97

REPORTS AND STUDIES

GESAMP
Joint Group of Experts on the
Scientific Aspects of Marine
Environmental Protection

THE MAGNITUDE AND IMPACTS OF ANTHROPOGENIC ATMOSPHERIC NITROGEN INPUTS TO THE OCEAN

GESAMP REPORTS AND STUDIES No. 97
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CONTENTS

EXECUTIVE SUMMARY

Nitrogen (N) is an essential macro-nutrient in marine ecosystems. Reactive nitrogen, N_r, enters the ocean through rivers, streams, groundwater, wastewater treatment facilities, and atmospheric deposition. Reactive nitrogen is also formed in the ocean through the fixation of dissolved N_2 gas by plankton known as diazotrophs. Human activities have significantly altered the input of N_r to both the coastal and open ocean, potentially leading to changes in marine biogeochemical cycles and perturbations of ocean ecosystems. Recent scientific and policy interest has focused on the input and subsequent impact of anthropogenic atmospheric N species on marine biogeochemistry. These inputs include both oxidized and reduced inorganic forms of N as well as poorly characterized organic nitrogen (ON) compounds. Several previous studies have addressed this issue. However, as a result of the increasing awareness of the potential biogeochemical impacts of the deposition of anthropogenic atmospheric N_r species, the availability of new data as well as recent model developments, it was determined that a fresh evaluation of this issue was in order. For this reason, the Joint Group of Experts on the Scientific Aspects of Marine Environmental Protection (GESAMP) asked the Working Group on The Atmospheric Input of Chemicals to the Ocean (WG 38) to address a series of new Terms of Reference. In abbreviated form, these were as follows:

- Update the geographical estimates of atmospheric anthropogenic nitrogen deposition to the global ocean;
- Re-evaluate the magnitude and impact of atmospheric nitrogen deposition on marine biogeochemistry;
- Provide a more reliable estimate of the impact of atmospheric anthropogenic nitrogen deposition on the production of additional nitrous oxide in the ocean and its subsequent emission to the atmosphere;
- Evaluate the extent to which anthropogenic nitrogen, delivered to the coastal zone via rivers, is transported to the open ocean; and
- Make a detailed estimate of the input and impact of anthropogenic nitrogen in the area of the Northern Indian Ocean and the South China Sea.

To address these new Terms of Reference, a highly successful and productive workshop, “The Atmospheric Deposition of Nitrogen and Its Impact on Marine Biogeochemistry” was held at the University of East Anglia in Norwich, United Kingdom from 11 to 14 February 2013. Six scientific papers resulting from the deliberations at that workshop have been published and a seventh will be submitted later in 2018. Summaries of each of those seven papers are presented in this report. Following are some of the primary findings from those papers.

Atmospheric N deposition to the ocean was estimated using the TM4-ECPL atmospheric chemistry model, which explicitly includes organic nitrogen (ON) deposition. The deposition fields estimated by this model for the inorganic N are consistent with other independently developed model estimates. The total atmospheric N_r input to the ocean, including the shelf seas, is estimated to be 39 TgN y^{-1} in 2005, with >30 TgN y^{-1} of this reaching the open ocean (1Tg = 10^{12}g or 10^6 tonnes). The models suggest that combustion processes are the primary source of oxidized nitrogen (HNO_3 and NO_3^- predominantly) - 55% from terrestrial anthropogenic combustion, 11% from biomass burning and 11% from shipping, with the remainder from natural sources. Almost two thirds of the reduced nitrogen (NH_3 and NH_4^+ predominantly) comes from agriculture. The ON of anthropogenic origin has increased from about 7% of the total ON in 1850 to about 30% of the total in 2005. However, the model suggests that about a quarter of the total input of reduced and organic N to the ocean arises from recycled material from the ocean itself, and therefore it does not constitute a net new N input to the ocean. The net atmospheric deposition to the oceans estimated here is thus significantly lower than that estimated by Duce et al. (2008), largely as a result of the now-recognized recycling of organic nitrogen and ammonia (NH_3). Atmospheric N inputs to the oceans are particularly large downwind of major source regions, especially in the western north Atlantic and north Pacific.

The present global modelling study of the N_r atmospheric cycle and N_r deposition to the ocean is the first that evaluates past, present and future N_r atmospheric deposition accounting for ON primary sources as well as for secondary ON chemical formation as a N-dependent process. While the total N_r deposition to the ocean is not expected to change significantly by 2050, the relative importance of oxidized and reduced N is expected to change, with an increasing proportion of ammonium compared to nitrate, resulting from more efficient controls on terrestrial emissions of nitrogen oxides (NO_x) compared to ammonia. This would result in a change in the acidity of the atmospheric deposition.

In order to evaluate more accurately the importance of atmospheric deposition of N to the ocean, a separate study was initiated to evaluate the riverine input of N_r to the ocean. It was determined that ~23 TgN y^{-1} as dissolved inorganic N enters the ocean via rivers, with ~17 TgN y^{-1} reaching the open ocean. There is an additional input of about 11 TgN y^{-1} from rivers as dissolved (DON), and the effectiveness of the shelf sea trapping of this organic component is uncertain. Atmospheric nitrogen inputs are thus more effective at delivering anthropogenic N to the open ocean than rivers, because of the role of the coastal ocean in trapping fluvial nitrogen. These estimated amounts of atmospheric and fluvial N inputs can be compared with our current estimate of 164 TgN y^{-1} for biological N fixation within the global ocean. Hence riverine and atmospheric inputs, which are both predominantly anthropogenic, are almost comparable in magnitude to natural inputs of N to the ocean predominantly by nitrogen fixation. An important uncertainty identified in the work of WG 38 is the extent to which this biological fixation might be suppressed by anthropogenic N inputs, since in terms of biochemical energy considerations planktonic organisms can be expected to use the most readily available N of nitrate and ammonium inputs rather than expend energy to fix N. Such suppression has the potential to provide a short-term negative feedback that would reduce the impacts of atmospheric N deposition on the oceans, but the scale of local increases in N deposition and particularly ambient concentrations, which trigger this feedback, are very uncertain.

Models represent the only method to derive global scale estimates of atmospheric deposition to the oceans, but these models need evaluation. Atmospheric deposition is routinely measured at many land stations around the world, including World Meteorological Organization (WMO) Global Atmosphere Watch (GAW) stations, and this can be used to validate the models. However, the collection of such model validation data at sea is more challenging. In order to evaluate the performance of atmospheric N deposition models, a database of measured aerosol nitrate (NO_3^-) and ammonium (NH_4^+) concentrations obtained on-board ships over the global ocean was compiled and compared to model simulation results. The database contained measurements of ~2900 samples collected from 1995–2012 and details of access to this database are available within the summary of paper 2.6 in this report. The model data comparisons suffered due to the scarcity of observational data and the large uncertainty in dry deposition velocities, v_d, used to derive deposition fluxes from concentrations. When considering modelled dry N_r deposition, the uncertainty in v_d probably implies that fluxes can be estimated to within no better than one order of magnitude. Uncertainties in v_d have been a major limitation on estimates of the flux of material to the oceans for several decades and we provide recommendations to help improve the situation. Overall the agreement of model and data based deposition estimates is very good at large ocean basin scales, but becomes less good at higher spatial resolution. It is therefore important to consider the time and space scales of interest when determining the appropriate model products being used.

Our Terms of Reference included focusing particularly on some regions where atmospheric deposition is especially high and changing. In one regional study of the impact of N deposition, the analysis of datasets for atmospheric N_r deposition, satellite chlorophyll-*a* (Chl-*a*), and air mass back trajectories revealed that transport of N originating from the populated east coasts of China and Indonesia and its deposition to the ocean contributed approximately 20% of the annual biological new production in the South China Sea. The airborne contribution of N_r to new production in this region is expected to grow considerably in the coming decades. Another regional study as part of the WG38 activities focused on the Northern Indian Ocean, which is another area of particular high and growing nitrogen deposition. The issue in this region is not just that this atmospheric deposition can stimulate increased ocean productivity, but also because the circulation of deep waters within the Northern Indian Ocean gives rise to one of the largest low oxygen regions of the world ocean. This is a natural phenomenon, but the increased atmospheric N_r deposition in this region can stimulate increased phytoplankton production, and the subsequent sinking of extra organic matter into deep ocean waters are predicted to increase this globally important region of low oxygen. Model calculations conducted as part of the WG 38 activities discussed below confirm this prediction and the associated increased production of the greenhouse gas N_2O.

Most ocean primary productivity is driven by internal recycling of the vast reservoirs of nutrients in the deep ocean. This productivity ultimately sustains life in the oceans and also mediates a vast exchange of carbon dioxide between the oceans and atmosphere. Ocean productivity depends on the availability of light and a range of nutrients beside N, particularly phosphorus and iron, and the iron also has an important atmospheric source. WG 38 deliberations focused on N, but the ocean modelling incorporated the role of all the factors limiting productivity. We estimate that atmospheric N deposition increases global ocean carbon sequestration via the biological pump by 0.15 Pg C y^{-1}, which is important within the context of an estimated ocean annual total C (as CO_2) uptake of 2.2 ± 0.5 Pg C y^{-1}. This estimated enhancement of carbon uptake is smaller than an estimate made earlier by members of WG 38 (Duce et al. 2008), reflecting lower net atmospheric nitrogen deposition estimates, particularly due to recycling of ammonia and organic nitrogen between the atmosphere and ocean. Increases in atmospheric deposition of nutrients have the potential to increase ocean productivity and thereby the sinking of organic matter into the deep ocean, thus increasing areas of low oxygen in the deep ocean, causing increases in denitrification (the breakdown of organic matter in areas of low oxygen using nitrate rather than oxygen as an oxidising agent) and the resulting emission of the greenhouse gas N_2O. However, we estimate that the increase in this N_2O flux will be small (1 to 3%) on the global scale, but perhaps 5-20% in the Arabian Sea region under likely scenarios over coming decades to 2050. A response has been identified in which increased ocean denitrification can create a negative feedback on the longer timescales (100s to 1000s of years) causing a decline in ocean nitrogen inventory and thus limiting the potential impact of atmospheric N_r deposition on marine productivity. The significance of this feedback clearly merits further study, particularly in Earth system models evaluating the longer term functioning of the ocean system.

Through these papers significant advances have been made in our understanding of atmospheric nitrogen deposition and its impacts on ocean biogeochemistry. A number of scientific gaps remain, providing new opportunities and challenges. Therefore we recommend additional work on:

- The sources of atmospheric organic nitrogen;
- The magnitude and significance of recycling of ammonia and organic nitrogen from the oceans;
- The parameterization of deposition, particularly dry deposition;
- An improved database, particularly on wet deposition over the oceans;
- The extent and thresholds for suppression of nitrogen fixation by ambient surface water dissolved nitrogen;
- The retention of nitrogen within shelf systems, and particularly how the rates of bacterial processes that convert fixed nitrogen back to dinitrogen gas (denitrification and the related anammox both of which convert N_r into inert nitrogen gas) vary with temperature;
- The impacts of possible change in aerosol acidity over coming decades on the atmospheric delivery of nutrients to the oceans; and
- Increasing atmospheric carbon dioxide concentrations are already driving ocean acidification and this is likely to increase in coming decades. The impacts of the changing ocean pH on ocean emissions of atmospherically important gases including nitrogen species, but also other gases, needs to be evaluated.

We also recommend further work in the following regions, which are particularly sensitive to likely changes in atmospheric deposition:

- The Northwest Pacific where deposition fluxes are expected to grow and where there may already be impacts from the current inputs;
- The Northern Indian Ocean, an important source region for N_2O, which receives a large atmospheric input that is argued to already be increasing plankton productivity; and
- Areas of the Mediterranean and North Atlantic where primary production is phosphorus- or iron-limited and hence where additional nitrogen deposition may lead to different nutrient biogeochemical responses to those in other ocean areas where nitrogen is the primary limiting nutrient.

1. INTRODUCTION

1.1 Brief history of Working Group 38 and early results

In the 1980s, GESAMP formed a working group sponsored by the World Meteorological Organization (WMO), the Intergovernmental Oceanographic Commission of the United Nations Educational, Scientific, and Cultural Organization (IOC-UNESCO), and the United Nations Environment Programme (UNEP) that developed a comprehensive review of the input of atmospheric trace species to the global ocean (GESAMP, 1989). That benchmark effort led to a scientific publication in *Global Biogeochemical Cycles* in 1991 (Duce et al., 1991) that for more than 15 years was the state-of-the-art reference in this area, leading to over 1500 citations in the literature. By 2008 that paper was quite out of date, and growing concern about the impact of atmospheric deposition of both natural and anthropogenic substances on ocean chemistry, biology, and biogeochemistry as well as climate led to the formation of a Working Group on The Atmospheric Input of Chemicals to the Ocean (WG 38).

Between its formation in 2008 and 2013, WG 38 held meetings at the University of Arizona, Tucson, AZ, in 2008; at the International Maritime Organization (IMO) in London in 2010; and in Malta in 2011. Sponsors of those WG 38 efforts have included the Global Atmosphere Watch (GAW) programme of WMO, IMO, the International Council for Science (ICSU)'s Scientific Committee on Oceanic Research (SCOR), the Swedish International Development Agency (SIDA), the European Commission Joint Research Centre, the University of Arizona, the International Environment Institute at the University of Malta, and the US National Science Foundation. Following the initial Terms of Reference and the above-mentioned meetings, described in detail in GESAMP Reports and Studies 84, *The Atmospheric Input of Chemicals to the Ocean*, a series of scientific papers were published in the peer-reviewed scientific literature. These were as follows:

[1] Okin, G., A.R. Baker, I. Tegen, N.M. Mahowald, F.J. Dentener, R.A. Duce, et al., 2011: "Impacts of atmospheric nutrient deposition on marine productivity: roles of nitrogen, phosphorus, and iron", *Global Biogeochemical Cycles*, 25, GB2022, doi:10.1029/2010GB003858.

[2] Hunter, K.A., P.S. Liss, V. Surapipith, F. Dentener, R A. Duce, M. Kanakidou, et al., 2011: "Impacts of anthropogenic SO_x, NO_x and NH_3 on acidification of coastal waters and shipping lanes", *Geophysical Research Letters,* 38, L13602, doi:10.1029/2011GL047720.

[3] Kanakidou, M., R. Duce, J. Prospero, A. Baker et al., 2012: "Atmospheric fluxes of organic N and P to the ocean", *Global Biogeochemical Cycles,* GB3026, doi:10.1029/2011GB004277.

[4] Schulz, M., J.M. Prospero, A.R. Baker, F. Dentener, L. Ickes, P.S. Liss et al., 2012: "The atmospheric transport and deposition of mineral dust to the ocean - Implications for research needs", *Environmental Science and Technology*, 46, doi:10.1021/es30073ul, 10,390-10,404.

[5] Hagens, M., K.A. Hunter, P.S. Liss and J.J. Middelburg, 2014: "Biogeochemical context impacts seawater pH changes resulting from atmospheric sulfur and nitrogen deposition", *Geophysical Research Letters*, 41, doi:10.1002/2013GL058796.

1.2 The nitrogen study and its Terms of Reference

Although the early work of Working Group 38 did consider some aspects of the deposition and impacts of atmospheric nitrogen species on the ocean, it was recognized that this was a significant and complex scientific issue that required a more in depth study. Thus, at GESAMP's 39th session in 2012, members approved additional Terms of Reference for continued work of GESAMP WG 38 to address issues related to the impact of the atmospheric deposition of anthropogenic nitrogen to the ocean. The additional Terms of Reference were as follows:

Terms of Reference

1. Update the geographical estimates of anthropogenic nitrogen deposition to the global ocean made in the 2008 paper in *Science (*Duce, R.A. et al., 2008: "Impacts of atmospheric anthropogenic nitrogen on the open ocean", *Science,* 320, 893-897), which were based on data from 2005 or earlier. This would utilize newer and more geographically distributed data on anthropogenic atmospheric nitrogen concentrations and deposition over the global ocean as well as improved models of these processes and impacts.

2. Considering issues related to Task 1 above, re-evaluate the impact of atmospheric nitrogen deposition on marine biogeochemistry, including re-estimating the amount of CO_2 that could be drawn down from the atmosphere into the ocean as a result of the increased productivity in the ocean derived from the additional anthropogenic nutrient nitrogen deposited. This would allow an update on the

impact of the atmospheric nitrogen deposition on atmospheric radiative properties outlined in the 2008 *Science* paper.

3. Provide a more reliable estimate of the impact of atmospheric anthropogenic nitrogen deposition on the production of additional nitrous oxide in the ocean and its subsequent emission to the atmosphere. This was one of the greatest uncertainties in the 2008 *Science* paper.

4. Evaluate the extent to which anthropogenic nitrogen delivered to the coastal zone via rivers, atmospheric deposition, etc. is transported to the open ocean, in which regions this may happen, and what its impact is there. In the 2008 *Science* paper it was assumed that all nitrogen delivered to the coastal zone was sequestered there and did not reach the open ocean, but this may not be true in all locations.

5. Make a more detailed estimate of the input and impact of anthropogenic nitrogen in the area of the Northern Indian Ocean (Arabian Sea, Bay of Bengal) and the South China Sea - the areas that are expected to show the greatest increase of anthropogenic nitrogen deposition over the next few decades.

To address these new Terms of Reference, a highly successful and productive workshop on “The Atmospheric Deposition of Nitrogen and Its Impact on Marine Biogeochemistry” was held at the University of East Anglia in Norwich, United Kingdom, from 11 to 14 February 2013. Twenty-three scientists participated in the workshop. The first day of the workshop was devoted to discussions of the five tasks identified above as the foci of the workshop. Two participants were asked to summarize the issues in each of these task areas and to lead the discussions that followed. On the basis of the task area discussions, the workshop participants broke up into subgroups on the second through fourth days of the workshop. These subgroups began the development of a number of scientific papers covering the task areas above. Six of these papers have been published in the peer-reviewed scientific literature and a seventh paper will be submitted later in 2018. Summaries of the scientific content of these seven papers are given in Section 2 below in the order of their publication. These summaries have been prepared by the lead authors and the style and focus of each is slightly different and designed to accentuate the key new findings in each particular paper. This nitrogen workshop was supported by WMO, IMO, the University of East Anglia and the US National Science Foundation through SCOR.

2. SUMMARIES OF THE SEVEN PAPERS DEVELOPED BY WG 38

2.1 Summary of "Impact of Atmospheric Nitrogen Deposition on Phytoplankton Productivity in the South China Sea"

(Tae-Wook Kim, Kitack Lee, Robert Duce and Peter Liss)

This paper was published in *Geophysical Research Letters*, 41, 3156-3162, doi: 10.1002/2014GL059665. (2014).

Introduction

The South China Sea (SCS) is adjacent to several rapidly growing economies, including China, Viet Nam, and Indonesia. Consequently, it receives substantial amounts of anthropogenic nitrogen (N) from these countries through riverine and atmospheric inputs. Furthermore, during the coming decades anthropogenic N deposition into the SCS is projected to increase at a rate exceeding that for other marginal seas (Duce et al., 2008). Phytoplankton productivity in the SCS is relatively low compared with other adjacent marginal seas (e.g. the East China Sea and the Yellow Sea) because it is located in a subtropical zone where the nutrient flux from deep water (a major nutrient source for supporting ocean productivity) is low. The combination of high levels of atmospheric N input and low existing ocean productivity (because of low rates of nutrient supply from deep water (Wong et al., 2007) makes the SCS more vulnerable to anthropogenic N deposition, and thus makes it an important location for investigating the impact of anthropogenic N deposition on ocean productivity. A previous study showed that atmospheric N deposition to the SCS accounted for 6-20% of the sinking nitrogen flux in the 1990s and that this proportion will increase to 15-30% by the 2050s (Krishnamurthy et al., 2007).

Results

Through analysis of multiple datasets, including those for air mass back trajectories, chlorophyll-a (Chl-*a*) and sea surface temperature (SST), we tested the hypothesis that N pollutants act to increase phytoplankton production in the SCS. We also estimated the contribution of N deposition to new production in the SCS. New production, is the plankton production sustained by external inputs rather than that sustained by internal recycling.

Air monitoring sites were grouped into urban, rural and remote stations to better distinguish the impact of pollutant N inputs on the sites selected for the study (Figure 1). The mean rates of wet N_r deposition (± the standard deviation) calculated for 16 urban, 7 rural, and 8 remote sites were 100 ± 52, 84 ± 55 and 50 ± 29 mmol N m^{-2} y^{-1}, respectively. For dry N_r deposition, the mean rates were 38 ± 34, 10 ± 5 and 5 ± 3 mmol N m^{-2} y^{-1} for the urban, rural and remote sites, respectively, values that were considerably lower than the corresponding rates of wet deposition. As a result, the ratio of wet to total (wet + dry) deposition was in the range 72-91%, with the lowest values found in the urban sites. The contributions of nitrate (NO_3^-) and ammonium (NH_4^+) to wet reactive nitrogen (N_r) deposition were approximately 36% and 64%, respectively. However, the contribution of particulate phase NO_3^- was a factor of two greater than that of the corresponding NH_4^+ contribution because of the greater association of NO_3^- with coarse particles (Nakamura et al., 2005).

In the mid-19th century, atmospheric N_r deposition to South-East Asia was 4-14 mmol N m^{-2} y^{-1} (Duce et al., 2008), which is substantially lower than the values found for the remote sites. The increase in N_r deposition at the remote sites and the contrast in N_r deposition between urban and remote sites (Figure 1) both indicate that the main source of N_r to all three sites (urban, rural and remote) was most likely human activities (use of fertilizers and fossil fuels). It is also noteworthy that the amount of NH_4^+ deposition in South-East Asia is considerably greater than that of NO_3^- deposition. The primary sources of atmospheric NH_4^+ are emissions from cropland and livestock, whereas atmospheric NO_3^- mostly arises from emissions from fossil fuel combustion engines, which are effectively controlled by legal regulations. This implies that anthropogenic N_r release into the atmosphere of South-East Asia may be difficult to control because of difficulties associated with reducing ammonia production from agricultural sources.

New production in the SCS was estimated to be 50-80 mg C m^{-2} d^{-1} based on satellite-derived primary productivity measurements multiplied by a published *f*-ratio (0.2), which is the ratio of new production to primary production (Yool et al., 2007) (Figure 2a), and is equivalent to 208-332 mmol N m^{-2} y^{-1} if a C:N ratio of 117/16 is used (T.-W. Kim et al., 2014). As a result, the total N deposition (organic + inorganic; ~55 mmol N m^{-2} y^{-1}) into the SCS reached approximately 20% of new production in the oligotrophic waters of the SCS (Figure 2c). This indicates that atmospheric N deposition is an important external source of N into the SCS. If this value is extrapolated to the entire oligotrophic SCS (~1.4×10^{12} m^2), the total N flux into this oligotrophic area is ~77 Gmol N y^{-1}. The combined N inputs (57-76 mmol N m^{-2} y^{-1}) from atmospheric deposition and N_2 fixation into the oligotrophic SCS contributed 20-35% of the new production in the oligotrophic SCS. Nonetheless, the vertical flux of N from deep water provided the N required for sustaining the remaining 65-80% of new production.

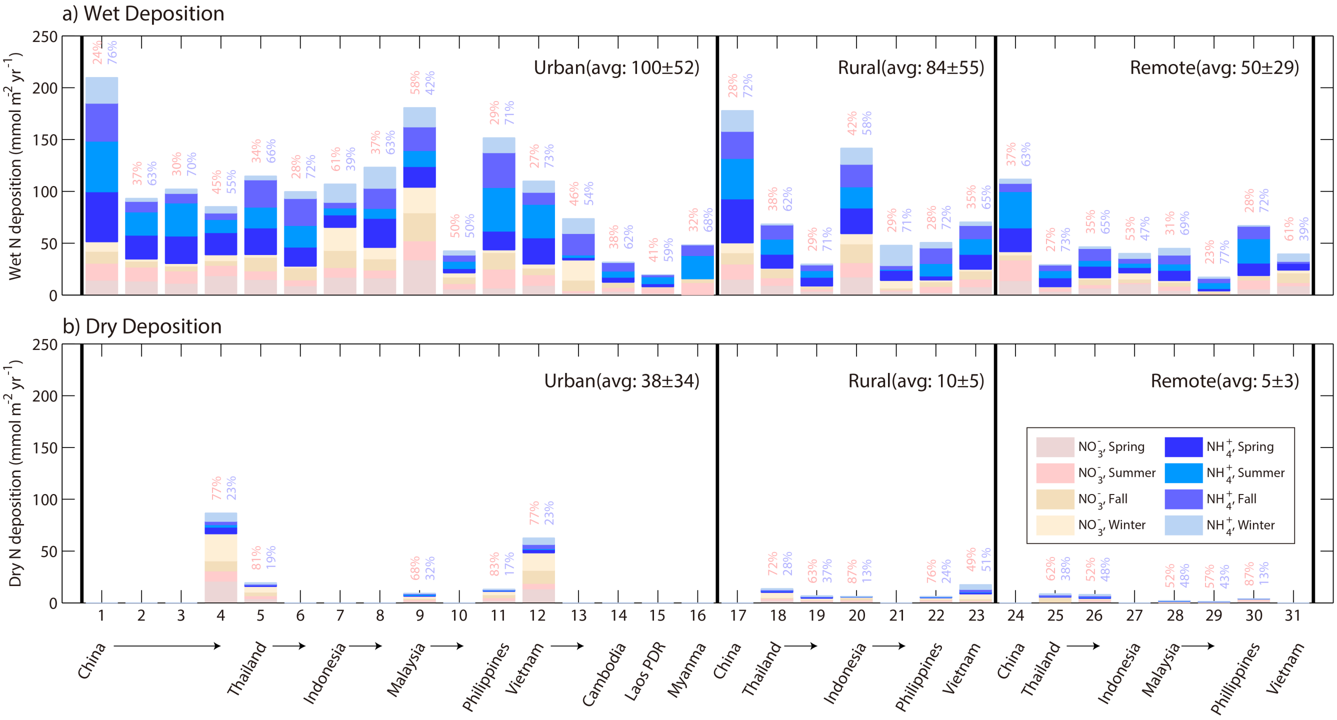

Figure 1. Wet (a) and dry (b) atmospheric N deposition in South-East Asia (mmol m^{-2} y^{-1}). Monitoring locations were grouped into urban, rural and remote sites (thick vertical lines). The numbers above the bars indicate the per cent deposition of NO_3^- (red) and NH_4^+ (blue). Seasonal mean values are presented using different colours (see the legend in Figure 1b). The locations of monitoring sites are shown in Figure 2a.

Source: T.-W. Kim et al., 2014

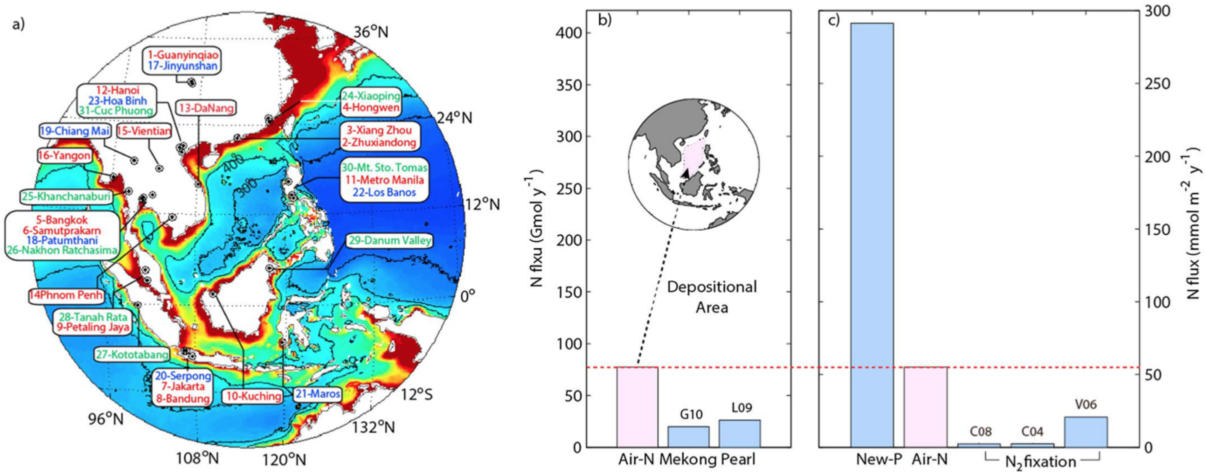

Figure 2. a) Primary production (contours: mg C m^{-2} d^{-1}; available at http://www.science.oregonstate.edu/ocean.productivity) and the Acid Deposition Monitoring Network in East Asia (EANET) monitoring sites adjacent to the SCS. The numbers assigned to the monitoring site names in a) are the site IDs shown in the *X*-axis in Figure 1. The red, blue and green characters indicate urban, rural and remote sites, respectively. b) Atmospheric N deposition (Air-N) to the oligotrophic SCS (marked by pink colour in the inset) and N fluxes from the Mekong and Pearl rivers. c) Estimated nitrogen necessary to support new production (New-P), atmospheric N deposition estimated in this study, and N_2-fixation from published literature (Grosse et al., 2010; Liu et al., 2009; Chen et al., 2008; Chen et al., 2004; Voss et al., 2006).

Source: T.-W. Kim et al., 2014

Analyses of multiple datasets - atmospheric N deposition data spanning 10 years, satellite-derived ocean productivity data, and air mass back trajectory data — reveal that anthropogenic N contained in air masses originating from populated areas enhanced Chl-*a* in the SCS. The implications of our results are not limited to the SCS because atmospheric N deposition into the ocean is a global phenomenon that will be particularly serious in ocean waters located downwind of Europe, North America and Asia generally. Future studies of this issue should focus in particular on changes in phytoplankton composition and the fate of newly supplied N. Such studies will improve understanding of the impacts of N deposition on ecosystems, biogeochemical cycles and climate change, including changes in the abundance of N_2 fixing microorganisms, the relative availability of nutrients, and N_2O production.

Conclusions

Human-induced alteration of the N cycle is one of the major global environmental threats in this century. The production of agricultural fertilizers transforms N_2 gas into reactive N ($N_r = NO_x + NH_y$), but much of the N_r generated is inadvertently released into the atmosphere (predominantly as NH_3) and rivers. The burning of fossil fuels also emits oxidized N_r into the atmosphere; approximately half of the anthropogenic N_r emitted to the atmosphere is eventually deposited to the ocean with the remainder onto land. The impacts of anthropogenic N deposition on the marine N cycle are only now being revealed, but the magnitudes of those impacts are largely unknown in time and space. The SCS is particularly subject to high anthropogenic N deposition because the adjacent countries are highly populated and have rapidly growing economies. Analysis of datasets for atmospheric N deposition, satellite Chl-*a*, and air mass back trajectories reveal that transport of N originating from the populated east coasts of China and Indonesia, and its deposition to the ocean, has been responsible for enhancements of Chl-*a* in the SCS. We found that atmospheric N deposition contributed approximately 20% of the annual biological new production in the SCS. The airborne contribution of N to new production in the SCS is expected to grow considerably in the coming decades.

2.2 Summary of "Past, Present and Future Atmospheric Nitrogen Deposition"

(Maria Kanakidou, Stelios Myriokefalitakis, Nikos Daskalakis, George Fanourgakis, Athanassios Nenes, Alex R. Baker, Kostas Tsigaridis and Nikolaos Mihalopoulos)

This paper was published in the *Journal of the Atmospheric Sciences,* 73, 2039-2047, DOI: 10.1175/JAS-D-15-0278.1. (2016).

Introduction

Reactive nitrogen (N_r) is emitted into the atmosphere in the form of inorganic (mainly nitrogen oxides, ammonia) and organic nitrogen (both reduced and oxygenated compounds). These emissions that have natural and anthropogenic origin are increasing due to human activities and as a consequence the atmospheric N deposition to the ocean and its impact on the marine environment are also changing. The human-induced atmospheric N deposition to the oceans was estimated by Duce et al. (2008) to account for up to about 3% of the global annual new oceanic primary productivity. However, that work was based mainly on inorganic nitrogen deposition model estimates and a rough estimate of the organic nitrogen deposition flux to the ocean.

Recently, Kanakidou et al. (2012) have developed a novel methodology to account for the organic nitrogen (ON) global atmospheric cycle and its deposition to the ocean in a global chemistry-transport model, accounting for both primary and secondary ON sources, and they have simulated the present day reactive nitrogen atmospheric deposition, accounting for ON. In particular, the primary emissions of ON have been estimated based on the particulate organic carbon (OC) emissions and the observed ratios of ON:OC in the various source categories, as reviewed in the supplementary material of the paper by Kanakidou et al. (2012). By applying that methodology to the past (from the Atmospheric Chemistry and Climate Model Intercomparison Project – ACCMIP - Lamarque et al., 2013) and future (from Representative Concentration Pathway – RCP- emission scenarios – van Vuuren et al., 2011) emission databases for the combustion emissions of OC and to the natural emissions as for the year 2005, the present work provides the first estimate of the evolution of the ON primary source strength, as summarized in Table 1 below (supplementary S1 in Kanakidou et al., 2016). This estimate does not account, however, for the year-to-year variability of the natural ON source. It indicates a preindustrial (1850) total ON source 25% lower than in 2005 and projects a small (less than 10%) decline in the total ON sources in the future (2050), driven by the anthropogenic source evolution. The ON of anthropogenic origin has increased from about 7% of the total in 1850 to about 30% of the total in 2005.

Table 1. Sources of primary ON (in Tg-N y^{-1}) taken into account in the global chemistry transport model TM4-ECPL following the methodology by Kanakidou et al. (2012) (supplementary Table S1 in Kanakidou et al., 2016).

	1850	*2005*	*2050 based on*	
Based on OC emissions from:	ACCMIP	RCP6.0	RCP6.0	RCP8.5
Combustion ON	**1.3**	**7**	**6.8**	**5.6**
Fossil fuel	0.9	2.6	2.2	2.1
Biomass burning	0.4	4.4	4.6	3.5
Natural ON	**16.1**	**16.1**	**16.1**	**16.1**
Vegetation/ primary biogenic particles	8.8	8.8	8.8	8.8
Soils	0.4	0.4	0.4	0.4
Ocean	6.9	6.9	6.9	6.9
TOTAL ON	**17.4**	**23.1**	**22.9**	**21.7**

With regard to primary inorganic nitrogen emissions, the historical emissions from the ACCMIP database show significant increases both in NOx (NO_2+NO gas) and NH_3 emissions. However, projections to the future (RCPs) indicate a decline in NOx emissions due to efforts to limit air pollution, while NH_3 emissions will continue to grow (see Figure 3, modified from Kanakidou et al., 2016). This figure also shows that ON has been a more important fraction of N_r emissions in the past than nowadays. Noticeable in Figure 3 are the differences in the projections between RCP 6.0 and RCP 8.5, which both show the above mentioned opposite emission trends in NOx and NH_3 but different overall change in the total inorganic nitrogen emissions (sum of NOx and NH_3).

During atmospheric transport, N_r compounds are chemically transformed before being deposited to the surface, partially contributing to the ON atmospheric pool and deposition. Therefore, the present study also accounts for N-containing secondary organic aerosols (SOA) that are formed as salts of marine amines or by oxidation of other volatile organic compounds (VOC). Compared to recent ON global modelling studies (Kanakidou et al., 2012; Ito et al., 2014, 2015) the present work takes into account the NOx availability when computing the formation of secondary ON. Precisely, it is here assumed that SOA formed under high NOx-to-VOC conditions contains ON (Kanakidou et al., 2016). Furthermore, due to the high uncertainty in, and potentially minor importance of, the sources of amines of continental origin, these have been neglected in this study.

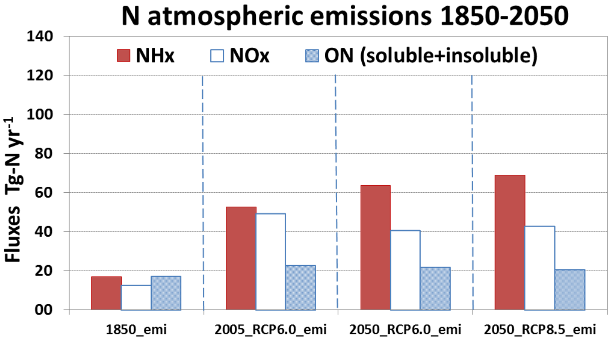

Figure 3. Global total emissions of NH_3 and NOx and those of particulate organic nitrogen (ON) used for this study for 1850, 2005 and 2050 from historical ACCMIP database and RCP6.0, RCP8.5 emissions (marked by 'year' emi). Natural emissions are kept constant as for the year 2005. (After Kanakidou et al., *Journal of the Atmospheric Sciences,* 73, 2039-2047, DOI: 10.1175/JAS-D-15-0278.1. (2016)).

Source: ©American Meteorological Society

Innovation compared to earlier studies

The present global modelling study of the reactive N atmospheric cycle and N_r deposition to the ocean is the first study that evaluates past, present and future N_r atmospheric deposition accounting for ON primary sources as well as for secondary ON chemical formation as a N-dependent process.

These considerations increase the total reactive N atmospheric source by about 20% compared to the source (inorganic N only) commonly taken into account in global models. In addition, secondary sources of gaseous and particulate ON (~4 to 13 Tg-N y^{-1}) are estimated to be 15-35% of the global total present day ON source. During this chemical formation of ON in the atmosphere, about 10% of primary NOx emissions are estimated to be transformed to ON. Only the part of the fraction of secondary ON source that leads to the formation of organic nitrates and peroxyacetyl nitrate-like (PANs) gases is taken into account in most global models. In this work this is estimated to be about 22% of the secondary source of ON. This in turn accounts for between ~3% and 10% of the total ON source.

Furthermore, the present study accounts for an important amount of marine N-recycling over the oceans. In particular, NH_3 marine emissions introduced in the model range between ~ 12 and 48% of the total NH_3 emissions, which are themselves 8-28% of the total inorganic N emissions. Primary marine ON is estimated to be roughly 30-40% of all primary ON emissions in the model. Oceanic emissions of N_r are however very uncertain and deserve further studies.

Methods

Ten simulations have been performed with the global atmospheric chemistry-transport model TM4-ECPL to calculate the atmospheric cycle of reactive nitrogen and derive the global distribution of total nitrogen deposition. For the first time, both inorganic and organic fractions of N deposition have been calculated for past and projected changes due to anthropogenic activities. The simulations have all been performed using ERA interim meteorology for the year 2005 (with appropriate spin-up time), the natural emissions corresponding to this year (desert dust, marine aerosol, biogenic terrestrial organic gases and aerosol emissions) calculated on or offline (as described in Myriokefalitakis et al. (2015) but for the year 2005), and the anthropogenic and biomass burning from ACCMIP historical and RCP6.0 and RCP8.5 emissions scenarios. Emissions for ON are calculated as described above, and more details can be found in Kanakidou et al. (2012) and (2016). NO_y is calculated by TM4-ECPL as the sum of NO_x, HNO_3, NO_3^-, HONO, HNO_4 and N_2O_5. Organic nitrates and PAN-like compounds that are reported in most models as NO_y are here included in the ON pool as appropriate.

Summary of results

Accounting for organic nitrogen (ON) primary emissions, the present-day global nitrogen atmospheric source is about 60% anthropogenic. Table 2 shows the global estimates of deposition fluxes for the 10 different simulations of TM4-ECPL and compares them with literature estimates. Almost a 3-fold increase over land (2-fold over the ocean) has been calculated for soluble N deposition due to the increase from 1850 to the present, primarily in the inorganic N flux that is driven by human activities. The annual global present-day atmospheric N deposition varies between 126 and 132 Tg-N y^{-1} with 54-58 Tg-N y^{-1} estimated to be deposited over the ocean (Figure 4). About 10% of the emitted

Table 2. Atmospheric total N_r deposition fluxes calculated by the TM4-ECPL model in Tg-N y^{-1}

Form of Nitrogen	***ACCMIP 1850***	***RCP6.0 2005***	***RCP6.0 2050***	***RCP8.5 2050***	***ACCMIP 2000***	***PhotoComp 2000***	***VET2014 2000****
NH_x	17 (10)	53 (19-20)	64-68 (21-22)	70 (21)	49±1	63	
NO_y	6-12 (3-6)	40-46 (20-22)	30-37 (15-18)	39 (20)	51±4	51	
DON	13-21 (8-10)	22-29 (12-14)	22-29 (12-14)	21 (12)	-	-	
ON insoluble	5-7 (1-2)	5-7 (1-2)	5-7 (1-2)	5 (2)			
Total N	48-51 (23-27)	126-132 (54-58)	129-138 (50-55)	135 (55)	101±2	114	122.2 (39)

Ranges provide results calculated based on different anthropogenic emission inventories and model configurations (see details in Kanakidou et al., 2016). The respective deposition fluxes over the oceans are provided in parentheses. For comparison purposes, the estimates from the ACCMIP multi-model simulations (Lamarque et al., 2013), from the PhotoComp simulations (Dentener et al., 2006) and from the data of Vet et al. (2014) (VET2014) for the year 2000 are also provided. For VET2014 parenthesis provides deposition flux on coastal and ocean systems. NO_y is calculated by TM4-ECPL as the sum of NO_x, HNO_3 and particulate NO_3^-; NH_x is the sum of NH_3 and NH_4^+ (This table is derived from Table 1 in Kanakidou et al., 2016).

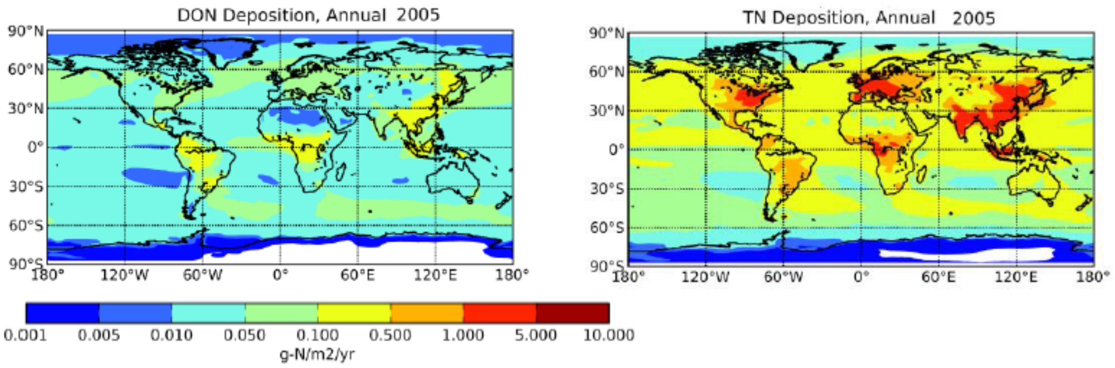

Figure 4. Atmospheric deposition of N in g-N.$m^{-2}y^{-1}$ computed by TM4-ECPL for DON (left) and TN (right), for the year 2005 (Extracted from Figure 3 from Kanakidou et al., *Journal of the Atmospheric Sciences,* 73, 2039-2047, DOI: 10.1175/JAS-D-15-0278.1. (2016)).

Source: © American Meteorological Society

NOx is deposited as ON instead of inorganic nitrogen (IN), as is considered in most global models. Present-day model results have been evaluated by Kanakidou et al. (2016) against available observations at monitoring stations over Europe and the US but also against the ACCMIP models ensemble product. In addition, the calculated IN aerosol surface concentrations and deposition fluxes over the oceans have been evaluated by Baker et al. (2017) by comparison with observations made from ships around the globe. About 20-25% of the total deposited N is calculated to be in the form of ON, in agreement with observations. Figure 5 shows the fraction of deposition of dissolved total N (DTN) that is deposited as dissolved ON (DON) as computed by the model base case simulation for the past, present and future atmospheres. This figure shows that the changes in atmospheric N emissions also lead to a decrease in the ratio of dissolved organic nitrogen (DON) to dissolved total nitrogen (DTN) from 0.77 in 1850 to 0.30 at the present time (in agreement with Duce et al.'s (2008) estimate) on an annual global-mean basis. Regionally-significant deviations from these mean ratios are computed; in particular the importance of marine DON recycling in the Southern Ocean and the north Atlantic is obvious.

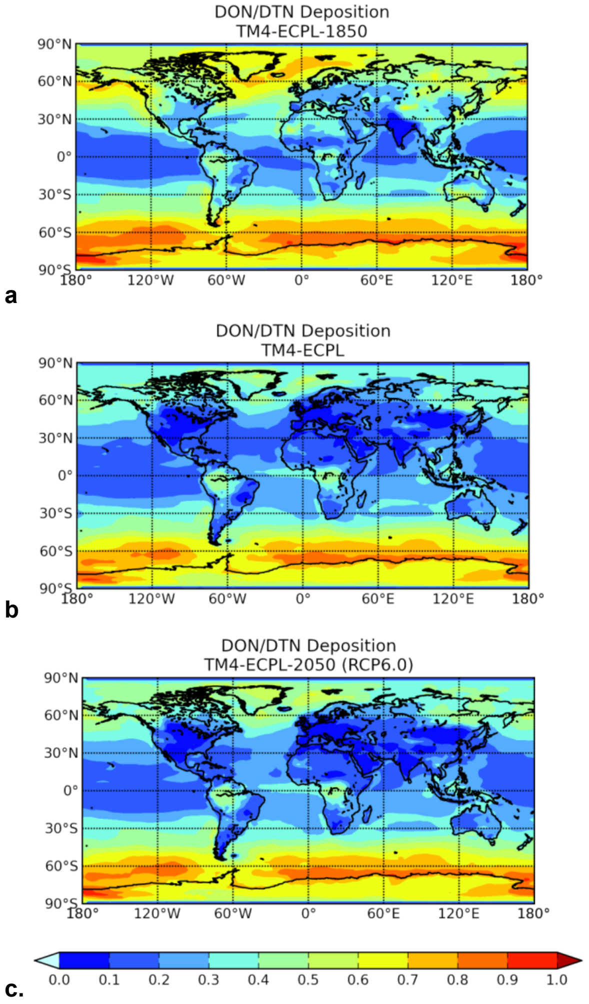

Figure 5. Ratios of DON to DTN annual deposition fluxes as computed by the base case simulation for a) 1850 simulation (global mean ratio 0.77), b) present day atmosphere (global mean ratio 0.30), c) 2050 simulation using RCP6.0 (global mean ratio 0.29) (Modified Figure S6 from Kanakidou et al., *Journal of the Atmospheric Sciences,* 73, 2039-2047, DOI: 10.1175/JAS-D-15-0278.1. (2016)).

Source: © American Meteorological Society

For the future, using RCP6.0 and RCP8.5 scenarios, the reduction in NO_x emissions is projected to be compensated by the continuing increase in NH_3 emissions, and as a result the global TN deposition is not expected to change much while its acidity will be significantly reduced. Regionally significant changes are projected, in particular over Asia. These results neglect future changes in biogenic emissions driven by climate. However, changes in natural emissions are expected to have much smaller impact on N deposition than changes in anthropogenic N emissions. The impact of the estimated changes in TN fluxes on the ecosystems requires dedicated biogeochemical ecosystem studies, and the results of the present study have been used by Jickells et al. (2017) to revisit the Duce et al. (2008) paper on the importance of human-driven atmospheric N deposition for the carbon cycle.

Uncertainties and recommendations for future work

On the global scale, significant changes since the preindustrial era are estimated and very small changes are projected in the future. Important changes have been computed in the regional distribution and chemical composition of the deposited N fluxes, with reduced compounds gaining importance relative to oxidized ones in the future (Figure 6).

Sensitivity simulations show associated large uncertainties in the modelled total soluble N deposition fluxes with the investigated model parameterizations of partitioning of inorganic nitrogen onto aerosols (at least within 10%) and of N chemical fixation on organics (between 25-35%). Much larger uncertainties were found for N emissions and in particular the ON primary emissions.

To improve the simulations of the atmospheric N cycle there is therefore a need for additional experimental data on the formation of secondary ON and on the primary sources of ON, its chemical characterization and its chemical ageing in the atmosphere during transport.

Changes in atmospheric acidity (aerosol, gases and precipitation) are a critical factor of uncertainty - in particular the changes in the aerosol acidity to which inorganic nitrogen partitioning to the aerosol phase is very sensitive (Weber et al., 2016). This finding points to the need for monitoring size-resolved aerosol composition as a whole and not only its N_r compound content.

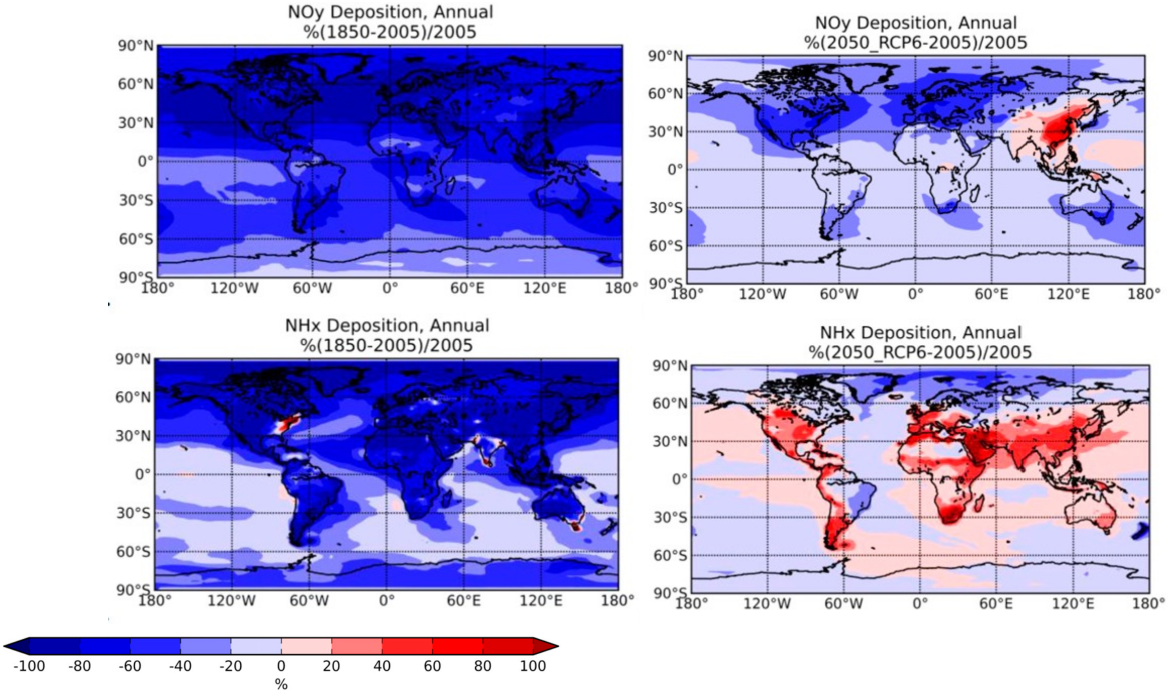

Figure 6. Percentage change relative to 2005 annual deposition flux computed by TM4-ECPL for (top) NOy; (bottom) NHx; due to (left) preindustrial emissions and (right) 2050 anthropogenic RCP6.0 emissions (Figure modified from Figure 4 in Kanakidou et al., *Journal of the Atmospheric Sciences,* 73, 2039-2047, DOI: 10.1175/JAS-D-15-0278.1. (2016)).

Source: © American Meteorological Society

Therefore, the mixing of natural aerosols containing crustal or sea-salt cations, as well as from biomass burning with anthropogenic acids, will be important for N partitioning to the aerosol phase and thus for atmospheric N deposition. These processes in the atmosphere need to be observed and modelled.

These many sources of uncertainty must be investigated through laboratory and field studies to enable a more robust view of the importance of atmospheric N deposition to the marine environment. Long term monitoring of size resolved atmospheric concentrations and deposition fluxes are needed for model evaluation.

2.3 Summary of "Limited Impact of Atmospheric Nitrogen Deposition on Marine Productivity due to Biogeochemical Feedbacks in a Global Ocean Model"

(Christopher J. Somes, Angela Landolfi, Wolfgang Koeve, and Andreas Oschlies)

This paper was published in *Geophysical Research Letters*, 43, 4500-4509, doi:10.1002/2016GL068335. (2016).

Introduction

Nitrogen is an essential element for life that limits marine productivity throughout much of the surface ocean. In the preindustrial ocean, bioavailable fixed nitrogen (fixed-N) was predominantly supplied by N_2-fixing microorganisms (diazotrophs) and was removed by bacterial processes of denitrification (including the bacterial anammox process) in oxygen deficient zones (ODZs, O_2<~10μM), where anaerobic respiration of organic matter occurs in the water column and in sediments (Gruber, 2008). Anthropogenic N emissions and subsequent deposition into the ocean are rapidly increasing (Duce et al., 2008) and approaching estimates of N_2-fixation by diazotrophs (Gruber, 2008), which may alter the balance of marine fixed-N inventory and productivity in the future ocean. It is not yet understood how marine ecosystems will respond to increasing levels of atmospheric N deposition, but it has been suggested that atmospheric N deposition may relieve N-limitation in the ocean and stimulate additional production (I.N. Kim et al., 2014) that could help buffer rising atmospheric CO_2 by sequestering carbon in the ocean.

The dynamic global response of marine N cycling processes to atmospheric N deposition has yet to be quantitatively understood. In this study, we use a global 3D ocean-biogeochemical model (Somes and Oschlies, 2015) that prognostically simulates N_2-fixation, water column denitrification, and sedimentary denitrification to quantify their dynamic response to idealized atmospheric N deposition model experiments and estimate their impact on marine productivity and ODZs.

Results

Local nitrogen addition simulations

Upon simulated N deposition (N-addition) throughout the tropical/subtropical open ocean where N_2-fixation generally occurs, N_2-fixation decreases nearly instantaneously, which buffers the input from N-addition (Figure 7a). N-addition to the surface ocean reduces N-limitation and thus diazotrophs' ecological niche in the model. Ordinary phytoplankton then outcompete diazotrophs for available phosphate (PO_4^{3-}) in the surface waters due to their higher growth rate (Hood, 2004). Some of the N-addition outside regions of N_2-fixation, if not immediately utilized by ordinary phytoplankton, eventually circulates into N_2-fixation regions and drives an additional decrease of N_2-fixation on decadal timescales (Figure 7b,c).

In regions where N_2-fixation is equal to or higher than local N-addition, the reduction in N_2-fixation compensates the N-addition and changes to the fixed-N inventory and productivity are small (Figure 7g-l). Since the current global anthropogenic N deposition estimate to the oceans (~70 Tg N y^{-1}) is much lower than modern global estimates by the model for N_2-fixation (133 Tg N y^{-1}), a reduction in N_2-fixation could, in principle, effectively compensate for the effect global N deposition on plankton productivity. This expectation assumes that the nitrogen deposition falls in regions with N_2-fixation, and that the diazotrophs and the remainder of the phytoplankton community respond to ambient fixed-N increases as hypothesized above (Landolfi et al., 2015).

Near ODZs where water column denitrification occurs, the local N-addition experiments show the counterintuitive result of a net loss to the marine fixed-N inventory (Figure 7d-i). This is due to denitrification consuming ~7 moles of NO_3 per mole of remineralizing organic nitrogen (*Richards*, 1965). Therefore, if phytoplankton assimilates all the N-addition into organic matter, which subsequently remineralizes via denitrification in the ODZ, N-addition at the surface would generate a hypothesized net N-loss cycle (Landolfi et al., 2013). The spatial extent of the net N-loss cycle depends on the efficiency by which N-addition is assimilated by phytoplankton and remineralized in and around ODZs, which expands with time as N-addition continually fuels more organic matter production (Figure 7g-i).

In our model simulations, the net N-loss cycle is most pronounced near the eastern tropical North Pacific ODZ (Figure 7i) due to strong N limitation in that region, where efficient assimilation of the N-addition directly above the ODZ occurs. Higher N_2-fixation exists above the North Indian ODZ due to more iron-replete conditions via atmospheric dust deposition compared to the eastern tropical Pacific. In the Northern Indian Ocean, N_2-fixation decreases in response to N-addition and buffers the potential to fuel a strong N-loss cycle (Figure 7 a-c). In the more iron-limited and NO_3-replete waters above the eastern tropical South Pacific ODZ, ordinary phytoplankton do not immediately assimilate a large portion of the N-addition to cause a strong N-loss cycle directly over that ODZ.

N-addition at higher latitudes accumulates in the surface open ocean in the model, which can potentially stimulate additional productivity. However, much of the high latitude ocean is either light or iron-limited so N-addition still has a limited immediate impact on local marine productivity (e.g. Southern Ocean, Figure 7 j-l). This additional fixed-N eventually circulates into the tropics and induces the same stabilizing N-cycle feedbacks described above. Since N_2-fixation occurs in closer spatial proximity to the Southern Ocean than denitrification, it responds before denitrification (Figure 7). The strength and timing of these stabilizing N-cycle feedbacks depends on how efficiently the N-addition circulates to these locations of N_2-fixation and denitrification, governed by ocean circulation.

On continental shelves, enhanced productivity from N-addition increases sedimentary denitrification in the model, which is predicted using an empirical benthic transfer function based on fluxes of organic carbon to the sediments, as well as bottom water oxygen and nitrate (Bohlen et al., 2012). The simulated sedimentary denitrification increase buffers much of the N-addition occurring over some continental shelves (e.g. North Sea, Figure 7 d-f). Since only part of the organic matter arrives at the sediment and, depending on bottom water nitrate and oxygen conditions, only part of the sedimentary respiration occurs via denitrification, the sedimentary denitrification feedback is less intense than in water column ODZs and never drives a net benthic N-loss cycle according to our model. Nonetheless, enhanced sedimentary denitrification still compensates much of the N-addition occurring over the continental shelves.

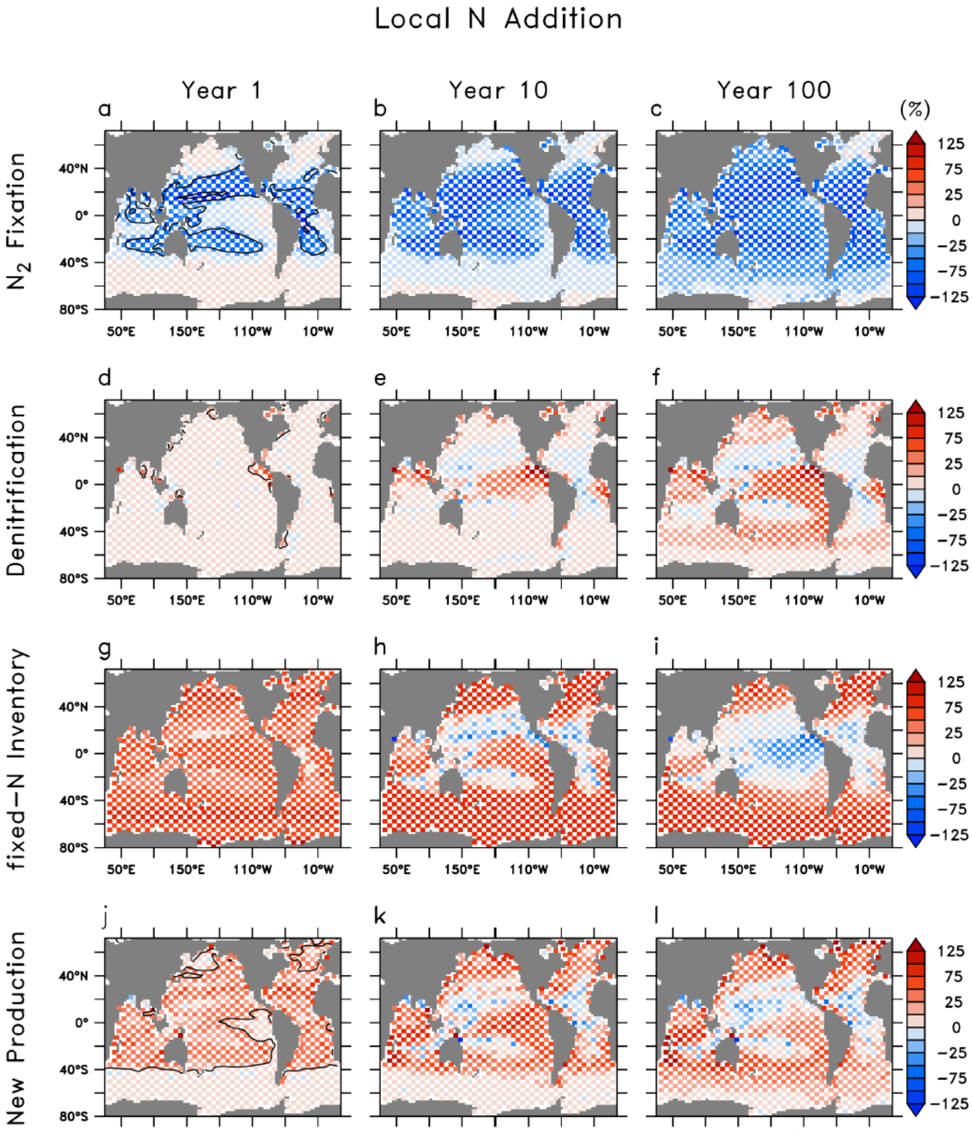

Figure 7. Local N-addition Simulations. The annual per cent changes of (a-c) global N_2-fixation, (d-f) global denitrification, (g-i) global marine fixed-N inventory, and (j-l) global new production normalized to the N-addition after 1, 10, and 100 years, respectively. Model experiments were performed on every other set of 2x2 grid boxes (coloured boxes) and white boxes indicate no experiment at that location. For N_2-fixation, denitrification, and new production, a value of ±100% means this process changed by the exact amount of the N-addition rate into the surface ocean grid boxes. For the global fixed-N inventory, the value of 100% means that all the N-addition has accumulated in the ocean, a value of 0% means that there is no net change, and negative values indicate a net loss of global fixed-N despite continuous N-addition. Contours of vertically-integrated (a) N_2-fixation (30 and 200 mmol N m^{-2} y^{-1}) and (d) total denitrification (200 mmol N m^{-2} y^{-1}), as well as (j) surface NO_3 (3 mmol m^{-3}) are shown from the N Feedbacks before N-addition was applied.

Source: Somes et al., 2016

Global Atmospheric Nitrogen Deposition Simulations

To better evaluate how these different N-cycle feedbacks interact on the global scale, we conducted additional idealized global atmospheric N deposition simulations from empirically estimated patterns and rates (Duce et al., 2008). These global atmospheric N deposition estimates were applied on two preindustrial model configurations: one including our standard configuration with a prognostic marine N budget including N_2 fixation, water column denitrification, and sedimentary denitrification (N Feedbacks, Figure 7) and another where these processes are removed, and thus N input via atmospheric deposition will cycle conservatively within the ocean circulation-biogeochemical system (N Conservative). These two model configurations were chosen to generally reflect the low (N Conservative) and high (N_Feedbacks) levels of complexity that global climate-biogeochemical models choose to represent marine N cycling (e.g. see table A3 in Cabré et al. (2015)). Both model versions produce similar large-scale patterns of common biogeochemical variables typically used to validate models such as PO_4, global net primary production, and ODZ volume.

The N_Conservative simulation was achieved by setting diazotrophs' growth rate to 0, which eliminates N_2-fixation, and removing the water column and sedimentary denitrification fluxes, while all other parameters remain identical. Thus, it behaves strictly according to the Redfield ratio (N:P=16) and cannot reproduce observed variations of N*. The switch to anaerobic respiration via denitrification is also removed, and thus oxygen can be consumed into negative concentrations in N_Conservative, which occurs in the core of the ODZs. N_Conservative was initialized with the quasi steady-state solution from N_Feedbacks and was run for an additional 2,500 years as the marine ecosystem-biogeochemical component approached its new steady-state with the same global marine fixed-N inventory, iron limitation, and ocean circulation as N_Feedbacks.

In our idealized experiments, we forced both model configurations with an empirical estimate of global N deposition directly from year 2000 (Figure 8a) (Duce et al., 2008) for 1,000 years, which was applied continuously to the preindustrial quasi steady-state solution. Atmospheric CO_2 is prescribed as a constant at preindustrial level and thus the seasonally cycling ocean circulation remains unchanged in the atmospheric N deposition perturbation simulations. However, the marine ecosystem-biogeochemical component dynamically reacts to the biogeochemical forcing induced by the extra N input, albeit with the same seasonally cycling iron limitation. While these simulations have implications for atmospheric nitrogen deposition's contribution to future changes in marine productivity, they should not be interpreted as direct predictions given the idealized nature of these simulations, which do not account for changes to temperature, ocean circulation and iron cycling.

The N_Conservative simulation shows a stronger response to the atmospheric N deposition compared to N_Feedbacks (Figure 8). Fixed-N limits productivity throughout much of the tropical/subtropical surface ocean in our model. Therefore, the N deposition input relieves N-limitation and stimulates additional productivity (+14.6%) and expansion of the ODZs (+94.6%) after 100 years in N_Conservative (Figure 8 d,e), while the changes predicted by N_Feedbacks to marine productivity (+1.8%) and ODZs (+8.6%) are smaller by nearly an order of magnitude. This illustrates that dynamic N-cycle feedbacks associated with N_2-fixation and denitrification compensate a substantial part of the atmospheric N deposition in the model.

The strong response in N_Conservative is likely affected by the initial NO_3:PO_4 conditions, which is set to 14.3 according to World Ocean Atlas observations. We chose these initial conditions because it is the most common protocol applied to global ocean-biogeochemical models. Since this ratio is lower than the canonical Redfield ratio applied to ordinary phytoplankton (N:P=16), the model is strongly N-limited and sensitive to N-input perturbations from atmospheric deposition. The N_Conservative simulation would likely be less sensitive to atmospheric N deposition if the global NO_3:PO_4 was initialized at the Redfield ratio, where the system may shift to P-limitation after N-input via atmospheric deposition.

In the N_Feedbacks simulation, N_2-fixation and denitrification both act to stabilize the fixed-N inventory and marine productivity in response to global atmospheric N deposition, but react on different temporal and spatial scales (Figure 8b). Since N_2-fixation occurs in surface waters that may be directly impacted by deposition, it significantly decreases almost instantaneously to N deposition with 48% of the total decrease during the entire millennial simulation occurring within the first year and only another 1.2% decrease between years 100 to 1,000. Denitrification shows a more gradual response as it increases by 17% in the first year as well as another 17% between years 100 and 1,000.

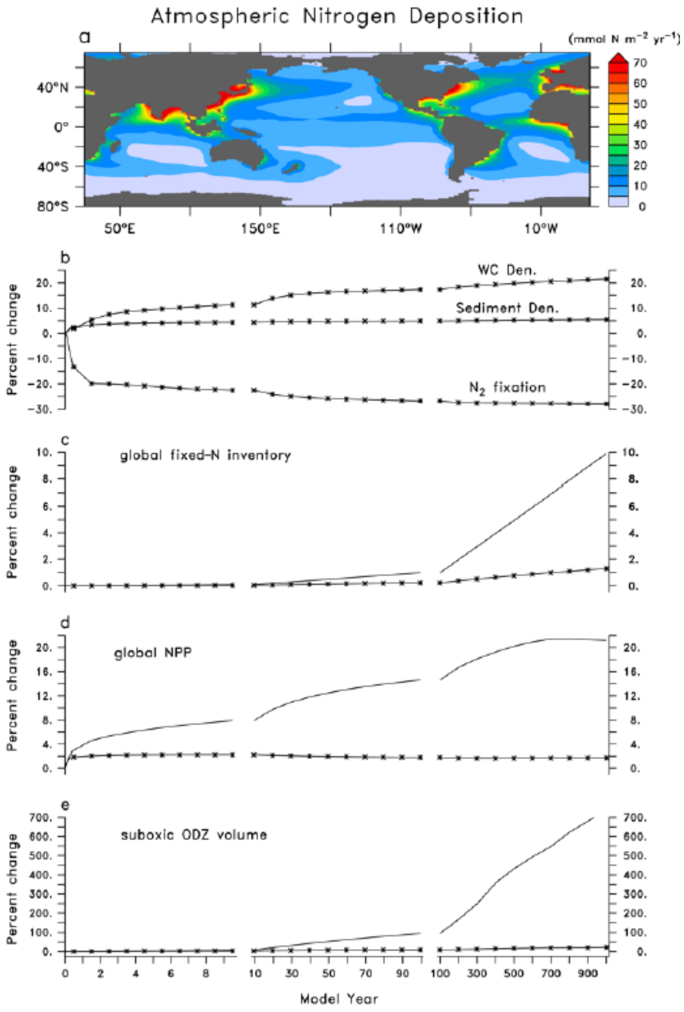

Figure 8. Global Atmospheric Nitrogen Deposition Simulation. Comparison of results from N Feedbacks (i.e. includes N_2 fixation and denitrification; lines with asterisks) and N_Conservative (i.e. excludes N_2 fixation and denitrification; straight lines), in which the (a) year 2000 deposition estimate (Duce et al., 2008) was applied continuously starting at model year 0, of (b) N_2 fixation and denitrification, (c) the global fixed-N inventory, (d) net primary production (NPP), and (e) volume of the suboxic ODZ (O_2<10 mmol m^{-3}). Per cent change is calculated relative to the preindustrial simulations of N_Feedbacks and N_Conservative, respectively, which exclude atmospheric nitrogen deposition.

Source: Somes et al., 2016

Atmospheric N deposition occurring in N-limited surface waters stimulates additional organic matter production (Figure 8d), most of which is respired aerobically and thereby leads to declining dissolved oxygen levels (Figure 8e). However, part of it is respired anaerobically in the slowly expanding ODZs and thereby increasing denitrification. This denitrification feedback acts as the main buffer to compensate the continuous global N deposition contributing to an increased global fixed-N inventory, marine productivity, and ODZ volume on long timescales in N Feedbacks compared to N Conservative (Figure 8).

Conclusions

Our modelling results suggest that oceanic feedbacks associated with N_2-fixation and denitrification limit the impact of atmospheric N deposition on marine productivity. We have identified different spatial and temporal scales of N cycle feedbacks on the marine fixed-N inventory in response to atmospheric nitrogen deposition according to our simulations. N_2-fixation provides an immediate stabilizing feedback by declining nearly instantaneously to N deposition. Denitrification, in addition, provides a more slowly operating stabilizing feedback with continuously increasing N-loss rates caused by additional remineralization of organic matter stimulated by N deposition near the slowly expanding ODZs. The water column denitrification feedback causes a net loss of marine fixed-N in ODZs in response to atmospheric nitrogen deposition in our model, which occurs due to its stoichiometry of consuming ~7 moles inorganic nitrogen for each mole of remineralizing organic nitrogen produced from deposition. This counterintuitive net fixed-N loss effect (Landolfi et al., 2013) makes water column denitrification a globally important nitrogen limiting feedback, despite the small size of ODZs in the global ocean. Our study emphasizes the importance of an adequate representation of the patterns and rates of N_2-fixation, water column denitrification and sedimentary denitrification to correctly predict the response of marine productivity to atmospheric N deposition and the future evolution of the global marine N cycle.

2.4 Summary of “What Proportion of Riverine Nutrients Reaches the Open Ocean?”

(Jonathan Sharples, Jack J. Middelburg, Katja Fennel and Timothy D. Jickells)

This paper was published in *Global Biogeochemical Cycles*, 31, 39-58, doi:10.1002/2016GB005483. (2017).

Introduction

Rivers are the primary conduit transporting weathered, leached and human-derived material from land to the sea. Rivers deliver 300-380 Tg of organic carbon per year to the coastal zone (Seitzinger et al., 2010), along with important quantities of nitrogen (37-66 Tg total N y^{-1}), phosphorus (4-11 Tg total P y^{-1}) (Seitzinger et al., 2005; Mayorga et al., 2010; Beusen et al., 2016), and silicate (340-380 Tg dissolved Si y^{-1}) (Tréguer et al, 1995; Beusen et al., 2009; Frings et al., 2016). Shelf sea ecosystems are affected by these fluvial inputs, and in turn modify the biogeochemistry of the open ocean by retaining, exchanging or breaking down nutrients and organic carbon. Our understanding and ability to predict global biogeochemical cycles currently lacks a realistic treatment of the role of shelf seas in modifying land-derived nutrient and carbon inputs to the open ocean.

In this paper we use the residence time of riverine plume water in shelf systems as a useful determinant of the fractions of dissolved N and P that are processed on the shelf before they can reach the open ocean (Nixon et al., 1996; Seitzinger et al., 2006). We develop a mechanistic description of plume behaviour that uses simple physical parameters combined with observational estimates of their global variability. We apply the method to the Nutrient Export from WaterSheds (NEWS) 2 database of riverine discharges and nutrient loads (Mayorga et al., 2010). The results provide a method for inclusion of riverine nutrient supplies in global models that is more realistic than simply assuming that either all or none of the riverine dissolved N and P load reaches the open ocean.

Methods

A low salinity plume of water exiting an estuary and entering the shelf sea will be diverted by the Earth's rotation against the coast to form a buoyancy-driven current. Our method is based on using the cross-shelf width of this buoyancy current compared to the width of the local continental shelf as a determinant of what processes transport riverine material across the shelf break and into the open ocean.

We assume that the width of a coastal buoyancy current, W_p, is related to the internal Rossby radius, R'_o, by $W_p = aR'_o$. At each river mouth in the NEWS 2 database we consider the question: is the width of the coastal buoyancy current less than or greater than the width of the shelf? If the buoyancy current extends beyond the shelf break, then we assume that the residence time of plume water over the shelf is governed by the cross-plume residual flows, wind-driven Ekman transport and horizontal dispersion within the buoyancy current. If the buoyancy current is narrower than the shelf, then we assume that exchange of plume water with the shelf water is far more effective than the exchange of shelf water across the shelf break and exchange between the shelf and the open ocean is constrained by the generally weak shelf break processes (e.g. (Huthnance, 1995)).

We use observations of plumes to yield useful global mean values for the relationship between W_p and R'_o, along with uncertainty estimates. For each river mouth the nearest shelf edge was identified, and the distance, W_s, between river mouth and shelf edge was calculated. The estimates of plume width were compared to the local shelf width to produce a global map of the ratio of plume width to shelf width:

$$S_p = \frac{W_p}{W_s} \qquad (1)$$

If $S_p > 1$ the low salinity plume reaches beyond the shelf edge into the open ocean, and the residence time of water over the shelf is assumed to be governed by the cross-shelf transport mechanisms within the buoyant plume. If $S_p < 1$ then the low salinity water will be confined on the shelf within the coastal buoyancy current; plume water will be gradually stirred into the shelf water and eventual exchange with the open ocean will be controlled by the processes that govern ocean-shelf exchange across the shelf break. A mean riverine water residence time on the shelf, T_{res}, is then calculated based on S_p and transport and exchange rates appropriate to whether S_p is greater than or less than 1.

The results of our calculation for the residence times compare well with available observations of residence times on a wide range of shelves and over a range of T_{res} from 1 week to > 1 year. Empirical relationships linking dissolved inorganic nitrogen (DIN) and dissolved inorganic phosphorous (DIP) processing on the shelf to T_{res} (Seitzinger et al., 2006; Nixon et al., 1996) are used to determine the proportion of DIN and DIP that escapes biogeochemical processing on the shelf to be exported to the open ocean for each river in the NEWS 2 database:

$$N_{export} = 100 - 23.9T_{res}^{0.2} \quad \% \qquad (2)$$

$$P_{export} = 80.5 - 13.8\ln T_{res} \quad \% \qquad (3)$$

Results

The global mean proportions of DIN and DIP that reach the ocean are 76% (minimum 72%, maximum 80%) and 79% (minimum 69%, maximum 85%) respectively. The largest contribution to the ranges in the analysis result from the range in the shelf-ocean exchange when $S_p < 1$. The largest source of random error in the results lies with the relationships used to empirically link residence time to DIN and DIP processing (equations (2) and (3)). Ignoring processing of nutrients within estuaries, so that the nutrient loads in the NEWS 2 database represent upper bounds on the amounts of DIN and DIP exiting estuaries onto the shelf, we can estimate DIN and DIP supplies to the open ocean (Figure 9, with supplies calculated on a 5°×5° global grid).

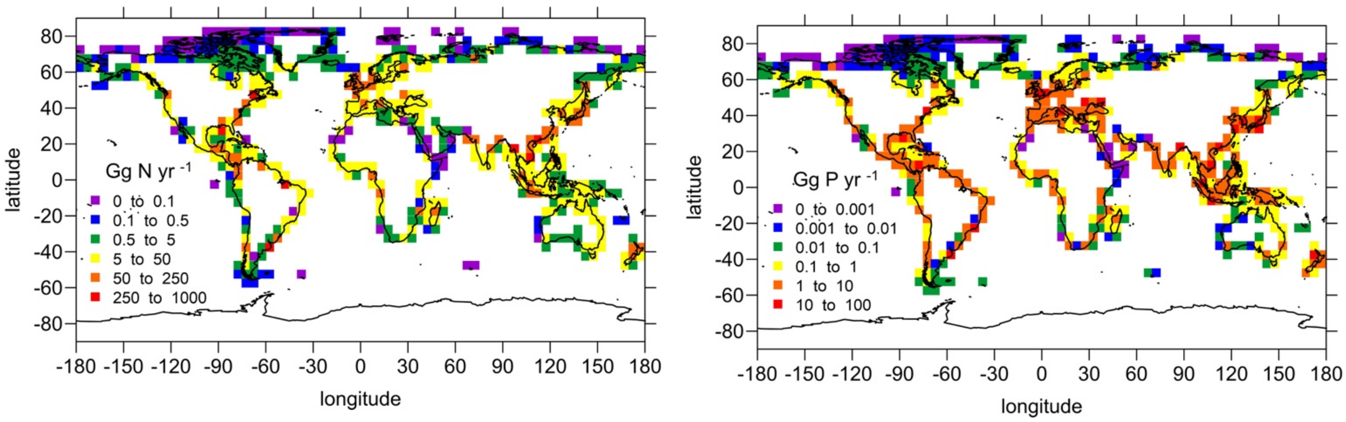

Figure 9. Global export of DIN (left) and DIP (right) from rivers to the open ocean

Source: Sharples et al., 2017

Discussion and implications

Despite the difficulty in addressing riverine nutrient delivery from over 6,000 rivers globally to the open ocean, we have made robust estimates of the proportion of DIN and DIP that could reach the open ocean. Comparing the globally integrated riverine DIN and DIP deliveries with our calculations of the global amount of DIN and DIP reaching the ocean results in an average removal of 25% DIN and 19% DIP on the shelf, which will be augmented by estuarine retention. This suggests that significant quantities of riverine dissolved nutrients reach the open ocean. Of the 23 Tg DIN that are delivered to the land-ocean interface every year, 6 Tg are retained on the shelf and the other 17 Tg are delivered to the open ocean. Similarly, of the 1.6 Tg DIP delivered by rivers, 0.4 Tg are estimated to be retained on the shelf and 1.2 Tg reaches the open ocean.

The uncertainties in the proportions of nutrients exported are typically ± 5% for DIN and ± 5-10% for DIP. The main sources of the uncertainties are the wide range of cross-plume and cross-shelf break exchange rates used in the analyses. The other important source of uncertainty not included in these error estimates, arises from the equations (2) and (3), typically ±15% in the proportions of DIN and DIP processed on the shelf.

Compared to recent estimates of atmospheric deposition of N to the oceans (Jickells et al., 2017) the annual riverine supply of 17 Tg DIN is similar to the total annual deposition of oxidized N, and is about 50% of the total atmospheric deposition of N. The annual riverine supply is about 10% of the supply via N fixation (Jickells et al., 2017). Our results also indicate important basin-scale contrasts in riverine DIN and DIP supply, with the western side of the northern hemisphere ocean basins tending to supply more DIN and DIP. Riverine N and P supplies tend to occur in different parts of the ocean compared to atmospheric N deposition or organic P delivery. Thus, the amounts of DIN and DIP delivered by rivers to the open ocean, combined with the spatial variability in the ability of shelf seas to process riverine nutrients, have the potential to alter global ocean productivity with a magnitude similar to other supply routes that are better quantified in global models. Riverine nutrient supply needs to be incorporated into global models of nutrient cycling in order to yield more accurate results beyond assuming that either all or none of the riverine nutrient supply reaches the open ocean.

While our analysis has focused on DIN and DIP, the variable shelf retention time will have similar implications for the transport of other components such as trace metals with similar reactivity and sensitivity to residence times, and for the issue of coastal eutrophication and “dead zones” (Diaz and Rosenberg, 2008). Spatially resolved estimates of nutrient processing on shelves are needed for proper assessment of climate-change induced modification of the hydrological cycle and sea level. Low sea levels during the last glaciation had greatly reduced shelf width and potentially altered nutrient and carbon delivery to the open ocean. Current evidence suggests that the ocean N cycle changed considerably during the most recent post-glacial warming (Galbraith et al., 2013). In the future, global warming is projected to lead to increased river discharge at high latitudes (Peterson et al., 2002) and nutrient flows may well increase with an increasing human population (Seitzinger et al., 2010). The latitudinal variation of DIN and DIP export will need to be incorporated into models to allow realistic projections of the future behaviour of both oceanic and coastal ecosystems.

2.5 Summary of “A Re-evaluation of the Magnitude and Impacts of Anthropogenic Atmospheric Nitrogen Inputs on the Ocean”

(Timothy D. Jickells, T.D., Eric Buitenhuis,, Katye E. Altieri, Alex R. Baker, Doug Capone, Robert A. Duce, Frank Dentener, Katja Fennel, Maria Kanakidou, Julie LaRoche., Kitack Lee, Peter S. Liss, Jack J. Middelburg, Keith Moore, Greg Okin, Andreas Oschlies, Monmohan M. Sarin, Sybil Seitzinger, Jonathan Sharples, Arvind Singh, Parv Suntharalingam, Mitsuo Uematsu, and Lauren M. Zamora)

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Introduction

Human activity has massively perturbed the nitrogen (N) cycle by increasing the total fluxes through the Earth System, particularly over the last hundred years following the invention of industrial N fixation processes with resultant damage to terrestrial and marine ecosystems (Fowler et al., 2013; Gruber and Galloway, 2008; Sutton et al., 2011). Duce et al. (2008) presented one of the first syntheses of atmospheric N deposition to the oceans and their impacts on primary production and ocean carbon dioxide (CO_2) uptake. Since that time research has intensified to try and understand the scale of the human impact on the N cycle. There have also been further attempts to understand and quantify the impacts of atmospheric deposition on the oceans. Therefore, the GESAMP Working Group 38 convened a group of experts, including many of the original authors, to re-evaluate the conclusions of Duce et al. 2008. This work included not only a critical assessment of the magnitude and distribution of atmospheric inputs to the oceans, but also a synthesis of the other main inputs of N to the oceans from land and from biological N fixation. The results of this assessment are presented in Jickells et al. (2017) and summarized here. This summary takes the form of quantification of each of the main inputs (fluvial, atmospheric and nitrogen fixation) and then a synthesis of the inputs and a model assessment of their impact.

Freshwater nitrogen inputs

River inputs dominate over groundwater inputs in terms of the total N input to the oceans. We were able to use the NEWS global database to estimate fluvial N inputs to the oceans (Seitzinger et al., 2010). Modern inputs are probably double those of one hundred years ago with modern inorganic N (mostly nitrate) inputs of 23 TgN y^{-1} and dissolved organic nitrogen (DON) inputs of 11 TgN y^{-1}. There are additional inputs of particulate organic nitrogen which are probably trapped in near shore systems, although they may still reach coastal waters and the ocean after degradation of the organic matter in coastal sediments. Duce et al. (2008) had suggested that all nitrogen from rivers was trapped on the shelf by denitrification processes. The WG38 activity evaluated this assumption critically and showed that while this may be true at high latitudes, where the Coriolis force acts to increase freshwater residence times on the shelf and thereby allows denitrification to consume much of the fluvial nitrate, this assumption is not true nearer the equator (within 20° of latitude) where the weaker Coriolis force allows more rapid transit across the shelf and less time for denitrification. The results of this work are discussed in detail in Sharples et al. (2017) included here (see Section 2.4 above). This allowed WG38 to estimate the total dissolved fluvial N inputs to the oceans, including the shelf seas, and the total input to the open ocean beyond the continental shelf (Table 3). As noted by Sharples et al. (2017) there are still uncertainties particularly about the fate of particulate organic nitrogen within the coastal seas and the rate of degradation of fluvial dissolved organic nitrogen (DON), and hence how much of this reaches the open ocean. Sharples et al. also used a single global estimate of denitrification rates and the rate may vary with temperature. Since temperature is related to latitude, this may offset the effects of Coriolis force if denitrification rates are faster at low latitudes and compensate for the shorter residence times as the authors note. Thus, the WG38 have not only improved our estimates of fluvial nitrogen inputs to the open oceans and their impacts, but also contributed to identifying research priorities for the future.

Nitrogen fixation

Nitrogen fixation is a natural process carried out by prokaryotic planktonic organisms which converts N_2 gas (which is normally chemically inert) from the atmosphere into biologically available N in the surface ocean. It occurs predominantly in tropical ocean surface waters. This process has a high biochemical energy demand and requires large amounts of iron and phosphorus. Since these latter elements are supplied to the ocean in relatively important amounts from the atmosphere, and the rates of biological N fixation are known to be limited by the availability of iron and phosphorus in some ocean areas (Jickells and Moore, 2015; Moore et al., 2013), there are direct links between the impacts of atmospheric N deposition and the deposition of other key plant nutrients (Okin et al., 2011). In the WG38 study, global N fixation rates were estimated using the PlankTOM ocean biogeochemical model (Buitenhuis et al., 2013) to be 164TgN y^{-1}, and Figure 10 shows the estimated geographical distribution of this process. These estimates are consistent with some other recent estimates, and generally somewhat higher than earlier estimates of the magnitude of this process. This reflects, in part, improvements in methodologies of measurement (Grosskopf et al., 2012). The WG 38 study, and some other recent ones, have highlighted a potentially important feedback in which increasing N concentrations in surface waters (such as could occur after atmospheric N deposition) suppress N fixation, as organisms choose to use readily available fixed N rather than expending the energy to fix N gas. The threshold at which this suppression of N fixation occurs is not well known or understood, but has the potential to be an important feedback regulating the ocean N cycle (Somes et al., 2016 and included here; Yang and Gruber, 2016), and we note this feedback deserves further study.

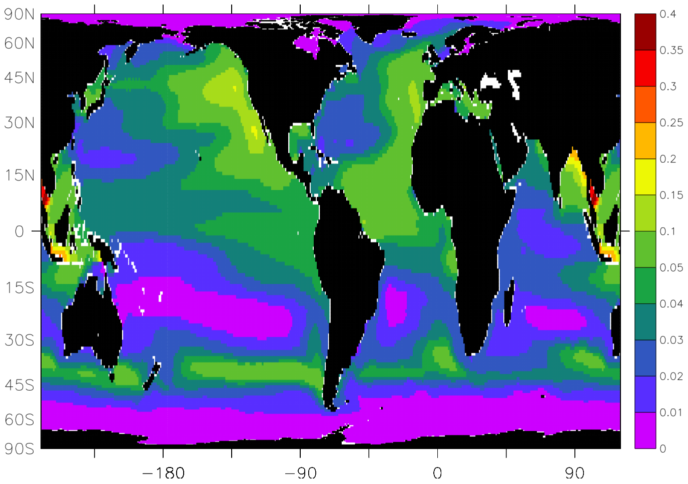

Figure 10. Nitrogen Fixation rate estimates from the PlankTOM model as mol N $m^{-2}y^{-1}$.

Source: Jickells et al., 2017

Atmospheric nitrogen deposition

Atmospheric N deposition was estimated using the TM4-ECPL atmospheric chemistry model (Daskalakis et al., 2015 included here; Kanakidou et al., 2016; Kanakidou et al., 2012), a model which explicitly includes organic nitrogen deposition. The model allowed us to estimate deposition of oxidised (nitrate aerosol and nitric acid), reduced (ammonia and ammonium aerosol) and organic nitrogen, a poorly understood source of nitrogen (Cape et al., 2011), for the present day, in the past and projecting into the future (Figure 11). WG38 has contributed important efforts towards validation of this and other atmospheric nitrogen transport models (Baker et al., 2017), also summarized here in Section 2.6. The three different sources of N contribute approximately equal amounts to the total N input to the oceans of 39TgN y^{-1} in 2005. Combustion processes are the main source of oxidised nitrogen; 55% from terrestrial anthropogenic combustion, 11% from biomass burning and 11% from shipping and the remainder from natural sources. In the case of reduced nitrogen, almost two thirds comes from agriculture. This pattern of emissions leads to the deposition field in Figure 11, which illustrates particularly large atmospheric N inputs to the oceans downwind of major source regions, particularly in the Northern Hemisphere in the western north Atlantic and Pacific. A very important result from the model results is that about a quarter of the total input of reduced and organic nitrogen to the ocean arises from recycled material from the ocean itself, and therefore it does not constitute a net new N input to the ocean. This result means that the net atmospheric deposition to the oceans estimated in Jickells et al. (2017) is significantly lower than estimated by Duce et al. (2008). While we have known for a long time that there are fluxes of ammonia and organic nitrogen from the oceans, this model estimate of the magnitude of this process is relatively large by comparison to other estimates, but also rather uncertain. The effect of this estimated N recycling on our estimate of the overall impact of N deposition on the oceans is quantitatively substantial. We conclude that an improved quantification of these fluxes would therefore allow a better quantification of the impacts of deposition.

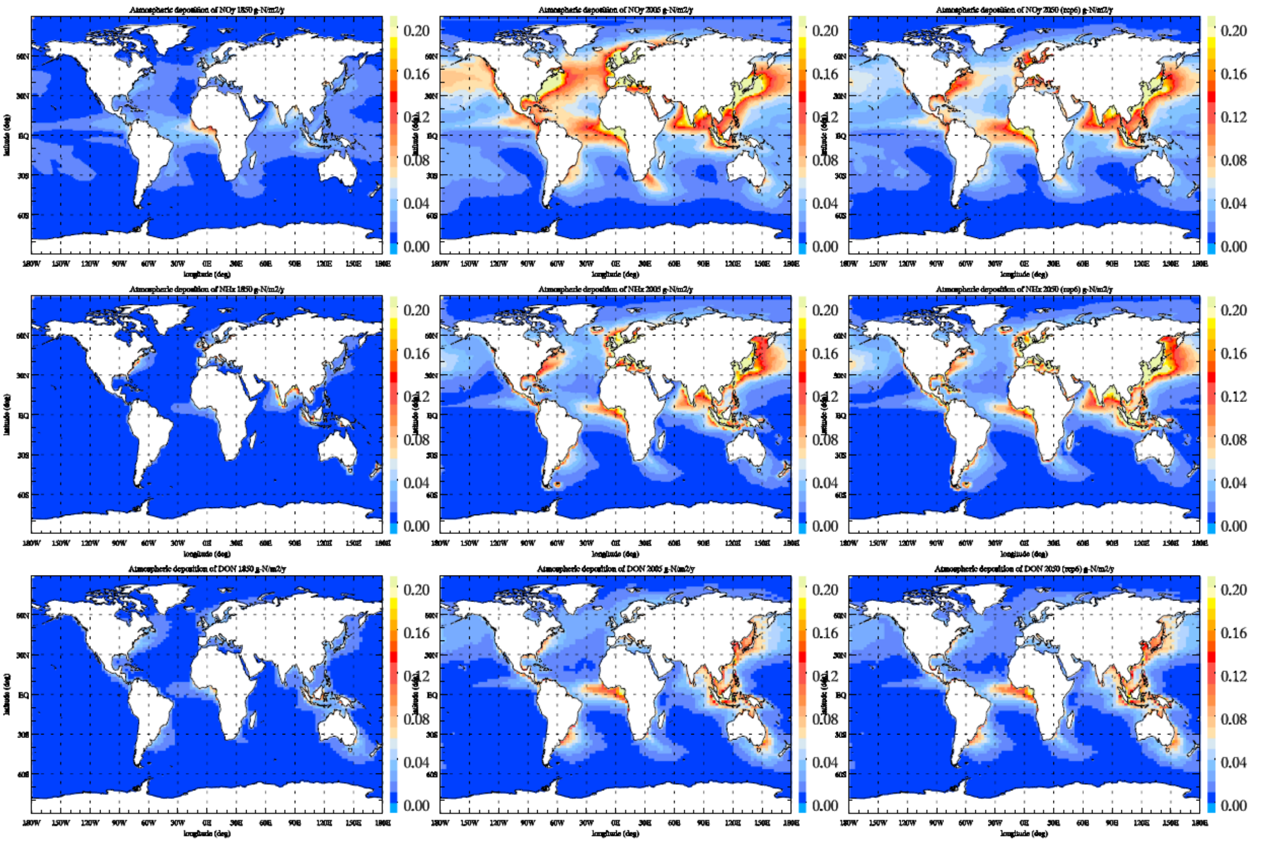

Figure 11. Atmospheric deposition of nitrogen to the oceans 1860 (left column, 2005 middle and 2050 right column for oxidised (top row), reduced (MIDDLE ROW) and organic nitrogen (bottom row).

Source: Jickells et al., 2017

Summary of nitrogen inputs to the oceans

Table 3 provides our best estimate of inputs to the oceans. This compilation confirms that the atmosphere represents the dominant route by which terrestrially-derived and mostly anthropogenic N reaches the open ocean, and this atmospheric source is of major importance relative to other inputs even on the shelf.

Table 3. Nitrogen fluxes to the ocean

Source	***Total nitrogen flux to the Ocean including the shelf seas TgN y^{-1}***	***Nitrogen flux to the open ocean beyond the shelf break TgN y^{-1}***
Atmosphere	39	>30
Fluvial	24	>17 - <28*
N_2 Fixation	164	164

**Depends on assumed fate of fluvial DON on the shelf, low value assumes it is degraded and denitrified, high vale assumes it is all resistant to degradation on the shelf*

Impact of atmospheric deposition

We have used the PlankTOM model to assess the impact of anthropogenic N deposition on the oceans. We estimate that atmospheric N deposition increases ocean carbon sequestration via the biological pump by 0.15 Pg C y^{-1} which is important within the context of an estimated ocean annual total CO_2 uptake of 2.2±0.5 Pg C y^{-1} (Denman et al., 2007). This estimated enhancement of uptake is smaller than the estimate of Duce et al, reflecting the smaller net atmospheric N deposition. These estimates are similar to others recently published (Krishnamurthy et al., 2007; Yang and Gruber, 2016).

Increases in atmospheric deposition have the potential to increase ocean productivity and the sinking of organic matter into the deep ocean, thereby increasing areas of low oxygen and causing increases in denitrification and the resulting emission of the greenhouse gas N_2O. However, we estimate that the increase in the N_2O flux will be small (1 to 3%) on the global scale, but perhaps 5-20% in the Arabian Sea region under likely scenarios over coming decades to 2050 (Suntharalingam et al., 2017 listed here (see Section 2.7)). Recent papers have highlighted a feedback in which increased denitrification can create a negative feedback on the longer timescales causing a decline in ocean nitrogen inventory (see Section 2.3), and we suggest this feedback merits further study (Somes et al., 2016).

We acknowledge that further research is necessary on the magnitude of all inputs, on improving ocean biogeochemistry models and in our understanding of the underlying processes and the role of organic nutrients in particular. We also identify that our estimate of impacts is sensitive to several of the uncertainties identified earlier, and we particularly recommend further work on the following;

- The magnitude and significance of recycling of ammonia and organic nitrogen from the oceans.

- The extent and thresholds for suppression of nitrogen fixation by ambient surface water dissolved nitrogen.

We also identified four regions which are particularly sensitive to likely changes in atmospheric deposition. These are;

- The Northwest Pacific where deposition fluxes are expected to grow and where there may already be impacts from the current inputs (Ito et al., 2016; I-N Kim et al., 2014; T.W. Kim et al., 2014), listed here – see Section 2.1 above.

- The Northern Indian Ocean which again receives a large atmospheric input and which is argued to already be increasing productivity (Singh et al., 2012). Inputs in this region are likely to grow. If these stimulate more productivity and sinking organic matter, the existing naturally occurring areas of deep water hypoxia may expand and increase N_2O fluxes (Suntharalingam et al., 2012), see also Section 2.7.

- Areas of the Mediterranean and North Atlantic where primary production is phosphorus or iron limited (Krom et al., 2010; Moore et al., 2009) and hence where additional nitrogen deposition may lead to different nutrient biogeochemistry compared to the remaining ocean areas, which are predominantly limited by nitrogen or iron supply (Moore et al., 2013; Zamora et al., 2010).

2.6 Summary of "Observation- and Model-Based Estimates of Particulate Dry Nitrogen Deposition to the Oceans"

(Alex R. Baker, Maria Kanakidou, Katye E. Altieri, Nikos Daskalakis, Gregory S. Okin, Stelios Myriokefalitakis, Frank Dentener, Mitsuo Uematsu, Manmohan M. Sarin, Robert A. Duce, James N. Galloway, William C. Keene, Arvind Singh, Lauren Zamora, Jean-Francois Lamarque, Shih-Chieh Hsu, Shital S. Rohekar and Joseph M. Prospero)

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Introduction

Anthropogenic nitrogen (N) emissions to the atmosphere have increased significantly the deposition of nitrate (NO_3^-) and ammonium (NH_4^+) to the surface waters of the open ocean. N is a limiting nutrient for primary producers over ~70% of the global ocean. Potential impacts of increased N deposition include enhancement of primary productivity in low-nitrogen marine ecosystems, ecological shifts caused by changing nutrient regimes and changes to the rates of denitrification and N_2O production and consumption. These impacts are thus significant for the global nitrogen and carbon cycles and for the budgets of radiatively-active trace gases in the atmosphere (Jickells et al., 2017). On-going implementation of emission controls (mostly affecting nitrogen oxides: NO_x) and global economic development will lead to further changes in both the magnitude and spatial distribution of nitrogen emissions, and their impacts, over the coming decades.

On relatively small spatial scales, the concentrations of NO_3^- and NH_4^+ over the oceans can be measured at coastal and island sites and from ships at sea. However, the availability of suitable land-based sites and the high cost of ship-based sampling makes monitoring of N inputs to the ocean over larger scales effectively impossible. Global-scale understanding of the impacts of N deposition to the oceans is therefore reliant on our ability to produce and validate models of nitrogen emission, atmospheric chemistry, transport and deposition (Baker et al., 2010). A number of such models have been developed, but due to the difficulties associated with making observations over the ocean, their effectiveness at reproducing N inputs to the ocean has not been evaluated previously.

In this work (Baker et al., 2017) a novel method to evaluate the performance of atmospheric N deposition models was assessed. This was achieved by compiling a database of measured aerosol NO_3^- and NH_4^+ concentrations made from ships over the global ocean and comparing these observations to model simulation products. Although scarce in comparison to observations over land, the database contained measurements from ~2900 samples collected in the period 1995-2012. Three ocean regions (the eastern tropical North Atlantic, the Northern Indian Ocean and Northwest Pacific) contained a relatively high density of observational data (Figure 12) and were the focus of the comparison to model products.

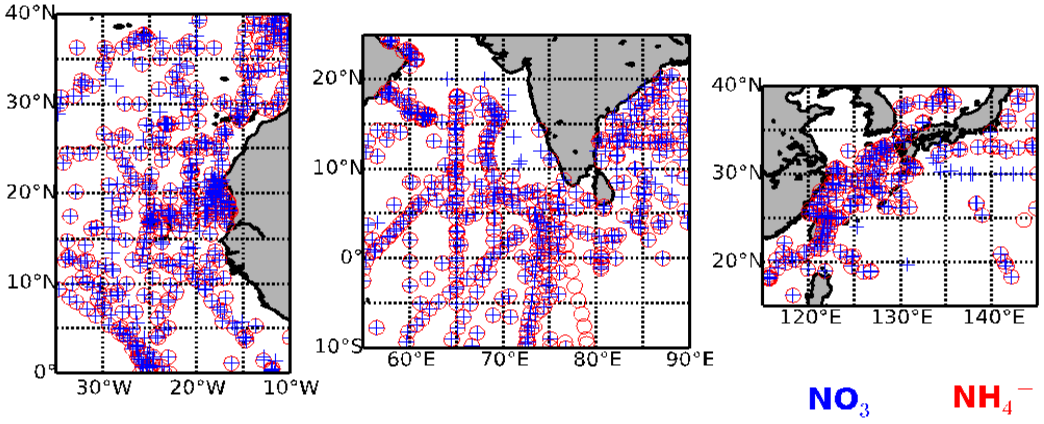

Figure 12. Aerosol sample collection start locations in the eastern tropical North Atlantic, Northern Indian Ocean and Northwest Pacific regions. Samples with NO_3^- observations are indicated with blue crosses and those with NH_4^+ observations by red circles.

Source: Baker et al., 2017

Two model products were selected for comparison to the observational database. The Atmospheric Chemistry and Climate Model Intercomparison Project (ACCMIP) evaluated the role of atmospheric chemistry in driving climate change in support of the Intergovernmental Panel on Climate Change (IPCC) Fifth Assessment Report (AR5). ACCMIP produced estimates of the deposition flux (due to both dry and wet deposition) of oxidised N (NO_y) and reduced N (NH_x) to the ocean surface, based on estimates of N emissions to the atmosphere for the year 2000 (Lamarque et al., 2013). These products were the averages of the output produced by several individual models (10 for NO_y, 5 for NH_x) and these averages are considered to be more robust than estimates produced by single models. While assessment of deposition flux is absolutely essential in order to assess the impacts of atmospheric N on marine systems, dry deposition due to aerosols cannot be measured over the remote ocean. Neither are the parameters reported by ACCMIP (NO_y and NH_x) measured over the ocean. Direct validation of the ACCMIP products was therefore not possible. The TM4-ECPL model was also included in the study (Kanakidou et al., 2016), as this was capable of reporting NO_y and NH_x dry deposition fluxes, as well as dry deposition fluxes of aerosol NO_3^- and NH_4^+ and the concentrations of these species. Thus, TM4 provided the capability to make direct comparisons of observations to model products and also to link these comparisons to the evaluation of the ACCMIP products.

Methods

The available observational data were averaged over a 5° x 5° grid and compared to the model products averaged over the same grid. Observed and modelled aerosol concentrations were compared directly. To compare to modelled dry deposition fluxes (ModDep), dry deposition fluxes (CalDep) were calculated from the product of the observed concentrations (C) and estimated dry deposition velocities (v_d). Dry deposition velocities are uncertain to within a factor of 2-3. Comparisons over regions larger than individual grid cells were made using the area- and sample number-averaged ratio ($R_{A,n}$) of modelled to observation-based parameter (concentration or deposition flux), as shown in Eq. 1, and normalized mean bias (NMB; Eq. 2) (where M is the modelled concentration or ModDep, O is the observed concentration or CalDep, A is the surface area and n is the number of observations for each grid cell).

$$R_{A,n} = \frac{\sum (M/O)\, A\, n}{\sum A\, n} \tag{1}$$

$$NMB = 100\, \frac{\sum (M-O)}{\sum O} \tag{2}$$

Summary of results

Comparison in the three study regions suggests that TM4 over-estimates NO_3^- concentrations ($R_{A,n}$ = 1.4-2.9) and under-estimates NH_4^+ concentrations ($R_{A,n}$ = 0.5-0.7), with spatial distributions in the tropical Atlantic and Northern Indian Ocean not being reproduced by the model (Figure 13). In the case of NH_4^+ in the Indian Ocean, this discrepancy was probably due to seasonal biases in the sampling.

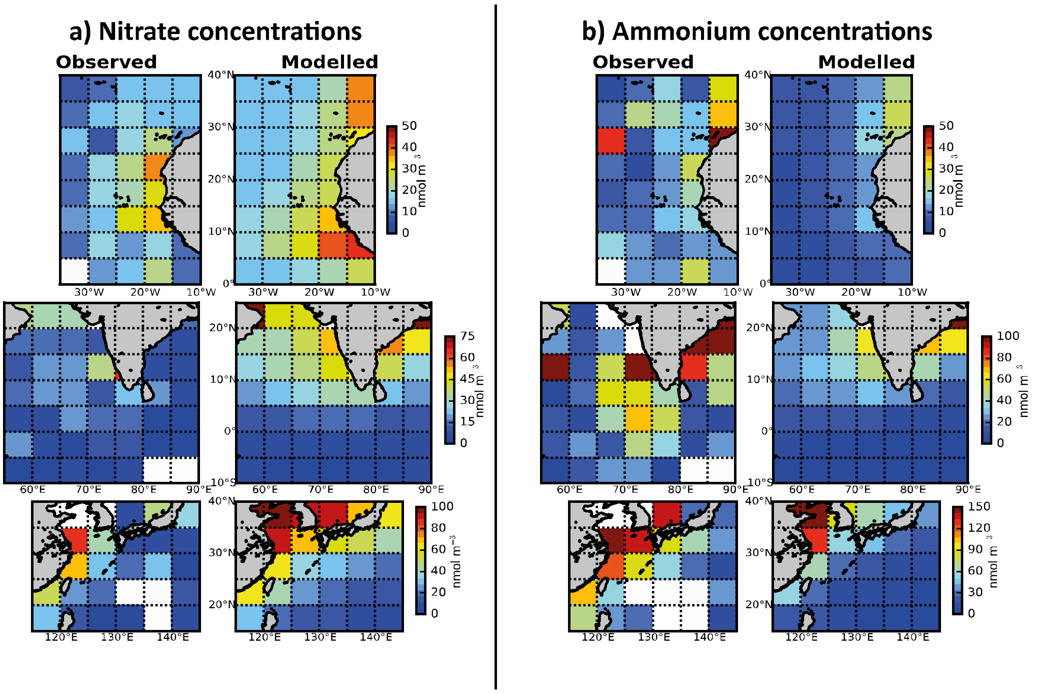

Figure 13. Aerosol concentrations of a) NO_3^- and b) NH_4^+ for the eastern tropical Atlantic, Northern Indian and Northwest Pacific study regions. For each panel, mean observed concentrations are shown on the left and concentrations simulated by TM4 on the right column.

Source: Baker et al., 2017

Similar patterns were observed in the various comparisons of CalDep to ModDep ($R_{A,n}$ = 0.6-2.6 for NO_3^-, 0.6-3.1 for NH_4^+). Values of $R_{A,n}$ for NH_x CalDep - ModDep comparisons were approximately double the corresponding values for NH_4^+ CalDep - ModDep comparisons, because a significant fraction of the NH_x model products is composed of gas-phase ammonia (NH_3) deposition which was not measured as part of the aerosol chemistry observations (Figure 14).

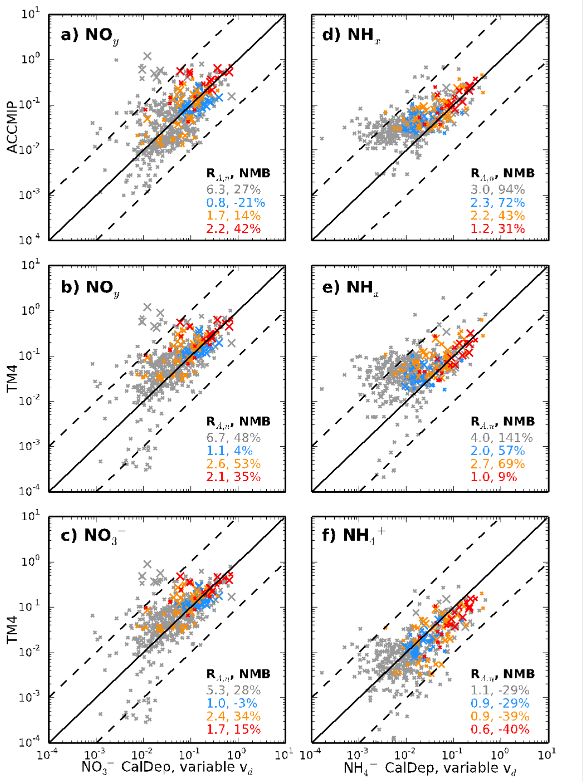

Figure 14. Scatter plots comparing dry deposition fluxes (mg N m^{-2} d^{-1}) of a) – c) NO_3^- and d) – f) NH_4^+ derived from the observational database with corresponding fluxes from model output. Panels represent comparisons to a) NO_y from ACCMIP, b) NO_y from TM4, c) NO_3^- from TM4, d) NH_x from ACCMIP, e) NH_x from TM4 and f) NH_4^+ from TM4. CalDep calculated by the variable v_d method. Data are plotted for each grid cell that contains observational data (grey), with cells from the eastern tropical North Atlantic, Northern Indian Ocean and Northwest Pacific regions coloured blue, orange and red respectively.

Source: Baker et al., 2017

Conclusions and recommendations for future work

Potential sources of bias in the model – observation comparison (arising from aerosol sampling techniques, non-uniformity in the spatial and seasonal distribution of the collected samples, modelling frameworks, and the model – observation comparison method) are discussed in the manuscript. All of the comparisons suffered due to the scarcity of observational data and the large uncertainty in dry deposition velocities used to derive deposition fluxes from concentrations. (Even the comparisons of observed and modelled aerosol concentrations are affected by uncertainties in dry deposition fluxes, because this parameter has a significant effect on modelled aerosol concentrations during transport through the atmosphere). When considering modelled dry N deposition, the uncertainty in v_d (when compounded with the other sources of uncertainty in the modelling) probably implies that fluxes can be estimated to within no better than an order of magnitude. The uncertainty in modelled dry deposition, in turn, leads to uncertainty in modelled wet deposition estimates. Uncertainties in v_d have been a major limitation on estimates of the flux of material to the oceans for several decades.

Recommendations for improved model predictions of atmospheric N inputs to the ocean include:

- Observations of N (e.g. gas-phase N speciation and deposition flux, in addition to particulate N speciation; detailed measurement of N species aerosol particle size distributions; long-term measurement of N species dry particulate deposition fluxes, concurrently with N species wet deposition measurements, at suitable remote island locations) that target key areas of uncertainty.

- Reducing uncertainties in v_d from studies of small-scale wind and aerosol property heterogeneity.

- Improvements in emissions estimates, such as through the use of satellite-derived fire radiative power to assess biomass-burning emissions.

- Updating model parameterisations of air – sea NH_3 exchange to take account of progress in this area over the last decade.

- More widespread simulation of organic N (typically 20-40% of atmospheric N) in atmospheric chemistry transport models.

- Comparison of observations to the corresponding absolute time in models, rather than over time-averaged periods as done here, in future model – observation comparisons.

Above all, validation of modelled dry deposition requires effective comparisons to observable aerosol-phase species concentrations and this cannot be achieved if model products only report dry deposition flux over the ocean.

2.7 Summary of "Increasing Inputs of Anthropogenic Nitrogen to the Northern Indian Ocean and Impacts on Marine N_2O Fluxes"

(Parvadha Suntharalingam, Lauren M. Zamora, Manmohan M. Sarin, Arvind Singh, Erik Buitenhuis, Hermann W. Bange, Srinivas Bikkina, Angela Landolfi, Laure Resplandy and Sybil Seitzinger)

This paper will be submitted for publication to *Environmental Research Letters* in 2018.

Introduction

Nitrous oxide (N_2O) production and the underlying nitrogen cycling mechanisms in the Northern Indian Ocean are fundamentally influenced by the unique combination of regional ocean biogeochemistry and circulation. The Arabian Sea is the location of one of the globe's largest permanent oxygen minimum zones (OMZs), and its upper oceanic circulation is characterized by strong seasonal upwelling driven by the regional South Asian monsoon. Previous estimates of N_2O fluxes to the atmosphere from this region highlight its significant contribution, which range from 2-35% of total ocean emissions (Law and Owens, 1990; Bange et al., 2001). N_2O emissions from the Northern Indian Ocean also have the potential to increase under pressure from a range of environmental stressors in the 21st century (Naqvi et al., 2010). Nutrient supply (N, P, Si, Fe) to marine ecosystems has increased significantly over the past century through increases in fossil fuel combustion, agricultural fertilizer application, and dust mobilization through land-use change. This has resulted in changes in marine productivity, organic matter export and remineralization, and coastal eutrophication (Galloway et al., 2003, Gruber and Galloway, 2008). These changes directly affect the oceanic N_2O source through changes in the magnitude of nitrification and denitrification fluxes, and through changes in extent of hypoxic and sub-oxic regions where N_2O production yields are higher than in the oxic ocean. Nitrogen inputs to oceanic ecosystems through deposition of anthropogenically derived nitrogen have been increasing over the past century (Duce et al., 2008), with the potential for increased N_2O production in coastal regions associated with high deposition fluxes (Suntharalingam et al., 2012). Model studies also suggest that non-linear feedbacks in the ocean biogeochemical system coupled with even relatively small nitrogen inputs from atmospheric N deposition could increase local rates of denitrification and cause net nitrogen losses from the system, potentially reducing biological productivity and influencing N_2O changes (Landolfi et al., 2017).

Here we bring together recent estimates of external nitrogen inputs to the Northern Indian Ocean from atmospheric deposition and riverine sources. We investigate the implications of this additional nutrient source for regional oceanic N_2O production using ocean biogeochemistry model analyses, identify uncertainties in these estimates, and highlight needs for further investigation.

Results

Inputs of anthropogenically derived nitrogen to the Northern Indian Ocean

Table 4 presents a compilation of recent estimates of nitrogen inputs to this region from atmospheric deposition and riverine fluxes. The sources for these estimates are described below.

Table 4. Riverine and atmospheric dissolved nitrogen inputs to the Arabian Sea and Bay of Bengal for the present-day (Year 2000). Units are in Tg N y^{-1}.

	Riverine	***Atmospheric***	***N_2 fixation***
Arabian Sea	0.38^{*} $(0.3)^{a}$	1.89^{**} $(0.92)^{b}$	$(0.03\text{-}15.4)^{c,\,d}$
Bay of Bengal	3.32^{*} $(9)^{a}$ $(0.9\text{-}2.0)^{d}$	1.20^{**} $(0.46)^{b}$ $(0.01\text{-}0.38)^{d}$	$0.4\text{-}8.2^{d}$

[*] *This study, year 2000 model data (includes dissolved organic and inorganic nitrogen, but not particulate nitrogen) (Seitzinger et al., 2010)*

[**] *This study, year 2000 model data (only includes inorganic nitrogen inputs)*

[a] *(Singh and Ramesh, 2011)* [b] *(Singh et al., 2012)*

[c] *(Gandhi et al., 2011)* [d] *(Srinivas and Sarin, 2013)*

Model derived deposition and riverine fluxes: Atmospheric inorganic nitrogen inputs are estimated from the NCAR Community Atmosphere Model (CAM) version 3.5 (Lamarque et al., 2011). Water soluble organic nitrogen atmospheric deposition was obtained from the Kanakidou et al. (2012) model. Riverine dissolved inorganic and organic nitrogen inputs were obtained from the NEWS model (Seitzinger et al., 2010) and other sources listed in Table 4.

Flux estimates from aerosol data: Data for nitrate and ammonium aerosol concentrations were obtained from the Surface Ocean Lower Atmosphere Study (SOLAS) Project Integration website (http://www.bodc.ac.uk/solas_integration/implementation_products/group1/aerosol_rain/), and from sources cited in Table 4. Given the aerosol concentration (C), the dry deposition flux (F) was estimated using the following equation: $F = CV_d$, and by assuming deposition velocities (V_d) of 1 cm s^{-1} for NO_3^- (Duce et al., 1991) and 0.6 cm s^{-1} for NH_4^+ (Spokes et al., 2000). Average interpolated values for the basin were obtained at 1° × 1° resolution for each basin (Arabian Sea and Bay of Bengal) and were then averaged to estimate total and dry deposition to the Northern Indian Ocean. The atmospheric model results suggest a total annual deposition of 2.5 kg N ha^{-1} y^{-1} over the entire Arabian Sea. The inorganic N inputs are in line with previous models (including Dentener et al. (2006) and Vet et al. (2014) estimates) and ground-based estimates (0.06-0.15 mg N m^{-2} d^{-1} (Baker et al., 2017; Singh et al., 2012).

Implications for oceanic N_2O production in the Northern Indian Ocean

Estimates from global models: The impacts of increasing levels of anthropogenically derived atmospheric nitrogen deposition on marine biogeochemistry and N_2O production at the global scale have been investigated by recent ocean biogeochemistry model analyses including Suntharalingam et al. (2012) and Jickells et al. (2017). These studies estimate relatively modest increases, at the global scale, in oceanic N_2O production (1-3%), due to changes in nitrogen deposition. However, they 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 suggested as one of the most potentially sensitive regions to changes in this anthropogenic nutrient input. This sensitivity is ascribed to a combination of high nitrogen deposition fluxes originating from the Indian sub-continental outflow, and enhanced N_2O formation in the region's hypoxic ocean zones. Here we summarize model derived changes in N_2O production in the Arabian Sea and Bay of Bengal from a range of analyses. The investigations of Suntharalingam et al. (2012) and Jickells et al. (2017) both employed the PlankTOM ocean biogeochemistry model 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). Further details of model specifications are given in the individual publications.

Table 5. Model-derived estimates of changes in global and Northern Indian Ocean N_2O production due to the impact of external reactive nitrogen input (atmospheric deposition and rivers). Estimates summarize results from the NEMO-PlankTOM model analyses of Suntharalingam et al. (2012) and Jickells et al. (2017), and the UVic2.9 Earth System Model of Landolfi et al. (2017).

REGION	***Present day oceanic N_2O source * (Tg N y^{-1})***	***Change in N_2O source due to Nr inputs (pre-industrial to present day) (Tg N y^{-1})***
Northern Indian Ocean	0.29 – 0.58	0.003 – 0.054
Global Ocean	4.5 – 4.7	0.037 – 0.165

** Present day values derived from Base Case model simulations for period 1995-2005*

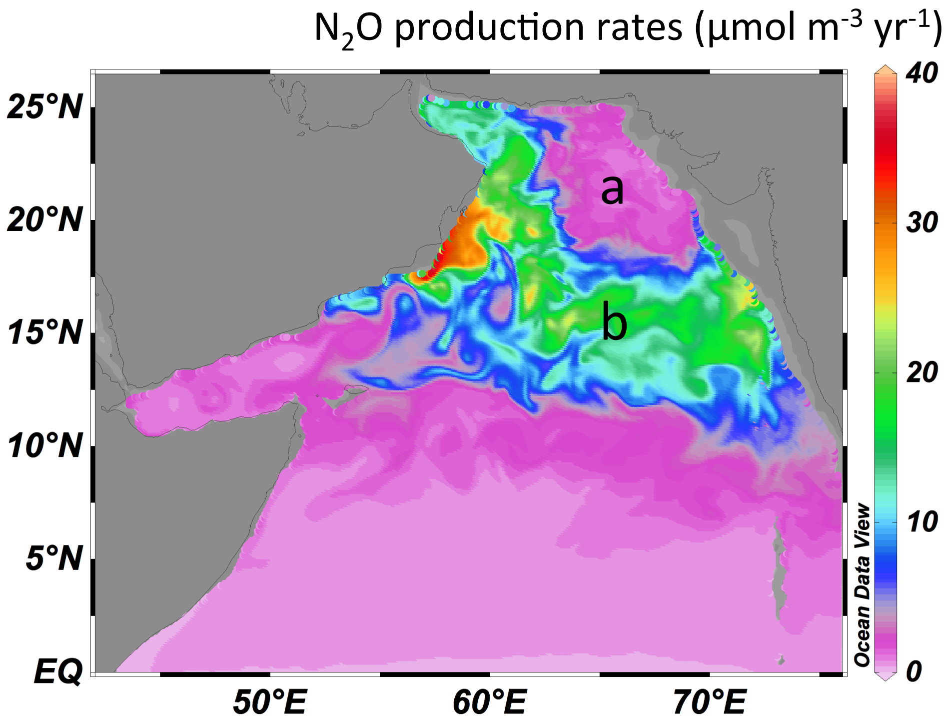

Figure 15. Modelled net N_2O production rates (μmol m^{-3} y^{-1}) in the Arabian Sea. Distributions are shown at a depth of 534 m, where the separate regions of (a) high N_2O consumption, and (b) high N_2O production are most distinct.

Estimated impact on Arabian Sea N_2O from diagnostic analyses of a high- resolution regional model: We also present a new diagnostically derived estimate of oceanic N_2O production in the Arabian Sea, based on biogeochemical fluxes and fields from the regional eddy-resolving (1/12 degree resolution) ocean model of Resplandy et al. (2011, 2012) (Figure 15). This high resolution model produces an improved representation of mesoscale variability, circulation patterns of the Arabian Sea (e.g. monsoonal upwelling), and local biological productivity, in comparison to global lower resolution models. Particularly relevant to N_2O are the good simulation of the regional oxygen distribution (e.g. ambient concentrations and oxygen utilization rates), and the improved representation of the Arabian Sea oxygen minimum zone.

Our diagnostic estimate of regional N_2O production is derived from the parameterizations of Suntharalingam et al. (2012) applied to the gridded biogeochemical and flux distributions (specifically oxygen and oxygen utilization rates) of the high-resolution simulations of Resplandy et al. (2012). We estimate gridded fields of N_2O production as the sum of: (i) N_2O from nitrification in oxygenated waters; (ii) enhanced N_2O production in low oxygen conditions (via denitrification and enhanced nitrification); (iii) N_2O consumption in conditions close to anoxia. The Standard model estimate of net N_2O production in the Arabian Sea region is 0.23 Tg Ny^{-1}, with a range of 0.1-0.56 Tg Ny^{-1} for sensitivity analyses conducted on variations in N_2O yield rates. This is consistent with previous observationally-derived estimates of N_2O flux for the Arabian Sea (e.g. 0.2-0.4 Tg Ny^{-1} (Bange et al., 2005). We derive diagnostic estimates of the impact of changes in reactive nitrogen deposition in this region by combining N_2O production estimates from the regional model together with nitrogen deposition fluxes from Duce et al. (2008), and constrained by N_2O rates of increase in the Arabian Sea from the model results of Suntharalingam et al. (2012). We estimate a resulting change in Arabian Sea N_2O production from the pre-industrial to the present of 0.012-0.05 Tg N (the range reflects sensitivity analyses on N_2O yield rates). This corresponds to an estimated increase of 5% - 20% of the Arabian Sea N_2O source, and is significantly larger than the average estimated change for global N_2O (1% - 3%). This high estimated impact highlights the need for more detailed evaluation of the influence of anthropogenic nitrogen inputs in the Northern Indian Ocean, through the development of high-resolution regional models of nitrogen and N_2O cycling, together with improved characterization, via in-situ and laboratory measurements, of the rates of N_2O cycling in these hypoxic and sub-oxic zones.

Implications of increasing nitrogen deposition for changes in water column denitrification and N_2O cycling: Following the analysis of Landolfi et al. (2013), we estimate the maximum fraction of the total pre-existing N pool that could be removed by atmospheric

N-driven denitrification (Figure 16). We analyse modelled N deposition fields together with regional oxygen data (Bianchi et al., 2012). To assess locations of most likely impact, we made the following simplifying assumptions: (1) atmospheric N 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 the suboxic zone curve from Van Mooy et al. (2002); (3) every mole of N remineralized within the OMZ is processed by denitrification; (4) standard denitrification stoichiometry in OMZ environments (e.g. Paulmier et al. (2009), such that for every 1 mol N from organic matter remineralised in sub-oxic waters, up to 7 additional moles of ambient seawater nitrate N may be lost to N_2 due to its role as an electron acceptor in the denitrification process (Codispoti et al., 2001; Landolfi et al., 2013). These assumptions do not account for such factors as advection-driven redistributions of deposited atmospheric N, non-Redfield stoichiometry of organic matter, or any deposition-driven expansion of OMZs. These processes will have impacts of differing sign and magnitude and are issues in need of further investigation.

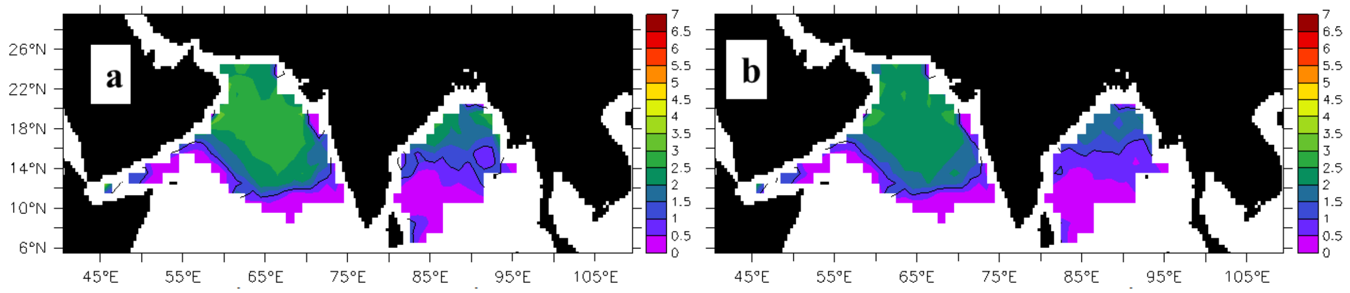

Figure 16. Illustration of the possible ratio of net N loss: net N gain (mol:mol) from atmospheric N deposition to OMZs in the North Indian Ocean assuming either a) the Martin curve, or b) the Van Mooy curve. Calculations based on World Ocean Atlas O_2 data.

3. OTHER ACTIVITIES OF WORKING GROUP 38 ON THE IMPACTS OF NITROGEN DEPOSITION

3.1 European Geosciences Union Symposia

In 2013, WMO requested Working Group 38 to organize a symposium at the 2014 European Geosciences Union Assembly in Vienna, Austria. The working group organized a symposium entitled "Air-Sea Exchanges: Impacts on Biogeochemistry and Climate", and the symposium was so successful that it has been repeated every year since at the EGU Assembly, including 2018. Papers have been presented by Working Group 38 members as well as other scientists, and the topics have included not only issues addressed by Working Group 38 during its meetings, but also many other topics in this general area.

3.2 Interactions with the WMO Global Atmosphere Watch (GAW)

WMO's Global Atmosphere Watch (GAW) has been the primary supporter of Working Group 38 since its inception. During 2016 and 2017, there has been additional interaction between Working Group 38 and its members and GAW. For example, GAW held an International Workshop on the Nitrogen Cycle in York, UK, on 13-14 April 2016. Working Group members presented 3 papers at that workshop:

> "GESAMP Working Group 38 and its oceanic nitrogen deposition studies" by Robert Duce
>
> "Atmospheric Nitrogen Deposition to the Oceans: Observation- and Model-Based Estimates" by Alex Baker
>
> "Atmospheric Nitrogen Inputs to the Oceans and their Impact" by Tim Jickells"

Robert Duce, representing WG 38, participated in the 3rd Scientific Steering Committee meeting of GAW held in Geneva in March 2016, and presented information on the current activities of Working Group 38.

Tim Jickells represented WG 38 (via a remote connection) at the GAW workshop on Measurement-Model Fusion for Global Total Atmospheric Deposition held in Geneva from 28 February to 2 March 2017, with a presentation on "Observation and Model based Estimates of Atmospheric Inputs to the Oceans".

Tim Jickells represented WG 38 and GESAMP at the GAW Symposium held in Geneva from 10 to 13 April 2017, giving one presentation to the plenary session on the activities of GESAMP and then a presentation on "Observations and Modelling Needs to Understand the Impacts of Nitrogen Inputs to the Oceans" at a side event on "How can GAW contribute to the N cycle assessment?"

Tim Jickells represented WG 38 and GESAMP at The Third Informal Meeting of the International Law Commission (ILC) on the Protection of the Atmosphere as part of the Dialogue with Scientists meeting held in Geneva on 5 May 2017, giving a presentation entitled "Linkages between the oceans and the atmosphere" and participating in the subsequent discussion.

3.3 Interactions with the International Nitrogen Management System (INMS)

An additional potential future activity in cooperation with GAW is the assessment of the impact of nitrogen on the marine environment as a contribution to the Integrated Nitrogen Management System (INMS). INMS is a global targeted research project with the aim to provide clear scientific evidence to inform future international nitrogen policy development. INMS's core funding comes from the Global Environment Facility (GEF) (the environment funding mechanism of the United Nations System) with UN Environment as the Implementing Agency and the UK Natural Environment Research Council (Centre for Ecology and Hydrology) as the Executing Agency acting on behalf of the International Nitrogen Initiative (INI). WG 38 is in an excellent position to bring together observational scientists and atmospheric modelling groups to address these issues. Funding for this effort would come from the INMS. Preliminary discussions with INMS have been held but no clear joint programme identified to date.

4. CONCLUSIONS AND FUTURE DIRECTIONS

This synthesis by GESAMP WG 38 provides new current best estimates of nitrogen inputs to the ocean from the atmosphere (39 TgN y^{-1}), and for context synthesises comparable estimates of inputs from rivers (34 TgN y^{-1}) and natural biological nitrogen fixation (164 TgN y^{-1}). Most of the atmospheric nitrogen input reaches the open ocean beyond the shelf break, while a substantial part of the riverine input is trapped on the shelf. Both the riverine and atmospheric nitrogen inputs have been substantially increased by human activity, with the atmosphere now the main vehicle by which anthropogenic nitrogen reaches the open ocean. The atmospheric input of nitrogen is estimated to now be almost 4 times that in 1850, and even in 1850 conditions were not pristine.

Atmospheric deposition of nitrogen to the oceans involves several distinct chemical components, each of approximately the same magnitude; oxidised nitrogen, primarily nitrate aerosol and nitric acid; reduced nitrogen, primarily ammonium aerosol and ammonia; and a poorly characterised organic nitrogen component. Identification of sources is important for the effective management of nitrogen inputs to the ocean. The main anthropogenic source of oxidised nitrogen is fossil fuel combustion on land plus an increasingly important source from fuel combustion on ships, while for reduced nitrogen the primary anthropogenic emission source is from intensive agriculture. There is also an important but poorly understood natural recycling of ammonia and organic nitrogen between the atmosphere and the oceans. The quantification of the net magnitude of atmospheric nitrogen inputs to the ocean and their impact is sensitive to the uncertainties in the magnitude of this recycling.

Atmospheric nitrogen emissions come predominantly from areas of high fossil fuel combustion and high rates of intensive agriculture. The largest emission sources are in North America, Europe, India and South-East Asia. Models based on future emission scenarios suggest that total nitrogen inputs to the oceans will change little between now and 2050, but that emissions are likely to increase over southern Asia and decline over North America and Europe. The largest inputs of nitrogen to the oceans occur downwind of these large emission sources over the North Atlantic, Northern Indian and north-west Pacific Oceans. Impacts of this atmospheric deposition on the marine environment have been previously suggested for the north-west Pacific, and impacts in this region and the Northern Indian Ocean are likely to increase in the future, based on the emission scenarios considered. Such impacts may include increases in phytoplankton production, and in the north-western Indian Ocean this may lead to increases in the emissions of the greenhouse gas N_2O from the low oxygen waters that occur naturally at depth in this region.

More generally the impact of nitrogen deposition to the ocean will be an increase in primary production in regions that are currently nitrogen limited, which include the surface waters of tropical ocean gyres. The increase in ocean production at the present day compared to 1850 levels is estimated to be about 0.4%, with an associated increase in the ocean uptake of CO_2 of 0.15Pg C y^{-1}. This estimate is very sensitive to assumptions about feedbacks that involve atmospheric nitrogen deposition suppressing nitrogen fixation. There is also, on a longer time scale, a sensitivity to feedbacks in which increasing nitrogen inputs to the ocean increase primary production and organic matter inputs to the deep ocean, increasing denitrification and anammox and leading to increased emissions of N_2 and N_2O gas.

Future Research Needs

WG 38 has identified a number of areas where further research could reduce uncertainties in both the estimated magnitude of net nitrogen input to the ocean and its impacts and thereby better informing policy makers.

1. We require a better understanding the sources and bioavailability of organic nitrogen as well as the recycling of both organic nitrogen and ammonia between the atmosphere and ocean.

2. The estimation of atmospheric deposition of nitrogen to the ocean depends on parameterisation of wet and dry deposition in models, and work to improve and validate these parameterisations would be valuable.

3. The work of WG 38 suggests that fluvial nitrogen is trapped on the shelf, particularly at high latitudes, and this prediction needs to be confirmed by further work.

4. Our assessment of the impacts of atmospheric deposition on ocean productivity are extremely sensitive to assumptions about the extent of suppression of nitrogen fixation by atmospheric deposition, and this sensitivity requires further research.

5. Further work is also required to test predictions from models of long term feedbacks between atmospheric inputs to the ocean and nitrogen loss by denitrification and anammox.

6. The confirmation that atmospheric inputs of nitrogen are the main vehicle by which the large scale perturbations of the global nitrogen cycle reach the open ocean suggest that further research generally is required to understand the impacts of this deposition. In addition, the WG 38 synthesis has identified the Northwest Pacific and Northern Indian Ocean as areas of the oceans that currently receive high inputs of atmospheric nitrogen and where there are also likely to be substantial changes in the future, with potential impacts at regional and global scales. We recommend further research in these regions to allow better predictions of the likely impacts of projected future increased atmospheric inputs. We also note that there are areas of the Mediterranean and North Atlantic where primary production is phosphorus- or iron-limited and hence where additional nitrogen deposition may lead to different nutrient biogeochemical responses to those in other ocean areas where nitrogen is the primary limiting nutrient. These areas also deserve further research.

Finally, we note that ongoing changes in nitrogen and sulphur atmospheric emissions will lead to decreases in atmospheric acidity, and the impact of this on the bioavailability of particularly atmospherically delivered trace metals to the ocean merits further work. In addition, continuing large-scale emissions of CO_2 from human activity is acidifying the ocean, and this is likely to change the emission of a variety of trace gases from the ocean including ammonia and N_2O. However, we do not know the scale or even the sign of such changes and this merits further research.

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ANNEX 1 - AGENDA

AGENDA

GESAMP Workshop on The Atmospheric Deposition of Nitrogen and Its Impact on Marine Biogeochemistry

11-14 February 2013

University of East Anglia, Norwich, United Kingdom

Sunday, 10 February
Participants arrive in Norwich

Monday, 11 February

0900-0915: Welcome, opening of workshop, logistics and schedules - Peter Liss

0915-0930: Scientific goals and approaches of the workshop - Bob Duce and Peter Liss

0930-1100: Presentation and discussion of updates on the modeling of the atmospheric input of nitrogen to the global ocean - led by Frank Dentener and Maria Kanakidou (**Topic 1)**

1100-1130: Coffee Break

1130-1300: Presentation and discussion of the impact of the atmospheric deposition of nitrogen on marine productivity and CO_2 drawdown - led by Doug Capone and Keith Moore (**Topic 2)**

1300-1400: Lunch

1400-1515: Presentation and discussion of the impact of the atmospheric deposition of nitrogen on marine nitrous oxide formation - led by Parv Suntharalingam and Sybil Seitzinger **(Topic 3)**

1515-1630: Presentation and discussion of the extent to which nitrogen delivered to the coastal zone is transported to the open ocean - led by Katja Fennel and Jonathan Sharples **(Topic 4)**

1630-1645: Coffee break

1645-1800: Presentation and discussion of the impact of atmospheric nitrogen in the northern Indian Ocean and South China Sea regions - led by Monmohan Sarin and Kitack Lee **(Topic 5)**

1800-1815: Summary and discussion of next day's activities - Peter Liss and Bob Duce

Tuesday, 12 February

0830-1300: Separate meetings and discussions within the five topic groups

1300-1400: Lunch

1400-1600: Continuation of topic group discussions

1600-1800: Plenary session, summary of the results from the topic group discussions

Wednesday, 13 February

0830-1300: Separate meetings and discussions within the five topic groups

1300-1400: Lunch

1400-1600: Continuation of topic group discussions

1600-1800: Plenary session, summary of the results from the topic group discussions

Thursday, 14 February

0830-0930: Final meetings of the individual topic groups

0930-1030: Final report and discussion of Topic 1

1030-1100: Coffee break

1100-1200: Final report and discussion of Topic 2

1200-1300: Final Report and discussion of Topic 3

1300-1400: Lunch

1400-1500: Final Report and discussion of Topic 4

1500-1600: Final Report and discussion of Topic 5

1600-1645: Workshop summary and outline of next steps

End of Workshop

ANNEX 2 - GESAMP Working Group 38 Membership during the Nitrogen Studies

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ANNEX 3 - GESAMP Reports and Studies publications

The following reports and studies have been published so far. They are available from the GESAMP website: http://gesamp.org

1. Report of the seventh session, London, 24-30 April 1975. (1975). Rep. Stud. GESAMP, (1):pag.var. Available also in French, Spanish and Russian
2. Review of harmful substances. (1976). Rep. Stud. GESAMP, (2):80 p.
3. Scientific criteria for the selection of sites for dumping of wastes into the sea. (1975). Rep. Stud. GESAMP, (3):21 p. Available also in French, Spanish and Russian
4. Report of the eighth session, Rome, 21-27 April1976. (1976). Rep. Stud. GESAMP, (4):pag. var. Available also in French and Russian
5. Principles for developing coastal water quality criteria. (1976). Rep. Stud. GESAMP, (5):23 p.
6. Impact of oil on the marine environment. (1977). Rep. Stud. GESAMP, (6):250 p.
7. Scientific aspects of pollution arising from the exploration and exploitation of the sea-bed. (1977). Rep. Stud. GESAMP, (7):37 p.
8. Report of the ninth session, New York, 7-11 March 1977. (1977). Rep. Stud. GESAMP, (8):33 p. Available also in French and Russian
9. Report of the tenth session, Paris, 29 May - 2 June 1978. (1978). Rep. Stud. GESAMP, (9):pag.var. Available also in French, Spanish and Russian
10. Report of the eleventh session, Dubrovnik, 25-29 February 1980. (1980). Rep. Stud. GESAMP, (10):pag.var. Available also in French and Spanish
11. Marine Pollution implications of coastal area development. (1980). Rep. Stud. GESAMP, (11):114 p.
12. Monitoring biological variables related to marine pollution. (1980). Rep. Stud. GESAMP, (12):22 p. Available also in Russian
13. Interchange of pollutants between the atmosphere and the oceans. (1980). Rep. Stud. GESAMP, (13):55 p.
14. Report of the twelfth session, Geneva, 22-29 October 1981. (1981). Rep. Stud. GESAMP, (14):pag.var. Available also in French, Spanish and Russian
15. The review of the health of the oceans.(1982).Rep. Stud. GESAMP, (15):108 p.
16. Scientific criteria for the selection of waste disposal sites at sea. (1982). Rep. Stud. GESAMP, (16):60 p.
17. The evaluation of the hazards of harmful substances carried by ships. (1982). Rep. Stud. GESAMP, (17):pag.var.
18. Report of the thirteenth session, Geneva, 28 February - 4 March 1983. (1983). Rep. Stud. GESAMP, (18):50 p. Available also in French, Spanish and Russian
19. An oceanographic model for the dispersion of wastes disposed of in the deep sea. (1983). Rep. Stud. GESAMP, (19):182 p.
20. Marine pollution implications of ocean energy development. (1984). Rep. Stud. GESAMP, (20):44 p.
21. Report of the fourteenth session, Vienna, 26-30 March 1984. (1984). Rep. Stud. GESAMP, (21):42 p. Available also in French, Spanish and Russian
22. Review of potentially harmful substances. Cadmium, lead and tin. (1985). Rep. Stud. GESAMP, (22):114 p.
23. Interchange of pollutants between the atmosphere and the oceans (part II). (1985). Rep. Stud. GESAMP, (23):55 p.
24. Thermal discharges in the marine Environment. (1984). Rep. Stud. GESAMP, (24):44 p.
25. Report of the fifteenth session, New York, 25-29 March 1985. (1985). Rep. Stud. GESAMP, (25):49 p. Available also in French, Spanish and Russian

26. Atmospheric transport of contaminants into the Mediterranean region. (1985). Rep. Stud. GESAMP, (26):53 p.

27. Report of the sixteenth session, London, 17-21 March 1986. (1986). Rep. Stud. GESAMP, (27):74 p. Available also in French, Spanish and Russian

28. Review of potentially harmful substances. Arsenic, mercury and selenium. (1986). Rep. Stud. GESAMP, (28):172 p.

29. Review of potentially harmful substances. Organosilicon compounds (silanes and siloxanes). (1986). Published as UNEP Reg. Seas Rep. Stud. (78):24 p.

30. Environmental capacity. An approach to marine pollution prevention. (1986). Rep. Stud. GESAMP, (30):49 p.

31. Report of the seventeenth session, Rome, 30 March - 3 April 1987. (1987). Rep. Stud. GESAMP, (31):36 p. Available also in French, Spanish and Russian

32. Land-sea boundary flux of contaminants: contributions from rivers. (1987). Rep. Stud. GESAMP, (32):172 p.

33. Report on the eighteenth session, Paris, 11-15 April 1988. (1988). Rep. Stud. GESAMP, (33):56 p. Available also in French, Spanish and Russian

34. Review of potentially harmful substances. Nutrients. (1990). Rep. Stud. GESAMP, (34):40 p.

35. The evaluation of the hazards of harmful substances carried by ships: Revision of GESAMP Reports and Studies No. 17. (1989). Rep. Stud. GESAMP, (35):pag.var.

36. Pollutant modification of atmospheric and oceanic processes and climate: some aspects of the problem. (1989). Rep. Stud. GESAMP, (36):35 p.

37. Report of the nineteenth session, Athens, 8-12 May 1989. (1989). Rep. Stud. GESAMP, (37):47 p. Available also in French, Spanish and Russian

38. Atmospheric input of trace species to the world ocean. (1989). Rep. Stud. GESAMP, (38):111 p.

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41. Report of the twentieth session, Geneva, 7-11 May 1990. (1990). Rep. Stud. GESAMP, (41):32 p. Available also in French, Spanish and Russian

42. Review of potentially harmful substances. Choosing priority organochlorines for marine hazard assessment. (1990). Rep. Stud. GESAMP, (42):10 p.

43. Coastal modelling. (1991). Rep. Stud. GESAMP, (43):187 p

44. Report of the twenty-first session, London, 18-22 February 1991. (1991). Rep. Stud. GESAMP, (44):53 p. Available also in French, Spanish and Russian

45. Global strategies for marine environmental protection. (1991). Rep. Stud. GESAMP, (45):34 p.

46. Review of potentially harmful substances. Carcinogens: their significance as marine pollutants. (1991). Rep. Stud. GESAMP, (46):56 p.

47. Reducing environmental impacts of coastal aquaculture. (1991). Rep. Stud. GESAMP, (47):35 p.

48. Global changes and the air-sea exchange of chemicals. (1991). Rep. Stud. GESAMP, (48):69 p.

49. Report of the twenty-second session, Vienna, 9-13 February 1992. (1992). Rep. Stud. GESAMP, (49):56 p. Available also in French, Spanish and Russian

50. Impact of oil, individual hydrocarbons and related chemicals on the marine environment, including used lubricant oils, oil spill control agents and chemicals used offshore. (1993). Rep. Stud. GESAMP, (50):178 p.

51. Report of the twenty-third session, London, 19-23 April 1993. (1993). Rep. Stud. GESAMP, (51):41 p. Available also in French, Spanish and Russian

52. Anthropogenic influences on sediment discharge to the coastal zone and environmental consequences. (1994). Rep. Stud. GESAMP, (52):67 p.

53. Report of the twenty-fourth session, New York, 21-25 March 1994. (1994). Rep. Stud. GESAMP, (53):56 p. Available also in French, Spanish and Russian

54. Guidelines for marine environmental assessment. (1994). Rep. Stud. GESAMP, (54):28 p.

55. Biological indicators and their use in the measurement of the condition of the marine environment. (1995). Rep. Stud. GESAMP, (55):56 p. Available also in Russian

56. Report of the twenty-fifth session, Rome, 24-28 April 1995. (1995). Rep. Stud. GESAMP, (56):54 p. Available also in French, Spanish and Russian

57. Monitoring of ecological effects of coastal aquaculture wastes. (1996). Rep. Stud. GESAMP, (57):45 p.

58. The invasion of the ctenophore Mnemiopsis leidyi in the Black Sea. (1997). Rep. Stud. GESAMP, (58):84 p.

59. The sea-surface microlayer and its role in global change. (1995). Rep. Stud. GESAMP, (59):76 p.

60. Report of the twenty-sixth session, Paris, 25-29 March 1996. (1996). Rep. Stud. GESAMP, (60):29 p. Available also in French, Spanish and Russian

61. The contributions of science to integrated coastal management. (1996). Rep. Stud. GESAMP, (61): 66 p.

62. Marine biodiversity: patterns, threats and development of a strategy for conservation. (1997). Rep. Stud. GESAMP, (62):24 p.

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