Authors' responses to reviewers' comments (bg-2016-42)

Biogeosciences Manuscript #: bg-2016-42

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Title: The silica-carbon biogeochemical cycle in the Bohai Sea and its responses to the changing terrestrial nutrient loading

Dear Reviewer,

Thank you very much for your attention and the useful comments and suggestions for improvement on our paper BG-2016-42. Based on the comments, we have made extensive modifications (point by point) of the original manuscript. We attach the revised manuscript with changes marked as PDF, as well as the document with our responses to your comments.

Kind regards,

Jun Liu, Lex Bouwman, Jiaye Zang, Chenying Zhao, Xiaochen Liu, Zhigang Yu, Ran Xiangbin

General comments:

The manuscript by Jun Liu and coworkers presents results from field measurements of Si and C in the water column and sediment of Bohai Sea. They estimated the Si and C budgets and assessed the impact of change in terrestrial loadings on primary production in the Bohai Sea.

Quantifying Si and C budgets and fluxes in the Bohai Sea, which is important for understanding the anthropogenic impacts on coastal biogeochemical cycles and ecosystem function, is a significant contribution to the field. My main concern is that the description and explanation of the Si and OC distributions are not well structured and supported. In addition, they are old news already. Therefore, there is some room for improvement in the focus of the manuscript before it is suitable for publication.

Response: Thanks for the referee's constructive and kind suggestions. We have made extensive modification on the manuscript based on the referee's suggestion and our new ideas. The description and explanation of the RSi and OC distributions have been revised as well as shortening them. Also, the processes of Si-C budget have been thorough discussed and consistent with the Si and OC distributions. Each flux had been recalculated and double checked based on our study and other latest studies (see below).

1. Manuscript focus

The distribution of silicon along with other nutrients in the Bohai Sea has been systematically studied by Li et al. (2003), Zhang et al. (2004) and Liu et al. (2011) and so on. It has been demonstrated that the Si distribution is highly influenced by diffusion from sediment and major river input (Li et al., 2003; Zhang et al., 2004). In general, the Si concentration is higher in the coastal areas than in the central Bohai Sea and higher in near bottom waters than in surface waters (Zhang et al., 2004; Liu et al., 2011; also shown in this study). Nutrients including Si are vertically well mixed in the coastal areas, while stratification takes place in the upper water of the central Bohai Sea especially in summer (Li et al., 2003; Liu et al., 2011). The Si concentration in the Bohai Sea is usually higher in autumn than in spring (Zhang et al., 2004; also shown in this study). Unfortunately, I couldn't recognize new added value in this manuscript to the current understanding of the nutrients distribution in the Bohai Sea. I would suggest that the authors rather focus on the nutrient budgets estimate and impact study, and eliminate the nutrients distribution

part. Unless the authors make a clear point on what is novel in their findings.

Response: Thanks for the referee's constructive suggestions. The manuscript was revised according to the comments. We condensed the section of 3.1, 3.2 and 4.1 and 4.2, and focused more on discussing the Si-C budget and impact processes in the revised manuscript. The reasons why silica and OC distributions should be reserved but more concise are: 1) it is the foundation to establish the budge model and some important hypotheses are originated from the C and Si distributions and C/Si ratios; 2) the budget can be tested and verified with the distribution. In order to concentrate our scientific aims, we move the figures of Figure 3 (Figure S2 in the revised manuscript) and Figure 4 (Figure S3 in the revised manuscript) to the *SUPPLEMENT INFORMATION*.

Additional, we recalculated the budget, some processes, like the benthic and exchange fluxes, were estimated on the basis of our study and available published results, which can improve our work greatly (see below).

2. Structure

The authors may want to consider reorganizing the "2.1 sampling and analytical methods" section by subsections (e.g. 2.1.1 sampling, 2.1.2 pretreatment of samples and 2.1.3 laboratory analysis) or grouping the related paragraphs together. L128-131 should be merged into the first paragraph of section 2.1. This would help readers to find information more efficiently.

The same for Section 4.3 that some paragraphs can be grouped together, otherwise information gets fragmented.

Response: We reorganized the section of 2.1 following the referee suggestion. L128-131 was also reorganized. Thus, the section of 2.1 has been divided into two subsections: 2.1.1 Sample collection and pretreatment, 2.1.2 laboratory analyses. Meanwhile, some relevant paragraphs in Section 4.3 have also been grouped together.

Specific comments:

(1) L25: They are old news already. See General comments 1.

Response: This suggestion was followed. It has been removed in the revised manuscript.

(2) L27: These are interesting results! State more numbers like 49% DSi input is from benthic diffusion and so on.

Response: This suggestion was followed. We state more clear numbers in the abstract, e.g. "Benthic diffusion contributes 49% of total reactive Si input to the Bohai Sea, and smaller amounts come from groundwater (26%), riverine input (17%|), surface runoff (7%) and atmospheric deposition (< 1%)".

(3) L40-41: Those estimates are likely upper limits of diatom contributions to the primary productivity, globally, as stated by Nelson et al. (1995). I would suggest to use "up to" instead of "more than".

Response: This suggestion was followed.

(4) L51-53: This statement cannot be supported by the two references listed here, especially the one by Conley (1997). Please provide appropriate references.

Response: This suggestion was followed. A more appropriate reference by Humborg et al. (2000) and Bernard et al. (2010) has been cited in this sentence.

(5) L56-58: Do you mean it will influence the function of coastal ecosystem as a biological pump?

Response: Yes, the Si cycle and C cycle in marine system are connected with each other mainly by primary production.

(6) L63: "have induced *significant* changes"

Response: This suggestion was followed.

(7) L69: "phytoplankton composition in the Bohai Sea"

Response: This suggestion was followed. The influenced area was limited in the Bohai Sea in the revised manuscript.

(8) L73: "... may have enhanced primary production by increasing export of ~~water~~ nutrients and

sediment to the Bohai Sea."

Response: Thanks for the referee's detailed guidance. This suggestion was followed.

(9) L75: "than in ~~other~~ open seas"

Response: Thanks for the referee's detailed guidance. This suggestion was followed.

(10) Section 2.1: How many replicants of water samples and sediment samples did you measure? Please state that in this section.

Response: This suggestion was followed. The data quality was monitored by inter-calibrations with the national/international standards, and samples were repeatedly analyzed for nutrients at the micromolar level. And the uncertainties for data were added as follows:

The BSi content in sediment was measured using the alkaline extraction method (1% Na_2CO_3, 85 ^oC, sequential extracted for over a total period of 8 h, during which the extract is sampled every 1 h and analyzed eight times) (DeMaster, 1981), with a relative standard deviation < 3% (n=5) in our study. DSi was calibrated against nutrient standard sample (from Second institute of Oceanography, State Oceanographic Administration, China), while TOC was calibrated against Reference Standard Materials (EuroVector, Code E11001).

By the way, we have ever got four interlaboratory samples from Dr. Daniel J Conley in 2004 when we started the work of BSi, please check in the results from the website (http://www.hyxb.org.cn/aos/ch/reader/create_pdf.aspx?file_no=20070419&year_id=2007&quarter_id=4&falg=1) for detail. Our results fell in the range of biogenic silica concentrations measured by 30 laboratories and were also very close to the mean values (Conley, 1998). Again, the measurements in the Bohai and Yellow Seas are consistent with other studies in the continental shelves of Eastern China (Liu et al., 2005; 2008a).

(11) L165-166: "The hydrography …" This sentence belongs to introduction.

Response: Yes, this sentence belongs to introduction, we present it here for a better understand of the water budget.

(12) L172: I would use “precipitation” instead of “atmospheric deposition” if you only refer to water.

Response: This suggestion was followed.

(13) L174-180: The water fluxes in the text have different precision as in Fig. 2. This results in a 10% difference in the estimate of surface runoff of 1 or 1.1 km^3 yr^{-1}. Please use the same precision in the text as in Fig. 2.

Response: This suggestion was followed.

(14) L209-210: Equation 2 and 3: Please give information on how the regression equations were derived, by showing figures (in the supplement) or citing references if they were taken from publications. Were the data (BSi, POC and SPM) sampled from the Yellow River? If so, please state the assumption that the relation between BSi/POC and SPM in the other rivers is similar as that in the Yellow River.

Response: These suggestions were followed. The information on how the regression equations were derived is provided in the *SUPPLEMENT INFORMATION* (Figure S1). The data (BSi, POC and SPM) were sampled from the Yellow River.

It should be noted that the Yellow River is the largest river in North China and most of the material sedimented in the North China Plain, where many of the rivers listed in Table 1 flow through, stems from the Yellow River. Hence, assuming that BSi and POC in river water generally have the same origin (Ran et al., 2015), we used the sediment content and BSi and POC concentrations for the Yellow River to develop regression equations that can be used to calculate BSi and POC concentrations for rivers with missing data:

(15) L221: The area of Bohai Sea should be introduced along with the average depth already in the introduction.

Response: This suggestion was followed. The area and average depth of Bohai Sea are introduced in the section 1. The new added relative text is as follows:

The Bohai Sea is a semi-enclosed, shallow shelf water body of the northwestern Pacific Ocean, with a surface area of 77,300 km^2 and an average depth of 18 m.

(16) L225-226: Is the DOC concentration in the coastal rainwater, reported by Willey et al. (2000), a global (coastal region) average value or from a certain coastal region? Is there a potential difference than the value in Bohai Sea?

Response: Yes, the DOC concentration in the original manuscript is in the coastal rainwater reported by Willey et al. (2000), which is an average value (coastal region) from Puerto Rico bulk, Costa Rica and Wilmington NC. Since there is rarely information about DOC of rain water in the Bohai Sea, and the DOC in rain water originates from multiple sources, with a significant portion coming from fossil fuels and biogenic fractions containing both marine and terrestrial organic carbon. Thus, there would be a potential difference of value between marine and continental rain, and this uncertainty has also been discussed in the section of 4.4 "The uncertainty of the budget". For a better estimation of DOC in atmospheric deposition, the DOC concentration in the coastal rainwater of Bohai sea is assumed equal to the average value (monthly data of 2014) in snow and rainfalls from Yantai (Xu et al., 2016) in the revised manuscript, which is connected to the southern Bohai Sea.

(17) L239: Equation 4: $\partial C/\partial C$ —> $\partial C/\partial z$

Response: We have corrected this equation in the revised manuscript.

(18) L254-255: "Sedimentation rate of BSi and OC … from the accumulation rate of total mass and the surface area." I don't understand what is "the surface area".

Response: Sorry, it was not clear explained here. "the surface area" represents the area of seafloor of the Bohai Sea. Since the surface water area of the Bohai Sea is 77300 km^2 and the average depth is 18 m; meanwhile, the seafloor of the Bohai Sea is flat; thus the surface area of seafloor in the Bohai Sea can be estimated as 77300 km^2.

(19) L290-291: First, they are not similar to me at all. Second, do you mean the DSi

concentrations are fairly low in the northwestern part of the Bohai Sea in the bottom water? Because it doesn't apply to the surface water.

Response: Yes, it was not clear described here. This paragraph has been rewritten as follows:

The average DSi concentration in fall exceeds that in spring (Table 2). In spring, DSi concentrations are fairly low in the northeast part of the Bohai Sea, but high in the Bohai Strait and Yellow River estuary. In autumn, DSi concentration in coastal areas of the Bohai Sea, especially for the Laizhou Bay and northern part of Bohai Sea, exceeds that in other areas (Figure S2).

(20) L302-303: Again, the distribution of BSi in surface water is not similar to that in bottom water to me.

Response: This suggestion was followed. This paragraph has been shorten and rewritten as follows:

The BSi concentration in fall exceeds that in spring by a factor of four (Table 2). In spring, surface water BSi concentrations are fairly high near the Bohai Bay, Laizhou Bay and Yellow River estuary, and lower in the central part of the Bohai Sea. Relatively high BSi concentrations occur in the central area of the Bohai Sea in the bottom water. In autumn, the BSi concentrations in the Yellow River estuary and other nearshore areas exceed those in the central zone (Figure S2).

(21) L306: I guess you meant lower, not higher. And please state here and where else in this section that it's the average concentration of all stations.

Response: This suggestion was followed. The comparison of RSi and OC values between spring and fall has been definitely stated in the revised manuscript.

(22) L317-318: The average BSi content … a large variability in both BSi and TOC among stations.

Response: Thanks for the reminding. This suggestion was followed.

(23) L328: The estimated deposition flux of what?

Response: Thanks for the reminding. It has been corrected as follows:

The estimated deposition flux of RSi in the Bohai Sea is 0.2 Gmol yr^{-1}, primarily (94%) from wet deposition.

(24) L334: "are similar" this statement is not appropriate and not necessary, please rephrase it.

Response: This suggestion was followed. It has been corrected as follows:

The inputs of RSi and OC from the Yellow Sea into the Bohai Sea are 2.8 Gmol yr^{-1} and 100 Gmol yr^{-1}, respectively, while the output fluxes of RSi (3.1 Gmol yr^{-1}) and OC (132 Gmol yr^{-1}) from the Bohai Sea to the Yellow Sea. Thus, net outputs of RSi and OC from Bohai Sea are 0.3 Gmol yr^{-1} and 32 Gmol yr^{-1}, respectively.

(25) L350: "The estimated surface runoff fluxes are 2.1 Gmol yr^{-1} for RSi and 6.4 Gmol yr^{-1} for OC according to Eq. (1) ."

Response: Thanks for the reminding. It has been corrected in the revised manuscript.

(26) Section 4.1: See General Comment 1.

Response: This suggestion was followed and the manuscript was revised according to the comments. We focused more on discussing the Si-C budget and impact processes in the revised manuscript.

(27) Section 4.2: The number of sampled stations in fall is 2.5 times as many as in spring. This is just a question to the authors whether the different sample sizes of two seasons have been taken into account in the analysis and the conclusion drawn from it.

Response: Thanks for the reminding. It has been corrected and stated that the comparison of seasonal variation of RSi and OC is based on the stations of the same position in spring and fall.

(28) L415-417 and L422-423: The dominant output should be sedimentation (99%), not sedimentation (99%) and export (1%). The same for L422-423.

Response: Thanks for the reminding. It has been corrected in this section and other parts in the revised manuscript.

(29) L435-436: The work by Treguer and De La Rocha (2013) can also be cited here.

Response: This suggestion was followed.

(30) L439-442: Whether the benthic flux of DSi plays a role in maintaining the primary production or not depends on the euphotic layer depth and vertical mixing level. Seasonal variation may exist also. Please elaborate on this point.

Response: This suggestion was followed. We added the relative text in the revised manuscript. The new text is as follows:

The primary production also depends on the euphotic layer depth, vertical mixing and presence of nutrients in the water column, which cause seasonal variation of primary production and RSi and OC in the Bohai Sea.

(31) L445: Before saying "another way", the authors should probably state "your way" first, which means to move the last sentence in front of "Another way...".

Response: Thanks for the reminding. It has been corrected in the revised manuscript.

(32) L452: "The ratio of BSi sedimentation flux to BSi production (3.3%) exceeds the average value for the world ocean (2.6%)."

Response: It has been corrected in the revised manuscript.

(33) L454: "~~similar to~~" → within the range of

Response: It has been corrected in the revised manuscript.

(34) L464: "Relative … and riverine Si input of Bohai Sea are similar to …." Please cite numbers here to demonstrate how similar they are.

Response: This suggestion was followed. We added the relative numbers in the revised

manuscript, and this paragraph has been rewritten as follows:

Our estimates for submarine groundwater and river Si inputs agree with previous study showing that Si inputs from submarine groundwater are 1–2 times higher than those associated with river discharge into the Bohai Sea (Liu et al., 2011). This agrees with data for the Yellow Sea where estimated submarine groundwater DSi and riverine RSi inputs are respectively 4–24 Gmol yr^{-1} and ~23 Gmol yr^{-1} (Kim et al., 2005) and with data for the Mediterranean Sea where estimated submarine groundwater and river inputs are 110 Gmol yr^{-1} and 30–40 Gmol yr^{-1}, respectively (Rodellas et al., 2015). The DOC input from submarine groundwater is also comparable to riverine input, indicating that submarine groundwater is also an important source of DOC in the Bohai Sea.

(35) L468: What are the "above studies"? Do you mean "(Liu et al., 2011)"? Please clarify here. In addition, this is only one study, please also modify the phrase "Previous studies" at the beginning of the paragraph accordingly.

Response: This suggestion was followed. This paragraph has been rewritten in the revised manuscript; please see comment No. 34 for detail.

(36) L480-482: This sentence belongs to the paragraph above.

Response: This suggestion was followed. It has been grouped to the paragraph above.

(37) L482: "silica" refers usually to the diatom shell, which is BSi as in the manuscript. Maybe you meant DSi or RSi here?

Response: Thanks for the reminding. It has been changed to "Si". "Available silica" represents reactive silicon.

(38) L495: It seems to me a conflict between "annual" and "in July".

Response: Thanks for the reminding. The word "annual" has been changed to "monthly".

(39) L496: "this year" can be misleading as 2016

Response: Thanks for the reminding. This sentence has been changed as follows:

The monthly DSi flux of Yellow River in July increased 5–10 fold and RSi flux by a factor of 3 since 2002; since the sediment load and water discharge regulation in spring has led to peak events (Figure 4) (Gong et al., 2015; Liu, 2015).

(40) L501: My suggestion: "This is confirmed by DOC increase in the Bohai Sea from 2.1 mg l^{-1} before (Zhang et al., 2006) to 2.6 …. , "

Response: This suggestion was followed. It has been corrected in the revised manuscript.

(41) L501-506: How much has the riverine input of OC changed? Can the increase of OC concentration in the Bohai Sea be attributed to increase of riverine input of OC?

Response: The riverine input of DOC from 2000 to 2014 has increased by ~400%, and there is a sharp increase after the water and sediment regulation of the Yellow River since 2002 (Liu et al., 2015). However, there is no long-term data of POC in the Yellow River, but the sediment discharge has increased significantly since 2002. Thus the POC flux would increase also. The increase of OC concentration in the Bohai Sea should attributed to the increase of riverine input of both OC and RSi, which need more field measurements in the future.

(42) L530-532: This sentence belongs to Introduction.

Response: This suggestion was followed. It has been removed in the revised manuscript.

(43) L532-534: The authors should indicate here that the 10% increase in PP comes from a calculation based on the relation between RSi and PP in year 2000-2008.

Response: This suggestion was followed. We added the relative text in the revised manuscript. The new text is as follows:

Based on the relation between RSi and primary production in year 2000–2008, primary production in the Bohai Sea has increased by 10% since 2002, as a result of the increasing riverine RSi input from the Yellow River due to water-sediment regulation.

(44) Section 5: It would be good to mention what the major uncertainty is in the budget estimate and to comment on the potential bias on the estimate due to the assumption of steady state at the end of this section.

Response: This suggestion was followed. We added the discussion of "4.4 *The uncertainty of the budget*" in the section 4.4 (Page 18–19) to illustrate the major uncertainty in the budget of RSi and OC in the Bohai Sea. The new added text is as follows:

4.4 The uncertainty of the Si and OC budgets

Table 1 presents the uncertainties in each flux estimate, in most cases based on the relative standard deviation in the measurement data used. By adding the absolute uncertainties we obtain an estimate of the uncertainty range of all incoming and outgoing fluxes. The results of the model simulation show that the benthic diffusive flux is the most important flux in the Si budget of the Bohai Sea with the largest absolute uncertainty (8.5 Gmol yr^{-1}). With the uncertainties associated with the river input (0.9 Gmol yr^{-1}), groundwater (1.8 Gmol yr^{-1}) and assuming that the flux from surface runoff and small rivers not included in F_R is 50% and that of atmospheric deposition 100%, the total uncertainty in the total of incoming fluxes is around 12 Gmol yr^{-1} (50% of total incoming flux). The uncertainty in the exchange fluxes between Bohai Sea and Yellow Sea is 50% in both the outgoing and the incoming flux, but since the RSi concentration in both exchanges is similar; the net flux has a minor contribution to the overall uncertainty. The uncertainty of the sedimentation flux is 40% (Table 1).

The water exchange between Bohai and Yellow Sea is the most important process influencing on the OC budget. However, the concentrations of OC in southern Bohai Sea and northern Yellow Sea are similar. The uncertainty in the net outgoing flux F_E can be assumed to be 30%, similar to that of each of the exchange fluxes. The uncertainty of the benthic diffusive OC flux is 5 Gmol yr^{-1} (55%). The estimated uncertainty caused by the spatial and temporal variability of OC in rivers draining into the Bohai Sea is 9 Gmol yr^{-1} (25%). With uncertainties associated with atmospheric deposition (6.1 Gmol yr^{-1}) and submarine groundwater discharge (2 Gmol yr^{-1}), and assuming 50% uncertainty in the flux from surface runoff and small rivers not included in F_R, the

total uncertainty in incoming fluxes is 34 Gmol yr^{-1} or 50% of the total incoming flux of 92 Gmol yr^{-1}.

Comments on figures and tables:

(45) Table 1: Main fluxes of reactive silica silicon and …

I found it difficult to find information in this table. The most interesting numbers might be the Si and C fluxes from different sources, which should be kept in the last two columns for each source, as you did for F_R. It would also be nice to have a sum of all rivers for Si and C fluxes as an additional row below "Daling River". Wherever possible, the same terms should be kept in the same column, e.g. for water exchange (F_E), just let the third column empty and move the rest six columns to the right.

Response: These suggestions were followed. Table 1 has been improved according to the comments in the revised manuscript.

(46) Table 2: Reactive ~~silica~~ silicon ...

I would suggest to move "Spring" and "Fall" in the same row as "Surface" to avoid confusion, or to draw a horizontal line to separate two seasons. The same suggestion goes to Table 3 for "Surface sediment" and "Core sediment".

Response: These suggestions were followed. "silica" has been change to "silicon", and Table 2 and Table 3 have been improved in the revised manuscript.

(47) Figure 1: Please state the core sediment sampling stations in the legend.

Response: This suggestion was followed. Sampling stations in the legend have been stated in Figure 1.

(48) Figure 3 and 4: They don't provide new information or systematic understanding, in my opinion. Therefore they can be removed. Also see the General Comment 1.

Response: Thanks for the reviewer's kind suggestion. We move these figures to the "*SUPPLEMENTARY INFORMATION*" as Figure S2 and S3, respectively. However, Figure S2 (points to Figure 3 in the previous version) and Figure S3 (points to Figure 4 in the previous

version) can give visualized distribution of RSi and OC in the water column and sediment, and these are the basis of Si and C budget. In addition, the figures also provide information to support the accuracy of our budget.

(49) Figure 5: reactive silica silicon …

This is a very nice figure! Maybe also state the unit in the legend?

Response: Thanks for the reviewer's encourages. This figure (Figure 3 in the revised manuscript) has been improved for a better understanding.

Reference

Li et al., 2003, Distributions of inorganic nutrients in the bohai sea of china, Journal of Ocean University of Qingdao , Volume 2, Issue 1, pp 112-116.

Response: This reference has been referred to in section 4.5 in the first paragraph to evaluate the nutrient change in the Bohai Sea.

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