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Tracking changes in nutrient delivery to western Lake Erie: Approaches to compensate for variability and trends in streamflow

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ABSTRACT

Tracking changes in stream nutrient inputs to Lake Erie over multidecadal time scales depends on the use of statistical methods that can remove the influence of year-to-year variability of streamflow but also explicitly consider the influence of long-term trends in streamflow. The methods introduced in this paper include an extended version of Weighted Regressions on Time, Discharge, and Season (WRTDS) modeling that explicitly considers nonstationary streamflow by incorporating information on changes in the frequency distribution of daily measured streamflow (discharge) over time. Soluble reactive phosphorus (SRP) trends in annual flow-normalized fluxes (loads) at five long-term monitoring sites in the western Lake Erie drainage basin show increases of 109 to 322% over the period 1995 to 2015. About one-third of the increase appears attributable to increasing discharge trends, while the remaining two-thirds appears to be driven by changes in concentration versus discharge relationships reflecting higher concentrations for any given discharge during recent years. Trends in total phosphorus and three nitrogen parameters (total nitrogen, nitrate-nitrite, and total Kjeldahl nitrogen) at the 10 sites analyzed were much less pronounced, and commonly show decreases in concentration-discharge relationships accompanied by increases in discharge, resulting in little net change in total flux. Trends in monthly SRP fluxes and discharge, dissolved versus particulate fractions of nutrients, and N:P flux ratios were also evaluated. The methods described here provide tools to more clearly discern the effectiveness of nutrient-control strategies and can serve as ongoing measures of progress, or lack of progress, towards nutrient-reduction goals.

Introduction

Concerns regarding nutrient loading linked to rapid increases in cyanobacteria harmful algal blooms (cHABs) in Lake Erie have galvanized efforts to increase agricultural conservation activities and expand tributary monitoring to assess effects of nutrient-reduction efforts (Betanzo et al., 2015a; International Joint Commission, 2017; Johnson et al., 2014; Ohio Environmental Protection Agency, 2017; U.S. Environmental Protection Agency, 2014, 2018; Shine, 2015). However, the nutrient trend drivers in the Lake Erie basin and Great Lakes region are complex, and include climatic changes and increases in streamflow (Coopersmith et al., 2014; Culbertson et al., 2016; DeMarchi, 2013; McDermid et al., 2015; U.S. Environmental Protection Agency, 2013), changing agricultural management practices (Jarvie et al., 2017; Michalak et al., 2013; U.S. Department of Agriculture, 2017), and changing precipitation-runoff response due to the combination of land-use practice and drainage modifications (King et al., 2015a; Michalak et al., 2013; Rittenburg et al., 2015).

Combined effects of climatic and land use/land management changes present challenges in tracking and assessing causes of changes in hydrologic system responses, such as changes in amounts of water and nutrients delivered to streams. Under such conditions, analysis of nutrient loading changes must incorporate tools to identify potential underlying trends in order to assess the effects of land management practices and nutrient-reduction efforts. In addition, it is crucial that such evaluations are not overly influenced by short-term (year-to-year) variations in nutrient fluxes (loads) related to the effects of random short-term changes in weather and streamflow. These types of streamflow variations, if not factored into trend evaluation, can result in misleading conclusions about improvement (or degradation) of water quality that may simply be artifacts of a particular sequence of wet and dry periods that have occurred over the period of record.

Stream nutrient loading is considered to be the primary driver of cHABs in Lake Erie, which have increased dramatically in recent years, associated with what has been termed the re-eutrophication of Lake Erie (International Joint Commission, 2014; Michalak et al., 2013;

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Scavia et al., 2014). Of particular concern are the recent dissolved phosphorus (P) increases in tributaries of the western Lake Erie drainage basin (WLEB, delineated by the area corresponding to U.S. Geological Survey 4-digit hydrologic unit code (HUC) 0410, Western Lake Erie subregion, at: https://water.usgs.gov/GIS/huc_name.html) (Baker et al., 2014a, b; Michalak et al., 2013; Reutter et al., 2011; Richards et al., 2010). The roles and interactions among all of the nutrient species contributing to algal community structure and blooms, and the catalysts for algal toxin release are complex (Chaffin et al., 2013, 2014; Gobler et al., 2016; Paerl et al., 2016; Pennuto et al., 2014; Reutter et al., 2011); nitrogen (N) supply and speciation, in addition to P, may be important in particular for *Planktothrix* bloom growth (and associated microcystin concentrations), which often dominates blooms in the lower tributaries and the bays of western Lake Erie, including Sandusky Bay (Chaffin et al., 2014; Davis et al., 2015).

WLEB streams have exhibited some strong and relatively rapid changes in water quality during the past 40 years. The historical perspective of factors associated with nutrient trends in tributaries of the western Lake Erie basin and their relations with increasing algal blooms in the lake have been summarized by Baker et al. (2014a, 2017), International Joint Commission (2014), and U.S. Department of Agriculture (2017). Land management changes reported to have occurred in the WLEB in recent decades include an increase in conservation tillage; changes in the chemical formulations, methods, and timing of fertilizer applications (shifts to surface broadcasting and to fall-winter applications); and expansion of subsurface drainage (Jarvie et al., 2017; Ohio Lake Erie Phosphorus Task Force, 2010, 2013; Smith et al., 2015a). Widespread conservation tillage has reduced particulate nutrient delivery to streams but has been related to a buildup of phosphorus in shallow soils (King et al., 2017; Powers et al., 2016; U.S. Department of Agriculture, 2017) and is considered a cause of increased delivery of dissolved nutrients including SRP to streams (Daloglu et al., 2012; Joosse and Baker, 2011; Kalcic et al., 2016; Sharpley et al., 2012). Tile drainage systems, extensively used in WLEB croplands, recently have been recognized as important transport pathways for both dissolved and particulate P in the WLEB (King et al., 2015b; Smith et al., 2015b). There is evidence that tile-drainage extent has increased across the WLEB during recent decades (Jarvie et al., 2017; Ohio Lake Erie Phosphorus Task Force, 2013; U.S. Department of Agriculture, 2017).

Climatic changes in this region have been related to increases in streamflow, precipitation, temperature, the frequency of intense rainfall events, and reduced snowfall and frozen ground conditions (Coopersmith et al., 2014; DeMarchi, 2013; DeMarchi et al., 2011; Karl and Knight, 1998; Mortsch et al., 2000). Changes in streamflow at multi-decadal time scales are likely the result of changes in climate as well as the changes in landscape characteristics (e.g. artificial land drainage) (Sekaluvu et al., 2018; Stow et al., 2015), and streamflow increases are expected to continue in the future (Bosch et al., 2014; U.S. Environmental Protection Agency, 2013). The role that changes in streamflow characteristics play in delivery of nutrients to the WLEB is not obvious. Increased streamflow can be expected to increase transport of nutrients (Bosch et al., 2014; Jarvie et al., 2017), including nutrients stored in soils, but part of that effect may simply be a change in the timing of nutrient delivery, with the annual total transport being primarily limited by the availability of the nutrients in the soils and groundwater.

Tracking progress towards the achievement of nutrient flux reductions presents serious challenges for the data analyst. For example: 1) the shape of the concentration-discharge relationships can vary from season-to-season and vary across a period of years and yet, many of the statistical methods for computing fluxes make assumptions that these relationships only change in terms of their intercept but not their overall shape, and 2) the random year-to-year variability in discharge can overwhelm watershed responses to nutrient-reduction efforts (e.g. Kleinman et al., 2015). Studies of annual, seasonal, and spatial patterns in WLEB tributary nutrient trends have focused principally on flow-weighted mean concentrations of P and P fluxes at downstream monitoring sites near Lake Erie (Baker et al., 2014a; Daloglu et al., 2012; Jarvie et al., 2017; Maccoux et al., 2016; Sekaluvu et al., 2018; Sharpley et al., 2012; Stets et al., 2015; Stow et al., 2015; Winter et al., 2015), but these measures are strongly influenced by interannual variability thus making it difficult to detect the signal of change amidst the noise of climate variability. Tracking trends at upstream monitoring sites as well as at points entering Lake Erie is also critical to address knowledge gaps related to nutrient source area identification and patterns of change across the region in nutrient transport influencing delivery of nutrients to Lake Erie.

This paper focuses on a recently developed methodology for water-quality trend analysis, Weighted Regressions on Time, Discharge, and Season (WRTDS: Hirsch and De Cicco, 2015; Hirsch et al., 2010), that is designed to remove the often overwhelming influence of year-to-year streamflow variability on water-quality flux (or concentration) trends. The terms streamflow, flow, and discharge are used interchangeably in this paper. They all refer to the volume of water per unit time passing a given cross-section of a river (Turnipseed and Sauer, 2010). WRTDS is currently being used for ongoing evaluations of nutrient-reduction progress at nearly 100 monitoring sites to guide adaptive management decisions in the Chesapeake Bay watershed (https://cbrim.er.usgs.gov/). Our objectives in this paper are to extend this methodology to improve the ability to identify trends in annual and seasonal water-quality fluxes under nonstationary flow conditions and then apply the method to investigate underlying trends and causes of trends at long-term (20 or more years) stream monitoring sites in the WLEB.

The analytical methods developed herein seek to address not only effects of short-term (1 year to a few years) variations in hydrologic conditions on water-quality trends, but also the challenges of identifying trends and drivers of trends in a region influenced by a combination of changing land use and(or) land management practices as well as substantial changes in climate. Strengths of this method compared to conventional trend methods include the ability to identify non-monotonic trend patterns (i.e. trends that reverse directions over the period of record) and the ability to differentiate between trends in concentration versus trends in flux; and, through the additional enhancements presented here, the ability to address long-term non-stationarity in streamflow (i.e. trends or changes in streamflow patterns over time), an issue of critical importance for the Lake Erie watersheds (Daloglu et al., 2012; Jarvie et al., 2017; Stow et al., 2015; Williams et al., 2016). The results from this method additionally provide increased understanding of the relative influences of streamflow changes and land use/land management changes on water-quality trends.

Data and methods

Discharge and water-quality trend analyses focused on monitoring sites with 20 or more years of record in the WLEB region. The discharge trend analysis included a recently developed graphical approach, “Quantile-Kendall plots” (Hirsch, 2018). The original WRTDS methodology (Hirsch and De Cicco, 2015) for water-quality trend analysis is briefly summarized and the extended WRTDS method is described in detail. The extended WRTDS method includes apportioning the water-quality trend into trend components attributable to changes in discharge (i.e. discharge trend) and to changes in the concentration-discharge relationship over time. In addition to the content in this section, further details on the construction of the Quantile-Kendall plots and the extended WRTDS methodology appear in Electronic Supplementary Material (ESM) Appendices S1 and S2, respectively. The ESM to this paper and Choquette et al. (2018) provide further details on the study sites, watershed characteristics, modeling input data, and modeling results. ESM figure and table numbers referred to in the following are prefaced by S (S# or S#-#) and can be found in the ESM.

Study sites and monitoring data

The focus of the study was on tributaries in the predominantly agricultural WLEB and the Cuyahoga River basin with minimal row crops and more extensive development (urban/residential) (Fig. 1 and Table 1). Water-quality constituents selected for analysis included soluble reactive phosphorus (SRP), total phosphorus (TP), nitrate plus nitrite (NO23), total nitrogen (TN), and total Kjeldahl nitrogen (TKN: ammonia plus organic nitrogen). Trend sites in the WLEB were selected based on screening of a multi-agency water-quality data set compiled by the U.S. Geological Survey (USGS) (Argue et al., 2014; Betanzo et al., 2015b) and discharge records retrieved from the USGS National Water Information System (NWIS; U.S. Geological Survey, 2016). Years throughout this paper, unless otherwise stated, refer to water years (October 1 through September 30; and the year label designated by the calendar year in which it ends). The trend analyses presented herein utilize the streamflow metric “daily discharge” from USGS stream gauge records. The daily discharge values refer to daily mean values determined from a higher frequency time-series of field measurements and specific analytical protocols (Nielsen and Norris, 2007; Olson and Norris, 2007; Turnipseed and Sauer, 2010).

Screening criteria used for water-quality site selection included sites sampled at least monthly for a minimum of 20 years for one or more of the five targeted water-quality constituents, through water year 2015, and located at or near a continuous-record stream gauge with flow record concurrent to the water-quality records. At the two locations where more than one water-quality monitoring site met these criteria (Maumee River near Waterville, Ohio, and St. Marys River near Ft. Wayne, Indiana), the site with the most complete water-quality record, based on record length and sampling frequency, was selected for the analysis. The USGS multi-agency water-quality data set was supplemented by record-updates from the Indiana Department of Environmental Management (IDEM) (2016) and from the National Center for Water Quality Research (NCWQR) (Heidelberg University, 2016).

Ten long-term water-quality monitoring sites and stream gauges (Fig. 1 and Table 1) were selected for analysis based on the screening criteria above. Discharge data were downloaded directly from NWIS (U.S. Geological Survey, 2016) using the EGRET software (Hirsch and De Cicco, 2015), and the nutrient monitoring records included data collected and analyzed by the NCWQR and the IDEM.

Water-quality sampling and analytical methods are described for the NCWQR sites in Baker et al. (2014a), Stow et al. (2015), and Jarvie et al. (2017), and for the IDEM sites in Beckman (2017). TP and TKN were analyses of unfiltered samples; SRP included only filtered samples (0.45-micron membrane filter); NO23 included either filtered (NCWQR sites) or unfiltered (IDEM sites) samples but sampled as such consistently over the period of record. SRP was measured only at the six NCWQR sites (Table 1). SRP is considered equivalent to dissolved orthophosphate or dissolved inorganic P, and excludes all but trace amounts of dissolved unreactive hydrolyzable P (polyphosphates plus some

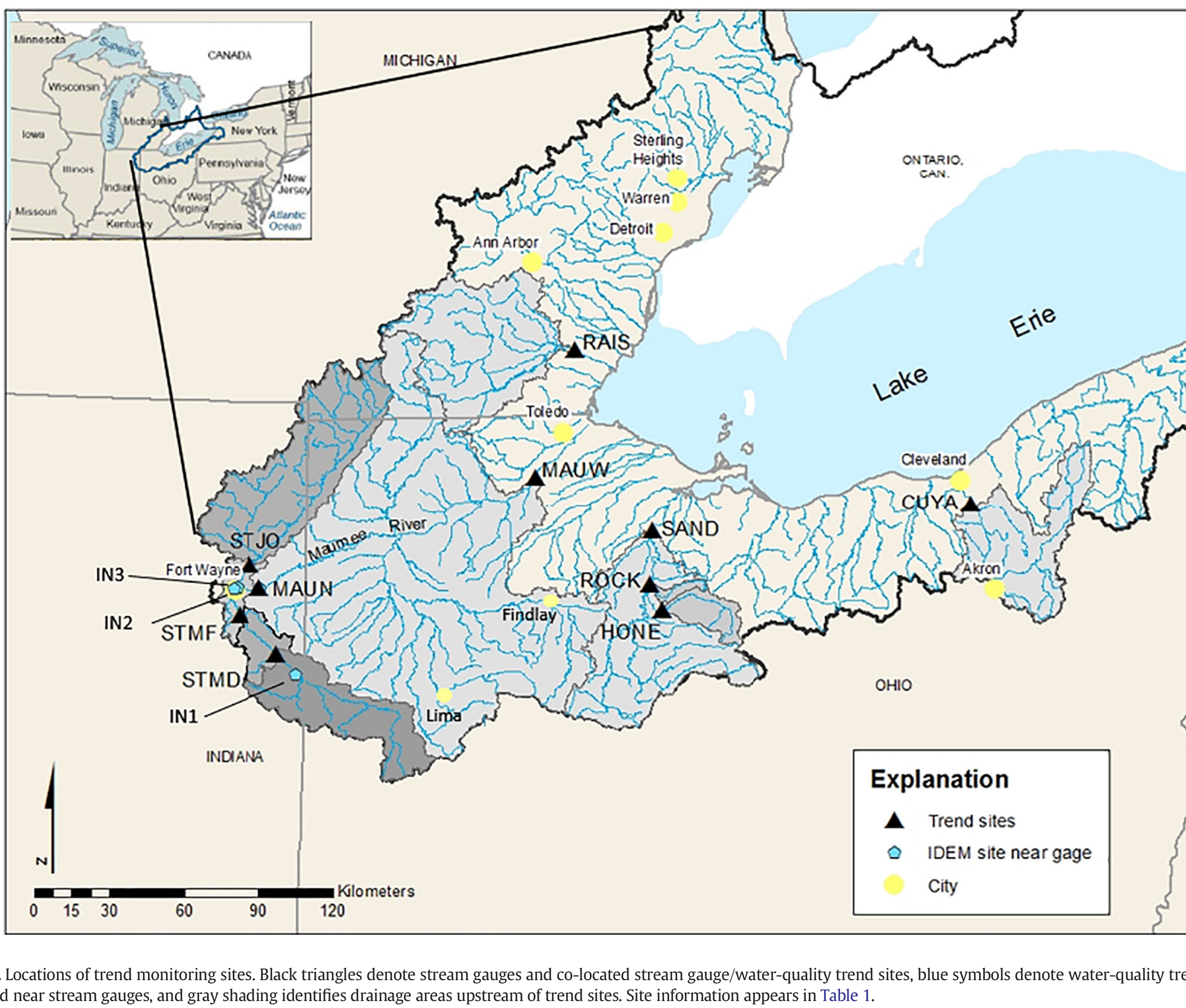

Fig. 1. Locations of trend monitoring sites. Black triangles denote stream gauges and co-located stream gauge/water-quality trend sites, blue symbols denote water-quality trend sites located near stream gauges, and gray shading identifies drainage areas upstream of trend sites. Site information appears in Table 1.
[IDEM, Indiana Department of Environmental Management].

Table 1
Summary of water-quality and streamflow monitoring sites included in the trend analyses. Site locations appear in Fig. 1.

Site abbreviation	Map ID in Fig. 1	Water-quality site number	Water-quality monitoring organization[b]	USGS streamgauge number	Site drainage area (km^2)	Start year of water-quality record	Site or streamgauge name	Land use in watershed[a] (in percent)				
								Developed	Forest	Row crops	Pasture-land	Other[c]
CUYA	CUYA	04208000	NCWQR	04208000	1831	1982	Cuyahoga River at Independence, OH	40	33	8.9	8.4	9.7
HONE	HONE	04197100	NCWQR	04197100[d]	386	1977	Honey Creek at Melmore, OH	6.7	9.7	81	1.5	1.1
MAUW	MAUW	Streamgauge		04193500	16,395		Maumee River at Waterville, OH	11	6.4	73	5.4	4.2
MAUW	MAUW	04193490[e]	NCWQR	04193500[f]	16,351	1976	Maumee River near Waterville, OH					
RAIS	RAIS	04176500	NCWQR	04176500	2699	1983	River Raisin near Monroe, MI	11	11	49	18	11
ROCK	ROCK	04197170	NCWQR	04197170[d]	90	1984	Rock Creek at Tiffin, OH	8.8	11	73	6.0	1.2
SAND	SAND	04198000	NCWQR	04198000	3240	1976	Sandusky River near Fremont, OH	8.1	8.8	78	3.1	2
MAUN	MAUN	LEM010-0014	IDEM	04183000	5095	1991	Maumee River at New Haven, IN	12	8.7	62	10	7.3
STJO	STJO	Streamgauge		04180500	2745		St. Joseph River near Fort Wayne, IN	8.5	11	53	17	10.5
STJO	IN3	LEJ100-0003	IDEM	04180500[f]	2812	1992	St. Joseph River in Fort Wayne, IN					
STMF	STMF	Streamgauge		04182000	1974		St. Marys River near Fort Wayne, IN	8.6	5.6	81	2.2	2.6
STMF	IN2	LES-06-0003[g]	IDEM	04182000[f]	2009	1991	St Marys River in Fort Wayne, IN					
STMD	STMD	Streamgauge		04181500	1608		St. Marys River at Decatur, IN	8.6	5.3	82	2.3	1.8
STMD	IN1	LES040-0007	IDEM	04181500[f]	1310	1992	St Marys River north of Pleasant Mills, IN					

[a] From National Land Cover Database 2006 (NLCD 2006: https://www.mrlc.gov/nlcd2006.php), in Falcone (2015).
[b] NCWQR, Heidelberg University National Center for Water Quality Research; IDEM, Indiana Department of Environmental Management.
[c] Includes sum of the NLCD 2006 land-use categories other than those listed in table.
[d] USGS Hydroclimatic Data Network (HCDN) site (Lins, 2012), and GAGES-II reference-quality classification (Falcone, 2015); water quality site moved from gauge to 4.02 km upstream as of 2/9/2010 (no sample record 8/14/2009–2/9/2010) (Baker et al., 2014a).
[e] Water quality sampling location is at USGS Site ID 04193490; NCWQR site ID is 04193500.
[f] Streamgauge located near water quality site.
[g] Prior site number was LES060-0004, moved to LES-06-003 on 8/18/2011.

organic phosphorus), and typically represents the majority of the total dissolved P fraction in natural waters (Jarvie et al., 2002, and Sprague et al., 2017, provide further details). TN was calculated as the sum of TKN plus NO23. Particulate phosphorus was estimated as the difference between TP and SRP (Baker et al., 2014a discuss the bioavailability of the dissolved and particulate phosphorus fractions).

Sampling frequency at the NCWQR sites was daily or more frequent; the IDEM sites were sampled at a monthly frequency. As a consequence, the sample sizes used in analyses for the IDEM sites were much smaller than for the NCWQR sites (for example for the 1995–2015 period, there were more than about 10,230 values per site for TP at the NCWQR sites, versus about 245 values per site for the IDEM sites). Both the NCWQR and the IDEM sampling records covered the range of low to high flows, however, the NCWQR samples included more complete coverage in the range of highest flows. The NCWQR monitoring protocol increases sampling to 3–4 times per day during periods of high flow or high turbidity. The potential effects of these differences between sampling protocols for this study are likely more accurate WRTDS estimates of annual and monthly nutrient fluxes for the NCWQR sites as demonstrated by Lee et al. (2016) who present information on the effects of different sampling protocols on the accuracy of load estimates from WRTDS and other load-estimation methods.

Land use, reservoir storage, and information on watershed attributes upstream of the trend sites, which includes 1982–2012 time-series data on agricultural practices (Falcone, 2017a, b), are summarized in Table 1 and ESM Appendix S3. The Cuyahoga River site (CUYA; Fig. 1) with limited row-crop agriculture (9% of the watershed) was included to provide a contrast with predominantly row-crop conditions in the nine WLEB watersheds, in which extent of row-crop agriculture ranged from 49% to 82% (mean 70%) of the watershed (Table 1). Soil runoff and erosion potential are generally highest in the St. Joseph and the upper reaches of the Raisin, Sandusky, and Maumee tributary watersheds (Betanzo et al., 2015b; Lund et al., 2011). Tile drainage is extensive in the WLEB and, although documentation is limited, there have been increases in the spatial extent, density, and efficiency of tile drainage in the region in recent years (Kalcic et al., 2016; LimnoTech, 2017; Wieczorek and LaMotte, 2010). Among these WLEB study sites, tile drainage is less prevalent in the Raisin and St. Joseph basins which include areas of more highly drained soils relative to the other watersheds (Betanzo et al., 2015b; Jarvie et al., 2017; Kalcic et al., 2016). The effects of contrasting characteristics across these watersheds on observed discharge and water-quality trends are discussed in "Results" and "Discussion".

Exploring streamflow trends

The modifications of WRTDS presented here specifically address statistically partitioning the effects of long-term streamflow trends on water-quality trends. Rather substantial streamflow trends over the past 3 decades have been recognized in the WLEB (Daloglu et al., 2012; Stow et al., 2015; Williams et al., 2016). These streamflow trends are believed to play a significant role in phosphorus trends in the western Lake Erie drainage basin (International Joint Commission, 2014; Jarvie et al., 2017; Michalak et al., 2013; Reutter et al., 2011; Richards et al., 2010; Stow et al., 2015). Maccoux et al. (2016) note in describing changes in total nutrient loading to Lake Erie, that phosphorus changes "are driven by early and recent improvements in point source discharges, but are confounded by recent increases in nonpoint source loads that may in turn be due to increasing trends in precipitation and river discharge". Clearly, additional tools are needed to increase the power of our analyses to detect trends in the water-quality parameters by better separating the effect of increasing streamflow; and possibly in the future, separate other factors of influence in addition to flow.

To gain some appreciation of the nature and extent of the streamflow changes in the WLEB, we explored temporal changes in daily discharge statistics across an annual time period including annual maximum day, minimum day, median daily, and mean daily using the EGRET software (Hirsch and De Cicco, 2015), and also used a recently developed type of plot, the Quantile-Kendall plot (Hirsch, 2018) described in ESM Appendix S1, to explore discharge trends at the study sites. The Quantile-Kendall plots, derived using daily discharge records, are used to evaluate discharge trends across the range of discharge values at a given site for a specified "season" or time-frame (e.g. annual, multiple-month season, or a specific month). The Quantile-Kendall plots for an annual time-frame show results of 365 Mann-Kendall trend tests, and the associated Thiel-Sen slope estimates, for each of the 365 ranks (order statistics) over a specified period of years (i.e. trends are evaluated for the annual minimum day discharge, the 2nd lowest discharge for each year, … the median daily discharge, … and the maximum day discharge).

In these plots, the trend slopes are computed using the Thiel-Sen slope estimator (Sen, 1968; Thiel, 1950) of the logarithms of the values shown and then transformed to percentage changes per year (Hirsch and De Cicco, 2015). The strength of the statistical evidence for these trends is evaluated using the Mann-Kendall trend test, adjusted for serial correlation using the adjustment method proposed by Yue et al. (2002). The strength of the evidence is characterized by the likelihood that the direction of the estimated trend is correct, computed from the Mann-Kendall test p-values as $[1 - (p / 2)]$ (see ESM Appendix S1 for details).

Evaluating nutrient trends

This paper introduces an extension of the existing WRTDS method, which we call "generalized flow-normalization" (and denote as FNG), used here to perform trend analysis on fluxes of five nutrient constituents (SRP, TP, TN, NO23, TKN) for the period 1995–2015. FNG allows for the explicit consideration of trends in discharge as well as trends in the concentration-discharge relationship (CQR). The FNG method was also used to explore the relative influences of trends in discharge and changes in the CQR on overall water-quality trends and to assess changes in ratios of nutrient species and in the seasonal patterns of SRP flux trends. The changes in these annual and seasonal flux patterns and nutrient ratios are much more stable than they would be if flow normalization was not used, and as such they can significantly improve the ability to interpret the underlying water-quality changes taking place, as opposed to the discharge-fluctuation-driven changes. The original WRTDS flow normalization method, defined below as "flow-normalized flux under the assumption of stationary discharge (FNS)", was used to evaluate 2005–2015 trends in SRP, TP, and TN fluxes.

Weighted regressions on time, discharge, and season

The WRTDS method and software package (EGRET) are described in Hirsch and De Cicco (2015) and Hirsch et al. (2010, 2015) and are briefly summarized here to illustrate the original method, which remains an integral part of the expanded methodology that addresses nonstationary flow conditions. WRTDS is a multivariate smoothing approach that uses water-quality data and an integrated probability distribution of discharge conditions to estimate a time series of "flow-normalized" (FN) fluxes. The purpose of flow-normalization in WRTDS is to reduce or eliminate effects of year-to-year variability in discharge on the record of trend in water quality. The WRTDS method defines the CQR at a daily scale, allowing the CQR to vary smoothly across seasons and years. This CQR is then integrated over a probability density function (pdf) of daily mean discharges that is associated with (i.e. indexed to) each day of the year, which then provides a daily time series of smoothly varying estimates of flow-normalized fluxes (or concentrations).

Here we focus on flow-normalized fluxes, as opposed to flow-normalized concentrations, because the cHAB concern is related to reducing the total nutrient inputs delivered to a large receiving water body (Lake Erie) and evaluating progress towards nutrient-reduction goals. The concept of WRTDS is that a data set of hundreds to thousands of water-quality samples collected at a river monitoring site in conjunction with daily discharge values at a nearby stream gauge can be effectively summarized by a CQR that changes gradually as a function of time and season. The WRTDS analytical system produces a CQR specific to every day of every year in the record. The CQR changes gradually as a function of time of year as well as year-to-year capturing gradual changes associated with changes in land use, land-use practices, and waste discharges that take place throughout the watershed.

WRTDS-estimated CQRs and the ability of the WRTDS method to capture changes in the CQR over time are illustrated by an example (Fig. 2a) showing SRP concentration-discharge relationships for two specific dates for the study site Honey Creek at Melmore, Ohio (HONE). For this particular time of year (early August), the expected value of concentration increased between 1995 and 2013 over a range of discharge values that covers more than two orders of magnitude. At low discharges (around 0.05 m^3/s) the increase in concentration was rather small (about 30%) but at high discharges (around 10 m^3/s) concentrations approximately doubled.

Placing these two CQRs in a broader context, Fig. 2b shows, in the form of a contour plot (the colors indicate the expected value of concentration for any combination of date and discharge), the set of CQRs for a period from 1990 to 2016. The two vertical lines are placed here on the dates for the specific CQRs shown in Fig. 2a. The pattern in this contour plot (Fig. 2b) clearly shows that the CQR is generally seasonal, that the highest concentrations are typically at the highest discharges, and that between the 1990s and about 2005 the concentrations associated with any given date and time of year increased substantially over those years. The plot also shows that there has been relatively little change in the CQR (beyond the seasonal variation) from about 2005 to 2016.

WRTDS uses the time-varying CQR (depicted in Fig. 2a and b) to determine estimates of the actual flux on each day in the period of record. In the following section, capital letters denote random variables, and the lower-case letters indicate functions or constants. For simplicity, the notation is written as if a year is always 365 days in length, but the computer code is designed to handle leap years correctly. The expected value of flux (an estimate of the actual flux) on each day in the period of record is:

$$\hat{F}_{i,j} = k \cdot \hat{w}_{i,j}(Q) \cdot Q_{i,j}$$

where

$\hat{F}_{i,j}$ = the estimated expected value of flux on day i of year j, given the observed value of discharge from that date, in kg/day.
$k = 86.4$, a conversion factor to compute flux in kg/day from concentration in mg/L and discharge in m^3/s.
$\hat{w}_{i,j}(Q)$ = a function that estimates the expected value of concentration based on discharge (Q) specific to day i, of year j. This function is what is plotted, for two specific dates, in Fig. 2a.
$Q_{i,j}$ = the observed mean daily discharge (in m^3/s) on day i of year j.

The $\hat{F}_{i,j}$ values can be summarized as average values for an individual month, season, or year (in kg/day). The time series of such annual or

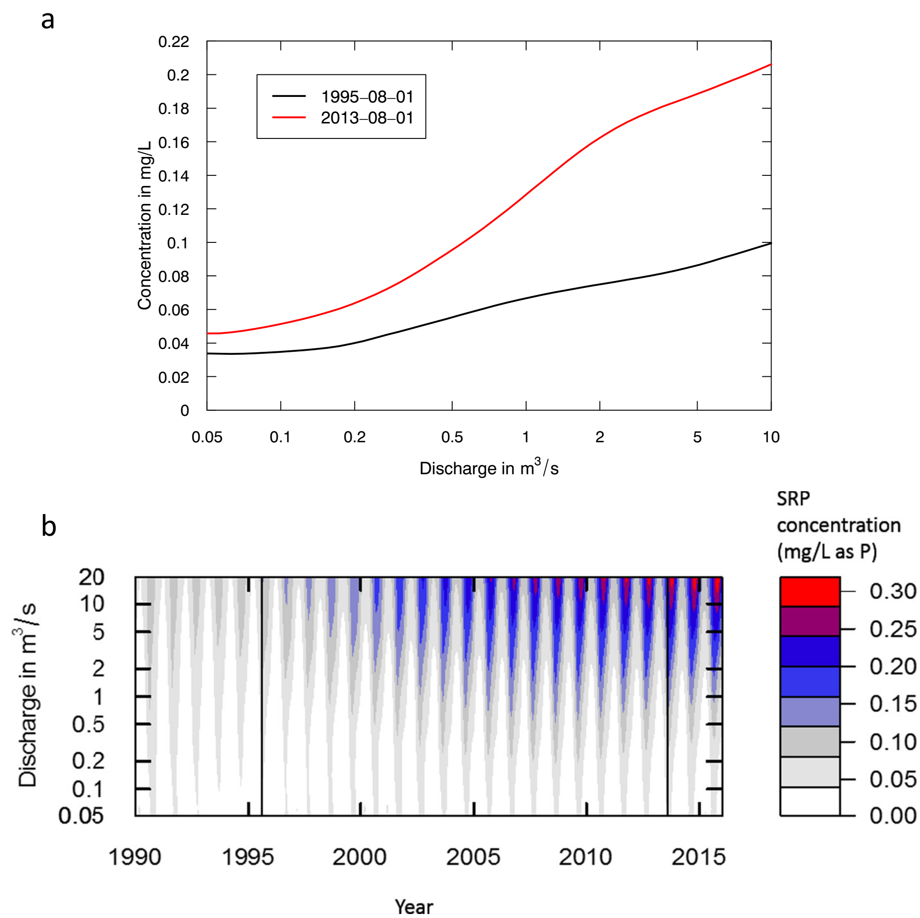

Fig. 2. (a) Example of the estimated concentration-discharge relationship (CQR) for two dates: August 1, 1995, and August 1, 2013, for soluble reactive phosphorus (SRP) for Honey Creek at Melmore, Ohio (HONE). Curves estimated by the WRTDS method. (b) A contour plot showing the SRP concentration-discharge relationship (CQR) for all dates from 1990 through 2016 for Honey Creek at Melmore, Ohio (HONE). The two vertical black lines are placed at August 1, 1995, and August 1, 2013. The colors of the contour plot indicate the expected value of SRP concentration for the given date and discharge.
[SRP data from Heidelberg University (2016) and streamflow data from the U.S. Geological Survey (2016)].

seasonal averages of the $\hat{F}_{i,j}$ are highly dependent on the set of daily discharges observed in the particular year and, as such, are a poor indicator of progress towards pollution reduction. Even when such annual average estimates are re-expressed as flow-weighted mean concentration values they remain highly correlated to discharge. This paper does not use the $\hat{F}_{i,j}$ values for our trend analysis, but it is useful to understand what they are in contrast to the FN flux values introduced below. The EGRET software does compute and display the $\hat{F}_{i,j}$ values.

It should also be noted that given the intense sampling used at HONE and the other NCWQR monitoring sites, the $\hat{F}_{i,j}$ estimates produced by WRTDS are undoubtedly less accurate than the flux estimates produced by the NCWQR, computed by a method described by Baker et al. (2014a). The NCWQR's highly intensive sampling effectively captures the event-specific details of nutrient fluxes, including hysteresis effects, that WRTDS is not designed to capture because it was developed for use with much lower sampling frequencies.

Consideration of the objective of the particular statistical analysis is crucial. If the purpose of the analysis is to create the most accurate estimate of the actual time series of nutrient fluxes, then methods such as that of Baker et al. (2014a) are ideal for such large data sets. But, if the objective is to learn about the changes in the behavior of the watershed in response to the full range of hydrologic conditions, in order to evaluate progress towards pollution reduction goals, then the WRTDS FN flux defined below is much better suited to the task.

In order to compute the WRTDS expected value of flux for any given date in a manner that does not depend on the specific discharge that happened on that date (what we call the FN flux) it is necessary to integrate the CQR over an estimated pdf of discharge (Q) for that day. The FN flux for a specific date $\hat{F}^*_{i,j}$ is defined as follows:

$$\hat{F}^*_{i,j} = k \cdot \int_0^\infty \hat{w}_{i,j}(Q) \cdot Q \cdot \hat{g}_{i,j}(Q)dQ$$

where

$\hat{g}_{i,j}(Q) =$ the estimated pdf of discharge (Q) for day i of year j.

The method used in WRTDS for estimating this discharge pdf is a non-parametric approach. It uses the observed values of discharge on day i from a set of years of discharge record that includes year j and other years in the record. The particular set of years used to determine the discharge pdf depends on whether the discharge pdf is assumed to be stationary or nonstationary over time, as discussed in the next section.

Flow normalization: extended methodology

Approaches to flow normalization. In the original formulation of WRTDS (Hirsch et al., 2010) a simplifying assumption was made to constrain the discharge pdf to be stationary over the study period. This decision was made based on the idea that for many watersheds, over the period for which adequate water-quality sample data are available (rarely

>30 years), the changes in the pdf of discharge are small enough to have little consequence to the calculated FN annual or seasonal fluxes. That decision also reflected the fact that at the time scales of 30 years, assessing discharge trends is complicated by the role of quasi-periodic oscillations that are typical of discharge records. Hirsch et al. (2010) recognized that the use of this stationarity assumption was a limitation of the method and stated that this limitation would need to be addressed as water-quality records become longer and as the extent of discharge trends become stronger due to both climatic factors and landscape changes. The extended methodology described here addresses the need that was expressed in Hirsch et al. (2010).

The definition of the "FN flux" used in the original WRTDS method (as described by Hirsch and De Cicco, 2015) assumed discharge stationarity, which under the expanded WRTDS method is equivalent to and is now explicitly termed the "flow-normalized flux under the stationary discharge assumption" (FNS), calculated as:

$$\hat{F}^{s}_{i,j} = k \cdot \int_{0}^{\infty} \hat{w}_{i,j}(Q) \cdot Q \cdot \hat{g}^{s}_{i}(Q)\, dQ$$

where

$\hat{g}^{s}_{i}(Q) =$ the estimated pdf of discharge for day i, using all years in the historical daily-discharge record.

Under the stationary discharge assumption, $\hat{g}^{s}_{i}(Q)$ is a pdf of discharge for day i, which is constant from year to year. As described in Hirsch et al. (2010), Hirsch and De Cicco (2015) and Hirsch et al. (2015), these estimated pdfs of discharge are actually point masses (based on the specific Q values observed in the record) rather than as continuous pdfs in the form of some specific distribution such as log-normal or gamma.

In the case of nonstationary discharge, where the discharge pdf is considered to be varying over time, the estimated $\hat{g}_{i,j}(Q)$ is determined using a pdf of discharge for day i which varies from year to year. The set of years used to determine the discharge pdf corresponds to a "time-window" during which the discharge pdf is assumed to be stationary. The difference between the calculations of the stationary $\hat{g}^{s}_{i}(Q)$ and a nonstationary $\hat{g}_{i,j}(Q)$ is that in the former case the estimated pdf is made using the discharge data from the entire period of discharge record, but in the latter case (introduced in this paper) the discharge pdf is based on a moving time-window. This time-window is of a set length, specified as a user option in the EGRET software, centered around each year. Therefore, the set of years in the time-window specific to year *j* will differ from the specific years in the time-window used to determine the pdfs for any other year. The details of the FNG method that capture the nonstationarity in the discharge record to produce the nonstationary $\hat{g}_{i,j}(Q)$ are described in ESM Appendix S2.

The time-window used in this study for constructing these nonstationary discharge pdfs is 15 years in duration. The choice of this time-window length is related to how rapidly the pdf of discharge evolves over time, and is distinct from the window width, used in the "modelEstimation" function in EGRET to determine the WRTDS expected values of water-quality concentration ($\hat{w}_{i,j}(Q)$) discussed earlier, which is related to how rapidly the water-quality conditions (for any given discharge and season) evolve over time (Hirsch and De Cicco, 2015 and Hirsch, 2018). However, the number of years selected for each of these two "window" values is set using subjective judgments rather than objective criteria.

We can now define three types of estimates (as averages in kg/day) for the flux for year j.

The WRTDS estimate of the actual flux for year j, based on the actual daily discharge values, is

$$\hat{F}_{j} = \frac{1}{365} \sum_{i=1}^{365} \hat{F}_{i,j}$$

The generalized flow-normalized (FNG) flux for year j is

$$\hat{F}^{*}_{j} = \frac{1}{365} \sum_{i=1}^{365} \hat{F}^{*}_{i,j}$$

The FNS flux (which assumes stationary discharge) for year j is

$$\hat{F}^{s}_{j} = \frac{1}{365} \sum_{i=1}^{365} \hat{F}^{s}_{i,j}$$

The FNS flux is what was called simply "flow-normalized flux" in previous versions of the EGRET code and associated publications.

For the FNG fluxes, we can express the change between two specific years, year j and year j + p (where p is a positive integer) as:

$$\Delta\hat{F}^{*}(j+p,j) = \hat{F}^{*}_{j+p} - \hat{F}^{*}_{j}$$

which has units of kg/day.

The uncertainty about the estimate of the overall change in FNG flux, $\Delta\hat{F}^{*}(j+p,j)$, including confidence intervals and the likelihood that the sign of the trend is correct, is determined using a block bootstrap procedure identical to the procedure described by Hirsch et al. (2015) except that here the representation of the discharge pdf is the year-specific distribution for each of the years (as described above and in ESM Appendix S2). The block bootstrap approach resamples the data set, with replacement, but it resamples entire time blocks of 200 days, rather than selecting samples one at a time. The justification for this approach is described by Hirsch et al. (2015).

For each bootstrap replicate the entire $\hat{w}_{i,j}(Q)$ function is estimated and then integrated over the pdf of discharge, $\hat{g}_{i,j}(Q)$, to compute a bootstrap estimate of $\hat{F}^{*}_{j}$ for that replicate. The bootstrap process, carried out using 500 replicates, provides estimates of the uncertainty that arises from the water-quality sampling in combination with the WRTDS method of computing the $\hat{w}_{i,j}(Q)$ function. It does not consider any uncertainty in the discharge pdfs because they are based on a complete enumeration of the record, rather than a random sample like the water-quality data. This bootstrap process is described in the documentation to the package EGRETci (EGRETci 2.0 at https://cran.r-project.org/).

It is worth noting, that although the results of this analysis are specifically defined as being the difference between 2015 and 1995, these results are strongly influenced by data collected over many years around those two years. The WRTDS method is fundamentally a smoothing method and the smoothing windows both for water quality and for discharge information are large (typically about 14 or 15 years wide). Thus, the 1995 value is not just a representation of what happened in 1995 but it is rather an integrated representation of the system (from both a concentration and a discharge perspective) over a much longer period.

The EGRET software provides a means of computing and graphing these smoothed annual flow-normalized flux values over the entire period of study (using the runSeries function) to show the broad outlines of how the system is changing over time. For simplicity in this paper we have used the approach of comparing the smoothed representation of the system for just two specific years (using the runPairs function in EGRET).

Calculating trend components. For the FNG fluxes, in addition to determining the overall size of the flux change from year j to year j + p, we are interested in knowing how much of that change can be attributed to the change in the CQR over that period and how much of it we can attribute to the change in the discharge pdf. The first of these components of the flux change, which we call the "CQ Trend Component" (CQTC), is computed as:

$$\Delta\hat{F}^{S}(j+p,j) = \hat{F}^{S}_{j+p} - \hat{F}^{S}_{j}$$

The CQTC is the average FNS flux for year j + p minus the average FNS flux for year j. By design, it eliminates from the calculation the influence of any nonstationarity in discharge. This means that the only source of change in this computation is a change in the CQR. The other component of the flux change, which we call the "Q Trend Component" (QTC), is computed as:

$$\Delta\hat{F}^{Q}(j+p,j) = \Delta\hat{F}^{*}(j+p,j) - \Delta\hat{F}^{S}(j+p,j)$$

These two components (CQTC and QTC) may be of interest when considering what part of the change in flux may be controllable by land and waste management versus the part of the overall change that may be a result of climate change. The CQTC is that portion of change that is a clear outcome of changes in availability of nutrients from the variety of point and non-point sources in the watershed, and arguably it may be feasible to make land-management and waste-water treatment changes that influence the CQTC. In contrast, the QTC is, at least in part, controlled by climate, which is outside the control of those who manage the landscape or point sources.

However, the dichotomy between CQTC and QTC "change-drivers" is only an approximate one because both climatic changes and human activities on the landscape can, in some situations, influence both of these trend components. For example, the QTC can also be influenced by land and water management activities in the watershed that substantially alter runoff responses or groundwater recharge (e.g. management of water impoundments or engineering changes in land drainage, such as tile drains, wetland drainage, or increased impervious surfaces). And the CQTC could be altered by climatic changes that could increase or reduce source supply and(or) delivery to streams (e.g. increased temperatures potentially could increase denitrification rates, in turn reducing nitrate source-availability and lowering the C-Q curve).

Examining water-quality trend results through the "lens" of the CQTC and the QTC, and comparison of their change patterns relative to each other, may provide insights into the changes taking place in the physical hydrologic and source-supply system. The case where the CQTC and QTC are both increasing, functioning as reinforcing influences, indicates that availability of a nutrient is increasing with an increase in discharge. This can occur either because the increased movement of water through the system is depleting stored nutrients or because land owners are increasing their rate of net delivery of the nutrient to the watershed.

In some cases, the CQTC and QTC changes may be in opposition, and the net result or "total trend" may indicate little change in the flow-normalized flux, effectively masking these underlying change patterns in the hydrologic system. For example, when the CQTC has a negative sign (i.e. changes in the CQR lead to lower concentrations over a wide range of discharge and season conditions) and the QTC has a nearly equal positive sign (as a result of higher discharges). An important implication of this type of case, which is commonly known as "supply limited", is that merely studying how concentrations have changed for given values of discharge and time of year may lead to erroneous conclusions because in such cases increasing discharge does not result in increasing flux.

One additional step taken with these flux results is to re-express these flux values as yield values (mass per unit time per unit of watershed area). This is helpful for comparing fluxes and flux changes over the set of sites being considered. Thus, the figures and tables present annual fluxes in units of kg/km^2. Flux trends, including FNG, CQTC, and QTC changes, are described here as the amount of change between the start and end years of the trend period, in units of yield (kg/km^2) or in percent (the ratio of change in yield to the yield for the initial year in the trend period).

The documentation and R-software for the FNS trend computations is in Hirsch and De Cicco (2015), and the documentation for the enhancements including the FNG trend method introduced here is in "Guide to EGRET 3.0 Enhancements" in the EGRET package version 3.0 (at: https://cran.r-project.org/). Documentation and R-software for the trend uncertainty estimates determined using the WRTDS bootstrap procedure is in Hirsch et al. (2015), and documentation for the enhancements that include FNG trends is in "Guide to EGRETci Enhancements" in the EGRETci package 2.0 (at: https://cran.r-project.org/).

Although this paper does not explicitly consider trends in concentration, the EGRET package considers not only trends in flux but also trends in concentration. The EGRET 3.0 software also can accommodate abrupt changes in the discharge regime (e.g. dam construction or removal) or in pollution sources (e.g. major upgrades of a treatment plant that is a dominant source of a pollutant in a watershed), but that approach was not applied for this study because such conditions were not present, and agricultural non-point sources dominate the nutrient budgets of the WLEB watersheds (George et al., 2015; Kane et al., 2014; Maccoux et al., 2016).

Results

Streamflow trends

Using the site Honey Creek at Melmore, Ohio (HONE), for the period from April 1, 1987, through March 31, 2016, as an example, trends in daily discharge statistics for an annual time-frame are shown in Fig. 3. Positive trends are apparent in all four annual statistics, although the slopes, indicating trend magnitude, are quite different. The largest slope is associated with the annual minimum day discharges (5.9% per year). The smallest slope is for the median daily discharge (1.3% per year). A trend of 5.9% per year in the slope of the annual minimum day indicates a total increase in the annual minimum day's discharge over this 29-year period of 427%. The trend in the mean daily discharge of 2.6% per year translates to a total increase of 111% over this period. Given this information, it is reasonable to suggest that the trend in discharge across the full range of the discharge distribution is positive and of substantial magnitude, particularly for the lower and upper extremes of the discharge distribution.

Continuing with the HONE streamgage as an example, trends in discharge across the range of discharge for HONE are shown in the Quantile-Kendall plot in Fig. 4. There are substantial positive trends at all parts of the flow duration curve, and these trends can be considered very likely or highly likely upwards for almost all order statistics in the lower quartile of the distribution, and for all discharges in the upper decile of the distribution. The percentage increases in low flows were much more substantial than those at high flows and were the least substantial near the median of the distribution (but even there, the trends are classified as likely upwards). Quantile-Kendall plots comparing discharge trend results among nine of the study sites for the period from April 1, 1987, through March 31, 2016, are shown in Fig. 5. The HONE period of record shows the overall greatest evidence for discharge trends among these sites. From the perspective of nutrient loading trends, the high-flow discharge trends tend to be the most influential due to the high proportion of total load transported during higher flows.

It is beyond the scope of this paper to fully describe the causes of the trends or the various similarities and differences among the flow-trend results for these sites. However, a few observations can be made regarding the high-flow discharge trends that we identify at 8 of the 9 sites. The first is that with only a few exceptions, the upper quartile of the daily discharge distribution shows very likely to highly likely positive trends, typically in the range of 2% per year. The exceptions are the

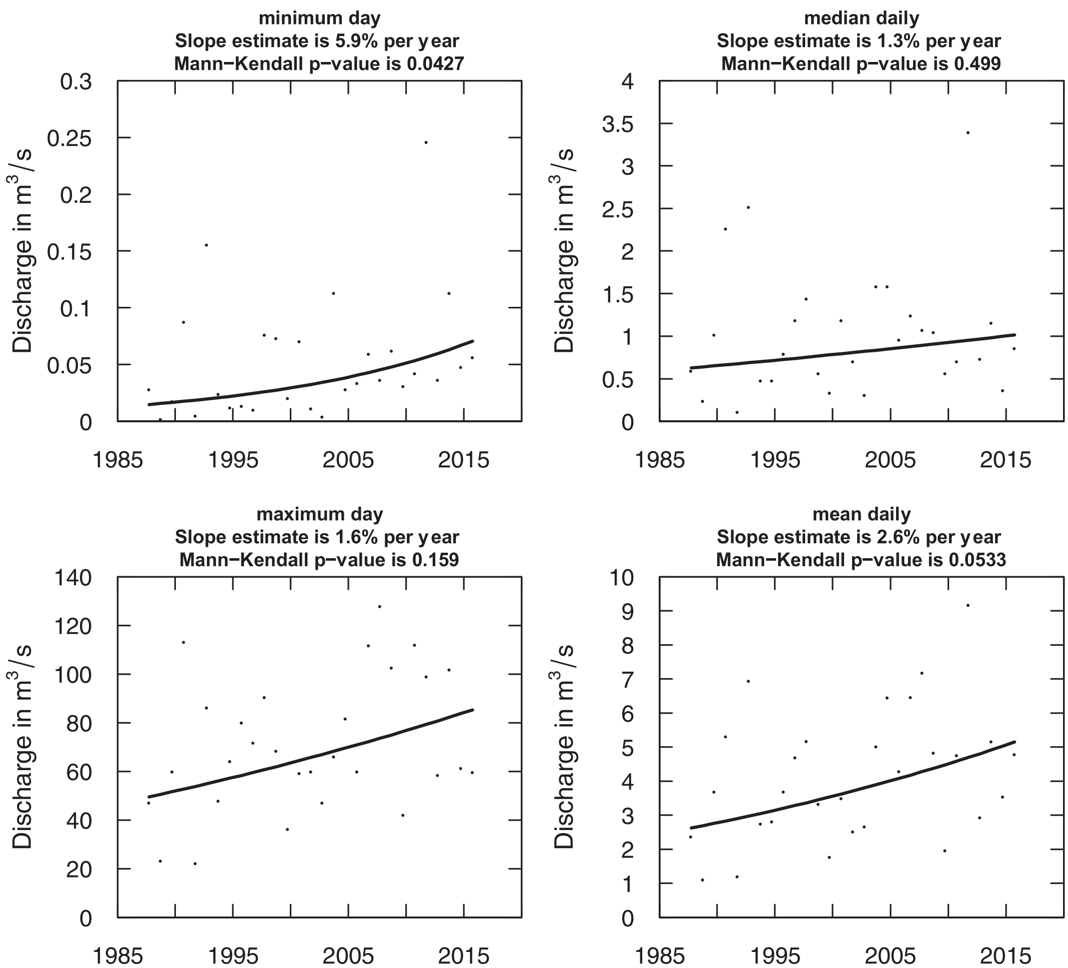

Fig. 3. Discharge (streamflow) trends for the period 1987–2016 for four annual discharge statistics: annual minimum day, maximum day, median daily, and mean daily. The statistics are determined from the daily discharge record for the stream gauge Honey Creek at Melmore, Ohio (HONE; Table 1 and Fig. 1). Each panel shows a Thiel-Sen slope estimate expressed in percentage change per year, and a two-sided p-value for the Mann-Kendall trend test.

Raisin and Cuyahoga River sites (RAIS and CUYA), both with likely to very likely upper-quartile trends of lower magnitude, and the St. Joseph River site (STJO) (Fig. 5) where the direction of the upper-quartile trend was rather uncertain. The lower magnitude or less clear high-flow trends at these three sites are possibly related to reservoir effects and may also be influenced by differences in land use and in extent of agricultural drainage modification. CUYA and RAIS have relatively substantial reservoir storage (volume as measured in days of mean discharge, ESM Appendix S3), which is about twice that of any other sites; STJO shows moderate reservoir storage located a short distance upstream of the streamgage as well as upstream inter-basin flow exports (ESM Table S3-1). These three watersheds also have the lowest row-crop land use and higher watershed proportion of forest and pastureland relative to the other trend sites (Table 1); they are located outside of the region where clay soils are most pervasive, and estimated tile drainage is less extensive in croplands of RAIS and STJO (Betanzo et al., 2015b; Jarvie et al., 2017). The similar land-use patterns for STJO and RAIS, showing less row crop and more pasture area relative to the other sites located in the WLEB, reflect the steeper topography in these watersheds that is less amenable to row cropping.

Trends in the range of low to moderate discharges lack consistency across the sites. Trends near the medians of the discharge distributions tend to be smaller positive values than the trends at the higher discharges and are even small negative values in some cases (Fig. 5). Trends at low flow are quite variable across all of the sites, although in those cases where they are highly likely they are positive trends. The three sites in the Sandusky River basin (HONE, ROCK, and SAND) and the Cuyahoga River site (CUYA) show the only substantial positive trends at low flow.

Monthly discharge trends at the sites during the 1994–2016 period, presented in ESM Appendix S4 and ESM Fig. S4-1, show variation across the year but also indicate some regionally consistent patterns. The majority of these sites show increasing high flows during March, November, and December, although the November increases were relatively uncertain. All of the sites show increases in December high flows at the upper 2 quartiles flow range with trend slopes as much as 6 to 10% per year and very likely to highly likely at 6 of the 10 study sites. During March, medium- to high-flow increases (1–4% per year) are apparent at all sites. Increases in low- to medium-flow trends (lower 2 quartiles) during January and June (3–8% per year) occurred at most of the sites. January low to medium flows show increases at all 10 sites except RAIS, and little or negative changes in higher flows. The three Sandusky basin sites (SAND, HONE, and ROCK) show increases across all flow ranges during late summer and fall (September through December) and low-flow increases, some of which are highly likely, extend from August through January.

Nutrient trends and sources of change

Multidecadal (1995–2015) nutrient flux trends were evaluated using the FNG method. The multi-decadal trend analyses focused on changes in annual and seasonal (monthly) nutrient fluxes, nutrient fractions (total versus dissolved), and nutrient ratios. The multidecadal trend results for the 2 phosphorus and 3 nitrogen parameters (Figs. 6

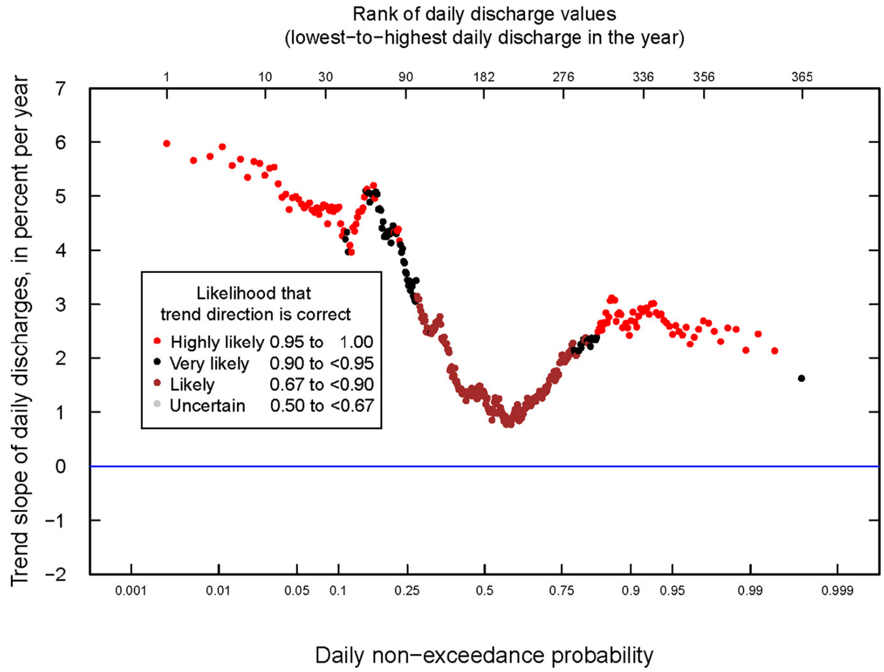

Fig. 4. Quantile-Kendall plot showing 1987–2016 trends in discharge at Honey Creek at Melmore, Ohio (HONE). Daily discharge values were ranked from 1 (the lowest rank) to 365 (the highest rank) within each year. Each point represents the estimated trend slope (expressed in percent change per year) for discharge values of the given rank (1 through 365). Low-flow trends are at the left and high-flow trends are at the right. The colors indicate, for each rank, the likelihood that the direction of the estimated trend is correct. See text and ESM Appendix S1 for additional description of the Quantile-Kendall plots.

and 7, respectively) include the 90% confidence intervals for the FNG flux change and the components of change (QTC and CQTC) attributable to systematic changes (long-term trends) in discharge (i.e. the discharge pdf) and to changes in the concentration versus discharge relationship (the CQR), discussed in above section "Flow normalization: extended methodology". Figs. 6 and 7 express the uncertainty of the trend directions using likelihood values (likelihood that the estimated trend value has the correct sign), rather than p-values (see Hirsch et al., 2015). This is similar to the trend assessment procedure described by McBride (2018). These trend results are summarized in ESM Table S1 and by map location in ESM Fig. S1. Recent decadal (2005–2015) trends in flux determined for SRP, TP, and TN, using the FNS method, are presented in ESM Appendix S5.

Dissolved phosphorus

The SRP annual flux trends at all 5 WLEB sites show the largest percentage change (+109% to +322%) among all nutrient parameters analyzed for the 1995–2015 period, with a highly likely probability of (+) trend direction (likelihood in excess of 0.999) at all of these sites (Fig. 6). At the CUYA site the SRP trend was minimal (−2%) with uncertain trend direction. The 1995 SRP yields were similar among all 6 sites; by 2015, SRP yields at the WLEB sites had substantially increased while CUYA showed essentially no change (ESM Table S1). The magnitude of the yield changes varied across the six sites, ranging from −0.3 kg/km^2 for CUYA, +8 at RAIS, +20 at MAUW, +24 at ROCK, +30 at SAND, and +46 at HONE, and shows strong positive correlation to extent of row-crop agriculture among these watersheds. The four largest positive (+) yield changes among these 6 watersheds occurred at the four sites having the highest percentage of row-crop land use (MAUW, SAND, HONE, and ROCK). In addition to proportion of basin in row crops, other differences among these watersheds likely contributed to spatial differences in SRP trends. The 2005–2015 SRP trends indicate minimal increases at 4 of the 5 WLEB SRP sites (+3 to +8%), and Honey Creek (HONE) shows a somewhat larger, highly likely increasing trend (+31%) (ESM Fig. S5-1 and Table S5-1).

For the five WLEB watersheds, the SRP CQTC, as percent of 1995 yield, averaged +137% (range: +77 to +227%) compared to an average QTC of +59% (range: +30 to +96%) (Fig. 6 and Table S1). The importance of this finding is that it suggests the SRP FNG flux trend can partly be attributed to the (+) change in runoff characteristics (the QTC, which is probably a function of both climate change and land-use change), but the (+)CQTC is the dominant part of the (+)FNG trends. Thus, factors such as the availability of SRP in the soil and groundwater system and the mobility of SRP have been the primary drivers of flux change, while changes in the amount and timing of water moving through the hydrologic system have been responsible for only about a third of the SRP flux changes at these sites. All 5 of the WLEB sites represent what we refer to as an "additive" CQTC and QTC situation because both components (QTC and CQTC) of trend are in the same direction resulting in a stronger FNG trend.

Total phosphorus and nitrogen parameters

The analyses of 1995–2015 TP, NO23, TN, and TKN flux trends included 10 sites, the six SRP sites along with four additional sites in the upper Maumee basin (Fig. 1 and Table 1). At site STMD, the trends for TKN and TN correspond to 1999–2015 (limited by available record). Annual TP fluxes at 5 of the 9 WLEB sites show highly likely trends, including 4 positive trends (34 to 71% change) and 1 negative trend (−36%) (Fig. 6 and ESM Fig. S1). The three largest increases included Honey Creek (HONE) and the two sites on the St. Marys River (STMD and STMF) showing changes of +68 to +82 kg/km^2. In contrast, the other major tributary of the Maumee River, the St. Joseph River (STJO), and RAIS had changes of −36 and −7 kg/km^2 (ESM Table S1). The STJO site was the only WLEB site with clear reductions in TP flux. Given that the St. Marys and the St. Joseph Rivers are the two primary tributaries draining into the upper Maumee River, it is not surprising that they roughly balance out such that the site on the upper main stem of the Maumee River (MAUN) had very small TP change that was about as likely to be positive as negative, which was similar at the lower Maumee site (MAUW) (+4 and +6 kg/km^2, respectively).

Of the sites evaluated for TP and the N species, only one site, STJO, shows negative values for the QTC changes in yield (Figs. 6 and 7; ESM Table S1) and these are all very small negative values. The Quantile Kendall plots (Fig. 5) show that the discharge trends for STJO are

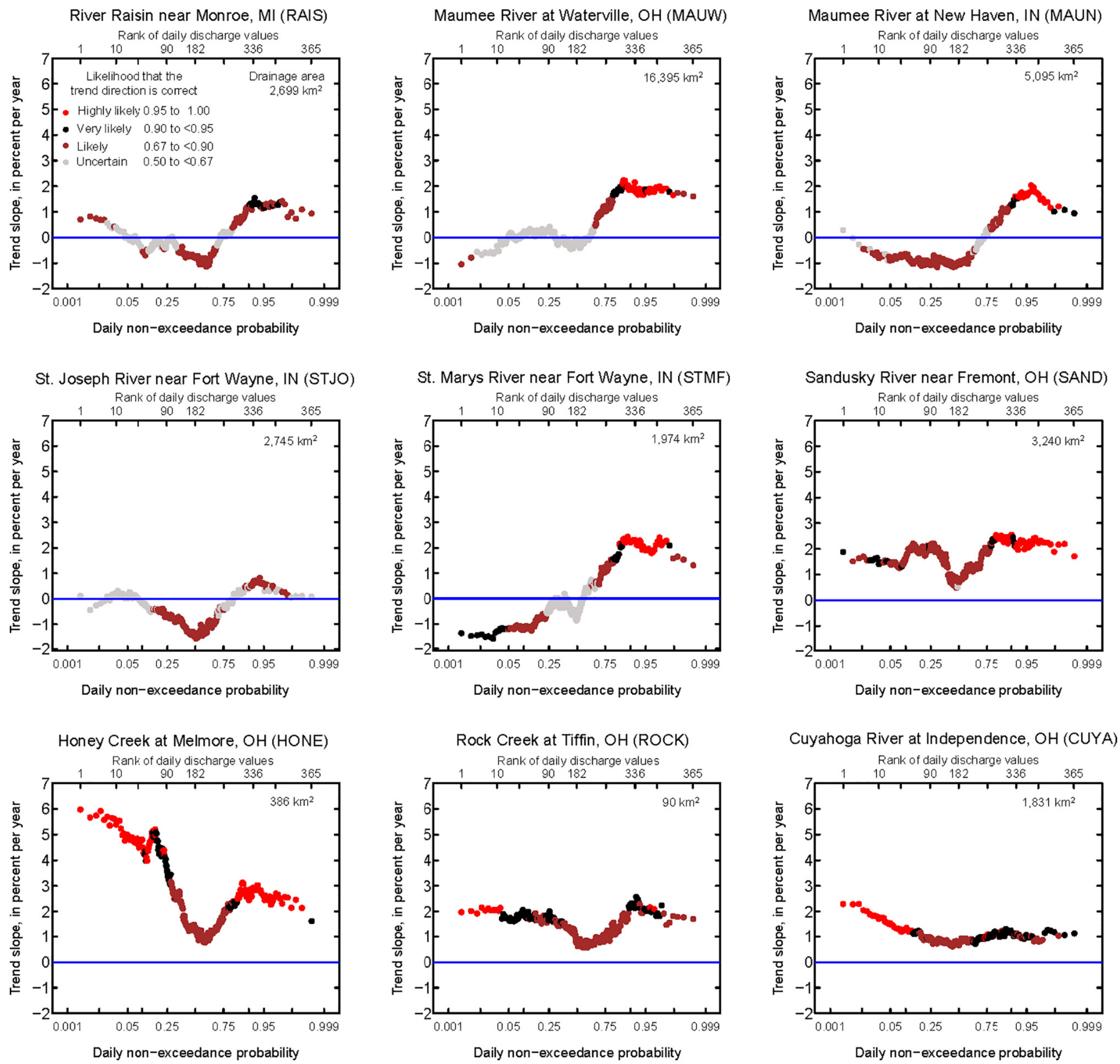

Fig. 5. Quantile-Kendall plots showing 1987–2016 trends in discharge at nine of the trend sites used in this study. The plots show the magnitude of the trends across the range from low to high flows (left side to right side, respectively) for each site, based on annually ranked daily discharge data. Colors indicate, for each rank, the likelihood that the direction of the estimated trend is correct. Upstream drainage area for these gages is listed in km^2. See text and ESM Appendix S1 for additional description of the Quantile-Kendall plots.

different from all of the other sites, particularly in the fact that they show virtually no trend throughout the upper quartile of discharge, possibly reflecting, as previously mentioned, upstream regulation, less extensive area in row crops relative to most of the other WLEB sites, and interbasin flow diversions (ESM Table S3-1). For all of the other 9 sites, if the CQTC changes were minimal (meaning that for a given discharge at a given time of year, expected concentration did not change) we would see an increase in yield equivalent to the QTC increase. In actuality at many of the sites part of the (+)QTC change is counterbalanced by a reduction in the CQTC, resulting in a smaller overall trend. Of these opposing cases for TP trends (Fig. 6), there are five sites where (+)QTC and (−)CQTC changes approximately cancel each other out, resulting in a yield change close to zero (RAIS, MAUW, MAUN, ROCK, and CUYA). Generally, the TP trends at these sites appear to follow a dilution type of behavior, where the increases in discharge are approximately balanced by a decrease in concentrations, resulting in only a very small change in yield. The other sites, SAND and HONE, where the (+)QTC change is much larger in absolute value than the (−)CQTC change, show highly likely positive overall trends in TP yield (35 and 58%, respectively). The only sites that show (+)CQTC changes for TP are the two St. Marys River sites (STMF, STMD) where about two-thirds of the increase can be attributed to the QTC increases and one-third to the CQTC increases. TP results for the STJO site were distinctly different from the rest of the sites, with a strongly negative trend that was entirely a result of the CQTC decreases.

If we consider the changes in annual TP yields in conjunction with the changes in SRP, on the basis of the six sites where both TP and SRP were monitored, the difference between the SRP change and the TP change can be used to infer a change in particulate phosphorus (PP). Of the five WLEB sites, there are three sites with substantial decreases in PP: RAIS, MAUW, and ROCK with changes of −15, −14, and −5 kg/km^2, respectively. There are two sites with increases in PP, HONE and SAND with an increase in PP of +22 and +11 kg/km^2, respectively. To generalize from these results we can say that efforts to

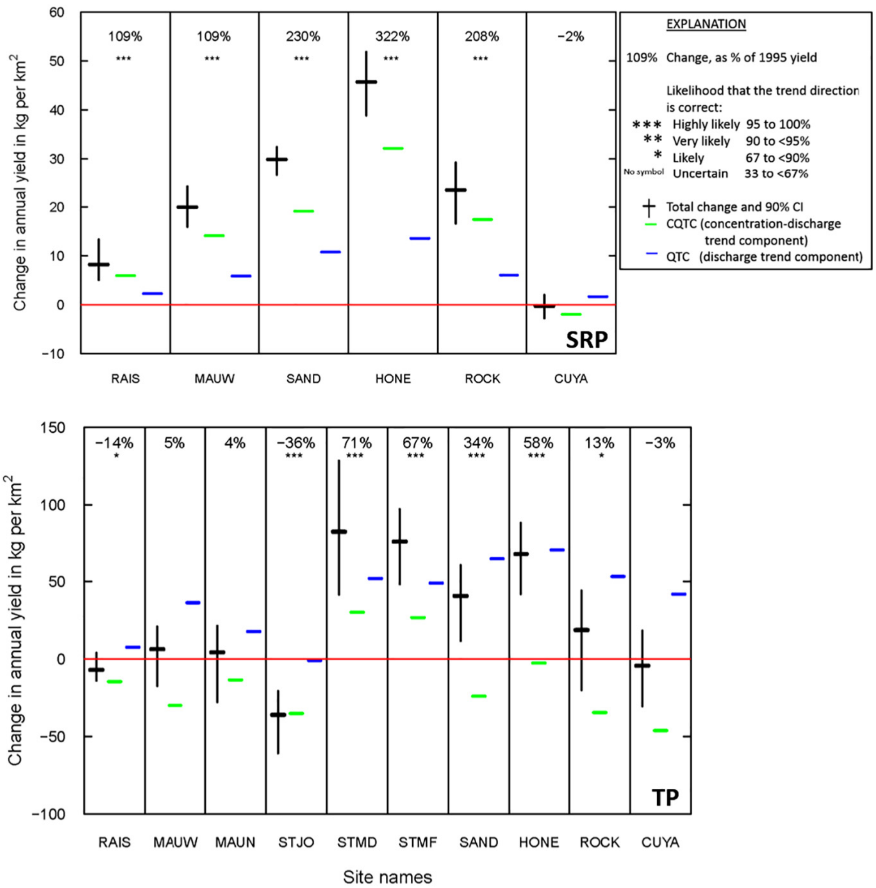

Fig. 6. Multidecadal trends (1995–2015) in annual yields of soluble reactive phosphorus (upper panel) and total phosphorus (lower panel) at each of the study sites, including confidence intervals and trend components. The trends were determined using WRTDS and the method of generalized flow normalization. Asterisk symbols indicate the likelihood that the estimated trend direction is correct. Top panel: SRP, soluble reactive phosphorus, as P; bottom panel: TP, total phosphorus, as P.

limit PP yield across the region are having some success, but the largely positive trends in SRP are canceling out those improvements and as a consequence, except in the RAIS basin where estimated PP decrease (−15 kg/km^2) exceeds the SRP increase (8 kg/km^2), TP is tending to increase or hold steady across the region.

The 1995–2015 trends for the N parameters (NO23, TKN, and TN) were also mixed and broadly speaking not very different from the TP results, but with generally smaller percentage change (Fig. 7). Expressed in percentage change, the maximum changes across the three N parameters at all 10 sites were generally less than +30% with several in the range of 15 to 20%. The largest increases for N parameters were for TKN (+38–56%) in the St. Marys basin; STMD was the only site showing a clear increasing CQTC for TKN.

If we focus on TN as the indicator of overall N changes, we see the QTC is positive at all sites except STJO where it is virtually zero. In almost all cases, the CQTC changes were in opposition and nearly equal to the QTC changes, meaning that the influence of increasing discharge is counterbalanced by a general decrease in concentration for any given discharge, and indicating dilution conditions, later discussed. The net result is that TN changes at the ten sites range from about −13% (RAIS) to +28% (STMF), and in 8 of the 10 cases the sign of the change is not highly certain. Of the WLEB sites, only 1 site shows TN trends in the range of very likely to highly likely, which is the positive trend at STMF. The negligible to small TN trends at RAIS, MAUW, SAND, HONE, and ROCK reflect dilution effects. An overall increase in TN, driven primarily by increased discharge, applies to the two St. Marys sites.

Results for NO23 (Fig. 7) are very similar to those for TN as might be expected given that NO23 constitutes a large fraction of the TN yield. The NO23 results show somewhat less uncertainty than TN, which is not surprising given the variability inherent in the suspended fraction of N transport. In all cases except for MAUN and STJO, the (+)QTC and (−)CQTC components were in opposition to each other. The largest percentage changes in NO23 yields were −13 to −15% at RAIS and ROCK, respectively, and + 20 to +29% at MAUN, STMD, and STMF.

TKN results (Fig. 7) were somewhat different from TN or NO23, with 5 of the 10 sites showing strong evidence of a trend in yield. There were highly likely increases at STMD, STMF, HONE, SAND, and CUYA (all in the range of 17% to 56%). Both STJO and RAIS show minimal QTC change; across 6 of the 8 remaining sites there was a tendency for the TKN CQTC decreases to be in opposition to the strong QTC increases.

During the 2005–2015 period, the majority of WLEB sites show very likely decreasing TP and TN trends, although the upper St. Marys basin (STMD) shows increases in TP and TN (by +15 to +16%) (ESM Appendix S5; ESM Fig. S5-1). Highly likely decreases in TN (−22 to −24%) occurred in RAIS, SAND, and ROCK; and all 3 sites in the Sandusky River basin show negative TN trends (ESM Fig. S5-1 and ESM Table S5-1).

Seasonal trends in dissolved phosphorus

Given that variability in seasonal fluxes over time can be strongly influenced by random, year-to-year shifts in the timing of high-flow events, the WRTDS method can be particularly helpful in seasonal trend evaluation, by providing a time-series of flow-normalized seasonal fluxes that are more indicative of the average seasonal pattern of yields. Among the six sites with SRP records (Fig. 8), the monthly trends for five WLEB sites (HONE, ROCK, SAND, MAUW, RAIS) all show relatively consistent patterns of monthly increases in yield between 1995 and 2015, with all months showing some increase. The largest increases at these sites occurred during December through March, with typically smaller February trends and moderate increases in the other months,

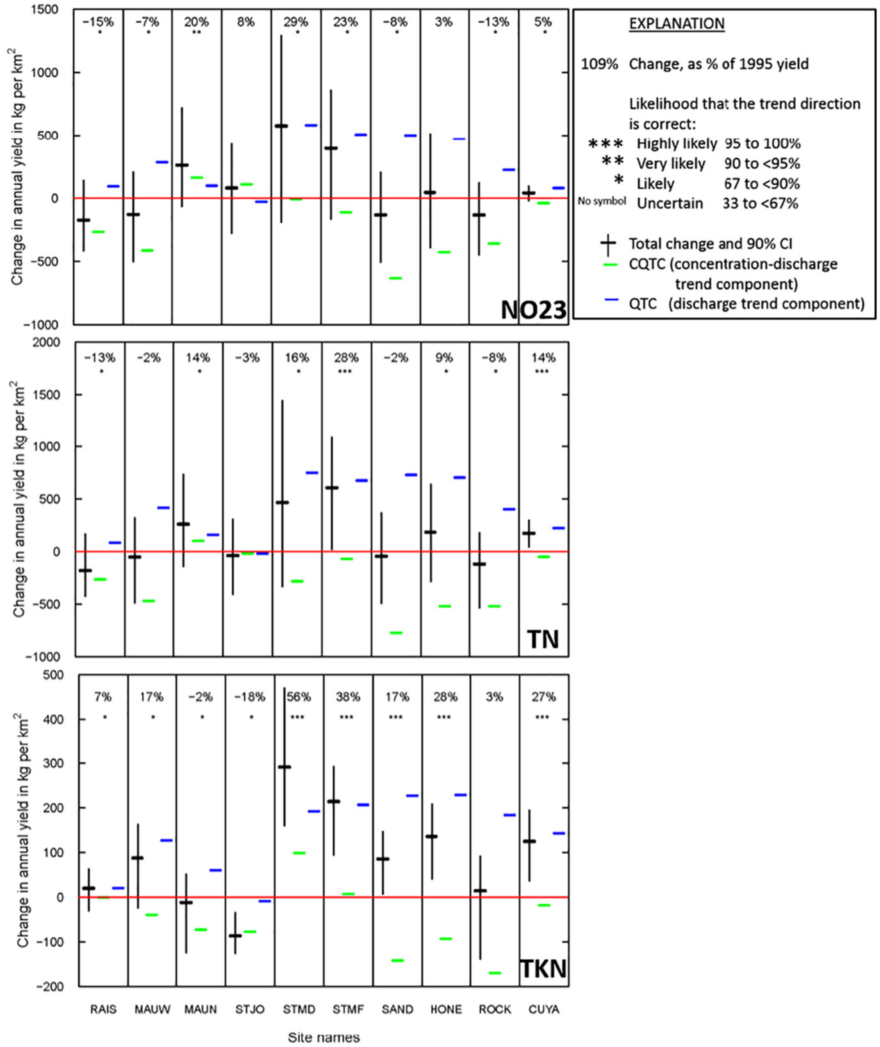

Fig. 7. Multidecadal trends (1995–2015) in annual yields of nitrate plus nitrite (upper panel), total nitrogen (center panel), and total Kjeldahl nitrogen (lower panel) at each of the study sites, including confidence intervals and trend components. The trends were determined using WRTDS and the method of generalized flow normalization. Asterisk symbols indicate the likelihood that the estimated trend direction is correct. NO23, nitrate plus nitrite, as N; TN, total nitrogen, as N; and TKN, total Kjeldahl nitrogen, as N. Site STMD compares years 1999 and 2015 for TKN and TN, based on available data.

except for August and September which show minimal changes at the four sites with greatest agricultural land use (HONE, ROCK, SAND, MAUW). The less extensive row-crop area in the RAIS and CUYA basins (Table 1) coincides with smaller monthly trends compared to the other sites shown in Fig. 8.

This seasonal pattern of the 1995–2015 SRP increases is consistent with high flow increases at these sites during these months, shown in the Quantile-Kendall monthly analysis (ESM Appendix S4), indicating that flow increases contribute to the SRP increases during these months, given the positive correlations between SRP and discharge at the WLEB sites. In the Sandusky River basin, the SRP increases at SAND, HONE, and ROCK extend into a longer period, including early summer and fall. The fact that the abrupt increases in monthly SRP flux trends starting with December coincide with winter increases in high flows at all of these sites (as much as 6 to 10% per year; ESM Appendix S4), also supports flow increases as contributing to these SRP increases, although other factors may also be contributing.

Changes in nutrient composition and ratios

Changes in nutrient composition (fractions by chemical species) and nutrient ratios (molar) across the study region during the 1995–2015 period were determined based on the 1995 and 2015 annual FNG fluxes. Among the 5 WLEB sites having both SRP and TP records, the ratio of SRP yield to TP yield between 1995 and 2015 increased dramatically, from an average of 0.12 to an average of 0.29, showing increases of nearly 2 to 3 times the 1995 ratios at these sites (Fig. 9 and ESM Table S2). The increase in the SRP fraction is evident even where 1995–2015 TP yields remained fairly constant (RAIS and MAUW); the SRP fraction remained unchanged only at the CUYA site. In contrast to phosphorus where the PP fraction dominates P fluxes, NO23, which occurs primarily in dissolved form, dominates the N fluxes representing on average 73% of TN flux among the WLEB sites during 1995 and 2015 (Fig. 9 and ESM Table S2). Seven of the 10 study sites show 1995 to 2015 reductions in NO23:TN ratios (ESM Table S2).

The 1995–2015 percentage change was considerably larger in NO23:SRP (decreasing) compared to TN:TP ratios, which show varying changes among the study sites (ESM Fig. S2; ESM Table S1). In comparison, these ratios at the non-agricultural site CUYA show little change, with a minimal increase in both ratios. The average 1995 and 2015 NO23:SRP ratios for the 5 WLEB sites with SRP records decreased from 256 in 1995 to 86 in 2015, and from 41 to 35 for TN:TP. For TN:TP, among all 10 study sites the largest 1995–2015 declines are in the St. Marys and Sandusky basins (STMF, STMD, SAND, and HONE) ranging from −24 to −32% change; and only 1 site (STJO) shows a notable TN:TP increase (+53%) reflecting the decrease in TP fluxes at this site

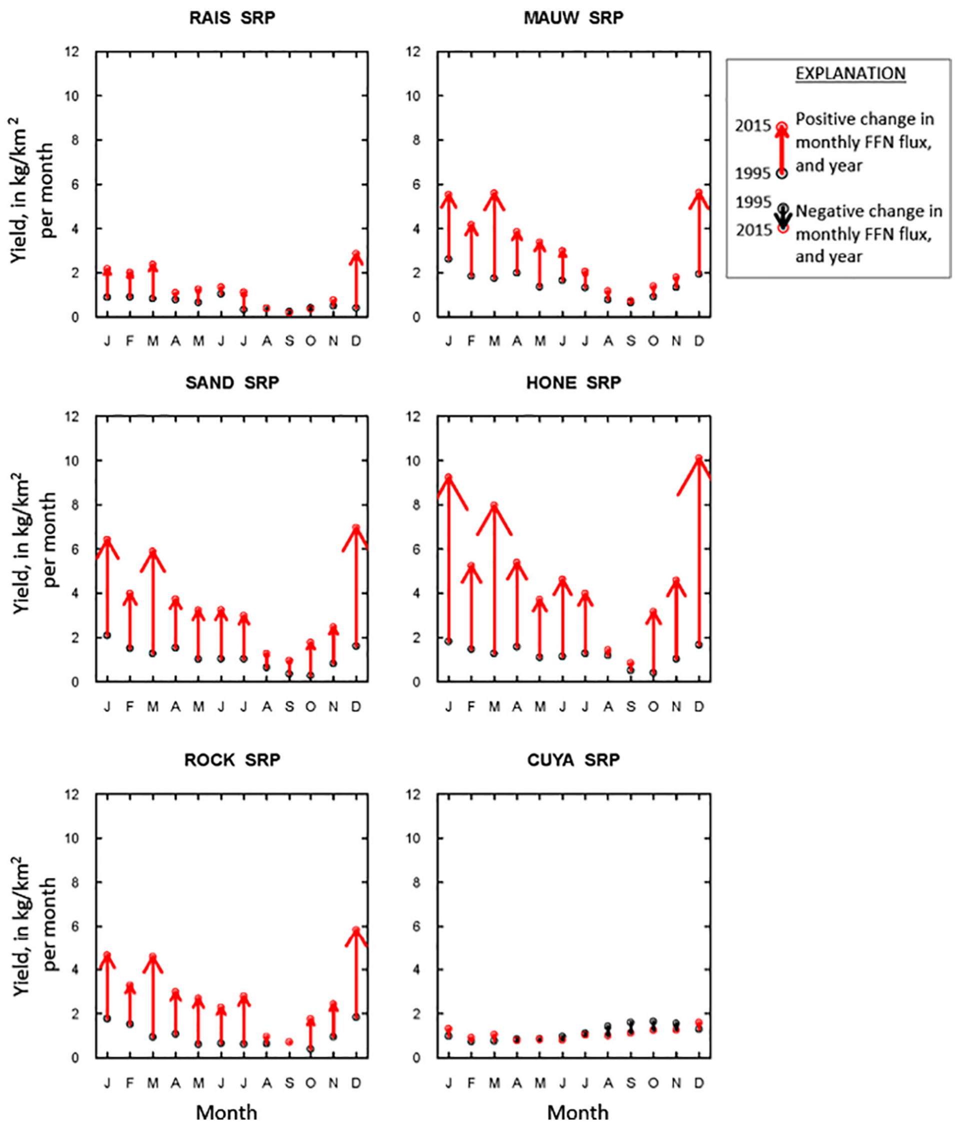

Fig. 8. Changes in monthly soluble reactive phosphorus (SRP) fluxes, as yield, between water years 1995 and 2015, using generalized flow normalization, at the six monitoring sites with long-term SRP records. The black circles indicate the monthly yield in 1995 and the red circles indicate the monthly yield in 2015. The red arrows indicate the magnitude of the increases, and the size of the arrowhead is proportional to the size of the increase. The black arrows indicate the magnitude of the decreases.

(ESM Table S1). Focusing on the 3 sites located at points of discharge to western Lake Erie (RAIS, MAUW, and SAND), the 1995–2015 changes in NO23:SRP and TN:TP flux ratios averaged −62 and −11%, respectively.

Spatial patterns in nutrient trends

The influence of row-crop agriculture on SRP changes in the WLEB is clearly evident in the 1995–2015 SRP trends and the contrast between the 5 WLEB sites' highly significant increasing trends (190 to 322%), with consistently positive CQTCs, and the CUYA site's lack of significant trend (Fig. 6 and ESM Fig. S1). These SRP trends (kg/km^2) show strong correlations with percent of basin in row crops among these 6 sites (Spearman rho = 0.99, 2-sided $p < 0.001$; Pearson r = 0.86, 2-sided $p = 0.03$). The influence of row-crop extent on trends was less clear for TP and the N-parameters.

Percentage change in SRP fluxes for the 1995–2015 period was 2 to 3 times greater at the 3 study sites in the Sandusky basin compared to the RAIS and lower Maumee River site (MAUW), and among these 5 SRP sites, the TP increases were also largest at the 3 Sandusky basin sites (Fig. 6). The largest TP changes among all study sites occurred at the 2 St. Marys basin sites (Fig. 6 and ESM Fig. S3), where SRP changes are unknown due to lack of long-term SRP records for the 4 upper Maumee basin sites.

The nutrient trends for all 4 parameters available at the two St. Marys sites show consistent increases and larger percent increases compared to all other study sites, as well as the largest annual TP and TN yields among these sites in 2015. Determining the sources of these nutrient flux patterns in the St. Marys basin will require additional study, but could be related in part to the estimated increases in livestock agriculture and manure in this basin relative to the other study sites (ESM Appendix S3). Estimates for the St. Marys basin (Falcone, 2017a, b, in ESM Appendix S3) indicate that swine doubled and poultry nearly tripled during 1982–2012, and that manure is a higher portion of agricultural P applied, 58 to 61% of fertilizer plus manure compared to about 20 to 30% for the other study sites during 2012. Ohio data indicate higher average P-content and lower N:P ratios for poultry manure compared to that of swine and dairy cattle (International Joint Commission, 2018). The 1995–2015 highly significant TKN increases at the St. Marys sites, which show the largest percentage change among N parameters and the only positive TKN CQTC (at STMD), are consistent with estimated increases in livestock and manure-N in this basin (ESM Appendix S3) as possible sources of ammonia and organic-N (components of TKN).

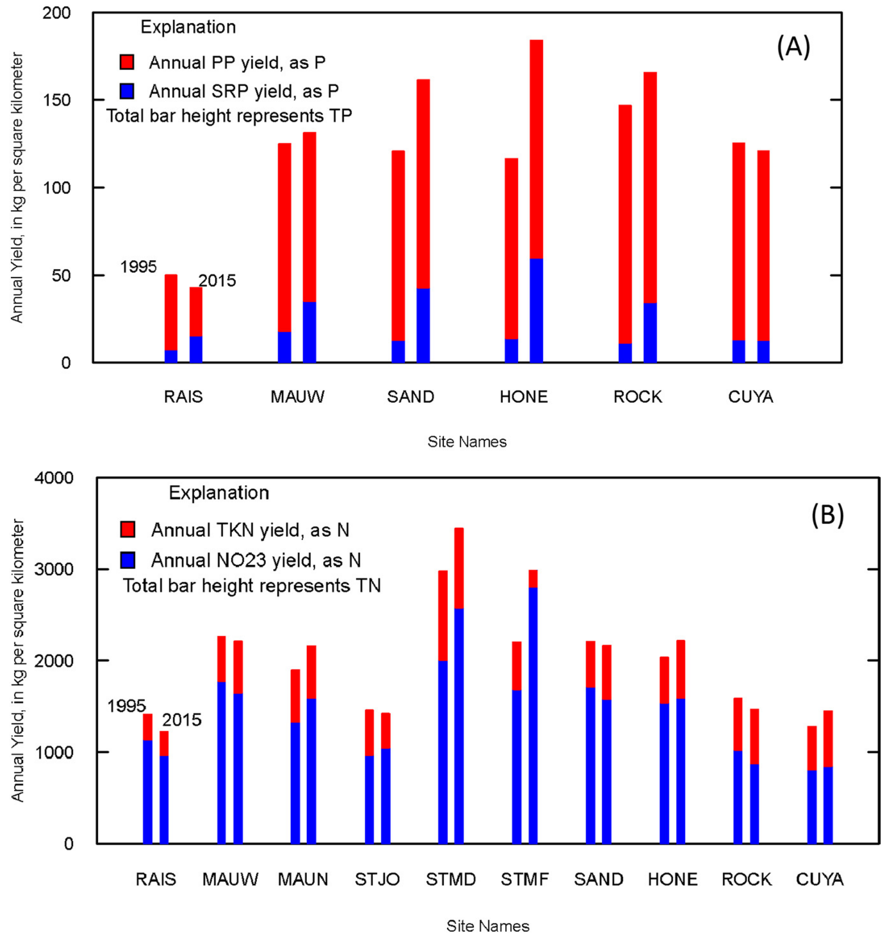

Fig. 9. Comparison of 1995 and 2015 fractions of (A) total phosphorus and (B) total nitrogen yields at the ten trend sites, determined using the method of generalized flow normalization. For site STMD, the years 1999 and 2015 are compared for TKN and TN, based on available data. NO23, nitrate plus nitrite, as N; PP, particulate phosphorus, as P; SRP, soluble reactive phosphorus, as P; TKN, total Kjeldahl nitrogen, as N; TN, total nitrogen, as N; TP, total phosphorus, as P.

The St. Joseph and St. Marys watersheds show highly significant opposing trends for TP and TN as well as strong differences in 2015 TP and TN yields (ESM Table S1); the combined influence of these 2 major tributaries are reflected in yield and trend patterns seen downstream at the upper Maumee River site (MAUN). In this case, the results for MAUN are not indicative of nutrient transport characteristics uniformly applicable to the entire upstream watershed region, but rather this site is an integrator of differing upstream conditions and nutrient inputs. Lower SRP and TP yields at RAIS and STJO, also the only sites with negative TP trends, likely relate to a combination of influences (see "Discussion") in addition to the smaller proportion of watershed in row crops in these basins. These two watersheds along with the Rock Creek and Cuyahoga River watersheds also show the lowest fluxes for NO23 and TN (Fig. 9). Possible factors related to spatial differences in fluxes and flux changes among these sites are further discussed below.

Discussion

The methodology presented herein provides an approach to statistically partition the effects of long-term stream discharge trends on water-quality trends in order to better discern land-use and climatic influences on water-quality changes. The method identifies trend components related to changes in discharge versus changes in the water-quality concentration-discharge relationship over time, which may provide insights into the changes taking place in the physical hydrologic and source-supply system.

Changes in discharge can result from a combination of land-use changes (e.g. related to tile drainage expansion) and climatic changes that could be related to changing concentrations of greenhouse gases or to patterns of decadal to multi-decadal climatic persistence that have existed long before human-caused increases in greenhouse gases. Given the close proximity of the WLEB watersheds, if climate change were the sole cause of changing discharge, then it is reasonable to expect that the Quantile Kendall plots of discharge trends would display very similar patterns (because they are likely to be responding to similar climatic forcing). If the driver of discharge change was more related to changes in land use (cropping patterns and land-drainage systems) that differed between watersheds, then the Quantile Kendall plots would tend to show inconsistencies in trend patterns. Although the analysis presented here does not attempt to explicitly attribute changes in discharge to land use versus climate, comparing the results across multiple contrasting watersheds in a given region can provide insight into their influences.

The Quantile-Kendall plots for the WLEB sites displayed some regionally consistent patterns but also some strong variations across the sites, suggesting that both climate and land-use/land-management changes may be contributing to some of the stream discharge trends. Annual trends for high flows (>90th percentile) show consistent increasing trends for all of the sites except STJO. In contrast, the annual trends in the low- to medium-flow range show variability among sites, which suggests land-use/land-management influences. The causes of the strong increasing flow trends (annual and monthly) across the range of low to high flows at the three Sandusky basin sites (SAND, HONE, and ROCK) are unclear, but may reflect expansion of tile drainage. Alternatively, some of these increased flows could be related to reduced evapotranspiration and increased runoff potential related to the 55–60% reduction in winter wheat production (serving as a cover crop) in these watersheds (ESM Appendix S3). Additional study would be required to confirm causes. The consistency displayed among all of the study sites in flow increases during December (high flows, as much as 6–10% per year) and January (low to medium flows, 2–8% per year) (ESM Appendix S4) suggests that winter climatic changes are contributing to these increases. The interpretation of such plots at multiple sites, in conjunction with climate data and hydrologic models is a topic in need of additional research.

Increases in discharge in recent decades are clearly contributing to the trend patterns in these study watersheds and include some regionally consistent seasonal increases for specific ranges of the flow distribution. The high-flow increases during fall-winter months across these sites may be important contributors to increased annual nutrient loading – if combined with increases in fall fertilizer applications, increases in the proportion of fall-winter precipitation as rain versus snow, and increased potential for rain on frozen ground, all of which have been reported for this region (Jarvie et al., 2017; Michalak et al., 2013; Ohio Lake Erie Phosphorus Task Force, 2010, 2013; Smith et al., 2015a). In addition, increases in high flows could increase mobilization through erosion of nutrients stored in streambank or bed sediments and their transport to downstream water bodies. The factors related to 1995–2015 changes in SRP fluxes appear to be strongly related to increases in nutrient sources and transport efficiency, with increasing discharge also playing a role in the increased transport. In contrast, TP and the N-parameters show mixed trends of lesser magnitude that reflect opposing components of increasing discharge but decreasing concentrations (dilution effect) across some or all of the concentration-flow relationship.

SRP trend results for the WLEB sites indicate that land use/land management-related changes (the CQTC; concentration-discharge trend component) and increases in discharge (the QTC) both contributed to the dramatic 1995–2015 increases at the five WLEB sites; however, the CQTC changes were the primary drivers of these trends, contributing on average 70% to the total observed change. Even without the discharge increases, SRP fluxes would be increasing as a result of increased SRP delivery to streams as a direct or indirect result of changes in land use/land management. This result is consistent with the partitioning of SRP trend drivers in Jarvie et al. (2017) who applied an alternative approach to identify sources of 2002–2014 SRP load changes in the Maumee, Sandusky, and Raisin Rivers. The SRP and TP trend results indicate PP reductions at some sites, likely reflecting the success of erosion control efforts, but the SRP increases are outweighing these PP reductions, emphasizing the need for measures directly focused on limiting P inputs to, and SRP losses from, soils and on controlling sources of P from human and animal waste. These results emphasize the need for reexamination of the efficacy of PP and SRP control strategies going forward (King et al., 2015a, b; Ohio Lake Erie Phosphorus Task Force, 2010, 2013; Smith et al., 2015b, 2016).

Given the fact that the increases in the QTC were substantial at the majority of the study sites, there are two sets of conditions to consider for why the 1995–2015 percentage changes in the nitrogen parameters (NO23, TKN, TN) were so much lower than the SRP changes (Figs. 6–7; ESM Table S1). One is simply a dilution condition, namely that there is annually only a finite amount of N available for transport from each watershed. This is the sum of N applied, atmospheric N fixation, and N carried over year-to-year in the soils and in the groundwater flow system minus N removed due to crop harvest and N losses to the atmosphere through denitrification. Given a fixed supply of N combined with increasing amounts of water moving through the system, it means that there is little change in the annual availability of N but, at the same time, discharge has been increasing, resulting in a dilution of N concentrations and a decrease in the CQTC.

The alternative condition is that waste management and agricultural practices are altering the available supply of N in the system, and this change in N supply is leading to changes in the concentration-discharge relationship (CQTC). The dilution condition dominated the TN trends among these study sites, although there was evidence of both conditions (dilution plus changes in supply) contributing to the trends for the N parameters at a few sites, particularly in the St. Marys basin. The product of this dilution condition or combined dilution plus supply-change condition indicates that transport of TN has been relatively fixed, with minimal changes over the last two decades. The TN flux trends in this system are most influenced by changes in NO23 fluxes, which generally exhibited small changes similar to those in TN; highly likely increases (or decreases) in TKN were also observed at a few sites.

Between 1995 and 2015, TN:TP flux ratios (molar) show small declines at most of the study sites, but reductions in ratios of the most bioavailable species NO23:SRP were quite dramatic, and on average among the WLEB study sites in 1995 were triple that of 2015. These changes in proportions of bioavailable nutrients in these tributaries may have implications for algal community structure and cyanobacteria toxicity (Chaffin and Bridgeman, 2014; Chaffin et al., 2014; Dodds and Smith, 2016; Gobler et al., 2016), as well as the nutrient composition of fluxes delivered to Lake Erie.

Regarding spatial differences in the nutrient flux trends among these sites, the higher nutrient yields and stronger trends for both P and N parameters in the St. Marys basin suggest the potential influence of estimated larger livestock numbers and increases in livestock and manure in recent decades (Falcone, 2017a, b; International Joint Commission, 2018; as discussed in ESM Appendix S3). Fanelli et al. (2018) note that manure appears to be a major driver of orthophosphate in regional analyses of agricultural watersheds of the Chesapeake Bay, based on strong relations between manure inputs and dissolved orthophosphate concentrations. Additional research is needed to determine current and anticipated future nutrient loading from manure sources, as well as instream storage and transport of total and dissolved phosphorus in the upper Maumee basin (St. Marys River) to downstream river reaches and Lake Erie.

The 1992–2012 changes in production by crop types in some of these watersheds included increases in soybeans (by 30 to 45%) at most of the sites, and a 50–60% reduction in winter wheat with a 20–25% increase in corn at the three sites in the Sandusky basin (ESM Appendix S3). Given differences in tillage, fertilizer needs (ESM Table S3-4), crop nutrient content, and growing seasons that are associated with each of these crop types, such crop changes may be influencing seasonal and annual nutrient and discharge trends.

The lower SRP and TP yields at RAIS and STJO, which also are the only sites with negative 1995–2015 TP trends (Fig. 6; ESM Table S1), may be related to several factors. Relative to the other WLEB trend sites, these basins are characterized by a smaller extent of row crops and a larger proportion in pastureland (Table 1), and higher soil drainage rates requiring less extensive tile-drainage (Betanzo et al., 2015b; Jarvie et al., 2017; Kalcic et al., 2016). The RAIS basin shows the largest estimated 1994–2012 fertilizer-P reduction (−47%) among the WLEB study sites (ESM Appendix S3), and a number of nutrient-reduction efforts have been documented in both of these basins (Michigan Department of Agriculture and Rural Development, 2012, 2016; St. Joseph River Watershed Initiative, 2011, and U.S. Department of Agriculture, 2016, 2017). Reservoir storage, which is larger in these two watersheds and can influence nutrient dynamics including reduction of downstream phosphorus transport (Bosch et al., 2009; Bosch, 2008; Walker et al., 2007), and(or) flow diversions in these two watersheds (ESM Appendix S3) could also contribute to the lower phosphorus yields. Jarvie et al. (2017) also suggest that smaller recent (2002–2014) increases in SRP in the Raisin basin compared to Maumee and Sandusky basins relate to the larger extent and likely increases in tile drainage in the Maumee and Sandusky basins.

The contrasts in trend results between the St. Joseph and St. Marys watersheds, which show highly significant opposing trends for TP and

TN as well as large differences in nutrient yields, illustrate the importance of strategic spatial coverage in trend network design to help delineate trend patterns and drivers which may vary in upstream areas and may not be apparent if monitoring, or analysis, is focused only on downstream reaches near discharge points into water bodies at risk. Evaluation of the effectiveness of TP control strategies will benefit particularly with examination of the contrast between changes in land use and land management in the St. Joseph River watershed (where substantial yield reductions have taken place) versus the St. Marys River and Honey Creek watersheds (where the largest increases have occurred).

Evaluation of the relative effects on streamflow from climate-driven versus land-use/land-management changes is an important research challenge not addressed in this paper, but crucial to management of nutrient delivery in the future. As stated in Lins et al. (2010), understanding the effects of climatic variability and change on water resources will require preservation of the continuity of long-term water data collection and analysis and interpretation of these data to determine how water resources are changing. Potential considerations for future research include linking climate or hydrologic models to these trend methods to improve the ability to attribute or further parse the QTC and CQTC changes to specific climatic and land-use drivers.

Conclusions

Herein we describe and utilize new analytical tools to help identify streamflow trends and to evaluate and understand multi-decadal water-quality trends in regions influenced by nonstationary streamflow. We applied these methods to 10 long-term monitoring sites located primarily in the western Lake Erie drainage basin, focusing on multidecadal changes (1990s to 2015) in stream discharge and in flow-normalized nutrient fluxes (SRP, TP, NO23, TKN, and TN). Regionally consistent discharge changes at these sites, likely driven by climatic changes, include increases in high flows (the upper quartile of the daily discharge distribution), important from the perspective of nutrient loading, and seasonal high-flow increases most notable during November, December, and March. Yet the presence of strong differences in flow trends among some of these watersheds, in relative close proximity, suggests that changes in land and water management play a substantial role in driving some of the discharge trends.

The largest and most pervasive 1995–2015 flux changes occurred for the parameter SRP, which showed increases in yield ranging from 109 to 322% among the sites in the predominantly agricultural WLEB. In contrast, the nearby CUYA site with only 9% of the watershed in row crops showed no discernable SRP flux change. The SRP flux trends can partly be attributed to the (+)QTC change in runoff characteristics (changes in the amount and timing of water moving through the hydrologic system and a function of both climate change and land-use change); however, the (+)CQTC is the dominant part of the (+)FNG trends, indicating that factors such as the availability of SRP in the soil and groundwater system, and its mobility have been the primary drivers of flux change.

The largest SRP flux (as yield) increases typically occurred during December through March with moderate increases during April–July and October–November, indicating winter and early spring may be important for prioritizing SRP-reduction actions. At the majority of the sites, TP and the N parameters showed "opposing" CQTC and QTC influences, with predominantly negative CQTC contributions and nearly universally positive QTC contributions to flux trends, the total trend being the net change (or sum) of these opposing influences. The result is that most of these trends are generally of small magnitude; had the source supply of these nutrients not been limited, these TP, TN, NO23, and TKN flux trends would likely have been much larger increases as a consequence of the increasing flows in this region. Additional research into land-use/land-management changes and nutrient sources in the St. Marys River basin is needed to identify causes for the TP trends that show positive CQTC influences, larger TP flux increases, and larger TN and TP yields in 2015 for the two St. Marys River sites relative to other WLEB study sites.

Understanding past changes in stream water quality and predicting change into the future must be done in full recognition of the factors that influence both water quality and water availability; this includes changes in water use, land-use practices, the design and operation of water infrastructure, changes in land drainage systems, alterations of groundwater levels and quality, and climate change. Here we offer tools to address a central challenge that researchers confront in evaluating nutrient flux changes in the Lake Erie watershed and other agricultural regions; that is, effects on nutrient trend patterns from complex interactions among climatic changes and land-use/land-management changes, such as tile drainage expansion and changes in tillage practices, all of which can influence streamflow trends over time.

The methods presented here center on flow-normalized nutrient fluxes, designed to be independent or more nearly independent of streamflow variability, in order to increase the trend signal-to-noise ratio and reduce risks of identifying flux trends biased by random interannual variations in streamflow. The ultimate goals of this approach are to provide estimates of stream nutrient transport, independent of flow fluctuations, as indicators of the influence of changes in land use and of nutrient-reduction activities and to provide information to help separate out effects of land-use activities from longer-term climate-related influences that also can be important nutrient-trend drivers and may become increasingly important in the future. The types of analyses demonstrated here, that explicitly address influences of streamflow effects and changes on water-quality trends, are critical to evaluating land-change effects on nutrient loading across the WLEB, or more broadly the Great Lakes region. Applied as periodic reassessments, the results of these analyses can serve as a "dashboard" for parties interested in or responsible for Great Lakes water quality to more clearly discern the influences of past and current policies and more effectively guide adaptive management decisions.

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Electronic supplementary material (ESM). Supplementary data

Supplementary data designated in this article as ESM (ESM appendices, figures, and tables) can be found online at https://doi.org/10.1016/j.jglr.2018.11.012. Also, Choquette et al. (2018) present additional electronic-format data including detailed site information, model input, and trend results.

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