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Toward disentangling the effect of hydrologic and

- ² nitrogen source changes from 1992 to 2001 on
- ³ incremental nitrogen yield in the contiguous United
- 4 States

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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 10 nitrogen yield for the contiguous United States. We ran different model scenarios to separate the 11 effects of changes in source contributions from hydrologic changes for the years 1992 and 2001, 12 assuming that only state conditions changed and that model coefficients describing the stream 13 water-quality response to changes in state conditions remained constant between 1992 and 2001. 14 Model results show a decrease of 8.2% in the median incremental nitrogen yield over the period 15 of analysis with the vast majority of this decrease due to changes in hydrologic conditions rather 16 than decreases in nitrogen sources. For example, when we changed the 1992 version of the model 17 to have nitrogen source data from 2001, the model results showed only a small increase in median 18 incremental nitrogen yield (0.12%). However, when we changed the 1992 version of the model to 19 have hydrologic conditions from 2001, model results showed a decrease of approximately 8.7% in 20 median incremental nitrogen yield. We did, however, find notable differences in incremental yield 21

estimates for different sources of nitrogen after controlling for hydrologic changes, particularly 22 for population related sources. For example, the median incremental yield for population related 23 sources increased by 8.4% after controlling for hydrologic changes. This is in contrast to a 2.8%24 decrease in population related sources when hydrologic changes are included in the analysis. 25 Likewise we found that median incremental yield from urban watersheds increased by 6.8% after 26 controlling for hydrologic changes – in contrast to the median incremental nitrogen yield from 27 cropland watersheds, which decreased by 2.1% over the same time period. These results suggest 28 that, after accounting for hydrologic changes, population related sources became a more signifi-29 cant contributor of nitrogen yield to streams in the contiguous United States over the period of 30 analysis. However, this study was not able to account for the influence of human management 31 practices such as improvements in wastewater treatment plants or Best Management Practices 32 (BMPs) that likely improved water quality, due to a lack of data for quantifying the impact of 33 these practices for the study area. 34

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

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

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

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

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

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

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

In the following section we provide details of the study methodology summarized in the previous paragraph. This section is followed by a presentation and discussion of the study results as a means for understanding changes in the spatial distribution of incremental nitrogen yield over the decadal study period. Finally we present a summary and concluding statements from this work, along with suggested extensions to the study 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

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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).

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

$$L_{i} = \left[\left\{ \sum_{j \in J(i)} L_{j} \right\} A(Z_{i}^{s}, Z_{i}^{r}; \kappa_{s}, \kappa_{r}) + \left\{ \sum_{n=1}^{N} S_{n,i} \beta_{n} \right\} D_{n}(Z_{i}^{D}; \alpha) A'(Z_{i}^{s}, Z_{i}^{r}; \kappa_{s}, \kappa_{r}) \right] \epsilon_{i}$$

$$(1)$$

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(.) 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(.) and if the waterbody *i* ¹³⁹ is a lake or reservoir, then Z_i^r and κ_r define the function A(.).

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

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¹⁴⁴ a conversion factor between the source units and flux units estimated by the model. The ¹⁴⁵ function D(.) 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'(.) 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].

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

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

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

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

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

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

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where κ_r is the reservoir decay coefficient estimated by the model and q_i^r is the areal 163 hydraulic loading (ratio of reservoir outflow to surface area, in units of distance per time). 164 The model is first calibrated using Equations 1-4 to estimate nitrogen loading for all 165 monitored reaches. The result of the regression is estimates of the coefficient values for 166 β , α , and κ that minimize errors between observed and predicted loadings for monitored 167 reaches. Once these coefficients have been determined, the model is applied to predict 168 nitrogen delivery for unmonitored reaches within the river network. The model results 169 in predictions of incremental and total nitrogen yield for each reach in the river network 170 dataset and for each nitrogen source considered by the model. While there are limitations 171 to the modeling approach used by SPARROW, as we will discuss in greater detail in 172 Section 3.3, we have used the model because it offers a practical blend of process-based 173 and empirical modeling appropriate for regional-scale assessments, where it is difficult to 174 parameterize physical-based models of hydrologic and biogeochemical processes. 175

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

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2001. Our initial approach was to calibrate the model to 1992 and 2001 observed loading 186 data separately, but there were insufficient instream concentration observations available 187 for 2001 to support this approach. Therefore we calibrated the SPARROW model for 188 1992 conditions and then simulated stream loads for 2001 conditions assuming that the 189 SPARROW model coefficients were unchanged and only state conditions changed. We 190 then used the SPARROW model coefficients from the Model I calibration to also predict 191 loadings for the Model II and Model III scenarios. Our justification for this assumption 192 is that because there were no major departures in state conditions between the decade 193 separating the two study periods, we can assume that the statistical relationship that 194 forms the basis of the model formulation is valid for both years. To verify our assumptions, 195 we evaluated the 2001 model by comparing predicted loadings to the limited set of available 196 observed loadings. A similar approach of calibrating the model for 1992 and then using 197 the calibration coefficients to predict for 2002 was used by Alexander et al. [2008] for the 198 Mississippi River Basin. 199

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

$$Yield_{i} = \left\{ \sum_{n=1}^{N} S_{n,i}\beta_{n} \right\} \times D_{n}(Z_{i}^{D};\alpha)A'(Z_{i}^{s},Z_{i}^{r};\kappa_{s},\kappa_{r}) / Area_{i}$$

$$(5)$$

where *i* is the number of incremental catchments, $Area_i$ is the area of the catchment and Yield_i is the incremental yield in kg ha⁻¹ 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

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

2.2. Data Preparation

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

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

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

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

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

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

$$ln(l) = \lambda_0 + \lambda_1 t + \lambda_2 sin(2\pi t) + \lambda_3 cos(2\pi t) + \lambda_4 ln(q) + \lambda_5 [ln(q)]^2 + \alpha$$
(6)

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

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

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

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

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

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

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

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

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

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

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

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

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

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

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3. Results and Discussion

3.1. Model Calibration

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

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

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

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

The model calibration resulted in instream loss coefficients (κ) that were greater for lowflow streams (Q < 28.3 m³ s⁻¹) compared to medium-flow streams (28.3 m³ s⁻¹ \leq Q \leq 283 m³ s⁻¹). These coefficients were found to be statistically significant for low flow streams (p < 0.05), but not for medium streams (p > 0.05). This is consistent with previous SPARROW model results and the conclusion that loss rate decreases with increasing stream size [*Alexander et al.*, 2000; *Hoos and McMahon*, 2009]. Model prediction for

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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

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

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

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

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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.

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

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

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

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

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

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

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

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

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

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sidered it as an urban watershed. We did the same for cropland and grass land. If the 585 land cover was more than 95% forest land, we considered it to be a forested watershed. 586 Results showed that while median incremental yield from urban watersheds experienced 587 only a 0.4% decrease from 1992 to 2001 (Model I vs Model IV results), this was due to an 588 offset between hydrologic changes and source contribution variability. If we consider only 589 changes in hydrology between 1992 and 2001 (Model I vs Model II), the results show a 590 decrease of 7.4% in median incremental yield. If we consider changes in source contribu-591 tion only over the same time period (Model I vs Model III), the results show an increase 592 of 6.8%. Urban watersheds showed the most significant increase in nitrogen yield due to 593 variability in source contribution after controlling for hydrologic changes, however this 594 result should be interpreted in light of the fact that the study does not account for reduc-595 tions in point sources of nitrogen that were implemented over the study period. Results 596 also showed an overall decrease in median incremental yield from cropland watersheds 597 of 16% from 1992 to 2001 (Model I vs Model IV). However, this decrease was primar-598 ily due to hydrologic changes and not to decrease in source contributions. Incremental 599 yield decrease for cropland due to hydrologic changes (Model I vs Model II) was 12%600 while decrease due to source contribution variability was 2.1% (Model I vs. Model III). 601 Forested watersheds showed an overall median incremental yield decrease of 5.8% (Model 602 I vs Model IV), but this change was primarily due to hydrologic changes (6.3%; Model603 I vs Model II) and not decreases in source contribution (0.5%; Model I vs Model III). 604 Finally grassland watersheds showed a 3.8% overall decrease (Model I vs Model IV) with 605 a 7% decrease when considering only hydrologic changes (Model I vs Model II) and 3.5%606 increase when considering only source contribution variability (Model I vs Model III) in 607

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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

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

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

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

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

4. Conclusion

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

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

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

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

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

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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.

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

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model predictions against the available instream nitrogen loading data and model showed
a good fit to these observed data. One possible extension of this work would be to identify
other reliable water quality datasets that could be used to improve estimates of instream
nitrogen loading in 2001 so that the SPARROW model can be re-calibrated for the 2001
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	$\rm kg \ km^{-2} \ yr^{-1}$	310.05	53.68	$<\!0.05$
Land to water delivery, α'				
Soil permeability	in hr^{-1}	-0.097	0.017	$<\!0.05$
Drainage density	$\rm km^{-1}$	2.006	0.479	$<\!0.05$
Mean annual air temperature	$^{\circ}\mathrm{C}$	-0.061	0.009	$<\!0.05$
Precipitation	cm	0.009	0.001	$<\!0.05$
In-stream decay, κ'				
$\kappa_1 (Q \le 28.3 m^3 s^{-1})$	day^{-1}	0.226	0.031	$<\!0.05$
$\kappa_2 (28.3 m^3 s^{-1} < Q < 283 m^3 s^{-1})$	day^{-1}	0.059	0.025	$<\!0.05$
Reservoir decay, κ_r	${\rm m~yr^{-1}}$	7.182	1.938	$<\!0.05$

 Table 2.
 Resulting model coefficients for the SPARROW model from the Model I

 $\left(1992\right)$ calibration.

Incremental Nitrogen Yield $(\text{kg ha}^{-1} \text{ yr}^{-1})$	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 $(\text{kg ha}^{-1} \text{ yr}^{-1})$ for the different model scenarios.

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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.

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Figure 6. Median incremental total nitrogen yield (kg $ha^{-1} yr^{-1}$) 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	
		10^{tn}	$25^{ au n}$	$50^{\tau n}$	$75^{\tau n}$	90 th	$({ m kg}~{ m ha}^{-1}~{ m yr}^{-1})$
Model I	Urban	12.61	17.82	32.2	66.65	113.75	1.6 - 38.5
(1992)	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	Urban	12.16	15.17	29.81	60.31	119.84	1.6 - 38.5
(2001 Hydrology)	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	Urban	13.46	19.24	34.38	55.35	105.06	1.6 - 38.5
(2001 Sources)	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	Urban	13	17.09	32.07	49.69	111.31	1.6 - 38.5
(2001)	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 (kg ha⁻¹ 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%)