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The influence of climate on average nitrogen export from large watersheds in the Northeastern United States

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Abstract. The flux of nitrogen in large rivers in North America and Europe is well explained as a function of the net anthropogenic inputs of nitrogen to the landscape, with on average 20 to 25% of these inputs exported in rivers and 75 to 80% of the nitrogen retained or denitrified in the landscape. Here, we use data for average riverine nitrogen fluxes and anthropogenic inputs of nitrogen over a 6-year period (1988–1993) for 16 major watersheds in the northeastern United States to examine if there is also a climatic influence on nitrogen fluxes in rivers. Previous studies have shown that for any given river, nitrogen fluxes are greater in years with higher discharge, but this can be interpreted as storage of nitrogen in the landscape during dry years and flushing of this stored nitrogen during wet years. Our analyses demonstrate that there is also a longer-term steady-state influence of climate on riverine nitrogen fluxes. Those watersheds that have higher precipitation and higher discharge export a greater fraction of the net anthropogenic inputs of nitrogen. This fractional export ranges from 10 to 15% of the nitrogen inputs in drier watersheds in the northeastern United States to over 35% in the wetter watersheds. We believe this is driven by lower rates of denitrification in the wetter watersheds, perhaps because shorter water residence times do not allow for as much denitrification in riparian wetlands and low-order streams. Using mean projections for the consequences of future climate change on precipitation and discharge, we estimate that nitrogen fluxes in the Susquehanna River to Chesapeake Bay may increase by 3 to 17% by 2030 and by 16 to 65% by 2095 due to greater fractional delivery of net anthropogenic nitrogen inputs as precipitation and discharge increase. Although these projections are highly uncertain, they suggest a need to better consider the influence of climate on riverine nitrogen fluxes as part of management efforts to control coastal nitrogen pollution.

Introduction

Over the past several decades, eutrophication in coastal marine ecosystems has grown and is now considered the biggest pollution problem in the coastal waters of the U.S. (Howarth et al. 2000; NRC 2000). Eutrophication lowers biotic diversity, leads to hypoxic and anoxic conditions, increases the incidence and duration of some types of harmful algal blooms, degrades the habitat quality of seagrass beds or even destroys them, and can lead to changes in ecological food webs that reduce fish and shellfish production (NRC 2000). The Environmental Protection Agency's National Coastal Condition Report (EPA 2001) lists eutrophic condition as one of the three greatest threats to the health of the nation's estuaries, along with poor benthic condition (a result, in part, of eutrophication) and wetland loss. Some 40% of the estuarine area in the conterminous U.S. is severely degraded from eutrophication, and 67% is degraded to some extent (Bricker et al. 1999; EPA 2001). In the northeastern United States (defined as Chesapeake Bay north through Maine), some 60% of the estuarine area shows a high expression of eutrophic condition (EPA 2001). Eutrophication of coastal marine ecosystems is driven primarily by nitrogen inputs (Howarth 1988; Nixon 1995; NRC 2000; Howarth and Marino, 2006). From 1960 to 1980, average nitrogen fluxes in rivers to the coastal waters of the United States are estimated to have increased by 67% (Howarth et al. 2002a). During the 1980's, nitrogen fluxes increased little if at all. However, riverine nitrogen fluxes in the United States are estimated to have again increased steadily over the past 15 years, although less rapidly than during the 1960s and 1970s (Howarth et al. 2002a).

Climate variability and climate change are likely to have a profound effect on the delivery of nutrients to coastal marine ecosystems, but there is great uncertainty as to the detailed responses expected (Scavia 2002). This uncertainty results in part from divergent predictions for future climate change, for example with some global models predicting a drier climate and some a wetter climate in the northeastern United States as atmospheric carbon dioxide levels continue to rise over the next century (Wolock and McCabe 1999). Further uncertainty results from the non-linearity in response of riverine freshwater discharge to changes in climate, with some models suggesting discharge will increase disproportionately to increases in precipitation, and others suggesting increases in discharge will be less than increases in precipitation (Najjar 1999; Wolock and McCabe 1999; Najjar et al. 2000). Beyond these uncertainties in the physical climate system and the hydrologic responses of watersheds, the biogeochemical responses to changes in climate and hydrology are difficult to predict, particularly for nitrogen. However, sustained changes in nitrogen processing within the landscape are likely to have very significant effects on the health of coastal marine ecosystems.

Watersheds with greater precipitation and discharge will tend to have higher erosion rates, and this leads to higher fluxes of phosphorus from the landscape since most of the phosphorus in large rivers is particle bound (Howarth et al. 1995, 2002b; Moore et al. 1997). Nitrogen moves through the landscape primarily in dissolved forms, and nitrogen fluxes seem to be primarily controlled by the sources and sinks of nitrogen in the landscape. For disturbed landscapes in the temperate zone, an average of 20 to 25% of the nitrogen inputs resulting from human activity is exported in rivers (Howarth et al. 1996, 2002b; Boyer et al. 2002). Is there a climatic influence on this relationship? For examining global patterns of nitrate flux in large rivers, some models have assumed that the non-point-source contribution is controlled in part by area-specific discharge (Seitzinger and Kroeze 1998; Caraco and Cole 1999), but in a direct comparison among these and other models, those without discharge or other climatic parameters proved to be at least as accurate and precise in predicting multi-year average fluxes (Alexander et al. 2002). For the Mississippi River basin, McIsaac et al. (2001) demonstrated that during dry years, nitrogen accumulates in the soil or groundwater, and during wet years, this stored nitrogen is flushed out. The time scale of response in their study was only a few years. What would be the consequences of a sustained change in climate over a longer period of time? Over longer time scales, the primary issues are not short-term storage and flushing, but rather whether there are changes in nitrogen sinks in the landscape (storage in soils and biomass, or in rates of denitrification). In this paper, we further address the influence of climate on average riverine nitrogen flux by examining the relationship of net anthropogenic nitrogen inputs (NANI) on 6-year mean nitrogen fluxes in 16 major rivers across a climate gradient in the northeastern United States. By studying this climatic gradient, we can ascertain the longer-term steady-state effects of climate on riverine nitrogen fluxes.

Methods

We build upon the analysis of anthropogenic nitrogen sources and riverine nitrogen fluxes for 16 major watersheds in the northeastern U.S. done by Boyer et al. (2002) for the time period of 1988 through 1993. These watersheds are (moving north to south, from Maine to Virginia) the Penobscot, Kennebec, Androscoggin, Saco, Merrimack, Charles, Blackstone, Connecticut, Mohawk, upper Hudson, Delaware, Schuylkill, Susquehanna, Potomac, Rappahannock, and James. This is the same set of large watersheds used by Alexander et al. (2002) in their comparison of models for predicting nitrate and total nitrogen fluxes. It is important to note that both our work and that of Boyer et al. (2002) are based on the watershed areas upriver of defined USGS monitoring stations, and so do not generally include the heavily urbanized areas immediately along the coast. So defined, the watershed areas vary from 475 km^2 for the Charles River basin to over 70,000 km^2 for the Susquehanna (Table 1). The single largest land-use type in all 16 watersheds is forest, ranging from 48% of the area of the Schuylkill to 87% of the area of the Saco. Agricultural land use varies from 1.5% of the land area in the Penobscot River basin to 38% in the Schuylkill, and urban land use varies from 0.4% of the area in the Penobscot to 22% in the Charles River basin. Population densities vary from 8 individuals per km^2 in the Penobscot basin to 556 individuals per km^2 in the Charles River basin (Table 1). Further information on the watersheds is given in Boyer et al. (2002).

We estimated annual average river discharges using daily discharge data from river gauging stations located at the outlet of each watershed (USGS 2005). Annual discharge for all 16 rivers for the period 1950 through 2003 is

Table 1. Characteristics of the 16 major watersheds of the northeastern U.S. during the period 1988 to 1993. Watersheds are defined as the area upstream of USGS gauging stations, as in Boyer et al. (2002).

	Area (km^2)	Population density (# km^{-2})	Mean discharge (mm $year^{-1}$)	Mean precipitation (mm $year^{-1}$)	Mean temp. (°C)	Mean N export (kg N km^{-2} $year^{-1}$)
Penobscot	20,109	8	588	1075	4.3	320
Kennebec	13,994	9	566	1085	4.3	330
Androscoggin	8,451	17	640	1151	4.6	400
Saco	3,349	16	672	1218	5.8	390
Merrimack	12,005	143	589	1148	7.4	500
Charles	475	556	583	1207	9.7	1760*
Blackstone	1,115	276	651	1260	9.0	1140
Connecticut	25,019	65	642	1160	6.3	540
Hudson	11,942	32	622	1126	6.6	500
Mohawk	8,935	54	548	1142	6.8	800
Delaware	17,560	85	547	1131	8.7	960
Schuylkill	4,903	293	488	1134	10.6	1760
Susquehanna	70,189	54	487	1022	8.9	980
Potomac	29,940	63	328	985	11.3	900
Rappahannock	4,134	24	360	1045	12.6	470
James	16,206	24	407	934	10.1	310

*Includes nitrogen in wastewater flows diverted out of the watershed.

shown in Figure 1. The period of analysis for our study (1988–1993) is indicated by grey shading in the figure. Note that discharges during our period of analysis and the preceding several years are broadly representative of the longer time frame, without unusually high or low discharge years. Annual average riverine nitrogen exports were estimated for the 1988–1993 period from USGS data on total nitrogen concentrations (collected at the gauging stations at approximately monthly intervals) using the estimator approach described in Cohn et al. (1992). This regression-based method is a flow-weighted interpolation of the concentration measurements (Boyer et al. 2002). Mean estimates for precipitation and temperature for the 6-year period were obtained from the VEMAP-II historical climate reconstruction (Kittel et al. 1997; Boyer et al. 2002).

We determined the net anthropogenic nitrogen inputs (NANI) to each watershed using the approach of Howarth et al. (1996). In this method, NANI is the sum of fertilizer use, nitrogen fixation in agro-ecosystems, the net import of nitrogen in human food and animal feeds, and the atmospheric deposition of oxidized nitrogen (NO_y). Note that wastewater discharges are not considered explicitly in this analysis, since the nitrogen in wastewater originates from food (either imported or grown within the region, with the source nitrogen from fertilizer use or agricultural nitrogen fixation). Similarly, the deposition of ammonia and ammonium is not considered an input in this approach, as the large majority of the ammonia and ammonium deposited in a watershed is assumed to have originated from emissions within the same watershed

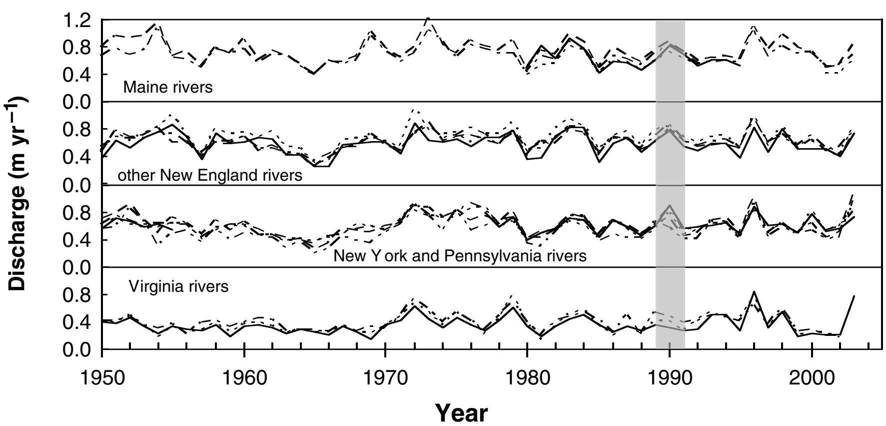

Figure 1. Annual freshwater discharge for each of the 16 northeastern US watersheds from 1950 to 2003. Top panel shows the 4 rivers that are mostly in Maine (Penobscot, Kennebec, Androscoggin, and Saco), the second panel from top shows the 4 other rivers in New England that are further to the west or south (Merrimack, Connecticut, Charles, and Blackstone), the third panel from the top shows the 5 rivers that are largely in New York State and Pennsylvania (Hudson, Mohawk, Delaware, Susquehanna, and Schuylkill), and the bottom panel shows the 3 rivers that are largely in Virginia and Maryland (Potomac, Rappahannock, and James). Our period of analysis for this study (1988–1993) is indicated by grey shade.

(Howarth et al. 1996). This is generally true in large watersheds and regions, but in smaller watersheds, there may be significant fluxes of ammonia and ammonium through atmospheric transport across different watersheds. Boyer et al. (2002) attempted to estimate these cross-boundary fluxes of ammonia and ammonium for the 16 major northeastern watersheds. However, this requires many highly uncertain assumptions, and in any case, the net ammonia/ammonium deposition due to cross-watershed transport in the atmosphere is small relative to NO_y deposition (Boyer et al. 2002). Therefore, we simply consider the NO_y term here. Note that in the Howarth et al. (1996) study, we used only the part of NO_y deposition estimated to originate from human activity in calculating NANI, rather than the total NO_y deposition we use here. On average for watersheds in the northeastern U.S., the total NO_y deposition is 2.3% greater than the anthropogenically derived NO_y deposition (Howarth et al. 1996).

We estimate the atmospheric deposition of NO_y (both wet and dry deposition) using the approach of Ollinger et al. (1993), based on a spatial model that extrapolates data from depositional monitoring networks (such as NADP) with a consideration of topographic effects. For most watersheds, we used the Ollinger et al. (1993) model, updated with more recent depositional velocities for dry deposition (Lovett and Rueth 1999). A few of the watersheds (Potomac, Rappahannock, and James) are outside of the geographic range for the regression equations used in Ollinger et al. (1993); for those three watersheds, we used the regression relationships put forth by Lovett and Lindberg (1993) that relate dry deposition to wet deposition (Boyer et al. 2002).

We estimate fertilizer use in each watershed using county-based sales data (Battaglin and Goolsby 1994) from 1991, scaled to the watersheds by weighting by the percentage of county area in each watershed (Boyer et al. 2002). Nitrogen fixation associated with agricultural crops is estimated from the area of particular types of crops (soybeans, alfalfa, snap beans, and hay and pasture) multiplied by literature-derived estimates of fixation rates associated with those individual crop types (Boyer et al. 2002). The net import of nitrogen in human food and animal feeds is estimated from a mass balance of needs versus production; that is, the difference between per capita estimates of the nitrogen in food and feed needs for humans and domestic animals and the nitrogen in foods and feeds produced within a watershed (Boyer et al. 2002). These estimates are somewhat sensitive to the assumed efficiency of nitrogen use in animal production; we use the values of van Horn (1998), which are based on U.S. agricultural practices.

Results

The average riverine nitrogen export from the 16 watersheds over the 6 year period from 1988 through 1993 ranged from a low of 310 to 330 kg N km^{-2} $year^{-1}$ for the James, Penobscot, and Kennebec River basins to a high of $\sim$1760 kg N km^{-2} year^{-1} for the Charles and Schuylkill River basins (Table 1). In comparison, without human disturbance average watersheds in the north temperate zone are estimated to export approximately 100 kg N km^{-2} year^{-1} (Howarth et al. 1996, 2002b; NRC 2000). The fluxes from the Charles and Schuylkill basins are quite high, and in fact exceed the average flux from the watersheds of the highly populated, heavily industrialized and agriculturally intensive watersheds that drain to the North Sea in Europe (1450 kg N km^{-2} year^{-1}; Howarth et al. 1996). Seven out of the 16 watersheds in the northeastern U.S. have nitrogen fluxes that exceed the average flow down the Mississippi River basin (570 kg N km^{-2} year^{-1}; Howarth et al. 1996).

The 16 watersheds vary in the relative importance of the various nitrogen inputs to the overall NANI estimate (Table 2). The majority of NANI comes from NO_y deposition in the 4 watersheds in Maine (the Penobscot, Kennebec, Androscoggin, and Saco River basins). In the watersheds further south, the NO_y deposition rates are higher than in Maine, but other sources increase even more (Table 2). The net importation of nitrogen in food and feed is quite important in watersheds with higher population densities, and this makes up more than half of NANI in the Charles and Blackstone River basins. In many watersheds, agricultural inputs from fertilizer use and nitrogen fixation are dominant, and these make up 50% or more of NANI in the Mohawk, Delaware, Potomac, Rappahannock, and James River basins (Table 2). Overall, for

Table 2. Average annual nitrogen inputs from anthropogenic sources to the 16 major watersheds of the northeastern U.S. for the period 1988 to 1993 (kg N km^{-2} year^{-1}).

	NO_y deposition	N fertilizer use	Agricultural N fixation	Net N import in foods and feeds	Total Net anthropogenic N inputs (NANI)
Penobscot	360	90	70	40	560
Kennebec	430	50	160	150	790
Androscoggin	500	80	150	240	970
Saco	570	40	100	100	810
Merrimack	610	150	210	710	1680
Charles	670	200	190	2090	3150
Blackstone	710	310	310	1500	2830
Connecticut	630	270	360	570	1830
Hudson	660	200	370	270	1500
Mohawk	710	410	1240	620	2980
Delaware	810	530	680	350	2370
Schuylkill	890	1210	1230	1950	5280
Susquehanna	820	620	1150	1100	3690
Potomac	710	1020	1170	1450	4350
Rappahannock	620	1030	1440	610	3700
James	650	360	700	400	2110
Area-weighted mean	680	560	740	740	2720
northeastern US mean (Howarth et al. 1996)	1200	600	750	1000	3550

these 16 watersheds, the area-weighted mean nitrogen inputs to the watersheds are reasonably evenly distributed between NO_y deposition (680 kg N km^{-2} year^{-1}), fertilizer use (560 kg N km^{-2} year^{-1}), nitrogen fixation in agro-ecosystems (740 kg N km^{-2} year^{-1}), and the net importation of nitrogen in foods and feeds (740 kg N km^{-2} year^{-1}; Table 2).

The analysis presented here is similar to that presented in Howarth et al. (1996) for the northeastern U.S. as a whole in terms of average riverine nitrogen fluxes, and the mean value for the entire northeastern U.S. presented in Howarth et al. (1996) sits in the center of, and is bracketed nicely by, the riverine nitrogen flux values for the 16 major watersheds. The analysis here also is similar in terms of the agricultural sources to the regions (fertilizer use and agricultural nitrogen fixation), but the estimates given in Howarth et al. (1996) are substantially greater for NO_y deposition and for the net importation of nitrogen in food and feeds (Table 2). For the importation of nitrogen in food and feeds, we attribute this difference to the inclusion of the heavily populated coastal margin cities (New York City, Boston, Washington, Providence, Philadelphia, etc.) within the area included in the Howarth et al. (1996) analysis but excluded from Boyer et al. (2002) and this study. For the most part, these urban centers in the northeastern U.S. are down-river from the USGS gauging stations which define the watershed areas used by Boyer et al. (2002) and this study.

For NO_y deposition, the mean value for the 16 watersheds reported in Boyer et al. (2002) and used here is 680 kg N km^{-2} year^{-1}, while for the entire northeastern U.S. Howarth et al. (1996) used an estimate of 1200 kg N km^{-2} year^{-1} (Table 2). As was the case with the net importation of nitrogen in food and feeds, the difference in these estimates may reflect the different geographic boundaries, with higher deposition in the more urbanized areas. Deposition in the more rural areas represented by the 16 watersheds (as defined up-river of the USGS gauging stations) may not reflect the potentially high levels of deposition that occur near emission sources in urban areas (Holland et al. 1999; Lovett et al. 2000; Howarth et al. 2002a). However, the different estimates also may be due in part to different methodologies. As stated above, Boyer et al. (2002) used depositional monitoring data for their estimate. The core data are from the National Acid Deposition Program (NADP), whose stations are purposefully located in rural areas where urban and agricultural influences are minimal (NADP 2005). Spatial coverage is sparse, and scaling the point observations over space and time is difficult (Meyers et al. 2001). Further, only wet deposition is measured at the NADP stations, and challenges remain in how to estimate contributions from dry deposition, given the complexity of factors controlling deposition velocities (Ollinger et al. 1993, Meyers et al. 2001). The estimate used in Howarth et al. (1996) comes from the GCTM model, which 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). A similar, more recent model (TM3) used by Galloway et al. (2004) for their global and regional nitrogen budgets yields a comparable estimate for the northeastern U.S. as did the GCTM model. These emission-based models are attractive, in that at least at very course spatial scales, they are as accurate as the emission data. However, they cannot easily be applied at a spatial scale fine enough to give estimates for the individual 16 northeastern watersheds. For the analysis in this paper, we therefore relied on the estimates from Boyer et al. (2002), which may well be robust for the rural areas represented by these watersheds. However, it is important to note that the actual total NO_y deposition to the northeastern U.S. may be substantially higher. If so, much of this additional NO_y deposition likely falls on the more urbanized landscape near the coast, where retention is low, and so it is likely to have a high percentage export to coastal waters (Howarth et al. 2002b).

The average annual riverine nitrogen fluxes from the 16 watersheds are highly correlated with NANI to each watershed (Figure 2). The relationship is very similar to that observed when comparing the large regional areas that drain into the North Atlantic Ocean, both from North America and from Europe (Howarth et al. 1996). Note in both cases the *y*-intercept of the linear regression is approximately 100 kg N km^{-2} $year^{-1}$, which has been used to provide an estimate of what the nitrogen flux off the landscape for temperate watersheds might be, absent human inputs of nitrogen (that is, NANI = 0; NRC 2000; Howarth et al. 2002b). Here, the slope of the regression is 0.26, indicating that on average only 26% of the human inputs of nitrogen to the landscape (NANI) are exported in downstream river export, and that 74% must be retained in the landscape or lost through denitrification. This is a similar slope to that observed in the coarser spatial-scale analysis of the North

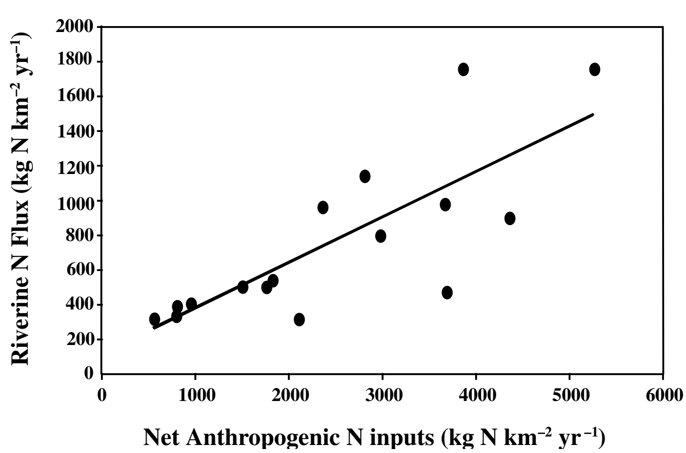

Figure 2. Average annual riverine nitrogen fluxes are strongly related to the net anthropogenic nitrogen inputs (NANI) to the watershed ($Y = 0.26X + 107$; $R^2 = 0.62$; $p = 0.0003$). Note that this relationship is similar to that reported in Boyer et al. (2002), although the latter was based on total nitrogen inputs, and not just the anthropogenic sources.

Atlantic drainage basin (Howarth et al. 1996). Note also that Boyer et al. (2002) presented a somewhat different analysis (in their figure 6). There, the x-axis is for total nitrogen inputs, and so includes an estimate for the natural rate of nitrogen fixation in forests. The Boyer et al. (2002) figure also includes some estimated net input of nitrogen from deposition of ammonia and ammonium which is not included in the NANI estimate here (see methods, above). The relationship shown in Boyer et al. (2002) looks very similar to that here, except that the intercept (7 kg N km^{-2} $year^{-1}$) was much closer to 0. This is consistent with the idea that a watershed that consistently receives no nitrogen inputs (from natural or anthropogenic sources) would export little or no nitrogen, and thus gives us greater confidence in using the intercept from Figure 2 (107 kg N km^{-2} $year^{-1}$) as an estimate of the riverine nitrogen flux for temperate watersheds which have only natural inputs of nitrogen.

The average riverine nitrogen flux from the 16 watersheds is fairly well explained just from NANI ($R^2 = 0.62$, $p = 0.0003$; Figure 2), and one could easily believe that much of the scatter results from quality of data or from differences among the watersheds in characteristics such as soil type and topography. However, we note that the points lying above the regression line tend to be watersheds with higher discharge and precipitation, while those below it are from ''less wet'' watersheds (Table 1). To evaluate whether some aspect of climate has an influence on the long-term average flux of nitrogen from these watersheds (in addition to the influence of NANI), we examined the fractional delivery of NANI and examined its relationship to climatically related parameters (precipitation, temperature, and discharge). We define the fractional delivery as the riverine nitrogen flux that is above the natural background flux expected absent any anthropogenic nitrogen inputs, divided by NANI for that watershed. That is,

$$F = (R - 107)/\mathrm{NANI} \tag{1}$$

where F is the fractional delivery of NANI, R is the long-term average riverine flux of nitrogen (kg N km^{-2} $year^{-1}$), 107 represents the natural background riverine nitrogen flux in the absence of human activity (kg N km^{-2} $year^{-1}$), and NANI is the net anthropogenic nitrogen input (kg N km^{-2} $year^{-1}$).

The fractional delivery of NANI is well correlated with both precipitation ($R^2 = 0.53$; $p = 0.0015$; Figure 3a) and discharge ($R^2 = 0.48$; $p = 0.003$; Figure 3b). Note that precipitation (P) and discharge (Q) are themselves correlated ($R^2 = 0.66$; plot not shown). Clearly, watersheds with greater precipitation and higher discharge have higher fractional deliveries, ranging from a high of 0.2 to 0.43 for watersheds with precipitation greater than 1,100 mm $year^{-1}$ and discharges greater than 500 mm $year^{-1}$ to 0.1 to 0.18 for watersheds with less precipitation and lower discharge. Temperature is not as good a predictor of fractional delivery of NANI, and the relationship, while suggestive, is at best marginally significant ($p = 0.11$) and has a lower R^2 value (0.17; Figure 3c). Note however the suggestion of an inverse relationship, with

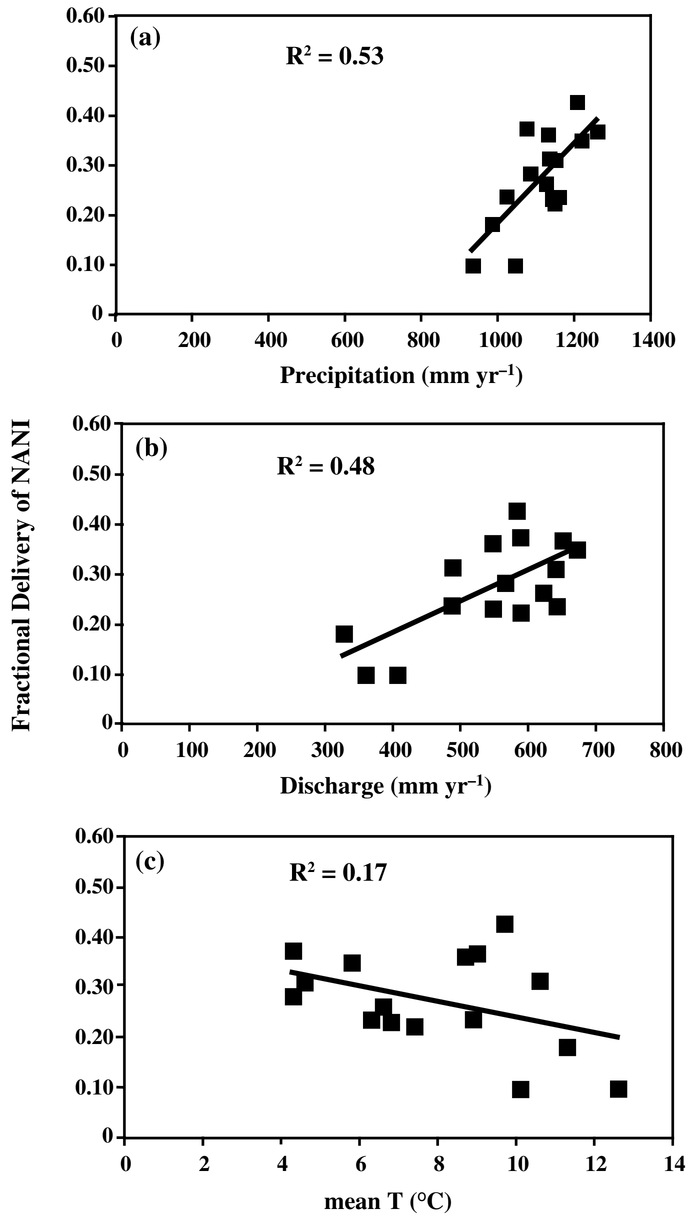

Figure 3. The fractional delivery of net anthropogenic nitrogen inputs (NANI) for the 16 watersheds plotted as a function of mean precipitation (a, top panel; $R^2 = 0.53$, $p = 0.0015$), mean discharge (b, middle panel; $R^2 = 0.48$, $p = 0.003$), and mean temperature (c, bottom panel; $R^2 = 0.17$, $p = 0.11$).

greater fractional delivery of NANI in the colder watersheds. While temperature is correlated with discharge ($R^2 = 0.56$; plot not shown), temperature is not well correlated with precipitation in these watersheds ($R^2 = 0.11$; $p = 0.20$; plot not shown).

To develop a predictive equation for the riverine nitrogen flux (R), we can re-arrange Equation (1) and substitute single variable relationships for F discussed above in an equation of the form:

$$R = F * \mathrm{NANI} + 107 \tag{2}$$

For example, we could use precipitation as a predictor of F (Figure 3a) since that relationship had the best explanatory power. Substituting the regression parameters for Figure 3a into Equation (2) yields:

$$R = (0.0008 * P - 0.62) * \mathrm{NANI} + 107 \tag{3}$$

where P is precipitation (mm year^{-1}). This equation can be written in the following form:

$$R = (0.0008 * P * \mathrm{NANI}) - (0.62 * \mathrm{NANI}) + 107 \tag{4}$$

Alternatively, to get the best parameter fit for an equation of the form of Equation (4), we can obtain coefficients that relate R to (P * NANI) and NANI by using a 2-variable linear regression with interacting terms. This yields the equation:

$$R = (0.00095 * P * \mathrm{NANI}) - (0.762 * \mathrm{NANI}) + 55 \tag{5}$$

The relationship is highly significant ($p < 0.000001$) and has an R^2 value of 0.875, or an adjusted R^2 of 0.855 (Table 3). Both the interaction term (P * NANI) and the NANI term contribute significantly to this relationship ($p = 0.0002$ and $p = 0.0024$, respectively; Table 3). Re-arranging Equation (5) into the form of Equation (3) yields:

$$R = (0.00095 * P - 0.762) * \mathrm{NANI} + 55 \tag{6}$$

The intercept of 55 kg N km^{-2} year^{-1} is lower than the 107 kg N km^{-2} year^{-1} determined from the NANI vs. riverine nitrogen flux regression (Figure 2), but not significantly so. The 95% confidence interval for the intercept determined in Equation (6) extends from −155 to +255 kg N km^{-2} year^{-1}. Note that the term (0.00095 * P−0.762) expresses the fractional delivery of NANI, or F.

We also tested a more complex model, including not only (P * NANI) and NANI as input terms but also P (that is, the complete interaction model for NANI and P). This 3-variable, interacting-term regression model is also significant (Table 4), but less so than the simpler model using just (P*NANI) and NANI. The addition of P alone does not contribute significantly ($p = 0.613$; Table 3), and its inclusion in the regression lessens the significance of the other two terms in comparison to the simpler 2-term model ($p = 0.036$ for (P*NANI) compared to $p = 0.0002$, and $p = 0.091$ for NANI, compared to $p = 0.0024$;

Table 3. Summary statistics for several linear regression models that predict riverine nitrogen flux (R) based on precipitation (P) and net anthropogenic nitrogen inputs (NANI).

	Interacting-term model with NANI and P *NANI	Interacting-term model with P, NANI, and P *NANI	non-interacting term model with P and NANI
Regression statistics			
R^2	0.875	0.877	0.821
Adjusted R^2	0.855	0.847	0.793
Standard error	179.5	184.8	749.3
Observations	16	16	16
ANOVA			
df for regression	2	2	2
df for residual	13	13	13
F	45.36	28.63	29.75
P	< 0.00001	0.00001	0.00001
Intercept			
Coefficient	55	920	−2710
Standard error	93	1670	749
t-statistic	0.59	0.55	−3.61
p-value	0.56	0.59	0.003
NANI			
Coefficient	−0.762	−1.03	0.287
Standard error	92.7	0.561	0.040
t-statistic	−3.76	−1.84	7.24
p-value	0.0024	0.091	0.00001
P			
Coefficient	–	−0.77	2.47
Standard error	–	1.49	0.65
t-statistic	–	−0.520	3.80
p-value	–	0.61	0.002
P *NANI			
Coefficient	0.00095	0.00095	–
Standard error	0.00018	0.00018	–
t-statistic	5.12	5.12	–
p-value	0.0002	0.036	

Table 3). This adds to our confidence in the approach we used to derive a predictive equation for riverine nitrogen flux by combining the relationship between NANI and riverine N flux (Equation (2); Figure 2) with the relationship which best predicts the fractional delivery of NANI as a function of precipitation (Figure 3a).

A regression that relates riverine discharge to P and NANI without an interaction of P and NANI yields the following equation:

$$R = (2.47 * P) + (0.29 * \mathrm{NANI}) - 2710 \qquad (7)$$

This relationship, too, is highly significant ($p = 0.00001$) with both the P and NANI terms contributing significantly to the regression ($p = 0.002$ and $p = 0.0001$, respectively; Table 3). However, the adjusted R^2 value (0.79) and F-

Table 4. Summary statistics for two interacting-term linear regression models that predict riverine nitrogen flux (R), based on discharge (Q) and net anthropogenic nitrogen inputs (NANI) or based on population density (D) and precipitation (P).

	Interacting-term model with NANI and Q*NANI	Interacting-term model with D and D*P
Regression statistics		
R^2	0.874	0.828
Adjusted R^2	0.855	0.801
Standard error	179.7	210.4
Observations	16	16
ANOVA		
df for regression	2	2
df for residual	13	13
F	45.28	31.25
P	< 0.000001	0.00001
Intercept		
Coefficient	−101	374
Standard error	101	73.5
t-statistic	−1.00	5.09
p-value	0.33	0.0002
NANI		
Coefficient	−0.096	–
Standard error	0.077	–
t-statistic	−1.24	–
p-value	0.24	–
D		
Co efficient	–	25.0
Standard error	–	8.6
t-statistic	–	2.90
p-value	–	0.012
Interacting term		
Coefficient	0.00087	−0.18
Standard error	0.00017	0.0071
t-statistic	5.11	−1.58
p-value	0.0002	0.023

ratio (28.9) are slightly lower than for equations 5 and 6, where P and NANI interact (compare Tables 3 and 5). As discussed below, predictors that do not include interaction terms, such as that in Equation (7), lead to dramatically different mechanistic interpretations than do the predictive equations which include such interactions, such as equations 5 and 6.

The slope of the regression line when riverine nitrogen fluxes predicted using NANI and precipitation (Equation 6) are plotted against the actual observed nitrogen fluxes is very close to 1:1, with a very good linear fit (Figure 4a). However, an exponential fit of the regression looks reasonable as well (Figure 4b), and statistically, the two fits are indistinguishable. The exponential fit indicates the possibility of a bias in Equation (6), with it under-predicting riverine nitrogen fluxes at both the low and high end of the relationship. At the low end, this would be consistent with a ''pristine'' riverine nitrogen flux

(NANI = 0) that is greater than the 55 kg N km^{-2} $year^{-1}$ predicted from Equation (6). The exponential fit instead suggests a "pristine" riverine nitrogen flux of 255 kg N km^{-2} $year^{-1}$ (Figure 4b). At the high end of the relationship, the exponential fit would be consistent with the concept of nitrogen saturation (Aber et al. 1998, 2003). That is, the percentage of nitrogen exported from the landscape may increase disproportionately with nitrogen loading to the landscape above a certain point. Aber et al. (2003) have shown that nitrogen losses from forests in the northeastern US increase dramatically and non-linearly as atmospheric deposition exceeds ~700 kg N km^{-2} $year^{-1}$, as occurs in several of the watersheds included in our data analysis.

Discharge is not quite as good a predictor of the fractional delivery of NANI as is precipitation, but it is still significant (Figure 3a and b). We can use

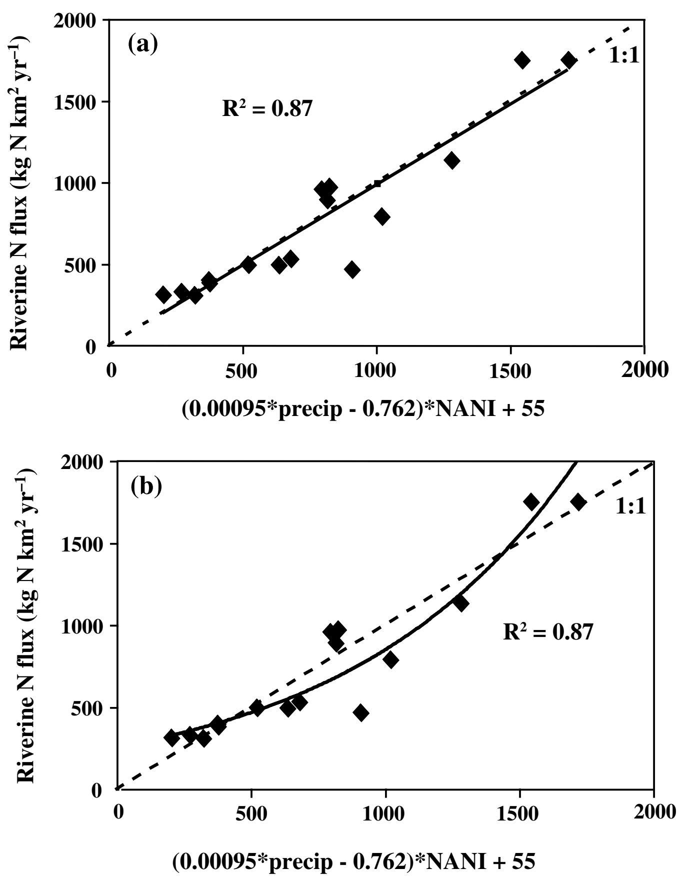

Figure 4. Equation (6) in the text ($R = (0.00095 * P - 0.762) * \mathrm{NANI} + 55$) is an excellent predictor of riverine nitrogen fluxes. Note the similarity of the regression line for the prediction and the 1:1 line (a, top). There is some suggestion of an exponential fit (b, bottom; $Y = 255\ e^{0.0012X}$), which may indicate nitrogen saturation in the landscape at higher inputs of anthropogenic nitrogen.

discharge instead of precipitation to develop a predictive equation for riverine nitrogen flux following the steps outlined above, using a 2-variable, interacting-term regression with Q and (Q*NANI). This also gives a highly significant relationship (Table 4) and yields the equation:

$$R = (0.00087 * Q - 0.096) * \mathrm{NANI} - 101 \tag{8}$$

The regression statistics are very similar to those for the 2-variable model using precipitation (compare Table 4 with Table 3), with one exception: the NANI term alone does not contribute significantly to this regression ($p = 0.24$; Table 4), and the regression is driven largely by the interactive (Q*NANI) term. Note also that the intercept is negative (−101 kg N km^{-2} year^{-1}). This intercept corresponds to the predicted riverine nitrogen flux in the situation where there was no human disturbance (NANI = 0), and a negative nitrogen flux from rivers is of course nonsensical. The 95% confidence limits for the intercept, however, extend from −319 to +116 kg N km^{-2} year^{-1}. Plots of the riverine nitrogen fluxes predicted from Equation (8) (using the NANI plus the interaction term Q*NANI) are shown in Figure 5a and b. These are very similar to those plots showing predictions based on (P*NANI) and NANI (Figure 4a and b).

It is also of interest to know whether simpler ''proxy variables'' for NANI have as much explanatory power. Population density is an example of such a variable, and has been used in many studies as an explanatory variable for nutrient discharge (Peierls et al. 1991; Smith et al. 2003). We tested a relationship of the same form Equation as 4, but substituting population density (D) for NANI, and obtained:

$$R = (-0.018 * P * D) + (25.0 * D) + 374 \tag{9}$$

With an adjusted R^2 of 0.80, it is a good relationship that is highly significant (Table 4), but not as predictive as using NANI (Table 3). Both the D term and the interaction term are significant ($p = 0.012$ and $p = 0.023$, respectively), but it is interesting to note that the signs are opposite those of the corresponding terms in Equation (5), suggesting that population density is not behaving as a simple proxy for NANI. As discussed above, equations 5 and 6 suggest that the slope of the relationship between R and NANI is itself a positive linear function of P; Equation (9) indicates that the corresponding slope of the relationship between R and D is a negative linear function of P.

Discussion

Given that many of the statistical models we explore do a good to excellent job of predicting riverine nitrogen flux, one must interpret them with care. Nonetheless, our analysis indicates a greater fractional export of NANI from the watersheds with greater precipitation (Figure 3a) and discharge (Figure 3b). Note that this greater fractional export of NANI is not due to

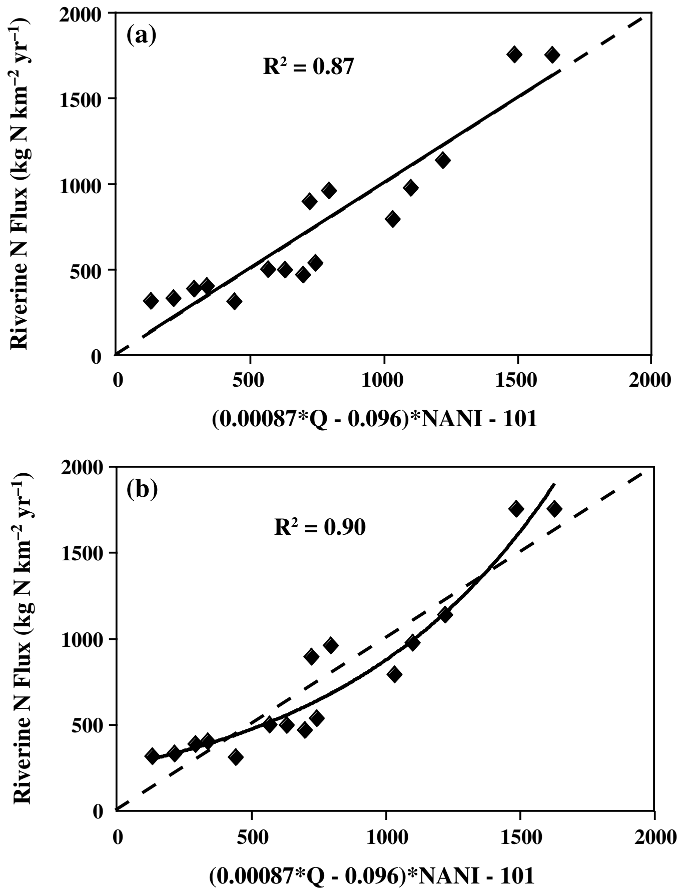

Figure 5. Equation (8) in the text ($R = (0.00087 * Q - 0.0.096) * \text{NANI} - 101$) is also an excellent predictor of riverine nitrogen fluxes. Note the similarity to Figure 4, including the close fit to the 1:1 line (a, top) and the suggestion of an exponential fit (b, bottom; $Y = 251\ e^{0.0021X}$).

flushing during wet years of nitrogen stored in the landscape during preceding dry years, as observed by McIsaac et al. (2001). Our analysis is based on mean fluxes of nitrogen over a 6-year period in watersheds across a climatic gradient, and the discharge from these watersheds during the period of analysis (1988–1993) is typical of the longer time scale observed over the last half century (Figure 1). We therefore believe that our results reflect the long-term influence of climate on the fate of NANI. The most plausible interpretation mechanistically is that the sinks for nitrogen in the landscape are smaller in watersheds with greater precipitation and discharge.

In general, the sinks for reactive nitrogen in the environment are poorly known (Galloway et al. 2004), but they are as well estimated for these 16 northeastern U.S. watersheds as for any other region on Earth. The best estimates are that for the NANI not exported in rivers from these 16 watersheds, roughly one third accumulates in soils or biomass or is exported from the watersheds in wood, while approximately two thirds is denitrified (van Breemen et al. 2002). It may seem paradoxical that a wetter climate would lead to either less storage of nitrogen in soils and biomass or less denitrification, as one might actually predict greater accumulation of organic matter in the soils of wetter environments, and greater rates of denitrification in wetter environments where soils are perhaps more likely to be waterlogged. We suggest that the major influence of climate on the nitrogen sinks is for less denitrification in the watersheds with greater precipitation and discharge, due to faster flushing of water through riparian wetlands and low-order streams. These riparian wetlands and low-order streams are likely to be sites of significant denitrification, and the amount of nitrogen that can be removed from these systems is directly related to the water residence time (Howarth et al. 1996; van Breemen et al. 2002; Seitzinger et al. 2002).

Lewis et al. (1999) have demonstrated that the nitrogen fluxes from undisturbed catchments in the tropics are greater where discharge is higher, a result which could be explained by higher rates of biological nitrogen fixation in the wetter environments, lower sinks for nitrogen in the wetter environments, or both. Lewis (2002) found a very similar relationship for small catchments in the United States where rates of deposition were relatively low (mean of 280 kg N km^{-2} $year^{-1}$, but note that deposition was greater than 400 to 500 kg N km^{-2} $year^{-1}$ in many of the catchments). For these catchments, it is unlikely that higher rates of nitrogen fixation can explain the pattern: assuming that only 20 to 25% of the nitrogen inputs (atmospheric deposition plus natural biological nitrogen fixation) are exported in stream flow (as for average NANI and total nitrogen inputs in larger temperate-zone watersheds; Howarth et al. 1996; Boyer et al. 2002), rates of biological nitrogen fixation would have to exceed 2,000 kg N km^{-2} $year^{-1}$ in the wetter catchments to support the observed nitrogen exports in streams. Such rates have not been observed in temperate-zone terrestrial ecosystems (Cleveland et al. 1999) and are an order of magnitude higher than estimates for the forests of the 16 major northeastern watersheds (Boyer et al. 2002). We suggest that the most likely explanation for the observation of Lewis (2002) is that the nitrogen sinks are smaller in the wetter environments, and that a higher fraction of NANI (and perhaps natural nitrogen fixation) is exported, as suggested in our analysis.

The statistical models we present in this paper fall into two general types: those that have an interactive term between NANI and precipitation or discharge (equations 5, 6, and 8) and those where there is no multiplicative interaction between NANI and the climate variable (Equation 7). These have very different physical interpretations. For the models shown in equations 6 and 8, the influence of climate is on the fractional export of NANI, and a greater fraction of NANI is exported in watersheds with more precipitation (Equation 6) and higher discharge (Equation 8). This sort of model indicates that the background flux associated with natural sources of nitrogen in the landscape is small, and that the climate might have a major influence on riverine nitrogen flux by altering the fractional delivery of NANI. For the non-interactive type of model (Equation 7), there is still a strong influence of climate on riverine nitrogen flux, with higher fluxes in wetter environments. However, there is no influence of climate (precipitation) on the amount of NANI that is exported, which remains constant at 29% (see Equation 7), but rather only on the background or "natural" flux of nitrogen. This flux must originate with the natural rate of biological nitrogen fixation. According to this model, then, the higher riverine nitrogen fluxes in the watersheds with more precipitation are due to higher rates of biological nitrogen fixation in forests. These rates of fixation are not well known, but in our earlier estimates, the rates of fixation are in fact lower in the wetter watersheds (Boyer et al. 2002). The physical interpretation of Equation 7 suggests that the rates of nitrogen fixation in the wetter watersheds must be of the magnitude of 2,800 kg N km^{-2} $year^{-1}$ higher than in the driest of the 16 northeastern watersheds. As noted above, these rates would be at least an order of magnitude higher than likely for temperate-zone forests (Cleveland et al. 1999; Boyer et al. 2002). Thus, while both classes of models provide very good to excellent statistical fits to the data, only the interacting-term models (such as equations 6 and 8) lead to realistic interpretations. We conclude that the effect of climate on riverine nitrogen export is very likely to be on the fractional delivery of NANI (as illustrated in Figure 3a and b) rather than the background natural flux of nitrogen.

We can use these interacting-term models (equations 6 and 8) to begin to estimate how future climate change might affect riverine nitrogen fluxes. Najjar et al. (2002) provide estimates for future changes in precipitation and discharge for the mid-Atlantic coastal region, which includes roughly half of the 16 watersheds in our study. Their estimates, based on both the Hadley Centre and Canadian Climate Centre models for global climate change, suggest a mean likely increase in precipitation of 4% and of discharge of 2% by 2030, with increases in precipitation and discharge by 2095 of 15 and 11% respectively. Such estimates are quite uncertain (Table 5), but they provide a context for

Table 5. Predicted consequences of climate change on riverine nitrogen flux in the Susquehanna River to Chesapeake Bay. Estimates rely on the range and mean projections of change in precipitation and discharge by 2030 and by 2095 from Najjar et al. (2000) and on our equations 6 and 8, which relate riverine nitrogen flux to NANI and either precipitation or discharge. NANI is assumed not to change. Mean projected values are shown, with the range given in parentheses.

	2030	2095
Change in precipitation	+4% (−1% to +8%)	+15% (+6% to +24%)
Change in discharge	+2% (−2% to +6%)	+11% (−4% to +27%)
Change in riverine nitrogen flux, based on precipitation	+17% (−4% to +35%)	+65% (+26% to +200%)
Change in riverine nitrogen flux, based on discharge	+3% (−3% to +8%)	+16% (−6% to +38%)

examining the consequences on nitrogen fluxes. In Table 5, we illustrate the potential magnitude of changes in riverine nitrogen fluxes for the Susquehanna River basin due to future climate change, using both precipitation and discharge as predictors, and using both the mean projections and the range of projections given by Najjar et al. (2002). We chose the Susquehanna River for this analysis both because it is the largest of the watersheds in the northeastern United States and because it is the major input of nitrogen to main stem of Chesapeake Bay (Hagy et al. 2004), one of the most nutrient-sensitive estuaries in the country (NRC 2000). Further, the range of predicted future values for discharge and precipitation in the Susquehanna River basin is within the range of values currently observed across the climate gradient for the 16 northeastern U.S. rivers, so we need not extrapolate our models beyond the observational data upon which they are based. This is critical, particularly for precipitation, where the relationship between precipitation and fractional delivery of nitrogen inputs is quite steep (Figure 3a), and undoubtedly is not linear when the precipitation is less than 800 or greater than 1,300 mm year^{-1}.

For these predictions of the consequences of climate change on nitrogen fluxes in the Susquehanna, we assume that NANI remains constant into the future. Note that the estimate based on NANI and discharge (Equation 8) over-predicts the flux from the 1988 to 1993 period by 13%, while the estimate based on NANI and precipitation (Equation 6) underestimates this flux by 16%. Note also that all of the nitrogen projections given in Table 5 respond in a non-linear way to climate forcing, with the nitrogen increases or decreases larger than the respective changes in either precipitation or discharge. This nonlinearity is particularly pronounced for the estimates based on precipitation. While our projections obviously carry a great deal of uncertainty, they suggest that compared to the 1988–1993 period, nitrogen fluxes down the Susquehanna in 2030 may be 3 to 17% greater and in 2095 may be 16 to 65% greater in response to climate change (Table 5, using mean estimates based on the discharge and precipitation models). Such changes would obviously make it much more difficult to achieve nitrogen reduction for Chesapeake Bay.

While riverine nitrogen fluxes for the northeastern watersheds are very well explained on the basis of nitrogen inputs to the landscape and climate (Figures 5 and 6), it must be noted that a variety of management options are available for greatly reducing nitrogen fluxes in rivers without necessarily decreasing the net anthropogenic nitrogen inputs (NANI; see Howarth in press and Howarth et al. 2006 for recent reviews of some of these options). For example, planting winter cover crops on agricultural fields or switching from annual to perennial crops can greatly reduce nitrogen losses from the fields even when there is no reduction in fertilizer application (Randall et al. 1997; Staver and Brinsfield 1998; Randall and Mulla 2001). That NANI so well explains the riverine nitrogen fluxes, therefore, suggests that farming and other nitrogen management practices (such as wastewater disposal) during the 1988–1993 period of our study were relatively uniform across the watersheds. Great opportunity exists to improve these management practices, and thereby help reduce nitrogen pollution in coastal waters. Nonetheless, our results indicate that climate plays a significant role in determining the magnitude of the flux in rivers of nitrogen from human-dominated landscapes.

Given the uncertainties in our analysis, our conclusions must be tempered, and our projections must be qualified. Nonetheless, the analysis of the 16 northeastern watersheds illustrates that climate probably has a pronounced, sustained influence on the flux of nitrogen in large rivers. The percentage of nitrogen inputs to the landscape that is exported to coastal ecosystems by rivers is greater in the watersheds with wetter climates, probably because the nitrogen sinks in the landscape (primarily denitrification) are less. We believe that the relationships developed here should be tested in a wider set of large watersheds across as broad a climate gradient as possible. Should the relationships prove robust, then the influence of future climate change must be an important consideration in any management plans to control nitrogen inputs to coastal marine ecosystems.

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