# Chapter 3
# Estimating Atmospheric Nitrogen Deposition in the Northeastern United States: Relevance to Narragansett Bay

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## 3.1 Introduction

Over the past several decades, nitrogen pollution has grown to be perhaps the largest pollution problem in the coastal waters of the United States (NRC, 2000). An estimated two-thirds of the coastal rivers and bays in the country are now believed to be moderately or severely degraded from this pollution (Bricker *et al.*, 1999). The nitrogen comes from many sources, including wastewater treatment plants, agriculture, and atmospheric deposition. Often, the relative importance of these sources for particular estuaries is not well known (NRC, 2000; Alexander *et al.*, 2001; Howarth *et al.*, 2002b). Much of the effort at reducing nitrogen pollution has been directed at wastewater treatment plants, in part because these sources are so obvious. While such point sources are dominant in some estuaries, in most ecosystems the non-point sources of nitrogen from agriculture and atmospheric deposition are more important (Howarth *et al.*, 1996, 2002a,b; NRC, 2000; Alexander *et al.*, 2001). However, in estuaries with high population densities in the watershed, wastewater inputs are sometimes the single largest sources (NRC, 1993). This is the case for Narragansett Bay, as discussed by Nixon and colleagues in Chapter 5 of this volume.

The nitrogen in atmospheric deposition originates both from fossil fuel combustion and from the volatilization of ammonia to the atmosphere from agricultural sources, particularly from animal wastes in confined animal feedlot operations. The importance of this source was virtually unrecognized before the pioneering paper by Fisher and Oppenheimer (1991) noted that the nitrate anion associated with nitric acid in acid rain may be a major source of nitrogen to Chesapeake Bay. Since then, the focus on atmospheric deposition as a source of nitrogen has intensified, and generally, estimates of the importance of this source have tended to increase over time as it has received more attention.

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## 3.2 Atmospheric Deposition as a Nitrogen Source to Coastal Waters

For the United States as a whole, we have estimated that atmospheric deposition of nitrogen that originates from fossil-fuel combustion contributes 30% of the total nitrogen inputs to coastal marine ecosystems, while another 10% of these nitrogen inputs come from ammonia volatized into the atmosphere from agricultural sources (Howarth and Rielinger, 2003). The rest of the nitrogen inputs to coastal waters come from runoff from agricultural sources (44%) and from municipal and industrial wastewater streams (~16%).

Some of the nitrogen from atmospheric deposition is deposited directly onto the surface of coastal waters. This direct deposition to surface waters often contributes between 1% and 40% of the total nitrogen inputs to coastal ecosystems (Nixon *et al.*, 1996; Paerl, 1997; Howarth, 1998; Paerl and Whitall, 1999; Valigura *et al.*, 2000). The direct deposition is most significant in very large systems, such as the Baltic Sea (Nixon *et al.*, 1996) or in coastal systems such as Tampa Bay which have relatively small watersheds in comparison to the area of their surface waters (Zarbock *et al.*, 1996).

In most coastal marine ecosystems, the major route whereby atmospheric deposition contributes nitrogen is not direct deposition onto surface waters, but rather deposition onto the terrestrial landscape with subsequent downstream export in streams and rivers. As discussed below, these fluxes are difficult to measure, leaving significant uncertainty and debate about their magnitude. In the northeastern US as a whole (Gulf of Maine through Chesapeake Bay), our studies have suggested that atmospheric deposition is the single largest source of nitrogen to coastal waters (Howarth *et al.*, 1996; Jaworski *et al.*, 1997; Boyer *et al.*, 2002), while other studies have concluded atmospheric nitrogen deposition is the second largest source after wastewater discharges from sewage treatment plants (Driscoll *et al.*, 2003). Our approach leads to the conclusion that atmospheric deposition of nitrogen onto the landscape—considering only the deposition of oxidized nitrogen compounds that originate from fossil fuel combustion (NOy)—contributes between 25% and 80% of the nitrogen flux in the different major rivers of New England (Fig. 3.1, Boyer *et al.*, 2002; Howarth and Rielinger, 2003) and approximately 25% of the nitrogen flux in the Mississippi River (NRC, 2000; Howarth *et al.*, 2002b). Using another approach—SPARROW, or Spatially Referenced Regression on Watershed attributes model—Alexander *et al.* (2001) concluded that atmospheric deposition onto the landscape contributed between 4% and 35% of the nitrogen flux in 40 major coastal watersheds across the United States, with the highest contribution in the northeastern and mid-Atlantic regions. As discussed later in this paper, the SPARROW model may significantly underestimate the role of deposition near emission sources.

The uncertainty over the contribution of atmospheric deposition as a nitrogen source to coastal marine ecosystems stems from two issues: uncertainty over

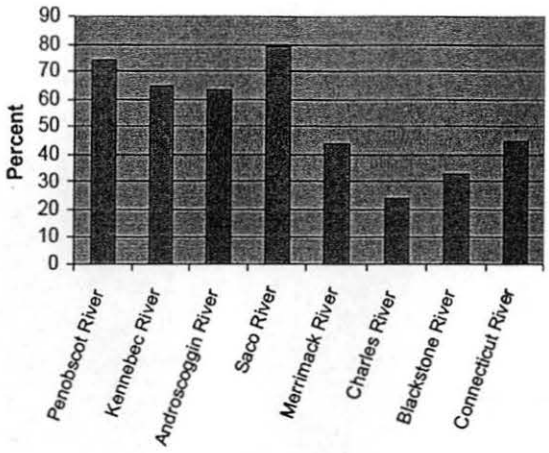


**Fig. 3.1** Percentage of nitrogen in major New England rivers that originates from fossil-fuel derived atmospheric deposition onto the landscape. Reprinted from Howarth and Rielinger (2003), based on data in Boyer *et al.* (2002)

the magnitude of nitrogen deposition onto watersheds, particularly from "dry deposition", and uncertainty over the amount of the deposited nitrogen that is subsequently exported downstream (NRC, 2000; Howarth *et al.*, 2002b). Each of these is discussed in some detail in the following sections.

## 3.3 Dry Deposition of Nitrogen as a Source

The vast majority of measurements of nitrogen deposition in the United States—including those made by the National Atmospheric Deposition Program (NADP)—measure only "wet deposition" (i.e., nitrogen in rainfall and snow). To estimate wet deposition onto an entire watershed, data at particular monitoring sites are extrapolated statistically considering factors such as local topography and precipitation (Ollinger *et al.*, 1993; Grimm and Lynch, 2005).

Substantial quantities of nitrogen can be deposited from the atmosphere as "dry deposition," which includes aerosols and other particles and uptake of gaseous forms of nitrogen by vegetation, soils, and surface waters. Both in the United States and Europe, the extremely sparse spatial coverage in networks for measuring dry deposition severely limits estimation of this process (Holland *et al.*, 2005). In the United States, dry deposition is routinely estimated only at sites that are part of the CASTNet and AIRMon-Dry programs. At the peak of these programs in the 1990s, these networks consisted of a total of 93 sites across the country, but the number is now down to 70 (http://www.epa.gov/castnet/). In the watersheds of Chesapeake Bay—an area of 165,000 $km^2$ that includes land in 6 states—there are only 8 stations for monitoring dry deposition. In New England, there are only 6 stations, with 3 in Maine and only one in southern New England. The vast majority of these dry deposition monitoring

stations across the country—and all of them in New England and New York State—are purposefully located far from sources of nitrogen emissions to the atmosphere.

In addition to the limited spatial extent of the dry deposition monitoring networks, these networks do not measure all of the components that can be deposited. For example, particulate $NO_3^-$ and $NH_4^+$ are routinely measured, as is nitric acid vapor. However, other gaseous nitrogen compounds that may play a significant role in deposition (i.e., NO, $NO_2$, HONO, peroxy and alkyl based organics, and ammonia gas) are not measured. NO and $NO_2$ are the major gases emitted from fossil fuel combustion, while ammonia is the major form of air pollution from agricultural sources. Ammonia is also released in vehicle exhaust, although at lesser amounts than for NO and $NO_2$ (Baum *et al.*, 2001; Cape *et al.*, 2004). To the extent these compounds are deposited, the dry depositional monitoring networks are underestimating total deposition. As currently measured, the dry deposition at the 8 CASTNet sites in the Chesapeake Bay watershed ranges from 23% to 38% of total deposition (T. Butler, pers. comm.), but the actual contribution when all forms of nitrogen gases are considered must certainly be higher.

The manner in which dry deposition rates are calculated—multiplying concentration data obtained at the monitoring sites by "depositional velocities"—may also result in underestimation of this process. For the AIRMon and CASTNet sites, these deposition velocities are estimated as a function of vegetation and meteorological conditions (Clarke *et al.*, 1997). Our knowledge of depositional velocities is based on studies in flat, homogenous terrain; as noted by Bruce Hicks (former Director of the NOAA Air Resources Lab), when estimating dry deposition "we are simulating the world on the assumption that our understanding of [these] special cases applies everywhere. We often display unwarranted confidence" in our estimates (Hicks presentation to the annual meeting of the American Society of Meteorology, October 2005). Complex terrain is likely to substantially increase depositional velocities. Vegetative cover is also important, and different models can vary in their estimates of spatial integrated dry deposition by more than 5-fold depending upon different assumptions of the effect of vegetation (particularly coniferous forests) on depositional velocities (Wesely and Hicks, 1999; Holland *et al.*, 2005).

## 3.4 Estimation of Total Nitrogen Deposition in the Northeastern US

Boyer *et al.* (2002) estimated the average deposition of oxidized nitrogen (NOy) onto the landscape of the major rivers of the northeastern United States (including both wet and dry deposition) following the approach of Ollinger *et al.* (1993) in using a statistical extrapolation of deposition monitoring data.

They estimated a range of values across these watersheds from ~360 kg N $km^{-2}$ $yr^{-1}$ in the Penobscot River basin in Maine to ~890 kg N $km^{-2}$ $yr^{-1}$ in the Schuylkill River basin in Pennsylvania (Boyer *et al.*, 2002). The average value for this set of watersheds was ~680 kg N $km^{-2}$ $yr^{-1}$.

Another approach for estimating nitrogen deposition onto the landscape can be obtained from models based on emissions to the atmosphere, with consideration of reaction and advection in the atmosphere, followed by deposition. We used one of these models (the GCTM model; Prospero *et al.*, 1996) to estimate nitrogen deposition in all of the regions—including the northeastern United States—that surround the North Atlantic Ocean (Howarth *et al.*, 1996). The GCTM model predicts depositional patterns globally at a relatively course spatial scale using emission sources as inputs and modeling atmospheric transformations and transport (Prospero *et al.*, 1996). For the northeastern United States, the GCTM model yielded an estimated total NOy deposition (wet plus dry) of ~1,200 kg N $km^{-2}$ $yr^{-1}$, a value 80% greater than that derived by Boyer *et al.* (2002) from extrapolation of deposition monitoring data (Fig. 3.2, Howarth *et al.*, in press). A similar, more recent emission-based model (TM3)

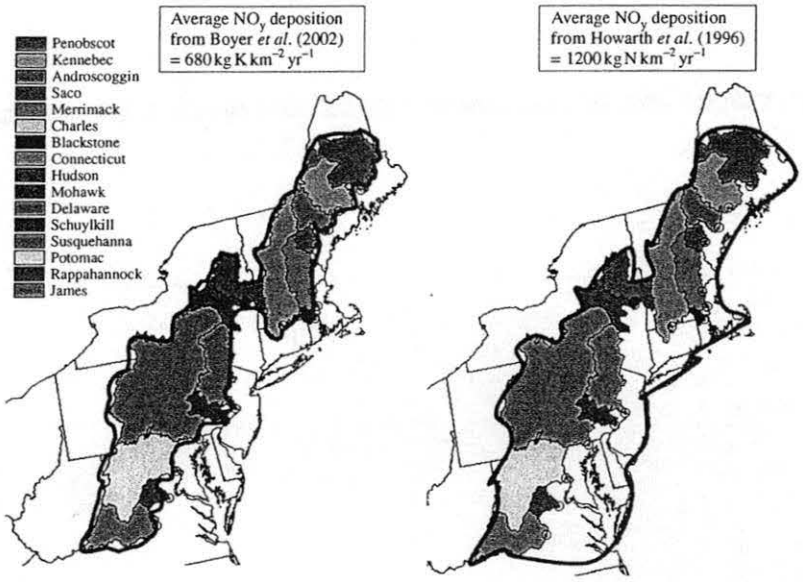


**Fig. 3.2** The geographic area considered by Boyer *et al.* (2002) was the area of 16 watersheds in the northeastern United States upriver from the lowest gauging station of the USGS (left). The area considered by Howarth *et al.* (1996) is somewhat larger, and includes the area on the coastal plain (right). Note that the average estimates for deposition of oxidized nitrogen pollution originating from fossil fuel combustion is ~80% greater in the Howarth *et al.* (1996) analysis, probably due to different approaches used for the estimation and/or the different area considered

developed by Frank Dentener and colleagues, and used by Galloway *et al.* (2004) for their global and regional nitrogen budgets, yields a comparable estimate for the northeastern United States as did the GCTM model (Howarth *et al.*, in press). These emission-based models are attractive, in that at least at very course spatial scales, they are as accurate as the emission data. However, these models are computationally demanding, and until very recently, had not been applied at a spatial scale fine enough to give estimates for the individual 16 northeastern watersheds. A new effort by NOAA/EPA's Atmospheric Sciences Modeling Division uses emissions data and the CMAQ model to estimate nitrogen deposition at a 36-km grid, but the model is still being tested as of late 2006 (presentation by R. Dennis at the National Atmospheric Deposition Program annual Technical Committee meeting, October 2006). This approach shows great promise for the future. Preliminary comparisons of this fine-scale model with the coarser scale output from GCTM and TM3 have shown good agreement (R. Dennis, pers. comm.).

Why is the estimate from the emission-based model (Howarth *et al.*, 1996) so much greater than that from estimates based on extrapolation of the wet deposition monitoring data (Boyer *et al.*, 2002)? There are three possible explanations, which are not mutually exclusive.

First, deposition on the relatively urbanized coastal plain may be much greater than in the watersheds away from the coast. The watershed areas considered by Boyer *et al.* (2002) are upriver from the coast and tend to be more rural than is the coastal plain downstream (Fig. 3.2). Recent studies have found evidence that deposition near emission sources can be much greater than deposition away from emission sources. For example, deposition within New York City was more than twice as high than in more rural areas to the north of the city (Lovett *et al.*, 2000), and deposition in the immediate vicinity of roads was much higher than a few hundred meters away (Cape *et al.*, 2004; presentation by R. Howarth, R. Marino, N. Bettez, E. Davidson, and T. Butler at the National Atmospheric Deposition Program annual Technical Committee meeting, October 2006);

Second, the estimate based on deposition monitoring data (Boyer *et al.*, 2002) may underestimate total deposition. This is of course likely, to the extent that dry deposition is underestimated. As noted above, not all of the important gases that may be deposited are routinely measured by the dry deposition monitoring networks, and depositional velocities may be underestimated in regions with major terrain features. Further, the deposition networks were not designed to measure deposition in the immediate vicinity of emission sources. In fact, most of the NADP wet deposition monitoring sites and most of the CASTNet dry depositon sites are intentionally located far away from urban emission sources.

Third, the estimate from emission-based modeling (Howarth *et al.*, 1996) may overestimate total deposition. This could occur if emissions are overestimated, which may well be true for ammonia emissions, but probably not for emissions of oxidized nitrogen to the atmosphere in the United States (Holland

*et al.*, 1999). The difference between the Howarth *et al.* (1996) and Boyer *et al.* (2002) estimates highlighted in this paper is for deposition of oxidized nitrogen (NOy). Alternatively, emission-based modeling may not accurately capture the spatial pattern of the deposition. These models rely on a mass balance of nitrogen in the atmosphere, so global deposition estimates are as accurate as the emissions data that feed them. However, deposition may be underestimated in some regions and correspondingly overestimated elsewhere.

Obviously, significant uncertainty exists in the overall magnitude of total nitrogen deposition in an area such as the northeastern United States. When considering the differences detailed above, it is important to note that extrapolations based on deposition monitoring (Ollinger *et al.*, 1993; Grimm and Lynch 2005) do not appear to capture any evidence of higher deposition near urban centers and transportation corridors. For reasons discussed in detail following, I believe it likely that traditional approaches that use deposition monitoring data to estimate total nitrogen deposition result in substantial underestimates, especially for total nitrogen deposition in the urbanized portions of the northeastern United States.

## 3.5 Using Throughfall to Estimate Total Nitrogen Deposition

The difficulty with measuring dry deposition of N (particularly of gaseous forms such as NO, $NO_2$, and $NH_3$) has led some investigators to use tree-canopy throughfall as a surrogate for total N deposition (Lajtha *et al.*, 1995; Lovett *et al.*, 2000; Weathers *et al.*, 2006; Schmitt *et al.*, 2005). Throughfall is the material that falls through the canopy of a forest, and so includes whatever is deposited on the canopy in both wet and dry deposition, plus or minus the net exchange of material with the vegetation. Most studies have found that the assimilation of nitrogen from deposition into leaves of the canopy is generally as great as or greater than the leaching of nitrogen out of leaves (Lindberg *et al.*, 1990; Johnson, 1992; Lovett and Lindberg, 1993; Dise and Wright, 1995; Lajtha *et al.*, 1995). Consequently, many experts on atmospheric deposition have argued that throughfall measurements provide a minimum estimate of total nitrogen deposition (Lindberg *et al.*, 1990; Johnson, 1992; Lovett and Lindberg, 1993; Dise and Wright, 1995; Lajtha *et al.*, 1995; Lovett *et al.*, 2000; Schmitt *et al.*, 2005).

The estimation of total nitrogen deposition from throughfall measurements can yield much higher rates than those inferred from extrapolation of deposition monitoring data. For example, in a forest in Falmouth, MA, on Cape Cod, Lajtha *et al.* (1995) measured wet deposition of 420 kg N $km^{-2}$ $yr^{-1}$ and estimated a total deposition rate of 840 kg N $km^{-2}$ $yr^{-1}$ by assuming that dry deposition equaled wet deposition. This estimate is quite similar to the deposition predicted for that location by the spatial extrapolation of Ollinger *et al.*

(1993). However, from their throughfall data, Lajtha *et al.* (1995) estimated that actual total nitrogen deposition at the site was 1,310 kg N $km^{-2}$ $yr^{-1}$, or more than 50% greater. In a more recent study, Weathers *et al.* (2006) compared throughfall data with more traditional approaches for estimating nitrogen deposition in Acadia National Park in Maine and in the Great Smoky Mountains National Park in North Carolina. In both locations, they found that total nitrogen deposition rates estimated from their throughfall data were 70% greater than those estimated from NADP and CASTNet wet and dry monitoring data. These throughfall estimates lend strength to the argument that the traditional approaches for estimating total deposition—such as we used in Boyer *et al.* (2002)—yield values that are too small.

## 3.6 The Fate of Nitrogen Deposited onto the Landscape

Forests are the dominant land cover in the northeastern United States (Boyer *et al.*, 2002), and so much of the nitrogen deposited onto the landscape falls on forests. Only a portion of this nitrogen is exported downstream, with much retained in the forests or denitrified and converted to non-reactive, molecular $N_2$. Productivity of most forests in the United States is limited by the supply of nitrogen (Vitousek and Howarth, 1991), so as forests receive more nitrogen from atmospheric deposition, production and storage of nitrogen in organic matter can be expected to increase. On average for the northeastern United States, approximately 60% to 65% of the nitrogen inputs to forests through natural nitrogen fixation as well as atmospheric deposition are retained in the forest (primarily accreted in woody biomass) or harvested from the forests in wood (Goodale *et al.*, 2002; van Breemen *et al.*, 2002). A little over 20% is exported from the forest in streams (primarily as nitrate, but also dissolved organic nitrogen), with the rest denitrified (van Breemen *et al.*, 2002). The ability of forests to store nitrogen, however, is limited, and forests can become nitrogen saturated when inputs exceed the needs of trees and the ability for soils to assimilate nitrogen (Aber *et al.*, 1989; Gundersen and Bashkin, 1994; Emmett *et al.*, 1998). Nitrogen export downstream can then increase dramatically (Emmet *et al.*, 1998; Howarth *et al.*, 2002b; Aber *et al.*, 2003).

A recent comparative study suggests that for the forests of northern New England and New York State, the nitrate concentrations in streams and small lakes just downstream increase dramatically as total nitrogen deposition increases above 600 to 800 kg N $km^{-2}$ $yr^{-1}$ (Fig. 3.3, Aber *et al.*, 2003), indicating a substantial increase in nitrogen export from the forests receiving the higher deposition. Figure 3.3 also indicates the estimated average NOy deposition for the northeastern United States in the Boyer *et al.* (2002) and Howarth *et al.* (1996) studies. Note that total deposition, including ammonia, ammonium, and organic nitrogen, would be greater by 20 to 40% (Boyer *et al.*, 2002; Howarth

*et al.*, 1996), but is also much more uncertain (Holland *et al.*, 1999; Howarth *et al.*, in press), so I have chosen to illustrate just the NOy component. Note also that the deposition estimates used in the Aber *et al.* (2003) analysis may also be low, since these are based on extrapolation of monitoring data. On the other hand, all of the data in the analysis of Aber *et al.* (2003) are from fairly rural sites, relatively far from emission sources; their deposition estimates may therefore be fairly reliable. Regardless, Fig. 3.3 suggests that nitrogen deposition onto the landscape on average in the northeastern United States is likely high enough to result in elevated losses of nitrogen from forests, particularly if the higher emission-based estimates used by Howarth *et al.* (1996) are valid.

While forests are often retentive of nitrogen, impermeable surfaces such as roads and parking lots are far less so. While not often studied, nitrogen runoff from these surfaces can be substantial. For example, runoff from highways near Providence, RI, is reported to be 1,700 kg N $km^{-2}$ $yr^{-1}$ of road surface (Nixon *et al.*, 1995). Most if not all of this nitrogen likely originated from atmospheric deposition, much of it from vehicle emissions on the highway.

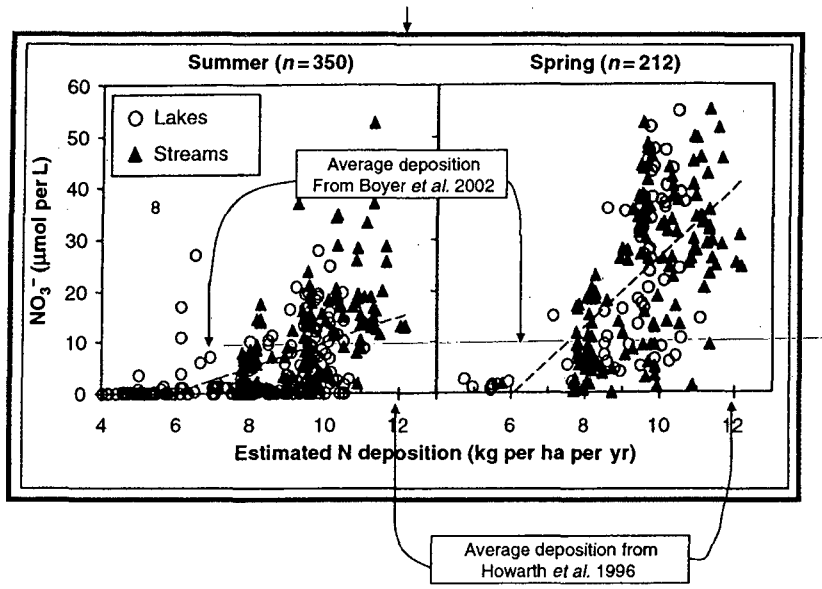


**Fig. 3.3** Concentrations of nitrate in small streams and lakes in forested catchments in northern New England in the spring (right) and summer (left) as a function of $NO_y$ deposition onto the landscape. Observe the non-linear response, with nitrate concentrations tending to increase as deposition exceeds 6–8 kg N per hectare per year (600–800 kg N $km^{-2}$ $yr^{-1}$). Arrows indicate the average deposition rates for oxidized nitrogen compounds ($NO_y$) estimated for the northeastern United States in Boyer *et al.* (2002) and Howarth *et al.* (1996), respectively. Modified from Aber *et al.* (2003)

## 3.7 A Closer Look at the SPARROW model

The SPARROW model is one of the best available tools for estimating the sources of nitrogen pollution in particular watersheds (NRC, 2000). The model statistically relates water quality data from US Geological Survey monitoring programs to spatial data on nutrient sources, landscape characteristics such as temperature and soil permeability, and stream properties such as residence time (Smith *et al.*, 1997). As noted previously in this chapter, the SPARROW model has been used to suggest that atmospheric deposition contributes from 4 to 35% of the total nitrogen inputs to a variety of US estuaries (Alexander *et al.*, 2001). One limitation of the SPARROW model as used in the Alexander *et al.* (2001) paper is that it used only wet deposition monitoring data as input for atmospheric deposition as a nitrogen source. Dry deposition data were not used, probably because the sparse spatial coverage of available data would have weakened the statistical analysis too greatly. In the SPARROW approach, the wet deposition data can serve as a surrogate for total deposition, if wet and dry deposition patterns are correlated in space (Howarth *et al.*, 2002b). However, increasingly it seems that wet and dry deposition are not correlated, and dry deposition is proportionately more important in more dry climates (Holland *et al.*, 1999) and in closer proximity to emission sources (presentation by R. Dennis at the National Atmospheric Deposition Program annual Technical Committee meeting, October 2006). This is probably particularly true for nitrogen from vehicle emissions, since relatively reactive gases are released very close to land and vegetation surfaces (Cape *et al.*, 2004; presentation by R. Howarth, R. Marino, N. Bettez, E. Davidson, and T. Butler at the National Atmospheric Deposition Program annual Technical Committee meeting, October 2006). Thus, the atmospheric deposition estimates given by the SPARROW model probably are low since they do not well represent dry deposition near emission sources.

In the version of the SPARROW model used by Alexander *et al.* (2001) to determine the relative importance of various sources of nitrogen inputs to estuaries, one of the identified sources of nitrogen pollution is called "non-agricultural non-point sources." This is nitrogen that is statistically associated with urban and suburban areas, but is not well represented by other nitrogen sources, such as wet deposition as indicated in the NADP monitoring program. Some of this nitrogen may come from home fertilizer use or from general disturbance of the landscape, but I suggest that much of it—perhaps even most of it—may in fact be associated with the dry deposition of nitrogen near vehicle emission sources. If so, the true estimate of the importance of atmospheric deposition as a nitrogen source to coastal systems may be better represented by the sum of the SPARROW estimates for atmospheric deposition and for non-agricultural non-point sources. This combined estimate ranges from 26% to 76% of the total nitrogen inputs to some representative coastal marine ecosystems in the northeastern United States (Table 3.1).

**Table 3.1** Estimates from the SPARROW model for the relative importance of atmospheric deposition, "non-agricultural non-point sources," and sewage wastewater as nitrogen inputs to several coastal marine ecosystems in the northeastern United States

| | Atmosphere | Non-agricultural non-point | Wastewater |
|---|---|---|---|
| Casco Bay | 22 | 54 | 13 |
| Great Bay | 9 | 58 | 23 |
| Merrimack River | 28 | 43 | 20 |
| Buzzards Bay | 12 | 14 | 63 |
| Narragansett Bay | 10 | 19 | 62 |
| Hudson River | 26 | 21 | 40 |
| Barnegat Bay | 19 | 28 | 43 |
| Delaware Bay | 22 | 17 | 35 |
| Chesapeake Bay | 28 | 22 | 8 |

Note that the atmospheric deposition terms are estimated just from wet deposition monitoring data. Note further that the "non-agricultural non-point sources" may include a substantial amount of input from dry atmospheric deposition near emission sources in urban and suburban environments, and this would not be included in the SPARROW estimate of the atmospheric deposition input. See text for further discussion. Based on Alexander *et al.* (2001). Values are percents (%).

## 3.8 Chesapeake Bay Case Study

Chesapeake Bay is the largest estuary in the United States, and one of the most sensitive to nutrient inputs (Bricker *et al.*, 1999; NRC, 2000). Nitrogen inputs to the Chesapeake have caused widespread loss of seagrasses and have greatly increased the volume of anoxic bottom waters (Boesch *et al.*, 2001). The role of atmospheric deposition as a source of nitrogen to the Chesapeake apparently was not considered until Fisher and Oppenheimer (1991) suggested that it may contribute 40% of the total inputs. Their analysis was simple and preliminary, and was not believed by many scientists who worked on Chesapeake Bay water quality. The most recent analyses by the Chesapeake Bay Program, while giving lower percentages, also suggest that deposition is important, contributing ~25% of the total nitrogen inputs to Chesapeake Bay (7% from direct deposition onto surface waters, and 19% from deposition onto the landscape with subsequent export to the bay ecosystem, using 2003 values; http://www.chesapeakebay.net/status.cfm?SID = 126; see also http://www.chesapeakebay.net/nutr1.htm).

Two lines of evidence suggest that the Chesapeake Bay Program model may be underestimating the inputs of nitrogen from atmospheric deposition: 1) the model may be underestimating the magnitude of deposition onto the landscape; and 2) the model may be underestimating the percentage of deposition onto the landscape that is subsequently exported downstream. Each of these is discussed below.

The Chesapeake Bay Program model relies on an estimate of total nitrogen deposition onto the watersheds of 1,210 kg N $km^{-2}$ $yr^{-1}$ (calculated from Fig. A-4 of EPA, 2003). The approach to derive this estimate is very similar to that used

by Boyer *et al.* (2002): extrapolation from deposition monitoring data for the 15 NADP wet sites and 8 CASTNet and Airmon dry deposition sties in the watersheds of the Chesapeake (Lewis Linker, Bay Program modeling coordinator, PowerPoint presentation by conference call, January 9, 2006), although the Boyer *et al.* (2002) estimate is in fact somewhat lower (1,010 kg N $km^{-2}$ $yr^{-1}$ for the area-weighted mean for the watersheds of the Susquehanna, Potomac, Rappannnock, and James Rivers up river of the USGS gaging stations). If we assume that the Boyer *et al.* (2002) estimate underestimates by 80% (based on comparison with the global-scale emission-based model used by Howarth *et al.*, 1996), then actual deposition on the Chesapeake watersheds may be as great as 1,550 kg N $km^{-2}$ $yr^{-1}$ (28% greater than assumed for the Chesapeake Bay Program model). This higher estimate is broadly consistent with the preliminary model runs from the CMAQ emission-based model discussed above (R. Dennis, pers. comm.). Note also that locally derived emissions from commercial chicken houses on the Delmarva Peninsula may contribute to the atmospheric deposition load to Chesapeake Bay (Siefert *et al.*, 2004), and this source is not well considered in the Chesapeake Bay Program model.

Perhaps of greater significance is the treatment of nitrogen retention in the landscape by the Chesapeake Bay model which assumes on average that 86% to 89% of total nitrogen deposition onto the landscape is retained, and only 11% to 14% is exported downstream to the Bay (calculated from Figure A-4, EPA, 2003). Most of this retention is assumed to occur in the 57% of the area of the watershed that is forested, with greater export of deposition onto agricultural lands and urban and suburban areas with impermeable surfaces. The model assumes that most of the forests in the Chesapeake Bay basin are not nitrogen saturated, and therefore leak little if any nitrogen (EPA, 2003).

The average export of nitrogen deposition from all land uses (12%) seems low in comparison with the estimate that average forests in the northeastern United States export over 20% of nitrogen deposition (Goodale *et al.*, 2002; van Breemen *et al.*, 2002). If the deposition in the Chesapeake basin is evenly distributed over land uses, then 43% falls on other land uses where much higher rates of export would be expected. If much of the deposition from nitrogen pollution that originates from vehicles falls near these emission sources (either onto impermeable surfaces or onto vegetation where the rate of deposition would be very high), then very high rates of export might be expected. The preliminary runs of the CMAQ model indeed suggests high deposition—particularly for dry deposition—near heavily populated urban areas. Obtaining better data on nitrogen retention in mixed land-use watersheds has been identified as a high national research need in a multi-agency federal planning document (Howarth *et al.*, 2003). But given current knowledge, it is probably as reasonable to assume that the percent export from atmospheric deposition onto the landscape of the Chesapeake Bay basin—including all land uses—is 30% as to assume the 12% used by the Chesapeake Bay model. Ranges from 20% to 40% and even higher can be reasonably inferred from studies of large watersheds (NRC, 2000; Howarth *et al.*, 2002b, in press; Boyer *et al.*, 2002).

Table 3.2 illustrates the sensitivity of nitrogen loading to Chesapeake Bay given various assumptions on the rate of deposition and on nitrogen retention in the landscape. Within this range of reasonable assumptions, the total input of nitrogen to Chesapeake Bay (both directly onto the surface waters and indirectly from deposition onto the landscape and subsequent export downstream) ranges from 34 to 92 thousand metric tons of nitrogen per year, and comprises from 25% to 50% of the total nitrogen load to Chesapeake Bay from all sources. Note that this is similar to the range of 28% to 50% determined from the SPARROW model for Chesapeake Bay (with the upper range including the "non-agricultural non-point sources; Table 3.2). Under the assumptions of greater deposition and lower retention in the landscape, the estimate for total nitrogen load to Chesapeake Bay increases substantially—from 130 to 188 thousand metric tons per year, or 45% greater total nitrogen load. Perhaps surprisingly, monitoring of the load of nitrogen to Chesapeake Bay is not adequate to constrain this total load estimate within this range of uncertainty. As with many other large coastal marine ecosystems, significant portions of the watersheds of Chesapeake Bay are not gaged because of the difficulty in gaging tidal streams and rivers (Valigura *et al.*, 2000; NRC, 2000; Howarth *et al.*, 2002b). These areas of the watershed are therefore not monitored for their nutrient inputs to the Bay. While the fluxes of nitrogen from the watersheds above gaging stations in the Chesapeake Basin are reasonably well known, the fluxes from the watershed in the more urbanized areas on the coastal plain—where nitrogen deposition may

**Table 3.2** Importance of atmospheric deposition as a source of nitrogen pollution to Chesapeake Bay under various assumptions. Fluxes are thousands of metric tons of nitrogen per year. Percentage values given in parentheses are percentages of total nitrogen load. The baseline run assumptions are from EPA (2003)

| | Total Load to Bay | Input to Bay from Direct Deposition onto Bay Water Surface | Input to Bay from Deposition onto Watersheds | Total Input to Bay from Deposition |
|---|---|---|---|---|
| Chesapeake Bay model (2000 conditions) | 130 | 9 (7%) | 25 (19%) | 34 (26%) |
| Deposition increased to 1,550 kg N $km^{-2}$ $yr^{-1}$ no change in retention assumptions | 140 | 12 (9%) | 32 (23%) | 44 (32%) |
| Chesapeake Bay model assumptions on deposition rate; assume 70% retention in landscape | 168 | 9 (5%) | 63 (38%) | 72 (43%) |
| Deposition increased to 1,550 kg N $km^{-2}$ $yr^{-1}$; assume 70% retention in landscape | 188 | 12 (6%) | 80 (43%) | 92 (49%) |

be much greater, and retention of nitrogen in the landscape much less—are estimated only from models and not from empirical monitoring data.

## 3.9 Application to Narragansett Bay

During the 1980s and early 1990s, Narragansett Bay received an average input of nitrogen of 29 g N $m^{-2}$ $yr^{-1}$ (when normalized over the entire surface area of the Bay; Nixon *et al.*, 1995; note that this corresponds to 29,000 kg N $m^{-2}$ $yr^{-1}$; in this paper, I express loadings per area of coastal ecosystem water surface in units of g N $m^{-2}$ $yr^{-1}$ and deposition of nitrogen onto the terrestrial landscape in units of kg N $km^{-2}$ $yr^{-1}$ so as to clearly distinguish the two). This estimate includes an input of 1.3 g N $m^{-2}$ $yr^{-1}$ from advection of ocean waters, and the input from land and atmosphere is slightly less than 28 g N $m^{-2}$ $yr^{-1}$. From the standpoint of the receiving water, this is a moderately high loading, comparable to that for Delaware Bay and the Potomac River estuary and twice that for Chesapeake Bay, but substantially less than the loading to the Hudson River estuary or to Boston Harbor during the 1980s (Nixon *et al.*, 1996; Howarth *et al.*, 2006).

The single largest input of nitrogen to Narragansett Bay is from rivers, estimated to be 17 g N $m^{-2}$ $yr^{-1}$ of surface area of the bay, on average (Nixon *et al.*, 1995). The second largest input of nitrogen to Narragansett Bay is the direct discharge of wastewater treatment plants (7.8 g N $m^{-2}$ $yr^{-1}$, Nixon *et al.*, 1995). Other inputs are the direct deposition of nitrogen onto the surface of the bay (1.3 g N $m^{-2}$ $yr^{-1}$) and runoff from urban areas adjacent to the bay (1.6 g N $m^{-2}$ $yr^{-1}$; Nixon *et al.*, 1995). It is important to note that compared to most estuaries, Narragansett Bay has a low ratio of watershed area to estuarine water surface area (13.2:1; Howarth *et al.*, 2006, LOICZ web site, http://data.ecology.su.se/mnode/index.htm). Thus, the loading expressed per area of estuarine area is moderately high, and the flux from the landscape per area of watershed is extremely high (2,000 kg N $km^{-2}$ $yr^{-1}$, considering wastewater, urban runoff, and river inputs). This is some 20-fold higher than one would expect from such a landscape absent human activity (Howarth *et al.*, 2002b). While such a high flux may not seem surprising given that much of the watershed is heavily urbanized, few other regions show such elevated fluxes. For example, human activity is estimated to have increased the nitrogen flux down the Mississippi River by only 5- to 6-fold (Howarth *et al.*, 2005) and into the Hudson River estuary adjacent to New York City by only 12-fold (Howarth *et al.*, 2006).

Even without the direct wastewater inputs, Narragansett Bay has a very high input of nitrogen from its watershed: ~1,400 kg N $km^{-2}$ $yr^{-1}$ (just considering river inputs and urban runoff). The sources of this nitrogen pollution in the landscape are not well known (Nixon *et al.*, 1995). How much of it might be due to atmospheric deposition onto land surfaces and subsequent export downstream to the bay? For the river inputs, we can evaluate this using the study of Boyer *et al.* (2002), which included the Blackstone River as one of 16 major rivers in the

northeastern US; the Blackstone River basin comprises 28% of the entire watershed of Narragansett Bay (Nixon *et al.*, 1995). By assuming that nitrogen exports in large rivers reflect the inputs of nitrogen to their watersheds (regardless of source; Howarth *et al.*, 1996, 2002a,b), the Boyer *et al.* (2002) analysis suggests that atmospheric deposition contributes one third of the nitrogen flux in the Blackstone River basin. While agriculture contributes some to this nitrogen flux, the majority probably comes from wastewater discharges into the Blackstone. As discussed earlier, Boyer *et al.* (2002) may have underestimated the rate of nitrogen deposition. On the other hand, the mass-balance watershed approach of Boyer *et al.* (2002) may underestimate the importance of wastewater inputs in more urbanized watersheds such as the Blackstone (Howarth *et al.*, 2006).

If atmospheric deposition contributes one third of the nitrogen flux from larger rivers into Narragansett Bay, and if most of the direct runoff from urban areas adjacent to the bay originate from atmospheric deposition, then overall atmospheric deposition (directly onto the bay and onto the landscape with subsequent export to the bay) makes up 30% of the total nitrogen inputs to the bay. Note that this is very similar to the SPARROW derived estimate, if the "non-agricultural non-point source" term is indeed associated with near-source deposition of vehicle exhaust (Table 3.1). While significant, atmospheric deposition is clearly less important as a nitrogen input to Narragansett Bay than are the inputs from wastewater treatment plants (Table 3.1).

Prudent management of nitrogen inputs to Narragansett Bay clearly should focus on the wastewater inputs. On the other hand, it may also make sense to further consider the inputs from atmospheric deposition. While there is little evidence of any increase in nitrogen loading from wastewater treatment plants to Narragansett Bay over the past several decades (see Nixon *et al.*, Chapter 5, this volume), atmospheric deposition may well have increased, particularly that in the near-vicinity of vehicles. While the population of Rhode Island grew by only 11% between 1970 and 2000, vehicle miles driven in the state increased by more than 70% (RI Statewide Planning Program, 2001). Improved technology for controlling NOx emissions from cars since the Clean Air Act Amendments of 1990 has resulted in some decrease in emissions per mile driven for cars, but overall the increase in miles driven, and an increased use of light trucks and SUVs—which are not as stringently regulated—resulted in more NOx emissions from vehicles in the eastern US during the 1990s (Butler *et al.*, 2005). Also, catalytic converters can actually increase the release of ammonia gas in car emissions due to over-reduction of NOx (Cape *et al.*, 2004).

## 3.10 Managing Atmospheric Deposition in the United States

Despite the widespread damage to coastal waters from nitrogen pollution, for the most part governments have been slow to systematically apply effective policies for controlling this problem in the United States or elsewhere (NRC,

2000; Howarth *et al.*, 2005). The reasons for this policy failure are many, but one major reason is that management of eutrophication or nutrient pollution often has focused on phosphorus rather than nitrogen since the early 1970s (Howarth and Marino, 2006; Howarth *et al*,. 2005). While this is appropriate for freshwater lakes, nitrogen is the larger problem in most coastal marine ecosystems (NRC, 2000; Howarth and Marino, 2006). Although some local or regional agencies have addressed nitrogen pollution in coastal waters over the past two decades, even today no national standards for coastal nitrogen pollution exist (NRC, 2000; Howarth *et al.*, 2005). Scientific evidence for the necessity of phosphorus control on eutrophication in freshwater lakes and nitrogen control in coastal marine ecosystems has steadily accumulated for many decades, but only in the past 5–10 years has this evidence begun to be fully accepted by water quality managers. Even when managers have recognized that nitrogen is the prime cause of eutrophication in coastal rivers and bays, management practices for non-point sources of nitrogen often have remained focused on those proven effective for managing phosphorus pollution, with insufficient recognition that other practices may be needed for nitrogen because of its much greater mobility in groundwater and through the atmosphere (NRC, 2000; Howarth *et al.*, 2005; Howarth and Marino, 2006).

Both fossil fuel combustion and agricultural practices contribute significantly to atmospheric fluxes of nitrogen but not phosphorus. The magnitude of the contribution of these atmospheric fluxes to coastal nutrient pollution remains uncertain, and understudied. Nonetheless, atmospheric deposition is clearly an important contributor to coastal nutrient pollution in many areas, including Narragansett Bay. This source demands more attention by water quality managers if the goal of reducing coastal nutrient pollution is to be met (NRC, 2000).

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