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Where did all the nitrogen go? Fate of nitrogen inputs to large watersheds in the northeastern U.S.A.

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Abstract. To assess the fate of the large amounts of nitrogen (N) brought into the environment by human activities, we constructed N budgets for sixteen large watersheds (475 to 70,189 km^2) in the northeastern U.S.A. These watersheds are mainly forested (48–87%), but vary widely with respect to land use and population density. We combined published data and empirical and process models to set up a complete N budget for these sixteen watersheds. Atmospheric deposition, fertilizer application, net feed and food inputs, biological fixation, river discharge, wood accumulation and export, changes in soil N, and denitrification losses in the landscape and in rivers were considered for the period 1988 to 1992. For the whole area, on average 3420 kg of N is imported annually per km^2 of land. Atmospheric N deposition, N_2 fixation by plants, and N imported in commercial products (fertilizers, food and feed) contributed to the input in roughly equal contributions. We quantified the fate of these inputs by independent estimates of storage and loss terms, except for denitrification from land, which was estimated from the difference between all inputs and all other storage and loss terms. Of the total storage and losses in the watersheds, about half of the N is lost in gaseous form (51%, largely by denitrification). Additional N is lost in riverine export (20%), in food exports (6%), and in wood exports (5%). Change in storage of N in the watersheds in soil organic matter (9%) and wood (9%) accounts for the remainder of the sinks. The presence of appreciable changes in total N storage on land, which we probably under-rather than overestimated, shows that the N budget is not in steady state, so that drainage and denitrification exports of N may well increase further in the future.

Introduction

Through processes such as manufacturing fertilizers, burning fossil fuels, and cultivating crops that fix nitrogen (N) symbiotically, humans have greatly accelerated the fixation of atmospheric N to plant-available forms (Galloway et al. 1995; Vitousek et al. 1997). As a result, the amount of reactive N that enters the biosphere each year worldwide has roughly doubled since pre-industrial times (Galloway et al. 1995; Howarth et al. 1996; Mosier et al. 2001). Because N is the second-most abundant plant nutrient (after CO_2) and limits primary production in many terrestrial, freshwater and near-coastal marine ecosystems, these anthropogenic activities have major environmental consequences (Vitousek et al. 1997).

Although the amounts of N fixed from natural and human activities have received much attention, the fate of this reactive N is poorly understood. Several recent budget studies have presented estimates of all known new N inputs to terrestrial landscapes. For example, Boyer et al. (2002) quantified N inputs to individual watersheds that drain to the northeast (NE) coast of the U.S.A., following the methodology put forth by Howarth et al. (1996) who estimated N inputs to all of the large regions that drain to the North Atlantic Ocean, including one value for the NE region as a whole. Despite the large differences in scale, both studies indicate that streamflow exports account for only about 25% of N inputs to the landscape (Figure 1). Regardless of how the input terms and boundary conditions are defined, nearly all such studies conclude that only 20–60% of N inputs are explained by N export in streamflow, whether considered at the scale of small watersheds (e.g. Campbell et al. 2000; Dise et al. 1998), large river basins (e.g. Jaworski et al. 1997; Castro et al. 2001), or regional drainage areas (e.g. Howarth et al. 1996). The N input in excess of riverine export has been termed the 'missing nitrogen', highlighting the uncertainty in the scientific community of how to quantify the fate and transport of N in the landscape. The fraction of N 'consumed' by the landscape and not delivered to streamflow is partly stored in pools with residence times exceeding decades to centuries (soil, wood, groundwater) and partly returned to the pool of highly inert atmospheric N_2 by denitrification. The relative sizes of these storage and loss terms of the N budget are highly uncertain. The aim of this paper is to increase our understanding of the amounts of N that are stored in these different slow but potentially reactive pools and that are lost to the atmosphere due to N transformation processes.

We chose 16 watersheds in the northeastern U.S.A. (Figure 2, taken from Boyer et al. 2002) for our analyses, a region where N cycling is of particular importance due to problems in coastal waters caused by over-enrichment of N (Bricker et al. 1999). Further, the availability of high-quality, long-term

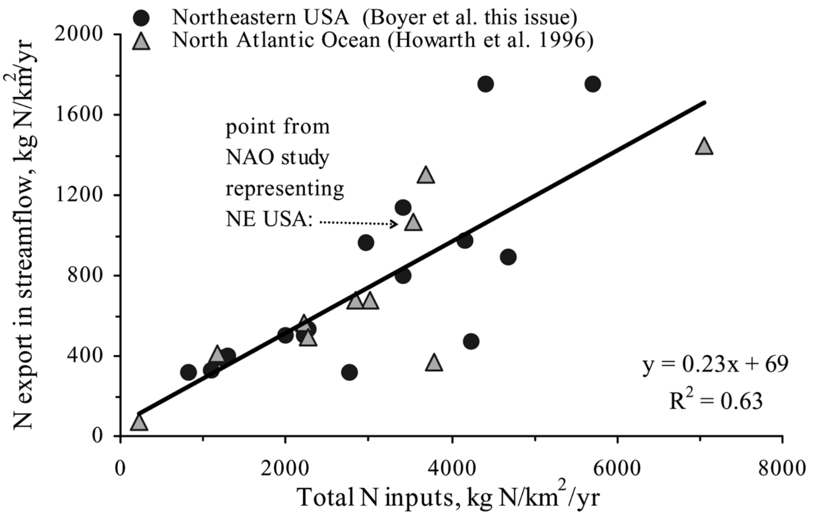

Figure 1. Nitrogen budget studies for large watersheds in the northeast (NE) U.S.A. (Boyer et al. 2002) and for regions in north America and in western Europe that drain to the north Atlantic Ocean (NAO, Howarth et al. 1996). Total N inputs are the net anthropogenic inputs from atmospheric deposition, fertilizer inputs, nitrogen fixation, and the net import of N in food & feed. The NAO regional analysis represents the entire NE U.S.A. as one region.

monitoring data in the northeast allowed us to quantify, with good confidence, N inputs to these regions as a starting point for investigating storage and losses of N in the landscape. The details of our calculations of N inputs and riverine exports for each watershed are presented in a companion paper (see Boyer et al. 2002). Nitrogen budgets were established by quantifying total annual inputs of N to each catchment. Most of the N inputs are derived from human activities, and include atmospheric deposition, fertilizer use, net imports (or exports) in food & feed, and biological fixation in agricultural areas and in forests. As shown in Figure 1, riverine export of N was well correlated with N inputs, but represented only a fraction (11–40%) of the total N inputs. The unresolved N inputs in excess of streamflow export are either stored (e.g. in vegetation, soil, or groundwater) or lost (e.g. denitrified, volatilized, or exported in transfers of commodities) in the watershed. In this paper, we attempt to close input-output N budgets for these watersheds by explaining the fate of the N inputs. We estimate storage and loss terms from a combination of statistical and process models and with data on land use change.

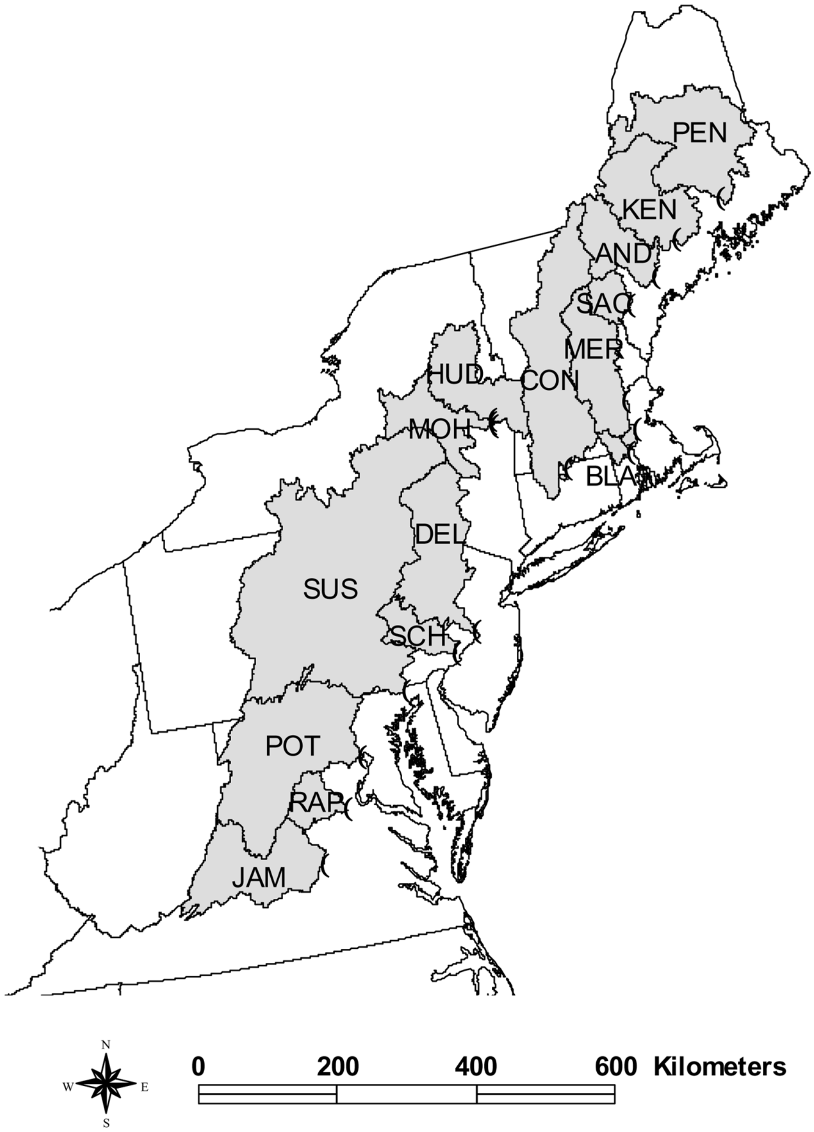

Figure 2. (from Boyer et al. 2002). Location of 16 watersheds draining to the northeast US coast. Watershed boundaries are delineated upstream of USGS stations (denoted with black circles) where streamflow and water quality characteristics were measured.

Methods

We begin with N inputs to 16 watersheds in the northeastern U.S.A., presented by Boyer et al. (2002), and refer to that paper for a detailed description of watershed characteristics, data sources, and methods used to estimate N inputs and riverine export. In brief, the watersheds provide the major drainage ways to the northeast coast, and are located in a latitudinal profile from Maine to Virginia. The basins range in size from 475 km^2 to 70,189 km^2. The combined total area of all watersheds is largely forested (72%) with some agricultural land (19%) and a small fraction of urban land (3%) (Table 1). Because we delineated the watershed boundaries upstream of suitable gaging stations (from which riverine streamflow and water quality data were obtained), most major coastal population centers are excluded. Data presented in this paper are representative of the early 1990's and, where possible, reflect average values over the period 1988–1993.

Sources of N to the 16 watersheds include atmospheric N deposition, nitrogenous fertilizer use, import of N in food & feed, and biological N fixation in crops and in forests (Table 2, after Boyer et al. 2002). The goal of this paper is to explain the fate of these N inputs. We take a mass balance approach to establish a complete N budget for each watershed, where inputs are balanced by outputs or changes in storage in the watershed. We utilize a conceptual model whereby N inputs are routed through one of the 4 major land use 'reservoirs' comprising the watershed: forested, agricultural, sub/urban, and water ecosystems (Figure 3). We establish a mass balance of N for each of the individual ecosystems, and aggregate their inputs and outputs to determine the total storages and losses from the watershed. In some cases, outputs from one ecosystem are inputs to another, transferring N internally but having no net effect on watershed output. In addition to the N exported in rivers, outputs of N from each watershed include removal in harvested wood, exports of food, ammonia volatilization losses, and gaseous losses due to denitrification (from sewage and waste water, from soil solutions in transit from soils to rivers, and within the water column). Changes in storage include the net accumulation of N in vegetation, in soil, and in groundwater.

Storage and losses in forest lands

Inputs of N to forested lands include atmospheric deposition and fixation, while losses include removal in harvested wood and denitrification (see Figure 3). Changes in storage include net accumulation in woody biomass, in dead wood and in green plant tissues (foliage & fine roots), and in forest soils. While not a loss when considered at the scale of the watershed, the internal transfer of N from the forest ecosystem to subsurface water reservoirs (e.g.

Table 1. Watershed Characteristics*

Watershed	Abbreviation	Area km^2	Sewer. Popul. $\#/km^2$	Unsew. Popul. $\#/km^2$	Temper. °C	Precip. mm yr^{-1}	Runoff yr^{-1}	Land Forest %	Land Agric. %	Land Urban %	Land Wat & Wetl. %	Land Other %
Penobscot	PEN	20109	3.0	4.5	4.3	1075	588	84	1	0	11	3
Kennebec	KEN	13994	4.4	4.8	4.3	1085	566	80	6	1	10	4
Androscoggin	AND	8451	8.2	8.6	4.6	1151	640	85	5	1	8	1
Saco	SAC	3349	7.6	8.7	5.8	1218	672	87	4	1	7	1
Merrimack	MER	12005	94.7	47.9	7.4	1148	589	75	8	9	8	1
Charles	CHA	475	428.9	127.4	9.7	1207	583	59	8	22	10	0
Blackstone	BLA	1115	217.5	58.5	9.0	1260	651	63	8	18	10	1
Connecticut	CON	25019	44.0	20.8	6.3	1160	642	79	9	4	7	1
Hudson	HUD	11942	19.7	12.6	6.6	1126	622	81	10	3	6	0
Mohawk	MOH	8935	32.3	22.0	6.8	1142	548	63	28	5	4	0
Delaware	DEL	17560	55.1	30.0	8.7	1131	547	75	17	3	5	0
Schuylkill	SCH	4903	249.9	42.8	10.6	1134	488	48	38	10	2	1
Susquehanna	SUS	70189	33.1	20.9	8.9	1022	487	67	29	2	2	1
Potomac	POT	29940	46.7	15.9	11.3	985	328	61	35	3	1	1
Rappahannock	RAP	4134	10.1	14.2	12.6	1045	360	61	36	1	1	1
James	JAM	16206	10.1	14.0	10.1	934	407	81	61	1	*1*	1
Area-wt. avg	—	*32666*	*38.0*	*19.0*	*8.0*	*1068*	*515*	*72*	*19*	*3*	*5*	*1*

*From Boyer et al. 2002. Watersheds are arranged by latitude of their outlets, from north to south.

Table 2. Sources of N to watersheds* (kg N per km^2 per year)

Water-shed	Total NO_y dep.	Total NH_x dep.[1]	Net Org. N dep.	N fertilizer use	Forest N fixation	Agricul. N fixation	Net N import in feed	Net N import in feed	Total Sources
PEN	362	129	88	91	58	74	55	0	857
KEN	428	154	105	54	50	164	171	0	1126
AND	495	176	121	80	69	146	247	0	1332
SAC	566	187	136	42	107	96	49	55	1237
MER	606	184	142	147	151	213	150	647	2240
CHA	674	178	153	197	218	187	62	2745	4415
BLA	707	190	162	307	260	305	217	1279	3426
CON	631	204	150	274	102	360	398	167	2286
HUD	658	234	161	204	103	374	251	20	2005
MOH	708	250	172	411	70	1239	758	0	3610
DEL	811	248	191	527	201	675	155	197	3005
SCH	885	253	205	1207	190	1225	1401	551	5917
SUS	816	269	195	615	179	1147	1554	0	4774
POT	714	255	174	1024	271	1173	2085	0	5696
RAP	615	256	157	1030	277	1439	898	0	4671
JAM	652	237	160	361	361	703	487	0	2961
Wt. Avg.	*677*	*288*	*163*	*474*	*167*	*740*	*887*	*86*	*3420*

*From Boyer et al. 2002. [1]Rather than using a net input term, wer treat depositional inputs of total NH_x and volatilization losses of NH_x separately.

groundwater) and to the river affects the input of N to these other ecosystems. We estimate all of the N fluxes in forested ecosystems (Table 3). The forest calculations for each watershed are presented in detail in a companion paper by Goodale et al. (2002). Forest inventory data were used to estimate changes in biomass and harvest export, and an ecosystem model was used to estimate changes in dead wood, green plant tissues, forest soils, and leaching of nitrate below the rooting zone.

Briefly, county-level data were obtained from the U.S. Forest Service's Forest Inventory and Analysis (FIA) program (Hansen et al. 1992) on the volume of wood growth, mortality, and harvests in eastern forests. These values were converted to estimates of biomass with two different approaches, and multiplied by literature-derived estimates of the N content in wood (0.19 + 0.08% for softwoods and 0.26 + 0.06% for hardwoods) to obtain estimates in terms of N. Values included in our study are the mean of the two biomass conversion approaches. Net N accumulation in biomass consists of growth

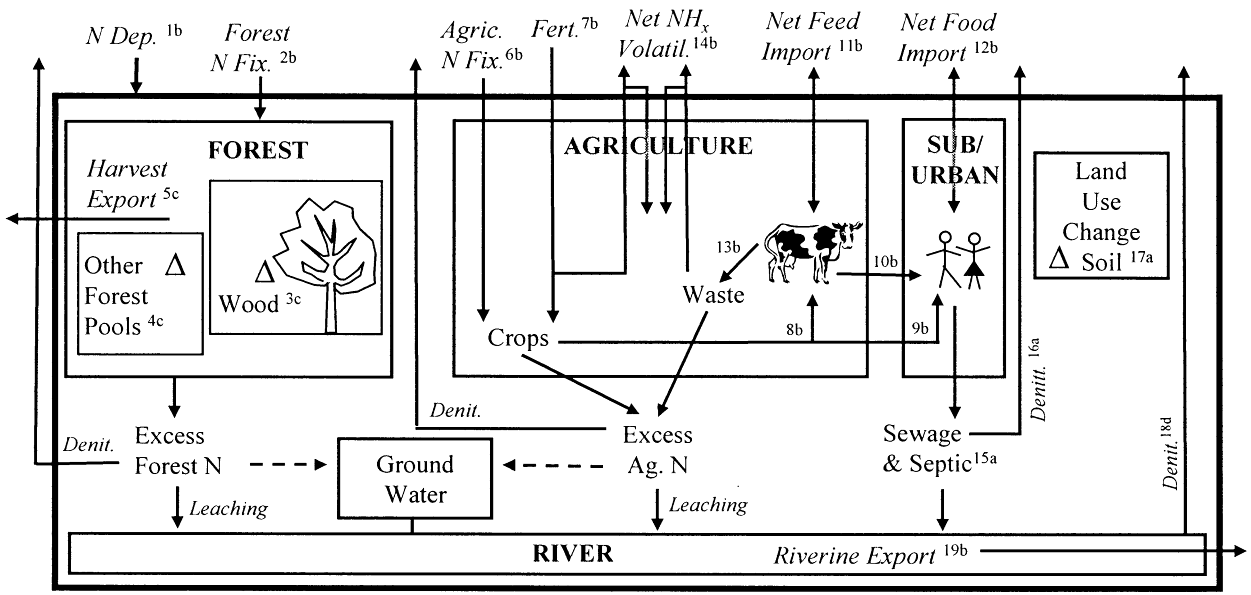

Figure 3. N fluxes to, from, and within each watershed: (1) NH_x, NO_y, and net organic N atmospheric deposition to whole watershed, (2) biological N fixation in forests, (3) increase in N stocks in woody biomass and (4) other forest pools (dead wood, forest soils, green tissues) (5) export of harvested wood, (6) biological nitrogen fixation in agricultural lands, (7) fertilizer use, (8) crop production for animal consumption and (9) for human consumption, (10) meat, milk and eggs, (11) Net import (or export) of animal feed and (12) human food, (13) production of animal waste, (14) Net NH_3 volatilization from animal waste and fertilizer, (15) sewage & septic waste and (16) denitrification during treatment, (17) increase in soil N due to land-use change, (18) in-stream denitrification, and (19) riverine export. Letters indicate the source of calculations: (a) this paper, (b) Boyer et al. (c) Goodale et al. (d) Seitzinger et al.

Table 3. N balance for forest land (kg N per km^2 of forest land per year)

Water-shed	% of land in forest	N Sources			N Storages and Losses							
		Atmos-pheric dep.	Forest N fixation	Total N inputs	Wood biomass storage[1]	Other forest storage[2]	Change in soil storage[3]	Harvest export[4]	NO_3 leaching	DON leaching	Total storage & loss	% inputs denitr. in soils[5]
PEN	84	579	70	649	–205	–6	–99	491	256	75	513	21
KEN	80	687	63	750	–220	–6	–96	524	312	75	589	21
AND	85	791	81	873	184	–103	55	313	259	75	783	10
SAC	87	889	123	1012	379	–113	191	181	218	75	931	8
MER	75	932	202	1134	267	–106	328	184	159	75	907	20
CHA	59	1006	368	1374	415	113	158	213	141	75	1115	19
BLA	63	1059	411	1470	263	131	98	257	174	75	997	32
CON	79	985	129	1114	385	–28	123	202	265	75	1021	8
HUD	81	1053	128	1181	426	–100	413	211	189	75	1212	–3
MOH	63	1131	111	1242	469	–40	357	130	222	75	1212	2
DEL	75	1250	269	1518	721	66	133	148	203	75	1346	11
SCH	48	1342	396	1738	649	–111	202	286	223	75	1325	24
SUS	67	1280	268	1548	550	167	123	215	179	75	1309	15
POT	61	1143	446	1589	570	182	69	193	231	75	1320	17
RAP	61	1027	451	1479	473	38	40	363	185	75	1173	21
JAM	81	1049	448	1497	392	200	–117	384	237	75	1171	22
Wt. Avg.	*72*	*1067*	*242*	*1309*	*395*	*66*	*102*	*259*	*218*	*75*	*1116*	*15*

[1,2,3,4]from Goodale et al. 2002. Change in storage in: [1]living wood biomas, [2]dead wood and live plant tissues, and [3]in forest land soils. [4]Removal of forest products from watershed. [5]In forest lands; estimated by difference of inputs – outputs.

minus losses due to natural mortality and harvests. Harvest export consists of only the harvested biomass removed from the forest; the rest of the harvested material is assumed to decompose within the watershed.

Accumulation of N in dead wood, green plant tissues, and soils was estimated with the forest ecosystem model PnET-CN (Aber et al. 1997; Aber & Driscoll 1997) as the balance between inputs from litter and logging slash, and losses from decomposition. Simulations were performed for a range of forest age classes as indicated by forest inventory data, and for two contrasting scenarios past land-use history: agriculture and forestry. Model-based N fluxes included here used the mean simulation results from these two land-use histories, weighted by the approximate prevalence of each history on each watershed.

Leaching of N from soils is significant in regions with excess N input, as in the northeastern U.S.A. Leaching losses of nitrate below the rooting zone were also estimated with the PnET-CN model. In the model structure, net mineralization from the soil is available for uptake by plants; the excess can nitrify and leach below the rooting zone with drainage water. Predicted losses of nitrate depended on the combined effects of N deposition, and both short- and long-term effects of disturbance (see Goodale et al. 2002). DON leaching losses from forested lands were estimated from literature values. In the northeastern U.S.A., DON losses to streams range from 30 to 240 kg km^{-2} yr^{-1}, and average about 75 kg km^{-2} yr^{-1} (Campbell et al. 2000; Lovett et al. 2000; Goodale et al. 2000). We assumed that the forested areas in each catchment leach 75 kg of DON km^{-2} yr^{-1} to streamwater. This conservative, constant estimate allows us to account for this important removal pathway while not biasing the pattern of residual uncertainties in our N budgets.

Some of the leached N is lost before the drainage water reaches rivers or groundwater, of which only part can be explained by plant uptake or retention in soils (Lajtha et al. 1995; Sollins & McCorison 1981). There is evidence for denitrification in the vadose zone or at the terrestrial-aquatic interface that is difficult to quantify (Montgomery et al. 1997; Valiela et al. 1997; Seely et al. 1998). Denitrification from well-drained, upland forest soils in the northeastern U.S.A. is generally very low (Groffman & Tiedje 1989; Bowden 1986). Overall, the forest ecosystem N balance was well constrained, with the estimated storage and loss terms accounting for approximately 85% of the inputs. The difference between N inputs and outputs, or the unresolved N, is attributed to denitrification or to changes in groundwater storage.

Storage and losses in sub/urban and agricultural lands

Sources of N to sub/urban lands include atmospheric deposition and food inputs. Food inputs come from two sources: from agricultural lands within the watershed (i.e. local crop and animal production of fruits, vegetables, meat, milk and eggs), and from imports of N from outside of the watershed (see Figure 3). Change in soil storage is potentially a significant sink for N. Watershed-scale losses of N from sub/urban lands include net export of N in food and denitrification of N during waste treatment.

Estimates of losses in food exports are presented by Boyer et al. (2002) in their discussion of the net import of food and feed to each watershed. About 1/2 of the watersheds produced more N in crop and animal products than could be consumed by the populations living there, and thus exported N in food sales to other regions (see Table 2). We calculated sewage treatment losses by comparing human consumption of N in food with data on N delivered to rivers in wastewater discharge. Data on populations with sewered waste were obtained from the 1990 U.S. Census (U.S. Dept. of Commerce 1990), and measurements of wastewater discharge and N concentration were obtained from a variety of agency reports (N Jaworski and L Hetling, personal communication). The correlation between the sewered population and wastewater discharge (r^2 = 0.95) yielded a mean per capita load of 3.1 kg N yr^{-1} per person (Figure 4). This is similar to the value of 3.3 kg N yr^{-1} per person reported by Meybeck et al. (1989). The difference between the per capita waste excretion and the per capita intake of 5 kg N yr^{-1} per person (Garrow et al. 2000), or 1.9 kg N yr^{-1} per person, is the amount of N that is either retained or, more likely, denitrified during sewage processing. We estimated total watershed losses from septic and sewage treatment by assuming the same per capita N loss from septic tanks as for the sewered population (1.9 kg N yr^{-1} per person). Although septic treatment is likely to retain N longer in the watershed than sewage treatment, the error introduced by this assumption is probably small because of the relatively low unsewered population (19%, see Table 1).

The northeastern U.S.A. has been experiencing rapid changes in land use, which could affect N storage in litter and soil organic matter stocks in all ecosystems (see Figure 3). Our analysis of data on land use change reveals that between 1982 and 1992 none of the watersheds gained land in agriculture, several gained small percentages of forest land, and all gained substantial amounts of sub/urban land (Figure 5). Therefore, we expected that changes in N in soil stores in sub/urban land uses to be significant factor in our N budgets, associated with the land use change itself (e.g. a shift from forest to urban land) or from land use legacies (e.g. land that had been previously fertilized that is not yet in steady state). Data on land use change were used to estimate net changes in soil organic N stocks using a modified version of the IPCC (Intergovernmental Panel on Climate Change) soil carbon (C) inventory procedure (IPCC 1997, Paustian et al. 1997a). The method employs

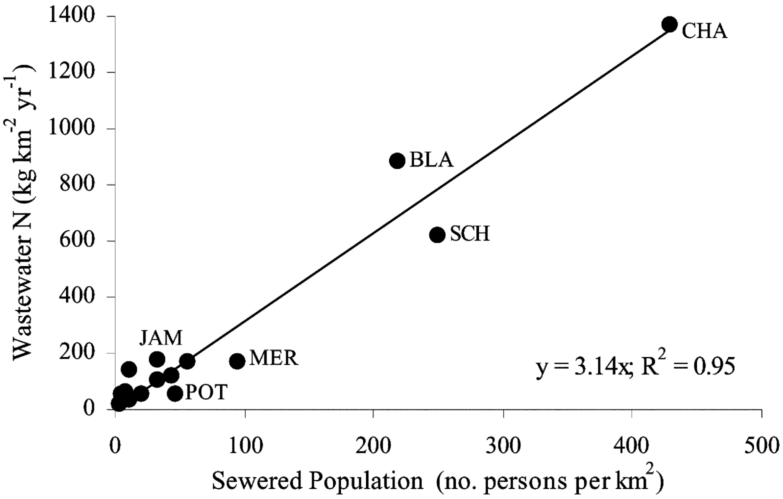

Figure 4. Relationship between sewered population and nitrogen fluxes due to sewage wastewater. The regression line indicates a per capita load in wastewater of 3.1 kg N yr^{-1} per person.

a net stock change approach that incorporates changes in areas according to multiple categories of land use and agricultural management systems, stratified by soil type and climate. Changes in soil C stocks in the upper 30 cm of the soil profile, which result from both land use change and management changes within continued agricultural use, were estimated using a series of coefficients based on climate, soil type, disturbance history, tillage intensity and C input rate (productivity).

We obtained data from the 1982 and 1992 National Resources Inventory (NRI), maintained by USDA's Natural Resources Conservation Service, to estimate land use and soil organic C and N changes. The NRI is a nationwide inventory of land cover and land use and management, comprised of >800,000 permanent sampling locations (Nusser & Goebel 1997). The inventory's statistical design includes an 'expansion factor' for each point location, which is used to estimate the total area represented by the point as specified by the hydrologic unit code (HUC) containing the point. A total of 55,289 NRI points representing 250,000 km^2 were located within the boundaries of the study watersheds.

Land use/land cover types included forest, agricultural, urban, rangeland, miscellaneous and non-cropland (includes abandoned, non-forested agricultural lands, non-forested wetlands). Within agricultural lands, crop rotations were grouped into several types of management systems (e.g. irrigated cropland, continuous row crops, row crop-small grain rotations, row crop-hay

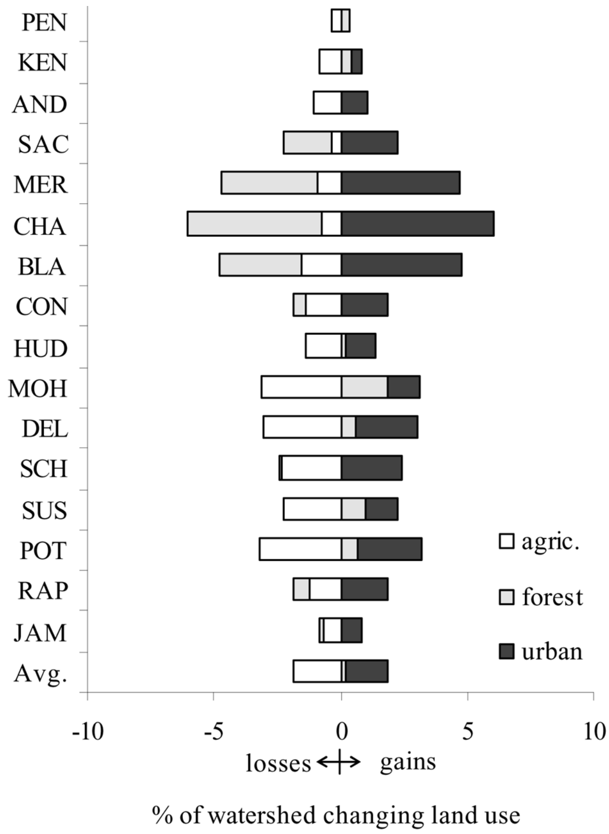

Figure 5. Changes in land use from 1982 to 1992 are shown as a percent of the land area of each watershed. Over the decade, all watersheds lost land in agriculture and gained urban land. Data are from the USDA-NRCS 1992 National Resources Inventory.

rotations, small grain-hay rotations, and vegetable crops), based on the cropping history information in NRI. Crop rotations were then aggregated into three groups, i.e. high, medium and low input systems, based on the rates of production and return of organic residues to soil. Management systems were further stratified into three categories of tillage: conventional, reduced and no-till. Data from the CTIC (Conservation Tillage Information Center) (www.ctic.purdue.edu; Dan Towery, personal communication) were used to estimate the areas under the three tillage management scenarios in 1982 and 1992. Areas for each of the land use/management systems were then calculated by watershed, for 1982 and 1992. To account for our use of a 10 year inventory cycle instead of the default 20-year IPCC inventory period, we changed the coefficients to yield only 50% of the expected change in C stocks over a twenty year period. Changes in organic N stocks were estimated from the C calculations, assuming a C:N ratio of 10 for agricultural and 15 for urban/suburban soils and early successional forests converted from agricultural use (Robertson & Vitousek 1981; Hamburg 1984; Zak et al. 1990).

Inputs of N to the agricultural ecosystem include atmospheric deposition, fixation, fertilizer, and net import (or export) in food & feed (see Figure 3). As in the sub/urban system, changes in soil N storage are potential N sinks. We calculate the change of N stored in agricultural soils due to land use change according to the IPCC methodology described above. Watershed-scale losses of N from agricultural lands include volatilization, feed exports, and denitrification. Internally, there are many important transfers. For example, a fraction of the crops produced and animal products (meat, milk & eggs) are outputs from the agricultural ecosystem but are input as food to the sub/urban ecosystem, thus having no net effect on the watershed-scale budget. We estimate both the internal and external cycling in agricultural lands (Table 4).

Estimates of net NH_3 volatilization losses are presented by Boyer et al. (2002) in their discussion of depositional inputs. Calculations of losses from each watershed in feed exports from agricultural lands are also presented by Boyer et al. (2002) in their discussion of net food & feed imports. None of the watersheds had net exports of N from agricultural lands in animal feed (see Table 2).

Leaching losses from applied N in agricultural fields to ground and surface water are difficult to quantify and depend on many factors. These losses increase with the amount of applied N and are generally higher in arable fields than in grasslands. Using literature values, we assumed that leaching losses from grassland equal $0.15 \times$ (N input – 500) kg km^{-2} yr^{-1} (Jordan et al. 1994; Scholefield et al. 1988; Magesan et al. 1996). Leaching from arable land was assumed to be $0.2 \times$ (N input – 500) kg km^{-2} yr^{-1} (Goss et al. 1988; Madramootoo et al. 1995; Watson et al. 1993; Wyland et al. 1996; Shephard 1992). N fertilizer inputs are normally somewhat higher on arable land than on pasture land, but varying the relative fertilizer application rates between 1 and 3 times higher on arable than on pasture land had no marked (<1%) effect on the estimated NO_3 leaching from agricultural land.

Denitrification in agricultural soils can be very significant, particularly in areas with high inputs of N fertilizers (Velthof et al. 1997). We estimated denitrification by difference between N total N inputs and the outputs from agricultural lands (soil N storage, NH_3 volatilization, food production removals, and NO_3 leaching). We attribute this budget discrepancy to denitrification, which appears to make up a significant fraction ($\sim$50%) of the N inputs to the agricultural ecosystem (Table 4).

Table 4. N balance for agricultural land (kg N per km^2 of agricultural land per year)

Water-shed	% of land in agric.	N Sources					N Storages and Losses						
		Atmos-pheric dep.	Fertilizer use	Agricul. N fixation	Net import in feed	Total N inputs	Crop products for food	Animal products for food	net NH_x vol.[1]	NO_3 leaching[2]	Change in soil storage[1]	Total storage & loss	% inputs denitr. in soils[4]
PEN	2	579	4550	3694	2726	11549	1581	1347	208	1779	901	5816	50
KEN	6	687	900	2727	2852	7165	34	1131	166	1095	1493	3918	45
AND	5	791	1600	2913	4936	10241	172	1888	253	1583	1998	5894	42
SAC	4	889	1050	2389	1224	5551	104	633	103	782	2062	3683	34
MER	8	932	1896	2749	1930	7507	23	921	146	1171	902	3164	58
CHA	8	1006	2343	2229	740	6318	8	477	115	1044	1831	3474	45
BLA	8	1059	3791	3764	2675	11290	28	1363	238	1902	680	4212	63
CON	9	985	3041	3999	4412	12437	90	1838	265	1969	945	5107	59
HUD	10	1053	1958	3586	2410	9008	62	1439	194	1556	1462	4713	48
MOH	28	1131	1469	4431	2711	9742	52	1553	199	1767	1104	4674	52
DEL	17	1250	3155	4044	929	9377	190	1313	222	1678	1038	4442	53
SCH	38	1342	3144	3191	3650	11328	267	2346	520	2062	1757	6951	39
SUS	29	1280	2155	4018	5446	12899	175	2642	498	2360	1095	6771	48
POT	35	1143	2961	3391	6028	13522	154	2866	1080	2097	1302	7499	45
RAP	36	1027	2873	4014	2504	10418	95	1173	375	1949	993	4585	56
JAM	16	1049	2312	4503	3116	10981	38	1462	616	2066	1031	5213	53
Wt. Avg.	*19*	*1067*	*2504*	*3727*	*3979*	*11278*	*243*	*1954*	*431*	*1921*	*1167*	*5716*	*49*

[1]Volatilized NH_x from fertilizers and animal waste that is transported outside of the watershed. [2]Subsurface leaching to ground water and to surface water. [3,4]In agricultural lands. [4]Estimated by difference of inputs – outputs.

Storage and losses in rivers

Inputs to the riverine ecosystem are made up of subsurface flow leached from agricultural, sub/urban, and forested landscapes, plus the very small fraction of atmospheric deposition that falls on the areas of water (see Figure 3). Watershed losses of N include riverine export and in-river denitrification losses. Calculations of N discharged from each watershed in riverine export are presented in Boyer et al. (2002).

The removal of N in the water column itself, which we attribute wholly to denitrification, is detailed in a companion paper by Seitzinger et al. (2002). Briefly, we estimated N loss in rivers using the robust inverse statistical relation observed with the ratio of water depth to water residence time in lakes or river stretches, integrated over whole river systems (Seitzinger et al. 2002). Both residence time of water and flow depth are surrogates to describe flow conditions, with high flows having higher depths of water and faster travel times. Under high flow conditions, there may be less settling of particulate N and less exchange with the subsurface sediments and hyporheic zone (Alexander et al. 2000). In general, the reduced contact times of N transported in streamflow that occur under high flow conditions result in less N loss to denitrification. To scale in-stream removal rates from short river stretches to whole watershed river systems, Seitzinger et al. (2002) used EPA-USGS reach network files for the sixteen watersheds, and their associated attributes describing depth and time of travel of each stream reach. The N loss values were calculated for each individual reach according to the inverse relationship between loss and flow conditions, then losses from all the reaches encompassing a river network were aggregated to provide a total loss estimate for each watershed. We estimate total edge-of-stream loading inputs to each river as the sum of the (insignificant) N deposition occurring on water areas plus the total N leaching losses calculated in the ecosystem budgets for agricultural, sub/urban (wastewater), and forest lands. We use the RivR-N model estimates of Seitzinger et al. (2002) based on the RF1 dataset, indicating the percentage of N inputs that are removed during transport through the river network, to estimate in-stream denitrification for each basin.

Results and discussion

Estimates of total watershed N inputs (Table 2) averaged 3420 kg N km^{-2} yr^{-1}. Atmospheric deposition was the largest single source input (31%), although the combination of N inputs from imports in food in feed (28%), by N_2 fixation in agricultural lands (22%), and from fertilizer inputs (14%) made agriculture the largest total source of N. Nitrogen fixation in forests contributed little (5%). (Note: Our presentation of the relative importance of the N input terms is slightly different than is reported for the same N sources in the accompanying manuscript by Boyer et al. They compare each input term to the total *net* anthropogenic N inputs. To facilitate our analysis of watershed N losses, we treat volatilization losses of ammonia and food exports as outputs from, rather than negative inputs to, each watershed, which accounts for the differences in the input terms between our papers). Total N inputs increase from around 1000 kg N km^{-2} yr^{-1} in the northern watersheds to between 2000 to 6000 kg N km^{-2} yr^{-1} in the southern watersheds. This increase is due mainly to increasing proportions of agricultural and urban land, at the expense of forested land, and the associated increase in N_2 fixation, and inputs of fertilizer and food and feed. In the three most densely populated, relatively small, watersheds (Charles, Blackstone & Schuylkill) net food and feed imports form the dominant N input category. Atmospheric deposition increased from north to south, but this increase in N inputs was small relative to that from agricultural and urban activities.

Aggregating the total N inputs to and outputs from the forested, agricultural, sub/urban, and riverine ecosystems yields a complete accounting of sources, storages, and losses occurring in the 16 watershed (Table 5). N inputs to each watershed are lost (as N removals or gaseous losses) or cause changes in storage within the system Loss terms make up, on average, 82% of the total sinks with the remaining 18% being stored in the watersheds.

Losses due to N removals

Riverine export

As we knew at the onset of this study, riverine export accounts for only a fraction of the total N inputs to each watershed, on average 20% of total N inputs. Our estimates of N inputs according to our riverine ecosystem N budget (see Figure 3) includes the (insignificant) fraction of N inputs from atmospheric deposition that lands on areas of water plus leaching of N from forest, agricultural, and sub/urban (wastewater) systems. These N inputs to the riverine ecosystem are well correlated with N losses in riverine export and with the sum of N discharged plus N removed in rivers due to denitrification (Figure 6). Although the rates of N drained from agricultural and forested areas are indirect model estimates, the N inputs to each river from wastewater discharge are based on independent measurements and are presumably reliable. Total N discharged from the watersheds in riverine export is about 84% of the N inputs to the river from N drainage from the landscape.

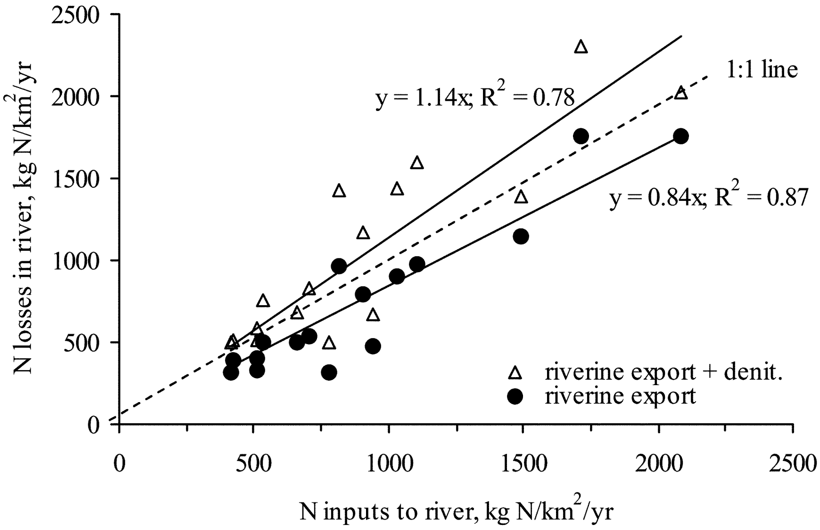

Figure 6. Total N inputs to the river from leaching of N from forested, agricultural, and urban (wastewater) landscapes, compared to the amount of N discharged from the watershed (export) and the sum of N discharged plus N denitrified in rivers (export + loss).

Export in commodities

N exports in commodities, including food products (6%, from removals in meat, milk & eggs) and forest products (5%, from removal of harvested timber) are small loss terms. However, these values do not adequately reflect the importance of commodity exports in individual watersheds (see Table 5). There were no net exports of N in food from most basins, and only in the Susquehanna, Rappahannock, and Potomac were the food exports significant fractions of outputs ($\sim$10%). In watersheds with the largest percentage of forested lands, harvest exports were among the dominant export terms. In the Penobscot, Kennebec, and Androscoggin watersheds, each supporting industrial timber production, exports of N in the forest harvest accounted for 44, 37, and 19% of the total outputs, respectively. We treated the entire wood harvest, which accounted for 5% of the losses on average, as an export term. However, some of the harvested wood will remain in the watershed for local use, and will decay at rates in the order of 0.1 to 0.005 yr^{-1} (Harmon et al. 1996), so this loss term may have been overestimated.

Gaseous losses

Denitrification in rivers
The relationship between N inputs to the river from leaching of N from forested, agricultural, and urban (wastewater) landscapes, and river N export (Figure 3) leaves room for denitrification inside the river system, but not as much as indicated by the analysis by Seitzinger et al. (2002). The sum of N discharged in riverine export plus N removed in the rivers via denitrification exceeds the estimated amount of N drained from the land: (N river discharge plus N denitrified in the rivers) = 1.14 * (N drained from the land), $R^2 = 0.78$. Given the uncertainties of both the land drainage and in-stream denitrification numbers, this good correlation is encouraging, and the overestimate of 14% is a reasonable error term. However, we used the 'low' estimate provided by Seitzinger et al. based on the RF1 dataset. Using their 'high' estimates of riverine denitrification based on the NHD dataset would yield an overestimate of N inputs by 38%: (N river discharge plus N denitrified in the rivers) = 1.38 * (N drained from the land), $R^2 = 0.92$. Assuming that the discharge values are correct, this would imply that either drainage from land has been underestimated or that in-river denitrification has been overestimated. If one equates the difference between N inputs to the river ecosystem and riverine export to in-stream denitrification, this estimate is in good agreement with the 'low' estimate of Seitzinger et al. Our best guess (according to the lower of the two Seitzinger et al. modeled estimates) indicates that, on average, in-stream denitrification accounts for 11% of the total N storage & loss terms.

Denitrification in the landscape
The estimates for denitrification on land must be considered as tentative, as they result from budget discrepancies of N, and therefore contain the accumulated uncertainties of the other estimates. Losses of N in septic and sewage treatment, like ammonia volatilization, accounted for only a small percentage (3%) of the overall N outputs from the watershed (3%). It was calculated in direct proportion to population, and was only significant as an output term in the basins with the highest percentages of urban populations: the Charles (23%), Blackstone (15%), and Merrimack (12%). Estimated soil denitrification was the dominant 'sink' for N inputs to the watersheds, accounting for 34% of the total storage and loss terms, on average. Though this estimate is very rough, it is believable given the vast amount of N sources to the agricultural landscape available to be denitrified. This estimate is similar to the to the value of 40% reported by Kroeze et al. (submitted) for the Netherlands where inputs and, therefore, losses are higher than in the watersheds considered here.

Table 5. Storages and losses of N in watersheds* (kg N per km^2 per year)

Water-shed	N losses							changes in N storage			total N source inputs[5]	total N storages & losses	% inputs unre-solved[6]
	NH_3 vol.[1]	Denit. in soils	Denit. of human waste	Denit. in river	Riverine N export[2]	Wood export	Food export	Forest wood storage[3]	Forest soil storage	Other soil storage[4]			
PEN	4	233	14	187	317	411	18	–177	–83	17	857	943	–10
KEN	10	324	17	173	333	417	17	–180	–76	102	1126	1137	–1
AND	13	297	31	178	404	265	10	68	47	119	1332	1431	–7
SAC	4	153	30	128	389	158	0	233	167	91	1237	1353	–9
MER	11	506	264	190	499	138	0	120	245	149	2240	2121	5
CHA	10	393	1029	270	1756	126	0	313	93	561	4415	4552	–3
BLA	19	872	511	245	1140	162	0	249	62	174	3426	3436	0
CON	24	734	120	294	538	160	0	282	97	123	2286	2371	–4
HUD	20	422	60	251	502	171	0	263	334	191	2005	2213	–10
MOH	56	1437	100	374	795	82	134	270	225	361	3610	3834	–6
DEL	37	953	158	461	961	111	0	588	99	208	3005	3576	–19
SCH	199	1879	542	547	1755	138	0	259	97	853	5917	6270	–6
SUS	142	1907	100	616	977	143	459	478	82	338	4774	5244	–10
POT	374	2247	116	538	897	117	633	457	42	484	5696	5904	–4
RAP	135	2279	45	199	470	223	291	313	24	370	4671	4349	7
JAM	96	1163	45	191	314	309	91	477	–94	176	2961	2768	7
Wt. Avg.	*108*	*1253*	*107*	*397*	*718*	*192*	*224*	*313*	*71*	*259*	*3420*	*3641*	–6

[1] NH_3 Volatilization of fertilizers and animal waste that is transported outside of the watershed boundary. [2] Includes 1112 kg km^{-1} yr^{-1} from wastewater export from the Charles watershed area that is not drained on the Charles River. [3] In living biomass, dead wood, and live plant tissues. [4] In agricultural and urban land. [5] From table 2. [6] Estimated by difference of inputs – outputs.

NH_3 volatilization losses
N is also lost from the landscape in agricultural lands via the volatilization of fertilizers and animal waste. NH_3 volatilization losses in Table 5, which refer to net transport of NH_3 outside the watershed, are among the smallest loss terms, accounting on average for 3% of all outputs. Gross NH_3 volatilization (not shown) is about four times higher, but about 75% of that is assumed to be re-deposited within the watershed boundaries, and therefore does not figure in the input/output budget (Boyer et al. 2002). NH_3 volatilization losses increased from less than 10 kg km^{-2} yr^{-1} in the northernmost watersheds to over 100 kg km^{-2} yr^{-1} in several watersheds in the mid-Atlantic region. This trend reflects animal waste production.

Changes in storage

Storage in biomass
Averaged across all watersheds, changes in storage in forests due to net increment of living wood biomass and net accumulation of N in dead wood and in green plant tissues accounted for 9% of the total N storage and losses. Sinks for N in wood were greatest on those watersheds with both high growth rates per unit forest area and high fractions of watershed area in forest cover. Net N uptake rates were high on most watersheds, as many of these forests are aggrading after past harvests or agricultural abandonment. Forests across the eastern U.S. were cleared for agriculture several centuries ago, and forest regrowth stores large quantities of N in accumulating wood (Goodale et al. 2002). With the abandonment of marginal farmlands in New England, wood stocks increased by 50% between 1952 and 1992 (Birdsey & Heath 1995). This recovering biomass represents a large but finite N sink in old-field forests, although forest harvest will continue to export substantial quantities of N (e.g. Hornbeck & Kropelin 1982; Johnson 1992).

Storage in soils
Even though forested land acreage greatly exceeds agricultural and sub/urban acreage, N storage in non-forested land (∼7%) is only slightly less than that in forest land (10%). This is mainly due to the growing sub/urban area, the land use category that increased most, by ∼3500 km^2, for the whole study area (Figure 5). Much of the landscape that was once under continuous row cropping was converted to urban/suburban expansion, especially in the watersheds in Maine and New York and southward; these systems are predicted to have higher soil organic matter stocks as a result. In the New England region, the predominant change was the conversion of forest to urban/suburban land use. Relatively little data is available on soil organic matter levels in urban lands. However, the management and high level of inputs (i.e. fertilizer and

irrigation) in urban forests and grasslands (i.e. lawns, parks, golf courses) are conducive to the buildup of soil organic matter and soil N. Groffman et al. (1995) reported 30% higher soil organic matter in urban forests compared to rural forests having the same species and soil types. Changes in land use resulted in a net increase of soil N stocks of almost 97,000 Mg N yr^{-1} for all watersheds combined. The Potomac and Susquehanna watersheds accounted for about 60% of the net soil organic N increase, because of significant areas of soils under continuous row cropping which were converted to urban lands, forests, and pasture. There was a significant increase in use of reduced till and no-till over the 10 year period which should increase soil organic C and N stocks on the land remaining in agriculture. Further, conversion of agricultural soils, which are relatively depleted in organic matter compared to native forests and grasslands (Paustian et al. 1997b) into grasslands also increased soil N. On average, the estimated increases in soil organic N stocks in forested, agricultural, and urban lands due to changes in land use and in land management were 330 kg km^{-2} yr^{-1}, accounting for 9% of the total N storages and losses.

Comparison of total sources and total storages & losses

Total N storage & losses for each watershed vary from 943 to 6270 kg km^{-2} yr^{-1}, with an area-weighted mean of 3641 kg km^{-2} yr^{-1} (Table 5). The percentage of inputs not accounted for by our estimates of storage & losses vary among the watersheds from –19% to +7%. On average, the budget discrepancy was small (–6%). Sources (inputs) and storages & losses (outputs) are very well correlated: inputs = 0.96 * outputs; $R^2 = 0.98$. Figure 7 shows our 'best guesses' for the N inputs to the watersheds and for the fate of these N inputs via storage and loss pathways.

Losses by denitrification in landscape soils are our most uncertain estimates, because they were calculated by difference between total inputs to and outputs from each ecosystem, and therefore contain accumulated errors from other estimates. This loss term by difference, which we ascribed to denitrification in forest, urban, or agricultural soils, may also reflect the change in N storage in groundwater. However, we assume that groundwater aquifers, though enriched with N in some areas in the northeast, are not gaining significant new N over the period of interest since fertilizer use rates have been relatively stable in the U.S.A. over the past decade. In-river denitrification estimates are also very uncertain (see multiple estimates presented in Seitzinger et al. 2002).

The presence of appreciable changes in total N storage on land (18% of total storage & losses) indicates that there is a non-steady state condition, presumably associated with increasing N inputs from commercial imports

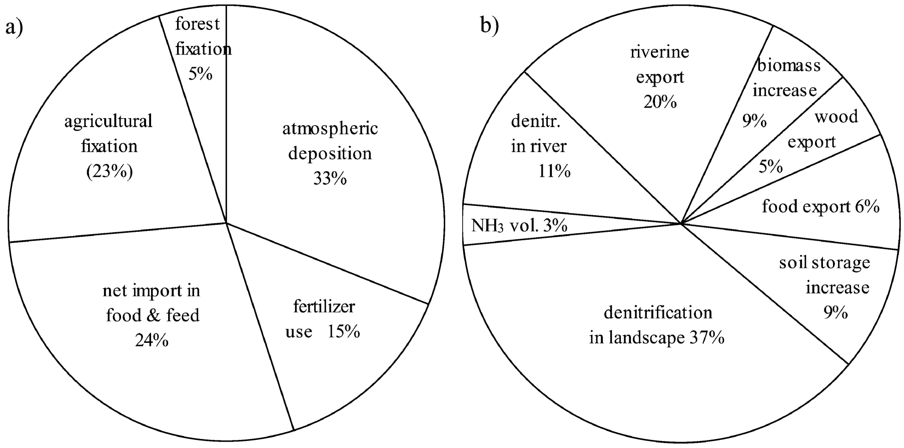

Figure 7. Best guess of (a) Nitrogen sources and (b) Nitrogen storages and losses. Values are the weighted average for the 16 watersheds.

and anthropogenically elevated atmospheric deposition. This in turn is ultimately due to the strongly increased industrial fixation of atmospheric N_2, and its application, mainly in agriculture. Increasing storage of N on land implies that drainage and denitrification exports of N are bound to increase further as storage terms reach a new steady state.

Understanding the sources and fate of N inputs to watersheds is necessary for mitigating N pollution problems in coastal and inland waters. Our estimates of N sources, storages, and losses are uncertain. Our ability to make these estimates is dependent on the availability a wide variety of statistical and spatial databases, and highlights the need for long-term monitoring. One element of uncertainty comes from the quality of these data themselves, and the methods used to scale information to the boundaries of our watersheds and to the timeframe of interest. For example, recent papers discuss challenges in estimating atmospheric N inputs (Meyers et al. 2001) and N loads in rivers (Brock 2001) based on incomplete and uncertain data from point monitoring networks. Another element of uncertainty comes from the empirical and process models from which we calculate storage and loss terms. Further research is needed to better understand the processes controlling N transport and transformations, and on how to best represent these processes in models that allow assessments at the scale of large regions.

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