

Response to Referees

Marco van Hulst et al.

15th December 2016

General response

All three reviews contain useful comments that resulted in discussion among the authors. Important themes of the discussion were the omission of a biological cycle and O_2 and pH dependency, as well as moving the focus on the observational data.

The observations at the GA02 transect have not been published and already give some of the insights reached or confirmed by the model runs. For this reason we restructured the paper, giving more space for a discussion on these observations.

As the reviewers correctly remarked, and as we acknowledged in our discussion paper, Mn does play an important role in biology. Therefore, we have added a biological cycle but a dependency of Mn redox on pH and O_2 was not possible to do within a reasonable amount of time.

In the public peer review, we suggested that we also use a different dust deposition field (reasons mentioned therein). However, finally, we have decided to stick with the original dust deposition field of Hauglustaine et al. (2004). While it would be beneficial to the surface dMn distribution, especially in the Pacific, if we were to use Mahowald et al. (1999), the Hauglustaine deposition is more similar to the broadly accepted dust flux of Jickells et al. (2005). As a consequence, our simulation is less accurate and highlights even more the regions where the model has shortcomings (especially the Pacific Ocean).

We are all very much aware that the reality in the oceans is far more complicated than what is actually being measured by seagoing oceanographers, and hence being simulated in models like ours. Most notably, all colleagues are aware that there exists for Mn, and other metals like Fe, a spectrum of colloids in a suite of size classes. If field data were available for at least one pool of colloidal Mn, then it would be worth considering to include such a separate pool in the model design. However, the datasets available thus far have been almost exclusively only for 0.2 micron or 0.4 micron seawaters filtered, i.e. operationally defined dissolved Mn, and one small dataset of particulate Mn, that is the Mn captured on a filter. Accordingly, the simulation model includes dissolved Mn and particulate Mn but, still, no colloidal Mn pool. More generally, the complexity of the model should not be (much) larger than what the observations can constrain. Of course, more extensive modelling and observational studies should be done to arrive at a more complete knowledge of Mn, but the current study is only a first model.

The following issues are finally different from what would have been if taking the Discussion Paper and then applying everything according to our response in the public peer review.

- We increased the dust dissolution flux from 30 to 40% in our new simulation, which is well within the observational range. This seems reasonable, because we included an extra sink of biological incorporation.

- The “in preparation” papers are in the meantime published.
- We did, after all, improve the comparison plot by changing the choice of colours in the concentration scale bar.
- We moved Figure 11 to the Results section, and discussed *OxidThreshold* as a proper simulation.

Our original responses in the public peer review now follow, with minor updates at places where original text is struck through. Furthermore, references to figures, tables and equations now refer to those in the Discussion Paper. And the tense is changes, as we finalised the new version of the manuscript.

Response to Anonymous Referee #1

We wish to thank the reviewer for the succinct analysis and criticism on our model.

The threshold M_{ox} that is used to account for the homogeneous background concentration of dissolved Mn of about 0.10 nM to 0.15 nM observed throughout most of the deep ocean, may result from over simplification of the model. There might be very different K_{ox} and K_{red} values at upper (above ~ 300 m) and deeper (below ~ 300 m) part of the ocean. M_{diss} may be mainly derived from remineralization of sinking organic matter in the upper ocean, and from an equilibration with colloidal or fine particles via absorption/colloid formation processes in the deeper ocean. One would expect very different k_{red} values at different depths.

The reduction and oxidation processes are indeed simplified a lot. We decided to devise a model that is as simple as possible, but reproduces reasonable dissolved Mn concentrations, and at the same time teaches us something useful. For this we have set k_{ox} to a constant, but realised on the outset that k_{red} must be much larger in the photic zone. Thus our most simple and still reasonable model was one where the oxidation rate is a constant and the reduction rate a two-valued step function (Eqs 7–9). Still, we do realise that bacterial activity, oxygen minimum zones and colloids influence the k values. We wrote that the dependence of oxygen would be advisable for future development, but decided that this was not needed for this first study, nor necessary to arrive at our conclusions. The influence by colloids would be difficult to verify, because the observations that we used do not include colloidal and “truly dissolved” fractions: the “dissolved Mn” in both the observations and the model includes both fractions. This means that the k values are really the effective k ’s between the operationally defined dissolved and particulate pools.

Concerning the relatively homogeneous background concentration, this could not be reached by only setting constant values for k_{ox} and k_{red} in the deep ocean. It is rather a consequence of a Mn oxide concentration threshold on the increased settling velocity (Eq. 11). There are two ways to look at this part of the model. One is to see it as a trick to get the right deep-ocean dissolved Mn concentration. The other is the interpretation of the threshold being a necessary minimum concentration of Mn oxides before aggregation and efficient settling can occur. Both are true, but the second is the interesting one here because it gives an actual, potential explanation for the constant background concentration.

In addition, M_{diss} may be mainly removed from the water column via oxidation to insoluble Mn(IV) with a rate that decreases with increasing depth due to lower dissolved oxygen concentration and lower pH at deeper depths, leading to very different k_{ox} values at these different

depths. Thus both k values and their ratios $K_{\text{red}}/k_{\text{ox}}$ are not homogeneous throughout the water column. Such difference may cause lower modelled M_{ndiss} than the observed values, thus requiring a threshold M_{nox} to account for higher M_{ndiss} at deeper depths.

Yes, in areas with lower oxygen concentrations and lower pH, the k_{ox} would be lower, resulting in a higher dissolved concentration which is what we want when the threshold were to be removed from the model. However, at the moment we think that nowhere in the deep ocean k_{ox} is actually low enough to accomplish this in our model. This may very well be related to the high deep-ocean settling velocity we chose in our model. However, if we would decrease this velocity, the hydrothermal plumes would extend too far (Section 4.2). If we would then furthermore reduce the hydrothermal input, the hydrothermal signals would not be reproduced by the model. Furthermore, as Fig. 16 shows, we have chosen at least the $k_{\text{ox}}/k_{\text{red}}$ ratio quite well, meaning that while minor features are likely to be improved by using an inhomogeneous k_{ox} , it could never capture the much larger effect that we achieve by the threshold on the increased settling velocity.

Response to Anonymous Referee #2

We wish to thank the reviewer for the analysis and suggestions on our manuscript.

While I am very supportive of the objectives of this work in general, the scope of the discussion, the wording and overall presentation of this manuscript gives the impression that the model is not sufficiently mature: at this stage of development, this paper reads more like a progress report than a fine-tuned, well-thought-out, scientific manuscript. The story in this manuscript announces a great start for Mn modelling, but comes across a bit short. Specifically, excluding the biological aspect of Mn cycling is a major omission (the authors themselves acknowledge that this is a major aspect of Mn cycling).

While the model does not include all the interesting processes, we believe that the model is sufficiently mature to publish as a first study. Concerning the biological Mn cycling, we agree and now have included a biological module in our simulation.

As a reader, it is not totally clear what has been learned about the Mn cycle by reading this manuscript. Interesting points are made in the last few lines of the abstract, but these are unfortunately not developed fully in the text. Most of the discussion focuses instead on technical limitations of the model and on a handful of parameterizations used to modify the model to make it fit data – with variable success. Since the bulk of the model presented here is essentially a scaled version of the existing companion Fe-model, the scope of the discussion presented here is somewhat limited.

We believe that our model can be used as a starting point for studies that teach us about the Mn cycle. In other words, this model is a first model that should be primarily considered as a proof of concept and basis for further study. Such further study is seen as doing more, and other type, of field measurements, as well as further development of the simulation model. These should go hand in hand, given the currently available type and amount of field data a more complex model would be overkill. However, we do put forward several insights in the cycling of Mn in the ocean (which should be considered as further supporting the proof of concept). This could still be considered somewhat limiting, which is why we decided to include more discussion in the paper by a more extensive discussion on the GA02 transect that has not been published in a paper before.

The four bullet points at the end of the abstract follow from, or are strongly suggested by, the manuscript. Our first point concerning the high concentration in the upper ocean was already to some extent established in the literature. Our second point on the deep AMOC transport is illustrated by both the observations and the model; the model makes it more intuitive because of the high deep settling velocity. Point three on the background concentration states what the model shows, and then goes on that this supports the idea that a minimal concentration of Mn oxides are needed before significant removal occurs (see simulation *NoThreshold*). It is outside the scope of this modelling study to test this; rather laboratory and/or field experiments are needed for this (and maybe different type of modelling that includes a more mechanistic description of aggregation). Point four on the hydrothermal signal also follows from the paper (see simulation *LowHydro*).

One aspect of the writing that is not helping convincing the reader that this is a conclusive piece of work, is the over-reliance of subjective statements to describe the simulations and on imprecise/loose statements. This undermines the results. For example, the model fields are repeatedly qualified as “realistic”, but this really doesn’t mean anything, as there are just as many paragraphs in the paper outlining model flaws. The figures also reveal quite a few regions where the model does not fit the data. There is also no real set of criteria that are presented to define what “realistic” means. The authors should instead define the features their model fits or doesn’t fit directly, without trying to oversell their product as “realistic”, and explain in a bit more detail why the model fits or doesn’t fit particular features. Other examples are: “The model reproduces observations accurately”, “This is mostly captured by the model”, “It is probably difficult to improve this feature of the model”, “Reference simulation gives a reasonably realistic distribution”, “LowHydro appears to give a much worse prediction”, “This first-order approach works well mostly, but shortcomings can be identified”, “the flux may be overall underestimated”, “this may still contribute to the dissolved pool in the bottom water”, “underestimation may be due to too strong vertical mixing”, etc. . . . Some of these propositions could be tested explicitly by the authors using additional sensitivity studies or appropriate diagnostics to produce objective conclusions instead of suggestions.

In the final version we will try to refrain from subjective statements, and make them more objective with statistics or other analyses, or remove them completely. The statement “LowHydro appears to give a much worse prediction” is subjective on its own, but it is supported by our statistical analysis.

One way forward could also be to develop a section that discusses the observations in more detail (i.e. expand what is currently in the Introduction and Figure 1 and tie this more closely to what the modelling section aims to achieve). Appendix A spends quite a bit of effort describing how measurements were made. Showcasing these (unpublished?) measurements in more depth early in the manuscript would help build a sense of expectations with regards to what the model is expected to do or not do. That could also be used as a “roadmap” to explain how the paper is organized and why. It is also not totally clear why the authors focus on only a few data sections in their discussion. Figure 5a&b show that the model struggles to fit observation in the upper Pacific on P16. This seems certainly worthy of further examination and commenting.

It would be a good idea to discuss all the observations, and also using all of them for the statistics; but we decided to focus on the GA02 transect. You are right that we can discuss the observations in more detail. The GA02 transect on which we focus has not been published before (except as a data product on the GEOTRACES website). It now has been decided among the co-authors, that

the field data and its primary interpretations, now will become part of this simulation modelling paper, instead of thus far chosen by a separate paper. We will restructure the paper such that the discussion of these observations is more extensive and prominent than it is now in the paper.

Other observations are used in a visual (more subjective) way, and detailed discussion and statistics on comparing those with the model (and GA02 observations) is not straightforward because they use other methods or are not intercalibrated. Future studies could include many more measurements for a statistical comparison; this would imply the prerequisite of evaluating every dataset for accuracy and consistency with the others. In the future there will also be more GEOTRACES sampling cruises in the Pacific Ocean and other basins. Analyses from those samples will be following the same quality protocols, thus be more suitable for model–data comparison than many of the already existing observations.

Since the authors emphasize goodness of fit as the main theme throughout their paper, it is not clear what is gained from the simulations in sections 3.2 & 3.3. (why these experiments were chosen and not others?) It is clear from Table 5 what the results of these sensitivity experiments are. In that sense, little new insight is gained from reading section 4.2 and 4.5. Discussion section 4 is also generally focused on outlining model flaws and omissions. While this honesty from the authors is appreciated, this discussion section has less scientific value than it could have as it caters more to the model developers themselves than to the general public interested in the Mn cycle.

Sections 3.2 shows that we need the combination of high hydrothermal input and high export to get such a good simulation as described in Section 3.1. Section 3.3 shows that it is not as simple as that: the high export needs to be limited to get the correct background concentration. We could have chosen other simulations as well to test further properties of the model, but we think that the chosen sensitivity simulations are the ones that provide the most important insight in the model. A general note on this: it is often useful to start with a realistic simulation, and do sensitivity simulations based on this (good) reference simulation.

Specific comments

- Please refrain from listing all the “in preparation” papers in the Introduction. The general public doesn’t have access to these manuscripts at this point and there is no guarantee “in prep” papers will be published. Since these “in prep” papers seem to be review papers, a few well-chosen published references would be much more appropriate.

Most “in preparation” papers in the Introduction are to be published in a special issue of the recent Royal Society GEOTRACES conference that connects closely with the goals of this paper. That special issue is very likely to be published before our Mn modelling paper. We also think that for the Introduction these review papers are the most useful for the reader, because they give an overview to all state-of-the art knowledge, and contain useful references. We will remove the citation to the Middag et al. “in preparation” paper from the manuscript, and instead what was aimed to be in that separate paper will now also be added to the current paper.

- The first line of the Introduction argues that Mn uptake by phytoplankton is important, even “crucial for photosynthesis”. Yet, the argument for not modelling the biological aspect of Mn cycling is that “not enough data is available to constrain the processes (L2223, p22)” and because “there is no clear evidence for typical uptake-reminalization processes” (L15, p26). It

seems that testing these ideas is very much what the value of a Mn model would be? Also, how can you say Mn is “crucial for photosynthesis” in the Introduction if there is no information available on biological uptake?

We know that Mn is needed in organisms, and it plays multiple roles in photosynthesis. We also have estimates of inner-cell Mn from Twining and Baines (2013). This means that Mn is incorporated into the cell during growth, but we do not know how this process takes place and what happens with the Mn after it is incorporated. Possibly it is very similar to what happens to phosphate, so we decided to simulate a biological Mn cycle by following P, using a Mn:P ratio from Twining and Baines (2013), consistent with Middag et al. (2013). In this way, we have tested the effect of the trivial case of an uptake-remineralisation proportional to phosphate. First results of this simulation show that it may be relevant for the equatorial region and at mid-latitudes, especially in the Pacific Ocean. We will include and discuss further this result in the paper.

- P22, L10: “the k values in our model are optimised to get values. . .”. How is this “optimisation” done? Is optimisation really what was done?

No, we did not use the right terminology here. We set $k_{\text{red,light}}$ simply to the mean value found by Sunda and Huntsman (1994), then used the dissolved and particulate profiles in Fig. 4 of Bruland, Orians and Cowen (1994) to derive k_{ox} and $k_{\text{red,dark}}$.

- For figures 5, 6, 7, 9 that compare simulations to data, consider adding panels showing the relative errors achieved by the model simulation with respect to the data. This would be helpful to interpret the meaning of the bulk statistical fit diagnostics reported in Table 5 and understand misfit patterns in more detail. The color scheme chosen is also not that great for these figures as it is very hard to distinguish dark blues from dark greens, even if these colors would imply errors of order 200–300 %.

~~We already spent quite some time in producing good comparison plots, and do not think we can improve on these notably.~~ Indeed, improvement was possible: we have now adjusted the colour palette; this has improved the visual comparison. Apropos, the statistics support the comparison in a quantitative manner.

- How does the Mn inventory evolve from one sensitivity study to the next? These sensitivity runs would make more sense if the global inventory of Mn was kept constant. Maybe discuss the inventories as part of Table 3.

We have added the inventories to Table 3. If one does a sensitivity simulation, it is not straightforward what is the best way to keep the inventory invariant compared to the reference simulation, or if this would be very useful. In the case of *NoThreshold* we want to see the change of inventory. In our new simulations, the amount has decreased compared to *Reference* from 440 to 373 Gmol.

- Appendix B, p29. From what I understand in this section, it seems that the interpolation of the model simulations to the sampling location is done in different ways. If that’s in fact the case, why?

Yes, for the horizontal coordinates we used the sampling locations, while for the vertical we did either nothing (for the plots) or used the model grid (for statistical comparison). For the plotting the reason for interpolating the model to the station coordinates was that there needed to be some

kind of interpolation onto a grid that includes the sampling locations; the simplest choice was the minimal one where the new grid is defined by the station coordinates. The reason for the statistics is that while we preferred a comparison of the model at the sampling locations, this would be less trivial in the vertical, since the sample depths vary per station. Since the vertical spacing of the model is similar to that of the samples, we decided to interpolate the observations onto the vertical model grid.

- Why is the dependency of Mn on O2 not considered? (p11, L9)

It would be better to include such a dependency, as the effect is clear in, for instance, the Pacific Ocean. Johnson et al. (1996) had tested this on a local scale. We decided not to repeat this exercise, even though it would be sensible to include it at some point in global Mn models. We found it not to be essential for our purposes at this point. The downside of adding such a dependency is that it would make the model more complicated. It would imply relying on the circulation and oxygen distribution of a biogeochemical carbon model that has uncertainties of its own. So far it has shown quite difficult to simulate OMZs and its effects in an accurate way. However, we have introduced biological cycling of Mn, akin to phosphate.

- Section 2.2.6. Can Mn scavenge on other particles besides autigenically formed Mn oxides in this model? Is the lack of particles in deep water what you are trying to approximate with the “aggregation threshold”?

This may be the case. We believe that a more mechanistic model than the one presented in the current manuscript would be much more difficult, as it should include several size classes of particles, different types of manganese and other complications. This should be done, but it is unfeasible for the current study.

- On p11, L21, you say “manganese oxide is buried when arriving at the ocean floor, which means that it is removed from the model domain”. If that is the case, what is the source of the sedimentary Mn from section 2.2.3, eq (3)? Is the amount of Mn buried substantially less/more(?) than the sedimentary Mn source? how do these compare? Is the sediment source totally decoupled from the burial sink in this model? Is that decoupling justified?

It is indeed decoupled. Coupling with a sediment model may be useful for future studies. We have compared burial with forced Mn input and said something about it in the updated manuscript (Section 4.1). ~~If useful, we might add a burial figure as well.~~ We decided to not add a burial figure. We found that the sediment source is large at shallow sediments, while the hydrothermal source generates by far most of the Mn burial. This is probably realistic, and suggests that coupling water column and sediment may not be useful, or very difficult, for a global study.

Response to Anonymous Referee #3

We wish to thank the reviewer for the critical but also positive comments on our manuscript. The reviewer said that this

approach of starting [with a] simple [model] and increasing complexity only after thorough analysis of model deficiencies is laudable. But it also means that this work is likely to be just a first step, and that more complex models will follow.

This is absolutely the idea. He or she continues:

The main victim of this approach of starting simple is that the model disregards the biological cycling of manganese, although it is known that manganese availability is necessary for oxygenic photosynthesis, and can be (co-)limiting to phytoplankton growth at times (Brand et al. 1983, Bruland et al. 1991). The authors argue that in the data they use for validating the model, there is no strong signal of uptake and remineralisation of Mn. I must admit that I am sceptical about this assumption:

- firstly, is the argument to neglect of biological Mn cycling maybe biased by using primarily Atlantic data? The secondary maximum of dissolved Mn in the North Pacific, shown in Bruland et al. 1991 seems hard to explain without remineralisation of Mn in biological material, and – in addition – a slower Mn(II) reduction in the oxygen minimum. This at least is what I have gotten out of the 1-d modelling study by Johnson et al. 1996.

We now have decided to include a module for biological cycling in the model, and results of first simulations including such a biological module are encouraging. Yes, right now we are missing the secondary maximum. This is especially clear in the Pacific Ocean where there is not only a secondary maximum around 800 m, but also a higher concentration in the upper 250 m; neither are captured by our model. There should really be a study more dedicated to these issues, including indeed the uptake-remineralisation processes as well as the effect of oxygen minima into consideration. Future GEOTRACES sampling expeditions will also help in that they would be following the same quality standards as, amongst other GEOTRACES transects, the GA02. Nonetheless, there are a large number of processes and uncertainties when doing a first model. Thus, we believe that for a first global model study it is not too bad that we chose a more simple approach.

- secondly, how consistent is the neglect of biological cycling with the magnitude of other vertical fluxes of Mn in the model? At least an estimate for the magnitude of the vertical export of Mn through biology could be obtained by multiplying Mn:P ratios in phytoplankton (Sunda and Huntsman, 1998, Twining 2013) with common estimates of global export production. The vertical sinking flux of particulate Mn (only the precipitated form, not the non-dissolved lithogenic part) from the model could be compared to that number.

Indeed the biological module that we now have included is based on a typical or world-average Mn/P element ratio of plankton.

There are other parameterisations in this model that are less ad-hoc than they appear at first sight: The authors use a constant rate for Mn(II) oxidation and a reduction rate for Mn(IV) that switches abruptly from a high value in the euphotic zone to a low value below. Although this is qualitatively justified by the existence of photoreduction, one wonders where the values come from, until one finds in the discussion that this simple parameterization was chosen to reproduce the particulate/dissolved Mn ratio at the Vertex site. I think figure 11 and the corresponding explanation should be shifted to the model description. Instead, in the discussion, I think the authors could perhaps discuss what they think is the driving process for this reduction and whether it is likely that the Mn(IV)/Mn(II) ratio in the deep ocean is the same in different ocean basins.

Yes, the abrupt change from high to low reduction from the photic to the aphotic zone is ad hoc. In retrospect, we could have used a continuous function of depth, which would probably even gotten a higher correlation index, but the choice of the function is not constrained by the literature. (Linear in light intensity may be a good approximation.) We moved Fig. 11 and the description to the Results section (now Fig. 16); it's better organised now, with *OxidThreshold* having a proper/more prominent place in the paper.

My last criticism concerns the parameterisation of the sinking removal of oxidised Mn; contrary to the spirit of starting a model with simple assumptions, I find this parameterisation not simple at all. At the surface and at concentrations below 25 pM, Mn particles are assumed to sink at 1 m/d. Has this value been estimated from some assumption on the size of the particles, using Stokes' law, or is it just an arbitrary choice? Most of the initially formed particles probably are so small they don't sink at all on their own and need aggregation with other organic particles to sink, but most aggregates sink quite a bit faster. The model assumes that above the threshold of 25 pM, Mn particles aggregate and sink faster, but the increasing sinking rate above the threshold is made dependent on water depth, so that it does not affect the removal of Mn near the surface. Is there any justification for the depth dependency of the sinking rate? Several biogeochemical models use a vertically increasing sinking rate for its detritus compartment, as an implicit way to account for aggregation, see e.g. Kriest and Oschlies 2010, but the absolute sinking rates for aggregates in these models are much higher than assumed here. I found the choices here quite ad-hoc and wondered how strongly the model results are affected by them.

The removal of oxidised Mn is somewhat arbitrary indeed. Most Mn probably sinks slowly with small lithogenic particles (p. 24, lines 6–9). This could not explain the strong removal rate near hydrothermal vents. One approach would be to include more species of particulate (and maybe dissolved) manganese. We chose to stick with only (observationally defined) two pools, namely dissolved and particulate manganese. This meant that we needed to set a high velocity near hydrothermal vents, but the removal needed to be small enough to maintain high surface concentrations and the (lower) background concentration. Our parameterisation works for this.

Summarizing, I think that the authors need to discuss the processes that lead to the vertical transport and ultimately removal of Mn in their model more, before the paper can be published. They have shown with their sensitivity runs that aggregation seems to be important for the removal of Mn from hydrothermal plumes, and that a lower threshold for this aggregation seems to be important for explaining the relatively uniform Mn background in the deep ocean. But if indeed aggregation is the main vector of vertical transport, then one might suspect that near the surface, biological export production plays a large role in determining the vertical Mn flux, either directly from incorporated Mn, or indirectly from driving aggregation. That would lead to a much less uniform vertical transport velocity and it would be good to have an idea how much that could influence model results.

In the photic zone the sinking speed equals 1.0 m/d in the model, while higher speeds can be found around and below 100 m depth. That said, there may already be some aggregation directly below the photic zone in the model, because accumulated (dissolved) Mn in that region quickly oxidises, hence exceeding the 25 pM threshold and having a higher speed than in the photic zone. This may be expected in the real ocean as well, but we cannot be sure whether our model does it in a correct way. The model suggests that aggregation would only be important in the deeper ocean.

We have addressed these issues in this new version of the manuscript.

Specific and minor comments

page 3, line 3-4: My suspicion is that the good correlation between lithogenic particles and Mn may be caused by Mn inside lithogenic material, not so much by scavenged Mn. I would therefore be cautious to cite that as an evidence for that 'lithogenic particles are likely to play a significant role in the removal .. on Mn'.

Lithogenic particles contains Mn, but in Roy-Barman et al. (2005) the correlation is between the authigenic Mn concentration and lithogenic tracer concentration such as ^{232}Th in sediment. In this study, the authigenic Mn content exceeds largely the lithogenic Mn content, so that the correlation is indeed between authigenic Mn and lithogenic particles.

We have rephrased this accordingly in the new version of the paper.

Equation 3 for the Middelburg et al. empirical formula for denitrification is incomplete, as the units of neither the denitrification rate nor the water depth are given. Probably it is metre for the latter, but for the first?

We have added the units. It is actually the logarithm of the denitrification flux; we've made this clear in the text.

p.4, line 17: It is on purpose that the concentration of O_2 is denoted by square brackets, but that of the OH^- ion by curly brackets?

Yes, this is how Johnson et al. (1996) have it. The $[\]$ is a concentration, $\{ \}$ activity. Since it is used only once, we now just use the words there, and not the symbols.

In my printout of the pdf file several of the equations appear mangled, with mathematical symbols replaced by small blocks, vertical lines replaced by some dots, the vertical bracket in eq. 20 missing etc. I think this is the consequence of using the a formula editor, which relies on software-specific fonts. The formulae should be checked out not only on a computer screen, but also in a printout.

Everything looks okay now (at my end) both on paper and on the screen.

Also, in my printout of Figure 2, all arrows are replaced with strange symbols, the ligated letters ff in 'sediment diffusion' are lost etc. I think this means that the pdf relies on the presence of some fonts that are present some type of computers, but not elsewhere. The ligated 'ff' in flux is also lost in the captions of the subfigures in figure 3.

If we cannot find (nor solve) the problem ourselves, we will ask the editors to have a look at this problem at the time of submission. Thank you for raising this issue.

It is very convenient that the caption to Figure 4 marks the cruise names in the same colours as the dots within the figure, but is this feature supported in the final journal form?

Good question. They state in the instructions that it is not allowed in tables, nothing about figures/captions. We'll just see what the editor says.

Several of the citations are incomplete, e.g. Charette et al.

We have checked and updated our citations (like this one) where needed.

The new manuscript version is included as the remaining pages of this document.