Trends in Bioavailable Phosphorus Loading to Lake Erie

Final Report

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Trends in Bioavailable Phosphorus Loading to Lake Erie

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Abstract

Not all of the phosphorus entering Lake Erie from its tributaries is readily available to support algal growth. The portion of the phosphorus that does support algal growth is referred to as bioavailable phosphorus. In this study, trends in bioavailable phosphorus loading to Lake Erie from the Maumee, Sandusky and Cuyahoga rivers have been assessed by (1) measuring the current proportions of particulate and dissolved phosphorus that are bioavailable, (2) comparing current bioavailability with similar studies done in 1982, and (3) adjusting historical trends in total phosphorus loads to trends in bioavailable phosphorus loading. The results indicate that for the major agricultural tributaries dissolved bioavailable phosphorus, after decreasing between the mid-1970 to the mid-1990, is now at record high levels. Bioavailable particulate phosphorus has remained constant or increased slightly. <u>The net effect is that bioavailable phosphorus loading from agricultural tributaries is now a record high levels, in spite of ~20 years of effort to reduce agricultural phosphorus loading to Lake Erie.</u> Most of the increase in bioavailable phosphorus loading is for dissolved phosphorus. In contrast with the agricultural rivers, bioavailable phosphorus loading from the Cuyahoga River has decreased substantially.

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Abbreviations

P = Phosphorus
PP = Particulate Phosphorus (does not pass through a 0.45 micron membrane filter)
DP = Dissolved Phosphorus (passes through a 0.45 micron membrane filter)

T = Total
TP = Total Phosphorus
TPP = Total Particulate Phosphorus
TDP = Total Dissolved Phosphorus (formerly TSP – Total Soluble Phosphorus)

Ba = Bioavailable
BaTP = Bioavailable Total Phosphorus
BaPP = Bioavailable Particulate Phosphorus
BaDP = Bioavailable Dissolved Phosphorus

<u>Special phosphorus forms:</u>

DRP = Dissolved Reactive Phosphorus (formerly SRP – Soluble Reactive Phosphorus)
DHP = Dissolved Hydrolysable Phosphorus (formerly SHP – Soluble Hydrolysable Phosphorus and generally equivalent to BaDP)
NaOH-P = Sodium hydroxide extractable particulate phosphorus (equivalent to bioavailable particulate phosphorus)
DOP = Dissolved Organic Phosphorus (calculated as difference between TDP and DHP)

<u>Other Abbreviations:</u>

SS = Suspended Solids
FWMC = Flow-weighted mean concentration

<u>Organizations:</u>

EPA-GLNPO = Environmental Protection Agency – Great Lakes National Program Office
LEPF = Lake Erie Protection Fund
NCWQR = National Center for Water Quality Research at Heidelberg University
USGS = United States Geological Survey

I. Introduction

In the 1960's and 1970's, excessive phosphorus loading was identified as a major cause of the eutrophication problems plaguing Lake Erie. Through various modeling studies of the relationships among phosphorus loading rates, phosphorus concentrations in the lake, algal biomass in relation to phosphorus concentrations, and oxygen depletion rates, it was concluded that if the total load of phosphorus entering the Lake could be reduced to 11,000 metric tons per year, problems associated with excessive algal growth would be greatly reduced (IJC, 1978; IJC, 1980). Programs were implemented in both the United States and Canada to reduce loading to 11,000 metric tons or below. These programs focused initially on reducing point sources of phosphorus. Subsequently, programs were implemented to reduce agricultural nonpoint sources of phosphorus (U.S. Army Corps of Engineers, 1979).

The target load was met for the first time in 1981, due primarily to reductions in point source phosphorus loading. Since that time, the target load has been met every year except for those years with much larger than average tributary discharges and associated nonpoint source phosphorus runoff. In the late 1980s through the mid-1990s, Lake Erie seemed to be responding as the models had predicted, with reductions in open-water phosphorus concentrations, algal blooms and central basin hypoxia. Unfortunately, those gains in Lake Erie water quality have been reversed in the late 1990's and 2000's. Identification of the causes of this reversal has been the focus of considerable research (Matisoff and Ciborowski, 2005) and much deliberation (OEPA, 2010). Some researchers are investigating the impacts of exotic species, such as zebra mussels, which may have fundamentally altered phosphorus cycling in the lake. Others have suggested that certain phosphorus sources, such as urban storm runoff and sewage treatment plant bypasses, may have been underestimated or that climate change, with its manifold impacts on the physical conditions in the lake, may be involved. Still others have suggested that changes in bioavailable phosphorus loading may not be paralleling changes in total phosphorus loading. This grant addresses the issue of changes in bioavailable phosphorus loading as a possible contributing factor to the decline in conditions in the Lake that began in the mid-1990s.

II. Background on Bioavailable Phosphorus Loading to Lake Erie

In the early 1970's, tributary loading measurements began to indicate that nonpoint sources of phosphorus represented significant portions of the total phosphorus load entering Lake Erie (Baker and Kramer, 1973; U.S. Army Corps of Engineers, 1979). Most of this nonpoint-derived phosphorus was attached to suspended sediments (i.e., particulate phosphorus). In contrast, most of the phosphorus entering the lake from point sources was dissolved phosphorus. In order to determine the relative effectiveness of point and nonpoint source phosphorus in stimulating algal growth in Lake Erie, research was initiated to determine the bioavailability of particulate phosphorus (Logan, 1978; Logan et al, 1979; Armstrong et al, 1979; DePinto et al, 1981). Dissolved reactive phosphorus, which made up the majority of point-source-derived-phosphorus, was known to be highly bioavailable.

The above studies compared a variety of techniques to determine phosphorus bioavailability, including sequential chemical extraction, anion exchange resins, and algal bioassays. Based on these studies, a NaOH-extraction procedure was found to correlate most closely with algal uptake bioassays (DePinto et al, 1981). The NaOH- extraction procedure was applied to large numbers of storm runoff samples from tributaries to Lake Erie and Lake Ontario (Baker, 1983). The studies by Baker indicated that from 23 – 33% of the particulate phosphorus from northwestern Ohio tributaries was bioavailable. These studies were in close agreement with the studies conducted by Logan, DePinto and Armstrong, as referenced above.

While most of the measurements of bioavailability have focused on particulate phosphorus, investigations of the bioavailability of dissolved phosphorus have also taken place (Figure 1). Dissolved phosphorus includes both inorganic and organic phosphorus that passes through a 0.45 micron membrane filter. Analysis of the filtrate using EPA method 365.2 yields *dissolved reactive phosphorus* (DRP). DRP is primarily composed of ortho-phosphate, but can also contain organic phosphate forms that breakdown under the analytical conditions of the colorimetric procedure. DRP, including both the inorganic orthophosphate and organic forms, is generally considered to be 100% bioavailable.

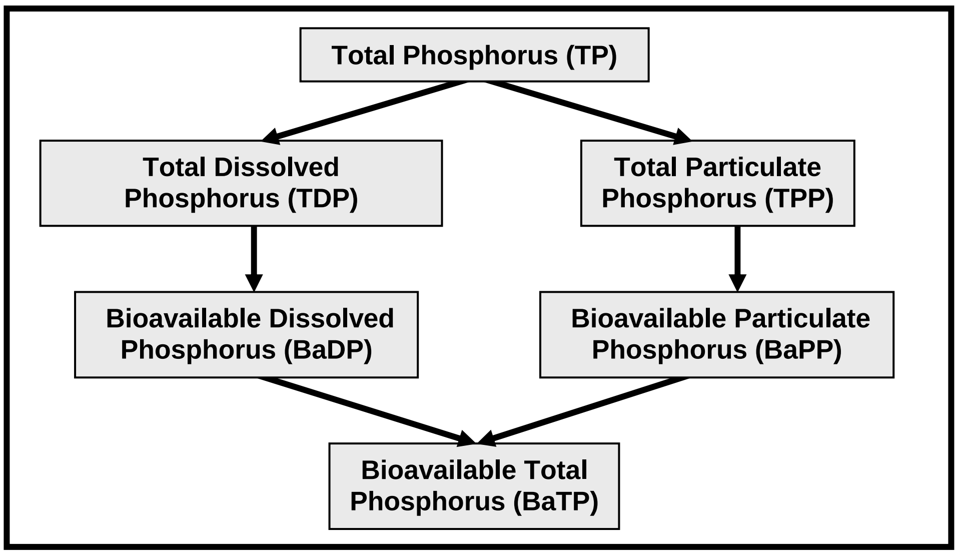

Figure 1. Major fractions of phosphorus under consideration in bioavailable phosphorus loading studies.

Analysis of the filtrate using the same procedures used to analyze total phosphorus in whole water samples (i.e., persulfate digestion, EPA Method 365.1) gives total dissolved phosphorus (TDP). The differences between TDP and DRP include both bioavailable phosphorus forms and "refractory" forms which are not readily bioavailable. In these studies, analysis of the filtrate using acid hydrolysis without persulfate is used to estimate the total bioavailable dissolved phosphorus fraction. Analysis of TDP is also needed to determine total particulate phosphorus (TPP). The latter is generally calculated by subtracting TDP from total phosphorus.

The studies of bioavailable phosphorus loading noted above were occurring at the same time that target loads for phosphorus for the Great Lakes were being established. Since the vast majority of data on phosphorus loading for the Great Lakes was limited to total phosphorus, the target loads were established in terms of total phosphorus loading. For Lake Erie, the target load was first met in 1981. By that time, major reductions in point source phosphorus loading had already occurred. Since most of the point source loading was in the form of DRP, point source reduction programs did result in major reductions of bioavailable phosphorus loading to Lake Erie. By the late 1980s and early 1990s, many of the problems associated with excessive phosphorus loading had diminished. Thus interests in issues of bioavailable phosphorus loading waned.

With the re-emergence of blue-green algal bloom and problems with attached algae including both Cladophora and Lyngbya, the issue of bioavailable phosphorus loading has once again surfaced. Since the mid-1990s, DRP loading to Lake Erie from agricultural tributaries has increased 2 to 3 fold. At the same time suspended sediment loads have decreased by about 30% and particulate phosphorus has undergone smalle decreases in loading. The increase in DRP directly represents an increase in bioavailable phosphorus loading. Although particulate phosphorus loading has decreased, it is uncertain whether the increases in DRP have been accompanied by increases in the bioavailability of particulate phosphorus. Likewise, it is uncertain whether the changes in total bioavailable dissolved phosphorus parallel the changes in DRP loading. This research project represents a pilot study regarding these issues.

This LEPF grant served as seed money for an expanded study of bioavailable phosphorus loading to Lake Erie that is being supported by a 2009 grant to the NCWQR from the EPA's Great Lakes National Program Office. Some of the initial data from that grant will be included in the results and discussion section of this report. Data from this study will be combined with data from the 2009 Bioavailability Grant for preparation of a paper for submittal to a refereed journal.

III. Study Objectives

The objectives of this study are to characterize the bioavailability of dissolved and particulate phosphorus loaded into Lake Erie during storm events from the Maumee, Sandusky and Cuyahoga rivers, and from Honey Creek, using the same methods employed in the early 1980s. The current characteristics of bioavailable phosphorus loading would then be compared to the conditions in the early 1980s. From these data, trends in bioavailable phosphorus loading from Ohio's major tributaries to Lake Erie will be developed. The results will then be conveyed to both management agencies and the scientific community through presentation at meetings and in publications.

Specific questions to be addressed include:

1. Has the proportion of total particulate phosphorus (TP) that is bioavailable (BaPP), as measured by NaOH-extraction procedures, changed from the 1980's to the current time?
2. Has the relationship between total dissolved phosphorus (TDP) and dissolved reactive phosphorus (DRP) changed between the 1980's and the current time?
3. Has the relationship between bioavailable dissolved phosphorus (BaDP), as measured by dissolved hydrolysable phosphorus, and DRP changed between the 1980's and the current time?
4. How do the above changes or lack thereof, interact with changes in dissolved reactive phosphorus loading and total particulate phosphorus loading to influence total bioavailable phosphorus export from the study watersheds.

IV. Methods

A. Analytical Procedures

The analytical procedures use to measure bioavailable phosphorus in water samples are summarized in Table 1. Each sample is analyzed for the five forms of phosphorus, with a sixth form (total particulate phosphorus) calculated from measurements of total phosphorus and total dissolved phosphorus. The relationships between these various analyses are shown in Figure 2.

Table 1. Summary of analytical procedures for various phosphorus forms used in phosphorus bioavailability studies.

Phosphorus form	Fraction analyzed	Sample Preparation/Digestion			Sample analysis
		Persulfate	Sulfuric acid	Autoclave digestion	
Total Phosphorus (TP)	Whole sample	+	+	+	Murphy-Riley colorimetric
Total Particulate P (TPP)	Calculated as TP - TDP	na	na	na	na
Total Dissolved P (TDP)	0.45 micron filtrate	+	+	+	Murphy-Riley colorimetric
Dissolved hydrolysable P (DHP)	0.45 micron filtrate	-	+	+	Murphy-Riley colorimetric
Dissolved Reactive P (DRP)	0.45 micron filtrate	-	-	-	Murphy-Riley colorimetric
NaOH Extractable P	Particulate matter on 0.45 micron nylon filter	Extract particulate matter on nylon filter with NaOH solution, neutralize and analyze as DRP			Murphy-Riley colorimetric

Additional descriptions of various phosphorus forms, either directly measured or calculated from the above measured forms, are presented below:

1. Total particulate phosphorus (TPP) -- TPP is calculated as TP – TDP. TPP is primarily phosphorus attached to inorganic sediment in storm runoff samples. Particulate mineral forms of phosphorus may also be present. TPP may also include phosphorus in algal or bacterial cells trapped on the 0.45 micron filters and/or glass fiber filters used in suspended sediment measurements.
2. Total dissolved phosphorus (TDP) -- TDP is needed to determine total particulate phosphorus. TDP can include both bioavailable and non-bioavailable dissolved phosphorus, although in agricultural tributaries, most of the TDP is bioavailable.
3. Dissolved hydrolysable phosphorus (DHP) -- DHP includes both dissolved reactive phosphorus (DRP) and an additional fraction of dissolved phosphorus that is readily hydrolysable. Both of these fractions are considered bioavailable.

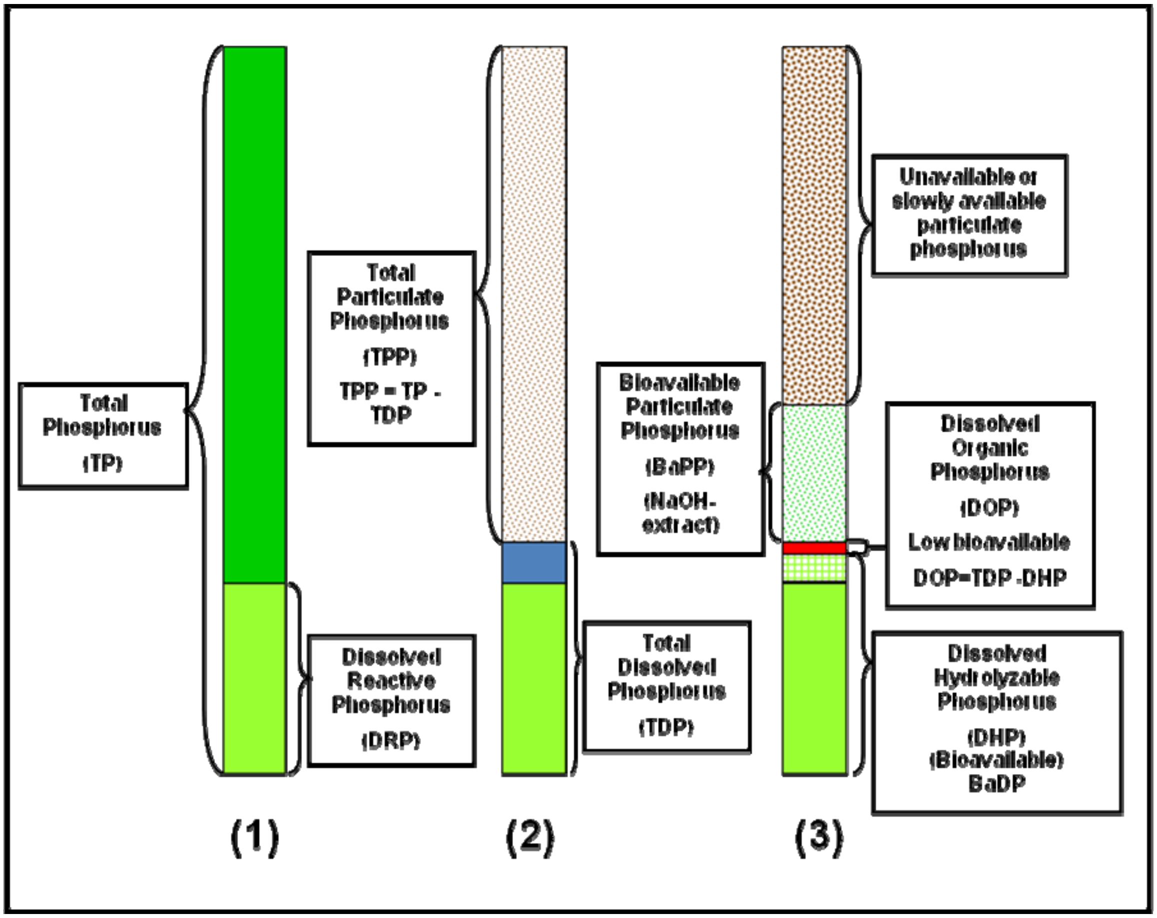

Figure 2. The relationships between the various forms of phosphorus included in phosphorus bioavailability studies.

4. Dissolved Reactive Phosphorus (DRP) -- DRP is considered to be 100% bioavailable to algae. In storm runoff samples, the majority of both the TDP and bioavailable dissolved phosphorus (BaDP) is comprised of DRP.
5. Dissolved Organic Phosphorus (DOP) -- DOP is calculated by subtracting DHP from TDP. This organic phosphorus is considered to have low bioavailability. It represents what is sometimes considered to be "refractory" phosphorus.
6. Sodium hydroxide extractable phosphorus (NaOH-P) -- NaOH-P represents the concentration of bioavailable particulate phosphorus BaPP in the sample. This portion of the particulate phosphorus can readily move into algal cells. The percentage of the total particulate phosphorus (TPP) that is bioavailable (%BaPP) can be calculated from the NaOH-P and TPP data.

The tributary monitoring programs of Heidelberg University's National Center for Water Quality Research routinely measure two of the above phosphorus forms – TP and DRP. Most monitoring programs for Great Lakes tributaries, and for point sources, measure only TP. Since the Heidelberg monitoring program does cover the major agricultural tributaries entering Lake

Erie, the existing DRP and TP data provide a unique starting point for estimating changes in bioavailable loading to Lake Erie. Our approach for estimating the total bioavailable phosphorus export from major watersheds involves measuring the three other phosphorus forms (TDP, DHP and NaOH-P) in representative samples having known TP and DRP concentrations and then extrapolating to BaDP and BaPP loading from the known loading of TP and DRP.

B. Sampling Procedures

As part of the NCWQR's tributary loading program, the NCWQR maintains refrigerated automatic samplers near the mouths of the Maumee, Sandusky, and Cuyahoga rivers and on Honey Creek. These samplers collect stream samples at 4 am, 12 noon, and 8 pm each day. Samples are returned to the NCWQR laboratories at weekly intervals. A more detailed description of the sampling procedure is contained on the NCWQR's tributary loading website (http://wql-data.heidelberg.edu/1.b.%20Sampling%20Stations%20and%20Methods.pdf).

During storm runoff events, all three samples from each day are analyzed for nutrients and sediments, while during non-storm periods, a single sample is analyzed per day. The bulk of the phosphorus loading to Lake Erie occurs during storm events. The sampling bottles contain sufficient volume to allow the analyses of bioavailable phosphorus forms on the same samples used for TP, DRP and suspended solids (SS). The added analytical costs for the bioavailable analyses described above preclude analysis of bioavailable phosphorus on all samples. Instead, selected storm event samples are chosen for analyses, with samples selected from the rising and falling limbs of the storm event. Thus, the selected samples are used to estimate the bioavailability of phosphorus for a given river system. It should be noted that each storm event represents a unique combination of watershed and river conditions interacting with unique rainfall intensity and distribution patterns. Consequently, the average suspended sediment and nutrient concentrations vary greatly from storm to storm for the same river. Also, storms at the same time can differ greatly from watershed to watershed.

V. Results

Note: Because the results section has 18 pages of graphs, these graphs have been included in a results section Appendix. This will consolidate the text materials and result in a more "readable" text section.

A. Sampling Program for 2007-08

The results of the 2007-2008 sampling program for bioavailable phosphorus forms are shown in Figures 3, 4, 5 and 6 for the Maumee River, Sandusky River, Honey Creek, and Cuyahoga River respectively. For each river, three graphs are shown: (A) dissolved reactive phosphorus as a function of total dissolved phosphorus, (B) dissolved hydrolysable phosphorus as a function of total dissolved phosphorus, and (C) NaOH-extractable phosphorus as a function of particulate phosphorus. For all graphs, linear regression lines are shown, along with the regression equation and the R-squared value. For graphs A and B, a one-to-one line is shown as a dashed line. Since the x-axis represents total dissolved phosphorus, neither the dissolved reactive phosphorus nor the dissolved hydrolysable phosphorus should exceed (lie above) the one-to-line.

For all four rivers (Figures 3-6 (A)), at least some of the dissolved reactive phosphorus values exceeded the corresponding total dissolved phosphorus value for that sample. For the Maumee and Sandusky Rivers, these results were largely limited to the lower concentration ranges (Figures 3 & 4). However, for Honey Creek and the Cuyahoga River, examples of DRP exceeding TDP occurred over a broader range of concentrations. Note that the concentration ranges differ among the four rivers. Possible explanations for occurrences where DRP exceeds TDP include the interaction of precision-associated variability for two separate measurements. The dissolved reactive phosphorus and total dissolved phosphorus analyses are done on different instruments, each with differing precisions. Where both measurements are done on the same instrument, as is the case for dissolved hydrolysable phosphorus (DHP) and total dissolved phosphorus (TDP) there are very few instances where the DHP values exceed TDP, even though the values are very close to the one-to-one line (Figures 3.B-6.B). A second possible reason for the cases where the DRP value exceeds the TDP value relates to differences in storage time within the laboratory. The DRP sample preparation and analysis are completed on a priority basis within 30 hours of arrival in the laboratory. Samples selected for TDP and DHP are generally prepared and analyzed after three full days of storage in the laboratory. The additional storage time for these samples could result in some changes in the soluble phosphorus fractions.

For the Maumee and Sandusky rivers, dissolved hydrolysable phosphorus (DHP) concentrations were very similar to total dissolved phosphorus (TDP) concentrations (Figure 3.B and 4.B). For Honey Creek (Figure 5.B), many of the DHP samples fell closely in line with the TDP samples, although there was a large cluster of samples where the DHP samples were much lower than the TDP samples. For the Cuyahoga River, there was more scatter in the DHP – TDP relationships, with the DHP values generally much lower than the TDP values (Figure 6.B).

In Figures 3.C- 6.C, the relationships between NaOH-extractable particulate phosphorus and total particulate phosphorus (TPP) concentrations are shown. In all cases, the particulate phosphorus was calculated by subtracting the total dissolved phosphorus concentrations, as shown in the “A” and “B” graphs, from the total phosphorus measurement for that sample. There was a strong correlation between the NaOH-extractable phosphorus and total particulate phosphorus for the three agricultural watersheds (Maumee – 3.C, Sandusky – 4.C and Honey Creek – 5.C) but less correlation for the Cuyahoga watershed Figure 6.C). In general, the relationships between particulate phosphorus and suspended sediments are very different in the agricultural watersheds (Maumee, Sandusky and Honey) from those in the more urban/forested watershed of the Cuyahoga River.

The relationships between the bioavailability of particulate phosphorus for individual samples, as expressed by NaOH-extractable phosphorus as a percentage of total particulate phosphorus, and stream discharge are shown in Figure 7 for all four rivers. For the Sandusky River (Figure 7.B), the percentage tended to increase as stream flow increased, while for the other three rivers, there were only small changes in the NaOH-extractable phosphorus as a percent of particulate phosphorus with increasing stream discharge. In all cases, samples at the higher stream flows contribute most to the flow-weighted concentrations of various phosphorus forms. Flow-weighted concentrations are used for extrapolation of the results of the bioavailability studies to overall loading estimates (see section V.C). It is noteworthy that the distribution of samples used for bioavailability analysis differed among the rivers, with proportionally more samples from high flow periods for the Maumee and Sandusky rivers than from Honey Creek and the Cuyahoga River. Storm flows persist for longer periods in the

Maumee and Sandusky rivers because of their larger watershed areas. Thus, the sampling regime of 3 samples per day yields more high flow samples for these two streams.

In Figure 8, the percent bioavailability of particulate phosphorus is shown as a function of particulate phosphorus concentrations for all four rivers. The Sandusky River (Figure 8.B) showed increasing bioavailability of particulate phosphorus as particulate phosphorus concentrations increased, while the Cuyahoga River showed an opposite trend (Figure 8.D.). Honey Creek and the Maumee River showed only slight changes in percent bioavailability as particulate phosphorus concentrations increased.

B. Comparisons of Current Results with Bioavailability Studies of 1980's

In Table 2, data are shown for samples analyzed for bioavailable phosphorus forms in a study at Heidelberg College supported by EPA-GLNPO in 1982. The table presents the flow-weighted concentrations of suspended sediments (SS), TP, DRP, TDP, DHP, NaOH-P and PP for samples from the same four rivers included in the current study. The data also include values for DRP as a percent of TDP and NaOH-P as a percent of PP. Forty samples were analyzed from each of the four rivers.

In Table 3, data for samples collected in the current study are presented in the same format as for Table 2. Again, flow-weighted-mean concentrations are presented. In Table 4, preliminary data from the on-going EPA-GLNPO Bioavailability Study are presented. The Table 4 data differ from that of Tables 2 & 3 in that the Table 4 concentration data represent average concentrations rather than flow-weighted-mean concentrations.

In comparing the flow-weighted concentrations of suspended sediments and phosphorus forms between the 1982 studies (Table 2) and the 2007-2008 studies (Table 3), the following similarities and differences are evident:

1. Suspended solids – For the Sandusky River and Honey Creek the SS concentrations were much higher in the 1982 studies than in the current study. For the Maumee River, SS concentrations were very similar during the two periods. For the Cuyahoga River, SS concentrations were much higher in the recent studies. These differences are likely due to characteristics of the individual storm runoff conditions available for analysis of bioavailable P forms rather than a reflection of longer-term trends in SS concentrations in the rivers.
2. Total phosphorus – The differences in TP concentrations among stations and periods paralleled those for SS. However the differences in TP concentrations were "muted" relative to the differences in SS concentrations because as SS concentrations increase, the ratios of particulate phosphorus to suspended sediment decrease. As suspended sediment concentrations increase, generally the average particle size composition also increases, resulting in a lower proportion of binding sites for phosphorus (i.e., relatively less clay) in the sample.
3. Dissolved reactive phosphorus – The DRP concentrations increased substantially in the three agricultural watersheds between the 1982 studies and the current studies. For the Cuyahoga River, the DRP concentrations decreased. These changes likely reflect the long-term trends in DRP concentrations at these stations.

Table 2. Flow-weighted mean concentrations of suspended sediments and phosphorus forms during bioavailability studies, 1982.*

River (Sample count)	SS mg/L	TP mg/L	DRP mg/L	TDP mg/L	DHP mg/L	NaOH-P mg/L	PP mg/L	DRP as % TDP	NaOH-P as % PP
Maumee (40)	329	.556	0.070	0.092	0.084	0.134	0.464	76.1%	28.6%
Sandusky (40)	775	.919	0.054	0.074	0.067	0.244	0.845	73.0%	28.8%
Honey Creek (40)	858	1.101	0.042	0.073	0.061	0.316	1.028	57.5%	30.7%
Cuyahoga (40)	434	.570	0.076	0.140	0.134	0.167	0.430	54.3%	38.8%

- Taken from Tributary Loading of Bioavailable Phosphorus into Lake Erie and Ontario, Draft Final Report, Part III, U.S. EPA Grant No. R005708-01, submitted to Great Lakes National Program Office, U.S. Environmental Protection Agency, Region V, 536 South Clark Street, Chicago, IL 60605 ,Table 4, prepared by David B. Baker, Heidelberg College, April 1983.

Table 3. Flow-weighted mean concentrations of suspended sediments and phosphorus forms during bioavailability studies, April 25, 2007 through February 10, 2008.

River (Sample count)	SS mg/L	TP mg/L	DRP mg/L	TDP mg/L	DHP mg/L	NaOH-P mg/L	PP mg/L	DRP as % TDP	NaOH-P as % PP
Maumee (37)	339	0.687	0.127	0.137	0.133	0.146	0.550	92.7%	26.5%
Sandusky (50)	327	0.696	0.124	0.142	0.136	0.176	0.554	87.3%	31.8%
Honey Creek (76)	188	0.524	0.152	0.176	0.157	0.119	0.347	86.3%	34.3%
Cuyahoga (45)	979	0.773	0.040	0.038	0.029	0.130	0.632	105%	20.6%

Table 4. Average concentrations of suspended sediments and phosphorus forms during GLNPO Bioavailability Study, 2009. Preliminary Data - Flow data not available at time of data review.

River (Sample count)	SS	TP	DRP	TDP	DHP	NaOH-P	PP	DRP as % TDP	NaOH-P as % PP
Maumee (12)	114	0.328	0.086	0.095	0.092	0.051	0.247	90.5	20.6
Sandusky (26)	306	0.559	0.089	0.105	0.096	0.139	0.458	84.7	30.3
Cuyahoga	917	0.660	0.065	0.069	0.065	0.204	0.579	94.2	35.2

(17)									

4. Total dissolved phosphorus – The changes in TDP concentrations for the three agricultural watersheds increased in parallel with the increases in DRP between the 1982 studies and the current studies. However, DRP as a percent of TDP increased for these three watersheds between the two studies. This suggests that among the dissolved phosphorus fractions, the increases between the two periods consisted primarily of increases in DRP. For the Cuyahoga River, the TDP concentrations decreased between the two study periods. During the 1982 period, TDP almost doubled the DRP concentrations, while in the current study TDP concentrations were essentially the same as the DRP concentrations.
5. Dissolved hydrolysable phosphorus – The DHP concentrations in the three agricultural watersheds fell between the DRP and the TDP concentrations for both periods. Thus the DHP concentrations were much higher during the recent studies than during the 1982 studies. For the Cuyahoga River, DHP was very similar to TDP in the 1982 study, but during the current study DHP was less than both the TDP and DRP.
6. Sodium hydroxide extractable P – The concentrations of NaOH-P for the agricultural watersheds largely parallel the concentrations of total phosphorus for study periods. Thus the concentrations of NaOH-P were higher in the 1982 study than in the current study. NaOH-P, as a percentage of particulate phosphorus (PP), has shown little change between the two study periods for the agricultural watersheds. In the case of the Cuyahoga River, NaOH-P as a percent of PP was much lower during the recent study.
7. Particulate phosphorus – The flow-weighted concentrations of PP are equivalent to the TP concentrations minus the TDP concentrations. As such, the differences in PP concentrations between the two study periods parallel the changes in TP and TDP for all four stations.

In Table 4, preliminary data from the current EPA-GLNPO study are presented. These results generally are consistent with those of the 2007-2008 study. However, the samples from the three rivers had very different average suspended-sediment concentrations (114, 306 and 917 mg/L respectively for the Maumee, Sandusky and Cuyahoga rivers).

C. Extrapolation of Bioavailability Study Results to Trends in Bioavailable Phosphorus Export at River Transport Stations.

A final objective of bioavailable phosphorus studies is to provide estimates of bioavailable phosphorus export from watersheds draining into Lake Erie. These estimates extend to the examination of trends in bioavailable phosphorus export that may be linked to changing conditions in Lake Erie. As noted in Section IV, the NCWQR's long-term tributary loading program provides detailed information on total phosphorus and dissolved reactive phosphorus loading for each station. Data from the bioavailability studies allow adjustments of the TP and DRP loads to the various bioavailable phosphorus forms, as indicated in Figure 2.

The data from Tables 2 and 3 provide information for the conversion factors shown in Table 5. The conversion of DRP to TDP allows calculation of particulate phosphorus by subtraction of TDP from TP. The conversion of DRP to DHP represents a direct conversion to bioavailable dissolved phosphorus (DHP ~ BaDP). The conversion of particulate phosphorus to bioavailable particulate phosphorus is based on the calculation of NaOH-P as a percent of PP.

Table 5. Factors used for converting DRP and TP to bioavailable loading estimates.

River	Date of bioavailability study	Conversion Factor (multiplier)		
		DRP to TDP	DRP to DHP	PP to BaPP
Maumee River	1982	1.31	1.20	0.286
	2007-2008	1.08	1.05	0.265
Sandusky River	1982	1.37	1.24	0.288
	2007-2008	1.15	1.10	0.318
Honey Creek	1982	1.74	1.45	0.307
	2007-2008	1.16	1.03	0.343
Cuyahoga River	1982	1.84	1.76	0.388
	2007-2008	1.00*	1.00*	0.206

- Conversion factors less than one were set equal to one since theoretically TDP and DHP cannot be less than DRP. The conversion factors based on data in Table 3 would be 0.95 and 0.72.

The conversion factors that show the least amount of change between the two study periods are for NaOH-P as a percent of PP (PP to BaPP). These factors are very similar among the three agricultural rivers and show only small changes between the two time periods. For the Cuyahoga River, the PP to BaPP conversion factor in 1982 was about twice as high as in 2007-2008. The flow-weighted sediment concentration in the Cuyahoga was very high during the 2007-2008 period, possibly accounting for the lower PP-BaPP conversion factor for that year.

The conversion factors for dissolved phosphorus forms (DRP to TDP and DRP to DHP) have changed more between the two time periods and show more variation from river to river. In general the dissolved phosphorus conversion factors are similar for the Maumee and Sandusky Rivers but are more variable for Honey Creek and the Cuyahoga River.

In using these conversion factors for estimating bioavailable phosphorus export from these watersheds, selection of the time period for application of the two conversion factors is arbitrary. Using the two points in time (1982 and 2007-2008) to calculate conversion factors that change continuously with time could be done, but it is unlikely that the changes in bioavailability would behave in that manner. To simplify the adjustments, the conversion factors from the 1982 study have been applied to the loading data through the 1994 water year and the conversion factors from the 2007-2008 water year studies have been applied to the 1995 and more recent water years. The 1994 to 1995 period represents the midpoint in the period when flow-weighted DRP concentrations switched from decreasing to increasing for the agricultural watersheds.

For each river nine graphs are shown. These graphs include:

A. Annual discharge – Loading of suspended sediments and nutrients from agriculture is strongly impacted by annual rainfall amounts and patterns and associated discharge.
B. Annual flow-weighted mean concentrations of total phosphorus – Flow-weighted mean concentrations are less variable than either discharge or loads and consequently are better suited for trend analysis.
C. Annual loads of total phosphorus.
D. Annual flow-weighted mean concentrations of dissolved reactive phosphorus.
E. Annual loads for dissolved reactive phosphorus.
F. Annual loads of particulate phosphorus (calculated as TP load minus TDP load by year).
G. Bioavailable particulate phosphorus (BaPP) loads.
H. Bioavailable dissolved phosphorus (BaDP) loads.
I. Bioavailable total phosphorus (BaTP) loads (sum of BaPP and BaDP loads).

The above graphs are shown in Figures 9, 10, 11 and 12 for the Maumee River, the Sandusky River, Honey Creek and the Cuyahoga River respectively. For each graph, linear trends are fitted to the loads or discharges, except for the loads of dissolved phosphorus which are clearly comprised of decreasing followed by increasing trends.

1. Annual Discharge -- Figures 9.A, 10.A, 11.A and 12.A show the annual discharge by water year for the period of record for nutrient and sediment export. For all four rivers, the annual discharge has increased during the period of record, although the discharge shows considerable variation from year to year. The general increasing trend in discharge for Lake Erie tributaries has been confirmed by the USGS (personal communication from Dan Button to the Ohio Lake Erie Task Force). The general correspondence between variations in discharge and variations in total phosphorus loads ("C" graphs) illustrates the dominant role of weather in determining total phosphorus export in any particular year. Consequently, consideration of trends in annual discharge is an essential component of examination of trends in nonpoint pollutant export.

2. Total Phosphorus Flow-weighted Mean Concentrations (FWMCs) -- The annual flow-weighted mean concentrations of total phosphorus are shown in the "B" graphs of Figures 9-12. For all of the streams, the TP flow-weighted mean concentrations decrease, although the decreases were very small for the Sandusky River and Honey Creek. Flow-weighted mean concentrations are calculated by dividing the total load of a chemical by the total discharge of water during a specified time interval, in this case for a water year. The annual discharge is based on continuous stage and discharge calculations by the USGS at transport stations while the annual loads are based on analyses of 450 to 550 storm samples at the transport station as part of the NCWQRs. Procedures for calculating flow-weighted mean concentrations are described at Heidelberg's tributary loading website (http://wql-data.heidelberg.edu/2.d.%20Time-weighted%20and%20Flow-weighted%20Mean%20Concentrations.pdf). It should also be noted that trends in TP FWMCs reflect a composite of trends for FWMCs of TDP and PP.

3. Total Phosphorus Loads – Total phosphorus loads are shown in the "C" graphs of Figures 9-12. Total phosphorus loads have remained relatively constant for the Maumee River, increased slightly for the Sandusky River and Honey Creek, and decreased for the Cuyahoga River.

4. Dissolved Reactive Phosphorus Flow-weighted Mean Concentrations – The DRP FWMCs for the three agricultural tributaries all show substantial decreases from the onset of the monitoring program through the mid-1990s (Figures 9.D, 10.D and 11.D). Since the mid-1990s, DRP FWMCs have increased dramatically matching their highest levels for the Maumee River and providing period-of-record highs for DRP for the Sandusky River and Honey Creek. For the Cuyahoga River, DRP FWMCs decreased very rapidly from the early 1980s through the early 1990s then increased through 2004 after which it has remained relatively constant (Figure 12.D). In the Cuyahoga River, DRP is largely derived from point sources, and trends in DRP concentrations largely reflect trends in point source discharges.

5. Dissolved Reactive Phosphorus Loads – The loads of dissolved reactive phosphorus are shown in the "E" graphs of Figures 9-12. For all three agricultural tributaries, the loads decreased from the onset of monitoring through the mid-1990s after which they increased rapidly, reaching record high DRP loads in the 2007 and 2008 water years. The causes of the increasing DRP loads have been the subject of much discussion within the Ohio Lake Erie Task Force (http://www.epa.state.oh.us/dsw/lakeerie/ptaskforce/index.aspx) and are outlined in the final report of the task force (http://www.epa.state.oh.us/portals/35/lakeerie/ptaskforce/Task_Force_Final_Report_April_2010.pdf).

The trends in DRP loads in the Cuyahoga River differ substantially from those of the agricultural watersheds. While the DRP loads in the agricultural tributaries show considerable year-to-year variability associated with weather conditions, the loading for the Cuyahoga River shows much less year-to-year variability. The dominance of point sources, which have relatively constant annual loads, accounts for the low weather-related effects in the Cuyahoga. The "smooth" changes over the years reflect variations in phosphorus loading and/or removal at the sewage treatment plants. The causes of the apparent increases in DRP loads in the Cuyahoga River between 1995 and 2004 have yet to be determined. Note that the absolute magnitude of the DRP loads from the Cuyahoga River is smaller than the loads from the Maumee and Sandusky rivers, and more comparable to the DRP export from Honey Creek, a relatively small agricultural sub-watershed of the Sandusky Watershed (see the y-axis scales of the "D" graphs).

6. Particulate Phosphorus Loads – The particulate phosphorus loads are shown in the "F" graphs of Figures 9-12. Particulate phosphorus loads are calculated by subtracting the total dissolved phosphorus loads (not shown) from the total phosphorus loads. Particulate phosphorus is phosphorus attached to soil particles or particulate minerals containing phosphorus generally associated with soils. The particulate phosphorus loads of the Maumee River (Figure 9.F) have decreased slightly over the period of record, while particulate phosphorus loads for the Sandusky River (Figure 10.F) and Honey Creek (Figure 11.F) have increased slightly. These increases or lack of decreases have occurred in spite of concerted effort by the agricultural community to reduce erosion and associated export of particulate phosphorus. Increases in stream discharge during the study period likely have contributed to the increases in particulate phosphorus loads.

Particulate phosphorus loads have also increased in the Cuyahoga River (Figure 12.F). Particulate phosphorus loads are affected by the conversion factors used to adjust DRP loads to total dissolved phosphorus loads. For all four rivers, the conversion factor decreased between 1982 and 2007-8, but the decrease was especially large for the Cuyahoga River (Table 5). Since TDP is subtracted from TP to yield PP, these differences in conversion factors have the effect of decreasing the particulate phosphorus prior to 1994 relative to 1995 and beyond.

7. Bioavailable Particulate Phosphorus Loads – The BaPP loads are shown in the “G” graphs of Figures 9-12. For the agricultural tributaries, the loading trends are very similar to those of the trends for particulate phosphorus, with a slight decrease in the Maumee River (Figure 9.G) and slight increases in the Sandusky River (Figure 10.G) and Honey Creek (Figure 11.G). For the agricultural tributaries, the factors for converting PP to BaPP (Table 5) did not change much between the two time periods. These conversion factors ranged from 0.265 to 0.343. For the Cuyahoga River the conversion factor from the 1982 study was 0.388 while the conversion factor for the recent period was 0.206. This resulted in an apparent large decrease in the BaPP loading from the Cuyahoga River (Figure 12.G).

8. Bioavailable Dissolved Phosphorus Loads – Trends in BaDP loads are shown in the ”H” graphs of Figures 9-12. These graphs reflect the trend graphs for DRP (“E” graphs), as adjusted by the conversion factors shown in Table 5. For all for rivers, the conversion factors were larger in the 1982 study than in the 2007-2008 study. This has the effect of increasing the BaDP levels in the early part of the study relative to the more recent years. Nevertheless, for the agricultural tributaries, the highest BaDP loading has occurred in the post-2000 time period. For the Cuyahoga River, the large decrease in the DRP to DHP conversion factor between the two time periods (Table 5) resulted in an even greater decrease in BaDP loading than the decrease in DRP loading (compare Graphs 12.H with 12.E).

9. Bioavailable total phosphorus loads – Trends in loads of bioavailable total phosphorus are shown in the “I” graphs of Figures 9-12. The bioavailable total phosphorus loads are the sums of the loads of bioavailable particulate and bioavailable dissolved phosphorus. For all three agricultural tributaries, the bioavailable total phosphorus loading reached its highest levels over the period-of-record during the post-2000 time periods. In contrast, for the urban-dominated Cuyahoga watershed, the BaTP loads during the mid-1980s were much higher than during the post-2000 period.

VI. Conclusions

Differences between trends in total phosphorus loading and trends in bioavailable phosphorus loads result from the following characteristics:

1. Particulate phosphorus comprises the majority of total phosphorus exported from tributaries.
2. Particulate phosphorus has much lower bioavailability than dissolved phosphorus.
3. The export of particulate phosphorus and dissolved phosphorus are largely independent of one another.
4. Dissolved reactive phosphorus, which comprises most of the total dissolved phosphorus, has trend patterns very different than the trends for particulate phosphorus.
5. Annual loads of both particulate and dissolved phosphorus are greatly impacted by weather conditions and annual discharge rates.

6. Changes in bioavailability of various phosphorus fractions between 1982 and 2007-8 do have impacts on estimates of trends in bioavailability, but these impacts are secondary to the major shifts in dissolved reactive phosphorus export from agricultural watersheds.
7. The proportion of the particulate phosphorus that is bioavailable changed by only 2-4% for individual agricultural watersheds between the two time periods.
8. For the agricultural watersheds, as the concentrations of DRP increased, DRP also increased as a percentage of total dissolved phosphorus between the two time periods.
9. Although agricultural pollution abatement measures have reduced suspended sediment concentrations over the study period (Richards,), reductions in particulate phosphorus concentrations have been smaller since the pollution abatement measures selectively reduce large particle size sediments that have lower phosphorus to sediment ratios.
10. The agricultural production systems that currently dominate the Maumee and Sandusky watersheds involve no-till and reduced till, along with broadcast fertilizer applications, both of which lead to increases in dissolved reactive phosphorus loading.

The net effects of the above set of characteristics and changes is that bioavailable phosphorus export from the Maumee and Sandusky rivers, while decreasing slightly through the mid-1990s, is now at the highest levels that have been observed during the monitoring period which extends back to the 1970s. Since the Maumee and Sandusky rivers are major sources of phosphorus for the western basin of Lake Erie, these high loads of bioavailable phosphorus are likely to be contributing to the reemergence of eutrophication problems in the Western Basin of Lake Erie (OEPA, 2010).

Even the particulate phosphorus loading, which has been the target of agricultural pollution abatement programs in Northwestern Ohio, have not shown decreases. Although annual flow-weighted mean concentrations of suspended sediment have decreased, the decreases in FWMCs of particulate phosphorus have been smaller. The erosion control practices adopted by farmers (no-till and reduced till, buffer strips, CRP) have been more effective in reduced large particle size sediments than the finer sediments. The finer sediments have higher phosphorus sediment ratios than do higher concentrations sediments with lower average particle sizes. The effectiveness of the erosion control measures have also been undercut by the increasing discharges of the rivers. While the effectiveness of agricultural pollution abatement measures can be corrected for variations in discharge, the actual loads that Lake Erie receives dictate lake responses.

The major changes in bioavailable loading to Lake Erie reflect the increasing dissolved phosphorus runoff from cropland under the prevailing management practices. Major changes in nutrient management will be necessary to reduce bioavailable phosphorus loading to the Lake.

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Appendix 1. Data Graph Figures for Final Report of LEPF Grant 315-07

May 24, 2010

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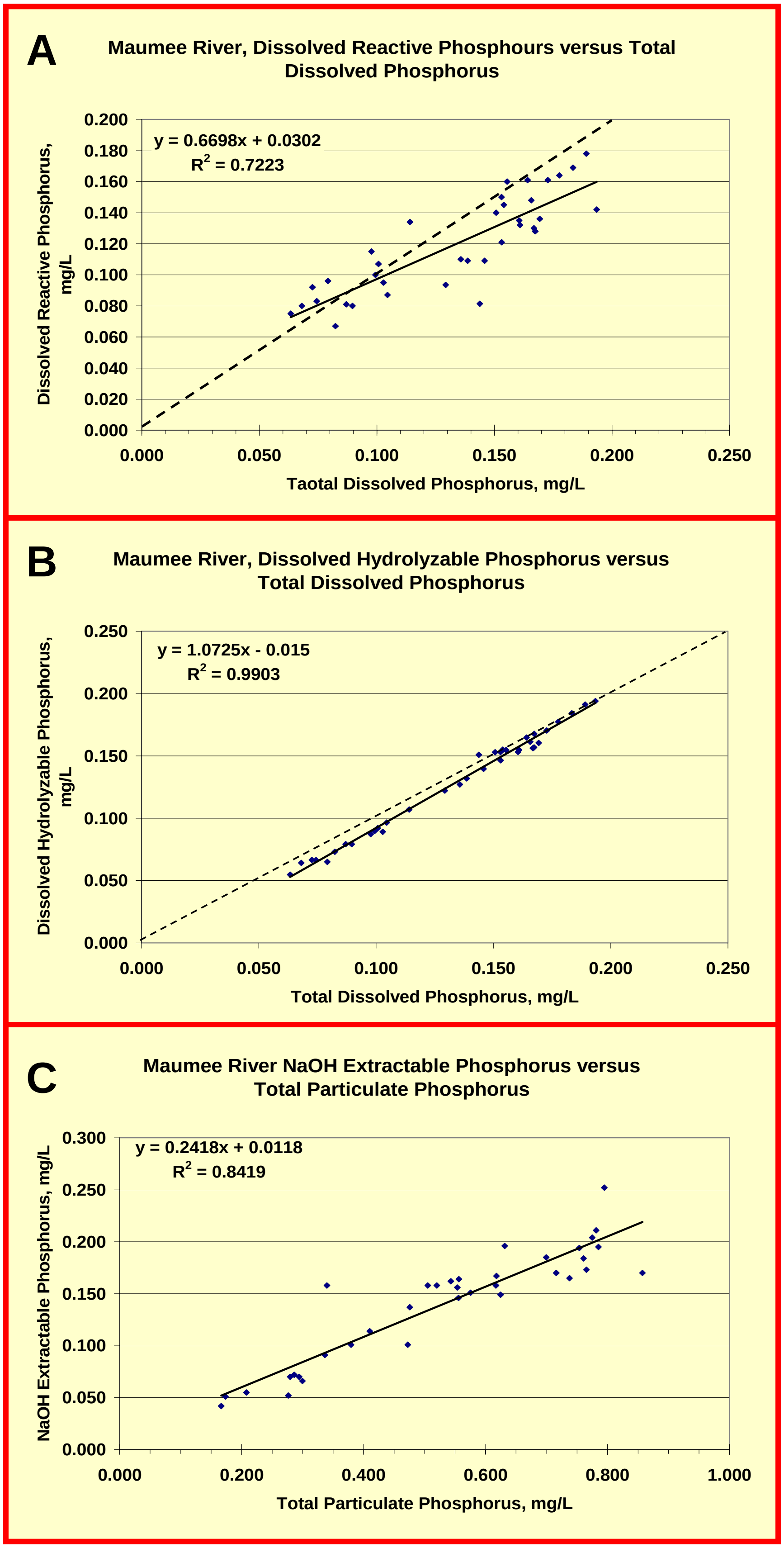
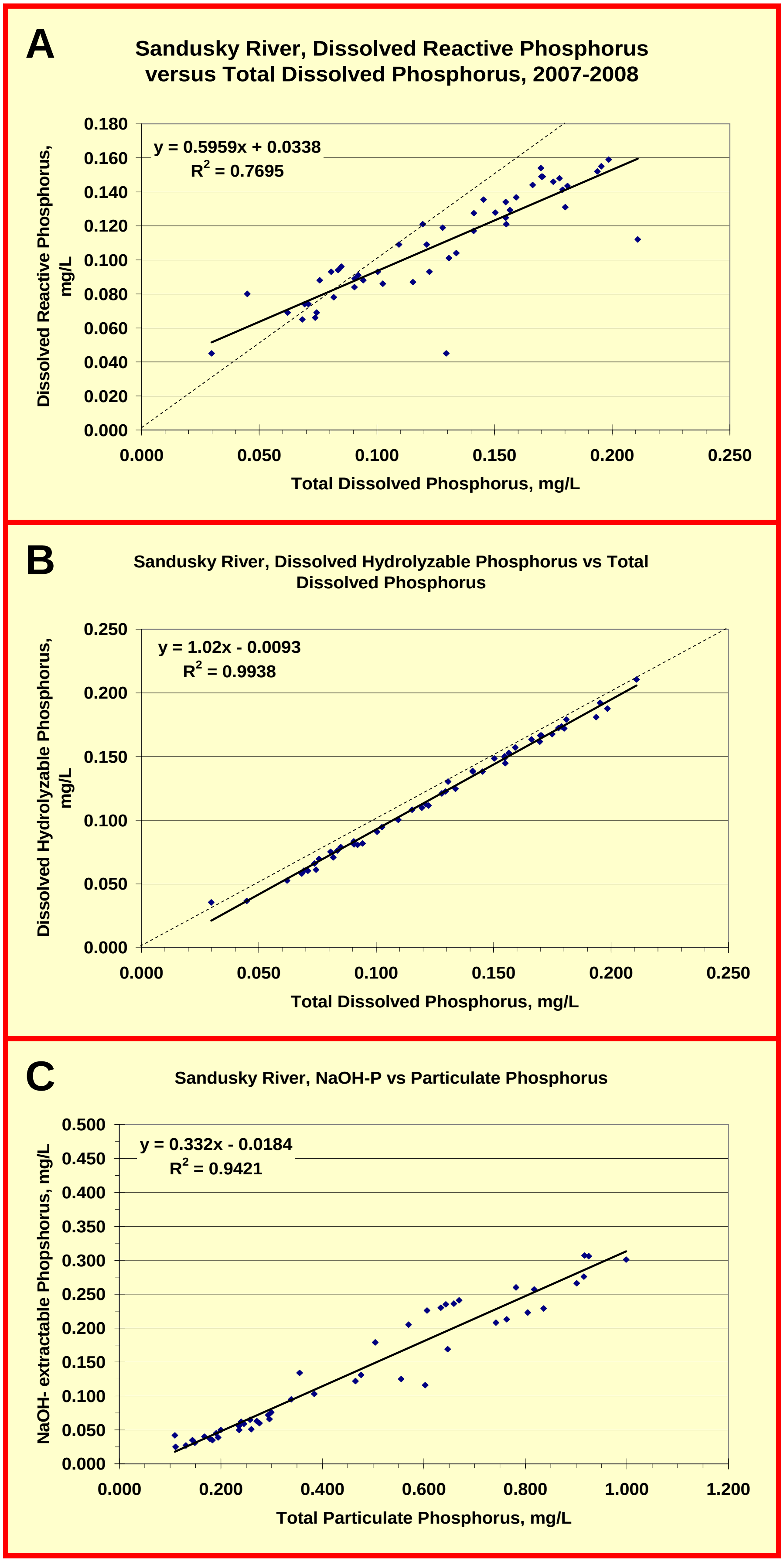


Figure 3. Analytical results for bioavailable phosphorus forms for the Maumee River. The relationship between (A) dissolved reactive phosphorus and total dissolved phosphorus, (B) dissolved hyrolysable phosphorus and total dissolved phosphorus and (C) NaOH-extractable phosphorus and total particulate phosphorus.



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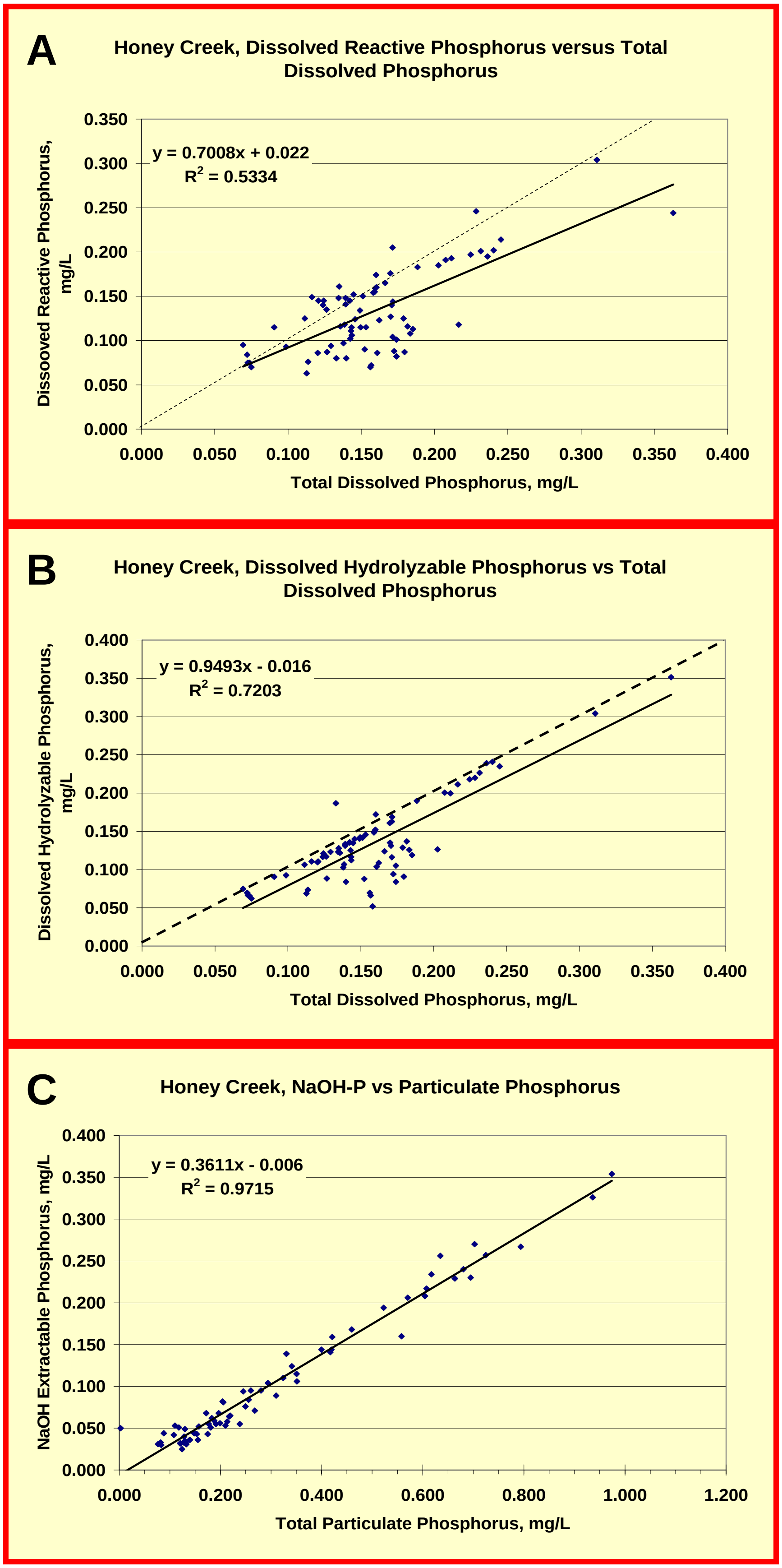

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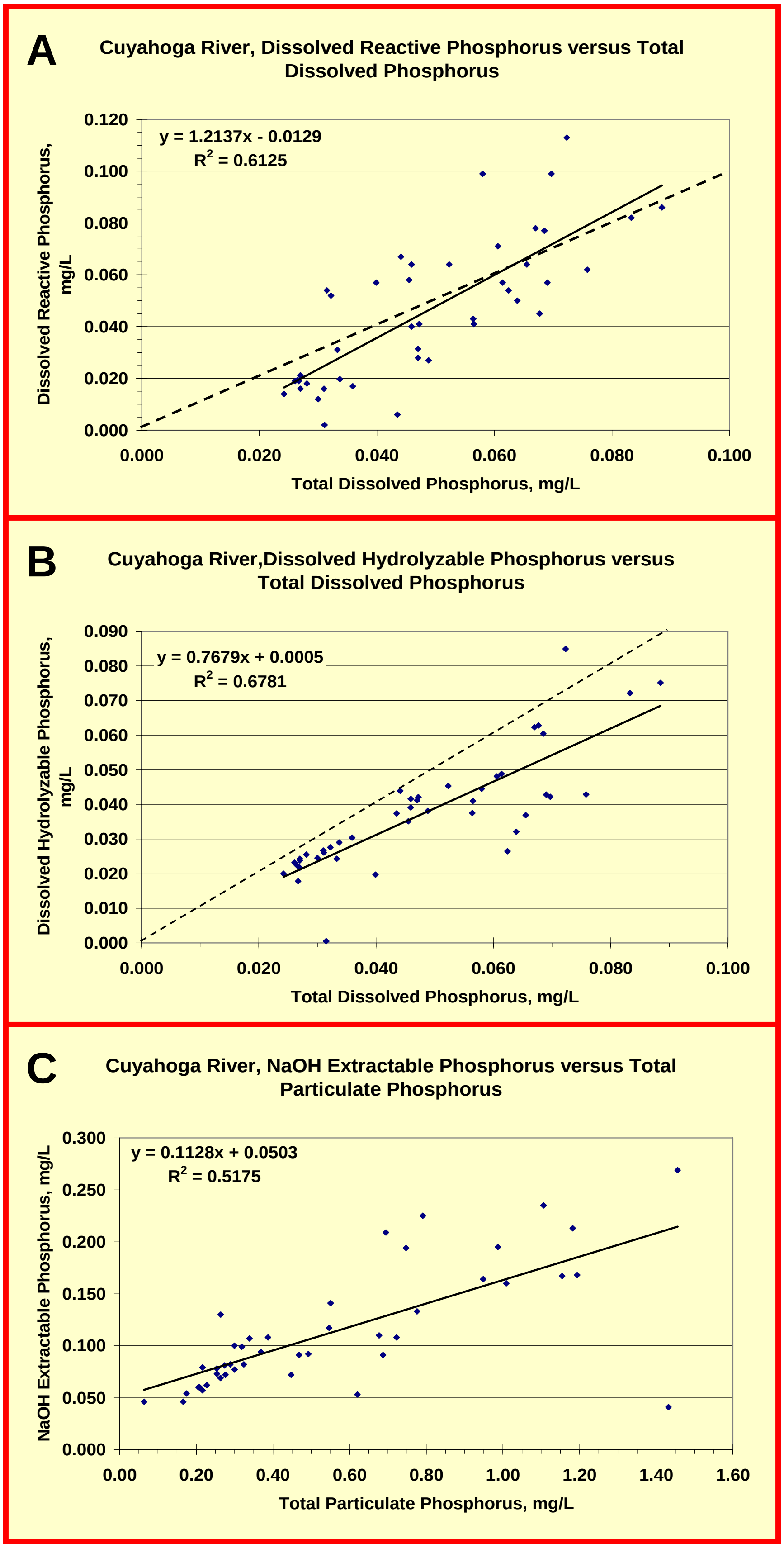


Figure 6. Analytical results for bioavailable phosphorus forms for the Cuyahoga River. The relationship between (A) dissolved reactive phosphorus and total dissolved phosphorus, (B) dissolved hyrolysable phosphorus and total dissolved phosphorus and (C) NaOH-extractable phosphorus and total particulate phosphorus.

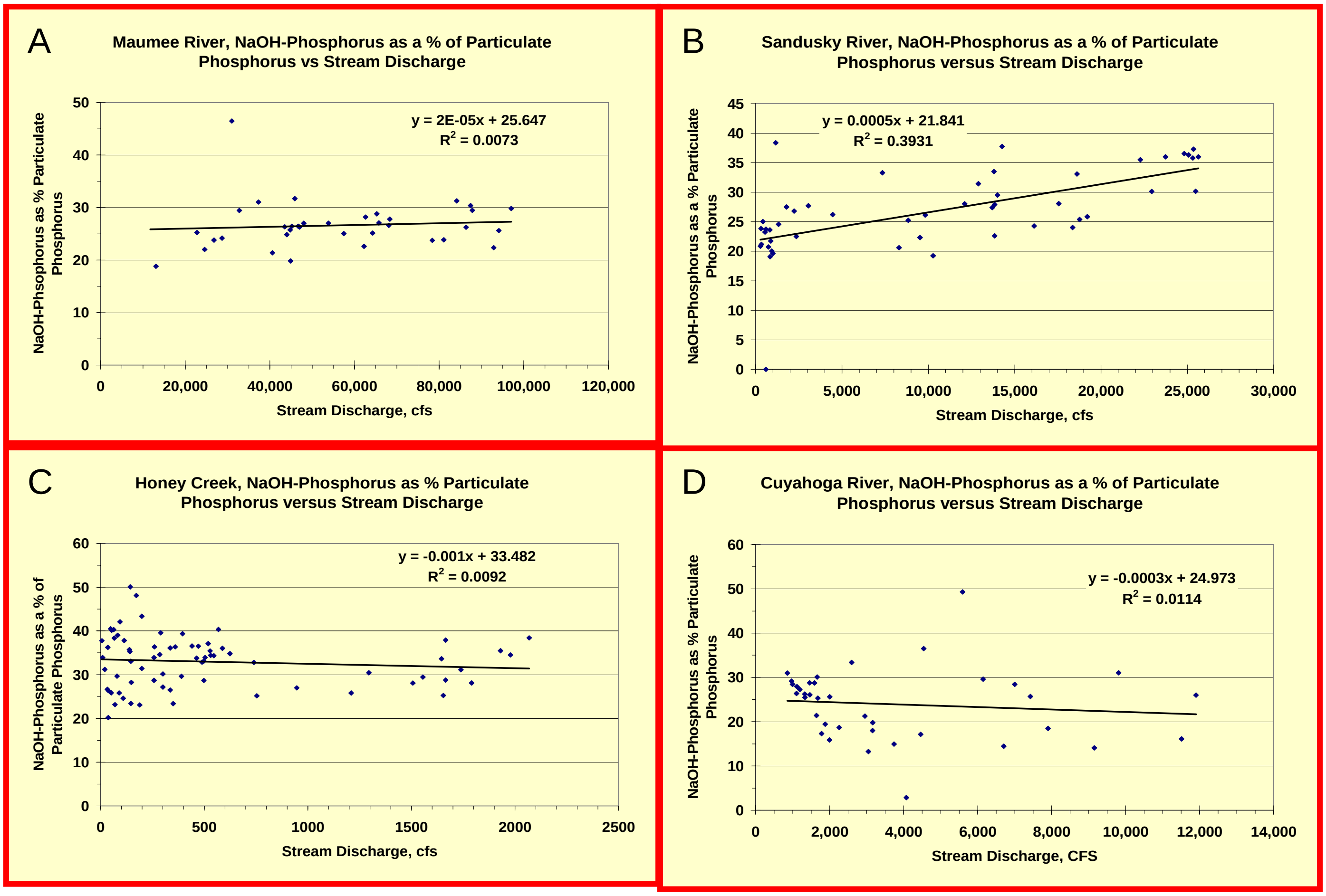
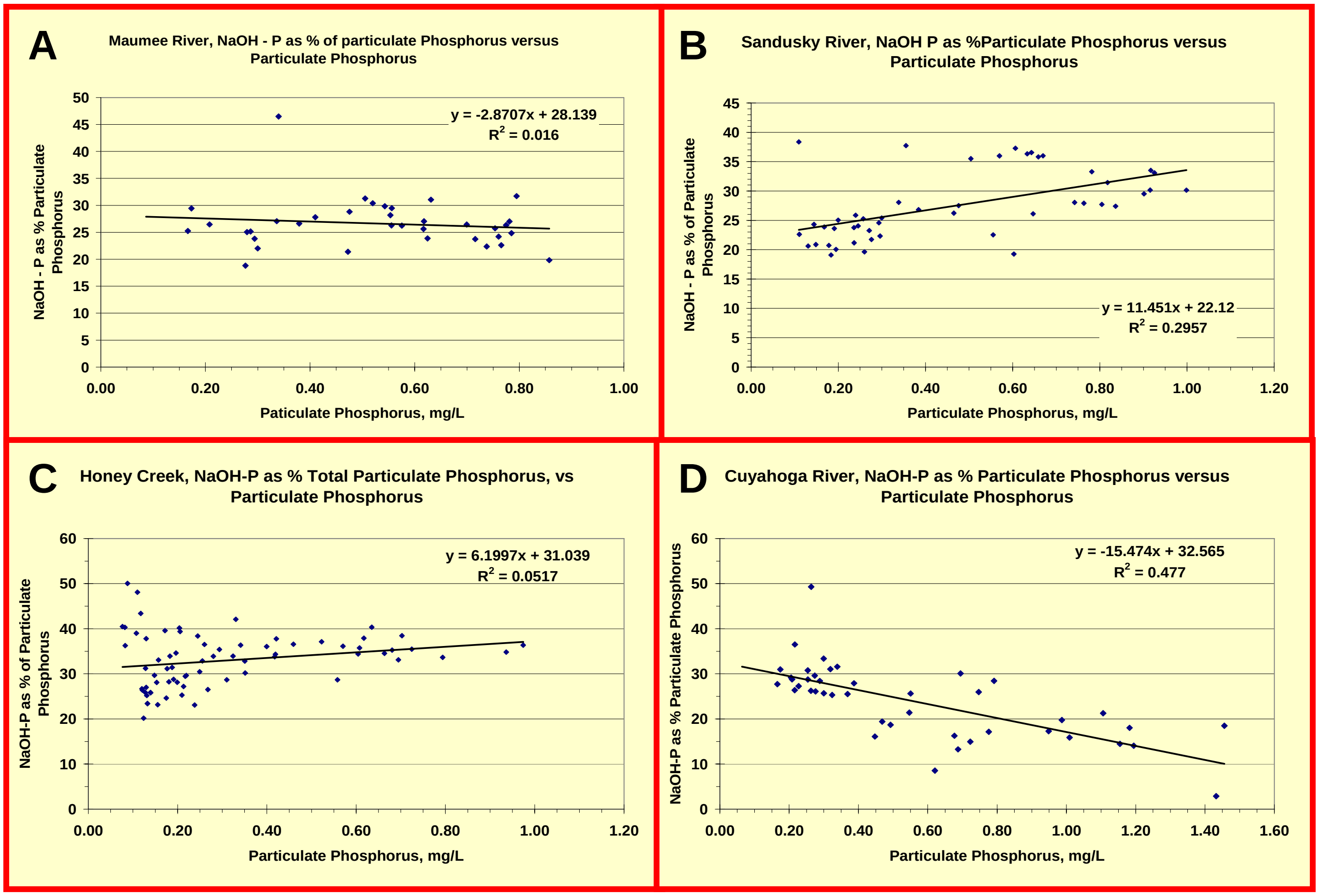


Figure 7. Relationships between percentage bioavailability of particulate phosphorus and stream discharge for the Maumee River (A), the Sandusky River (B), Honey Creek (C), and the Cuyahoga River (D).



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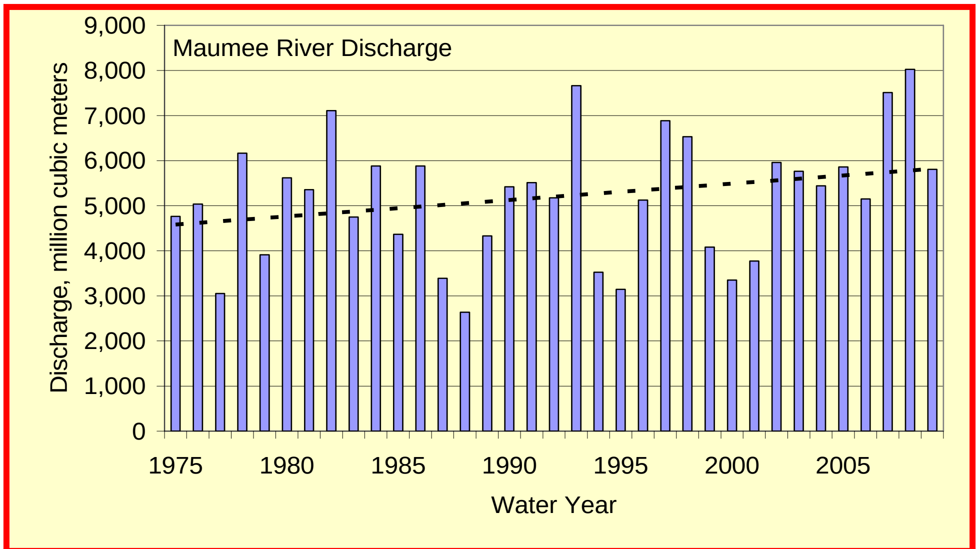


Figure 9.A. Annual discharges for the Maumee River for the period of record.

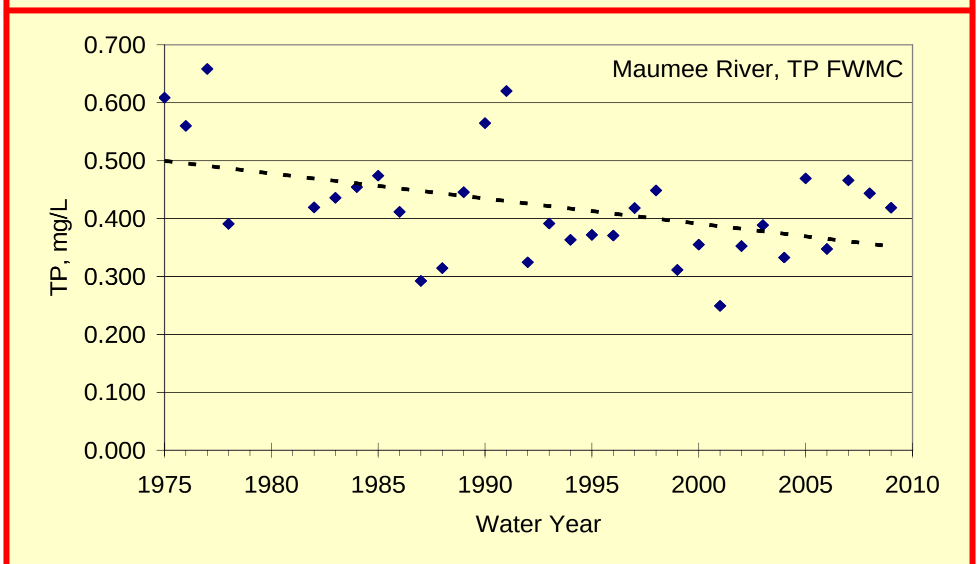
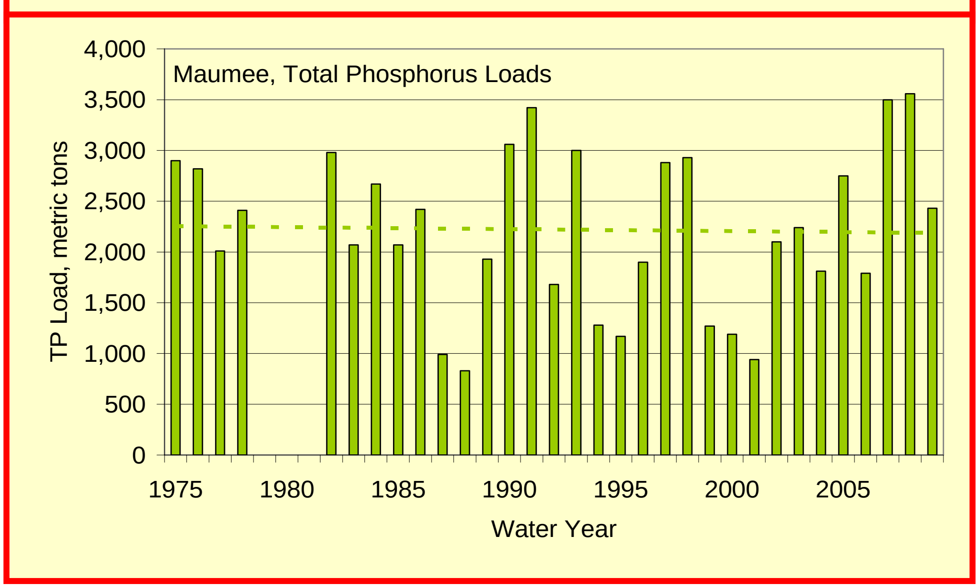


Figure 9.B. Annual flow-weighted mean concentrations of total phosphorus for the Maumee River for the period of record.



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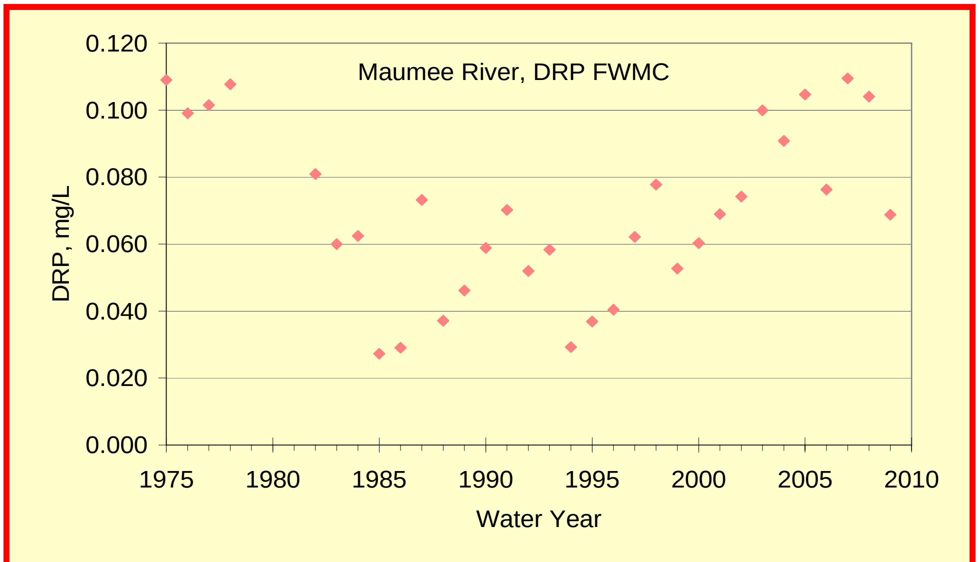

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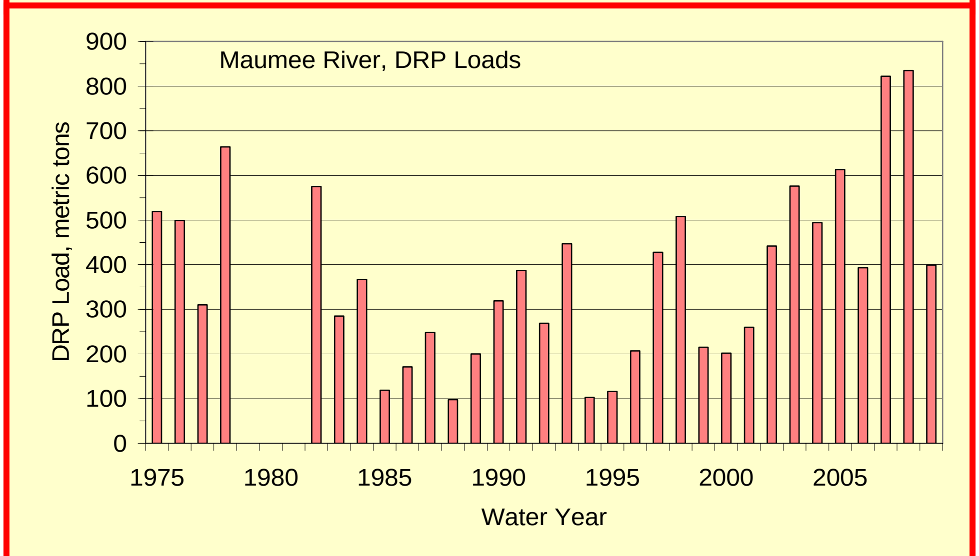

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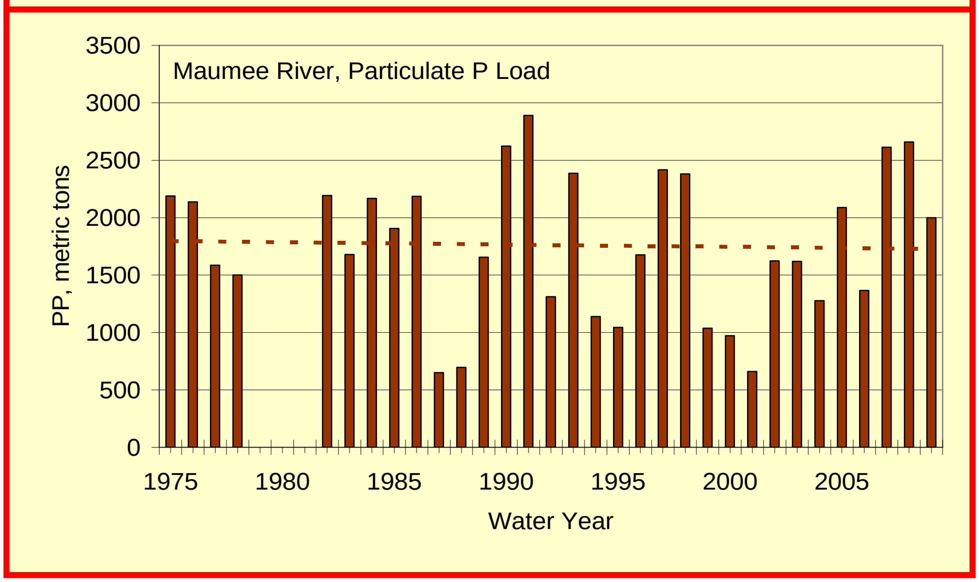

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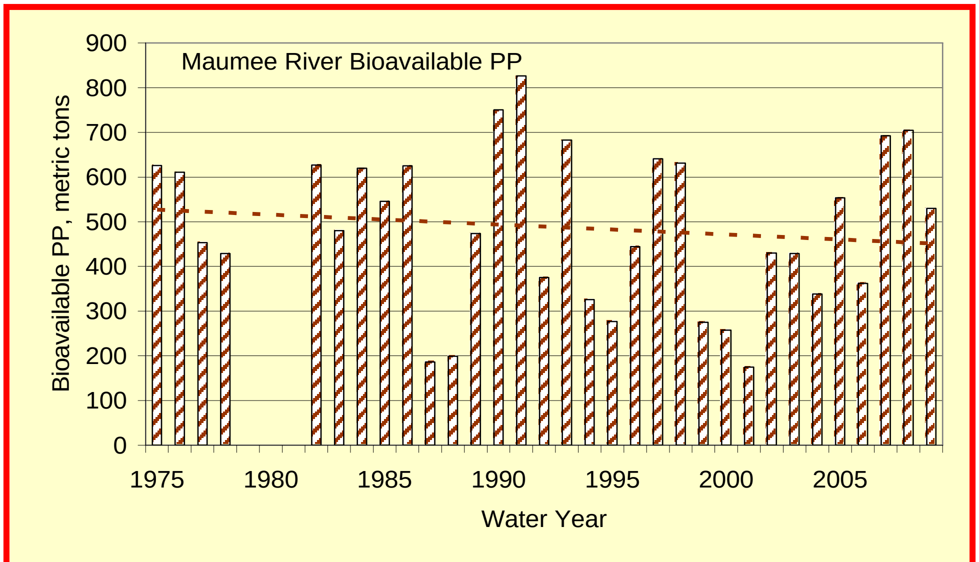

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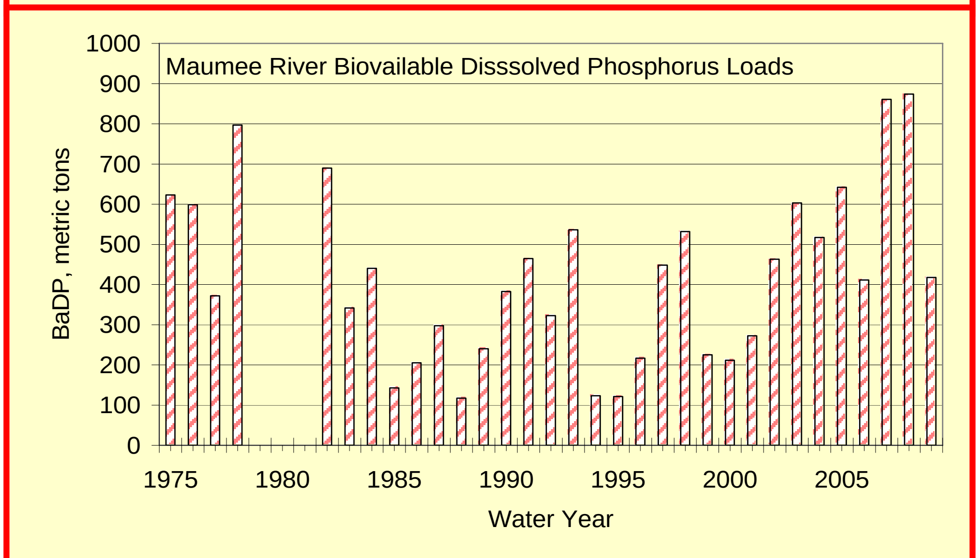

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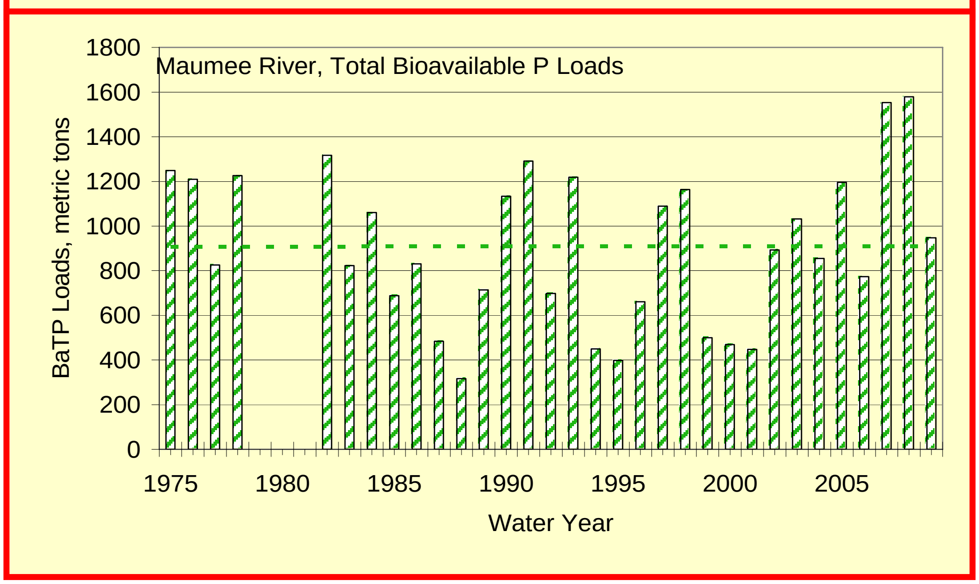

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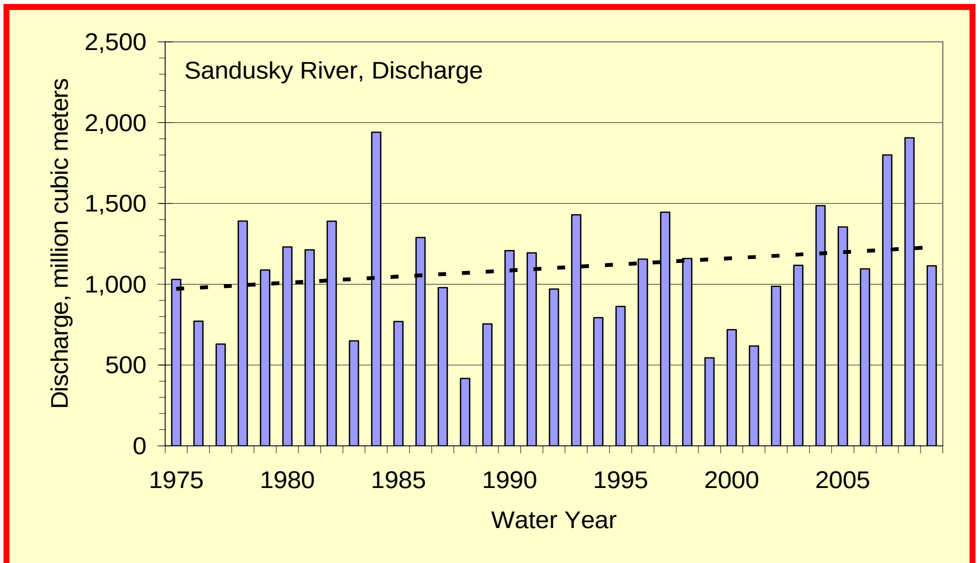

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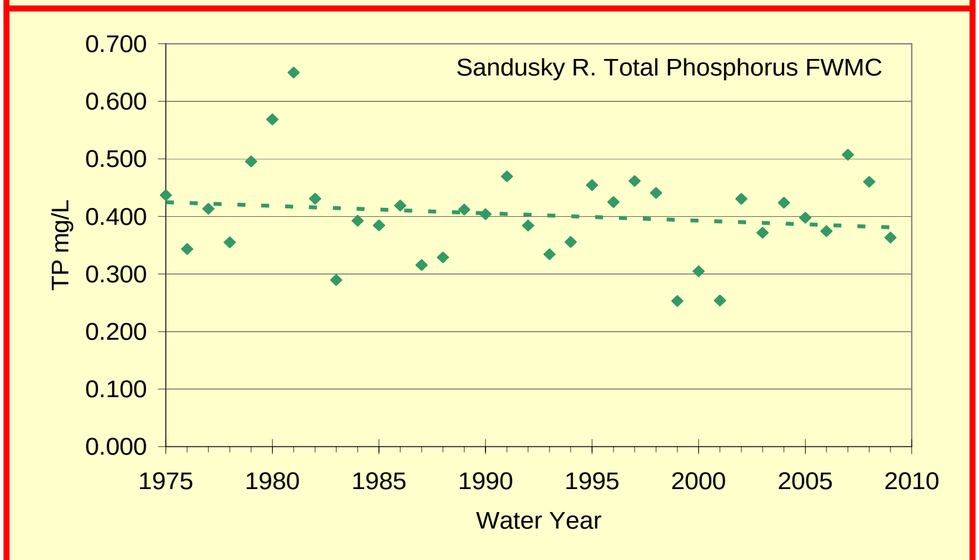

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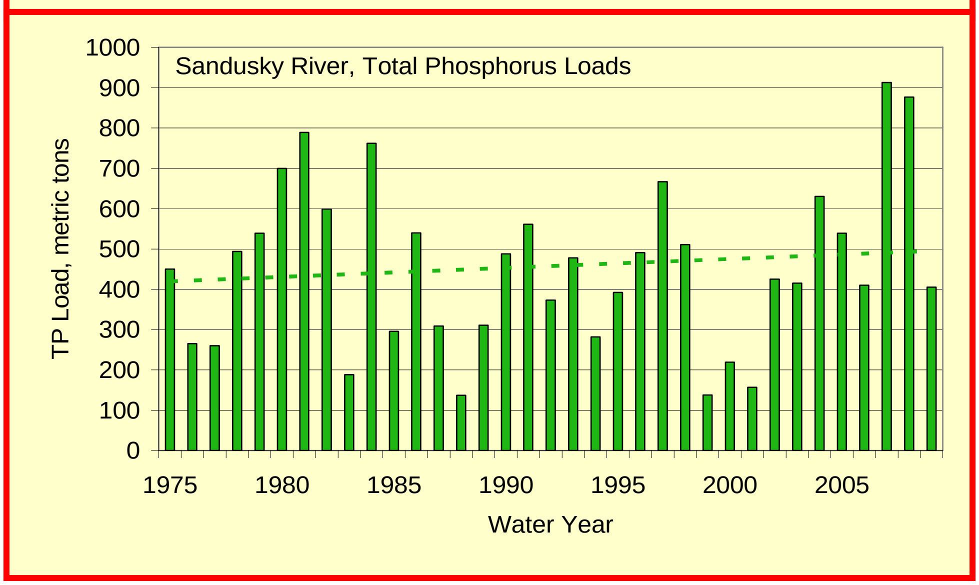

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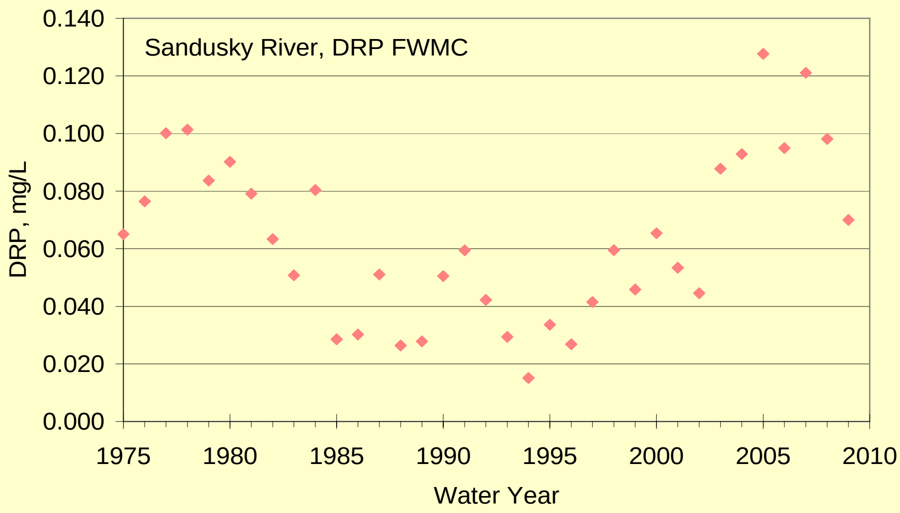

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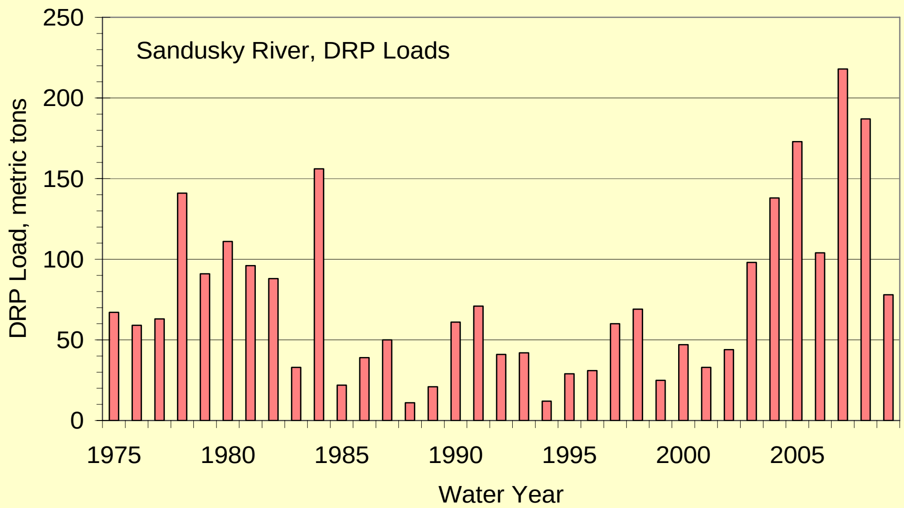

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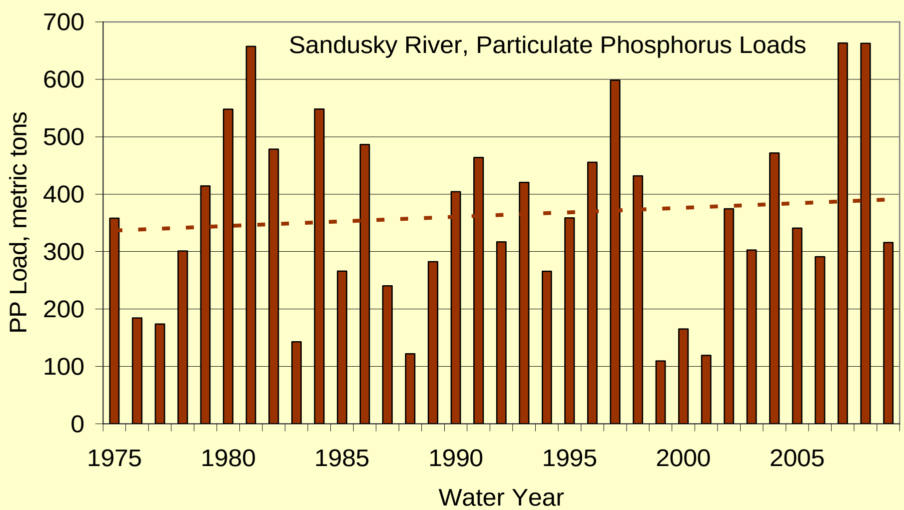

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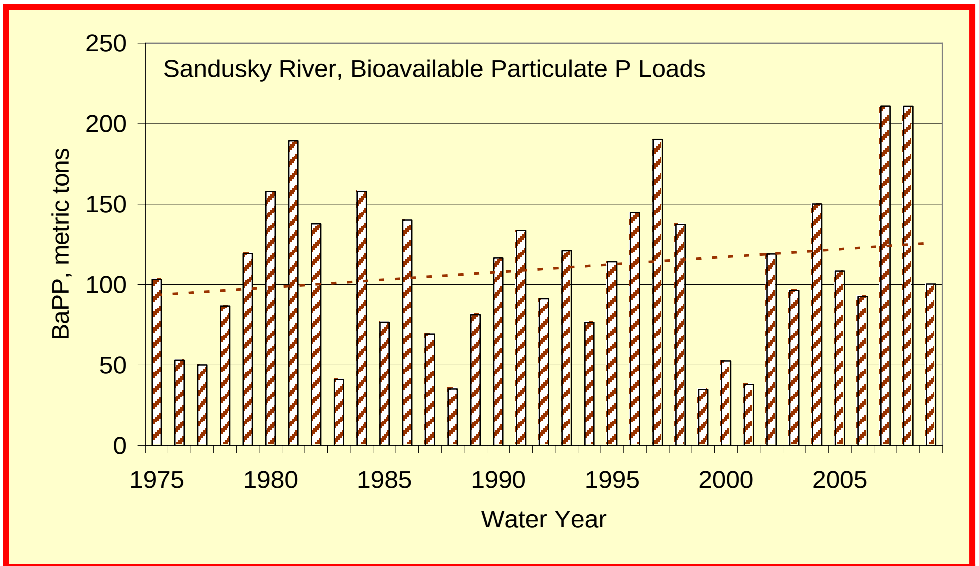

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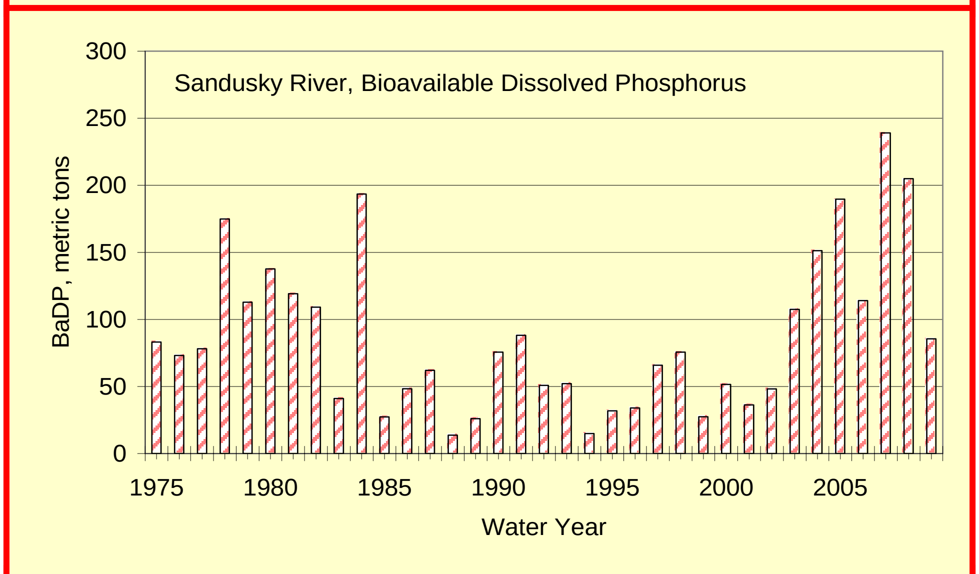

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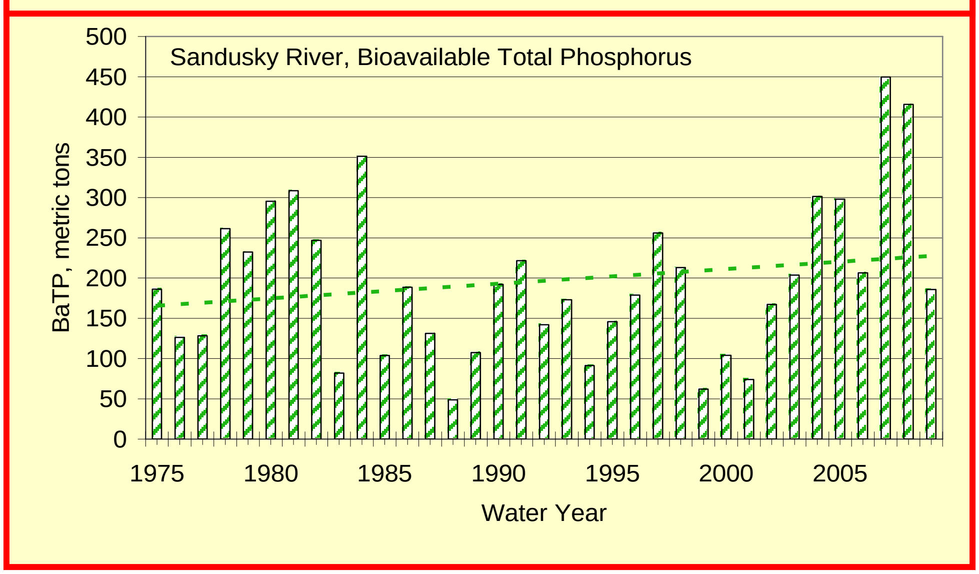

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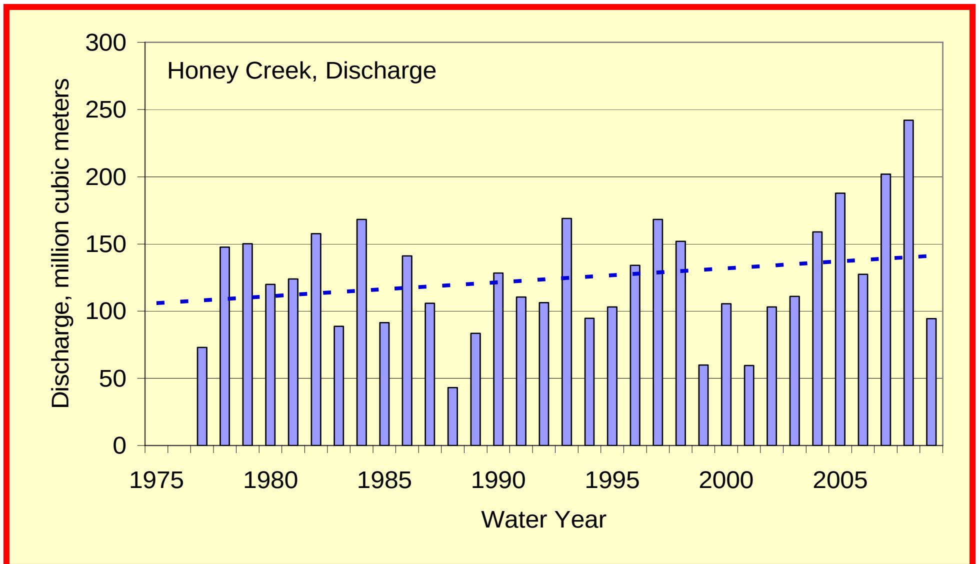
Figure 10. Trends in annual discharge and annual FWMCs and loads for selected phosphorus forms in the Sandusky River. Continued.



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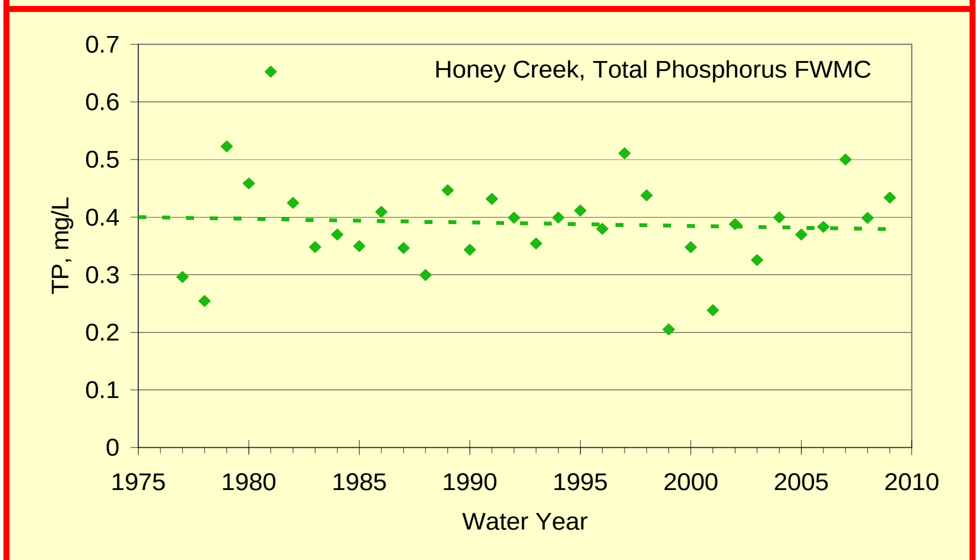

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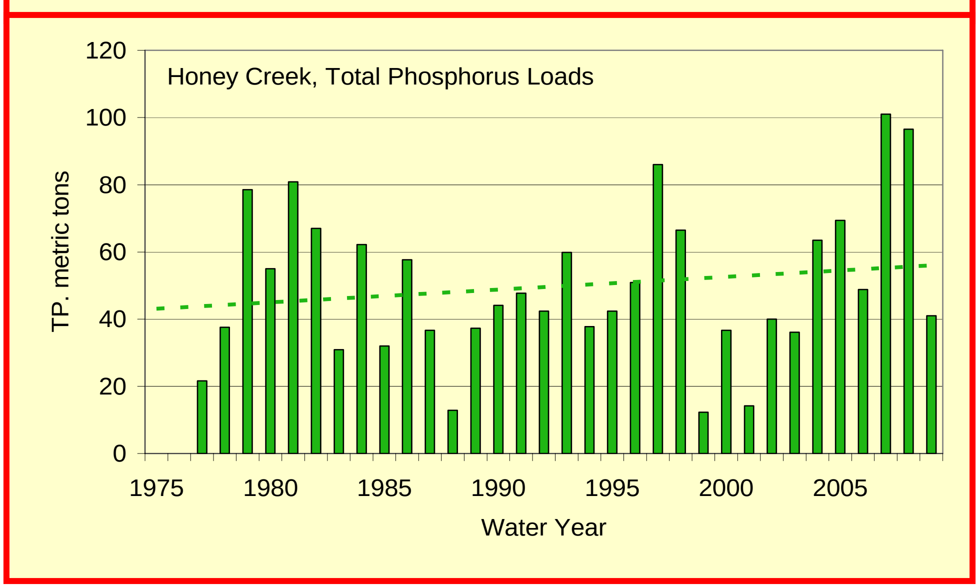

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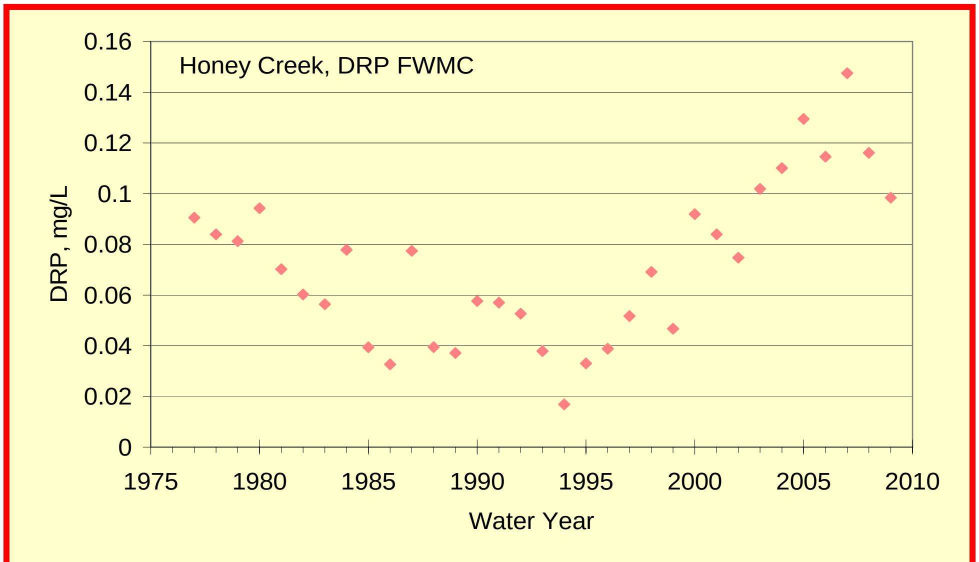

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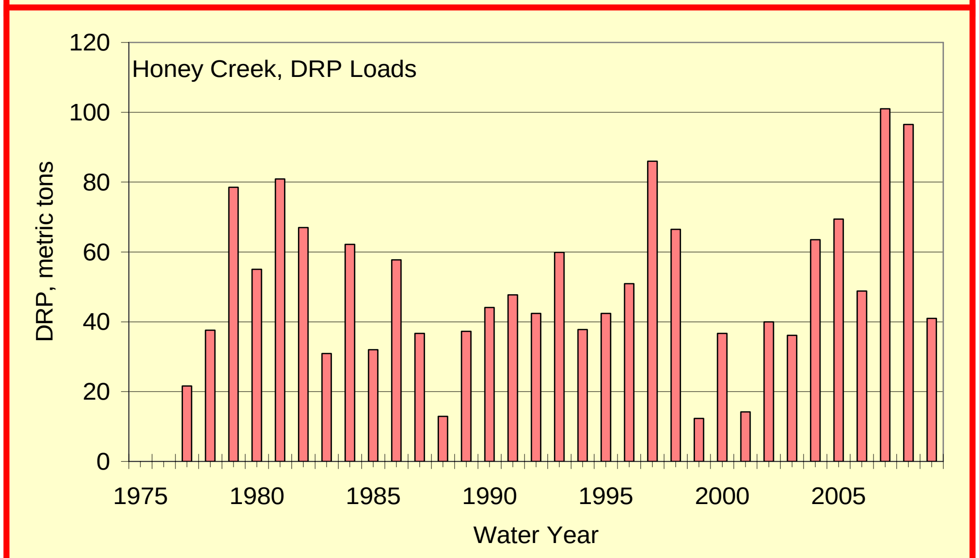

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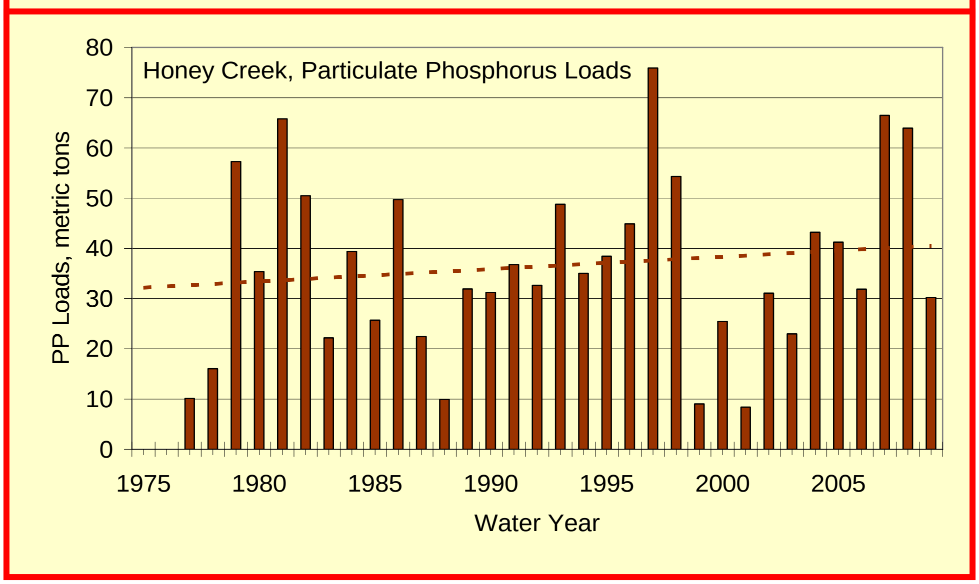

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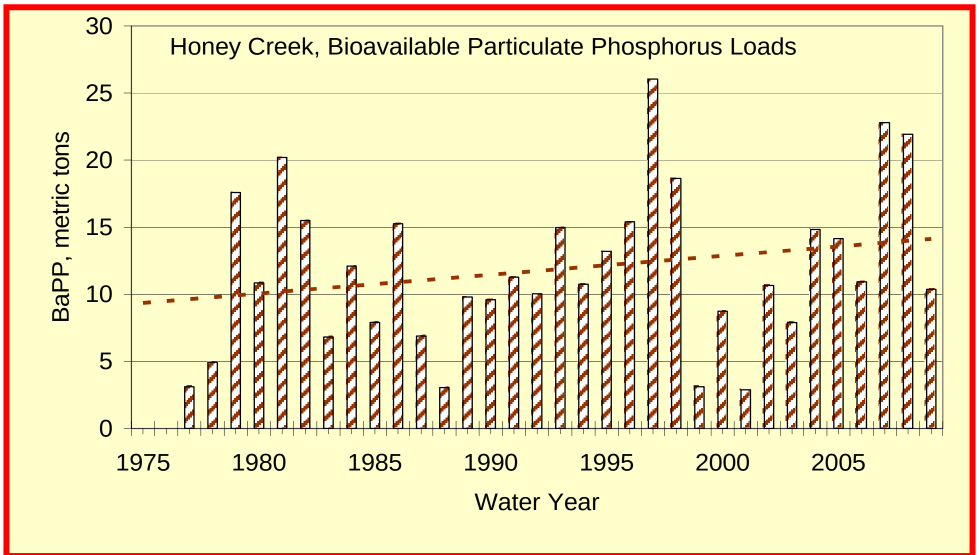

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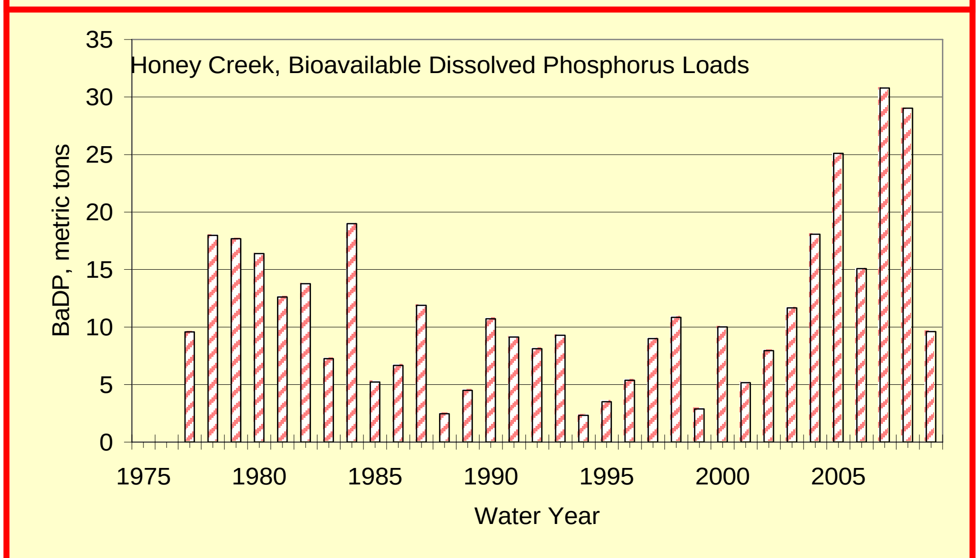

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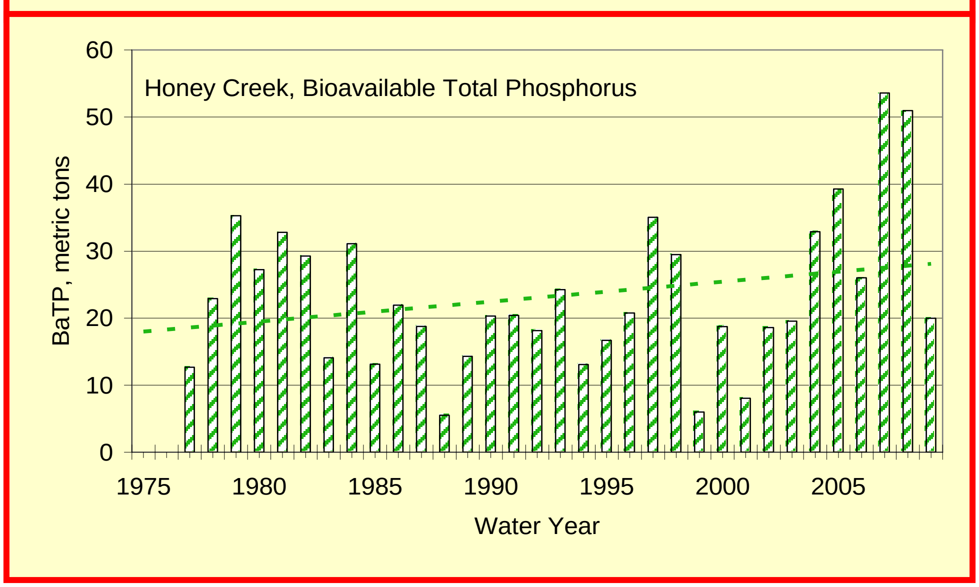

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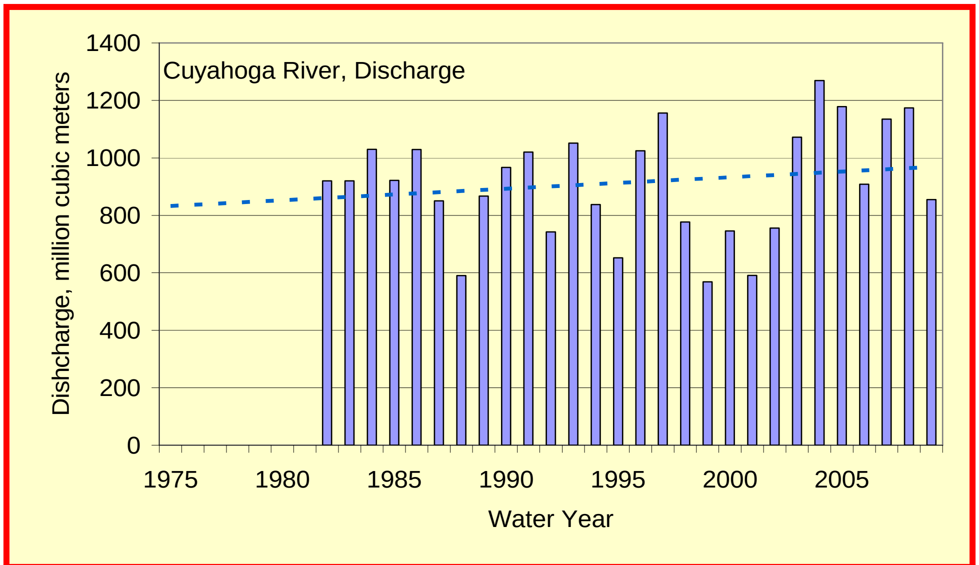

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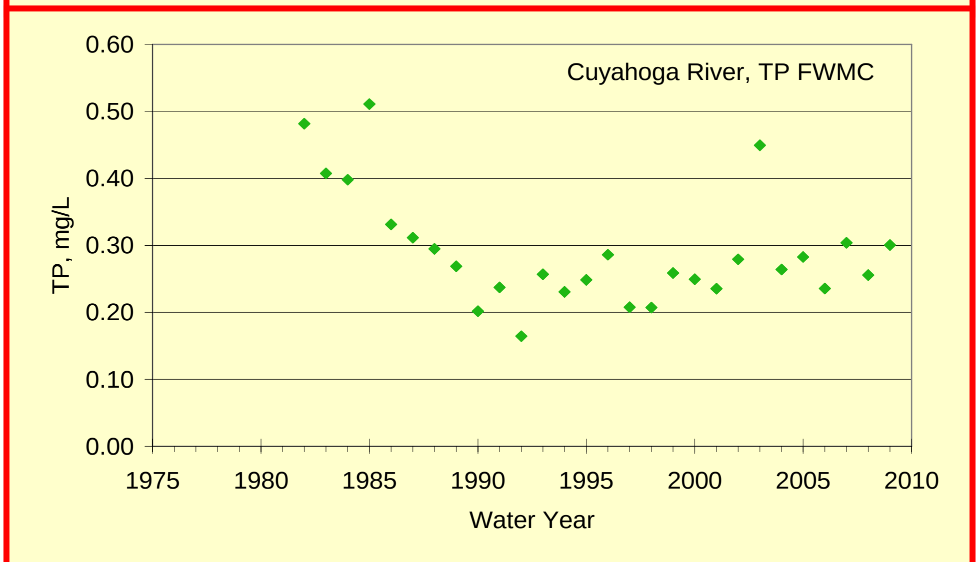


Figure 12.B. Annual flow-weighted mean concentrations of total phosphorus for the Cuyahoga River for the period of record.

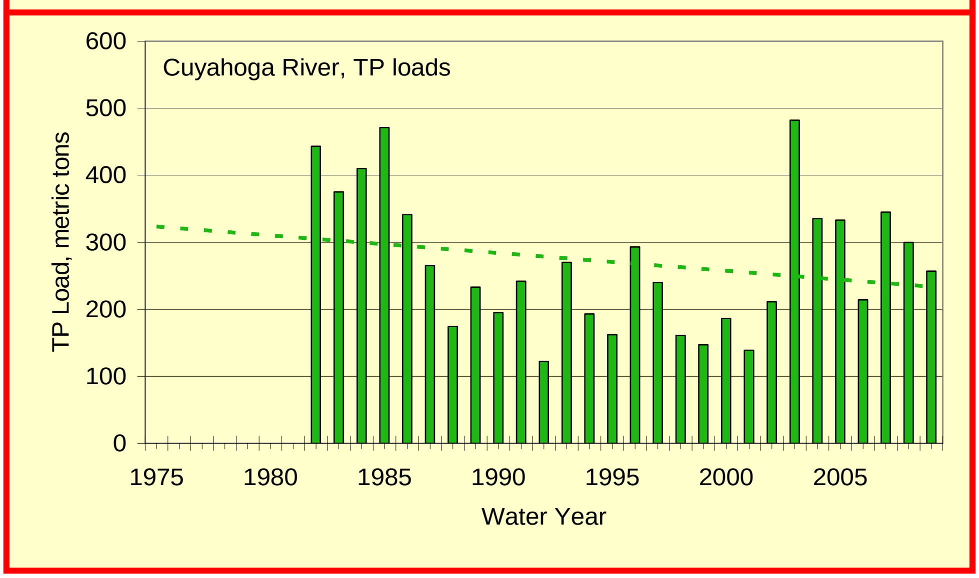


Figure 12.C. Annual total phosphorus loads for the Cuyahoga River for the period of record.

Figure 12. Trends in annual discharge and annual FWMCs and loads for selected phosphorus forms in the Cuyahoga River.

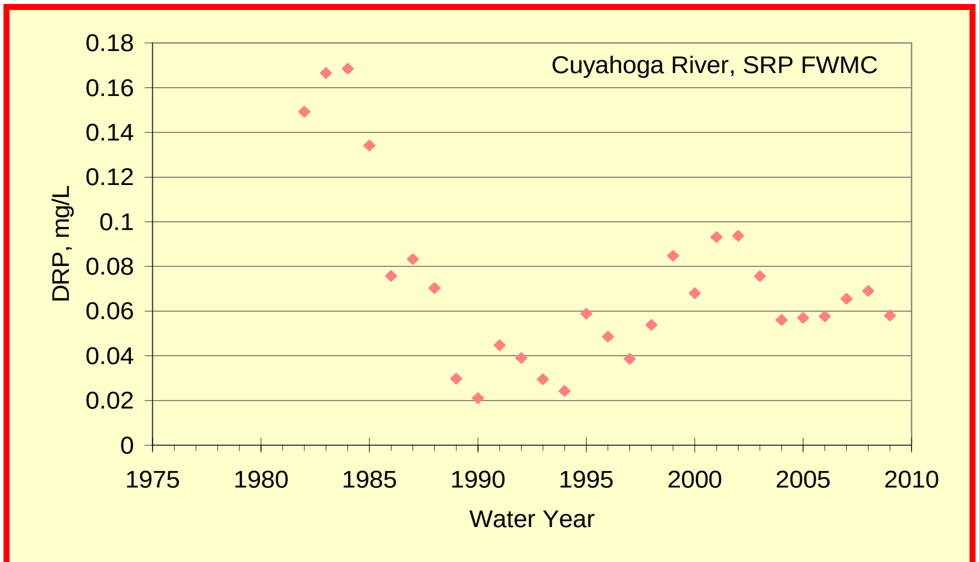


Figure 12.D. Annual flow-weighted mean concentrations of dissolved reactive phosphorus for the Cuyahoga River for the period of record.

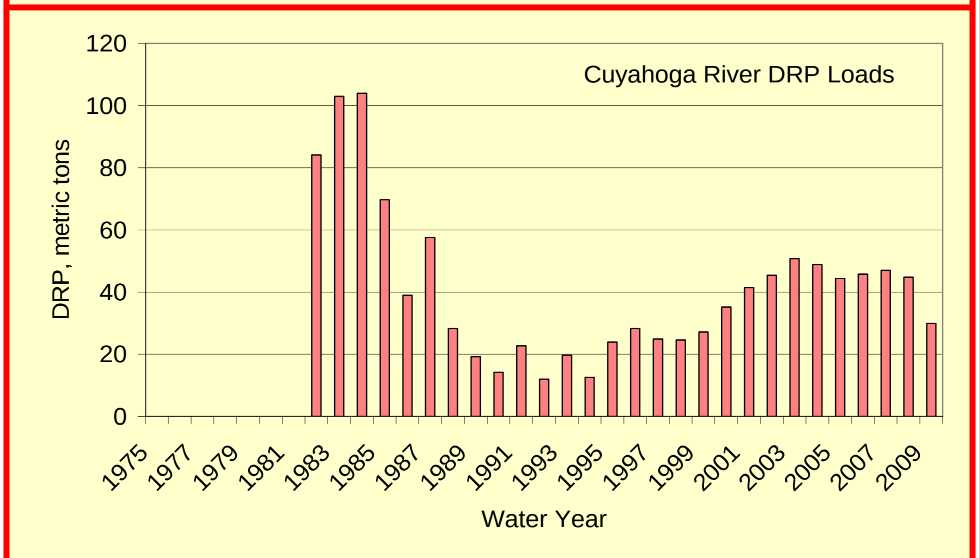


Figure 12.E. Annual loads of dissolved reactive phosphorus for the Cuyahoga River for the period of record.

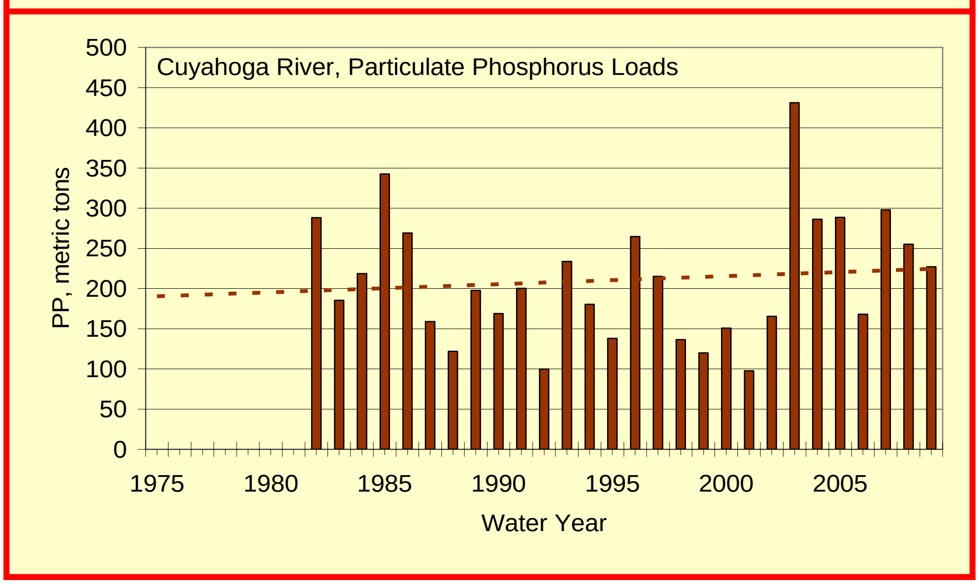


Figure 12.F. Annual loads of particulate phosphorus for the Cuyahoga River for the period of record.

Figure 12. Trends in annual discharge and annual FWMCs and loads for selected phosphorus forms in the Cuyahoga River. Continued.

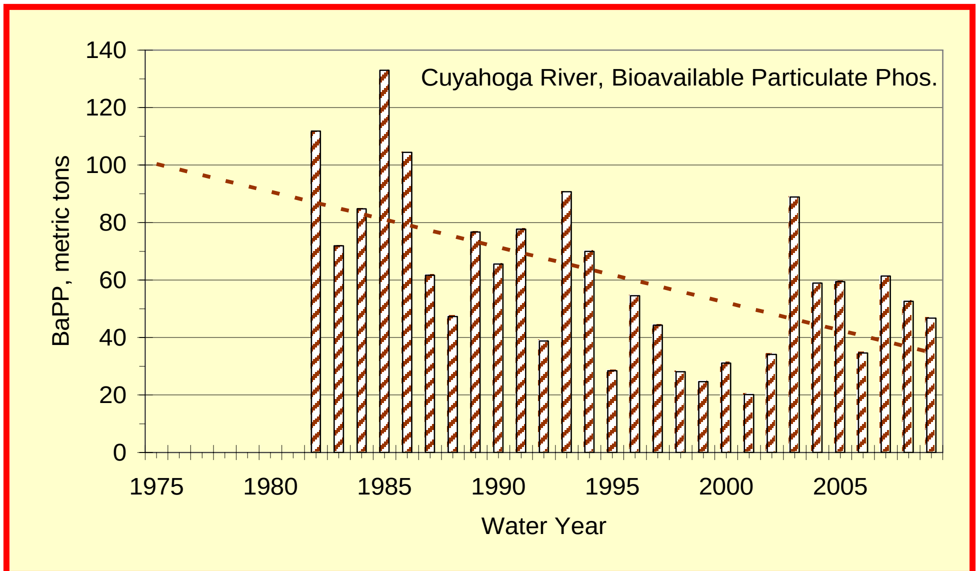


Figure 12.G. Annual loads of bioavailable particulate phosphorus for the Cuyahoga River for the period of record.

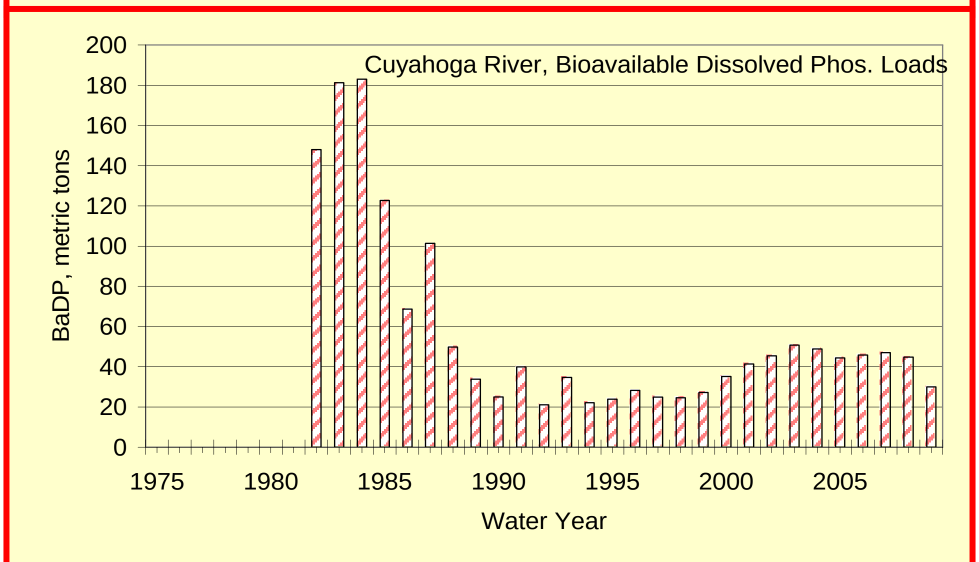


Figure 12.H. Annual loads of bioavailable dissolved phosphorus for the Cuyahoga River for the period of record.

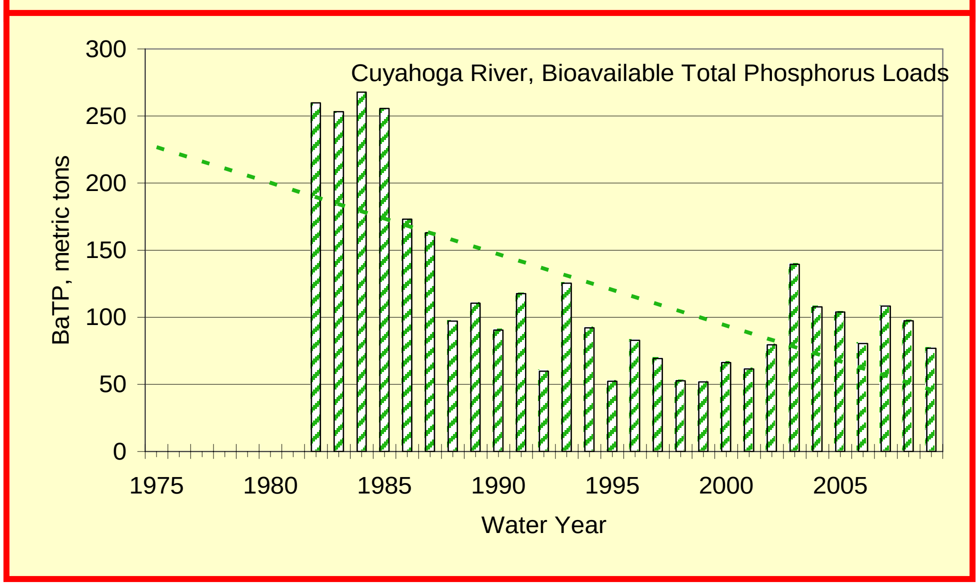
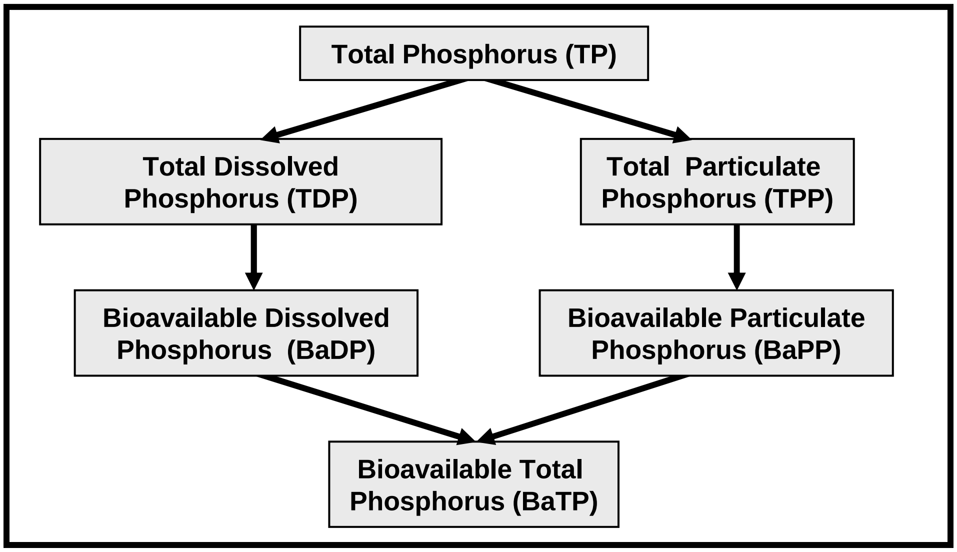
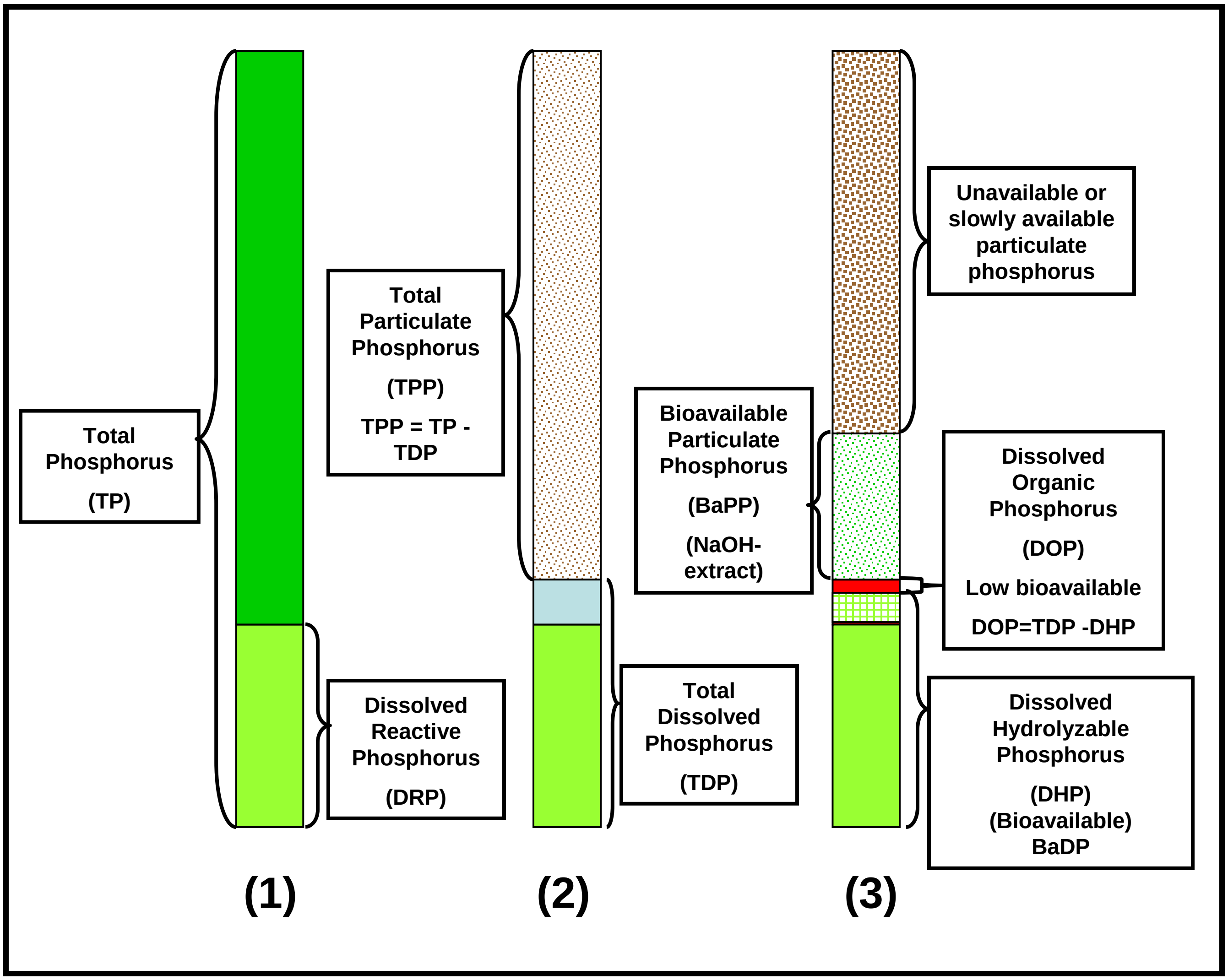


Figure 12.I. Annual loads of bioavailable total phosphorus for the Cuyahoga River for the period of record.

Figure 12. Trends in annual discharge and annual FWMCs and loads for selected phosphorus forms in the Cuyahoga River. Continued.


Total Phosphorus (TP)
Total Dissolved Phosphorus (TDP)
Total Particulate Phosphorus (TPP)
Bioavailable Dissolved Phosphorus (BaDP)
Bioavailable Particulate Phosphorus (BaPP)
Bioavailable Total Phosphorus (BaTP)



Total Phosphorus
(TP)
Dissolved Reactive Phosphorus
(DRP)
Total Particulate Phosphorus
(TPP)
TPP = TP - TDP
Total Dissolved Phosphorus
(TDP)
Bioavailable Particulate Phosphorus
(BaPP)
(NaOH-extract)
Unavailable or slowly available particulate phosphorus
Dissolved Organic Phosphorus
(DOP)
Low bioavailable
DOP=TDP -DHP
Dissolved Hydrolyzable Phosphorus
(DHP)
(Bioavailable)
BaDP
(1)
(2)
(3)

Appendix 2. Results Dissemination and Leveraging

A. State Agency Interactions

Data on the increasing loading of bioavailable phosphorus to Lake Erie was conveyed to the Ohio EPA by NCWQR staff in 2007, leading to the formation of the Ohio Lake Erie Phosphorus Task Force. NCWQR staff participated fully in the task force deliberations, leading to the Publication of the Task Force Final Report in 2010. The final report contains graphs of the estimates of total bioavailable phosphorus loading to Lake Erie for the Maumee and Sandusky rivers, using the procedures detailed in these studies.

B. LEPF bioavailability grant as seed money

The work initiated under this $10,000 grant from the LEPF was the basis for a grant successful grant application to the U.S. EPA-Great Lakes National Program Office for $100,000 for additional studies of bioavailable phosphorus loading to Lake Erie. This work includes: (1) extension of the type of work done under the current grant, with the addition of the use of ferric oxide strips as a means of bioavailable phosphorus measurements; (2) studies of the fate and effects of particulate and dissolved phosphorus as they move through the lower Maumee River, Maumee Bay and the adjacent waters of the western basin of Lake Erie during storm events in the Maumee River; (3) studies of the bioavailability of current sewage treatment plant effluents; and (4) studies of the relationship between phosphorus soil test levels and the potential for dissolved phosphorus runoff from soils.

C. Presentation of results to scientific and public meetings (selected examples)

1. Lake Erie Millennium Network/ Research Needs Workshop 4.3 (Teleconference)
   Location: (Multiple via Teleconferencing)
   Date: December 9-10, 2008
   Title: Background Materials on Nonpoint Source Loading of Nutrients and Sediments from the Maumee and Sandusky Rivers. (In three parts).

2. International Association of Great Lakes Research
   Location: State College, PA
   Date: June 1, 2007
   Title: Increased bioavailability of nonpoint-derived phosphorus loads to Lake Erie from the Maumee and Sandusky rivers.

3. Ohio Academy of Science
   Location: Toledo, Ohio
   Date: April 12, 2008
   Title: Increasing Trends in Dissolved Phosphorus Loading to Lake Erie from Northwestern Ohio Watersheds. (Lake Erie Symposium)

4. Great Lakes Phosphorus Forum
   Location Windsor, Ontario
   Date: July 29, 2009
   Title: What changes have we seen in phosphorus coming out of tributaries?

5. Conservation Tillage and Technology Conference
   Location: Ohio Northern University, Ada, OH
   Date: February 27, 2009
   Title: Phosphorus Stratification, No-till and Water Quality