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Anthropogenic nitrogen inputs and impacts on oceanic N<sub>2</sub>O fluxes in the northern Indian Ocean: The need for an integrated observation and modelling approach

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**1 ABSTRACT**

2 Anthropogenically-derived nitrogen input to the northern Indian Ocean has  
3 increased significantly in recent decades, based on both observational and model-  
4 derived estimates This external nutrient source is supplied by atmospheric  
5 deposition and riverine fluxes, and has the potential to affect the vulnerable  
6 biogeochemical systems of the Arabian Sea and Bay of Bengal, influencing  
7 productivity and oceanic production of the greenhouse-gas nitrous-oxide (N<sub>2</sub>O). We  
8 summarize current estimates of this external nitrogen source to the northern Indian  
9 Ocean from observations and models, highlight implications for regional marine N<sub>2</sub>O  
10 emissions using model-based analyses, and make recommendations for  
11 measurement and model needs to improve current estimates and future predictions  
12 of this impact. Current observationally-derived estimates of deposition and riverine  
13 nitrogen inputs are limited by sparse measurements and uncertainties on accurate  
14 characterization of nitrogen species composition. Ocean model assessments of the  
15 impact of external nitrogen sources on regional marine N<sub>2</sub>O production in the  
16 northern Indian Ocean estimate potentially significant changes but also have large  
17 associated uncertainties. We recommend an integrated program of basin-wide  
18 measurements combined with high-resolution modeling and more detailed  
19 characterization of nitrogen-cycle process to address these uncertainties and  
20 improve current estimates and predictions.

**21**  
**22**  
**23 1. INTRODUCTION**

24 The 2<sup>nd</sup> International Indian Ocean Expedition program [Hood et al. 2016] was  
25 launched in December 2015 on the occasion of the 50<sup>th</sup> anniversary of the first  
26 International Indian Ocean Expedition program (1959-1965) as a joint effort of the  
27 Scientific Committee on Oceanic Research (SCOR), the Intergovernmental  
28 Oceanographic Commission (IOC) and the Indian Ocean Global Observing System  
29 (IOGOOS). IIOE-2 was set up to advance Indian Ocean initiatives and projects  
30 addressing emerging scientific issues of the Indian Ocean in the 21<sup>st</sup> century. To this  
31 end, a deeper understanding of the biogeochemical and socio-economic feedbacks  
32 associated with ongoing changes in the Indian Ocean is critical to better project and  
33 mitigate the consequences of the anthropogenic activities for the Indian Ocean  
34 region and beyond.

35 The biogeochemical systems of the northern Indian Ocean are vulnerable to  
36 the increasing levels of bioavailable reactive nitrogen derived from human activities  
37 (e.g., fossil-fuel combustion, agriculture) and supplied to the coastal and open ocean  
38 through atmospheric pollution outflow and riverine inputs [von Glasow et al. 2013;  
39 Ramanathan et al. 2005; Duce et al. 2008; Seitzinger et al. 2010]. These external  
40 inputs of reactive nitrogen provide an additional nutrient source to marine  
41 ecosystems and can have significant impacts on regional biological productivity and  
42 the associated generation of greenhouse gases such as nitrous oxide (N<sub>2</sub>O). The  
43 activities of the Joint Group of Experts on the Scientific Aspects of Marine  
44 Environmental Protection (GESAMP) Working Group 38 on the Atmospheric Input of  
45 Chemicals to the Ocean (<http://www.gesamp.org/work/groups/38>) have previously  
46 highlighted the increasing levels of anthropogenically derived nitrogen provided to  
47 global ocean ecosystems via atmospheric deposition and riverine fluxes, and their

48 the potentially complex impacts on these ecosystems (e.g., Duce et al. 2008;  
49 Dentener et al. 2006; Suntharalingam et al. 2012; Jickells et al. 2017). The northern  
50 Indian Ocean, in particular, was noted to be a region subject to high and rapidly  
51 increasing anthropogenic nitrogen deposition [Dentener et al. 2006; Baker et al.  
52 2017], and the resulting impacts on local ocean biogeochemistry are poorly  
53 characterized. This commentary results from GESAMP Working Group 38 activities to  
54 examine these regional impacts in the northern Indian Ocean, and in particular, to  
55 assess implications for oceanic emissions of the greenhouse gas nitrous-oxide ( $N_2O$ );  
56 the Arabian Sea region is a known site of dynamic  $N_2O$  cycling [Bange et al. 2001;  
57 Naqvi et al. 2006, 2010a]. In this study we provide a synthesis of current estimates  
58 of sources of external nitrogen inputs to the northern Indian Ocean, and discuss the  
59 implications for regional oceanic emissions of  $N_2O$  using existing estimates from  
60 global model analyses and a new estimate based on a high-resolution model of the  
61 Indian Ocean. An important aim of this commentary is to highlight much needed  
62 advances in regional observations and model analyses to achieve more reliable  
63 estimates of current and future  $N_2O$  emissions in this region.

64

### 65 **Nitrous oxide in the northern Indian Ocean**

66 Oceanic  $N_2O$  production and consumption pathways display significant sensitivity to  
67 the ambient dissolved oxygen level [Codispoti et al. 2001; Bange et al. 2010]. In well-  
68 oxygenated waters,  $N_2O$  is produced during nitrification processes involving the  
69 microbial oxidation of ammonia to nitrate [Cohen and Gordon, 1979]. Culture  
70 studies indicate enhanced yields associated with decreasing ambient oxygen levels  
71 [Goreau et al. 1980; Frame and Casciotti, 2010; Löscher et al., 2012]. In suboxic and  
72 anoxic waters, denitrification processes dominate, and  $N_2O$  is formed as an  
73 intermediate product of the microbial reduction of nitrate to nitrogen gas during  
74 organic matter remineralization (i.e., incomplete denitrification) [Naqvi et al. 2000].  
75 As ambient oxygen levels approach anoxia,  $N_2O$  can be further reduced to  $N_2$ , thus  
76 denitrification processes also provides a sink of  $N_2O$  at very low oxygen levels. The  
77 turnover of  $N_2O$  at the sub-oxic to oxic interface of low oxygen waters is observed to  
78 be highly dynamic, with gross production and consumption fluxes significantly larger  
79 (~20 times) than the residual net  $N_2O$  yield from incomplete denitrification [Babbin  
80 et al. 2015].

81  $N_2O$  production and the underlying nitrogen cycling mechanisms in the  
82 northern Indian Ocean (defined here as the Indian Ocean region located north of the  
83 Equator) are fundamentally influenced by the unique characteristics of the region's  
84 oceanic circulation and associated biogeochemistry. This region encompasses the  
85 Arabian Sea and the Bay of Bengal and hosts two of the most intense oxygen  
86 minimum zones (OMZs) worldwide [Paulmier and Ruiz-Pino, 2006; Morrison et al.  
87 1999; Bristow et al. 2017]. The extensive Arabian Sea OMZ is a region noted for  
88 intense denitrification [Naqvi et al. 2006; Ward et al. 2009], and a 'hotspot' for  
89 generation of marine  $N_2O$  [Naqvi et al. 2010a]. Production rates of  $N_2O$  here are  
90 hypothesized to reach 10,000 times the ocean average [Codispoti, 2010]. In  
91 contrast, although widespread low oxygen levels have been reported in the Bay of  
92 Bengal [e.g., submicromolar  $O_2$ , Bristow et al. 2017], extensive anoxia and  
93 denitrification have not been detected here, possibly due to the persistence of trace  
94 levels of oxygen that inhibit this process [Bristow et al. 2017]. The large riverine

95 freshwater discharge in the north of the Bay of Bengal from the Ganges outflow, in  
96 conjunction with riverine discharge from the peninsular subcontinent into the south-  
97 west of the Bay of Bengal has resulted in a latitudinal gradient in surface N<sub>2</sub>O  
98 concentrations along the coast of the western Bay of Bengal characterized by  
99 equilibrium or under-saturation in the northwest of the basin and small  
100 supersaturation and emission in the south-west [Rao et al. 2013]. According to Rao  
101 et al. [2013] this complex behavior results from a combination of factors, including  
102 seasonal undersaturation in N<sub>2</sub>O associated with monsoonally-governed variations in  
103 Ganges freshwater outflow, and higher nitrification rates and associated N<sub>2</sub>O  
104 supersaturation in the coastal upwelling regions in the south-west of the Bay of  
105 Bengal.

106 The Asian monsoon system plays an important role in regulating the  
107 temporal variation of the region's N<sub>2</sub>O production and emission to the atmosphere.  
108 Monsoonal winds drive a seasonally reversing upper ocean circulation with  
109 associated changes in seasonal upwelling and regional biological productivity in both  
110 the Arabian Sea and the Bay of Bengal [McCreary et al., 2013; Schott and McCreary,  
111 2001; Madhuratap et al. 1996, and references therein]. This upwelling is most  
112 intense in the NW Arabian Sea and leads to especially high regional N<sub>2</sub>O [De Wilde  
113 and Helder, 1996; Bange et al. 2001]. The Asian monsoon system also accounts for  
114 pronounced rainfall and river discharge in the northern Bay of Bengal, playing an  
115 important role in the hydrography and associated biogeochemistry of the basin.  
116 These freshwater fluxes reduce surface salinity and enhance water column  
117 stratification, yielding lower productivity levels in the Bay of Bengal than in the  
118 neighbouring Arabian Sea [Rao et al. 2013; Singh and Ramesh, 2015 and references  
119 therein].

120 Previous estimates of oceanic N<sub>2</sub>O emission from the northern Indian Ocean  
121 highlight its significant contribution to global fluxes [Law and Owens 1990; Naqvi and  
122 Noronha 1991; Lal and Patra 1998; Bange et al. 2001]. The Arabian Sea, in particular,  
123 is a region of intense oceanic N<sub>2</sub>O production, and is estimated to provide a flux to  
124 the atmosphere of 0.2 – 0.6 Tg N yr<sup>-1</sup> [Naqvi et al., 2010b]. This flux represents 2 – 31  
125 % of global total oceanic N<sub>2</sub>O emissions (N<sub>2</sub>O emissions (1.9 – 12.3 Tg N yr<sup>-1</sup>) from  
126 the open ocean, coastal and estuarine sources [Ciais et al. 2013]. In contrast N<sub>2</sub>O  
127 fluxes to the atmosphere from the Bay of Bengal are considerably lower (0.027 -  
128 0.077 Tg N yr<sup>-1</sup>) [Naqvi et al. 1994; Naqvi et al. 2010a], a result of less pronounced  
129 upwelling and lower productivity levels.

130

### 131 **Vulnerability to climate change and anthropogenic nutrient input**

132 The circulation and biogeochemistry of the northern Indian Ocean basins are  
133 vulnerable to the impacts of climate warming and associated changes; for example,  
134 to (a) ocean deoxygenation and the predicted expansion of ocean OMZs [Stramma et  
135 al. 2010; Naqvi et al. 2010b; Banse et al 2014], and (b) potential increases in the  
136 intensity of the monsoonal circulation and associated upwelling and productivity in  
137 the Arabian Sea [De Castro et al. 2016; Goes et al. 2005]. Such changes could result  
138 in significant increases in oceanic N<sub>2</sub>O emissions from the region [Codispoti, 2010].  
139 These hypotheses are supported by recent model analyses that predict the  
140 deepening and intensification of the Arabian Sea OMZ due to strengthening of the  
141 region's monsoonal winds [Lachkar et al. 2018], and increases in northern Indian

142 Ocean N<sub>2</sub>O production associated with expansion of oceanic low-oxygen regions  
143 under scenarios of 21<sup>st</sup> century warming [Martinez-Rey et al. 2015]. A key  
144 vulnerability of the Bay of Bengal results from the observed widespread low oxygen  
145 levels. The regional biogeochemical system is on the brink of hypoxia, and further  
146 oxygen depletion (via physical or biological forcing) could tip this ocean region into a  
147 regime of denitrification with significant increases in N<sub>2</sub>O and N<sub>2</sub> emission to the  
148 atmosphere [Naqvi et al. 2008; Bristow et al. 2017].

149 N<sub>2</sub>O emissions from the Arabian Sea and the Bay of Bengal could also  
150 respond to the rapidly rising supply of anthropogenically-derived nutrient inputs to  
151 these basins [Naqvi et al. 2010a]. Nutrient supply (of N, P, Si, Fe) to marine  
152 ecosystems has increased significantly over the past century; this increase is  
153 projected to continue in the future due to higher levels of fossil fuel combustion,  
154 agricultural fertilizer application, and dust mobilization through land-use change  
155 [Duce et al., 2008; Seitzinger et al. 2010; Fowler et al. 2013; Sattar et al., 2014].  
156 Previous global-scale analyses of the impact of increasing anthropogenic nitrogen  
157 inputs estimate that highest impacts will occur on ocean ecosystems in coastal  
158 regions downwind of atmospheric pollution outflow and influenced by riverine  
159 discharge [Doney et al. 2007; Krishnamurthy et al. 2007; Seitzinger et al. 2010]. Such  
160 increases in nitrogen inputs are predicted to influence marine productivity, organic  
161 matter export and remineralization, and cause coastal eutrophication [Galloway et  
162 al. 2003; Gruber and Galloway, 2008]. These changes will also have an impact on the  
163 oceanic N<sub>2</sub>O source by influencing the magnitude of regional nitrification and  
164 denitrification fluxes, and by altering the extent of hypoxic and sub-oxic regions  
165 where N<sub>2</sub>O production yields are higher than in the oxic ocean [Duce et al. 2008].  
166 Analyses of the impacts of increases in nutrient deposition on productivity have  
167 highlighted the potential for especially high impacts in the northern Indian Ocean  
168 [Naqvi et al., 2000; 2009; Capone and Hutchins, 2013; Suntharalingam et al. 2012].  
169 This region is also noted to be vulnerable to marine nitrogen cycle feedbacks,  
170 whereby relatively small nitrogen inputs from atmospheric nitrogen deposition could  
171 increase local rates of denitrification and cause large net nitrogen losses [Somes et  
172 al. 2016; Yang and Gruber 2016; Landolfi et al. 2017].

173 Current estimates of the anthropogenic nitrogen supply to the northern  
174 Indian Ocean and its impact on the region's ecosystems and biogeochemistry are  
175 derived from sparse measurements and a limited set of model analyses. In this  
176 commentary, we bring together recent estimates of external nitrogen inputs to the  
177 Arabian Sea and Bay of Bengal from atmospheric deposition and riverine sources  
178 (section 2). We also discuss the implications of this additional nutrient source for  
179 oceanic N<sub>2</sub>O production in the northern Indian Ocean based on (i) a synthesis of  
180 previous global ocean biogeochemistry model studies (section 3.1), and (ii) a newly-  
181 derived estimate of Arabian Sea N<sub>2</sub>O production using a regional high-resolution  
182 model (section 3.2). We discuss key uncertainties in these model estimates (sections  
183 3.3 and 3.4), and highlight the need for a comprehensive and integrated  
184 measurement and modeling effort towards more reliable prediction of future  
185 changes in N<sub>2</sub>O emissions from the northern Indian Ocean (section 4).

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187

188

**189 2. Inputs of externally derived nitrogen to the northern Indian Ocean**

190 There has been an approximately 12-fold increase in the rates of Indian agricultural  
191 nitrogen fertilizer consumption since the 1970s associated with rapid economic  
192 development [Ramesh et al. 2007]. Some part of this increased nitrogen load on  
193 land is transported into the northern Indian Ocean's estuaries and coastal waters  
194 (e.g., Martin et al. [2011]; Singh and Ramesh [2011]; Krishna et al. [2016]) likely  
195 resulting in rapid increases in aquatic organic and inorganic nitrogen levels [Liu et al.  
196 2008]. Based on results from the Global Nutrient Export from WaterSheds (NEWS)  
197 model [Pedde et al. 2017; Seitzinger et al. 2010], we estimate that there was an  
198 ~50% ( $1.22 \text{ Tg N yr}^{-1}$ ) increase in riverine dissolved nitrogen input into the northern  
199 Indian Ocean between 1970-2000, 87% of which was directed into the Bay of Bengal.

200 Atmospheric chemistry models also estimate substantial increases in  
201 atmospheric nitrogen deposition to the northern Indian Ocean over recent decades  
202 [Dentener et al. 2006; Kanakidou et al. 2012, 2016; Lamarque et al. 2013]. This trend  
203 is supported by observations; e.g., since the 1980s,  $\text{NO}_x$  emissions over India have  
204 more than doubled [Garg et al. 2006], and there have been significant increases in  
205 observed wet nitrate deposition at different coastal and open ocean sites [Attri and  
206 Tyagi, 2010; Safai et al., 2004]. Cruise measurements made between 2001-2009 in  
207 the Bay of Bengal show similar trends for aerosol  $\text{NH}_4^+$  concentrations [Srinivas et al.,  
208 2011]. Based on a combination of output from the NCAR-CAM (version 3.5) model  
209 [Lamarque et al., 2011] and the TM4-ECPL model [Kanakidou et al., 2012] models  
210 (see Appendix A for details), we estimate an ~226% ( $1.42 \text{ Tg N m}^{-2}$ ) increase in the  
211 amount of soluble nitrogen being deposited to the northern Indian Ocean between  
212 1850 and 2005, 59% of which went into the Arabian Sea.

213 Table 1 presents estimates of the relative contribution of external (i.e., non-  
214 recycled) soluble nitrogen sources to the present-day northern Indian Ocean. The  
215 sources and calculation methods for these estimates are described in Appendix A.  
216 The inorganic nitrogen inputs are in line with previous estimates derived from  
217 models, including those here used [Dentener et al., 2006; Kanakidou et al., 2012,  
218 2016; Lamarque et al., 2013; Vet et al., 2014] and ground-based observations [Baker  
219 et al., 2017; Singh et al., 2012; Srinivas et al., 2011; Srinivas and Sarin, 2013]. Note  
220 that models and observations are not always directly comparable, as the models do  
221 not represent all processes with sufficient accuracy, and the observations do not  
222 provide comprehensive spatial and temporal coverage. The estimates of Table 1  
223 suggest that recent rapid increases in riverine and atmospheric nitrogen are non-  
224 negligible influences on the external nitrogen fluxes to these ocean basins.

225 For biologically-mediated nitrogen sources, there are very few estimates of  
226 nitrogen ( $\text{N}_2$ ) fixation in the northern Indian Ocean. Direct estimates, made using  
227 isotopically enriched ( $^{15}\text{N}_2$ ) tracers, suggest extremely high rates of  $\text{N}_2$  fixation in the  
228 Arabian Sea during springtime (Table 1, Gandhi et al., 2011). Srinivas and Sarin  
229 [2013] provide indirect estimates of  $\text{N}_2$  fixation rates, derived with respect to  
230 atmospheric deposition inputs and assuming Fe and P limitation in the ocean; these  
231 estimates suggest a large variability in regional  $\text{N}_2$  fixation rates, linked to the  
232 uncertainty in atmospheric deposition fluxes. The role of  $\text{N}_2$  fixation must also,  
233 therefore, be better constrained for a more accurate assessment of anthropogenic  
234 nutrient impacts on the biogeochemistry of the northern Indian Ocean.

235

236

**237 Uncertainties in nitrogen inputs**

238 Data available to validate model estimates for the region are still sparse, particularly  
239 for the Bay of Bengal. For example, we found references to only three cruises that  
240 sampled aerosol water soluble organic nitrogen (WSON) (compiled in Srinivas et al.,  
241 [2011] and Srinivas and Sarin [2013]). Inorganic dry nitrogen deposition is better  
242 constrained, with over 400 individual ship-based observations, but sample sizes are  
243 still insufficient to adequately characterize the north Indian Ocean's high inter-  
244 annual and seasonal variability, and the expected increases in deposition with time  
245 (e.g., more than half of the observations were collected prior to the year 2000).  
246 There are particularly high uncertainties in wet deposition estimates due to small  
247 numbers of observations over the open ocean.

248 As with atmospheric and riverine inputs, observations of  $N_2$  fixation to the  
249 Arabian Sea are also sparse, particularly over the Bay of Bengal, where there have  
250 only been a few reports of rates from *Trichodesmium* [Jyothibabu et al., 2017]. The  
251 relatively low number of observations for the different external nitrogen sources  
252 makes it challenging to validate regional models, resulting in high uncertainties in  
253 descriptions of the mean system state. The paucity of the data, along with rapid and  
254 large-scale increases in anthropogenic nutrient inputs, highlight the need for a more  
255 detailed evaluation of external nutrient inputs to the region.

256

**257 3. Implications for oceanic  $N_2O$  production in the northern Indian Ocean**

258 **3.1 Estimates from global models:** The impacts of increasing levels of  
259 anthropogenically derived atmospheric nitrogen deposition on marine  
260 biogeochemistry and  $N_2O$  production have been investigated by recent ocean  
261 biogeochemistry model analyses (e.g., Suntharalingam et al. [2012], Jickells et al.  
262 [2017], Landolfi et al. [2017]). While these studies estimate relatively modest  
263 increases in oceanic  $N_2O$  production at the global scale (1-3%), they all suggest more  
264 significant regional impacts in regions of high nutrient deposition downwind of  
265 continental outflow in the vicinity of oceanic OMZs. In particular, the northern  
266 Indian Ocean, and specifically the Arabian Sea, is noted as one of the most sensitive  
267 regions to changes in this anthropogenic nutrient input. Table 2 summarizes  
268 estimated changes in  $N_2O$  production in the northern Indian Ocean from the above  
269 three global model analyses. The ocean biogeochemical models include ecosystems  
270 of varying complexity, but all represent marine cycles of carbon, nitrogen,  
271 phosphorus, oxygen and  $N_2O$ . The three model analyses synthesized in Table 2 also  
272 employed different model-derived estimates of atmospheric nitrogen deposition to  
273 the ocean, which differ in some aspects of flux composition (e.g., organic nitrogen  
274 levels, proportion of oxidized and reduced nitrogen species [Jickells et al. 2017]).  
275 These details, and the individual ocean model specifications, are given in Appendix B  
276 and in the individual publications. Despite differences in their representation of  
277 ocean ecosystem processes and nutrient inputs all model analyses estimate  
278 increases in surface biological productivity and organic matter export in response to  
279 increases in nitrogen deposition from the pre-industrial to the present day. These  
280 analyses also estimate proportionately larger changes in  $N_2O$  production in the  
281 northern Indian Ocean than on the global scale (e.g., increases of 1%-10% for the  
282 northern Indian Ocean in comparison to 1%-3% on the global scale). This regional



283 enhancement of N<sub>2</sub>O production primarily results from the combination of increased  
284 export production resulting from high levels of nitrogen deposition from Indian sub-  
285 continental outflow, in conjunction with higher yields of N<sub>2</sub>O associated with the  
286 Arabian Sea OMZ [Suntharalingam et al. 2012].

287 Limitations of these model-based estimates include potential errors in model  
288 circulation arising from the coarse resolution of the global models (i.e., horizontal  
289 resolution ranging from 1.5° to 3.7°), and the nitrogen cycle parameterizations  
290 employed, which did not always account for more complex interactions and  
291 feedbacks. For example, Suntharalingam et al. [2012] report the highest increases in  
292 N<sub>2</sub>O production (e.g., of over 25% in net N<sub>2</sub>O production in the 300-1000 m depth  
293 range of the north-eastern Arabian Sea), however, this may be an overestimate, as  
294 this model analysis did not account for the potential suppression of N<sub>2</sub> fixation in  
295 response to increases in available nitrogen from deposition. The nitrogen cycle  
296 dynamics of estuarine and shelf regions (e.g., sedimentary denitrification of riverine  
297 nitrogen input) are also not well represented in coarse-resolution models, leading to  
298 uncertainties in the impacts of riverine nitrogen on the biogeochemistry of the  
299 northern Indian Ocean (see further discussion of nitrogen cycle related uncertainties  
300 in section 3.4). Here, we first discuss uncertainties arising from representation of the  
301 regional circulation of the northern Indian Ocean in global models.

302 Coarse grid ocean models (e.g., with horizontal resolution > ~1°) are not able  
303 to represent the circulation dynamics (i.e., those regulating equatorial currents,  
304 lateral mixing and ventilation) that control the extent, intensity and evolution of  
305 tropical OMZs [Coco et al. 2013; Bopp et al. 2013; Stramma et al. 2012;  
306 Gnanadesikan et al. 2012]. This presents challenges for accurate model simulation of  
307 N<sub>2</sub>O production and consumption processes in hypoxic, suboxic and anoxic waters,  
308 as the nitrification and denitrification processes controlling N<sub>2</sub>O production and  
309 consumption demonstrate significant sensitivity to even small shifts in local oxygen  
310 levels; e.g., shifts of < 5 μmol L<sup>-1</sup> in a sub-oxic regime can result in changes from net  
311 N<sub>2</sub>O production to net consumption via denitrification, thus affecting the region's  
312 net N<sub>2</sub>O fluxes [Zamora and Oschlies, 2014].

313 Eddy-resolving model analyses (1/12° resolution) of the Arabian Sea [e.g.,  
314 Resplandy et al. 2011, 2012; Lachkar et al. 2018] indicate that representation of the  
315 mesoscale dynamics at this resolution improves simulation of the region's seasonal  
316 biological productivity, associated remineralization, and the position, extent and  
317 intensity of the Arabian Sea OMZ. While higher-resolution regional models (< 1°)  
318 have previously investigated aspects of the biological productivity and oxygen  
319 distribution of the northern Indian Ocean [Wiggert et al. 2000; Resplandy et al. 2011,  
320 2012], to our knowledge, the regional nitrogen cycle, and in particular, the impact of  
321 nitrogen deposition on oceanic N<sub>2</sub>O in the Arabian Sea, has not been investigated at  
322 these spatial scales. Below, we use results from a regional high-resolution ocean  
323 model to derive a diagnostic estimate of this impact on N<sub>2</sub>O; the aim is to highlight  
324 the need for more detailed representation of the Arabian Sea's circulation and  
325 biogeochemistry when assessing nitrogen cycle changes and predicting the future  
326 evolution of N<sub>2</sub>O fluxes from this critical region.

327  
328

### 329 **3.2 Estimating Arabian Sea N<sub>2</sub>O production with output from a high-resolution** 330 **regional model**

331 We present here a diagnostically derived estimate of oceanic N<sub>2</sub>O production in the  
332 Arabian Sea, using biogeochemical fluxes and fields from the regional eddy-resolving  
333 (1/12° resolution) ocean model of Resplandy et al. [2011, 2012]. In comparison to  
334 global coarse resolution models, this high resolution regional model produces an  
335 improved representation of the mesoscale variability and circulation of the Arabian  
336 Sea (e.g., monsoonal upwelling), and of the local biological productivity. Particularly  
337 relevant to N<sub>2</sub>O are the good simulation of the regional oxygen distribution (e.g.,  
338 ambient concentrations and oxygen utilization rates), and the improved  
339 representation of the Arabian Sea OMZ [Resplandy et al. 2011, 2012].

340 Our diagnostic estimate of N<sub>2</sub>O production in the Arabian Sea (Figure 1) is  
341 derived from the N<sub>2</sub>O cycle parameterizations of Suntharalingam et al. [2012]  
342 applied to the gridded biogeochemical and flux distributions (specifically oxygen and  
343 oxygen utilization rates) of the high-resolution simulations of Resplandy et al. [2012].  
344 We estimate gridded fields of N<sub>2</sub>O production as the sum of (i) N<sub>2</sub>O from nitrification  
345 in oxygenated waters, (ii) enhanced N<sub>2</sub>O production in low oxygen conditions (via  
346 denitrification and enhanced nitrification), and (iii) N<sub>2</sub>O consumption in  
347 conditions close to anoxia (see Appendix C for calculation details). The model  
348 estimate of net N<sub>2</sub>O production in the Arabian Sea region is 0.23 Tg N yr<sup>-1</sup>, with a  
349 range of 0.1 - 0.56 Tg N yr<sup>-1</sup> when accounting for sensitivity analyses conducted on  
350 variations in N<sub>2</sub>O yield rates (yields taken from Law and Owens [1990], Naqvi and  
351 Noronha [1991], and Patra et al. [1999], see Table C1 in Appendix C). This is  
352 consistent with previous observationally-derived estimates of N<sub>2</sub>O flux for the  
353 Arabian Sea (e.g., 0.2-0.6 Tg N yr<sup>-1</sup> [Naqvi et al., 2010a]). We calculate empirically-  
354 based estimate of the impact of changes in nitrogen deposition in this region by  
355 combining N<sub>2</sub>O production estimates from the regional model together with the  
356 deposition-induced changes in N<sub>2</sub>O yield in the Arabian Sea from the model results  
357 of Suntharalingam et al. [2012] (see Appendix C for details of the estimation). We  
358 estimate a resulting change in Arabian Sea N<sub>2</sub>O production from the pre-industrial to  
359 the present of 0.01-0.07 Tg N (the range reflects sensitivity analyses on N<sub>2</sub>O yield  
360 rates). This corresponds to an estimated increase of 5% - 30% of the Arabian Sea N<sub>2</sub>O  
361 source. This estimate is derived diagnostically, thus has limited applicability for  
362 predictive purposes. However, the relatively large estimated impact, and the large  
363 uncertainty, highlights the need to develop high-resolution regional process models  
364 of nitrogen and N<sub>2</sub>O cycling, that also account for the specific nitrogen-cycling  
365 pathways important in low-oxygen regions, in order to more accurately assess the  
366 impacts of anthropogenic and climate induced changes in the northern Indian  
367 Ocean.

### 368 369 **3.3 Uncertainties in model representation of the N<sub>2</sub>O and nitrogen cycles**

370 The representation of the N<sub>2</sub>O cycle in the current generation of global  
371 biogeochemical models relies on parameterized functions derived from a relatively  
372 limited set of laboratory process studies, and optimized using in-situ oceanic  
373 measurements of N<sub>2</sub>O and related biogeochemical quantities [Buitenhuis et al. 2018;  
374 Battaglia and Joos, 2017; Zamora and Oschlies, 2014; Martinez-Rey et al. 2015;  
375 Suntharalingam et al. 2012]. These parameterizations have some success in

376 representing the N<sub>2</sub>O distribution in the well-oxygenated ocean where nitrification  
377 processes dominate N<sub>2</sub>O production. However, global models do not simulate well  
378 the complex interplay of N<sub>2</sub>O production and consumption pathways, primarily  
379 involving denitrification, that regulate N<sub>2</sub>O in low oxygen regions such as the Arabian  
380 Sea OMZ [Suntharalingam et al. 2000; Martinez-Rey et al. 2015; Zamora and Oschlies  
381 2014]. A key model challenge in these regions is the accurate representation of the  
382 net N<sub>2</sub>O yields resulting from the competing effects of dynamic production and  
383 consumption processes at the sub-oxic to anoxic interface, and in simulating the  
384 associated steep gradients in N<sub>2</sub>O observed at the oxycline boundaries of OMZs  
385 [Babbin et al. 2015, Ji et al. 2015; Kock et al., 2016]. The vertical spatial scale of these  
386 gradients in N<sub>2</sub>O and oxygen at OMZ boundaries are on the order of ~10s of metres,  
387 thus their representation remains challenging in the current generation of global  
388 biogeochemical models with relatively low spatial resolution.

389 A further challenge for biogeochemical models is the accurate representation  
390 of background nitrogen cycle processes (e.g., N<sub>2</sub> fixation), and potential regulation of  
391 these processes by changes in external nitrogen sources to marine ecosystems (e.g.,  
392 from atmospheric deposition and riverine sources). Observational evidence suggests  
393 that N<sub>2</sub> fixation provides an important 'new nitrogen' source to the northern Indian  
394 ocean [Table 1 and references therein]. Regional estimates of this source are based  
395 on very sparse measurements and have significant uncertainties (see Table 1).  
396 Gandhi et al. [2010ab] additionally note significant temporal and spatial variability of  
397 the episodic N<sub>2</sub> fixation blooms in the Arabian Sea, and highlight the need for more  
398 comprehensive measurements to improve characterization of this nitrogen source  
399 and quantification of its magnitude. Previous model analyses have included  
400 interactions between nitrogen cycle processes, for example, by accounting for  
401 suppression of N<sub>2</sub> fixation in the presence of bio-available nitrogen from deposition  
402 [Krishnamurthy et al. 2009; Yang and Gruber 2016; Jickells et al. 2017, Landolfi et al.  
403 2017]. However, challenges remain in accurately simulating such interactions, and in  
404 representing the role of micronutrients such as iron [Moore and Doney, 2007'  
405 Martino et al., 2014; Weber and Deutsch, 2014] in regulating the supply of fixed  
406 nitrogen to surface ocean ecosystems.

407 On longer time-scales of decades to centuries, a potentially important  
408 feedback process associated with the oxygen depleted basins of the northern Indian  
409 Ocean involves the interaction between nitrogen deposition, denitrification and N<sub>2</sub>  
410 fixation [Landolfi et al. 2013; Somes et al. 2017]. We discuss this in more detail in  
411 section 3.4 below.

412

### 413 **3.4 Implications of increasing nitrogen deposition for changes in water column** 414 **denitrification**

415 While increased nitrogen loading may initially fertilize biological production, the  
416 onset of biogeochemical feedbacks may lead to an overall nitrogen impoverishment  
417 of the region [Landolfi et al. 2013] with impacts on biological productivity [Somes et  
418 al. 2017]. In oxygen deficient regions, increased organic matter production may  
419 exacerbate oxygen consumption and stimulate anaerobic remineralization via  
420 denitrification, which consumes fixed nitrogen. A simplified analysis combining  
421 modeled nitrogen deposition fields with regional oxygen data [Bianchi et al., 2012]  
422 suggests that the nitrogen removed by atmospheric nitrogen-driven denitrification

423 may be larger than the nitrogen gain (Figure 2). This is based on the following  
424 simplifying assumptions: 1) atmospheric nitrogen deposited onto surface waters  
425 overlying the OMZ produces biomass in Redfield ratios and sinks at those locations,  
426 2) export flux within the OMZs can be represented either by the standard Martin  
427 curve [Martin et al., 1987] or following the fit computed by Van Mooy et al. [2002]  
428 from sub-oxic zone regions, 3) organic matter within the OMZ is remineralized under  
429 anoxic conditions by denitrification, following 4) standard denitrification  
430 stoichiometry [e.g. Paulmier et al. 2009], such that for every mole of nitrogen from  
431 organic matter remineralised in sub-oxic waters, up to 7 additional moles of ambient  
432 seawater nitrate may be lost to  $N_2$  due to its role as an electron acceptor in the  
433 denitrification process [Codispoti et al., 2001]. These assumptions do not account for  
434 such factors as advection-driven redistributions of deposited atmospheric nitrogen,  
435 any deposition-driven expansion of OMZs and other nitrogen-inventory stabilizing  
436 feedbacks, such as the reduction of fixed nitrogen inputs by marine  $N_2$  fixers. These  
437 processes have impacts of differing sign and magnitude and may counteract each  
438 other. Using the more comprehensive model of Landolfi et al. [2017] we find that  
439 following atmospheric deposition of nitrogen in the light-lit surface waters,  $N_2$  fixers  
440 lose their competitive advantage (6% decline). The oxygenic remineralization of  
441 organic matter at depth promotes the expansion of low-oxygen waters in the  
442 Arabian Sea and Bay of Bengal triggering nitrogen loss via water column and benthic  
443 denitrification (12% increase). In this model, the increased atmospheric nitrogen  
444 load ( $+2.7 \text{ Tg N yr}^{-1}$ ) leads to an additional loss of  $0.53 \text{ Tg N yr}^{-1}$ , that is a  $\sim 10\%$  larger  
445 nitrogen-cycle imbalance relative to preindustrial conditions [Landolfi et al. 2017].  
446 This highlights a potential negative feedback that acts to stabilize the oceanic  
447 nitrogen inventory when subject to additional anthropogenic nitrogen inputs.

448

#### 449 **4. Summary and Recommendations**

450 Nitrogen cycle processes regulating  $N_2O$  emissions from the northern Indian Ocean  
451 basins are vulnerable to the ongoing rapid increase in the regional outflow of  
452 anthropogenically derived nitrogen to the two ocean basins. The Arabian Sea OMZ is  
453 currently a globally significant site for oceanic  $N_2O$  production, where  $N_2O$  emissions  
454 could increase under this increased nitrogen loading, especially in conjunction with  
455 other climate-induced stressors such as ocean deoxygenation. Biogeochemical  
456 systems in regions of the Bay of Bengal are close to hypoxia, and could shift into a  
457 denitrifying regime, with loss of  $N_2O$  and  $N_2$  to the atmosphere, if oxygen levels are  
458 further depleted by biogeochemical or physical drivers. Many of the biogeochemical  
459 processes governing these changes are currently poorly characterized by  
460 observations, and their representation in models (ocean and atmospheric) have  
461 significant uncertainties.

462

463 To enable more accurate estimation of the impacts of anthropogenic nitrogen inputs  
464 on current and future  $N_2O$  emissions from this region, we recommend further  
465 investigation into the following key issues:

466 (1) Reduction of uncertainties on external nitrogen inputs to the northern Indian  
467 ocean by :

468 a) improved characterization of the magnitude, composition (i.e., ammonium,  
469 nitrate, organic nitrogen), and variability of the atmospheric nitrogen

470 deposition flux to the Arabian Sea and Bay of Bengal. This requires an  
471 observational strategy involving regular monitoring of aerosol composition at  
472 representative sites, as well as targeted field campaigns to account for  
473 monsoonal influences, and to identify the major pathways of pollution  
474 outflow from the sub-continent. These data also require consolidation to  
475 derive regional atmospheric deposition nitrogen fluxes using high resolution  
476 atmospheric modeling. In turn, this requires well calibrated, up-to-date high-  
477 resolution emissions of the precursors to nitrogen deposition (such as NO  
478 and NH<sub>3</sub> emissions).

479 b) implementation of a long-term nutrient monitoring network in rivers draining  
480 into the Arabian Sea and Bay of Bengal. We recommend regular sampling  
481 (e.g., monthly) of the outflow of nutrient fluxes, and process studies in  
482 estuarine and shelf systems to characterize local nitrogen cycling dynamics  
483 and to assess the proportion of riverine nitrogen reaching open ocean  
484 waters.

485 c) accurate quantification of the contribution of N<sub>2</sub> fixation to the regional  
486 nitrogen budget through a comprehensive program of direct measurements.  
487 In addition, improved understanding is required on the processes regulating  
488 activity of nitrogen-fixing organisms (for example, concerning potential  
489 suppression of activity under nitrogen input from deposition, and regulation  
490 by micro- and macro- nutrients).

491

492 2) Improved assessment of the potential impacts of these anthropogenic nitrogen  
493 inputs on the regional oceanic N<sub>2</sub>O fluxes. This requires:

494 (a) a comprehensive observational approach providing in-situ measurements and  
495 process-knowledge to improve current understanding on the evolution of N<sub>2</sub>O in the  
496 low-oxygen waters of both basins. Such a strategy will include:

497 i. basin-wide networks of regular measurements of nitrogen cycle process rates  
498 (e.g., nitrification, denitrification, N<sub>2</sub> fixation) in the water column at selected  
499 sites in the Arabian Sea and Bay of Bengal to decipher the short- and long-  
500 term trends of the N<sub>2</sub>O and background nitrogen cycle.

501 ii. regular time-series measurements of N<sub>2</sub>O, oxygen and nutrient depth profiles  
502 at representative sites in the coastal and open ocean of the Arabian Sea and  
503 the Bay of Bengal.

504 iii. high-resolution measurements of N<sub>2</sub>O in the surface ocean and the  
505 atmospheric boundary layer on voluntary observing ship (VOS) lines crossing  
506 the Arabian Sea and Bay of Bengal.

507 iv. further development of high-spatial-resolution satellite and aircraft  
508 observations of near-surface atmospheric nitrogen species (e.g., NO<sub>x</sub> and  
509 N<sub>2</sub>O) and surface-ocean phytoplankton composition, which can be used to  
510 evaluate atmospheric deposition and its impacts on marine ecosystems.

511

512

513 (b) A targeted modeling strategy involving :

514 i. development of customized regional biogeochemical process models that  
515 build on recent advances in eddy resolving models of the Arabian Sea and  
516 Bay of Bengal, and also incorporate the key nitrogen and N<sub>2</sub>O cycling

517 processes specific to the region's low-oxygen waters. Specific improvements  
518 required to current model parameterizations include accurate  
519 characterization of the oxygen thresholds and denitrification processes  
520 regulating N<sub>2</sub>O production, consumption, and the net yield in the region's  
521 hypoxic and sub-oxic waters.

522 ii. a synthesis of these regional model analyses with those from global  
523 biogeochemical models and Earth System Models. This will enable evaluation  
524 of the relative contribution of the Arabian Sea and Bay of Bengal to global  
525 oceanic N<sub>2</sub>O emissions, and also provide mechanistic process knowledge to  
526 inform the development of new biogeochemical parameterizations in the  
527 global models towards improved predictive capability of N<sub>2</sub>O-climate  
528 feedbacks.

529

530 National and international collaboration will be an integral aspect of the envisioned  
531 multidisciplinary studies. An improved understanding of the Indian Ocean nitrogen  
532 cycle will contribute to our understanding of the crucial role of the nitrogen cycle in  
533 societal-relevant issues such as climate change, eutrophication, air quality (pollution)  
534 and the overall health of the ocean. To this end, we propose that nitrogen research  
535 should be a continuing long-term focus of international research initiatives such as  
536 the 2<sup>nd</sup> International Indian Ocean Expedition (IIOE-2: [www.iioe-2.incois.gov.in](http://www.iioe-2.incois.gov.in)),  
537 SOLAS (Surface Ocean Lower Atmosphere Study: [www.solas-int.org](http://www.solas-int.org)) and Future  
538 Earth ([www.futureearth.org](http://www.futureearth.org)).

539

540

541

**542 Appendix A : Derivation of flux estimates for section 2**

543 For the flux estimates calculated here, the Arabian Sea area is defined as the oceanic  
544 region within the polygonal area bounded by (0°N, 42°E), (0°N, 76°E), (26.5°N, 76°E)  
545 and (26.5°N, 59°E), and the Bay of Bengal as the region with bounds 5°-24°N and  
546 76°-97.5°E.

547

548 Model derived atmospheric N-deposition and riverine N-fluxes : Atmospheric  
549 inorganic nitrogen inputs are estimated from CAM version 3.5 model [Lamarque et  
550 al., 2011]. Water soluble organic nitrogen (WSON) atmospheric deposition was  
551 obtained from the Kanakidou et al. [2012] model. All global models have difficulties  
552 in reproducing the spatial distribution of reduced and oxidized inorganic reactive  
553 nitrogen deposition fluxes. In particular, in the northern Indian Ocean, global models  
554 overestimated nitrate dry deposition fluxes while underestimating ammonium dry  
555 deposition fluxes [Baker et al., 2017]. These discrepancies that can be partially  
556 explained by seasonal biases in the sampling or by the pH dependence of the nitrate  
557 and ammonium partitioning to the aerosol phase [Weber et al., 2016; Kanakidou et  
558 al., 2018]. Riverine dissolved inorganic and organic nitrogen inputs were obtained  
559 from the NEWS model [Seitzinger et al., 2010; Pedde et al. 2017] and other sources  
560 listed in Table 1.

561

**562 Flux estimates from aerosol data:**

563 Data for nitrate ( $\text{NO}_3^-$ ) and ammonium ( $\text{NH}_4^+$ ) aerosol concentrations were obtained  
564 from the Surface Ocean Lower Atmosphere Study (SOLAS) Project Integration  
565 website

566 ([http://www.bodc.ac.uk/solas\\_integration/implementation\\_products/group1/aerosol\\_rain/](http://www.bodc.ac.uk/solas_integration/implementation_products/group1/aerosol_rain/)), and from sources cited in Table 1. The dry deposition fluxes of aerosol  
567 WSON from the Arabian Sea were adopted from the mean value of the two cruises  
568 sampled in Srinivas and Sarin [2013]. Given the aerosol concentration (C), the dry  
569 deposition flux (F) was estimated using the following equation:  $F = C \cdot V_d$ , and by  
570 assuming deposition velocities ( $V_d$ ) of  $1 \text{ cm s}^{-1}$  for  $\text{NO}_3^-$  [Duce et al., 1991] and  $0.6 \text{ cm s}^{-1}$   
571 for  $\text{NH}_4^+$  [Spokes et al., 2000]. The  $V_d$  is difficult to quantify and is affected by  
572 several factors such as wind speed, surface roughness and, particle size etc. Due to  
573 the inherent uncertainties associated with the assumed dry deposition velocities, the  
574 estimates of atmospheric dry deposition of nitrogen carry additional uncertainty by  
575 up to a factor of three.

577 Wet deposition is only collected at a few long-term coastal monitoring sites, and  
578 hardly ever over the open ocean. Therefore, following Singh et al. [2012], wet  
579 deposition fluxes ( $F_w$ ) were estimated from the following formula:

580  $F_w = P \cdot S \cdot C_d \cdot \rho_a^{-1} \cdot \rho_w$ , where P is the precipitation rate from the 1981-2010 Global  
581 Precipitation Climatology Project long term mean [Adler et al., 2003], S is the  
582 assumed scavenging ratio (330 and 200 for nitrate and ammonium, respectively  
583 [Singh et al., 2012]), and 131 for WSON (the mean from Zamora et al. [2013]),  $C_d$  is  
584 the aerosol concentration, and  $\rho_a$  and  $\rho_w$  are the densities of air and water,  
585 respectively.

586 Interpolated values for each basin (the Arabian Sea and the Bay of Bengal) were  
587 obtained at  $1^\circ \times 1^\circ$  resolution), and then averaged to estimate wet and dry  
588 deposition to the northern Indian Ocean.

589

590 **Appendix B : Global biogeochemistry models used in analysis of 3.1**

591 The investigations of Suntharalingam et al. [2012] and Jickells et al. [2017] both  
 592 employed the PlankTOM ocean biogeochemistry model (PlankTOM5 and  
 593 PlankTOM10 respectively) embedded in the NEMO ocean general circulation model,  
 594 v 3.1 [Madec, 2008], and the diagnostic N<sub>2</sub>O model of Suntharalingam et al. [2012].  
 595 The model analysis of Landolfi et al. [2017] employed the UVic2.9 Earth System  
 596 Model, and the N<sub>2</sub>O parameterization of Zamora and Oschlies [2014]. The model  
 597 analyses all evaluated the impact of changes in external nutrient input from the pre-  
 598 industrial to the present, but have differences in their representation of the specific  
 599 nutrient inputs considered (deposition, riverine and N<sub>2</sub> fixation), and in their  
 600 parameterization of the nitrogen cycle. Further details of model specifications and  
 601 assumptions are given in the individual publications.

602

603 **Appendix C: Estimation of N<sub>2</sub>O Production and Impact of Nitrogen Deposition in  
604 section 3.2**

605 For the regional model calculations of section 3.2, we derive a diagnostic estimate of  
 606 the net N<sub>2</sub>O source from the parameterizations of Suntharalingam et al. [2012, 2000]  
 607 which estimate N<sub>2</sub>O production as a function of organic matter remineralization via  
 608 the local oxygen consumption rate. The N<sub>2</sub>O production parameterizations are  
 609 applied to the gridded biogeochemical model fields (specifically oxygen and oxygen  
 610 utilization rates) of the high-resolution model simulations (1/12 degree) of  
 611 Resplandy et al. [2012]. N<sub>2</sub>O production is estimated from two separate pathways:  
 612 (i) nitrification in the oxygenated open ocean, and (ii) higher N<sub>2</sub>O yield processes  
 613 (denitrification, enhanced nitrification) in low-oxygen zones. In addition, N<sub>2</sub>O loss by  
 614 denitrification is represented below a specified oxygen threshold ( $[O_2] < [O_2]_{denit}$ ).  
 615 Overall, the net N<sub>2</sub>O production is estimated as:

$$616 \quad \text{Net N}_2\text{O production} = \alpha \cdot [\text{O}_2 \text{ consumption}] + \beta \cdot f(\text{O}_2) \cdot [\text{O}_2 \text{ consumption}] - [\text{Anoxic N}_2\text{O loss}]$$

617

618 Here, the scalar parameter  $\alpha$  (mol [N<sub>2</sub>O]/mol [O<sub>2</sub>]) represents the N<sub>2</sub>O yield from  
 619 nitrification and is quantified from observed correlations between excess N<sub>2</sub>O  
 620 ( $\Delta\text{N}_2\text{O}$ ) and Apparent Oxygen Utilization (AOU). The parameter  $\beta$  represents the  
 621 higher yield of N<sub>2</sub>O in sub-oxic zones, and  $f(\text{O}_2)$  represents the non-linear functional  
 622 dependence of N<sub>2</sub>O yield on oxygen level. N<sub>2</sub>O loss in anoxic conditions is  
 623 represented as (-1) x the value of total N<sub>2</sub>O production below oxygen threshold  
 624 levels of  $[O_2]_{denit}$ ; i.e., all N<sub>2</sub>O produced in these zones is assumed to be consumed by  
 625 denitrification processes, and this represents an upper bound on N<sub>2</sub>O loss in these  
 626 zones. For the analyses of this section we take the threshold level of  $[O_2]_{denit}$  as 5  
 627  $\mu\text{mol L}^{-1}$ . Refer to Suntharalingam et al. [2012, 2000] and Buithenhuis et al. [2018]  
 628 for additional details on the N<sub>2</sub>O parameterization.

629 Table C1 presents the regionally aggregated results from the set of scenarios  
 630 constructed for this analysis. These scenarios evaluate the sensitivity of estimated  
 631 regional N<sub>2</sub>O production to variation in the N<sub>2</sub>O yield parameters  $\alpha$  and  $\beta$ . The  
 632 'Standard' model takes  $\alpha$  and  $\beta$  values from Suntharalingam et al. [2012]. In addition  
 633 we evaluate scenarios based on N<sub>2</sub>O measurement analyses from the Arabian Sea  
 634 (Law and Owens [1990], Naqvi and Noronha [1991], and Patra et al. [1999]). For  
 635 these scenarios, the N<sub>2</sub>O yield parameters were derived from the  $\Delta\text{N}_2\text{O}/\text{AOU}$



636 correlations reported in the individual studies. Figure 1 presents the column  
 637 integrated distribution of N<sub>2</sub>O production from the Standard scenario for the Arabian  
 638 Sea region.

639 We derive an empirical estimate of the impact of changes in nitrogen deposition in in  
 640 the Arabian Sea on regional N<sub>2</sub>O production by applying the regionally averaged  
 641 fractional change in N<sub>2</sub>O yield for the Arabian Sea region ( $\gamma_{N_2O_{dep}}=0.125$ ) from the  
 642 nitrogen deposition of Suntharalingam et al. [2012] to the N<sub>2</sub>O production estimates  
 643 derived above for the regional model:

644 i.e.,

$$\text{Change in N}_2\text{O} = (\gamma_{N_2O_{dep}}) \times [\text{N}_2\text{O production}]$$

647 **Table C1 : Regional model analysis of N<sub>2</sub>O production scenarios and estimates of**  
 648 **the impact of nitrogen deposition**

649

<b>N<sub>2</sub>O Production Scenarios</b>	<b>N<sub>2</sub>O Production (Tg N yr<sup>-1</sup>) Nitrification Pathway</b>	<b>N<sub>2</sub>O Production (Tg N yr<sup>-1</sup>) Low-Oxygen Pathway</b>	<b>N<sub>2</sub>O loss (Tg N yr<sup>-1</sup>)</b>	<b>Net N<sub>2</sub>O production (Tg N yr<sup>-1</sup>)</b>	<b>Impact of Nitrogen Deposition on N<sub>2</sub>O production* (Tg N yr<sup>-1</sup>)</b>
<b>Standard</b> $\alpha=0.75 \times 10^{-4}$ ; $\beta=0.01$	<b>0.055</b>	<b>0.27</b>	<b>0.096</b>	<b>0.23</b>	<b>0.029</b>
<b>LOW <math>\alpha \beta</math> scenario</b> $\alpha=0.5 \times 10^{-4}$ ; $\beta=0.005$	0.037	0.135	0.048	0.124	0.015
<b>HIGH <math>\alpha \beta</math> scenario</b> $\alpha=2.0 \times 10^{-4}$ ; $\beta=0.02$	0.147	0.54	0.124	0.564	0.070
<b>Law and Owens 1990 LOW scenario</b> $\alpha=0.33 \times 10^{-4}$ ; $\beta=0.01$	0.024	0.27	0.097	0.197	0.025
<b>Law and Owens 1990 HIGH scenario</b> $\alpha=3.1 \times 10^{-4}$ ; $\beta=0.01$	0.228	0.27	0.098	0.40	0.05
<b>Naqvi and Noronha 1991 scenario</b> $\alpha=1.7 \times 10^{-4}$ ; $\beta=0.01$	0.125	0.27	0.097	0.298	0.037
<b>Patra 1999 scenario</b> $\alpha=1.5 \times 10^{-4}$ ; $\beta=0.01$	0.110	0.27	0.097	0.283	0.036

650

651 \*Impact of nitrogen deposition estimated as Change =  $\gamma_{N_2O_{dep}} \times N_2O$  production

652

653

#### 654 **Appendix D : Acronyms**

655 NCAR-CAM : National Center for Atmospheric Community Atmosphere Model

656 TM4-ECPL : Transport Model version 4-Environmental Chemical Processes  
 657 Laboratory

658 NEMO : Nucleus for European Modelling of the Ocean

659 PlankTOM : Plankton Types Ocean Model

660

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678

679

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**Table 1.** Estimates of present-day external(non-recycled) dissolved nitrogen inputs (Tg N yr<sup>-1</sup>) to the Arabian Sea and Bay of Bengal from riverine, atmospheric, and diazotrophic sources.

	Riverine	Atmospheric	N <sub>2</sub> Fixation
<b>Arabian Sea</b>			
Model-derived	0.4 <sup>a</sup>	1.4 <sup>d</sup>	2.6 <sup>f</sup> -6.2 <sup>g</sup>
Obs-based	0.1-0.4 <sup>b</sup>	1.8 <sup>e</sup>	0.0-20.4 <sup>h,i</sup>
<b>Bay of Bengal</b>			
Model-derived	5 <sup>a</sup>	1.1 <sup>d</sup>	4.2 <sup>f</sup> -4.3 <sup>g</sup>
Obs-based	0.9-2.0 <sup>c</sup>	1.6 <sup>e</sup>	0.6-11.3 <sup>i</sup>

<sup>a</sup>This study, year 2000 model output includes dissolved organic and inorganic nitrogen [Seitzinger *et al.* 2010; Pedde *et al.* 2017]

<sup>b</sup>[Singh and Ramesh, 2011]; [Singh *et al.*, 2012]

<sup>c</sup>[Srinivas and Sarin, 2013] and references therein.

<sup>d</sup>This study, year 2005 model output includes water soluble inorganic and organic nitrogen [Lamarque *et al.*, 2011; Kanakidou *et al.*, 2012]; See Appendix A for further details.

<sup>e</sup>This study, see Appendix A for further details.

<sup>f</sup>[Jickells *et al.* 2017]

<sup>g</sup>[Le Quéré *et al.* 2016]

<sup>h</sup>[Gandhi *et al.*, 2011] estimated N<sub>2</sub> fixation rates using <sup>15</sup>N<sub>2</sub> with less than 6% uncertainty.

<sup>i</sup>[Srinivas and Sarin, 2013] and references therein, converted to the larger open ocean regions used in this study (6.6x10<sup>6</sup> km<sup>2</sup> for the Arabian Sea, and 3.0x10<sup>6</sup> km<sup>2</sup> for the Bay of Bengal). Nitrogen fixation rates were estimated with respect to atmospheric inputs by assuming Fe and P limitation in the ocean.

**Table 2** : Model-derived estimates of changes in global and northern Indian ocean N<sub>2</sub>O production due to the impact of external nitrogen inputs from atmospheric deposition. Estimates shown summarize results from the model analyses of Suntharalingam et al. [2012], Jickells et al. [2017], and the NDEP simulation of Landolfi et al. [2017].

REGION	Present day oceanic N <sub>2</sub> O source * (Tg N yr <sup>-1</sup> )	Change in N <sub>2</sub> O source due to nitrogen inputs (pre-industrial to present day) (Tg N yr <sup>-1</sup> )
Northern Indian Ocean**	0.3 – 0.6	0.003 – 0.05
Global Ocean	2.5-4.5	0.04 – 0.15

\* Present day values derived from model simulations for period 1995-2005.

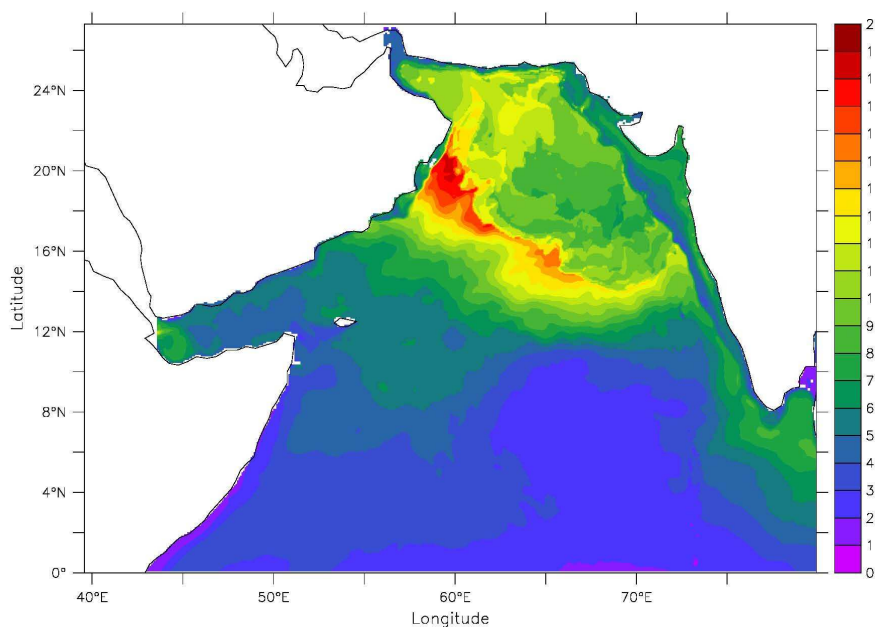
\*\* Defined here as the ocean region north of the Equator and within the longitudinal range 42°E-100°E.

**Figure 1.** Net column-integrated N<sub>2</sub>O production (units : Mg N m<sup>-2</sup> yr<sup>-1</sup>) in the Arabian Sea derived from the regional high-resolution model analysis reported in section 3.2. The net N<sub>2</sub>O production is derived from the methodology outlined in Appendix C and is the sum of N<sub>2</sub>O production from the nitrification and low-oxygen pathways, and N<sub>2</sub>O loss below an oxygen threshold of 5 μmol L<sup>-1</sup>.

**Figure 2 :** Illustration of the possible ratio of net nitrogen loss: net nitrogen gain (mol:mol) from atmospheric nitrogen deposition to OMZs in the northern Indian Ocean assuming either a) the Martin curve, or b) the Van Mooy curve. Calculations based on World Ocean Atlas O<sub>2</sub> data [Bianchi et al. 2012].

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**Figure 1.** Net column-integrated  $\text{N}_2\text{O}$  production (units :  $\text{Mg N m}^{-2} \text{ yr}^{-1}$ ) in the Arabian Sea derived from the regional high-resolution model analysis reported in section 3.2. The net  $\text{N}_2\text{O}$  production is derived from the methodology outlined in Appendix C and is the sum of  $\text{N}_2\text{O}$  production from the nitrification and low-oxygen pathways, and  $\text{N}_2\text{O}$  loss below an oxygen threshold of  $5 \mu\text{mol L}^{-1}$ .



**Figure 2** : Illustration of the possible ratio of net nitrogen loss: net nitrogen gain (mol:mol) from atmospheric nitrogen deposition to OMZs in the northern Indian Ocean assuming either a) the Martin curve, or b) the Van Mooy curve. Calculations based on World Ocean Atlas O<sub>2</sub> data [Bianchi et al. 2012].

