PERGAMON

Chemosphere – Global Change Science 2 (2000) 267–279

CHEMOSPHERE–
GLOBAL CHANGE
SCIENCE

Global distribution of N_2O emissions from aquatic systems: natural emissions and anthropogenic effects

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Received 9 July 1999; accepted 12 October 1999

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 N_2O emission is from aquatic sources (rivers, estuaries, continental shelves) and the terrestrial sources comprise the remainder. Over 80% of aquatic anthropogenic N_2O 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 N_2O emissions because a substantial portion enters aquatic systems and results in increased N_2O production. Thus, wise management of N in the terrestrial environment could help reduce/control both aquatic and terrestrial N_2O emissions.

Main Abstract: The global distribution of N_2O 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 N_2O emissions; oceanic emissions comprise the remainder. Oceanic N_2O 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. N_2O 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 N_2O emissions from both land and aquatic ecosystems, because a substantial portion of the anthropogenic N applied to watersheds enters rivers, estuaries and continental shelves.

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

1. Introduction

Total global N_2O 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

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PII: S1465-9972(00)00015-5

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_2O 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_2O 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_2O emissions from a number of other sources, including arable land, grasslands, and animal excreta (Bouwman et al., 1995). Much of the N_2O emission from these aquatic systems is anthropogenic and thus contributes to the current increase in atmospheric N_2O. In addition, large increases in future N_2O 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_2O emissions.

In the current paper we compare: (1) N_2O emissions from rivers, estuaries and continental shelves with N_2O emissions from oceanic regions, (2) anthropogenic and natural emissions from aquatic systems, and (3) aquatic and terrestrial N_2O 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°×1°) estimates of N_2O emissions from published studies are used in these analyses.

2. Methods

We calculated N_2O emissions for the following latitudinal zones for the northern and southern hemisphere: 0°–20°, 20°–45°, 45°–66° and 66°–90°.

Emissions from oceans: We obtained the gridded (1°×1°) global N_2O 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°×2.8° grid of Nevison (1994) to a 1°×1° 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°×1°) 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_2O 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_2O emissions from oceans by latitudinal zones then were calculated by summing the N_2O emissions from all gridcells with an average depth >200 m.

Emissions from continental shelves: We used the gridded (1°×1°) global N_2O 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°×1°) global N_2O 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°×1°) global N_2O 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_2O emissions to latitudinal zones; they assigned the N_2O emissions from an entire watershed to the latitude of the river mouth for that watershed.

Terrestrial emissions: We used the gridded (1°×1°) global N_2O 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_2O. 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_2O 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 ($\sim$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 emissions of N_2O during both nitrification and denitrification. The global geographic distribution of dissolved inorganic N (DIN) export by world rivers to coastal marine ecosystems and associated N_2O emissions in rivers and coastal marine ecosystems were estimated in Seitzinger and Kroeze (1998). The detailed methodology is presented in Seitzinger and Kroeze (1998). A brief description of the approach is presented here.

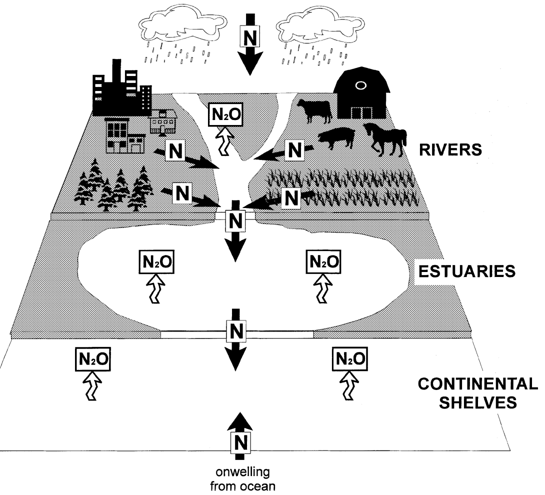


Fig. 1. Schematic of N inputs to aquatic ecosystems. N inputs to rivers and estuaries are linked to N inputs to the surrounding watersheds from both natural and anthropogenic sources. Continental shelves receive N inputs from both terrestrial sources (river/estuarine export and atmospheric deposition) 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.

DIN input to and export by rivers was modeled as a function of N inputs to the watersheds of rivers from synthetic fertilizer use, atmospheric NO*y* deposition, human sewage, and water runoff. (Input and export refer to fluxes.) (N input from the watershed areas below the mouth of a river, for example from cities located on estuaries below river mouths, also was included in the estimated river export.) N_2O emissions in rivers and their estuaries were estimated as a function of denitrification and nitrification using the emission factors described below; denitrification and nitrification were estimated as a function of external N inputs to the rivers or estuaries. Emission factors of N_2O for nitrification and denitrification rates were 0.3% for rivers and estuarine gridcells, except in gridcells with external N inputs exceeding 10 kg N ha^{-1} y^{-1}; for these, an emission factor of 3% was used. These emissions factors are based on a variety of studies which suggest that the ratio of N_2O:N_2 fluxes is generally within the range of 0.1–0.5%, with ratios up to 6% in heavily polluted sediments (e.g., Nishio et al., 1983; Jensen et al., 1984; Seitzinger, 1988).

N_2O emissions in continental shelf regions also were estimated as a function of nitrification and denitrification, however with a lesser amount of detail than for rivers and estuaries (Seitzinger and Kroeze, 1998). N_2O production associated with denitrification was estimated by extrapolating previously estimated denitrification rates for three latitude zones (0°–20°, 20°–45° and 45°–90°) in the North Atlantic (Seitzinger and Giblin, 1996) to shelf areas in those latitudes in other ocean basins. N_2O production associated with pelagic nitrification was estimated by applying the depth average rate of nitrification measured in shelf waters of the western US to the global shelf area. Continental shelves were defined as areas with water depths less than 200 m using the USDOC/NOAA ETOPO-5 bathymetry database (Edwards, 1986; Pilson and Seitzinger, 1996). An emission factor of 0.3% was applied for N_2O for nitrification and denitrification rates in continental shelf regions.

Relative magnitude of all aquatic sources: Model predicted N_2O emissions from global rivers (1.1 Tg N y^{-1}), estuaries (0.2 Tg N y^{-1}) and continental shelves (0.6 Tg N y^{-1}) total 1.9 Tg N_2O-N y^{-1} (Seitzinger and Kroeze, 1998) (Fig. 2). This substantially increases the only other gridded inventory of global N_2O emissions

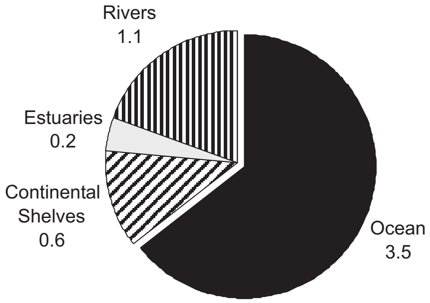


Fig. 2. Comparison of global emissions of N_2O from all aquatic ecosystems (no global emissions estimates for lakes). Units: Tg N y^{-1} (emissions estimates from Seitzinger and Kroeze, 1998 and Nevison et al., 1995).

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_2O 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_2O 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_2O 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_2O 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_2O 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_2O 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_2O 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_2O emissions from aquatic sources (Fig. 3; Table 1). Oceanic N_2O 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_2O 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_2O 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_2O 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_2O 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_2O fluxes were calculated by Nevison et al. (1995) by coupling sea–air gas transfer coefficient fields to a map of surface water N_2O supersaturations obtained from cruise data. In general, the latitudinal distribution of surface N_2O reflects both wind velocities and the amount of mixing between N_2O rich deep water and surface waters (Nevison et al., 1995). An overall view of the latitudinal distribution of ocean N_2O emission per unit area (fluxes) was obtained by combining the ocean area (Fig. 4(A)) with the ocean N_2O emission for each latitudinal zone (Fig. 3). A pattern of decreasing N_2O

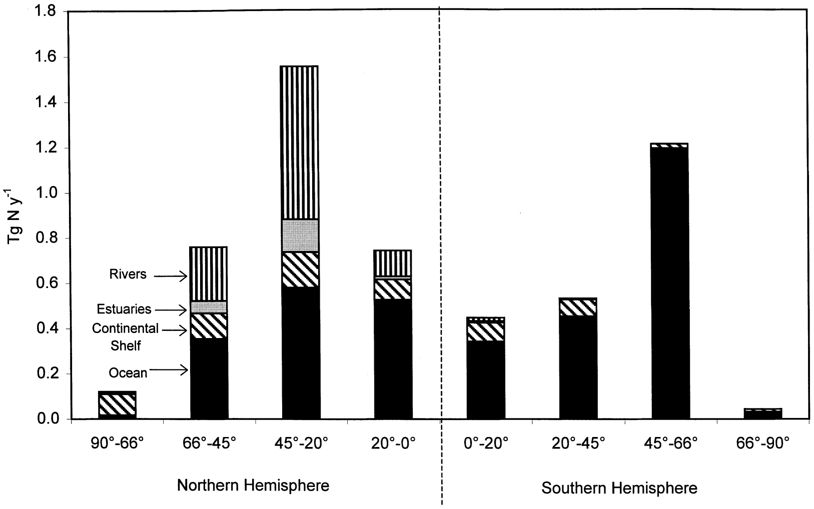


Fig. 3. Comparison of the latitudinal distribution of total N_2O emissions from rivers, estuaries, continental shelves and oceans Units: Tg N y^{-1}.

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_2O 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_2O flux, as defined in the current calculations, does not vary greatly among latitudinal zones. This results in the general decrease in total N_2O emitted from continental shelf regions per latitudinal zone going from north to south (Fig. 3).

The latitudinal distribution of river and estuary N_2O 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_2O emissions: In the current analysis we attribute N_2O 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_2O 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 y^{-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 y^{-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 y^{-1}) of a total of 20.8 Tg y^{-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) y^{-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_2O emissions from rivers are estimated to be 1.1 Tg N y^{-1} (Seitzinger and Kroeze, 1998). The emission factor used by Seitzinger and Kroeze (1998) to relate N_2O emissions to denitrification in rivers varies as a function of N inputs to rivers (see above). Therefore, to estimate natural N_2O emissions from rivers, we assumed a natural watershed DIN yield of 40 kg N (km^{-2} watershed) y^{-1} (Meybeck, 1982) and applied the approach of Seitzinger

Table 1

N_2O emissions summed by latitudinal zones for all sources (Gg N yr^{-1}). Emissions inventories for oceans from Nevison et al. (1995), other aquatic sources from Seitzinger and Kroeze (1998), and for terrestrial sources from Bouwman et al. (1995)

Source	N_2O emissions by latitudinal zones (Gg N y^{-1})								
	90°N–66°N	66°N–45°N	45°N–20°N	20°N–0°	0°–20°S	20°S–45°S	45°S–66°S	66°S–90°S	Total
Aquatic									
Ocean	16.0	354	579	525	342	452	1193	32.2	3493
Cont. shelf	93.6	113	157	90.2	83.3	75.1	19.2	11.5	643
Estuaries	1.29	54.8	145	13.0	7.15	1.39	0.003	0	222
Rivers	8.83	239	671	114	14.3	3.57	0.362	0	1051
Total aquatic	120	761	1552	742	447	532	1212	43.7	5409
Terrestrial									
Abiogenic									
Biofuel	0.003	2.91	31.0	16.7	7.37	2.21	0.012	0	60.3
Energy/transport	0.109	43.2	107	5.30	1.5	5.94	0.026	0	163
Insustrial processes	0.455	229	194	10.7	5.4	16.9	0.029	0	457
Total abiogenic	0.567	275	332	32.7	14.3	25.0	0.067	0	680
Biogenic									
Arable land	0.053	278	560	80.0	25.4	18.6	0.111	0	963
Animal excretion	0.001	193	435	181	88.6	118	6.33	0	1021
Biomass burning	0	0	35.5	222	162	32.7	0	0	452
Agricultural waste burning	0.003	17.8	46.9	18.4	5.34	5.02	0.006	0	93.4
Natural soils	28.1	431	1190	2054	2317	568	6.62	0	6594
Total biogenic	28.2	920	2267	2555	2598	742	13.1	0	9123
Total anthropogenic terrestrial[a]	0.624	764	1409	534	296	199	6.51	0	3210
Total terrestrial	28.7	1195	2599	2588	2613	767	13.1	0	9804
Total terrestrial and aquatic	149	1956	4151	3330	3060	1299	1225	43.7	15213

[a] Total anthropogenic terrestrial includes all terrestrial sources except natural soils.

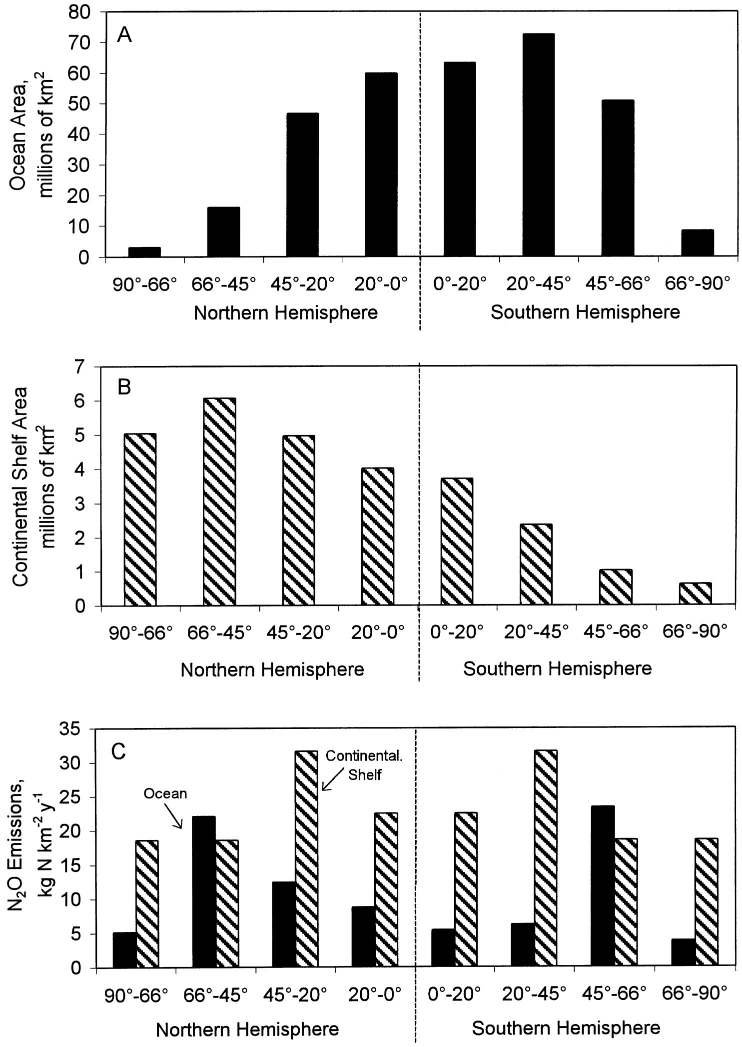


Fig. 4. Comparison of the latitudinal distribution of: (A) ocean area (10^6 km^2); (B) continental shelf area (10^6 km^2); (C) N_2O emissions for oceans and continental shelves per unit area of ocean or shelf (kg N km^{-2} y^{-1}).

and Kroeze (1998). Natural N_2O emissions from rivers are estimated to be approximately 5% (0.05 Tg N y^{-1}) (Table 2). The remaining 95% of global N_2O 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, N_2O 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 N_2O 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 N_2O emissions (0.20 Tg N y^{-1}) were calculated as the difference between the total estuarine N_2O emissions (0.22 Tg N y^{-1}) and natural emissions. Therefore, anthropogenic emissions account for 91% and natural emissions only 9% of the N_2O from estuaries.

Continental shelf N_2O emissions are a combination of anthropogenic and natural. Again, we estimated the relative amount of natural and anthropogenic N_2O 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-

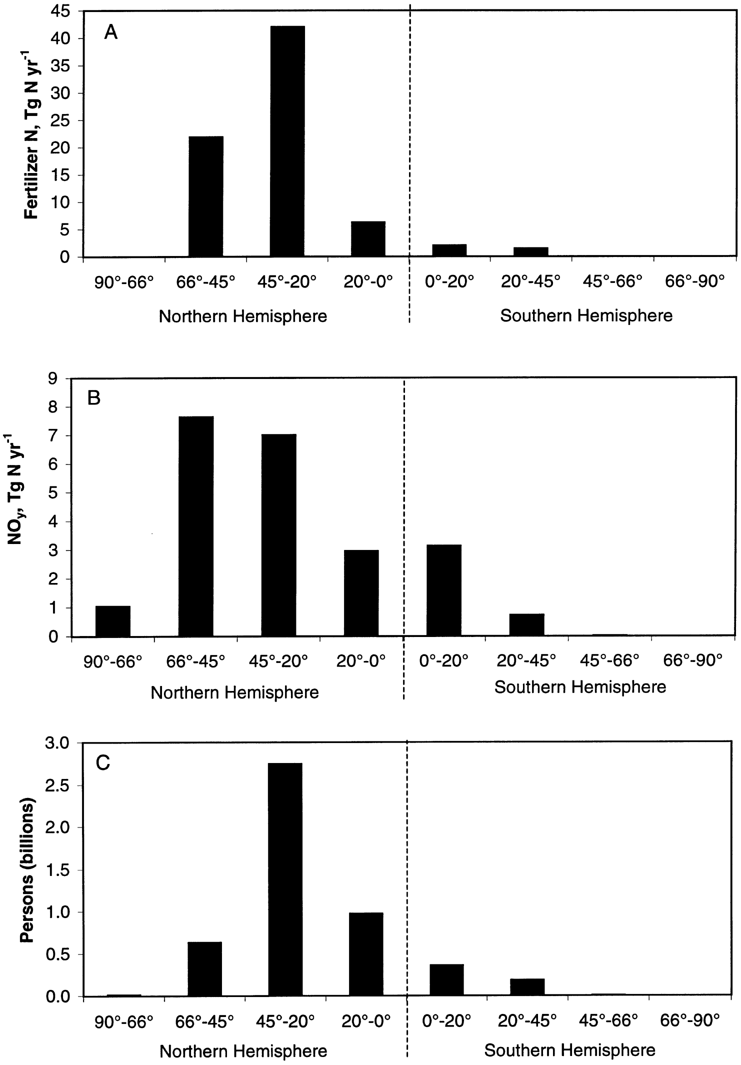


Fig. 5. The latitudinal distribution (for exoreic watershed areas only) of: (A) fertilizer use (Tg N y^{-1}); (B) atmospheric NO*y* deposition (Tg N y^{-1}); (C) human population density (billions).

restrial and ocean N inputs to shelves globally is not well known. However, an analysis was recently done for the North Atlantic (Seitzinger and Giblin, 1996).

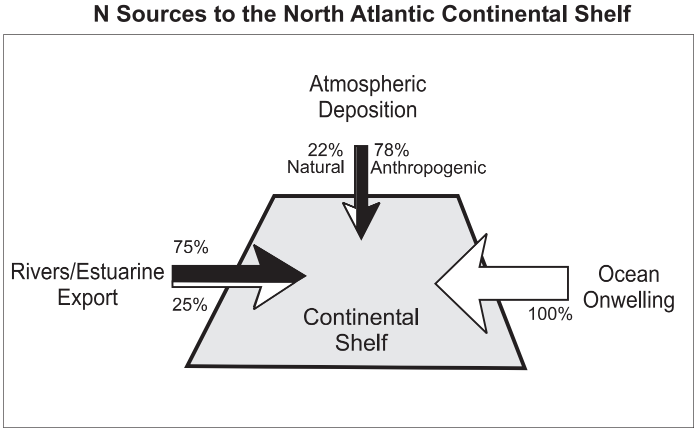
For the North Atlantic continental shelf, ocean sources (10–13.5 Tg N y^{-1}) are estimated to contribute about 1.5 times as much N as terrestrial (rivers+atmosphere) sources (8.2–10.6 Tg N y^{-1}) (Fig. 6). As noted above, ocean sources are considered to be natural N. The anthropogenic and natural components 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 (NO*y*, NH*x*, dissolved organic N) also contains both natural and anthropogenic components. Pre-industrial (i.e., natural) and current anthropogenic NO*y* 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 NO*y* 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%) NO*y* deposition to the North Atlantic shelf are the average of that to continents (17% natural and 83% anthropogenic) and oceans (27% natural and 73%

Table 2
Estimated N_2O emissions (Tg N y^{-1}) from aquatic ecosystems, including distribution of natural and anthropogenic emission (see text for assumptions)

Source	Total N_2O emission	Natural N_2O emission	Anthropogenic N_2O emission
Continental shelves	0.6[a]	0.4 (66%)	0.2 (34%)
Estuaries	0.22[a]	0.02 (9%)	0.20 (91%)
Rivers	1.1[a]	0.05 (5%)	1.05 (95%)
Subtotal	1.9	0.47 (25%)	1.42 (75%)
Oceans	3.5[b]	3.5 (100%)	0 (0%)
Total	5.4	4.0(74%)	1.4 (26%)

[a] Seitzinger and Kroeze (1998).

[b] Nevison et al. (1995) after omitting emissions from continental shelf gridcells.



Sources	TN Inputs (Tg N y^{-1}) Natural	Anthropogenic	Sum
River/Estuarine Export	1.6-2.2	4.8-6.6	6.4-8.8
Atmosphere	0.4	1.4	1.8
Ocean Onwelling	10-13.5	-	10-13.5
Total	12-16.1 66%	6.2-8 34%	**18.2-24.1**

Fig. 6. N inputs to North Atlantic continental shelf from estuarine export plus large river discharge, atmospheric deposition, and ocean inputs (e.g., onwelling, advection) (from Seitzinger and Giblin, 1996). The range of inputs from rivers/estuarine export is due to the range in estimates of N removal by denitrification within estuaries before N is exported to the shelf (Nixon et al., 1996). Ocean onwelling was calculated by difference to balance N losses through burial and denitrification (from Seitzinger and Giblin, 1996).

anthropogenic). 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_2O 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_2O 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_2O emissions with approximately 75% attributed to anthropogenic N_2O emission and 25% to natural (Table 2). However, when ocean emissions are added the pattern reverses; now approximately 26% (1.4 Tg N y^{-1}) of the total aquatic emissions are attributed to an anthropogenic N source and 74% (4 Tg N y^{-1}) to a natural N source.

3.2. Comparison of aquatic and terrestrial N_2O emissions

Overall comparison of natural and anthropogenic emissions: Bouwman et al. (1995) summarized the global source distribution of N_2O emissions from terrestrial sources and from the ocean. We now compare the updated aquatic N_2O emissions to the terrestrial N_2O emissions. Aquatic emissions account for about a third (5.4 Tg N y^{-1}) and total terrestrial emissions about two-thirds of the total estimated global N_2O emissions (~15.2 Tg N y^{-1}) (Fig. 7; Table 1). The total terrestrial emissions include both biogenic (natural soils and agriculture) (9.1 Tg N y^{-1}) and abiogenic (biofuel, industry, and energy/transport) (0.7 Tg N y^{-1}) sources.

Natural terrestrial N_2O emissions are from natural soils (6.6 Tg N y^{-1}). Anthropogenic terrestrial emissions total 3.2 Tg N y^{-1} and include a wide range of sources: arable lands (1.0 Tg y^{-1}), N excretion from animals (1.0 Tg N y^{-1}), biomass burning (0.5 Tg N y^{-1}), agricultural waste burning (0.1 Tg N y^{-1}), industry (0.5 Tg N y^{-1}) and combustion processes (0.2 Tg N 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_2 fixation, and sewage treatment plants. These sources may amount to over 2 Tg N y^{-1} at present (Mosier et al., 1998). The industrial and energy sources included in EDGAR are about 1 Tg N y^{-1} lower than more recent IPCC estimates by Prather et al. (1995).

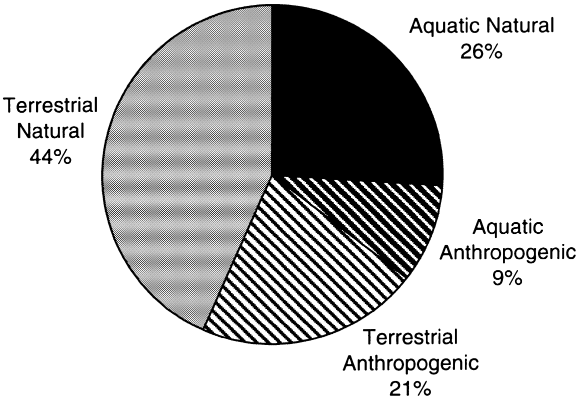


Fig. 7. Comparison of N_2O emissions from aquatic (rivers, estuaries, continental shelves and oceans) and terrestrial systems. The anthropogenic and natural component of each is also indicated.

Based on the above, approximately 67% (6.6 Tg N y^{-1}) of total terrestrial N_2O emissions (9.8 Tg N y^{-1}) are considered natural and 33% (3.2 Tg N y^{-1}) anthropogenic (Fig. 7). This is similar to the distribution of total aquatic N_2O emission, with approximately 75% considered natural and 25% anthropogenic. Globally, then, about one third of N_2O emissions are anthropogenic (more than 4 Tg N y^{-1}). This is consistent with the observed increase in atmospheric N_2O which indicates that net additions to the atmosphere amounted to 4–5Tg N 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_2O emissions: The above comparisons were at the global scale. The geographic distribution of aquatic and terrestrial N_2O 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_2O 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_2O emissions from land, but also are primarily

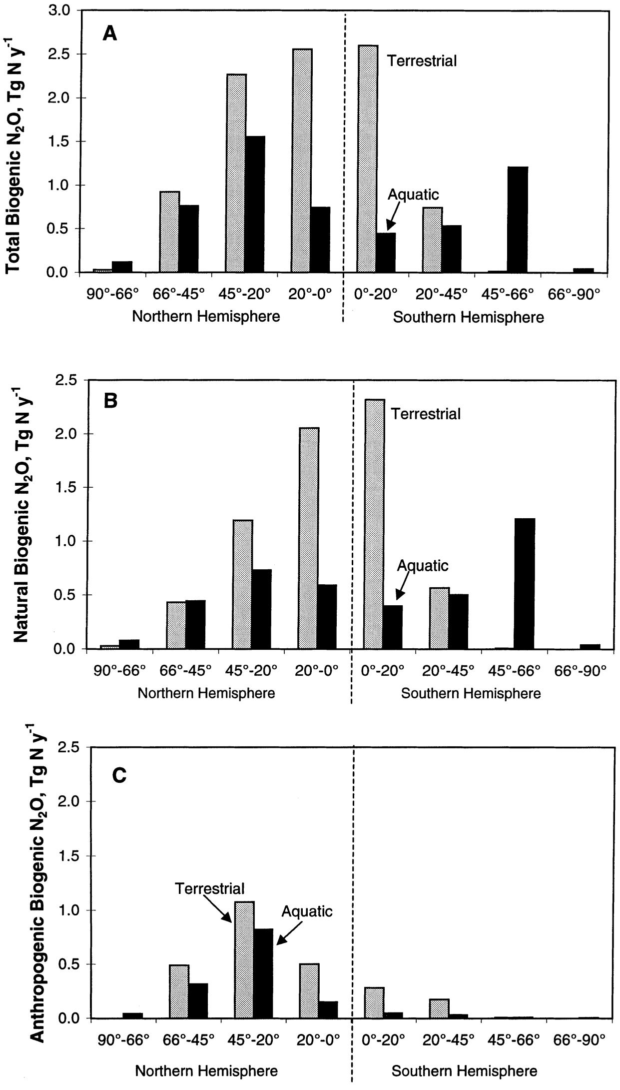


Fig. 8. Comparison of the latitudinal distribution of biogenic N_2O emissions from terrestrial and aquatic systems: (A) totals; (B) natural; (C) anthropogenic emissions.

responsible for anthropogenic N_2O 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_2O (Fig. 1).

Our analysis indicates that most emissions of N_2O 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^{-1}) is higher than that of Prinn et al. (1990) (13 Tg N y^{-1}), which is also consistent with the fact that Prinn et al. (1990) used a larger atmospheric lifetime for N_2O (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.

N_2O emissions linked to N inputs from terrestrial sources versus N inputs from aquatic sources: A final perspective from which to examine biogenic N_2O emissions is to consider whether the N driving the N_2O 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_2O emissions associated with aquatic N sources include all ocean N_2O 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_2O emissions attributed to a terrestrial N source include all biogenic N_2O 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_2O–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.

Table 3
N_2O emissions (Tg N y^{-1}) linked to N inputs from terrestrial sources versus N inputs from aquatic sources[a]

Region	N_2O from aquatic N	N_2O from terrestrial N
Oceans	3.5	
Continental shelves	0.3	0.3
Estuaries		0.2
Rivers		1.1
Terrestrial soils		
Natural and anthropogenic		9.1
Total	3.8	10.7

[a] Inventories from Nevison et al. (1995), Bouwman et al. (1995) and Seitzinger and Kroeze (1998) (see text for details).

4. Concluding remarks

It should be kept in mind that the above analyses are based on N_2O 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_2O 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_2O emissions so that effective strategies can be developed to control emissions. The relatively large magnitude of freshwater and coastal marine N_2O 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_2O 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).

Acknowledgements

We would like to thank L. Bouwman and J. Olivier for assistance with obtaining the terrestrial and ocean data bases from EDGAR. This work was supported in part with funding from NOAA NJ Sea Grant (SPS) (NJSG-99-423), NOAA Coastal Ocean Program (SPS) and Wageningen University (CK).

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