ABSTRACT

STRICKLING, HAYDEN LELAND. A Bayesian Hierarchical Model for Estimating Nutrient Export Rates in a Developing Watershed. (Under the direction of Dr. Daniel Obenour).

Watershed models based on nonlinear regression are useful tools for estimating the magnitude of loading rates (i.e., export coefficients) for various pollutant sources within large-scale river basins. Few such models, however, have incorporated temporal variability in either source distributions or climate, despite evidence that precipitation is the primary driver in interannual variability in loading rates. The model developed here includes changes in precipitation, land-use, point-source discharge, and livestock operations to capture temporal variability in nitrogen loads. Precipitation is incorporated directly in the formulation of export rates using hierarchically related coefficients that vary by source type. In-stream and reservoir retention of nitrogen is included to account for nitrogen sinks within the watershed. A Bayesian-hierarchical approach is employed to integrate uncertainty in loading estimates, include prior knowledge of parameters, address intra-watershed correlation, and estimate export coefficients probabilistically. This method is applied to three North Carolina river basins that have experienced substantial growth in urban development and livestock operations in the past few decades, and where eutrophication-related water quality problems are common. Results indicate that livestock operations are a significant contributor of nitrogen throughout much of the study area. Precipitation is shown to have a larger influence on export rates for agricultural than for developed lands, creating a system dominated by agricultural TN during high precipitation years and by developed regions during low precipitation years.

A Bayesian Hierarchical Model for Estimating Nutrient Export Rates in a Developing Watershed

by
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INTRODUCTION

Excessive nitrogen in surface waters can cause algal blooms, hypoxia, and fish kills, resulting in diminished water quality and impacted fishing and recreational industries (Conley et al., 2009). With increasing trends in nutrient delivery from urbanization and livestock production seen in the past decades (Billen et al., 2010), determining the sources and fate of nitrogen within a watershed is imperative for implementing effective programs for nutrient mitigation. Hybrid empirical/process-based models, such as the Spatially Referenced Regressions of Contaminant Transport on Watershed Attributes (SPARROW), can be important tools for modeling nutrient loadings in large river basins where complex process-based models can be computationally intensive (Smith et al., 1997; Borah & Bera, 2004). Hybrid models spatially relate watershed and land-use characteristics to water-quality data by incorporating simple mechanistic processes in a nonlinear regression framework, allowing for the identification of sources and sinks of nutrients within a watershed (Smith et al., 1997). The statistical estimation of a small set of model parameters allows for a robust assessment of parameter and nutrient loading estimate uncertainty, and avoids issues of equifinality and over-parameterization (Beven, 2006; Beck, 1987) that can occur with processed-based models (Schwarz et al., 2006; Cao et al., 2006). The increasing availability of regional datasets provides an opportunity for hybrid models to incorporate rich spatial and temporal information to characterize nutrient sources and watershed dynamics, and provide a data-driven line of evidence to support watershed management (McMahon et al., 2003).

Hybrid watershed models typically characterize pollutant sources in terms of land-use export rates (kg/ha/yr), also known as export coefficients (ECs), which can be useful for forecasting outcomes of nutrient management or development scenarios. The EC concept has been effective in relating anthropogenic land-use practices to downstream water quality in past studies (Vollenweider et al., 1971; Norvell et al., 1979; Johnes, 1996), and in the development of Total Maximum Daily Load (TMDL) allowances (Mattson & Isaac, 1999). Often, ECs are estimated through field studies of small single land-use watersheds (Beaulac & Reckhow, 1982). These estimates, however, may not be available for a particular region or may not represent a mean rate for a basin-scale study where heterogeneity of land-use practices and characteristics exist (Shrestha et al., 2008). Hybrid models, on the other hand, can provide an estimate of mean loading rates over large regions through empirical estimation based on available water quality data, watershed characteristics, and land-use data (Alexander et al., 2004). Statistical approaches can also quantify uncertainty in EC estimates, which is important in assessing the reliability of model estimates for considering risk in watershed management (Reckhow, 2003; Xia et al., 2016).

A limitation of both the EC concept and hybrid models, however, is that temporal variability in export rates is rarely considered. ECs are static estimates representative of mean conditions (Beaulac & Reckhow, 1982), and most hybrid models estimate export rates from temporally-averaged land-use and watershed characteristics data (e.g., Smith et al.,1997). In reality nutrient export is influenced by temporally varying factors such as precipitation and land-use trends that can cause highly variable loading rates (Fraterrigo & Downing, 2008; Kaushal et al., 2008). Addressing temporal variability in loading estimates can facilitate

understanding of the causes of adverse loading moments, which can inform more effect management strategies (McClain et al., 2003).

Few efforts have been made to include interannual variability in hybrid watershed models. Wellen et al. (2012) added temporally variable climatic variables as an additive component to the SPARROW regression and Xia et al (2016) included land-use change. In addition, both of these studies used dynamic parameter estimation, allowing relationships to vary over time. However, for hybrid models, precipitation has yet to be incorporated into the formulation of nutrient export, and the interplay between changes in land use and climate has received little attention. Since both precipitation and land use are primary drivers of interannual variability in nitrogen export (Haith & Shoemaker, 1987; Sinha & Michalak, 2016), their inclusion within the mechanistic formulation of hybrid models could improve our understanding of watershed loading dynamics.

Bayesian hierarchical modeling is a potentially powerful tool in addressing the complex nutrient loading dynamics of a watershed (Reckhow & Chapra, 1999). Model development within a Bayesian framework readily allows for the integration of spatial and temporal variability at different organizational levels, uncertainty in model inputs, and prior knowledge of watershed mechanics (Qian et al., 2010; Gronewold & Borsuk, 2010; Zobrist & Reichert, 2006; Rode et al., 2010). Bayesian model results consist of posterior distributions that describe both parameters and predictions, which can be used to evaluate hypotheses about the relative importance of different sources and sinks of pollution, and which can be used to assess the efficacy of different management scenarios with in a risk-based framework (Reckhow & Chapra, 1999; Qian et al., 2005).

Understanding temporal variability in watershed nutrient loading is particularly relevant for watersheds subject to changing land use practices. Our study consists of three such watersheds in eastern North Carolina: Neuse River Basin (NRB), Cape Fear River Basin (CFRB), and Tar Pamlico River Basin (TPRB). Estuaries in these basins have been subject to toxic algal blooms, hypoxia and fish kills in the past decades, resulting in diminished water quality, and their inclusion in the United States Environmental Protection Agency (EPA) 303(d) list of impaired waters in the 1990s (Paerl et al., 1998; Burkholder et al., 2006). The causes of impairment were identified as chlorophyll a level exceedance for the NRB and TPRB, and low dissolved oxygen (DO) levels for the CFRB, both attributed to up-stream nitrogen loadings (NCDEQ, 2017). As mandated by the Clean Water Act (Clean Water Act, 1972) for impaired water bodies, the North Carolina Division of Water Quality developed a Total Maximum Daily Load (TMDL) requiring point and distributed source total nitrogen (TN) loading reductions of 30% in the NRB and TPRB, relative to 1990-1994 base-line years (NCDENR, 1999; NCDENR, 1994). By 2008, however, the 30% reduction goal had not been achieved (NCDENR, 2009; NCDEQ, 2010).

While TMDLs have been shown effective in decreasing nitrate export from point sources, a recent increase in organic nitrogen (ON) has been reported (Lebo et al., 2012). The causes of this increasing trend remain undetermined though hypotheses have been made. Some have suggested waste from concentrated animal feeding operations (CAFOs) and noted the trends in increased livestock production in eastern North Carolina (Burkholder et al., 2006; Cahoon et al., 1999; Mallin et al., 2015). The EPA defines a CAFO as an animal feeding operation comprising over 2,500 hogs and 30,000 chickens (USEPA, 2000). From 1985 to

1995 hog and chicken production increased 200% and 50% respectively in NC, prompting the implementation of a state-wide moratorium in 1997 on new hog operations, still effective to date (Cahoon et al., 1999; S.L. 1997-458, 1997). Chicken operations expanded steadily since the 1990s making North Carolina one of the nation's top producers of chickens by 2012 (USDA NASS, 2014). CAFOs in North Carolina are mostly located in the coastal plains region of eastern NC, where their proximity to coastal ecosystems make nutrient contamination from waste particularly concerning (Harden, 2015). Most chicken operations in NC store waste as dry-litter which is sold as fertilizer for surrounding fields (Harden, 2015). Hog operations, on the other hand, tend to store waste in one or more on-site lagoons and are permitted to apply effluent as spray-irrigation on nearby fields during the growing season (NCDENR, 2014). Despite permitting restrictions, effluent is sometimes applied in excess to the assimilative capacities of crops and excess nutrients can be conveyed to streams through drainage systems (Kellogg et al., 2000; Evans et al., 1984). Elevated levels of nitrogen have been observed in the groundwater and streams of CAFO dominated NC watersheds (Mallin et al., 2015; Gilmore et al., 2016). A recent United States Geological Survey (USGS) study tested 45 NC coastal plain streams over the course of a year, and found a correlation of hog and chicken density and spray-field acreage with downstream nutrient contamination (Harden, 2015). Groundwater contamination from poorly-lined lagoon pits and the infiltration of excess nutrients from agricultural soils has been reported and could account for off-site groundwater nutrient loadings into surface waters, especially when sub-surface tile drains are used in fields (Dukes & Evans, 2006; Harden & Spruill, 2008; Harden, 2015; Spruill et al., 2005). Flooding of

lagoons from extreme weather can also contaminate surface waters, a concern especialy for the hurrican-prone regions of eastern NC (Burkholder et al., 1997).

Another potential driver of this increasing ON loading into NC estuaries is land-use activities such as agriculture and urban development, resulting from population growth nearly doubling from 1990 to 2010 in the three basins (U.S. Census Bureau, 2017). Urban development increases nutrient transport to surface waters through the destruction of riparian forested and wetland buffers, disruption of soils resulting in sediment transport of nutrients, and increased impervious development that accelerates nutrient wash-off from lawn fertilizers and pet waste (Lee et al., 2006; Reavie & Baratono, 2007; Hopkinson & Vallino, 1995; Hogan & Walbridge, 2007; Faulkner, 2004). Agricultural cultivation also affects water quality through transport of fertilizer inputs through ditches and sub-surface tile drains (Cambardella et al., 1999; Robertson & Vitousek, 2009) though agricultural cultivation has not increased substantially in the study area in recent decades. To investigate the relationship between land-use and nutrient loadings of dissolved organic nitrogen (DON) in the NRB, Osburn et al. (2016) matched water sample fluorescence tracers with that of various DON sources and found soil DON to be the dominant type in the NRB, just above chicken litter and street runoff. Soil DON likely comes from disruption of soils caused by urban development and leaching of nitrogen from agricultural soils during precipitation events (Osburn et al., 2016). Thus, the combined effects of urban and agricultural land-use practices could account for the increase in ON noted above.

Hybrid models have been employed to assess sources of nutrient loading in NC's coastal watersheds. McMahon et al. (2003) developed a SPARROW model to estimate TN

export from distributed and point-sources in the NRB, TPRB, and CFRB. SPARROW model predictions for the NRB show a watershed nitrogen budget dominated by agricultural inputs from the lower part of the basin where shorter transport distances allow for fewer in-stream losses (McMahon et al., 2003). Qian et al. (2005) built upon this model by introducing a Bayesian hierarchical framework to test different model formulations and account for spatial autocorrelation of residuals and variability among watersheds. While providing useful estimates of nutrient delivery for this region generally reflective of conditions prior to 2000, neither of these models directly address precipitation variability and changing land-use practices, nor include CAFOs as potential sources of nitrogen.

Developed here is a novel adaptation of the hybrid approach to watershed modeling, focused on estimating pollutant ECs and instream loadings over time, in a developing watershed. Our study includes three river basins in eastern NC that have seen substantial increases in CAFOs and urban development. Building on previous modeling efforts, changes in land-use and precipitation rates are integrated into the model to capture temporal variability in nitrogen export. CAFO-related livestock data are also included for the first time as a potential source of nitrogen loading, given their prominence in this region. In addition, uncertainty in loading estimates and prior knowledge of parameters are integrated into the model using a Bayesian hierarchical framework, which also allows for a probabilistic understanding of watershed loading.

METHODS

Model development involved several steps. First, annual loading rates for sub-watersheds were estimated from available TN concentration and flow data, and the uncertainty in these estimates were determined through sub-sampling. Next annual data for the TN sources used in the model were collected and aggregated for each sub-watershed. A basin-wide stream and water-body network was developed to account for TN retention and annual precipitation data was obtained for each sub-watershed to address the effect of precipitation on nutrient delivery rates. Next a non-linear regression model formulation was developed to relate TN source, stream and water-body network, and precipitation data to the estimated loading rates. Finally, the model was developed in a Bayesian-hierarchical framework that incorporates the uncertainty in the loading rate estimates, a random effects term grouped by sub-watershed, and the prior distributions of model parameters, obtained from the literature.

Study Area

The NRB, CFRB and TPRB boundaries used in this study are the USGS HUC 6 delineations, with areas of 15718, 23943, and 16625 km^2 respectively. The three basins extend from the relatively hilly and urban Piedmont region in the west, to the flatter and largely agricultural Coastal Plains in the east. The Piedmont is characterized by having clay, erodible soils; and the Coastal Plain by sandy, well-drained soils, wetlands and estuarine habitats. All geospatial data processing was performed using ArcGIS by ESRI (ESRI, 2011). Watersheds were delineated for each sampling station using a 30-meter digital elevation model from the

USGS National Elevation Dataset (USGS, 2017) (Figure 1). The study period is 1994 to 2012, as limited by nutrient-source data availability.

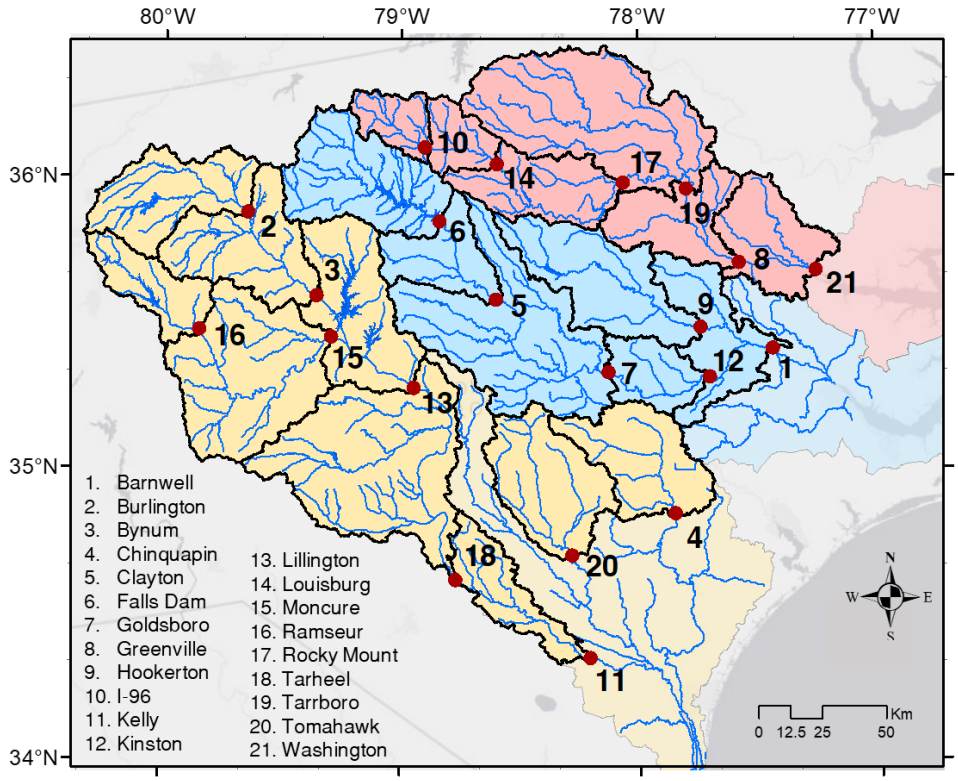

Figure 1. The North Carolina river basins used in this study: Tar Pamlico (red), Neuse (blue), and Cape Fear (yellow). Darker shades outline the portion of each river basin used in this study, where available water-quality data exists. Red points are water-quality and flow monitoring stations used to delineate the modeled incremental watersheds.

Estimates of Annual TN Loads

Annual TN loadings were estimated from stream flow and water quality data. Streamflow were obtained from the USGS NWIS database, and water quality data were obtained from the Water Quality Portal, a compilation of state and federal water quality datasets (National Water Quality Monitoring Council, 2010). A total of 21 sampling stations throughout all three basins were selected for having continuous flow and at least 10 years of water quality data (Figure 1). The inclusion of a station required total Kjeldahl nitrogen, nitrate, and nitrite data, since TN is calculated as the sum of these nitrogen species. Flow data were continuous for all stations except Barnwell, the furthest downstream station on the Neuse, where flows preceding the year 2000 were estimated by linear regression using flows at Kinston and Hookerton as predictor variables (r^2 = .96). To estimate annual TN loads, the USGS Weighted Regressions on Time, Discharge and Season (WRTDS) method was used (Hirsch & Cicco, 2015). WRTDS imputes missing nutrient concentration data based on a regression with time of year (season), time (in years), and discharge. Furthermore, WRTDS uses a flexible semi-parametric approach where unique regression coefficients are determined for each estimation date, by more heavily weighting observations with similar seasonality, year, and discharge (Hirsch et al., 2010). This flexible approach is particularly important for our study area, where the magnitudes of different nutrient source types are changing over time. Incremental nitrogen loadings for each incremental watershed were calculated by subtracting the WRTDS estimate of each monitroing station by the estimates from the directly contributing upstream station or stations.

Uncertainty Analysis for Load Estimates

Uncertainty in WRTDS estimates was determined by relating error variance to sampling frequency at monitoring stations. This relationship was developed by sub-sampling at four stations, selected for having nitrogen concentration samples taken nearly every day over a span of at least 7 years. Subsampling of concentration data was implemented based on different sampling schedules: samples taken every two months, 1 month, 2 weeks, 1 week, and 4, 3, and 2 days. Samples were selected at random within a sliding window corresponding to each sampling schedule to simulate real-life variability in sample collection. For each sampling schedule, WRTDS was run 300 times (simulations) with a different randomly selected subsample used for each simulation. A 3-year half-window width was used in the WRTDS estimation, and annual loading estimates were taken from year(s) within 3 years from the start and finish of the simulation. The window width refers to the time-span for the distribution of weights used for the WRTDS regression analysis, where data outside this window will be assigned a weight nearly zero (Hirsch et al., 2010). Residuals were determined by subtracting WRTDS loading estimates based on subsamples from those obtained using the complete dataset. For each year (t) of each simulation, a coefficients of variation (CV) was calculated:

$$CV_t = \frac{\sigma_t}{\mu_t} \tag{1}$$

where σ_t is the standard deviation of residuals, μ_t is the mean loading estimate (for a simulation in year *t*). Results from subsampling are shown in Appendix D, where CV values are plotted as a function of number of samples per year. A power function was fit to this distribution and used to estimate CV based on number of samples taken per year ($R^2 = 0.95$):

$$CV_{i,t} = 0.9662 * X_{i,t}{}^{-0.783} \tag{2}$$

where $CV_{i,t}$ is the coefficient of variation for watershed *i* and year *t*, and $X_{i,t}$ is the number of samples taken. Equation 2 was applied to calculate the CV for each annual WRTDS loading estimate for each of the 21 sampling stations used in this study. These CVs were then used to determine the standard deviation of each loading estimate, consistent with equation 1. The incremental error variance, representing the uncertainty in the change in loading across an incremental watershed, is calculated based on the relationship between two correlated random variables (Kottegoda & Rosso, 2008):

$$\tilde{\sigma}_{i,t}^2 = \sigma_{z,i+1,t}^2 + \sigma_{z,i,t}^2 - 2\rho_{i+1,i}\sigma_{z,i+1}\sigma_{z,i} \tag{3}$$

where $\tilde{\sigma}_{i,t}^2$ is the incremental error variance for incremental watershed *i* and year *t*, $\sigma_{z,i,t}^2$ is the WRTDS error variance for station i, $\sigma_{z,i+1,t}^2$ is the error variance for the upstream station to station *i*, $\rho_{i+1,i}$ is the correlation of loadings between watershed *i* and the upstream watershed, $\sigma_{z,i+1}$ is the WRTDS standard deviation of station *i*, and $\sigma_{z,i}$ is the WRTDS standard deviation of the upstream station.

Nitrogen Source Inputs

Three types of nitrogen sources were included in the model: distributed sources associated with land use, point-source dischargers, and CAFOs. Land-use data were acquired from the U.S. conterminous wall-to-wall anthropogenic land-use trend (NWALT) dataset for the years 1992, 2002 and 2012 (Appendix A) (Falcone, 2015). For the purposes of this study, land use classifications were aggregated into three general categories: *developed, agriculture,* and *wild*. Both *low-use* and *low-use/conservation* classifications were grouped into *wild*, and three classifications within *production* make up the *agriculture* group: *Crops, Pasture/Hay, and Grazing Potential*. The developed group comprises a range of land-use classifications including low-, medium-, and high-density commercial, industrial, recreational, and residential areas. Land-use area for each of the three groups was calculated on the Hydrologic Unit Code (HUC 12) spatial scale. Land-use areas for years in between the NWALT datasets were calculated through linear interpolation.

National Pollutant Discharge Elimination System (NPDES) data for dischargers in the three river basins was obtained from the North Carolina Department of Environmental Quality (Marla Sink, personal communication, 2016). NPDES data only exists from 1994 to 2016, which limits the starting year for this study. This dataset includes monthly concentration and flow rate estimates from 71 major (> 3785 m^3/d) and 242 minor dischargers located in the NRB, TPRB and CFRB. Because monthly estimates in a given year were often incomplete, annual loading estimates for each discharger were calculated from mean monthly concentrations and flow rates for each year. The locations and mean TN discharge of point-sources are shown in Appendix E and total annual loadings for each basin are shown in Figure

2. The largest sources are waste water treatment plants in the up-stream Raleigh-Durham, Greensboro and Fayetteville metropolitan areas. In general TN loadings from point-source dischargers have been decreasing since the early 1990s, though there has been an increase in TPRB since the mid-2000s (Figure 2).

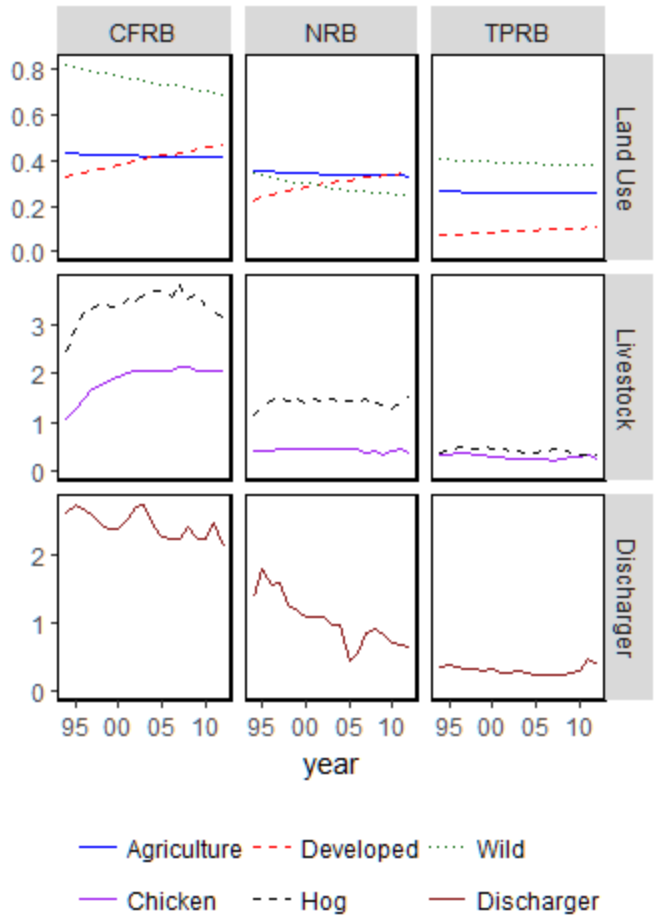

Figure 2. Nitrogen source data for each river basin (1994-2012). Included are the area of distributed sources, Agriculture, Developed, and Wild ($10^6\ ha$), chicken (10^8) and hog (10^6) counts, and TN loadings from point source dischargers ($10^6\ \frac{kg}{yr}$).

County-level livestock operation estimates were derived from United States Department of Agriculture (USDA) chicken (broilers and layers) and hog quinquennial census and annual survey counts (USDA-NASS, 2016). Most hogs and chickens are raised in CAFOs in NC (Burkholder et al., 2007) and are thus reflective of the size of CAFOs. Annual survey estimates for hog inventory are available for the period of this study, 1994- 2012, whereas

annual estimates for broilers and layers are only available from 2006-2012. Layer and broiler production counts for years preceding 2006 were estimated using 5-year census data with linear interpolation to estimate in-between years. Livestock densities were distributed across incremental watersheds by area-weighting. County level counts are the best available source of livestock data, since comprehensive CAFO records are only available after 2003 (J.R. Joshi, NC DEQ, personal communication, 2017). Nitrogen content of both chicken and hog waste was calculated using the 2017 North Carolina Agricultural Chemical Manual estimates of manure volume produced per animal per year and nitrogen content per volume. Broilers, layers and hogs were estimated to produce 0.19, 0.52, and 6.3 kg/animal/yr of nitrogen respectively (NC State University, 2016). In general, there was a dramatic increase in livestock production in the CFRB in the 1990s, with a leveling off in the 2000s (Figure 2).

Annual precipitation estimates were included within the model to assess the effect of rainfall on nutrient delivery. Annual precipitation estimates were obtained from the NC Climate Retrieval and Observation Network of the Southeast Database (CRONOS) (State Climate Office of North Carolina, 2016). CRONOS data were obtained for 143 weather stations for the years 1994-2012. Surface rasters of precipitation for each year across all three river basins were created using inverse distance weighting, and spatial averages were obtained for each incremental watershed.

Nitrogen Retention

Nitrogen retention due to settling and denitrification was integrated into this model for both streams and water bodies (lakes and reservoirs). River-reach and water body

characteristics were obtained from the National Hydrography Dataset (USGS 2013) acquired from the EPA's Better Assessment Science Integrating point & Non-point Sources (BASINS) GIS application (US EPA, 2015). NHD is a compilation of geospatial hydrologic datasets and includes stream reach characteristics such as mean flow, velocity, area, and depth. Nitrogen retention was estimated as a function of residence time, mean depth, and mass transfer coefficient for each stream, and hydraulic loading rate (ratio of flow to surface area) and mass transfer coefficient for each water body. This relationship was proposed by Kelly et al. (1987) for lakes, was adapted to streams by Howarth et al. (1996), and was expressed as first-order exponential decay by Birgand et al. (2007). The mass transfer coefficient is the water column height from which nitrate is removed per unit of time, calculated from the ratio of areal denitrification to nitrate concentration (Kelly et al., 1987; Howarth et al., 1996; Birgand et al., 2007). Nitrogen retention (r) is calculated by the following equation:

$$r = 1 - e^{\frac{-\rho}{K}} \tag{4}$$

where ρ is the mass transfer coefficient (m/yr) for streams (ρ_s) or water-bodies (ρ_{wb}), and K is the depth to residence-time ratio for streams (m/yr), and hydraulic loading rate for water-bodies (m/yr). Nitrogen retention in streams is estimated for point and distributed sources within a watershed along the stream path from their location (the outlet of a HUC 12 subbasin for distributed sources) to the incremental watershed outlet. To determine the effective K for a given source stream path, the depths of stream segments along the stream path were weight-averaged by residence time, and this weighted depth was then divided by the cumulative

residence time. Retention is calculated individually for each water body intersected by the stream path from a source location. A single (overall) retention value for each source location within a watershed is calculated as follows:

$$r_{i,x} = 1 - (e^{\frac{-\rho_s}{K_{i,x,s}}} * \prod_{w} e^{\frac{-\rho_{wb}}{K_{i,x,w}}}) \tag{5}$$

where $r_{i,x}$ is the retention of source *x* in watershed *i*, $K_{i,x,s}$ is the aggregated *K* value for the stream path from source *x*, and $K_{i,x,w}$ is the *K* value for each waterbody (*w)* that is intersected by the stream path of source *x*.

Bayesian Model Formulation

The model formulation used in this study includes both deterministic and stochastic components:

$$y_{i,t} = \hat{y}_{i,t} + \alpha_i + \varepsilon_{i,t} \tag{6}$$

where $y_{i,t}$ is the incremental loading (kg/yr) for incremental watershed *i* and year *t*, $\hat{y}_{i,t}$ is the deterministic prediction of incremental nitrogen loading (kg/yr), α_i is a normally distributed watershed-level "random effect" (Gelman et al., 2014), and $\varepsilon_{i,t}$ is a log-normally distributed residual error term. The random effect α_i represents systematic variability among watersheds, whereas the $\varepsilon_{i,t}$ term is the predictive error typical of regression models.

The WRTDS incremental loading estimate is related to the unknown actual incremental loading ($y_{i,t}$), through a normal distribution:

$$\tilde{y}_{i,t} \sim N\left(y_{i,t}, \tilde{\sigma}_{i,t}\right) \tag{7}$$

where $\tilde{y}_{i,t}$ is the WRTDS incremental loading estimate and $\tilde{\sigma}_{i,t}$ is its standard deviation (calculated from equations 3). A normal distribution is used here because incremental loadings may be negative if the retention of upstream loadings exceeds the loading contribution of sources within the incremental watershed. The log-transformed actual incremental loading estimate is normally distributed and centered on the log-transformed deterministic prediction plus the random intercept term:

$$L(y_{i,t}) \sim N\left(L(\hat{y}_{i,t} + \alpha_i), \sigma_\epsilon\right) \tag{8}$$

where *L(y)* is the natural log of $y + 10^6$ (kg/yr), and σ_ϵ is the residual standard deviation. The log transformation is performed to address the non-normality of loading residuals on the original scale (Schwarz et al., 2006). The 10^6 kg offset accounts for the possibility of negative incremental loadings (as noted above), and is a common practice when applying power transformations (Faraway, 2016). The loading estimate $\hat{y}_{i,t}$ is determined by summing watershed loading contributions from point sources, distributed sources (agriculture, developed, and wild), and CAFOs, and subtracting instream losses associated with the upstream load:

$$\hat{y}_{i,t} = s_{i,t,a} + s_{i,t,d} + s_{i,t,w} + s_{i,t,h} + s_{i,t,c} + s_{i,t,p} - z_{i,t} * r_{i,z} \quad (9)$$

where $s_{i,t,a}$, $s_{i,t,d}$, $s_{i,t,w}$, $s_{i,t,h}$, $s_{i,t,c}$, and $s_{i,t,p}$ are the watershed- and year-specific source contributions of agriculture, urban development, wildlands, hogs, chickens, and point sources, respectively (after accounting for in-stream losses); and $z_{i,t}$ and $r_{i,z}$ are the upstream WRTDS loading estimate and associated retention within the incremental watershed. The source specific contributions are calculated as:

$$s_{i,t,x} = \beta_x\left(1 + \beta_{p,x} * p_{i,t}\right) * \boldsymbol{w}_{\boldsymbol{i,t,x}}^{\boldsymbol{T}} * \left(1 - \boldsymbol{r}_{\boldsymbol{i,x}}\right) \quad (10)$$

where $s_{i,t,x}$ is the contribution of a given source type (x = {a, d, w, h, c, p}) in kg/yr, β_x is the source's export (EC) or delivery coefficient (DC), $\beta_{p,x}$ is the source's precipitation impact coefficient (PIC), and $p_{i,t}$ is the normalized watershed- and year-specific precipitation. The transposed vector $\boldsymbol{w}_{\boldsymbol{i,t,x}}^{\boldsymbol{T}}$ includes the area (ha) of land-use types and loadings (kg/yr) of CAFO and discharger types from different locations around the incremental watershed, and the vector $\boldsymbol{r}_{i,x}$ is the retention losses associated with these different locations. ECs are in units of kg/ha/yr while DCs are dimensionless, representing the percentage of nitrogen that is delivered to streams.

Both the EC and DC terms represent baseline nitrogen contribution rates under mean annual precipitation (when $p_{i,t}$ is zero). Because precipitation is centered at zero, the PIC term β_p can have either a negative or positive impact on the baseline EC. While PICs differ by

source type, they are hierarchically related to each other by a common distribution with standard deviation, σ_p. The assumption is that the intensity of runoff related nutrient transport is dependent on surface characteristics such as imperviousness, drainage practices, and riparian mitigation, which can vary by land-use type. The PIC term for point sources is set to 0 since year-to-year variability in point sources is already accounted for in the point source loading dataset.

Prior Distributions

Each parameter is assigned a prior distribution within the model (Table 1). Vague uniform priors were used for parameters for which there is no known prior information. Priors for agriculture, developed, and wild ECs were obtained from a compilation of over 70 studies conducted nationally, and previously used in a watershed planning study of the Albemarle-Pamlico Estuary in NC (Dodd et al., 1992). Priors for both developed and agriculture land-use ECs have high standard deviations, given the diversity of storm-water management practices, crop types and hydrological factors that influence nitrogen loading rates. A log-normal distribution for the mass transfer coefficient for streams, ρ_s, was determined from quantile values given from a study of over 70 streams across the U.S. using nitrogen stable isotropic tracers to determine denitrification rates (Mulholland et al., 2008; Alexander et al., 2009). The distribution for the mass transfer coefficient for water bodies, ρ_{wb}, was determined from studies of 35 temperate lakes (Harrison et al., 2009). All EC, DC, and PIC parameters were given a lower bound of zero, since negative values are mechanistically implausible.

Table 1. Model parameters with their prior distributions (or hierarchical distributions in the case of PIC parameters).

Parameter	Description	Units	Prior Distribution	Source
$\beta_{e,a}$	Agriculture EC	kg/ha/yr	N(9,7)	Dodd, et al., 1992
$\beta_{e,d}$	Developed EC	kg/ha/yr	N(8,3)	Dodd, et al., 1992
$\beta_{e,w}$	Wild EC	kg/ha/yr	N(2,2)	Dodd, et al., 1992
$\beta_{d,c}$	Chickens DC	-	U(0,1)	—
$\beta_{d,h}$	Hogs DC	-	U(0,1)	—
$\beta_{d,p}$	Point Source DC	-	N(1,0.1)	—
$\beta_{p,a}$	Agriculture PIC	-	N(0,σ_p)	—
$\beta_{p,d}$	Developed PIC	-	N(0,σ_p)	—
$\beta_{p,w}$	Wild PIC	-	N(0,σ_p)	—
$\beta_{p,c}$	Chickens PIC	-	N(0,σ_p)	—
$\beta_{p,h}$	Hogs PIC	-	N(0,σ_p)	—
ρ_s	Coef. Streams	m/yr	LogN(3,1)	Mulholland, et al., 2008
ρ_{wb}	Coef. w.b.	m/yr	N(5,5)	Harrison, et al., 2009
σ_{res}	Residual s.d.	kg/yr	U(0,5$x10^6$)	—
σ_α	Random int. s.d.	kg/yr	U(0,5$x10^6$)	—
σ_p	PIC s.d.	-	U(0,1)	—

Model parameters were estimated using Hamiltonian (Markov Chain) Monte Carlo (HMC) sampling with the Rstan software operated in R (R. Core Team, 2013; Stan Development Team, 2016). Three parallel sampling chains were run 20,000 iterations each with the first 10,000 discarded as the burn-in period, resulting in 30,000 posterior samples total. Convergence was considered achieved when the scale reduction statistic $\hat{R}$—the square root of the ratio of total variance to within-chain variance of the posterior distributions—was approximately equal to one (Gelman & Rubin, 1992).

Performance Assessment

Model performance was determined by the Nash-Sutcliffe Efficiency (NSE) (Nash & Sutcliffe, 1970) and mean absolute error (MAE). NSE, which is commonly used in hydrologic modeling, is equivalent to the coefficient of determination as it is often defined in statistic texts (e.g., Faraway, 2016), but different from the square of the Pearson's correlation coefficient. NSE and MAE were calculated based on the means of the Bayesian posterior predictive distributions. For comparison, these performance metrics were calculated in two different ways – assuming the incremental watershed-specific random effects are known, and assuming they are unknown (set to zero).

The performance of the full model (calibrated to all data) is compared to a null-source model, and is further evaluated through a 3-fold cross-validation (Elsner & Schmertmann, 1994) where each of the three river basins are treated as a fold. The null-source model is a linear regression where watershed area and precipitation multiplied by watershed area were used as predictor variables of incremental loads within watersheds. The null-source model serves as a benchmark to gage the extent to which representation of specific source types (i.e., land uses, point sources, and CAFO sources) and their spatiotemporal variability improves model performance. For cross-validation, the data were divided into three sections (i.e., folds) by river-basin, where the model is trained by two and tested by one of the three segments, considering all three possible combinations in turn. The cross-validation serves to test the model's predictive skill, demonstrated by the NSE and MAE, and by comparing parameter estimates from the validation folds to the full model. A robust model should have similar parameter estimates regardless of which regions are used for training or testing.

RESULTS

Annual TN Loads

Loading estimates from the WRTDS analysis suggest an increase in TN entering NC estuaries since the late 1990s, particularly in the CFRB. Annual and flow-normalized loading estimates for each river basin are shown in Figure 3. Flow-normalized loadings are determined by taking the mean of loading estimates calculated from all historical flows for each particular day, and are useful in analyzing long-term loading trends in that they eliminate short-term variability in flow rates caused by droughts or wet weather (Hirsch et al., 2010; Lebo et al., 2012). Flow normalized loadings suggest there has been a steady increase in TN loadings in the CFRB since the mid-1990s. In the NRB and TPRB, TN loadings declined until around 2000 and then began to increase. Loading estimates at individual stations along the Neuse suggest larger contributions from the mostly agricultural watersheds of Goldsboro to Ft. Barnwell than the urban watersheds of Falls Dam and Clayton (Appendix G). The mean annual incremental loading (loading produced within each incremental watershed) is approximately 680,000 kg/yr, with a standard deviation of 450,000 kg/yr TN. Mean annual incremental loading and standard deviation normalized by watershed area are 4.15 kg/ha/yr and 3.07 kg/ha/yr, respectively.

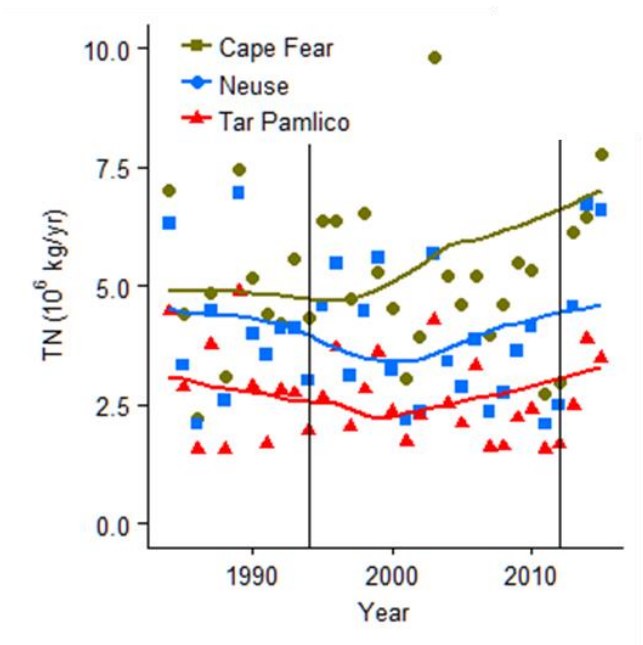

Figure 3. WRTDS estimates of TN loadings at downstream locations on the CFRB, NRB, and TPRB (Kelly, Ft. Barnwell, and Washington, respectively). Points represent annual loading estimates and lines are flow-normalized loads. Vertical lines mark the beginning and end of the time-series used in the watershed model (1994-2012).

Model Parameterization and Performance

Model parameters that characterize nutrient sources, nutrient retention, and error distributions were determined through Bayesian inference (Table 2). Marginal posterior distributions of model parameters are given as density plots along with their prior distributions in Figure 4. For EC (β_e) and DC (β_d) parameters, the posterior distributions appear more influenced by the data than the vaguely distributed priors (Figure 4). The probability of a given parameter exceeding another parameter is determined by the frequency of exceedance among posterior samples (Appendix B). There is an 87% and 91% probability that the agriculture EC ($\beta_{e,a}$) is greater than the ECs for developed and wild, respectively; but only a 53% probability

that the developed EC is greater than the wild EC. In terms of magnitude, our results suggest that agricultural EC is 2.0 and 1.8 times higher than wild and developed ECs, respectively (Table 2). The mean DC value for hogs is very similar to that of chickens. Mean PIC values are largest for agricultural land and smallest for developed land and chickens, suggesting that nutrient export from agriculture lands is most heavily influenced by rainfall rates, while developed land and chicken operations export a more constant rate of TN regardless of precipitation. While the credible intervals (CIs) for PICs are wide and substantially overlap, there is an 80% probability that the PIC for agriculture land is greater than the PIC for developed land (Appendix B).

Table 2. Mean parameter estimates with 95% credible intervals for models with and without informative priors on EC and DC. Mean parameter estimates are also shown for each fold of the cross validation. Parameter units are provided in Table 1.

	Full model with all priors		Full model without EC & DC priors		Validation TP & N	Validation N & CF	Validation CF & TP
Symbol	Mean	95% CI	Mean	95% CI	Mean	Mean	Mean
$\beta_{e,a}$	4.9	2.3-7.7	5.0	2.0-8.4	5.0	4.3	5.8
$\beta_{e,d}$	2.7	0.47-5.0	2.6	0.11-5.6	3.6	2.8	2.5
$\beta_{e,w}$	2.5	0.92-4.3	3.0	1.1-5.2	3.8	2.5	1.9
$\beta_{d,c}$	0.028	0.010-0.053	0.026	0.0-0.053	0.029	0.031	0.032
$\beta_{d,h}$	0.025	0.0-0.050	0.023	-0.008-0.056	0.053	0.025	0.022
$\beta_{d,p}$	0.87	0.68-1.1	0.70	0.39-1.0	0.96	0.89	0.87
$\beta_{p,a}$	0.43	0.21-0.79	0.43	0.21-0.83	0.31	0.46	0.45
$\beta_{p,d}$	0.25	0.015-0.67	0.24	0.01-0.72	0.27	0.25	0.24
$\beta_{p,w}$	0.36	0.071-0.77	0.32	0.08-0.69	0.40	0.39	0.31
$\beta_{p,c}$	0.23	0.015-0.64	0.25	0.011-0.73	0.31	0.22	0.28
$\beta_{p,h}$	0.36	0.025-1.1	0.35	0.022-0.99	0.43	0.34	0.34
ρ_s	4.6	1.3-9.9	5.0	1.2-11	11	4.4	4.0
ρ_{wb}	11	7.9-14	11	7.5-14	6.7	12	11
σ_{res}	0.096	0.084-0.11	0.095	0.085-0.11	0.083	0.11	0.096
σ_{α}	1.5	0.98-2.3	1.6	1.0-2.4	1.5	1.3	1.6
σ_p	0.49	0.21-1.2	0.49	0.20-1.2	0.55	0.52	0.51

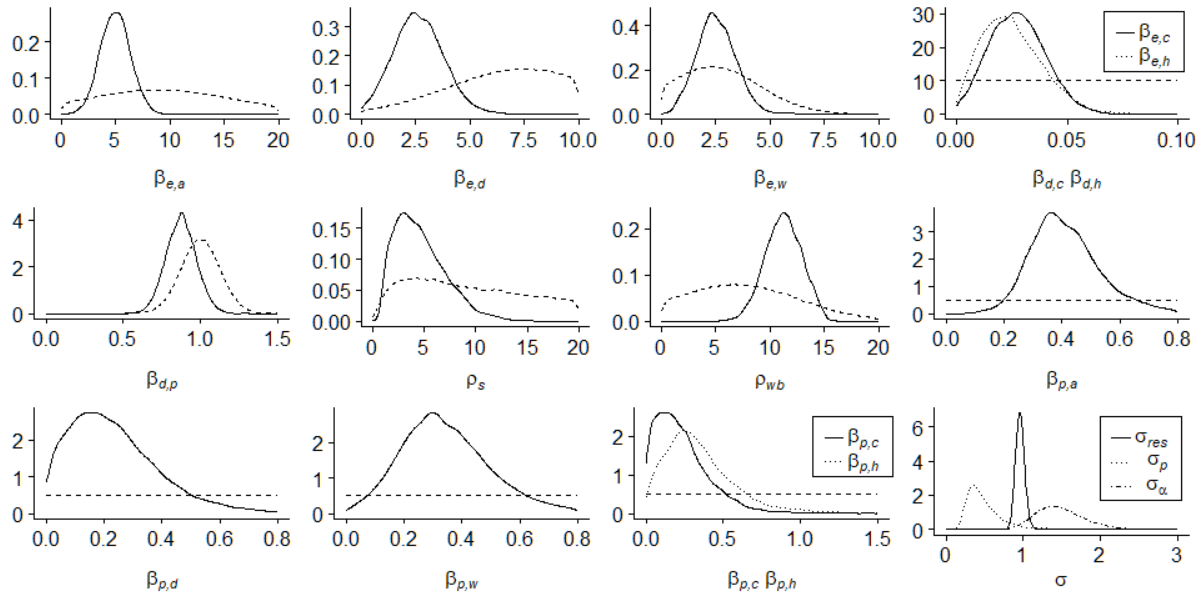

Figure 4. Marginal posterior distributions of model parameters with prior distributions (dashed lines), as determined for the full model. Parameter units are provided in Table 1.

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Nitrogen retention is determined by the mass-transfer coefficients for streams and water bodies. The mean mass transfer coefficient for in-stream retention (ρ_s) is on the lower-end of literature values while that of water body retention (ρ_{wb}) is on the upper end (as reflected by the prior distribution, Figure 4). Retention in the two largest reservoirs, Jordan Lake and Falls Lake, are 84% and 60%, respectively, which is in the higher range of retention for lakes in general (Harrison et al., 2009), though for Falls Lake within range from a previous study (Garrett, 1990). Overall, the percentages of TN retained in streams and water bodies are 13.4% and 12.2%, respectively. In total, stream and water body losses retain 24% of TN in all three basins. Basin-wide retention from HUC12 subbasins to river basin outlet are shown in Appendix H.

Hydraulic retention times for both Jordan Lake and Falls Lake are relatively long, potentially causing a lag in downstream station monitoring for upstream TN loadings that exceeds the annual time-step. Mean residence times from 1984 to 2017, calculated by dividing mean outflow by reservoir volume (USACE, 2017), for Jordan Lake and Falls Lake are 0.52 and 0.78 years respectively, with standard deviations of 0.60 and 0.35 years respectively. Both Lillington and Falls Dam watersheds, containing Jordan Lake and Falls Lake, have NSEs of -0.48 and 0.33 respectively, indicating that the large retention times in these watersheds may be contributing to poor estimations of incremental loadings. Still, these watersheds were included in the model since their mean residence times were less than a year, and for the sake of maintaining continuity of TN transport within the river basins.

The model was also calibrated using non-informative priors for EC and DC parameters, so that the influence of the prior information could be more clearly assessed. In most cases, parameter estimates without priors were similar to parameter estimates with priors (Table 2). A notable exception is the EC for hogs, where the 95% CI intersects zero slightly (though the 90% CI does not). While this "no-prior" model is not preferred because prior information from previous studies is ignored and because negative TN loadings are mechanistically impossible, the no-prior model highlights the fact that agricultural and chicken sources are the most strongly identifiable distributed and CAFO source types, respectively, in this study area.

Model skill is assessed in terms of NSE and MAE, based on model predictions with watershed random effects, with fixed effects only, and from cross validation (Figure 5). With random effects included and removed (set to zero), the full model (calibrated to all data) explains 78% and 73% of variability, respectively. Using log-transformations for predicted

and observed values, the model explains 74% and 68% of variability with random effects included and removed respectively (Appendix L). Temporal autocorrelation in residuals is minimal, with lag-1 correlation coefficients averaging 0.075 across watersheds. In cross validation (without random effects), the model explains 63% of the variability. Mean parameter estimates for all three basin-level folds of the cross validation are similar to each other and to the full model (Table 2), affirming the model is reasonably robust to variations in the training dataset. In comparison, the null-source model explained just 49% of variability using only watershed area and precipitation as predictors.

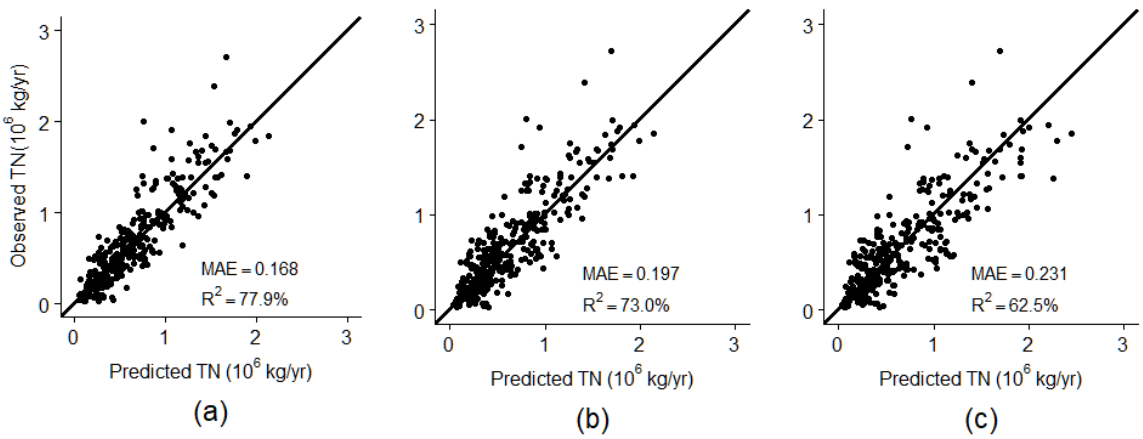

Figure 5. Observed vs. predicted plots of incremental TN loadings using (a) full model with fixed and random effects, (b) full model with fixed effects only, and (c) cross validation with fixed effects only.

Nutrient Loading Source Apportionment

Throughout the study period, from 1994 to 2012, TN export estimates reflect a decrease in point-sources, moderate changes in land-use, and a notable increase in CAFOs in many watersheds. Figure 6 shows source-specific TN contributions for of each basin, both before and after accounting for in-stream and water body retention, and using long-term mean precipitation values for each watershed. From 1994 to 2012, point-source discharges of TN decreased by 10% and 61%, in the CFRB and NRB, respectively, and increased by 1.3% in the TPRB. Developed land TN increased by nearly 50% in all three river basins coinciding with a decrease in agriculture land TN of 3.7%, 5.9%, and 3.7% and wild TN of 16%, 29%, and 6.8% in the CFRB, NRB, and TPRB, respectively. Chicken TN export increased by 97% in the CFRB and decreased slightly by 7% and 15% in the NRB and TPRB respectively. Hog TN export increased by 29% and 35% in the CFRB and NRB, respectively, and decreased by 14% in the TPRB. The largest changes in basin-wide percent composition of TN export occurred in the NRB, where in 1994, point, land-use, and CAFO sources made up 26%, 9%, and 65% of basin-wide TN export, respectively; whereas in 2012, the same sources made up 9%, 12%, and 79%, respectively. For the CFRB, livestock sources increased from 12% to 20% and point and land-use sources decreased from 28% to 24% and 60% to 56%, respectively, of the total TN export composition from 1994 to 2012. Percent composition of source contributions changed relatively little for the TPRB.

In-stream and water body retention reduces TN loadings before they reach the downstream station of each river basin (Figure 6, DS results). In 1994, these losses reduced TN loadings by 24%, 25%, and 20%, and changed slightly in 2012 to 22%, 26%, and 19% in

the CFRB, NRB, and TPRB, respectively. TN loading contributions are largest in downstream regions, characterized by higher annual precipitation and lower retention rates (fewer reservoirs), and where most livestock operations and agricultural land is situated (Figure 7). The largest increases in TN loading rates from 1994 to 2012 are in the downstream CFRB because of the growth of chicken and hog operations. Upstream decreases in loading rates are attributable to decreases in point-source loads in the CFRB and NRB and decreases in agriculture land and livestock production in the TPRB. Precipitation has a larger effect on TN loadings in downstream agricultural areas, given that the PIC for agriculture is higher than that of developed and wild land (Appendix I).

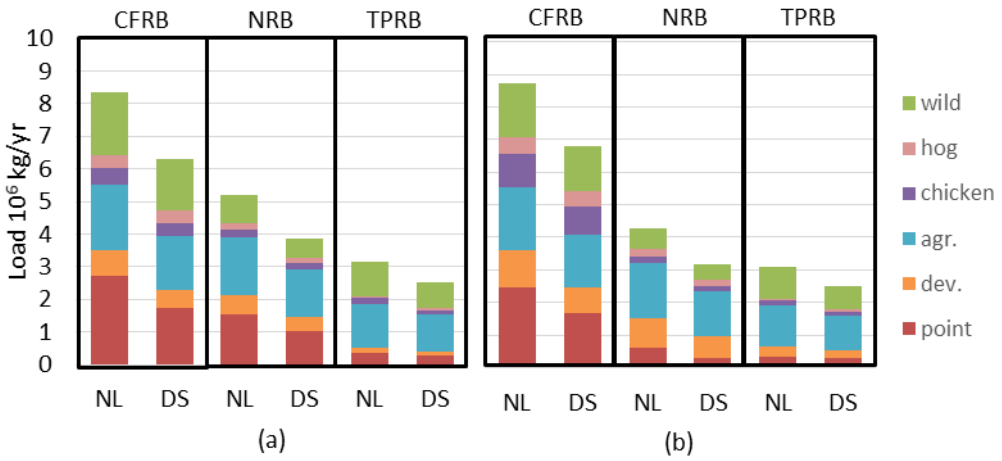

Figure 6. TN loading contributions in the CFRB, NRB, and TPRB for (a) 1994 and (b) 2012, by source type. Mean precipitation values are used in calculating loads. Shown are total loadings entering the stream network, including no stream and water body losses (NL), and loadings at the downstream (DS) basin outlet accounting for stream and water body losses. Downstream locations for each basin are Kelly (CFRB), Ft. Barnwell (NRB), and Washington (TPRM). Chinquapin and Tomahawk watersheds are also included in the CFRB loading calculations.

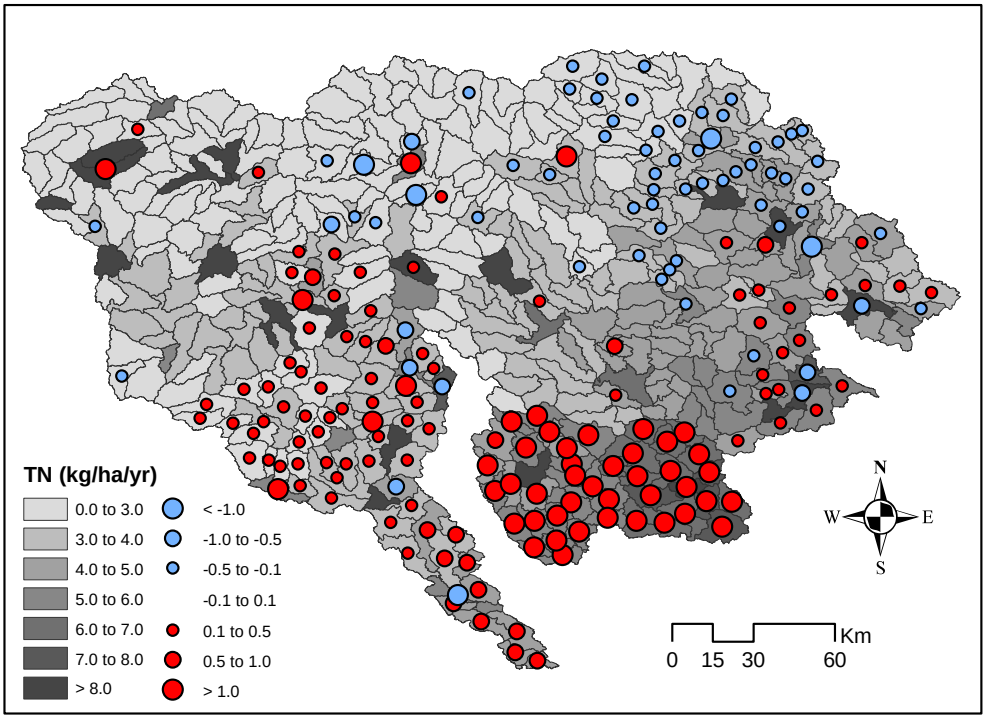

Figure 7. TN loading rates for the year 2002 (grey shading) and change in loading rate from 1994 to 2012 (points) for each HUC 12. Stream and reservoir retention is included and rates reflect loadings to downstream locations for each basin (Kelly for CFRB, Ft. Barnwell for NRB, and Washington for TPRB). HUC 12 mean precipitation values are used in calculating loads.

DISCUSSION

In this study, we introduce a new approach for probabilistically estimating nutrient export rates and predicting nutrient loadings over time. The approach builds on hybrid empirical/process-based watershed models where nutrient loading, retention, and transport are represented within a nonlinear regression framework (Smith et al., 1997; McMahon et al., 2003; Qian et al., 2005). However, while most previous applications of hybrid watershed modeling have been developed for particular reference years, our approach leverages interannual variability in nutrient sources and precipitation over a multi-decadal (*19*-year) study period. The temporal variability considered in this work provides richer information to characterize nutrient export rates and explore the drivers of interannual variability. Here, source-specific export and delivery coefficients (ECs and DCs) are modified based on annual precipitation anomalies (i.e., deviations from mean precipitation). This approach reflects the expectation that precipitation will increase surface wash-off and groundwater fluxes of nutrients to streams (e.g., Haith & Shoemaker, 1987). The extent to which precipitation anomalies affect nutrient loading is determined by hierarchically-related, source-specific PICs. The interannual variability in nutrient loading is, of course, further controlled by changes in source distributions over time. Overall, the deterministic component of our model accounts for 73% of the spatiotemporal variability in incremental watershed loadings (78% if watershed random effects are included, Figure 3). If, for example, variability due to precipitation is removed from the model (i.e., PICs are set to zero), then the model only explains 58% of the

loading variability. If land use variations are removed, then only 67% of the variability is explained.

While this is perhaps the first study to incorporate temporal changes in both precipitation and source distributions in a nonlinear regression model for watershed loading, similar modeling studies have explored temporal variability using alternative approaches. Wellen et al. (2012) added both interannual climatic variables and dynamic parameter estimation to SPARROW (Smith et al., 1997), thus treating climatic and watershed processes as primary agents of interannual variability. However, unlike our approach, this configuration doesn't account for land-use change, and precipitation is included as an additive component to the nonlinear regression rather than as a source-specific modifier of TN export (Wellen et al., 2012). Xia et al (2016) also considered dynamic parameter estimation and land-use change in a nonlinear regression formulation to account for interannual variability in TN export, but did not consider precipitation directly (Xia et al., 2016). Less mechanistic, linear regression models have also been used to study temporal variability in watershed TN loading. For example, Sinha and Michalak (2016) found inter-annual variability in large-scale watershed TN loadings to be driven primarily by both annual precipitation and extreme precipitation events. The effects of precipitation timing and extreme events could also be considered within context of nonlinear regression modeling, perhaps as a future enhancement to the model developed here.

In this study, we quantify TN export rates from several management-relevant source types. For the first time, using the hybrid modeling approach, we explicitly include CAFO-related livestock counts as potential sources, and empirically estimate the fraction of livestock

waste TN delivered to streams. Posterior estimates suggest that hog and chicken DCs are very similar and both less than 5% (Figure 4). A 2002 EPA study of CAFO waste management practices across the nation found a mean edge-of-field delivery loss rate of 6% and 9% for hogs and chickens, respectively (Whitman et al., 2002), which are somewhat higher than the DCs determined here. A possible reason for the lower estimates in this study is that some of the livestock contribution may be reflected in the agricultural land EC. To the extent that chicken and hog waste replace other types of fertilizer, these wastes do not constitute an additional load. Furthermore, our DCs do not represent edge-of-field delivery, but rather delivery to the HUC-12 outlet (Figure 7). In any case, our results indicate that chicken and hogs are substantial TN contributors (Figure 6), with mean ECs (calculated by dividing the mean livestock waste delivered to streams by total agriculture land area) of 1.3 and 0.82 kg/ha/yr, respectively. Adding these livestock ECs to the EC for agricultural land (4.9 kg/ha/yr) gives a total effective agricultural EC of 7.0 kg/ha/yr, which is near previous estimates for agricultural land in this region (McMahon et al., 2003; Qian et al., 2005) and the mean literature value (Dodd et al., 1992; Lin, 2004; Harmel et al., 2006).

Non-agricultural TN sources, including point source dischargers, urban development, and wildland sources are also included in the model. The wild land EC for this study is similar to mean EC values in the literature (Dodd et al., 1992; Beaulac & Reckhow, 1982). The point source DC has a mean of 0.87 and a CI that includes 1.0. A value of near 1.0 is expected for point sources, assuming that there are no substantial biases in the point source loading data (Schwarz et al., 2006). We note that point source DCs below 1.0 are common in many hybrid model applications, including previous applications in this study area (Smith et al., 1997;

McMahon et al., 2003; Qian et al., 2005), and may suggest over-reporting of discharger loading rates (Wellen et al., 2012; Schwarz et al., 2006). Developed land EC is on the lower end of literature values for mixed-intensity urban development. Our urban EC estimates are more typical of low-density development (Beaulac & Reckhow, 1982), which comprises the largest portion of developed land-use in the three river basins (Appendix A). The mean EC for point sources (calculated by dividing the mean annual point-source TN delivered to streams by total developed land area) is 5.0 kg/ha/yr, which added to developed land EC gives an effective total developed EC of 7.7 kg/ha/yr, somewhat higher than the effective total agricultural EC (above).

In this study, interannual variability in TN export and delivery rates is controlled directly by annual precipitation and source-specific PICs (Appendix C). The PIC represents the fractional change in TN export corresponding to one standard deviation change in precipitation, relative to the mean precipitation. The PIC estimate is highest for agricultural land (0.43) and lowest for developed land and chicken operations (0.25 and 0.23, respectively). The lower PIC for developed land suggests that TN export from developed areas is less responsive to precipitation on an annual time scale, consistent with the finding of Shields, et al. (2008) that urban areas export most TN during small precipitation events, which are prevalent in all years. Because of the differing PICs, there is substantial interannual variability in the relative magnitude of different source types. During a high precipitation year (95th percentile) agriculture land TN export rates are nearly 2.3 and 2.1 times higher than that of developed and wild lands, respectively, whereas during the low precipitation year (5th

percentile), agriculture land TN export is nearly equal to that of wild land, and developed land export becomes 1.8 times higher than agriculture.

Because not all spatial variation is explained by the deterministic component of this model, watershed random effects account for much of the remaining spatial variability. The random effects term can also indicate spatial correlation among watersheds that might result from regional differences in export rates and watershed processes (e.g. Coastal Plain versus Piedmont). While a previous NC study using data at higher spatial resolution (McMahon et al., 2003; Qian et al., 2005) suggested spatial correlation, the random effects in this study do not exhibit substantial spatial correlation (Appendix J). Systematic differences in nutrient export rates between Piedmont and Coastal Plain watersheds, as suggested in a previous Chesapeake-area study (Jordan et al., 1997), are also not apparent based on the random effects of our study (Appendix I).

Our results show a general decline in point-source, wild, and agricultural land TN contributions, and an increase in livestock and developed land TN contributions in the NRB and CFRB, and relatively little change in the TPRB. Regression analysis of land use practices between 1992 and 2001 by Rothenberger et al. (2009) show a strong correlation between Neuse River TN loading and urban land and CAFO expansion in the NRB, consistent with our results. Alameddine et al. (2011) used a Bayesian changepoint-threshold model, for 1979 to 2008, to determine a change in the flow-concentration relationship, indicating a regime-change in 1999 from a point-source to agriculturally dominated system. Lebo et al (2012) noted a nearly 15% increase and 15% decrease in ON and nitrate loads, respectively, into the NRB from 1995 to 2008, a trend that is concurrent with the decline in point-source TN and increase in TN loadings

from animal waste found in our study. A study by Osburn et al. (2016) found anthropogenic ON in the Neuse to derive mostly from chicken litter, urban street runoff, and eroded sediments either from urban or agricultural sources. Hog waste, however, was found to contribute little ON, though some of this waste could be converted to and be present as soil-derived ON (Osburn et al., 2016). The results of our study generally suggest similar overall contributions from hogs and chickens in the NRB (Figure 6).

Annual effective EC estimates for total agriculture (sum of chicken, hog, and agriculture land ECs) and total urban (sum of point source and developed land ECs) show declining urban and increasing agricultural TN export rates, on average, across the study area (Appendix M). Total urban EC decreased from 10 to 6.4 kg/ha/yr, while total agricultural EC increased from 6.8 to 7.2 kg/ha/yr from 1994 to 2012 during a mean precipitation scenario (Appendix M). The decrease in the total urban export rate results from a decrease in point sources loadings concurrent with an increase in developed area. The increase in TN export rate for total agriculture is attributable to the increase in CAFOs. If CAFOs were not included in the model, the total agricultural EC would remain constant, and there would be no mechanism for exploring the implications of CAFO development within the study region.

Results can be analyzed in terms of total annual export (kg/yr) in addition to areal export rates (kg/ha/yr). Annual TN export from total agricultural and total developed sources varies by basin and by precipitation scenario. The TPRB experienced little change in total agricultural and developed TN export over the study period (Appendix M). Total developed TN export (kg/yr) decreases only in the NRB due to substantial reductions in point-source discharge (Appendix M). In the NRB, agricultural export is greater than developed export

under all but the lowest (5th percentile) flow conditions, where agricultural and developed export are approximately equal. This outcome highlights the effectiveness of TMDLs at reducing point sources in the NRB and the remaining challenge of reducing nonpoint sources. Total agricultural TN export increases substantially in the CFRB due mainly to CAFO contributions, while total developed land export increases slightly as a result of increasing urbanization with little reduction in point source loadings (Figure 2, Appendix M).

This study effectively incorporates temporal variability in sources and precipitation data into a hybrid non-linear regression model for determining interannual variability in watershed loads. The inclusion of precipitation as a modifier of source-specific export rates can be useful in assessing environmental impacts of nutrient loadings during different land-use and precipitation scenarios. The Bayesian-hierarchical modeling framework provides a platform for incorporating uncertainty in model inputs, for including watershed-level random effects that address spatial variability not otherwise accounted for by the deterministic component of the model, for including prior knowledge from the literature, and for providing probabilistic estimation of model parameters. Incorporating interannual variability within a probabilistic framework is an important development for hybrid watershed models, and can provide rich insights into the dynamics of nutrient loading within watersheds. This is especially important for informing effective water-resources management policies for mitigating nutrient loadings in developing watersheds.

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APPENDICES

Appendix A. *NWALT Land-Use Classifications*

NWALT land-use classifications with their respective areas (ha) and % of total area in all three river basins *(Falcone, 2015)*.

	Area (hectares)			% of total area		
Land Use Classification	1992	2002	2012	1992	2002	2012
Water						
Water	21548	21548	21960	1.2	1.2	1.2
Wetlands	130783	130783	130783	7.3	7.3	7.3
Developed						
Major Transportation	26197	28453	30156	1.5	1.6	1.7
Commercial/Services	30030	35111	38592	1.7	2.0	2.2
Industrial/Military	10842	12392	13884	0.6	0.7	0.8
Recreation	6997	8049	8669	0.4	0.5	0.5
Residential, High Density	14185	20450	26935	0.8	1.1	1.5
Residential, Low-Medium Density	64792	75844	90279	3.6	4.3	5.1
Developed, Other	22929	17494	12631	1.3	1.0	0.7
Semi-Developed						
Urban Interface High	25584	23495	21489	1.4	1.3	1.2
Urban Interface Low Medium	129582	217673	267704	7.3	12.2	15.0
Anthropogenic Other	599	531	448	0.0	0.0	0.0
Production						
Mining/Extraction	1031	1060	1136	0.1	0.1	0.1
Crops	335551	318468	318840	18.9	17.9	17.9
Pasture/Hay	165608	165823	156385	9.3	9.3	8.8
Grazing Potential	8403	8258	8363	0.5	0.5	0.5
Low Use						
Low Use	784342	693572	630749	44.1	39.0	35.4
Very Low Use, Conservation						
Very Low Use, Conservation	375	375	375	0.0	0.0	0.0

Appendix B. *Probabilistic Comparison of Parameter Magnitudes*

Table values represent the percent probability of row parameter exceeding column parameter.

	$\beta_{e,a}$	$\beta_{e,d}$	$\beta_{e,w}$
$\beta_{e,a}$	—	87	90
$\beta_{e,d}$	13	—	54
$\beta_{e,w}$	10	46	—

	$\beta_{d,c}$	$\beta_{d,h}$
$\beta_{d,c}$	—	57
$\beta_{d,h}$	43	—

	$\beta_{p,a}$	$\beta_{p,d}$	$\beta_{p,w}$	$\beta_{p,c}$	$\beta_{p,h}$
$\beta_{p,a}$	—	80	62	83	65
$\beta_{p,d}$	20	—	30	53	34
$\beta_{p,w}$	38	70	—	70	55
$\beta_{p,c}$	17	47	30	—	34
$\beta_{p,h}$	35	66	45	66	—

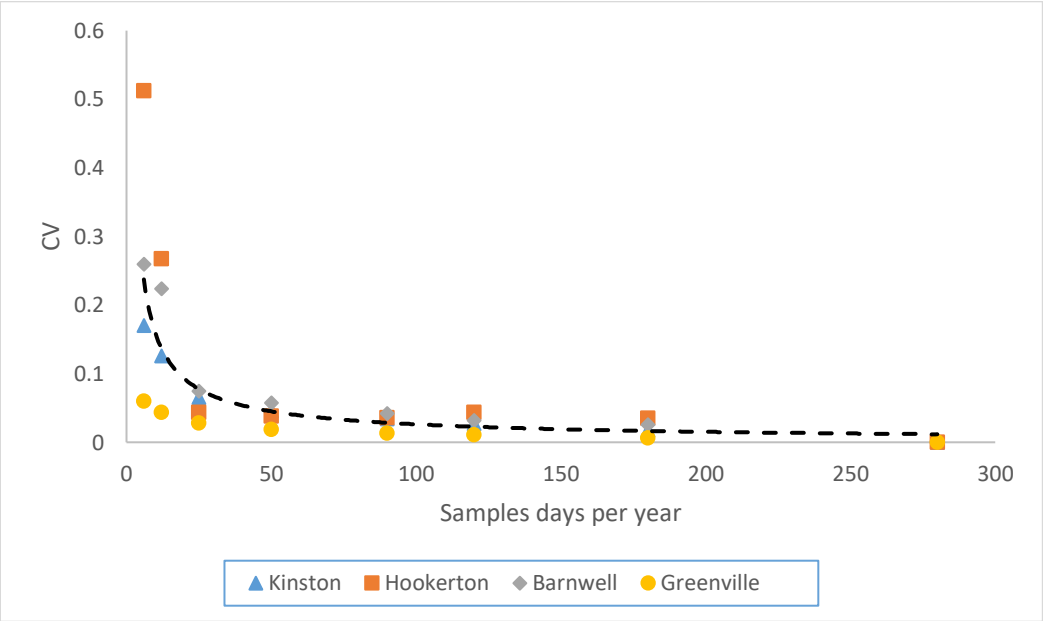
Appendix C. *Modified Export Coefficients Including the PIC for Each Source*

The table below lists the modified export coefficients, including the PIC for each source type (kg/ha/yr). Included are TN export for mean precipitation (prec. = 0) and positive and negative standard deviation of precipitation ranges (prec. = ± 1) and the 95 percentile of precipitation ranges (prec. = ± 1.96).

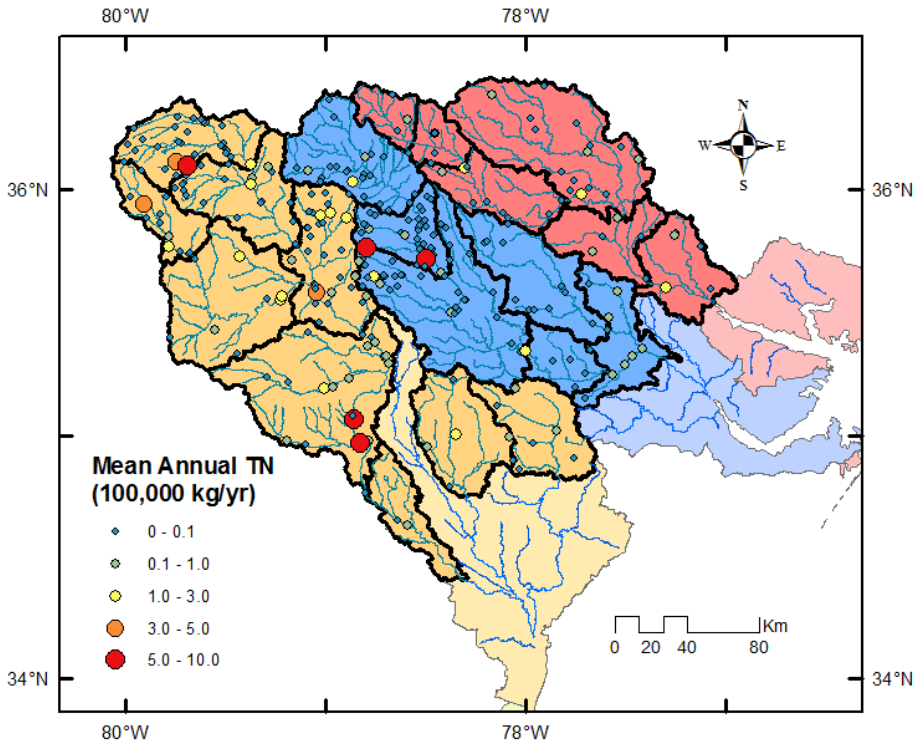
Source	prec. = 0	$prec. = \pm 1$	$prec. = \pm 1.96$
Agriculture	4.9	2.8-7.0	0.77-9.0
Developed	2.7	2.0-3.4	1.4-4.0
Wild	2.5	1.6-3.4	0.74-4.3
Chicken	0.028	0.022-0.034	0.015-0.041
Hog	0.025	0.016-0.034	0.0074-0.042

Appendix D. *Coefficient of Variation (CV) Values from the WRTDS Uncertainty Analysis*

The figure below shows the Coefficient of Variation (CV) values for the 8 sampling schedules used in the WRTDS uncertainty analysis by station location. The dashed line is the fit power regression used in this study.

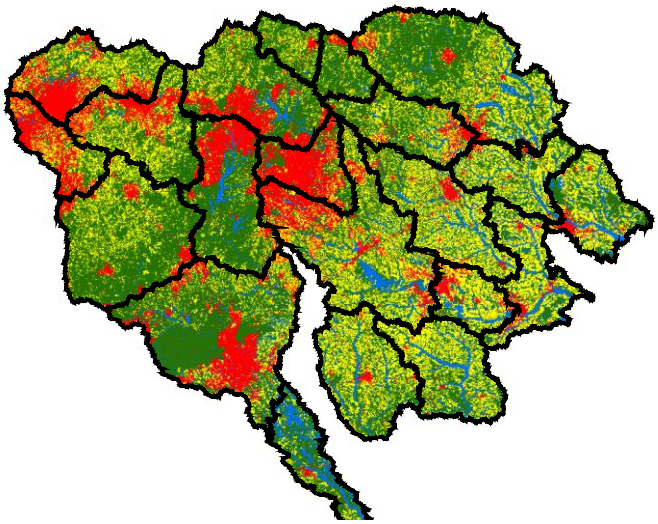


Appendix E. *Mean Annual TN Loads from Point Source Dischargers from 1994 to 2012*

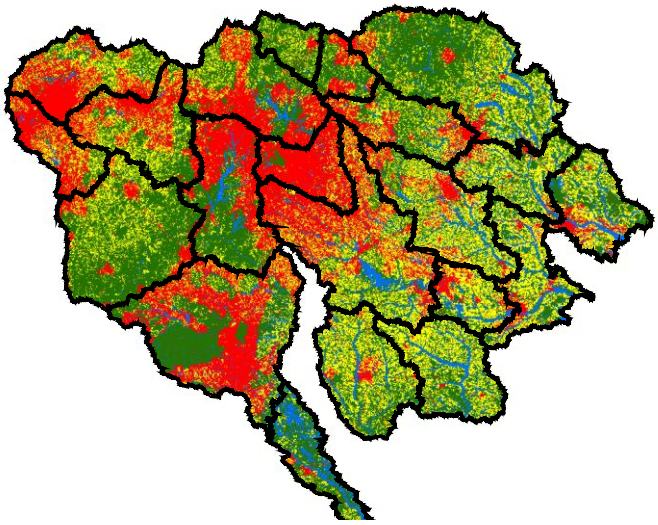


Appendix F. *Land Use Change Maps*

The figures below show land use change for 1992 (a) and 2012 (b). Land use types included are agriculture (yellow), developed (red), wild (green), and water/wetlands (blue). Black outlines are the watersheds used in this study.



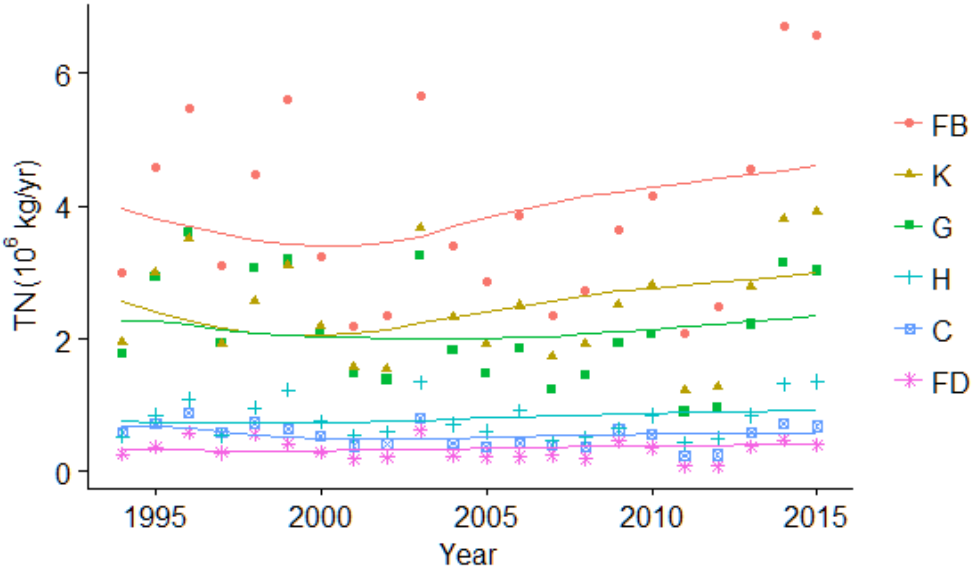
(a)



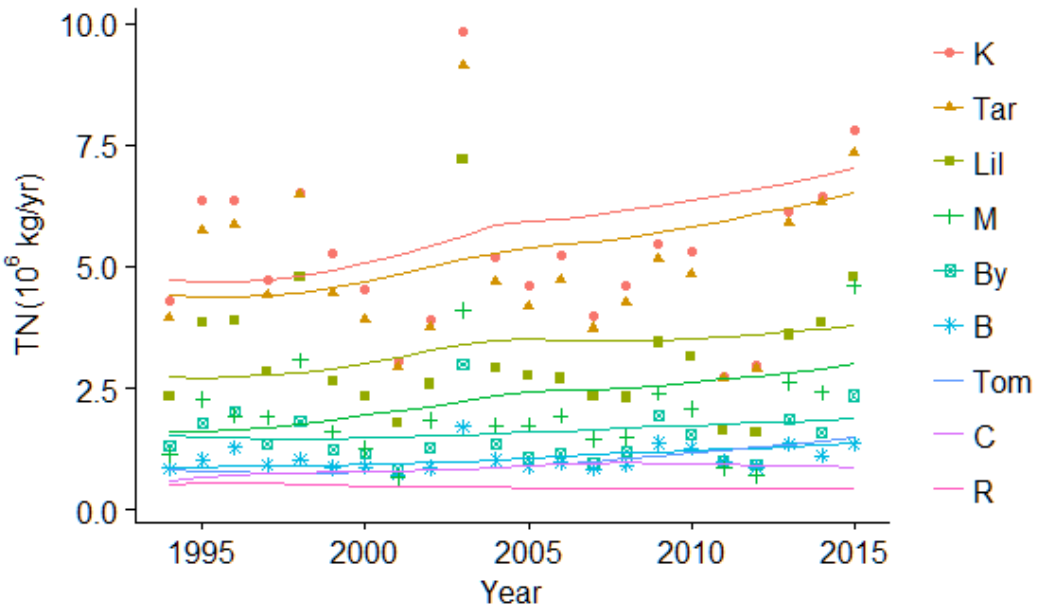
(b)

Appendix G. *WRTDS TN Estimates for Stations in Each River Basin*

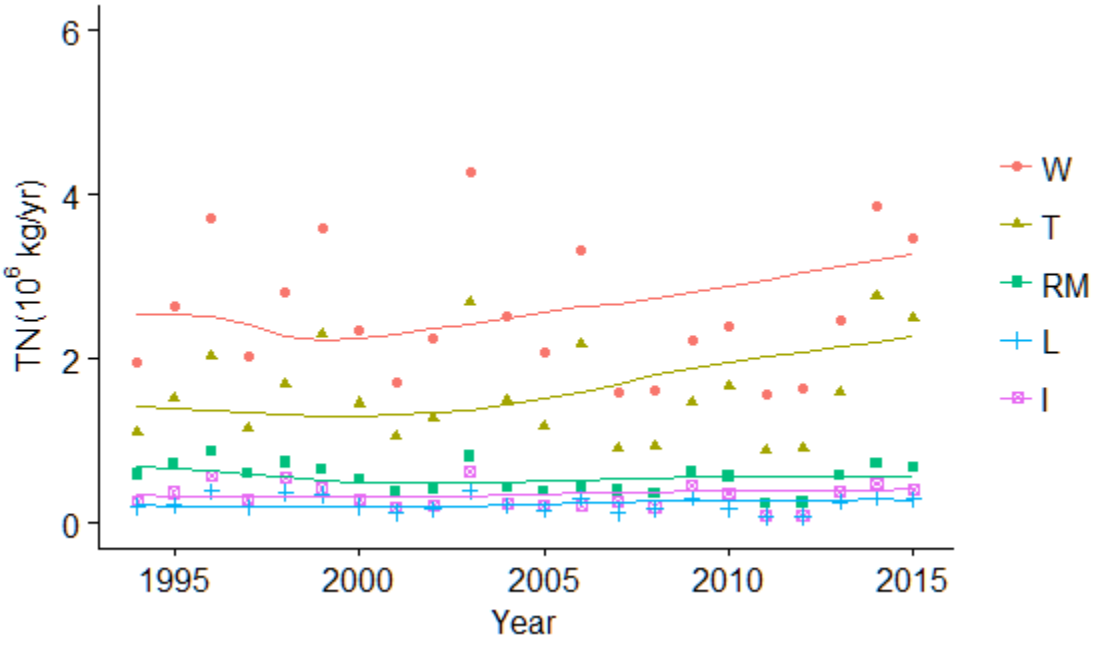
The figures below show WRTDS TN estimates for stations in the NRB (a), CFRB (b), and TPRB (c) from 1994 to 2015. Points are annual estimates and lines are flow-normalized estimates. Stations for the NRB include Falls Dam (FD), Clayton (C), Goldsboro (G), Kinston (K), and Ft. Barnwell (FB). Stations for the CFRB include Kelly (K), Tarheel (Tar), Lillington (Lil), Moncure (M), Bynum (By), Burlington (B), Tomahawk (Tom), Chinquapin (C), and Ramseur (R). Stations for the TPRB include Washington (W), Tarrboro (T), Rocky Mount (RM), Louisburg (L), and I96 (I).



(a)


10.0
7.5
5.0
2.5
0.0
TN (10^6 kg/yr)
1995
2000
2005
2010
2015
Year
K
Tar
Lil
M
By
B
Tom
C
R

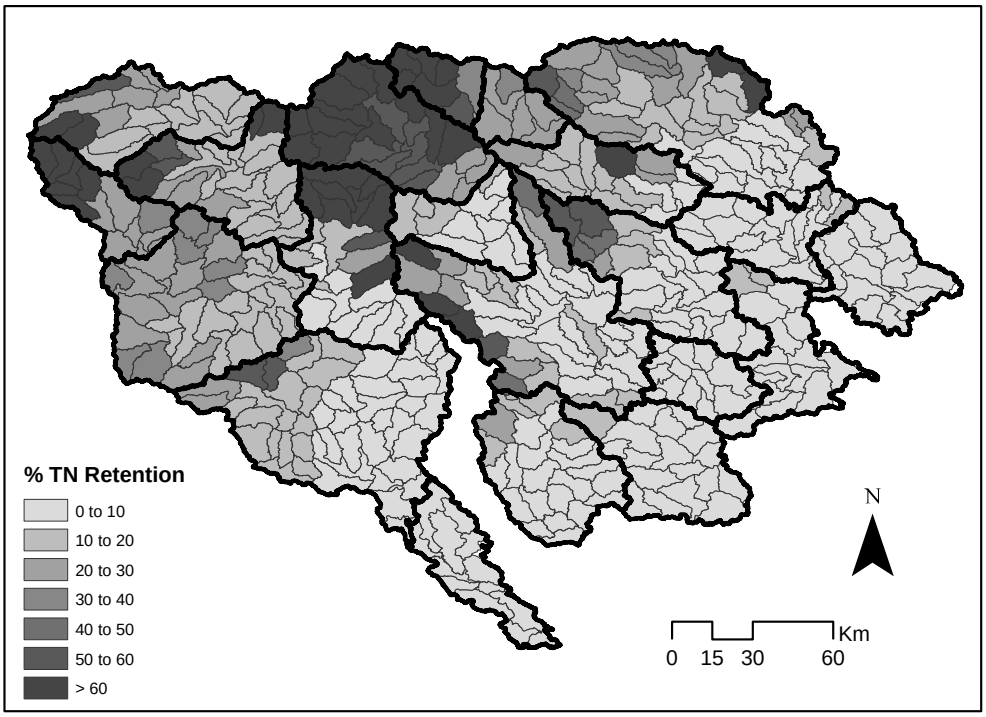
(b)


6
4
2
0
TN (10^6 kg/yr)
1995
2000
2005
2010
2015
Year
W
T
RM
L
I

(c)

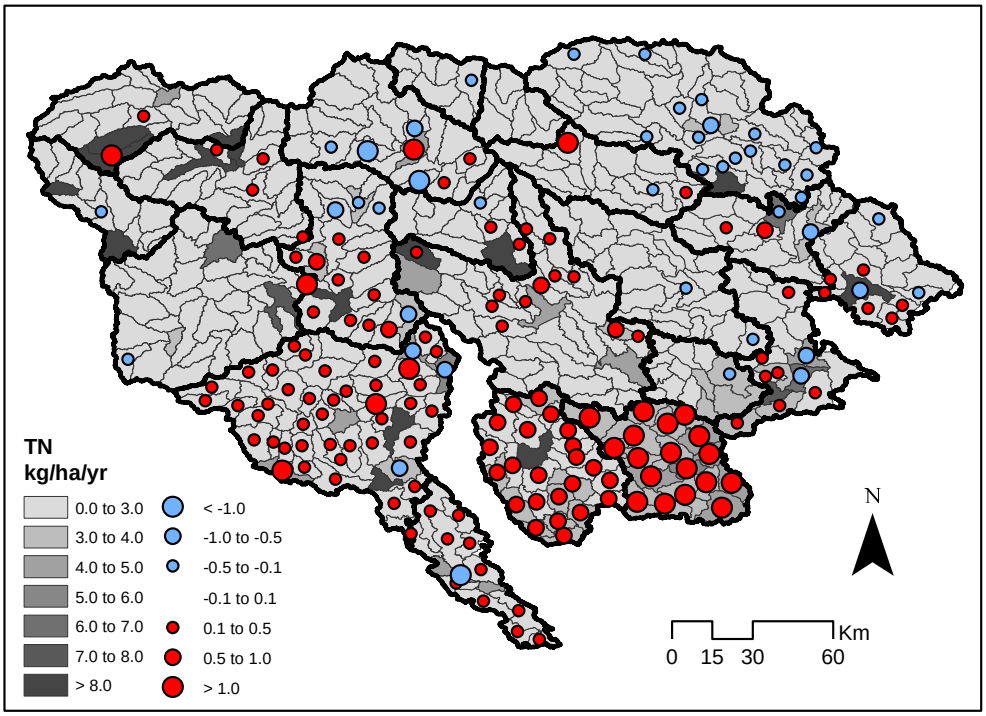
Appendix H. *Percent TN Retention Map*

The map below shows percent TN retention from each HUC12 to downstream points for each basin (Kelly for all CFRB watersheds except Chinquapin and Tomahawk, Ft. Barnwell for NRB, and Washington for TPRB). Black outlines are the watersheds used in this study.

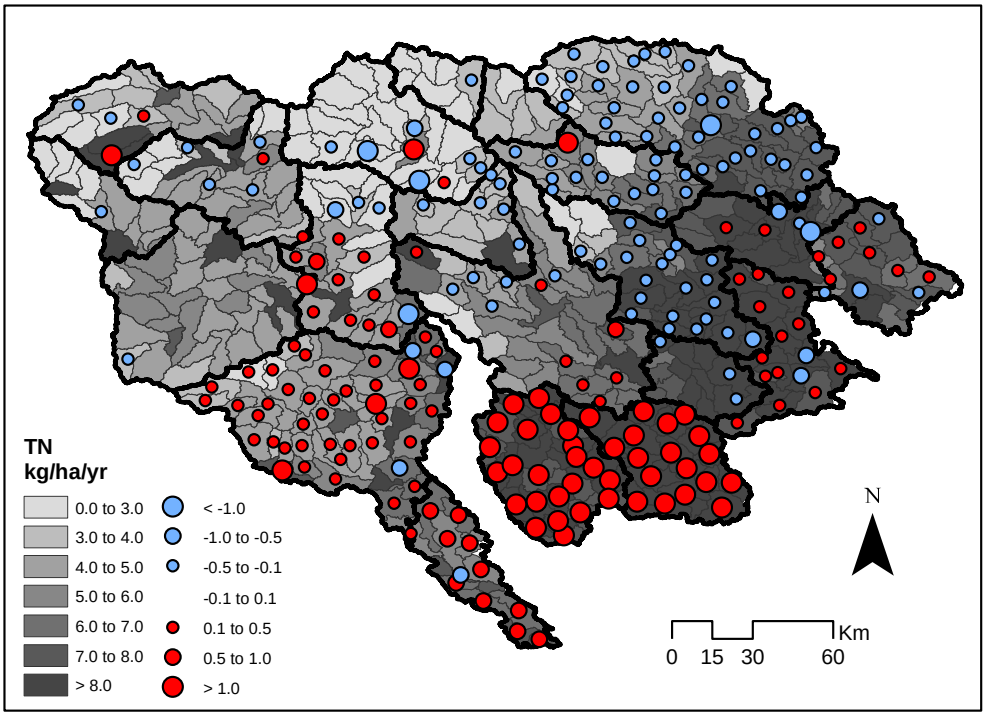


Appendix I. *TN Loading Rate Maps*

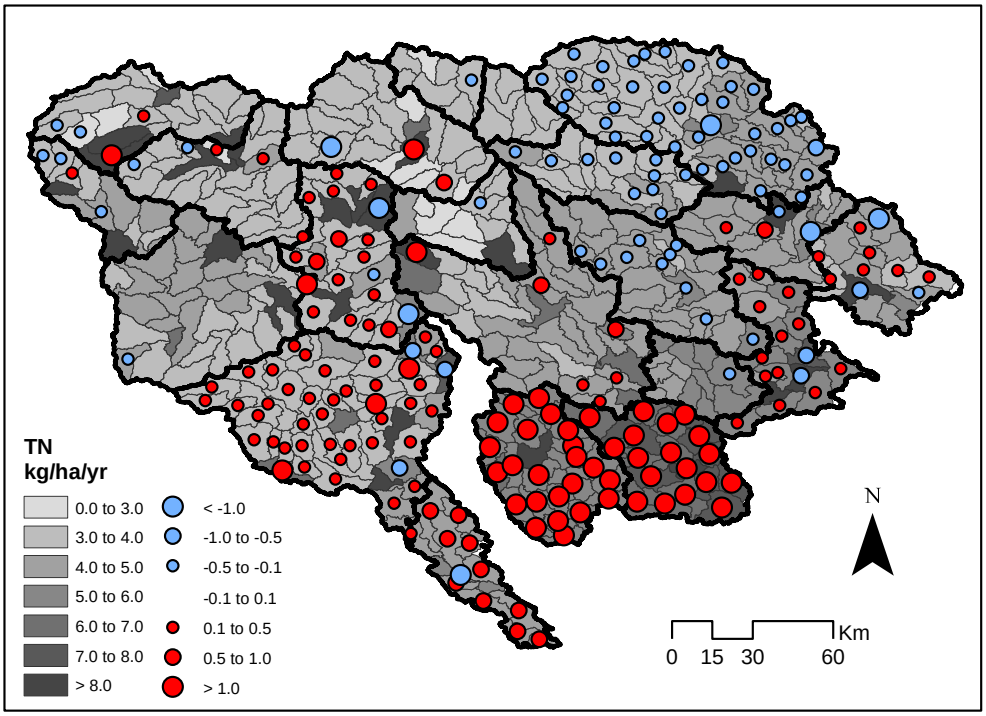
The maps below show TN loading rates (kg/ha/yr) using 5th (a) and 95th (b) percentile precipitation values for each HUC 12. Also shown are TN loading rates not including stream and water body retention using mean precipitation values (c) for each HUC 12. Loading rates for the year 2002 are in grey shading and points reflect change in rates from the year 1994 to 2012. In maps (a) and (b) stream and reservoir retention is included and rates reflect loadings to downstream locations for each basin (Kelly for CFRB, Ft. Barnwell for NRB, and Washington for TPRB).



(a)

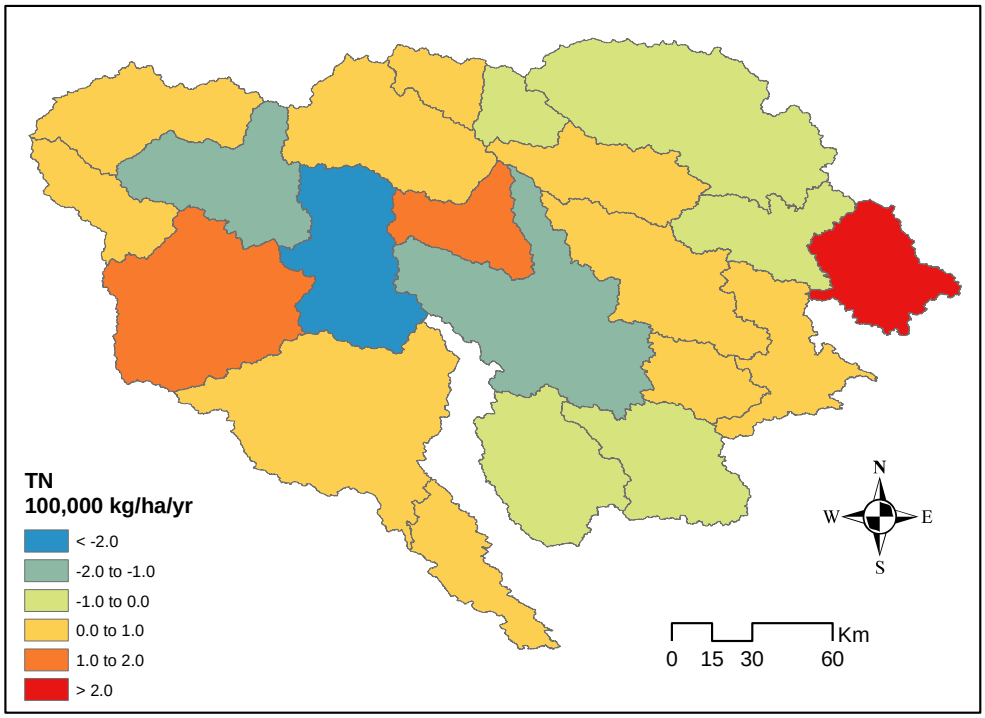

TN
kg/ha/yr
0.0 to 3.0
3.0 to 4.0
4.0 to 5.0
5.0 to 6.0
6.0 to 7.0
7.0 to 8.0
> 8.0
< -1.0
-1.0 to -0.5
-0.5 to -0.1
-0.1 to 0.1
0.1 to 0.5
0.5 to 1.0
> 1.0
N
0 15 30 60
Km

(b)

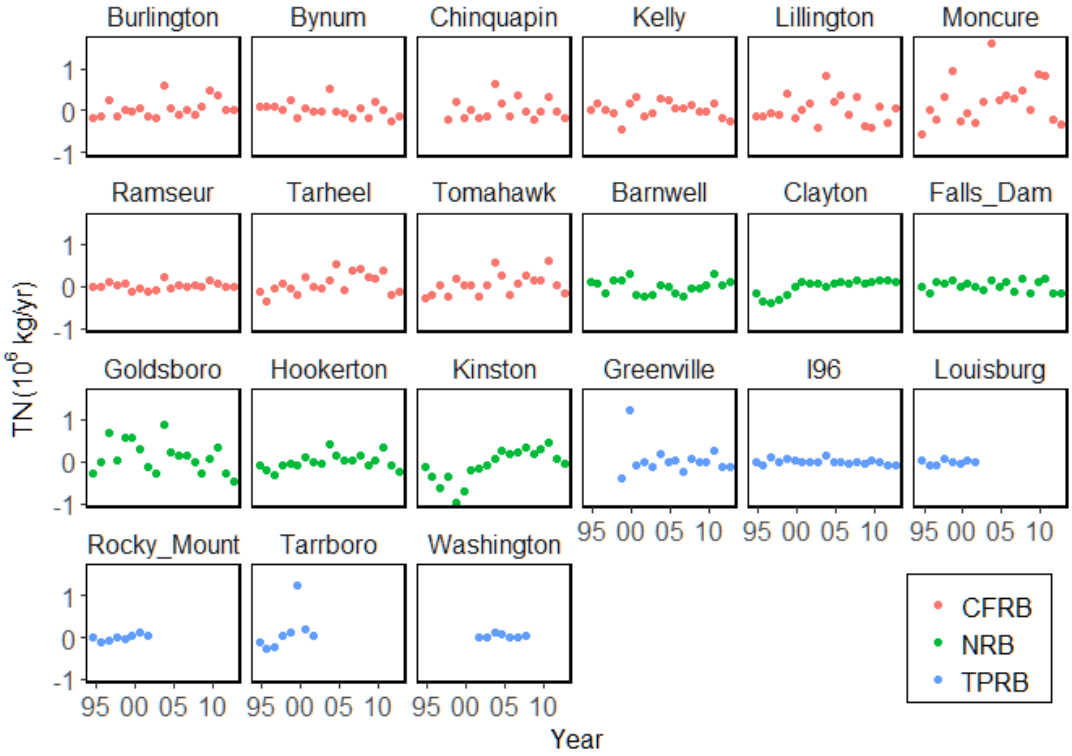

TN
kg/ha/yr
0.0 to 3.0
3.0 to 4.0
4.0 to 5.0
5.0 to 6.0
6.0 to 7.0
7.0 to 8.0
> 8.0
< -1.0
-1.0 to -0.5
-0.5 to -0.1
-0.1 to 0.1
0.1 to 0.5
0.5 to 1.0
> 1.0
N
0 15 30 60
Km

(c)

Appendix J. *Values of Random Intercept Term α by Watershed*

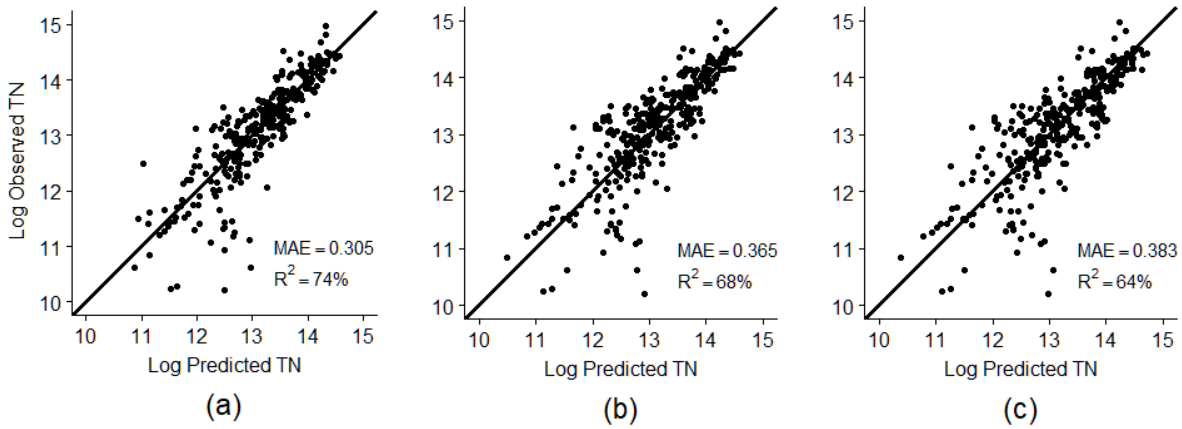


Appendix K. *Plot of Residuals as a Function of Time for Each Watershed*



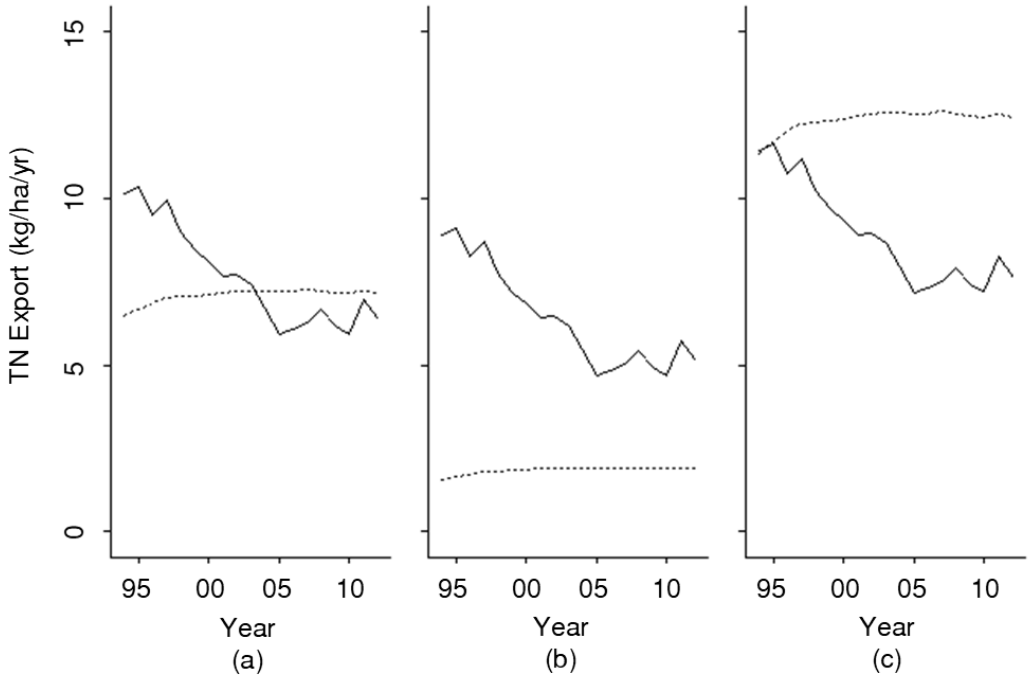
Appendix L. *Log Observed vs. Predicted Plots of Incremental TN*

Shown below are plots of log observed vs. predicted incremental TN loadings using (a) full model with fixed and random effects, (b) full model with fixed effects only, and (c) cross validation with fixed effects only.

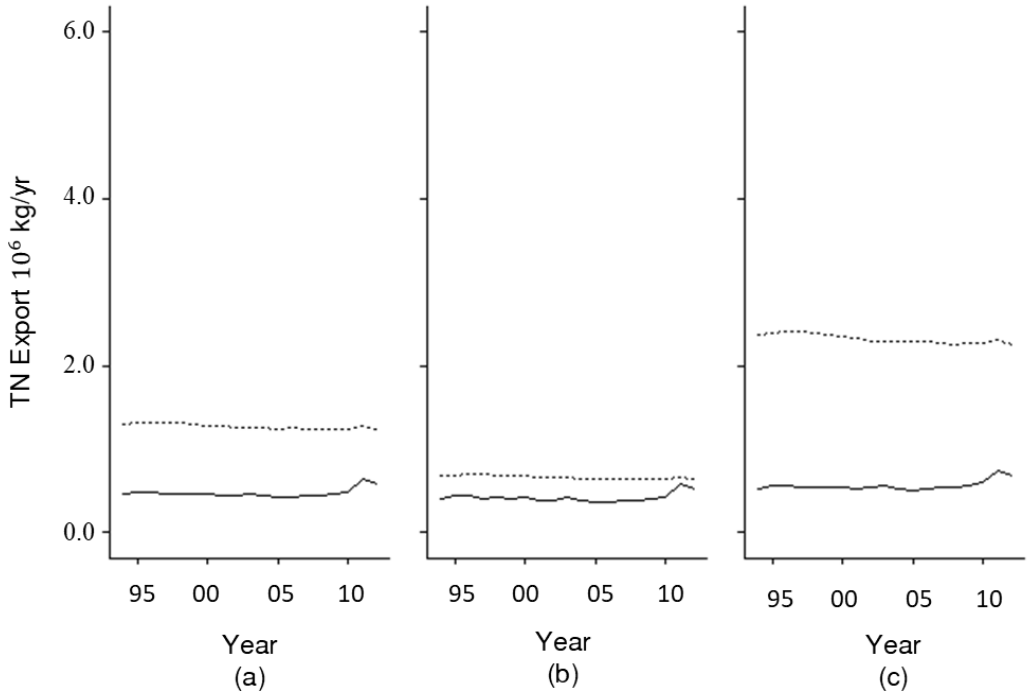


Appendix M. *Effective Export Rates and Loadings*

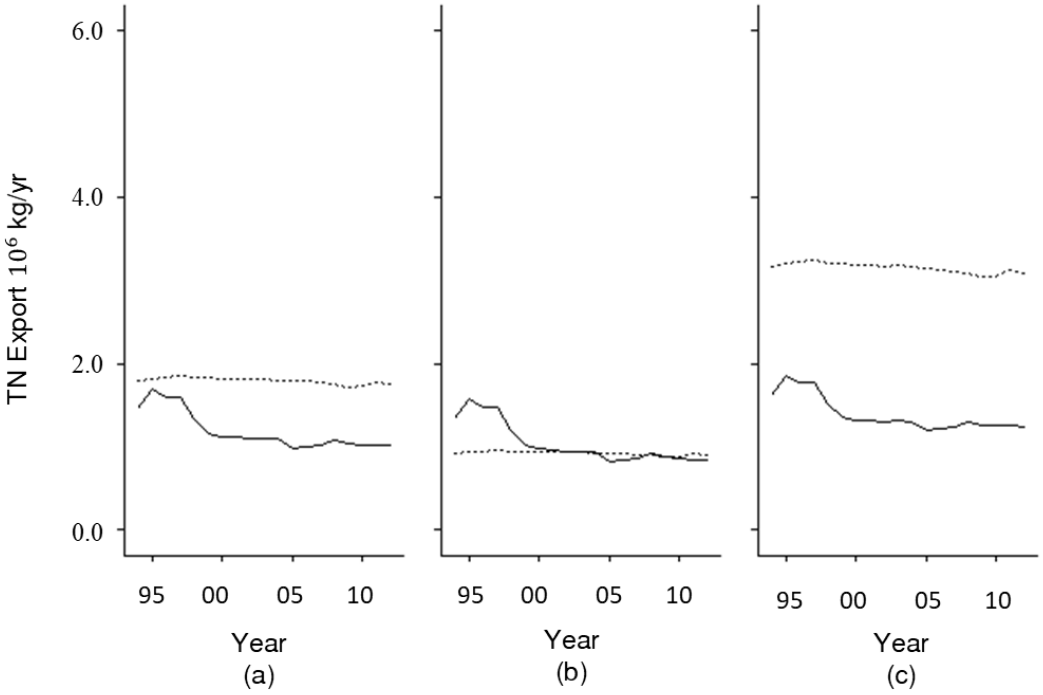
Effective export rates (kg/ha/yr) for all three basins (i) and effective loadings (kg/yr) for the TPRB (ii), NRB (iii), and CFRB (iv), are shown in the figures below. Developed (solid) and agriculture (dashed) effective loadings or export rates are calculated using the mean (a), 5th (b) and 95th (c) percentile precipitation values.



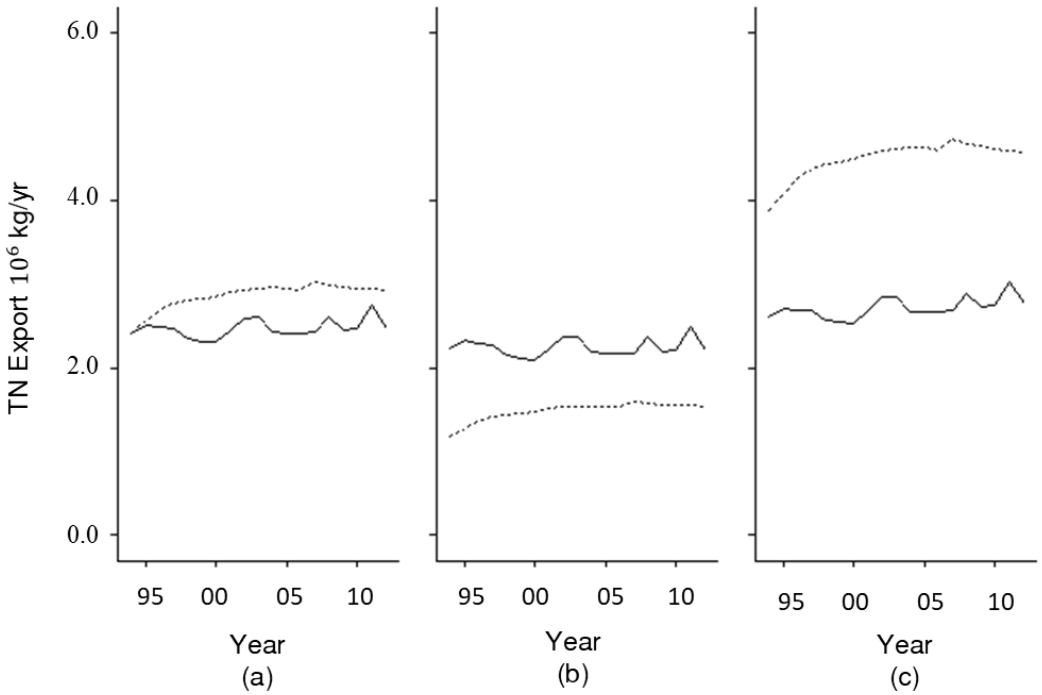
(i)


6.0
4.0
2.0
0.0
TN Export 10^6 kg/yr
95
00
05
10
Year
(a)
(b)
(c)

(ii)


6.0
4.0
2.0
0.0
TN Export 10^6 kg/yr
95
00
05
10
Year
(a)
(b)
(c)

(iii)


6.0
4.0
2.0
0.0
TN Export 10^6 kg/yr
95
00
05
10
Year
(a)
95
00
05
10
Year
(b)
95
00
05
10
Year
(c)

(iv)