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Recent trends in nutrient and sediment loading to coastal areas of the conterminous U.S.: Insights and global context

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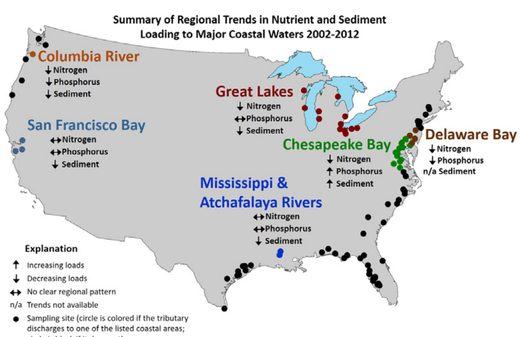
HIGHLIGHTS

- Examined decadal trends in riverine nutrient and sediment loads to coastal waters
- N loading decreased at more coastal sites than P loading, especially in urban areas.
- Nutrient loading from undeveloped watersheds was low but increased 2002–2012.
- N was often elevated relative to P, despite recent decreases.
- Additional N and P reductions in coastal rivers would benefit coastal ecosystems.

GRAPHICAL ABSTRACT



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ABSTRACT

Coastal areas in the U.S. and worldwide have experienced massive population and land-use changes contributing to significant degradation of coastal ecosystems. Excess nutrient pollution causes coastal ecosystem degradation, and both regulatory and management efforts have targeted reducing nutrient and sediment loading to coastal rivers. Decadal trends in flow-normalized nutrient and sediment loads were determined for 95 monitoring locations on 88 U.S. coastal rivers, including tributaries of the Great Lakes, between 2002 and 2012 for nitrogen (N), phosphorus (P), and sediment. N and P loading from urban watersheds generally decreased between 2002 and 2012. In contrast, N and P trends in agricultural watersheds were variable indicating uneven progress in decreasing nutrient loading. Coherent decreases in N loading from agricultural watersheds occurred in the Lake Erie basin, but limited benefit is expected from these changes because P is the primary driver of degradation in the lake. Nutrient loading from undeveloped watersheds was low, but increased between 2002 and 2012, possibly indicating degradation of coastal watersheds that are minimally affected by human activities. Regional differences in trends were evident, with stable nutrient loads from the Mississippi River to the Gulf of Mexico, but commonly decreasing N loads and increasing P loads in Chesapeake Bay. Compared to global rivers, coastal rivers of the conterminous U.S have somewhat lower TN yields and slightly higher TP yields, but similarities exist among land use, nutrient sources, and changes in nutrient loads. Despite widespread decreases in N loading in coastal watersheds, recent N:P ratios remained elevated compared to historic values in many areas. Additional progress in reducing N and P loading to U.S. coastal waters, particularly outside of urban areas, would benefit coastal ecosystems.

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1. Introduction

Coastal areas in the U.S. and worldwide have experienced massive changes in population, land use, and inputs from interior continental sources (Seitzinger et al., 2010). Between 1970 and 2010, the population in U.S. coastal areas increased by 45% to 163.8 million people, comprising just over half of the U.S. population (National Oceanic and Atmospheric Administration, 2013). By 2020, the coastal population is expected to increase by another 8%. Further development may increase eutrophication and its associated symptoms (Bricker et al., 2007). Changes to land use and water quality in watersheds adjacent to coastal areas are expected to have the largest and most direct effects on coastal ecosystems (National Oceanic and Atmospheric Administration, 2013).

Development in coastal watersheds has resulted in significant degradation of estuaries and coastal ecosystems through numerous interacting factors including habitat loss, nutrient pollution, and changes to sediment delivery (Carpenter et al., 1998; Foley, 2005; Lotze, 2006; Mallin et al., 2000; Seitzinger et al., 2010; U.S. Environmental Protection Agency, 2016; Vitousek et al., 2009). Population growth and increased human activity in coastal areas have increased nutrient inputs to coastal waters through higher volumes of agricultural and urban nonpoint-pollution, higher demand for wastewater treatment, and consumption of fossil fuels (Bricker et al., 2007; U.S. Environmental Protection Agency, 2016). Sediment loading to estuaries is complicated and possible negative consequences can result from either too much or too little. Land clearing and disturbance increases erosion and can lead to enhanced coastal loading (Dauer et al., 2000; Syvitski, 2005; Thrush et al., 2004) while hydrologic modification from the development of dam and reservoir systems can decrease sediment loads (Meade and Moody, 2009). Increases in sediment loads reduce water clarity and contribute to the loss of submerged aquatic vegetation (Bricker et al., 2007; Orth et al., 2006; Waycott et al., 2009). However, sufficient sediment loading is necessary to sustain coastal wetlands and estuaries which provide critical habitat for wildlife and fisheries and serve as buffers against storms and sea-level rise (Bricker et al., 2007; National Research Council, 2000; U.S. Environmental Protection Agency, 2016).

Eutrophication through excess nutrient pollution is a primary driver of coastal ecosystem degradation (Bricker et al., 1999; National Research Council, 2000). Widespread coastal eutrophication has been well-documented in scientific literature (Boyer et al., 2006; Cai et al., 2011; Carpenter et al., 1998; Diaz and Rosenberg, 2008; Howarth et al., 2011). Two previous national assessments found that approximately two-thirds of the coastal waters in the U.S. are moderately or severely degraded from nitrogen (N) pollution, which is causing extensive eutrophication and associated effects (Bricker et al., 2007, 1999). Globally, N pollution has increased due to the increased creation of reactive N for fertilizer use and, inadvertently, from the combustion of fossil fuels (Galloway et al., 2004). In coastal waters, N pollution is greatest where agricultural activity and urbanization are greatest (Howarth and Marino, 2006). For the U.S., human activity likely has increased N loads to the coast by approximately 4 to 8-fold (Howarth et al., 2002). Nonpoint sources are the dominant inputs of N and phosphorus (P) to most U.S. surface waters, but in urban areas, while diffuse sources affect a larger number of streams, point sources contribute >50% of the N and P mass reaching rivers (Carpenter et al., 1998; Preston et al., 2011). P tends to be elevated in urban areas due to point-source sewage inputs and because of fertilizer and manure usage in agricultural areas (U.S. Environmental Protection Agency, 2016). N is the largest pollution problem in U.S. coastal waters and one of the greatest threats to the ecological functioning of these ecosystems (Howarth et al., 2000; National Research Council, 2000) although elevated P concentrations in nearshore coastal waters are common (U.S. Environmental Protection Agency, 2016).

Relatively few national-scale assessments have addressed the changes in water quality in rivers and streams that flow to coastal waters. A comprehensive assessment of nutrient enrichment and eutrophic conditions in U.S. estuaries was published by National Oceanic and Atmospheric Administration (NOAA) in 1999 (Bricker et al., 1999), followed by a similar assessment in 2007 (Bricker et al., 2007) that included information about N and P loading to estuaries. However, assessment of the change in eutrophic condition for individual estuaries in the second assessment was impeded by reduced reporting and a change in the data collection method (Bricker et al., 2007). The U.S. Environmental Protection Agency (USEPA) has conducted regular assessments of coastal marine waters and the results are published in four National Coastal Condition Reports (NCCR) that assess the overall condition of U.S. coastal waters, but do not provide individual estuary results. (U.S. Environmental Protection Agency, 2012). In 2010 and 2015, the USEPA conducted National Coastal Condition Assessments (NCCA) using a statistical survey of U.S. nearshore coastal waters during the summer and concluded that P was the most widespread stressor to nearshore water quality (U.S. Environmental Protection Agency, 2016).

Riverine inputs dominate nutrient loading to coastal waters (National Research Council, 2000; Sharples et al., 2017). Therefore, determining the trends in nutrient and sediment loading from streams is critical to understanding whether progress is being made in reducing nutrient loads and whether differences in trends are evident between sites with different land use and nutrient sources.

This study focuses on recent trends in nutrient and sediment loads in rivers and streams that flow to coastal waters of the conterminous U.S., including the Great Lakes. We focused on results from individual sites, rather than aggregated regional fluxes, to better understand where nutrient and sediment loads were increasing or decreasing as well as the land-use factors contributing to those changes. Understanding national trends in surface-water quality is a primary goal of the U.S. Geological Survey's (USGS) National Water Quality Assessment project (NAWQA). For the first time, in an effort meant to be as inclusive as possible, data from multiple sources were aggregated, screened, standardized, and used to support a comprehensive assessment of surface-water-quality trends in the U.S. (De Cicco et al., 2017; Oelsner et al., 2017). Coastal sites were selected from this larger trend project to describe the trends in nutrient and sediment loads in rivers and streams draining to coastal waters. This analysis allows us to address recent (2002−2012) trends in nutrient and sediment loadings to U.S. coastal waters. We also explore regional differences in nutrient and sediment loadings as well as compare trends in nutrient and sediment loading among land-use categories. To provide a broader context for these recent trends, we compared estimates of total nitrogen (TN) and total phosphorus (TP) yield between 1972 and 2012 for the larger rivers in this study to provide a longer-term perspective and compared the TN and TP yield estimates to values for rivers outside of the conterminous U.S. to provide a global perspective. By examining the long-term data compiled for this study, we can address whether progress has been made in reducing nutrients compared to historic values.

2. Methods

2.1. Data sources and handling

2.1.1. Water-quality data

Water-quality data (ammonium (NH4-N), nitrate (NO3-N), TN, soluble reactive phosphorus (SRP), TP, total suspended solids (TSS), and suspended sediment concentration (SSC)) were acquired from multiple national, state, and local sources. Because the data originated from a variety of sources, it was necessary to harmonize parameter names, sample fraction, reporting units and speciation, and remark codes across all sources. NO3-N includes samples collected and analyzed for NO_3^- as well as $NO_3^- + NO_2^-$ because these samples were shown to be equivalent for the purposes of this study (Oelsner et al., 2017). In other parts of the larger trend study (De Cicco et al., 2017; Oelsner et al., 2017), SRP is referred to as orthophosphate and the two terms can be considered interchangeable for the purposes of this analysis. During the data screening and trend analysis, filtered and unfiltered SRP data were treated as separate parameters. However, since there were only two coastal sites with unfiltered SRP trends, the results were combined and called "SRP" in the results and figures of this manuscript. Data from these sources were used in two ways: (1) a trend analysis of water quality in coastal rivers; and, (2) an analysis of molar TN:TP ratios in historic (pre-1979) and recent (2002–2012) samples.

Trends presented in this study were determined for four periods between 1972 and 2012, (1) 1972–2012, (2) 1982–2012, (3) 1992–2012, and (4) 2002–2012 as data allowed. For the trend analysis, sites were required to have at least one sample per quarter per year (with a flexible quarterly distribution as described in Oelsner et al. (2017)) in the first two and last two years in the trend period. The beginning year and ending year in the trend analysis were allowed to vary by one year such that the start year could be 2002 or 2003 and end year could be 2011 or 2012. Additionally, sites were required to have at least quarterly samples in 70% of the years in the trend period (i.e. 7 of 10 years); longer gaps were allowed for the 30 and 40-year trend periods. Sites were also required to have a representative number of samples collected during high-flow periods as described in Oelsner et al. (2017). All sites that passed the screening criteria were included in the initial trend analysis. Since no sites in Alaska or Hawaii passed the screening criteria, the trend analysis pertains only to rivers in the conterminous U.S. The criteria, guided by sensitivity analyses, are described in greater detail in Oelsner et al. (2017). The screened and finalized data were used in the subsequent trend analyses presented in this manuscript. Input data, trend models, and trend results were published separately (De Cicco et al., 2017).

Although SSC and TSS are the two predominant measures of suspended sediment in rivers and streams, the analytical methods are not comparable and should not be used interchangeably (Gray et al., 2000). SSC uses the dry weight of the suspended sediment load in the entire water sample, whereas TSS uses the dry weight of suspended sediment from a subsample. In a previous study, SSC was found to be a more reliable measure of suspended sediment in natural waters, especially when >25% of the suspended sediment was sand-sized (Gray et al., 2000). We present trend results for both SSC and TSS as measures of sediment loading to coastal waters, but the results are not directly comparable and are not expected to always agree.

2.1.2. Discharge data

Daily stream discharge data, which is required for the trend analysis, originated primarily from the USGS streamgage network and was obtained from the USGS National Water Information System (NWIS) (https://waterdata.usgs.gov/nwis). Five sites used discharge data from non-USGS sources: the U.S. Army Corps of Engineers ($n = 2$), Oregon Water Resources Department ($n = 2$), and the International Boundary and Water Commission ($n = 1$). In most cases the monitoring site and streamgage were co-located. Otherwise, the monitoring site was paired with a nearby streamgage when the difference in respective watershed areas was within 10% and there were no major inputs between the streamgage and the monitoring site. Detailed descriptions of the gage matching routines are given in Oelsner et al. (2017).

2.2. Trend analysis

Trend analysis was performed on the screened data using the Weighted Regressions on Time, Discharge, and Seasons (WRTDS) model (Hirsch et al., 2010; Hirsch and De Cicco, 2015). The model was implemented using the R packages EGRET (version 2.2.0) and EGRETci (version 1.0.4) to produce trend results and the associated likelihood analysis (Hirsch et al., 2015).

The WRTDS model estimates concentration (c) for every day of the period of record at each site as

$$\ln(c) = \beta_0 + \beta_1 t + \beta_2 \ln(Q) + \beta_3 \sin(2\pi t) + \beta_4 \cos(2\pi t) + \varepsilon \qquad (1)$$

where ln is natural log, βi are fitted coefficients, Q is daily mean discharge, t is decimal time, and ε is the unexplained variation. The fitted coefficients in the WRTDS model are not the same throughout the entire domain of the data; a unique set of coefficients for each day and discharge are included in the model (Q_i, t_i). The observations used to calibrate each model are selected using half-window widths. Half-window widths of 7 years and 1/2 year were used for the time and seasonal dimensions, respectively, and 1 or 2 natural log units were used for the discharge dimension for sites with drainage areas greater or <250,000 km^2, respectively (Oelsner et al., 2017). The weights on each observation (Q_i, t_i) used in each calibration are based on their similarity in terms of time, season, and discharge to the day being calibrated (Q_o, t_o). This process results in fitted coefficients that vary over the period of record which minimizes bias in estimates of daily concentration and load. Estimates of daily concentration are multiplied by the respective daily mean discharge to estimate daily load.

WRTDS also allows the effects of random and systematic variability in discharge to be accounted for through a process called flow normalization. Flow-normalization was implemented using the assumption of a stationary discharge regime over the period of record and removes the variation in concentration and load that is due to random and systematic variations in discharge but retains the influence of nonrandom seasonal variations. Flow-normalized (FN) estimates of concentration and load are produced as estimates of concentration and load that occur at the mean discharge for each calendar day of the year over the trend period. Basing FN estimates on a single mean value for discharge embeds an assumption of stationary flow (no trends). Thus, temporal changes in the FN estimates show changes in water quality apart from random and systematic changes in discharge and serve as an indicator of non-discharge related changes occurring in the watershed, often related to human actions and decisions. This approach serves as a better indicator of the effect of changes in the watershed on water quality as opposed to an approach that does not explicitly account for different types of discharge variability. Based on these advantages, FN load estimates and trends determined by the WRTDS model assuming stationarity in the discharge regime are presented in this manuscript.

For the trend analyses presented here, daily FN loads were aggregated to water-year (October 1 through September 30) means. Trends in annual mean FN load were calculated as the net change (in percentage) between the start and end of the trend period. The confidence intervals and associated significance level of the trend were determined through a block bootstrap approach based on a time interval of 200 days to avoid oversampling any of the more densely sampled periods during the overall period of record and to broadly maintain samples from individual high or low discharge events (Hirsch et al., 2015). The bootstrap method was used to generate a 90% confidence interval on the magnitude of the trend (calculated as the net change in load in percentage) and a likelihood statistic that is the functional equivalent to the two-sided p-value. The likelihood statistic provides information on whether the null hypothesis that there is no trend over the period of record should be rejected and provides a measure of the strength of evidence that the trend is occurring. Models with >0.7 likelihood that net change in load (in percentage) was either positive or negative were considered to have evidence for a trend, whereas models with likelihoods <0.7 were considered as likely to have an upward trend as a downward trend. More detail on the bootstrapping method and trend likelihood is given in Hirsch et al. (2015). Figures presenting the original concentration data used in the model, the seasonal distribution of the concentrations, the observed and estimated concentration and flux values from the model, and model predictions of the annual mean concentration and flux and flow-normalized annual mean concentration and flux along with the 90% confidence intervals are available in the online mapper (https://nawqatrends.wim.usgs.gov/swtrends/) and the underlying data are available in De Cicco et al. (2017).

Model output was checked visually for fit, residual structure, and residual bias according to the procedures described in Hirsch and De Cicco (2015) and problematic models were excluded from the study. Including all the trend models reviewed as part of the Oelsner et al. (2017) study, not just models from coastal sites, approximately 22% of the nutrient models and 54% of the sediment models were rejected (Oelsner et al., 2017). Additional details regarding trend model review procedures are described in Oelsner et al. (2017) along with a complete list of rejected models.

Trend results were used in two ways in this study. First, FN trend results from the most recent trend period (2002–2012) were presented for rivers and streams that discharge to an ocean or one of the Great Lakes to understand the recent trends in nutrient and sediment loading to coastal waters of the conterminous U.S. Based on the large range of watershed sizes (Table S1) and load estimates, yield and percent change were used to compare trends in loads across sites. Yield was calculated by dividing the FN load estimates by the watershed area and is expressed in kg ha^{-1} yr^{-1}. The percent change in FN load is equivalent to the percent change in FN yield. To determine whether water quality was improving in areas with high nutrient and sediment loads, we compared FN estimates of yield in 2002 to the percent change in yield between 2002 and 2012. Second, trend model estimates of FN TN yield and FN TP yield from the start of each trend period (1972, 1982, 1992, and 2002) and from 2012 were used to provide a longer-term context to how TN and TP yields have changed over time in medium to large coastal rivers (annual mean discharge >30 cms or watershed area >1500 km^2) of the conterminous U.S.

2.3. Site selection

Coastal sites included in this study were selected from the larger group of national nutrient and sediment trend sites that passed both trend screening criteria and subsequent model quality checks (described in Sections 2.1.1 and 2.2 and in Oelsner et al. (2017)) based on several criteria including stream level, proximity to coastal waters, intervening influences, and number of available trends. First, stream level information from the NHDPlus was used to determine trend sites on streams that drain to coastal waters. Stream level is a numbering system in the NHDPlus based on the hierarchy of streams from the mouth (the reverse of stream order) (McKay et al., 2012); stream reaches with a stream level of 1 empty into the ocean (or Great Lake) and streams with a stream level of 2 are tributaries to stream level 1 streams. For this study, all trend sites on stream level 1 reaches were initially included. Sites on stream level 2 or 3 reaches were initially included if they were major streams that ultimately drained to coastal waters or entered a lower-level stream downstream of a trend site. For example, the Appomattox River was included in the study, but it has a stream level of 2 because it flows into the James River before entering Chesapeake Bay. Sites on higher level streams (>2) were excluded if they were upstream of another trend site on a lower level stream. Second, sites were excluded if the straight-line distance to the coast exceeded 160 km. Additionally, trend analysis methods do not work with positive and negative discharge values, so the trend sites need to be located above the head-of-tide, which is often not close to the coast. Occasionally, there were multiple trend sites along the same stream or river. In these cases, sites closest to the coast were selected except when an upstream site had trends for more parameters. Sites were excluded if they could not be determined to drain into an estuary, ocean, or one of the Great Lakes. Additionally, sites upstream of lakes and reservoirs were excluded because the nutrient and sediment loads would not be representative of loads entering coastal waters. The Colorado River (Arizona) trend site was not included in this analysis for two reasons (1) the Colorado River only reaches the Sea of Cortez during unusually wet periods or under special flood experiments (Jarchow et al., 2017) and (2) the water quality monitored at the Northern International Boundary is over 220 km away from the Sea of Cortez and is not representative of the water reaching coastal waters on rare occasions. In most cases, we only included one site from each coastal river or stream. However, trend results from two sites on the same stream were both included when the two unique sites were on the same stream reach and had no intervening influences such that water quality at the two sites could be assumed reasonably comparable. From the larger set of national trend results, we identified 95 trend sites from 88 streams and rivers (and watersheds) in coastal areas with recent (2002–2012) nutrient and sediment trends that are included in this study (Fig. S1, Table S1).

From the 95 recent (2002–2012) coastal trend sites available for this study, there were 295 nutrient and sediment trend results. The number and direction of recent coastal trends included in this study varied among parameter groups (N, P, and sediment). Trends in N parameters (NH4-N, NO3-N, and TN) composed more than half (161) of the trends included in this study, followed by P parameters (SRP and TP, 88), and sediment (SSC and TSS, 46). The number of trends for each parameter for the recent trend period (2002–2012) are provided in Table S2 and trend results for all sites are reported in Table S3.

Average annual gage-adjusted discharge values for each site and stream level 1 reach (the QE_MA value from NHDPlus) (McKay et al., 2012) were used to determine the percentage of flow to coastal waters represented by the trend sites. Average annual discharge values from the most downstream reach of each stream level 1 stream in the conterminous U.S. were summed to determine the total annual average discharge from the conterminous U.S. to coastal waters. Because we include the Great Lakes as coastal waters in this study, we subtracted the average annual discharge for the Detroit, Niagara, and St. Lawrence rivers from the total value because they represent water transfers between or out of the Great Lakes. The proportion of discharge to the coastal waters of the conterminous U.S. represented by the trend sites was calculated by dividing the sum of the average annual discharge from the trend sites by the sum of the average annual discharge from all stream level 1 reaches.

2.4. Regional and land-use categories

Coastal trend sites were categorized into five regions similar to the regional designations used by NOAA (Fig. S1, Table S1) (Bricker et al., 2007). There were two regional categories on the East Coast of the U.S. Sites located north of the Virginia/North Carolina state boundary (designated by the dashed line extending from the East coast) were grouped into the North and Middle Atlantic (North/Mid-Atlantic) region and sites south of this boundary were grouped into the South Atlantic region. Sites along the Gulf of Mexico (the states of Florida, Louisiana, and Texas) were grouped into the Gulf region. All the sites on the West Coast of the U.S. were categorized into the Pacific region, and all the sites in the northern U.S. around one of the Great Lakes were grouped into the Great Lakes region.

We also examined the distribution of land use in each of the regions to determine whether the observed regional patterns in nutrient and sediment trends could be a function of land use or whether geographic regions were dominated by a single land use. Land use in each watershed was classified using percentages of land use from the NAWQA Wall-to-Wall Anthropogenic Land Use Trends (NWALT) dataset (Falcone, 2015). Watersheds were categorized by predominant land use into four classes, based on 2012 land use in the NWALT data set (definitions in Table S4 and see Falcone (2015) for detailed land-use class descriptions). The four broad classes were undeveloped, agricultural, urban, and mixed. “Undeveloped” watersheds were defined as those with high percentages of natural vegetation and low anthropogenic use, “agricultural” watersheds were defined as those that were not undeveloped, had relatively high percentages of agriculture, and low percentages of urbanization. "Urban" watersheds were defined as those that had relatively high percentages of urbanization and relatively low percentages of agriculture but were not undeveloped nor agricultural. "Mixed" watersheds were those which had a mix of land uses not falling into any of the other three categories. Land-use categories were not assigned to the largest rivers and watersheds (Mississippi River, Atchafalaya River, Columbia River, and Rio Grande) because aggregated land use at such large scales is not a useful predictor of water quality in these large, heterogeneous watersheds. Instead, these 4 large rivers with watershed areas >200,000 km^2 are shown separately in figures with land-use categories as "Large Rivers."

Only small changes in urban and agricultural land use occurred between 2002 and 2012 at the coastal sites based on the NWALT dataset (Falcone, 2015). The change in the percentage of agricultural land use in the individual watersheds between 2002 and 2012 ranged from 1% to −4% at all but one site, the Little Manatee River near Wimauma, FL, where the percentage of agricultural land use in the watershed was 7% lower in 2012. The change in the percentage of developed land use in the individual watersheds between 2002 and 2012 ranged from 0% to 6%.

A majority of the trend watersheds were classified as having urban land use in the North/Mid-Atlantic and South Atlantic regions (Fig. S2a). In the Pacific region, a majority of the trend watersheds were classified as "undeveloped" and the North/Mid-Atlantic region had a similar number of "undeveloped" watersheds, although a smaller percentage (Fig. S2b). None of the regions had many watersheds with agricultural land use, but the majority occurred in the Great Lakes and Gulf regions (Fig. S2b). The Gulf region had the largest number of watersheds with mixed land use, followed by the North/Mid-Atlantic region, and the South Atlantic region (Fig. S2b). Notably, 3 of the "Large River" watersheds (Mississippi River, Atchafalaya River, and the Rio Grande) are in the Gulf region.

Densities of housing, population, and major wastewater facilities as well as N and P loading from manure and fertilizer in the trend watersheds were compared across the land-use categories to examine how the land-use categories represented these factors influencing water quality based on datasets in Falcone (2017). In general, the urban watersheds had the highest population and housing densities while undeveloped watersheds had the lowest population and housing densities (Fig. S3). Urban watersheds had the highest density of major wastewater facilities (Fig. S4) and agricultural watersheds received the highest amounts of N and P from fertilizer and manure (Fig. S5). Based on these comparisons, we determined that the land-use categories were useful for comparing trend results and their potential causes.

2.5. Coastal estuaries and eutrophication susceptibility

In order to examine the influence of water-quality trends on estuaries, each site was assigned to its respective terminal coastal waters or estuary based on the NOAA Coastal Assessment Framework (CAF) (National Oceanic and Atmospheric Administration, 2007). The CAF is primarily composed of Estuarine (and sub-estuarine) Drainage Areas (EDAs), Fluvial Drainage Areas (FDAs), and Coastal Drainage Areas (CDAs). Estuary assignments were completed at the sub-estuary level (the EDASUBEDA variable in the CAF dataset). The susceptibility of each estuary to eutrophication was based on the "Influencing Factors" designation in the National Estuarine Eutrophication Assessment (NEEA) Update (Bricker et al., 2007). After manually checking the assignments, several adjustments were made to better assess the influence of the trends on estuary water quality. The Susquehanna River sites and the two tributaries that drain to the Patapsco River were assigned eutrophication influencing factor levels for the Chesapeake Bay Mainstem estuary. The estuary name, code, and the "Influencing Factors" rating was assigned to each site based on its associated estuary (Table S5). Trend results were grouped by the susceptibility of estuaries to eutrophication, the "Influencing Factors" rating, to understand how the water-quality trends may affect eutrophication conditions in estuaries.

2.6. Global TN and TP yield estimates

We conducted a literature review to compile recent (>2000) estimates of TN and TP yields in coastal rivers outside of the conterminous U.S. An effort was made to find estimates based on monitoring data rather than regional models. Six sources were used in this manuscript. Estimates of TN and TP yields primarily derived from monitoring data included HELCOM (2018) for the Baltic Sea, and Romero et al. (2013) for Southwest Europe. Estimates of dissolved inorganic nitrogen (DIN) and SRP yields using recent concentration data for 6 rivers in China and one river in Korea were published in Liu et al. (2009). Modeled estimates for TN and TP yields based on the Global NEWS 2 model were included for 10 river basins that drain to the Bay of Bengal (BOBLME, 2014) and 6 additional global rivers with both dissolved and particulate N and P estimates (Beusen et al., 2005; Mayorga et al., 2010). All published yield estimates were converted to kg N or P ha^{-1} yr^{-1}.

2.7. N:P calculations

In order to determine the extent to which the primary productivity of these coastal rivers was potentially limited by N or P and to put the short-term recent trends into a larger context, the N:P ratios were determined for each site using molar concentrations of TN and TP. In addition, this N:P ratio may shed light on the likelihood for N or P limitation in the receiving coastal waters, however the riverine N:P is not the determining factor for N or P limitation in the estuary, where physical conditions are also important factors. We used median concentration N:P ratios because load estimates from the trend analysis were not always available, particularly for the historic period. N:P ratios were calculated separately for each sample pair (unique date and time) at each site and then median N:P ratios were determined for each site for two time periods: historic (1965–1979) and recent (2002–2012). Because there were a limited number of sites with both TN and TP trends, N:P ratios were calculated using TN and TP concentrations at any site included in this study, even if sufficient TN and TP data for trend analysis were not available. All concentration data used in N:P calculations, but not in trend analysis, are available publicly though the USGS National Water Information System (NWIS) database (https://waterdata.usgs.gov/nwis), the USEPA STOrage and RETrieval Data Warehouse, Water Quality Exchange (STORET) (https://www.epa.gov/waterdata/water-quality-data-wqx), or the Texas Commission on Environmental Quality's Surface Water Quality Information System online database and the CRP Data Tool (www80.tceq.texas.gov/SwqmisWeb/public/crpweb.faces). The data source for TN and TP data for each river is provided in Table S6.

3. Results

All trend results are presented for the flow-normalized (FN) yields of N, P, and sediment.

3.1. Overall recent trends (2002–2012)

Nationally, a greater proportion of coastal sites had decreasing N (TN, NO3-N, NH4-N) loads than had decreasing P (TP, SRP) or sediment loads (Fig. 1). NH4-N loads decreased at the largest proportion of sites (31 sites, 67%) followed by NO3-N loads (36 sites, 59%), and TN loads (27 sites, 50%). Approximately 34% of the sites had decreasing SRP and TP loads. In fact, there were more sites with increasing SRP and TP loads than decreasing loads. The two sediment parameters, TSS and SSC, had substantially different proportions of sites with increasing and decreasing loads. SSC loads decreased at as many sites as they increased while TSS loads increased at more than double the number of

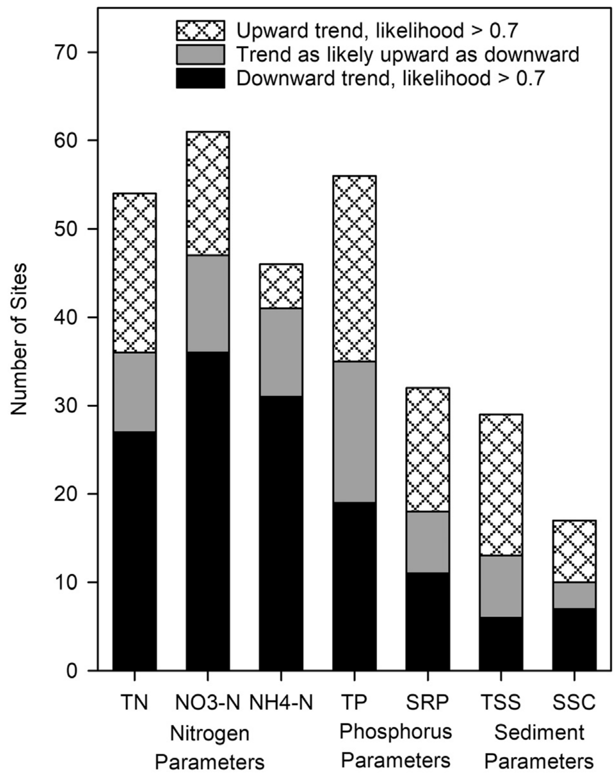

Fig. 1. Summary of the flow-normalized (FN) trends in load in coastal rivers and streams between 2002 and 2012. The number of coastal trend sites with increasing and decreasing FN trends in load (likelihood >0.7) and FN trends in load that are as likely to be upward as downward (likelihood <0.7) are presented for ammonium (NH4-N), nitrate (NO3-N), total nitrogen (TN), orthophosphate (SRP), total phosphorus (TP), total suspended solids (TSS), and suspended sediment concentration (SSC).

sites than they decreased. The proportion of sites with no measurable change in SSC or TSS ranged from 18% to 24%.

3.2. Recent regional trends (2002–2012)

There were regional differences in both the number of trends for each parameter and the trend directions (Fig. 2). The Gulf and North/Mid-Atlantic regions had the most trend sites (29 and 30, respectively) and trend results for all parameters (75 and 114, respectively), while the South Atlantic and Pacific regions had the fewest trend sites (13 and 10, respectively) and trend results for all parameters (30 and 26, respectively).

TN loads decreased at a majority of the sites in the North/Mid-Atlantic and Great Lakes regions, but only 30% of the sites in the Gulf region had decreasing TN loads and there were no decreasing TN loads in the South Atlantic region (Fig. 2a). Similar to TN loads, NO3-N loads decreased at a majority of sites in the North/Mid-Atlantic and Great Lakes regions while less than half of the NO3-N loads in the South Atlantic and Gulf regions decreased (Fig. 2b). Although NH4-N loads decreased at most sites across all regions, the North/Mid-Atlantic and Gulf regions were the only two regions where any NH4-N loads increased (Fig. 2c).

There were fewer regional differences between the P parameters. In each of the Great Lakes, North/Mid-Atlantic, and Pacific regions there were similar proportions of decreasing and increasing TP and SRP loads (Fig. 2d and e). However, in the Gulf region, there were more increasing TP and SRP loads than decreasing TP and SRP loads (Fig. 2d and e).

Sediment trends varied both regionally as well as between the two sediment parameters, SSC and TSS, which is not surprising given the differences in the parameters (Fig. 2f and g). The North/Mid-Atlantic region was the only region where TSS and SSC had a similar proportion of sites with increasing (67% and 55%, respectively) and decreasing (22% for both) loads. In the South Atlantic, 2 sites had increasing TSS loads and 3 sites had loads that were as likely to be increasing as decreasing, and the 1 SSC site had increasing loads. A majority of the TSS loads in the Gulf region were increasing while the two sites with SSC trends had decreasing loads. Unlike other regions with many increasing TSS loads, two-thirds of the TSS loads were decreasing in the Great Lakes region. The Pacific region had the largest difference between trend results for the sediment parameters. The three sites with TSS trends, located in OR and WA states, had increasing loads. SSC loads decreased at 2 of the 3 sites located in the San Francisco Bay area and in the Columbia River.

3.3. Recent changes in nutrient and sediment yield in relation to land use (2002–2012)

We examined how changes in nutrient and sediment loads were related to land use and initial conditions. For N parameters, many sites with the highest yields in 2002 also had decreasing yields over the recent trend period (2002–2012). The majority of sites with high TN yields in 2002 were agricultural sites and TN yield decreased at all of them (Fig. 3a). Notably, these agricultural sites were located within the Great Lakes Region (Fig. S6). Increases in TN yield mostly occurred at undeveloped sites with low TN yield and at agricultural sites with moderate TN yield. Similar to TN, four agricultural sites with high NO3-N yield in 2002 had decreasing NO3-N yields (−10% to −62%) and were located in the Great Lakes Region (Fig. S6). However, unlike TN, the three sites with the highest NO3-N yield in 2002 had very small changes in yield or increases in yield (Fig. 3b). Undeveloped sites had low NO3-N and NH4-N yields in 2002, and both increasing and decreasing yields. NH4-N yields decreased at the urban sites with the highest NH4-N yield in 2002, while NH4-N yield increased at several mixed land-use sites with low yield in 2002 (Fig. 3c). Changes in land use between 2002 and 2012 were minimal at most sites, but the one site where the percentage of agricultural land use in the watershed decreased by 7%, the Little Manatee River near Wimauma, FL, also had likely decreases in NH4-N yield (−13%) and NO3-N yield (−9.6%).

Relationships between trend results and initial yield were not as clear for TP as they were for TN. Decreasing TP yields were common among urban sites with high initial TP yields, but the pattern was not evident for other land-use types (Fig. 3d). Other than urban sites and large rivers, most sites in other land-use types experienced increasing TP yields, regardless of initial TP yield (Fig. 3d). SRP trends were similarly mixed with agricultural sites having more increases than decreases in yield and other land-use types having approximately equal numbers of both (Fig. 3e). Like TN, TP yields were comparatively low at undeveloped sites, but increased between 2002 and 2012.

Both SSC and TSS yields decreased at sites with the highest yields in 2002 (Fig. 3f and g). Initial SSC yield was highest at urban and agricultural sites. Increasing SSC yields occurred predominantly at sites with low yield in 2002 (Fig. 3f). In contrast, many of the sites with increasing TSS yields had mid-range yields in 2002 (Fig. 3g). Three of four agricultural sites had increasing SSC yields while the two undeveloped sites had mixed trend results. TSS yields increased at the six undeveloped sites, while agricultural and urban sites had mixed trend results (Fig. 3g). Interestingly, three large river sites (Mississippi, Atchafalaya, and Columbia rivers) all had similar decreases in SSC, between 27% and 32%.

3.4. Trend relationships between TN, NO3-N, and TP (2002–2012)

Trend results for TN were compared with results for either NO3-N or TP at the same sites to examine the extent to which component species reflected changes in total trends as well as to determine the relationship between TN and TP changes. For sites with both NO3-N (as N) and TN trends ($n = 47$), the trends in yield for the two parameters were similar

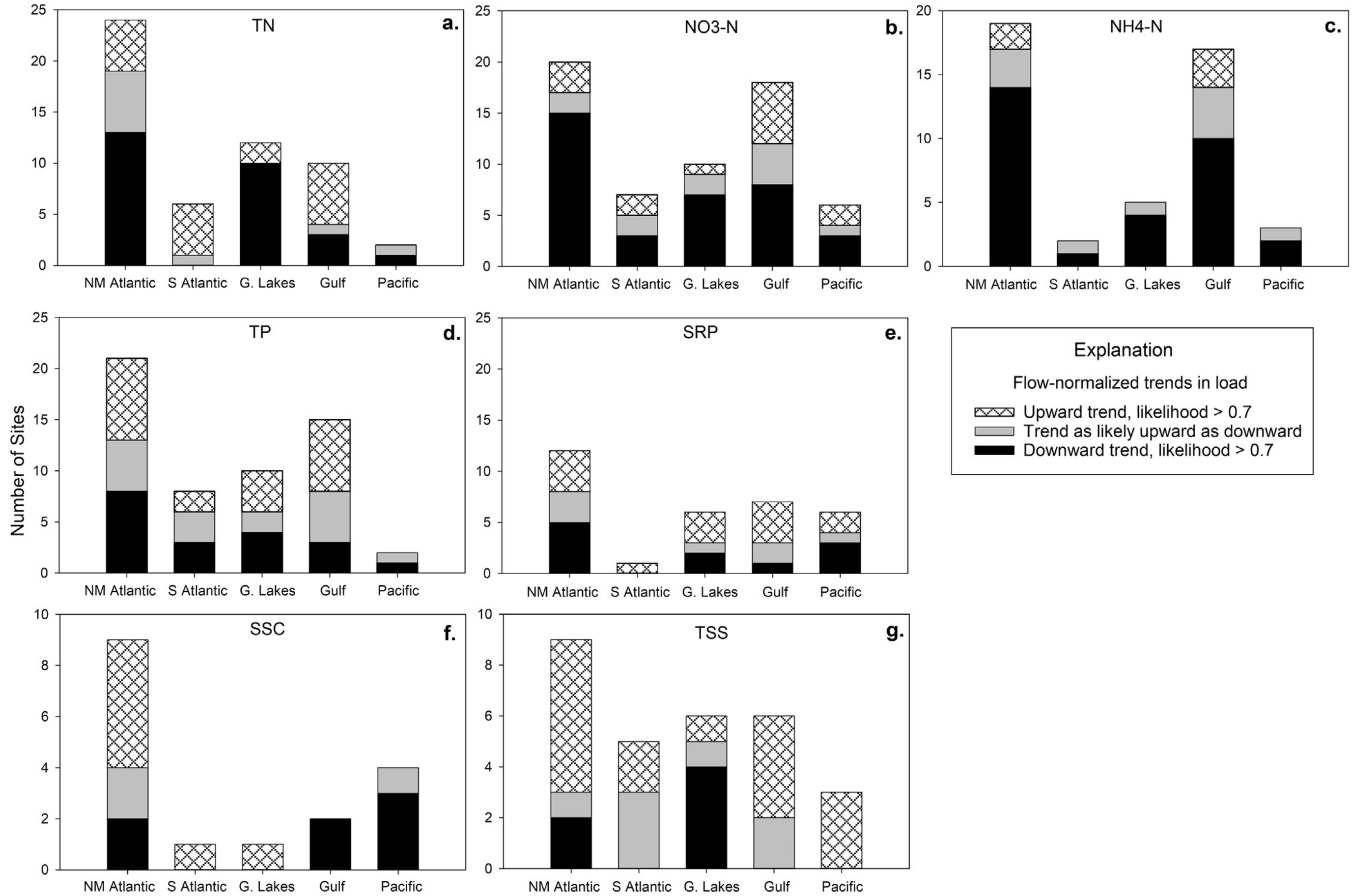

Fig. 2. Number of sites in each region with increasing and decreasing flow-normalized (FN) trends in loads (likelihood >0.7) and FN trends in loads that are as likely to be upward as downward (likelihood <0.7) for total nitrogen (TN) (a), nitrate (NO3-N) (b), ammonium (c), total phosphorus (TP) (d), orthophosphate (SRP) (e), suspended sediment concentration (SSC) (f), and total suspended solids (TSS) (g) between 2002 and 2012. Regional abbreviations are as follows: NM Atlantic = North/Mid-Atlantic, S Atlantic = South Atlantic, G. Lakes = Great Lakes, and Gulf = Gulf of Mexico.

and plot mostly along a 1:1 line (Fig. S7), indicating that the changes in TN were primarily driven by changes in NO3-N. The comparison of sites with both TN and TP trends ($n = 45$) indicates a distinct clustering of sites by land use (Fig. 4). TN and TP yields decreased at urban sites and increased at undeveloped sites. Mixed land-use sites had predominantly decreasing TN yields, but both increasing and decreasing TP yields. Agricultural sites had widely varying combinations of TN and TP yield trends, indicating a lack of overall progress in decreasing nutrient yields to coastal waters from agricultural watersheds.

3.5. Nutrient loading to estuaries

The 95 trend sites drained into 56 unique coastal estuaries including 11 in the Great Lakes region, 6 FDAs (freshwater portions of a watershed upstream of an EDA), 5 CDAs, and 34 EDAs (Table S1). Of these 56 unique coastal estuaries, 40 including the 34 EDAs and the 6 FDAs were assessed in the NEEA report (Bricker et al., 2007). Most of the sites ($n = 66$) were assigned an “influencing factors” rating and there were 10 sites with trend results that had insufficient data to be assigned an “influencing factors” rating in the NEEA report.

Between 2002 and 2012, NO3-N yield decreased in a majority of the streams and rivers that flow into the most sensitive estuaries, those that had “high” influencing factors for eutrophication, especially in the North/Mid-Atlantic region and the Gulf region (Fig. 5a). However, in the South Atlantic, only half of the trend sites along rivers draining to sensitive estuaries had decreasing NO3-N yields. Fewer TN yields than NO3-N yields decreased in the streams and rivers that flow into the most sensitive estuaries in the Gulf and North/Mid-Atlantic regions and TN yields increased at all trend sites in the South Atlantic that flow into sensitive estuaries (Fig. 5b). In contrast, among estuaries with a high susceptibility to eutrophication, about an equal number of sites had increasing as decreasing TP yields (Fig. 5c). At the sites with insufficient data to be assigned an “influencing factors” rating, trend results for NO3-N, TN, and TP indicate both increasing and decreasing yields. Additionally, for TN and TP, the percent change was larger for the increasing yields than for the decreasing yields (Fig. 5b and c).

3.6. Comparison of long-term N and P yields in large U.S. and global coastal rivers

To provide a longer-term perspective on the recent trends, TN yield estimates from 1972, 1982, 1992, 2002, and 2012 in the largest coastal rivers with trend results were examined (Fig. 6a and b, Table S7). TN yields generally decreased between 1972 and 2012, and the 2012 TN yields were usually the lowest yields for the available trend period (Fig. 6a). Decreases in TP yield estimates were less common (Fig. 6b). For both TN and TP, the changes in yield were often inconsistent over time, varying in both trend magnitude and direction although changes in trend direction were more common for TP (Table S7). In the coastal rivers of the conterminous U.S., TN yields in 2012 were all <22 kg N ha^{-1} yr^{-1} (with most <10 kg N ha^{-1} yr^{-1}) and TP yields were all <2 kg P ha^{-1} yr^{-1} (with most <1 kg P ha^{-1} yr^{-1}). Many of the highest TN and TP yields occurred in the medium-sized watersheds (<10,000 km^2 and <100 cms), while TN and TP yields at the largest coastal rivers were small by comparison.

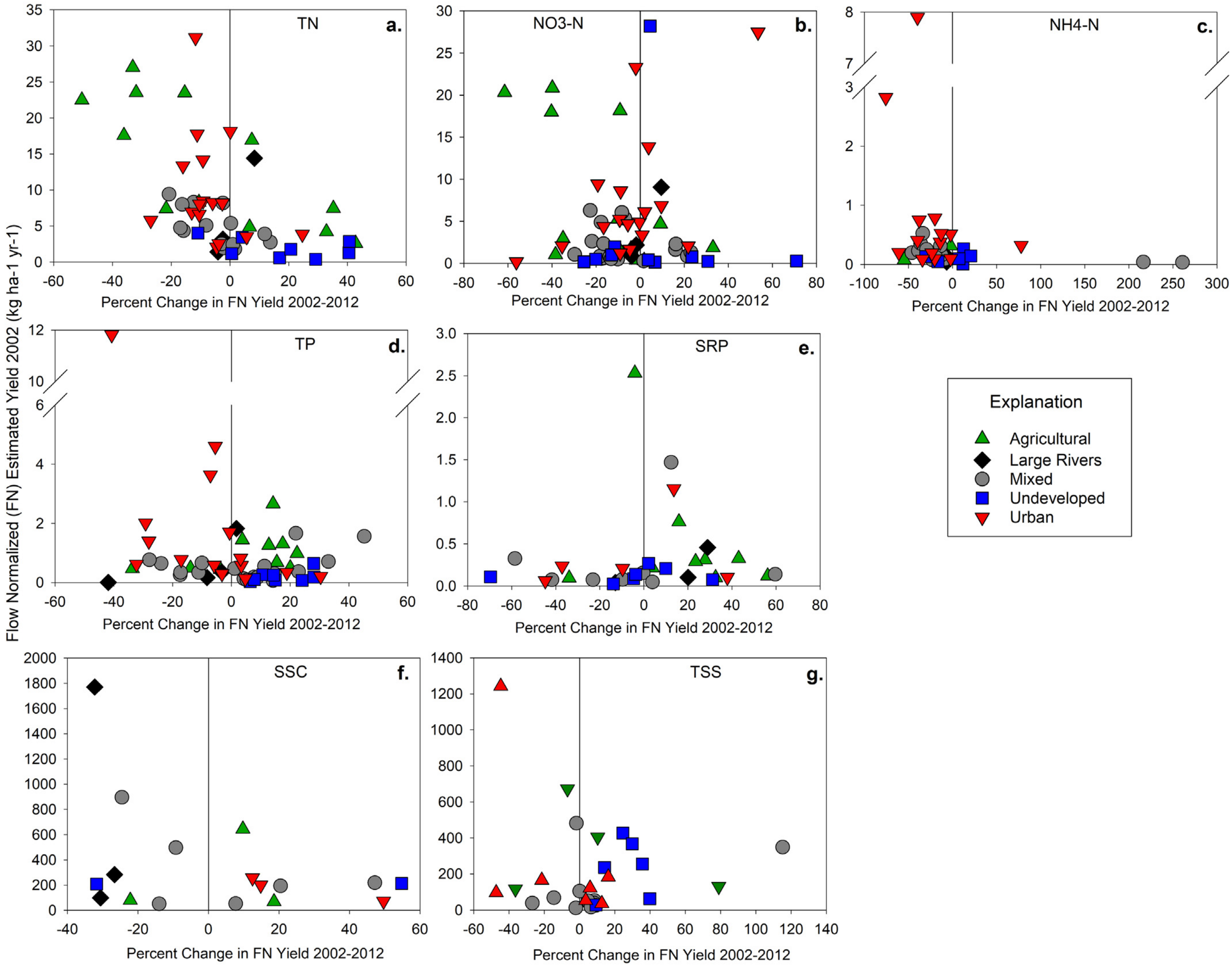

Fig. 3. Comparison of flow-normalized (FN) yield in 2002 to the percent change (trend) in FN yield between 2002 and 2012 at each site by land use for total nitrogen (TN) (a), nitrate (NO3-N) (b), ammonium (c), total phosphorus (TP) (d), orthophosphate (SRP) (e), suspended sediment concentration (SSC) (f), and total suspended solids (TSS) (g) between 2002 and 2012.

TN yields for medium to large coastal rivers in the conterminous U.S. were somewhat lower than other global coastal rivers, while TP yields were slightly higher in coastal rivers in the conterminous U.S. (Tables 1; S7). The median TN yield in the conterminous U.S. rivers was 4.4 kg N ha^{-1} yr^{-1}, approximately 32% lower than the median TN yield for the other global rivers (6.5 kg N ha^{-1} yr^{-1}). Approximately 34% of the global rivers in our comparison had TN yields >10 kg N ha^{-1-} yr^{-1}. Median TP yield in the conterminous U.S. rivers was approximately 26% higher than was the median TP yield for the other global rivers (0.34 kg P ha^{-1} yr^{-1} and 0.26 kg P ha^{-1} yr^{-1}, respectively) but the difference between the median yields was small. Approximately 16% of the coastal rivers in both the conterminous U.S. (6 rivers) and globally (9 rivers) had TP yields >1 kg P ha^{-1} yr^{-1}. The subset of coastal rivers in Europe had somewhat higher median TN and TP yields (6.8 kg N ha^{-1} yr^{-1} and 0.30 kg P ha^{-1} yr^{-1}, respectively) than when combined with other global rivers, but the comparisons to TN and TP yields from coastal rivers of the conterminous U.S. is similar.

3.7. Comparison of historic and recent N:P ratios

Comparison of historic (<1979) and recent (2002–2012) N:P values can provide a context for the recent trends and an estimate of the potential for nutrient limitation in the rivers and streams discharging to coastal waters. There were 51 coastal sites that had sufficient TN and TP data to compare historic and recent N:P ratios. The majority of median N:P ratios at all sites for both time periods were much higher than the Redfield ratio of 16, indicating a potential for P limitation (Fig. 7) (Redfield, 1958). Additionally, a majority of median N:P ratios for more recent years (2002–2012) were elevated relative to historic (<1979) values. Median N:P values were <16 at 33% of sites prior to 1979 (17 sites) whereas only 18% of sites (9 sites) had median N:P values <16 after 2002.

We compared the change in the median historic and recent concentrations of TN and TP to understand possible causes of the change in the N:P ratios (Table S6). Among the sites with elevated recent N:P ratios, recent median TN concentrations were greater than historic median TN concentrations at 18 sites. Of these 18 sites, recent median TP concentrations were less than historic median TP concentrations at 12 sites. Conversely, there were 21 sites with elevated recent N:P ratios where recent median TN concentrations were less than historic median TN concentrations. At most of these sites, recent median TP concentrations were also less than historic median TP concentrations. Elevated recent N:P ratios are primarily driven by two scenarios of changing TN and TP concentrations. Recent median N:P ratios were understandably elevated when median TN concentrations increased and median TP concentrations decreased relative to historic values. Additionally, at the sites where median TN and TP concentrations have both decreased

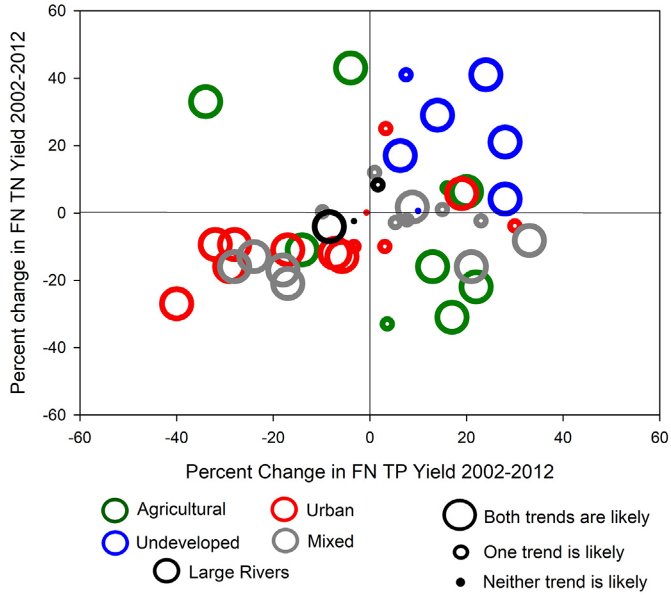

Fig. 4. Relationship between trends in flow-normalized (FN) total nitrogen (TN) yield and FN total phosphorus (TP) yield for the recent (2002–2012) trend period. Symbol color indicates watershed land use. Symbol size represents the trend likelihood for both trends on the figure. Likelihood was determined by bootstrap analysis and trend results with a likelihood >0.7 were considered "likely" while those with likelihood <0.7 were considered "unlikely." Large circles indicate both trends are likely, small circles indicate that only one of the trends is likely, and dots indicate that neither trend is likely.

relative to historic values, TP decreased more than TN, resulting in greater N concentrations relative to P.

4. Discussion

4.1. Regional and land-use differences in N and P loading trends

Decreases in N loading to U.S. coastal waters in the recent trend period (2002–2012) and in the longer-term trend periods (1972–2012) were more common than decreases in P loading, reflecting some success in controlling N loading. The differences in recent N and P trends were strongly related to differences in land use which often reflects both nutrient sources and pollution control efforts. Some of the most successful efforts to reduce nutrient loading have come from improvements to wastewater treatment, combined sewer overflows, and stormwater management in places like Chesapeake Bay (Fisher et al., 2006; Liner et al., 2017; Rice et al., 2017; Sparkman et al., 2017), the Hudson River and Raritan Bay (Hickman and Hirsch, 2017), and in places with tertiary treatment of sewage (Carey and Migliaccio, 2009). The urban watersheds in this study had higher densities of major wastewater facilities than watersheds with other dominant land uses (Fig. S2). In agricultural areas, fertilizer and manure, often from concentrated animal feeding operations (CAFOs), are the dominant nutrient sources, and agricultural runoff is not specifically regulated by the Federal Water Pollution Control Act of 1972 (known as the Clean Water Act). In many watersheds, agriculture remains the largest source of nutrients (Boyer et al., 2006; Howarth et al., 2002; Preston et al., 2011; Robertson and Saad, 2013) and the agricultural watersheds in our study had higher estimates of N and P loading from manure and

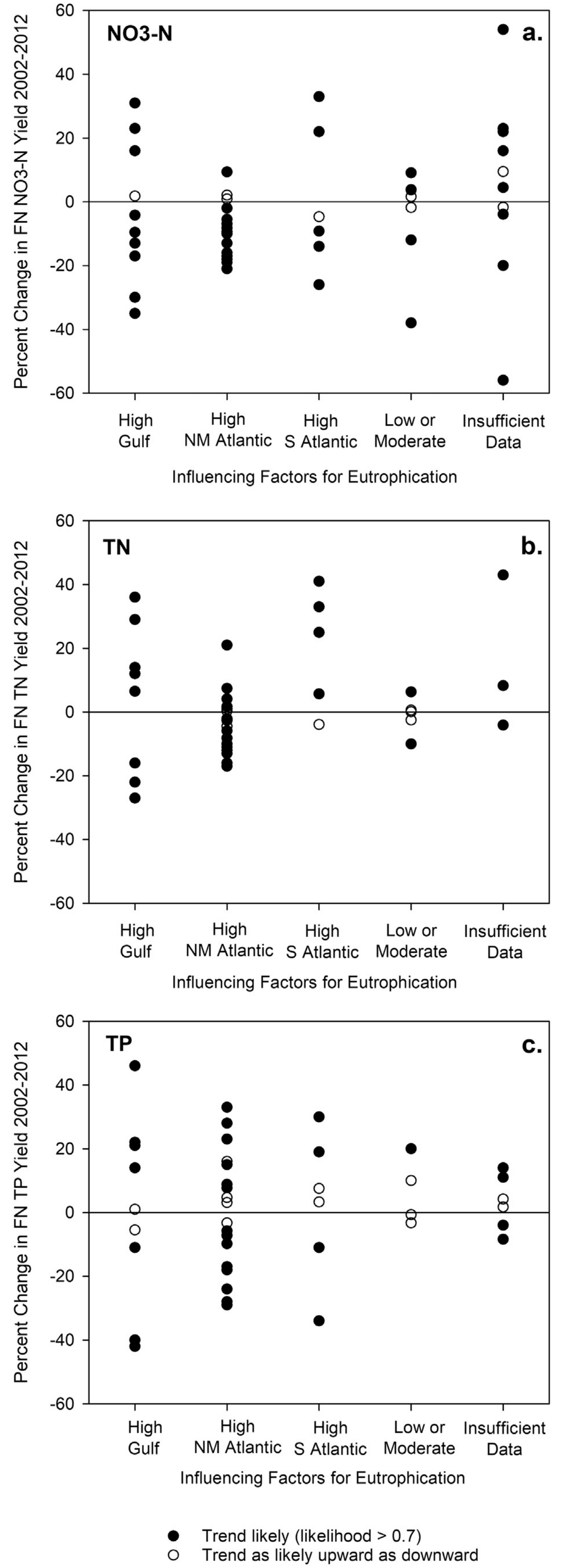

Fig. 5. Trends in flow-normalized (FN) total nitrogen (TN) (a), nitrate (NO3-N) (b), and total phosphorus (TP) (c) grouped by estuary susceptibility to eutrophication (as measured by the influencing factors rating from Bricker et al., 2007). Each point represents the trend result for a unique coastal river or stream. For estuaries with a "High" influencing factors rating, rivers and streams were further divided by region where: Gulf = Gulf of Mexico, NM Atlantic = North/Mid-Atlantic, and S Atlantic = South Atlantic.

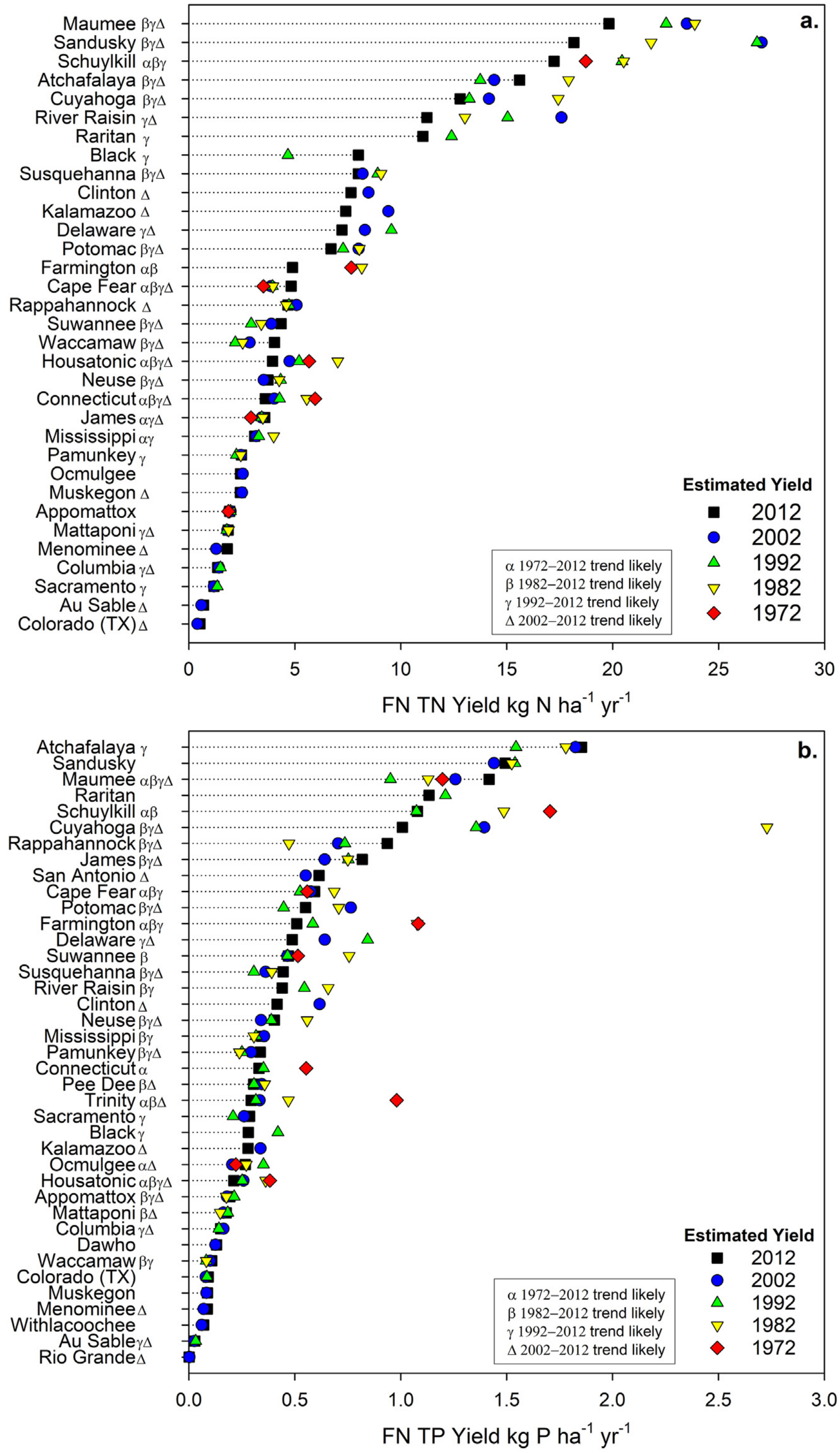

Fig. 6. Comparison of flow-normalized (FN) annual yield estimates for the medium to large coastal rivers (average annual discharge >30 cms or watershed area >1500 km^2) over the trend periods for (a) total nitrogen (TN) and (b) total phosphorus (TP). The different colored and shaped symbols represent the available yield estimates at the start of the four trend periods: 1972, 1982, 1992, and 2002. The black squares represent the yield estimates in 2012, at the end of the trend periods. Greek letters following the name of the river on the y-axis indicate whether the trend for each available period was likely (likelihood >0.7). Note that not all sites had sufficient data for trend analysis for all four trend periods.

fertilizer than watersheds in other land-use categories (Fig. S3). Atmospheric deposition from fossil fuel combustion can be a source of N to aquatic ecosystems irrespective of the type or intensity of development in the watershed (Boyer et al., 2006; Howarth et al., 2002; Preston et al., 2011). Reductions in N loading to watersheds occurred in many regions following passage of the Clean Air Act Amendments (Eshleman et al., 2013; U.S. Environmental Protection Agency, 2017).

The fewer number of coastal sites with decreases in P loads as compared with N loads between 2002 and 2012 as well as the variability in P trend directions over time (1972–2012) could be due to differences in source and in-stream behavior. P loading from wastewater was reduced following two regulatory efforts (1) phosphate detergent was banned beginning in 1971 and (2) the Clean Water Act specifically targeted point sources and resulted in widespread upgrades to wastewater

Table 1
Compilation of published estimates of TN and TP yield from other global rivers.

River	Receiving Water	Area (km^2)	TN yield (kg N ha^{-1} yr^{-1})	TP yield (kg P ha^{-1} yr^{-1})	Source
Neva	Baltic Sea	271,800	1.9	0.07	a
Vistula	Baltic Sea	194,420	7.5	0.47	a
Odra	Baltic Sea	118,840	8.7	0.38	a
Nemunas	Baltic Sea	97,920	4.5	0.18	a
Daugava	Baltic Sea	86,530	4.5	0.18	a
Kemijoki	Baltic Sea	51,130	1.3	0.04	a
Göta älv	Baltic Sea	50,230	2.9	0.08	a
Loire	Atlantic (France)	116,981	12	0.82	b
Garonne	Atlantic (France)	55,703	6.3	0.31	b
Dordogne	Atlantic (France)	23,902	7.3	0.14	b
Adour	Atlantic (France)	16,861	11	0.65	b
Vilaine	Atlantic (France)	10,490	15	0.33	b
Blavet	Atlantic (France)	2057	35	0.53	b
Aulne	Atlantic (France)	1687	27	0.43	b
Douro	Atlantic (Portugal)	97,682	3.4	0.19	b
Tagus	Atlantic (Portugal)	81,947	3.3	0.29	b
Seine	English Channel	75,989	17	0.66	b
Scheldt	English Channel	21,860	13	0.82	b
Somme	English Channel	6223	10	0.25	b
Orne	English Channel	2948	26	0.41	b
Po	N Adriatic	71,057	16	1.1	b
Rhone	W Mediterranean	96,619	12	0.54	b
Ebro	W Mediterranean	85,000	3.4	0.10	b
Jucar	W Mediterranean	21,578	0.27	0.01	b
Segura	W Mediterranean	19,525	0.01	0	b
Tiber	W Mediterranean	17,375	16	0.84	b
Arno	W Mediterranean	8228	15	0.60	b
Aude	W Mediterranean	5226	4.9	0.23	b
Argens	W Mediterranean	2762	2.0	0.13	b
Herault	W Mediterranean	2625	3.1	0.18	b
Touloubre	W Mediterranean	1576	2.0	0.15	b
Orb	W Mediterranean	1556	8.7	0.39	b
Vidourle	W Mediterranean	827	4.2	0.15	b
Gapeau	W Mediterranean	566	5.1	0.42	b
Changjiang	East China Sea	1,809,000	5.5	0.07	c
Qiantangjiang	East China Sea	41,000	12.1	0.26	c
Zhujiang	South China Sea	590,000	9.4	0.19	c
Huanghe	Yellow Sea	752,000	2.0	0.01	c
Huaihe	Yellow Sea	270,000	1.3	0.12	c
Han	Yellow Sea	26,200	25.9	0.86	c
Daguhe	Yellow Sea	5600	2.2	0.01	c
Ganges	Bay of Bengal	1,626,470	26	5.8	d
Irrawaddy	Bay of Bengal	405,481	24	5	d
Godavari	Bay of Bengal	317,127	6.7	1.1	d
Salween	Bay of Bengal	273,038	9.9	3	d
Krishna	Bay of Bengal	266,291	2.9	0.19	d
Mahanadi	Bay of Bengal	141,040	8.5	0.86	d
Cauweri	Bay of Bengal	78,587	2.9	0.21	d
Damodar	Bay of Bengal	59,591	17	4.7	d
Brahmani	Bay of Bengal	57,289	10	1.3	d
Penner	Bay of Bengal	53,845	0.88	0.02	d
Amazon	Atlantic Ocean	5,846,870	8.5	1.38	e, f
Orinoco	Atlantic Ocean	1,038,130	10.8	2.21	e, f
Lena	Arctic Ocean	2,438,900	1.2	0.06	e, f
Kolyma	Arctic Ocean	664,851	1.88	0.04	e, f
Yana	Arctic Ocean	224,724	0.73	0.02	e, f
Yukon	Pacific Ocean	854,690	3.35	0.09	e, f

Note. Sources for TN and TP yield estimates are as follows: [a]HELCOM, 2018; [b]Romero et al., 2013; [c]Liu et al., 2009, note: TN yield is for DIN, TP yield is PO_4^{3-}-P; [d]BOBLME, 2014; [e]Mayorga et al., 2010; and [f]Beusen et al., 2005.

treatment plants (Litke, 1999). However, P from non-point sources continues to be widespread (Carpenter et al., 1998; Litke, 1999; Preston et al., 2011; Stoddard et al., 2016). Recently, the predominant sources of elevated P in streams are fertilizer and manure from agricultural areas and, to a lesser extent, wastewater effluent and urban runoff in urbanized areas (Carpenter et al., 1998; Daniel et al., 1998; Howarth et al., 2002; Preston et al., 2011). The P trend results presented in this study reflect the different P sources and control efforts; P loads decreased in many urban watersheds and increased in many agricultural watersheds. Variability in longer-term P trends likely reflect the variable timing of phosphate detergent bans and improvements to wastewater treatment facilities (both secondary and tertiary treatment) in the watersheds, and the increasing influence of diffuse agricultural sources of P as point source loadings are reduced.

In the Lake Erie basin, agricultural watersheds with high TN yield experienced recent decreases in N loading, but not concurrent decreases in P loading. Changing agricultural practices including manure application and conservation tillage have been suggested as possible reasons for decreasing TN yields (Stow et al., 2015). Reductions in atmospheric deposition of N has been proposed as a reason for the decline in TN yields in the Great Lakes area (Stow et al., 2015) and would not be accompanied by decreases in TP. Unfortunately, decreases in N loading may have limited influence on water quality in Lake Erie, where P is the primary driver of eutrophication (Muenich et al., 2016; Watson

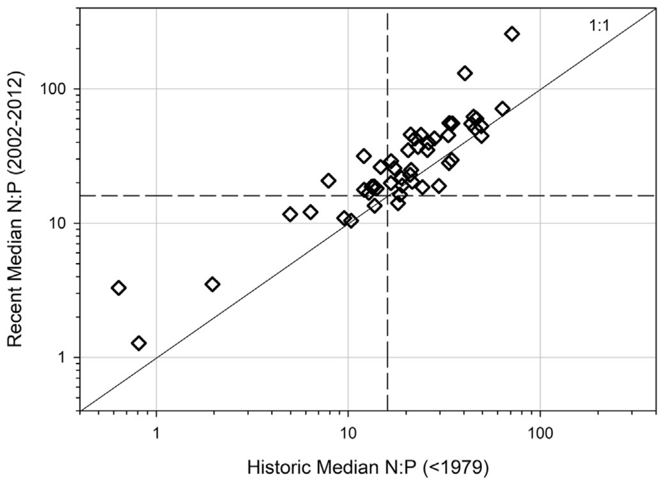

Fig. 7. Comparisons of historic (pre-1979) and recent (2002–2012) N:P ratios at coastal trend sites. Dashed lines denote a 16:1 N:P ratio. Median total nitrogen (TN) and total phosphorus (TP) concentrations at the coastal trend sites for both the historic and recent time periods are listed in Table S6.

et al., 2016). Increases in TP yields within the Lake Erie basin have been attributed to legacy P, while changing agricultural practices, including the implementation of conservation tillage, have been suggested as causes for increases in reactive P (Baker et al., 2014; Daloğlu et al., 2012; Muenich et al., 2016; Stow et al., 2015).

Trends in nutrient loads (NO3-N, TN, and TP) in the Mississippi River at St. Francisville, LA during 2002–2012 were near-level (−2% with an uncertain likelihood) and indicate that upstream improvements in water quality are not sufficient nor widespread enough to significantly reduce nutrient loading downstream. Our findings are similar to other recent studies showing small or uncertain trends in Mississippi River nutrients in recent decades (Justić et al., 2002; Murphy et al., 2013; Oelsner et al., 2017; Sprague et al., 2011). Nutrient loading has decreased in parts of the Mississippi River basin; most notably N loads decreased in the Iowa and Illinois Rivers and to a lesser extent in the Ohio River from 2000 to 2010, and TN and NO3-N loads decreased 2002–2012 in the Iowa, Illinois, and Ohio Rivers (Murphy et al., 2013; Oelsner et al., 2017). However, this has not translated into decreases downstream at St. Francisville, LA. This result has been attributed to legacy N from groundwater or other sources offsetting trends in different parts of the basin (David et al., 2010; Murphy et al., 2013; Van Meter et al., 2017). The large watershed size and distributed nature of agricultural nutrient sources are a challenge for reducing nutrient loads to the Gulf of Mexico from both the Mississippi and Atchafalaya Rivers and in other large river watersheds.

TN and TP loading from undeveloped watersheds to U.S. coastal waters was low but generally increased, which could indicate degradation of relatively pristine coastal watersheds of the Nation. Nutrient loading from the undeveloped sites was almost universally low as compared with other U.S. coastal rivers in more developed watersheds and indicates that these watersheds represent areas that are minimally affected by human activities. TN and TP yields from the undeveloped sites are within the range of the lower third of the TN and TP yields from other global rivers (Tables 1; S7). Importantly, some undeveloped sites flow into estuaries that have a moderately high susceptibility to eutrophication like the Suwannee River estuary, Matagorda Bay, Rio Grande estuary, and Winyah Bay (Table S5, Bricker et al., 2007). Previous work has also reported increasing TP in undeveloped catchments (Stoddard et al., 2016). While nutrient loading from these undeveloped sites may not be immediate threats to eutrophication, they emphasize the need for continued and increased monitoring of undeveloped areas to preserve watersheds that are minimally affected by human activities.

A further concern is that nutrient increases were common in estuaries with relatively little information on eutrophication status, suggesting that a lack of monitoring information could be hampering our ability to determine high-risk estuaries and track related changes in nutrient loading to those estuaries. For example, Choctawhatchee Bay, San Antonio Bay, Stono/North Edisto Rivers, Tillamook Bay, and Umpqua River all had insufficient data at the time of the NOAA NEEA Update to be assigned an "Influencing Factors" rating for eutrophication risk (Bricker et al., 2007), but rivers and streams draining into these estuaries had increasing nutrient loads. In fact, despite exhaustive efforts of this study to include as many sites as possible, trend sites represented ≥50% of the freshwater inputs to only 18 estuaries (including 5 within Chesapeake Bay) suggesting that changes in riverine nutrient loading to estuaries is not well-understood.

While substantial progress has been made toward improving water quality in Chesapeake Bay (Zhang et al., 2018), additional pollution control measures are required to reduce TP loads and continue reducing TN loads to the Bay. Long-term decreases in TN and TP are occurring at monitoring stations throughout the watershed (Moyer and Blomquist, 2017). However, increasing TP loads were more prevalent in the recent period (2002–2012) indicating that P may be a re-emerging problem (Moyer and Blomquist, 2017). Various explanations have been proposed for this phenomenon (Fanelli et al., 2019; Hirsch, 2012; Hogan, 2008; Zhang et al., 2016), but it may be a leading indicator of further water-quality degradation in other areas of the Nation as initial reductions in TP from the Clean Water Act are overwhelmed by emerging sources.

Sediment loading decreased into the Columbia River, San Francisco Bay, and the Gulf of Mexico even where there were minimal changes in nutrient loading. The construction of upstream dams, erosion control structures and practices that trap sediment, and channelization can reduce sediment loads (Keown et al., 1986; Meade and Moody, 2009; Syvitski, 2005; Turner and Rabalais, 2003). Changes in sediment loading can be either beneficial or detrimental to coastal ecosystems depending on many interrelated factors. Sediment can transport pollutants to an estuary and increase turbidity, which is detrimental to seagrass and other aquatic vegetation (Horowitz et al., 2012; Orth et al., 2006; Walling and Collins, 2008). While reduced sediment loads have decreased mercury and PCB loading to San Francisco Bay (Bricker et al., 2007; Cloern, 2001), sediment-starved wetlands in the Mississippi River delta are being lost to coastal erosion, sea-level rise, and subsidence (Coastal Protection and Restoration Authority, 2012; Horowitz, 2009). Reduced sediment loads can also increase eutrophication risk as light limitation is reduced (Bricker et al., 2007). These contrasting examples underscore the complexity of interpreting sediment loads at a continental scale.

4.2. Global context of N and P yields and trends

The finding of somewhat lower TN yields and slightly higher TP yields from medium to large coastal rivers of conterminous U.S. from this study as compared to global rivers is not inconsistent with regional yield estimates from the Global NEWS 2 model for 2000 (Mayorga et al., 2010). DIN yield is particularly high in many areas of Europe and South Asia and somewhat lower in the eastern U.S. (Mayorga et al., 2010). The differences in TP yield between coastal rivers of conterminous U.S. and global rivers is small, and the slightly lower median TP yield from the global rivers could be related to only having DIN and SRP yield estimates for rivers in China and the Korean Peninsula, where particulate P yields are large (Mayorga et al., 2010). TP export from North America is estimated to be higher than TP export from Europe, but lower than TP export from South Asia and South America (Mayorga et al., 2010). Globally, anthropogenic nonpoint (diffuse) sources of N from agricultural activities dominate DIN yields (Boyer et al., 2006; Seitzinger et al., 2005) and anthropogenic point sources from sewage dominate DIP yields (Seitzinger et al., 2005). Similarly, high N and P yields from the conterminous U.S. are likely due to predominantly anthropogenic sources of agriculture and wastewater (Howarth et al., 2002). Estimates of TN and TP yield for rivers outside the conterminous U.S. based on empirical data are relatively uncommon (particularly outside Europe) and reflect the need for more widespread water-quality monitoring to improve nutrient loading and trend estimates. Models such as Global NEWS are useful for making global comparisons of nutrient loading and would also be improved with increased water-quality monitoring data for model calibration.

Global trends in N and P loading often vary by river but generally reflect some of the same land-use influences observed in this study. TN and TP inputs to the Baltic Sea and North Sea have decreased in recent decades, although the change in loads has not been consistent and there have been some periods of increased loading (Grizzetti et al., 2012; HELCOM, 2018). In southwestern Europe, P loads decreased significantly in most rivers while N loads have remained relatively constant or increased (Romero et al., 2013). Many countries in Europe have established reduction goals to limit nutrient loading to coastal waters through four different international conventions: HELCOM, OSPAR, Barcelona, and Bucharest (Grizzetti et al., 2012). Additionally, there are several European regulation policies that have the goal to reduce nutrient loading to coastal waters: the Nitrates Directive (91/676/EEC), targeting diffuse agricultural sources of N, the Urban Waste Water Treatment Directive (91/271/EEC), targeting nutrients from urban wastewater; the Water Framework Directive (2000/60/EC), aiming for good status for all surface and ground waters by River Basin District, and the recent Marine Strategy Directive, (Directive 2008/56/EC), to achieve Good Environmental Status by 2020 (Grizzetti et al., 2012; Romero et al., 2013). To date, efforts to reduce P have been more successful than policies targeted to reduce N (Grizzetti et al., 2012; Ludwig et al., 2010; Romero et al., 2013), which may be related to the difference between primary N and P sources. P is derived primarily from point sources and phosphate detergent bans coupled with wastewater treatment improvements have been widely successful, whereas N is derived primarily from diffuse sources, including agriculture, and has proven more difficult to control (Grizzetti et al., 2012; Ludwig et al., 2010; Romero et al., 2013; Seitzinger et al., 2010). In the Changjiang River, 6- to 10-fold increases in DIN concentrations over the last 30 years are attributed to the intensification of agriculture and increased use of fertilizers (Liu et al., 2009). Interestingly, the storage of P in soils may have resulted in relatively stable SRP concentrations in the Changjiang River over the same time period (Liu et al., 2009), suggesting that it may become an important source in the future.

4.3. Implications of historic and current N:P ratio comparison

While our observations that recent (2002–2012) N:P ratios are generally elevated as compared with historic (prior to 1979) values may be produced by multiple combinations of trends in N or P, the results nevertheless have two important likely implications for current nutrient inputs to coastal waters. We interpret these results with the assumption that the growth of primary producers in coastal waters can be nutrient limited, although it is acknowledged that disturbances, light limitation, and trophic controls may also limit the growth of primary producers, in which case N and P loading may both exist in excess of maximum ecological requirements.

First, greater N concentrations relative to P suggest that P limitation of estuaries may be of greater importance now than historically, despite reductions of N at many sites. N is generally regarded as the primary driver of coastal eutrophication (Elser et al., 2007; Howarth and Marino, 2006), although scientific opinion about this has changed over time. The recent NCCA report concluded that P was the most widespread stressor of water quality in coastal marine waters (U.S. Environmental Protection Agency, 2016). However, the change analysis in the NCCA report was limited to recent data (>2000) and did not address whether N loads have decreased enough to return to historic values. Therefore, while P may be limiting in the most immediate sense, that condition should be understood to be a product of the elevated N concentrations relative to P concentrations in many of these tributaries to estuaries.

Second, while P limitation may currently be more prevalent, focus should not be lost on the need to reduce N concentrations in estuaries. Our results are consistent with a recent study that determined N:P ratios increased globally over the 20th century, perhaps due to the stagnation of P fertilizer but an increase in N fertilizer (Beusen et al., 2016). In a large study that compiled the results of many N and P enrichment field experiments, additions of N and P both resulted in enhanced growth in freshwater and marine systems and, importantly, the additions of N and P had synergistic effects producing higher responses than the single nutrient additions (Elser et al., 2007). While each situation is different, our finding that N:P ratios remain elevated compared to historic values, despite decreases in N and P concentrations, suggests that elevated N concentrations relative to P is a common condition and attention should be paid to reducing both N and P as a way of controlling eutrophication of estuaries.

4.4. Geographic gaps in coastal trends

Despite an unprecedented data compilation effort for a national trend assessment that resulted in trend results for 95 sites at 88 rivers and streams that drain to estuaries and coastal waters including the Great Lakes, substantial geographic gaps remained. The combined median flows of these 95 sites captured 62% of the freshwater flows to coastal areas. From a population perspective, the trend sites only included about 21 million people or approximately 13% of the NOAA population estimate of people living in coastal watersheds in 2012. Some areas with long-standing monitoring programs were well-represented, such as the Chesapeake Bay, Delaware Bay, Mississippi and Atchafalaya Rivers, Columbia River, and San Francisco Bay. However, there were noticeable geographic gaps in the New England, South Atlantic, and Pacific regions and many of the major metropolitan areas located near the coast. Additionally, five major rivers (St. Lawrence, Hudson, Altamaha, Mobile, and Brazos) did not have sufficient data to be included in this study. The lack of systematic monitoring and reporting of coastal water quality and inadequate coverage of U.S. coasts has been previously reported (National Research Council, 2000) and results in monitoring records for many coastal rivers that are either missing or inadequate for a detailed, national-scale trend analysis such as the one conducted in this study. Our conclusions are somewhat limited by these geographic gaps and highlights the need for systematic monitoring and reporting and better coverage of U.S. coasts as suggested by the National Research Council (2000) and NOAA (Bricker et al., 2007).

5. Conclusions

To address questions about how water quality is changing in the U.S., the USGS NAWQA Project compiled data from multiple public sources for trend assessment. Coastal sites were selected from this larger pool of sites to examine trends in nutrient and sediment loading to coastal waters, including the Great Lakes. A total of 95 sites and 295 recent (2002–2012) nutrient and sediment trends were included in this study. This analysis allowed us specifically to address recent (2002–2012) trends in nutrient and sediment loadings to U.S. coastal waters. We also explored regional differences in nutrient and sediment loadings as well as compared trends in nutrient and sediment loading among land-use categories. To provide a broader context for these recent trends, we compared estimates of TN and TP yield between 1972 and 2012 for the larger rivers in this study to provide a longer-term perspective and compared the TN and TP yield estimates to values for rivers outside of the conterminous U.S. to provide a global perspective. By examining the long-term data compiled for this study, we addressed whether progress has been made in reducing nutrients compared to historic values.

Overall, N loads decreased at approximately 60% of the coastal sites whereas P loads decreased at approximately 33% of sites and sediment loads decreased at 25–50% of sites. However, there were differences in nutrient and sediment trends among land-use categories. In general, N and P loading from urban watersheds to U.S. coastal waters decreased between 2002 and 2012. In contrast, there were both increases and decreases of N and P loading to U.S. coastal waters from agricultural watersheds, although increases in P loading were common indicating uneven progress in reducing nutrient loading from agricultural sources. Decreases in N loading from agricultural watersheds were primarily in the Lake Erie basin. These decreases may have little impact on water quality in Lake Erie, where P is the primary driver of eutrophication. Consideration of the results differentiated by land use and region can provide insight into the processes affecting coastal nutrient loading and therefore allow generalization of the results beyond the direct monitoring network. Decreases in nutrient loading are often the result of targeted legislation. In both the U.S. and Europe, efforts to reduce point-sources of nutrients (often the dominant source in urban areas) have been more successful than efforts to reduce diffuse sources of nutrients which are more commonly the primary source in agricultural areas.

Nutrient and sediment loading between 2002 and 2012 to Chesapeake Bay and the Gulf of Mexico, two of the coastal areas with the highest risk of regional eutrophication, were largely consistent with previous studies. N loading to the Chesapeake Bay decreased in a majority of tributaries between 2002 and 2012, but P loading to the Bay generally increased. N and P loading to the Gulf of Mexico from the Mississippi River was relatively stable between 2002 and 2012. These trend results highlight the need for continuing and new pollution control measures to reach nutrient reduction targets.

TN and TP loading from undeveloped watersheds to U.S. coastal waters was relatively low but increased between 2002 and 2012, which could indicate degradation of coastal watersheds that are minimally affected by human activities. Additionally, nutrient loading increased to many estuaries with insufficient data to be assigned an "influencing factors" for eutrophication risk score, which indicates the need for the continuation of monitoring and assessment in some of the less-developed estuaries.

TN yields for the coastal rivers in the conterminous U.S. were somewhat lower than other global coastal rivers, while TP yields were slightly higher in coastal rivers in the conterminous U.S. However, the underlying patterns of nutrient sources and land use were similar for many of the rivers. Both globally and within the conterminous U.S., reductions in nutrients, especially P, from point sources in urban areas are common, especially in Europe. On the other hand, many watersheds with primarily diffuse agricultural sources of nutrients have steady or increasing N and P loads.

The overall lack of decreasing P loads at many sites, especially outside of urban areas, could suggest that P is the primary threat to coastal water eutrophication and should be the focus of nutrient reduction plans. However, a comparison of historic (pre-1979) to recent (2002–2012) N:P values at the trend sites indicate that N remained elevated relative to P even in rivers where there were concurrent decreases in N and P concentrations. This indicates that more progress is needed in reducing both N and P loading to U.S. coastal waters, particularly in agricultural areas other than the Lake Erie basin. However, substantial geographic gaps remain that provide a challenge for interpretation and suggest that improved monitoring will be needed to fully describe progress in controlling coastal eutrophication.

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Appendix A. Supplementary information

Supplementary information to this article can be found online at https://doi.org/10.1016/j.scitotenv.2018.10.437.

References

Baker, D.B., Confesor, R., Ewing, D.E., Johnson, L.T., Kramer, J.W., Merryfield, B.J., 2014. Phosphorus loading to Lake Erie from the Maumee, Sandusky and Cuyahoga rivers: the importance of bioavailability. J. Great Lakes Res. 40, 502–517. https://doi.org/10.1016/j.jglr.2014.05.001.

Beusen, A.H.W., Dekkers, A.L.M., Bouwman, A.F., Ludwig, W., Harrison, J., 2005. Estimation of global river transport of sediments and associated particulate C, N, and P. Glob. Biogeochem. Cycles 19. https://doi.org/10.1029/2005GB002453.

Beusen, A.H.W., Bouwman, A.F., Van Beek, L.P.H., Mogollón, J.M., Middelburg, J.J., 2016. Global riverine N and P transport to ocean increased during the 20th century despite increased retention along the aquatic continuum. Biogeosciences 13, 2441–2451. https://doi.org/10.5194/bg-13-2441-2016.

BOBLME, 2014. Understanding Nutrient Loading and Sources in the Bay of Bengal Large Marine Ecosystem. BOBLME-2014-Ecology-18.

Boyer, E.W., Howarth, R.W., Galloway, J.N., Dentener, F.J., Green, P.A., Vörösmarty, C.J., 2006. Riverine nitrogen export from the continents to the coasts. Glob. Biogeochem. Cycles 20. https://doi.org/10.1029/2005GB002537.

Bricker, S.B., Clement, C.G., Pirhalla, D.E., Orlando, S.P., Farrow, D.R., 1999. National Estuarine Eutrophication Assessment: Effects of Nutrient Enrichment in the Nation's Estuaries. US National Oceanographic and Atmospheric Administration, National Ocean Service, Special Projects Office and the National Center for Coastal Ocean Science, Silver Spring, MD.

Bricker, S.B., Longstaff, B., Dennison, W., Jones, A., Boicourt, K., Wicks, C., Woerner, J., 2007. Effects of Nutrient Enrichment in the Nation's Estuaries: A Decade of Change. NOAA Coastal Ocean Program Decision Analysis SeriesNational Centers for Coastal Ocean Science, Silver Spring, MD.

Cai, W.-J., Hu, X., Huang, W.-J., Murrell, M.C., Lehrter, J.C., Lohrenz, S.E., Chou, W.-C., Zhai, W., Hollibaugh, J.T., Wang, Y., Zhao, P., Guo, X., Gundersen, K., Dai, M., Gong, G.-C., 2011. Acidification of subsurface coastal waters enhanced by eutrophication. Nat. Geosci. 4, 766–770. https://doi.org/10.1038/ngeo1297.

Carey, R.O., Migliaccio, K.W., 2009. Contribution of wastewater treatment plant effluents to nutrient dynamics in aquatic systems: a review. Environ. Manag. 44, 205–217. https://doi.org/10.1007/s00267-009-9309-5.

Carpenter, S.R., Caraco, N.F., Correll, D.L., Howarth, R.W., Sharpley, A.N., Smith, V.H., 1998. Nonpoint pollution of surface waters with phosphorus and nitrogen. Ecol. Appl. 8, 559–568. https://doi.org/10.1890/1051-0761(1998)008[0559:NPOSWW]2.0.CO;2.

Cloern, J., 2001. Our evolving conceptual model of the coastal eutrophication problem. Mar. Ecol. Prog. Ser. 210, 223–253. https://doi.org/10.3354/meps210223.

Coastal Protection and Restoration Authority, 2012. Louisiana's Comprehensive Master Plan for a Sustainable Coast. Coastal Protection and Restoration Authority of Louisiana, Baton Rouge, L.A.

Daloğlu, I., Cho, K.H., Scavia, D., 2012. Evaluating causes of trends in long-term dissolved reactive phosphorus loads to Lake Erie. Environ. Sci. Technol. 46, 10660–10666. https://doi.org/10.1021/es302315d.

Daniel, T.C., Sharpley, A.N., Lemunyon, J.L., 1998. Agricultural phosphorus and eutrophication: a symposium overview. J. Environ. Qual. 27, 251. https://doi.org/10.2134/jeq1998.00472425002700020002x.

Dauer, D.M., Weisberg, S.B., Ranasinghe, J.A., 2000. Relationships between benthic community condition, water quality, sediment quality, nutrient loads, and land use patterns in Chesapeake Bay. Estuaries 23, 80. https://doi.org/10.2307/1353227.

David, M.B., Drinkwater, L.E., McIsaac, G.F., 2010. Sources of nitrate yields in the Mississippi River basin. J. Environ. Qual. 39, 1657. https://doi.org/10.2134/jeq2010.0115.

De Cicco, L., Sprague, L.A., Murphy, J.C., Oelsner, G.P., Johnson, H.M., Falcone, J.A., Stets, E.G., 2017. Water-Quality and Streamflow Datasets Used in the Weighted Regressions on Time, Discharge, and Season (WRTDS) Models to Determine Trends in the Nation's Rivers and Streams, 1972–2012: U.S. Geological Survey Data Release. https://doi.org/10.5066/F7KW5D4H.

Diaz, R.J., Rosenberg, R., 2008. Spreading dead zones and consequences for marine ecosystems. Science 321, 926–929. https://doi.org/10.1126/science.1156401.

Elser, J.J., Bracken, M.E.S., Cleland, E.E., Gruner, D.S., Harpole, W.S., Hillebrand, H., Ngai, J.T., Seabloom, E.W., Shurin, J.B., Smith, J.E., 2007. Global analysis of nitrogen and phosphorus limitation of primary producers in freshwater, marine and terrestrial ecosystems. Ecol. Lett. 10, 1135–1142. https://doi.org/10.1111/j.1461-0248.2007.01113.x.
Eshleman, K.N., Sabo, R.D., Kline, K.M., 2013. Surface water quality is improving due to declining atmospheric N deposition. Environ. Sci. Technol. 47, 12193–12200. https://doi.org/10.1021/es4028748.
Falcone, J.A., 2015. U.S. conterminous wall-to-wall anthropogenic land use trends (NWALT), 1974–2012: U.S. Geological Survey Data Series 948. https://doi.org/10.3133/ds948 33 p. plus appendixes 3–6 as separate files.
Falcone, J.A., 2017. Watershed Characteristics for Study Sites of the Surface Water Trends Project. National Water Quality Program: U.S. Geological Survey Data Release https://doi.org/10.5066/F7TX3CKP.
Fanelli, R.M., Blomquist, J.D., Hirsch, R.M., 2019. Point sources and agricultural practices control spatial-temporal patterns of orthophosphate in tributaries to Chesapeake Bay. Sci. Total Environ. 652, 422–433. https://doi.org/10.1016/j.scitotenv.2018.10.062.
Fisher, T.R., Hagy, J.I.D., Boynton, W.R., Williams, M.R., 2006. Cultural eutrophication in the Choptank and Patuxent estuaries of Chesapeake Bay. Limnol. Oceanogr. 51, 435–447. https://doi.org/10.4319/lo.2006.51.1_part_2.0435.
Foley, J.A., 2005. Global consequences of land use. Science 309, 570–574. https://doi.org/10.1126/science.1111772.
Galloway, J.N., Dentener, F.J., Capone, D.G., Boyer, E.W., Howarth, R.W., Seitzinger, S.P., Asner, G.P., Cleveland, C.C., Green, P.A., Holland, E.A., Karl, D.M., Michaels, A.F., Porter, J.H., Townsend, A.R., Vorosmarty, C.J., 2004. Nitrogen cycles: past, present, and future. Biogeochemistry 70, 153–226. https://doi.org/10.1007/s10533-004-0370-0.
Gray, J.R., Glysson, G.D., Turcios, L.M., Schwarz, G.E., 2000. Comparability of suspended-sediment concentration and total suspended solids data: U.S. Geological Survey Water-Resources Investigations Report 2000–4191. https://doi.org/10.3133/wri004191 14 p.
Grizzetti, B., Bouraoui, F., Aloe, A., 2012. Changes of nitrogen and phosphorus loads to European seas. Glob. Chang. Biol. 18, 769–782. https://doi.org/10.1111/j.1365-2486.2011.02576.x.
HELCOM, 2018. Sources and pathways of nutrients to the Baltic Sea. Baltic Sea Environment Proceedings No. 153 Available online at. http://www.helcom.fi/Lists/Publications/BSEP153.pdf.
Hickman, R.E., Hirsch, R.M., 2017. Trends in the quality of water in New Jersey streams, water years 1971–2011: U.S. Geological Survey Scientific Investigations Report 2016–5176. https://doi.org/10.3133/sir20165176 58 p.
Hirsch, R.M., 2012. Flux of nitrogen, phosphorus, and suspended sediment from the Susquehanna River basin to the Chesapeake Bay during tropical storm lee, September 2011, as an indicator of the effects of reservoir sedimentation on water quality: U.S. Geological Survey Scientific Investigations Report 2012–5185. 17 p. Available online at. https://pubs.usgs.gov/sir/2012/5185/.
Hirsch, R.M., De Cicco, L.A., 2015. User guide to Exploration and Graphics for RivEr Trends (EGRET) and dataRetrieval: R packages for hydrologic data (version 2.0, February 2015): U.S. Geological Survey Techniques and Methods book 4, chap. A10. https://doi.org/10.3133/tm4A10 93 p.
Hirsch, R.M., Moyer, D.L., Archfield, S.A., 2010. Weighted regressions on time, discharge, and season (WRTDS), with an application to Chesapeake Bay river inputs. J. Am. Water Resour. Assoc. 46, 857–880. https://doi.org/10.1111/j.1752-1688.2010.00482.x.
Hirsch, R.M., Archfield, S.A., De Cicco, L.A., 2015. A bootstrap method for estimating uncertainty of water quality trends. Environ. Model. Softw. 73, 148–166. https://doi.org/10.1016/j.envsoft.2015.07.017.
Hogan, D.M., 2008. Management of urban stormwater runoff in the Chesapeake Bay watershed: U.S. Geological Survey Fact Sheet 2008–3101. 3 p. Available online at. https://pubs.usgs.gov/fs/2008/3101.
Horowitz, A.J., 2009. A quarter century of declining suspended sediment fluxes in the Mississippi River and the effect of the 1993 flood. Hydrol. Process. https://doi.org/10.1002/hyp.7425.
Horowitz, A.J., Stephens, V.C., Elrick, K.A., Smith, J.J., 2012. Concentrations and annual fluxes of sediment-associated chemical constituents from conterminous US coastal rivers using bed sediment data. Hydrol. Process. 26, 1090–1114. https://doi.org/10.1002/hyp.8437.
Howarth, R.W., Marino, R., 2006. Nitrogen as the limiting nutrient for eutrophication in coastal marine ecosystems: evolving views over three decades. Limnol. Oceanogr. 51, 364–376. https://doi.org/10.4319/lo.2006.51.1_part_2.0364.
Howarth, R.W., Anderson, D.B., Cloern, J.E., Elfring, C., Hopkinson, C.S., Lapointe, B., Maloney, T.J., Marcus, N., McGlathery, K., Sharpley, A.N., Walker, D., 2000. Issues in ecology: nutrient pollution of coastal rivers, bays, and seas. Issues Ecol. 7 (17 p).
Howarth, R.W., Sharpley, A., Walker, D., 2002. Sources of nutrient pollution to coastal waters in the United States: implications for achieving coastal water quality goals. Estuaries 25, 656–676. https://doi.org/10.1007/BF02804898.
Howarth, R.W., Chan, F., Conley, D.J., Garnier, J., Doney, S.C., Marino, R., Billen, G., 2011. Coupled biogeochemical cycles: eutrophication and hypoxia in temperate estuaries and coastal marine ecosystems. Front. Ecol. Environ. 9, 18–26. https://doi.org/10.1890/100008.
Jarchow, C.J., Nagler, P.L., Glenn, E.P., Ramírez-Hernández, J., Rodríguez-Burgueño, J.E., 2017. Evapotranspiration by remote sensing: an analysis of the Colorado River Delta before and after the minute 319 pulse flow to Mexico. Ecol. Eng. 106, 725–732. https://doi.org/10.1016/j.ecoleng.2016.10.056.
Justić, D., Rabalais, N.N., Turner, R.E., 2002. Modeling the impacts of decadal changes in riverine nutrient fluxes on coastal eutrophication near the Mississippi River Delta. Ecol. Model. 152, 33–46. https://doi.org/10.1016/S0304-3800(01)00472-0.
Keown, M.P., Dardeau, E.A., Causey, E.M., 1986. Historic trends in the sediment flow regime of the Mississippi River. Water Resour. Res. 22, 1555–1564. https://doi.org/10.1029/WR022i011p01555.
Liner, B., Lueckenhoff, D., McNally, D., Brown, S., 2017. Chesapeake Bay Progress: Wastewater Pollution Reduction Leads the Way. Presented at the American Water Works Association Annual Conference & Exhibition (ACE) 2017, Philadelphia, PA.
Litke, D.W., 1999. Review of phosphorus control measures in the United States and their effects on water quality: U.S. Geological Survey Water-Resources Investigations Report 99-4007. https://doi.org/10.3133/wri994007.
Liu, S.M., Hong, G.-H., Zhang, J., Ye, X.W., Jiang, X.L., 2009. Nutrient budgets for large Chinese estuaries. Biogeosciences 6, 2245–2263. https://doi.org/10.5194/bg-6-2245-2009.
Lotze, H.K., 2006. Depletion, degradation, and recovery potential of estuaries and coastal seas. Science 312, 1806–1809. https://doi.org/10.1126/science.1128035.
Ludwig, W., Bouwman, A.F., Dumont, E., Lespinas, F., 2010. Water and nutrient fluxes from major Mediterranean and Black Sea rivers: past and future trends and their implications for the basin-scale budgets. Glob. Biogeochem. Cycles 24. https://doi.org/10.1029/2009GB003594.
Mallin, M.A., Williams, K.E., Esham, E.C., Lowe, R.P., 2000. Effect of human development on bacteriological water quality in coastal watersheds. Ecol. Appl. 10, 1047–1056. https://doi.org/10.1890/1051-0761(2000)010[1047:EOHDOB]2.0.CO;2.
Mayorga, E., Seitzinger, S.P., Harrison, J.A., Dumont, E., Beusen, A.H.W., Bouwman, A.F., Fekete, B.M., Kroeze, C., Van Drecht, G., 2010. Global nutrient export from WaterSheds 2 (NEWS 2): model development and implementation. Environ. Model. Softw. 25, 837–853. https://doi.org/10.1016/j.envsoft.2010.01.007.
McKay, L., Bondelid, T., Dewald, T., Johnston, C., Moore, R., Rea, A., 2012. NHDPlus version 2—User guide, 2012. U.S. Environmental Protection Agency, http://www.horizon-systems.com/NHDPlus/index.php.
Meade, R.H., Moody, J.A., 2009. Causes for the decline of suspended-sediment discharge in the Mississippi River system, 1940-2007. Hydrol. Process. https://doi.org/10.1002/hyp.7477.
Moyer, D.L., Blomquist, J.D., 2017. Summary of Nitrogen, Phosphorus, and Suspended-Sediment Loads and Trends Measured at the Chesapeake Bay Nontidal Network Stations: Water Year 2016 Update.
Muenich, R.L., Kalcic, M., Scavia, D., 2016. Evaluating the impact of legacy P and agricultural conservation practices on nutrient loads from the Maumee River watershed. Environ. Sci. Technol. 50, 8146–8154. https://doi.org/10.1021/acs.est.6b01421.
Murphy, J.C., Hirsch, R.M., Sprague, L.A., 2013. Nitrate in the Mississippi River and its tributaries, 1980–2010: an update. U.S. Geological Survey Scientific Investigations Report 2013–5169. https://doi.org/10.3133/sir20135169 31 p.
National Oceanic and Atmospheric Administration, 2007. Coastal Assessment Framework (CAF) NOAA/NOS Special Projects Office-Coastal Geospatial Data Project. Digital Geospatial Data. Silver Spring, MD. (Current version at https://www.ncei.noaa.gov/).
National Oceanic and Atmospheric Administration, 2013. National Coastal Population Report Population Trends from 1970 to 2020, NOAA State of the Coast Report Series. https://aamboceanservice.blob.core.windows.net/oceanservice-prod/facts/coastal-population-report.pdf.
National Research Council, 2000. Clean Coastal Waters: Understanding and Reducing the Effects of Nutrient Pollution. National Academies Press, Washington, D.C. https://doi.org/10.17226/9812.
Oelsner, G.P., Sprague, L.A., Murphy, J.C., Zuellig, R.E., Johnson, H.M., Ryberg, K.R., Falcone, J.A., Stets, E.G., Vecchia, A.V., Riskin, M.L., De Cicco, L.A., Mills, T.J., Farmer, W.H., 2017. Water-quality trends in the nation's rivers and streams, 1972–2012—data preparation, statistical methods, and trend results: U.S. Geological Survey Scientific Investigations Report. https://doi.org/10.3133/sir20175006 136 p.
Orth, R.J., Carruthers, T.J.B., Dennison, W.C., Duarte, C.M., Fourqurean, J.W., Heck, K.L., Hughes, A.R., Kendrick, G.A., Kenworthy, W.J., Olyarnik, S., Short, F.T., Waycott, M., Williams, S.L., 2006. A global crisis for seagrass ecosystems. Bioscience 56, 987. https://doi.org/10.1641/0006-3568(2006)56[987:AGCFSE]2.0.CO;2.
Preston, S.D., Alexander, R.B., Schwarz, G.E., Crawford, C.G., 2011. Factors affecting stream nutrient loads: a synthesis of regional SPARROW model results for the continental United States 1: factors affecting stream nutrient loads: a synthesis of regional SPARROW model results for the continental United States. J. Am. Water Resour. Assoc. 47, 891–915. https://doi.org/10.1111/j.1752-1688.2011.00577.x.
Redfield, A.C., 1958. The biological control of chemical factors in the environment. Am. Sci. 11, 205–221.
Rice, K.C., Moyer, D.L., Mills, A.L., 2017. Riverine discharges to Chesapeake Bay: analysis of long-term (1927–2014) records and implications for future flows in the Chesapeake Bay basin. J. Environ. Manag. 204, 246–254. https://doi.org/10.1016/j.jenvman.2017.08.057.
Robertson, D.M., Saad, D.A., 2013. SPARROW models used to understand nutrient sources in the Mississippi/Atchafalaya River basin. J. Environ. Qual. 42, 1422. https://doi.org/10.2134/jeq2013.02.0066.
Romero, E., Garnier, J., Lassaletta, L., Billen, G., Le Gendre, R., Riou, P., Cugier, P., 2013. Large-scale patterns of river inputs in southwestern Europe: seasonal and interannual variations and potential eutrophication effects at the coastal zone. Biogeochemistry 113, 481–505. https://doi.org/10.1007/s10533-012-9778-0.
Seitzinger, S.P., Harrison, J.A., Dumont, E., Beusen, A.H.W., Bouwman, A.F., 2005. Sources and delivery of carbon, nitrogen, and phosphorus to the coastal zone: an overview of global nutrient export from watersheds (NEWS) models and their application. Glob. Biogeochem. Cycles 19. https://doi.org/10.1029/2005GB002606.
Seitzinger, S.P., Mayorga, E., Bouwman, A.F., Kroeze, C., Beusen, A.H.W., Billen, G., Van Drecht, G., Dumont, E., Fekete, B.M., Garnier, J., Harrison, J.A., 2010. Global river nutrient export: a scenario analysis of past and future trends. Glob. Biogeochem. Cycles 24. https://doi.org/10.1029/2009GB003587.

Sharples, J., Middelburg, J.J., Fennel, K., Jickells, T.D., 2017. What proportion of riverine nutrients reaches the open ocean?: riverine nutrients reaching the ocean. Glob. Biogeochem. Cycles 31, 39–58. https://doi.org/10.1002/2016GB005483.

Sparkman, S.A., Hogan, D.M., Hopkins, K.G., Loperfido, J.V., 2017. Modeling watershed-scale impacts of Stormwater management with traditional versus low impact development design. J. Am. Water Resour. Assoc. 53, 1081–1094. https://doi.org/10.1111/1752-1688.12559.

Sprague, L.A., Hirsch, R.M., Aulenbach, B.T., 2011. Nitrate in the Mississippi River and its tributaries, 1980 to 2008: are we making Progress? Environ. Sci. Technol. 45, 7209–7216. https://doi.org/10.1021/es201221s.

Stoddard, J.L., Van Sickle, J., Herlihy, A.T., Brahney, J., Paulsen, S., Peck, D.V., Mitchell, R., Pollard, A.I., 2016. Continental-scale increase in Lake and stream phosphorus: are oligotrophic systems disappearing in the United States? Environ. Sci. Technol. 50, 3409–3415. https://doi.org/10.1021/acs.est.5b05950.

Stow, C.A., Cha, Y., Johnson, L.T., Confesor, R., Richards, R.P., 2015. Long-term and seasonal trend decomposition of Maumee River nutrient inputs to Western Lake Erie. Environ. Sci. Technol. 49, 3392–3400. https://doi.org/10.1021/es5062648.

Syvitski, J.P.M., 2005. Impact of humans on the flux of terrestrial sediment to the global Coastal Ocean. Science 308, 376–380. https://doi.org/10.1126/science.1109454.

Thrush, S., Hewitt, J., Cummings, V., Ellis, J., Hatton, C., Lohrer, A., Norkko, A., 2004. Muddy waters: elevating sediment input to coastal and estuarine habitats. Front. Ecol. Environ. 2, 299–306. https://doi.org/10.1890/1540-9295(2004)002[0299:MWESIT]2.0.CO;2.

Turner, R.E., Rabalais, N.N., 2003. Linking landscape and water quality in the Mississippi River basin for 200 years. Bioscience 53, 563. https://doi.org/10.1641/0006-3568(2003)053[0563:LLAWQI]2.0.CO;2.

U.S. Environmental Protection Agency, 2012. National Coastal Condition Report IV (Office of Research and Development/Office of Water No. EPA-842-R-10-003). Washington, D.C. https://www.epa.gov/national-aquatic-resource-surveys/national-coastal-condition-report-iv-2012.

U.S. Environmental Protection Agency, 2016. National Coastal Condition Assessment 2010 (EPA 841-R-15-006) (Office of Water and Office of Research and Development). Washington, D.C. http://www.epa.gov/national-aquatic-resource-surveys/ncca.

U.S. Environmental Protection Agency, 2017. 2015 Program Progress - Cross-State Air Pollution Rule and Acid Rain Program. Washington, D.C. https://www3.epa.gov/airmarkets/progress/reports/index.html.

Van Meter, K.J., Basu, N.B., Van Cappellen, P., 2017. Two centuries of nitrogen dynamics: legacy sources and sinks in the Mississippi and Susquehanna River Basins. Glob. Biogeochem. Cycles 31, 2–23. https://doi.org/10.1002/2016GB005498.

Vitousek, P.M., Naylor, R., Crews, T., David, M.B., Drinkwater, L.E., Holland, E., Johnes, P.J., Katzenberger, J., Martinelli, L.A., Matson, P.A., Nziguheba, G., Ojima, D., Palm, C.A., Robertson, G.P., Sanchez, P.A., Townsend, A.R., Zhang, F.S., 2009. Nutrient Imbalances in Agricultural Development. Science 324, 1519–1520. https://doi.org/10.1126/science.1170261.

Walling, D.E., Collins, A.L., 2008. The catchment sediment budget as a management tool. Environ. Sci. Pol. 11, 136–143. https://doi.org/10.1016/j.envsci.2007.10.004.

Watson, S.B., Miller, C., Arhonditsis, G., Boyer, G.L., Carmichael, W., Charlton, M.N., Confesor, R., Depew, D.C., Höök, T.O., Ludsin, S.A., Matisoff, G., McElmurry, S.P., Murray, M.W., Richards, R.P., Rao, Y.R., Steffen, M.M., Wilhelm, S.W., 2016. The re-eutrophication of Lake Erie: harmful algal blooms and hypoxia. Harmful Algae 56, 44–66. https://doi.org/10.1016/j.hal.2016.04.010.

Waycott, M., Duarte, C.M., Carruthers, T.J.B., Orth, R.J., Dennison, W.C., Olyarnik, S., Calladine, A., Fourqurean, J.W., Heck, K.L., Hughes, A.R., Kendrick, G.A., Kenworthy, W.J., Short, F.T., Williams, S.L., 2009. Accelerating loss of seagrasses across the globe threatens coastal ecosystems. Proc. Natl. Acad. Sci. 106, 12377–12381. https://doi.org/10.1073/pnas.0905620106.

Zhang, Q., Hirsch, R.M., Ball, W.P., 2016. Long-term changes in sediment and nutrient delivery from Conowingo dam to Chesapeake Bay: effects of reservoir sedimentation. Environ. Sci. Technol. 50, 1877–1886. https://doi.org/10.1021/acs.est.5b04073.

Zhang, Q., Murphy, R.R., Tian, R., Forsyth, M.K., Trentacoste, E.M., Keisman, J., Tango, P.J., 2018. Chesapeake Bay's water quality condition has been recovering: insights from a multimetric indicator assessment of thirty years of tidal monitoring data. Sci. Total Environ. 637–638, 1617–1625. https://doi.org/10.1016/j.scitotenv.2018.05.025.