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Science of the Total Environment 452–453 (2013) 208–221

Contents lists available at SciVerse ScienceDirect

Science of the Total Environment

journal homepage: www.elsevier.com/locate/scitotenv

Long-term seasonal trends of nitrogen, phosphorus, and suspended sediment load from the non-tidal Susquehanna River Basin to Chesapeake Bay

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HIGHLIGHTS

► Flow-normalized loads of N, P, and SS from the Susquehanna River were evaluated.
► SS and particulate-bound P and N from the Susquehanna to Chesapeake Bay are rising.
► N, P, and SS loads have declined in the Susquehanna River above its major reservoirs.
► The Conowingo Reservoir has neared its capacity to trap SS and particulate P and N.
► The reservoir will pose challenges to attainment of nutrient and sediment reduction.

ARTICLE INFO

Article history:
Received 23 August 2012
Received in revised form 8 February 2013
Accepted 8 February 2013
Available online 19 March 2013

Keywords:
Susquehanna River Basin
Chesapeake Bay
Nutrient and sediment loadings
Long-term seasonal trends
Flow normalization

ABSTRACT

Reduction of nitrogen (N), phosphorus (P), and suspended sediment (SS) load has been a principal focus of Chesapeake Bay Watershed management for decades. To evaluate the progress of management actions in the Bay's largest tributary, the Susquehanna River, we analyzed the long-term seasonal trends of flow-normalized N, P, and SS load over the last two to three decades, both above and below the Lower Susquehanna River Reservoir System. Our results indicate that annual and decadal-scale trends of nutrient and sediment load generally followed similar patterns in all four seasons, implying that changes in watershed function and land use had similar impacts on nutrient and sediment load at all times of the year. Above the reservoir system, the combined loads from the Marietta and Conestoga Stations indicate general trends of N, P, and SS reduction in the Susquehanna River Basin, which can most likely be attributed to a suite of management actions on point, agricultural, and stormwater sources. In contrast, upward trends of SS and particulate-associated P and N were generally observed below the Conowingo Reservoir since the mid-1990s. Our analyses suggest that (1) the reservoirs' capacity to trap these materials has been diminishing over the past two to three decades, and especially so for SS and P since the mid-1990s, and that (2) the Conowingo Reservoir has already neared its sediment storage capacity. These changes in reservoir performance will pose significant new kinds of challenges to attainment of total maximum daily load goals for the Susquehanna River Basin, and particularly if also accompanied by increases in storm frequency and intensity due to climate change. Accordingly, the reservoir issue may need to be factored into the proper establishment of regulatory load requirements and the development of watershed implementation plans.

1. Introduction

Chesapeake Bay has experienced persistent summertime hypoxia in its bottom waters that has been attributed to a combination of anthropogenic nutrient inputs from the watershed (Boynton and Kemp, 2000; Hagy et al., 2004; Kemp et al., 2005; Malone et al., 1988; Murphy et al., 2011) and naturally occurring vertical stratification (Boicourt, 1992; Murphy et al., 2011; Pritchard and Schubel, 2001). On one hand, high nutrient inputs – primarily nitrogen (N) and phosphorus (P) – can stimulate phytoplankton growth that can exert considerable biochemical oxygen demand when the algal matter sinks to the deep channel (Boynton and Kemp, 2000; Cloern, 2001; Kemp et al., 2009). On the other hand, freshwater flow acts to strengthen water column stratification that can isolate the deep water hypoxic zones, thus preventing oxygen replenishment from the surface water (Boicourt, 1992; Goodrich et al., 1987; Pritchard and Schubel, 2001). In addition, suspended sediment (SS) can reduce light penetration and thus inhibit the growth of beneficial submerged aquatic vegetation (Brakebill et al., 2010). Of the two influences, anthropogenic inputs and stratification, the more attainable means of controlling hypoxia is the reduction of N, P, and SS load, and this has been a principal focus of Chesapeake Bay Watershed (CBW) management for decades. In 2010, the strength of this endeavor was

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http://dx.doi.org/10.1016/j.scitotenv.2013.02.012

increased, with the introduction of regulations of total maximum daily loads (TMDLs) for N, P, and SS (US Environmental Protection Agency, 2010).

To aid the assessment of reduction progress, the U.S. Geological Survey (USGS) and collaborators have been collecting and analyzing water quality data at many monitoring sites in the CBW for decades (Langland et al., 2007; Sprague et al., 2000). For example, the USGS River Input Monitoring (RIM) Program has been monitoring streamflow and water quality at nine stations at the fall-line of major tributaries since the mid-1980s (US Geological Survey, 2012a). In a comprehensive study, Langland et al. (2007) detected significantly decreasing trends in flow-adjusted annual concentration of total N (TN), total P (TP), and SS from 1985 to 2006 at about 74%, 68%, and 32% of 34 selected monitoring sites in the CBW, respectively.

As part of on-going efforts to analyze load trends in the major tributaries to Chesapeake Bay, this study focuses on the Susquehanna River because it is the largest tributary in terms of freshwater discharge (60%), TN load (62%), and TP load (34%) (Belval and Sprague, 1999). Encouragingly, McGonigal (2010) detected significantly decreasing trends in flow-adjusted annual concentration of TN, TP, and SS at most monitoring sites in the Susquehanna River Basin (SRB) from 1986 to 2009 (Fig. 1). Consistent with these findings, a later study by Langland et al. (2012) also reported generally decreasing trends of flow-adjusted annual concentration of these pollutants in the SRB from 1985 to 2010. In addition, Langland et al. (2012) reported reducing trends of TN and SS, but non-significant changes in TP at the Conowingo Station at the fall-line of the Susquehanna River. A common feature of these studies is the adoption of annual resolution in trend analyses. However, in order to capture impacts of seasonality such as variations in temperature and rainfall, fertilizer application, and benthic recycling of P and denitrification in river channels and reservoirs, seasonal trends of these anthropogenic pollutants need to be investigated, and attention needs to be given to calculations not only of concentration but also of load, which is more complex because of the need to temporally match concentration with flow.

With regard to prior estimates of historical load, the USGS has been using a tool developed in 1989 called ESTIMATOR (Cohn et al., 1989) to compute and report daily nutrient and sediment load in Chesapeake Bay tributaries. In recent work, Murphy et al. (2011) combined some of these loading estimates for the Susquehanna River at Conowingo with some interpolations and extrapolations of upstream data that had been previously developed by Hagy et al. (2004) to plot a 60-year history of Susquehanna winter-spring (i.e., Jan–May) TN load, which has been of

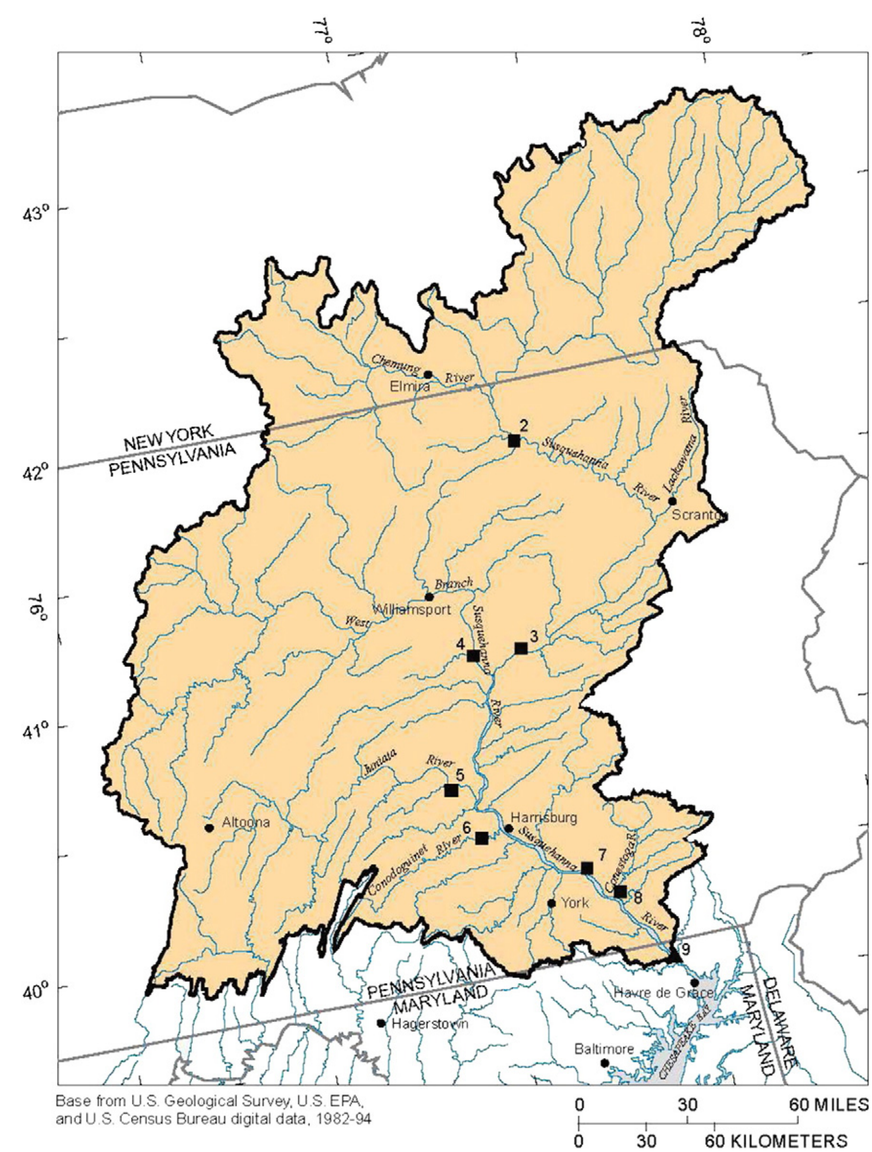

Fig. 1. Map of Susquehanna River Basin and long-term monitoring sites. Sites in this study include two main-stem stations, Marietta (No. 7) and Conowingo (No. 9), and one tributary station, Conestoga (No. 8). The non-tidal Susquehanna River Basin, shaded in yellow, covers portions of New York, Pennsylvania, and Maryland.
This figure was reproduced from Fig. 6 in Sprague et al. (2000) with permission.

special interest because of its reported correlation with summer-time oxygen depletion in Chesapeake Bay (Hagy et al., 2004). The 60-year TN load history (*cf.* Fig. 3c of Murphy et al. (2011)), although replete with flow-related inter-annual variations, suggests a general decreasing trend of Jan–May TN load in recent decades. In addition, Murphy et al. (2011) have reported correlation between the Jan–May TN loads and bay hypoxia during the late-summer period (by contrast, long-term trends in early-summer hypoxia were found to be correlated with some long-term flow-unrelated trends of increasing stratification). Because the reported correlation is dependent on good understanding of the TN load history, it is important that such trends be evaluated using the best tools available.

When considering overall riverine inputs of nutrient and sediment, it is also important to recognize that such loadings can be greatly complicated by impacts of sediment retention and release in reservoirs. In particular, reservoirs in early stages of their lifespan can effectively retain sediment and associated N and P (Friedl and Wüest, 2002; Jossette et al., 1999; Medeiros et al., 2011), thus providing efficient removal of N and P from streamflow, mainly through denitrification and particle deposition, respectively (Friedl and Wüest, 2002; Jossette et al., 1999). In the SRB, the most studied reservoir system is probably the Lower Susquehanna River Reservoir System (hereafter, "the reservoir system"), which includes Lake Clarke, Lake Aldred, and the Conowingo Reservoir (Fig. 2) (Langland, 2009; Langland and Hainly, 1997). The system has been reported to trap about 2%, 45%, and 70% of annual TN, TP, and SS load, respectively, from the Susquehanna River to the bay (Langland and Hainly, 1997), thus alleviating the pollutant load considerably. As the most downstream and the largest reservoir in the system, the Conowingo Reservoir (hereafter, "the reservoir") is the only one that was reported not to have reached its sediment storage capacity (SSC) (Langland, 2009). Based on assumptions in SS input load and sediment deposition rate in the reservoir, Langland (2009) estimated an additional service life of 15–20 years (from 2009) before the reservoir would be filled up. By that time, annual loads of TN, TP, and SS to the bay have been projected to increase by 2%, 70%, and 250%, respectively (Langland and Hainly, 1997). In addition, a previous study has suggested that a flow of 400,000 ft^3/s (11,300 m^3/s) was the "scour threshold" for the Conowingo Reservoir, and that major floods above this level would further increase nutrient and sediment delivery to Chesapeake Bay (Langland and Hainly, 1997). More recently, Hirsch (2012a) has analyzed discharge and water-quality data at the Conowingo Dam, and detected upward trends in annual TP and SS load. The author has evaluated some hypotheses and presented a "scour hypothesis" as a most likely explanation to the observed trends. This hypothesis states that when reservoirs are near capacity, the water channel would become smaller, thus resulting in faster water flow, and correspondingly, higher likelihood of scour

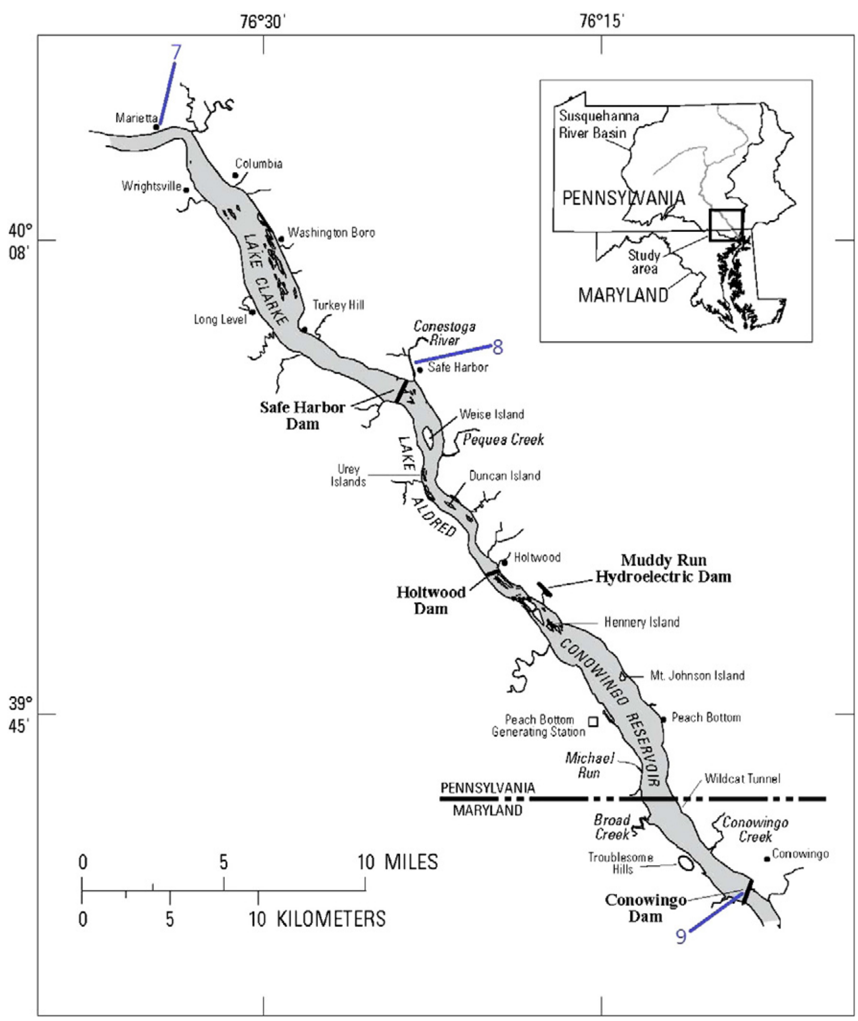

Fig. 2. Map of Lower Susquehanna River Reservoir System and the study sites. The reservoir system consists of Lake Clarke, Lake Aldred, and the Conowingo Reservoir. The Marietta Station (No. 7) is just above the reservoirs and the Conowingo Station (No. 9) is just below the reservoirs. The Conestoga Station (No. 8) monitors streamflow from the Conestoga River, a major tributary to the Susquehanna River. See Fig. 1 of this article for locations of the three sites in the non-tidal Susquehanna River Basin.
This figure was adapted from Fig. 1 in Langland (2009) with permission.

(Hirsch, 2012a). Therefore, increased net scouring of sediment in the Conowingo Reservoir may already be occurring at flow rates much lower than the above-cited scour threshold (Blankenship, 2012). Hirsch (2012a) has suggested that filling processes in the Conowingo Reservoir are already approaching a final asymptotic stage and that further monitoring and evaluation of the reservoir performance are critical to evaluation of management plans.

In the above context, the work described herein was undertaken concurrently with the efforts of Hirsch (2012a) and with the goal of more closely examining many issues raised in that work and similarly motivated by prior studies. Particular new contributions of this work are as follows:

(1) analysis of multiple N and P species to examine potential differences as related to particulate versus dissolved fractions;
(2) direct comparison of above- and below-reservoir data at multiple sites to evaluate changes in sediment and nutrient inventory within the reservoirs;
(3) application of a new and reportedly more accurate loading estimation method called "weighted regressions on time, discharge, and season (WRTDS) (Hirsch et al., 2010)" to develop the first set of estimates for sites above the reservoirs;
(4) analysis of flow-normalized trends to better understand long-term trends independent of random streamflow. The WRTDS method is capable of providing both "true-condition" and "flow-normalized" estimates of concentration and load. In comparison, most previous studies (e.g., Langland et al., 2012, 2007; McGonigal, 2010) have used the ESTIMATOR model, which is not able to produce the flow-normalized trends; and
(5) analysis of seasonal loads and trends using both the "true-condition" and "flow-normalized" approaches. Most previous studies of SRB sites (e.g., Hirsch, 2012a; Hirsch et al., 2010; Langland et al., 2012, 2007; McGonigal, 2010) have focused on analysis of annual loads and trends.

More specifically, we have examined the long-term seasonal history of N, P, and SS loads in the Susquehanna River, both above and below the reservoir system, through the following three broad types of analysis:

(1) reconstruction of our best understanding of the long-term history of concentrations and loads of nitrate (NO_x) and loads of TN from the Susquehanna River to Chesapeake Bay for a 67-year period (1945–2011), in terms of both "true-condition" and "flow-normalized" estimates, using the latest available method (WRTDS) and the longest available records of concentration and flow at the Conowingo Station;
(2) estimation of seasonal "flow-normalized" loads for multiple species of nutrients and sediment for two locations just above the reservoir system for a 26-year period (1986–2011) over which relevant concentration and flow data are available – major species studied include SS, TP, particulate P (PP), dissolved P (DP), TN, particulate N (PN), and dissolved N (DN); and
(3) similar estimation of seasonal "flow-normalized" loads for the same species of nutrients and sediment at the Conowingo Station for a 34-year period (1978–2011) over which relevant data are available.

Our objectives in undertaking these analyses were as follows:

(1) to compare long-term trends in N loading with prior estimates published by Murphy et al. (2011) and to thus verify whether the long-term trends are still apparent;
(2) to evaluate progress in reduction of N, P, and SS load from the non-tidal SRB at seasonal resolution; and
(3) to compare the relative changes in N, P, and SS loads discharging into and emanating from the reservoirs at seasonal resolution, thus allowing an evaluation of reservoir performance and service life in terms of sediment and nutrient retention.

2. Methods

2.1. Study sites

The RIM station at the Conowingo Dam is about 10 miles from the Susquehanna River mouth and receives 99% of the streamflow from the SRB (Fig. 1) (Belval and Sprague, 1999). This station is also located at the river fall-line, a physical fall that provides distinct separation of the tidal and non-tidal basins. Upstream, seven additional sites at Towanda, Danville, Lewisburg, Newport, Hogestown, Marietta, and Conestoga have been monitored by the Susquehanna River Basin Commission (SRBC) through the Susquehanna Nutrient Assessment Program (SNAP) since the mid-1980s (Susquehanna River Basin Commission, 2012). Since all the stations are above the fall-line (i.e., not influenced by tides), trends observed there can be used to assess nutrient and sediment reduction progress in their respective upstream watersheds within the SRB (Sprague et al., 2000).

Sites examined in the present study include the RIM station at Conowingo and two SNAP stations at Marietta and Conestoga (Table 1). The Marietta Station is the most downstream SNAP station on the river mainstem and represents the vast majority (~96%) of the watershed area represented by the Conowingo Station and with a median streamflow that is slightly higher (Table 1). However, one major distinction between the two stations is their locations relative to the Lower Susquehanna River Reservoir System — Marietta is upstream and Conowingo is downstream of the reservoirs (Fig. 2). The Conestoga Station on the Conestoga River (a major tributary to the Susquehanna located between Marietta and Conowingo; Fig. 2), monitors surface runoff from the small but heavily agricultural Conestoga basin (Table 1). In general, the combined nutrient and sediment load from the Marietta and Conestoga Stations represents a majority of input to the reservoirs, whereas load at Conowingo represents the output. Comparisons between the input and output are thus well suited for examining the possible impacts of the reservoirs on long-term seasonal trends of nutrient and sediment loads.

2.2. Statistical methods

Because of concomitant constraints on labor, time, and funding, water-quality samples have been collected only once or several times

Table 1
Details of the study sites[a].

USGS ID	Station name	Upstream land area (mi^2)	Upstream land use (percent)				Flow statistics[b] (cubic feet per second)		
			Urban	Agricultural	Forested	Other	Min	Median	Max
01576000	Susquehanna River at Marietta, PA	25,990	4	30	64	2	24,370	36,280	63,560
01576754	Conestoga River at Conestoga, PA	470	14	60	23	3	217	664	1140
01578310	Susquehanna River near Conowingo, MD	27,100	2	29	67	2	23,560	35,575	65,540

[a] Modified from Table 4 in Sprague et al. (2000) with permission.
[b] Calculated based on annual average flow data from 1985 to 2010 (US Geological Survey, 2012b).

each month at the study sites. Therefore, appropriate statistical methods are required to make predictions for unsampled days. Selection of best methods of estimation for nutrient and sediment concentrations and loads based on available monitoring data has been an important topic of discussion since at least the late 1980s (Cohn et al., 1989). To date, the USGS has been applying an estimation tool known as the ESTIMATOR model (Cohn et al., 1989) to estimate daily nutrient and sediment concentration and load in Chesapeake Bay tributaries. More recently, however, Hirsch et al. (2010) have described the need for new statistical methods that can both (a) better describe temporal variations in concentration and load, and (b) more effectively remove the influence of random flow variation. More importantly, the new methods should not rely on questionable assumptions such as a constant concentration-flow relation, constant seasonal trends in concentration, or the existence of specific functional forms of these trends (Hirsch et al., 2010). Hirsch et al. (2010) incorporated these considerations into the development of the WRTDS method, which has recently been applied to several large data sets and is fully described elsewhere, e.g., Hirsch et al. (2010), Sprague et al. (2011). For the convenience of readers here, we have briefly summarized the basic structure and application of WRTDS in Appendix A.

The WRTDS method produces two types of estimates for both concentration and load – so-called "true-condition" and "flow-normalized" estimates, as described in more detail in Appendix A. Hirsch et al. (2010) have pointed out that the true-condition estimates are useful to help understand the real history of riverine nutrient (or sediment) and downstream ecological impact, whereas the flow-normalized estimates are more helpful to evaluate management progress in the watershed — i.e. with respect to nutrient loading factors that are less related to river-flow. The flow-normalization algorithm, described in more detail in Appendix A, can greatly remove the sometimes dramatic influence of random variations in streamflow by linking the estimation to the full history of hydrological flows over long-term cycles, thus rendering longer-term inter-annual trends easier to detect and understand than they would be with true-condition estimates. For these reasons, we have focused most of our attention in this work to analyses of flow-normalized load.

One major assumption of the flow-normalization method is the stationarity of streamflow time series during the study period, as more fully discussed elsewhere, e.g., Hirsch et al. (2010), Sprague et al. (2011). In this regard, one should be aware that flow-normalized estimates can potentially be misleading if stationarity is violated — that is, if the probability distribution of streamflow on a given day of the year has changed significantly over time, and if such change has been able to exert substantial impacts on the relation between flow and water quality (Hirsch et al., 2010). In the mid-Atlantic region where Chesapeake Bay is located, for example, one might have concerns that watershed development has altered the "flashiness" of streamflow (Jarnagin, 2007), and that these changes, if they exist, could challenge the validity of the stationarity assumption. At present, however, we have no means to further explore this issue. According to Hirsch et al. (2010), there is currently no formal procedure to defend or reject the appropriateness of the stationarity assumption in streamflow, and this is an area where future research is needed to improve WRTDS.

Another issue to consider for any given application of WRTDS is the selection of "half-window widths" for the estimation process, as described in more detail in Appendix A. In this study, the half-window widths were defined as 10 years and 0.5 years for time and season, respectively. For the discharge dimension, the window was selected such that, for a given discharge Q (as reference), non-zero weights would be assigned only to discharges falling between $Q_{min} = Q/\exp(2)$ and $Q_{max} = Q\exp(2)$, or in other words, $\ln(Q_{max}/Q) = \ln(Q/Q_{min}) = 2$. In their analyses of Chesapeake Bay tributaries including Susquehanna, Hirsch et al. (2010) used the above half-window widths and considered them as appropriate based on testing. We agreed with this assessment — preliminary independent analysis with our own data set also suggested that this subjective choice would have little impact on model estimates and load trends, as long as reasonable values are assumed, within ranges suggested by Hirsch et al. (2010).

2.3. Data compilation and analyses

We collected streamflow and water-quality data at Conowingo (1978–2011) from the USGS National Water Information System Web Interface (USGS–01578310; US Geological Survey, 2012b), and at Marietta (1986–2011) and Conestoga (1984–2011) from the SRBC SNAP website (Susquehanna River Basin Commission, 2012). The collected water-quality data included information for eight nutrient and sediment constituents, namely, SS, TP, DP, TN, DN, dissolved orthophosphate (DOP), dissolved nitrate plus nitrite (DNO_x), and dissolved ammonia plus organic N (DKN). We implemented the WRTDS method using the statistical package R (R Development Core Team, 2011) to produce the true-condition and the flow-normalized estimates for every day in the period of record for each species. The daily estimates of load in units of kg day^{-1} were used to calculate the seasonal averages of load for each of the four seasons, defined as January–March, April–June, July–September, and October–December, respectively. In addition, since there was no measurement of PP or PN, their seasonal loads were inferred by subtracting DP and DN from TP and TN seasonal loads, respectively. Thus, the signals of particulate and dissolved fractions could be separated. Similarly, dissolved hydrolysable P (DHP), or the "non-labile" fraction of DP (refer to Table 1 in Neal et al. (2010) for terminology), was inferred by subtracting DOP from DP. In contrast to DP, individually measured data were directly available for DN and DNO_x to the present date and for DKN up to May 1995, after which DKN concentration in water samples has been reported as the difference between measured DN and DNO_x concentrations. For each of the four seasons studied, we observed that the DN loads estimated using WRTDS on measured DN data fell between 95% and 105% of the values (for the same season and location) that were calculated from the sum of estimated DNO_x load plus estimated DKN load.

To reconstruct the 67-year history of NO_x concentration and load at the Conowingo Station (1945–2011), our first step was to close the data gaps in streamflow discharge (1945–1968) and NO_x concentration (1945–1978) based on upstream data at Harrisburg (USGS–01570500; US Geological Survey, 2012b; see Fig. 1 for location). We first compiled the daily streamflow data at Harrisburg from 1945 to 1968 and converted them to Conowingo flow data using the ratio reported by Hagy et al. (2004) (i.e., Conowingo flow $= 10/9 \times$ Harrisburg flow). We then compiled the NO_x concentration data at Harrisburg from 1945 to 1978 and converted them to NO_x concentration at Conowingo using monthly ratios reported by Hagy et al. (2004). These manipulated records, together with observational data (1968–2011 for flow; 1978 to 2011 for NO_x concentration), constituted the 67-year full records at Conowingo (Fig. 3a, b, c, d). On that basis, we estimated the true-condition and the flow-normalized estimates for NO_x at Conowingo from 1945 to 2011.

In addition, to reconstruct the 67-year history of true-condition Jan-May TN load at Conowingo (1945–2011), we needed to convert the pre-1978 true-condition NO_x load to TN load, due to lack of TN concentration data for that period. We first developed linear regression models relating monthly TN to NO_x loads at Conowingo for each month of the year based on available TN and NO_x load estimates from 1981 to 2010 (refer to Appendix B for more detail). We then used these linear models to convert the pre-1978 true-condition NO_x load (described above) to true-condition TN load in respective months. These monthly TN loads were then combined with directly estimated (post-1978) monthly TN loads to reconstruct the 67-year monthly TN loads from 1945 to 2011. Finally, the Jan–May true-condition TN loads at Conowingo were obtained by averaging the monthly TN loads from January to May in each year.

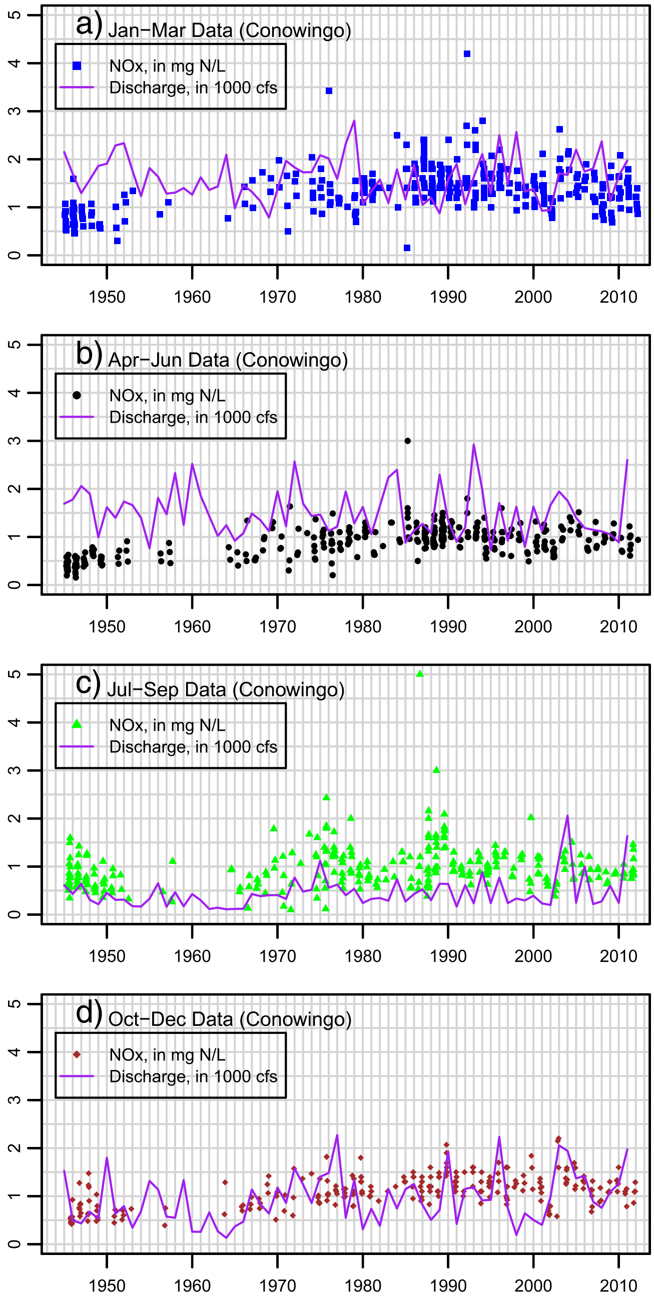

Fig. 3. Observed data of NO_x concentration and seasonal streamflow discharge in (a) Jan–Mar, (b) Apr–Jun, (c) Jul–Sep, and (d) Oct–Dec, in the Susquehanna River at the Conowingo Station for the period from 1945 to 2011. Note that pre-1968 streamflow and pre-1978 NO_x concentration data have been estimated based on observed data at Harrisburg – see text.

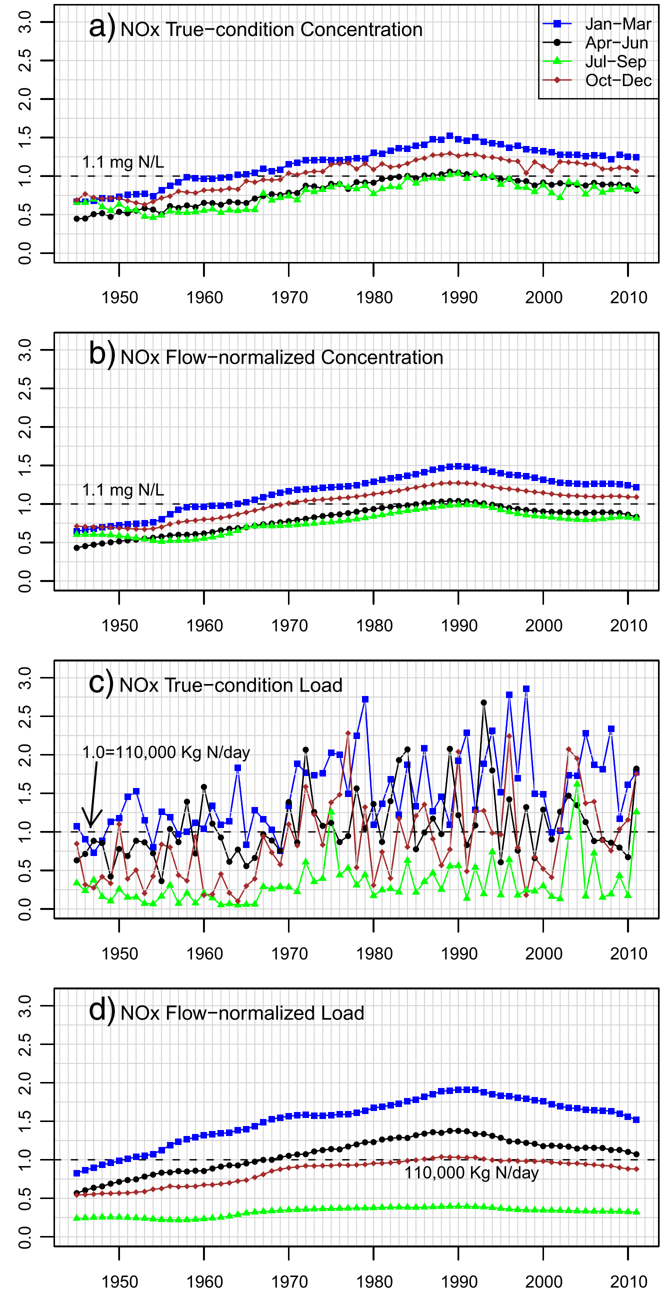

Fig. 4. Seasonal averages of NO_x (a) true-condition concentration, (b) flow-normalized concentration, (c) true-condition load, and (d) flow-normalized load in the Susquehanna River at the Conowingo Station. All estimates have been normalized by the median of respective long-term annual averages at the Conowingo Station (located at y = 1.0 in each panel).

3. Results and discussion

In the sections below, we present our results with regard to the three major sets of tasks and objectives identified in Section 1. First, we present the 67-year analysis of NO_x and TN trends at Conowingo in terms of both "true-condition" and "flow-normalized" results. Second, we analyze the combined loads of SS, P, and N from the Marietta and Conestoga Stations to evaluate progress of management actions in the non-tidal SRB above the reservoirs. This watershed covers portions of New York, Pennsylvania, and Maryland. Finally, we present and discuss the seasonal trends of SS, P, and N load at the Conowingo Station to examine the evolving behavior of the reservoirs in modulating sediment and nutrient load at seasonal resolution.

3.1. History of NO_x and TN load at the Conowingo Station (1945–2011)

3.1.1. Results

Our retrospective analyses of the 67-year record of Susquehanna River NO_x concentration and load are presented in Fig. 4. With regard to the concentration results, the true-condition (Fig. 4a) and the flow-normalized (Fig. 4b) estimates both show similar annual- and decadal-scale trends among all four seasons, with a steady rise from 1945 to around 1990, followed by a steady decline. With regard to the load results, trends in the true-condition loads (Fig. 4c) are difficult to discern, owing to the high degree of inter-annual variability in streamflow. Removal of this influence is in fact a primary motivation for the consideration of the flow-normalized loads (Fig. 4d), which show more clear trends (see Section 3.1.2 below). In general, trends in the flow-normalized loads are similar to trends in the flow-normalized concentrations.

The lack of raw data at Conowingo for TN concentration prevents a similar calculation of flow-normalized concentration or load. For this constituent, our interest is primarily in the "true-condition" load estimates during the period of Jan–May, for the purpose of comparison with values used in earlier analyses by Murphy et al. (2011). These results are shown in Fig. 5, together with the values from Murphy et al. (2011) and also differences between the two sets of loads. In general, results using either method exhibit a similar long-term trend, with generally much lower peak loads prior to 1970, increased variability since about 1980, and a general trend of stabilized or decreasing loads since that time.

3.1.2. Discussion

The flow-normalized NO_x results presented in Fig. 4 (b and d) show similar trends in all four seasons. More generally, however, such consistency in seasonal trends should not necessarily be expected. In fact, our on-going study of trends in other tributaries of Chesapeake Bay has revealed substantially different trends in some cases. For example, from the late 1970s to the early 1990s, the flow-normalized NO_x concentrations in the Potomac River show a trend of slight decline in Jan–Mar, but strong upward trends in the other three seasons (data not shown).

Perhaps the most important point to observe from Fig. 4 is the manner in which the flow-normalized NO_x loads (Fig. 4d) remove the effects of the highly variable streamflow during each season (shown as solid lines in Fig. 3). In this regard, Fig. 4d reveals a smooth trend in load change that is similar in its basic aspects to the trends in estimated concentration (Fig. 4a and b) and devoid of the flow-induced variations evident in Fig. 4c. From this example, we can see that the flow-normalized loads are more helpful to evaluate progress of management actions in the watershed. Differences between trends in flow-normalized loading and in flow-normalized concentration are presumably the result of flow influences on concentration and resulting effects on the regressions that account for flow. Although the exact meanings of these differences are complex, it has been suggested that trends in flow-normalized concentration are more representative of changes in point sources, which are presumed to be less heavily influenced by flow (Hirsch et al., 2010).

As noted in Section 1, the 67-year true-condition TN load history at Conowingo was reconstructed using the WRTDS method in order to verify whether Jan–May trends reported by Murphy et al. (2011) would still be observed. Our results (Fig. 5) confirm that true-condition estimates of TN loading with WRTDS are generally similar to those previously assumed by Murphy et al. (2011). Overall, the WRTDS estimates range between 0.84 and 1.09 times of the previously reported TN load values, with largest differences occurring in the 1970s, which were relatively

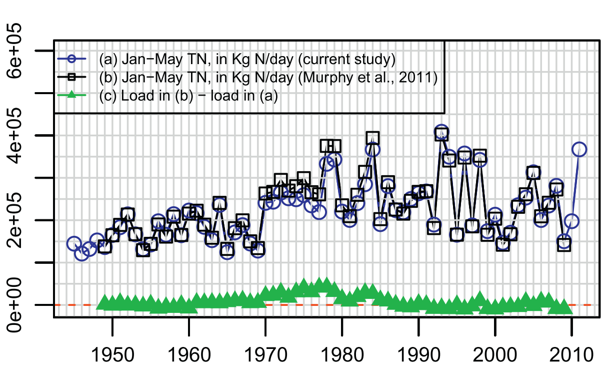

Fig. 5. Estimates of "true-condition" Jan–May TN load in the Susquehanna River at the Conowingo Station for the period 1945 to 2011. Plot (a) shows TN loads obtained in this study. Plot (b) shows TN loads reported by Murphy et al. (2011) for the period 1949 to 2009. For that study, TN loads prior to 1980 were obtained using regression equations between TN and NO_x load developed by Hagy et al. (2004), and TN loads from 1981 to 2009 were directly obtained from the USGS RIM Program website (US Geological Survey, 2012a). Plot (c) shows differences in estimated loads between (a) and (b).

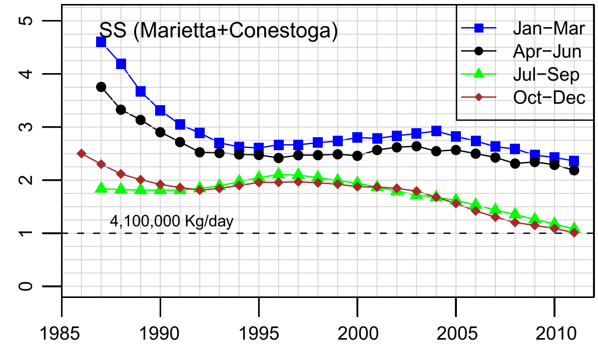

Fig. 6. Seasonal averages of flow-normalized SS load from the Marietta and Conestoga Stations. All loads have been normalized by the median of long-term annual SS loads at the Conowingo Station (located at y = 1.0).

wet years. These inconsistencies likely resulted from the different treatment of high-flow samples in ESTIMATOR and WRTDS. For a given estimation day, the ESTIMATOR model always gives equal weight to high-flow samples and low-flow samples, whereas the WRTDS method assigns higher weight to samples whose corresponding flows are closer to the flow on the estimation day.

3.2. History of SS, P, and N Load from the Marietta and Conestoga Stations (1986–2011)

3.2.1. Results

The combined flow-normalized SS loads from Marietta and Conestoga show consistently downward trends in all four seasons (Fig. 6), with decelerated reduction or slight rise between the mid-1990s and the mid-2000s.

The combined flow-normalized TP loads from Marietta and Conestoga also show downward trends in all four seasons, with decelerated reduction from the mid-1990s to the early 2000s (Fig. 7a). Within TP, PP contributed to the TP reduction only since around 2000 (Fig. 7b), whereas DP contributed to the TP reduction throughout the study period (Fig. 7c). Within DP, DOP increased consistently until 2002 and started to contribute to the DP reduction thereafter (Fig. 7d), whereas DHP decreased substantially in the earlier period (up until 2002) and has remained low since that time (Fig. 7e).

The combined flow-normalized TN loads from Marietta and Conestoga also show consistently downward trends in all four seasons (Fig. 8a). Within TN, PN decreased rapidly until the late 1990s, and thereafter increased slightly in Jan–Mar and Apr–Jun but continued to reduce in Jul–Sep and Oct–Dec (Fig. 8b). DN shows downward trends in all four seasons (Fig. 8c), similar to the TN trends previously noted. Both DN and PN contributed to the TN reduction until the late 1990s, thereafter primarily DN contributed to the TN reduction. The two fractions of DN, i.e., DNO_x (Fig. 8d) and DKN (Fig. 8e), were estimated separately using available data and both show downward trends throughout the study period.

3.2.2. Discussion

The flow-normalized SS, P (TP, PP, and DP), and N (TN, PN, and DN) loads from Marietta and Conestoga all show downward trends from 1986 to 2011, suggesting that management controls have been effective in reducing watershed inputs of these pollutants in the non-tidal SRB above the reservoirs.

To more fully understand the above-noted "positive progress," it is useful to review factors affecting the source, transport, and transformation of nutrient and sediment in the SRB. In terms of source, Ator et al. (2011) reported that mean annual TN loads from the non-tidal SRB, calculated for the year 2002 by using statistical representations of long-term mean hydrological conditions, have originated mainly from non-point source inputs of fertilizer, fixation, and manure (58%), followed by

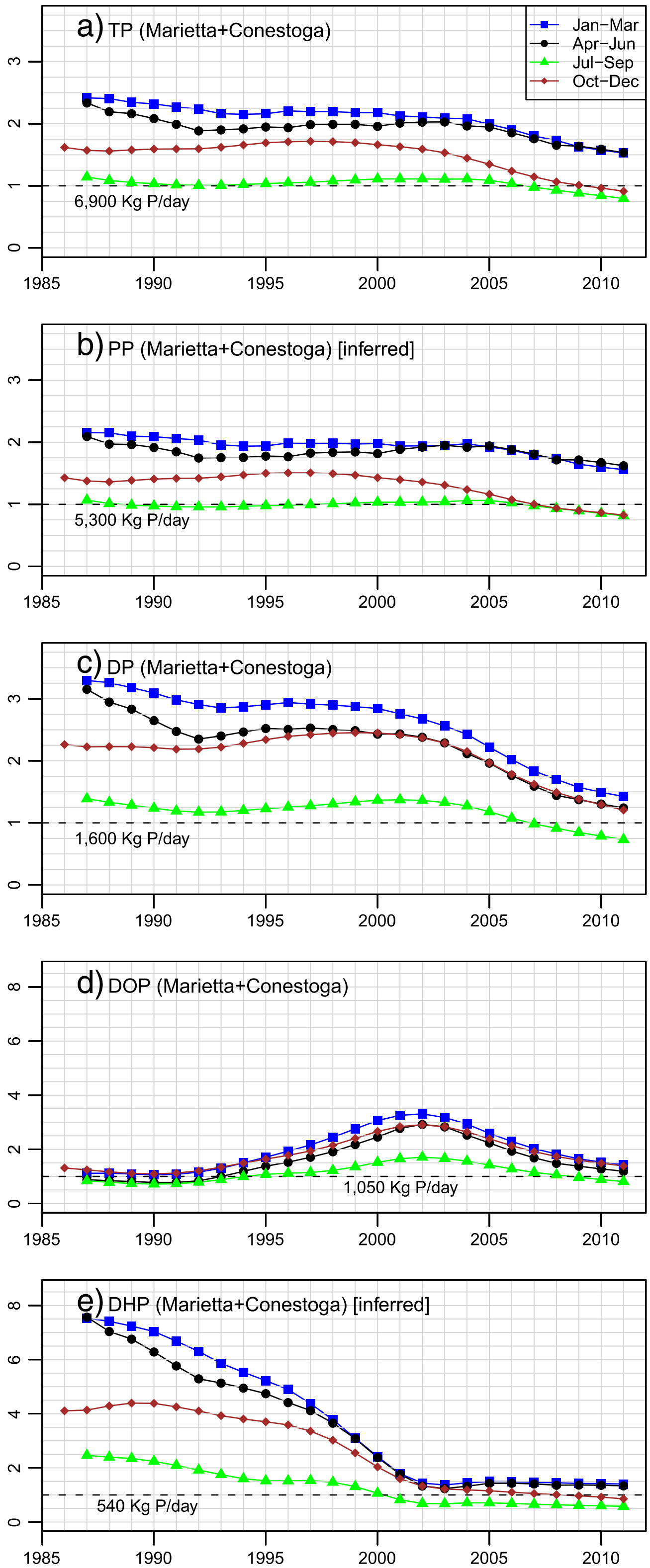

Fig. 7. Seasonal averages of flow-normalized load of (a) TP, (b) PP, (c) DP, (d) DOP, and (e) DHP from the Marietta and Conestoga Stations. All loads have been normalized by the median of respective long-term annual loads at the Conowingo Station (located at y = 1.0 in each panel). Estimates for panels labeled as "inferred" were obtained from data that were inferred rather than measured (i.e., PP = TP − DP; DHP = DP − DOP).

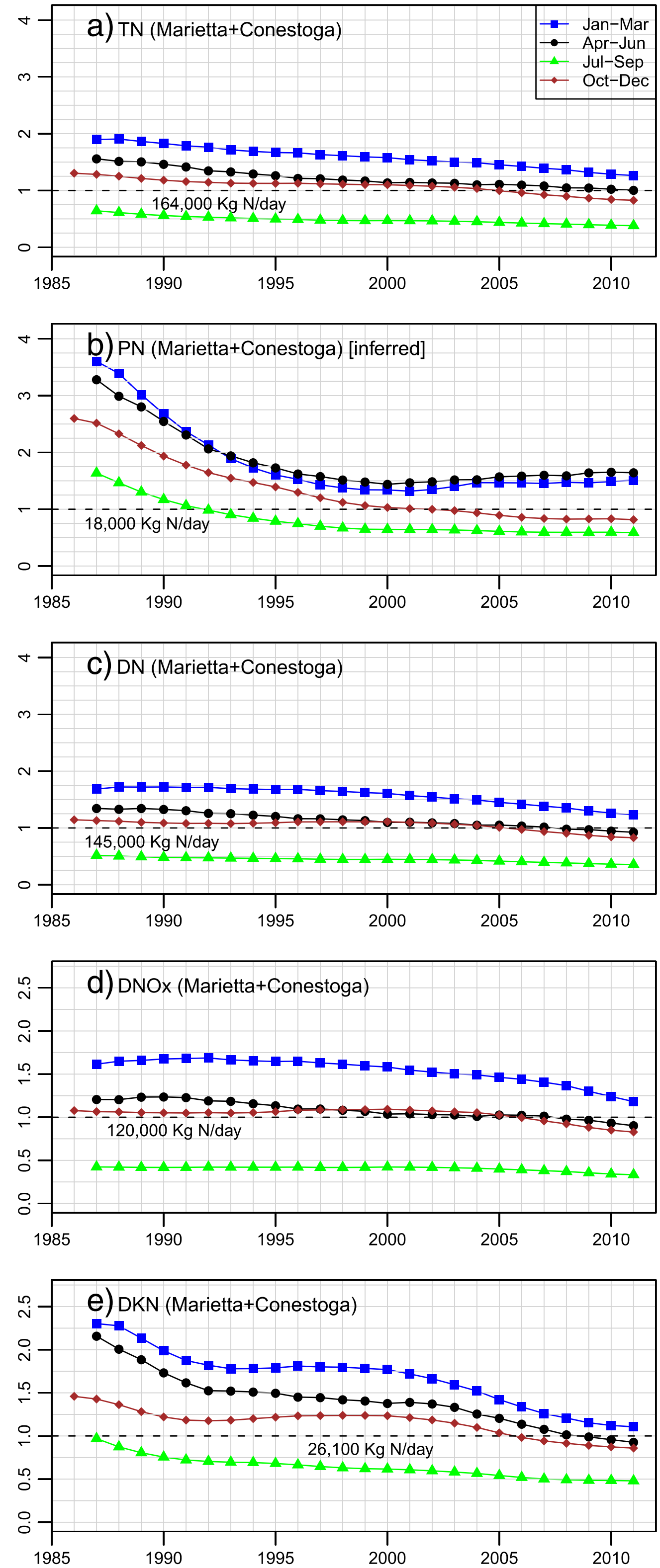

Fig. 8. Seasonal averages of flow-normalized load of (a) TN, (b) PN, (c) DN, (d) DNO_x, and (e) DKN from the Marietta and Conestoga Stations. All loads have been normalized by the median of respective long-term annual loads at the Conowingo Station (located at y = 1.0 in each panel). Estimates for panel labeled as "inferred" were obtained from data that were inferred rather than measured (i.e., PN = TN–DN).

atmospheric deposition (20%), urban sources (12%), and point sources (10%). Using the same method, TP loads were reported to have originated mainly from point sources (39%) as well as from fertilizer and manure (32%), followed by erosion of rocks (22%) and urban sources (7%). In addition, Brakebill et al. (2010) determined agricultural land as the greatest overall source and urban development as the source with highest yield (load per unit area) for SS in the CBW. Once the N and P are generated from these sources, they can be temporarily stored in the system (e.g., land surface, riparian buffer, river channels, and reservoirs), transformed chemically or biologically (e.g., plant uptake, mineralization and denitrification of N, and precipitation of P), or transported downstream (Ator et al., 2011; Brakebill et al., 2010). SS, however, exhibits a more conservative behavior since it cannot be readily transformed (Brakebill et al., 2010).

In the last few decades, various changes in watershed practices have been implemented to control N, P, and SS load in the SRB, some focusing on reduction by controlling pollutant transport or transformation, but more focusing on control at the pollutant source. An overview of some historical changes in source-based management and practices in the SRB is provided in Appendix C. As further discussed in Appendix C, it is likely that the source-based management strategies and associated controls on transport and transformation processes, were responsible for the downward nutrient and sediment trends in the SRB at locations above the reservoirs. Indeed, Brakebill et al. (2010) have suggested that effective SS control measures should include both source reduction (e.g. settlement ponds, soil conservation practices, and riparian buffers) and streambank protection (e.g. directing erosive flow and flood-plain stabilization). However, identification of the extent of implication and relative contribution of these different management actions is well beyond the scope of our current study.

3.3. History of SS, P, and N load at the Conowingo Station (1978–2011)

3.3.1. Results

The flow-normalized SS loads at Conowingo show generally “fall-and-then-rise” trends in all four seasons (Fig. 9). In Jan–Mar, Apr–Jun, and Jul–Sep, SS load generally decreased until around 1990, stabilized for about one decade, and increased rapidly since the late 1990s. In Oct–Dec, SS load displayed much weaker variation. Overall, the SS load at Conowingo has then digressing increasingly far from the TMDL goal in recent years in all seasons except Oct–Dec.

The flow-normalized TP loads at Conowingo show very similar “fall-and-then-rise” trends in all four seasons (Fig. 10a), closely following the SS trend. Overall, the TP load at Conowingo has also digressed increasingly far from the TMDL goal. The effect is clearly related to particulate species — PP shows the same “fall-and-then-rise” trend (Fig. 10b), whereas DP shows downward trends in all four seasons (Fig. 10c). Both DP and PP contributed to the TP reduction until the mid-1990s, and PP alone contributed to the TP rise thereafter. Within DP, DOP shows downward trends particularly in the early period (Fig. 10d), whereas DHP shows bi-modal patterns in all four seasons (Fig. 10e).

The flow-normalized TN loads at Conowingo also show long-term trends that are similar among all four seasons, but opposite to those of SS and TP (i.e., “rise-and-then-fall”), with the peak load occurring in the late 1980s (Fig. 11a). Overall, the TN load at Conowingo has been brought

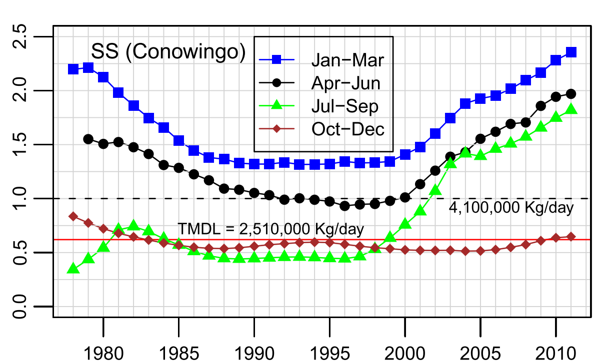

Fig. 9. Seasonal averages of flow-normalized SS load in the Susquehanna River at the Conowingo Station. All loads have been normalized by the median of long-term annual SS loads at Conowingo (located at y = 1.0). The TMDL of 2,510,000 kg day^{-1} set for the Susquehanna River (US Environmental Protection Agency, 2010) is inserted for comparison.

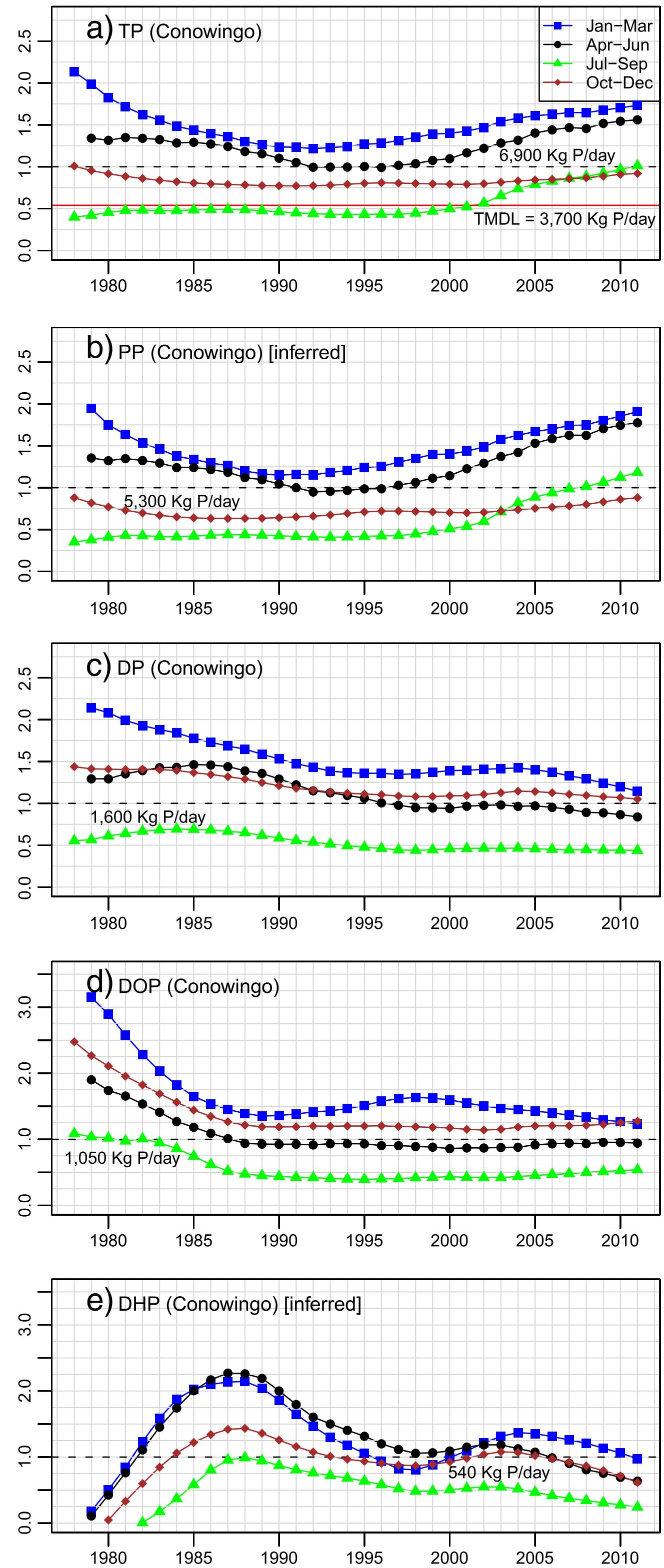

Fig. 10. Seasonal averages of flow-normalized load of (a) TP, (b) PP, (c) DP, (d) DOP, and (e) DHP in the Susquehanna River at the Conowingo Station. All loads have been normalized by the median of respective long-term annual loads at Conowingo (located at y = 1.0 in each panel). The TMDL of 6,900 kg P day^{-1} set for the Susquehanna River (US Environmental Protection Agency, 2010) is inserted in (a) for comparison. Estimates for panels labeled as “inferred” were obtained from data that were inferred rather than measured (i.e., PP = TP − DP; DHP = DP − DOP).

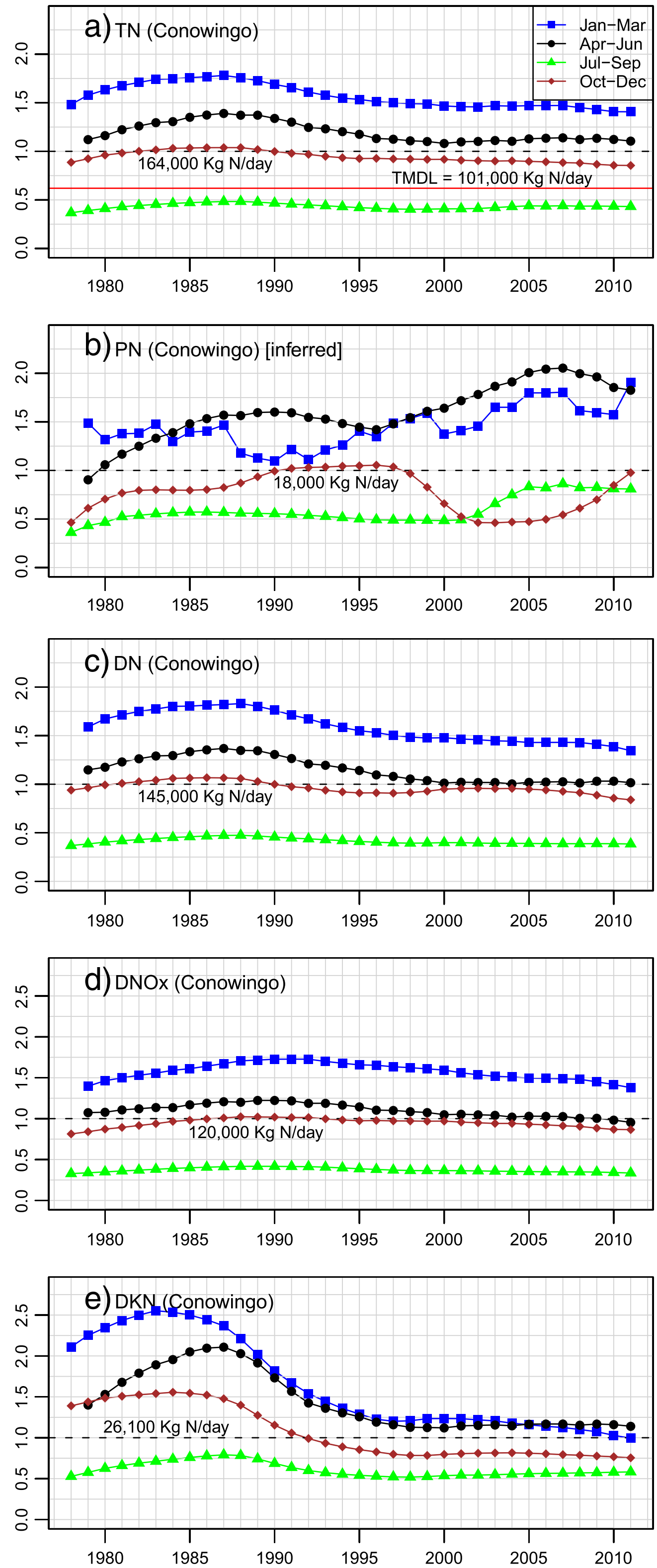

Fig. 11. Seasonal averages of flow-normalized load of (a) TN, (b) PN, (c) DN, (d) DNO_x, and (e) DKN in the Susquehanna River at the Conowingo Station. All loads have been normalized by the median of respective long-term annual loads at Conowingo (located at y = 1.0 in each panel). The TMDL of 101,000 kg N day^{-1} set for the Susquehanna River (US Environmental Protection Agency, 2010) is inserted in (a) for comparison. Estimates for panel labeled as "inferred" were obtained from data that were inferred rather than measured (i.e., PN = TN − DN).

closer and closer to the TMDL goal in recent years. Within TN, PN shows upward trends in Jan–Mar, Apr–Jun, and Jul–Sep (Fig. 11b), whereas DN shows similar trends as those of TN (Fig. 11c). Both DN and PN contributed to the TN rise until the late 1980s, and DN alone contributed to the TN reduction thereafter. Within DN, DNO_x shows "rise-and-then-fall" trends (Fig. 11d), and DKN shows similar trends but with the start of the "fall" occurring 3–7 years earlier than DNO_x (Fig. 11e).

3.3.2. Discussion

3.3.2.1. Dissolved and particulate nutrient fractions at the Conowingo Station. As with the previously discussed "above-reservoir" results (Section 3.2), the below-reservoir results also show an overall trend of reducing load for both DN and DP. On the other hand, however, there is a clear upward trend of SS load at the Conowingo Station since the late 1990s and accompanying increases in PP and PN loads. In terms of TN, there is still a trend of overall decline in all seasons because PN is a small portion of TN and so PN has not reversed the progress achieved through DN reduction. For phosphorus, however, PP is the major fraction of TP, and the recent rise of PP has caused the TP rise since the mid-1990s.

In terms of impact on Chesapeake Bay, the observed increases in particulate fractions of P and N are of concern. Although these particulate species are not as immediately available for algal consumption as are the DN (e.g. DNO_x) and DP (e.g. DOP) species, a portion of the particulate species can undergo decomposition and generate bioavailable N and P to sustain algae growth (Kemp and Boynton, 1984). Such generation of bioavailable nutrients from particulate phases can be strongly promoted at conditions of high temperature (Kemp and Boynton, 1984) and low dissolved oxygen concentration (Boynton et al., 1996), which are coupled characteristics of Chesapeake Bay in summer.

3.3.2.2. The reservoirs' role in sediment and particulate nutrient retention. Considering that the watershed monitored by Conowingo has almost identical streamflow, watershed area, and land use pattern to that monitored by the Marietta and Conestoga Stations (Table 1), the deteriorating situation of SS load at Conowingo can be largely attributed to the impact of the reservoirs, as evidenced by a comparison between these loads and those observed upstream (Fig. 6). In fact, the reservoir system appears to have been gradually losing its sediment storage capacity (SSC) especially since the mid-1990s, with concurrent effects on PP and PN. Correspondingly, the upward trends of PP and PN load at Conowingo suggest negative progress in particulate nutrient control for the overall non-tidal SRB, which can be largely attributed to the impact of the reservoirs, as evidenced by comparisons between these loads and those observed upstream (Figs. 7b and 8b). Coupled with the gradually diminishing SSC, the reservoir system seems to be trapping less PP and PN in recent years than in the early years. The seasonal effluent trends at Conowingo are consistent with those observed by Hirsch (2012a) (see his Figs. 13 and 17) using annual load estimates, and our new analysis of upstream data now further support his suggestion that recent changes reflect alterations in reservoir performance.

3.3.2.3. Trends in rate of change in sediment inventory (storage) within the reservoirs (1986–2011). To further explore the evolving performance of the reservoirs in modulating N, P, and SS load, we considered the reservoir system as the control volume (CV), the combined load from Marietta and Conestoga as the input, and the load at Conowingo as the output. For simplicity, we ignored watershed processes within the CV (i.e., a small watershed area below Marietta (site No. 7 in Fig. 1) and Conestoga (site No. 8 in Fig. 1) and above Conowingo (site No. 9 in Fig. 1), which corresponds to roughly 2.4% of the total watershed area above Conowingo). We then used the difference between our flow-normalized estimates of the SS input and output rates to roughly represent the rate of change in SS inventory within the reservoirs, thus reflecting rates of storage or

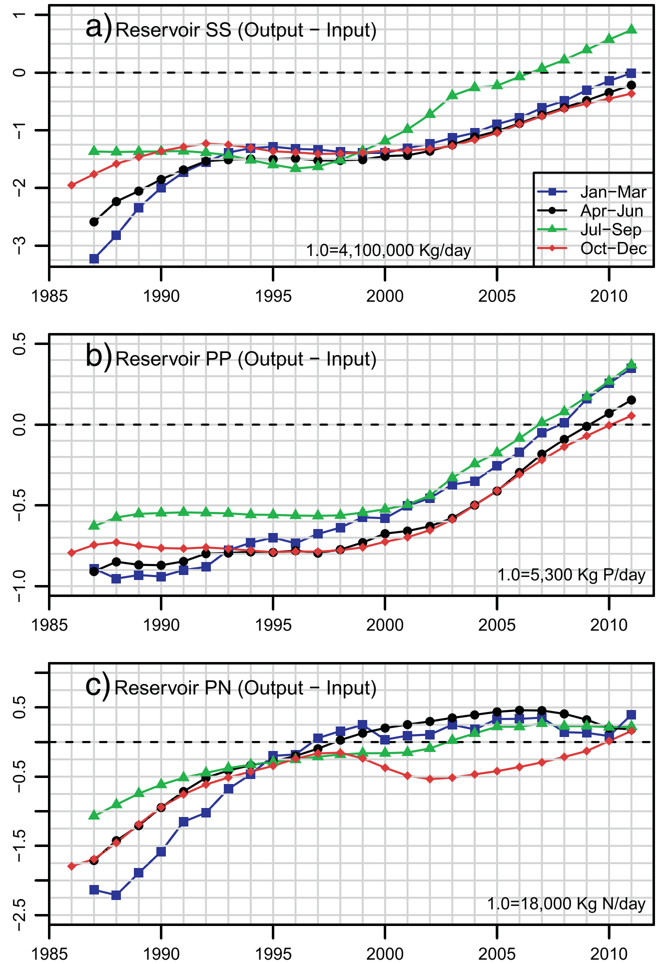

Fig. 12. Rates of storage change in (a) SS, (b) PP, and (c) PN within the reservoir system based on flow-normalized load. All rates of change have been calculated as the differences between the loads at Conowingo (system output) and the combined loads from Marietta and Conestoga (system input), and then normalized by the median of respective long-term annual loads at the Conowingo Station.

release. Similarly, we evaluated the rates of inventory change for PP and PN within the reservoirs.

Changes in SS load across the reservoirs show net storage of SS in most years in all four seasons (Fig. 12a), but the reservoirs' capacity to trap new SS input has been gradually diminishing since the beginning of the record, albeit with an apparent plateau in net storage rate occurring in the 1990s. On a net basis, these flow-normalized results suggest that the reservoirs may have started to lose SS in Jul–Sep since 2007 and in Jan–Mar since 2011, and it appears to be on a trajectory to start losing SS in Apr–Jun and Oct–Dec soon. Note that net loss of sediment is presumably related to scouring and that the Jul–Sep values since 2007 are likely the result of the historical hurricanes and storms that occur predominantly in this season. In fact, the flow-normalized estimate of net loss of SS in Jul–Sep for 2011 was sufficient to exceed the estimates of net storage in the other seasons, such that the estimate for that year would represent an overall annual net loss and for the first time in the history of the Conowingo Dam. Similarly, we observed gradually diminishing capacity of the reservoirs for trapping new input of PP (Fig. 12b) and PN (Fig. 12c) in all four seasons in the last 26 years. For PP, the flow-normalized results imply net loss from the reservoirs in Jul–Sep since 2007, Jan–Mar since 2008, and Apr–Jun and Oct–Dec since 2011. For PN, there has been net loss from the reservoirs in Jan–Mar since 1997, Apr–Jun since 1998, Jul–Sep since 2003, and Oct–Dec since 2010. In terms of the corresponding estimates of annual change across the reservoirs, flow-normalized output reach the estimates of input for PN and PP in 2003 and 2009, respectively, but with PP showing a much more rapid rise since the late 1990s. The different trajectories of PP and PN are possibly related to differences in the size fractions with which P and N are predominantly associated in the inlet, the outlet, or both (Hirsch, 2012b). In addition, there may also be substantial differences with regard to the overall (net) transformation of N and P between dissolved and particulate fractions, as might be expected from fundamental dissimilarities in the biogeochemical processes that affect each nutrient.

It should be noted that sediment or particulate nutrient retention in the reservoirs in any given year would be affected by a combination of highly dynamic and complex processes. For example, short-term changes in sediment storage can occur due to scouring in storm events (Hirsch, 2012a; Langland, 2009) or due to short-term changes in reservoir stratification and biochemistry. In this regard, for example, the Jul–Sep results may have been especially influenced by some storm events in recent years, and the annual net SS loss in 2011 does not necessarily mean that this situation will continue in the coming years. Overall, such complications can confound the ability of the method to evaluate the "capacity to trap materials" under normal flow conditions. In addition, it is especially important to recall that flow-normalized trends in input and output loadings do not reflect the best estimate of "true conditions" for any given year. Nevertheless, it is evident from Fig. 12 that there is a clear decadal-long trend of steady decline in the reservoirs' ability to trap sediment and particulate nutrients and there is reason for serious concern that the Conowingo Reservoir may be already at or near its storage capacity.

3.3.2.4. Cumulative SS deposition in the Conowingo Reservoir (1987–2010). In order to assess the Conowingo Reservoir's remaining SSC, one must consider both the available capacity in the reservoir and the on-going rate of sediment deposition. Considering the latter issue first, we note that WRTDS-based "true-condition" calculations of upstream and downstream SS loadings provide new estimates of annual SS deposition in the reservoir that are useful for comparisons against prior estimates made by others using other methods. In previous studies, Langland (2009) reported 1.47×10^7 U.S. tons of SS deposition from 1996 to 2008 (averaging 1.23×10^6 tons/year) using a bathymetry mapping method, and 1.69×10^7 tons of SS deposition for the same period (averaging 1.41×10^6 tons/year) using monitored loading estimates. In comparison, our true-condition estimates suggest an average deposition loading of 1.55×10^6 tons/year for 1996 to 2008, which matches reasonably well with those reported by Langland (2009), being roughly 27% and 10% higher, respectively.

In terms of the remaining SSC in the Conowingo Reservoir, this was reported to be 4.2×10^7 tons in 1996 based on bathymetry mapping (Langland, 2009). Our true-condition estimate of cumulative SS deposition from 1996 to 2010 is 2.0×10^7 tons. Thus, ~47% of the 1996 capacity had already been consumed and there was only 2.2×10^7 tons of remaining SSC as of 2010, which was about 11% of the original 1928 SSC (2.04×10^8 tons) reported by Langland and Hainly (1997). This result clearly indicates that the Conowingo Reservoir is approaching its SSC.

3.3.2.5. Evolving behavior of the reservoirs in sediment and nutrient retention. In addition, we investigated the evolving pattern of seasonal SS concentration in relation to streamflow discharge at Marietta (reservoirs inlet) and Conowingo (reservoirs outlet), as an alternative method to examine the evolving behavior of the reservoirs in SS retention. For each season, we selected the middle day as representative of the season. At Marietta, the SS concentration vs. discharge relation appears to be similar for the selected years between 1990 and 2010 in all four seasons (Fig. 13a, b, c, d). At Conowingo, however, this relation has gradually shifted upward since the beginning of the study period in all four seasons (Fig. 13e, f, g, h). The results suggest that, for a given flow condition, there are higher SS concentrations at Conowingo in recent years than in the early years in all four seasons. In addition, a significant shift in this relation appears to have occurred around 2000 in all four seasons,

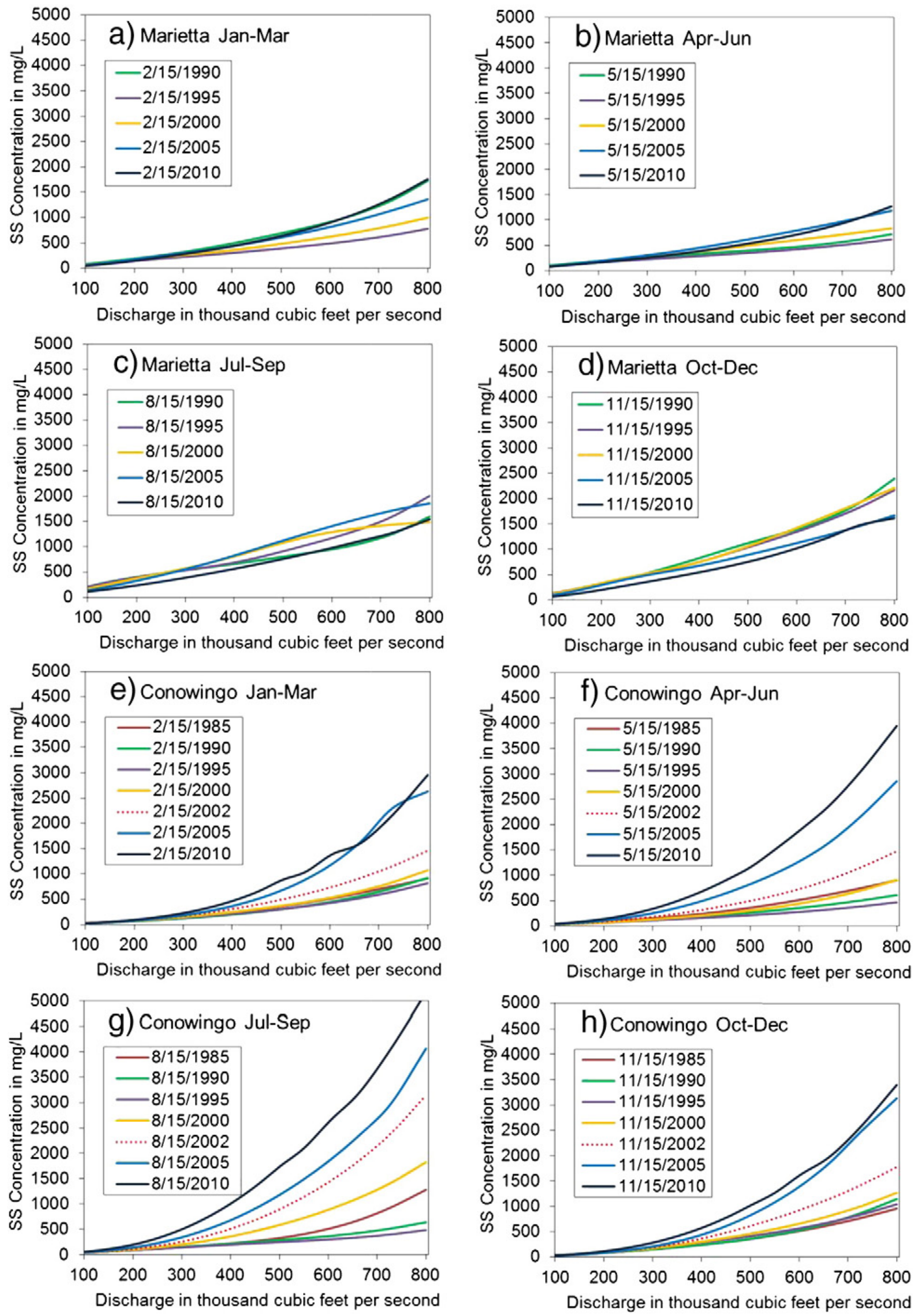

Fig. 13. The evolving patterns of SS concentration vs. streamflow discharge in each season at (a–d) the Marietta and (e–h) the Conowingo Stations. A 5-year interval was selected to show the evolution. Patterns in 2002 (dashed line) were also added in (e) to (h) to aid comparison.

consistent with our previous inference that the reservoir system has been losing its SSC especially since the late 1990s (Fig. 12). Similar patterns were also observed for PN and PP (data not shown). Thus, the concentration results (Fig. 13) tend to confirm our earlier conclusion that the reservoir system is now less efficiently trapping SS, PP, and PN than in the earlier years, and particularly so during high flow conditions. Considering only the date of September 1 as a point of comparison among years, Hirsch (2012a) has also observed rising patterns of TN, TP, and SS concentration as a function of streamflow discharge (Hirsch, 2012a; see his Figs. 8, 12, 16).

3.3.2.6. Summary and broader implications. The above analyses have two important implications. First, the flow-normalized reservoir input and output trends (Fig. 12) suggest that the reservoir system has been steadily losing its storage capacity for SS, PP, and PN over the past two to three decades, and especially so for SS and P since the mid-1990s. Second, both these trends and the concentration vs. discharge plots (Fig. 13) show that the reservoir system is becoming increasingly sensitive to scour events and that the Conowingo Reservoir has neared its storage capacity. Despite earlier predictions that the reservoir may not reach its total SSC until 2024–2029 (Langland, 2009), it is evident that increasingly substantial amounts of SS, PP, and PN are already entering Chesapeake Bay as the result of major reductions in reservoir performance toward sediment retention. Moreover, one might expect such increases to be further intensified if there are more frequent and intense major storms as the result of changing climate (Najjar et al., 2010; Rabalais et al., 2009).

On a seasonal basis, these findings complement the annual analyses recently provided by Hirsch (2012a). The current study adds additional information about flow-normalized seasonal trends of multiple nutrient and sediment species, with a special focus on directly comparing above- and below-reservoir loading estimates as a means of considering long-term trends in reservoir performance.

Although recent rises in loadings of particulate-based nutrients have been at least in part counter-acted by reductions in the more readily available dissolved species, the changes in reservoir performance will pose significant new kinds of challenges to attainment of TMDL goals for the SRB. In this regard, our results reinforce recommendations recently made by Hirsch (2012a) — i.e., that these changes need to be factored into the proper establishment of regulatory load requirements and the development of watershed implementation plans. As better described elsewhere (Susquehanna River Basin Commission Sediment Task Force, 2002), a wide range of riverine, upland, and reservoir management options will need to be considered for controlling the sediment load in the non-tidal SRB.

4. Conclusions

This paper presents our analyses of long-term seasonal trends of flow-normalized N, P, and SS loads from the non-tidal Susquehanna River to Chesapeake Bay. Major findings include:

- Long-term trends of flow-normalized N, P, and SS load generally followed similar patterns in all four seasons, implying that changes in watershed function and land use had similar impacts on nutrient and sediment load at all times of the year.
- 67-year concentration and load histories of NO_x at the fall-line of the Susquehanna River show a steady rise from 1945 to 1990 and a steady decline thereafter.
- Flow-normalized loads of N, P, and SS have been generally reduced in the SRB above the Lower Susquehanna River Reservoir System (representative of about 96% of the non-tidal SRB) in the last 26 years, which can most likely be attributed to a suite of management control actions on point, agricultural, and stormwater sources.
- Flow-normalized loads of SS, PP, and PN at the outlet of the Conowingo Reservoir have been generally rising since the mid-1990s. The reservoirs' capacity to trap these materials has been diminishing, and the Conowingo Reservoir has neared its sediment storage capacity. These important changes will pose significant new kinds of challenges to attainment of TMDL goals for the SRB.

Conflict of interest

The authors declare no conflict of interest.

Acknowledgement

This research was funded by the U.S. Water Environment Research Federation under Grant No. U4R09 and the National Science Foundation under Award No. 0854329. The authors benefited greatly from suggestions and comments from anonymous reviewers and Robert Hirsch (USGS), and would also like to thank Rebecca Murphy for useful discussions. This work would not have been possible without the streamflow and concentration data made available through the USGS National Water Quality Assessment Program and the SRBC Susquehanna Nutrient Assessment Program. The authors also want to thank Mike Langland (USGS) and Lori Sprague (USGS) for permission to adapt Table 1 and Figs. 1 and 2 from their reports.

Appendix A. Supplementary data

Supplementary data to this article (Appendices A, B, and C) can be found online at http://dx.doi.org/10.1016/j.scitotenv.2013.02.012.

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