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Anthropogenic nitrogen inputs and impacts on oceanic N_2O 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_2O). 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_2O
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_2O 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 2nd International Indian Ocean Expedition program [Hood et al. 2016] was
25 launched in December 2015 on the occasion of the 50th 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 21st 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 at 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_2O). 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
Nitrous oxide in the northern Indian Ocean
65
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

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freshwater discharge in the north of the Bay of Bengal from the Ganges outflow, in conjunction with riverine discharge from the peninsular subcontinent into the south-west of the Bay of Bengal has resulted in a latitudinal gradient in surface N_2O concentrations along the coast of the western Bay of Bengal characterized by equilibrium or under-saturation in the northwest of the basin and small supersaturation and emission in the south-west [Rao et al. 2013]. According to Rao et al. [2013] this complex behavior results from a combination of factors, including seasonal undersaturation in N_2O associated with monsoonally-governed variations in Ganges freshwater outflow, and higher nitrification rates and associated N_2O supersaturation in the coastal upwelling regions in the south-west of the Bay of Bengal.

The Asian monsoon system plays an important role in regulating the temporal variation of the region's N_2O production and emission to the atmosphere. Monsoonal winds drive a seasonally reversing upper ocean circulation with associated changes in seasonal upwelling and regional biological productivity in both the Arabian Sea and the Bay of Bengal [McCreary et al., 2013; Schott and McCreary, 2001; Madhuratap et al. 1996, and references therein]. This upwelling is most intense in the NW Arabian Sea and leads to especially high regional N_2O [De Wilde and Helder, 1996; Bange et al. 2001]. The Asian monsoon system also accounts for pronounced rainfall and river discharge in the northern Bay of Bengal, playing an important role in the hydrography and associated biogeochemistry of the basin. These freshwater fluxes reduce surface salinity and enhance water column stratification, yielding lower productivity levels in the Bay of Bengal than in the neighbouring Arabian Sea [Rao et al. 2013; Singh and Ramesh, 2015 and references therein].

Previous estimates of oceanic N_2O emission from the northern Indian Ocean highlight its significant contribution to global fluxes [Law and Owens 1990; Naqvi and Noronha 1991; Lal and Patra 1998; Bange et al. 2001]. The Arabian Sea, in particular, is a region of intense oceanic N_2O production, and is estimated to provide a flux to the atmosphere of $0.2 - 0.6$ Tg N yr^{-1} [Naqvi et al., 2010b]. This flux represents $2 - 31$ % of global total oceanic N_2O emissions (N_2O emissions ($1.9 - 12.3$ Tg N yr^{-1}) from the open ocean, coastal and estuarine sources [Ciais et al. 2013]. In contrast N_2O fluxes to the atmosphere from the Bay of Bengal are considerably lower ($0.027 - 0.077$ Tg N yr^{-1}) [Naqvi et al. 1994; Naqvi et al. 2010a], a result of less pronounced upwelling and lower productivity levels.

Vulnerability to climate change and anthropogenic nutrient input

The circulation and biogeochemistry of the northern Indian Ocean basins are vulnerable to the impacts of climate warming and associated changes; for example, to (a) ocean deoxygenation and the predicted expansion of ocean OMZs [Stramma et al. 2010; Naqvi et al. 2010b; Banse et al 2014], and (b) potential increases in the intensity of the monsoonal circulation and associated upwelling and productivity in the Arabian Sea [De Castro et al. 2016; Goes et al. 2005]. Such changes could result in significant increases in oceanic N_2O emissions from the region [Codispoti, 2010]. These hypotheses are supported by recent model analyses that predict the deepening and intensification of the Arabian Sea OMZ due to strengthening of the region's monsoonal winds [Lachkar et al. 2018], and increases in northern Indian

142 Ocean N_2O production associated with expansion of oceanic low-oxygen regions
143 under scenarios of 21st 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_2O and N_2 emission to the
148 atmosphere [Naqvi et al. 2008; Bristow et al. 2017].

149   N_2O 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_2O 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_2O 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_2O 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_2O 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_2O emissions from the northern Indian Ocean (section 4).

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

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

Atmospheric chemistry models also estimate substantial increases in atmospheric nitrogen deposition to the northern Indian Ocean over recent decades [Dentener et al. 2006; Kanakidou et al. 2012, 2016; Lamarque et al. 2013]. This trend is supported by observations; e.g., since the 1980s, NO_x emissions over India have more than doubled [Garg et al. 2006], and there have been significant increases in observed wet nitrate deposition at different coastal and open ocean sites [Attri and Tyagi, 2010; Safai et al., 2004]. Cruise measurements made between 2001-2009 in the Bay of Bengal show similar trends for aerosol NH_4^+ concentrations [Srinivas et al., 2011]. Based on a combination of output from the NCAR-CAM (version 3.5) model [Lamarque et al., 2011] and the TM4-ECPL model [Kanakidou et al., 2012] models (see Appendix A for details), we estimate an ~226% (1.42 Tg N m^{-2}) increase in the amount of soluble nitrogen being deposited to the northern Indian Ocean between 1850 and 2005, 59% of which went into the Arabian Sea.

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

For biologically-mediated nitrogen sources, there are very few estimates of nitrogen (N_2) fixation in the northern Indian Ocean. Direct estimates, made using isotopically enriched ($^{15}N_2$) tracers, suggest extremely high rates of N_2 fixation in the Arabian Sea during springtime (Table 1, Gandhi et al., 2011). Srinivas and Sarin [2013] provide indirect estimates of N_2 fixation rates, derived with respect to atmospheric deposition inputs and assuming Fe and P limitation in the ocean; these estimates suggest a large variability in regional N_2 fixation rates, linked to the uncertainty in atmospheric deposition fluxes. The role of N_2 fixation must also, therefore, be better constrained for a more accurate assessment of anthropogenic nutrient impacts on the biogeochemistry of the northern Indian Ocean.

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Uncertainties in nitrogen inputs

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

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

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

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

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283 enhancement of N_2O 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_2O 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^o to 3.7^o), 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_2O production (e.g., of over 25% in net N_2O 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_2 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_2O production and consumption processes in hypoxic, suboxic and anoxic waters,
308 as the nitrification and denitrification processes controlling N_2O production and
309 consumption demonstrate significant sensitivity to even small shifts in local oxygen
310 levels; e.g., shifts of < 5 µmol L^{-1} in a sub-oxic regime can result in changes from net
311 N_2O production to net consumption via denitrification, thus affecting the region's
312 net N_2O fluxes [Zamora and Oschlies, 2014].

313   Eddy-resolving model analyses ($1/12^o$ 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_2O 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_2O; 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_2O fluxes from this critical region.

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328

329 **3.2 Estimating Arabian Sea N$_2$O production with output from a high-resolution**
330 **regional model**
331 We present here a diagnostically derived estimate of oceanic N$_2$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$_2$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$_2$O production in the Arabian Sea (Figure 1) is
341 derived from the N$_2$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$_2$O production as the sum of (i) N$_2$O from nitrification
345 in oxygenated waters, (ii) enhanced N$_2$O production in low oxygen conditions (via
346 denitrification and enhanced nitrification), and (iii) and N$_2$O consumption in
347 conditions close to anoxia (see Appendix C for calculation details). The model
348 estimate of net N$_2$O production in the Arabian Sea region is 0.23 Tg N yr^{-1}, with a
349 range of 0.1 - 0.56 Tg N yr^{-1} when accounting for sensitivity analyses conducted on
350 variations in N$_2$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$_2$O flux for the
353 Arabian Sea (e.g., 0.2-0.6 Tg N yr^{-1} [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$_2$O production estimates from the regional model together with the
356 deposition-induced changes in N$_2$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$_2$O production from the pre-industrial to
359 the present of 0.01-0.07 Tg N (the range reflects sensitivity analyses on N$_2$O yield
360 rates). This corresponds to an estimated increase of 5% - 30% of the Arabian Sea N$_2$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$_2$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$_2$O and nitrogen cycles**
370 The representation of the N$_2$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$_2$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

representing the N$_2$O distribution in the well-oxygenated ocean where nitrification processes dominate N$_2$O production. However, global models do not simulate well the complex interplay of N$_2$O production and consumption pathways, primarily involving denitrification, that regulate N$_2$O in low oxygen regions such as the Arabian Sea OMZ [Suntharalingam et al. 2000; Martinez-Rey et al. 2015; Zamora and Oschlies 2014]. A key model challenge in these regions is the accurate representation of the net N$_2$O yields resulting from the competing effects of dynamic production and consumption processes at the sub-oxic to anoxic interface, and in simulating the associated steep gradients in N$_2$O observed at the oxycline boundaries of OMZs [Babbin et al. 2015, Ji et al. 2015; Kock et al., 2016]. The vertical spatial scale of these gradients in N$_2$O and oxygen at OMZ boundaries are on the order of ~10s of metres, thus their representation remains challenging in the current generation of global biogeochemical models with relatively low spatial resolution.

A further challenge for biogeochemical models is the accurate representation of background nitrogen cycle processes (e.g., N$_2$ fixation), and potential regulation of these processes by changes in external nitrogen sources to marine ecosystems (e.g., from atmospheric deposition and riverine sources). Observational evidence suggests that N$_2$ fixation provides an important 'new nitrogen' source to the northern Indian ocean [Table 1 and references therein]. Regional estimates of this source are based on very sparse measurements and have significant uncertainties (see Table 1). Gandhi et al. [2010ab] additionally note significant temporal and spatial variability of the episodic N$_2$ fixation blooms in the Arabian Sea, and highlight the need for more comprehensive measurements to improve characterization of this nitrogen source and quantification of its magnitude. Previous model analyses have included interactions between nitrogen cycle processes, for example, by accounting for suppression of N$_2$ fixation in the presence of bio-available nitrogen from deposition [Krishnamurthy et al. 2009; Yang and Gruber 2016; Jickells et al. 2017, Landolfi et al. 2017]. However, challenges remain in accurately simulating such interactions, and in representing the role of micronutrients such as iron [Moore and Doney, 2007' Martino et al., 2014; Weber and Deutsch, 2014] in regulating the supply of fixed nitrogen to surface ocean ecosystems.

On longer time-scales of decades to centuries, a potentially important feedback process associated with the oxygen depleted basins of the northern Indian Ocean involves the interaction between nitrogen deposition, denitrification and N$_2$ fixation [Landolfi et al. 2013; Somes et al. 2017]. We discuss this in more detail in section 3.4 below.

3.4 Implications of increasing nitrogen deposition for changes in water column denitrification

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

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

4. Summary and Recommendations

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

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

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

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

deposition flux to the Arabian Sea and Bay of Bengal. This requires an observational strategy involving regular monitoring of aerosol composition at representative sites, as well as targeted field campaigns to account for monsoonal influences, and to identify the major pathways of pollution outflow from the sub-continent. These data also require consolidation to derive regional atmospheric deposition nitrogen fluxes using high resolution atmospheric modeling. In turn, this requires well calibrated, up-to-date high-resolution emissions of the precursors to nitrogen deposition (such as NO and NH_3 emissions).

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

    c) accurate quantification of the contribution of N_2 fixation to the regional nitrogen budget through a comprehensive program of direct measurements. In addition, improved understanding is required on the processes regulating activity of nitrogen-fixing organisms (for example, concerning potential suppression of activity under nitrogen input from deposition, and regulation by micro- and macro- nutrients).

2) Improved assessment of the potential impacts of these anthropogenic nitrogen inputs on the regional oceanic N_2O fluxes. This requires:

(a) a comprehensive observational approach providing in-situ measurements and process-knowledge to improve current understanding on the evolution of N_2O in the low-oxygen waters of both basins. Such a strategy will include:

    i. basin-wide networks of regular measurements of nitrogen cycle process rates (e.g., nitrification, denitrification, N_2 fixation) in the water column at selected sites in the Arabian Sea and Bay of Bengal to decipher the short- and long-term trends of the N_2O and background nitrogen cycle.

    ii. regular time-series measurements of N_2O, oxygen and nutrient depth profiles at representative sites in the coastal and open ocean of the Arabian Sea and the Bay of Bengal.

    iii. high-resolution measurements of N_2O in the surface ocean and the atmospheric boundary layer on voluntary observing ship (VOS) lines crossing the Arabian Sea and Bay of Bengal.

    iv. further development of high-spatial-resolution satellite and aircraft observations of near-surface atmospheric nitrogen species (e.g., NOx and N_2O) and surface-ocean phytoplankton composition, which can be used to evaluate atmospheric deposition and its impacts on marine ecosystems.

(b) A targeted modeling strategy involving :

    i. development of customized regional biogeochemical process models that build on recent advances in eddy resolving models of the Arabian Sea and Bay of Bengal, and also incorporate the key nitrogen and N_2O 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_2O 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_2O 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_2O-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 2nd International Indian Ocean Expedition (IIOE-2: www.iioe-2.incois.gov.in),
537 SOLAS (Surface Ocean Lower Atmosphere Study: www.solas-int.org) and Future
538 Earth (www.futureearth.org).

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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 (NO_3^-) and ammonium (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/aeros
567 ol_rain/), and from sources cited in Table 1. The dry deposition fluxes of aerosol
568 WSON from the Arabian Sea were adopted from the mean value of the two cruises
569 sampled in Srinivas and Sarin [2013]. Given the aerosol concentration (C), the dry
570 deposition flux (F) was estimated using the following equation: F = C*Vd, and by
571 assuming deposition velocities (Vd) of 1 cm s^{-1} for NO_3^- [Duce et al., 1991] and 0.6 cm
572 s^{-1} for NH_4^+ [Spokes et al., 2000]. The Vd is difficult to quantify and is affected by
573 several factors such as wind speed, surface roughness and, particle size etc. Due to
574 the inherent uncertainties associated with the assumed dry deposition velocities, the
575 estimates of atmospheric dry deposition of nitrogen carry additional uncertainty by
576 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 (Fw) were estimated from the following formula:
580 Fw = P*S*Cd*ρ_a^{-1}*ρ_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]), Cd is
584 the aerosol concentration, and ρ_a and ρ_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° × 1° resolution), and then averaged to estimate wet and dry
588 deposition to the northern Indian Ocean.

589

Appendix B : Global biogeochemistry models used in analysis of 3.1

The investigations of Suntharalingam et al. [2012] and Jickells et al. [2017] both employed the PlankTOM ocean biogeochemistry model (PlankTOM5 and PlankTOM10 respectively) embedded in the NEMO ocean general circulation model, v 3.1 [Madec, 2008], and the diagnostic N_2O model of Suntharalingam et al. [2012]. The model analysis of Landolfi et al. [2017] employed the UVic2.9 Earth System Model, and the N_2O parameterization of Zamora and Oschlies [2014]. The model analyses all evaluated the impact of changes in external nutrient input from the pre-industrial to the present, but have differences in their representation of the specific nutrient inputs considered (deposition, riverine and N_2 fixation), and in their parameterization of the nitrogen cycle. Further details of model specifications and assumptions are given in the individual publications.

602

Appendix C: Estimation of N_2O Production and Impact of Nitrogen Deposition in section 3.2

For the regional model calculations of section 3.2, we derive a diagnostic estimate of the net N_2O source from the parameterizations of Suntharalingam et al. [2012, 2000] which estimate N_2O production as a function of organic matter remineralization via the local oxygen consumption rate. The N_2O production parameterizations are applied to the gridded biogeochemical model fields (specifically oxygen and oxygen utilization rates) of the high-resolution model simulations (1/12 degree) of Resplandy et al. [2012]. N_2O production is estimated from two separate pathways: (i) nitrification in the oxygenated open ocean, and (ii) higher N_2O yield processes (denitrification, enhanced nitrification) in low-oxygen zones. In addition, N_2O loss by denitrification is represented below a specified oxygen threshold ($[O_2] < [O_2]_{denit}$). Overall, the net N_2O production is estimated as:

Net N_2O production = $\alpha \cdot [O_2$ consumption$] + \beta \cdot f(O_2) \cdot [O_2$ consumption$]$ – [Anoxic N_2O loss]

Here, the scalar parameter α (mol [N_2O]/mol [O_2]) represents the N_2O yield from nitrification and is quantified from observed correlations between excess N_2O (ΔN_2O)and Apparent Oxygen Utilization (AOU). The parameter β represents the higher yield of N_2O in sub-oxic zones, and $f(O_2)$ represents the non-linear functional dependence of N_2O yield on oxygen level. N_2O loss in anoxic conditions is represented as (-1) x the value of total N_2O production below oxygen threshold levels of $[O_2]_{denit}$; i.e., all N_2O produced in these zones is assumed to be consumed by denitrification processes, and this represents an upper bound on N_2O loss in these zones. For the analyses of this section we take the threshold level of $[O_2]_{denit}$ as 5 μmol L^{-1}. Refer to Suntharalingam et al. [2012, 2000] and Buithenhuis et al. [2018] for additional details on the N_2O parameterization.

Table C1 presents the regionally aggregated results from the set of scenarios constructed for this analysis. These scenarios evaluate the sensitivity of estimated regional N_2O production to variation in the N_2O yield parameters α and β. The 'Standard' model takes α and β values from Suntharalingam et al. [2012]. In addition we evaluate scenarios based on N_2O measurement analyses from the Arabian Sea (Law and Owens [1990], Naqvi and Noronha [1991], and Patra et al. [1999]). For these scenarios, the N_2O yield parameters were derived from the ΔN_2O/AOU

636 correlations reported in the individual studies. Figure 1 presents the column
637 integrated distribution of N_2O 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_2O production by applying the regionally averaged
641 fractional change in N_2O yield for the Arabian Sea region (γ_{N2Odep} =0.125) from the
642 nitrogen deposition of Suntharalingam et al. [2012] to the N_2O production estimates
643 derived above for the regional model:
644 i.e.,

$$\text{Change in } N_2O = (\gamma_{N2Odep}) \times [N_2O \text{ production}]$$

645

646

647 **Table C1 : Regional model analysis of N_2O production scenarios and estimates of**
648 **the impact of nitrogen deposition**

649

N_2O Production Scenarios	N_2O Production (Tg N yr^{-1}) Nitrification Pathway	N_2O Production (Tg N yr^{-1}) Low-Oxygen Pathway	N_2O loss (Tg N yr^{-1})	Net N_2O production (Tg N yr^{-1})	Impact of Nitrogen Deposition on N_2O production [*] (Tg N yr^{-1})
Standard $\alpha=0.75\times10^{-4}$; $\beta=0.01$	**0.055**	**0.27**	**0.096**	**0.23**	**0.029**
LOW α β scenario $\alpha=0.5\times10^{-4}$; $\beta=0.005$	0.037	0.135	0.048	0.124	0.015
HIGH α β scenario $\alpha=2.0\times10^{-4}$; $\beta=0.02$	0.147	0.54	0.124	0.564	0.070
Law and Owens 1990 LOW scenario $\alpha=0.33\times10^{-4}$; $\beta=0.01$	0.024	0.27	0.097	0.197	0.025
Law and Owens 1990 HIGH scenario $\alpha=3.1\times10^{-4}$; $\beta=0.01$	0.228	0.27	0.098	0.40	0.05
Naqvi and Noronha 1991 scenario $\alpha=1.7\times10^{-4}$; $\beta=0.01$	0.125	0.27	0.097	0.298	0.037
Patra 1999 scenario $\alpha=1.5\times10^{-4}$; $\beta=0.01$	0.110	0.27	0.097	0.283	0.036

650

651 *Impact of nitrogen deposition estimated as Change = $\gamma_{N2Odep} \times N_2O$ 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

17

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

	Riverine	Atmospheric	N$_2$ Fixation
Arabian Sea			
Model-derived	0.4[a]	1.4[d]	2.6[f]-6.2[g]
Obs-based	0.1-0.4[b]	1.8[e]	0.0-20.4[h,i]
Bay of Bengal			
Model-derived	5[a]	1.1[d]	4.2[f]-4.3[g]
Obs-based	0.9-2.0[c]	1.6[e]	0.6-11.3[i]

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

[b][*Singh and Ramesh*, 2011]; [*Singh et al.*, 2012]

[c][*Srinivas and Sarin*, 2013] and references therein.

[d]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.

[e]This study, see Appendix A for further details.

[f][Jickells et al. 2017]

[g][Le Quéré et al 2016]

[h][Gandhi et al., 2011] estimated N$_2$ fixation rates using ^{15}N$_2$ with less than 6% uncertainty.

[i][*Srinivas and Sarin*, 2013] and references therein, converted to the larger open ocean regions used in this study (6.6x10^6 km^2 for the Arabian Sea, and 3.0x10^6 km^2 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_2O 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_2O source * (Tg N yr^{-1})	Change in N_2O source due to nitrogen inputs (pre-industrial to present day) (Tg N yr^{-1})
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^oE-100^oE.

Figure 1. Net column-integrated N_2O production (units : Mg N m^{-2} yr^{-1}) in the Arabian Sea derived from the regional high-resolution model analysis reported in section 3.2. The net N_2O production is derived from the methodology outlined in Appendix C and is the sum of N_2O production from the nitrification and low-oxygen pathways, and N_2O loss below an oxygen threshold of 5 µmol 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_2 data [Bianchi et al. 2012].

Figure 1. Net column-integrated N_2O production (units : Mg N m^{-2} yr^{-1}) in the Arabian Sea derived from the regional high-resolution model analysis reported in section 3.2. The net N_2O production is derived from the methodology outlined in Appendix C and is the sum of N_2O production from the nitrification and low-oxygen pathways, and N_2O loss below an oxygen threshold of 5 µmol L^{-1}.
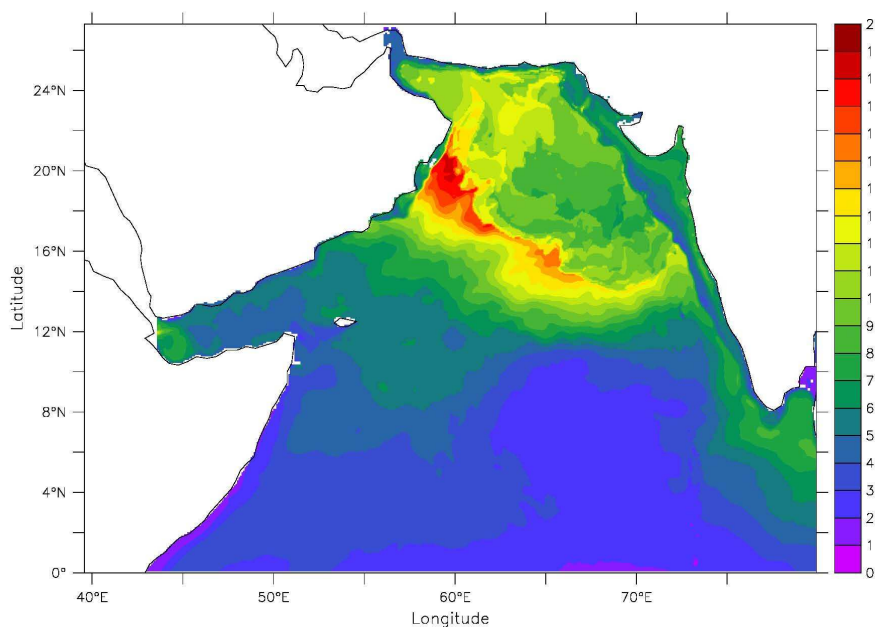
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