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EFFECTIVENESS AND LONGEVITY OF ALUM TREATMENTS IN LAKES

Eugene B. Welch
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Water Resources Series
Technical Report No.145
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FOREWARD

Alum has been used extensively to control lake eutrophication by inactivating sediment phosphorus. While alum is generally considered to be effective and relatively long lasting, specific information on effectiveness and longevity of past treatments had not been critically evaluated as a whole. This project was therefore undertaken to provide an in-depth, critical evaluation of initial compared to long-term effectiveness of that lake restoration method.

ABSTRACT

Effectiveness and longevity of alum treatments were evaluated in 21 lakes across the U.S.; 9 were shallow, unstratified and 12 were deeper and thermally stratified during the summer. Effectiveness was judged from reductions in lake phosphorus content and the trophic state index (Secchi transparency, chlorophyll and total phosphorus), both initially and over a period of from 8 to 20 years, depending on the lake, following treatment. Effectiveness of treatments in unstratified, shallow lakes was described separately from that in stratified lakes, primarily because the availability of internally loaded phosphorus to affect epilimnetic trophic state is uncertain in stratified lakes. Effectiveness was substantial and relatively long-lasting in the majority of lakes evaluated. The fate of added aluminum was also evaluated by examining sediment profiles. Specific findings are presented in the Conclusions.

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EFFECTIVENESS AND LONGEVITY OF ALUM TREATMENTS IN LAKES

INTRODUCTION

Excessive biomass levels of planktonic algae, especially blue-green algae, are common in many North American eutrophic lakes, reservoirs, and ponds during summer months. The consequences of this problem are well known and include degraded lake quality, impaired recreation, and taste, odor, and disinfection by-products in potable water supplies. Long-lasting controls of nuisance algae involve significant reductions in external loading and in-lake nutrient concentrations. Most often the management target for reducing macronutrients has been phosphorus (P). This is because P has a sedimentary biogeochemical cycle which permits its removal or complexation from nutrient sources and lake water without concern for the additional atmospheric inputs which would be expected for nitrogen and carbon. Also, P is often the biomass-limiting nutrient, or can be made so by lowering its in-lake concentration.

Mass balance models of in-lake P concentration indicate that concentration should decline approximately as external P loading declines, modified by the rate of dilution and by P sedimentation. Advanced wastewater treatment, diversion, and/or nutrient interception normally provide the reduction in P loading. Marsden (1989) has described two types of responses to a significant load reduction.
Lake Washington's response (Edmondson and Lehman, 1991), which illustrates one of the types, involved a decline in annual mean total P (TP) which was almost exactly that predicted by a mass balance model. This occurred because the lake is deep, has a rapid water renewal rate, an oxic hypolimnion, and has a relatively short history of enrichment. The response of most lakes to reduced loading has been of the second type wherein mean lake total phosphorus concentration (TP) has remained significantly higher than expected for many years. This response is due to sediment P release or internal P loading, a process which is particularly

common in lakes with long histories of sediment enrichment (see reviews by Cullen and Forsberg, 1988; Sas et al., 1989; Cooke et al., 1993a). Examples of this type of response include Lakes Norrviken and Vallentuna, SWE (Ahlgren, 1977, 1980), Shagawa Lake, MN (Larsen et al, 1979),and Lake Sammamish, WA (Welch et al., 1986).

Aerobic and anaerobic sediments can be significant P sources to the water column (Marsden, 1989), especially during warm summer months when microbial activity is high, pH from photosynthesis increases, and low or zero dissolved oxygen concentrations occur in water overlying sediment and/or at night in the littoral zone. Also, dilution may be low from reduced summer inflows, allowing nutrients to accumulate in the water column.

There are several in-lake management methods which have been employed to control sediment P release following a reduction in external P loading. These include sediment skimming and dredging, addition of dilution water, hypolimnetic withdrawal and aeration, sediment oxidation, and P inactivation (see Cooke et al., 1993a for complete review). Except for P inactivation, these technques have been used infrequently to accelerate lake recovery after a reduction in external loading, and long term follow-up studies of effectiveness and longevity have been even fewer.

Phosphorus inactivation is a method of controlling sediment P release by binding inorganic sediment P with an aluminum (Al) salt such as aluminum sulfate or sodium aluminate. Recently, there also have been applications of calcium or iron salts for this purpose, but only a few case histories are known (see Cooke et al., 1993a,b for review of the procedure). Phosphorus inactivation has been used for 25 years for this purpose, in both Europe and North America, and in shallow and deep lakes. Before this present study there were conflicting reports regarding its effective use in shallow lakes. As well, control of P release has been reported to last from less than one year to as long as 15 or more years. Thus, while this technique to control internal P loading following reduction in external loading has been used often, and some case histories have been followed by lake managers for years, questions remain regarding how long the method, in the majority of cases, could be expected to control sediment P release and how effective it would continue to be in limiting algal biomass by limiting P concentration in a lake's photic zone.

The purpose of this project is to evaluate aluminum sulfate/sodium aluminate treatments of lakes for their effectiveness/longevity of control of sediment P release and improving lake trophic state. Selected shallow (polymictic) and deep (dimictic) lakes in Washington, Wisconsin, Ohio, New York, Vermont, New Hampshire, and Maine, all treated prior to 1987, were studied (Table 1). In addition, a deep water core sample of Delavan Lake, WI was obtained in July, 1991, two months after the lake's alum treatment.

CONCLUSIONS

Treatments were effective in six of the nine shallow lake basins (two lakes each had two separate basins), controlling lake phosphorus (P) by an average of 48% (29 to 75%) for at least eight years on average (four of six treatments were still effective after eight years). Control of internal loading *per se* was even more effective. Trophic state index (TSI) improved in all cases where sediment P inactivation was effective and remained lower in four of the six effectively treated lakes after eight years. Dense macrophyte crops were probably responsible for ineffectiveness or short longevity in three lakes/lake basins, because they cause uneven floc distribution or recycle sediment P from below the floc layer through senescence and decay. The results suggest that iron redox may have been the most controlling mechanism for P release in lakes where alum was effective, even though anaerobic conditions in sediment-overlying water is usually not obvious in shallow unstratified lakes.

Alum applications to stratified lakes were highly effective and long lasting in controlling sediment P release. In two cases (Mirror and Shadow Lakes), control lasted at least 13 years, and in Snake Lake, control has been evident for at least 19 years. In these cases percent reduction in internal P loading has been continuously above 80%. Trophic state of stratified treated lakes improved, but the significance of alum treatments usually cannot be separated from that of nutrient diversion. At West Twin Lake diversion was more important than control of P release from hypolimnetic sediments in improving trophic state. The significance of vertical P entrainment should be considered before treating dimictic lakes. Alum treatment of lakes with high external loading was not effective.

Table 1. Characteristics and alum doses of project lakes. AS = aluminum sulfate, SA = sodium aluminate

	Lake name & location	Treatment Date	Chemicals Used	Dose gm Al/m^3	Application Depth (m)	Lake Area(Km^2)	Maximum Depth(m)	Mean depth (m)	Alkalinity mg/l $CaCO_3$	Mixis	Reference
1.	Annabessacook Winthrop ME	8/78	AS:SA 1:1.6	25	hypolimmnion	5.75	12.0	5.4	20	dimictic	Dominie, 1980
2.	Cochnewagon Winthrop ME	6/86	AS:SA 2:1	18	hypolimnion	1.56	9.0	5.7	13-15	dimictic	Dennis and Gordon, 1991
3.	Kezar Sutton NH	6/84	AS:SA 2:1	30	hypolimnion	0.74	8.2	2.7	3-10	dimictic	Connor and Martin, 1989
4.	Morey Fairlee VT	5-6/86	AS:SA 2.1:1	11.7	hypolimnion	2.20	13.0	8.4	35-54	dimictic	Smeltzer, 1990
5.	Irondoquoit Bay Rochester NY	7-9/86	AS	28.7	hypolimnion	6.79	23.7	6.9	170	dimictic	Spittal and Burton, 1991
6.	Dollar, Kent OH	7/74	AS	20.9	90% hypolimnion 10% surface	0.02	7.5	3.9	101-127	dimictic	Cooke et al., 1978
7.	West Twin, Kent OH	7/75	AS	26	hypolimnion	0.34	11.5	4.4	102-149	dimictic	Cooke et al., 1978
8.	Pickeral Stevens Point WI	4/73	AS	7.3	surface	0.20	4.6	3.0	110	polymictic	Knauer and Garrison, 1980
9.	Mirror, Waupaca WI	5/78	AS	6.6	hypolimnion	0.05	13.1	7.8	222	dimictic	Garrison and Ihm, 1991
10.	Shadow, Waupaca WI	5/78	AS	5.7	hypolimnion	0.17	12.4	5.3	188	dimictic	Garrison and Ihm, 1991
11.	Snake Woodruff WI	5/72	AS:SA ratio unknown	12 (80% of lake v)	surface	0.05	5.5	2.0	50	dimictic	Garrison and Knauer, 1984
12.	Horeshoe Manitowoc WI	5/70	AS	2.6	surface	0.09	16.7	4.0	218-278	dimictic	Garrison and Knauer, 1984
13.	Eau Galle Spring Valley WI	5/86	AS	4.5	hypolimnion	0.60	9.0	3.2	144	dimictic	Barko et al., 1990
14.	Long, Port Orchard WA (Kitsap Co.)	9/80	AS	5.5	surface	1.40	3.7	2.0	10-40	polymictic	Welch, et al., 1988
15.	Long, Tumwater WA (Thurston Co.)	9/83	AS	7.7	surface	1.30	6.4	3.6	45	polymictic	Welch, et al., 1988
16.	Erie, Mt. Vernon WA	9/85	AS	10.9	surface	0.45	3.7	1.8	80-90	polymictic	Welch, et al., 1988
17.	Campbell Mt. Vernon WA	10/85	AS	10.9	surface	1.50	6.0	2.4	80-90	polymictic	Welch, et al., 1988
18.	Pattison Tumwater WA	9/83	AS	7.7	surface	1.10	6.7	4.0	45	polymictic	Welch, et atl., 1988
19.	Wapato, Parkland WA	7/84	AS	7.8	surface	0.12	3.5	1.5	NA	polymictic	Welch, et al., 1988

Treatment effectiveness wanes probably due to downward distribution of Al floc in the sediment over time as evidenced by the lack of distinct Al markers (higher concentrations) in sediment cores and the uniform, but high Al content in a treated lake compared to a connected, untreated lake. This Al floc sinking phenomenon (and other evidence) indicates that the relatively high, soluble (filterable) Al concentrations (average = 150 μg/L) determined in several treated lakes may be largely natural, organically bound and, hence, probably nontoxic.

MATERIALS AND METHODS

SAMPLING WASHINGTON AND WISCONSIN LAKES

All lakes were sampled at one station corresponding to the deepest portion of the lake, with the exception of Long and Pattison (Thurston County) Lakes. These two lakes each consist of two distinct basins that were sampled and evaluated separately. The Washington study lakes were sampled for constituents shown in Table 2 eight times in the summer (June-September) of 1990 and nine times in the summer of 1991. In addition, Campbell and Erie Lakes were sampled in summers, 1992 and 1993. The Wisconsin lakes were sampled by the Wisconsin Department of Natural Resources (WDNR) on four occasions and by project personnel once during the summer of 1991.

WATER ANALYSIS FOR WASHINGTON LAKES

Temperature and dissolved oxygen (DO) were determined at one-meter intervals at the deep station in each lake using a YSI model 57 oxygen meter. Air-calibrated meter DO values were compared to DO values at two depths in each profile determined using the azide modification of the Winkler method (APHA, 1989). Transparency was determined with a Secchi disc.

Discrete water column samples were taken using a Van Dorn bottle at three or four depths at each site, and were analyzed for P, pH, alkalinity, phytoplankton biovolume, and chlorophyll a (chl a). Samples for soluble reactive P were filtered through pre-soaked 0.45 μm filters and frozen until analyzed. SRP (soluble

TABLE 2. PHYSICAL AND CHEMICAL CHARACTERISTICS DETERMINED IN WATER SAMPLES

Characteristic	Method of Measure
Dissolved Oxygen (DO)	Winkler-azide modification
Temperature	Thermometer
pH	Corning probe
Transparency	Secchi disc
Alkalinity	Potentiometric titration
Soluble Reactive Phosphorus (SRP)	Ascorbic-molybdate, 10 cm cell
Total Phosphorus (TP)	Persulfate digestion/ascorbic-molybdate
Residual Aluminum (TAR, SAl)	ICP
Chlorophyll a (chl a)	Acetone extraction
Phytoplankton biovolume	Direct count-Palmer cell

reactive P) was determined in a 10 cm cell using the ascorbic acid method (APHA, 1989). TP was determined by analyzing for SRP in preserved (3 drops H_2SO_4/250 ml) whole water samples after persulfate digestion.

Total Al and soluble aluminum (SAl) samples were collected on at least two occasions from each lake in Washington, the midwest and east. SAl samples were filtered using a 0.22 µm pre-soaked filter, and all samples were acidified to pH less than 2 with analytical grade concentrated nitric acid and stored in acid-washed polyethylene bottles. Al was determined using an ICP (inductively-coupled argon plasma emission) spectrophotometer at the 396.15 nm emission line.

Samples for chl a were filtered through Gelman 25 mm glass fiber filters. Filters were ground in a $MgCO_3$-saturated, 90%-acetone solution. Chl a absorbance was read at 665 and 750 nm following grinding of the filter, 24-hour extraction with 90% acetone and correction for phaeophytin.

Field plus laboratory precision for SRP, TP, Al and chl a analyses was determined by replicating 10 percent of the field samples for the Washington lakes. Most variability was due to constituent distribution, because analytical precision was within 3 percent (Table 3). The accuracy of P and chl a analyses was determined by comparison with EPA standards, and that for Al was determined by analysis of in-house primary standards (Table 3).

Discrete samples for the analysis of phytoplankton taxa were taken from the surface and mid-depth on each sampling data and preserved with Lugol's reagent. A 30-ml subsample was concentrated 10-fold by centrifuging for 20 minutes and aliquots were counted in a 0.2-ml Palmer-Maloney cell. Fifty grids were counted for each sample. Identification was performed to genera and biovolume and percent blue-greens were determined.

SAMPLING OHIO AND EASTERN LAKES AND WATER ANALYSIS

Ohio

Dollar, East, and West Twin Lakes were sampled 12-15 times per summer, between early June and mid-September, except in 1989 when only five samples were taken. Samples were taken at the deepest site, at 1-m intervals, using a 2 liter Kemmerer bottle. Temperature was determined with a YSI telethermometer, and

TABLE 3. PRECISION AND ACCURACY OF WATER QUALITY ANALYSES. FIELD PRECISION IS EXPRESSED AS THE RANGE IN PERCENT SD, LAB PRECISION AS PERCENT SD, AND ACCURACY AS MEAN PERCENT RECOVERY (ALL N=2).

Parameter	Field Precision	Lab Precision	Accuracy
aluminum	0.9% - 3.3%	1.7%	99.5%
SRP	0.0% - 17.2%	1.1%	104.3%
TP	1.6% - 13.9%	3.1%	100.5%
chlorophyll a	2.6% - 26.1%	2.6%	86.7%

dissolved oxygen (DO) was determined by titration, using the azide modification of the Winkler method. A 20 cm black/white Secchi disk was used for transparency.

Total P was determined from triplicate subsamples of each water sample, using the ascorbic acid method (APHA, 1989) following persulfate--H_2SO_4 digestion in a pressure cooker for 45 minutes at 121°C and 15 PSI. Analytical precision was always within 3%, based on a daily standard curve with known concentrations spanning the expected unknown concentrations. Chlorophyll a (chl a) was determined from absorbances at 665 and 750 nm, using Lorenzen's (1967) equations, after filtration through Gelman GF/C filters and extraction in 90% acetone (Long and Cooke, 1971). Results were not corrected for pheophytin.

Irondequoit Bay, New York

Depth profiles for temperature, dissolved oxygen, and other variables were obtained using a Hydrolab unit, calibrated before use. Water samples were obtained with a peristaltic pump strapped to the Hydrolab sonde. Two sites on the bay were sampled at weekly intervals during summer months. Sample analysis followed Standard Methods (APHA, 1989) (Spittal and Burton, 1991).

Lake Morey, Vermont

Lake Morey was sampled weekly at 1-m intervals at a central station during 1981-82 and 1986-87. In other years, spring TP was determined at turnover, followed by weekly monitoring for Secchi disc transparency. Standard Methods (APHA, 1989) were used for analytical procedures (Smeltzer, 1990).

Kezar Lake, New Hampshire

The lake was sampled biweekly at its deepest point, at 2, 4, and 6 m (mid epilimnion, metalimnion, hypolimnion). DO and temperature were determined with a YSI meter, chl from a 0-6 m integrated sample using the trichromatic equations, and TP using the persulfate digestion method (APHA, 1989) (Connor and Martin, 1989).

Cochnewagon Lake and Annabessacook Lake, Maine

Dissolved oxygen and temperature were determined biweekly at three stations at 1-m intervals. Transparency was determined with a 20 cm disk using a viewing scope. Chl was determined from 0-7-m depth integrated samples (procedure not specified). TP samples, collected at one meter intervals, were analyzed with a modified, automated single reagent method, using Standard Methods (APHA, 1989) (Dennis and Gordon, 1991). Annabessacook Lake data were obtained with essentially identical methods. In 1991, samples were sent to Kent State University (see Ohio lakes for procedure) for total P determinations.

SEDIMENT SAMPLING

Two sediment cores were taken in each lake in Washington, the midwest and east in 1991 for the determination of P, Al, Fe and Pb. Cores were sectioned at 2-cm intervals and analyzed for Al to determine the current depth of the aluminum hydroxide layer. The determination of stable Pb profiles can provide an estimate of sedimentation rates. The beginning (circa 1930) and end (circa 1972) of leaded gasoline use provide dates in profiles, which were used to determine if there had been any marked changes in P deposition since treatment.

SEDIMENT ANALYSIS AND PHOSPHORUS RELEASE

Twenty-four separate sediment cores were also taken for the determination of anaerobic phosphorus release rates from both whole and sectioned cores. Replicate 30-cm sediment cores were collected from the deep station in each lake in Washington, the midwest and east in 1991 with a gravity corer for the determination of sediment constituent profiles. Cores were sectioned at 2-cm intervals, placed in plastic bags and kept at 4° C until dried and digested.

Sediment samples were digested and analyzed as follows (APHA, 1989). Samples were dried overnight at 105° C and approximately one gram from each sample was subsequently placed in a 7-ml Taylor tube. Five ml of reagent grade nitric acid was added and the samples were allowed to stand overnight to prevent foaming, followed by digestion at 150° C for three hours in a block digester. Two blanks and a liquid standard were digested with each batch. After cooling to room temperature, samples were brought up to volume using deionized water. The tubes were covered with parafilm, inverted several times, and the remaining particulates allowed to settle. The digestate was decanted into acid-washed polyethylene bottles for storage until analysis. Samples were analyzed for Al, Fe, and Pb using ICP. Sediment total TP was determined by analyzing the digestate for SRP.

Sediment P release rates from intact cores and from cores with the top 2 cm and the top 6 cm removed were determined in order to estimate whether control of anaerobic P release was still apparent at these depths in the sediments. Two dozen cores for the determination of anaerobic P release rates were collected from each lake and transported intact to the laboratory. Surficial sediment was removed (for the 2- and 6-cm sections) using a vacuum aspirator. The head space in each core was filled with lake bottom water collected at the time of coring. For each section, replicate cores were incubated in an anaerobic environment at 20° C for approximately 4, 8, 16, and 32 days. After each period samples for total soluble phosphorus (TSP) and TP were collected at a constant depth of 8 cm above the sediment in a nitrogen atmosphere. TSP samples were filtered through a 0.45 μm presoaked filter, and the filtrate P was subsequently determined in the same manner as TP described above.

EVALUATION OF TREATMENT EFFECTIVENESS AND LONGEVITY

Longevity of alum treatments is defined as the number of years in which the rate of treated sediment P release is less than pretreatment rates, or less than the rates of a control lake. Effectiveness has two components: (1) effectiveness in controlling sediment P release (e.g. percent reduction) and (2) improvement in lake trophic state. These components must be separated because control of sediment P release with alum cannot affect lake trophic state unless sediment-released P reached the photic zone in significant amounts prior to treatment. In the case of polymictic lakes (all Washington lakes plus Pickerel Lake, WI), a direct transport from a temporary hypolimnion to the entire water column may occur with each mixing event. In dimictic lakes, vertical entrainment and/or P diffusion from metalimnion to epilimnion during summer months may or may not be significant, depending upon lake morphometry, shoreline features which protect it from wind (trees, steep bluffs, etc.), and the P concentration gradient, all of which can influence the effects of the lake's metalimnetic P on epilimnetic P (Jones and Welch, 1990; Kortmann et al., 1982; Osgood, 1988). The alum treatment could be effective at controlling hypolimnetic sediment P release but have little impact on controlling epilimnetic algal blooms.

A second problem arises when evaluating the effectiveness of an alum treatment (or any in-lake method) following a reduction in external nutrient loading. How can the effects of the alum application on lake trophic state be separated from the effects of reduced loading? In the case of the polymictic Washington lakes, summer external nutrient loading is minimal or absent due to low rainfall. This means that lake sediments, and processes such as bioturbation, macrophyte senescence, and sediment oxygen demand and/or pH increases are at least the primary or possibly the only phosphorus source to the water column during the summer. Moreover, in none of the Washington lakes were there reductions in point sources of nutrients. However, watershed BMPs were implemented for Long (Thurston) and Pattison, which should provide long-term reductions in inputs but probably not account for the lake-P decrease observed in those lakes the first several years. Stormwater was diverted from Wapato, which did not show a positive effect on that or alum. For the Wisconsin, Ohio, and eastern lakes, the effects of reduced external loading cannot be easily separated from those resulting from alum-caused reduced internal phosphorus loading (after alum application) unless there is a detailed annual phosphorus budget for pre- and post-treatment years or an untreated control or reference lake. While that is true for trophic state indicators in the epilimnion, it is less so for hypolimnetic P, the increase of which has been shown to be due largely to release from sediment (Nurnberg, 1987). Also, sediment release rates have been shown to remain rather constant for many years after external P diversion (Ahlgren, 1988).

Stratified Lakes

Effectiveness of P internal loading control in stratified lakes was simply the percent reduction in directly measured sediment P release rate:

$$\text{Percent effectiveness} = [(R_{pre} - R_{post})/R_{pre}]*100$$

where R_{pre} and R_{post} are, respectively, the anaerobic sediment release rates of P measured prior to and following treatment. This approach was possible only for pre- treatment P release rates available from WDNR for Mirror and Shadow Lakes in Wisconsin.

Where no direct measurement of pre-treatment release rates were available, pre and post-treatment P internal loading was determined from the observed rate of increase in TP in the hypolimnion:

$$\text{Sediment P Release Rate, mg/m}^2\text{-day} = [(TP_2\text{-}TP_1)*V/(t_2\text{-}t_1)]/A$$

By assuming that the observed increases in summer hypolimnetic TP were due solely to internal loading (i.e., summer inflow TP mass is negligible), the increase in hypolimnetic TP concentration ($TP_2 - TP_1$), the hypolimnetic volume (V), and the hypolimnetic sediment area (A) were used to compute the net mass of P entering the overlying water per unit surface area from the sediments for a given time period (t_2-t_1). These are net sediment release rates (gross minus sedimentation), but have been shown to be very similar to directly measured rates in the laboratory under anaerobic conditions (Welch, 1977; Nurnberg, 1987).

Unstratified (Polymictic) Lakes

Anaerobic P release rates measured directly in cores may not be indicative of total internal P loading in unstratified lakes that apparently remain aerobic during the summer. In those cases, effectiveness of internal loading control was determined from changes in whole lake TP concentration. If annual external loading were assumed to have remained constant since treatment, as should be the case for lakes with relatively undeveloped watersheds (especially Erie and Campbell), then a comparison of pre- and post-treatment whole lake TP concentrations, which during summer months are due to internal loading only, should indicate treatment effectiveness. To determine the amount of TP increase due to internal loading requires subtraction of the portion originating from external sources. Effectiveness was thus assessed from observed post-treatment mean summer TP concentration in the lake (TP_{post}), the pre-treatment mean summer TP concentration (TP_{pre}), the average inflow TP concentration (TP_{in}), and the sediment retention coefficient (R), which was assumed to be approximated by $1/(1+\rho^{0.5})$ (Larsen and Mercier, 1976.), according to:

$$\text{Percent effectiveness} = \{1-[(TP_{post} - TP_{in}(1-R))/(TP_{pre} - TP_{in}(1-R))]\}*100$$

The above equation estimates the TP concentration due to external loading, subtracts it from the observed post- and pre-treatment observed TP levels, thereby providing an estimate of the percentage control of internal loading following treatment. The assumptions made in the use of this equation are the following: 1) the retention coefficient (R) for external TP remains constant for pre- and post-treatment conditions; and 2) the external loading has remained essentially constant since treatment. As for assumption one, (P_{in}-P_{out}/P_{in}) has been observed to remain relatively constant for a specific lake without internal loading before and after sewage diversion (Edmondson and Lehman, 1981). Also, retention as a function of flushing rate (as used here) has remained constant from year to year in a lake with internal loading (Shuster et al., 1986). Assumption two, on the other hand, may not be valid and a lack of data makes it difficult to confirm or disprove that assumption for a given lake. Sediment-P profiles in cores were not useful in detecting recent changes in P deposition (see Results).

A second method in shallow lakes is analogous to that used to determine internal loading in stratified lakes from the observed increase in hypolimnetic TP. Assuming that the mass input of TP to the lake during summer is negligible, which is typical for western Washington, a net rate of internal loading was calculated based on the increase in whole-lake TP. From the maximum amplitude during the summer period, an areal sediment P release rate was calculated:

$$\text{Sediment P Release Rate, mg/m}^2\text{-day} = [(TP_2\text{-}TP_1)*V/(t_2\text{-}t_1)]/A$$

In this case, the change in phosphorus concentration (TP_2-TP_1), the volume (V), and the area (A) are on a whole-lake basis. While this technique does not separate loading from sources such as macrophytes, it does allow a working comparison of observed pre- and post-treatment internal loading resulting from <u>all</u> mechanisms occurring in the lake.

Thirdly, a simple comparison of pre- and post-treatment mean-summer, whole-lake TP was used to evaluate effectiveness. This approach would theoretically give the least direct estimate of internal loading change because it does not measure a rate and there is no attempt to separate internal from external sources.

The objective with all these methods is to determine the percent reduction in internal loading, i.e., the non-macrophyte P transport from sediment to water. That is the process which alum controls.

The Carlson (1977) Trophic State Index (TSI) was used to determine if trophic state changed following nutrient diversion and alum treatments. Unlike some other indices, the TSI is based upon a trophic continuum idea, and thus provides a quantitative estimate from TP, chl and/or transparency of the degree of eutrophy (or oligotrophy) of a lake with little or no nonalgal turbidity. TSI values between 40 and 50 have been widely accepted as "mesotrophic." Values above 60 are considered to approach "hypereutrophy." Changes of 10 TSI units approximate a doubling or halving of algal biomass, depending upon the direction of change, according to Carlson (1977). When algal biomass (chl) is P-limited, and transparency is determined by algal biomass, the TSI values from the three variables will be essentially identical. Deviations from this suggest other controls of algal biomass (e.g. grazing, light).

For this report, TSI values from TP, chl, and transparency were based on mixed layer (whole water column) for Washington polymictic lakes, and on surface or epilimnion data for the dimictic lakes. In the case of Wisconsin lakes, only water column volume-weighed values were available from some dates for pre- and immediate post-treatment.

RESULTS

HISTORY OF UNSTRATIFIED LAKES

Campbell and Erie Lakes

These two shallow lakes, which are connected by a small stream, are relatively unaffected by development. Only about 1% of the watershed is developed, including about 21 and 32% of their respective shorelines. Forest covers 72-77% of their combined watersheds. Nevertheless, dense blue-green algal blooms were commonplace in both lakes producing scums, odors and occasional fish kills. Although both lakes have <10% of their surface area covered with submersed macrophytes, emergent plants cover 51-75% of the shoreline in Erie and 76-100% of Campbell. Prior to the alum treatment (Table 1), internal loading contributed 63% of the P to Erie Lake and 27% to Campbell Lake on an annual basis (Entranco, 1983).

Pattison Lake

This shallow lake consists of a smaller north basin (33 ha) and larger south basin (77 ha) separated by a shallow sill with basin depths being similar ($\bar{z}$ = 4.2 versus 4.0 m). Much of the lake's shoreline is developed and, in summer, nearly 100% of the south basin surface is covered with native submersed macrophytes. The outlet from the south basin of Pattison Lake enters the south basin of Long Lake - Thurston.

Long Lake - Thurston County

This shallow lake also consists of north (70 ha)and south (60 ha) basins separated by an isthmus with basin mean depths of 3.6 m and 3.8 m, respectively. The outflow from Pattison Lake enters the south basin. The shoreline around the north basin is almost completely developed, while about 50% of the south basin shoreline is developed. Algal blooms and dense macrophyte stands (mostly *Myriophyllum spicatum*) threatened the recreational quality of both basins (Entranco, 1987). Because most of the controllable P entering both Long and Pattison Lakes was from internal sources, both lakes were treated with alum in autumn 1983 (Table 1).

Wapato Lake

This shallow lake received stormwater from residential and commercial areas in two major drainage basins as well as from a nearby interstate highway. Recreational use of the lake as part of a park was limited by turbidity, algal blooms and oil and grease. A multifaceted restoration project was begun in 1982 in which part of the lake was dammed off for treatment of winter stormwater, which was subsequently bypassed around the major lake area (Entranco, 1986).. Dilution water was also added to the lake for two years following stormwater diversion. Although TP dropped 46% after diversion/dilution and chl a dropped 38%, post-treatment peak TP and algal blooms still persisted due largely to internal loading. So the lake was treated with alum in 1984 (Table 1). Unfortunately, dilution water was discontinued after the alum treatment, so there are no data from a pre-alum treatment/no dilution period to evaluate alum.

Long Lake - Kitsap County

This lake has been the subject of study and restoration activity since 1976. After 3.5 years of post-treatment study, the lake's level was drawn down nearly 2 m in June of 1979. The former level was re-established in October. About one-third of the lake bottom was exposed during the summer. Although the exposed sediments reduced the macrophytes for one year only, summer lake TP inexplicably declined in 1980 following drawdown (Jacoby et al., 1982).

In September 1980 the lake was treated with alum to control internal P loading (Table 1). A second treatment of the same magnitude was employed in October 1991, although the lake's quality usually indicated that the first treatment was still partially successful. In the meantime (1988-1990), a macrophyte harvesting and removal project was carried out in an attempt to determine if macrophyte removal would lower lake TP concentration (Welch et al., 1993). Efforts are being renewed by the local residents to more effectively control the macrophyte crop.

Pickerel Lake, Portage County, Wisconsin

Pickerel Lake is a shallow (mean depth 2.6 m), polymictic, hardwater lake (alkalinity = 110 mg/L). A naturally eutrophic lake with no surface inflows or outflows, Pickerel lake had a history of algal blooms and winter fishkills. In 1973 the lake was treated with alum at a dose of 7.3 mg Al/L--the only treatment ever performed on the lake (Table 1). Immediate results showed a decrease in summer chl and TP concentrations, but after mixing in July a large algal bloom dominated by the blue-green alga *Microcystis aeruginosa* occurred. Garrison and Knauer (1984) hypothesized that the short-lived effectiveness of the alum treatment was due to the redistribution of the alum floc to the center of the lake, thus leaving the majority of lake sediments uncovered and not subject to control by alum. Only TP data are available prior to treatment. The 1972 summer mean of 35 μg/L is not significantly different from those in 1973, 1982 and 1991 (44, 23, 30 μg/L). Given its high alkalinity, a much larger alum dose would have been appropriate and may have been more effective.

EFFECTIVENESS/LONGEVITY IN UNSTRATIFIED LAKES

Phosphorus

Effectiveness and longevity of alum treatment in controlling sediment P release was judged by the third method, a comparison of pre- and post-treatment mean, whole-lake TP concentrations, because data were more complete for that method. However, effectiveness determined by TP concentration was usually less than if the two more direct measures of internal loading were used (Table 4). The highest effectiveness was shown with calculated internal loading. That method, which used external loading estimates, could not be used for Long and Pattison Lakes (Thurston Co.). Those lakes each had two basins that showed marked differences in response to treatment and external loading could be applied to the whole lake only. Observed sediment P release rate also showed greater, or in one case essentially equal (Erie), effectiveness compared to using mean summer TP concentration (Table 4).

The greater effectiveness shown by more direct measures of internal loading than summer mean TP concentration was expected because P inactivation with alum controls internal loading only. While mean summer TP concentration is in large part a result of summer internal loading, external loading also contributes to that concentration. Thus, using summer TP concentration would be expected to give a lower effectiveness; 52 versus 68 percent on average for the five most successful treatments shown in Table 4. Nevertheless, TP concentration alone was relied upon to judge effectiveness/longevity overall because data were more complete.

TABLE 4. EFFECTIVENESS OF PHOSPHORUS INACTIVATION IN SHALLOW LAKES (T = THURSTON), COMPARISON OF THREE METHODS[1,2,3] FOR DATA BEFORE AND IMMEDIATELY AFTER TREATMENT (1 to 4 YEARS) AND AFTER SEVERAL YEARS (5-10) AS A RESULT OF THIS STUDY. VALUES ARE MEAN PERCENT REDUCTIONS.

Lake	Observed Release Rate[1]		Calculated Internal Loading[2]		TP Concentration[3]	
Erie	79(1)	82(5-6)	92(1)	97(5-6)	77(1)	84(5-6)
Campbell	57(1)	64(5-6)	57(1)	76(5-6)	43(1)	57(5-6)
Long (North-T)	84(1-2)	79(7-8)			60(1-2)	56(7-8)
Pattison (North-T)	81(1-2)	73(5-7)			43(1-2)	29(5-7)
Long (Kitsap)	62(1-4)	40(7-10)	78(1-4)	51(7-10)	48(1-4)	33(7-10)

[1]P release rate determined from the maximum rate of increase in whole-lake TP during summer.

[2]P internal loading calculated assuming constant external loading and retention coefficient for external loading since treatment.

[3]Mean summer, whole-lake TP concentration.

Alum was initially effective at controlling internal loading, and hence lake TP concentration, in 6 of 9 lakes/lake basins (Long and Pattison each have two basins). In four lakes/basins, treatments remained effective for 5 and 8 years (Table 5). Although chl a and TP concentrations indicated that effectiveness was finished in Pattison-north after 5 years, observed sediment release rate showed that the persistence of inactivation was longer (Table 4). The relative persistence of P inactivation in the 6 successful treatments, as indicated by mean TP concentration, is shown in Figure 1. Alum was initially ineffective in Pattison south, Wapato, and Pickerel (Table 5).

Submersed macrophytes were abundant throughout the lake/basin in three of the cases in which the treatment either failed initially or longevity was short (Wapato, Pierce County; Long-south and Pattison-south, Thurston County). Pattison-south, which was covered with several native species of rooted macrophytes, drained to Long-south providing a source of P from decomposing plants. The thick stands may also have prevented a uniform coverage of alum and been partly responsible for the poor effectiveness initially. *Myriophyllum spicatum* was abundant in Long Lake-south. Although TP was initially reduced in Wapato Lake, it returned to pre-treatment levels within a month, presumably due to P released from the late-summer senescence of a greatly increased biomass of *Ceratophyllum* and to P released from sediments possibly due to the plant-caused pH increase to 10 (Entranco, 1986). That judgment is based on using a pre-treatment level of TP from the two years when the lake received dilution water and after stormwater was diverted. Those treatments had already reduced TP 47 percent. When alum was subsequently applied, the lake was no longer being diluted. Curtailed dilution, as well as the large macrophyte biomass, probably contributed to the increased TP (24 percent, Table 5) and apparent failure of the alum treatment. If pre-dilution, pre-diversion TP values are used for comparison, there was a post-alum reduction in TP.

The second alum treatment of Long Lake (Kitsap) in October, 1991 was more successful initially than the first treatment (Table 5). TP concentration during summer averaged 32 µg/L during the first four years following the first treatment, but was 20 µg/L the summer following the second treatment, a 20 percent greater decrease relative to TP prior to the first treatment. The greater effectiveness of the second alum treatment is also evident in Figure 2, which shows lake TP concentration corrected for inflow concentration. The most

TABLE 5. EFFECTIVENESS AND LONGEVITY OF PHOSPHORUS INACTIVATION BASED ON MEAN SUMMER, WHOLE-LAKE TP CONCENTRATIONS. T = THURSTON AND K = KITSAP COUNTIES.

Lake	Pre Treatment TP μg/L (yrs)	% Reduction TP Initial (yrs)	Last (yrs)	Treatment Longevity, yrs
Erie	115 (2)	77 (1)	75 (5-8)	>8
Campbell	49 (2)	43 (1)	46 (5-8)	>8
Long (T)				
North	42 (3)	60 (1-2)	56 (7-8)	>8
South	31 (3)	32 (1-2)	50 (4-5)	5
Pattison (T)				
North	28 (3)	43 (1-2)	29 (5-7)	7**
South	30 (3)	-7 (1-2)	----	<1
Long (K)	63 (3)	48 (1-4) 68 (1)*	30(7-11)	>11
Wapato	46 (2)	-24 (1-2)	----	<1
Pickerel	35 (1)	-26 (1)	----	<1

*Second treatment in 1991 at same dose as 1980.

**See Table 4--observed release rate.

negative values, indicating high sedimentation and low internal loading are during the four post-alum years and the first post-second alum year.

Erie and Campbell Lakes in Skagit County represent the most successful cases for alum in shallow, unstratified lakes. Treatment effectiveness has remained as high after eight years as initially (Table 5, Figure 1). Likewise, treatment effectiveness was long lasting, although less effective, in Long Lake (Kitsap). The Long Lake treatment was thought to have lost its effectiveness during the fifth post-treatment year (1985) when TP returned to the pre-treatment level (63 µg/L, Figure 1). The returning high internal loading is also shown in Figure 2. That high-TP summer was later attributed to the sharp decline of the principle macrophyte (*Egeria densa)* to 10 percent of its previous biomass (see Welch and Kelly, 1990). However, TP returned to generally lower levels (mean = 41 µg/L) during years 7 to 11, but not as low (mean = 32 µg/L) as during the first 4 post-treatment years (Figure 1). Internal loading was evident, but not as high as in pre-treatment years (Figure 2). Effectiveness was also low in year 10 (1990), possibly due to intense harvesting of *Egeria* that year (Welch et al., 1993). Although effectiveness declined overall, alum apparently still had a beneficial effect for 11 years.

Trophic State

Lake trophic state (TSI) in the shallow lakes responded to alum treatments similarly to TP. The initial response in Erie and Campbell Lakes was to change from eutrophic to mesotrophic and the improved trophic state has persisted for 8 years (Figures 3 and 4). Chl a and Secchi transparency have been more variable than TP, but in general have remained lower than pre-treatment values in those lakes. The three TSI indicators track rather closely in Campbell demonstrating that algal biomass is dependent on TP. However, pre-treatment transparency in Erie was lower than expected from TP and chl a, probably due to the lake's brown (humic matter) color.

Absolute reductions in chl a and increases in transparency are not well indicated by changes in TSI values in Figures 3 and 4 because of the $\log_2$ transformations of the data in the TSI calculation. Summer chl a the last four years (5-8 years post-treatment) averaged 82% and 43% below the pre-treatment levels for Erie and Campbell Lakes, respectively, while average transparency was 17% and 50% greater than pre-treatment in

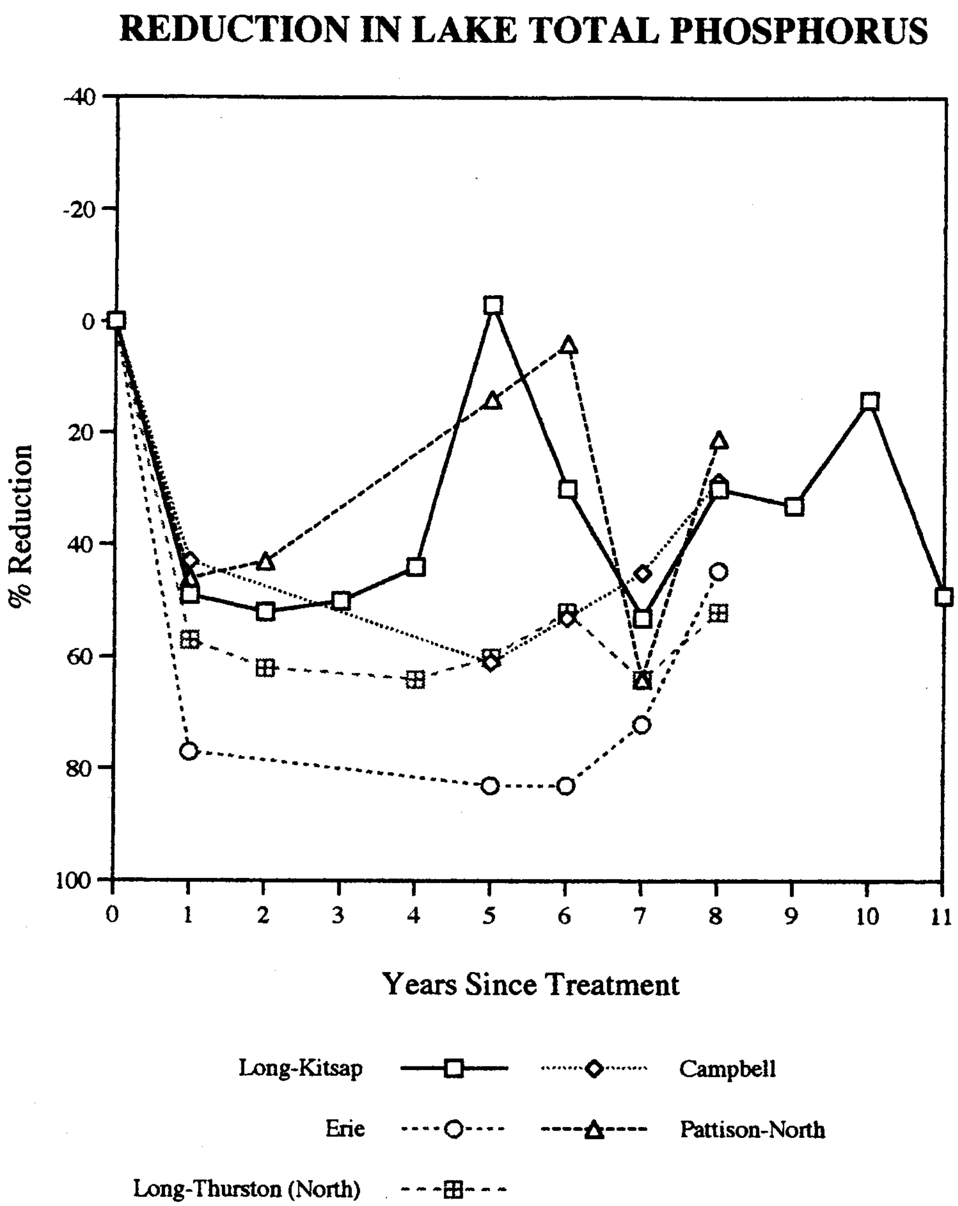

Figure 1. Percent reduction in whole-lake TP content following alum treatments in five shallow lakes (basins) in Washington state

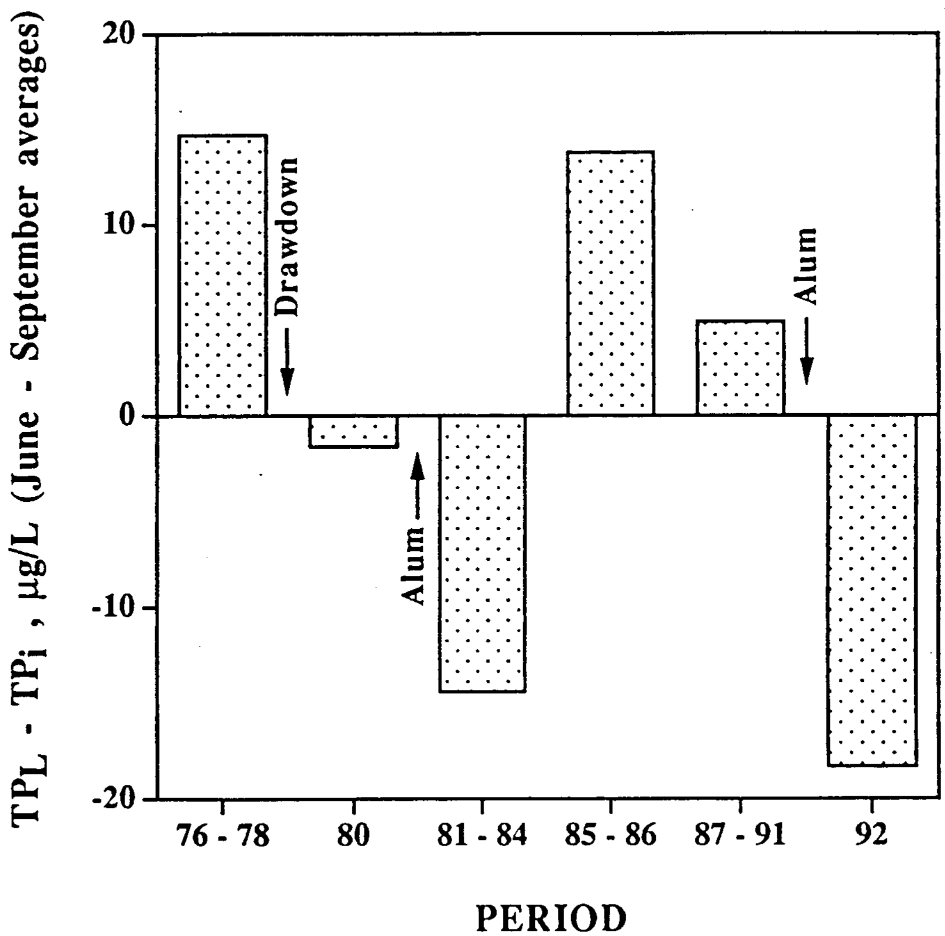

Figure 2. Mean difference between volume-weighted lake TP_L and inflow TP_i concentration during June-September for years before and after the alum treatments in Long Lake - Kitsap County

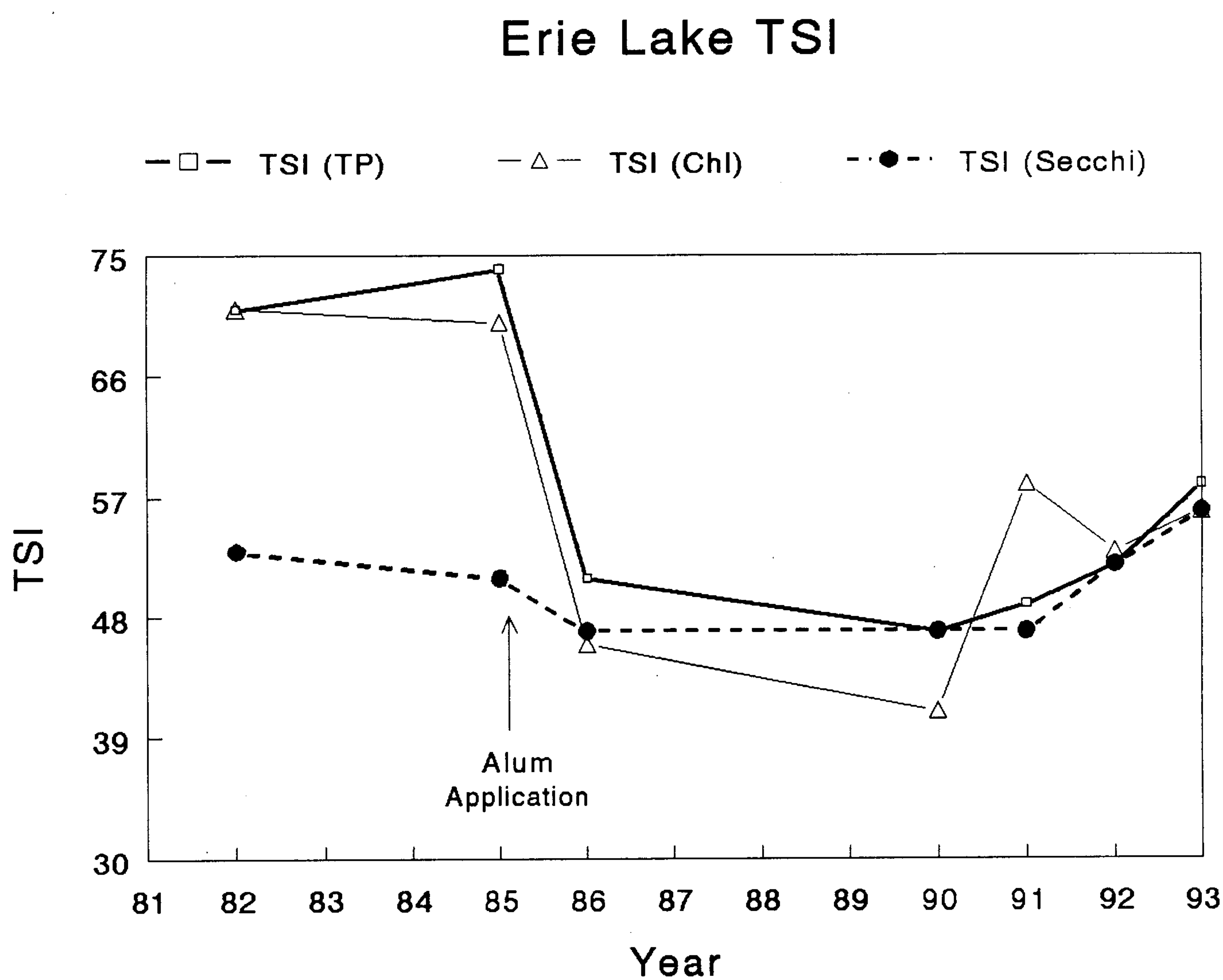

Figure 3. Trophic state index values based on summer means for respective variables in Erie Lake

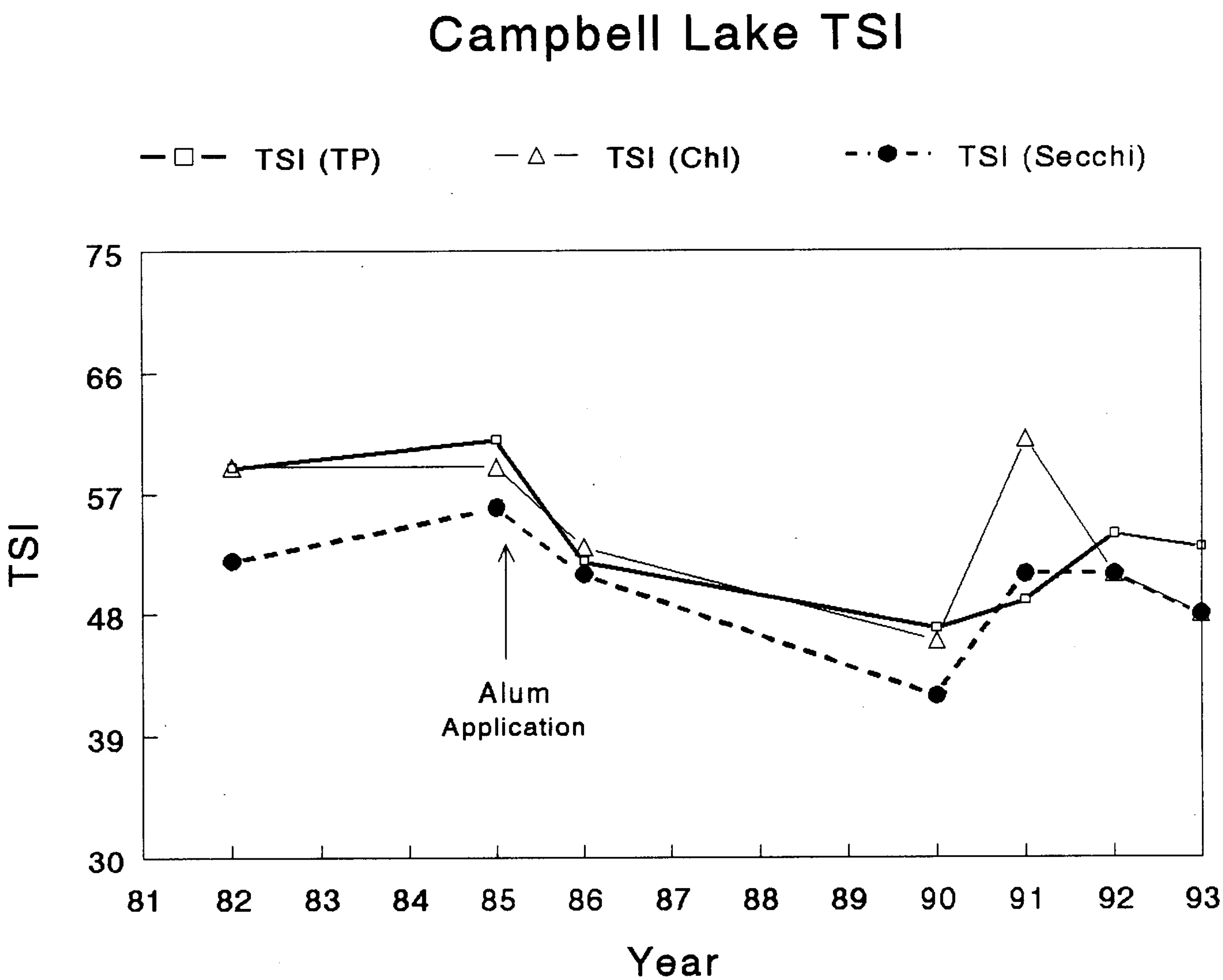

Figure 4. Trophic state index values based on summer means for respective variables in Campbell Lake

the two lakes. Transparency in Erie Lake has been low and similar to the pre-treatment level the past two years in spite of consistently low chl a.

Three basins in Long and Pattison Lakes (Long-north and south and Pattison-north) all showed initial reductions in TSIs to mesotrophic states (Figures 5 and 6) consistent with TP reductions (Table 5). Absolute reductions in mean chl a were 89%, 64% and 40% for the three basins, respectively, while transparency increased 39%, 47% and 87%. Persistence of the initial effect varied, however. While control on TP persisted for at least 8 years in Long-north (Table 5), chl a increased to near the pre-treatment level after year 5 (Figure 5). All three variables increased after 5 years in Long-south (Figure 5) as well, accounting for the 5-year longevity suggested in Table 5. Likewise, the Pattison-north treatment, although effective at reducing internal P loading for 7 years (Table 4), was subsequently considered no longer effective due to high TP and a large algal bloom at depth in 1991. However, mean transparency in Pattison-north during post-treatment years 5-8 still remained 45% greater than pre-treatment levels.

Except for 1985 and 1990 (years 5 and 10 post-treatment), chl a and transparency in Long Lake-Kitsap have remained well below the pre-treatment levels in 1977-1979, although still eutrophic (Figure 7). On an absolute scale, post-treatment summer chl a averaged 53% less than levels in the pre-treatment years and transparency averaged 43% greater during the same nine post-treatment years. Events involving macrophytes during 1985 and 1990, which influenced lake TP and TSIs, were referred to earlier.

Algal Composition

Pre- and post-treatment data on the taxonomic composition of algae are available from only three of the shallow lakes; Erie, Campbell and Long-Kitsap Lakes. Percent of the biovolume represented by blue-green algae before and after treatment in those lakes is shown in Table 6. Although blue-greens are still present, their relative contribution to biomass was substantially reduced initially and has remained less in all three lakes.

Of probably more significance is the near complete disappearance of *Aphanizomenon* in Erie and Campbell Lakes. Although the summer-long blue-green bloom in those lakes was composed almost exclusively of *Aphanizomenon* prior to treatment, that taxon has been nearly unrepresented for at least eight years since the treatment. Although blue-greens are still present, the disappearance of *Aphanizomenon*,

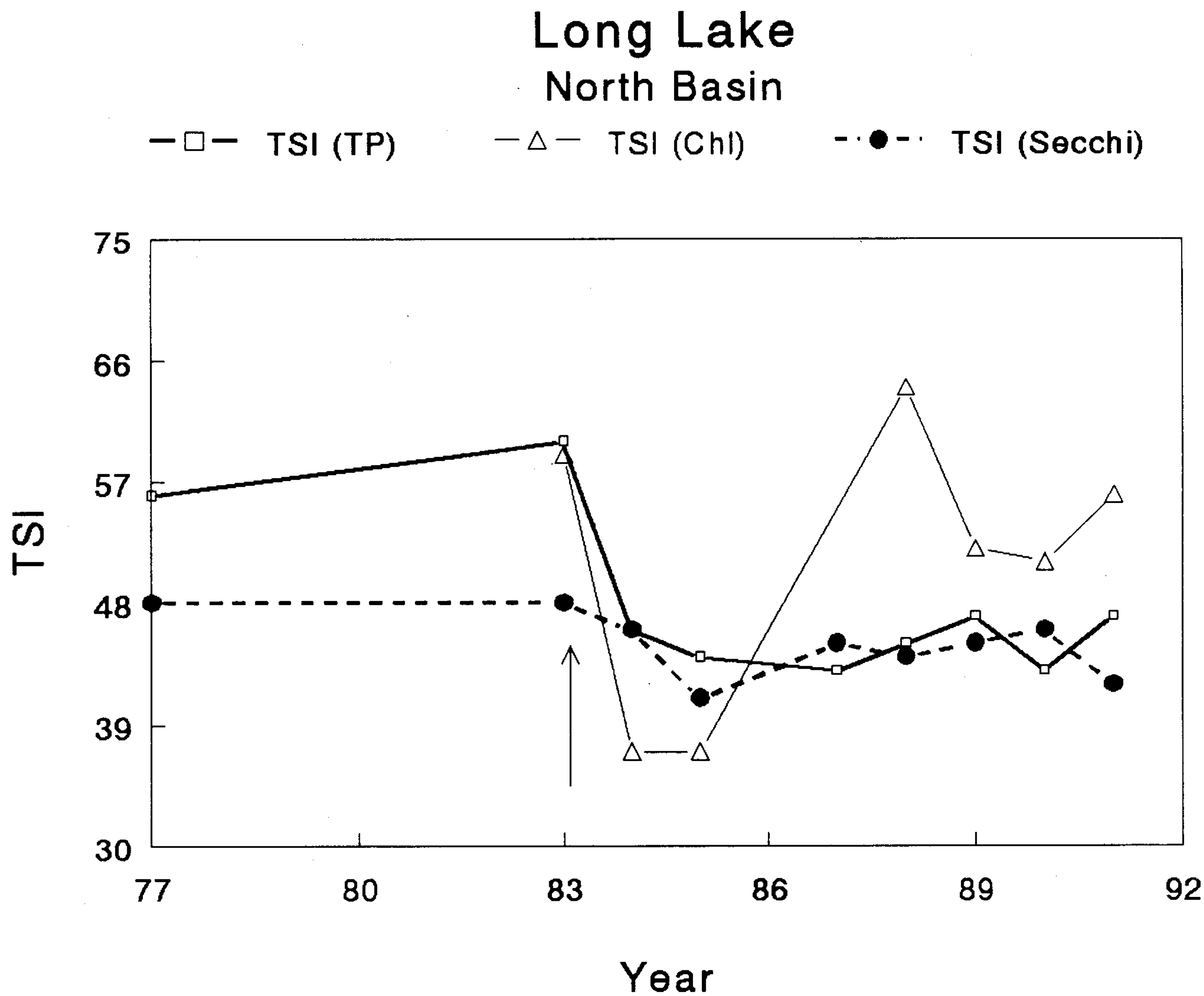

Figure 5. Trophic state index values based on summer means for respective variables in Long Lake, north basin, Thurston County

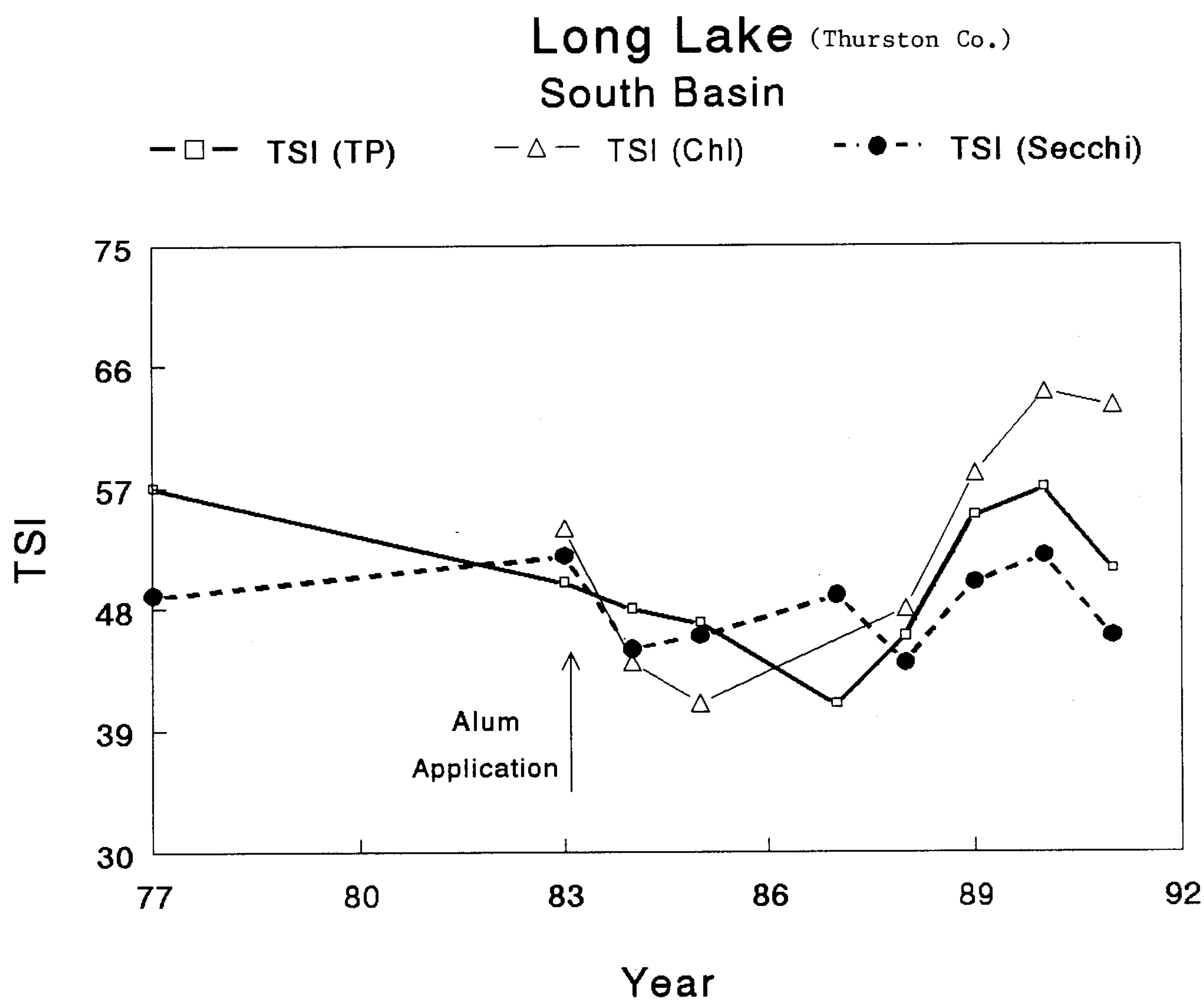

Figure 6. Trophic state index values based on summer means for respective variables in Long Lake, south basin, Thurston County

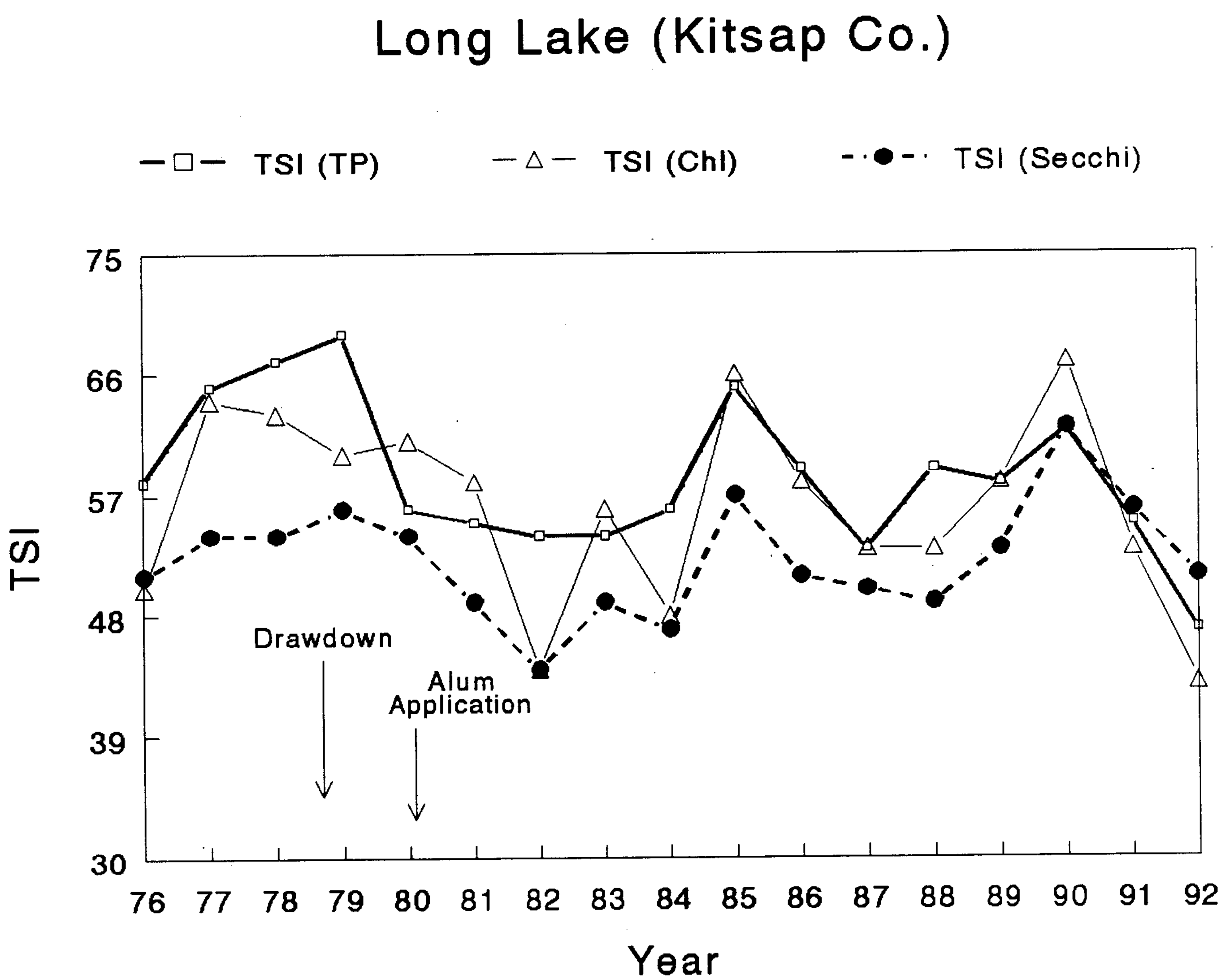

Figure 7. Trophic state index values based on summer means for respective variables in Long Lake, Kitsap County

TABLE 6. PERCENT BLUE-GREEN ALGAL BIOVOLUME DURING SUMMERS IN LONG (KITSAP), ERIE AND CAMPBELL LAKES BEFORE AND AFTER ALUM TREATMENT. YRS - YEARS AFTER TREATMENT.

Lake	Before	Initially After	Latest (yrs)
Long	91	30	58, 55 (8,9)
Erie	38*	1**	10, 48, 13 (5-7)**
Campbell	70*	59**	33, 38, 45 (5-7)**

*Aphanizomenon at 100% of late summer biovolume.

**Aphanizomenon not detected.

coupled with much less biomass (82 and 43% reduction in Erie and Campbell chl a), has greatly enhanced the appearance of those lakes. Prolonged green scums covering the lake surface are no longer present.

The algal composition in Long Lake-Kitsap changed from one of primarily blue-greens to a more mixed assemblage with cryptomonads, diatoms and green algae (Welch et al., 1982). Although still well-represented, blue-greens were dominant for one-fourth of the summer only compared to all summer previously (Table 6). Blue-greens returned to summer-long dominance four years after treatment and were especially abundant in 1985, when high TP returned, but in the other nine years (except 1985, 1990) since treatment they have remained less representative than during pre-treatment years (Table 6).

Treatment effectiveness on the blue-green component cannot be evaluated in the other test lakes because pre-treatment results are unavailable. However, analyses of samples collected in 1990-1991 showed that blue-greens did not represent on average more than about half the algal biovolume in Wapato Lake (20%), Long Lake-north/south (56, 51%) and Pattison Lake-north/south (22, 53%). Qualitative observations indicate that the blue-green component in Long Lake-Thurston may have been substantially less the first year following treatment.

Long Lake-Thurston was treated with the herbicide Sonar four times during July and August of 1991 to control *Myriophyllum*. Although average TP and chl a were not markedly different that summer, Secchi transparency was substantially greater. Also, a large surface bloom of *Aphanizomenon* was observed in both basins of the lake following the herbicide application. Another point of interest, but not linked to the Long Lake herbicide, was a one-half m dense layer of the flagellate, *Gonyostomum*, occurred 0.5-0.7 m above the bottom in Pattison Lake-north on several occasions during the summer of 1991. That phenomenon was never observed in any of the other lakes.

EFFECTIVENESS/LONGEVITY IN STRATIFIED LAKES

Horseshoe Lake, Wisconsin

Horseshoe Lake, located near Manitowoc, WI became eutrophic from agricultural runoff, and also from the direct drainage of the waste lagoon of a cheese and butter factory during 1963-1965. Prior to the dairy discharges, the lake had a sport fishery. Winter fish kills in 1964-1966, severe blue-green algal blooms from

1963-69, and nuisance macrophytes curtailed use of the lake. The dairy plant closed in 1965. In 1970 Horseshoe Lake became the first lake in the United States to receive an alum application (J.O. Peterson, et al., 1973; Table 1)

Alum was effective in lowering volume-weighted, whole-lake mean TP from 140 µg/L in 1966 to 40 µg/L in 1971. In 1982, volume-weighted mean hypolimnetic TP was less than in 1966, but by 1991 hypolimnetic concentrations were very high, increasing from 300 µg/L in June to 975 µg/L in early September (Peterson, 1973; Garrison and Knauer, 1984; Schrieve, 1992). Nevertheless, sediment-P release rate has remained at 50% of the pre-treatment rate (Figure 8).

Trophic state of Horseshoe Lake, based on surface TP, improved from a 1966 value of 71 (hypereutrophic) to 62 and 59 (eutrophic) in 1971 and 1972 (data from Peterson, 1973). The use of copper sulfate before and after alum application prevented a meaningful calculation of TSI values from transparency and chl. The effect of eliminating dairy wastes cannot be entirely separated from the effect of the alum treatment. Longevity and effectiveness of using alum to improve Horseshoe Lake's trophic state following diversion is therefore not known.

Snake Lake, Wisconsin

Wastewater was added to Snake Lake, beginning in 1942, producing a large fish kill in 1942-43 and algal blooms in the years which followed. Diversion was accomplished in 1964 but blooms persisted. Dilution was attempted in 1970 with only a minor reduction in nutrients (Born et al., 1973). In 1969, volume-weighted whole-lake mean TP was 240 µg/L, with occasional values exceeding 2,700 µg/L in bottom, anoxic waters. Alum and sodium aluminate were added in 1972 (Garrison and Knauer, 1984; Table 1).

Whole-lake, volume-weighted mean TP declined to 100 µg/L, and TP in anoxic bottom water declined to 130 µg/L in 1973. Similar values were found in 1982 and 1991. Algal blooms persisted in 1982, due in part to storm sewer runoff (Garrison and Knauer, 1984; Schrieve, 1992). Rates of hypolimnetic P increase declined by 90% and have apparently remained low (Figure 8). These results indicate that alum has controlled sediment P release for 19 years. However, its effects on trophic state cannot be separated from the effects of diversion.

REDUCTION IN INTERNAL PHOSPHORUS LOADING

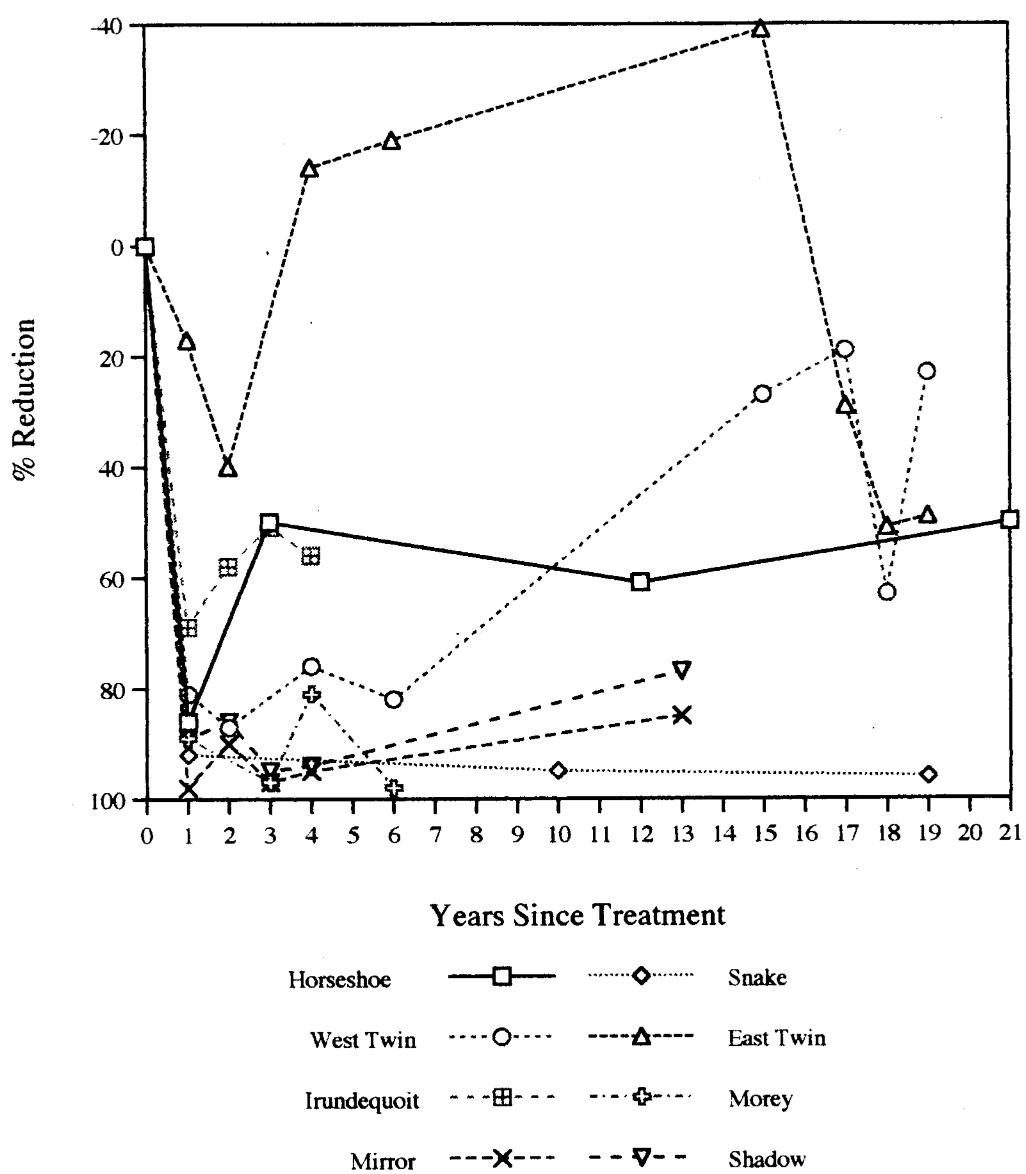

Figure 8. Percent reduction in sediment P release (rate of hypolimnetic P buildup) for seven treated, stratified lakes and one untreated, stratified lake (East Twin)

Eau Galle Reservoir, Wisconsin

Eau Galle is a small Army Corps of Engineers flood control impoundment near Spring Valley, WI (Table 1). Algal blooms occur in the summer, driven by high external and internal P loading, including ground water (Gaugush, 1984). Although no diversion of external loading occurred, the reservoir's hypolimnion was treated with alum in 1986 (Kennedy et al., 1987).

The treatment was effective in controlling sediment P release for one summer (1986), but high external loading of nutrients and silt overwhelmed the effects of alum (James et al., 1991). Monitoring and sediment P release studies in 1991 as part of this project confirmed this conclusion. Eau Galle Reservoir is a case history which clearly demonstrates the futility of an alum treatment without abatement of external loading, especially in highly flushed systems.

Mirror and Shadow Lakes, Wisconsin

Mirror and Shadow Lakes are located in Waupaca, WI (Table 1). Storm water drainage to them began in the 1930s, and by the 1960s residents of the area complained of winter fish kills and odors from algal blooms. In 1972-73, storm sewer loading accounted for 65% and 58% of annual external P loading to Mirror and Shadow Lakes, respectively. Diversion of storm drainage was completed in 1976, reducing the watersheds of Mirror and Shadow to 13.1 ha and 56.7 ha, respectively. Both watersheds are primarily residential, although Shadow has drainage from some undeveloped areas. Diversion reduced external P loading to Mirror from 0.34 to 0.12 $g/m^2 \cdot y$, and from 0.24 to 0.10 $g/m^2 \cdot yr$ to Shadow. Mirror Lake exhibited brief and weak spring/fall mxing due to its very small surface area relative to mean depth (Osgood Index, $\bar{z} / \sqrt{A_0}$, where A_0 is area in Km^2 = 35), so artificial circulation was used in November and at ice-out to prevent winter fish kills. Spring TP concentration remained almost unchanged after storm sewer diversion due to sediment P release. Alum was added to the hypolimnion of both lakes in May 1978 (Garrison and Knauer, 1983; Garrison and Ihm, 1991).

Diversion immediately reduced epilimnetic TP, mean volume-weighted TP, and volume-weighted dissolved reactive P. However, concentrations remained in the range expected for eutrophic lakes and it was hypothesized that internal loading prevented further improvement. The alum application reduced mean volume-weighted TP from about 93 μg/L to about 20 μg/L in Mirror and from 55 μg/L to 23μ g/L in Shadow

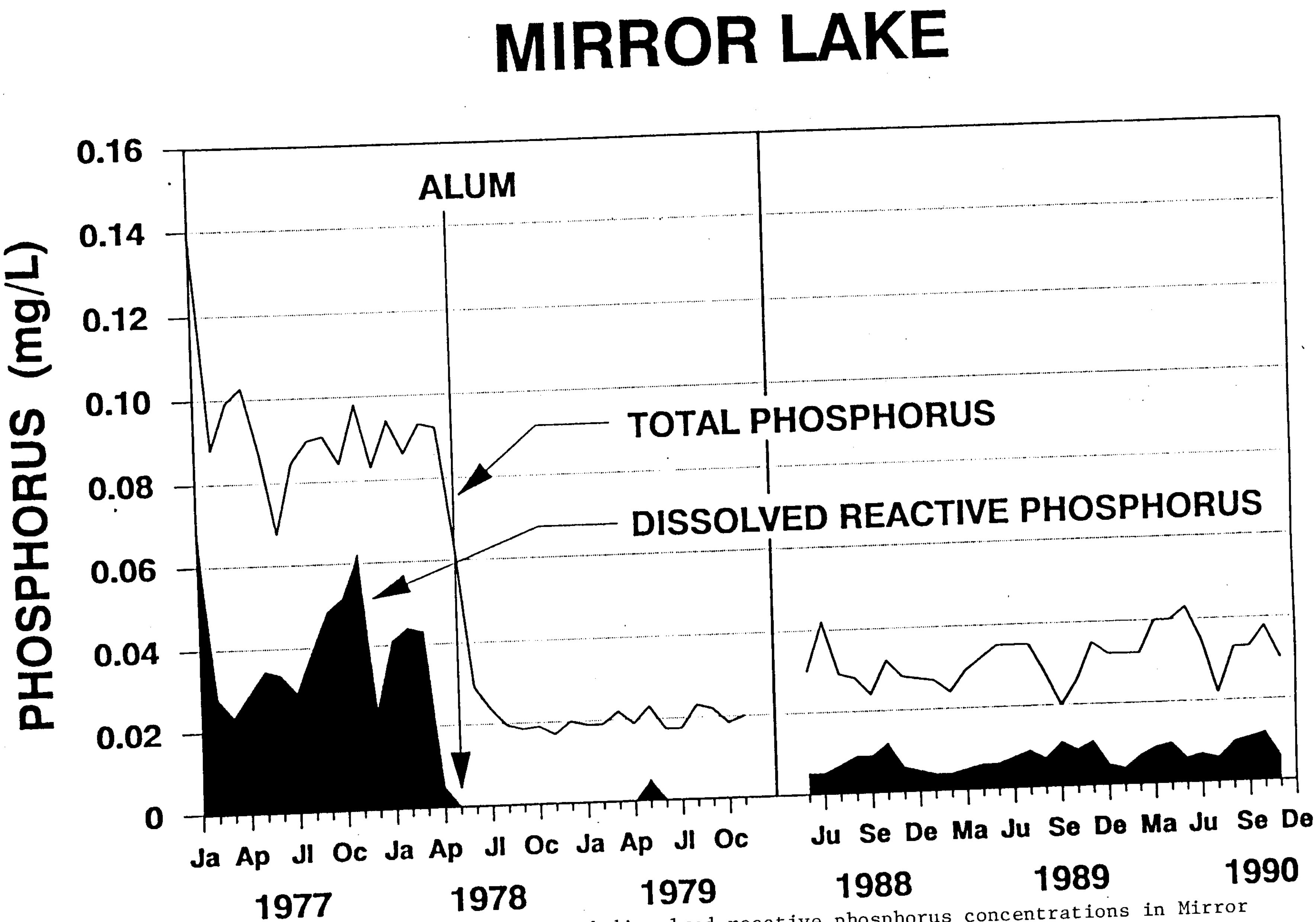

Figure 9. Changes in volume-weighted total and dissolved reactive phosphorus concentrations in Mirror Lake, WI before and after alum application (from Garrison and Ihm, 1991)

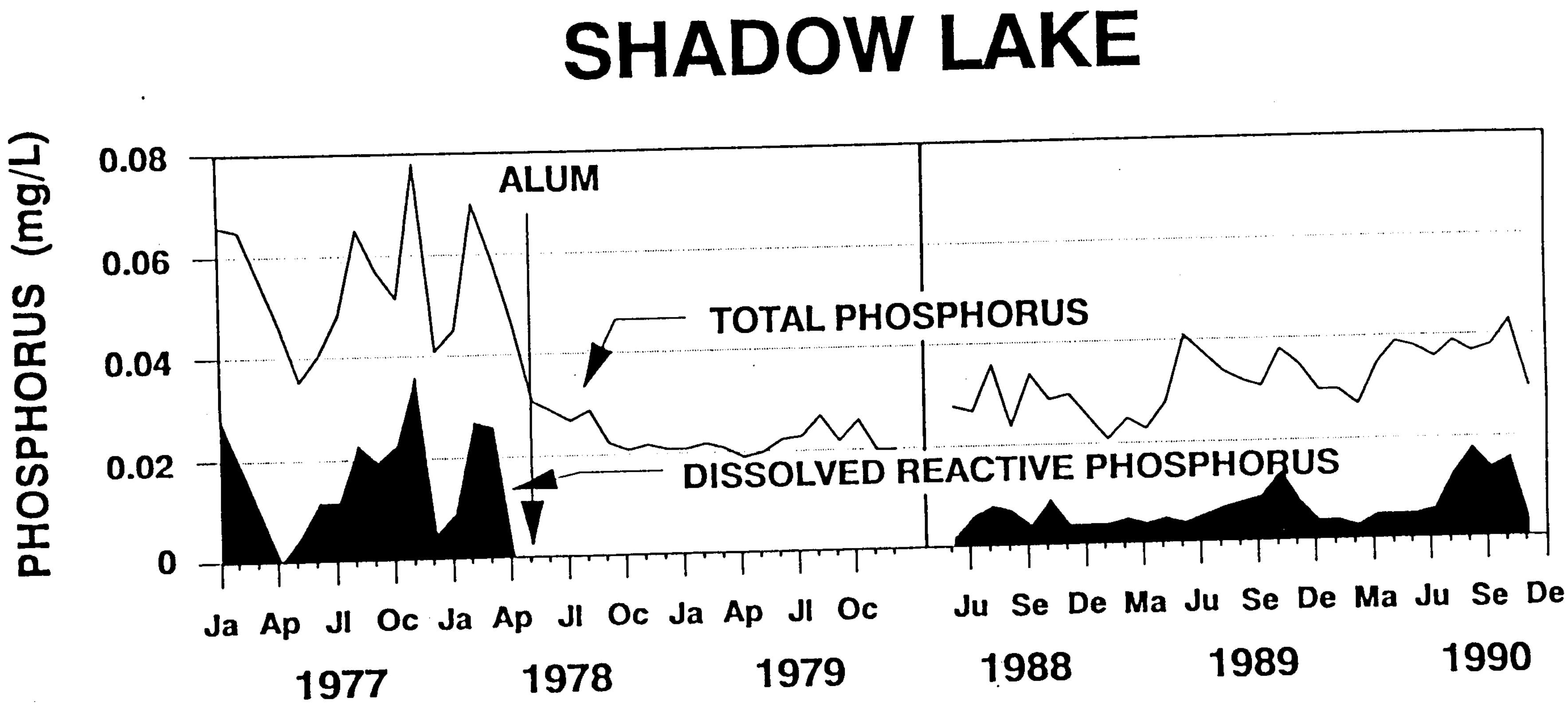

Figure 10. Changes in volume-weighted total and dissolved reactive phosphorus concentrations in Shadow Lake, WI before and after alum application (from Garrison and Ihm, 1991)

Lake (Figures 9 and 10). These lower concentrations, due primarily to a reduction in hypolimnetic P, remained at least three years. Sediment P release rates, determined with in situ release chambers, were lowered 90-95% (Table 7; Figure 8), and remained low through 1990 (12 years). The 1991 rates (Table 7) were obtained from sediment cores in the laboratory (this study) and may not be comparable to the in situ values. Maximum TP concentrations in the hypolimnion of Mirror Lake were less than 100 μg/L for three years after alum treatment (compared to more than 500 μg/L before treatment). In 1990 Mirror's maximum hypolimnetic TP was about 300 μg/L (Garrison and Knauer, 1983; Garrison and Ihm, 1991). In July 1991 (this study), TP in the stratum above the hypolimnetic sediments was 121 μg/L in Mirror and 106 μg/L in Shadow. These modest increases, along with the very small increases in P release rates through 1990 (Table 7), strongly suggest that the layer of $Al(OH)_3$ (clearly visible in 1991 (this study) at the 8-10 cm depth in sediment cores from both lakes), remains highly effective in retarding sediment P release 13 years after application.

Figures 11 and 12 illustrate changes in the TSI of Mirror and Shadow Lakes following diversion and alum application (data from Garrison and Knauer, 1983; Garrison and Ihm, 1991; this study). Diversion from Mirror Lake improved trophic state (based on TP) from 62 (1972-1974 mean), typical of eutrophic lakes, to 52 in 1977. After the alum application, trophic state fell from 52 to 43 in 1978 and has remained at that level (borderline oligotrophic) through 1991 (13 years). After the alum application, there has been a close correspondence of the three TSI values (based on TP, chl, and transparency), suggesting P limitation of algal biomass.

The effects of storm water diversion on trophic state improvement in Mirror and Shadow Lakes cannot be separated from the effects of the alum treatment. Without diversion, algal blooms would have continued, and the alum application would have had little impact on trophic state. Because these lakes are deep relative to surface area, and are surrounded by trees and bluffs (especially Mirror), wind-driven vertical entrainment of P-rich hypolimnetic waters during the summer may be small, though significant diffusion from hypolimnion to epilimnion is possible. Alum may have acted to improve these lakes by maintaining low spring overturn concentrations, which had remained unchanged after storm sewer diversion (Garrision and Knauer, 1983), and by eliminating the steep gradient of P between hypolimnion and epilimnion and thereby eliminating P diffusion as a major internal source to the epilimnion. Significant and permanent diversion of external loading, coupled

TABLE 7. PHOSPHORUS RELEASE RATES DETERMINED FROM IN SITU P RELEASE CHAMBERS (FROM GARRISON AND IHM, 1991)

	P Release Rate mg/m²·day	
Year	Mirror Lake	Shadow Lake
1977	1.30	1.30
1978 (alum)	0.03	0.14
1979	0.13	0.18
1980	0.04	0.07
1981	0.07	0.08
1990	0.20	0.30
*1991	5.83	2.43

*determined from sediment cores, incubated for 16 days at room temperature under anoxia (this study).

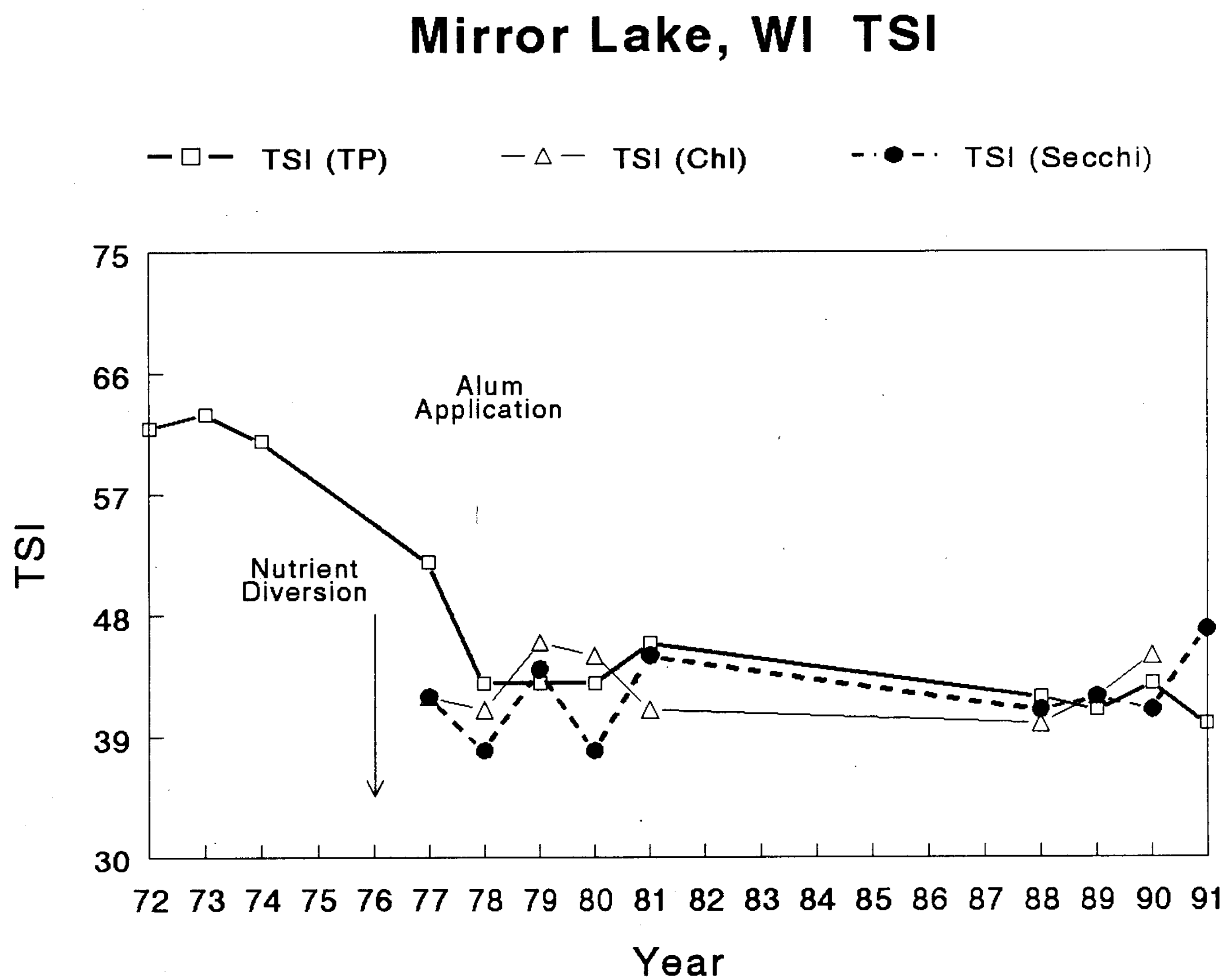

Figure 11. Changes in mean trophic state (TSI) variables of Mirror Lake, WI following diversion of storm water in 1976 and alum application in 1978

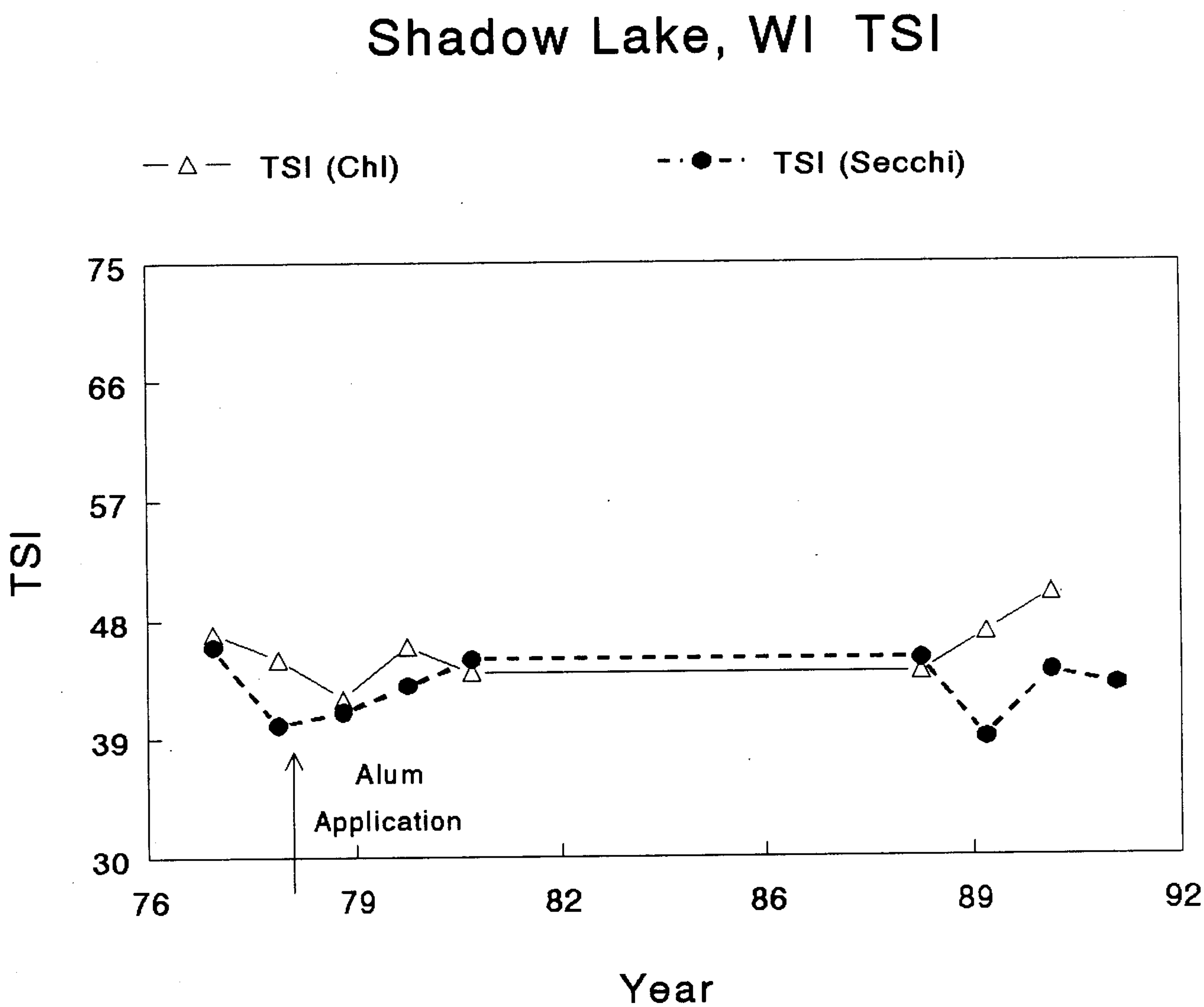

Figure 12. Changes in mean trophic state (TSI) variables in Shadow Lake, WI following storm water diversion in 1976 and alum application in 1978

with a highly effective (90-95% reduction in sediment P release) and long lasting (at least 13 years) alum treatment, have improved these lakes to near pre-disturbance conditions.

West Twin and East Twin Lakes, Ohio

East and West Twin Lakes, OH are of glacial origin ("kettle lakes") and found in the same residential and forested watershed as Dollar Lake (Table 1). In the late 1960s, the lakes were very eutrophic (TSI 60-70) from prolonged septic drainfield seepage, either into ditches flowing to the lake or directly into the lake. Extensive home construction, including wetland destruction through dredging, was also a major nutrient source. Wastewater was diverted from the watershed in 1971-1972. Recovery of the lakes was expected to be delayed because internal loading, especially into the anoxic hypolimnion, was extensive. Mass balance and hypolimnetic accumulation rates were used to determine this (Cooke et al., 1977, 1993b). Alum was applied to West Twin's hypolimnion in July 1975, using the maximum dose of 26.0 g Al/m^3, as determined by the method of Kennedy and Cooke (1982). East Twin, a nearby (200 m), downstream lake was used as a reference or untreated lake because of its identical history and highly similar morphometry, hydraulics, and biota. The hypothesis was that West Twin would recover sooner from diversion because of its reduced internal loading rate following alum application. This unique experiment-control (but see Edmondson (1993) for comments about whole-lake "experiments") situation allowed a separation of the impact of the alum treatment on trophic state from the impact of nutrient diversion (Cooke et al., 1977, 1978, 1982, 1993b).

The $Al(OH)_3$ layer was visible on the sediment surface in 1975, following treatment, as a white "blanket." In 1989, 1991, and 1992 it could be distinguished visually in sediment cores at about 10 cm below the sediment-water interface. In 1993 it could not be seen. The longevity and effectiveness of this $Al(OH)_3$ layer in controlling P release during anoxic conditions was determined by computing net P release in the 10-11 m stratum, the deepest zone of either lake, and by incubating sediment cores in the laboratory under oxic and anoxic conditions. Table 8 and Figure 8 show a comparison of net sediment P release rates of the 10-11 m stratum for West and East Twin Lakes during the early June - late August interval of lake thermal stratification. Prior to alum application in 1975, release rates were slightly higher in East Twin. After the treatment of West Twin, East Twin's release rates were 4-7 times greater through 1980 (5 years). There were insufficient data to with a highly effective (90-95% reduction in sediment P release) and long lasting (at least 13 years) alum treatment, have improved these lakes to near pre-disturbance conditions.

West Twin and East Twin Lakes, Ohio

East and West Twin Lakes, OH are of glacial origin ("kettle lakes") and found in the same residential and forested watershed as Dollar Lake (Table 1). In the late 1960s, the lakes were very eutrophic (TSI 60-70) from prolonged septic drainfield seepage, either into ditches flowing to the lake or directly into the lake. Extensive home construction, including wetland destruction through dredging, was also a major nutrient source. Wastewater was diverted from the watershed in 1971-1972. Recovery of the lakes was expected to be delayed because internal loading, especially into the anoxic hypolimnion, was extensive. Mass balance and hypolimnetic accumulation rates were used to determine this (Cooke et al., 1977, 1993b). Alum was applied to West Twin's hypolimnion in July 1975, using the maximum dose of 26.0 g Al/m^3, as determined by the method of Kennedy and Cooke (1982). East Twin, a nearby (200 m), downstream lake was used as a reference or untreated lake because of its identical history and highly similar morphometry, hydraulics, and biota. The hypothesis was that West Twin would recover sooner from diversion because of its reduced internal loading rate following alum application. This unique experiment-control (but see Edmondson (1993) for comments about whole-lake "experiments") situation allowed a separation of the impact of the alum treatment on trophic state from the impact of nutrient diversion (Cooke et al., 1977, 1978, 1982, 1993b).

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TABLE 8. PHOSPHORUS RELEASE RATES FROM DEEP WATER (10-11 M) SEDIMENTS IN EAST AND WEST TWIN LAKES, OH (mg P/m^2·day) DURING JUNE THROUGH AUGUST

Year	West Twin	East Twin
1972	2.55	3.30
1973	4.24	2.83
1974	1.51	2.80
1975 (Pre-alum)	2.68	2.76
1975 (Post-alum)	-0.53	2.44
1976	0.37	1.76
1978	0.67	3.34
1980	0.49	3.47
1989	2.02	4.05
1991	2.22	2.07
1992	1.01	1.42
1993	2.13	1.49

compute a release rate between 1980-1988. In 1989 (14 years after treatment), some effectiveness remained. By 1991 there appeared to be little difference in release rates between the two lakes..

Table 9 shows a comparison of P release rates from intact sediment cores, under oxic and anoxic laboratory conditions. East Twin's anaerobic release rate in 1989 was 2.6 times greater than West Twin's, a ratio very similar to that of the *in situ* release rate difference in 1989 (Table 8). This suggests that the alum layer retained some effectiveness for 14 years. Under aerobic laboratory conditions, sediments of both lakes had identical net release rates of P in 1989, suggesting iron control of their P cycles (see also results on Fe/P). Extensive determination of P release in cores collected in 1992 failed when dissolved oxygen could not be eliminated from the cores which were intended to be anaerobic.

Figures 13-16 illustrate the changes in the lakes following diversion of wastewaters, cessation of most home construction by 1980, and the 1975 alum application to West Twin's hypolimnion. Alum produced a sharp decline in West Twin's mean hypolimnetic P concentration which lasted until at least 1980 (5 years) (Figure 13). By 1989, mean hypolimnetic P was identical in the two lakes, and remained highly similar through 1993. The very low standard errors of the mean concentration in West Twin from 1975-1980 demonstrate its consistently low hypolimnetic TP concentration over the summer, even as the redox potential presumably fell steadily during the continuously anoxic summer period. East Twin's hypolimnetic TP increased steadily through the summer, as expected during a continuously anoxic period. However, from 1989-1993, West Twin's hypolimnion once again demonstrated a steadily increasing TP over the summer's duration (Figure 13), indicating a reduction in the effectiveness of the alum treatment at some point between 1980 and 1989.

West Twin was expected to improve in trophic state before East Twin because it was assumed that the P-rich hypolimnion of East Twin would provide a P subsidy to its epilimnion via vertical entrainment and diffusion. Figure 14 shows that this hypothesis must be rejected. Mean epilimnetic TP concentrations of the treated and untreated lakes were essentially the same after alum application. East and West Twin trophic states, based on surface water samples, recovered at the same rates following diversion. Inflows to East Twin from the treated West Twin were minimal or absent during dry summer months, eliminating dilution by West Twin as a cause of East Twin's improvement. Therefore, diversion of wastewater appears to have been the major

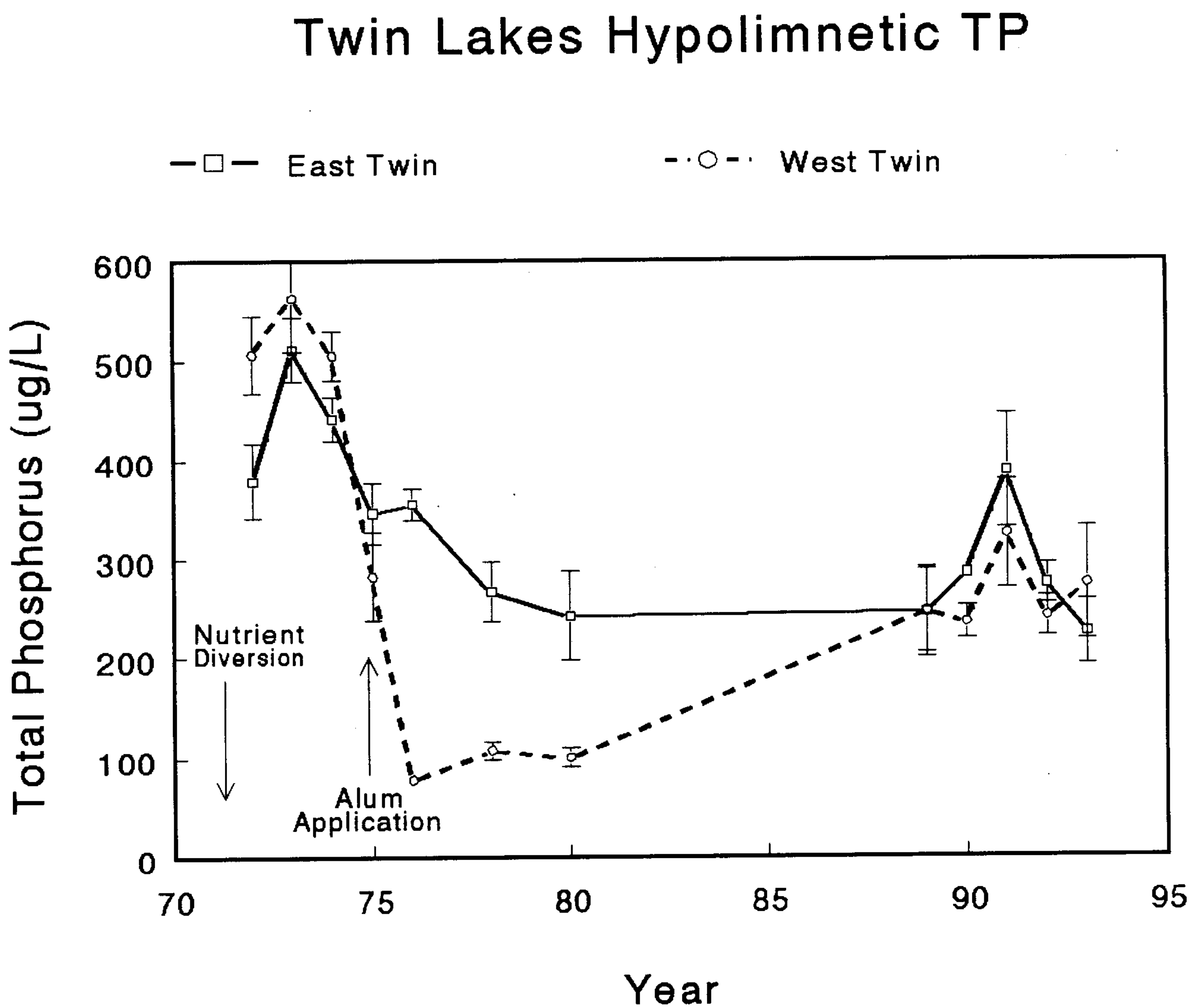

Figure 13. Changes in total phosphorus concentration at 10 meters in East and West Twin Lakes, OH following nutrient diversion and alum application to West Twin

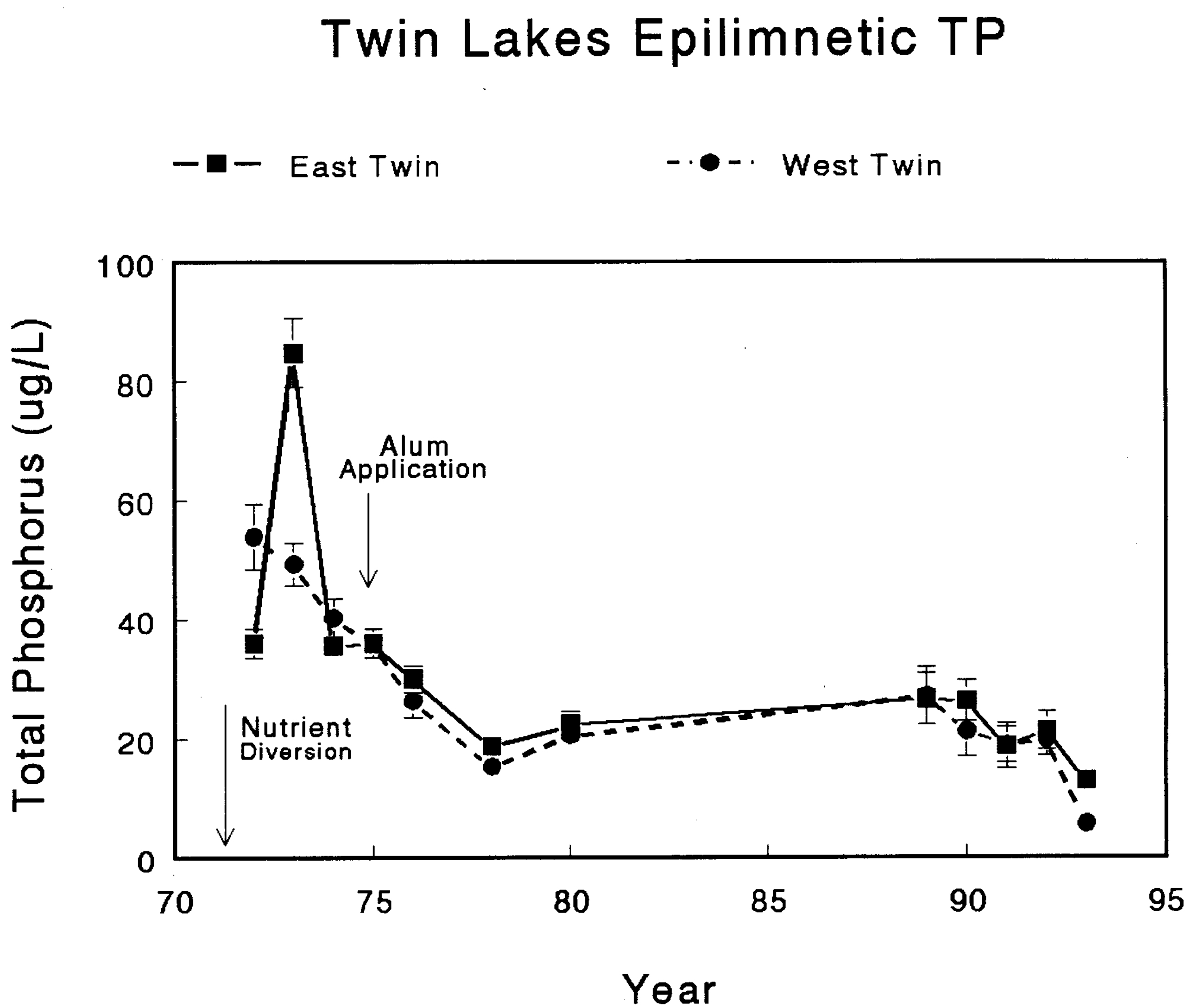

Figure 14. Changes in epilimnetic (1 meter) total phosphorus concentration in East and West Twin Lakes, OH following nutrient diversion and alum application to West Twin

factor contributing to the simultaneous lake improvements. Even though the alum application was effective in lowering P release from hypolimnetic sediments of West Twin, the use of the control lake has shown that apparently this was not a major factor in reducing the average summer epilimnetic TP concentration. An analysis of vertical P transport mechanisms in these lakes may provide additional information about West Twin's response to alum application.

Figures 15 and 16 illustrate changes in the TSI of the lakes before and after diversion and alum application. The TSI values based on TP were above 60, and TSI values from chl were above 70 (hypereutrophic) prior to and just after diversion. The wide deviation between mean TSI values for chl and TP in both East and West Twin is probably due to the existence of blue-green "scums" (*Aphanizomenon, Anabaena, Microcystis*), which put much more algal biomass in surface waters during daytime samples throughout the study. By 1980, trophic state in both lakes was about 50 (marginally eutrophic) for TP and transparency, again indicating the major role of nutrient diversion in improving the lakes. The lakes have continued to change, and in 1993 they had trophic states of about 40 (borderline oligotrophic).

The increasingly clear waters of East and West Twin may have allowed the development of a greatly increased area of submergent plants, mainly *Myriophyllum spicatum* and *Ceratophyllum demersum*. This also appears to be unrelated to the alum application because both lakes improved at the same rate and both have macrophyte problems.

The Twin lakes experiment, unique among alum treated lakes, demonstrated that nutrient diversion had far greater impact on lake trophic state improvement than the alum treatment. Spring algal blooms in West Twin Lake were generally much less than East Twin's, suggesting that the alum treatment did have an impact at that time period.

Dollar Lake, Ohio

Dollar Lake is a small, dimictic, alkaline bog lake located adjacent to East Twin Lake (Table 1). Urban drainage, contaminated with wastewater from septic tanks, stimulated large blue-green algal blooms. Septic tank flows were diverted in 1971-1972. Dollar Lake was alum-treated in 1974, the world's first alum treatment

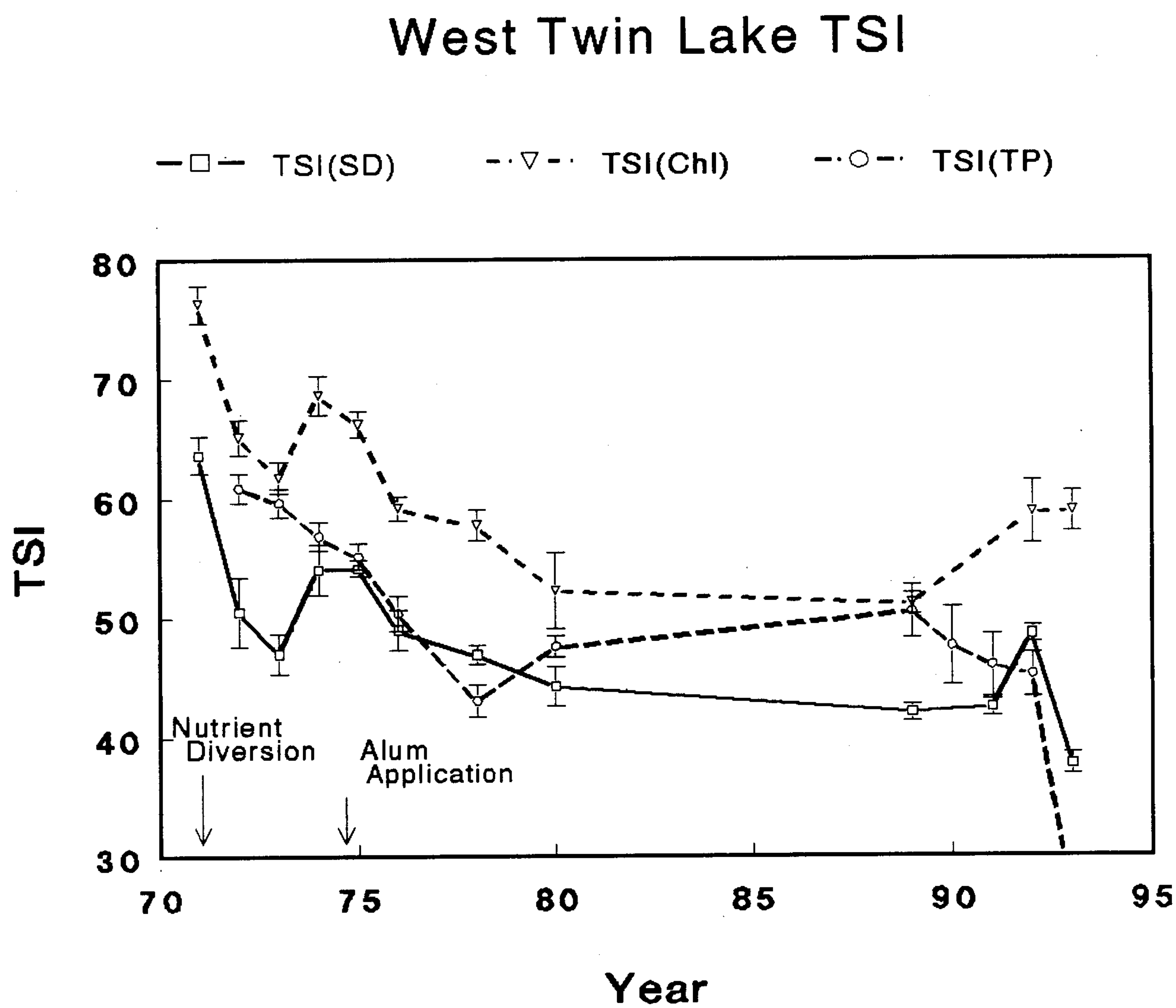

Figure 15. Changes in mean trophic state (TSI) variables of West Twin Lake, OH following nutrient diversion and alum application

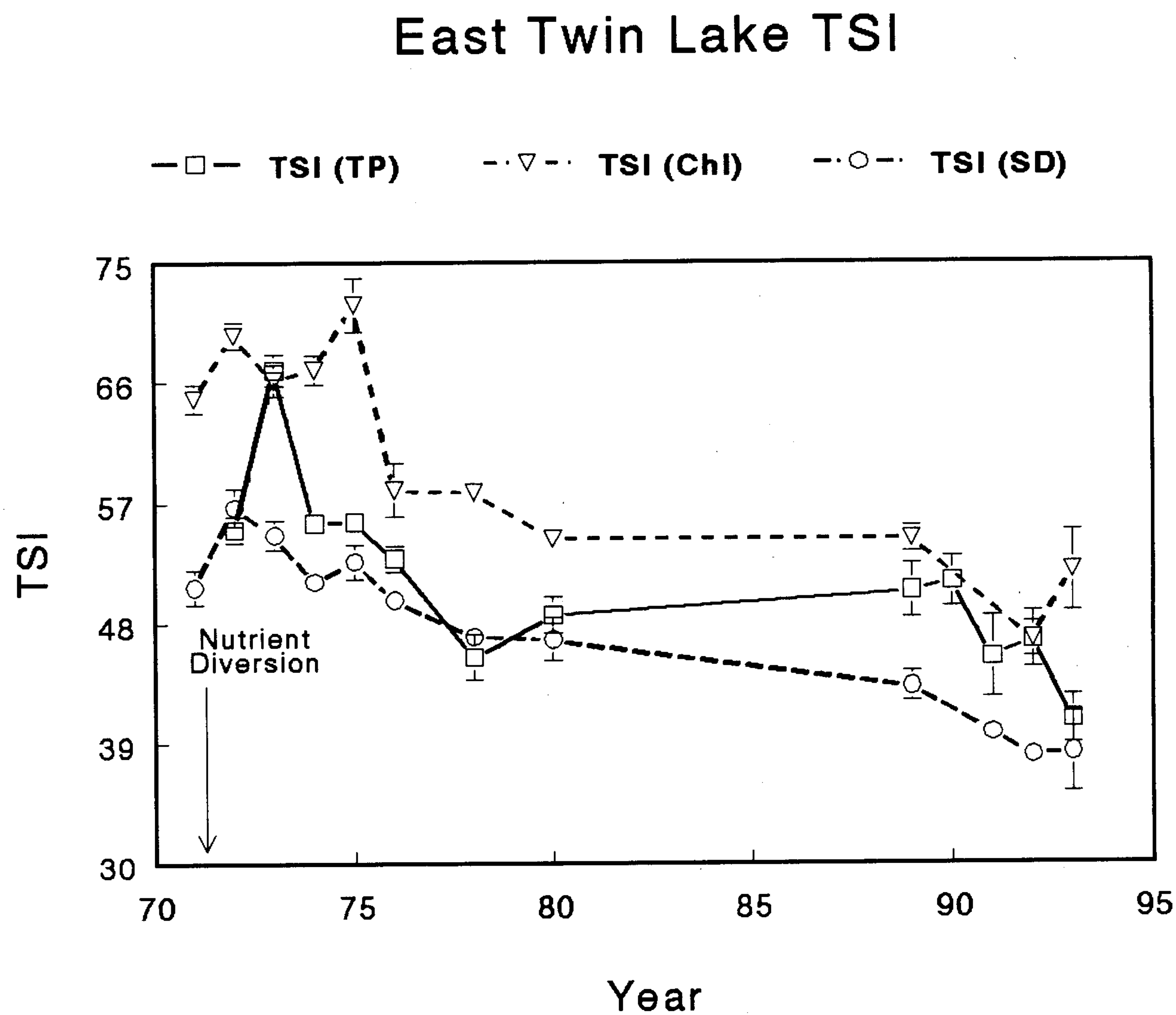

Figure 16. Changes in mean trophic state (TSI) variables of East Twin Lake, OH following nutrient diversion and alum application

of a lake's hypolimnion. A light treatment (10% of total dose) was also made to the lake's surface (Kennedy, 1978).

Figures 17 and 18 illustrate the changes in Dollar Lake following diversion and alum application, though the relative effects of these events cannot be separated. The lake was very eutrophic in 1968-1973 (TP TSI = 65). Alum lowered hypolimnetic TP in 1974, an effect which lasted until at least 1980 (no data available for 1981-1991). Hypolimnetic TP in 1991-1993 increased by a factor of 2-4 from 1980 levels. Surface TP was greatly reduced between 1974 and 1975, following alum application, and has remained low through 1993. Chl fell and transparency increased. Lake trophic state has deteriorated slightly in recent years, as indicated by TSI values for transparency and TP (Figure 19). Chl has increased, as evidenced by surface blooms of colonial blue-greens in 1992-1993. Except for this problem, the lake's trophic state in 1993 is borderline eutrophic, a state which is similar to 1975, the first post-alum year (Figure 19).

The significance of the alum treatment in improving lake trophic state cannot be separated directly from the effect of diversion. However, in early summer 1974, prior to the late July alum application, surface TP of Dollar Lake averaged 90.8 ± 6.3 µg/L. This value is similar to the 1973 summer mean of 81.6 ± 14.2 µg/L. After the alum addition, 1974 surface TP averaged 24.3 ± 2.7 µg/L, a value very similar to surface TP in all post-treatment years through 1993. The initial reduction of epilimnetic TP was probably due to the light treatment of the lake's surface. Low epilimnetic TP in subsequent years may be due to low external loading and to low spring TP and lowered hypolimnetic TP from the effects of alum. Mean hypolimnetic TP nearly doubled between 1992 and 1993, and mean surface TP also increased in that interval, suggesting the effectiveness of alum in Dollar Lake has ended after 18 years and/or that external loading has increased. It also suggests that vertical P transport in this lake is significant even though its morphometry (Osgood Index = 27.6) indicates that wind-driven entrainment is unlikely.

<u>Irondequoit Bay, New York</u>

Irondequoit Bay is the largest (6.79 km^2) of the alum-treated lakes discussed in this report (Table 1). Urban and agricultural drainage from the Rochester area produced eutrophic conditions. Wastewater effluents and dry weather combined sewer discharges were diverted from the watershed in1978-80, and by 1986

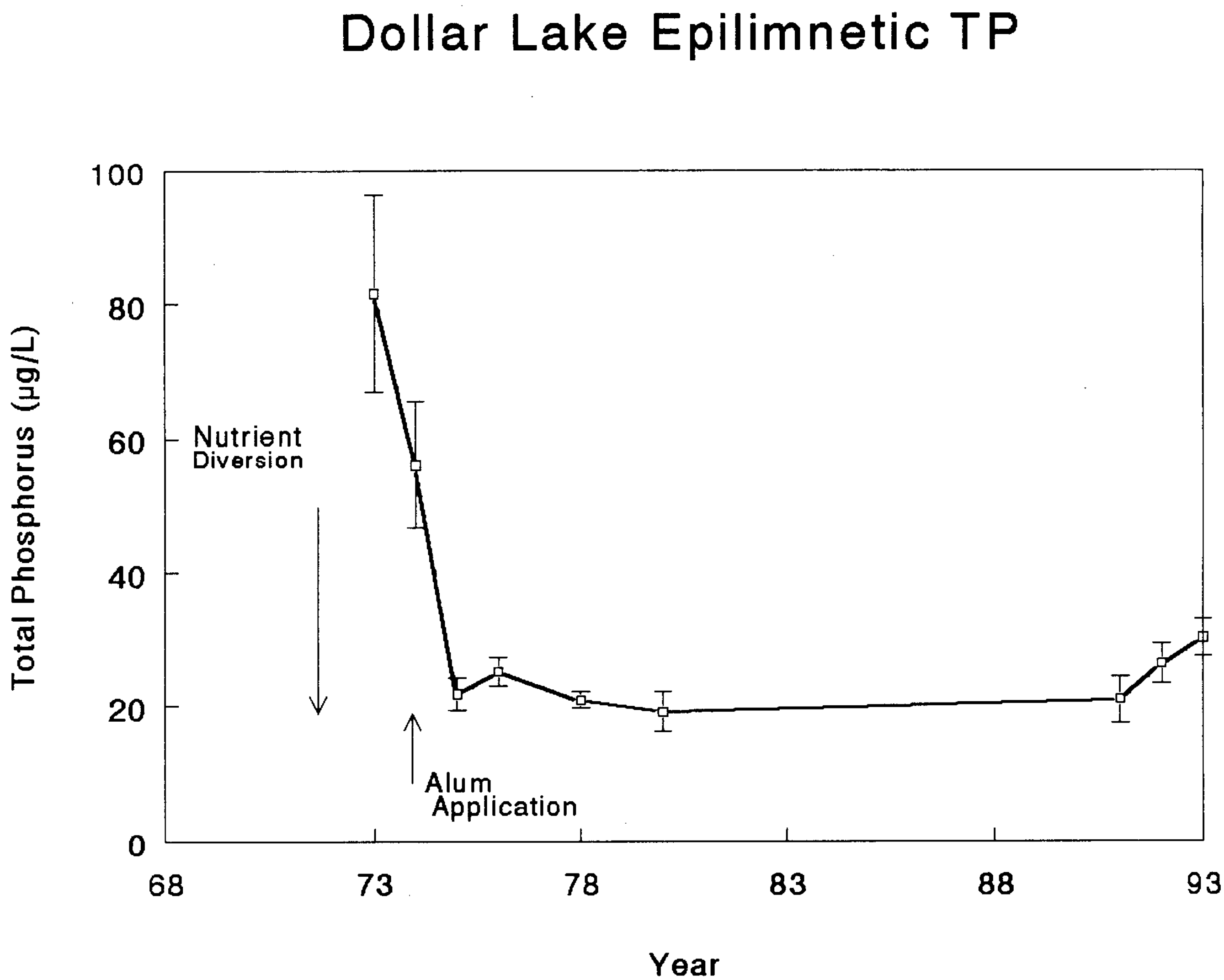

Figure 17. Changes in epilimnetic (1 meter) total phosphorus concentrations in Dollar Lake, OH following nutrient diversion and alum application

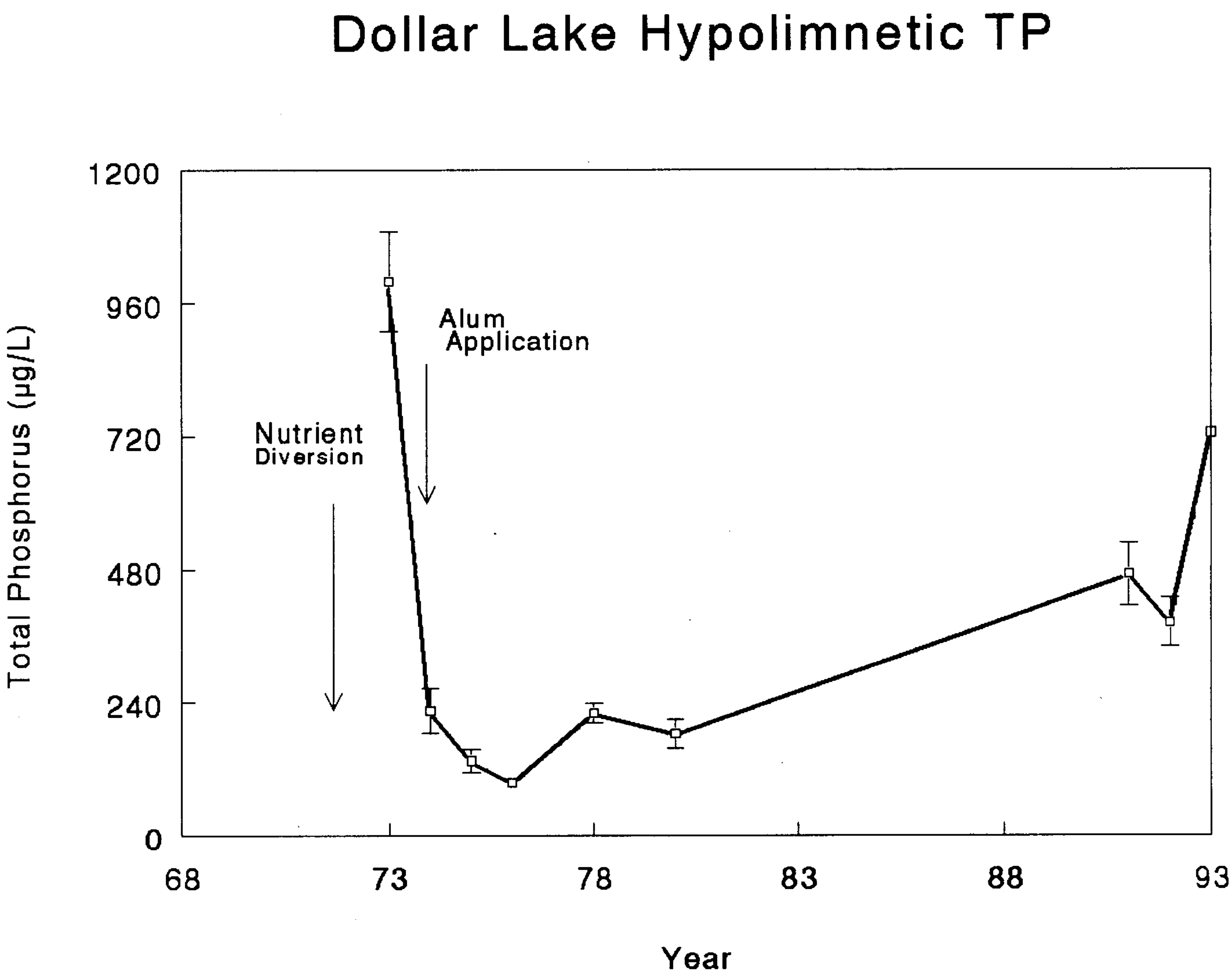

Figure 18. Changes in hypolimnetic (6 meters) total phosphorus concentrations in Dollar Lake, OH following nutrient diversion and alum application

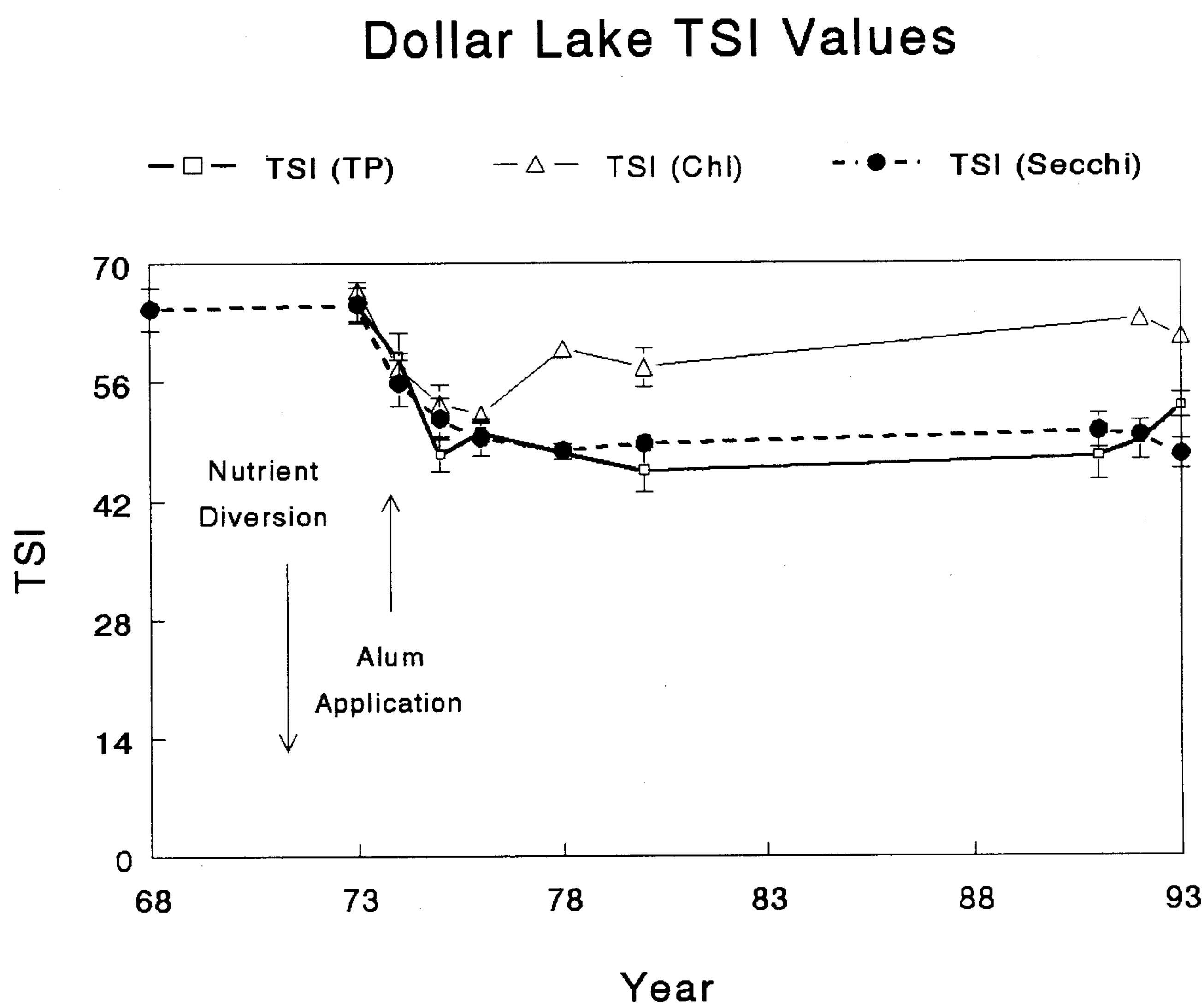

Figure 19. Changes in Dollar Lake, OH mean trophic state (TSI) variables following nutrient diversion and alum application

combined sewer storm overflows were eliminated. Blue-green algal blooms were common during summer months. Diffusive transfer of P from metalimnetic and hypolimnetic sediments was estimated to be 16 $mg/m^2 \cdot day$ during summer prior to alum treatment. Eddy diffusion was believed to be a mechanism which transported P to epilimnetic waters. The large lake surface area relative to depth (Osgood Index of 2.66) suggests a high probability of vertical entrainment in Irondequoit Bay. Alum was applied to metalimnetic and hypolimnetic sediments (about 45% of the lake's total area) at a dose of 28.7 g Al/m^3, between 1 July and 9 September 1986 (Spittal and Burton, 1991).

Hypolimnetic volume-weighted TP declined following diversion, declined again sharply in 1986-87 after the alum application, and then increased slightly in 1989-1990 (Figure 20). P release rates, calculated from hypolimnetic P accumulation (Table 10), declined sharply from a 1982-1985 pretreatment mean of 14.4 $\pm$ 4.2 $mg/m^2 \cdot day$ to a 1987-1990 post-treatment mean of 8.1 $\pm$ 0.74 $mg/m^2 \cdot day$ (data for calculations from L. Spittal, Monroe County, NY Department of Health, Rochester, NY, personal communication). Post-treatment rates appear to have increased slightly each year (Table 9 and Figure 8). Effectiveness in controlling sediment P release has lasted at least 4 years (no post-1990 data), although net sediment P release rates after application would be considered to be high (c.f. Tables 8, 9, and 11 with Table 10; also see Nurnberg, 1984 for a range of values). Epilimnetic mean TP fell following diversion and may not have been greatly affected by the alum application because mean concentrations in 1987-1989 were almost identical to the summer 1984 mean (Figure 21).

Changes in trophic state variables following diversion and alum application are summarized in Figure 22. Chl (as TSI) concentrations peaked in 1985, fell every year through 1988, and then increased. Transparency was primarily influenced by chl (Figure 22). The lake's trophic state based on TP changed little following the diversions from 1978 through 1986. A slight improvement to a TSI of about 55 (mildly eutrophic) occurred after alum was added.

Alum application to Irondequoit Bay does not appear to have greatly influenced trophic state, despite the high dose, some control of sediment P release, and the suggestion that vertical entrainment might be significant. The relatively high epilimnetic TP suggests that external P loading and/or continued sediment P release have maintained this lake's eutrophic condition.

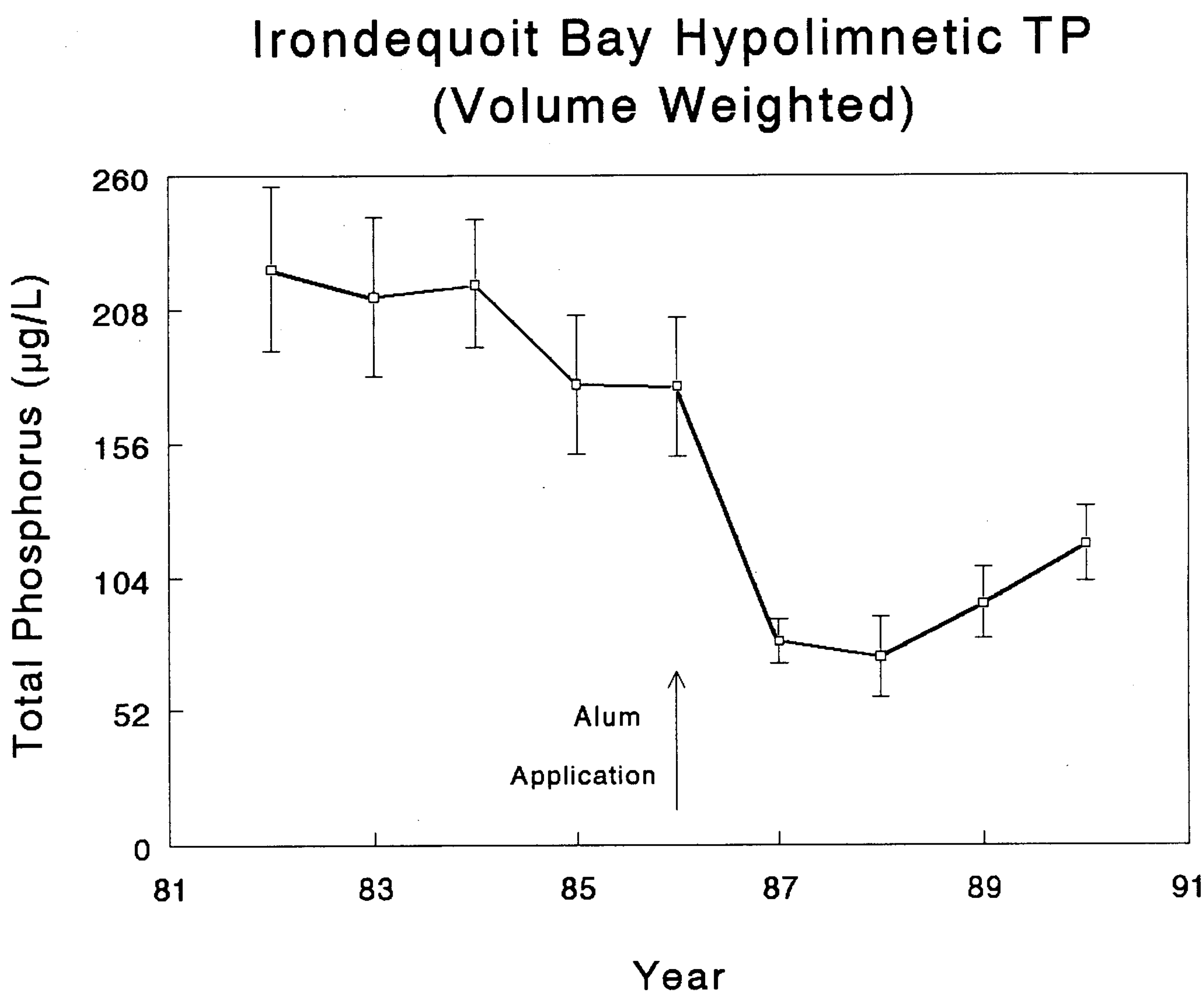

Figure 20. Changes in volume-weighted hypolimnetic total phosphorus in Irondequoit Bay, NY following storm water diversion (1978-1986) and alum application

TABLE 9. MEAN (±/S.D.) INTACT CORE P RELEASE RATES (MG P/M^2·DAY) DURING 5 DAY INCUBATION IN 1989

	ANAEROBIC	AEROBIC
West Twin (N = 3)	1.69 ± 0.13	-4.22 ± 0.23
East Twin (N = 3)	4.35 ± 2.66	-4.65 ± 0.15

TABLE 10. HYPOLIMNETIC PHOSPHORUS ACCUMULATION RATES IN IRONDEQUOIT BAY, NY

	$mg/m^2 \cdot day$
1982	21.8
1983	20.1
1984	17.4
1985	18.4
1986	alum application
1987	6.0
1988	8.1
1989	9.5
1990	8.6

TABLE 11. HYPOLIMNETIC PHOSPHORUS ACCUMULATION RATE (11-13 METER CONTOUR) IN LAKE MOREY, VT.

	$mg/m^2 \cdot day$
1981	5.05
1982	3.32
1986	alum application
1987	0.48
1989	0.14
1990	0.81
1991	0.43
1992	0.08

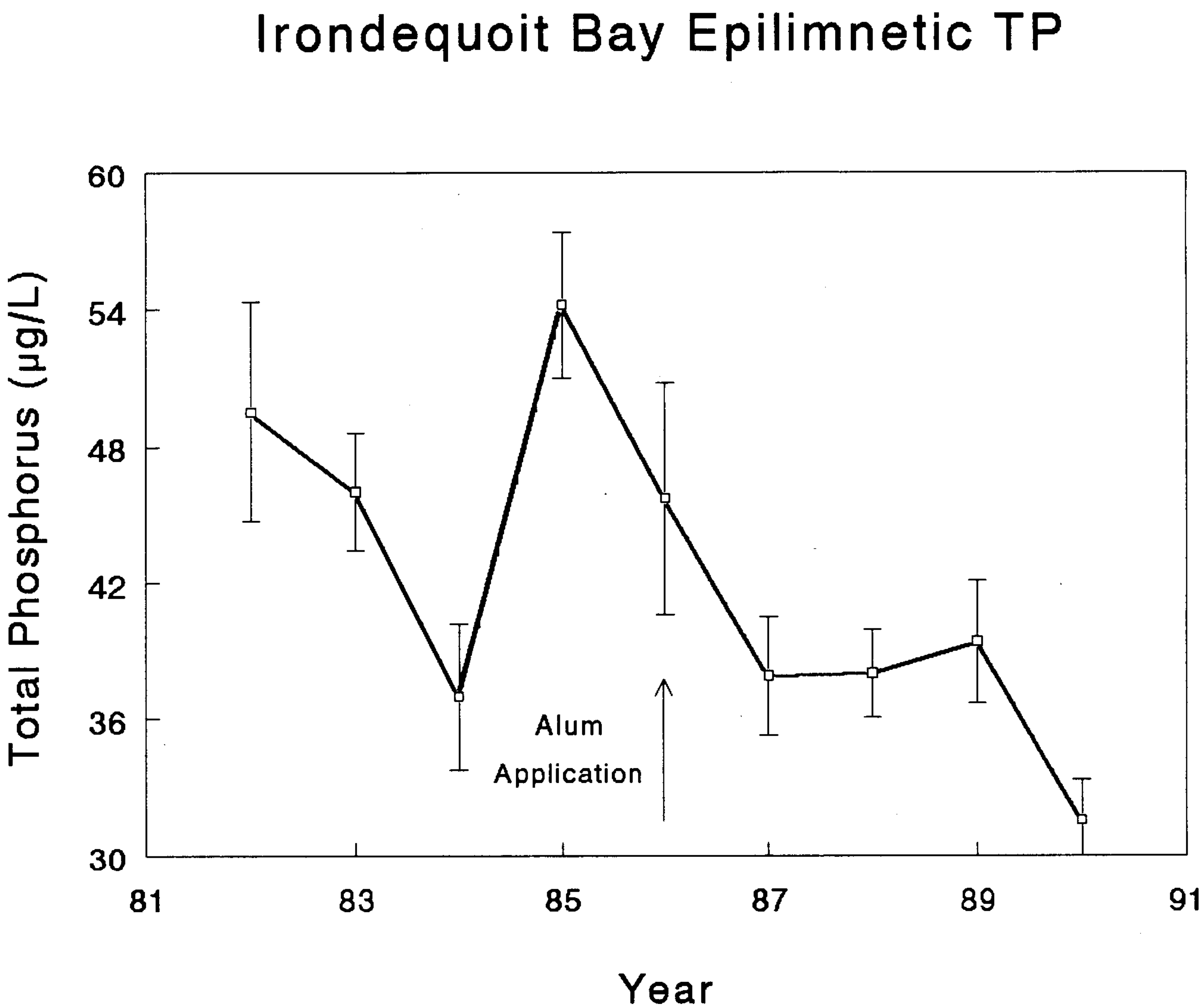

Figure 21. Changes in epilimnetic total phosphorus concentrations (surface) in Irondequoit Bay, NY following storm water diversion (1978-1986) and alum application

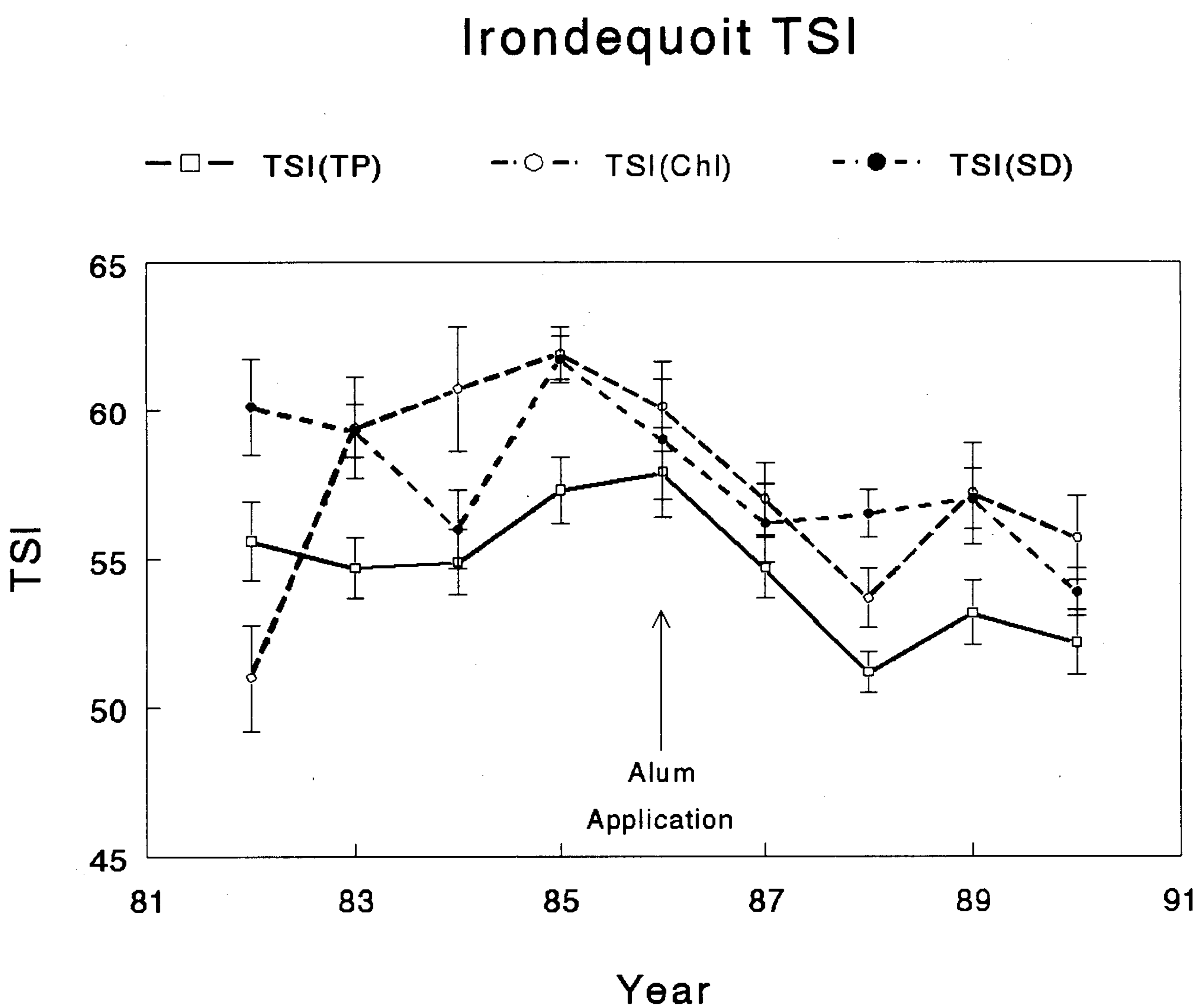

Figure 22. Changes in mean trophic state (TSI) variables of Irondequoit Bay, NY following storm water diversion (1978-1986) and alum application

Lake Morey, Vermont

Lake Morey had been impaired by decades of summer blue-green algal blooms, and a massive fish kill occurred in summer 1985. The lake was considered to be eutrophic. These problems were unexpected because of the lake's mountainous, heavily forested (92%) watershed. Initially, high nutrient loading from septic systems for homes, resorts, and summer camps was suspected. A detailed P budget in1981-82 revealed that septic tank drainfield leachate was insignificant and that internal loading greatly exceeded external loading. The lake's hypsograph is unusal in that two-thirds of the lake's area lies below the metalimnion. Lake Morey's hypolimnion is a zone characterized by low or zero dissolved oxygen and high P concentrations from sediment release. The extensive area of P-rich hypolimnion, therefore, should be associated with significant P diffusion and vertical entrainment to surface waters. It was concluded that internal P loading would have to be controlled in order to improve lake trophic state. A dose of alum and sodium aluminate of 44 g Al/m^2 (11.7 g Al/m^3) was applied to the hypolimnion in 1986.

Table 11 lists net P release rates, based on summer accumulation rates in the 11-13 m contour. Alum treatment reduced net sediment P release by an order of magnitude, an effect which has persisted at least 6 years (Figure 8; no data available for 1993). Total P concentration at 11 m (maximum depth = 13 m) declined from pre-treatment summer means above 200 μg/L to means ranging from 18-86 μg/L in the post-treatment years of 1986-93 (Figure 23). Mean summer surface TP fell from a pre-treatment mean of about 13 μg/L in 1981 to mean values ranging from 4-6 μg/L in 1989-93 (Figure 24). Algal biomass (chlorophyll) decreased from pre-treatment bloom levels (31 μg/L in 1985) to about 1 μg/L in 1990-93. Secchi disk transparency increased accordingly. Trophic state of Lake Morey improved from mesotrophic (TSI-TP about 45) before treatment to oligotrophic (TSI-TP about 30) following the treatment (Figure 25). The alum application's longevity of control of P release and its role in improving lake trophic state has lasted at least 7 years (Smeltzer, 1990; E. Smeltzer, Vermont Dept. Environ. Conserv., Waterbury, VT, personal communication).

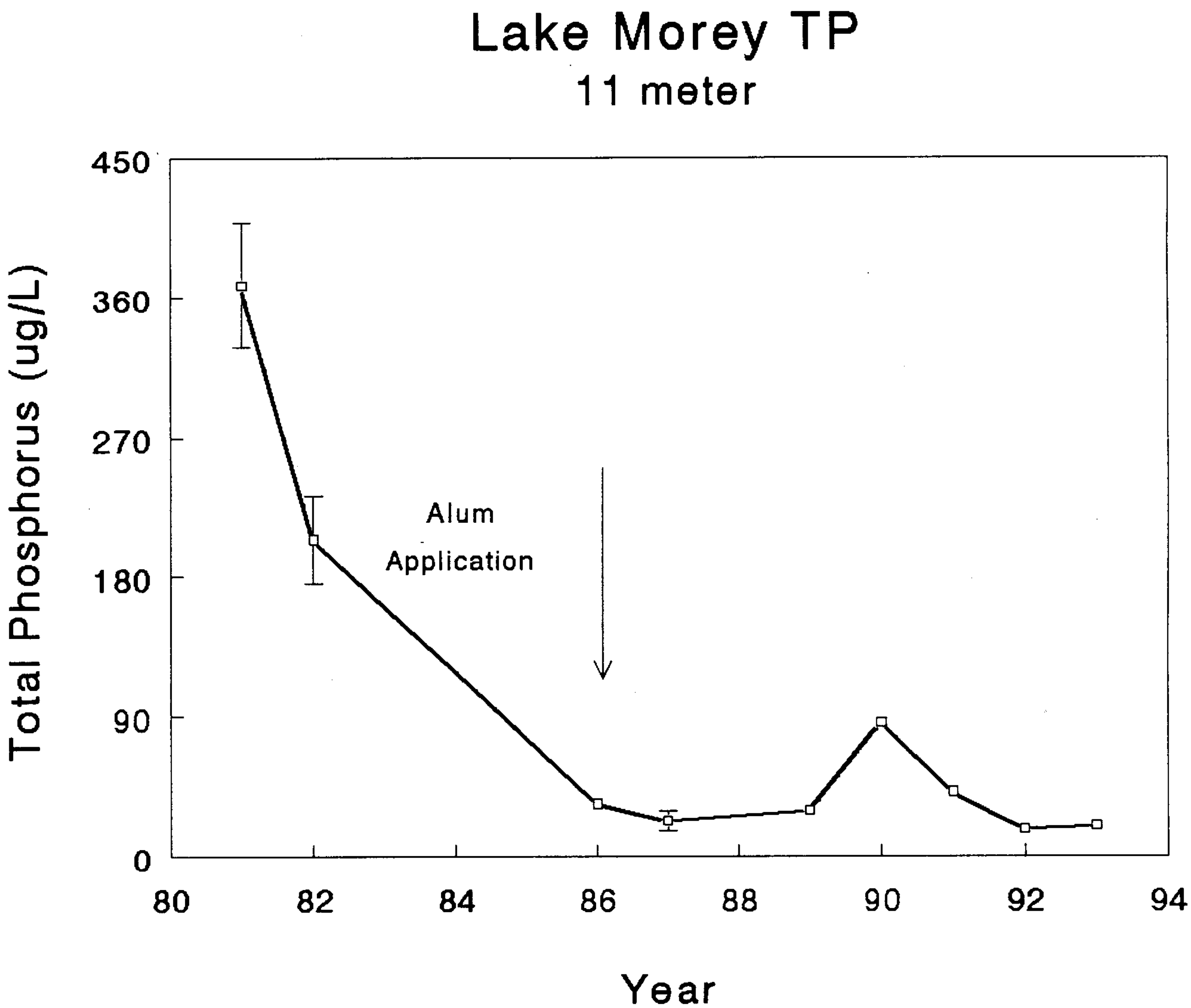

Figure 23. Changes in total phosphorus concentration at 11 meters in Lake Morey, VT following alum application

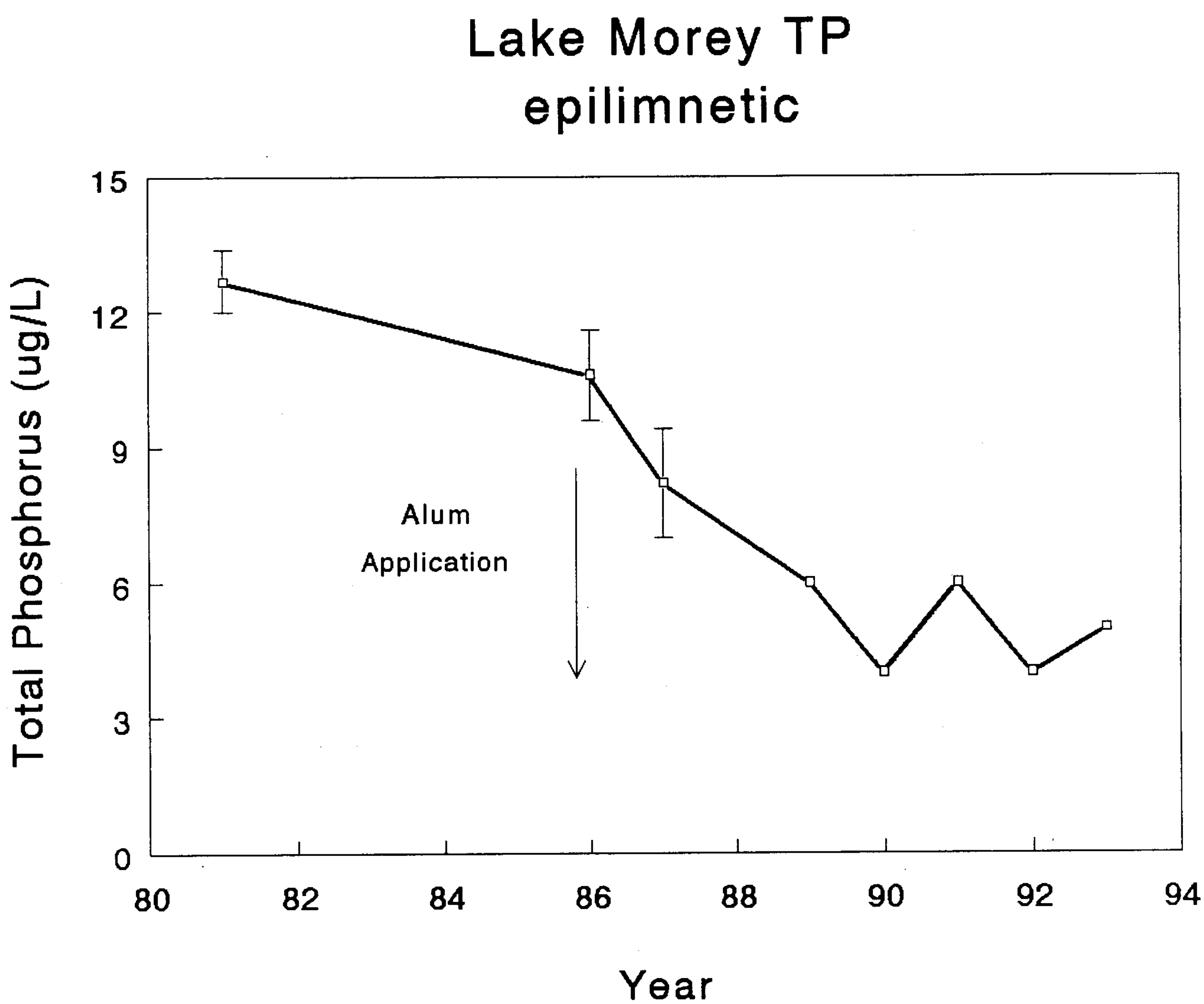

Figure 24. Changes in surface water total phosphorus concentration in Lake Morey, VT following alum application

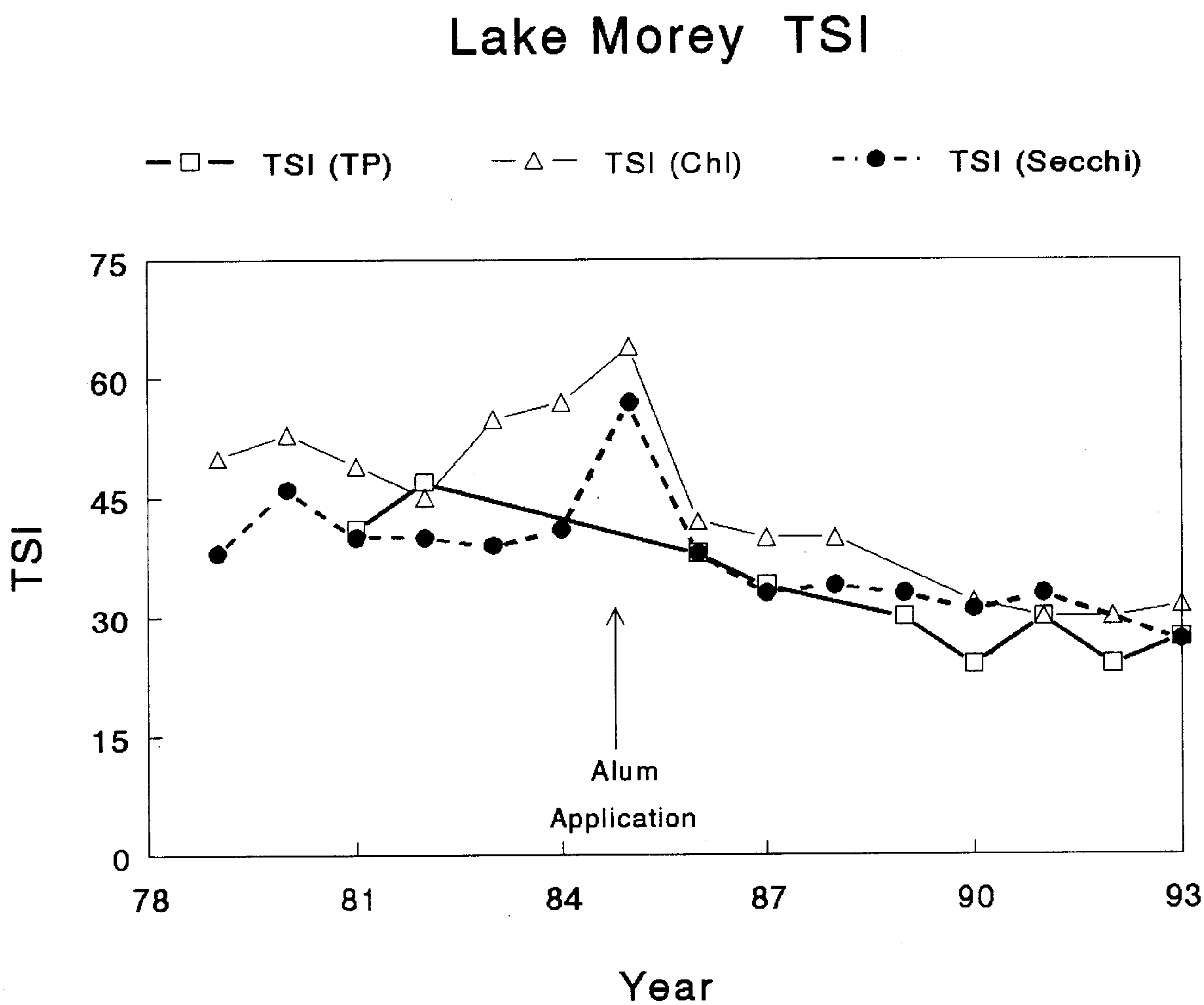

Figure 25. Changes in mean trophic state (TSI) variables of Lake Morey, VT following alum application

Kezar Lake, New Hampshire

Blue-green algal blooms in Kezar Lake began in the 1960s, following years of receiving wastewater effluents. Massive fish kills and property devaluations followed. Artificial circulation and copper sulfate treatments were ineffective. In 1981, some effluents were diverted, reducing the external P load by an estimated 71%. While no empirical measures of internal P loading were obtained, modeling and sediment core analyses suggested that sediment P release could be a significant P source to the water column. Even though the lake is dimictic, its shallowness suggests a high probability that partial mixing of P-rich hypolimnetic waters with surface waters could subsidize algal growth. Alum was applied to the hypolimnion in1984, following a copper sulfate application of 4.5 lbs/acre-10 feet (Connor and Martin, 1989).

Figures 26-27 illustrate changes in trophic state indicators in Kezar Lake following diversion and the copper and alum treatment. Mean TP concentration at 6 m (maximum depth = 8 m) declined sharply in 1984 when the lake was treated with alum, steadily returned to higher levels each year through 1987, and then declined again in 1988-1991 to levels near the 1984 post-alum treatment mean (Figure 26). Mean TP at 2 m declined from 1980 through 1992 (Figure 27). Diversion in 1981 produced a sharp decrease and so did the alum application in 1984. The post-treatment increase, then a decrease in mean TP at 6 m also occurred at 2 m, though less dramatically. This suggests that vertical entrainment in this dimictic lake was important, a conclusion supported by the lake's comparatively low Osgood (1988) Index of 3.14.

Changes in TSI values for Kezar Lake are shown in Figure 28. Mean chl (as TSI) declined after diversion and after alum application, and then increased and decreased as 2 m and 6 m mean TP changed. Mean transparency was the inverse of mean chl and, therefore, followed chl closely as TSI. Prior to diversion, Kezar Lake was eutrophic (TSIs > 50). Based on TP, diversion improved the lake to a mesotrophic state, although chl and transparency TSI values remained high. The alum application in 1984 further improved trophic state.

The effectiveness of the Kezar Lake alum treatment on trophic state cannot be separated from the effects of diversion. The increase in TP at 2 m and 6 m between 1985 and 1988, and the associated deterioration in trophic state, suggests a temporary increase in external P loading. Mass balance or hypolimnetic P release data are unavailable, but it appears that alum has been effective for 8 years in controlling sediment P release.

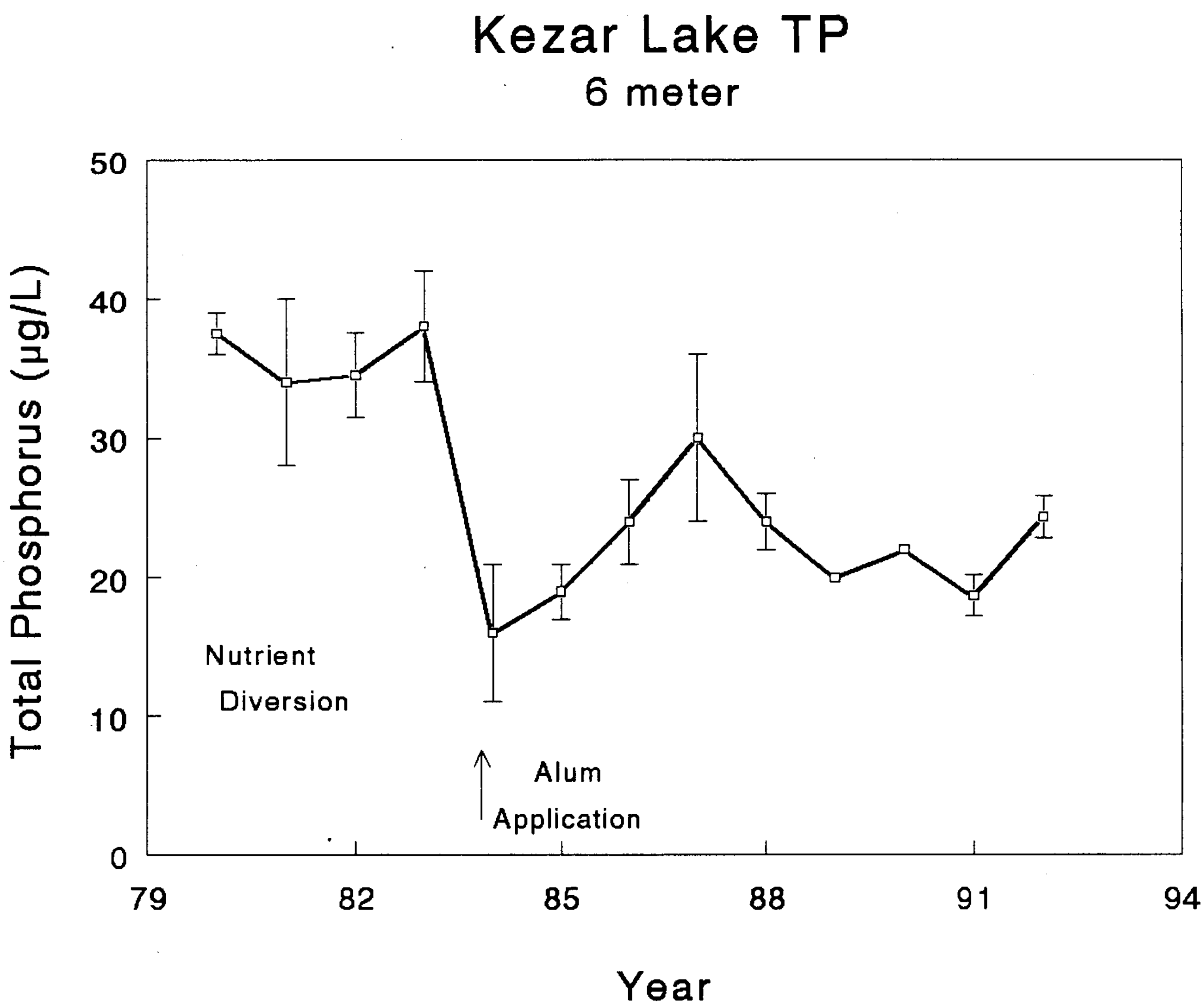

Figure 26. Changes in total phosphorus concentrations at 6 meters in Kezar Lake, NH following nutrient diversion and alum application

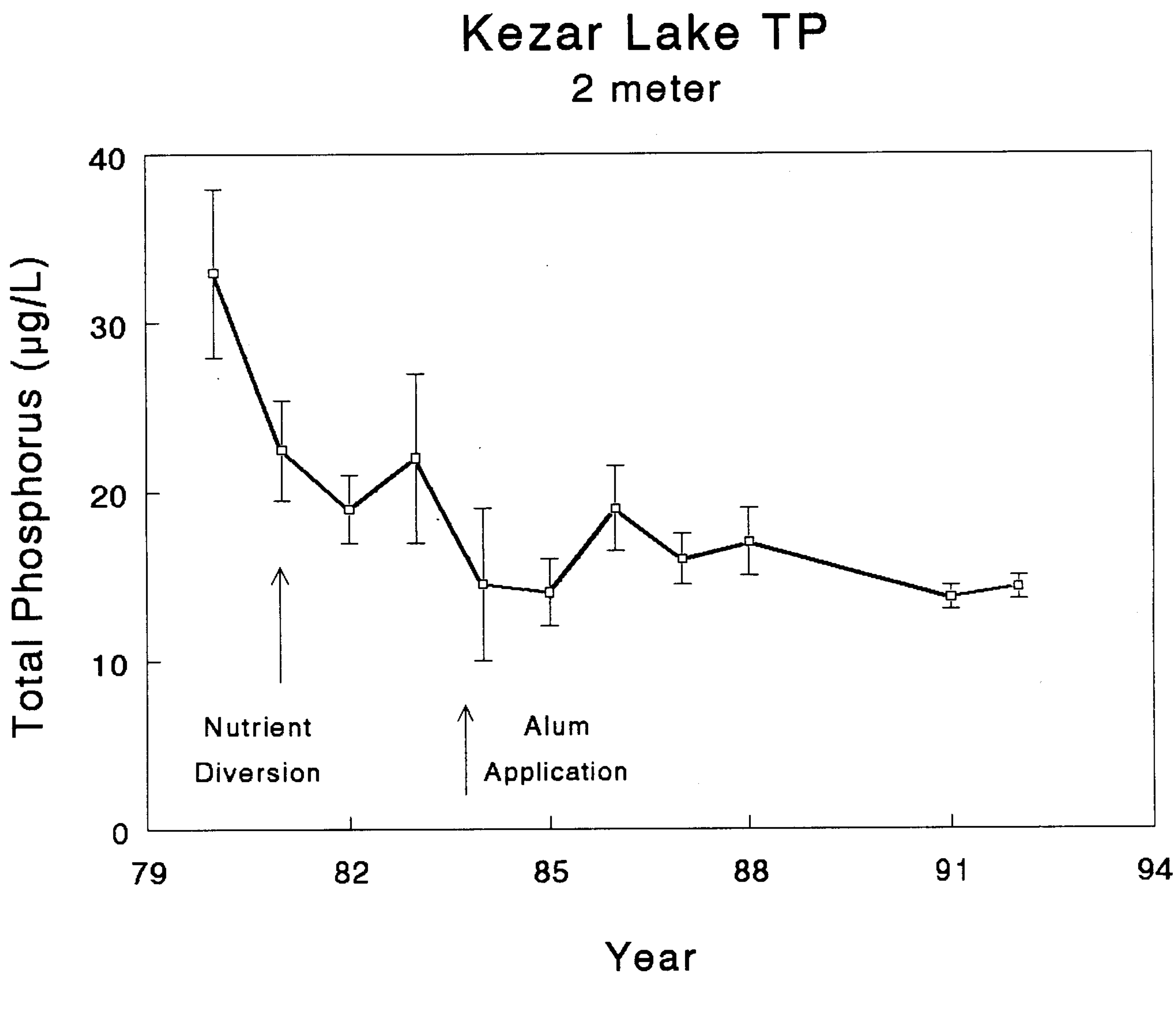

Figure 27. Changes in total phosphorus concentrations at 2 meters in Kezar Lake, NH following nutrient diversion and alum application

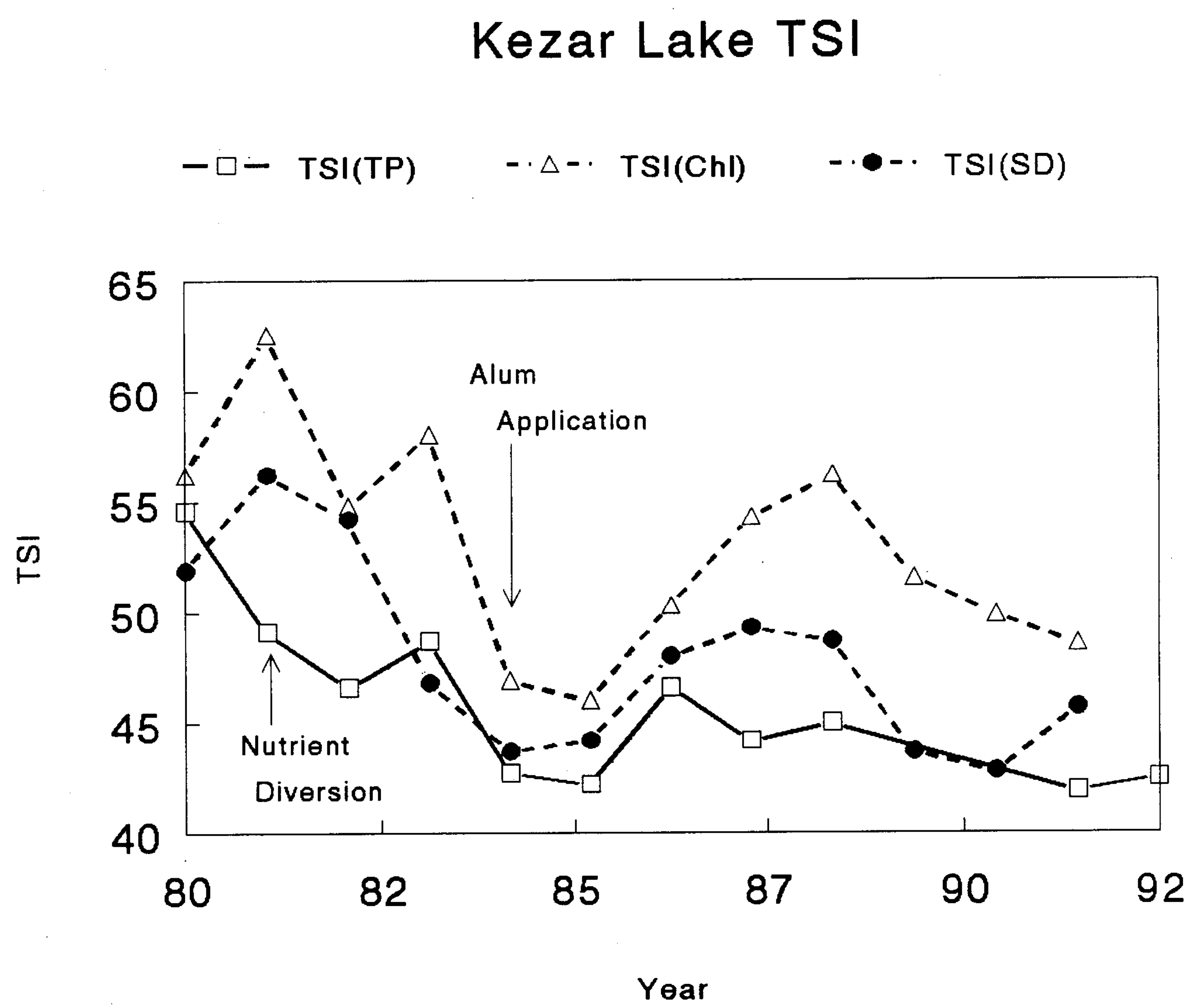

Figure 28. Changes in mean trophic state (TSI) variables of Kezar Lake, NH following nutrient diversion and alum application

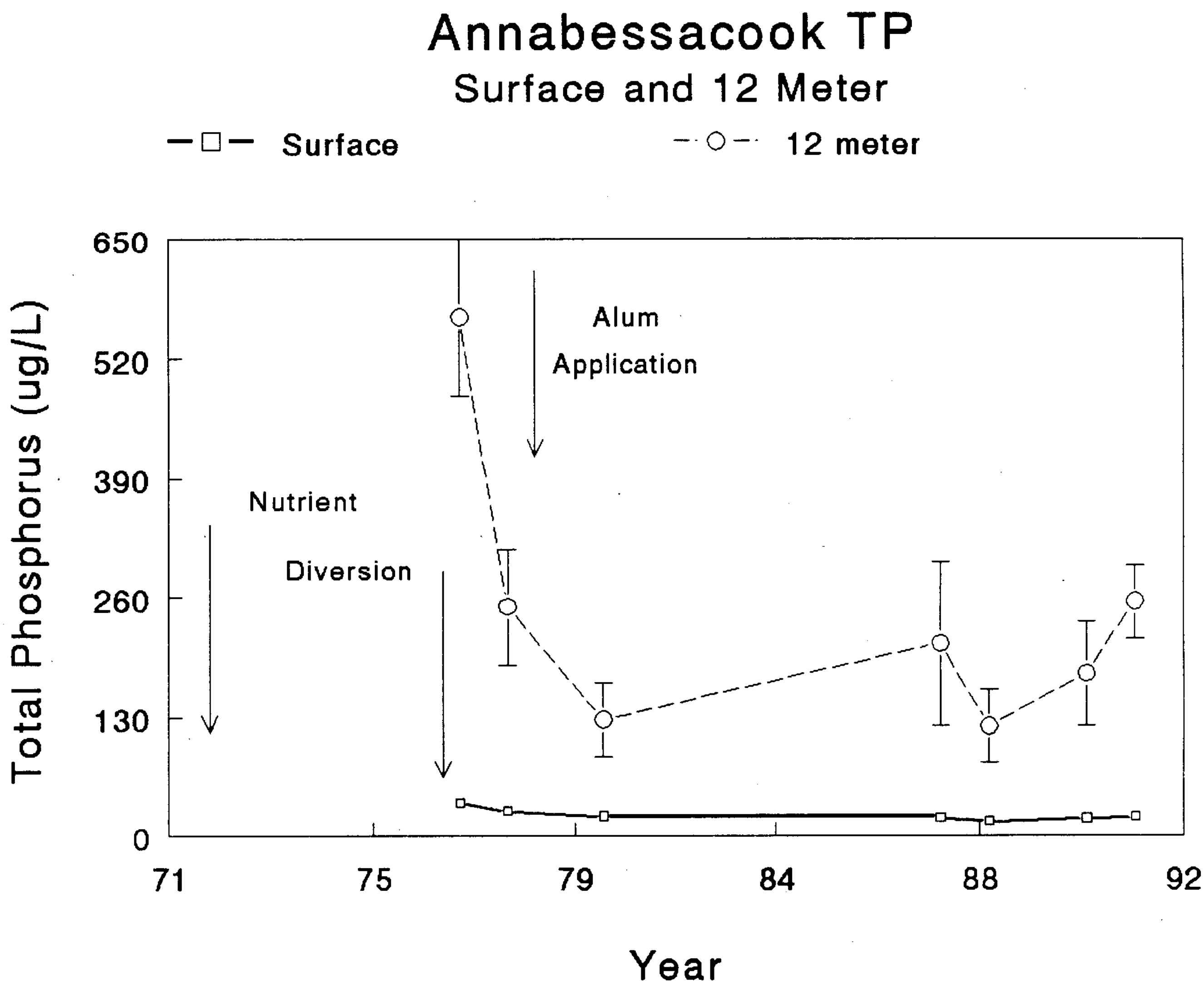

Figure 29. Changes in surface and 12 meter total phosphorus concentrations of Annabessacook Lake, ME following nutrient diversions and alum application

Annabessacook Lake, Maine

The lake received direct discharges of domestic and industrial wastes, and agricultural development in the watershed contributed nonpoint nutrients. Algal blooms have been common since the 1940s. In 1972, approximately 80% of the external P load was diverted, and in 1976 the remaining point sources were eliminated. Algal blooms persisted, and continued nonpoint agricultural loading and internal loading were identified as possible nutrient sources. Alum and sodium aluminate were applied to the hypolimnion in 1978 (Table 1), and agricultural waste management was implemented at the same time (Dominie, 1980; Gordon, 1980; Manfredonia, 1980). Since then, residential development on the watershed and along the shoreline accelerated and estimated external P loading in 1990 exceeded values from 1974 (Dennis, 1991, Cobbossee Watershed District, E. Winthrop, ME, personal communication). Areal internal loading rates during any of these periods are unavailable.

Figure 29 illustrates mean TP at surface and 12 m from 1975-1991 (from D. Dominie, personal communication; Dominie, 1980; Cobbossee Watershed District reports to USEPA; W. Dennis, personal communication; and Cobbossee Watershed District Water Quality Summary reports). The alum treatment lowered hypolimnetic TP from a 1977 pre-treatment mean ($n = 2$) at 12 m (deepest point) of 565 ± 85 µg/L to levels between 120 and 250 µg/L (Figure 29). The effect of the treatment on hypolimnetic P appears to have lasted through 1991 (13 years), assuming that the two pre-treatment data points in 1977 are indicative of concentrations prior to alum application.

The impact of the alum treatment on trophic state cannot be separated from the effects of nutrient diversion. Figure 30 shows an improvement in lake condition (based on surface TP and transparency) from eutrophic in 1977-78 (TSI > 50) to borderline mesotrophic (TSI = mid 40s). Algal blooms have persisted as evidenced by high chl TSI values. Mean transparency and chl (as TSI; Figure 30) in the 1990s did not differ from means in the mid-1970s. The close correspondence between TSI values for transparency and TP, with much higher values for chl, suggests that algal blooms are composed of large blue-green colonies. Sampling frequency and variances were not reported for transparency and chl.

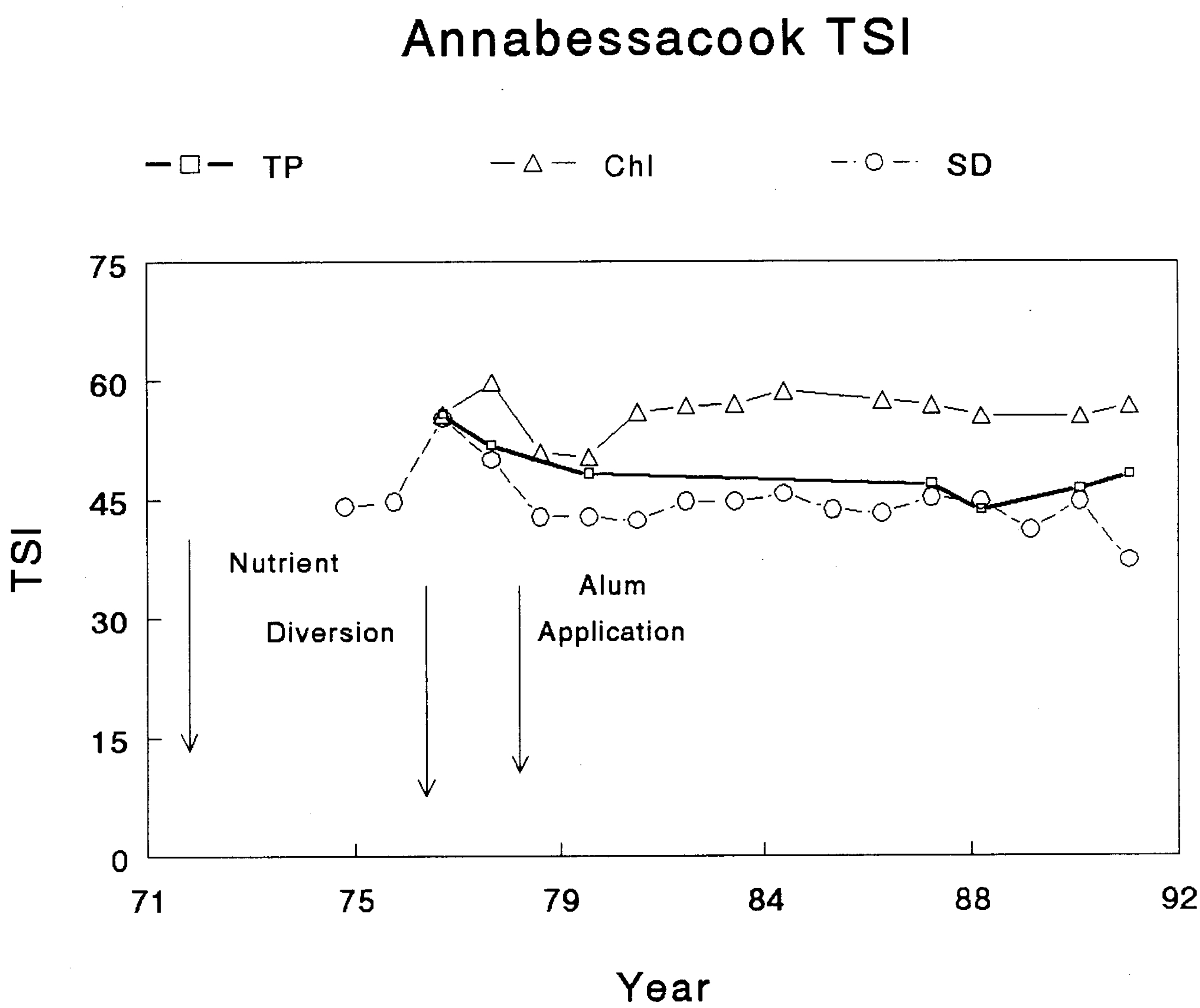

Figure 30. Changes in mean trophic state (TSI) variables in Annabessacook Lake following nutrient diversions and alum application

The most important force in improvement of Annabessacook Lake may have been the diversion of municipal and agricultural loading, rather than alum treatment. However, the lake is dimictic with an Osgood Index of 2.25, a value which indicates a high probability of vertical transport. The lowest surface TP values were found in June, prior to the modest post-alum hypolimnetic P increases of late summer. The highest epilimnetic values were always in late summer. This suggests that vertical transport increased epilimnetic TP when the hypolimnion became P-enriched. The alum treatment may have reduced the significance of this late summer P source, though pre-treatment data are missing. Based on this reasoning, the alum application to Annabessacook Lake probably contributed to an improved trophic state, and its effects have lasted for at least 13 years.

Cochnewagon Lake, Maine

Cochnewagon Lake, also found in the Cobbossee Watershed District with Annabessacook Lake, began having significant blue-green algal blooms in 1980. Prior to this time, the lake was mesotrophic. Significant reductions in agricultural and residential sources of nutrients took place, beginning in 1979-80. Internal P loading into the lake's anoxic hypolimnion was believed to be a major P source. For example, in summer 1984, the input of P to the water column from this source was estimated to be 1.5 times external loading. In Cochnewagon Lake sediment P release could be a significant contributor to surface algal blooms because the lake occasionally destratifies, which would allow P-rich hypolimnetic waters to mix with the entire water column. Alum and sodium aluminate was added to the hypolimnion (Table 1) in 1986 to control this P source (Dennis and Gordon, 1991).

Mean algal biomass (chl) declined sharply and transparency increased after diversion (Figure 32). Mean TP at 8 m (maximum depth = 9 m) was very high in 1982, possibly due to prolonged stratification and anoxia (Figure 31). Though a somewhat elevated mean TP at 8 m was also noted in 1985, the general pattern was a decrease following diversion. Alum application in 1986 eliminated the elevated TP at 8 m, even during sustained anoxia in 1989, and mean concentrations at 8 m became nearly identical with mean surface concentrations. Surface (1 m) mean TP concentrations ranged from 10.0 ± 2.0 to 16.0 ± 1.7 µg/L between 1980 and 1991 (before and after the 1986 alum treatment). Variance in these data, where sample sizes ranged

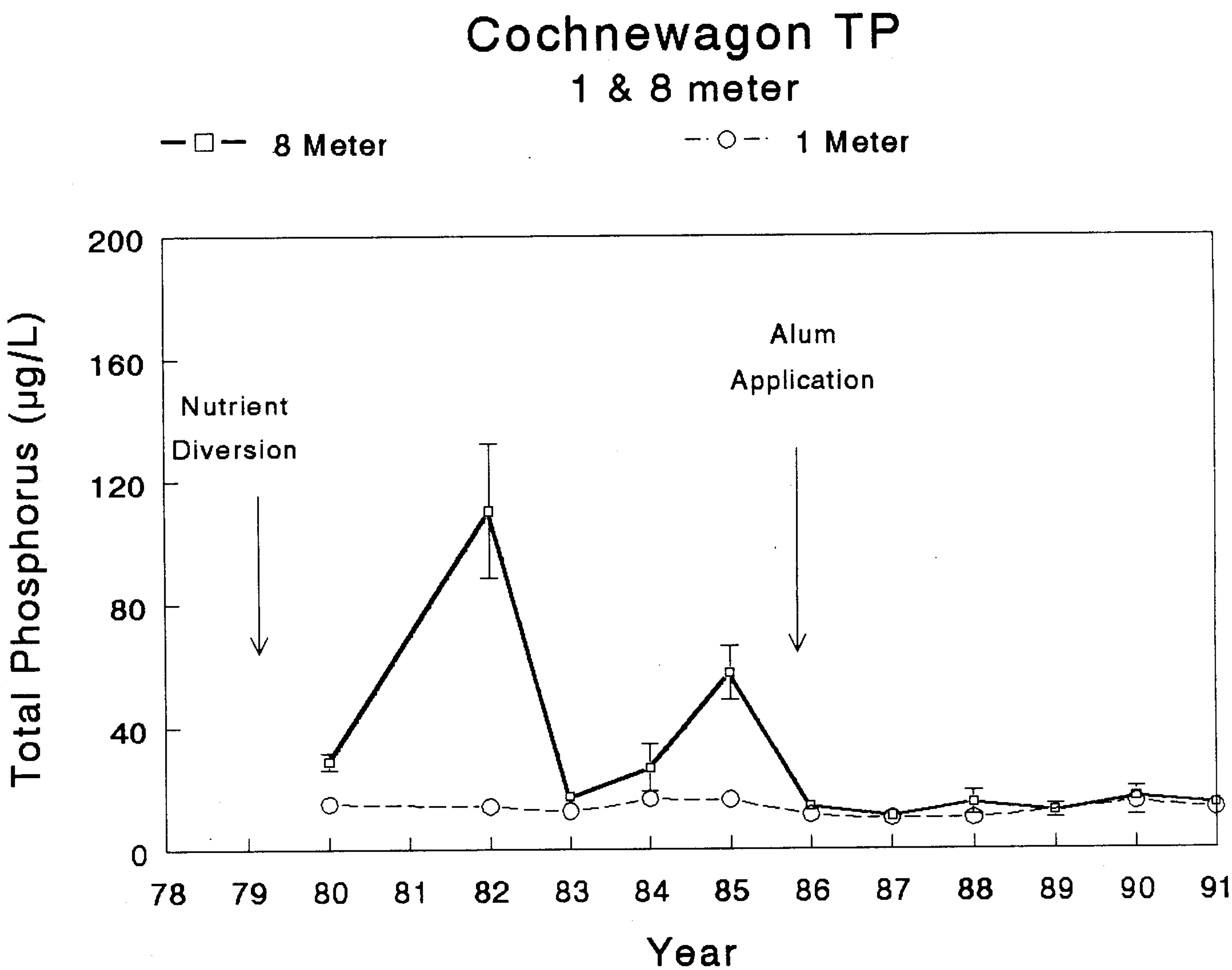

Figure 31. Changes in surface and 8 meter total phosphorus concentrations in Cochnewagon Lake, ME following nutrient diversion and alum application

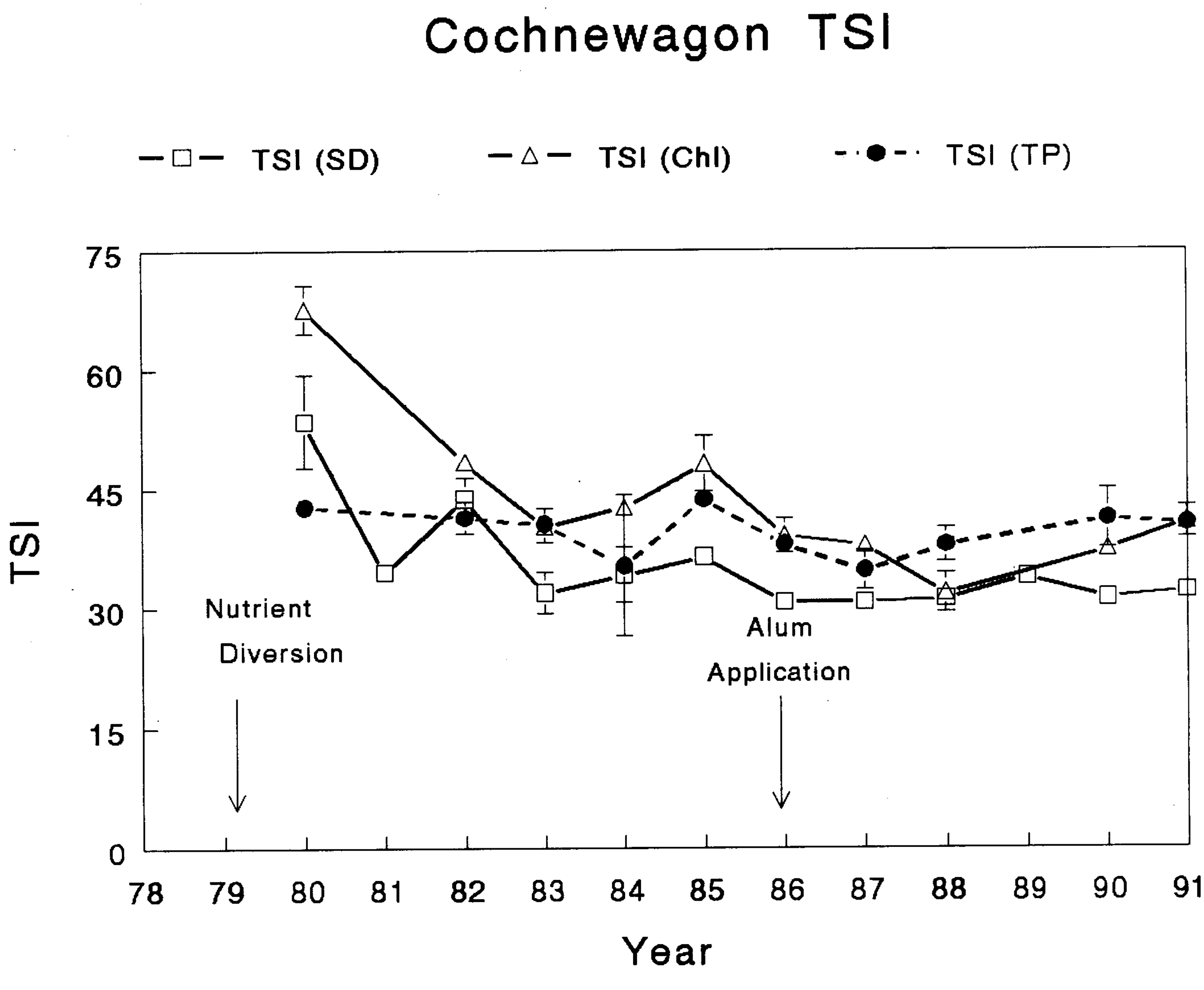

Figure 32. Changes in mean trophic state (TSI) variables in Cochnewagon Lake, ME following nutrient diversion and alum application

from 4-6 before alum treatment to 11-12 per summer after treatment, was extremely small. The alum application does not appear to have lowered surface TP concentration, suggesting that vertical P transport prior to treatment was not a significant P source to surface waters (Dennis and Gordon, 1991; W. Dennis, personal communication).

Figure 32 illustrates changes in lake trophic state following diversion and alum application. Chl (as TSI) declined sharply after diversion and transparency increased. The lake was eutrophic in 1980 based on chl, though TP concentrations at surface and 8 m were low. Trophic state steadily improved after diversion, with a strong correlation among variables appearing in 1982, suggesting P limitation of algal biomass. The lake reached oligotrophic status in 1983, prior to alum application, and has varied since then around a trophic state of 40. Slight increases in 1990-91 suggest increased external loading. The hypolimnetic alum treatment in 1986, while apparently effective in controlling P release from anoxic hypolimnetic sediments, does not appear to have been a major factor in improving lake trophic state when compared to diversion of external loading. Effectiveness of alum in controlling sediment P release has lasted at least five years.

FATE OF ADDED ALUMINUM--ALL LAKES

The added Al was not readily visible as a distinguishable layer in most sediment cores. Only in Mirror and possibly Campbell Lakes did the analytical results indicate a point in the core with higher Al content. That is surprising, because a distinct grayish band of alum floc was obvious at a depth of about 5 to 10 cm in the Mirror, Shadow, and West Twin Lake cores. Possibly the 2-cm increment samples were too crude to effectively detect that narrow band.

Nevertheless, enough Al was added to have been detectable in the cores. While the digestion procedure does not release Al from silicates, other forms of Al and Fe, such as the $Al(OH)_3$ floc, should have been solubilized. Based on the quantity of Al added and background levels of Al in the sediment, residuals from the alum dose should have been detectable. For example, had the added Al remained in the upper 2 cm, Al in the sediment should have been increased to over 100% of background in the majority of cases. So the Al added with most alum treatments represents a substantial fraction of the background sediment content.

The added Al may have been mixed vertically over time by bioturbation and physical settling through sediment due to density difference and possibly due to wind in shallow lakes. That could account for the lack of obvious concentration peaks in most cases. There is only one example where a control lake will allow testing of that hypothesis in this data set and that is West and East Twin Lakes (OH). The addition of 26 mg/L of Al to West Twin with a mean depth of 4.3 m should have raised the Al concentration in the 20-cm sediment column (core length) an average of 12.7 mg/g (assuming 4.4% solids), or 62% over the background mean concentration of 20.5 $\pm$ 3.0 mg/g. Figure 33 shows that Al levels throughout the 20-cm cores are consistently higher in West Twin, the treated lake, than in East Twin, the control. That average difference, however, is only about half (6.7 mg/g) the expected difference. The high concentrations of Al at 16-20 cm in the West Twin core suggests that the added Al was distributed even deeper than 20 cm (Figure 33).

Surficial sediment (0-4 cm) collected in Delavan Lake, WI, in 1991 contained 10.6 mg/g of Al, almost twice the concentrations at 4-8 and 8-12 cm of 6 and 6.7 mg/g, respectively. Delavan Lake was treated only two months before the core was collected and at the time of sampling the alum floc was highly visible on the sediment surface. These observations support a downward redistribution of a floc layer with time.

Except for the low background Al in Delavan Lake sediments, sediment Al ranged from about 10 to 30 mg/g in the test lakes. Fe ranged in concentration from about 7 to 30 mg/g and P from about 0.5 to 2.5 mg/g in most lakes. Table 12 shows ratios of Al/P, Fe/P and Al/Fe in the surficial sediments of the lakes. Al/Fe ratios in Erie, Campbell, Long-north and Long-Kitsap were considerably greater than the ratios in Long-south and the two basins of Pattison Lake. Moreover, sediment TP was correlated more closely with Al in Erie and Campbell Lakes ($r^2 = 0.95, 0.98$), while TP was correlated more closely with Fe in Long-south and Pattison-north ($r^2 = 0.98, 0.99$). That may partly explain the different treatment effectiveness/longevity among those lakes. Al/Fe ratios are 1.0 or more in the three Wisconsin lakes with effective long-lasting treatments (Mirror, Shadow and Horseshoe). The high pH from macrophytes in Wapato Lake may have been the most important cause for the ineffective alum treatment there, in spite of the rather high Al/Fe ratio. In contrast, treatment effectiveness/longevity was five or more years in West Twin Lake, while the Al/Fe ratio was rather low, although it was considerably increased over that in East Twin Lake.

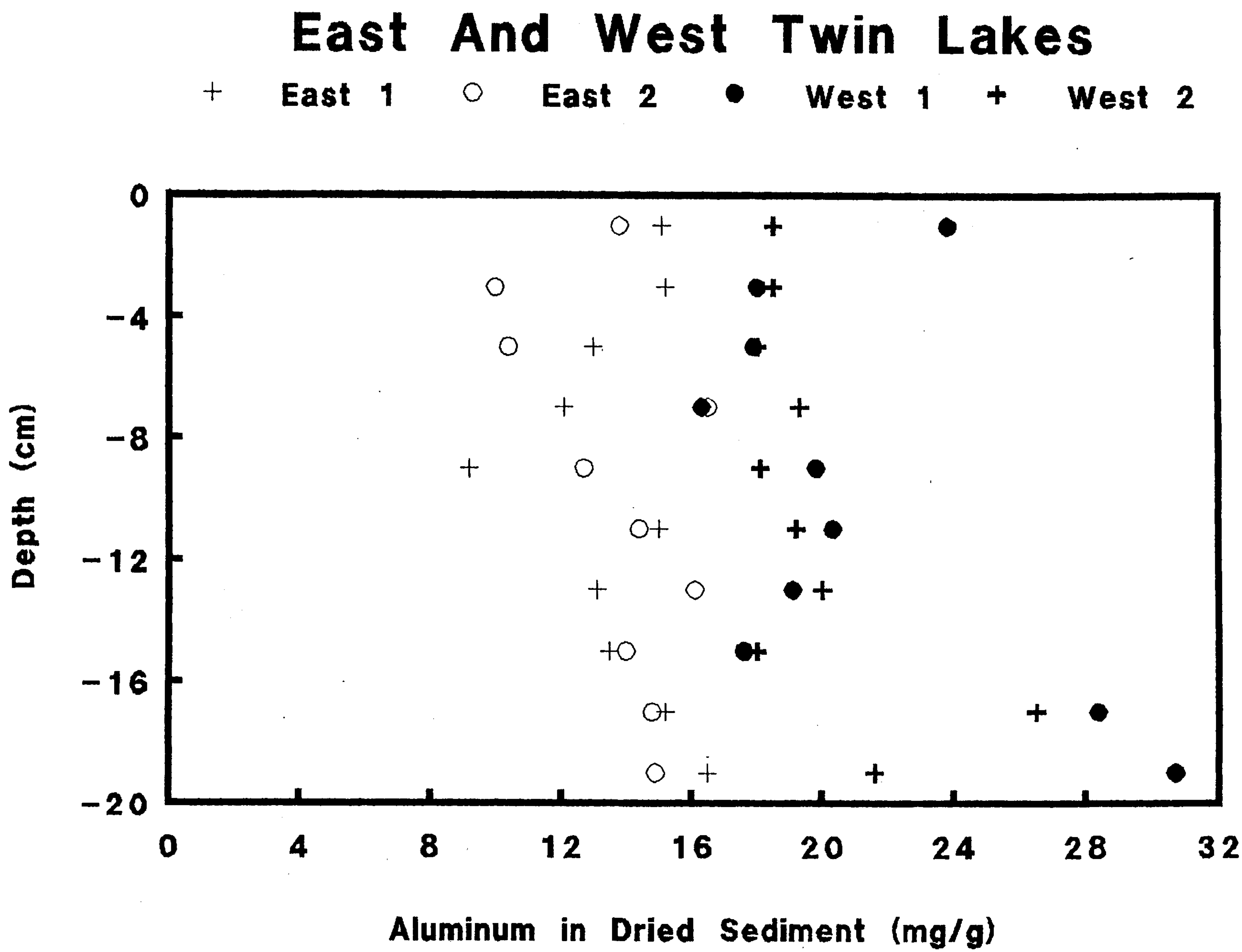

Figure 33. Distribution of aluminum in sediment cores of East and West Twin Lakes, OH. West Twin Lake was treated with aluminum sulfate.

TABLE 12. TP CONTENT AND RATIOS AMONG P, AL AND FE IN SURFICIAL (0-6, 0-8 CM) SEDIMENT (N=2 or 1*) OF STUDY LAKES

	TP	Al/P	Fe/P	Al/Fe
Campbell	2.3	7.4	6.8	1.09
Erie	1.6	8.0	7.7	1.04
Long (north)	1.5	7.5	8.1	0.93
Long (south)	1.6	6.5	8.7	0.75
Pattison (north)	2.3	5.2	10.0	0.52
Pattison (south)	1.9	4.8	7.2	0.67
Long (Kitsap-pretreat)	1.8	21.6	16.3	1.33
Wapato	2.6	12.2	11.2	1.09
Mirror	2.1	6.0	6.2	0.97
Shadow	2.5	5.8	3.1	1.87
Horseshoe	2.6	4.7	2.6	1.81
Eau Galle	4.1	4.8	4.8	1.00
West Twin	1.7	12.3	17.4	0.71
East Twin (untreated)	1.4	10.2	20.8	0.49
Dollar	2.4	17.5	18.3	0.95
Kezar*	1.3	15.7	10.2	1.54
Annabessacook*	2.8	6.8	11.3	0.60
Morey*	0.6	27.5	44.0	0.63
Delevan*	0.8	10.0	12.1	0.82
Cochnewagon*	1.0	22.6	24.1	0.94
Irondequoit*	1.0	14.4	18.4	0.78

SEDIMENT-P PROFILES AND CORE RELEASE RATES

Sediment-P content was determined in cores in an attempt to detect if changes in external loading (changed P deposition) had occurred since alum treatment. However, the core data proved to be inadequate to answer that question. Besides the problem of substantial variation between profiles in replicate cores, P content usually increased near the sediment surface, even in lakes where no change in human development or use had occurred. While such an increase could conceivably be due in part to increased external loading in recent years, such concentration toward the surface is more likely the result of migration of redox solubilized P toward the sediment surface where it is retained through cycles of oxia/anoxia.

Release rates of P determined in cores sectioned at different depths (intact, 2 cm removed and 6 cm removed) and incubated under anaerobic conditions, did not provide useful information for evaluating alum treatment effectiveness. While release rates from intact cores were usually higher than rates from cores from which 2 cm of sediment had been removed, a similar pattern was shown in cores from an untreated lake. The depth stratification of release rates is apparently natural and may be a result of depth-varying P fractions (Bostrom and Petterson, 1981).

DISCUSSION

EFFECTIVENESS/LONGEVITY IN UNSTRATIFIED LAKES

These results clearly show that P inactivation with alum in shallow lakes is effective and has longevity. Effectiveness of inactivation (P release) averaged 48% for the 6 successful cases. Further, treatment effectiveness of P inactivation lasted an average of at least 8 years for those 6 cases. Treatments in 4 of the 6 cases still remained effective, so their longevity is probably greater than 8 years.

Trophic state improved initially in all shallow lakes where TP was effectively controlled. However, treatment longevity was not as apparent with TSI in some cases as with only TP. TSI remained lower than pre-treatment levels for at least eight years in four of the six successful treatments. Moreover, the blue-green algal component was reduced in the three lakes where pre-treatment data were available, and in two lakes *Aphanizomenon* and its scum forming blooms have been virtually absent for the eight years since treatment.

Of the lakes investigated, the least effective treatment was in Pattison Lake-south, which was very shallow and completely covered with macrophytes. How macrophytes reduce the effectiveness of alum is not entirely clear. They apparently intercept the settling alum floc and prevent a uniform covering on the sediment surface. That is supported by lower Al content of surficial sediment in Pattison-south and Long-south than in either north basins. Also, for species that senesce during summer and release P to the water through decomposition (e.g., *Myriophyllum spicatum*; Smith and Adams, 1986), internal loading of P would continue because their roots penetrate well below the sediment surface and alum layer. That could explain the poor effectiveness in Pattison-south. Macrophyte coverage in Long-south was not as dense, but Pattison-south drained to Long-south, so much of the P released from senescent plants in Pattison-south would have been transported to Long-south, possibly accounting for the shorter longevity in Long-south.

The other problems presented by macrophytes in shallow, alum-treated lakes is due to increased transparency. Macrophytes are apt to be limited by light in eutrophic lakes with dense algal blooms. The resulting improved water clarity in shallow alum-treated lakes, with Secchi Disk exceeding maximum depth in many cases, will stimulate plant growth and could result in greater internal P loading even if macrophytes senescence were not important initially. Macrophyte biomass (largely *Elodea canadensis*) increased greatly following the alum treatment in Wapato Lake, from 90 and 37 g/m^2 and minimal surface coverage determined the two years before treatment to 129-130 g/m^2 and 70-90% coverage for the two post-treatment years (Entranco, 1986). The plants were estimated to have contributed 57 and 60 percent of the total loading of P the two years following treatment, based on senescence only, whereas contributions from the previous two years were estimated at only 7 percent (Entranco, 1986).

Macrophytes may not always increase and contribute significantly to internal loading, however. The dominant macrophyte in Long Lake (Kitsap) is *Egeria densa*, which did not increase following the 1980 alum treatment. Because it is very slow to senesce, it does not contribute significantly to internal P loading in that lake (Welch et al., 1993). Moreover, it apparently protects the sediment surface from wind mixing, thereby retarding entrainment of P, which is an important mechanism of internal loading in shallow lakes (Jones and

Welch, 1990). That was especially noteworthy in 1985 when *Egeria* biomass was only 10 percent of previous levels, leaving large shallow areas with a bare sediment surface, and the highest observed post-treatment whole-lake TP.

The mechanism(s) of P release, from sediments in shallow, unstratified lakes that are effectively controlled by alum, is not clear. All these test lakes are softwater and very sensitive to pH increase from algal photosynthesis. Aluminum hydroxide complexed P is released when pH increases, in contrast to this molecule's insensitivity to anoxia. That mechanism (hydroxide exchange for PO_4 on iron hydroxide complexes at high pH) was probably responsible for some of the pre-treatment internal loading in Long Lake-Kitsap (Jacoby, et al., 1982). High pH (9-10) was observed throughout the water column in 1976-1978 prior to alum treatment. However, high pH did not occur following the alum treatment. On the other hand, high pH (10) in Wapato Lake was blamed for the failure of alum in that lake. The high pH was apparently due to a large biomass of *Ceratophyllum* and *Elodea canadensis* (see earlier data). *Ceratophyllum* is non-rooted and tends to cover the bottom, so its photosynthesis may have produced high pH in sediment overlying water. *Ceratophyllum* was also abundant over the bottom of broad areas of Long Lake (Kitsap) in 1985 after *Egeria* had declined to a low level and TP reached pretreatment levels. High pH may have occurred in sediment overlying water and been partially responsible for the TP increase in that case as well. Thus, alum may not continue to inactivate P if pH at the sediment-water interface increases from a post-treatment increase in macrophytes.

In addition to high pH, brief periods of anoxia in water immediately overlying the sediment, when winds are calm, are often considered a more likely explanation as the principal cause of internal loading in unstratified lakes. Low DO and high SRP have been observed in sediment overlying water in Long lake-Kitsap (Welch et al., 1988). High pH may have had a secondary role, however. Once P is entrained from a narrow (probably undetectable) anoxic layer overlying the sediments, a high pH in the water column due to algal photosynthesis (resulting from the released P) would maintain solubility and continuous availability of P (Ryding, 1985), as well as further release from sediment. The effectiveness of internal P loading control by alum in most of these shallow lakes indicates that iron redox is probably an important mechanism that initiates P release because aluminum hydroxide-P complexes are not redox sensitive. P translocation via blue-green algal recruitment from sediment is apparently not affected by alum floc (Perakis et al., 1995). If the iron redox mechanism is blocked by aluminum hydroxide, then algal-photosynthetically-caused high pH will not occur (assuming macrophytes are not a problem) and sediment P will remain inactivated.

Migration of blue-green algae from sediment to water in shallow lakes can also represent a significant portion of the summer increase in TP (Barbiero and Welch, 1992). The sharp curtailment of blue-green algae in lakes with effective alum treatments may indicate their previous importance to internal loading (e.g., Erie, Campbell and Long-Kitsap Lakes). The near complete absence of *Aphanizomenon* for eight years in Erie and Campbell Lakes is noteworthy. Such a sharp reduction in blue-green algae has usually not occurred in lakes where lake TP was reduced externally. The reduction in the fraction of *Oscillatoria* and other blue-greens in Lake Washington was greatly delayed beyond the decrease in TP and algal biomass (Edmondson and Litt, 1982). The blue-green fraction in Moses Lake, Washington remained relatively unchanged for 12 years after the start of dilution and subsequent sewage diversion had caused the reduction of TP by 50 percent and chl a by 70 percent (Welch et al., 1992). While Lake Washington is deep and sediment derived inoculation of the water column may not be important, sediment-water migration of *Aphanizomenon* in shallow Moses Lake is pronounced.

Al in the sediment of alum-treated lakes was usually indistinguishable above background either chemically or visually, even though the amounts added should have at least doubled the concentration had it remained in the top 2-3 cm. Therefore, the alum floc probably settles gradually through the usually low-density sediments of most lakes. For example in West Twin Lake, OH calculated versus observed increase compared to the control lake indicated that the floc apparently redistributed rather evenly to more than 20 cm between 1975 and 1991. The floc should be easily distinguishable near the surface, both chemically and visually, before it has had a chance to settle, as was the case with Delevan Lake sediments 2 months after treatment. On the other hand, the floc layer was quite visible between about 5 and 10 cm in Shadow Lake sediments 13 years after treatment, but was not detected chemically. Also, the floc layers in Green Lake and Long Lake (Kitsap, Washington) were indistinguishable visually after only two weeks following treatment (Leinenbach, 1993). In general, however, an alum floc tends to distribute downward in the low-density sediment of lakes and become indistinguishable, both chemically and physically, from background levels over time.

There is some indication that treatment effectiveness may be related to the ultimate Al/Fe ratio. For the Washington and Wisconsin lakes, effectiveness/longevity were greater where sediment Al/Fe ratios were greater than 1.0, which may have merit in judging the alum dose in lakes where P is controlled by Fe.

Al content in lake water (188 $\pm$46 µg/L for WA lakes) was relatively high regardless of the time since treatment (Schrieve, 1992). Moreover, nearly all the Al was soluble (filterable). However, these high levels are not considered to be an adverse effect of alum treatments. That is because in one instance TAl and SAl actually decreased after treatment (Long-Kitsap) and another where SAl concentrations six years after treatment were much higher than the first year after treatment (Erie and Campbell). Also, a clear Al layer in the sediment was either indistinguishable or buried in most lakes with sediment of high water content, so that the high levels are more likely due to natural Al sources than the alum addition.

EFFECTIVENESS/LONGEVITY IN STRATIFIED LAKES

These long term monitoring data from alum treated lakes in Wisconsin, Ohio, and the eastern states clearly emphasize the major role of nutrient diversion in bringing about lake trophic state improvement. In the extreme case, Eau Galle Reservoir, WI was alum treated without any attempt to reduce external loading. Sediment P release was briefly reduced, as expected, but algal blooms persisted because loading was unabated. Data from Kezar Lake, NH and Irondequoit Bay, NY suggest that the effectiveness of their alum treatments was affected by continued high loading (in the case of Irondequoit Bay), or by episodes of loading (Kezar Lake) after treatment. In at least one lake (Cochnewagon Lake, ME), external loading appeared to be the only major nutrient source so that diversion alone, not alum, was responsible for most of its improvement.

Aluminum sulfate, or aluminum sulfate plus sodium aluminate, was found to be extremely effective and long lasting in reducing sediment P release. Evidence from in situ release chambers (Mirror and Shadow Lakes, WI) and from hypolimnetic P accumulation rates (Horseshoe, Snake, East and West Twin, Irondequoit Bay and Lake Morey) shows that control has lasted at least up to 12 years (Mirror and Shadow) and probably as long as 20 years (Horseshoe and Snake). Evidence of effectiveness and longevity from reduced hypolimnetic P concentrations suggests that treatments may have controlled P release for periods of 13 years in

Annabessacook and perhaps up to 18 years in Dollar. The West Twin Lake treatment appears to have diminished effectiveness, a change which occurred somewhere between 6 and 14 years after application.

A more significant assessment of alum treatment effectiveness involves a determination of its impact on lake trophic state. Unlike the Washington lakes, summer external loading to the lakes of Wisconsin, Ohio and eastern states can be significant. However none of these project lakes has had a water and P input-output budget done each year. This would have allowed a separation of the effects of P diversion on lake concentration from the effects of the alum treatment.

In stratified lakes, it has been conventional wisdom to conclude that the P-rich hypolimnion, the target of alum treatments, provides a significant internal load of P to the epilimnion via diffusion and/or by vertical transport during wind events. In six of the stratified lakes (Eau Galle, Irondequoit Bay, Morey, Kezar, Annabessacook, and Cochnewagon) the Osgood Index $\left(\bar{z}/\sqrt{A_0}\right)$ values (Osgood, 1988) were low enough (< 6) to expect vertical P transport and thus reduced epilimnion P when alum was applied. The Eau Galle treatment failed due to continued external loading. Of the remaining lakes, only Cochnewagon did not have a decrease in epilimnetic P which could be inferred to have been associated with the alum treatment. Sediment P release at Lake Cochnewagon was not shown to be a significant P source to the water column prior to treatment.

Six of the stratified project lakes had Osgood Index values below 6, and four had index values above 13. These latter lakes would be predicted to have little or no reduction of epilimnetic P from a reduction in hypolimnetic P following an alum treatment, unless a steep gradient of P concentration between hypolimnion and epilimnion had been a significant source of P to the epilimnion through diffusion. This prediction cannot be substantiated for Horseshoe, Snake, Shadow, and Dollar Lakes because the role of diversion cannot be separated from the effects of alum. Mirror Lake appears to have been improved from the alum, though this effect could have been due to a greatly reduced spring mixing P which carried into the summer. Only the West Twin Lake treatment has had the benefit of a reference or control lake. This project demonstrated that diversion and not alum was the primary factor in the decrease of epilimnetic P concentrations. The alum application was effective and long lasting in reducing sediment P release in these deeper lakes, but it appears that diversion of external loading was a major reason for improved trophic state.

A significant reduction in external nutrient loading remains as the critical first step in reducing algal blooms of lakes. In stratified lakes, it appears that control of hypolimnetic sediment P release with alum is most likely to result in lowered epilimnetic P when lake morphometry had permitted partial mixing during summer months, when significant ice-cover P accumulation is controlled so that spring mixing concentrations are less, or when diffusion of P from hypolimnion to epilimnion was a major P source.

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