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Reactive nitrogen inputs to US lands and waterways: how certain are we about sources and fluxes?

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Reactive nitrogen inputs to US lands and waterways: how certain are we about sources and fluxes?

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An overabundance of reactive nitrogen (N) as a result of anthropogenic activities has led to multiple human health and environmental concerns. Efforts to address these concerns require an accurate accounting of N inputs. Here, we present a novel synthesis of data describing N inputs to the US, including the range of estimates, spatial patterns, and uncertainties. This analysis shows that human-mediated N inputs are ubiquitous across the country but are spatially heterogeneous, ranging from < 0.1 to 34.6 times the background N input for individual water-resource units (8-digit Hydrologic Unit Codes). The Midwest, Mid-Atlantic, central California, and portions of the Columbia River valley currently receive the highest N loads. Major opportunities to advance our understanding of N sources can be achieved by: (1) enhancing the spatial and temporal resolution of agricultural N input data, (2) improving livestock and human waste monitoring, and (3) better quantifying biological N fixation in non-cultivated ecosystems.

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Reactive nitrogen (N) includes all biologically, photochemically, and radiatively active forms of N within Earth's atmosphere and biosphere. Anthropogenic activities have at least doubled annual input of N to global land surfaces as compared with N input during pre-industrial times, largely to support the food and fuel needs of the world's rapidly growing human population (Galloway *et al.* 2004). Although increased N input benefits society, it also causes many human health and environmental problems, including water and air pollution, loss of biodiversity, eutrophication of aquatic ecosystems, and increased emissions of nitrous oxide (N_2O), a potent greenhouse and ozone-depleting gas (SAB 2011). Balancing positive and negative impacts associated with human-mediated N inputs represents one of the most important challenges facing environmental managers this century (Schlesinger 2009).

In a nutshell:

- In the US, human activities have tripled continental-scale annual reactive nitrogen (N) loading over pre-European levels, with local increases up to 35-fold
- Synthetic fertilizer is the single largest anthropogenic N input to 41% of the water-resource units we analyzed, followed by atmospheric deposition and agricultural biological N fixation (33% and 22%, respectively)
- High agreement exists for datasets describing synthetic fertilizer or atmospheric deposition, but other N inputs are moderately to very uncertain
- Credible, appropriately scaled, spatially resolved estimates of N sources are necessary to support the formulation and implementation of effective N mitigation strategies

Sound information on N inputs and sources is critical for improving efficiency of N use in industrial and agricultural systems and for reducing environmental N pollution. Many global-scale assessments of N sources have offered strategies to manage human-mediated N inputs (eg Delwiche 1970; Smil 1999; Van Drecht 2009; Bouwman *et al.* 2009). However, continental, regional, and watershed-scale N assessments remain critical for management (Hong *et al.* 2011; SAB 2011; Swaney *et al.* 2012). The European Union recently completed a continental-scale N assessment and, consequently, has adopted a proactive, multi-scale N-management framework (Sutton *et al.* 2011). In contrast, most US-based assessments have been conducted at small-to-medium spatial scales, to assist with local-to-regional N pollution problems (eg David *et al.* 1997; Han and Allan 2008; TBEP 2012). This approach has shifted with the introduction of several coordinated efforts to assess national-level N inputs and environmental effects (Howarth *et al.* 2002; SAB 2011). The Science Advisory Board to the US Environmental Protection Agency (EPA) recently called for an integrated national N-management strategy, with an overall recommendation of reducing N inputs to the US by 25% (SAB 2011). This synthesis of datasets describing national N sources and their spatial distribution strongly supports this strategy. Many previous regional and national assessments have selected only one value to describe a specific N source and have failed to fully address the potential uncertainties inherent in estimates. Comparisons of multiple N-source datasets can provide insights into how well N inputs from specific sources are known and help prioritize future research, thereby improving decision-making related to N management at local, regional, and national scales.

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Here, we synthesize and analyze data on N sources to terrestrial and aquatic systems in the US to (1) evaluate existing estimates of the nation's N inputs, highlighting their range of variation and underlying uncertainties; (2) provide spatial representation of all N inputs to those systems; and (3) identify information gaps and suggest ways to improve domestic N input data.

Data described herein form the basis of a national N-source inventory, which includes both spatial and non-spatial data obtained from surveys, direct measurements, and model outputs, and will be modified and expanded as new N input datasets are added. The national N-source inventory is currently available at http://www.epa.gov/wed/pages/research/nitrogen/us-national-nitrogen-inventory-20121207.xls

A primer on N inputs to US lands and waterways

Domestic N inputs to terrestrial and aquatic systems involve both "new" and "recycled" N. New N inputs originate directly from the conversion of N_2 gas (a non-reactive form of N that constitutes 78% of the atmosphere) to reactive N. This conversion requires a large amount of energy, and occurs naturally via lightning strikes and biological N fixation (BNF), by either free-living or symbiotic bacteria (Galloway *et al.* 2004). In contrast, recycled N inputs entail the redistribution of N through transfers in air, water, and organisms (eg livestock manure disposal, release of wastewater to surface waters, and, to an extent, atmospheric N deposition).

Technological advances in the past 200 years have introduced or expanded three human-mediated inputs of new N to US lands: (1) cultivation of crops that have symbiotic relationships with N-fixing bacteria (agricultural BNF), (2) fossil-fuel-mediated creation of N for synthetic fertilizers and industrial products, and (3) deposition of reactive N created as a byproduct during fossil-fuel combustion. New N originating from anthropogenic activities cascades through air, land, and water, where it can have both positive (eg enhanced crop growth) and negative (eg air pollution, greenhouse-gas [N_2O] emissions, groundwater contamination, and coastal eutrophication; Galloway *et al.* 2003) effects.

Major human-mediated inputs of recycled N in the US include the deposition of livestock manure during grazing, application of livestock manure from Confined Animal Feedlot Operations (CAFOs) to croplands, effluent discharge of wastewater, and the widespread deposition of N compounds volatilized from agricultural and urban areas. Trade can also introduce or remove N in synthetic fertilizer, industrial N, or food and feed products.

Assembly and analysis of the US national N-source inventory

We applied queries to ISI Web of Knowledge and Google Scholar using relevant keywords, including: "nitrogen", "N", "deposition", "agriculture", "fertilizer", "manure", "fixation", "biological N fixation", "BNF", "industrial", "livestock", "input", and "United States" to search for peer-reviewed literature describing N inputs to the US. We also acquired datasets from several private groups and government agencies. Detailed descriptions of data analysis and the complete national N-source inventory are provided online (WebPanel 1; WebTable 1). All estimates in this paper refer to the conterminous US.

Different N-source estimates were often derived from a single dataset. We therefore cannot provide descriptive statistics for N-source estimates because doing so would violate the assumption of statistical independence. Instead, we present the range of estimates and offer a "best available estimate", which was determined as the estimate with the most robust underlying data. If multiple estimates were of similar quality, we chose the one with the widest spatial and temporal coverage for the study period (1990s–2000s).

For spatial N input data, we selected the best available spatial data according to the criteria described above and scaled N inputs to the resolution of 8-digit Hydrologic Unit Codes (HUC-8s), which are unique identifiers – created by the US Geological Survey – of specific drainage areas in the US and are commonly used in water resource management (Seaber *et al.* 1987). Although N inputs vary at spatial scales finer than those characterized by HUC-8s, the resolution of HUC-8s provide good graphical representations for national analyses. Finer-scale-resolution estimates, where available, have been included in the national N-source inventory (WebTable 1).

Background N input

BNF in non-cultivated lands accounts for nearly all of the new background N input to the US in pre-European and contemporary (1990s–2000s) times (Figures 1 and 2; Table 1). The most robust calculation of pre-European non-cultivated BNF is 7.9 teragrams of N per year (Tg N yr^{-1}, where 1 Tg = 1×10^{12} g), derived from the correlation between BNF rates for specific terrestrial ecosystem types and evapotranspiration (Table 1; WebPanel 1; Cleveland *et al.* 1999). The amount of N fixed by lightning strikes is small by comparison (< 0.1 Tg N yr^{-1}; Table 1; Galloway *et al.* 2004). The range of pre-European natural BNF is 3.8–12.7 Tg N yr^{-1}, based on six estimates derived from a literature review (Cleveland *et al.* 1999) and scaled to the US by terrestrial ecosystem type or evapotranspiration (Table 1; WebPanel 1).

The best available estimate of current, non-cultivated BNF in the US is 3.6 Tg N yr^{-1}. This value is calculated using terrestrial-ecosystem-specific BNF rates and excludes agricultural and urban land cover types (Bouwman *et al.* 2009). This estimate, when as compared with the pre-European estimate, suggests that conversion of natural landscapes to agriculture and urban areas has reduced non-cultivated BNF in the US by >50%.

Nevertheless, the range of estimates for current non-cultivated BNF is large (0.5–12.2 Tg N yr^{-1}; Figure 1; Table 1). Minimum and maximum estimates for this range are derived from Jordan and Weller (1996), who applied two different combinations of uniform BNF rates over large areas of the country (WebPanel 1).

Estimates of non-cultivated BNF at the national level remain highly uncertain because of several factors, including (1) the difficulty of measuring BNF in field settings, (2) the consequent lack of empirical field data, and (3) an incomplete understanding of controls on BNF (Cleveland *et al*. 1999; Vitousek *et al*. 2002; Herridge *et al*. 2008). Spatial data that describe the distribution of N_2-fixing plants are currently unavailable for most of the US; such data could greatly improve estimates of non-cultivated BNF.

■ New human-mediated N inputs

Using the most robust available datasets, we suggest that human-mediated N input to the US is currently 26.9 Tg N yr^{-1} (Figure 1; Table 2). However, total new N input could be as little as 14.9 Tg N yr^{-1} or as much as 35.3 Tg N yr^{-1} (Figure 1; Table 1). Our analysis suggests that human-mediated N inputs to the US are currently more than three times as large as pre-European background N inputs. This human-mediated increase above background can range from one- to nine-fold, depending on what combinations of datasets are used (Figure 1). Our ranking of new, human-mediated N input by magnitude for the 1990s–2000s, from largest to smallest, is: (1) synthetic fertilizer, (2) agricultural BNF, (3) N fixed for industrial products, and (4) atmospheric N deposition (Figure 2; Table 2).

Synthetic N fertilizer

We estimate current annual synthetic N fertilizer input as 10.9 Tg N yr^{-1}, based on average (1990–2001) annual US county-level N fertilizer applications (Figure 2; Table 2; Ruddy *et al*. 2006). Synthetic N fertilizer input could range from 7.9 to 13.0 Tg N yr^{-1}, according to eight additional estimates for the study period (Table 2), but the most probable range of inputs is 10.9–13.0 Tg N yr^{-1} because the lower-bound estimate (7.9 Tg N yr^{-1}) includes only 80% of US croplands (Potter *et al*. 2006). Given that this range of estimates is similar to the range of inter-annual variability for individual estimates during the study period (1–4 Tg N yr^{-1}; Ruddy *et al*. 2006; Kelly and Matos 2008; FAO 2011; USDA-ERS 2011), disagreement between national-level estimates for synthetic N fertilizer may be attributed mostly to differences in the period of reporting.

We consider synthetic N fertilizer to be the best-documented N input in the US because of the consistent record-keeping and frequent compilation of these data. Improved information for rates and timing of fertilizer applications to specific crop types and to non-agricultural lands, however, is still needed. Fertilizer applications to non-agricultural lands (eg residential lawns) are poorly recorded; current estimates rely on weak correlations between fertilizer use and population density (Ruddy *et al*. 2006).

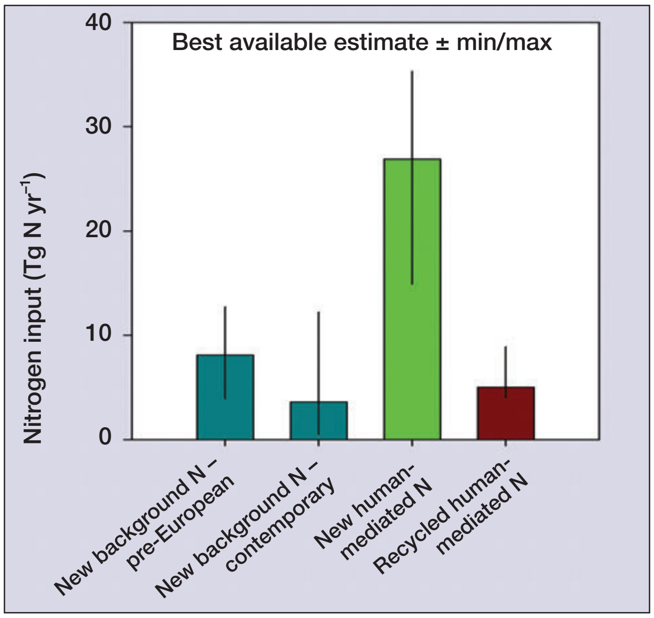

Figure 1. *Nitrogen (N) input to the conterminous US during the 1990s–2000s. Bars represent combinations of best available estimates for individual N-source terms (see Tables 1–3); whiskers display the range between minimum and maximum estimates based on all possible combinations of datasets.*

Agricultural BNF

The best available estimate of current agricultural BNF in the US is 7.7 Tg N yr^{-1}, originating from crop-specific areal BNF rates (Figure 2; SAB 2011), yet the range of

Table 1. Estimates of new background N inputs to the conterminous US

New background N source	*Tg N yr^{-1}*	*Time period*	*Source*
Non-cultivated BNF	3.9[a]	Pre-European	Cleveland *et al*. (1999)[c]
	3.9[b]	" "	" "
	7.9[b]	" "	" "
	8.1[a]	" "	" "
	12.0[b]	" "	" "
	12.7[a]	" "	" "
Non-cultivated BNF	0.5	Early 1990s	Jordan and Weller (1996)[c]
	3.6	2000	Bouwman *et al*. (2009)
	6.4	Early 2000s	SAB (2011)
	12.2	Early 1990s	Jordan and Weller (1996)[c]
Atmospheric N deposition	***<0.1***	Constant	Galloway *et al*. (2004)

Notes: [a]Based on the distribution of ecosystem types in the conterminous US (WebTable 2); [b]Based on correlation between non-cultivated BNF for ecosystem types and evapotranspiration; [c]Multiple estimates provided by one reference (see WebPanel 1 for details). BNF = biological N fixation. Bold italics identify the best available estimates.

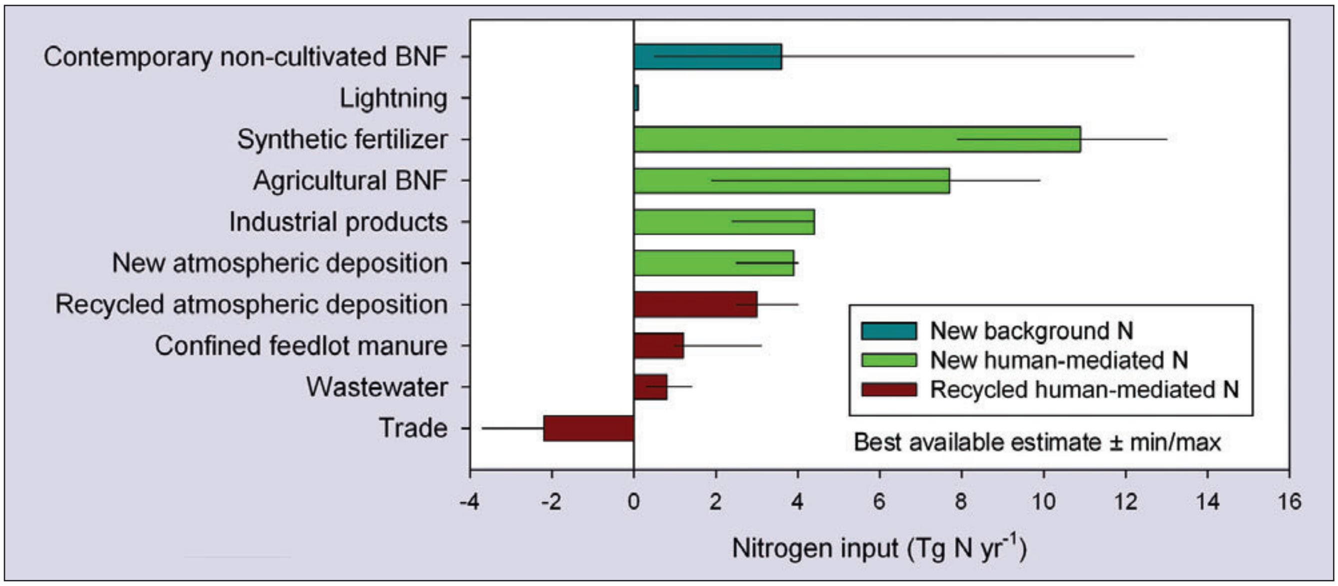

Figure 2. *Compiled estimates of individual N inputs to the conterminous US for the 1990s to the early 2000s. BNF = biological N fixation. Bars are best available estimates ± minimum and maximum of all available estimates. See Tables 1–3 for data used in the graph.*

estimates suggests that high uncertainty surrounds this important N source (1.9–9.9 Tg N yr^{-1}; Table 2). Some variability may be due to the 30% increase in soybean (*Glycine max*) production that took place between 1990 and 2010 (USDA 2010), although stark differences in the methods used in the calculation of agricultural BNF also contribute to the wide range of estimates (Herridge *et al.* 2008; WebPanel 1). Improvements in calculating this large and uncertain source would substantially improve understanding of anthropogenic N inputs.

Table 2. Estimates of new human-mediated N inputs to the conterminous US

New human-mediated N source	*Tg N yr^{-1}*	*Time period*	*Source*
Synthetic fertilizer	7.9	1997[a]	Potter *et al.* (2006)
	10.9	1990–2001	Ruddy *et al.* (2006)
	11.0	1990–2009	FAO (2011)
	11.0	1990–2009	IFA (2011)
	11.0	1990–2007	USDA-ERS (2011)
	12.1	2000	Bouwman *et al.* (2009)
	12.2	2000	Liu *et al.* (2010)
	13.0	1990–2003	Kelly and Matos (2008)
Agricultural BNF	1.9	2000	Bouwman *et al.* (2009)
	4.4	2000	Liu *et al.* (2010)
	4.7	Mid 1990s	Smil *et al.* (1999)
	5.9	2000	Howarth *et al.* (2002)
	6.6	Early 1990s	Jordan and Weller (1996)
	7.7	2002	SAB (2011)
	9.9	1997[a]	Potter *et al.* (2006)
Industrial products	2.4	1990–2003	Kelly and Matos (2008)
	4.4	2002–2009	FAO (2011)
Atmospheric N deposition[b]	2.7	1978–1994	Holland *et al.* (2005)[c]
	3.3	1978–1994	Holland *et al.* (2005)[c]
	3.6	1990–2001	Ruddy *et al.* (2006)
	3.7	Early 1990s	Jordan and Weller (1996)
	3.9	2002	USEPA (2010a)
	4.3	2000	Bouwman *et al.* (2009)
	4.4	1993	Dentener (2006)
	8.0	1997	Howarth *et al.* (2002)

Notes: [a]Represents 80% of cropland in the conterminous US; [b]Assumes organic N is 30% of total atmospheric N deposition (Neff *et al.* 2002; SAB 2011; see WebPanel 1 for details); [c]Multiple estimates provided by one reference (see WebPanel 1 for details). BNF = biological N fixation. Bold italics identify the best available estimates.

Industrial products

We suggest 4.4 Tg N yr^{-1} as the best available estimate of new N created for industrial products, derived from average non-fertilizer N use for 2002–2009 (Figure 2; Table 2; FAO 2011). One other estimate indicates that 2.4 Tg N yr^{-1} is currently fixed for the industrial production of plastics, synthetics, and explosives in the US (Table 1; Kelly and Matos 2008). Although industrial N fixation is a substantial source of new N, the amount of fixed N that enters the environment is currently unknown. The long-term fate and potential ecological impacts of N contained in manufactured goods also warrant further study.

Atmospheric deposition

Current deposition of atmospheric N derived from fossil-fuel combustion (new N) to the US is 3.9 Tg N yr^{-1} (Figure 2; Table 2). This estimate originates from Community Multiscale Air Quality (CMAQ) model output for deposition of oxidized inorganic N in 2002 (USEPA 2010a), and is adjusted upward by 30% to account for deposition by organic N compounds (WebPanel 1; Neff *et al.* 2002). The corresponding range of estimates (n = 8) is 2.7–8.0 Tg N yr^{-1}. Estimates developed specifically for the US (Jordan and Weller 1996; Howarth *et al.* 2002; Holland *et al.* 2005; Ruddy *et al.* 2006; USEPA 2010a) are consistently lower (by 10–66%) than estimates derived from the global TM3 model (Howarth *et al.* 2002; Dentener 2006; Bouwman *et al.* 2009). A decline in emissions of new N to the atmosphere has occurred over the study period, probably as a result of the implementation of the 1990 amendments to the US Clean Air Act (NAPAP 2005; USEPA 2008).

One way to improve estimates of new atmospheric N deposition would be to expand monitoring and modeling of organic N deposition, which is broadly characterized here as 30% of total N deposition (Neff *et al.* 2002). However, organic N deposition patterns across space are highly uncertain N (Neff *et al.* 2002).

Recycled human-mediated N input

Combined N input to the US from recycling and waste disposal processes currently amounts to 5.0 Tg N yr^{-1} according to the best available datasets (Figure 1; Kellogg *et al.* 2000; Van Drecht *et al.* 2009; USEPA 2010a). The range of recycled N input estimates is 4.0–8.9 Tg N yr^{-1} (n = 13 estimates; Table 3). Best available estimates for individual recycled N sources, in order of decreasing magnitude, are: (1) atmospheric N deposition, (2) livestock manure, and (3) wastewater N (Figure 2).

Atmospheric deposition

We estimate deposition of recycled N to the US as 3.0 Tg N yr^{-1}, according to CMAQ model output (Table 2; Figure 2; USEPA 2010a), although it could range from 2.7–4.4 Tg N yr^{-1} (n = 6 estimates; Table 3). Many of the same patterns and issues related to atmospheric deposition of new N also apply to recycled N deposition; these include higher estimates from global models relative to those for the US (23–39% higher; Table 3), a sparse monitoring network, and lack of information on the organic fraction of N deposition. Nevertheless, several issues associated with this source are unique. Modeling ultimate sources of recycled N deposition remains challenging for several reasons, including uncertainties associated with livestock manure disposal and N volatilization following synthetic N fertilizer application to fields (WebPanel 1; Holland *et al.* 2005; USEPA 2010a). Improving our ability to attribute human-mediated sources of recycled N deposition is important because emissions of recycled N continue to increase across the nation, despite reductions in other emission sources (USEPA 2008).

Livestock manure

The best available estimate of N input to the US from the land application of livestock manure produced on CAFOs is 1.2 Tg N yr^{-1} (Figure 2; Table 3); during the study period, the associated range of estimates is 1.0 to 3.1 Tg N yr^{-1} (Table 3). The spatial allocation of the actual amount of CAFO manure N distributed on agricultural lands remains uncertain, in that estimates provide only a potential manure N input estimate to agricultural lands. Improved on-farm reporting of manure production and dispersal would facilitate assessing the amount and timing of CAFO manure N application to agricultural lands.

Approximately 3.2 Tg N yr^{-1} is derived from manure N deposited during grazing (Kellogg *et al.* 2000; Ruddy *et al.* 2006). However, this estimate does not account for N lost to the atmosphere via ammonia volatilization, and inclusion of grazing-related manure would double-count atmospheric deposition of recycled human-mediated N. Much of the manure N deposited during grazing probably represents local recycling of new N (BNF, atmospheric deposition, and fertilizer) taken up by plants, while an unknown portion may derive from N compounds added to supplement livestock feed (Stanton and Whittler 2006). National-level and spatially explicit estimates of volatilization-corrected manure deposited during grazing would improve our understanding of N inputs and associated effects related to livestock waste disposal across the US.

N in wastewater

Point-source, non-industrial wastewater N input to the US during the 1990s–2000s is 0.8 Tg N yr^{-1}, ranging from

Table 3. Estimates of recycled human-mediated N inputs in the conterminous US

Recycled human-mediated N source	*Tg N yr^{-1}*	*Time period*	*Source*
Atmospheric N deposition[a]	2.7	1978–1994	Holland *et al.* (2005)[b]
	3.0	2002	USEPA (2010a)
	3.3	1978–1994	Holland *et al.* (2005)[b]
	4.3	2000	Bouwman *et al.* (2009)
	4.4	1993	Dentener (2006)
CAFO[c] manure	1.0	1997	Potter *et al.* (2006)
	1.2	1992, 1997	Kellogg *et al.* (2000); Ruddy *et al.* (2006)
	2.3	2000	Liu *et al.* (2010)
	3.1	1997	Howarth *et al.* (2002)
Point-source wastewater	0.3	2007	USEPA (2010b)
	0.8	2000	Van Drecht *et al.* (2009)
	1.3	2002	SAB (2011)
	1.4	1997	Howarth *et al.* (2002)

Notes: [a]Assumes organic N deposition is 30% of total N deposition (Neff *et al.* 2002; SAB 2011; see WebPanel 1 for details); [b]Multiple estimates provided by one reference (see WebPanel 1 for details); [c]Confined Animal Feedlot Operations. Bold italics identify the best available estimates.

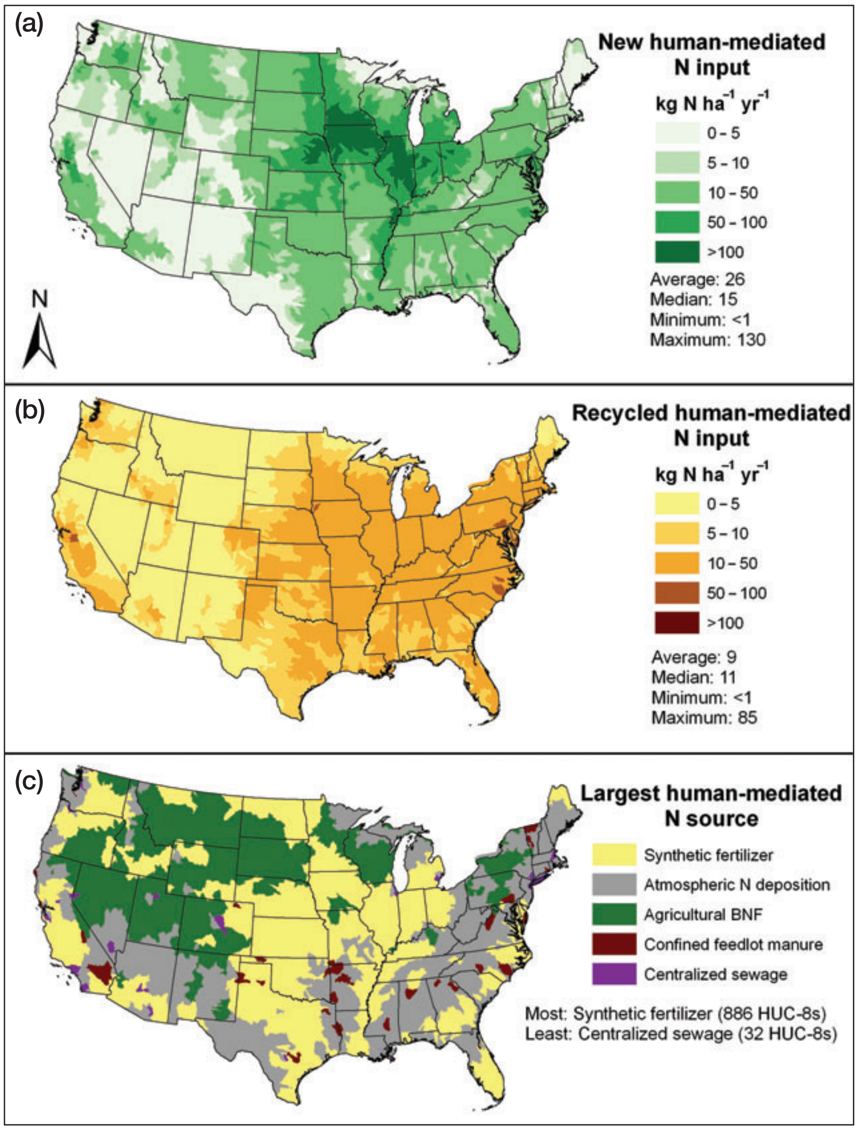

Figure 3. *Combinations of best available estimates for spatial distribution on N input to 8-digit Hydrologic Unit Codes (HUC-8s) in the conterminous US for the 1990s–2000s. Estimates of (a) new human-mediated N input, (b) recycled N input, and (c) single-largest human-mediated N source (new or recycled). WebPanel 1 and WebFigure 1 describe spatial datasets used to construct maps. Deposition of ≤1 kg oxidized N ha^{-1} yr^{-1} was assumed to originate from background sources. Uncertainties exist for all N-source estimates, with N inputs from agricultural BNF, CAFO manure, and centralized sewage among the least certain (see Figure 2).*

0.3–2.0 Tg N yr^{-1} depending on which data source is used. Point-source datasets differ regarding assumptions about the calculation of human N excretion and wastewater treatment (Table 3). N removal is most efficient when wastewater undergoes a tertiary nitrification/denitrification step, which results in the removal of up to 90% of N in wastewater as N_2 and N_2O gases (Scheele and Doorn 2001). Wastewater treatment plants that carry out tertiary treatment serve less than 1% of the US population (USEPA 2011). Wastewater is a small flux, but point-source reductions can be disproportionately important for water quality because wastewater is released directly into water bodies.

Point-source wastewater N input in the US can also include industrial effluent. Although the availability of point-source data in specific regions or for specific facilities has recently expanded (USEPA 2010b; Maupin and Ivanhnenko 2011), the last complete national estimate of N input from all sewage and industrial point sources (1.4 Tg N yr^{-1}) occurred outside our study period, in 1978 (Gianessi and Peskin 1984). Availability of N data from industrial and municipal effluent in the EPA's Permit Compliance System remains inconsistent, despite calls to improve reporting (NITG 2009) and advances in data access (USEPA 2010b).

Wastewater N can also enter ecosystems via septic tank leaching and land application of treated sewage. Of the estimated maximum 0.4 Tg N yr^{-1} that enters septic treatment systems (assuming minimal, non-sanitary disposal of waste), 0.2 Tg N yr^{-1} could currently leach from US septic systems. The EPA has compiled data on land application of treated sewage and suggests that ~0.1 Tg N yr^{-1} is currently applied to land surfaces (USEPA 2008).

Other important land and water N fluxes

A net export of 2.2 Tg N yr^{-1} is associated with international trade (mainly as grain; SAB 2011). While US product-based N export are currently larger than N imports, approximately 40% of synthetic fertilizer applied in the country originates from overseas markets (Kelly and Matos 2008).

Release of N from N-rich sedimentary rocks may represent another important input to terrestrial and aquatic environments (Morford *et al.* 2011). However, the spatial extent of geologic N and the relative importance of this source have yet to be explored in the US.

Spatial distribution of N inputs and sources

The national N-source inventory provides the basis for an integrated spatial representation of the total N input, dominant N sources, and degree of human alteration to N inputs across the conterminous US. The average input of new human-mediated N to hydrologic units (HUC-8s) in the US is 26 kg N ha^{-1} yr^{-1}, with a minimum and maximum of <1 kg N ha^{-1} yr^{-1} and 130 kg N ha^{-1} yr^{-1}, respectively (Figure 3a). The average input of recycled human-mediated N to HUC-8s is 9 kg N ha^{-1} yr^{-1}, ranging from <1 kg N ha^{-1} yr^{-1} to 85 kg N ha^{-1} yr^{-1} (Figure 3b).

Across the conterminous US, synthetic N fertilizer and atmospheric N deposition are the largest and second-largest overall human-mediated N sources (the single-largest sources in 41% and 33% of HUC-8s, respectively; Figure 3c). Atmospheric N deposition represents the single-largest source in many non-agricultural areas (eg portions of the East Coast, the Upper Great Lakes region, the Southwest, and the Pacific Northwest; Figure 3c). Agricultural BNF has the third-largest extent (the single-largest source in 22% of HUC-8s), dominating portions of the Northeast, Upper Midwest, Northwest, Southwest, and Intermountain West (Figure 3c). As for the remaining sources, distribution of manure N from CAFOs to adjacent agricultural lands (mainly in areas with high livestock population densities, such as eastern North Carolina, northern Georgia, and western Arkansas) and wastewater N (which can be important in densely populated urban areas) were the largest N sources to the landscape in only 2% and <2% of HUC-8s, respectively (Figure 3c and WebFigure 1). Spatial distribution of the rates of human-mediated N inputs are provided in WebFigure 1.

Human-mediated N inputs are present in every HUC-8 in the US; the magnitude of those inputs relative to pre-European-settlement background levels is spatially variable (Figure 4; WebFigure 1). Humans have doubled total new N inputs to nearly 60% of HUC-8s in the US, and have increased rates of N inputs in 15% of HUC-8s by more than five-fold over pre-European-settlement estimates (Figure 4a). The highest rates of N input occur in agricultural regions of the Midwest, the Mid-Atlantic, California, Idaho, and eastern Washington State (Figure 4a). As for recycled human-mediated N inputs, they are at least equivalent to pre-European-settlement input of background N for 19% of HUC-8s (Figure 4b) and are highest in densely populated urban areas and in agricultural areas with substantial animal production in the Mid-Atlantic, the Southeast, the upper Midwest, the southwestern Great Plains, southern Idaho, and southern California (Figure 4b).

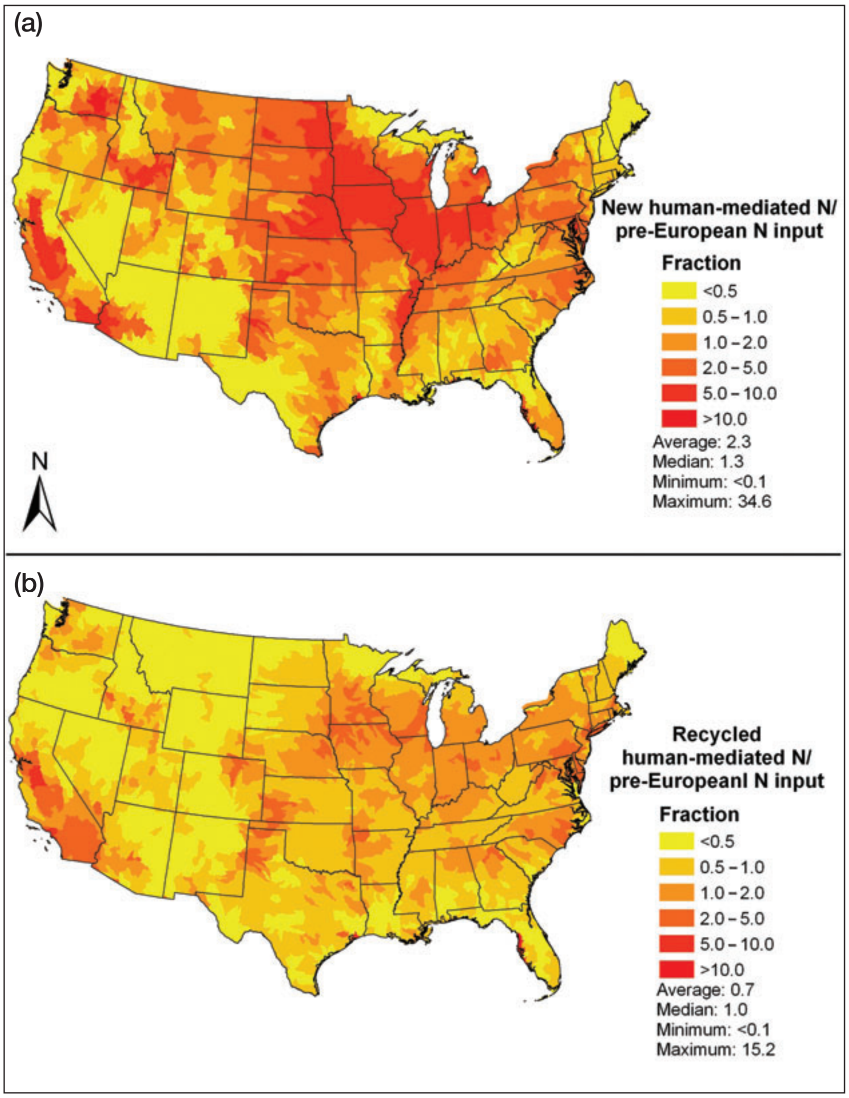

Figure 4. *Comparisons of background N input in pre-European times with (a) new human-mediated N input and (b) recycled N input to the conterminous US for the 1990s–2000s at the spatial resolution of 8-digit Hydrologic Unit Codes. See WebPanel 1 and WebFigure 1 for methods and data. Uncertainties exist for both human-mediated and background N input estimates, with estimates of non-cultivated BNF in pre-European times being the least certain of all N sources (see Table 1).*

There are two important caveats for our spatial representation of N inputs. First, a great deal of uncertainty surrounds estimates of non-cultivated BNF and many of the human-mediated N sources, particularly for agricultural BNF, CAFO-manure N, and wastewater N. This uncertainty influences the determination of dominant human-mediated sources at the resolution of an individual HUC-8. Second, identifying the single-largest N source does not mean that other N sources are unimportant. For example, in nearly 60% of HUC-8s where agricultural BNF was the single-largest N source, the difference between fertilizer application and agricultural BNF rates was small (< 5 kg N ha^{-1} yr^{-1}). Both caveats highlight the need for improved information and spatial resolution of background and human-mediated N sources.

Conclusions

This synthesis and analysis expands on previous studies of N cycling at regional and national scales. In addition to confirming that humans have dramatically altered the magnitude of N inputs across the nation, this national N-source inventory provides new insights regarding the precision and spatial patterns of current N inputs to the US. Best available data suggest current activities associated with food production and energy consumption have tripled the annual input of reactive N to land and waterways over pre-European-settlement N input at the national level, while all available data indicate that a two- to nine-fold increase is possible. Most datasets identified synthetic N fertilizer as the single-largest source of N in the US. The national ranking of other N sources is much less certain, owing to the wide variation in estimates resulting from our inability to precisely quantify specific N-source terms, in particular agricultural BNF, non-cultivated BNF, livestock manure, and wastewater inputs.

Available data suggest that human alteration of the N cycle in terrestrial and aquatic systems is widespread, occurring across all HUC-8s within the conterminous US, although the magnitude of alteration is spatially heterogeneous. Some hydrologic units have probably experienced more than a 35-fold increase in N loading in the 20th and 21st centuries, whereas other areas have experienced only minor increases in N inputs. Some of the most highly altered regions include the Upper Midwest, Mid-Atlantic, central California, and the interior Pacific Northwest. Synthetic N fertilizer or agricultural BNF dominate total N inputs in many of these areas, although manure N from CAFOs can be a major N source in parts of the Mid-Atlantic and California. Regions where N inputs have been least modified include sparsely populated regions like northern Maine, the northern Great Lakes region, the northern Rocky Mountains, and the central Great Basin. Human-mediated N inputs to these areas are probably mostly through atmospheric deposition.

Several key areas for improvement to our understanding of N sources and their spatial distribution are identified here:

- *Information regarding land-cover data and quantitative linkages between land-cover and synthetic N fertilizer application data.* While synthetic fertilizer information is tracked sub-annually and annually, land-cover data are reported less frequently, limiting the temporal resolution at which fertilizer inputs can be estimated. Given that synthetic N fertilizer is the single-largest N source to large areas of the country, this is a pressing need.
- *National, spatially explicit estimates of BNF in agricultural systems.* We suggest that crop yield-based modeling linked to data on soil N content and local climate is a worthwhile approach. Better information on agricultural BNF is needed because it is an important N source in several areas of the country.
- *Information and models of BNF in non-cultivated ecosystems.* This is another potentially large and uncertain N source to the nation. Information on the spatial distribution of N_2 fixers and factors that affect BNF rates is required.
- *National monitoring of the disposal of livestock manure, including the amount and timing of manure deposited to fields, pastures, and rangelands.* Current estimates rely on equations that convert agricultural census data regarding livestock to manure inputs but do not fully describe the application rates and timing to all US cropland types. Although relatively small when compared with other agricultural N sources, livestock manure N inputs can be locally important and are linked to large-scale air- and water-quality problems (Bouwman *et al.* 2005; USEPA 2011).
- *Complete and publicly accessible national reporting of N discharge from wastewater treatment plants (municipal and industrial).* Although these N inputs are generally monitored, regional methodological differences in monitoring and reporting complicate developing a reliable, national-scale representation of these N sources.
- *Monitoring and modeling of the organic fraction of atmospheric N deposition.* Organic N is an important but poorly constrained component of N deposition.

Spatial analysis of N sources could contribute to regionally tailored management that could help achieve the recommended 25% reduction in N loading to the nation (SAB 2011). Our analysis suggests that management to reduce N loading to the upper Midwest landscape should include consideration of the inputs and effects of both synthetic fertilizers and agricultural BNF. Managers in regions dominated by recycled N sources should concentrate on manure management in livestock-dominated regions and on sewage treatment technologies in densely populated areas. Improvements to N-source data at all levels of spatial and temporal resolution would help to rationally target future research investments (Hufnagl-Eichiner *et al.* 2011) and to benefit national N management efforts, including traditional regulatory approaches used by states and the Federal Government and emerging N management approaches such as markets for nutrient trading and ecosystem services (Compton *et al.* 2011).

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WebPanel 1. Synthesis and analysis of the US nitrogen inventory

More than 913 000 individual peer-reviewed publications, referring to nitrogen (N) input in the US, were generated by our search engine-based queries. The vast majority (>99.9%) of papers identified in this way were too focused in scope or geographic area to be included in this synthesis. National-level N input estimates were, for the most part, taken directly from peer-reviewed literature or government publications (for a complete list, see WebTable 1). However, in some cases (eg for several estimates of non-cultivated biological N fixation [BNF], sewage wastewater, and inorganic N deposition) peer-reviewed estimates/models were re-analyzed to extract US-specific information. Here, we provide a description of the methods used for (1) extrapolation of N input estimates/models that were developed at scales smaller than the conterminous US, or (2) subsampling of N input estimates/models that were developed for scales larger than the continental US.

Non-cultivated BNF

Jordan and Weller (1996) provided two estimates of non-agricultural BNF (Table 1). For the lower estimate (0.5 Tg N yr^{-1}), we assumed a constant non-agricultural N fixation rate of 1 kg N ha^{-1} yr^{-1} for US Geological Survey (USGS) Water Resource Regions (2-digit Hydrologic Unit Codes [HUC-2s]) east of the Mississippi River and 0.5 kg N ha^{-1} yr^{-1} for HUC-2s west of the Mississippi River (Jordan and Weller 1996). For the higher estimate (12.2 Tg N yr^{-1}), we assumed a constant 1 kg N ha^{-1} yr^{-1} for everywhere except western arid USGS Water Resource Regions (California, Great Basin, Colorado, and Texas-Gulf-Rio Grande). For these four regions, we assumed a constant N fixation rate of 25 kg N ha^{-1} yr^{-1} (Jordan and Weller 1996).

Estimates of non-cultivated BNF calculated according to terrestrial ecosystem types (Cleveland *et al.* 1999) in Table 1 were developed by applying BNF rates of 5%, 15%, and 25% N-fixer coverage for specific ecosystem types to Level I ecoregions (www.epa.gov/wed/pages/ecoregions/na_eco.htm). WebTable 2 presents the pairings of Level I ecoregions with ecosystem type as described in Cleveland *et al.* (1999).

Non-cultivated BNF utilized the correlation between BNF and evapotranspiration (Cleveland *et al.* 1999) in Table 1, including the following regression models:

Conservative estimate:

$$\text{BNF (kg N ha}^{-1}\text{ yr}^{-1}) = 0.102 * \text{evapotranspiration (cm yr}^{-1}) + 0.524 \quad (1)$$

Central estimate:

$$\text{BNF (kg N ha}^{-1}\text{ yr}^{-1}) = 0.234 * \text{evapotranspiration (cm yr}^{-1}) - 0.172 \quad (2)$$

Upper-bound estimate:

$$\text{BNF (kg N ha}^{-1}\text{ yr}^{-1}) = 0.367 * \text{evapotranspiration (cm yr}^{-1}) - 0.754 \quad (3)$$

We downloaded global data (0.5 × 0.5 degrees grid resolution) describing actual evapotranspiration (http://climate.geog.udel.edu/~climate/html_pages/README.wb_ts2.html) to construct these estimates.

Synthetic N fertilizer

Many of the estimates regarding synthetic N fertilizer application in the US share common links to a single data source. Two such estimates (Ruddy *et al.* 2006; IFA 2011) share a link to fertilizer sales data compiled by the American Association of Plant Food Control Officials (www.aapfco.org). Three estimates (Bouwman *et al.* 2009; Liu *et al.* 2010; FAO 2011) are based on, or have been modeled using, data collected by the Food and Agricultural Organization of the United Nations (FAO; www.fao.org). One estimate, provided in the National Nutrient Loss and Soil Carbon (NNLSC) database, derives from surveys of ~75 000 farms and 15 major crops for 1997 (Potter *et al.* 2006).

Agricultural BNF

Methods used to estimate agricultural BNF included in the N inventory fall into three categories. The first method relies on mechanistic models – applying laboratory- or field-based data to model BNF as a function of crop type and limiting environmental factors, most commonly soil N availability (negative effect on BNF), moisture (positive effect), and temperature (non-linear effect; Liu *et al.* 2011) – to calculate agricultural BNF. The process-based estimate included in this inventory originates from the Environmental Policy Integrated Climate (EPIC) model, a process-based model of crop production that includes an N-fixing component for several N-fixing crop types (Potter *et al.* 2006). The EPIC model has been used to produce a national, spatially explicit estimate of agricultural BNF based on 1997 crop and soil conditions as part of the NNLSC database (Potter *et al.* 2006).

The second method applies a crop-specific per-area BNF rate to the area planted with that crop. National- and county-specific estimates derive from area planted with N-fixing crops described in databases maintained by the US Department of Agriculture (USDA) or the FAO (WebTable 1; Jordan and Weller 1996; Smil 1999; Howarth *et al.* 2002; Liu *et al.* 2010; SAB 2011).

The third method uses a crop yield-based approach and assumes that all N acquired by the plant during the growing season originates from agricultural BNF (Bouwman *et al.* 2009). Other recently developed regional or crop-specific models combine crop yield with measured data describing the fraction of N acquired by BNF relative to N assimilated from the soil (Han and Allan 2008; Herridge *et al.* 2008).

Atmospheric N deposition

Accurate source attribution of atmospheric N deposition is extremely difficult (Holland *et al.* 2005). For atmospheric deposition datasets that do not designate the origin of deposited N, we assume that either (1) 50% of the estimate is new N and 50% is recycled N, or (2) the NO_y component of deposition represents new N and the NH_x component represents recycled N (Holland *et al.* 2005). These approximations have been used in previous N assessments to designate N sources (eg Howarth *et al.* 2002; Holland *et al.* 2005), although future research on deposition source attribution is clearly needed. Finally, in order to estimate total N deposition from inorganic N deposition, we assumed that organic N deposition is 30% of total N deposition and augmented estimates of inorganic N deposition accordingly (Neff *et al.* 2002).

Data for atmospheric N deposition to the US originate from one of two sources. Data collected as part of the National Atmospheric Deposition Program (NADP; http://nadp.sws.uiuc.edu/) and/or the Clean Air Status and Trend Network (CASTNET; http://epa.gov/castnet/javaweb/index.html) provide information for four of the estimates (Jordan and Weller 1996; Holland *et al.* 2005; Ruddy *et al.* 2006). Data from these networks have been extrapolated from point measurements (250 NADP sites and 86 CASTNET sites) to estimate national-level N deposition. Although these estimates are based on observed data, the networks of sites

continued

WebPanel 1. – *continued*

used to construct the estimates are sparse and often located in areas with minimal human impact. Large-scale models based on data from industrial and agricultural emissions to the atmosphere form the basis of the remaining estimates (Howarth *et al*. 2002; Dentener 2006; Bouwman *et al*. 2009; USEPA 2010a). Modeled estimates have the advantage of including spatial patterns of deposition not captured by monitoring sites, and can help inform the selection of future monitoring locations.

Atmospheric N deposition estimates provided by the TM3 model (Dentener *et al*. 2006) are global in extent for 1993. We downloaded gridded data (5 × 3.75 degrees) available from the Oak Ridge National Laboratory (Dentener 2006) and, after downscaling data to 30 arcseconds (to minimize data exclusion at US boundaries), used Spatial Analyst in ArcMap 9.3 (ESRI Inc, Redlands, CA) to estimate US-specific atmospheric N deposition.

Livestock manure

Estimates of confined feedlot manure N ultimately derive from livestock population data collected for the USDA Census of Agriculture (USDA 2011). Differences among datasets reflect differences in methods used to translate animal populations into land application of manure and to account for volatilization/leaching (up to 50% of manure N produced on feedlots [Bouwman *et al*. 2005b]). Kellogg *et al*. (2000) and Ruddy *et al*. (2006) provided county-level data on feedlot manure N input to the US for 1982, 1987, 1992, and 1997. The best estimate incorporates a wide range of livestock categories, considers farm-specific life cycles of livestock (eg only the time spent on a feedlot by cattle for slaughter is considered), includes any supplemental N compounds added to feed (Stanton and Whittler 2006), and accounts for N loss to ammonia (NH_3) volatilization (Kellogg *et al*. 2000; Ruddy *et al*. 2006).

Wastewater

We estimated point-source inputs of non-industrial wastewater from the US population in 2000 (280 million). We used a constant per capita N excretion rate (6.1 kg N person^{-1} yr^{-1}), the fraction of the population connected to a centralized sewage system (~80% of the US population; USEPA 2011), and a fractional removal of N during the wastewater treatment process (~46%; WebPanel 1; Van Drecht *et al*. 2009). Although N removal in septic systems can be extremely variable and depend on the system's age, capacity, and technology (USEPA 2012), we assumed a similar fractional removal of N (46%) in on-site septic tanks to that of centralized treatment for the estimate of N input from septic system leaching.

Spatial distribution of N inputs

We compiled the spatial distribution of new N inputs, the magnitude of new human-mediated N input above background levels, and the largest human-mediated N source for the US from the 1990s to the early 2000s (Figures 3 and 4; WebFigure 1). For these estimates, we chose spatial datasets that offered complete coverage of the conterminous US land area, the highest spatial resolution, and complete metadata describing data acquisition and representation. To facilitate comparisons across datasets, we summarized inputs at the spatial resolution of the 8-digit Hydrologic Unit Code (HUC-8; Seaber *et al*. 1987) using the Zonal Statistics tool in the Spatial Analyst feature of ArcMap 9.3.

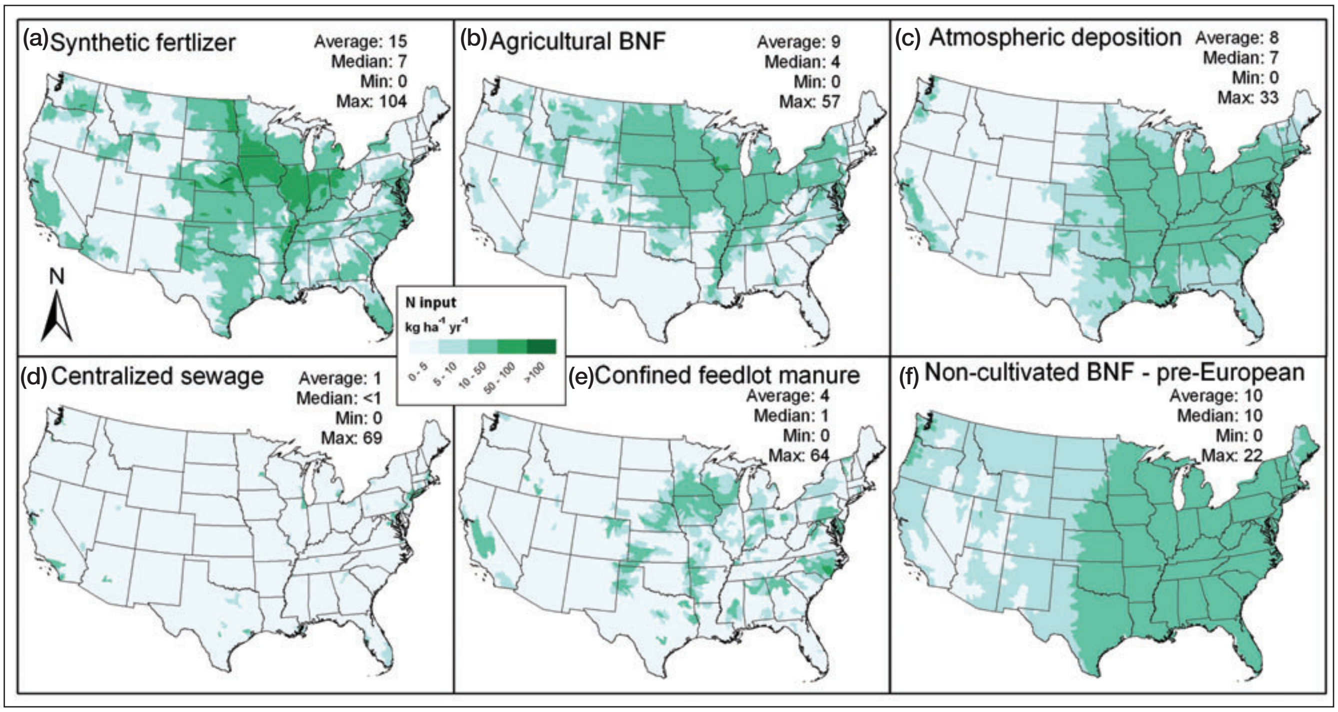
For agricultural N inputs (synthetic fertilizer, agricultural BNF, and confined feedlot manure), we used county-level data for 1997 (Ruddy *et al*. 2006; USDA 2011). All county-level estimates originate from Ruddy *et al*. (2006) except agricultural BNF, which was estimated by applying coefficients described in Smil (1999) and Howarth *et al*. (2002) to areas planted in N-fixing crops or in pasture for 1997 (USDA 2011).

We estimated the spatial distributions of non-fertilizer N inputs (non-cultivated BNF, wastewater, and inorganic N deposition) to the US using the following methods. For non-cultivated BNF, the regression model for the central estimate of non-cultivated BNF described above (Cleveland *et al*. 1999) was applied to a gridded (30-arcsecond resolution) dataset on actual evapotranspiration (derived from http://climate.geog.udel.edu/~climate/html_pages/README.wb_ts2.html) in the conterminous US. For wastewater, we applied the treatment corrected per capita excretion rate of N (2.8 kg N person^{-1} yr^{-1}; Van Drecht *et al*. 2009) to a 1-km × 1-km gridded dataset of the US population in 2000 (http://lwf.ncdc.noaa.gov/oa/climate/research/population/; rounded to the nearest 10 000). Lastly, we used 36-km × 36-km gridded data modeled by CMAQ for 2002 (USEPA 2010a) to estimate atmospheric inorganic N deposition in the US, assuming that NO_y originates primarily as new N and ammonium originates as recycled N (Holland *et al*. 2005).

We summed all new human-mediated N inputs and recycled N inputs for each HUC-8 in the US (Figure 4a). These sums represent gross input and do not account for N removed during harvest. We calculated the degree to which new human-mediated N input has increased total N input at the HUC-8 level by dividing the sum of all new human-mediated N inputs by background N input (WebFigure 1; non-cultivated BNF plus $\leq$1 kg N ha^{-1} yr^{-1} of inorganic N deposition; Boring *et al*. 1988). We identified the single largest human-mediated N input (new or recycled) by identifying the largest per area N input to each HUC-8 (Figure 3c).

Global context of N inputs to the US

On the basis of the best available estimates, new human-mediated N input to the US represents 15% of annual new human-mediated N input to Earth's land surface (WebTable 3). This suggests that human-mediated N input is currently three-fold higher in the conterminous US than would be expected if new human-mediated N input was distributed equally across Earth's land surface (the US contains 5% of global land area). In contrast to new human-mediated N input, new background N and recycled N inputs to the US are not exceptional relative to other world regions (4% and 7% of global estimates, respectively). Recycled N input is 17% of new N input in the US, whereas recycled N input is 47% of new N input to global land surfaces (WebTable 3).



WebFigure 1. *Spatial information on nitrogen (N) inputs available for the conterminous US. Spatial units are 8-digit Hydrologic Unit Codes. (a) Synthetic N fertilizer input, (b) agricultural biological N fixation (BNF), (c) atmospheric N deposition, (d) centralized sewage N, (e) confined feedlot manure N, and (f) non-cultivated BNF in pre-European times. See WebPanel 1 for data sources and method details. All N source estimates possess some level of uncertainty, with agricultural BNF, centralized sewage, confined feedlot manure, and non-cultivated BNF in pre-European times being the least certain.*

WebTable 1. Data resources available for estimating N input to the US (1990s–2000s)

Name	Link	Description	Modeled or measured?	Temporal extent	Temporal resolution	Spatial extent	Spatial resolution
New N							
Association of American Plant Food Control Officials	www.aapfco.org/	Fertilizer sales	Measured	1982–2001	Annual	US	County
Farm Business and Household Survey Data	www.ers.usda.gov/Data/ARMS/	Fertilizer application rates	Measured	1996–2005	Subannual	Nation/select states	Farm field/ crop type
Agricultural Chemical Usage Reports	www.nass.usda.gov/Statistics_by_ Subject/Environmental/index.asp	Fertilizer application by crop	Measured	1991–2010	Annual	Select states	Cropland acre
US Census Bureau Industrial Reports	www.census.gov/manufacturing/cir/ historical_data/mq325b/index.html	Economy of commercial fertilizer	Measured	1941–2010	Quarterly	US	US
USGS Mineral Handbook: Nitrogen Statistics and Information	http://minerals.usgs.gov/minerals/ pubs/commodity/nitrogen/index. html#myb	Economy of N consumption	Measured	1994–2010	Annual	US	US
Historical Statistics for Mineral and Material Commodities in the United States	http://minerals.usgs.gov/ds/2005/140/	Economy of N consumption	Measured	1943–2010	Annual	US	US
County-Level Estimates of Nitrogen and Phosphorus Fertilizer Use in the United States, 1945 to 1985	http://pubs.usgs.gov/of/1990/ data.html	N fertilizer use	Measured/ modeled	1945–1985	Annual	US	County
USDA Economic Research Service: Fertilizer Use and Price	www.ers.usda.gov/Data/FertilizerUse/	N fertilizer use by crop type	Measured	1960–2008	Annual	US	Select states
Global patterns of terrestrial biological nitrogen (N_2) fixation in natural ecosystems	www.agu.org/pubs/crossref/1999.../ 1999GB900014.shtml	Review of terrestrial N-fixation	Measured	--	Annual	Global	0.5 × 0.5 degrees
FERTISTAT: Fertilizer Use Statistics	www.fao.org/ag/agl/fertistat/index_ en.htm	N fertilizer use by crop type	Measured	1998	Annual	Global	Crop type
Nitrogen in rock: Occurrences and biogeochemical implications	www.agu.org/pubs/crossref/2002/ 2002GB001862.shtml	N released during weathering	Measured	--	Annual	Global	Hectare
Global inputs of biological nitrogen fixation in agricultural systems	www.springerlink.com/content/ 75063j57488126/	Agricultural N-fixation	Measured	2005	Annual	Global	US
Spatial Data in Geographic Information System Format on Agricultural Chemical Use, Land Use, and Cropping Practices in the United States	http://pubs.usgs.gov/wri/wri944176/ bat000.html	Agricultural N flow	Measured	1985–1991	Annual	US	County
Nitrogen in crop production: An account of global flows	www.agu.org/journals/ABS/1999/ 1999GB900015.shtml	Agricultural N flow	Measured/ modeled	1997–2050	Annual	Global	US
A high-resolution assessment on global nitrogen flows in cropland	www.pnas.org/content/107/17/ 8035.full	Agricultural N flow	Modeled	2000	Annual	Global	5 arc-minutes
IFADATA: fertilizer data provided by the International Fertilizer Institute	www.fertilizer.org/ifa/ifadata/search	Synthetic N fertilizer	Measured	1961–2009	Annual	Global	US
FAOSTAT: Fertilizer data provide by the Food and Agricultural Organization of the United Nations	http://faostat.fao.org/site/575/ default.aspx#ancor	Synthetic N fertilizer and industrial N	Measured	1961–2009	Annual	Global	US
Nitrogen's role in industrial systems	http://onlinelibrary.wiley.com/doi/ 10.1162/108819801753358517/ abstract	Industrial N	Measured	1996	Annual	US	US
County-level estimates of nitrogen and phosphorus from commercial fertilizer for the conterminous US	http://pubs.usgs.gov/sir/2012/5207/	N inputs from farm and non-farm fertilizer application	Both	1987–2006	Annual	US	County
Recycled N							
Manure Nutrients Relative to the Capacity of Cropland and Pastureland to Assimilate Nutrients: Spatial and Temporal Trends for the United States	www.nrcs.usda.gov/technical/NRI/ pubs/manntr.html	Manure N input to agricultural systems	Measured/ modeled	1982–1997	Annual	US	County

continued

WebTable 1. – *continued*

Name	*Link*	*Description*	*Modeled or measured?*	*Temporal extent*	*Temporal resolution*	*Spatial extent*	*Spatial resolution*
Models of Infectious Disease Agent Study: Poultry farms	www.epimodels.org/midas/pubsyntdata1.do	Estimates of poultry farms in US	Modeled	2008	Annual	US	Farm
US EPA Envirofact Permit Compliance System Water Discharge Permit Query form	www.epa.gov/enviro/html/pcs/pcs_query_java.html	Wastewater discharge	Measured	1970–2009	Daily	US	Point source
Discharge Monitoring Report (DMR) Pollutant Loading Tool	http://app6.erg.com/icisloader/	Wastewater discharge	Measured	2007	Annual	US	Point source
Clean Watershed Needs Survey	http://water.epa.gov/scitech/datait/databases/cwns/index.cfm	Wastewater discharge	Measured	1970–2008	Annual	US	Point source
Methods for Estimating Annual Wastewater Nutrient Loads in the Southeastern United States	http://pubs.usgs.gov/of/2007/1040/	Wastewater discharge	Modeled	2002	Annual	US	Point source
US Census Data on Small Community Housing and Wastewater Disposal and Plumbing Practices	http://water.epa.gov/infrastructure/wastewater/septic/census_index.cfm	Wastewater and septic systems	Measured	1990	Annual	US	Census reporting unit
Global nitrogen and phosphate in urban wastewater for the period 1970 to 2050	www.agu.org/pubs/crossref/2009/2009GB003458.shtml	Urban wastewater	Modeled	1970–2050	Annual	Global	Continental
An overview of the RFF Environmental Data Inventory	www.worldcat.org/title/rff-environmental-data-inventory/clc/019292227	Point sources	Measured	1978	Annual	US	Point
Nutrient loadings to streams of the continental United States from muncipal and industrial effluent	http://onlinelibrary.wiley.com/doi/10.1111/j.1752-1688.2011.00576.x/abstract	Point sources	Measured/modeled	1992–2002	Annual	US	Major river basins
USGS survey of nitrate deposits in the United States	http://pubs.usgs.gov/bul/0838/report.pdf	Geologic N	Measured	Pre-1932	--	US	Survey locations by state
Toxic Release Inventory	www.epa.gov/tri/	Point sources	Measured	1987–2010	Annual	US	Individual facilities
New and recycled N							
National Atmospheric Deposition Program: National Trends Network	http://nadp.sws.uiuc.edu/ntn/	Wet deposition of NH_4^+ and NO_3^-	Measured	1978–2008	Weekly	US	Collection site
National Atmospheric Deposition Program: Atmospheric Integrated Research Monitoring Network	http://nadp.sws.uiuc.edu/airmon/	Wet deposition of NH_4^+ and NO_3^-	Measured	1992–2008	Daily	US	Collection site
County Level Estimates of Nutrient Inputs to the Land Surface of the Conterminous United States	http://pubs.usgs.gov/sir/2006/5012/	TN input of fertilizer and manure; wet NH_4^+ and NO_3^-	Measured/modeled	1982–2001	Annual	US	County
USEPA Clean Air Status and Trends Network (CASTNET)	www.epa.gov/castnet/index.html	Dry deposition of inorganic N species	Measured/modeled	1986–2008	Daily	US	Collection site
Community Multiscale Air Quality (CMAQ) Modeling System	www.cmascenter.org/index.cfm	Total deposition of inorganic N species	Modeled	2002–2006, 2020	Hourly	US	4, 12, or 36 km grid
USDA Census of Agriculture	www.agcensus.usda.gov/	Fertilizer, livestock, crop harvest	Measured	1840–2007	Annual	US	State or county
Nitrogen Deposition onto the United States and Western Europe	http://daac.ornl.gov/CLIMATE/guides/nitrogen_deposition.html	Total deposition of inorganic N species	Measured/modeled	1978–1994	Annual	US	0.5 degree grid cells
Global Maps of Atmospheric Nitrogen Deposition, 1860, 1993, 2050	http://daac.ornl.gov/CLIMATE/guides/global_N_deposition_maps.html	Total deposition of inorganic N and NO_y	Modeled	1860, 1993, 2050	Annual	Global	5 × 3.75 degree cells
National Nutrient and Soil Carbon Loss Database	www.brc.tamus.edu/data-resources/national-nutrient-soil--carbon-losses	Agricultural N-fixation, inorganic and manure N fertilizers, N loss	Measured/modeled	1997	Subannual	US	Unique resource unit
Measured Annual Nutrient loads form Agricultural Environments (MANAGE)	www.ars.usda.gov/Research/docs.htm?docid=11079	Measured nutrient loads for agricultural and forest land	Measured	1952–2003	Annual	US	Farm field

continued

WebTable 1. – *continued*

Name	Link	Description	Modeled or measured?	Temporal extent	Temporal resolution	Spatial extent	Spatial resolution
US Environmental Protection Agency National Emissions Inventory	www.epa.gov/ttnchie1/trends/	Emissions of NH_3 and NO_x	Measured	1900–2008	Annual	US	US
USGS NAQWA SPARROW Model	http://water.usgs.gov/nawqa/sparrow/	N loading values	Measured/ modeled	1992/2002	Annual	US	Watershed segments
USGS attributes for NHDPlus catchments for the conterminous United States	http://water.usgs.gov/nawqa/ modeling/nhdplusattributes.html	N loading values	Measured/ modeled	2002	Annual	US	Watershed segments
Human Contributions to Terrestrial Nitrogen Flux	www.jstor.org/stable/1312895	N input and flux in the US	Measured/modeled		1990–1993	Annual	HUC-2
Reactive Nitrogen and the World: 200 Years of Change	www.bioone.org/doi/abs/10.1579/ 0044-7447-31.2.64	N input to regions of the world	Measured	1860–2050	Annual	Continental	Continental
Human alteration of the global nitrogen and phosphorus soil balances for the period 1970–2050	www.agu.org/pubs/crossref/2009/ 2009GB003576.shtml	Agricultural N inputs to the world	Measured/ modeled	1970–2050	Annual	Continental	Continental
Nitrogen Use in the United States from 1961–2000 and Potential Future Trends	http://pinnacle.allenpress.com/doi/ pdf/10.1579/0044-7447-31.2.88	N flows in the US	Measured/ modeled	1961–1997	Annual	US	US
Other available resources							
The Net Anthropogenic Nitrogen Inputs (NANI)	www.eeb.cornell.edu/biogeo/nanc/ nani/nani.htm	Anthropogenic N	Measured/ modeled	1987–1997	Annual	US	Watershed Toolbox
Nutrient Use Geographic Information System (NuGIS) for the US	www.ipni.net/NuGIS	GIS-based tool for a national N balance	Modeled	1987–2007	Annual	US	HUC-8
USEPA Watershed Deposition Tool	www.epa.gov/AMD/EcoExposure/ depositionMapping.html	Tool to estimate N deposition using CMAQ output	Modeled	2002–2006, 2020	Seasonal/ annual	US	4, 12, or 36 km grid
National Geochemical Survey Database	http://tin.er.usgs.gov/geochem/	N in soil and stream sediment samples	Measured	1967–2007	--	US	Sample point
CENTURY Soil Organic Matter Model	www.nrel.colostate.edu/projects/ century5/	Model of plant and soil N dynamics	Modeled	--	Monthly	Global	Biome
Estimated anthropogenic nitrogen and phosphorus inputs to the land surface of the conterminous United States–1992, 1997, and 2002	http://pubs.er.usgs.gov/publication/ sir20125241	Net input of anthropogenic N inputs to the US	Both	1992, 1997, 2002	Annual	US	County and watershed

WebTable 2. Pairing of Level I ecoregions (www.epa.gov/wed/pages/ecoregions/na_eco.htm) with ecosystem types described in Cleveland *et al*. (1999) for estimates of non-cultivated biological N fixation (BNF)

Level I ecoregion	*Cleveland* et al. *(1999) ecosystem type*	*BNF rate (kg N ha^{-1} yr^{-1})* 5%	15%	25%
North American Desert	Desert	4.84	7.81	10.78
Mediterranean California	Mediterranean Shrubland	1.52	2.51	3.51
Tropical Wet Forest	Tropical Evergreen Forest	14.73	25.40	36.07
Temperate Sierras	Temperate Forests	6.59	16.04	26.58
Southern semi-arid highlands	Arid Shrubland	9.47	22.04	33.93
Great Plains	Tall/Medium/Short Grassland	2.35	2.70	3.05
Eastern Temperate Forest	Temperate Forests	6.59	16.04	26.58
Marine West Coast Forest	Temperate Forests	6.59	16.04	26.58
Northwestern Forested Mountains	Temperate Forests	6.59	16.04	26.58
Northern Forests	Temperate Forests	6.59	16.04	26.58

Notes: Subheadings of 5%, 15%, and 25% refer to percent coverage of N-fixing plant species.

WebTable 3. Comparison between best available estimates of N inputs to the US (Tables 1 and 2) and global estimates of N input to land surfaces for the 1990s–2000s

N source	*US (Tg N yr^{-1})*	*Global (Tg N yr^{-1})*	*US contribution (%)*
New human-mediated N			
Synthetic fertilizer	10.9	100.0[a]	11
Agricultural BNF	7.7	31.5[a]	24
Non-fertilizer industrial	4.4	28.5[b]	15
Atmospheric deposition	3.9	31.2[c]	13
Total:	**26.9**	**184.5**	**15**
Recycled human-mediated N			
Atmospheric deposition	3.0	52.6[a]	6
Confined feedlot manure	1.2	17.0[c]	7
Wastewater	0.8	6.0[d]	13
Total recycled N:	**5.0**	**75.6**	**7**
New background N			
Non-cultivated BNF – pre-European	8.1	195.0[e]	4
Non-cultivated BNF – contemporary	3.6	195.0[e]	2
Atmospheric deposition	<0.1	5.4[a]	<2

Notes: [a]Galloway *et al.* (2004); [b]FAO (2008); [c]Liu *et al*. (2010); [d]Van Drecht *et al.* (2009); [e]Cleveland *et al*. (1999). BNF = biological N fixation.

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