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Global distribution and sources of dissolved inorganic nitrogen export to the coastal zone: Results from a spatially explicit, global model

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[1] Here we describe, test, and apply a spatially explicit, global model for predicting dissolved inorganic nitrogen (DIN) export by rivers to coastal waters (NEWS-DIN). NEWS-DIN was developed as part of an internally consistent suite of global nutrient export models. Modeled and measured DIN export values agree well (calibration $R^2 = 0.79$), and NEWS-DIN is relatively free of bias. NEWS-DIN predicts: DIN yields ranging from 0.0004 to 5217 kg N km^{-2} yr^{-1} with the highest DIN yields occurring in Europe and South East Asia; global DIN export to coastal waters of 25 Tg N yr^{-1}, with 16 Tg N yr^{-1} from anthropogenic sources; biological N_2 fixation is the dominant source of exported DIN; and globally, and on every continent except Africa, N fertilizer is the largest anthropogenic source of DIN export to coastal waters.

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1. Introduction

[2] In many watersheds, nutrient delivery by rivers to coastal waters has been increasing as a result of human activities such as increased use of artificial fertilizer in agriculture, population growth and cultivation of legumes [e.g., *Carpenter et al.*, 1998; *Galloway et al.*, 2004; *Vitousek et al.*, 1997]. This increased nutrient delivery has been blamed for the formation and increased extent of coastal "dead zones," as well as loss of seagrass habitat, decreases in coastal biodiversity, and increased frequency and severity of harmful and nuisance algae blooms [*Diaz et al.*, 2003]. One of the most important nutrients in this respect is nitrogen (N), because nitrogen is often the most limiting nutrient [*Justic et al.*, 1995; *Turner and Rabalais*, 1994; *Vince and Valiela*, 1973]. Dissolved inorganic nitrogen (DIN) is often the most abundant and bioavailable form of nitrogen, and therefore contributes significantly to coastal eutrophication [*Veuger et al.*, 2004].

[3] To date, three models have been applied at the global scale to model river DIN export in a spatially explicit manner [*Green et al.*, 2004; *Seitzinger and Kroeze*, 1998; *Smith et al.*, 2003]. These models have greatly improved our understanding of global patterns and magnitudes of DIN export. *Smith et al.* [2003] presented an empirical multiple regression model predicting DIN export simply as a function of runoff and population density. *Seitzinger and Kroeze* [1998] developed a river DIN export model at 1 × 1° resolution that included some DIN sources and sinks. However, important DIN sources such as N_2 fixation and manure were not included in this model. More recently, *Green et al.* [2004] have developed a model at the 0.5 × 0.5° scale that includes a wider range of input parameters as well as loss terms. However, like the other two models, the model of *Green et al.* [2004] was calibrated using data from multiple decades, leading to large differences between measurement times of inputs and outputs of these models. Furthermore, none of the three have been validated with global data not used in model calibration. Finally, none of these models were developed in a way that allows for easy comparison of output with predicted export of other nutrients or nutrient forms.

[4] Here we describe, evaluate, and apply a spatially explicit, global DIN export model called NEWS-DIN. NEWS-DIN was developed as part of a multi-investigator effort to model river export of multiple bioactive elements and element forms (particulate/dissolved, organic/inorganic) called Global Nutrient Export from WaterSheds (Global NEWS; see other papers in this special section [*Harrison et al.*, 2005a, 2005b] (see also A. H. W. Beusen et al., Estimation of global river transport of sediments and associated particulate C, N and P, submitted to *Global Biogeochemical Cycles*, 2005) (hereinafter referred to as Beusen et al., submitted manuscript, 2005)). To the extent possible, models developed within Global NEWS use the same input data (e.g., population, runoff, and basin delineation data), are calibrated or developed using data for the 1990s, have the same resolution, and have the potential to run scenarios for future nutrient export to coastal waters. Eventually, we hope to use NEWS-DIN, in conjunction with other NEWS models, to compare global spatial patterns of export of different nutrients and nutrient forms. To date, this has not

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Table 1. Overview of Data Used as Inputs and for Calibration of NEWS-DIN, Including Their Resolution and Source

Data	Resolution	Source
Land use	0.5°	*Bouwman et al.* [2005b]
Stream network and basin delineations	0.5°	STN30 [*Vörösmarty et al.*, 2000b]
Runoff	0.5°	*Fekete et al.* [2000]
Discharge	river basin	USGS [*Alexander et al.*, 1996], European Environmental Agency [*EEA*, 1998], and *Meybeck and Ragu* [1995]
Population data	country	*United Nations* [1998], *FAO* [2001] and *WorldBank* [2001]
NO_3^--N and NH_4-N concentration	river basin	USGS [*Alexander et al.*, 1996], the European Environmental Agency [*EEA*, 1998], and *Meybeck and Ragu* [1995]
Anthropogenic river water removal	river basin	*Dynesius and Nilsson* [1994]
Dam properties	subbasin	*Vörösmarty et al.* [2003]
Manure N addition	0.5°	*Bouwman et al.* [2005b]
Fertilizer N addition	0.5°	*Bouwman et al.* [2005b]
Biological N_2 fixation	0.5°	*Green et al.* [2004]
Atmospheric NO_3^--N deposition	5 × 3.75° interpolated to 0.5°	F. Dentener[a]
Crop N export	0.5°	*Bouwman et al.* [2005b]
Sewage point sources	country	*Bouwman et al.* [2005a]

[a]Dentener, F., Global patterns and magnitudes of nitrogen deposition: A revised approach, submitted to *Global Biogeochemical Cycles*, 2004.

been possible owing to the disparate input data requirements and structures of existing global nutrient export models. Here we use NEWS-DIN to estimate global DIN export for 1995 in a spatially explicit manner, and indicate dominant sources and sinks of DIN export.

2. Methodology

[5] Building on past work by *Caraco and Cole* [1999] and *Seitzinger and Kroeze* [1998], we developed a new model for predicting DIN export from watersheds (NEWS-DIN). NEWS-DIN includes several input variables missing from the original model reported by *Seitzinger and Kroeze* [1998] (N-model) such as manure N and biological N_2 fixation. NEWS-DIN also includes retention and loss terms that were absent from the original N-model, including N retention in river networks, N retention in dammed reservoirs, N loss via consumptive water use, and N loss via harvesting and grazing. NEWS-DIN also includes a more sophisticated treatment of sewage point sources than was included in the original N-model, incorporating estimates of sewage treatment, sewage connectivity, and variable N-excretion rates. The spatial resolution of input data and basin delineations was increased from 1 × 1 to 0.5 × 0.5° grids. We also used an enhanced data set for calibration and validation; the model has now been calibrated primarily for one specific period of time: 1990 to 1997, rather than for data spanning over 3 decades of measurements. This restriction gives a better temporal match between input data and DIN export data used in calibration. Finally, we use more river basins for calibration than was used in formulation of the original N-model (61 versus 35 basins).

2.1. Model Form and Input Data

2.1.1. NEWS-DIN

[6] NEWS-DIN can be summarized as

$$DIN = FE_{riv} \cdot \left[DIN_{sew} + \left(FE_{ws} \cdot TN_{diff}\right)\right], \tag{1}$$

where *DIN* is modeled DIN yield per river basin (kg N km^{-2} yr^{-1}), DIN_{sew} is DIN from sewage point sources (kg N km^{-2} yr^{-1}) and TN_{diff} is total nitrogen (TN) from diffuse sources that is mobilized from the watershed soils and sediments (kg N km^{-2} yr^{-1}). FE_{riv} is a river export fraction representing the fraction (0–1) of total point and diffuse DIN inputs to the river that is exported as DIN, FE_{ws} is a watershed export fraction representing the fraction (0–1) of TN from diffuse sources in the watershed that leaches to rivers as DIN (see the Notation section). To estimate total DIN export per basin (kg N basin^{-1} yr^{-1}) we multiplied DIN yield by basin area (km^2, derived from *Vörösmarty et al.* [2000a, 2000b]).

[7] Sub-models and input data used to calculate the terms in equation (1) are described in sections 2.1.2 to 2.1.5. Most input data (Table 1) were available as a 0.5 × 0.5° grid and were selected to be representative of the years between 1990 and 1997. Before using this gridded data in the model, it was averaged over river basins delineated from an updated version of the 0.5 × 0.5° STN30-p global river network [*Vörösmarty et al.*, 2000a, 2000b], unless stated otherwise. Many input data sources, such as those for land use, basin delineations, runoff, discharge, population, dam properties, fertilizer, manure, harvesting losses, and sewage are the same as those used for other Global NEWS models (see other papers in this special section) including NEWS-DOC, NEWS-DON, NEWS-DOP [*Harrison et al.*, 2005b], NEWS-DIP [*Harrison et al.*, 2005a] and a particulate export model by Beusen et al. (submitted manuscript, 2005).

2.1.2. Aquatic Retention

[8] Aquatic retention ($1 - FE_{riv}$) is the retention of DIN (0–1) in reservoirs and in the STN-30 basin river network. The fraction of river DIN inputs exported to the coastal zone as DIN, FE_{riv}, is defined as

$$FE_{riv} = (1 - L_{den}) \cdot (1 - Q_{rem}) \cdot (1 - D), \tag{2}$$

where ($1 - L_{den}$) is the fraction of DIN not retained in river reaches, ($1 - D$) is the fraction of DIN not retained in dammed reservoirs, and ($1 - Q_{rem}$) is the fraction that is not diverted to other basins or removed for irrigation.

[9] DIN loss by denitrification during transport throughout entire river networks (L_{den}) is estimated according to *Seitzinger et al.* [2002] as

$$L_{den} = c \cdot \ln(A) - d, \tag{3}$$

where A is basin area (km^2) from *Vörösmarty et al.* [2000a, 2000b] and c and d are fitted coefficients equal to 0.0605 and 0.0443, respectively ($r^2 = 0.88$), using data on A and modeled L_{den} for 16 rivers in the northeastern United States as in the work by *Seitzinger et al.* [2002]. Estimation of c and d was done using rivers with an L_{den} smaller than 0.65. Therefore the maximum L_{den} was set at 0.65 to avoid extrapolation error.

[10] The impact of anthropogenic removal of river water (containing DIN) was estimated as

$$Q_{rem} = \frac{Q_{irr} + Q_{div}}{Q_{nat}}, \tag{4}$$

where Q_{rem} is the fraction of DIN retained, owing to the anthropogenic removal of river water (containing DIN), Q_{div} is the amount of discharged water lost from the river by anthropogenic transfer of water out of the basin (km^3 yr^{-1}), Q_{irr} is the amount of discharge removed for irrigation, minus the amount of irrigation water that ultimately flows back into the river (i.e., Q_{irr} is extracted irrigation water that evaporates on irrigated fields) (km^3 yr^{-1}), and Q_{nat} is the river discharge before any direct human manipulations on the river system (km^3 yr^{-1}). Values for Q_{irr}, Q_{div}, and Q_{nat} were from *Dynesius and Nilsson* [1994]. If these values were unavailable, then Q_{rem} was assumed to equal 0.039, the average of Q_{rem} for the 115 basins where Q_{irr} and Q_{div} were available.

[11] DIN retention in reservoirs (D) within a river basin is modeled according to *Seitzinger et al.* [2002] as

$$D = \frac{1}{Q} \cdot \sum_{i=1}^{n} 0.8845 \cdot \left(\frac{DEPT_i}{Rt_i}\right)^{-0.3677} \cdot Q_i, \tag{5}$$

where $DEPT_i$ is reservoir depth (m), Rt_i is water residence time (years), and i is the reservoir identification number within a basin ($\{i = 1 \ldots i = n\}$). The retention in each reservoir of a river basin is aggregated to a basin average (D) by taking a weighted arithmetic average of the retention in reservoirs. The modeled retention in a reservoir is weighted by the fraction of total basin discharge that the reservoir intercepts. Q is total basin discharge and Q_i is the discharge intercepted by dam i. Maximum D is set at 0.965, because that is the maximum D in the set of basins used for calibration. Per reservoir values for $DEPT_i$, Rt_i, and Q_i were from *Vörösmarty et al.* [2003].

2.1.3. Point Sources

[12] DIN input into rivers from point sources is estimated by first estimating the amount of TN from human excreta and industrial wastewater in sewage effluents, and by subsequently multiplying this amount with an estimated fraction of TN that is DIN in sewage effluents. TN in sewage effluents is estimated as described by *Bouwman et al.* [2005a],

$$TN_{sew} = H \cdot (1 - T_N) \cdot I \cdot E_N, \tag{6}$$

where TN_{sew} is the annual sewage TN discharged to surface water per km^2 of basin (kg N km^{-2} yr^{-1}), H is population density (individuals km^{-2}) from *United Nations* (*UN*) [1998], *Food and Agriculture Organization* (*FAO*) [2001] and *WorldBank* [2001], T_N is a country by country fraction of TN removed by wastewater treatment compiled by *Bouwman et al.* [2005a], I is a country by country fraction of population connected to sewer systems compiled by *Bouwman et al.* [2005a], and E_N is the per capita human TN emission estimated as described by *Bouwman et al.* [2005a] (kg N individual^{-1} yr^{-1}). For those countries where I and T_N were not available on a country by country basis, we used regional estimates according to *Bouwman et al.* [2005a].

[13] Sewage effluent DIN emitted to rivers is estimated as

$$DIN_{sew} = TN_{sew} \cdot \left[0.485 + \frac{T_N}{\max(T_N)} \cdot 0.255\right], \tag{7}$$

where DIN_{sew} is DIN in sewage effluents for a basin (kg N km^{-2} yr^{-1}), max is the maximum of all countries for which T_N is known ($\max(T_N) = 0.8$ in Finland), 0.485 is an estimate of the fraction of TN that is DIN in sewage effluent [*Seitzinger*, 1995] without treatment, and 0.255 is the maximum increase in DIN to TN ratio that can be achieved by sewage treatment [*Seitzinger*, 1995].

2.1.4. Watershed Export

[14] The watershed export fraction, FE_{ws}, in equation (1) is the fraction of TN from diffuse sources that is transported as DIN from soils to rivers. In NEWS-DIN, FE_{ws} is defined as

$$FE_{ws} = e \cdot R, \tag{8}$$

where R is runoff (m yr^{-1}) and, e, the watershed export coefficient, is a calibrated parameter that defines the slope of the assumed linear relationship between R and FE_{ws}. Values for R were obtained from *Fekete et al.* [2000]. FE_{ws} was not allowed to exceed a value of 1.

[15] This way of defining transport of DIN from soils to rivers is consistent with *Caraco and Cole* [1999], who used a similar relationship for transport of NO_3^- from soils to rivers. Runoff has been found to be an important predictor of DIN transport through soils, brooks and small river reaches [*Behrendt and Opitz*, 2000; *Goolsby et al.*, 2000].

2.1.5. Diffuse Sources

[16] The total amount of diffuse source nitrogen that is mobilized annually (TN_{diff}) (kg N km^{-2} yr^{-1}) is estimated in NEWS-DIN as

$$TN_{diff} = TN_{am} + TN_{fe} + TN_{dep} + TN_{fix} - TN_{exp}, \tag{9}$$

where TN_{am} is animal manure N addition (kg N km^{-2} yr^{-1}), TN_{fe} is fertilizer N addition (kg N km^{-2} yr^{-1}), TN_{dep} is atmospheric NO_y deposition (kg N km^{-2} yr^{-1}), TN_{fix} is biological N_2 fixation (kg N km^{-2} yr^{-1}) and TN_{exp} is N in crops and grassland that is removed from the land by harvesting and grazing (kg N km^{-2} yr^{-1}). TN_{am}, TN_{fe} and TN_{exp} were calculated as by *Bouwman et al.* [2005b]. TN_{dep} is annual average deposition of atmospheric NO_y-N modeled for 1995 [*Lelieveld and Dentener*, 2000]. TN_{fix} includes both natural and agricultural biological N_2 fixation. Biological N_2 fixation data were obtained from *Green et al.*

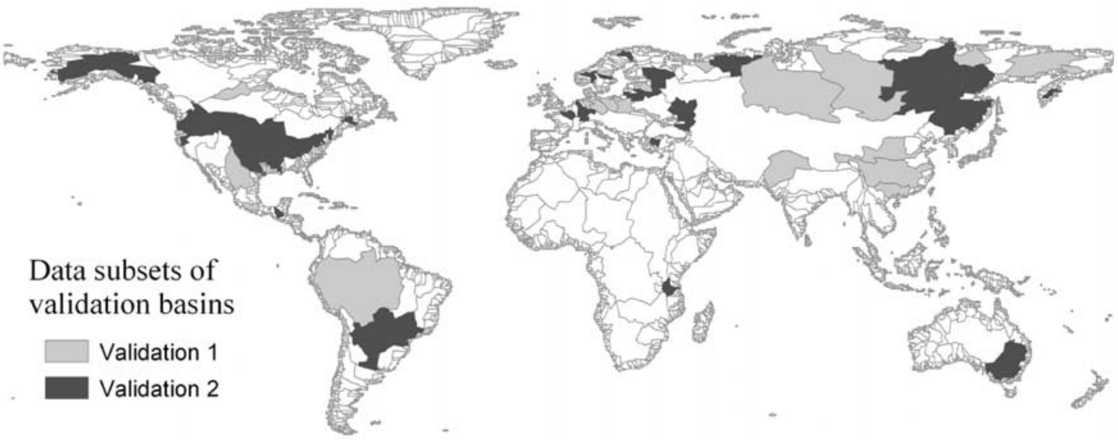

Figure 1. Geographic distribution of basins used to calibrate and validate NEWS-DIN (see section 2.2.2).

[2004]. Subtraction of TN_{exp} in equation (9) makes it so that TN_{diff} represents only the fraction of N from diffuse sources that is directly available for leaching to surface water, thereby avoiding double counting of N inputs. For example, in equation (9) we do not count fertilizer N that is removed by harvesting or grazing, and that is subsequently available for leaching to surface water after reapplication as animal manure or as point source from human sewage.

[17] Equation (9) is separately applied to each 0.5 × 0.5° grid cell. In individual cases, calculated TN_{diff} may be negative because crop and grazing N export exceeds N additions. However, we set negative TN_{diff} values to zero, because there is generally not a net flux of N from rivers to agricultural fields. Subsequently, TN_{diff} for each grid cell was averaged over the basin and this average TN_{diff} was used in equation (1).

[18] In order to avoid double counting of deposited NH_x that has volatilized from manure produced in the same year and basin, we did not include NH_x deposition in equation (9).

2.1.6. Source Contributions

[19] In evaluating model output, we separately estimated the contributions of atmospheric NO_y deposition, fertilizer addition, manure addition and agricultural and non-agricultural biological N_2 fixation to DIN export. This was done by removing the point sources term (DIN_{sew}) from equation (1) and by replacing TN_{diff} with the amount of N mobilized from atmospheric NO_y-N deposition, fertilizer N addition, manure N addition, agricultural N_2 fixation or non-agricultural N_2 fixation, respectively. The contribution to exported DIN from manure, fertilizer and agricultural N_2 fixation inputs was obtained by multiplying the amount of N from each of these sources by the fraction remaining after harvest and grazing. This fraction (G) was calculated as

$$G = 1 - \frac{TN_{\mathrm{exp}}}{TN_{am} + TN_{fe} + \left(0.23 \cdot TN_{fix}\right)}, \tag{10}$$

where 0.23 is an estimate of the fraction of TN_{fix} occurring on agricultural land, calculated on the basis of work by *Galloway et al.* [2004]. We assumed that NO_y deposition and natural N_2 fixation were not subject to removal of N by harvesting or grazing.

[20] We also estimated the contribution of point sources to model output by removal of TN_{diff} from equation (1).

[21] Source contributions are either expressed in Tg N yr^{-1} or as a fraction of model output, where the source with the highest fraction is referred to as the dominant source.

2.1.7. Measured DIN Export From Watersheds

[22] Discharge, DIN concentration, and basin surface area data for 61 basins were compiled from several sources for use in model calibration and validation (Appendix A). Together, these 61 basins account for 33% of global exoreic discharge. We restricted our data set to include only long-term (>4 years) annual averages with at least 85% of the measurements taken between 1990 and 1997. These annual averages were obtained from *Meybeck and Ragu* [1995] (median), *European Environment Agency* [1998] (median), and *U.S. Geological Survey* (*USGS*) [2003] (arithmetic average). We also excluded basins encompassing fewer than 11 grid cells owing to uncertainties associated with smaller basins [*Harrison et al.*, 2005a; *Vörösmarty et al.*, 2000b]. Despite these requirements for inclusion, both calibration and validation data sets include basins with a broad range of sizes, land uses, climates, topographies, and ecosystems (Figure 1). Data on DIN yield have an interannual variation ranging from a factor 2 to 13 [*USGS*, 2003].

[23] Measured DIN yield was obtained as follows:

$$DIN_{meas} = \left(\left[NO_3^-\right] + \left[NH_4^+\right]\right) \cdot \frac{Q}{A}. \tag{11}$$

DIN_{meas} is DIN yield near the river mouth (kg N km^{-2} yr^{-1}). $[NO_3^-]$ and $[NH_4^+]$ are measured NO_3^--N and NH_4^+-N concentrations, respectively, measured near the river mouth (kg N km^{-3}). Q is measured basin discharge (km^3 yr^{-1}) and A is basin area (km^2).

2.2. Calibration and Model Analyses

2.2.1. Model Fit

[24] Two indicators of model fit are used in this paper: model efficiency and model error. The quality of the one to

Table 2. Metrics of Model Performance During Validation of NEWS-DIN and Three Other Global DIN Export Models on Two Validation Subsets (After Being Calibrated on Independent Subsets of Rivers)[a]

			R^2		Model Errors, %					
Model	Validation	Fitted Parameter	Yield	Export	IQR[b]	Minimum	25th	Median Error	75th	Maximum
NEWS-DIN	Val. 1	1.2[c]	0.78	0.83	168	−78	−41	19	127	332
	Val. 2	1.1[d]	0.54	0.72	109	−74	−49	−21	60	781
Smith et al. [2003]	Val. 1	na	0.36	0.71	130	−88	−51	29	79	1513
	Val. 2	na	0.20	0.59	146	−81	−45	34	102	488
Seitzinger and Kroeze [1998]	Val. 1	na	0.40	0.56	214	−97	−84	6	130	2155
	Val. 2	na	0.33	0.52	124	−95	−74	−23	51	1070
Green et al. [2004]	Val. 1	na	0.54	0.52	157	−96	−58	6	99	541
	Val. 2	na	0.25	0.25	162	−99	−51	3	111	623

[a] Data sets Val. 1 and Val. 2 contained 30 and 31 basins, respectively. Model errors (%) and R^2 are computed as defined in section 2.2.1 for DIN export (kg N yr^{-1}) and DIN yield (kg N km^{-2} yr^{-1}). Abbreviation na denotes "not applicable."

[b] IQR denotes interquartile range (difference between the 25th and 75th percentiles of the distribution of errors).

[c] This is parameter e_1.

[d] This is parameter e_2.

one linear relationship between the logarithm of measurements and model estimates is expressed as model efficiency (R^2, distinct from r^2, the coefficient of determination) [*Nash and Sutcliffe*, 1970].

[25] Model error, *ME* (%), is expressed for the *i*th basin according to *Alexander et al.* [2002],

$$ME_i = \frac{Mod_i - Obs_i}{Obs_i} \cdot 100, \qquad (12)$$

where *Obs* is observed DIN export for a basin and *Mod* is modeled DIN export for the same basin.

2.2.2. Model Calibration and Validation

[26] Equations (3) and (8) contain calibrated parameters. The watershed export coefficient (*e*; equation (8)) was calibrated separately from parameters *c* and *d* (equation (3)). Equation (3) was calibrated using model-predicted %N removed in river networks [*Seitzinger et al.*, 2002]. Parameter *e* in equation (8) was calibrated by optimizing the linear one to one relation between log measurements and log model outputs for 61 basins, hereinafter referred to as the calibration basins.

[27] To validate, the set of available DIN export measurements (n = 61) was split randomly into two subsets of 31 and 30 measurements. These two data subsets (subsequently referred to as Data subset 1 and Data subset 2) were given a basin area frequency distribution that was as equal as possible. As a result, the basins belonging to each subset covered similar surface areas and each basin area class is equally represented in the two subsets (Figure 1). In the first validation (Validation 1), Data subset 2 was used to recalibrate parameter *e* (equation (8)) and Data subset 1 was used to evaluate model fit. In the second validation (Validation 2), data subset 1 was used to recalibrate parameter *e* and Data subset 2 was used to evaluate model fit. This approach was taken in order to avoid equifinality [*Beven*, 1993]. The recalibrated values of parameter *e* for Validation 1 and 2 are hereinafter referred to as e_1 and e_2, respectively.

2.2.3. Model Efficiency and Sensitivity Analyses

[28] We conducted a model efficiency analysis, in order to test the relative contribution of NEWS-DIN's model parts in explaining DIN export. In this analysis, we evaluated how model efficiency (R^2) changed when individual model parts, such as *D* or TN_{dep}, were removed sequentially. Changes in R^2 are used to calculate the fraction of otherwise unexplained variation that is explained by inclusion of the removed model part.

[29] To evaluate model sensitivity we calculated the change in modeled DIN yield for each basin resulting from 5% changes in model inputs and parameters. We then used this to calculate the global mean and maximum basin change.

3. Results and Discussion

3.1. NEWS-DIN Performance

[30] Despite substantial uncertainties associated with model inputs, a comparison of modeled versus measured DIN yield and export indicates that NEWS-DIN's predictive capacity is quite high. NEWS-DIN explains 54–78% of the variability in DIN yield (kg N km^{-2} yr^{-1}), and 72–83% of DIN export (kg N $basin^{-1}$ yr^{-1}) in validation basins (Table 2). Median model error ranges between −21 and 19% and the interquartile range (IQR) between 109 and 168% in validation basins (Table 2). The values of the calibrated parameter, e_1 and e_2, differed only slightly after being calibrated on either of the two independent data subsets, suggesting that the uncertainty in this parameter is low. Model performance of *Smith et al.* [2003] and *Seitzinger and Kroeze* [1998] when validated on Data subset 1 (Validation 1) and Data subset 2 (Validation 2) was lower than found for NEWS-DIN, regarding fit (R^2), precision (IQR) and bias (absolute median error). The model of *Green et al.* [2004] had smaller bias, equal precision, and lower fit, compared to NEWS-DIN when validated on Data subsets 1 and 2.

[31] Fit, precision and bias obtained during calibration on the calibration basins are similar to those obtained during validation. Parameter *e* was set to 1.1 by calibration. On calibration basins, 70% of the variability in DIN yield (kg N km^{-2} yr^{-1}) (Figure 2a), and 79% of DIN export (kg N $basin^{-1}$ yr^{-1}) (R^2 = 0.79) (Figure 2b) was explained. Distribution of residuals in Figures 2a and 2b suggests that NEWS-DIN yield and export estimates are relatively unbiased. The median percent error of 7% also suggests a low model bias. The calibration R^2 value for

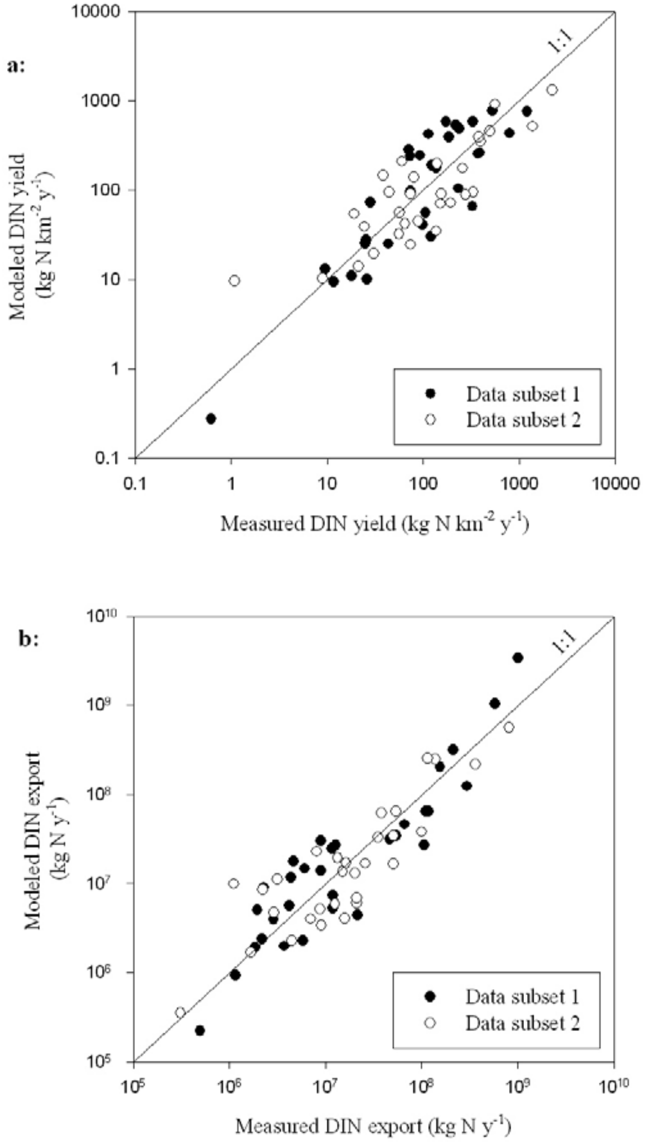

Figure 2. (a) Modeled versus measured DIN yield (kg N km^{-2} yr^{-1}). $R^2 = 0.70$. (b) Same as Figure 2a, but for DIN export (kg N yr^{-1}). $R^2 = 0.79$.

DIN yield (0.70) is slightly greater than calibration values reported for DIN yield for two other comparable calibrated global models: 0.59 [*Smith et al.*, 2003], and 0.68 [*Green et al.*, 2004].

3.2. Predicted Spatial Distributions

3.2.1. DIN Export

[32] Predicted DIN yields spanned 7 orders of magnitude, ranging from 0.0004 to 5217 kg N km^{-2} yr^{-1} (Figure 3). NEWS-DIN's highest DIN yields were typically predicted for tropical humid basins such as the Amazon and the Zaire, densely populated basins with a high GDP (gross domestic product) such as the Rhine and the Thames (England), and basins with much intensive agriculture such as the Ganges and the Chang Jiang (Eastern China). Lowest yields were predicted for basins with low population densities such as most basins at high latitudes, and also for basins in arid regions such as the Tamanrasett (Africa) or the Nile.

[33] Largest DIN export by basin was predicted for the Amazon (3.4 Tg N yr^{-1}), followed by the Ganges (2.2 Tg N yr^{-1}), Chang Jiang (1.0 Tg N yr^{-1}), Zaire (0.8 Tg N yr^{-1}) and Mississippi (0.6 Tg N yr^{-1}). Together, these high export-basins account for 32% of the global total.

[34] NEWS-DIN-predicted distribution of high and low DIN yields is somewhat different from that predicted by other DIN export models. For example, *Seitzinger and Kroeze* [1998] predicted a much lower DIN yield in humid tropical areas than is predicted by NEWS-DIN. This can be explained by the fact that their model did not account for biological N_2 fixation. *Green et al.* [2004] also predicted much lower DIN yields in tropical areas than NEWS-DIN. This is probably due the fact that the *Green et al.* [2004] model includes a positive relationship between N retention and temperature, whereas NEWS-DIN does not.

3.2.2. Source Contributions to River DIN Export

[35] According to NEWS-DIN, biological N_2 fixation is the dominant source of exported DIN (not to be confused with N loading onto land) over much of the Earth's surface (Figure 4). N_2 fixation constitutes the dominant source of DIN export to the coast in many tropical, subtropical and boreal basins, including basins in Canada, Russia, central Africa, Indonesia and Brazil (Figure 4). Anthropogenic N, especially from fertilizer, is the dominant source of DIN export in southern and eastern Asia, western Europe, and the central United States. Fertilizer is the dominant source of exported DIN in two thirds of the basins with DIN yields exceeding 1000 kg N km^{-2} yr^{-1}. Sewage point sources are predicted to dominate DIN export mainly in arid basins (in, for example, Mexico, north Africa, west Australia, Arabia). This is because arid basins have low modeled export of DIN from diffuse sources, owing to a low FE_{ws} (equation (8)). Sewage point sources are also often the dominant source of DIN export in numerous small ($\leq$15,000 km^2) densely populated basins, owing to high human N emission and their relatively low predicted aquatic retention (section 3.3). NEWS-DIN predicts that manure addition is mainly the dominant source of exported DIN in parts of the eastern United States (e.g., Florida, Georgia), south-eastern Australia and Argentina.

[36] The magnitude of predicted basin DIN export is related to the NEWS-DIN-predicted dominant DIN source. Largest DIN export is generally predicted to occur in basins where fertilizer dominates DIN export, whereas lowest DIN export is often predicted to occur in basins in which point sources dominate DIN export.

3.3. DIN Retention

[37] Global patterns of N retention and their underlying causes are relatively uncertain [*Nixon et al.*, 1996; *Seitzinger*, 1988; *Van Breemen et al.*, 2002]. NEWS-DIN enables us to make predictions regarding global patterns of DIN retention in river systems.

[38] Owing to model assumptions, NEWS-DIN predicts that the highest aquatic retention is in basins with dams on

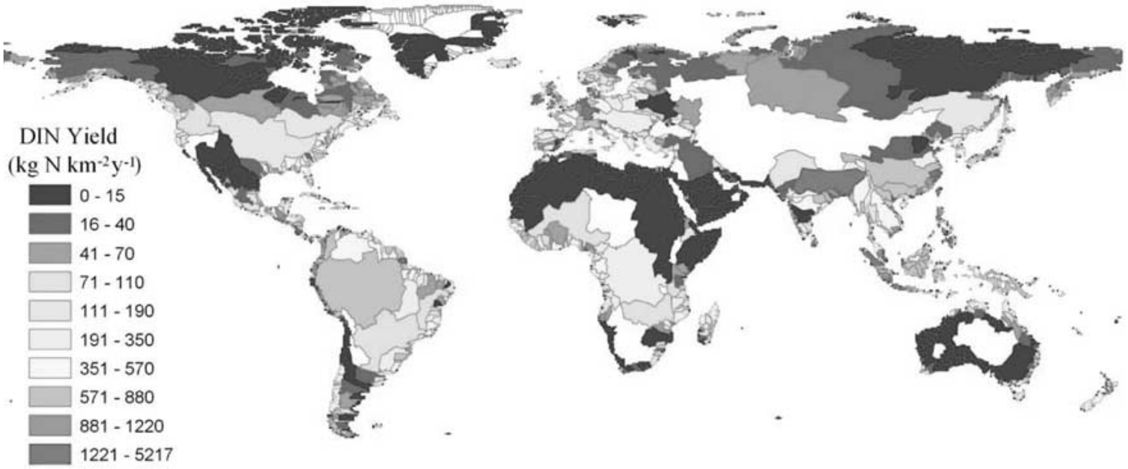

Figure 3. Modeled DIN yield by exoreic basin in kg N km^{-2} yr^{-1}. See color version of this figure at back of this issue.

their main stem such as the Rio Grande or Huang He. Lowest aquatic retention was generally predicted in small basins close to oceans.

[39] Dams greatly increased predicted DIN retention in some rivers. For example, in dam-influenced rivers such as the Colorado, Rio Grande, Orange, and Huang He, the predicted dam induced retention ranged from 16% to 97% percent of total aquatic retention. However, according to NEWS-DIN, dam induced retention has a relatively small impact on DIN retention at the global scale. NEWS-DIN predicts that just 6% of the DIN retained in aquatic systems globally is due to dams. This is consistent with an analysis by *Seitzinger et al.* [2002] indicating average contribution of 2% by dams to aquatic retention in 16 northeastern U.S. watersheds with small to medium sized dammed reservoirs. Nevertheless, dams will likely become more important in the future because the number of dams is projected to increase much in the coming decades [*Vörösmarty et al.*, 2003].

[40] The percentage of N loaded onto basins from point and non-point sources (both anthropogenic and natural) exported as DIN ranged from 0.0001% to 43% for basins larger than ten 0.5 × 0.5° grid cells (Figure 5). Lowest retention rates were predicted for basins with high runoff (>1 m yr^{-1}), because FE_{ws} is one in these basins (section 2.1.4). Highest retention rates were predicted for basins with extensive damming or very low annual precipitation and runoff. According to NEWS-DIN, in exoreic dry areas of Africa, Asia, Australia and North America where runoff is less than 0.1 m yr^{-1} (30% of the exoreic world), 95% (on average) of N applied to watersheds is not exported to coastal waters as DIN. Export from these arid systems is predicted to be low because there is very little runoff to transport soluble nitrogen (nitrate) from the soil and unsaturated zone to streams.

3.4. Global and Regional Analyses

[41] NEWS-DIN predicts a global rate of river DIN export of 24.8 Tg N yr^{-1}. NEWS-DIN's estimate is somewhat higher than other estimates for the global total of DIN export: 20.8 Tg N yr^{-1} by *Seitzinger and Kroeze* [1998], and 14.5 Tg N yr^{-1} by *Green et al.* [2004]. The ratio of

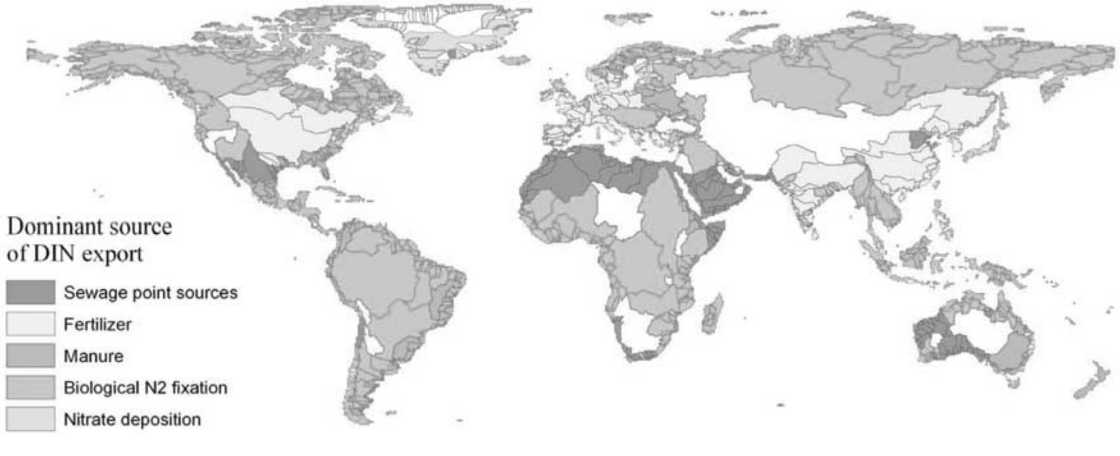

Figure 4. NEWS-DIN-predicted dominant sources of DIN export in exoreic basins. See color version of this figure at back of this issue.

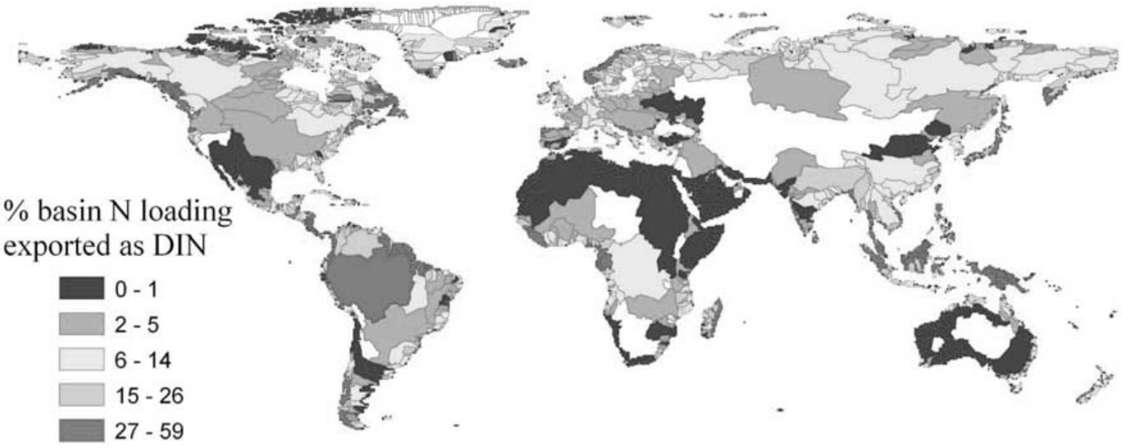

Figure 5. Percentage of N loaded onto basins from point and nonpoint sources that NEWS-DIN predicts is exported to coastal waters as DIN. See color version of this figure at back of this issue.

NEWS-DIN's prediction of global DIN export to predictions of global TN export (48.7 Tg N yr^{-1} [*Galloway et al.*, 2004], 54 Tg N yr^{-1} [*Van Drecht et al.*, 2003], 40 Tg N yr^{-1} [*Green et al.*, 2004]) suggests that DIN is responsible for 46–62% of global TN export by rivers.

[42] NEWS-DIN predicts that of the 24.8 Tg N yr^{-1} exported by rivers, 15.8 Tg N yr^{-1} is anthropogenic. *Meybeck* [1982] estimated global anthropogenic DIN export as 7 Tg yr^{-1} for the late 1970s. This suggests an approximate doubling of anthropogenically derived DIN export between the late 1970s and the mid 1990s. This is reasonable considering that sources of N to surface waters have increased dramatically, including a 1.6-fold increase in world population [*UN*, 1998], increase in animal populations [*FAO*, 1996], 2.6-fold increase in rates of N fertilizer application [*Bouwman et al.*, 2005b], and the widespread development of enhanced sewer systems, which more efficiently export sewage, and hence DIN, to rivers. Our estimate of DIN export from natural sources (9.3 Tg yr^{-1}) is higher than, previous estimates of non-anthropogenically derived DIN export (5 Tg N yr^{-1} [*Meybeck*, 1982; *Seitzinger and Kroeze*, 1998] and 2.4 Tg N yr^{-1} [*Green et al.*, 2004]). The much lower estimate of natural DIN export by *Green et al.* [2004] is probably due to an assumption that a very small fraction of modeled organic nonpoint sources (N_2 fixation and manure) is exported as DIN. Furthermore, as mentioned previously, *Green et al.* [2004] assume that DIN delivery to coastal waters is reduced by high temperatures, reducing their predicted DIN export from tropical basins where natural sources are relatively important. The proportion of NEWS-DIN-modeled riverine DIN from natural sources (36%) is slightly lower than found for TN by *Van Drecht et al.* [2003] (51%). This difference is consistent with the notion, supported by existing data [*Van Breemen*, 2002], that natural systems export N mainly in forms other than DIN.

[43] NEWS-DIN predicts that of all the continents, Asia exports the most DIN to its coasts, owing in part to its large surface area, but also to its large population and cultivated land area. This is consistent with predictions by previous DIN export models [e.g., *Seitzinger and Kroeze*, 1998]. The relative share of DIN export from Africa and South America in global DIN export predicted by NEWS-DIN is higher than predicted by *Seitzinger and Kroeze* [1998]. Absolute NEWS-DIN predicted DIN export from Africa and South America is also higher than predicted by *Seitzinger and Kroeze* [1998]. This is probably due to the fact that N_2 fixation, which is a relatively important source of DIN export on these two continents, is not explicitly represented in the model used by *Seitzinger and Kroeze* [1998].

[44] According to NEWS-DIN, biological N_2 fixation is the dominant DIN export source for the majority of continents, including North America, South America, Africa and the aggregate of Australia, Indonesia and Oceania, where it is predicted to account for 39, 73, 72 and 70% of exported DIN, respectively. There is substantial variation in other predicted DIN sources between continents. Contributions to DIN export by manure and fertilizer vary most between continents. Contribution to continental DIN export by manure and fertilizer spans from 10 to 30% and from 1 and 39%, respectively (Figure 6).

[45] According to NEWS-DIN, natural N_2 fixation is the single largest source of river exported DIN globally, accounting for 36% (9 Tg N yr^{-1}) of the total global DIN exported from watersheds. Inorganic fertilizer application is the second most important source of river-exported DIN, accounting for 21% (5.3 Tg N yr^{-1}) of the total global export. Manure application, agricultural N_2 fixation, NO_y deposition, and sewage point sources, are less important, though still significant, sources of DIN to coastal waters, accounting for 18, 15, 8, and 2% of global DIN export, respectively (4.5, 3.8, 1.9, and 0.4 Tg N yr^{-1}). NEWS-DIN predicts that agricultural sources of DIN (inorganic fertilizer, animal manure and agricultural N_2 fixation) account for about half of the total DIN export globally.

3.5. Efficiency and Sensitivity of Model Parts

[46] We analyzed the change in model efficiency upon removing different parts of NEWS-DIN (2.2.3). For the modeled DIN yield per basin, the change in model efficiency ranged from 0 to 87% depending upon the portion of the model removed (Table 3). The largest decrease

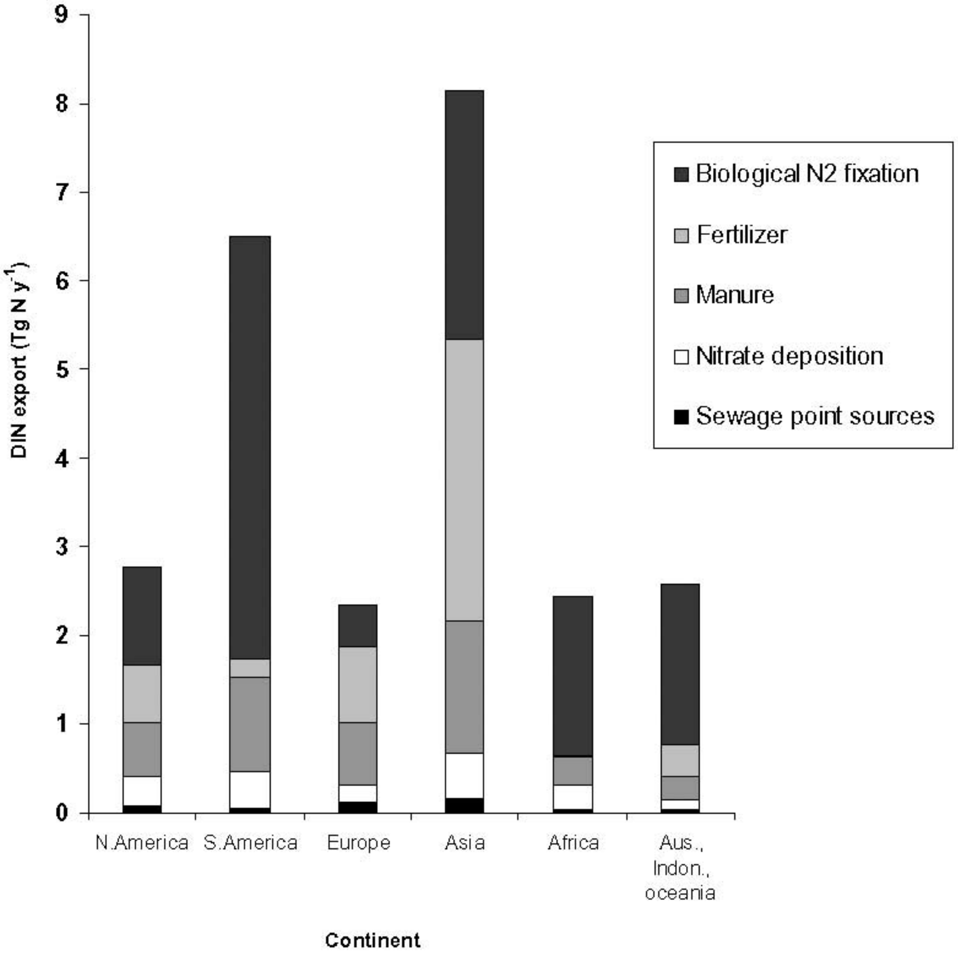

Figure 6. Modeled DIN export to the coastal zone of each continent by modeled source.

in model efficiency occurred when *R* or river network retention terms were removed from NEWS-DIN. This suggests that future improvements to NEWS-DIN may depend much upon a better understanding of river network retention and the factors controlled by runoff within watersheds.

[47] Removal of the sewage point source term from NEWS-DIN decreased model efficiency only slightly, which is consistent with the low model sensitivity for sewage (Table 4). Removing the calibrated watershed export coefficient, *e*, from NEWS-DIN also decreased model efficiency only slightly, suggesting that NEWS-DIN is relatively insensitive to uncertainty in this calibrated parameter. The small change in model efficiency resulting from removal of irrigation and river diversion terms from NEWS-DIN suggests that anthropogenic river water removal has a relatively small impact on DIN export at the global scale. However, at a local scale, anthropogenic river water removal can have an important impact on DIN export. For example, in the Colorado (southwest United States) and the Eastmain (east Canada), anthropogenic river water removal accounts for 99.5% and 93% of DIN retention, respectively.

[48] The effect of 5% changes in inputs and parameters on the mean percent change in output (basin DIN export or yield) ranges from 0.4 to 9.6% (Table 4). Highest mean percentage change in output was found for coefficient *c* (equation (3)). Sensitivity analysis suggests that NEWS-DIN predictions are quite sensitive to treatment of its retention terms (Table 4), which is consistent with results of the efficiency analyses. In this regard, NEWS-DIN is similar to previous DIN export models [e.g., *Seitzinger et al.*, 2002; *Smith et al.*, 1997]. Despite the sensitivity of DIN

Table 3. Decrease in NEWS-DIN Model Efficiency as a Function of Removal of Individual Model Components[a]

Model Component	Percent Decrease in Model Efficiency Resulting From Component Removal
River network retention, fraction	61
Anthropogenic removal of river water, fraction	2
Dam reservoirs, fraction	36
Sewage point sources, kg N km^{-2} yr^{-1}	0
e (equation (8))	1
Runoff, m yr^{-1}	87
Atmospheric NO_3^--N deposition, kg N km^{-2} yr^{-1}	12
Net biological N_2 fixation, kg N km^{-2} yr^{-1}	46
Fertilizer, kg N km^{-2} yr^{-1}	63
Manure, kg N km^{-2} yr^{-1}	36
Harvesting and grazing N removal, kg N km^{-2} yr^{-1}	18

[a] Described in section 2.2.3.

Table 4. Results of Sensitivity Analysis: Mean and Maximum Change in Modeled DIN Yield Per Basin as a Function of Changing Inputs and Model Parameters by +5% and −5%

Input or Parameter	Mean Output Change, %	Maximum Change, %
River network retention, fraction	5.0	5
c (equation (3))	9.6	31
d (equation (3))	0.4	1
Anthropogenic removal of river water, fraction	2.5	995
Dam reservoirs, fraction	0.6	25
Sewage point sources, kg N km^{-2} yr^{-1}	0.7	5
e (equation (8))	3.8	5
Runoff, m yr^{-1}	3.8	5
Atmospheric NO_3^- deposition, kg N km^{-2} yr^{-1}	0.7	5
Net biological N_2 fixation, kg N km^{-2} yr^{-1}	2.7	5
Fertilizer, kg N km^{-2} yr^{-1}	0.9	7
Manure, kg N km^{-2} yr^{-1}	1.1	6
Harvesting and grazing N removal, kg N km^{-2} yr^{-1}	1.2	7

models to river N retention, estimates of river N retention are quite uncertain, owing largely to a lack of N-retention studies at the appropriate scale. To improve these estimates it will be necessary to make measurements of denitrification throughout river networks at appropriate temporal and spatial scales to refine understanding of the magnitude of denitrification and controlling factors in river networks.

[49] The sensitivity of NEWS-DIN to parameters and inputs in individual basins can be much larger than its average sensitivity of DIN export in all basins of the world. This is especially true for model parts related to aquatic retention such as coefficient *c*, irrigation and river diversion and dam reservoirs parts. The frequency of occurrence of basins, with high sensitivity to inputs related to aquatic retention such as damming, irrigation and river water transfer, is expected to increase in the future [*Dynesius and Nilsson*, 1994; *Vörösmarty et al.*, 2003]. Therefore it is important to improve our understanding of the relationship between damming, anthropogenic river water removal and DIN export for future predictions.

Appendix A

[50] Table A1 includes data used in NEWS-DIN calibration and validation. Columns include river names, continents, basin area (km^2), measured DIN yield (kg N km^{-2} yr^{-1}), validation subset ID, and sources of concentration and discharge data.

Notation

A	basin area, km^2.
c	fitted parameter in relationship between basin area (A) and river network retention (L_{den}); in NEWS-DIN, coefficient c was set to equal 0.0605.
d	fitted parameter in relationship between basin area (A) and river network retention (L_{den}); in NEWS-DIN, coefficient d was set to equal 0.0443.
D	fraction of DIN retained in dammed reservoirs (0–1).
$DEPT_i$	depth of reservoir i, m.
DIN	DIN yield modeled per river basin, kg N km^{-2} yr^{-1}.
DIN_{meas}	DIN yield measured per river basin, kg N km^{-2} yr^{-1}.
DIN_{sew}	DIN from sewage point sources along rivers, kg N km^{-2} yr^{-1}.
e	coefficient defining the relationship between runoff and EF_{ws} if runoff is less than 0.91 (m yr^{-1}); in NEWS-DIN coefficient e was set to equal 1.1.
E_N	per capita human N emission (kg N $individual^{-1}$ yr^{-1}).
FE_{riv}	fraction of total point and nonpoint DIN inputs to the river that is exported as DIN (0–1).
FE_{ws}	fraction of N from diffuse sources in the watershed that leaches to the river as DIN (0–1).
G	fraction of N from manure, fertilizer and agricultural N_2 fixation available for leaching after harvest and grazing.
H	population density, individuals km^{-2}.
I	fraction of the population connected to sewerage systems (0–1).
L_{den}	fraction of DIN lost in the basin river network (0–1).
Q	basin discharge, km^3 yr^{-1}.
Q_{div}	amount of discharged water lost from the river by anthropogenic transfer of water out of the basin, mostly by artificial channels, km^3 yr^{-1} [*Dynesius and Nilsson*, 1994].
Q_i	discharge intercepted by dam i, km^3 yr^{-1} [*Fekete et al.*, 2000].
Q_{irr}	amount of discharge removed for irrigation, minus the amount of irrigation water that ultimately flows back into the river, i.e., extracted irrigation water that evaporates on irrigated fields, km^3 yr^{-1} [*Dynesius and Nilsson*, 1994].
Q_{nat}	amount of discharge if Q_{irr} and Q_{div} did not occur, km^3 yr^{-1} [*Dynesius and Nilsson*, 1994].
Q_{rem}	fraction of DIN retained, owing to the anthropogenic removal of (DIN containing) river water (0–1).
R	precipitation minus evaporation, m yr^{-1}.
Rt_i	water residence time of reservoir i, years.
T_N	fraction of N removed by wastewater treatment (0–1).
TN_{am}	addition of manure N, kg N km^{-2} yr^{-1}.
TN_{dep}	atmospheric deposition of NO_y-N, kg N km^{-2} yr^{-1}.
TN_{diff}	N from diffuse sources that is mobilized from the watershed soils and sediments, kg N km^{-2} yr^{-1}.
TN_{exp}	N in crops and grassland that is removed from the land by harvesting and grazing, kg N km^{-2} yr^{-1}.
TN_{fe}	addition of fertilizer N, kg N km^{-2} yr^{-1}.

Table A1. River Basin Data Used for Calibration and Validation

River	Continent	Basin Area, km^2	Measured DIN Yield, kg N km^{-2} yr^{-1}	Validation Subset ID[a]	Data Source[b]
Altamaha	North America	41,450	113.2	1	2
Amazon	South America	5,833,000	172.5	1	1
Amur	Asia	1,748,000	79.7	2	1
Anabar	Asia	98,550	11.7	1	1
Appalachicola	North America	54,660	235.1	1	2
Balsas	North America	122,600	73.1	1	1
Brazos	North America	124,600	56	2	2
Bug	Europe	68,980	28.3	1	3
Chang Jiang	Asia	1,788,000	327.5	1	1
Churchill (Hudson Bay)	North America	302,400	9.5	1	1
Colorado (Texas)	North America	120,800	24.2	2	2
Columbia	North America	729,300	74.1	2	1
Copper	North America	66,990	325.2	1	2
Dalalven	Europe	29,820	56.7	2	1
Daugava	Europe	83,160	151.3	2	3
Don	Europe	421,600	19.1	2	1
Elbe	Europe	148,000	795.4	1	1
Glama	Europe	47,310	191.8	2	1
Huang He	Asia	890,500	120.5	1	1
Hudson	North America	43,070	381.1	2	1
Indus	Asia	1,139,000	136.9	1	1
Kamchatka	Asia	50,370	88.8	2	1
Klamath	North America	32,080	71	1	2
Kolyma	Asia	663,200	18	1	1
Kuban	Europe	63,630	330.9	2	1
Kuskowin	North America	115,400	136.9	2	2
Lena	Asia	2,433,000	21.1	2	1
Mezen	Europe	75,430	24.8	1	1
Mississippi	North America	3,191,000	255.6	2	1
Murray	Australia	1,028,000	1.1	2	1
Narva	Europe	58,010	73.3	1	3
Nemanus	Europe	96,630	138.4	2	1
Neva	Europe	283,500	74.1	2	1
Nushagak	North America	35,300	105.3	1	2
Ob	Asia	3,015,000	98	1	1
Odra	Europe	119,400	389.8	1	1
Paraiba do Sul	South America	62,760	185.2	1	1
Parana	South America	2,654,000	43.9	2	1
Pechora	Europe	313,100	64.7	2	1
Pee Dee	North America	27,640	219.3	1	2
Penzhina	Asia	85,540	25.5	1	1
Potomac	North America	38,300	395.7	2	1
Rhine	Europe	164,500	2200.4	2	1
Rio Grande (United States)	North America	801,900	0.6	1	1
Rufiji	Africa	186,100	275.9	2	1
Sacramento	North America	58,690	38.1	2	1
Saint John	North America	52,850	59.8	2	1
Sakarya	Asia	56,830	155	2	1
Seine	Europe	73,190	1364.9	2	1
Stikine	North America	51,170	233	1	2
Susquehanna	North America	71,860	493.2	2	1
Tejo	Europe	73,090	122.8	1	1
Tornionjoki	Europe	34,510	9	2	1
Trinity	North America	47,380	92.2	1	2
Usumacinta	North America	67,890	562.2	2	1
Weser	Europe	45,470	1204.7	1	1
Wisla	Europe	180,000	371.8	1	1
Yana	Asia	224,200	25.9	1	1
Yenisei	Asia	2,569,000	43.1	1	1
Yukon	North America	852,700	30.6	2	1
Zhujiang	Asia	407,100	523.3	1	1

[a] Validation subsets 1 and 2 as described in section 2.2.2.

[b] References are as follows: 1, *Meybeck and Ragu* [1995]; 2, USGS [*Alexander et al.*, 1996]; 3, European Environmental Agency [*EEA*, 1998].

TN_{fix}	natural and agricultural biological N_2 fixation, kg N km^{-2} yr^{-1}.
TN_{sew}	N from sewage effluents discharged to surface water, kg N km^{-2} yr^{-1}.

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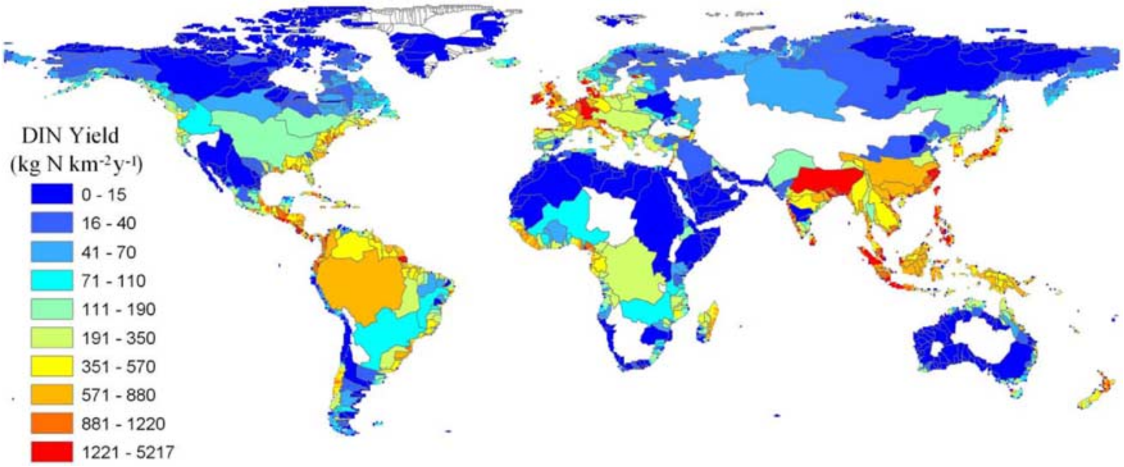

Figure 3. Modeled DIN yield by exoreic basin in kg N km^{-2} yr^{-1}.

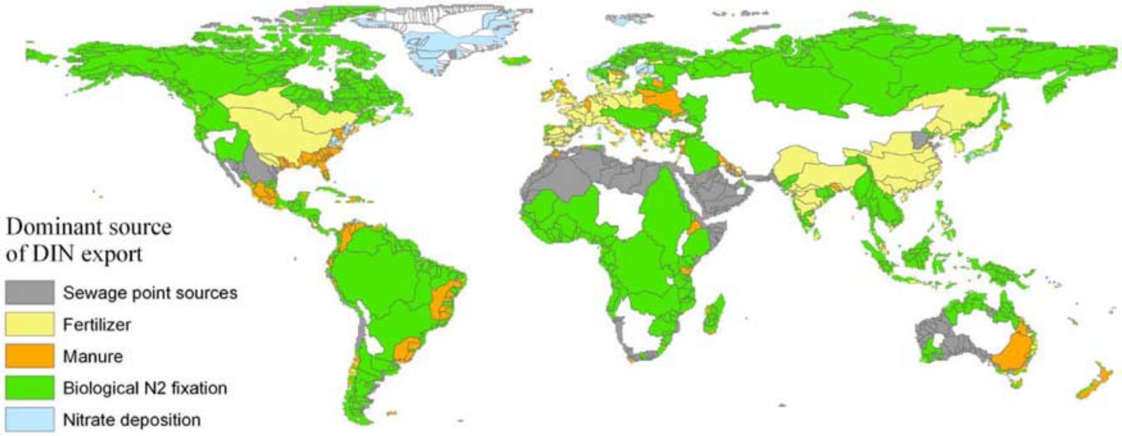

Figure 4. NEWS-DIN-predicted dominant sources of DIN export in exoreic basins.

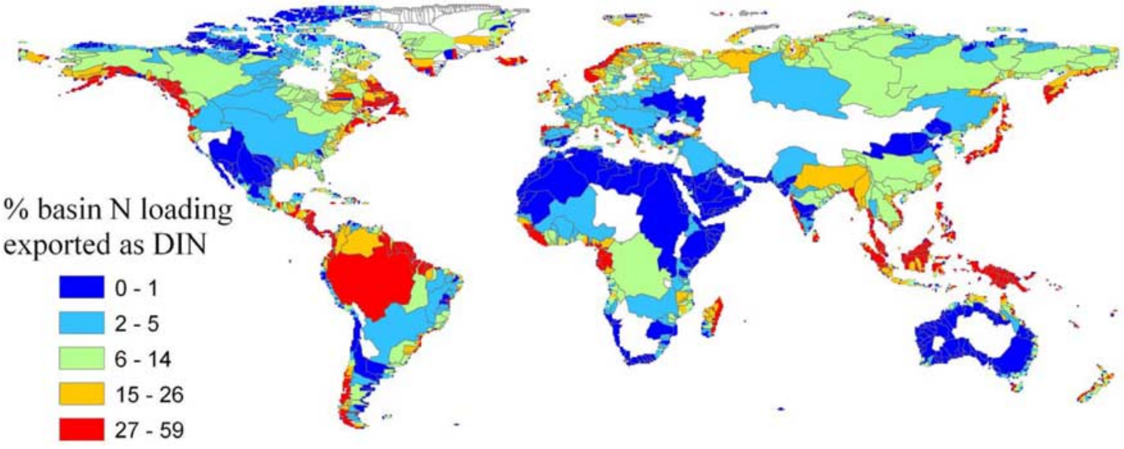

Figure 5. Percentage of N loaded onto basins from point and nonpoint sources that NEWS-DIN predicts is exported to coastal waters as DIN.