High Inter- and Intra-lake Variation in Sediment Phosphorus Pools in Shallow Lakes

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Key Points:

- The size and chemical composition of the sediment phosphorus pool are spatially heterogeneous both within and among shallow lakes
- Differences in sediment phosphorus chemistry among lakes indicate that internal loading mechanisms may also vary across lakes
- Understanding intra-lake variation in sediment P pools informs the spatial sampling resolutions necessary for accurate stock assessments

Abstract

Phosphorus (P) release from lakebed sediments may fuel algal blooms, especially in shallow systems. A primary mechanism that controls internal P loading is the size and chemical composition of the sediment P pool. However, variation in sediment P speciation within and among shallow lakes remains poorly quantified, limiting efforts to scale and model sediment P pools. We measured the degree of spatial heterogeneity in the size and composition of the sediment P pool, both within and among lakes, for seven shallow glacial lakes by measuring sediment P fractions from 10 cores in each lake. There was a 1.6x difference in total sediment P among the study lakes, with sediment P composition varying based on the dominant watershed soil series. We also found that higher mobile P (as a fraction of total P) in the profundal sediments was significantly correlated with higher average chlorophyll-*a* concentrations ($p=0.04$), indicating the influence of sediment P composition on algal biomass in shallow lakes. Additionally, we measured substantial within-lake heterogeneity in total and loosely-bound sediment P among the 10 sites sampled in each lake. Concentrations were positively correlated with the depth of the water column above the sediments such that extrapolating measurements from the deep site alone could overestimate whole-lake mean P concentrations. Our results provide insight into the magnitude and pattern of inter- and intra-lake variation in sediment P pools that should be accounted for when sampling, scaling measurements, and modeling sediment P dynamics.

Plain Language Summary

Phosphorus (P) is an essential nutrient in freshwater ecosystems; however, in excess it can cause algal blooms. P enters lakes from the watershed (external loading) or when released from

lakebed sediments into the overlying water (internal loading). Sediment P chemistry controls internal loading because some forms of P are more susceptible to release. However, we lack a quantitative understanding of how sediment P chemistry can vary within individual lakes and among different waterbodies. We measured variation in sediment P composition within and among seven shallow lakes. We found that sediment P composition varied across lakes based on watershed soil characteristics. We also found that a higher abundance of mobile P forms in the sediments was correlated with higher concentrations of algal pigments, indicating that sediment P composition may influence algal biomass in shallow lakes. Within individual study lakes, sediment P was highly variable, meaning that multiple samples are required to capture this heterogeneity. Our results reveal the scale of spatial variation in sediment P forms in shallow lakes. Since sediment P composition is a primary mechanism controlling sediment P release, this knowledge is critical to building accurate models to predict internal P loading.

1 Introduction

Sediments are an integral component of aquatic ecosystems, especially shallow lakes. The sediments regulate whole-lake biogeochemical cycles through diagenetic processes and material exchange across the sediment-water interface (Forsberg, 1989; Golterman, 2004). For example, phosphorus (P) stored in sediments may reenter the water column due to disturbance, microbial activity, or changes in chemical conditions at the sediment surface (Boström et al., 1988; Søndergaard et al., 2003). In many ecosystems, this sediment P release (i.e., internal P loading) can maintain high water column P concentrations, even if external nutrient inputs are reduced (Søndergaard et al., 1999; Jeppesen et al., 2005; Søndergaard et al., 2013). Excess P availability supports algal production (Elser et al., 2007; Schindler et al., 2016), which can reach nuisance levels, threatening ecosystem services and public health (Schindler & Vallentyne, 2008). The morphometric features of shallow lakes make these systems particularly susceptible to internal P loading. Specifically, shallow basins have a large sediment surface area relative to water volume. As a result, sediment-water interactions are highly influential in determining whole-lake water chemistry (Welch & Cooke, 1995, Søndergaard et al., 2003). Although it is known that internal P loading is an important process in shallow lakes, variation in the underlying mechanisms remains poorly quantified for these ecosystems.

A fundamental mechanism controlling internal P loading in shallow lakes is the size and composition of the sediment P pool. Sediment P may be found in a wide variety of minerals and organic materials as well as sorbed to particle surfaces (North et al., 2015). This chemical speciation determines the conditions in which P is mobilized and released as different P forms are vulnerable to different internal loading processes (Orihel et al., 2017). For example, changes in redox potential due to fluctuations in dissolved oxygen and alternative electron acceptor concentrations can mobilize P associated with redox-sensitive minerals (i.e., iron- and manganese-bound P; Mortimer, 1941; Orihel et al., 2015). Changes in pH may release other mineral-bound P forms (e.g., calcium-, and aluminum-bound; Jensen & Andersen, 1992), and microbial decomposition can liberate P incorporated in labile organic materials (Joshi et al., 2015; Song & Burgin, 2017; Frost et al., 2019). Sediment resuspension due to wind disturbance or bioturbation releases pore water and surface-sorbed P (i.e., loosely-bound P *sensu* James 2017b; Ekholm et al., 1997). Loosely-bound P is likely an important form of sediment P in shallow lakes as these ecosystems experience frequent water column mixing and wind disturbance (Bengtsson & Hellström, 1992). Although internal P loading occurs under a variety of conditions, the dominant mechanisms are determined by the chemical composition of the sediment P pool.

Sediment P content and speciation vary among lakes due to differences in watershed features and external loading as well as autochthonous processing. External P loading includes particulate and dissolved forms that are transported from the watershed into lakes through surface and subsurface flows (Carpenter et al., 1998; Pease et al., 2018). The P speciation in catchment soils will drive the chemical composition of particulate P inputs, in turn influencing the composition of the sediment P pool (Kerr et al., 2011; Tang et al., 2019). However, selective soil erosion and transport processes may alter sediment composition relative to the soil matrix if various P fractions are not distributed evenly across grain sizes. Fine particles that are more easily transported are often enriched in exchangeable, organic, and iron- and aluminum-bound P and depleted in calcium-bound P relative to larger grain sizes (Stone & English, 1992). As a result, lakebed sediments may have more labile P forms than the surrounding soils (Kerr et al., 2011). In addition to exogeneous loading of particulate forms, P may also enter waterbodies in an aqueous form or desorb from settling particles before reaching the sediments (Pease et al., 2018). This soluble P is readily taken up by aquatic primary producers. Following biological uptake, P may be cycled among producers and consumers or it may settle out of the water column as necromass, algal detritus, or macrophyte leaf litter (Orihel et al., 2017). The sources of sedimented organic matter influence sediment P composition and lability (Twilley et al., 1986). For example, if algal detritus dominates organic matter sedimentation, then sediment organic P will be associated with more labile materials. Overall, sediment P content and composition vary among different lakes due to differences in exogeneous inputs and *in situ* processing.

The spatial distribution of the sediment P pool is also heterogeneous within individual shallow lakes following variation in sediment transport, deposition, and resuspension processes (Mackay et al., 2012). Sediment total P generally increases with water depth as a result of sediment focusing, which concentrates fine-grained, P-enriched sediments in deeper portions of the lake basin (White & Stone, 1996; Hou et al., 2014; Cardoso-Silva et al., 2018). Dynamic disturbance and biological activity across the lakebed further drive spatial variation in sediment P speciation in shallow waterbodies (Forsberg, 1989; Nowlin et al., 2005; Kowalczewska-Madura et al., 2019a). Variable sediment disturbance has the greatest influence on the distribution of loosely-bound P forms. Smaller stocks of loosely-bound P are expected in shallow sediments that experience wind or wave disturbance while higher loosely-bound P content is expected in deeper, more protected sediments. (James, 2017a). Additionally, the location and density of macrophyte beds can influence the spatial distribution of sediment P pools as macrophytes take up pore water P from the sediments and mobilize other P forms in order to meet their nutrient requirements (Xing et al., 2018). Macrophyte root structures also stabilize and oxygenate rhizosphere sediments, which can protect stocks of loosely-bound and redox-sensitive P species by reducing resuspension and maintaining aerobic conditions (Carpenter et al., 1983; Madsen, 2001; Lai et al., 2011). Sediment focusing and disturbance as well as the presence of macrophytes are expected in shallow waterbodies (Kelton & Chow-Fraser, 2005). As a result, the spatial distribution of sediment P content and composition are expected to be heterogeneous within shallow lakes. Accounting for this spatial variation is essential for accurate inventories of sediment P pools, burial rates, and internal loading potential (Mackay et al., 2012; Orihel et al., 2017). However, the degree of spatial heterogeneity in sediment P content and composition in shallow lakes remains poorly quantified.

In order to quantify the spatial heterogeneity of sediment P species in shallow lakes we measured the sediment P pools of seven shallow, glacial lakes in northwest Iowa. Specifically, we collected sediment cores from ten locations within each lake and analyzed the sediments for total P as well as pore water and loosely-bound P. We quantified spatial variation in these P forms and tested how sampling resolution influenced estimates of mean sediment P concentrations. Water depth is expected to drive horizontal variation in sediment P within lakes, with higher concentrations found in deeper areas of the lake. We further hypothesize that lakes with more variable bathymetry will have greater variability in loosely-bound P, driven by variation in sediment disturbance and macrophyte occurrence. The sediment core from the deepest site in each study lake was additionally analyzed for total P and five sediment P species (loosely-bound P, redox-sensitive, labile organic, and calcium-, and aluminum-bound fractions). We hypothesize that the variation in the composition of the sediment P pool among lakes will relate to watershed soil composition, land cover, and long-term algal biomass. Understanding spatial variability in sediment P pools within and among shallow lakes is necessary to measure, scale, and model lentic P dynamics. A quantitative understanding of this variation is a critical step to being able to predict internal P loading potential based on lake and watershed characteristics.

2 Materials and Methods

2.1 Study Lakes

We surveyed seven shallow, glacial lakes in northwest Iowa, USA (Table 1). The spatial extent of the study lakes covers approximately 5,600 km^2 on the western edge of the Des Moines Lobe. Although the study systems cover a wide range of basin characteristics, all are shallow (average mean depth = 2.2 m) and follow a discontinuous cold polymictic stratification and mixing pattern (Lewis 1983; Table S1). These lakes were formed from kettle depressions at the end of the Wisconsin glaciation. The watershed soils developed from loamy glacial till and are now heavily influenced by row crop agriculture (Arbuckle & Downing, 2001; Tables S2-3). The watersheds of Storm and Swan Lake also contain soils developed from loess deposits as these catchments are located on the boundary of the Des Moines Lobe and the Loess Prairies of western Iowa. All of the study lakes are classified as eutrophic or hypereutrophic due to high phosphorus concentrations (Carlson, 1977).

2.2 Sediment Core Collection and Analysis

We collected ten sediment cores from each study lake once between mid-June and early July of 2018 using a gravity corer (inner diameter 5 cm). Sampling sites included the deep point of the lake (hereafter "profundal sediments"), from which all sediment P species and total P were measured, as well as nine shallower sites distributed across the lakebed, from which only loosely-bound and total P were measured. These additional sites were selected using a randomized sampling scheme stratified by water depth. Specifically, sites were randomly selected from an evenly spaced grid of potential sites within each depth contour, defined as 0.5 or 1 meter intervals depending on the maximum depth of the lake. All sediment cores were sliced in the field immediately following collection. Cores from the profundal site, the zone of sediment accumulation, were sliced every 2 cm up to 10 cm deep and then every 5 cm up to 20 cm deep. One slice from 0-10cm was collected for each of the other nine shallow sediment cores. Sediment samples were sealed in plastic bags with the air removed and transported and stored at 4°C until analysis. All analyses began within 36 hours of sample collection.

The sediments from all ten collection sites were thoroughly homogenized within the sample bags before subsampling. For analysis of physical characteristics, a 0.4 to 0.5 g subsample was first measured into a pre-weighed, oven-dried aluminum weigh boat. The subsamples were then placed in a drying oven at 104 $\pm$ 1°C for at least 2 hours to dry to a constant mass. The wet and dry masses of each subsample were used to calculate sediment moisture content (MC; Text S1, Eq. 1). The samples were then combusted in a muffle furnace for 2 hours at 550°C and weighed again to calculate organic matter content as loss-on-ignition (Text S1, Eq. 2) and estimate bulk density (Text S1, Eq. 3; Håkanson & Jansson 2002).

An additional sediment subsample was analyzed for P speciation via sequential extraction following the methods of James (2017b). This method was modified from the methods of Nürnberg (1988) and Psenner and Puckso (1988). To begin the sequential P extractions, subsamples of fresh sediment equivalent to 0.2 g of dry sediment (Text S1, Eq. 4) were weighed into 50 mL polyethylene centrifuge tubes. The same sediment pellet was used throughout the sequential extraction procedure. All extractions were performed at 25°C unless otherwise stated. Centrifuge time and speed (30 minutes at 3000 rpm) as well as shaker table speed (120 rpm) were consistent across extractions.

To quantify loosely-bound P (all sampling sites in each lake), sediment was extracted in 25 mL of 1 M NH_4Cl. The samples were placed on a shaker table for two hours and then centrifuged. After pouring off and saving the supernatant, we repeated this extraction on the same sediment pellet. The combined, filtered (0.45µm GF/C filters) supernatants were analyzed for soluble reactive phosphorus (SRP; Standard Methods 4500-P E) to determine the concentration of sediment pore water and loosely-bound P (Text S1, Eq. 5).

For profundal sediment samples, additional extraction steps were performed on each core slice immediately following the NH_4Cl extraction to quantify other P species. Redox-sensitive P species were extracted in 12.5 mL of a 0.11 M bicarbonate – 0.1 M sodium dithionate solution. Sample tubes were placed in a 40°C water bath for 30 minutes and centrifuged. The supernatant was bubbled with air for at least 30 minutes before filtering and analyzing for SRP. Next, we determined the amount of labile organic P and P associated with aluminum oxides with a two-step 0.1 M NaOH extraction. We added 25 mL of the extractant to the sample tubes and shook the samples for 17 hours. After centrifuging, the supernatant pH was adjusted to between 6-8 using 0.1 M HCl. A subsample of the supernatant was filtered and analyzed for SRP to determine the aluminum-bound P concentration while the remainder underwent a persulfate digestion (Standard Methods 4500-P B.5) before SRP analysis to determine the concentration of both aluminum-bound and labile organic P. The labile organic P fraction was determined as the difference between the total NaOH-extractable P and the subsample analyzed for aluminum-bound P. Unfortunately, the persulfate digestion for total NaOH-extractable P failed for sediments from three of the lakes (Swan, North Twin, and South Twin) due to an autoclave malfunction. As such, we were unable to determine labile organic P for these lakes. Since the error occurred during supernatant processing, we are confident that the sequential extractions for other P fractions were not affected. For the final extraction for calcium-bound P, we added 25 mL of 0.5 M HCl to the remaining sediment pellet, shook for 24 hours, and centrifuged. We then used 0.1 M NaOH to adjust the supernatant pH to within 6-8 before SRP analysis. All SRP concentrations were corrected for pH adjustments and standardized by extractant volume and sediment mass to determine the sediment concentration of each P species (Text S1, Eq. 5-9).

To determine the total P concentration, we performed a hot acid digestion on an additional subsample from every sediment sample (both profundal and shallow sites). Sediments were dried for 24 hours, ground into a fine powder with a mortar and pestle, and stored in glass scintillation vials until analysis. For the acid digestion, we first weighed 0.2 g of the dried, ground sediment and combusted it at 550°C for 2 hours. We then boiled the sediment samples on a digestion block in 50 mL of 1 M HCl for 2 hours at 150°C. Following digestion, we brought the samples back up to volume using 50 mL of deionized water. Samples were then pH adjusted within 6-8 using 0.1 M NaOH before analysis of TP (Standard Methods 4500-P B.5, E). All concentrations were corrected for pH adjustments and dilution and standardized by the sediment mass used (Text S1, Eq. 10).

2.3 Aquatic Macrophyte Survey

We evaluated spatial relationships between macrophyte beds and sediment P pools in one of the study lakes, Swan Lake, as there were extensive beds of submersed and floating-leaf macrophytes in this waterbody. High turbidity limited macrophyte colonization in the other study lakes; therefore, we focused our investigation of macrophytes and sediment P pools to Swan Lake. We surveyed aquatic macrophyte community composition and bed density on Swan Lake in July of 2018 (peak of seasonal growth) using the point-intercept method. We followed a grid of 98 sampling sites, evenly-spaced 65 m apart across the lakebed (Ortiz & Wilkinson, 2021). At each sampling point, a two-side rake was lowered to the bottom and used to collect submersed and floating-leaf macrophytes from an approximately 0.4 m^2 area (Mikulyuk et al., 2011). Species presence and a qualitative estimate of density were recorded at each sampling site. We scored bed density on a scale of 0-3 based on whether the site yielded no plants on the sampling rake or in sight of the boat (score of 0), a few plants on the rake or visible within 2 m of the boat (score of 1), enough plants to fill the rake but leave the tines visible (score of 2), or enough plants to completely fill the rake leaving no part of the metal rake head visible (score of 3).

2.4 Statistical Analyses

To determine if profundal sediment P speciation varied among study lakes, we used compositional data analysis and principal components analysis (PCA) on the covariance matrix. Compositional data analysis tests for a difference of proportions among multivariate observations that convey relative information (i.e., parts of a whole). This statistical approach allowed us to test differences in the relative abundance of P fractions among the study lakes without biasing the analysis to the most abundant fractions (Filzmoser et al., 2018). We used the concentrations of sediment P species measured at each depth interval in the core from the deep site of each study lake in this analysis. However, one interval slice from North Twin (0-2cm) and two from Center Lake (2-4cm and 10-15cm) were excluded from the analysis due to insufficient preserved sediment to determine total P. The compositional analysis was defined by the concentrations of loosely-bound (porewater and surface sorbed), redox-sensitive (Fe- and Mn-bound), aluminum-bound, calcium-bound, and total organic P (defined as the sum of labile and refractory organic P). Refractory organic P was calculated as the difference between total P and the sum of loosely-bound, redox-sensitive, aluminum-bound, calcium-bound, and labile organic P. For the study lakes where we were unable to measure labile organic P directly, we calculated total organic P as the difference between total P and the sum of loosely-bound, redox-sensitive, aluminum -bound, and calcium-bound P (Table 2). The sediment P concentrations were center logratio transformed prior to PCA and further analyses. We fit vectors of environmental data to the PCA biplot to explore how watershed land cover, basin morphology, and sediment physical characteristics corresponded to patterns in profundal sediment P speciation among lakes.

In order to understand the effect that the bioavailability of sediment P fractions may have on algal biomass in the study lakes, we performed a linear regression analysis of the percent of the total sediment P pool at the profundal site that is in a mobile form (the combination of the loosely-bound, total organic, and redox-sensitive species) versus the time-averaged chlorophyll-*a* concentrations for the ice-free season, as an idex of algal biomass. Chlorophyll-*a* concentrations are determined based on the U.S. EPA method 445 from a depth integrated sample up to the thermocline or 2 m deep, whichever is shallower (Arar & Collins, 1997; Standard Methods 10200-H). These measurements are made 3-5 times each summer from May through September as a part of the Iowa Department of Natural Resources Ambient Lake and Shallow Lakes monitoring programs. Annual monitoring began in 2000 in all lakes except South Twin where monitoring began in 2006.

To quantify spatial heterogeneity in loosely-bound and total P both within and among lakes, we used measures from the nine spatially-distributed shallow sediment cores and the deep site core slices averaged over depth interval slices. To make comparisons of the variability among lakes we calculated the coefficient of variation for both total P and loosely-bound P in each lake. To test if water depth at the sampling site correlated with the concentration of total P and loosely-bound P, we used linear mixed effects regression with lake as a random effect on the intercept. We used likelihood ratio tests to evaluate the importance of random effects of the lake on model intercepts. We further assessed within-lake variation in Swan Lake by mapping spatial patterns in macrophyte bed density and sediment P concentrations.

We used a rarefaction analysis to determine the spatial sampling resolutions needed for accurate inventories of sediment P pools (Ortiz & Wilkinson, 2021). Specifically, we tested how the number of sampling sites affected the whole-lake estimate of mean total and loosely-bound P concentrations. For each study lake and P form, the data were randomly subset to between 2 and 9 sites. The mean P concentration for this sampling subset was then calculated and compared to the "true" mean of all 10 sites based on the root mean square error (RMSE). This analysis was repeated for 1000 iterations and RMSE values were averaged for each sampling subset and normalized to the mean for all 10 sites to express the estimated error a proportion of the "true" mean concentration.

All data are available in Albright et al. (2020). All statistical analyses were completed in R version 3.6.0 (R Core Team, 2019) using the tidyverse (Wickham et al., 2019), robCompositions (Filzmoser et al., 2018), vegan (Oksanen et al., 2019), lmerTest (Kuznetsova et al., 2017), infer (Bray et al., 2021),and sf packages (Pebesma, 2018).

3 Results

3.1 Inter-lake variation in profundal sediment P

The size and composition of the profundal sediment P pool varied considerably among the study lakes. Total sediment P concentrations, averaged across the depth intervals of profundal cores, ranged from 738.2 to 1,164.7 µg P g^{-1} dry sediment. The chemical speciation of the profundal sediment P pool also differed among the study lakes (Table 2). Redox-sensitive and organic P fractions were consistently the most abundant components of the total sediment P pool while aluminum and calcium-bound P were usually present in the lowest concentrations. We used PCA as part of a compositional analysis to explore patterns in profundal sediment P chemistry among our study lakes and to identify lakes with similar sediment P composition (Figure 1A). The first principal component (PC1) explained 45.27% of the variation in the dataset and was most closely associated with the prevalence of redox-sensitive and aluminum-bound P. The second principal component (PC2) explained 36.42% of the variation and was highly correlated with organic P content. Together, the first two principal components explained 81.69% of the variance in the dataset. Sediment P speciation defined groups of similar lakes (Table 2, Figure 1A). Center, Five Island, Storm, and Silver Lake formed the largest group, based on higher calcium-bound P concentrations. Silver Lake was slightly separated from this group due to higher concentrations of loosely-bound P. Abundant organic and loosely-bound P placed North and South Twin Lake in a group together. Swan Lake was distinguished from the other study lakes due to high redox-sensitive and aluminum-bound P concentrations.

The grouping of study lakes based on sediment P composition followed patterns in dominant watershed soil series (Table S2). North and South Twin Lake had very similar watershed soil composition, primarily characterized by Webster, Nicollet, Clarion, and Canisteo soil series. Additionally, Swan Lake, which was unique in sediment P speciation, had a different watershed soil composition, the majority of which were Marshall, Colo-Judson, and Exira series formed in loess, rather than glacial till. There was variation in dominant watershed soils within the Center, Five Island, Silver and Storm Lake group. Soils in the Storm Lake catchment were predominantly Sac, Primghar, Marcus, and Galva series formed in loess while the other watershed soils were composed of series formed in glacial till.

In addition to watershed soils, some landcover classes corresponded to the observed groups of study lakes based on profundal sediment P chemistry (Table S3, Figure 1B). Specifically, the proportions of grassland and forest cover in the catchment were strong predictors of sediment P composition, separating Swan Lake from the other waterbodies. Lake basin morphology and sediment characteristics were also strong predictors of lake groupings. The Center, Five Island, Storm, and Silver Lake group was associated with greater maximum lake depth and higher sediment bulk density. The North and South Twin Lake group was associated with higher sediment organic matter content and greater volume development, indicating bowl-shaped basins. The spread of lakes along PC1 seems to be driven by watershed characteristics while variation along PC2 was more closely associated with lake basin and sediment features.

In order to evaluate the ecosystem-level implications of the observed variation in profundal sediment P speciation among our study lakes, we tested the relationship between the relative availability of mobile P fractions in the sediment and algal biomass in each lake. We preformed a linear regression of the percent of total sediment P that is mobile (loosely-bound, total organic, and redox-sensitive) versus time-averaged chlorophyll-*a* concentrations. There was a significant positive relationship between the percentage of total P that is mobile and chlorophyll-*a* concentrations among the seven study lakes ($F_{1,5} = 7.584$, $p = 0.0401$, adjusted $R^2 = 0.52$, $\beta_1 = 3.953$ [0.263, 7.642]; Figure 2).

3.2 Intra-lake variation in sediment P

There was substantial spatial variation in total and loosely-bound P within individual study lakes. Loosely-bound P had greater spatial heterogeneity than total P within most lakes. There was a 1.2 to 2.2-fold within-lake difference in sediment total P and a 1.7 to 11.3-fold difference in loosely-bound P. Values for the coefficient of variation for loosely-bound P (range = 11.4 to 39.7%, mean = 25.7%) were also greater than the values for total P (range = 6.4 to 22.4%, mean = 14.0%; Table 2). South Twin was the only lake for which the coefficient of variation for loosely-bound P was less than the coefficient of variation for total P. The depth of the water column at the coring location explained 63.1% of the variation in sediment total P and 84.6% of the variation in loosely-bound P when lake was included as a random effect on the intercept (Figure 3). Including random effects on the model intercepts substantially improved the model fits ($p<0.0001$ for both models). For both the total sediment P and loosely-bound fractions, concentrations increased with depth. The increase in P with water depth was greater for total P ($\beta_1 = 65.28$ [27.57, 102.49]) than loosely-bound P ($\beta_1 = 12.79$ [7.18, 18.33]).

We further explored the influence of lake basin morphology on spatial variation in loosely-bound P through an analysis of volume development (D_v). The D_v score is a ratio of the volume of a lake basin to the volume of a perfect cone with a base area equal to the surface area of the lake and a height equal to the maximum water depth. The D_v ratio is used to characterize lake basin shape. Lakes with low D_v values have cone-shaped basins with localized deep holes while higher D_v ratios correspond to shallow, bowl-shaped bathymetries (Hutchinson, 1957). We found an inverse relationship between D_v and the variability in loosely-bound P, measured as the coefficient of variation (Figure S1). Five Island, Storm, and Swan Lakes had the greatest within-lake variation in loosely-bound P (CV range = 32.0 to 39.7%), and these waterbodies also had low D_v values (range = 0.65 to 1.36). Conversely, we found much less variability in loosely-bound P (CV range = 11.4 to 24.4%) in Center, Silver, North and South Twin Lakes, which all had higher D_v scores near 2 (range = 1.98 to 2.12).

To evaluate additional explanations for within-lake variation in sediment P, we surveyed spatial relationships between macrophyte colonization and sediment P in Swan Lake. Swan Lake had high variation in both loosely-bound and total P across the lakebed (Figure 4). There were extensive beds of sago pondweed (*Stuckenia pectinata*) within the 1-2 m depth contour across the northern half of Swan Lake and along the eastern shoreline. These beds were especially dense on the east side of the lake. There was also an isolated but dense bed of American lotus (*Nelumbo lutea*) in the center of the northern half of the lake rooted in 1.7 m of water. Both total and loosely-bound sediment P concentrations were lowest in the shallow, northern portion of the lake. Concentrations increased in the deeper, southern portion of the lake. There was no clear pattern between sediment P concentration and macrophyte bed density.

A better understanding of intra-lake variation in sediment P concentrations will inform appropriate spatial sampling resolutions to accurately quantify sediment P pools. We used a rarefaction analysis to test how the number of sampling sites affected the estimate of mean total and loosely-bound P in each study lake. For sediment total P, normalized RMSE values began to plateau when 6 or more sampling sites were included (Figure 5A). In subsets of 6 or more sampling sites, the normalized RMSE was less than 5 percent for all study lakes (range = 1.37 to 4.71%). Some study lakes had relatively low normalized RMSE values regardless of how many sites were included while others demonstrated sharp declines in normalized RMSE as the number of sites increased. These differences among lakes did not follow patterns with basin D_v or maximum water depth. Compared to total P, the number of sampling sites had a greater impact on the estimate of mean loosely-bound P within each study lake (Figure 5B). Normalized RMSE values did not reach obvious plateaus for any study lakes. Lakes with low D_v values (i.e., Five Island, Storm, and Swan Lakes) had higher normalized RMSE values and more drastic improvements in RMSE with increasing number of sample sites. Conversely, lakes with higher D_v values (i.e., Center, Silver, North and South Twin Lakes) had lower normalized RMSE and more gradual improvements in RMSE with more sampling sites.

In addition to quantifying the effects of spatial sampling resolution on P pool estimates, we also tested the influence of sample site location, specifically whether P concentrations at the deep site were representative of mean P concentrations across the lakebed. We compared total and loosely-bound P concentrations from the deep site of each lake to the whole-lake mean (Table S4). For sediment total P, concentrations at the deep site exceeded the whole-lake average for all lakes except Swan Lake. Normalized RMSE values ranged from 0.3 to 11.3 percent (mean = 4.6 %). Loosely-bound P concentrations at the deep site were consistently greater than whole-lake means. Normalized RMSE values were much greater than for total P (range = 2.2 to 52.5%, mean = 26.2%).

4 Conclusions

4.1 Inter-lake variation in sediment P content and composition

We quantified spatial heterogeneity in sediment P content and composition in shallow lakes and found high variation both within individual waterbodies and among different systems. Whole-lake means of sediment total P content ranged from 897.0 to 1,184.9 µg P g^{-1} dry sediment across the study lakes, with a grand mean of 946.2 µg P g^{-1} dry sediment. These concentrations align with observed ranges in other shallow, eutrophic lakes (Søndergaard et al., 2013) as well as other productive waterbodies (Doig et al., 2017; Kowalczewska-Madura et al., 2019b; Tao & Lu, 2020). Overall, sediment total P concentrations from our study lakes tend to be either similar to or lower than average values reported for other productive waterbodies (Table S5). The chemical composition of the profundal sediment P pool also varied among our study lakes; however, redox-sensitive and organic P were consistently the most abundant components. Other eutrophic waterbodies also have high levels of redox-sensitive P (Song &

Burgin, 2017; Randall et al., 2019; Papera et al., 2021) and organic P forms (Nowlin et al., 2005; Frost et al., 2019). More generally, aquatic sediments tend to be enriched in redox-sensitive, organic, loosely-bound, and aluminum-bound P because these forms are commonly associated with fine sediments, which are more easily eroded and transported from the source material (i.e., terrestrial soils; Stone & English, 1992; Kerr et al., 2011).

In order to compare sediment P composition among our study lakes, we used PCA as part of a compositional analysis to explore patterns and identify lakes with similar sediment P chemistry. The analysis grouped Center, Five Island, Storm, and Silver Lake together based on higher calcium-bound P levels. Abundant organic and loosely-bound P grouped North and South Twin Lake together while high redox-sensitive and aluminum-bound P concentrations placed Swan Lake apart from the other study lakes. The North and South Twin Lake and Swan Lake groupings reflect patterns in dominant watershed soil series. However, there was variation in watershed soil characteristics within the Center, Five Island, Silver and Storm Lake group as soils in the Storm Lake catchment were formed in loess while the other watershed soils were formed in glacial till. Variation in watershed soils within this group reinforces that aquatic sediment P composition is not a direct reflection of the bulk soil matrix due to selective erosional and sediment transport processes as well as *in situ* processing (Kerr et al., 2011).

The dominant mechanisms that drive internal P loading in a given waterbody depend on the chemical composition of the sediment P pool (Orihel et al., 2017). Our finding that sediment P composition varies among different lakes implies that the processes driving sediment P release also vary across systems. For example, redox-sensitive P was the dominant fraction in Swan Lake while North and South Twin Lake a higher prevalence of organic P. Based on these differences in sediment P composition, we would expect sediment P release in these lakes to respond differently to dissolved oxygen levels at the sediment-water interface. Specifically, we would anticipate that anoxia would prompt sediment P release in Swan Lake due to the dissolution of redox-sensitive minerals under reducing conditions (Mortimer, 1941; Orihel et al., 2015). Anaerobic sediment P release from redox-sensitive P minerals could also occur in North and South Twin Lake; however, due to high organic P levels, we would also expect to see sediment P release under oxic conditions due to decomposition of sediment organic matter and subsequent P mineralization (Joshi et al., 2015; Song & Burgin, 2017; Frost et al., 2019). Hypolimnetic aeration, a management approach to limit anaerobic internal P loading, might protect redox-sensitive minerals and reduce sediment P release in Swan Lake, but the same approach in North and South Twin Lakes could stimulate decomposition and enhance aerobic P release (Horppila et al., 2017, Tammeorg et al., 2017). Inter-lake variation in sediment P composition means that management strategies to prevent and control internal P loading are unlikely to translate well across lakes with different sediment P chemistry.

Across the study lakes we found a strong relationship between the average contribution of mobile P fractions to the total sediment P pool and long-term, average chlorophyll-*a* concentrations, as an index of algal biomass. This relationship indicates the substantial contribution of internally-loaded P to algal production in these shallow lakes. Total organic P was the largest fraction of total sediment P in all of the study lakes, driving the among-lake variability in mobile P contributions. Given the prevalence of organic P in the sediments, it is likely that decomposition of sediment organic matter and subsequent P mineralization are key processes for internal P loading in the study lakes (Joshi et al., 2015; Frost et al., 2019). In hypereutrophic waterbodies rampant algal production and detritus enhance organic matter sedimentation, increasing organic P in lakebed sediments. Large pools of organic P are associated with high extracellular enzyme activities, which mobilize and release sediment P (Song & Burgin, 2017). Our study lakes are likely examples of systems in which biologically-mediated cycling of organic P pools drives internal P loading.

4.2 Intra-lake variation in sediment P pools

In addition to variation in sediment P content and composition across different lakes, we also identified spatial heterogeneity in total and loosely-bound P pools within individual lakes. Consistent with our expectations, loosely-bound P concentrations were most variable in lakes with low volume development indices, indicative of conical basins with localized deep holes. This bathymetric variability produces both shallow regions where frequent sediment disturbance releases loosely-bound P and deeper areas where loosely-bound P is protected (James, 2017a). As a result, loosely-bound P concentrations are more variable in lake basins with more complex bathymetry. In contrast, lakes with high volume development scores, associated with shallow, bowl-shaped basins, had more consistent loosely-bound P concentrations, as sediment disturbance was likely more uniform across the lakebed. We also hypothesized that greater water depths would correlate with higher concentrations of total and loosely-bound sediment P across the lakebed, which was supported by the mixed model analysis. In fact, the depth of the water at the sediment sampling location described the vast majority (84.1%) of the variation in loosely-bound P and over half of the variation (63.1%) in total P concentrations. The pattern of higher concentrations at greater depths is likely driven by sediment focusing and greater net sediment deposition in deeper portions of the lake (White & Stone, 1996; Hou et al., 2014; Cardoso-Silva et al., 2018). The direct relationship that we observed between water depth and sediment P concentrations is consistent with other studies of spatial variation in sediment P pools (Nowlin et al., 2005; Kowalczewska-Madura et al., 2019b). However, water depth did not fully explain the observed variation in sediment total and loosely-bound P, indicating that factors beyond basin morphology contribute to spatial variation in sediment P pools.

A factor that may influence the spatial distribution of the sediment P pool is the location and density of macrophyte beds. In most of the study lakes, macrophytes were limited to emergent taxa; therefore, the macrophyte beds did not overlap with sediment sampling sites. However, extensive submersed and floating-leaf vegetation across Swan Lake allowed us to sample sediment P across a gradient of macrophyte bed density. We found no clear evidence of a spatial pattern between macrophyte bed density and sediment P. Total and loosely-bound P concentrations were lowest in the shallow, heavily-vegetated northern portion of the lake. Sediment P concentrations were generally higher in the deeper, non-vegetated areas of the lake. However, there were two vegetated sites along the eastern edge of the lake that also had high sediment total and loosely-bound P concentrations. Thus, it is difficult to establish a spatial relationship between macrophyte beds and sediment P. Macrophytes could be expected to either increase sediment P stocks by stabilizing sediments and reducing resuspension (Carpenter et al., 1983; Madsen, 2001) or decrease sediment P pools via biological uptake (Xing et al., 2018). Exploring the balance of this complex relationship is an avenue for future research. Specifically, quantifying sediment resuspension could better explain the spatial distribution of P stocks in Swan Lake (James, 2017a). Sediment resuspension is expected to interact with both water depth and macrophytes as shallow, non-vegetated sediments are the most susceptible to disturbance (Horppila & Nurminen, 2001; Nurminen & Horppila, 2009). Quantifying sediment disturbance on Swan Lake could further illuminate interactions between water depth and macrophyte bed density and the resultant effects on sediment P pools.

Our study provides a valuable characterization of spatial heterogeneity in sediment P pools. Although sediment P content and chemical composition are expected to vary over time as a balance of sedimentation, mobilization and release into the water column, and post depositional transformations (Ostrofsky, 2012; Heathcote et al., 2013;), other studies of natural, productive lakes have found limited temporal variation in sediment P concentrations on intra-annual scales (Kowalczewka-Madura et al., 2019a; Kowalczewka-Madura et al., 2019b). As such, our results provide a fair snapshot of expected spatial variation in sediment P pools within shallow lakes. A natural progression of this research is to explore temporal variation in sediment P content and composition.

4.3 Applications and significance

A quantitative understanding of intra-lake heterogeneity in sediment P concentrations reveals the spatial sampling resolutions necessary for accurate inventories of sediment P pools. Our rarefaction analysis suggests that sampling six or more locations across the lakebed may be sufficient to estimate the mean, whole-lake concentration of sediment total P. When six or more sites were included, estimate error (normalized RMSE) dropped below five percent for all study lakes and remained stable even as more sampling sites were included. This result aligns with research on lacustrine burial rates that proposes as few as five sediment cores can be used to accurately assess sediment P accumulation rates (Rippey et al., 2008; Engstrom & Rose, 2013). Compared to the rarefaction analysis for total P, estimate errors for mean loosely-bound P concentrations were greater for most study lakes, and there were no clear plateaus in error values as more sampling sites were considered. However, the relationship between sampling sites and estimate error did follow a pattern with basin morphology. Lakes with more complex bathymetry (low D_v; Five Island, Storm, Swan Lake) had more drastic improvements in RMSE values as more sampling sites were considered. Lakes with simple, bowl-shaped basins (high D_v; Silver, Center, South Twin, North Twin Lake) had lower RMSE values overall and more gradual improvements with increased sampling resolution. This pattern makes sense in light of our finding that lakes with more complex basin morphology had greater spatial variation in loosely-bound P concentrations and suggests that more sampling sites are needed to inventory loosely-bound P stocks compared to total P, especially in lakes with complex bathymetry.

Comparing P concentrations at the deep site of each study lake to whole-lake means demonstrates that scaling values from the deep site to the entire lakebed will tend to overestimate sediment P stocks. We found that total P concentrations at the deep site were greater than the whole-lake average for all lakes except for Swan Lake, where the values were very similar, and that extrapolating values from the deep site could overestimate the whole-lake mean by as much as 11.3 percent. Loosely-bound P concentrations at the deep site were consistently greater than the whole-lake mean and extrapolating from the deep site overestimated loosely-bound P stocks by 2.2 to 52.5 percent (mean 26.2%). Our findings support other studies suggesting that single-core analyses produce unreliable estimates of whole lake carbon and nutrient stocks (Mackay et al., 2012; Lin et al., 2022). Our analysis further reinforces that a good approach for addressing this bias and obtaining an accurate estimate of whole-lake sediment P stocks is to collect multiple sediment cores across a range of water depths (Engstrom & Rose, 2013). Accurate inventories of sediment P stocks are necessary to parameterize models of lentic P cycling (Hansen et al., 2020). Lake response to simulated changes in external P loading will be sensitive to the mass of P held in the sediments that is available for recycling. As such, accurate estimates of sediment P stocks, accounting for spatial variation, are critical for modeling lentic P cycles and forecasting lake response to changes in watershed nutrient loading.

A quantitative understanding of inter- and intra-lake variation in sediment P content and composition is essential for accurately sampling, scaling, and modeling sediment P pools, yet this variability has been largely undescribed for shallow lakes. Our study builds on our understanding of regional patterns in lacustrine sediment P speciation and contributes novel perspectives on the causes and consequences of spatial heterogeneity in sediment P pools within shallow lakes. Our finding of inter-lake variation in sediment P composition reinforces that management strategies to control internal P loading will also differ among lakes and that effective solutions will consider site-specific features, including sediment P chemistry. Our analysis further supports that shallow lakes are spatially heterogenous ecosystems, and accounting for this variation is necessary to accurately characterize sediment P pools. The large within-lake variability in loosely-bound and total P documented in this study reinforces the importance of sampling multiple sites across the lakebed when possible and using caution when extrapolating measures from the deep site to the whole system. Accurate sediment P inventories will allow for more empirical parameterization of sediment-water interactions in models of lake P cycling. Our quantification of spatial heterogeneity in sediment P pools within and among lakes is a critical step to being able to predict internal P loading potential based on lake and watershed characteristics.

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Data Availability Statement: The data supporting the conclusions are publicly available in Albright et al. (2020), with a CC0 1.0 Universal Public Domain Dedication license. The analysis code is available in the Github repository https://github.com/AlbrightE/Sediment_P_2018, which will be archived using Zenodo upon acceptance of the manuscript.

Tables

Table 1. *Location, Basin Morphology, and Watershed Features of the Study Lakes*

	Center Lake	Five Island	Silver Lake	Storm Lake	North Twin	South Twin	Swan Lake
Latitude	43.41263	43.1545	43.44145	42.61977	42.47563	42.45847	42.03568
Longitude	-95.1357	-94.648	-95.3353	-95.1857	-94.6405	-94.6536	-94.8416
Surface area (ha)	104	405.6	431.4	1270.7	187.4	224.6	48.6
Maximum depth (m)	5.5	8	3	6.2	3.7	1.6	3.8
Mean depth (m)	3.7	1.7	2	2.6	2.7	1.1	1.7
Catchment/surface area	2.7	9.2	10.7	4.4	4.6	6	6.1
Chlorophyll-*a* (µg L^{-1})	36.64	45.42	28.38	15.94	51.2	87.2	75.71
% Cropland	14.3	75.5	78.1	60.2	82.5	78.4	45.1
Dominant soil origin	Glacial till	Glacial till	Glacial till	Loess	Glacial till	Glacial till	Loess

Note. Chlorophyll-*a* values are the 20-year mean (2000-2019) of measurements made during the ice-free season (n=55-57 measurements per lake) with the exception of South Twin Lake which is the 14-year mean (2006-2019) of measurements made during the ice-free season (n=55)

Table 2. *Variation in Profundal Sediment P Among Lakes and Within-Lake Heterogeneity*

	Center Lake	Five Island	Silver Lake	Storm Lake	North Twin	South Twin	Swan Lake
Total P (µg P g^{-1} dry sediment)	938.3 (±191.4)	1,013.5 (±160.9)	936.5 (±97.0)	738.2 (±29.5)	1,134.1 (±288.3)	939.1 (±113.1)	1,164.7 (±306.2)
% Redox-sensitive	24.45	30.30	22.81	31.16	9.62	11.81	35.58
% Labile organic	31.45	25.02	21.79	19.06	--	--	--
% Refractory organic	21.85	15.98	17.53	22.35	--	--	--
% Total Organic	53.31	41.00	39.32	41.41	74.81	70.48	50.14
% Loosely-bound	7.75	15.04	21.79	9.88	10.74	12.09	3.16
% Al-bound	7.02	8.21	7.50	6.91	2.79	1.96	9.72
% Ca-bound	7.47	5.44	8.58	10.64	2.04	3.64	1.40
CV Total P (%)	10.2	19.0	6.4	17.5	7.3	22.4	15.2
CV Loosely-bound (%)	17.5	32.0	17.7	37.3	24.4	11.4	39.7

Note. Total P values are the mean (± standard deviation) for all intervals of the deep site core. The precent contribution of each P species to the total sediment P pool is an average value across the deep site core profile. The coefficient of variation for total and loosely-bound P was calculated for all ten sediment cores collected across each study lake.

Figures

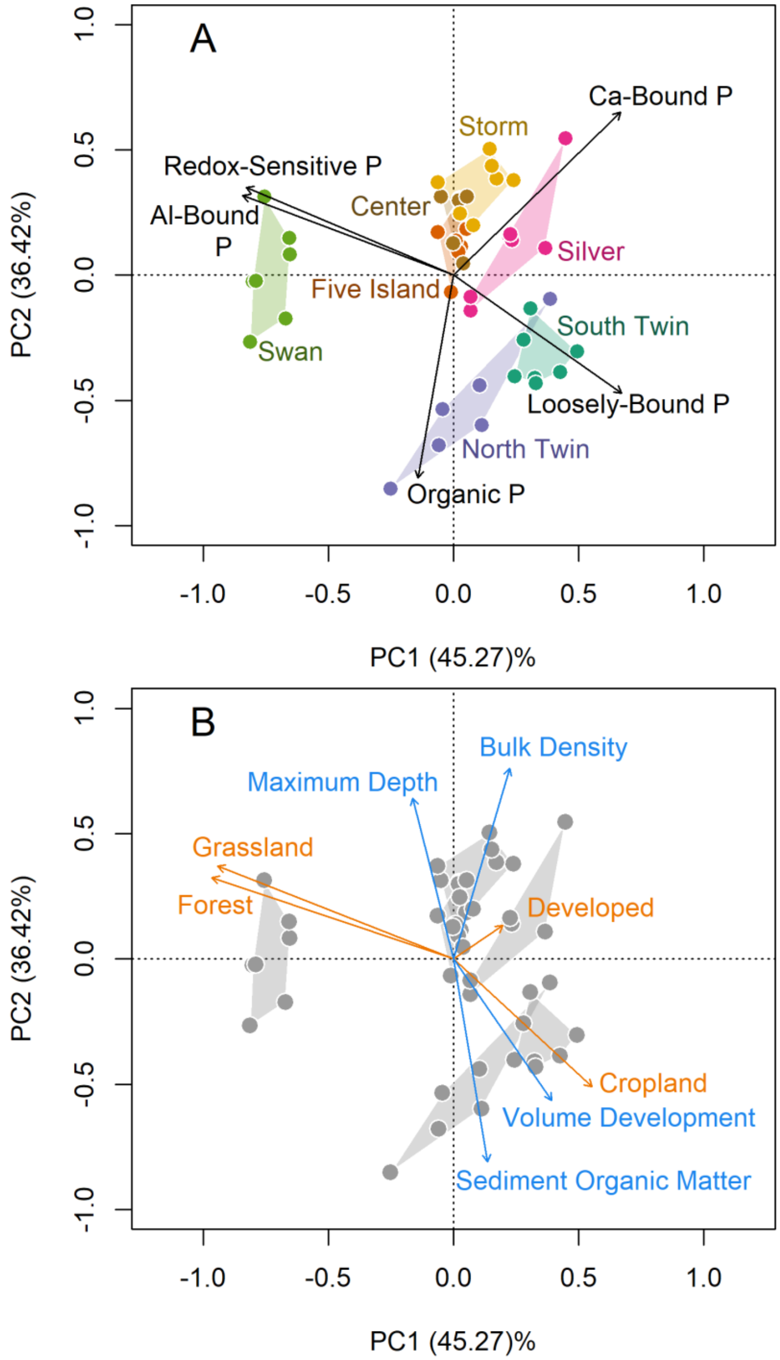

Figure 1. (A) PCA biplot based on a compositional data analysis of profundal sediment P pools. The compositional analysis was defined by the concentrations of loosely-bound (porewater and surface sorbed), redox-sensitive (Fe- and Mn-bound), aluminum-bound, calcium-bound, and organic P (including labile and refractory components). Arrows represent center log-ratio transformed concentrations of the P fractions. **(B)** Interpretation of PCA biplot with key environmental variables. Watershed land cover classes are shown in orange while lake and sediment characteristics are plotted in blue.

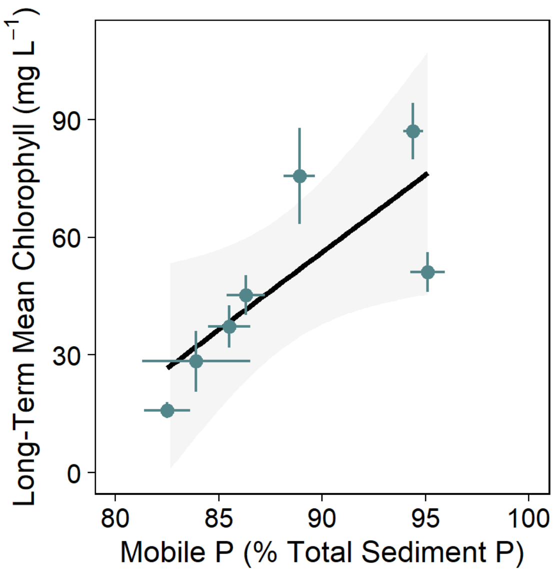

Figure 2. The relationship between the fraction of the total sediment P pool that is comprised of mobile species and the long-term average chlorophyll-*a* concentrations in each study lake ($F_{1,5}$ = 7.584, p = 0.0401, adjusted R^2 = 0.52, β_1 = 3.953 [0.263, 7.642]).

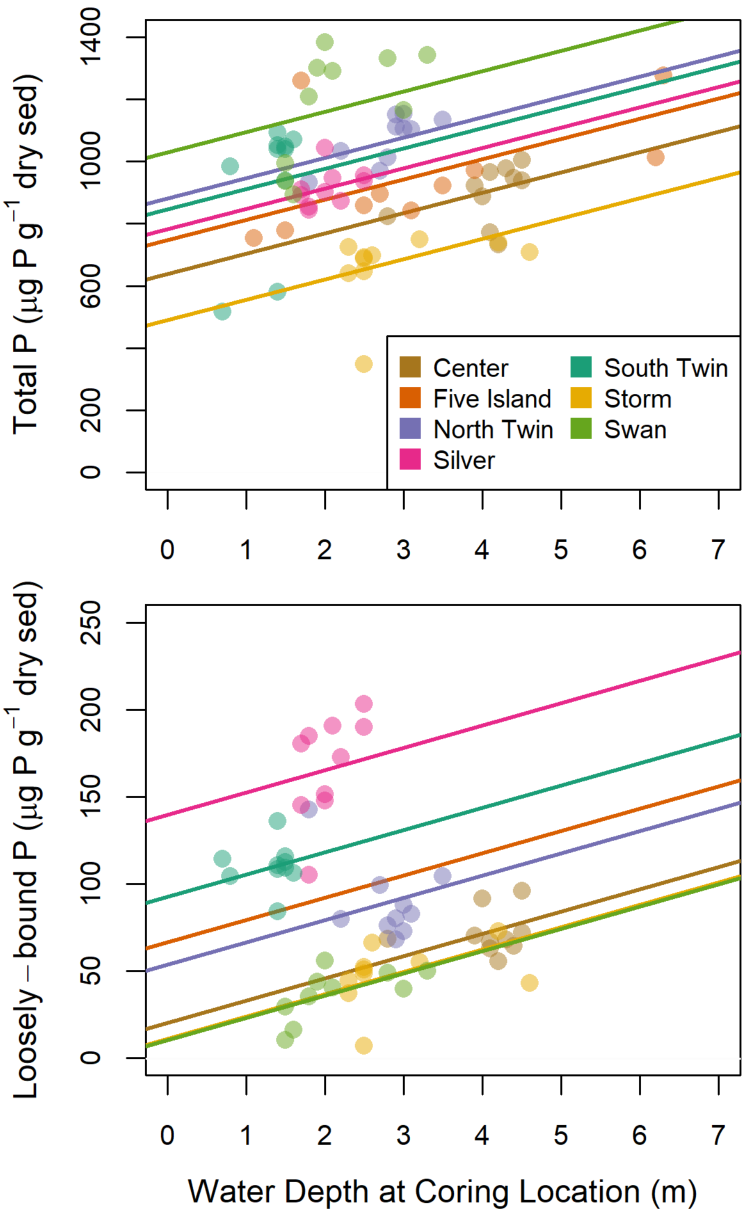

Figure 3. Mixed model regression effects of water depth at the coring location on total sediment P (β_{1}=65.28 [27.5661, 102.4886]; top panel) and loosely-bound sediment P (β_{1}=12.787 [7.180, 18.334]; bottom panel) by lake.

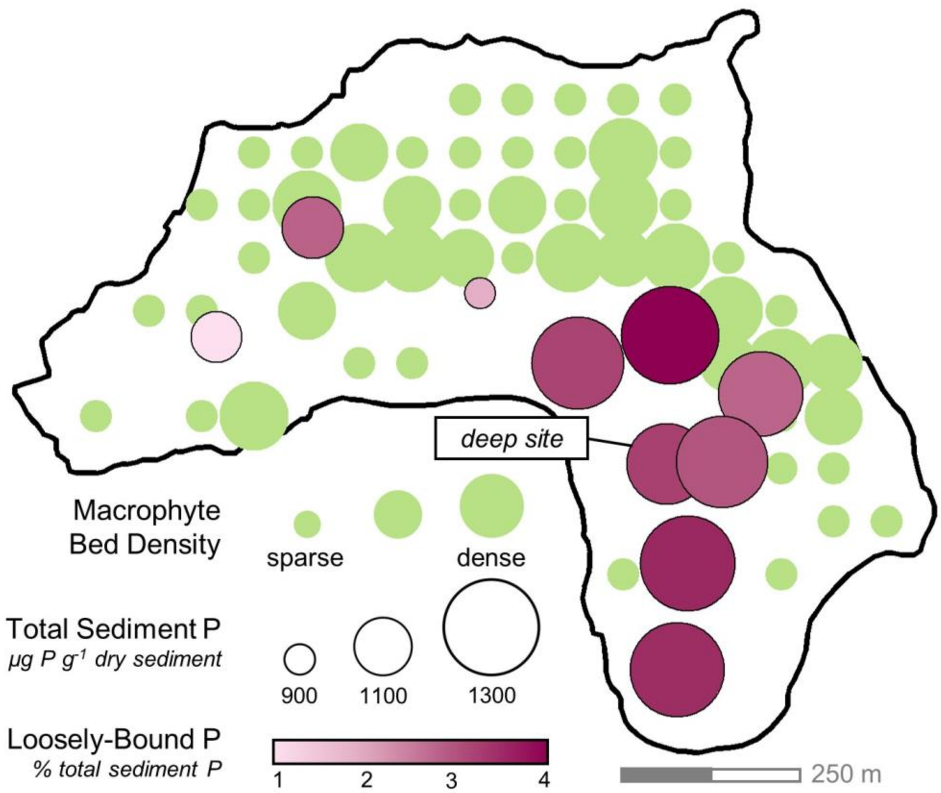

Figure 4. Within-lake variation in sediment loosely-bound and total P in Swan Lake. The ten sediment cores collected across Swan Lake are plotted with pink circles. The point size corresponds to total sediment P concentration which the shade of pink represents the relative proportion of loosely-bound P at that site. The qualitative estimate (1-3) of macrophyte bed density is shown in green circles. Blank areas of the map indicated bare sediment while areas with larger green dots represent very dense beds of *Stuckenia pectinata* and *Nelumbo lutea.*

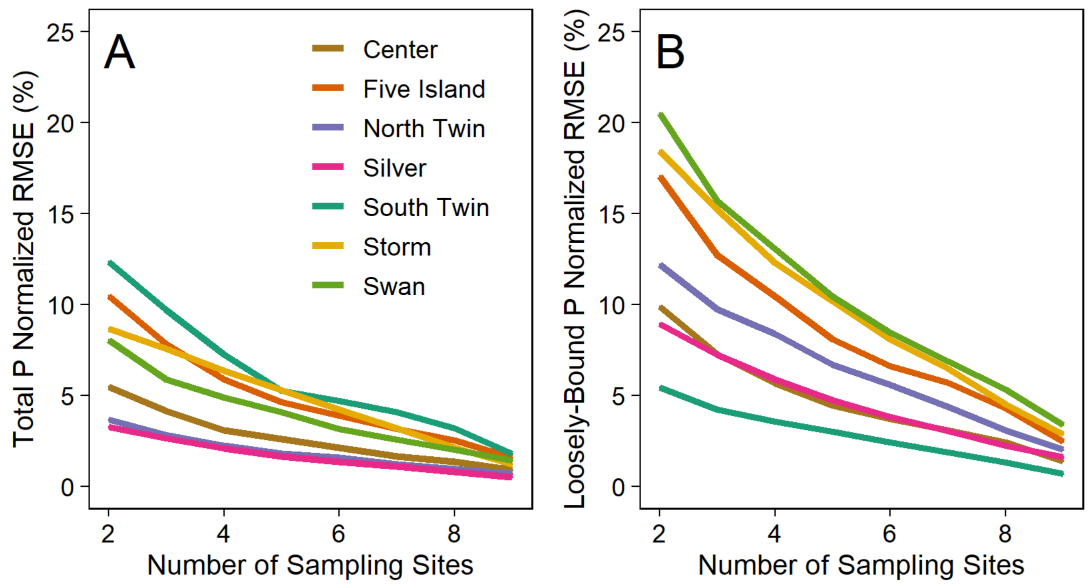

Figure 5. Rarefaction analysis of mean total **(A)** and loosely-bound **(B)** P concentrations based on the number of sampling sites. RMSE values are normalized as a percent of the mean P concentration when all 10 sampling sites are included.

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Supporting Information for

High Inter- and Intra-lake Variation in Sediment Phosphorus Pools in Shallow Lakes

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Contents of this file

Introduction

The supporting information contains detailed explanations of the equations used to determine the sediment physical characteristics and the concentrations of sediment phosphorus (P) fractions (Text S1). Table S1 details the frequency of thermal stratification in the study lakes as well as dissolved oxygen conditions at the sediment-water interface. Tables S2 and S3 contain more detailed information on watershed soil series and landcover respectively. Table S4 compares the total and loosely-bound P concentrations at the deep site of each study lake to the whole-lake mean values. Table S5 summarizes how sediment total P concentrations in our study lakes compare to other values in the literature. We have also included a visualization of the relationship between the coefficient of variation in loosely-bound P across the lakebed and lake basin volume development for the study lakes (Figure S1).

Text S1.

Eq. 1 Moisture Content (MC)

$$\text{Moisture Content (\%)} = \left[\frac{(W_w - W_t) - (W_d - W_t)}{W_w - W_t}\right] \times 100 \quad (1)$$

Where W_t is the weight of the aluminum weigh boat, W_w is the weight of the weigh boat and fresh sediment sample, and W_d is the weight of the weigh boat and dry sediment.

Eq. 2 – Organic Matter Content as Loss-on-Ignition (LOI)

$$\text{LOI Organic Matter Content (\%)} = \left[\frac{(W_d - W_t) - (W_a - W_t)}{W_d - W_t}\right] \times 100 \quad (2)$$

Where W_a is the weight of the weigh boat and the ashed sediment after combustion.

Eq. 3 – Bulk Density

$$\text{Bulk Density } (g/cm^3) = \frac{260}{100 + 1.6 \times \left[MC + \left(\frac{LOI}{100 \times (100\text{-}MC)}\right)\right]} \quad (3)$$

Eq. 4 – Dry Mass Equivalent of Fresh Sediment Used

$$\text{Dry Mass Equivalent (g)} = \text{Mass Fresh Sediment (g)} \times (100 - MC) \quad (4)$$

Eq. 5 – Loosely sorbed and pore water P

$$\text{Loosely-Bound P (mg P/g dry sediment)} = \frac{\text{Concentration SRP (mg/L)} \times \text{Extractant Volume (L)}}{\text{Dry Mass Equivalent of Sediment Used (g)}} \quad (5)$$

The concentration of SRP used should reflect the average of lab duplicate measures. The extractant volume should equal the total volume of 1M NH_4Cl used across both extractions, (0.05 L). The dry mass equivalent of the fresh sediment used is estimated based on MC (Eq. 4) and will be the same for the calculations of each subsequent extraction (Eq. 6-9).

Eq. 6 – Redox-sensitive P (Fe- and Mn-bound)

$$\text{Redox-Sensitive P (mg P/g dry sediment)} = \frac{\text{Concentration SRP (mg/L)} \times \text{Extractant Volume (L)}}{\text{Dry Mass Equivalent of Sediment Used (g)}} \quad (6)$$

The concentration of SRP used should reflect the average of lab duplicate measures. The extractant volume should equal the volume of 0.11 M bicarbonate – 0.1 M sodium dithionate solution used (0.0125 L).

Eq. 7 – Aluminum-Bound P

$$\text{Al-Bound P (mg P/g dry sed.)} = \frac{\frac{\left[\text{Concentration SRP}\left(\frac{mg}{L}\right) \times \frac{\text{(Post pH (g)-Tare (g))}}{\text{(Pre pH (g)-Tare (g))}}\right]}{1.00152} \times \text{Extractant Volume (L)}}{\text{Dry Mass Equivalent of Sediment Used (g)}} \quad (7)$$

The concentration of SRP used should reflect the average of lab duplicate measures and must be corrected for the pH adjustment. Tare is the mass of the I-chem jar in which the adjustment is performed; Pre pH is the mass of the jar and the extraction supernatant; and Post pH is the mass of the jar, supernatant, and titrant used for the pH adjustment. The constant 1.00152 is used for 0.1 M HCl as the titrant. The corrected SRP concentration can then be corrected for the extractant volume (0.025 L) and sediment mass.

Eq. 8 – Labile Organic P

$$\text{Labile Organic P (mg P/g dry sed.)} = \left[\frac{\text{Concentration SRP, digested (mg/L)} \times \text{Extractant Volume (L)}}{\text{Dry Mass Equivalent of Sediment Used (g)}}\right] - \text{Al-Bound P} \quad (8)$$

The concentration of SRP from the *digested* supernatant should first be corrected for the volume of NaOH used (0.05 L) and the sediment mass. This value represents the concentration of aluminum-bound and labile organic P in the sediment pellet. The concentration of labile organic P is calculated as the difference between this value and the Al-bound P concentration (Eq. 7).

Eq. 9 – Calcium-Bound P

$$\text{Ca-Bound P (mg P/g dry sed.)} = \frac{\frac{\left[\text{Concentration SRP}\left(\frac{\text{mg}}{\text{L}}\right) \times \frac{(\text{Post pH (g)-Tare (g)})}{(\text{Pre pH (g)-Tare (g)})}\right]}{1.00452} \times \text{Extractant Volume (L)}}{\text{Dry Mass Equivalent of Sediment Used (g)}} \quad (9)$$

The concentration of SRP used should reflect the average of lab duplicate measures and must be corrected for the pH adjustment. The constant 1.00452 is used for 0.1 M NaOH as the titrant. The corrected SRP concentration can then be corrected for the extractant volume (0.025 L) and sediment mass.

Eq. 10 – Total P

$$\text{Total P (mg P/g dry sediment)} = \frac{\frac{\left[\text{Concentration TP}\left(\frac{\text{mg}}{\text{L}}\right) \times \frac{(\text{Post pH (g)-Tare (g)})}{(\text{Pre pH (g)-Tare (g)})}\right]}{1.00452} \times \text{Dilution Volume (L)}}{\text{Dry Mass of Sediment Used (g)}} \quad (10)$$

The concentration of TP used should reflect the average of lab duplicate measures and must be corrected for the pH adjustment. The constant 1.00452 is used for the 0.1 M NaOH as the titrant. This corrected SRP concentration can then be corrected for the volume to which the sample was diluted after boiling (0.05 L) and the mass of dry sediment used.

Table S1. Long-term stratification patterns and dissolved oxygen conditions

	Frequency of Summer Thermal Stratification (%)		Dissolved Oxygen at Sediment-Water Interface (mg L^{-1})
Lake Name	Long-Term Record*	2018 Field Season†	2018 Summer Mean‡
Center	26.2	28.9	4.81
Five Island	26.8	18.9	1.28
North Twin	9.3	21.4	7.26
Silver	8.1	26.7	8.20
South Twin	NA	0.8	9.18
Storm	2.5	NA	4.72
Swan	17.4	30.8	2.87

* Obtained from the Iowa Department of Natural Resources AQuIA data repository. Records are from 2006-2018 for Storm and Silver Lakes and from 2005-2018 for the remaining study lakes. The frequency of stratification was determined as the percent of observations where a thermocline was present at the deep site of the lake out of the total observations in the long-term monitoring record. Lakes were sampled three times between May and October each year.

† Determined from high-frequency water temperature loggers deployed at regular depth intervals at the deep site of each study lake from around May to August of 2018. The frequency of stratification was determined as the number of days when a thermocline was present as a percent of the total days the sensors were deployed. Sensors on Storm Lake were lost during a Storm and never recovered.

‡ Data were obtained from the Iowa Department of Natural Resources Ambient Lake Monitoring Program's profile data. Dissolved oxygen concentrations from early and mid-summer sampling events were averaged to estimate conditions at the time of our sampling. South Twin is not monitored in this program, so dissolved oxygen values were obtained from a surface dissolved oxygen logger and assumed to represent conditions at the sediment-water interface since the lake was well-mixed at this time.

Table S2. Dominant watershed soil texture and series

Lake name	Dominant watershed soil texture	Dominant watershed soil series
Center	Loam, clay loam, silty clay loam	Webster, Nicollet, Clarion, Canisteo, Okabena, Waldorf
Five Island	Loam, clay loam, silty clay loam	Webster, Nicollet, Clarion, Canisteo, Okoboji
North Twin	Loam, clay loam,	Webster, Nicollet, Clarion, Canisteo
Silver	Loam, clay loam, silty clay loam	Webster, Nicollet, Clarion, Canisteo, Okabena, Waldorf
South Twin	Loam, clay loam,	Webster, Nicollet, Clarion, Canisteo
Storm	Silty clay loam	Sac, Primghar, Marcus, Galva
Swan	Silty clay loam	Marshall, Colo-Judson, Exira

Table S3. Watershed land cover

Lake name	Cropland (%)	Grassland (%)	Water (%)	Forest (%)	Urban (%)
Center	14.3	26.6	34.3	7.5	17.2
Five Island	75.5	9.6	12.0	1.8	1.7
North Twin	82.5	11.9	19.8	0.5	2.2
Silver	78.1	12.8	7.5	0.8	0.8
South Twin	78.4	2.2	30.7	0.1	10.4
Storm	60.2	12.6	18.5	1.4	7.2
Swan	45.1	28.8	13.8	8.5	3.9

Table S4. RMSE of deep site P concentrations

	Total P (µg P g dw^{-1})				Loosely-Bound P (µg P g dw-1)			
Lake	Whole-Lake Mean	Deep Site	RMSE	Normalized RMSE (%)	Whole-Lake Mean	Deep Site	RMSE	Normalized RMSE (%)
Center	897.0	938.3	41.3	4.6	71.6	100.6	29.0	40.5
Five Island	956.7	1013.5	56.8	5.9	108.3	156.9	48.6	44.9
Silver	915.6	936.5	20.9	2.3	167.2	203.3	36.1	21.6
Storm	663.1	738.2	75.1	11.3	47.7	72.8	25.1	52.5
North Twin	1070.4	1134.1	63.7	5.9	89.4	101.7	12.3	13.8
South Twin	936.0	939.1	3.1	0.3	110.2	112.6	2.4	2.2
Swan	1184.9	1164.7	20.2	1.7	37.0	39.9	2.9	7.7
		AVERAGE	40.1	4.6		AVERAGE	22.3	26.2

Table S5. Literature comparison of sediment total P concentrations

			Whole-lake Total P across study lakes (μg P g-1 dw)	
Reference	Waterbodies	Location	Range	Grand Mean
This Study	7 shallow lakes	Iowa, USA	897.0 - 1,184.9	946.2
Søndergaard et al., 2013	6 shallow lakes	Denmark	740 - 4,100	2,397
Doig et al., 2017	9 eutrophic lakes reservoirs	Canadian Prairie Provinces	533 - 2,310	1,594
Tao & Lu, 2020	83 lakes and reservoirs	Yangtze, Huaihe River catchments, Eastern China	360 - 2,180	820
Kowalczewska-Madura et al., 2019b	2 meso-eutrophic lakes	Poland	840 - 1,300	1,072

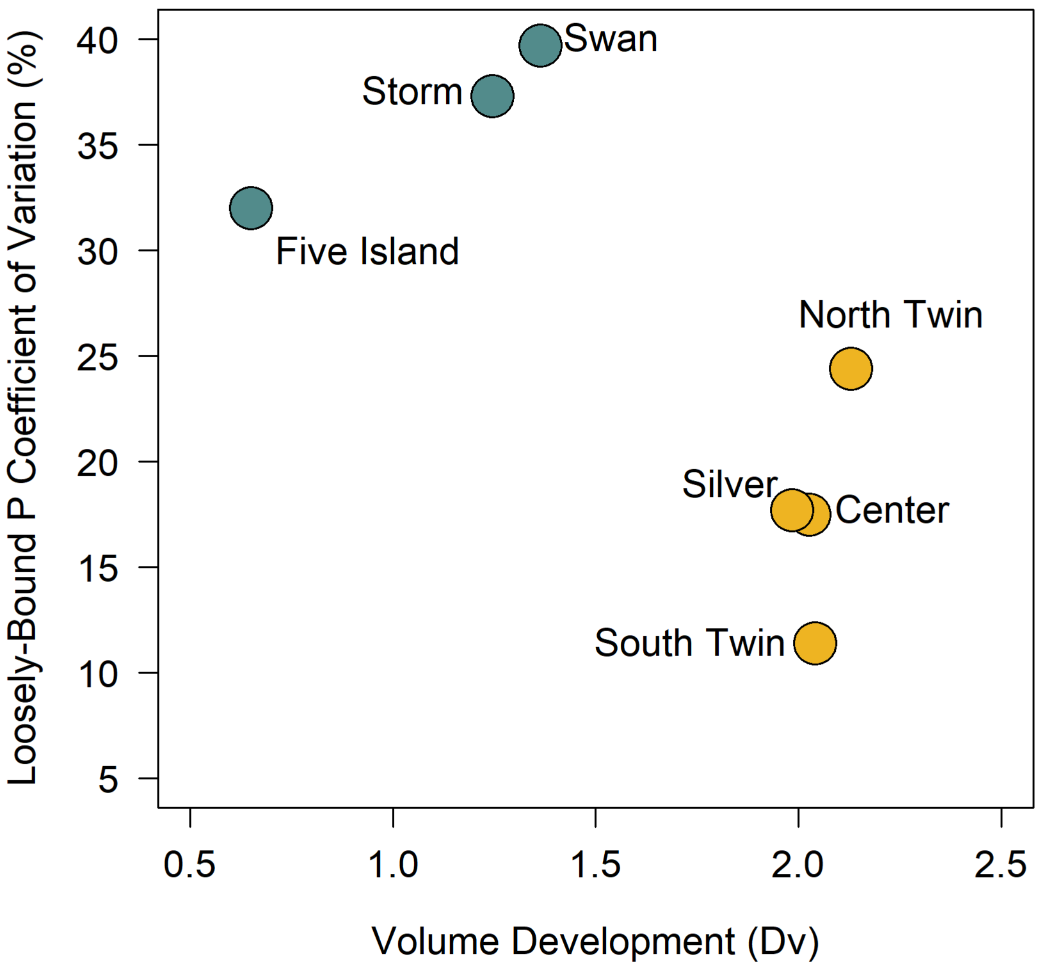

Figure S1. Variation in sediment loosely-bound P across lake basin volume development. The coefficient of variation for loosely-bound P was highest in lakes with low volume development scores (blue points), indicating more variable basin shape. Lakes with higher volume development ratios (yellow points) had less intra-lake variation in loosely-bound P.