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Shifts in biogenic carbon flow from particulate to dissolved forms under high carbon dioxide and warm ocean conditions

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[1] Photosynthesis by phytoplankton in sunlit surface waters transforms inorganic carbon and nutrients into organic matter, a portion of which is subsequently transported vertically through the water column by the process known as the biological carbon pump (BCP). The BCP sustains the steep vertical gradient in total dissolved carbon, thereby contributing to net carbon sequestration. Any changes in the vertical transportation of the organic matter as a result of future climate variations will directly affect surface ocean carbon dioxide (CO_2) concentrations, and subsequently influence oceanic uptake of atmospheric CO_2 and climate. Here we present results of experiments designed to investigate the potential effects of ocean acidification and warming on the BCP. These perturbation experiments were carried out in enclosures (3,000 L volume) in a controlled mesocosm facility that mimicked future pCO_2 (~900 ppmv) and temperature (3°C higher than ambient) conditions. The elevated CO_2 and temperature treatments disproportionately enhanced the ratio of dissolved organic carbon (DOC) production to particulate organic carbon (POC) production, whereas the total organic carbon (TOC) production remained relatively constant under all conditions tested. A greater partitioning of organic carbon into the DOC pool indicated a shift in the organic carbon flow from the particulate to dissolved forms, which may affect the major pathways involved in organic carbon export and sequestration under future ocean conditions. **Citation:** Kim, J.-M., K. Lee, K. Shin, E. J. Yang, A. Engel, D. M. Karl, and H.-C. Kim (2011), Shifts in biogenic carbon flow from particulate to dissolved forms under high carbon dioxide and warm ocean conditions, *Geophys. Res. Lett.*, *38*, L08612, doi:10.1029/2011GL047346.

1. Introduction

[2] The oceanic biological carbon pump (BCP) is one of the key natural processes that regulate carbon dioxide (CO_2) levels in the atmosphere [*Archer et al.*, 2000]. The net production of organic matter by photosynthetic organisms in surface waters results in a corresponding decrease in the surface partial pressure of CO_2 (pCO_2), and in the vertical concentration gradient of dissolved inorganic carbon, which acts as a driving force for the flux of CO_2 from the atmosphere into the ocean [*Emerson et al.*, 1997; *Laws et al.*, 2000; *Lee*, 2001]. Whereas primary production includes both dissolved and particulate organic carbon (DOC and POC respectively), only POC leads to rapid and efficient carbon export to the deep ocean when it is associated with sinking biogenic inorganic particles, including those containing ballast minerals (e.g., silicate and carbonate) [*Margalef*, 1978; *Armstrong et al.*, 2001; *Klaas and Archer*, 2002]. In contrast, newly produced dissolved organic matter in surface waters is mostly recycled by bacteria back into dissolved inorganic forms. Some refractory dissolved organic matter is known to be exported to the ocean interior by vertical mixing only in oceanic regions where winter overturning ventilates the deep ocean layers [*Carlson et al.*, 1994; *Ducklow et al.*, 1995]. Finally, a small percentage of DOC may be sequestered for centuries to millennia in recalcitrant DOC molecules [*Jiao et al.*, 2010]. Therefore, the overall efficiency of the BCP is largely controlled by the export of particulate organic matter.

[3] It is currently not possible to predict how the functioning of the BCP is likely to evolve in coming centuries, because our current knowledge of how marine ecological systems will respond to emerging global environmental perturbations (i.e., ocean acidification and warming) is far from perfect. Most information to date has been derived from modeling experiments [*Intergovernmental Panel on Climate Change* (*IPCC*), 2007]. We report here the use of a controlled mesocosm facility to directly investigate the effect of pCO_2 concentration and the combined effects of pCO_2 concentration and elevated temperature on the production of organic matter, in both particulate and dissolved forms.

2. Materials and Methods

[4] Experimental settings: the manipulative experiment utilized large volume (3,000 L) mesocosm enclosures, and was carried out over 20 days in the coastal waters of Korea (34.6°N and 128.5°E) from 21 November 2008 to 11 December 2008. The experiment included acidification (~900 ppmv CO_2/ambient temperature) and greenhouse (~900 ppmv CO_2/~3°C warmer than ambient temperature) treatments to simulate likely future oceanic conditions and a contemporary ocean control (~400 ppmv CO_2/ambient temperature). The simulated CO_2 (~900 ppmv) and temperature (~3°C warmer than ambient) values chosen are close to the conditions predicted for the year 2100, based on model projections under the A2

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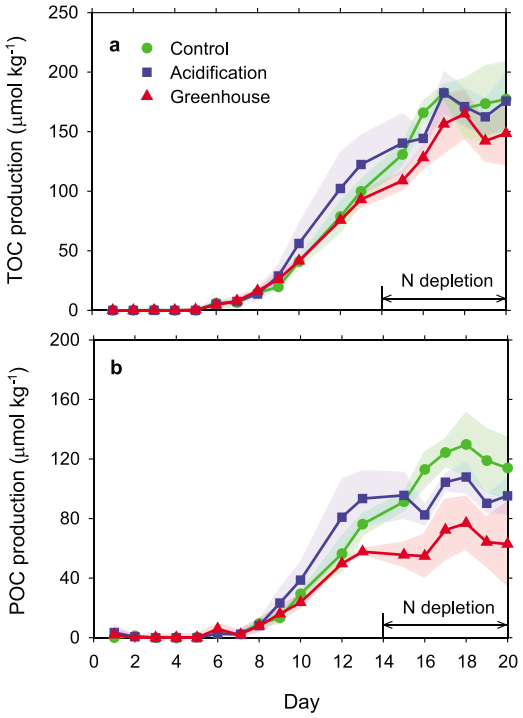

Figure 1. (a) Net production (μmol kg^{-1}) of total organic carbon (ΔTOC = TOC$_{day-n}$ − TOC$_{ref}$) and (b) particulate organic carbon (ΔPOC = POC$_{day-n}$ − POC$_{ref}$) in the control (green; ~400 ppmv CO_2/ambient temperature), acidification (blue; ~900 ppmv CO_2/ambient temperature), and greenhouse (red; ~900 ppmv CO_2/~3°C warmer than ambient temperature) enclosures during the study period. TOC$_{ref}$ and POC$_{ref}$ are defined as the mean values of total and particulate organic carbon production from day 0 to day 8, respectively. The color shading represents one standard deviation (1σ) from the mean (colored symbols and lines) of the replicate enclosures. Beginning on day 14 the dissolved inorganic nutrient concentrations were undetectable.

Scenario of the Intergovernmental Panel on Climate Change *Special Report on Emissions Scenarios* [*IPCC*, 2007].

[5] The target seawater pCO_2 levels were achieved in the enclosures by mixing CO_2-saturated seawater with ambient seawater that had been passed through a 100 μm pore size filter to remove large zooplankton (Figure S1a of the auxiliary material).[1] The 3°C elevation in seawater temperature in the greenhouse treatments was achieved within 7 h by circulating warm water (~10°C warmer than ambient) through a 30 m length of carbon tubing wrapped around the lower parts of three pillar-type seawater mixers; the elevated seawater temperature was maintained throughout the experiment (Figure S1b). When the seawater pCO_2 concentration in an enclosure reached the target value, the atmospheric pCO_2 levels were maintained at the target value by continuous addition to the enclosure atmosphere of air containing the appropriate pCO_2 concentration. On day 0, identical quantities of nutrients were added to each enclosure to initiate a phytoplankton bloom. Following addition the nutrient concentrations in each enclosure at day 1 were ~33 μM nitrate (DIN), ~2.5 μM phosphate (DIP), and ~50 μM silicate. To enhance the homogeneity of biogenic particles and solutes we gently mixed the enclosure seawater for 20 min prior to daily sampling, using bubble-mediated mixers. This mixing procedure resulted in the biogenic particles (particulate organic matter) being evenly distributed throughout the enclosure. The mesocosm facility and its performance over a range of experimental conditions have been described in detail elsewhere [*Kim et al.*, 2008].

3. Results and Discussion

[6] The key parameters in the present experiment are the production of DOC and POC and the DOC:POC production ratio. At the time of sampling, biogenic particles (measured as POC) could be either in suspension or settled on the bottom of the enclosures, and the relative proportions of suspended and settled particles should be the same in each enclosure. The settled proportion of POC was estimated as the difference between the biological utilization of dissolved inorganic carbon (DIC = $[CO_2(aq)] + [HCO_3^-] + [CO_3^{2-}]$) and the corresponding production of suspended total organic carbon (TOC = POC + DOC). The biological utilization of DIC in each enclosure was indirectly calculated from the total consumption of DIN multiplied by a DIC to DIN ratio of 6.6 [*Redfield et al.*, 1963]. In this calculation, we did not use measured DIC data because the data collected during the latter half of the experimental period were not reliable due to malfunctioning of our analytical apparatus. The POC settled at the bottom of each enclosure was estimated to account for 13 ± 6% of the total POC production, with no significant variations among the test enclosures (Figure S2). Although our bubble-mediated mixing procedure efficiently suspended most biogenic particles, some degree of particle loss was inevitable due to particle adhesion to the inner surfaces of the enclosures. The total POC production values for all enclosures were scaled up by including the loss of POC.

[7] The upward trend in the net production of TOC was stoichiometrically related to the downward trend in the concentrations of added nutrients (Figures 1a and S3). In all enclosures the concentrations of nitrate and phosphate rapidly decreased from day 0 to days 13–15 (Figure S3), while the production of TOC reached maximum levels (160 ± 20 μmol kg^{-1}) at days 17–18, then dropped slightly (Figure 1a). It is not clear what caused the slight decrease in TOC concentration without the concomitant increase in nutrient concentrations from day 17 to day 20. The maximum TOC values in the treatment (acidification and greenhouse) and control enclosures were not significantly different, which is not surprising given that the same quantities of nutrients were added to all enclosures.

[8] In contrast to the similar levels of TOC production among the treatment and control enclosures, production of DOC was highest in the greenhouse enclosures, intermediate in the acidification enclosures, and lowest in the control enclosures. During the nutrient replete period (days 6–13), DOC production was more rapid in the treatment enclosures than in the control enclosures. As a result, more DOC accumulated in the treatment enclosures, although the differences in DOC production among all enclosures were marginal. However, during the nutrient depletion period (days 14–20) the differences in DOC production among enclosures were statistically significant (ANOVA, $p < 0.05$).

[1] Auxiliary materials are available in the HTML. doi:10.1029/2011GL047346.

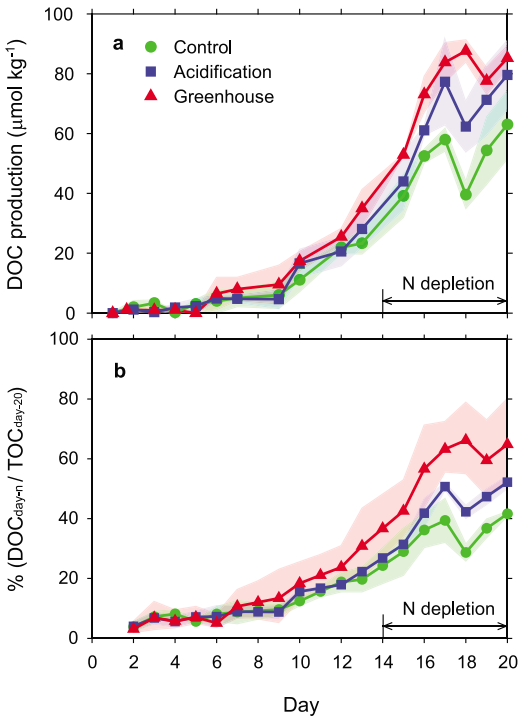

Figure 2. (a) Net production (μmol kg^{-1}) of dissolved organic carbon (ΔDOC = $DOC_{day-n} - DOC_{ref}$) and (b) percentage ratio (% = $DOC_{day-n}/TOC_{day-20} \times 100$) of daily net dissolved organic carbon production (DOC_{day-n}) to maximum total organic carbon production (measured at day 20; TOC_{day-20}) in the control (green), acidification (blue), and greenhouse (red) enclosures during the study period. DOC_{ref} is defined as the mean value of dissolved organic carbon from day 0 to day 8. The color shading represents one standard deviation (1σ) from the mean (colored symbols and lines) of the replicate enclosures.

In particular, the DOC production values in the acidification and greenhouse enclosures were 20% and 35% higher, respectively, than in the control enclosures (Figure 2a). More importantly, the DOC components of TOC production in the acidification and greenhouse enclosures were 20% to 40% higher, respectively, than in the control enclosures (Figure 2b). The disproportionate enhancement of DOC production in the treatment enclosures was concomitant with a reduction in POC production (Figure 1b), which is consistent with the TOC production remaining similar in the treatment and control enclosures (Figure 1a). Thus, the data suggest that the greater the contribution of DOC to TOC, the smaller the POC production.

[9] The observed differences in DOC production between the treatment and control enclosures likely stem from one or more of the following mechanisms: extracellular release by phytoplankton; release and excretion by grazers; grazing or viral lysis; and transformation of POC to DOC by bacteria or chemical hydrolysis [*Carlson*, 2002]. Of these DOC production pathways, DOC excretion by grazers, whereby POC is transformed to DOC by sloppy feeding, egestion and excretion is the most likely [*Strom et al.*, 1997]. In all enclosures, heterotrophic dinoflagellates (~90% of the total carbon biomass of microzooplankton) fed largely on diatoms including *Skeletonema costatum*, *Chaetoceros* spp. and *Eucampia zodiacus* [*Kim et al.*, 2010]. Among these major prey species, only *S. costatum* showed a significant positive growth response to increased pCO_2 (ANOVA, $p < 0.05$), consistent with the previous findings [*Kim et al.*, 2006]. As a result, the grazing rate during the TOC production period (days 8–16) was significantly higher in the treatment enclosures (acidification and greenhouse) than in the control enclosures (Figure S4) [*Kim et al.*, 2010], suggesting that more DOC was produced in the treatment enclosures than in the controls. This mechanism, however, cannot explain why DOC production was higher in the greenhouse enclosures than in the acidification enclosures, because the grazing rate was higher in the latter.

[10] Extracellular release by phytoplankton is another possible mechanism of DOC accumulation, and would explain the greater DOC production in the greenhouse enclosures than in the acidification enclosures, since extracellular release is directly associated with photosynthetic activity of phytoplankton [*Fogg*, 1983; *Karl et al.*, 1998]. This process is especially common during nutrient-depleted growth conditions where phytoplankton exude DOC to the environment to lower the energy costs associated with storing surplus compounds [*Fogg*, 1966; *Wood and Van Valen*, 1990]. DOC can also be passively released to the environment due to the concentration gradient across the cell membrane [*Fogg*, 1966; *Bjørnsen*, 1988]. Our observation of an upward trend in the DOC concentration in all enclosures (days 4–17) exactly coincided with the upward trend in POC production, with no lag period (Figures 2a and 1b), providing strong evidence of cellular carbon overflow. Previous studies have indicated that elevated CO_2 could enhance the extracellular release of DOC [*Engel*, 2002; *Engel et al.*, 2004; *Riebesell et al.*, 2007], because higher rates of photosynthesis result in increases in the amount of surplus carbohydrates. In addition, direct DOC excretion by some phytoplankton species may be temperature dependent under conditions favorable for photosynthesis (e.g., 15–30°C). [*Berman and Holm-Hansen*, 1974; *Verity*, 1981; *Zlotnik and Dubinsky*, 1989]. Extracellular release of photosynthetic products was enhanced in warm ocean conditions [*Morán et al.*, 2006; *Wohlers et al.*, 2009; *Engel et al.*, 2011]. This could explain the difference in DOC production under acidification and greenhouse conditions.

[11] We also evaluated the extent of bacterial lysis, another trophic interaction that may contribute to the observed differences in DOC production. A recent study showed that increased cell-specific activity of extracellular enzymes at high CO_2 levels leads to higher solubilization of POC [*Piontek et al.*, 2010]. Because the enclosures were sealed at the bottom, all POC was trapped within the enclosures. This mesocosm design did not provide an escape for sinking particles as would have happened under natural in situ conditions. Therefore, some of the trapped POC particles could be transformed into DOC by bacteria, thereby contributing to DOC production in both the treatment and control enclosures. However, this process alone is probably not adequate to explain the differences in DOC production between the treatment and control enclosures because there was no significant difference in bacterial abundance among the enclosures (treatments and control) (Figure S5).

[12] The present study indicates that, in all enclosures (regardless of treatment), the molar ratio of TOC to TON production (comparable to that of DIC to DIN drawdown)

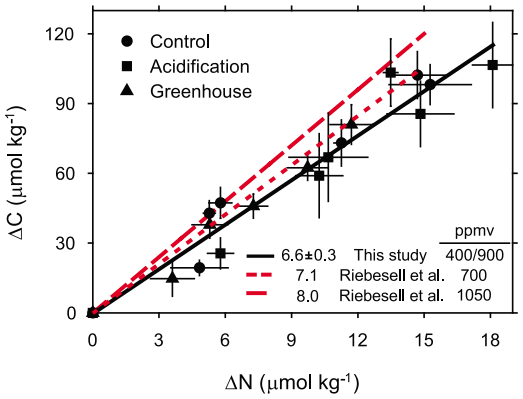

Figure 3. Ratios of TOC to TON increment in the control (filled circle), acidification (filled square), and greenhouse (filled triangle) enclosures. Error bars represents one standard deviation (1σ) from the mean of the replicate enclosures. The solid line represents the mean ratio of TOC to TON increment measured under the three different treatments used in this study. The red dotted and dashed lines indicate the ratios of DIC to DIN drawdown at 700 ppmv and 1050 ppmv, respectively [*Riebesell et al.*, 2007].

was insensitive to increasing pCO_2 concentration and was close to the Redfield ratio of ~6.6 (Figure 3). Our results do not agree with those of a previous study [*Riebesell et al.*, 2007], in which the ratio of DIC to DIN drawdown increased with increasing pCO_2 concentration and was greater than the Redfield ratio (Figure 3). The results obtained by *Riebesell et al.* [2007] indicate excess DIC consumption per unit DIN utilization in high pCO_2 oceans, in line with a strengthening of the biological pump in high pCO_2 oceans. Our study, by contrast, found no increase in the ratio of TOC to TON production with increasing pCO_2 concentration. This discrepancy may be explained by species-specific responses to increased temperature and CO_2 concentration; however, the exact cause of this apparent discrepancy is currently unknown. Additional experiments in a range of oceanic regions are needed to resolve this key issue.

4. Conclusions

[13] Our results show that elevated seawater pCO_2 concentration and temperature stimulated two main processes responsible for enhancing DOC production (release and excretion by grazers and the extracellular release by phytoplankton). An increase in the DOC:POC production ratio (with similar levels of TOC production under all test conditions) implies a shift in the organic carbon flow; that is, net POC production decreased while net DOC production increased. Although the lability of DOC produced under elevated CO_2 and temperature conditions was not determined in our experiment, the resulting excess DOC production will probably remain in the upper ocean for extended periods, during which time some fractions may be transformed into dissolved inorganic carbon via microbial degradation. Both a reduction in the vertical flux of POC and a release of CO_2 from the labile fraction of DOC may act to increase the CO_2 concentration in surface waters, thereby decreasing (or delaying) the net flux of CO_2 from the atmosphere. As a result, excess DOC production may act as a positive feedback to increase the atmospheric CO_2. However, the extent to which our results can be extrapolated to likely future oceanic conditions (i.e., elevated pCO_2 and temperature) can only be fully assessed as more experimental data become available.

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