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Abstract Management efforts to control excess algal growth in the Neuse River and Estuary, North Carolina began in the 1980s, with an initial focus on phosphorus (P) input reduction. However, continued water quality problems in the 1990s led to development of a Total Maximum Daily Load (TMDL) for nitrogen (N) in 1999 to improve conditions in N-sensitive estuarine waters. Evaluation of the effectiveness of management actions implemented in the Neuse River basin is a challenging endeavor due to natural variations in N export associated with climate. A simplified approach is presented that allows evaluation of trends in flow-normalized nutrient loading to provide feedback on effectiveness of implemented actions to reduce N loading to estuarine waters. The approach is applied to five watershed locations, including the headwaters of the Neuse Estuary. Decreases in nitrate + nitrite (NO_3–N) concentrations occurred throughout the basin and were largest just downstream of the Raleigh metropolitan area. Conversely, concentrations of total Kjeldahl N (TKN) increased at many stations, particularly under high flow conditions. This indicates a relative increase in organic N (Org-N) inputs since the mid-1990s. Overall, patterns in different N fractions at watershed stations indicate both partial success in reducing N inputs and ongoing challenges for N loading under high flow conditions. In downstream waters, NO_3–N concentrations decreased concurrent with TMDL implementation in the upper portion of the estuary but not in the middle and lower reaches. The lack of progress in the middle and lower reaches of the estuary may, at least in part, be affected by remineralization of settled particle-bound N deposited under high river flows.

Keywords TMDL · Nutrient management · Eutrophication · Nitrogen · Phosphorus · Trends

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Introduction

Excessive loading of nutrients to coastal areas is a widespread problem, adversely impacting the overall health of estuaries (Bricker and others 2007). Nutrient levels in estuaries can affect productivity at all trophic levels, with degradation of the ecosystem when high levels lead to pervasive algal blooms, including toxic species, bottom water hypoxia or anoxia, fish kills and habitat loss (Nixon 1995; Diaz and Rosenberg 2008). Although overall stresses to coastal environments include a range of physical, chemical, and biological alterations (e.g., Cloern 2001), it is generally recognized for many systems that inputs of nitrogen (N), and possibly phosphorus (P), need to be reduced (National Research Council 2000; Boesch and others 2001; Conley and others 2009). This broad need for nutrient reductions is illustrated by the current management efforts in the watersheds draining to the Gulf of Mexico (Rabalais and others 2009), Chesapeake Bay (Fisher and others 2006), and many other estuaries, including the Neuse Estuary in North Carolina (Paerl and others 2006a; Rothenberger and others 2009). The potential implementation of numeric nutrient criteria throughout the United States will likely further increase the need for broad scale nutrient reductions (United States Environmental Protection Agency 2001). Techniques to assess progress and provide feedback on effective strategies toward reduction goals are an essential component of these efforts.

The delivery of nutrients to estuarine waters is a product of both landscape and instream processes throughout the watershed. In the context of land-based inputs of nutrients, the mobility of nutrients can be affected by a number of factors, including the amount, timing, and composition of fertilizer applied, the location and extent of land disturbance, the magnitude, location and intensity of precipitation events, and the efficiency of stormwater controls (United States Environmental Protection Agency 2001). Once nutrients enter surface waters, the downstream transport may be affected by instream processes (e.g., sedimentation and resuspension, biological uptake, denitrification, etc.), with less effective trapping generally occurring in larger tributaries and rivers (Alexander and others 2000, 2008; Giffin and Corbett 2003). Assessing the effectiveness of management actions to reduce nutrient exports is confounded by large climatic variation among and within years. In this regard, the hydrologic impacts of climatic changes, including a recent increase in tropical cyclone activity (Webster and others 2005), resultant flooding (Bales 2003; Paerl and others 2006a, 2010) and regional droughts need to be incorporated in assessments and management of nutrient loads to estuarine and coastal waters, as typified by the tropical cyclone and drought-prone Neuse River (NR) watershed (Lebo and others 2002; Peierls and others 2003; Burkholder and others 2006; Paerl and others 2006a, 2010).

Management efforts to control nutrient-driven excessive algal growth, including nuisance cyanobacterial blooms in the NR and hypoxia-generating and ichthyotoxic dinoflagellate blooms in the Estuary began in the 1980s and initially focused on P controls (e.g., Paerl and others 2004). A P detergent ban and total P (TP) discharge limits on point sources in 1988 were implemented to address freshwater algal blooms, which effectively decreased P concentrations and algal blooms in the tidal freshwater portion of the system (Stow and others 2001; Lebo and others 2002; Paerl and others 2004). However, fish kills and low bottom water DO levels were regularly observed in downstream waters in the 1990s (Paerl and others 2004; Burkholder and others 2006), leading to a re-examination of management actions (NCDWQ 2009). Nutrient Sensitive Waters (NSW) Management Rules, approved in December 1997, required a 30% reduction in total N (TN) loading to the head of the Neuse Estuary at New Bern, NC (15A NCAC 2B 0.0232) (Fig. 1).

The NSW rules developed for the NR seek to achieve the required 30% reduction in TN loading by controls on dominant sources (NCDWQ 1999). Apportionment of TN inputs in the late 1990s included in the TMDL was 24% from point sources and 76% from nonpoint sources. Relative contribution from different land uses was urban (8%), cropland (67%), managed herbaceous (3%), and forests (20%), with atmospheric deposition on open waters accounting for 2% of the total. In the TMDL, point and nonpoint sources were each required to reduce TN loading to the estuary by 30%. The 10 largest municipalities and five counties were required to develop stormwater management plans while a 30% reduction was required from agriculture through implementation of best management practices (BMPs). Implementation of new Clean Air Act provisions also was expected to decrease atmospheric TN inputs. Control of TN inputs from forests was accomplished by protection of existing riparian buffers along all perennial and intermittent waterbodies in the basin. These control actions provided the means to implement the Total Maximum Daily Load (TMDL) developed for the NR basin (NCDWQ 1999, 2001).

An accurate evaluation of the effectiveness of nutrient controls must separate the impacts of the practices or limits implemented from expected variation in nutrient transport

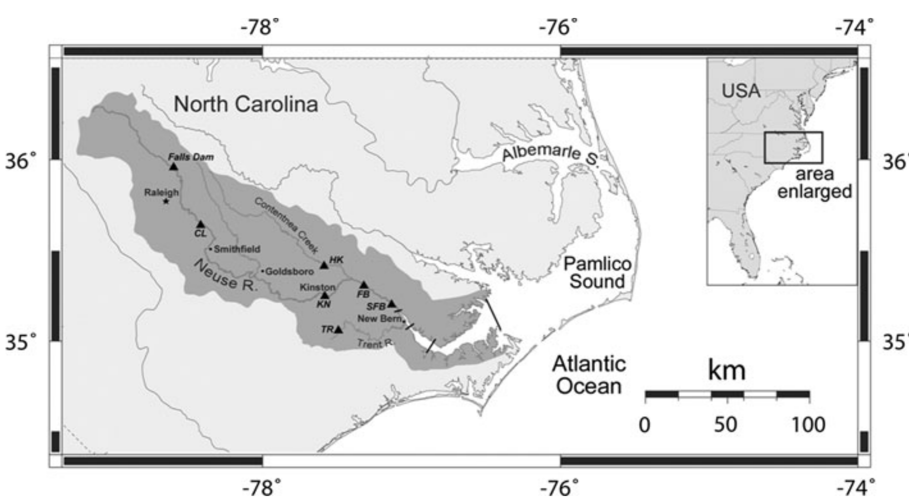

Fig. 1 Neuse River watershed identifying locations and sub-drainage areas. Locations are: *CL* Neuse River at Clayton, *KN* Neuse River at Kinston, *SFB* Neuse River at Streets Ferry Bridge, *HK* Contentnea Creek at Hookerton, and *TR* Trent River at Trenton. *Bars* across the estuarine portion of the system denote estuarine zones, starting with River (at SFB) and proceeding downstream with Upper (at New Bern), Middle, and Lower Estuary

driven by variation in rainfall and flow. Achieving a successful outcome for the NR basin must ultimately include reduction of TN loading to estuarine waters, but year-to-year loading reflects the combined effects of river flow and N inputs. Previous efforts to evaluate trends of N and P levels in the NR system have shown a decrease in TP beginning in 1988, following implementation of P controls and increased nitrate (NO_3–N) and TN concentrations in the middle of the basin through the mid-1990s (Lebo and others 2002; NCDWQ 1999; Qian and others 2000; Stow and others 2001). Interpreting nutrient trend data over the past decade is confounded by the series of tropical storms and hurricanes that have impacted Eastern North Carolina since the mid-1990s (Paerl and others 2010). Two previous studies (Lebo and others 2002; Stow and Borsuk 2003) considered river flow impacts in the structure of the analysis. However, the approaches used were data-intensive and involved considerable statistical manipulation.

The focus of this contribution is to describe and apply a relatively simple approach to account for the impact of flow variation on nutrient trends. Because of prior actions taken on TP reductions in the Neuse River system that were effective in arresting upstream freshwater cyanobacterial blooms, the emphasis here is on reducing N inputs, which has been identified as the most effective approach for controlling downstream brackish water blooms (Paerl and others 2004; NCDWQ 2009). Because data are available on both dissolved inorganic N and total Kjeldahl N concentrations, we are able to distinguish inorganic from organic fractions of the TN. This allowed us to examine relative trends in these N fractions. Both fractions are relevant to downstream eutrophication problems, since prior and ongoing studies indicate that they are capable of stimulating and sustaining algal production (Peierls and Paerl 1997; Harrington 1999) at levels that exceed the State of North Carolina's chlorophyll *a* standard of 40 µg/l (Paerl and others 2007, 2010), which is the target for the Neuse Estuary N-based TMDL (NCDWQ 2009). The objective of this study is to utilize available data to characterize overall change in N delivery to the Neuse Estuary as well as change associated with different flow regimes and N fractions. By examining these components of N loading, feedback to management is increased, providing an opportunity for additional insight on relative effectiveness of individual actions implemented.

Materials and Methods

Site Description

The Neuse River (NR) Basin covers approximately 16,000 km^2 of eastern North Carolina, USA and stretches from the northern Piedmont west of Durham to Pamlico Sound (Christian and others 1991) (Fig. 1). Over its southeasterly course of >320 km, the headwater NR flows past several cities, in particular the Raleigh metropolitan area, and through several smaller cities and predominantly rural drainage areas to the east. Rapid population growth has occurred in the basin over the past several decades, with >70% increases between 1970 and 1990 in both the Raleigh and coastal regions (North Carolina Division of Water Quality (NCDWQ) 1998); developed areas (and associated wastewater discharges) in the Upper Neuse drainage account for 17.0% of total area compared with a basin average of 11.5% based on 2001 data (NCDWQ 2009). Overall, considerable nutrient loading occurs to the NR and its tributaries from nonpoint sources, including runoff from row-crop and confined animal operations, and discharges of treated wastewaters from municipal and industrial facilities (e.g., Stow and others 2001). The rural character of the Middle Neuse and Contentnea drainage areas is reflected in the proportions of 34.4 and 44.1%, respectively, of the landscape classified in the National Land Cover Database for 2001 as agriculture (NCDWQ 2009). Confined animal operations are concentrated throughout the middle portions of the basin, including Contentnea, Middle Neuse, and Trent River areas (NCDWQ 2009).

The NR Estuary is relatively shallow with average depths ranging from 2.4 m at New Bern to 5.2 m near Pamlico Sound (Giese and others 1985). Due to restricted flow through the Outer Banks between Pamlico Sound and the Atlantic Ocean, the typical tidal range in the NR Estuary is <0.3 m, and short-term variation in water levels and salt wedge movement are primarily controlled by winds (e.g., Giese and others 1985). The NR is a highly productive system (Mallin and others 1991; Boyer and others 1993), and since the early 1970s it has had sufficient nutrient loads to support algal blooms (Hobbie and Smith 1975). Algal blooms with chlorophyll *a* (Chl*a*) concentrations exceeding the State water quality standard of 40 µg/l have been reported along the lower river upstream of New Bern and throughout the estuary (e.g. Lebo and others 2002). Although algal blooms are not a new phenomenon in the NR, Paerl and others (1995, 2007) indicated the magnitude and frequency of blooms in the estuarine portion of the system appeared to have increased between the 1970s and the 1990s. Nitrogen is generally the nutrient in shortest supply and hence potentially limiting (Christian and others 1991; Rudek and others 1991). On a system level, increased river flow following large rainfall events has been shown to increase algal production in the NR Estuary and affect the algal community composition (Mallin and others 1993; Paerl and others 2006b, c; Pinckney and others 1999).

Data Sources

Water quality data were compiled from State, University, and NGO (Weyerhaeuser) research and monitoring efforts to characterize variation in nutrient and Chl*a* levels at selected watershed locations and along the NR Estuary from 1980 to 2009. Data were obtained from surface and mixed layer (i.e., photic zone) depths, either collected as discrete grab samples or photic zone composites (using a tube sampler) at a range of stations located along a longitudinal transect of the system from microtidal freshwaters at Streets Ferry Bridge (SFB) upstream of New Bern to the confluence with Pamlico Sound (Fig. 1). Riverine data were compiled from four stations either along the mainstem of the Neuse River (Clayton and Kinston) or from major tributaries (Trent River at Trenton and Contentnea Creek at Hookerton). Nutrient data from bottom water samples at estuarine stations were omitted from the primary data base, since sufficient data for trend evaluation were not available throughout the period evaluated. Lebo and others (2002) provide additional information on data compiled through 2000. Exact monitoring locations within the Neuse Estuary for the different sampling programs varied over time, and thus, the data for SFB to Pamlico Sound were grouped into four estuarine regions similar to the approach used by Harned and Davenport (1990) (Fig. 1).

Nutrient fractions evaluated included ammonium (NH_4–N), nitrate + nitrite (NO_3–N), total Kjeldahl N (TKN), total N (TN), and total P (TP), as well as Chl*a*. When not reported in the data base, TN was computed as TKN plus NO_3–N. For programs reporting both TKN and NH_4–N, the organic N (Org-N) fraction was calculated as TKN minus NH_4–N. The majority of sampling was done by surface grabs, except data collected by the NC Division of Water Quality at the Kinston location (Fig. 1), which also was collected by grab sampling and, after 1995, collection by automated sampler. With the exception of 2001–2003, grab sample data from NC Division of Water Quality were used for Kinston so that the number of sampling dates across periods were generally similar. For 2001–2003 when grab sample data were not available, available data collected by automated sampler were subsampled to generate two survey dates per month (closest sampling date to the 1st and 15th), comparable to the frequency of data collection throughout the analysis period. Parameter values listed as less than the detection limit were included at the detection limit (DL) provided the DL was equal to or less than 0.05 mg/l for NH_4–N, NO_3–N, and TP or less than or equal to 0.1 mg/l for TKN and TN. Table 1 lists data used by source, period, and location.

Flow data for evaluated sites were obtained from U.S. Geological Survey data records, available online. Table 2 lists the periods for which flow data were available to define the flow distribution for each site. Daily average flows for each gauging location were used in the analysis, with the exception of the SFB location on the Neuse River. For SFB, flow data from upstream gages at Kinston and Hookerton were used to estimate flow intervals for a longer time period than could be done using the gage installed at Fort Barnwell in 1996. Estimated daily flow at SFB, except during September–October 1999 following Hurricane Floyd, was computed as the sum of reported flow at Kinston and 1.84 times the flow at Hookerton. Flows used were 2-day average flows for the current and prior day to account for downstream time of travel, and the multiplier of 1.84 was used to account for ungaged drainage area. For the period affected by Hurricane Floyd (September–October 1999), estimated flow for the Fort Barnwell location was used to better represent flow at SFB due to excessive rainfall in the Contentnea Creek watershed that was not representative of overall ungaged drainage. A second factor taken into account in the flow data utilized was the creation of Falls Reservoir in 1982. In the case of the Neuse River at Clayton, the flow record used was limited to the period after 1982 when Falls Reservoir was built due to changes in the overall flow distribution associated with flood control and low-flow augmentation objectives of the project.

Table 1 Nutrient and Chl*a* data sources used in analysis

Data source[a]	Years[b]	Location[c]
ECU	1980–1987	Streets Ferry, estuary stations
LNBA	1995–2009	Clayton, Kinston
NCDWQ	1980–2009	Clayton, Kinston, Streets Ferry, Trenton, Hookerton, estuary stations
UNC	1981–2009	Streets Ferry, estuary stations
USGS	1980–1999	Clayton, Kinston, Hookerton, Trenton
Weyerhaeuser	1980–2002	Streets Ferry, estuary stations

[a] *ECU* East Carolina University, *LNBA* Lower Neuse Basin Association, *NCDWQ* North Carolina Division of Water Quality, *UNC* University of North Carolina at Chapel Hill, *USGS* United States Geological Survey

[b] Additional data before 1980 available from NCDWQ and USGS programs

[c] Data period of record varies by location for some programs. Data may not be available for all locations for the full period listed

Flow-Normalized Trends

Evaluation of trends in nutrient concentrations throughout the NR watershed was done by comparing average values for data grouped into three flow intervals. Selection of three flow intervals was done to provide feedback on changes under base flow conditions (low flows), which are more affected by point source and groundwater inputs, and

Table 2 Flow information for monitoring locations evaluated

Location	USGS ID	Data record	Flow averages (m^3/s)[a]			DWQ ID
			Low	Middle	High	
Clayton	02087500	1983–2007[b]	7.69	13.9	69.4	J4170000
Kinston	02089500	1930–2007	18.7	51.2	171.8	J6150000
Streets Ferry	No gage[c]	1930–2007	26.5	76.8	258.7	J7930000
Hookerton	02091500	1930–2007	3.67	13.0	49.0	J7450000
Trenton	02092500	1951–2007	0.454	2.48	13.4	J8690000

Average flows are listed for the three flow intervals used to group nutrient concentration data. Intervals based on tertiles of long-term flow data at each location

[a] Average flows by interval for data collected through 2007

[b] Building of Falls Reservoir impacted flow distribution beginning in 1983

[c] Flows at Kinston and Hookerton were used to estimate long-term flow range except following Hurricane Floyd when Fort Barnwell station on Neuse River, ID = 02091814, was used

storm flows (high flows) more affected by nonpoint source inputs. Thus, the approach allows inferences, within the variability of the data, about effectiveness of measures to control point and nonpoint sources. A second factor in selecting three flow intervals was the practical limitation of the number of sampling points to derive trends provided by typical monitoring done on a monthly or biweekly schedule.

Long-term flow records for the NR basin were used to define the 33rd and 66th percentile of daily flows for locations with either gaged or estimated (i.e., SFB) flow (Table 2). These flow thresholds were then used to group nutrient data by low (0–33%), middle (34–66%), and high (67–100%) flow regimes. Average nutrient concentrations, by flow interval, were computed as the mean of available data for each nutrient fraction and as the flow-weighted average of the same data set. Values reported here for watershed locations were the average of the mean and flow-weighted average for each time period and flow interval of interest. The rationale for using both approaches to derive average concentrations is to provide a more robust metric for trend assessment than either approach individually.

Nutrient loads, for the long-term cumulative flow distribution, were computed by parameter and location. These flow-normalized loads were the product of average concentrations (C_i) described above and average flow volume (Q_i) contributed by each of the flow intervals (L, M, and H) over the full period of record multiplied by a constant to convert values to kg/year:

$$\text{Load} = \text{Constant} \cdot \sum_{i=\text{L}}^{\text{H}} Q_i \cdot C_i \quad (1)$$

The rationale for this approach was based on the fact that climate affects river flow while management actions primarily affect nutrient concentrations for a given river flow. Change over time associated with implementation of management actions for the NR basin was evaluated by comparing loads to benchmark periods either established in regulation (i.e., 1991–1995 for TN) or just prior to management action implementation (pre-1988 for TP).

Annual Nutrient Loads

Estimated annual loads of nutrients in the NR at SFB were calculated from the derived average parameter values by flow interval and reported river flows. To ensure sufficient data were included to derive reliable average nutrient concentrations for both low and high flow regimes, nutrient concentration data from a 3-year moving window centered on the year of interest were used to calculate annual averages. Flow volumes used in the calculation for each of the three flow intervals were only from the year of interest. Annual load was computed as the sum of loads derived for each of the three flow intervals.

Results and Discussion

Detection of Trend in Context of TMDLs

Evaluation of instream response, in terms of water quality improvement, to implemented TMDL actions is a key component of tracking progress toward attainment of water quality standards. Determining progress can be constrained by natural variability in nutrient concentrations, interannual variation in river flow associated with climate, and limited resources to collect essential monitoring data. Previous researchers have developed approaches to account for predictable seasonal and flow dependencies of nutrient concentrations by a combination of partitioning data by season and normalizing concentrations to particular flows of interest (e.g., Stow and others 2001). Often, these methods are computationally intensive, such as the approach recently proposed by Hirsch and others (2010) accounting for variation due to season and flow in the evaluation of change in nutrient concentrations over time. The approach applied in this study evaluates feedback on nutrient trends that can be provided by averaging concentrations by flow interval and across multi-year periods.

The accuracy of load predictions based on the averaging approach used was evaluated using a fabricated 10-year daily record based on actual 1990–1999 flow data reported for the Neuse River at Kinston and simulated concentrations of NO_3–N, TKN, and TP. As in the overall data analysis, TN concentration was calculated as the sum of NO_3–N and TKN. The NH_4–N fraction was not included in the evaluation due to inclusion in TKN and low overall contribution to TN. An average concentration-flow relationship was developed for each fraction to predict a hypothetical concentration value to which a random component was added to account for variability in actual data available for the Kinston location during 1990–1999. A normal distribution was assumed for non-flow related variation in the data. Daily values were independently derived to allow maximum variability in the evaluation of potential method error. Reference annual nutrient loads for determination of error were computed as the sum of calculated daily loads. Subsets of the simulated daily record were extracted using a systematic sampling frequency of 7, 15, and 30 days throughout the 10 year period. The extraction process was done seven times for each monitoring frequency, starting on a different day of the record.

Available data to compute nutrient loads based on three defined flow intervals varied by year in the simulated record due to years of drought and with high rainfall. On an annual basis, the number of years with three or more data points for all three flow intervals was 30, 84, and 94% for sampling intervals of 30, 15, and 7 days, respectively. However, available data for the 15 day sampling interval increased to >10 for all three intervals for 99% of simulated years when a 3-year moving window was used. A key limitation of analysis of trends is the availability of data with which to assess change over time. The benefit of the approach used here is that reliable trends can be derived for typical monitoring program sampling frequency through averaging across multi-year periods. Average error for subsamples based on the 15-days sampling interval with 3-year averaging, relative to computed values from the full daily data record, for predicted TN and TP annual loads was 3.6 and 6.5%, respectively, with average error of 5.3 and 4.3%, respectively, for the NO_3–N and TKN fractions. The 95th percentile for individual years were 10, 10, 8, and 14% differences from the "actual" reference value, respectively, for NO_3–N, TKN, TN, and TP.

The uncertainty of predictions of change in nutrient loads derived from the flow interval approach was evaluated using simulated data sets, as described above, for the NR at Kinston for the post-TMDL period of 2000–2009. The approach utilized statistically derived data sets without trend to characterize changes in N fractions that could results from natural variability in concentrations even if there were no change associated with implemented actions. A total of 45 runs were done to develop probability distributions of percent change in NO_3–N, TKN, TN, and TP due to natural variability in concentrations. Consistent with the sampling record for the NR, a 15 day sampling interval was used in the uncertainty analysis. To generate the 45 runs, subsamples at the 15 day interval were extracted starting on each of the first 15 days of the simulated 10-year data record for three independently developed data sets. Averaging periods of 3 and 5 years to derive trends were evaluated. For the analysis, the first 3 (2000–2002) or 5 (2000–2004) years of predictions in each run were used as the benchmark to which each successive multi-year average was compared. Figure 2 presents a summary of the results of the uncertainty analysis. Changes in flow-normalized nutrient loading greater than the values in the figure would have a confidence greater than the percentile shown. For example, a change in NO_3–N of 6.0% using the 5-year averaging approach would have a 90% confidence of indicating actual change but not a 95% confidence. Clearly, longer averaging period can detect smaller changes in flow-normalized loading; using a 5-year period decreased the 95% confidence value for TN to 4.7% compared with 7.6% using a 3-year period for the same data set.

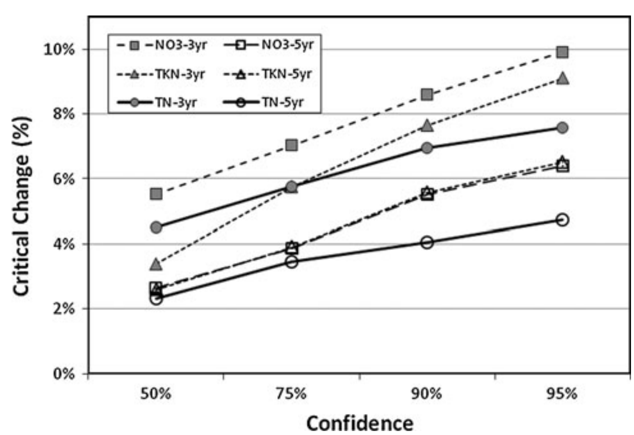

Fig. 2 Uncertainty analysis for flow interval approach using 3 and 5-year averaging periods. Values shown are percentiles of relative change from reference period for each N fraction. Confidence percentiles for TP ranged from 6.8% (50th) to 14.2% (95th) for a 5-year averaging period

Nutrient Delivery to Estuarine Waters

Annual TN loading at SFB ranged from 2.0 to 8.4 × 10^6 kg/year for years during 1980–2009, with a median value of 3.7 × 10^6 kg/year (Fig. 3). With regards to interannual variation, annual TN loading was highly correlated ($r^2 = 0.93$) with annual mean river flow calculated at SFB as has been previously reported for the NR system (e.g., Stow and others 2001). Overall, average contributions of NH_4–N, NO_3–N, and Org-N to the total for 1980–2009 were 5, 53 and 41%. These relative proportions did not vary consistently with river flow across all years. However, there was an increase in the contribution of the Org-N fraction to TN loading at SFB in the mid-1990s; Org-N contribution increased from 35% of TN in 1980–1994 to 48% for 1995–2009. For TP loading at SFB, annual values ranged from 0.25 to 1.1 × 10^6 kg/year (median = 0.45 × 10^6 kg/year). TP loading also was highly correlated ($r^2 = 0.77$) with annual mean river flow.

Evaluation of the effectiveness of management actions implemented to reduce TN loading to the NR Estuary was done by comparing estimated loading under long-term average flow conditions (Fig. 4). These flow-normalized loads provide feedback on the impact of concentration changes over time on average loading to the NR Estuary. Loads of NO_3–N, TKN, and TN were computed by 5-year moving averages and compared with the corresponding value for the 1991–1995 reference period. Negative numbers presented in figures indicate decreased loading under average flow conditions. Tracking relative change in TN loading to NR Estuary by fraction showed that reductions in flow-normalized NO_3–N loading have occurred but not in TKN loading (primarily Org-N, see Fig. 3). Flow-normalized NO_3–N loading decreased beginning in the 1992–1996 period prior to TMDL implementation and reached a minimum value of −25% in 1999–2003. The achieved decrease was >15% for all periods beginning

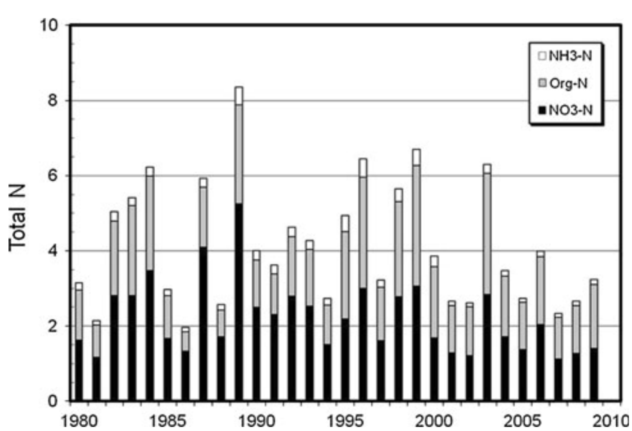

Fig. 3 Estimated annual loading for Neuse River at SFB by TN fraction in 10^6 kg/year

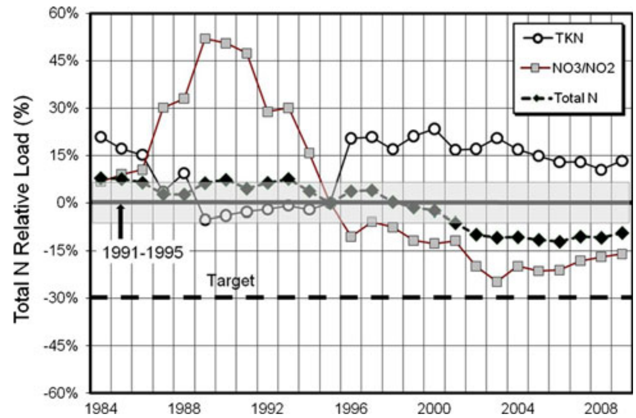

Fig. 4 Flow-normalized loading for Neuse River at SFB by TN fraction. Values are for a 5-year averaging period plotted in year 5 and are expressed as a percent difference from the 1991 to 1995 baseline load define in the TN TMDL. Target is a 30% decrease in TN from baseline. *Shaded envelope* provides approximate 95th confidence values from uncertainty analysis (see Fig. 2)

with 1998–2002 (Fig. 4). Conversely, the flow-normalized TKN loading at SFB has been consistently higher than the 1991–1995 reference period throughout the past 15 years. The abrupt increase in TKN loading in 1996 concurrent with the beginning of a period of elevated tropical storm activity for North Carolina indicates impacts from large storm events (e.g., Paerl and others 2006b) may be a contributing factor to the pattern. For TN, the combination of the patterns for NO_3–N and TKN yields a total reduction of 9.3–12% for periods ending in 2002–2009. Based on the uncertainty analysis summarized in Fig. 2, changes in flow-normalized NO_3–N, TKN, and TN are all much higher than expected from natural variations in nutrient concentrations.

Examining the average concentrations of N fractions by flow interval at SFB shows the changes integrated in Fig. 4 do not occur equally across the flow distribution (Table 3). For the NO_3–N fraction, average concentrations have decrease more for the low and middle flow intervals than for high flows. For example, reductions in NO_3–N for the 2005–2009 period from corresponding values for 1991–1995 were 43, 25, and 8%, respectively, for the low, middle, and high flow interval. The pattern for TKN, in contrast, shows higher average values for the middle and high flow intervals when data from the 2000s are compared with 1991–1995.

Watershed Source Regions

Potential differences in effectiveness of N and P management efforts were evaluated by comparing patterns in flow-normalized loading at three regions of the NR basin with different predominant sources for N inputs. Locations examined were the NR near Clayton below the Raleigh

Table 3 Average NO_3–N and TKN concentrations (mg/l) at SFB by 5-year period and flow interval

Parameter	Period	Low	Middle	High
NO_3–N	1981–1985	0.66	0.82	0.59
NO_3–N	1986–1990	0.98	0.94	0.87
NO_3–N	1991–1995	0.83	0.79	0.51
NO_3–N	1996–2000	0.71	0.66	0.45
NO_3–N	2001–2005	0.48	0.57	0.43
NO_3–N	2005–2009	0.47	0.59	0.47
TKN	1981–1985	0.61	0.58	0.53
TKN	1986–1990	0.52	0.46	0.44
TKN	1991–1995	0.49	0.47	0.46
TKN	1996–2000	0.48	0.53	0.60
TKN	2001–2005	0.49	0.50	0.55
TKN	2005–2009	0.51	0.53	0.53

Values are typically based on 20 or more data points

Table 4 Loading of N and P for 1980–2009 normalized to total drainage area

Parameter	Neuse@ Clayton	Cont@ Hookerton	Trent@ Trenton	Neuse@ SFB
NH_3–N	0.30	0.45	0.19	0.21
NO_3–N	2.16	2.71	1.97	2.11
Org-N	1.49	1.86	1.87	1.63
Total P	0.64	0.62	0.32	0.50

Values are in kg/ha

metropolitan area, Contentnea Creek near Hookerton, and the Trent River near Trenton. Table 4 summarizes average loading for each location for 1980–2009, compared with the NR at SFB. All three of the watershed source regions exhibited TN and TP areal loadings equal to or greater than the average values at SFB, with the exception of TP from the Trenton station. Overall, average TN and TP loadings ranged from 4.0 to 5.0 and 0.32 to 0.64 kg/ha, respectively.

Patterns in N and P loading over time for the NR near Clayton differed by parameter and flow interval (Fig. 5a). The flow-normalized loadings of NO_3–N and TKN both show periods of increasing and decreasing values while there is an overall decrease over time in TN loading. For NO_3–N, there was a progressive increase for 5-year periods ending in 1988 through 1997, followed by a large decrease through 2003 to values 48–54% below 1991–1995. The gradual change shown in Fig. 5a is actually an artifact of the 5-year averaging done to compare with the established reference approach; the NO_3–N concentration decreased 47, 37, and 21%, respectively, for low, middle, and high flows between 1997 and 1998. Conversely, TKN flow-normalized loading peaked in the 1980s and again in the 2000s. The combination of these two patterns yields a decrease in TN both in the early 1980s associated with a decline in TKN and in the late 1990s through early-2000s associated with decreased NO_3–N. Total N flow-normalized loading reached a minimum of approximately 30% below 1991–1995 levels before increasing again in recent years. For TP, there was a large decrease (>50%) between 1987 and 1988 concurrent with implementation of P control actions before a gradual pattern of increased loading

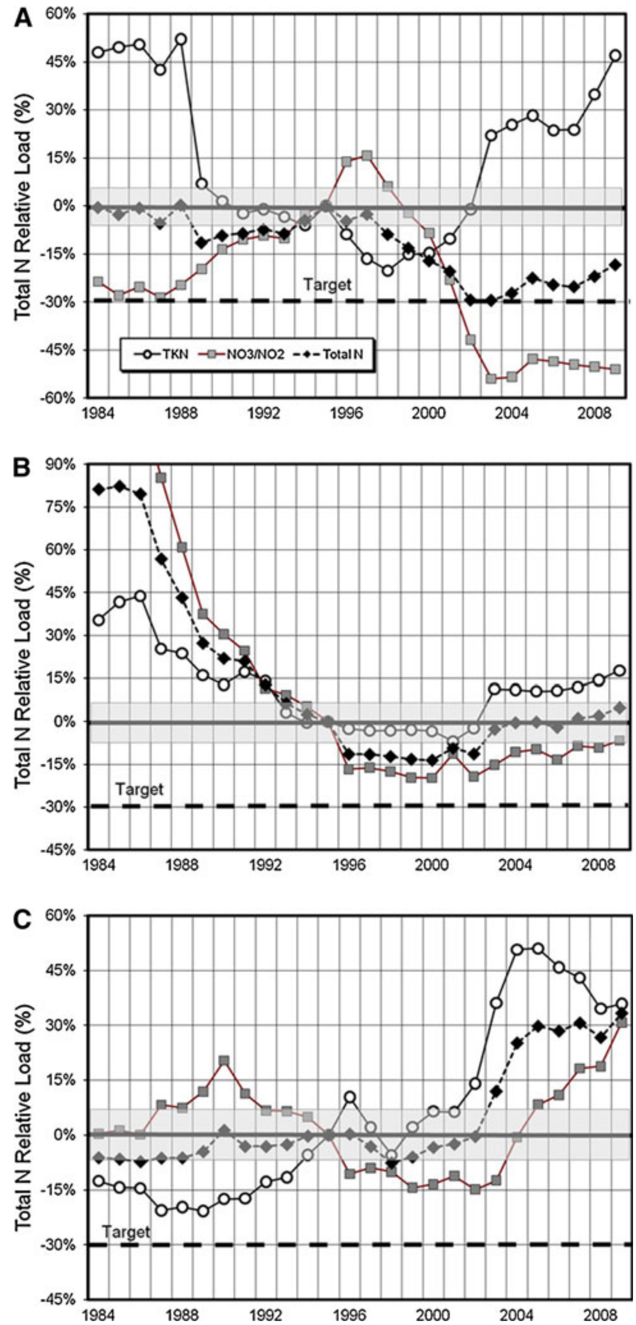

Fig. 5 Flow-normalized loading at watershed locations: **a** Neuse River at Clayton; **b** Contentnea Creek at Hookerton; **c** Trent River at Trenton. See Fig. 4 for more explanation

over the past decade to approximately 30% below the 1983–1987 value (data not shown).

Monitoring data on suspended sediment and turbidity levels in the NR near Clayton were compiled and averaged by the flow intervals to examine whether the trends in TKN levels in Fig. 5a can be explained by changes in particle-bound N. For the periods in the 1980s and 2000s, turbidity levels in the NR downstream of the Raleigh metropolitan area were significantly higher ($P < 0.01$) than in the 1990s (48 and 56 NTU vs. 25 NTU) for high flow conditions; turbidity for low and middle flows were not significantly ($P > 0.07$) different among the periods (Fig. 6). A similar analysis with data from the site just below Falls Dam (Fig. 1) did not show the same temporal pattern of elevation turbidity over the past decade. Thus, changes in the level of suspended sediment material may be a factor in the observed TKN pattern, and the source of potential particle-bound N appears to be downstream of Falls Dam.

The second watershed location examined is the Contentnea Creek subwatershed draining rural areas in the middle portion of the NR basin. Flow-normalized values for the Hookerton station for NO_3–N, TKN, and TN peaked in the mid-1980s and then decreased through the mid- to late-1990s (Fig. 5b). The largest change during periods ending in 1983–1996 occurred for the NO_3–N fraction (+122 to −17% relative to 1991–1995), accounting for 57% of TN. After this early decline, all three fractions remained relatively constant until 2002, with TN 11–14% below the 1991–1995 reference period. Then, both TKN and NO_3–N fractions increased again in periods ending in 2003–2009. The recent increase in NO_3–N and TKN flow-normalized loadings is mainly due to increases for the middle and high flow intervals (data not shown). For TP, flow-normalized loading was highest in the early 1980s and actually began to decrease before P control actions were implemented in early 1988. Review of turbidity and total suspended solids data for Hookerton did not show evidence for increases in recent years.

The third watershed location examined is the Trent River subwatershed draining rural areas in the lower portion of the NR basin. Figure 5c compares NO_3–N, TKN, and TN flow-normalized loads by 5-year period to the 1991–1995 reference period. The NO_3–N and TKN fractions at Trenton varied considerably over the past 30 years; NO_3–N peaked in the late 1980s and in recent years, while TKN peaked in the early 1980s and mid-2000s. Overall, the flow-normalized TN load was relatively constant at the 1991–1995 level for the mid-1980s through the early 2000s until increasing by 30% for periods ending in 2005–2009. Variation in TP concentration was limited to <0.05–0.15 mg/l over time, except in the late 1990s during a period of several large hurricane events with widespread flooding; TP concentrations of 0.2–0.8 mg/l occurred on many dates during 1996–1999.

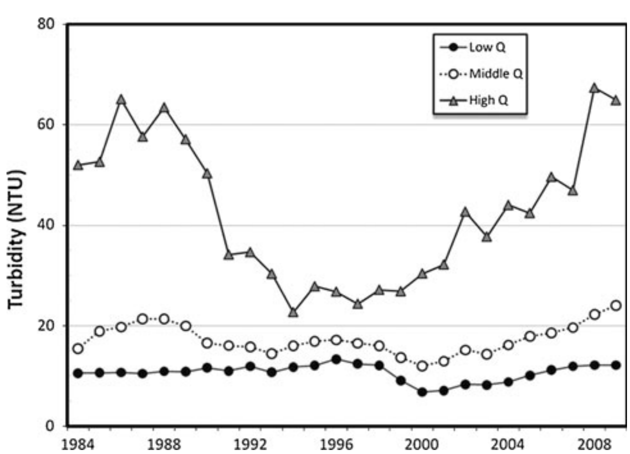

Fig. 6 Turbidity in Neuse River at Clayton. Values are 5-year averages by flow interval plotted in year 5 of each period consistent with average nutrient values reported for watershed locations

Estuarine Nutrient Patterns

Monitoring data on N and P concentrations along the NR Estuary from SFB to Pamlico Sound were averaged (mean) by zone, year, and decade (mean of annual means) to assess whether observed patterns in N loading yielded discernable estuarine response patterns. Typically, each of the four zones (River, Upper Estuary, Middle Estuary, and Lower Estuary) had multiple sampling locations for dates on which water samples were collected. Concentrations of NH_4–N varied considerably among years but accounted for only about 6–9% of TN, with the highest contribution in the Upper Estuary (Fig. 7). In terms of change in NH_4–N over time, the average concentration decreased significantly ($P < 0.03$) by 42–51%, depending on zone, between the 1980s and 2000s. The NH_4–N concentration also was consistently higher in the River and Upper Estuary zones than in the Middle and Lower Estuary. Indeed, peak NH_4–N concentrations for the 1980s and 2000s occurred in the Upper Estuary, indicating input of NH_4–N to the water column in that region. The most likely source of NH_4–N to the Upper Estuary zone is sediment release, although external inputs near the City of New Bern or Trent River inflow may contribute to a lesser extent. Concentrations of NH_4–N showed a lack of correlation along the NR Estuary either with calculated NH_4–N or TN loading at SFB when averaged by individual year or decade.

Variation in annual average NO_3–N concentrations in the River and Upper Estuary zones reflects the temporal pattern at Streets Ferry (decrease beginning in the mid-1990s), as depicted by flow-normalized loads at SFB (Fig. 4). Figure 7 illustrates the temporal pattern of a significant decrease in NO_3–N in the River zone between the

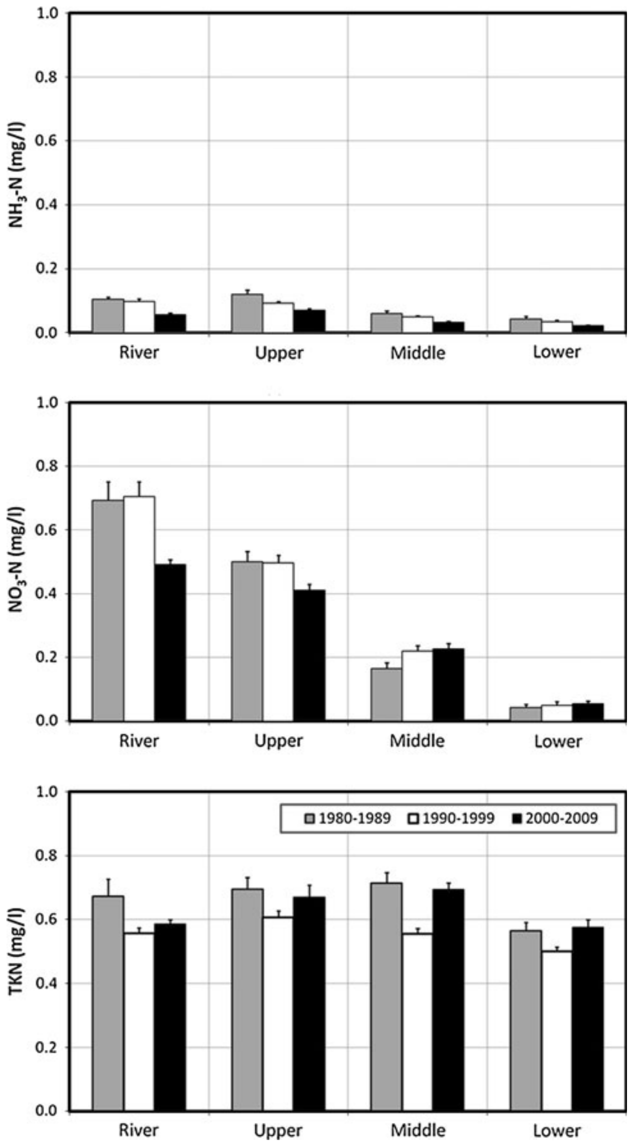

Fig. 7 Decadal mean concentrations of N fractions by zone in the NR Estuary. *Error bars* indicate mean ± SE for each respective period of annual mean values

1990s and 2000s. Spatially, NO_3–N concentrations were highest in the River and Upper Estuary, intermediate in the Middle Estuary, and lowest in the Lower Estuary. Despite the decrease in NO_3–N in the upper portions of the Neuse Estuary, there was not a corresponding decrease in NO_3–N average concentrations in the Middle (0.2 ± 0.1 mg/l) and Lower (0.1 ± 0.1 mg/l) Estuary zones concurrent with implementation of the Neuse Estuary TN TMDL. However, this lack of change in average NO_3–N between the 1990s and 2000s is an artifact of seasonal averaging; winter (December–March) NO_3–N concentrations were lower, on average, in the 2000s than the 1990s for both the Middle and Lower Estuary, but summer (June–September) values were higher. Comparing annual NO_3–N loading to the estuary at SFB to annual average concentrations did not show any obvious correlations in NO_3–N concentrations and mass of annual loading.

Variation in the TKN concentration in the NR Estuary among zones and across years was less than for the DIN fractions. Figure 7 shows annual average TKN in the NR Estuary by zone and decade. In terms of the annual values used to derive the decadal averages, there were many years where TKN concentration was lowest in the Lower Estuary while concentrations similar to upstream zones occurred in other years (e.g., 2003–2006). Temporally, the TKN concentrations in the NR Estuary may have been lower in the late-1980s to early-1990s, reflected mainly in the 1990s averages, and then increased by the mid-1990s to ~0.6 ± 0.1 mg/l. Peak TKN concentrations, on average, occurred in the Middle Estuary, with annual average TKN of about 0.7 mg/l during 2001–2006. The increase in TKN in the Middle Estuary, without a corresponding increase in NH_4–N, would indicate accumulation of Org-N (e.g., algal biomass) in the zone through uptake of NO_3–N or internal sources of NH_4–N. As with other N fractions, there was no clear correspondence between years with high TKN loading and elevated TKN concentrations.

Total P concentrations decreased throughout the NR Estuary in the 1990s following the P controls enacted in 1988, as has been documented in a number of prior analyses (e.g., Stow and Borsuk 2003; Paerl and others 2004). Spatially, TP concentration was highest in the River zone and decreased to minimum values in the Lower Estuary. In terms of response to P controls, River and Lower Estuary average TP concentration decreased from 0.24 and 0.15 mg/l, respectively, in the 1980s to 0.15 and 0.09 mg/l in the 1990s, with intermediate concentrations in the Upper and Middle Estuary. For the 2000s, TP concentration decreased further to 0.12 mg/l in the River zone to 0.07 mg/l in the Lower Estuary.

Estuarine Chlorophyll *a* Pattern

Annual average Chl*a* values varied considerably by year, with the highest values typically occurring in the Middle and Lower Estuary (Fig. 8). From a temporal perspective, Chl*a* concentrations in the River and Upper Estuary zones decreased in the same time-frame as P and N controls in the Neuse River watershed (P controls in 1988 mainly affecting the River Zone and N controls beginning in the late 1990s affecting Upper Estuary Chl*a*). Otherwise, it appears that the elevated loading of TN, with a substantial portion as Org-N, may have contributed to higher availability of N during 2000–2009 and higher annual average Chl*a* values, although there was considerable variability among years. Recent studies have shown that a significant fraction of watershed-derived Org-N is assimilated by estuarine and

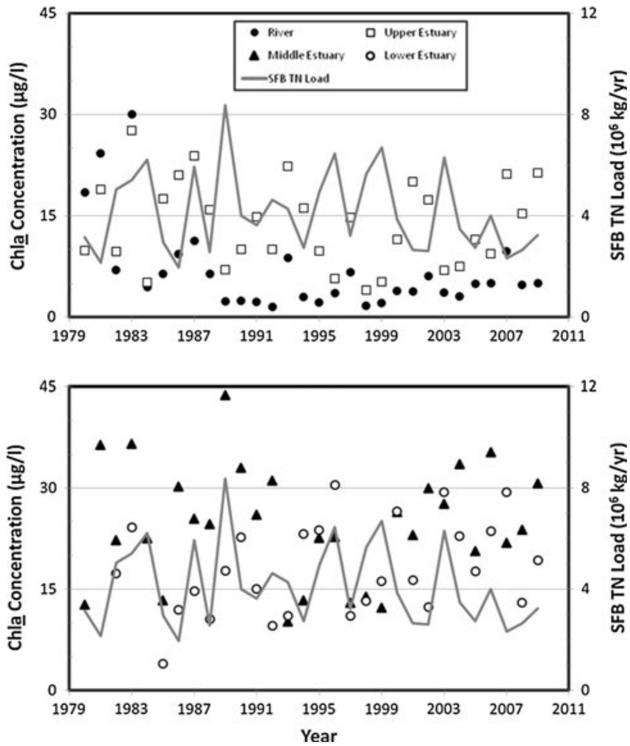

Fig. 8 Annual mean Chl*a* by zone in NR Estuary compared with TN loading for NR at SFB

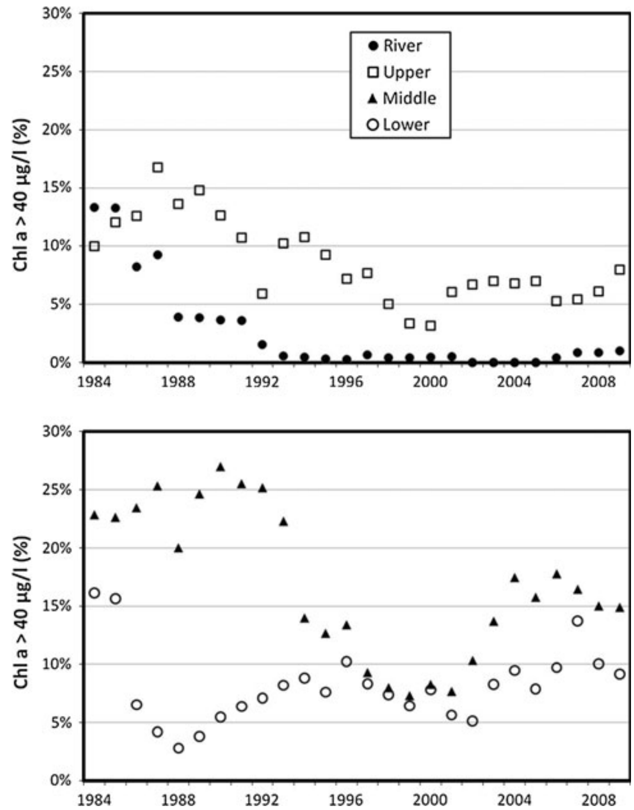

Fig. 9 Cumulative frequency of Chl*a* greater than 40 μg/l standard by 5-year assessment period and zone along NR Estuary. Values are plotted in year 5 of each period

coastal phytoplankton (Bradley and others 2010a, b) and, hence, should be included in the N fractions evaluated as part of estuarine eutrophication assessment. In contrast to high loading years, low flow years would have reduced TN loading, and an upstream migration of salt water would shift the zone of maximum algal production to the Upper Estuary. Consistent with this predicted change, 4 of the 5 years with average Chl*a* <15 μg/l in the Middle Estuary were low flow years (1980, 1985, 1994, and 1997).

In terms of compliance with the Chl*a* standard for the NR Estuary, it is the frequency of large algal blooms with measured photic zone average Chl*a* concentrations greater than 40 μg/l that is the key metric to evaluate. Figure 9 plots changes in the fraction of data points in each zone greater than the 40 μg/l for rolling 5-year periods (i.e., 1984 = 1980–1984). Frequency of large blooms clearly has decreased in the River and Upper Estuary zones, with cumulative 5-year frequencies <10% since 1986 and 1995, respectively. In contrast, frequency of Chl*a* >40 μg/l have been 13.7–17.8 and 7.9–13.7%, respectively, for the Middle and Lower Estuary for 5-year periods ending 2003 through 2009. One difference between the Middle and Lower Estuary zones is the lower frequency of large algal blooms in the Lower Estuary than in the Middle Estuary, with a number of 5-year periods with a frequency <10%.

Interpreting Progress

Accurate assessment of TMDL management action effectiveness requires a comprehensive analysis over several years to a decade or more. The time factor needed is affected by a number of confounding factors such as in-stream transformations among forms, potential for time lags for decreased inputs to the stream network, settling and resuspension of particle-bound nutrients, and sediment release of regenerated NH_4–N and PO_4–P (e.g., United States Environmental Protection Agency 2001; Meals and others 2010). The approach described in this study provides an effective means to evaluate long-term progress toward achieving water quality goals. For assessments associated with TMDLs, it is critical to provide managers with a reasonable expectation for when improvements in nutrient concentrations and eutrophication symptoms can be expected. In defining a reasonable expectation, potential continued impacts from past inputs or practices on existing conditions should be taken into account. For example, algal biomass and production continued to be elevated in the lower NR Estuary for several years following the severe flooding in 1999 associated with three major tropical storms (Christian and others 2004; Paerl and others 2006b).

The NC Division of Water Quality has recently reviewed progress in the implementation of management actions required under the Neuse Estuary TN TMDL (NCDWQ 2009). For point sources, NPDES compliance data showed an overall decrease of 65% in projected TN delivered to the NR Estuary from point sources by 2006 compared with the established 1995 benchmark, reflected in an approximately 50% decrease in NO_3–N below Raleigh for the flow-adjusted trend (i.e., Fig. 5a). Conversely, TSS and turbidity data below Raleigh show an overall increase in recent years, reflected in higher TKN concentrations. The general pattern of constant or increased TKN concentration at all watershed locations evaluated, particularly under high flows, indicates actions to date may not have collectively addressed Org-N inputs. It is also possible that excessive loading of eroded soil to the stream network during the large hurricane events in 1996 and 1999 have contributed to ongoing TKN loading to estuarine waters. Overall, the lack of instream improvement for high flow conditions contradicts estimated reductions of 45% in TN export from agricultural lands and implementation of urban stormwater plans in many areas of the basin (NCDWQ 2009). Additional evaluation is needed on sources of NO_3–N and Org-N under high flow conditions as well as effectiveness of implemented controls on agricultural lands and for urban stormwater. For example, a portion of the N reduction credit for agricultural land is based on an inferred efficiency of best management practices (BMPs), such as vegetated buffers and controlled drainage, in reducing TN export. It will be important in future evaluations to determine whether the current lack of instream progress is due to time lags between implementation of BMPs and stream benefits (e.g., Meals and others 2010) or to insufficient controls on TN inputs, particularly for sources important under high flow conditions. Application of trend analysis at additional monitoring locations representing predominantly different land uses may also help to provide insight on effectiveness of actions implemented to date.

Observed trends in concentrations of N fractions at NR watershed stations were only partially translated to decreased concentrations in downstream estuarine waters. Decreased NO_3–N concentrations along the mainstem of the NR from the Clayton station below Raleigh downstream to SFB were also observed in the Upper Estuary. However, average NO_3–N concentrations in the Middle and Lower Estuary were actually higher in the 2000s than in the 1990s before the TMDL was established. This apparent contradiction in temporal patterns may reflect regeneration of particle-bound N deposited in the Middle and Lower Estuary of the NR following periods of high flow associated with moderate to large storm events. A potential link between particle-bound N input and increased NO_3–N within the photic zone of the Middle Estuary is supported by increased TKN loading at SFB in the 2000s, by high rates of NH_4–N release from sediments in the Neuse Estuary (e.g., Fisher and others 1982), and reported increased NH_4–N concentrations in nearshore areas along the Middle Estuary (Burkholder and others 2006) during the same time period as increased TKN loading. The observed increase in NH_4–N in the Upper Estuary compared to River stations also supports the importance of regenerated N from sediments. This potential link between loading of particle-bound N and increased NO_3–N concentrations (after nitrification) in the Middle Estuary has implications for management of eutrophication in the NR and merits additional investigation. Ultimately, successful implementation of the TMDL and management of eutrophication in the Neuse Estuary will involve controlling sources of N contributing to elevated algal production in estuarine waters across the full range of hydrologic conditions.

Conclusions

Restoration of impaired waters typically involves implementation of a variety of actions targeting decreased loading of the pollutant of concern. The relatively simple analytical approach described here and applied to nutrient concentrations in the Neuse River watershed provided essential feedback for managers without extensive statistical manipulations. Further, applying the approach to different N fractions in the case example showed there was progress achieved in reducing NO_3–N inputs but not TKN, particularly under high flow conditions. The information provided by the analysis has the promise to help guide evaluation of potential input sources or spatial areas throughout a given watershed where further evaluation or management actions are needed. This is true for the Neuse River watershed or any other system to which the approach may be used to evaluate trends in flow-impacted pollutants.

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