

A comparison of NEWS and SPARROW models to understand sources of nitrogen delivered to US coastal areas

Michelle L. McCrackin · John A. Harrison ·
Jana E. Compton

Received: 11 June 2012 / Accepted: 3 November 2012
© US Government 2013

Abstract The relative contributions of different anthropogenic and natural sources of in-stream nitrogen (N) cannot be directly measured at whole-watershed scales. Hence, source attribution estimates beyond the scale of small catchments must rely on models. Although such estimates have been accomplished using individual models, there has not yet been a comparison of N source attribution predictions at large spatial scales. We compared results from two models applied to the continental US: Nutrient Export from WaterSheds (NEWS) and SPATIally Referenced Regressions On Watersheds (SPARROW). NEWS and SPARROW predictions for total N delivery to the US coastal zone were 373 and 429 kg N km⁻² year⁻¹, respectively, for the contemporary period. Despite differences in how inputs were represented and defined by the models, NEWS and SPARROW both identified the same single-largest N sources for 67 % of the surface area that drains to the US coastal zone. When only anthropogenic sources were considered,

agreement increased to 78 % of surface area. Fertilizer and crop N-fixation were dominant in the Mississippi River Basin, where the bulk of agricultural lands are located. Sewage and population-related sources were most important in urban areas and natural N (primarily N-fixation on non-agricultural land) was most important in the Pacific Northwest. Attribution to fertilizer plus crop N-fixation, atmospheric deposition, and sewage and population-related sources was generally greater by SPARROW than NEWS, and the reverse was true for manure and natural sources. Nonetheless, both models agreed in attributing 62–81 % of N delivered to the coastal zone in the continental US to human activities.

Keywords Nitrogen · Water quality · Atmospheric deposition · Sewage · Fertilizer · Model · Coastal zone · Nutrient export

M. L. McCrackin (✉)
National Research Council, National Academies
of Science, Washington, DC 20001, USA
e-mail: michelle.mccrackin@vancouver.wsu.edu

M. L. McCrackin · J. A. Harrison
School of the Environment, Washington State University,
Vancouver Campus, Vancouver, WA 98686, USA

J. E. Compton
Western Ecology Division, U.S. Environmental Protection
Agency, 200 SW 35th Street, Corvallis, OR 97333, USA

Introduction

Through activities associated with food and energy production and consumption, humans have significantly altered the global nitrogen (N) cycle (Schlesinger 2009), more than doubling the rate at which N is fixed and made available to the terrestrial biosphere. Excess N in soils, groundwater, rivers, and the oceans has degraded inland freshwater and coastal ecosystems (Galloway et al. 2003; Dodds et al. 2009). Chronic N loading can saturate

the N-retention capacity of ecosystems and compromise the services that they provide (Compton et al. 2011). In the continental United States, 70 % of estuaries have been degraded by nutrient loading that can result in hypoxic “dead zones” and nuisance or toxic algal blooms (Bricker et al. 2007).

In the US, recent attention has focused on sources of N in the environment, ecological impacts, and mitigation strategies at the national scale (e.g., Davidson 2012, Suddick and Davidson 2012). The Science Advisory Board to the US Environmental Protection Agency (SAB) has recommended an integrated, national approach to managing N and explored options to reduce environmental N inputs by 25 % (SAB 2011). A necessary precursor for evaluating and developing policies to improve coastal water quality at this scale is the ability to differentiate between contributions from agricultural activities, atmospheric deposition, sewage, and natural lands to coastal N delivery by rivers. One challenge to source apportionment is that N compounds are transformed by biological and physical processes as they move within and between ecosystems. As a result, the relative contribution of land-based N inputs may not be in proportion to their contribution to delivery at the coast. There is currently no method by which the magnitude of different N sources can be measured across large spatial scales. Stable isotope analyses have been used to distinguish nitrate (NO_3^-) sources (Silva et al. 2000) but are often inconclusive due to source mixing and fractionation occurring during biological processing. Further, NO_3^- is only one component of total N (TN) load and may have different sources than other N forms. In the absence of empirical measurements, models capable of source apportionment can be used to evaluate potential management options and determine strategies that will have the greatest impact (Whitall et al. 2004).

A number of approaches have been used to model the sources and fates of N on land, in groundwater, and river networks to improve our understanding of N biogeochemistry. Such models include Hydrologic Simulation Program Fortran (HSPF, <http://www.epa.gov/ceampubl/swater/hspf>), Net Anthropogenic Nitrogen Inputs (NANI, <http://www.eeb.cornell.edu/bigeo/nanc/nani>), Net Total Nitrogen Inputs (NTNI, Howarth et al. 2012), nitrogen loading model (NLM, Valiela et al. 1997), nutrient export from watersheds (NEWS, Mayorga 2010), Soil and Water Assessment Tool (SWAT, <http://swatmodel.tamu.edu>), SPATIally

Referenced Regressions On Watersheds (SPARROW, <http://water.usgs.gov/nawqa/sparrow>), and Watershed Assessment Tool for Evaluating Reduction Strategies for Nitrogen (WATERSN, Castro 2001). Except for NANI, TNNI, and SWAT, all of these models consider catchment retention and removal of land-based N inputs in the determination of N export (e.g., kg N year^{-1} or $\text{kg N km}^{-2} \text{ year}^{-1}$) and are capable apportioning N between different point and diffuse sources. Most modeling efforts in the US have focused on regional and local scales. Of the models capable of source attribution, only SPARROW and NEWS have been applied to the continental US.

Model inter-comparisons have been used to build confidence in model-derived insights in several fields, including weather and climate forecasting and predicting riverine nutrient concentrations (Tebaldi and Knutti 2007; Hejzlar et al. 2009; Exbrayat et al. 2011). Comparing model results can lend credibility to predictions and identify areas for future improvement. To date, most comparisons of predictions between N export models have been made at global (Boyer et al. 2006) or basin scales (Alexander et al. 2002; Valiela et al. 2002) and have focused on primarily on N export rates (e.g., kg N year^{-1} or $\text{kg N km}^{-2} \text{ year}^{-1}$). Alexander et al. (2002) and Valiela et al. (2002) also compared N source attribution predictions of different models for specific watersheds based on common input data. In both studies the authors found substantial differences between the models' attribution to anthropogenic sources, illustrating the potential risk of relying on the predictions of a single model.

Here we compare predictions by NEWS and SPARROW for source apportionment and TN yield ($\text{kg N km}^{-2} \text{ year}^{-1}$) delivered by rivers to coastal areas of the continental US. This comparison complements recent efforts to understand N dynamics at the national scale (SAB 2011; Davidson 2012). Managing coastal water quality requires understanding the factors that influence water quality across large spatial scales, particularly because large rivers transport nutrients such as N across local and state political boundaries. Our objectives are to (1) provide information to policymakers and resource managers about important sources of N delivered to coastal areas of the US, (2) identify areas of agreement and disagreement between source-attribution predictions by NEWS and SPARROW, and (3) identify key factors that contribute to differences in predictions.

Methods

Overview of models

Both NEWS and SPARROW use mass balance approaches that consider different N sources and retention on land and in river networks to predict N export and source apportionment. NEWS and SPARROW differ in several respects, including: (1) how inputs are represented and defined by the models, (2) the sources of input data, and (3) the spatial scales at which predictions are made. NEWS is a spatially explicit, global model that combines empirical, mechanistic, and statistical approaches to predict the export of different nutrients (N, phosphorus, silica, and carbon) and nutrient forms (dissolved, inorganic, and particulate) for more than 6,000 global rivers (Table 1; Mayorga 2010). NEWS attributes sources of dissolved inorganic N (DIN) and dissolved organic N (DON) to household sewage, fertilizer, biological N-fixation by crops, manure, atmospheric deposition, and N-fixation in non-agricultural (natural) areas. Model structure does not allow for source attribution predictions for PN. The transfer efficiency of N inputs from land surfaces to river networks is estimated as a function of runoff. Riverine sinks for N in NEWS are denitrification, consumptive water withdrawals, and retention behind dams. Retention of N both on land and in rivers is estimated at the whole-basin scale. NEWS predicts loads (kg N year^{-1}) and yields ($\text{kg N km}^{-2} \text{ year}^{-1}$) delivered to the mouths of coastal rivers and was calibrated at the global scale for basins $>25,000 \text{ km}^2$ (Mayorga 2010). Catchment delineations used by NEWS are from the global simulated topographical network (STN) (Vörösmarty and Fekete 2011). Data inputs used to drive NEWS were developed globally at 30 min spatial resolution and averaged over the area of each basin (Table 2).

SPARROW uses a hybrid statistical and mechanistic approach to estimate the sources, transport and transformation of nutrients in watersheds (Table 1). Model parameters are statistically estimated at the reach scale by calibration to measured in-stream water quality measurements (Alexander et al. 2008). The reach network was defined from 1 km digital elevation model watersheds (http://water.usgs.gov/GIS/metadata/usgswrd/XML/erf1_2.xml). SPARROW estimates N delivered to river reaches and coastal areas from household and industrial sewage, urban runoff, fertilizer

applied to specific crops (e.g., wheat, corn), crop N-fixation, manure, atmospheric deposition, and runoff from forest, shrub, and barren lands. Transfers of N from watersheds to streams are calculated as a function of six catchment characteristics, including: temperature, soil permeability, and stream density (Alexander et al. 2008). In-stream N processing is modeled using a first-order decay process as a function of water travel time and water depth. SPARROW predictions of delivered TN load (kg N year^{-1}), and source attribution percentages for 2002 were available for 250 coastal and estuarine drainage areas as defined by the US National Oceanic and Atmospheric Administration's Coastal Assessment Framework (CAF, <http://coastalgeospatial.noaa.gov>). This excludes the Colorado, St. Lawrence, and Nelson rivers, because the mouths of these rivers are located outside the US. Model input data were developed at county to national scales from a variety of sources (Table 2).

Predictions by NEWS and SPARROW are based on estimates of N inputs to catchments for 2000 and 2002, respectively (Alexander et al. 2008; Mayorga 2010). Both models address year-to-year variation in riverine nutrient fluxes though calibration with long-term average annual water quality data (1975–1995 for SPARROW, 1980–1997 for NEWS). We consider predictions of TN export and source attribution by both NEWS and SPARROW to represent average fluxes for the “contemporary” period and, thus, to be comparable. Throughout the paper, we define N export as yield ($\text{kg N km}^{-2} \text{ year}^{-1}$) and load (kg N year^{-1}) of N delivered to the coast by catchments after retention on land and in the river network. Load and yield are not used to refer to N sources to land surfaces or intermediate water bodies along the land-to-ocean continuum. The term “input” is used to refer to data used to run the models.

Model comparisons

We compared predictions of TN yield and source attribution between NEWS and SPARROW for rivers that drain to coastal areas of the continental US. Basin delineations used by NEWS were generally at coarser scales than SPARROW, so we aggregated the SPARROW delivered TN loads and source-share attribution (portion of load) into the encompassing NEWS basin. We manually compared basin delineations, drainage areas, and the location of river mouths between

Table 1 Summary of NEWS and SPARROW model structure

	NEWS	SPARROW
Basin delineations	Global river and reservoir networks defined in Simulated Topological Networks http://www.wsag.unh.edu/Stn-30/stn-30.html	Streams, reservoirs, and drainage topography defined in Enhanced River Reach File http://water.usgs.gov/GIS/metadata/usgswrd/XML/erf1_2.xml
Scale	Applied at global scale for > 6,000 basins	Applied to continental US
N forms	Source attribution predictions for DIN, DON	Source attribution predictions for TN
Watershed N retention	Removal by harvest, function of runoff. Estimated at whole-basin scale	Function of catchment characteristics including stream network density, temperature, precipitation Estimated at reach scale
N retention in river network	Denitrification, dams, and irrigation water withdrawals Estimated at whole-basin scale	First-order decay as a function of water travel time and stream depth Estimated at reach scale
Model performance	Nash–Sutcliffe Efficiency	Root mean square error Coefficient of determination

Table 2 Comparison of model input data used in NEWS and SPARROW

Model input	NEWS		SPARROW	
	Description	Sources	Description	Sources
Fertilizer	Fertilizer application rates by country	IFA/IFDC/FAO 2003	Estimates of commercial fertilizer inputs by US county	Ruddy 2006
N-fixation by agricultural crops	Estimates of biologically fixed N ₂ for legume crops	Cleveland et al. 1999	Estimates of biologically fixed N ₂ for soybean, alfalfa, and hay crops by US county	McIsaac et al. 2002; NASS 2002
Atmospheric N deposition	Global modeled wet and dry NH _y and NO _x deposition	Dentener et al. 2006	Mean annual measured wet NO ₃ ⁻ deposition	NADP 1993
Sewage and population-related	Estimated as a function of population density and gross domestic product	Van Drecht 2009	Human population as a surrogate for municipal waste-water effluent, septic systems, and urban runoff	GeoLytics 2000
Manure	Estimated animal stocks and production per animal	Alcamo et al. 2006; Bouwman, 2009	Estimates of the nutrients in livestock manure, based on animal census by US county	Kellogg et al. 2000
Natural/non-agricultural land	Biologically fixed N ₂ on non-agricultural lands	Cleveland et al. 1999	Forest, shrub, and barren land cover as a surrogate for runoff from undeveloped lands	Vogelmann et al. 2001

hydrography models (CAF and STN) to determine which SPARROW-defined basins aligned with NEWS-defined basins. SPARROW TN loads and source shares were summed into the encompassing NEWS basin. Yields were determined by dividing aggregated loads by aggregated basin area. For sub-continental analysis, we further combined individual basins into five regions that differed in terms of

dominant land uses and climate. Regions were defined as Northeast and mid-Atlantic (29 basins), Southeast (39 basins), Mississippi River (1 basin), western Gulf of Mexico (13 basins), and West Coast (33 basins). While our primary interest was comparing source attribution predictions, we quantified differences in TN yield predictions as the ratio of the NEWS to SPARROW prediction for each river basin.

SPARROW and NEWS predictions of N yield were previously calibrated with observed water quality data. SPARROW used root mean square error (RMSE) and the coefficient of determination (r^2) to measure the differences between modeled and observed data (Alexander et al. 2008). RMSE for the TN model was 0.55 and r^2 for TN yield was 0.87. Alexander et al. (2008) used a bootstrapping approach to estimate source-attribution prediction intervals for the Mississippi and Atchafalaya rivers; such data are not available for SPARROW at the national scale (R. Smith, personal communication).

NEWS model performance was evaluated using the Nash–Sutcliffe efficiency (R^2), a measure of how well the linear relationship of observed and modeled yield data conform to unity (Nash and Sutcliffe 1970). Values of R^2 between 0 and 1 indicate a model is more accurate than simply using the mean of measurements to predict DIN export and 1 is unity (when all points fall on the 1:1 line). R^2 was 0.54 and 0.71 for NEWS predictions of DIN and DON yield, respectively (Mayorga 2010). Estimates of prediction error were not available for NEWS-derived source attributions.

Source attribution was determined as the portion of total load for each N source for: (1) individual basins (based on NEWS delineations), (2) regions (e.g., summation of loads for all rivers in a region), and (3) the US as a whole (e.g., summation of loads for all rivers that drain to the US coast). Because NEWS and SPARROW defined or named N sources somewhat differently, we aggregated or assigned source attribution predictions from both models into the following categories for the comparison: fertilizer plus crop N-fixation, atmospheric deposition, manure, sewage and population-related sources, and natural. The “translation” of the source categories between models was mostly related to agricultural and natural sources (Table 3). NEWS attributed N separately to fertilizer and crop N-fixation while these sources were combined and reported by crop type in SPARROW. Accordingly, we aggregated SPARROW predictions for corn and soybeans, wheat, alfalfa, and other crops as “fertilizer plus crop N-fixation”. SPARROW predictions for forest, shrub and barren lands sources were combined as “natural” for comparison to NEWS.

There is a potential mismatch in source attribution predictions between models because NEWS is only capable of source apportionment for DIN and DON, not PN. Particulate N averaged 19 % of TN yield for

Table 3 Translation of source attribution terms used by NEWS, SPARROW, and for their comparison (as in Table 4; Figs. 2–5)

NEWS	SPARROW	Model comparison (this paper)
Fertilizer	Corn and soybeans	Fertilizer plus crop N-fixation
Other diffuse runoff from agricultural land	Wheat Alfalfa Other crops	
Biological fixation by crops		Fertilizer plus crop N-fixation
Animal manure	Pasture/rangeland	Manure
Biological fixation in non-agricultural areas	Forest Shrub land Barren land	Natural
Atmospheric deposition	Atmospheric deposition	Atmospheric deposition
Human waste	Urban and population-related sources	Sewage and population-related sources

the Mississippi River and 9–12 % for the Northeast and mid-Atlantic, western Gulf of Mexico, and the Southeast. For the West Coast, the fraction of PN was greater and averaged 24 % of TN yield because of a number of small, steep river basins. We evaluated the potential for PN to influence the comparison of source attribution predictions by quantifying the fraction of TN yield composed of PN for each river. There was no statistical difference ($P = 0.7$) in the PN fraction of TN for basins where single-largest N-source predictions agreed and basins where there was disagreement as to dominant source between models.

Results

National overview

Predictions of TN yields to coastal areas of the continental US were 373 kg N km⁻² year⁻¹ according to NEWS and 429 kg N km⁻² year⁻¹ according to SPARROW. Corresponding TN loads were 2.3 and 2.7 Tg N year⁻¹ for NEWS and SPARROW, respectively. Among river basins, yields spanned more than two orders of magnitude and there was a wide range of predictions for both NEWS (2–2,686 kg N km⁻² year⁻¹) and SPARROW (22–5,173 kg N km⁻² year⁻¹). For all basins, TN yield predictions by NEWS

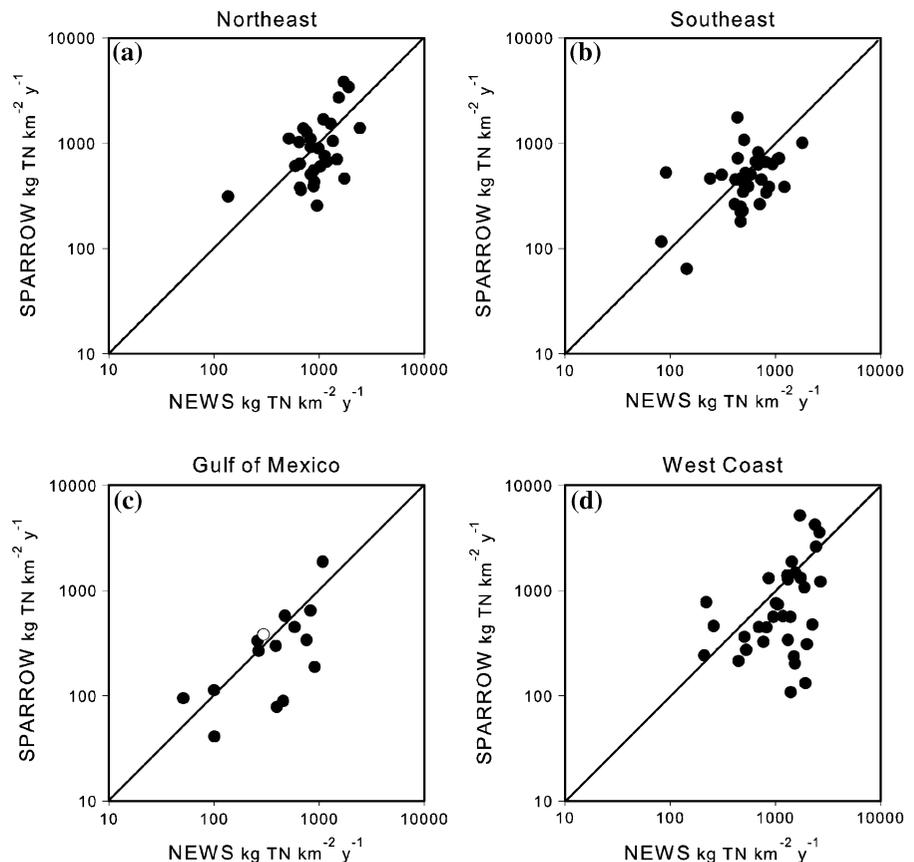
tended to be greater than those by SPARROW, by a median factor of 1.4 (interquartile range, IQR, 0.7–3.6) (Fig. 1). NEWS TN predictions were within a factor of two of SPARROW predictions for 56 % of rivers and were within a factor of 4 for 80 % of rivers. For about 50 % of all basins, differences in model predictions fell outside the RMSE of SPARROW predictions, and in these cases NEWS predictions were greater than SPARROW by a median factor of 2.6 (IQR, 1.7–4.9). Yields were most similar for basins in the northeastern and mid-Atlantic region (Fig. 1a, median NEWS: SPARROW ratio = 1.0, IQR 0.6–1.8) and least similar for the West Coast (Fig. 1d, median ratio = 1.4, IQR 0.9–2.2). There was no relationship between the difference in model predictions and basin size ($r^2 = 0.01$), which ranged from 2,000 to 3 million km².

For the continental US, NEWS and SPARROW predicted that anthropogenic N accounted for 62 and 81 % of coastal N loading, respectively (Fig. 2). NEWS attributed less to fertilizer plus crop N-fixation (31 %) than SPARROW (42 %), while the reverse

was true for manure (12 vs. 5 %, for NEWS and SPARROW, respectively). N export attributable to all agricultural activities (fertilizer, crop N-fixation, and manure) was comparable between models (SPARROW: 47 %, NEWS: 43 %). There was also good agreement for atmospheric N deposition (SPARROW: 21 %, NEWS: 17 %). Agreement between models was poorest for sewage, population-related, and natural N sources. SPARROW attributed more than twice as much exported N to sewage and population-related sources (21 %) compared to NEWS (9 %). NEWS attributed 31 % of coastal loads to natural N on undeveloped land, while SPARROW attributed 11 %.

Regionally, the dominant (single-largest) sources of N predicted by both SPARROW and NEWS reflect land-use patterns across the US (Fig. 3a, b; Table 4). Atmospheric deposition, sewage, and population-related sources were important along densely populated areas of the East and West Coasts, while fertilizer and crop N-fixation dominated in central regions. Natural N sources were most important in the Pacific

Fig. 1 Each point represents TN yield ($\text{kg N km}^{-2} \text{ year}^{-1}$) predicted by NEWS and SPARROW for the **a** Northeast, **b** Southeast, **c** Gulf of Mexico (open circle is Mississippi River), and **d** West Coast



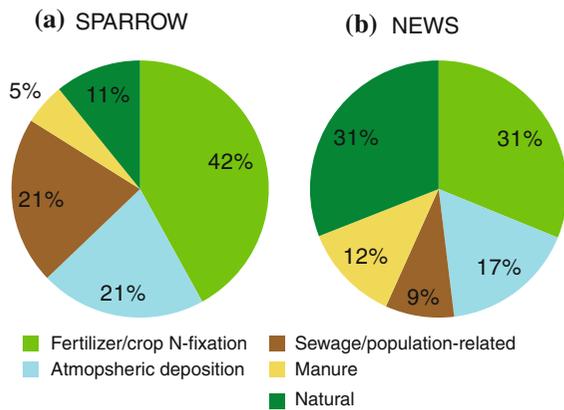


Fig. 2 Model predictions of source apportionment of N to the coastal zone for the continental US for **a** SPARROW and **b** NEWS. Apportionments were estimated as percentages of total load (kg N year^{-1}) for 113 basins. The anthropogenic portion is estimated as the sum of fertilizer plus crop N-fixation, 60 % of atmospheric deposition (as in Galloway et al. 2003), sewage and population-related sources, and manure, and totals 81 and 62 % for all N sources for SPARROW and NEWS, respectively

Northwest. The dominant N sources identified by both models agreed for 67 % of the drainage area and 63 % of the discharge of the continental US (excluding the Colorado, Nelson, and St. Lawrence River Basins, Fig. 3c). There were no basins where both models identified atmospheric deposition or livestock manure as the dominant N source. Mean predicted TN yield did not differ between basins where the predicted dominant N source agreed and basins where there was disagreement as to dominant source ($P > 0.05$).

When only anthropogenic N sources were considered, the area of agreement between models increased to 75 % of the modeled drainage area and 79 % of discharge. Fertilizer plus crop N-fixation constituted the single largest N source for the majority of the continental US (Fig. 4a, b). Additionally, there were a number of smaller basins along the Atlantic and Pacific coasts where NEWS and SPARROW found sewage, population-related N, and atmospheric deposition to be the most important sources (Fig. 4c).

Regional summaries

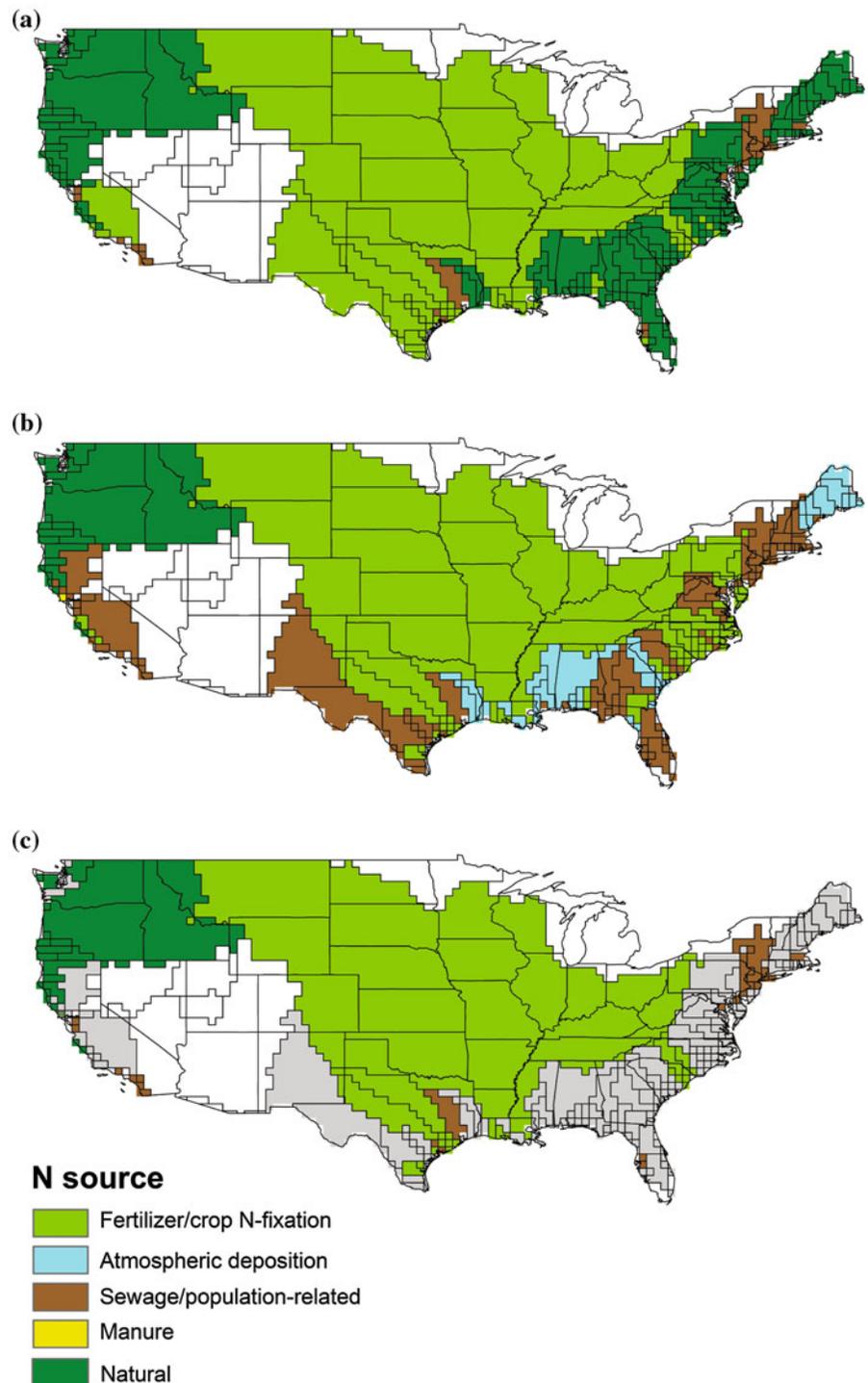
We prepared regional summaries to understand how model comparisons varied for different land uses and climates. Predicted yields for river basins in the region

encompassing northeast and mid-Atlantic watersheds were similar: 801 and 838 $\text{kg N km}^{-2} \text{ year}^{-1}$ for NEWS and SPARROW, respectively. This agreement did not extend to source attribution predictions; NEWS found the single-largest source of coastal N was from natural sources (45 % of all sources) while SPARROW identified sewage and population-related sources (43 %) (Fig. 5; Table 4). The models agreed atmospheric N deposition was the second-largest source (NEWS: 23 %; SPARROW: 25 %). Chesapeake Bay represents about 44 % of the drainage area for northeast and mid-Atlantic region and is the focus of a number of eutrophication studies. NEWS predicted that TN delivery to Chesapeake Bay was 964 $\text{kg N km}^{-2} \text{ year}^{-1}$ compared to 765 $\text{kg N km}^{-2} \text{ year}^{-1}$ according to SPARROW. NEWS found that natural sources of N were most significant (29 %) for this region, followed by fertilizer and crop N-fixation (22 %). SPARROW predicted that sewage and population-related sources (34 %) and fertilizer and crop N-fixation (29 %) were most important to Chesapeake Bay.

For the southeastern US, which includes 8 states that drain to the Atlantic Ocean and eastern Gulf of Mexico, the regional TN yield was 637 and 439 $\text{kg N km}^{-2} \text{ year}^{-1}$ according to NEWS and SPARROW, respectively. NEWS found N-fixation in non-agricultural areas to be most important (43 %) and SPARROW identified sewage and population-related sources (32 %) (Fig. 5; Table 4). Both models identified fertilizer plus crop N-fixation as the second-largest N source delivered to coastal areas (NEWS: 18 %; SPARROW: 24 %).

The Mississippi River Basin comprises nearly 80 % of the US drainage area for the Gulf of Mexico. In NEWS, the Mississippi River is delineated as a single basin that included the Atchafalaya River, while in SPARROW this region covers about 25,000 sub-catchments. NEWS and SPARROW estimated TN yields to be 293 and 382 $\text{kg N km}^{-2} \text{ year}^{-1}$ (Fig. 1c), respectively and attributed 54 and 61 % of coastal export to fertilizer plus crop N-fixation, respectively (Fig. 5; Table 4). For rivers draining to the western Gulf of Mexico, TN yields were similar between models (NEWS: 136 $\text{kg N km}^{-2} \text{ year}^{-1}$, SPARROW: 122 $\text{kg N km}^{-2} \text{ year}^{-1}$). Here, NEWS attributed 35 % of N export to fertilizer plus crop N-fixation and 17 % to sewage and population-related sources. The reverse was true for SPARROW, which identified sewage and population-related sources (40 %) and

Fig. 3 Single-largest sources of N exported by 113 individual catchments as predicted by **a** NEWS and **b** SPARROW. Map **(c)** shows areas where models agree as to the dominant source of riverine N; *gray shaded* areas indicate no agreement. *White areas* are regions not included in the comparison



fertilizer plus crop N-fixation (27 %) as the first- and second-largest sources of N, respectively, exported to the western Gulf.

Predicted regional TN yields for estuaries along the coasts of California, Oregon, and Washington were similar between NEWS and SPARROW at 427 and

Table 4 Source attribution predictions for major US regions

Region	Model	Fertilizer and crop N-fixation (%)	Atmospheric deposition (%)	Sewage and population-related sources (%)	Manure (%)	Natural N sources (%)
Northeast and mid-Atlantic	SPARROW	18	25	43	3	10
	NEWS	8	23	18	7	45
Southeast	SPARROW	24	25	32	7	11
	NEWS	18	21	7	11	43
Mississippi River	SPARROW	61	18	10	6	5
	NEWS	54	15	5	15	11
West Coast	SPARROW	15	18	24	4	39
	NEWS	16	10	8	10	56
W. Gulf of Mexico	SPARROW	27	19	40	7	7
	NEWS	35	16	17	15	16

457 kg N km⁻² year⁻¹, respectively. For the region as a whole, both models found N originating on undeveloped land to be the dominant source of coastal N, but attribution was greater by NEWS (56 %) than SPARROW (39 %). NEWS identified fertilizer and crop N-fixation (16 %) as the second-most important source while SPARROW identified sewage and population-related sources (24 %) (Fig. 5; Table 4).

Discussion

Sources of coastal nitrogen

Agriculture

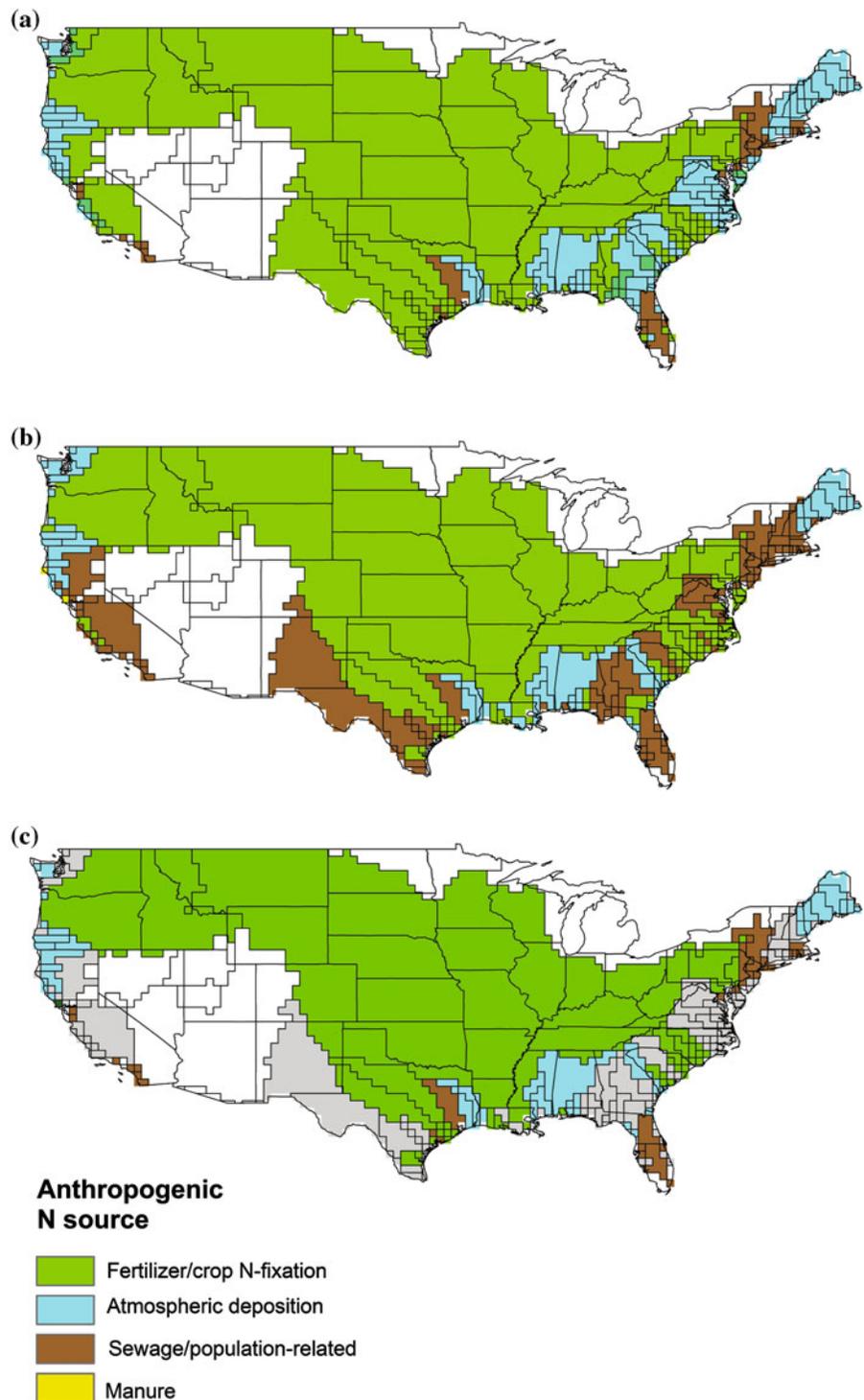
At the continental scale, SPARROW apportioned a third more riverine N to fertilizer plus crop N-fixation than NEWS (Fig. 2). At regional scales, attribution to this source was most similar for the Mississippi River, western Gulf of Mexico, and West Coast and least comparable for the Northeast and Southeast (Table 4). It is likely that attribution predictions differ in part because of the underlying input data used to drive the models (Table 2). A recent analysis by Sobota et al. (2013) found relatively strong agreement between different estimates of N fertilizer inputs for the continental US and poor agreement for biological N-fixation. This finding is reflected in the models' input data in that total fertilizer inputs were more similar between models (SPARROW:

18.2 Tg year⁻¹; NEWS: 14 Tg N year⁻¹) than crop N-fixation (SPARROW: 7.3 Tg N year⁻¹, NEWS 1.9 Tg N year⁻¹) (Alexander et al. 2008; Bouwman 2009). These differences highlight the need for improved estimates of land-based N inputs mediated by humans.

In addition to the magnitude of input data, another important factor to consider when comparing model output is transfer efficiency. For example, NEWS apportioned more than twice as much N export to manure than SPARROW for the US as a whole and this pattern is seen across all regions (Table 4). Manure inputs were somewhat greater for NEWS (7 Tg N year⁻¹, Bouwman 2009) compared to SPARROW (5.8 Tg N year⁻¹, Kellogg et al. 2000), but not enough to explain differences in source attribution. However, we estimate that NEWS transferred about 17 % of agricultural N from land to water in contrast to 6–16 % for SPARROW (depending on crop). Thus, the combination of differences in N inputs and transfer efficiencies could likely explain differences in source attribution predictions between models.

Predictions by NEWS and SPARROW for the Mississippi River and areas of the western Gulf of Mexico are supported by a number of other models, including NANI and NLM, which found fertilizer and crop N-fixation to be significant N sources to the Gulf of Mexico (Howarth et al. 1996; Castro et al. 2003; Green et al. 2004; David et al. 2010). Stable isotope studies of N sources in the Mississippi River

Fig. 4 Single-largest source of anthropogenic N exported by 113 individual catchments as predicted by **a** NEWS and **b** SPARROW. Map **(c)** shows areas where models agree as to the dominant source of riverine N; *gray shaded* areas indicate no agreement. *White areas* are regions not included in the comparison



suggesting that NO_3^- originates primarily in agricultural areas of the upper Midwest (Goolsby et al. 2000; Battaglin 2001) are also consistent with model source

attributions. In this region, multiple lines of evidence indicate an important role for exports of agricultural N to the Gulf of Mexico.

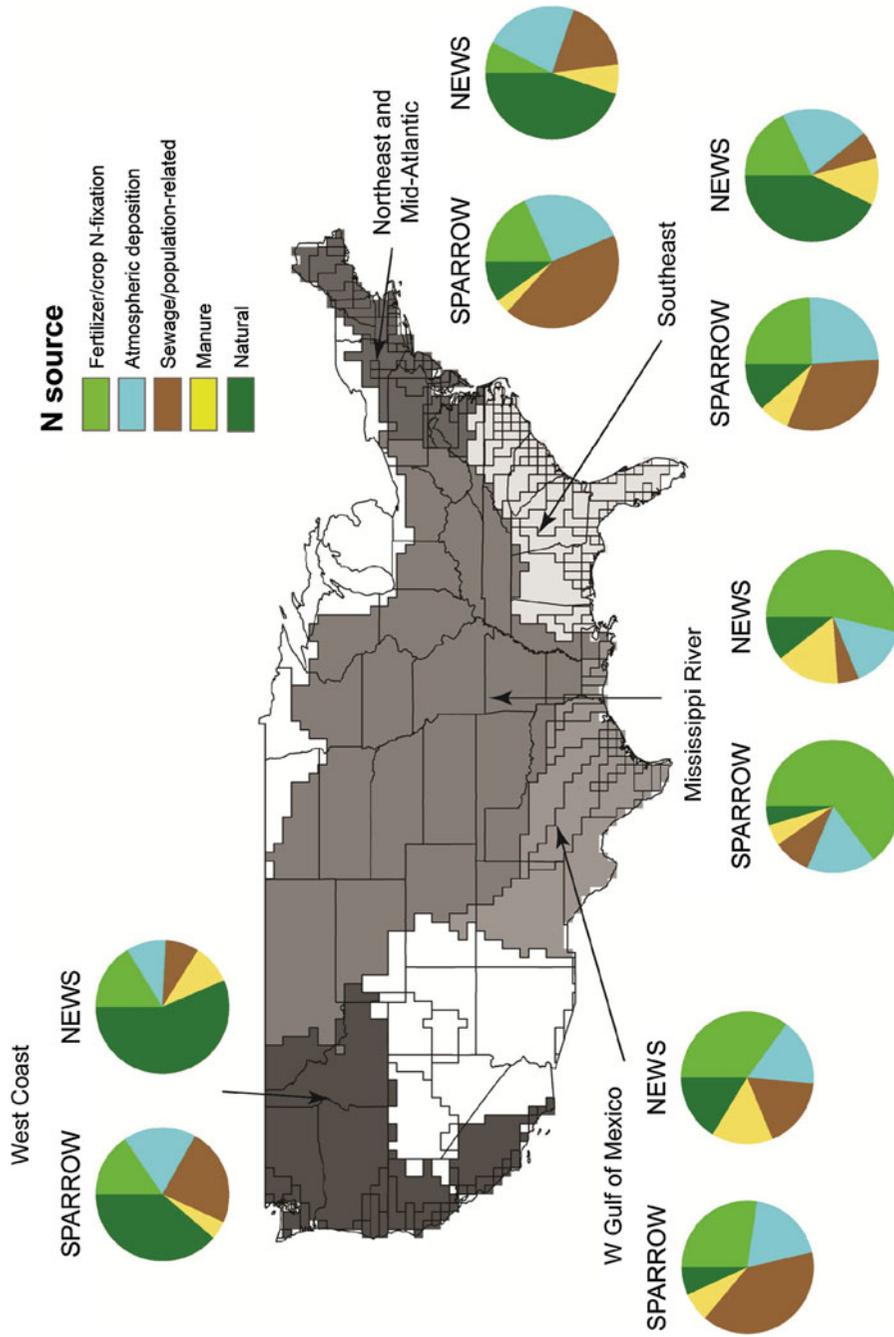


Fig. 5 Comparison of N source attribution between SPARROW and NEWS models for regions of the US. *White areas* are regions not included in the comparison. Corresponding *pie chart* values are shown in Table 4

Atmospheric deposition

There was good agreement between NEWS and SPARROW attribution to N deposition for the US as a whole (Fig. 2) and for all regions except the West Coast (Table 4), although SPARROW consistently attributed more to this source than NEWS. For individual rivers, there were no cases where both models identified N deposition as the single largest N source (Fig. 3c). As seen with fertilizer and crop N-fixation, there was improved agreement between models when only anthropogenic N sources were considered (Fig. 4c). It is important to note that our analysis only considers N deposited to land surfaces that is subsequently processed in the watershed and river network before export to the coastal zone. Neither SPARROW nor NEWS includes direct deposition of N to estuaries, which is significant in some areas (Whitall et al. 2003; Howarth 2008).

A number of factors likely contribute to differences in attribution to atmospheric deposition. First, SPARROW uses wet NO_3^- deposition rates to represent total atmospheric N deposition as a predictor for in-stream N, while NEWS uses global estimates of oxidized and reduced forms of wet and dry deposition (Table 2). SPARROW might overestimate or underestimate the contribution of atmospheric deposition to river N export to the extent that wet NO_3^- deposition is not proportional to wet plus dry deposition (Howarth 2008). To investigate this potential, we compared the relationship between wet NO_3^- deposition rates used by SPARROW (<http://nadp.sws.uiuc.edu/nadpdata>) to TN deposition rates used in NEWS. We found no relationship ($r^2 < 0.01$), suggesting there is no systematic over- or under-estimation based on the models' input data. Second, transfer efficiencies could explain why SPARROW tended to attribute a greater portion of riverine N to atmospheric deposition than NEWS. SPARROW estimated that about 70 % of deposition is transferred from land to water while NEWS transfer efficiencies were ~ 30 %. These values aren't strictly comparable because NEWS used total wet and dry N deposition for data inputs, while the SPARROW used wet NO_3^- deposition, but could explain differences between the models. Lastly, there were differences in model structure. NEWS assumed that all atmospheric deposition is exported in the form of DIN and there was potential for NEWS to under-apportion riverine N to atmospheric deposition by not accounting for export as

DON. Also, the regression approach used by SPARROW may over- or under-allocate atmospheric deposition to in-stream N if, for example, N runoff from non-agricultural lands was more strongly correlated with wet NO_3^- deposition than with the area of forest, shrub, and barren land. In such cases, in-stream N attributed to atmospheric deposition could include natural N sources as well.

SPARROW found N deposition to be the dominant source of delivered N to estuaries along the Atlantic Coast and Gulf of Mexico (Fig. 3c) and these predictions were generally consistent with those by NANI, WATERSN, NLM, and the regional SPARROW model for the Southeast (Howarth et al. 1996; Boyer et al. 2002; Castro et al. 2003; Hoos and McMahon 2009; Latimer and Charpentier 2010). The regional SPARROW model for the Northeast and mid-Atlantic found that deposition was dominant source in estuaries of the Connecticut, Kennebec, and Penobscot rivers (Moore 2011). For the region as whole, however, atmospheric deposition was the third largest source of TN load (20 %), behind agriculture (37 %) and sewage and population-related sources (28 %).

There are surprisingly few isotope studies to which we could compare the models' predictions and the few that exist were generally inconclusive. For example, studies of watersheds in the Northeast found evidence that in-stream NO_3^- was derived primarily from nitrification of NH_4^+ from indeterminate sources rather than direct atmospheric deposition (Mayer et al. 2002; Barnes et al. 2008). However, NH_4^+ can represent about 20 % of total N deposition in this region (<http://nadp.sws.uiuc.edu/nadpdata>) that can be subsequently nitrified or assimilated and mineralized before being exported. In such cases, isotopic analysis could underestimate the contribution of atmospheric deposition to in-stream N. In the absence of measurements data, output from a number of models suggests that atmospheric N deposition is an important source of N delivered to many estuaries along the Atlantic coast.

Sewage and population-related sources

There was poor agreement between NEWS and SPARROW for sewage and population-related sources at national and regional scales (Fig. 2; Table 4) and for individual rivers (Fig. 3c). In all cases, attribution by SPARROW was greater than that by NEWS. This is

likely due to differences in how source input data are defined and estimated. SPARROW used human population as a proxy for N from municipal sewage effluent, septic systems, and other urban runoff that included atmospheric deposition and non-agricultural fertilizer use. NEWS model inputs considered per capita rates of N excretion and connectivity to sewage systems to estimate human N emissions (Van Drecht 2009). Unlike SPARROW, NEWS input data did not include industrial waste or other urban sources and this likely explains why attribution to sewage and population-related sources was consistently greater by SPARROW than NEWS.

The predictions of local and regional models were generally more consistent with SPARROW than NEWS for attribution to sewage and population-related sources. Regional SPARROW models, NLM, WATERSN, and other models identified this source as most important for a number of estuaries in the Northeast and Gulf of Mexico (Castro et al. 2003; Latimer and Charpentier 2010; Moore 2011; Rebich 2011). Valiela et al. (2000) reported strong isotopic evidence in support of NLM predictions that wastewater contributed nearly half of the N delivered to Waquoit Bay. Otherwise, there are relatively few stable isotope studies addressing N sources in estuaries and those that exist focus on the assimilation of sewage by macrophytes and mollusks in northeastern estuaries, such as Narragansett Bay and Cape Cod (Pruell et al. 2006; Wigand et al. 2007; Carmichael et al. 2008). While these approaches provide evidence for incorporation of anthropogenic N into the food chain, they do not reveal the contribution of sewage relative to other sources, such as atmospheric deposition. Nonetheless, a number of studies qualitatively suggest that sewage and population-related sources are important for densely populated coastal regions in the northeastern US.

Natural N sources

There was disagreement between NEWS and SPARROW in apportionment to natural N sources at both national and regional scales (Fig. 2; Table 4). For individual basins, NEWS and SPARROW predictions of dominant N sources are most comparable when only anthropogenic sources were considered because of considerable differences between models in their estimates of N originating in undeveloped lands

(Figs. 3–4). In all cases, attribution by NEWS was greater than that by SPARROW. Input data used to drive NEWS is based on the method of Cleveland et al. (1999) as a function of evapotranspiration on non-agricultural land. More recent models of N fixation based on temperature suggest that there is little to no symbiotic N-fixation in mature temperate forests (Houlton et al. 2008), which occupy large areas of the US. That said, NEWS inputs of N fixation (3.6 Tg year^{-1}) are at the low end of the range of estimates for the continental US ($0.5\text{--}12 \text{ Tg N year}^{-1}$) (Sobota et al. 2013). Model inputs cannot be easily compared because SPARROW uses forest, shrub, and barren land cover as a proxy for diffuse N from undeveloped areas. SPARROW predictions could differ from NEWS if the distribution of important N-fixing species, such as alder, had a disproportionate effect relative to their area cover (Compton et al. 2003). Similarly, atmospheric deposition could be included in the SPARROW attribution to runoff from undeveloped land because of the possibility these variables were spatially correlated. The effect of such a correlation is not clear, because SPARROW consistently attributes more coastal N to atmospheric deposition and less to natural sources than NEWS.

In contrast to SPARROW and other regional models, NEWS identified natural N-fixation as the dominant source for a number of basins along the Atlantic coast. Interestingly, regional SPARROW models for the northeastern and southeastern US considered forest and shrub land in exploratory models, but later excluded these cover types from final models because they were not significant predictors of N export (Hoos and McMahon 2009; Moore 2011). The national SPARROW model likely encompasses broader gradients of non-agricultural land cover types than regional models, which could explain why land cover was a significant predictor in the national but not regional applications. Of the regions examined, the Pacific Northwest is the only area where NEWS and SPARROW both identified natural N as the largest riverine source (Fig. 3c). Achieving substantial reductions in coastal N loading could be more difficult in this region because of the magnitude of background nutrient sources.

There were only two other models applied to the West Coast with which we could compare NEWS and SPARROW. The recently published SPARROW model for the Pacific Northwest, which is the only

SPARROW model to explicitly include an N-fixation term, also found natural N sources to be most dominant (Wise and Johnson 2011). An application of NTNI (NANI plus natural N-fixation) found fertilizer and atmospheric deposition to represent the majority of land-based N sources for select rivers along the West Coast (Schaefer et al. 2009). But most of the basins included in this NTNI study were sub-basins of NEWS-delineated catchments, so there is a scale mismatch between NEWS and NTNI.

While NEWS and SPARROW predictions were more comparable when only anthropogenic sources were considered, we do not suggest that natural N sources should be ignored in modeling efforts. In order to develop effective nutrient management strategies, it is important to understand how anthropogenic activities compare to background (natural) coastal N loading to show the magnitude of human perturbation and indicate how much potential there is to reduce coastal N loading. Better understanding the magnitude of biological N-fixation in the US (Sobota et al. 2013) and the portion of this source that is exported from catchments will inform future models.

Uncertainties

It was not possible to directly evaluate uncertainty associated with modeled source attributions because there are no appropriately scaled independent measurements of coastal N sources with which to compare model-derived estimates. In the absence of such measurements, one way to partially assess uncertainty associated with source attribution is via bootstrapping. This approach has been applied with SPARROW to estimate the range of uncertainty (90 % prediction intervals) associated with individual SPARROW model parameters in the Mississippi and Atchafalaya basins and yielded the following range of source attributions: 29–93 % for fertilizer plus crop N-fixation, 0–23 % for manure, 6–30 % for atmospheric deposition, 6–14 % for sewage and urban-related sources, and 0–13 % for natural sources (Alexander et al. 2008). Such an approach, however, does not address uncertainty associated with model structure or model inputs. Another way to assess uncertainty is to compare and contrast output from multiple models. Indeed, there are a number of modeling applications in addition to nutrient-export source attribution where no calibration data exist, such as climate change. In this

case, multiple model comparisons are the primary means to evaluate predictions, as we have done throughout this paper for source attribution. Further assessing the uncertainty associated with nutrient source attribution poses an important challenge for future studies.

Other structural factors

Aside from data inputs, differences in model structure could potentially contribute to differences in source attribution predictions between models. For example, the models treated N retention in the catchment and river network quite differently. SPARROW determined land-to-water delivery of TN as a function of six factors including temperature, soil permeability, and catchment area (Alexander et al. 2008). In-stream removal and retention of N by denitrification, sedimentation and biotic assimilation was estimated as a function of water travel time and stream depth. NEWS determined land-to-water delivery for dissolved N forms as a function of runoff (Dumont 2005; Mayorga 2010). Aquatic DIN retention was estimated by NEWS for three different sinks: denitrification, withdrawals of water for human uses, and retention in reservoirs. For DON, the only sink considered was consumptive water use and this could explain why NEWS attributed a greater source of in-stream N yield to natural N sources compared to SPARROW. If DON was excluded from NEWS source apportionment predictions for natural N, however, agreement between models did not improve, perhaps because a substantial portion of riverine N attributed to natural sources by NEWS was in the form of DIN.

Conclusions

Here we presented the first large-scale comparison of source attribution predictions for NEWS and SPARROW. The objectives of this paper were to understand important sources of N delivered to coastal areas of the US by comparing published model results. Despite differences in model structure and input data (Tables 1, 2), dominant source attribution predictions agreed for 67 % of the area of the US (Fig. 3). The area of agreement was even greater if only anthropogenic sources were considered (Fig. 4). NEWS and SPARROW source attribution predictions were most

similar where the predominant land use was agriculture (Mississippi River Basin) or undeveloped (Pacific Northwest). A principal strength of model comparisons is that agreement between models using different approaches lends credibility to predictions. Such agreement can give resource managers greater confidence and leverage in implementing and assessing actions that reduce coastal N loading. Our findings suggest that managing fertilizer losses from cropland in the Midwestern US could result in substantially reduced N export to the Gulf of Mexico and that reducing coastal N loading in the Pacific Northwest will be difficult because of the magnitude of background sources.

Predictions diverged most widely for basins with a mix of urban, agricultural, and undeveloped land. One approach to resolving these differences is to apply the models at smaller spatial scales with finer-resolution input and calibration data. Indeed, SPARROW and other models discussed in this paper have been used at regional and small-basin scales (e.g., Latimer and Charpentier 2010; Moore 2011). We found that the spatial extent of published regional applications varied widely and, as a result, comparing existing model output for specific regions of the US could be challenging. Future regional analyses of coastal N sources should consider the output of multiple models that use consistent data inputs and spatial scales (as in Alexander et al. 2002 and Valiela et al. 2002). Even in instances where model predictions agree, however, regions with heterogeneous land uses, such as along the Atlantic and California coasts, will likely require the management of N from multiples sources (such as in Chesapeake Bay, www.epa.gov/chesapeakebaytmdl).

While there was good agreement between the models in many respects, NEWS tended to attribute more to natural N and manure than SPARROW, while the reverse was true for atmospheric deposition, sewage and population-related sources, and fertilizer plus crop N-fixation (Figs. 2, 5). Areas where source attribution predictions disagreed pose a challenge because of the lack of measured N-source data with which to validate model predictions. Identifying differences in predictions of source attribution between models, as we have done here, is an important first step towards improving their performance. We found that differences in both model structure and input data contributed to differences in model predictions. Differences due to model structure are inevitable

and reflect the interests of the model developer and the specific need for which the model was developed. Differences due to model inputs might be more easily addressed. While beyond this scope of this paper, it would be informative to compare NEWS and SPARROW output that were generated using more consistent input datasets.

Acknowledgments We thank Richard Smith for a helpful review of this manuscript and, along with Richard Alexander, for providing SPARROW data. Three anonymous reviewers provided comments that greatly improved this paper. The information in this document was funded by the National Academies of Science Research Associateship Program and the US Environmental Protection Agency. It has been subjected to review by the National Health and Environmental Effects Research Laboratory's Western Ecology Division and approved for publication. Approval does not signify that the contents reflect the views of the Agency, nor does mention of trade names or commercial products constitute endorsement or recommendation for use.

References

- Alcamo J, Van vuuren DP, Cramer W (2006) Changes in ecosystem services and their drivers across the scenarios. In: Carpenter SR (ed) *Ecosystems and human well-being: scenarios*. Island Press, Washington, DC, pp 279–354
- Alexander RB, Johns PJ, Boyer EW, Smith RA (2002) A comparison of models for estimating the riverine export of nitrogen from large watersheds. *Biogeochemistry* 57:295–339
- Alexander RB, Smith RA, Schwarz GE, Boyer EW, Nolan JV, Brakebill JW (2008) Differences in phosphorus and nitrogen delivery to the Gulf of Mexico from the Mississippi River basin. *Environ Sci Technol* 42:822–830
- Barnes RT, Raymond PA, Casciotti KL (2008) Dual isotope analyses indicate efficient processing of atmospheric nitrate by forested watersheds in the northeastern US. *Biogeochemistry* 90:15–27
- Battaglin WA, Kendall C, Chang CCY, Silva SR, Campbell DH (2001) Isotopic and chemical composition of inorganic and organic water quality samples from the Mississippi River Basin, 1997–98. U.S. Geological Survey: Water-Resources Investigations Report 01-4095, Denver
- Bouwman AF, Beusen AHW, Billen G (2009). Human alteration of the global nitrogen and phosphorus soil balances for the period 1970–2050. *Global Biogeochem Cycles* 23:GB0A04. doi:10.1029/2009GB003576
- Boyer EW, Goodale CL, Jaworski NA, Howarth RW (2002) Anthropogenic nitrogen sources and relationships to riverine nitrogen export in the northeastern USA. *Biogeochemistry* 57:137–169
- Boyer EW, Alexander RB, Parton WJ, Li CS, Butterbach-Bahl K, Donner SD, Skaggs RW, Del Gross SJ (2006) Modeling denitrification in terrestrial and aquatic ecosystems at regional scales. *Ecol Appl* 16:2123–2142
- Bricker S, Longstaff B, Dennison W, Jones A, Boicourt K, Wicks C, Woerner J (2007) Effects of nutrient enrichment

- in the nation's estuaries: a decade of change. NOAA Coastal Ocean Program, Decision Analysis Series No. 26. National Centers for Coastal Ocean Science, Silver Spring, MD
- Carmichael RH, Hattenrath T, Valiela I, Michener RH (2008) Nitrogen stable isotopes in the shell of *Mercenaria mercenaria* trace wastewater inputs from watersheds to estuarine ecosystems. *Aquat Biol* 4:99–111
- Castro MS, Driscoll CT, Jordan TE, Reay WG, Boynton WR, Seitzinger SP, Styles RV, Cable JM (2001) Contribution of atmospheric deposition to the total nitrogen loads to thirty-four estuaries on the Atlantic and Gulf Coasts of the United States. In: Valigura RA, Alexander RB, Castro MS, Meyers TP, Paerl HW, Stacey PE, Turner RE (eds) Nitrogen loading in coastal water bodies: an atmospheric perspective. American Geophysical Union
- Castro MS, Driscoll CT, Jordan TE, Reay WG, Boynton WR (2003) Sources of nitrogen to estuaries in the United States. *Estuaries* 26:803–814
- Cleveland CC, Townsend AR, Schimel DS, Fisher H, Howarth RW, Hedin LO, Perakis SS, Latty EF, Von Fischer JC, Elseroad A, Wasson MF (1999) Global patterns of terrestrial biological nitrogen (N_2) fixation in natural ecosystems. *Global Biogeochem Cycles* 13:623–645
- Compton JE, Church MR, Larned ST, Hogsett WE (2003) Nitrogen export from forested watersheds in the Oregon Coast Range: the role of N-2-fixing red alder. *Ecosystems* 6:773–785
- Compton JE, Harrison JA, Dennis RL, Greaver TL, Hill BH, Jordan SJ, Walker H, Campbell HV (2011) Ecosystem services altered by human changes in the nitrogen cycle: a new perspective for US decision making. *Ecol Lett* 14:804–815
- David MB, Drinkwater LE, McIsaac GF (2010) Sources of nitrate yield in the Mississippi River Basin. *J Environ Qual* 39:1657–1667
- Davidson EA, David MB, Galloway JN, Goodale CL, Haeuber R, Harrison JA, Howarth RW, Jaynes DB, Lowrance RR, Nolan BT, Peel JL, Pinder RW, Porter E, Snyder CS, Townsend AR, Ward MH (2012) Excess nitrogen in the US environment: trends, risks, and solutions. *Issues in Ecology*
- Dentener F, Stevenson D, Ellingsen K, van Noije T, Schultz M, Amann M, Atherton C, Bell N, Bergmann D, Bey I, Bouwman L, Butler T, Cofala J, Collins B, Drevet J, Doherty R, Eickhout B, Eskes H, Fiore A, Gauss M, Hauglustaine D, Horowitz L, Isaksen ISA, Josse B, Lawrence M, Krol M, Lamarque JF, Montanaro V, Muller JF, Peuch VH, Pitari G, Pyle J, Rast S, Rodriguez J, Sanderson M, Savage NH, Shindell D, Strahan S, Szopa S, Sudo K, Van Dingenen R, Wild O, Zeng G (2006) The global atmospheric environment for the next generation. *Environ Sci Technol* 40:3586–3594
- Dodds WK, Bouska WW, Eitzmann JL, Pilger TJ, Pitts KL, Riley AJ, Schloesser JT, Thornbrugh DJ (2009) Eutrophication of U.S. freshwaters: analysis of potential economic damages. *Environ Sci Technol* 43:12–19
- Dumont E, Harrison J, Kroeze C, Bakker EJ, Seitzinger S (2005) Global distribution and sources of dissolved inorganic nitrogen export to the coastal zone: results from a spatially explicit, global model. *Global Biogeochem Cycles* 19:GB4S02. doi:10.1029/2005GB002488
- Exbrayat J-F, Viney NR, Frede H-G, Breuer L (2011) Probabilistic multi-model ensemble predictions of nitrogen concentrations in river systems. *Geophys Res Lett* 38:L12401. doi:12410.11029/12011GL047522
- Galloway JN, Aber JD, Erisman JW, Seitzinger SP, Howarth RW, Cowling EB, Cosby BJ (2003) The nitrogen cascade. *Bioscience* 53:341–356
- GeoLytics I (2000) Census 2000: demographic data and estimates
- Goolsby DA, Battaglin WA, Aulenbach BT, Hooper RP (2000) Nitrogen flux and sources in the Mississippi River Basin. *Sci Total Environ* 248:75–86
- Green PA, Vorosmarty CJ, Meybeck M, Galloway JN, Peterson BJ, Boyer EW (2004) Pre-industrial and contemporary fluxes of nitrogen through rivers: a global assessment based on typology. *Biogeochemistry* 68:71–105
- Hejzlar J, Anthony S, Arheimer B, Behrendt H, Bouraoui F, Grizzetti B, Groenendijk P, Jeuken M, Johnsson H, Lo Porto A, Kronvang B, Panagopoulos Y, Siderius C, Silgram M, Venohr M, Zaloudik J (2009) Nitrogen and phosphorus retention in surface waters: an inter-comparison of predictions by catchment models of different complexity. *J Environ Monit* 11:584–593
- Hoos AB, McMahon G (2009) Spatial analysis of instream nitrogen loads and factors controlling nitrogen delivery to streams in the southeastern United States using spatially referenced regression on watershed attributes (SPARROW) and regional classification frameworks. *Hydrol Process* 23:2275–2294
- Houlton BZ, Wang YP, Vitousek PM, Field CB (2008) A unifying framework for dinitrogen fixation in the terrestrial biosphere. *Nature* 454:327–334
- Howarth RW (2008) Coastal nitrogen pollution: a review of sources and trends globally and regionally. *Harmful Algae* 8:14–20
- Howarth RW, Billen G, Swaney D, Townsend A, Jaworski N, Lajtha K, Downing JA, Elmgren R, Caraco N, Jordan T, Berendse F, Freney J, Kudeyarov V, Murdoch P, Zhu ZL (1996) Regional nitrogen budgets and riverine N and P fluxes for the drainages to the North Atlantic Ocean: natural and human influences. *Biogeochemistry* 35:75–139
- Howarth R, Swaney D, Billen G, Garnier J, Hong B, Humborg C, Johnes P, Mörth C-M, Marino R (2012) Nitrogen fluxes from the landscape are controlled by net anthropogenic nitrogen inputs and by climate. *Front Ecol Environ* 10:37–43
- IFA/IFDC/FAO (2003) Fertilizer use by crop, 5th edn. International Fertilizer Industry Association (IFA), International Fertilizer Development Center (IFDC), and the Food and Agriculture Organization (FAO) of the United Nations, Rome
- Kellogg RL, Lander CH, Moffitt DC, Gollehon N (2000) Manure nutrients relative to the capacity of cropland and pastureland to assimilate nutrients: spatial and temporal trends for the U.S. USDA-NRCS Economic Research Service, Pub. No. nps00-0579. <http://www.nrcs.usda.gov/technical/land/pubs/manmr.html>.
- Latimer JS, Charpentier MA (2010) Nitrogen inputs to seventy-four southern New England estuaries: application of a watershed nitrogen loading model. *Estuar Coast Shelf Sci* 89:125–136

- Mayer B, Boyer EW, Goodale C, Jaworski NA, Van Breemen N, Howarth RW, Seitzinger S, Billen G, Lajtha LJ, Nosal M, Paustian K (2002) Sources of nitrate in rivers draining sixteen watersheds in the northeastern US: isotopic constraints. *Biogeochemistry* 57:171–197
- Mayorga E, Seitzinger S, Harrison AF, Dumont E, Beusen AHW, Bouwman AF, Fekete BM, Kroeze C, Van Drecht G (2010) Global nutrient export from watersheds 2 (NEWS 2): model development and implementation. *Environ Model Softw*. doi:10.1016/j.envsoft.2010.1001.1007
- McIsaac GF, David MB, Gertner GZ, Goolsby DA (2002) Relation of net nitrogen input in the Mississippi River Basin to nitrate flux in the lower Mississippi River: a comparison of approaches. *J Environ Qual* 31:1610–1622
- Moore RB, Johnston CM, Smith RA, Milstead B (2011) Source and delivery of nutrients to receiving waters in the Northeastern and mid-Atlantic regions of the United States. *J Am Water Resour Assoc* 47:965–990
- NADP (1993) Annual data summary: precipitation chemistry in the United States 1983 to 1993. National Atmospheric Deposition Program (NADP), National Resources Ecology Laboratory, Colorado State University, Fort Collins
- Nash JE, Sutcliffe JV (1970) River flow forecasting through conceptual models: a discussion of principles. *J Hydrol* 10:282–290
- NASS (2002) Census of agriculture: census quick stats: Ag statistics for U.S., state, and county. National Agricultural Statistics Service (NASS), U.S. Department of Agriculture
- Pruell RJ, Taplin BK, Lake JL, Jayaraman S (2006) Nitrogen isotope ratios in estuarine biota collected along a nutrient gradient in Narragansett Bay, Rhode Island, USA. *Mar Pollut Bull* 52:612–620
- Rebich RA, Houston NA, Mize SV, Pearson DK, Ging PB, Hornig CE (2011) Sources and delivery of nutrients to the northwestern Gulf of Mexico from streams in the south-central United States. *J Am Water Resour Assoc* 47:1061–1086
- Ruddy BC, Lorenz DL, Muellwe DK (2006) County-level estimates of nutrient inputs to the land surface of the conterminous United States, 1982–2001. U.S. Geological Survey, Scientific Investigations Report 2006-5012
- SAB (2011) Reactive nitrogen in the United States: an analysis of inputs, flows, consequences, and management options. US Environmental Protection Agency, Washington, DC, EPA-SAB-11-013
- Schaefer SC, Hollibaugh JT, Alber M (2009) Watershed nitrogen input and riverine export on the west coast of the US. *Biogeochemistry* 93:219–233
- Schlesinger WH (2009) On the fate of anthropogenic nitrogen. *Proc Natl Acad Sci USA* 106:203–208
- Silva SR, Kendall C, Wilkison DH, Ziegler AC, Chang CCY, Avanzino RJ (2000) A new method for collection of nitrate from fresh water and the analysis of nitrogen and oxygen isotope ratios. *J Hydrol* 228:22–36
- Sobota DJ, Compton JE, Harrison JA, Neale AC (2013) Reactive nitrogen inputs to land and waterways in the United States: how certain are we about sources and fluxes? *Front Ecol Environ*. doi:10.1890/110216
- Suddick EC, Davidson EA (2012) The role of nitrogen in climate change and the impacts of nitrogen-climate interactions on terrestrial and aquatic ecosystems, agriculture, and human health in the United States: a technical report submitted to the US National Climate Assessment. North American Nitrogen Center of the International Nitrogen Initiative (NANC-INI), Woods Hole Research Center, 149 Woods Hole Road, Falmouth, MA, 02540-1644 USA
- Tebaldi C, Knutti R (2007) The use of the multi-model ensemble on probabilistic climate projections. *Philos Trans R Soc A* 365:2053–2075
- Valiela I, Collins G, Kremer J, Lajtha K, Geist M, Seely B, Brawley J, Sham CH (1997) Nitrogen loading from coastal watersheds to receiving estuaries: new method and application. *Ecol Appl* 7:358–380
- Valiela I, Geist M, McClelland J, Tomasky G (2000) Nitrogen loading from watersheds to estuaries: verification of the Waquoit Bay nitrogen loading model. *Biogeochemistry* 49:277–293
- Valiela I, Bowen JL, Kroeger KD (2002) Assessment of models for estimation of land-derived nitrogen loads to shallow estuaries. *Appl Geochem* 17:935–953
- Van Drecht G, Bouwman AF, Harrison J, Knoop JM (2009) Global nitrogen and phosphate in urban wastewater for the period 1970 to 2050. *Global Biogeochem Cycles* 23:GB0A03. doi:10.1029/2009GB003458
- Vogelmann JE, Howard SM, Yang L, Larson CR, Wylie BK, Van Driel N (2001) Completion of the 1990's National Land Cover Data Set for the conterminous United States from Landsat Thematic Mapper data and ancillary data sources. *Photogramm Eng Remote Sensing* 67:650–652
- Vörösmarty C, Fekete B (2011) ISCS-CP II River routing data (STN-30p). In: Hall FG, Collatz G, Meeson B, Los S, Brown de Colstoun E, Landis E (eds) ISLSCP initiative II collection. Available via <http://daac.ornl.gov> from Oak Ridge National Laboratory Distributed Active Archive Center, Oak Ridge
- Whitall D, Hendrickson B, Paerl H (2003) Importance of atmospherically deposited nitrogen to the annual nitrogen budget of the Neuse River estuary, North Carolina. *Environ Int* 29:393–399
- Whitall D, Castro M, Driscoll C (2004) Evaluation of management strategies for reducing nitrogen loadings to four US estuaries. *Sci Total Environ* 333:25–36
- Wigand C, McKinney RA, Cole ML, Thursby GB, Cummings J (2007) Varying stable nitrogen isotope ratios of different coastal marsh plants and their relationships with wastewater nitrogen and land use in New England, USA. *Environ Monit Assess* 131:71–81
- Wise DR, Johnson HM (2011) Surface-water nutrient concentrations and sources in the United States Pacific Northwest. *J Am Water Resour Assoc* 47:1110–1135