

We thank referee n°1 for their constructive comments. We have replied to these comments below:

*Modifications introduced in main text and Supplemental Information after posting comments on the web site are in italics.*

**1) Steady-state assumption.** The authors make value judgements on the data used to calculate fluxes sometimes discarding studies, sometimes averaging a wide range of numbers, although this reviewer feels comfortable with their calculations. However, after a large number of assumptions the authors arrive at a “balanced budget” interpreted that the global Si cycle in the oceans is at a steady-state. I am not sure the success of achieving a balanced budget warrants the conclusion that the budget is in steady state especially because there is a large range of input and output fluxes, many of them uncertain, contributing to this revised budget. Are there fluxes that are underestimated (a la Jeandel) or estimates of fluxes that are over-stated (sponge BSi deposition) that could put the budget far away from an apparent balance?

Answer : We believe there is a misunderstanding regarding our approach to build a budget for the marine Si cycle.

This review paper aims to show the best knowledge concerning the processes that controlled the input fluxes of dSi and the output fluxes of Si to and from the ocean.

What we show is that a best estimate of the total average of the input fluxes (14.8 Tmol-Si yr-1) and the total average of the output fluxes (15.6 Tmol-Si yr-1) are unbalanced. Therefore, we disagree with the reviewer that « the authors arrived at a balance budget ».

However, given a possible underestimate of the *input fluxes* (see sections 5.2.3 and 6.), we do not exclude that the two fluxes balance each other, and thus that the Si cycle in the modern ocean could be at steady state (Figure 1). This is clearly stated:

- in the abstract («we address the steady state hypothesis »),
- in section 5.2.3 (« A possible steady-state scenario »), and
- in the « Conclusions/recommendations » section 6 (« The main question that still needs to be addressed is whether the contemporary marine Si cycle is at steady state,... »).

**2) Specific unresolved questions (starting at line 537).** The manuscript presents two case studies (Chinese Seas and The North-East Pacific dSi Anomaly) that add little to the main theme of the manuscript. The conclusion of this section starting at line 581 “a process that requires further studies” could be used on many of the processes contributing to fluxes. I do not know if these case studies are important to have as part of this manuscript.

Answer :

Reviewer n°1 asks if these two cases studies add value to the manuscript. A principal aim of this review paper is to provide guidelines for future research programs.

We chose to highlight these two case studies in the Pacific Ocean, which is considered a « silica ocean » (Honjo et al. 2008 ), since more knowledge is required for the Si cycle in this ocean.

As explained in section 5.3.1, the Chinese marginal seas are unique over the world ocean marginal seas, particularly because of the massive entrainment of siliceous soils through the hydrographic network of large rivers, making it difficult to quantify the bSi content in this system using a classic alkaline leach's method. As mentioned in section 6, we recommend the Chinese marginal seas to be one of the major test sites for the international comparative analytical exercise to be organised in the future. Thus, a better understanding of the processes that control the Si cycle in the Chinese marginal seas is a priority for a better understanding for the Si cycle of the world ocean.

As explained in section 5.3., the North Pacific dSi-very rich plume is unique in the world ocean. Among the hypotheses that explain the formation and maintainance of this plume (which required a flux of 1-2 Tmol-Si yr-1) is remobilization of relatively old bSi that accumulated over a long time interval, by dissolution in a hydrothermally active environment. If this hypothesis is correct, the total average net dSi inputs to the ocean would be between 15.8 and 16.8 Tmol-Si yr-1, i.e. in balance or in small excess to the present extimate of the total Si output flux.

Process studies in these two sites of the Pacific ocean are a priority for future research on the Si cycle, and if the Editor agrees, we prefer to maintain this section.

**3) A discussion of the uncertainties and how changes in the ocean Si cycle would be affected by global change (Section 5.4) is important to include, however, this section needs to be more coherent and better presented.**

**A carefully revised manuscript would greatly increase our knowledge of the global Si cycle and would provide new hypothesis and assumptions to be examined by the community.**

Answer :

For the sake of simplicity, this section is now limited to "the impacts of climate change on the Si cycle" (section 5.4):

- 5.4.1 (impacts on riverine inputs of dSi and aSi),
- 5.4.2 (Abondance of marine pelagic and benthic silicifiers), and
- 5.4.3 (Predictions for the ocean phytoplankton production and bSi production).

The impacts of anthropogenic activities not associated with climate change are now discussed in a different section 5.5. ("Other anthropogenic impacts").

**Minor comments:**

**-Line 157 "this represents a realistic upper limit" – What is the justification for this statement?**

Answer : Cho et al (2018)'s used a  $^{228}\text{Ra}$  inverse model and groundwater dSi/ $^{228}\text{Ra}$  ratios to estimate the total SGD dSi flux to the ocean. Because this total flux comprises the contributions of both the terrestrial (net) and the marine (net + recycled) components it represents the upper limit of the net SGD dSi flux, if we assume that the marine component would be a net flux, which obviously is not the case.

The text has been changed as follows :

«... this represents an upper limit... ».

**-Line 168 “Only one value currently exists” – See Hirst et al. published on 26 June 2020 and Hatton et al. published on 14 July 2020 in Frontiers that have new numbers for Antarctica.**

Answer : These studies are now acknowledged.

**-Line 232 and 233 - Why are you using units of “g yr-1” instead of “Tmol yr-1” as in the rest of the manuscript?**

Answer : we used Tmol yr-1 when we refer to silica fluxes, but used g yr-1 when we refer to water fluxes (hydrothermal fluid or river water fluxes).

**-Line 235 Changed “determined” to “calculated”**

Answer : corrected.

**-Line 337 “canonical” is used several times in the manuscript. I have looked up the word and it has many meanings depending on the field of study. I looked in the original citation and “canonical” is not there either. Can you please use a less ambiguous word?**

Answer : Corrected

The sentence “The “canonical” value for global gross marine bSi pelagic production is 240 ( $\pm 40$ ) Tmol-Si yr-1 (Nelson et al., 1995)” has been rewritten to read

“The last evaluation of global marine silica production was by (Nelson et al., 1995) who estimated global gross marine bSi pelagic production to be 240 ( $\pm 40$ ) Tmol-Si yr-1”

**-Line 455 Change “do” to “does”. Fix the rest of the sentence “being little reliable”**

Answer : We have reworded this sentence. Thank you!

“...If the bSi production that is being accumulated as standing stock in the living sponge populations annually is assumed to become constant in a long term equilibrium state, the global annual deposition rate of sponge bSi can be considered as a reliable estimate of the minimum value that the annual bSi production by the sponges can reach in the global ocean. The large associated SD value does not derive from the approach being unreliable but from the spatial distribution of the sponges

on the marine bottom being extremely heterogeneous, with some ocean areas being very rich in sponges and sponge bSi in sediments at different spatial scales while other areas are completely deprived from these organisms....

**-Lines 504 (Section 5.2.2) I do not really understand what you are trying to say here, especially the last sentence. You state that climate change or anthropogenic impacts affect dSi (Bernard et al. 2010, 2011) leading to an imbalance. Do you mean there will be no changes in ocean production of siliceous organisms if the Si balance in the oceans change? If so, is that only speculation or is there evidence?**

Answer : (cf. lines 485-486) over a given time scale, an elemental cycle is at steady state if the outputs balance the inputs, and the mean concentration of the dissolved element remains constant. The output flux depends on the export flux of biogenic silica which itself depends on the gross production of biogenic silica.

The text has been corrected as follows :

“In the modern ocean, as shown above, the main control over silica burial and authigenic formation rate is the bSi production rate of (pelagic + benthic) silicifiers. The gross production of bSi due to diatoms depends on the dSi availability in the surface layer (Figure 1), which is controlled external inputs, inputs from below, and Si recycling within the surface layer. Actually, this production is not Si-limited or not severely limited in several zones of the world ocean, which include the coastal zones, and the HNLC zones (Tréguer & De La Rocha, 2013). Thus, on short timescales, there are no strong negative feedbacks, between supply rates and production or burial rates, which would necessarily keep the marine Si cycle in balance. For this reason, impacts of climatic changes or anthropogenic that, at short timescales affect dSi inputs to the ocean by rivers (Bernard et al., 2010, 2011) and/or other pathways (see section 5.4.1), could lead to an imbalance of Si inputs and outputs in the modern ocean.”

**-Line 542 I would add “particulate Si inputs” to needing a better understanding**

Answer : Corrected :

« ... but also to a better understanding of the processes that control the Si cycle, such as SGD, reverse weathering, and particulate Si inputs... »

**-Line 552 “unusual” – Why is it unusual for coastal systems?**

Answer : In coastal systems, according to TDLR 2013 the D : P (dissolution to production) ratio averages 0.51. In coastal Chinese seas this ratio varies between 0.62 and 0.90.

To make it clearer the text has been amended as follows :

« ... Secondly, the bSi production seem to be mostly (62-90%) maintained by a recycling of Si (Li et al., 2019; Liu et al., 2005; Liu et al., 2016; Wu et al., 2017; Wu et al., 2020), which is particularly high for coastal systems (Tréguer & De La Rocha, 2013) ».

**-Line 563 “recommend additional attention” – Why**

-Answer : As explained in section 5.3.1, the Chinese marginal seas are unique over the world ocean marginal seas, particularly because of the massive entrainment of siliceous soils through the hydrographic network of large rivers, making it difficult to quantify the bSi content in this system using a classic alkaline leach's method. As mentioned in section 6, lines 689 and following, we recommend the Chinese marginal seas to be one of the major test sites for the international comparative analytical exercise to be organised in the future. Thus, a better understanding of the processes that control the Si cycle in the Chinese marginal seas is a priority for a better understanding for the Si cycle of the world ocean.

**-Line 580 Need reference to these numbers presented.**

Answer :

we are not sure we understand the question. All the numbers given in section 5.3.2 are taken from the two references already cited in this section (Talley et al. 1992 ; Johnson et al., 2006)

**-Lines 590, 598, 600 Should be “precipitation”**

Answer : Corrected

**-Line 601 Spell out what you mean by “contradictory impacts”**

Answer : let us explain the two contradictory impacts :

There is more precipitation in the tropical convergence zones, which means there is more weathering of soils and more dSi delivery to the ocean. When there is less precipitation in tropical subsidence zones, this means there is less weathering of soils and less dSi delivery to the ocean.

**-Line 616 Why use the word “corroborated”?**

Answer : Modified as follows

... « the melting of Antarctic ice platforms has been noticed to trigger impressive population blooms of highly silicified sponges (Fillinger et al. 2013). ».

**-Line 652 “These uncertainties suggest” – If we can not use models to help us understand how the silica biogeochemical cycle will change in a future ocean, then what do we use? Seems like it was written by someone who does not like models.**

Answer: the second reviewer shared your criticisms and requested a few additional details.

Below is our answer to both reviewers.

Section 5.4.3 has been modified as follows :

“In the 21st century, climate change affects ocean circulation, stratification and upwelling thus affecting the cycles of nutrients (Aumont et al., 2003; Bopp et al., 2005, 2013).

With increase stratification, reduced dSi supply from below (Fig. 1 and 4) leads to less siliceous phytoplankton production in surface compartments of lower latitudes and possibly the North Atlantic (Tréguer et al., 2018).

Based on field studies, the impact of climate change on the phytoplankton production of higher latitudes is highly debated as melting of sea ice decreases light limitation. Regarding the Arctic Sea, increase nutrients (at the least for silicic acid) availability will occur through the Transpolar Drift delivering nutrient rich river- and shelf derived waters as potential sources for primary production, including bSi production (e.g. Charette et al., 2020). Regarding the Southern Ocean, a well known area for low dissolved iron (dFe) concentrations (Tagliabue et al 2016), except in the CCMZ (e.g. Annett et al. 2015 ; Sherrell et al. 2018), bSi production might increase in areas impacted by dFe inputs from icebergs melting or from increased convection that feeds surface waters in dFe from below (Boyd et al., 2016 ; Hutchins & Boyd, 2016 ; Tréguer et al., 2018). Changes in Southern Ocean phytoplankton population due to climate change have been observed already. For example, along the West Antarctic Peninsula, phytoplankton community are quite sensible to the ice coverage, with the haptophyte and cryptophyte community increase while the diatom decrease (Henley et al., 2019). Globally, it is therefore possible that a warmer and acidified ocean alters the pelagic bSi production rates, thus modifying the export production, and the Si burial rates at short time scales.

Although uncertainty is substantial, model studies (Bopp et al., 2005; Dutkiewicz et al., 2019; Laukötter et al, 2015) suggest regional shifting of bSi pelagic production due to climate change. Climate change models suggest a global decrease in diatom biomass and productivity over the course of the 21st century (Bopp et al., 2005, Dutkiewicz et al., 2019, Laukötter et al., 2015), which would lead to a reduction in the pelagic biological flux of silica. Regional responses however differ, with most models suggesting a decrease in diatom productivity in the lower latitudes and many predicting an increase in diatom productivity in the Southern Ocean (Laukötter et al, 2015). Holzer et al. (2019) suggest that changes in supply of dFe will alter bSi production mainly by inducing floristic shifts, not by relieving kinetic limitation. Increased primary productivity come from reduction in sea-ice and the

faster growth rates with warmer waters and longer growing seasons in the high latitudes. However, many models have very simple ecosystems including only diatoms and a small phytoplankton. In these models, increased primary production in the Southern Ocean is mostly from diatoms. Some models with more complex ecosystem (i.e. including additional phytoplankton groups) suggest that increased primary productivity in the future Southern Ocean will be due to other phytoplankton types (e.g. pico-eukaryote) and that diatoms biomass will decrease (Dutkiewicz et al, 2019; also see model PlankTOM5.3 in Laufkötter et al, 2015), except in regions where sea-ice has melted. Differences in the complexity of the ecosystem and parameterizations, in particular in terms of temperature dependences of biological process, between models lead to widely varying predictions (Dutkiewicz et al., 2019; Laufkötter et al., 2015) thus constraining our capacity to predict what will happen with the silica biogeochemical cycle in a future ocean.”

We thank the reviewer n°2 for their constructive comments, which we addressed below:

*Modifications introduced in main text and Supplemental Information after posting comments on the web site are in italics.*

**-L63 “Silicifiers use . . . structure”. I will suggest moving this sentence later. Something like Silicifiers are among the most important aquatic organisms, including microorganisms () and macro-organisms (). They all depend on dSi that they precipitate and form biogenic silica to build their internal () and/or external () structures.**

Answer : The introduction has been modified accordingly :

« ... Silicifiers are among the most important aquatic organisms, including micro-organisms (e.g. diatoms, rhizarians, silicoflagellates, several species of choanoflagellates), and macro-organisms (e.g. siliceous sponges). They use dSi to precipitate biogenic silica (bSi; SiO<sub>2</sub>) as internal (Moriceau et al., 2019) and/or external (Maldonado et al., 2019) structures...”

**-L71: independently?**

Answer : corrected

**-L74 – 86: Here you present the different aspect/section/points of the paper, why not underline the benthic fluxes as they are emerging studies since the last review Treguer & De La Rocha (TDLR) 2013?**

Answer : Actually, we would not qualify « benthic studies » as « emerging studies » because the potential importance of reverse weathering (FRW), and of sponges (FSP) as Si outputs was already mentioned in TDLR 2013. *Hayes et al. recent synthesis article on the burial rates of various elements including Si is now taken into account.* So, in this review article we attract attention on recent studies which show that FB and FRW are much higher and FSP much lower than TDLR 2013's estimates. The total outflux is now 37% higher than TDLR 2013's estimate, which is already highlighted in the abstract.

**-L89: Silicic acid is already defined in the introduction, keep only dSi**

Answer : corrected

**-L91: One standard deviation or two standard deviation? Could you precise to avoid any mistake when people use the numbers in the future.**

Answer : Thank for this comment. The text has been modified accordindly :

« All fluxes are given with one standard deviation ».

**-L93: 60% not 60 %**

Answer : Corrected

**-L106: FA is only defined in the title, might need to define it also in the text.**

Answer : Corrected

**-L108-109: it seems that a verb is missing in the sentence. If not could you rephrase?**

Answer : The text has been modified as follows :

“As shown in Fig. 2, the low-temperature dissolution of siliceous minerals in seawater and from sediments feeds a dSi flux, Fw, through two processes: (1) the dissolution of river-derived lithogenic particles deposited along the continental margins and shelves, and (2) the dissolution of basaltic glass in seawater, processes at work mostly in deep waters. About 15-20 Gt yr-1 river-derived lithogenic particles...”

**-L108-129: This paragraph is a bit confusing. Fw is meant to represent benthic fluxes but a numerous of studies have not been mentioned here. The paragraph seems to focus on a flux representing the dissolution of lithogenic material deposited via river inputs. This input is of importance but do not represent all benthic Si fluxes. This section can be extended and also subdivided with the different type of benthic fluxes like mineral dissolution, benthic fluxes due to early diagenesis of biogenic opal (e.g. Ehlert et al., 2016 Stable silicon isotope signatures of marine pore waters – Biogenic opal dissolution versus authigenic clay mineral formation; Marz et al., 2015 Silica diagenesis and benthic fluxes in the Arctic Ocean, Ng et al., 2019 Sediment efflux of silicon on the Greenland margin and implication for the marine silicon cycle.**

**Furthermore, it might be of importance to mention the difference in flux magnitudes between abyssal plain and continental shelves for example.**

Answer : Thanks to reviewer n°2 to point out this ambiguity. Actually FW strictly represents dissolution of minerals and not « benthic fluxes » sensu lato. This sentence has been modified to avoid ambiguity (see below).

As a matter of fact section 2 deals with dSi inputs and not with Si outputs. Early diagenesis processes are discussed in section 3. Processes dealing with the dissolution of lithogenic material through submarine groundwater pathways (FGW, Figure 2), are discussed in section 2.3.

Section 2.2, has been rephrased (see below) to recall that, as shown in Fig. 2, FW is fed by two processes :

- the low-temperature dissolution of river-derived lithogenic particles deposited along the continental margins and shelves,
- and the low-temperature dissolution of basaltic glass in seawater, processes at work mostly in deep waters.

As explained in the manuscript, numerous experiments have been set up to measure the flux of dissolution of lithogenic silica materials, demonstrating that effectively lithogenic silica and/or basaltic glass are able to dissolve at low temperature (Jones et al. 2012 ; Morin et al. 2015 ; Oelkers et al. 2011 ; Pearce et al. 2013). However, extrapolations of these experimental data to estimate FW are difficult since the experimental conditions are far for the environmental conditions, which is why they not been retained in our manuscript.

Another approach to get an estimate for FW is to retain the benthic fluxes corresponding to dSi diffusive efflux from siliceous mineral sediments, as did Frings (2017) and TDLR 2013. Frings et al's high value for FW is not retained because of a possible bias due to the presence of biogenic silica in some of his « non-biogenic silica sediments ». This is why we finally retained TDLR (2013)'s estimate. Note, that this a world ocean total, because we are not able to make a difference between the contribution of abyssal plains and continental shelves. A point that would be mentioned in the perspective section (n° 6).

Modified text :

## “2.2 Dissolution of minerals (FW)

As shown in Fig. 2, the low-temperature dissolution of siliceous minerals in seawater and from sediments feeds a dSi flux, Fw, through two processes: (1) the dissolution of river-derived lithogenic particles deposited along the continental margins and shelves, and (2) the dissolution of basaltic glass in seawater, processes at work mostly in deep waters. About 15-20 Gt yr-1 river-derived lithogenic particles are deposited along the margins and shelves (e.g. Syvitskia et al., 2003, also see Fig. 2). Dissolution experiments with river sediments or basaltic glass in seawater showed that 0.08-0.17% of the Si in the solid phase was released within a few days to months (e.g., Jones et al., 2012; Morin et al., 2015; Oelkers et al., 2011; Pearce et al., 2013). However, the high solid-to-solution ratios in these experiments increased the dSi concentration quickly to near-equilibrium conditions inhibiting further dissolution, which prevents direct comparison with natural sediments. Field observations and subsequent modelling of Si release range around 0.5 – 5 % yr-1 (e.g., Arsouze et al., 2009; Jeandel and Oelkers, 2015). On the global scale, Jeandel et al. (2011) estimated the total flux of dissolution of minerals to range between 0.7 - 5.4 Tmol-Si yr-1, i.e. similar to the dSi river flux. However, this estimate is based on the assumption of 1 - 3 % congruent dissolution of sediments for a large range of lithological composition which, so far, is not proven.

Another approach to get an estimate of Fw is to consider the benthic efflux from sediments devoid of biogenic silica deposits. Frings (2017) estimates that “non-biogenic silica” sediments (i.e. clays and

calcareous sediments, which cover about 78% of the ocean area) may contribute up to 44.9 Tmol-Si yr-1 via benthic diffusive Si flux. However, according to lithological descriptions given in GSA Data Repository 2015271 some of the “non-biogenic silica” sediment classes described in this study may contain significant bSi, which might explain Frings’ high estimate for FW. In agreement with Tréguer et al. (1995), Tréguer and De La Rocha (2013) considered benthic efflux from non-siliceous sediments ranging between ~10-20 mmol m-2 yr-1. If extrapolated to 120 Mkm2 zone of opal-poor sediments in the global ocean, this gives an estimate of  $F_w = 1.9 (\pm 0.7)$  Tmol-Si yr-1.”

**-L122: 1-3% in LaTeX cod 1-3\% :**

Answer : Corrected.

**-L152: “the potential flux of dissolution of quartz from sandy beaches” to avoid a series of “of”**

Answer : Corrected

« ...calculated that the potential flux of dissolution of quartz (sandy beaches), »

**-L 163 -179 : Recent study could be added to this section : Hirst et al. 2020 ; Hatton et al. 2020**

Answer : We thank the reviewer for pointing out these recent publications and have added some additional text in this section. To clarify, the endmembers from these studies are not used in the calculation of flux estimates from Antarctica. They are considered to originate from inputs too small to make any significant difference to estimates (e.g. the meltwater flux from the McMurdo Dry Valleys and Antarctic Peninsula is very small compared to the subglacial flux estimated for the Antarctic continent; van Wessem et al., 2017 ; Lyons et al., 2015). We therefore still use only the subglacial lake endmember (Michaud et al., 2016) to estimate Antarctic Ice Sheet fluxes, which remains the best estimate of concentrations from waters most of which exits the subglacial environment of the Antarctic Ice Sheet beneath ice shelves.

-Van Wessem et al. (2017) Characteristics of the modelled meteoric freshwater budget of the western Antarctic Peninsula, Deep Sea Research Part II: Topical Studies in Oceanography, 139, 31-39

-Lyons et al. (2015) Antarctic streams as a potential source of iron for the Southern Ocean, Geology, doi: doi:10.1130/G36989.1

**-L177: Are the benthic fluxes from Ng et al., 2020 included in the (sub)polar glaciers estimation? If that's the case I think they should be removed and added to the benthic fluxes in the previous section. The citation reference of Hendry et al., 2019 is missing in the References section**

Answer : The benthic fluxes from Ng et al. (2020) are not included in the flux estimates for (sub)polar glaciers – only freshwater fluxes. This citation was added to emphasise the potential importance of indirect glacial fluxes, which are often not considered. The aSi flux would presumably indicate the most likely phase to dissolve in these benthic environments.

**-L186: what do you mean by focused?**

Answer : Focused hydrothermal venting generally refer to high-temperature hydrothermal fluids from ridge axis, forming sulfide chimney deposits. We added an explanation in the text.

“..A major challenge limiting our current models of both heat and mass flux (e.g. Si flux) through the seafloor is estimating the distribution of the various forms of hydrothermal fluxes, including focused (i.e. high-temperature) vs. diffuse (i.e. low temperature) and ridge axis vs. ridge flank fluxes. Estimates of the Si flux for each input are detailed below.” ...

**-L194: “is required” for what? This sentence feels to not be finished.**

Answer: Text corrected:

...“the required seawater flux is 5.9 ( $\pm 0.8$ )  $10^{16}$  g yr<sup>-1</sup> (Mottl, 2003).”

**-L197: do you have a reference list for these 100 discrete vent fluid data? If yes could you add it in the text.**

Answer : This dataset is available here;

Mottl, M. 2012. Explanatory Notes and Master Chemical Item Spreadsheet for the VentDB Data Collections housed in the EarthChem Library, Version 1.0. Interdisciplinary Earth Data Alliance (IEDA). <https://doi.org/10.1594/IEDA/100207>.

The text is amended as follows:

“...which is the average concentration in hydrothermal vent fluids that have an exit temperature > 300°C (Mottl, 2012).” ...

**-L206: Either “... (Mottle 1983; Von Damm et al, 1991).” or change to “... (Mottle 1983; Von Damm et al, 1991), and it is possible ...”**

Answer: Text corrected:

“...and is mainly controlled by the solubility of secondary minerals such as quartz (Mottl 1983; Von Damm et al. 1991),..”

**-L223: a word is missing in the sentence or the sentence needs to be rephrased.**

Answer : Text corrected

“...Using basaltic formation fluids from the 3.5 Ma crust on the eastern flank of the Juan de Fuca Ridge (Wheat and McManus, 2005), a global flux of 0.011 Tmol-Si yr-1 for warm ridge flank is calculated....”

**-L234: “Si anomaly of 0.07 . . . North Pond (S18)” could you add a reference here?**

Answer : Reference added:

Meyer, J.L., Jaekel, U., Tully, B.J., Glazer, B.T., Wheat, C.G., Lin, H.-T., Hsieh, C.-C., Cowen, J.P., Hulme, S.M., Girguis, P.R. and Huber, J.A. (2016) A distinct and active bacterial community in cold oxygenated fluids circulating beneath the western flank of the Mid-Atlantic ridge. *Scientific Reports* 6, 22541.

**L246: “the error propagation from Bevington and Robinson, 2003.**

Answer : Corrected

**-L267: no space for the ratio, Si:C, same for L271**

Answer : Corrected

**-L275: replace Aller et al., 1996 with the more recent Aller et al 2014**

Answer : Corrected

**-L279: “ $^{32}\text{Si}$  activities . . . delivery to the sediments” this sentence is missing a word or needs to be rephrased.**

Answer : text corrected as follows :

«  $^{32}\text{Si}$  activities signal represent approximately ~50% of initial bSiopal delivery to sediments (Rahman et al., 2017) »

**-L284: Did you mean “Fb of 7.0”?**

Answer : text corrected as follows :

« ... revised global total burial flux, *FB*, of 9.2 ( $\pm 1.6$ ) Tmol-Si yr-1,...»

**-L285: 11%**

Answer : Corrected

**-L290-293: I agree that if a sponge lived without disturbance the bSi accumulation through biosilicification is a long process and it is likely that it is long compared to the deposition to the sediment but do we really know how long it take for the spicules to be deposited and then buried within the sediment and being considered as bSi accumulated?**

**The supplementary material does not add more details neither reference for the rapid process lasting days to months.**

Answer : When a sponge dies, its organic parts are rapidly degraded and disappear (Rützler and McIntyre, 1978 as new reference), but the spicules remain in a patch that slowly disaggregate on the seafloor. There are several references testifying for this latter slow process. We have now included them along with a more detailed explanation on this process in the Supplementary “Section 3- The output fluxes”

Rützler, K. and Macintyre, I. G. Siliceous sponge spicules in coral reefs sediments. *Marine Biology*, 49 (2), 147–159 (1978).

Laguionie-Marchais, C., Kuhnz, L. A., Huffard, C. L., Ruhl, H. A., & Smith Jr., K. L. (2015). Spatial and temporal variation in sponge spicule patches at Station M, northeast Pacific. *Marine Biology*, 162(3), 617–624. <https://doi.org/10.1007/s00227-014-2609-1>

Bett, B. J., & Rice, A. L. (1992). The influence of hexactinellid sponge (*Pheronema carpenteri*) spicules on the patchy distribution of macrobenthos in the Porcupine Seabight (bathyal NE Atlantic). *Ophelia*, 36, 217–226. <https://doi.org/10.1080/00785326.1992.10430372>

Regarding the question of how long takes for the spicules to be buried and considered as accumulated bSi, it will depend mostly on the local rate of sediment deposition, also on bioturbation (Katz et al., 2016). In the only study addressing this issue so far, the rates varied across marine environments, ranging from 471 years to 74,074 years. See table 2 in the below reference Maldonado et al. 2019:

Katz, T., Yahel, G., Tunnicliffe, V., Herut, B., Whitney, F., Snelgrove, P. V. R., & Lazar, B. (2016). The silica cycle in a Northeast Pacific fjord; the role of biological resuspension. *Progr. Oceanogr.* 147, 20-21 (2016).

Maldonado, M., López-Acosta, M., Sitjà, C., García-Puig, M., Galobart, C., Ercilla, G., & Leynaert, A. (2019). Sponge skeletons as an important sink of silicon in the global oceans. *Nature Geoscience*, 12(10), 815–822.

The Fsp provided in the current manuscript has been calculated by re-analyzing sediment samples from 19 cores worldwide distributed, as indicated in the above reference Maldonado et al. 2019.

In response to this comment by the reviewer, we have added all the above information and the pertinent references to the supplementary section 3.

**-L293: Did you mean Supplement, section 3? L306: change to “considering Maldonado et al., 2019, the new best estimate for FSP is . . .”**

Answer : Yes, we made a mistake and really meant Supplement, section 3. We have also correct the extra-bracket in L306. Thanks

**-L321: you could add Pickering et al., 2020, Geilert et al., 2020 to the reference list.**

Answer : These two papers are now listed in the reference list and the text is corrected accordingly.

« Recent direct evidence supporting the rapid formation of authigenic clays comes from tropical and subtropical deltas (Michalopoulos 320 & Aller, 1995; Rahman et al., 2016, 2017; Zhao et al., 2017) and several geochemical tools show that authigenic clays may form ubiquitously in the global ocean (Baronas et al., 2017; Ehlert et al., 2016a; Geilert et al., 2020, Michalopoulos & Aller, 2004, Pickering et al. 2020). »

**-L345-358: This section is slightly confusing if I understand well the ocean basins are composed of a number of provinces and the domain is the provinces subdivided as coast, SO and open domain. Could you rephrase saying something like “for the domain estimate, each province was categorised either as a coast or an open domain. Only the SO is defined as a whole domain.”**

Answer : Each of the 56 provinces defined by Longhurst (1995) is part of a domain (which I propose to rename "biomes" for less confusion). For the domain estimate, five biomes are delineated: the coastal, upwelling, trades, westerlies and polar biomes (Table 2). Data from provinces within the same biomes are averaged. The "coastal" biome and the "upwelling" biome are then added together to represent the "coastal domain". The "westerlies" and "trades" biomes are combined to represent the "open ocean". The SO is defined as a whole biome.

Consequently, in the Supplemental Information, Annex 1, the last table has been modified accordingly.

**-Table 2 and section 4.1: To compare with the pelagic production, did you used the same parameters values (within 300km of shore) to estimate the coastal domain? It is actually very interesting to see that the production at the coast is almost half of the open ocean for the model estimation, obviously the surface area is much bigger for the open ocean. Do you think that the chlorophyll level here can be biased by other organisms for the open ocean area or maybe the lower resolution in the coastal area as many models do not cover the coast as well as the open ocean?**

**-Answer :** The concept of coastal domain differs according to the approaches, but it cannot explain the difference between model estimation and those based on field data.

The coastal area in the satellite productivity models is defined as the area <300 km from coastlines and is 52 106 km<sup>2</sup>.

For the estimation based on field data, the coastal domain is defined as the region whose general ocean circulation is significantly modified by the interaction with the topography and by the coastal wind regime (Longhurst, 2007). It represents a surface area of 37 106 km<sup>2</sup> (this area includes upwelling biomes which covers 8 106 km<sup>2</sup>).

Concerning the model resolution: Chl based models have good coverage in coastal waters as their resolution is 1/6 degree.

The issue of 'bias by other organisms' pertains to uncertainty in the fraction of productivity by diatoms in each chl category. This, we took from DARWIN which averaged 29% but was 9%, 30%, and 59% for oligotrophic, mesotrophic, and eutrophic waters. This is certainly a source of uncertainty.

At the end of section 4.1.1, the text has been modified as follows:

"Other sources of uncertainty in our bSi production estimates include poorly-constrained estimates of the Si:C ratio and dissolution: production ratios (see Supplemental Information). The errors incurred by these choices are more likely to cancel out in the global average, but could be significant at regional scales, potentially contributing to the discrepancies in productivity across the various methods."

**-There is some disagreement between the main text and SI: L408 in main text: "these models are likely to overestimate the role that diatoms play, especially in the SO" L146 in SI: "potential biases in SO chlorophyll concentration (and consequently, NPP), which may be underestimated in the Southern Ocean". As said in the SI the data disparity is a major problem in global estimates, it might be worth putting it in the main text at the end of section 4.1.3.**

**Answer :** The text has been modified as follows :

"In the Southern Ocean (SO), a key area for the world ocean Si cycle (DeMaster, 1981), there is some disagreement among the different methods of estimating bSi production. Field studies give an estimate of 67 Tmol-Si yr-1 for the annual gross production of silica in the SO, close to the estimate of 60 Tmol-Si yr-1 calculated using satellite productivities models (Table 2). However, the bSi

production in the SO estimated by ocean biogeochemical models is about twice as high, at 129 Tmol-Si yr-1 (Table 2). The existing in-situ bSi production estimates are too sparse to be able to definitively settle whether the lower estimate or the higher estimate is correct, but there is reason to believe that there are potential biases in both the satellite NPP models and the ocean biogeochemical models. Southern Ocean chlorophyll concentrations may be underestimated by as much as a factor of 3-4 (Johnson et al., 2013), which affects the NPP estimates in this region and hence our bSi production estimates by this method. The bSi production estimated by ocean biogeochemical models is highly sensitive to vertical exchange rates in the Southern Ocean (Gnanadesikan and Toggweiler, 1999), and is also dependent on the representation of phytoplankton classes in models with explicit representation of phytoplankton. Models that have excessive vertical exchange in the Southern Ocean (Gnanadesikan and Toggweiler, 1999), or that represent all large phytoplankton as diatoms, may overestimate the Si uptake by plankton in the Southern Ocean. Other sources of uncertainty in our bSi production estimates include poorly-constrained estimates of the Si:C ratio and dissolution: production ratios (see Supplemental Information).

The errors incurred by these choices are more likely to cancel out in the global average, but could be significant at regional scales, potentially contributing to the discrepancies in productivity across the various methods."

**-L451-457: why not replicate the same method used for the pelagic production, i.e. separating the coastal area and the abyssal plain. Abyssal plains are more likely going to represent the long-term equilibrium state than the coastal areas where sponges are sensitive to currents, particle/sediment accumulation, animals, seabed destruction etc.**

Answer : We understand the idea and would be delighted to be able to repeat the diatom approach for sponges. However, regarding sponges, the deep-ocean compartment remains poorly investigated and the available data are neither comprehensive nor accurate as to derive calculations from their own. There are currently several on-going, international, multidisciplinary research project to palliate this lack of knowledge. At the present moment, caution advices to provide just a global calculation for sponge bSi production and, if the reviewer and Editor agree, we would like to keep it as it is now rather than dealing with excessive speculation.

**-L451: "If the production bSi that . . ." did you mean if the bSi production that . . .?**

Answer : Yes, thank you. We changed it (see above)

**-L482: Could you define the number, something like FB/Gross bSi pelagic prod = (7.0/255) = 2.8%.**

Answer : text corrected

« ... The new estimate for the global average preservation efficiency of bSi buried in sediments is (FB = 9.2 /FPgross = 255=) 3.6 %,...»

**-L496-L501: This sentence is very long, could you break it?**

Answer : This sentence is simplified as follows :

... « Skeletal underdevelopment (Maldonado et al., 1999), and low performance in dSi consumption (Maldonado et al., 2020) occur in sponges when they use dSi at relatively modest concentrations, typical of most environments in the modern ocean. »

**-L511-513: What about changes in phytoplankton population due to climate changes. For example, along the West Antarctic Peninsula, phytoplankton community are quite sensible to the ice coverage, with the haptophyte and cryptophyte community increase while the diatom decrease (Henley et al., 2019. Variability and change in the west Antarctic Peninsula marine system: Research priorities and opportunities. Progress in Oceanography). It is something that need to be more studied but worth considering as these changes will impact the pelagic production such as the burial flux**

Answer : Thank for the suggestion. Referee n°2's inputs fits more in section 5.4.2 of the discussion (climate change), where Henley et al. 2019 is now included.

**-L524: you could add Pickering et al., 2020.**

Answer : Added.

« ... or sponge spicules that are abundant in sediments (Maldonado et al., 2019; Pickering et al. 2019). »...

**-L535: remove one of the brackets, (94.3 Tmol-Si yr-1)**

Answer : Corrected.

**-L575: the reference Johnson et al., 2006 is incorrect in the Bibliography list.**

Answer : Apologies ! The correct reference is now in the reference list :

« Johnson, H.P., Hautala, S.L., Bjorklund, T.A., Zarnetske, M.R.: Quantifying the North Pacific silica plume, Geoch. Geophys. Geosyst., <https://doi.org/10.1029/2005GC001065> »

**-L628-644:**

**-L628-629: Coastal and continental zone in the Southern Ocean are not as limited as we think by iron for example see Annett et al., 2015 Comparative roles of upwelling and glacial iron sources in Ryder Bay, coastal western Antarctic Peninsula, Sherrell et al., 2018, A 'shallow bathtub ring' of local sedimentary iron input maintains the Palmer Deep biological hotspot on the West Antarctic Peninsula shelf.**

**-L641-644: In coastal area the reduction of sea-ice has been shown to reduce the primary productivity not to increase it. I will suggest that the author give more details about which part of the ocean is considered, coastal or open ocean**

Here we are providing answers to the two reviewers for L628-629 and L641-L644.

The text has been modified as follows :

"In the 21st century, climate change affects ocean circulation, stratification and upwelling thus affecting the cycles of nutrients (Aumont et al., 2003; Bopp et al., 2005, 2013).

With increase stratification, reduced dSi supply from below (Fig. 1 and 4) leads to less siliceous phytoplankton production in surface compartments of lower latitudes and possibly the North Atlantic (Tréguer et al., 2018).

Based on field studies, the impact of climate change on the phytoplankton production of higher latitudes is highly debated as melting of sea ice decreases light limitation. Regarding the Arctic Sea, increase nutrients (at the least for silicic acid) availability will occur through the Transpolar Drift delivering nutrient rich river- and shelf derived waters as potential sources for primary production, including bSi production (e.g. Charette et al., 2020). Regarding the Southern Ocean, a well known area for low dissolved iron (dFe) concentrations (Tagliabue et al 2016), except in the CCMZ (e.g. Annett et al. 2015 ; Sherrell et al. 2018), bSi production might increase in areas impacted by dFe inputs from icebergs melting or from increased convection that feeds surface waters in dFe from below (Boyd et al., 2016 ; Hutchins & Boyd, 2016 ; Tréguer et al., 2018). Changes in Southern Ocean phytoplankton population due to climate change have been observed already. For example, along the West Antarctic Peninsula, phytoplankton community are quite sensible to the ice coverage, with the haptophyte and cryptophyte community increase while the diatom decrease (Henley et al., 2019). Globally, it is therefore possible that a warmer and acidified ocean alters the pelagic bSi production rates, thus modifying the export production, and the Si burial rates at short time scales.

Although uncertainty is substantial, model studies (Bopp et al., 2005; Dutkiewicz et al., 2019; Laukötter et al, 2015) suggest regional shifting of bSi pelagic production due to climate change. Climate change models suggest a global decrease in diatom biomass and productivity over the course of the 21st century (Bopp et al., 2005, Dutkiewicz et al., 2019, Laukötter et al., 2015), which would lead to a reduction in the pelagic biological flux of silica. Regional responses however differ, with most models suggesting a decrease in diatom productivity in the lower latitudes and many predicting an increase in diatom productivity in the Southern Ocean (Laukötter et al, 2015). Holzer et al. (2019) suggest that changes in supply of dFe will alter bSi production mainly by inducing floristic shifts, not by relieving kinetic limitation. Increased primary productivity come from reduction in sea-ice and the faster growth rates with warmer waters and longer growing seasons in the high latitudes. However,

many models have very simple ecosystems including only diatoms and a small phytoplankton. In these models, increased primary production in the Southern Ocean is mostly from diatoms. Some models with more complex ecosystem (i.e. including additional phytoplankton groups) suggest that increased primary productivity in the future Southern Ocean will be due to other phytoplankton types (e.g. pico-eukaryote) and that diatoms biomass will decrease (Dutkiewicz et al, 2019; also see model PlankTOM5.3 in Laufkötter et al, 2015), except in regions where sea-ice has melted. Differences in the complexity of the ecosystem and parameterizations, in particular in terms of temperature dependences of biological process, between models lead to widely varying predictions (Dutkiewicz et al., 2019; Laufkötter et al., 2015) thus constraining our capacity to predict what will happen with the silica biogeochemical cycle in a future ocean."

**-L694-697: As mentioned before, some very recent papers (i.e. Ng et al., 2020, Pickering et al., 2020, Geilert et al., 2020) have not been included in this review.**

Answer : *These 3 references are now included in the revised version.*

*Ng et al. 2020 is cited in section 2.4. The two last references are now included in the list of references (see section 3.3).*