

We have submitted a revised manuscript (ms), which includes all the modifications described in our two response letters. The main differences from our ms published in BGD (referred to original ms below) are as follows:

1. Abstract is modified in the revised ms to include the estimated fluxes according to the suggestion from the second reviewer.
2. Several sentences are modified in the Introduction section of the revised ms to mainly explain or introduce:
 - Why does benthic denitrification replace anammox as the fixed-N loss process in the model;
 - DON and its bioavailability scenarios are included in our analysis;
 - Why is riverine nitrogen input excluded from of our analysis.
3. One paragraph is added in Section 2.1 Circulation and biogeochemical model (Page 6 of the revised ms) to describe ordinary phytoplankton and nitrogen fixers in detail.
4. A brief description of the nine main model configurations is added in Section 2.2 Model configurations of the revised ms.
5. The explanation to why phosphate deposition is excluded from our analysis is modified in Section 2.3 Atmospheric nitrogen deposition (Pages 7–8 of the revised ms).
6. Three new model sensitivity experiments are performed and added in the revised ms:
 - Model sensitivities to uncertainties underlying the atmospheric nitrogen deposition, atmospheric DON deposition and its bioavailability, and predicted increasing nitrogen deposition;
 - The influence of aphotic nitrogen fixation on the nitrogen budget of our model domain;
 - Model sensitivity to spatially variable Martin Curve exponents b .
7. The Section 2.6 Synthesis configurations has been integrated into Section 2.2 Model configurations in the revised ms.
8. The ship-collected net primary production from Pennington et al. (2016) is compared to our NPP estimates, which is discussed on Pages 19–20 of the revised ms.
9. Flux comparison with other model or observational estimates are summarized in Table 3 of the revised ms, which are discussed on Pages 20–21 of the revised ms.
10. The Discussion and conclusions section of the revised ms is improved according to the comments from our first reviewer, to have a better understanding of the eastern tropical South Pacific in the light of more recent findings.
11. A new Summary section is added on Page 23 of the revised ms according to the suggestion from the second reviewer.
12. Table 1 of the original ms is modified by explaining the process abbreviations in the caption, which is the new Table 1 of the revised ms.
13. Tables 2 and 3 of the original ms are integrated into one table, which is the new Table 2 of the revised ms. The references of the relative estimates are added in the new table.
14. The sequence of the figures are changed and some of them are modified in the revised ms:
 - Fig. 1 in the original ms → Fig. 1 in the revised ms: a panel is added to show the geographical location of our model domain.
 - Fig. 2 in the original ms → Fig. 2 in the revised ms: the nitrogen deposition and nitrogen fixation are shown separately for the U and S boxes.

- Fig. 3 in the original ms → Fig. 3 in the revised ms.
- Fig. 4 in the original ms → Fig. 4 in the revised ms: the nitrogen deposition and nitrogen fixation are shown separately for the U and S boxes.
- Fig. 5 in the original ms → Fig. 6 in the revised ms.
- Fig. 6 in the original ms → Fig. 7 in the revised ms.
- Fig. 7 in the original ms → Fig. 8 in the revised ms.
- Fig. 8 in the original ms → Fig. S1 in the supplementary materials of the revised ms: the nitrogen deposition and nitrogen fixation are shown separately for the U and S boxes .
- Fig. 5 in the revised ms is new.

15. A supplement is added to the revised ms with Appendices A–D.

16. Appendix A of the original ms is moved to the supplement of the revised ms.

17. The term phosphate regeneration in the original ms is changed to phosphorus regeneration in the revised ms, which we think is better.

Please see our response letters for details.

The detailed differences between the original and revised ms are also shown in the marked-up PDF file processed with *latexdiff*, which we have submitted together with the revised ms.

We have submitted two letters respectively in response to the comments from our two referees already. Here we summarise our detailed point-by-point response to referee comments and specify all changes in the revised manuscript (ms) in response to the relevant comments. A list of all relevant changes made in the revised ms is also submitted together with this response. We will refer to the ms published in BGD as the original ms, and all figure and table numbers refer to the original ms unless otherwise noted.

Response to Referee #1:

Comment 1: Based on a prognostic 5 box model of the OMZ region in the ETSP published earlier in 2015, the authors focus here on the effects and feedbacks between major sources and sinks in the marine N cycle. They consider in particular the atmospheric deposition and benthic remineralisation. I found this work rather interesting as it tends to show that those processes are important for N cycle in the studied domain. I see however several points concerning the numerous assumptions and/or simplification that have been done in the model that should be addressed before publication in Biogeosciences.

I also find that the short discussion-conclusion section was too much centered on model results. I would thus recommend to take a step back on the results in order to draw a discussion (and conclusions) that may have a stronger and broader impact for the understanding of that complex area in light of recent findings.

Response 1: The authors thank the reviewer for these generally positive comments and the suggestions on the structure of the manuscript. Please see our modifications to the “Discussion and Conclusion” section in the revised ms (Pages 19–23), in which more recent findings as suggested by both reviewers have been included to provide a better understanding of the eastern tropical South Pacific (ETSP).

Comment 2: One can wonder how the uncertainties linked to those numerous assumptions make this version of the model really solid. For example, only heterotrophic denitrification was considered for fixed-N loss process; for atmospheric deposition, DON is not considered (because of a lack of data but it has been shown recently that this fraction could be very important — see below); also riverine inputs are not considered; also phosphorus atmospheric deposition is not considered. And last but not least, aphotic N₂ fixation process is not mentioned in the study. I have not too much problems with simplification but at one point, these simplification should be also part of the discussion: how the omission of all those impact/process influence or not the results/conclusions.

Response 2: The Question about atmospheric nitrogen and phosphorus deposition and aphotic N₂ fixation is addressed below in response to **Comments 3 and 4**. Denitrification as the only fixed-N loss processes was already explained on Page 14444 Lines 10–14 of the original BGD ms:

Even though anammox has been observed to contribute significantly to the fixed-N loss in the ETSP, both denitrification and anammox are ultimately driven by the flux of organic matter into the OMZ. Because the NH₄⁺ for anammox from either ammonification or dissimilatory nitrate reduction to ammonium (DNRA) is ultimately determined by the organic matter fluxes into the OMZ [Lam et al., 2009, Koeve and Kähler, 2010].

We rephrase these sentences to clarify this point on Page 4 Lines 8–13 in the revised ms:

Anammox has been observed to be a major fixed-N loss process in the eastern tropical South Pacific (ETSP) [Lam et al., 2009, Kalvelage et al., 2013]. However, the essential substrates for anammox are ultimately provided by heterotrophic processes [Koeve and Kähler, 2010, Kalvelage et al., 2013], such as organic-matter remineralisation or dissimilatory nitrate reduction to ammonium (DNRA). Thus, both denitrification and anammox are ultimately driven by the flux of organic matter into the OMZ.

No large rivers flow into the ETSP from the coast of Peru. According to Seitzinger and Kroeze [1998], riverine DIN input into the ETSP only accounts for less than 1.5 % of the total riverine nitrogen input into the Pacific and increases the NO_3^- concentration of our study area by less than $2 \times 10^{-6} \mu\text{mol N L}^{-1}$, assuming that all the riverine DIN was distributed evenly, which indicates a very minor role in the nitrogen budget of the ETSP. Therefore, riverine nitrogen input is excluded from our model analysis. We rephrase the text on Page 14445, Lines 13–17 of the original BGD ms to read:

Using a conceptually simple and computationally efficient box model, we here attempt a synthesis considering all essential sources and sinks and their mutual interactions, with the only exception of riverine input, which is excluded because it contributes only negligibly to the nitrogen inventory in the ETSP [Seitzinger and Kroeze, 1998].

on Page 5, Lines 12–15 in the revised ms.

Atmospheric phosphorus deposition is also excluded from our analysis. We clarify the statement from Page 14447, Lines 10–15 of the original BGD ms as follows on Page 7, Line 25–Page 8, Line 2 of the revised ms, also considering the comments from the other reviewer:

Atmospheric phosphorus deposition is excluded from our analysis because its amount is much smaller than the Redfield equivalent of nitrogen atmospheric deposition [Duce et al., 1991]. This results in N/P (mole/mole) ratios of more than 100, much higher than the average elemental N/P ratio required by phytoplankton [Duce et al., 2008, Mahowald et al., 2008].

Comment 3: I would recommend to give more detail on nitrogen atmospheric deposition used in the model. Inclusion of atmospheric deposition in your model is a hint of the paper: it needs more solid assessments. This is an important addition to the previous model and it is important to provide more information on the data used. The section on atmospheric deposition is very short, and estimates of DIN deposition used need to be more explained. Considering that this area has only been validated by scarce field data, the uncertainty on the flux (from models) are quite high. How these uncertainty impact your model results? Also concerning the fact that atmospheric Organic Nitrogen was not considered in the model although recent work have shown how important this fraction can be for total nitrogen inputs. For ex., Kanakidou et al., 2012 indicate an average of 35 % of Organic Nitrogen of the total soluble N in wet deposition: this deserves to be discussed as atmospheric deposition used in your model is in fact most likely underestimated: how this can impact the results?

Response 3: We thank the reviewer for this suggestion. We modify the “Model description” section about atmospheric deposition on Page 11, Lines 13–26 in the revised ms. Since the atmospheric nitrogen deposition data from Lamarque et al. [2011] only include one chemistry-climate model results, a multi-model perspective could offer additional insights into the influence of uncertainties in nitrogen deposition on our model results. Three recent inter-model comparisons [Dentener et al., 2006, Lamarque et al., 2013, Vet et al., 2014] show very similar performance over our model domain, therefore we choose the results from Dentener et al. [2006], which is also applied in a number of benchmark papers such as Duce et al. [2008]. The results are shown in new Fig. 5 in the revised ms.

Dissolved organic nitrogen (DON) accounts for about 30 % of the total dissolved N deposition in South America [Cornell et al., 2003, Kanakidou et al., 2012], and about 30 % of the deposited DON is available to primary producers, increasing by about 13 % the bioavailable nitrogen input into the model domain. Considering also a suggestion from the other reviewer, we investigate further scenarios for different DON bioavailability and future emission increases. We summarize the results in the new Fig. 5, and on Page 17, Lines 2–12 of the revised ms. The results are discussed on Page 19, Lines 18–22 in the “Discussion and conclusions” section of the revised ms.

Comment 4: There is one process that should be taken into consideration or at least discuss why it is not and how it could change the presented budget: this is the aphotic N₂ fixation in that area, a process that was recently evidenced to be very important in ETSP according to Bonnet et al., 2013. In your study, N₂ fixation was only considered in the top 100m layer. Bonnet et al., clearly state in their conclusion: ‘These new sources of N could potentially compensate for as much as 78 % of the estimated N loss processes in ETSP, indicating that they need to be taken into account in marine N budgets’. How can this important question be addressed in your work? How this actual process and important source of fixed N will affect your proposed nitrogen-balancing mechanism in that area? Note also that the same authors find that N₂ fixation was never inhibited after NO₃⁻ addition, an interesting finding that could also be discussed.

Response 4: We thank the reviewer for this information. We now include the aphotic nitrogen fixation estimates from the 2010 and 2011 cruises reported by Bonnet et al. [2013] in our model analysis. Supplementary Fig. S5 shows the nitrogen fluxes after including aphotic nitrogen fixation. Photic nitrogen fixation decreases by 39 % and 15 % in the AphoticNfix1 and AphoticNfix2 configurations. Water-column denitrification rate stays steady because more nitrogen input by aphotic nitrogen fixation does not increase export production into the OMZ. The lateral fixed-N effluxes in the AphoticNfix1 and AphoticNfix2 configurations are about 33 and 4 times of that in the control configuration, accounting for 91 % and 78 %, respectively, of extra nitrogen input by aphotic nitrogen fixation. Tracer concentrations at steady state are presented in supplementary Fig. S6. Aphotic N₂ fixation has little effect on most tracers except NO₃⁻, which increases by 110 % and 87 % respectively in the UM box and the model domain in AphoticNfix1. While the lower estimate of aphotic N₂ fixation (AphoticNfix2) brings the NO₃⁻ concentrations closer to the WOA2009 data, the high estimate (AphoticNfix1) leads to a strong overestimate. We conclude that aphotic N₂ fixation is likely closer to the lower (2011) estimate of Bonnet et al. [2013]. We incorporate this process as sensitivity experiment and describe it on Page 12, Lines 1–11 on the revised ms. We also describe the results and discuss this topic on Page 17, Lines 13–26 and Page 20, Lines 14–24 of the revised ms.

Bonnet et al. [2013] found that N₂ fixation by non-cyanobacteria diazotrophs was never inhibited after NO₃⁻ addition in the OMZ where ambient NO₃⁻ concentration are in a range of 30–40 $\mu\text{mol L}^{-1}$. In their results, the reasons for this are not resolved because the metabolic potential of diazotrophs is not characterized. In our AphoticNfix1 and AphoticNfix2 model configurations, we have simply integrated aphotic nitrogen fixation as fixed-N input fluxes into the respective regions, thus avoiding the issue of NO₃⁻ inhibition for N₂ fixations in the aphotic zones. In the surface boxes, PO₄³⁻ is the only limiting nutrient for nitrogen fixers and NO₃⁻ does not inhibit the growth of nitrogen fixers.

Comment 5: I found that the model concept and results was often quite decoupled from actual field knowledge and data for the given area. This is the case for my comment regarding atmospheric deposition, N₂ fixation; this is also the case for the estimation of the rain rate POC. The ‘classical’ b=0.82 is taken into consideration although it is well known that b depends on a number of parameter and is not constant over the ocean. In the recent regionalization study from Guidi et al. 2015, it is well demonstrated that ‘b’ is a non constant number resulting from non uniform remineralisation. We are all aware of that but I believe that it is important to take into account recent findings and at least discuss the limit of your hypothesis in light of those recent findings. See their table 2 for the regions included in ETSP (CHIL, PEQD and SPSG), actually, their ‘b’ is close to the Berelson value (although lower for the SPSG domain). I think this is an interesting point to better discuss in light of recent data.

Response 5: In the main configuration of our model, a constant Martin Curve exponent $b = 0.82$ is applied for the whole ETSP, because this values is a global average and also very close to the average value estimated for the ETSP by Berelson [2001] (Fig. 2 in Berelson [2001]). A lower b value ($b = 0.4$) under anoxic conditions, as suggested by Van Mooy et al. [2002], is applied in our sensitivity analysis, which can be considered as a lower limit for the Martin curve exponent b . In order to consider the recent findings by Guidi et al. [2015], we also consider spatial variations in b within our model domain.

In the UM-box, $b = 0.83$, which corresponds to the Peru-Chile upwelling region (CHIL), is applied. $b = 0.85$ is applied in the D-box, which is the average of the b values in regions named CHIL, PEQD, SPSG, and WARM in Guidi et al. [2015].

The nitrogen fluxes in configurations with different b values (bars marked ‘C’ in supplementary Fig. S7) are in good agreement with those in our main configuration (bars marked ‘A’ in supplementary Fig. S7). Nitrogen fixer (NF) concentrations in the surface boxes are also robust after including spatially variable b values (supplementary Fig. S8). Thus, regionally varying b values as implied by Guidi et al. [2015] have only minor effects on our model results. Our corresponding sensitivity experiment is described and its results are presented on Page 12, Lines 22–29 and Page 18, Lines 9–12 of the revised ms, respectively.

Comment 6: Define MBD and DBD also in the text (only in caption Table 1). This will make it easier for the reader.

Response 6: The text below is included on Page 7, Lines 6–17 of the revised manuscript to define the configurations briefly in the text:

In the NDEP configuration, atmospheric nitrogen input into the surface ocean according to the estimate by Lamarque et al. [2011] is included; MBD and DBD are configurations in which model- and data-based benthic denitrification are included in the control configuration; MPR and DPR represent configurations with model- and data-based benthic phosphorus regeneration respectively. Detailed information of all processes is presented in Sects. 2.3, 2.4 and 2.5.

Nitrogen deposition, benthic denitrification and phosphate regeneration are integrated into the synthesis model configurations to explore the model sensitivity to each process and their mutual interactions in the ETSP. Synthesis configuration Syn1 includes model-based benthic denitrification and phosphorus regeneration; Syn2 includes the data-based benthic denitrification and phosphorus regeneration; Syn3 also includes atmospheric deposition in addition to the processes in Syn1; Syn4 includes atmospheric deposition in addition to the processes in Syn2. The synthesis configurations Syn1 to Syn4 are also summarized in Table 1.

Comment 7: I would rather call the atmospheric source of nitrogen that enters the open ocean available for biota ‘reactive’ and not ‘fixed’ (although it is commonly used).

Response 7: We think that “bioavailable” may be a better term, and change it in the revised ms.

Comment 8: I would add a figure of the actual model domain showing the ETSP.

Response 8: We add a panel to Fig. 1 in the revised ms, showing the geographical location of the model domain.

Response to Referee #2:

Comment 1: Su et al. used a prognostic 5-box circulation model from a previous published manuscript (Su et al., Biogeosciences, 2015) to investigate the effects of atmospheric nitrogen deposition and benthic remineralization on the nitrogen cycle of the Eastern Tropical South Pacific (ETSP). Their main findings are that 1) N deposition is offset by half by reduced N₂ fixation with the other half exported out of their model domain, 2) sedimentary denitrification and phosphate regeneration under suboxic conditions acts to increase N₂ fixation, and 3) this increased N₂ fixation is partly removed by stronger water-column denitrification. Overall, they claim that these stabilizing feedbacks keep a

balanced nitrogen inventory in the ETSP. While their results are interesting, I have some major issues with the paper and recommend revisions before publication in *Biogeosciences*.

Response 1: The authors thank the reviewer for the generally positive comments and recommendation for publication in *Biogeosciences* after revision.

Comment 2: First, I feel that many simplifications have been made in their model. For instance, the authors assume that N₂ fixation is ultimately limited by P supply but omit to consider the important role of Fe. Fe has been shown to control patterns of N fixation, even in regions where Fe depositions are higher than in the ETSP (e.g., see Moore et al., *Nature Geoscience*, 2009). In a recent study published in *Global Biogeochemical Cycles*, Dekaezemacker et al. (2013) reported that N₂ fixation was stimulated by Fe addition in the ETSP. Therefore, I would like them to describe the role of Fe limitation on N₂ fixation in their model.

Response 2: While Fe deposition is low in this region, Fe concentrations are not, as also reported by Dekaezemacker et al. [2013]. It is somewhat unclear to us how to interpret the results of the Fe-enrichment experiments of Dekaezemacker et al. [2013] in terms of modeling. Stimulation of N₂ fixation was inversely related to ambient Fe concentrations, so that the Fe enrichment raised N₂ fixation by 200 % with low ambient Fe, but by more than 450 % with 10-times higher ambient Fe, with only minor differences in ambient nitrate and phosphate concentrations. These findings are counter to what would be expected if N₂ fixation was mainly Fe limited. Dekaezemacker et al. apparently came to the same conclusion, writing in their Discussion that “Finally, the high positive Fe* (Table 1) in 2011 indicates that there is enough iron to support the complete consumption of PO₄³⁻”. In our view, the role of Fe limitation in the region of our model domain thus remains too unclear to warrant inclusion in our model.

Comment 3: Furthermore, they also neglected DON, that represents ~ 30% of total dissolved N wet depositions in South America (see Cornell et al., *Atmospheric Environment*, 2003). While I understand that the bioavailability of DON is still unclear, some estimates are available, for instance, Peierls and Paerl (*Limnology and Oceanography*, 1997) suggested that ~ 20 – 30% of atmospheric organic N is readily available to primary producers. Therefore, I believe that they could test different scenarios regarding DON bioavailability in their model. I would also like to see a scenario with increased N depositions that reflects predicted future changes. Second, they separate the coastal and open ocean regions in their model (e.g., U and S boxes) but fail to discuss these separately in their discussion. Coastal regions are highly productive compared to the open ocean, therefore I would expect fluxes to be significantly different, as shown in their sensitivity analysis (Figures 3, 5 and 6). However, only global fluxes for the two regions are shown in Figures 2 and 4. I would like them to separate their model results for these two regions and better discuss these results in their discussion.

Response 3: Dissolved organic nitrogen (DON) accounts for about 30% of the total dissolved N deposition in South America [Cornell et al., 2003, Kanakidou et al., 2012], and about 30% of the deposited DON is available to primary producers [Paerl and Whitall, 1999], increasing by about 13% the bioavailable nitrogen input into the model domain. Predicted increasing nitrogen deposition is estimated according to the RCP8.5 scenario projections for 2100 [Lamarque et al., 2011], and we have tested this scenario in our model.

We now also show the results separately for the upwelling region and open ocean, and discuss them separately in the revised ms. These results are shown in the new Fig. 5 in the revised ms. The model description about atmospheric DON deposition and increasing nitrogen deposition predicted by Lamarque et al. [2011] is included on Page 11, Lines 13–26 of the revised ms. The results are presented and discussed on Page 16, Line 22–Page 17, Line 12 and Page 19, Lines 18–22, respectively, of the revised ms.

Comment 4: Third, I feel a comparison of their fluxes with direct measurements from previous studies is needed in their discussion. For example, do their N₂ fixation and N-loss fluxes match what

can be derived from direct rate measurements from past studies (e.g., Kalvelage et al., 2013 for N-loss and Dekaezemacker et al., 2013 and Lscher et al., 2014 for N₂ fixation) for the considered region?

Response 4: We thank the reviewer for this very helpful comment. The flux comparison is included in the new Table 3 of the revised ms. The discussion of this comparison is included on Page 20, Line 25–Page 21, Line 13 of the revised ms.

Comment 5: Abstract: Should include actual numbers (ranges) for global fluxes derived from their model.

Response 5: We now compare the fluxes in our model with other estimates in Table 3 of the revised ms. We also include some of these numbers in our abstract in the revised ms. We would like to stress, though, that the quantification of these numbers is not the main goal of our analysis.

Comment 6: P. 14442, line 20: This statement appears to be incorrect. How can the ESTP be a NO₃⁻ source when we observe such high N deficits in the Oxygen Deficient Zone (ODZ) of this region (e.g., Codispoti, Biogeosciences, 2007)? N-loss rates (up to 36 nmol N l⁻¹ d⁻¹; Kalvelage et al., 2013) from direct measurements are also generally at least 1–2 orders of magnitude higher than N₂ fixation rates in the ETSP (0.01 to 0.9 nmol N l⁻¹ d⁻¹; Dekaezemacker et al., 2013 and Lscher et al., 2014).

Response 6: We agree that the coastal region could be a sink of fixed-N, particularly when the high rate of nitrogen-loss by benthic denitrification is considered. But we would like to point out here that the criticised sentence does **not** say that the ETSP is a NO₃⁻ source but states explicitly that the **water column** of our model domain could be a NO₃⁻ source, and that the ETSP could be a NO₃⁻ sink when benthic denitrification is considered. Also, our model domain is the whole ETSP, encompassing the OMZ and the adjacent ocean, whose size is \approx 20 times the area of the OMZ.

Specifically, [Codispoti, 2007] reported a significant imbalance of the global nitrogen budget, which was mainly due to high rates of benthic denitrification, but the nitrogen inputs exceed the losses if benthic denitrification is excluded from his analysis. Kalvelage et al. [2013] only estimated the N budget of the actual OMZ. The OMZ is represented in our model by the UM box, which is indeed a strong NO₃⁻ sink. The point of our statement is to clarify the different roles of the water-column and the benthic for the fixed-N budget of the model domain. We reformulate this sentence on Page 2, Lines 20–22, slightly to make it easier to understand.

Comment 7: Page 14444, Line 1: Noffke et al., Limnol. Oceanograph., 57, 851–867, 2012, who estimated benthic Fe and P fluxes in the ETSP, should be cited here.

Response 7: The reference is added on Page 3, Line 28 of the revised ms.

Comment 8: Page 14444, Lines 10–14: First, I found this sentence a bit confusing to read. Please rephrase. Second, dissimilative nitrate reduction to ammonium could also be a source of NH₄⁺ for anammox, as claimed in Lam et al., 2009, i.e., not all NH₄⁺ is necessarily derived from organic matter oxidation in the water-column. How this would affect their water-column estimate of N-loss in their model?

Response 8: We agree with the reviewer that NH₄⁺ for anammox could be from either organic matter oxidation or dissimilatory nitrate reduction to ammonium (DNRA). Nevertheless, DNRA is also heterotrophic and thus organic matter is also required for DNRA [Lam et al., 2009, Koeve and Kähler, 2010]. We rephrase these sentences to clarify this point in the revised ms on Page 4, Lines 8–13:

Anammox has been observed to be a major fixed-N loss process in the eastern tropical South Pacific (ETSP) in recent studies [Lam et al., 2009, Kalvelage et al., 2013]. However, the essential substrates for anammox are ultimately provided by heterotrophic processes

[Koeve and Kähler, 2010, Kalvelage et al., 2013], such as organic-matter remineralisation or dissimilatory nitrate reduction to ammonium (DNRA). Thus, both denitrification and anammox are hence driven by the flux of organic matter into the OMZ.

Comment 9: Page 14444, Lines 25–26: Perhaps also cite Kim et al.: Increasing anthropogenic nitrogen in the North Pacific Ocean. *Science*, 346, 1102–1106, 2014.

Response 9: We thank the reviewer for pointing out this reference. This reference is added on Page 4, Line 25 of the revised ms.

Comment 10: Page 14445, Lines 5–8: I think DON should be considered in their model, with different scenarios regarding bioavailability, since it can represent a significant fraction of total atmospheric N depositions. See my general comments above.

Response 10: We have answered this question together with the response to the general question about DON in the atmospheric N deposition. Please refer to **Response 3** above.

Comment 11: Page 14445, Lines 16–17: I think they should cite published studies that quantify N riverine inputs in the Pacific Ocean, and if possible, the ETSP. For example, Seitzinger and Kroeze (Global Biogeochemical Cycles, 1998) reported a value of 4 Tg N yr⁻¹ for the Pacific Ocean.

Response 11: No major rivers flow into the ETSP from the coast of Peru. According to Seitzinger and Kroeze [1998], riverine DIN input into the ETSP only accounts for less than 1.5% of the total nitrogen riverine input into the Pacific and increases the NO_3^- concentration of our study area for less than $2.0 \times 10^{-6} \mu\text{mol N L}^{-1}$ if we assume that all the riverine DIN is distributed evenly, which indicates a very minor role in the nitrogen budget of the ETSP. Therefore, riverine nitrogen input is excluded from our model analysis. We will rephrase the statement on Page 14445, Lines 13–17 of the BGD ms to:

Using a conceptually simple and computationally efficient box model, we here attempt a synthesis considering all essential sources and sinks and their mutual interactions, with the only exception of riverine input, which is excluded from our model analysis because it contributes only negligibly to the nitrogen inventory in the ETSP [Seitzinger and Kroeze, 1998].

on Page 5, Lines 12–15 in the revised ms.

Comment 12: Overall: It is a bit unclear to me how N₂ fixation is modeled. Maybe add a short section giving more detail about this?

Response 12: To provide more details about how we model N₂ fixation, the following sentences are added on Page 6, Lines 8–17 of the revised ms to replace the content on Page 14446, Lines 9–12 of the BGD ms:

We represent two phytoplankton types in the biogeochemical model, ordinary phytoplankton (Phy) and nitrogen fixers (NF) as defined in Su et al. [2015]. Both Phy and NF concentrations are determined by the steady-state balance between net primary production (NPP) and mortality (M) respectively in the U and S boxes. Phy requires both phosphate and nitrate, and growth of ordinary phytoplankton is described by a Blackman-type dependence on the nitrate and phosphate limitation terms. NF can fix N₂ as long as PO₄³⁻ is available. A quadratic mortality term is adopted for both Phy and NF, considering possible viral lysis, phytoplankton aggregation, or a feedback between zooplankton grazing and phytoplankton concentration. N₂ fixers are given a lower maximum growth rate, which is 1/3 of the maximum growth rate of ordinary phytoplankton, in order to account for the high cost of nitrogen fixation [La Roche and Breitbarth, 2005].

Comment 13: Page 14445, Line 23: It is unclear to me, and maybe to other non-modelers, why they calibrated their physical parameters to fit “the average $\delta^{14}\text{C}$ of each box”. Perhaps clarify?

Response 13: To clarify why the average $\delta^{14}\text{C}$ of each box is applied to calibrate the physical parameters of the model, the following sentences are added on Page 5, Lines 20–24 of the revised ms to replace the sentence on Page 14445, Lines 22–24 of the BGD ms:

Briefly, the physical parameters were calibrated to fit the average $\delta^{14}\text{C}$ [Key et al., 2004] of each box and biogeochemical parameters are constrained by literature data. $\delta^{14}\text{C}$ is the ^{13}C fractionation-corrected ratio of $^{14}\text{C}/^{12}\text{C}$, which is commonly used in ocean modelling to evaluate and calibrate model physics, because it tends to cancel the effect of the biotic downward transport of ^{14}C with the rain of organic particles produced by marine organisms.

Comment 14: Page 14446: Lines 2–3: They separated their model into coastal upwelling region and open ocean, but their model results are then merged for the two regions in Figures 2 and 4. I think it would be helpful to distinguish between these two different regions in Figures and in the discussion and conclusions section.

Response 14: We thank the review for the constructive comments. We have modified all the figures, model results and discussion in the revised ms to distinguish between the two different regions. Please see the Figs. 2 and 4 in the revised ms, and supplementary Fig. S1.

Comment 15: Page 14447, Line 6: Are there any uncertainties associated with these estimates of N deposition rates? If so, I think these should also be reflected in their modeled fluxes.

Response 15: The data applied in the BGD ms are from a chemistry-climate model [Lamarque et al., 2011], and it is not possible for us to estimate the uncertainties underlying these model results. Instead, we apply an inorganic nitrogen-deposition estimate by Dentener et al. [2006], which is the average of 23 atmospheric chemistry transport models, and investigate the impact of uncertainties on our model results. We include these results in the new Fig. 5 in the revised ms. The model description is included on Page 11, Lines 13–18 of the revised ms. The results are presented and discussed on Page 16, Line 22–Page 17, Line 2 and Page 19, Lines 18–22, respectively, of the revised ms.

Comment 16: Page 14447, Line 10–15: It is a bit unclear what they wish to communicate in this paragraph. I suppose that they want to point out that DIP depositions are low, thus justifying neglecting it in their model. I suggest rewriting this paragraph to expose this point more clearly.

Response 16: We rephrase this paragraph to read

Atmospheric phosphorus deposition is excluded from our analysis because its amount is much smaller than the Redfield equivalent of nitrogen atmospheric deposition [Duce et al., 1991]. This results in N/P (mole/mole) ratios of more than 100, much higher than the average elemental N/P ratio required by phytoplankton [Duce et al., 2008, Mahowald et al., 2008].

on Page 7, Line 25–Page 8, Line 2 of the revised ms.

Comment 17: Page 14448, equation 2: Katsev and Crowe, Geology, 43(7), 2015 (doi: 10.1130/G36626.1) recently suggested a correction to the power law of remineralization under anoxic conditions. How this correction would affect their results?

Response 17: Unfortunately, we cannot include the power-law correction of remineralization under suboxic conditions of Katsev and Crowe [2015], because it would require resolving the time course

of particle export and remineralisation. However, our analysis does not resolve this short time scale. Nevertheless, we would like to point out that the variability in the Martin-curve exponent b implied by Katsev and Crowe [2015] is well within the range of b values reported by Guidi et al. [2015]. Variations in b play only a very minor role in our model, which we report on Page 18, Lines 9–12 of the revised ms and in supplementary Figs. S7 and . S8.

Comment 18: Page 14449, Line 1: “Martin-curve values” refer to the second part of equation 2 only, as EP_U and EP_S represents the export production (F). Perhaps clarify?

Response 18: We rephrased Page 14449, Lines 1–3 in the BGD ms to

where EP_U and EP_U+EP_S represent the export production (F in Eq. 2) in the upwelling region and the whole model domain, respectively; AMC_{UM} and AMC_D ($(\frac{z}{100})^{-b}$ in Eq. 2) are the average Martin-curve values corresponding to the actual water depth (z) in the ETOPO2 data; SD_{UM} and SD_D represent the percentages in contact with the sediment in the UM and D boxes, respectively (Table 2).

on Page 9, Lines 10–14 in the revised ms.

Comment 19: Page 14449, section 2. 4. 2: Their data-based estimate of benthic denitrification is derived from primary production estimates from satellite data. I would like them to also use other more direct ship-based measurements of primary productivity for the area or at least discuss how the two compare. In this respect, see review by Pennington et al., Progress in Oceanography, 69, 285–317, 2006.

Response 19: We now discuss these processes also with regard to ship-collected data from Pennington et al. [2006] on Page 19, Line 25–Page 20, Line 13 in the revised ms.

Comment 20: Page 14452, Line 11: Again, what is the error associated with this N deposition estimate?

Response 20: Please refer to **Response 15** of this response letter.

Comment 21: Page 14453, Line 13: Is that local or global NO₃[−] inventories? Please clarify.

Response 21: These are the NO₃[−] inventories of the UM and D boxes, respectively. The sentence is modified to

Due to the small sediment-area percentages, the annual nitrogen loss by benthic denitrification is 0.17 and 0.82 Tg N yr^{−1} in the UM and D boxes, accounting for only about 0.14 and 0.0051 % year^{−1}, respectively, of the NO₃[−] inventories in these boxes (Table 2).

on Page 14, Lines 6–9 of the revised ms.

Comment 22: Page 14453, Line 4, Page 14454, Line 2 and Page 14455, lines 1–3: Can these results be included as Supplementary Materials?

Response 22: We now provide Figs. S2, S3, and S4 in the supplementary material of the revised ms.

Comment 23: Page 14457, Line 2: What about N₂ fixation limitation by Fe? See my general comments above.

Response 23: Please see our **Response 2** above in this response letter.

Comment 24: Page 14457, lines 7–10: Should also include a model scenario with correspondingly higher future N depositions

Response 24: The results for a future N deposition scenario are explained in **Response 3** of this response letter.

Comment 25: Page 14459, lines 5–8: This is essentially the same sentence as in the introduction. See my comment for Page 14444, Lines 10–14.

Response 25: We remove this sentence from the revised ms and rephrase the sentences on Page 14459, Lines 9–13 to:

Even though water-column denitrification has been considered the major fixed-N loss processes for simplicity, the stimulatory effects between nitrogen fixation and fixed-N loss, and phosphorus regeneration and fixed-N loss still apply even if anammox replaced water-column denitrification as the fixed-N loss pathway. Thus, the nitrogen-balancing mechanism in the ETSP should not depend on whether the fixed-N is lost through denitrification or anammox.

This will be added on Page 22, Lines 18–23 of the revised ms.

Comment 26: Page 14459, lines 16–24: I think the fact that many other models and observational results found that the ETSP is a NO_3^- sink might rather indicate that their model is inaccurate. Again, if the ETSP was a NO_3^- source, we would not observe large N deficits (see my previous comment P. 14442, line 20).

Response 26: We do not state that the ETSP is a NO_3^- source, but rather that the water column of our model domain including both the upwelling region and the vast open ocean is NO_3^- source. Please refer to **Response 6** of this response letter.

Comment 27: I find that the discussion/conclusion section ends rather abruptly. I recommend adding a short summary paragraph, including the major implications of their findings.

Response 27: A Summary section is added on Pages 23–24 of the revised ms:

The influence of atmospheric nitrogen deposition and benthic remineralization on the nitrogen budget of the ETSP is investigated with a conceptually simple and computationally efficient box model. Additional nitrogen input by atmospheric nitrogen deposition is offset by about 48 % by reduced nitrogen fixation with the remainder being transported horizontally out of the model domain, irrespective of uncertainties underlying the actual rate of nitrogen deposition and bioavailability of deposited DON. All our data- and model-based estimations for benthic remineralization are comparable with those of previous studies based on both models and observations. Modeled responses to these fluxes indicate stabilizing feedbacks, which tend to balance the nitrogen inventory. Variations in these fluxes due to variations in NPP, aphotic nitrogen fixation and organic matter remineralization under different oxygen conditions do not change these feedbacks. In the high-BD configuration, our model domain turns into a NO_3^- sink, which indicates that the whole tropical South Pacific, including the upwelling region, the open ocean, and the sediments, might be a NO_3^- sink. More research is needed to better constrain the fluxes of nitrogen and phosphorus, including the amount of bioavailable nitrogen deposition, nitrogen fixation and benthic denitrification, to allow a more reliable estimation of the nitrogen budget of the ETSP.

Comment 28: Table 1. I found this table rather confusing. The terms are defined both in the legend and in the upper part of the table. I suggest defining all terms in the legend and only including the lower part of the table, explaining the different model configurations (e.g., Syn1, Syn2, Syn3 and Syn4 in different columns and MBD, MPR, DBD, DPR, and N-DEP in rows).

Response 28: We agree that the Table 1 in the original BGD ms was confusing. MBD, MPR, DBD, N-DEP, Syn1, Syn2, Syn3 and Syn4 are all model configuration names, so we can not separate them into different rows and columns. However, we modify the caption of the table to improve its clarity. Please see Table 1 in the revised ms.

Comment 29: Tables 2 and 3. Another more comprehensive table summarizing all data based estimates (e.g. nitrogen deposition, N₂-fixation, benthic denitrification and phosphate regeneration) used in their model as well as references for these data would be useful.

Response 29: We thank the reviewer for this suggestion. N₂-fixation is a variable of our model, therefore we do not combine it together with the data-based estimates (nitrogen deposition, benthic denitrification, phosphorus regeneration). We combine Tables 2 and 3 of the BGD ms, and all the data-based estimates as well as their references in one table (Table 2 in the revised ms).

Comment 30: Aesthetic detail: Why is the space between N-fix and WC-denif in legend in Figures 2, 4 and 8 so large?

Response 30: This is now corrected in all figures in the revised ms.

Comment 31: Page 14456, Line 15: I would remove “below the water column” and change the sentence to: “...reaching the sea floor under suboxic conditions...”

Response 31: This is changed on Page 18, Line 13 of the revised ms.

Comment 32: Page 14459, Line 29: I would change to: “Based on our findings...”

Response 32: This is changed on Page 23, Lines 7–8 of the revised ms.

References

W. M. Berelson. The flux of particulate organic carbon into the ocean interior: A comparison of four U. S. JGOFS regional studies. *Oceanography*, 14(4):59–67, 2001.

S. Bonnet, J. Dekaezemacker, K. A. Turk-Kubo, T. Moutin, R. M. Hamersley, O. Grosso, J. P. Zehr, and D. G. Capone. Aphotic N₂ fixation in the eastern tropical south pacific ocean. *PLoS one*, 8 (e81265):1–14, December 2013. doi: 10.1371/journal.pone.0081265.

L. Codispoti. An oceanic fixed nitrogen sink exceeding 400 $\text{tg n } a^{-1}$ vs the concept of homeostasis in the fixed-nitrogen inventory. *Biogeosciences*, 4:233–253, May 2007. doi: 10.5194/bg-4-233-2007.

S. Cornell, T. Jickells, J. Cape, A. Rowland, and R. Duce. Organic nitrogen deposition on land and coastal environments: a review of methods and data. *Atmos. Environ.*, 37:2173–2191, 2003. doi: 10.1016/S1352-2310(03)00133-X.

J. Dekaezemacker, S. Bonnet, O. Grosso, T. Moutin, M. Bressac, and D. Capone. Evidence of active dinitrogen fixation in surface waters of the eastern tropical south pacific during el niño and la niña events and evaluation of its potential nutrient controls. *Global Biogeochem. Cycles*, 27:768–779, Nov. 2013. doi: 10.1002/gbc.20063.

F. Dentener, J. Drevet, J. F. Lamarque, I. Bey, B. Eickhout, A. M. Fiore, D. Hauglustaine, L. W. Horowitz, M. Krol, U. C. Kulshrestha, M. Lawrence, C. Galy-Lacaux, S. Rast, D. Shindell, D. Stevenson, T. V. Noije, C. Atherton, N. Bell, D. Bergman, T. Butler, J. Cofala, B. Collins, R. Doherty, K. Ellingsen, J. Galloway, M. Gauss, V. Montanaro, J. F. Muller, G. Pitari, J. Rodriguez, M. Sanderson, F. Solmon, S. Strahan, M. Schultz, K. Sudo, S. Szopa, and O. Wild25. Nitrogen and sulfur deposition on regional and global scales: A multimodel evaluation. *Global Biogeochem. Cycles*, 20(GB4003):1–21, Oct. 2006. doi: 10.1029/2005GB002672.

R. Duce, P.S.Liss, J. Merrill, E. Atlas, P. Buat-Menard, B. B. Hicks, J. Miller, J.M.Prospero, R. . Arimoto, T. Church, W. Ellis, J. alloway, L. . Hansen, T. Jickells, A. H. Knap, K. Reinhardt, B. Schneider, A. Soudine, J. J. Tokos, S. Tsunogai, R. . Wollast, and M. Zhou. The atmospheric input of trace species to the world ocean. *Global Biogeochem. Cycles*, 5(3):193–259, Sep. 1991.

R. A. Duce, J. L. Roche, K. Altieri, K. R. Arrigo, A. R. Baker, D. G. Capone, S. Cornell, F. Dentener, J. Galloway, R. S. Ganeshram, R. J. Geider, T. Jickells, M. M. Kuypers, R. Langlois, P. S. Liss, S. M. Liu, J. J. Middelburg, C. M. Moore, S. Nickovic, A. Oschlies, T. Pedersen, J. Prospero, R. Schlitzer, S. Seitzinger, L. L. Sorensen, M. Uematsu, O. Ulloa, M. Voss, B. Ward, and L. Zamora. Impacts of atmospheric anthropogenic nitrogen on the open ocean. *Science*, 320(5878):893–897, May 2008.

L. Guidi, L. Legendre, G. Reygondeau, J. Uitz, L. Stemmann, and S. A. Henson. A new look at ocean carbon remineralization for estimating deepwater sequestration. *Global Biogeochem. Cycles*, 29:1044–1059, July 2015. doi: 10.1002/2014GB005063.

T. Kalvelage, G. Lavik, P. Lam, S. Contreras, L. Arteaga, C. R. Löscher, A. Oschlies, A. lien Paulmier, L. Stramma, and M. M. M. Kuypers. Nitrogen cycling driven by organic matter export in the South Pacific oxygen minimum zone. *Nature Geosci.*, 6:228–234, Mar. 2013. doi: 10.1038/NGEO1739.

M. Kanakidou, R. A. Duce, J. M. Prospero, A. R. Baker, C. Benitez-Nelson, F. J. Dentener, K. A. Hunter, P. S. Liss, N. Mahowald, G. S. Okin, M. Sarin, K. Tsigaridis, M. Uematsu, L. M. Zamora, and T. Zhu. Atmospheric fluxes of organic n and p to the global ocean. *Global Biogeochem. Cycles*, 26(GB3026):1–12, 2012. doi: 10.1029/2011GB004277.

S. Katsev and S. A. Crowe. Organic carbon burial efficiencies in sediments: The power law of mineralization revisited. *geology*, 43(7):607–610, May 2015. doi: 10.1130/G36626.1.

R. M. Key, A. Kozyr, C. L. Sabine, K. Lee, R. Wanninkhof, J. L. Bullister, R. A. Feely, F. J. Millero, C. Mordy, and T.-H. Peng. A global ocean carbon climatology: Results from Global Data Analysis Project (GLODAP). *Global Biogeochem. Cycles*, 18(GB4031):1–23, Dec. 2004. doi: 10.1029/2004GB002247.

W. Koeve and P. Kähler. Heterotrophic denitrification vs. autotrophic anammox – quantifying collateral effects on the oceanic carbon cycle. *Biogeosciences*, 7:2327–2337, Aug. 2010. doi: 10.5194/bg-7-2327-2010.

J. La Roche and E. Breitbarth. Importance of the diazotrophs as a source of new nitrogen in the ocean. *J. Sea Res.*, 53:67–91, 2005. doi: 10.1016/j.seares.2004.05.005.

P. Lam, G. Lavika, M. M. Jensen, J. van de Vossenberg, M. Schmidb, D. Woebken, D. Gutiérrez, R. Amanna, M. S. M. Jettenb, and M. M. M. Kuypers. Revising the nitrogen cycle in the Peruvian oxygen minimum zone. *Proc. Natl. Acad. Sci. USA*, 106(12):4752–4757, Mar. 2009. doi: 10.1073/pnas.0812444106.

J.-F. Lamarque, G. P. Kyle, M. Meinshausen, K. Riahi, S. J. Smith, D. P. van Vuuren, A. J. Conley, and F. Vitt. Global and regional evolution of short-lived radiatively-active gases and aerosols in the Representative Concentration Pathways. *Climatic Change*, 109:191–212, 2011. doi: 10.1007/s10584-011-0155-0.

J.-F. Lamarque, F. Dentener, J. McConnell, C.-U. Ro, M. Shaw, R. Vet, D. Bergmann, P. Cameron-Smith, S. Dalsoren, R. Doherty, G. Faluvegi, S. J. Ghan, B. Josse, Y. H. Lee, I. A. MacKenzie, D. Plummer, D. T. Shindell, R. B. Skeie, D. S. Stevenson, S. Strode, G. Zeng, M. Curran, D. Dahl-Jensen, S. Das, D. Fritzsche, and M. Nolan. Multi-model mean nitrogen and sulfur deposition from the Atmospheric Chemistry and Climate Model Intercomparison Project (ACCMIP): evaluation of historical and projected future changes. *Atmos. Chem. Phys.*, 13:7997–8018, Aug. 2013. doi: 10.5194/acp-13-7997-2013.

N. Mahowald, T. D. Jickells, A. R. Baker, P. Artaxo, C. R. Benitez-Nelson, G. Bergametti, T. C. Bond, Y. Chen, D. D. Cohen, B. Herut, N. Kubilay, R. Losno, C. Luo, W. Maenhaut, K. A. McGee, G. S. Okin, R. L. Siebert, and S. Tsukuda. Global distribution of atmospheric phosphorus sources, concentrations and deposition rates, and anthropogenic impacts. *Global Biogeochem. Cycles*, 22 (GB4026):1–19, Dec. 2008. doi: 10.1029/2008GB003240.

H. W. Paerl and D. R. Whitall. Anthropogenically-derived atmospheric nitrogen deposition, marine eutrophication and harmful algal bloom expansion: Is there a link? *Ambio*, 28:307–311, 1999.

J. T. Pennington, K. L. Mahoney, V. S. Kuwahara, D. D. Kolber, R. Calienes, and F. P. Chavez. Primary production in the eastern tropical Pacific: A review. *Prog. Oceanogr.*, 69:285–317, Mar. 2006.

S. P. Seitzinger and C. Kroeze. Global distribution of nitrous oxide production and N inputs in freshwater and coastal marine ecosystems. *Global Biogeochem. Cycles*, 12(1):93–113, March 1998. doi: 10.1029/97GB03657.

B. Su, M. Pahlow, H. Wagner, and A. Oschlies. What prevents nitrogen depletion in the oxygen minimum zone of the eastern tropical South Pacific? *Biogeosciences*, 12:1113–1130, Feb. 2015. doi: 10.5194/bg-12-1113-2015.

B. A. S. Van Mooy, R. G. Keil, and A. H. Devol. Impact of suboxia on sinking particulate organic carbon: Enhanced carbon flux and preferential degradation of amino acids via denitrification. *Geochim. Cosmochim. Acta*, 66(3):457–467, 2002. doi: 10.1016/S0016-7037(01)00787-6.

R. Vet, R. S. Artz, S. Carou, M. Shawa, C.-U. Ro, W. Aas, A. Baker, V. C. Bowersox, F. Dentener, C. Galy-Lacaux, A. Hou, J. J. Pienaar, R. Gillett, M. C. Forti, S. Gromov, H. Hara, T. Khodzherm, N. M. Mahowald, S. Nickovic, P. Rao, and N. W. Rei. A global assessment of precipitation chemistry and deposition of sulfur, nitrogen, sea salt, base cations, organic acids, acidity and ph, and phosphorus. *Atmos. Environ.*, 93:3–100, Aug. 2014. doi: 10.1016/j.atmosenv.2013.10.060.

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Box-modeling of the impacts of atmospheric nitrogen deposition and benthic remineralization on the nitrogen cycle of the eastern tropical South Pacific

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Abstract

Both atmospheric deposition and benthic remineralization influence the marine nitrogen cycle, and hence ultimately also marine primary production. The biological and biogeochemical relations ~~of in~~ the eastern tropical South Pacific (ETSP) ~~to among~~ nitrogen deposition, benthic denitrification and ~~phosphate phosphorus~~ regeneration are analysed in a prognostic box model of the oxygen, nitrogen and phosphorus cycles in the ETSP. ~~In the model, atmospheric nitrogen deposition-based-on estimates~~ Atmospheric nitrogen deposition ($\approx 1.5 \text{ Tg N yr}^{-1}$ for the years 2000–2009) is offset by half ~~in the model~~ by reduced N_2 fixation, with the other half transported out of the model domain. ~~Both model-Model-~~ and data-based benthic denitrification ~~are found to in our model domain are responsible for losses of 0.19 and 1.0 Tg N yr^{-1} , respectively, and both~~ trigger nitrogen fixation, partly compensating for the NO_3^- loss. ~~Model- and data-based estimates of enhanced phosphate release via sedimentary phosphorus regeneration under suboxic conditions are 0.062 and 0.11 Tg P yr^{-1} , respectively.~~ Since phosphate is the ultimate limiting nutrient in the model, ~~enhanced sedimentary phosphate regeneration under suboxic conditions stimulates even very small additional phosphate inputs stimulate~~ primary production and subsequent export production and NO_3^- loss in the oxygen minimum zone (OMZ). A sensitivity analysis of the local response to both atmospheric deposition and benthic remineralization indicates dominant stabilizing feedbacks in the ETSP, which tend to keep a balanced nitrogen inventory, i.e., nitrogen input by atmospheric deposition is counteracted by decreasing nitrogen fixation; NO_3^- loss via benthic denitrification is partly compensated by increased nitrogen fixation; enhanced nitrogen fixation stimulated by phosphate regeneration is partly ~~removed by the~~ ~~counteracted by~~ stronger water-column denitrification. Even though the water column in our model domain acts as a NO_3^- source, the ETSP including benthic denitrification might ~~become be~~ a NO_3^- sink.

1 Introduction

Marine primary production (PP) by phytoplankton is a key factor controlling the strength of the oceanic biological carbon pump and the amount of CO_2 that is stored in the ocean (Gruber, 2004; Okin et al., 2011). PP is controlled by light and nutrients, such as nitrogen, phosphorus or iron, necessary for the production of phytoplankton. These nutrients are supplied to the light-lit surface waters by upwelling, turbulent entrainment of subsurface water, riverine inputs, biological nitrogen fixation, atmospheric deposition and benthic remineralisation (Falkowski et al., 1998; Kasai et al., 2002; Duce et al., 2008; Bakun and Weeks, 2008; Moore and Braucher, 2008).

Nitrogen is often the limiting nutrient for phytoplankton in the ocean (Moore et al., 2013). On the other hand, oceanic nitrogen is thought to adjust, via nitrogen gain and loss processes, to the marine phosphorus inventory on geological time scales, making phosphorus the ultimate limiting nutrient and nitrogen the proximate limiting nutrient (Tyrrell, 1999). The ocean's nitrogen inventory has a turnover time of a few thousand years, being affected by relatively large interacting nitrogen sinks and sources. The exact mechanisms and timescales of the interactions are not well understood. Estimates of oceanic nitrogen fixation, the main fixed-N source into the ocean, vary from 106 to 330 Tg N yr^{-1} based on both in-situ observations and models (Codispoti et al., 2001; Brandes and Devol, 2002; Gruber and Sarmiento, 2002; Gruber, 2004; Großkopf et al., 2012). Water-column denitrification and anaerobic ammonium oxidation (anammox) in oxygen minimum zones (OMZs), accounting for 100–300 Tg N yr^{-1} , and benthic denitrification, estimated as 95–300 Tg N yr^{-1} , mainly determine the oceanic fixed-N sink (Gruber and Sarmiento, 2002; Gruber, 2004; Codispoti, 2007; Bohlen et al., 2012; Eugster and Gruber, 2012; DeVries et al., 2012). Due to the large uncertainty uncertainties in the major sources and sinks of the global nitrogen cycle, the balance of the nitrogen inventory in the ocean is still a matter of debate (Gruber, 2004; Codispoti, 2007; DeVries et al., 2012).

Phosphate can be the ultimate limiting nutrient on geological time scales even in regions with fixed nitrogen deficits with respect to the Redfield-equivalent of the phosphate concentration (Tyrrell, 1999; Su et al., 2015; Augu  res and Loreau, 2015). The ocean's phosphorus

budget has been suggested to be unbalanced in the modern ocean with sedimentary burial as the major sink exceeding phosphorus sources (Wallmann, 2010). This condition might be alleviated by benthic ~~phosphate~~phosphorus regeneration, which can be enhanced under low-oxygen bottom waters ($O_2 < 20 \mu\text{mol L}^{-1}$) (Slomp and Van Cappellen, 2007; Wallmann, 2010)

5 **(Slomp and Van Cappellen, 2007; Wallmann, 2010; Noffke et al., 2012)**. Input of bioavailable phosphorus into the ocean stimulates primary production, and decomposition of subsequent export production enhances O₂ consumption in the ocean, in turn increasing the volume of oceanic oxygen-deficit water and the fixed-N loss. Consequently, **phosphate-phosphorus** regeneration is expected to be enhanced by enlarging OMZs, possibly leading to a positive feedback 10 loop (Van Cappellen and Ingall, 1994; Wallmann, 2003).

OMZs also play an important role in the global marine fixed-N budget as they are responsible for a large fraction of total marine fixed-N loss (Canfield, 2006). The relative contribution of heterotrophic denitrification and autotrophic anammox to the total oceanic fixed-nitrogen sink remains debated (Lam et al., 2009; Ward et al., 2009). ~~Even though anammox~~ Anammox has been observed to be a ~~major~~ fixed-N loss process in the eastern tropical South Pacific (ETSP) (Lam et al., 2009; Kalvelage et al., 2013). However, the essential substrates for anammox are ultimately provided by heterotrophic processes (Koeve and Kähler, 2010; Kalvelage et al., 2013), such as organic-matter remineralisation or dissimilatory nitrate reduction to ammonium (DNRA). Thus, both denitrification and anammox are ~~ultimately~~ driven by the flux of organic matter into the OMZ(~~Koeve and Kähler, 2010; Kalvelage et al., 2013~~). For simplicity, heterotrophic denitrification is considered the major fixed-N loss process in the present study. Continental shelves and the upper continental slopes are the most important sites for benthic fixed-N loss (Christensen et al., 1987; Devol, 1991). However, Bohlen et al. (2011) found that the continental shelf and upper continental slope of the ETSP across a section at 11° S are sites of nitrogen recycling rather than fixed-N loss, because of relatively low rates of denitrification and high rates of NH_4^+ release from Dissimilatory Nitrate Reduction to Ammonium (DNRA). This illustrates that the NH_4^+ released from DNRA should be taken into account when the benthic fixed-N sink is estimated.

In the last few decades, a number of ~~model and data-based model- and data-based~~ investigations have been carried out on the importance of atmospheric fixed-N input into the ocean for marine biogeochemical cycles (Duce, 1986; Duce et al., 1991; Krishnamurthy et al., 2007, 2010; Okin et al., 2011; Mouriño-Ca
5 Duee et al. (2008) indicate (Duce, 1986; Duce et al., 1991; Krishnamurthy et al., 2007, 2010; Okin et al., 2011; Mouriño-Ca
Duce et al. (2008) suggest that anthropogenic nitrogen deposition is rapidly approaching ~~the global oceanic estimates for estimates for global oceanic~~ N_2 fixation, while preindustrial deposition was an order of magnitude lower ~~than~~ fixation. However, the response of nitrogen-fixation and denitrification to atmospheric nitrogen deposition remains an open question. Atmospheric nitrogen inputs into the global ocean are dominated by inorganic nitrogen from anthropogenic sources (Warneck, 1988; Paerl and Whitall, 1999). The exact magnitude of organic nitrogen deposition ~~is not clear and its bioavailability is still under appraisement~~ due to a lack of ~~observations~~ (Cornell et al., 2003; Duce et al., 2008; Zamora et al., 2011). ~~Thus, the contribution of DON to enough observations (Duce et al., 2008; Zamora et al., 2011). Therefore, we apply the finding of Cornell et al. (2003); Kanakidou et al. (2012) that DON accounts for 30% of total nitrogen deposition is still uncertain and the distribution, bioavailability and lifetime are also not clear (Seitzinger and Sanders, 1999; Duarte et al., 2006; Duce et al., 2008). Therefore, DON deposition is excluded from our analysis, in our model, and investigate its role on the nitrogen budget of the ETSP with the bioavailability measured by Paerl and Whitall (1999). Several scenarios with different DON bioavailability are assessed to analyse uncertainties regarding the bioavailability of DON~~

Various biogeochemical models have addressed the effects and feedbacks between the major sources and sinks in the marine nitrogen cycle (Van Cappellen and Ingall, 1994; Deutsch et al., 2001, 2007; Krishnamurthy et al., 2007; Somes et al., 2013; Landolfi et al., 2013). However, most of them have explored only a subset of the atmospheric, pelagic and benthic nitrogen sources and sinks. Using a conceptually simple and computationally efficient box model, we here attempt a synthesis considering all essential sources and sinks and their mutual interac-

tions, with the only exception of riverine input, which is excluded from our model analysis because ~~of its unknown and presumably minor role it contributes negligibly to the nitrogen inventory~~ in the ETSP (Seitzinger and Kroeze, 1998).

2 Model description

5 2.1 Circulation and biogeochemical model

The circulation model is the same as in Su et al. (2015), which is a prognostic 5-box model to explore the interactions among oceanic circulation, nitrogen fixation and water-column denitrification in the OMZ of the ETSP. Briefly, the physical parameters were calibrated to fit the average $\delta^{14}\text{C}$ (~~Key et al., 2004~~) of each box and biogeochemical parameters are constrained by literature data. $\delta^{14}\text{C}$ is the ^{13}C fractionation-corrected ratio of $^{14}\text{C}/^{12}\text{C}$, which is commonly used in ocean modelling to evaluate and calibrate model physics, because it tends to cancel the effect of the biotic downward transport of ^{14}C with the rain of organic particles produced by marine organisms. All the simulations in this manuscript employ the Open-boundary + Reduced-denitrification (OBRD) configuration of Su et al. (2015), which allows for exchange of deep and intermediate ETSP waters with the Southern Ocean (“SO” in Fig. 1) and applies reduced remineralisation rates under suboxic conditions. The model domain consists of five boxes representing the water column of an upwelling region and an adjacent ocean basin. The U box represents the upper upwelling region. The UM box is the OMZ below, where suboxia is expected to develop. The S box represents the surface ocean away from the upwelling zone. Below the S box sits the I box, which represents water of intermediate depth and exchanges water with UM. D is the deep box, which represents water deeper than 500 m (model configuration shown in Fig. 1).

~~There are~~ We represent two phytoplankton types in the biogeochemical model, ordinary phytoplankton, ~~Phy~~, (~~Phy~~) and nitrogen fixers, ~~NF~~, (~~NF~~) as defined in Su et al. (2015). Both Phy and NF require phosphate, whereas nitrate is required in addition to phosphate only by Phy, and NF concentrations are determined by the steady-state balance between net primary

production (NPP) and mortality (M) respectively in the U and S boxes. Phy requires both phosphate and nitrate, and growth of Phy is described by a Blackman-type dependence on the nitrate and phosphate limitation terms. NF can fix N_2 as long as PO_4^{3-} is available. A quadratic mortality term is adopted for both Phy and NF, considering possible viral lysis, phytoplankton aggregation, or a feedback between zooplankton grazing and phytoplankton concentration. N_2 fixers are given a lower maximum growth rate, which is 1/3 of the maximum growth rate of ordinary phytoplankton, in order to account for the high cost of nitrogen fixation (La Roche and Breitbarth, 2005).

Dead phytoplankton is immediately remineralized in the surface layer and underlying boxes according to the pre-defined remineralization fractions. Remineralization occurs preferentially via aerobic respiration, with anaerobic denitrification and the associated nitrogen loss setting in only when all O_2 has been consumed by aerobic respiration. When oxygen is exhausted in the OMZ, remineralization is assumed to slow down by a factor of 5, and accordingly denitrification within the UM box is responsible for 1/5 of the remaining organic matter remineralization and the remainder will be remineralized in the D box.

In order to represent the nitrogen and phosphate fluxes across the water-sediment interface, remineralization of particulate organic carbon reaching the sediment (POC rain rate, RRPOC) is included additionally in the UM and D boxes. RRPOC is calculated according to the method introduced in [Section 2.4](#), and we assume that all the POC is buried in the sediment.

20 2.2 Model configurations

The above descriptions define the control configuration. In order to investigate the model sensitivity to atmospheric nitrogen deposition and benthic remineralization, we employ another nine model configurations incorporating either a subset or all of these processes, which are summarized in Table 1.

25 In the NDEP configuration, atmospheric nitrogen input into the surface ocean according to the estimate by Lamarque et al. (2011) is included; MBD and DBD are configurations in which model- and data-based benthic denitrification rates are included in the control configuration; MPR and DPR represent configurations with model- and data-based benthic phosphorus

regeneration respectively. Detailed information of all processes is presented in Sects. 2.3, 2.4 and 2.5.

Nitrogen deposition, benthic denitrification and phosphate regeneration are integrated into the synthesis model configurations to explore the model sensitivity to each process and their mutual interactions in the ETSP. Synthesis configuration Syn1 includes model-based benthic denitrification and phosphorus regeneration; Syn2 includes the data-based benthic denitrification and phosphorus regeneration; Syn3 includes atmospheric deposition in addition to the processes in Syn1; Syn4 includes atmospheric deposition in addition to the processes in Syn2. The synthesis configurations Syn1 to Syn4 are summarized in Table 1.

10 2.3 Atmospheric nitrogen deposition

Years 2000–2009 levels of dry and wet inorganic nitrogen deposition following the RCP 4.5 scenario (Lamarque et al., 2011) are examined in our work. Inferred atmospheric inorganic nitrogen deposition rates are 0.081 and 1.4 Tg N yr⁻¹ (73.1 and 64.9 mg N m⁻² yr⁻¹) for the U and S box, respectively. Note that the circulation remains constant in our model, and only atmospheric nitrogen deposition fluxes are included as an additional annual nitrogen input into the surface (U and S) boxes.

Duce et al. (1991) estimated atmospheric dissolved inorganic phosphorus (DIP) input into the global ocean, which indicates high N/P (mole/Atmospheric phosphorus deposition is excluded from our analysis because its amount is much smaller than the Redfield equivalent of nitrogen atmospheric deposition (Duce et al., 1991). This results in N/P (mole/mole) ratios of more than 100 on a global scale (Dentener et al., 2006; Duce et al., 2008; Mahowald et al., 2008), much higher than the average elemental N/P N/P ratio required by phytoplankton in the ocean. Thus, we do not explore the influence of atmospheric phosphorus deposition in our analysis. (Duce et al., 2008; Mahowald et al., 2008).

2.4 Benthic denitrification

The empirical transfer function of Bohlen et al. (2012) is applied to predict benthic inorganic nitrogen loss (L_{DIN} in $\mu\text{mol N m}^{-2} \text{d}^{-1}$) through benthic denitrification, which can account for the net loss of dissolved inorganic nitrogen (DIN) from the sediment.

$$5 \quad L_{\text{DIN}} = (0.06 + 0.19 \cdot 0.99^{(\text{O}_2 - \text{NO}_3^-)_{\text{bw}}}) \cdot \text{RRPOC} \quad (1)$$

where NO_3^- and O_2 are bottom-water nitrate and oxygen concentrations in $\mu\text{mol kg}^{-1}$, and the rain rate (RRPOC) is in $\mu\text{mol C m}^{-2} \text{d}^{-1}$. Since the bottom-water NO_3^- and O_2 concentrations are well known in the ETSP, the uncertainty in our estimation of benthic denitrification 10 comes mostly from uncertainties in the rain rate, which, in turn, depends on biological production, as a function of phytoplankton biomass and its physiological status. Simulated phytoplankton concentrations in the surface boxes of the model roughly agree with estimates by Behrenfeld et al. (2005) from Aqua-Modis satellite data and the Redfield C:N ratio (U-box: 1.06 $\mu\text{mol N kg}^{-1}$ simulated vs. 0.68 $\mu\text{mol N kg}^{-1}$ from Aqua-Modis; S-Box: 0.23 $\mu\text{mol N kg}^{-1}$ 15 simulated vs. 0.28 $\mu\text{mol N kg}^{-1}$ from Aqua-Modis).

2.4.1 Model-based estimation of benthic denitrification

Fixed-N losses via benthic denitrification (L_{DIN}) in the UM and D boxes are obtained according to Eq. (1), with the respective simulated actual NO_3^- and O_2 concentrations taken as the bottom-water concentrations, and RRPOC is estimated from the export production out of the U and S 20 boxes (EP_U and EP_S) and the Martin curve (Eq. 2) (Martin et al., 1987):

$$\text{RRPOC} = F \cdot \left(\frac{z}{100} \right)^{-b} \quad (2)$$

where RRPOC is the rain rate, F is the export production from both surface boxes, and z is the water depth. The bathymetry of the regions of the UM and D boxes is derived 25 from ETOPO2 (http://www.ngdc.noaa.gov/mgg/gdas/gd_designagrid.html). We apply $b = 0.82$

in Eq. (2), which is the global average according to Berelson (2001) and also close to his estimate for the ETSP. An exponent of 0.4 for Eq. (2) in suboxic water is implied by Van Mooy et al. (2002). Therefore, sensitivity experiments are performed with $b = 0.4$. From Eq. (2) and the fraction of the lower boundary of the respective box in contact with the seafloor, the RRPOC at the sediment surfaces of the UM and D boxes is calculated according to Eqs. (3) and (4):

$$\text{RRPOC}_{\text{UM}} = \text{EP}_{\text{U}} \cdot \text{SD}_{\text{UM}} \cdot \text{AMC}_{\text{UM}} \quad (3)$$

$$\text{RRPOC}_{\text{D}} = (\text{EP}_{\text{U}} + \text{EP}_{\text{S}}) \cdot \text{SD}_{\text{D}} \cdot \text{AMC}_{\text{D}} \quad (4)$$

where EP_{U} and $\text{EP}_{\text{U}} + \text{EP}_{\text{S}}$ represent the export production (F in Eq. 2) in the upwelling region and the whole model domain, respectively; AMC_{UM} and AMC_{D} ($(\frac{z}{100})^{-b}$ in Eq. 2) are the average Martin-curve values corresponding to the actual water depth (z) in the grid of ETOPO2 respectively data; SD_{UM} and SD_{D} represent the percentages in contact with the sediment in the UM and D boxes, respectively (Table 2).

2.4.2 Data-based estimation of benthic denitrification

For a second and independent estimate of L_{DIN} , we combine observations from different datasets. O_2 and NO_3^- concentrations for our model domain are obtained from the annual objectively analyzed mean concentrations of the WOA 2009 $1^\circ \times 1^\circ$ data (Garcia et al., 2010a,b), and interpolated over the region of our model domain to match the resolutions of the other datasets.

RRPOC is estimated from primary production following Bohlen et al. (2012). According to the carbon-based approach of Behrenfeld et al. (2005), average annual primary production is derived from photosynthetically available radiation (PAR), the diffuse attenuation coefficient at 490 nm (K490), chlorophyll *a* (Chl *a*) and mixed layer depth (MLD). PAR, K490 and Chl *a* are from the Aqua-Modis satellite data (2005–2010) (<http://oceancolor.gsfc.nasa.gov/>), and MLD is from the Hybrid Coordinate Ocean Model (HYCOM, <http://orca.science.oregonstate.edu/1080.2160.monthly.hdf.mld.hycom.php>). Export production is estimated from primary production and sea-surface-temperature (SST) (Dunne et al., 2005), where SST is from the WOA 2009 annual average $1^\circ \times 1^\circ$ temperature data (Locarnini et al., 2010). The rate of particle transport

at each grid cell to the seafloor is calculated using the Martin curve (Eq. 2) (Martin et al., 1987). To obtain more accurate estimates for RRPOC of our regional box model, all data processed in this experiment are interpolated on a grid of $2' \times 2'$ in the UM box and $20' \times 20'$ in the D box, and the ETOPO2 data ($2' \times 2'$) are averaged within each $20' \times 20'$ grid cell in the D box.

5 The Aqua-Modis data ($5' \times 5'$) and NO_3^- and O_2 concentrations from WOA 2009 dataset are interpolated or averaged horizontally to match these resolutions. The vertical resolution of the NO_3^- and O_2 concentrations are interpolated to resolve the bathymetry of the ETOPO2 data, and the NO_3^- and O_2 concentrations closest to the sediment are applied in Eq. (1) for the bottom water NO_3^- and O_2 concentrations.

10 Finally, the L_{DIN} derived from observational datasets are averaged over the regions represented by UM and D boxes to produce an annual NO_3^- loss term.

2.5 Phosphate—Phosphorus regeneration

15 **Phosphate—Phosphorus** regeneration is estimated according to Wallmann (2010) and Flögel et al. (2011) with both model- and data-based estimates for the rain rate. We estimate benthic PO_4^{3-} regeneration (resupply of benthic PO_4^{3-} into the water column, Ben_{DP}) from the RRPOC degradation ratio (r_{REG}) and the POC burial rate in the sediments (BURPOC) according to:

$$\text{Ben}_{\text{DPUM}} = \frac{\text{RRPOC}_{\text{UM}} - \text{BURPOC}_{\text{UM}}}{r_{\text{REG}}} \quad (5)$$

$$20 \quad \text{Ben}_{\text{DPD}} = \min \left(\frac{\text{RRPOC}_{\text{D}} - \text{BURPOC}_{\text{D}}}{r_{\text{REG}}}, \frac{\text{RRPOC}}{106} \right) \quad (6)$$

where RRPOC is estimated with the methods described in [SeetsSections](#). 2.4.1 and 2.4.2. A minimum condition is introduced in the D-box to prevent Ben_{DP} exceeding the rain rate of particulate organic phosphate ($\text{RRPOP} = \text{RRPOC} / 106$) to the deep ocean, but not for the UM box, because there are possible extra sources of RRPOP, such as inputs via weathering or eolian deposition, onto the continental shelf, which is contained in the UM box in our model.

BURPOC is estimated from Eq. (7) for the continental shelf (UM box) and Eq. (8) for the deep-sea sediment (D box), and r_{REG} is the C:P regeneration ratio estimated via Eq. (9) following the empirical relations of Wallmann (2010).

$$\text{BURPOC}_{\text{UM}} = 0.14 \cdot \text{RRPOC}_{\text{UM}}^{1.11} \quad (7)$$

$$5 \quad \text{BURPOC}_{\text{D}} = 0.014 \cdot \text{RRPOC}_{\text{D}}^{1.05} \quad (8)$$

$$r_{\text{REG}} = 123 + (-112) \cdot \exp\left(-\frac{\text{O}_2}{32}\right) \quad (9)$$

where O_2 is the oxygen concentration in the ambient bottom water (in $\mu\text{mol kg}^{-1}$). r_{REG} in Eq. (9) is higher than the Redfield ratio in oxic water, resulting in preferential P burial under 10 oxic conditions; it is much smaller than the Redfield ratio when $\text{O}_2 < 20 \mu\text{mol kg}^{-1}$, indicating excess phosphate release from the sediment under suboxic conditions.

2.6 Synthesis configurations

15 Nitrogen deposition, benthic denitrification and phosphate regeneration are integrated into the synthesis model configurations to explore the model sensitivity to each process and their mutual interactions in the ETSP. The configurations (Syn1 to Syn4) with different benthic processes and atmospheric inputs are summarized in Table 1.

2.6 Model sensitivity experiments

20 Since the atmospheric nitrogen deposition data from Lamarque et al. (2011) only include results of a single chemistry-climate model, a multi-model perspective could offer additional insights into the influence of uncertainties in nitrogen deposition on our model results. Three recent inter-model comparisons (Dentener et al., 2006; Lamarque et al., 2013; Vet et al., 2014) show very similar performance over our model domain, therefore we choose the results from Dentener et al. (2006), which is also applied in a number of benchmark papers such as Duce et al. (2008). The influence of DON from the atmospheric nitrogen deposition on

the nitrogen budget is investigated by applying that DON accounts for 30% of the total dissolved nitrogen deposition suggested by Cornell et al. (2003); Kanakidou et al. (2012). The bioavailability of the deposited DON is also considered by assuming that 30% of it is available to primary producers according to Paerl and Whitall (1999). 10% and 50% bioavailability is also applied in our model to account for the underlying uncertainties. Considering the rapid rise of nitrogen deposition (Duce et al., 2008), we also apply the RCP 8.5 scenario for the year 2100 predicted by Lamarque et al. (2011) in our model domain.

Bonnet et al. (2013) reported a large quantity of aphotic nitrogen fixation in the ETSP, which can account for as much as 90% of the total fixed-N input via nitrogen fixation there. To test the effect of aphotic nitrogen fixation on the nitrogen budget of the ETSP, we include the aphotic nitrogen fixation rate measured by Bonnet et al. (2013) as additional NO_3^- input in two sensitivity experiments AphoticNfix1 and AphoticNfix2. Due to the very low sampling density of their data, we extrapolate their data to our model domain and assume that the nitrogen fixation rate in the open ocean is the same as that measured at the coast. Aphotic nitrogen fixation is responsible for 0.0711, 0.0528 and 0.0528 $\mu\text{mol N kg}^{-1} \text{yr}^{-1}$ (0.44, 6.5 and 25.6 Tg N yr^{-1}) fixed-N input into the UM, I and D boxes, respectively, with estimates from the 2010 cruise (AphoticNfix1). It contributes 0.0109, 0.0057 and 0.0059 $\mu\text{mol N kg}^{-1} \text{yr}^{-1}$ (0.067, 0.70 and 2.9 Tg N yr^{-1}) when applying estimates for the 2011 cruise (AphoticNfix2).

Since our model domain only includes the top 2000 m of the water column, the sediments only account for a small portion of the whole sediment of the ETSP (Table. 2). A sensitivity experiment high-BD/PR is performed with the assumption that all of the bottom of the D box is in contact with the sediment below 500 m (high benthic denitrification (high-BD), or high phosphate-phosphorus regeneration (high-PR)) including all NO_3^- losses by benthic denitrification and phosphate release by phosphate regeneration of phosphorus regeneration at the sediment.

The original work of Martin et al. (1987) and Van Mooy et al. (2002) indicate a lower value for the exponent b of Eq. (2) in suboxic water. We perform an additional sensitivity experiment with $b = 0.4$ according to the suggestion by Van Mooy et al. (2002), to explore the influence of benthic denitrification and phosphate-phosphorus regeneration under conditions of slower

POC remineralization. We perform another sensitivity experiment to explore the influence of organic matter remineralization on the benthic denitrification and phosphorus regeneration with more recent findings (Guidi et al., 2015), where we apply the variable Martin curve exponent b values in our model domain. In the UM-box, $b = 0.83$, which corresponds to the Peru-Chile upwelling region, is applied, whereas $b = 0.85$ is applied in the D-box, which is the average of the b values for the regions named Chile-Peru Current Coastal (CHIL), Pacific Equatorial Divergence (PEQD), South Pacific Subtropical Gyre (SPSG) and Western Pacific Warm Pool (WARM) in Guidi et al. (2015).

3 Results

10 3.1 Nitrogen deposition

Due to the low NO_3^- concentrations in the surface U and S boxes, the annual nitrogen input by atmospheric nitrogen deposition accounts for 63 % and 10 %, respectively, of nitrogen inventories of the U and S boxes. Figure Fig. 2 indicates that the extra fixed-N bioavailable nitrogen input by nitrogen deposition reduces the growth of nitrogen fixers in the surface ocean by about 0.72–0.73 mainly in the U box, even though more nitrogen is deposited into the S box. Nitrogen fixation is reduced by about 0.7 and 0.1 Tg N yr^{-1} (about 12 % respectively in the U and S boxes (about 18 % of total nitrogen fixation), which and 5 % of the total). The reduction in nitrogen fixation accounts for about 48 % of the total fixed-N bioavailable nitrogen inputs into surface waters from atmospheric deposition (1.5 Tg N yr^{-1}). Water-column denitrification stays almost unchanged because the increase in export production (EP) of Phy by Phy (ordinary phytoplankton) is almost exactly compensated by the decrease in EP of NF nitrogen fixers (NF), resulting in essentially unchanged total EP. As a result of the $\sim 50 \approx 50$ % of the nitrogen deposition not immediately compensated by a decline in compensated by lower nitrogen fixation, the model domain becomes a larger fixed-N source (Fig. 2). The N-loss fixed-N loss through the lateral boundary increases from $0.93 \text{ Tg N yr}^{-1}$ in the control configuration to 1.7 Tg N yr^{-1} in the configurations including nitrogen deposition, leading to about $0.78 \text{ Tg N yr}^{-1}$ extra fixed-N

loss from the model domain, i.e. about 50 % of the total ~~fixed-N input by nitrogen~~ ~~bioavailable nitrogen input from atmospheric~~ deposition. Thus, almost all the extra nitrogen input into the model domain via nitrogen deposition is offset by ~~lower reduced~~ nitrogen fixation and ~~nitrogen loss by enhanced~~ lateral transport out of the model domain.

There is ~~Nitrogen deposition has~~ no significant influence ~~of nitrogen deposition on the~~ ~~biogeochemical tracer concentrations of the model at steady state: ordinary phytoplankton (Phy)~~ ~~)concentrations increase~~ ~~Phy concentration increases~~ by 3 % in the U box and even smaller changes occur in the S box, which can be attributed to the stronger nitrogen deficit in the region above the OMZ (U box) than in the open ocean (S box) (~~not shown~~). ~~Largest changes are found for Fig. S2~~. The largest effect is a ~~decrease by about 9 % of~~ the concentration of nitrogen fixers (NF) ~~that decrease by about 9 %~~ in the U box, ~~partly~~ counteracting the nitrogen input via nitrogen deposition (Fig. 3). ~~Again in the S box~~, NF concentration stays almost unaltered ~~in the S box~~ (Fig. 3). ~~There are also slight~~ ~~Slight~~ variations of the NO_3^- concentration ~~occur~~ in the UM box and of ~~the~~ O_2 concentrations in the I and D boxes (~~not shown~~ Fig. S2).

3.2 Benthic denitrification

The data-derived benthic denitrification and ~~phosphate phosphorus~~ regeneration in the UM and D boxes are shown in Table 3. ~~The modeled~~ ~~2. Modeled~~ net primary production (NPP) in the surface ocean above the UM and D boxes is, respectively, 1.4 and $0.87 \text{ g C m}^{-2} \text{ day}^{-1}$, indicating higher NPP in the coastal upwelling region and lower NPP in the open ocean adjacent to the upwelling region, which is consistent with the estimate by Behrenfeld et al. (2005). Due to the small ~~sediment-area~~ ~~sediment-area~~ percentages, the annual nitrogen loss by benthic denitrification is 0.17 and $0.82 \text{ Tg N yr}^{-1}$ in the UM and D boxes, accounting for only about 0.14 % and $5.1 \times 10^{-3} \text{ % year}^{-1}$, respectively, of the NO_3^- inventories ~~in these boxes~~ (Table 32). The higher sedimentary NO_3^- sink in the UM box can be attributed to the anoxic conditions and larger RRPOC ~~there~~ ~~(rain rate)~~.

Our simulated biogeochemical tracer concentrations at steady state are quite robust with respect to ~~including~~ benthic denitrification (Fig. 3). ~~There are~~ ~~Including~~ benthic denitrification ~~causes~~ only minor deviations ~~of~~ ~~in~~ the MBD and DBD configurations ~~from~~ ~~compared to~~ the

control run. Nitrogen fixation rates ~~increases-increase~~ by about 2.9 % and 5.8 % respectively, in the MBD and DBD configurations (A-bars in panels MBD and DBD of Fig. 4). ~~Most of this increase occurs in the U box, which receives water with a strong N-deficit via upwelling.~~ Obviously, the response is stronger in the DBD configuration than in the MBD configuration, because ~~there is an approximately 5-times larger~~ fixed-N loss via benthic denitrification in the DBD configuration ~~is approximately 5-times larger~~ (A-bars in Fig. 4). The DBD configuration results in stronger responses of nitrogen fixation and lateral fluxes to benthic denitrification: the increase in nitrogen fixation ~~can not~~ ~~cannot~~ fully compensate the nitrogen loss by benthic denitrification, ~~thus,~~ ~~Thus,~~ the model domain becomes a ~~-~~smaller fixed-N source, about 25 % of that in the control configuration. ~~Otherwise~~ ~~In other respects,~~ the steady-state solutions of the MBD and DBD configurations are almost identical to ~~that~~ ~~those~~ of the control configuration after including benthic denitrification ~~-(Fig. S3)~~. The temporal development of biogeochemical tracer concentrations is also insensitive to ~~including the presence or absence of~~ benthic denitrification (~~not shown~~ Fig. S3).

15 3.3 Phosphate Phosphorus regeneration

Phosphate release by ~~phosphate-phosphorus~~ regeneration accounts for about 0.23 % and $2.2 \times 10^{-3} \text{ % year}^{-1}$, respectively, of the total phosphate inventories in the UM and D boxes (Table 2). The higher sedimentary PO_4^{3-} source in the UM box can be attributed to the anoxic conditions and larger RRPOC ~~there~~^{rain rate}. The phosphate release associated with benthic ~~phosphate-phosphorus~~ regeneration can stimulate nitrogen fixation and EP from the surface ocean, followed by higher water-column denitrification, owing to enhanced decomposition of exported organic matter (A-bars in MPR and DPR panels ~~ef in~~ Fig. 4). In the MPR configuration, nitrogen fixation ~~and water-column denitrification increase by 12 and 11 %, respectively, while in the~~ ~~increases by about 18 % in the U box, and stays almost unchanged in the S box. In the~~ DPR configuration, nitrogen fixation ~~and water-column denitrification increase by about 17 and 15 % (A-bars in~~ ~~also increases by about 23 % in the U box when benthic phosphate release is included (Fig. 4)~~. Water-column denitrification increases by 10 % and 14 %, respectively, in the MPR and DPR panels ~~of~~ configurations (Fig. 4). ~~Compared with~~ ~~Compared to~~ the MBD and

DBD configurations, phosphate regeneration cannot benthic phosphorus regeneration does not turn our model domain into a -smaller fixed-N source even though there is-, in spite of higher water-column denitrification, because nitrogen fixation can compensate for the nitrogen loss by water-column denitrification enhanced nitrogen fixation compensates for the extra nitrogen loss (A-bars in Fig. 4).

While changes in nitrogen deposition and benthic denitrification are to a -large extent compensated by adjustments via simulated in nitrogen fixation, phosphate is the ultimate limiting nutrient in our model domain (Su et al., 2015). Hence, the extra phosphate input into the model domain by phosphate-benthic phosphorus regeneration has a -more significant influence on the steady-state model results than the perturbations of the nitrogen input or loss applied in the model inputs or losses (Fig. 3). Phy concentration in the DPR configuration decreases in the U box but remains unchanged in the S box (not shown Fig. S2). Phy concentrations in the U and S boxes remain almost unaltered in the MPR configuration. Compared with the control configuration, NF concentrations in the U and S boxes increase by 11 % and 1.6 % respectively, respectively, in the MPR configuration, and 14 % and 1.6 % respectively, respectively, in the DPR configuration (Fig. 3). The nitrate concentration in the UM box decreases by about 3.84.2 % in the MPR configuration and 5.2 % in the DPR configuration (not shown Fig. S2). The temporal development of biogeochemical tracer concentrations is also appears robust to benthic phosphate regeneration (not shown phosphorus regeneration (Fig. S4).

20 3.4 Synthesis configurations

In the synthesis configurations (Table 1), phytoplankton, nutrient and oxygen concentrations are quite robust with respect to the various fluxes associated with nitrogen input or removal and phosphate release from the sediment into the water column (Fig. 3S2). However, the interactions among nitrogen fixation, water-column denitrification, benthic denitrification, and phosphate and benthic denitrification and phosphorus regeneration result in different sensitivities of nitrogen fixation and the lateral flux-fluxes to atmospheric N deposition in the presence of benthic denitrification and phosphate phosphorus regeneration (Fig. 3). In contrast to the N-DEP to the NDEP configuration, nitrogen fixation rates in the Syn3 and Syn4 configura-

tions increase by about 1.7 % and 8.5 %, in spite of the additional nitrogen input into model domain by atmospheric nitrogen deposition. The lateral Most of this increase occurs in the U box, whereas almost no change happens in the S box. The lateral fixed-N flux out of the model domain (NO_3^- source) increases by about $0.97 \text{ Tg N yr}^{-1}$ in the Syn3 configuration, which accounts for about 65 % of the total atmospheric nitrogen deposition, so that i.e., more than half of the extra nitrogen supplied by nitrogen deposition is not utilised locally. However, in the Syn4 configuration, the increase in lateral NO_3^- efflux increase only accounts for about 25 % of the total nitrogen deposition, with 75 % of the deposited nitrogen utilised in-within the model domain. Less fixed-N is lost laterally from the model domain in the configurations including data-based estimates than in those including model-based estimates, due to more NO_3^- loss within the model domain (Fig. 4). Thus, the sensitivity of lateral fluxes and the sensitivity fixed-N budget to nitrogen deposition are also is strongly controlled by benthic denitrification and phosphate regeneration rather than nitrogen deposition only phosphorus regeneration.

3.5 Model sensitivity

The nitrogen deposition rate estimated by Dentener et al. (2006) is about 48 % and 14 % higher, respectively, in the U and S boxes than the estimate of Lamarque et al. (2011). However, this increase induces only a 3.1 % decrease of nitrogen fixation in the U box and a 5.9 % increase in lateral nitrogen flux, while water-column denitrification and nitrogen fixation in the S-box remain unchanged (A and B bars in Panel NDEP of Fig. 5). Whereas the uncertainty associated with the nitrogen-deposition estimate of Dentener et al. (2006) amounts to about $\pm 40\%$, nitrogen fixation rate in the U box and lateral flux only vary by about $\pm 9.7\%$ and $\pm 20\%$, respectively (panels NDEP-low and NDEP-up in Fig. 5). Effects of accounting for atmospheric deposition of bioavailable DON are investigated in three sensitivity experiments with different scenarios for DON bioavailability (panels NDEP-DON(10%), NDEP-DON(30%) and NDEP-DON(50%) in Fig. 5). Including bioavailable atmospheric DON in addition to DIN deposition causes only minor changes, i.e., slightly lower nitrogen fixation and slightly higher lateral nitrogen efflux. The RCP 8.5 scenario projects an about 7.2 % increase in nitrogen deposition for the year 2100 compared to our main experiment (2000–2009 average according

to the RCP 4.5 scenario), causing only negligible changes to the nitrogen budget in our model domain (A bars in panels NDEP and NDEP-2100 of Fig. 5). These sensitivity experiments show that variations in nitrogen deposition are largely offset by changes in nitrogen fixation and lateral nitrogen flux out of the model domain, tending to keep a balanced nitrogen inventory.

5 The effect of aphotic nitrogen fixation is investigated in the AphoticNfix1 and AphoticNfix2 configurations, where photic nitrogen fixation decreases by 39 % and 15 %, respectively (Fig. S5). Water-column denitrification remains unchanged because more nitrogen input by aphotic nitrogen fixation does not increase export production into the OMZ. The lateral fixed-N effluxes in the AphoticNfix1 and AphoticNfix2 configurations are about 33 and 4 times of those in the control configuration, accounting for about 91 % and 78 %, respectively, of extra nitrogen input by aphotic nitrogen fixation (Fig. S5). Aphotic N₂ fixation has little effect on most tracers except NO₃⁻, which increases by 110 % and 87 % respectively in the UM box and the whole model domain for AphoticNfix1, which is a strong overestimate compared to WOA2009 data (Fig. S6). The lower estimate of aphotic N₂ fixation (AphoticNfix2) brings the NO₃⁻ concentrations closer to the WOA2009 data (Fig. S6), and the associated changes in nitrogen fluxes are similar to our other sensitivity configurations (Figs. 5 and S5). As for the sensitivity with respect to atmospheric nitrogen deposition, these changes are largely compensatory, leading to only small changes in the nitrogen budget of our model domain.

Fig. 6 shows the results of the sensitivity experiments with high-BD and high-PR. Compared with Fig. 3, the influence on the biogeochemical tracer concentrations at steady state is stronger, due to the larger NO_3^- loss via benthic denitrification and PO_4^{3-} release via phosphate phosphorus regeneration (Table 3). High-BD or high-BD together with high-PR can even turn our model domain into a NO_3^- sink (B-bars in panels DBD and DBD+DPR of Fig. 4).

25 Applying the Martin Curve exponent $b = 0.4$ also amplifies the influence of benthic denitrification and **phosphate**-**phosphorus** regeneration on phytoplankton and biogeochemical tracers, although the effect is weaker than in the high-BD and high-PR configurations. For example, NF_U increases by as much as 33 % in the DBD+DPR configuration, and NF_S increase about 15 % (Fig. 7). Compared with A-bars in Fig. 4, this enhanced influence results from the higher NO_3^- loss through benthic denitrification and phosphate input via **phosphate**-**phosphorus** re-

generation (C-bars in Fig. 4). Spatial variations in the Martin Curve exponent b as suggested by Guidi et al. (2015) result in nitrogen fluxes and concentrations at steady-state which are in good agreement with those in our main configurations (A and C bars in Fig. S7 and Fig. S8), because the b values from Guidi et al. (2015) are all very close to $b = 0.82$, as used in our main configurations.

Due to the higher RRPOC reaching the sea floor ~~below the water column with under~~ suboxic conditions, benthic denitrification increases by about 42 % and 198 % (A- and C-bars of panels MBD and DBD in Fig. 4) and phosphate-phosphorus regeneration increases about 36 % and 200 % ~~respectively, respectively~~, in model- and data-based estimations in the sensitivity experiments with Martin Curve value $b = 0.4$. Our model domain switches to a NO_3^- sink in the DBD and DBD+DPR configurations with $b = 0.4$ (C-bars in Fig. 4). Comparing A- and C-bars of panel DBD in Fig. 4, we find that higher benthic denitrification can stimulate nitrogen fixation, but ~~the water-column~~ denitrification remains constant. However, comparing A- and C-bars of panel DBD+DPR in Fig. 4, we find that higher benthic denitrification can increase nitrogen fixation and water-column denitrification, indicating an important role of PO_4^{3-} in the balanced balancing nitrogen inventory. This shows a positive feedback between water-column denitrification in the OMZ and benthic denitrification below, caused by slower remineralisation under ~~2~~ anoxic conditions, which results in more RRPOC reaching the sea floor. All above comparisons indicate that phosphate limitation could be responsible for breaking this positive feedback under the assumption of our model that PO_4^{3-} is the only limiting factor for the growth of nitrogen fixers.

4 Discussion and conclusions

The impact of nitrogen deposition on the ETSP has rarely been investigated so far, since this region is believed to receive less ~~atmospheric fixed-N deposition compared with bioavailable nitrogen from atmospheric deposition than~~ the coasts of western Europe, South and East Asia (Dentener et al., 2006; Duce et al., 2008). The influence of anthropogenic nitrogen deposition on the biogeochemical cycles of the open ocean is increasing and the increase in atmospheric nitro-

gen deposition will probably induce an approximately 10 % rise in carbon sequestration on land and in the ocean by 2030 (Duce et al., 2008; Reay et al., 2008). The ETSP, a -typical N-deficit region due to denitrification in the OMZ, is likely to be sensitive to anthropogenic nitrogen deposition. We find that, in our model, nitrogen deposition can inhibit N₂ fixation by relieving nitrogen limitation for Phy: ~~this~~ which counteracts the effect of atmospheric nitrogen input. This is in line with the finding that N₂ fixation decreases with increasing nitrogen deposition in global-scale models that use essentially the same assumptions about the environmental controls on marine nitrogen fixation ([Krishnamurthy et al., 2007, 2009, 2010; Zamora et al., 2010](#)) ([Krishnamurthy et al., 2007, 2009, 2010; Zamora et al., 2010](#)). Another portion of the deposited nitrogen is exported out of the model domain since not all the deposited nitrogen can be taken up by Phy locally ~~because of~~, owing to phosphate limitation (Fig. 2). [Appendix A shows the model results when there is](#) The coastal upwelling region (the U box) in our model is more sensitive to nitrogen deposition due to the N-deficit water supplied by upwelling (Fig. 5). In spite of the uncertainties in the magnitude of atmospheric bioavailable nitrogen deposition and the bioavailability of deposited DON, atmospheric deposition appears unable to exert a strong influence of the fixed-N budget of our model domain, as nitrogen deposition is mostly counteracted by decreased nitrogen fixation and enhanced nitrogen export out of the model domain. Replacing obligate N₂ fixation in our model by facultative N₂ fixation, ~~which~~ slightly enhances the strength of the negative feedback between nitrogen fixation and nitrogen deposition (see Appendix A in the supplementary material for details).

In a 3-D biogeochemical model study of the nitrogen fixation response to benthic denitrification, Somes et al. (2013) found that an increase of benthic denitrification can stimulate The net primary production (NPP) estimated in our study is on average $1.4 \text{ gC m}^{-2} \text{ day}^{-1}$ and $0.87 \text{ gC m}^{-2} \text{ day}^{-1}$, respectively, in the surface ocean above the UM and D boxes, according to the carbon-based approach of Behrenfeld et al. (2005). Pennington et al. (2006) estimated NPP from ship-collected data as, respectively, $1.2 \text{ gC m}^{-2} \text{ day}^{-1}$ and $0.67 \text{ gC m}^{-2} \text{ day}^{-1}$ for the surface ocean above the UM and D boxes, whereby the NPP for the surface ocean above the D box could be somewhat overestimated because the western boundary for their data is 140°W . Our estimates are about 17 % and

30 % higher than Pennington et al.'s for the surface ocean above the UM and D boxes, because Behrenfeld et al.'s carbon-based approach yields considerably higher values than other approaches for tropical regions. Export production is linearly related to RRPOC (Eq. 2), as is RRPOC to BD (Eq. 1). PR and RRPOC are related through a power law with exponents of 1.11 and 1.05 for the UM and D boxes, respectively (Eqs. 5–8). Fixed-N loss via BD and PO_4^{3-} release by PR estimated with ship-collected data should thus be within the range corresponding to the NPP estimates from Behrenfeld et al. (2005) and Pennington et al. (2006). However, in our model-based estimation of BD and PR, the fixed-N loss by BD and PO_4^{3-} release by PR are, respectively, 421 % and 306 % higher than our data-based estimates. Therefore, the NPP estimated from ship-collected data lead to benthic remineralization fluxes between our data-based and model-based estimations.

Aphotic nitrogen fixation, i.e., below the euphotic zone, has been considered an important contribution to the nitrogen budget of the ETSP (Bonnet et al., 2013). Our model configurations including aphotic N_2 fixation, ~~but this occurred under the condition that~~ are in line with this view, as the large amount of additional nitrogen input in the AphoticNfix1 configuration induces overestimation of NO_3^- concentrations in the model domain, whereas NO_3^- concentration is closer to the WOA2009 data in the AphoticNfix2 configuration, which has a much lower aphotic nitrogen fixation rate. Due to the very sparse data for aphotic N_2 fixation was tuned to compensate for the ~~the~~, we had to extrapolate the data for the coastal region to the vast open ocean of the ETSP, which could have led to an overestimation of aphotic nitrogen fixation. Thus, we expect that aphotic N_2 fixation is likely closer to the lower (2011) estimate of Bonnet et al. (2013), as the resulting NO_3^- concentrations are closer to the WOA2009 data.

Table 3 shows our modeled fluxes in comparison with other model-based and observational estimates. N_2 fixation in the upwelling region of our model is higher than those reported by Somes et al. (2010), DeVries et al. (2013), and Dekaezemacker et al. (2013), but within the range suggested by Loescher et al. (2014). However, N_2 fixation rate in the open ocean of our model is lower than those by Somes et al. (2010) and DeVries et al. (2013), but within the range suggested by Halm et al. (2012). Water-column denitrification is comparable with

Somes et al. (2010) and DeVries et al. (2013), but lower than that from Kalvelage et al. (2013). Our predicted fixed-N loss by reducing mortality and predation rates. Nitrogen fixation can also be stimulated by a N-deficit (Deutsch et al., 2007), which can result from benthic denitrification. In our 2-D model, we also find that loss by benthic denitrification is comparable to other estimates. Phosphorus regeneration in our analysis spans a wide range, but is comparable with the evaluation of Bohlen et al. (2012) for the full depth of the model domain. Major nitrogen and phosphorus fluxes in our study also span wide ranges because fluxes both from the upper 2000 meter and full-depth of the ocean are assessed with both data and model-based evaluations, in each case accounting for organic matter remineralization under different oxygen conditions (Table 3). Currently, both global and regional estimates of nitrogen fixation and benthic remineralization rates are rather uncertain, owing to temporal and spatial variations and problems associated with measuring methods (e.g. Mohr et al., 2010). Thus, we had to apply rather wide ranges in order to investigate the potential influence of these processes on the nitrogen budget of our model domain.

Under the assumption that N_2 fixation compensates for any fixed-N deficit (Deutsch et al., 2007), nitrogen fixation can be enhanced by the N-deficit caused by benthic denitrification, so that nitrogen fixation can somehow compensate for the nitrogen stimulated by benthic denitrification. Somes et al. (2013) found that benthic denitrification stimulates N_2 fixation in their 3-D biogeochemical model, which was tuned under the condition that the global fixed-N budget was balanced. Even though we make no a priori assumption about the association between N_2 fixation and fixed-N loss processes, we also find that a fixed-N deficit can stimulate N_2 fixation, thus compensating for the fixed-N loss.

We find a strong increase in primary production after incorporating benthic phosphate regeneration that incorporating benthic phosphorus regeneration strongly increases primary production, which is mainly attributed to nitrogen fixation (Panels MPR and DPR in Fig. 4). Phosphate regeneration will be Phosphorus regeneration is enhanced under O_2 deficit conditions, and the enhanced phosphate release will stimulate stimulates primary production, finally resulting in the expansion of OMZs and a possible possibly causing a positive feedback loop leading to more benthic phosphate phosphorus regeneration (Van Cappellen and Ingall, 1994;

Wallmann, 2010). However, our model domain only represents the upper 2000 m of the ocean and its sediments only account for a small fraction of ~~total sediment~~ the total sediment area in the ETSP. The model results incorporating benthic denitrification and ~~phosphate regeneration for a ease where phosphorus regeneration, and assuming that~~ all of the ~~bottom of the~~ D box is ~~assumed~~ in contact with the sediment, are shown in Figs. 4 and 6. ~~For our parameterization of nitrogen fixation being favoured~~ 6. Our parameterization allows nitrogen fixation to be favored in N-deficit waters, ~~since~~ the increase of water-column denitrification can ~~always~~ be compensated by increased nitrogen fixation when ~~there is only phosphate regeneration~~ ~~phosphorus regeneration is sufficient~~ (panels MPR and DPR in Fig. 4). Our model domain can only turn 10 into a fixed-N sink after including high-BD of the full sediment.

Due to the ~~The~~ simplicity and computational efficiency of our box model, ~~it is relatively easy to explore facilitates exploring~~ model sensitivity to ~~various~~ processes related to the nitrogen budget of the ETSP. Even though ~~some details of~~ spatial and temporal variations are missing compared with results from ~~the~~ 3D global circulation models (Krishnamurthy et al., 2007, 2010; Zamora et al., 2010), we can efficiently diagnose the regional impacts at steady state. ~~Stimulatory effect can be identified~~ We identify stimulatory effects, respectively, between nitrogen fixation and water-column denitrification, ~~phosphate phosphorus~~ regeneration and nitrogen fixation, ~~phosphate phosphorus~~ regeneration and water-column denitrification, and atmospheric deposition and lateral NO_3^- transport (Fig. 4). Depressive effects occur between atmospheric deposition and nitrogen fixation, and between benthic denitrification and ~~the~~ lateral NO_3^- transport (Fig. 4). The model sensitivity to processes related to the nitrogen budget of the OMZ in the ETSP is illustrated in Fig. 8. ~~The Nitrogen fixation rate~~ Nitrogen fixation can be enhanced by benthic denitrification, compensating for part of the NO_3^- loss. The stimulatory effect between nitrogen fixation and water-column denitrification ~~can help to keep a help~~ balancing the fixed-N balance in the ocean budget. The extra fixed-N input by nitrogen deposition ~~can be is partly~~ counteracted by decreased nitrogen fixation and ~~partially partly~~ removed by lateral flux. All of these local responses combined constitute a nitrogen-balancing mechanism in the ETSP. ~~Even though anammox has been suggested as a possible~~ Even though water-column denitrification has been considered the major fixed-N loss in the water column 25

of the ETSP (Lam et al., 2009; Kalvelage et al., 2013), both denitrification and anammox are ultimately driven by the flux of organic matter (Koeve and Kähler, 2010) and create a fixed-N deficit. Hence, the processes for simplicity, the stimulatory effects between nitrogen fixation and fixed-N loss, and phosphate-phosphorus regeneration and fixed-N loss still apply even if anammox replaced water-column denitrification as the fixed-N loss pathway. Thus, the nitrogen-balancing mechanism in the ETSP should not depend on whether the fixed-N is lost through denitrification or anammox.

In the high-BD sensitivity experiment, our model domain turns into a NO_3^- sink (Fig. 4). The NO_3^- inventory in the ETSP is determined by nitrogen fixation, water-column denitrification, benthic denitrification, and lateral NO_3^- flux. Since ~~nitrogen fixation and water-column denitrification occur only in the surface ocean and the OMZ, which are included in our model domain, our model domain (except in the high-BD sensitivity configuration) encompasses only the water column and a small fraction of the corresponding sediment area,~~ we cannot rule out that the ETSP including sedimentary denitrification is a NO_3^- sink, which is consistent with many ~~model-derived or observational~~ model- or data-derived results (Ganachaud and Wunsch, 2002; Kalvelage et al., 2013). Extra phosphate input into the model domain via ~~phosphate phosphorus~~ regeneration can increase water-column denitrification significantly due to the increase ~~of~~ in EP from the surface ocean. However, ~~phosphate phosphorus~~ regeneration alone can not turn our model domain into a NO_3^- sink, ~~which corroborates that the water column of ETSP is net source of~~.

The remineralization rate of organic matter is thought to be reduced under anoxic conditions (Martin et al., 1987; Van Mooy et al., 2002), ~~which results in more RRPOC resulting in a higher rain rate (RRPOC)~~ reaching the sediments. According to the analysis of Bohlen et al. (2012), benthic denitrification is very sensitive to RRPOC, i.e., higher RRPOC results in higher benthic denitrification. Based on ~~the our~~ findings that higher benthic denitrification can increase nitrogen fixation, higher nitrogen fixation could result in higher water-column denitrification and the expansion of ~~OMZs and finally a~~ ~~the~~ OMZ, and hence a positive feedback between water-column and benthic denitrification. But this positive feedback is only observed in ~~the~~ configurations with phosphate input via ~~phosphate phosphorus~~ regeneration, which indicates

that PO_4^{3-} limitation could prevent this positive nitrogen play an important role in preventing this positive fixed-N loss feedback.

It is known from laboratory experiments that diazotrophic phytoplankton can also utilize DIN for growth (e.g., Holl and Montoya, 2005). In contrast to our NF model where NF can only fix from the atmosphere, Schmittner et al. (2008) introduced a formulation to allow also NO_3^- uptake by diazotrophs. In Schmittner et al. (2008)'s model, nitrogen fixers preferentially use nitrate when available and cover only the residual N demand by N_2 fixation, denoted as facultative N_2 -fixation. Thus, we explore the effect of facultative N_2 -fixation on our model results with extra fixed-N input by nitrogen deposition.

Compared to results from the configurations in which NF can only fix N_2 (Fig. 3), both Phy and NF in the U-box are more robust to the extra nitrogen input via nitrogen deposition, for instance, Phy increases by 1.5 % (facultative N_2 -fixation) vs. 2.9 % (obligate N_2 -fixation), and NF decreases by 3.9 % (facultative N_2 -fixation) vs. 10 % (obligate N_2 fixation). Again, the biogeochemical concentrations at steady state are relatively insensitive to nitrogen deposition (not shown).

There is a stronger negative feedback between nitrogen deposition and facultative N_2 -fixation, since nitrogen fixation is reduced by about 21 % (facultative N_2 -fixation) compared to 12 % (obligate N_2 fixation) (Fig. 8). The increased lateral fluxes of NO_3^- only account for about 21 % of the extra nitrogen input by nitrogen deposition (facultative N_2 -fixation) compared to 50 % (obligate N_2 fixation) (Fig. 8). Thus, facultative N_2 -fixation can adjust nitrogen fixation in response to nutrient concentrations in the surface boxes and control the magnitude of our model domain being a NO_3^- source.

5 Summary

The influence of atmospheric nitrogen deposition and benthic remineralization on the nitrogen budget of the ETSP is investigated with a conceptually simple and computationally efficient box model. Additional nitrogen input by atmospheric nitrogen deposition is offset by about 48 % by reduced nitrogen fixation with the remainder being transported horizontally out of the

model domain, irrespective of uncertainties underlying the actual rate of nitrogen deposition and bioavailability of deposited DON. All our data- and model-based estimations for benthic remineralization are comparable with those of previous studies based on both models and observations. Modeled responses to these fluxes indicate stabilizing feedbacks, which tend to balance the nitrogen inventory. Variations in these fluxes due to variations in NPP, aphotic nitrogen fixation and organic matter remineralization under different oxygen conditions do not change these feedbacks. In the high-BD configuration, our model domain turns into a NO_3^- sink, which indicates that the ETSP, including the upwelling region, the open ocean, and the sediments, might be a NO_3^- sink. More research is needed to better constrain the fluxes of nitrogen and phosphorus, including the amount of bioavailable nitrogen deposition, nitrogen fixation and benthic denitrification, to allow a more reliable estimation of the nitrogen budget of the ETSP.

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25 References

Augu  res, A.-S. and Loreau, M.: Regulation of Redfield ratios in the deep ocean, *Global Biogeochem. Cy.*, 29, 254–266, doi:10.1002/2014GB005066, 2015.

Bakun, A. and Weeks, S. J.: The marine ecosystem off Peru: what are the secrets of its fishery productivity and what might its future hold?, *Prog. Oceanogr.*, 79, 290–299, 2008.

Behrenfeld, M. J., Boss, E., Siegel, D. A., and Shea, D. M.: Carbon-based ocean productivity and phytoplankton physiology from space, *Global Biogeochem. Cy.*, 19, 1–14, GB1006, doi:10.1029/2004GB002299, 2005.

Berelson, W. M.: The flux of particulate organic carbon into the ocean interior: a comparison of four U.S. JGOFS regional studies, *Oceanography*, 14, 59–67, 2001.

Bohlen, L., Dale, A. W., Sommer, S., Mosch, T., Hensen, C., Noffke, A., Scholz, F., and K. Wallmann: Benthic nitrogen cycling traversing the Peruvian oxygen minimum zone, *Geochim. Cosmochim. Ac.*, 75, 6094–6111, 2011.

Bohlen, L., Dale, A. W., and K. Wallmann: Simple transfer functions for calculating benthic fixed nitrogen losses and C:N:P regeneration ratios in global biogeochemical models, *Global Biogeochem. Cy.*, 26, 1–16, GB3029, doi:10.1029/2011GB004198, 2012.

[Bonnet, S., Dekaezemacker, J., Turk-Kubo, K. A., Moutin, T., Hamersley, R. M., Grosso, O., Zehr, J. P., and Capone, D. G.: Aphotic N₂ fixation in the eastern tropical south pacific ocean, PLoS one, 8\(e81265\), 1–14, doi:10.1371/journal.pone.0081265, 2013.](#)

Brandes, J. A. and Devol, A. H.: A global marine-fixed nitrogen isotopic budget: Implications for holocene nitrogen cycling, *Global Biogeochem. Cy.*, 16, 1–14, doi:10.1029/2001GB001856, 2002.

Canfield, D. E.: Models of oxic respiration, denitrification and sulfate reduction in zones of coastal upwelling, *Geochim. Cosmochim. Ac.*, 70, 5753–5765, doi:10.1016/j.gca.2006.07.023, 2006.

Christensen, J. P., Murray, J. W., Devol, A., and Codispoti, L. A.: Denitrification in the continental shelf sediment has major impact on the oceanic nitrogen budget, *Global Biogeochem. Cy.*, 1, 97–116, 1987.

Clarke, L., Edmonds, J., Jacoby, H., Pitcher, H., Reilly, J., and Richels: Scenarios of Greenhouse Gas Emissions and Atmospheric Concentrations: Synthesis and Assessment Product 2.1a Report by the U.S. Climate Change Science Program and the Subcommittee on Global Change Research, Technical Report, Department of Energy, Office of Biological and Environmental Research, Washington, D.C., USA, 2007.

Codispoti, L. A.: An oceanic fixed nitrogen sink exceeding 400 Tg N a⁻¹ vs the concept of homeostasis in the fixed-nitrogen inventory, *Biogeosciences*, 4, 233–253, doi:10.5194/bg-4-233-2007, 2007.

Codispoti, L. A., Brandes, J., Christensen, J., Devol, A., Naqvi, S. W., Paerl, H., and Yoshinari, T.: The oceanic fixed nitrogen and nitrous oxide budgets: moving targets as we enter the anthropocene?, *Sci. Mar.*, 65, 85–105, doi:10.3989/scimar.2001.65s285, 2001.

Cornella

Cornell, S. E., Jickells, T. D., Cape, J. N., Rowland, A. P., and Duce, R. A.: Organic nitrogen deposition on land and coastal environments: a review of methods and data, *Atmos. Environ.*, 37, 2173–2191, doi:10.1016/S1352-2310(03)00133-X, 2003.

Dentener, F., Drevet, J., Lamarque, J. F., Bey, I., Eickhout, B., Fiore, A. M., Hauglustaine, D., Horowitz, L. W., Krol, M., Kulshrestha, U. C., Lawrence, M., Galy-Lacaux, C., Rast, S., Shindell, D., Stevenson, D., Noije, T. V., Atherton, C., Bell, N., Bergman, D., Butler, T., Cofala, J., Collins, B., Doherty, R., Ellingsen, K., Galloway, J., Gauss, M., Montanaro, V., Müller, J. F., Pitari, G., Rodriguez, J., Sanderson, M., Solmon, F., Strahan, S., Schultz, M., Sudo, K., Szopa, S., and Wild, O.: Nitrogen and sulfur deposition on regional and global scales: a multimodel evaluation, *Global Biogeochem. Cy.*, 20, 1–21, GB4003, doi:10.1029/2005GB002672, 2006.

Dekaezemacker, J., Bonnet, S., Grosso, O., Moutin, T., Bressac, M. and Capone, D.: Evidence of active dinitrogen fixation in surface waters of the eastern tropical south pacific during el niño and la niña events and evaluation of its potential nutrient controls, *Global Biogeochem. Cycles*, 27, 768–779, doi:10.1002/gbc.20063, 2013.

Deutsch, C., Gruber, N., Key, R. M., Sarmiento, J. L., and Ganachaud, A.: Denitrification and N₂ fixation in the Pacific Ocean, *Global Biogeochem. Cy.*, 15, 483–506, 2001.

Deutsch, C., Sarmiento, J. L., Sigman, D. M., Gruber, N., and Dunne, J. P.: Spatial coupling of nitrogen inputs and losses in the ocean, *Nature*, 445, 163–167, 05392, doi:10.1038/nature05392, 2007.

Devol, A. H.: Direct measurement of nitrogen gas fluxes from continental shelf sediments, *Nature*, 349, 319–321, 1991.

DeVries, T., Deutsch, C., Primeau, F., Chang, B., and Devol, A.: Global rates of water-column denitrification derived from nitrogen gas measurements, *Nature*, 5, 547–550, doi:10.1038/NGEO1515, 2012.

DeVries, T., Deutsch, C., Rafter, P. A., and Primeau, F.: Marine denitrification rates determined from a global 3-d inverse model, *Biogeosciences*, 10, 2481–2496, doi:10.5194/bg-10-2481-2013, 2013.

Duarte, C. M., Dachs, J., Llabrés, M., Alonso-Laita, P., Gasol, J. M., Tovar-Sánchez, A., ~~Wihemy~~ ^{Wilhemey} ~~Sañudo-Wilhemey~~, S. S., and Agustí, S.: Aerosol inputs enhance new production in the subtropical northeast Atlantic, *J. Geophys. Res.*, 111, 1–8, G04006, 2006.

Duce, R. A.: The impact of atmospheric nitrogen, phosphorus, and iron species on marine biological productivity, in: The Role of Air–Sea Exchange in Geochemical Cycling, vol. 185 of NATO ASI Series, edited by: P. Buat-Ménard, Springer, New York, 497–529, 1986.

Duce, R. A., Liss, P. S., Merrill, J. T., Atlas, E. L., Buat-Menard, P., Hicks, B. B., Miller, J. M., Prospero, J. M., Arimoto, R., Church, T. M., Ellis, W., Galloway, J. N., Hansen, L., Jickells, T. D., Knap, A. H., Reinhardt, K. H., Schneider, B., Soudine, A., Tokos, J. J., Tsunogai, S., Wollast, R.,

and Zhou, M.: The atmospheric input of trace species to the world ocean, *Global Biogeochem. Cy.*, 5, 193–259, 1991.

Duce, R. A., LaRoche, J., Altieri, K., Arrigo, K. R., Baker, A. R., Capone, D. G., Cornell, S., Denter, F., Galloway, J., Ganeshram, R. S., Geider, R. J., Jickells, T., Kuypers, M. M., Langlois, R., Liss, P. S., Liu, S. M., Middelburg, J. J., Moore, C. M., Nickovic, S., Oschlies, A., Pedersen, T., Prospero, J., Schlitzer, R., Seitzinger, S., Sorensen, L. L., Uematsu, M., Ulloa, O., Voss, M., Ward, B., and Zamora, L.: Impacts of atmospheric anthropogenic nitrogen on the open ocean, *Science*, 320, 893–897, 5878, 2008.

Dunne, J. P., Armstrong, R. A., Gnanadesikan, A., and Sarmiento, J. L.: Empirical and mechanistic models for the particle export ratio, *Global Biogeochem. Cy.*, 19, 1–16, GB4026, doi:10.1029/2004GB002390, 2005.

Eugster, O. and Gruber, N.: A probabilistic estimate of global marine N-fixation and denitrification, *Global Biogeochem. Cy.*, 26, 1–15, GB4013, doi:10.1029/2012GB004300, 2012.

Falkowski, P. G., Barber, R. T., and Smetacek, V.: Biogeochemical Controls and Feedbacks on Ocean Primary Production, *Science*, 281, 200–206, 1998.

Flögel, S., Wallmann, K., Poulsen, C. J., Zhou, J., Oschlies, A., Voigt, S., and Kuhnt, W.: Simulating the biogeochemical effects of volcanic CO₂ degassing on the oxygen-state of the deep ocean during the Cenomanian/Turonian Anoxic Event (OAE2), *Earth Planet. Sc. Lett.*, 305, 371–384, 2011.

Ganachaud, A. and Wunsch, C.: Oceanic nutrients and oxygen transports and bounds on export production during the world ocean circulation experiment, *Global Biogeochem. Cy.*, 16, 1–14, doi:10.1029/2000GB001333, 2002.

Garcia, H. E., Locarnini, R. A., Boyer, T. P., Antonov, J. I., Baranova, O. K., Zweng, M. M., and Johnson, D. R.: Volume 3: Dissolved oxygen, apparent oxygen utilization, and oxygen saturation, in: *World Ocean Atlas 2009*, edited by: Levitus, S., NOAA Atlas NESDIS 70, U.S. Government Printing Office, Washington, D.C., p. 344, 2010a.

Garcia, H. E., Locarnini, R. A., Boyer, T. P., Antonov, J. I., Zweng, M. M., Baranova, O. K., and Johnson, D. R.: Volume 4: Nutrients (phosphate, nitrate, silicate), in: *World Ocean Atlas 2009*, edited by: Levitus, S., NOAA Atlas NESDIS 71, U.S. Government Printing Office, Washington, D.C., p. 398., 2010b.

Großkopf, T., Mohr, W., Baustian, T., Schunck, H., Gill, D., Kuypers, M. M. M., Lavik, G., Schmitz, R. A., Wallace, D. W. R., and LaRoche, J.: Doubling of marine dinitrogen-fixation rates based on direct measurements, *Nature*, 488, 361–364, 2012.

Gruber, N.: The dynamics of the marine nitrogen cycle and its influence on atmospheric CO₂ variations, in: The Ocean Carbon Cycle and Climate, vol. 40 of NATO Science Series, chapt. 4, edited by: Follows, M. and Oguz, T., Kluwer Academic, P.O. Box 17,3300 AA Dordrecht, the Netherlands, 97–148, 2004.

5 Gruber, N. and Sarmiento, J. L.: Large-scale biogeochemical-physical interactions in elemental cycles, in: Biogeochemical/Physical Interactions in Elemental Cycles, vol. 12 of The Sea, chapt. 9, edited by: Robinson, A. R., Macarthy, J. J., Rothschild, B. J., J. Wiley and Sons, Harward University Press, 337–399, New York, 2002.

Guidi, L., Legendre, L., Reygondeau, G., Uitz, J., Stemmann, L. and Henson, S. A.: A new look at 10 ocean carbon remineralization for estimating deepwater sequestration, *Global Biogeochem. Cycles*, 29, 1044–1059, doi:10.1002/2014GB005063, 2015.

Halm, H., Lam, P., Ferdelman, T. G., Lavik, G., Dittmar, T., LaRoche, J., D'Hondt, S., and 15 Kuypers, M. M.: Heterotrophic organisms dominate nitrogen fixation in the South Pacific Gyre, *ISME J.*, 6, 1238–1249, doi:10.1038/ismej.2011.182, 2012.

Holl, C. M. and Montoya, J. P.: Interactions between nitrate uptake and nitrogen fixation in continuous 20 cultures of the marine diazotroph *Trichodesmium* (Cyanobacteria), *J. Phycol.*, 41, 1178–1183, doi:10.1111/j.1529-8817.2005.00146.x, 2005.

Kalvelage, T., Lavik, G., Lam, P., Contreras, S., Arteaga, L., Löscher, C. R., Oschlies, A., Paulmier, A., 25 Stramma, L., and Kuypers, M. M. M.: Nitrogen cycling driven by organic matter export in the South Pacific oxygen minimum zone, *Nat. Geosci.*, 6, 228–234, doi:10.1038/NGEO1739, 2013.

Kanakidou, M., Duce, R. A., Prospero, J. M., Baker, A. R., Benitez-Nelson, C., Dentener, F. J., Hunter, K. A., Liss, P. S., Mahowald, N., Okin, G. S., Sarin, M., Tsigaridis, K., Uematsu, M., Zamora, L. M., and Zhu, T.: Atmospheric fluxes of organic n and p to the global ocean, *Global Biogeochem. Cycles*, 26(GB3026), 1–12, doi:10.1029/2011GB004277, 2012.

25 Kasai, A., Kimura, S., Nakata, H., and Okazaki, Y.: Entrainment of coastal water into a frontal eddy of the kuroshio and its biological significance, *J. Marine Syst.*, 37, 185–198, 2002.

Key, R. M., Kozyr, A., Sabine, C. L., Lee, K., Wanninkhof, R. J., Bullister, L., Feely, R. A., Millero, F. J., 30 Mordy, C., and Peng, T.-H.: A global ocean carbon climatology: Results from Global Data Analysis Project (GLODAP), *Global Biogeochem. Cycles*, 18(GB4031), 1–23, doi:10.1029/2004GB002247, 2004.

Kim, I.-N., Lee, K., Gruber, N., Karl, D. M., Bullister, J. L., Yang, S., and Kim, T.-W.: Increasing 35 anthropogenic nitrogen in the North Pacific Ocean, *Science*, 346(6213), 1102–1106, doi:10.1126/science.1258396, 2014.

Koeve, W. and Kähler, P.: Heterotrophic denitrification vs. autotrophic anammox – quantifying collateral effects on the oceanic carbon cycle, *Biogeosciences*, 7, 2327–2337, doi:10.5194/bg-7-2327-2010, 2010.

Krishnamurthy, A., Moore, J. K., Zender, C. S., and Luo, C.: Effects of atmospheric inorganic nitrogen deposition on ocean biogeochemistry, *J. Geophys. Res.*, 112, 1–10, G02019, doi:10.1029/2006JG000334, 2007.

Krishnamurthy, A., Moore, J. K., Mahowald, N., Luo, C., Doney, S. C., Lindsay, K., and Zender, C. S.: Impacts of increasing anthropogenic soluble iron and nitrogen deposition on ocean biogeochemistry, *Global Biogeochem. Cy.*, 23, 1–15, GB3016, doi:10.1029/2008GB003440, 2009.

Krishnamurthy, A., Moore, J. K., Mahowald, N., Luo, C., and Zender, C. S.: Impacts of atmospheric nutrient inputs on marine biogeochemistry, *J. Geophys. Res.*, 115, 1–13, G01006, doi:10.1029/2009JG001115, 2010.

Lam, P., LavikaLavik, G., JensenJensen, M. M., van de Vossenberg, J., SchmidtSchmid, M., WoebkenWoebken, D., GutiérrezGutiérrez, D., AmannaAmann, R., JettenJetten, M. S. M., and KuypersKuypers, M. M. M.: Revising the nitrogen cycle in the Peruvian oxygen minimum zone, *P. Natl. Acad. Sci. USA*, 106, 4752–4757, doi:10.1073/pnas.0812444106, 2009.

Lamarque, J.-F., Kyle, G. P., Meinshausen, M., Riahi, K., Smith, S. J., van Vuuren, D. P., Conley, A. J., and Vitt, F.: Global and regional evolution of short-lived radiatively-active gases and aerosols in the Representative Concentration Pathways, *Climatic Change*, 109, 191–121, doi:10.1007/s10584-011-0155-0, 2011.

Lamarque, J. F., Dentener, F., McConnell, J., Ro, C. U., Shaw, M., Vet, R., Bergmann, D., Cameron-Smith, P., Dalsoren, S., Doherty, R., Faluvegi, G., Ghan, S. F., Josse, B., Lee, Y. H., MacKenzie, I. A., Plummer, D., Shindell, D. T., Skeie, R. B., Stevenson, D. S., Strode, S., Zeng, G., Curran, M., Dahl-Jensen, D., Das, S., Fritzsche, D., and Nolan, M.: Multi-model mean nitrogen and sulfur deposition from the Atmospheric Chemistry and Climate Model Intercomparison Project (ACCMIP): evaluation of historical and projected future changes, Atmos. Chem. Phys., 13, 7997–8018, doi:10.5194/acp-13-7997-2013, 2013.

Landolfi, A., Dietze, H., Koeve, W., and Oschlies, A.: Overlooked runaway feedback in the marine nitrogen cycle: the vicious cycle, *Biogeosciences*, 10, 1351–1363, doi:10.5194/bg-10-1351-2013, 2013.

La Roche, J. and Breitbarth, E.: Importance of the diazotrophs as a source of new nitrogen in the ocean, J. Sea Res., 53, 67–91, doi:10.1016/j.seares.2004.05.005, 2005.

Locarnini, R. A., Mishonov, A. V., Antonov, J. I., Boyer, T. P., Garcia, H. E., Baranova, O. K., Zweng, M. M., and Johnson, D. R.: Volume 1: Temperature, in: *World Ocean Atlas 2009*, NOAA

Atlas NESDIS 68, edited by: Levitus, S., U.S. Government Printing Office, Washington, D.C., p. 184, 2010.

[Loescher, C. R., Grokopf, T., Desai, F. D., Gill, D., Schunck, H., Croot, P. L., Schlosser, C., Neulinger, S. C., Pinnow, N., Lavik, G., Kuypers, M. M., LaRoche, J., and Schmitz, R. A.: Facets of diazotrophy in the oxygen minimum zone waters off Peru, ISME J., 8\(11\), 2180–2192, doi:10.1038/ismej.2014.71, 2014.](#)

5 Mahowald, N., Jickells, T. D., Baker, A. R., Artaxo, P., Benitez-Nelson, C. R., Bergametti, G., Bond, T. C., Chen, Y., Cohen, D. D., Herut, B., Kubilay, N., Losno, R., Luo, C., Maenhaut, W., McGee, K. A., Okin, G. S., Siefert, R. L., and Tsukuda, S.: Global distribution of atmospheric phosphorus sources, concentrations and deposition rates, and anthropogenic impacts, *Global Biogeochem. Cy.*, 22, 1–19, GB4026, doi:10.1029/2008GB003240, 2008.

10 Martin, J. H., Knauer, G. A., Karl, D. M., and Broenkow, W. W.: VERTEX: carbon cycling in the north-east Pacific, *Deep-Sea Res.*, 34, 267–285, 1987.

15 [Mohr, W., Grosskopf, T., Wallace, D. W. R., La Roche, J.: Methodological underestimation of oceanic nitrogen fixation rates, PLoS ONE, 5, doi:10.1371/journal.pone.0012583, 2010.](#)

20 Moore, C. M., Mills, M. M., Arrigo, K. R., Berman-Frank, I., Bopp, L., Boyd, P. W., Galbraith, E. D., Geider, R. J., Guieu, C., Jaccard, S. L., Jickells, T. D., LaRoche, J., Lenton, T. M., Mahowald, N. M., Marañón, E., Marinov, I., Moore, J. K., Nakatsuka, T., Oschlies, A., Saito, M. A., Thingstad, T. F., Tsuda, A., and Ulloa, O.: Processes and patterns of oceanic nutrient limitation, *Nature Geosci.*, 6, 701–710, doi:10.1038/NGEO1765, 2013.

25 Moore, J. K. and Braucher, O.: Sedimentary and mineral dust sources of dissolved iron to the world ocean, *Biogeosciences*, 5, 631–656, doi:10.5194/bg-5-631-2008, 2008.

20 Moss, R. H., Edmonds, J. A., Hibbard, K. A., Manning, M. R., Rose, S. K., van Vuuren, D. P., Carter, T. R., Emori, S., Kainuma, M., Kram, T., Meehl, G. A., Mitchell, J. F. B., Nakicenovic, N., Riahi, K., Smith, S. J., Stouffer, R. J., Thomson, A. M., Weyant, J. P., and Wilbanks, T. J.: The next generation of scenarios for climate change research and assessment, *Nature*, 463, 747–756, doi:10.1038/nature08823, 2010.

30 Mouriño-Carballido, B., Pahlow, M., and Oschlies, A.: High sensitivity of ultra-oligotrophic marine ecosystems to atmospheric nitrogen deposition, *Geophys. Res. Lett.*, 39, 1–6, L05601, doi:10.1029/2011GL050606, 2012.

[Noffke, A., Hensen, C., Sommer, S., Scholz, F., Bohlen, L., Mosch, T., Graco, M., and Wallmann, K.: Benthic iron and phosphorus fluxes across the Peruvian oxygen minimum zone, *Limnol. Oceanogr.*, 57\(3\), 851–867, doi:0.4319/lo.2012.57.3.0851, 2012.](#)

Okin, G. S., Baker, A. R., Tegen, I., Mahowald, N. M., Dentener, F. J., Duce, R. A., Galloway, J. N., Hunter, K., Kanakidou, M., Kubilay, N., Prospero, J. M., Sarin, M., Surapipith, V., Uematsu, M., and Zhu, T.: Impacts of atmospheric nutrient deposition on marine productivity: roles of nitrogen, phosphorus, and iron, *Global Biogeochem. Cy.*, 25, 1–10, GB2022, 2011.

5 Paerl, H. W. and Whitall, D. R.: Anthropogenically-derived atmospheric nitrogen deposition, marine eutrophication and harmful algal bloom expansion: is there a link?, *Ambio*, 28, 307–311, 1999.

Pennington, J. T., Mahoney, K. L., Kuwahara, V. S., Kolber, D. D., Calienes, R. and Chavez, F. P.: Primary production in the eastern tropical Pacific: A review, *Prog. Oceanogr.* 69, 285–317, 2006.

10 Reay, D. S., Dentener, F., Smith, P., Grace, J., and Feely, R. A.: Global nitrogen deposition and carbon sinks, *Nature Geosci.*, 1, 430–437, 2008.

15 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. Cy.*, 22, 1–21, GB1013, doi:10.1029/2007GB002953, 2008.

Seitzinger, S. P. and Kroeze, C.: Global distribution of nitrous oxide production and N inputs in freshwater and coastal marine ecosystems, *Global Biogeochem. Cycles*, 12(1), 93–113, doi:10.1029/97GB03657, 1998.

20 Seitzinger, S. P. and Sanders, R. W.: Atmospheric inputs of dissolved organic nitrogen stimulate estuarine bacteria and phytoplankton, *Limnol. Oceanogr.*, 44, 721–730, 1999.

25 Slomp, C. P. and Van Cappellen, P.: The global marine phosphorus cycle: sensitivity to oceanic circulation, *Biogeosciences*, 4, 155–171, doi:10.5194/bg-4-155-2007, 2007.

Somes, C. J., Schmittner, A., Galbraith, E. D., Lehmann, M. F., Altabet, M. A., Montoya, J. P., Letelier, R. M., Mix, A. C., Bourbonnais, A., and Eby, M.: Simulating the global distribution of nitrogen isotopes in the ocean, *Global Biogeochem. Cycles*, 24(GB4019), 1–16, 2010.

30 Somes, C. J., Oschlies, A., and Schmittner, A.: Isotopic constraints on the pre-industrial oceanic nitrogen budget, *Biogeosciences*, 10, 5889–5910, doi:10.5194/bg-10-5889-2013, 2013.

Su, B., Pahlow, M., Wagner, H., and Oschlies, A.: What prevents nitrogen depletion in the oxygen minimum zone of the eastern tropical South Pacific?, *Biogeosciences*, 12, 1113–1130, doi:10.5194/bg-12-1113-2015, 2015.

Tyrrell, T.: The relative influences of nitrogen and phosphorus on oceanic primary production, *Nature*, 400, 525–531, 1999.

Van Cappellen, P. and Ingall, E. D.: Benthic phosphorus regeneration, net primary production, and ocean anoxia: a model of the coupled marine biogeochemical cycles of carbon and phosphorus, *Paleoceanography*, 9, 677–692, 1994.

Van Mooy, B. A. S., Keil, R. G., and Devol, A. H.: Impact of suboxia on sinking particulate organic carbon: enhanced carbon flux and preferential degradation of amino acids via denitrification, *Geochim. Cosmochim. Ac.*, 66, 457–467, doi:10.1016/S0016-7037(01)00787-6, 2002.

Vet, R., Artz, R. S., Carou, S., Shawa, M., Ro, C., Aas, W., Baker, A., Bowersox, V. C., Dentener, F., Galy-Lacaux, C., Hou, A., Pienaar, J., J., Gillett, R., Forti, M. C., Gromov, S., Hara, H., Khodzherm, T., Mahowald, N. M., Nickovic, S., Rao, P. S. P., and Rei, N. W.: IA global assessment of precipitation chemistry and deposition of sulfur, nitrogen, sea salt, base cations, organic acids, acidity and pH, and phosphorus, *Atmos. Environ.*, 93, 3–100, doi:10.1016/j.atmosenv.2013.10.060, 2014.

Wallmann, K.: Feedbacks between oceanic redox states and marine productivity: a model perspective focused on benthic phosphorus cycling, *Global Biogeochem. Cy.*, 17, 1–10, 2003.

Wallmann, K.: Phosphorus imbalance in the global Ocean, *Global Biogeochem. Cy.*, 24, 1–12, GB4030, doi:10.1029/2009GB003643, 2010.

Ward, B. B., Devol, A. H., Rich, J. J., Chang, B. X., Bulow, S. E., Naik, H., Pratihary, A., and Jayakumar, A.: Denitrification as the dominant nitrogen loss process in the Arabian Sea, *Nature*, 461, 78–81, doi:10.1038/nature08276, 2009.

Warneck, P.: Chemistry of the Natural Atmosphere, vol. 71 of International Geophysics Series, Elsevier, New York, 1988.

Wise, M., Calvin, K., Thomson, A., Clarke, L., Bond-Lamberty, B., Sands, R., Smith, S. J., Janetos, A., and Edmonds, J.: Implications of limiting CO₂ concentrations for land use and energy, *Science*, 324, 1183–1189, doi:10.1126/science.1168475, 2009.

Zamora, L. M., Landolfi, A., Oschlies, A., Hansell, D. A., Dietze, H., and Dentener, F.: Atmospheric deposition of nutrients and excess N formation in the North Atlantic, *Biogeosciences*, 7, 777–793, doi:10.5194/bg-7-777-2010, 2010.

Zamora, L. M., Prospero, J. M., and Hansell, D. A.: Organic nitrogen in aerosols and precipitation at Barbados and Miami: implications regarding sources, transport and deposition to the western subtropical North Atlantic, *J. Geophys. Res.*, 116, 1–17, D20309, doi:10.1029/2011JD015660, 2011.

Table 1. Summary of model configurations including different processes. Process abbreviations are “N-DEP”, “Model BD and Data BD represent model- and data-based benthic denitrification”, respectively; “Model PR” and “Data PR are model- and data-based benthic phosphate regeneration, respectively”. N-DEP represents the atmospheric nitrogen input into the surface ocean according to the estimate by Lamarque et al. (2011); Model BD and Data BD represent model- and data-based benthic denitrification, respectively; Model PR and Data PR are model- and data-based benthic phosphorus regeneration, respectively.

| Configuration | Processes | | | | | N-DEP |
|---|-----------|---------|----------|---------|-------|-------|
| | Model BD | Data BD | Model PR | Data PR | N-DEP | |
| Control | | | | | | |
| N-DEP | ± | | | | + | |
| MBD | | + | | | | |
| MPR | | | + | | | |
| DBD | | | | + | | |
| DPR | | | | | ± | |
| Synthesis configuration configurations | | | | | | |
| MBD+MPR (Syn1) | | + | | + | | |
| DBD+DPR (Syn2) | | | + | | | |
| MBD+MPR+ N-DEP (Syn3) | + | + | | + | | |
| DBD+DPR+ N-DEP (Syn4) | + | | + | | + | |

± indicates that the process is included.

Table 2. Sediment percentage and average Martin curve value used for the model-based estimation Summary of benthic denitrification data-based flux estimates. “N-deposition” is the annual nitrogen input via atmospheric deposition; “Sediment percentage” is the percentage of the surface areas of the UM and D boxes in contact with the sediment; “Average Martin curve value fraction” represents the average of fractions (calculated from the Martin curve fractions) of export production reaching the sediment for each grid point of the topography data; “NPP” is the net primary production estimated from Aqua-Modis satellite data; “Data BD” and “Data PR” represent fixed-N loss via benthic denitrification and phosphate release via phosphorus regeneration in the UM and D boxes respectively. High-BD indicates that the full sediment of the D box is included to estimate NO_3^- loss via benthic denitrification and phosphate release via phosphorus regeneration.

| | N-deposition (Tg N yr^{-1}) | Sediment percentage (SD, %) | Average Martin curve fraction (AMC, %) | NPP ($\text{mg C m}^{-2} \text{ day}^{-1}$) | Rain rate | Data BD (Tg N yr^{-1}) | Data PR (Tg P yr^{-1}) |
|--------------------|---|-----------------------------------|--|--|-------------------|--------------------------------------|--------------------------------------|
| U box | 0.081 ^a 0.12 ± 0.052^b | | | | | | |
| S box | 1.4 ^a 1.6 ± 0.063^b | | | | | | |
| UM box | 0.81 ^c | 53.04 ^{c,d} | 1374.7 ^e | 148.4 ^f | 0.17 ^g | 0.058 ^h | |
| D box | 2.25 ^c | 12.51 ^{c,d} | 873.9 ^e | 12.2 ^f | 0.82 ^g | 0.056 ^h | |
| D box (high-BD/PR) | | | 873.9 ^e | 12.2 ^f | 8.8 ^g | 0.56 ^h | |
| Equations | | Eq. 2 | | Eqs. 3,4 | Eqs. 1 | Eqs. 5–9 | |

^aNitrogen deposition estimated from a chemistry-climate model by Lamarque et al. (2011).

^bAverage nitrogen deposition estimated from 23 atmospheric chemistry transport models by Dentener et al. (2006).

^c2-Minute Gridded Global Relief Data (http://www.ngdc.noaa.gov/mgg/gdas/gd_designagrid.html).

^dThe Martin curve exponent $b = 0.82$ is from Berelson (2001).

^eNPP is estimated according to Behrenfeld et al. (2005). PAR, K490, Chl a are from the Aqua-modis satellite data (<http://oceancolor.gsfc.nasa.gov/>); MLD data are from the Hybrid Coordinate Ocean Model (HYCOM, <http://orca.science.oregonstate.edu/1080/by.2160.monthly.hdf.mld.hycom.php>).

^fExport production is estimated from NPP and SST according to Dunne et al. (2005); SST is from the World Ocean Atlas annual average $1^\circ \times 1^\circ$ temperature (Locarnini et al., 2010).

^gBD is estimated from RRPOC applying the emeprical transfer function of Bohlen et al. (2012).

^hPR is estimated from RRPOC following the emeprical relationship of Wallmann (2010) and Flögel et al. (2011).

Table 3. Model flux comparison with model-based and observational estimates.

| Reference | N ₂ -fixation | | WC-denif (Tg N yr ⁻¹) | BD | PR (Tg Pyr ⁻¹) |
|--|--------------------------|---------------------|--------------------------------------|-------------------------|-------------------------------|
| | U box | S box | | | |
| This study ^a | 2.8–6.7 | 1.9–3.2 | 4.9–6.6 | 0.19–9.0 ^{f,g} | 0.062–0.62 ^{f,g} |
| Somes et al. (2010) ^{a,b} | 0.086 | 4.4 | 5.7 | 0.86 ^f | |
| Halm et al. (2012) ^c | | 1.4–21 ^e | | | |
| Bohlen et al. (2012) ^a | | | | 2.0 ^f | 0.34 ^f |
| Kalvelage et al. (2013) ^c | | | 10 | 1.2 ^h | |
| DeVries et al. (2013) ^{a,b} | 0.4±0.1 | 13±4.0 | 7.0±2.0 | 6.0±2.0 ^f | |
| Dekaezemacker et al. (2013) ^c | 0.023–0.30 ^d | | | | |
| Loescher et al. (2014) ^c | 0.22–18.7 ^d | | | | |

^aModel results.^bPersonal communication.^cObservational estimates.^dValue extrapolated to the area of the U box in our model.^eValue extrapolated to the area of the S box in our model.^fFor the whole sediment area below our model domain.^gTop 2000m of our model domain.^hTop 600m of the OMZ region.

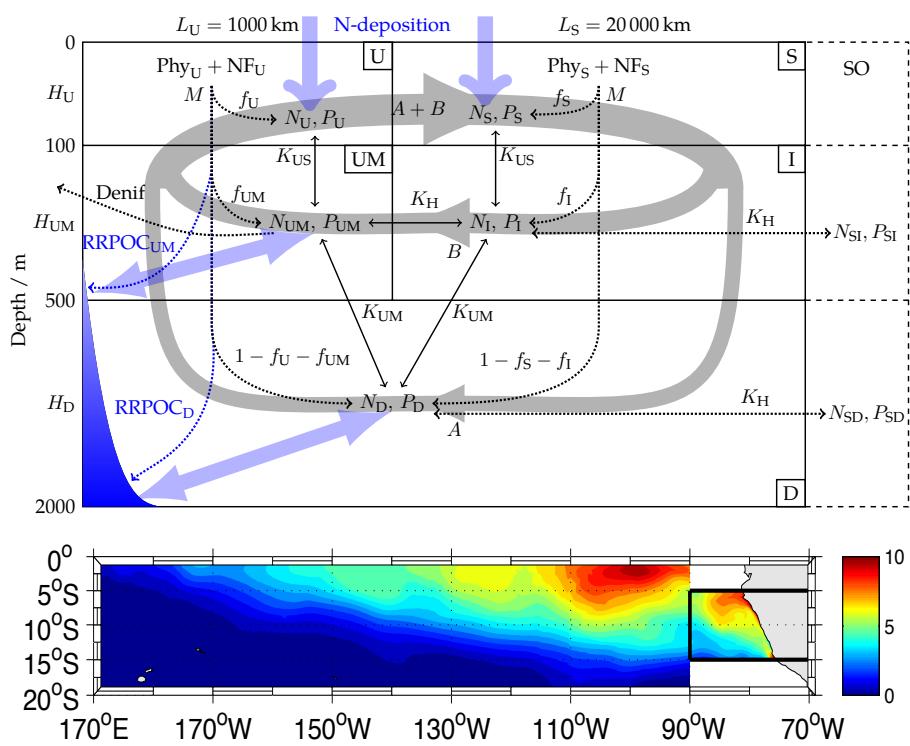


Fig. 1. Model structure. The model domain comprises five boxes representing the top 100 m of an upwelling region (U), the underlying oxygen minimum zone (UM), and an adjacent open-ocean basin divided into a surface (S) and an intermediate-depth box (I). A deep box (D) underlies both the upwelling region and the open ocean. The large-scale circulation is represented by deep (A) and shallow (B) convection (thick grey lines). Mixing between boxes is implemented via mixing coefficients (K). Remineralisation derived from primary production by ordinary (Phy) and diazotrophic (NF) phytoplankton in the surface boxes consumes oxygen. Under anoxic conditions remineralisation is fueled by anaerobic remineralization (Denif). In the configuration employed in this study, the model domain exchanges nutrients and oxygen with the Southern Ocean (right, denoted as “SO”). Nitrogen deposition and benthic remineralization are included additionally to represent their influence on the local water-column nutrient concentrations (thick light blue arrows). The graph above is a schematic figure of our model domain; the graph below shows the surface of our model domain, and the colorbar is nitrate concentration in $\mu\text{mol L}^{-1}$.

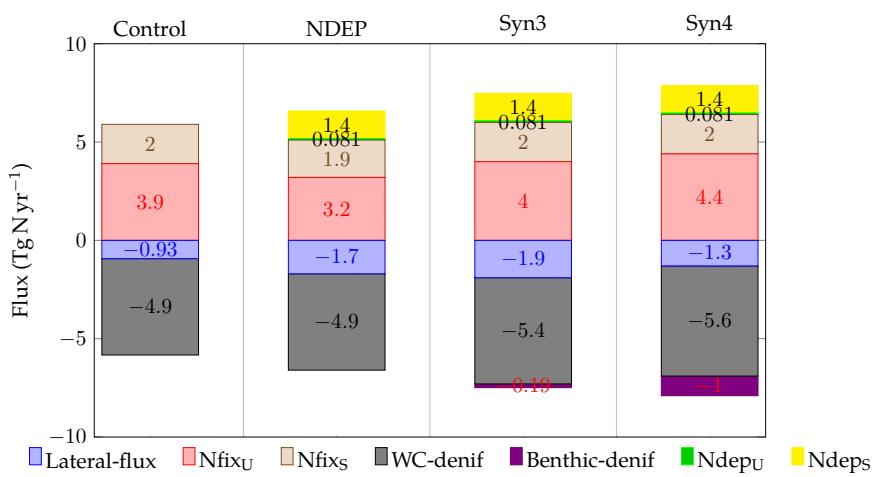


Fig. 2. Nitrogen fluxes after including atmospheric nitrogen deposition in the control, Syn3 and Syn4 configurations defined in Table 1. Lateral-flux identifies is the nitrogen efflux or influx through the southern boundary; N-fix represents Nfix_U and Nfix_S represent the nitrogen fixation rate by NF respectively in the U and S boxes; WC-denif is water-column denitrification; N-dep is Ndeps and Ndeps are the nitrogen input into surface ocean U and S boxes via nitrogen deposition.

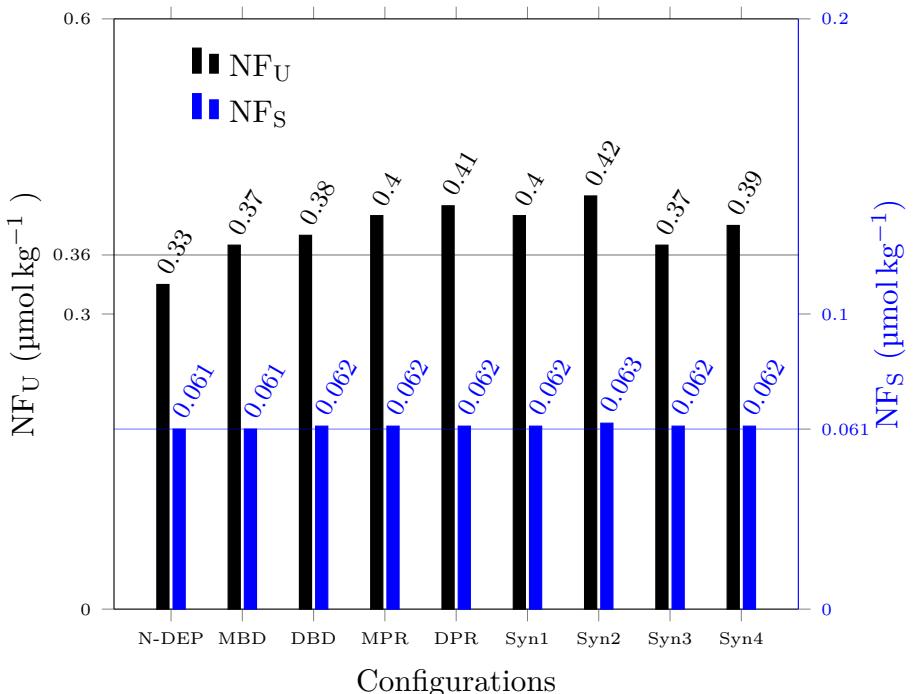


Fig. 3. Sensitivity of simulated steady-state concentrations of nitrogen fixers NF_U and NF_S in the U and S boxes respectively. Horizontal grey and light blue lines represent the NF_U and NF_S concentrations in the control configuration respectively. Syn1, Syn2, Syn3 and Syn4 denote the “MBD + MPR”, “DBD + DPR”, “MBD + MPR + ~~N-DEP~~NDEP”, and “DBD + DPR + ~~N-DEP~~NDEP” synthesis configurations defined in Table 1.

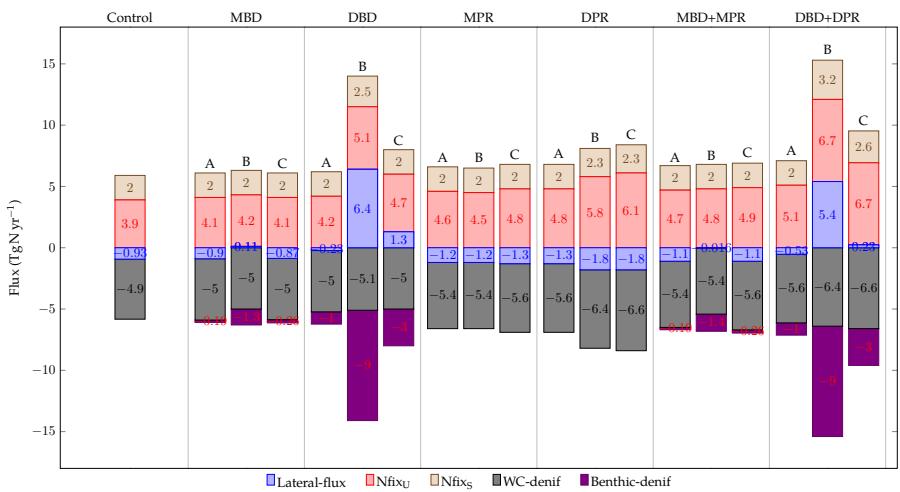


Fig. 4. Nitrogen fluxes after including benthic denitrification or/and phosphate-phosphorus regeneration. Lateral-flux identifies is the nitrogen efflux or influx through the southern boundary; N-fix Nfix_U and Nfix_S represents the nitrogen fixation rate by NF in the U and S boxes respectively; WC-denif is water-column denitrification; Benthic-denif represents the fixed-N loss via benthic denitrification in the model domain. Bar labels: A, main experiments; B, sensitivity experiments with high-BD; C, sensitivity experiments with Martin Curve exponent $b = 0.4$.

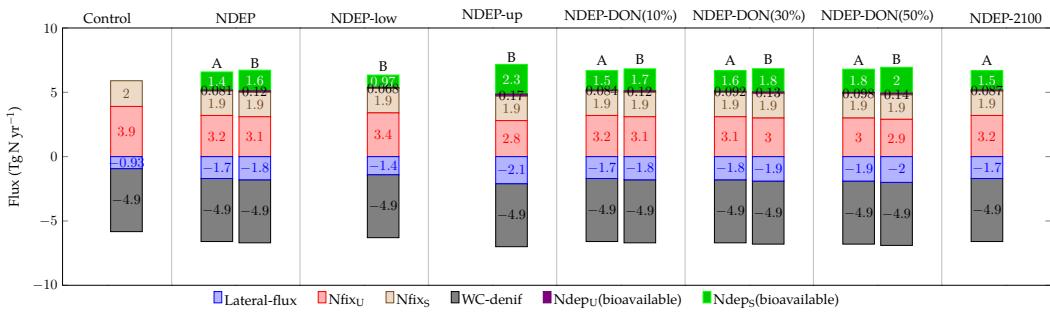


Fig. 5. Sensitivity of nitrogen fluxes to atmospheric inorganic and organic nitrogen deposition and associated uncertainties. Lateral-flux is the nitrogen efflux or influx through the southern boundary; Nfix_U and Nfix_S represent the bioavailable nitrogen fixation rate by NF in the U and S boxes respectively; WC-denif is water-column denitrification; Ndep_U and Ndep_S are the nitrogen input into surface ocean (U and S boxes) via nitrogen deposition. Bar labels: A, nitrogen deposition data from Lamarque et al. (2011); B, nitrogen deposition data from Dentener et al. (2006). In NDEP-low and NDEP-up, the lower and upper limit of nitrogen deposition fluxes are included; in NDEP-DON(10%), NDEP-DON(30%) and NDEP-DON(50%), the bioavailability of deposited DON is assumed to 10%, 30% and 50% respectively; in NDEP-2100, nitrogen deposition is estimated according to the RCP8.5 scenario projections for 2100 (Lamarque et al., 2011).

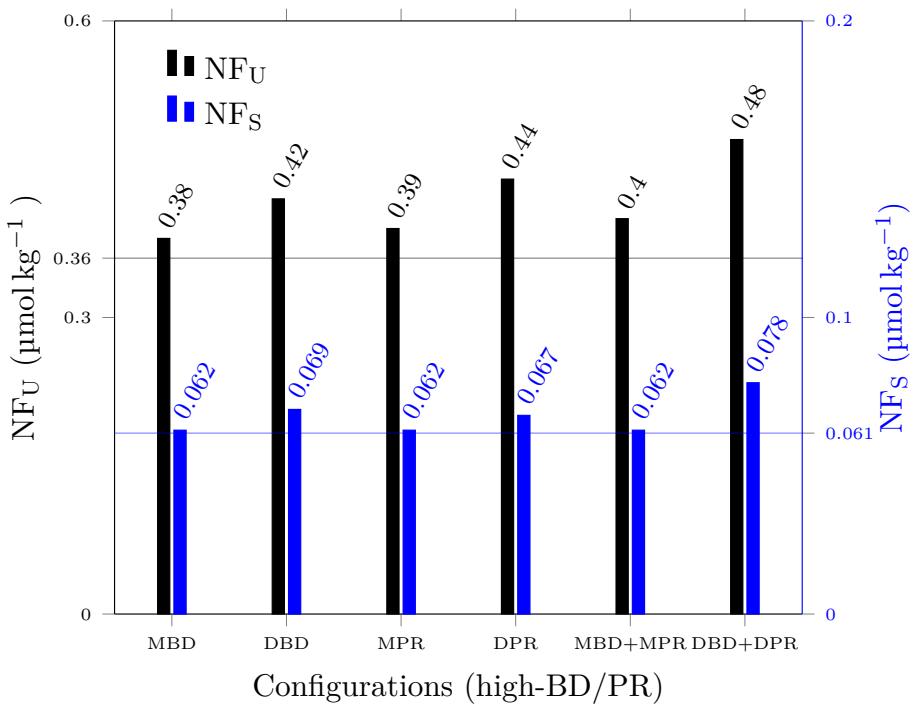


Fig. 6. Sensitivity of simulated steady-state concentrations of nitrogen fixers (NF_U and NF_S) in the U and S boxes respectively after incorporating high-BD and high-PR. Horizontal grey and light blue lines represent the NF_U and NF_S concentrations in the control configuration.

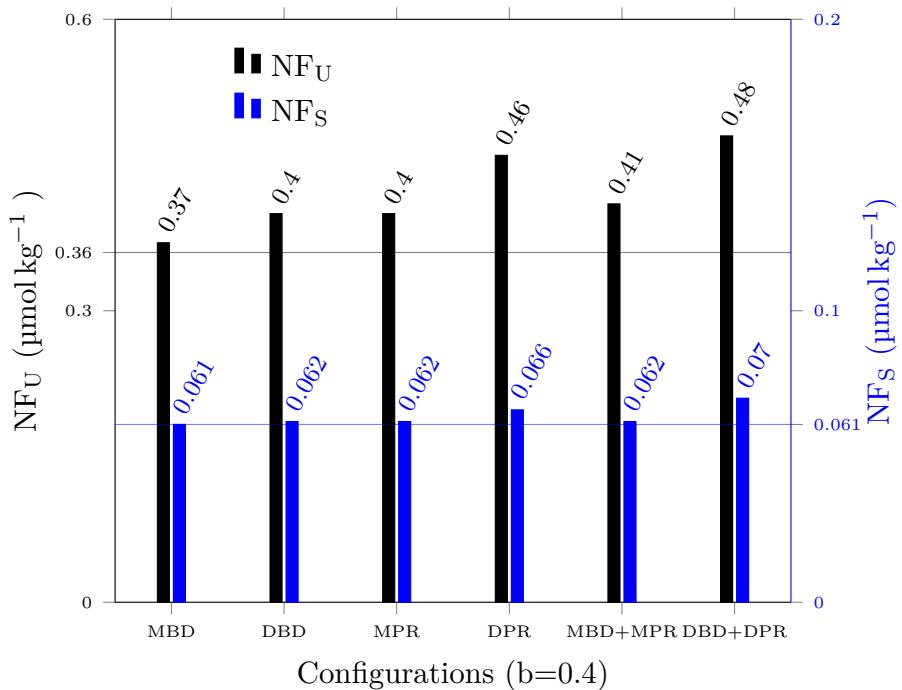


Fig. 7. Sensitivity of simulated steady-state concentrations of nitrogen fixers (NF_U and NF_S) in the U and S boxes respectively after applying $b = 0.4$ for Eq. (2). Horizontal grey and light blue lines represent the NF_U and NF_S concentrations in the control configuration.

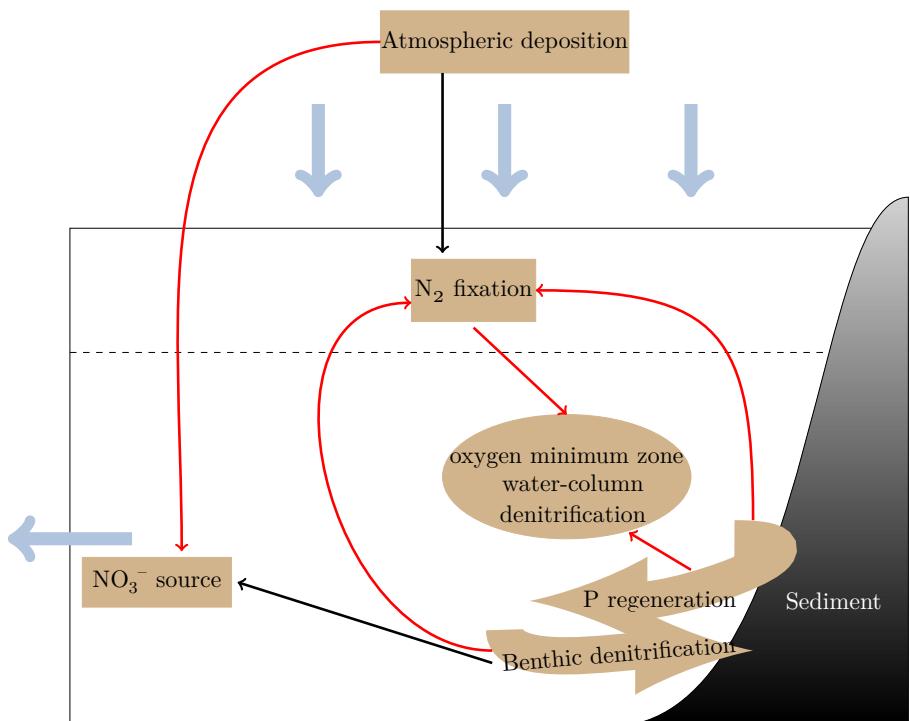


Fig. 8. Schematic of the model sensitivity to different processes related to the nitrogen budget of the ETSP. The red solid lines ~~present represent~~ stimulatory effects, and the black solid lines represent depressive effects.

~~Nitrogen fluxes after including atmospheric nitrogen deposition in the model with facultative fixation. Labels are the same as those in Fig. 2.~~