SUPPORTING INFORMATION

INFLUENCE OF CLIMATE AND HUMAN ACTIVITIES ON THE RELATIONSHIP BETWEEN WATERSHED NITROGEN INPUT AND RIVER EXPORT

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DATA COLLECTION

Major sources of data to construct NANI budgets for watersheds of the LMB at 5-year intervals from 1974 to 1992 include the United States Department of Agriculture (USDA), National Atmospheric Deposition Program/National Trends Network (NADP/NTN), Clean Air Status and Trends Network (CASTNET), Environmental Protection Agency (EPA), USDA National Agricultural Statistics Service (USDA/NASS), United States Geological Survey (USGS), and others.

1. Census of Agriculture

1.1 Crop data

For this study, the term "crop" includes all corn for grain and silage, wheat, oats, barley, rye, soybeans, potatoes, sorghum, alfalfa hay, other hay (consisting of all hay excluding alfalfa hay), cropland pasture, and non-cropland pasture (consisting of all pastureland excluding cropland pastureland). Most county-level data on crop acreages as well as crop production were retrieved from the Census of Agriculture for 1974 to 1992 at five-year intervals, for which an electronic version is available from the USDA web site (http://www.nass.usda.gov) and the Mann Library, Cornell University (http://agcensus.mannlib.cornell.edu/).

1.2 Animal data

The Census of Agriculture also reports the calendar end-of-year inventory and sales data for livestock groups at the county level. This study included all cattle and calves, hogs and pigs, poultry, horses, and sheep and lambs as the livestock associated with N dynamics in agriculture. **Table M1** summarizes the inventory and sales data for the number of head of these livestock groups reported in the Census of Agriculture. Data availability varies among states or counties, years, and livestock types. Moreover, to protect the confidentiality of respondents, for counties that have only one farm operation for a specific group of livestock, the Census of Agriculture does not publish the population data, marking them "non-disclosed". We estimated these non-disclosed or missing data following others (*1, 2*) as shown in **Table M1**.

2. Fertilizer data

Historical N inputs from fertilizer application in the Lake Michigan watersheds from 1974 to 1992 were estimated using three different fertilizer datasets, including county-level fertilizer data for the years 1974 to 1982 provided by the USGS Branch of Systems Analysis, the county-level fertilizer sales data for 1987 provided by USGS Water Resources Division (WRD) (*3*) and county-level fertilizer input for years 1992 to 2002 provided by USGS National Water-Quality Assessment Program (http://water.usgs.gov/pubs/sir/2006/5012/excel/Nutrient_Inputs_1982-2001jan06.xls)(*1*).

The fertilizer use or sales datasets for 1974 to 1982 and for 1987 to 2002 were processed under different assumptions and computations to disaggregate state-level fertilizer use or sales data to the county level. For the 1974 to 1982 dataset, county-level fertilizer use was assumed to be directly proportional to a county's fertilized acreage, which refers to the total acreage of cropland, pastureland, and rangeland treated with chemical fertilizer. This was estimated using Equation 1:

$$FC_{ik} = FS_i \times \frac{FAC_{ik}}{FAS_i} \qquad (1)$$

where FC_{ik} is county-level fertilizer use for the i^{th} state and k^{th} county, FS_i is state-level fertilizer use for the i^{th} state, FAC_{ik} is county fertilized acreage for the i^{th} state and k^{th} county, and FAS_i is state fertilized acreage for the i^{th} state.

To determine annual county-level fertilizer sales data for 1987 to 2002, estimates of annual state-level sales were multiplied by the ratio of county to state expenditures for commercial fertilizer, which were calculated from the Census of Agriculture for the corresponding years (*4-6*).

3. Atmospheric N deposition and national emission data

Data describing wet and dry deposition of N species are available from the National Atmospheric Deposition Program/National Trends Network (NADP/NTN) (*7*) and from CASTNET (Clean Air Status and Trends Network (CASTNET) (*8*). The GIS point coverages for all NADP/NTN and CASTNET stations within OH, IN, IL, MI, and WI were obtained from EPA's Clean Air Mapping and Analysis Program (C-MAP) GIS electronic database.

Annual dry deposition of particulate ammonium (NH_4^+), gaseous nitric acid (HNO_3), and particulate nitrate (NO_3^-) was obtained for the period from 1989 through 2004 from CASTNET. Only sites meeting data completeness criteria for each year were included to create isopleth maps of inorganic N deposition for this study.

Because data availability for wet deposition of N for the years prior to 1980 within the study region is very limited, atmospheric deposition of NO_y was estimated from national trends in nitrogen oxide (NO_x) emissions for 1960 to 2000. National estimates of NOx emissions were compiled from two EPA emission trend reports for the years 1960-1989 (*9*), and for 1990-2000 (*10*).

Historical trends in NH_3 emissions for the U.S. were constructed using data from the NH_3 emission inventory of the Hundred Year Database for Integrated Environment Assessments (HYDE) for the United States from version 2.0 of the Emission Database for Global Atmospheric Research (EDGAR 2.0) for the years 1890 through 1990, at 10 year intervals (*11*). In this source, NH_3 emissions are calculated using an emission factor approach based on historical activity statistics and selected emission factors. In addition, these data sources provide the NH_3 emission inventory for four anthropogenic source categories with consistent source definitions: 1) fuel combustion (for power supply, domestic, industry, and transportation uses), 2) industrial processes, 3) agriculture (from livestock agriculture and fertilizer application), and 4) waste handling (landfill, agricultural waste burning, and wastewater treatment).

Table M1. Inventory and sales data for livestock groups used in this research and reported by the Census of Agriculture, along with the computation to estimate livestock numbers if the population was reported as non-disclosed or missing (This table is modified from two studies (*1, 2*).

Livestock group from census of agriculture		Computation used if the population was reported as non-disclosed
Cattle and Calves		
End-of-year inventory	Cattle and calves	—
	Cows and heifers that had calved	—
	Beef cows	0.5 × Cows and heifer that had calved
	Milk cows	0.5 × Cows and heifer that had calved
	Heifer and heifer-calf	0.5 ×(Total Cattle and calves – (Cows and heifers that had calved))
	Steers, steer calves, bulls and bull calves	0.5 ×(Total Cattle and calves – (Cows and heifers that had calved))
Sales	Calves sold weighing less than 500 pounds	—
	Cattle and calves sold weighing more than 500 pounds	—
	Number of fattened cattle	—
Hogs and pigs		
Inventory	Hogs and pigs used for breeding	—
	Other hogs and pigs	0.5 × (Hogs and pigs sold including feeder pigs-feeder pigs sold for further feeding)
Sales	Hogs and pigs sold including feeder pigs	—
	Feeder pigs sold for further feeding	—
Sheep and lambs		
Inventory	Sheep and lambs	—
Sales	Sheep and lambs sold	—
Horses		
Inventory	Horses	—
Sales	Horses sold	—

Table M1. Continued

Livestock group from census of agriculture		Computation used if the population was reported as non- disclosed
Poultry		
end-of-year inventory	Chickens 3 months old or older	—
	Hens and pullets of laying age	Chickens 3 months old or older
	Pullets 3 months old or older, not of laying age	—
	Pullet chicks and pullets under 3 months old	—
	Broilers and other meat-type chickens	—
	Total Turkeys	
	Turkeys for slaughter	(Total turkeys - turkeys for breeding) or Total turkeys
	Turkeys for breeding	
Sales	Chickens 3 months old or older sold	—
	Hens and pullets of laying age sold	Chickens 3 months old or older
	Pullets 3 months old or older, not of laying age sold	—
	Pullet chicks and pullets under 3 months old sold	—
	Broilers and other meat-type chickens sold	—
	Turkeys sold	
	Turkeys for slaughter sold	Turkeys sold

4. Population census data

County-level annual population estimates for 1970 to 2004 were obtained from the United States Census Bureau(*12-15*).

5. Land use

All available GIS data on land use or land cover were obtained from USGS and EPA, including the 1:250,000-scale Geographic Information Retrieval and Analysis System (GIRAS) Land Use and Land Cover (LU/LC) and the National Land Cover Data (NLCD) derived from 30-meter Landsat thematic mapper (TM) data. The GIRAS LU/LC data were created from high-altitude aerial photographs from the mid-1970s to early 1980s (*16*) and were coded using the Anderson classification system (*17*), which is a hierarchical system of general (level 1) to more specific (level 2 and higher) characterization. The NLCD are more recent, and include data for 1992, 2000, and the enhanced version of 1992 NLCD (hereafter referred as to NLCDe). The latter, published in 2005, includes four new classifications in addition to the original 21 land cover classifications of NLCD92. **Table M2** lists the 21 land use classifications for the NLCD and NLCDe. We used the classifications "Row crops (code 82)", "Small grains (83)", "Fallow (84)", "Orchards/vineyards/others (61)", "LULC orchards/vineyards/other (62)", "Low intensity residential (21)", "LULC residential (25), "NLCD/LULC forested residential (26)," and "urban recreational grasses (85)" when computing the fertilized area.

Table M2. The numeric codes and land cover classifications of the "enhanced" version of the National Land Cover Data 1992 (NLCDe 92)

Code	Classification	Code	Classification
11	Open water	51	Shrubland
12	Perennial Ice and Snow	61	Orchards/vineyards/other
21	Low intensity residential	62	LULC orchards/vineyards/other
22	High intensity residential	71	Grasslands/herbaceous
23	Commercial/industrial/transportation	72	LULC tundra
25	LULC residential	81	Pasture/hay
26	NLCD/LULC forested residential	82	Row crops
31	Bare Rock/Sand/Clay	83	Small grains
32	Quarries/Strip mines/gravel pits	84	Fallow
33	Transitional	85	Urban/Recreational grasses
41	Deciduous forest	91	Woody wetlands
42	Evergreen forest	92	Emergent herbaceous wetlands
43	Mixed forest		

NANI BUDGETING METHODS

1. Fertilizer

We estimated fertilizer use in each watershed from county-based N fertilizer use data for 1974, 1978, and 1982 (*18*) and from sales for 1987 (*3*) and 1992 (*19*), aggregated to the watershed scale using the fraction of land that is included within the watershed boundary.

2. Net trade of N in food and feed

Net trade of N in food and feed was calculated as crop and animal production minus human and animal consumption requirements. Human N consumption was estimated by multiplying annual human population estimates (*12-14*) by per capita N consumption rates obtained from the USDA Economic Research Service (*20*). Animal-specific N consumption rates from the National Research Council (*21-24*) were combined with the average numbers of animals during a given census year to estimate animal N consumption. Using Equation 2, the average animal population for a year was quantified based on information on the multiple marketings per year for individual classes of livestock (**Table M3**) and using data on sales and inventory of livestock from the Census of Agriculture (*2*).

$$AL = \left\{ \left(inventory \times \frac{1}{Cycles} \right) + \left(\frac{Sales}{Cycles} \times \frac{Cycles - 1}{Cycles} \right) \right\} \qquad \text{Equation 2}$$

where *AL* is the annual average number of livestock, *inventory* is the number from the end-of-year inventory data, *Sales* is the number from sales data, and *Cycles* is the duration of the life cycle (the number of days from birth to market) per year, equating to

$$\frac{365}{Life\ Cycle\ (numbers\ of\ days\ from\ birth\ to\ market)}$$

Supporting information Table M3. Computation of the average number of each livestock type during the year, along with the life cycle time from birth to market, and the products produced by the livestock types during the life cycle or after slaughter (Adapted from Kellogg et al. (*2*))

Livestock group		**Computation of the average number of head on livestock type during the year**		**Life cycle Days**	**Average live weight [a] (kg/head/yr)**	**N content of manure [b] (kg-N/head/yr)**	**Emission factors [b] (kg-N/head/yr)**
Cattle and Calves							
Cows	Milk cows	Milk cow inventory		365	650	99.9	40.2
	Beef cows	Beef cow inventory		365	460	67.2	5.4
Slaughtered cattle (Fattened cattle*)		Fattened cattle sales × (140/365)		140	403	46.4	18.6
Young (milk+beef) Calves		Cattle less than 500 pounds sold × (150/365)		150	98	9.6	0.8
Heifer	Beef heifer for replacement herd	0.15 × Beef cow inventory × (150/365)	Heifer and heifer calves inventory × (200/365)	150	403	35.3	2.3
	Dairy heifer for replacement herd	0.2 × Milk cow inventory × (150/365)		150	489	32.5	2.8
Beef stockers		(Beef stockers inventory [c] + beef stockers sold [d]) ×(200/365)		200	266	25.2	10.1
Hogs and Pigs							
Hogs for breeding		Breeding hog inventory		140	114	16.0	8.2
Hogs for slaughter		Other hogs and pigs inventory × (1/2) + (Hogs and pigs sold – Feeder pigs sold for further feeding) × (1/2) × (1/2)		180	34	11.6	6.0

Supporting information Table M3. Continued-

Livestock group		Computation of the average number of head on livestock type during the year	Life cycle Days	Average live weight [a] (kg/head/yr)	N content of manure [b] (kg-N/head/yr)	Emission factors [b] (kg-N/head/yr)
Poultry						
Hens (Laying eggs)		Hens of laying age inventory	365	2	0.56	0.22
Pullets (Before egg laying)	Pullets more than 3 months	Inventory of pullets × (146/365) + Sales of pullets× (146/365) × (1.25/2.25)	146	1.5	0.41	0.18
	Pullets less than 3 months			1.5	0.23	0.1
Broiler		Broiler Inventory × (60/365) + Sales of Broiler× (60/365) × (5/6)	60	1.7	0.40	0.18
Turkeys	Turkeys for breeding	Turkey hens for breeding inventory	365	8.5	1.68	0.75
	Turkeys for slaughter	Slaughter Turkeys inventory × (1/2) + Slaughter Turkeys sold × (1/2) × (1/2)	180	6.4	1.85	0.83
Horse		Horses Inventory	365	NA [g]	68.9	13.78
Sheep and Lambs		Sheep and Lambs Inventory	365	NA [g]	3.0	2.01

[a] Average live weight for the period 1987-1992
[b] Average values for four states (IL, IN, MI and WI) for period 1990-2000s
[c] Beef Stockers Inventory = Steers[e] + Heifer and Heifer calves Inventory - Beef and Dairy Heifer for replacement Herd
[d] Beef stockers sold = Cattle more than 500 pounds sold – Fattened Cattle Sold –Beef and Diary Cow sold
[e] Steers = Steers and Bulls inventory –Bulls [f]
[f] Bulls= minimum of (0.05×beef cow inventory) or steer and bull inventory
[g] Not available, so assumed to be the same as beef cows.

Crop N content (*25, 26*) was combined with county-level crop yield data from the Census of Agriculture for corn, soybean, wheat, alfalfa hay, other hay, sorghum, barley, oats, rye, and potatoes. Assumptions about crop products lost to spoilage or other causes as well as allocation of crop products to animals and humans were applied (*27*) (**Table M4**).

Table M4. N content of harvested crops and partitioning ratio used to classify crops as livestock feed or human food, by commodity, modified from three studies (*25-27*)

Crop type	Yield unit (YU)	Nitrogen content (kg-N/YU)	Fraction of crops fed to humans (%)	Fraction of crops fed to animals (%)	Proportion remaining after handling loss (%)
Field corn, for grain	Bushel	0.80	4	96	90
Field corn for silage	Ton	3.22	0	100	100
Wheat	Bushel	0.50	61	39	90
Oats	Bushel	0.27	6	94	90
Barley	Bushel	0.41	3	97	90
Sorghum for grain	Bushel	0.44	0	100	90
Sorghum for silage	Bushel	6.70	0	100	100
Irish potatoes	Cwt.	0.16	100	0	90
Rye for grain	Bushel	0.49	17	83	90
Alfalfa hay	Ton	22.87	0	100	100
Other hay	Ton	9.86	0	100	100
Soybean	Bushel	1.61	2	98	90
Crop pasture	Acre	2000.00	0	100	90
Non-crop pasture	Acre	1000.00	0	100	90

Animal production was estimated from data summarizing the sale of slaughtered livestock, combined with N content of their edible portion and the varying weights of slaughtered livestock by year (**Table M5**). The N content of the edible portion was obtained from the USDA National Nutrient Database for Standard Reference (*28*), and annual average live weights of cattle, calves, swine, sheep and lambs for each state (IN, IL, MI, and WI) for the period from 1974 to 1992 were obtained from USDA NASS state-level annual and monthly livestock slaughter summary reports (*29, 30*).

Table M5. Summary of rates used to estimate animal N products.

Animal	Animal edible product	Average weight per animal in kg [a]	Edible portion yield [c] as % of live weight	N % in edible portion = Protein (%) × 0.16
Cattle	Beef	463	42.2	4.8
Calf	Veal	103	41	3.2
Pigs & Hogs	Pork	112	53.6	0.52
Sheep	Lamb	44.6	49.8	4.8
Layer	Chicken	2.16	73	2.16
	Egg	0.058	89[d]	1.76
Broiler	Broiler	1.71	69	1.71
Turkey	Turkey	8.51	79	2.93
	Milk	9091[b]	100	0.496

[a] The average live weight per animal at the market during 1974-1992
[b] The weight of milk production per head of milk cow in kg/head/yr
[c] Edible portion only includes separate lean, trimmed to 0" fat, excluding hair, skins, bones, fats and intestines
[d] The proportion of the edible portion of a whole egg excluding shell

3. Crop N fixation

Crop N fixation associated with non-alfalfa and crop-pasture was estimated based on the size of harvested acreage multiplied by average values of N fixed per unit area taken from various literature sources for non-alfalfa hay (11,600 kg-N km^{-2} yr^{-1}) and for crop pasture (1,500 kg-N km^{-2} yr^{-1}) (*27, 31, 32*). We calculated N fixation by soybean and alfalfa as the product of estimates of total plant N production for these crops and the percentage of this N that can be attributed to fixation (*33*). For each watershed, the corresponding proportion of total legume N derived from N fixation was determined from tabulated values (*33*) and the estimates of average soil N mineralization (kg-N km^{-2} yr^{-1}) calculated by following the method put forth by two studies (*34, 35*) (**Table M6**).

Table M6. Estimates of average soil N mineralization and the corresponding proportion of plant N from N fixation by soybean and alfalfa hay for the 18 Lake Michigan watersheds

ID	Catchment	Soil N mineralization (kg-N km^{-2} yr^{-1})	Proportion of plant N from fixation		Crop N fixation rate	
			Soybean (%)	Alfalfa (%)	Soybean (kg-N km^{-2} yr^{-1})	Alfalfa (kg-N km^{-2} yr^{-1})
1	Root	6,861	0.73	0.83	10,947	20,860
2	Milwaukee	6,726	0.74	0.84	10,800	20,685
3	Sheboygan	7,343	0.71	0.81	10,511	19,574
4	Fox	6,771	0.74	0.84	10,882	18,799
5	Oconto	5,471	0.75	0.80	10,775	16,844
6	Peshtigo	7,007	0.72	0.82	10,595	15,265
7	Menominee	7,926	0.68	0.78	8,232	12,847
8	Ford	13,688	0.53	0.73	4,031	11,507
9	Escanaba	9,608	0.59	0.69	4,427	10,894
10	Manistique	12,422	0.57	0.77	3,773	12,168
11	Manistee	3,935	0.81	0.84	9,292	11,793
12	Pere Marquette	4,092	0.80	0.84	8,819	15,075
13	Muskegon	4,955	0.77	0.82	9,999	13,569
14	Grand	6,547	0.75	0.85	11,027	19,516
15	Kalamazoo	4,843	0.78	0.82	10,835	18,530
16	St. Joseph	4,922	0.77	0.82	11,563	18,635
17	Trail Creek	3,677	0.82	0.85	10,781	18,191
18	Burns Ditch	8,430	0.65	0.75	13,159	21,641

4. Net atmospheric deposition

Net atmospheric deposition of NO_Y, NH_X, and organic nitrogen were each estimated separately. Annual precipitation-weighted mean wet deposition of NH_4^+ and NO_3^- and dry deposition of particulate ammonium (NH_4^+), gaseous nitric acid (HNO_3), and particulate nitrate (NO_3^-) were obtained for all sites in five states for the years 1980-2004 from NADP/NTN (*7*) and for the years 1989 to 2004 from CASTNET (*8*), respectively. During the period 1980-1988, dry deposition of NH_4^+, HNO_3, and NO_3^- was estimated to be on average 14% and 51% of wet deposition based on estimates of dry and wet deposition from 11 CASTNET sites where both dry and wet deposition were monitored during 1989 through 2004. Since atmospheric organic nitrogen (AON) can be a substantial input of N , the amounts of dust AON and organic nitrate as new inputs were estimated to be one-half of the median value of 20 kg-N km^{-2} yr^{-1} from AON dust deposition and one-half of 110 kg-N km^{-2} yr^{-1} from the TM_3 model (*36*). Ammonia deposition as a component of the net atmospheric NH_X input term was adjusted by assuming that

75% of NH_x emissions are re-deposited locally and the remaining 25% are transported outside the area (*27*).

To estimate NH_X emissions from animal manure, manure N first was calculated by multiplying the estimates of average annual livestock populations by N excretion rate, and the resultant values were then multiplied by emission factors for eighteen individual livestock categories (See **Table M3**). The parameters used for manure production and emission for each livestock class were derived from two references (*2, 25*), and the EPA emission inventory report (*37*). Volatilization losses from fertilizer were calculated as a percentage of fertilizer application in each watershed: 15% for urea, 2% for ammonium nitrate, 8% for nitrogen solution, 1.0 % for anhydrous ammonia, and 4.4% for other combined fertilizers (*38, 39*). In this study, N lost via volatilization from crops was also estimated using crop acreage data from the USDA Census of Agriculture, assuming volatilization rates to be 6,000 kg-N km^{-2} yr^{-1} for corn, 4,500 kg-N km^{-2} yr^{-1} for soybean, and 3,500 kg-N km^{-2} yr^{-1} for wheat (*32*). Further details of NANI estimation are given in (*35*).

PANEL DATA REGRESSION METHODS

A panel data set includes observations on multiple entities, where each entity is observed at two or more points in time. Because panel data are typically larger than cross-sectional or time series data sets, and explanatory variables vary over two dimensions (space and time) rather than one, the estimators of the regression based on panel data are quite often more accurate than from other cross-sectional or times series regression (*40*). The estimates of coefficients derived from ordinary least square (OLS) regression may be subject to omitted variable bias. Here, omitted variable bias represents a problem that arises if a variable that is correlated with the included variables is excluded from the model. This problem can result from incomplete model specification or because omitted variables are un-measurable or unknown. They can be either of time-invariant (that vary across spatial units but do not vary over time) or spatial unit-invariant (e.g. that vary by years but do not vary across space). With panel data, it is possible to control some types of omitted variables even without observing them, by observing changes in the dependent variable over time or over space. The four main types of panel data analytic models include 1) constant coefficient, 2) fixed effects, 3) random effects, and 4) random coefficient. The constant coefficient model pools all data and runs an OLS regression model, because the coefficients of both intercepts and slopes are constant, which means there are neither significant spatial units nor significant temporal effects. The fixed effects model, called least square dummy variable model, has constant slopes but intercepts that differ according to the spatial unit or time. For example, hypothetically, consider that TN export from a river Y is determined linearly by watershed N input X and we have observations on 18 watersheds in each of five time period (**Figure M1**).

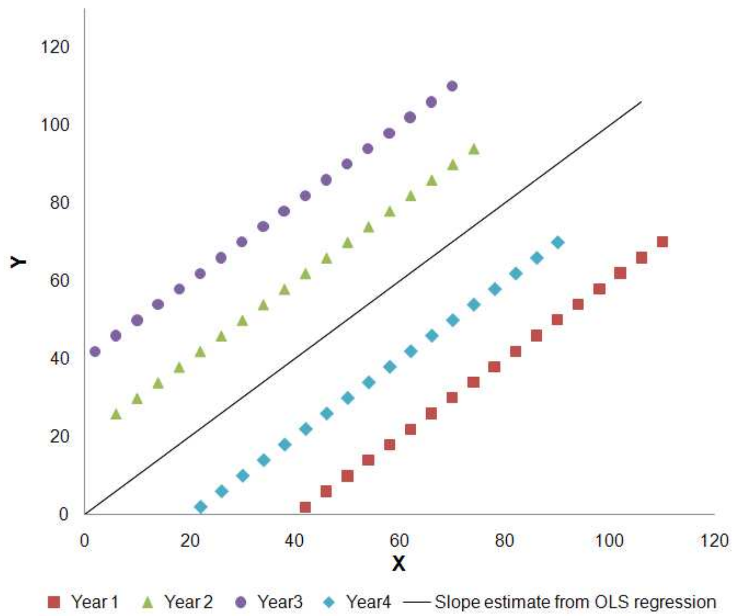


Figure M1. Panel data showing four observations on each of four years (Adapted from (*41*))

This figure shows that although each year in this example has the same slope, the five years all have different intercepts, indicating that the unmeasured variables that determine Y result in a different intercept for each year. The fixed effects model deals with the presence of a year-specific intercept term by using a dummy for each year and running OLS with all dummy variables to guard against omitted variable bias. However, this may require too many dummy variables, reducing the number of degrees of freedom and thus statistical power. This can be avoided, however, by subtracting the average values of NANI within a year (i.e., mean of values of NANI for 18 different watersheds for a given year) from the individual watershed values of NANI. An ordinary least squares regression (OLS) in then run using the transformed data.

The alternative approach, the "random effects" model, allows for different intercepts which are interpreted as random variables and treated as a part of the error term. The random effect model uses the variance-covariance matrix of the errors with a non-spherical pattern (i.e. all off-diagonal elements are not zero) and transforms data to have a spherical (i.e. all off-diagonal elements are zero) variance-covariance matrix of the errors. An OLS is again run using the transformed data. Although this model substantially reduces the number of parameters that must be estimated, this year-specific error term must be uncorrelated with the errors of the explanatory variables. Finally, the random coefficient panel data model can be applied to the case of heterogeneity of slopes. Neither the fixed effect model (varying intercept) not the random effect model (error components) allows for an interaction of individual specific and/or time varying differences with the included explanatory variables, x. However, this third model allows both random intercept and slope to vary around common means. The random coefficients can be considered outcomes of a common mean plus an error term, representing a mean deviation for each individual or year.

PERFORMANCE OF THE PANEL REGRESSION MODEL

The best regression model following the panel data approach, using both linear and exponential relationships between NANI and N export, found that the exponential equation had higher precision than the linear regression based on an error analysis (Figure M2) and R^2 comparison (exponential: 0.87, linear: 0.75).

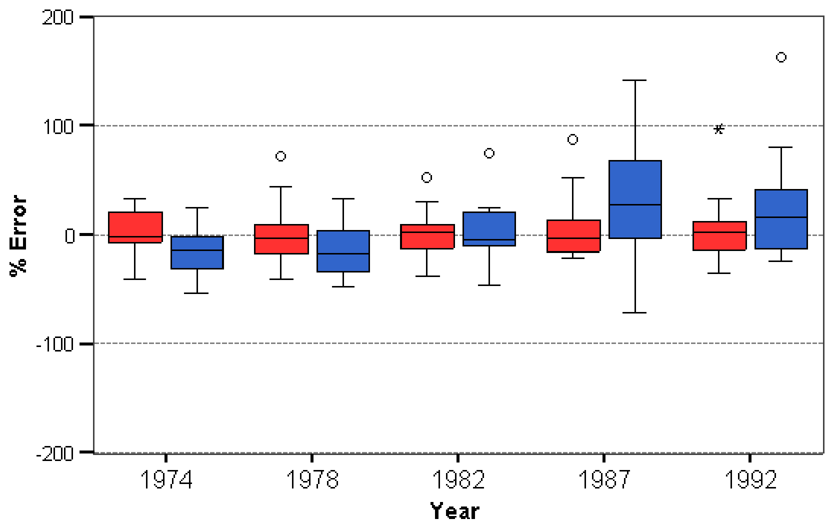


Figure M2. Boxplots showing how the magnitude and distribution of prediction errors generated from the panel regression model using exponential (red boxes) and linear (blue boxes) relationship between NANI and river N export (circles and asterisks represent outliers and extremes, respectively).

The greatest variability and bias in prediction errors associated with our model were evident when the model was used to predict historical change in riverine TN exports for each watershed, rather than spatial distribution of riverine TN exports across watersheds for each year. In other words, this model more successfully accounts for spatial variation in riverine TN exports across years than for temporal variation in riverine TN exports across watersheds. In addition, the highly urbanized watershed with the highest population density (the Root watershed) had a negative median error as well as a negative value of IQR, suggesting that the model has a tendency to under-predict riverine TN exports for urbanized watersheds. This could be the result of underestimation of N inputs such as hotspots of organic N deposition, because NADP/NTN networks are not located in urban areas; and also failure to include nitrate associated with roadways and construction activities that are common in urban areas (*42, 43*). However, if N inputs are adequately estimated, then the proportionally higher export of N by urban streams may be attributable to lower rates of in-stream denitrification as a consequence of their flashier hydrology and altered geomorphic structure, which would not be captured by our model (*44*).

ERROR ANALYSIS

The error analysis put forth by Alexander (2002) employs box and whisker plots of summary statistics, including the median, interquartile range (difference between the 75th and 25th percentiles), and minimum and maximum values for the prediction errors for the 18 watersheds for each of the five years. Each error term was computed as the difference between the predicted and measured river TN exports, expressed as a percentage of the measured exports.

FORECASTING WATERSHED N INPUTs AND RIVER EXPORT TO 2020

1. Scenario1 (Status-quo)

We assumed that all components of NANI such as fertilizer, crop N fixation, net atmospheric N deposition, and net trade of N in feed for livestock remain constant in 2020 except for net trade of N in food for humans. This term was updated to reflect future population change and the corresponding human N consumption in 2020.

2. Scenario 2 (Organic-farming)

To estimate how NANI might change between baseline (1992-2002 averages) and 2020, we assumed that the composition of leguminous plants (e.g. soybean, hay, cropland used only for pasture or grazing) will be as observed in the western region of the Lake Michigan watersheds as shown in **Table M7**. However, total harvested area of leguminous plants and non-leguminous crops for each watershed will remain constant. Based on this assumption, future N fixation for the watersheds of the eastern and southern regions of the Lake Michigan basin were estimated by multiplying the areas of legumes harvested in the watersheds of the eastern and southern Lake Michigan Basin by the combined rate of crop N fixation per unit of the harvested area of the legumes for the western region of the Lake Michigan basin.

Table M7

	Composition of major leguminous plants					Total area of legumes harvested
	soybean	Hay		Pasture		
		Alfalfa	Non-alfalfa	Cropland	Non-cropland	
Western region	7%	37%	27%	14%	16%	3,175 km^2
Eastern region	57%	18%	4%	9%	`12%	6,474 km^2

Similarly, to estimate future N inputs of N fertilizer, net trade of N in feed and food, and N volatilization from agricultural sources, the rates of fertilizer application, crop N production for food and feed, animal N consumption, and animal N manure production per unit area of agricultural land for the western region of the Lake Michigan Basin were multiplied by the area of agricultural land for the watershed within the eastern and southern Lake Michigan basin.

3. Scenario 3 (Expanded corn-production for bio-ethanol production expansion)

According to the USDA 2017 projection for expanded corn-based ethanol production (*45*), we assume that corn and soybean production increases by 54% and 41%; however other grains such as sorghum, barley, oats, and wheat decrease to 60%, 54%, 64% and10 % compared with their baselines, respectively. The amount of future fertilizer application was adjusted according to changes in crop production by multiplying the projected acreages of corn, soybean, and wheat by the rates of fertilizer application for each crop (corn: 136 lbs/treated acre, soybean: 25 lbs/treated acre, wheat: 68 lb/treated acre) obtained from the 2006 USDA AREI report (*46*). In addition, we assume livestock population will be adjusted in response to high grain and soybean meal prices due to the expansion of corn-based ethanol production and the extra supply of distiller grains, a co-product of ethanol production that can be used in livestock rations. For this study, beef cows, other cattle, hogs, broilers, turkeys and egg production are assumed to increase by 6%, 1%, 21%, 40%, 5% and 11%; however, milk cow production is expected to decrease by 8% from its base line based on USDA projection. After developing the projected crop, animal, and fertilizer use data for 2020, our automated macro model of NANI budget coded by Visual Basic application is run to estimate future NANIs for the 18 Lake Michigan watersheds.

FUTURE CLIMATE SCENARIOS

A number of studies project increases in precipitation in the Great Lakes region over the next 20 to 50 years (*47*). Precipitation levels over four of the five (all but Superior) Great Lakes have shown statistically significant increases from 1930-2000, and if trends continue increases may be as great as 20% (*48*). River discharge may show modest increases or not change greatly, because increased precipitation is expected to be offset by increased evapotranspiration due to global warming. The frequency of heavy rainfall events measured over 24-hour and 7-day periods is projected to more than double relative to the 1900-2000 average by 2100, and increases in intensity may also occur (*49*). We reviewed available information that makes a plausible case for increased precipitation and discharge, and we selected increases of 5% and 10% because they are reasonably modest, within the historical range, and similar to Howarth's (2006) future discharge values (*50*).

Table S1. Watershed characteristics and land use statistics for the 18 watersheds of the Lake Michigan Basin

ID	Watershed	USGS station	Area (km^2)	Mean temp. (°C)[1]	Population Density [2] (capita km^{-2})	Land use[3] (%)			
						Agric	Forest	Urban	Wetland
1	Root	4087242	510	8.8	397	76.7	3.1	19.0	0.1
2	Milwaukee	4087010	1818	8.0	201	73.9	7.9	12.2	4.6
3	Sheboygan	4086000	1106	8.1	31	82.0	7.2	2.5	7.0
4	Fox	4085059	15825	7.1	32	51.1	27.2	2.4	13.3
5	Oconto	4071775	2543	6.1	10	27.5	52.1	0.7	17.2
6	Peshtigo	4069500	2797	5.8	9	20.7	54.7	0.9	21.7
7	Menominee	4067651	10541	5.0	7	7.1	73.1	0.7	16.3
8	Ford	4059500	1165	5.2	3	7.1	53.5	0.2	39.0
9	Escanaba	4059000	2253	5.0	9	5.4	66.7	1.1	23.6
10	Manistique	4049500	883	5.7	3	5.0	49.5	0.3	40.2
11	Manistee	4126520	4343	6.7	8	18.3	73.1	1.0	5.9
12	Pere Marquette	4122500	1764	7.3	8	17.6	71.2	0.7	8.1
13	Muskegon	4122150	6941	6.9	27	33.6	47.7	2.8	11.3
14	Grand	4120250	14292	8.6	85	75.4	13.9	5.5	3.7
15	Kalamazoo	4108670	5164	8.8	83	75.1	12.6	6.1	4.2
16	St. Joseph	4102533	12095	9.4	68	80.4	9.3	5.5	2.4
17	Trail Creek	4095380	153	10.0	237	50.0	27.7	19.6	0.5
18	Burns Ditch	4095090	857	10.1	286	63.7	13.3	20.0	1.1

[1] Source: PRISM historical climate GIS data set (*51*) and values represent averages of estimates for five census years from 1974 to 1992;[2] averaged values over five census years from 1974 and 1992; [3] averaged values from the GIRAS LULC (for mid 1970s- early1980s) (*52*) and the 1992 NLCD land use data (*53*)

Supporting Information Table S2. Comparison of the performances of the simple linear regressions using NANI, individual N inputs, climatic variables and population density to account for spatial variation in riverine TN exports across the 18 Lake Michigan watersheds for each of five different years

Regressors	R^2 value				
	1974	1978	1982	1987	1992
NANI	**0.91**	**0.78**	**0.81**	**0.69**	**0.81**
Fertilizer N	**0.51**	**0.38**	**0.47**	**0.61**	**0.66**
Fixation N	**0.32**	**0.44**	**0.46**	**0.31**	**0.74**
Net import of N in food	**0.44**	**0.42**	**0.43**	**0.49**	**0.25**
Net import of N in feed	**0.39**	**0.46**	**0.49**	**0.63**	**0.67**
Net atmospheric N deposition	**0.46**	**0.46**	**0.38**	**0.13**	**0.55**
Annual precipitation	0.13	**0.36**	0.07	**0.72**	**0.30**
Annual water discharge	0.07	0.00	0.07	0.12	0.00
Population	**0.72**	**0.75**	**0.79**	**0.66**	**0.44**

* Bolded numbers are significant ($p<0.05$)

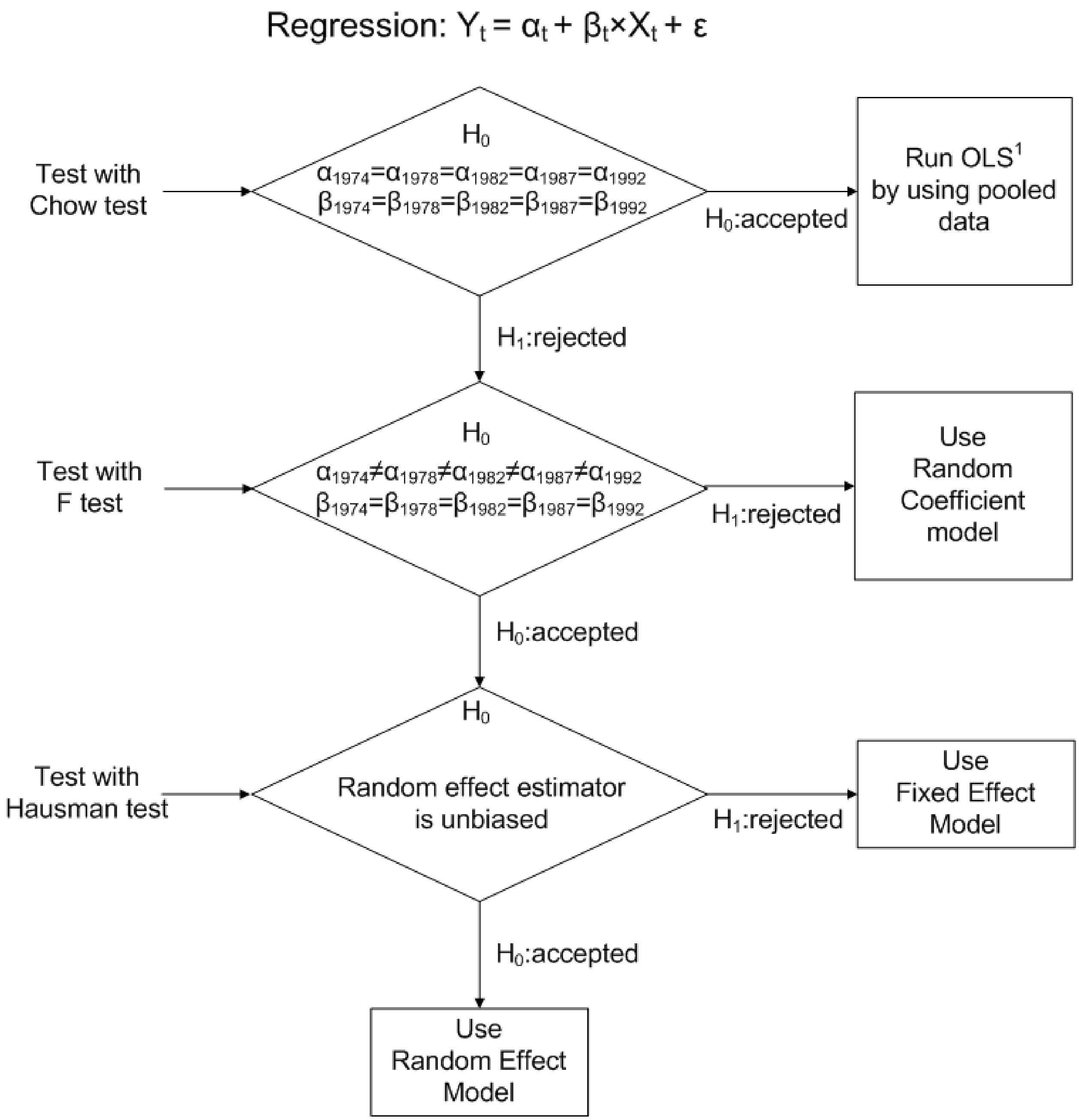
Supporting Information Table S3. Comparison of the performances of the simple linear regressions using NANI and climatic variables to account for temporal variation in riverine TN exports across the five census years from 1974 to 1992.

ID	Name	Regressors		
		NANI (kg-N km^{-2} yr^{-1})	water discharge (mm/yr)	Precipitation (mm/yr)
1	Root	0.35	<u>0.65</u>	0.16
2	Milwaukee	0.29	<u>0.75</u>	0.29
3	Sheboygan	0.07	**0.90**	0.54
4	Fox	<u>0.68</u>	**0.85**	0.60
5	Oconto	0.01	0.44	0.37
6	Peshtigo	0.01	<u>0.76</u>	0.56
7	Menominee	0.01	<u>0.68</u>	**0.95**
8	Ford	0.01	<u>0.64</u>	**0.85**
9	Escanaba	0.16	**0.76**	**0.93**
10	Manistique	0.08	**0.95**	**0.88**
11	Manistee	**0.77**	0.05	0.00
12	Pere Marquette	0.45	0.17	0.10
13	Muskegon	0.03	0.53	0.18
14	Grand	0.01	<u>0.74</u>	<u>0.74</u>
15	Kalamazoo	0.34	0.35	0.53
16	St. Joseph	0.01	**0.77**	0.20
17	Trail Creek	<u>0.70</u>	0.10	0.25
18	Burns Ditch	0.12	0.31	0.32

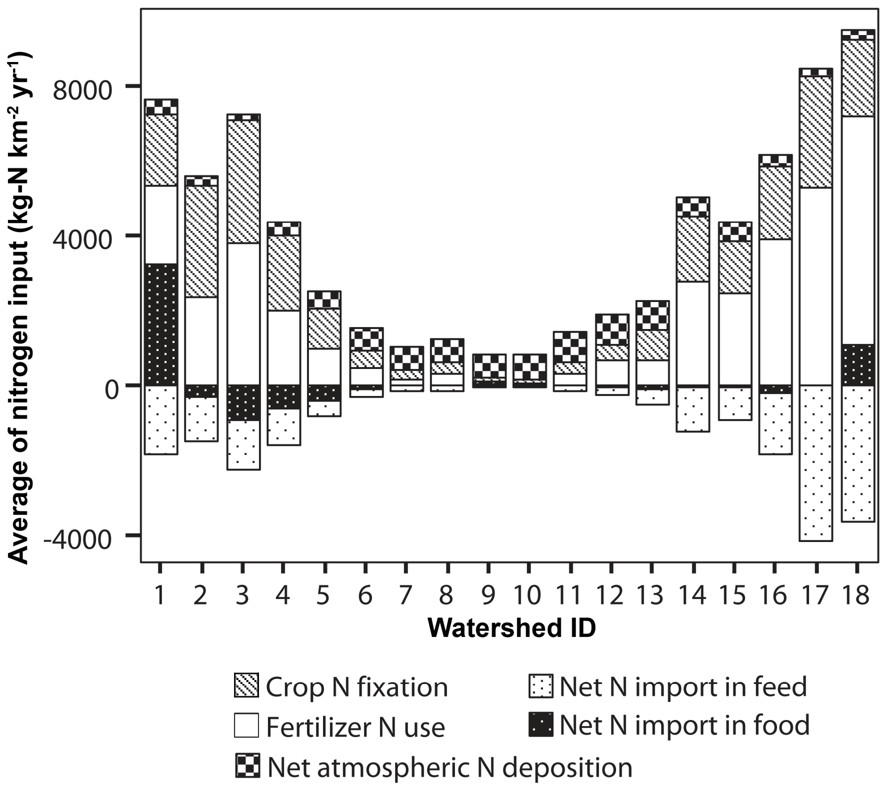
* Bolded numbers are significant ($p<0.05$) ; underlined numbers indicate $p <0.1$.

Supporting information Table S4. Summary of statistics for evaluating the accuracy of the prediction of riverine TN exports for the random effect panel data regression model using NANI and discharge across years and watersheds. Prediction errors are computed as the difference between the predicted and measured values of riverine TN exports expressed as a percentage of the measured export

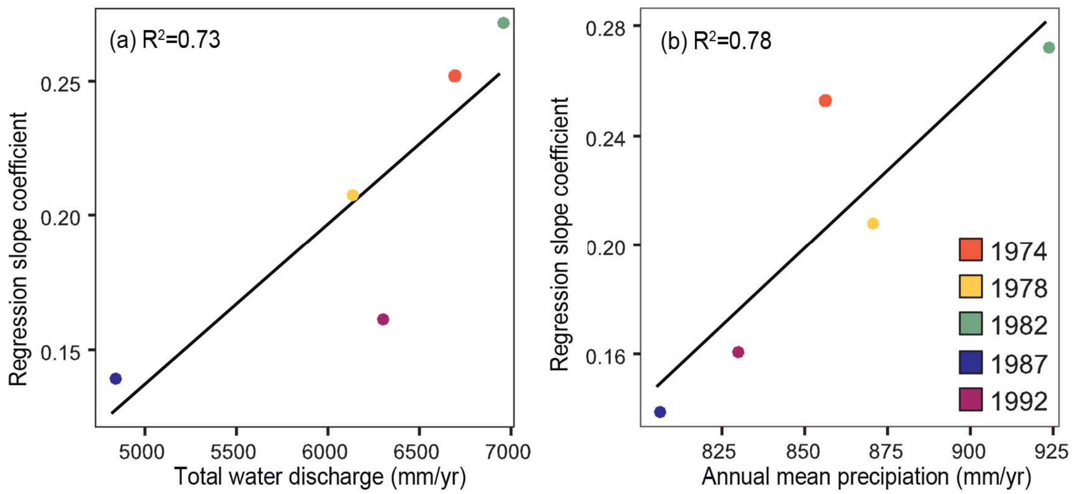
Year	Prediction error (%)					
	Median	IQR	Min.	25th percentile	75th percentile	Max.
1974	-1.70	29.2	-41.6	-7.7	21.5	33.3
1978	-3.35	28.8	-40.4	-17.7	11.1	72.3
1982	2.05	23.6	-37.6	-12.9	10.7	52.8
1987	-3.15	30.9	-21.9	-15.8	15.1	87.1
1992	1.70	29.9	-35.4	-14.4	15.5	96.8
Watershed	Median	IQR	Min.	25th percentile	75th percentile	Max.
Root	-15.5	33.4	-37.6	-27.9	-4.3	-3.3
Milwaukee	-0.4	39.1	-19.9	-12.1	19.2	35.0
Sheboygan	-12.5	92.2	-31.8	-24.5	60.3	87.1
Fox	1.5	29.4	-14.1	-8.4	15.3	18.8
Oconto	-31.7	71.9	-41.6	-41.0	30.3	52.8
Peshtigo	9.0	38.0	-24.4	-9.2	13.6	17.6
Menominee	2.2	39.1	-14.1	-13.4	25.0	33.3
Ford	7.9	40.0	-11.3	-2.0	28.7	32.2
Escanaba	0.5	33.7	-16.6	-12.3	17.1	27.6
Manistique	-12.7	16.2	-21.9	-18.5	-5.7	-0.1
Manistee	29.8	39.0	9.8	16.8	48.7	52.8
Pere Marquette	-6.4	74.3	-21.2	-19.4	53.1	96.8
Muskegon	5.0	38.2	-17.6	-8.2	20.6	26.8
Grand	-19.2	27.8	-37.5	-36.4	-9.8	-0.6
Kalamazoo	-6.0	21.0	-12.2	-9.3	8.7	13.9
St. Joseph	-6.9	30.8	-24.0	-17.4	6.8	11.2
Trail Creek	9.2	31.1	-5.7	-3.4	25.4	29.9
Burns Ditch	12.3	58.4	-9.9	-9.4	48.4	72.3



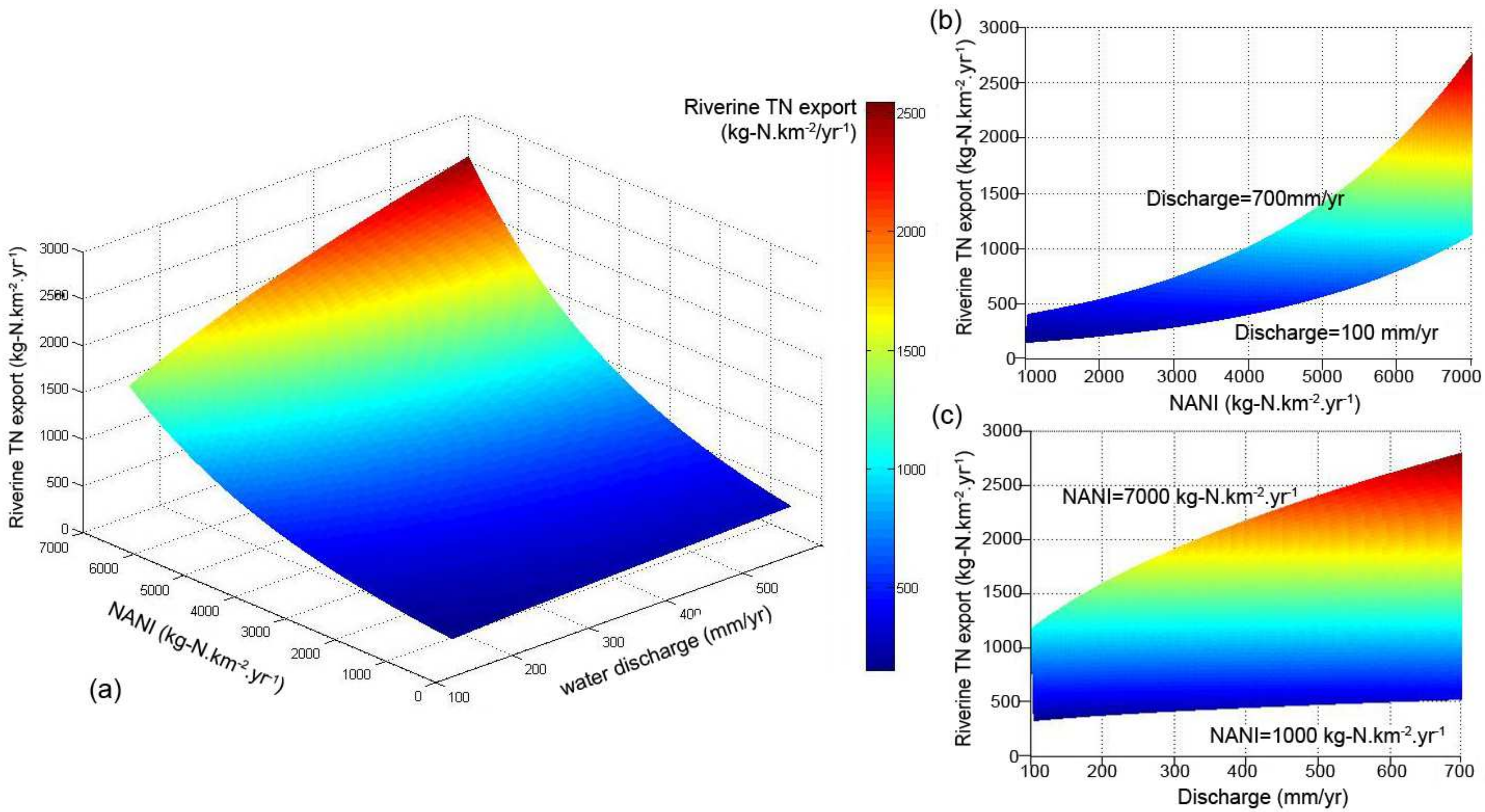
Supporting information Figure S1. Developing a panel data model by testing a hierarchical sequence of hypotheses. [1]OLS: ordinary least regression



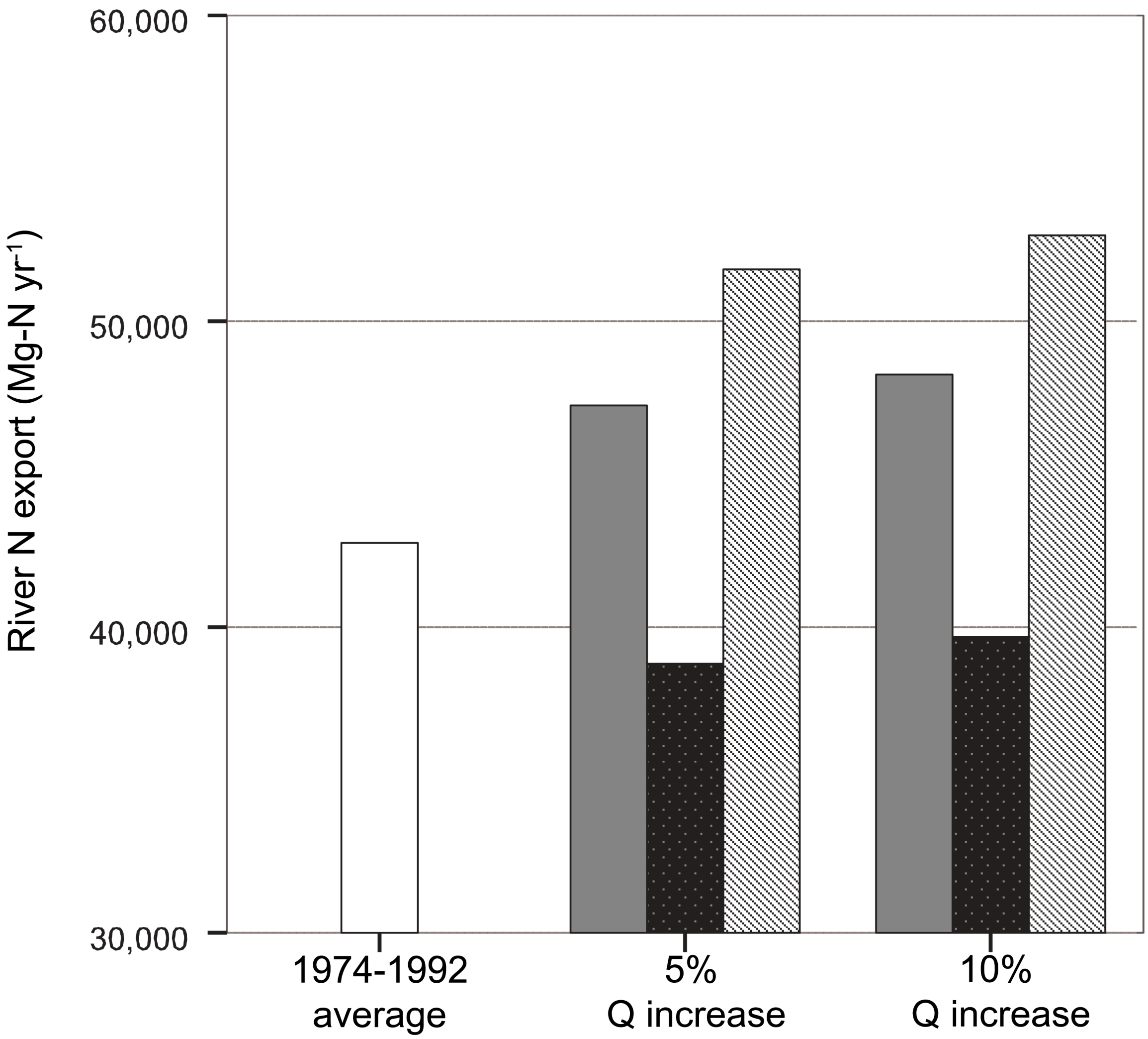
Supporting information Figure S2. Relative importance of individual N inputs averaged over five agricultural census years (1974, 1978, 1982, 1987, and 1992) for each of 18 Lake Michigan watersheds. See **Supporting information Table S1** for watershed names.



Supporting information Figure S3. The slope coefficients of the linear regressions of river TN exports vs. watershed N inputs of the 18 Lake Michigan watersheds for each of five years are positively correlated with (a) annual total water discharge and (b) annual mean precipitation of the 18 Lake Michigan watersheds for the corresponding years.



Supporting Information Figure S4. (a) A three-dimensional contour map illustrating how estimated TN export varies as a non-linear function of discharge and NANI based on equation 5, (b) estimated change in riverine TN exports across the values of NANI ranging from 1,000 to 7,000 kg-N km^{-2} yr^{-1}, for different amounts of discharge ranging from 100 to 700 mm/yr, and (c) estimated change in riverine TN export as a function of discharge (100 to 700 mm/yr) at different magnitudes of NANI from 1000 to 7000 kg-N km^{-2} yr^{-1}.



Supporting Information Figure S5. Forecasting of loads (Mg-N/yr) of river N exports for the combined 18 Lake Michigan watersheds, based on three scenarios of future N inputs (Scenario 1: status-quo (solid gray bar); Scenario 2: organic (black with white dots); Scenario 3: expanded corn-based ethanol production (diagonal lines) and two assumptions of future climate change (water discharge increases of 5% and 10%), in comparison with measured river TN export (open bar), averaged over five census years (1974- 1992).

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