Estuaries Vol. 25, No. 4b, p. 656–676 August 2002

Sources of Nutrient Pollution to Coastal Waters in the United States: Implications for Achieving Coastal Water Quality Goals

ROBERT W. HOWARTH[1,2,*], ANDREW SHARPLEY[3], and DAN WALKER[4,5]

[1] *The Ecosystems Center, Marine Biological Laboratory, Woods Hole, Massachusetts 02534*
[2] *Department of Ecology and Evolutionary Biology, Cornell University, Ithaca, New York 14853*
[3] *U.S. Department of Agriculture, Agricultural Research Service, Pasture Systems and Watersheds, State College, Pennsylvania 16802*
[4] *Oceans Studies Board, The National Academies, Washington, D.C. 20007*
[5] *The Marine Policy Center, Woods Hole Oceanographic Institution, Woods Hole, Massachusetts 02543*

ABSTRACT: Some 60% of coastal rivers and bays in the U.S. have been moderately to severely degraded by nutrient pollution. Both nitrogen (N) and phosphorus (P) contribute to the problem, although for most coastal systems N additions cause more damage. Globally, human activity has increased the flux of N and P from land to the oceans by 2-fold and 3-fold, respectively. For N, much of this increase has occurred over the past 40 years, with the increase varying by region. Human activity has increased the flux of N in the Mississippi River basin by 4-fold, in the rivers of the northeastern U.S. by 8-fold, and in the rivers draining to the North Sea by more than 10-fold. The sources of nutrients to the coast vary. For some estuaries, sewage treatment plants are the largest single input; for most systems nonpoint sources of nutrients are now of relatively greater importance, both because of improved point source treatment and control (particularly for P) and because of increases in the total magnitude of nonpoint sources (particularly for N) over the past three decades. For P, agricultural activities dominate nonpoint source fluxes. Agriculture is also the major source of N in many systems, including the flux of N down the Mississippi River, which has contributed to the large hypoxic zone in the Gulf of Mexico. For both P and N, agriculture contributes to nonpoint source pollution both through losses at the field scale, as soils erode away and fertilizer is leached to surface and ground waters, and from losses from animal feedlot operations. In the U.S. N from animal wastes that leaks directly to surface waters or is volatilized to the atmosphere as ammonia may be the single largest source of N that moves from agricultural operations into coastal waters. In some regions, including the northeastern U.S., atmospheric deposition of oxidized N from fossil-fuel combustion is the major flux from nonpoint sources. This atmospheric component of the N flux into estuaries has often been underestimated, particularly with respect to deposition onto the terrestrial landscape with subsequent export downstream. Because the relative importance of these nutrient sources varies among regions and sites, so too must appropriate and effective mitigation strategies. The regional nature and variability of nutrient sources require that nutrient management efforts address large geographic areas.

Introduction

Nutrient pollution is currently the largest pollution problem in the coastal rivers and bays of the U.S. (National Research Council 2000; Howarth et al. 2000) and is likely to increase globally as human use of inorganic fertilizers and fossil fuels, the two dominant sources of nutrients, continues to intensify, at least on a global basis. Much remains to be learned about the geographic extent and severity of eutrophication, the relative susceptibility of different coastal ecosystems, and the most effective nutrient control strategies. There is also a great need to better translate scientific knowledge into effective policy and management strategies, which require an understanding of the complex oceanic, estuarine, and watershed processes that contribute to eutrophication. With this better understanding, more effective techniques can be developed for reducing and preventing nutrient pollution, eutrophication, and associated effects.

In an effort to provide advice to federal, state, and local government agencies charged with addressing the growing problems of nutrient over-enrichment, the National Research Council (NRC) created the Committee on the Causes and Management of Coastal Eutrophication. Through the efforts of this committee and other volunteers, the NRC published *Clean Coastal Waters: Understanding and Reducing the Effects of Nutrient Pollution* (NRC 2000). To a large degree this paper summarizes findings presented in that report, while providing greater context for the significance of that report to the scientific community at large.

Although eutrophication and other adverse ef-

* Corresponding author; e-mail: rwh2@cornell.edu.

© 2002 Estuarine Research Federation

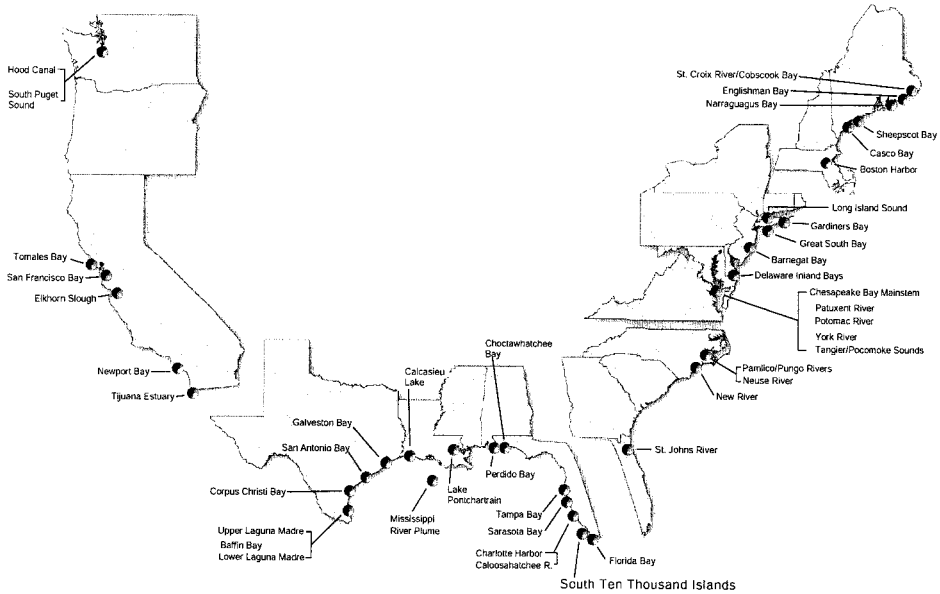

Fig. 1. Forty-four estuaries along all the nation's coasts were showing high expressions of nutrient over-enrichment by the National Oceanic and Atmospheric Administration National Estuarine Eutrophication Assessment (Bricker et al. 1999). In most of these estuaries, one or more of the following symptoms occur over large areas of the system, annually, persistently, or for long durations: high chlorophyll levels, macroalgal abundance problems, epiphyte abundance problems, low dissolved oxygen concentrations, loss of submerged aquatic vegetation, and harmful algal blooms. An additional 36 estuaries (not shown) show moderate effects of nutrient over-enrichment.

fects of nutrient pollution are recognized as growing problems in many of the nation's estuaries and coastal areas, ranging from Long Island Sound to the Chesapeake Bay to the plume of the Mississippi River in the Gulf of Mexico, the nation's capability to respond has been limited by lack of knowledge about the extent, severity, and characteristics of the problem. To fill this void, the National Oceanic and Atmospheric Administration (NOAA) designed the National Estuarine Eutrophication Assessment to gather consistent data nationwide and provide the basis for a national strategy for research, monitoring, and management of this pervasive problem. Bricker et al. (1999) summarize the assessment results. This report concludes that symptoms of eutrophication are present in many of the nation's estuaries, with high expression of symptoms in roughly one-third of the estuaries studied (44 of the 139 sites; Fig. 1). The report asserts that, left unabated, two out of every three estuaries will have impaired use by 2020. Problems occur in estuaries along all coasts, but are most prevalent in estuaries along the Gulf of Mexico and mid-Atlantic coasts where human influences are substantial and exchange with the open ocean tends to be slow.

Sources of Nutrient Inputs to Estuaries and Coastal Waters

Human activity has an enormous influence on the global cycling of nutrients, especially on the movement of nutrients to estuaries and other coastal waters. For phosphorus (P), global fluxes are dominated by the essentially one-way flow of P carried in eroded materials and wastewater from the land to the oceans, where it is ultimately buried in ocean sediments (Hedley and Sharpley 1998). The size of this flux is currently estimated at 22 Tg P yr^{-1} (Howarth et al. 1995). Prior to increased human agricultural and industrial activity the flow is estimated to have been 8 Tg P yr^{-1} (Howarth et al. 1995), suggesting that current human activities cause an extra 14 Tg of P to flow into the ocean sediment sink each year, or approximately the same as the amount of P fertilizer (16 Tg P) applied to agricultural land annually (NRC 2000).

The effect of human activity on the global cycling of nitrogen (N) is equally immense, and the rate of change in the pattern of use is much greater (Galloway et al. 1995). The single largest global change in the N cycle comes from increased reliance on synthetic inorganic fertilizers, which accounts for more than half of the human alteration of the N cycle (Vitousek et al. 1997). The process for making inorganic N fertilizer was invented early in the 20th century (Smil 2001), but was not widely used until the 1950s. The rate of use increased steadily until the late 1980s, when the collapse of the former Soviet Union led to great disruptions in agriculture and fertilizer use in Russia and much of eastern Europe. These disruptions

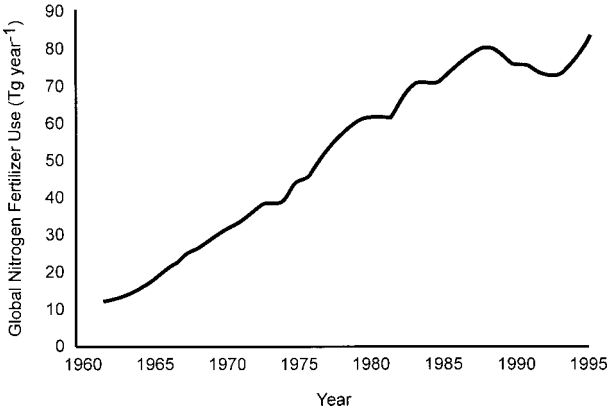

Fig. 2. Annual global nitrogen fertilizer consumption for 1960–1995 (1 Tg = 10^{12} g; data from FAO 1999).

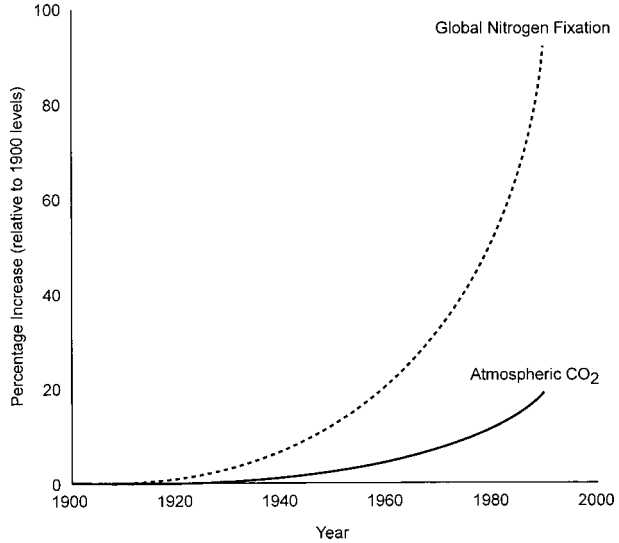

Fig. 3. The relative increase in creation of reactive, biologically available nitrogen by human activities globally compared to the increase in carbon dioxide in the atmosphere that contributes to global warming (modified from Vitousek et al. 1997).

resulted in a slight decline in global N fertilizer use for a few years (Matson et al. 1997). By 1995 the global use of inorganic N fertilizer was again growing rapidly (Fig. 2), with much of the growth driven by increased use in China (NRC 2000). As of 1996 use was approximately 83 Tg N yr^{-1}. Approximately half of the inorganic N fertilizer that was ever used on Earth has been applied during the last 15 years.

Production of N fertilizer is the largest process whereby human activity mobilizes N globally. Other human-controlled processes, such as combustion of fossil fuels and production of N-fixing crops in agriculture, convert atmospheric N into biologically available forms of N. Overall, human fixation of N (including production of fertilizer, combustion of fossil fuels, and production of N-fixing agricultural crops) increased globally 2-fold to 3-fold between 1960 and 1990 and continues to grow (Galloway et al. 1995). By the mid-1990s human activities made new N available at a rate of 140 Tg N yr^{-1} (Vitousek et al. 1997), matching the natural rate of biological N fixation on all the land surfaces of the world (Vitousek et al. 1997; Cleveland et al. 1999). The rate at which humans have increased the supply of reactive, biologically available N, leading to global eutrophication, far exceeds the rate at which humans have increased CO_2 in the atmosphere, leading to global warming (Fig. 3; Vitousek et al. 1997).

The human alteration of nutrient cycles is not uniform over the Earth, and the greatest changes are concentrated in the areas of greatest population density and greatest agricultural production. Some regions of the world have seen very little change in the flux of either N or P to the coast (Howarth et al. 1995, 1996), while in other places the change has been tremendous. Human activity is estimated to have increased N inputs to the coastal waters of the northeastern U.S. generally and to Chesapeake Bay specifically by 6-fold to 8-fold (Boynton et al. 1995; Howarth et al. 1996; Howarth 1998). Atmospheric deposition of N has increased even more than this in the northeast (Holland et al. 1999). The time trends in human perturbation of nutrient cycles can also vary among regions. For example, while the global use of inorganic N fertilizer continues to increase, the use of N fertilizer in the U.S. has increased less rapidly since 1985 (Fig. 4; Howarth et al. 2002).

Note that the use of N fertilizer in the U.S. in the next century may increase to support greater exports of food to developing countries (Howarth et al. 2002). Countries such as China have been largely self-sufficient in food production for the

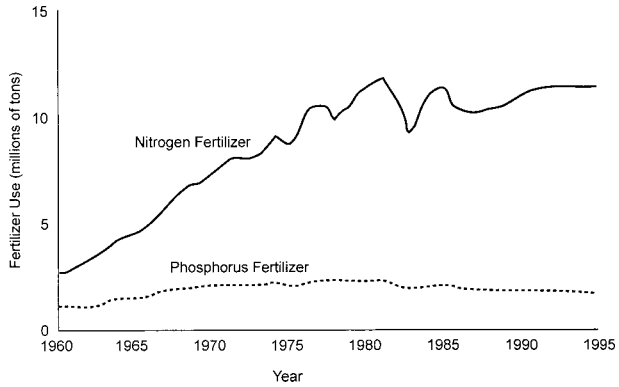

Fig. 4. U.S. commercial fertilizer use (modified from Evans et al. 1996).

past two decades, in part because of increased use of N fertilizer. The use of fertilizer in China is now very high, almost 10-fold greater than in the U.S. per area of cultivated land, and further increases in fertilizer use are less likely to lead to huge increases in food production as they have in the past. If China's population continues to grow, it may once again be forced to import food from the U.S. and other developed countries, leading to more use of N fertilizer in the U.S.

Most water quality management emphasizes control of discharges from wastewater treatment plants and other point sources; generally of greater concern for nutrients and coastal eutrophication are nonpoint sources of nutrients (NRC 1993a). A regional-scale analysis of fluxes of N from the landscape to the coast of the North Atlantic Ocean demonstrated that nonpoint sources of N exceeded sewage inputs for all regions in both Europe and North America (Howarth et al. 1996). Overall, sewage contributed only 12% of the flux of N from the North American landscape to the North Atlantic Ocean (Howarth et al. 1996). Nonpoint sources also dominate for P inputs to surface waters in the U.S. (Sharpley and Rekolainen 1997; Carpenter et al. 1998), and because of an effort to control P point source pollution, nonpoint sources of P have grown in relative importance since 1980 (Jaworski 1990; Sharpley et al. 1994; Litke 1999). Reduction in eutrophication and other effects of nutrient pollution of most estuaries requires the management of nutrient inputs from nonpoint sources in addition to those of wastewater treatment plants and industrial sources (NRC 1993a).

The Changing Role of Agricultural and Fossil Fuel Use

One of the greatest changes in agriculture has been the use of inorganic fertilizers, which expanded dramatically after World War II in response to the demand for increased agricultural output. In the developed countries large processing plants were built to manufacture nitrogenous fertilizers and convert imported rock phosphate into a variety of water-soluble and partially water-soluble P fertilizer products. Basic slag, a by-product from the steel industry, also became widely used in the manufacturing of P fertilizer. In the U.S. the use of inorganic P fertilizer rose rapidly in the 1940s and 1950s, but has been relatively constant since 1960. The rate of use of inorganic N fertilizer, on the other hand, continued to rise rapidly until the early 1980s (Fig. 4). This relative gain in N use over P use resulted primarily from favorable crop yield responses, especially corn, to N fertilizers.

Over the last 30 years, agricultural production systems in the U.S. have become more specialized and concentrated. During this time, overall agricultural production has more than doubled (Evans et al. 1996) and is occurring on less agricultural land and on fewer but larger farms (Evans et al. 1996). Since 1950 U.S. farmland has decreased from 1.2 billion to 970 million acres (20%) and the number of farms has dropped from 5.6 million to 2.1 million (63%), while average farm size has increased from 213 to 469 acres (120%).

In many states animal feeding operations (AFOs) are now a major source of agricultural income. The rapid growth of the animal industry in certain areas of the U.S. has been coupled with an intensification of operations. Current census information shows an 18% increase in the number of hogs in the U.S. over the last 10 years at the same time as a 72% decrease in numbers of hog farms. Over the same 10 years, the number of dairy farms decreased 40%, but herd size increased 50%. A similar intensification of the poultry and beef industries has also occurred, with 97% of poultry production in the U.S. coming from operations with more than 100,000 birds and over a third of beef production coming from just under 2% of the feedlots (Gardner 1998). Driving this intensification is an increased demand for animal products and improved profitability because of advances in transportation, processing, and marketing. Animal feeding operations pose significant challenges with the management of wastes produced.

Prior to World War II, farming communities tended to be self-sufficient, in that enough feed was produced locally to meet animal requirements and the manure nutrients could be effectively recycled to meet crop needs (NRC 2000). After World War II, increased fertilizer use in crop production fragmented farming systems and created specialized crop and animal operations that efficiently coexisted in different regions. Because farmers did not need to rely on as fertilizers (the primary source until fertilizer production and distribution became cheap), grain and animal production could be separated spatially. By 1995 the major animal-producing states imported over 80% of their grain for feed (Lanyon and Thompson 1996), and less than a third of the grain produced on farms today is fed to animals on the farm where it is grown (U.S. Department of Agriculture 1989).

This evolution of agricultural systems is resulting in a major transfer of nutrients from grain-producing areas to animal-producing areas and, consequently, accumulation of N and P in soils of the animal-producing areas. The potential for N and P surplus at the farm scale can be much greater in AFOs than in cropping systems, because nutrient inputs become dominated by feed rather than fer-

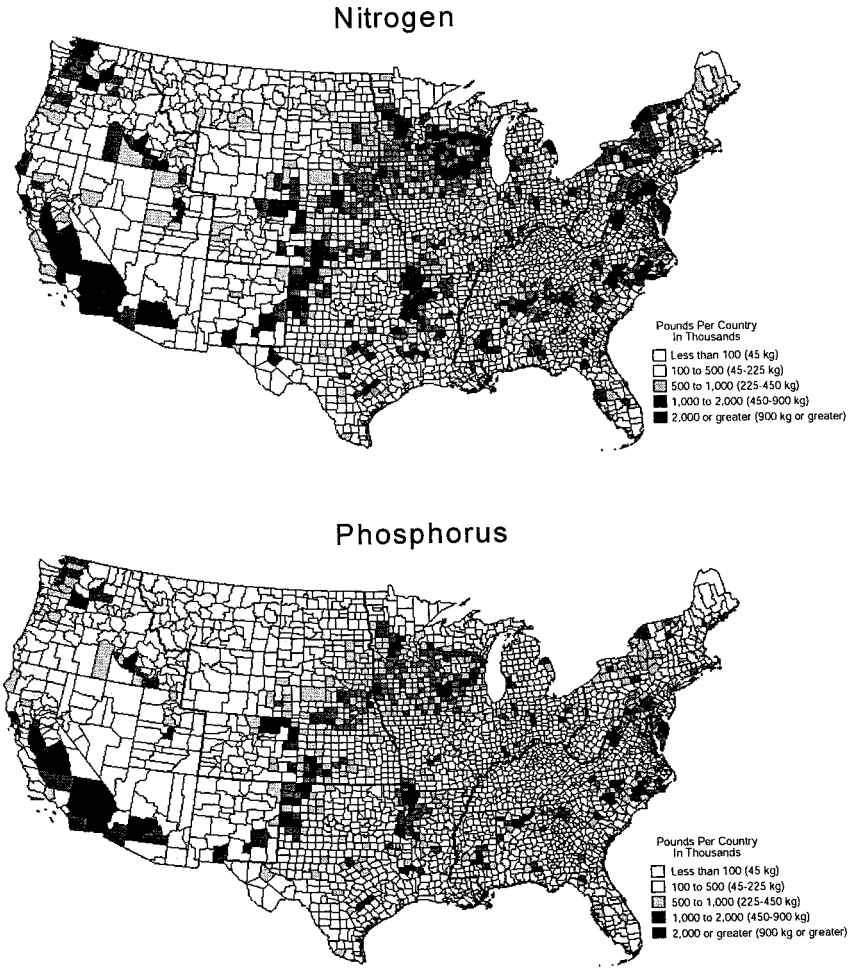

Fig. 5. Estimated manure nitrogen and phosphorus production from confined livestock (modified from Lander et al. 1998).

tilizer (Isserman 1990; NRC 1993b). Many water quality concerns are a result of this imbalance in system inputs and outputs of N and P, which have been brought about by an increase in AFOs. Lander et al. (1998) calculated the amounts of N and P produced by manure in confined AFOs on a country-wide basis (Fig. 5). From this and crop yield information, Lander et al. (1998) were able to identify those counties where manure production exceeds the total nutrient need for crop production (Fig. 6; assuming that manure is applied only to non-legumes and harvested crop land and hay land).

The number of U.S. counties where manure N and P exceeds the potential crop uptake has been steadily increasing since 1950 (Fig. 7). This increase has been greater for P than for N (Kellogg and Lander 1999). In those areas with an excess of N and P relative to crop needs, there is a greater risk of nutrient export from agricultural watersheds to surface and ground waters (Fig. 8). This excess of nutrients in manure tends to occur in areas where downstream export is likely due to relatively wet climates, since high water availability is conducive to animal feeding operations.

Export of Phosphorus from Agricultural Systems

Several surveys of U.S. watersheds have clearly shown that P loss in runoff increases as the forested portion of the watershed decreases and agriculture increases (Omernik 1977; Rast and Lee 1978). In general, forested watersheds conserve P, with P input in dust and rainfall usually exceeding outputs in stream flow (Taylor et al. 1971; Hobbie and Likens 1973; Schreiber et al. 1976). Surface runoff from forests, grasslands, and other non-cultivated soils carries little sediment, so P fluxes are low and the export that occurs is generally dominated by dissolved P. The loss of P from forested land tends to be similar to that found in subsurface or dissolved base flow from agricultural land (Ryden et al. 1973; House and Casey 1988). The cul-

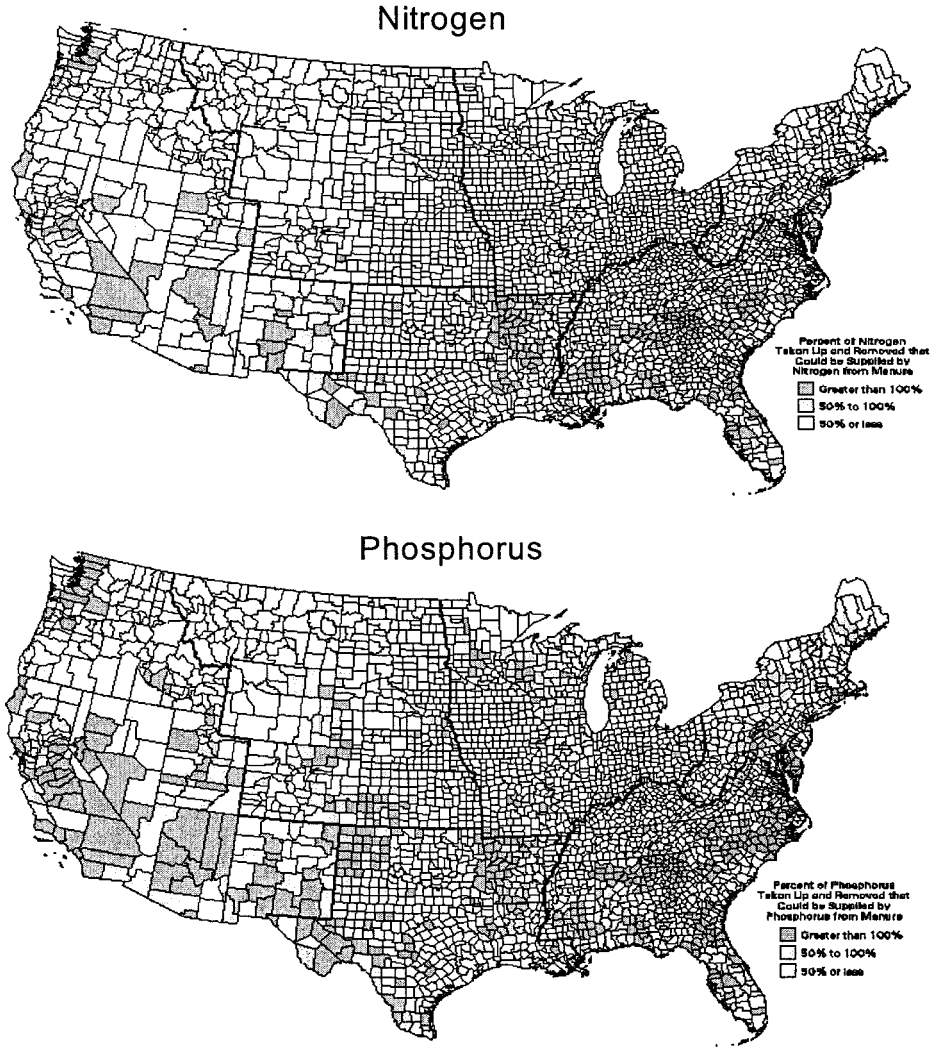

Fig. 6. Potential for nitrogen and phosphorus available from animal manure to meet or exceed plant uptake and removal on non-legume, harvested cropland and hayland (modified from Lander at al. 1998).

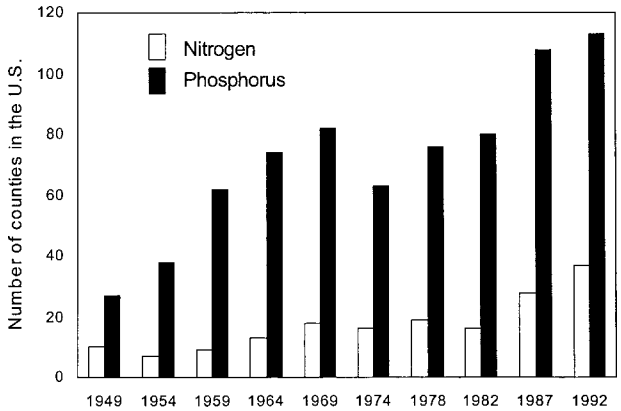

Fig. 7. Number of counties where manure nutrients exceed the potential plant uptake and removal, including pastureland application (modified from Kellogg and Lander 1999).

tivation of land in agriculture greatly increases erosion and with it the export of particle-bound P. Particulate fluxes constitute 60% to 90% of P exported from most cultivated land (Sharpley et al. 1995). In the eastern U.S. conversion of land from forests to agriculture between 1700 and 1900 resulted in a 10-fold increase in soil erosion and a presumed similar increase in P export to coastal waters, even without any addition of P fertilizer (Meade 1988; Howarth et al. 1996). The soil-bound P includes both inorganic P associated with soil particles and P bound in organic material eroded during flow events. Some of the sediment-bound P is not readily available (Howarth et al. 1995), but much of it can be a long-term source of P for aquatic biota (Sharpley 1993; Ekholm 1994).

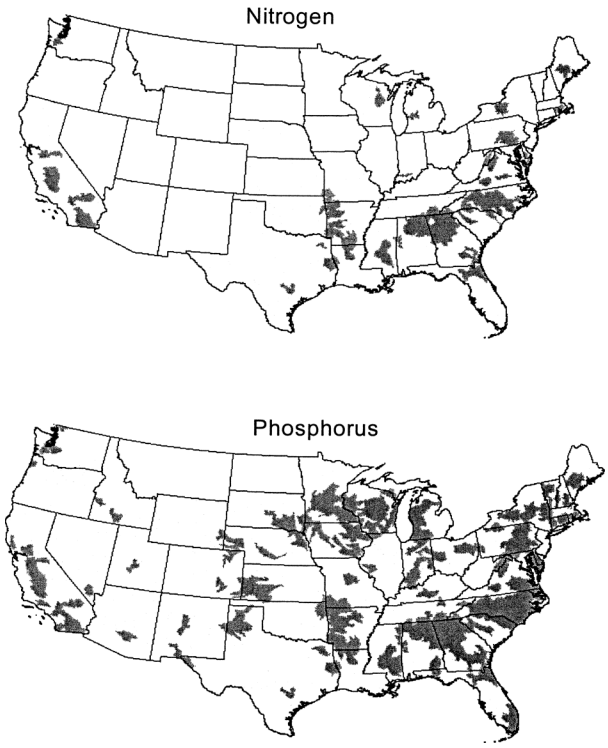

Fig. 8. Watersheds with a high potential for soil and water degradation from manure nitrogen and phosphorus (modified from Kellogg and Lander 1999).

Increases in P export from agricultural landscapes have been measured after the application of P (Sharpley and Rekolainen 1997). P export is influenced by the rate, time, and method of P application; form of fertilizer or manure applied, amount and time of rainfall after application, and land cover. These losses are often small from the standpoint of farmers (generally less than 200 kg P km^{-2}), represent a minor proportion of fertilizer or manure P applied (generally less than 5%), and are not of economic importance to farmers in terms of irreplaceable fertility. They can contribute to eutrophication of downstream aquatic ecosystems.

While P export from agricultural systems is usually dominated by surface runoff, important exceptions occur in sandy, acid organic, or peaty soils that have low P adsorption capacities and in soils where the preferential flow of water can occur rapidly through macropores (Sharpley and Syers 1979; Bengston et al. 1992; Sharpley et al. 1998; Sims et al. 1998). Soils that allow substantial subsurface export of dissolved P are common in parts of the Atlantic coastal plain and Florida and are thus important to consider in the management of coastal eutrophication in these regions (NRC 2000). Although there exists a good understanding of the chemistry of P in soil-water systems, the hydrologic pathways linking spatially variable P sources, sinks, temporary storages, and transport processes in landscapes are less well understood. This information is critical to the development of effective mitigation programs that address the reduction of P export from agricultural watersheds.

Most of the P exported from agricultural watersheds comes from only a small part of the landscape during a few relatively large storms, where hydrologically active areas of a watershed contributing surface runoff to streamflow are coincident with areas of high soil P (Gburek et al. 2000; Pionke et al. 2000). To be most effective, mitigation strategies must consider critical source areas, which are specific identifiable areas within a watershed that are most vulnerable to P loss in surface runoff (Heathwaite and Johnes 1996; Gburek and Sharpley 1998). Critical source areas are dependent on the coincidence of transport (surface runoff, erosion, subsurface flow, and channel processes) and source or site management factors (functions of soil, crop, and management; Table 1). These are typically well defined and reflect land-use patterns related to soil P status, fertilizer and manure P inputs, and tillage (Table 1). Even in

TABLE 1. Factors influencing phosphorus loss.

Factors	Description
Erosion	Total P loss strongly related to erosion.
Surface runoff	Water has to move off or through a soil for P to move.
Subsurface flow	In sandy, organic, or P-saturated soils, P can leach through the soil.
Soil texture	Influences relative amounts of surface and subsurface flow.
Irrigation runoff	Improper irrigation management can induce surface runoff and erosion of P.
Connectivity to stream	The closer the field to the stream, the greater the chance of P reaching it.
Proximity of P-sensitive water	Some watersheds are closer to P-sensitive waters than others (i.e., point of impact).
Sensitivity P input	Shallow lakes with large surface area tend to be more vulnerable to eutrophication.
Soil P	As soil P increases, P loss in surface runoff and subsurface flow increases.
Applied P	The more P (fertilizer or manure), the greater the risk of P loss.
Application method	P loss increases in this order: subsurface injection, plowed under, and surface broadcast with no incorporation.
Application timing	The sooner it rains after P is applied, the greater the risk for P loss.

regions where subsurface flow pathways dominate, areas contributing P to drainage waters appear to be localized to soils with high soil P saturation and hydrologic connectivity to the drainage network (Schoumans and Breeuwsma 1997). Understanding how water moves and where surface runoff occurs is critical to a more detailed understanding of P export from agricultural watersheds.

Export of Nitrogen from Agricultural Systems

The fate of N applied as fertilizer to agricultural fields has received extensive study. Generally for the U.S., 45% to 75% of the N in fertilizer is removed in crop harvest (Bock 1984; Nelson 1985; NRC 1993b). Of the remainder some is stored as organic N in the soil, some is volatilized to the atmosphere, and some leaches to ground and surface waters. A variety of factors including soil type, climate, fertilizer type, and farming practices influence the fate of fertilizer use (Howarth et al. 1996). For typical farming practices in the U.S. the percentage of fertilizer that leaches to ground and surface waters varies between 10% and 40% for loam and clay soils and 25% and 80% for sandy soils (Howarth et al. 1996). Overall in North America it is estimated that 20% of the fertilizer N applied to agricultural fields leaches into ground and surface waters (Howarth et al. 1996), although much of that is lost to denitrification in downstream wetlands, streams, and rivers before reaching estuaries or coastal waters (Alexander et al. 2000, 2002; Seitzinger et al. 2002; van Breemen et al. 2002).

A variety of factors affect the volatilization of N from fertilizer to the atmosphere, including soil type, climate, farming practices, and type of fertilizer (Bouwman et al. 1997). When ammonium sulfate is applied to a soil with a pH below 5.5, less than 2% of the ammonium is volatilized to the atmosphere (in the form of ammonia) but when ammonium sulfate is applied to calcareous soil (which has a higher pH), up to 50% of the N can be volatilized as ammonia gas to the atmosphere (Whitehead and Raistrick 1990; Bouwman et al. 1997). For typical farming practices, climate, and soils in the U.S. and Europe, Bouwman et al. (1997) estimated that on average 8% of the N in ammonium sulfate and 15% of the N in urea is volatilized to the atmosphere. The percentages are greater in tropical climates because of warmer temperatures. And the volatilization from nitrate-based fertilizers is much less than for urea-based or ammonium-based fertilizers. While emissions of nitric oxide to the atmosphere are an important N loss from fertilized fields in tropical areas, this is generally a very small flux in temperate regions, including the U.S. (Holland et al. 1999). Virtually all the N volatilized from agricultural fields is eventually redeposited back onto the landscape and can reach estuaries and coastal waters (Howarth et al. 1996). Generally, this N is redeposited quite close to the point of emission, and deposition rates fall of exponentially with distance from the source (Fahey et al. 1999; Holland et al. 1999).

Because 45% to 75% of the N applied as fertilizer is harvested in crops, tracing the fate of N in food and feedstock is important for understanding N inputs to natural waters (Howarth et al. 1996). In the U.S. most crops are fed to animals (Bouwman and Booij 1998), and most of this in turn is excreted by the animals. For such animals as poultry, hogs, and cows kept in barns or sheds, 36% of the excreted N on average is volatilized to the atmosphere as ammonia; keeping cows in meadows instead of barns reduces the atmospheric volatilization by more than 50% (Bouwman et al. 1997). Assuming that 65% of the N applied as fertilizer is removed in crops (NRC 1993b); two-thirds of the crop production in the U.S. is fed to animals (Bouwman and Booij 1998); the N growth efficiency for animals is 10% (Bouwman and Booij 1998); and 36% of the N excreted by animals is volatilized to the atmosphere (Bouwman et al. 1997), then 14% of all N applied in fertilizer is eventually volatilized to the atmosphere as ammonia after being consumed by animals. This is in addition to direct volatilization of ammonia from fertilizers and from sewage treatment plants.

Ammonia volatilization to the atmosphere from agricultural systems in the U.S. is of the same order of magnitude as nitrate leaching from agricultural fields into surface waters. Although losses are poorly documented, animal wastes also contribute N directly to surface waters (Howarth 1998). In a regional comparison of N cycling in major regions of the U.S. and Europe, Howarth (1998) found that estimates of N consumption by domestic animals were far better as predictors of nonpoint-source N fluxes in rivers than were rates of application of inorganic N fertilizer.

Fate of Nitrogen in Atmospheric Deposition

Reactive N in the atmosphere contains both reduced compounds (NH_x) and oxidized compounds (NO_y). These come from a variety of sources, including fossil-fuel combustion, biomass burning, lightning, and emissions from soils. In the U.S. most NO_y comes from fossil-fuel combustion and most NH_x comes from emissions from agricultural sources (Howarth et al. 1996; Prospero et al. 1996; Bouwman et al. 1997; Holland et al. 1999). The lifetime in the atmosphere for many of these reactive N compounds is short, from hours to a few days, and a large portion of the N is deposited near its source (Holland et al. 1999). NO_y contributes

to acid rain, but estuarine waters are well buffered and are not directly susceptible to acidification. The threat from NO_y discussed here comes from its role as a contributor of N to coastal eutrophication.

N deposition directly onto the water surfaces of estuaries and coastal waters can be substantial, although this is difficult to measure. Monitoring stations for atmospheric input of N tend to be scarce in coastal areas (NRC 2000). Where monitoring stations exist, they tend to measure only the N deposited in precipitation (wet deposition). Dry deposition of N (the impaction of particles and gases of N onto water, plant, or land surfaces) has proven difficult to measure in any type of ecosystem, and usually only wet deposition or at best some portion of dry deposition are measured at monitoring sites (Holland et al. 1999).

Evidence indicates that deposition directly onto the water surfaces of estuaries and coastal waters tends to contribute from 1 to 40% of the total N inputs (Nixon et al. 1996; Paerl 1997; Howarth 1998; Paerl and Whitall 1999; Valigura et al. 2000), with the Baltic Sea (Nixon et al. 1996) and Tampa Bay (Zarbock et al. 1996) at the upper end of this range. Evidence suggests a significant movement of N in the atmosphere from the eastern U.S. to the coastal and even offshore waters of the North Atlantic Ocean where it is deposited (Prospero et al. 1996; Holland et al. 1999). This flux could be as large as half the entire amount of reactive N emitted into the Earth's atmosphere from the U.S. Because of the large natural flux of N from the deepwater of the North Atlantic Ocean onto the continental shelf off the eastern U.S., this atmospheric deposition probably contributes less than 10% of the total input of N to the surface waters of the continental shelf (Howarth 1998).

Much of the reactive N deposited from the atmosphere falls onto terrestrial ecosystems. This can affect estuaries and coastal waters to the extent that it is exported from land. The fate of N deposition in forests has received extensive study. Productivity of most U.S. forests in their natural state is limited by the supply of N (Vitousek and Howarth 1991). As more N is made available to these forests from atmospheric deposition, production and storage of N in organic matter can be expected to increase temporarily. The ability of forests to store N, however, is limited. Forests become saturated with respect to N when inputs exceed the total amount needed by trees and the assimilation capacity (through microbial and abiotic processes) of soil organic matter (Aber et al. 1989; Gundersen and Bashkin 1994; Magill et al. 1997; Emmett et al. 1998). Once a forest is saturated with respect to N, losses both to the atmosphere and to downstream ecosystems can increase rapidly. In European forests that have received high levels of N deposition for some time, the downstream export of N can be high, often greater than 500 kg N km^{-2} yr^{-1} (van Breemen et al. 1982; Hauhs et al. 1989; Schulze et al. 1989; Durka et al. 1994). Some evidence indicates that the process whereby forests switch from retaining N to exporting N as they become N saturated can be self-accelerating due to related changes in biogeochemical cycling and ecosystem decline (Schulze et al. 1989; Howarth et al. 1996).

Ecological theory suggests that young aggrading forests tend to retain more N and be less likely to become N saturated than old-growth mature forests (Vitousek and Reiners 1975; Aber et al. 1989). Forests that have been logged or burned within the past several decades to a century can therefore be expected to retain more N from deposition. A variety of factors, in addition to land-use history, can affect the ability of a forest to retain N, including the species composition of trees, climate, and soil type (Howarth et al. 1996; Aber and Driscoll 1997; Aber et al. 1997; Magill et al. 1997; Emmett et al. 1998). Lajtha et al. (1995) found that only half the N input from atmospheric deposition was retained by forests at Cape Cod, Massachusetts, whether the forests were young or mature, apparently because the sandy soils there allow N to pass through quickly. Forests on stony and sandy loam soils in western Massachusetts retained over 85% of N inputs even when heavily fertilized with N over a 6-yr period (Magill et al. 1997). In this fertilization experiment there was some evidence that N retention decreased over time as the N content of the forest increased and that N saturation occurred more rapidly in pine forests than in hardwood forests.

In a review of N retention in U.S. forests, Johnson (1992) found no relationship between N inputs and N losses to downstream ecosystems; the percentage of N deposition that was retained varied among forests from nearly none to virtually all. Much of this variation could have been caused by differences in land use, soil type, and dominant tree species (Lajtha et al. 1995; Aber and Driscoll 1997; Magill et al. 1997; Emmett et al. 1998). Some of the variation, however, could have been due to the exclusion of dissolved organic N fluxes in the budgets considered by Johnson (1992), all of which included losses only of inorganic N. Losses of organic N can be considerable from some forests (Hedin et al. 1995; Lewis et al. 1999; Lewis 2002). Even in northern New England where atmospheric deposition of N is moderately high, most of the dissolved N leaving forests is organic N rather than inorganic N (Campbell et al. 2000). Many of the budgets reviewed by Johnson (1992) were based on short-term studies, and losses of N

from forests can show considerable year-to-year variation in response to climatic variation (Aber and Driscoll 1997). Finally, dry deposition of N is difficult to estimate (Howarth et al. 1996; Holland et al. 1999; Valigura et al. 2000), and may not have been characterized correctly in some of the budgets summarized by Johnson (1992).

Emmett et al. (1998) proposed that the extent of N leaching from a forest can be predicted easily from the N status of the forest as measured by the ratio of organic carbon (C) to N in the forest floor. They experimentally illustrated that forests with a low N status (forest floor C:N greater than 30:1) retain most of the N added (well over 90%), whereas forests with a high N status (forest floor C:N less than 25:1) retain less than half the N added through deposition and fertilizer. Campbell et al. (2000) also demonstrated that the ratio of organic carbon to organic N in streamwater draining forests in the northeastern U.S. is a good predictor of export of inorganic N from those forests. Export of inorganic N increased dramatically in systems where the organic carbon to organic N ratio of the streamwater was below 20:1 to 25:1 (Campbell et al. 2000). A variety of factors control whether a forest acts as a sink or source of N, but forests experiencing long periods of high N inputs through atmospheric deposition will tend to become saturated with respect to N (Emmett et al. 1998). Over time a forested watershed that experiences high inputs of N deposition will reach its capacity to store N and will begin to act as a source of N to the streams that drain it. It is not surprising that experiments showed that N leaching from a forest slowed quickly after deposition was reduced through the use of roof exclosures (Bredemeier et al. 1998). This lead Emmet et al. (1998, p. 357) to suggest that "immediate benefits in water quality could be expected following any reduction in nitrogen deposition loading."

To manage lake acidification in some areas of Europe, managers have adopted the critical load concept (Bashkin 1997). This approach sets a goal of keeping atmospheric deposition below some level where it is thought that downstream release will be kept small enough to keep any ecological damage at an acceptable level. Research supports the conclusion that downstream release of N (and associated acid) can be expected to occur when a critical load value of 1,000 kg N km^{-2} yr^{-1} from atmospheric deposition is reached (Schulze et al. 1989; Pardo and Driscoll 1993; Emmett and Reynolds 1996; Williams et al. 1996; Skeffington 1999; Fig. 9). Average levels of N deposition (wet plus dry) currently exceed 1,000 kg N km^{-2} yr^{-1} for the northeastern U.S. and for much of Europe (Howarth et al. 1996; Prospero et al. 1996; Holland et al. 1999; Valigura et al. 2000).

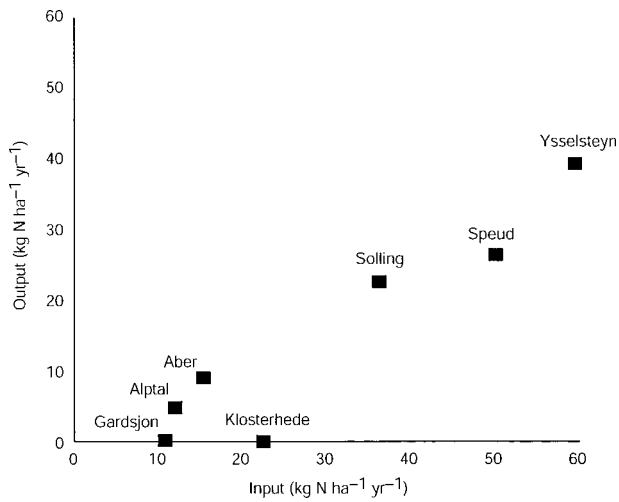

Fig. 9. Ambient inputs or nitrogen from deposition and experimental fertilization compared to leaching losses at the nitrogen saturation experiment sites (modified from Emmett et al. 1998).

The export of N following deposition onto terrestrial ecosystems other than forests has received less study. Some evidence indicates that grasslands are as retentive of N as forests, or even more so (Dodds et al. 1996). When N from the atmosphere is deposited onto agricultural fields, its fate may be similar to the fate of N fertilizer applied to such fields, although N deposited during the non-growing season could be prone to greater loss to downstream ecosystems (NRC 2000). The fate of atmospheric N deposited onto urban and suburban landscapes appears to be unstudied, although the N content of stormwater runoff from urban environments is high (Environmental Protection Agency 1983). N deposition (wet or dry) in urban environments is poorly measured because most deposition monitoring sites are in rural environments (Holland et al. 1999). Deposition onto urban landscapes should be high because much of the reactive N in the atmosphere is deposited near sources, and it is reasonable to expect that the export of this deposition to coastal waters is also high. Uncertainty over the extent of N deposition in urban environments is one of the greatest uncertainties in the N budget for the U.S. (Holland et al. 1999).

Nutrient Fluxes to the Coast: Insights from a Regional Analysis

At the scale of individual estuaries it has proven exceedingly difficult to determine the ultimate source of N inputs and the magnitude of the load each source contributes. Numerous obstacles exist

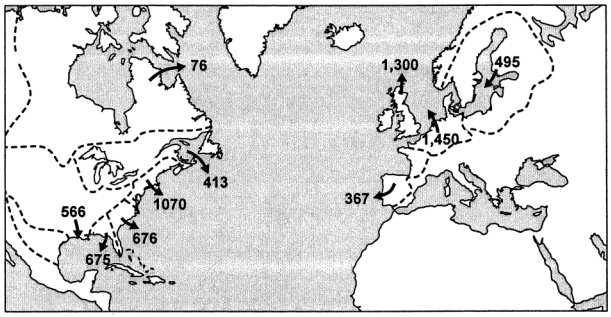

Fig. 10. Export of total nitrogen from large regions to the North Atlantic Ocean in rivers and from sewage treatment plants (kg N km^{-2} yr^{-1}; modified from Howarth 1998).

to understanding nutrient fluxes to the coast, including the existence of multiple sources (fossil-fuel combustion from both mobile and stationary sources plus agricultural sources), the difficulty in estimating dry deposition at the scale of whole watersheds, the difficulty in measuring gaseous losses from ecosystems (including denitrification of nitrate to N and volatilization of ammonia and NO_y compounds), and the multiple pathways for N flows (surface waters, ground waters, and atmosphere). These problems are far less significant when dealing with P fluxes. Much insight into N fluxes to coastal waters has been gained recently by analyzing fluxes at relatively large spatial scales.

The International SCOPE Nitrogen Project synthesized and analyzed information on N fluxes as of the late 1980s or early 1990s at the scale of large regions, such as the combined watersheds of the North Sea, the combined watersheds of the northeastern U.S. from Maine through the Chesapeake Bay, and the Mississippi River basin (Howarth et al. 1996; Howarth 1998). There are large variations in the export of N to the North Atlantic from regions in the temperate zone, with fluxes per area of watershed varying from as low as 76 kg N km^{-2} yr^{-1} for the watersheds of northern Canada to 1,450 kg N km^{-2} yr^{-1} for the watersheds of the North Sea (Fig. 10). As stated above, the export of N from nonpoint sources dominates the N fluxes in all regions (Howarth et al. 1996; Howarth 1998). The flux of N from a region per area of watershed, both the total flux and the flux from nonpoint sources, is weakly correlated with population density (Fig. 11).

At the scale of large regions, net imports of reactive N include the use of N fertilizer, N fixation by agricultural crops, deposition from the atmosphere of oxidized forms of N (which are presumed to come primarily from fossil-fuel combustion in the temperate zone; Holland et al. 1999), and the import or export of N in food and animal feedstocks (Howarth et al. 1996). Sewage is not

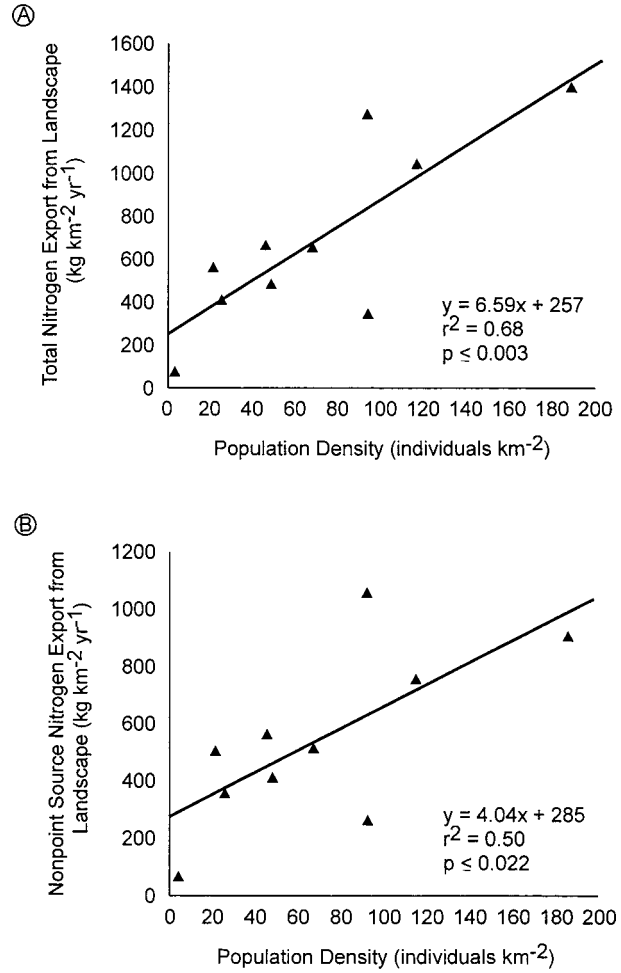

Fig. 11. The relationship between population density and the export of nitrogen in rivers to the coast for temperate regions surrounding the North Atlantic Ocean. Each point represents one region. (A) The total nitrogen export from the region in rivers. (B) The flux of nitrogen from nonpoint sources in the region, independent of upstream sources (modified from Howarth 1998).

considered a net source because it is a recycling of N that was brought into a region for agricultural purposes or directly as N in food. Deposition of ammonium and organic N from the atmosphere is also not considered a net input since, at large spatial scales, this is largely a recycling of N volatilized into the atmosphere from agricultural sources within the same region (Howarth et al. 1996; Howarth 1998).

The export of N from the landscape to the coast in the temperate zones of North America and Europe is well described as a linear function of the import of reactive N forms into the region by human activity (Fig. 12) (Howarth et al. 1996; Howarth 1998). On average for the temperate regions of North America and Europe, 20% of the N in-

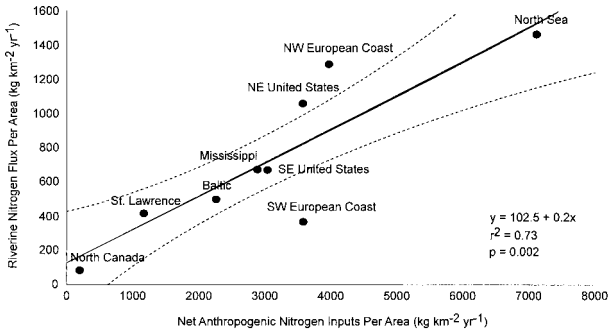

Fig. 12. A comparison of human-controlled inputs of nitrogen to a region and nitrogen export from the region to the coast in rivers for temperate regions surrounding the North Atlantic Ocean. The dashed lines refer to the 95% confidence limits around the regression line (solid line; modified from Howarth et al. 1996).

puts under human control flow out of regions to coastal waters. The remaining 80% of the human-controlled N inputs are either denitrified or stored within the regions. The nature of these sinks, including whether they will change with time, is poorly known (Howarth et al. 1996).

Regression analyses of N from flux from the landscape with human-related N inputs (Fig. 13a–d) suggest that deposition from fossil-fuel sources (NO_y deposition) per unit mass introduced into the landscape is a better predictor of N export to coastal waters ($r^2 = 0.81$) than is fertilizer application ($r^2 = 0.28$; Howarth 1998). A simple multiple regression that used both NO_y deposition and agricultural inputs (i.e., the sum of fertilizer, N fixation in agriculture, and net movements of N in foodstocks) was constructed to predict N export to the coast. The best overall fit was obtained by a

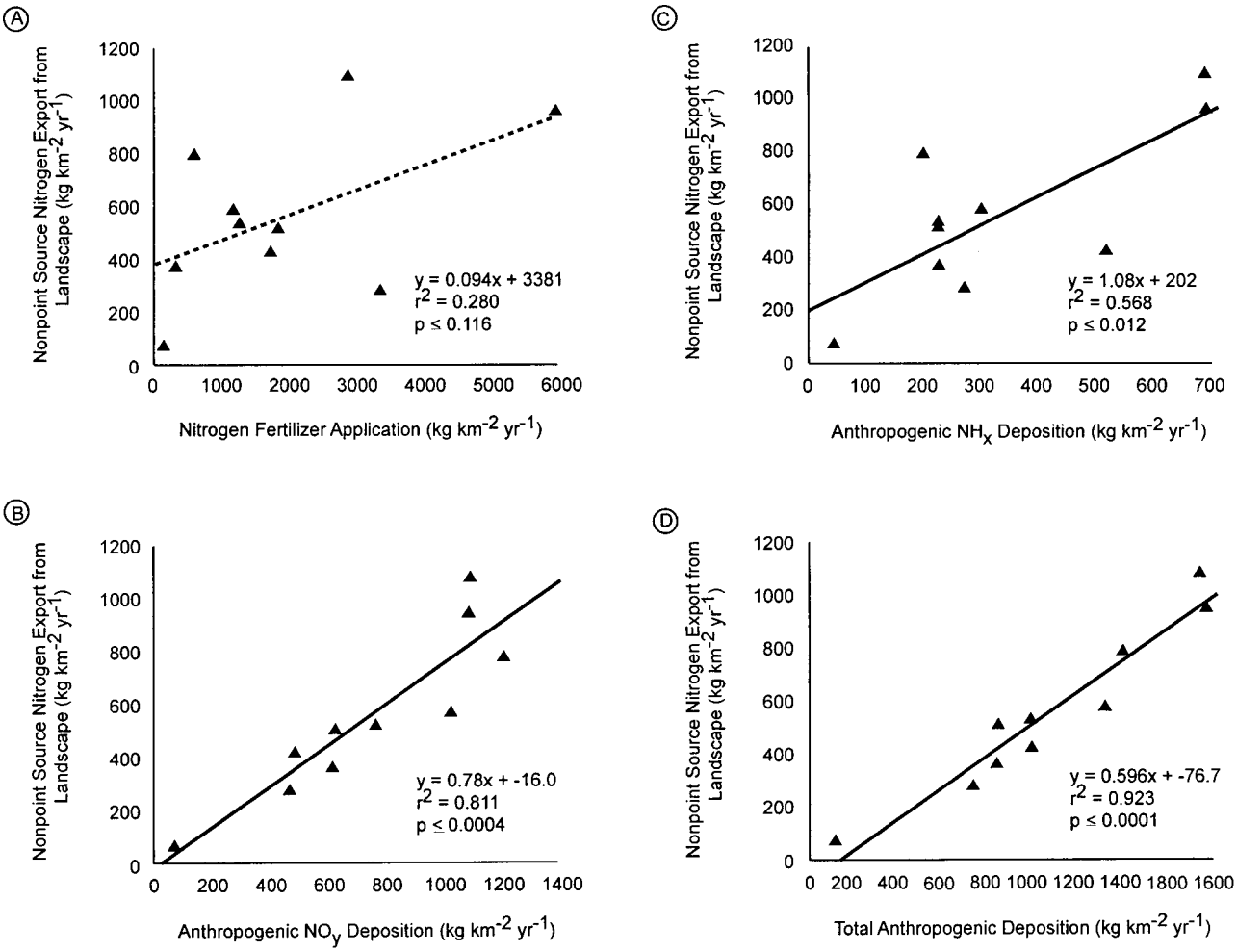

Fig. 13. Export of nitrogen per watershed area from nonpoint sources to the North Atlantic Ocean, where each point represents a large region (as in Fig. 11b) plotted as a function of nitrogen fertilizer inputs (A), atmospheric deposition of NO_y from anthropogenic sources (B), atmospheric deposition of NH_x from anthropogenic sources (C), and total nitrogen deposition (NO_y plus NH_x) from anthropogenic sources (D) (modified from Howarth 1998).

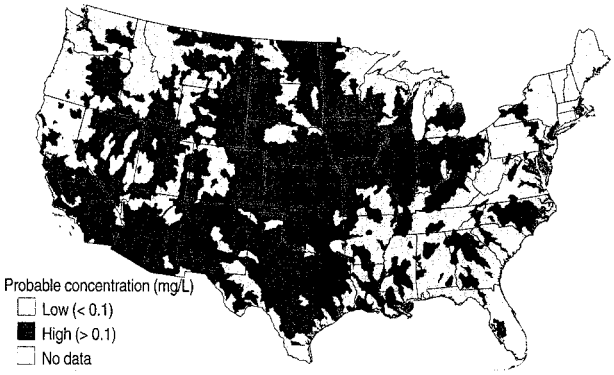

Fig. 14. Classification of predicted total phosphorus concentrations in surface waters of the U.S. as estimated from the SPARROW model (Smith et al. 1997).

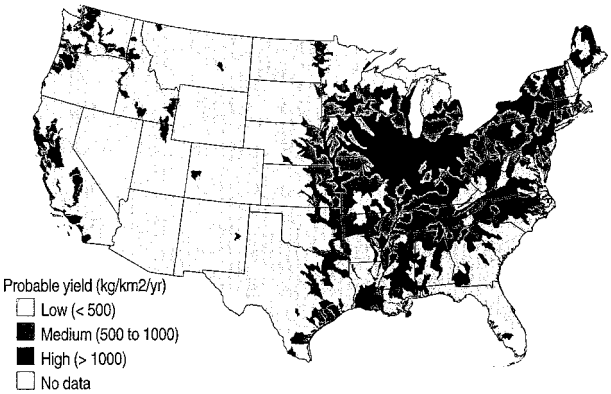

Fig. 15. Predicted local total nitrogen yield in hydrologic cataloging units of the conterminous U.S. Local yield refers to transport per unit area at the outflow of the unit due to nitrogen sources in the unit, independent of upstream sources (Smith et al. 1997).

curve in which the NO_y deposition term was seven times greater than the agricultural input term (Howarth et al. 1996). This suggests that, per unit mass, N from fossil-fuel sources may contribute more to the N flux in rivers to the coast than do agricultural sources. Of course, in many areas the total inputs of N as fertilizer are far greater than are the inputs from NO_y deposition. In the Mississippi River basin the total inputs of N as fertilizer far exceed those from NO_y deposition; consequently, agriculture is the greatest contributor to the N export from that basin (Goolsby et al. 1999).

The best regression fit for the export of N from nonpoint sources for the temperate regions of the North Atlantic Ocean results from using the sum of NO_y deposition and ammonium deposition (wet plus dry) to predict N export (Fig. 13d; $r^2 = 0.92$; Howarth 1998). Ammonium deposition is strongly tied to livestock densities (Bouwman et al. 1997), which suggests that livestock wastes contribute disproportionately to the N pollution of surface waters by agriculture and, together with the fossil-fuel source, are often major factors in N export to the coastal oceans at the scale of large regions (Howarth 1998).

Insights from the SPARROW Model Applied to the National Scale

Another useful large-scale approach to assessing sources of N and P in surface waters was taken by Smith et al. (1997). By applying the Spatially Referenced Regressions on Watersheds (SPARROW) model to a set of data from 414 stations in the National Stream Quality Accounting Network, Smith et al. (1997) concluded that just over half of the streams and rivers in the U.S. have total P concentrations in excess of 0.1 mg l^{-1} (Fig. 14). They concluded that livestock waste production was the single largest source of P contamination leading to elevated P concentrations nationally (Smith et al. 1997). Mean values for land-water delivery factors (the fraction of the original source of P that actually reaches surface waters) were estimated at approximately 0.07 and 0.11, respectively, for P from fertilizer application and P from livestock wastes. Smith et al. (1997) suggested that, per mass of P, P from livestock wastes was 50% more likely to be exported to surface waters. These delivery factors are estimated as part of the model in determining the best fit between nutrient sources and concentrations.

With respect to N, Smith et al. (1997) concluded that much of the U.S. exports less than 500 kg N km^{-2} yr^{-1}, but that export is probably much higher in much of the Mississippi River basin and in the watersheds of the northeastern U.S. (Fig. 15). For the areas of export over 1,000 kg N km^{-2} yr^{-1}, Smith et al. (1997) concluded that fertilizer was the largest source of N overall (48%), followed by atmospheric deposition (18%) and livestock wastes (15%). To some degree this result is driven by the large area of the Mississippi River basin; this basin represents 41% of the area of the lower 48 states and is a region where fertilizer application greatly exceeds NO_y deposition (Howarth et al. 1996; Goolsby et al. 1999). In the northeastern U.S. atmospheric deposition is the largest nonpoint source of N to surface waters (Howarth et al. 1996; Jaworski et al. 1997; Smith et al. 1997).

For the portions of the U.S. where total N export was over 1,000 kg N km^{-2} yr^{-1} the SPARROW model estimated land-water delivery factors of 0.24 for livestock wastes, 0.32 for fertilizer application, and 1.62 for atmospheric deposition (Smith et al. 1997). For both livestock waste and fertilizer, the delivery factors are greater for N than for P (by 2-fold to 4-fold). This is consistent with the known

greater mobility of N in dissolved forms in surface and ground water and in volatile forms in the atmosphere.

There are some biases in the land delivery factors for N fertilizer and for atmospheric deposition because Smith et al. (1997) did not include the N fixation by agricultural crops or dry deposition from the atmosphere in their analysis. N fixation by agricultural crops tends to be correlated with N fertilizer application in the U.S., and both are sources of N to downstream ecosystems (Howarth et al. 1996). Howarth et al. (1996) also demonstrated that, on average, for the portions of the U.S. that export over 1,000 kg N km^{-2} yr^{-1} N fertilizer makes up just over 60% of the sum of fertilizer application plus N fixation by agricultural crops. Adjusting the land-water delivery factor from Smith et al. (1997) to include N fixation as a source of N yields a new land-water delivery factor of 0.20 for the combined N from fertilizer and N fixation (a value comparable to that for N loss from livestock waste determined by Smith et al. [1997]).

For atmospheric deposition Smith et al. (1997) reported a land-water delivery coefficient of 1.62, suggesting that more N runs off the landscape from a depositional source than actually falls in deposition. The most likely explanation for this overestimated delivery factor is that the deposition estimates used for input were only for wet deposition of NO_y, and did not include NO_y dry deposition or wet or dry deposition of ammonium and organic N (Smith personal communication). For areas in the U.S. receiving fairly high levels of atmospheric deposition, wet NO_y deposition is approximately 25% of total atmospheric deposition (wet and dry of both reduced and oxidized forms). Although there is uncertainty associated with this estimate (Johnston and Lindberg 1992; Lovett and Lindberg 1993; Whelpdale et al. 1997; Holland et al. 1999; Valigura et al. 2000), using this value as a correction factor for the land-water delivery factor for N deposition of Smith et al. (1997) leads to a land delivery factor of approximately 0.40 in areas of high N export in the U.S.

A comparison of these revised delivery factors of 0.40 for total atmospheric deposition of N (NO_y) and 0.20 for N fertilizer application plus N fixation by agricultural crops leads to the conclusion that N from depositional sources is about twice as mobile in the landscape as is N running off agricultural fields. This conclusion is consistent with the regional analysis of inputs to the North Atlantic Ocean discussed earlier, which also demonstrated the greater mobility of N from NO_y deposition (Howarth et al. 1996). While the global mobilization of newly available N is greater through fertilizer production than through fossil-fuel combustion (Galloway et al. 1995; Vitousek et al. 1997), the N from fossil fuel sources may be disproportionately important to coastal eutrophication and other adverse effects of nutrient over-enrichment.

The conclusions reached by Smith et al. (1997) from their SPARROW analysis agree remarkably well with the conclusions of the International SCOPE Nitrogen Project (Howarth et al. 1996; Howarth 1998) with one exception. Results from the SCOPE project show that livestock wastes are a more significant source of N to surface waters than predicted by Smith et al. (1997); the SPARROW analysis finds livestock wastes to be the major source of P, but a lesser source of N. These two independent studies based on completely independent data sources largely agree on the major source terms of N in the landscape.

Nutrient Budgets for Specific Estuaries and Coastal Waters

Knowledge of nutrient inputs to an estuary is essential for management of nutrient over-enrichment problems, and nutrient budgets have now been prepared for many estuaries. Several of these have been summarized by Valigura et al. (2000). Often, nutrient inputs are estimated as part of some larger scientific research project and are published in the peer-reviewed scientific literature. More frequently, the budgets are prepared as management tools and are either not published or are published as government or consulting company reports (Valigura et al. 2000). Documentation of the data sources and approaches used is sometimes missing and is seldom fully adequate for independent review.

No standard methods exist for estimating nutrient inputs to estuaries, and many different approaches have been used. N utrient budgets are often based on export-coefficient models, where nutrient exports are estimated from literature values as a function of land-use types without independent verification of fluxes (NRC 2000). In other cases budgets are based on empirically derived loading coefficients for the actual watershed. These approaches work well for determining the importance of point source inputs such as wastewater treatment plants. Without proper calibration, estimates for nutrient inputs from nonpoint sources can be misleading. Estimating the importance of atmospheric deposition as a source is particularly problematic when using export-coefficient models.

Export-coefficient models simply take empirical data, and apply it through a series of relatively straightforward calculations to obtain an estimate of the total load. In the simplest form (which is

often the form used), the approach uses published coefficients for various land-use types, and often the coefficients are derived for regions other than that where the watershed resides. In a simple hypothetical watershed published coefficients might suggest that farmland exports X kg N km^{-2} yr^{-1}, forests export Y kg N km^{-2} yr^{-1}, and urban lands export Z kg N km^{-2} yr^{-1}. These values are multiplied by the area of each land type in the watershed to get the export for the watershed as a whole. Atmospheric deposition of N (NO_y) presents an immediate problem in that these models have historically not differentiated whether the export coefficients used were derived for areas with high or low atmospheric deposition of N. Atmospheric deposition has been ignored, and the export from forests is generally treated as a background, natural flux. This erroneously implies that no amount of atmospheric deposition of N will increase the export of N from forests. Presumably, the approach could be improved so that forest export varied depending on deposition, but to date no efforts to address this problem have been successfully completed.

Almost all nutrient budgets for estuaries rely on gauged stream discharge data when available. For many estuaries (including such major ones as Chesapeake Bay, Delaware Bay, and the Hudson River) significant portions of the watersheds are not gauged because of the difficulty in gauging tidal streams and rivers (Valigura et al. 2000). Where available, data on concentrations of total P and N are used in these budgets, but for many estuaries only inorganic dissolved nutrients are measured (Valigura et al. 2000). These problems add considerable error to the nutrient budgets.

Methods for determining the sources of nutrients and the magnitude of the load contributed by each are poorly developed at the scale of individual estuaries, and there is an urgent need for developing better approaches, particularly with regard to atmospheric deposition of N onto the landscape. The large-scale and regional analyses discussed above (the International SCOPE Nitrogen Project and the SPARROW analysis) provide a potential framework based on quantifying inputs to the watershed, but these analyses are relatively recent and have not yet been applied to the management of most estuaries. In an effort to determine the validity of using SPARROW-derived estimates for a given estuary, Valigura et al. (2000) conducted a preliminary comparison of SPARROW-derived estimates with independently derived estimates of N loading to 27 estuaries on the Atlantic and Gulf of Mexico coasts of the U.S. Based on that comparison Valigura et al. (2000) concluded that while SPARROW accurately predicted the mean loading to the estuaries as a group, it did a poor job of predicting the load to any one particular estuary (i.e., a linear regression of the SPARROW estimates and the locally derived estimates had a slope of 1 and a r^2 of 0.49). As with many such analyses involving locally derived information, the observed data from each estuary vary in quality and quantity and the methods used to calculate estimates vary as well. The locally derived estimates were not obtained from directly comparable data sets and most were not verified. The poor match between SPARROW predictions and local estimates may lie with the quality of the individual estimates for the 27 estuaries (NRC 2000).

Perhaps the greatest uncertainty with N budgets for estuaries concerns the contribution of atmospheric deposition. In most classical estuarine studies N inputs from the atmosphere were completely ignored. This has changed since Fisher and Oppenheimer (1991) pointed out the potential importance of atmospheric deposition as a source of N to Chesapeake Bay, and since Paerl (1985) showed the importance of atmospheric deposition as a N source to the coastal waters of North Carolina. Even many nutrient budgets constructed during the last decade have no estimate for the input of N from atmospheric deposition. In many other estuaries budgets estimate the importance only of direct deposition onto the surface waters of the estuary itself (and generally only wet deposition, not dry deposition) and do not estimate deposition onto the landscape with subsequent export to the estuary.

Available evidence (although constrained by limited monitoring) indicates that direct deposition onto the water surface alone (not including the contribution of N, which falls on the landscape and is then exported to estuaries) contributes between 1% and 40% of the total N input to an estuary—depending in large part on the relative area of the estuary and its watershed (Nixon et al. 1996; Valigura et al. 2000). In estuaries where the ratio of the area of the estuary to the area of its watershed is greater than 0.2, direct atmospheric deposition usually makes up 20% or more of the total N loading (Valigura et al. 2000). Where the ratio of the estuarine area to watershed area is less than 0.1, atmospheric deposition directly onto the water surface generally makes up less than 10% of the total N input (Valigura et al. 2000).

For estuaries that have relatively large watersheds, the deposition of N from the atmosphere onto the landscape with subsequent runoff into the estuary is probably greater than the deposition of N directly onto the water surface. The magnitude of this flux is poorly characterized for most estuaries. The deposition onto the landscape can

be estimated for most watersheds, although the error associated with these estimates can be considerable due to inadequate monitoring and the difficulty of measuring dry deposition. The larger problem is determining what portion of the N deposition is retained in the landscape and what portion is exported to rivers and the coast. The two major approaches for making this determination are to use statistical models or to use process-based models of N retention in the watershed. In their application to estuaries both approaches are quite recent and are relatively untested. There is an urgent need for further development and evaluation of these techniques; it appears that the statistical approaches have led to more reliable estimates, for reasons discussed below.

Both the SPARROW model and regressions comparing N flux in rivers to sources of N across landscapes (used by the International SCOPE Nitrogen Project) represent examples of statistical approaches that appear to provide reliable estimates of the portion of the N deposition retained in the landscape versus what is exported to rivers and coastal areas. Jaworski et al. (1997) used a similar approach in the northeastern U.S., comparing atmospheric deposition and riverine flux for 10 watersheds with relatively little agricultural activity or sewage inputs. This led to the conclusion that 64% of the wet inorganic deposition onto the landscape was exported to the rivers (Jaworski et al. 1997). Assuming that dry deposition is 40% of the total inorganic deposition (Jaworski et al. 1997), this corresponds to 40% of the total inorganic deposition (wet plus dry) being exported from the landscape, a value remarkably similar to the results from applying the SPARROW model at the national scale. By applying this result to other watersheds in the northeast, including those with agricultural activity, Jaworksi et al. (1997) estimated that between 36% and 80% of the total N flux in rivers was originally derived from atmospheric deposition onto the landscape.

In another recent effort a National Oceanic and Atmospheric Administration sponsored project brought together researchers from around the U.S. to examine atmospheric deposition to coastal waters (Valigura et al. 2000). Valigura et al. (2000) summarized and compared the four different approaches included in the NOAA project, including a process-based model and an application of the statistical approach used by SPARROW. They report that, for 42 estuaries in the U.S., atmospheric deposition onto the landscape contributed between 6% and 50% of the total N load to the receiving body. Jaworski et al. (1997) and Valigura et al. (2000) give estimates in common for only one river/estuary—the Hudson-Raritan—and for this system, the Jaworski et al. (1997) estimates are similar to the statistical model results, but quite different from the process-based model estimates. Jaworksi et al. (1997) estimate that 34% of the N flux in the Hudson comes from atmospheric deposition onto the landscape, after correction for the point source inputs from New York City (Hetling et al. 1996). Estimates from the process-based model indicated 9% of the N flux of the Hudson-Raritan total N load comes from N deposition onto the landscape. The statistical SPARROW model approach estimated the flux to the estuary from atmospheric deposition onto the watershed as 26% for this system.

Great uncertainty about the importance of atmospheric deposition as a N source to specific estuaries may exist. There is little doubt that the relative importance of fossil-fuel combustion versus agricultural activity in controlling atmospheric deposition of N to estuaries depends both on the nature and extent of farming activities in the watershed and on the nature and extent of fossil-fuel combustion in the airsheds upwind of the watershed. In estuaries fed by watersheds with little agricultural activity but significant loads of atmospheric pollution (such as the Connecticut and Merrimack Rivers and most of the northeastern U.S.) atmospheric deposition of N from fossil-fuel combustion can account for up to 90% or more of the N contributed by nonpoint sources. For watersheds such as the Mississippi basin where agricultural activity is high and atmospheric pollution from fossil-fuel combustion is relatively low (Fig. 16), agricultural sources dominate the fluxes of N. The major areas of agricultural activity that dominate the N fluxes for the Mississippi and Gulf of Mexico are far from the Gulf, in Iowa, Illinois, Indiana, Minnesota, Wisconsin, and Ohio (Goolsby et al. 1999).

A recent report from the Environmental Protection Agency (1999) estimates that 10% to 40% of the total N input to estuaries comes from atmospheric deposition, including deposition directly onto the water surface and the watershed. It must be stressed that few of the individual studies upon which this conclusion is based had adequate methods for determining the input of N from atmospheric deposition, particularly the indirect input through atmospheric deposition onto the landscape with subsequent runoff into the estuary. Many of these studies have probably underestimated the importance of this pathway, and it seems likely that atmospheric deposition is a greater input to estuaries than suggested by Environmental Protection Agency (1999).

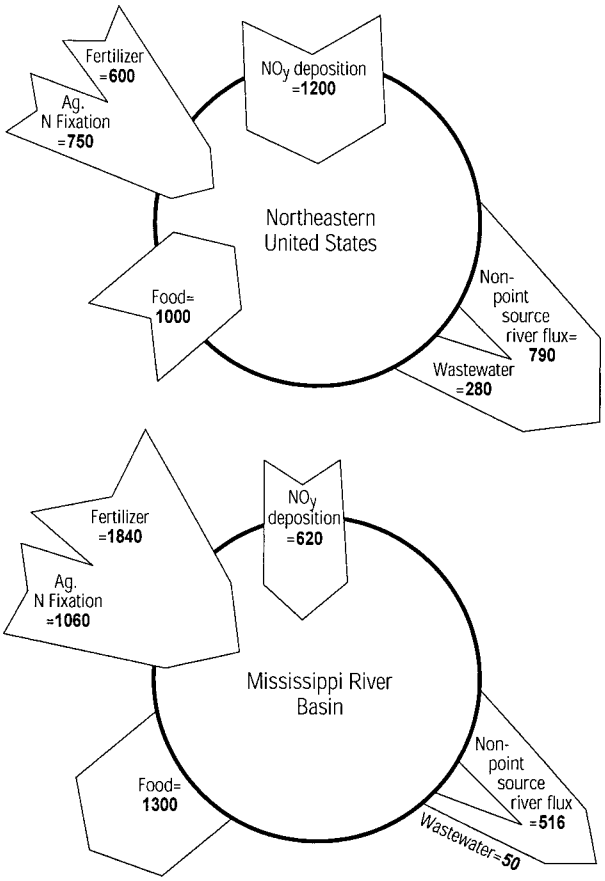

Fig. 16. A comparison of human-controlled inputs of nitrogen and nitrogen losses (kg N km^{-2} yr^{-1}) as food exports and in riverine exports between the northeastern U.S. and the Mississippi River basin (modified from NRC 2000 and based on data in Howarth et al. 1996).

Rate of Change of Nutrient Inputs to the Coast

Historical data on fluxes of total N in rivers are rare, but data for trends in nitrate concentrations are available for many rivers going back to the early 1900s. Because human activity preferentially mobilizes nitrate over other forms of N in rivers (Howarth et al. 1996), these historical nitrate data are valuable in tracking the effects of humans on N fluxes to the coast. For the Mississippi River the nitrate flux to the Gulf of Mexico is now 3-fold larger than 30 years ago, and most of this increase occurred between 1970 and 1983 (Fig. 17; Goolsby et al. 1999; Goolsby and Battaglin 2000; McIsaac et al. 2001). Nitrate fluxes in many rivers in the northeastern U.S. have increased 2-fold to 3-fold or more since 1960, with much of this increase occurring between 1965 and 1980 (Fig. 18; Jaworski et al. 1997). Most of the increase in nitrate in the Mississippi River was due to increased use of N fertilizer (Goolsby et al. 1999), whereas most of the increase in nitrate in the northeastern rivers was

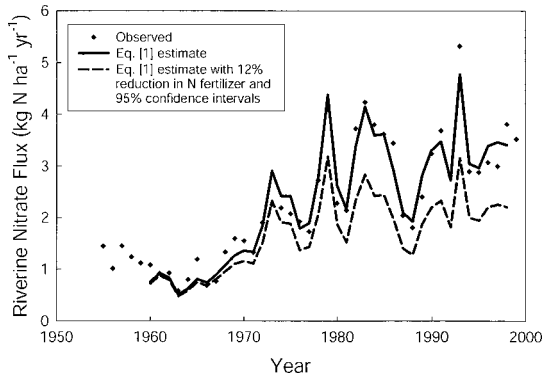

Fig. 17. Flux of nitrate down the Mississippi River to the Gulf of Mexico. Points indicate actual observations at St. Francisville, Louisiana, from 1955 to 1998. Solid line indicates the flux predicted from a simple model based on annual water yield and average annual net anthropogenic nitrogen inputs to the basin. Dashed line indicates the flux predicted from the same model, but assuming a 12% reduction in nitrogen fertilizer use throughout the time period (modified from McIsaac et al. 2001).

due to increased N deposition from the atmosphere onto the landscape, with the N originating from fossil-fuel combustion (Jaworski et al. 1997). The increase in nitrate flux in the northeastern rivers during the 1960s and 1970s and its stabilization since then closely parallels the trend in human inputs of N to the landscape during that time (Jaworski et al. 1997).

In contrast to N, P fluxes to estuaries have often changed little over the past several decades. For the Mississippi River, data on total P flux are only available since the early 1970s, but there has been no statistically significant change since (Goolsby et al. 1999). Smith et al. (1987) used data from 300 river locations throughout the U.S. to compare water quality trends from 1974 to 1981. Many rivers showed no trend during that time; rivers that had a trend in total P flux were equally divided between those that showed an increase and those that showed a decrease. Where total P fluxes increased

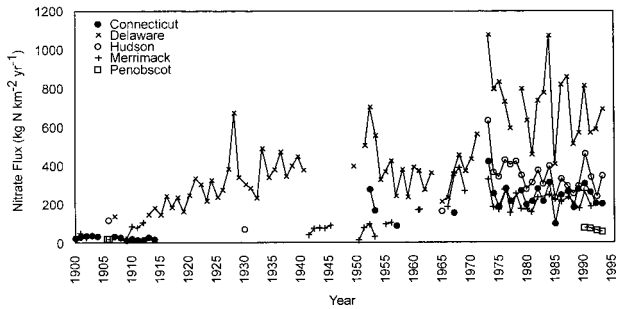

Fig. 18. Flux of nitrate nitrogen from five major rivers in the northeastern U.S. from the early 1900s to 1994 (modified from Jaworski et al. 1997).

it was generally attributable to increased use of P fertilizer in the watershed. Decreases in total P fluxes were generally a result of point source reductions (Smith et al. 1987). Smith et al. (1987) also analyzed the national river data for trends in nitrate flux from 1974 to 1981. For nitrate most rivers showed a marked increase in flux during that time, particularly for rivers in the eastern U.S. The increased nitrate flux was attributed both to agricultural activity and N deposition (Smith et al. 1987).

Conclusions and Implications for Achieving Source Reductions

Human activity has an enormous effect on the cycling of nutrients and especially on the movement of such nutrients as N and P into estuaries and other coastal waters. Although much effort has been made in the U.S. to improve control of point sources of pollution, nonpoint sources, such as urban runoff, agricultural runoff (particularly from animal feeding operations), and atmospheric deposition, are generally of greater concern in terms of effect on nutrient enrichment and eutrophication of coastal waters. While sewage inputs dominate in some estuaries, nonpoint sources dominate nationally. Insufficient effort has been expended on controlling nonpoint sources of N and P, and there are few comprehensive plans for managing nutrient enrichment of the nation's coastal waters, particularly from nonpoint sources. Efforts to manage nonpoint and point sources of N and P are needed to reduce adverse effects of nutrient over-enrichment in the nation's rivers, lakes, and coastal waters.

There is compelling evidence that both atmospheric deposition of N from fossil-fuel combustion and agricultural sources of N contribute N to coastal waters. The relative importance of these inputs varies among estuaries, but recent evidence indicates that the amount of N from deposition has been historically underestimated as an input to many estuaries, particularly by the indirect pathway of N deposited onto the landscape that is then exported to the estuary. Recent evidence suggests that per unit input to the landscape, N from fossil-fuel combustion may be more important than N from fertilizer and in turn contributes disproportionately to the input of N to coastal waters.

Much uncertainty remains regarding the fluxes of N from the atmosphere to the landscape and to estuaries, and this is a critically important research priority. N deposition and fate in urban and suburban areas is poorly known. There is only a limited understanding of dry deposition in any environment, and understanding this in coastal areas and over water is challenging.

Although understanding some details regarding the atmospheric transport and fate of biologically available N will require additional research, the significant role atmospheric deposition of N plays in nutrient over-enrichment in some regions is clear. Addressing this component of the problem will require coordinated efforts over many states, clearly dictating a federal role in the effort. The regional nature of the atmospheric component of N loading argues that nutrient management should be a significant component of efforts to reduce air pollution and should be a key consideration during re-authorization of the Clean Air Act.

Changes in agricultural production systems are concentrating large amounts of nutrients in localized areas, thereby increasing the risk of nutrient leakage to the environment. Most of this concentration is associated with animal feedlots and with the long-distance transport of feedstocks. Changes in farm practices are driven by economics, and this concentration and long-range transport provide economic advantages to the producers; the larger costs, such as the external cost of nutrient exports to estuaries, remain to be addressed.

In general, sources of nutrients to estuaries have been poorly characterized and in some cases sources have been mistakenly characterized because some land-use export-coefficient models used for characterization are inadequately verified. There are currently no easy-to-use and reliable methods for a manager of an estuary to determine the sources of nutrients flowing into that estuary. A balanced and cost-effective nutrient management strategy will require an understanding of both the relative importance of various sources of nutrients and the economic costs associated with reducing the loads attributable to each (NRC 2000).

Acknowledgments

This manuscript closely follows Chapter 5, Sources of Nutrient Inputs to Estuaries and Coastal Waters, of the report of the NRC Committee on the Causes and Management of Coastal Eutrophication (NRC 2000). Members of that committee included Robert W. Howarth (chair), Cornell University; Donald M. Anderson, Woods Hole Oceanographic Institution; Thomas M. Church, University of Delaware; Holly Greening, Tampa Bay Estuary Program; Charles S. Hopkinson, Marine Biological Laboratory; Wayne C. Huber, Oregon State University; Nancy Marcus, Florida State University; Robert J. Naiman, University of Washington; Kathleen Segerson, University of Connecticut; Andrew N. Sharpley, U.S. Department of Agriculture; William J. Wiseman, Jr., Louisiana State University. We thank each member of the committee for their thoughtful participation in producing the overall report, and for their comments on Chapter 5. The efforts of the committee were supported by the U.S. Environmental Protection Agency, the National Oceanic and Atmospheric Administration, the U.S. Geological Survey, and the Electric Power Research Institute.

Literature Cited

Aber, J. D. and C. T. Driscoll. 1997. Effects of land use, climate variation, and nitrogen deposition on nitrogen cycling and carbon storage in northern hardwood forests. *Global Biogeochemical Cycles* 11:639–648.

Aber, J. D., K. J. Nadelhoffer, P. Steudler, and J. M. Melillo. 1989. Nitrogen saturation in northern forest ecosystems. *BioScience* 39:378–386.

Aber, J. D., S. V. Ollinger, and C. T. Driscoll. 1997. Modeling nitrogen saturation in forest ecosystems in response to land use and atmospheric deposition. *Ecological Modeling* 101:61–78.

Alexander, R. B., P. J. Johnes, E. A. Boyer, and R. A. Smith. 2002. A comparison of models for estimating the riverine export of nitrogen from large watersheds. *Biogeochemistry* 57: 295–339.

Alexander, R. B., R. A. Smith, and G. E. Schwarz. 2000. Effect of stream channel size on the delivery of nitrogen to the Gulf of Mexico. *Nature* 403:758–761.

Bashkin, V. N. 1997. The critical load concept for emission abatement strategies in Europe: A review. *Environmental Conservation* 24:5–13.

Bengston, L., P. Seuna, A. Lepisto, and R. K. Saxena. 1992. Particle movement of meltwater in a subdrained agricultural basin. *Journal of Hydrology* 135:383–398.

Bock, B. R. 1984. Efficient use of nitrogen in cropping system, p. 273–294. *In* R. D. Hauck (ed.), Nitrogen in Crop Production. American Society of Agronomists, Madison, Wisconsin.

Bouwman, A. F. and H. Booij. 1998. Global use and trade of feedstuffs and consequences for the nitrogen cycle. *Nutrient Cycling in Agroecosystems* 52:261–267.

Bouwman, A. F., D. S. Lee, A. H. Asman, F. J. Dentener, K. W. van der Hoek, and J. G. J. Olivier. 1997. A global high-resolution emission inventory for ammonia. *Global Biogeochemical Cycles* 11:561–587.

Boynton, W. R., J. H. Garber, R. Summers, and W. M. Kemp. 1995. Inputs, transformations, and transport to nitrogen and phosphorus in Chesapeake Bay and selected tributaries. *Estuaries* 18:285–314.

Bredemeier, M., K. Blanck, Y. J. Xu, A. Tieteam, A. W. Boxman, B. A. Emmett, F. Moldan, P. Gundersen, P. Schleppi, and R. F. Wright. 1998. Input-output budgets at the NITREX sites. *Forest Ecology and Management* 101:57–64.

Bricker, S. B., C. G. Clement, D. E. Pirhalla, S. P. Orlando, and D. G. G. Farrow. 1999. National Estuarine Eutrophication Assessment: Effects of Nutrient Enrichment in the Nation's Estuaries. National Ocean Service, National Oceanic and Atmospheric Administration, Silver Spring, Maryland.

Campbell, J. L., J. W. Hornbeck, W. H. McDowell, D. C. Buso, J. B. Shaley, and G. E. Likens. 2000. Dissolved organic nitrogen budgets for upland, forested ecosystems in New England. *Biogeochemistry* 49:123–142.

Carpenter, S. R., N. F. Caraco, D. L. Correll, R. W. Howarth, A. N. Sharpley, and V. H. Smith. 1998. Nonpoint pollution of surface waters with phosphorus and nitrogen. *Issues in Ecology* 3:1–12.

Cleveland, C. C., A. R. Townsend, D. S. Schimel, H. Fisher, R. W. Howarth, L. O. Hedin, S. S. Perakis, E. F. Latty, J. C. von Fischer, A. Elseroad, and M. F. Wasson. 1999. Global patterns of terrestrial biological nitrogen (N_2) fixation in natural ecosystems. *Global Biogeochemical Cycles* 13:623–645.

Dodds, W. K., J. M. Blari, G. M. Henebry, J. K. Koelliker, R. Ramundo, and C. M. Tate. 1996. Nitrogen transport from tall grass prairie watersheds. *Journal of Environmental Quality* 25: 973–981.

Durka, W., E. D. Schultze, G. Gebauer, and S. Voerkelius. 1994. Effects of forest decline on uptake and leaching of deposited nitrate determined from ^{15}N and ^{18}O measurements. *Nature* 372:765–767.

Ekholm, P. 1994. Bioavailability of phosphorus in agriculturally loaded rivers in southern Finland. *Hydrobiologia* 287:179–194.

Emmett, B. A., D. Boxman, M. Bredemeier, P. Gundersen, O. J. Kjønaas, F. Moldan, P. Schleppi, A. Tietema, and R. F. Wright. 1998. Predicting the effects of atmospheric deposition in conifer stands: Evidence from the NITREX ecosystem-scale experiments. *Ecosystems* 1:352–360.

Emmett, B. A. and B. Reynolds. 1996. Nitrogen critical loads for spruce plantations in Wales: Is there too much nitrogen? *Forestry* 69:200–214.

Environmental Protection Agency. 1983. Results of the Nationwide Urban Runoff Program. U.S. Government Printing Office, Washington, D.C.

Environmental Protection Agency. 1999. Deposition of air pollutants to the great waters. Third Report to Congress. U.S. Government Printing Office, Washington, D.C.

Evans, R., L. C. Cuffman-Neff, and R. Nehring. 1996. Increases in agricultural productivity, 1948–1993. *In* Updates on Agricultural Resources and Environmental Indicators No. 6. U.S. Department of Agriculture-Economic Research Service, U.S. Government Printing Office, Washington, D.C.

Fahey, T. J., C. J. Williams, J. N. Rooney-Varga, C. C. Cleveland, K. M. Postek, S. D. Smith, and D. R. Bouldin. 1999. Nitrogen deposition in and around an intensive agricultural district in central New York. *Journal of Environmental Quality* 28:1585–1600.

Fisher, H. B. and M. Oppenheimer. 1991. Atmospheric nitrate deposition and the Chesapeake Bay estuary. *Ambio* 20:102.

Galloway, J. N., W. H. Schlesinger, C. Levy, A. Michaels, and J. L. Schnoor. 1995. Nitrogen fixation: Anthropogenic enhancement and environmental response. *Global Biogeochemical Cycles* 9:235–252.

Gardner, G. 1998. Recycling organic wastes, p. 96–112. *In* L. Brown, C. Flavin, and H. French (eds.), State of the World. W. W. Norton, New York.

Gburek, W. J. and A. N. Sharpley. 1998. Hydrologic controls on phosphorus loss from upland agricultural watersheds. *Journal of Environmental Quality* 27:267–277.

Gburek, W. J., A. N. Sharpley, A. L. Heathwaite, and G. J. Folmar. 2000. Phosphorus management at the watershed scale: A modification of the phosphorus index. *Journal of Environmental Quality* 29:130–144.

Goolsby, D. A. and W. A. Battaglin. 2000. Nitrogen in the Mississippi River basin: Estimating sources and predicting flux to the Gulf of Mexico. U.S. Geological Survey Fact Sheet, FS-135-00. U.S. Geological Survey, Reston, Virginia.

Goolsby, D. A., W. A. Battaglin, G. B. Lawrence, R. S. Artz, B. T. Aulenbach, R. P. Hooper, D. R. Kenney, and G. J. Stensland. 1999. Flux and sources of nutrients in the Mississippi-Atchafalaya River basin: Topic 3, Report for the integrated assessment of hypoxia in the Gulf of Mexico. NOAA Coastal Ocean Program Decision Analysis Series No. 17. National Oceanic and Atmospheric Administration Coastal Ocean Office, Silver Spring, Maryland.

Gundersen, P. and V. Bashkin. 1994. Nitrogen cycling. *In* N. Moldan and J. Cerny (eds.), Biogeochemistry of Small Catchments: A Tool for Environmental Research. John Wiley and Sons, Chichester, U.K.

Hauhs, M., K. Rost-Siebert, G. Ragen, T. Paces, and B. Vigerus. 1989. Summary of European data: The role of nitrogen in the acidification of soils and surface waters. *Miljorapport* 10: 92.

Heathwaite, A. L. and P. J. Johnes. 1996. The contribution of nitrogen species and phosphorus fractions to stream water quality in agricultural catchments. *Hydrological Processes* 10: 971–983.

Hedin, L. O., J. J. Armesto, and A. H. Johnson. 1995. Patterns of nutrient loss from unpolluted old-growth temperate forests: Evaluation of biogeochemical theory. *Ecology* 76:493–509.

HEDLEY, M. J. AND A. N. SHARPLEY. 1998. Strategies for global nutrient cycling, p. 70–95. *In* L. Currie (ed.), Long-Term Nutrient Needs for New Zealand's Primary Industries: Global Supply, Production Requirements and Environmental Constraints. The Fertilizer and Lime Research Centre, Massey University, Palmerston North, New Zealand.

HETLING, L. J., N. A. JAWORSKI, AND D. J. GARETSON. 1996. Comparison of input loadings and riverine export fluxes in large watersheds. *In* Third International Conference on Diffuse Pollution. International Association of Water Quality, Edinburgh, Scotland.

HOBBIE, J. E. AND G. E. LIKENS. 1973. Output of phosphorus, organic carbon, and fine particulate carbon from Hubbard Brook watersheds. *Limnology and Oceanography* 18:734–742.

HOLLAND, E. A., F. J. DENTENER, B. H. BRASWELL, AND J. M. SULZMAN. 1999. Contemporary and pre-industrial global reactive nitrogen budgets. *Biogeochemistry* 46:7–43.

HOUSE, W. A. AND H. CASEY. 1988. Transport of phosphorus in rivers, p. 253–282. *In* H. Tiessen (ed.), Phosphorus in the Global Environment. John Wiley, New York.

HOWARTH, R. W. 1988. Nutrient limitation of net primary production in marine ecosystems. *Annual Reviews in Ecology and Systematics* 19:89–110.

HOWARTH, R. W. 1998. An assessment of human influences on inputs of nitrogen to the estuaries and continental shelves of the North Atlantic Ocean. *Nutrient Cycling in Agroecosystems* 52: 213–223.

HOWARTH, R., D. ANDERSON, J. CLOERN, C. ELFRING, C. HOPKINSON, B. LAPOINTE, T. MALONE, N. MARCUS, K. MCGLATHERY, A. SHARPLEY, AND D. WALKER. 2000. Nutrient pollution of coastal rivers, bays, and seas. *Issues in Ecology* 7:1–15.

HOWARTH, R. W., G. BILLEN, D. SWAENY, A. TOWNSEND, N. JAWORSKI, K. LAJTHA, J. A. DOWNING, R. ELMGREN, N. CARACO, T. JORDAN, F. BERENDSE, J. FRENEY, V. KUDEYAROV, P. MURDOCH, AND Z. ZAHO-LINA. 1996. Regional nitrogen budgets and riverine N and P fluxes for the drainages to the North Atlantic Ocean: Natural and human influences. *Biogeochemistry* 35:75–139.

HOWARTH, R. W., E. BOYER, W. PABICH, AND J. GALLOWAY. 2002. Nitrogen use in the United States from 1961–2000, and estimates of potential future trends. *Ambio* 31:88–96.

HOWARTH, R. W., H. S. JENSEN, R. MARINO, AND H. POSTMA. 1995. Transport to and processing of phosphorus in nearshore and oceanic waters, p. 323–345. *In* H. Tiessen (ed.), Phosphorus in the Global Environment. John Wiley, New York.

ISSERMAN, K. 1990. Share of agriculture in nitrogen and phosphorus emissions into the surface waters of western Europe against the background of their eutrophication. *Fertilizer Research* 26:253–269.

JAWORSKI, N. A. 1990. Retrospective study of the water quality issues of the upper Potomac estuary. *Aquatic Science* 3:11–40.

JAWORSKI, N. A., R. W. HOWARTH, AND L. J. HETLING. 1997. Atmospheric deposition of nitrogen oxides onto the landscape contributes to coastal eutrophication in the northeast United States. *Environmental Science and Technology* 31:1995–2004.

JOHNSON, D. W. 1992. Nitrogen retention in forest soils. *Journal of Environmental Quality* 21:1–12.

JOHNSTON, D. W. AND S. E. LINDBERG (EDS.). 1992. Atmospheric Deposition and Forest Nutrient Cycling: A Synthesis of the Integrated Forest Study. Springer-Verlag, New York.

KELLOGG, R. L. AND C. H. LANDER. 1999. Trends in the potential for nutrient loadings from confined livestock operations. *In* The State of North America's Private Land. U.S. Government Printing Office, Washington, D.C.

LAJTHA, K., B. SEELY, AND I. VALIELA. 1995. Retention and leaching losses of atmospherically derived nitrogen in the aggrading coastal watershed of Waquoit Bay, Massachusetts. *Biogeochemistry* 28:33–54.

LANDER, C. H., D. MOFFITT, AND K. ALT. 1998. Nutrients Available from Livestock Manure Relative to Crop Growth Requirements. Resource Assessment and Strategic Planning Working Paper 98-1. U.S. Department of Agriculture, Natural Resources Conservation Service, Washington, D.C.

LANYON, L. E. AND P. B. THOMPSON. 1996. Changing emphasis of farm production, p. 15–23. *In* M. Salis and J. Popow (eds.), Animal Agriculture and the Environment: Nutrients, Pathogens, and Community Relations. Northeast Regional Agricultural Engineering Service, Ithaca, New York.

LEWIS, W. M. 2002. Yield of nitrogen from minimally disturbed watersheds of the United States. *Biogeochemistry* 57:375–385.

LEWIS, W. M., J. M. MELACK, W. H. MCDOWELL, M. MCCLAIN, AND J. E. RICHEY. 1999. Nitrogen yields from undisturbed watersheds in the Americas. *Biogeochemistry* 46:149–162.

LITKE, D. W. 1999. Review of phosphorus control measures in the United States and their effects on water quality. Water-Resources Investigations Report 99-4007. U.S. Geological Survey, Denver, Colorado.

LOVETT, G. M. AND S. E. LINDBERG. 1993. Atmospheric deposition and canopy interactions of nitrogen in forests. *Canadian Journal of Forest Research* 23:1603–1616.

MAGILL, A. H., J. D. ABER, J. J. HENDRICKS, R. D. BOWDEN, J. M. MELILLO, AND P. A. STEUDER. 1997. Biogeochemical response of forest ecosystems to simulated chronic nitrogen deposition. *Ecological Applications* 7:402–415.

MATSON, P. A., W. J. PARTON, A. G. POWER, AND M. J. SWIFT. 1997. Agricultural intensification and ecosystem properties. *Science* 277:504–509.

MCISAAC, G. F., M. B. DAVID, G. Z. GERTNER, AND D. A. GOOLSBY. 2001. Net anthropogenic N input to the Mississippi River basin and nitrate flux to the Gulf of Mexico. *Nature* 414:166–167.

MEADE, R. H. 1988. Movement and storage of sediment in river systems, p. 165–179. *In* A. Lerman and M. Meybeck (eds.), Physical and Chemical Weathering in Geochemical Cycles. Kluwer, Dordrecht.

NATIONAL RESEARCH COUNCIL. 1993a. Managing Wastewater in Coastal Urban Areas. National Academy Press, Washington, D.C.

NATIONAL RESEARCH COUNCIL. 1993b. Soil and Water Quality: An Agenda for Agriculture. National Academy Press, Washington, D.C.

NATIONAL RESEARCH COUNCIL. 2000. Clean Coastal Waters: Understanding and Reducing the Effects of Nutrient Pollution. National Academy Press, Washington, D.C.

NELSON, D. 1985. Minimizing nitrogen losses in non-irrigated eastern areas, p. 173–209. *In* Plant Nutrient Use and the Environment. Fertilizer Institute, Washington, D.C.

NIXON, S. W., J. W. AMMERMAN, L. P. ATKINSON, V. M. BEROUNSKY, G. BILLEN, W. C. BOICOURT, W. R. BOYTON, T. M. CHURCH, D. M. DITORO, R. ELMGREN, J. H. GARBER, A. E. GIBLIN, R. A. JAHNKE, N. J. P. OWENS, M. E. Q. PILSON, AND S. P. SEITZINGER. 1996. The fate of nitrogen and phosphorus at the land-sea margin of the North Atlantic Ocean. *Biogeochemistry* 35:141–180.

OMERNIK, J. M. 1977. Nonpoint Source–Stream Nutrient Level Relationships: A Nationwide Study. EPA-600/3-77-105. Environmental Protection Agency, Corvallis, Oregon.

PAERL, H. W. 1985. Enhancement of marine primary production by nitrogen-enriched acid rain. *Nature* 316:747–749.

PAERL, H. W. 1997. Coastal eutrophication and harmful algal blooms: The importance of atmospheric and groundwater as "new" nitrogen and other nutrient sources. *Limnology and Oceanography* 42:1154–1165.

PAERL, H. W. AND R. WHITALL. 1999. Anthropogenically derived atmospheric nitrogen deposition, marine eutrophication and harmful algal bloom expansion: Is there a link? *Ambio* 28:307–311.

PARDO, L. AND C. DRISCOLL. 1993. A critical review of mass balance methods for calculating critical loads of nitrogen for forested ecosystems. *Environment* 1:145–156.

PIONKE, H. B., W. J. GBUREK, AND A. N. SHARPLEY. 2000. Critical source area controls on water quality in an agricultural watershed located in the Chesapeake basin. *Ecological Engineering* 14:325–335.

PROSPERO, J. M., K. BARRETT, T. CHURCH, F. DENTNER, R. A. DUCE, J. N. GALLOWAY, H. LEVY, J. MODDY, AND P. QUINN. 1996. Atmospheric deposition of nutrient to the North Atlantic basin. *Biogeochemistry* 35:75–139.

RAST, W. AND G. F. LEE. 1978. Summary Analysis of the North American OECD Eutrophication Project: Nutrient Loading, Lake Response Relationships, and Trophic State Indices. EPA 600/3-78-008. U.S. Environmental Protection Agency, Corvallis, Oregon.

RYDEN, J. C., J. K. SYERS, AND R. F. HARRIS. 1973. Phosphorus in runoff and streams. *Advances in Agronomy* 25:1–45.

SCHOUMANS, O. F. AND A. BREEUWSMA. 1997. The relation between accumulation and leaching of phosphorus: Laboratory, field and modelling results, p. 361–363. *In* H. Tunney, O. T. Carton, P. C. Brookes, and A. E. Johnston (eds.), Phosphorus Loss from Soil to Water. CAB International Press, Cambridge, U.K.

SCHREIBER, J. D., P. D. DUFFY, AND D. C. MCCLURKIN. 1976. Dissolved nutrient losses in storm runoff from five southern pine watersheds. *Journal of Environmental Quality* 5:201–205.

SCHULZE, E. D., W. DE VRIES, M. HAUHS, K. ROSEN, L. RASMUSSEN, O. C. TANN, AND J. NILSSON. 1989. Critical loads for nitrogen deposition in forest ecosystems. *Water, Air, and Soil Pollution* 48:451–456.

SEITZINGER, S. P., R. V. STYLES, E. W. BOYER, R. ALEXANDER, G. BILLEN, R. HOWARTH, B. MAYER, AND N. VAN BREEMEN. 2002. Nitrogen retention in rivers: Model development and application to watersheds in the northeastern U.S. *Biogeochemistry* in press.

SHARPLEY, A. N. 1993. Assessing phosphorus bioavailability in agricultural soils and runoff. *Fertilizer Research* 36:259–272.

SHARPLEY, A. N., S. C. CHAPRA, R. WEDEPOHL, J. T. SIMS, T. C. DANIEL, AND K. R. REDDY. 1994. Managing agricultural phosphorus for protection of surface waters: Issues and options. *Journal of Environmental Quality* 23:437–451.

SHARPLEY, A. N., W. J. GBUREK, AND G. FOLMAR. 1998. Integrated phosphorus and nitrogen management in animal feeding operations for water quality protection, p. 72–94. *In* R. W. Masters and D. Goldman (eds.), Animal Feeding Operations and Ground Water: Issues, Impacts, and Solutions. National Ground Water Association, Westerville, Ohio.

SHARPLEY, A. N., M. J. HEDLEY, E. SIBBESEN, A. HILLBRICHT-ILKOWSK, W. A. HOUSE, AND L. RYSZKOWSKI. 1995. Phosphorus transfers from terrestrial to aquatic ecosystems, p. 173–242. *In* H. Tiessen (ed.), Phosphorus in the Global Environment. John Wiley, Chichester, U.K.

SHARPLEY, A. N. AND S. REKOLAINEN. 1997. Phosphorus in agriculture and its environmental implications, p. 1–54. *In* H. Tunney, O. T. Carton, P. C. Brookes, and A. E. Johnston (eds.), Phosphorus Loss from Soil to Water. CAB International Press, Cambridge, U.K.

SHARPLEY, A. N. AND J. K. SYERS. 1979. Loss of nitrogen and phosphorus in tile drainage as influenced by urea application and grazing animals. *New Zealand Journal of Agricultural* Research 22:127–131.

SIMS, J. T., R. R. SIMARD, AND B. C. JOERN. 1998. Phosphorus losses in agricultural drainage: Historical perspective and current research. *Journal of Environmental Quality* 27:277–293.

SKEFFINGTON, R. A. 1999. The use of critical loads in environmental policy making: A critical appraisal. *Environmental Science and Technology* 33:245.

SMIL, V. 2001. Enriching the Earth. MIT Press, Cambridge, Massachusetts.

SMITH, R. A., R. B. ALEXANDER, AND M. G. WOLMAN. 1987. Water-quality trends in the nation's rivers. *Science* 235:1607–1615.

SMITH, R. A., G. E. SCHWARZ, AND R. B. ALEXANDER. 1997. Regional interpretation of water-quality monitoring data. *Water Resources Research* 33:2781–2798.

TAYLOR, A. W., W. M. EDWARDS, AND E. C. SIMPSON. 1971. Nutrients in streams draining woodland and farmland near Coshocton, Ohio. *Water Resources Research* 7:81–90.

U.S. DEPARTMENT OF AGRICULTURE. 1989. Fact Book of Agriculture. Miscellaneous Publications No. 1063. Office of Public Affairs, Washington, D.C.

VALIGURA, R. A., R. B. ALEXANDER, M. S. CASTRO, T. P. MEYERS, H. W. PAERL, P. E. STACEY, AND R. E. TURNER (EDS.). 2000. Nitrogen Loading in Coastal Water Bodies. An Atmospheric Perspective. Coastal and Estuaries Series, No. 57. American Geophysical Union, Washington, D.C.

VAN BREEMEN, N., E. W. BOYER, C. L. GOODALE, N. A. JAWORSKI, S. SEITZINGER, K. PAUSTIAN, L. HETLING, K. LAJTHA, M. EVE, B. MAYER, D. VAN DAM, R. W. HOWARTH, K. J. NADELHOFFER, AND G. BILLEN. 2002. Nitrogen budgets for 16 watersheds draining to the northeastern U.S. coast: Storage and losses of nitrogen inputs. *Biogeochemistry* in press.

VAN BREEMEN, N., P. A. BURROUGHS, E. J. VELTHORST, H. F. VAN DOBBEN, T. DE WIT, T. B. RIDDER, AND H. F. R. REIJNDERS. 1982. Soil acidification from atmospheric ammonium sulphate in forest canopy throughfall. *Nature* 299:548–550.

VITOUSEK, P. M., J. ABER, S. E. BAYLEY, R. W. HOWARTH, G. E. LIKENS, P. A. MATSON, D. W. SCHINDLER, W. H. SCHLESINGER, AND G. D. TILMAN. 1997. Human alteration of the global nitrogen cycle: Causes and consequences. *Ecological Issues* 1:1–15.

VITOUSEK, P. M. AND R. W. HOWARTH. 1991. Nitrogen limitation on land and sea: How can it occur? *Biogeochemistry* 13:87–115.

VITOUSEK, P. M. AND W. A. REINERS. 1975. Ecosystem succession and nutrient retention: A hypothesis. *BioScience* 25:376–381.

WHELPDALE, D. M., P. SUMMER, AND E. SANHUEZ. 1997. A global overview of atmospheric acid deposition fluxes. *Environmental Monitoring and Assessment* 48:217–227.

WHITEHEAD, D. C. AND N. RAISTRICK. 1990. Ammonia volatilization from five nitrogen compounds used as fertilizers following surface application to soils. *Journal of Soil Science* 41:387–394.

WILLIAMS, M. W., J. S. BARON, N. CAINE, R. SOMMERFELD, AND R. J. SANFORD. 1996. Nitrogen saturation in the Rocky Mountains. *Environmental Science and Technology* 30:640–646.

ZARBOCK, H. W., A. J. JANICKI, AND S. S. JANICKI. 1996. Estimates of Total Nitrogen, Total Phosphorus, and Total Suspended Solids to Tampa Bay, Florida. Tampa Bay National Estuary Program Technical Publication #19-96. St. Petersburg, Florida.

SOURCES OF UNPUBLISHED MATERIALS

FOOD AND AGRICULTURE ORGANIZATION. 1999. FAOSTAT Agriculture Data. http://apps.fao.org/cgi-bin/nph-dp.pl?subset=agriculture.

SMITH, R. A. Personal Communication. United States Geological Survey, Reston, Virginia.

Received for consideration, January 29, 2001
Accepted for publication, January 2, 2002