

## Factors influencing internal P loading in a western Michigan, drowned river-mouth lake

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**Abstract.** Release of P from lake sediments may account for a significant portion of a lake's total P (TP) load. Previous studies using sediment cores showed that ~65% of the total P load entering Spring Lake, Michigan, came from the sediments, and that an alum concentration of 24 mg Al/L effectively inactivated P release in experimental sediment-core tubes. In 2004, we studied the influence of alum concentration and sediment resuspension on P release rates from the sediments. Based on laboratory incubations using alum concentrations of 0, 5, 10, 15, 20, and 25 mg/L, we determined that P release rates at alum concentrations  $\geq 15$  mg/L were no different than at release rates at concentrations of 25 mg/L. Resuspension of sediments substantially increased TP concentrations, even at high alum concentrations, but total soluble P concentrations remained low in the water when alum was present. Alum application may be an effective tool to reduce P flux from sediments in shallow lakes, but external P load reduction must accompany alum application to address the long-term impacts associated with cultural eutrophication.

**Key words:** internal phosphorus loading, alum, shallow lakes, cultural eutrophication, sediment resuspension, Michigan.

Internal loading of P refers to the release of P from lake sediments. P often accumulates in sediments when external loading of P (from the watershed and atmosphere) is high. Sediment P can be transferred to the water column by: 1) diffusion of soluble P from sediments during periods of anoxia, and 2) resuspension-driven processes whereby soluble P is desorbed from particulate matter or released from the pore water (Selig 2003). Internal loading is a frequent phenomenon in shallow, eutrophic lakes throughout the world and can account for a substantial amount of the total P (TP) load entering the water column. Indeed, internal loading may prevent lake water quality from recovering even after external loads are reduced (Sas 1989, Granéli 1999) because of the release of P from sediments (Bjork 1985, Granéli 1999, Steinman et al. 1999).

Spring Lake has some of the highest P concentrations measured in western Michigan lakes, with TP levels averaging 100  $\mu\text{g/L}$  and ranging from 6 to 631  $\mu\text{g/L}$  during ice-free periods from 1999 through 2002 (T. Groves, Progressive AE, personal communication). In response to concerns from residents regarding

impaired water quality in the watershed, laboratory-based studies, conducted in 2003 using sediment cores from Spring Lake, indicated that internal loading accounted for between 55 and 65% of the TP entering the lake water column on an annual basis. Also, an alum application of 24 mg Al/L was extremely effective at reducing TP release from the sediments in the laboratory (Steinman et al. 2004). The results from these experiments provided evidence that internal loading was important and that alum had the potential to effectively mitigate the process. However, uncertainties emerged from that study, including: 1) the effectiveness of alum concentrations  $< 24$  mg/L for reducing internal P loading, and 2) the effect of sediment resuspension on P inactivation by alum. Our present study was designed to examine these 2 uncertainties.

### Methods

#### *Study site*

Spring Lake is located in west-central Michigan and drains into the Grand River, ~1 km east of Lake Michigan (Fig. 1). This drowned river-mouth lake has a complex morphometry, with a surface area of 5.25 km<sup>2</sup>, mean and maximum depths of 6 m and 13 m, respectively, and a water residence time of

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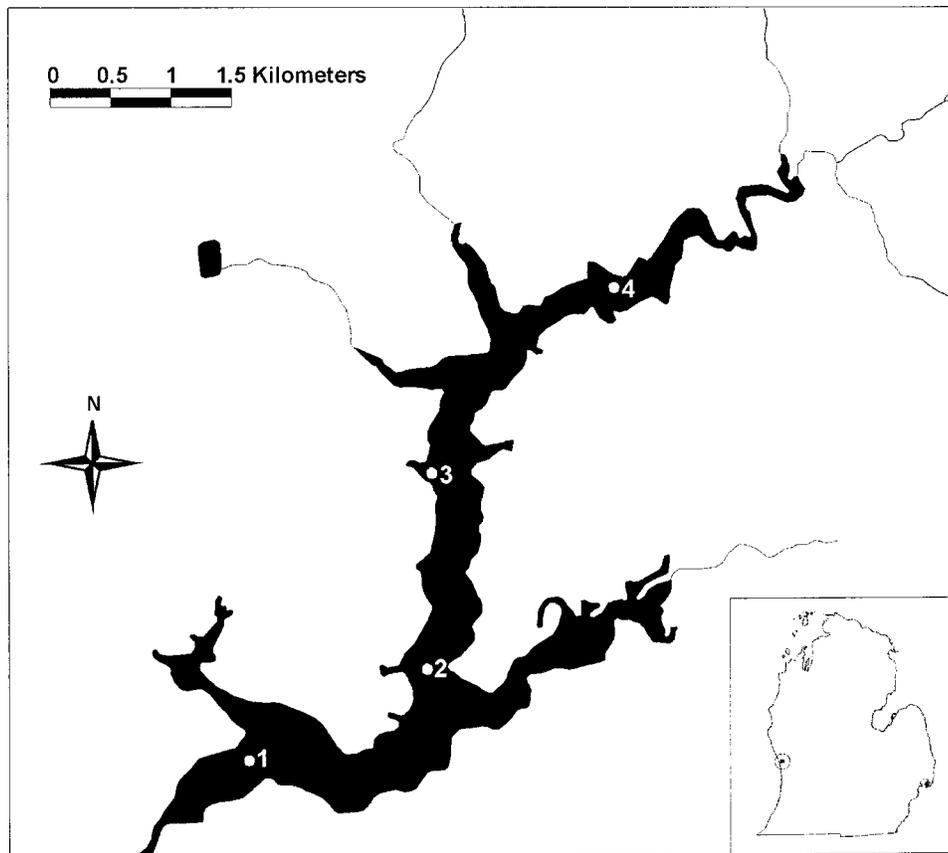


FIG. 1. Sampling sites in Spring Lake, Michigan. Inset shows location of Spring Lake (encircled dot) within the lower peninsula of Michigan.

~5 mo in the winter and ~11 mo in the summer. The watershed is 134 km<sup>2</sup>, and land use is split among forest/undeveloped (57%), urban (19%), agriculture (18%), and wetland (6%) (Lauber 1999). The lake's shoreline is densely populated with primary residences, and macrophytes cover ~27% of the lake's surface area (Lauber 1999). The mixing regime of Spring Lake is complex, varying among years and locations in the lake; in some years the lake remains stratified throughout the summer, whereas in other years stratification breaks down and reforms throughout the summer.

*Experiment 1: effect of alum concentration on P release rate*

*Field methods.*—Sediment cores were collected from 4 sites in Spring Lake on 20 April 2004. The sites were selected to provide spatial coverage of different parts of the lake (Fig. 1) and to be as close as possible to sampling locations from prior investigations to take advantage of historical information.

At each site, vertical profiles of dissolved O<sub>2</sub>, pH, temperature, specific conductance, chlorophyll *a*, and total dissolved solids were measured using a Hydrolab

DataSonde 4a equipped with a Turner Designs fluorometer. Water samples for nutrient analysis were collected with a Van Dorn bottle and maintained on ice until delivery to the laboratory. P analyses were performed on a BRAN+LUEBBE Autoanalyzer (AA) using the automated ascorbic acid method (USEPA 1983). A 15-mm flow cell and a 660-nm filter were used for the analyses, and the AA was calibrated using a 6-point calibration curve. The standard detection limit for TP and soluble reactive P (SRP) was 10 µg/L. Sample volumes were limited (10 mL) to minimize the volume of replacement water in each core tube; these limited volumes required all P samples to be diluted by 2× to complete the digestion. Quality assurance/quality control (QA/QC) procedures followed method guidelines including use of 10% method blanks and 10% matrix spikes/matrix spike duplicates (precision and accuracy ±15%).

Six sediment cores were collected from each site using a piston corer (Fisher et al. 1992, Steinman et al. 2004). The corer was constructed of a graduated 0.6-m-long polycarbonate core tube (7-cm inner diameter)

and a polyvinyl chloride (PVC) attachment assembly for coupling to Al drive rods. The piston was advanced 20 to 25 cm prior to deployment to maintain a water layer on top of the core during collection. The corer was positioned vertically at the sediment–water interface and pushed downward with the piston cable remaining stationary. After collection, the core was brought to the surface, and the bottom was sealed with a rubber stopper prior to removal from the water, resulting in an intact sediment core that was ~20 cm in length, with a 25-cm overlying water column. The piston was then bolted to the top of the core tube to keep it stationary during transit. Core tubes were placed in a vertical rack and maintained at ambient temperature during transit.

*Laboratory methods.*—The 24 cores (6/site) were placed in a Revco environmental growth chamber, with the temperature maintained to match ambient conditions in Spring Lake at the time of collection. Cores from each site were exposed to each of 6 alum concentrations (0, 5, 10, 15, 20, or 25 mg Al/L) by introducing ~0, 114, 227, 341, 454, or 568 mg of alum (as aluminum sulfate solution; General Chemical Corporation, River Rouge, Michigan), respectively, into the water column of each core tube. The alum formed a floc at the water–sediment surface. Alum treatments began within 10 h of core collection in the field. Initial samples (time 0) were taken prior to alum introduction. The water column of each core tube was bubbled with N<sub>2</sub> (with 330 ppm CO<sub>2</sub>) to create buffered anaerobic conditions.

Internal load estimates were made using the methods outlined in Moore et al. (1998), with minor modifications (Steinman et al. 2004). In brief, a 40-mL water sample was removed by syringe through the sampling port of each core tube at 2 h, 8 h, 1 d, 2 d, 4 d, 8 d, 12 d, 16 d, 20 d, and 24 d after time 0, and replaced with an equal volume of filtered lake water. Immediately after extraction, a 20-mL subsample was refrigerated for analysis of TP, and a 20-mL subsample was filtered through a 0.45- $\mu$ m membrane filter and frozen for analysis of total soluble P (TSP). P was analyzed as described above.

Flux (P release rate) calculations were based on the increase in water-column TP or TSP using the following equation (Steinman et al. 2004):

$$P_{rr} = (C_t - C_0)V/A$$

where,  $P_{rr}$  is the net P release rate or retention per unit surface area of sediments,  $C_t$  is the TP or TSP concentration in the water column at time  $t$ ,  $C_0$  is the TP or TSP concentration in the water column at time 0,  $V$  is the volume of water in the water column, and  $A$  is the planar surface area of the sediment cores. P release

rate was calculated from the time when P concentrations stabilized in the water column (~day 1) until the date on which an asymptote was reached.

Following the incubations, cores were centrifuged to remove excess pore water and the top 10 cm of each core was removed and analyzed for TP according to the methods described above.

*Statistical analysis.*—Given the effectiveness of alum in binding P, the P concentration of many samples was below our detection limit (10  $\mu$ g/L). These data were assigned a value of ½ the detection limit. This approach is commonly used despite its limitations (Helsel 2005) but alternative approaches to estimate P concentrations for values below our detection limit, such as maximum likelihood estimation and regression on order statistics, failed because of the small sample sizes in our data set (cf. Helsel 1990, Shumway et al. 2002). Differences in TP and TSP release rates among 4 sites (blocks) and 6 alum treatments were analyzed by a randomized block 2-way analysis of variance (ANOVA). P release data were successfully normalized by ln-transformation (as determined by the Kolmogorov–Smirnov test). If the overall model was significant, the Holm–Sidak multiple comparison test (MCT) was used to analyze differences in TP and TSP release rates among alum treatments. Sediment TP concentration also was analyzed with a randomized block 2-way ANOVA; data transformation was not necessary as determined by the Kolmogorov–Smirnov test.

#### *Experiment 2: effect of sediment resuspension on P concentration*

*Laboratory methods.*—At the end of experiment 1 (day 24), all cores were vigorously agitated by increasing the flow rate of gas to induce resuspension. Flow rates were increased to the point where resuspension of sediments was similar among cores. Water samples were collected at 12, 24, and 48 h during resuspension and replaced with an equal volume of filtered lake water; both TP and TSP were sampled, processed, and analyzed using the methods described above.

*Statistical analysis.*—Differences in TP and TSP concentrations among alum concentration treatments were analyzed by a randomized block 2-way ANOVA. P concentration, rather than release rate, was measured in this experiment because the 48-h duration of the experiment was too short to determine maximum rates of release. However, this time period was considered appropriate because sediment resuspension events tend to be short-lived. P concentrations were normalized by ln-transformation (as determined by the Kolmogorov–Smirnov test), and if the overall

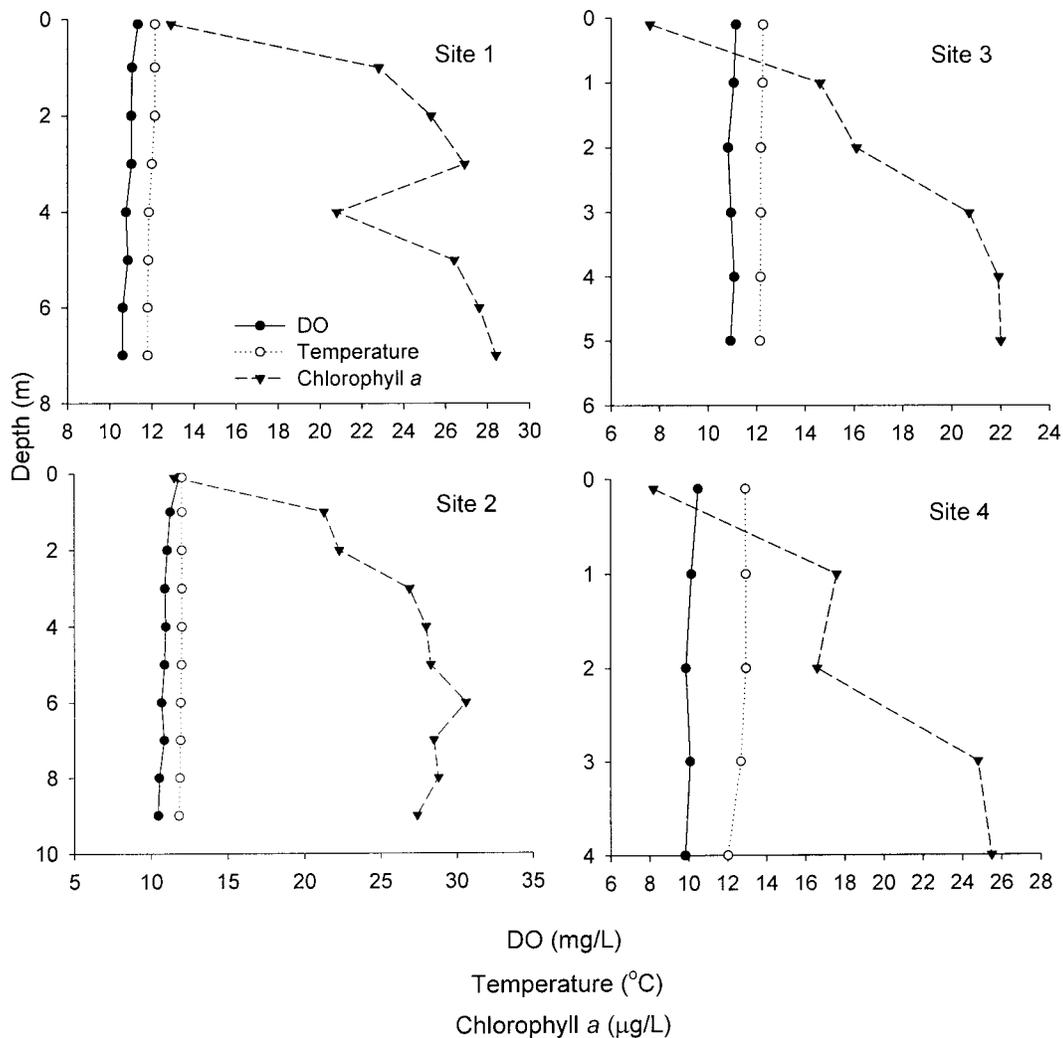


FIG. 2. Dissolved O<sub>2</sub> (DO), temperature, and chlorophyll *a* values in Spring Lake, sampled 20 April 2004.

model was significant, mean differences were compared using MCT. Data that were below our detection limits were assigned a value of ½ the detection limit, as described above.

## Results

### *Experiment 1: effect of alum concentration on P release rate*

**Field results.**—Water quality was similar among sites during the April cruise (Fig. 2). Temperature and dissolved O<sub>2</sub> (DO) concentration were constant through the water column, with temperature ~12°C and DO ~10 mg/L. Chlorophyll *a* concentrations generally increased with depth at all sites; this increase may have been caused by photoinhibition near the water surface as irradiance levels ranged from 861 to 2717 µmol photon m<sup>-2</sup> s<sup>-1</sup> at the surface and declined

to between 6 and 31 µmol photon m<sup>-2</sup> s<sup>-1</sup> at the 3-m depth. Secchi-disk values were 1.0 m at all sites except site 3, which was slightly less turbid (Secchi depth = 1.25 m). Mean surface and near-bottom water TP concentrations from all 4 sampling sites were 50 µg/L (SD = 11 µg/L) and 58 µg/L (SD = 11 µg/L), respectively.

**Laboratory results: P release rates.**—Even low concentrations of alum were effective at reducing the release of TP from Spring Lake sediments (Fig. 3). In the absence of alum, TP concentrations reached their maxima by days 16 to 20, with maximum TP concentrations in the water column reaching 194, 374, 66, and 48 µg/L at sites 1 to 4, respectively (Fig. 3). TP concentrations were usually near, or below, our 10-µg/L detection limit when alum was present, regardless of the alum concentration (Fig. 3). Sediment TP concentrations (mg/kg dry mass) were signifi-

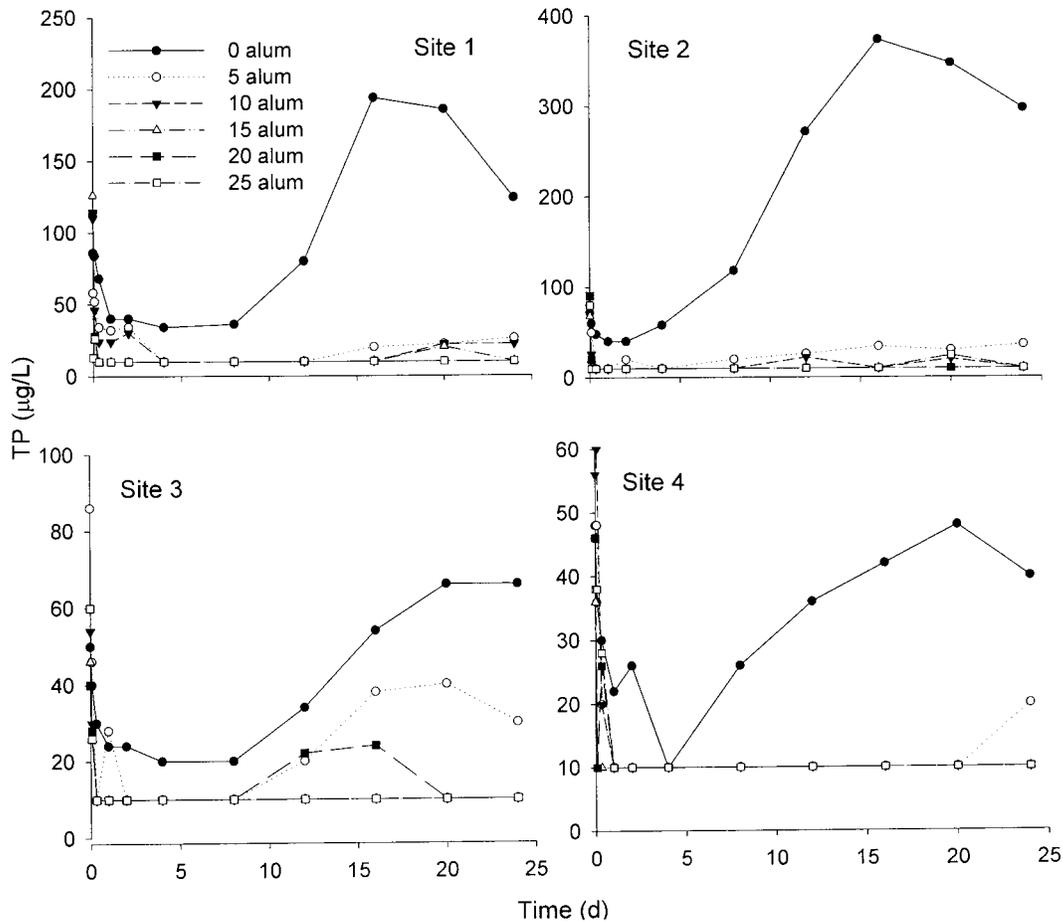


FIG. 3. Total P (TP) concentrations in water columns of core tubes exposed to alum concentrations of 0, 5, 10, 15, 20, or 25 mg/L. Note the different scales for the y-axes.

cantly greater at sites 1 and 2 than sites 3 and 4 ( $p < 0.001$ ; means  $\pm$  SD:  $1512 \pm 179$ ,  $1592 \pm 84$ ,  $1152 \pm 84$ ,  $1171 \pm 84$  at sites 1–4, respectively). Hence, sites with the most sediment TP also released the most P into the water column of the core tubes, although this relationship was not proportional.

Mean TP release rates ranged from 0 to  $5.86 \text{ mg m}^{-2} \text{ d}^{-1}$ . Alum treatments had a statistically significant effect on P release rates ( $F_{5,15} = 4.56$ ,  $p < 0.02$ ), but release rates did not differ among sites ( $F_{3,15} = 1.45$ ;  $p > 0.26$ ). Release rates were significantly greater in cores without alum than in the 15-, 20-, and 25-mg Al/L alum treatments (MCT,  $p < 0.05$ ; Fig. 4). TP release rates in the 15-, 20-, and 25-mg Al/L alum treatments were  $< 0.10 \text{ mg m}^{-2} \text{ d}^{-1}$  and did not differ (MCT,  $p > 0.05$ ; Fig. 4).

Mean TSP concentrations were rarely above detection limits in cores with alum, regardless of the concentration applied. Even in the 0-mg/L alum treatment, detectable concentrations of TSP were not measured until day 12 in cores from any site except

site 2, where detectable TSP was measured on day 1 (data not shown).

*Experiment 2: effect of sediment resuspension on P release rate*

The increase in mean TP concentration (i.e., TP at 48 h after resuspension minus TP on day 24 [before resuspension began]) ranged from 129 (0-mg Al/L alum treatment) to  $487 \text{ } \mu\text{g/L}$  (20-mg Al/L alum treatment) (Fig. 5A). Resuspension was a major source of TP in the water column, and concentrations were much greater after 48 h of resuspension than after 24 d of P release in all alum treatments (Fig. 5A). Alum treatment had no statistically significant effect on the increase in TP concentration ( $F_{5,15} = 1.67$ ;  $p > 0.20$ ), largely because of the high variances in the 15-, 20-, and 25-mg/L alum treatments (Fig. 5A). Site had a significant effect ( $F_{3,15} = 9.41$ ;  $p < 0.001$ ), with the increased TP concentration at site 2 ( $653 \text{ } \mu\text{g/L}$ ) greater than at the other sites (306, 159, and  $50 \text{ } \mu\text{g/L}$  at sites 1, 3, and 4, respectively).

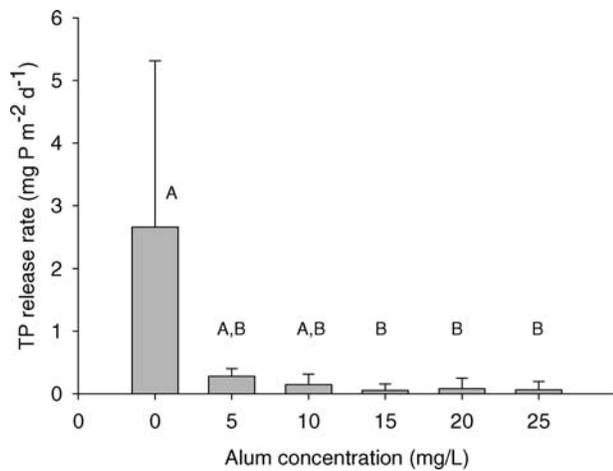


FIG. 4. Mean total P (TP) release rates from sediments exposed to alum concentrations of 0, 5, 10, 15, 20, or 25 mg/L. Release rates were based on changes in TP concentration between days 1 and 20.

The increase in mean TSP concentration ranged from 0 (20- and 25-mg Al/L alum) to 80  $\mu\text{g/L}$  (0-mg Al/L alum) (Fig. 5B). Alum treatment significantly affected the increase in TSP concentration ( $F_{5,15} = 9.06$ ;  $p < 0.001$ ). Increases in mean TSP were significantly higher in the 0-mg Al/L alum treatment than in all alum treatments, whereas increases in mean TSP concentration did not differ among alum treatments (MCT,  $p < 0.05$ ). Similar to TP, site had a significant effect ( $F_{3,15} = 5.23$ ;  $p < 0.02$ ), with the increased TSP concentration at site 2 (27  $\mu\text{g/L}$ ) greater than at sites 1 and 4, but not at site 3. However, given the large number of values below detection (60%), these differences should be viewed with caution.

### Discussion

Internal loading can be a significant source of nutrients in shallow, eutrophic lakes, and can result in serious impairment of water quality (Welch and Cooke 1995, Moore et al. 1998, Granéli 1999, Søndergaard et al. 2001). One restoration technique that can be applied in these systems is chemical inactivation, such as the use of alum (Cooke et al. 1993, Haggard et al. 2005). A laboratory-based study using Spring Lake sediments was conducted in 2003, but only one alum concentration was examined, and the influence of resuspension on sediment P inactivation by alum was not investigated (Steinman et al. 2004).

#### Concentration of alum needed to inactivate sediment P

Steinman et al. (2004) tested the maximum environmentally safe dose of alum for Spring Lake (24 mg Al/L). This concentration was chosen based on the

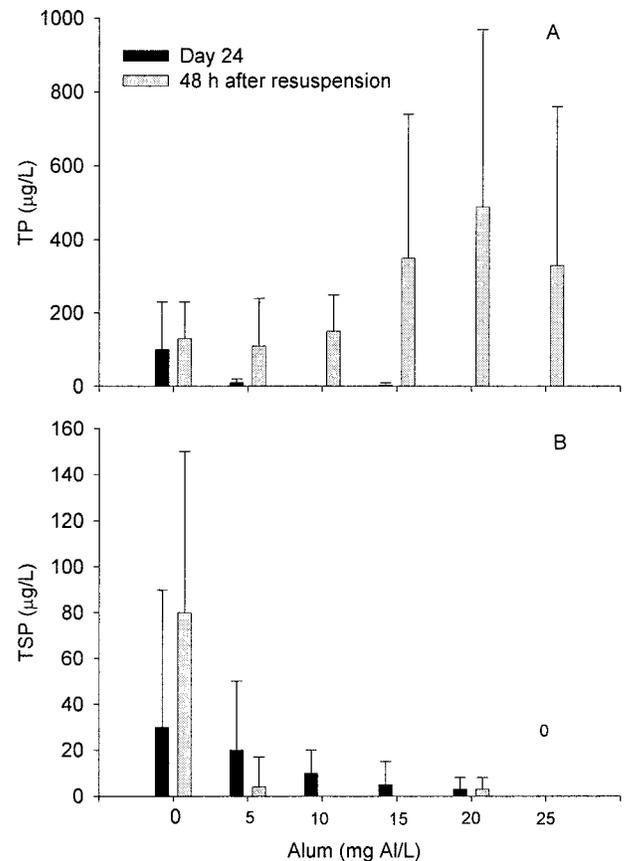


FIG. 5. Mean (+1 SD) total P (TP; A) and total soluble P (TSP; B) concentrations in water columns of core tubes, averaged across 4 sites. Day 24 values (black bars) reflect the net change in TP or TSP from time 0 to day 24 under static conditions. Values 48 h after resuspension (grey bars) reflect net changes in TP (or TSP) 48 h after resuspension minus TP (or TSP) on day 24 before resuspension began.

approach developed by Kennedy and Cooke (1982), who used lakewater alkalinity to establish maximum environmentally safe doses and then applied this maximum dose to the sediments (Cooke et al. 1993). A dose of 24 mg Al/L effectively inactivated P in the Spring Lake sediment cores (Steinman et al. 2004), but it was unclear if a lower concentration would have resulted in an equal level of P inactivation. Our current results suggest that an alum application of 15 mg/L should be as effective as one of 25 mg/L at reducing TP concentrations in the water column, and an alum application of 5 mg/L should be as effective as one of 25 mg/L at reducing TSP concentrations. However, our experiments were conducted under static laboratory conditions, where the alum remained in place and could bind efficiently to P. In a lake environment, wind and wave action result in sediment resuspension and mixing, and the effectiveness of alum application probably will be lower than in the laboratory. Instead

of a stationary layer of alum floc, as was observed in the core tubes, alum coverage may be variable, with some areas having thick layers of floc and other areas having little or none.

Application of higher concentrations of alum results in better physical coverage of lake sediments and allows surplus Al to adsorb to bioavailable P mobilized from sediments (Gächter et al. 1988, Gächter and Meyer 1993). However, the fate of the Al is of environmental concern. Rydin et al. (2000) found that stable Al-P complexes formed in the sediments of alum-treated lakes, and that P adsorbed by Al was transformed from mobile P to permanently bound P. Welch and Cooke (1999) found that Al concentrations in the sediment of 21 alum-treated lakes generally were no higher than background Al concentrations. They attributed the high background Al levels to sedimented clays and the downward migration of the floc in the low-density sediment of the lakes. Hence, although an alum concentration of 15 mg/L should provide a similar degree of TP inactivation as 24 mg/L, the higher concentration is not expected to result in negative environmental consequences based on the alkalinity and pH levels present in Spring Lake (Kennedy and Cooke 1982, Cooke et al. 1993). Moreover, the higher alum concentration increases the likelihood of a successful field application.

#### *P release from the sediments during resuspension*

P release from sediments is enhanced by physical mixing (e.g., Andersen 1974, Ahlgren 1980, Holdren and Armstrong 1980, Søndergaard et al. 1992, Reddy et al. 1996). Boström et al. (1982) identified several mechanisms that could increase P concentrations from wind-induced turbulence: 1) diffusive flux will increase because dissolved P leaving the sediments will disperse rapidly, maintaining a steep concentration gradient across the sediment-water interface; 2) rapid dispersion of dissolved P from the sediments should accelerate equilibrium-regulated processes; and 3) resuspension of particles may occur, increasing TP in the water column. SRP concentration in the water column also increases after experimental resuspension events (e.g., Søndergaard et al. 1992). The increase in water-column SRP concentration typically occurs as a result of liberation of dissolved P from sediment porewater or its desorption from particles suspended in the water column. However, sediments also can serve as a sink for P because mixing events and bioirrigation can create oxidized conditions in the sediment that cause P to precipitate after its association with Fe oxyhydroxides (Olila and Reddy 1993, Hansen

et al. 1997, Lewandowski and Hupfer 2005). Ultimately, the equilibrium P concentration will dictate whether or not sediments serve as a sink or a source of P to the water column of any particular system.

Sediment resuspension introduced a substantial amount of TP into the water column, even in the presence of alum. At the pH of Spring Lake water, alum dissociates to give trivalent  $\text{Al}^{3+}$  ions, which hydrolyze rapidly to form soluble monomeric and polymeric species and an amorphous  $\text{Al}(\text{OH})_3$  floc. The monomeric species can precipitate soluble P as  $\text{Al}(\text{PO}_4)$ , whereas the floc can remove soluble and particulate forms of P by adsorption or physical entrapment (Bottero et al. 1980, Galarneau and Gehr 1997, Omoike and Valoon 1999). The relatively low-density, flocculent layer of alum can strip P from the water column as it settles to the sediment surface. However, this P-rich, flocculent material is very susceptible to resuspension, and resuspension of sediment with its alum-bound P can raise P concentrations in the water column compared to conditions without sediment resuspension. It is unclear if resuspension will result in nutrient heterogeneity within the water column in nature. Despite very different P release rates among sites in our study, the Spring Lake water-column P levels were relatively similar. Advective mixing, biotic uptake, and sediment focusing all may mask the localized differences in nutrient concentration associated with internal loading. Collectively, however, this process can account for a substantial load to shallow, eutrophic lakes.

The significant increase in TSP after resuspension in the 0-mg Al/L alum treatment indicated that much of the P liberated from the sediment was in a bioavailable form, consistent with results from previous studies that showed increases in SRP after resuspension (Søndergaard et al. 1992, Reddy et al. 1996). James and Barko (2004) reported that P desorption from total suspended solids is an important source of bioavailable P in the Mississippi River, and that, in the presence of alum, TSP concentrations remained low even during resuspension. This result also was observed in our study and suggests that, at least in the short term, an alum treatment in Spring Lake should result in relatively little bioavailable P for algal uptake, even during sediment-mixing events.

#### *Long-term effectiveness of alum treatments*

Welch and Cooke (1999) evaluated the effectiveness and longevity of alum applications in 21 lakes across the USA. In polymictic lakes, the internal loading rate was reduced by alum treatment in 6 of 9 cases; the average reduction was ~67% and lasted 5 to 11 y. In

dimictic lakes, the internal loading rate was reduced in 7 of 7 cases; the average reduction was ~80% and lasted for 4 to 21 y. The major constraint in polymictic lakes was that macrophytes interfered with application. In 1998, submerged aquatic plants covered ~27% of the sediment surface of Spring Lake (Lauber 1999), and coverage was greatest in shallow arms (bayous) of the lake (Fig. 1). Hence, to optimize the alum effectiveness in Spring Lake, applications should either avoid the bayous or be conducted during periods when macrophyte biomass is low.

Our results suggest that doses lower than maximum environmentally safe concentrations have the potential to reduce P release from sediments in shallow lakes. However, our results also have several qualifications. First, the study was conducted under optimal conditions for P binding (i.e., anaerobic, static water column). In Spring Lake, sediment resuspension is common and macrophytes may be present. Thus, the recommended alum dose (24 mg/L) for Spring Lake (Cooke et al. 1993) is more likely to ensure P inactivation than lower concentrations, although the cost may be greater. Second, many samples, especially TSP (86% in experiment 1; 60% in experiment 2), were below the analytical detection limits. This reflected the effective sorption capacity of alum, but in many cases it also prevented determining the actual degree to which P was reduced by alum. Even without an exact determination, these P reductions represent a significant ecological improvement in Spring Lake given its eutrophic status.

Alum application is a short-term solution to the longer-term problem of internal P loading. Welch and Cooke (1999) concluded that a reasonable expectation of longevity of benefits from alum treatments is 10 y in polymictic and 15 y in dimictic lakes. However, impacts associated with nonpoint sources of P in impaired lakes can last for hundreds or thousands of years (Carpenter 2005). Hence, it is critical that we address the underlying reasons for impaired water quality. External load reduction must complement any chemical addition (Hansson et al. 1998) regardless of the long-term effectiveness of alum treatment. Reducing stormwater discharge, conversion of septic systems to sewers, use of low- or no-P fertilizer, and implementation of other best management practices should be emphasized and promoted wherever possible in the Spring Lake watershed and other similarly impacted systems (Walsh et al. 2005).

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