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BGD

9, C5302–C5317, 2012

Interactive Comment

Interactive comment on "Nitrous oxide dynamics in low oxygen regions of the Pacific: insights from the MEMENTO database" by L. M. Zamora et al.

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GENERAL COMMENTS

Zamora et al. present analysis using the MEMENTO oceanic database to characterize marine N_2O dynamics with a focus on the low oxygen zones of the Eastern Tropical Pacific (ETP). The study includes estimation of net N_2O production and consumption rates in the ETP based on the MEMENTO data, and accompanying sensitivity analyses from an ocean model (UVic-ESCM). The study presents some useful analysis of net production and consumption rates within the ETP, however, the primary aims, layout, and description of the analysis are sometimes not very clear. Below I include some general comments on these issues.





1) AIMS: Is the main aim (a) to quantify net N_2O production/consumption rates in the ETP based on the oceanic data? or (b) to provide a set of guidelines for ocean N_2O models in the low oxygen zones of the ETP?

Thanks for pointing out that this was confusing. In answer to the question, our main aims lean more towards (b) listed above, although as part of providing good guidelines, we also tried to constrain N_2O consumption rates. To clarify the aims we have changed the last paragraph of the introduction to read (with changes in bold):

"The MEMENTO database, which is the largest and most recent collection of 100+ cruises in which N₂O data were sampled (Bange et al., 2009), provides the unique opportunity to re-evaluate net marine in situ N₂O production and consumption rates, thus complementing laboratory studies. Here we quantitatively investigate the dynamics of marine N₂O in the eastern tropical Pacific, one of the ocean's largest suboxic zones. **Our goal is to provide a set of guidelines for ocean N₂O models of the ETP.** We estimate subsurface N₂O production rates at low O₂ concentrations in order to determine whether rates increase exponentially as previously predicted. We also examine in situ N₂O consumption rates in the ETP and estimate the O₂ concentration at which net N₂O production switches to net N₂O consumption. Finally, we test the sensitivity of N₂O distributions simulated by a global ocean model to uncertainties in these values."

If the aim is (b), then there should be significantly more focus, and detail presented on the *individual* processes that contribute to N_2O formation and destruction in low oxygen regions.

To address this point, we have added in discussion of how individual microbial processes might affect the interpretation of our results. Much of this new discussion has resulted from the suggestions of the referees. In the N₂O production rate section, we now discuss 1) the impact of non-bacterial nitrifiers on N₂O production rates at low

BGD

9, C5302–C5317, 2012

Interactive Comment

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



 O_2 , and 2) the impact of using N_2O_{xs} /AOU ratios as a proxy for N_2O produced during organic matter remineralization when ideally production would be described more directly by N_2O_{xs} / NH_4^+ yields. We have added to our section on N_2O consumption rates a discussion of uncertainties in instantaneous N_2O consumption rates caused by individual N_2O production processes that might occur at O_2 concentrations <10 μ M, such as denitrifier nitrification. Finally, we have discussed several previously unmentioned uncertainties in the switching point section. These uncertainties include: 1) how the O_2 threshold may vary in different oceanic environments that include different individual processes, and 2) how using NO_2 accumulation from denitrification as a proxy for net N_2O consumption might affect the estimation of the correct threshold value. We hope that taken together, these changes will help address the referee's concern.

In addition, we have added several paragraphs to the introduction describing these relevant microbial processes. Changes to the introductory text are listed below (with new text in bold):

"The processes that contribute to N₂O concentrations in the ocean are complex and O₂ -dependent. In aerobic environments, N₂O is primarily formed through nitrification (both via the ammonia oxidation and nitrite oxidation/nitrifier-denitrification steps) (Bange, 2008). In suboxic conditions, N₂O can also be formed early in the denitrification process during nitrite reduction (NO₃ \rightarrow NO₂ \rightarrow NO \rightarrow N₂O) (Ward, 2008). However, during the last step of denitrification (N₂O \rightarrow N₂O), N₂O is consumed. Whether net N₂O is produced or consumed during denitrification depends on several factors. Oxygen concentrations play a large role, as denitrifiers generally produce net N₂O at the high end of their O₂ range (Knowles et al., 1981; Payne et al., 1971) and consume N₂O in stably suboxic to anoxic environments. At complete anoxia, i.e., O₂ =0 μ M, denitrification does not occur at all (Bange et al., 2010). Additionally, the composition of the microbial community present plays a major role, particularly because more denitrifying microbes have the enzymatic ap-

BGD

9, C5302–C5317, 2012

Interactive Comment

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



paratus to reduce $NO_3 \rightarrow NO_2$ than $N_2O \rightarrow N_2$ (Zumft, 1997).

Currently, the O_2 concentrations that determine which of these various processes are dominant are not clearly defined. Although nitrification is generally dominant in aerobic conditions and denitrification is generally dominant in suboxic conditions, studies indicate that these processes can co-occur within a large O_2 concentration range. Ammonia-oxidizers can flourish at low O_2 concentrations (Carlucci and McNally, 1969; Goreau et al., 1980), and aerobic ammonia oxidation can even be active at O₂ concentrations around 2 μ M (Kalvelage et al., 2011). Conversely, denitrification has been reported within the anaerobic interiors of marine snow in waters with O₂ concentrations up to 20 μ M (Alldredge and Cohen, 1987; Yoshida et al., 1989). In the Peruvian OMZ, recent results indicate that anaerobic NO₃ reduction might be partially active at O_2 levels even higher than this (Kalvelage et al., 2011). As previously mentioned, laboratory studies indicate that N_2O production by ammonia-oxidizing bacteria and heterotrophic denitrifying bacteria increases non-linearly at low O₂ (Goreau et al., 1980; Knowles et al., 1981; Payne et al., 1971).

As the many processes contributing to N_2O concentrations are highly complex, it is problematic that much of the data available on N_2O production and consumption rates are based on simplified laboratory studies, which only assess specific organisms and conditions that are not necessarily representative of the open ocean. *Archaea*, for example, were only recently recognized as major contributors to nitrification (e.g., Beman et al., 2012) and marine N_2O production (Santoro et al., 2011; Löscher et al., 2012). Information on their N_2O production rates is still very limited. N_2O production rates from anammox (Kartal et al., 2007) and nitrifier denitrification (e.g. Charpentier et al. (2007)) are also not well constrained.

The MEMENTO database, which is the largest and most recent collection of 100+ cruises in which N_2O data were sampled (Bange et al., 2009), provides the unique

9, C5302–C5317, 2012

Interactive Comment

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



opportunity to re-evaluate net marine in situ N₂O production and consumption rates, thus complementing laboratory studies. Here we quantitatively investigate the dynamics of marine N₂O in the eastern tropical Pacific, one of the ocean's largest suboxic zones. Our goal is to provide a set of guidelines for ocean N₂O models of the ETP. We estimate subsurface N₂O production rates at low O₂ concentrations in order to determine whether rates increase exponentially as previously predicted. We also examine in situ N₂O consumption rates in the ETP and estimate the O₂ concentration at which net N₂O production switches to net N₂O consumption. Finally, we test the sensitivity of N₂O distributions simulated by a global ocean model to uncertainties in these values. By geochemically evaluating the dynamics of net N₂O production and consumption, we constrain some of the uncertainties derived from the complex dynamics of individual microbial processes in the ocean."

The present focus on 'net' production and consumption does not adequately account for the range of possible formation pathways with significantly different N_2O yields that operate in low oxygen zones (e.g., nitrification, nitrifier-denitrification, heterotrophic denitrification, see e.g., Frame and Casciotti 2010, Westley et al. 2006, Lipschultz et al. 1990, for specific pathways). A successful predictive ocean model simulation should depend on how the yields from these individual pathways may change as the extent of oceanic low oxygen zones change.

Unfortunately, the data presented in this paper are limited to bulk chemical measurements and so we cannot use the MEMENTO database to determine with certainty each pathway that contributes to the net signal. That being said, the only reason that there would be a substantial deviation from our predictions given an expansion of OMZ regions is if for some reason the relative contribution of different processes occurring at a given O_2 range changes substantially. While we cannot rule out that this may occur, we currently do not have any reason to believe that it would.

BGD

9, C5302–C5317, 2012

Interactive Comment



Printer-friendly Version

Interactive Discussion



In theory, expansion of OMZs into new regions with different temperatures or mixing conditions could affect yields at a given O_2 concentration- and it is true that we cannot extrapolate to other conditions that our data don't cover. However, we can interpolate over the environmental conditions that are included in the data, and these data were gathered from a wide range of conditions. Given our relatively wide range of temperature data (see Fig. 8), temperature does not seem to be a major influence on net N₂O production. Additionally, our data include a wide range of mixing conditions within the ocean (see Fig. A1 and related discussion in Appendix 1) and the data were also aathered from both eutrophic and oligotrophic regions (see our Figures 2 and A2). The great deal of scatter in Fig. 5 where we derived the relationship between net N_2O production and O_2 concentrations might be due to some of these variations in temperature, mixing, and other factors related to geographic distribution. However, despite the scatter, the trend in the relationship between N_2O_{xs} /AOU ratios and O_2 concentrations shown in Figure 5 remained relatively consistent for all the data, indicating that the relationship overall is not particularly sensitive to changes in these conditions. The same is true for the data shown in Fig. 8 that we used to derive the switching point between net N₂O production and consumption.

Also, note that the ETP already experiences large changes in the OMZ locations due to climatic fluctuations such as the Pacific Decadal Oscillation (PDO) and the El Niño Southern Oscillation (ENSO) (Czeschel et al., 2012), not to mention seasonal changes. Thus, the MEMENTO data, which were collected between 1976-2009 during various phases of the PDO and ENSO already incorporate much of the variability that future climate change might accentuate. This is particularly true because the core of the OMZ should remain in the same location in the future- it is only the edges of the OMZ that will be subject to change from climatic fluctuations.

Because it seems unlikely that the relative contribution of each process contributing to N_2O production at a given O_2 concentration will change substantially, and because our data span a large range of O_2 conditions (O_2 values range from 0 to 278 μ M),

BGD

9, C5302–C5317, 2012

Interactive Comment



Printer-friendly Version

Interactive Discussion



we believe that our model does offer some predictive capacity with respect to future changes in the OMZ.

As a final note, we would like to mention that it would probably be possible to use an ecological/biogeochemical ocean model to study how sensitive each of the formation pathways the referee mentions might be to potential future large changes in T, O_2 , and nutrients. While not within the scope of this paper, this would be an interesting topic for a future study. To address the referee's concern, we have added the following cautionary text at the beginning of the results section:

"Thus, N₂O production derived from N₂O_{xs} /AOU ratios is best described by a slope of two times the observed slope with respect to O₂ (in this case, 0.00048 nmol N₂O / μ mol O₂). It is important to note that while this relationship holds within the conditions that the data cover, we cannot extrapolate this relationship to other conditions (i.e., temperature, O₂, etc.) not included in this dataset. "

2) LAYOUT AND USE OF MODEL ANALYSES: The current layout of the manuscript, and in particular, the role of the model simulations, is not always clear. E.g., the methods section (section 2) begins with the ocean model description, however the rationale for some of the parameterizations of N_2O production and consumption does not appear until much later in section 3. It may be clearer to present the data analysis early on (before the model discussion), so that the reasons for the specific model simulations are clearer.

We have made the suggested change.

There is also insufficient detail on how the model was used (see, e.g., Comment 4 in Specific Comments), and on the validity of using a coarse-resolution ocean model to characterize nitrogen cycling processes and oxygen distributions in the ETP that display significant heterogeneity on scales of 1-100 km.

9, C5302–C5317, 2012

Interactive Comment



Printer-friendly Version

Interactive Discussion



As requested, we have provided more details on how the model was used (please see the response to "Specific comment" 4 below for a more complete description of these changes).

The referee also was concerned about the validity of using a coarse-resolution model. We think that the model was valid for the purposes it was used in this study (i.e. to measure overall sensitivity of bulk N₂O concentrations to N₂O consumption rates and to switching points). This is because N₂O concentrations in the model were based on only O₂ concentrations and O₂ consumption rates. While there is meso-scale heterogeneity of both of these components in the real ocean, the majority of variation in O₂ concentrations and consumption is captured on regional scales.

There is some discussion on pg. 10023 (lines 6-10) of using a higher resolution MOM4 model for 'improved accuracy of current velocities', however, there is no discussion on whether the model's nutrient, oxygen and N₂O fields were also treated at high-resolution, or of the possibilities of inconsistency if using different models for velocity fields vs. for the N₂O and O₂ fields.

To clarify, the higher resolution MOM4 model was only used for currents, not for biogeochemistry, and only in the context of deriving transport velocity of water masses in the PCUC for estimating N₂O consumption rates in equation 5. Only one model was used for biogeochemical parameters, and there was no inconsistency between modeled N₂O and O₂ fields. For further clarification and revisions to the manuscript regarding this question, please also see response to Specific comment 4.

Overall, the manuscript could be improved by some restructuring and refocusing on a single major aim, and by additional detail on the model and on how the model analyses were used.

Thank you for your suggestion, we hope that the changes described in this response have made the paper more clear.

BGD

9, C5302–C5317, 2012

Interactive Comment



Printer-friendly Version

Interactive Discussion



SPECIFIC COMMENTS

1) Abstract: Lines beginning "In light of the expected deoxygenation... it is possible that N_2O concentrations may decrease rather than increase as oxygen minimum zones expand". As it is currently worded, this conclusion seems speculative and is not substantiated by any specific finding in this study. The main conclusions that can be drawn are that a significant degree of uncertainty surrounds N_2O production and consumption in the low oxygen zones, if these oxygen minimum zones expand.

We have made minor changes to the abstract:

"Based on these findings, it appears that recent studies substantially overestimated N_2O production in the ETP. In light of expected deoxygenation and the higher than previously expected point at which net N_2O production switches to consumption, there is enough uncertainty in future N_2O production that even the sign of future changes is still unclear."

The referee felt that we did not initially provide enough support for this final statement in the abstract, and indicated that all that is really known is that the uncertainty is large. We agree with the referee that the uncertainty is large, but we differ in that we believe that the uncertainty is large enough that it includes the possibility that future N₂O concentrations might decrease. In other words, we currently have no guarantee that the sign for N₂O changes in the ETP won't be negative. The factors that affect N₂O concentration (namely future O₂ extent and the switching point at which N₂O consumption and production occurs) are highly uncertain. As pointed out by the referees, although we think that the switching point of 10 μ M O₂ is as good a value as can be obtained from our dataset, there is still high uncertainty in this value. Our model results shown in Fig. 6a indicate that the switching point is a vital parameter for O₂ concentrations. If consumption regions expand at the expense of production regions, N₂O concentrations could theoretically decrease. 9, C5302–C5317, 2012

Interactive Comment

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



We would also like to point out that although not discussed in the paper, we have run a future model scenario with the UVic-ESCM model used in this paper going to the year 2100 using the RCP8.5 future atmospheric CO₂ and N₂O scenarios (and using our recommended switching point of 10 μ M O₂ and a N₂O consumption rate of 0.1 mmol m-3 yr-1). Those model data indicate a 20% reduction in N₂O in the ETP (although note again that there is substantial uncertainty in modeled future O₂ concentrations). For further discussion on the difficulties comparing model O₂ and observations, see Stramma et al. (2012).

2) Pg 10021 (lines 15-18): It is not clear why different O_2 thresholds should not apply in different oceanic environments; i.e., the threshold may depend on which mechanisms for N_2O production or consumption dominate for that given environment, and this would be reflected in the range of threshold levels reported in the literature.

This is a good point. We have reworded this sentence as follows:

"It is also not clear whether there is a single O₂ value that is representative of the point at which net N₂O production switches to N₂O consumption. Current estimates for this value within the ETP range from 1–20 μ M O₂ (e.g. Farias et al., 2009, and Suntharalingam et al., 2000)."

3) Pg 10022 lines 23-25 and pg 10024 (lines 1-4): As discussed above in the General Comments, it would be clearer if the rationale for the selection of these parameterizations for N_2O production and consumption rates were discussed early on, preferably before the model discussion.

We have done this.

4) Pg 10024, lines 6-11: Please provide a more detailed description of how exactly the MOM4 model was used in these analyses. Were only the velocity fields from MOM4 used? Were they averaged to the larger grid size of the UVic-ESCM? How were the

BGD

9, C5302–C5317, 2012

Interactive Comment

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



model's nutrient, N_2O and oxygen fields treated? Would there be a problem of inconsistency between the velocity values (from MOM4) and biogeochemical distributions from the UVic–ESCM?

We have clarified the text as the author suggested (see text below with changes in bold):

For improved accuracy of current velocities in the Peru-Chile Undercurrent (section 3.3), we used a higher resolution version of the global ocean model (the Geophysical Fluid Dynamics Laboratory Modular Ocean Model 4, or MOM4) (Zamora et al., 2010). The MOM4 model was only used to calculate velocity fields; the sensitivity analyses and derivation of biogeochemical distributions (e.g., N₂ O, O₂) were derived separately and solely from the UVic-ESCM discussed previously. This version of the MOM4 model had a 10 longitude by 10 latitude resolution with 50 vertical layers.

5) Pg 10024 (lines 15-16): Discussion of the TTD method. Please clarify how the 'modeled N_2O_{eq} ' value is obtained.

We have added the following text to the Methods section in question:

"The TTD method cannot be tested with in situ observations since it is impossible to know the actual N₂O_{eq} values with certainty. Therefore, we tested the validity of the TTD method by comparing modeled N₂O_{eq} with estimated N₂O_{eq}. Modeled N₂O_{eq} was obtained from our UVic-ESCM model by turning biological N₂O consumption and production off, so that the only factor affecting modeled N₂O concentrations was air-sea gas exchange, atmospheric N₂ O, and physiochemical properties of the water affecting this exchange (salinity, temperature, etc.). We then compared modeled N₂O_{eq} with estimated N₂O_{eq}, whereby atmospheric N₂O was estimated by three methods: (1) the current model

BGD

9, C5302–C5317, 2012

Interactive Comment

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



year (which would correspond to sample date if this method were used for observational data, as in Elkins et al., 1978), (2) atmospheric N₂O as estimated by modeled pCFC-12 age (which was calculated by matching observed pCFC-12 concentrations to the historic pCFC-12 concentration that would arise given the salinity and temperature of that water mass using solubility calculations from Warner and Weiss, 1985), and (3) the TTD age based on modeled pCFC-12 ages."

6) Pg 10026 (lines 27+) to Pg10027 (first paragraph): Filtering of data to exclude samples with $O_2 < 10$ micromol in calculating N_2O production. Will this procedure also exclude regions of potential N_2O production though mechanisms of nitrifier-denitrification or heterotrophic denitrification?

It is possible that N₂O production by these two mechanisms does occur at O₂ <10 μ M, and we would not be able to observe it in the bulk chemical data because N₂O consumption removes this N₂O before it accumulates. We have added the following qualifier in the text concerning this point:

"Another uncertainty is that N_2O production in regions of net N_2O consumption via nitrifier-denitrification or heterotrophic denitrification could reduce the net consumption rate observed here."

7) Pg 10029 (lines 25+) to pg 10030 (lines 1-3): Discussion of N₂O production from pathways other than bacterial nitrification: It should be noted that the yields of N₂O from these other pathways (which operate primarily at low O₂ levels) can be significantly larger than from bacterial nitrification, therefore the impact on overall N₂O yield could be a non-linear increase as O₂ levels decline.

We agree that non-bacterial nitrification may be of great importance. With regard to non-linearity, the actual (combined bacterial and non-bacterial) production appears to be quite linear, which contradicts a simultaneously dominant and non-linear contribu-

BGD

9, C5302–C5317, 2012

Interactive Comment



Printer-friendly Version

Interactive Discussion



tion from non-bacterial sources. We have added the following changes to the paragraph in question (changes in bold):

"For comparison in Fig. 5, we plot the expected exponential relationship between N_2O_{xx} /AOU ratio and O_2 based on Goreau et al. (1980) as defined in Eq. (6) in Nevison et al. (2003). Nevison et al. (2003) hypothesized that the lack of observed in situ exponential behavior might have been due to mixing with N_2 O-depleted waters from OMZs. In addition to mixing, it is also possible that the lack of observed exponential behavior could be explained by a smaller than previously expected volume of water in which conditions exist where exponential behavior would be observable (as the data of Frame and Casciotti, 2010, seem to suggest). It is also worthwhile to emphasize that the Goreau et al. (1980) study was limited to bacterial nitrification. Therefore, it is possible that N₂O production from other sources, such as denitrification (e.g. Farias et al., 2009) or archaeal nitrification, is larger than previously thought and that these sources do not exponentially increase as O_2 declines (although laboratory data do suggest some sort of increase in yields of N₂O at low O₂ levels from these processes (Knowles et al., 1981; Loescher et al., 2012; Payne et al., 1971)). Independent of mechanism, it appears that the best description of net N_2O production in the ETP is a linear, not exponential, function of decreasing O_2 ."

8) Pg 10032-10033 Section 3.3: "Switch between net N₂O production and consumption." The rationale for and conclusions of this section need to be laid out more clearly. Currently it is not clear whether it is focused on identifying oxygen thresholds for application in ocean models, or identifying nitrite distribution characteristics that mark the onset of denitrification in the ETP. Since the authors also note the reporting of lower oxygen thresholds for nitrite accumulation from other studies that may reflect differences between oceanic environments (lines 25+ on pg 10033), they should be cautious about recommending a globally applicable higher O_2 threshold for use in ocean

BGD

9, C5302–C5317, 2012

Interactive Comment

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



models.

We have added a sentence into the introductory paragraph for this section in order to clarify our goals. To answer the referee's question, our goal was to identify O_2 thresholds for application in ocean models. However, the method we used to do so involved relating the beginning of net N_2O consumption with observable accumulation of NO_2 -, and perhaps this is where the confusion arose.

The rationale for the conclusions have also been laid out more clearly in this context (please note also the revised text and the related response to referee 2 below).

The referee also suggested that we be more cautious about suggesting a globally applicable higher O_2 threshold for use in ocean models. Although recommending that our results be applied on a global scale was not our intent (we are not sure how this impression arose), we recognize that this is an important point to address. In response to these concerns, we have changed the text as follows:

"It is well known that as oxygen concentrations are reduced, eventually net N₂O production switches to net N₂O consumption, presumably due to denitrification. **However, the exact O₂ concentration at which net N₂O consumption begins is not well defined and may not be consistent among regions.** Models estimating N₂O production have previously used values of 1-4.5 μ M O₂ (Suntharalingam et al., 2000; Jin and Gruber, 2003; Nevison et al., 2003; Freing et al., 2012; Bianchi et al., 2012). However, literature estimates specific to the ETP place the switching point between 5-20 μ M O₂ (Nevison et al., 2003; Farías et al., 2009; Cornejo and Farías, 2012; Ryabenko et al., 2012), indicating that the switching point may be higher than models have so far accounted for in this region. In this section, we identify the O₂ concentration that best represents the point at which net N₂O production switches to net N₂O consumption for application in biogeochemical models of the ETP."

BGD

9, C5302–C5317, 2012

Interactive Comment

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



9) Pg 10035 Lines 16-18: Sentence beginning "If there is an increase in the volume....". As also noted for the Abstract, this conclusion seems too speculative; the main point should be that there is significant uncertainty on how N_2O production and consumption will change if the extent of the ocean's low oxygen zones change.

Please see our response to "Specific comment"1 above.

10) Table 1: It would be useful if more details on the calculations and different model versions used in Table 1 were presented somewhere in the manuscript; e.g., in an Appendix.

We have added this information to new Appendix C.

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Interactive Comment

BGD

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



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BGD

9, C5302–C5317, 2012

Interactive Comment

Full Screen / Esc

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Interactive Discussion



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