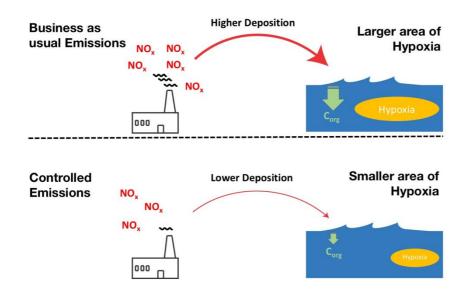
1	Quantifying the impact of anthropogenic atmospheric nitrogen deposition on the
2	generation of hypoxia under future emission scenarios in Chinese coastal waters
3	
4	
5	Yu Yan YAU <sup>1</sup> , David M BAKER <sup>2</sup> & Benoit THIBODEAU <sup>1</sup> *
6	
7	<sup>1</sup> Department of Earth Sciences and Swire Institute of Marine Science, The University of
8	Hong Kong
9	<sup>2</sup> School of Biological Sciences and Swire Institute of Marine Science, The University of
10	Hong Kong
11	
12	*Corresponding author: bthib@hku.hk
13	





15 TOC ART

## 16 Abstract

Atmospheric deposition is an important source of nitrogen to coastal waters. In nitrogen-17 18 limited waters, the atmosphere can contribute significantly to eutrophication and hypoxia. This is especially true in China, where nitrogen emissions have increased dramatically and are 19 20 projected to further increase in the future. Here, we modelled the potential future impact of 21 change in atmospheric nitrogen deposition on hypoxia in Chinese coastal seas. We used changes in nitrogen deposition under two IPCC scenarios that included emission regulation 22 and climate change (representative concentration pathways RCP 4.5 and 8.5) to evaluate the 23 24 impacts of such deposition on hypoxia in the 2030s and 2100s. We found that, by 2030, the 25 extent of hypoxic areas would increase up to 5% in China seas under RCP 8.5 due to the 26 projected increase in nitrogen deposition. However, hypoxia extent was projected to decrease by up to 9% by 2100 once emission regulations included in RCP 4.5 and 8.5 are implemented. 27 28 The South China Sea was found to be the most sensitive region to changes in nitrogen loads, 29 which indicates that more effort in emissions control is needed in order to avoid expansion of 30 the hypoxic zones in that specific region.

## 32 Introduction

33 Humans have significantly altered the cycling of atmospheric nitrogen (N) since the industrial revolution.<sup>1-3</sup> Over the past decades, industrial and agricultural advancements in addition to 34 35 urbanisation have contributed to a rapid increase of N production. Nitrogen is often the limiting nutrient controlling productivity in aquatic environments;<sup>4–6</sup> therefore, excess N can directly 36 impact the biogeochemical cycling in lakes, coastal regions and even the open ocean.<sup>7-12</sup> 37 Eutrophication and hypoxia are usually triggered by excessive nutrients. Many studies have 38 39 identified that hypoxia is associated with anthropogenic activities, such as industrial and 40 agricultural practices, which promote enhanced nutrient delivery via rivers and waste discharge.<sup>13</sup> These phenomena may provide additional dissolved inorganic nitrogen to coastal 41 42 regions, triggering the growth of phytoplankton and increasing oxygen consumption.

43

Atmospheric deposition is a non-negligible source of N to coastal waters. Elevated anthropogenic N emissions sourced from fossil fuel combustion and fertilizer use increases N deposition to the ocean.<sup>14,15</sup> The atmospheric N deposition supports up to 3% of annual new marine biological production globally.<sup>16–18</sup> China is one of the hotspots for N deposition, which has increased dramatically from 13.2 kg ha<sup>-1</sup> yr<sup>-1</sup> to 21.1 kg ha<sup>-1</sup> yr<sup>-1</sup> from 1980 to the 2000s<sup>16,19</sup>. Evidence shows that this elevated N could lead to environmental problems such as eutrophication and hypoxia.<sup>20</sup>

51

Atmospheric N deposition in N-limited aquatic systems can trigger the growth of phytoplankton and zooplankton.<sup>3,7,16,21–23</sup> Marine organic matter produced from their growth sinks to bottom waters and accumulates in sediments, where it is decomposed by bacteria, consuming dissolved oxygen in the process.<sup>24–26</sup> The high demand of oxygen consumption from microbial respiration and nitrification can thus decrease the concentration of dissolved

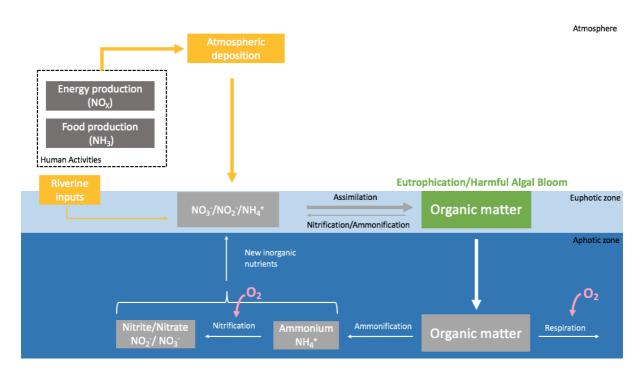
oxygen in poorly-ventilated bottom waters and thereby lead to hypoxia.<sup>25,27,28</sup> Moreover, N in 57 58 organic matter is remineralized and can be returned as new inorganic nutrients to the euphotic zone by vertical mixing and upwelling, producing the so-called N cascade (Figure 1).<sup>16,29–31</sup> 59 60 Increases in ocean productivity can thus lead to the expansion of hypoxic zones, often called dead zones, which are frequently observed in the Gulf of Mexico, Chesapeake Bay, the St. 61 Lawrence Estuary, the Baltic Sea and many other coastal ecosystems.<sup>32–37</sup> Hypoxic areas are 62 usually located near productive fisheries crucial to the economy of adjacent communities and 63 multiple stakeholders, such as in the case of the Gulf of Mexico.<sup>35</sup> Fish death due to hypoxia 64 can therefore cause considerable economic losses.<sup>38–41</sup> As such, increases in atmospheric N 65 deposition may induce a cascade of ecological, economic and social consequences.<sup>22,31,42,43</sup> 66

67

Eutrophication and seasonal hypoxia have been major environmental issues in China.<sup>44–48</sup> 68 69 During the summer of 2006, a hypoxic zone of more than 15,000 km<sup>2</sup> was reported in the East China Sea near the Yangtze River.<sup>49,50</sup> The East China Sea is one of the largest coastal oxygen-70 71 depleted regions globally, and is simultaneously a major Chinese fishery.<sup>50,51</sup> Although river and groundwater discharge contributes to the high nutrient input to coastal waters in China,<sup>52</sup> 72 atmospheric deposition also plays a key role in supplying N (Figure 1).<sup>53-56</sup> Due to 73 74 industrialization, increased use of vehicles and food production<sup>57</sup>, China is one of the largest emitters of anthropogenic N globally.<sup>58</sup> Anthropogenic N emissions in China increased 6-fold 75 from the 1980s to 2010s, and are predicted to increase until 2030.<sup>59</sup> This rapid increase of N 76 77 emissions on land causes a large increase of atmospheric N deposition on both land and in the surrounding ocean.<sup>54,60–67</sup> The deposition of this emitted N rose by 60% from 1980 to 2000 in 78 China.<sup>68</sup> Despite increased emission and deposition of N in Asia, the northwestern Pacific 79 ocean and the east part of the Sea of Japan is predicted to remain N-limited.<sup>69</sup> Although the 80 Pearl River and Yangtze River are rich in nitrogen, beyond the plume area of South China Sea, 81

East China Sea and Yellow Sea have a relatively low N:P ratio<sup>70–73</sup>. Hence, increases in atmospheric N deposition are likely to directly affect the primary productivity and nutrient cycling in the South China Sea and the other regions considered here.





86

87 Figure 1. Schematic graph of the contribution of atmospheric deposition to hypoxia in the coastal marine environment. The atmosphere receives N from combustion of fossil fuels 88 and the use of fertilizers. Because of the relative short residential time in the atmosphere, N 89 90 can directly be deposited to the coastal ocean. Consequently, additional nutrients in the surface 91 ocean may stimulate the rapid production of organic matter in N-limited coastal oceans and 92 thus enhance primary productivity. Thus, this addition of N can increase the risk and duration 93 of harmful algal blooms. When organic matter sinks to bottom waters, oxygen is consumed by 94 different pathways such as respiration, degradation of organic matter and nitrification. When 95 oxygen consumption exceeds oxygen supply in the water column, hypoxia is generated. 96

97 Climate change can influence hypoxia through various mechanisms. A warmer climate may 98 stratify the water column, which inhibits the vertical exchange of oxygen and thus the 99 replenishment of oxygen.<sup>74</sup> Higher temperatures also reduce the solubility of oxygen in 100 seawater and increases microbial activity, supporting enhanced degradation of organic matter 101 in bottom waters and thus contributing to increased oxygen demand.<sup>28,75,76</sup> Changes in 102 precipitation will also affect the amount of N deposited, since precipitation is one of the main

factors controlling the wet NOy deposition.<sup>77,78</sup> Precipitation is predicted to increase in eastern 103 104 south China.<sup>79,80</sup> and therefore could be an additional mechanism for enhancing atmospheric nitrogen deposition and increasing hypoxia. In addition to China, this effect may also be 105 particularly significant throughout India and Southeast Asia<sup>81</sup>. Hence, it is crucial to understand 106 107 how climate change and anthropogenic emissions manifest changes in N deposition, and its 108 particular impact on Chinese coastal waters. While various studies have estimated atmospheric N deposition over continental China.<sup>82,83</sup> and its implications for primary productivity in 109 Chinese coastal seas, <sup>54,84</sup> the indirect biogeochemical consequences of atmospheric deposition 110 111 such as the generation of hypoxia are seldom included. Moreover, it is difficult to isolate the 112 effect of a single source of nitrogen when analysing field data. Thus, the impact of deposited 113 N on hypoxia in China coastal waters is still unresolved and unquantified.

114

115 Here, we quantified the future impact of changes in N deposition on the extent of hypoxic areas in Chinese coastal waters using the future changes in atmospheric deposition over coastal 116 117 China under the impact of climate change and emissions scenarios. We used the projection 118 results from the Atmospheric Chemistry and Climate Model Intercomparison Project 119 (ACCMIP), which accounts for the Representative Concentration Pathways (RCPs) adopted in 120 the fifth assessment report of the Intergovernmental Panel on Climate Change (IPCC). The 121 RCPs are used to predict potential trends of atmospheric greenhouse gas concentrations based on different socio-economic factors.<sup>85,86</sup> These scenarios considered the effect of climate 122 123 change and emission control regulations, and thus can be used to model the potential impacts 124 of future anthropogenic emissions. In RCP 4.5, anthropogenic emissions of greenhouse gases are predicted to increase and peak in the 2030s ("stabilisation" scenario), while in RCP 8.5 125 emissions continually increase (i.e., a typical "business-as-usual" scenario).<sup>87-91</sup> We 126 incorporated the ACCMIP modelling results into a global biogeochemical model 127

### Manuscript accepted in Environmental Science & Technology doi: 10.1021/acs.est.0c00706

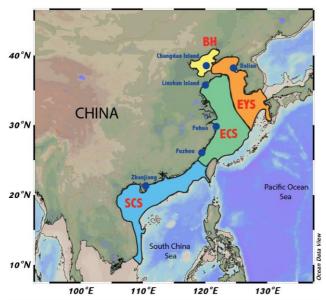
(COOLBEANS) to compute hypoxic areas under RCP 4.5 and 8.5 scenarios and the related change in N deposition. After discussing the limitations of the model, we analysed the impact of N-atmospheric on carbon fluxes and hypoxia as well as the temporal and spatial variability of the extent of hypoxia under ACCMIP results using RCP 4.5 and 8.5, which include both the effect of climate change and socio-economic factors on the deposition of N. Finally, we discuss the implication of our results with regard to the potential reduction of hypoxic areas following emission regulation and stabilisation of emission in China.

135 **2. Materials and Methods** 

# 136 Estimating new marine primary productivity and associated carbon fluxes derived from

### 137 total N deposition

We define "modern-day" N deposition based on the measurement of the atmospheric N 138 deposition (dry and wet) described by Luo et al.<sup>82</sup>. They measured the N deposition in various 139 140 coastal Chinese cities; Dalian, Changdao, Linshandao, Fenghua, Fuzhou and Zhanjiang (Figure 141 2). We assume a similar rate of N deposition in the coastal cities and the surrounding coastal 142 Seas. We use the four coastal zones of China defined in COSCAT; East Yellow Sea, Bohai Sea, East China Sea and South China Sea as our study area (Table S1).<sup>92,93</sup> Thus, we take the 143 average N deposition of the nearby coastal cities as "modern-day" N deposition in each study 144 145 zones (Figure 2).



147 100°E 110°E 120°E 130°E
148 Figure 2. Coastal marine environments considered in this study. The East Yellow Sea
149 (EYS), Bohai Sea (BH), East China Sea (ESC) and South China Sea (SCS) are defined in
150 COSCAT (Table S1).<sup>92</sup> The circles represent coastal cities used in Luo et al.<sup>82</sup> to measure N
151 deposition which is used in this study to establish the modern-day N deposition in each zones.
152 This figure was created using Ocean Data View.<sup>94</sup>

154 To estimate future changes in N deposition in China, we used the projected percentage changes 155 of dry and wet N deposition in the 2030s (2030-2039) and 2100s (2100-2109) under RCP4.5 and RCP8.5 scenarios reported in Zhang et al.<sup>95</sup>, which used the results from ACCMIP models. 156 The future scenarios considered both the impact of climate change and emissions. In the 157 projection of future N deposition, the scenarios account for the change in both dry and wet 158 deposition. Thus, changes in precipitation are included in our scenarios. Our study zones are 159 slightly different from Zhang et al.<sup>95</sup> because we followed the COSCAT model<sup>96</sup> which 160 accounts for river catchment and discharge. We therefore used the raw data provided by Zhang 161 et al.<sup>95</sup> and recalculated the future percentage change of N deposition based on each COSCAT 162 zones<sup>96</sup>. Then we averaged dry and wet N deposition and seasonal changes in each coastal 163 164 ocean as the regional mean changes (in %) relative to the modern-day deposition (Table S3).

166	Using the modern-day N deposition measured by Luo et al. <sup>82</sup> and future average percentage
167	change, we calculated the future N deposition (2030 and 2100) under RCP 4.5 and RCP 8.5
168	scenarios in each region. The difference of N deposition in each region between the future
169	scenario (2030 and 2100) and modern-day scenario (2010) was then converted into changes in
170	carbon flux using equation (1), assuming that all deposited N is assimilated by phytoplankton.
171	It is further assumed that a C:N ratio in coastal and shelf systems is 10:1, which is slightly
172	higher than 6.625 in the Redfield ratio because some of the nitrogen-rich organic matter is lost
173	in the water column before sedimentary deposition.93,97,98 Hence, carbon flux can be derived
174	from the total N deposition according to

- 175
- 176
- 177

178 where *C* represents the carbon flux (mol C m<sup>-2</sup> yr<sup>-1</sup>) and *N* represents the N deposition (mol N 179 m<sup>-2</sup> yr<sup>-1</sup>).

C = 10N

180

### 181 Calculating maximum hypoxic areas in the coastal Chinese seas

COOLBEANS (Coastal Ocean Oxygen Linked to Benthic Exchange And Nutrient Supply) is 182 a model that links changes in nutrient fluxes with changes in coastal hypoxia.<sup>93</sup> The model 183 184 quantifies bottom oxygen demand using relationships with aerobic respiration, organic matter 185 decomposition, vertical exchange and iron and sulphate reduction. Parameters include water 186 depth, surface salinity, surface temperature, primary productivity and surface oxygen for each zone to account for spatial variations (Table S2). Thus, it can be used to predict maximum 187 188 coastal hypoxic areas under shifts in nutrient loads. COOLBEANS uses a modified version of 189 the COSCAT zones and only consider the biochemically active part of the zone (i.e., the part where 80% of the productivity is located) to account for the fact that the impact on hypoxia 190

### Manuscript accepted in Environmental Science & Technology doi: 10.1021/acs.est.0c00706

will be concentrated where primary productivity is. A detailed description and validation of the 191 model was published previously<sup>93</sup> and only the most important parts will be presented here, we 192 refer the reader to the supplementary information and the original publication for more details. 193 194 Here, we used COOLBEANS to estimate hypoxic areas caused by future changes in N 195 deposition by applying changes in carbon fluxes calculated from modern-day and future N 196 deposition (equations S1 to S5 in SI). We further compared the maximum hypoxic area from future scenarios and present-day hypoxic areas to assess the impacts of future N deposition on 197 the area of hypoxia over coastal Chinese waters. Therefore, we assume riverine input remains 198 199 constant in future scenarios in order to isolate the impact from atmospheric deposition. The 200 uncertainty of the extent of hypoxic areas predicted for future scenarios is calculated based on 201 the standard deviations of future N deposition in the ACCMIP models, which was reported in Zhang et al.<sup>95</sup>. 202

203

### 204 **Results**

## 205 Calculation of new primary productivity and carbon fluxes derived from N deposition

In this study, we assumed that all N deposition in surface waters would be assimilated by phytoplankton and contributed to carbon fluxes.<sup>93</sup> We found that modern-day atmospheric N deposition would translate to a carbon flux of 25.6 g C m<sup>-2</sup> yr<sup>-1</sup> (East Yellow Sea), 28.1 g C m<sup>-2</sup>  $^{2}$  yr<sup>-1</sup> (Bohai Sea), 27.9 (East China Sea) and 37.0 g C m<sup>-2</sup> yr<sup>-1</sup> (South China Sea) (Figure 3), which indicates that the atmospheric deposition of N can support 2 to 12% of the total carbon flux. The East Yellow Sea has the highest percentage contribution of atmospheric to the total carbon flux (12.3%) (Table 1).

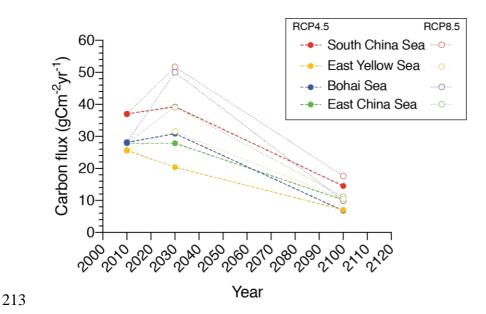


Figure 3. Modelled changes in carbon fluxes in Chinese coastal waters under RCP 4.5 (dots) and RCP 8.5 scenarios (open dots). All zones have higher carbon flux in 2030 under high atmospheric deposition (RCP 8.5) compared to 2010. We also note that in 2100 the carbon flux is lower is lower under both scenarios that includes emission regulation and climate change. Dots represent our modelled data and the dashed lines represent interpolation between our datapoint.

N deposition is shown to result in an increase in carbon fluxes in 2030 and a decrease in 2100 221 (Figure 3). We estimate the carbon flux to be 20.4 and 31.5 g C m<sup>-2</sup> yr<sup>-1</sup> (East Yellow Sea), 222 30.9 and 50.0 g C m<sup>-2</sup> yr<sup>-1</sup> (Bohai Sea), 27.5 and 39.2 g C m<sup>-2</sup> yr<sup>-1</sup> (East China Sea) and 51.7 223 and 59.3 g C m<sup>-2</sup> yr<sup>-1</sup> (South China Sea) in 2030 for scenarios RCP 4.5 and 8.5 respectively 224 225 (Figure 3). All carbon fluxes are projected to peak in 2030, with the exception of the East China Sea, where the carbon flux decreased slightly (Figure 3). By 2100, the atmospheric contribution 226 to the carbon flux decreased and was estimated to be 7.1 and 10.4 g C m<sup>-2</sup> yr<sup>-1</sup> (East Yellow 227 Sea), 6.8 and 9.9 g C m<sup>-2</sup> yr<sup>-1</sup> (Bohai Sea), 10.2 and 11.2 g C m<sup>-2</sup> yr<sup>-1</sup> (East China Sea) and 17.6 228 and 14.5 g C m<sup>-2</sup> yr<sup>-1</sup> (South China Sea) for scenarios RCP 4.5 and 8.5 respectively. The Bohai 229 230 Sea is the most affected area, with an increase in its carbon flux of 78% in 2030 and a decrease of 80% from 2030-2100 under RCP 8.5 (Figure 3). Overall, projected atmospheric N deposition 231 would support between 2 - 15% and 1 - 6% of the total coastal carbon flux in 2030 and 2100 232 233 respectively.

# 235 Table 1. The contribution of atmospheric N deposition to the total carbon flux in each

236	zone in	percent
		T

		<b>RCP 4.5</b>		<b>RCP 8.5</b>	
_	Modern day	2030	2100	2030	2100
East Yellow Sea	12.3	9.8	3.4	15.2	5
Bohai Sea	2.1	2.3	0.5	3.7	0.7
East China Sea	4.4	4.4	1.6	5.2	1.8
South China Sea	8.5	11.9	6	13.3	4.8

<sup>237</sup> 

### 238 Future changes in maximum hypoxic areas

239 Comparing to the modern-day maximum hypoxia area, projected maximum hypoxic areas in 2030 are estimated to change from -2.3% to +3.3% under RCP 4.5 and by +1.8% to +4.7%240 under RCP 8.5 depending on the region. Yet, by the end of 2100 the area affected by hypoxia 241 is projected to decrease by 1.6% to 8.7% in RCP 4.5 and 1.3% to 7.1% in RCP 8.5 relative to 242 243 the modern-day scenario due to a reduction in anthropogenic emissions reflecting predicted governmental regulations (Figure 4 and Table S3). Compared to the maximum hypoxic area 244 reached in 2030, the hypoxia area of the China's coastal ocean decreases up to 6.6% under 245 RCP 4.5 and up to 9.9% under RCP 8.5 by 2100. The South China Sea has the largest rate of 246 change in hypoxia area per unit N loading (1.12 km<sup>2</sup>/mg N m<sup>-2</sup> vr<sup>-1</sup>) followed by East Yellow 247 Sea (0.96 km<sup>2</sup>/mg N m<sup>-2</sup> yr<sup>-1</sup>), East China Sea (0.46 km<sup>2</sup>/mg N m<sup>-2</sup> yr<sup>-1</sup>) and Bohai Sea (0.05 248  $km^2/mg N m^{-2} yr^{-1}$  (Table S3). Moreover, the South China Sea has the largest proportion of its 249 biogeochemical active area that is hypoxia (31%) compared to other Chinese coastal oceans 250 251 which is around 13%.

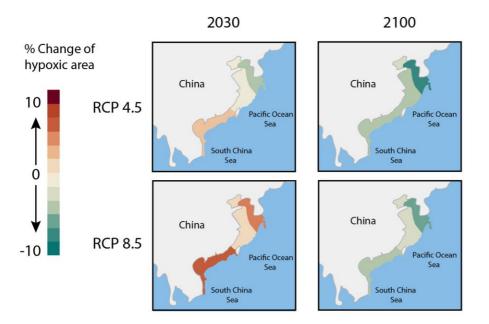


Figure 4. Modelled hypoxic areas (km<sup>2</sup>) in Chinese coastal seas in modern-day and future N deposition under RCP 4.5 and RCP 8.5 emissions scenarios. The figure highlights the high sensitivity of dissolved oxygen to nitrogen load, especially in the South China Sea (left panels) but also the potential of improvement of water quality under emission reduction policy (right panels). This figure was created using Ocean Data View.<sup>94</sup>

# 259 **Discussion**

# 260 Impact of atmospheric deposition on primary productivity and carbon fluxes

Modern-day modelled atmospheric deposition accounts for about 30 g C m<sup>-2</sup> yr<sup>-1</sup> on average to the carbon flux in coastal Chinese waters (1.5 - 6.8 Tg C), which contributes to 2-12% of the total carbon flux (Figure 3). This is consistent with previous studies, whose estimations range from 2.3 to 68 g C m<sup>-2</sup> yr<sup>-1</sup>, contributing to 0.1 - 37% of the new primary productivity in the region.<sup>54,56,99–101</sup> This highlights the importance of atmospheric N deposition to the total carbon flux in coastal Chinese waters, and validates the effective performance of COOLBEANS in reproducing field data from this region.

268

Our calculations estimate that atmospheric input contributes around 12% of the total carbonflux in the East Yellow Sea, which is the highest among the coastal ocean in our study (Table

### Manuscript accepted in Environmental Science & Technology doi: <u>10.1021/acs.est.0c00706</u>

1). This may be explained by the low total carbon flux characteristic of the East Yellow Sea compared to other coastal zones, therefore the contribution of atmospheric input will be more significant in the region. Moreover, the East Yellow Sea is deeper than other sites considered here (150 m), which is used by COOLBEANS to calculate the total flux of organic carbon. Thus, the depth of the East Yellow Sea translates into a lower carbon flux compared to our other sites. These results highlight the spatial differences in the impact of changes in atmospheric deposition on the total carbon flux in Chinese coastal waters.

278

### 279 Temporal variation of coastal hypoxia

280 Under climatic scenarios RCP 4.5 and RCP 8.5, the average hypoxic areas in China coastal 281 water will increase by about 1.6% relative to the modern-day by 2030. Our results are slightly 282 higher but on the same order of magnitude as the modelled cumulative effect of climate change (increase air temperature of +3 °C) and river discharge (increase 10% of river discharge) 283 reported by Lehrter et al.<sup>102</sup> in the Gulf of Mexico (+1%). This indicates that increases in 284 285 atmospheric deposition due to human activity and climate change has an enhanced effect on the extent of hypoxia than climate change. For year 2100, we estimate an average decrease in 286 287 carbon fluxes (-72% compared to modern-day) and thus hypoxia area (-4%) in coastal Chinese waters under RCP 4.5 and RCP 8.5 scenarios. This decreasing trend is similar to Cabré et al.<sup>103</sup>, 288 289 who modelled a decrease of 37% in total primary productivity in the low-latitude upwelling 290 area under climate change, which would also lead to a decrease in the area of hypoxia.

Our results suggest that hypoxic conditions will peak in 2030 and decrease in 2100, following the predicted trend of anthropogenic emissions. This illustrates how governmental control on emissions is important to control hypoxia. However, total N deposition and NO<sub>X</sub> and NH<sub>3</sub> emissions in China began to stabilize in recent years, earlier than what RCP 4.5 and 8.5 predicted, due to stricter environmental policies in China.<sup>104,105</sup> While this doesn't change our quantification of the extent of hypoxia under a given reduction in N deposition, it could meanthat the extent of hypoxic areas may start to stabilize and decrease earlier than 2030.

298

While scenarios RCP 4.5 and RCP 8.5 consider the effect of climate change on N deposition, physicochemical consequences of a warmer ocean are not included in COOLBEANS. Yet, impending sea surface warming may enhance stratification of water column and result in less mixing and oxygenation of bottom water, thereby increasing in the extent of coastal hypoxia.<sup>27,103,106,107</sup> Moreover, warmer ocean also decreases the solubility of oxygen and increase remineralization rate, which may further worsen the hypoxia.<sup>107</sup> Thus, our estimates might be considered conservative in this regard.

306

## 307 Spatial variation of sensitivity to nutrient input: The South China Sea

308 We observed spatial variation in forecasted future maximum hypoxic areas and their rates of change in coastal Chinese waters. The highest rate of hypoxia expansion per unit N added per 309 310 year was found in the South China Sea, followed by the East Yellow Sea, East China Sea and 311 Bohai Sea (Table S4). This can be attributed to the sensitivities of each respective coastal ocean to nutrient loading in COOLBEANS, which are mainly controlled by the bottom oxygen 312 demand and vertical exchange of oxygen.<sup>93</sup> The bottom oxygen demand in the model is driven 313 314 by depth and primary productivity in each zone. The South China Sea has a higher calculated 315 bottom oxygen demand among the four zones in the model because of its relatively deep shelf 316 and low vertical exchange of oxygen, resulting in lower modelled concentrations of bottom oxygen and a larger area of hypoxia.<sup>13,45</sup> Thus, controlling N input to the South China Sea, 317 318 including atmospheric emissions, is critical to reduce coastal hypoxia. This is a grand challenge 319 due to the densely populated regions on its coastline, including the Greater Bay Area, which 320 houses about 120 million people and multiple industries. The high sensitivity of South China Sea might increase when considered atmospheric inputs together with the effects of global
 warming such as enhanced stratification and the interplay of ongoing ocean acidification,
 eutrophication and hypoxia.<sup>46</sup>

324

## 325 Effect of a reduced emissions scenario on hypoxia

326 Hypoxia reduction targets should be set in China in order to reduce the ecological, social and economic impacts associated with the phenomenon. Taking the Gulf of Mexico as an example, 327 328 the intergovernmental Gulf Task Force set a goal of reducing the hypoxic zone by about 70% (~5000 km<sup>2</sup> over a 5-year average) by 2035 in Gulf of Mexico and reducing 20% of the spring 329 N loading from the Mississippi River.<sup>108</sup> Although this exact target may not be applicable in 330 331 China, it can be used as a reference to evaluate measurable policy actions required to achieve 332 acceptable reductions in N emissions and hypoxia. From our model, a reduction of 30 to 90% of N input, which includes atmospheric deposition and riverine inputs, is necessary to keep 333 coastal hypoxic zones below 5000 km<sup>2</sup> in Chinese coastal waters. Therefore, strict controls on 334 335 atmospheric emissions in China would have a substantial impact on the development of hypoxia in coastal waters. 336

337

### 338 Model limitations

While we took into consideration increase in N deposition due to both climate change (e.g., precipitation) and emission regulation, we do not include changes in riverine loads to predict its effects on future changes of hypoxic areas. Thus, our results might overlook the cumulative or competing effects of these parameters. Moreover, as highlighted before, COOLBEANS assumes that all nutrients will be assimilated and transferred to organic carbon. However, changes in nutrient loading may affect nutrient dynamics, plankton communities and the food web, which all play a role in coastal hypoxia formation.<sup>27</sup> While the efficiency of

### Manuscript accepted in Environmental Science & Technology doi: <u>10.1021/acs.est.0c00706</u>

COOLBEANS to reproduce observational data was previously demonstrated (Nash-Sutcliffee 346 efficiency = 0.71), <sup>93</sup> the model does not consider seasonal and inter-annual physicochemical 347 variability of the coastal environment but rather a yearly-averaged nitrogen deposition, primary 348 349 productivity, carbon fluxes and biochemical oxygen demand. Thus, some discrepancy can be expected when comparing to field data that are often representing a snapshot in time. Our 350 351 results should be considered as a first-order approximation for potential future trend in hypoxia under certain emissions scenarios, and therefore we focussed the discussion on relative change 352 353 in the proportion of hypoxia over the biochemically active regions of COSCAT.

354

355 Atmospheric N deposition is a non-negligible source of nutrient supply from the land to coastal 356 Chinese waters. Here, we quantified the impact of atmospheric N deposition on the extent of 357 hypoxia in the coastal marine environment and highlighted regional differences in nutrient load 358 sensitivities. This should serve as critical information when looking at N emissions control in 359 nearby cities to avert the expansion of coastal hypoxia in China. Regions that are especially 360 sensitive to N deposition, such as the South China Sea, should be prioritized. Implementing 361 targets and emissions regulations in China is crucial to improve the water quality in coastal 362 ecosystems.

363

### 364 Acknowledgement

Yau Yu Yan was partly funded by the Theme-based Research Scheme (T21-602/16-R) of the Hong Kong Research Grants Council co-awarded to DMB and the Stephen S.F. Hui Trust Fund awarded to BT. We thank Dr. Gao for providing the data of future changes in nitrogen deposition. We also thank John Doherty for proofreading. Data generated for this study are available in the supplementary material.

# 371 Supporting Information Available

- 372 Description of the COOLBEANS model; Input used to run the COOLBEANS model;
- 373 Change of total deposition; Modelled change in hypoxic area; Modelled rate of change of
- 374 hypoxia
- 375

# 376 **References**

- Galloway, J. N.; Winiwarter, W.; Leip, A.; Leach, A. M.; Bleeker, A.; Erisman, J. W.
  Nitrogen Footprints: Past, Present and Future. *Environ. Res. Lett.* 2014, *9* (11),
  115003. https://doi.org/10.1088/1748-9326/9/11/115003.
- Kanakidou, M.; Myriokefalitakis, S.; Daskalakis, N.; Fanourgakis, G.; Nenes, A.;
  Baker, A. R.; Tsigaridis, K.; Mihalopoulos, N. Past, Present, and Future Atmospheric
  Nitrogen Deposition. J. Atmos. Sci. 2016, 73 (5), 2039–2047.
- 383 https://doi.org/10.1175/JAS-D-15-0278.1.
- 384 (3) Dentener, F.; Drevet, J.; Lamarque, J. F.; Bey, I.; Eickhout, B.; Fiore, A. M.; 385 Hauglustaine, D.; Horowitz, L. W.; Krol, M.; Kulshrestha, U. C.; Lawrence, M 386 Galv-Lacaux, C.; Rast, S.; Shindell, D.; Stevenson, D.; Van Noije, T.; Atherton, C. 387 Bell, N.; Bergman, D.; Butler, T.; Cofala, J.; Collins, B.; Doherty, R.; Ellingsen, K.; 388 Galloway, J.; Gauss, Michael.; Montanaro, V.; Müller, J. F.; Pitari, G.; Rodriguez, J. 389 Sanderson, M.; Solmon, F.; Strahan, S.; Schultz, M.; Sudo, K.; Szopa, S.; Wild, O. 390 Nitrogen and Sulfur Deposition on Regional and Global Scales: A Multimodel 391 Evaluation. Global Biogeochem. Cycles 2006, 20 (4), GB4003.
- 392 https://doi.org/10.1029/2005GB002672.
- Graneli, E.; Wallstrom, K.; Larsson, U.; Graneli, W.; Elmgren, R. Nutrient Limitation
  of Primary Production in the Baltic Sea Area. *Ambio* 1990, *19* (3), 142–151.
- Howarth, R. W.; Marino, R. Nitrogen as the Limiting Nutrient for Eutrophication in
  Coastal Marine Ecosystems: Evolving Views over Three Decades. *Limnol. Oceanogr.* **2006**, *51* (1part2), 364–376. https://doi.org/10.4319/lo.2006.51.1\_part\_2.0364.
- 398 (6) Voss, M.; Bange, H. W.; Dippner, J. W.; Middelburg, J. J.; Montoya, J. P.; Ward, B.
  399 The Marine Nitrogen Cycle : Recent Discoveries , Uncertainties and the Potential
  400 Relevance of Climate Change The Marine Nitrogen Cycle : Recent Discoveries ,
  401 Uncertainties and the Potential Relevance of Climate Change. *Phil Trans R Soc B*402 **2013**, *368* (1621).
- 403 (7) Paerl, H. W. Coastal Eutrophication and Harmful Algal Blooms: Importance of
  404 Atmospheric Deposition and Groundwater as "new" Nitrogen and Other Nutrient
  405 Sources. *Limnol. Oceanogr.* 1997, 42 (5), 1154–1165.
- 406 (8) Galloway, J. N.; Dentener, F. J.; Capone, D. G.; Boyer, E. W.; Howarth, R. W.;
  407 Seitzinger, S. P.; Asner, G. P.; Cleveland, C. C.; Green, P. A.; Holland, E. A.; Karl, D.
  408 M.; Michaels, A. F.; Porter, J. H.; Townsend, A. R.; Vörösmarty, C. J. Nitrogen
  409 Cycles: Past, Present, and Future. *Biogeochemistry* 2004, 70 (2), 153–226.
- 410 https://doi.org/10.1007/s10533-004-0370-0.
- 411 (9) Troost, T. A.; Blaas, M.; Los, F. J. The Role of Atmospheric Deposition in the
  412 Eutrophication of the North Sea: A Model Analysis. J. Mar. Syst. 2013, 125, 101–112.
  413 https://doi.org/10.1016/j.jmarsys.2012.10.005.
- 414 (10) Thibodeau, B.; Helie, J.; Lehmann, M. F. Variations of the Nitrate Isotopic

- 415 Composition in the St. Lawrence River Caused by Seasonal Changes in Atmospheric
  416 Nitrogen Inputs. *Biogeochemistry* 2013, *115*, 287–298.
- 417 https://doi.org/10.1007/s10533-013-9834-4.
- 418 (11) Gao, Y.; Zhou, F.; Ciais, P.; Miao, C.; Yang, T.; Jia, Y.; Zhou, X.; Klaus, B.-B.; Yu,
  419 G.; Yang, T. Human Activities Aggravate Nitrogen Deposition Pollution to Inland
- 420 Water over China. *Natl. Sci. Rev.* **2019**, No. June, 1–11.
- 421 https://doi.org/10.1093/nsr/nwz073.
- 422 (12) Zhang, Y.; Liu, C.; Liu, X.; Xu, W.; Wen, Z. Atmospheric Nitrogen Deposition around
  423 the Dongting Lake, China. *Atmos. Environ.* 2019, 207 (March), 197–204.
  424 https://doi.org/10.1016/j.atmosenv.2019.03.034.
- 425 (13) Dai, M.; Cai, P.; Zhai, W.; Tang, T.; Wang, L.; Cai, W.-J.; Wang, B.; Yuan, L.; Guo,
  426 X. Oxygen Depletion in the Upper Reach of the Pearl River Estuary during a Winter
  427 Drought. *Mar. Chem.* 2006, *102* (1–2), 159–169.
  428 https://doi.org/10.1016/i.morgham.2005.00.020
- 428 https://doi.org/10.1016/j.marchem.2005.09.020.
- 429 (14) Da, F.; Friedrichs, M. A. M.; St-Laurent, P. Impacts of Atmospheric Nitrogen
  430 Deposition and Coastal Nitrogen Fluxes on Oxygen Concentrations in Chesapeake
  431 Bay. J. Geophys. Res. Ocean. 2018, 123 (7), 5004–5025.
- 432 https://doi.org/10.1029/2018JC014009.
- 433 (15) Zhang, H.; Li, S. Effects of Physical and Biochemical Processes on the Dissolved
  434 Oxygen Budget for the Pearl River Estuary during Summer. *J. Mar. Syst.* 2010, 79 (1–
  435 2), 65–88. https://doi.org/10.1016/j.jmarsys.2009.07.002.
- (16) Duce, R. A.; LaRoche, J.; Altieri, K.; Arrigo, K. R.; Baker, A. R.; Capone, D. G.;
  (16) Duce, R. A.; LaRoche, J.; Altieri, K.; Arrigo, K. R.; Baker, A. R.; Capone, D. G.;
  (17) Cornell, S.; Dentener, F.; Galloway, J.; Ganeshram, R. S.; Geider, R. J.; Jickells,
  (18) T.Kuypers, M. M.; Langlois, R.; Liss, P. S.; Liu, S. M.; Middelburg, J. J.; Moore, C.
  (19) M.; Nickovic, S.; Oschlies, A.; Pedersen, T.; Prospero, J.; Schlitzer, R.; Seitzinger, S.;
  (10) Sorensen, L. L.; Uematsu, M.; Ulloa, O.; Voss, M.; Ward, B.; Zamora, L. Impacts of
  (10) Atmospheric Anthropogenic Nitrogen on the Open Ocean. *Science (80-. ).* 2008, 320
  (10) Sorensen, 1120 (asignese 1150260)
- 442 (5878), 893–897. https://doi.org/10.1126/science.1150369.
- 443 (17) St-Laurent, P.; Friedrichs, M. A. M.; Najjar, R. G.; Martins, D. K.; Herrmann, M.;
  444 Miller, S. K.; Wilkin, J. Impacts of Atmospheric Nitrogen Deposition on Surface
  445 Waters of the Western North Atlantic Mitigated by Multiple Feedbacks. *J. Geophys.*446 *Res. Ocean.* 2017, *122* (11), 8406–8426. https://doi.org/10.1002/2017JC013072.
- 447 (18) Jickells, T. D.; Buitenhuis, E.; Altieri, K.; Baker, A. R.; Capone, D.; Duce, R. A.;
  448 Dentener, F.; Fennel, K.; Kanakidou, M.; LaRoche, J.; Lee, K; Liss, P.; Middelburg, J.
  449 J.; Moore, J. K.; Okin, G.; Oschlies, A.; Sarin, M.; Seitzinger, S.; Sharples, J.; Singh,
  450 A.; Suntharalingam, P.; Uematsu, M.; Zamora, L. M. A Reevaluation of the Magnitude
  451 and Impacts of Anthropogenic Atmospheric Nitrogen Inputs on the Ocean. *Global*452 *Biogeochem. Cycles* 2017, *31* (2), 289–305. https://doi.org/10.1002/2016GB005586.
- 453 (19) Liu, L.; Zhang, X.; Wang, S.; Lu, X.; Ouyang, X. A Review of Spatial Variation of
  454 Inorganic Nitrogen (N) Wet Deposition in China. *PLoS ONE*. 2016, pp 1–17.
  455 https://doi.org/10.1371/journal.pone.0146051.
- 456 (20) Kim, T. W.; Lee, K.; Duce, R.; Liss, P. Impact of Atmospheric Nitrogen Deposition on
  457 Phytoplankton Productivity in the South China Sea. *Geophys. Res. Lett.* 2014, 41 (9),
  458 3156–3162. https://doi.org/10.1002/2014GL059665.
- 459 (21) Nixon, S. W. Coastal Marine Eutrophication: A Definition, Social Causes, and Future
  460 Concerns. *Ophelia* 1995, *41* (1), 199–219.
- 461 https://doi.org/10.1080/00785236.1995.10422044.
- 462 (22) Nixon, S. W. Eutrophication and the Macroscope. *Hydrobiologia* 2009, 629 (1), 5–19.
   463 https://doi.org/10.1007/s10750-009-9759-z.
- 464 (23) Erisman, J. W.; Galloway, J.; Seitzinger, S.; Bleeker, A.; Butterbach-Bahl, K. Reactive

165		Nitrogen in the Environment and Its Effect on Climate Change, Curr Onio, Environ
465 466		Nitrogen in the Environment and Its Effect on Climate Change. <i>Curr. Opin. Environ. Sustain.</i> <b>2011</b> , <i>3</i> (5), 281–290. https://doi.org/10.1016/j.cosust.2011.08.012.
400 467	(24)	Cole, J. J.; Findlay, S.; Pace, M. L. Bacterial Production in Fresh and Saltwater
468	(27)	Ecosystem: A Cross-System Overview. <i>Mar. Ecol Prog. Ser.</i> <b>1988</b> , <i>43</i> , 1–10.
469	(25)	Diaz, R. J.; Rosenberg, R. Marine Benthic Hypoxia : A Review of Its Ecological
470	(23)	Effects and the Behavioural Responses of Benthic Macrofauna. <i>Oceanogr. Mar. Biol.</i>
471		<i>an Annu. Rev.</i> <b>1995</b> , <i>33</i> , 245–303. https://doi.org/10.1021/je700185m.
472	(26)	Fennel, K.; Levin, J.; Moisan, J.; Wilkin, J.; O'Reilly, J.; Haidvogel, D. Nitrogen
473	(=0)	Cycling in the Middle Atlantic Bight: Results from a Three-Dimensional Model and
474		Implications for the North Atlantic Nitrogen Budget. <i>Global Biogeochem. Cycles</i>
475		<b>2006</b> , 20 (3), GB3007. https://doi.org/10.1029/2005gb002456.
476	(27)	Diaz, R. J.; Rosenberg, R. Spreading Dead Zones and Consequences for Marine
477	~ /	Ecosystems. Science (80). 2008, 321 (5891), 926–929.
478		https://doi.org/10.1126/science.1156401.
479	(28)	Thibodeau, B.; de Vernal, A.; Mucci, A. Recent Eutrophication and Consequent
480		Hypoxia in the Bottom Waters of the Lower St. Lawrence Estuary:
481		Micropaleontological and Geochemical Evidence. Mar. Geol. 2006, 231 (1-4), 37-50.
482		https://doi.org/10.1016/j.margeo.2006.05.010.
483	(29)	Collos, Y.; Döhler, G.; Biermann, I. Production of Dissolved Organic Nitrogen during
484		Uptake of Nitrate by Synedra Planctonica: Implications for Estimates of New
485		Production in the Oceans. J. Plankton Res. 1992, 14 (8), 1025–1029.
486		https://doi.org/10.1093/plankt/14.8.1025.
487	(30)	Seitzinger, S. P.; Harrison, J. A.; Böhlke, J. K.; Bouwman, A. F.; Lowrance, R.;
488		Tobias, C.; Drecht, G. Van. Denitrification across Landscapes and Waterscapes : A
489		Synthesis. Ecol. Appl. 2006, 16 (6), 2064–2090.
490	(31)	Galloway, J. N.; Erisman, J. W.; Howarth, R. W.; Cowling, E. B.; Aber, J. D.; Cosby,
491		B. J.; Seitzinger, S. P. The Nitrogen Cascade. <i>Bioscience</i> 2003, 53 (4), 341–356.
492		https://doi.org/10.1641/0006-3568(2003)053[0341:tnc]2.0.co;2.
493	(32)	Diaz, R. J. Overview of Hypoxia around the World. J. Environ. Qual. 2001, 30 (2),
494		275–281.
495	(33)	Pena, M. A.; Katsev, S.; Oguz, T.; Gilbert, D. Modeling Dissolved Oxygen Dynamics
496	(2.1)	and Hypoxia. <i>Biogeosciences</i> <b>2010</b> , <i>7</i> , 933–957.
497	(34)	Conley, D. J.; Destouni, G.; Gustafsson, B. O. G.; Hietanen, S.; Kortekaas, M.; Kuosa,
498 499		H.; Meier, H. E. M.; Norkko, A. L. F.; Gertrud, N. U.; Rosenberg, R.; Savchuk, O. P.;
499 500		Slomp, C. P.; Voss, M. Hypoxia-Related Processes in the Baltic Sea. <i>Environ. Sci.</i> <i>Tachnol.</i> <b>2000</b> , <i>43</i> (10), 3412, 3420
500 501	(35)	<i>Technol.</i> <b>2009</b> , <i>43</i> (10), 3412–3420. Dodds, W. K. Nutrients and the "Dead Zone": The Link between Nutrient Ratios and
502	(33)	Dissolved Oxygen in the Northern Gulf of Mexico. <i>Ecol. Soc. Am.</i> <b>2006</b> , <i>14</i> (1), 3–4.
502 503	(36)	Murphy, R. R.; Kemp, W. M.; Ball, W. P. Long-Term Trends in Chesapeake Bay
503 504	(30)	Seasonal Hypoxia, Stratification, and Nutrient Loading. <i>Estuaries and Coasts</i> <b>2011</b> , <i>34</i>
505		(6), 1293–1309. https://doi.org/10.1007/s12237-011-9413-7.
506	(37)	Gilbert, D.; Sundby, B.; Gobeil, C.; Mucci, A.; Tremblay, GH. A Seventy-Two-Year
507	(37)	Record of Diminishing Deep-Water Oxygen in the St. Lawrence Estuary: The
508		Northwest Atlantic Connection. Am. Soc. Linmnology Oceanogr. 2005, 50 (5), 1654–
509		1666.
510	(38)	Huang, L.; Smith, M. D.; Craig, J. K. Quantifying the Economic Effects of Hypoxia on
511	()	a Fishery for Brown Shrimp Farfantepenaeus Aztecus. Mar. Coast. Fish. 2010, 2 (1),
512		232–248. https://doi.org/10.1577/c09-048.1.
513	(39)	Rabotyagov, S. S.; Kling, C. L.; Gassman, P. W.; Rabalais, N. N.; Turner, R. E. The
514	. ,	Economics of Dead Zones: Causes, Impacts, Policy Challenges, and a Model of the
		-

- 515
   Gulf of Mexico Hypoxic Zone. *Rev. Environ. Econ. Policy* 2014, 8 (1), 58–79.

   516
   https://doi.org/10.1093/reep/ret024.
- 517 (40) Breitburg, D.; Levin, L. A.; Oschlies, A.; Grégoire, M.; Chavez, F. P.; Conley, D. J.;
  518 Garçon, V.; Gilbert, D.; Gutiérrez, D.; Isensee, K.; Jacinto, G. S.; Limburg, K. E.;
- 519 Montes, I.; Naqvi, S. W.A.; Pitcher, G. C.; Rabalais, N. N.; Roman, M. R.; Rose, K.
- 520 A.; Seibel, B. A.; Telszewski, M.; Yasuhara, M.; Zhang, J. Declining Oxygen in the
- 521 Global Ocean and Coastal Waters. *Science* (80-. ). **2018**, 359 (6371), 46.
- 522 https://doi.org/10.1126/science.aam7240.
- 523 (41) Zhang, J.; Gilbert, D.; Gooday, A. J.; Levin, L.; Naqvi, S. W. A.; Middelburg, J. J.;
  524 Scranton, M.; Ekau, W.; Peña, A.; Dewitte, B.; Oguz, T.; Monteiro, P. M.S.; Urban,
- 525 E.; Rabalais, N. N.; Ittekkot, V.; Kemp, W. M.; Ulloa, O.; Elmgren, R.; Escobar526 Briones, E.; Van Der Plas, A. K. Natural and Human-Induced Hypoxia and
  527 Consequences for Coastal Areas: Synthesis and Future Development. *Biogeosciences*
- 528 **2010**, 7 (5), 1443–1467. https://doi.org/10.5194/bg-7-1443-2010.
- (42) Phoenix, G. K.; Emmett, B. A.; Britton, A. J.; Caporn, S. J. M.; Dise, N. B.; Helliwell,
  R.; Jones, L.; Leake, J. R.; Leith, I. D.; Sheppard, L. J.; Sowerby, A.; Pilkington, M.
  G.; Rowe, E. C.; Ashmore, M. R.; Power, S. A. Impacts of Atmospheric Nitrogen
  Deposition: Responses of Multiple Plant and Soil Parameters across Contrasting
- 533Ecosystems in Long-Term Field Experiments. Glob. Chang. Biol. 2012, 18 (4), 1197–5341215. https://doi.org/10.1111/j.1365-2486.2011.02590.x.
- 535 (43) Howarth, R. W.; Sharpley, A.; Walker, D. Sources of Nutrient Pollution to Coastal
  536 Waters in the United States : Implications for Achieving Coastal Water Quality Goals.
  537 *Estuaries* 2002, 25 (4), 656–676.
- 538 (44) Zhang, J.; Liu, S. M.; Ren, J. L.; Wu, Y.; Zhang, G. L. Nutrient Gradients from the
  539 Eutrophic Changjiang (Yangtze River) Estuary to the Oligotrophic Kuroshio Waters
  540 and Re-Evaluation of Budgets for the East China Sea Shelf. *Prog. Oceanogr.* 2007, 74
  541 (4), 449–478. https://doi.org/10.1016/j.pocean.2007.04.019.
- 542 (45) Lu, Z.; Gan, J.; Dai, M.; Liu, H.; Zhao, X. Joint Effects of Extrinsic Biophysical
  543 Fluxes and Intrinsic Hydrodynamics on the Formation of Hypoxia West off the Pearl
  544 River Estuary. J. Geophys. Res. Ocean. 2018, 123 (9), 6241–6259.
  545 https://doi.org/10.1029/2018JC014199.
- (46) Qian, W.; Gan, J.; Liu, J.; He, B.; Lu, Z.; Guo, X.; Wang, D.; Guo, L.; Huang, T.; Dai,
  M. Current Status of Emerging Hypoxia in a Eutrophic Estuary: The Lower Reach of
  the Pearl River Estuary, China. *Estuar. Coast. Shelf Sci.* 2018, 205, 58–67.
  https://doi.org/10.1016/j.ecss.2018.03.004.
- 550 (47) Tang, D.; Di, B.; Wei, G.; Ni, I.; Oh, I. S.; Wang, S. Spatial, Seasonal and Species
  551 Variations of Harmful Algal Blooms in the South Yellow Sea and East China Sea.
  552 *Hydrobiologia* 2006, 568, 245–253. https://doi.org/10.1007/s10750-006-0108-1.
- Hu, C.; Li, D.; Chen, C.; Ge, J.; Muller-Karger, F. E.; Liu, J.; Yu, F.; He, M. X. On the
  Recurrent Ulva Prolifera Blooms in the Yellow Sea and East China Sea. J. Geophys. *Res. Ocean.* 2010, 115 (5). https://doi.org/10.1029/2009JC005561.
- (49) Zhu, Z. Y.; Wu, H.; Liu, S. M.; Wu, Y.; Huang, D. J.; Zhang, J.; Zhang, G. Sen.
  Hypoxia off the Changjiang (Yangtze River) Estuary and in the Adjacent East China
  Sea: Quantitative Approaches to Estimating the Tidal Impact and Nutrient
  Regeneration. *Mar. Pollut. Bull.* 2017, *125* (1–2), 103–114.
  https://doi.org/10.1016/i.marpolbul.2017.07.029
- 560 https://doi.org/10.1016/j.marpolbul.2017.07.029.
- 561 (50) Chen, C. C.; Gong, G. C.; Shiah, F. K. Hypoxia in the East China Sea: One of the
  562 Largest Coastal Low-Oxygen Areas in the World. *Mar. Environ. Res.* 2007, 64 (4),
  563 399–408. https://doi.org/10.1016/j.marenvres.2007.01.007.
- 564 (51) Zhu, Y.; Mccowan, A.; Cook, P. L. M. Effects of Changes in Nutrient Loading and

- Composition on Hypoxia Dynamics and Internal Nutrient Cycling of a Stratified
  Coastal Lagoon. *Biogeosciences* 2017, *14*, 4423–4433.
  Archana, A.; Thibodeau, B.; Geeraert, N.; Xu, M. N.; Kao, S. J.; Baker, D. M.
  Nitrogen Sources and Cycling Revealed by Dual Isotopes of Nitrate in a Complex
  Urbanized Environment. *Water Res.* 2018, *142*, 459–470.
  https://doi.org/10.1016/j.watres.2018.06.004.
- 571 (53) Archana, A.; Li, L.; Shuh-Ji, K.; Thibodeau, B.; Baker, D. M. Variations in Nitrate
  572 Isotope Composition of Wastewater Effluents by Treatment Type in Hong Kong. *Mar.*573 *Pollut. Bull.* 2016, *111* (1–2), 143–152.
- 574 https://doi.org/10.1016/j.marpolbul.2016.07.019.
- 575 (54) Zhang, Y.; Yu, Q.; Ma, W.; Chen, L. Atmospheric Deposition of Inorganic Nitrogen to
  576 the Eastern China Seas and Its Implications to Marine Biogeochemistry. J. Geophys.
  577 Res. Atmos. 2010, 115 (11), 1–10. https://doi.org/10.1029/2009JD012814.
- 578 (55) Yang, J. Y. T.; Hsu, S. C.; Dai, M. H.; Hsiao, S. S. Y.; Kao, S. J. Isotopic Composition
  579 of Water-Soluble Nitrate in Bulk Atmospheric Deposition at Dongsha Island: Sources
  580 and Implications of External N Supply to the Northern South China Sea.
- 581 *Biogeosciences* **2014**, *11* (7), 1833–1846. https://doi.org/10.5194/bg-11-1833-2014.
- 582 (56) De Leeuw, G.; Spokes, L.; Jickells, T.; Skjøth, C. A.; Hertel, O.; Vignati, E.; Tamm,
  583 S.; Schulz, M.; Sørensen, L. L.; Pedersen, B.; Klein, L.; Heinke Schlünzen, K.
- 584Atmospheric Nitrogen Inputs into the North Sea: Effect on Productivity. Cont. Shelf585Res. 2003, 23 (17–19), 1743–1755. https://doi.org/10.1016/j.csr.2003.06.011.
- 586 (57) Wang, M.; Ma, L.; Strokal, M.; Ma, W.; Liu, X.; Kroeze, C. Hotspots for Nitrogen and
  587 Phosphorus Losses from Food Production in China: A County-Scale Analysis.
  588 *Environ. Sci. Technol.* 2018, 52 (10), 5782–5791.
  589 https://doi.org/10.1021/acs.est.7b06138.
- 589 (58) Liu, X.; Duan, L.; Mo, J.; Du, E.; Shen, J.; Lu, X.; Zhang, Y.; Zhou, X.; He, C.;
- 591 Zhang, F. Nitrogen Deposition and Its Ecological Impact in China: An Overview.
  592 *Environ. Pollut.* 2011, 159 (10), 2251–2264.
- 593 https://doi.org/10.1016/j.envpol.2010.08.002.
- 594 (59) Zhao, B.; Wang, S. X.; Liu, H.; Xu, J. Y.; Fu, K.; Klimont, Z.; Hao, J. M.; He, K. B.;
  595 Cofala, J.; Amann, M. NOx Emissions in China: Historical Trends and Future
  596 Perspectives. *Atmos. Chem. Phys.* 2013, *13* (19), 9869–9897.
  597 https://doi.org/10.5194/acp-13-9869-2013.
- 598 (60) Xie, Y.; Xiong, Z.; Xing, G.; Yan, X.; Shi, S.; Sun, G.; Zhu, Z. Source of Nitrogen in
  599 Wet Deposition to a Rice Agroecosystem at Tai Lake Region. *Atmos. Environ.* 2008,
  600 42 (21), 5182–5192. https://doi.org/10.1016/j.atmosenv.2008.03.008.
- (61) Xu, W.; Zhao, Y.; Liu, X.; Dore, A. J.; Zhang, L.; Liu, L.; Cheng, M. Atmospheric
  Nitrogen Deposition in the Yangtze River Basin: Spatial Pattern and Source
  Attribution. *Environ. Pollut.* 2018, 232, 546–555.
- 604 https://doi.org/10.1016/j.envpol.2017.09.086.
- (62) Zhang, J.; Zhang, G. S.; Bi, Y. F.; Liu, S. M. Nitrogen Species in Rainwater and
  Aerosols of the Yellow and East China Seas: Effects of the East Asian Monsoon and
  Anthropogenic Emissions and Relevance for the NW Pacific Ocean. *Global Biogeochem. Cycles* 2011, 25 (3), 1–15. https://doi.org/10.1029/2010GB003896.
- 609 (63) Wang, X.; Wu, Z.; Shao, M.; Fang, Y.; Zhang, L.; Chen, F.; Chan, P.; Fan, Q.; Wang,
- 610 Q.; Zhu, S.; Bao, R. Atmospheric Nitrogen Deposition to Forest and Estuary
- Environments in the Pearl River Delta Region, Southern China. *Tellus B* 2013, 65, 1–
  13. https://doi.org/10.3402/tellusb.v65i0.20480.
- 613 (64) Uno, I.; Uematsu, M.; Hara, Y.; He, Y. J.; Ohara, T.; Mori, A.; Kamaya, T.; Murano,
  614 K.; Sadanaga, Y.; Bandow, H. Numerical Study of the Atmospheric Input of

- 615 Anthropogenic Total Nitrate to the Marginal Seas in the Western North Pacific Region. Geophys. Res. Lett. 2007, 34 (17). https://doi.org/10.1029/2007GL030338. 616 Zhao, Y.; Zhang, L.; Pan, Y.; Wang, Y.; Paulot, F.; Henze, D. K. Atmospheric 617 (65) 618 Nitrogen Deposition to the Northwestern Pacific: Seasonal Variation and Source 619 Attribution. Atmos. Chem. Phys. 2015, 15 (18), 10905–10924. 620 https://doi.org/10.5194/acp-15-10905-2015. 621 Kim, T. W.; Lee, K.; Najjar, R. G.; Jeong, H. D.; Jeong, H. J. Increasing N Abundance (66) 622 in the Northwestern Pacific Ocean due to Atmospheric Nitrogen Deposition. Science 623 (80-.). 2011, 334 (6055), 505–509. https://doi.org/10.1126/science.1206583. 624 Cui, S.; Shi, Y.; Groffman, P. M.; Schlesinger, W. H.; Zhu, Y. Centennial-Scale (67) 625 Analysis of the Creation and Fate of Reactive Nitrogen in China (1910-2010). Proc. 626 Natl. Acad. Sci. U. S. A. 2013, 110 (6), 2052-2057. 627 https://doi.org/10.1073/pnas.1221638110. Liu, X.; Zhang, Y.; Han, W.; Tang, A.; Shen, J.; Cui, Z.; Vitousek, P.; Erisman, J. W.; 628 (68) Goulding, K.; Christie, P.; Fangmeier, A.; Zhang, F. Enhanced Nitrogen Deposition 629 over China. Nature 2013, 494 (7438), 459–462. https://doi.org/10.1038/nature11917. 630 631 (69) Kim, T. H.; Kim, G. Changes in Seawater N: P Ratios in the Northwestern Pacific 632 Ocean in Response to Increasing Atmospheric N Deposition: Results from the East (Japan) Sea. Limnol. Oceanogr. 2013, 58 (6), 1907–1914. 633 634 https://doi.org/10.4319/lo.2013.58.6.1907. 635 (70)Grosse, J.; Bombar, D.; Doan, H. N.; Nguyen, L. N.; Voss, M. The Mekong River Plume Fuels Nitrogen Fixation and Determines Phytoplankton Species Distribution in 636 637 the South China Sea during Low- and High-Discharge Season. Limnol. Oceanogr. 638 **2010**, *55* (4), 1668–1680. https://doi.org/10.4319/lo.2010.55.4.1668. Yin, K.; Qian, P. Y.; Wu, M. C. S.; Chen, J. C.; Huang, L.; Song, X.; Jian, W. Shift 639 (71) 640 from P to N Limitation of Phytoplankton Growth across the Pearl River Estuarine 641 Plume during Summer. Mar. Ecol. Prog. Ser. 2001, 221, 17–28. 642 https://doi.org/10.3354/meps221017. 643 (72)Hu, J.; Peng, P.; Jia, G.; Mai, B.; Zhang, G. Distribution and Sources of Organic 644 Carbon, Nitrogen and Their Isotopes in Sediments of the Subtropical Pearl River 645 Estuary and Adjacent Shelf, Southern China. Mar. Chem. 2006, 98 (2-4), 274-285. 646 https://doi.org/10.1016/j.marchem.2005.03.008. 647 Wang, B. D.; Wang, X. L.; Zhan, R. Nutrient Conditions in the Yellow Sea and the (73) 648 East China Sea. Estuar. Coast. Shelf Sci. 2003, 58 (1), 127-136. 649 https://doi.org/10.1016/S0272-7714(03)00067-2. Kanakidou, M.; Tsigaridis, K.; Mahowald, N.; Baker, A. R.; Hunter, K. A.; Sarin, M.; 650 (74)651 Duce, R. A.; Zhu, T.; Liss, P. S.; Zamora, L. M.; Dentener, F. J.; Benitez-Nelson, C.; Okin, G. S.; Uematsu, M.; Prospero, J. M. Atmospheric Fluxes of Organic N and P to 652 653 the Global Ocean. Global Biogeochem. Cycles 2012, 26 (3), 1–12. 654 https://doi.org/10.1029/2011gb004277. Nydahl, A.; Panigrahi, S.; Wikner, J. Increased Microbial Activity in a Warmer and 655 (75)656 Wetter Climate Enhances the Risk of Coastal Hypoxia. FEMS Microbiol. Ecol. 2013, 85 (2), 338-347. https://doi.org/10.1111/1574-6941.12123. 657 Veraart, A. J.; de Klein, J. J. M.; Scheffer, M. Warming Can Boost Denitrification 658 (76)659 Disproportionately due to Altered Oxygen Dynamics. *PLoS One* **2011**, 6 (3), 2–7. https://doi.org/10.1371/journal.pone.0018508. 660 Kryza, M.; Werner, M.; Dore, A. J.; BłaŚ, M.; Sobik, M. The Role of Annual 661 (77) 662 Circulation and Precipitation on National Scale Deposition of Atmospheric Sulphur 663 and Nitrogen Compounds. J. Environ. Manage. 2012, 109 (x), 70-79.
- 664 https://doi.org/10.1016/j.jenvman.2012.04.048.

- (78) Wałaszek, K.; Kryza, M.; Dore, A. J. The Impact of Precipitation on Wet Deposition
  of Sulphur and Nitrogen Compounds. *Ecol. Chem. Eng. S* 2013, 20 (4), 733–745.
  https://doi.org/10.2478/eces-2013-0051.
- (79) Chong-Hai, X.; Ying, X. The Projection of Temperature and Precipitation over China
  under RCP Scenarios Using a CMIP5 Multi-Model Ensemble. *Atmos. Ocean. Sci. Lett.*(67) 2012, 5 (6), 527–533. https://doi.org/10.1080/16742834.2012.11447042.
- (80) Wang, L.; Chen, W. A CMIP5 Multimodel Projection of Future Temperature,
  Precipitation, and Climatological Drought in China. *Int. J. Climatol.* 2014, *34* (6),
  2059–2078. https://doi.org/10.1002/joc.3822.
- 674 (81) Sinha, E.; Michalak, A. M.; Balaji, V. Eutrophication Will Increase during the 21st
  675 Century as a Result of Precipitation Changes. *Science* (80-.). 2017, 357 (6349), 405–
  676 408. https://doi.org/10.1126/science.aan2409.
- 677 (82) Luo, X. S.; Tang, A. H.; Shi, K.; Wu, L. H.; Li, W. Q.; Shi, W. Q.; Shi, X. K.;
  678 Erisman, J. W.; Zhang, F. S.; Liu, X. J. Chinese Coastal Seas Are Facing Heavy
  679 Atmospheric Nitrogen Deposition. *Environ. Res. Lett.* 2014, *9* (9).
  680 https://doi.org/10.1088/1748-9326/9/9/095007.
- (83) Chen, N.; Hong, H.; Huang, Q.; Wu, J. Atmospheric Nitrogen Deposition and Its
  Long-Term Dynamics in a Southeast China Coastal Area. *J. Environ. Manage.* 2011,
  92 (6), 1663–1667. https://doi.org/10.1016/j.jenvman.2011.01.026.
- (84) Taketani, F.; Aita, M. N.; Yamaji, K.; Sekiya, T.; Ikeda, K.; Sasaoka, K.; Hashioka, T.;
  Honda, M. C.; Matsumoto, K.; Kanaya, Y. Seasonal Response of North Western
  Pacific Marine Ecosystems to Deposition of Atmospheric Inorganic Nitrogen
  Compounds from East Asia. *Sci. Rep.* 2018, 8 (1), 1–9.
  https://doi.org/10.1038/s41598-018-27523-w.
- (85) van Vuuren, D. P.; Edmonds, J.; Kainuma, M.; Riahi, K.; Thomson, A.; Hibbard, K.;
  Hurtt, G. C.; Kram, T.; Krey, V.; Lamarque, J. F.; Masui, T.; Meinshausen, M.;
- 691 Nakicenovic, N.; Smith, S. J.; Rose, S. K. The Representative Concentration
- 692Pathways: An Overview. Clim. Change 2011, 109 (1), 5–31.
- 693 https://doi.org/10.1007/s10584-011-0148-z.
- 694 (86) IPCC. *Climate Change 2014: Synthesis Report*; IPCC: Geneva, Switzerland, 2014.
   695 https://doi.org/10.1046/j.1365-2559.2002.1340a.x.
- (87) Clarke, L.; Edmonds, J.; Jacoby, H.; Pitcher, H.; Reilly, J.; Richels, R. Scenarios of
  Greenhouse Gas Emissions and Atmospheric Concentrations. Sub-Report 2.1A of
  Synthesis and Assessment Product 2.1 by the U.S. Climate Change Science Program
  and the Subcommittee on Global Change Research; 2007.
- (88) Wise, M.; Calvin, K.; Thomson, A.; Clarke, L.; Bond-lamberty, B.; Sands, R.; Smith,
  S. J.; Janetos, A.; Edmonds, J. Implications of Limiting CO2 Concentrations for Land
  Use and Energy. *Science* (80-. ). 2009, 1183 (2), 1183–1187.
  https://doi.org/10.1126/science.1168475.
- Riahi, K.; Grübler, A.; Nakicenovic, N. Scenarios of Long-Term Socio-Economic and
  Environmental Development under Climate Stabilization. *Technol. Forecast. Soc. Change* 2007, 74 (7), 887–935. https://doi.org/10.1016/j.techfore.2006.05.026.
- (90) Thomson, A. M.; Calvin, K. V.; Smith, S. J.; Kyle, G. P.; Volke, A.; Patel, P.;
  Delgado-Arias, S.; Bond-Lamberty, B.; Wise, M. A.; Clarke, L. E.; Edmonds, J. A.
  RCP4.5: A Pathway for Stabilization of Radiative Forcing by 2100. *Clim. Change*
- 710 **2011**, *109* (1), 77–94. https://doi.org/10.1007/s10584-011-0151-4.
- 711 (91) Riahi, K.; Rao, S.; Krey, V.; Cho, C.; Chirkov, V.; Fischer, G.; Kindermann, G.;
- Nakicenovic, N.; Rafaj, P. RCP 8.5-A Scenario of Comparatively High Greenhouse
  Gas Emissions. *Clim. Change* 2011, *109* (1), 33–57. https://doi.org/10.1007/s10584011-0149-y.

- 715 (92)Laruelle, G. G.; Dürr, H. H.; Lauerwald, R.; Hartmann, J.; Slomp, C. P.; Goossens, N.; 716 Regnier, P. A. G. Global Multi-Scale Segmentation of Continental and Coastal Waters 717 from the Watersheds to the Continental Margins. Hydrol. Earth Syst. Sci. 2013, 17 (5), 718 2029–2051. https://doi.org/10.5194/hess-17-2029-2013. 719 Reed, D. C.; Harrison, J. A. Linking Nutrient Loading and Oxygen in the Coastal (93)720 Ocean: A New Global Scale Model. Glob. Biogeochem. Cycles 2016, 30 (3), 447-459. 721 https://doi.org/10.1002/2015GB005303.Received. 722 (94)Schlitzer, R. Ocean Data View odv.awi.de. 723 Zhang, J.; Gao, Y.; Ruby Leung, L.; Luo, K.; Liu, H.; Lamarque, J. F.; Fan, J.; Yao, (95) 724 X.; Gao, H.; Nagashima, T. Impacts of Climate Change and Emissions on 725 Atmospheric Oxidized Nitrogen Deposition over East Asia. Atmos. Chem. Phys. 2019, 726 19 (2), 887–900. https://doi.org/10.5194/acp-19-887-2019. Meybeck, M.; Dürr, H. H.; Vörösmarty, C. J. Global Coastal Segmentation and Its 727 (96)728 River Catchment Contributors: A New Look at Land-Ocean Linkage. Global 729 Biogeochem. Cycles 2006, 20 (1), 1–15. https://doi.org/10.1029/2005GB002540. 730 (97) Boudreau, P. A Method-of-Lines Code for Carbon and Nutrient Diagenesis in Aquatic
- Boudreau, P. A Method-of-Lines Code for Carbon and Nutrient Diagenesis in Aquatic
   Sediments. *Comput. Geosci.* 1996, 22 (5), 479–496.
- Wang, Y.; Van Cappellen, P. A Multicomponent Reactive Transport Model of Early
  Diagenesis: Application to Redox Cycling in Coastal Marine Sediments. *Geochim. Cosmochim. Acta* 1996, 60 (16), 2993–3014. https://doi.org/10.1016/00167037(96)00140-8.
- (99) Qi, J. H.; Shi, J. H.; Gao, H. W.; Sun, Z. Atmospheric Dry and Wet Deposition of
  Nitrogen Species and Its Implication for Primary Productivity in Coastal Region of the
  Yellow Sea, China. *Atmos. Environ.* 2013, *81*, 600–608.
  https://doi.org/10.1016/j.atmosenv.2013.08.022.
- (100) Wu, S.-P.; Dai, L.-H.; Wei, Y.; Zhu, H.; Zhang, Y.-J.; Schwab, J. J.; Yuan, C.-S.
  Atmospheric Ammonia Measurements along the Coastal Lines of Southeastern China: Implications for Inorganic Nitrogen Deposition to Coastal Waters. *Atmos. Environ.*2018, 177, 1–11. https://doi.org/10.1016/j.atmosenv.2017.12.040.
- (101) Nakamura, T.; Matsumoto, K.; Uematsu, M. Chemical Characteristics of Aerosols
  Transported from Asia to the East China Sea: An Evaluation of Anthropogenic
  Combined Nitrogen Deposition in Autumn. *Atmos. Environ.* 2005, *39* (9), 1749–1758.
  https://doi.org/10.1016/j.atmosenv.2004.11.037.
- (102) Lehrter, J. C.; Ko, D. S.; And, L. L. L.; Penta, B. Predicted Effects of Climate Change
  on Northern Gulf of Mexico Hypoxia. *Springer International Publishing AG*. 2017, pp
  174–214. https://doi.org/10.1007/978-3-319-54571-4.
- (103) Cabré, A.; Marinov, I.; Leung, S. Consistent Global Responses of Marine Ecosystems
  to Future Climate Change across the IPCC AR5 Earth System Models. *Clim. Dyn.* **2015**, 45 (5–6), 1253–1280. https://doi.org/10.1007/s00382-014-2374-3.
- (104) Yu, G.; Jia, Y.; He, N.; Zhu, J.; Chen, Z.; Wang, Q.; Piao, S.; Liu, X.; He, H.; Guo, X.;
  Wen, Z.; Li, P.; Ding, G.; Goulding, K. Stabilization of Atmospheric Nitrogen
  Deposition in China over the Past Decade. *Nat. Geosci.* 2019, *12* (6), 424–429.
  https://doi.org/10.1038/s41561-019-0352-4.
- (105) Zheng, B.; Tong, D.; Li, M.; Liu, F.; Hong, C.; Geng, G.; Li, H.; Li, X.; Peng, L.; Qi,
  J.; Yan, L.; Zhang, Y.; Zhao, H.; Zheng, Y.; He, K.; Zhang, Q. Trends in China's
  Anthropogenic Emissions since 2010 as the Consequence of Clean Air Actions. *Atmos. Chem. Phys.* 2018, *18* (19), 14095–14111. https://doi.org/10.5194/acp-18-14095-2018.
- (106) Meire, L.; Soetaert, K. E. R.; Meysman, F. J. R. Impact of Global Change on Coastal
  Oxygen Dynamics and Risk of Hypoxia. *Biogeosciences* 2013, *10* (4), 2633–2653.
  https://doi.org/10.5194/bg-10-2633-2013.

- (107) Voss, M.; Bange, H. W.; Dippner, J. W.; Middelburg, J. J.; Montoya, J. P.; Ward, B.
  The Marine Nitrogen Cycle: Recent Discoveries, Uncertaintiesand the Potential
  Relevance of Climate Change. *Philos. Trans. R. Soc. B Biol. Sci.* 2013, *368* (1621).
  https://doi.org/10.1098/rstb.2013.0121.
- 769 (108) Mississippi River/Gulf of Mexico Watershed Nutrient Task Force. 2017 Report to
   770 Congress; 2017.
- 771