Response to review

Dear Editor

I have gone through the reviewer's comments and fully agree that the description of the SE calculations has not been good enough. The confusion stems from a combination of a few mistakes in the 'n' number and the description of how it was calculated.

I have carefully gone through the manuscript and made appropriate corrections/changes, mainly in the materials and methods chapter and the Table and Figure legends. When going through the manuscript again I found a few minor things (e.g. spelling) that were also corrected in this version. A detailed response to each point is given below, plus the full manuscript with track changes.

Sincerely,

Kristian Spilling

All the issues and points raised by the reviewer followed with our response

I am afraid I still don't understand correctly as it seems that answers provided and the new paragraph that was added do not match. If I understand correctly, rates of change (i.e. Delta DOC) were calculated as the difference between the start (2 first days) of each period. My first question would be: how did you do for the last period?

Author response – it was done from the average of the last two days and this information has been added to the text (M&M chapter) and to the Table 3 legend.

In the added paragraph, it does not seem to be explained correctly. The same is true for the legends of Table 1-3, as it is said that: "… net community production estimated based on organic carbon pools (NCPo) are all average for Phase I in mmol C m-2 d-1 ± SE (n = 16)." How can you have n = 16 if based on a difference between 2 time points?

Author response – It was not based on the difference between two time points, as all variables were measured throughout the different phases, and the SE was calculated from the full set of data (please see also our explanation to your example of DOC below). You are right that the n = 16 is inaccurate. NCPo was calculated based on Eq. 5, for all the measured variables we have now placed the exact n number (it was 16 only for TR).

Moreover, regarding measured rates (i.e. BP), I doubt that they were measured on a daily basis (n is not 16), but I might be wrong.

Author response – You are partly right. NPP and TR were measured on a daily basis (but with a loss of some number of NPP as the incubation platform at some point disappeared),

but other parameters including BP was measured every 2-3 days. In the figure legend we now specify the exact n for all variables.

If I take the example of Delta DOC in M1 for Phase 1, that would be 16.4 mmol C/m2/d (considering a 16 day period, 7435 for the start of phase 1 - 7172 for the start of phase 2), a value of 15.5 is reported. I end up with a NCPo estimate of 18.2 mmol C/m2/d.

Author response – Phase I, M1: the total period counted was from t0 to t16, which is 17 days when including day 0 as the first day, ending up with 15.5 instead of 16.4. This affects also NCPo.

More importantly, I don't understand that, if indeed you propagated errors the way you explained (i.e. square root of the sum of variance), how you end up with a propagated error that is lower than one of the terms. For instance, still for M1 in Phase 1, delta DOC should have an error of SQRT(87^2+38^2) = ca. 95. Doing the same for delta TPC (SE ~40), you would end up with a propagated error on NCPo of SQRT(95^2+40^2+0.1^2) = 103, far from the value of 33 that is reported.

Author response –The confusion most likely stems from an error in the n value set for the Delta (Δ) variables, and that the explanation of SE calculations was not elaborate enough. In your given example the n value is 8 and not 2. The SE for the Δ variables was not calculated from SE of the pools, e.g. DOC_{pool} as in your example. Rather, the change was calculated between each measuring point and the SE for Δ variables calculated for the full range. We have corrected the n value in the figure legend, and elaborated the description in the materials and methods chapter.

I might have misunderstood, however if this is the case, first I apologize, but also I would suggest the authors to better clarify their methodology.

Author response - We fully agree and have hopefully managed to make the description of the SE calculations clear.

Very minor corrections:

L18-21: Affiliations have been swapped
Author response – Corrected

L50: Please remove "fixed"
Author response – removed

L180: A parenthesis is missing in the equation
Author response – corrected

L650: since you calculate GPP from your budgets, you should change to: (i.e. NCP + TR)
Author response - L650 is in the reference section, I looked through all places where GPP appeared, but did not find any apparent place where '(i.e. NCP + TR)' should be inserted.

Effects of ocean acidification on pelagic carbon fluxes in a mesocosm experiment

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Running title: Modified pelagic carbon fluxes

Key words: Carbon fluxes, carbon budget, gross primary production, respiration, bacterial production, sinking carbon flux, CO_2 exchange with atmosphere

Abstract

About a quarter of anthropogenic CO_2 emissions are currently taken up by the oceans decreasing seawater pH. We performed a mesocosm experiment in the Baltic Sea in order to investigate the consequences of increasing CO_2 levels on pelagic carbon fluxes. A gradient of different CO_2 scenarios, ranging from ambient (~370 µatm) to high (~1200 µatm), were set up in mesocosm bags (~55 m^3). We determined standing stocks and temporal changes of total particulate carbon (TPC), dissolved organic carbon (DOC), dissolved inorganic carbon (DIC) and particulate organic carbon (POC) of specific plankton groups. We also measured carbon flux via CO_2 exchange with the atmosphere and sedimentation (export); and biological rate measurements of primary production, bacterial production and total respiration. The experiment lasted for 44 days and was divided into three different phases (I: *t0-t16*; II: *t17-t30*; III: *t31-t43*). Pools of TPC, DOC and DIC were approximately 420, 7200 and 25200 mmol C m^{-2} at the start of the experiment, and the initial CO_2 additions increased the DIC pool by ~7% in the highest CO_2 treatment. Overall, there was a decrease in TPC and increase of DOC over the course of the experiment. The decrease in TPC was lower, and increase in DOC higher, in treatments with added CO_2. During Phase I the estimated gross primary production (GPP) was ~100 mmol C ~~fixed~~ m^{-2} d^{-1}; from which 75-95% were respired, ~1% ended up in the TPC (including export) and 5-25% added to the DOC pool. During Phase II, the respiration loss increased to ~100% of GPP at the ambient CO_2 concentration, whereas respiration was lower (85-95% of GPP) in the highest CO_2 treatment. Bacterial production was ~30% lower, on average, at the highest CO_2 concentration compared with the controls during Phases II and III. This resulted in a higher accumulation DOC standing stock and lower reduction in TPC in the elevated CO_2 treatments at the end of Phase II extending throughout Phase III. The "extra" organic carbon at high CO_2 remained fixed in an increasing biomass of small-sized plankton and in the DOC pool, and did not transfer into large, sinking aggregates. Our results revealed a clear effect of increasing CO_2 on the carbon budget and mineralization, in particular under nutrient limited conditions. Lower carbon loss processes (respiration and bacterial remineralization) at elevated CO_2 levels resulted in higher TPC and DOC pools compared with the ambient CO_2 concentration. These results highlight the importance to address not only net changes in carbon standing stocks, but also carbon fluxes and budgets to better disentangle the effects of ocean acidification.

1 Introduction

Combustion of fossil fuels and change in land use, have caused increasing atmospheric concentrations of carbon dioxide (CO_2). Ca. 25% of the anthropogenic CO_2 is absorbed by the oceans, thereby decreasing surface water pH, a process termed ocean acidification (Le Quéré et al., 2009). Ocean acidification and its alterations of aquatic ecosystems have received considerable attention during the past decade, but there are many open questions, in particular related to consequences for planktonic mediated carbon fluxes.

Some studies on ocean acidification have reported increased carbon fixation (Egge et al., 2009; Engel et al., 2013), bacterial production (Grossart et al., 2006) and bacterial degradation of polysaccharides (Piontek et al., 2010) at enhanced CO_2 levels, with potential consequences for carbon fluxes within pelagic ecosystems and export to the deep ocean, i.e. the biological carbon pump. Increasing carbon fixation in a high CO_2 environment can translate into an enhanced sequestration of carbon (Riebesell et al., 2007), but this depends on numerous environmental factors including phytoplankton community composition, aggregate formation and nutrient availability. For example, if the community shifts towards smaller cell sizes and/or enhanced cycling of organic matter carbon, export from the upper water layers may decrease (Czerny et al., 2013a).

The effect of ocean acidification has mostly been studied in marine ecosystems under high phytoplankton biomass. Brackish water has lower buffering capacity than ocean water and the pH fluctuates more. The limited number of studies of ocean acidification in brackish water and indications that ocean acidification effects are greatest under nutrient limitation (De Kluijver et al., 2010), motivated this mesocosm study in the Baltic Sea during low nutrient, summer months.

The Baltic Sea is functionally much like a large estuary, with a salinity gradient ranging from approximately 20 in the South-West to <3 in the Northernmost Bothnian Bay. It is an almost landlocked body of water with a large population in its vicinity (~80 million). Human activities (e.g. agriculture, shipping and fishing) cause a number of environmental problems such as eutrophication and pollution. As a coastal sea projected to change rapidly due to interaction of direct and indirect anthropogenic pressures, the Baltic Sea can be seen as a model ecosystem to study global change scenarios (Niiranen et al., 2013).

Most primary data from this experiment are published in several papers of this Special Issue (Riebesell et al., 2015). The aim of the present paper is to provide an overarching synthesis of

all information related to carbon standing stocks and fluxes. This enabled us to calculate carbon budgets in relation to different CO_2 levels.

2 Materials and methods

2.1. Experimental set-up

Six Kiel Off-Shore Mesocosms for future Ocean Simulations (KOSMOS; with a volume of ca. 55 m^3) were moored at Storfjärden, on the south west coast of Finland (59° 51.5' N; 23° 15.5' E) on 12 June 2012 (nine KOSMOS units were originally deployed but three were lost due to leaks). A more detailed description of the set-up can be found in Paul et al. (2015). The mesocosms extended from the surface down to 19 m depth and had a conical bottom end, which enabled quantitative collection of the settling material. Different CO_2 levels in the bags were achieved by adding filtered (50 µm), CO_2-saturated seawater. The CO_2 enriched water was evenly distributed over the upper 17 m of the water columns and added in 4 consecutive time steps (*t0 – t3*). Two controls and four treatments were used, and for the controls, filtered seawater (without additional CO_2 enrichment) was added. The CO_2 fugacity gradient after all additions ranged from ambient (average throughout the experiment: ~370 µatm fCO_2) in the two control mesocosms (M1 and M5), up to ~1200 µatm fCO_2 in the highest treatment (M8). We used the average fCO_2 throughout this experiment (from *t1 – t43*) to denote the different treatments: 365 (M1), 368 (M5), 497 (M7), 821 (M6), 1007 (M3) and 1231 (M8) µatm fCO_2. On *t15*, additional CO_2–saturated seawater was added to the upper 7 m in the same manner as the initial enrichment, to counteract outgassing of CO_2.

We sampled the mesocosm every morning, but some variables were determined only every second day. Depth-integrated water samples (0 – 17 m) were taken by using integrating water samplers (IWS, HYDRO-BIOS, Kiel). The water was collected into plastic carboys (10 L) and taken to the laboratory for sub-sampling and subsequent determination of carbon stocks.

2.2. Primary variables

For more detailed descriptions of the primary variables and the different methods used during this CO_2 mesocosm campaign, we refer to other papers in this joint volume: i.e. total particulate carbon (TPC), dissolved organic carbon (DOC), and dissolved inorganic carbon (DIC) are described by Paul et al. (2015); micro and nanophytoplankton enumeration by Bermúdez et al. (2016); picophytoplankton, heterotrophic prokaryotes and viruses by Crawfurd et al. (2016); zooplankton community by Lischka et al. (2015); primary production and respiration by Spilling et al. (2016); bacterial production (BP) by Hornick et al. (2016); and sedimentation by Boxhammer et al. (2016); and Paul et al. (2015).

Briefly, samples for TPC (500 mL) were GF/F filtered and determined using an elemental analyszer (EuroAE). DOC was measured using the high temperature combustion method (Shimadzu TOC –VCPN) following Badr et al. (2003). DIC was determined by infrared absorption (LI-COR LI-7000 on an AIRICA system). The DIC concentrations were converted from µmol kg^{-1} to µmol L^{-1} using the average seawater density of 1.0038 kg L^{-1} throughout the experiment. Settling particles were quantitatively collected every other day from sediment traps at the bottom of the mesocosm units and the TPC determined from the processed samples (Boxhammer et al., 2016) as described above.

Mesozooplankton was collected by net hauls (100 µm mesh size), fixed (ethanol) and counted in a stereomicroscope. Zooplankton carbon biomass (CB) was calculated using the displacement volume (DV) and the equation of Wiebe (1988): (log DV + 1.429)/0.82 = log CB. Micro and nanoplankton (zoo- and phytoplankton) CB was determined from microscopic counts of fixed (acidic Lugol's iodine solution) samples, and the cellular bio-volumes were determined according to Olenina et al. (2006) and converted to POC by the equations provided by Menden-Deuer and Lessard (2000).

Picophytoplankton were counted using flow cytometry and converted to CB by size fractionation (Veldhuis and Kraay, 2004) and cellular carbon conversion factors (0.2 pg C $µm^{-3}$ (Waterbury et al., 1986). Prokaryotes and viruses were determined according to Marie et al. (1999) and Brussaard (2004), respectively. All heterotrophic prokaryotes, hereafter termed bacteria, and viruses were converted to CB assuming 12.5 fg C $cell^{-1}$ (Heinänen and Kuparinen, 1991) and 0.055 fg C $virus^{-1}$ (Steward et al., 2007), respectively.

The respiration rate was calculated from the difference between the O_2 concentration (measured with a Fibox 3, PreSens) before and after a 48 h incubation period in a dark, climate controlled room set to the average temperature observed in the mesocosms.

Bacterial protein production (BPP) was determined by ^{14}C-leucine (^{14}C-Leu) incorporation (Simon and Azam, 1989) according to Grossart et al. (2006). The amount of incorporated ^{14}C-Leu was converted into BPP by using an intracellular isotope dilution factor of 2. A conversion factor of 0.86 was used to convert the produced protein into carbon (Simon and Azam, 1989).

Net primary production (NPP) was measured using radio-labeled $NaH^{14}CO_3$ (Steeman-Nielsen, 1952). Samples were incubated for 24 h in duplicate, 8 ml vials moored on small incubation platforms at 2, 4, 6, 8 and 10 m depth next to the mesocosms. The areal primary production was calculated based on a simple linear model of the production measurements from the different depths (Spilling et al., 2016).

2.3. Gas exchange

In order to calculate the CO_2 gas exchange with the atmosphere (CO_{2flux}), we used N_2O as tracer gas, and this was added to mesocosm M5 and M8 (control and high CO_2 treatment) according to Czerny et al. (2013b). The N_2O concentration was determined every second day using gas chromatography. Using the N_2O measurements, the fluxes across the water surface (F_{N2O}) was calculated according to:

$$F_{N2O} = I_{t1} - I_{t2} / (A * \Delta t) \quad (1)$$

where I_{t1} and I_{t2} is the bulk N_2O concentration at time: t_1 and t_2; A is the surface area and Δt is the time difference between t_1 and t_2.

The flux velocity was then calculated by:

$$K_{N2O} = F_{N2O} / (C_{N2Ow} - (C_{N2O\,aw})) \quad (2)$$

where C_{N2Ow} is the bulk N_2O concentration in the water at a given time point, and C_{N2Oaw} is the equilibrium concentration for N_2O (Weiss and Price, 1980).

The flux velocity for CO_2 was calculated from the flux velocity of N_2O according to:

$$k_{CO2} = k_{N2O} / (Sc_{CO2}/Sc_{N2O})^{0.5} \quad (3)$$

where Sc_{CO2} and Sc_{N2O} are the Schmidt numbers for CO_2 and N_2O, respectively. The CO_2flux across the water surface was calculated according to:

$$F_{CO2} = k_{CO2} (C_{CO2w} - C_{CO2aw}) \quad (4)$$

where C_{CO2w} is the water concentration of CO_2 and C_{CO2aw} is the equilibrium concentration of CO_2. CO_2 is preferentially taken up by phytoplankton at the surface, where also the atmospheric exchange takes place. For this reason, we used the calculated CO_2 concentration (based on the integrated CO_2 concentration and pH in the surface) from the upper 5 m as the input for equation 5.

In contrast to N_2O, the CO_2 flux can be chemically enhanced by hydration reactions of CO_2 with hydroxide ions and water molecules in the boundary layer (Wanninkhof and Knox, 1996). Using the method outlined in Czerny et al. (2013b) we found an enhancement of up to 12% on warm days and this was included into our flux calculations.

2.4. Data treatment

The primary data generated in this study comprise of carbon standing stock measurements of TPC, DOC, DIC, as well as carbon estimates of meso- and microzooplankton, micro-, nano- and picophytoplankton, bacteria and viruses. Flux measurements of atmospheric CO_2 exchange and sedimentation of TPC, as well as the biological rates of net primary production (NPP_{14C}), bacterial production (BP) and total respiration (TR) enabled us to make carbon budget.

Based on the primary variables (Chl *a* and temperature), the experiment where divided into three distinct phases: Phase I: *t0-t16*; Phase II: *t17-t30* and Phase III: *t31-t43*, where e.g. Chlorophyll *a* (Chl *a*) concentration was relatively high during Phase I, decreased during Phase II and remained low during Phase III (Paul et al. 2015). Measurements of pools and rates were average for the two first sampling points of each experimental phase (n = 2) and where normalized to m^2 knowing the total depth (17 m, excluding the sedimentation funnel) of the mesocosms. For Phase III we used the average of the last two measurements as the end point (n = 2).

For fluxes and biological rates we used the average for the whole periods normalized to days (day^{-1}),. The same was done for rates of change (ΔTPC, ΔDOC and ΔDIC), which accounted for the ~~were the~~ difference between the start and end of each phase for all carbon pools (TPC_{pool}, DOC_{pool}, DIC_{pool}). All error estimates were calculated as standard error (SE), and this was calculated using all measurements within each phase (e.g. calculating the ΔTPC SE

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using the difference between each TPC measurement). The three different phases of the experiments were of different length and each variable had a slightly different sampling regime (every 1-3 days, and some measurements missing due to technical problems). The exact sample number (n) for each SE is presented in the Table legends 1-3. ~~with n = 16, n = 14 and n = 13 for Phases I – III respectively.~~ The SE for estimated rates were calculated from the square root of the sum of variance for all the variables (Eq 5-10 below) The primary papers mentioned above (section 2.2.) present detailed statistical analyses and we only refer to those here.

NPP was measured directly and we additionally estimated the net community production (NCP). This was done in two different ways from the organic (NCP_o), dissolved plus particulate and inorganic (NCP_i) fractions of carbon. NCP_o was calculated from changes in the organic fraction plus the exported TPC (EXP_{TPC}) according to:

$$NCP_o = EXP_{TPC} + \Delta TPC + \Delta DOC \quad (5)$$

Direct measurements using ^{14}C isotope incubations should in principal provide a higher value than summing up the difference in overall carbon balance (our NCP_o), as the latter would incorporate total respiration and not only autotrophic respiration. NCP_i was calculated through changes in the dissolved inorganic carbon pool, corrected for CO_2 gas exchange with the atmosphere (CO2flux) according to:

$$NCP_i = CO_{2flux} - \Delta DIC \quad (6)$$

In order to close the budget we estimated gross primary production (GPP) and DOC production (DOC_{prod}). GPP is defined as the photosynthetically fixed carbon without any loss processes (i.e. NPP + autotrophic respiration). GPP can be estimated based on changes in organic (GPP_o) or inorganic (GPP_i) carbon pools, and we used these two different approaches providing a GPP range:

$$GPP_o = NCP_o + TR \quad (7)$$

$$GPP_i = TR + CO_{2flux} - \Delta DIC \quad (8)$$

During Phase III, TR was not measured and we estimated TR based on the ratios ~~bwteen~~between NCP_o and BP to TR during Phase II. The minimum production of DOC (DOC_{minp}) in the system was calculated assuming bacterial carbon uptake was taken from the DOC pool according to:

250 $DOC_{minp} = \Delta DOC + BP$ (9)

251 However, this could underestimate DOC_{prod} as a fraction of bacterial DOC uptake is respired.
252 Without direct measurement of (heterotrophic prokaryote) bacterial respiration, (BR), we
253 estimated BR from TR. The share of active bacteria contributing to bacterial production is
254 typically in the range of 10-30% of the total bacterial community (Lignell et al., 2013). We
255 used the fraction of bacterial biomass (BB) of total biomass (TB) as the maximum limit of
256 BR (BR $\leq$ BB/TB), and hence calculated max DOC production (DOC_{maxp}) according to:

257 $DOC_{maxp} = \Delta DOC + BP + (BB * TR / TB)$ (10)

258 We assumed that carbon synthesized by bacteria added to the TPC pool.

259 There are a number of uncertainties in these calculations, but this budgeting exercise provides
260 an order-of-magnitude estimate of the flow of carbon within the system and enables
261 comparison between the treatments. The average of the two controls (M1 and M5) and two
262 highest CO_2 treatments (M3 and M8) were used to illustrate CO_2 effects.

263

264 **3. Results and discussion**

265 **3.1 Change in plankton community, from large to small forms over time**

266 The overall size structure of the plankton community decreased over the course of the
267 experiment. Fig 1 illustrates the carbon content in different plankton groups in the control
268 mesocosms. During Phase I, the phytoplankton abundances increased at first in all treatments
269 before starting to decrease at the end of Phase I (Paul et al., 2015). At the start of Phase II
270 (t17), the phytoplankton biomass was higher than at the start of the experiment (~130 mmol
271 C m^{-2} in the controls) but decreased throughout Phase II and III. The fraction of
272 picophytoplankton increased in all treatments, but some groups of picophytoplankton
273 increased more in the high CO_2 treatments (Crawfurd et al., 2016).

274 Nitrogen was the limiting nutrient throughout the entire experiment (Paul et al., 2015), and
275 primary producers are generally N-limited in the main sub-basins of the Baltic Sea
276 (Tamminen and Andersen, 2007). The surface to volume ratio increases with decreasing cell
277 size, and consequently small cells have higher nutrient affinity, and are better competitors for
278 scarce nutrient sources than large cells (Reynolds, 2006). The prevailing N-limitation was
279 likely the reason for the decreasing size structure of the phytoplankton community.

Micro and mesozooplankton standing stock was approximately half of the phytoplankton biomass initially, but decreased rapidly in the control treatments during Phase I (Fig 1). In the CO_2 enriched treatments the zooplankton biomass also decreased but not to the same extent as in the control treatments (Spilling et al., 2016). Overall, smaller species benefitted from the extra CO_2 addition, but there was no significant negative effect of high CO_2 on the mesozooplankton community (Lischka et al., 2015).

Bacterial biomass was the main fraction of the plankton carbon throughout the experiment. The bacterial numbers largely followed the phytoplankton biomass with an initial increase then decrease during Phase I; increase during Phase II and slight decrease during Phase III (Crawfurd et al., 2016). The bacterial community was controlled by mineral nutrient limitation, bacterial grazing and viral lysis (Crawfurd et al., 2016), and bacterial growth is typically limited by N or a combination of N and C in the study area (Lignell et al., 2008; Lignell et al., 2013).

The bacterial carbon pool was higher than the measured TPC. Part of the bacteria must have passed the GFF filters (0.7 µm), and assuming pico- to mesoplankton was part of the TPC, >50% of the bacterial carbon was not contributing to the measured TPC. The conversion factor from cells to carbon is positively correlated to cell size, and there is consequently uncertainty related to the absolute carbon content of the bacterial pool (we used a constant conversion factor). However, bacteria is known to be the dominating carbon share in the Baltic Sea during the N-limited summer months (Lignell et al., 2013), and its relative dominance is in line with this.

Although there are some uncertainty in the carbon estimate (Jover et al. 2014), virus make up (due to their numerical dominance) a significant fraction of the pelagic carbon pool. Of the different plankton fractions the virioplankton have been the least studied, but their role in the pelagic ecosystem is ecologically important (Suttle, 2007; Brussaard et al., 2008; Mojica et al., 2016). Viral lysis rates were equivalent to the grazing rates for phytoplankton and for bacteria in the current study (Crawfurd et al., 2015). As mortality agents, viruses are key drivers of the regenerative microbial food web (Suttle, 2007; Brussaard et al., 2008). Overall, the structure of the plankton community reflected the nutrient status of the system. The increasing N-limitation favoring development of smaller cells, and increasing dependence of the primary producers on regenerated nutrients.

3.2. The DIC pool and atmospheric exchange of CO_2

The DIC pool was the largest carbon pool: 3-4 fold higher than the DOC pool and roughly 60-fold higher than the TPC pool (Tables 1-3). After the addition of CO_2, the DIC pool was ~7% higher in the highest CO_2 treatment compared to the control mesocosms (Table 1). The gas exchange with the atmosphere was the most apparent flux affected by CO_2 addition (Tables 1-3). Seawater in the mesocosms with added CO_2 were supersaturated, hence CO_2 outgassed throughout the experiment. The control mesocosms were initially undersaturated, hence ingassing occurred during Phases I and II (Fig 2). In the first part of Phase III, the control mesocosms reached equilibrium with the atmospheric fCO_2 (Fig. 2). The gas exchange had direct effects on the DIC concentration in the mesocosms (Fig. 3). From the measured gas exchange and change in DIC it is possible to calculate the biologically mediated carbon flux. In the mesocosms with ambient CO_2 concentration, the flux measurements indicated net heterotrophy throughout the experiment. The opposite pattern, net autotrophy, was indicated in the two mesocosms with the highest CO_2 addition (Fig 3; see also section 3.7.).

3.3. The DOC pool, DOC production and remineralization

The DOC pool increased throughout the experiment in all mesocosm bags, but more in the treatments with elevated CO_2 concentration. The initial DOC standing stock in all treatments was approximately 7200 mmol C m^{-2}. At the end of the experiment, the DOC pool was ~2% higher in the two highest CO_2 treatments compared to the controls (Fig. 4), and there is statistical support for this difference between CO_2 treatments (Phase III, $p = 0.05$) (Paul et al., 2015). Interestingly, the data does not point to a substantially higher release of DOC at high CO_2 (Figs 4 and 5). The bacterial production was notably lower during Phases II and III in the high CO_2 treatments (Hornick et al., 2016), and of similar magnitude as the rate of change in DOC pool (Table 2 and 3), indicating reduced bacterial uptake and remineralization of DOC. The combined results suggest that the increase in the DOC pool at high CO_2 was related to reduced DOC loss (uptake by bacteria), rather than increased release of DOC by the plankton community, at elevated CO_2 concentration.

The Baltic Sea is affected by large inflow of freshwater containing high concentrations of refractory DOC such as humic substances, and the concentration in Gulf of Finland is typically 400-500 µmol C L^{-1} (Hoikkala et al., 2015). The large pool of DOC and turn over

times of ~200 days (Tables 1-3) is most likely a reflection of the relatively low fraction of labile DOC, but bacterial limitation of mineral nutrients can also increase turn over times (Thingstad et al., 1997).

The DOC pool has been demonstrated to aggregate into transparent exopolymeric particles (TEP) under certain circumstances, which can increase sedimentation at high CO_2 levels (Riebesell et al., 2007). We did not have any direct measurements of TEP, but any CO_2 effect on its formation is highly dependent on the plankton community and its physiological status (MacGilchrist et al., 2014). No observed effect of CO_2 treatment on carbon export suggests that we did not have a community where the TEP production was any different between the treatments used.

3.4. The TPC pool and export of carbon

There was a positive effect of elevated CO_2 on TPC relative to the controls. At the start of the experiment, the measured TPC concentration in the enclosed water columns was 400-500 mmol C m^{-2} (Table 1). The TPC pool decreased over time but less in the high CO_2 treatment and at the end of the experiment, the standing stock of TPC was ~6% higher (Phase III, $p = 0.01$; Paul et al. (2015) in the high CO_2 treatment (Fig. 4).

The export of TPC was not dependent on the CO_2 concentration but varied temporally. The largest flux of TPC out of the mesocosms occurred during Phase I with ~6 mmol C m^{-2} d^{-1}. It decreased to ~3 mmol C m^{-2} d^{-1} during Phase II and was ~2 mmol C m^{-2} d^{-1} during Phase III (Table 1-3). The exported carbon as percent of average TPC standing stock similarly decreased from ~1.3% during Phase I to 0.3-0.5% during Phase III. The initial increase in the autotrophic biomass was the likely reason for relatively more of the carbon settling in the mesocosms in the beginning of the experiment whereas the decreasing carbon export was most likely caused by the shift towards a plankton community depending on recycled nitrogen. This reduced the overall suspended TPC and also the average plankton size in the community.

3.5. Biological rates: respiration

Total respiration (TR) was always lower in the CO_2 enriched treatments (Tables 1-3). The average TR was 83 mmol C m^{-2} d^{-1} during Phase I, and initially without any detectable

375 treatment effect. The respiration rate started to be lower in the high CO_2 treatments,
376 compared with the controls, in the beginning of Phase II. At the end of Phase II there was a
377 significant difference ($p = 0.02$; Spilling et al., 2016) between the treatments (Table 2), and
378 40% lower respiration rate in the highest CO_2 treatment compared with the controls (Spilling
379 et al., 2016).

380 Cytosol pH is close to neutral in most organisms, and reduced energetic cost for internal pH
381 regulation (e.g. transport of H^+) and at lower external pH levels could be one factor reducing
382 respiration (Smith and Raven, 1979). Hopkinson et al. (2010) found indirect evidence for
383 decreased respiration and also proposed that increased CO_2 concentration (i.e. decreased pH)
384 reduced metabolic cost of remaining intracellular homeostasis. Mitochondrial respiration in
385 plant foliage decreases in high CO_2 environments, possibly affected by respiratory enzymes
386 or other metabolic processes (Amthor, 1991; Puhe and Ulrich, 2012). Most inorganic carbon
387 in water is in the form of bicarbonate (HCO_3^-) at relevant pH, and many aquatic autotrophs
388 have developed carbon concentrating mechanisms (CCMs) (e.g. Singh et al., 2014) that could
389 reduce the cost of growth (Raven, 1991). There are some studies that have pointed to savings
390 of metabolic energy due to down-regulation of carbon concentrating mechanisms (Hopkinson
391 et al., 2010) or overall photosynthetic apparatus (Sobrino et al., 2014) in phytoplankton at
392 high CO_2 concentrations. Yet, other studies of the total plankton community have pointed at
393 no effect or increased respiration at elevated CO_2 concentration (Li and Gao, 2012; Tanaka et
394 al., 2013), and the metabolic changes behind reduced respiration, remains an open question.
395 Membrane transport of H+ is sensitive to changes in external pH, but the physiological
396 impacts of increasing H+ needs further study to better address effects of ocean acidification
397 (Taylor et al., 2012). An important aspect is also to consider the microenvironment
398 surrounding plankton; exchange of nutrients and gases takes place through the boundary
399 layer, which might have very different pH properties than bulk water measurements (Flynn et
400 al., 2012).

401

402 **3.6. Biological rates: bacterial production**

403 Bacterial production (BP) became lower in the high CO_2 treatment in the latter part of the
404 experiment. During Phase I, BP ranged from 27 to 46 mmol C m^{-2} d^{-1} (Table 1). The
405 difference in BP between treatments became apparent in Phases II and III of the experiment.
406 The average BP was 18% and 24% higher in the controls compared to the highest CO_2

407 treatments during Phases II and III, respectively (Tables 2 and 3). Statistical support ($p \leq 0.01$)
408 for a treatment effect during parts of the experiment is presented in Hornick et al. (2016).

409 The lower bacterial production accounted for ~40% of the reduced respiration during Phase
410 II, and the reduced respiration described above could at least partly be explained by the lower
411 bacterial activity. This raises an interesting question: what was the mechanism behind the
412 reduced bacterial production/respiration in the high CO_2 treatment? There are examples of
413 decreased bacterial production (Motegi et al 2013) and respiration (Teira et al., 2012) at
414 elevated CO_2 concentration. However, most previous studies have reported no change
415 (Allgaier et al., 2008) or a higher bacterial production at elevated CO_2 concentration
416 (Grossart et al., 2006; Piontek et al., 2010; Endres et al., 2014). The latter was also supported
417 by the recent study of Bunse et al. (2016), describing up-regulation of bacterial genes related
418 to respiration, membrane transport and protein metabolism at elevated CO_2 concentration;
419 albeit, this effect was not evident when inorganic nutrients had been added (high Chl *a*
420 treatment).

421 In this study, the reason for the lower bacterial activity in the high CO_2 treatments could be
422 due to either limitation and/or inhibition of bacterial growth or driven by difference in loss
423 processes. Bacterial grazing and viral lysis was higher in the high CO_2 treatments during
424 periods of the experiment (Crawfurd et al., 2016), and would at least partly be the reason for
425 the reduced bacterial production at high CO_2 concentration.

426 N-limitation increased during the experiment (Paul et al., 2015), and mineral nutrient
427 limitation of bacteria can lead to accumulation of DOC, i.e. reduced bacterial uptake
428 (Thingstad et al., 1997), similar to our results. Bacterial N limitation is common in the area
429 during summer (Lignell et al., 2013), however, this N-limitation was not apparently different
430 in the controls (Paul et al., 2015), and CO_2 did not affect N-fixation (Paul et al., 2016). In a
431 scenario where the competition for N is fierce, the balance between bacteria and similar sized
432 picophytoplankton could be tilted in favor of phytoplankton if they gain an advantage by
433 having easier access to carbon, i.e. CO_2 (Hornick et al., 2016). We have not found evidence
434 in the literature that bacterial production will be suppressed in the observed pH range inside
435 the mesocosms, varying from approximately pH 8.1 in the control to pH 7.6 in the highest
436 fCO_2 treatment (Paul et al., 2015), although enzyme activity seems to be affected even by
437 moderate pH changes. For example, some studies report on an increase in protein degrading
438 enzyme leucine aminopeptidase activities at reduced pH (Grossart et al., 2006; Piontek et al.,
439 2010; Endres et al., 2014), whereas others indicate a reduced activity of this enzyme

(Yamada and Suzumura, 2010). A range of other factors affects this enzyme, for example the nitrogen source and salinity (Stepanauskas et al., 1999), and any potential interaction effects with decreasing pH are not yet resolved. Any pH-induced changes in bacterial enzymatic activity could potentially affect bacterial production.

3.7. Biological rates: primary production

There was an effect of CO_2 concentration on the net community production based on the organic carbon fraction (NCP_o). NCP_o was higher during Phase I than during the rest of the experiments and during this initial phase without any apparent CO_2 effect. There was no consistent difference between CO_2 treatments for NPP_{14C} ($p > 0.1$), but NCP_o increased with increasing CO_2 enrichment during Phase II (Phase II; linear regression $p = 0.003$; $R^2 = 0.91$). This was caused by the different development in the TPC and DOC pools. The pattern of gross primary production (GPP) was similar to NCP_o during Phases I and II. During Phase III there were no respiration or NPP_{14C} measurements and the estimated GPP is more uncertain. The NCP_o and GPP indicated a smaller difference between treatments during Phase III compared with Phase II.

The measures of NPP_{14C} and NCP_o were of a similar magnitude (Tables 1-3). During Phase I, $NPP_{14C} < NCP_o$ (Table 1), this relationship reversed for most treatments during Phase II, with the exception of the highest CO_2 levels (Table 2). The difference between NPP_{14C} and NCP_o suggests that observed reduction in respiration at elevated CO_2 could be mainly heterotrophic respiration. However, in terms of the $NPP_{14C} < NCP_o$, the uncertainty seems to be higher than the potential signal of heterotrophic respiration. This would also indicate that the NPP_{14C} during Phase I have been underestimated, in particular for the control mesocosm M1. During Phase II, the NPP_{14C} was higher than NCP_o, except for the two highest CO_2 treatments, more in line with our assumption of $NPP_{14C} > NCP_o$. The systematic offset in NPP_{14C} during Phase I could be due to changed parameterization during incubation in small volumes (8 mL, Spilling et al., 2016), for example increased loss due to grazing.

The results of the DIC pool and atmospheric exchange of CO_2 provides another way of estimating the net community production based on inorganic carbon (NCP_i). There was some discrepancy between the NCP_o and NCP_i as the latter suggested net heterotrophy in the ambient CO_2 whereas the high CO_2 treatments were net autotrophic during all three phases of the experiment (Fig. 3). For the NCPo there was no indication of net heterotrophy at ambient

CO_2 concentration. In terms of the absolute numbers, the NCPi estimate is probably more uncertain than NCP_o. Calculating the CO_2 atmospheric exchange from the measurements of a tracer gas involves several calculation steps (Eq 1-4), each adding uncertainty to the calculation. However, both estimations (NCPi and NCPo) indicate that increased CO_2 concentrations lead to higher overall community production, supporting our overall conclusion.

3.8 Budget

A carbon budget for the two control mesocosms and two highest CO_2 additions is presented in Fig. 5. During Phase I the estimated gross primary production (GPP) was ~100 mmol C fixed m^{-2} d^{-1}; from which 75-95% were respired, ~1% ended up in the TPC (including export) and 5-25% added to the DOC pool. The main difference between CO_2 treatments became apparent during Phase II when the NCP_o was higher in the elevated CO_2 treatments. The respiration loss increased to ~100% of GPP at the ambient CO_2 concentration, whereas respiration was lower (85-95% of GPP) in the highest CO_2 treatment. Bacterial production was ~30% lower, on average, at the highest CO_2 concentration compared with the controls during Phase II. The share of NCP_o of GPP ranged from 2% to 20% and the minimum flux to the DOC pool was 11% to 18% of TPC.

The overall budget was calculated by using the direct measurements of changes in standing stocks and fluxes of export, respiration and bacterial production rates. The most robust data are the direct measurements of carbon standing stocks and their development (e.g. ΔTPC). These are based on well-established analytical methods with relatively low standard error (SE) of the carbon pools. However, the dynamic nature of these pools made the relative SE for the rate of change much higher, reflecting that the rate of change varied considerably within the different phases.

The rate variables~~parameters~~, calculated based on conversion factors, have greater uncertainty, although their SEs were relatively low, caused by uncertainty in the conversion steps. For example, the respiratory quotient (RQ) was set to one, which is a good estimate for carbohydrate oxidation. For lipids and proteins the RQ is close to 0.7, but in a natural environment RQ is often >1 (Berggren et al., 2012), and is affected by physiological state e.g. nutrient limitation (Romero-Kutzner et al., 2015). Any temporal variability in the conversion

factors would directly change the overall budget calculations, e.g. RQ affecting total respiration and gross primary production estimates. However, the budget provides an order-of-magnitude estimate of the carbon flow within the system. Some of the variables~~parameters~~ such as GPP were estimated using different approaches, providing a more robust comparison of the different treatments.

The primary effect of increasing CO_2 concentration was the higher standing stocks of TPC and DOC compared with ambient CO_2 concentration. The increasing DOC pool and relatively higher TPC pool were driven by reduced respiration and bacterial production at elevated CO_2 concentration. Decreasing respiration rate reduced the recycling of organic carbon back to the DIC pool. The lower respiration and bacterial production also indicates reduced remineralization of DOC. These two effects caused the higher TPC and DOC pools in the elevated CO_2 treatments. The results highlight the importance of looking beyond net changes in carbon standing stocks to understand how carbon fluxes are affected under increasing ocean acidification.

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Table 1. The standing stock of total particular carbon (TPC_{pool}), dissolved organic carbon (DOC_{pool}) and dissolved inorganic carbon (DIC_{pool}) at the start of Phase I in mmol C m^{-2} ± SE (n = 2). The DOC_{pool} was missing some initial measurements and is the average for all mesocosms assuming that the DOC concentration was similar at the onset of the experiment. The net change in TPC (ΔTPC), DOC (ΔDOC) and DIC (ΔDIC) are average changes in the standing stocks during Phase I in mmol C m^{-2} d^{-1} ± SE (n = ~~16~~13, 9, 16, 7 and 11 for CO_{2flux}, EXP_{TCP}, TR, BP and NPP_{14C} respectively). SE for NCPo was calculated from the square root of the sum of variance of the three variables used in Eq 6. The NCP_o was calculated from the net change in carbon pools plus carbon export, whereas NPP_{14C} was measured carbon fixation using radiolabeled ^{14}C over a 24 h incubation period *in situ*. TR was measured as O_2 consumption and for comparison with carbon fixation we used a respiratory quotient (RQ) of 1. CO_{2flux} was only calculated for the period after full addition of CO2 (*t4-t16*). A total budget of carbon fluxes for ambient and high CO_2 treatments is presented in Fig 5.

Phase I (*t0-t16*)

CO_2 treatment (µatm *f*CO_2)	**365**	**368**	**497**	**821**	**1007**	**1231**
Mesocosm number	**M1**	**M5**	**M7**	**M6**	**M3**	**M8**
TPC_{pool}	417 ± 38	425 ± 39	472 ± 48	458 ± 38	431 ± 48	446 ± 57
DOC_{pool}	7172 ± 87	7172 ± 87	7172 ± 87	7172 ± 87	7172 ± 87	7172 ± 87
DIC_{pool}	25158 ± 9	25182 ± 10	25628 ± 8	26295 ± 22	26637 ± 36	26953 ± 48
ΔTPC	-4.6 ± 15	-5.2 ± 13	-8.3 ± 13	-8.2 ± 17	-7.0 ± 13	-6.3 ± 20
ΔDOC	15.5 ± 58	18.3 ± 30	18.5 ± 33	25.0 ± 36	18. 5 ± 73	18.1 ± 63
ΔDIC	5.5 ± 5.2	6.9 ± 9.2	-6.1 ± 11	-24 ± 14	-32 ± 20	-49 ± 42
CO_{2flux}	4.4 ± 0.2	4.8 ± 0.3	-0.8 ± 0.5	-11 ± 1.0	-17 ± 1.4	-23 ± 2.0
EXP_{TPC}	6.6 ± 0.10	5.6 ± 0.04	5.4 ± 0.07	6.0 ± 0.07	5.6 ± 0.06	6.0 ± 0.05
TR	107 ± 9	82 ± 7	81 ± 6	80 ± 8	75 ± 8	74 ± 8
BP	27 ± 8	41 ± 6	43 ± 8	41 ± 4	36 ± 5	46 ± 9
NPP_{14c}	4.8 ± 0.8	11.4 ± 2.1	14.9 ± 3.6	12.3 ± 2.3	11.3 ± 2.4	14.5 ± 2.7
NCP_o	17.4 ± 33	18.7 ± 20	15.6 ± 30	22.8 ± 28	17.1 ± 25	17.8 ± 28

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Table 2. The standing stock of total particular carbon (TPC_{pool}), dissolved organic carbon (DOC_{pool}) and dissolved inorganic carbon (DIC_{pool}) at the start of Phase II in mmol C m^{-2} ± SE (n = 2). The net change in TPC (ΔTPC), DOC (ΔDOC) and DIC (ΔDIC) are average changes in the standing stocks during Phase II in mmol C m^{-2} d^{-1} ± SE (n = ~~2~~7). Flux measurements of atmospheric gas exchange (CO_{2flux}) and exported carbon (EXP_{TPC}) plus biological rates: total respiration (TR), bacterial production (BP), measured (NPP_{14C}) and net community production estimated based on organic carbon pools (NCP_o), are all average for Phase II in mmol C m^{-2} d^{-1} ± SE (n = ~~1~~4, 7, 14, 5 and 14 for CO_{2flux}, EXP_{TCP}, TR, BP and NPP_{14C} respectively). See Table 1 legend for further details.

Phase II (*t17-t30*)						
CO_2 treatment (µatm *f*CO_2)	**365**	**368**	**497**	**821**	**1007**	**1231**
Mesocosm number	**M1**	**M5**	**M7**	**M6**	**M3**	**M8**
TPC_{pool}	339 ± 14	337 ± 20	331 ± 22	318 ± 9	312 ± 12	339 ± 23
DOC_{pool}	7435 ± 38	7483 ± 37	7487 ± 43	7597 ± 37	7487 ± 61	7479 ± 37
DIC_{pool}	25247 ± 34	25269 ± 34	25639 ± 8	26177 ± 25	26413 ± 28	26757 ± 45
ΔTPC	-2.4 ± 5	-2.3 ± 8	-1.6 ± 14	0.3 ± 6	2.8 ± 4	3.2 ± 8
ΔDOC	-0.6 ± 39	2.4 ± 30	3.6 ± 40	8.4 ± 31	11.3 ± 58	9.1 ± 36
ΔDIC	22.4 ± 12	17.6 ± 8.1	-0.4 ± 4.5	-10.5 ± 16	-14.2 ± 10	-23.1 ± 13
CO_{2flux}	1.7 ± 0.3	1.2 ± 0.3	-2.6 ± 0.3	-10 ± 0.5	-14 ± 0.6	-19 ± 1.0
EXP_{TPC}	3.3 ± 0.08	2.6 ± 0.06	2.5 ± 0.08	2.6 ± 0.06	2.8 ± 0.07	2.9 ± 0.06
TR	140 ± 7	127 ± 5	103 ± 3	103 ± 4	101 ± 5	86 ± 4
BP	66 ± 17	57 ± 8	61 ± 7	57 ± 7	43 ± 6	47 ± 6
NPP_{14c}	3.8 ± 0.6	11.2 ± 1.9	10.8 ± 2.0	14.3 ± 2.8	10.4 ± 2.1	12.0 ± 2.5
NCP_o	0.3 ± 20	2.7 ± 15	4.5 ± 22	11.4 ± 16	16.9 ± 19	15.2 ± 16

Table 3. The standing stock of total particular carbon (TPC_{pool}), dissolved organic carbon (DOC_{pool}) and dissolved inorganic carbon (DIC_{pool}) at the start of Phase III in mmol C m^{-2} ± SE (n = 2). The net change in TPC (ΔTPC), DOC (ΔDOC) and DIC (ΔDIC) are average changes in the standing stocks during Phase III in mmol C m^{-2} d^{-1} ± SE (n = ~~2~~6), using the average of the last two sampling days as the end point. Flux measurements of atmospheric gas exchange (CO_{2flux}) and exported carbon (EXP_{TPC}) plus biological rates: ~~total respiration (TR),~~ bacterial production (BP)~~, measured (NPP_{14C})~~ and net community production estimated based on organic carbon pools (NCP_o), are all average for Phase III in mmol C m^{-2} d^{-1} ± SE (n = ~~1~~3, 6, and 7 for CO_{2flux}, EXP_{TCP}, and BP respectively). See Table 1 legend for further details. During Phase III we did not have direct measurements of net primary production (NPP_{14C}) or total respiration (TR).

Phase III (*t31-t43*)						
CO_2 treatment (µatm *f*CO_2)	**365**	**368**	**497**	**821**	**1007**	**1231**
Mesocosm number	**M1**	**M5**	**M7**	**M6**	**M3**	**M8**
TPC_{pool}	306 ± 12	304 ± 20	309 ± 20	323 ± 2	351 ± 13	384 ± 16
DOC_{pool}	7426 ± 16	7469 ± 20	7485 ± 92	7553 ± 20	7593 ± 30	7562 ± 38
DIC_{pool}	25557 ± 9	25545 ± 10	25648 ± 13	26030 ± 19	26197 ± 31	26371 ± 32
ΔTPC	-3.8 ± 10	0.3 ± 7	3.3 ± 14	3.3 ± 10	-1.4 ± 8	-4.8 ± 8
ΔDOC	9.8 ± 5	8.8 ± 7	8.9 ± 43	9.2 ± 10	5.7 ± 17	16.3 ± 20
ΔDIC	4.3 ± 3.9	5.5 ± 8.7	6.2 ± 11	-12.3 ± 7.2	-16.3 ± 14	-20.1 ± 14
CO_{2flux}	-0.3 ± 0.7	-0.8 ± 0.6	-3.0 ± 0.5	-7.3 ± 0.5	-9.4 ± 0.6	-13 ± 0.6
EXP_{TPC}	1.5 ± 0.07	1.4 ± 0.05	0.4 ± 0.07	1.9 ± 0.05	1.6 ± 0.04	1.7 ± 0.05
BP	31 ± 6.8	37 ± 1.4	38 ± 1.4	27 ± 2.1	17 ± 3.8	28 ± 2.3
NCP_o	7.6 ± 16	10.5 ± 13	12.7 ± 20	14.3 ± 13	6.0 ± 10	13.2 ± 14

Figure legends

Fig. 1. The different fractions of carbon in the control mesocosms (M1 and M5) at the start of Phase I (t0), II (t17) and III (t31) in mmol C m^{-2} ± SE (n = 2). The differences between the controls and elevated CO_2 concentration are discussed in the text. The size of the boxes indicates the relative size of the carbon standing stocks.

Fig 2. The calculated exchange of CO_2 between the mesocosms and the atmosphere. Positive values indicate net influx (ingassing) and negative values net outflux (outgassing) from the mesocosms. The flux was based on measurements of N_2O as a tracer gas and calculated using equations 2-5.

Fig 3. Change in dissolved inorganic carbon (DIC) pool and the atmospheric CO_2 exchange (Fig. 2). All values are average mmol C m^{-2} d^{-1} ± SE for the three different phases (n = ~~16~~13, ~~14~~8 and ~~13~~7 for Phases I – III respectively) in the control mesocosms (M1 + M5) and high CO_2 mesocosms (M3 + M8). Black, solid arrows indicated measured fluxes. Grey, dashed arrows are estimated by closing the budget, and indicate the net community production based on inorganic carbon budget (NCP_i), which equals biological uptake or release of CO_2.

Fig 4. Standing stocks of total particulate carbon (TPC) and dissolved carbon (DOC) at the last day of the experiment (*t43*), plus the sum of exported TPC throughout the experiment; all values are in mmol C m^{-2} ± SE (n = 2). The values are averages of the two controls (M1 and M5) and the two highest CO_2 treatments (M3 and M8). Red circles indicate statistically significant higher standing stocks in the high CO_2 treatments (further details in text). The size of the boxes indicates the relative size of the carbon standing stocks and export.

Fig 5. Average carbon standing stocks and flow in the control mesocosms (M1 + M5) and high CO_2 mesocosms (M3 + M8) during the three phases of the experiment. All carbon stocks (squares): dissolved inorganic carbon (DIC), total particulate carbon (TPC) and dissolved organic carbon (DOC), are average from the start of the period in mmol C m^{-2} ± SE (n = 2). Fluxes (arrows) and net changes (Δ) are averages for the whole phase in mmol C m^{-2} d^{-1} ± SE (n presented in Table legends 1-3~~= 2~~) . Black, solid arrows indicated measured fluxes (Tables 1-3): total respiration (TR), bacterial production (BP), exported TPC (EXP_{TPC}). Grey, dashed arrows are estimated by closing the budget: gross primary production (GPP) using equations 7 and 8; DOC production (DOC_{prod}) using equations 9 and 10.

Bacterial respiration was calculated using equation 10 and is a share of TR (indicated by the parenthesis). Aggregation was assumed to equal BP. Red circles indicate statistically higher values compared with the other CO_2 treatment ($p < 0.05$, tests presented in the primary papers described in section 2.2.). The size of the boxes indicates the relative size of the carbon standing stocks.

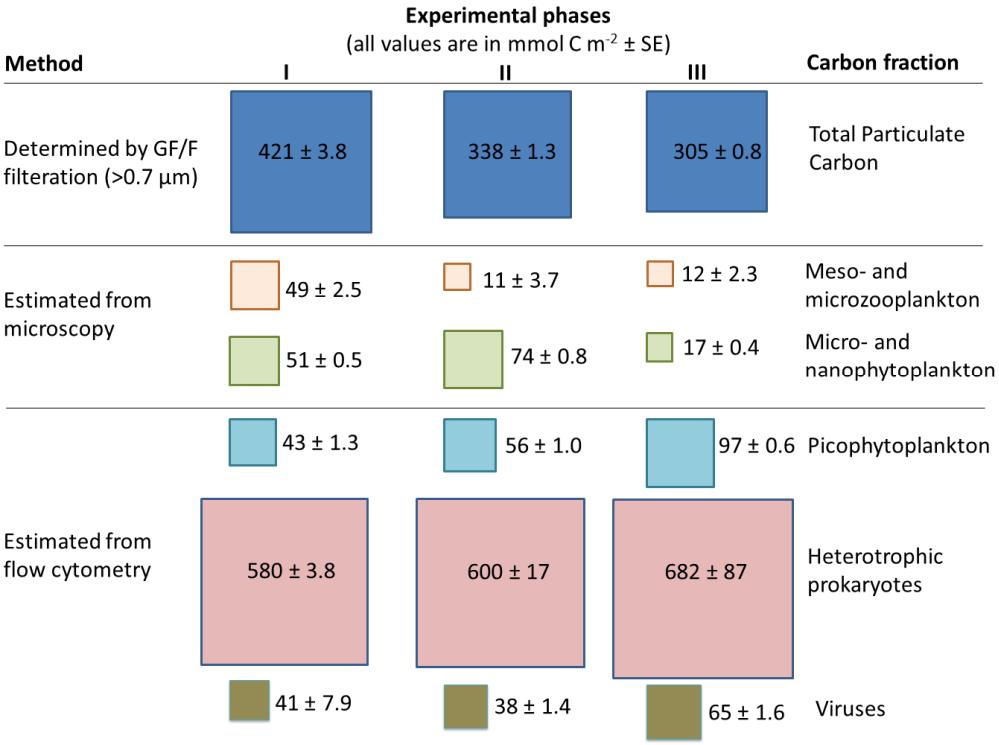
Experimental phases
(all values are in mmol C m-2 ± SE)
Method
I
II
III
Carbon fraction
Determined by GF/F filteration (>0.7 µm)
421 ± 3.8
338 ± 1.3
305 ± 0.8
Total Particulate Carbon
Estimated from microscopy
49 ± 2.5
11 ± 3.7
12 ± 2.3
Meso- and microzooplankton
51 ± 0.5
74 ± 0.8
17 ± 0.4
Micro- and nanophytoplankton
43 ± 1.3
56 ± 1.0
97 ± 0.6
Picophytoplankton
Estimated from flow cytometry
580 ± 3.8
600 ± 17
682 ± 87
Heterotrophic prokaryotes
41 ± 7.9
38 ± 1.4
65 ± 1.6
Viruses

Fig 1

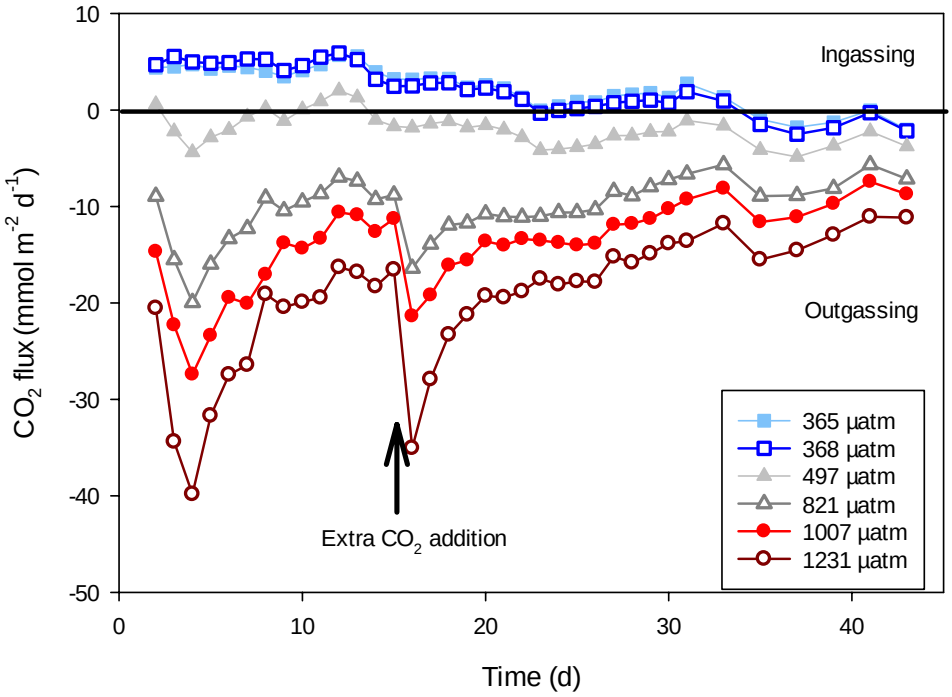
Ingassing
Outgassing
Extra CO2 addition
365 µatm
368 µatm
497 µatm
821 µatm
1007 µatm
1231 µatm
CO2 flux (mmol m-2 d-1)
Time (d)
10
0
-10
-20
-30
-40
-50
0
10
20
30
40

Fig 2

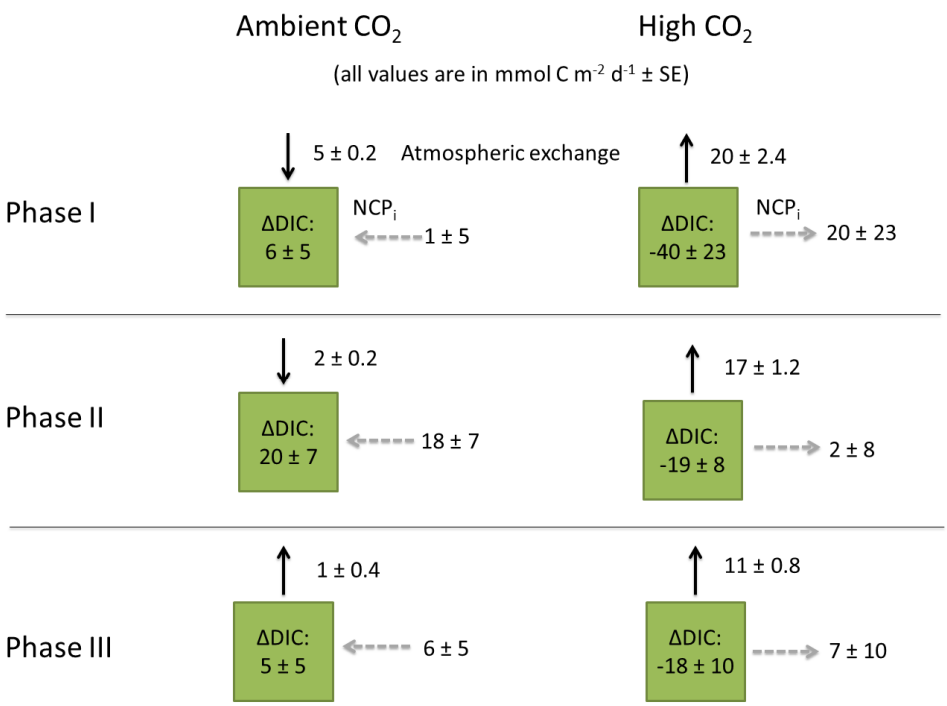
Ambient CO_2
High CO_2
(all values are in mmol C m^{-2} d^{-1} ± SE)
5 ± 0.2
Atmospheric exchange
20 ± 2.4
Phase I
ΔDIC:
6 ± 5
NCP_i
1 ± 5
ΔDIC:
-40 ± 23
NCP_i
20 ± 23
2 ± 0.2
17 ± 1.2
Phase II
ΔDIC:
20 ± 7
18 ± 7
ΔDIC:
-19 ± 8
2 ± 8
1 ± 0.4
11 ± 0.8
Phase III
ΔDIC:
5 ± 5
6 ± 5
ΔDIC:
-18 ± 10
7 ± 10

Fig 3

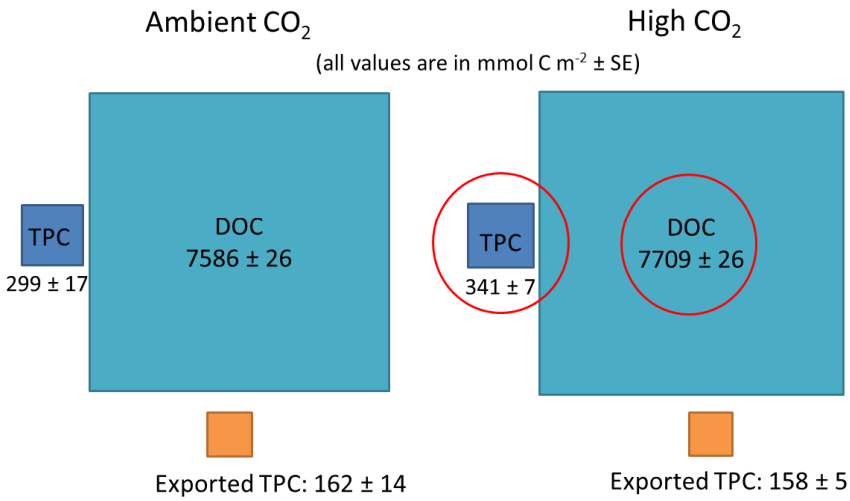
Ambient CO_2
High CO_2
(all values are in mmol C m^{-2} ± SE)
TPC
299 ± 17
DOC
7586 ± 26
TPC
341 ± 7
DOC
7709 ± 26
Exported TPC: 162 ± 14
Exported TPC: 158 ± 5

Fig 4

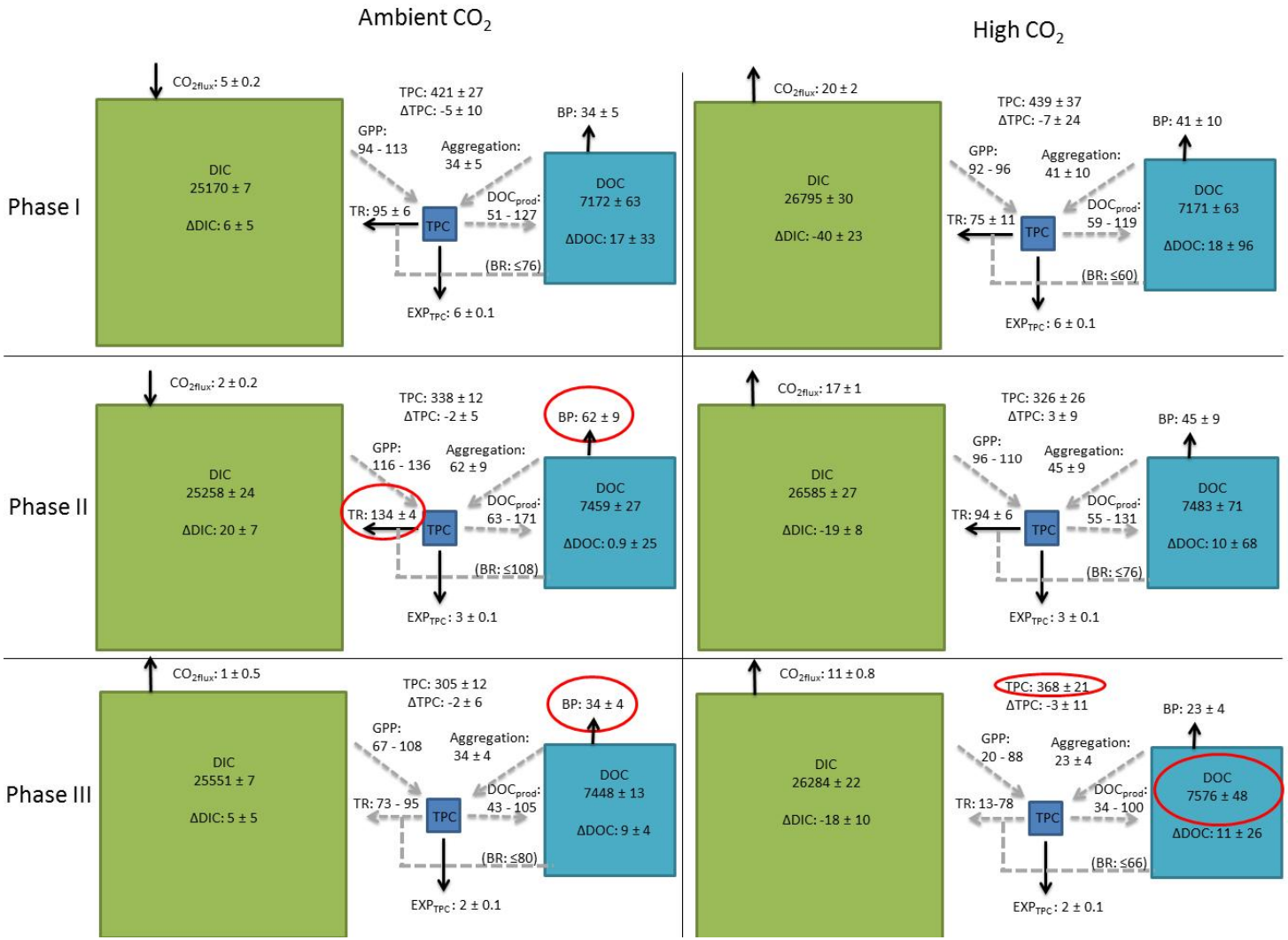
Ambient CO_2
High CO_2
Phase I
Phase II
Phase III
CO_{2flux}: 5 ± 0.2
DIC 25170 ± 7
ΔDIC: 6 ± 5
TPC: 421 ± 27
ΔTPC: -5 ± 10
GPP: 94 - 113
Aggregation: 34 ± 5
TR: 95 ± 6
TPC
DOC_{prod}: 51 - 127
BP: 34 ± 5
DOC 7172 ± 63
ΔDOC: 17 ± 33
(BR: ≤76)
EXP_{TPC}: 6 ± 0.1
CO_{2flux}: 20 ± 2
DIC 26795 ± 30
ΔDIC: -40 ± 23
TPC: 439 ± 37
ΔTPC: -7 ± 24
GPP: 92 - 96
Aggregation: 41 ± 10
TR: 75 ± 11
TPC
DOC_{prod}: 59 - 119
BP: 41 ± 10
DOC 7171 ± 63
ΔDOC: 18 ± 96
(BR: ≤60)
EXP_{TPC}: 6 ± 0.1
CO_{2flux}: 2 ± 0.2
DIC 25258 ± 24
ΔDIC: 20 ± 7
TPC: 338 ± 12
ΔTPC: -2 ± 5
GPP: 116 - 136
Aggregation: 62 ± 9
TR: 134 ± 4
TPC
DOC_{prod}: 63 - 171
BP: 62 ± 9
DOC 7459 ± 27
ΔDOC: 0.9 ± 25
(BR: ≤108)
EXP_{TPC}: 3 ± 0.1
CO_{2flux}: 17 ± 1
DIC 26585 ± 27
ΔDIC: -19 ± 8
TPC: 326 ± 26
ΔTPC: 3 ± 9
GPP: 96 - 110
Aggregation: 45 ± 9
TR: 94 ± 6
TPC
DOC_{prod}: 55 - 131
BP: 45 ± 9
DOC 7483 ± 71
ΔDOC: 10 ± 68
(BR: ≤76)
EXP_{TPC}: 3 ± 0.1
CO_{2flux}: 1 ± 0.5
DIC 25551 ± 7
ΔDIC: 5 ± 5
TPC: 305 ± 12
ΔTPC: -2 ± 6
GPP: 67 - 108
Aggregation: 34 ± 4
TR: 73 - 95
TPC
DOC_{prod}: 43 - 105
BP: 34 ± 4
DOC 7448 ± 13
ΔDOC: 9 ± 4
(BR: ≤80)
EXP_{TPC}: 2 ± 0.1
CO_{2flux}: 11 ± 0.8
DIC 26284 ± 22
ΔDIC: -18 ± 10
TPC: 368 ± 21
ΔTPC: -3 ± 11
GPP: 20 - 88
Aggregation: 23 ± 4
TR: 13-78
TPC
DOC_{prod}: 34 - 100
BP: 23 ± 4
DOC 7576 ± 48
ΔDOC: 11 ± 26
(BR: ≤66)
EXP_{TPC}: 2 ± 0.1

Fig 5