

## History of nutrient inputs to the northeastern United States, 1930–2000

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[1] Humans have dramatically altered nutrient cycles at local to global scales. We examined changes in anthropogenic nutrient inputs to the northeastern United States (NE) from 1930 to 2000. We created a comprehensive time series of anthropogenic N and P inputs to 437 counties in the NE at 5 year intervals. Inputs included atmospheric N deposition, biological N<sub>2</sub> fixation, fertilizer, detergent P, livestock feed, and human food. Exports included exports of feed and food and volatilization of ammonia. N inputs to the NE increased throughout the study period, primarily due to increases in atmospheric deposition and fertilizer. P inputs increased until 1970 and then declined due to decreased fertilizer and detergent inputs. Livestock consistently consumed the majority of nutrient inputs over time and space. The area of crop agriculture declined during the study period but consumed more nutrients as fertilizer. We found that stoichiometry (N:P) of inputs and absolute amounts of N matched nutritional needs (livestock, humans, crops) when atmospheric components (N deposition, N<sub>2</sub> fixation) were not included. Differences between N and P led to major changes in N:P stoichiometry over time, consistent with global trends. N:P decreased from 1930 to 1970 due to increased inputs of P, and increased from 1970 to 2000 due to increased N deposition and fertilizer and decreases in P fertilizer and detergent use. We found that nutrient use is a dynamic product of social, economic, political, and environmental interactions. Therefore, future nutrient management must take into account these factors to design successful and effective nutrient reduction measures.

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### 1. Introduction

[2] Globally, humans have increased the availability of the often-limiting nutrients nitrogen (N) and phosphorus (P) [Vitousek *et al.*, 1997b; Falkowski *et al.*, 2000; Galloway *et al.*, 2008]. Increased nutrient availability has had many positive effects on human well-being globally, primarily by increasing crop yields and therefore human food supply. However, it has also led to ecologically damaging and economically costly eutrophication problems [Carpenter *et al.*, 1998]. Humans have changed N and P cycles differently; whereas global N availability has doubled, P availability has quadrupled since the preindustrial age [Falkowski *et al.*, 2000]. At the same time, regulatory controls for P in the United States have typically been more widespread and more successful than those for N (e.g., P detergent bans [Litke,

1999]; P turf fertilizer bans [Lehman *et al.*, 2009]). However, human nutrient use does not respond to regulations alone. Although there have been no regulations directly controlling the use of agricultural fertilizer, inputs of fertilizer P have declined in the United States since 1980 [Alexander and Smith, 1990]. Global nutrient cycles are frequently depicted as systems spiraling out of control [Vitousek *et al.*, 1997a; Childers *et al.*, 2011]; yet declines in P inputs from agricultural fertilizer, in the absence of direct regulation, suggest that anthropogenic nutrient cycles may respond to a diversity of drivers and thus may be subject to additional socio-ecological feedback (in the sense of Liu *et al.* [2007]).

[3] Because of the long-term legacy effects of human environmental management [McGuire *et al.*, 2001; Pastore *et al.*, 2010; Bain *et al.*, 2012; MacDonald *et al.*, 2012], a historical approach is critical for understanding how absolute quantities and geographic patterns of nutrient use emerge over time [Barles, 2007; Billen *et al.*, 2007] and provides a context for understanding the state of modern socio-ecosystems [Foster *et al.*, 2003; Billen *et al.*, 2007]. Importantly, Barles [2007] notes that changes in human-nutrient systems are “not necessarily...continuous, systematic and deliberate.” Human nutrient use is intricately tied to how we produce food and how we deal with waste [Jordan and Weller, 1996; Barles, 2007; Billen *et al.*, 2007; Cordell *et al.*, 2009]. It is therefore also tied to the technologies that society has available for these two activities (e.g., fertilizer, water treatment), human perceptions regarding waste [e.g., Barles, 2007], and the political and economic conditions

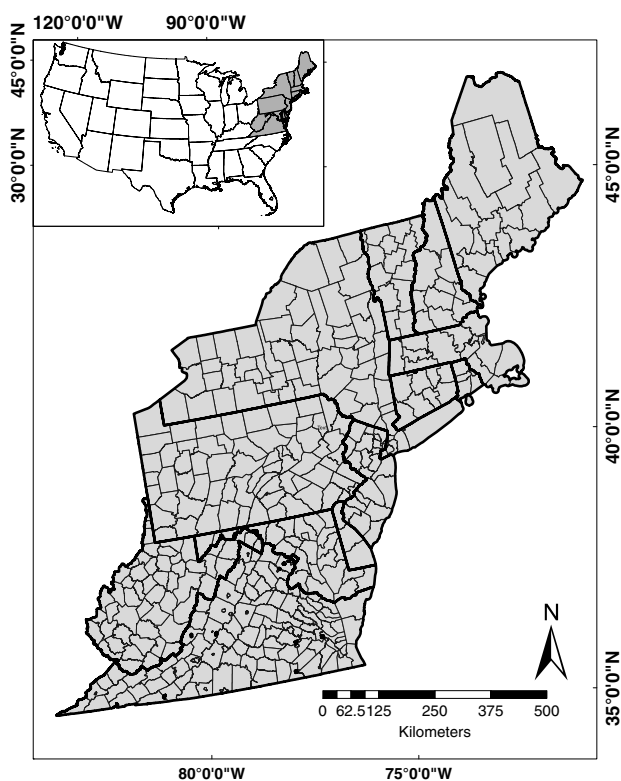
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**Figure 1.** Study area includes the 13 northeastern states and 437 counties.

within which these activities take place (e.g., U.S. food policy, global grain markets).

[4] Historical approaches are also useful for understanding ecological consequences of nutrient inputs and the development of scientific knowledge regarding the pollution resulting from those inputs [Howarth and Marino, 2006]. Recent research suggests that increases in anthropogenic N inputs relative to P are a global phenomenon [Peñuelas *et al.*, 2012] that may be causing shifts in nutrient limitation as well as species composition in both fresh [Elser *et al.*, 2009] and marine waters [Justic *et al.*, 1995; Turner *et al.*, 2003; Billen *et al.*, 2007; Grizzetti *et al.*, 2012]. Understanding the mechanisms underlying historic and ongoing changes in nutrient inputs is critical to designing effective solutions to current and future nutrient pollution [Foster *et al.*, 2003] and improving the current watershed management emphasis on problem remediation rather than prevention [Vörösmarty *et al.*, 2010].

[5] Nutrient use by humans is variable not only temporally but also spatially. Differences in the absolute quantities, drivers, and stoichiometry of nutrient use vary across regional and global scales [Jordan and Weller, 1996; Boyer *et al.*, 2002; Vitousek *et al.*, 2009] as a result of differences in land use [Jordan and Weller, 1996; Boyer *et al.*, 2002] and economic development [Vitousek *et al.*, 2009]. Many recent assessments of anthropogenic nutrient inputs to the United States have focused on areas dominated by row-crop agriculture, especially the Mississippi River basin [David and Gentry, 2000; Donner *et al.*, 2004; Alexander *et al.*, 2008; Broussard and Turner, 2009]. Relatively little research has focused on the historical nutrient patterns of the northeastern United States (NE, Figure 1) from 1930 to 2000. The NE is one of the most densely populated regions of the

U.S., and over the twentieth century, it experienced a significant reduction in cropland concurrent with a near doubling of the human population. However, the NE is also a major meat-producing region for the U.S., and so livestock agriculture is a major driver of nutrient cycling [Boyer *et al.*, 2002]. Furthermore, the NE has vast areas of forested land that is subject to high rates of atmospheric N deposition [Boyer *et al.*, 2002].

[6] The objectives of this paper are to (i) describe changes in the geographic patterns of nutrient inputs to the NE region over a near-century timeframe (1930–2000), (ii) assess how nutrient inputs have responded to changing demography, land use, technology, and legislation during this period, and (iii) identify potential ecological consequences of changes in nutrient inputs over time.

## 2. Methods

[7] We used a mass balance approach [Green *et al.*, 2004] to estimate net anthropogenic inputs of N and P to the NE during the twentieth century. We created nutrient budgets for the 437 NE counties at 5 year time steps from 1930 to 2000. For the present study, we measured the net inputs of nutrients to or from each county as fertilizer, atmospheric deposition (N only), biological N<sub>2</sub> fixation, livestock feed, human food, and detergent phosphates. To avoid double counting, the total net inputs of N and P took into account transfers within the county (e.g., crops consumed as human food). Where local production of food and feed exceeded local consumption, the balance was negative and was defined as a net export from the county. Exports included only excess agricultural production and ammonia volatilization. We use “net inputs” to refer to inputs associated with a single source of nutrients and “total net inputs” to refer to inputs from all sources. We made the simplifying assumption that P inputs from geologic weathering were unchanging and did not include them here. Manure and human sewage were calculated but were not considered additional inputs as they result from internal recycling of nutrients from fertilizer, food, and feed imports. Our budgets estimated the net inputs or exports of nutrients to or from each county and to watersheds, but we did not track nutrient transport or processing in receiving waters downstream. We also did not account for management strategies, such as riparian buffers or wastewater treatment, which may reduce the pathways and fluxes of nutrients in aquatic ecosystems.

### 2.1. Atmospheric Deposition

[8] Data on N deposition rates are limited in time and space. To describe temporal changes in atmospheric deposition, we estimated atmospheric deposition of N to the whole region using relationships between gaseous N emissions (as nitrogen oxides [NO<sub>x</sub>], and ammonia [NH<sub>3</sub>]) and N deposition. State-level NO<sub>x</sub> emissions data for 1930 to 2000 came from Gschwandtner *et al.* [1985] and the Environmental Protection Agency (EPA) National Emissions Inventory (NEI, <http://www.epa.gov/ttn/chieftrends/index.html>). Although state-level NH<sub>3</sub> emissions were available for 2000 from the EPA, they were not available for early parts of the century. We therefore calculated historic NH<sub>3</sub> emissions using manure and fertilizer data from our data set (see below for manure and fertilizer methods) and NH<sub>3</sub> volatilization

**Table 1.** N<sub>2</sub> Fixation Rates for Various Crops With References

Crop	N <sub>2</sub> Fixation Rate (kg N ha <sup>-1</sup> yr <sup>-1</sup> )	Reference
Soybeans	78	Barry <i>et al.</i> [1993] and Messer and Brezonik [1983]
Peanuts	86	Barry <i>et al.</i> [1993] and Messer and Brezonik [1983]
Nonlegume crops	5	Barry <i>et al.</i> [1993] and Messer and Brezonik [1983]
Alfalfa hay	218	Keeney [1979]
Nonalfalfa hay	116	Keeney [1979]
Dry edible beans	40	Keeney [1979] and Stevenson [1982]
Nonwooded pasture	15	Keeney [1979]

coefficients from *Battye et al.* [1994] and *Boyer et al.* [2002]. Our calculated NH<sub>3</sub> emissions were well correlated with EPA emissions data for the year 2000 ( $r=0.998$ ,  $p < 0.001$ ). We obtained atmospheric deposition rates for the year 2000 from the National Atmospheric Deposition Network (NADP, <http://nadp.sws.uiuc.edu/>). The NADP collects data on annual wet deposition rates for nitrate (NO<sub>3</sub><sup>-</sup>) and ammonium (NH<sub>4</sub><sup>+</sup>) from 41 sites throughout the NE. We estimated total deposition (wet+dry) by assuming that dry deposition of NH<sub>4</sub><sup>+</sup> is 18% of wet deposition and dry deposition of NO<sub>3</sub><sup>-</sup> is 48% of wet deposition [*Bowen and Valiela*, 2001]. These data were then spatially interpolated in ArcGIS (Environmental Systems Research Institute, Redlands, California) using inverse distance weighting (following NADP protocols) to create a continuous loading surface and calculate average state-level deposition. We regressed state-level emissions against state-level deposition data for year 2000 independently for NO<sub>3</sub><sup>-</sup> ( $R^2=0.769$ ,  $p < 0.001$ ) and NH<sub>4</sub><sup>+</sup> ( $R^2=0.729$ ,  $p < 0.001$ ). These relationships were then applied to historic NO<sub>x</sub> and NH<sub>3</sub> emissions data to estimate past N deposition levels at the state level, which were added to estimate total N deposition to the region.

[9] To describe changes in the spatial pattern of N deposition, we used point-scale deposition data from the NADP and earlier literature [*Eriksson*, 1952; *Fisher*, 1968; *Pearson and Fisher*, 1971; *Cogbill and Likens*, 1974] to estimate atmospheric deposition based on latitude, longitude, and year. Deposition data extended from 1920 (four sites) to 2000 (41 sites). We developed separate regression equations for the wet deposition of NO<sub>3</sub><sup>-</sup> and NH<sub>4</sub><sup>+</sup>. These regression equations (equations (1) and (2)) were then used to create a grid (7 km resolution) of N deposition rates for each study year in ArcGIS. Mean N deposition rates were then calculated for each county and each year in ArcGIS:

$$\text{NO}_3^- = -489.9 - 1.01 \times \text{Longitude} + 0.97 \\ \times \text{Latitude} + 0.19 \times \text{Year} (R^2 = 0.24, p < 0.0001) \quad (1)$$

$$\text{NH}_4^+ = -50.68 - 0.21 \times \text{Longitude} + 0.20 \\ \times \text{Latitude} + 0.01 \times \text{Year} (R^2 = 0.27, p < 0.0001) \quad (2)$$

## 2.2. Fertilizer Application

[10] Fertilizer application rates (kg N or P ha<sup>-1</sup> yr<sup>-1</sup>) for 1945 to 2002 were obtained at the county level from two U.S. Geological Survey (USGS) reports (for years 1945–1985 [*Alexander and Smith*, 1990] and for years 1982–2001 [*Ruddy et al.* 2006]). To estimate fertilizer application rates for earlier decades, we used state-level fertilizer sales and nutrient content from fertilizer use surveys [*Smalley*, 1929, 1939] to calculate

inputs of nutrients to each state (equation (3)). State-level data were then disaggregated to county level using county harvested cropland (1930 and 1940) as a proportion of total state cropland data from the Census of Agriculture [*U.S. Census Bureau (USCB)*, 1932, 1942]. All inputs were calculated as kg N or P county<sup>-1</sup> yr<sup>-1</sup> and then divided by county area to obtain inputs rates in units of kg N or P ha<sup>-1</sup> yr<sup>-1</sup>:

$$F_{ik} = F_i \times N_i \times C_{ik}/C_i \quad (3)$$

where

- $F_{ik}$  inputs of fertilizer N or P for the  $k$ th county in the  $i$ th state (kg);
- $F_i$  fertilizer sales for the  $i$ th state (kg);
- $N_i$  nutrient content of fertilizer in the  $i$ th state (%);
- $C_{ik}$  area of harvested cropland for the  $k$ th county in the  $i$ th state (ha);
- $C_i$  area of harvested cropland for the  $i$ th state (ha).

## 2.3. Biological Nitrogen Fixation

[11] Biological N<sub>2</sub> fixation was calculated by multiplying crop and pasture areas [*USCB*, 1932, 1942; *U.S. Department of Agriculture (USDA)*, 1980, 1990, 1993, 1999, 2004] by rates of N<sub>2</sub> fixation obtained from *Jordan and Weller* [1996] and sources cited therein (Table 1). Because land use data for our study period were unavailable and rates of N<sub>2</sub> fixation in nonagricultural lands are usually low [*Jordan and Weller*, 1996], we assumed nonagricultural land had negligible N<sub>2</sub> fixation rates.

## 2.4. Crop-Livestock Balance

[12] For each county at each time step, we calculated N and P in crops harvested, feed imported for livestock, and manure production to calculate the net input of N and P as feed and food to or from the county (equations (4)–(6)). All inputs were calculated in units of kg county<sup>-1</sup> yr<sup>-1</sup> and then divided by county area (in ha) to obtain net inputs as kg N or P ha<sup>-1</sup> yr<sup>-1</sup>. We used a spoilage rate of 10% for all food and feed, following *Jordan and Weller* [1996].

[13] For livestock feed,

$$LF_k = LD_k - LS_k \times S \quad (4)$$

where

- $LF_k$  net inputs of nutrients in livestock feed for the  $k$ th county (kg);
- $LD_k$  demand for nutrients by livestock in the  $k$ th county (kg);
- $LS_k$  supply of nutrients for livestock feed by local crop production in the  $k$ th county (kg);
- $S$  rate of spoilage (%).

[14] For consumption of N and P by humans,

$$HF_k = HCD_k - HCS_k \times S + HLD_k - HLS_k \times S \quad (5)$$

where

- $HF_k$  net inputs of nutrients in human food for the  $k$ th county (kg);  
 $HCD_k$  demand for nutrients from crops by humans in the  $k$ th county (kg);  
 $HCS_k$  supply of nutrients for human food by local crop production in the  $k$ th county (kg);  
 $HLD_k$  demand for nutrients from livestock by humans (e.g., meat, milk, and eggs) in the  $k$ th county (kg);  
 $HLS_k$  supply of nutrients for human food from local live stock production in the  $k$ th county (kg).

[15] The local livestock production is defined as

$$HLS_k = (LD_k - LM_k) \quad (6)$$

where  $LM_k$  is the production of manure by livestock in the  $k$ th county (kg).

[16] Total nutrients in crop harvest were estimated by multiplying county-level crop production data [USCB, 1932, 1942; USDA, 1980, 1990, 1993, 1999, 2004] by crop-specific nutrient content [Lander and Moffit, 1996] for the following crops: corn for grain, wheat, oats, barley, rye, soybeans, potatoes, sorghum, alfalfa hay, and nonalfalfa hay [Boyer et al., 2002]. We assumed that the nutrient content of each crop was constant over time. We assumed that the net input of food and feed was the difference between county-level supply and county-level demand. Therefore, if livestock feed supply was less than livestock feed demand, we assumed a net input of feed to make up the difference. Conversely, if supply was greater than demand, the balance was negative and the excess was assumed to be exported. Boyer et al. [2002] used this approach to estimate anthropogenic N inputs to the NE for a single year and found a strong correlation between feed imports calculated using this method and imports estimated from feed expenditure data. Local crop production is consumed by either livestock or humans or is exported. After subtracting a 10% spoilage rate [Jordan and Weller, 1996], we made the following proportions of crops available for human consumption (i.e.,  $HCS_k$ ): 100% of potatoes, 61% of wheat, 17% of rye, 4% of corn, 6% of oats, and 3% of barley [Jordan and Weller, 1996]. The remaining crops were made available for livestock consumption (i.e.,  $LS_k$ ) [Jordan and Weller, 1996]. Any crops not consumed by humans or livestock were exported.

[17] Livestock nutrient demand ( $LD_k$ ) was calculated from county-level inventories of livestock (cattle, chickens, turkeys, hogs, and pigs) from the Census of Agriculture [USCB, 1932, 1942; USDA, 1980, 1990, 1993, 1999, 2004] and published nutrient consumption rates [Van Horn et al., 1996]. Nutrient loss as manure ( $LM_k$ ) was calculated using livestock inventories and published manure production rates per animal [Van Horn et al., 1996]. The difference between feed inputs and manure losses, minus a spoilage rate of 10% [Jordan and Weller, 1996] was assumed to go to human food products ( $HLS_k$ ). Any production in excess of local demand was exported.

## 2.5. Human Food and Waste

[18] To calculate net inputs of food nutrients consumed by humans, we calculated dietary demand for N and P using county-level population [USCB, 1995, 2002] and estimates

of per-capita N and P consumption rates. Based on the average protein consumption in the United States ( $80 \text{ g d}^{-1}$ ) [Geissler and Powers 2005], we estimated N intake to be  $4.7 \text{ kg cap}^{-1} \text{ yr}^{-1}$ . This is similar to other values used in the literature [Boyer et al., 2002; Han and Allen, 2012]. The USDA recommended daily allowance of P is  $0.256 \text{ kg cap}^{-1} \text{ yr}^{-1}$  [Geissler and Powers 2005], and available P in the food supply averaged  $0.55 \text{ kg cap}^{-1} \text{ yr}^{-1}$  during our study period [Gerrior et al., 2004]. Assuming that P consumption is higher than recommended values but lower than that available in food supply, we averaged these values to obtain a per capita P consumption rate of  $0.4 \text{ kg P yr}^{-1}$ . This value is similar to other estimates in the literature [Meybeck et al., 1989; David and Gentry, 2000]. We assumed no net accumulation of individuals for a given year; that is, demand for food nutrients was assumed to be equal to nutrients in human waste. We assumed that human protein (and therefore nutrient) demand from animal and crop sources was 70% and 30%, respectively [Food and Agriculture Organization of the United Nations, 2012]. Net nutrient input or export to each county as food was calculated as the difference between human food demand and local supply. This was calculated separately for animal and crop sources (equation (5)).

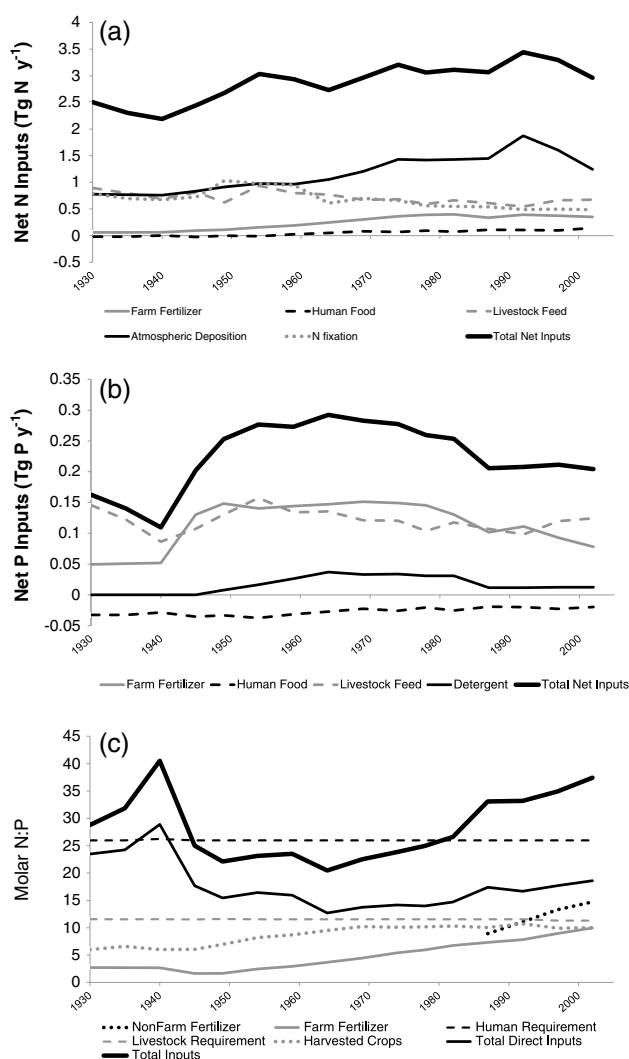
## 2.6. Detergent Phosphates

[19] Phosphate-containing detergents were not used until 1945, but by 1970 inputs from detergents had reached  $0.8 \text{ kg P cap}^{-1} \text{ yr}^{-1}$  [Chapra, 1980]. We assumed a linear increase in per-capita phosphate use from 1945 to 1970. The first bans on detergent phosphates emerged in 1971. We used data from Litke [1999] on detergent bans (ban dates and phosphate limits for each state) to estimate state-level per-capita inputs for each decade. To convert the Litke data, given as detergent phosphate concentrations, to per-capita inputs, we assumed a pre-ban detergent P content of 12% for calculations [Litke, 1999].

## 2.7. Temporal and Spatial Statistics

[20] To determine the significance of trends in nutrient inputs to the region over time, we performed a linear regression using year as the predictor variable for annual net inputs to the region (total and for individual sources).

[21] We investigated the degree to which nutrient inputs were distributed or concentrated across the NE throughout the twentieth century by spatial autocorrelation. Spatial autocorrelation measures the degree to which data from locations close to each other are more similar than from remote locations [O'Sullivan and Unwin, 2003]. Positive spatial autocorrelation, the most commonly observed type, indicates that the data values for spatial entities located near each other (e.g., contiguous counties) are similar. Global spatial autocorrelation details the degree to which statistically significant spatial clustering of high or low data values occurs throughout the study area. We used Moran's I [Chang, 2008] to measure the global spatial autocorrelation for the NE for 5 year periods between 1930 and 2000. Moran's I does not provide detail on where within the study area nutrient inputs were concentrated. Therefore, we also used a local spatial autocorrelation metric, Local Indicators of Spatial Association (LISA) [Anselin, 1995; Franczyk and Chang, 2009], to identify the locations of statistically significant spatial clustering within the study area.



**Figure 2.** Temporal trends in net regional inputs of (a) nitrogen and (b) phosphorus (c) the molar stoichiometry of nutrient inputs.

### 3. Results and Discussion

#### 3.1. Patterns in Nutrient Inputs to the NE Over Time

[22] Total net N inputs over the entire region increased steadily and significantly over the study period (Figure 2a;  $R^2=0.70$ ,  $p < 0.001$ ). Farm-fertilizer inputs of N to the landscape increased significantly ( $R^2=0.91$ ,  $p < 0.001$ ), despite a decline in cropland area, reflecting agricultural intensification. Food N was exported most years, but exports decreased significantly over the study period ( $R^2=0.91$ ,  $p < 0.001$ ; Figure 2a). As the population nearly doubled (from 35 to 67.6 million), crop production for human food declined (12% to 7% of total crop production). Although human food N was exported throughout most of the century, the food system as a whole was an importer of N via fertilizer and livestock feed (Figure 2a).

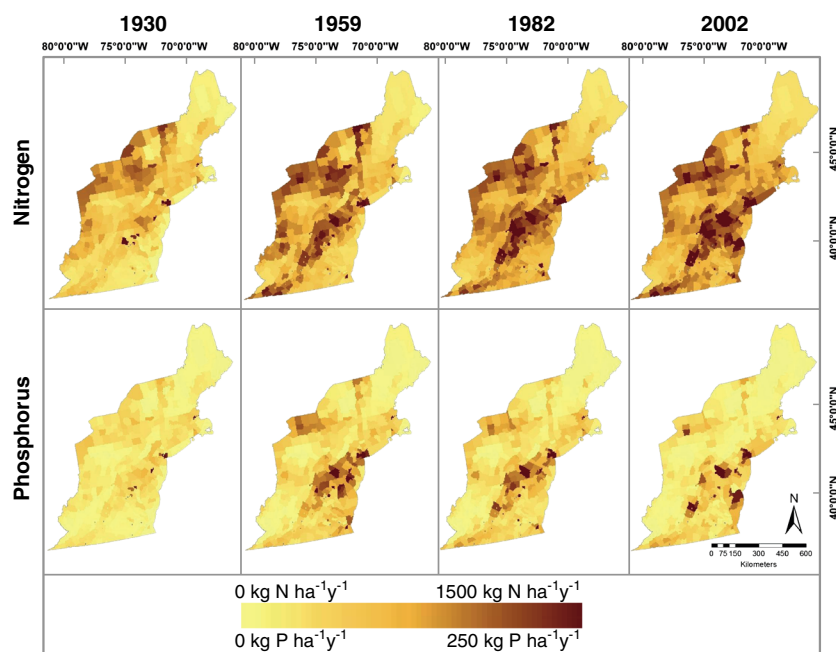
[23] Atmospheric N deposition increased significantly over the study period ( $R^2=0.88$ ,  $p < 0.0001$ ) and contributed as much as 47% of net N inputs to the NE by the end of the century. Since the mid-1990s, however, N deposition has decreased slightly due to reduced  $\text{NO}_x$  emissions (EPA NEI,

<http://www.epa.gov/ttn/chief/trends/index.html>). Inputs of N as livestock feed ( $R^2=0.39$ ,  $p < 0.05$ ) and  $\text{N}_2$  fixation ( $R^2=0.36$ ,  $p < 0.05$ ) decreased throughout the study period; however, these remained large contributors to total inputs, 23% and 15%, respectively, as of 2002.

[24] In contrast to N, there was no consistent linear trend in total net P inputs across the entire study period (Figure 2b). Instead, total net P inputs increased nearly fourfold from 1940 to 1969, from  $0.08 \text{ Tg P yr}^{-1}$  to  $0.29 \text{ Tg P yr}^{-1}$ , followed by a significant decline in the 1970s ( $R^2=0.79$ ,  $p < 0.01$ ). Human food P was exported throughout the study period, but exports declined significantly over time ( $R^2=0.71$ ,  $p < 0.001$ ), tracking the pattern of food N. Both detergent and fertilizer P inputs peaked around 1970 and thereafter declined significantly (detergent:  $R^2=0.75$ ,  $p < 0.05$ ; fertilizer:  $R^2=0.90$ ,  $p < 0.01$ ). Inputs of livestock feed P did not demonstrate any significant trend over the study period.

[25] Since temporal patterns of net N and P inputs differed over the study period, the N:P of nutrient inputs also changed over time (Figure 2c). The N:P of total net inputs increased from 1930 to 1940 as a result of several smaller changes in livestock populations and crop production. From 1940 to the mid-1960s, the trend reversed as inputs of P fertilizer and detergents increased more rapidly than fertilizer N. The N:P of total inputs steadily increased from 1965 to 2002 due to concurrently increasing N inputs and declining P inputs. Across the study period, the N:P of total net inputs was consistently greater than the N:P requirements of humans. When we excluded atmospheric deposition of N from the total (i.e., “Total direct inputs,” Figure 2c) as an indicator of food system N:P, the N:P of inputs was high from 1930 to 1940, largely due to very low inputs of P fertilizer and high inputs of N via biological  $\text{N}_2$  fixation. From 1940 onward, the N:P of total direct inputs was bounded between the livestock requirement of  $\sim 11$  and the human requirement of  $\sim 26$ . This not only suggests that the major drivers of nutrient requirements were demands for human food and livestock feed but also indicates that P and N inputs were well matched with regard to demands. Excluding detergent P and nonfarm fertilizer inputs of N and P does not substantially change this pattern: N:P remains bounded within 11 and 26 (data not shown).

[26] Two other important trends of note are the increases in N:P of farm fertilizer since 1950 (Figure 2c) and nonfarm fertilizer since the mid-1980s (data not shown). The N:P of farm fertilizer was much lower than that of harvested crops throughout much of the study period, but the two lines converged at the end of the century, indicating that fertilizer additions more closely matched crop needs. Of course, N is also added to croplands via  $\text{N}_2$  fixation. The stoichiometry of all agricultural inputs ( $\text{N}_2$  fixation and N fertilizer, P fertilizer) is much above that of crop removal throughout the study period, which indicates an oversupply of N to crop systems. Although the stoichiometry of inputs was at most 6.5 times higher than crop uptake, the N:P of inputs moved toward the N:P of crop removal over time, i.e., nutrient additions were more in balance with crop removal. One final consideration with regard to nutrient additions to agricultural soils is that P is much more likely to accumulate in soils, whereas N is more likely to leach from the soil column. That is, the



**Figure 3.** Net N and P inputs ( $\text{kg ha}^{-1} \text{yr}^{-1}$ ) for 1930, 1960, 1982, and 2002.

average residence time of N and P may be different, and therefore, annual inputs of fertilizers may not reflect the N:P of plant-available nutrients in agricultural soils.

### 3.2. Spatial Patterns of Nutrient Inputs

[27] The overriding trend for the twentieth century NE has been a major reorganization of the landscape, as agriculture shifted southward, human population density increased, and livestock populations became more concentrated. The major pattern was a spatial separation of food production from food consumption. At the end of the twentieth century, agricultural inputs (livestock feed, fertilizer, and  $\text{N}_2$  fixation) remained the largest inputs of nutrients at the regional scale and were the largest source of nutrients for most counties. However, the decline in agricultural inputs for most counties (N: 71%, P: 64% of counties) mirrored an increase in urban N and P inputs (human food, nonfarm fertilizer, and detergent P, 72% of counties). The key feature of these trends was their spatial pattern: declines in agricultural inputs were collocated with increases in urban inputs (Figure 4; N:  $r = -0.22$ , P:  $r = -0.32$ ), suggesting a specialization of the landscape into separate urban and agricultural subregions (Figure 4).

[28] Spatially, N and P inputs became more clustered throughout the region, as measured by Moran's I. Moran's I of N inputs ranged from 0.24 to 0.54 and increased significantly over the study period ( $R^2 = 0.34$ ,  $p = 0.02$ ); Moran's I values for P inputs ranged from 0.36 to 0.65 and also increased significantly over the study period ( $R^2 = 0.76$ ,  $p < 0.0001$ ). Differences in the changes in clustering between N and P are likely due to fertilizer use patterns (less widespread for P than N) and N deposition (higher rates in forested areas of the NE). Moran's I was also consistently higher for P than for N throughout the study period. Hot spots of nutrient inputs—clusters of counties with statistically high nutrient inputs as identified by LISA [Anselin, 1995; Franczyk and Chang, 2009]—were similar for N and P (Figure 3). The spatial statistical analysis revealed persistent hot spots of N and P inputs

around the New York metropolitan area (Figure 3). Since 1970, hot spots emerged around the Chesapeake Bay in Virginia, Pennsylvania, Maryland, and Delaware.

[29] Nutrient inputs shifted southward over the study period (Figure 4). Changes in nutrient inputs from 1930–2000 were significantly negatively correlated with latitude for both N ( $R^2 = 0.09$ ,  $p < 0.0001$ ) and P ( $R^2 = 0.07$ ,  $p < 0.0001$ ). These shifts were related to significant southward shifts in agricultural inputs. Changes in livestock nutrient demand from 1930 to 2002 were significantly negatively correlated with latitude (N and P:  $R^2 = 0.11$ ,  $p < 0.0001$ ), as were changes in fertilizer (N:  $R^2 = 0.05$ ,  $p < 0.0001$ ; P:  $R^2 = 0.11$ ,  $p < 0.0001$ ) and  $\text{N}_2$  fixation ( $R^2 = 0.33$ ,  $p < 0.0001$ ) from 1930 to 2002 (Figure 4). Changes in human population density over the study period were not significantly related to latitude.

### 3.3. Drivers of Changes Over Space and Time

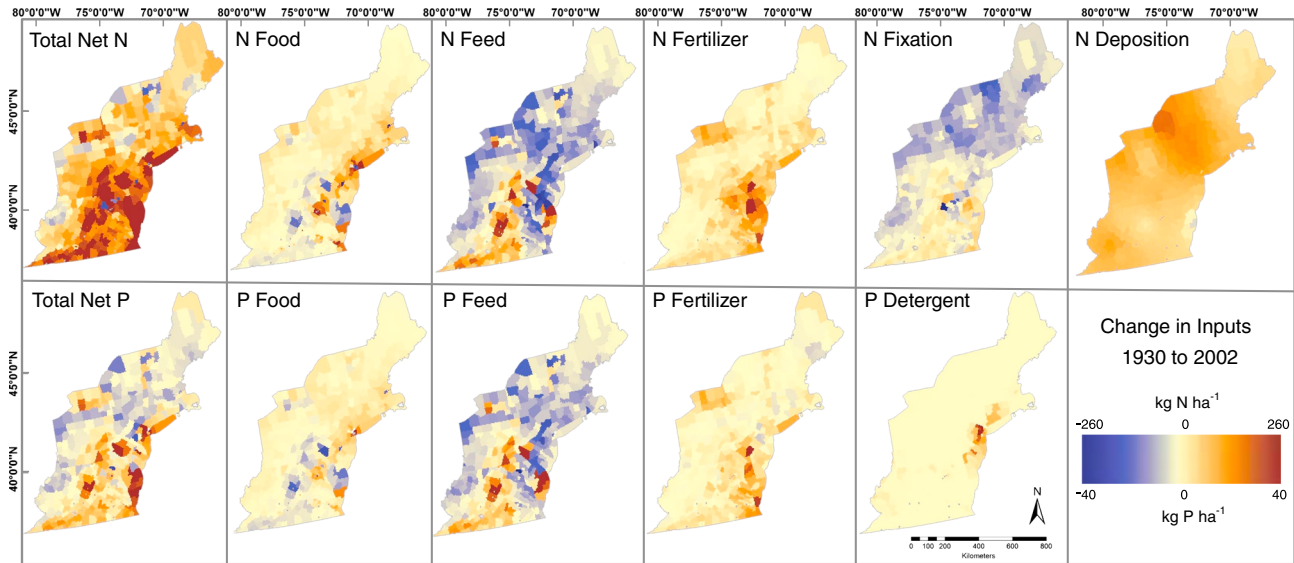
[30] Major changes in N and P inputs are apparent at the regional scale over space and time (Figures 2–4). These changes resulted from changes in land use, technology, fertilizer and food production, and nutrient emission control legislation.

#### 3.3.1. Role of Livestock Agriculture

[31] Previous work on the theory of ecological stoichiometry suggests that human activities disproportionately affect P cycling in order to bring nutrient ratios toward the N:P of the human body [Sturner and Elser, 2002]. Our stoichiometry results suggest that livestock and human nutritional requirements are key drivers of nutrient inputs to the NE (Figure 2c). This pattern is in stark contrast to nutrient inputs in the central part of the U.S., which are driven by row-crop agriculture [Alexander et al., 2008; Broussard and Turner, 2009].

[32] Livestock husbandry was a defining feature of the NE nutrient landscape during the twentieth century. The majority of the inputs of nutrients to the region were used to support





**Figure 4.** Change in N and P inputs from 1930 to 2002 for total net N and P inputs and individual nutrient sources. Data are presented as kg P (or N)  $\text{ha}^{-1}$ . Blue colors indicate a decrease in nutrient inputs from 1930 to 2002, orange colors indicate an overall increase in nutrient inputs from 1930 to 2002, and tan colors indicate no net change from 1930 to 2002.

livestock agriculture, either directly as livestock feed or indirectly as fertilizer, most of which was used on feed crops. Despite declines in cropland, crop production increased during the twentieth century, peaking in 1992. Much of this production was for livestock feed crops, and production of food crops for human consumption did not change significantly since 1930 (Figures 5a and 5b). Despite massive inputs of fertilizer to produce feed crops, the crop system only provided 12 to 50% (33% on average) of the nutrients required by livestock. The remaining nutrient demand was met with imported feed (Figures 5c and 5d). This system was highly inefficient in terms of nutrient use. The greater part of feed nutrients was converted to manure, and only 6–24% of the nutrient inputs to the regional livestock system were consumed by humans locally (Figures 5c and 5d).

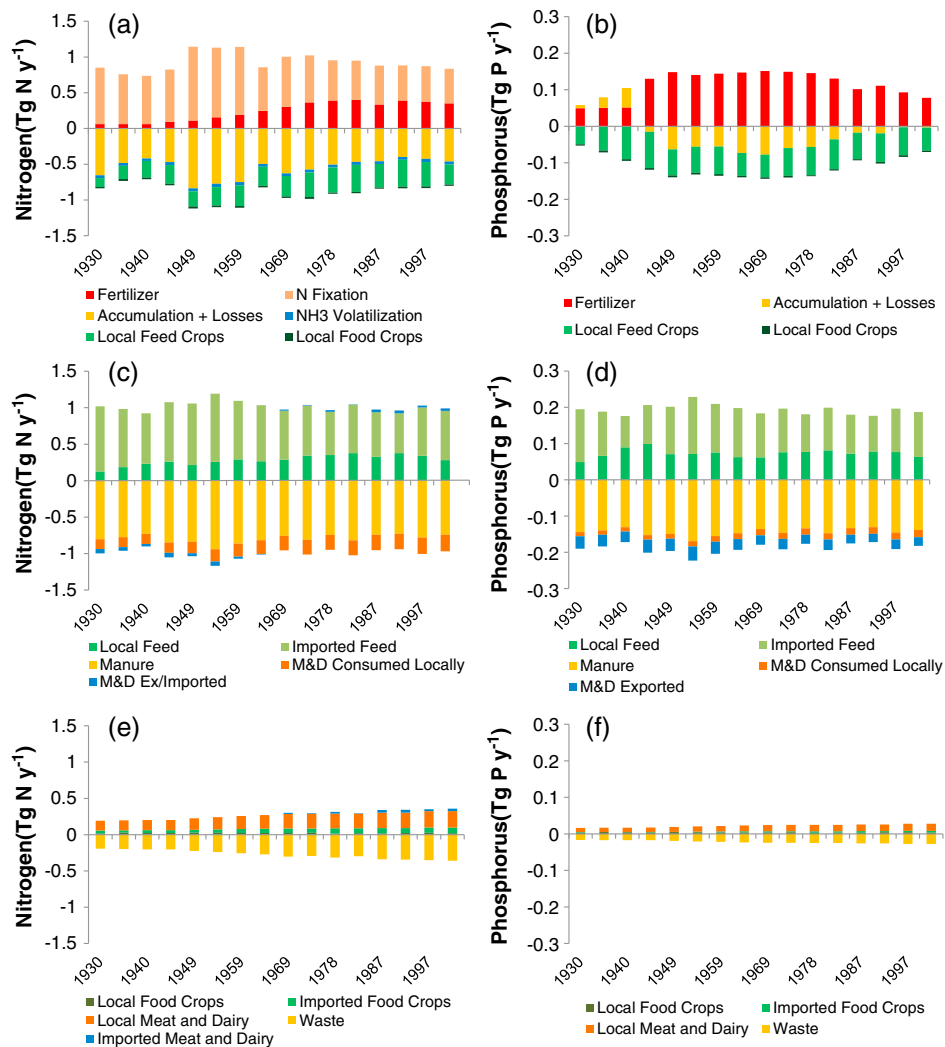
[33] Spatial patterns also demonstrate the importance of livestock to NE nutrient inputs. To understand how the drivers of nutrient inputs varied between counties, we categorized counties as human, livestock, or crop driven based on which had the largest demand for nutrients. We then regressed total net N and P inputs ( $\text{kg N}$  or  $\text{P ha}^{-1} \text{yr}^{-1}$ ) against human population density, total livestock nutrient requirements (a proxy for livestock population density), and crop uptake. Across all counties, human population density was the best predictor of total net nutrient inputs  $\text{ha}^{-1} \text{yr}^{-1}$  across time (correlation coefficients ranged from 0.88 to 0.98), and the highest nutrient inputs were in counties with the highest population density ( $>5$  people  $\text{ha}^{-1}$ ,  $N=25$  counties in 1930 and 63 counties in 2002; Figure 6). However, for counties with low population density ( $<5$  people  $\text{ha}^{-1}$ ,  $N=412$  counties in 1930 and 374 counties in 2002), livestock nutrient requirements  $\text{ha}^{-1}$  (an integrative proxy for livestock population density that incorporates ranges in livestock body mass and nutrient demand) was the best predictor of total net N and P inputs  $\text{ha}^{-1}$  (Figure 6). Furthermore, although human population densities were the best predictor of nutrient inputs overall, livestock were the best predictor

of nutrient inputs for the majority of counties. However, there was a decrease in livestock-driven counties over time, from 84% to 61%, and an increase in human-driven counties, from 15% to 31%, and crop-driven counties, from 0.5% to 9%.

[34] Although the number of livestock-driven counties decreased over the study period, average nutrient inputs to livestock-driven counties increased from 35 to 58  $\text{kg N ha}^{-1}$  and 2.7 to 3.7  $\text{kg P ha}^{-1}$ . Median livestock densities (as measured by total livestock nutrient demand) declined over the study period ( $R^2=0.61$ ,  $p=0.0002$ ), yet maximum densities increased ( $R^2=0.94$ ,  $p<0.0001$ ), reflecting the rise of concentrated industrial animal agriculture. Importantly, total livestock populations for the region have not changed significantly over the study period. Rather, it is the shifting spatial distribution of these populations into smaller areas that is driving changes in nutrient inputs and possibly increasing the clustering of nutrient inputs as measured by Moran's I. Meanwhile, human population densities demonstrated the opposite pattern, where median human population density increased ( $R^2=0.98$ ,  $p<0.0001$ ), and the maximum human population density decreased ( $R^2=0.73$ ,  $p<0.0001$ ), suggesting declining urban populations and increased suburbanization or exurbanization, a trend shared with much of the Midwest and eastern U.S. [Brown *et al.*, 2005].

### 3.3.2. Crop Agriculture and Fertilizers

[35] The largest nutrient inputs to the NE during the twentieth century were to support agriculture—fertilizers,  $\text{N}_2$  fixation, and livestock feed (Figure 5). Figure 5 illustrates that accumulation and/or losses accounted for a large portion of nutrients added to agricultural crop systems as fertilizer and  $\text{N}_2$  fixation. Although total N inputs to the crop system did not change over time, fertilizer replaced  $\text{N}_2$  fixation as the dominant input to the crop system. The majority of N inputs to the crop system either accumulated in soils or were lost to runoff, leaching, or denitrification (Figure 5a). Inputs of P to crop systems were less than crop uptake from 1930 to 1940, meaning that farmers were mining soils for P. Because crop



**Figure 5.** Detailed nutrient inputs and their fates for crop, livestock, and human subsystems over time: (a) N inputs to and outputs from crop system. “Accumulation” in crop subsystem includes accumulation in soils (change in storage), or denitrification, leaching, and runoff losses (export from system); (b) P inputs to and outputs from crop system; (c) N inputs to and outputs from livestock system; (d) P inputs to and outputs from livestock system; (e) N inputs to and outputs from human subsystem; and (f) P inputs to and outputs from human subsystem. Note that scales are the same for each element. “M&D” indicates meat and dairy.

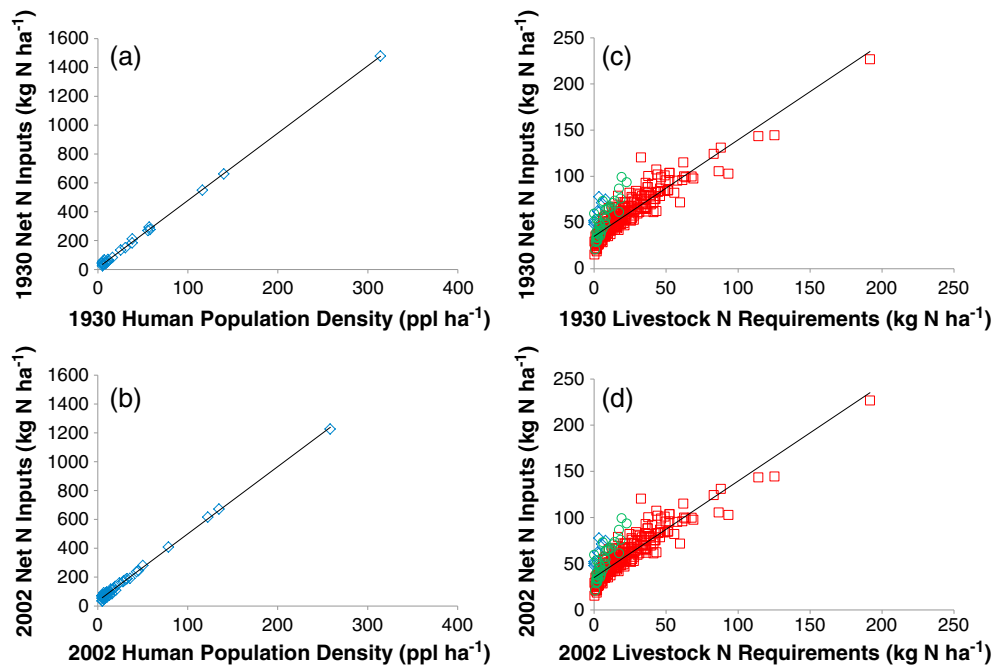
uptake of P did not change significantly over the study period, changes in P fertilizer use primarily affected the amount of P accumulating in soils and lost downstream as eroding soils (Figure 5b).

[36] Over the study period, N and P fertilizer use followed very different patterns (Figures 2a and 2b). We can understand the differences between N and P fertilizer use by comparing the absolute amount and stoichiometry of fertilizer nutrients to that removed from soils by crops. The ratio of fertilizer P inputs to P removed by harvested crops has moved toward 1 since 1970, indicating more efficient use of fertilizers (Figure 7). This pattern was not apparent for agricultural N inputs. N fertilizer inputs increased throughout the century, although until the mid-1990s inputs were lower than N removed by crops (Figure 7). Including biological  $N_2$  fixation pushed agricultural N inputs far above crop removal, and declines in N inputs during the second half of the century were directly related to reductions in cropland area rather

than reductions in N fertilizer use (Figure 7). Another way of understanding fertilizer use efficiency is to compare the stoichiometry of nutrients (i.e., N:P) added as fertilizer to the stoichiometry of nutrients removed as crops. Ideally, these would be equal, otherwise the nutrient added in excess would be unused and therefore vulnerable to downstream loss or, in the case of N, denitrification. In the NE, the N:P of fertilizer application was much lower than that of crop harvest during much of the study period (Figure 2c), indicating an over-application of P relative to N. The N:P ratios of fertilizer and crop harvest converge by the end of the century, indicating a more efficient application of fertilizer at the regional scale. When  $N_2$  fixation is included, however, the N:P of inputs was far in excess of that removed by crops, a possible contributor to N pollution in rivers—for example, throughout New England [Moore *et al.*, 2004].

[37] The increasing efficiency of P fertilizer use was driven by a confluence of factors: Better science allowed farmers to





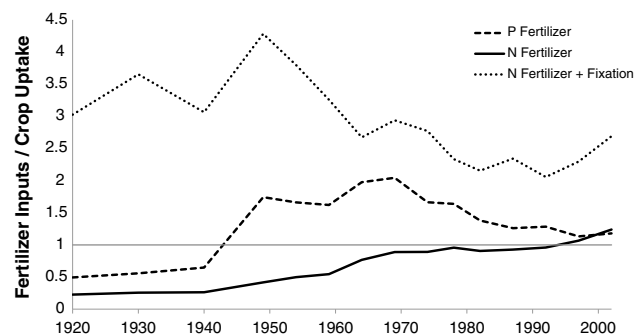
**Figure 6.** (a and b) Total net N inputs at the county level are strongly correlated with human population density for counties with population densities  $>5$  people  $\text{ha}^{-1}$ . (c and d) For counties with  $<5$  people  $\text{ha}^{-1}$ , livestock N demand is the best predictor of net N inputs. Blue colors indicate counties where net N inputs are dominated by human food and nonfarm fertilizer. Red colors indicate counties where net N inputs are dominated by livestock feed. Green colors indicate counties where net N inputs are dominated by fertilizer and  $\text{N}_2$  fixation.

calculate optimal rates of fertilization [e.g., Bray, 1945], and the availability of individual nutrient fertilizers rather than multielement fertilizers allowed farmers to apply fertilizers in ratios appropriate to their crops, soil, and climate. Previous over-fertilization meant that many soils had high levels of P that crops could mine [Parker, 1950; MacDonald and Bennett, 2009], and a major spike in the cost of fertilizer in the early 1970s acted as an incentive for farmers to use fertilizers judiciously [Stewart, 2004; Economic Research Service, 2011]. Fertilizer N was not unaffected by these changes, but the effects were less dramatic. Rather than a drop in fertilizer N use, we see a slight leveling off. One potential reason for continued use of N fertilizers is that N is prone to leaching and denitrification and therefore less likely to accumulate in soils, regardless of over-fertilization. Continued use of N despite increased concern for N pollution compared to P with regard to water quality [Kurtz, 1970] suggests that changes in society's environmental ethic were not important drivers of the changes in P fertilizer use during the 1970s.

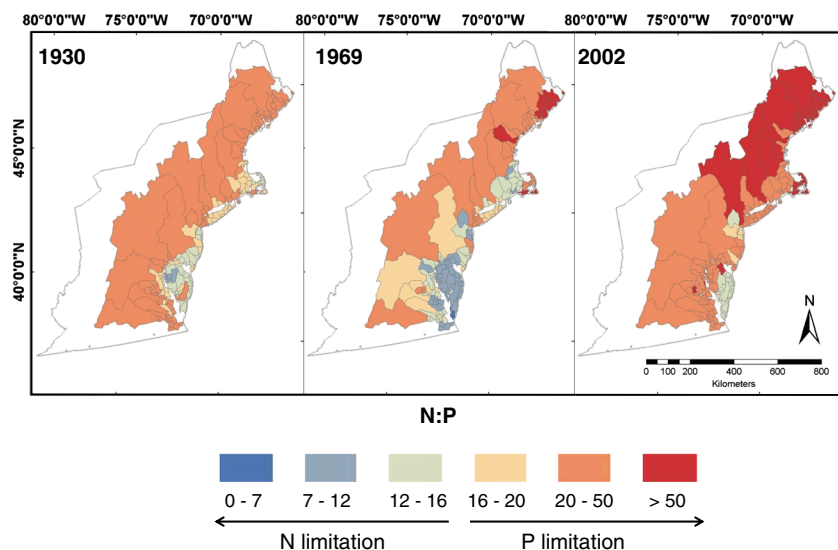
### 3.3.3. Nutrient Legislation

[38] Although agricultural fertilizer use was never directly regulated, detergent P and nonfarm P fertilizers have been subject to restrictive legislative controls. Most nutrient legislation during the latter part of the century focused on P reduction strategies and was to some degree successful at reducing P concentrations in streams and rivers [Lettenmaier et al., 1991; Litke, 1999; Lehman et al., 2009]. The legislative focus on P during the 1970s, to the exclusion of N, was an important reason for several of the divergent patterns of N and P inputs and was in part due to the scientific understanding of

nutrient limitation at the time [Howarth and Marino, 2006]. The science of the limnological tradition held that productivity in freshwater and marine systems was P limited, and therefore, the most effective strategy to reduce eutrophication was to reduce inputs of P. Although there was research demonstrating that many marine receiving waters were N limited [Ryther and Dunstan, 1971], water managers doubted the results and mistrusted the bioassay methods used in marine studies [Lee, 1973; Cloern, 2001; Howarth and Marino, 2006]. As a result, the contemporary knowledge of ecosystem function at the time had a strong influence on which pollution management strategies were pursued, with a long-term legacy effect on pollution patterns regionally.



**Figure 7.** Ratio of fertilizer inputs to nutrients removed in crop harvest for the NE over time. Points above the 1:1 line indicate over-application of fertilizer (and  $\text{N}_2$  fixation); points below the line indicate under-application of agricultural inputs.



**Figure 8.** Molar N:P of nutrient inputs for watersheds draining into the Atlantic Ocean in 1930, 1970, and 2000. Ratios are displayed relative to the Redfield ratio.

[39] Point sources of pollution were addressed in legislation before nonpoint sources because they were relatively easy to manage and their management had an identifiable impact on water resources [Litke, 1999; Carpenter *et al.*, 1998]. However, the NE hosts a remarkable example of effective nonpoint source pollution legislation in the regulation of nonfarm fertilizers. Nonfarm fertilizer was one of the most rapidly increasing inputs of N and P to the NE. Although still a small percentage of total N (2%) and of P (4%) inputs by 2002, nonfarm fertilizer increased substantially from 4% of total fertilizer P inputs in 1987 (when records began) to 10% in 2002 (and from 5% to 18% for N). Spatially, areas with concentrated nonfarm fertilizer inputs (e.g., suburban and urban areas) were distinct from areas with high fertilizer use for agriculture (data not shown). Although agricultural P fertilizer use is not regulated [Environmental Protection Agency, 1999; The Fertilizer Institute, 2003], there has been a recent emergence of P fertilizer bans for urban and suburban lawns due to eutrophication of local water bodies [e.g., Lehman *et al.*, 2009]. Legislation has emerged across spatial and political scales at the municipal, county, and state level. At this time, 11 states in the U.S., five of which are in the NE (Maine, Maryland, New York, New Jersey, and Virginia), have passed laws banning the use of P fertilizers for turf grass.

#### 3.3.4. Atmospheric N

[40] Some variations between trends in N and P are due to differences in their biogeochemical cycling potential. The N cycle has a large inert atmospheric component while the P cycle is geologic. These differences have major consequences for the stoichiometry of NE nutrient inputs. There are three major pathways by which humans convert inert  $N_2$  gas into reactive N species: (1) biological  $N_2$  fixation, (2) industrial  $N_2$  fixation (fertilizer production), and (3)  $NO_x$  production as a by-product of the combustion of fossil fuels. These types of human activities influence the N cycle without affecting P inputs. While industrial  $N_2$  fixation for fertilizer manufacture is tightly controlled, N deposition is an inadvertent result of human activity, and biological  $N_2$  fixation is indirectly controlled by farmers as a result of crop choices.

[41] We found a consistent pattern when considering only total direct inputs of N (i.e., no atmospheric sources): The stoichiometry (N:P) of inputs and absolute amounts of N matched nutritional needs (livestock and human requirements, crop uptake). The N:P of total inputs to crop systems was substantially higher than the N:P of crop uptake and the N:P of the total nutrient inputs for the region was higher than the N:P of any of the major consumers in the system (humans, livestock, crops; Figure 2c). This is evidence that the atmospheric component of N cycle in the NE has been poorly managed and that inputs of N from N deposition and  $N_2$  fixation have not been adequately accounted for by nutrient users. The lack of attention to atmospheric inputs of N has led to increased N:P of nutrient inputs at the regional scale. The excess N entering the system is then especially vulnerable to downstream losses because it is not needed by the systems to which it is applied, with severe consequences for downstream ecosystems.

#### 3.4. Potential Ecological Consequences

[42] Accounting for anthropogenic nutrient inputs is easiest within human boundaries, such as municipalities and counties, but nutrient inputs are transported by water downstream, and thus ecological effects must consider ecological boundaries, in this case watersheds. We calculated the N:P of nutrient inputs to 256 watersheds draining to the NE coast (Figure 8). These estimates do not account for processing (by ecosystems or technology such as waste water treatment) or transport processes that occur within the watershed and therefore ignore the large percentage of nutrient inputs that may be retained by watersheds [e.g., Seitzinger *et al.*, 2002; Hong *et al.*, 2012]. An additional caveat is the potential for land use legacies to have a strong effect on downstream loading [Foster *et al.*, 2003]. P cycles much more slowly than N due to binding with soils and sediments, and therefore watersheds may be more retentive of P than of N [e.g., Hong *et al.*, 2012]. As a result, the N:P of inputs for a year may not be a good predictor of the N:P of nutrients delivered downstream. P inputs from fertilizer to agricultural soils are likely to build

up over time [Dobermann and Cassman, 2002; MacDonald and Bennett, 2009; MacDonald et al., 2012]. Since soil P content is the best predictor of P transport downstream [Carpenter et al., 1998], it is likely that in agricultural areas, cumulative P inputs could be a better predictor of downstream P export than annual inputs. Finally, we calculated nutrient inputs on an annual basis, but riverine exports are likely to vary seasonally [Carpenter et al., 1998] due to seasonal use of nutrients by humans as well as variability due to runoff patterns. Previous research has shown significant seasonal fluctuations in nutrient limitation of aquatic systems [Howarth, 1988]. However, these estimates do provide a qualitative spatial and temporal assessment of the stoichiometry of nutrient inputs to coastal areas.

[43] The absolute amounts of nutrients entering coastal areas are critical for determining ecological effects. However, due to nutrient limitation, the ratios of elements are often just as, if not more important than, the total amounts. The ratios of nutrients entering estuaries and coastal areas can determine whether or not pollution will cause eutrophication and may cause significant shifts in phytoplankton community structure [Justic et al., 1995; Smith, 2003]. Although a comprehensive evaluation of the ecological consequences of coastal nutrient loading over time is beyond the scope of this work, we show that the N:P ratios of nutrient inputs have changed dramatically over time, with potentially important ecological consequences. Figure 8 illustrates changes in watershed stoichiometry from 1930 to 1970 to 2000 relative to the Redfield ratio (16:1), the theoretical ratio of N:P in marine phytoplankton and ocean waters [Redfield, 1958]. In 1930, there was a distinct pattern where larger, more inland watersheds had N:P greater than 16, and smaller coastal watersheds had N:P less than 16. There was also a latitudinal pattern, where coastal watersheds along Maine were great than 16, whereas watersheds along the coast from Massachusetts southward had inputs with an N:P of less than 16. The ratio of N:P decreased dramatically by 1970 due to increased P inputs as fertilizer and detergent. The N:P of nutrient inputs decreased for most watersheds, with the exception of northern watersheds in Maine and Cape Cod. More watersheds in 1970 experienced inputs with N:P less than the Redfield ratio compared to 1930. By 2002, however, there was a shift again in the opposite direction as P inputs decreased and atmospheric deposition of N increased. The ratios by the end of the century and for much of the northern part of the region reached an order of magnitude higher than the Redfield ratio. These findings are consistent with global trends [Peñuelas et al., 2012]. The latitudinal pattern in watershed N:P was strongest in 2002. This spatial pattern is likely related to latitudinal trends in atmospheric N deposition, as well as increases in livestock agriculture (associated with relatively low N:P demand; Figure 2c) in the southern portion of the study area. Although in general marine systems are currently considered N-limited, this ratio of nutrient loading could shift receiving systems from N to P limitation, depending on loading relative to water volumes and flows as well as cycling rates in receiving waters. This is not unprecedented. Billen et al. [2007] found that legislative P controls and uncontrolled increases in N loading to the Seine River in France led to a shift from N to P limitation in the marine system. N inputs from atmospheric deposition have also been found to shift freshwater systems from N to P limitation [Elser et al., 2009]. Shifts in nutrient limitation of NE coastal and freshwaters over time

and space not only have implications for ecosystems and the economies that depend on them but must also be taken into account when designing effective nutrient legislation.

### 3.5. Sources of Uncertainty and Limitations

[44] Due to the scope of our work, particularly its historical nature, we relied entirely on publically available data sets for our data sources and for calculating nutrient inputs associated with each. Here we discuss the sources of uncertainty and the resulting limitations of our research. The two main sources of uncertainty are those associated with the data themselves, including their spatial and temporal resolution, and the coefficients used to calculate the nutrient budgets.

#### 3.5.1. Uncertainty in Data Sources

[45] The majority of our data were obtained from the U.S. Census of Agriculture reported at the county scale. These data are self-reported, and therefore, there is a certain level of error that can be expected in these data. However, due to the large number of counties (437), we have confidence in the general spatial and temporal patterns generated by this resolution.

[46] There is also uncertainty associated with our N deposition estimates. We have the most confidence in our estimates based on NADP data, for which there were 41 data points available since 1978. Interpolating regional N deposition from this number of points certainly ignored smaller-scale variation in deposition rates. Jaworski et al. [1997] demonstrated that riverine N export from watersheds with minimal agricultural or urban inputs was strongly predicted by N deposition estimated from NADP data, suggesting that this resolution of data is appropriate for regional-scale studies. Uncertainty increases for earlier years, where data were limited or nonexistent. County-level deposition rates were estimated using multiple regressions (equations (1) and (2)), whereas total regional deposition rates were estimated from emissions data. These two methods yielded similar deposition estimates for the whole region from 1974 to 2002, but estimates increasingly diverged back in time, so that in 1930 our emissions-based deposition for the region was 0.92 Tg and our regression-based estimate was 0.39 Tg. These differences do lead to differences in total N input estimates of up to 20% (for 1930, estimated net N inputs are 2.11 Tg using spatial model and 2.64 Tg using the emissions model). However, temporal trends in atmospheric N inputs and total N inputs are robust to the N deposition model selection, as are trends in nutrient stoichiometry. N:P of nutrient inputs varied by as much as 7 (molar ratio, 1930) using different N deposition models, but the overall pattern was robust. N:P peaked in 1940 using both models, though N:P values were 47 using the emissions model and 40 using the spatial model. The lowest N:P value was in 1964 and was 22 using emissions data and 20 using the spatial model.

[47] A second source of uncertainty in our N deposition data was the calculation of dry deposition and organic N deposition. Estimates of dry deposition as a proportion of total deposition in the eastern U.S. range from 25% to 70% for  $\text{NO}_3^-$  and 2% to 33% for  $\text{NH}_4^+$  deposition [Bowen and Valiela, 2001] and are likely to be variable over space and time. However, since there are little data on how dry deposition varies over space, we used a consistent coefficient from the literature [Bowen and Valiela, 2001]. Spatial and temporal variation in dry deposition might have either damped or strengthened the patterns that we observed.

### 3.5.2. Uncertainty in Data Generated

[48] Nutrient demand, consumption, and production by crops, livestock, and humans likely varied over time and space during our study period as agricultural practices and human diets changed [Gerrior *et al.*, 2004; Metson *et al.*, 2012]. Because of data limitations and the scope of our research, we made the simplifying assumption that coefficients used to calculate input rates did not vary.

[49] Biological N<sub>2</sub> fixation rates range widely in the literature, though our estimates fall in the middle [Smil, 1999]. We calculated N<sub>2</sub> fixation based on the area of cropland planted in various crops. This is consistent with other nutrient inputs studies [e.g., Jordan and Weller, 1996; Boyer *et al.*, 2002; Hong *et al.*, 2012; Howarth *et al.*, 2012]. However, other research has suggested that N<sub>2</sub> fixation varies with crop yield [Herridge *et al.*, 2008], and therefore it is possible that N<sub>2</sub> fixation rates per area increased as yields increased over our study period. Similarly, we assumed that the nutrient content of crops remained constant over the study period. Fertilizer use typically increases not only the yield but also the nutrient content of crop plants [e.g., Lawlor, 2002], thus nutrient uptake per crop yield has likely increased over time. Since we used contemporary values of crop nutrient content, estimates of nutrient accumulation in agricultural soils are possibly underestimated during the early part of our study period, though the magnitude of this uncertainty is unknown.

[50] Fertilizer use rates from 1945 to 2002 were obtained from USGS reports [Alexander and Smith, 1990; Ruddy *et al.*, 2006] where county-level fertilizer use was estimated from county-level fertilizer sales. For earlier years (1930–1940), fertilizer use at the county level was estimated based on state fertilizer sales disaggregated to the county level using harvested crop area. If fertilizer use patterns varied within states, spatial patterns for these years could be stronger than our estimates.

[51] Livestock nutrient demand was estimated based on inventories and published coefficients for livestock nutrient requirements. However, livestock nutrient demand per animal likely increased over time due to changing agricultural practices, which would strengthen the spatial and temporal trends that we described. Similarly, human nutrient demand likely increased over the study period as protein consumption increased in the United States [Gerrior *et al.*, 2004]. Thus, our estimates of livestock and human nutrient demand are likely liberal in terms of inputs during the early part of our study period but conservative in terms of changes in inputs over time, particularly the last few decades (i.e., including variation in coefficients would likely strengthen the trends observed).

[52] In calculating the local supply of nutrients in human food and livestock feed, we assumed that a certain percentage of each crop type was used for food or feed (e.g., 4% of corn went to human food). These percentages are likely to vary over both time and space, especially as livestock husbandry practices have changed. Because the NE imported the majority of feed and crop food across the study period, we think that this is likely a small source of uncertainty. Furthermore, we did not account for pasture grazing as a source of livestock feed. This is likely a small source of local nutrients by the end of the century due to changing livestock raising practices, but may have been more important at the beginning of the study period.

[53] We assumed constant spoilage rates across food and feed types and over space and time. This assumption is consistent with previous anthropogenic nutrient budgeting literature [e.g., Boyer *et al.*, 2002; Hong *et al.*, 2011]. This spoilage rate includes many types of waste along the food production chain, including waste in production, transportation, retail, and consumer waste. The FAO publishes spoilage rates for various feed and food types that are higher and more variable than the rate we used (e.g., 20–60% [Gustavsson *et al.*, 2011]). However, given that spoilage rates likely vary over time and space, especially in response to changing agricultural, transport, and consumer practices, we think that a consistent value facilitates interpretation.

## 4. Conclusions

[54] Anthropogenic nutrient use is highly dynamic both spatially and temporally and responds to scientific understanding, policy changes, technology, and land-use and demographic changes. Over the twentieth century, we found that agriculture, and livestock agriculture in particular, was the major driver of spatial patterns of nutrient use. Livestock consumed the majority of nutrient inputs to the NE, and the spatial concentration of livestock populations over time drove changes in the spatial patterns of nutrient inputs. As a result, spatial and temporal changes in nutrient inputs mirror the history of agricultural policies which have shifted the locations of U.S. agriculture, particularly the movement of row crop agriculture to the west and the development of concentrated livestock agriculture in the NE. Similarly, human demographic trends—suburbanization in particular—led to increases in human food nutrient inputs across the region. These changes have led to major spatial and temporal changes in the stoichiometry of nutrient inputs, with important potential ecological consequences for N and P limitation of receiving waters.

[55] Future nutrient management strategies will need to take into account the multiple pathways through which humans affect nutrient inputs. Our study period included major developments in environmental legislation, including the pioneering Clean Air and Water Acts. Our results show that environmental regulations that regulate direct emissions have been successful in reducing some inputs of P, namely, detergents and nonfarm fertilizers. Agricultural fertilizers remain a major contributor to nutrient inputs in the NE and are a difficult management issue due to the distributed nature of the inputs, the lability and multiphase nature of N and P, and the difficulty of enforcement. However, we did find that P fertilizer use responded strongly to economic drivers, suggesting that indirect economic mechanisms may be a viable option for fertilizer use management. Management of N remains more difficult than of P due to the atmospheric component of the N cycle, highlighting the importance of including the atmospheric component in managing N inputs. Of strategic importance to regional nutrient management is the fact that N uptake by agricultural crops is much higher than N fertilizer additions, suggesting the critical importance of both managed applications (i.e., fertilizer) and N<sub>2</sub> fixation which today are equivalent to double crop needs. A complete accounting may facilitate fertilizer use management and minimize water quality management challenges associated with fertilizer application.

[56] Optimizing nutrient management to cobalance agricultural production and environmental protection remains a difficult task in the NE. Additional challenges will also be associated in refining the debate on carbon management, the use of cropland for biofuels, and preparing the region for future climate change. The variety of drivers of nutrient use presents a challenge for decision makers who must take in account the interactions between economics, biogeochemistry, technology, and policy. However, the diversity of drivers presents an opportunity as well—policy makers have many levers at their disposal beyond directly managing nutrient use. A historical approach can illustrate when and where different approaches may or may not succeed and facilitate a multifaceted approach to nutrient management that takes advantage of the multiple social, political, economic, and environmental drivers of human nutrient use.

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