

## Role of algal aggregation in vertical carbon export during SOIREE and in other low biomass environments

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[1] Additions of iron to surface regions of the ocean have induced an increase in phytoplankton biomass, but do not necessarily trigger increases in carbon export from surface waters. Using new size-characterization of settling particles from the Southern Ocean Iron Release Experiment (SOIREE) and an extremely simple, mechanistic aggregation model, we show that removal of phytoplankton from surface waters via sedimentation was controlled by the formation of larger, faster sinking particles via coagulation. We demonstrate that in low biomass regions, where concentrations do not reach the critical concentration needed for massive sedimentation, carbon export is nevertheless still contingent upon the detailed mechanics of particle aggregation. There is therefore a direct dependence of export flux on the extent of aggregation during blooms, verification critical to the success of hydrodynamic models in predicting export. **Citation:** Jackson, G. A., A. M. Waite, and P. W. Boyd (2005), Role of algal aggregation in vertical carbon export during SOIREE and in other low biomass environments, *Geophys. Res. Lett.*, 32, L13607, doi:10.1029/2005GL023180.

### 1. Introduction

[2] In regions of the world's oceans where light and macronutrient concentrations could support higher algal biomass than is present [Martin *et al.*, 1994], large-scale in situ experiments have investigated the role that the micronutrient iron has on phytoplankton growth. Iron addition experiments in the Equatorial Pacific [Coale *et al.*, 1996], Southern Ocean [Boyd *et al.*, 2000; Gervais *et al.*, 2002] and subarctic Pacific [Tsuda *et al.*, 2003] increased biomass of the larger diatoms for days to weeks. Iron enrichment of the ocean is also viewed as a potential mitigation strategy to reduce atmospheric concentrations of CO<sub>2</sub> via settling of algal carbon from the surface mixed layer [Chisholm *et al.*, 2001], though no comprehensive observations of iron-enhanced downward flux have yet been reported from field experiments. Here we aim to elucidate the fundamental mechanisms by which such flux may be controlled.

[3] Downward particle flux depends, among other factors, on particle size [Aldredge and Gotschalk, 1988],

which is controlled by the balance between dispersive and coagulation processes [Jackson, 1990], mitigated by patch size [Waite and Johnson, 2003]. In the context of recent work highlighting the role of aggregation in triggering flux from mesoscale SOIREE and IronEx II [Boyd *et al.*, 2002], it is crucial to examine the assumption that aggregation is in fact the primary process driving vertical flux.

[4] Models suggest aggregation is always occurring in natural populations, but that the process rate is concentration-dependent and non-linear, happening most rapidly during intense phytoplankton blooms [Jackson, 1990]. Key analyses therefore hinge around the calculation of a critical cell concentration,  $C_{cr}$ , at which aggregation occurs rapidly enough to overwhelm continued algal division, and massive sedimentation is triggered via a sudden increase in mean particle size [Jackson, 1990]. However, in many open ocean regions, physical dispersion can keep cell concentrations significantly lower than  $C_{cr}$ , raising the possibility that aggregation would no longer control export. Can aggregation effect export at concentrations significantly below  $C_{cr}$ ?

[5] Here we test this hypothesis using the results from SOIREE, where the vertical flux was low but unusually well characterized in field measurements. We analyze hitherto unpublished particle size spectra from SOIREE, generated for the first time from sediment trap gels. We assess the ability of the simplest aggregation model to simulate correctly the observed changes in particle size distribution of the flux at these low cell concentrations. We hypothesize that even at such low cell concentrations, aggregation may represent the key process governing the rate at which algal cells sink from the euphotic zone.

[6] SOIREE occurred over 13 d in the polar Southern Ocean, south-west of New Zealand [Boyd *et al.*, 2000]. The impact of addition of dissolved iron to 50 km<sup>2</sup> of the surface mixed layer was observed for two weeks [Boyd *et al.*, 2000; Boyd and Law, 2001]. The main findings of SOIREE included increases in chlorophyll *a* and diatom concentrations. *Fragilariopsis kerguelensis* was the dominant species as carbon and biovolume. The mean settling speed of large diatoms decreased from 1.31 to 0.57 m d<sup>-1</sup> after iron addition [Waite and Nodder, 2001], and there was no mesozooplankton grazing [Zeldis, 2001]. If dispersive losses are incorporated as a decrease in net growth rate  $\mu'$ ,  $\mu' \sim 0.15 \text{ d}^{-1}$  [Boyd *et al.*, 2002; Boyd, 2002; Abraham *et al.*, 2000].

[7] Downward export flux as measured with surface-tethered free-drifting sediment traps increased by a factor of 2 [Nodder and Waite, 2001]. <sup>234</sup>Th concentrations suggested no enhanced export [Charette and Buesseler, 2000], though <sup>234</sup>Th and particulate organic carbon do not always track the same particles [Burd *et al.*, 2000; Moran *et al.*, 2003].

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## 2. Methods

[8] New data from SOIREE were used here. Free-drifting sediment traps were deployed for 2 d starting on day 11 of the experiment at locations IN and OUT of the fertilized region at depths of 110 m. Polyacrylamide gel in the bottom of the cylinders immobilized particles [Waite and Nodder, 2001], providing information on size-dependent export. Material in the traps consisted of phytoplankton cells and aggregates, primarily diatoms. Gels were scanned on video and image analysis executed at three different magnifications. Measurements included particle area,  $A$ ; a spherical-equivalent particle diameter,  $d$ , was calculated for each particle:  $d = 2(A/\pi)^{0.5}$ . Particles were sorted into size categories for which the upper spherical-equivalent volume was twice the lower. The flux size spectrum ( $\# \text{ m}^{-2} \text{ d}^{-1} \text{ cm}^{-1}$ ) was calculated as number of particles per size category per cross-sectional area sampled, per the time interval (2d), per bin size. Imaging techniques undersample particles at the lower ends of their size ranges [Jackson et al., 1997], producing an anomalous peak in size spectra. Only spectral flux values with diameters greater than the diameter of this peak were used for analysis. Also, statistics are poor when too few particles are in a size bin. For a Poisson distribution, the number of particles per bin has a counting error that goes as the square root of the mean number of particles in that bin. We thus excluded values with 5 or fewer particles in the size range from our analysis. Particle spectra calculated for the different microscope magnifications were adjusted by multiplying the spectral values obtained for the two higher magnifications by 4 and 24, providing the best fit for results.

[9] Because we wished to test the simplest iteration of the model using the raw particle size spectra, we executed no curve fitting, selected all initial model parameters (hereafter STD parameters) on the basis of field results and first principles alone, and executed only the most basic sensitivity analysis. The model was similar to that of Jackson and Lochmann [1992], with  $\mu' = 0.15 \text{ d}^{-1}$ , considering only a single dominant algal cell with properties similar to those of the dominant diatom, *F. kerguelensis*: equivalent spherical diameter was  $21 \mu\text{m}$ , and algal density was  $\Delta\rho = 0.0288 \text{ g cm}^{-3}$  inside the patch and  $\Delta\rho = 0.06938 \text{ g cm}^{-3}$  outside the patch [Waite and Nodder, 2001]. The initial concentration of algal cells was  $35 \text{ cells cm}^{-3}$ .

[10] The initial particle size spectra were calculated to be at equilibrium with the initial diatom concentration. We used a fractal coagulation kernel [Jackson, 2001], no algal divisions in aggregates greater than  $2^3 = 8$  cells in size and no disaggregation. The assumed fractal dimension in the first instance was 2.33 [Jackson et al., 1997]. The observed mixed layer depth was 65 m, and average turbulent shear rate  $\gamma = 0.1 \text{ s}^{-1}$  [Abraham et al., 2000]. The critical concentration  $C_{cr} = \mu'/(10.4\gamma \nu^3)$  was  $1.4 \times 10^3 \text{ cells cm}^{-3}$  [Jackson and Burd, 1998]. The assumed algal stickiness ( $\alpha$ ) was 1, the first-order assumption used in other similar models [Jackson, 2005]. Assumptions regarding  $\alpha$  are not straightforward, however. Field investigations have assumed lower  $\alpha$  and then modified other model parameters to enhance the fit, [Riebesell, 1991a, 1991b] where  $\alpha = 1$  would have achieved the same result. Laboratory-determined values of  $\alpha$  fit curves to aggrega-

tion data and are generally  $<1$ , but are possibly biased by pre-aggregation in culture experiments and the poor fit of the assumed stickiness model to the data [Waite et al., 1997]. On the whole we believe that  $\alpha$  is a poorly resolved quantity in both laboratory and field measurements, and that an  $\alpha$  of 1 is the simplest robust assumption for our model (though we also test  $\alpha = 0.1$ , see below). Concentrations and fluxes for waters outside the patch were calculated assuming that the particle size distribution was in steady state with the imposed particle concentration of the dominant diatom. An assessment of model fit to the data was achieved by using a combined spectrum of highest recorded values from S2, S3 or S4 within the acceptable ranges as defined above, resulting in 14 measured diameters between 0.0027 and 0.055 cm. Goodness of fit was calculated by using the data from the combined spectrum and comparing this with model results interpolated to these 14 diameters. The observed and model spectral values were  $\log_{10}$ -transformed and used to calculate a root-mean-squared (RMS) value of their differences.

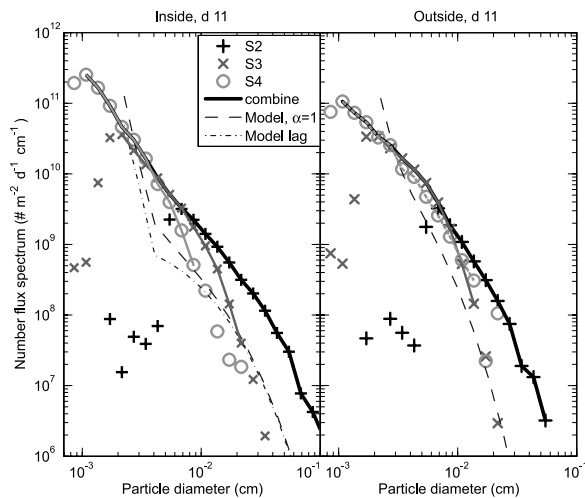
[11] We executed a limited comparison of model fits of STD parameters with other plausible parameters including higher shear ( $1 \text{ s}^{-1}$ ), lower stickiness (0.1), twice the initial number of particles (to  $70 \text{ cm}^{-3}$ ), and lower fractal dimension (2). Each model run was compared to the raw data using the method as indicated above.

## 3. Results

[12] Both the number flux spectrum and mass flux were greatest for small particles ( $<300 \mu\text{m}$ , Figure 1), consistent with single cells dominating the sinking flux. There was a measurable increase in the flux of large particles with time. Although total mass flux was only marginally higher inside the patch than outside after 11 d, fluxes of the larger particles ( $1000 \mu\text{m}$ ) were significantly greater inside ( $p < 10^{-11}$ ), confirming some aggregation in the population, although at slow rates because of the low particle abundances.

[13] The coagulation model for the iron-enriched patch shows production of larger particles at low rates for the first 10 d after fertilization. The concentrations of larger aggregates then increase and remain relatively constant, as do the total mass fluxes within the patch. The model displays a slight increase in the flux after day 10, so that the total flux has almost doubled by day 19. Given the variability in the field flux estimates, these time scales are remarkably consistent with the observed sediment trap fluxes.

[14] We tested the effect of transit time from the base of the mixed layer at 65 m to the sediment traps at 110 m by correcting the fluxes of the different size fractions for the lag associated with falling the 45 m between the two depths. For example, a particle falling at  $15 \text{ m d}^{-1}$  would take 3 d to make the transit, so that its concentration at 110 m on day 11 is that of the same size particles at day 8 at 65 m. Any particle sizes for which the lag was greater than 11 days (the experiment start) were given concentrations for day 0. Resulting concentrations were slightly lower for the smallest particles, but almost the same for particles  $100 \mu\text{m}$  and larger because their larger fall velocities reduced the time lags to insignificant levels (Figure 1).



**Figure 1.** Measured particle flux spectra at day 11 of SOIREE and flux predicted from the model using standard (STD) parameters, INside the fertilized patch (L); OUTside the fertilized patch (R). S2, S3 (4x S2), and S4 (24x S2) are different magnifications used to measure particle sizes. All data points are shown, but lines connect acceptable points (see text). Solid black line denotes the amalgamated particle flux spectrum. Solid blue line denotes the results of the model at 65 m; the dashed blue line represents the flux expected at 110 m if the flux from at 65 m is corrected for the travel times of the different size fractions to reach 110 m on the 11th day (lagged model). RMS difference between field data and STD model was 0.83 (IN) or 1.57 (OUT). See color version of this figure in the HTML.

[15] Values of log RMS difference for the raw data within the patch, with no curve fitting, was 0.89 ( $10^{0.89} = 7.75$ ) indicating that the model results were within a factor of 10 of the observations, with flux ranging over 6 orders of magnitude (Figure 2). Other parameter choices in the validation yielded values from 0.44 to 2.3. Of all the extra parameters tested, only increasing the initial particle concentration improved the fit (to RMS = 0.44, or within a factor of 3 of the data; see Figure 2).

#### 4. Discussion

[16] The small but measurable increase in particle size inside the patch documented in the flux spectra from the field data is replicated in its detail via aggregation theory in the model. The largest particles are still a small fraction of the total flux, and the formation of these particles and their contribution to export is only measurable using size-specific flux data. Despite this, the model predicts both the existence of these larger particles and their heightened contribution to export relative to single cells.

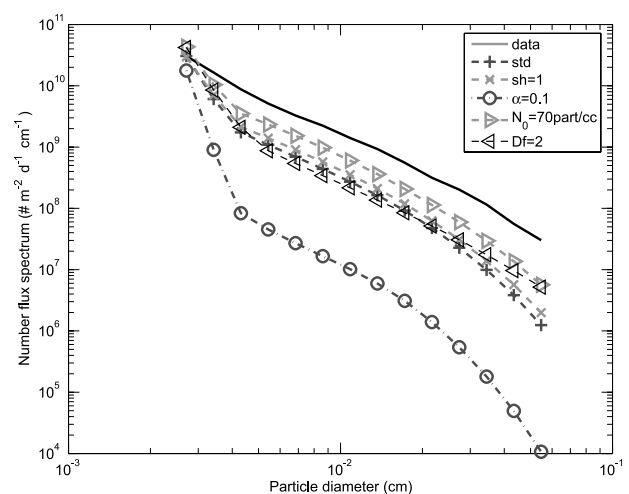
[17] The lack of a substantial ( $\sim 10X$ ) difference in downward particle flux between Fe-enriched waters and the control region after 11 d is consistent with the relatively small increase in biomass (3X increase in POC) and the decrease of sinking rate ( $\sim 3X$ ) observed in SOIREE, and the dominance of the biomass by unaggregated particles. The flux of solitary cells would have been the dominant feature driving export flux over the first 10 d. For the largest particles, however, the model indicates a clear increase in

the flux with time, suggesting that it is possible to resolve the accelerated flux in the large size fraction associated with small amounts of aggregation occurring at low mean cell concentrations, as observed in the field data [Waite and Nodder, 2001]. We note the remarkable power of a simplistic coagulation model in replicating the relatively fine detail of the particle size spectra from the field. This suggests there was no significantly enhanced export over the course of the experiment because aggregation-mediated particle sinking rate increases were confined to a small, though measurable, fraction of the available biomass.

[18] The largest differences between observed and predicted size spectra are for the smallest particles, where the simple assumption of a single dominant alga is least likely to be valid. The only improvement to the fit of the original STD model was achieved using a higher initial concentration of particles, suggesting that particles other than *F. kerguelensis* cells were in fact important components of the particle dynamics overall. This highlights the possible importance of the nanoplankton [Gall *et al.*, 2001], and other particles participating in aggregation (e.g., transparent exopolymeric particles [Passow, 2002]) not represented here.

[19] While we did not incorporate any processes changing particle concentrations between the base of the mixed layer and the sediment traps, we believe that these were small for the SOIREE site: measured zooplankton grazing rates were low near the surface and should have been lower deeper but no information is available for the deepest layers; bacterial solubilization of particles should have been slow because of the low water temperatures (2 C); lower shear rates deeper in the water column should have slowed coagulation rates.

[20] The model indicates it would take diatoms in the patch a minimum of 25 d to reach the critical concentration ( $C_{cr}$ ) where aggregation entirely overwhelms growth [Jackson, 1990; Jackson and Burd, 1998]. However, phytoplankton losses to grazing and to lateral diffusion



**Figure 2.** Effect of varying model parameters on RMS differences (in parentheses). STD standard values (0.83), higher shear =  $1 \text{ s}^{-1}$  (0.71); lower stickiness  $\alpha = 0.1$  (2.31); twice the initial particle concentrations  $N_0 = 70 \text{ cm}^{-3}$  (0.45); lower fractal index  $Df = 2$  (0.73). See color version of this figure in the HTML.

decrease the net accumulation rate [Boyd *et al.*, 2002]. Initial patch size also alters this balance, since larger patches are more likely to trigger downward flux [Waite and Johnson, 2003].

[21] Simple coagulation models have proven useful in predicting the maximum phytoplankton concentrations attained during *in situ* iron addition experiments [Boyd *et al.*, 2002; Jackson, 2005]. Overall, we confirm that even at cell concentrations below  $C_{cr}$ , aggregation is the primary process by which vertical carbon flux is delivered from primary production. In this dilute case, simple mass sedimentation at  $C_{cr}$  is replaced by a more sensitive dependence of flux on the details of coagulation dynamics. Future mesoscale perturbation experiments must include observations of particle dynamics if we are to understand the removal of particulate carbon from the surface mixed layer.

[22] Other factors would need to be incorporated to more accurately describe a given flux regime, including accurate descriptions of the physical regime, a thorough assessment of the particle stickiness, and a description of the processes between the base of the mixed layer and the depth of the sediment traps.

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