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Trends in the nutrient enrichment of U.S. rivers during the late 20th century and their relation to changes in probable stream trophic conditions

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Abstract

We estimated trends in concentrations of total phosphorus (TP) and total nitrogen (TN) and the related change in the probabilities of trophic conditions from 1975 to 1994 at 250 nationally representative riverine monitoring locations in the U.S. with drainage areas larger than about 1,000 km^2. Statistically significant ($p < 0.05$) declines were detected in TP and TN concentrations at 44% and 37% of the monitoring sites, and significant increases were detected at 3% and 9% of the sites, respectively. We used a statistical model to assess changes in the probable trophic-state classification of the sites after adjusting for climate-related variability in nutrient concentrations. The probabilistic assessment accounts for current knowledge of the trophic response of streams to nutrient enrichment, based on a recently proposed definition of ‘‘eutrophic,’’ ‘‘mesotrophic,’’ and ‘‘oligotrophic’’ conditions in relation to total nutrient concentrations. Based on these trophic definitions, we found that the trophic state improved at 25% of the monitoring sites and worsened at fewer than 5% of the sites; about 70% of the sites were unchanged. Improvements in trophic-state related to declines in TP were more common in predominantly forested and shrub–grassland watersheds, whereas the trophic state of predominantly agricultural sites was unchanged. Despite the declines in TP concentrations at many sites, about 50% of all monitoring sites, and more than 60% of the sites in predominantly agricultural and urban watersheds, were classified as eutrophic in 1994 based on TP concentrations. Contemporaneous reductions in major nutrient sources to streams, related to wastewater treatment upgrades, phosphate detergent bans, and declines in some agricultural sources, may have contributed to the declines in riverine nutrient concentrations and associated improvements in trophic conditions.

Cultural sources of nutrients to streams changed appreciably during the last three decades of the 20th century in the U.S. Advances in wastewater treatment, enabled by the 1972 Clean Water Act, and state-level phosphate detergent bans greatly reduced municipal point sources of nutrients to streams (Litke 1999). Diffuse nutrient sources, including fertilizers and fossil-fuel emissions, rose to peak levels in the late 1970s and early 1980s and have varied considerably since that time (Howarth et al. 2002). Soil conservation and government-sponsored retirements of farmlands have received increasing attention since the 1980s (USDA 1997), although knowledge of their effects on stream nutrients are limited.

Despite these widespread changes in nutrient sources and management-related activities, there is surprisingly little information about how the trophic state of U.S. streams has changed over the past several decades, especially in response to changes in nutrient enrichment. Biennial reports of stream-use impairment from state water-quality inventories are generally of limited use for tracking historical changes in water quality and trophic conditions (Mayio and Grubbs 1993; Litke 1999). Moreover, previous studies of nutrient trends in major U.S. rivers (Smith et al. 1987, 1993; Lurry and Dunn 1997) and biological measures in state waters (e.g., Ohio Environmental Protection Agency 1998) have not provided a comprehensive assessment of changes in the trophic conditions of the nation's streams. A major problem is that long-term monitoring records of algal biomass are too sparse to allow a representative assessment of the nation's rivers. State biological assessments have only recently been implemented in most states. Prior to this, few states had adopted direct measures of stream algal biomass, such as the measures of chlorophyll that have been successfully used to manage lakes (U.S. EPA 1996, 2002). An additional challenge is that the highly variable response of stream algae to nutrient enrichment (e.g., Van Nieuwenhuyse and Jones 1996) has complicated efforts to develop a trophic-state classification system for streams such as that employed for lakes (Dodds et al. 1998; U.S. EPA 2000). Moreover, published trend information has limited utility for assessing the ecological relevance of historical changes in stream nutrients; frequently, trends are reported as an absolute or percentage change in mean concentrations without any information about how the trends relate to trophic conditions.

Recently, some progress has been made in developing a general method for identifying the trophic state of streams based on concentrations of nutrients and chlorophyll (Dodds et al. 1998; U.S. EPA 2000). This proposed trophic classification is supporting initial efforts to identify national- and state-level criteria for nutrients in streams and rivers (U.S. EPA 2000). Here, we use this classification system as a framework for evaluating the potential trophic importance of changes in the nutrient enrichment of U.S. rivers over approximately the last quarter of the 20th century. Using long-term monitoring data collected at 250 locations on major rivers throughout the U.S. from 1975 to the 1990s, we sta-

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tistically model changes in ambient nutrient concentrations and concentrations standardized for seasonal and stream flow variability. The statistical models provide a consistent method for explaining climate-related changes in nutrient concentrations and adjusting for subtle differences in monitoring record lengths and sampling frequencies. These models make it possible to more reliably compare nutrient trends for similar seasons and years across sites and evaluate them for possible anthropogenic causes. Our analysis extends conventional trend detection methods by evaluating changes in nutrients in relation to concentration ranges that have been shown to generally correspond to different trophic states of flowing waters. The method quantifies changes in the probability that nutrient concentrations observed at the stream monitoring sites fall within the concentration intervals of the trophic classes of the recently proposed classification system (i.e., oligotrophic, mesotrophic, and eutrophic; Dodds et al. 1998). This assessment method addresses the difficulty of using conventional measures of trend (e.g., absolute or percentage change) and their statistical uncertainties to evaluate the relation of changes in stream nutrient concentrations to concentrations that have relevance to trophic conditions. The probability-based approach effectively integrates current knowledge of the trophic response of streams to nutrient enrichment with statistical models that characterize the likely range of variation in stream nutrient concentrations and the level of statistical confidence in estimated trends.

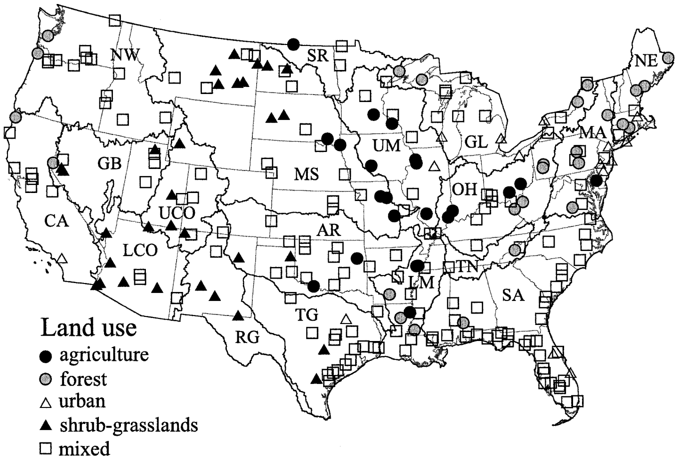


Fig. 1. Location of the 250 USGS water-quality monitoring sites with predominant land use of the watersheds and major regional drainage basins. Regional drainage basin symbols are NE (New England), MA (Mid-Atlantic), SA (South-Atlantic-Gulf), GL (Great Lakes), OH (Ohio), TN (Tennessee), UM (Upper Mississippi), LM (Lower Mississippi), SR (Souris-Red-Rainy), MS (Missouri), AR (Arkansas-Red-White), TG (Texas-Gulf), RG (Rio Grande), UCO (Upper Colorado), LCO (Lower Colorado), GB (Great Basin), NW (Pacific Northwest), CA (California). The method for identifying the predominant land use of the watershed of each monitoring site is described in the text.

Methods

Water-quality monitoring data—Regularly collected water-quality data were obtained from long-term monitoring stations in the U.S. Geological Survey's National Stream Quality Accounting Network (NASQAN; for details, *see* Alexander et al. 1996; Hooper et al. 2001). We selected 250 stations (*see Fig. 1*) with data spanning the period of 1973 to 1994—fewer than 10% of the station records began after 1973 (i.e., in the 1974–1975 period), and fewer than 20% of the station records ended prior to 1994 (i.e., in the 1992–1993 period). Water-quality records extend through 2001 at 27 of the stations, the latest year for which data were retrieved. The set of 250 stations provides a geographically representative description of the water quality of major rivers of the U.S. (Smith et al. 1993; Alexander et al. 1996; *see Tables 1 and 2*). Table 2 compares land use and population characteristics of the monitoring stations with those for the hydrologic cataloging units, a national set of officially defined watersheds (Seaber et al. 1987) that are generally representative of the physiography, land use, and population in the drainage basins of large rivers of the United States (the characteristics of the monitoring station watersheds in Table 2 are generally similar to those reported for the national set of hydrologic cataloging units). Each of the station records has at least 70 observations of total nitrogen (TN) and total phosphorus (TP)—sampling frequencies were at least quarterly at all stations with bimonthly sampling conducted at a majority of the sites; monthly sampling was common at most sites prior to 1982. Total nitrogen measurements were calculated as the sum of measurements of total kjeldahl nitrogen (ammonia plus organic) and dissolved nitrate plus nitrite. Dissolved nitrate–nitrite and phosphorus were analyzed on filtered water samples (based on use of 0.45-μ pore size filters). Nondetected nutrient values—values reported as less than laboratory reporting limits—typically comprise less than 25% of the observations at the sites (interquartile range for the stations: TN, 3% to 23%; TP, 0 to 4%)—fewer than 10% of the sites have more than 39% and 14% nondetected values for TN and TP, respectively. The nondetects for TN most frequently result from nondetected values of nitrate–nitrite. All nondetected values are included in the statistical analyses (*see next section*). Measurements of suspended sediment, turbidity, and fecal coliform bacteria, an indicator of human and animal wastes, were also tested for trend to provide ancillary measures of water-quality properties that are frequently associated with nutrient sources. Daily mean values of stream flow were paired with all water-quality measurements and used in statistical models to adjust for flow variability. The turbidity method changed from a Jackson candle turbidimeter (JTU) to a nephelometer (NTU) in 1978. Comparisons of available turbidity measurements, where both methods were used (about 1,000 pairs at 100 stations), indicated that differences were typically less than about 10% (interquartile range of the difference in JTU and NTU expressed as a percentage of the total = -9.1% to 10.5%) with NTU measurements slightly higher on average than JTU measurements (linear regression equation for $n = 1{,}004$: NTU = 0.888 + 0.999 JTU). Composite turbidity records were constructed to span the 1973 to 1994 time period.

Statistical models of water-quality concentrations—We developed statistical models of historical concentrations of

Table 1. Selected water-quality and watershed characteristics for the 250 USGS stream monitoring stations. The reported percentiles for ''drainage area'' are based on the total drainage area upstream of each site. DIN is dissolved inorganic nitrogen computed as the sum of nitrate–nitrite and ammonia.

Station property	Percentiles of the station mean-annual estimates				
	10th	25th	Medium	75th	90th
Total nitrogen (TN) (mg L^{-1})	0.60	0.81	1.27	1.92	3.4
Nitrate–nitrite–N, dissolved (mg L^{-1})	0.11	0.19	0.39	0.82	2.0
Kjeldahl nitrogen, total (mg L^{-1})	0.41	0.54	0.77	1.16	1.6
Ratio of DIN to TN	0.22	0.30	0.42	0.59	0.70
Total phosphorus (TP (mg L^{-1})	0.04	0.06	0.12	0.25	0.46
Dissolved phosphorus (mg L^{-1})	0.02	0.02	0.05	0.10	0.23
Ratio of dissolved phosphorus to TP	0.37	0.45	0.57	0.68	0.78
Turbidity (NTU)	2	6	20	58	146
Suspended sediment (mg L^{-1})	8	18	77	222	832
Fecal coliform bacteria (counts 100 ml^{-1})	37	118	329	950	2345
Streamflow (m^3 s^{-1})	5	17	62	250	807
Drainage area (km^2)	1,094	3,543	16,096	43,805	167,832

nitrogen (total, total kjeldahl, and dissolved nitrate–nitrite) and phosphorus (total and dissolved) for each monitoring station. The models predict stream concentration as a function of mean stream flow, season of the year, and long-term linear trend in concentration. The linear trend describes long-term annual changes in concentration that may relate to the transport or supply of pollutants to streams. Similar models were also developed for turbidity, suspended sediment, fecal coliform bacteria, and stream flow; only the seasonal and long-term linear trend variables were used to model stream flow. Water-quality models of this general type have been used previously in trend studies of data from the U.S. Geological Survey (USGS) water-quality stations examined in this study (e.g., Cohn et al. 1989, 1992; Langland et al. 2000). In this study, these models were extended to account for both the contemporaneous and antecedent stream flow conditions associated with each water-quality observation. Persistent antecedent flow conditions over time periods of 1 to 5 yr, caused by extended periods of drought or high precipitation and runoff, recently have been found to be important in explaining temporal variability in water-quality concentrations in major rivers of the U.S. (Vecchia 2003). Therefore, we modeled water-quality concentrations according to

$$\ln C_i = \beta_0 + \beta_1 T_i + \beta_2 Q_{0,i} + \beta_3 Q_{1,i} + \beta_4 Q_{5,i} + \beta_5 \sin(2\pi T_i) + \beta_6 \cos(2\pi T_i) + \varepsilon_i \qquad (1)$$

where ln is the natural logarithm, C_i is the instantaneous concentration (units are mg L^{-1}, except NTU for turbidity and colonies per 100 ml for fecal bacteria) for the *i*th observation; T_i is decimal time in years (the descriptor of annual linear trend in concentration); $Q_{5,i}$ is the 5-yr antecedent stream flow value calculated as the mean of the daily flow deviations, $Q^*_{D,i}$ (obtained as the difference between the log daily flow and the period-of-record mean of log daily flows),

Table 2. Land-use and population characteristics for the 250 USGS stream monitoring stations and hydrologic cataloging units of the United States. The 2,049 hydrologic cataloging units (HCU) are an officially defined set of U.S. drainage basins (Seaber et al. 1987) of generally uniform size (drainage area interquartile range from 2,100 to 4,800 km^2).

Property	Percentiles of the station and HCU properties				
	10th	25th	Median	75th	90th
USGS monitoring stations					
Land use (% of total drainage area)					
Agriculture	2	7	21	38	60
Forest	4	13	33	59	74
Shrub–grassland	0	0	11	52	69
Urban	<1	<1	1	3	9
Population density (persons km^{-2})	2	4	14	37	85
Hydrologic cataloging units					
Land use (% of total drainage area)					
Agriculture	<1	4	18	44	71
Forest	1	5	25	58	77
Shrub–grassland	0	0	11	58	83
Urban	<1	<1	1	2	6
Population density (persons km^{-2})	1	2	11	27	76

for the 5 yr preceding and inclusive of the current observation; $Q_{1,i}$ is the 1-yr antecedent stream flow calculated as the mean of the deviations of values of $Q^*_{D,i}$ from the 5-yr antecedent stream flow values for the 1 yr preceding and inclusive of the current observation; $Q_{0,i}$ reflects contemporaneous daily flow variation and is defined as the deviation of $Q^*_{D,i}$ from the sum of the 5- and 1-yr antecedent stream flow values; the trigonometric functions of decimal time describe seasonal variation according to a single annual cycle; ε_i is the model error assumed to be independently and identically distributed as a normal random variable with mean zero and standard deviation σ; and β_0, β_1, . . ., β_6 are estimated regression coefficients. Model residuals were evaluated for normality and constant variance following calibration of the models. The contemporaneous and antecedent stream flow expressions in Eq. 1 are defined to be mutually orthogonal. Equation 1 provides an estimate of trend in climatically and seasonally adjusted water-quality concentrations. We also developed a log-linear model of water-quality concentrations without the flow and seasonal adjustments of Eq. 1 to describe long-term annual trends in ambient (i.e., nonadjusted) concentrations in the streams. This model is of the form

$$\ln C_i = \beta_0 + \beta_1 T_i + \varepsilon_i \tag{2}$$

We estimated the model parameters of Eqs. 1 and 2 by applying censored regression techniques in the SAS Lifereg procedure. The models in this application are of the classic Tobit form for left-censored data because selected nutrient observations are reported as below laboratory reporting limits. The Lifereg procedure was applied to records where fewer than 70% of the observations were censored. The procedure uses maximum likelihood estimation (MLE) based on a Newton-Raphson algorithm to estimate parameter values. The standard errors of the parameter estimates are based on large sample approximations to the normal distribution.

Long-term changes in ambient water-quality concentrations and concentrations standardized for seasonal and stream flow–related variability were quantified according to two methods. First, we used conventional trend detection methods to evaluate the sign and statistical significance (α = 0.05) of the linear time trend coefficient (β_1) in Eqs. 1 and 2.

Second, we used the water-quality concentration models to estimate changes in the probable trophic state of the monitoring stations by evaluating changes from 1975 to 1994 (and 2001 for the subset of 27 stations) in the probability that concentrations fall within each of three trophic-state classes—oligotrophic, mesotrophic, eutrophic (*see Table 3*). The years 1975 and 1994 were selected to provide a consistent measure of the broadest change in conditions over the time period of the monitoring records. In this approach, we first used the models in Eqs. 1 and 2 to predict the mean log concentration, $\bar{C}_P$, with standard error of prediction, σ_{SE}, corresponding to the summer period of the years 1975, 1994, and 2001. In Eq. 1, these predictions were also based on mean stream flow conditions for the period of record (i.e., antecedent and contemporaneous deviations from the long-term mean stream flow were set to zero). The standard error of prediction includes both the effects of coefficient and model error—i.e., statistical errors caused by uncertainty in the fitted model coefficients and by random deviations between the modeled and actual concentrations. Next, the probability, p^*, of observing log concentrations larger than the specified log concentration boundary for a trophic-state class, C^*, was obtained from a standard normal distribution with mean $\bar{C}_P$ and standard deviation σ_{SE} and evaluated according to a standard normal quantile value, z^*, such that

Table 3. Proposed classification of stream trophic state from Dodds et al. (1998).

Water-quality metric	Oligotrophic–mesotrophic boundary	Mesotrophic–eutrophic boundary
Mean benthic chlorophyll (mg m^{-2})	20	70
Sestonic chlorophyll (μg L^{-1})	10	30
Total nitrogen (mg L^{-1})	0.7	1.5
Total phosphorus (mg L^{-1})	0.025	0.075

$$z^* = \frac{C^* - \bar{C}_P}{\sigma_{\mathrm{SE}}} \tag{3}$$

Values of p^*, evaluated separately for the oligotrophic–mesotrophic and mesotrophic–eutrophic concentration boundaries, are referenced as p^{OM} and p^{ME}, respectively. The probabilities (p^{O}, p^{M}, p^{E}) that nutrient concentrations fall within the oligotrophic, mesotrophic, and eutrophic classes, respectively, were then computed for each time period and monitoring location according to

$$p^{\mathrm{E}} = p^{\mathrm{ME}}, \qquad p^{\mathrm{M}} = p^{\mathrm{OM}} - p^{\mathrm{ME}}, \qquad p^{\mathrm{O}} = 1 - p^{\mathrm{OM}} \tag{4}$$

We identified the ‘‘most probable’’ trophic state as the trophic class with the highest concentration probability in Eq. 4. An illustration of the method is presented for a monitoring station record for total nitrogen in Fig. 2.

Inferences about the causes of nutrient trends—A comprehensive investigation of the causes of the trends in nutrient concentrations is beyond the scope of this study; however, limited inferences can be made from ancillary information for the stations. Nutrient trends were examined for consistency with trends in other water-quality constituents, including turbidity, suspended sediment, and fecal bacteria, that may be influenced by changes in nutrient-related sources. The monitoring sites were also classified according to the predominant land use in their watersheds (i.e., agriculture, forest, shrub–grasslands, and urban; *see Fig. 1*) to provide information for interpreting the station trophic-state classifications and estimated changes in the trophic state. The initial criteria for identifying the predominant land use of a monitoring site specified that the percentage of the total drainage area upstream of the site must equal or exceed one of the following percentages of land use: 70% for forest, 60% for shrub–grasslands, 50% for agriculture, or 10% for urban, based on land-use data for 1992 (National Land Cover Data; http://www.epa.gov/mrlc/nlcd.html; Vogelmann et al. 2001). A population density (GeoLytics 1996) of more than 100 persons km^{-2} was also required for urban sites and a density of less than 100 persons km^{-2} was required for the

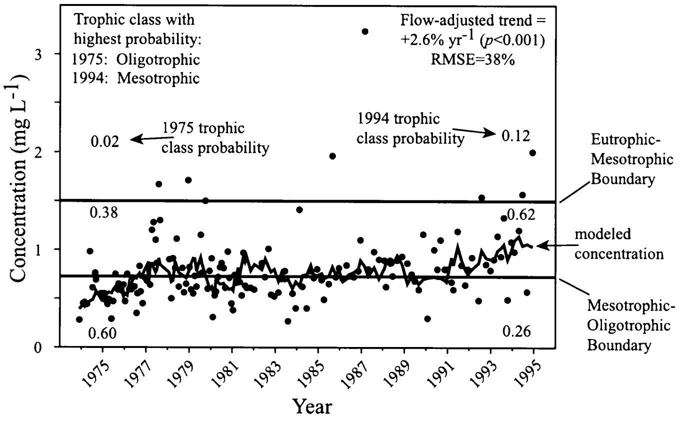


Fig. 2. Application of the trophic-state classification method to measured total nitrogen concentrations from the Pascagoula River near Benndale, Mississippi, 1973 to 1994. The modeled concentrations are based on the application of Eq. 1 to the measured concentrations. The trophic-state concentration boundaries are from a previously proposed (Dodds et al. 1998) trophic-state classification for streams. The ''trophic class probability'' indicates the probability that the total nitrogen concentrations during the summer period of the specified years (1975 and 1994) for long-term mean stream flow conditions fall within the concentration boundaries of each of the three trophic classes. The designation of the trophic classification for the site in 1975 (''oligotrophic'') and 1994 (''mesotrophic'') is based on the trophic class with the highest probability in each of these years. RMSE is the root mean square error expressed as a percentage. One observation of 5.6 mg L^{-1} in 1982 is not shown.

other site classifications. We further conditioned the station land-use classification by using empirical estimates of the contributions of nutrients at the monitoring stations from major point and diffuse sources within the watersheds, based on long-term mean-annual estimates from the spatially referenced regression on watershed attributes model (SPARROW; Smith et al. 1997; Alexander et al. 2000); the monitoring stations in this study are a subset of the SPARROW calibration data set. Municipal/industrial point sources and agricultural sources of nutrients contributed less than 10% and 40% of the total stream nutrient flux, respectively, at the forested and shrub–grassland sites; agricultural nutrient sources contributed more than 50% of the total stream nutrient flux at the agricultural sites; and agricultural nutrients contributed less than 50% of the total stream nutrient flux at the urban sites. Based on these criteria, 94 of the 250 monitoring stations were classified as having predominantly one of the four land uses; the remaining 156 were classified as having ''mixed'' land uses (*see Fig. 1*).

Major national changes in municipal wastewater and diffuse nutrient sources and agricultural management during the study period are also highlighted for comparison with the nutrient trends. Detailed comparisons of the nutrient trends with contemporaneous changes in agricultural nutrient sources were made for the 24 monitoring stations located in predominantly agricultural watersheds. Changes in fertilizers (Alexander and Smith 1990; B. Ruddy pers. comm.; USGS 2004) and livestock manure (Puckett et al. 1998) were estimated based on county-level data assembled for the total watershed drainage area upstream of these monitoring sites. We computed the percentage change in the mean fertilizer nutrients from the 1973–1976 period to the 1991–1994 period. The percentage change in livestock manure nutrients was computed as the difference in annual estimates between 1982 and 1992.

Based on a review of available historical information on USGS field and laboratory methods (Schertz et al. 1994; Alexander et al. 1996), we evaluated the nutrient trends for the effects of possible measurement errors during the 1980 to 1986 period, related to the use of mercuric chloride tablets as a sample preservative. A previous study of laboratory blanks indicated that kjeldahl nitrogen measurements during this period potentially contained nitrogen contamination (<0.5 mg L^{-1}) related to the use of a tablet form of a mercuric chloride preservative (Schertz et al. 1994). In addition, semiannual laboratory analyses of spiked standard reference samples for dissolved substances, conducted as part of a regular external audit program, indicated a possible positive measurement error in phosphorus during 1980–1981 (Alexander et al. 1996). We assessed the effects of these potential measurement errors by testing the sensitivity of the nutrient trend results to the measurements reported for the 1980–1986 time period; in the analysis, we eliminated all total nutrient observations from the station records during the period 1980–1986 and retested the records for trends.

Results

Estimation of the water-quality concentration models—The accuracy of the ambient concentration and climatically adjusted concentration models in Eqs. 1 and 2 is shown in Table 4. The accuracy is expressed according to the root mean square error (RMSE)—a conventional measure of the average prediction error of the model—which, in this application, indicates the average percentage error expected for a prediction of a daily concentration. The RMSE of the nutrient models ranged from a low of about 20% to more than 100%; values for most stations, based on the interquartile ranges, fall between 31% to 63% for total nitrogen and 45% to 84% for total phosphorus (*see Table 4*). The models of fecal coliform bacteria display the largest RMSEs. In general, the accuracy of the trend models tends to be highest (i.e., lowest RMSE) in the less hydrologically variable eastern rivers and for monitoring stations that drain large watersheds. Evaluations of the log residuals indicated that the model assumptions of constant variance and normality are generally satisfied to an acceptable degree.

Accounting for climatically induced variability in concentration by applying the ''flow-adjusted'' model from Eq. 1 improved model accuracy; model prediction errors for nutrients were typically reduced by about 15% (i.e., median reduction) with maximum reductions in error of nearly 60% (Table 4). Of the combined stream flow terms in the total nitrogen and total phosphorus models, the contemporaneous daily flow is among the most important stream flow variable—it is statistically significant ($\alpha = 0.05$) at approximately 70% of the stations. A majority of the stations (70%) display a positive relation between concentration and contemporaneous daily flow. The 1- and 5-yr antecedent flow

Table 4. Accuracy of the statistical models of water-quality concentrations for the USGS monitoring sites. Root mean square error (RMSE) expressed in percentage is a measure of the average percentage error in a predicted value of concentration. The ''concentration'' model is described by Eq. 2 and the ''flow-adjusted'' concentration model is described by Eq. 1.

Model	Percentiles of the station RMSE (%)*				
	Min.	25th	Median	75th	Max.
Total phosphorus					
Concentration	30	54	65	84	204
Flow-adjusted	24	45	55	71	144
Error reduction†	0	8	14	21	53
Dissolved phosphorus					
Concentration	38	60	71	85	143
Flow-adjusted	29	52	61	72	121
Error reduction†	0	6	10	18	49
Total nitrogen					
Concentration	21	37	45	63	138
Flow-adjusted	18	31	39	50	107
Error reduction†	0.4	8	13	22	57
Dissolved nitrate plus nitrite					
Concentration	20	58	85	120	198
Flow-adjusted	19	45	68	93	159
Error reduction†	2	12	21	28	62
Kjeldahl nitrogen, total					
Concentration	28	46	56	66	120
Flow-adjusted	26	42	50	60	97
Error reduction†	0	6	9	14	34
Turbidity					
Concentration	45	78	103	133	288
Flow-adjusted	40	62	77	97	190
Error reduction†	0	14	23	31	66
Suspended sediment					
Concentration	39	80	97	122	240
Flow-adjusted	33	62	73	85	156
Error reduction†	1	12	25	34	55
Fecal coliform					
Concentration	80	131	158	179	288
Flow-adjusted	62	108	127	147	224
Error reduction†	0	11	17	24	48
Stream flow (monthly sum)					
Concentration	8	65	89	124	273
Flow-adjusted	7	53	70	109	262
Error reduction†	0.3	8	14	22	39

† The percentage reduction in RMSE with the addition of variables in the flow-adjusted model.

* Note that the expression of RMSE as a percentage is an approximation and significantly overstates the magnitude of the error for percentages above about 60%.

condition coefficients are statistically significant at about 40% of the stations, although the 5-yr coefficient is statistically significant at fewer stations (28%) for total phosphorus. For total phosphorus, nearly equal numbers of positive and negative 1- and 5-yr antecedent flow coefficients are observed for the stations. By contrast, the total nitrogen models exhibit twice as many negative as positive 5-yr coefficients and nearly twice as many positive as negative 1-yr coefficients. Accounting for climatic variability in the physical constituents—suspended sediment and turbidity—led to even larger reductions in error (typically from 15% to 35%) than observed for the nutrients. For turbidity and suspended sediment, the contemporaneous and 1- and 5-yr antecedent flow coefficients are statistically significant at 90%, 50%, and 35% of the stations, respectively. The coefficients are also predominantly positive in these models ($>$70% of stations). For nutrients and the physical water-quality measurements, the seasonal coefficients are statistically significant at typically about two-thirds of the stations.

Trends in concentrations and physical water-quality measures—A national summary of the trend results for the 1975 to 1994 time period, based on an evaluation of the sign and statistical significance ($\alpha = 0.05$) of the linear time trend coefficient in Eqs. 1 and 2, is presented in Table 5. A map of the station trend results is presented in Fig. 3 for the flow-adjusted nutrient concentrations, based on an evaluation of the time coefficient in Eq. 1. The trend results for nutrients and the physical measures of water quality show many similarities for ambient concentrations (Eq. 2) and climatically or flow-adjusted concentrations (Eq. 1). The largest difference was observed for total nitrogen where the application of the flow-adjusted model led to a nearly 50% reduction in the number of statistically significant uptrends from 41 to 23 stations. Overall, about 6% to 10% fewer statistically significant time trends ($\alpha = 0.05$) were detected in the flow-adjusted nutrient models compared with the trends detected in the non–flow-adjusted models of ambient nutrient concentrations.

Application of the models in Eqs. 1 and 2 indicates that ambient and flow-adjusted concentrations of total nitrogen and total phosphorus showed more statistically significant declines than increases at stations from 1975 to 1994. Significant downtrends in flow-adjusted concentrations were observed at 37% and 44% of the monitoring stations for total nitrogen and total phosphorus, respectively. Downtrends in TN and TP are coincident at 54 stations, equivalent to 59% and 49% of all sites with downtrends in TN and TP, respectively. Downtrends in dissolved phosphorus concentrations occurred at six times as many sites as uptrends and are also coincident at nearly 60% of the stations with significant TP downtrends. By contrast, uptrends in dissolved nitrate–nitrite concentrations occurred at more than twice as many sites (26%) as downtrends (11%). About 25% of the sites with downtrends in TN also showed significant downtrends in nitrate–nitrite N; a negative trend slope (regardless of significance level) was indicated for dissolved nitrate–nitrate concentrations at 70% of the sites with significant downtrends in TN. We also detected a much larger number of significant downtrends than uptrends nationally in turbidity and suspended sediment concentrations; for suspended sediment by a 3 to 1 margin and for turbidity by more than a 10 to 1 margin. Among stations with significant downtrends in TN and TP concentrations, downtrends in turbidity and suspended sediment concentrations were coincident at approximately 50% and 40% of the stations, respectively. Nationally, four times as many stations were estimated to have

Table 5. Trends in ambient and flow-adjusted water-quality concentrations at the USGS monitoring stations, 1975 to 1994. The ''concentration'' model is described by Eq. 2 and the ''flow-adjusted'' concentration model is described by Eq. 1. An α value of 0.05 was used to classify statistically significant ''up'' and ''down'' trends.

		Up trends				Down trends			
Consitituent	No. sites	No. significant sites	% of total sites	Mean slope (% yr^{-1})	Min. slope (% yr^{-1})	No. significant sites	% of total sites	Mean slope (% yr^{-1})	Min. slope (% yr^{-1})
Total nitrogen									
Concentration	249	41	17	2.0	0.8	88	35	−3.9	−0.8
Flow-adjusted	249	23	9	2.5	0.7	92	37	−4.0	−0.8
Dissolved nitrate									
Concentration	245	77	31	4.0	0.9	27	11	−5.7	−0.8
Flow-adjusted	245	63	26	4.3	1.1	26	11	−5.8	−1.0
Kjeldahl nitrogen									
Concentration	249	18	7	2.2	1.2	98	39	−3.1	−1.0
Flow-adjusted	249	12	5	3.9	1.4	103	41	−3.9	−1.1
Total phosphorus									
Concentration	250	15	6	4.0	1.5	120	48	−4.1	−1.0
Flow-adjusted	250	8	3	3.7	1.7	111	44	−4.2	−1.1
Dissolved phosphorus									
Concentration	220	14	6	5.5	2.7	85	39	−5.6	−1.4
Flow-adjusted	220	12	5	5.0	2.5	77	35	−6.1	−1.2
Suspended sediment									
Concentration	233	25	11	4.6	1.8	75	32	−6.1	−1.6
Flow-adjusted	233	21	9	6.5	2.1	75	32	−6.0	−1.3
Turbidity									
Concentration	243	6	3	5.9	5.3	93	38	−5.4	−1.5
Flow-adjusted	243	8	3	8.5	4.6	103	42	−5.7	−1.3
Fecal coliform									
Concentration	220	15	7	7.5	4.4	57	26	−12.2	−4.4
Flow-adjusted	220	15	7	10.0	4.1	56	26	−13.0	−3.5
Stream flow (monthly sum)									
Concentration	249	30	12	4.7	0.9	76	31	−3.3	−0.4
Flow-adjusted	249	39	16	4.2	0.9	87	35	−3.0	−0.4

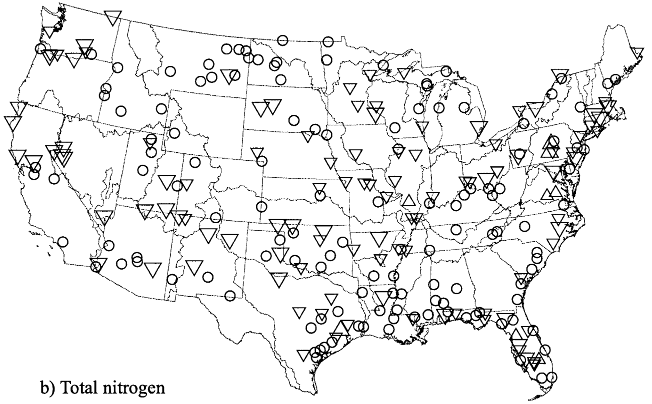
downtrends in fecal coliform concentrations (24%) as uptrends (6%); among stations with significant TN and TP downtrends, downtrends in fecal coliform were observed at eight sites and uptrends at one site (negative trend slopes were observed at two-thirds of the sites).

The downtrends in TN and TP concentrations occurred from 1975 to 1994 at river locations throughout the U.S. (*see Fig. 3*). Downtrends were observed in total phosphorus at more than half of the stations in the following regional drainage basins (*see regional basin locations in Fig. 1*): New England (six of eight stations), Upper Mississippi (9 of 12), Arkansas-Red (10 of 19), Great Basin (five of eight), and Pacific Northwest (8 of 15). Downtrends were observed in total nitrogen at more than half of the stations in the following regional drainage basins: New England (five of nine stations), Souris-Red-Rainy (two of three), upper and lower Colorado (14 of 18), Pacific Northwest (9 of 15), and California (six of eight).

Uptrends in total nitrogen occurred most frequently in the South Atlantic Gulf (6 of 41 stations), Upper Mississippi (4 of 12), and Missouri (4 of 31). Some of the largest watersheds showing uptrends included the Neuse River in North Carolina, Cumberland River in Tennessee, Mississippi River in Iowa, South Platte River in Colorado, and Missouri River in Missouri. Uptrends in total phosphorus were detected at only eight monitoring stations; most of the sites were located in the Mid-Atlantic and South Atlantic Gulf regions.

Trends in trophic-state classifications—The national percentage of the water-quality monitoring sites that are classified according to each of the trophic states is presented in Table 6 separately for total nitrogen and total phosphorus. The results in Table 6 are reported for the ''concentration'' (Eq. 2) and ''flow-adjusted'' (Eq. 1) trend models for the years 1975 and 1994. The ''most probable'' trophic state of each station was identified as the trophic class with the highest probability of occurrence. The mean probability among all stations within each trophic class is reported in Table 6 as the ''mean concentration probability.'' These mean probabilities range from 0.59 to 0.78 for total nitrogen and from 0.57 to 0.86 for total phosphorus; the ''eutrophic'' class typically has the highest probabilities among the trophic-state

a) Total phosphorus



b) Total nitrogen

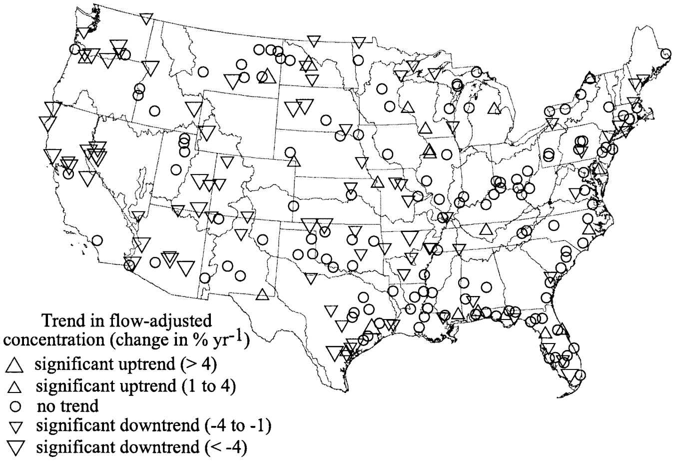


Fig. 3. Trends in flow-adjusted concentrations at the water-quality monitoring sites from 1975 to 1994 for (a) total phosphorus and (b) total nitrogen. The trends are based on an evaluation of the sign, slope (change in concentration expressed as percentage per year), and statistical significance ($\alpha = 0.05$) of the linear time trend coefficient in Eq. 1.

classes. Among stations with changes in their probable trophic-state classification based on predicted nutrient concentrations in 1975 to 1994, the trophic-state class probability changed by a median of 0.35 probability units (interquartile range: 0.15 to 0.49, TP; 0.25 to 0.45, TN). The trophic-state classifications of the monitoring stations, based on the percentage of stations within each trophic class, are generally similar among the concentration and flow-adjusted trend models. This is indicative of the somewhat small differences between many of the flow-adjusted and ambient-concentration trends (Table 5); moreover, the differences in the trophic class concentration probabilities associated with these trends were rather small and resulted in few differences in the trophic-state classifications (Table 6).

Changes in the trophic-state classifications from 1975 to 1994, based on total nitrogen and total phosphorus concentrations from the flow-adjusted trend models in Eq. 1, are shown for the monitoring sites in Fig. 4. Nationally, the trophic state is estimated to have improved from 1975 to 1994 in relation to total nitrogen and total phosphorus (i.e., change in classification from eutrophic to mesotrophic and mesotrophic to oligotrophic) at 26% and 23% of the sites, respectively. The trophic state declined at about 6% and 2% of the sites in relation to these nutrients, respectively. For total nitrogen, 41% of the stations were classified as mesotrophic in 1975, with about one-third of the stations classified as eutrophic. The percentage of stations classified as eutrophic fell to about 25% in 1994, with a corresponding increase from 1975 to 1994 in the percentage of stations classified as oligotrophic (change from 25% to 39%). About 50% of the stations with significant uptrends in total nitrogen were classified as eutrophic in 1975 and 1994; these included two stations in the Upper Mississippi region and four stations in the Missouri region. The results for total phosphorus show that a much larger percentage of the stations (66%) were classified as eutrophic in 1975 than observed for total nitrogen (33%). Stations classified as eutrophic, based on total phosphorus concentrations, declined to 55% in 1994 (a 16% reduction in the percentage of stations). A complementary rise from 1975 to 1994 was observed for total phosphorus in stations classified as oligotrophic and mesotrophic—increasing from 8% to 18% and from 25% to 27%, respectively. Six of the eight stations with significant uptrends in total phosphorus were classified as eutrophic in 1975 and 1994.

Trophic-state classifications based on total nitrogen and total phosphorus concentrations are coincident at nearly 50% of the monitoring sites (Table 6). Based on the flow-adjusted model results for the stations with coincident estimates of the trophic-state classification, the percentage of stations showing eutrophic conditions in relation to both total nutrient concentrations decreased from 64% in 1975 to 45% in 1994. The percentage of stations with oligotrophic conditions was 13% in 1975 and increased to 32% in 1994. Coincident eutrophic classifications in relation to total nutrients occurred most frequently in 1975 in the Ohio (8 of 16 stations), Upper Mississippi (8 of 11), and Missouri (16 of 30) regions. By 1992, the percentage of stations with coincident eutrophic classifications slightly declined in all of these regions (Ohio—6 of 16 stations; Missouri—12 of 30) except for the Upper Mississippi, where there was no change in the trophic status. Coincident oligotrophic classifications for total nutrients occurred most frequently at stations in the Great Lakes region, where this trophic state was reported for 6 of 16 stations in 1975 and increased to eight stations by 1994. The frequency of oligotrophic classifications in relation to total nitrogen and phosphorus also increased from two of eight stations in 1975 in the California region to five of eight stations in 1994.

Trophic-state trends in relation to land use—Changes in the trophic-state classifications of the monitoring sites from 1975 to 1994 are shown in Fig. 5 in relation to the predominant land use upstream of the monitoring sites (*see Fig. 1* for the land-use classification of the stations). The results in Fig. 5 generally illustrate the effect of land use on water-quality concentrations and the estimated trophic state of waters at the monitoring sites. Sites identified as having predominately agricultural or urban lands in their drainage were more frequently classified ($>$50% of the sites) as eutrophic in 1975 than sites in watersheds draining other types of land uses. Concentrations of total phosphorus were sufficiently

Table 6. Percentage of the USGS stream monitoring stations classified according to oligotrophic, mesotrophic, and eutrophic conditions in 1975 and 1994, based on estimated nutrient concentrations. The ''most probable'' trophic state of each station was identified as the trophic class with the highest probability that nutrient concentrations are observed within its corresponding concentrations interval (based on application of Eq. 4 according to the trophic concentration intervals defined by Dodds et al. 1998—*see Table 3*). The mean probability among all stations within each trophic class is reported as the ''mean concentration probability.'' The ''concentration'' model is described by Eq. 2, and the ''flow-adjusted'' concentration model is decribed by Eq. 1. Estimates for the ''flow-adjusted'' model are standardized to reflect concentrations during the summer period for long-term mean streamflow conditions. The percentage of stations classified according to each trophic state is also reported for a subset of the 250 stations for which the trophic-state classification for both total nitrogen and total phosphorus was identical.

				Trophic-state classification [percentage of stations (mean concentration probability)]		
Constituent	Model type	Year	No. of sites	Oligotrophic	Mesotrophic	Eutrophic
Total nitrogen (TN)	Concentration	1975	249	27.7 (0.67)	37.4 (0.59)	34.9 (0.73)
		1994	249	38.6 (0.76)	33.3 (0.59)	28.1 (0.75)
Total nitrogen (TN)	Flow-adjusted	1975	249	25.3 (0.71)	41.4 (0.63)	33.3 (0.75)
		1994	249	39.4 (0.78)	36.1 (0.63)	24.5 (0.78)
Total phosphorus (TP)	Concentration	1975	250	10.0 (0.62)	26.8 (0.57)	63.2 (0.81)
		1994	250	20.8 (0.66)	26.8 (0.57)	52.4 (0.81)
Total phosphorus (TP)	Flow-adjusted	1975	250	8.4 (0.66)	25.2 (0.62)	66.4 (0.86)
		1994	250	18.4 (0.67)	26.8 (0.59)	54.8 (0.85)
Stations with identical trophic-state classifications						
TN, TP	Concentration	1975	125	15.2	22.4	62.4
		1994	133	30.8	22.6	46.6
TN, TP	Flow-adjusted	1975	117	12.8	23.1	64.1
		1994	118	32.2	22.9	44.9

high at the agricultural sites such that none of the stations were classified as oligotrophic. By contrast, most of the predominately forested sites (>70%) were classified as either mesotrophic or oligotrophic in 1975. Among the forested sites, 27% of the sites were classified as eutrophic in 1975 based on total phosphorus concentrations. A large percentage (~75%) of the shrub–grasslands and mixed land-use sites were classified as eutrophic in 1975, based on concentrations of total phosphorus. However, fewer shrub–grassland sites were classified as eutrophic (nearly 45%), based on total nitrogen concentrations.

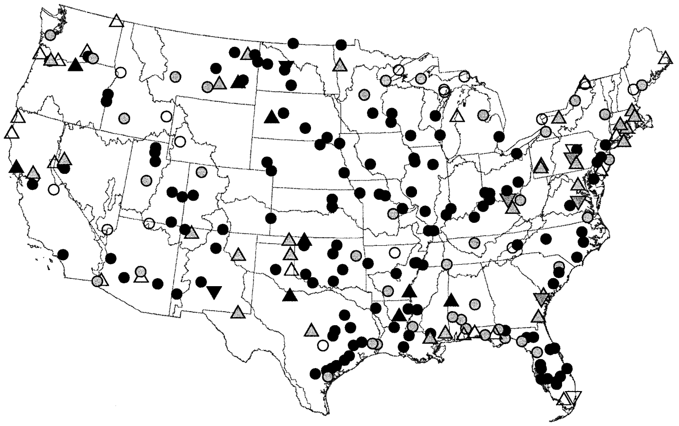
The percentage of sites that showed improvements from 1975 to 1994 in their trophic state (i.e., change in classification from eutrophic to mesotrophic or mesotrophic to oligotrophic) ranged from 20% to 50% within each of the land-use classes with the exception of total phosphorus at agricultural sites, which showed no changes in trophic state (Fig. 5); fewer than 10% of the sites in the various land-use categories displayed a worsening of the trophic-state conditions. Land uses with the largest percentage of sites showing improvements in trophic state were shrub–grassland sites for total nitrogen (52%) and forested sites for total phosphorus (42%). Most of these shrub–grassland sites (11 of 16 sites) are located on rivers in the southwestern U.S., including tributaries to the Colorado River and Rio Grande. Some of the predominantly forested sites that showed improvements in trophic state drain large eastern watersheds with moderate population densities; these include the Connecticut River at Thompsonville, Connecticut; Susquehanna River at Harrisburg, Pennsylvania; and sites on the Allegheny and Monongahela Rivers in Pennsylvania.

Improvements in trophic state in relation to total nitrogen concentrations were observed at about one-third of the agricultural sites ($n = 8$). Six of the eight agricultural sites showing improvements in relation to total nitrogen were located in the Missouri and Lower Mississippi regions. Downtrends also occurred in suspended sediment at seven of the sites (four of the downtrends were statistically significant). Among all agricultural sites, trends in total nitrogen were down at 13 stations, of which nine were statistically significant. At sites with statistically significant downtrends, manure nitrogen declined from 1982 to 1992 by an average of 15%, whereas nitrogen fertilizer increased from 1973 to 1994 by an average of about 60%.

Statistically significant uptrends were observed in total nitrogen at four agricultural sites. Three of these sites were classified as eutrophic in 1975 and 1994 and included the Mississippi River in Iowa, Rock River in Illinois, and Maumee River in Ohio; the classification of the Choptank River in Maryland changed from mesotrophic to eutrophic from 1975 to 1994. Nitrogen fertilizer increased at these sites from 1973 to 1994 by an average of 35%, whereas livestock manure nitrogen declined by an average of 19%.

Although the trophic-state classification remained unchanged from 1975 to 1994 at agricultural sites in relation to total phosphorus, downtrends were observed in total phosphorus concentrations over this period at 21 of the stations (statistically significant trends at 11 stations). Phosphorus declines were also observed in livestock manure from 1982 to 1992 (median change = −14%) and in fertilizer from 1973 to 1994 (median = −17%) at nearly all of these sites.

Among the shrub–grassland sites with improved trophic conditions in relation to total nitrogen concentrations, suspended sediment downtrends occurred at 60% of the sites and turbidity and fecal coliform downtrends occurred at nearly 40% of the sites. Suspended sediment, turbidity, and


a) Total phosphorus

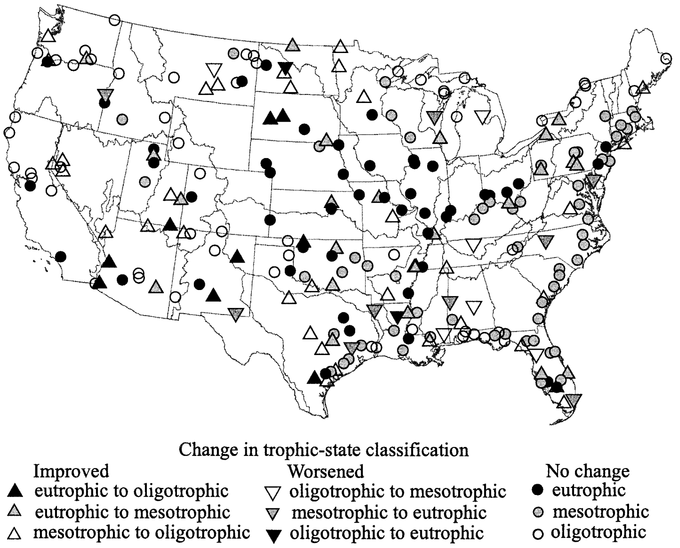

b) Total nitrogen

Fig. 4. Change in the probable trophic-state classification of the water-quality monitoring sites from 1975 to 1994, based on concentrations of (a) total phosphorus and (b) total nitrogen. The "most probable" trophic state of each station in 1975 and 1994 was identified as the trophic class with the highest probability that nutrient concentrations during the summer period of the specified years for long-term mean stream flow conditions are observed within the corresponding concentration interval of the trophic class (estimates are based on application of Eq. 4 according to the trophic concentration intervals defined by Dodds et al. 1998 in table 3). Watershed boundaries are shown for the regional drainage basins shown in Fig. 1.

fecal coliform downtrends occurred at about one-third of the forested sites showing improved trophic conditions in relation to total phosphorus concentrations. Among the agricultural sites with improved trophic conditions based on total nitrogen concentrations, 50% of the sites also showed downtrends in suspended sediment concentrations from 1975 to 1994.

Recent trends in nutrient concentrations and trophic conditions, 1994–2001—We examined trends in nutrients over the period 1994 to 2001 at 27 sites with available data to determine whether conditions had changed appreciably since 1994. The stations are located in the Mississippi (10 sites), New England (one), Mid-Atlantic (one), Southeast-Atlantic-Gulf (three), Great Lakes (one), Colorado and Great Basin (three), Pacific Northwest (five), and California (three). For the period 1975–1994, significant downtrends were observed at 15 of the 27 sites for total phosphorus and 12 of 27 sites for total nitrogen; one significant uptrend was observed in total nitrogen. For the period 1994–2001, significant trends were detected at only three monitoring stations for total nitrogen (two up, one down) and total phosphorus (three up); nearly equal numbers of positive and negative trend slopes (regardless of the significance levels) were found for the other sites. The three stations showing significant uptrends in total phosphorus were the Connecticut River in Connecticut, Ohio River in Illinois, and the Missouri River in Missouri. These three stations had previously shown downtrends from 1975 to 1994. Uptrends were observed in total nitrogen from 1994 to 2001 at the Alabama River in Alabama and the Columbia River in Oregon. Downtrends were previously observed at these sites for the 1975–1994 period. Of the 27 sites, 65% and 40% were classified as eutrophic in 1994 in relation to total phosphorus and total nitrogen concentrations, respectively. Improving trophic conditions from 1994 to 2001 were noted at one site for total phosphorus and three sites for total nitrogen; trophic conditions worsened at five sites for total phosphorus and two sites for total nitrogen. Stations showing a worsening of trophic conditions in relation to total phosphorus included the Connecticut River in Connecticut, Yellowstone River in Montana, Sacramento River in California, and the Columbia and Willamette Rivers in Oregon.

Effects of laboratory measurement errors on nutrient trends—We evaluated the sensitivity of the 1975–1994 nutrient trend results to potential measurement errors by eliminating nutrient observations from the station records for the years 1980–1986, a period when measurement errors may have occurred (*see Methods*). We then retested the records for trend over the 1975–1994 period. Changes in the sign and statistical significance of the nutrient trend slopes were relatively small, and the effects on the trophic-state classifications for total phosphorus and total nitrogen, especially those in relation to land use, were generally negligible. For total phosphorus, sufficient data were available to test for trends at 233 sites or 94% of the original number of sites; the percentage of sites showing downtrends (41%) and uptrends (3%) was similar to that reported for the unedited data (44% and 3%, respectively). The trend results were unchanged at 82% (192) of the sites; significant downtrends occurred at 90 sites and uptrends at five sites. For total nitrogen, sufficient data were also available to test for trends at 94% of the original number of sites; 31% showed downtrends and 12% showed uptrends as compared with 37% and 9% for the unedited data, respectively. The trend results were unchanged at 75% (174) of the sites; significant downtrends occurred at 65 sites and uptrends at 19 sites.

Discussion

Trend assessment method—The trend assessment method used here provides an ecological context for describing long-

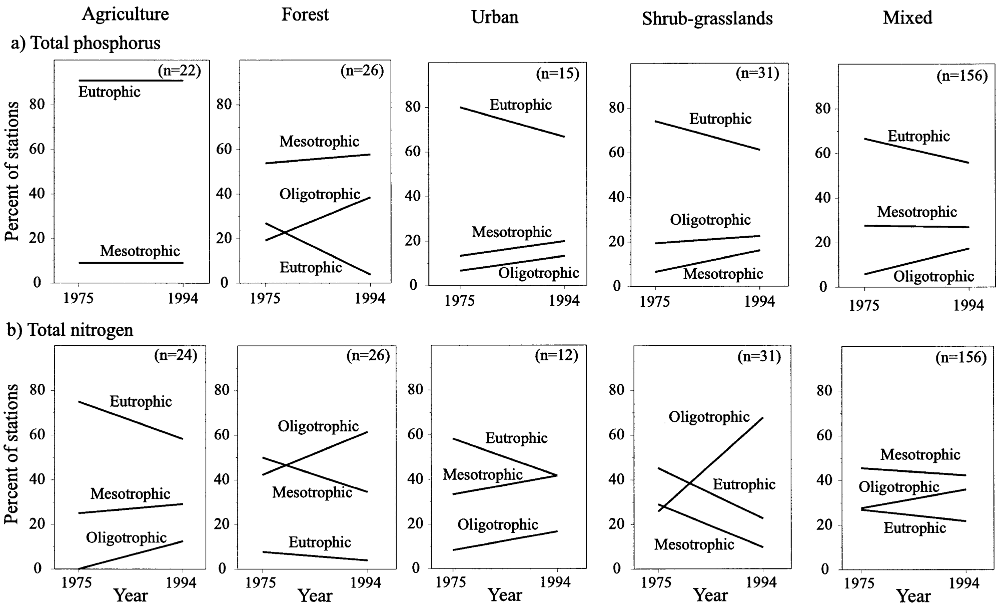


Fig. 5. Percentage of the water-quality monitoring sites within each land-use category, classified according to their probable trophic state in 1975 and 1994, based on modeled nutrient concentrations. The modeled concentrations are standardized to reflect nutrient conditions during the summer period for long-term mean stream flow conditions. The number of stations classified according to each land-use category is shown in parentheses; the locations of these sites are shown in Fig. 1.

term changes in the nutrient concentrations of streams. The method was applied to monitoring data collected from 1975 to the mid-1990s at 250 locations that are generally representative of the physiography, land use, and population in major U.S. river basins with drainage areas larger than about 1,000 km^2. We developed statistical models of water-quality concentrations as a function of stream flow, season of the year, and long-term linear trend. Measures of long-term trends in concentration, although frequently reported by scientists, cannot be used directly to assess the ecological importance of changes in stream nutrients. Scientists and water-resource managers require additional techniques that integrate the model estimates of trend and their uncertainties with information on the relevance of these trends to trophic conditions. We accomplished this by comparing the concentration predictions from the trend models in 1975 and 1994 with the concentration intervals of a recently proposed trophic classification system; these ranges of nutrient concentrations have been shown to generally correspond to different trophic states of streams (Dodds et al. 1998). Our method quantifies changes in the probability that nutrient concentrations fall within the concentration intervals of the trophic-state classes, thereby directly incorporating information about the statistical uncertainties of modeling variability in nutrient concentrations into the assessment of the probable trophic state of monitored stream waters.

The proposed trophic classification system for streams and rivers (Dodds et al. 1998) was developed as a relative index of trophic conditions based on the boundaries corresponding to the lower and upper thirds of the frequency distributions of nutrient and chlorophyll concentrations reported in the literature. These concentration data are from temperate, lotic systems representative of a variety of conditions, ranging from low-order streams to large rivers and pristine to highly developed watersheds. The concentration boundaries of the trophic classes approximately correspond to total nutrient concentrations that have relevance to the trophic state of flowing waters and lakes (Dodds et al. 1998). The relations of total nutrients to chlorophyll concentrations are generally more variable for streams than those observed for lakes, possibly related to the presence of larger quantities of suspended nonalgal particles in streams and the effects of stream hydraulics on periphyton biomass (Van Nieuwenhuyse and Jones 1996; Dodds et al. 1998, 2002). Our conclusions about changes in the probable trophic conditions at the monitoring sites are based exclusively on estimated long-term changes in total nutrient concentrations (both observed and those adjusted for seasonal and flow effects) in relation to the nutrient concentration boundaries of the trophic-state classes; information on changes in chlorophyll concentrations or other measures of algal biomass was unavailable for these sites. The estimates of changes in the probable trophic conditions assume that algal production at the sites was primarily controlled by nutrient availability; these estimates do not account for cases where algal growth was primarily limited by physical properties (e.g., light, temperature, water velocity and depth, and substrate conditions). The trophic-state classification system (Dodds et al. 1998) was proposed with the understanding that the concentration boundaries would be periodically reassessed as additional literature data on algal biomass and nutrient concentration relations becomes available. The nutrient-based trophic classifications of the monitored waters presented here may potentially change in response to new boundary definitions and, therefore, would also need to be reassessed in the event of changes in the concentration boundaries.

The method used here for characterizing changes in trophic conditions is based on shifts in the proportion of observations lying above or below the trophic-state concentration boundaries. The method is potentially sensitive to changes in concentration when a large proportion of the observations resides near the margins of the trophic-state boundaries. This may explain the trend results for sites where a change in the probable trophic-state classification is shown (Fig. 4), but no statistically significant trend is detected in nutrient concentrations (Fig. 3). Conversely, at sites where a large proportion of the nutrient concentrations do not reside near the trophic concentration boundaries, statistically significant trends in concentration may not result in a change in the probable trophic-state classification. Most of the trophic-state classifications of the monitoring stations (Table 6) were based on a probability of about 0.60 of observing concentrations within the trophic-state concentration interval (higher probabilities were observed for stations classified as ‘‘eutrophic’’). Among stations where the trophic-state classification changed between 1975 and 1994, the highest probability of observing concentrations within the trophic-state interval typically changed by about 0.35 probability units. Note that the assessment method does not provide information about increasing concentrations (i.e., worsening conditions) at sites classified as eutrophic at the beginning of the water-quality record and no information is provided about declining concentrations (i.e., improving conditions) at sites classified as oligotrophic at the beginning of the record; results from the method should be interpreted with conventional measures of trends to understand the nature of such changes.

Trend assessment results—We found that the statistically estimated nutrient trend models predicted individual concentrations with an acceptable level of accuracy; the accuracy ranges from 30% to 80% at most stations, based on the average measure of model uncertainty (i.e., root mean square error). Model accuracy of this magnitude for nutrients is generally state of the art for monitoring records of the length and number of samples analyzed here; similar levels of model accuracy have been previously reported for these and other regularly collected water-quality monitoring data (e.g., Cohn et al. 1992; M. J. Langland USGS pers. comm. 2004). In addition, we found that for the typical numbers of observations analyzed here ($n = 120$ to 150), the trend models were capable of detecting statistically significant changes in concentration of as little as 3% to 4% per year; changes of as little as 1% per year were detected in the most frequently sampled records.

A number of noteworthy results emerged from the assessment of trends and their relation to changes in probable trophic conditions at the monitoring sites. Statistically significant ($p < 0.05$) declines were detected in total phosphorus and total nitrogen flow-adjusted concentrations at 44% and 37% of the monitoring sites, and significant increases were detected at 3% and 9% of the sites, respectively. The downtrends in concentrations were geographically widespread, occurring at monitored river locations in many regions throughout the U.S. The results are generally consistent with previous national (Smith et al. 1987, 1993) and regional (Mississippi basin; Lurry and Dunn 1997) analyses of trends in total nutrients for similar time periods and monitoring stations as those analyzed here. A study of trends over the 1980–1989 period (Smith et al. 1993) noted that total phosphorus downtrends outnumbered uptrends by a factor of five to one and expanded on a geographic pattern of downtrends that was first evident in a study of total phosphorus trends from the mid-1970s to 1981 period (Smith et al. 1987).

Despite statistically significant declines in nutrient concentrations at many monitoring sites from 1975 to 1994, improvements in trophic state, defined as a change in the probable trophic-state classification (i.e., ‘‘eutrophic’’ to ‘‘mesotrophic’’ or ‘‘mesotrophic’’ to ‘‘oligotrophic’’), occurred at only about 25% of the sites. A worsening of trophic conditions occurred at fewer than 5% of the sites over this period; no change occurred in the trophic-state classification at about 70% of the sites. The largest percentages of sites with improved trophic state were located in predominantly forested and shrub–grassland watersheds. Some of these predominantly forested sites drain large eastern watersheds with moderate population densities; most of the shrub–grassland sites with improved trophic state are located on rivers in the southwestern U.S. Despite the declines in total phosphorus concentrations at many sites, about 50% of all monitoring sites, and as many as 60% of the sites in predominantly agricultural and urban watersheds, were classified as eutrophic in 1994 based on total phosphorus concentrations. Moreover, no changes were observed in the trophic-state classification of the agricultural sites from 1975 to 1994 in relation to total phosphorus concentrations despite statistically significant declines in total nutrients at many of these sites. The findings of elevated nutrient concentrations in agricultural and urban watersheds are consistent with previous studies of nutrients in small- to medium-sized streams during the 1990s (USGS 1999; Litke 1999) and in major rivers during the 1980s (Smith et al. 1993).

Statistically significant increases in flow-adjusted concentrations of total nutrients occurred at fewer than 10% of all monitoring sites, with more sites showing increases in concentrations of total nitrogen (9%) than total phosphorus (3%). About one-third of these sites showed a worsening of the probable trophic state; of these sites, most were classified as eutrophic in 1994.

Among the 27 stations for which records were available for the more recent 1994 to 2001 period, relatively few trends were observed in nutrient concentrations (~10% of sites) and trophic conditions (nearly 20% of sites), indicating that water-quality conditions were generally unchanged since 1994 at this sample of river locations.

Standardization of water-quality concentrations—Because the monitoring station records of concentrations reflect the effects of different climatic conditions and sampling characteristics (i.e., modest differences in record length and sampling frequency), we used the statistical trend models to standardize water-quality concentrations so that changes could be reliably compared for similar seasons and years across the network of sites. The statistical models account for variability in water-quality concentrations related to the season of the year, contemporaneous and antecedent stream flow conditions, and long-term linear changes (or trends) in concentration. The seasonal and stream flow components of the model account for the effects of both within- and between-year changes on water-quality concentrations caused by climate-related processes during the study period. Climatic effects may include the long-term storage of nutrients caused by extended periods of drought, the flushing and persistent transport of nutrients caused by lengthy periods of high precipitation, and the more immediate effects of individual storms on transport. Seasonal variations in nutrient concentrations, in addition to reflecting natural fluctuations in uptake by stream biota, may also reflect seasonal variations in human activities, such as those caused by agricultural practices. Our principle use of the seasonally and flow-adjusted trend models was to standardize the assessment of changes in the probable trophic-related levels of the nutrient concentrations at all monitoring stations (Table 6); nutrient concentrations were predicted for the summer period of the years 1975, 1994, and 2001 under long-term mean stream flow conditions. These statistical models were also used in a conventional assessment of water-quality trends (Table 5) to quantify long-term annual changes in seasonally and flow-adjusted concentrations. An adjustment for seasonal and flow-related variability improves the sensitivity of the method for detecting trends in water-quality concentrations that are related to long-term changes in pollutant sources and management activities (Hirsch et al. 1991).

Considering the effects of antecedent stream flow conditions on water-quality concentrations, as was done here, is a relatively new approach to modeling trends (Vecchia 2003) and provides an effective technique to account for annual and multiyear climatic effects on stream concentrations. In models of both contemporaneous and antecedent flow conditions, we found that contemporaneous daily variations in stream flow were most strongly correlated with nutrient concentrations at about 70% of the stations; however, antecedent flows with persistent anomalous variation over 1 to 5 yr were also strongly correlated with nutrient concentrations at as many as 40% of the stations. There was evidence that the inclusion of the antecedent flow variable adjusted for the effects of long-term trends in daily flow on concentrations. First, we found that as many as 10% fewer statistically significant trends were detected in nutrient concentrations, adjusted for contemporaneous and antecedent flow conditions for the 1975–1994 period, as compared with the trends in concentrations that were not flow adjusted. We found that this effect was even more pronounced on trends in total nutrients over the shorter 1994–2001 period, since approximately 60% fewer significant trends were detected after adjustment for antecedent flow conditions. In general, the effects of antecedent flow conditions on nutrient trends tend to be more detectable in monitoring records of 5–10 yr in length than in the longer records that were tested. Adjustment of water-quality concentrations for only contemporaneous variability in stream flow frequently improves the precision of trend detection and generally leads to larger numbers of detected trends compared with the trend results for ambient, non–flow-adjusted concentrations (e.g., Hirsch et al. 1991). Here, the results suggest that an important net effect of the adjustments for antecedent flow conditions was to correct for long-term annual trends in concentration caused by persistent changes or long-term trends in stream flow.

Second, among sites showing significant trends in both daily flow and total nutrients, there is evidence that differences in the magnitude of the nutrient trend slopes for flow-adjusted and nonadjusted concentrations are generally consistent with the direction of the concentration-flow correlation. For example, among the 36 sites showing significant downtrends in both total phosphorus and daily flow, larger magnitude flow-adjusted concentration slopes (relative to the unadjusted slopes) were found at all 19 stations where concentrations were negatively correlated with 5-yr antecedent flows. For the complementary set of 17 stations among the 36 sites, smaller magnitude flow-adjusted concentration slopes were found at 11 of 17 stations where concentrations were positively correlated with 5-yr antecedent flows. Despite the apparent effectiveness of these methods, there remains for any statistical flow-adjustment procedure the possibility that the water-quality concentration models have not fully corrected for the effects of year-to-year variations in stream flow at all stations; more complex model specifications than those applied here and additional data may be required in some cases to account for these effects.

Inferences about the causes of nutrient trends—We examined various ancillary information to evaluate potential causes of the observed nutrient trends and changes in probable trophic state. First, we found that the nutrient trends were generally consistent with those observed in turbidity, suspended sediment, and, to a lesser extent, fecal bacteria, properties that may have been influenced by changes in nutrient-related sources. For example, among the predominantly shrub and grasslands sites with improved trophic conditions from 1975 to 1994 in relation to total nitrogen, downtrends were also observed in suspended sediment, turbidity, and fecal coliform at 40% to 60% of the sites. Agricultural sites with improved trophic conditions in relation to total nitrogen ($n = 8$ sites) were also associated with declines in suspended sediment at seven of the sites. Among forested sites showing improved trophic conditions in relation to total phosphorus concentrations, downtrends were observed in suspended sediment, turbidity, and fecal coliform at about one-third of the sites. These ancillary water-quality properties are potentially related to various activities that affect the supply and mobilization of nutrients in the watersheds. The turbidity of stream waters may reflect both the contributions of nonalgal constituents of water (e.g., sediment) and in situ algal production in streams. Stream sediment concentrations potentially reflect soil erosion and cultural activities that protect or disturb soils, including conservation tillage practices and livestock grazing. Phosphorus, which is frequently transported in a particulate form, may also be influenced by processes affecting sediment supply and transport. Sediment is less likely to play a direct role in controlling the supply and transport of total nitrogen (more than 90% of the total nitrogen analyzed here is in a dissolved form) but may be associated with activities that disturb soils and mobilize inorganic and organic forms of nitrogen. Sediment concentrations are also sensitive to changes in flow regulation such as dam construction (Williams and Wolman 1984) and the effects of flow dynamics on streambank erosion and the resuspension and transport of sediment, although the flow-adjustment procedures applied in this analysis, if properly specified, would be expected to account for these factors. Fecal coliform bacteria indicate human and animal sources of fecal contamination and may also be related to the pathways by which anthropogenic sources of nitrogen and phosphorus enter watersheds and streams.

Many changes occurred in the anthropogenic sources of nutrients to U.S. streams from the mid-1970s to mid-1990s that may have contributed to the observed stream nutrient trends during this period. Wastewater treatment improved considerably from the 1970s to mid-1990s (*see* Litke 1999 for a detailed summary), following passage of the Clean Water Act in 1972, and may have influenced stream nutrients in more populated watersheds at sites downstream of wastewater facilities. By 1996, primary treatment was replaced by secondary and higher levels of treatment in most plants; secondary treatment typically removes from 10% to 20% of the phosphorus in the influent (Litke 1999). Tertiary treatment, which removes as much as 99% of the phosphorus in the influent, was installed in 25% of all wastewater treatment plants by 1996. As a result, phosphorus in municipal wastewater effluent fell by as much as 50% over this period, with state bans on phosphate detergent use responsible for most of the decline. By the mid-1990s, phosphorus consumption in detergents was 10% of that in the mid-1970s.

Changes also occurred in major agricultural sources of nutrients, including fertilizers and livestock wastes, from the mid-1970s through the 1990s. Nationally, nitrogen and phosphorus fertilizer use increased severalfold from the 1960s through 1981 when use peaked. Nitrogen fertilizer use varied appreciably after 1981 but showed a net average increase of 25% over the period 1973 to 1994. By contrast, phosphorus fertilizer use showed a net average decline of a similar magnitude over this same time period. Nationally, nutrients released in livestock manure increased steadily from the 1960s to a peak in 1980 (Litke 1999). Manure nutrients declined by about 15% from 1980 to 1990, but by 1997 had increased nearly to 1980 levels. From 1982 to 1997, despite few overall changes nationally in the number of animals, the number of livestock operations declined by 24%, but the number and capacity of large farms with confined animals increased by about 25% (Kellogg et al. 2000). Regulation of these concentrated animal feeding operations (CAFOs) also increased during this period, although only about 10% of 6,600 designated CAFOs were permitted by 1995. The comparisons of changes in livestock manure and fertilizer nutrients to the stream nutrient trends in the predominantly agricultural watersheds were generally more consistent for total phosphorus where declines in both stream nutrient concentrations and agricultural nutrient sources were common at most of the sites. For total nitrogen, downtrends in stream concentrations were somewhat more frequently observed than uptrends; however, decreases were only observed in livestock manure nitrogen, whereas increases were observed in fertilizer nitrogen in all of the watersheds of the predominantly agricultural sites.

Atmospheric deposition is an important source of nitrogen to streams in the Mid-Atlantic and Northeast, representing more than 40% of the total nitrogen input to streams, but contributes less than 10% of the nitrogen in western streams (Smith and Alexander 2000). Although air emissions of nitrogen approximately doubled from 1960 to 1975 when emissions peaked, relatively little net change occurred nationally from 1975 through the mid-1990s (Howarth et al. 2002).

Improved management of farm lands occurred in selected agricultural areas of the U.S. during the study period as a result of two conservation efforts; however, the effect of these changes on stream nutrients is not well understood. Conservation tillage practices (defined as >30% residue on fields) were applied to about 35% of the acres planted in the nation in 1994, an increase of about 40% from 1989 (USDA 1997). The retirement of cropland in the Conservation Reserve Program (CRP) began in 1986 and idled approximately 15 million hectares of cropland by 1993 (~10% of cultivated land area nationally; USDA 1997). CRP lands were concentrated in the Great Plains and western Corn Belt states and showed an average reduction in soil erosion of 20% compared with conditions prior to CRP enrollment (USDA 1997). From 1982 to 1987, data from the National Resources Inventory indicated that sheet and rill erosion on rural land, including crop and rangelands, declined by 13% (Smith et al. 1993).

An assessment of potential causes of the nutrient trends (or lack of trends) may ultimately need to account for temporal lags in the response of stream nutrients and trophic conditions to changing nutrient inputs. In general, comparisons of nutrient trends with contemporaneous changes in nutrient sources and management activities are limited by uncertainties over the timing of the response of riverine concentrations to such changes. For example, older waters (>1 yr in age) may constitute an average of nearly 50% of total river flow, based on available tritium measurements at a small number of U.S. river locations (Michel 1992); the estimated mean residence times of these older waters ranged from 10 to 20 yr (Michel 1992; Focazio et al. 1997). Temporal lags, ranging from 2 to 9 yr, have been estimated in the mean-annual nitrogen flux near the outlet of the Mississippi River (McIsaac et al. 2001). Moreover, recent studies of European rivers have noted many instances where, despite very substantial declines in agricultural fertilizer use and livestock manure production, such as those evidenced after the fall of the former Soviet Union, decreases in riverine nutrient concentrations were not observed in subsequent sampling periods that ranged from 5 to 10 yr (Stalnacke et al. 2003). Detailed watershed modeling investigations of nutrient sources in relation to long-term monitoring data and estimates of water and nutrient residence times may perhaps improve understanding of the importance of time lags in explaining the observed changes (or absence of changes) in stream nutrients.

Summary—We used a trend assessment method to estimate changes in the probable trophic state (i.e., ‘‘oligotrophic,'' ''mesotrophic,'' and ''eutrophic'') at 250 monitored locations on major U.S. rivers, based on measured changes in nutrient enrichment (i.e., total nitrogen and total phosphorus concentrations) at these sites over approximately the last quarter of the 20th century. The watersheds of the monitored rivers are generally representative of the physiography, land use, and population in major U.S. river basins with drainage areas larger than about 1,000 km^2. The assessment method provides a new approach for integrating current knowledge of the trophic response of streams to nutrient enrichment, based on a recently proposed trophic-state classification system, with information on the statistical uncertainties in quantifying long-term changes in stream nutrient concentrations.

The trend assessment indicated that, despite evidence of statistically significant ($p < 0.05$) declines in nutrient concentrations at an appreciable percentage of the monitoring sites from 1975 to 1994 (37% and 44% for total nitrogen and total phosphorus, respectively), improvements in trophic state, defined as a change in the trophic-state classification (i.e., eutrophic to mesotrophic or mesotrophic to oligotrophic), occurred at only about 25% of the sites. No change occurred in the trophic-state classification at approximately 70% of the sites. As many as one-quarter to one-half of the monitoring sites, and more than 60% of the sites in predominantly agricultural and urban watersheds, were classified as eutrophic in 1994.

The trends in stream nutrient concentrations were generally consistent with those observed over the same period in turbidity, suspended sediment, and, to a lesser extent, fecal coliform bacteria, properties that may have been influenced by changes in nutrient-related sources. The declines in riverine nutrient concentrations and related improvements in trophic conditions may have been influenced by contemporaneous reductions in nutrient inputs from several major nutrient sources, including those related to wastewater treatment upgrades, phosphate detergent bans, and declines in certain agricultural sources.

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