Biogeosciences Discuss., 6, C4149–C4181, 2010
www.biogeosciences-discuss.net/6/C4149/2010/

Biogeosciences
Discussions

Interactive comment on “Atmospheric deposition of nutrients and excess N formation in the North Atlantic” *by* L. M. Zamora et al.

L. M. Zamora et al.

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Received and published: 26 January 2010

Dear Dr. Middelburg, editor:

Below follow comments on the manuscript: “Atmospheric deposition of nutrients and excess N formation in the North Atlantic” by L. M. Zamora et al.

We thank the three referees for their thoughtful reviews, and their useful and constructive comments on our manuscript. In the first submitted version, several important parts of our methods were unclear to some of the referees. Their suggestions have helped us clarify and improve the text. We now hope that the manuscript in its revised form will be acceptable for publication in “Biogeosciences.” Below, we have given our response

in italics following the individual referee comments.

General comments

Please note that we would like to add a supplementary appendix which gives a more comprehensive description of the biogeochemical model. We feel that this is necessary since, as referee #3 put it, “it is hardly possible to follow the description and evaluate the necessary details of the different models. Here some clarification in the text is necessary.”

Referee comments and our responses

Anonymous Referee 1

Assessment

The individual sections are well written with a good summary of processes and methods. I just had a few minor suggestions and comments that I am stating below:

Comment 1: In the Introduction section where you discuss N*, DINxs, TNxs, it might be helpful to define N* as well. Also, please clarify the difference (if any) between N* and DINxs.

> *Response: To address the reviewer’s concern we have modified the paragraph in the original draft on page 9852, line 5 so that it reads:*
>
> “. . .We employ the index DINxs, which is defined as the excess of inorganic N relative to inorganic P expected from the Redfield relationship:
>
> $$\text{DINxs} = [\text{NO}_3^-] - 16 * [\text{PO}_4^{3+}] \tag{1}$$
>
> The difference between DINxs and the definition of N* (Gruber and Sarmiento, 1997) as modified by Deutsch et al. (2001) is the constant 2.9:

$$\mathrm{N}^{*} = DINxs + 2.9 \quad (2)$$

The constant 2.9 imposes a global mean N* value of zero, but the spatial gradients of the excess nitrate are the same, whether calculated as N* or DINxs. Because here we focus only on the North Atlantic and for this reason do not require the global mean N* value to be zero, we use DINxs instead of N*."

Comment 2: In section 3.3.1, in the last sentence the parenthesis needs to be closed.

Response: Thank you, we have corrected this.

Referee 2 (Natalie Mahowald)

Assessment

The ocean biogeochemistry explorations were crude, but seemed interesting and insightful, but I am an atmospheric modeler, so I may miss some of the subtleties here.

Some comments:

Comment 3: How did you model the atmospheric deposition of SRP? It doesn't appear to be in the description. Or are you just comparing your estimated TP deposition to SRP observations? You appear to be saying that your TP deposition is too low by an order of magnitude, but SRP observations and TP modeled values match ok. I'm a little confused what you did with this information. "N deposition (e.g. Markaki et al., 2003; Chen et al., 2007), our errors in SRP deposition estimation should have only minor effects on the amount of excess N in deposition" this implies you ignored your order of

magnitude error? Why bother to include P deposition if you are doing it so poorly? I'm confused. Why not just adjust your model values to match the data? But do you really want SRP instead of TP anyway? What is the relationship between SRP and TP in the atmosphere?

Response: Here, many of the referee's questions appear to be due to a lack of clarity in the text. We will answer these questions from the referee one at a time, and then we will indicate how we plan to change the text to improve the clarity:

Comment 3a: How did you model the atmospheric deposition of SRP?

Response: We will elaborate on our Methods section 2.1 p. 9854 l. 14 by changing it from:

"Observations of soluble reactive phosphorus (SRP) deposition are sparse; because there are more data for comparison, we focused on comparison with total phosphorus."

To:

"Observations of soluble reactive phosphorus (SRP) deposition are sparse; therefore, instead of modeling SRP directly, it was assumed that all phosphorus deposition was in the form of bioavailable SRP. Our motivation for making this assumption was twofold: First, there are more total phosphorus (TP) data available than there are SRP data, and so the accuracy of the modeled P deposition was easier to gauge by using TP data as a basis for comparison. Secondly, because in reality, SRP is much lower than TP (between 7-100%, averaging ~32% in the North Atlantic (Baker et al., 2006a; Baker et al., 2006b; Mahowald et al., 2008)),

> we intended this method to err by overestimating the amount of bioavailable P deposition to the ocean. Thus, any errors due to faulty P deposition would be more likely to underemphasize the role of excess N deposition rather than falsely over-emphasize it, leading to a conservative estimate of how deposition affects the relevant biogeochemical processes studied in this experiment."

And to avoid confusion and repetition, we will remove the following section from the Results section 3.1. (p. 9860, line 11):

> "Only the bioavailable fraction of deposited TP (which is predominantly SRP) will affect the surface nutrient balance. Here, in part because there are more TP deposition data than there are SRP deposition data, we attempted to obtain an overestimate of SRP deposition by modeling TP deposition and assuming all TP to be bioavailable."

Comment 3b: You appear to be saying that your TP deposition is too low by an order of magnitude, but SRP observations and TP modeled values match ok. I'm a little confused what you did with this information. "N deposition (e.g. Markaki et al., 2003; Chen et al., 2007), our errors in SRP deposition estimation should have only minor effects on the amount of excess N in deposition" this implies you ignored your order of magnitude error? Why bother to include P deposition if you are doing it so poorly? I'm confused. Why not just adjust your model values to match the data? But do you really want SRP instead of TP anyway?

Response: This confusion was probably due to in part to our poor Methods explanation of how we modeled SRP (which we have clarified in the response to comment 3a). To avoid further confusion, we will remove our discussion of the comparison between the model and observed P deposition because it is not directly relevant to our methods and findings.

Instead we will focus on the comparison between modeled P deposition and SRP observations. The revised paragraph on this topic originally from Results section 3.1 (p. 9860 starting on line 15) will now say:

> "Although the original intent of assuming that all deposited phosphorus was bioavailable was to overestimate SRP deposition (section 2.1), modeled P deposition actually slightly under-represented wet SRP deposition (dry SRP deposition was not compared due to scarcity of data). The model reproduced 67±43% of observed wet deposition SRP and only 19±35% of observed TP deposition (based on the references in auxiliary material Table S2b in Mahowald et al., 2008, as presented in Fig. 9). As the initial objective was to represent SRP values and not total deposited P, a 67% matchup between modeled P deposition and observed SRP deposition was deemed acceptable. However, an under-representation of P could result in errors in the magnitude of excess N deposition."

The referee's suggestion of adjusting the model values to match the observed TP data is a very good one, which we might have done if we had thought of it at the time. However, because we are most interested in SRP deposition and because we feel that our current 67% recovery is already acceptable, we believe fitting the data might add some additional degree of error but might only nominally help us. In the following paragraph of the Results section (p. 9860, line 24), we addressed the issue of errors in P deposition. We discuss that errors in P deposition estimates may affect the rest of our study by increasing the amount of excess N in deposition to the NASTG. However, the observed values of SRP deposition are low enough in comparison to soluble N deposition values that an error in SRP deposition of 40% (based off the relationship to observations of 67±43%) is expected to be negligible except perhaps in the SE corner of the gyre. Even in this corner of the gyre, all deposition still has N:P ratios of at least

37N:1P, which is well above the Redfield ratio of 16N:1P. Therefore, as we state in the text, although P deposition errors could slightly change the magnitudes of excess N deposition effects, excess N should still be deposited and the actual processes affected by excess N should still be visible.

However, we will incorporate an additional sentence in the conclusions stating that while the signals and processes we observe as being caused by atmospheric deposition are still relevant, there is some error associated with the magnitudes of the processes being studied due to incomplete and therefore, potentially inaccurate deposition values.

Comment 3c: What is the relationship between SRP and TP in the atmosphere?

Response: To answer the reviewer's question for the reader's benefit, we have inserted the following sentence into the Methods section 2.1:

"Secondly, because in reality, SRP is much lower than TP (between 7-100%, averaging ~32% in the North Atlantic (Mahowald et al., 2008, Baker et al. 2006a,b)), this method would err by overestimating the amount of bioavailable P deposition to the ocean."

Comment 4: "The mechanisms and time scales by which atmospheric deposition will be transported out of the surface waters are unclear, although it appears that non-Redfield processes in the surface are important in the NASTG": can't you get these numbers from your model?

Response: The sentence Dr. Mahowald refers to is in our Conclusions section (p. 9869, line 10). While we do model the mechanisms and time

scales of the effects of atmospheric deposition in the ocean, what we had intended to convey with this sentence is that because our model study scenarios represent ideal cases, they are not necessarily representative of what goes on in the real ocean. Therefore, we will reword this sentence to say:

"The actual mechanisms and time scales by which atmospheric deposition is transported out of the surface waters cannot be obtained from this series of idealized biogeochemical scenarios. However, our model results do allow insight about which scenarios are most likely to be occurring. Because modeled purely Redfield conditions produced unrealistic results, non-Redfield processes in the surface appear to be important in the NASTG."

Comment 5: Could you clarify your conclusions a bit: How much of your results are new to this paper? Be very clear in the conclusions what is new, what agrees with previous studies and what disagrees. What are the errors in your study because you are effectively kludging nitrogen fixation, and remineralization, and denitrification, instead of including them in the model explicitly? Please be explicit in your conclusions about what is believable about your study and why.

Response: This is an excellent suggestion. We plan to re-write the conclusions as follows (with changes in the text highlighted in bold):

Conclusions

"**Atmospheric nutrient deposition has long been thought to contribute to the high DINxs development in the North Atlantic (Fanning, 1992; Michaels et al., 1996; Hansell et al., 2007), thereby affecting the geochemical estimates of N_2 fixation in the gyre. Previous works have estimated the various sources of DINxs in the main thermocline of the NASTG, including the contribution from**

N_2 fixation (Gruber and Sarmiento, 1997; Hansell et al., 2004; Hansell et al., 2007; Landolfi et al., 2008), atmospheric deposition (Hansell et al., 2007; Landolfi et al., 2008), and preferential P remineralization (Landolfi et al., 2008). This work provides the first mechanistic understanding of how deposition might contribute to the development of DINxs. We determine that atmospheric nutrient deposition could account for up to 19% of current annual DINxs production rates in the main thermocline and that atmospherically deposited nutrients of natural and anthropogenic origin have probably been affecting DINxs levels in the main thermocline of the NASTG for some time.

Because the inputs of nutrient deposition are not well understood, there is considerable uncertainty in how much atmospheric nutrient deposition contributes to high DINxs development in the main thermocline. Due to a paucity of data, we did not include bio-available organic nitrogen and phosphorus deposition in this study. Excluding organic nutrients could potentially lead to a significant underestimation of the effects of nutrient deposition on the ocean. Conversely, the comparison between the modeled phosphorus deposition and the measurements made at various marine-influenced stations tended to underestimate P deposition by ~40%, which may cause an overestimate of the effects of nitrogen deposition. Despite these errors, N:P ratios in deposition were consistently higher than Redfield N:P ratios (as expected based on numerous observations). Therefore, even though the magnitude of atmospheric nutrient deposition is somewhat uncertain, we were still able to simulate the direction of biogeochemical effects caused by high N:P deposition to the ocean.

Non-Redfield processes in the surface appear to be important in the NASTG because under purely Redfield conditions, biological processes produced unrealistic results. **Previously, both preferential P remineralization and/or export of particles with high N:P ratios have been suggested as possible mechanisms for exporting excess N from the surface to the main thermocline. Our study, which represents a first attempt to model the interactions of these non-Redfield processes with atmospheric nutrient deposition,** reveals that either

(or both) mechanisms could be occurring, but that preferential P remineralization requires N_2 fixation or higher levels of deposition to co-occur in order to produce realistic nutrient distributions.

Based on all three modeled scenarios, we also observe that N deposition should be causing P depletion in the surface. **This result is supported by the findings of others that indicate that atmospheric deposition is also increasing P limitation in the Mediterranean (Herut et al., 1999), the global ocean (Krishnamurthy et al., 2009), and in lakes (Elser et al., 2009).**

Due to the regional rather than global focus of this study, we were able to resolve some of the finer details that affect deposition fate in the NASTG. Deposition location, circulation of NASTG waters, and the rate of local export were very important to the fate of deposition in this region. Rapid ventilation of the gyre enabled atmospherically influenced surface water to be transported to the main thermocline, particularly affecting the northeastern part of the gyre in all scenarios. Because most atmospherically deposed nutrients landed near the Gulf Stream, a rapid export meant that when particles remineralized in deeper waters, they did not re-enter the gyre. In contrast, ADINxs transported in surface waters could re-enter. For this reason and due to the importance of surface water subduction in the NASTG, future studies would benefit by a more comprehensive inclusion of DOM. Finally, from the excess N uptake scenario it appears that convective upwelling may enhance the effects of deposition by resupplying atmospheric nutrients to the surface waters from below.

Some sources of error in this study are the unquantified interactions of atmospheric deposition with DOM and N_2 fixation/denitrification. Although marine DOM is indirectly simulated in the Redfield scenario, we cannot separate the specific contributions of DOM and inorganic nutrients to ADINxs development, and so any biogeochemical effects due to delayed DOM remineralization in subducting water are unaccounted for. In addition, we did not include interactions between nutrient deposition and N_2 fixation. For example, atmospheric deposition may stimulate N_2 fixation by providing Fe (e.g.

Moore et al. (2009)). Alternatively, N from deposition could enable other organisms to outcompete diazotrophs. We currently do not understand these opposing interactions very well, and cannot include them in our model at this time; therefore, our results must be interpreted with some caution. Despite the uncertainty in the magnitude of the ADINxs signal, however, this study is the first to provide useful information on the types of effects that atmospheric nutrient deposition has on the North Atlantic subtropical gyre and on the most likely physical and biological mechanisms that affect the fate of deposited nutrients in this region.

Our results **support an increasing body of geochemical research that indicates that** atmospheric nutrient deposition is important enough to be included in future mass balance assessments of excess N in the NASTG. Based on our results, atmospheric N deposition will probably increase new production and affect surface as well as deep water concentrations of nutrients. Our results reinforce the idea that it is inappropriate to assume Redfield stoichiometry in this area."

Anonymous Referee 3

Assessment

These approaches are generally meaningful and plausible to answer the question. There are, however, several major problems with the modelling, which may partly come from an incomplete description. For me as a non-modeller it is hardly possible to follow the description and evaluate the necessary details of the different models. Here some clarification in the text is necessary.

Comment 6: The authors use the coarse grid model (70 years) and not the finer resolution (42 years) because they want to save computation time. This statement is not quite satisfying because the difference may have important implications for the model results – if that is not the case it needs to be better demonstrated.

Response: We used a state-of-the-art ocean circulation model with admittedly rather coarse resolution. We agree also that our intercomparison with the higher resolution model could have been more extensive. On the other hand, even our "high resolution" (1 degree) model is, probably, a rather rough representation of the real ocean as essential processes such as eddy dynamics are not explicitly resolved and the parameterization applied to account for such effects is still subject of ongoing research which is beyond the focus of this paper. We feel also that an extensive analysis focused on the sensitivity of ocean circulation on horizontal resolution is beyond the focus of this paper and hope that the reviewer sees, nevertheless, merit in our research. We do present some results, however, which indicate that a change to the 1 degree resolution will probably not revise our conclusions.

Changing from coarse to 1 degree resolution at year 42 resulted in a change in atmospheric nutrient accumulation from 0.80 to 0.78 mmol N m^{-3} *at year 42. This results in only a 3% difference between the two models. Because the nutrient deposition accumulation signal includes both the relevant physical and biological dynamics, we do not believe that much would be gained by running the model at the 1 degree resolution. To emphasize why we think that the fine resolution model was acceptable, we will change the text from:*

"...at the end of the fine resolution model run, nutrient deposition caused an accumulation of 0.80 mmol NO_3^- m^{-3}, whereas in the fine resolution model there was an accumulation of 0.78 mmol NO_3^- m^{-3} at year 42. Because the nutrient deposition accumulation signal includes both the relevant physical and biological dynamics, we determined that the coarse mode yielded similar results to the fine model, and so for computational reasons, the coarse resolution model was used for the sensitivity studies described in the remainder of the study."

To this (changes are in bold):

"**The nutrient deposition accumulation signal includes both the physical and biological dynamics relevant to this study.** At the end of the fine resolution model run, nutrient deposition caused an accumulation of 0.80 mmol NO_3^- m^{-3}, whereas in the fine resolution model there was an accumulation of 0.78 mmol NO_3^- m^{-3} at year 42. **The 3% difference between the fine and coarse models was small enough that we determined that running the model at a finer resolution was acceptable.** For computational reasons, the coarse resolution model was used for the sensitivity studies described in the remainder of the study."

Comment 7: It seems that a globally homogenous forcing was used even for the atmospheric deposition which is certainly heterogeneous. It is written on p 9855 l 20-24 that it is for conceptual simplicity. I have the feeling that it is over-simplification. The homogeneity suppresses horizontal gradients which are a typical feature for atmospheric deposition and necessary for the specific questions of the manuscript.

Response: We believe there was a misunderstanding here- deposition applied to the ocean was not horizontally homogeneous. As represented in Figures 4a and 4b, nutrient deposition was spatially heterogeneous.

The referee refers to the following line,
"For conceptual simplicity, we used year 2000 deposition estimates for each year of the model simulation. Although the deposition was applied over the entire global ocean, we can neglect the impact of atmospheric deposition in other ocean basins on the North Atlantic Ocean due to the decadal time scales considered

here."

To avoid this confusion in the future, we will change this passage to say that:

"Year 2000 deposition was applied in amounts corresponding to Figs 4a and 4b each year that the model ran. Although deposition was applied over the entire global ocean, we can neglect the impact of atmospheric deposition in other ocean basins because it would take longer than the 70 years of the model run for deposition in other basins to affect the biogeochemistry of the North Atlantic."

Comment 8: Why is oxygen a modelled variable? It does not appear anywhere and seems not to be necessary.

Response: The referee is correct that the presence of oxygen as a variable should not affect any of our results. We will remove it from the list of prognostic variables.

Comment 9: No lateral advection is produced in the water column with this approach if I understood this correctly. But advection is essential for the whole modelling exercise and the particle distribution and degradation (see page 9857 l 26). Furthermore lateral advection results are later discussed for the modelled ocean box.

Response: We believe that here there was also a misunderstanding. A full suite of circulation was present in the model, including lateral advection. Lateral advection at 5 and 500 m is pictured in Figures 8a and 8b.

The source of this misunderstanding was a poorly-worded portion of our *methods section in which we stated,*

> "Our assumptions allow no time delay for atmospheric nutrient export and ignore the impacts of lateral transport on the sinking particles."

In context, this sentence is part of a paragraph whose purpose was to defend the biogeochemical conditions and assumptions that defined the excess N scenario. In this scenario, our assumptions were twofold: 1) that deposited nutrients were remineralized immediately, and 2) at any given location, the distribution of these remineralized deposited nutrients in the water column was proportional to the existing remineralization profile at that location. In the sentence that confused our referee, we meant to point out a potential fault with the second assumption. Particles generated by deposition can be moved laterally with time in the real world, and so deposition that falls on the surface at one point may not influence the remineralization profile at, say 400 m, until the particle is moved further downstream from where the deposition fell.

To avoid similar confusion in the future, we have re-worded the problematic sentence to say:

> "The biogeochemical assumptions specific to the excess N scenario allow no time delay for atmospheric nutrient export and do not take into account that particles generated by deposition can be moved laterally with time in the real ocean."

Comment 10: The authors assume uptake of all atmospheric excess N (page 9857 l 18). How does this work under a Redfield scenario – all excess N becomes DON?

Response: To clarify, we only assume organisms can take up all excess N in one of our biogeochemical scenarios: the excess N uptake scenario

(not in the Redfield or preferential P remineralization scenarios).

We are not sure what the referee meant when they asked how the uptake of excess N would work in a Redfield scenario. If the referee is asking about what happens to excess N from deposition when Redfield conditions hold true, in short, excess N from deposition is not taken up at all by organisms unless it lands in or is transported to a location where there is excess P in the surface water (more detail can be found in section 3.3.1). This is because in the Redfield scenario, organisms can only take up N in ratios of 1N:16P (Redfield ratios). Thus, if Redfield conditions were strictly adhered to in the real-world subtropical N. Atlantic, there should be an increasing amount of N in the surface, which perhaps could be stored as DON.

Hopefully this answers the intended question?

Comment 11: Under chapter 2.4 the problems of DOM exclusion are named. DOM is a major sink for atm. deposition and it is a much larger fraction than DIN or particles. How can such a major pool be neglected is still not clear to me. (page 9858 l 10-13)

Response: The referee raises a good point and we agree that including DOM would have been ideal. While the majority of N and P in the subsurface ocean is in inorganic form, the majority of N and P in the surface gyre is in organic form. Also, the surface DOM pool is a reservoir for excess N that will contribute to high DINxs in the main thermocline upon remineralization. Unfortunately, there are not sufficient DOM data to provide good enough information to map DOM distributions in this region.

However, despite our lack of a DOM compartment we maintain that even without explicitly including DOM dynamics, our results are still valid within *the context that we provided. In the Methods text, we state that:*

"...excluding DOM was not a major problem because DOM does not sink, and any non Redfield DOM formed from atmospheric nutrient deposition should exhibit a similar pattern as ADINxs in the Redfield (physical transport) scenario."

We think that this argument is important and valid. Therefore, we will flesh out a more complete description of how DOM behavior is approximated in this model in the Methods section 2.4:

"As our focus is purely on the biogeochemical effects of atmospheric deposition (see section 2.4), DOM is primarily relevant to this study is as a reservoir for high ADINxs. A non-Redfield DOM reservoir can be formed through three processes: atmospheric deposition, N_2 fixation, and preferential P remineralization (Landolfi et al., 2008). Unfortunately, there are many uncertainties regarding DOM remineralization rates, distributions, and the relative importance of each of these processes on non-Redfield DOM formation. Therefore, a DOM compartment was not explicitly modeled in this study.

However, the indirect effects of DOM were included in ADINxs estimates. It is realistic to assume that within the fraction of DOM which is labile (i.e. which is available to surface biology on timescales shorter than a year), the DOP fraction would cycle faster than the DON fraction (Vidal et al., 1999; Abell et al., 2000; Aminot and Kérouel, 2004). Therefore, the effects of non-Redfield DOM on ADINxs development would be included in the preferential P remineralization scenario. Alternatively, if DOM is refractory (i.e. not taken up by biology at the surface until timescales of greater than a year), then the effect of DOM on DINxs would be mimicked by the Redfield scenario. Based on transient tracers, water advects across the surface NASTG within 7 to 10 years before subduction. DOM

and ADINxs in the Redfield scenario should behave similarly. Upon subduction, transects of DON in the North Atlantic indicate that the majority of subducted DON remineralizes within 15 years (Hansell et al., 2007). Therefore, within the >70 year timescale of our model study, remineralized DOM should create inorganic nutrient patterns similar to those in the Redfield scenario. Although the preferential P remineralization and Redfield scenarios do not provide any indication of which fraction (organic or inorganic) ultimately contributes the most to the ADINxs signal, they do indicate both the location and the overall contribution of the original atmospheric deposition source to ADINxs."

We will also emphasis more strongly in the conclusions the uncertainties caused by not having a DOM pool by adding in the following statement (also see response to comment #5).

"Other sources of error in this study are the unquantified interactions of atmospheric deposition with DOM and N_2 fixation/denitrification. Although the contribution of marine DOM to ADINxs is indirectly simulated in the Redfield and preferential P remineralization scenarios, we cannot separate the specific contributions of DOM and inorganic nutrients to ADINxs development...."

Comment 12: What is the mass balance technique page 9858 line 21?

Response: The context of line to which the referee refers is reproduced here:

"We could not realistically model N_2 fixation in the North Atlantic by simply assuming that diazotrophs have an advantage in surface waters with low excess N, i.e. a surplus of P over N (Deutsch et al., 2007; Schmittner et al., 2008).

Its exclusion meant that we could not reproduce the observed DINxs pool nor maintain a realistic steady state of DINxs in the NASTG. Models such as that of Coles and Hood (2007) have dealt with the difficulty in maintaining realistic diazotroph levels by assuming that N_2 fixation equals the rate of development of the excess N signal. Since the goal here was to isolate the atmospheric contribution to the DINxs signal, this method was not an option. **(Line 21)** We therefore did not include N_2 fixation in any of the model simulations and instead, we compared model runs with and without deposition, making the assumption that the various sources of DINxs are sufficiently independent to be superimposed linearly (see Fig. 7)."

I think that perhaps we were not clear enough in this passage, and so we might have confused the referee into thinking that a mass balance technique was used. In fact, we intended to put across that we did NOT use a mass balance technique as others have done because a mass balance technique would not have allowed us to differentiate between mechanisms of DINxs formation (in this case, atmospheric deposition and N_2 fixation).

To avoid this confusion in the future we will re-write the above paragraph (with changes marked in bold) to say:

"We could not realistically model N_2 fixation in the North Atlantic by simply assuming that diazotrophs have an advantage in surface waters with low excess N, i.e. a surplus of P over N (Deutsch et al., 2007; Schmittner et al., 2008). **The** exclusion of **N_2 fixation** meant that we could not reproduce the observed DINxs pool nor maintain a realistic steady state of DINxs in the NASTG. Models such as that of Coles and Hood (2007) have dealt with the difficulty in maintaining realistic diazotroph levels by assuming that N_2 fixation equals the rate of development of the excess N signal. Since the goal here was to isolate the

atmospheric contribution to the DINxs signal, this **mass balance** method was not an option. We therefore did not include N_2 fixation in any of the model simulations."

Additionally, the referee asked us to better explain this aspect of our methods. An understanding of this section is key to our paper and to interpreting our results. Therefore, we will also clarify the subsequent paragraph (changes are in bold).

"Instead, we compared model replicates that did and did not include deposition. The difference between the replicates with and without deposition was taken as the isolated effect of deposition for that scenario. **All other complexities within the system, such as those stemming from the lack of N_2 fixation and denitrification, were the same for each replicate. Therefore, the only differences between the two replicates were the exclusive effects of atmospheric deposition. The major assumption of this method is** that the various sources of DINxs are sufficiently independent to be superimposed linearly (see Fig. 7) **(e.g. N_2 fixation does not affect ocean response to atmospheric deposition and vice versa).**"

Comment 13: Export seems to be a problem in the model too (besides DOM) because the flux is not well constrained (see also page 9867 line 13-14) causing variations in the estimates by more than an order of magnitude. How critical is that for the overall model results?

Response: Export fluxes are not well known for the North Atlantic, and we address this issue further in the text (see below). First though, we want to clarify that the export problem does not specifically lie with our model. The sentence that the referee refers to begins as:

“Unfortunately, export flux of PON from the surface of the NASTG to below the euphotic zone is not well constrained. Depending on the method used and the site and time sampled, N export flux in the NASTG ranges from <0.02 to 0.63±0.15 mol N m^{-2} yr^{-1} (Jenkins, 1982; Jenkins and Goldman, 1985; Jenkins, 1988; Oschlies, 2002; Roussenov et al., 2006). If high and low-end estimates of export and DINxs annual production are used, anywhere between 5 and nearly 100% of original P in exported particles would need to remain in the surface. In fact, the low end estimates are too small to account for the DINxs itself. Given this wide range, we instead primarily focus on export flux and remineralization rates as relevant within our model.”

To avoid confusion, in this passage we meant the reader to understand that field data are highly variable and that they support a wide range of export flux estimates; variability in the field data is what causes our understanding of export fluxes to be poorly constrained. While our model cannot be said to be accurate, because there is no actual consensus on what actual export fluxes are in the field, it is not necessarily any faultier than any other model; in fact, we use state-of-the-art export flux models, and so it is probably as good as current models can get. An exhaustive study of export fluxes is beyond the scope of this paper. However, we see no reason to believe that it is inherently bad- it lies between the high and lower end flux estimates, it is based on what we consider to be realistic assumptions (see Kriest and Oschlies, 2008), and export in the NASTG is high enough to theoretically support the observed DINxs accumulation (p. 9867, line 19) in the preferential P remineralization scenario.

Furthermore, errors in export flux would have only minor impacts on the results of our study. In the excess N uptake scenario, export flux is irrelevant since we are assuming 100% flux of atmospherically deposited nutrients to

below the surface anyways to obtain an upper estimate of how this process could affect the NASTG. In the Redfield and preferential P remineralization scenarios, an unrealistic export flux would not change the direction of the ocean response to nutrient deposition, but it could affect the magnitude of the response. There are two ways in which export flux could contain error. The first is if N:P ratios in sinking matter are wrongly assigned. Understanding the degree to which N:P ratios matter in sinking matter to ocean biogeochemistry is one of the primary goals of this study, and so errors in N:P ratios will not be discussed specifically since this was tested.

Therefore, particle sinking flux is the main source of error in export fluxes in these two scenarios. In the Redfield scenario, the surface quickly goes to P limitation, and so sinking flux is very low and most export of nutrients to the main thermocline is via subduction. Therefore, errors in sinking flux for the Redfield scenario are negligible. However, in the preferential P remineralization scenario, an unrealistic sinking flux could affect the magnitude of our results because the amount of P retained in the surface would change along with the amount of excess N that is exported to the main thermocline. We address this in detail on page 9867, starting on line 10.

Comment 14: The authors explain why they did not consider nitrogen fixation (page 9858 l 13-20). But it is well known how important nitrogen fixation is in the tropical Atlantic Ocean, especially under light of all the unicells and proteobacteria nitrogen fixation activities. To omit this major source of excess nitrogen is problematic.

Response: The referee states that omitting N_2 fixation is problematic because N_2 fixation is a major source of excess N. We believe that this is not entirely correct. While N_2 fixation is a major source of excess N, this fact is not very relevant to the focus of our study. The scope of our study *was only the fate of atmospheric deposition and it's unique contribution to DINxs. Given this focus, the only way N_2 fixation (or lack thereof) would affect our results is if there are interactions between atmospheric deposition and N_2 fixation. Because this misunderstanding arose, it is clear to use that we need to go back and re-emphasize our goals in the introduction of the paper.*

First though, let us emphasize that it was necessary to separate the atmospheric deposition signal from the N_2 fixation signal to address the scope of the paper, which is why N_2 fixation was not included. Because both this referee and referee # 2 ask how omitting N_2 fixation affects our results, we will add further detail in both the Methods section 2.4 (p. 9859, line 4) as follows:

"N_2 fixation is known to be important to the North Atlantic biogeochemistry. The potential of this important process to affect our study results depends on whether deposition is sufficiently independent from the process of N_2 fixation to be superimposed linearly as we do in our model. Here, we assume that the biogeochemical effects of N_2 fixation do not have any interactions with the biogeochemical impacts of atmospheric deposition. If this is the case, then omitting the N_2 fixation source of excess N has no bearing on the formation of high DINxs from atmospheric deposition and thus, can be ignored. However, if atmospheric deposition does affect N_2 fixation in a major way, or if N_2 fixation affects the ocean's response to atmospheric deposition, then our model would have an unquantified source of error. One way that atmospheric deposition could affect N_2 fixation could be by providing Fe (e.g. Moore et al. (2009)). Alternatively, N from deposition could enable other organisms to outcompete diazotrophs. We currently do not understand these opposing interactions very well, and cannot include them in our model at this time. Therefore, our results must be interpreted

with some caution. However, the main goal of this study is to understand the maximum potential role of atmospheric deposition in DINxs development. While uncertain feedbacks between N_2 fixation and atmospheric deposition probably affect the role of ADINxs in the main thermocline, the likely mechanisms of ADINxs transport remain the same."

And in the conclusions (also see response to comment #5):

"Other sources of error in this study are the unquantified interactions of atmospheric deposition with DOM and N_2 fixation/denitrification. . . .we did not include interactions between nutrient deposition and N_2 fixation. Some interactions could include the stimulation of N_2 fixation by atmospheric iron deposition or the suppression of N_2 fixation by N enhancements from deposition. We currently do not understand these opposing interactions very well, and cannot include them in our model at this time; therefore, our results must be interpreted with some caution. Despite the uncertainty in the magnitude of the ADINxs signal, however, this study is the first to provide useful information on the types of effects that atmospheric nutrient deposition has on the North Atlantic subtropical gyre and on the most likely physical and biological mechanisms that affect the fate of deposited nutrients in this region."

Comment 15: And nitrogen fixers – at least the colonial species like *Trichodesmium* - are known to release a major fraction of the fixed N as dissolved organic N which is automatically also not considered.

Response: The referee is correct that diazotrophs affect DOM dynamics and thus can indirectly affect DINxs. However, the same argument applies here as in the response to comment # 14: in this paper we are purely focused on the factors influencing ADINxs (the atmospherically-derived por-

tion of DINxs). The only reason diazotrophic activity would affect ADINxs, through DOM release or otherwise, is if diazotrophy affects the ocean response to atmospheric deposition or vice versa. As stated in the response to comment #14, these potential interaction effects could include inhibition of N$_2$ fixation due to N deposition or an enhancement of N$_2$ fixation due to iron inputs associated with deposition. Unfortunately, not much is known accurately known about these interaction effects in the actual environment and so in our model we make the assumption that these feedbacks are not important. The referee is correct that the degree of error associated with this assumption could be significant. To address this concern, we have added a paragraph in the conclusions stating that this error exists and may significantly affect the magnitudes of the processes considered, although not the direction of the biogeochemical effects (see also response to comment #5).

The other effects of N$_2$ fixation and its release of DOM are beyond the scope of this paper. Please also see our previous response pertaining to DOM (comment #11) for more about the effects of DOM in our study.

Comment 16: Moreover it seems problematic to use the WOCE data set for comparison in which nitrogen from fixation is included.

Response: We can see how the referee might have been confused, but we do not directly compare our model's nutrient data with the WOCE dataset.

The first case where there might be some confusion is with Fig. 10, where we relate the locations of modeled DINxs production with the locations of modeled ADINxs accumulation. As a reminder, ADINxs is the fraction of

DINxs from atmospheric deposition. ADINxs does not include the effects of N$_2$ fixation, but DINxs does.

What might have confused the reviewer is the way in which we included N$_2$ fixation into the DINxs production rates. As described in section 2.5, for Fig. 10, we began a model simulation with the WOCE data. The WOCE DINxs data represent all real world sources of excess N, including DOM remineralization, atmospheric deposition, and N$_2$ fixation. To determine the rate of loss of DINxs from the system, the WOCE nutrient pool was then subjected to modeled ocean circulation in the absence of any source of DINxs. By assuming steady state conditions, we equated the rate of DINxs loss to the rate of DINxs production. Because ADINxs is a fraction of DINxs, the rates of production/ accumulation of the two can be validly compared.

The second case where there might be confusion is with Fig. 11, where we show profiles of DINxs, PO$_4^{3+}$, and NO$_3^-$ in the real ocean (using WOCE data) next to profiles that indicate the amount of modeled change in DINxs, PO$_4^{3+}$ and NO$_3^-$ caused by deposition in the three scenarios. The purpose of showing WOCE data in this figure is only to give perspective to the size of changes from atmospheric deposition shown in 11b,c, and d. To make this clearer to future readers, we will change the figure caption to state this.

Comment 17: Some other comments: It would be nice to give a short summary of the hydrography/ currents of the modelled area.

Response: This is a good idea. We propose to insert and reference the following hydrographic description of our study region in section 3.2.2, which is where we discuss the modeled spatial distributions of ADINxs:

> "The NASTG is bounded to the west and northwest by the Gulf Stream. In the northwestern part of the gyre (north of ~19°N and west of 45°W), spring subduction of subtropical mode water (18° water) ventilates the main thermocline to mean depths of about 287 m with a mean thickness of about 200 m (Peng et al. (2006) and refs therein). The NASTG is bounded to the north by the eastward propagating North Atlantic current (between ~30-40° N). Another subduction zone (Madeira mode water) is found northeast of the NASTG between Madeira and the Azores front, subducting south/southwestward along the isopycnal surface σ_θ = 26.5 (Siedler et al., 1987). At 35.6°N, dense Mediterranean outflow water enters the North Atlantic and flows south and west between 600-1500m (Baringer and Price, 1997). Along the south, the NASTG is ventilated by cross equatorial flow. Within the gyre (centered around 20°N, 35°W), hot salty water subducts inside of the upper thermocline (~upper 200 m) to form subtropical underwater (O'Connor et al., 2005)."

Comment 18: P 9853 l 26: I guess it is not possible to compare production rates because the production by nitrogen fixers is not considered.

Response: The contribution of N_2 fixation actually is considered in the estimation of total DINxs production (also see response to comment 16). The referee refers to a statement in the introduction where we stated that,

> "By observing the biogeochemical signatures of the three mechanisms in the water column, we are able to... compare the production rates of DINxs and ADINxs (where ADINxs is the fraction of the DINxs pool in the main thermocline coming from atmospheric deposition)."

As a reminder, we estimated DINxs production by assuming that losses of DINxs equal production of DINxs (i.e. steady-state). We modeled the rate

of loss of DINxs when N_2 fixation, DOM and atmospheric deposition did not occur, starting from the WOCE dataset (which incorporates all sources of DINxs, including N_2 fixation).

N_2 fixation was not considered in ADINxs because this fraction is specifically defined as the fraction of DINxs from deposition. It is valid to compare ADINxs with DINxs because ADINxs is just a portion of DINxs.

Comment 19: P 9854 l20: a distinction between a coarse and fine fraction is made here for P. But later the two fractions are not used. Coarse and fine is used in two different ways – for the model and for P-particles. It may be better (clearer) to use different wordings.

Response: The text the referee refers to is as follows:

> "TP was estimated from BC deposition by assuming TP is equally divided between coarse and fine fractions and by using published BC:TP molar ratios of 0.02 (for coarse particles) and 0.0029 (for fine particles) (Mahowald et al., 2005)."

To add clarity, we will change this text to say:

> "We assume that TP in aerosol particles is equally divided between coarse and fine size fractions. The two size fractions have different compositions; taking this into account, we use published BC:TP molar ratios of 0.02 for coarse particles and 0.0029 for fine particles (Mahowald et al., 2005) in order to obtain the final value of TP in the deposition."

Comment 20: Page 9866 l 19-25 where is the estimate of N_2 fixation to sustain the observed nutrient pattern in steady state?

> *Response: The referee refers to the following passage:*
>
> "For previously mentioned reasons, we did not include N_2 fixation in our model. Nonetheless, we can estimate the amount of N_2 fixation necessary in this scenario to sustain observed nutrient patterns in steady state: to balance the excess P at the surface, excess N from N_2 fixation should be equal to 16 times that of excess P. Or, in other words, if preferential P remineralization is the sole mechanism responsible for the high DINxs pool, excess N supplied at the surface from N_2 fixation should have the same magnitude as the amount of DINxs in the main thermocline."
>
> *We will add in the following:*
>
> "In this idealized case, the portion of DINxs that is due to N_2 fixation is 2.3x10^{11} mol N yr^{-1} for the region defined as the NASTG (this value is of course subject to some error because it assumes preferential P remineralization rates are correct and that the DINxs pool is primarily formed due to preferential remineralization)."

Comment 21: I like the conclusions which are very clear and interesting to read. Unfortunately all the questions in the model approach and the numerous assumptions make me question how reliable these conclusions really are. The manuscript would certainly benefit from a concise and clear description of the approach.

> *Response: The majority of concerns this referee had with the approach and assumptions in this study were based on misunderstandings within the methods text. We have clarified the text and why we believe our approach and assumptions are valid. We have also considerably edited the conclusions section to include an assessment of what is believable in the study and why, as also suggested by the second referee (see comment #5 and response). Hopefully, those edits will also address this referee's concern.*

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