibly by hydrodynamic effects leading to overtrapping (Murray et al., 1996; Charette et al., 1999). Variation in the carbon to ^{234}Th ratio may affect estimates of carbon export from the thorium deficit (Buesseler et al., 2006; Waite and Hill, 2006). However, such a potential variation would not have affected the measurement of zero carbon export in SOIREE, nor would it have significantly changed the upper limit for carbon export from ^{234}Th, which was determined by a propagation of measurement uncertainties. The variation in the sediment trap fluxes has been attributed to a sampling bias (Nodder and Waite, 2001).

Given the importance of export data for accurate carbon budgets, it might be best to use both methods in parallel until the superiority of one method is beyond doubt. The comparison of NCP with changes in POC and DOC should provide an independent assessment of carbon export in budget studies (following the approach of Sweeney et al., 2000) and would allow an investigation of the errors relating to the measurements of carbon export. Ideally NCP is determined from the *in situ* evolution of DIC or O_2 (Bakker et al., 1997, 2005; Sweeney et al., 2000; this study). New techniques, such as measurements of the oxygen to argon ratio (Kaiser et al., 2005), may provide an additional assessment of NCP.

The absence of a blank correction could have resulted in erroneously high POC_{Nis} values due to absorption of DOC to the filters (Moran et al., 1999; Gardner et al., 2003). However, this effect is likely to have been small, as the POC concentration was higher than 5 μM in SOIREE.

The POC stock from the Niskin bottles exceeded that from the pumps with a ratio of 1.2. Ratios of

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1–200 have been observed in other Southern Ocean waters with temperatures below 5 °C (Gardner et al., 2003). Gardner et al. (2003) argue that bottle data are closer to true POC than the pump data. The latter may have a methodological bias as a result of damage to bacteria and plankton cells by high-pressure differences during the filtration of large volumes of water.

Inaccurate estimates of carbon biomass, which rely on empirical conversions, visual counting of biota and measurement of the dimensions of biota, are a prime suspect for causing the excess of carbon biomass over POC, especially as the carbon content of plankton may have changed upon iron addition. An overestimation of mesozooplankton stock may have originated from the depth correction. Alternatively, the POC stock may have been underestimated. The 2-l samples for POC_{Nis} may not have fully accounted for sparsely distributed mesozooplankton (Sweeney et al., 2000). It is unlikely that large particles would have sunk below the spigots (Gardner et al., 1993), which were in the base of the Niskin bottles. Size fractionation and microscopic study of POC samples will demonstrate where discrepancies between POC and carbon biomass originate in future studies.

Phytoplankton and bacterial carbon biomass can also be estimated by flow cytometry in size-fractionated samples (Veldhuis and Kraaij, 2004). This method reduces analysis time and observer bias but also has some disadvantages. An alternative technique for determining phytoplankton carbon consists of a combination of flow cytometry and the analysis of DNA content (Veldhuis and Kraaij, 2004). All these methods require accurate conversion factors, which should be quantified repeatedly during an experiment.

Carbon-14 uptake was similar to NCP in SOIREE, but was about twice NCP along 6°W in austral spring (Bakker et al., 1997; using ^{14}C data by Jochem et al., 1995). Horizontal dispersion was not quantified along 6°W, which may have resulted in an underestimation of NCP. However, the latter effect is unlikely to fully explain the different ratios of ^{14}C uptake to NCP at 6°W and in SOIREE. These observations illustrate the difficulty in relating ^{14}C uptake rates to a specific production term.

BCD is generally estimated from BP and BGE (Carlson et al., 1999). The analysis of BP in radiotracer incorporation assays requires the conversion of incorporation rates to number of cells, factors for which vary by a factor 4 (Wiebinga et al., 1997). Evidence of an increase in BGE with the progression of a phytoplankton bloom has been found in the Ross Sea, where BGE increased from 9% and 19% in early spring to 32% and 38% in late summer (Carlson et al., 1999). Carbon budget studies will strongly benefit from the frequent assessment of BGE and bacterial carbon conversion factors.

Time consuming, indirect methods for the analysis of zooplankton respiration and grazing, requiring sample incubation and empirical conversions, were used in SOIREE. Quicker and more accurate measurement techniques should be developed, such that frequent and precise quantification of these zooplankton characteristics will be possible in future budget studies.

4.2. A comparison to carbon budgets by modelling

Carbon budgets for the 65-m deep mixed layer of the SOIREE bloom have also been made with two marine ecosystem models. The models indicate lower NCP (503 mmol m^{-2} in the SWAMCO model by Hannon et al. (2001), 664–695 mmol m^{-2} in Fasham et al. (2006), both after 13.0 d) than this study (837 ± 176 mmol m^{-2} after 12.0 d), possibly as a result of the use of ^{14}C uptake as a proxy for NPP in the models. The models predict low carbon export from the upper 65 m (21 mmol m^{-2} in SWAMCO, 38–96 mmol m^{-2} by Fasham et al.), values within the range determined by the thorium method (0 mmol m^{-2} with an upper limit of 125 mmol m^{-2}). The SWAMCO model suggests an accumulation of POC and DOC in the mixed layer of 482 mmol m^{-2} (here 407–648 mmol m^{-2}), while Fasham et al. estimate a build-up of biomass and detritus of 510–531 mmol m^{-2}. Comparison of the model results to our budget highlights how different interpretations of the same data set give different values for carbon parameters, in particular NCP. The models provide a complementary tool for studying pelagic carbon cycling.

4.3. Carbon cycling in Southern Ocean blooms

Biological activity is relatively low in large parts of the Southern Ocean, where low iron concentrations limit phytoplankton growth, as demonstrated in the SOIREE, EisenEx, SOFeX (Boyd et al., 2000; Gervais et al., 2002; Coale et al., 2004), and later iron fertilisation experiments. Seasonal algal blooms occur in frontal systems and downstream of islands

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and other shallow topography (Sullivan et al., 1993), fuelled by specific iron inputs (De Baar et al., 1995; Bucciarelli et al., 2001). The SOIREE bloom resembled natural blooms in iron-replete systems in the Southern Ocean in many respects. Importantly, the bloom was dominated by large diatoms (*Fragilariopsis kerguelensis*) (Boyd et al., 2000; Gall et al., 2001a), while spring blooms near the APF at 6°W had the diatoms *F. kerguelensis*, *Corethron inerme* and *Corethron criophilum* as dominant species (Bathmann et al., 1997). However, the SOIREE bloom did not resemble natural blooms in all respects, as will be discussed below.

Daily NCP in the SOIREE bloom was high ($70\,mmol\,m^{-2}\,d^{-1}$) and was exceeded only by that in summertime blooms in the Ross Sea (Table 4). High daily NCP in SOIREE, which corresponded to an fCO_2 reduction of $3.8\,\mu atm\,d^{-1}$, resulted from the sudden availability of iron upon fertilisation, thus enabling the algae to grow at the maximum rate for the specific environmental conditions (Bakker et al., 2001). Average daily NCP in the blooms near the APF at 6°W ($56\,mmol\,m^{-2}\,d^{-1}$) was lower than in the SOIREE bloom, partly because the regional average for 6°W included both highly productive and moderately productive areas (Figs. 6 and 7 in Bakker et al., 1997). Locally the decrease in fCO_2 was up to $2.2\,\mu atm\,d^{-1}$, or $2.7\,\mu atm\,d^{-1}$ upon correction for surface-water warming, in the main bloom areas at 6°W (after Bakker et al., 1997). Southern Ocean blooms at 170°W had lower daily NCP than SOIREE, possibly as a result of longer periods between site visits (Table 4).

Horizontal dispersion is a major term in the budgets for organic and inorganic carbon (23% of NCP) in the SOIREE bloom and is an even larger term (36% of NCP) in the budget of the EisenEx bloom (Table 4) (Bakker et al., 2005). Horizontal dispersion has not been determined for natural blooms in the open Southern Ocean but is likely to have been a major process in the dynamics of algal blooms of limited extent, notably near the APF at 6°W. Thus NCP in the core of the APF blooms, after correction for horizontal dispersion, would have been considerably higher than the average value of $56\,mmol\,m^{-2}\,d^{-1}$ for the APF region between 47° and 51°S.

Uptake of atmospheric CO_2 and vertical diffusion had little effect on DIC (<6% of NCP) in the blooms in SOIREE or EisenEx, or at 6°W (Table 4). Air-sea exchange of CO_2 was 12–28% of NCP along 170°W (Morrison et al., 2001) (Table 4).

Satellite images suggest that the SOIREE bloom persisted for at least 42 d (Abraham et al., 2000). The carbon export (0–29% of NCP) in the first 12.0 d (this study) and the inferred small carbon export from the long-lived SOIREE bloom (Abraham et al., 2000) are in the lower range of observations elsewhere. Major carbon export, equivalent to 38–73% of NCP, occurred within 21–31 d of the development of large diatom blooms near the APF at 6°W (Bakker et al., 1997; Rutgers van der Loeff et al., 1997) (Table 5). Substantial carbon export, 27–46% of annual ^{14}C uptake, was also observed between 55.0° and 68.5°S along 170°W (Nelson et al., 2002) (Table 5).

The relative increase in POC stock (41% of NCP) was smaller in SOIREE than in natural algal blooms (57% of NCP near the APF at 6°W and 72–82% of NCP in the Ross Sea) (Table 5). Estimates of the net DOC release in SOIREE (8–37% of NCP) have a large range as a result of uncertainty in carbon export. No increase in DOC stock was observed at 6°W (0% of NCP) (Kähler et al., 1997). Production of DOC constituted 0–34% of NCP with an average of 11% in different seasons in the Ross Sea (Carlson et al., 2000). The ratio of BP to ^{14}C uptake of 4–7% in SOIREE was similar to values of 4% in spring and 11% in summer in Ross Sea blooms (Ducklow et al., 2000).

5. Conclusions

The difficulty in matching carbon pools and flows in the Lagrangian SOIREE experiment highlights fundamental caveats in the sampling and analysis of carbon parameters, which hamper our understanding of the marine carbon cycle. Absence of DOC data and major uncertainty in carbon export prevent closure of the SOIREE carbon budget. Uncertainties in carbon conversion factors and infrequent measurements have made it difficult to address the role of bacteria and zooplankton in carbon cycling.

Closure of a carbon budget critically depends on the careful assessment of all relevant carbon parameters, in particular DIC, POC, DOC, PIC, carbon export and horizontal dispersion. Future budget studies should include frequent measurements of carbon conversion factors, zooplankton grazing and respiration, and bacterial growth parameters. Modelling is an additional tool for studying bloom dynamics and pelagic carbon cycling.

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Table 4
Net community production (NCP_{DIC}) and DIC changes in iron fertilisation experiments and in natural algal blooms in the Southern Ocean (Eq. (2))

Study	Period	Length (d)	NCP_{DIC} ($mmol\,m^{-2}\,d^{-1}$)	ΔDIC_{net} (%)	ΔDIC_{VDif} (%)	ΔDIC_{AirSea} (%)	ΔDIC_{HDis} (%)	Reference
Iron addition								
SOIREE, 61°S, s-APF	Feb	12.0	70 ± 14	75 ± 3	0.9 ± 0.9	2 ± 1	23 ± 17	This study
EisenEx, 48°S, F eddy	Nov	18.3	38 ± 14	57 ± 9	3 ± 3	5 ± 3	36 ± 22	Bakker et al. (2005)
Natural blooms								
47–51°S, 6°W, APF	Oct–Nov	20.8	56	85	9	6	-	Bakker et al. (1997), as corrected in Bakker, 1998
55°±1°S, 170°W, n- APF	Nov–Jan	~60	25-42	87	-	13	-	Morrison et al., 2001; Green and Sambrotto, 2006
60°±1°S, 170°W, APF	Nov–Jan	~60	28-35	88	-	12	-	Morrison et al., 2001; Green and Sambrotto, 2006
62°±1°S, 170°W, s-APF	Nov–Jan	~60	30-45	78	-	28	-	Morrison et al., 2001; Green and Sambrotto, 2006
65°±1°S, 170°W, n-SACCF	Nov–Jan	~60	23-37	77	-	23	-	Morrison et al., 2001; Green and Sambrotto, 2006
65.5°-68.5°S, Ross Gyre	Dec–Mar	100	37	-	-	-		Nelson et al. (2002)
~76.5°S, Ross Sea	Nov–Dec	32	44	-	-	-	-	Sweeney et al. (2000)
~76.5°S, Ross Sea	Dec–Feb	60	95	-	-	-	-	Sweeney et al. (2000)

Net changes in DIC (ΔDIC_{net}, ΔDIC_{fCO_2} in this study), changes in DIC by vertical diffusion (ΔDIC_{VDif}), air-sea exchange (ΔDIC_{AirSea}), and horizontal dispersion (ΔDIC_{HDis}) are indicated as a percentage of ΔDIC_{NCP}. '-' indicates the absence of a value. The abbreviations are: SACCF—Southern Antarctic Circumpolar Current Front, APF—Antarctic Polar Front, n-APF and s-APF—north and south of the APF, respectively.

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Table 5
The fate of net community production (NCP_{DIC}) in Southern Ocean blooms (Eq. (3)) for studies in Table 4

	Period	NCP_{DIC} ($mmol\,m^{-2}\,d^{-1}$)	ΔPOC (%)	ΔDOC (%)	Export (%)	ΔDIC_{HDis} (%)	Reference
Iron addition							
SOIREE, 61°S, s-APF	Feb	70±14	41±7	8–37	0–29	23±17	This study
Natural blooms							
47–51°S, 6°W, APF	Oct–Nov	56	57	0	38–73 (43)	-	Bakker et al. (1997), as corrected in Bakker, 1998
55°±1°S, 170°W, n-APF	Nov–Jan	25–42	-	-	36[a]	-	Morrison et al. (2001); Nelson et al. (2002)
60°±1°S, 170°W, APF	Nov–Jan	28–35	-	-	27[a]	-	Morrison et al. (2001); Nelson et al. (2002)
62°±1°S, 170°W, s-APF	Nov–Jan	30–45	-	-	31[a]	-	Morrison et al. (2001); Nelson et al. (2002)
65°±1°S, 170°W, n-SACCF	Nov–Jan	23–37	-	-	31[a]	-	Morrison et al. (2001); Nelson et al. (2002)
65.5°−68.5°S, 170°W, Ross Gyre	Dec–Mar	37	-	-	46[a]	-	Nelson et al. (2002)
~76.5°S, Ross Sea	Nov–Dec	44	82	6	(14)	-	Sweeney et al. (2000)
~76.5°S, Ross Sea	Dec–Feb	95	72	19	(7)	-	Sweeney et al. (2000)

Changes in particulate and dissolved organic carbon stocks (ΔPOC, ΔDOC), carbon export and horizontal dispersion of organic carbon are presented as a percentage of NCP_{DIC}. Values calculated by difference are between round (from the literature) and square brackets (this study). The abbreviations are as in Table 4.

[a]The percentage of annual carbon export along 170°W, as calculated from total annual ^{14}C uptake in Nelson et al. (2002).

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NCP converted 837 mmol m^{-2} of inorganic carbon into organic carbon within 12.0 d of the first SOIREE iron addition. Horizontal dispersion had removed 23% of newly produced organic carbon after 12.0 d, while 41% of NCP remained as POC in the mixed layer of the patch centre and 0–29% had been exported. Estimates of net DOC production range from 8–37% of NCP.

Carbon cycling in the iron-fertilised SOIREE bloom resembled that in natural algal blooms in the Southern Ocean in many respects, but not in all. Daily NCP in the SOIREE bloom exceeded that in most natural blooms in the open Southern Ocean, while the build-up of POC and carbon export as a fraction of NCP were relatively low in SOIREE in comparison to natural blooms. It is possible that the SOIREE bloom differed in other, unidentified ways from natural algal blooms. Few algal blooms and carbon cycling within them have been extensively studied in the Southern Ocean. This makes it difficult to compare the SOIREE bloom to a natural algal bloom in a similar Southern Ocean region, as bloom evolution, bloom composition and carbon cycling are likely to vary with latitude and position within the frontal zonation of the Southern Ocean.

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Appendix A

A summary of the methods and the calculation of the RE (relative error) for the carbon parameters in SOIREE (Table A.1). The parameters were obtained by direct measurement (M), by indirect estimates, or as the resultant (R) or sum (S) of other parameters. These REs are subjectively set here for steps in the estimation procedure: 30% for an empirical conversion (E), 10% for visual counting of biota (V), 10% for the visual measurement of the dimensions of biota (V), 5% for counting of biota by flow cytometry (F), 5% for measurement of dimensions by flow cytometry (F), 10% for an incubation (I), 10% for a depth conversion (D), and 20% for an extrapolation as a function of carbon biomass (X). The numbers of the equations in the text are between round brackets. References are [1] Bakker et al. (2001); [2] Bakker et al. (2005); [3]This study; [4] Wanninkhof (1992); [5] Law et al. (2003); [6] Abraham et al. (2000); [7] Charette and Buesseler (2000); [8] Trull and Armand (2001); [9] Nodder and Waite (2001); [10] Hall and Safi (2001); [11] Gall et al. (2001a); [12] Zeldis (2001); [13] Gall et al. (2001b) (Table A.1).

Table A.1

Carbon parameter	Timing	Analysis	Method	Relative error (%)	Reference
FCO_{2w}	5 min	Infrared analysis, ± 1.0 µatm, interpolation to daily maps, IC grid points from fit of fCO_{2w} to SF_6, RE = 100%/380	M	0.3	Bakker et al. (2001)
FCO_{2air}	30 min	Infrared analysis, ± 1.0 µmol mol^{-1}	M	0.3	Bakker et al. (2001)
DIC	Daily CTD	Coulometry, ± 2.7 µmol kg^{-1}, RE = 100% × 2.7/2130	M	0.1	Bakker et al. (2001)
$\Delta DIC_{fCO_{2w}}$	Daily map, Daily CTD	fCO_{2w} at IC grid points, conversion to DIC, $\Delta DIC/\Delta z = 0$ for 0−50 m, $\Delta DIC/\Delta t = 0$ below 50 m, RE from sensitivity analysis	E	4	Bakker et al. (2005); This study
ΔDIC_{AirSea}	Daily map	fCO_{2w} at IC grid points, fCO_{2air}, gas transfer velocity k_w from fit for short term, 10 min shipboard wind speed, RE = 50% (k_w) + 2% (fCO_2 maps)	E	52	Bakker et al. (2005); Wanninkhof (1992)

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Table A.1 (*continued*)

Carbon parameter	Timing	Analysis	Method	Relative error (%)	Reference
ΔDIC_{VDif}	daily map, daily CTD	DIC_{fCO2w} at IC grid points; $\Delta DIC/\Delta z$ for 50–90 m, $K_z = 0.11 \pm 0.20\,cm^2\,s^{-1}$, RE = 100% ($K_z$)	E	100	Bakker et al. (2005); Law et al. (2003)
ΔDIC_{HDis}	Daily map	$DIC_{fCO_{2w}}$ at IC grid points, $7 \pm 3\%\ d^{-1}$, RE from sensitivity analysis	E	83	Bakker et al. (2005); Abraham et al. (2000)
$\Delta DIC_{NCP\_fCO_2}$	Daily	$\Delta DIC_{fCO_{2w}}$, ΔDIC_{VDif}, ΔDIC_{AirSea}, ΔDIC_{HDis}, RE from terms	R (2)	21	Bakker et al. (2005); This study
ΔDIC_{NCP_bio}	Variable	$\Delta DIC_{^{14}C}$, ΔDIC_{BR}, ΔDIC_{ZR}, ΔDIC_{HDis_bio}, RE from terms	R (4)	80	This study
$\Delta DIC_{NCP\_^{14}C}$	Daily	$\Delta DIC_{^{14}C}$, $\Delta DIC_{HDIS\_^{14}C}$, RE from terms	R (5)	50	This study
POC_{Nis}	Daily CTD	2 l sample, >0.7 µm, NO fuming with HCl, CHN analysis, RE from excess POC_{Nis} to POC_M	M	17	This study
POC_{cp}	daily casts	Calibration of transmission to POC_{Nis}, RE as RE of POC_{Nis}	M	17	This study
POC_M	~2 d	Pump on CTD, 400 l sample, 0–100 m depth, >1.2 µm, fuming with HCl, CHN analysis, RE from deficit POC_M to POC_{Nis}	M	20	Charette and Buesseler (2000)
POC_H	~2 d	Hose pump at 10, 30, 60, 100 m depth, several hundreds of litres sample, >0.7 µm, fuming with HCl, CHN analysis, RE from deficit POC_M to POC_{Nis}	M	20	Trull and Armand (2001)
$Export_{Th}$	daily	^{234}Th depletion 0–100 m, depth conversion, RE = 30% (E) + 10% (D)	E, D	40	Charette and Buesseler (2000)
$Export_{trap}$	3 times	Floating sediment traps, 110 m depth, depth conversion, RE = 20% (M) + 10% (D)	M, D	30	Nodder and Waite (2001)
ΔDOC	daily	NCP_{DIC}, ΔPOC_{Nis}, Export, ΔDIC_{HDis}, RE from terms	R (3)	100	This study
Bacterial carbon biomass	daily CTD	Cell counts by flow cytometry, $12.4\,10^{-15}\,g\,C\,cell^{-1}$, RE = 5% (F) + 30% (E)	F, E	35	Hall and Safi (2001)
Picophytoplankton carbon biomass	daily CTD	Counts and diameter of cells by flow cytometry, cell volume from diameter, $0.22\,10^{-12}\,g\,C\,\mu m^{-3}$, RE = 2 × 5% (F) + 30% (E)	2F, E	40	Hall and Safi (2001)
Nanoflagellate carbon biomass	daily CTD	Counts and size by microscopy, volume from size and shape of 200 cells, $0.24\,10^{-12}\,g\,C\,\mu m^{-3}$, RE = 2 × 10% (V) + 30% (E)	2V, E	50	Hall and Safi (2001)
Dinoflagellate carbon biomass	daily CTD	Counts and size by microscopy, volume from size and shape, conversion to carbon biomass, RE = 2 × 10% (V) + 30% (E)	2V, E	50	Gall et al. (2001a)
Diatom carbon biomass	Daily CTD	Counts and size by microscopy, volume from size and shape, conversion to carbon biomass, RE = 2 × 10% (V) + 30% (E)	2V, E	50	Gall et al. (2001a)
Ciliate carbon biomass	Daily CTD	Counts and size by microscopy, volume from size and shape of 10–20 animals per taxon, $0.19\ 10^{-12}$ g C μm^{-3}, RE = 2 × 10% (V) + 30% (E)	2V, E	50	Hall and Safi (2001)

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Table A.1 (*continued*)

Carbon parameter	Timing	Analysis	Method	Relative error (%)	Reference
Mesozooplankton carbon biomass	Daily tow, 0–91 m	Counts and length of animals by microscopy, conversion of taxon lengths to carbon biomass, depth conversion, RE = 2 × 10% (V) + 30% (E) + 10% (D)	2V, E, D	60	Zeldis (2001)
Total carbon biomass	Daily	Sum of carbon biomass of all biota, RE from terms	S	50	This study
^{14}C uptake	Daily CTD	^{14}C incorporation, 24 hour incubation, dark correction, factor 12/14, RE = 10% (I) + 10% (R)	I, R	20	Gall et al. (2001b); This study
Bacterial production (BP)	Daily CTD	methyl-^{3}H thymidine incorporation, conversion to 2.4 10^{18} cells mol^{-1} thymidine, 20 10^{-15} g C $cell^{-1}$, RE = 10% (I) + 2 × 30% (E)	I, 2E	70	Hall and Safi (2001)
Bacterial respiration (BR)	Daily CTD	From bacterial production and bacterial growth efficiency of 0.3, RE = 10% (I) + 3 × 30% (E)	I, 3E	100	This study
Nano- + microzooplankton grazing	Day 12	Dilution technique, scale to nano- + microzooplankton biomass, RE = 10% (I) + 30% (E) + 20% (X)	I, E, X	60	Hall and Safi (2001)
Nano- + microzooplankton respiration	Day 0	Balance carbon flows after 0 d, scale to nano- + microzooplankton carbon biomass, RE from terms + 20% (X)	R, X	100	This study
Mesozooplankton grazing	~2 d	Ingestion of large copepods, factor 2 for all mesozooplankton, RE = 10% (I) + 30% (E) + 10% (factor 2)	I, E, factor 2	50	Zeldis (2001)
Mesozooplankton respiration	~2 d	Difference between grazing and egestion for large copepods, factor 2 for all mesozooplankton, RE = 10% (I) + 30% (E) + 10% (factor 2) + 30% (R)	R, I, E, factor 2	80	Zeldis (2001)

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