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Variation in NO_3 Export from Flowing Waters of Vastly Different Sizes: Does One Model Fit All?

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Abstract

Flowing waters receive nitrogen (N) from the surrounding watershed and ultimately export much of this N to coastal waters, which in turn can be substantially affected by these inputs. Although the control of N export is complex, for large rivers among-system variation is predicted relatively well by simple models of human activity. Using data from 249 predominantly North Temperate watersheds that varied in size from 0.1 to over 1,000,000 km^2, we examined whether these simple models lose their predictive power at smaller scales. We found that the relationship between human population density and NO_3 export becomes weaker at smaller scales, and that for watersheds less than 100 km^2, it explains only 8% of the 1000-fold variation in NO_3 export. However, NO_3 export predicted from a simple loading model related well to measured NO_3 export across all scales; linear regressions of log modeled versus log measured export for small (less than 100 km^2), mid-sized (100–10,000 km^2), and large (more than 10,000 km^2) watersheds were all highly significant ($P < 0.01$) and had r^2 values of 0.78, 0.63, and 0.77, respectively. For the smallest systems, however, the model was biased and predicted higher NO_3 export than was measured. The bias suggests slightly greater storage or gaseous N loss in smaller watersheds, whereas the tight correlation between predicted and measured export indicates that for small as well as large systems, among-system variation in NO_3 export is controlled primarily by anthropogenic N loads rather than site-specific variations in soil or vegetation characteristics. Across all scales, however, predictive models can be improved by the inclusion of these local parameters.

Key words: NO_3 export; rivers; watersheds; human impacts; N biogeochemistry.

Introduction

Flowing waters are a major pathway of solute transport between landscapes (Meybeck 1982; Fisher and others 1998; Aumont and others 2001). These flows deplete upland soils of essential nutrients over time (Likens and others 1996) while enriching floodplain soils, as well as lakes, reservoirs, and coastal waters (Carpenter and others 1998). Exports are also of interest because in many ways they are integrated measures of the ecosystem function of a watershed (Likens and Bormann 1995). The scale of this integration varies with the size of the watershed. Thus, running waters can be used to understand not only how ecosystem characteristics (for example, water runoff, human population density) affect integrated ecosystem function, but also how size itself influences this function (Turner 1989; Peterson and Parker 1998).

Because nitrogen (N) is an important limiting nutrient in both terrestrial and aquatic systems (D'Elia 1987; Howarth 1988; Vitousek and Howarth 1991; Rabalais and others 1996), there are many studies of N export in flowing waters. These studies include watersheds that vary from less than 1 km^2 in size to large rivers with watersheds over 10^6 km^2

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(Meybeck 1982). These studies tend to have different foci at different scales of study. At the large, regional scale, there has been an emphasis on using correlations and very simple models to predict N export across systems (see, for example, Cole and others 1993; Howarth and others 1996; Jordan and Weller 1996). For example, in large rivers with global distribution, NO_3 export has been found to relate relatively well to just one variable—human population density in the watershed, which predicts 50% of the cross-system variance (Peierls and others 1991; Cole and others 1993). For these same systems, a simple model that includes anthropogenic N loads and water runoff explains over 80% of the variation in NO_3 export (Caraco and Cole 1999a). Studies of small watersheds, on the other hand, frequently emphasize the complex interactions and the proximate as well as the ultimate exogenous sources of N that drive N export (Sjodin and others 1997; Dodds and others 2000). In addition, small watershed studies have tended to focus on interannual variability or comparisons between relatively similar systems in a single region (see, for example, Vitousek and Reiners 1975; Gosz 1980; Mitchell and others 1999), rather than on broad-scale comparisons among very different systems. This difference in emphasis could be due either to a difference in control and complexity of control at different scales or to a difference in focus on the part of the researchers.

Nitrogen export from small watersheds might be less easily predicted from simple models because, as compared to large watersheds, many of the factors driving variability in export may operate more strongly. For example, whereas a small watershed may be severely impacted by disturbances such as an insect defoliation and as a result show greater N export (Eshleman and others 1998, 2001), such events may not greatly impact large watersheds, where only a small fraction of the watershed may be strongly disturbed (Turner 1989). Additionally, simple correlations that occur at large scales may not operate at small scales. For example, at the large scale, human population density may correlate well with both N loads to ecosystems (Cole and others 1993; Caraco and Cole 1999a) and the destruction of wetlands and riparian areas that can retain N (Correll and others 1992; Cole and others 1993; Jansson and others 1998). These correlations may be absent or far weaker at smaller scales. Thus, due to a number of potential factors, the relationship of N export to either human population density or anthropogenic N loads could become weaker at the small scales. Alternatively, these relationships could be strong but quantitatively different (for example, less or greater export per unit N load to the watershed) due to greater or lesser N retention in small as compared to large watersheds. This difference in retention could occur in the terrestrial ecosystem, in riparian areas, or within aquatic systems (Billen and others 1991; Mulholland 1992; Vörösmarty and others 1997; Behrandt and Bachor 1998; Fisher and others 1998; Lampman and others 1999; Alexander and others 2000; Peterson and others 2001).

In this study, we examined the scale dependency of N export. Previous studies have examined scale-dependent N export within single watersheds (see, for example, Dent and others 2001). Here we examined scale dependency among a suite of watersheds. Specifically, we used NO_3 export data from 249 watersheds that vary from less than 1 to more than 1,000,000 km^2 and tested to see whether simple correlations and models developed at the large scale break down at small scales due to increased variance or bias. Nitrogen is exported in a variety of forms. We focus on NO_3 export because models developed at the large scale accurately predict NO_3 export (Caraco and Cole 1999a); NO_3 is the most frequently measured form of N in river studies (Meybeck 1982); and NO_3 responds markedly to disturbance (Likens and others 1970; Hedin and others 1995; Caraco and Cole 1999b). We did not consider xeric systems (less than 0.1 m y^{-1} runoff) because large-scale models developed for predominantly mesic systems do not accurately predict the export from highly xeric systems (Caraco and Cole 2000). Thus, desert and dry grassland sites are not included. Furthermore, although the systems considered here are widely distributed (Cole and others 1993), they are predominantly located in North Temperate areas of the continental United States. As such, tropical and boreal areas are not well represented.

Methods

Data Sources

Of the 249 systems considered, 87 are large (more than 10,000 km^2). Thirty of these systems comprise the globally distributed data set used to construct the NO_3 export models (Cole and others 1993). The remaining large systems are from the US Geologic Survey (USGS) national monitoring network (WQN), (Alexander and others 1996). An additional 89 systems were moderately sized (100–10,000 km^2) and are primarily from the WQN (Alexander and others 1996) and from USGS data for the Hudson River Basin (Hudson–NWQA systems) (Phillips and Hanchar 1996), but additionally include rivers/streams in northern Italy (Montorsi and Dell'Arhiprete 1990; Marchetti and Verna 1992). The remaining 73 systems are between 0.1 and 100 km^2. The data for these systems are from a greater diversity of sources and include forested areas of New York (n = 39) (Lovett and others 2000), New Hampshire (n = 5) (G. E. Likens unpublished), New Mexico (n = 7) (Gosz 1980), Ohio (n = 1) (Taylor and others 1971), and Puerto Rico (n = 3) (McDowell and others 1990), as well as areas of mixed land use in New York (n = 14) (DCEMC 2000), Italy (n = 3) (Marchetti and Verna 1992; Montorsi and Dell'Arhiprete 1990), Wisconsin (n = 1) (Alexander and others 1996), and Ohio (n = 1) (Taylor and others 1971).

Large River Predictions

We first tested to see whether human population density per se relates well to NO_3 export (in kg km^{-2} (watershed) y^{-1}), as it does for large rivers with global distributions (Cole and others 1993; Caraco and Cole 1999a). We then tested to see whether a simple loading model developed for the same large rivers (Caraco and Cole 1999a) accurately predicted the export of NO_3 across all sizes of watershed. This simple loading model predicts NO_3 export from sewage loads directly to the river (Sew) and watershed loads by atmospheric deposition of NOy (Atm) and inorganic fertilizer application (Fert). Thus,

$$NO_3 \text{ export} = F\text{River}_{\text{export}}[(\text{Sew} + F\text{WS}_{\text{export}}(\text{Atm} + \text{Fert})]$$

where $F\text{River}_{\text{export}}$ and $F\text{WS}_{\text{export}}$ are the fraction of NO_3 loaded to the river and watershed, respectively—that is, exported (rather than being stored in soils or plant biomass or lost to the atmosphere). $F\text{River}_{\text{export}}$ is taken as 0.7 for all systems; $F\text{WS}_{\text{export}}$ is a function of water runoff (Runoff) in m y^{-1}. Thus,

$$F\text{WS}_{\text{export}} = 0.45\ \text{Runoff}^{0.82}$$

For water runoffs of 0.3 and 0.6 m y^{-1}, this implies that $F\text{WS}_{\text{export}}$ is 0.17 and 0.30, respectively.

Parameter Calculation

Average annual NO_3 export (kg km^{-2} (watershed) y^{-1}) was either given (Taylor and others 1971; Gosz 1980; Cole and others 1993; McDowell and Asbury 1994; Phillips and Hanchar 1996; Lovett and others 2000) or was calculated from water runoff (m y^{-1}) and average annual NO_3 concentration (Alexander and others 1996; DCEMC 2000). The data cover between 1 and 5 years for the various systems.

Methods for evaluating predictive parameters varied between data sets. The methods to evaluate human population density, as well as to assess Atm, Fert, Sewage, and Runoff for the 30 large systems used to develop the relationships in Eq. (1) and Eq. (2), are described in Cole and others (1993) and Caraco and Cole (1999a). For the remaining data sets, we relied as much as possible on previously compiled information on the above parameters, but in some cases it was necessary to calculate several parameters.

For the Hudson-NWQA systems, human population data, as well as sewage, atmospheric and fertilizer loads and runoff, are from Phillips and Hanchar (1996). For the WQN sites, runoff was from flow and watershed size data (Alexander and others 1996). Sewage loads were calculated as the product of sewered population in the watershed and a per-person sewage N input of 1.9 kg N y^{-1} (Caraco and Cole 1999a). Sewered population was in turn calculated as the product of population in the watershed (Alexander and others 1996) and percent of population sewered in each state (van der Leeden and others 1990). Fertilizer inputs are from cropland area in each watershed (Alexander and others 1996) and estimates of fertilizer use per unit crop area for the state where these rivers occurred (NRC 1993). For estimates of both sewage and fertilizer inputs, where watersheds were present in several states, we used weighted average values from various states included in the watershed.

Atmospheric loads for the WQN systems were averages of NADP estimates for sampling locations in or near the watersheds (nadp.sws.uiuc.edu). The NO_3 deposition from NADP is wet deposition. To convert these numbers to total NO_3 deposition, we multiplied by 2 (Caraco and Cole 1999a). For the rivers in northern Italy, human population and fertilizer loads are from Marchetti and Verna (1992). Atmospheric inputs were values for the Po River in northern Italy (Caraco and Cole 1999a). For the small forested and agriculture streams in Ohio, loads and runoff are from Taylor and others (1971). For watersheds of Dutchess County, New York, with mixed land use, fertilizer was calculated from the area of cropland in each watershed (DCEMC 2000) and countywide fertilizer use per unit crop area (Alexander and Smith 1990). Human population was calculated from estimates of land area in low-, mid-, and high-density development (DCEMC 2000). Atmospheric inputs were based on measurements at the Institute of Ecosystem Studies, located in Dutchess County, New York (www.

Table 1. Characteristics of the Three Size Classes of Watersheds in this Study

		Mean	Min	Max
Forest	Small	86	10	100
	Mid-sized	44	2	94
	Large	33	2	92
Agriculture	Small	7	0	69
	Mid-sized	37	3	92
	Large	53	3	93
Developed	Small	4	0	36
	Mid-sized	8	0.2	43
	Large	3	0.2	19
NO3 export	Small	360	5	4567
	Mid-sized	630	25	2884
	Large	443	3	1873
Runoff	Small	0.89	0.1	3.6
	Mid-sized	0.54	0.12	1.1
	Large	0.42	0.1	1.4
Population density	Small	38	0	995
	Mid-sized	117	0.1	707
	Large	53	0.1	300
Watershed load	Small	1195	190	4468
	Mid-sized	2239	170	11,803
	Large	2197	84	8758

For small (less than 100 km^2), mid-sized (100–10,000 km^2) and large (more than 10,000 km^2) watersheds, average and range of values are shown.
The first three parameters (Forest, Agriculture, Developed) are watershed land-use variables and are given as % areal coverage of the watersheds.
NO_3 export is in kg N km^{-2} (watershed) y^{-1}, water runoff is in m y^{-1}, human population density is in ind. km^{-2}, and loads to the watershed from atmospheric (Atm) and fertilizer (Fert) inputs are in kg N km^{-2} (watershed) y^{-1}.

ecostudies.org). For the completely forested small systems (where sewage and fertilizer loads do not occur), atmospheric loads were given for the individual watersheds where NO_3 export and runoff was measured (Gosz 1980; McDowell and Asbury 1994; McDowell and others 1990; Lovett and others 2000).

Results

The watersheds examined in this study are primarily North Temperate but vary substantially in land use among the 249 watersheds (Table 1). Land use was considered in the following three categories: forest, developed lands, and agriculture (nrcs.usda.gov/technical/land/lgiv/m51501.gif). Here agriculture includes crop, pasturelands, and rangeland. On average, forest and agricultural areas account for 55% and 31%, respectively, of the land area, and developed areas account for 5%. The range of variation between different land uses for the entire data set are (a) forest, 1%–100%; (b) agriculture, 0–95%; and (c) developed, 0–43%. All three size classes of watersheds had large among-system variation in land use (Table 1). On average, however, small watersheds had a substantially greater proportion of forested areas and a lower proportion of agricultural areas than the moderate and large watersheds (Table 1). However, all three size classes include systems dominated by both forested area and agriculture.

In addition to the variation in land use, there was a large among-system variation in NO_3 export and the anthropogenic variables that might drive this export. NO_3 export varied between 3 and 4567 kg N km^{-2} (watershed) y^{-1} across the different watersheds; human population density varied between 0 and 995 ind. km^{-2}; and N loads to the watershed from a combination of fertilizer and atmospheric inputs varied between 84 and 11,800 kg N km^{-2} (watershed) y^{-1}. The variation in both NO_3 export and potential controlling variables was high for not only the entire data set but also for each of the three size classes of watersheds (Table 1).

Across all size classes, the variation in NO_3 export was significantly related to human population density ($P < 0.05$, linear regression) (Figure 1A–C), but the strength of this relationship varied with watershed size. For the entire data set human population density explained 22% of the variation in NO_3 export. For the small, mid-sized, and large watersheds, human population density explained 8%, 28%, and 46%, respectively, of the variance in NO_3 export. When the 30 large systems used to develop the relationship between human population density and NO_3 export are eliminated from the regression analysis, 44% of the variance in NO_3 export is explained by human population density in the watershed. As compared to the original 30 systems with global distribution, the 57 remaining systems, which are all in the continental United States, had higher NO_3 export for a given human population density in the watershed (Figure 1C).

The variation in NO_3 export from the 249 watersheds showed a significant positive relation with water runoff in a linear regression analysis ($P <$ 0.05). However, runoff explained only 8% of the variance of the entire data set and 24%, 11%, and 9% of the variance in NO_3 export in the small, mid-sized, and large watersheds, respectively. A simple model (Eq. [2]) that included both water runoff and anthropogenic N loads predicted the variation in NO_3 export far more accurately. For the entire data set, the variation in modeled and measured NO_3 export was well correlated ($P < 0.001$, $r^2 = 0.71$). Furthermore, the modeled export correlated well to measured NO_3 export for all size classes of watersheds. For small, mid-sized, and

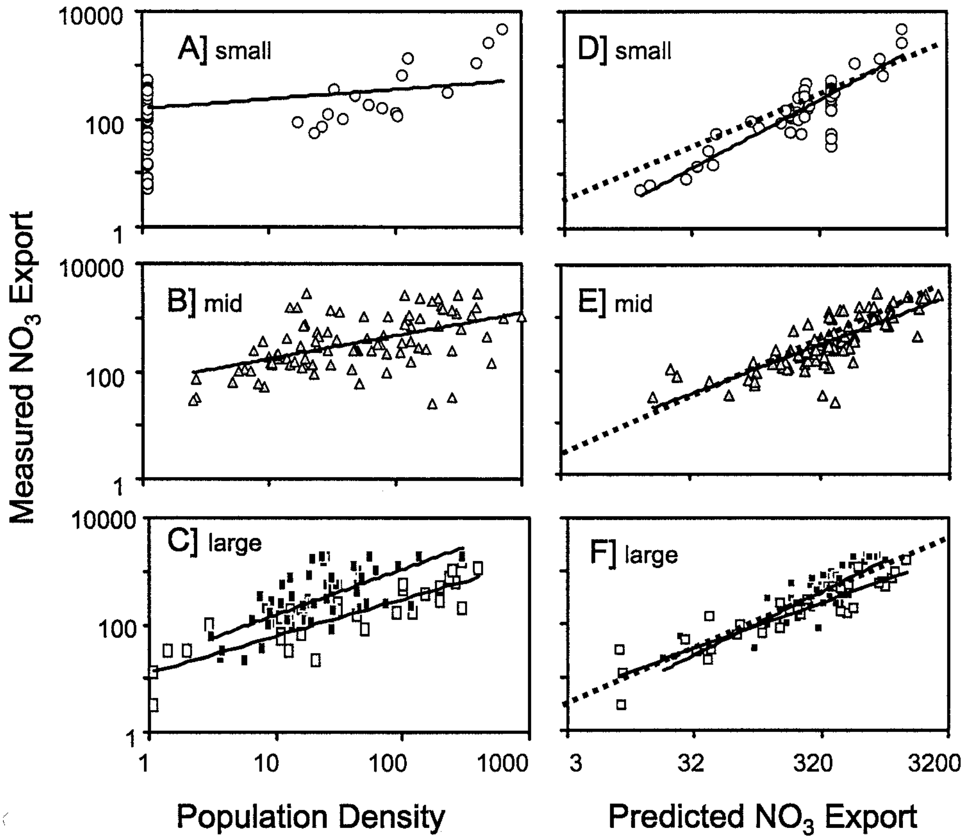

Figure 1. Annual NO_3 export (kg N km^{-2} (watershed) y^{-1}) from streams and rivers versus human population density in the watershed (**A–C**) and export predicted from a simple loading model (**D–F**). Watersheds are divided into three size classes: less than 100 km^2 (A and D, *open circles*), 100–10,000 km^2 (B and E, *triangles*), and more than 10,000 km^2 (C and F, *squares*). For the large watersheds, the original 30 systems used to develop the relationship between N load and human population density (C) and the prediction of NO_3 export based on N loads (F) are shown with open squares; the remaining 57 systems are shown with closed squares. In A through F, solid lines represent linear regression lines; for D–F, the dashed lines are the 1:1 line for observed versus predicted export. For the large systems, the regression lines of the original data set of 30 and the new data set of 57 are shown separately. Parameters for the regression lines are shown in Table 2.

large watersheds, 78%, 63%, and 77% of the variance in log-NO_3 export was explained, respectively (Figure 1D and F). Eliminating the 30 large watersheds used to develop the modeled export does not significantly change these results; in the remaining 57 large watersheds, 76% of the variation in NO_3 export was explained by the loading model (Figure 1F).

Although modeled export related well to measured export for small watersheds (Figure 1D), the model was biased and on average predicted higher export than was actually observed (paired *t*-test, $P < 0.001$). For the small watersheds, the model predicted NO_3 export greater than was actually measured in 71% of the cases (29% underpredicted); significant overpredictions (greater than twofold off) occurred in 19% of the cases, as compared to only 1% of significant underpredictions (greater

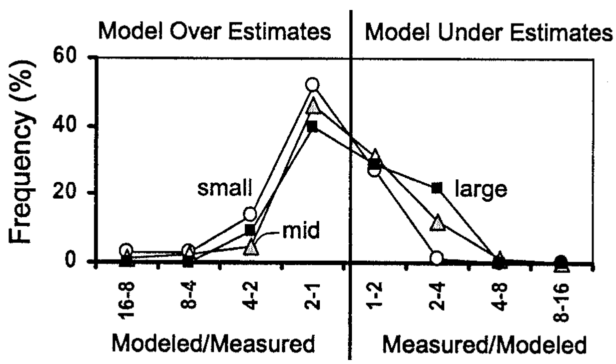

Figure 2. Frequency distribution model (Eq. [2]) of overpredictions and underpredictions of NO_3 export for small (*circles*), mid-sized (*triangles*), and large watersheds (*squares*). The overpredictions are expressed as Modeled/Measured NO_3 export; the underpredictions are expressed as Measured/Modeled NO_3 export. In both cases, data are divided in log 2–based categories.

Table 2. Parameters Describing the Relationship between NO_3 Export and Human Population Density in the Watershed (Population) and Modeled NO_3 Export (Modeled Export) for 249 Watersheds by size Class

		Small	Mid-sized	Large (all)	Large (new)	Large (global)
Population	Int.	2.19	1.82	1.42	1.31	1.11
	Slope	0.18	0.43	0.67	0.88	0.69
Modeled export	Int.	–0.79	0.11	0.05	–0.35	0.25
	Slope	1.29	0.95	0.99	1.18	0.86

Both slopes and intercepts (Int.) are for log-log regressions.
The parameters for the large watersheds are shown for the entire data set (all), for the global data set used to develop the log-log relationships (global), and for the new data (new) added in analysis for this manuscript.

than twofold off) (Figure 2). For the mid-sized and large watersheds, on the other hand, the model did not significantly overpredict actual export (paired *t*-test, $P > 0.05$). For mid-sized watersheds, the model predicted greater NO_3 export than was observed in 54% of the cases (46% underpredicted). For large watersheds, overpredictions and underpredictions were balanced at 50%.

Discussion

For small, mid-sized and large watersheds, the among-system variation in NO_3 export appears to be driven predominantly by human activities. Anthropogenic N loads may be particularly important in this regard. Control by anthropogenic N loads is supported by the fact that, although water runoff poorly predicts the NO_3 export, simple models that consider only anthropogenic N loads and water runoff (Eqs. [1] and [2]) predict this export relatively well at all scales (Figure 1D–F). Furthermore, the fact that human population density itself does not predict export well at small scales appears to be due to a poor correlation between human population and anthropogenic N loads at the small scale. For medium and large watersheds, there is a significant positive relationship between human population density and either sewage loads, fertilizer loads, or atmospheric loads ($P < 0.05$, log-log regression). For small watersheds, although human population density is related significantly to fertilizer inputs and point sewage loads ($P < 0.05$, log-log regression), there was no significant relationship between human population and atmospheric loads ($P = 0.62$, log-log regression).

Predictions from a simple loading model developed at the large scale correlate well with measured NO_3 export across all scales. However, these models generally overpredict export at small scales (Figures 1D and 2). Thus, $F\text{River}_{export}$ and/or $F\text{WS}_{export}$ in small watersheds are somewhat higher than values in large or moderate-sized watersheds (Eqs. [1] and [2]). If atmospheric N loads, which in large part are deposited on forested areas, are stored in watersheds to a greater degree than fertilizer is loaded to agricultural lands, a lower $F\text{WS}_{export}$ in small watersheds could be explained by the greater proportion of forest area in the small watersheds of our data set (Table 1).

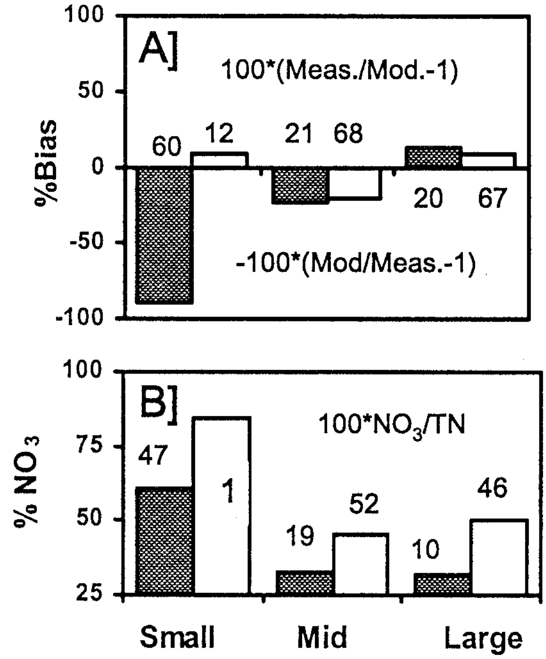

Figure 3. (**A**) Average bias of NO_3 export predicted from simple load model for small, mid-sized, and large watersheds (see Figure 1D–F). For each size class, watersheds are divided into those with N inputs dominated by atmospheric loads (*dark bars*) and those not dominated by atmospheric loads (*light bars*). The equations for calculating bias are shown on the figure. Mod. and Meas. are export predicted from the model (Eq. [2]) and measured, respectively. (**B**) Average importance of NO_3 export relative to the export of other forms of N (%NO_3 export) for the same size and atmospheric dominance classes as in A. For both panels, numbers next to bars are the size of the data set in each of the six subcategories.

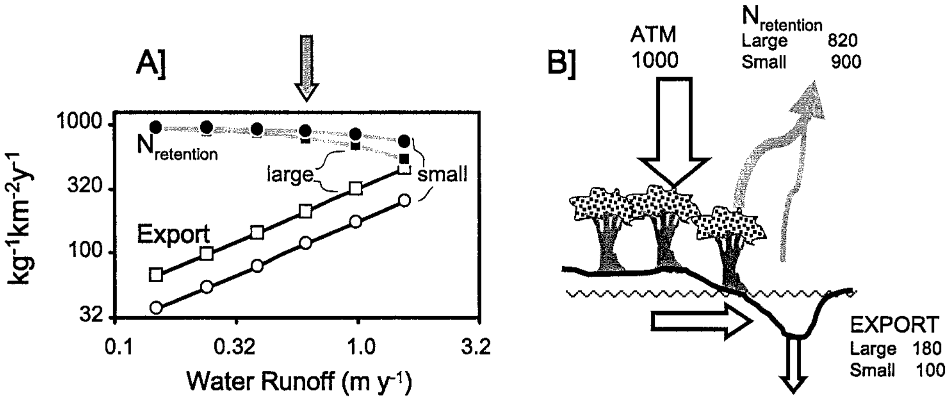

Figure 4. (**A**) NO_3 export (kg N $m^{-2}y^{-1}$) (*open circles* and *squares*) and the sum of N storage and N gas emissions ($N_{retention}$) (*closed circles* and *squares*) versus water runoff for an atmospheric NO_y load (Atm, Eq. [2]) of 1000 kg N $m^{-2}y^{-1}$. Both export and $N_{retention}$ are shown for large watersheds (*squares*) and small watersheds (*circles*). The large watershed results are based on Eq. (2). The small watershed results are based on the observed 80% lower export from small as compared to large or mid-sized watersheds (see Figure 3A). The arrow at the top of the graph marks the data for 0.5 m y^{-1} water runoff. (**B**) Cartoon of fates of atmospheric N loads in large and small predominantly forested watersheds for a water runoff of 0.5 m y^{-1}. Note that to explain a 1.8-fold lower NO_3 export from small watersheds, only a very small fractional increase in $N_{retention}$ is required.

When we consider, however, only watersheds dominated by atmospheric inputs (atmospheric N loads comprise more than 50% of total loads), our conclusions about model overpredictions do not change; the model predicts significantly higher NO_3 export than was observed for small systems ($P < 0.05$, paired *t*-test) but not for medium or large ones (Figure 3A). Lower NO_3 export does not necessarily imply lower total N export, because it is possible that small watersheds have a greater proportion of N export in other forms, such as dissolved organic N (Hedin and others 1995; Perakis and Hedin 2002). However, our data suggest the opposite: Small watersheds, on average, have a higher proportion of N export as NO_3 than larger watersheds (Figure 3B).

Thus, our data suggest that the sum of N storage and gaseous N loss ($N_{retention}$) (Figure 4A, B) is somewhat greater in small watersheds than in moderately sized or large watersheds. The surplus $N_{retention}$ could occur within streams themselves (Alexander and others 2000; Peterson and others 2001; Bernhardt and others 2002), riparian areas (Fisher and others 1998), or in the soils of small watersheds (Lawrence and others 2000).

Although small watersheds, or at least small forested ones, may have slightly greater $N_{retention}$ than moderate-sized or large watersheds, across all sizes of watersheds $N_{retention}$ appears to be quite large (Figure 4A, B). When $N_{retention}$ is large, relatively small changes in storage or gaseous N loss can result in relatively large increases in NO_3 export. For example, our N loading model (Eqs. [1] and [2]) suggests that for a system with a water runoff of 0.5 m y^{-1} decreasing watershed retention by 10% will nearly double NO_3 export (Figure 4B). This example shows that a knowledge of the controls on $N_{retention}$ is critical to understanding and accurately predicting N export. The simple loading model considered here uses only water runoff from the watershed to predict the fraction of N loaded to the watershed that is exported. Although this variable is likely an important control on N export from watersheds (McIsaac and others 2001), N export and $N_{retention}$ are clearly controlled by a great number of additional variables, including wetland and reservoir abundance and distribution, watershed disturbance, soil organic content, vegetation, and past N loading (Likens and others 1970; Eshelman and others 1998; Lovett and others 2000; Lawrence and others 2000; McIsaac and others 2001). Parameterization of these variables for use at a number of scales would allow inclusion of these variables into simple models. The revised models could not only enable better predictions of present-day NO_3 export from small, mid-sized, and large watersheds, but also help us to understand and predict future changes in NO_3 export from these systems.

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