

Reply to anonymous referees one and two:

We thank the two anonymous referees for their critical remarks on our manuscript “Elemental budgets in an Arctic CO₂ perturbation study”. Corrections and amendments as suggested by the referees helped to improve the quality of the manuscript substantially.

General comments:

By the authors:

The approach of calculating elemental budgets and analysing elemental fluxes by mass balance is the objective of our manuscript, and seems to be a logical approach when elements in all possible major forms were measured in a closed system. This is in so far a novel concept for carbon in mesocosm research as we are the first to present actual measurements on all major carbon pools and fluxes. Pool X should be theoretically zero in a closed system, as changes in each dataset should be balanced by changes in one or several others. Therefore Pool X is presented to demonstrate that all individual measurements cannot be simply summed up to close the carbon budget in our experiment. We assumed that information given in Figure 3 was sufficient to demonstrate for the replicated control mesocosms that (1) bulk variability in Pool X for carbon is caused by variability in DOC measurements and (2) increase or decrease in pools beside DOC was balanced by changes in other pools so that virtually no carbon was available in the system to form DOC or to be lost into undetermined pools. We agree with the referees' opinion that this might not be sufficient to justify the calculation of DOC, DON and POP from mass balances and we will improve the revised manuscript by including more data to define uncertainties of this approach (Tab. A,B, Fig. A).

Besides clarification of these technical issues of mass balance calculation, textual changes in the abstract and the objective section at the end of the introduction will be added to the revised manuscript to better introduce the reader to the concept.

1. Remark by referee 1: *Thus, although in the abstract they define the intention of considering all the elemental pools, they were not able to measure with sufficient accuracy all the dissolved elements (DOC, DON and DOP), they did not measure zooplankton contribution and the flux of particulate organic matter was only partially considered. Therefore to close the gap for each element budget the authors use a set of “Pool Xs” which should include the dissolved pools, part of the sedimentation, and the larger zooplankton.*

We will mention our lack of sufficient precision in determining DOC, DON and POP in the abstract and include worst case estimates of the uncertainties in mass balance calculations, which are for instance the unaccounted contribution of zooplankton

The “dubious” name “Pool X” for calculated changes in DOC, DON and POP (including uncertainties listed in the table A) will be changed to DOC_{calc}, DON_{calc} and POP_{calc}. This should prevent confusion of the loss fraction “Pool X” (including all measured variables) with mass balance estimates for selected variables.

In table A we show that these unaccounted pools are minor compared to uncertainties in DOC and also DON and POP determination. They are roughly one order of magnitude lower than estimated changes in DOC, DON and POP calculated by the mass balance. Moreover unaccounted losses can only cause a gradual increase of Pool X not a decrease, whereas both, increases as well as decreases of DOC_{calc}, DON_{calc} and POP_{calc} were observed.

Table A

Estimates for the contribution of undetermined carbon pools (or not daily determined pools) to Pool X or DOC_{calc}. Sediment losses were estimated on the basis of the ratio of surface areas of the funnel relative to the gap around the funnel ($1 \cdot \text{radius of the sediment trap flotation ring}$) and the average cumulative sediment trapped during each phase. Copepod biomass

changes were estimated from average numbers and carbon content determined from weekly net hauls, assuming all copepods were not represented in the particulate carbon measurements. Wall growth for phases II and III (after nutrient addition) was estimated, assuming exponential growth of the wall grown carbon measured on t30 ($8.31 \pm 3.1 \mu\text{mol kg}^{-1}$) at rates of 1 to 0.3 per day (Hagseth et al. 1992). “Maximum contribution of undetermined pools” depicts the sum of estimated mean changes within the three listed pools plus one standard deviation, or in the case of wall growth, the largest estimate.

| Phase | I | II | III |
|---|-------------------------------------|------------------------------------|------------------------------------|
| | $\mu\text{mol kg}^{-1}$ | $\mu\text{mol kg}^{-1}$ | $\mu\text{mol kg}^{-1}$ |
| Sediment lost into dead volume | 0.046 ± 0.007 | 0.10 ± 0.015 | 0.17 ± 0.063 |
| Copepods escaped | -0.33 ± 0.44 | 0.26 ± 0.43 | 1.11 ± 0.78 |
| Wall growth | non | 0.081 to 0.82 | 1.0 to 3.4 |
| Maximal sum of undetermined pools | 0.12 | 1.63 | 5.52 |
| Observed range of DOC_{calc} on the last day of the phase | -0.93 to 11.09 | -7.02 to 8.81 | 19.32 to 30.5 |

Zooplankton biomass was not excluded from particulate carbon filtrations. Hence it is at least partly included in mass balance calculations. Zooplankton was dominated by cirriped nauplii and copepods of the genus *Calanus*. While cirriped nauplii were probably well represented on our filters copepodid stages are probably not, owing to their ability to escape our water sampling device. Large copepods were observed on filters only sporadically and are probably one of the main reasons for day to day variability in results for water column particles.

There was no open gap around the sediment funnel, but the upper edge of the funnel formed by the floatation ring was round (D=6cm). Therefore, particles settling on the outer 3cm radius of this ring could be transported away from the funnel towards the bag obviously seeping into the dead volume. A potential gap area of 5.9% of the total mesocosm area was calculated on the bases of one radius of the flotation ring. (Tab. A)

2 Remark by referee 1: *They use the variation of these poolXs for most of their conclusions assuming such pools mainly constituted by dissolved elements (DOC, DON and DOP). This assumption is not supported by any results presented in this or other studies.*

We will include and discuss correlations of the total loss fractions (Pool X) to changes in a single variable, for instance changes in DOC (Fig. A).

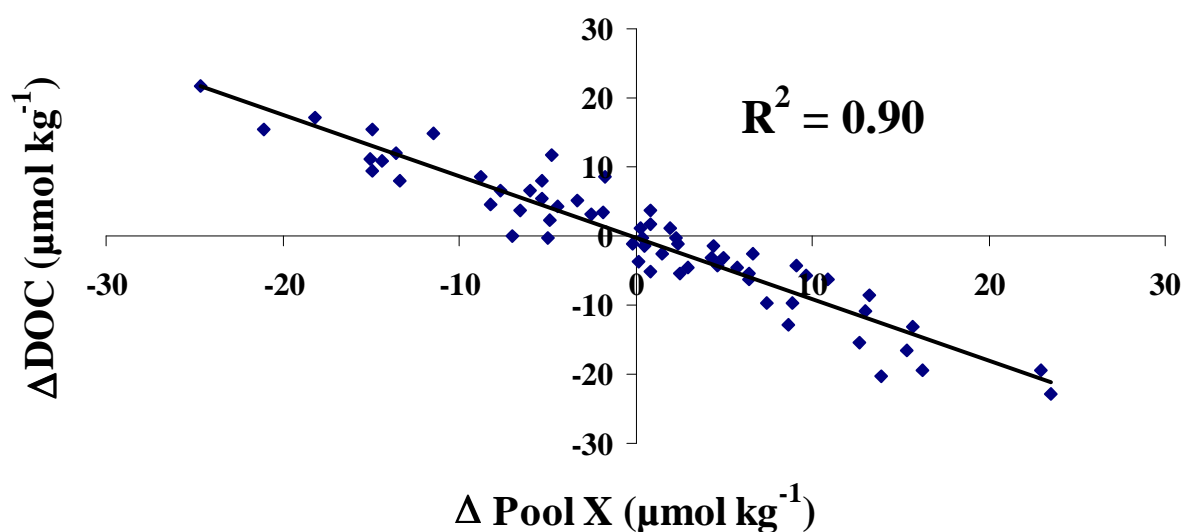


Figure A

Linear correlation of changes in Total Pool X ($\Sigma \Delta PC$, ΔDOC , ΔCT , ΔSed , ΔGX) to changes in measured concentrations of DOC in all mesocosms during phase II.

Up to 90% of the variance in Pool X can be explained by the variance in DOC. The intercept of the linear model is close to zero ($-0.19 \mu\text{mol kg}^{-1}$), supporting the conclusion that there were no significant changes in DOC or carbon losses during this period. Statistical results of correlations of Total Pool X to other carbon pools are listed in Table B.

| Correlation of Pool X to: | | PC | DOC | CT | GX | Sed |
|---------------------------|-------|------|------|------|------|------|
| Phase I | r^2 | 0.01 | 0.85 | 0.01 | 0.00 | 0.03 |
| | p | | | | | |
| Phase II | r^2 | 0.00 | 0.90 | 0.11 | 0.09 | 0.10 |
| | p | | | | | |
| Phase II | r^2 | 0.08 | 0.51 | 0.24 | 0.08 | 0.22 |
| | p | | | | | |

Table B

Regression coefficients depict the share of variances in Pool X explained by variances in the datasets it is based on.

In phase I and II, variances in Pool X can be almost entirely explained by variances in DOC, while in phase III inorganic carbon absorbed by wall growth as well as sediment not yet sampled on the last day might also have contributed considerably to Pool X (Tab. A). Trends for DOC are significant on a <5% level precise p values will be included in the revised manuscript (the Statistica server is not running at the moment)

Similar results as shown in table B were obtained in equivalent analyses for the nitrogen and phosphorus budget. Here 72% of the variance observed in Pool X could be explained by variance in DON and 56% by variance in POP for phosphorus respectively.

3 Remark by referee 2: *“Unfortunately, the authors do not stick to the observations, but create a dubious pool X, which is not directly measured, but estimated by difference assuming mass balance in arbitrarily selected variables.”*

See answers above.

4 Remark by referee 1:

„ the numerous parallel studies performed during the same experiment constitutes an invaluable opportunity to understand this particular ecosystem functioning. It is therefore disappointing to read through the MS and to discover that the authors did not profit of such opportunity building their conclusions more on assumptions than on solid data.”

We agree with the referee’s opinion that conclusions made in this manuscript could profit from comparison with data presented in parallel studies within this special issue. However, it is not within the scope of this manuscript to understand ecosystem functioning. Data presented on changes in bulk elemental pools are not specific enough to draw conclusions on physiological or ecological processes. The focus of our manuscript is on elemental cycling and export. We will clarify this at the end of the introduction.

We will also include comparisons of our results with data within other manuscripts, such as:

1. We will indicate the range of bacterial carbon demand measured by Motegi et al. (2012) as well as community respiration measured by Tanaka et al. (2012) in direct comparison to carbon export fluxes. This should give indication on whether the ecosystem was dominated by recycling or export.
2. We will discuss findings on export and particulate carbon composition as well as production rates by de Kluijver et al. (2012) in the context of determined overall carbon fluxes.
3. We will compare trends in uptake ratios of inorganic nutrients presented by Silyakova et al. (2012) to biomass build up.
4. We will improve referring to Schulz et al. (2012) and Brussard et al. (2012).

5 Remark by referee 1:

The results of parallel studies during the same experiment are only used when they confirm the authors' conclusions.

Due to the large quantity of data gathered during this experiment we only refer to selected results relevant to our results and useful for understanding measured effects on carbon fluxes. However, we could not find results directly contradictory to our conclusions.

6 Remark by referee 2:

“However, nutrients (inorganic N and P) were added in equal amounts to all mesocosms on Day 13 to boost phytoplankton growth, and this complicates extrapolation of the results to natural conditions, as CO₂ and nutrient treatment effects cannot be statistically differentiated with this design.”

We do agree with the referee’s opinion that the addition of nutrients further complicates the extrapolation of the results to natural conditions. If nutrients would not have been added the observed system would have been simpler.

We do not agree with the remark that nutrient addition is affecting statistical power to detect CO₂ effects. Within a developing ecosystem other than within a chemostat, nearly none of the detected changes can be attributed to be a direct treatment response anyways. If phytoplankton grows faster it might be due to increased carbon availability at high CO₂, but might also be an indirect effect of changed nutrient speciation or enhanced remineralisation due to elevated bacterial exoenzymatic activity in response to lowered pH. After an initial treatment mediated response inside a mesocosm many important growth parameters have

changed due to differences in uptake or production of matter. With time the detection of direct CO₂ effects becomes more and more difficult. Whether or not nutrients were added is here of secondary importance. We think that especially those indirect effects are most interesting and probably highly relevant in natural systems.

We will improve the manuscript so that it becomes clearer that we detect correlations to treatment CO₂ but do not imply that we detected simple direct effects of CO₂ on common eco-physiological processes.

Specific comments by referee 1:

1 (page 11886, lines 10-13) *The sentence “all relevant element pools and fluxes of carbon, nitrogen and phosphorus were measured, using an improved experimental design intended to narrow down some of the mentioned uncertainties” set the reader expectation very high.*

We will include a description of the Pool X concept, and the approach to calculate DOC, DON and POP from mass balances in the abstract.

2 (page 11886, lines 19-20) *“Enhanced carbon consumption appears to result in accumulation of dissolved organic compounds under nutrient recycling summer conditions” This is not supported by data*

Enhanced carbon consumption at high CO₂ during phase I is supported by CT data corrected for gas exchange (see also NCP by Silyakova et al., 2012). There is no carbon pool other than DOC to accommodate this consumption, as indicated by the mass balance calculation being subject of this manuscript. Beside these results, general DOC production during this time can be even detected in statistical examination of the contaminated DOC samples (Engel et al., 2012; Schulz et al., 2012). Elevated DOC production at high CO₂ was also measured by ¹⁴C DOC production (Engel et al., 2012).

3 (page 11886, lines 22-24) *“The out-competing of large diatoms by comparatively small algae in nutrient uptake caused reduced production rates under future ocean CO₂ conditions in the end of the experiment” The authors only assume that they were competing for nutrients.*

We will rephrase this sentence because competition for nutrients by phytoplankton growing in two different phases of the experiment is obviously misleading.

4 (page 11888, lines 3-5) *This is not entirely correct. Pteropods are important only in limited area of the ocean as also reported by the cited author.*

We will restrict the statement to Arctic regions.

Technical comments:

5 (page 11888, line 20) *“but surface ocean warming” can be removed since this is a consequence of the increasing temperature.*

Entirely removing would change the meaning of the sentence but we will improve it.

6 (page 11888, line 6) *Remove “often” and change “is keeping” with “may keep” (page 11888, line 11)*

Will be changed accordingly.

7 (page 11888, line 11) *“in global carbon flux models” add “some” and add more references.*

We will additionally cite Bopp et al. (2001) and Schmittner et al. (2008)

8 (page 11888, line 28 continuing at page 11889 lines 1-2) *remove*

Removed.

9 *It is not clear why the mesocosms were closed at day 7*

This is a misunderstanding by the reviewer; the closing day is stated (page 11890 line 15) to be t-5 which is 5 days before the start of the experiment (sampling of the manipulated mesocosm). On t-7 (page 11890 line 15) the bags were lowered into the water covered with a net on both openings. For clarification, we will re-write the respective sentences.

10 *Please explain (page 11891 line 13) Should I understand that the authors performed two salt additions during the experiment? When was the first addition performed? Please clarify.*

This was understood right! For clarification, we will re-write the respective sentences.

11 *There is some confusion, at least for me, in the total duration of the experiment and the duration of the analysis. Please clarify*

We do not exactly understand what analyses are referred to by the reviewer. We will insert a sentence on page 11890 line 24:

"The entire experiment from filling (t-7) to the last sampling (t31) of the mesocosms lasted 39 days. Sampling for all variables started at t-3, three days prior to the beginning of the CO₂ manipulation and lasted until t31 for most variables presented here." We also will improve the paragraph describing the reason for the particular starting point chosen for carbon mass balance (Page 11893 line 23 and 24 to Page 11894 line 1-5).

12 *The text is difficult to follow because of the use of different terminology: e.g. "Dissolved substances" (page 11890 line 22) "Dissolved and particulate parameters" (page 11891 line 21) "Particulate and dissolved substances" (page 11891 line 26) "Particulate matter" (page 11892 line 1) "sediment" (page 11892 line 3) "particles" (page 11892 line 5) Please be more precise and consistent.*

In the revised manuscript we will consistently use the term "matter" for water column material and "sediment" for settled material.

13 *If the sampling device reach 12m depth and the sediment collector is at 15m depth this means that there is about 4.2 m³ of water, 8.4-5.6% of the total volume which should be sampled daily to "measure all the relevant element pools and fluxes"*

We chose to sample in safe distance from the sediment funnel in order to prevent dispersion of sediment. With the lower end of the water sampler on 13m, the inlet is on the upper end 12m. The integrating water sampler is taking a water sample representative for the center of the mesocosm over the upper 12 of the 15m total water depth. We are confident that the rest of the water inside the mesocosm has very similar properties to our samples and we will clarify this in the method section.

14 *It is not clear which is the volume of water sampled every day from the sedimentation trap. Please clarify*

We will indicate the volume of the samples (it is ~3 l) and describe gravimetric sub-sampling for microscopic inspection.

15 *In the results is reported the presence of cirripeda larvae in the sediment (page 11900 line 6). I presume therefore that the authors performed microscopic analysis of the sediments but it is not explained. Please clarify.*

We will include a sentence describing the microscopic analyses of sediment samples. Results are reported by Niehoff et al. (2012).

16 *(page 11893 lines 13-15) It is acceptable to cite another source for describing a technique but the short description left should be understandable....I am not sure to get what "implementing measured CO₂ gradients". Please explain better.*

We will improve this paragraph in the revised version of the manuscript.

17 (page 11894 lines 1-2) *Of which equilibrium are you talking about and how was it measured? Do you mean the CT concentration between the water column and the dead volume? Please explain it better.*

We are talking about equilibration of the dead volume with the water column. We rely on the assessment by Silyakova et al. (2012) and Bellerby et al. (2012) that performed measurements. We will improve this section.

Technical comments:

18 (page 11890 line 15) *“were unfolding themselves” Please rephrase.*
We will rephrase.

19 (page 11890 line 12) *“mesh” not “mash”.*
Changed

20 (page 11890 lines 22-24) *“nonetheless. . .” Please rephrase.*
We will rephrase.

21 (page 11891 lines 5-7) *“The replicate measured volume. . .” This sentence is really unclear, please rephrase it.*
This section will be rephrased.

22 (page 11891 line 13) *Please remove “early” (page 11891 line 21) “dissolved and particulate parameters” Please be more precise.*
“Early” will be replaced by “Before routine sampling”
“dissolved and particulate parameters” will be replaced by introduction of these parameters and their abbreviations.

23 (page 11891 line 22) *“Which is the IWS total volume?”*
Will be clarified

24 (page 11891 line 26) *The word “substances” is too vague. Please be more precise.*
Will be improved using the term “matter “ as defined in response 22

25 (page 11892 lines 1-3) *“Installed . . .” Please rephrase*
The sentence will be simplified

26 (page 11892 lines 3-4) *“Sediment suspended. . .” Please rephrase with something like: ??*
L of water was sampled inside a glass bottles applying a vacuum pressure to the end of the tube.

Will be rephrased as:

“Sediment dispersed in approximately 3 l of water was drawn from the silicone tube into a glass bottle by low vacuum.”

27 (page 11892 line 8) *“Particulate matter” please change with “water” Please indicate the volume filtered for each analysis.*
Changed

28 (page 11893 line 1) *Dissolved inorganic carbon should be indicated with “DIC”*

We decided to stick to CT as an abbreviation for total inorganic carbon to provide consistency to Bellerby et al. (2012) and Silyakova (2012), who provided the data. Dissolved inorganic carbon will be now consistently called total inorganic carbon CT throughout the revised manuscript.

29 (page 11893 line 6) *I am not sure to understand what "slowly freeze dried" means. Are this the POM pellets which were stored in the -80 freezer?..I do not understand. Please explain better and rephrase the entire paragraph. (page 11893 line 12)*

Slowly freeze dried means that no other energy (heat) was applied than the one coming in from the surrounding. This was done to preserve the natural phytopigments as good as possible. These pellets were cooled to -80°C to prevent melting during the time when vacuum was building up in the freeze dryer.

We will rephrase in the sampling section and the analytic section.

30 *Please change “deliberate tracer” with “gas tracer” (page 11893 line 18)*

Changed

31 *Please remove “on” and change “days” with “day”*

Changed

Results specific comments

32 (page 11896 line 16) *Isn't it “t-7” the day in which mesocosms were closed (page 11890 line 10)?*

t-7 is the day when mesocosms were filled. Two weeks later on t7 is the first bloom peak. We will clarify the nomenclature of the experimental days in the description of the duration of the experiment.

33 (page 11896 lines 16-18) *This sentence should be in “discussion” since does not refer to reported data*

The sentence will be deleted.

34 (page 11897 lines 7-9) *What ratio was used for phase three?*

In the BGD manuscript we used as stated:

”a ratio of 0.036 gChl *a* gC⁻¹ for the phase III peak, when diatoms were abundant.”

Detailed assessment of water column phytopigments revealed that the contribution of diatoms to water column phytoplankton biomass was rather small throughout the experiment, in a revised manuscript we will use 0.02 gChl *a* gC⁻¹ for the entire experiment.

35 (page 11897 lines 9-11) *I am surprised to not see an increase in chl *a* in day 30 after the brushing of the wall. Should I assume that the brushing was performed after the Chl *a* measurement? Please clarify.*

The referee is right. We will clarify this issue when describing the in the wall growth estimate in the method part that wall cleaning was done after routine sampling.

36 (page 11897 lines 14-15) *Sedimentation does not mirror the Chl *a* bloom development.*

Right, sedimentation in this phase is not a big issue and we will change the sentence accordingly.

37 (page 11898page 11898 C5191 line 12) “averaged over all mesocosms” Does this mean that on average on each bag walls there were $7.3 \mu\text{mol C kg}^{-1}$. In this case it would be interesting to know the standard deviation or at least the range.

Wall growth of POC, PON, POP was measured only once in each bag except bag 7 that was opened to remove the sediment funnel used to build the wall brush and bag 1 that was destroyed by the first attempt to use it. Results of the remaining seven bags were rather variable with average carbon being $8.31 \pm 3.1 \mu\text{mol C kg}^{-1}$ among the seven mesocosms and there was no CO_2 effect detectable. We now included a table (Tab. A) assessing the contribution of wall growth to Pool X based on all carbon wall growth measurements.

38 (page 11899 lines 22-23) Figure 5d is PoolX for phase one...I do not understand?
Sorry, we intended to refer to Figure 5e.

39 (page 11900 line 1) Growth of what? Phytoplankton?
We re-phrased with “phytoplankton production”.

40 (page 11900 lines 5-10) If this is reported in "results" then the zooplankton analysis method should be reported in M&M

A table (Tab. A) with zooplankton biomass and its contribution to Pool X was added and methods are reported in the sampling and analytic sections. (See also reply to comment **15**)

41 (page 11900 lines 11-14) Or it goes to Discussion or has to be described in M&M

We will move the paragraph describing the pteropod addition to the section describing the setup.

42 (page 11900 lines 21-23) Why does it seem possible if it is not indicated by the data?

It seems possible because of the differential nutrient situation at the onset of phase III. See point **43**. Because data on wall growth have poor resolution and Pool X during phase III is an equation with two unknowns, an undetected correlation of wall growth to CO_2 seems possible.

43 (page 11900 lines 25-28) From Schulz et al 2012 I can not detect this difference in N and P concentration between High CO_2 and low CO_2 in phase III (page 11901 lines 1-3)

It could be clearer in Schulz et al 2012 (p12557 line 16-20)

“ NO_3^- and PO_4^{3-} were then readily taken up by the plankton community, declining towards detection limits until the end of the experiment. Immediately after nutrient addition, however, nutrient utilization of both NO_3^- and PO_4^{3-} was faster at higher CO_2 levels during phase II, while being slower during phase III (Fig. 6b and d). This observation was statistically significant.”

We will include DIN and DIP concentrations on t20 in the results part of this manuscript. We also constructed linear correlations of inorganic nutrient availability at the onset of phase III to actual pCO_2 , as well as of nutrient availability to cumulative sedimentation of carbon during phase III (Fig. B,C,D).

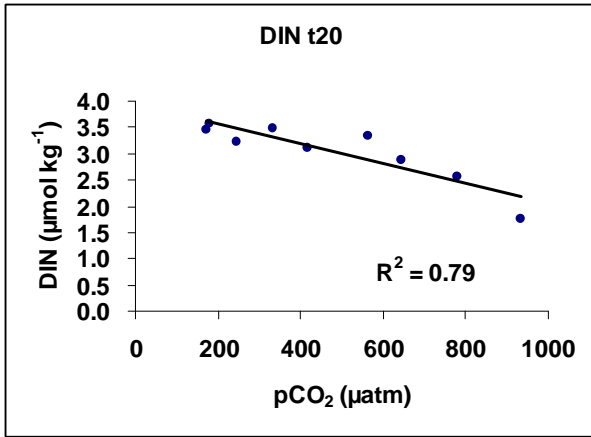


Figure B

Linear correlation of dissolved inorganic nitrogen (DIN) to treatment CO₂ on the beginning of phase III (t20)

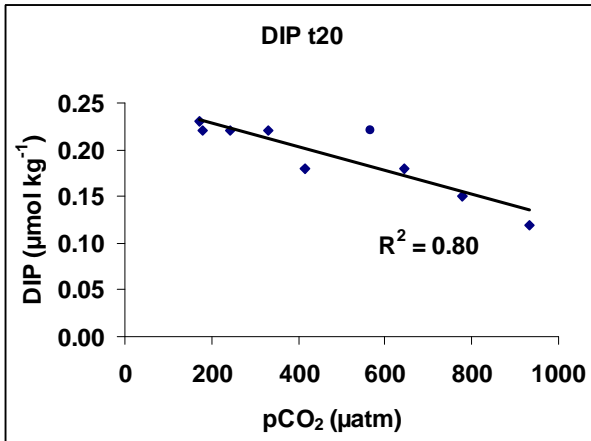


Figure C

Linear correlation of dissolved inorganic phosphorus (DIP) to treatment CO₂ on the beginning of phase III (t20)

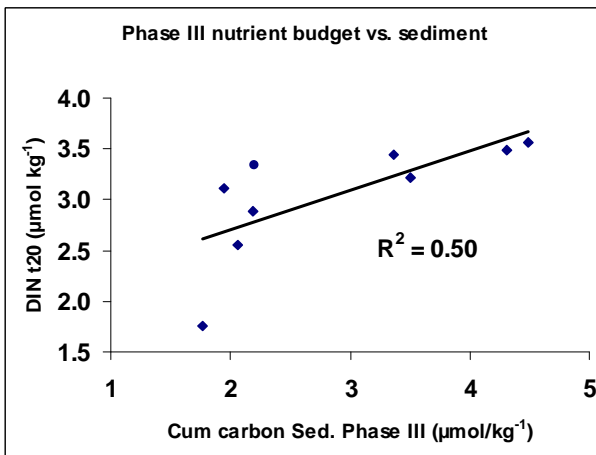


Figure D

Linear correlation of cumulative carbon sediment during phase III (t20-t27) to dissolved inorganic nitrogen (DIN) on the beginning of phase III (t20)

At the beginning of phase III (t20) inorganic nutrient concentrations (Fig. B and C) were correlated to treatment CO₂. The amounts of inorganic nutrients available in the low CO₂ control treatments were roughly double of the amount available in the highest CO₂ treatment. Therefore, indirect effects of CO₂ via nutrient availability caused by CO₂ correlated uptake during phase II are presumably responsible for increased production at low CO₂ during phase III. Sedimentation during phase III is negatively correlated to CO₂ and positively to nutrient availability at t20 (Fig. D). Direct CO₂ effects during phase III are possible but can not be distinguished from nutrient effects.

44 (page 11901 lines 1-3) *How did you estimate the net autotrophic growth rate?*

We estimated net community inorganic carbon uptake (ΔCT_{GX}). We will change the wording to “net community carbon production”

45 *Figure 3 (page 11920) If the wall brushing was performed at day 30, why the dot is plotted at day 27?*

We plotted it on t27 because t30 would be out of scale. Wall growth from t30 is not directly comparable to the dataset ending on t26. Therefore, the data point was removed and wall growth is reported in the text and a table (Tab. A).

Discussion Specific comments

46 (page 11902 lines 13-14) *Viral lyses of what? Grazing by whom?*

Now that an interpretation of the Brussard data is on the web it is no problem to include this information.

47 (page 11902 lines 19-22) *I am not able to find the base of these assumptions. Please explain better*

Rapid cycling of matter during phase I due to high growth rates and high loss rates is documented by Brussard et al. (2012) (loss rates), de Kluiver et al. (2012) (growth rates and BP/PP), Montegi et al. (2012) (bacterial carbon demand) and Tanaka et al. (2012) (community respiration).

We will include reference to measured heterotrophic processes.

Mass balance is telling us that dissolved carbon formed at high CO₂ was stable at least during phase II (then wall growth is interfering with further interpretation of mass balance). Net production of DOC during phase I and stable values for phase II is also supported by interpretations by Engel et al. (2012) based on statistics on DOC measurements and C¹⁴ DOC production.

48 (page 11903 lines 21-24) *This hypothesis is not supported by the data of Brussard et al 2012.*

The referee is right. But the general hypotheses, that surface layer DOC production is rather promoting remineralisation than export, can be stated here independently of whether or not effects on bacterial abundance were directly measured during the limited experimental period or not. If DOC would be of a refractory nature owing to lifetimes in the range of deep ocean turnover times, it could also be considered to be export (Ridgwell 2011). We will include this aspect into the syntheses and corrected tense of the sentence.

The fact that there is no positive or even a negative response (phase III) of bacterial abundance to CO₂ reported by Brussard et al. (2012) does not mean that bacterial growth was not “supported” by substrate availability. The hypothesis of enhanced cycling of organics by a larger microbial community is presented in the conclusions by Brussard et al. (2012) in a similar way and also by de Kluiver et al. (2012).

49 (page 11904 line 15) *How the authors can assert that Pool X was mainly constituted by DOC. It could be entirely due to the "undetermined pools" and not to DOC*

See reply to general comment one and two.

Even if 50% of the sediment would have been lost or all zooplankton escaped from our water samplers (we know that was not the case). This would be not enough carbon to affect our interpretation of the mass balance.

50 (page 11904 lines 23-25) *I do not see any differences in nutrient concentration between the treatments.*

See point **43**

Synthesis Specific comments

51 (page 11907 lines 5-7) *I am not able to find the bases for this statement.*

Enhanced carbon uptake is evident in ΔCT_{GX} , indication that it was obviously channelled into DOC is the result of our mass balance calculation (as discussed before in general comments and specific comment **2**, and **47**).

52 (page 11907 lines 11-13) *The authors can not state this on the base of their data. I am not able to see how pH controlled the ecosystem productivity.*

We do neither know how nor if pH or CO₂ controlled ecosystem productivity. We simply detect that nutrient uptake as well as carbon uptake and partitioning was obviously correlated to our pH/CO₂ treatment.

53 (page 11907 lines 16-17) *What the authors mean with "characteristic effect of CO2"? Please explain better.*

We rephrased this sentence

54 (page 11907 lines 17-19) *This, in my opinion, means that are nutrients more than the Co2 to control the productivity of the system.*

This is generally true and does not need to be discussed as CO₂ is not a limiting nutrient in seawater. But it is nicely shown that CO₂ also affects productivity and productivity affects nutrients.

55 (page 11908 lines 4-6) *I have not seen evidence of increased DOM due to CO2 and I have not seen any data regarding the size of phytoplankton.*

We are now talking about the experiment in a whole; we will make this clearer by replacing the word "study" by "experiment".

56 (page 11908 lines 19-21) *May be Brussard et al did it but the data are not presented in this study.*

See point **55**. We rephrased the sentence.

57 (page 11908 line 23) *What are follow up effects? Please be more precise.*

We rephrased:

"Whereas the growth effect on picoeucaryotes itself had no effect on carbon export fluxes and could not even be clearly detected in POM, their footprint in the nutrient budget was indeed of biogeochemical relevance."

Specific comments by referee 2:

1 The ms title “Element budgets in an Arctic mesocosm CO₂ perturbation study” is misleading, since only chlorophyll a results are shown in absolute values, all particulate and dissolved nutrient pools being presented as treatment responses or temporal changes in 3 growth phases vs. (subtracted) initial reference value.

The title was changed to “Elemental mass balance in an Arctic mesocosm CO₂ perturbation study”.

2 It is not clear why the response of the indigenous plankton community from Day 0 to Day 8 is left out of the treatment (CO₂ perturbation and “bag effect”) examinations, when CO₂ treatment started already on Day -1 (and continued until Day 4)?

We will make this clearer in the method section:

“The addition of CO₂ saturated water caused major changes in the CT budget that could only be precisely quantified by direct measurements of CT inside the mesocosms. The earliest reference points for CT budgets could be measured after equilibration between the water column above and the dead volume below the sediment traps was achieved. While CT values were found to be stable in the non-manipulated control and some low CO₂ mesocosms earlier, it took until t8 for the high CO₂ mesocosms to deliver CT budgets sufficiently equilibrated with the dead volume to perform mass balance calculations.”

3 I strongly oppose the way of introducing ‘Pool X’, where the authors aim to assign the measured changes in inorganic C, N and P that can not be accounted for by the combined changes in pools of dissolved and particulate organics, cumulative gas exchange and sedimentation. However, dissolved organic C and N measurements as well as particulate organic P were excluded from the corresponding mass balance calculations because “measurement uncertainties of these parameters were larger than the size of Pool X and would therefore compromise mass balance calculations.” From statistical (quantitative) point of view this approach is unacceptable. For example, the authors justify in Discussion the exclusion of direct DOC measurements from respective Pool X estimates by contamination of DOC samples. However, temporal development of DOC observations (given in Schulz et al, Fig. 8D, Biogeosciences Discuss., 9, 12543–12592, 2012) does not support the contamination argument.

The statement that variables were excluded because they would compromise calculations was misleading and will be removed. They were excluded to estimate their possible development by mass balance.

It can be stated that especially DOC and DON are variables that are hard to quantify as compared to CT, DIN or particulate carbon for example. Also the HTC method used to measure DOC has a low nominal uncertainty of $\sim \pm 0.5 \mu\text{mol kg}^{-1}$ (Qian and Mopper, 1996). In our dataset it was on average $1.5 \mu\text{mol kg}^{-1}$ but more than $\pm 10 \mu\text{mol kg}^{-1}$ variability between samples was found. Qualitative statistical analyses of DOC measurements as the ones presented by Engel et al. (2012), and Schulz et al. (2012), resulted in similar trends but had a comparatively low quantitative validity (please compare scales). The contamination problem is discussed by Engel et al. (2012) on page 10298. The DOC dataset in BGD Schulz et al. (2012) was a preliminary version with only half of the measurements included; therefore it will be updated to be coherent with data discussed herein. DON measurements had obvious problems with measured reference material as indicated by strong parallel concentration shifts in all samples (see supplementary graphs (scales!). The source of POP variability could not be identified

4 The authors state in Abstract that “CO₂ treatments induced a shift away from diatoms towards smaller phytoplankton and enhanced cycling of dissolved organics was pushing the system towards a retention type food chain with overall negative effects on export potential.”

However, virtually no diatoms were found in mesocosms, until the major part of nutrient additions were depleted near the end of the experiment, so diatoms could not be outcompeted by smaller algae. Moreover, the phytoplankton succession in the mesocosms seemed to be mainly governed by the combination of nutrient availability and cascading grazing effects, which were then mostly positively modified (but not controlled or induced) by elevated pCO₂.

We agree with the reviewer's opinion that outcompetition was not the right term and diatoms were never really abundant in the water column. We will re-phrase this section in the abstract.

Microscopic inspection of the sediment samples as well as Si:C ratios and pigment data (not shown) indicate that diatoms dominated the sediment material from t24 onwards. This is also supported by PLFA results by de Kluijver et al. (2012). There is indication that diatoms (probably *Fragilariopsis*) were abundant and contributed considerably to the biomass growing on the walls. The low abundance of diatoms in the water samples compared to sediments might be due to their faster sinking rates. We will discuss this aspect in the revised version of the manuscript.

Literature

Bellerby, R. G., Silyakova, A., Nondal, G., Slagstad, D., Czerny, J., De Lange, T., and Ludwig, A.: Marine carbonate system evolution during the EPOCA Arctic pelagic ecosystem experiment in the context of simulated future Arctic ocean acidification, *Biogeosci. Discuss.*, this volume, 2012.

Bopp, L., Monfray, P., Aumont, O., Dufresne, J.-L., Le Treut, H., Madec, G., Terray, L. and Orr, J. C. (2001) Potential impact of climate change on marine export production. *Global Biogeochemical Cycles*, 15, (1), 81-99.

de Kluijver, A., Soetaert, K., Czerny, J., Schulz, K. G., Riebesell, U., and Middelburg, J. J.: A ¹³C labelling study of carbon fluxes in Arctic plankton communities under elevated CO₂ levels *Biogeosci. Discuss.*, this volume, 2012.

Engel, A., Borchard, C., Piontek, J., Riebesell, U., Schulz, K. G., and Bellerby, R. G.: CO₂ increases ¹⁴C-primary production in an arctic plankton community, *Biogeosci. Discuss.*, this volume, 2012.

Hegseth, E. s.: Sub-ice algal assemblages of the Barents Sea: Species composition, chemical composition, and growth rates, *Polar Biology*, 12, 485-496, 10.1007/bf00238187, 1992.

Motegi, C., Tanaka, T., Gattuso, J. P., and Weinbauer, M. G.: Effect of CO₂ enrichment on bacterial production and respiration and on bacterial carbon metabolism in Arctic waters, *Biogeosci. Discuss.*, this volume, 2012.

Niehoff, B., Knüppel, N., Daase, M., Czerny, J., and Boxhammer, T.: Mesozooplankton community development at elevated CO₂ concentrations: Results from a mesocosm experiment in a high Arctic fjord, *Biogeosci. Discuss.*, this volume, 2012.

Riebesell, U., Czerny, J., von Bröckel, K., Boxhammer, T., Büdenbender, J., Deckelnick, M., Fischer, M., Hoffmann, D., Krug, S. A., Lenz, U., A., L., Mucbe, R., and K.G., S.: A mobile sea-going mesocosm system: New opportunities for ocean change research, *Biogeosci. Discuss.*, this volume, 2012.

Qian, J., and Mopper, K.: Automated High-Performance, High-Temperature Combustion Total Organic Carbon Analyzer, *Analytical Chemistry*, 68, 3090-3097, 10.1021/ac960370z, 1996.

Ridgwell, A.: Evolution of the ocean's "biological pump", *Proceedings of the National Academy of Sciences*, 108, 16485-16486, 10.1073/pnas.1112236108,

Schmittner, A., Oschlies, A., Matthews, H. D., and Galbraith, E. D.: Future changes in climate, ocean circulation, ecosystems, and biogeochemical cycling simulated for a business-as-usual CO₂ emission scenario until year 4000 AD, *Global Biogeochem. Cycles*, 22, GB1013, 10.1029/2007gb002953, 2008.

Schulz, K. G., Bellerby, R. G. J., Brussaard, C., Büdenbender, J., Czerny, J., Fischer, M., Koch-Klavnsen, S., Krug, S. A., Lischka, S., Ludwig, A., Meyerhöfer, M., Nondal, G., Silyakova, A., Stuhr, A., and Riebesell, U.: Temporal biomass dynamics of an Arctic plankton bloom in response to increasing levels of atmospheric carbon dioxide, *Biogeosci. Discuss.*, this volume, 2012.

Silyakova, A., Bellerby, R. G. J., Nondal, G., Czerny, J., Schulz, K. G., Engel, A., Tanaka, T., De Lange, T., and Riebesell, U.: Net community production and stoichiometry of nutrient consumption in a pelagic ecosystem of a northern high latitude fjord: mesocosm CO₂ perturbation study, *Biogeosci. Discuss.*, this volume, 2012.

Tanaka, T., Alliouane, S., Schulz, K. G., Czerny, J., and Gattuso, J.-P.: Metabolic balance of a plankton community in Arctic coastal waters in response to increased pCO₂, *Biogeosci.*, 2012.