Dear Dr. Hoppema,

We want to thank you for your help, and the three reviewers for their valuable comments about our manuscript. They have raised important points that helped us to improve clarity and became an article on a high level.

Below is a list of our correctness based on your comments.

All revised area is changed in the final manuscript with yellow highlighted.

Topic Editor Decision: Publish subject to minor revisions (review by editor) (07 Nov 2019) by Mario Hoppema

Comments to the Author: Dear Drs. Kim, Chapman and co-authors,

Your revisions are satisfactory and the manuscript is almost ready for acceptance. I went through it and below my final comments are listed.

Abstract L1 coastal sea (no capitals)

 \rightarrow corrected

Abstr L2 nitrogen mass balance (not N here)

 \rightarrow corrected

Abstr L5 no need for (AN-D); it is not used in the Abstract anymore

 \rightarrow corrected

Abstr L6 I think "however" is not needed here.

 \rightarrow deleted

Abstr L11 "more than 2" is not very precise and looks strange here. Can you please modify this? \rightarrow we changed word "over 2 gC~"

L24 Please define SGD

 \rightarrow L24, submarine groundwater discharge (SGD)

L67 I guess you mean high winds and high nitrate concentrations

 \rightarrow L66-69, we changed the sentence more clearly.

L117 Please explain how data interpolation was done

 \rightarrow 117-118, we explained more detail into the text

L124 Please change format: kg ha-1 year-1. Is this according to SI? ha is hectare, right? Even when this is the usual notation in this field of science, please also use SI units.

 \rightarrow L125, corrected

L129 dito

 \rightarrow L130, corrected

L145 ml/L is not a SI unit. Please (also) give the value as SI.

 \rightarrow L146, corrected

L148-149 "which means that the two layers have different biological processes." I do not agree. The rates could just be different.

→ L149-150, corrected

L151-152 "While chlorophyll can be found below the pycnocline (DiMarco and Zimmerle, 2017)." This is not a complete sentence. Please correct.

 \rightarrow L152-155, corrected, "While chlorophyll can be found below the pycnocline (DiMarco and

Zimmerle, 2017), the fact that it is typically associated with low oxygen concentrations suggests that the phytoplankton are either inactive or, more likely, producing at a very slow rate."

L208 "nitrate (NO2-) and nitrite (NO3-)" This is an error. It should be exactly the other way around.

\rightarrow L211, corrected

L277 Please use format 4 March 2005

\rightarrow L279-280, corrected

L295-296 The sentence was not clear. I suggest: We defined the brown zone as having the PPP rate of over 2 gC m-2 day-1 because ...

→ L298-299, corrected

L328-329 ... determined the integrated PP rates with 14C measurements during 2004 ... (because twice in the sentence a word with "measure")

\rightarrow L331-332, corrected

L335 MR has not been defined before. Please consider not using that many abbreviations, because they reduce the readability.

\rightarrow Defined and corrected in L245

L508 suggest (instead of explained)

\rightarrow L507, corrected

L513-514 "in the near future both AN-D flux and riverine N flux need to be considered for managing nitrogen in coastal waters." This is double info and it appears in the previous paragraph and at other place in the manuscript.

\rightarrow deleted

L533-534 "We identified the brown zone close to the Keum River mouth and the green and blue zones further away from the coast of Korea." This is trivial and does not contain much information. If you would like to state something here, please give more details.

\rightarrow deleted

As to the references: L555 No journal name \rightarrow L557-563, corrected L580 No journal name \rightarrow L585-591, corrected L685,688 Any more info on these reports? Report number, Place of publication? \rightarrow L812-819, corrected L723 Journal must be: Journal of the Atmospheric Sciences \rightarrow L731-732, corrected L731 upper case for 222 \rightarrow L739, corrected L804 Any more info for this report? \rightarrow L821-823, corrected L855 Please change format of reference \rightarrow L872, corrected L893 Estuarine (typo) \rightarrow L910, corrected L929 Any more info for this report? Where available? \rightarrow L945-947, corrected

Figure 1 caption. Please use Gulf of Mexico full, not GOM. a) Please add some geographical

names in the figure for orientation. What does the drawn line represent? What about abbreviations and percentages? Where exactly is the sampling area? Is it the whole region? Please be more specific.

b) Please explain the colors

→ L962-969, corrected, "Study sites and sampling areas in the Gulf of Mexico and Korea. (a) shows the sampling area within the northern Gulf of Mexico. Flow in the Mississippi/ Atchafalaya River System is split 30% to the Atchafalaya River, 70% to the Mississippi River. The box is the sampling area. (b) shows station positions from March 2005. Note that MCH project data are widely distributed across the region. Red, grey, and blue stations correspond to sub-regions A (near the Mississippi River), B (between the Mississippi and Atchafalaya), and C (near the Atchafalaya) respectively. (c) shows the sampling area off the west coast of Korea. (d) shows all of the station positions."

L1020, 1022 delete "Figure"

 \rightarrow Delected

L1025 describes, not described

Figure 2 Please define RMEPs

 \rightarrow L973, corrected

Figure 9 Please do not use abbreviations here

 \rightarrow L1011-1012, corrected

Table 1 Please use data format like 5 April 2004

Table 4 Please use data format like 5 April 2004

L1084, 1088 There should be a minus sign in the ratio

→ Table 1, L1032-1035, 1039-1040, corrected

Thank you and best wishes Mario Hoppema

Implications of different nitrogen input sources for potential production and carbon flux estimates in the coastal Gulf of Mexico (GOM) and Korean coastal waters

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Abstract

The coastal Gulf of Mexico (GOM) and coastal sea off Korea (CSK) both suffer from human-induced eutrophication. We used a nitrogen (N) mass balance model in two different regions with different nitrogen input sources to estimate organic carbon fluxes and predict future carbon fluxes under different model scenarios. The coastal GOM receives nitrogen predominantly from the Mississippi and Atchafalaya Rivers and atmospheric nitrogen deposition is only a minor component in this region. In the CSK, groundwater and atmospheric nitrogen deposition are more important controlling factors. Our model includes the fluxes of nitrogen to the ocean from the atmosphere, groundwater, and rivers, based on observational and literature data, and identifies three zones (brown, green and blue waters) in the coastal GOM and CSK with different productivity and carbon fluxes. Based on our model results, the potential primary production rate in the inner (brown water) zone are over 2 gC m⁻² day⁻¹ (GOM) and 1.5 gC m⁻² day⁻¹ (CSK). In the middle (green water) zone, potential production is between 0.1 to 2 (GOM) and 0.3 to 1.5 gC m⁻² day⁻¹ (CSK). In the offshore (blue water) zone, productivity is less than 0.1 (GOM) and 0.3 (CSK) gC m⁻² day⁻¹. Through our model scenario results, overall oxygen demand in the GOM would increase approximately 21% if we fail to reduce riverine N input, likely increasing considerably the area affected by hypoxia. Comparing the results from the U.S. with those from Korea shows the importance of considering both riverine and atmospheric inputs of nitrogen. This has direct implications for investigating how changes in energy technologies can lead to changes in the production of various atmospheric contaminants that affect air quality, climate and the health of local populations.

Keywords:

Chemical tracers, Biological processes, Shelf-seas, Gulf of Mexico, Yellow Sea.

1 Introduction

2 Industrial expansion and anthropogenic emissions are major factors leading to increased coastal productivity and potential eutrophication (Sigman and Hain 2012). Coastal primary 3 4 production is controlled largely by nitrogen (N) and phosphorus (P), and the relative supply of 5 each determines which element limits production (Paerl 2009); freshwater inputs and the 6 distance from sources such as river mouths are also important (Dodds and Smith 2016). 7 Changes in nutrient loading from air-borne, river-borne and groundwater sources can also affect 8 which element limits coastal productivity (Sigman and Hain 2012). Most coastal regions are N-9 limited, however, at certain times conditions can change from N-limited to P-limited (Dodds and 10 Smith 2016; Howarth and Marino 2006). Sylvan et al. (2006), for example, suggested that the 11 coastal GOM, especially near the Mississippi River delta mouth, is P-limited at certain times.

12 Several studies have shown that increasing atmospheric nitrogen deposition (AN-D) is 13 contributing to ocean production globally, including to eutrophication, and is potentially of 14 future importance in the GOM (Cornell et al., 1995; Doney et al., 2007; Duce et al., 2008; He et 15 al., 2010; Kanakidou et al., 2016; Kim 2018; Kim (TW) et al., 2011; Lawrence et al., 2000; Paerl 16 et al., 2002). Recently, Kim (TW) et al. (2011), using a model simulation showed that AN-D 17 controls approximately 52% of the coastal productivity in the Yellow Sea. Global NOx 18 emissions have increased but appear to be changing differently in the US and Asia (Kim (JY) et 19 al., 2010; Luo et al., 2014; Shou et al., 2018; Zhao et al., 2015), and may affect not only coastal 20 productivity but also global total nitrogen budgets. This study uses a box model to define 21 potential carbon fluxes based on different nitrogen input sources in two different regions, the 22 Coastal Gulf of Mexico (GOM) and the Coastal Sea off Korea (CSK). The GOM and CSK 23 were selected in this study because while the major input source to the coastal ocean in both

regions is riverine, the AN-D and submarine groundwater discharge (SGD) are considerably more important in the CSK region (Wade and Sweet, 2008; Zhao et al., 2015).

Most previous model studies in the GOM have been used to predict the size of the 26 hypoxic zone (e.g., Fennel et al., 2006, 2011, 2013; Green et al., 2008; Hetland and DiMarco 27 28 2008; Justic et al., 2002; Scavia et al., 2004; Turner et al. 2006, 2008), although Bierman et al. 29 (1994), used a mass balance model to estimate carbon flux and oxygen exchange. The mass 30 balance model is a useful tool to calculate nutrient or carbon fluxes and to estimate production in 31 the coastal ocean (Kim (JS) et al, 2010; Kim (G) et al., 2011), and such models have been 32 successfully used in many regions and individual coastal systems to estimate ecosystem 33 metabolism, e.g., in the Patuxent River estuary of the Chesapeake Bay (Hagy et al. 2000; Testa 34 et al., 2008) and in the LOICZ (Land Ocean Interactions in the Coastal Zone) project (e.g., 35 Ramesh et al., 2015). However, there are few such model studies in the GOM and CSK. All previous models for the GOM and the CSK have considered only riverine N as the predominant 36 37 input source, and no one has considered AN-D as an input in either region.

38 In this study, we aimed to: 1) build a mass balance model considering not only riverine N 39 input but also air-borne and groundwater-borne N; 2) use it to calculate potential primary 40 production in the three regions defined by Rowe and Chapman (2002, henceforth RC02, see next 41 section) and their associated coastal productivity; and 3) use the mass balance model to test the 42 RC02 hypothesis. Because RC02 did not quantify their model with nutrient data and no one 43 has applied this model to another region, we tested the RC02 hypothesis using data from both the GOM and the CSK that include low salinity samples. We used historical data from the mid-44 45 western part of the CSK and evaluated the theoretical model of RC02 in both areas where 46 freshwater with high terrestrial nutrient input mixes into the coastal ocean.

47

48 Study areas

49 The Texas-Louisiana (LATEX) shelf in the northern Gulf of Mexico is affected by 50 coastal nutrient loading, leading to hypoxia, coming from two major terrestrial sources (the 51 Mississippi and Atchafalaya Rivers that together form the Mississippi-Atchafalaya River System 52 MARS). These two major rivers have different nutrient concentrations. The Gulf of Mexico 53 (GOM) is a semi-enclosed oligotrophic sea and the MARS is the major source of nutrients and 54 freshwater to the northern GOM (Alexander et al., 2008; Rabalais et al., 2002; Robertson and 55 Saad, 2014). The MARS drains 41% of the contiguous United States (Milliman and Meade, 1983) and discharges approximately 20,000 m³ s⁻¹, or about 60% of the total freshwater flow, 56 (about 10.6 x 10^{11} m³ year⁻¹ or 3.4 x 10^4 m³ s⁻¹) to the northern side of the GOM. The 57 58 remainder comes from other U.S. rivers, Mexico and Cuba (Nipper et al., 2004).

59 At the Old River Control Structure on the lower Mississippi River approximately 25% of 60 the Mississippi River's water is diverted into the Atchafalaya River, where it mixes with the 61 water in the Red River. The flow in the Atchafalaya River totals 30% of the total MARS flow 62 (Figure 1a). Several projects have investigated the relationship between nutrients and the 63 marine ecosystem, and how this leads to hypoxia in the GOM (e.g. Bianchi et al., 2010; Diaz and Rosenberg, 1995, 2008; Forrest et al., 2011; Hetland and DiMarco, 2008; Laurent et al., 2012; 64 65 Quigg et al., 2011; Rabalais and Smith, 1995; Rabalais et al., 2007; Rabalais and Turner 2001; 66 Rowe and Chapman 2002). Strong stratification due to the high freshwater discharge from the MARS, local topography (DiMarco et al., 2010), wind direction, and high nitrate concentration 67 all affect hypoxia formation, with upwelling-favorable wind facilitating its development (Feng et 68 69 al., 2012, 2014).

70 In the Northern GOM, the major factor controlling coastal productivity is riverine N input. 71 Rowe and Chapman (2002), defined three theoretical zones over the LATEX shelf close to the Mississippi and Atchafalaya River mouths to predict the effects of nutrient loading on hypoxia 72 73 along the river plumes and over the shelf. They named these the brown, green, and blue zones 74 (Figure 2). Nearest the river mouths is a 'brown' zone, where the nutrient concentrations are 75 high, but the discharge of sediment from the river reduces light penetration and limits primary 76 productivity within the plume. Further away from the river plume is a stratified 'green' zone 77 with available light and nutrients that result in high productivity. In this region, the rapid 78 depletion of nutrients is due to biological uptake processes that depend on the season and river 79 flow (Bode and Dortch, 1996; Dortch and Whitledge, 1992; Lohrenz et al., 1999; Turner and 80 Rabalais, 1994). Still further offshore, and also along the river plume to the west, there is the 81 so-called 'blue' zone, defined arbitrarily by nitrate concentrations of 1 μ M or less, which is 82 dominated by intense seasonal stratification and a strong pycnocline, so that in the surface layer 83 nutrients are limiting at this distance from the rivers and most primary production is fueled by 84 recycled nutrients (Dortch and Whitledge, 1992). It is important to note that RC02 makes clear 85 that the edges of the zones (geographical regimes) are not static, but change over time depending 86 on season, river flow, and biological processes (Figure 2).

The coastal sea off western Korea (CSK) forms the eastern side of another semi-enclosed basin (the Yellow Sea) and is affected by freshwater discharge from river plumes in the same way as the coastal GOM, although the freshwater flow is considerably less. The Yellow Sea covers about 380,000 km² area with an average water depth of 44 m, and numerous islands are located on its eastern side (Liu et al., 2003). Our specific study area is the mid-western coastal region from the Taean Peninsula to Gomso Bay (Figures 1c and 1d).

93 There is a strong tidal front in the coastal area near the Taean Peninsula due to sea floor 94 topography and the coastal configuration (Park, 2017; Park et al., 2017). The region also 95 contains several bays (Garolim Bay, Gomso Bay and Cheonsu Bay), and is affected by 96 discharges from a large artificial lake (Saemangeum lake) as well as the freshwater discharge 97 from the Keum river plume that contains high concentrations of nutrients (Lim et al., 2008). 98 Conditions in the mid-western CSK near the Taean peninsula are similar to the coastal GOM, 99 because of mixing of two different water masses from Gyunggi Bay (Han River) and the Keum 100 River (Choi et al., 1998, 1999). The annual mean flow rates within the Keum River were about 101 70 m³ s⁻¹ (normal period) and 170 m³ s⁻¹ (flood period) (Yang and Ahn 2008). Precipitation 102 within the catchment was 1,208 mm year⁻¹ during 2003 to 2005 (Yang and Ahn 2008).

Unlike the coastal GOM, the CSK has increased nitrogen inputs from atmospheric nitrogen deposition (AN-D, which is approximately five times higher than in the GOM, Table 2) (Kim (JY) et al., 2010; Luo et al., 2014; Shou et al., 2018; Zhao et al., 2015) and nutrient inputs from the groundwater discharge (Kim (JS) et al., 2010; Kim (G) et al., 2011). AN-D has increased in the CSK owing to industrial development in China during the last few decades, which has led to increased atmospheric N emission.

109

110 Data and Methods

111 Riverine N data

Hydrographic data from the MCH (Mechanisms Controlling Hypoxia – MCH Atlas) projects in the Gulf of Mexico were collected from the National Oceanographic Data Center (https://www.nodc.noaa.gov) covering the period from 2004 through 2007 (Table 1). We excluded cruises MCH M6 and M7 because the threat of hurricanes led to sampling stations in different areas from the other cruises. The study sites and sampling areas are shown in Figure
117 1b. Quality control removed inconsistencies and anomalies in the data (e.g., removing outliers,
missing data found by linear interpolation). Hydrographic data from the CSK (nutrients,
salinity, oxygen) were collected during several cruises (Table 1 and Figure 1c and 1d), and the
data were put through similar QA/QC routines.

121

122 Atmospheric Nitrogen Deposition (AN-D) data

123 AN-D data from around the US are sparse (Table 2). Most US data have been collected 124 along the east coast of the US and the only data in the GOM region were collected near Corpus Christi (~1 g m⁻² year⁻¹; Wade and Sweet, 2008), Considerable AN-D could be expected, 125 126 however, from the large number of petrochemical and fertilizer plants in southern TX, especially 127 near Houston and along the Mississippi. While there are more data from the Yellow Sea (Kim 128 (JY) et al., 2010; Luo et al., 2014; Shou et al., 2018; Zhao et al., 2015), they are still limited 129 owing to the broad sampling coverage. While AN-D data in the Asian region were up to 14 g 130 m⁻² year⁻¹, data from the eastern side of the US were under 1 g m⁻² year⁻¹, even lower than in the 131 GOM, suggesting there is currently not a large contribution from AN-D to total N loads to the 132 North Atlantic Ocean. The approximate order of magnitude difference in AN-D concentrations 133 between the GOM and the CSK is due to the continuing industrial development in East Asia and 134 the resulting N emissions (Wang et al., 2016; Zhao et al., 2015). Lamarque et al., (2013) 135 reported model results, which covers our study regions, and their model appears to underestimate 136 AN-D at the sampling sites compared with observational data in the GOM (Wade and Sweet, 137 2008). However, the pattern of AN-D inputs between GOM and CSK from Lamarque et al.,

138 (2013) shows around five times difference between the two regions, which agrees with our data.

139 Thus, in our model, we used observational data for both regions, as shown in Table 2.

140

141 *Methodology: N-mass balance model*

142 Our model consists of three sub-regions based on sampling locations during MCH cruises 143 (Figure 3), each of which contains a series of one-quarter degree square boxes, as followed by 144 Belabbassi (2006). The quarter degree boxes in this study were separated into an upper box and 145 a lower box, based on pycnocline depth, as defined by a sharp change in density and coincides 146 generally with a minimum change in oxygen concentration of 22.33 μ M. We assume steady 147 state conditions, and estimate potential production, which we count as an estimate of potential 148 carbon flux (Figure 3a). Primary production (PP) above the pycnocline is expected to be higher 149 than below it (Anderson 1969; Sigman and Hain, 2012), which means that the two layers have 150 different production rates. The difference in PP between upper and lower boxes also depends 151 on the freshwater discharge rate, which determines nutrient input to the upper layer, seasonal 152 variability, and transfer processes between the layers. While chlorophyll can be found below the pycnocline (DiMarco and Zimmerle, 2017), the fact that it is typically associated with low 153 oxygen concentrations suggests that the phytoplankton are either inactive or, more likely, 154 155 producing at a very slow rate.

The N-mass balance box model is modified from previous models to calculate the net removal of dissolved inorganic nitrogen (DIN) inside each box, which represents potential primary production (PPP) (De Boer A.M. et al., 2010; Kim (G) et al., 2011) (Equation 1). In this model, DIN concentration includes ammonium (NH_4^+), nitrate (NO_3^-), and nitrite (NO_2^-).

161
$$F_{River}^{DIN} + F_{Atmo}^{DIN} + F_{Bott}^{DIN} - F_{Export}^{DIN} - F_{Deni}^{DIN} = F_{Removal}^{DIN} - \text{Eq. 1}$$

162

where, F_{River}^{DIN} , an input term, is DIN flux from each river discharge and calculated with C_{Box}^{DIN} , 163 the DIN concentration in each box, A_{Bott} , the bottom area of each quarter degree box, and 164 F_{River} , river discharge rate $(C_{Box}^{DIN} \times A_{Bott} \times F_{River})$. As another input term, F_{Atmo}^{DIN} is the 165 flux from atmospheric nitrogen deposition. F_{Bott}^{DIN} , the benthic flux is additional input term in 166 167 the sub-pycnocline layer box. The one quarter degree blue boxes located closest to the 168 Mississippi and Atchafalaya river mouths were assumed to be the only ones affected by riverine input (Figure 3b). As an output term, F_{Export}^{DIN} as an advection term was calculated from the 169 170 current velocity in each region from observations (Nowlin et al., 1998a, b) and from literature 171 data (Jacob et al., 2000; Lim et al., 2008) and the exchange between boxes from the residence 172 time in each box. Note that water and nutrient exchange can take place through all four sides of each box, so the array is two-dimensional. F_{Export}^{DIN} for water mixing was calculated from these 173 factors; C_{EX}^{DIN} is the difference in DIN concentration between adjacent boxes, V_S is the water 174 volume of each box, and λ_{Mix} is the mixing rate of each box $(C_{EX}^{DIN} \times V_S \times \lambda_{Mix})$. We used 175 176 a reciprocal of the water residence time that we considered to represent horizontal mixing, i.e. dispersion. Another output term is F_{Deni}^{DIN} , denitrification process from the water column, and 177 $F_{Removal}^{DIN}$ is removal by biological production. The details of the model definitions are given 178 179 below in Table 3 and shown in Figure 3. Each arrow indicates input (blue) and output (red) 180 terms (Figure 3). Input/output terms vary based on whether the boxes are above/below the 181 pycnocline, while there are separate inputs from the Mississippi and Atchafalaya rivers in the 182 GOM and Keum and Han rivers in the CSK, respectively.

In order to calculate the net removal of DIN in a box above the pycnocline layer, we usedour N-mass balance model in Equation 2.

185

186
$$F_{River}^{DIN} + F_{Atmo}^{DIN} - F_{Export}^{DIN} - F_{Sink}^{DIN} = F_{Removal}^{DIN} - \text{Eq. 2}$$

187

The boxes above the pycnocline layer have two input terms: 1) F_{River}^{DIN} , riverine N, 188 which affects only a subset of boxes along the edge of each region, and 2) F_{Atmo}^{DIN} , atmospheric 189 190 nitrogen deposition (AN-D), which affects every box equally. The mean value of Asian data, as 191 shown in Table 2 (Kim (JY) et al., 2010; Luo et al., 2014; Shou et al., 2018; Zhao et al., 2015), is used for F_{Atmo}^{DIN} of the CSK region, which is initially five times higher than that of the GOM (1.4 192 193 X 10⁵ mol day⁻¹; Wade and Sweet, 2008). We also considered vertical sinking as an input for the sub-pycnocline layer box and as an output from the upper layer. Other possible input 194 195 factors might be upwelling/downwelling processes; however, these factors are neglected in the 196 model because both regions are shallow and close inshore (Feng et al., 2014; Lim et al., 2008) 197 and we have no observational data on upwelling/downwelling rates. The output terms are the following: 1) F_{Export}^{DIN} , the exchange rate between each box (obtained from the different N 198 concentrations in each box and the mass transfer between them), and 2) F_{Sink}^{DIN} , removal by 199 200 biological production, including sinking (assuming that any other removal factors are neglected 201 above the pycnocline). We tested the RC02 three zone hypothesis in the upper box laver, in 202 which we can also examine the horizontal influence (horizontal extent) of the river plume based on production rates. 203

Below the pycnocline layer we used the revised Equation 3.

$$F_{Bott}^{DIN} + F_{Sink}^{DIN} - F_{Export}^{DIN} - F_{Deni}^{DIN} = F_{Removal}^{DIN} - Eq. 3$$

207

Equation 3 has two separate input terms; 1) The benthic flux F_{Bott}^{DIN} term contains all the 208 209 potential input from the bottom sediment (defined here as net DIN release from the bottom 210 sediment) including nutrient regeneration by bacteria, groundwater nutrient inputs, and an uptake 211 of nitrate (NO₃) and nitrite (NO₂) mainly by sedimentary denitrification (McCarthy et al., 2015; Nunnally et al., 2014), and 2) F_{Sink}^{DIN} term as a vertical sinking from the box above the 212 pycnocline layer, for which we used data from Qureshi (1995). The unit of F_{Sink}^{DIN} was 213 converted to mol day-1 from the unit of original data (gN m⁻² day-1) with area of box (0.25 m x 214 215 0.25 m) and molar mass of N (14 g mol⁻¹).

216 In the GOM, benthic sediments provide excess ammonium to overlying water by 217 regeneration processes such as remineralization (Lehrter et al., 2012; Nunnally et al., 2014; 218 Rowe et al., 2002). Generally, there is an uptake of nitrate and nitrite mainly by sedimentary 219 denitrification (McCarthy et al., 2015) or dissimilatory nitrate reduction to ammonium (DNRA) 220 and assimilation by benthic microalgae (Christensen et al., 2000; Dalsgaard, 2003; Thornton et al., 2007). Due to this, net DIN flux was used as the value of F_{Bott}^{DIN} , which shows DIN release 221 222 from bottom sediments to overlying water column. For example, in the GOM, the sum of 223 nitrate and nitrite fluxes to bottom sediments (e.g., May: -10.05, July -61.9, August: -48.42 µmol N m⁻² h⁻¹) were similar or smaller than the flux of ammonium from bottom sediments (e.g., May: 224 203, July: 152, August: 156 μ mol N m⁻² h⁻¹) off Terrebonne bay (McCarthy et al., 2015). In 225 226 the CSK, the sum of nitrate and nitrite flux to bottom sediments and ammonium flux are $0.5 \sim$ 1.4 mmol N m⁻² d⁻¹ and 1.3 ~ 9.6 mmol N m⁻² d⁻¹, respectively, which indicated that excess 227 ammonium with additional nitrate and nitrite were released from sediments in this region (Lee et 228

229 al., 2012). The release of nitrate and nitrite in the CSK unlike the GOM can be estimated due to 230 high inputs of nitrogen by groundwater in the CSK (Kim (G) et al., 2011) even though there is minor uptake of nitrate and nitrite. Diffusion from groundwater can probably be ignored in the 231 232 GOM as Rabalais et al. (2002) reported that the groundwater discharge is very low in coastal 233 Louisiana, but is likely important elsewhere and is known to be important in the CSK. Based 234 on this, we averaged and sum the fluxes data of nitrate, nitrite, and ammonium from McCarthy et 235 al., 2015 for the GOM and Lee et al., 2012 for the CSK, respectively, and then applied F_{Bott}^{DIN} value as 1.2 mmol N m⁻² day⁻¹ in the GOM and 6.2 mmol N m⁻² day⁻¹ in the CSK. Thus, 236 in equation 3, the benthic flux term is calculated from existing literature results after considering 237 238 all DIN fluxes as above (Lee et al., 2012; McCarthy et al., 2015), and then multiplied by the area 239 of each box.

The output terms are; 1) F_{Export}^{DIN} , the exchange rate between each box in the lower layer, 240 and 2) F_{Deni}^{DIN} , the denitrification rate from the water column. Due to high stratification at the 241 pycnocline, upward transfer of dissolved material from the lower layer to the upper layer is 242 243 assumed not to occur in our model. Also, denitrification from the water column below the 244 pycnocline is a significant N removal process, which removes up to a maximum 68% of total N input from the Mississippi River (MR) in the GOM (McCarthy et al., 2015). As the value of 245 F_{Deni}^{DIN} in the GOM, we used a direct measurement of denitrification rates from the McCarthy et 246 al., (2015) in the water column (88 µmol m⁻² h⁻¹, which converted to 2.1 mmol N m⁻² dav⁻¹) 247 248 where the stations were exactly same as our sub-region A, B, and C. We assumed this applied 249 only below the pychocline where oxygen concentrations decrease. However, in the CSK, there 250 is no water column denitrification data because the dissolved oxygen concentration has never been down below about 4 mg L⁻¹ during our data periods. Based on this, we estimated that 251

there is a very little water column denitrification in the CSK, so we did not count this term in theCSK. Thus, we only considered the sedimentary denitrification term for the CSK region.

254 Water transport in the region is generally from the east, i.e., from near the Mississippi 255 River in Sub-region A to the west, near the Atchafalaya River in Sub-region C during non-256 During summer, the winds change direction from easterly to westerly, summer periods. blocking the water flow to the west (Cho et al., 1998). We calculated advection from current 257 258 meter data collected during the LATEX program (Nowlin et al., 1998a, b) from April 1992 to 259 December 1994, from which we determined U (west to east flow) and V (south to north flow) 260 components (cm s⁻¹). Figure 4 shows the mean values of coastal ocean current velocities. The 261 annual range of the currents is 0 to 30 cm s⁻¹ for the longshore component, with standard deviation of about 8 cm s⁻¹, and 0 to 7 cm s⁻¹ for the cross-shelf component, with a similar 262 263 standard deviation, but these current velocities are not constant and change depending on time 264 and day. The annual current velocities in the CSK are more affected by tidal exchange and the 265 presence of the Yellow Sea Current, but velocities are similar to those in the GOM (Jacob et al., 266 2000; Lim et al., 2008). The annual range of the currents is around 0 to 28 cm s⁻¹ and 0 to 7 cm s^{-1} for the cross-shelf component. Thus, we used the mean value of the current velocity for the 267 268 time of year during each cruise in both the GOM and the CSK for calculating the advective flow 269 in both alongshore and onshore/offshore directions.

To run the box model, we assumed three factors: 1) the study area is in a steady state condition, with equal input sources and outputs, 2) AN-D is evenly distributed across each area, and 3) DIN is fully utilized by phytoplankton growth in the layer above the pycnocline, so we can neglect other removal factors. However, in the layer below the pycnocline, as we mentioned above, denitrification, which leads to a main loss of DIN as nitrogen gas, is

considered as another output term in Equation 3. Because we assumed that all DIN removed is fully consumed by primary production above the pynocline, we can calculate potential carbon fluxes and oxygen consumption using the Redfield ratio (C: N: $-O_2$: P = 106: 16: 138: 1). The PPP can be compared with ¹⁴C measurement data (Lohrenz et al., 1998, 1999; Redalje et al., 1994; Quigg et al., 2011) and dissolved oxygen data from MCH mooring C at 29° N, 92° W (4 March 2005 ~ 10 July 2005) (Bianchi et al., 2010).

- 281
- 282 **Results**

283 An N-mass balance model for the Texas-Louisiana Shelf

284 The existence of the three zones suggested by RC02 has been verified from winter data 285 using nutrient/salinity relationships (Kim 2018). Figure 5 shows the contour graph based on the 286 mean concentration of DIN at each station during the MCH M4 (March 2005) cruise. For 287 operational and modeling purposes, stations were grouped into three sub-regions - near the 288 Mississippi (A), near the Atchafalaya (C) and an intermediate region (B) between ~90°-91°W. 289 During summer, it is hard to use nutrient/salinity relationships directly because riverine nutrient 290 inputs are lower and phytoplankton growth causes rapid nutrient consumption over the shelf, 291 leading to low overall nutrient surface concentrations. We calculated the mean [DIN] in each 292 box, and then used the relationship between DIN and salinity to define the edges of the three 293 zones. Near the coast salinity was consistently low, with high turbidity from the river water 294 discharge. This was labelled the brown (river) zone.

A range of N input values from various sources were used in the N-mass balance model to estimate PPP and carbon fluxes in the coastal GOM. The PPP rates were highest near the river mouth and we set the boundaries of production for each zone based on our N-mass balance

model results and mean [DIN] data. We defined the brown zone as having the PPP rate of over 2 gC m⁻² day⁻¹ because of the high input of N from the river, AN-D, and benthic fluxes, and the rate in the blue zone is less than 0.1 gC m⁻² day⁻¹. The PPP rate in the green zone is then between 0.1 and 2 gC m⁻² day⁻¹. Basically, these PPP ranges were set based on synthesized measured ranges of coastal GOM primary production, as defined for near, mid, and far fields of the coastal GOM (Dagg and Breed 2003; Lohrenz et al., 1999). Note that our model results of the PPP might overestimate the actual production because of light limitation, following RC02.

305 The edges of the three zones above and below the pycnocline layer, based on our N-mass 306 balance model results, are shown in Figures 6a and b. The patterns of the boundaries above and 307 below the pycnocline differ from the edges of the zones. The brown zone was found above the 308 pycnocline on all cruises close to the Mississippi River mouth because of the high nutrient 309 concentrations, but only appeared off the Atchafalaya River in March 2005 (MCH M4). 310 However, below the pycnocline it was found only in April 2004 (MCH M1) in sub-region A. 311 This suggests that vertical transport across the pycnocline rapidly removes the high levels of 312 suspended material that cause light limitation above the pycnocline. In the green zones, the 313 nutrient source is mostly supported directly by the river, with minor additional sources of N from 314 vertical sinking, AN-D, and benthic fluxes. We utilized the vertical sinking flux from the 315 sediment trap data from Qureshi (1995) below the pycnocline layer to estimate PPP. This varied between 0.1-1.0 gN m⁻² day⁻¹ (Table 3). Typically, in the blue zone where biological 316 317 production is low, vertical sinking followed by local decomposition is assumed to be the major factor that changes the nutrient concentration in the lower layer. The blue zone is always more 318 319 extensive below the pycnocline than above it, which suggests there is little or no sub-pycnocline 320 production except close to the coast and/or the river mouths, and reinforces the assumption that any chlorophyll below the pycnocline is inactive (Figure 6b). Thus, we can identify the horizontal influence of the river plume in the layer below the pycnocline and the variation in the boundaries of the three zones, based on the observed nutrient data from a bottom layer and our N-mass balance model. The model suggests that regions of moderate potential productivity extend offshore at least as far as 28° 30'N in sub-region B, both above and below the pycnocline.

326

327 An N-mass balance model calibration

328 The model calibration was done with historic literature data. Literature data suggest that observed PP rates in the green and brown zones of the coastal GOM vary between 0.4 gC m⁻² 329 day⁻¹ (winter) and ~ 8 gC m⁻² day⁻¹ (summer) (Dagg et al., 2007; Lohrenz et al., 1998, 1999; 330 331 Redalje et al., 1994). Recently, Quigg et al. (2011) determined the integrated PP rates with ¹⁴C 332 measurements during 2004 in the coastal GOM. The highest integrated PP rates were found near the Mississippi River at 3.5 gC m⁻² day⁻¹ (in July), and near the Atchafalaya River at 2.7 \sim 333 5.9 gC m⁻² day⁻¹ (in May to July) (in the brown and green zones). However, lowest integrated 334 335 PP rates were on the outer part of the LATEX shelf (the blue zone) at 0.07 gC m⁻² day⁻¹ (in March), $0.04 \sim 0.15$ gC m⁻² day⁻¹ (in May), and $0.33 \sim 0.91$ gC m⁻² day⁻¹ (in July). Additionally, 336 337 Quigg et al., (2011) pointed out that these higher PP values were affected by high riverine 338 nutrients input from the MR that flows westward during that time period.

The actual PP ranges were similar with our model-based PPP (Figure 6). However, this was different from RC02's brown zone. This might be due to the differences between methods such as ¹⁴C, our N-mass balance model, and RC02's theoretical model. Typically, RC02 assumed that the brown zone is light limited due to high sediment turbidity, but our model does not account for this and only considered DIN concentrations. Except for this, our PPP results are similar to direct productivity measurements from the ¹⁴C incubations (Quigg et al., 2011).
Our model result (PPP) showed the same range of values as ¹⁴C incubations (e.g., Dagg et al., 2007; Lohrenz et al., 1998, 1999; Quigg et al., 2011; Redalje et al., 1994) in the three subregions.

Note that our model assumed all the biological uptake could be converted directly to production rates, which we considered as PPP. The PPP from cruises MCH M1 ~ M8 for samples from above the pycnocline calculated using our model is reasonable based on comparison with previous PP values (Figure 6a). The PPP ranges $(0.01 \sim 5.05 \text{ gC m}^{-2} \text{ day}^{-1})$ were similar to previous ¹⁴C measurement PP values of between $0.04 \sim 5.9 \text{ gC m}^{-2} \text{ day}^{-1}$.

353 Based on our model calculation, which assumes all the nutrients are available for 354 production, the PPP showed maxima at all times in sub-region A (near the Mississippi river) and 355 minima in sub-region B (between the Mississippi and Atchafalaya River), except for MCH M2 356 in June 2004, when sub-region C had the lowest PPP (Figure 6a). The high values in sub-357 region A are due largely to underutilization of nutrients in regions of high turbidity. As the 358 water flows west under the influence of the Coriolis effect, PPP is expected to decrease as a 359 result of declining nutrient concentrations because of dilution and nutrient uptake during 360 biological production while the water flows to sub-region B. In sub-region C, MCH M4 361 (March 2005) had the highest PPP among the all MCH cruises. This probably depended on 362 high nutrient concentrations being present during the winter period, when the region was affected 363 by Atchafalaya River nutrient input.

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³⁶⁵ *Model scenarios in the Gulf of Mexico (GOM)*

366 We tested the sensitivity of the model to changes in input/output parameters such as 367 increasing AN-D and decreasing riverine N input. Assuming the model is robust, we 368 investigated three model scenarios based on the nutrient distributions seen during the MCH1 369 cruise (note that using data from other cruises gives very similar results). In the first scenario, 370 we cut riverine N input 60% and increased the AN-D input by a factor of two based on 371 increasing N emission predictions (Duce et al., 2008; He et al., 2010; Kanakidou et al., 2016; 372 Kim (T) et al., 2011; Lawrence et al., 2000; Paerl et al., 2002). In the second scenario, we 373 doubled the amount of AN-D as in scenario 1 and decreased riverine N input by 30% based on 374 the hypoxia management plan goal (Gulf Hypoxia Action Plan Report, 2001, 2008; Rabalais et al. 375 2009). In the third scenario, we increased riverine N input by 20%, assuming the failure of the 376 hypoxia management plan, while we set the AN-D amount equal with the first and second 377 Based on our N-mass balance model calculation and model scenarios, we can scenarios. 378 initially estimate carbon fluxes from our PPP rate, and, using the Redfield carbon to oxygen 379 stoichiometry ratio (106:138), the overall oxygen balance within the coastal GOM (Table 4).

380 As can be seen in the scenario results for MCH M1 data (Table 4), the riverine N input 381 source is still the major controlling factor in the coastal GOM region even when its contribution 382 is greatly reduced and the AN-D source is doubled. For instance, if we fail to reduce riverine N 383 input in the future (scenario 3), the potential carbon fluxes will increase by 17% relative to 384 current conditions. In contrast, the AN-D input source only increased to a maximum of 5% of 385 the total input term and this indicates that AN-D input is still a minor factor in the GOM. If the 386 production is increased, overall oxygen demand will also be increased. The MCH M1 scenario 387 result indicated that the overall oxygen demand would increase approximately 21% if we fail to 388 reduce riverine N input, likely increasing considerably the area of the hypoxia.

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An N-mass balance model in the Coastal Sea off Korea (CSK)

As we have done in the GOM, we used our N-mass balance model to estimate the PPP in the CSK and define the three different zones (Figure 7). Similar to the GOM region, the PPP rates were highest near the river mouth, and we set the boundaries of each zone based on our Nmass balance model results. Based on nutrient data, as was done for the GOM, we defined the brown zone as having a PPP rate above $1.5 \text{ gC m}^{-2} \text{ day}^{-1}$ because of the increased N sources from the river, AN-D, and the sediment flux. We defined the green zone as having PPP rates between 0.3 to $1.5 \text{ gC m}^{-2} \text{ day}^{-1}$ and the blue zone as having rates of less than 0.3 gC m⁻² day⁻¹.

398 The seasonal results shown in Figures 7a and b show that the boundaries of the three 399 zones above and below the pycnocline layer were roughly consistent with the main change 400 coming in summer (August), which is the wet season and sees the highest river discharge. The 401 large size of the green zone in all seasons suggests that AN-D is consistently adding extra 402 nitrogen to the surface ocean along with the riverine N input. This is supported by the fact that 403 the PPP in the blue zone is an order of magnitude higher than for the GOM. Around 90% of the 404 grid cells in the CSK are in the same zones above and below the pycnocline (Figure 7 a and b) 405 during all four cruises; however, in the GOM (Figure 6 a and b) this was found for fewer than 406 half of the grid cells. This is probably due to the difference in freshwater discharge rate in the 407 two regions, which leads to a much larger stratified area in the GOM than in the CSK.

408 One question that has not been investigated is the temperature dependence of primary 409 productivity in the two areas. While the GOM is temperate throughout the year, winter 410 temperatures in the CSK fall to \sim 5°C. However, according to the ocean color remote sensing 411 images from near the CSK river mouth reported by Son et al., (2005), primary production in the

412 CSK does not appear to be strongly affected by temperature. The PPP results of our model (0.2 413 to 2.2 gC m⁻² day⁻¹) agreed with their ocean color remote sensing results (0.4 to 1.6 gC m⁻² day⁻¹) 414 in the CSK. Also, during all seasons, the Keum River consistently supplies high amounts of 415 DIN (average: < 60 μ M) (Lim et al., 2008) to the coastal zone (especially close to the Keum 416 mouth). We believe, therefore, that the higher value of PPP in winter near the Keum mouth 417 (brown zone in figure 7a), is reasonable.

The AN-D input source comes mainly from the Chinese side of the East China Sea (ECS) and this affects the boundaries of the green and blue zones above the pycnocline as it is deposited uniformly across the region. There is also nutrient input from offshore, as the Yellow Sea Bottom Cold Water Mass can up-well during the mixing process and is assumed to supply additional nutrients to the outer shelf (Lim et al., 2008).

423

424 Model scenarios in Mid-Western Coastal Sea off Korea (CSK)

425 AN-D is currently considerably more important (by approximately an order of magnitude) 426 in the CSK than in the GOM), and it is anticipated that AN-D will likely be a major controlling 427 factor here in the future (Duce et al., 2008; He et al., 2010; Kim (T) et al., 2011; Lawrence et al., 428 2000; Paerl et al., 2002). Because of the lack of research on potential hypoxia scenarios in 429 Korea, we used the same three scenarios in the CSK as were used for the GOM. Similar to 430 GOM results, riverine N input remains the major controlling factor; however, in this area, the 431 AN-D source is more critical than in the GOM region (Table 5). The AN-D input source 432 increased from 20% to 47% of the total input under scenario 1, while based on our scenario 3 433 results, increases in the AN-D input source and riverine N input together will affect biological

production by increasing carbon fluxes up to 25% and oxygen demand up to 32% if we fail toreduce N input in future (Table 5).

436

437 **Discussion**

438 Most previous model studies in the GOM were focused on predicting the hypoxia area 439 (Bierman et al., 1994; Fennel et al., 2011, 2013; Justic et al., 1996, 2002, 2003; Scavia et al., 440 2004). For example, Justic et al., (1996; 2003) used a two-layer model incorporating vertical 441 oxygen data, from one station (LUMCON station C6; 28.867°N, 90.483°W), to predict the size 442 of the hypoxia area. Similarly, Fennel et al. (2011; 2013) used her more complex simulation 443 model, which included oxygen concentration as well as a plankton model from Fasham et al. 444 (1990), to predict the size of the hypoxia region in the GOM. Our N-mass balance model, in 445 contrast, uses historical data from the LATEX shelf to estimate potential carbon fluxes in the 446 GOM, and calculate the overall oxygen demand from those carbon fluxes. While this affects 447 the total area subject to hypoxia it does not estimate the size of the hypoxic zone.

448 In contrast to our model, traditional predictive models have also ignored different 449 nitrogen input sources such as AN-D and SGD. While this is probably reasonable on the 450 Texas-Louisiana shelf, where riverine inputs dominate, it may not apply in other coastal regions. 451 As a result, model studies in this region have concluded that reducing riverine N input is the only 452 solution to decrease the size of the hypoxia area in the GOM (Gulf Hypoxia Action Plan Report, 453 2001, 2008; Rabalais et al. 2009; Scavia et al., 2013). According to our model results, AN-D is 454 still a minor controlling factor in the GOM; however, in the CSK, the AN-D contributed more to 455 the total nitrogen budget and may be a major controlling factor in the future. This indicates that 456 AN-D should be considered as another input term for nutrient managements, especially in Asia 457 or in other regions where high concentrations are expected. Similarly, nitrogen input from458 either sediment fluxes or groundwater also need to be considered.

Our zonal boundaries can be compared with the results of Lahiry (2007), who used salinity to define the edges of each zone for the three cruises MCH M1, M2, and M3 (Figure 8) and defined the edges of the RC02 zones in the coastal GOM based solely on salinity. Her limited simulation results indicated similar patterns to our model based on DIN concentration near the Mississippi River mouth (e.g., during MCH M1, M2, and M3). Mixing was more conservative in this region than further west because the low salinity water with high nutrients was less diluted with offshore water.

466 Away from the MR in sub-regions B and C, however, her results gave very different 467 boundaries for the three zones compared with our results (Figure 8). In particular, the results 468 near the Atchafalaya River were very different (compare Figures 6 and 8). For example, our 469 data showed only green and blue zones off Atchafalaya Bay during MCH M1, with no brown 470 zone. Similarly, the extent of the blue zones in sub-region C during MCH M2 and M3 is also 471 very different. We believe that our N-model based classification can cover more complex 472 biological processes than the Lahiry (2007) method, which considers only advection and mixing 473 and that our N-model is a more sensible way to look at biological processes in the GOM.

Our results also agree with previous studies that demonstrated that both the GOM and CSK regions are N-limited for most of the year (Kim (G) et al., 2011; Turner and Rabalais, 2013). This compares with the results of Sylvan et al., (2007), who reported that the coastal GOM could be P-limited in the MR delta mouth area where our brown zone is located, while RC02 suggested light-limitation rather than N- or P-limitation. However, this P-limited condition appears to occur when N concentrations are very high. In particular, the N/P ratios in

480 the both the GOM and CSK during our sampling were less than 16, indicating that both regions 481 were N-limited, although a few stations in the brown zone near the MR river area had ratios of 482 between 16 and 18 (Figure 9). These higher N to P ratios may result from the high sediment 483 turbidity causing light-limited conditions in this zone near the river mouth (Rowe and Chapman, 484 2002).

It should be remembered, however, that the arithmetic N:P value per se is unimportant in determining nutrient limitation. As long as both nutrients can be measured, it is theoretically possible for phytoplankton to continue to grow. The MARS has generally such an excess of N relative to P that N:P ratios >>16 can be expected as P concentrations fall, but this does not necessarily mean that productivity is limited, and we never found P concentrations of zero in any of our sub-regions; the lowest P concentration measured during all cruises in the GOM and CSK was 0.2μ M.

492 Both the GOM and CSK regions receive nitrogen inputs from AN-D, rivers, and benthic 493 fluxes. These different nitrogen input sources control coastal productivity, and this may reflect 494 the different nitrogen cycling in the two regions. In the GOM, the riverine N input source 495 consistently dominates coastal productivity and eutrophication, while, in the CSK, AN-D is also 496 becoming a critical controlling factor. In the CSK, however, there is strong tidal mixing of 497 freshwater from the Keum River and/or Gyunggi Bay with nearby coastal water, which results in 498 a tidal front along the offshore region and off the Taean Peninsula during spring and summer. 499 It is this physical mixing that mostly controls the spatial distribution patterns of nutrients and 500 salinity here, particularly below the pycnocline (Lim et al., 2008). The brown zone in the upper 501 layer in the CSK (August 2008) changed to a green zone region below the pycnocline layer as a 502 result of the strong coastal tidal mixing.

RC02 considered their model to be theoretical. In the brown zone, close to the river mouth, they assumed turbidity leads to light-limited conditions. Their results agree well with measured ¹⁴C PP numbers from Quigg et al. (2011) who found the lowest integrated PP is near the MR delta mouth. However, our N-mass balance model did not consider light limitation and therefore PPP in the brown zone is high. Such good agreement suggests that our model can be applied to a wide region, while ¹⁴C measurements are typically conducted at a few specific points, as long as such limitations are taken into account.

In the CSK, most previous production studies focused on inshore areas such as estuaries or rivers. Our research focused for the first time on the coastal ocean off Korea. Our results suggest that diverse nitrogen sources need to be recognized as potential issues for future nutrient management concerned with hypoxia, eutrophication, or other environmental issues. The agreement between our results and the pattern of production based on satellite-sensing in the CSK (Son et al., 2005), suggests that our model is reasonable.

516 The results of our changing scenarios represent how the biological processes in these 517 coastal regions may vary as individual nutrient sources change. While our model cannot 518 predict the area of the hypoxic zone, we can investigate the effects of potential flux changes of 519 each factor, such as AN-D, riverine input, or benthic fluxes, and calculate the effects of changes 520 in each on PPP and on the overall oxygen balance for the region. We have only considered 521 different input terms of our N-mass balance model; output terms such as water mixing rates and 522 the residence time for each box need more detailed study in future work to calculate more 523 realistic production changes in each box.

524

525 Conclusion

526 The model suggests that the three zone theory of RC02 can be applied not only in the 527 northern GOM but also in the CSK region and that three zones can be distinguished based on 528 their nutrient concentration. As a result, we believe that using our N-mass balance model to 529 separate different zones based on RC02 may be appropriate not only for large-scale regions like 530 the GOM and CSK but also at small scales such as river or estuary systems. The model also 531 estimates potential primary production and carbon flux based on the inclusion of AN-D data that 532 have not been considered previously (e.g. Bierman et al., 1994; Kim (T) et al., 2011). Our 533 results agree well with previous ¹⁴C measurements in the GOM (Quigg et al., 2011) and ocean 534 color remote sensing in the CSK (Son et al., 2005).

535 Based on CSK cruise data results, we can initially determine where the three different 536 zones are in the CSK. We evaluated our model and tested its sensitivity based on three 537 different scenarios. Through our scenario results, we assume that the AN-D is a considerable 538 factor in the CSK as well as the riverine N input from the Keum river. Reducing nutrient input 539 from the river is critical for hypoxia management policy (Gulf Hypoxia Action Plan Report, 540 2001, 2008; Rabalais et al. 2009). In addition, these model scenarios will be helpful in future 541 coastal nutrient management or hypoxia management studies in the CSK, especially as AN-D 542 sources become more important.

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961 List of Figures

- 962 Figure 1. Study sites and sampling areas in the Gulf of Mexico and Korea. (a) shows the 963 sampling area within the northern Gulf of Mexico. Flow in the Mississippi/ 964 Atchafalava River System is split 30% to the Atchafalava River, 70% to the Mississippi 965 River. The box is the sampling area. (b) shows station positions from March 2005. 966 Note that MCH project data are widely distributed across the region. Red, grey, and 967 blue stations correspond to sub-regions A (near the Mississippi River), B (between the 968 Mississippi and Atchafalaya), and C (near the Atchafalaya) respectively. (c) shows the sampling area off the west coast of Korea. (d) shows all of the station positions. 969 970
- Figure 2. The Rowe and Chapman three zone hypothesis, which describes the physical and biochemical processes that initiate and sustain hypoxia on the Texas-Louisiana Shelf,
 [Rowe and Chapman, 2002]. RMEPs are Reduced Metabolic End Products. *Reprinted with permission of Gulf of Mexico Science.*
- Figure 3. (a) Input (blue) and output (red) sources for each 0.25° box in the GOM and CSK (see
 text for details); (b) Area of each sub-region (red) and boxes affected by direct riverine
 input (blue) in the GOM. Export N (Mixing) represents the advective transport term.
 The processes of biogeochemical and transport processes of both regions are the same
 and each in/out put factor is the same in the GOM and CSK. Note that transfer
 between boxes occurs in both directions alongshore and onshore/offshore and is not a
 one-dimensional process as suggested in the diagram.
- Figure 4. Mean ocean current velocities (a) and standard deviations (b) for biweekly periods
 from August 1993 through December 1994 based on data from LATEX project.
 Positive values of U show eastward flow; positive values of V show northward flow.
- Figure 5. Extent of the three zones defined by RC02 based on the mean concentration of nutrient (DIN) at each station during the MCH M4 cruise in March 2005, showing their correspondence to the three sub-regions used in the box model. Red, grey and blue stations correspond to sub-regions A (near the Mississippi River), B (between the Mississippi and Atchafalaya), and C (near the Atchafalaya) respectively.
- Figure 6a. Areal distributions of the three zones using data from above the pycnocline, based on
 N-mass balance model results. Colors and numbers represent boxes found in each of
 the three zones in terms of potential productivity (Unit: gC m⁻² day⁻¹).
- Figure 6b. As for 6a, using data from below the pycnocline.
- 1000Figure 7a. The distribution of the three zones off Mid-western Korea (CSK) above the1001pycnocline based on the RC02 hypothesis applied to the N-mass balance model.1002Colors and numbers represent boxes found in each of the three zones in terms of1003potential productivity (Unit: gC m⁻² day⁻¹).
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1005 Figure 7b. As for 7a, using data from below the pycnocline

1006	
1007	Figure 8. Distribution of the three zones during cruises MCH M1-M3 based on salinity data
1008	(Lahiry, 2007). Areas shaded in three colors represent the brown, green and blue
1009	zones respectively.
1010	
1011	Figure 9. Dissolved inorganic nitrogen (DIN) against dissolved inorganic phosphorus (DIP)
1012	during sampling periods in the Gulf of Mexico (GOM) and Mid-western Korea (CSK).
1013	Nearly all samples had an N:P ratio of < 16 , which indicated potential N-limited
1014	condition. At a few points near the brown zone the ratio was between 16 -18; this is
1015	where light-limitation is expected according to RC02.
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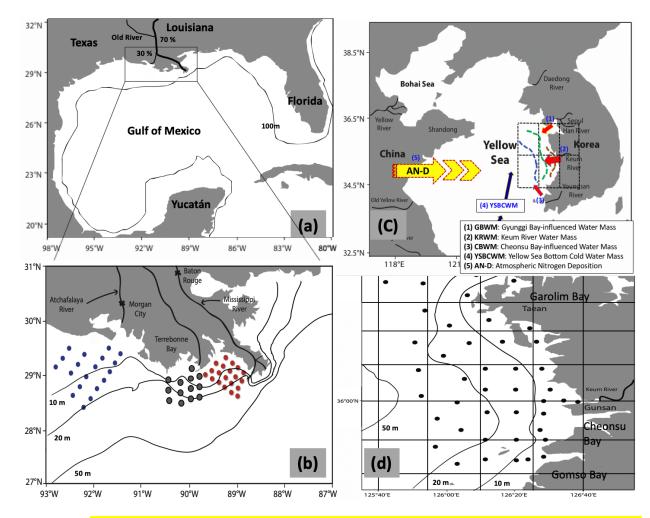
1017 List of Tables

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- Table 1. Sampling dates for data from Gulf of Mexico projects and the coastal sea of Korea.
 Winter data are listed for the Gulf of Mexico cruises.
- 1021 Table 2. Atmospheric Nitrogen Deposition (AN-D) in the USA and in the Yellow Sea.
- 1023Table 3. Definitions and values used in N-mass balance model to calculate DIN removal by1024biological production. (a) Each one quarter degree box; (b) Wade and Sweet 2008 for1025GOM region; (c) McCarthy et al., 2015 (d) Lee et al., 2012; (e) McCarthy et al., 2015;1026(f) Qureshi 1995. * F_{Atmo}^{DIN} of CSK region is used as mean values of Asia data in Table 2,1027which is initially 5 times higher than that of GOM (1.4 X 10⁵ mol day⁻¹). ** The unit1028of F_{Sink}^{DIN} was converted to mol day⁻¹ from the unit of original data (gN m⁻² day⁻¹) with1029area of box (0.25 m x 0.25 m) and molar mass of N (14 g mol ⁻¹). All unit were1030converted to mol day⁻¹ multiplied by area of box (0.25 m x 0.25 m).
- 1032Table 4. Simulation results for selected model scenarios based on MCH M1 (5 ~ 7 April 2004),1033as described in the text. Biological production is calculated using our N-mass balance1034model, while oxygen demand is calculated by the Redfield stoichiometry ratio (C: $-O_2$ 1035= 106: 138) (Unit: gC m⁻² day⁻¹).
- 1037Table 5. Simulation results for selected model scenarios based on CSK (February 2008) data, as1038described in the text. Biological production is calculated using our N-mass balance1039model, while oxygen demand is calculated by the Redfield stoichiometry ratio (C: $-O_2 =$ 1040106: 138) (Unit: gC m⁻² day⁻¹).
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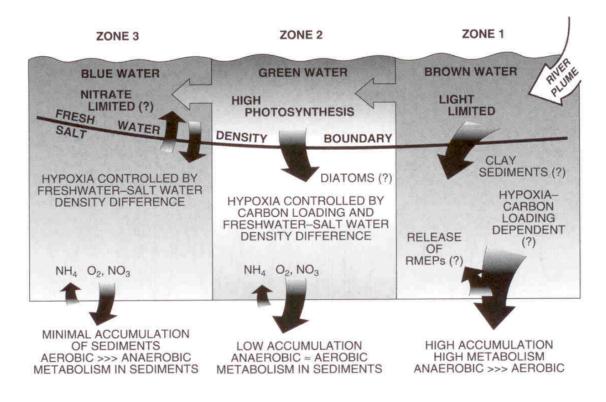
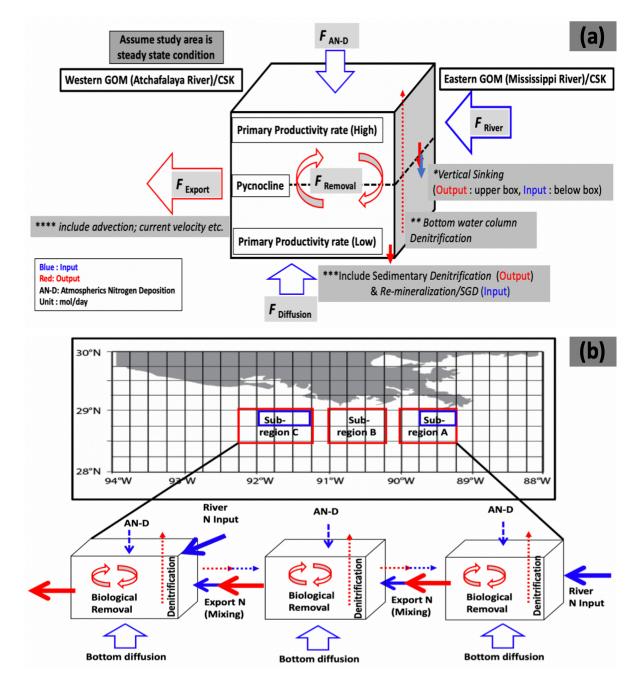


Figure 2. The Rowe and Chapman three zone hypothesis, which describes the physical and biochemical processes that initiate and sustain hypoxia on the Texas-Louisiana Shelf, [Rowe and Chapman, 2002]. RMEPs are Reduced Metabolic End Products. *Reprinted with permission of*

Gulf of Mexico Science.



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Figure 3. (a) Input (blue) and output (red) sources for each 0.25° box (see text for details); (b) Area of each sub-region (red) and boxes affected by direct riverine input (blue). Export N (Mixing) represents the advective transport term. The processes of biogeochemical and transport processes of both regions are the same and each in/out put factor is the same in the GOM and CSK. Note that transfer between boxes occurs in both directions alongshore and onshore/offshore and is not a one-dimensional process as suggested in the diagram.

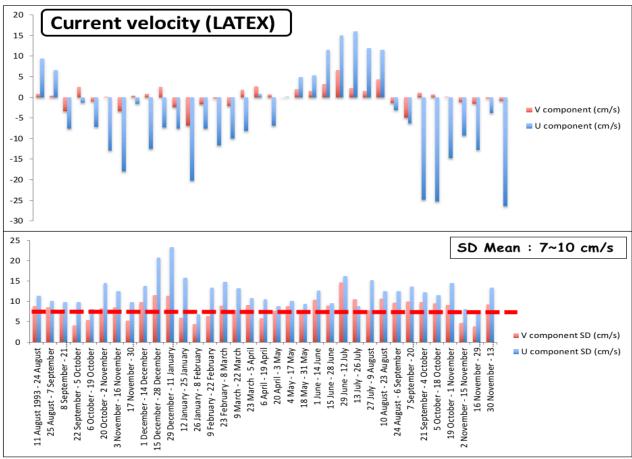


Figure 4. Mean ocean current velocities (a) and standard deviations (b) for biweekly periods from August 1993 through December 1994 based on data from LATEX project. Positive values of U show eastward flow; positive values of V show northward flow.

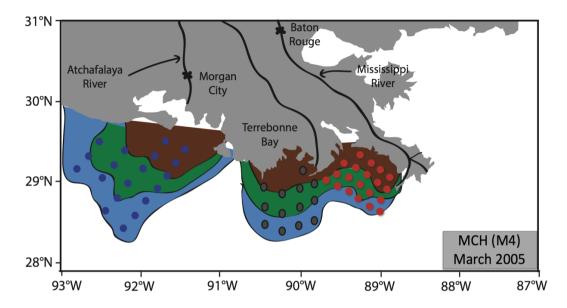


Figure 5. Extent of the three zones defined by RC02 based on the mean concentration of nutrient (DIN) at each station during the MCH M4 cruise in March 2005, showing their correspondence to the three sub-regions used in the box model. Red, grey and blue stations correspond to sub-regions A (near the Mississippi River), B (between the Mississippi and Atchafalaya), and C (near the Atchafalaya) respectively.

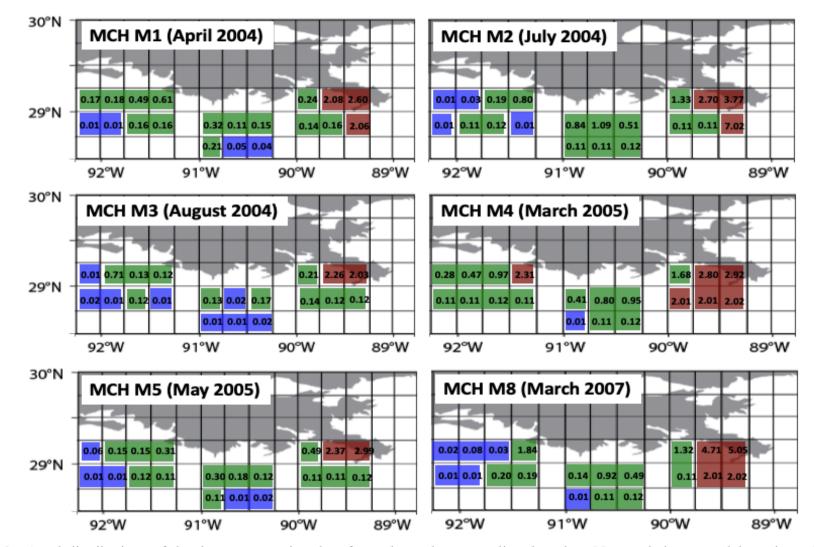


Figure 6a. Areal distributions of the three zones using data from above the pycnocline, based on N-mass balance model results. Colors and numbers represent boxes found in each of the three zones in terms of potential productivity (Unit: $gC m^{-2} day^{-1}$).

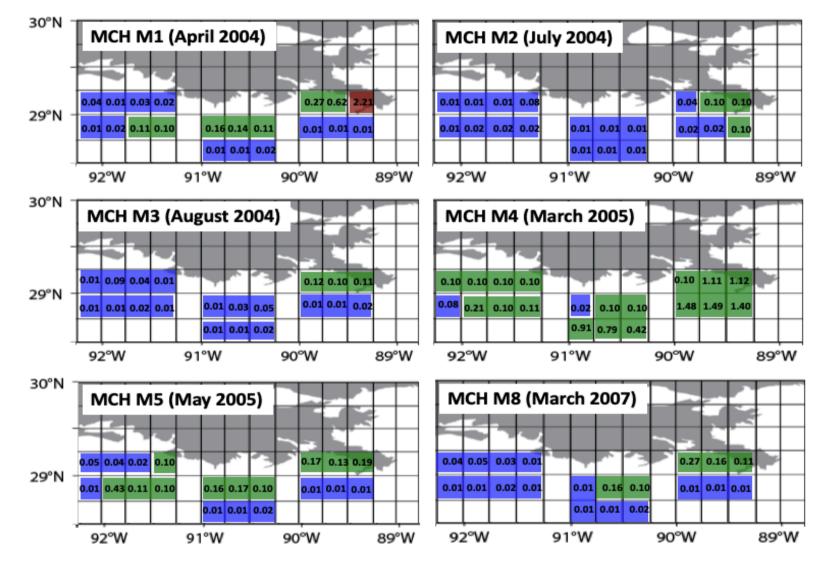


Figure 6b. As for 6a, using data from below the pycnocline.

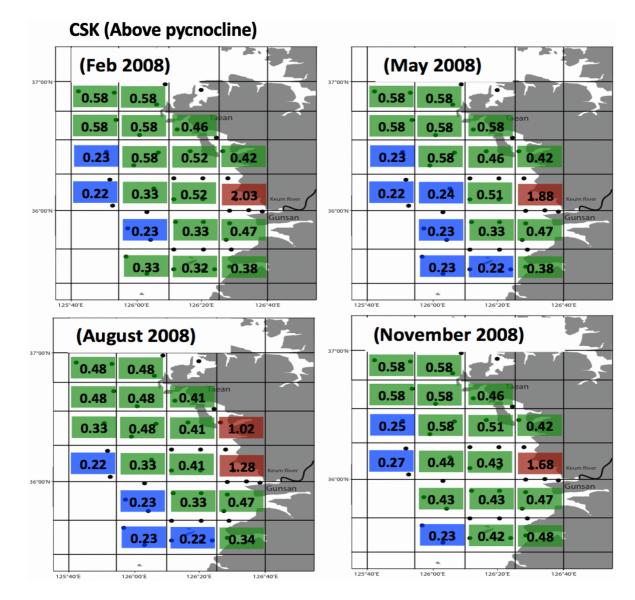
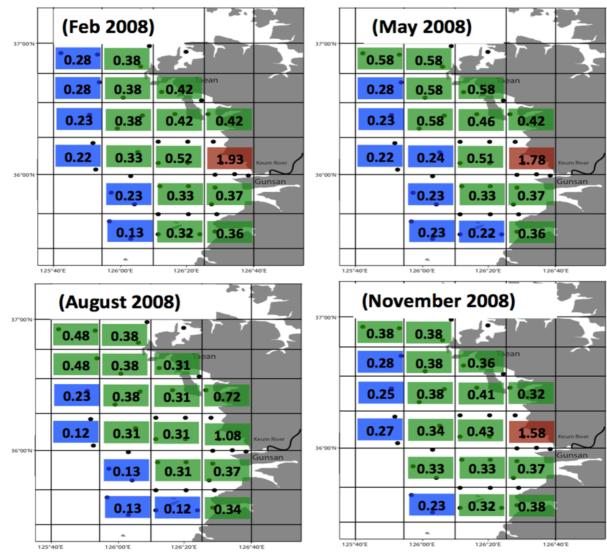


Figure 7a. The distribution of the three zones off Mid-western Korea (CSK) above the pycnocline based on the RC02 hypothesis applied to the N-mass balance model. Colors and numbers represent boxes found in each of the three zones in terms of potential productivity (Unit: $gC m^{-2} day^{-1}$).



CSK (Below pycnocline)

Figure 7b. As for 7a, using data from below the pycnocline.

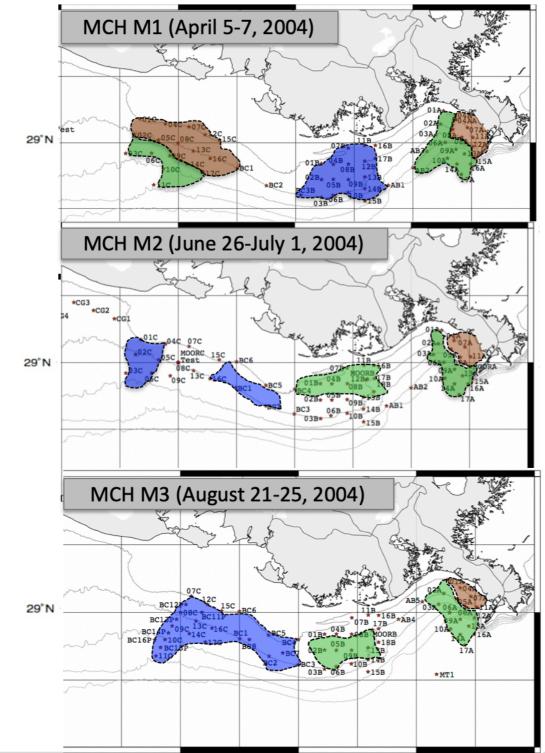
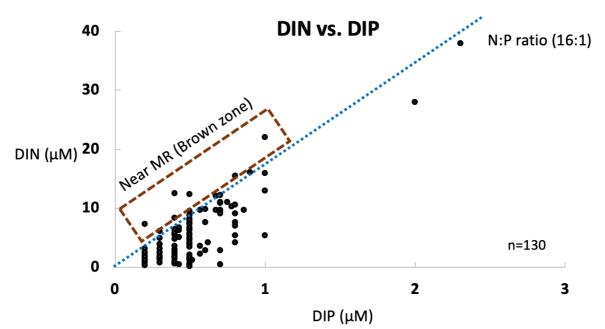


Figure 8. Distribution of the three zones during cruises MCH M1-M3 based on salinity data (Lahiry, 2007). Areas shaded in three colors represent the brown, green and blue zones respectively.



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1092 Figure 9. Dissolved inorganic nitrogen (DIN) against dissolved inorganic phosphorus (DIP)
1093 during sampling periods in the Gulf of Mexico (GOM) and Mid-western Korea (CSK).
1094 Nearly all samples had an N:P ratio of < 16, which indicated potential N-limited condition.
1095 At a few points near the brown zone the ratio was between 16 -18; this is where light1096 limitation is expected according to RC02.

	Study area	Date	Cruise number	
	Gulf of Mexico MCH	<mark>5 ~ 7 April 2004</mark>	MCH M1	
		26 June ~ 1 July 2004	MCH M2	
		21 ~ 25 August 2004	MCH M3	
		23 ~ 27 March 2005	MCH M4	
		<mark>20 ~ 26 May 2005</mark>	MCH M5	
		23 ~ 29 March 2007	MCH M8	
	Korea CSK	Feb, May, Aug	g, Nov (2008)	

1098 Table 1. Sampling dates for data from Gulf of Mexico projects and the coastal sea of Korea.
1099 Winter data are listed for the Gulf of Mexico cruises.

Watersheds	AN-D (g m ⁻² year ⁻¹)	References
Casco Bay, ME	0.15	Castro and Driscoll. 2002
Merrimack River, MA	$0.12 \sim 0.4$	Alexander et al. 2001
Long Island Sound, CT	0.18	Castro and Driscoll. 200
Delaware Bay, DE	$0.22 \sim 0.44$	Castro and Driscoll. 200 Goolsby. 2000
Chesapeake Bay	0.14 ~ 1.74	Alexander et al. 2001 Castro, M. S et al. 2001 Castro and Driscoll. 200 Goolsby. 2000
Gulf of Mexico	1~1.15	Wade and Sweet. 2008
Bohai Sea	6.42 ~ 14.25	Shou et al. 2018
Yellow Sea (China on the west side)	$1.61 \sim 1.84$ $2.99 \sim 3.28$ $3.81 \sim 9.24$	Zhao et al. 2015 Luo et al. 2014 Shou et al. 2018
Yellow Sea (Korea on the east side)	1.5 ~ 5.82	Kim (JY) et al. 2010

Table 2. Atmospheric Nitrogen Deposition (AN-D) in the USA and in the Yellow Sea.

1106**Table 3.** Definitions and values used in N-mass balance model to calculate DIN removal by biological production. (a) Each one quarter degree1107box; (b) Wade and Sweet 2008 for GOM region; (c) McCarthy et al., 2015 (d) Lee et al., 2012; (e) McCarthy et al., 2015; (f) Qureshi 1995.1108 $*F_{Atmo}^{DIN}$ of CSK region is used as mean values of Asia data in Table 2, which is initially 5 times higher than that of GOM (1.4 X 10⁵ mol day⁻¹).

1109 ** The unit of F_{Sink}^{DIN} was converted to mol day⁻¹ from the unit of original data (gN m⁻² day⁻¹) with area of box (0.25 m x 0.25 m) and molar

1110 mass of N (14 g mol⁻¹). All unit were converted to mol day⁻¹ multiplied by area of box (0.25 m x 0.25 m).

Unit	Definitions	Value
A_{Bott} (m ²)	Area of box	6.2 X 10 ⁸ m ² (a)
C_{Box}^{DIN} (µM)	DIN concentration in each area (box)	
V_{s} (m ³)	Water volume of box	ABott X Pycnocline depth
C_{EX}^{DIN} (mmol m ⁻³)	Different concentration between each box C _{EX} = (C _{On} - C _{Off}) or (C _{East} - C _{West}) for DIN	
λ_{Mix} (day ⁻¹)	Mixing rate of each box to box (A reciprocal of the water residence time)	
$\mathbf{F}_{\mathbf{River}}(\mathbf{day}^{-1})$	River discharge	
F_{River}^{DIN} (mol day ⁻¹)	DIN flux from each river discharge	
F_{Atmo}^{DIN} (mol day ⁻¹)	Diffusive flux from Atmospheric deposition (Bulk N deposition rate x A _{Bott} (A _{surface of ocean}) for DIN	1.4 X 10 ⁵ mol day ⁻¹ * (b)
F_{Bott}^{DIN} (mol day ⁻¹)	Benthic flux from the bottom sediments (Net DIN release considered regeneration, groundwater inputs, and uptake of NO_2/NO_3)	1.2 mmol N m ⁻² day ⁻¹ (c) 6.2 mmol N m ⁻² day ⁻¹ (d)
F_{Export}^{DIN} (mol day ⁻¹)	An advection term which calculated from the current velocity	
F_{Deni}^{DIN} (mol day ⁻¹)	Denitrification in the water column	2.1 mmol N m ⁻² day ⁻¹ (e)
F_{Sink}^{DIN} (mol day ⁻¹)	Vertical sinking of DIN flux from sediment trap data	$0.1 \sim 1 \text{ gN m}^{-2} \text{ day}^{-1} ** (f)$
F ^{DIN} _{Removal} (day ⁻¹)	Removal by biological production (Assuming that the other removal factors are negligible above the pycnocline layer)	

Table 4. Simulation results for selected model scenarios based on MCH M1 ($5 \sim 7$ April 2004). Biological production is calculated by our N-mass balance model. Oxygen demand is calculated by Redfield stoichiometry ratio (C: $-O_2 = 106$: 138) (Unit: gC m⁻² day⁻¹).

	F River	F _{AN-D}	F Bott/SGD	Biological production	Oxygen demand
Nominal Value	1.4 x 10 ⁷ (~98 %)	1.4 x 10 ⁵ (~1 %)	1.4 x 10 ⁵ (~1 %)	Base line	
Scenario 1	5.6 x 10 ⁶	2.8 x 10 ⁵	1.4 x 10 ⁵	~45%	~58%
	(~93 %)	(~5%)	(~2%)	decreased	decreased
Scenario 2	9.8 x 10 ⁶	2.8 x 10 ⁵	1.4 x 10 ⁵	~22%	~28%
	(~96 %)	(~3%)	(~1%)	decreased	decreased
Scenario 3	1.7 x 10 ⁷	2.8 x 10 ⁵	1.4 x 10 ⁵	~17%	~21%
	(~97 %)	(~2%)	(~1%)	increased	increased

Table 5. Simulation results for selected model scenarios based on CSK (February 2008)1116data. Biological production is calculated by our N-mass balance model. Oxygen1117demand is calculated by the Redfield stoichiometry ratio (C: $-O_2 = 106$: 138) (Unit: gC m⁻²1118day⁻¹).

	F River	F _{AN-D}	F Bott/SGD	Biological production	Oxygen demand
Nominal Value	1.9 x 10 ⁶ (~60%)	6.0 x 10 ⁵ (~20%)	6.0 x 10 ⁵ (~20%)	Base line	
Scenario 1	7.2 x 10 ⁵	1.2 x 10 ⁶	6.0 x 10 ⁵	~13%	~16%
	(~29%)	(~47%)	(~24%)	decreased	decreased
Scenario 2	1.3 x 10 ⁶	1.2 x 10 ⁶	6.0 x 10 ⁵	~2%	~2%
	(~41%)	(~39%)	(~20%)	decreased	decreased
Scenario 3	2.2 x 10 ⁶	1.2 x 10 ⁶	6.0 x 10 ⁵	~25%	~32%
	(~55%)	(~30%)	(~15%)	increased	increased