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Uneven rise in N inputs to the Lake Michigan Basin over the 20th century corresponds to agricultural and societal transitions

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Abstract By constructing nitrogen (N) budgets from 1880 to 2002 for watersheds that have undergone urbanization, intensive agricultural specialization or experienced minimal change, we document an uneven timeline of increase in anthropogenic N inputs. N loading to the watersheds of the Lake Michigan Basin grew six-fold from 1880 to 2002, peaking in 1987. Human activities influenced N inputs as early as 1880, and the magnitude and timing of increase differed markedly across regions in accord with population growth, land use, and type of agriculture. The greatest increase occurred from 1950 to 1980, corresponding with rapidly accelerating use of artificial fertilizers, but increases in atmospheric deposition and shifting patterns in crop and livestock production also affected trends. Net anthropogenic N inputs have changed little since about 1980, showing a modest decline due to a leveling out of fertilizer use and greater export of animal feed and products. Using a model that predicts riverine N export from watershed N loadings and river discharge, we found that river TN fluxes from all tributaries increased approximately threefold from 1900 to 2000 but have stabilized or declined over the past two decades, consistent with national surveys that show near-constant or declining riverine TN concentrations. For the LMB, the past two decades has been a period of relative stasis in N inputs to its terrestrial systems and N export from watersheds. This retrospective analysis also points to the challenge of forecasting future trends in N budget terms, which can both increase and decline in response to policy and societal transitions.

Keywords Nitrogen · NANI · N export · River · Catchment · Nutrient budget · Nutrient loading · Lake Michigan

Abbreviations

AON	Atmospheric organic nitrogen
CASTNET	Clean air status and trends network
LMB	Lake Michigan Basin
NANI	Net anthropogenic nitrogen input
NADP/NTN	National atmospheric deposition program/national trends network
NOx	Nitrogen oxide

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Introduction

Global production of reactive nitrogen has increased approximately tenfold since 1860 due to widespread cultivation of N-fixing crops, combustion of fossil fuels, and extensive use of synthetic fertilizer (Galloway et al. 2003; Schlesinger 2009). Much of this increase has taken place since 1960 (Galloway and Cowling 2002). For the US, anthropogenic N inputs doubled between 1961 and 1997, with most of the increase in the 1960s and 1970s (Howarth et al. 2002). Increased use of inorganic N fertilizer accounted for the largest fraction of this increase, followed by emissions of nitrogen oxide (NO_x) from fossil fuel combustion. N-fixation, the largest single source of reactive N in1960, increased overall but declined as a fraction.

N export from watersheds is estimated to have increased two to 20-fold as a consequence of increased N loading to terrestrial systems (Howarth et al. 1996), causing degradation of water quality and contributing to the formation of oxygen depleted bottom waters in coastal systems (Carpenter et al. 1998; Rabalais et al. 2002). Watershed studies find that as much as 20–25 percent of the added N to the terrestrial system is exported from rivers to the oceans or inland basins (Howarth et al. 1996; Boyer et al. 2006), however, some lower estimates of fractional export may indicate as yet unexplained regional variation (Parfitt et al. 2006; Schaefer and Alber 2007; Han et al. 2009). Although long-term records are few, studies have shown that nutrient concentrations in streams and rivers of the coterminous United States are higher today than a century ago. Observed concentrations of total N in the Mississippi River have doubled from 1950 to the present (Turner and Rabalais 2003). Measured nitrate–N concentrations for 63 rivers of the coterminous US were three to four times higher at the close of the 20th century as compared with the beginning (Broussard and Turner 2009). Using minimally impacted US reference basins for comparison, Smith et al. (2003) estimated that current TN concentrations exceed background levels by a factor of 6.4.

However, trends in riverine nutrient concentrations over the latter part of the 20th century are less pronounced. For 250 US locations monitored over 1975–1994, statistically significant declines in TN concentrations were much more common than increases (Alexander and Smith 2006), consistent with other reports (Smith et al. 1987; 1993; Lurry and Dunn 1997). The cause of these declines is uncertain, but source reductions, nutrient processing and storage in impoundments, wastewater treatment upgrades and improvements in farmland management may all have contributed. A subsequent analysis for 1993–2003 also reported that riverine TN concentrations have stabilized or are declining in many areas of the USA (Sprague and Lorenz 2009). Since river export equals nutrient concentration × water discharge, loads also are likely to have shown little change.

Thus, while there is no question that N application to the land and export by rivers are greatly elevated over 19th century values, the detailed timeline of change over the past 100-plus years is not well known. In the present study we report net anthropogenic nitrogen inputs to the terrestrial system from 1880 to 2002 for 25 watersheds surrounding Lake Michigan that differ substantially in land use, human population, and anthropogenic N sources. This unusually detailed spatial and temporal accounting allows us to relate changes in N sources to changing human drivers, including historical changes in farm practices and other socio-economic factors in the region. In addition, using a previously published model (Han et al. 2009), we provide hind-cast estimates of river N export including probable near-baseline conditions.

Methods

Study area

The 25 watersheds cover a total area of approximately 115,318 km^2, range in size from 1,063 to 12,151 km^2, and encompass a wide variety of land use, population density, and human activities (Fig. 1, Supplementary Material, Table S1). The study area includes nearly all of the 118,000 km^2 that is generally given as the total drainage area of the Lake Michigan Basin (LMB). Land cover in the LMB was 46% agricultural, 36% forest, 11% wetland, and 4% urban for the 1970s through 1980s, although land cover varies greatly over the basin and over time. The

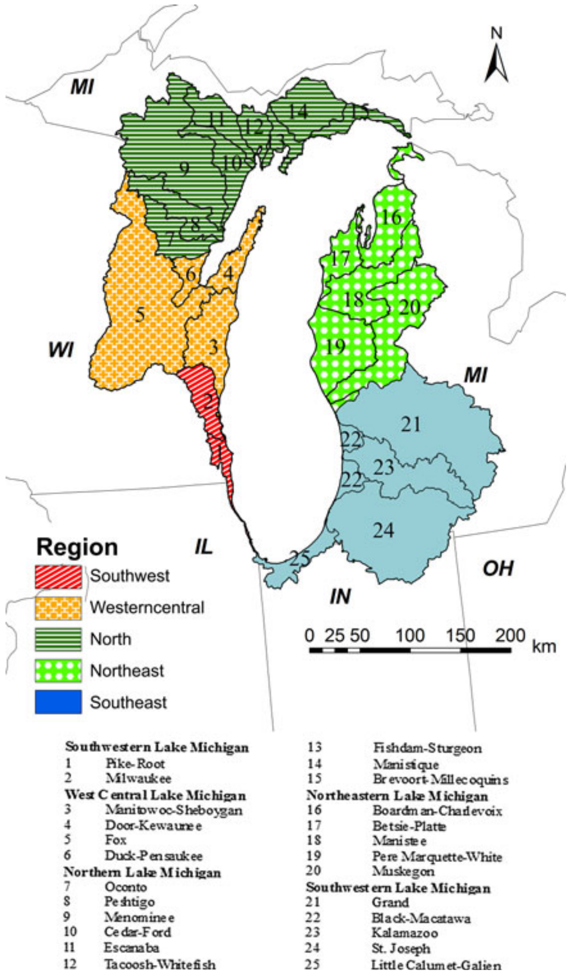

Fig. 1 The boundaries of the 25 Lake Michigan watersheds used in constructing N budgets

area-weighted average total annual precipitation is highly variable from year to year, ranging from about 610 mm/year in 1910 to 910 mm/year in 1954 (Daly and Gibson 2002). Three droughts occurred in the agricultural census years from 1880 to 2002: in 1910 (610 mm/year), 1934 (713 mm/year) and 1944 (720 mm/year).

N budget construction

We constructed N budgets for the 25 LMB watersheds for each of the 22 agricultural census years from 1880 to 2002, quantifying all known anthropogenic N inputs (fertilizer, crop fixation, atmospheric deposition, imports of N in crop, and animal products), outputs (volatilization of N from applied manure and fertilizer and crop senescence, and exports of N in food and feed) as well as the net balances between inputs and outputs, resulting in an estimate of net anthropogenic N inputs (NANI). Table S2 (Supplementary Material) provides data sources, temporal and spatial resolution, and extent of data used in this study; methods of estimation follow those described by Boyer et al. (2002) and Han and Allan (2008).

Fertilizer N

Estimates of historical N input from fertilizer prior to 1945–1991 were based on county-level fertilizer use from Alexander and Smith (1990) for 1945–1985, from Battaglin (1994) for 1986–1991, and from Ruddy et al. (2006) for 1992–2001. For years prior to 1945, the Statistical Reporting Service, Crop Reporting Board (CRB) reports nationwide fertilizer sales for 1850–1945 (USDA (U.S. Department of Agriculture) 1966). National data were disaggregated to county-level data using the county to state and state to nation ratios of expenditures for commercial fertilizer, which were obtained from the Census of Agriculture for the corresponding years.

Atmospheric N deposition and volatilization

Atmospheric deposition was estimated separately for NO_y (NO_3^- and HNO_3), NH_x (NH_4^+ and NH_3), and atmospheric organic nitrogen (AON). Annual wet deposition of NH_4^+ and NO_3^- for all stations in five states in the vicinity of LMB watersheds (IL, IN, MI, OH, and WI) for 1980–2004 and 1989–2004 were obtained from National Atmospheric Deposition Program/National Trends Network (NADP/NTN) (NADP(National Atmospheric Deposition Program/National Trends Network) 2006) and Clean Air Status and Trends Network (CASTNET) (CASTNET (Clean Air Status and Trends Network) 2006), respectively. In addition, annual estimates of dry deposition of particulate ammonium (NH_4^+), gaseous nitric acid (HNO_3), and particulate nitrate (NO_3^-) for the period 1989–2004 were retrieved from CASTNET (CASTNET (Clean Air Status and Trends Network) 2006).

Dry deposition for the years 1980–1988 was estimated by multiplying wet deposition of inorganic

N by the estimated ratio of dry to wet deposition of NH_4^+ (0.14), HNO_3 (0.51), and NO_3^- (0.51), which were calculated using measurements of dry and wet deposition from 11 stations for the period 1989–2004. For the years prior to 1980 for which data were not available, total atmospheric deposition of NO_y and NH_x was extrapolated using historical trends in national NO_x emission (USEPA 2000; 2003) and ammonia (NH_3) emission (Van Aardenne et al. 2001) based on the relationship between emission and atmospheric deposition of NO_3 and NH_4 for 1981–2004 (David and Gentry 2000).

Atmospheric organic N (AON) deposition includes reduced AON, biological and particulate AON, and organic nitrates. However, only new inputs of dust AON and organic nitrates were included in the budget calculations, because reduced AON has a short life time, making it unlikely to be transported into or exported from the LMB, and biological AON is likely to be negligible. We estimated new inputs of dust AON and organic nitrates to be 10 and 55 kg-N/km^2/year from the TM_3 model (Neff et al. 2002), respectively. Similar to the historical inorganic N extrapolation, historical organic nitrates were extrapolated using the historical trends in national NO_x emission as a surrogate and dust AON was assumed to be constant over the study period (Han and Allan 2008).

The estimated export of atmospheric N through volatilization of ammonia from applied fertilizer, animal manure, and crop senescence was calculated based on the assumption that 75% of NH_x emissions are re-deposited locally and the remaining 25% are exported from the study watershed, following Boyer et al. (2002). Following methods in Han and Allan (2008), we first estimated average annual animal population numbers by animal group, then calculated the amount of N excreted from each livestock type via feces and urine, and combined these with the rates of volatilization of ammonia according to individual livestock type and manure management practices to calculate the total amount of N emitted from manure, by livestock type.

Biological N-fixation

We estimated biological N-fixation by legume crops including soybeans, alfalfa, other non-alfalfa hay, and crop pasture. Crop N-fixation in association with non-alfalfa and crop pasture was estimated from size of harvested acreage and average values of N fixed per unit area for non-alfalfa hay (11,600 kg-N/km^2/year) and for crop pasture (1,500 kg-N/km^2/year) (Boyer et al. 2002; Burkart and James 1999). The amount of N fixed by soybean and alfalfa, which are principal legumes within the LMB, was estimated following the yield-based approach of Meisinger and Randall (1991). For soybeans and alfalfa these estimates were adjusted for the amount of N mineralized from soil organic matter following Goolsby et al. (1999) and Meisinger and Randall (1991), and converted from grain harvested to total plant N derived from N_2 fixation using the ratio of grain yield to total plant mass.

Trade of N in food and feed

Net import of N in food and feed is estimated by subtracting human and animal consumption from crop and animal production, based on the assumption that human and animal food and feed requirements are met by local agricultural production and by imports. Historical human consumption of N in food over the 1880–2002 time interval was estimated using yearly estimates of population from the U.S. Bureau of the Census, multiplied by per capita consumption of N in food for a given year. Values for per capita N consumption were obtained from estimates of annual per capita protein consumption at the national level from the USDA (U.S. Department of Agriculture) (2006), multiplied by 0.16 (N percentage of protein). Annual estimates of county-level population for the years 1970–2004 were obtained from the U.S. Bureau of the Census (USBC 1982; 1990; 1992; 1995; 2003). For years prior to 1970, the human population was interpolated from decadal censuses to obtain estimates corresponding to agricultural census years. Following Han and Allan (2008), N consumption by livestock was calculated from estimates of average annual livestock populations and the N requirement per animal for each livestock type. The amount of N harvested with crops and animal products to be consumed by humans and animals in food and feed was calculated by multiplying crop production and livestock weights by the nitrogen content of each harvested crop type and animal product.

Historical estimates of riverine N export

To generate historical estimates of riverine TN fluxes from all tributaries draining to Lake Michigan, we applied a previously developed model (Han et al. 2009) that incorporated spatial and temporal variation in the relationship between river TN export (RNEXPORT in kg-N/km^2/year), NANI (NANI in kg-N/km^2/year) and annual water yield (Q in mm/year) for 18 Lake Michigan watersheds where adequate TN export data was available ($i = 1$–18) for five agricultural census years ($t = 1$–5) between 1974 and 1992:

$$\mathrm{RNEXPORT}_{it} = 11.65 \times Q_{it}^{0.46} \times \exp\left(3.49 \times 10^{-4} \times \mathrm{NANI}_{it}\right) \quad (1)$$

To reconstruct historical water yields, we first collected all relevant riverine discharge estimates from USGS gauging stations of major tributaries to Lake Michigan (Supplementary Material, Table S3). For years without measurements we extrapolated historical annual water discharge using two methods: (1) a linear regression relating annual water yield at the location with missing data to some longer and more complete dataset from neighboring watersheds and (2) a linear regression relating annual water yield to annual precipitation (mm/year) data available for all 25 Lake Michigan watersheds from 1900 to 2002 from PRISM (Daly and Gibson 2002). We selected and used the regression model with the highest R^2 and smallest error (Supplementary Material, Table S3 for selected regressions used for historical discharge extrapolation). Hindcasts of river TN exports for the 20th century (from 1900 to 2002, precipitation data were not available prior to 1900) followed the methods described above using annual NANI and water yield estimated at 5 year intervals. We also estimated atmospheric N deposition directly to Lake Michigan during the same period following methods described previously for atmospheric N deposition to watersheds.

Uncertainty analysis

To evaluate uncertainty in the estimation of NANI and prediction of river TN flux we conducted a multivariate Monte Carlo uncertainty analysis allowing all parameters and input drivers used in estimating N terms and four additional parameters (water discharge and the three regression coefficients of Eq. 1) to vary stochastically. All parameters varied independently according to a normal distribution with expectation equal to 100% and standard deviation equal to ±5% of the nominal value. A total of 100 model runs were simulated for each of the 25 watersheds for the 22 census years from 1880 to 2002 for NANI terms and for the 20 census years from 1900 and 2002 for river TN exports. We then calculated means and standard deviations within the set of 100 runs for individual modeled N inputs and outputs, NANI, and river TN exports for each census year, in each watershed.

Results

Human activities in the LMB

From 1880 to 2002 the human population within the LMB increased steadily from ~1,606,000 to ~8,874,000 persons, a 5.5-fold increase due mainly to urban growth in and around major cities (Table 1). Farming practices underwent substantial change, while area of cropland increased and then declined (Fig. 2). Average farm size grew modestly over 1880–1950, and then experienced a steady rise so that average farm size at the end of the 20th century was approximately twice that in the first half of the century. Wheat was the dominant crop in 1880, as it had been for some decades, but declining yields encouraged crop diversification (Granger and Kelly 2005). A diversity of crops were cultivated well into the early 20th century, including corn, oats, potatoes, barley, rye, vegetables, and fruits (Fig. 3). Alfalfa and corn for animal feed increased after about 1920, and greater commodity specialization is apparent after 1940 as corn, soybean and alfalfa hay came to dominate crop production and wheat and other crops contributed proportionately less. Soybean cultivation and production dramatically expanded across the entire cropland of the LMB after the 1980s (Fig. 3). By 2002, the principal crops in the LMB were corn (35% of total), soybean (22%), and alfalfa hay (13%).

Livestock populations varied over time. Dairy cattle increased steadily until about mid-century and then declined so that numbers at the end of the 20th

Table 1 Temporal changes from 1800 to 2002, for the entire Lake Michigan Basin in populations of humans and livestock

Year	Human	Beef cow	Milk cow	Hog and pig	Chicken
1880	1,606,439	23,737	338,617	710,156	2,679,700
1890	2,055,619	18,153	392,510	760,355	3,753,413
1900	2,450,523	34,832	447,004	843,735	4,753,413
1910	2,956,821	111,212	626,870	954,124	6,151,190
1920	3,118,852	32,428	678,472	822,123	6,760,365
1924	3,366,830	37,579	703,822	676,463	7,758,198
1929	3,636,596	11,943	640,327	616,509	7,018,363
1934	3,633,804	14,946	781,942	457,914	7,626,394
1939	3,834,502	22,496	818,531	475,214	6,406,953
1944	4,567,821	115,027	837,690	538,139	6,662,948
1949	4,466,315	26,565	746,421	692,456	5,662,506
1954	4,808,162	46,613	762,878	910,180	7,544,641
1959	5,261,233	52,586	669,882	1,018,416	5,841,756
1964	5,652,198	76,449	650,682	804,024	4,743,306
1969	6,100,649	100,066	525,554	774,560	4,087,521
1974	6,493,912	121,031	523,554	659,049	4,513,253
1978	6,660,229	83,973	491,773	899,219	4,816,355
1982	6,636,872	80,710	554,621	963,284	4,633,070
1987	7,304,594	63,729	517,912	1,038,806	3,618,488
1992	7,992,209	66,955	464,479	1,089,146	2,094,625
1997	8,362,118	67,910	433,306	824,422	2,142,590
2002	8,874,457	70,123	426,912	685,364	1,706,397

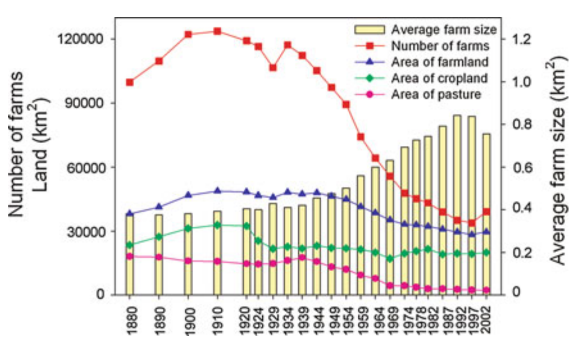

Fig. 2 Temporal changes from 1880 to 2002 for the entire Lake Michigan Basin in agricultural metrics including average farm size, number of farms, and area of farmland, cropland, and pasture

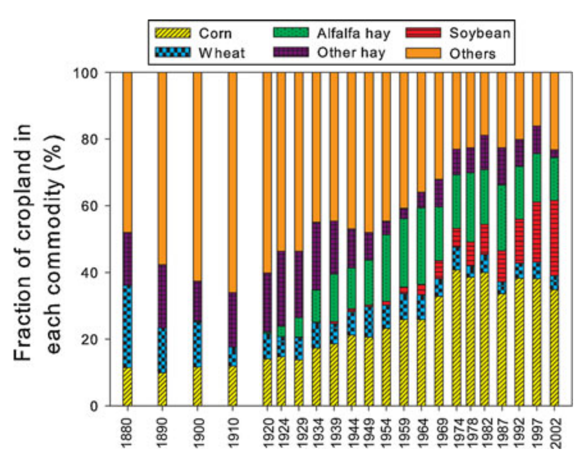

Fig. 3 Temporal changes from 1880 to 2002, for the entire Lake Michigan Basin in the fraction of cropland in major crops. Wheat, hay and a diversity of crops early in the historical record have been replaced by corn and soybean dominance in the past few decades

century were similar to those in 1880 (Table 1). By the late 1950s, specialization and consolidation into larger livestock enterprises led to a decline in the number of livestock farms and poultry and dairy cow populations. The number of hogs and pigs went through several episodes of increase and decline. The substantial decline over 1920–1940 coincided with federal loan programs that required farmers to participate in hog and corn reduction (Bowers et al. 1984).

N inputs to the Lake Michigan Basin

Fertilizer N

Fertilizer N inputs to the entire LMB increased from only 194 ± 4 (mean ± standard deviation) in 1880–204,034 ± 3,715 Mg–N/year in 2002 (Fig. 4, Supplementary Material Table S4). By 1940 fertilizer use had increased to 5,449 ± 110 Mg–N (47 ± 1 kg-N/km^2/year), but was still limited by the high price of domestic production and low availability of imported fertilizer (Baum and Clement 1958). From the 1940s to the 1980s N fertilizer use increased by a factor of 40, to 226,008 ± 4,249 Mg–N (1960 ± 37 kg-N/km^2/year) in 1982, and ranged from 486 ± 8 kg-N/km^2/year in the forested north to 3,763 ± 101 kg-N/km^2/year in the southeastern LMB (Supplementary Material, Fig. S1a). Fertilizer inputs to most watersheds leveled off or declined after 1982, particularly in the west-central and southwest of the LMB.

Biological N-fixation

N-fixation by crops in the LMB increased approximately 2.8-fold, from 61,081 ± 1,414 Mg–N/year in 1880 to 172,949 ± 4,219 Mg–N/year in 2002 (Fig. 4). Crop fixation was 93% of all positive inputs of anthropogenic N to the LMB in 1880, 56% in 1964, and 39% in 2002, as fertilizer N replaced crop fixation as the dominant input overall N-fixation in forested northern LMB watersheds increased almost five-fold over 1880–2002, from 89 ± 2 to 431 ± 11 kg-N/km^2/year, whereas the more agricultural southern region experienced high N-fixation even in 1880 at 926 ± 23 kg-N/km^2/year, and this value more than doubled by 2002–2421 ± 67 kg-N/km^2/year. Changes in N-fixation correspond with increased alfalfa cultivation (Fig. 3) and pasture for dairy farming (Table 1) after about 1920, especially in the southwest and west-central regions of the LMB, and the subsequent expansion of soybean cultivation after the 1980s across the entire basin and especially in the southeastern region of the LMB. Counter to the overall trend, however, crop N-fixation in the southwestern region of the LMB decreased over 1978–2002, from 3058 ± 169–2097 ± 99 kg-N/km^2/year (Fig. S1b), as the expansion of soybean coincided with reduced cultivation of alfalfa hay.

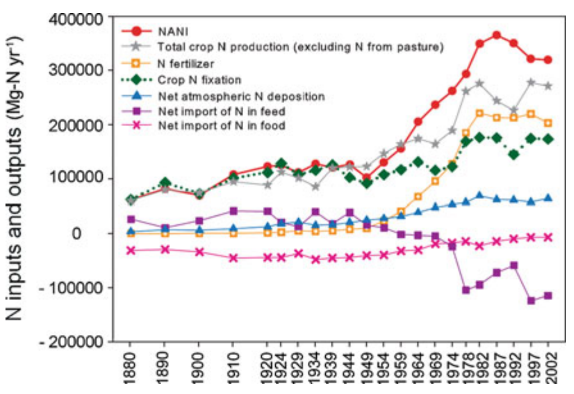

Fig. 4 Temporal changes from 1880 to 2002, for the entire Lake Michigan Basin, in individual N input and output terms. NANI is the sum of N inputs as fertilizer, net atmospheric deposition, crop N-fixation, and trade (shown here as net import) in human food and animal feed. Total crop N production (excluding N from pasture), which is not a budget term, is also shown

Atmospheric N deposition

Net atmospheric N deposition to the entire LMB increased from 4,136 ± 668 Mg–N/year in 1880 to 61,916 ± 1,205 Mg–N/year in 2002 (Fig. 4). Because other N inputs were minimal, net atmospheric N deposition was the most important source of N to the forested northern LMB in 1880 (66% of NANI), and even in 2002 contributed 48% of total N inputs to that region. Although considerable uncertainty in early estimates of atmospheric deposition must be acknowledged, modest increases in N deposition through about 1940 are expected to result from volatized ammonia transported by the prevailing westerly winds into the LMB from manure produced by high livestock populations in western WI. Net atmospheric deposition to the entire LMB land area is estimated to have risen steadily after 1880 to approximately 66,146 ± 1,400 Mg–N/year by 1982 as a result of increased emission of NO_x and NH_3, and thereafter was approximately constant over 1982–2002 at approximately 60, 600 Mg–N/year (526 kg-N/km^2/year) (Fig. 4, Supplementary Material, Table S4). Net deposition was consistently higher in the forested northeast and north relative to the agricultural southeastern and southwestern regions of the LMB (Fig. S1e), because of the greater amount of volatilization estimated for agricultural land.

Net trade of N in food and feed

Prior to 1940, N demand by humans and livestock within the LMB was well balanced by the local supply of N from crop and animal products, supplemented by relatively small amounts of N imported from outside the LMB. However, the amount of N exported in food and feed increased from 5,946 ± 4,073 Mg–N/year in 1944 to about 121,121 ± 5,584 Mg–N/year in 2002 (Fig. 4), because of large increases in crop N production, particularly in corn and soybean after the 1950s (Fig. 3), and a moderate decrease in animal consumption (Table 1), leading to a large N surplus. Shifts in the net trade of food and feed showed regional trends, not only between agriculture and urban areas, but also between regions focusing on livestock versus crop production. By 1940 pronounced differences were apparent between the dairy farming southwest region, which imported feed, and the corn-oriented southeastern region, which exported animal feed (Supplementary Material, Fig. S1d). The steady population increase in and around the cities of Milwaukee and Chicago led to increases in N imported as human food in the southwestern basin, from 56 ± 91 kg-N/km^2/year in 1924 to 2115 ± 128 kg-N/km^2/year in 2002 (Supplementary Material, Fig. S1c). In contrast, the southeastern region of the LMB experienced a comparable increase in the net export of N as animal feed, from 1415 ± 103 kg-N/km^2/year in 1978 to 2130 ± 144 kg-N/km^2/year in 2002, as soybean production expanded during this period (Supplementary Material, Fig. S1d).

Total anthropogenic N inputs

Net inputs from all sources of anthropogenic N to the LMB increased nearly sixfold from approximately 64,779 ± 2,495 Mg–N (562 ± 22 kg-N/km^2/year) in 1880 to a peak of 354,283 ± 7,209 Mg–N (3072 ± 63 kg-N/km^2/year) in 1987, and then declined to 309,834 ± 6,575 Mg–N (2684 ± 61 kg-N/km^2/year) in 2002 (Fig. 4). Between 1880 and 2002, N inputs to each of the 25 watersheds had increased by different amounts, ranging from approximately two- to 11-fold (Fig. 5). Disparities among watersheds increased notably, with greatest growth occurring in the more agricultural and settled southern regions of the LMB (Supplementary Material, Fig. S1f). Especially striking is the growth of a "NANI hotspot" in the southeastern region of LMB after about 1950, so that by 1982 N inputs to southeastern and southwestern regions of the LMB were similar. After the 1980s, however, anthropogenic N inputs appear to enter a phase of relative stasis, with declines in some areas (Fig. S1f).

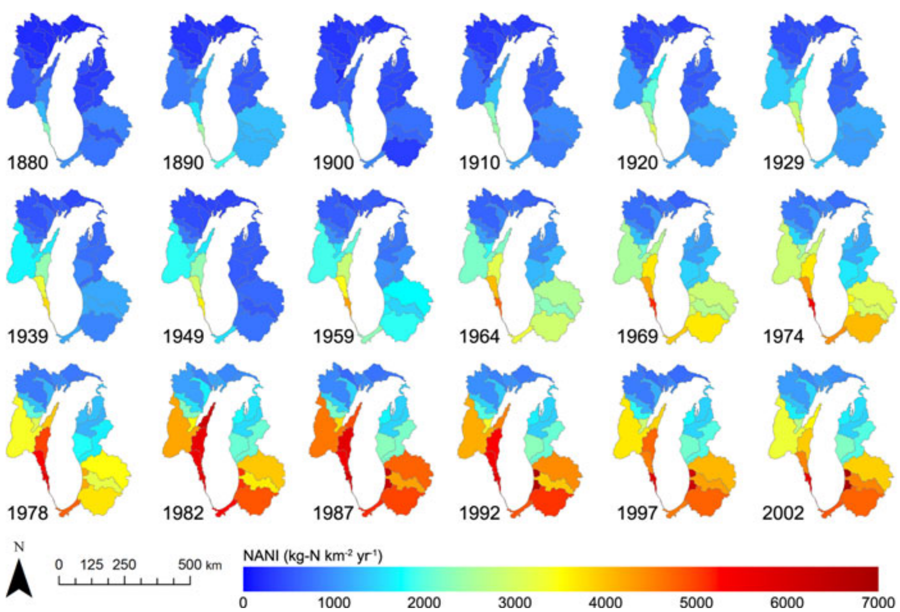

Fig. 5 Trends in anthropogenic N inputs (NANI in kg-N/km^2/year) for the 25 Lake Michigan watersheds for selected census years from 1880 to 2002

Historical changes in riverine N fluxes

Applying Eq. 1 to historical estimates of total N inputs (Fig. 4) and mean annual water discharge (Fig. S2a) for the 25 Lake Michigan watersheds from 1900 to 2002 reveals that the annual flux of TN from all major tributaries draining to Lake Michigan underwent more than a three-fold increase over the twentieth century. Estimated river export of TN increased from approximately 21,923 ± 1,161 Mg–N/year in 1900 to a peak of 66,465 ± 5,326 Mg–N/year in 1982, and then declined to 54,490 ± 3,807 Mg–N/year in 2002 (Fig. 6) in response to decreases in both N inputs and streamflow. The intensively cultivated southeastern and western regions of the LMB (53% of total drainage area) consistently contributed over 60% of TN river export to Lake Michigan prior to 1965 and as much as about 65% in subsequent years.

Because anthropogenic N inputs, primarily from agriculture, were evident as early as 1880 and we were able to estimate river discharge only after 1900, our earliest estimates of riverine TN export still reflect human influence. To approximate pre-disturbance conditions, we estimated TN yield for the LMB using the range of annual streamflow for 1900–2002 and setting anthropogenic inputs to zero in Eq. 1. Under presumed pristine conditions, N export by rivers of the LMB is estimated to be 161 kg-N/km^2/year (at the mean water yield of 302 mm/year), and ranged from 128 kg-N/km^2/year (at the lowest water yield of 184 mm/year) to 191 kg-N/km^2/year (at the highest water yield of 439 mm/year).

Atmospheric deposition of N directly to the surface of Lake Michigan increased about 4.3-fold, from 11,360 ± 1,172 Mg–N/year in 1900 to a peak of 49,406 ± 4,855 Mg–N/year in 1992, then leveling off near 48,853 ± 4,690 Mg–N/year in 2002 (Fig. 6). Relative to tributary inputs, atmospheric N deposition to the lake surface has contributed from roughly one-third to one-half of total N flux into Lake Michigan over the 20th century, increasing from 35% in 1900 to 54% in 1964, and remaining between 40 and 50% thereafter.

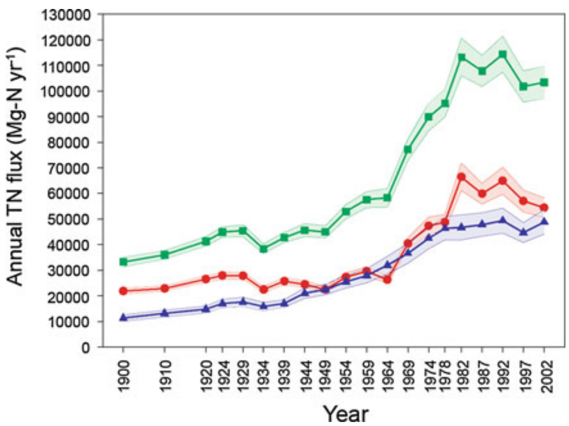

Fig. 6 Estimated annual export of N (Mg–N/year, circles) by major rivers draining to Lake Michigan from 1900 to 2002. Direct atmospheric input of N to Lake Michigan (Mg–N/year, *triangles*) is less than tributary input but nonetheless substantial. Total estimated N loading to Lake Michigan from tributary and atmospheric sources combined (*squares*) shows a rapid increase from 1960 to 1980, and no change or some decline thereafter. The shaded region around each *trend line* indicates ~95% confidence intervals based on a multivariate Monte Carlo uncertainty analysis that simulated 100 model runs for each of the 25 watersheds for the 20 census years from 1900 to 2002

Discussion

Construction of historical anthropogenic nitrogen budgets for 25 LMB watersheds over 1880–2002 illustrates that the rise in anthropogenic nitrogen loading to the terrestrial Lake Michigan Basin is non-linear, and different budget components show regional and sometimes off-setting patterns in response to changing economic, policy and technological drivers that influence inputs of anthropogenic N. Understanding the factors driving the uneven historic trends in major terms of the LMB nitrogen budget emphasizes the difficulty in forecasting, as well as the potential for shifts in policy or societal transitions to cause major changes in the regional nitrogen budget. Inputs of reactive nitrogen to LMB watersheds increased more than sixfold over the past 120 years, consistent with prior global (Galloway and Cowling 2002) and US (Howarth et al. 2002) analyses. Greatest change occurred after 1950 (Fig. 4), associated with large increases in fertilizer use and N-fixation, and a trend towards increased export of N in feed (seen as a negative import term). Net anthropogenic inputs averaged 650 kg-N/km^2/year (range 230–2700 kg-N/km^2/year) during 1880–1900, demonstrating human influence more than a century ago. By the end of the

20th century watersheds of the LMB received a basin-wide average of 3170 kg-N/km^2/year in 1987, and 2770 kg-N/km^2/year in 2002 (range 600–7,000 kg-N/km^2/year). These values are comparable to estimates for watersheds in the NE USA (3,400 kg-N/km^2/year, range 900–6300 kg-N/km^2/year, Van Breemen et al. 2002) and for New Zealand (3,650 kg-N/km^2/year, range: 1,200–6,900 kg-N/km^2/year, Parfitt et al. 2006), despite quite different agricultural practices and population densities. The relative importance of inputs varies considerably within and between studies, however. Agricultural watersheds of the LMB in recent times are dominated by fertilizer inputs followed by biological fixation, similar to those in NE USA (Boyer et al. 2002). New Zealand is unusual among temperate countries in that biological fixation by pasture legumes contributes the major fraction of N inputs (Parfitt et al. 2006).

Many of the most important drivers of rising nitrogen inputs to the LMB (Figs. 4 and 5) were national or global in scope, especially the greatly expanded use of inorganic fertilizer (Galloway and Cowling 2002). Across the USA from 1900 to 2002, total farmland area increased by 11.5%, the number of farms fell by more than half and the average farm size doubled (Broussard and Turner 2009), very similar to what we report for the LMB (Fig. 2). Agricultural intensification and specialization over the 20th century resulted in changing patterns in crop cultivation and livestock rearing and greater reliance on mechanization, and was influenced by changes in market forces and government-enacted policies. As others have noted, the decades after World War II saw explosive growth in N use, driven by rising prosperity as well as the rapidly expanding availability of inorganic fertilizer. Farm policies that encouraged farm consolidation and greater commodity specialization, together with advances in agricultural production (Dimitri et al. 2005), led to greater specialization in corn, soy, and non-ruminant animal production, as well as increased trade in animal feed and products. This era contrasts with a period of relative stasis following the end of the First World War, associated with a decline in foreign demand for American food and economic depression (Bowers et al. 1984; Granger and Kelly 2005). The era after about 1980 also differs in a number of ways from the explosive growth of the preceding several decades. Soybean cultivation in the LMB increased but corn production and fertilizer use stabilized, resulting in a moderate decline in net anthropogenic nitrogen inputs. Livestock production within the LMB fell and shifted from ruminants to non-ruminants, even as total crop production increased, which caused food and feed import to shift from a positive to a negative term. This is consistent with the increasing globalization of trade in animal feed, of which maize and soy are primary components, and the rapid growth in trade in animal feed and animal products (Galloway et al. 2007). The stasis of the past few decades, which perhaps runs counter to some public perception, may suggest that N inputs to the terrestrial system have reached a plateau.

Historical reconstruction of riverine TN export

The amount of N exported by rivers to receiving water bodies and the oceans is estimated to be as much as twenty times greater than under pre-settlement conditions (Howarth et al. 1996). Estimates of riverine TN export prior to about 1970 are few, however, due to the scarcity of measurements of TN concentrations in rivers, necessary for the estimation of riverine fluxes. Thus, our hind-casts of river export (Fig. 6) provide a unique perspective on the time-course of change. We estimate export of TN from LMB rivers to have increased three-fold over the 20th century, consistent with Broussard and Turner's (2009) report that measured nitrate–N concentrations for a number of US rivers were three to four times higher at the close of the 20th century as compared with the beginning. In addition, our estimated river export is relatively unchanging after 1980, consistent with studies that find little trend or declines in TN concentrations of US rivers since about 1975 (Alexander and Smith 2006; Sprague and Lorenz 2009). Results for the LMB correspond well to estimated TN yield to the Gulf of Mexico from the Mississippi River Basin. Goolsby and Battaglin (2001) reported a 2.3 fold increase from 200 kg-N/km^2/year for the 1955-70 average to 490 kg-N/km^2/year for the 1980–1999 average. For the same time period, the estimated yield of TN to Lake Michigan also increased twofold, from 280 kg-N/km^2/year for the 1954–1969 average to 550 kg-N/km^2/year for the 1982–1997 average.

Because anthropogenic N inputs, primarily from agriculture, were evident as early as 1880 and we were able to estimate river discharge only after 1900, we estimated pre-disturbance TN yield for rivers of the LMB to be 160 kg-N/km^2/year (range 130–190 kg-N/km^2/year depending on water yield). Using similar approaches, pre-disturbance TN export have been estimated to be 110 kg-N/km^2/year for watersheds of the NE USA (Howarth et al. (2002) and 45–110 kg-N/km^2/year for the Seine, France (Billen et al. 2007). These estimates are lower than minimally disturbed USA rivers (260 kg-N/km^2/year, Lewis 2002); however, greater TN yields from current undisturbed watersheds relative to modeled pristine conditions can be explained by higher atmospheric N deposition in modern time. Present-day inorganic wet deposition to the undisturbed forested watersheds studied by Lewis (2002) was 280 kg-N/km^2/year, versus an estimated ~30 kg-N/km^2/year deposited to LMB watersheds in 1880.

Total N inputs to Lake Michigan

Total N inputs to Lake Michigan from river transport and direct atmospheric deposition have risen more than three-fold over the 20th century (Fig. 6). Direct atmospheric deposition of N to the surface of Lake Michigan appears to have been nearly as large a term as river export throughout. This is explained partly by the large surface area of Lake Michigan (58,194 km^2), which is roughly half of the land area of the LMB. In addition, although net anthropogenic N inputs to the land are considerably larger than direct atmospheric deposition to the lake surface, only ~20% of inputs to the terrestrial system are exported by rivers (Han et al. 2009). Similar results were found for Lake Superior, where inputs of atmospheric N were lower but comparable on an aerial basis to our estimates for Lake Michigan, and atmospheric N deposition exceeded tributary inputs for essentially the entire 20th centuary (McDonald et al. 2010). The even greater relative contribution of atmospheric N to Lake Superior is unsurprising, as its watersheds are much less agricultural than those of Lake Michigan.

Conclusions

Our estimates of N inputs to watersheds of the LMB are comparable to other estimates of present-day N inputs (Van Breemen et al. 2002; Parfitt et al. 2006), of differences associated with land use (Boyer et al. 2002), of the magnitude of change over the past century or more (Galloway and Cowling 2002), and of inputs during earlier periods of small-scale farming (Billen et al. 2007). Although others have compared N production (Galloway and Cowling 2002) and riverine N concentrations at the beginning and end of the 20th century (Broussard and Turner 2009), our study is unusual in attempting to reconstruct a detailed timeline of N inputs to watersheds and N exports by rivers at 5–10 year intervals since 1880 or 1900. We thus provide insight into the uneven rise in N inputs to the landscape over time and among regions of the Lake Michigan basin, a land area of approximately 118,000 km^2. Because total N inputs are the sum of multiple sources, each of which may be affected differently by change in farming practices, technologies, wider economic forces and governmental policy, the shifting importance of inputs and even overall declines should be expected. Our retrospective analysis also suggests that there are opportunities for policy and societal transitions to alter nitrogen budgets in the future, as they clearly have done so in the past.

Finally, while we recognize the many sources of uncertainty in our reconstructions of historical trends in N inputs to watersheds and export by rivers, we believe that the estimates for the past several decades are the most reliable of the time series. In particular, the recent decline in fertilizer sales in some regions of the LMB is well documented. These data indicate relative stasis since about 1980, and provide an alternative to the expectation of further increases in N inputs based on the trend from 1900 to 2000. Although N loading to terrestrial systems and export to aquatic systems have unquestionably increased many-fold over the past century or more, our analysis shows that growth for the LMB to be episodic and punctuated by periods of stasis, including the present. These uneven periods of rapid increase reflect societal transitions that affect one or more drivers in ways best understood in hindsight, which underscores the challenge of forecasting of future trends in N loading to ecosystems.

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