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Anthropogenic nitrogen sources and relationships to riverine nitrogen export in the northeastern U.S.A.

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Abstract. Human activities have greatly altered the nitrogen (N) cycle, accelerating the rate of N fixation in landscapes and delivery of N to water bodies. To examine relationships between anthropogenic N inputs and riverine N export, we constructed budgets describing N inputs and losses for 16 catchments, which encompass a range of climatic variability and are major drainages to the coast of the North Atlantic Ocean along a latitudinal profile from Maine to Virginia. Using data from the early 1990's, we quantified inputs of N to each catchment from atmospheric deposition, application of nitrogenous fertilizers, biological nitrogen fixation, and import of N in agricultural products (food and feed). We compared these inputs with N losses from the system in riverine export.

The importance of the relative sources varies widely by catchment and is related to land use. Net atmospheric deposition was the largest N source (>60%) to the forested basins of northern New England (e.g. Penobscot and Kennebec); net import of N in food was the largest source of N to the more populated regions of southern New England (e.g. Charles & Blackstone); and agricultural inputs were the dominant N sources in the Mid-Atlantic region (e.g. Schuylkill & Potomac). Over the combined area of the catchments, net atmospheric deposition was the largest single source input (31%), followed by net imports of N in food and feed (25%), fixation in agricultural lands (24%), fertilizer use (15%), and fixation in forests (5%). The combined effect of fertilizer use, fixation in crop lands, and animal feed imports makes agriculture the largest overall source of N. Riverine export of N is well correlated with N inputs, but it accounts for only a fraction (25%) of the total N inputs. This work provides an understanding of the sources of N in landscapes, and highlights how human activities impact N cycling in the northeast region.

Introduction

Human activities have greatly altered the nitrogen (N) cycle, accelerating the rate of N fixation in landscapes and delivery of N to water bodies (Galloway et al. 1995; Howarth et al. 1996; Smil 1997; Vitousek et al. 1997; Caraco

& Cole 1999). In most estuaries, over-enrichment of N leads to eutrophication, presently the greatest pollution problem in coastal marine waters of the United States (NRC 2000). Over 40% of the estuaries in the U.S. are degraded from eutrophication, with particularly severe problems in the New England and mid-Atlantic regions (Bricker 1999). Nitrogen loadings in major U.S. rivers have increased during recent decades (e.g. Stoddard 1991; Turner & Rabalais 1991; Puckett et al. 1995; Jaworski et al. 1997). Most N delivered to coastal waters in the U.S. comes from non-point sources in the landscape, with agricultural sources and atmospheric deposition being major contributors (Howarth et al. 1996; Smith et al. 1997; Goolsby et al. 1999; Castro et al. 2000). Understanding the sources of N loadings is essential to developing nutrient management strategies.

To examine relationships between N inputs and riverine N export, we established N budgets for 16 catchments in the northeast (NE) U.S.A. These basins encompass a range of climatic variability and are major drainages to the coast of the North Atlantic Ocean along a latitudinal profile from Maine to Virginia. Nitrogen budgets were established by quantifying all new inputs of N to each catchment, where ‘new’ refers to N that is either newly fixed within or transported into each catchment. Budget terms included inputs of N from atmospheric deposition, fertilizer use, net imports in food and feed, and biological fixation in agricultural areas and in forests. The total net inputs were compared with N losses from the system in riverine export.

Our N budgets allow us to assess the importance of N sources, highlighting how human activities have impacted N cycling in the NE region. The relative importance of the input terms varied widely by catchment and is related to land use. Over the combined area of the catchments, net atmospheric deposition was the largest single source input (31%), followed by imports of N in food and feed (25%), fixation in agricultural lands (24%), fertilizer use (15%), and fixation in forests (5%). Riverine export of N is well correlated with N inputs, but represents only a fraction (25%) of the total N inputs, with inputs exceeding outputs. This implies that large percentages of the N inputs are stored (e.g. in vegetation, soil, or groundwater) or lost (e.g. denitrified) in the catchment.

Methods

Study area

We selected sixteen river basins (Figure 1) draining to the NE coast of the U.S.A. The catchments include the Penobscot, Kennebec, Androscoggin, and Saco Rivers flowing into the Gulf of Maine; the Merrimack and Charles

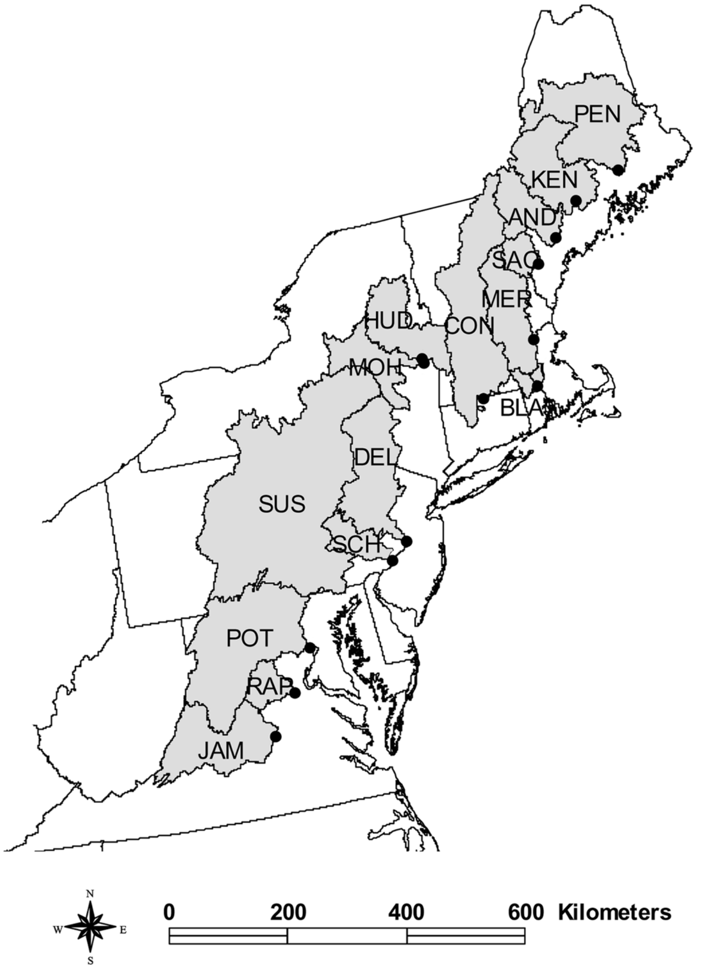

Figure 1. The basin boundaries are delineated upstream of USGS stations (denoted with black circles) where streamflow and water quality characteristics were measured. From north to south, the catchments include: Penobscot (PEN), Kennebec (KEN), Androscoggin (AND), Saco (SAC), Merrimack (MER), Charles (CHA), Blackstone (BLA), Connecticut (CON), Hudson (HUD), Mohawk (MOH), Delaware (DEL), Schuylkill (SCH), Susquehanna (SUS), Potomac (POT), Rappahannock (RAP), and James (JAM).

flowing into Massachusetts Bay; the Blackstone flowing into Narragansett Bay, the Connecticut flowing into Long Island Sound; the Mohawk and Upper Hudson flowing into the Hudson Estuary; the Delaware and Schuylkill flowing into Delaware Bay; and the Susquehanna, Potomac, Rappahannock, and James rivers flowing into Chesapeake Bay. We focused specifically on portions of the catchments upstream from individual USGS gaging stations, where long-term measurements of streamflow and water quality exist (Alexander et al. 1998). These stations are generally located upstream from the major coastal population centers of Portland, Boston, Providence, New York, Philadelphia, Washington D.C., and Richmond. We delineated catchment boundaries upstream of the gaging stations based on topography. The catchments cover a total area of approximately 250,000 km^2, and range in size from 475 km^2 (Charles) to 70,189 km^2 (Susquehanna).

Spatial data describing land use, population, and climate were aggregated to the scale of catchments using GIS software by weighting each county- or grid- estimate by the fraction of land area that is included within the catchment boundaries. Land use maps were obtained on a 30-m grid from the National Land Cover Database (NLCD), which represents land use during the early 1990's (MRLC 1995). County-level population data were obtained from the 1990 U.S. census (U.S. Dept. of Commerce 1990). Long-term monthly temperature and precipitation data were obtained on a half-degree grid from Kittel et al. (1997).

The combined landscapes of the 16 catchments were 72% forested, 19% agricultural, and 3% urban in the early 1990's, although land use varied greatly by catchment (Table 1). Portions of the Rappahannock, Potomac, Susquehanna, Schuylkill and Mohawk basins support intense crop and animal production, while the Penobscot and Kennebec largely support industrial timber production. Over 14 million people lived within the catchment boundaries in 1990, averaging 58 people km^{-2}. Population densities were highest in the Charles, Schuylkill, Blackstone, and Merrimack catchments with 556, 293, 276, and 143 people km^{-2}, respectively, and lowest in the Penobscot and Kennebec with only 8 and 9 people km^{-2}.

Climate data were obtained on a half degree grid for 1988–1993 from the VEMAP-II historical climate reconstruction (Kittel et al. 1997). Estimated average annual precipitation during 1988-1993 ranged from 930 to 1260 mm yr^{-1} with a mean of 1110 mm yr^{-1} (Table 1). Annual runoff during 1988–1993 ranged from 330 mm yr^{-1} in the Potomac to 670 mm yr^{-1} in the Saco. Evapotranspiration, as estimated from the annual water budget, varied with regional differences in mean temperature, from 44–50% of precipitation on the cool northern catchments (the Penobscot, Saco, Merrimack, and Connecticut), to more than 65% of precipitation on Potomac and Rappahannock. On average across the 16 catchments, evapotranspiration was 570 mm yr^1 or approximately 50% of precipitation.

Nitrogen budgets

We constructed N budgets that represent conditions during the early 1990's, following the approach put forth by Howarth et al. (1996). We quantify new inputs of N to each catchment, most of which are derived from human activ-

Table 1. Watershed characteristics

River Basin	Abbreviation	USGS[1] station	Area km^2	Persons[4] #km^2	Mean[2] Temp °C	Precip[2] mm/yr	Flow[1,3] mm/yr	Land Use[5] (% watershed area)					
								Forest	Agric.	Urban	Wetl.	Water	Other
Penobscot	PEN	1036390	20109	8	4.3	1075	588	83.8	1.5	0.4	5.2	6.2	3.0
Kennebec	KEN	1049265	13994	9	4.3	1085	566	79.6	5.9	0.9	3.6	6.4	3.6
Androscoggin	AND	1059000	8451	17	4.6	1151	640	84.6	4.8	1.1	3.4	4.6	1.5
Saco	SAC	1066000	3349	16	5.8	1218	672	87.4	3.6	0.8	3.9	3.1	1.1
Merrimack	MER	1100000	12005	143	7.4	1148	589	74.7	7.8	8.7	3.1	5.0	0.8
Charles	CHA	1103500	475	556	9.7	1207	583	59.3	8.4	22.2	7.2	2.5	0.5
Blackstone	BLA	1112500	1115	276	9.0	1260	651	63.3	8.1	17.6	6.8	3.4	0.8
Connecticut	CON	1184000	25019	65	6.3	1160	642	79.0	9.0	4.0	4.7	2.2	1.1
Hudson	HUD	1357540	11942	32	6.6	1126	622	80.8	10.4	2.7	2.5	3.4	0.2
Mohawk	MOH	1357500	8935	54	6.8	1142	548	63.1	28.0	4.7	2.6	1.5	0.1
Delaware	DEL	1463500	17560	85	8.7	1131	547	74.7	16.7	3.3	2.5	2.4	0.4
Schuylkill	SCH	1474500	4903	293	10.6	1134	488	48.1	38.4	10.2	0.7	1.2	1.5
Susquehanna	SUS	1578310	70189	54	8.9	1022	487	66.7	28.5	2.4	0.5	1.1	0.8
Potomac	POT	1646500	29940	63	11.3	985	328	60.8	34.6	2.6	0.5	0.7	0.8
Rappahannock	RAP	1668000	4134	24	12.6	1045	360	61.3	35.9	1.4	0.2	0.4	0.7
James	JAM	2035000	16206	24	10.1	934	407	80.6	15.6	1.4	0.6	0.7	1.1
Total Area	Total	—	*248326*	—	—	—	—	*72.2*	*19.3*	*2.9*	*2.1*	*2.4*	*1.1*

Watersheds listed in order from north to south. [1]USGS stream gaging station number; [2]Average temperature and precipitation for 1988–1993 from Kittel et al. 1997; [3]Average streamflow for water years 1988–1993 from USGS daily values; [4]U.S. Census 1990; [5]National land-cover database for the early 1990's from MRLC 1995.

ities: net atmospheric deposition, fertilizer application, agricultural and forest biological N fixation, and the net import of N in food and feed. Animal waste (manure) and human waste (sewage) are not considered as new inputs, as they represent recycling within a region; both of these terms are accounted for in our estimates of N transferred in food and feed. We compare the total N inputs to N exported in riverine streamflow. Throughout this paper, all graphs and tables show trends for the catchments arranged in geographical order from north to south, and all N fluxes are expressed in terms of kg N per km^2 of catchment area per year; for readers more accustomed to hectares, 100 kg km^{-2} yr^{-1} = 1 kg ha^{-1} yr^{-1}.

Input: Net atmospheric deposition

Associated with industrial, automotive, and biogenic N emissions, rates of N deposition in the eastern U.S. are the highest in the country, providing significant N inputs to our 16 catchments (NADP 2000). We considered wet and dry deposition of NO_y (NO_3^- and HNO_3), NH_x (NH_4^+ and NH_3), and AON (atmospheric organic nitrogen) in our budgets. To avoid double-accounting of N, we wanted to exclude all N that is both emitted and re-deposited within the catchment boundaries. Therefore, we quantify the new, net atmospheric deposition of NO_y, NH_x, and AON to each catchment via atmospheric deposition as described below.

Inorganic N deposition

Wet deposition of NO_3^- and NH_4^+ is measured regularly at a network of monitoring stations across the US called the National Atmospheric Deposition Program/National Trends Network (NADP/NTN). We chose data from 1991 to be representative of atmospheric deposition in the early 1990's since it was an average year for annual precipitation over the combined area of the catchments during the period 1988–1993. We obtained annual precipitation-weighted wet deposition values for 1991 for all stations in the New England and the Mid-Atlantic states from the NADP/NTN electronic database (NADP 2000). Using GIS software, we plotted the annual values observed at each sampling location, kriged the values to create isopleth maps, overlaid the catchment boundaries, and finally calculated the average value of wet deposition of NO_3^- and NH_4^+ for each catchment.

Inferential estimates of dry deposition can vary widely, due largely to different assumptions regarding deposition velocity values for various N species. For our budgets, we compared three methods for quantifying dry deposition (Figure 2(a)). First, we applied the commonly-used method of Lovett and Lindberg (1993), who observed that in eastern North America, total (wet + dry) N deposition (from NO_y and NH_x) is approximately twice measured wet deposition: *[total deposition (kg* ha^{-1} yr^{-1}*) = –0.72 + 2.07 * wet deposition (kg* ha^{-1} yr^{-1}*);* R^2 *= 0.91].* However, the data on which this equation is based (from the Integrated Forest Study, Johnson & Lindberg 1991) included just 2 low-elevation sites from the NE, and one of those (Maine) had a very limited data set (G. Lovett, personal communication).

Ollinger et al. (1993) observed that in the eastern US, spatial patterns of dry deposition do not correlate directly with patterns of wet deposition, leading to substantial differences in the ratio of wet to dry deposition across the region. They used deposition data from NADP/NTN and other monitoring networks to derive linear regressions predicting the concentrations of both wet and dry N species as a function of latitude and longitude in the northeastern U.S. Wet deposition of NO_3^- and NH_4^+ was estimated by multiplying by precipitation, and dry deposition of HNO_3 vapor and NO_3 and NH_4 aerosols was estimated by multiplying by deposition velocity constants (Ollinger et al. 1993). As a second method to consider, we applied this model to 13 of our 16 catchments using the precipitation data reported in Table 1. The Potomac, James and Rappahannock catchments are outside of the latitudinal range in which the regression equations are suitable.

Estimates of dry deposition using the Ollinger et al. (1993) model are lower than those obtained by Lovett and Lindberg (1993) or Holland et al. (1999), largely because of differences in the deposition velocities used in these models. As a third method of calculating dry N deposition, we take advantage of additional information regarding deposition velocities in our study area. Lovett and Rueth (1999) compiled several years' worth of data from 7 sites in the NE and report updated deposition velocities for HNO_3 vapor (2.14 cm s^{-1}) and NO_3 and $(NH_4)_2SO_4$ aerosols (0.12 cm s^{-1}). The reported deposition velocity for HNO_3 vapor, a significant form of dry N deposition in the eastern U.S. is substantially higher than that used by Ollinger et al. (1993) (1.3 cm s^{-1}). As a third approach, we combined the spatial model of Ollinger et al. (1993) with the revised deposition velocities reported in Lovett and Rueth (1999).

Estimates of total (wet + dry) N deposition are generally similar using the three methods (Figure 2(a)). For our N budgets we chose the third method, which most reflects current understanding of dry deposition to the region (G. Lovett, personal communication). This method – the spatial model of Ollinger et al. (1993) updated with deposition velocities of Lovett and Rueth (1999) – provides estimates of the NO_y and NH_x components of total N deposition (Figure 2(b)). For the Potomac, James and Rappahannock catchments (which are out of the latitudinal range in which the Ollinger et al. regression equations apply), we used the first method of Lovett and Lindberg (1993) to quantify deposition.

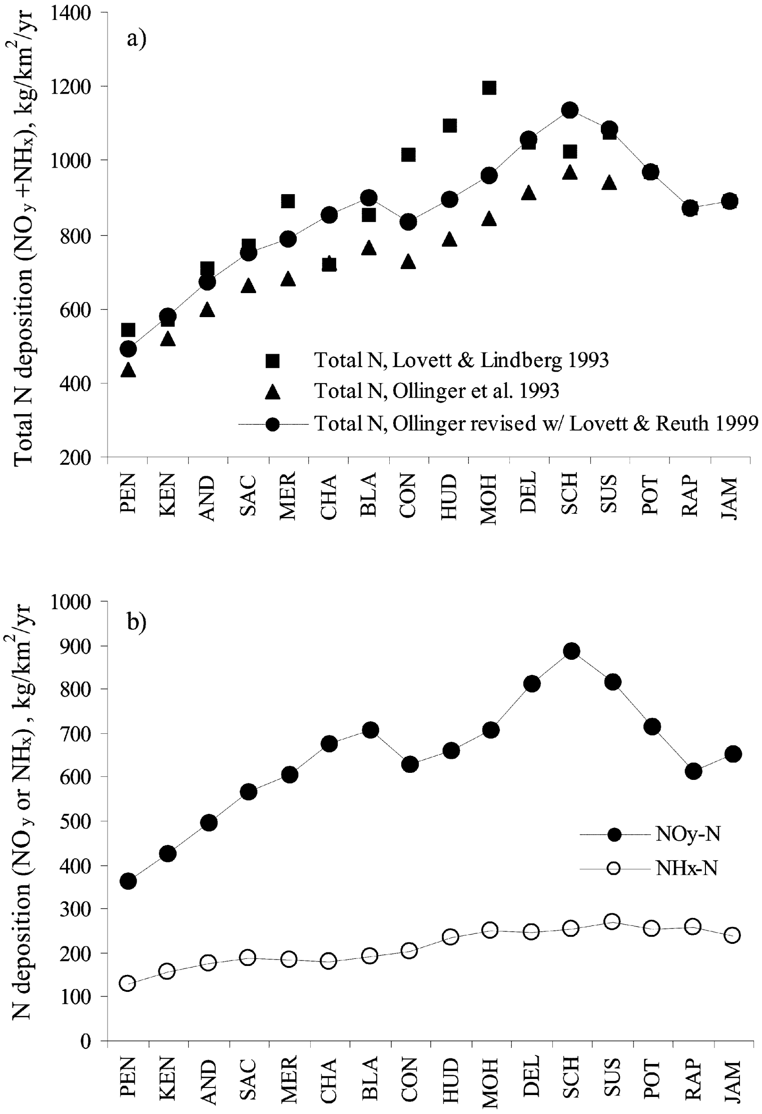

Figure 2. (a) Comparison of 3 models describing total (wet + dry) depositional inputs of inorganic N (NO_y + NH_x) to each catchment. Values indicated by dashed lines were used in budget calculations. (b) Portions of the total N input to each catchment from NO_y-N (in filled circles) and NH_x-N (in hollow circles), calculated using the revised Ollinger model.

Net NH_x Input

Approximately 90% of NH_x in the atmosphere comes from agricultural sources (Dentener & Crutzen 1994), with major emissions coming from animal wastes (manure) and lesser contributions from volatilization of fertilizers. Because NH_x is short-lived in the atmosphere, with residence times ranging from hours to a few weeks (Fangmeier et al. 1994), NH_x may redeposit within the same region from which it was emitted (Schlesinger & Hartley 1992; Prospero et al. 1996). Therefore, several studies over large spatial scales have simply assumed that NH_x deposition reflects local recycling and have ignored it as a new input (Howarth et al. 1996; Jordan & Weller 1996; Castro et al. 2000). The volatilization and deposition cycle may be complete over the scale of a large region, but it is unlikely that this recycling is complete over shorter distances, and several recent studies have highlighted long-range transport of NH_x (Dentener & Crutzen 1994; Galperin & Sofiev 1998). For example, with an area of 32,820 km^2, Belgium is almost 1/2 the size of the Susquehanna basin and is larger in size than all of our other study catchments. Source-receptor matrices produced by the Program for Monitoring and Evaluation of the Long-Range Transmission of Air Pollutants in Europe show that Belgium received transboundary imports of NH_x deposition from more than 7 nearby countries (EMEP 2001). As illustrated by this example, our catchments may be too small in size for complete recycling of NH_x to occur within the basin boundaries. Therefore, we explicitly consider both inputs of NH_x in atmospheric deposition and outputs of NH_x from volatilization to quantify the net depositional input of NH_x to each catchment.

We estimated total (wet + dry) input of NH_x using the revised Ollinger spatial model, as described above. Volatilization from animal waste (manure) is estimated based on NH_x emission factors that have been developed for animal populations (Table 2). These factors are highly variable because they vary with agricultural management practices and with the size, dietary intake, and excretion of each animal, all of which can vary substantially over space and time. We estimated NH_x volatilization for each catchment (Figure 3) based on 10 different published sets of emission factors. For our budgets, we used factors from Battye et al. (1994) because they are current estimates that are recommended to describe agricultural management practices in the U.S.A.

Fertilizers, especially those applied as urea, are potentially volatilized. Data on fertilizer types used in each catchment are described below (see ‘input: nitrogenous fertilizer use’). We estimated volatilization losses (Figure 3) as a percentage of fertilizers applied: 15% of urea, 2% of ammonium nitrate, 2.5% of nitrogen solutions (mixed urea and ammonium

Table 2. Ammonia emission rates from animals*, kg N $animal^{-1}$ yr^{-1}

Animal	Cass et al. 1982 *cited in Battye et al. 1994*	ApSimon et al. 1987	Buijsman et al. 1987	Kruse et al. 1989 *cited in Battye et al. 1994*	Moller & Schieferdecker 1989	Asman 1990 *cited in Battye et al. 1994*	Lee 1994 *cited in Battye et al. 1994*	Battye et al. 1994	Bouwman et al. 1997	van der Hoek 1998
Beef cattle	—	15.87	—	—	22.10	—	—	**18.83**	7.80	11.76
Dairy cattle	27.00	15.87	14.80	19.31	22.10	20.70	27.00	**18.83**	20.40	23.44
Young cattle	—	15.87	—	—	22.10	—	—	**10.72**	7.80	11.76
Pigs & hogs	4.50	2.35	2.30	2.86	5.20	3.96	1.60	**4.20**	4.00	3.98
Sheep	2.70	2.20	2.55	2.68	3.00	1.57	0.70	**2.77**	0.64	1.10
Goats	—	—	—	—	—	—	—	**5.26**	0.58	1.10
Horses	33.00	25.99	7.73	31.60	15.00	10.30	—	**10.03**	7.60	6.58
Chickens (layers)	0.24	0.19	0.21	0.23	0.22	0.26	0.12	**0.20**	0.20	0.30
Chickens (broilers)	0.24	0.19	0.21	0.23	0.22	0.26	0.12	**0.14**	0.20	0.30
Turkeys	0.66	0.19	0.21	0.23	0.22	0.26	0.10	**0.71**	0.20	0.76

* — No data. Values chosen for our nitrogen budgets, which best represent current agricultural management practices in the USA, are in boldface. If a more general ammonia emission factor was reported, we used the value for “cattle” for beef, dairy, and young, and we used the value for “poultry” fro layers, broilers, and turkeys.

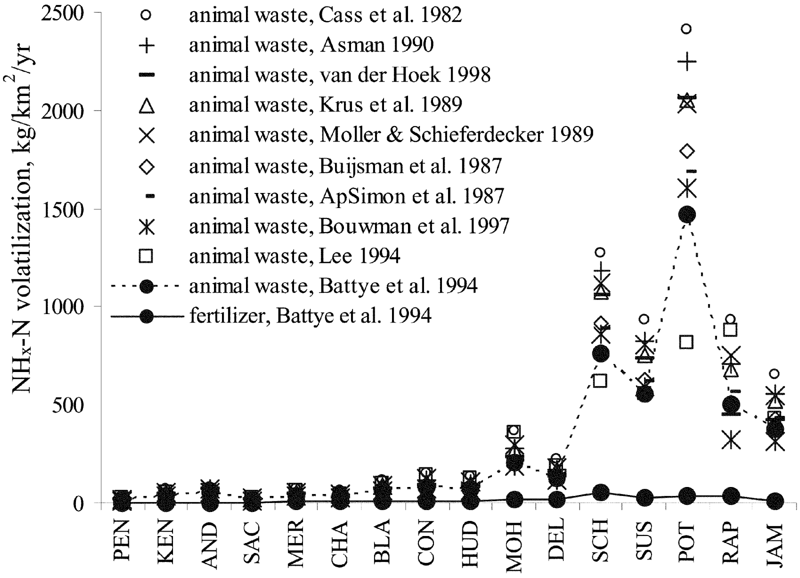

Figure 3. Estimates of NH_x volatilization from animal waste based on 10 published sets of animal emission factors. Values reported by Battye et al. 1994, which are recommended to describe U.S. agricultural practices, were used in our budget calculations. Estimates of NH_x volatilization from fertilizers are much lower in magnitude than the emissions from animal populations.

nitrate), 0.1% of anhydrous ammonia, and 2% of other combined fertilizers (Battye 1994).

An uncertain amount of reduced N redeposits within the same region from which it volatilized, but the existing wet deposition monitoring network may miss much of this redeposited N. NADP monitoring sites are generally located in forested areas rather than immediately downwind of farms and animal feedlots, and so NADP measurements likely capture long-range transport of NH_x rather than local sources. If we had detailed information on NH_x deposition downwind of these agricultural regions, we could simply have subtracted the NH_x volatilization emissions from the complete NH_x depositional inputs to estimate net NH_x inputs to each catchment. As a first approximation, we assumed that 75% of the ammonia volatilization that occurs from animal wastes and fertilizers is re-deposited locally, and thus is not an output from the catchment but rather represents a recycling within its boundaries. We assumed that the remaining 25% of the NH_x emissions from agricultural sources are transported long-range; this is treated as a volatilization output of NH_x-N from each catchment. Net NH_x gains or losses were then calculated as the difference between the depositional input and this volatilization output for each catchment.

Net organic nitrogen input

In addition to depositional inputs of inorganic N species, inputs of atmospheric organic nitrogen (AON) also can be substantial (see review by Neff et al. this volume). Measured fluxes of bulk DON deposition in New England ranged from 60 to 190 kg km^{-2} yr^{-1} (Campbell et al. 2000; Currie et al. 1996). Compiling data measurements from 41 different environments, Neff et al. found that the percentage contribution of organic nitrogen to total deposition is consistently around 30%. For our N budgets, we must consider how much of this AON is a new input to the region. Some of this organic N reflects recycling within the boundaries of our catchments, as it comes from natural biological sources (e.g. pollen) and from agricultural sources. However, AON is also formed as reaction byproducts between NO_y and hydrocarbons and is transported regionally (Neff et al. 2002). Modeled estimates of the long-range transport of AON (that is, new inputs of AON) predict about 44 kg km^{-2} yr^{-1} of AON deposition to the NE, largely derived from the Midwest (Neff et al. this volume). Given both measured and modeled estimates, we know that a significant fraction of the AON is a new input to the region. We assume that half of the AON (or 15% of total atmospheric N) is a new N input that is transported to each catchment (J. Neff, personal communication).

Input: Nitrogenous fertilizer use

The United States produces and consumes large amounts of fertilizers. In 1990, over 10 million metric tons of nitrogenous fertilizers were used in the U.S. (Battaglin & Goolsby 1994). We obtained digital data from the USGS on the N content of fertilizers sold within each county in 1991 (Battaglin & Goolsby 1994). These spatial maps are based on state data from the U.S. EPA, which were disaggregated to the county level by the Tennessee Valley Authority National Fertilizer and Environmental Research Center. Fertilizer sales estimates are available for each county, broken down by form: ammonium nitrate, anhydrous ammonia, nitrogen solutions, urea, and miscellaneous forms. We assumed that fertilizers are applied in the same county in which they are sold, which is a potential source of error. We aggregated the county level fertilizer data to the catchment level using GIS software by weighting each county estimate by the percent of county covered by each catchment.

Input: Net N import in human food and animal feed

Both humans and animals require food and feed, and these demands are met both by local agricultural production and by imports from other regions. Transfers of agricultural products can be important sources of N to a region. For example, Howarth et al. (1996) estimate that the net import of N in food and feed was 28% of total N inputs to the NE U.S. region as a whole. For each of the 16 NE catchments, we used the general method put forth by Jordan and Weller (1996), who quantified the net import of N as the balance between production of N in crops and animal products and the consumption of N by both humans and animals. We obtained crop and animal production data from the U.S. Department of Agriculture, National Agricultural Statistics Service (USDA/NASS). An agricultural census is conducted in the U.S. every 5 years (USDA/NASS 1992). We used county-level data from the 1992 U.S. Agriculture Census on the number of cows (beef and dairy), horses, pigs, sheep, chickens (layers and broilers), and turkeys, as well as information on pasture acreage and the annual production of crops typically used as food and feed, such as corn grain, corn silage, wheat, barley, oats, soybeans, and hay. These animal census and crop data were aggregated from the county to the catchment level by weighting the numbers reported for each county by the percent of county covered by each catchment.

Food and feed consumption

Human consumption of N in food was estimated by multiplying population density for each catchment (see Table 1) by a per capita intake of 5.0 kg N per year, a value typical of western populations on a high protein diet (Garrow et al. 2000). Animals are usually fed according to relatively straightforward dietary prescriptions designed for maintaining or gaining weight (van Horn et al. 1996). We estimated total demand for N in animal feed by multiplying per animal annual N requirements by the animal inventories from the 1992 agricultural census. We compared four sets of published values for the typical feed intake, or consumption, of N per animal type (Table 3). For our budgets, we chose the values reported by van Horn (1998) because they were developed based on current U.S. agricultural practices.

Crop production

To determine how much of the demands for N in consumption of food and feed can be satisfied by crop production, we calculated the N content of the entire crop harvest in each catchment. Crop production data were obtained from the 1992 agricultural census (USDA/NASS 1992), and nutrient contents for each crop type were assigned based on conversion factors reported by Lander and Moffitt (1996). Although most crops are produced for animal

Table 3. N consumption and waste production rates from animals*, kg N animal^{-1} yr^{-1}

Animal	Consumption: N intake rates				Waste production: N excretion rates					
	Thomas & Gilliam 1977	Bleken & Bakken 1997	van Horn 1998	van der Hoek & Bouwman 1999	Thomas & Gilliam 1977	Bleken & Bakken 1997	van Horn 1998	van der Hoek & Bouwman 1999	Smil 1999	SCS 1992
Beef cattle	56.00	74.60	**66.75**	51.30	44.00	66.60	**58.51**	40.70	50.00	41.72
Dairy cattle	166.00	126.70	**156.00**	177.00	120.00	93.90	**121.00**	140.50	80.00	79.47
Young cattle	—	57.70	—	74.10	—	36.45	—	67.70	—	36.26
Pigs & hogs	8.70	5.85	**8.51**	14.93	6.10	4.34	**5.84**	10.46	10.00	19.70
Sheep	—	9.76	**5.97**	27.60	—	6.70	**5.00**	25.00	5.00	13.04
Goats	—	13.90	**5.97**	23.70	—	12.30	**5.00**	19.90	—	—
Horses	—	—	**44.80**	—	—	50.00	**40.00**	45.00	35.00	27.81
Chickens (layers)	1.10	0.94	**0.84**	1.17	0.83	0.61	**0.55**	0.81	0.30	0.21
Chickens (broilers)	0.82	0.08	**0.13**	1.03	0.39	0.03	**0.07**	0.57	0.30	0.55
Turkeys	2.12	1.29	**0.62**	—	1.29	0.34	**0.39**	0.50	0.30	0.37

* — No data. Values chosen for our nitrogen budgets, which best represent current agricultural management practices in the USA, are in boldface.

consumption, a small fraction is grown for humans. To partition crop yields, we followed the distribution given by Jordan and Weller (1996), and assumed that 4% of corn, 61% of wheat, 6% of oats, 3% of barley, 17% of rye, 2% of soybeans, and 100% of potatoes were for human consumption. The remaining percentages of those crops went to feed animals, as did 100% of sorghum, hay, and pasture production. Following Jordan and Weller (1996), we assumed that pests, spoilage, and processing caused a 10% loss of all crops but hay and silage.

Animal production

Humans consume both animal and plant products. We quantified animal N production (i.e. meat, milk, and eggs) as the difference between animal feed consumption (intake) and animal excretion (waste production). Estimates of typical per animal feed intake and waste production vary significantly between studies (Table 3), because they depend on animal weights and efficiencies (for example, with the amount of milk a dairy cow produces) and on agricultural management practices. We compare values for N intake and excretion reported in the literature for Norway from Bleken and Bakken (1997), for the Netherlands and Europe from van der Hoek and Bouwman (1999), for the U.S. from Thomas and Gilliam (1977) and for the U.S. from van Horn et al. (1998). For our budgets, we chose the values reported by van Horn (1998), as these are the most current values that we could find that are based on U.S. agricultural practices. We assumed that spoilage and inedible components caused a 10% loss of animal products available for consumption.

Net import in food and feed

We estimated the net import of N in food and feed using a mass balance of needs versus production. We assumed that N import in feed equaled the difference between animal N demands and N produced in crops grown for animal consumption, and that N import in food equaled the difference between human N demands and N produced in food for humans. Imports were assumed to have come from regions outside of each catchment boundary. Thus: *[net import in food and feed = human consumption + animal consumption – crop production for animal consumption – crop production for human consumption – animal production for human consumption]*. In some cases the balances were negative, with crop and animal production exceeding human and animal demands; this indicates a net export of N in food and feed.

Calculations of net N import in food and feed were sensitive to the coefficients used to describe rates of animal intake and excretion (Figure 4(a)). For our N budgets, we chose the rates reported by van Horn (1998), which are based on current agricultural practices in the U.S. Considering just imports in feed, we checked our estimate of the net import of N in feed based

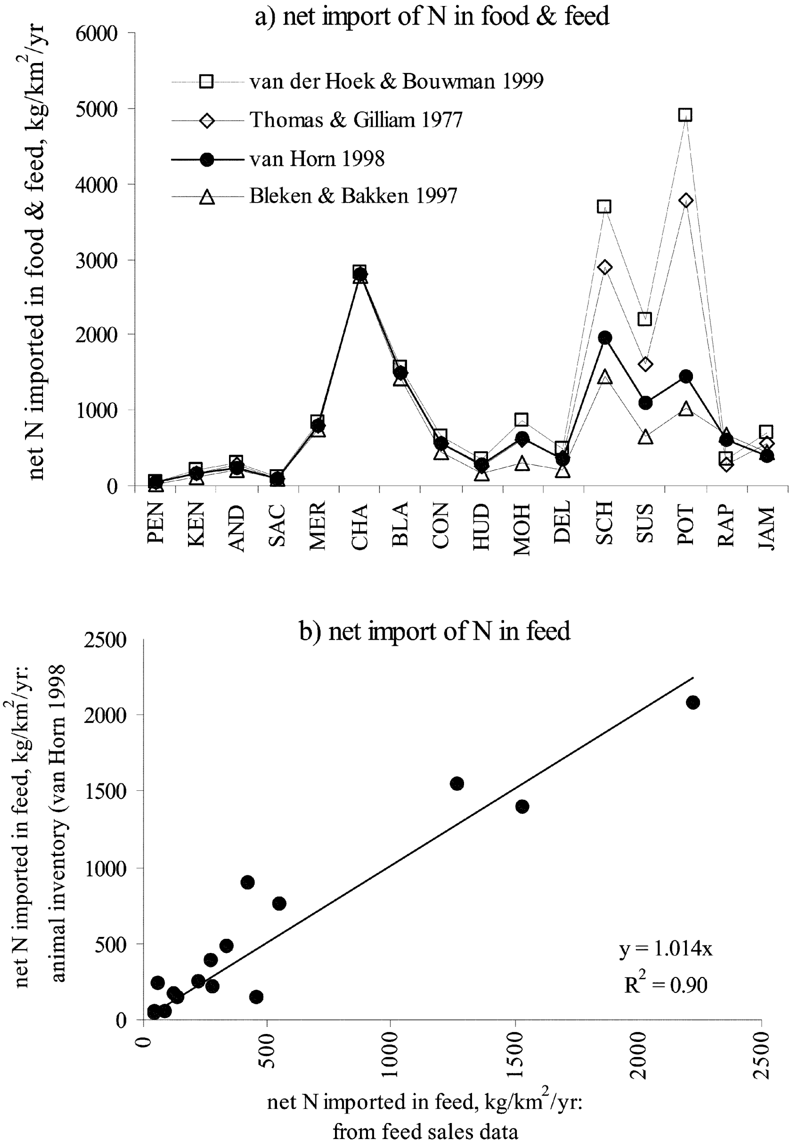

Figure 4. (a) We compare 4 estimates of N inputs to each catchment from net imports of food and feed, each based on a different set of coefficients describing the consumption and the waste production of N by animals. All estimates indicate that import of food is significant in the highly populated Charles and Blackstone catchments, while import of feed is significant in the highly agricultural Susquehanna, Potomac, and Rappahannock catchments. For our N budgets, we chose the rates reported by van Horn (1998), which are based on U.S. agricultural practices. (b) We checked our estimate of net N import in animal feed based on animal inventory data against an independent estimate based on feed expenditures. This nearly 1:1 relationship provides support that the coefficients of van Horn (1998) are appropriate to describe animal production in the northeastern U.S.

on animal inventory data against an independent estimate based on feed expenditure data (Figure 4(b)). Net N imports in feed were quantified from the animal inventory data using the coefficients for intake and excretion by van Horn 1998 that we used in our N budgets. County-level data on feed expenditures are reported in the 1992 Census of Agriculture. Feed expenditures were converted to estimates of N in feed by multiplying by average feed costs for the northeast region of approximately 3.0 kg dry matter per U.S. dollar, or 0.15 kg N per dollar (USDA/NASS 1997). The nearly 1:1 relationship between the net N import in feed calculated from the animal inventory data versus those calculated from feed expenditures within each catchment provide support that the coefficients of van Horn (1998) are appropriate to describe animal production in the northeastern U.S.

Input: Nitrogen fixation

Agricultural land

Alfalfa is the major N-fixing crop grown in the catchments, accounting for 23% of the N in total crop production, and 47% of the total N fixed in agricultural land. We estimated agricultural N fixation rates by multiplying the area of N-fixing species by literature-derived N fixation rates. We obtained data on the area of soybeans, alfalfa, hay, pasture, and snap beans from the 1992 agricultural census (USDA/NASS 1992). We used a fixation rate of 9600 kg km^{-2} yr^{-1} for soybeans (average of reported values by Rennie et al. 1978; Ham and Caldwell 1978; Deibert et al. 1979) and 22400 kg N km^{-2} yr^{-1} for alfalfa (Heichel et al. 1984); both rates are based on measurements in U.S. agroecosystems by the ^{15}N isotope dilution technique. We calculated the area of non-alfalfa leguminous hay by subtracting the reported area of grass hays (which do not fix N) and the area of alfalfa hay from the total area of hay. We assume that the remaining leguminous hay contains largely clovers and vetches, and assume a fixation rate of 11700 kg N km^{-2} yr^{-1}, based on averages of reported values for clover varieties (Brink 1990; Labandera et al. 1988; Rice 1980). Pasture was assumed to fix 1500 kg N km^{-2} yr^{-1} (after Jordan & Weller 1996). Snap beans grown in the catchments were assigned a rate of 90 kg N ha^{-1} yr^{-1} (Westerman et al. 1981). Other leguminous crops (such as other beans, peas, seeds, and peanuts) were not grown in significant quantities in the boundaries of our catchments. Although N-fixation rates for crops can vary significantly, the values we use for our N budgets are within the range of those considered by Smil (1999) to be most reliable.

Forested land

Both symbiotic and non-symbiotic N fixation can occur in eastern U.S. forests. Studies using the acetylene-reduction technique consistently report extremely low rates of N fixation by free-living microbes in soils and woody litter, with a range of 0.2–200 kg km^{-2} yr^{-1} (Tjepkema 1979; Roskowski 1980; Grant & Binkley 1987; Hendrickson 1990; Barkmann & Schwintzer 1999; Cleveland et al. 1999). We assumed a constant value of 40 kg km^{-2} yr^{-1} for non-symbiotic N fixation in all forests in the region. Rates of N fixation in eastern species with symbiotic associations can be quite high (∼3,000–7,500 kg km^{-2} yr^{-1}; Boring & Swank 1984), but both N fixation rates and the spatial distribution of these species are extremely variable and not well quantified. Black locust (*Robinia pseudoacacia*), a legume, occurs largely in southern Appalachia, where it ranges from 71% of the basal area in young stands to <1% in old, uneven-aged stands (Boring & Swank 1984). We obtained information on forest types from the USDA Forest Service's Forest Inventory and Analysis program (Hansen et al. 1992). For lack of additional information, we assumed that locust made up an average of 10% of oak-hickory stands, and that it fixed N at a mean rate of 5,000 kg km^{-2} yr^{-1} when in a pure stand (Boring & Swank 1984). Speckled alder (*Alnus incana* spp. rugosa) and common or hazel alder (*A. serrulata*) are actinorhizal N-fixing shrubs that occur in wet soils across the region. We assumed that alder covered 10% of wetland areas, and that it fixed N at a mean rate of 4,000 kg km^{-2} yr^{-1} when in a pure stand, as observed in the Adirondack Mountains, New York (Hurd et al. 2001).

Output: Riverine export

N export, or loading, in streamflow was quantified by multiplying stream N concentration by the flow rate of water. We obtained daily discharge values for the stream gaging stations located at the outlet of each catchment (see Table 1) from the USGS National Water Information System (USGS 2000). Stream N concentrations (of total N, including NO_3^-, NH_4^+ and dissolved organic nitrogen) were sampled at each of these sites approximately monthly, and were obtained from the databases of the USGS water quality monitoring networks (Alexander et al.1998). Stream N export was calculated from these data using the USGS 'estimator' software, which is a regression-based approach that allows flow-weighted interpolation of the discrete measurements of concentration, and has bias corrections (Cohn et al. 1992). We report average values of the annual N exports (or mass loadings) computed over the years 1988–1993.

Results

Atmospheric deposition was an important pathway by which N entered the catchments (Table 4). Total new inputs of N in deposition ranged from 575 kg km^{-2} yr^{-1} in the Penobscot to 1212 kg km^{-2} yr^{-1} in the Delaware (Table 4). Over the combined area of the catchments, NO_y, NH_x, and AON accounted for 69%, 15%, and 16% of the net N inputs, respectively, and new inputs of N in atmospheric deposition accounted for 31% of the total N inputs to the landscape.

Total atmospheric inputs from deposition of NO_y were lowest in the northern Maine catchments and highest in the Mid-Atlantic region, ranging from 362 kg km^{-2} yr^{-1} in the Penobscot to 885 kg km^{-2} yr^{-1} in the Schuylkill. Total net NH_x deposition, representing the fraction of total NH_x that is a new input to each catchment, averaged 120 kg km^{-2} yr^{-1}. Ammonia volatilization from fertilizer ranged from 1 kg km^{-2} yr^{-1} in the Saco to 58 kg km^{-2} yr^{-1} in the Schuylkill, while ammonia volatilization from animal waste ranged from 13 kg km^{-2} yr^{-1} in the Penobscot to 1459 kg km^{-2} yr^{-1} in the Potomac (see Figure 3). Net NH_x input was negative in the heavily agricultural Potomac basin (–119 kg km^{-2} yr^{-1}), with net volatilization outputs exceeding net depositional inputs due largely to the large number of poultry in the catchment and their associated emissions. Total net AON input, representing the fraction of AON that is a new input to each catchment, ranged from 88 kg km^{-2} yr^{-1} in the Potomac to 205 kg km^{-2} yr^{-1} in the Schuylkill.

Annual fertilizer N inputs increased with the percent of catchment area in agriculture, from less than 60 kg km^{-2} yr^{-1} on the forested catchments of the Saco and Kennebec to over 1000 kg km^{-2} yr^{-1} on the Schuylkill, Potomac and Rappahannock, where approximately 35% of the land area is in agriculture. Across the 16 catchments, N inputs from nitrogenous fertilizers averaged 474 kg km^{-2} yr^{-1} or 15% of the total N inputs. Fertilizer was applied mostly in compound forms (56%), although significant quantities of mixed urea and ammonium nitrate solutions (24%) and urea (14%) were applied. Much smaller quantities were applied in the form of ammonium nitrate (3%) and anhydrous ammonia (3%). The fractions containing urea are most susceptible to ammonia volatilization.

Human N consumption is proportional to the distribution of population, which is greatest in the Charles, Blackstone, Schuylkill, and Merrimack basins (Table 1). Animal N consumption was greatest on the Potomac, Schuylkill, Susquehanna, and Rappahannock, driven by large cattle herds and poultry demands (Table 5). Human and animal dietary N demands are satisfied by consumption of crops (forage, grains, fruits, vegetables) and animal products (meat, milk, and eggs). Across the 16 catchments, hay dominated the crop N production (38%), followed by pasture forages (23%), corn grain

Table 4. Atomospheric nitrogen deposition*, kg N km^2 yr^{-1}

Watershed	Total NO_y dep.	Total NH_x dep.	Total inorganic NO_y+NH_x dep.	NH_x vol. from fertilizer	NH_x vol. from an Waste	Total NH_x vol.	Net NH_x dep.	Net atm. N dep.	Total net N dep.
Penobscot	362	129	491	3	13	17	125	88	575
Kennebec	428	154	582	2	38	40	144	105	677
Androscoggin	495	176	671	3	48	51	163	121	779
Saco	566	187	753	1	15	16	183	136	885
Merrimack	606	184	790	5	40	45	173	142	921
Charles	674	178	852	8	31	39	168	153	996
Blackstone	707	190	897	12	66	77	171	162	1040
Connecticut	631	204	835	12	84	95	181	150	962
Hudson	658	234	893	8	73	81	214	161	1033
Mohawk	708	250	958	16	207	223	195	172	1075
Delaware	811	248	1059	22	127	149	211	191	1212
Schuylkill	885	253	1138	58	740	798	53	205	1143
Susquehanna	816	269	1085	28	540	569	127	195	1138
Potomac	714	255	969	35	1459	1494	–119	174	769
Rappahannock	615	256	871	33	506	538	121	157	893
James	652	237	889	12	373	384	141	160	953
Area-wght avg.	*677*	*228*	*904*	*19*	*412*	*431*	*120*	*163*	*959*

*See text for description of the net depositional terms. Boldface values were used in our budgets.

Table 5. Nitrogen transfers in food and feed, kg N km^{-2} yr^{-1}

Watershed	Animal N consumption	Human N consumption	Crop production N for animals	Crop production N for humans	Animal N products for humans	Net N import in feed for animals	Net N import in food for humans	**Net N import in food and feed**
Penobscot	122	38	67	32	24	55	–18	36
Kennebec	293	46	122	2	61	171	–17	154
Androscoggin	373	84	126	9	85	247	–10	237
Saco	123	82	74	4	23	49	55	104
Merrimack	335	713	185	2	64	150	647	797
Charles	201	2781	138	1	36	62	2745	2807
Blackstone	501	1380	284	2	99	217	1279	1495
Connecticut	764	324	367	8	149	398	167	565
Hudson	687	161	436	6	135	251	20	271
Mohawk	1977	272	1219	14	391	758	–134	624
Delaware	1001	426	846	32	197	155	197	352
Schuylkill	3523	1464	2122	102	810	1401	551	1952
Susquehanna	3097	270	1543	50	679	1554	–459	1095
Potomac	3822	313	1737	53	892	2085	–633	1452
Rappahannock	2673	121	1775	34	379	898	–291	607
James	1299	120	812	6	205	487	–91	395
Area-wght. Avg.	*1847*	*289*	*960*	*31*	*397*	*887*	*–139*	*748*
Sum	*20791*	*8595*	*11855*	*358*	*4230*	*8936*	*4008*	*12944*

(15%) and corn silage (12%). Wheat, barley, and oats each contributed less than 2% of total N in crop production; soybeans contributed an average of 6% with peak production in the Schuylkill catchment. Overall, N produced by crop and animal production was used to satisfy human and animal dietary needs. Of the total N produced in food and feed within the catchments, 69% was from crops that were fed to animals, 2% was from crops that were fed to humans, and 29% was from animals that were fed to humans. In catchments where the dietary demands for N could not be met by local crop and animal production, N was imported in food and feed (Table 5). Import of N in animal feed was most significant in the largely agricultural basins of the mid-Atlantic: the Potomac (2085 kg km^{-2} yr^{-1}), Susquehanna (1554 kg km^{-2} yr^{-1}), Schuylkill (1401 kg km^{-2} yr^{-1}), and the Rappahannock (898 kg km^{-2} yr^{-1}). All catchments had some import of N in feed, and averaged 887 kg km^{-2} yr^{-1} for animal feed. Import of N in human food was largest in the Charles (2745 kg km^{-2} yr^{-1}), Blackstone (1279 kg km^{-2} yr^{-1}), Schuylkill (551 kg km^{-2} yr^{-1}), and Merrimack (647 kg km^{-2} yr^{-1}), which are the 4 basins with the highest population densities (see Table 1). Many of the basins with large agricultural production or low population densities export foods to other regions.

Another important source of N was agricultural N fixation. N fixation from leguminous crop species averaged 740 kg km^{-2} yr^{-1} and varied directly with total land area in agriculture. Fixation rates ranged from 74 kg km^{-2} yr^{-1} on the Saco to 1439 kg km^{-2} yr^{-1} on the Rappahannock (Table 6). Alfalfa and other leguminous (e.g. clover) hays accounted for the vast majority of total agricultural fixation inputs (47% and 41%, respectively), with smaller inputs from fixation occurring in the small areas on which soybeans were harvested (6%), from eastern pasture (5%), and from other crops (<1%).

Forest fixation was a small N source to each catchment (Table 6). These estimates were largely dependent on assumptions regarding the abundance and mean N fixation rate of black locust, the main N-fixing tree species in the region. Estimated total forest N fixation rates per catchment ranged from 50 kg km^{-2} yr^{-1} on the Kennebec to 361 kg km^{-2} yr^{-1} on the James, with a mean of 167 kg km^{-2} yr^{-1}. Heterotrophic N fixation accounted for over half of the small amount of N (50–70 kg km^{-2} yr^{-1}) fixed in the northern Maine catchments, whereas symbiotic N fixation associated with black locust contributed over 70% of the N fixed in the Massachusetts, Pennsylvania, and Virginia catchments. N fixation associated with alder was most important in northern Maine, but never exceeded 30 kg km^{-2} yr^{-1}.

Total nitrogen budgets were established by aggregating the N input terms (net deposition, fertilizer use, fixation, and net inputs in food & feed) for each catchment (Table 6). Total new inputs of N to each catchment ranged from

Table 6. Overall nitrogen budget, kg N km^{-2} yr^{-1}

	Net Atmos. pheric N Dep.	Nitro- genous Fertilizer use	N fixation in forest lands	N fixation in agricult. lands	Net N import in food & feed	Total nitrogen inputs	Stream- flow N export*	% of N inputs exported in stream flow	% of N inputs stored or lost in basin
Penobscot	575	91	58	74	36	835	317	38	62
Kennebec	677	54	50	164	154	1099	333	30	70
Androscoggin	779	80	69	146	237	1310	404	31	69
Saco	885	42	107	96	104	1233	389	32	68
Merrimack	921	147	151	213	797	2228	499	22	78
Charles	996	197	218	187	2087	4406	1756	40	60
Blackstone	1040	307	260	305	1495	3407	1140	33	67
Connecticut	962	274	102	360	565	2262	538	24	76
Hudson	1033	204	103	374	271	1985	502	25	75
Mohawk	1075	411	70	1239	624	3420	795	23	77
Delaware	1212	527	201	675	352	2967	961	32	68
Schuylkill	1143	1207	190	1225	1952	5717	1755	31	69
Susquehanna	1138	615	179	1147	1095	4173	977	23	77
Potomac	769	1024	271	1173	1452	4689	897	19	81
Rappahannock	893	1030	277	1439	607	4246	470	11	89
James	953	361	361	703	395	2773	314	11	89
Area-wght. Avg.	*959*	*474*	*167*	*740*	*748*	*3088*	*718*	*25*	*75*

– no data. *Export of N in streamflow for the Charles River was 644 kg N km^{-2} yr^{-1}. The value shown includes 1112 kg N km^{-2} yr^{-1} of wastewater that originated within the Charles watershed but is diverted out of the basin boundary.

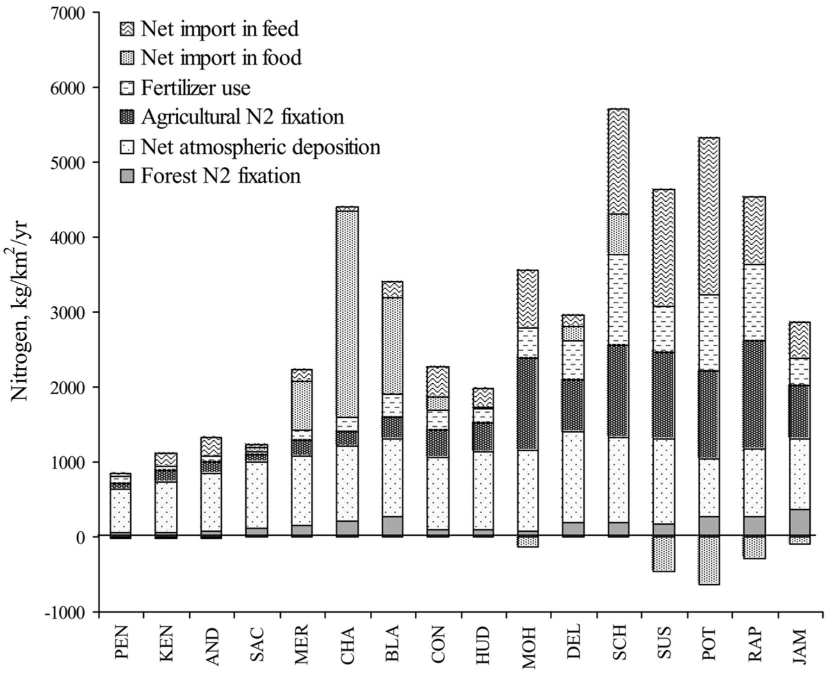

Figure 5. The relative importance of the N sources varies widely by catchment. Net atmospheric deposition was the largest N source (>60%) to the forested basins of northern New England (e.g. Penobscot & Kennebec); net import of N in food was the largest source of N to the more populated regions of southern New England (e.g. Charles & Blackstone); and agricultural inputs were the dominant N sources in the Mid-Atlantic region (e.g. Schuylkill & Potomac).

835 kg km^{-2} yr^{-1} (Penobscot) in the forested northeast to 5717 kg km^{-2} yr^{-1} (Schuylkill) in the agricultural southeast. The magnitude and relative importance of the N inputs vary widely between the catchments (Figure 5). Losses of N in riverine export ranged from 314 kg km^{-2} yr^{-1} on the James to 1,755 kg N km^{-2} yr^{-1} on the Schuylkill, with an average of 747 kg N km^{-2} yr^{-1} (Table 6). R^2 values describing the amount of variance in streamflow N export that can be explained by the variance in the individual N input terms with simple, pairwise regressions were 0.83 for net food & feed imports, 0.33 for atmospheric deposition, 0.21 for fertilizer, and 0.10 for fixation. N loss in streamflow was strongly related to the total N inputs (R^2 = 0.62, Figure 6). However, only 25% of the total N inputs are represented by N losses in streamflow export.

A correlation matrix provides further insight into the relationships between streamflow N loading (of nitrate-N and total N) and individual N inputs (Table 7). Atmospheric deposition terms are more closely correlated

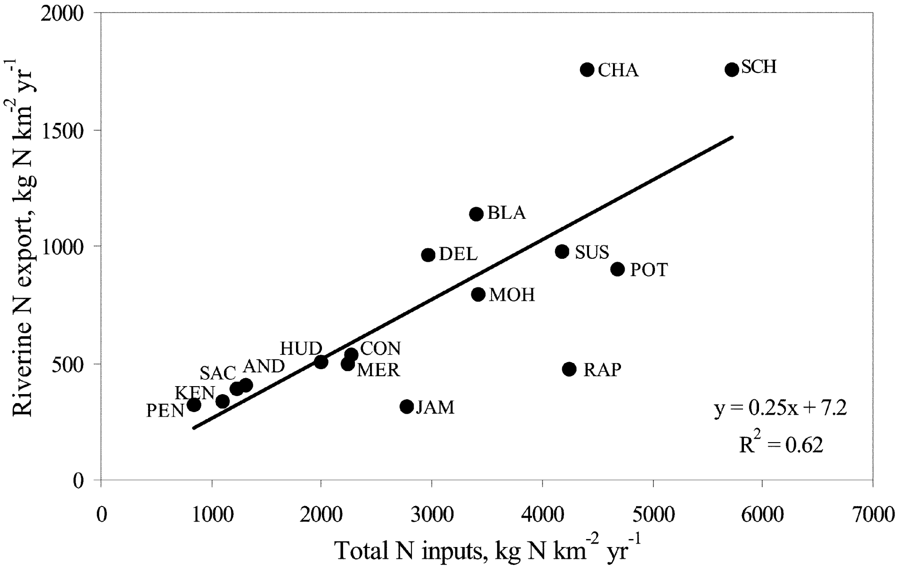

Figure 6. N export in streamflow is strongly related to total new inputs of nitrogen to each catchment.

with nitrate-N loss from the stream than with total-N loss in the stream. NO_y-N deposition is the most highly correlated input term with NO_3^-N export in streamflow (R = 0.82). In contrast, net import of N in food and feed is the most highly correlated input term with total-N export in streamflow (R = 0.91), yet this is less strongly correlated with nitrate-N export in streamflow (R = 0.56).

Discussion

We quantified nitrogen budgets for 16 catchments in the northeastern U.S., comparing new inputs of N from deposition, fertilizer, fixation, and inputs in food & feed to outputs of N transported in streamflow. The importance of the relative N sources varies widely by catchment and related strongly to land use (see Figure 5). These results emphasize that landscape management plans need to be developed on a watershed-by-watershed basis. Given the extreme variability among basins in both the sources of nutrients and controls on their transport, average values of the importance of the individual input terms across broad areas tend to be inappropriate descriptors of the individual catchments. For example, net atmospheric deposition was the most important N source (>60%) to the forested basins of northern New England (e.g. Penobscot and Kennebec); net import of N in food was the largest source of N to the

Table 7. Pearson correlation coefficients (R) relating streamflow N export (as NO_3-N and total-N) to watershed N inputs

	NO_3-N export in streamflow	Total N export in streamflow
NO_y-N dep	0.82	0.70
NH_x-N dep	0.56	0.29
Net AON dep	0.78	0.63
Total net N dep	0.66	0.57
Net import in feed	0.55	0.32
Net import in food	0.11	0.63
Food & feed N import	0.56	0.91
Fertilizer N use	0.70	0.46
Agric. N fixation	0.57	0.30
Forest N fixation	0.22	0.31
Total Agric. N inputs	0.74	0.78
Total N inputs	0.76	0.79
NO_3-N export in streamflow	1.00	0.78
Total N export in streamflow	0.78	1.00

more populated basins in southern New England (e.g. Charles & Blackstone); and agricultural inputs were the dominant N sources to the basins in the Mid-Atlantic (e.g. Schuylkill & Potomac). Over the region covering all of the NE catchments, net atmospheric deposition was the largest single source input (31%), followed by net imports of N in food and feed (25%), fixation in agricultural lands (24%), fertilizer use (15%), and fixation in forests (5%). The combined effect of fertilizer use, fixation in agricultural lands, and food & feed imports (64%) makes agriculture the largest overall source of N to the region. However, it is important to highlight that the net import of N in food and feed accounts for, among other things in the food production cycle, human and animal waste.

Although all 16 of the catchments are predominantly forested, shifts in land use to include even relatively small percentages of agricultural or urban land (see Table 1) have profound impacts on the annual N budgets. Total N inputs have a strong negative correlation with the fraction of land area in forest (Figure 7(a), $R^2 = 0.77$). N inputs increase directly with the fraction of land area in agriculture (Figure 7(b), $R^2 = 0.70$). The two outliers (in Figure 7(b), $\sim$10% agricultural land) are the highly urbanized catchments of the Charles (with a population density of 556 persons per km^2, 8.4% agricultural land use, and 22.2% urban areas) and Blackstone (with a population density of 276 persons per km^2, 8.1% agricultural land use, and 17.6% urban areas). This indicates the importance of urbanization, in addition to agriculture, as a large human-derived source of N to the region. Taking the sum of agricultural and urban lands, there is a direct and strong relationship between these disturbed landscapes and total N loading (Figure 7(c), $R^2 = 0.96$), highlighting the effects of anthropogenic manipulations.

Over the combined area of the catchments, 44% of food and feed requirements had to be supplied from imports from outside of the catchment boundaries. Animal demands exceeded crop production in all of the basins, and import of feed was necessary (see Table 5). Food was imported for human consumption in the New England and northern Mid-Atlantic region (including the Saco, Merrimack, Charles, Blackstone, Connecticut, Hudson, Delaware, and Schuylkill catchments). The more heavily agricultural catchments of the southern Mid-Atlantic region exported food, possibly supporting the demands to the north and to the urban centers below the points of watershed delineation for these analyses. These transfers of N illustrate the de-coupling of production and consumption inherent in many contemporary agricultural ecosystems, requiring the transfer of large quantities of N in food and feed across large distances (e.g. Jordan & Weller 1996). Because the outlets of our 16 catchments are located above many of the large population centers along the east coast, it is expected that the net import of N in food and feed is probably even more important to these larger drainage basins than our results would suggest.

Total N inputs greatly exceeded losses of N in riverine export. The fraction of N inputs represented by riverine export ranged from 11% to 40% and averaged 25%. This result is consistent with the findings of other studies; only a small fraction of N inputs to the landscape are explained by export in streamflow, whether considered at the scale of small catchments (e.g. Campbell et al. 2000), large river basins (Jaworski et al. 1997; Castro et al. 2000), or continents (Howarth et al. 1996). Questions remain about the fate of N that was attenuated by the catchment; i.e. converted to gaseous forms through denitrification, and/or stored in biomass, groundwater, or soils of the landscape. Because most of the nitrogen added to regions through human activity is stored within the region or denitrified, it is critical to understand the other major controls over loss and storage of this N. The role of these processes in the 16 catchments is evaluated in an accompanying manuscript (see Van Breemen et al. 2002).

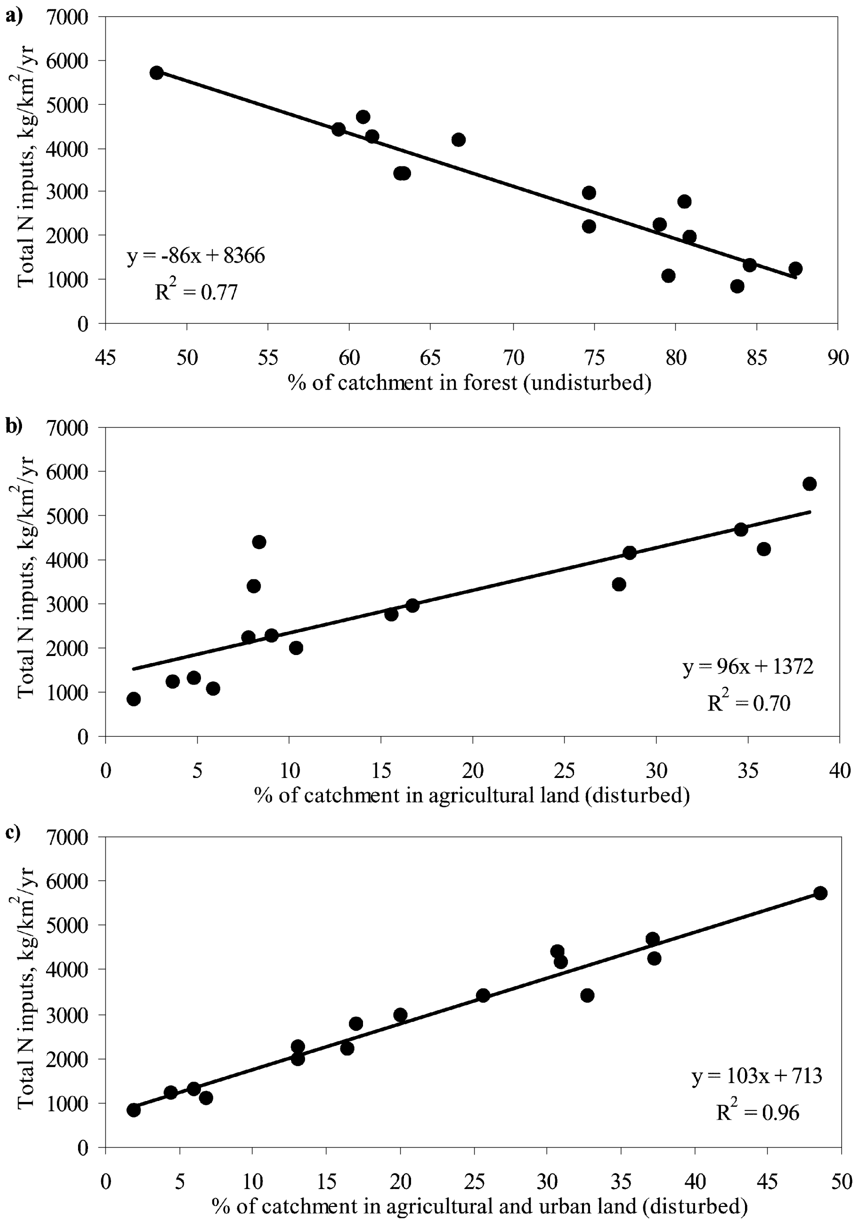

Figure 7. Nitrogen inputs to each catchment are related to land use, having: a negative correlation with land in forest (a); a positive correlation with land in agriculture (b); and a strong positive correlation with urbanized and agricultural lands (c). This highlights the effects of anthropogenic manipulations of the landscape.

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