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Sources of Nitrogen to Estuaries in the United States

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ABSTRACT: The purpose of this study was to quantify the nitrogen (N) inputs to 34 estuaries on the Atlantic and Gulf Coasts of the United States. Total nitrogen (TN) inputs ranged from 1 kg N ha^{-1} yr^{-1} for Upper Laguna Madre, Texas, to 49 kg N ha^{-1} yr^{-1} for Massachusetts Bay, Massachusetts. TN inputs to 11 of the 34 estuaries were dominated by urban N sources (point sources and septic systems) and nonpoint source N runoff (5% of total); point sources accounted for 36–86% of the TN inputs to these 11 urban-dominated estuaries. TN inputs to 20 of the 34 estuaries were dominated by agricultural N sources; N fertilization was the dominant source (46% of the total), followed by manure (32% of the total) and N fixation by crops (16% of the total). Atmospheric deposition (runoff from watershed plus direct deposition to the surface of the estuary) was the dominant N source for three estuaries (Barnegat Bay, New Jersey: 64%; St. Catherines-Sapelo, Georgia: 72%; and Barataria Bay, Louisiana: 53%). Six estuaries had atmospheric contributions ≥ 30% of the TN inputs (Casco Bay, Maine: 43%; Buzzards Bay, Massachusetts: 30%; Great Bay, New Jersey: 40%; Chesapeake Bay: 30%; Terrebonne-Timbalier Bay, Louisiana: 59%; and Upper Laguna Madre: 41%). Results from our study suggest that reductions in N loadings to estuaries should be accomplished by implementing watershed specific programs that target the dominant N sources.

Introduction

Nitrogen (N) inputs to estuaries on the Atlantic and Gulf Coasts of the United States are now 2 to 20 times greater than during pre-industrialized times (Boynton et al. 1995; Howarth et al. 1996; Jaworski et al. 1997; Goolsby 2000). Total nitrogen (TN) inputs to the Chesapeake Bay are now 6 to 8 times greater than during pre-colonial times (Boynton et al. 1995). Increased N inputs are of great concern because N often controls primary production in N-limited estuaries (Ryther and Dunstan 1971; Nixon 1986, 1995; Fisher and Oppenheimer 1991; D'Elia et al. 1992; Howarth et al. 2000). Chronic N additions to N-limited estuaries can accelerate primary production and eutrophication, leading to many undesirable responses, such as increased frequency of harmful algal blooms, hypoxic (< 4 mg l^{-1}) and anoxic bottom waters, loss of emergent plants, and reduced fish stocks (Valiela and Costa 1988; Paerl 1988, 1995, 1997; Valiela et al. 1990; Hallengraeff 1993; Boynton et al. 1995). To manage these adverse water quality problems we must identify the N sources and implement cost-effective controls.

Developing a plan to manage N inputs to estuaries is difficult because N originates from many different sources. Point sources of N include discharge from sewage treatment plants and industrial facilities. N also enters estuaries as runoff from forests, agricultural lands, and urban areas. Atmospheric N deposition is another sources of N that originates from the emissions of N oxides from automobiles, urban and industrial sources, and ammonia emissions from agricultural sources. An effective management plan to control N inputs to estuaries must start with an assessment of these diverse N sources. The purpose of our study was to quantify the sources of N to 34 estuaries on the Atlantic and Gulf Coasts of the U.S. Results from our assessment can be used to develop cost-effective management strategies to reduce N inputs to estuaries in the U.S.

Methods

Characteristics of the Watershed-Estuary Systems

We studied 34 watershed-estuary systems on the Atlantic and Gulf Coasts of the U.S. (Castro et al.

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2000). Human population densities in the study watersheds ranged from 0.3 to 8.1 persons ha^{-1}. Highest population densities were in watersheds in the northeastern U.S. Upland forests accounted for 45–77% of the total watershed area for watersheds in the northeast, except for Massachusetts Bay, Massachusetts (22% of watershed area). Urban areas were important in the northeast, accounting for 16–75% of the total watershed area. Watersheds in the Mid-Atlantic and southeast regions were dominated by upland forests (42–63%) and agricultural lands (24–37%); except for the St. Catherines-Sapelo, Georgia, and Indian River, Florida, watersheds. St. Catherines-Sapelo contained virtually no agriculture (1%), but was dominated by upland forests (63%) and wetlands (30%). The Indian River watershed was dominated by urban areas (37%) and agriculture (27%). Watersheds on the eastern Gulf Coast were dominated by rangelands and agriculture (Charlotte Harbor and Tampa Bay, Florida), agriculture and upland forests (Apalachee Bay and Apalachicola Bay, Florida; Mobile Bay, Alabama; and West Mississippi Sound, Louisiana), or wetlands (Barataria Bay and Terrebonne-Timbalier, Louisiana). Watersheds on the western Gulf Coast were dominated by either upland forests and agriculture (Calcasieu Lake, Louisiana; Sabine Lake, Texas) or rangeland and agriculture (Galveston Bay, Matagorda Bay, Corpus Christi Bay, and Upper and Lower Laguna Madre, Texas).

N Inputs to Watersheds and Estuaries

N is essential for life, but most of the N in the biosphere is unreactive and unavailable to organisms. Unreactive N can be converted into reactive N by natural processes and human activities. We estimated the amount of reactive N that was either used or produced by human activities in our study watersheds. Reactive N associated with human activities included the application of N fertilizers to crops and lawns, biotic N_2 fixation by leguminous crops and pastures, the net import of N in food for human consumption, the net import of N in feed for livestock, and atmospheric deposition of inorganic N (NO_3^- and NH_4^+). Details of these calculations are described in Castro et al. (2000), but the results presented here are different because we considered atmospheric deposition of NH_4^+ in this study.

We used land-use specific approaches similar to those described in Castro et al. (2000) to estimate N export from watersheds to our study estuaries. For this study, we improved our original approach and estimated the amount of N available for transport to estuaries from agricultural lands, urban areas, and upland forests (Fig. 1). These three land uses account for 69–99% of the total land area in our 34 study watersheds. Our revised approach includes the following modifications that were not used in our previous study (Castro et al. 2000): better estimates of denitrification losses from agricultural lands, N deposition to surface waters in the watersheds, assumption that forests export organic N, watershed-specific N excretion rates from the human population to estimate septic system N losses, different retention estimates of N during riverine transport, and a different approach to estimate the amount of N retained and stored in the landscape. Here we describe the modifications to our original approach.

Denitrification in Agricultural Lands

We adjusted denitrification rates in agricultural areas to reflect temperature influences by use of mean watershed temperatures (Castro et al. 2000) and a denitrification activity Q_{10} value of 2 (Stanford et al. 1975; Maag et al. 1997); the Mid-Atlantic region served as the reference for temperature adjustment.

Atmospheric N Deposition to Surface Waters in the Watershed

To estimate TN deposition to surface waters (streams, canals, lakes, and reservoirs) in the watershed, we multiplied the wet and dry deposition rates of NO_3^- and NH_4^+ by the total surface water area in the watershed (Pacheco 1999). Wet deposition rates of NO_3^- and NH_4^+ were obtained from the National Atmospheric Deposition program. Dry deposition rates of NO_3^- and NH_4^+ were estimated using data from the Clean Air Status and Trends Network and the Regional Atmospheric Deposition Model (RADM). Details of our atmospheric N deposition rates can be found in Meyers et al. (2000).

N Export from Upland Forests

We assumed that dissolved organic nitrogen (DON) is exported from upland forests. The contribution made by DON to the TN export from forests in our study watersheds is not known. We assumed that the DON contribution to the TN loads was equal to 50% of the inorganic N load exported from forests. We estimated inorganic N exported from upland forests using a non-linear regression relationship between wet deposition of NH_4^+ and NO_3^- and stream water N export of dissolved inorganic N (Castro et al. 2000).

Per Capita N Excretion Rates

To estimate per capita N excretion rates for septic systems, we divided the total amount of point source N (organic and inorganic forms) released

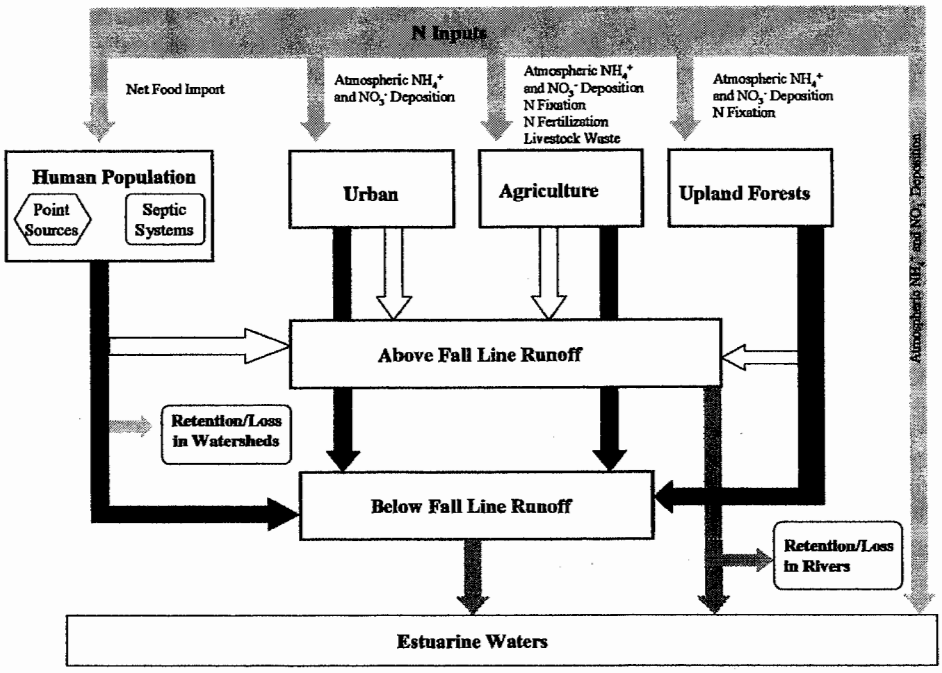

Fig. 1. Schematic diagram of the approach used to estimate the contribution made by different nitrogen sources to the total nitrogen inputs to our study estuaries. Grey colored arrows show the nitrogen inputs to the watershed and estuary and nitrogen losses and retention in the watershed, streams and rivers. The open arrows show the nitrogen inputs to the watershed above the fall line. The black arrows show the nitrogen inputs to the watershed below the fall line. Patterned arrows show nitrogen inputs to the estuary from the different portions of the watershed.

from wastewater treatment plants by the human population on sewers that contributed to these point source N (Pacheco 1999).

Watershed and Riverine N Retention and Losses

To validate our approach, we compared our estimates to independent riverine N fluxes measured at the fall line of 18 watersheds using data from the U.S. Geological Survey (USGS) National Stream Quality Accounting Network (NASQAN) program (Pacheco 1999). Our predicted fluxes were generally higher than N fluxes measured at the fall line (Fig. 2). This discrepancy was expected because our approach does not account for riverine and watershed N sinks.

To improve our estimates, we accounted for watershed and riverine sinks of N. The amount of N removed during transport in the watershed is likely to differ depending on the source of N. We examined the implications of assuming various rates of removal of N from different N sources. We assumed that point sources of N and runoff from upland forests were not attenuated during transport in the watershed. Point sources of N that were discharged from wastewater treatment plants into rivers and estuaries bypass all of the natural watershed retention processes. We accounted for N retention and losses in urban areas. Some of the N available to be exported from agricultural land and septic systems is likely to be lost by watershed removal or storage processes and needed to be accounted for in our analysis. As a result, we varied our watershed removal rates (20%, 30%, 40%, 50%, 60%, and 80%) of the N available to be exported from agricultural lands and septic systems. We found for some watersheds that the removal of more than 40% of the excess N from agricultural lands and septic systems implied little or no N loss in rivers, which is unrealistic (Table 1). We assumed that 40% was a reasonable approximation of the amount of available N from agriculture lands and septic systems that was retained or lost during drainage water transport in the watershed. We also accounted for N losses during riverine transport to the estuary. We assumed that N is retained or lost during transport to the fall line. We adjusted our river N loss rates until predicted N fluxes matched the N fluxes measured at the fall line (Fig. 2). Our estimated riverine N loss rates (Table 1) were similar to those reported in other studies (Howarth et al. 1996; Alexander et al. 2000; Castro et al. 2000). We assumed no N losses during riverine transport below the fall line because of the short travel times to the estuaries. This is consistent with the observation that small streams have much

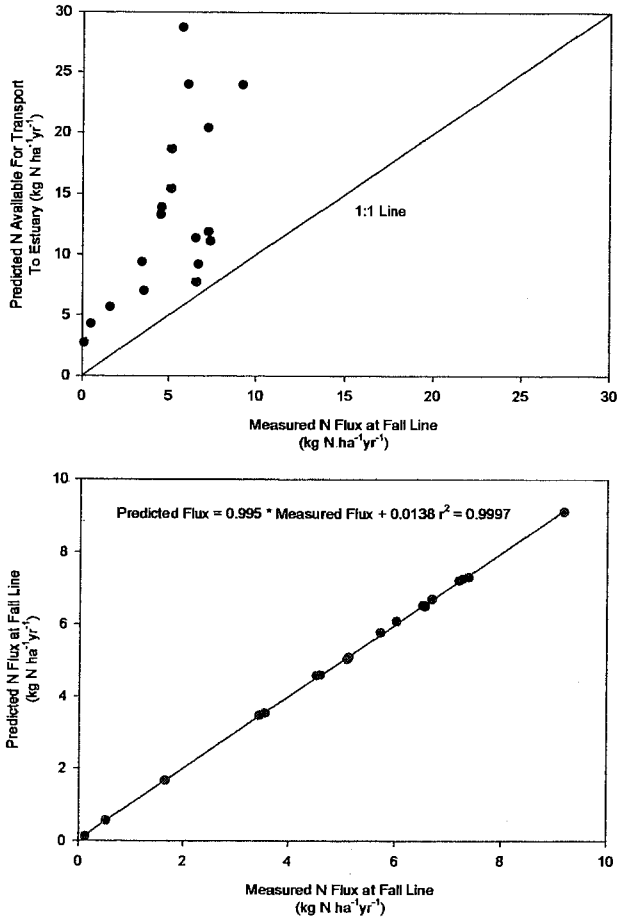

Fig. 2. Comparison of nitrogen fluxes measured at the watershed fall line and nitrogen available for transport from the watershed to the fall line before and after accounting for watershed and riverine nitrogen retention and losses.

higher N loss rates (0.45 versus 0.005 kg N d^{-1}) than larger river systems (Alexander et al. 2000b)

Results and Discussion

Anthropogenic N Inputs to the Watershed

There were important differences in the anthropogenic N sources across our study regions (Table 2). Net food import of N was greatest in the Northeastern U.S.; it was 5–24 times greater than net food import of N in all other regions (Table 2). The net import of N in food accounted for 46% of the total anthropogenic N inputs to watersheds in the Northeastern U.S. On average, the net import of N in feed was lowest in watersheds in the Northeast and the western Gulf Coast. The Mid-Atlantic region had the greatest net import of N in feed. N fixation by crops and pastures was lowest in the northeast and greatest in the Mid-Atlantic region, followed by the Southeast and eastern Gulf Coast. N fertilization was lowest in the Northeast and was approximately 3 times greater in all other regions compared to the northeast. The average contribution made by atmospheric N deposition was greatest in the Northeast and Mid-Atlantic regions and lowest in the Southeast and western Gulf Coast (Table 2).

At the watershed scale, the net import of N in food ranged from −1.0 kg N ha^{-1} yr^{-1} (negative values denote net export from the watershed) for the Charlotte Harbor watershed to 50.1 kg N ha^{-1} yr^{-1} for Massachusetts Bay (Table 2). The mean was 6.2 kg N ha^{-1} yr^{-1}. On average net food import was 19% of the total anthropogenic N inputs for the 34 watersheds. In several of the watersheds, particularly in the Northeast, net food import was a major (> 30%) component of total anthropogenic N inputs. This regional pattern reflects the need to import food to support the large human populations in the Northeastern U.S.

Average net import of N in feed was 2.9 kg N ha^{-1} yr^{-1}, with values ranging from −5.1 kg N ha^{-1} yr^{-1} for Lower Laguna Madre, to 14.8 kg N ha^{-1} yr^{-1} for Tampa Bay. On average, the net import of N in feed was 9% of the total anthropogenic N input for all watersheds. Net feed import was a major source (> 25%) of anthropogenic N for Chesapeake Bay, Charleston Harbor, South Carolina; Altamaha River, Georgia, Mobile Bay, West Mississippi Sound, and Sabine Lake. High net feed import to these watersheds, particularly the Chesapeake Bay watershed, was needed to support the large populations of cows, hogs, and poultry.

Rates of N fixation ranged from 0.3 kg N ha^{-1} yr^{-1} for St. Catherines-Sapelo to 10 kg N ha^{-1} yr^{-1} for Pamlico-Pungo Sound, North Carolina, with a mean value of 3.7 kg N ha^{-1} yr^{-1} for all watersheds. On average, N fixation was 12% of the total anthropogenic N input and was a significant fraction (> 20%) of the total anthropogenic N flux for Pamlico-Pungo Sound (23%), St. Helena Sound, South Carolina (24%), Appalachee Bay (21%), Apalachicola Bay (25%), and Calcasieu Lake (27%). High biotic N_2 fixation in the Pamlico-Pungo Sound watershed was due to biotic N_2 fixation by soybeans, which was the dominant N_2 fixing crop in this system.

N fertilization rates ranged from 0.5 kg N ha^{-1} yr^{-1} for St. Catherines-Sapelo to 48.2 kg N ha^{-1} yr^{-1} for Indian River, with an average of 12.0 kg N ha^{-1} yr^{-1}. N fertilization accounted for 38% (averaged over all watersheds) of total anthropogenic N inputs to the 34 watersheds. For 18 of 34 watershed-estuary systems, N fertilization was the largest source of total anthropogenic N inputs. N fertilization rates were very high in several Florida watersheds (Indian River: 48.2 kg N ha^{-1} yr^{-1}; Charlotte

TABLE 1. Percent of the N retained or lost in the river above the fall line with different amounts of watershed N retention of the excess N from agricultural lands and septic systems. Sites without values do not have fall lines.

Watershed Location	20%	40%	50%	60%	80%
Northeast					
Casco Bay, Maine					
Great Bay, New Hampshire					
Merrimack River, Massachusetts	12	8	4	1	0
Massachusetts Bay, Massachusetts					
Buzzards Bay, Massachusetts					
Narragansett Bay, Rhode Island	61	59	58	56	54
Long Island Sound, Connecticut	69	65	64	61	56
Hudson River-Raritan Bay, New York	44	35	29	22	5
Barnegat Bay, New Jersey					
Great Bay, New Jersey					
Mid-Atlantic					
Delaware Bay, Delaware	20	9	3	0	0
Chesapeake Bay	22	5	0	0	0
Pamlico-Pungo Sound, North Carolina	76	69	65	58	32
Southeast					
Wynah Bay, South Carolina	69.5	61	55	47	15
Charleston Harbor, South Carolina					
St. Helena Sound, South Carolina	55	40	29	13	0
St. Catherines-Sapelo, Georgia					
Altamaha River, Georgia	61	50	43	33	0
Indian River, Florida					
Eastern Gulf Coast					
Charlotte Harbor, Florida					
Tampa Bay, Florida					
Apalachee Bay, Florida	56.1	42	31	14	0
Apalachicola Bay, Florida	61	51	43	33	0
Mobile Bay, Alabama	32	17	5	0	0
West Mississippi Sound, Louisiana					
Barataria Bay, Louisiana					
Terrebonne-Timbalier Bays, Louisiana					
Western Gulf Coast					
Calcasieu Lake, Louisiana	27	9	0	0	0
Sabine Lake, Texas	60	49	42	31.5	0
Galveston Bay, Texas	69	63	59	53.8	39
Matagorda Bay, Texas	84	81	79	76	60
Corpus Christi Bay, Texas	94	93	92	90	83
Upper Laguna Madre, Texas					
Lower Laguna Madre, Texas					

Harbor: 35.8 kg N ha^{-1} yr^{-1}; Tampa Bay: 43 kg N ha^{-1} yr^{-1}) and for the Upper and Lower Laguna Madre (36.9 and 29.6 kg N ha^{-1} yr^{-1}, respectively). High N fertilization rates in Florida and Texas reflect the longer growing seasons with multiple crops in the same year.

Total (wet plus dry) atmospheric N deposition rates ranged from 4.1 kg N ha^{-1} yr^{-1} in Corpus Christi Bay to 11.7 kg N ha^{-1} yr^{-1} at Barataria Bay, with an average value of 4.8 kg N ha^{-1} yr^{-1}. With the exception of the northernmost watersheds in Maine (Casco Bay: 4.5 kg N ha^{-1} yr^{-1}) and New Hampshire (Great Bay: 5.6 kg N ha^{-1} yr^{-1}), atmospheric N deposition was fairly constant across the Northeast and northern Mid-Atlantic regions, ranging from 7 to 10 kg N ha^{-1} yr^{-1}. In the Southeast, atmospheric N inputs decreased to between 4.9 and 6.6 kg N ha^{-1} yr^{-1}. On the Gulf Coast, atmospheric N deposition rates ranged from 4.7 to 11.7 kg N ha^{-1} yr^{-1} and 4.1 to 8.9 kg N ha^{-1} yr^{-1} for watersheds on the eastern and western Gulf Coast, respectively. The contribution made by atmospheric N deposition to the total anthropogenic N inputs ranged from 7% for Tampa Bay to 71% at St. Catherines-Sapelo Island and the average for all 34 watersheds was 22%.

Total NH_4^+ deposition rates ranged from 1.3 kg N ha^{-1} yr^{-1} for the Indian River watershed to 3.3 kg N ha^{-1} yr^{-1} for the West Mississippi Sound and Barataria Bay watersheds (Meyers et al. 2000). As expected, regional patterns were evident, with relatively low deposition (< 1.6 kg N ha^{-1} yr^{-1}) in northern New England (Casco Bay and Great Bay). Atmospheric NH_4^+ deposition generally increased

TABLE 2. Net anthropogenic N inputs (kg N ha^{-1} yr^{-1}) to each watershed and the percentage (in parentheses) of the total net anthropogenic N inputs to each watershed from different anthropogenic sources. Negative food and net feed import values indicate that N has been exported out of the watershed.

Watershed Location	Net Food Import	Net Feed Import	Nitrogen Fixation	Fertilization	Atmospheric N Deposition	Total
Northeast						
Casco Bay, Maine	4.4 (32)	2.2 (16)	0.9 (7)	1.6 (12)	4.5 (33)	13.7
Great Bay, New Hampshire	5.2 (32)	3.3 (21)	0.8 (5)	1.1 (7)	5.6 (35)	15.9
Merrimack River, Massachusetts	9.3 (45)	1.5 (7)	0.9 (4)	1.5 (7)	7.6 (37)	20.8
Massachusetts Bay, Massachusetts	50.1 (79)	1.0 (2)	0.8 (1)	3.9 (6)	7.9 (12)	63.7
Buzzards Bay, Massachusetts	18.9 (46)	2.7 (7)	1.4 (4)	9.9 (24)	7.9 (19)	40.9
Narragansett Bay, Rhode Island	21.7 (54)	3.1 (8)	1.2 (3)	5.5 (14)	8.8 (22)	40.3
Long Island Sound, Connecticut	9.3 (37)	4.0 (15)	1.5 (5)	3.4 (13)	9.1 (33)	27.4
Hudson River-Raritan Bay, New York	20.2 (48)	2.5 (6)	4.4 (10)	4.7 (11)	10.0 (24)	41.8
Barnegat Bay, New Jersey	17.3 (51)	−0.4 (−1)	2.1 (6)	4.5 (13)	10.3 (30)	33.8
Great Bay, New Jersey	13.1 (37)	−0.9 (−2)	3.4 (10)	9.2 (26)	10.3 (29)	35.1
Regional Average	16.9 (46)	1.9 (9)	1.7 (5.5)	4.5 (13)	8.2 (27)	33.3
Mid-Atlantic						
Delaware Bay, Delaware	9.8 (22)	4.0 (9)	7.5 (17)	11.6 (27)	10.5 (24)	43.5
Chesapeake Bay	0.3 (1)	10.4 (28)	7.1 (19)	8.9 (24)	7.3 (28)	34.1
Pamlico-Pungo Sound, North Carolina	0.1 (0.1)	2.6 (6)	10.0 (23)	23.4 (54)	7.0 (16)	43.2
Regional Average	3.4 (8)	5.7 (14)	8.2 (20)	14.6 (35)	8.3 (23)	40.3
Southeast						
Wynah Bay, South Carolina	−0.3 (−1)	6.9 (20)	6.9 (20)	14.4 (42)	6.6 (19)	34.6
Charleston Harbor, South Carolina	2.3 (11)	5.3 (25)	2.5 (12)	5.8 (27)	5.4 (25)	21.2
St. Helena Sound, South Carolina	0.4 (2)	−0.5 (−2)	4.8 (24)	9.3 (47)	4.3 (29)	18.4
St. Catherines-Sapelo, Georgia	1.3 (15)	0.3 (4)	0.3 (4)	0.5 (6)	6.0 (71)	5.8
Altamaha River, Georgia	0.0 (0)	7.8 (27)	4.4 (15)	9.8 (35)	6.3 (22)	28.3
Indian River, Florida	1.9 (3)	1.9 (3)	5.4 (9)	48.2 (77)	4.9 (8)	62.5
Regional Average	0.9 (5)	3.6 (13)	4.0 (14)	14.7 (39)	5.6 (29)	28.5
Eastern Gulf Coast						
Charlotte Harbor, Florida	−1.0 (−2)	5.5 (10)	7.4 (14)	35.8 (68)	5.3 (10)	53.0
Tampa Bay, Florida	10.0 (13)	14.8 (19)	5.7 (7)	43.0 (54)	5.8 (7)	79.6
Apalachee Bay, Florida	−0.1 (−1)	−0.5 (−3)	4.2 (21)	11.8 (59)	4.7 (23)	19.9
Apalachicola Bay, Florida	−0.6 (−2)	3.2 (11)	7.6 (25)	14.7 (48)	5.5 (18)	30.4
Mobile Bay, Alabama	0.1 (0)	7.1 (30)	3.8 (16)	6.3 (26)	6.7 (28)	24.1
West Mississippi Sound, Mississippi	0.8 (3)	9.3 (32)	2.9 (10)	7.8 (27)	8.6 (29)	29.3
Barataria Bay, Louisiana	8.4 (30)	0.3 (1)	1.6 (6)	5.7 (21)	11.7 (42)	27.7
Terrebonne-Timbalier Bays, Louisiana	2.1 (12)	0.4 (2)	0.8 (4)	3.6 (20)	10.9 (61)	17.8
Regional Average	2.5 (7)	5.0 (13)	4.2 (13)	16.1 (40)	7.4 (27)	35.2
Western Gulf Coast						
Calcasieu Lake, Louisiana	−0.3 (−1)	−0.8 (−4)	6.4 (27)	9.8 (41)	8.9 (37)	23.9
Sabine Lake, Texas	−0.7 (−2)	8.2 (30)	3.9 (14)	8.7 (32)	7.3 (27)	27.4
Galveston Bay, Texas	5.8 (16)	2.6 (7)	7.1 (20)	12.3 (35)	7.6 (21)	35.4
Matagorda Bay, Texas	−0.5 (−3)	0.2 (1)	2.3 (14)	9.8 (60)	4.6 (28)	16.4
Corpus Christi Bay, Texas	−0.4 (−4)	−0.1 (−1)	1.6 (13)	7.5 (59)	4.1 (32)	12.7
Upper Laguna Madre, Texas	0.5 (1)	−2.9 (−7)	2.4 (6)	36.9 (89)	4.4 (11)	41.3
Lower Laguna Madre, Texas	0.3 (1)	−5.1 (−17)	1.9 (6)	29.7 (92)	4.2 (13)	31.0
Regional Average	0.7 (1)	0.3 (2)	3.7 (14)	16.4 (59)	5.9 (24)	26.9

southward through southern New England and the Mid-Atlantic region, peaking at the Chesapeake Bay watershed (3.2 kg N ha^{-1} yr^{-1}). Deposition rates decreased in the southeast (2.0 kg N ha^{-1} yr^{-1}), with relatively low rates (< 1.7 kg N ha^{-1} yr^{-1}) for the Florida watersheds. Ammonium deposition increased along the Gulf Coast reaching a maximum (3.3 kg N ha^{-1} yr^{-1}) in Louisiana (West Mississippi Sound, Barataria Bay) and decreasing along the western Gulf Coast (< 2.0 kg N ha^{-1} yr^{-1}).

Source of the N Inputs

A goal of this analysis was to quantify the major sources of N to our study estuaries. These inputs included: four sources of N from agricultural lands (atmospheric N deposition, fertilizer N, manure N, N fixation), three sources of N derived from urban areas (direct point source discharge, leachate from septic fields, N runoff from urban lands), N exported from forests derived from atmospheric N deposition, and direct deposition of atmospheric N to the surface of the estuary. To facilitate our

TABLE 3. Nitrogen exported (kg N ha^{-1} yr^{-1}) to the estuaries from different watershed N sources and the percentage of N exported from different sources (in parentheses). Watershed types were based on the dominant sources of N to the estuary. Watersheds dominated by urban N sources (point, septic, and Nonpoint Source runoff) were classified as urban, watersheds dominated by agricultural N sources (fertilization, fixation, and manure) were classified as agriculture, and watersheds dominated by atmospheric N deposition were classified as atmospheric. Effluent from sewage treatment plants in the Barnegat Bay watershed is discharged offshore; sewage inputs to the Barnegat Bay estuary are from septic systems in the watershed.

Watershed-Estuary System	Watershed Type	Agriculture Runoff	Urban Nonpoint Source	Upland Forest Runoff	Human Sewage	Atmospheric Deposition	Total
Casco Bay, Maine	Urban	0.7 (12.6)	0.8 (15.0)	0.3 (5.5)	1.9 (36.3)	1.6 (30.6)	5.3
Great Bay, New Hampshire	Urban	1.3 (19.2)	1.4 (19.9)	0.3 (4.6)	2.5 (36.4)	1.4 (19.8)	6.8
Merrimack River, Massachusetts	Urban	0.8 (8.5)	0.5 (5.1)	0.6 (6.5)	5.7 (59.7)	1.9 (20.2)	9.5
Massachusetts Bay, Massachusetts	Urban	1.8 (3.7)	0.1 (0.3)	0.1 (0.2)	41.9 (85.5)	5.0 (10.3)	49.0
Buzzards Bay, Massachusetts	Urban	5.2 (23.8)	0.3 (1.2)	0.3 (1.5)	12.3 (56.4)	3.7 (17.1)	21.8
Narragansett Bay, Rhode Island	Urban	3.0 (11.1)	0.7 (2.7)	0.4 (1.5)	19.3 (70.7)	3.8 (13.9)	27.2
Long Island Sound, Connecticut	Urban	2.0 (15.4)	0.5 (4.1)	0.9 (6.8)	7.3 (56.7)	2.2 (17.0)	12.9
Hudson River-Raritan Bay, New York	Urban	2.8 (11.5)	0.8 (3.4)	0.8 (3.1)	15.7 (65.5)	3.9 (16.4)	24.0
Delaware Bay, Delaware	Urban	6.4 (31.9)	0.4 (2.1)	0.7 (3.6)	8.8 (43.8)	3.7 (18.5)	20.2
Charleston Harbor, South Carolina	Urban	3.9 (28.6)	0.2 (1.5)	0.2 (1.4)	8.4 (62.2)	0.9 (6.4)	13.5
Terrebonne-Timbalier Bays, Louisiana	Urban	1.4 (13.2)	0.2 (1.6)	0.002 (0.02)	5.1 (47.9)	3.9 (37.3)	10.6
Average		2.7 (16)	0.5 (5)	0.4 (3)	11.7 (57)	2.9 (19)	18.3
Great Bay, New Jersey	Agriculture	4.7 (47.4)	0.1 (1.2)	0.7 (6.8)	1.4 (14.1)	3.0 (30.5)	9.9
Chesapeake Bay	Agriculture	7.2 (53.4)	0.2 (1.5)	1.0 (7.5)	2.1 (15.3)	3.0 (22.3)	13.5
Pamlico-Pungo Sound, North Carolina	Agriculture	13.5 (74.1)	0.1 (0.3)	0.3 (1.7)	2.6 (14.4)	1.8 (9.6)	18.2
Wynah Bay, South Carolina	Agriculture	8.9 (70.2)	0.1 (0.6)	0.3 (2.1)	2.2 (17.5)	1.2 (9.5)	12.7
St. Helena Sound, South Carolina	Agriculture	4.7 (81.8)	0.0 (0.2)	0.1 (2.4)	0.1 (1.2)	0.8 (14.5)	5.7
Altamaha River, Georgia	Agriculture	6.6 (70.6)	0.1 (0.6)	0.3 (2.8)	1.6 (16.7)	0.9 (9.3)	9.4
Indian River, Florida	Agriculture	21.3 (73.1)	0.3 (1.1)	0.01 (0.05)	4.1 (13.9)	3.4 (11.8)	29.1
Charlotte Harbor, Florida	Agriculture	15.4 (85.2)	0.03 (0.2)	0.02 (0.12)	1.1 (6.2)	1.5 (8.4)	18.1
Tampa Bay, Florida	Agriculture	21.1 (78.3)	0.2 (0.7)	0.02 (0.07)	2.7 (9.9)	3.0 (11.0)	26.9
Apalachee Bay, Florida	Agriculture	4.5 (81.2)	0.1 (0.9)	0.2 (3.5)	0.2 (3.5)	0.6 (10.8)	5.6
Apalachicola Bay, Florida	Agriculture	7.2 (72.1)	0.1 (1.3)	0.2 (2.2)	1.6 (16.3)	0.8 (8.0)	10.0
Mobile Bay, Alabama	Agriculture	4.6 (54.3)	0.1 (1.3)	0.4 (4.7)	2.3 (26.9)	1.1 (12.8)	8.5
West Mississippi Sound, Mississippi	Agriculture	5.7 (63.1)	0.1 (1.3)	0.6 (6.8)	1.6 (18.1)	1.0 (10.7)	9.1
Calcasieu Lake, Louisiana	Agriculture	5.9 (50.7)	0.1 (0.55)	0.5 (4.5)	2.6 (22.3)	2.6 (22.0)	11.7
Sabine Lake, Texas	Agriculture	6.1 (66.3)	0.1 (0.79)	0.3 (3.8)	1.6 (17.3)	1.1 (11.9)	9.3
Galveston Bay, Texas	Agriculture	7.8 (47.2)	0.2 (1.36)	0.1 (0.8)	6.7 (40.4)	1.7 (10.2)	16.5
Matagorda Bay, Texas	Agriculture	3.0 (75.7)	0.04 (0.91)	0.0 (1.2)	0.5 (13.6)	0.3 (8.6)	4.0
Corpus Christi Bay, Texas	Agriculture	1.6 (67.5)	0.01 (0.35)	0.0 (1.7)	0.6 (25.0)	0.1 (5.4)	2.4
Upper Laguna Madre, Texas	Agriculture	0.7 (70.3)	0.01 (0.91)	0.0 (1.1)	0.1 (8.9)	0.2 (18.7)	1.0
Lower Laguna Madre, Texas	Agriculture	6.6 (76.7)	0.03 (0.34)	0.0 (0.2)	1.3 (15.0)	0.7 (7.8)	8.7
Average		7.9 (68)	0.1 (0.8)	0.3 (3)	1.8 (16)	1.4 (13)	11.5
Barnegat Bay, New Jersey	Atmospheric	2.6 (34.9)	0.1 (2.0)	0.6 (8.4)	0.3 (3.7)	3.7 (51.0)	7.3
St. Catherines-Sapelo, Georgia	Atmospheric	0.3 (14.7)	0.4 (15.5)	0.3 (11.3)	0.01 (0.64)	1.3 (57.9)	2.3
Barataria Bay, Louisiana	Atmospheric	2.3 (27.5)	0.2 (2.1)	0.003 (0.04)	2.6 (31.8)	3.2 (38.6)	8.3
Average		1.7 (26)	0.2 (7)	0.3 (7)	1.0 (12)	2.8 (49)	6.0

analysis, we classified the watershed-estuary systems on the basis of the major sources of N to estuaries (Table 3). Systems were classified as urban, agriculture, or atmospheric, depending on whether N inputs to the estuaries were dominated by urban, agriculture, or atmospheric sources, respectively (Table 3).

Urban N Sources

Eleven of the 34 Atlantic and Gulf Coast estuaries investigated showed the major source of N to be derived from urban sources (Table 3). Eight of these 11 estuaries were located in the Northeastern U.S. For these 11 urban-dominated watersheds, the total anthropogenic N inputs to the watersheds were strongly related ($r^2 = 0.88$) to population density (Fig. 3), and a significant ($p < 0.05$) relationship between the urban sources (point sources, septic systems, and nonpoint source runoff) of N and the total input of N to the estuary was observed (Fig. 4; TN input to estuary [kg N ha^{-1} yr^{-1}] = 1.1 × urban N sources [kg N ha^{-1} yr^{-1}] + 4.8; $r^2 = 0.96$). Sewage effluents from both point sources and septic tanks were the dominant urban N sources, accounting for 57% (average over all 11 watersheds) of the TN inputs to these urban-dominated estuaries. Septic systems contributed 10% and urban nonpoint source runoff supplied 5% of the TN inputs to these systems. The relative contribution of the different urban N sources was var-

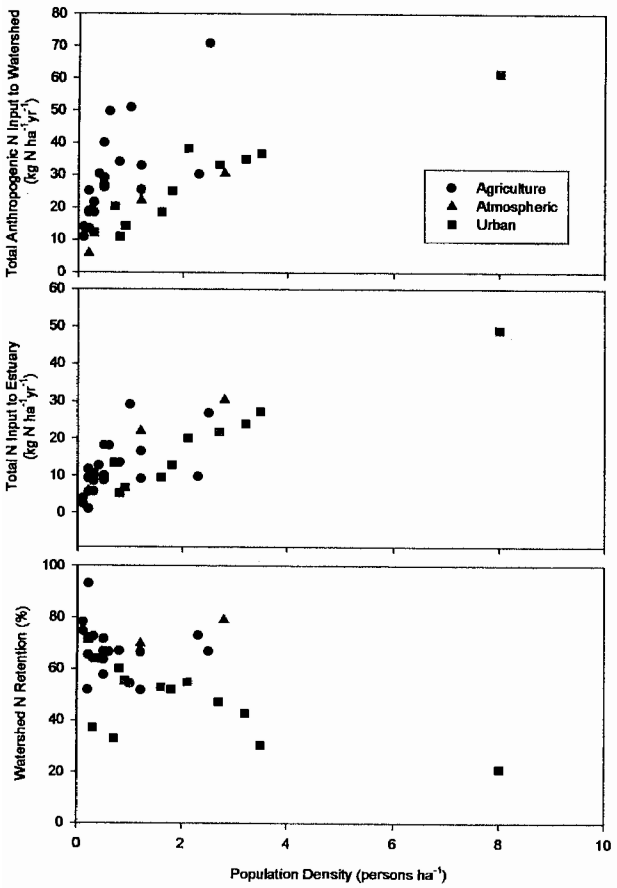

Fig. 3. Relationships between population density and total anthropogenic nitrogen inputs to watersheds, total nitrogen inputs to estuaries, and watershed nitrogen retention. Systems with estuary nitrogen sources dominated from urban inputs, agricultural inputs, and atmospheric deposition are indicated.

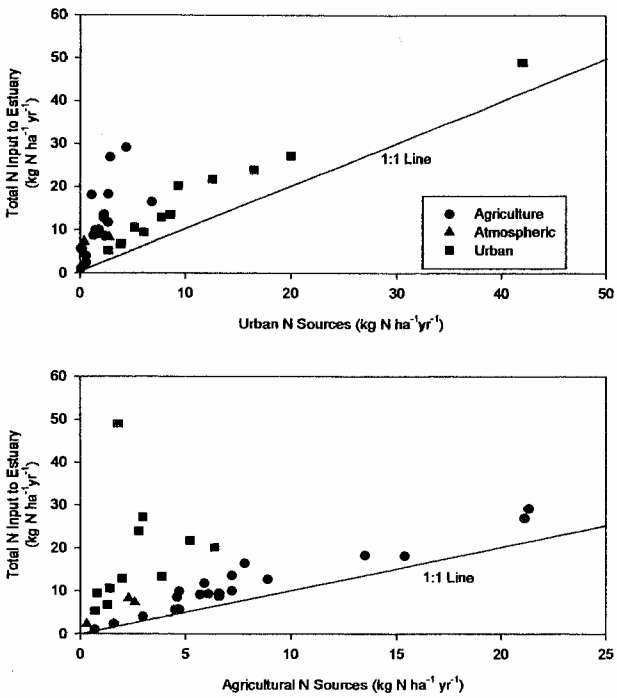

Fig. 4. Relationships between total nitrogen inputs to estuaries from urban nitrogen sources and agricultural nitrogen sources. Sites with estuary nitrogen sources dominated from urban inputs, agricultural inputs, and atmospheric deposition are indicated. The 1:1 line is indicated.

iable across watersheds (Table 3). Casco Bay (15%) and Great Bay (20%) had significant contributions from urban nonpoint source runoff. Some watersheds, such as Buzzards Bay, Massachusetts (16%), Great Bay (13%), Narragansett Bay, Rhode Island (11%), Charleston Harbor (15%), and Terrebonne-Timbalier Bays (16%), had relatively large contributions from septic systems.

We observed a significant relationship between N inputs to the urban-dominated estuaries and the human population in the watershed (total estuary N input [kg N ha^{-1} yr^{-1}] = 5.6 × population [persons ha^{-1}] + 5.3; r^2 = 0.93; Fig. 3). The slope of this relationship suggests that each human supplied 5.6 kg N yr^{-1} to the urban-dominated estuaries, which is consistent with previous estimates of the per capita N load from human N excretion (4.4–5.2 kg N $person^{-1}$ yr^{-1}; Howarth et al. 1996) and wastewater effluent (3.3 kg N $person^{-1}$ yr^{-1} from Meybeck et al. 1989; 2.0 to 7.23 kg N $person^{-1}$ yr^{-1} from the U.S. Environmental Protection Agency 1980). We also observed a similar relationship between estuarine N input and population for all the study watersheds, but it was not as strong as that derived from the 11 urban-dominated watersheds (estuary input of N [kg N ha^{-1} yr^{-1}] = 5.7 × population [persons ha^{-1}] + 21; r^2 = 0.37; $p < 0.05$).

Agriculture N Sources

Agricultural activities were the major source of N inputs to 20 of the Atlantic and Gulf Coast estuaries (Table 3). Estuaries largely influenced by agriculture N sources were generally located south of Delaware Bay, Delaware. Fertilizer (46%, the mean for all watersheds) and manure (32.3%) N applications were the primary inputs of N to these agriculture lands. N fixation (15.2%, the mean for all watersheds) and atmospheric deposition (6.1%) were relatively small N sources. The importance of agricultural inputs to the supply of N to Atlantic and Gulf Coast estuaries is evident through the relationship between TN input to the estuary and N runoff from agriculture-dominated watersheds (TN input to estuary [kg N ha^{-1} yr^{-1}] = 1.2 × agricultural N sources [kg N ha^{-1} yr^{-1}] + 1.8; r^2 = 0.92; Fig. 4). This relationship suggests that virtu-

ally all the N exported to these agriculture-dominated estuaries is supplied by runoff from agricultural lands. We also observed a relationship between population density and TN inputs to agriculture-dominated estuaries (estuary input of N [kg N ha^{-1} yr^{-1}] = 6.1 × population [persons ha^{-1}] + 7.3; $r^2 = 0.32$; $p < 0.05$), but the relationship was not as strong as observed for urban-dominated estuaries (Fig. 3).

Atmospheric N Sources

Atmospheric N deposition to the watershed (including surface waters in the watershed) was the dominant watershed N source for 3 of the 34 watershed-estuary systems (Table 3). Atmospheric N deposition accounted for 51%, 58%, and 39% of the TN inputs to estuaries of Barnegat Bay, New Jersey, St. Catherines-Sapelo Island, and Barataria Bay, respectively (Table 3). Atmospheric N deposition from the watershed was also a major contributor (37%) of the TN inputs to the estuaries of the Terrebonne-Timbalier Bays, which are adjacent to the Barataria Bay estuary. The relatively high atmospheric contribution associated with the Barnegat Bay estuary occurred primarily because the effluent from sewage treatment plants in the watershed is discharged offshore, bypassing the estuary.

The relatively high atmospheric N input to the St. Catherines-Sapelo estuary could be a consequence of either the small TN load (2.3 kg N ha^{-1} yr^{-1}) or relatively high atmospheric N deposition rates. Atmospheric N deposition to the St. Catherines-Sapelo estuary was not unlike atmospheric N deposition rates to estuaries in the same region. The contribution made by direct deposition to the total atmospheric N input to St. Catherines-Sapelo estuary was not different than most estuaries (Table 4). The relatively large atmospheric N input to the St. Catherines-Sapelo estuary appears to be related to the small TN load.

Barataria Bay and the Terrebonne-Timbalier Bays had relatively high atmospheric N inputs from the watershed. The dominant atmospheric N source for these systems was atmospheric N deposition to surface waters in the watershed. Surface water areas accounted for 15% and 26% of total watershed areas for Barataria and Terrebonne-Timbalier, respectively, and were not unlike the contribution made by surface waters to the total watershed area in other systems. These two systems had high atmospheric N deposition rates compared to all other systems (10.9 and 11.7 kg N ha^{-1} yr^{-1} for Terrebonne-Timbalier and Barataria, respectively). Most of the atmospheric N input was dominated by NO_3^- deposition (71% of the TN input), which is likely to be derived from fossil fuel combustion associated with electric utilities and automobile exhaust in major Gulf Coast cities (Galveston and Houston, Texas) upwind of these systems. These two systems and others in this region (from West Mississippi Sound to Galveston Bay) had some of the highest NH_4^+ deposition rates (2.4–3.3 kg N ha^{-1} yr^{-1}). These high NH_4^+ deposition rates suggest that there are important local sources of NH_4^+ from livestock operations, fossil fuel combustion, and oil refinery facilities.

Atmospheric N deposition also enters estuaries directly as wet and dry N deposition to the surface of the estuaries. The contribution made by direct atmospheric N deposition to the total atmospheric N inputs was generally < 30% (Table 4). Eleven out of 34 watershed-estuary systems had direct atmospheric N inputs that exceeded 30% of the total atmospheric N inputs (Table 4). For these 11 systems, the contribution made by atmospheric N deposition to the TN inputs ranged 13–59%, with a mean value of 30%. The other 23 systems with direct atmospheric deposition contributions < 30% had total atmospheric N deposition contributions that ranged 5–72% of the TN inputs, with an average of 21%. High direct N deposition to the surface of the estuary is not a prerequisite for high atmospheric contributions to the TN inputs to estuaries.

Watershed Retention of N Inputs

We found a significant relationship between total anthropogenic N inputs to the watershed and N inputs to the estuary for all systems (N input to estuary [kg N ha^{-1} yr^{-1}] = 0.57 × total anthropogenic N input to watershed [kg N ha^{-1} yr^{-1}] − 2.7; $r^2 = 0.72$, n = 34; Fig. 5). This linear relationship suggests that on average 57% of the total anthropogenic N inputs to watersheds were exported to the Atlantic and Gulf Coast estuaries. Over all watersheds watershed N retention rates ranged from 21% for the urban-dominated Massachusetts Bay watershed to 97% for the agricultural-dominated Upper Laguna Madre watershed.

We estimated a large range of watershed N retention values for different land uses. The percentage of TN retained by forests and agricultural lands was high. Retention of atmospheric N deposition by upland forests averaged 92%; values ranged from a low of 85% for Chesapeake Bay to a high of 95% for upland forests in many watersheds. Retention of N by agricultural lands was somewhat less than N retention by forests. Average retention of the TN inputs by agricultural lands was 79%, with Indian River exhibiting the lowest retention of 65% and Upper Laguna Madre the greatest N retention of 97%. In contrast to forests and agricultural lands, urban-dominated water-

TABLE 4. Atmospheric N inputs to each of the selected watershed-estuary systems. Note: the percent contribution made by atmospheric N deposition to the total N input to the estuary was calculated as the sum of the direct and watershed atmospheric N inputs divided by the sum of the total watershed and direct deposition N inputs. [a] indicates data from Castro et al. (2000).

Watershed Location	Atmospheric Nitrogen Deposition to Surface of Estuary[a] (10^6 kg N yr^{-1})	Atmospheric Nitrogen From Watershed to Estuary (10^6 kg N yr^{-1})	Total Watershed Nitrogen to Estuary (10^6 kg N yr^{-1})	Contribution Made by Atmospheric Deposition to the Total Nitrogen Inputs to the Estuary (% of total N input)
Northeast				
Casco Bay, Maine	0.20	0.4	1.1	43.2
Great Bay, New Hampshire	0.03	0.3	1.6	22.3
Merrimack River, Massachusetts	0.01	2.4	11.0	21.7
Massachusetts Bay, Massachusetts	0.57	1.1	10.2	15.0
Buzzards Bay, Massachusetts	0.39	0.4	2.2	30.1
Narragansett Bay, Rhode Island	0.30	1.5	10.8	16.5
Long Island Sound, Connecticut	2.70	8.9	48.9	22.6
Hudson River-Raritan Bay, New York	0.68	14.3	82.7	17.9
Barnegat Bay, New Jersey	0.16	0.5	0.9	63.9
Great Bay, New Jersey	0.24	1.0	2.8	39.5
Mid-Atlantic				
Delaware Bay, Delaware	1.87	11.5	57.2	22.7
Chesapeake Bay	10.53	48.4	186.1	30.0
Pamlico-Pungo Sound, North Carolina	3.57	4.4	43.4	17.0
Southeast				
Wynah Bay, South Carolina	0.05	5.2	51.1	10.3
Charleston Harbor, South Carolina	0.04	3.5	54.4	6.6
St. Helena Sound, South Carolina	0.09	1.0	6.1	17.5
St. Catherines-Sapelo, Georgia	0.10	0.3	0.4	71.8
Altamaha River, Georgia	0.02	3.2	60.8	5.3
Indian River, Florida	0.36	0.8	6.9	16.5
Eastern Gulf Coast				
Charlotte Harbor, Florida	0.24	1.2	13.1	10.5
Tampa Bay, Florida	0.46	1.5	13.0	14.5
Apalachee Bay, Florida	0.76	0.9	7.3	20.0
Apalachicola Bay, Florida	0.28	3.8	46.0	8.9
Mobile Bay, Alabama	0.65	12.3	87.4	14.7
West Mississippi Sound, Mississippi	3.22	3.7	33.6	18.9
Barataria Bay, Louisiana	0.71	1.3	3.2	52.6
Terrebonne-Timbalier Bays, Louisiana	1.02	0.8	2.1	58.8
Western Gulf Coast				
Calcasieu Lake, Louisiana	0.20	2.5	10.1	26.4
Sabine Lake, Texas	0.16	5.7	44.6	13.0
Galveston Bay, Texas	0.85	10.2	95.7	11.4
Matagorda Bay, Texas	0.42	3.9	43.4	9.8
Corpus Christi Bay, Texas	0.21	0.6	10.5	7.3
Upper Laguna Madre, Texas	0.34	0.2	0.9	41.3
Lower Laguna Madre, Texas	0.54	0.9	10.6	12.7

sheds exhibited the lowest watershed retention of N (21–60% of the N inputs). Watershed retention of TN by Massachusetts Bay was only 21% and in Narragansett Bay, watershed retention of TN was 29%. For urban-dominated watersheds, we observed a decrease in watershed retention of N with increasing population density (watershed retention of N [%] = $-3.6 \times$ population [persons ha^{-1}] + 52.6; $r^2 = 0.39$; $p < 0.05$; Fig. 3). For these urban-dominated watersheds, net food import is an important source of the TN input (Table 2). This N input is consumed by humans and discharged to surface waters with little attenuation in wastewater treatment plants. This pathway of N transport bypasses many of the mechanisms of N retention and loss that application of N to the land surface are subjected to, thereby decreasing watershed retention.

Conclusions and Management Implications

Urban and agricultural activities are the dominant sources of N to the 34 watershed-estuary system examined in this study. N inputs to 11 of these estuaries were dominated by urban sources. Most of these 11 urban-dominated estuaries were located in the northeastern U.S. (Table 3). Sewage ef-

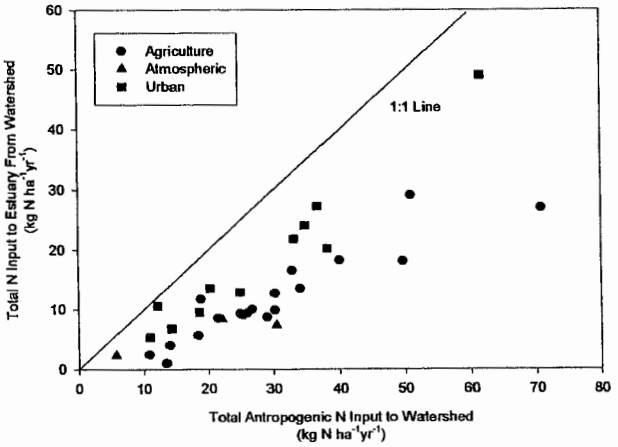

Fig. 5. Relationship between total anthropogenic nitrogen inputs to estuaries and total nitrogen inputs to watersheds. Sites with estuary nitrogen sources dominated from urban inputs, agricultural inputs, and atmospheric deposition are indicated. The 1:1 line is indicated. The vertical distance between an observation and the 1:1 line represents the quantity of nitrogen retained in a watershed.

fluents from both point sources and septic tanks were the dominant urban N sources, accounting for 57% (average over all 11 watersheds) of the TN inputs to these urban-dominated estuaries. Septic systems contributed 10% and urban nonpoint source runoff supplied 5% of the TN inputs to these systems. Since the supply of N to these estuaries was largely the result of wastewater effluent, the most effective approach to control N inputs to these estuaries would be through tertiary treatment of wastewater to remove N.

Agricultural activities were the major source of N inputs to 20 Atlantic and Gulf Coast estuaries (Table 3). The studied estuaries, which are heavily influenced by agriculture N sources, were generally located south of Delaware Bay. Fertilizer (46%, the mean for these 20 watersheds) and manure (32.3%) N applications were the primary inputs of N to these agriculture lands. To minimize N runoff from these agricultural sources, agricultural practices should be modified to improve the efficiency of the use of N fertilizers and manure during crop production.

Atmospheric N deposition was generally not a dominant N source for most systems. It did make an important contribution to three watershed-estuary systems since atmospheric N deposition generally contributed < 30% of the TN load to most of the 34 watershed-estuary systems. Efforts to reduce emissions of NO_x from combustion and NH_3 emissions from fertilizer and livestock wastes are unlikely to make a substantial contribution towards reducing TN inputs except to a few study sites.

There is much uncertainty about the fate of N in watersheds. We found that watersheds retained about 57% of the TN input. The percentage of TN retained by upland forests and agricultural lands was high. For forests, we estimated the mean retention of atmospheric N deposition of about 92% with a range of 85–95%. Watersheds with N losses dominated by agriculture N sources had an average retention of 79% and a range of 65–97%. Urban-dominated watersheds exhibited the lowest watershed retention of N, ranging 21–65%. More research is needed to determine the fate of the N that is retained and stored by these different land uses.

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