We thank the reviewers for their valuable and insightful comments, which have improved our manuscript substantially. We provide our responses (plain text with blue color) to all comments (italic text) below.

- Reviewer #1 -

#### 1. General comment

(1) The authors present a modeling study whereby they looked for estimating  $N_2O$  levels in nearbottom waters of Northern Gulf of Mexico (the nGOM) for the period comprised between 1985 and 2007 (summer period). These estimates may be important for understanding the marine/estuarine  $N_2O$  cycle and its environmental controls, and thus this manuscript could be an important contribution to the field.

 $\rightarrow$  We appreciate to the reviewer's supportive statement.

\*Note: We cite Kim and Min (2013) instead of Kim (2012). The manuscript of Kim and Min (2013) is under revision.

# 2. Major points

(1) Important pieces of information are omitted throughout the manuscript and there is a lack of logical sequence that triggers a series of questions throughout the whole manuscript. It had been very difficult for me to understand the used conceptual model (rationale) and the relationships among  $O_2$ ,  $N_2O$  and  $NO_3^-$  in order to reproduce  $N_2O$  concentration in the bottom water under hypoxia condition, most of the time. I guess that the exercise was only done for bottom water to prevent further advection (vertical) and diffusion effects, but what about the role of lateral advection, resuspention, pelagic benthic coupling, etc.?

→ We apologize for the insufficient explanation about the conceptual model (Equation 1). In providing a more inclusive explanation we discovered that the equilibrium term  $([N_2 O]_{equilibrium}^{(T,S)})$  had been neglected. We have therefore re-calculated all estimates and revised all results. None of our conclusions have changed.

 $\rightarrow$  A. Detailed explanation for the conceptual model is now provided in the manuscript and is included below for the convenience of the review.

The tracer continuity equation for  $\Delta N_2 O$  (=  $[N_2 O]_{estimated}$  –  $[N_2 O]_{equilibrium}$ ) in the bottom layer associated with microbial processes (i.e. nitrification and denitrification by bacteria and archaea) is expressed as follows:

$$\Gamma(\Delta N_2 0) = J_{nitrification}(\Delta N_2 0) + J_{low oxygen}(\Delta N_2 0) - J_{low oxygen}(\Delta N_2 0)$$
(1)

, where  $J(\Delta N_2 O)$  represents the function describing net production (i.e. source) minus consumption (i.e. sink) for each process.  $J_{\text{nitrification}}$  ( $\Delta N_2 O$ ) denotes the source term for nitrification,  $+J_{\text{low oxygen}}$  ( $\Delta N_2 O$ ) denotes the N<sub>2</sub>O production during low O<sub>2</sub>, and  $-J_{\text{low oxygen}}$ ( $\Delta N_2 O$ ) denotes the N<sub>2</sub>O consumption during low O<sub>2</sub> (Fig. A1). The operator  $\Gamma$  is the transport and time rate of change and is given as:

$$\Gamma(C) = \frac{\partial C}{\partial T} + U \cdot \nabla C - \nabla \cdot (D \cdot \nabla C)$$
<sup>(2)</sup>

, where C is any tracer concentration,  $\bigtriangledown$  denotes the gradient operator in the x, y, and z directions, U is the 3-dimensional velocity field, and D is the eddy diffusivity.

Under low oxygen conditions, such as suboxic and anoxic conditions, anaerobic ammonium oxidation (anammox) and dissimilatory nitrate reduction to ammonium (DNRA) could be additional sources for N<sub>2</sub>O (Naqvi et al., 2010). However, the pathways and yields of N<sub>2</sub>O production during these two processes are poorly known. Furthermore, measurements of anammox and DNRA in the nGOM are in short supply (Dagg et al., 2007). As a result, we did not explicitly include the N<sub>2</sub>O production by anammox and DNRA in our model. Instead, we

assumed that the terms of low oxygen ( $J_{low oxygen}$ ) are determined by denitrification alone. In our conceptual model, we assumed a tri-linear  $\Delta N_2 O/O_2$  relationship (Fig. A1).



Fig. A1. Conceptual illustration showing a tri-linear relationship between  $O_2$  and  $\Delta N_2 O$ .

In reality, the  $\Delta N_2 O/O_2$  relationship may show a non-linear behavior due to the complex inter actions of physical and biogeochemical processes (Nevison et al., 2003); however, the exact shape of the  $\Delta N_2 O/O_2$  non-linear relationship is not known. The use of a simple tri-linear  $\Delta N_2 O/O_2$  relationship may increase the uncertainty of our modeled results but can be taken as a simple best-guess approach. Using the empirically-derived linear relationships (Equation 1),  $\Delta N_2 O$  can be computed as follows:

$$\Delta N_2 O = \alpha \frac{\Delta N_2 O}{AOU} \cdot AOU + \beta \frac{\Delta N_2 O}{\Delta N_{deni}} \cdot \Delta N_{deni} - \gamma \frac{\Delta N_2 O}{\Delta N_{deni}} \cdot \Delta N_{deni}$$
(3)

where AOU is the apparent oxygen utilization – the difference between the measured  $O_2$  concentration and the equilibration value,  $\alpha$  indicates the relationship between  $\Delta N_2O$  and AOU, and  $\beta$  and  $\gamma$  are the relationships between  $\Delta N_2O$  and the amount of denitrification ( $\Delta N_{deni}$ ).

Finally,  $[N_2O]_{est}$  can be estimated as:

$$[N_2 0]_{est} = \Delta N_2 0 + [N_2 0]_{equilibrium}^{(T,S)}$$
(4)

 $\rightarrow$  B. The question on lateral advection is answered in e2. Sensitivity, uncertainty, and a caveat below and is included in the revised manuscript.

#### $\rightarrow$ C. The question on resuspension and pelagic benthic coupling is answered as follows:

The referee correctly pointed out that the following statement "A contribution of N<sub>2</sub>O released from the underlying sediments is expected to be negligible (Visser, 2009) and was, therefore, not accounted for in our model" in Section 2.2 is incorrect. Our intended meaning was that our model cannot directly account for possible  $N_2O$  sediment fluxes, as the data used for our analysis were based on near-bottom hydrographic parameters. Nevertheless, it is likely that these hydrographic parameters are influenced by interaction between bottom waters and sediments. Therefore, we assumed that the estimated N<sub>2</sub>O concentrations from this study were resulted from bottom water and sedimentary processes combined (i.e. benthic coupling processes). In summer, water column stratification caused by density gradients is intensified (Dagg et al., 2007), and therefore the bottom layer is highly stable. Although resuspension enhanced by bottom currents may occur continuously on small scales in the bottom layer in spite of the strong stability, large scale resuspension would require external forcing, such as might be provided through intense storm systems. Walker et al. (2010) reported enhanced N<sub>2</sub>O production after storm, resulting in reoxygenation through vertical redistribution of organic matters in the water column. As the data in Table S1 and the discussion in Section 3.1 indicate, the data sets used for this analysis were little influenced by storm/hurricane events. The statement above that "the estimated N<sub>2</sub>O concentrations in the bottom layer from our analysis are derived from the combined results of bottom water and sedimentary processes" therefore includes small scales of resuspension by bottom currents but neglects large storm events whose effects are not apparent in the data. We will include this statement in the revised manuscript (Section 3.2).

(2) (1) As authors mentioned, nGOM is an extended continental shelf area with hypoxic and eutrophic condition. In this kind of ecosystems, we expect a huge N<sub>2</sub>O accumulation as a product of in situ biological production and/or discharge from rivers, estuaries; particularly as they are being affected by anthropogenic nitrogen (N) inputs. The estimated N<sub>2</sub>O values are really low, even at levels of under saturation, making boast of: → **D**. The range of bottom N<sub>2</sub>O measurements during the summer (July-August) of 2008 was from 4.63 to 30.02 nmol L<sup>-1</sup> (Visser, 2009; Walker et al., 2010). Visser (2009) also measured bottom water N<sub>2</sub>O concentrations in September 2007 and April 2008, and they ranged from 8.44-11.87 and 6.53-9.54 nmol L<sup>-1</sup>, respectively. We compared our estimates with the measurements by Visser (2009) and Walker et al. (2010) (Fig. A2). Our estimates are comparable with the previously published summer measurements. Overall, summer N<sub>2</sub>O concentrations are higher than spring/fall N<sub>2</sub>O concentrations. Comparing our estimates with those from other regions (see Table 2b of Naqvi et al., 2010 and references therein), it can be seen that maximum subsurface N<sub>2</sub>O concentrations in coastal hypoxic systems have a range from 9.8 nmol L<sup>-1</sup> in the Chesapeake Bay to 62.6 nmol L<sup>-1</sup> in the Changjiang Estuary, to 139 nmol L<sup>-1</sup> Tokyo Bay. Estimates from the nGOM (~30 nmol L<sup>-1</sup>) lie in the middle of this range according Naqvi et al. (2010). The maximum N<sub>2</sub>O concentration estimated in our analysis was ~52 nmol L<sup>-1</sup> (Table 2).



Fig. A2 Interannual variation of estimated bottom  $N_2O$  concentrations (blue square) in the nGOM during July 1985-2007. Direct measurements are included (black, green, cyan, and pink squares: September 2007, April 2008, July 2008, and August 2008, respectively).

The description of this comparison is included in the revised manuscript (Section 3.3).

(2) Lack of sensitivity and robustness in the used model - A N<sub>2</sub>O consumption by denitrification, but as it proceeds only in anoxic environments, N<sub>2</sub>O reduction to N2 is unlikely because hypoxic

and suboxic conditions always remain in the bottom water; - A  $N_2O$  consumption by the sediments (denitrification) but in this ms. the role of sediment is ignored.

 $\rightarrow$  E. We have addressed the concern that our model lacks sensitivity and does not produce robust results:

e1. The  $O_2$  criteria for determining nitrogen processes that dominate  $N_2O$  production and consumption

We used Eq. 4 along with observations to estimate the N<sub>2</sub>O concentrations in the near-bottom waters in the nGOM, which nitrogen processes (i.e. nitrification and denitrification) dominate N<sub>2</sub>O production/consumption depend primarily on O<sub>2</sub> concentrations. As little information is available on the O<sub>2</sub> threshold for N<sub>2</sub>O production by denitrification, we deduced  $\beta$  and  $\gamma$  from the results of Farías et al. (2009) for our analysis. However, it should be kept in mind that these values were based on measurements on eastern tropical South Pacific, and may not represent processes occurring in the nGOM.

To evaluate the dependence of our N<sub>2</sub>O estimation to these O<sub>2</sub> conditions, we consider two cases (Table A1). Case 1 (Table A1) characterizes the O<sub>2</sub> conditions as stated above. Under oxic conditions, N<sub>2</sub>O is produced by nitrification only, and thus the concentration is calculated as  $\alpha \times$  AOU (Yoshinari, 1976; Cohen and Gordon, 1979; Oudot et al., 1990). Recently, Farías et al. (2009) showed net N<sub>2</sub>O production around hypoxic O<sub>2</sub> levels in the eastern tropical South Pacific (see their Table 1). Under hypoxic conditions, during which nitrification and denitrification are both involved in N<sub>2</sub>O production (Naqvi et al., 1998; Nevison et al., 2003), our N<sub>2</sub>O concentration is calculated as  $\alpha \times AOU + \beta \times \Delta N_{deni}$ . It has been reported that N<sub>2</sub>O consumption occurs at O<sub>2</sub> < ~4  $\mu$ M (≈0.13mg L<sup>-1</sup>) (Nevision et al., 2003 and references therein). Under such suboxic-anoxic conditions, denitrification produces N<sub>2</sub>O (Cohen and Gordon, 1978; Elkins et al., 1978; Yamagishi et al., 2007), and nitrification produces N<sub>2</sub>O via nitrite reduction (nitrifier denitrification: NH<sub>4</sub><sup>+</sup> → NO<sub>2</sub><sup>-</sup> → N<sub>2</sub>O) (Poth and Focht, 1985; Wrage et al., 2001). Therefore, under these conditions N<sub>2</sub>O concentration is calculated as  $\alpha \times AOU - \gamma \times \Delta N_{deni}$ .

Case II (Table A1) is given and was computed for three conditions: (1) oxic-hypoxic conditions as  $O_2 > 0.14 \text{mg L}^{-1}$  ( $\alpha \times \text{AOU}$ ), (2) suboxic conditions as  $0.07 \le O_2 \le 0.14 \text{mg L}^{-1}$  ( $\alpha \times \text{AOU} + \beta \times \Delta N_{\text{deni}}$ ), and (3) suboxic-anoxic conditions as  $0 \le O_2 < 0.07 \text{mg L}^{-1}$  ( $\approx 2.2 \ \mu\text{M}$ ) ( $\alpha \times \text{AOU} - \gamma \times \Delta N_{\text{deni}}$ ).

Case I					
O <sub>2</sub> levels	Conditions	N process N <sub>2</sub> O estimatio			
2 < O <sub>2</sub>	Oxic	Nitrification	$\alpha \times AOU$		
$0.14 \le O_2 \le 2$	Hypoxic	Nitrification + denitrification (+)	$\alpha \times AOU + \beta \times \Delta N_{deni}$		
$0 \le O_2 < 0.14$	Suboxic-anoxic	Nitrification + denitrification (-)	$\alpha \times AOU - \gamma \times \Delta N_{deni}$		
Case II					
O <sub>2</sub> levels	Conditions	N process N <sub>2</sub> O estimation			
$0.14 < O_2$	Oxic-hypoxic	Nitrification	$\alpha \times AOU$		
$0.07 \le O_2 \le 0.14$	suboxic	Nitrification + denitrification (+) $\alpha \times AOU + \beta \times \Delta N_{deni}$			
$0 \le O_2 < 0.07$	Suboxic-anoxic	Nitrification + denitrification (-)	$\alpha \times AOU - \gamma \times \Delta N_{deni}$		

Table A1. Summary of Case I and II conditions

The revised manuscript includes the description above in Section 2.3.

### e2. Sensitivity, uncertainty, and a caveat

We assigned 0.048, 0.83, and 0.83 for  $\alpha$ ,  $\beta$ , and  $\gamma$ , respectively, to estimate bottom water N<sub>2</sub>O concentrations using the empirical relationship presented in Section 3.1. To investigate the validity of chosen values of  $\alpha$ ,  $\beta$ , and  $\gamma$ , we used a Monte Carlo technique generating random numbers for individual  $\alpha$ ,  $\beta$ ,  $\gamma$  values within expected ranges looking for those coefficients that produced estimated bottom N<sub>2</sub>O concentrations within the observed range. The value of  $\alpha$  ranges from 0.048 to 0.31 in ocean environments, including the nGOM (Suntharalingam and Sarmiento, 2000 and references therein). For our model simulation we generated random numbers for  $\alpha$  between 0 and 0.31. Although we deduced  $\beta$  and  $\gamma$  from the results of Farías et al. (2009),

information on  $\beta$  and  $\gamma$  outside the tropical South Pacific lacks. Therefore, we increased the possible range in these values that generated random numbers for  $\beta$  and  $\gamma$  between 0 and 2.5. In the nGOM, Visser (2009) and Walker et al. (2010) directly measured water column N<sub>2</sub>O concentrations during the summer of 2008, and reported that bottom N<sub>2</sub>O concentrations ranged from 4.25 to 30.02 nmol L<sup>-1</sup> (July) and from 5 to 30 nmol L<sup>-1</sup> (August). Bottom N<sub>2</sub>O concentrations were similar in both months, so we used the median of measured values (17–18 nmol L<sup>-1</sup>) as the acceptable zone, and then applied the random  $\alpha$ ,  $\beta$ , and  $\gamma$  values to the data sets to estimate N<sub>2</sub>O concentrations. When the mean N<sub>2</sub>O concentration reproduced by random  $\alpha$ ,  $\beta$ , and  $\gamma$  values fell in the acceptable range (i.e. 17 < mean [N<sub>2</sub>O] < 18), they were saved and averaged. We generated a thousand of random numbers for individual  $\alpha$ ,  $\beta$ , and  $\gamma$  coefficients averaged over all the simulations were 0.051±0.003, 1.27±0.15, 1.16±0.16, respectively.



Fig. A3 Simulation results of randomly generated  $\alpha$ ,  $\beta$ , and  $\gamma$  producing N<sub>2</sub>O estimates in the acceptable range (i.e. 17 < mean [N<sub>2</sub>O] < 18).

The  $\alpha_{=0.048}$  obtained from the empirical relationship compared well with the simulated  $\alpha_{=0.051\pm0.003}$ . The  $\beta_{=0.83}$  and  $\gamma_{=0.83}$  are somewhat lower than the simulated  $\beta_{=1.27\pm0.15}$  and  $\gamma_{=1.16\pm0.16}$ . The change of N<sub>2</sub>O concentration according as  $\alpha$ ,  $\beta$ , and  $\gamma$  individually change with 0.01 interval was ~1.3, ~0.03, and ~0.001 nmol L<sup>-1</sup>, respectively. The dependence of  $\alpha$  among three components in the estimation was larger.

To examine the sensitivity on the threshold  $O_2$  values for  $N_2O$  production/consumption by denitrification (Table A1), Case I estimates were compared with Case II. On average, the  $N_2O$  concentrations estimated for Case I were ~1.3 nmol L<sup>-1</sup> higher than those for Case II (Fig. A4). The magnitude of this difference is dominated by 6 individual years out of 20 years. Also, both temporal trends were similar. Thus, the results of Case I are primarily used for our study.



Fig. A4 Interannual variation of estimated bottom N<sub>2</sub>O concentrations according to Case I and II formula.

We used  $\Delta N_{deni}$  estimated from the eOMP analysis by Kim and Min (2013) for representing N<sub>2</sub>O production/consumption by denitrification. Kim and Min (2013) defined 4 different water masses (i.e. SUW: Subtropical UnderWater, TLCW: Texas-Louisiana Coastal Water, ADW: Atchafalaya Discharge Water, and MDW: Mississippi Discharge Water) for the eOMP analysis in the study area. Unlike the others, SUW is a foreign water mass that occasionally intrudes into the study area and is dependent on eddy development/circulation patterns. Although it would be difficult to estimate preformed N<sub>2</sub>O amounts for each water mass from this analysis, because SUW locally occupies the bottom layer and is derived from outside the study area, it is possible to approximate the amount N<sub>2</sub>O advected by SUW using a plot of the estimated bottom N<sub>2</sub>O

concentrations vs. the mixing ratios of SUW (Fig. A5). Higher mixing ratios of SUW imply that the water properties are close to those of source water mass, and by using a constraint on SUW mixing ratios of > 90% (blue dots), the N<sub>2</sub>O concentrations produced through advection of SUW into the region can be ~13.3±2.5 nmol L<sup>-1</sup>, which is mean value averaged from the N<sub>2</sub>O concentrations of blue dots (Fig. A5). This estimate is close to the mean concentration of ~14.5±2.3 nmol L<sup>-1</sup>. However, SUW locally distributes in the study area (~89.5°W–92W and 28.5–29.0°N deeper than ~40m isobath), and its occupation in the nGOM bottom waters has been decreased since ~1998 (Kim and Min, 2013). Overall, the effect of N<sub>2</sub>O advected by SUW on our estimation is small.



Fig. A5 Plot of estimated bottom N<sub>2</sub>O concentrations vs. SUW mixing ratios (%).

Here we estimated bottom N<sub>2</sub>O concentrations using the tri-linear relationships between O<sub>2</sub> and  $\Delta$ N<sub>2</sub>O. This approach provided a representation of interannual N<sub>2</sub>O variations in the nGOM. However, we have several limitations, such as: i) the lack of  $\alpha$  variability in the nGOM (i.e. interannual and seasonal), ii) little information on  $\beta$  and  $\gamma$  in the nGOM, and iii) unknown the exact shape of the  $\Delta$ N<sub>2</sub>O/O<sub>2</sub> relationship. Due to such limitations, we constrain that our estimates are valid for the summertime within the nGOM. In order to accurately estimate N<sub>2</sub>O concentrations using modeling approaches, integrated information on  $\alpha$ ,  $\beta$ , and  $\gamma$  based on direct measurements will be needed to establish a non-linear equation in the future. We will include this discussion in the revised manuscript (Section 3.2).

 $\rightarrow$  **F**. The concern that the role of sediments is ignored is answered in our response **C** above.

### 3. Minor points

(1) There are inconsistencies among the presented background in introduction section, the used conceptual biogeochemical model, results and others. Since the study is focused on a hypoxic region, it precludes the existence of anaerobic process as canonical denitrification or DNRA, I wonder if the study area has representative anoxia period. Let include information about frequency of anoxic periods. Table 1 reveals DO as low as 40 nmol  $L^{-1}$ , it is far to be suboxic (4.4 µmol  $L^{-1}$ ). Introduction section only mentioned N<sub>2</sub>O producing processes; how about N<sub>2</sub>O consuming process?. However, conceptual model incorporates a terms of N<sub>2</sub>O consuming,. Again, how you weigh or ponders the role of canonical denitrification in the study area? The modeled mean N<sub>2</sub>O concentration in the bottom water was 7.7±6.7 nmol  $L^{-1}$ ; it means a range from 10% to 234% saturation. I wonder if there are data with under saturation levels in other eutrophicated and hypoxic ecosystems. Under saturated N<sub>2</sub>O concentrations are usually found in suboxic/anoxic and nitrite-rich waters, as observed of the oxygen minimum zones or the sediments. This pattern has been clearly ascribed to canonical denitrification to N<sub>2</sub> under anoxic condition.

 $\rightarrow$  G. We did not preclude the low oxygen processes (e.g. denitrification). Under low oxygen conditions, such as suboxic and anoxic conditions, anaerobic ammonium oxidation (anammox) and dissimilatory nitrate reduction to ammonium (DNRA) might be additional sources for N<sub>2</sub>O (Naqvi et al., 2010). However, the pathways and yields of N<sub>2</sub>O production during these two processes are unknown. In addition, measurements of anammox and DNRA in the nGOM are not

available (Dagg et al., 2007). Therefore, we did not explicitly include N<sub>2</sub>O production by anammox and DNRA in our model approach and assume that the terms for low oxygen conditions ( $J_{low oxygen}$ ) in Equation (1) are determined via denitrification (More details are described in our response A). Additionally, it should be noted that anoxic conditions (O<sub>2</sub>=0 mg L<sup>-1</sup>) did not develop in the nGOM during the study period (Fig. A6).



Fig. A6 Interannual variation of bottom  $O_2$  concentrations (mg L<sup>-1</sup>) in the nGOM during July 1985-2007. The dotted line indicates the threshold  $O_2$  value for suboxic conditions (0<O<sub>2</sub><0.14 mg L<sup>-1</sup>).

Table 1 and Fig. 3 were used to simulate  $\alpha$  variability due to change in physical conditions, for example, pre-storm vs. post-storm. As the table and figure appear to be cause confusion, they were removed from the revised manuscript.

The reviewer is correct that role of  $N_2O$  consumption had been left out of Introduction. In the revised manuscript, the role of  $N_2O$  consumption is added to Introduction. However, the role of denitrification was included in our model (see Eq. 3).

Our equations did not include the equilibrium term  $([N_2 O]_{equilibrium}^{(T,S)})$ . So we recalculated all estimates and include all revised estimates in the revision (Details are described in our response **A**). We thank the reviewer for finding this inadequacy.

The revised mean  $N_2O$  concentration estimated in the bottom layer is 14.5±2.3 nmol L<sup>-1</sup>.  $N_2O$  saturation is computed as follows:

$$N_2 O saturation(\%) = \frac{[N_2 O]_{estimated}}{[N_2 O]_{equilibrium}^{(T,S)}} \times 100$$
(5)

The resulting bottom N<sub>2</sub>O saturations estimates are shown in Fig. A7.



Fig. A7 Interannual variation of bottom N<sub>2</sub>O saturation (%) in the nGOM.

Mean N<sub>2</sub>O saturation (%) for bottom water was ~238±40%, which is somewhat higher than, but still comparable to the July 2008 measured value of  $171\pm111\%$  reported by Visser (2009). We divided the mean bottom N<sub>2</sub>O concentrations into 4 components (Fig. A8): production by nitrification, production by denitrification, consumption by denitrification, and N<sub>2</sub>O equilibrium. N<sub>2</sub>O saturation was relatively low in 1998, but relatively high in 2002 (Fig. A7). In 1998, N<sub>2</sub>O consumption by denitrification (sink) was higher, causing the lower saturation level, while in 2002 N<sub>2</sub>O production by denitrification (source) was higher, resulting in a higher saturation level.



Fig. A8 The four components of estimated  $N_2O$  (purple stars): production by nitrification (red squares), production by denitrification (green squares), consumption by denitrification (blue squares), and  $N_2O$  equilibrium (black squares).

(2) In my opinion the production of  $N_2O$  is very sensitive to changes in oxygenation but also depends on several factors. If authors assume that oxygen is the dominating factor (80%) controlling  $N_2O$  cycling and that there are not limitations for ammonium (electron donor for nitrification) or nitrate/nitrite (electron acceptor for denitrifies), Thus, those assumptions should be clearly established. Regarding the scale used to separate concepts of oxia, hypoxia, suboxia and anoxia, it is confusing because authors use the same range to separate anoxic suboxia, and I think this is a mistake, because the nitrogen cycle (particularly  $N_2O$ ) behaves very differently depending on  $O_2$  traces or not exist in the environment. The authors must to include an anoxic term. I think that the best definition is those stated out by Naqvi who defined anoxia when  $O_2=0$  $\mu$ mol  $L^{-1}$  and  $NO_2^- > 0$ . I believe that  $NO_2^-$  distribution should really help to define  $O_2$  ranges. Please include that data.

 $\rightarrow$  Please refer to our response **e1** above.

(3) The rationale of  $N_2O$  conceptual model is very difficult to follow to me. Dynamic patterns of dissolved  $N_2O$  concentrations in marine waters emerge from complex interactions among physical, biogeochemical processes. Therefore, since the model only includes a microbiological term, it is a very biased conceptual framework. What is  $\Delta O_2$  is the Eq. 1 ? and What include the term  $\Delta N_{deni}$ ,  $NO_3^-$  and  $NO_2^-$  loss as  $N_2$ ? What do you think about N-loss by anammox? Could  $\Delta N_{deni}$ , be being underestimated? cWhat kind of (lineal exponential, polynomial) relationship was used to estimate  $\alpha$ ,  $\beta$ , and  $\gamma$ ? It is widespread knowledge that and relationships between  $O_2$ (AOU) and  $N_2O$  ( $\Delta N_2O$ ) and even  $NO_3^-$  are not linear. For example, these relationships depend on (besides oxygen) water masses mixing, the microbial communities (functional and even phylogenetic diversity) among others. So, it is no possible to use parameterizations obtained in other ecosystems, like the OMZ of the eastern South Pacific. Finally, N<sub>2</sub>O cycling should comprise the conceptual model: Oxic Condition should include aerobic ammonium and nitrite oxidation (AAO and ANO, respectively) by Bacteria and Archaea (only NH4 oxidation) Hypoxic Condition: AAO ANO, partial denitrification, if you a priori preclude nitrifier denitrification Suboxic Condition: idem to hypoxic condition Anoxic Condition: total o canonical denitrification, if you a priori preclude DNRA.

## $\rightarrow$ H. Please refer to our response A above.

To avoid confusion we now use AOU instead of  $\Delta O_2$  (see Equation 3 above).

Kim and Lee (2013) estimated  $\Delta N_{deni}$  using the extended OMP analysis. The  $\Delta N_{deni}$  is estimated as the difference between observed and estimated N, i.e., N deficit, assuming that a N deficit results from the denitrification processes. As mentioned in our response A, measurements of anammox and DNRA in the nGOM are not yet available (Dagg et al., 2007). If anammox is a dominant process for N removal in the nGOM, as the reviewer suggests, then our  $\Delta N_{deni}$  could be underestimated. In principle, a completely anoxic environment is required for anammox to occur, but during the study period in the nGOM, O<sub>2</sub> levels never reached anoxic levels (Fig. A6). Although beyond the scope of this work, certainly, future investigations into the nitrogen cycle in the nGOM would do well to take a closer look at this question. The exact shape of the  $\Delta N_2O/O_2$ non-linear relationship is not known, but as discussed above, we assumed a simple best-guess approach using a tri-linear  $\Delta N_2O/O_2$  relationship (Fig. A1) to represent what is likely a complex interplay of physical and biogeochemical processes (Nevison et al., 2003) that results in nonlinear behavior. It may be true as the reviewer suggests that it is not possible to base our approach on "*parameterizations obtained in other ecosystems, like the OMZ of the eastern South Pacific*", due to the different physical characteristics, diversity of bacterial communities, and so on. We agree. However, one advantage of the modeling approach is that it allows representation of the target we want to predict with definable constraints, even though there are large uncertainties. To support the validity of our approach, we provided the results of sensitivity analysis (Details are described in our response e2). In addition, since there is little information on a threshold of O<sub>2</sub> value for N<sub>2</sub>O production by denitrification, we deduced it from the results of Farías et al. (2009) for our analysis. We recognize and state in the revised manuscript that it is uncertain whether this threshold is representative of processes in the nGOM. Also, we examined the sensitivity on the threshold O<sub>2</sub> values for N<sub>2</sub>O production/consumption by denitrification (Details are described in our responses e1-2). Nitrification consists of aerobic ammonium and nitrite oxidation. Here we assume that both bacterial and archaea are primarily responsible for the microbial processes (i.e. nitrification and denitrification) associated with Equation (3).

(4) Regarding results, I am truly surprising respect to the separation of data between pre or post storm, why not the authors previously present these dynamics as a background. But the existence of pre and post storm dynamics means that there was another temporal scale of variation, which overlaps with some seasonal and annual variability, etc. You should include an analysis of this perturbation each summer (July). In this regard, I could not understand how simulation of  $\alpha$  (pre and post storm) was performed. Table 2 present estimated N<sub>2</sub>O data; given the high standard deviation of the data, I see that no significant differences exist in N<sub>2</sub>O levels among years, with a mean range from 3.7 to 13.5 and a SD range from 4.3 to 12.2. Another point, during July 1998 a value of 3,7 ±12.2 was reproduced, it means that negative value could be estimated, I ask myself what is the sensitivity of this model?. Relative a N<sub>2</sub>O production/consumption (Figure 4), I realize that there is not a trend in N<sub>2</sub>O produced by nitrification, but if nitrification is main N<sub>2</sub>O producing process, How the authors justify found correlation between estimated N<sub>2</sub>O levels and areal hypoxia. I found the estimated N<sub>2</sub>O levels extremely low for an eutrophic area where ammonium levels should be high (close to sediment-water interface). If you have in mind that  $N_2O$  values in bottom water as high as 533 nM were found in the western continental shelf of India (Naqvi et al, 2000), 7.7 nM seems to be low. Finally, the authors omitted information about nutrient and the influence of river in the coastal area, such information can contribute to the discussion of this ms.

→ I. Walker et al. (2010) measured N<sub>2</sub>O in the waters of the nGOM in August 2008. They discussed pre-storm N<sub>2</sub>O production vs. post-storm N<sub>2</sub>O production. They reported enhanced N<sub>2</sub>O production after storm, resulting in reoxygenation from the vertical redistribution of organic matters in the water column. They estimated  $\alpha$  value under both pre-storm (0.048) and post-storm (0.096) conditions from the relationship between  $\Delta$ N<sub>2</sub>O and AOU based on the direct measurements (see their Fig. 4). As stated in our response **C**, the data sets used for the present analysis were not influenced by storm/hurricane events, which enhanced N<sub>2</sub>O production only for very short periods. Therefore, we adopted  $\alpha_{=0.048}$  as a representative summertime nGOM coefficient for production N<sub>2</sub>O by nitrification. We then checked  $\alpha$  sensitivity using a Monte Carlo procedure (Details are described in our response **e2**).

Revised Table 2 (shown below) and Fig. A2 show interannual variations: magnitudes of which are comparable to the measured values. The range of mean estimated N<sub>2</sub>O concentrations was 11.0-20.6 nmol  $L^{-1}$ , and that of standard deviations was 3.5-11.3 nmol  $L^{-1}$ . Maximum values ranged from 21.3 to 54.9 nmol  $L^{-1}$  (Table 2).

Our data indicated that  $N_2O$  equilibrium (Fig. A8) remained constant in time relative to the other  $N_2O$  components in the nGOM. Variation in  $N_2O$  production by nitrification was also small in the bottom waters. On the other hand, linear regression analysis showed that  $N_2O$  production by nitrification was significantly correlated with hypoxic area (Fig. A9), which has changed from year to year.



Fig. A9 Correlation between N<sub>2</sub>O production by nitrification and hypoxic area.

We will include in the revised manuscript (Section 3.3) the information concerning the total freshwater discharge from the Mississippi and Atachafalaya Rivers accumulated for January-July and the magnitude of remineralization (instead of nutrients) estimated by Kim and Min (2013).

Table 2. Mean bottom N<sub>2</sub>O concentrations (nmol L<sup>-1</sup>) during the study period estimated from the equation (4) using  $\alpha_{=0.048}$ ,  $\beta_{=0.83}$ , and  $\gamma_{=0.83}$ . Mean is calculated as  $[N_2O]_{est}^{mean} = \frac{\sum_{i=1}^{n} [N_2O]_{est}^i}{n}$ , where *i* is station, and *n* is the number of total stations. Standard deviations (±) is calculated as

 $\sqrt{\frac{\sum_{i=1}^{n} \left( \left[ N_2 O \right]_{est}^{i} - \left[ N_2 O \right]_{est}^{mean} \right)^2}{n}}.$ 

Year	$[N_2 O]_{est}^{mean}$	maximum	remark	
(for July)				
1985	13.0±7.6	35.9		
1986	16.7±10.0	52.4		
1987	13.6±4.9	32.9		
1988	ND	ND		
1989	ND	ND		
1990	ND	ND		
1991	13.7±5.6	38.4		
1992	14.3±7.0	42.7		
1993	15.4±6.1	31.7		
1994	14.7±4.4	30.8	Visser (2009)	
1995	13.8±4.5	32.0	Sept. 2007: 7.59-11.87 (mean: 9.95±1.07)	
1996	13.5±3.9	22.5	April 2008: 6.53-9.54 (mean: 7.10±0.70)	
1997	14.0±3.5	21.3	July 2008: 4.25-30.02 (mean: 11.00±6.95)	
1998	11.4±10.7	31.8		
1999	16.6±9.7	51.5		
2000	11.0±4.5	29.1	Walker et al. (2010)	
2001	15.0±6.6	34.4	August 2008: 5-30 (prestorm)	
2002	20.6±11.3	54.9	7-47 (poststorm)	
2003	12.7±4.7	27.6		
2004	17.9±9.9	51.9		
2005	12.4±5.7	23.3		
2006	13.9±7.4	42.9		
2007	16.0±7.2	42.6		

ND: not determined

- References -

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