Global distribution of N₂O emissions from aquatic systems: natural emissions and anthropogenic effects

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Abstract

Context Abstract: Atmospheric concentrations of nitrous oxide, a greenhouse gas, are increasing due to human activities. Our analysis suggests that a third of global anthropogenic N₂O emission is from aquatic sources (rivers, estuaries, continental shelves) and the terrestrial sources comprise the remainder. Over 80% of aquatic anthropogenic N₂O emissions are from the Northern Hemisphere mid-latitudes consistent with the geographic distribution of N fertilizer use, human population and atmospheric N deposition. These N inputs to land have increased aquatic as well as terrestrial anthropogenic N₂O emissions because a substantial portion enters aquatic systems and results in increased N₂O production. Thus, wise management of N in the terrestrial environment could help reduce/control both aquatic and terrestrial N₂O emissions.

Main Abstract: The global distribution of N₂O emissions from rivers, estuaries, continental shelves, and oceans are compared to each other, and to terrestrial emissions, using existing gridded inventories. Rivers, estuaries and continental shelves (1.9 Tg N y⁻¹) account for about 35% of total aquatic N₂O emissions; oceanic emissions comprise the remainder. Oceanic N₂O emissions are approximately equally distributed between the Northern and Southern Hemispheres; however, over 90% of emissions from estuaries and rivers are in the Northern Hemisphere. N₂O emissions from rivers, estuaries, and continental shelves combined equal oceanic emissions in both the 20°–45°N and 45°–66°N latitudinal zones. Over 90% of river and estuary emissions are considered anthropogenic (1.2 Tg N y⁻¹); only 25% of continental shelf emissions are considered anthropogenic (0.1 Tg N y⁻¹); oceanic emissions are considered natural. Overall, approximately one third of both aquatic and of terrestrial emissions are anthropogenic.

Natural terrestrial emissions are highest in tropical latitudes while natural aquatic emissions are relatively evenly distributed among latitudinal zones. Over half of both the anthropogenic terrestrial and aquatic emissions occur between 20° and 66°N. Anthropogenic N inputs to the terrestrial environment drive anthropogenic N₂O emissions from both land and aquatic ecosystems, because a substantial portion of the anthropogenic N applied to watersheds enters rivers, estuaries and continental shelves. © 2000 Elsevier Science Ltd. All rights reserved.

Keywords: Nitrous oxide; Aquatic; Rivers; Estuaries; Continental shelves; Anthropogenic; Global emissions; Geographic distribution

1. Introduction

Total global N₂O emissions include a wide range of sources from terrestrial and aquatic systems, with both natural and anthropogenic components (e.g., Khalil and Rasmussen, 1992; Matthews, 1994; Bouwman et al., 1995; Bange et al., 1996). A clear understanding of the
magnitude of the various natural and anthropogenic sources and their geographic distribution is necessary to quantify factors contributing to the current increases in atmospheric N$_2$O concentrations, to predict future potential increases, and to develop realistic and cost-effective strategies to reduce future emissions. There have been a number of analyses of the magnitude and global distribution of terrestrial and deep ocean sources (e.g., Nevison, 1994; Bouwman et al., 1995); however only recently have such analyses been done for freshwater and coastal marine systems (Seitzinger and Kroeze, 1998).

N$_2$O emissions from freshwater and coastal marine ecosystems are estimated to amount to 1.9 Tg N y$^{-1}$ (Seitzinger and Kroeze, 1998), which is similar in magnitude to estimated N$_2$O emissions from a number of other sources, including arable land, grasslands, and animal excreta (Bouwman et al., 1995). Much of the N$_2$O emission from these aquatic systems is anthropogenic and thus contributes to the current increase in atmospheric N$_2$O. In addition, large increases in future N$_2$O emissions from these aquatic systems are predicted as a result of increases in human activities (Kroeze and Seitzinger, 1998). Bouwman et al. (1995) presented an analysis of the total and latitudinal distribution of most of the known terrestrial sources and Nevison et al. (1995) has done so with oceanic sources. We now extend that analysis by adding freshwater and coastal marine N$_2$O emissions.

In the current paper we compare: (1) N$_2$O emissions from rivers, estuaries and continental shelves with N$_2$O emissions from oceanic regions, (2) anthropogenic and natural emissions from aquatic systems, and (3) aquatic and terrestrial N$_2$O emissions, both natural and anthropogenic. These comparisons are done at a global scale as well as on a latitudinal basis to provide insight into the geographic distribution of the different sources. Gridded (1$^\circ$×1$^\circ$) estimates of N$_2$O emissions from published studies are used in these analyses.

2. Methods

We calculated N$_2$O emissions for the following latitudinal zones for the northern and southern hemisphere: 0$^\circ$–20$^\circ$, 20$^\circ$–45$^\circ$, 45$^\circ$–66$^\circ$ and 66$^\circ$–90$^\circ$.

**Emissions from oceans:** We obtained the gridded (1$^\circ$×1$^\circ$) global N$_2$O emission inventory for ocean emissions from Nevison (1994) through the Emission Database for Global Atmospheric Research (EDGAR) (Olivier et al., 1996). The EDGAR database had been converted from the original 2.8$^\circ$×2.8$^\circ$ grid of Nevison (1994) to a 1$^\circ$×1$^\circ$ grid, which resulted in a loss of 0.2 Tg N y$^{-1}$ from the original 3.8 Tg N y$^{-1}$ estimate (Bouwman et al., 1995). That database (1$^\circ$×1$^\circ$) is based on Nevison’s “best” estimate emissions which she calculated using the water–air flux calculation of Erickson (1993). It is also available from the Global Emissions Inventory Activity (GEIA) web site (http://blueskies.sprl.umich.edu/geia). We excluded from Nevison’s database the N$_2$O emissions from the area that is considered continental shelf by Seitzinger and Kroeze (1998), i.e., the gridcells with an average depth <200 m. The totals for N$_2$O emissions from oceans by latitudinal zones then were calculated by summing the N$_2$O emissions from all gridcells with an average depth >200 m.

**Emissions from continental shelves:** We used the gridded (1$^\circ$×1$^\circ$) global N$_2$O inventory from Seitzinger and Kroeze (1998). The totals for latitudinal zones were calculated by summing the emissions from the continental shelf gridcells within these zones.

**Emissions from estuaries:** We used the gridded (1$^\circ$×1$^\circ$) global N$_2$O inventory from Seitzinger and Kroeze (1998). The totals for latitudinal zones were calculated by summing the emissions from the estuarine gridcells within these zones.

**Emissions from rivers:** We used the gridded (1$^\circ$×1$^\circ$) global N$_2$O inventory from Seitzinger and Kroeze (1998). The total emissions for latitudinal zones were calculated by summing the emissions from the watershed gridcells within a latitudinal zone. This differs from the approach used by Seitzinger and Kroeze (1998) to assign river N$_2$O emissions to latitudinal zones; they assigned the N$_2$O emissions from an entire watershed to the latitude of the river mouth for that watershed.

**Terrestrial emissions:** We used the gridded (1$^\circ$×1$^\circ$) global N$_2$O emission inventory for terrestrial emissions from different source categories from Bouwman et al. (1993, 1995) which we obtained through the EDGAR (Olivier et al., 1996). It is also available through the GEIA website (http://blueskies.sprl.umich.edu/geia). The terrestrial emission source categories distinguished by Bouwman et al. (1995) include natural soils, arable land, animal excreta, biomass burning, agricultural waste burning, post-clearing effects of deforestation, industry, energy and fuelwood combustion (Bouwman et al., 1995). In the EDGAR database several of these sources have been broken down into subcategories, totalling 16 sources of N$_2$O. Emissions from a 17th subcategory, fossil fuel combustion from air transportation, were not available as a gridded database from EDGAR, so they were not included in our terrestrial totals. N$_2$O emissions from this source are 0.003 Tg N y$^{-1}$, which is equivalent to 0.09% of the total anthropogenic terrestrial emissions (Olivier et al., 1996).

We used the aquatic inventories identified above in the current analysis because as far as we are aware they are the only gridded inventories available for rivers, estuaries, continental shelves or oceans. Details of the methods used to estimate emissions from each of these sources and uncertainties in each inventory are discussed in the original publications (Nevison et al., 1995; Kroeze...
and Seitzinger, 1998; Seitzinger and Kroeze, 1998). There have been a number of estimates of terrestrial \( N_2O \) emission from particular sources (e.g., Matthews, 1994; Nevison, 1994) and some additional sources or modifications to the above estimates (discussed below) since Bouwman et al. (1995). We used the inventories of Bouwman et al. (1995) because of their relative completeness and availability in gridded format. The reader is referred to Bouwman et al. (1993, 1995) for discussion of the uncertainties in those terrestrial inventories.

3. Results and discussion

3.1. Aquatic \( N_2O \) emissions

\( N_2O \) emissions from rivers, estuaries and continental shelves: Global watersheds receive inputs of nitrogen (N) from a variety of anthropogenic sources (Fig. 1). The three anthropogenic sources of new fixed N are synthetic fertilizer, combustion of fossil fuels and biological N\(_2\)-fixation associated with cultivated crops. Together, these sources contribute an amount of N (about 90–130 Tg N y\(^{-1}\)) similar to the natural biological fixation of N\(_2\) in non-agricultural soils (~140 Tg N y\(^{-1}\)) (Galloway et al., 1995). A considerable amount of the anthropogenic N inputs to terrestrial ecosystems eventually enters rivers through leaching, runoff and direct discharge. A portion of this N in rivers is transported to coastal marine ecosystems (estuaries and continental shelves). The biological cycling of natural and anthropogenic nitrogen through these aquatic ecosystems results in \( N_2O \) emissions from each of these aquatic systems (estuaries and continental shelves). The biological cycling of natural and anthropogenic nitrogen through these aquatic ecosystems results in \( N_2O \) emissions from each of these aquatic systems (estuaries and continental shelves).

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Fig. 1. Schematic of N inputs to aquatic ecosystems. N inputs to rivers and estuaries are linked to N inputs to the surrounding watersheds from both natural and anthropogenic sources. Continental shelves receive N inputs from both terrestrial sources (river/estuarine export and atmospheric deposition) and from oceanic (e.g., onwelling, advection) sources. Subsequent cycling of N through nitrification and denitrification pathways results in \( N_2O \) emissions from these aquatic systems.
from aquatic ecosystems which is that from oceanic regions (3.5 Tg N y\(^{-1}\); Nevison’s (1994) inventory minus emissions from continental shelf gridcells). Global emissions from lakes have not been estimated. Overall the N\(_2\)O emissions from freshwater and coastal systems (1.9 Tg N y\(^{-1}\)) account for about 35% of the total aquatic emissions (5.4 Tg N y\(^{-1}\)) (Fig. 2).

The uncertainties in the freshwater and coastal marine N\(_2\)O emissions estimates are considerable. For rivers, estuaries and continental shelves (1.9 Tg N y\(^{-1}\)), the estimated uncertainty range is 0.9–9.0 Tg N y\(^{-1}\), based on uncertainties in emission factors only (Seitzinger and Kroeze, 1998). The emission estimates were derived by using different emission factors, which depended on the external N inputs (0.3 or 3% of nitrification and denitrification rates; see above). A sensitivity analysis indicated that a twofold increase or decrease in the threshold N input value used to determine the emission factor would not markedly alter the overall conclusions of the study by Seitzinger and Kroeze (1998), given the other model uncertainties. Their analysis furthermore indicated that the calculated emissions from rivers are less sensitive than emissions from estuaries to changes in the threshold N input. Unfortunately, there are few rivers or estuaries where N\(_2\)O emissions have been measured in conjunction with N input over the temporal (yearly) and spatial (entire river or estuary) scales necessary to compare with the model output. A comparison of the modeled N\(_2\)O emissions from the Amazon River and Tamar and Narragansett estuaries indicated that the modeled emissions agree within a factor of 2–5. However, additional comparisons across a range of rivers and estuaries are needed to validate the modeled N\(_2\)O fluxes.

The estimates for aquatic emissions of nitrous oxide are likely to be more uncertain than emissions from terrestrial systems, primarily because there are many more studies of N\(_2\)O emissions in terrestrial systems. Van Aardenne et al. (1998) analyzed the uncertainties in agricultural emissions of nitrous oxide from terrestrial systems for the Netherlands, following the methodology in Mosier et al. (1998). Van Aardenne et al. (1998) also estimated aquatic N\(_2\)O emissions associated with external N inputs (leaching) to aquatic systems from agricultural sources using the methodology in Mosier et al. (1998) which is similar to that of Seitzinger and Kroeze (1998). Van Aardenne et al. (1998) concluded from a Monte Carlo analysis, that of the different model parameters analyzed, relatively large contributions to the total uncertainty were due to uncertainties in the emission factors from aquatic systems and agricultural fields, and in the estimated fraction of agricultural nitrogen subject to leaching and runoff.

**The latitudinal distribution of aquatic sources:** There are obvious patterns in the latitudinal distribution of N\(_2\)O emissions from aquatic sources (Fig. 3; Table 1). Oceanic N\(_2\)O emissions are approximately equally distributed between the northern (42%) and southern (58%) hemispheres. Approximately 70% of the total continental shelf emissions are in the northern hemisphere. However, N\(_2\)O emissions from estuaries and rivers are almost exclusively in the northern hemisphere (96% and 98%, respectively).

A more detailed look at the distribution of emissions (Tg N y\(^{-1}\)) by latitudinal zones (0°–20°, 20°–45°, 45°–66° and 66°–90° in Northern and Southern Hemispheres) indicates further differences in the pattern among aquatic sources. For example, the total amount of N\(_2\)O emitted from the oceans in each of the five latitudinal zones between 45°S and 66°N is similar, with total oceanic emissions between 45°S and 66°S being at least twice as great (Fig. 3). Continental shelf emissions also are similar in each of the five latitudinal zones between 45°S and 66°N, although there is a slight trend of decreasing emissions going from north to south. In contrast, N\(_2\)O emissions from rivers or estuaries show considerable variation among latitudinal zones with the 20°–45°N and 45°–66°N zones having the highest total emissions. N\(_2\)O emissions from rivers, estuaries, and continental shelves combined equal oceanic emissions in both the 20°–45°N and 45°–66°N latitudinal zones.

These differences in latitudinal distributions among aquatic sources are related to a variety of factors. Oceanic N\(_2\)O fluxes were calculated by Nevison et al. (1995) by coupling sea–air gas transfer coefficient fields to a map of surface water N\(_2\)O supersaturations obtained from cruise data. In general, the latitudinal distribution of surface N\(_2\)O reflects both wind velocities and the amount of mixing between N\(_2\)O rich deep water and surface waters (Nevison et al., 1995). An overall view of the latitudinal distribution of ocean N\(_2\)O emission per unit area (fluxes) was obtained by combining the ocean area (Fig. 4(A)) with the ocean N\(_2\)O emission for each latitudinal zone (Fig. 3). A pattern of decreasing N\(_2\)O...
emission per unit area of ocean is apparent between 66°N and 45°S (Fig. 4(C)). In the Southern Hemisphere between 45°–66°S there is a large increase in rate per unit area (flux) due to upwelling of N$_2$O rich deep water (Nevison, 1994; Nevison et al., 1995). There is a strong pattern of decreasing continental shelf area going from the Northern to the Southern Hemisphere (Fig. 4(B)); however, the N$_2$O flux, as defined in the current calculations, does not vary greatly among latitudinal zones. This results in the general decrease in total N$_2$O emitted from continental shelf regions per latitudinal zone going from north to south (Fig. 3).

The latitudinal distribution of river and estuary N$_2$O emissions (Fig. 3) is consistent with the distribution of N inputs to watersheds from anthropogenic activities such as fertilizer use (FAO, 1990 modified by Bouwman et al., 1995), human population density (Lerner et al., 1988 updated to 1990 by Bouwman) and atmospheric NO$_y$ deposition (Dentener and Crutzen, 1994) which are dominant in the northern hemisphere between 20°N and 66°N (Fig. 5(A)–(C)).

**Relative magnitude of natural and anthropogenic aquatic N$_2$O emissions:** In the current analysis we attribute N$_2$O emissions to natural or anthropogenic sources depending on the origin (natural or anthropogenic) of the N source to the aquatic system that is nitrified or denitrified. N inputs to oceanic regions are considered to be essentially all natural in that they are primarily from N$_2$-fixation in the oceans; thus N$_2$O emissions in the oceans linked to nitrification and denitrification of that N are considered natural (Table 2). Two potential additional sources of N to oceanic regions are atmospheric deposition and rivers. All N inputs from rivers are likely removed by denitrification and burial in the sediments of estuaries and continental shelves (Christensen et al., 1987; Seitzinger and Giblin, 1996) and thus do not contribute to N inputs to oceanic regions. Anthropogenic NO$_y$ deposition to coastal and oceanic regions is estimated at 9 Tg N yr$^{-1}$ (Galloway et al., 1995). However, given that anthropogenic NO$_y$ deposition is relatively small compared to N inputs from oceanic N$_2$-fixation (wide range of estimates but generally $>$80 Tg N yr$^{-1}$; Pilson, 1998), and that only a portion of the deposition is to oceanic regions, in this analysis it is considered negligible.

N inputs to rivers are from a combination of both natural and anthropogenic sources. Knowing what component of the N in rivers is from natural or anthropogenic sources is difficult to estimate because of a paucity of measurements of N in rivers with no anthropogenic activity in their watersheds. In a recent model of DIN export by world rivers, roughly 25% (5 Tg N yr$^{-1}$) of a total of 20.8 Tg yr$^{-1}$ was estimated to be natural N (Seitzinger and Kroeze, 1998). This estimate was made by simply assuming that natural export of DIN is 40 kg N (km$^{-2}$ watershed) yr$^{-1}$, as estimated by Meybeck (1982) based on available data for relatively unpolluted rivers globally. The actual value is likely to vary geographically (Howarth et al., 1996), however, at present a better estimate is not available. Globally, N$_2$O emissions from rivers are estimated to be 1.1 Tg N yr$^{-1}$ (Seitzinger and Kroeze, 1998). The emission factor used by Seitzinger and Kroeze (1998) to relate N$_2$O emissions to denitrification in rivers varies as a function of N inputs to rivers (see above). Therefore, to estimate natural N$_2$O emissions from rivers, we assumed a natural watershed DIN yield of 40 kg N (km$^{-2}$ watershed) yr$^{-1}$ (Meybeck, 1982) and applied the approach of Seitzinger...
<table>
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<th>Source</th>
<th>N\O emissions by latitudinal zones (Gg N yr(^{-1}))</th>
<th>Total terrestrial and aquatic (Gg N yr(^{-1}))</th>
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<td>Rivers</td>
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<tr>
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<tr>
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<td>149 1958 41.51</td>
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\(^{a}\) Total anthropogenic terrestrial includes all terrestrial sources except natural soils.
Natural \( \text{N}_2\text{O} \) emissions from rivers are estimated to be approximately 5% (0.05 Tg N y\(^{-1}\)) (Table 2). The remaining 95% of global \( \text{N}_2\text{O} \) emissions from rivers, 1.05 Tg N y\(^{-1}\), are considered anthropogenic. In addition to DIN, organic N (both dissolved and particulate) is transported by, and transformed within, rivers; however, \( \text{N}_2\text{O} \) emissions associated with transformations of that organic N have not been estimated.

N inputs to estuaries were estimated based on modeled N export by rivers. Natural \( \text{N}_2\text{O} \) emissions from rivers are estimated to be approximately 5% (0.05 Tg N y\(^{-1}\)) (Table 2). The remaining 95% of global \( \text{N}_2\text{O} \) emissions from rivers, 1.05 Tg N y\(^{-1}\), are considered anthropogenic. In addition to DIN, organic N (both dissolved and particulate) is transported by, and transformed within, rivers; however, \( \text{N}_2\text{O} \) emissions associated with transformations of that organic N have not been estimated.

Fig. 4. Comparison of the latitudinal distribution of: (A) ocean area \( (10^6 \text{ km}^2) \); (B) continental shelf area \( (10^6 \text{ km}^2) \); (C) \( \text{N}_2\text{O} \) emissions for oceans and continental shelves per unit area of ocean or shelf \( (\text{kg N km}^{-2} \text{ y}^{-1}) \).

and Kroeze (1998). Natural \( \text{N}_2\text{O} \) emissions from rivers are estimated to be approximately 5% (0.05 Tg N y\(^{-1}\)) (Table 2). The remaining 95% of global \( \text{N}_2\text{O} \) emissions from rivers, 1.05 Tg N y\(^{-1}\), are considered anthropogenic. In addition to DIN, organic N (both dissolved and particulate) is transported by, and transformed within, rivers; however, \( \text{N}_2\text{O} \) emissions associated with transformations of that organic N have not been estimated.

N inputs to estuaries were estimated based on modeled N export by rivers. Natural \( \text{N}_2\text{O} \) emissions from estuaries (0.02 Tg N y\(^{-1}\); Table 2) were calculated using the above estimate of natural DIN export by rivers (5 Tg N y\(^{-1}\)) and the approach of Seitzinger and Kroeze (1998). Anthropogenic \( \text{N}_2\text{O} \) emissions (0.20 Tg N y\(^{-1}\)) were calculated as the difference between the total estuarine \( \text{N}_2\text{O} \) emissions (0.22 Tg N y\(^{-1}\)) and natural emissions. Therefore, anthropogenic emissions account for 91% and natural emissions only 9% of the \( \text{N}_2\text{O} \) from estuaries.

Continental shelf \( \text{N}_2\text{O} \) emissions are a combination of anthropogenic and natural. Again, we estimated the relative amount of natural and anthropogenic \( \text{N}_2\text{O} \) emissions based on the N sources to the shelf. Continental shelves receive total N (TN) inputs from both terrestrial sources and from the oceans (Fig. 6). The oceanic component (e.g., onwelling, across shelf transport) is considered to be natural as discussed above. Terrestrial N inputs to the shelf include river export (direct discharge of large rivers to shelves as well as export of N from estuaries) and atmospheric deposition directly to shelf waters. The relative magnitude of ter-
For the North Atlantic continental shelf, ocean sources (10–13.5 Tg N y⁻¹) are estimated to contribute about 1.5 times as much N as terrestrial (rivers+atmosphere) sources (8.2–10.6 Tg N y⁻¹) (Fig. 6). As noted above, ocean sources are considered to be natural N. The anthropogenic and natural components of river and atmospheric N inputs to the shelf are estimated as follows. Twenty-five percent of river DIN export to coastal systems globally are estimated to be natural (see above). We use this percentage to estimate the portion of total river N inputs (6.4–8.8 Tg N y⁻¹) to the North Atlantic continental shelf that are natural (1.6–2.2 Tg N y⁻¹) and anthropogenic (4.8–6.6 Tg N y⁻¹) (Fig. 6). Atmospheric N deposition (NOₓ, NH₃, dissolved organic N) also contains both natural and anthropogenic components. Pre-industrial (i.e., natural) and current anthropogenic NOₓ deposition to the global oceans (3.3 and 9 Tg N y⁻¹, respectively) and to continents (4.5 and 22 Tg N y⁻¹, respectively) have been estimated by Galloway et al. (1995), however, the relative contribution of natural and anthropogenic NOₓ deposition to shelves was not estimated. Since continental shelves are located between the terrestrial and the oceanic regions, we assume that the relative amounts of natural (22%) and anthropogenic (78%) NOₓ deposition to the North Atlantic shelf are the average of that to continents (17% natural and 83% anthropogenic) and oceans (27% natural and 73%
We also, somewhat simplistically, assumed the same distribution for NH$_x$. Of the total NO$_y$ and NH$_x$ deposition to the North Atlantic continental shelf (1.8 Tg N y$^{-1}$; Nixon et al., 1996), we estimate that 0.4 Tg N y$^{-1}$ are natural and 1.4 Tg N y$^{-1}$ are anthropogenic (Fig. 6). (No estimate of organic N deposition to continental shelf areas was available.) Considering ocean, atmospheric and river inputs of N to the North Atlantic shelf, and the distribution of natural and anthropogenic components in each, we estimate that about 66% of the N inputs to the shelf are from natural sources and 34% from anthropogenic sources. Following the approach for rivers, estuaries and oceans, we would then proportion the estimated total N$_2$O emissions from this continental shelf region between natural and anthropogenic sources according to the proportion of natural and anthropogenic N inputs.

For shelf areas outside the North Atlantic, no comparable analyses of the relative contributions of the sources (ocean, atmospheric, rivers) exist and they are likely to differ in other regions. The oceanic N contribution to continental shelves outside of the North Atlantic is especially uncertain. For lack of additional information, we have used the North Atlantic scenario to estimate the proportion of the total global N$_2$O emissions from continental shelves (0.6 Tg N y$^{-1}$) that is from natural (66%; 0.4 Tg N y$^{-1}$) and anthropogenic (34%; 0.2 Tg N y$^{-1}$) N sources (Table 2).
Based on the above analyses, anthropogenic emissions dominate the freshwater and coastal marine N$\textsubscript{2}$O emissions with approximately 75% attributed to anthropogenic N$\textsubscript{2}$O emission and 25% to natural (Table 2). However, when ocean emissions are added the pattern reverses; now approximately 26% (1.4 Tg N$\textsubscript{y}^{-1}$) of the total aquatic emissions are attributed to an anthropogenic N source and 74% (4 Tg N$\textsubscript{y}^{-1}$) to a natural N source.

### 3.2. Comparison of aquatic and terrestrial N$\textsubscript{2}$O emissions

**Overall comparison of natural and anthropogenic emissions:** Bouwman et al. (1995) summarized the global source distribution of N$\textsubscript{2}$O emissions from terrestrial sources and from the ocean. We now compare the updated aquatic N$\textsubscript{2}$O emissions to the terrestrial N$\textsubscript{2}$O emissions. Aquatic emissions account for about a third (5.4 Tg N$\textsubscript{y}^{-1}$) and total terrestrial emissions about two-thirds of the total estimated global N$\textsubscript{2}$O emissions ($\sim$15.2 Tg N$\textsubscript{y}^{-1}$) (Fig. 7; Table 1). The total terrestrial emissions include both biogenic (natural soils and agriculture) (9.1 Tg N$\textsubscript{y}^{-1}$) and abiogenic (biofuel, industry, and energy/transport) (0.7 Tg N$\textsubscript{y}^{-1}$) sources.

Natural terrestrial N$\textsubscript{2}$O emissions are from natural soils (6.6 Tg N$\textsubscript{y}^{-1}$). Anthropogenic terrestrial emissions total 3.2 Tg N$\textsubscript{y}^{-1}$ and include a wide range of sources: arable lands (1.0 Tg N$\textsubscript{y}^{-1}$), N excretion from animals (1.0 Tg N$\textsubscript{y}^{-1}$), biomass burning (0.5 Tg N$\textsubscript{y}^{-1}$), agricultural waste burning (0.1 Tg N$\textsubscript{y}^{-1}$), industry (0.5 Tg N$\textsubscript{y}^{-1}$) and combustion processes (0.2 Tg N$\textsubscript{y}^{-1}$) (Bouwman et al., 1995; obtained through EDGAR (Olivier et al., 1996)). Since Bouwman et al. (1995) compiled the gridded inventory, some additional sources have been quantified on a global scale but as yet are not available in gridded inventories. These sources include animal waste management systems, agricultural soil emissions due to crop residues and biological N$\textsubscript{2}$ fixation, and sewage treatment plants. These sources may amount to over 2 Tg N$\textsubscript{y}^{-1}$ at present (Mosier et al., 1998). The industrial and energy sources included in EDGAR are about 1 Tg N$\textsubscript{y}^{-1}$ lower than more recent IPCC estimates by Prather et al. (1995).

Based on the above, approximately 67% (6.6 Tg N$\textsubscript{y}^{-1}$) of total terrestrial N$\textsubscript{2}$O emissions (9.8 Tg N$\textsubscript{y}^{-1}$) are considered natural and 33% (3.2 Tg N$\textsubscript{y}^{-1}$) anthropogenic (Fig. 7). This is similar to the distribution of total aquatic N$\textsubscript{2}$O emission, with approximately 75% considered natural and 25% anthropogenic. Globally, then, about one third of N$\textsubscript{2}$O emissions are anthropogenic (more than 4 Tg N$\textsubscript{y}^{-1}$). This is consistent with the observed increase in atmospheric N$\textsubscript{2}$O which indicates that net additions to the atmosphere amounted to 4–5 Tg N$\textsubscript{y}^{-1}$ in the period 1977–1987 (Khalil and Rasmussen, 1992), although we know that we are missing several sources as discussed above. It should be noted, however, that including the sources for which no gridded inventories are yet available, would increase the relative contribution of anthropogenic sources. It also should be realized that the net additions of nitrous oxide to the atmosphere may be smaller than the total anthropogenic source, since emissions from natural systems may have decreased as a result of human activities (Kroeze et al., 1999).

**Geographical distribution of natural and anthropogenic biogenic N$\textsubscript{2}$O emissions:** The above comparisons were at the global scale. The geographic distribution of aquatic and terrestrial N$\textsubscript{2}$O emissions, including natural and anthropogenic, was obtained by summing the gridded (1°×1°) emissions inventories by latitudinal zones (0°–20°, 20°–45°, 45°–66°, 66°–90°) in each hemisphere. The latitudinal distribution of aquatic N$\textsubscript{2}$O emissions differs from the biogenic terrestrial emissions (Fig. 8A). Overall, the aquatic sources are somewhat more evenly distributed among these latitudinal zones compared to the terrestrial sources, which are concentrated between 20°S and 45°N. Additional patterns emerge from an examination of natural and anthropogenic emissions from aquatic and terrestrial systems. Natural terrestrial emissions dominate in the tropical latitudes (20°S to 20°N) while over half of the anthropogenic terrestrial sources are in the mid-latitudes of the Northern Hemisphere (20° to 66°N) (Fig. 8B and C). Natural aquatic sources are relatively evenly distributed among the latitudinal zones, however anthropogenic aquatic emissions show a definite peak in the mid-latitudes of the Northern Hemispheric (20° to 66°N) as do the anthropogenic terrestrial emissions. The similarity in the latitudinal distribution of anthropogenic terrestrial and aquatic sources is reasonable. Anthropogenic N inputs to the terrestrial environment not only drive anthropogenic N$\textsubscript{2}$O emissions from land, but also are primarily
responsible for anthropogenic N\textsubscript{2}O emissions from aquatic ecosystems. This is because a substantial portion of the anthropogenic N applied to watersheds enters rivers, their estuaries, and continental shelves, resulting in the production of N\textsubscript{2}O (Fig. 1).

Our analysis indicates that most emissions of N\textsubscript{2}O take place in the Northern Hemisphere (63%), with largest contributions from the mid-latitudes; 37% of the global emissions are from the Southern Hemisphere. This is consistent with Prinn et al. (1990) who concluded from analysis of atmospheric trends between 1978 and 1988 that emissions into the 90°–30°N, 30°–0°N, 0°–30°S and 30°–90°S latitudinal belts contribute 22–34%, 32–39%, 20–29%, and 11–15%, respectively, to the global emissions. Bouwman et al. (1995) also estimated, for their case 4b, that 60% of the total emissions are from the Northern Hemisphere and 40% from the Southern. Adding the emissions from rivers, estuaries and continental shelves to the Bouwman et al. inventory slightly improved the comparison to the Prinn et al., mid-point estimate for the North (63%)–South (37%) distribution as reported in Bouwman et al. (1995). Our estimated total global source (15 Tg N y
\textsuperscript{–1}) is higher than that of Prinn et al. (1990) (13 Tg N y
\textsuperscript{–1}), which is also consistent with the fact that Prinn et al. (1990) used a larger atmospheric lifetime for N\textsubscript{2}O (about 170 year) than was used in more recent studies (120 years; Prather et al., 1995). As a result, Prinn et al. (1990) may have underestimated total global emissions.

Fig. 8. Comparison of the latitudinal distribution of biogenic N\textsubscript{2}O emissions from terrestrial and aquatic systems: (A) totals; (B) natural; (C) anthropogenic emissions.
N$_2$O emissions linked to N inputs from terrestrial sources versus N inputs from aquatic sources: A final perspective from which to examine biogenic N$_2$O emissions is to consider whether the N driving the N$_2$O emissions in an ecosystem was originally from land or from aquatic sources. Aquatic N sources include biological N$_2$-fixation in aquatic ecosystems. In the framework of the current analysis, N$_2$O emissions associated with aquatic N sources include all ocean N$_2$O emissions (3.5 Tg N y$^{-1}$) and that portion (55%) of continental shelf emissions that are associated with onwelling or advection of ocean N (0.3 Tg N y$^{-1}$; Fig. 6) (Table 3). N$_2$O emissions attributed to a terrestrial N source include all biogenic N$_2$O from terrestrial ecosystems (2.5 Tg N y$^{-1}$ anthropogenic, 6.6 Tg N y$^{-1}$ natural soils), rivers (1.1 Tg N y$^{-1}$) and estuaries (0.22 Tg N y$^{-1}$) plus that portion of continental shelf emissions associated with atmospheric N deposition and river export (0.3 Tg N y$^{-1}$) (Tables 1–3; Fig. 6). Thus, of the 14.5 Tg of biogenic N$_2$O–N emitted per year, approximately 75% (10.7 Tg N y$^{-1}$) is due to terrestrial N and 25% (3.8 Tg N y$^{-1}$) is from oceanic N sources.

### 4. Concluding remarks

It should be kept in mind that the above analyses are based on N$_2$O emissions inventories currently available in gridded format. As noted above, there are a number of missing sources (Mosier et al., 1998), as well as more recent estimates of abiotic sources (Prather et al., 1995). Emissions from all sources have considerable uncertainty associated with them. As we improve our understanding of factors controlling N$_2$O emissions in ecosystems worldwide, most certainly our estimates will need to be reassessed.

As anthropogenic activities associated with the use and cycling of N in the environment increase in the future, it will be particularly important to understand factors controlling N$_2$O emissions so that effective strategies can be developed to control emissions. The relatively large magnitude of freshwater and coastal marine N$_2$O emissions in combination with their large anthropogenic component indicates the need to obtain better quantification of these sources and of the factors controlling them. This is illustrated by recent estimates of future N$_2$O emissions from rivers, estuaries and continental shelves which indicate that emissions from these sources alone may increase from 2 Tg N y$^{-1}$ in 1990 to 5 Tg N y$^{-1}$ by 2050 due to projected increases in N inputs to these aquatic ecosystems from anthropogenic activities (Kroeze and Seitzinger, 1998).

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### References


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