Research

Regional Nutrient Trends in Streams and Rivers of the United States, 1993–2003

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Received December 23, 2008. Revised manuscript received March 03, 2009. Accepted March 05, 2009.

Trends in flow-adjusted concentrations (indicators of anthropogenic changes) and observed concentrations (indicators of natural and anthropogenic changes) of total phosphorus and total nitrogen from 1993 to 2003 were evaluated in the eastern, central, and western United States by adapting the Regional Kendall trend test to account for seasonality and spatial correlation. The only significant regional trend was an increase in flow-adjusted concentrations of total phosphorus in the central United States, which corresponded to increases in phosphorus inputs from fertilizer in the region, particularly west of the Mississippi River. A similar upward regional trend in observed total phosphorus concentrations in the central United States was not found, likely because precipitation and runoff decreased during drought conditions in the region, offsetting the increased source loading on the land surface. A greater number of regional trends would have been significant if spatial correlation had been disregarded, indicating the importance of spatial correlation modifications in regional trend assessments when sites are not spatially independent.

Introduction

Nutrients are essential for plant and animal life, but in high concentrations they can act as contaminants in water. Overenrichment of streams and rivers with nutrients contributes to the formation of algal blooms, which can lead to disruption of recreational activities (*1*), taste and odor problems in drinking water supplies (*2*), creation of carcinogenic trihalomethanes during drinking water treatment (*3, 4*), production of algal toxins harmful to animal and human health (*5–7*), hypoxic conditions that contribute to the release of metals from streambed sediments (*8*) and fish kills (*9*), and economic losses in property values and recreational use (*10*).

Understanding how and why nutrient concentrations are changing over time in streams and rivers is essential for effectively managing and protecting these water resources. Natural changes in precipitation and streamflow and anthropogenic changes in nutrient sources (such as fertilizer application) or transport to streams (such as implementation of management practices like conservation tillage) can influence nutrient concentrations in streams (*11*). Natural and anthropogenic changes often occur simultaneously, and they may counteract or supplement one another. For example, nutrient concentrations in a stream may change very little if a decrease in concentration resulting from upgrades at a wastewater treatment plant is offset by decreased in-stream dilution resulting from a natural decrease in precipitation and streamflow.

Two types of concentration trends can help discriminate the effects of natural and anthropogenic changes on nutrient concentrations in streams. In the analysis of trends in flow-adjusted concentrations, the effects of changes in streamflow on concentration are removed. Because changes in streamflow are tied to natural changes in precipitation and evapotranspiration, flow adjustment of nutrient concentrations allows trends caused by other, largely anthropogenic, changes to be more directly assessed. In some streams, flow adjustment may not account for all natural influences on nutrient concentrations and/or may account for anthropogenic influences on streamflow (such as irrigation or reservoir storage); nonetheless, the adjustment process will provide a more direct assessment of the effects of anthropogenic changes in nutrient sources and transport. In the analysis of trends in observed nonflow-adjusted concentrations, no adjustments are made for any natural or anthropogenic influence. Therefore, the net effects of all simultaneous influences on concentration are evaluated, allowing for the assessment of nutrient concentrations in streams relative to water quality standards and the condition of aquatic communities. A comparison of trends in flow-adjusted and observed concentrations can provide insight into the effects of changes in streamflow and climate on in-stream nutrient concentrations. Our primary objective was to evaluate and compare spatial patterns in trends of flow-adjusted and observed concentrations of total nitrogen and total phosphorus in major geographic regions of the United States between 1993 and 2003.

The presence of spatial correlation of nutrient records can complicate such evaluations of regional trend patterns. Large- and small-scale physical factors such as climate, geology, land use, and vegetation have the potential to induce similar patterns in nutrient records at two or more stream locations. If the distance between these locations is smaller than the areal extent of influential physical factors, some degree of spatial correlation may be present (*12*). When spatial correlation is present, the assumption of independence required by most trend tests can be violated (*12*). The effect is often a more liberal hypothesis test, where the null hypothesis of no regional trend will tend to be more frequently rejected than it should (*13*). Our secondary objective was to examine the effects of spatial correlation on the regional trend results.

Methods

Nutrient data publicly accessible through the online U.S. Geological Survey National Water Information System (NWIS) database at http://waterdata.usgs.gov/usa/nwis/qw initially were surveyed for this study. Additional data from the online U.S. Environmental Protection Agency STORET database at http://www.epa.gov/storet/dbtop.html also were surveyed in some areas. Sites were selected for analysis of trends between 1993 and 2003 on the basis of the following minimum criteria (*14*): nutrient data record beginning in water year

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10.1021/es803664x

Published on Web 04/07/2009

FIGURE 1. Study sites for (A) total phosphorus and (B) total nitrogen.

1993 or earlier and ending in water year 2003 or later, approximately quarterly sampling each year, contemporaneous record of daily mean discharge at that site or a nearby representative site, data gaps no longer than two years and only during the middle six years of record, and representative coverage of samples over the hydrograph and across seasons to avoid bias toward particular streamflows or seasons. In addition, sites with a data record consisting of two time periods separated by a gap longer than two years were included for step trend evaluation if data in the two periods met the remaining minimum criteria. The final 171 sites for total phosphorus and 137 sites for total nitrogen are shown in Figure 1.

The 1993−2003 time frame was selected to capitalize on increased nutrient monitoring by the U.S. Geological Survey through its National Water Quality Assessment program during this period. Trends from 1993 to 2003, however, will not reflect the widespread increase in stream nutrient concentrations that occurred over the last century (*15*), when nutrient inputs to streams from fertilizer, industrial emissions, and other anthropogenic sources increased substantially (*16, 17*). Instead, trends from 1993 to 2003 will reflect more recent changes, including management strategies designed to reverse or reduce the pace of earlier increases. Because interannual variability in stream concentrations can be large, and because watershed conditions often change slowly, the 11 year period examined may be too short in some cases to show a statistically significant trend.

Regional trends in flow-adjusted and observed concentrations were examined in the eastern, central, and western United States (Figure 1), using what we have termed the Regional Seasonal Kendall test. The Regional Seasonal Kendall test is an extension of the Regional Kendall test (*18*) and the Seasonal Kendall test (*19*), both of which are based on the simpler Mann−Kendall test.

In the Mann−Kendall test, the test statistic S is calculated for a site by comparing each of n (t, y) data pairs, where t is time and y is the trend variable of interest, to all other (t, y) data pairs in a pairwise fashion, and subtracting the number of data pairs where y decreases as t increases (M) from the number of data pairs where y increases as t increases (P), with ties not counted (*20*).

$$S = P - M \tag{1}$$

S is asymptotically normal, with mean of 0 and variance of

$$\mathrm{Var}(S) = \frac{n(n-1)(2n+5)}{18} \tag{2}$$

If there are ties, an adjustment is made to the computation of variance (*20*).

The Seasonal Kendall test accounts for seasonality by calculating S within each of m seasons separately (*19*).

$$S_{\mathrm{K}} = \sum_{S=1}^{m} S_S \tag{3}$$

where S_K is the Seasonal Kendall test statistic, m is the number of seasons, and S_S is the Mann−Kendall test statistic for each of the $S = 1$ to m seasons. S_K is asymptotically normal with mean of 0 and variance of

$$\mathrm{Var}(S_K) = \sum_{S=1}^{m} \mathrm{Var}(S_S) + 2\sum_{S=1}^{m-1} \sum_{Q=1+S}^{m} \mathrm{Cov}(S_S, S_Q) \tag{4}$$

where $\mathrm{Var}(S_S)$ is the variance of S_S and $\mathrm{Cov}(S_S,S_Q)$ is the covariance between seasons S and Q. The covariance is computed as described in ref *21*, building on work presented in ref *22*, to account for serial dependence. The Sen slope, an estimate of the trend magnitude, may be computed as the median of all slopes between data observations in the same season (*19, 23*).

The Regional Kendall test substitutes location for season in the Seasonal Kendall test and computes the Mann−Kendall test statistic, S_L, for each of n locations (*18*). The overall regional test statistic, S_{R}, and its variance can be computed following eq 3 and eq 4, respectively, with S_L substituted for S_S. Calculating the variance following eq 4 accounts for spatial correlation among the sites (*13*).

The Regional Seasonal Kendall test incorporates features of both the Seasonal Kendall test and the Regional Kendall test. The overall regional and seasonal test statistic, S_{T}, is the sum of the individual Mann−Kendall test statistics, S_{SL}, from each of the m seasons at each of the n locations.

$$S_{\mathrm{T}} = \sum_{S=1}^{m} \sum_{L=1}^{n} S_{SL} \tag{5}$$

The variance of S_{T}, $\mathrm{Var}(S_{\mathrm{T}})$, is computed from the covariance between each season at each site, as with the Seasonal Kendall test, and between each site for each season.

$$\mathrm{Var}(S_{\mathrm{T}}) = \sum_{S=1}^{m} \sum_{L=1}^{n} \mathrm{Var}(S_{SL}) + 2\sum_{S=1}^{m-1} \sum_{Q=1+S}^{m} \sum_{L=1}^{n} \mathrm{Cov}(S_{SL}, S_{QL}) + 2\sum_{S=1}^{m} \sum_{L=1}^{n-1} \sum_{K=1+L}^{n} \mathrm{Cov}(S_{SL}, S_{SK}) \tag{6}$$

where $\mathrm{Var}(S_{SL})$ is the variance of S_{SL}, $\mathrm{Cov}(S_{SL},S_{QL})$ is the covariance between seasons S and Q at location L, and $\mathrm{Cov}(S_{SL},S_{SK})$ is the covariance between locations L and K in season S

$$\mathrm{Cov}(S_{SL}, S_{SK}) = \frac{K_{S,LK}}{3} + \frac{(n^3 - n)r_{S,LK}}{9} \tag{7}$$

where

$$K_{S,LK} = \sum_{i<j} \mathrm{sgn}[(x_{SKj} - x_{SKi})(x_{SLj} - x_{SLi})] \quad (8)$$

$$r_{S,LK} = \frac{3}{(n^3 - n)} \sum_{i,j,h} \mathrm{sgn}[(x_{SKj} - x_{SKi})(x_{SLj} - x_{SLh})] \quad (9)$$

and x_{SKj} is the observed value in season S at location K for year j. If there are no ties and no missing values, then $r_{S,LK}$ is Spearman's correlation coefficient for locations L and K in season S. The computation of $r_{S,LK}$ does not change if there are ties or missing values, but it is not equivalent to Spearman's correlation coefficient in that case. The remaining covariance is assumed to be 0.

Var(S_T) can be used to compute the test statistic Z_T and p-value of the normal approximation for the Regional Seasonal Kendall test, with a continuity correction for S_T.

$$Z_T = \begin{cases} \dfrac{S_T - 1}{\sqrt{\mathrm{Var}(S_T)}} & \text{if } S_T \text{ is greater than } 0 \\ 0 & \text{if } S_T \text{ is equal to } 0 \\ \dfrac{S_T + 1}{\sqrt{\mathrm{Var}(S_T)}} & \text{if } S_T \text{ is less than } 0 \end{cases} \quad (10)$$

The inclusion of Cov(S_{SL},S_{SK}) in the calculation of Var(S_T) in eq 6 accounts for spatial correlation among sites across multiple scales. To evaluate the effects of spatial correlation on the Regional Seasonal Kendall test results, Var(S_T) was computed with and without Cov(S_{SL},S_{SK}), and the resulting p-values were compared. Although this approach accounts for spatial correlation, it cannot address the limitations of sparse coverage and the possible extrapolation of trend results to unsampled areas, which may have occurred in this study (Figure 1).

The null hypothesis of the Regional Seasonal Kendall test is that there is no regional trend. Rejection of the null hypothesis will occur when a change in the same direction occurs at many of the individual locations, even if the trend at each individual location is not always significant on its own (*18*). Failure to reject the null hypothesis may result from a lack of trend at most locations or from trends in opposite directions at different locations canceling one another out (*18*). Regional trend results were considered significant if the p-value was less than or equal to 0.10.

Flow-adjusted concentrations were calculated as residuals from a lowess smooth of log-transformed concentrations on log-transformed streamflow with a span of 0.75; a maximum of 5% censoring was accepted in the lowess flow adjustment. For evaluation of trends in observed concentration, concentrations were recensored to the highest detection limit for each constituent among all sites, and a maximum of 50% censoring was accepted. This involved some loss of information and possibly loss of power to detect true trends. For both observed and flow-adjusted concentrations, the number of seasons was set to four, the highest annual sampling frequency inclusive of the most sites. When multiple observations occurred in a single season, the observation closest to the midpoint of that season was used. Sites not meeting the above requirements were excluded from further analysis.

To aid in interpreting any regional trends, temporal changes in key nitrogen and phosphorus sources were determined for each site. Nitrogen in atmospheric deposition was derived from wet deposition data from the National Atmospheric Deposition Program (*16*). Phosphorus and nitrogen inputs from fertilizer (including farm and nonfarm uses) were derived from sales and expenditures data from the Association of American Plant Food Control Officials and the U.S. Census of Agriculture (*14, 16*). Phosphorus and nitrogen inputs from manure (including confined and unconfined livestock) were derived from livestock population data from the U.S. Census of Agriculture (*16*). Population density was derived from census block groups and population counts from the U.S. Census of Population and Housing (*24, 25*). A time series for each source was generated using geographic information system (GIS) software (*26*) for a series of spatial overlay analyses of the site drainage area boundaries with digital maps of nutrient sources. With the annual data series from 1993 to 2003 for atmospheric deposition and fertilizer, the Sen slope estimate was determined for each site, normalized to the value in 1993 for that site, and multiplied by the number of years in the study period. With the periodic data series for manure (1992, 1997, and 2002) and population density (1990 and 2000), the percentage change between the first and last year was determined for each site.

When significant regional trends were detected, the strength of the correspondence between trends in nutrient concentrations and changes in nutrient sources at sites within the region was evaluated using Kendall's tau rank correlation (*20*). The goal of this analysis was not to explain all factors affecting each trend at individual sites—that was beyond the scope of this study. Rather, the goal was to identify on a broader scale individual factors potentially affecting trends at a regional grouping of sites. Other factors affecting nutrient concentrations likely were important at some or all of the sites, but consistent and comprehensive national estimates of these factors are not currently (2008) available for the study period.

Results and Discussion

Effect of Spatial Correlation. The results of the regional trend tests were substantially different with and without correction for spatial correlation. Without this correction, a greater number of the regional trends would have been significant, including flow-adjusted concentrations of total phosphorus in the eastern and western United States and total nitrogen in the central United States as well as observed concentrations of total phosphorus and total nitrogen in the western United States (Table 1). The more frequent rejection of the null hypothesis without the spatial correlation correction indicates that the use of the corrected Regional Seasonal Kendall test is appropriate in regional trend assessments where sites are not spatially independent.

Spatial covariance was greater for observed concentrations than for flow-adjusted concentrations, particularly with total phosphorus (Table 1). Broad-scale climate patterns that affect streamflow and in-stream nutrient concentrations likely contributed to spatial correlation within a region. The effects of these patterns were reduced during flow adjustment, leading to lower relative spatial covariance in flow-adjusted concentrations.

Regional Trends in Flow-Adjusted Concentration. After correction for spatial correlation, the only significant regional trend in flow-adjusted concentrations of total phosphorus occurred in the central United States; there were no significant regional trends in flow-adjusted concentrations of total nitrogen (Table 1). The number of sites with general increases in flow-adjusted concentrations was comparable to the number of sites with decreases for total nitrogen in the eastern, central, and western United States, and many of these changes resulted in trends that were nonsignificant (Figure 2A). There were more sites with general increases than decreases in flow-adjusted concentrations of total phosphorus in the eastern and central United States and more sites with general decreases than increases in the western United States. Of the sites with increases or decreases, the greatest percentage was significant for total phosphorus in the central United States (Figure 2A).

TABLE 1. Regional Seasonal Kendall Test Results for Flow-Adjusted (FA) and Observed (OBS) Concentrations of Total Phosphorus and Total Nitrogen

region	type	n	S_T	Var(S_T)	Cov(S_{SL},S_{QL})	Cov(S_{SL},S_{SK})	p-value, without correction for spatial correlation	p-value, with correction for spatial correlation
				total phosphorus				
eastern United States	FA	40	345	23816	6927	299209	0.050	0.549
	OBS	58	−37	21167	9155	629828	0.836	0.965
central United States	FA	38	1099	20607	9855	325905	<0.001	0.066
	OBS	56	297	26186	9255	628203	0.116	0.716
western United States	FA	29	−444	18150	3738	164610	0.003	0.305
	OBS	51	−493	16809	2615	491884	<0.001	0.491
				total nitrogen				
eastern United States	FA	51	29	30240	5240	544149	0.882	0.971
	OBS	56	−57	33158	10781	655919	0.789	0.947
central United States	FA	40	829	21312	5385	303090	<0.001	0.149
	OBS	46	−147	24954	5965	411648	0.406	0.826
western United States	FA	18	−162	10777	4637	66762	0.195	0.574
	OBS	31	−616	16542	6157	207000	<0.001	0.199

Trends in flow-adjusted concentrations of total phosphorus in the central United States were significantly related to changes in phosphorus inputs from fertilizer (Table 2). Increases in phosphorus inputs from fertilizer were more widespread than increases in population density or phosphorus inputs from manure in the central United States during the study period (Supporting Information), and changes in population density and phosphorus inputs from manure were not significantly related to trends in flow-adjusted concentrations of total phosphorus regionally (Table 2). The rank correlation coefficient with phosphorus inputs from fertilizer, while significant, was low. This may have been due in part to geographic differences in the change in phosphorus inputs from fertilizer; these inputs generally increased west of the Mississippi River, but generally decreased east of the Mississippi River (Supporting Information). It may also have been due in part to localized changes in these phosphorus sources that were not apparent at the county level. In addition, other changes in phosphorus sources or transport such as wastewater discharge or management practices like conservation tillage likely were important either regionally or at individual sites.

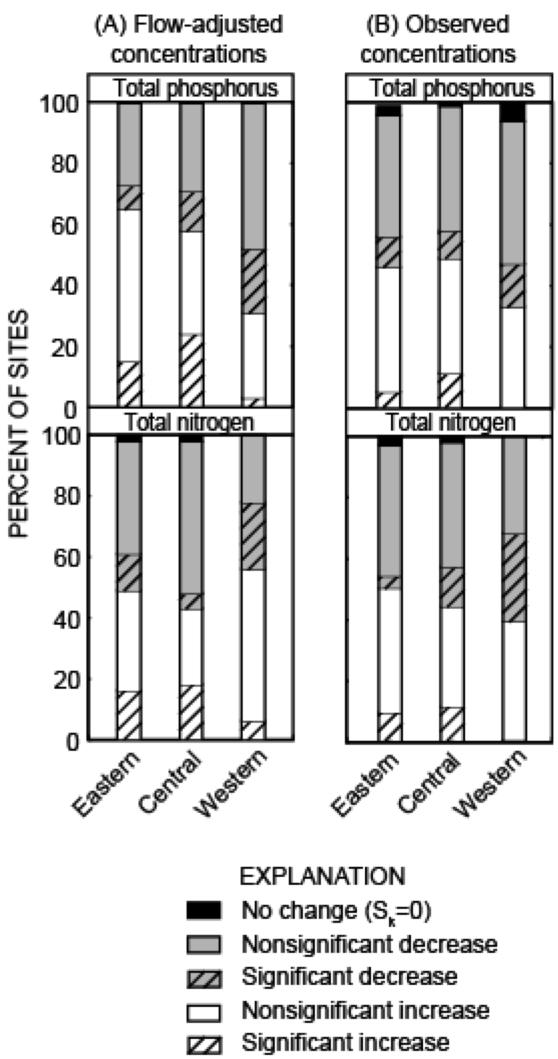

FIGURE 2. Seasonal Kendall trend results (α = 0.10) for the (A) flow-adjusted and (B) observed concentrations at individual sites in each region.

TABLE 2. Kendall's Tau Rank Correlation between Trends in Flow-Adjusted Concentrations of Total Phosphorus at Study Sites in the Central United States and Changes in Phosphorus Sources in the Watershed of Those Sites

change in source	number of sites	tau	p-value
fertilizer	38	0.22	0.054
manure	38	0.14	0.213
population density	38	0.13	0.260

Although changes in nitrogen inputs from fertilizer often mirrored changes in phosphorus inputs from fertilizer (Supporting Information), a similar upward regional trend in flow-adjusted total nitrogen concentrations in the central United States was not found. One possible explanation for the difference in phosphorus and nitrogen trends is that soils in this region are at or approaching phosphorus saturation, which can occur in areas that have been subject to long-term manure or fertilizer application (*27*). Because the amount of phosphorus in surface runoff can increase with the phosphorus content of soils (*28, 29*), over time a greater proportion of the phosphorus load applied to the land surface may be reaching streams, particularly in agricultural areas. Another possibility is that a substantial source of nitrogen in some of the streams is groundwater (*30*). Because it often can take years to decades for groundwater to travel to streams (*31*), there may be a lag between changes on the land surface and changes in total nitrogen concentration in streams. Nitrogen typically is transported through groundwater to a greater extent than phosphorus, which sorbs more readily to soil; therefore, nitrogen trends in streams are more likely to be affected by groundwater lag times than are phosphorus trends.

Regional Trends in Observed Concentration. After correction for spatial correlation, there were no significant regional trends in observed concentrations of either total phosphorus or total nitrogen (Table 1). The number of sites with general increases in observed concentrations was comparable to the number of sites with decreases for total phosphorus and total nitrogen in the eastern and central United States, and many of these changes resulted in trends that were nonsignificant (Figure 2B). There were more sites with general decreases than increases in observed concentrations of total phosphorus and total nitrogen in the western United States. Only a small percentage of those decreases was significant for total phosphorus; a greater percentage was significant for total nitrogen (Figure 2B). Many of the decreases in observed total nitrogen concentrations, however, occurred at closely spaced sites in the Willamette River Basin in Oregon (*14*), indicating that spatial correlation may have been a substantial influence in the regional analysis.

The upward regional trend in flow-adjusted total phosphorus concentrations in the central United States was not found for observed total phosphorus concentrations (Table 1). Precipitation decreased during drought conditions in the latter part of the study period in many parts of the central United States (*32, 33*), contributing to downward trends in streamflow at many sites in the region (*14*). The effects of increased source loading indicated by the upward regional trend in flow-adjusted concentrations likely were offset by the effects of decreased runoff indicated by the downward trends in streamflow, resulting in a nonsignificant regional trend in observed total phosphorus concentrations. Without the decrease in streamflow and associated runoff at these sites, in-stream concentrations of total phosphorus might have been higher than actually occurred.

The trend results from this study are notably different than those from previous studies of nutrient trends in United States rivers from 1978 to 1987 (*34*) and from 1975 to 1994 (*35*). From 1978 to 1987 and from 1975 to 1994, there were more downward than upward trends in total phosphorus concentrations; the infrequent upward trends were confined largely to parts of the eastern United States. From 1978 to 1987, there were more upward than downward trends in total nitrogen concentrations, but from 1975 to 1994, there were more downward than upward trends in total nitrogen concentrations. The disparate results in the three trend periods, which likely reflect differences in contemporaneous and prior (lagged) anthropogenic changes, climatic changes, and stream conditions at the start and end of each record, underscore the need for continued long-term monitoring of streams and rivers in the United States.

Acknowledgments

The authors thank E. Trench, D. Harned, D. Robertson, R. Rebich, N. Bauch, G. Fuhrer, D. Wise, C. Kratzer, and D. Saleh for their contributions to trend analysis and interpretation, K. Hitt, N. Nakagaki, J. Gronberg, J. Falcone, and D. Wolock for GIS data compilation, and D. Clow, J. Kiang, and three anonymous reviewers for their helpful comments on the manuscript. This research was funded by the U.S. Geological Survey National Water Quality Assessment program.

Supporting Information Available

Maps showing changes in phosphorus loading from manure, phosphorus loading from fertilizer, population density, nitrogen loading from manure, nitrogen loading from fertilizer, and nitrogen loading from atmospheric deposition in the United States during the study period. This information is available free of charge via the Internet at http://pubs.acs.org.

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ES803664X