

Toward disentangling the effect of hydrologic and nitrogen source changes from 1992 to 2001 on incremental nitrogen yield in the contiguous United States

Md Jahangir Alam¹ and Jonathan L. Goodall²

The goal of this research was to quantify the relative impact of hydrologic and nitrogen source changes on incremental nitrogen yield in the contiguous United States. Using nitrogen source estimates from various federal data bases, remotely-sensed land use data from the National Land Cover Data (NLCD) program, and observed instream loadings from the United States Geological Survey (USGS) National Stream Quality Accounting Network (NASQAN) program, we calibrated and applied the spatially-referenced regression model SPARROW to estimate incremental nitrogen yield for the contiguous United States. We ran different model scenarios to separate the effects of changes in source contributions from hydrologic changes for the years 1992 and 2001, assuming that only state conditions changed and that model coefficients describing the stream water-quality response to changes in state conditions remained constant between 1992 and 2001. Model results show a decrease of 8.2% in the median incremental nitrogen yield over the period of analysis with the vast majority of this decrease due to changes in hydrologic conditions rather than decreases in nitrogen sources. For example, when we changed the 1992 version of the model to have nitrogen source data from 2001, the model results showed only a small increase in median incremental nitrogen yield (0.12%). However, when we changed the 1992 version of the model to have hydrologic conditions from 2001, model results showed a decrease of approximately 8.7% in median incremental nitrogen yield. We did, however, find notable differences in incremental yield

estimates for different sources of nitrogen after controlling for hydrologic changes, particularly for population related sources. For example, the median incremental yield for population related sources increased by 8.4% after controlling for hydrologic changes. This is in contrast to a 2.8% decrease in population related sources when hydrologic changes are included in the analysis. Likewise we found that median incremental yield from urban watersheds increased by 6.8% after controlling for hydrologic changes – in contrast to the median incremental nitrogen yield from cropland watersheds, which decreased by 2.1% over the same time period. These results suggest that, after accounting for hydrologic changes, population related sources became a more significant contributor of nitrogen yield to streams in the contiguous United States over the period of analysis. However, this study was not able to account for the influence of human management practices such as improvements in wastewater treatment plants or Best Management Practices (BMPs) that likely improved water quality, due to a lack of data for quantifying the impact of these practices for the study area.

Keywords: Water quality, anthropogenic activities, land use, non-linear regression modeling, hydrology.

1. Introduction

Anthropogenic activities are altering the nitrogen-cycle and resulting in increased contributions of excess nitrogen to aquatic ecosystems [Howarth *et al.*, 1996; Galloway *et al.*, 1995; Boyer *et al.*, 2002]. This excess nitrogen can have serious environmental impacts including algal blooms that contribute to anoxic and hypoxic conditions in waterbodies

¹Graduate Research Assistant,

41 [NRC, 2000]. The frequency and magnitude of hypoxic areas in coastal waterbodies in the
42 United States and abroad have increased during the latter half of the twentieth century
43 [Diaz, 2001; Rabalais *et al.*, 2002]. In the Gulf of Mexico in particular, increased nitro-
44 gen loading from the Mississippi River has caused eutrophication and chronic seasonal
45 hypoxia in the shallow waters of the Louisiana Shelf [Alexander *et al.*, 2000]. Many large
46 ecosystems that are now severely stressed by hypoxia face declines in fishery production,
47 reductions in species diversity, and changes to food web structures [Diaz, 2001]. Studies
48 suggest that future riverine nitrogen export is likely to increase by as much as 24% in re-
49 sponse to heavier fertilizer use, expanded corn production to meet the increased demand
50 of food and bio-fuel production, and an increase in annual river discharge under future
51 climate conditions [Han *et al.*, 2009]. These factors make it important to understand how
52 regional-scale anthropogenic activities, in addition to direct changes to nitrogen sources
53 and hydrology, will impact the delivery of nitrogen to waterbodies.

54 Nitrogen sources and delivery involve interrelationships between human, economic, and
55 physical systems. Anthropogenic activities are not only altering the spatial distribution of
56 nitrogen sources, but also the hydrologic cycle by changing land use, which in turn changes

Department of Civil and Environmental

Engineering, University of South Carolina,

Columbia, SC, USA

²Assistant Professor, Department of Civil

and Environmental Engineering, University

of South Carolina, Columbia, SC, USA

57 evapotranspiration rates, precipitation rates, runoff volumes, infiltration rates, and air
58 temperature [Feddema *et al.*, 2005; Foley *et al.*, 2005; Juang *et al.*, 2007]. Application of
59 nitrogen, increased in the form of fertilizer, is driven by food, and more recently bio-fuel,
60 production needs [Stonestrom *et al.*, 2009]. Human activities are also responsible for the
61 change of animal waste input and atmospheric deposition. Nitrogen delivery is dependent
62 on a watershed's ecological and biophysical characteristics including soil properties, stream
63 availability, average annual temperature and precipitation [Smith *et al.*, 1997; Jones *et al.*,
64 2001]. For these reasons, quantifying the impacts of anthropogenic nitrogen sources and
65 hydrology/climate related changes in the contiguous United States, the main focus of this
66 study, remains a major research challenge.

67 Past data collection efforts in the United States have resulted in spatially detailed in-
68 formation on nitrogen sources, the physical environment, and instream nitrogen concen-
69 trations. Modelers have leveraged these historical data to understand the relationships
70 that drive nitrogen fate and transport at regional spatial scales. The SPAtially Refer-
71 enced Regressions On Watershed Attributes (SPARROW) model [Smith *et al.*, 1997] is
72 an example of a regional-scale nitrogen fate and transport model that uses a combination
73 of physically-based process representations (e.g., mass balances, overland and instream
74 losses) and statistical regression to explain observed spatial variability in instream nitro-
75 gen concentrations. SPARROW has been applied to the Mississippi River Basin [Alexan-
76 der and Smith, 2005; Alexander *et al.*, 2008], the Chesapeake Bay Watershed [Roberts and
77 Prince, 2010], the contiguous United States [Smith *et al.*, 1997], and other regions in the
78 United States and abroad [Elliott *et al.*, 2005; Hoos and McMahon, 2009]. SPARROW is
79 unique as a statistical water quality model because it incorporates spatial referencing of

80 the watershed and river network when modeling the transfer of nutrients from the land-
81 scape to streams and through the river network. *Smith et al.* [1997] demonstrated that
82 spatial referencing increases model accuracy by reducing commonly encountered problems
83 of network sparseness, bias, and basin heterogeneity. Although the model has been widely
84 used for regional scale analysis, only one study has used the model to understand tempo-
85 ral changes [*Alexander et al.*, 2008], and this study was limited to the Mississippi River
86 Basin.

87 The objective of this study was to quantify the relative effects of changes in cli-
88 mate/hydrology compared to changes in nitrogen source contributions to incremental
89 nitrogen yield for the contiguous United States over a decadal time period (1992-2001).
90 We applied SPARROW to estimate changes in the delivery of nitrogen to waterbodies
91 for the years 1992 and 2001 due to changes in source contributions and hydrological fac-
92 tors. We used these two years because of the availability of National Land Cover Dataset
93 (NLCD) that includes a land use change product dataset for 1992 and 2001. Using datasets
94 described in the following section, we parameterized and then calibrated SPARROW for
95 the contiguous United States using the best available information for the year 1992. We
96 then used the calibrated model coefficients to predict incremental nitrogen yield for 1992,
97 2001, and two other hypothetical scenarios: one where we set the hydrologic conditions to
98 1992 levels and source contributions to 2001 levels, and a second where we set hydrologic
99 conditions to 2001 levels and source contributions to 1992 levels. For model parameters
100 that we assumed were constant over the period of analysis (soil permeability and drainage
101 density), we used the base data available as part of the 2.8 version of the SPARROW
102 model [*Schwarz et al.*, 2006]. For time dependent variables (e.g., observed loading, ap-

103 plication rates, and land use conditions), we determined the state conditions for the two
104 study years. Our analysis focused on the incremental total nitrogen yield estimated by
105 the model for over 60,000 river reaches in the contiguous United States. Incremental yield
106 is defined in the SPARROW model as the total flux delivered from the incremental water-
107 shed to the reach, normalized by the watershed drainage area with units of $\text{kg ha}^{-1} \text{yr}^{-1}$.
108 The model output quantifies nitrogen yield for each nitrogen source considered within the
109 model. A key assumption of our study was to neglect human management practices meant
110 to reduce nitrogen loading (e.g., reductions wastewater point source loadings and the use
111 of Best Management Practices (BMPs) to control nitrogen runoff). This assumption was
112 necessary because of a lack of data for quantifying the impact of these activities, however
113 the results of the analysis should be interpreted in light of this key model limitation.

114 In the following section we provide details of the study methodology summarized in
115 the previous paragraph. This section is followed by a presentation and discussion of
116 the study results as a means for understanding changes in the spatial distribution of
117 incremental nitrogen yield over the decadal study period. Finally we present a summary
118 and concluding statements from this work, along with suggested extensions to the study
119 materials and methodology that could be accomplished through future research.

2. Materials and Methods

2.1. Model Description

120 SPARROW is a non-linear regression model that relates measured loadings at moni-
121 toring stations to point and nonpoint source loadings, waterbody properties, and water-
122 shed attributes in order to predict long-term mean annual instream load for unmonitored
123 reaches [Schwarz *et al.*, 2006]. The measured instream loadings serve as the dependent

124 variable in the regression, while point and nonpoint sources, waterbody properties, and
 125 watershed attributes serve as the independent variables. Nitrogen source terms consid-
 126 ered by the model include direct sources such as population related sources, atmospheric
 127 deposition, fertilizer application and animal waste, as well as indirect sources such as
 128 non-agricultural land use. Watershed attributes typically considered in SPARROW in-
 129 clude precipitation, temperature, soil permeability and stream density, while waterbody
 130 attributes include flow rate, velocity and hydraulic loading (for lakes and reservoirs).

131 The SPARROW model formulation used in this study states that the mean annual total
 132 nitrogen loading observed at reach i is

$$L_i = \left[\left\{ \sum_{j \in J(i)} L_j \right\} A(Z_i^s, Z_i^r; \kappa_s, \kappa_r) + \left\{ \sum_{n=1}^N S_{n,i} \beta_n \right\} D_n(Z_i^D; \alpha) A'(Z_i^s, Z_i^r; \kappa_s, \kappa_r) \right] \epsilon_i \quad (1)$$

133 where the first summation term is the amount of flux that leaves each of the adjacent up-
 134 stream reaches for reach i where L_j represents the measured or estimated flux leaving the
 135 adjacent upstream reach j . The function $A(\cdot)$ represents the loss of mass due to transport
 136 to the downstream node of reach i (Figure 1). The vectors Z_i^s and Z_i^r are measured stream
 137 and reservoir characteristics, while κ_s and κ_r are corresponding coefficient vectors. If the
 138 waterbody i is a stream, then Z_i^s and κ_s define the function $A(\cdot)$ and if the waterbody i
 139 is a lake or reservoir, then Z_i^r and κ_r define the function $A(\cdot)$.

140 The second summation term is the amount of mass originating within the watershed
 141 that is delivered to the waterbody i . N is the total number of mass sources, n is an
 142 individual mass source, and $S_{n,i}$ is the contribution from mass source n in reach i . β_n is a
 143 regression coefficient that represents an array of source-specific coefficients and serves as

144 a conversion factor between the source units and flux units estimated by the model. The
 145 function $D(\cdot)$ represents the land-to-water delivery process where α is the estimated vector
 146 of coefficients and Z_i^D is the vector of watershed attributes. The term $A'(\cdot)$ represents
 147 the instream mass loss for reach i . Finally, ϵ_i is the multiplicative error term defined
 148 by the model, which is independent and identically distributed across watersheds in the
 149 intervening drainage area between monitoring stations [*Alexander and Smith, 2005; Hoos*
 150 *and McMahon, 2009*].

151 Land-to-water delivery is represented in our model by Equation 2 that represents a
 152 first-order decay process

$$D_n(Z_i^D; \alpha) = \exp(-\alpha' Z_i^D) \quad (2)$$

153 where α' is an array of the model coefficients that describe the land-to-water delivery
 154 for each element in the watershed attribute array, Z_i^D . Instream transport is represented
 155 within the model by Equation 3 that models loss as a first-order decay process

$$A(Z_i^s, Z_i^r; \kappa_s, \kappa_r) = \exp(-\kappa_s' T_{i,j}) \quad (3)$$

156 where κ_s' is the array of decay coefficients for streams classified by their mean annual flow
 157 rates. The decay coefficients are estimations of mass loss per unit stream length. $T_{i,j}$ is
 158 an array of waterbody characteristics for the flow path. The first-order decay is modeling
 159 losses due to physical processes occurring within the stream such as denitrification and
 160 sedimentation [*Alexander and Smith, 2005*]. Finally, for reaches that represent lakes or
 161 reservoirs, $A(Z_i^s, Z_i^r; \kappa_s, \kappa_r)$ takes the form of Equation 4 that models nitrogen loss as a
 162 settling rate

$$A(Z_i^s, Z_i^r; \kappa_s, \kappa_r) = \left(1 + \frac{\kappa_r}{q_i^r}\right)^{-1} \quad (4)$$

163 where κ_r is the reservoir decay coefficient estimated by the model and q_i^r is the areal
164 hydraulic loading (ratio of reservoir outflow to surface area, in units of distance per time).

165 The model is first calibrated using Equations 1-4 to estimate nitrogen loading for all
166 monitored reaches. The result of the regression is estimates of the coefficient values for
167 β , α , and κ that minimize errors between observed and predicted loadings for monitored
168 reaches. Once these coefficients have been determined, the model is applied to predict
169 nitrogen delivery for unmonitored reaches within the river network. The model results
170 in predictions of incremental and total nitrogen yield for each reach in the river network
171 dataset and for each nitrogen source considered by the model. While there are limitations
172 to the modeling approach used by SPARROW, as we will discuss in greater detail in
173 Section 3.3, we have used the model because it offers a practical blend of process-based
174 and empirical modeling appropriate for regional-scale assessments, where it is difficult to
175 parameterize physical-based models of hydrologic and biogeochemical processes.

176 In this study we considered four model scenarios: Model I (1992), Model II (2001
177 Hydrology), Model III (2001 Sources) and Model IV (2001). Model I represents the
178 1992 conditions. All the sources, land-to-water delivery terms, and mean annual loadings
179 represent the year 1992. Model II represents Model I modified so that the precipitation
180 and mean air temperature are set to 2001 conditions. We use this model scenario to
181 estimate how the hydrologic changes (precipitation and evaporation, which is related
182 to air temperature) impacted nitrogen delivery. Model III represents Model I modified
183 so that the source variables are set to 2001 conditions. This model scenario allows us to
184 estimate loading changes due to changes in nitrogen sources. Finally, Model IV represents
185 the 2001 scenario where all the sources and land-to-water delivery terms represent the year

2001. Our initial approach was to calibrate the model to 1992 and 2001 observed loading
 data separately, but there were insufficient instream concentration observations available
 for 2001 to support this approach. Therefore we calibrated the SPARROW model for
 1992 conditions and then simulated stream loads for 2001 conditions assuming that the
 SPARROW model coefficients were unchanged and only state conditions changed. We
 then used the SPARROW model coefficients from the Model I calibration to also predict
 loadings for the Model II and Model III scenarios. Our justification for this assumption
 is that because there were no major departures in state conditions between the decade
 separating the two study periods, we can assume that the statistical relationship that
 forms the basis of the model formulation is valid for both years. To verify our assumptions,
 we evaluated the 2001 model by comparing predicted loadings to the limited set of available
 observed loadings. A similar approach of calibrating the model for 1992 and then using
 the calibration coefficients to predict for 2002 was used by *Alexander et al.* [2008] for the
 Mississippi River Basin.

Once the model has been calibrated and predicted, the incremental yield for each catch-
 ment was estimated as

$$Yield_i = \left\{ \sum_{n=1}^N S_{n,i} \beta_n \right\} \times D_n(Z_i^D; \alpha) A'(Z_i^s, Z_i^r; \kappa_s, \kappa_r) / Area_i \quad (5)$$

where i is the number of incremental catchments, $Area_i$ is the area of the catchment and
 $Yield_i$ is the incremental yield in $\text{kg ha}^{-1} \text{ yr}^{-1}$. Incremental yield gives the yield estimates
 that originated in that specific catchment without considering the upstream contributions.
 The incremental yield model output was the primarily focus of this study because we were

206 interested in how loads were delivered to streams under different hydrologic and sourcing
207 conditions.

2.2. Data Preparation

208 It is typical to use the Enhanced River Reach File 1 (ERF1) dataset as the river network
209 representation for continental-scale SPARROW applications [Nolan *et al.*, 2002]. ERF1
210 is a digital stream network at a 1:500,000 spatial scale for the contiguous United States
211 that is an improvement over the earlier River Reach File 1 (RF1) dataset [DeWald *et al.*,
212 1985; Alexander *et al.*, 1999]. The stream network consists of more than 60,000 reaches
213 with mean reach length of 17 km and total reach length of 1 million km. Approximately
214 2,000 of the river reach features represent large reservoirs with a capacity greater than 6
215 million m³ [Smith *et al.*, 1997]. Mean streamflow, stream velocity, and time of travel are
216 included as reach attributes in the dataset and used to model instream loss rates due to
217 sedimentation and denitrification in the SPARROW model [DeWald *et al.*, 1985]. The
218 dataset also includes attributes of stream morphology and hydraulic properties, for exam-
219 ple incremental and total drainage area, drainage density, mean water depth, and areal
220 hydraulic load. Mean stream velocity was estimated using a regression based approach
221 that relates stream velocity to long term mean streamflow and stream order. Travel time
222 was then estimated by dividing the reach channel length by the mean stream velocity
223 [DeWald *et al.*, 1985].

224 Watersheds for each reach within the ERF1 dataset were derived using terrain pro-
225 cessing algorithms and the Hydro 1K Digital Elevation Model [U.S. Geological Survey,
226 2000]. Watershed attributes considered in previous applications of SPARROW include
227 precipitation, long term average streamflow, incremental drainage area, soil permeability,

228 slope, drainage density, reach length and mean air temperature [*Smith et al.*, 1997]. How-
229 ever, this previous work found that only soil permeability, drainage density and mean air
230 temperature were statistically significant for explaining the nitrogen fate and transport at
231 the regional spatial scale [*Smith et al.*, 1997]. Later work has shown precipitation to also
232 be a statistically significant watershed attribute in nitrogen modeling using SPARROW
233 [*Hoos and McMahon*, 2009]. Therefore, we selected soil permeability, drainage density,
234 mean air temperature, and precipitation as watershed attributes in our model formula-
235 tion. Soil permeability was estimated for each watershed using the State Soil Geographic
236 (STATSGO) database [*Schwarz and Alexander*, 1995]. Drainage density is defined as the
237 the ratio of stream length to the drainage area and was calculated from the ERF1 reach
238 length and the area of the Hydro 1K-derived watershed associated with that reach.

239 We used PRISM (Parameter-elevation Regressions on Independent Slopes Model) data
240 [*PRISM*, 2004] for temperature and precipitation estimates. After creating the maximum
241 and minimum temperature grid from the data available in PRISM, we averaged the max-
242 imum and minimum grids to estimate the average temperature for the years 1992 and
243 2001. After creating the average temperature grid, we estimated temperatures for each
244 ERF1 reach catchment (Figure 4). We created the precipitation grid for the year 1992 as
245 the average from the PRISM data for the years 1991, 1992, and 1993. We followed the
246 same procedure to estimate the average precipitation for the year 2001 by using the years
247 2000, 2001, and 2002. We took this approach to dampen the variability that can occur
248 in annual data. PRISM precipitation data indicated that 2001 was dryer than 1992 for
249 most of the United States, especially for the West, Midwest and Southwest (Figure 4).

250 Total nitrogen loading, the dependent variable in the model, was quantified using data
 251 from the United States Geological Survey (USGS) National Stream Quality Accounting
 252 Network (NASQAN). NASQAN was established by the USGS in 1974 to provide a long-
 253 term, systematically collected baseline water chemistry dataset for the nation [*Ficke and*
 254 *Hawkinson, 1975; Alexander et al., 1996*]. We define Total Nitrogen (TN) in this study
 255 as nitrate, nitrite, and total Kjeldahl nitrogen in unfiltered samples. Annual loading was
 256 estimated by using the Fluxmaster program [*Schwarz et al., 2006*]. Fluxmaster predicts
 257 the continuous daily load from continuous daily observed streamflow and discontinuous
 258 nitrogen concentration data by using a nonlinear regression model, and then calculates
 259 annual loading by taking averages of the daily estimates of total nitrogen loading. In the
 260 estimation process we used stations that have at least 15 observations to reduce estimation
 261 uncertainty.

262 We selected the following Fluxmaster model to relate measured nitrogen concentration
 263 to streamflow and other explanatory variables.

$$\begin{aligned} \ln(l) = & \lambda_0 + \lambda_1 t + \lambda_2 \sin(2\pi t) \\ & + \lambda_3 \cos(2\pi t) + \lambda_4 \ln(q) + \lambda_5 [\ln(q)]^2 + \alpha \end{aligned} \quad (6)$$

264 where l is the instantaneous nitrogen transport, t is decimal time to account for temporal
 265 trends [*Robertson et al., 2006*], q is instantaneous discharge and λ_0 through λ_5 are regres-
 266 sion coefficients. The term α is the sampling and model error assumed to be independent
 267 and identically distributed, while the trigonometric terms approximate seasonal variations
 268 in transport. The mean annual loading was calculated as

$$L = \frac{1}{365} \sum_{i=1}^{365} \exp[\lambda_0 + \lambda_1 t_i + \lambda_2 \sin(2\pi t_i)]$$

$$+\lambda_3\cos(2\pi t_i) + \lambda_4\ln(q_i) + \lambda_5[\ln(q_i)]^2]V_f \quad (7)$$

269 where t_i is the i^{th} day of the base year in decimal format, q_i is the average streamflow
 270 of the i^{th} day of the year over the multi-year period of the streamflow data, and V_f is
 271 the minimum variance bias re-transformation correction factor. The minimum variance
 272 unbiased estimator procedure [Cohn *et al.*, 1989] was used in the model. The time period
 273 used for estimating the average annual loads was 1970 to 2006, as described in the following
 274 paragraph, although for many stations the availability of data was only 1970 to 1995 due
 275 to budget reductions and the resulting discontinuation of monitoring for certain stations
 276 in 1995.

277 We obtained total nitrogen concentration, instantaneous streamflow, and daily average
 278 streamflow observations for NASQAN sites for the time period 1970-2008. We auto-
 279 mated the data retrieval process by using web services provided through the Consortium
 280 of Universities for the Advancement of Hydrologic Science, Inc. (CUAHSI) Hydrologic
 281 Information System (HIS) [Maidment, 2008; Goodall *et al.*, 2008]. We used nitrogen con-
 282 centration and daily streamflow data from 1970 to 2000 and detrended the water quality
 283 and flow model in Fluxmaster to 1992 to estimate long term mean loading. Using this
 284 time period, we were able to estimate loading for 354 monitoring stations for our 1992
 285 analysis. We also estimated loading for 2001 with a flow-concentration relationship more
 286 closely targeted to the study year using nitrogen concentration and daily streamflow data
 287 from 1996 to 2006 and then detrended to 2001. In this way we were able to estimate
 288 loading for 122 stations. Again, this 2001 loading dataset was not used for calibration of
 289 the SPARROW model because there were too few stations, but instead for comparison

290 of the estimated loading for 2001 from monitoring stations with the predicted loading for
291 2001 from the SPARROW Model IV simulation.

292 Nitrogen sources considered in the model are population related sources, atmospheric de-
293 position, fertilizer application, livestock waste, and non-agricultural land. Using publicly
294 available datasets, we quantified each source for the years 1992 and 2001. We used human
295 population estimates for the population related sources, similar to previous SPARROW
296 studies [*Alexander et al.*, 2008]. The assumption is that contribution from population
297 related sources like wastewater effluent, municipal waste, and urban runoff are related to
298 human population. We used county level population estimates from the United States
299 Census Bureau [*U.S. Bureau of Census*, 2010] for 1992 and 2001 and ArcGIS® to calcu-
300 late the population density for the contiguous United States at 1km resolution. We then
301 estimated the population count for each ERF1 watershed for the years 1992 and 2001.

302 Atmospheric deposition is a well known and important source of nitrogen, specifically
303 nitrate, to streams [*Jones et al.*, 2001]. We considered wet deposition of inorganic nitro-
304 gen (nitrate and ammonia) (kg yr^{-1}) as a measure of atmospheric deposition following
305 the approach used in previous SPARROW model applications [*Smith et al.*, 1997]. We
306 did not include ammonia deposition to avoid double counting of agricultural nitrogen
307 input [*Howarth et al.*, 1996]. Annual estimates from the National Atmospheric Deposi-
308 tion Program [*NADP*, 2010] were used to estimate wet deposition of inorganic nitrogen.
309 These monitoring station estimates were converted to a continuous grid of 1km resolution
310 using an inverse-distance weighting interpolation method. Similar to precipitation data,
311 we averaged 1991, 1992, and 1993 to create the 1992 deposition input dataset. We then
312 summarized the atmospheric deposition loadings for each watershed in the study region.

313 We followed the same procedure for the 2001 nitrogen deposition, averaging deposition
314 estimates for 2000, 2001, and 2002.

315 Fertilizer application to both agricultural lands and to urban lands for lawn maintenance
316 is another major source of instream nitrogen. The Association of American Plant Food
317 Control Officials (AAPFCO) [*Ruddy et al.*, 2006] used state total sales rather than county
318 level sales to estimate fertilizer application because county level fertilizer sales data are
319 not reliable. Sometimes multi-county distributors report their sales for a single county
320 rather than the actual counties. Moreover farmers can buy fertilizer from one county and
321 use it in another county, so the spatial distribution of fertilizer sales may be inaccurately
322 represented. Data on the state-level annual sales of commercially produced fertilizer from
323 American Plant Food Control Officials (AAPFCO) and data on the county-level fertilizer
324 expenditure that were obtained from the Census of Agriculture were used to estimate
325 county level nitrogen input from farm fertilizer use in kilograms of nitrogen [*Ruddy et al.*,
326 2006]. State level non-farm fertilizer sales were estimated from the population data and
327 converted to nitrogen inputs in kilograms of nitrogen [*Ruddy et al.*, 2006]. This county
328 level nitrogen input was then summarized to SPARROW watershed level estimates at 1
329 km resolution for both 1992 and 2001.

330 Finally, livestock waste was considered as another source of instream nitrogen loading in
331 our model. Nitrogen input from livestock waste was estimated from county-level livestock
332 population data collected by the Census of Agriculture. *Ruddy et al.* [2006] presented
333 a county level estimate of nitrogen in the livestock waste from confined and unconfined
334 animals. Both recoverable manure from confined animals and unrecoverable manure from
335 confined and unconfined animals were included in this estimation. We estimated SPAR-

336 ROW watershed level nitrogen input from the county level dataset for 1992 and 2001 at
337 1km resolution.

338 We used the National Land Cover Dataset (NLCD) 1992/2001 Retrofit Land Cover
339 Change Product (Fry et al., 2009) to assess land use in the years 1992 and 2001. It
340 is not possible to directly use the NLCD 1992 and 2001 land cover products because
341 the products were generated using different classification methodologies, source image
342 seasonality, georegistration approaches, mapping methodologies, and land use classes [*U.S.*
343 *Geological Survey*, 2001]. We instead reconstructed two land use datasets from the recently
344 produced NLCD Land Cover Change Product that include major land use types of urban,
345 forest, crop land, grass land and non-agricultural land. Urban land includes areas of low,
346 medium, and high intensity development with a mixture of constructed materials and
347 vegetation [*U.S. Geological Survey*, 2001]. Forest land includes deciduous forest, evergreen
348 forest, and mixed forest; Cropland includes cultivated crops and pasture lands; Pasture
349 lands include both grasses and legumes for livestock grazing; Grassland includes both
350 grassland and shrubs; Non-agricultural land represents the combination of urban, forest,
351 and grasslands. Land use grids for the two study years were used to estimate the total
352 area of each land use type, for each year, and for each of the watersheds.

353 We used the data described in the previous paragraphs to construct the inputs for the
354 four previously described model scenarios. For Model I, we both calibrated the model
355 and used the calibrated model to predict 1992 loadings. For the remaining three model
356 scenarios, we used the parameters from the Model I calibration to predict loadings. We
357 used the 2.8 version of SPARROW, the latest version of the model at the time of this
358 study.

3. Results and Discussion

3.1. Model Calibration

359 Model I (1992) resulted in R^2 values for predicted log of flux of 0.885 and R^2 values for
360 predicted log of yield of 0.802 (Table 1, Figure 2). These R^2 values are similar to those
361 obtained in previous SPARROW model applications. For example, *Smith et al.* [1997]
362 reported results from a national SPARROW model for total nitrogen using 414 NASQAN
363 stations in which their R^2 values for log of flux were 0.874 and *Hoos and McMahon* [2009]
364 reported R^2 values for flux and yield were 0.96 and 0.68, respectively for an application of
365 SPARROW for the Southeastern United States. The model residuals (Figure 2) showed
366 some signs of a spatial bias with the highest and lowest loads corresponding to the largest
367 and smallest rivers, suggesting an over-prediction for larger rivers and an under-prediction
368 for small rivers. Some possible bias was also present in specific regions such as the Pacific
369 Northwest, which showed over-predictions in general, and the Midwest, which showed
370 under-predictions in general. SPARROW predicted loading for Model IV (2001) was also
371 compared to actual loading observations estimated using the Fluxmaster program based
372 on the flow and concentration data available from 1996 to 2006 for 122 stations (Figure
373 3). The R^2 value for this predicted log of flux vs actual log of flux was 0.890 for these
374 stations. It should be noted that the high R^2 value for the 2001 evaluation may be due in
375 part to the 2001 dataset consisting of primarily larger streams when compared to the 1992
376 dataset, or to a time lag introduced by using long term flow and concentration data for
377 model estimation. Nonetheless, we argue that the evaluation of the model against 2001
378 observed loads provides a reasonable level of confidence that the model is able to predict
379 loads in this year based on calibrated model coefficients for the 1992 model.

380 The SPARROW model includes three coefficients, α , β and κ , that are fit during the
381 model calibration process (Table 2). The coefficients have physical meaning in that they
382 allow one to understand losses due to land-to-river and instream transport processes.
383 The α coefficient results for soil permeability, drainage density, mean annual air tem-
384 perature, and precipitation were consistent with expectations and previous SPARROW
385 results [Smith *et al.*, 1997; Alexander *et al.*, 2008; Hoos and McMahon, 2009]. All α coeffi-
386 cients were statistically significant ($p < 0.05$). The magnitude of the α coefficients for the
387 different watershed attributes provides important information on how soil permeability,
388 drainage density, mean annual air temperature and precipitation influence the efficiency
389 with which nitrogen is delivered from the land to waterbodies. As expected, the coeffi-
390 cients for soil permeability and temperature were negative. Highly permeable soils will
391 have higher absorption capacities for nutrients, which act to resist nitrogen transport to
392 streams [Smith *et al.*, 1997]. Temperature has a negative correlation with nitrogen trans-
393 port because increased temperature results in an increased denitrification rate, therefore
394 decreasing the proportion of nitrogen transported to waterbodies [Smith *et al.*, 1997].
395 Drainage density, the ratio of stream length to drainage area for a watershed, is positively
396 correlated with nitrogen delivery because having a higher drainage density increases the
397 ability for nitrogen to be delivered from the landscape to waterbodies. Similarly, precip-
398 itation is positively correlated with nitrogen transport as higher precipitation indicates
399 higher runoff.

400 For direct nitrogen sources, the β coefficients account for possible variation in source es-
401 timates and will vary between sources [Smith *et al.*, 1997]. For indirect nitrogen sources,
402 the β coefficients represent a source term and provide information about the quantity

403 of nitrogen originating from different sources. The β coefficient for population related
404 sources was $3.57 \text{ kg person}^{-1} \text{ yr}^{-1}$ and was statistically significant ($p < 0.05$). The β coef-
405 ficients for atmospheric deposition, fertilizer application, and livestock waste production
406 were 0.28, 0.25, and 0.08, respectively, and the β coefficient for non-agricultural land was
407 $310 \text{ kg km}^{-2} \text{ yr}^{-1}$. The β coefficients for fertilizer application and non-agricultural land
408 were statistically significant ($p < 0.05$), but the β coefficients for atmospheric deposition
409 and livestock waste production were not statistically significant ($p > 0.05$). Land use
410 types urban, crop, forest, and grass land were used as variables in the preliminary model
411 simulations but later dropped based on the following criteria. After considering statistical
412 significance, if a variable was not statistically significant, we used variance inflation factor
413 (VIF) to determine if the multicollinearity was responsible for this. We also used eigen-
414 spread values to identify multicollinearity. If the eigenspread value was greater than 100,
415 we dropped that predictor variable from the model [Schwarz *et al.*, 2006]. Our final selec-
416 tion of the predictor variables was based on constant variance and minimum correlation.
417 Cropland was dropped because of the strong collinearity between cropland and fertilizer
418 application. For the same reason only the population related source was included and the
419 urban land source was dropped from the final model.

420 The model calibration resulted in instream loss coefficients (κ) that were greater for low-
421 flow streams ($Q < 28.3 \text{ m}^3 \text{ s}^{-1}$) compared to medium-flow streams ($28.3 \text{ m}^3 \text{ s}^{-1} \leq Q \leq 283$
422 $\text{m}^3 \text{ s}^{-1}$). These coefficients were found to be statistically significant for low flow streams
423 ($p < 0.05$), but not for medium streams ($p > 0.05$). This is consistent with previous
424 SPARROW model results and the conclusion that loss rate decreases with increasing
425 stream size [Alexander *et al.*, 2000; Hoos and McMahon, 2009]. Model prediction for

426 the reservoir loss coefficient (κ_r) was 7.18 m yr^{-1} in 1992, was statistically significant (p
427 < 0.05), and was consistent with *Hoos and McMahon* [2009], which reported a slightly
428 higher reservoir loss rate of 13.1 m yr^{-1} for the Southeastern United States. The standard
429 errors for the coefficients (Table 2) provide a measure of the confidence intervals of the
430 coefficient estimates. The standard error values are in line with prior SPARROW studies
431 and could be used through future work to quantify uncertainty of the model estimates
432 reported in this study.

3.2. Model Predictions

433 When comparing overall change in loading between Model I (1992) and Model IV (2001),
434 the results indicate a decrease in the median incremental nitrogen yield of 0.67 kg ha^{-1}
435 yr^{-1} (or 8.21%) from $8.16 \text{ kg ha}^{-1} \text{ yr}^{-1}$ in 1992 to $7.49 \text{ kg ha}^{-1} \text{ yr}^{-1}$ in 2001 (Table 3).
436 Table 3 presents various statistical quantities for the predictions including the standard
437 deviation for predicted incremental total nitrogen yield ($\text{kg ha}^{-1} \text{ yr}^{-1}$) that provide a
438 measure of the distribution of yield results across the entire study area. We have chosen
439 to summarize the incremental nitrogen yield prediction results using the median value
440 because it is less influenced by very high incremental yields predicted for larger river
441 basin systems. That said, we acknowledge that because first-order streams dominate
442 the stream network, the median will be weighted toward changes in smaller streams.
443 *Alexander et al.* [2008] also indicated a slight overall decrease in both simulated loadings
444 based on a SPARROW model, as well as monitoring-based loadings of nitrogen in streams
445 over a similar time period for the Mississippi River Basin. Comparing Model I vs Model
446 II (2001 Hydrology) and then Model I vs Model III (2001 Sources) reveals that this
447 decrease was mainly due to hydrologic differences rather than variability in nitrogen source

448 input. The estimates of median incremental nitrogen yield between Model II and Model
449 I indicate a decrease of $0.71 \text{ kg ha}^{-1} \text{ yr}^{-1}$ (or 8.70%). This decrease is the result of
450 changes in precipitation and mean annual air temperature between 1992 to 2001. The
451 precipitation map [*PRISM*, 2004] indicated a decrease in precipitation in 2001 compared
452 to 1992 (Figure 4), and precipitation is an important nitrogen delivery variable for land-
453 to-water transport. Comparing estimates between Model III and Model I indicates an
454 increase of $0.01 \text{ kg ha}^{-1} \text{ yr}^{-1}$ (or 0.12%) in incremental nitrogen yield (Table 3). This
455 result suggests that only taking into account changes in sources does not account for the
456 changes in incremental nitrogen yield over the period of study. Previous studies in the
457 Mississippi River Basin also showed an increase in nitrogen sources and loading to streams
458 before the early 1980s, but after that time no significant trend was observed [*Goolsby et al.*,
459 1999; *National Agricultural Statistics Service (NASS)*, 1998; *Alexander and Smith*, 1990;
460 *Council of Environmental Quality (CEQ)*, 1989].

461 The total nitrogen yield map (Figure 5a) shows the overall incremental yield scenario
462 for 1992 (Model I). This figure shows that the incremental yield estimates are higher in the
463 Upper Mississippi, the Ohio, the southeastern portion of the Missouri, the Tennessee and
464 the Lower Mississippi Basins, but also in the Northeastern and the Pacific West Coast
465 regions. A common characteristic of these regions is that they have either significant
466 agricultural lands or higher human populations, which account for the higher incremental
467 yields. *Alexander et al.* [2008] showed a similar pattern of nitrogen yield for the Mississippi
468 River Basin. The results shown in Figure 5a also suggest that the incremental nitrogen
469 yield estimates are highest in the wettest areas of the country and lowest in the driest
470 areas. Nitrogen can be more easily transported in wetter climates because of higher

471 precipitation, runoff, and discharge. On the other hand, in arid and semiarid climates,
472 low amounts of precipitation and high evaporation rates result in limited runoff and, as a
473 result, reduced nitrogen yields. This may be the reason for the lower nitrogen yield from
474 large populated cities in the Southwestern United States.

475 Figure 5b shows differences in the incremental yield scenario if hydrologic inputs (mean
476 annual air temperature and precipitation) are set to 2001 conditions but nitrogen source
477 data is held at 1992 conditions (Model I vs Model II). Analyzing the input data shows
478 that the change in precipitation map has a similar pattern as seen in 5b, indicating
479 the importance of precipitation in particular in nitrogen delivery due to nonpoint source
480 pollution transport. Figure 5c shows the results of a scenario where we used 2001 source
481 contribution but 1992 hydrologic conditions to quantify how change in sources only would
482 affect incremental nitrogen yield (Model I vs Model III). Comparing Figure 5c with Figure
483 5a suggests that the region in the Mississippi Basin with the highest nitrogen yield in 1992
484 actually saw a decrease in yield when considering only changes in nitrogen sources and
485 controlling for hydrologic/climate changes. Finally Figure 5d presents the overall change
486 in incremental yield during the period 1992 to 2001 (Model I and Model IV). This map
487 shows that some regions of the contiguous United States produced more incremental
488 nitrogen yield, despite the fact that the median yield decreased during the period of
489 analysis. Furthermore, by comparing Figures 5b and 5c to Figure 5d, we can observe
490 changes in incremental nitrogen yield over the study period and determine whether they
491 were due to variability in nitrogen sources or changes in hydrologic/climate conditions.
492 For example, the increase in the Ohio and Tennessee Basins appears to be primarily due

493 to hydrologic changes, while the increase in the northern portion of the Missouri Basin
494 appears to be primarily due to increases in nitrogen sources.

495 Estimates of median incremental yield for each model scenario, separated by each ni-
496 trogen source, are presented in Figure 6. Comparing Model I to Model IV provides
497 an estimate of median incremental yield difference between 1992 and 2001 and indi-
498 cates a decrease of $0.01 \text{ kg ha}^{-1} \text{ yr}^{-1}$ (2.8%) for population related sources, 0.14 kg ha^{-1}
499 yr^{-1} (15%) for atmospheric deposition, $0.08 \text{ kg ha}^{-1} \text{ yr}^{-1}$ (5.4%) for fertilizer application,
500 $0.03 \text{ kg ha}^{-1} \text{ yr}^{-1}$ (6.1%) for livestock waste production, and $0.22 \text{ kg ha}^{-1} \text{ yr}^{-1}$ (8.0%) for
501 non-agricultural lands. Comparison of Model I vs Model III results, which controls for
502 hydrologic changes, provides insight into how changes in nitrogen sources alone during
503 the study period impacted nitrogen yield to streams. The results show that population
504 related sources had the largest percent increase in nitrogen yield (8.4%), fertilizer applica-
505 tion had the second largest percent increase (2.9%), and livestock waste had the smallest
506 (0.7%) increase. Median incremental nitrogen yield from atmospheric deposition and non-
507 agricultural land decreased by 6.6% and 0.15%, respectively. The decrease in nitrogen
508 yield from atmospheric deposition was likely due to the fact that we used wet deposition
509 as an input dataset and, by doing so, this source already accounts for hydrologic changes.
510 Controlling for variability in nitrogen sources (Model I vs Model II) showed population
511 related sources had the largest percent decrease (10%) in nitrogen yield. Thus, while yield
512 from population related sources had an overall decrease of 2.8% when comparing Model
513 I and Model IV, considering also the results from Models II and III suggests that this
514 decrease was primarily due to 2001 being a drier year than 1992. Finally, we noted that
515 the percent yield decrease when controlling for sources (Model I vs Model II) was similar

516 for all sources (ranging between a 7% and 10% decrease for the different nitrogen sources
517 considered). When interpreting these results, it should be noted that even though human
518 population is used as a surrogate for point source and non point urban sources, population
519 is diffuse in nature and the relationship between changes in point sources and changes in
520 population is not ideal but necessary given available information. It should also be noted
521 that fertilizer application and livestock waste were assigned to reaches based on county
522 level data, and this data might exaggerate these sources for many counties outside of the
523 agricultural Midwest. Also, as mentioned previously, management practices intended to
524 reduce nitrogen in waters such as improvements in wastewater treatment, construction of
525 urban retention ponds as Best Management Practices (BMPs), preserving riparian buffer
526 zones, and reductions in automobile emission (if not reflected in NADP measurements) are
527 not considered in this study because we are unaware of accurate national-scale datasets
528 for quantifying the impacts of these activities.

529 The spatial distribution of changes in yield for each source when comparing Model
530 I vs Model IV suggests that nitrogen yields from population related sources increased
531 in the western United States, Texas, and Florida (Figure 7). However, it also suggests
532 an increase in the Tennessee and Ohio Basins. This is the same region that showed
533 higher precipitation rates between 2001 and 1992, which may explain the higher yield
534 from population sources as precipitation may have caused an increase in nonpoint source
535 pollution from populated areas in this region. Nitrogen yields from atmospheric deposition
536 show a pattern nearly identical to the input dataset of changes in nitrogen deposition
537 over the study period. These data show widespread decreases in wet nitrate deposition
538 in the Midwest and Northeast over the period of analysis [NADP, 2010]. Fertilizer yield

539 increased in the West, upper portion of the Missouri Basin, Ohio Basin, and the Northeast,
540 while it decreased in the Southeast and Upper Mississippi Basin. *Alexander et al.* [2008]
541 found that the fertilizer sources decreased in the Upper Mississippi Basin and increased
542 in the Missouri Basin due to expanded corn and soybean production, which agree with
543 our findings. However, *Alexander et al.* [2008] found a decrease in fertilizer application
544 for the Ohio Basin, but we found an increase for a portion of this Basin in our study.
545 The increases due to fertilizer in the Ohio Basin and the upper portion of the Missouri
546 Basin follow the pattern shown in the overall incremental yield change map for 1992 to
547 2001 (Figure 5d), suggesting that fertilizer increases were the primary source of overall
548 nitrogen yield increase in these regions. Increases due to livestock waste showed a scattered
549 pattern overall with decreases in the lower portion of the Mississippi Basin. Changes due
550 to the non-agricultural land followed the same pattern as precipitation changes because
551 non-agriculture land is well distributed across the study region.

552 Figure 8 presents source share results for each nitrogen source considered in the model.
553 Source share is defined as the incremental yield for a specific source of nitrogen divided
554 by the total incremental yield for a given watershed. Given the definition of incremental
555 yield (Equation 5), it can be shown that the watershed property term (Z^D) and the in-
556 stream transport terms (Z^s and Z^r) cancel out when calculating source share. In other
557 words, because the transport factors are applied equally to all sources, the changes in cli-
558 mate/hydrology transport factors do not impact the incremental measures of the source
559 shares, which are applied equally for all sources. Therefore, Model I source share results
560 match Model II source share results and, similarly, Model III source share results match
561 Model IV source share results. We presented Model I and Model IV source share re-

562 sults in Figure 8. The model results show an increase in source share contribution from
563 population related sources and livestock waste, and a decrease in source share from at-
564 mospheric deposition, fertilizer application, and non-agricultural land. Over the period of
565 analysis population increased by approximately 8% in the contiguous United States [*U.S.*
566 *Bureau of Census*, 2010] and population related source share increased by 11.5%. The
567 source share for atmospheric deposition decreased by 6.17%, however this decrease may
568 be related to lower precipitation in 2001 compared to 1992, as the model considered wet
569 deposition of inorganic nitrogen (nitrate and ammonia) as an estimate for atmospheric
570 deposition. The decrease in source share from fertilizer application was 3.0%. While the
571 fertilizer application rate data from USDA indicated increases in fertilizer use for some
572 regions of the United States, the model results suggest an overall decrease in source share
573 contribution for fertilizer. This decrease in fertilizer source share could simply be the
574 result of an increase in source share contribution from population related sources. How-
575 ever, this fertilizer source share decrease did not account for changes in farm management
576 practices such as conservation tillage, nutrient management, and BMPs, and nitrogen
577 removal by increasing crop yields, which all likely contributed to reducing nitrogen yield
578 from agricultural lands over the period of analysis. Source share from livestock waste
579 production increased by 1.2%, while source share from non-agricultural land decreased by
580 1.5%. Both sources showed little change as these sources remain nearly constant over the
581 period of analysis.

582 To understand the impact of land use type on incremental yield, we estimated incre-
583 mental yield loading from watersheds that have a dominant land use type (Figure 9 and
584 Table 4). If the land cover of the watershed was more than 90% urban land, we con-

585 sidered it as an urban watershed. We did the same for cropland and grass land. If the
586 land cover was more than 95% forest land, we considered it to be a forested watershed.
587 Results showed that while median incremental yield from urban watersheds experienced
588 only a 0.4% decrease from 1992 to 2001 (Model I vs Model IV results), this was due to an
589 offset between hydrologic changes and source contribution variability. If we consider only
590 changes in hydrology between 1992 and 2001 (Model I vs Model II), the results show a
591 decrease of 7.4% in median incremental yield. If we consider changes in source contribu-
592 tion only over the same time period (Model I vs Model III), the results show an increase
593 of 6.8%. Urban watersheds showed the most significant increase in nitrogen yield due to
594 variability in source contribution after controlling for hydrologic changes, however this
595 result should be interpreted in light of the fact that the study does not account for reduc-
596 tions in point sources of nitrogen that were implemented over the study period. Results
597 also showed an overall decrease in median incremental yield from cropland watersheds
598 of 16% from 1992 to 2001 (Model I vs Model IV). However, this decrease was primar-
599 ily due to hydrologic changes and not to decrease in source contributions. Incremental
600 yield decrease for cropland due to hydrologic changes (Model I vs Model II) was 12%
601 while decrease due to source contribution variability was 2.1% (Model I vs. Model III).
602 Forested watersheds showed an overall median incremental yield decrease of 5.8% (Model
603 I vs Model IV), but this change was primarily due to hydrologic changes (6.3%; Model
604 I vs Model II) and not decreases in source contribution (0.5%; Model I vs Model III).
605 Finally grassland watersheds showed a 3.8% overall decrease (Model I vs Model IV) with
606 a 7% decrease when considering only hydrologic changes (Model I vs Model II) and 3.5%
607 increase when considering only source contribution variability (Model I vs Model III) in

608 median incremental yield. The median incremental yield from the different land use types
609 were similar to previously published values (Table 4; *Frink* [1991]; *Ritter* [1988]; *Beaulac*
610 *and Reckhow* [1982]).

3.3. Model Limitations and Assumptions

611 While we have made note of limitations of our model, data, and methodology through-
612 out this discussion, we highlight the most significant of these limitations and assumptions
613 here. One key limitation is that SPARROW is a semi-empirical model and therefore has
614 inherit differences compared to a more mechanistic modeling approach. However, there
615 are challenges and issues associated with using a more mechanistic model at a national
616 scale including data availability, computational requirements, the need for some empirical
617 assumptions even in mechanistic models, subgrid heterogeneity, and problems of param-
618 eter estimation and calibration [*Beven*, 1989; *Smith et al.*, 2008; *Oreskes et al.*, 1994].
619 Therefore, while there are advantages to using a more mechanistic model for answering
620 our research question, such an approach is not without its own problems and so there
621 is still a role to play for simplified versions of process-based models like SPARROW, a
622 widely applied model both in the United States and abroad [*Elliott et al.*, 2005; *Hoos*
623 *and McMahon*, 2009]. While we are on one hand making an argument for SPARROW
624 as an appropriate model for our research question, we acknowledge the possibility that
625 SPARROW may be overly simplified for addressing our research question because it does
626 not, for example, include representations for processes such as nitrogen contributions from
627 groundwater, which may be significant for many regions of the country [*Ator and Ferrari*,
628 1997]. Also previous studies suggest potential problems with the statistical approaches
629 used in SPARROW. For example, *Qian et al.* [2005] suggested a structural weakness in

630 the SPARROW model that may cause spatial autocorrelation. Similarly, the fitted SPAR-
631 ROW model may be biased because it uses statistical inference of a nonlinear regression
632 based on the normality assumption [Fuller, 1995].

633 In this study we assumed that some watershed properties and river conditions were
634 constant from 1992 to 2001 including soil permeability, drainage density, streamflow rates,
635 and stream velocities. Streamflow rates and stream velocities are estimated based on long
636 term average condition. We therefore do not suspect large changes in these attributes
637 over the short time period of analysis, and we believe it to be a justifiable assumption
638 that changes in these parameters from 1992 to 2001 will not significantly alter the findings
639 reported in this study. Another important limitation of the study was our inability to
640 estimate observed nitrogen loading using NASQAN monitoring stations for 2001 at a
641 sufficient number of stations in order to calibrate SPARROW. Due to inadequate observed
642 nitrogen data, we were only able to estimate loading for 1992, when sufficient data were
643 available to estimate flow-concentration relationships. This estimation was based on long
644 term flow condition for 1970-2000 and long term mean load detrended to 1992. To address
645 the limitation of lack of monitoring data for 2001 we considered an alternative approach
646 to only calibrate the SPARROW model for 1992 and then use the calibration coefficients
647 to simulate the loading for 2001. This approach assumes that the model coefficients are
648 unchanged and only state conditions change. Alexander *et al.* [2008] presented a similar
649 approach for Mississippi River Basin SPARROW model study. We kept streamflows and
650 velocities (which are used to estimate travel times) as long term averages in all four
651 model scenarios to be consistent with the long term nitrogen loading estimates. Because
652 we were primarily interested in incremental nitrogen yield, we assumed that precipitation

653 would capture hydrologic changes and that keeping streamflow and velocity as long term
654 averages, to be consistent with instream nitrogen loading data, would be justifiable. An
655 extension to this study would be to identify nitrogen concentration and flow observations
656 from other datasets outside of NASQAN to use in the analysis to obtain better estimates
657 of instream nitrogen loading at monitoring stations for both time periods. In this case,
658 the streamflow and velocity estimates should also be updated to each of the base years of
659 the simulation.

4. Conclusion

660 The goal of this research was to better understand how the variability in source contribu-
661 tions (anthropogenic and non anthropogenic) and the changes in hydrology/climate affect
662 incremental nitrogen yield within the contiguous United States. We used the SPARROW
663 model, land use change products from the National Land Cover Dataset (NLCD), other
664 source inputs, watershed characteristics and instream nitrogen loading observations to
665 quantify these impacts for more than 60,000 watersheds in the contiguous United States.
666 We built four model scenarios to isolate these changes: Model I was a simulation of 1992
667 conditions, Model II was a modification of Model I where hydrologic inputs (eg. precipita-
668 tion and mean air temperature) were set to 2001 conditions, Model III was a modification
669 of Model I where source contribution inputs were set to 2001 conditions, and Model IV
670 was a simulation of 2001 conditions.

671 The results of this study suggest a decrease of 8.2% in median incremental nitrogen
672 yield from 1992 to 2001 (Model I vs Model IV). The decrease was 15% for atmospheric
673 deposition, 8.0% for non-agricultural land use, 6.1% for livestock waste, 5.4% for fertilizer
674 use, and 2.8% for population related sources. If only changes in nitrogen source contri-

675 butions were considered (Model I vs Model III), we observe only a small increase (0.1%)
676 in median incremental nitrogen yield. However, if only hydrology related changes were
677 considered (Model I vs Model II), we observe a decrease of 8.7% in median incremental
678 nitrogen yield. Therefore results of this analysis suggest that hydrologic changes – and
679 not decreases in nitrogen source contributions – were primarily responsible for changes
680 in nitrogen yield over the period of analysis. The results confirm previous research find-
681 ings that suggested significant changes in nitrogen sources to the Mississippi River Basin
682 were not observed after the early 1980s [*Goolsby et al.*, 1999; *National Agricultural Statis-*
683 *tics Service (NASS)*, 1998; *Alexander and Smith*, 1990; *Council of Environmental Quality*
684 *(CEQ)*, 1989]. The results also highlight the importance of precipitation and temperature
685 changes on regional scale nitrogen transport.

686 The model results suggest decreases in incremental nitrogen yield from some of the
687 highest yield producing areas (e.g., Upper Mississippi Basin). After separating hydrologic
688 and source contributions using the model scenarios we found that, although some of this
689 reduction was due to hydrologic differences between the two years (e.g., 2001 was a drier
690 year than 1992), the change was also due to reductions in source contributions, particularly
691 in the Mississippi Basin. The model results also show some areas that experienced an
692 increase in incremental nitrogen yield over the study period. From the model scenarios
693 we observe in some regions this increase was due to increases in source contribution (e.g.,
694 the upper portion of the Missouri Basin), but for other regions this increase was due
695 primarily to differences in hydrology (e.g., the Pacific Northwest).

696 We found from the model scenarios how each source was dependent on hydrologic vs
697 source contribution changes. For example, although overall median incremental nitrogen

698 yield decreased for population related sources (2.8%; Model I vs Model IV), this overall
699 decrease was due to an offset between an increase in source contribution (8.4%; Model I
700 vs Model III) and a decrease due to hydrologic changes (10%; Model I vs Model II). When
701 incremental nitrogen yield for each source is viewed spatially, we found that changes in
702 fertilizer application in particular was responsible for the overall decrease in nitrogen yield
703 for the Upper Mississippi Basin and the overall increase in nitrogen yield for the upper
704 portion of the Missouri Basin.

705 We found that source share of the total nitrogen budget for incremental yield increased
706 by 11.5% for population related sources and decreased by 6.17% and 3.0% for atmo-
707 spheric deposition and fertilizer application, respectively. Source share for livestock waste
708 and non-agricultural land remained nearly constant over the period of analysis. By group-
709 ing results for watersheds with dominate land use types, we found that urban watersheds
710 showed the largest percent increase in incremental nitrogen yield (6.8%) and cropland
711 had largest percent decrease (2.1%), after controlling for hydrologic changes. These re-
712 sults suggest that nitrogen from population related sources may becoming a significant
713 contributor of incremental nitrogen yield to streams, however it is important to stress
714 that this study was not able to account for changes in human management practices over
715 the period of analysis that are known to have occur but are not easy to quantify at the
716 scale of this study. Another key limitation of our model was that – because of an in-
717 sufficient amount of instream nitrogen observation data in and around the year 2001 –
718 we were unable to calibrate the model for 2001 conditions. Therefore we assumed that
719 model coefficients in SPARROW that describe such properties as instream and land-to-
720 water transport were constant between 1992 and 2001. However, we did evaluate the 2001

721 model predictions against the available instream nitrogen loading data and model showed
722 a good fit to these observed data. One possible extension of this work would be to identify
723 other reliable water quality datasets that could be used to improve estimates of instream
724 nitrogen loading in 2001 so that the SPARROW model can be re-calibrated for the 2001
725 model scenario.

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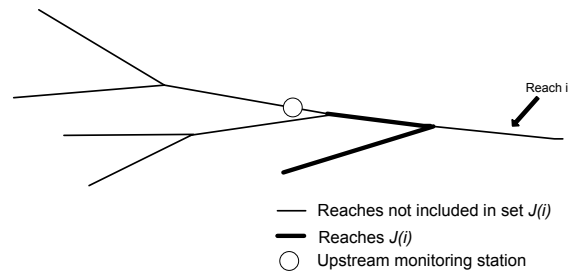


Figure 1. Schematic illustration for SPARROW reaches where $J(i)$ is the set of adjacent reaches upstream of reach i .

R^2 flux	0.885
R^2 yield	0.802
Mean square error	0.404
Number of observations	354

Table 1. Results of Model I (1992) calibration

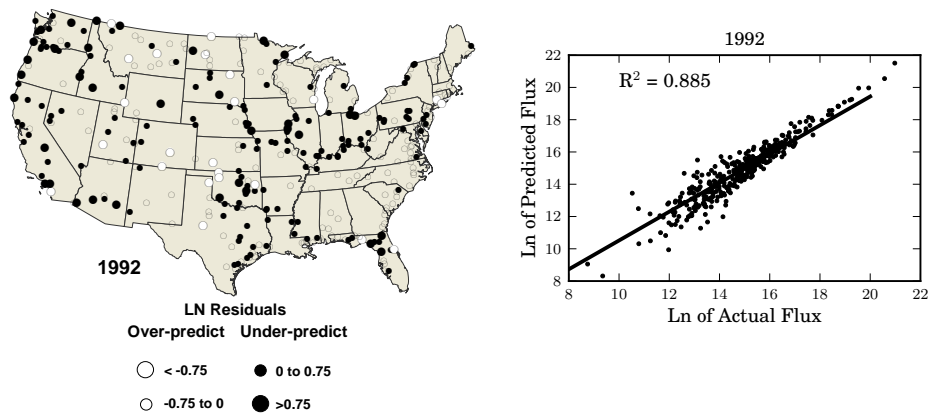


Figure 2. Predicted flux from Model I (1992) versus actual flux of total nitrogen for the 354 monitoring stations for 1992

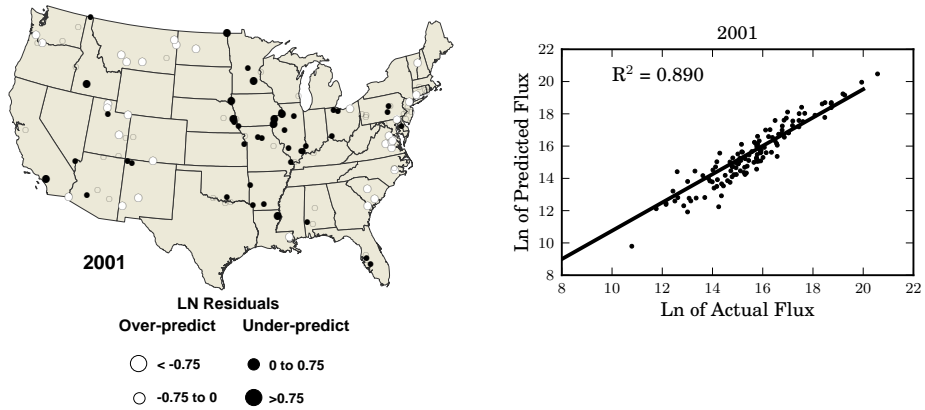


Figure 3. Predicted flux in Model IV (2001) versus actual flux of total nitrogen for the 122 monitoring stations for 2001

Parameter	Units	Coefficient	Standard Error	P-Value
Nitrogen Sources, β				
Population related Sources	kg person ⁻¹ yr ⁻¹	3.565	0.801	<0.05
Atmospheric deposition	dimensionless	0.282	0.179	0.117
Fertilizer application	dimensionless	0.248	0.046	<0.05
Livestock waste production	dimensionless	0.083	0.074	0.260
Non Agricultural Land	kg km ⁻² yr ⁻¹	310.05	53.68	<0.05
Land to water delivery, α'				
Soil permeability	in hr ⁻¹	-0.097	0.017	<0.05
Drainage density	km ⁻¹	2.006	0.479	<0.05
Mean annual air temperature	°C	-0.061	0.009	<0.05
Precipitation	cm	0.009	0.001	<0.05
In-stream decay, κ'				
κ_1 ($Q \leq 28.3 \text{ m}^3 \text{ s}^{-1}$)	day ⁻¹	0.226	0.031	<0.05
κ_2 ($28.3 \text{ m}^3 \text{ s}^{-1} < Q < 283 \text{ m}^3 \text{ s}^{-1}$)	day ⁻¹	0.059	0.025	<0.05
Reservoir decay, κ_r	m yr ⁻¹	7.182	1.938	<0.05

Table 2. Resulting model coefficients for the SPARROW model from the Model I (1992) calibration.

Incremental Nitrogen Yield (kg ha ⁻¹ yr ⁻¹)	10 th	25 th	50 th	75 th	90 th	Mean	Standard Deviation
Model I (1992)	2.9	4.52	8.16	14.11	24.64	17.91	603.51
Model II	2.66	4.19	7.45	13.12	22.98	16.66	528.83
Model III (2001 Sources)	2.95	4.64	8.17	14.18	24.63	18.03	600.64
Model IV (2001)	2.71	4.3	7.49	13.2	22.91	16.8	525.7
Total Change (kg ha⁻¹ yr⁻¹) Model IV - Model I	-0.19	-0.22	-0.67	-0.91	-1.73	-1.11	-
Total Change (%) Model IV - Model I	-6.55	-4.87	-8.21	-6.45	-7.02	-6.20	-
Change due to Hydrology (%) Model II - Model I	-8.28	-7.30	-8.70	-7.02	-6.74	-6.98	-
Change due to Sources (%) Model III - Model I	1.72	2.65	0.12	0.50	-0.04	0.67	-

Table 3. Distribution of incremental total nitrogen yield (kg ha⁻¹ yr⁻¹) for the different model scenarios.

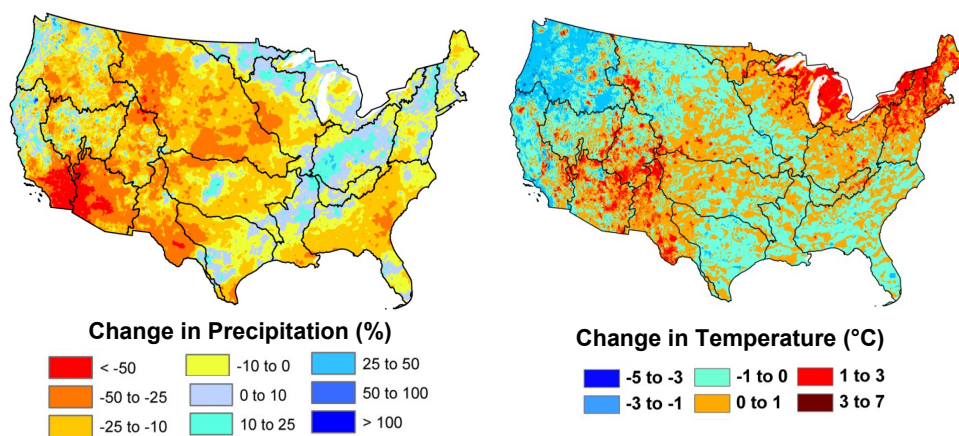


Figure 4. Percent change in precipitation and temperature between the input datasets used for the 1992 and 2001 models.

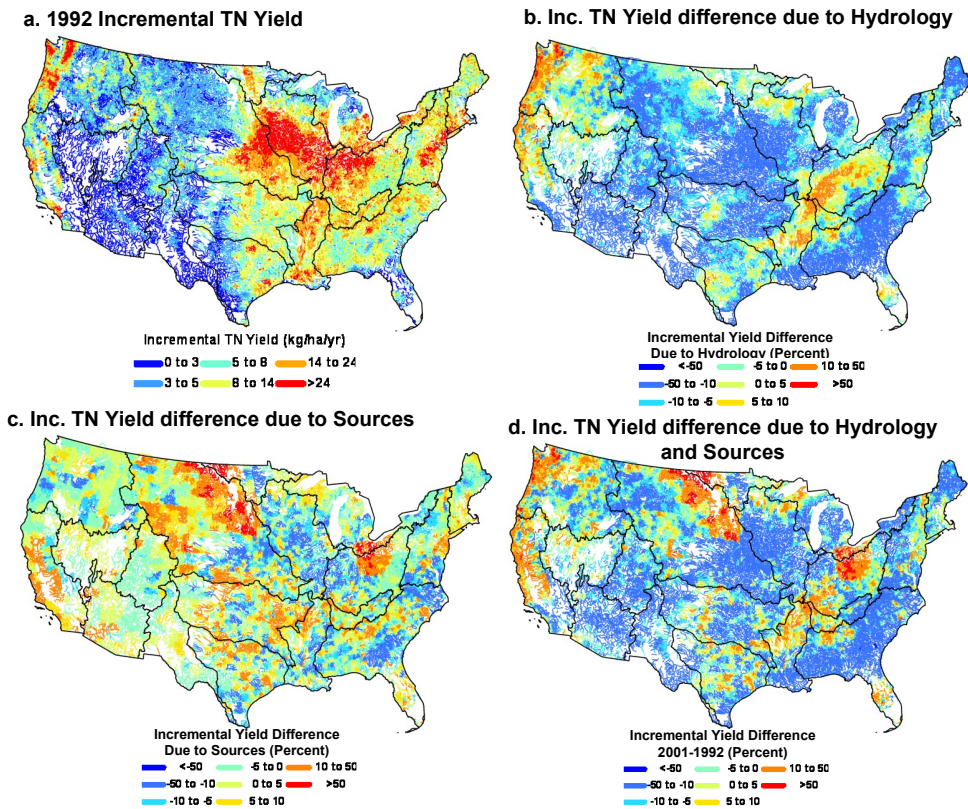


Figure 5. The top left map (a.) shows incremental nitrogen yield scenario in 1992. The top right map (b.) shows the percent difference in incremental nitrogen yield because of change in hydrology between 1992 and 2001. The bottom left map (c.) shows the percent difference in incremental nitrogen yield due to the change in source contribution between 1992 and 2001. The bottom right (d.) map shows the percent difference of incremental nitrogen yield due to overall change between 1992 and 2001.

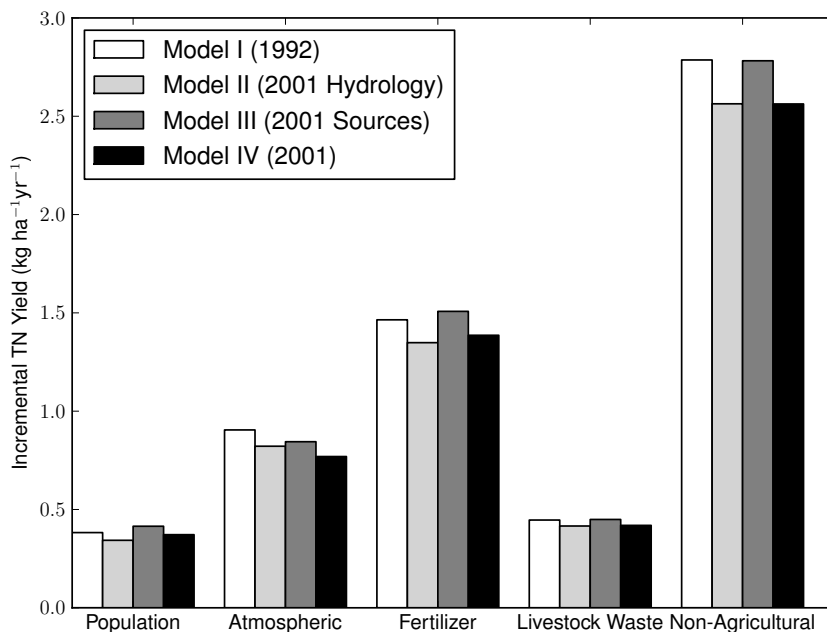


Figure 6. Median incremental total nitrogen yield ($\text{kg ha}^{-1} \text{ yr}^{-1}$) for different sources of nitrogen and for each model scenario.

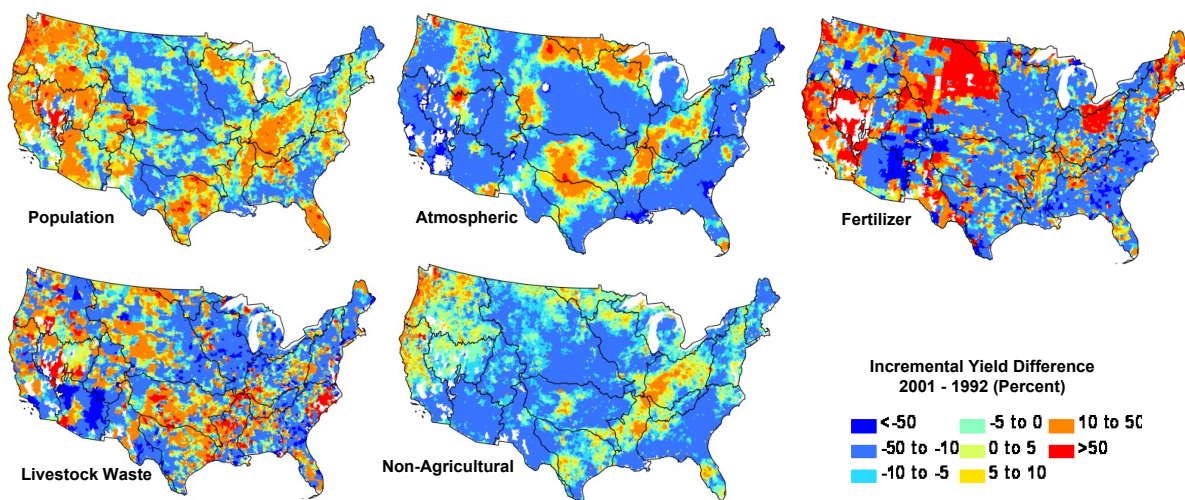


Figure 7. Percent difference of incremental nitrogen yield between Model I (1992) and Model IV (2001) for different sources of nitrogen

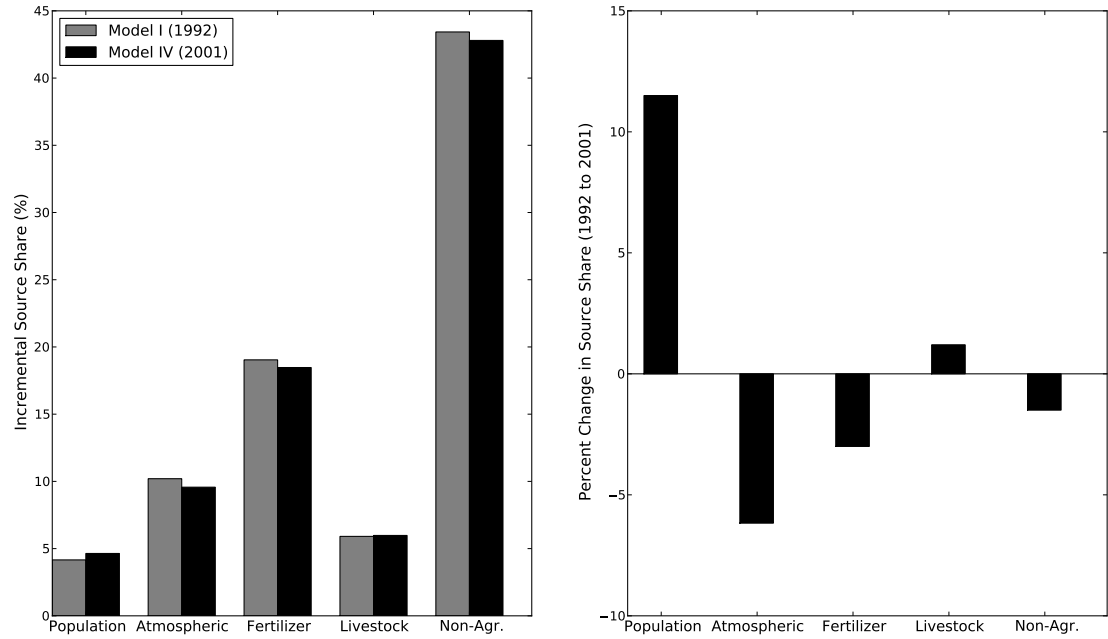


Figure 8. Comparison of median incremental total nitrogen source share (%) contribution for Model I (1992) and Model IV (2001).

Model No	Watershed Type ¹	Distribution of Incremental Yield (kg ha ⁻¹ yr ⁻¹)					Range of Yield values from Literature ² (kg ha ⁻¹ yr ⁻¹)
		10 th	25 th	50 th	75 th	90 th	
Model I (1992)	Urban	12.61	17.82	32.2	66.65	113.75	1.6 - 38.5
	Forest	5.49	7.77	11.13	16.38	30.21	0.1 - 10.8
	Crop	8.09	13.61	23.73	32	40.83	0.8 - 79.6
	Grass	1.73	2.49	3.67	5.26	8.26	0.1 - 30.8
Model II (2001 Hydrology)	Urban	12.16	15.17	29.81	60.31	119.84	1.6 - 38.5
	Forest	5.26	7.42	10.43	16.09	30.72	0.1 - 10.8
	Crop	7.67	12.75	20.81	27.29	34.91	0.8 - 79.6
	Grass	1.49	2.27	3.41	4.9	7.52	0.1 - 30.8
Model III (2001 Sources)	Urban	13.46	19.24	34.38	55.35	105.06	1.6 - 38.5
	Forest	5.62	7.82	11.08	16.37	29.92	0.1 - 10.8
	Crop	7.91	14.21	23.23	30.93	41.63	0.8 - 79.6
	Grass	1.73	2.55	3.8	5.45	8.61	0.1 - 30.8
Model IV (2001)	Urban	13	17.09	32.07	49.69	111.31	1.6 - 38.5
	Forest	5	7.5	10.49	16	29.96	0.1 - 10.8
	Crop	7.72	13	19.89	26.21	35.31	0.8 - 79.6
	Grass	2	2.32	3.53	5.1	7.86	0.1 - 30.8

Table 4. Incremental total nitrogen yield (kg ha⁻¹ yr⁻¹) for different model scenarios and literature estimates for watersheds with dominate landuse type in the United States.

¹The land cover types are based on the following percentage of land use area in SPARROW watersheds: urban(>90%), forest (>95%), crop land (>90%), grass (>90%)

² Literature reported values for incremental total nitrogen yield [*Frink, 1991; Ritter, 1988; Beaulac and Reckhow, 1982*].

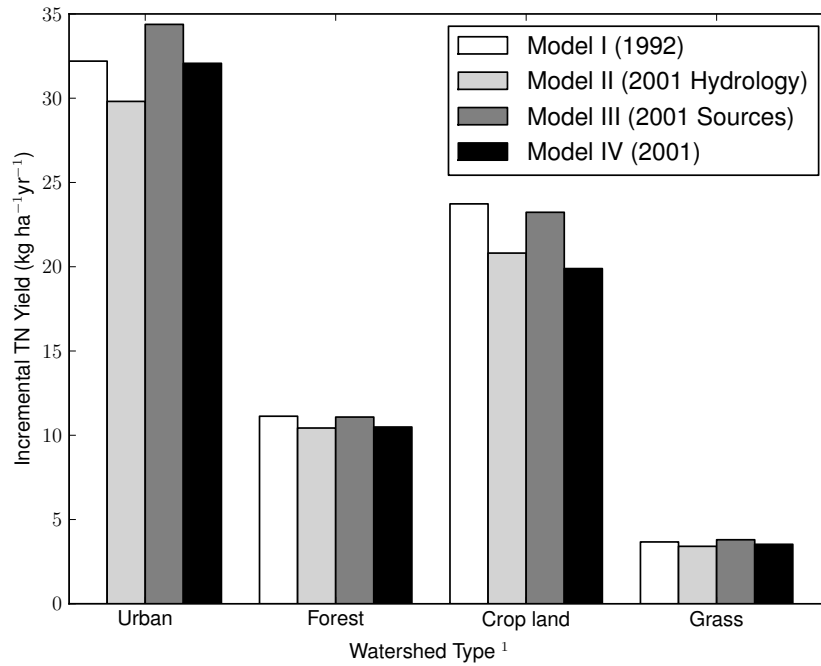


Figure 9. Median incremental total nitrogen yield ($\text{kg ha}^{-1} \text{ yr}^{-1}$) for major land use types in the United States for different model scenario.

¹The land cover types are based on the following percentage of land use area in SPARROW watersheds: urban (>90%), forest (>95%), crop land (>90%), grass (>90%)