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Interannual variation in summer N₂O concentration in the hypoxic region of the northern Gulf of Mexico, 1985–2007

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Simulating summer N₂O concentration in the nGOM

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Abstract

We present evidence of temporal variation in nitrous oxide (N$_2$O) concentrations in the bottom waters of the northern Gulf of Mexico (nGOM) hypoxic zone. The analysis is based on a conceptual model simulating N$_2$O biogeochemical processes in conjunction with water-column O$_2$ levels, derived from summer Texas–Louisiana shelf-wide hydrographic data for twenty Julys between 1985 and 2007. The mean modeled nGOM N$_2$O concentration was 7.7 ± 6.7 nmol L^{-1}, and was significantly correlated with the areal extent of hypoxia. Our modeling analysis indicates that the nGOM is a persistent summer source of N$_2$O, and nitrification is a primary factor leading to its production in this region. Based on the ongoing increase in the areal extent of hypoxia in the nGOM, we conclude that N$_2$O emission from this environmentally stressed region will continue to increase into the future contributing to the global increase in greenhouse gases.

1 Introduction

Nitrous oxide (N$_2$O) has a lifetime of ~ 120 yr in the atmosphere, where it has two major effects: it contributes to greenhouse effects in the troposphere, and destroys ozone in the stratosphere (IPCC, 2007; Ravishankara et al., 2009). The atmospheric N$_2$O concentration has increased rapidly since the 18th century, primarily because of anthropogenic activities (IPCC, 2007; Machida et al., 1995). The oceans are a major source of atmospheric N$_2$O, with oceanic emissions of N$_2$O accounting for approximately 20 % (4–7 Tg N yr^{-1}) of the total annual emissions (16–34 Tg N yr^{-1}) (Nevison et al., 1995; Seitzinger et al., 2000; Bange, 2006). Among identified oceanic sources, coastal oceans account for up to 60 % of the total oceanic N$_2$O emissions (Bange et al., 1996). The production of N$_2$O in coastal oceans is projected to increase worldwide (Bange, 2000; Naqvi et al., 2010), in proportion to the extent and intensity of eutrophication and hypoxia (Diaz and Rosenberg, 2008; Rabalais et al., 2009). The amount of N$_2$O produced during nitrification and denitrification (Fig. 1), which are the major

production processes of N$_2$O in the ocean, strongly depends on the prevailing oxygen (O$_2$) concentration. N$_2$O production is significantly enhanced under low (i.e. hypoxic or suboxic) O$_2$ concentrations (Codispoti, 2010).

The northern Gulf of Mexico (nGOM) is widely considered to be a "dead zone" because of extreme eutrophication arising from nutrient loads that enter from adjacent rivers, including the Atchafalaya and Mississippi (Malakoff, 1998; Justić et al., 2003; Turner and Rabalais, 2004). As a result, the area affected by hypoxia, (defined here as an O$_2$ concentration $\leq 2\,\mathrm{mg\,L^{-1}}$) increased to $\sim 20\,000\,\mathrm{km^2}$ during the past two decades (Rabalais et al., 2002) and has likely been emitting increasing quantities of N$_2$O to the atmosphere. The limited measurements of dissolved N$_2$O concentrations made in the nGOM hypoxic zone (available only for April and July–September 2008) show the area to be a significant source of atmospheric N$_2$O (Visser, 2009; Walker et al., 2010). However, it is not known how nGOM N$_2$O emissions have evolved. More importantly, cause and effect associations remain unclear between variations in the strength of N$_2$O emissions and the degree of hypoxia in the nGOM. Addressing these unknowns will help to establish a modeling framework for the prediction of the evolving release of N$_2$O from the nGOM as well as other coastal areas throughout the globe. Such prediction is important because in the future these regions are expected to become more vulnerable to hypoxia as eutrophication continues to increase with population and development.

Using a simple conceptual model framed in terms of measured seawater O$_2$ levels, and long-term summer hydrographic data collected from the nGOM, we simulate the evolution of seawater N$_2$O concentrations in Texas–Louisiana shelf waters for the month of July from 1985 to 2007. The model used in the present study incorporates all biogeochemical processes known to be involved in the production and consumption of N$_2$O in seawater.

2 Methods

2.1 Study area and data

The continental shelf area of the nGOM broadly extends seaward, and is shallower than 100 m depth (Fig. 2). The characteristics of nGOM seawater are primarily determined by mixing of Gulf of Mexico saltwater with freshwater discharged from the Atchafalaya and Mississippi Rivers (Fig. 2). In our analysis, we used observations (temperature, salinity, dissolved O_2, nitrite + nitrate, phosphate and silicate) from summer Texas–Louisiana shelf-wide surveys (approximately 80 stations were occupied during each of the surveys). These July data for the period 1985–2007 (excluding the three years between 1988 and 1990) are available online: http://www.nodc.noaa.gov and http://www.aoml.noaa.gov/ocd/necop/. The website also provides information concerning the hydrographic cruise and the analytical methods used for nutrient analysis. Nutrients were measured at only two depths (near-surface and near-bottom). Our calculation of N_2O concentrations is based on the water samples collected near the shelf bottom.

2.2 A conceptual biogeochemical model: for simulating N_2O production and consumption

In the ocean environment, N_2O is produced during nitrification (α), and is produced (β) and consumed (γ) during denitrification (Bange et al., 2010). Specific microbes are involved in the production and consumption of N_2O, and their activities are sensitive to the seawater O_2 concentration (Bianchi et al., 2012). Therefore, our conceptual model, which simulates the production and consumption of N_2O, is based on seawater O_2 concentrations (Fig. 1), and is formulated as follows:

$$[N_2O]_{est} = \alpha \left[\frac{\Delta N_2O}{\Delta O_2} \right] \times AOU + \beta \left[\frac{\Delta N_2O}{\Delta N_{deni}} \right] \times \Delta N_{deni} - \gamma \left[\frac{\Delta N_2O}{\Delta N_{deni}} \right] \times \Delta N_{deni} \qquad (1)$$

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where ΔN_2O is the difference between the measured N_2O concentration and the equilibration value. AOU is the apparent oxygen utilization (the difference between the measured O_2 concentration and the equilibration value), and ΔN_{deni} is the loss of nitrate (NO_3^-) as a consequence of denitrification. The coefficient α indicates the relationship between ΔN_2O and AOU, and the coefficients β and γ indicate the relationships between ΔN_2O and the rate of denitrification. The values of these coefficients applicable to the nGOM are empirically determined to be 0.048, 0.7, and 0.7 for α, β, and γ, respectively. The rationale for choosing these values is described below.

We use Eq. 1 with the observations to estimate the N_2O concentration in the near-bottom waters of the nGOM (approximately 1 m above the sediment-water interface) based on the O_2 concentration, and characterized the condition as: (1) oxic ($O_2 > 2\,\mathrm{mg\,L^{-1}}$), (2) hypoxic-suboxic ($0.13 < O_2 \leq 2\,\mathrm{mg\,L^{-1}}$), and (3) suboxic-anoxic ($O_2 \leq 0.13\,\mathrm{mg\,L^{-1}}$) (Naqvi et al., 2010). Under oxic conditions, N_2O 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). Under hypoxic-suboxic conditions, during which nitrification and denitrification are both involved in N_2O production (Naqvi et al., 1998; Nevison et al., 2003), the N_2O concentration is calculated as $\alpha \times$ AOU $+\beta \times \Delta N_{deni}$. Under suboxic-anoxic conditions, nitrification produces N_2O via nitrite reduction (nitrifier denitrification: $NH_4^+ \rightarrow NO_2^- \rightarrow N_2O$) (Poth and Focht, 1985; Wrage et al., 2001), but the competing process (denitrification) consumes N_2O (Cohen and Gordon, 1978; Elkins et al., 1978; Yamagishi et al., 2007). As a result, the N_2O concentration is calculated as $\alpha \times$ AOU $- \gamma \times \Delta N_{deni}$, where ΔN_{deni} was estimated using extended optimum multi-parameter (eOMP) analysis. The basic structure for the eOMP analysis is given as:

$$\mathbf{A} \cdot x - d = R, \tag{2}$$

where: the matrix $\mathbf{A}$ is composed of the physicochemical characteristics of end-members that participate in physical mixing in the study area, and the Redfield ratios that represent biogeochemical processes; the vector x consists of unknowns including

mixing ratios among different pre-defined water masses, the amount of remineralized phosphate, and denitrification (N_{deni}); the vector d contains the observed values, and the vector R represents the constraint residuals. Further details can be found in Kim (2012). Here, we used the results of denitrification (N_{deni}) estimated from the eOMP analysis by Kim (2012) for this study.

The procedures for estimating N_2O concentrations in the nGOM are schematically presented in Fig. S1. A contribution of N_2O released from the underlying sediments is expected to be negligible (Visser, 2009) and was, therefore, not accounted for in our model.

3 Results and discussion

3.1 Determination of the coefficients α, β and γ in the conceptual model

The coefficient α in Eq. (1) is known to vary as a result of intrusions of foreign water masses into the nGOM, changes in the rate of nitrification, and variations in the chemical composition of organic matter produced in situ (Cohen and Gordon, 1978; Elkins, 1978; Nevison et al., 2003). In our analysis, α is derived using ΔN_2O and AOU data for both pre-storm and post-storm periods observed during August 2008 according to Walker et al. (2010). The values of α applicable to the nGOM were determined to be 0.048 (pre-storm) and 0.096 (post-storm). Implicit in chosen value of α is the assumption that α only reflects changes in ΔN_2O and AOU associated with biological processes (i.e. nitrification). However, physical processes in the marine environment can also affect α by modifying the solubilities of N_2O and O_2.

We therefore explored the possibility that the α difference between pre- and post-storm periods (0.048) could have been a consequence of changes in the physical properties of the water that occurred during the transition from pre-storm to post-storm period. During this transition, changes in the properties (temperature, salinity, and O_2 concentration) of near-bottom water masses in the nGOM had a negligible influence

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on the solubility of N$_2$O but considerable, disproportional influence on the solubility of O$_2$, thereby increasing the value of α (Table 1). Our sensitivity test of the variability of α associated with changes in water mass properties indicates that the value of α for the post-storm period ($\alpha_{\text{post-storm}}$) could be as much as a factor of ~ 1.7 higher

5 than the value for the pre-storm period ($\alpha_{\text{pre-storm}}$), close to the measured post-storm α (0.096) (Fig. 3). It also indicates that the use of the pre-storm $\alpha_{\text{pre-storm}} = 0.048$ is probably a more suitable value for the nGOM. One line of evidence supporting the use of this value is that all the data used were related to the summer months, during which there were no or few hurricanes or storm events (Table S1). We therefore

10 use $\alpha_{\text{pre-storm}} = 0.048$ to simulate the summer N$_2$O concentration in the nGOM, and assume that the resulting value is valid for all summer periods included in this study. It will not be possible to evaluate the validity of this assumption until multi-year measurements are available for the nGOM and other coastal areas. Such measurements made in the future will enable more accurate calculation of α. It is assumed that it is

15 reasonable to set $\alpha = 0.096$ as an upper limit, because the factors known to influence α in the nGOM are probably at their most extremes immediately following the storm event.

We assign a value 0.7 to the coefficient β for the nGOM. This value was derived from an incubation experiment in the eastern tropical South Pacific (ETSP) (see Fig. 4 of

20 Farías et al., 2009). We adopt this estimate in our modeling because (i) no comparable data from the nGOM are available and (ii) the nitrogen cycle in oxygen minimum zone of the ETSP is comparable to that in the nGOM hypoxic zone. During the denitrification process N$_2$O is also consumed. The amount consumed is quantitatively related to the O$_2$ level. Farías et al. (2009) varied the ratio of N$_2$O production vs. consumption from

25 0.36 to 6.55 according to oxygen levels in the ETSP (see Fig. 5 of Farías et al., 2009). It was determined that the ratio under suboxic-anoxic conditions is 1 on average. Here, we assign $\gamma \approx \beta$ for the purpose of our modeling exercise. The choice of values for the coefficients β and γ is less critical than the choice of α, because N$_2$O production

associated with β and γ typically represents less than $\sim 25\%$ of the total production (Fig. 4).

3.2 Mechanisms of N$_2$O production and consumption in the nGOM

For the period 1985–2007, the July mean N$_2$O concentration in near-bottom waters of the nGOM is estimated to be 7.7 nmol L^{-1} (min: 3.7 nmol L^{-1}, max: 13.5 nmol L^{-1}), with considerable associated interannual variability ($\sigma = \pm 6.7$ nmol L^{-1}, where σ is the standard deviation) (Fig. 5 and Table 2). This estimate appears reasonable as measured bottom N$_2$O concentrations in July–August 2008 in the nGOM typically ranged from ~ 4 to 15 nmol L^{-1} (Visser, 2009; Walker et al., 2010), although higher concentrations of up to 30 nmol L^{-1} were also detected. Our estimated nGOM N$_2$O concentrations are comparable to those observed in other hypoxic-anoxic areas, including Chesapeake Bay, and the Baltic and Black Seas (6–20 nmol L^{-1}) (Naqvi et al., 2010).

The N$_2$O concentrations for July 1996 and 1998 (4.5 and 3.7 nmol L^{-1}, respectively) were lower during the study periods, but the areal extents of hypoxia were relatively high ($> 12\,000$ km^2) (Fig. 5). It was because the estimated N$_2$O concentrations were somewhat influenced by N$_2$O consumption For July 1996 and 1998 (Fig. 4). July 2000 N$_2$O (~ 5.1 nmol L^{-1}) was also relatively low compared to other years, however in contrast to 1996 and 1998, the areal extent of hypoxia was smaller (~ 4400 km^2) (Figs. 5 and 6). The following scenario may explain the lower N$_2$O concentration and smaller areal extent of hypoxia found in July 2000. The total freshwater discharge from the Mississippi and Atchafalaya Rivers in the period January–July 2000 was less than in the same period in other years, resulting in decreased production of organic matter through biological productivity (Walker and Rabalais, 2006). Consequently, the oxidation of organic matter was probably decreased, and the net effect was a reduction in the areal extent of hypoxia in July 2000. Another contrasting example is evident for the month of July 2002. The mean N$_2$O concentration was highest (~ 13.1 nmol L^{-1}) and the size of the hypoxic zone was largest ($\sim 22\,000$ km^2) (Fig. 5). The production

of N_2O during denitrification was also highest in July 2002, and its contribution to the overall N_2O production was significant (Fig. 4). These conditions probably resulted in July 2002 having the highest N_2O concentration (Fig. 6). Overall, interannual variation in the estimated N_2O concentrations is significantly correlated with the areal extent of hypoxia ($R = 0.52$; $p < 0.05$; Fig. 5).

To identify the primary mechanism(s) of N_2O production in the nGOM, we estimate the contribution to N_2O concentration of each mechanism: production by nitrification and denitrification, and consumption by denitrification (Fig. 4). Our result indicates that $\sim 75\%$ of the total N_2O is produced under oxic and hypoxic-suboxic conditions during nitrification, and the remainder ($\sim 20\%$) is produced under hypoxic-suboxic conditions during denitrification. The result also shows that N_2O is consumed exclusively under suboxic-anoxic conditions during denitrification, but this makes only a small contribution to the N_2O concentration (approximately 5% of total N_2O production). Our study indicates that the nitrification is the major process responsible for the production of N_2O in the nGOM.

Based on the significance of the correlation between the estimated N_2O concentrations and the areal extent of hypoxia (Fig. 5), it is expected that the strength of the nGOM as a source of N_2O will increase into the future, with the expansion in the hypoxic region (Bange, 2000; Naqvi et al., 2000; Codispoti, 2010).

4 Summary

The implications of our modeled results are profound. The nGOM is receiving ever-increasing loads of nutrients through rivers from anthropogenic activities, and this has led to more intense and widespread hypoxic conditions (Rabalais et al., 2009; Bianchi et al., 2010). As the areal extent of nGOM hypoxia continues to expand, it is expected that N_2O emissions will be enhanced. This study area is particularly vulnerable to human-induced catastrophic events. A prime example is the recent "Deep-Water Horizon" oil spill, which may greatly increase the areal extent of hypoxia in the nGOM.

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Table 1. Mean surface and bottom temperature, salinity, DO, AOU and ΔN$_2$O values for pre-storm and post-storm periods based on Walker et al. (2010) used to simulate α variability due to changes in physical environment. Since the pre-storm and post-storm N$_2$O differences are small within the standard deviations (see their Fig. S1), we used the fixed values of mean surface and bottom N$_2$O concentrations for the simulation.

condition	T (°C)	S (psu)	DO (μmol L^{-1})	AOU (μmol L^{-1})[a]	ΔN$_2$O (nmol L^{-1})[b]
N$_2$O = 8 nmol L^{-1} at surface					
pre-storm	29	23	200	11.7	1.9
post-storm	29	30	190	13.6	2.1
N$_2$O = 16 nmol L^{-1} at bottom					
pre-storm	24	36	40	171.7	9.4
post-storm	26	33	100	110.4	9.7

[a] AOU = [O$_2$]$_{saturation (T, S)}$ − [O$_2$]$_{observed}$.
[b] ΔN$_2$O = [N$_2$O]$_{observed}$ − [N$_2$O]$_{saturation (T, S)}$.

Table 2. N_2O concentrations (nmol L^{-1}) during the study period estimated from the Eq. (1) using $\alpha_{=0.048}$, $\beta_{=0.7}$, and $\gamma_{=0.7}$. Standard deviations (±) indicate spatial variability within the study area.

Year (for July)	$[N_2O]_{est}$
1985	6.5 ± 8.0
1986	10.1 ± 8.7
1987	7.3 ± 4.5
1988	ND
1989	ND
1990	ND
1991	7.5 ± 4.9
1992	7.6 ± 6.2
1993	8.5 ± 6.2
1994	7.9 ± 4.5
1995	6.6 ± 6.1
1996	4.5 ± 6.4
1997	6.9 ± 4.8
1998	3.7 ± 12.2
1999	10.0 ± 8.4
2000	5.1 ± 4.3
2001	8.4 ± 6.4
2002	13.5 ± 9.8
2003	6.2 ± 4.3
2004	11.0 ± 8.7
2005	6.1 ± 5.8
2006	7.8 ± 7.1
2007	9.5 ± 6.3

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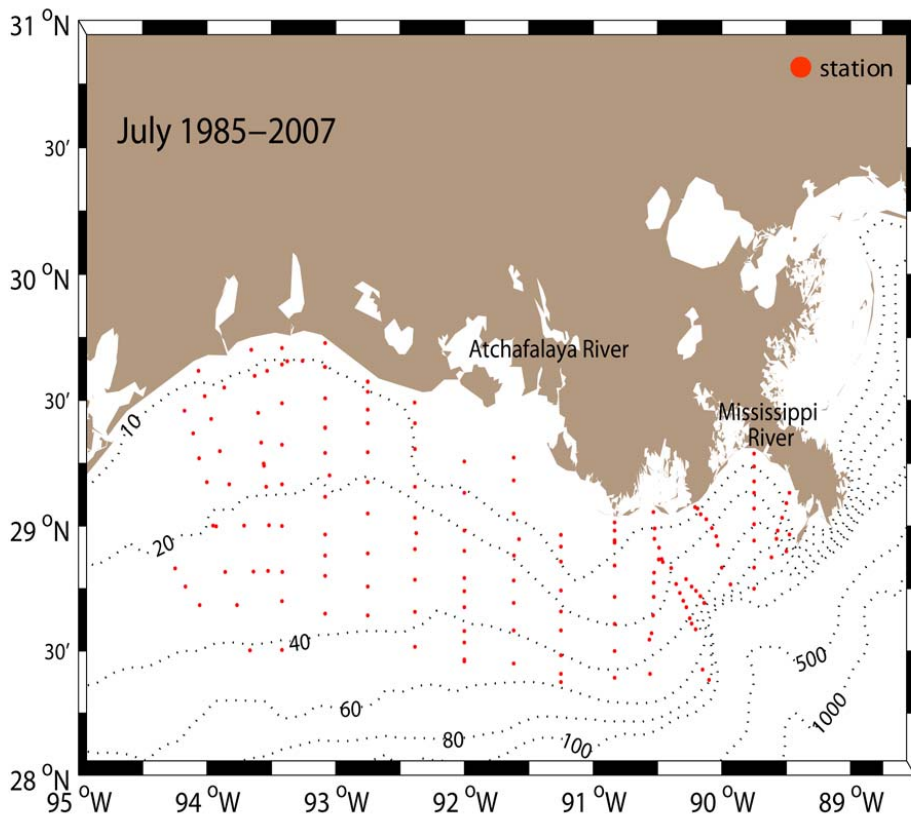Tables | Figures

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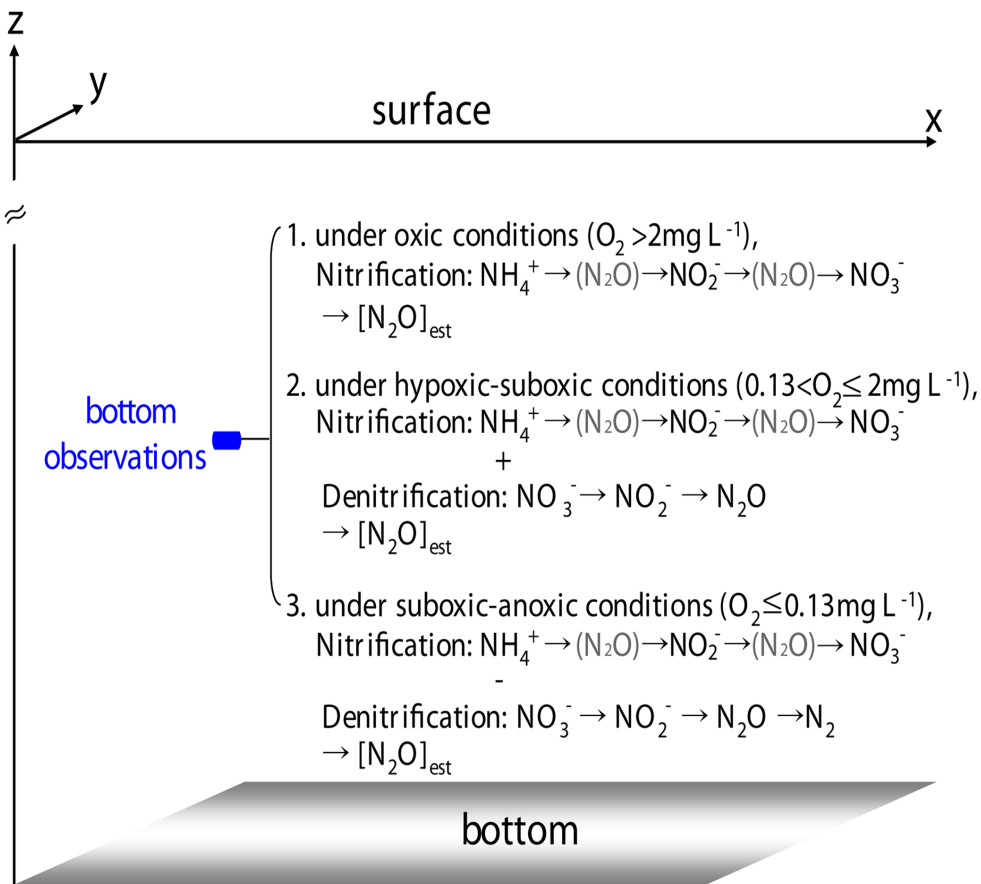
Fig. 1. A conceptual model of the processes affecting production and consumption of N_2O in near-bottom waters of the study area.

Fig. 2. The study area map showing the summer Texas–Louisiana shelf-wide cruise stations (red dots) during July 1985–2007 with bathymetry contours (black dotted-lines) in the northern Gulf of Mexico.
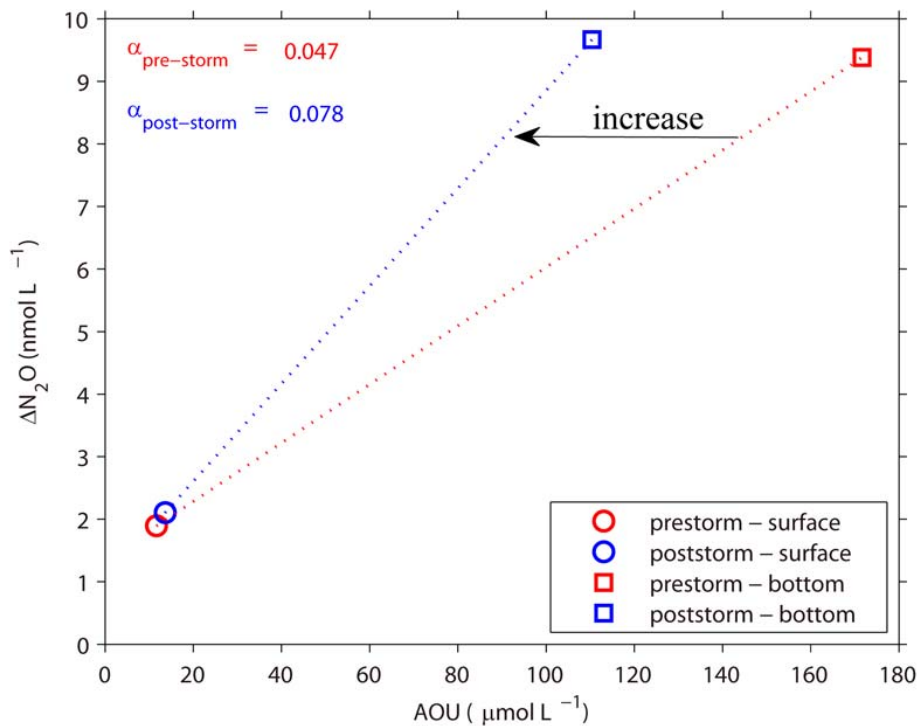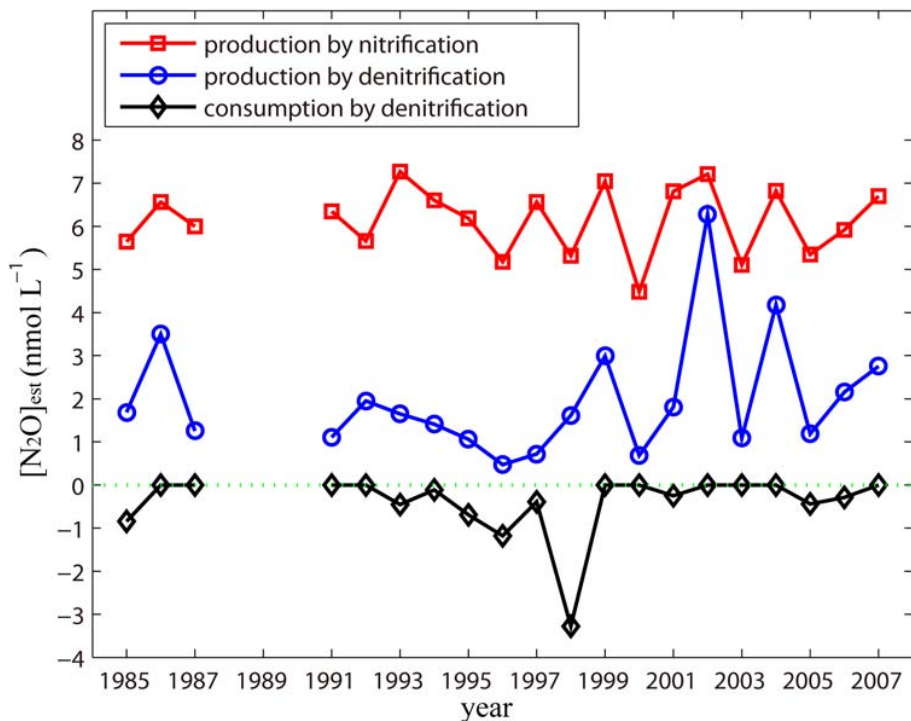
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Fig. 3. A simple simulation of α (= ΔN_2O/AOU) variability using the pre-and post-storm information found in Table 1.
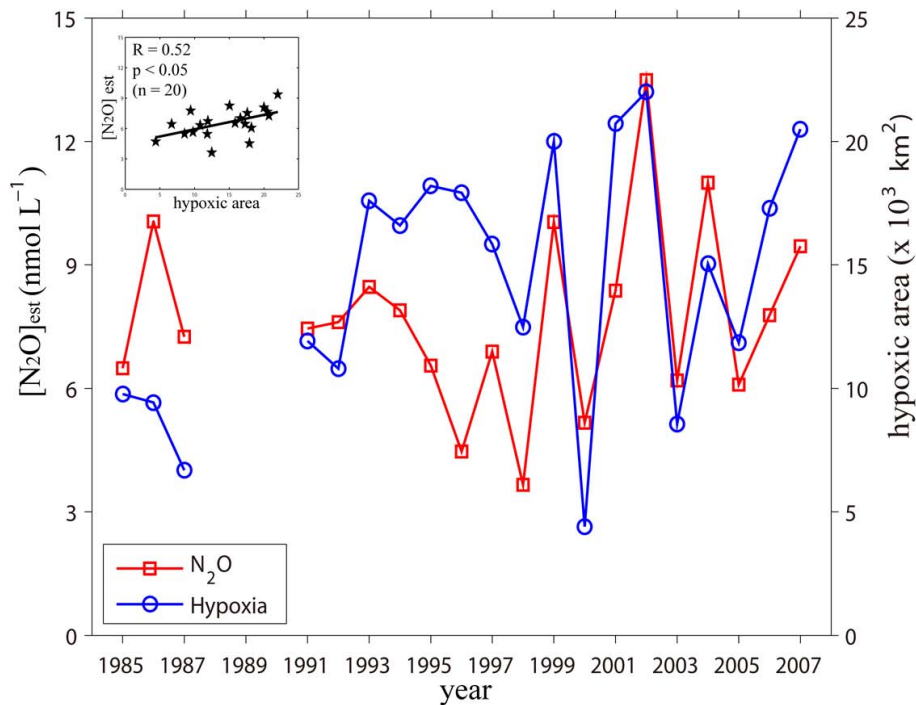
Fig. 4. The individual contributions of nitrification (red squares, production), denitrification (blue circles, production), and denitrification (black diamonds, consumption) to the net N$_2$O concentrations in the nGOM from July 1985 to 2007.

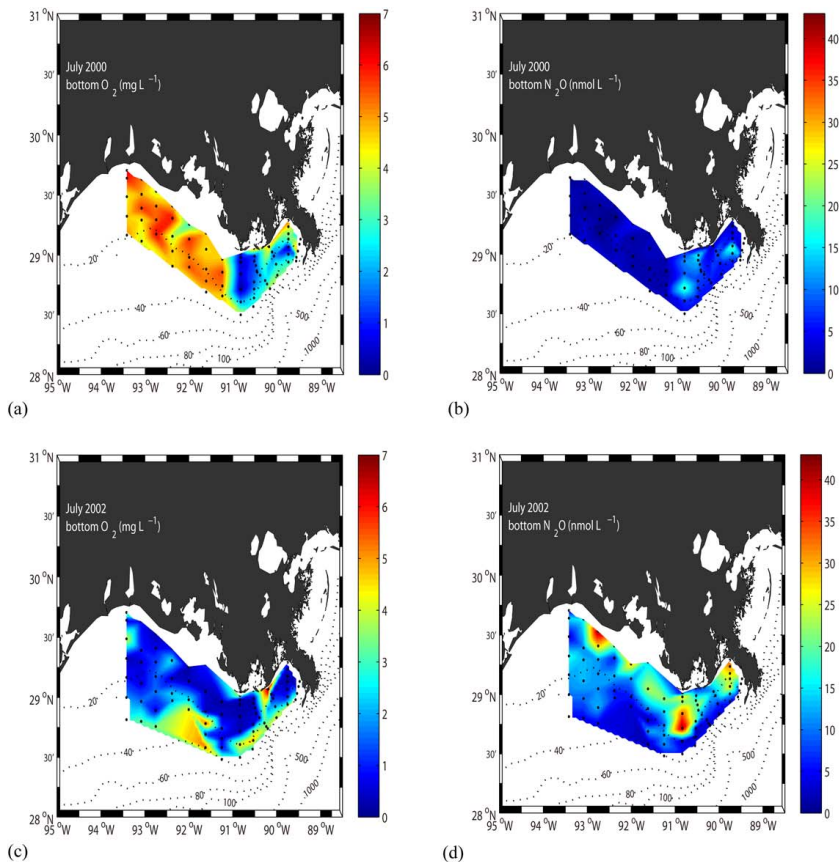**Fig. 5.** Interannual variations in summer (July) N_2O concentration (nmol L^{-1}, red squares) and the areal extent of hypoxia (km^2, blue circles). The estimated N_2O concentrations were significantly correlated with the areal extent of hypoxia ($R = 0.5$; $p < 0.05$ with $n = 20$). Data concerning the areal extent of hypoxia is available at http://www.gulfhypoxia.net/Research/ Shelfwide%20Cruises/ (Data source: N.N. Rabalais, Louisiana Universities Marine Consortium, R.E. Turner, Louisiana State University).

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Fig. 6. Distributions of bottom O_2 and estimated N_2O concentrations in July 2000 and 2002, which are min and max hypoxic area during the study periods, respectively. **(a)** and **(b)** bottom O_2 and N_2O spatial distributions in July 2000, **(c)** and **(d)** bottom O_2 and N_2O spatial distributions in July 2002.