

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

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13 contact the coresponding author.
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15 **Key Points:**

- 16
- The size and chemical composition of sediment phosphorus pools are spatially
17 heterogeneous both within and among shallow lakes
 - Lake basin morphology, water depth, and watershed characteristics drive variation in
18 sediment P pools within and among lakes
 - Understanding intra-lake variation in sediment P informs the spatial sampling resolutions
19 necessary for accurate P stock assessments
20
21

22 **Abstract**

23 Phosphorus (P) release from lakebed sediments may fuel phytoplankton blooms, especially in
24 shallow waterbodies. A primary mechanism that controls internal P loading is the size and
25 chemical composition of the sediment P pool. However, variation in sediment P within and
26 among shallow lakes remains poorly quantified. We measured the degree of spatial heterogeneity
27 in the size and composition of sediment P pools, both within and among seven shallow lakes.
28 There was a 1.6x difference in total sediment P among the study lakes, and P composition varied
29 across lakes based on differences in watershed soils and land cover and lake basin morphology.
30 Differences in sediment P composition among lakes imply that the dominant mechanisms of
31 internal loading will also vary among lakes. We also found that higher mobile P (as a fraction of
32 total P) in the profundal sediments was positively correlated with long-term mean chlorophyll-*a*
33 concentrations ($p=0.04$), indicating the reciprocal relationship between sediment P composition
34 and phytoplankton biomass in shallow lakes. Additionally, we measured substantial within-lake
35 heterogeneity in total and loosely-bound sediment P within each lake. Concentrations were
36 positively correlated with water depth such that extrapolating measurements from the deep site
37 alone could overestimate whole-lake mean P concentrations, reinforcing that single station
38 methods produce unreliable estimates of mean sediment P stocks. Our results provide insight into
39 the magnitude and pattern of inter- and intra-lake variation in sediment P pools that should be
40 accounted for when sampling, scaling measurements, and modeling sediment P dynamics.

41

42 **Plain Language Summary**

43 Phosphorus (P) is an essential nutrient in freshwater ecosystems; however, in excess it can cause
44 blooms of algae and cyanobacteria. P enters lakes from the watershed (external loading) or when

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

58

59 **1 Introduction**

60 Sediments are an integral component of aquatic ecosystems, especially shallow lakes.
61 The sediments regulate whole-lake biogeochemical cycles through diagenetic processes and
62 material exchange across the sediment-water interface (Forsberg, 1989; Golterman, 2004). For
63 example, phosphorus (P) stored in sediments may reenter the water column due to disturbance,
64 microbial activity, or changes in chemical conditions at the sediment surface (Boström et al.,
65 1988; Søndergaard et al., 2003). In many ecosystems, this sediment P release (i.e., internal P
66 loading) can maintain high water column P concentrations, even if external nutrient inputs are
67 reduced (Søndergaard et al., 1999; Jeppesen et al., 2005; Søndergaard et al., 2013). Excess P

68 availability supports phytoplankton production (Elser et al., 2007; Schindler et al., 2016), which
69 can reach nuisance levels, threatening ecosystem services and public health (Schindler &
70 Vallentyne, 2008). The morphometric features of shallow lakes make these systems particularly
71 susceptible to internal P loading. Specifically, shallow basins have a large sediment surface area
72 relative to water volume. As a result, sediment-water interactions are highly influential in
73 determining whole-lake water chemistry (Welch & Cooke, 1995, Søndergaard et al., 2003).
74 Although it is known that internal P loading is an important process in shallow lakes, variation in
75 the underlying mechanisms remains poorly quantified for these ecosystems.

76 The chemical composition of the sediment P pool is an important mechanism controlling
77 internal loading because some forms of P are more mobile than others. Sediment P may be found
78 in a wide variety of minerals and organic materials as well as sorbed to particle surfaces (North
79 et al., 2015). This chemical speciation determines the conditions in which P is mobilized and
80 released as different P forms are vulnerable to different internal loading processes (Orihel et al.,
81 2017). For example, changes in redox potential due to fluctuations in dissolved oxygen and
82 alternative electron acceptor concentrations can mobilize P associated with redox-sensitive
83 minerals (i.e., iron- and manganese-bound P; Mortimer, 1941; Orihel et al., 2015). Changes in
84 pH may release other mineral-bound P forms (e.g., calcium-, and aluminum-bound; Jensen &
85 Andersen, 1992), and microbial decomposition can liberate P incorporated in labile organic
86 materials (Joshi et al., 2015; Song & Burgin, 2017; Frost et al., 2019). Sediment resuspension
87 due to wind disturbance or bioturbation releases pore water and surface-sorbed P (i.e., loosely-
88 bound P *sensu* James 2017b; Ekholm et al., 1997). Loosely-bound P is likely an important form
89 of sediment P in shallow lakes as these ecosystems experience frequent water column mixing
90 and wind disturbance (Bengtsson & Hellström, 1992). Although internal P loading occurs under

91 a variety of conditions, the dominant mechanisms are determined by the chemical composition of
92 the sediment P pool.

93 Sediment P content and chemical composition vary among lakes due to differences in
94 watershed features and external loading as well as autochthonous processing. External P loading
95 includes particulate and dissolved forms that are transported from the watershed into lakes
96 through surface and subsurface flows (Carpenter et al., 1998; Pease et al., 2018). The P
97 speciation in catchment soils will drive the chemical composition of particulate P inputs, in turn
98 influencing the composition of the sediment P pool (Kerr et al., 2011; Tang et al., 2019).
99 However, selective soil erosion and transport processes may alter sediment composition relative
100 to the soil matrix if various P fractions are not distributed evenly across grain sizes. Fine
101 particles that are more easily transported are often enriched in exchangeable, organic, and iron-
102 and aluminum-bound P and depleted in calcium-bound P relative to larger grain sizes (Stone &
103 English, 1992). As a result, lakebed sediments may have more labile P forms than the
104 surrounding soils (Kerr et al., 2011). In addition to exogeneous loading of particulate forms, P
105 may also enter waterbodies in an aqueous form or desorb from settling particles before reaching
106 the sediments (Pease et al., 2018). This soluble P is readily taken up by aquatic primary
107 producers. Following biological uptake, P may be cycled among producers and consumers or it
108 may settle out of the water column as necromass, phytoplankton detritus, or macrophyte leaf
109 litter (Orihel et al., 2017). The sources of sedimented organic matter influence sediment P
110 composition and lability (Twilley et al., 1986). For example, if phytoplankton detritus dominates
111 organic matter sedimentation, then sediment organic P will be associated with more labile
112 materials (Jiang et al., 2022). Inter-lake variation in sediment P composition, due to differences

113 in exogenous inputs and *in situ* processing, may also cause the dominant mechanisms of
114 internal P loading to vary among different waterbodies.

115 In addition to variation among different lakes, the spatial distribution of the sediment P
116 pool is also heterogeneous within individual lakes following variation in sediment transport,
117 deposition, and resuspension processes (Mackay et al., 2012). Sediment total P generally
118 increases with water depth as a result of sediment focusing, which concentrates fine-grained, P-
119 enriched sediments in deeper portions of the lake basin (White & Stone, 1996; Hou et al., 2014;
120 Cardoso-Silva et al., 2018). Dynamic disturbance and biological activity across the lakebed
121 further drive spatial variation in sediment P speciation (Nowlin et al., 2005; Trolle et al., 2006;
122 Kowalczywska-Madura et al., 2019a). Variable sediment disturbance has the greatest influence
123 on the distribution of loosely-bound P forms. Smaller stocks of loosely-bound P are expected in
124 shallow sediments that experience wind or wave disturbance while higher loosely-bound P
125 content is expected in deeper, more protected sediments. (James, 2017a). Additionally, the
126 location and density of macrophyte beds can influence the spatial distribution of sediment P as
127 macrophytes take up pore water P to meet their nutrient requirements (Xing et al., 2018) but also
128 protect stocks of loosely-bound P by reducing resuspension (Madsen, 2001). Sediment focusing
129 and disturbance as well as the presence of macrophytes are expected in shallow waterbodies
130 (Kelton & Chow-Fraser, 2005). As a result, the spatial distribution of sediment P content and
131 composition are expected to be heterogeneous within shallow lakes (Mackay et al., 2012; Orihel
132 et al., 2017). However, research on spatial variation in sediment P pools has focused on large or
133 deep waterbodies, and the degree of spatial heterogeneity in sediment P content and composition
134 in smaller, polymictic lakes remains poorly quantified.

135 In order to quantify the spatial heterogeneity of sediment P species in shallow lakes, we
136 measured the sediment P pools of seven shallow, glacial lakes in northwest Iowa, USA.
137 Specifically, we collected sediment cores from ten locations within each lake and analyzed the
138 sediments for total P as well as pore water and loosely-bound P. We quantified spatial variation
139 in these P forms and tested how sampling resolution influenced estimates of mean sediment P
140 concentrations. Water depth is expected to drive horizontal variation in sediment P within lakes,
141 with higher concentrations found in deeper areas. We further hypothesize that lakes with more
142 variable bathymetry, as quantified by volume development, will have greater variability in
143 loosely-bound P, driven by variation in sediment disturbance and macrophyte occurrence. The
144 sediment core from the deepest site in each study lake was additionally analyzed for total P and
145 five sediment P species (loosely-bound P, redox-sensitive, labile organic, and calcium-, and
146 aluminum-bound fractions). We hypothesize that variation in the composition of the sediment P
147 pool among lakes will relate to watershed soil composition, land cover, and long-term
148 phytoplankton biomass. Accurate inventories of sediment P stocks as well as estimates of P
149 burial rates and internal loading potential must account for spatial variation in sediment P content
150 and composition. By quantifying spatial heterogeneity in sediment P within shallow lakes, our
151 analysis suggests appropriate spatial resolutions for sampling sediment P pools; constrains the
152 uncertainty associated with scaling single measurements of P across the lakebed; and informs
153 models of lentic P cycling, such as those used for regulatory planning (e.g., total maximum daily
154 load calculations for P-impaired waterbodies). Our analysis of variation in sediment P pools
155 among different waterbodies illuminates how the mechanisms of internal P loading may also
156 vary among lakes and is a critical step to being able to predict internal P loading potential based
157 on lake and watershed characteristics.

158

159 **2 Materials and Methods**

160 2.1 Study lakes

161 We surveyed seven shallow, glacial lakes in northwest Iowa, USA (Table 1). The spatial
162 extent of the study lakes covers approximately 5,600 km² on the western edge of the Des Moines
163 Lobe. Although the study systems cover a wide range of basin characteristics, all are shallow
164 (average mean depth = 2.2 m) and follow a discontinuous cold polymictic stratification and
165 mixing pattern (Lewis 1983; Table S1). These lakes were formed from kettle depressions at the
166 end of the Wisconsin glaciation. The watershed soils developed from loamy glacial till and are
167 now heavily influenced by row crop agriculture (Arbuckle & Downing, 2001; Tables S2-3). The
168 watersheds of Storm and Swan Lake also contain soils developed from loess deposits as these
169 catchments are located on the boundary of the Des Moines Lobe and the Loess Prairies of
170 western Iowa. All of the study lakes are classified as eutrophic or hypereutrophic due to high
171 phosphorus concentrations (Carlson, 1977), but are characterized as P-limited based on long-
172 term TN:TP molar ratios (Table 1; Figure S1; Arbuckle & Downing, 2001).

173

174 2.2 Sediment core collection

175 We collected ten sediment cores from each study lake once between mid-June and early
176 July of 2018 using a gravity corer (inner diameter 5 cm). Sampling sites included the deep point
177 of the lake (hereafter “profundal sediments”), from which all sediment P species and total P were
178 measured, as well as nine shallower sites distributed across the lakebed, from which only
179 loosely-bound (i.e., NH₄Cl-extractable, including pore water and surface-sorbed P) and total P
180 were measured. These additional sites were selected using a randomized sampling scheme

181 stratified by water depth. Specifically, sites were randomly selected from an evenly spaced grid
182 of potential sites within each depth contour, defined as 0.5 or 1 meter intervals depending on the
183 maximum depth of the lake. All sediment cores were sliced in the field immediately following
184 collection. Cores from the profundal site, the zone of sediment accumulation, were sliced every 2
185 cm up to 10 cm deep and then every 5 cm up to 20 cm deep. One slice from 0-10cm was
186 collected for each of the other nine shallow sediment cores. Sediment samples were sealed in
187 plastic bags with the air removed and transported and stored at 4°C until analysis. All analyses
188 began within 36 hours of sample collection.

189

190 2.3 Sediment physical and chemical analyses

191 Sediments from each sampling site were thoroughly homogenized within their own
192 sample bags before subsampling for physical and chemical analyses. For analysis of physical
193 characteristics, a 0.4 to 0.5 g subsample was measured into a pre-weighed, oven-dried aluminum
194 weigh boat. The subsamples were then placed in a drying oven at $104 \pm 1^\circ\text{C}$ for at least 2 hours
195 to dry to a constant mass. The wet and dry masses of each subsample were used to calculate
196 sediment moisture content (MC; Text S1, Eq. 1). The samples were then combusted in a muffle
197 furnace for 2 hours at 550°C and weighed again to calculate organic matter content as loss-on-
198 ignition (Text S1, Eq. 2) and estimate bulk density (Text S1, Eq. 3; Håkanson & Jansson 2002).

199 An additional sediment subsample was analyzed for P speciation via sequential extraction
200 following the methods of James (2017b). This method was modified from the methods of
201 Nürnberg (1988) and Psenner and Puckso (1988). To begin the sequential P extractions,
202 subsamples of fresh sediment equivalent to 0.2 g of dry sediment (Text S1, Eq. 4) were weighed
203 into 50 mL polyethylene centrifuge tubes. The same sediment pellet was used throughout the

204 sequential extraction procedure. All extractions were performed at 25°C unless otherwise stated.
205 Centrifuge time and speed (30 minutes at 3000 rpm) as well as shaker table speed (120 rpm)
206 were consistent across extractions.

207

208 2.3.1 Loosely-bound P

209 To quantify loosely-bound P (all sampling sites in each lake), sediment was extracted in
210 25 mL of 1 M NH₄Cl. The samples were placed on a shaker table for two hours and then
211 centrifuged. After pouring off and saving the supernatant, we repeated this extraction on the
212 same sediment pellet. The combined, filtered (0.45µm GF/C filters) supernatants were analyzed
213 for soluble reactive phosphorus (SRP; Standard Methods 4500-P E) to determine the
214 concentration of sediment pore water and loosely-bound P (Text S1, Eq. 5).

215

216 2.3.2 Additional P species

217 For profundal sediment samples, additional extraction steps were performed on each core
218 slice immediately following the NH₄Cl extraction to quantify other P species. Redox-sensitive P
219 species were extracted in 12.5 mL of a 0.11 M bicarbonate – 0.1 M sodium dithionate solution.
220 Sample tubes were placed in a 40°C water bath for 30 minutes and centrifuged. The supernatant
221 was bubbled with air for at least 30 minutes before filtering and analyzing for SRP. Next, we
222 determined the amount of labile organic P and P associated with aluminum oxides with a two-
223 step 0.1 M NaOH extraction. We added 25 mL of the extractant to the sample tubes and shook
224 the samples for 17 hours. After centrifuging, the supernatant pH was adjusted to between 6-8
225 using 0.1 M HCl. A subsample of the supernatant was filtered and analyzed for SRP to determine
226 the aluminum-bound P concentration while the remainder underwent a persulfate digestion

227 (Standard Methods 4500-P B.5) before SRP analysis to determine the concentration of both
228 aluminum-bound and labile organic P. The labile organic P fraction was determined as the
229 difference between the total NaOH-extractable P and the subsample analyzed for aluminum-
230 bound P. Unfortunately, the persulfate digestion for total NaOH-extractable P failed for
231 sediments from three of the lakes (Swan, North Twin, and South Twin) due to an autoclave
232 malfunction. As such, we were unable to determine labile organic P for these lakes. Since the
233 error occurred during supernatant processing, we are confident that the sequential extractions for
234 other P fractions were not affected. For the final extraction for calcium-bound P, we added 25
235 mL of 0.5 M HCl to the remaining sediment pellet, shook for 24 hours, and centrifuged. We then
236 used 0.1 M NaOH to adjust the supernatant pH to within 6-8 before SRP analysis. All SRP
237 concentrations were corrected for pH adjustments and standardized by extractant volume and
238 sediment mass to determine the sediment concentration of each P species (Text S1, Eq. 5-9).

239

240 2.3.3 Sediment total P

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

249 concentrations were corrected for pH adjustments and dilution and standardized by the sediment
250 mass used (Text S1, Eq. 10).

251

252 2.4 Long-term chlorophyll-*a* data

253 In order to explore the relationship between sediment P composition and phytoplankton
254 biomass, we used chlorophyll-*a* concentrations as an index of phytoplankton biomass.

255 Chlorophyll-*a* data were obtained from the Iowa Department of Natural Resources AQuIA
256 public data repository. Chlorophyll-*a* concentrations are determined based on the U.S. EPA
257 method 445 from a depth integrated sample up to the thermocline or 2 m deep, whichever is
258 shallower (Arar & Collins, 1997; Standard Methods 10200-H). These measurements are made 3-
259 5 times each summer from May through September as a part of the Iowa Department of Natural
260 Resources Ambient Lake and Shallow Lakes monitoring programs. Water samples for these
261 analyses were collected from a single station at the deepest site of each study lake. Annual
262 monitoring began in 2000 in all lakes except South Twin where monitoring began in 2006. Data
263 from the first year of monitoring through the summer of 2018 when sediment samples were
264 collected were used to capture a long-term perspective of lake chlorophyll-*a* concentrations.

265

266 2.5 Aquatic macrophyte survey

267 We evaluated spatial relationships between macrophyte beds and sediment P pools in one
268 of the study lakes, Swan Lake, as there were extensive beds of submersed and floating-leaf
269 macrophytes in this waterbody. High turbidity limited macrophyte colonization in the other study
270 lakes; therefore, we focused our investigation of macrophytes and sediment P pools to Swan

271 Lake. We surveyed aquatic macrophyte community composition and bed density on Swan Lake
272 in July of 2018 (peak of seasonal growth) using the point-intercept method (Text S2).

273

274 2.6 Statistical analyses

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

293 the PCA biplot to explore how watershed land cover, basin morphology, and sediment physical
294 characteristics corresponded to patterns in profundal sediment P speciation among lakes.

295 In order to understand the relationship between sediment P bioavailability and
296 phytoplankton biomass, we performed a linear regression of the mean proportion of the total
297 sediment P pool found in a mobile form (the combination of the loosely-bound, total organic,
298 and redox-sensitive species) in the surface sediments (0-6 cm) at the profundal site versus the
299 long-term (13 or 19 years) mean chlorophyll-*a* concentration, as an index of phytoplankton
300 biomass, in the mixed surface layer of each lake. Sediment P and chlorophyll-*a* concentrations
301 are spatially congruent as both were measured at the deep site of each lake. Additionally, these
302 values represent a similar timescale. The surface sediments (0-6 cm) at the profundal sampling
303 sites represent roughly 45 years of sediment accumulation (Heathcote et al., 2013). As such, we
304 chose to compare these values to the best estimate available for long-term mean chlorophyll-*a*
305 concentrations. Using time-averaged chlorophyll-*a* values for each study lake also allowed us to
306 capture seasonal and inter-annual variation in phytoplankton biomass, as would be reflected in
307 the accumulated sediments.

308 To quantify spatial heterogeneity in loosely-bound and total P both within and among
309 lakes, we used measures from the nine spatially-distributed shallow sediment cores and the deep
310 site core slices averaged over depth interval slices. To make comparisons of the variability
311 among lakes we calculated the coefficient of variation for both total P and loosely-bound P in
312 each lake. To test if water depth at the sampling site correlated with the concentration of total P
313 and loosely-bound P, we used linear mixed effects regression with lake as a random effect on the
314 intercept. We used likelihood ratio tests to evaluate the importance of random effects of the lake
315 on model intercepts. We also tested the influence of lake basin volume development (D_v) on

316 spatial variation in loosely-bound P. The D_v score is a ratio of the volume of a lake basin to the
317 volume of a perfect cone with a base area equal to the surface area of the lake and a height equal
318 to the maximum water depth. The D_v ratio is used to characterize lake basin shape. Lakes with
319 low D_v values have cone-shaped basins with localized deep holes while higher D_v ratios
320 correspond to shallow, bowl-shaped bathymetries (Hutchinson, 1957).

321 We used a rarefaction analysis to determine the spatial sampling resolutions needed for
322 accurate inventories of sediment P pools (Ortiz & Wilkinson, 2021). Specifically, we tested how
323 the number of sampling sites affected the whole-lake estimate of mean total and loosely-bound P
324 concentrations. For each study lake and P form, the data were randomly subset to between 2 and
325 9 sites. The mean P concentration for this sampling subset was then calculated and compared to
326 the “true” mean of all 10 sites based on the root mean square error (RMSE). This analysis was
327 repeated for 1000 iterations and RMSE values were averaged for each sampling subset and
328 normalized to the mean for all 10 sites to express the estimated error a proportion of the “true”
329 mean concentration.

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

334

335 **3 Results**

336 3.1 Inter-lake variation in profundal sediment P

337 The size and composition of the profundal sediment P pool varied considerably among
338 the study lakes. Total sediment P concentrations, averaged across the depth intervals of

339 profundal cores, ranged from 738.2 to 1,164.7 $\mu\text{g P g}^{-1}$ dry sediment. The chemical speciation of
340 the profundal sediment P pool also differed among the study lakes (Table 2). Redox-sensitive
341 and organic P fractions were consistently the most abundant components of the total sediment P
342 pool while aluminum and calcium-bound P were usually present in the lowest concentrations.
343 We used PCA as part of a compositional analysis to explore patterns in profundal sediment P
344 chemistry among our study lakes and to identify lakes with similar sediment P composition
345 (Figure 1A). The first principal component (PC1) explained 45.27% of the variation in the
346 dataset and was most closely associated with the prevalence of redox-sensitive and aluminum-
347 bound P. The second principal component (PC2) explained 36.42% of the variation and was
348 highly correlated with organic P content. Together, the first two principal components explained
349 81.69% of the variance in the dataset. Sediment P speciation defined groups of similar lakes
350 (Table 2, Figure 1A). Center, Five Island, Storm, and Silver Lake formed the largest group,
351 based on lower organic P concentrations. Silver Lake was slightly separated from this group due
352 to higher concentrations of loosely-bound P. Abundant organic and loosely-bound P placed
353 North and South Twin Lake in a group together. Swan Lake was distinguished from the other
354 study lakes due to high redox-sensitive and aluminum-bound P concentrations.

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

362 predominantly Sac, Primghar, Marcus, and Galva series formed in loess while the other
363 watershed soils were composed of series formed in glacial till.

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

375 In order to evaluate the ecosystem-level implications of the observed variation in
376 profundal sediment P speciation among our study lakes, we tested the relationship between the
377 relative availability of mobile P fractions in the sediment and phytoplankton biomass in each
378 lake. We performed a linear regression of the percent of total sediment P that is mobile (loosely-
379 bound, total organic, and redox-sensitive) versus long-term mean chlorophyll-*a* concentrations.
380 There was a significant positive relationship between the percentage of total P that is mobile and
381 chlorophyll-*a* concentrations among the seven study lakes ($F_{1,5} = 7.6$; $p = 0.04$; adjusted $R^2 =$
382 0.52 ; $\beta_1 = 4.9$, $CI_{95\%} = [0.33, 9.5]$; Figure 2).

383

384 3.2 Intra-lake variation in sediment P

385 There was substantial spatial variation in total and loosely-bound P within individual
386 study lakes. Loosely-bound P had greater spatial heterogeneity than total P within most lakes.
387 There was a 1.2 to 2.2-fold within-lake difference in sediment total P and a 1.7 to 11.3-fold
388 difference in loosely-bound P. Values for the coefficient of variation for loosely-bound P (range
389 = 11.4 to 39.7%, mean = 25.7%) were also greater than the values for total P (range = 6.4 to
390 22.4%, mean = 14.0%; Table 2). South Twin was the only lake for which the coefficient of
391 variation for loosely-bound P was less than the coefficient of variation for total P. The depth of
392 the water column at the coring location explained 63.1% of the variation in sediment total P and
393 84.6% of the variation in loosely-bound P when lake was included as a random effect on the
394 intercept (Figure 3). Including random effects on the model intercepts substantially improved the
395 model fits ($p < 0.0001$ for both models). For both the total sediment P and loosely-bound
396 fractions, concentrations increased with depth. The increase in P with water depth was greater for
397 total P ($\beta_1 = 65.3$, $CI_{95\%} = [27.6, 102.5]$) than loosely-bound P ($\beta_1 = 12.8$, $CI_{95\%} = [7.2, 18.3]$).

398 We further explored the influence of lake basin morphology on spatial variation in
399 loosely-bound P through an analysis of volume development (D_v). We found an inverse
400 relationship between D_v and the variability in loosely-bound P, measured as the coefficient of
401 variation (Figure 4). Five Island, Storm, and Swan Lakes had the greatest within-lake variation in
402 loosely-bound P (CV range = 32.0 to 39.7%), and these waterbodies also had low D_v values
403 (range = 0.65 to 1.36). Conversely, we found much less variability in loosely-bound P (CV range
404 = 11.4 to 24.4%) in Center, Silver, North and South Twin Lakes, which all had higher D_v scores
405 near 2 (range = 1.98 to 2.12). To evaluate additional explanations for within-lake variation in
406 sediment P, we surveyed spatial relationships between macrophytes and sediment P in Swan
407 Lake (Figure S2). Both total and loosely-bound sediment P concentrations were lowest in the

408 shallow, northern portion of the lake, and concentrations increased in the deeper, southern
409 portion of the lake. There was no clear pattern between sediment P concentration and
410 macrophyte bed density.

411 A better understanding of intra-lake variation in sediment P concentrations will inform
412 appropriate spatial sampling resolutions to accurately quantify sediment P pools. We used a
413 rarefaction analysis to test how the number of sampling sites affected the estimate of mean total
414 and loosely-bound P in each study lake. For sediment total P, normalized RMSE values began to
415 plateau when 6 or more sampling sites were included (Figure 5A). In subsets of 6 or more
416 sampling sites, the normalized RMSE was less than 5 percent for all study lakes (range = 1.37 to
417 4.71%). Some study lakes had relatively low normalized RMSE values regardless of how many
418 sites were included while others demonstrated sharp declines in normalized RMSE as the
419 number of sites increased. These differences among lakes did not follow patterns with basin D_v
420 or maximum water depth. Compared to total P, the number of sampling sites had a greater
421 impact on the estimate of mean loosely-bound P within each study lake (Figure 5B). Normalized
422 RMSE values did not reach obvious plateaus for any study lakes. Lakes with low D_v values (i.e.,
423 Five Island, Storm, and Swan Lakes) had higher normalized RMSE values and more drastic
424 improvements in RMSE with increasing number of sample sites. Conversely, lakes with higher
425 D_v values (i.e., Center, Silver, North and South Twin Lakes) had lower normalized RMSE and
426 more gradual improvements in RMSE with more sampling sites.

427 In addition to quantifying the effects of spatial sampling resolution on P pool estimates,
428 we also tested the influence of sample site location, specifically whether P concentrations at the
429 deep site were representative of mean P concentrations across the lakebed. We compared total
430 and loosely-bound P concentrations from the deep site of each lake to the whole-lake mean

431 (Table S4). For sediment total P, concentrations at the deep site exceeded the whole-lake average
432 for all lakes except Swan Lake. Normalized RMSE values ranged from 0.3 to 11.3 percent (mean
433 = 4.6 %). Loosely-bound P concentrations at the deep site were consistently greater than whole-
434 lake means. Normalized RMSE values were much greater than for total P (range = 2.2 to 52.5%,
435 mean = 26.2%).

436

437 **4 Discussion**

438 4.1 Patterns and mechanisms of inter-lake variation in sediment P pools

439 We quantified spatial heterogeneity in sediment P content and composition in shallow
440 lakes and found high variation both within and among different waterbodies. Across the study
441 lakes, whole-lake means of sediment total P content ranged from 897.0 to 1,184.9 $\mu\text{g P g}^{-1}$ dry
442 sediment, with a grand mean of 946.2 $\mu\text{g P g}^{-1}$ dry sediment. These concentrations align with
443 ranges from other shallow, eutrophic lakes (Søndergaard et al., 2013) and other productive
444 waterbodies more generally (Doig et al., 2017; Kowalczywska-Madura et al., 2019b; Tao & Lu,
445 2020). Overall, sediment total P concentrations in our study lakes were either similar to or lower
446 than average values reported for other productive waterbodies (Table S5). Sediment P
447 composition also varied among the study lakes; however, redox-sensitive and organic P were
448 consistently the most abundant fractions. Other eutrophic waterbodies also have high levels of
449 redox-sensitive P (Song & Burgin, 2017; Randall et al., 2019; Papera et al., 2021) and organic P
450 forms (Nowlin et al., 2005; Frost et al., 2019). More generally, aquatic sediments tend to be
451 enriched in redox-sensitive, organic, loosely-bound, and aluminum-bound P because these forms
452 are commonly associated with fine sediments, which are more easily eroded from the source
453 material (i.e., terrestrial soils; Stone & English, 1992; Kerr et al., 2011).

454 Variation in sediment P composition among the study lakes followed differences in
455 watershed and lake basin characteristics. The North and South Twin Lake and Swan Lake
456 groupings reflect patterns in dominant watershed soil series. However, there was variation in
457 watershed soil characteristics within the Center, Five Island, Silver and Storm Lake group as
458 soils in the Storm Lake catchment were formed partially in loess while the other watershed soils
459 were formed in glacial till. Variation in watershed soils within this group reinforces that aquatic
460 sediment P composition is not a direct reflection of the bulk soil matrix due to selective erosional
461 and sediment transport processes as well as *in situ* processing (Kerr et al., 2011). For example,
462 similarities in P composition within this group were related to maximum depth and sediment
463 bulk density, suggesting that these waterbodies may experience more similar sediment transport,
464 deposition, and resuspension processes compared to other study lakes (Mackay et al., 2012).

465 In addition to the role of watershed and lake basin features in driving differences in
466 sediment P composition across the study lakes, we also found a direct relationship between long-
467 term mean chlorophyll-*a* concentrations, as an index of phytoplankton biomass, and the average
468 contribution of mobile P species to the total sediment P pool in surface sediments. This
469 relationship suggests a positive feedback loop between mobile sediment P forms, heightened
470 internal P loading rates, and phytoplankton biomass (Katsev et al., 2006; Song & Burgin, 2017).
471 Specifically, internally-loaded P from mobile forms in the sediments supports phytoplankton
472 production, while sedimentation of phytoplankton detritus supplies more mobile P to the
473 sediments. Organic P was the largest fraction of total sediment P in all of the study lakes, driving
474 the among-lake variability in mobile P contributions. Given the prevalence of organic P in the
475 sediments, it is likely that decomposition of sediment organic matter and subsequent P
476 mineralization are key processes for internal P loading in the study lakes (Joshi et al., 2015; Frost

477 et al., 2019). In hypereutrophic waterbodies rampant phytoplankton production and detritus
478 enhance organic matter sedimentation, increasing organic P in lakebed sediments. Large pools of
479 organic P are associated with high extracellular enzyme activities, which mobilize and release
480 sediment P (Song & Burgin, 2017). Our study lakes are likely examples of systems in which
481 biologically-mediated cycling of organic P pools drives internal P loading and high
482 phytoplankton production in turn increases mobile P forms in the sediments.

483

484 4.1.1 Implications of inter-lake variation in sediment P pools

485 For a given waterbody, the dominant mechanisms of internal P loading depend on the
486 chemical composition of the sediment P pool (Orihel et al., 2017). The variation that we
487 measured in sediment P composition among different lakes implies that the processes driving
488 sediment P release also vary across these ecosystems. For example, redox-sensitive P was the
489 dominant fraction in Swan Lake while North and South Twin Lake a higher prevalence of
490 organic P. Based on these differences in sediment P composition, we would expect sediment P
491 release in these lakes to respond differently to dissolved oxygen levels at the sediment-water
492 interface. Although these lakes are polymictic, they are also very productive such that periods of
493 thermal stratification can cause anoxia in bottom waters. Anoxia would likely prompt sediment P
494 release in Swan Lake due to the dissolution of redox-sensitive minerals under reducing
495 conditions (Mortimer, 1941; Orihel et al., 2015). Anaerobic sediment P release from redox-
496 sensitive P minerals could also occur in North and South Twin Lake; however, due to high
497 organic P levels, we would also expect to see sediment P release under oxic conditions due to
498 decomposition of sediment organic matter and subsequent P mineralization (Joshi et al., 2015;
499 Song & Burgin, 2017; Frost et al., 2019). Bottom water aeration, a management approach to

500 limit anaerobic internal P loading, might protect redox-sensitive minerals and reduce sediment P
501 release in Swan Lake, but the same approach in North and South Twin Lakes could stimulate
502 decomposition and enhance aerobic P release (Horppila et al., 2017, Tammeorg et al., 2017).
503 Inter-lake variation in sediment P composition means that management strategies to control
504 internal P loading are unlikely to translate well across lakes with different sediment P chemistry.

505

506 4.2 Mechanisms of intra-lake variation in sediment P pools

507 In addition to variation in sediment P content and composition across different lakes, we
508 also quantified spatial heterogeneity in total and loosely-bound P pools within individual lakes.
509 Consistent with our expectations, loosely-bound P concentrations were most variable in lakes
510 with low volume development indices, indicative of conical basins with localized deep holes.
511 This bathymetric variability produces both shallow regions where frequent sediment disturbance
512 releases loosely-bound P and deeper areas where loosely-bound P is protected (James, 2017a).
513 As a result, loosely-bound P concentrations are more variable in lake basins with more complex
514 bathymetry. In contrast, lakes with high volume development scores, associated with shallow,
515 bowl-shaped basins, had more consistent loosely-bound P concentrations, as sediment
516 disturbance was likely more uniform across the lakebed.

517 We also hypothesized that greater water depths would correlate with higher
518 concentrations of total and loosely-bound sediment P across the lakebed, which was supported
519 by the mixed model analysis. In fact, the depth of the water at the sediment sampling location
520 described the vast majority (84.1%) of the variation in loosely-bound P and over half of the
521 variation (63.1%) in total P concentrations. The pattern of higher concentrations at greater depths
522 is likely driven by sediment focusing and greater net sediment deposition in deeper portions of

523 the lake (White & Stone, 1996; Hou et al., 2014; Cardoso-Silva et al., 2018). The direct
524 relationship that we observed between water depth and sediment P concentrations is consistent
525 with other studies of spatial variation in sediment P pools (Nowlin et al., 2005; Kowalczevska-
526 Madura et al., 2019b). However, water depth did not fully explain the observed variation in
527 sediment total and loosely-bound P, indicating that factors beyond basin morphology contribute
528 to spatial variation in sediment P pools.

529 A factor that may influence the spatial distribution of the sediment P pool is the location
530 and density of macrophyte beds. However, contrary to our expectations, we found no clear
531 spatial pattern between macrophyte bed density and sediment P pools. High turbidity limited the
532 depth of macrophyte colonization in most of our study lakes such that we were only able to test
533 these spatial relationships in one lake. Another complicating factor is that macrophytes could be
534 expected to either increase sediment P stocks by stabilizing sediments and reducing resuspension
535 (Carpenter et al., 1983; Madsen, 2001) or decrease sediment P pools via biological uptake (Xing
536 et al., 2018). Exploring the balance of this complex relationship in more waterbodies is an
537 avenue for future research.

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

545 natural progression of this research is to explore temporal variation in sediment P content and
546 composition.

547

548 4.3 Applications and significance

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

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

587 A quantitative understanding of inter- and intra-lake variation in sediment P content and
588 composition is essential for accurately sampling, scaling, and modeling sediment P pools, yet
589 this variability has been largely undescribed for shallow lakes. Our study builds on our
590 understanding of regional patterns in lacustrine sediment P speciation and contributes novel

591 perspectives on the causes and consequences of spatial heterogeneity in sediment P pools within
592 shallow lakes. Our finding of inter-lake variation in sediment P composition reinforces that
593 management strategies to control internal P loading will also differ among lakes and that
594 effective solutions will consider site-specific features, including sediment P chemistry. Our
595 analysis further supports that shallow lakes are spatially heterogenous ecosystems, and
596 accounting for this variation is necessary to accurately characterize sediment P pools. The large
597 within-lake variability in loosely-bound and total P documented in this study reinforces the
598 importance of sampling multiple sites across the lakebed when possible and using caution when
599 extrapolating measures from the deep site to the whole system. Accurate sediment P inventories
600 will allow for more empirical parameterization of sediment-water interactions in models of lake
601 P cycling. Our quantification of spatial heterogeneity in sediment P pools within and among
602 lakes is a critical step to being able to predict internal P loading potential based on lake and
603 watershed characteristics.

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

618 **Tables**619 **Table 1.** *Location, Basin Morphology, and Watershed Features of the Study Lakes (Iowa, 2018)*

	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- <i>a</i> ($\mu\text{g L}^{-1}$)	37.58	47.08	30.62	16.65	53.89	85.22	77.94
% 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
Mean TN:TP (molar)	56.2	116.6	118.7	50.9	77.5	51.5	20.5

620 *Note.* Chlorophyll-*a* concentrations and TN:TP molar ratios are the 19-year mean (2000-2018) of measurements
621 made during the ice-free season (n=36-49 measurements per lake) with the exception of South Twin Lake
622 which is the 13-year mean (2006-2018) of measurements made during the ice-free season (n=35).
623

624 **Table 2.** *Variation in Profundal Sediment P Among Lakes and Within-Lake Heterogeneity (Iowa, 2018)*
625

	Center Lake	Five Island	Silver Lake	Storm Lake	North Twin	South Twin	Swan Lake
Total P ($\mu\text{g P g}^{-1}$ 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

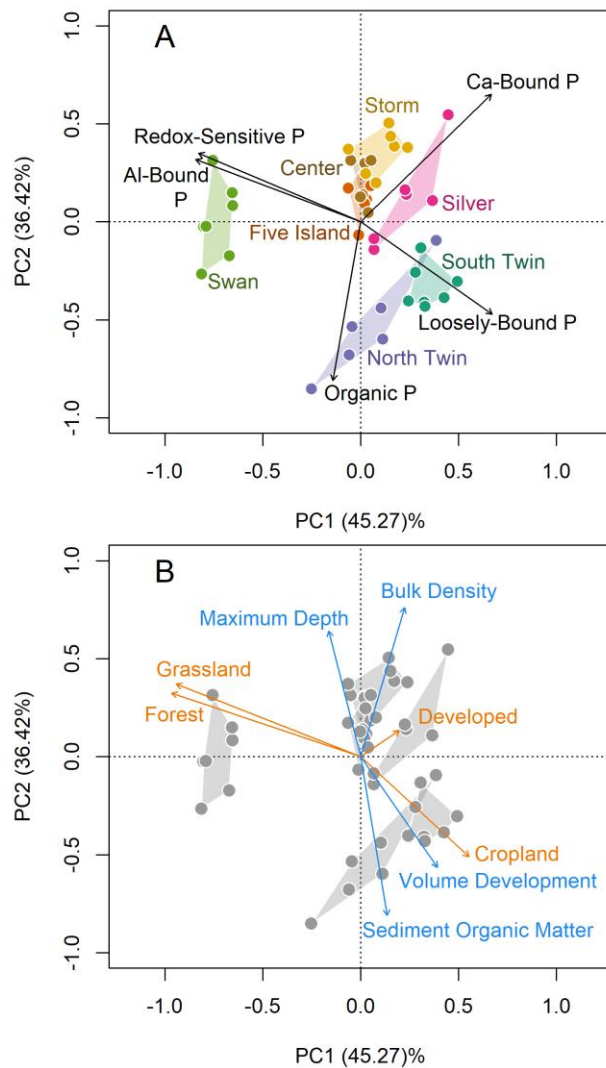
626 *Note.* Total P values are the mean (\pm standard deviation) for all intervals of the deep site core. The percent contribution of
627 each P species to the total sediment P pool is an average value across the deep site core profile. The coefficient of
628 variation for total and loosely-bound P was calculated for all ten sediment cores collected across each study lake.
629

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631

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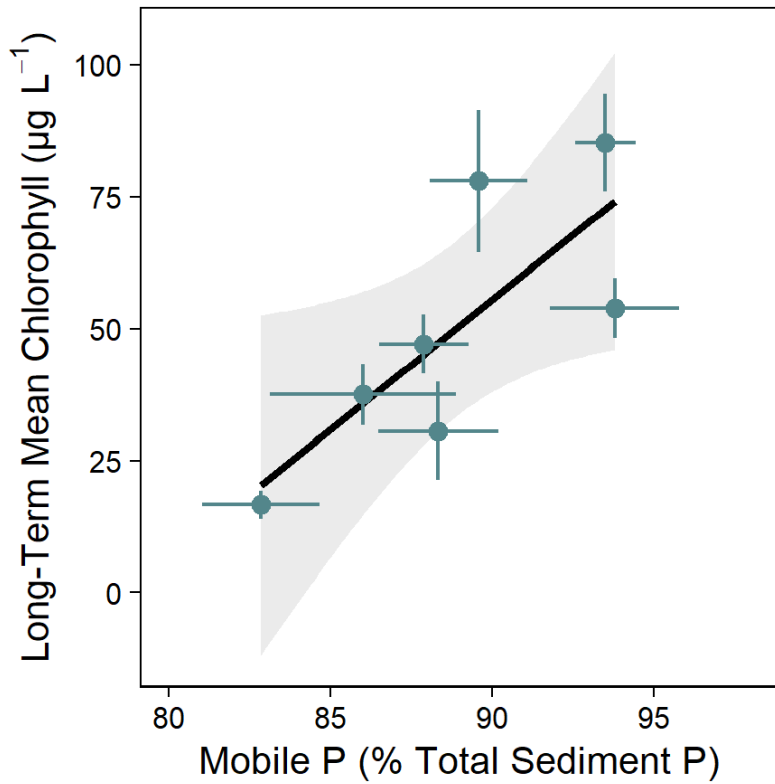
633 **Figures**
 634



635 **Figure 1. (A)** PCA biplot based on a compositional data analysis of profundal sediment P pools.
 636

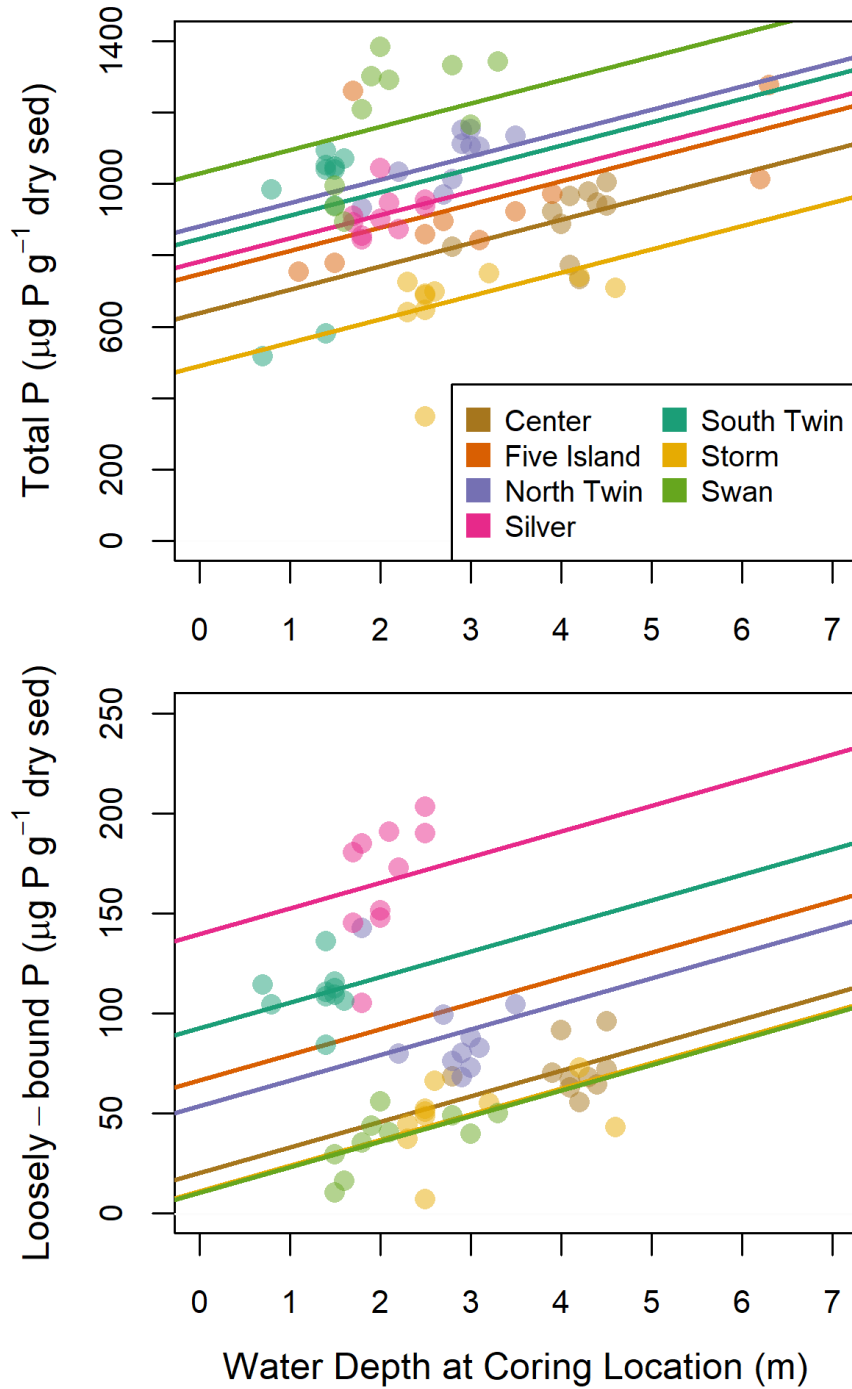
637 The compositional analysis was defined by the concentrations of loosely-bound (porewater and
 638 surface sorbed), redox-sensitive (Fe- and Mn-bound), aluminum-bound, calcium-bound, and
 639 organic P (including labile and refractory components). Arrows represent center log-ratio
 640 transformed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

643



644

645 **Figure 2.** The relationship between the fraction of the total sediment P pool that is comprised of
646 mobile species (averaged over 0-6 cm at the profundal sampling site) and the long-term (2000-
647 2018 for most study lakes or 2006-2018 for South Twin Lake) mean chlorophyll-*a*
648 concentrations in each study lake ($F_{1,5} = 7.6$; $p = 0.04$; adjusted $R^2 = 0.52$; $\beta_1 = 4.9$, $CI_{95\%} =$
649 $[0.33, 9.5]$). The grey shaded area indicates the 95% confidence interval.

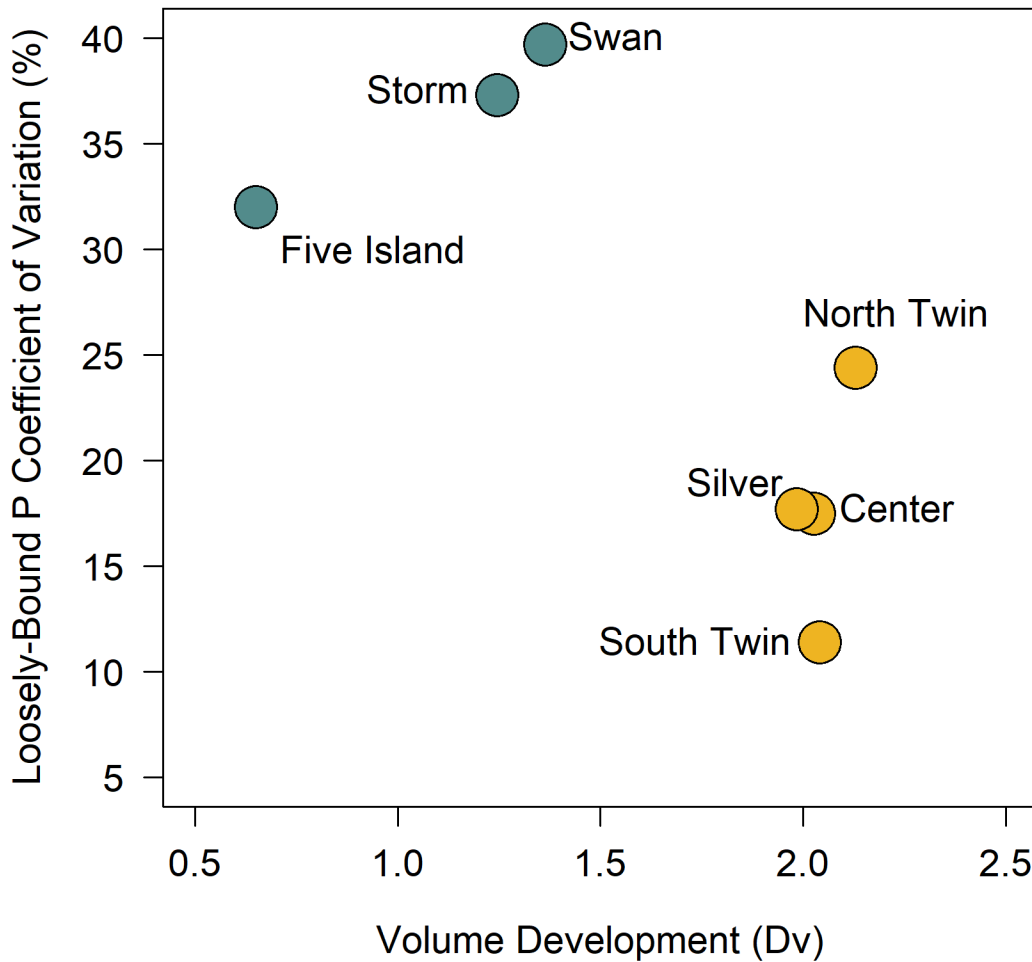


650

651 **Figure 3.** Mixed model regression effects of water depth at the coring location on total sediment

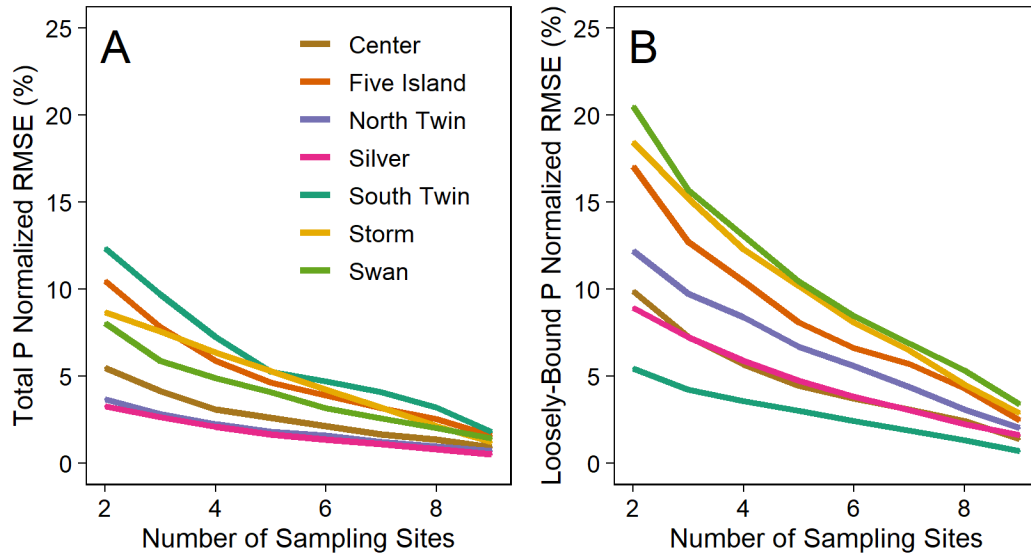
652 P ($\beta_1 = 65.3$, $CI_{95\%} = [27.6, 102.5]$; top panel) and loosely-bound sediment P ($\beta_1 = 12.8$, $CI_{95\%} =$

653 $[7.2, 18.3]$; bottom panel) by lake.



654

655 **Figure 4.** Variation in sediment loosely-bound P across lake basin volume development. The
 656 coefficient of variation for loosely-bound P was highest in lakes with low volume development
 657 scores (blue points), indicating more variable basin shape. Lakes with higher volume
 658 development ratios (yellow points) had less intra-lake variation in loosely-bound P.



659

660 **Figure 5.** Rarefaction analysis of mean total (A) and loosely-bound (B) P concentrations based

661 on the number of sampling sites. RMSE values are normalized as a percent of the mean P

662 concentration when all 10 sampling sites are included.

663

664 **References**

- 665 1. Albright, E.A., Wilkinson, G.M., Fleck, R.A., & Shingai, Q. (2020). Analysis of Sediment
666 Phosphorus Pools in Shallow Lakes. figshare. Dataset.
667 <https://doi.org/10.6084/m9.figshare.13362971.v1>
- 668 2. Arar, E. J., & Collins, G.B. (1997). Method 445.0 *in vitro* determination of chlorophyll a and
669 pheophytin a in marine and freshwater algae by fluorescence. U.S. Environmental
670 Protection Agency, Washington, DC.
- 671 3. Arbuckle, K.E., & Downing, J.A. (2001). The influence of watershed land use on lake N:P in
672 a predominantly agricultural landscape. *Limnology and Oceanography*, 46(4), 970-975.
673 <https://doi.org/10.4319/lo.2001.46.4.0970>
- 674 4. Bengtsson, L., & Hellström, T. (1992). Wind-induced resuspension in a small shallow lake.
675 *Hydrobiologia*, 241, 163-172. <https://doi.org/10.1007/BF00028639>
- 676 5. Boström, B., Andersen, J.M., Fleischer, S., & Jansson, M. (1988). Exchange of phosphorus
677 across the sediment-water interface. *Hydrobiologia*, 170, 229-244.
678 <https://doi.org/10.1007/BF00024907>
- 679 6. Bray, A., Ismay, C., Chasnovski, E., Baumer, B., & Cetinkaya-Rundel, M. (2021). infer:
680 Tidy Statistical Inference. R package version 0.5.4. [https://CRAN.R-](https://CRAN.R-project.org/package=infer)
681 [project.org/package=infer](https://CRAN.R-project.org/package=infer)
- 682 7. Cardoso-Silva, S., Ferreira, P.A., Figueira, R.C.L., Silva, D.C., Moschini-Carlos, V., &
683 Pompêo, M.L.M. (2018). Factors that control the spatial and temporal distributions of
684 phosphorus, nitrogen, and carbon in the sediments of a tropical reservoir. *Environmental*
685 *Science and Pollution Research*, 25, 31776–31789. [https://doi.org/10.1007/s11356-018-](https://doi.org/10.1007/s11356-018-2923-0)
686 [2923-0](https://doi.org/10.1007/s11356-018-2923-0)

- 687 8. Carlson, R.E. (1977). A trophic state index for lakes. *Limnology and Oceanography*, 22(2):
688 361-369. <https://doi.org/10.4319/lo.1977.22.2.0361>
- 689 9. Carpenter, S.R., Caraco, N.F., Correll, D.L., Howarth, R.W., Sharpley, A.N., & Smith, V.H.
690 (1998). Nonpoint pollution of surface waters with phosphorus and nitrogen. *Ecological*
691 *Applications*, 8(3): 559-568. [https://doi.org/10.1890/1051-](https://doi.org/10.1890/1051-0761(1998)008[0559:NPOSWW]2.0.CO;2)
692 [0761\(1998\)008\[0559:NPOSWW\]2.0.CO;2](https://doi.org/10.1890/1051-0761(1998)008[0559:NPOSWW]2.0.CO;2)
- 693 10. Carpenter, S.R., Elser, J.J., & Olson, K.M. (1983). Effects of roots of *Myriophyllum*
694 *verticillatum* L. on sediment redox conditions. *Aquatic Botany*, 17(3-4), 243-249.
695 [https://doi.org/10.1016/0304-3770\(83\)90060-8](https://doi.org/10.1016/0304-3770(83)90060-8)
- 696 11. Doig, L.E., North, R.L., Hudson, J.J., Hewlett, C., Lindenschmidt, K.-E., & Liber, K. (2017).
697 Phosphorus release from sediments in a river-valley reservoir in the northern Great Plains
698 of North America. *Hydrobiologia*, 78(1): 323-339. [https://doi.org/10.1007/s10750-016-](https://doi.org/10.1007/s10750-016-2977-2)
699 [2977-2](https://doi.org/10.1007/s10750-016-2977-2)
- 700 12. Ekholm, P., Malve, O., & Kirkkala, T. (1997). Internal and external loading as regulators of
701 nutrient concentrations in the agriculturally loaded Lake Pyhäjärvi (southwest Finland).
702 *Hydrobiologia*, 345, 3-14. <https://doi.org/10.1023/A:1002958727707>.
- 703 13. Elser, J.J., Bracken, M.E.S., Cleland, E.E., Gruner, D.S., Harpole, W.S., Hillebrand, H., et al.
704 (2007). Global analysis of nitrogen and phosphorus limitation of primary producers in
705 freshwater, marine and terrestrial ecosystems. *Ecology Letters*, 10, 1135-1142.
706 <https://doi.org/10.1111/j.1461-0248.2007.01113.x>
- 707 14. Engstrom, D.R., & Rose, N.L. (2013). A whole-basin, mass-balance approach to
708 paleolimnology. *Journal of Paleolimnology*, 49: 333-347.
709 <https://doi.org/10.1007/s10933-012-9675-5>

- 710 15. Filzmoser, P., Hron, K., & Templ, M. (2018). *Applied Compositional Data Analysis with*
711 *Worked Examples in R*. Spring Series in Statistics. Springer International Publishing,
712 Cham, Switzerland.
- 713 16. Forsberg, C. (1989). Importance of sediments in understanding nutrient cyclings in lakes.
714 *Hydrobiologia*, 176/177, 263-277. <https://doi.org/10.1007/BF00026561>
- 715 17. Frost, P. C., Prater, C., Scott, A. B., Song, K., & Xenopoulos, M. A. (2019). Mobility and
716 Bioavailability of Sediment Phosphorus in Urban Stormwater Ponds. *Water Resources*
717 *Research*, 55(5), 3680-3688. <https://doi.org/10.1029/2018WR023419>
- 718 18. Golterman, H.L. (2004). *The Chemistry of Phosphate and Nitrogen Compounds in*
719 *Sediments*. Springer Science+Business Media, B.V.
- 720 19. Hanson, P.C., Stillman, A.B., Jia, X., Karpatne, A., Dugan, H.A., Carey, C.C., et al. (2020).
721 Predicting lake surface water phosphorus dynamics using process-guided machine
722 learning. *Ecological Modelling*, 430: 109136.
723 <https://doi.org/10.1016/j.ecolmodel.2020.109136>
- 724 20. Heathcote, A.J., Filstrup, C.T., & Downing, J.A. (2013). Watershed sediment losses to lakes
725 accelerating despite agricultural soil conservation efforts. *PLoS ONE*, 8(1): e 53554.
726 <https://doi.org/10.1371/journal.pone.0053554>
- 727 21. Hou, D., He, J., Lü, C., Dong, S., Wang, J., Xie, Z., et al. (2014). Spatial variation and
728 distributions of phosphorus and nitrogen in bottom sediments from a typical north-
729 temperate lake, China. *Environmental Earth Sciences*, 71, 3063-3079.
730 <https://doi.org/10.1007/s12665-013-2683-6>
- 731 22. Horppila, J., Holmroos, H., Niemistö, J., Massa, I., Nygrén, N., Schönach, P., et al. (2017).
732 Variations of internal phosphorus loading and water quality in a hypertrophic lake during

- 733 40 years of different management efforts. *Ecological Engineering* 103: 264-274.
734 <http://dx.doi.org/10.1016/j.ecoleng.2017.04.018>
- 735 23. Hutchinson, G.E. (1957). *A treatise on limnology*. Volume 1. New York, NY: Wiley.
- 736 24. Håkanson, L., & Jansson, M. (2002). *Principles of lake sedimentology*. Caldwell, NJ: The
737 Blackburn Press. <https://doi.org/10.1002/iroh.19850700318>
- 738 25. James, W.F. (2017a). Internal phosphorus loading contributions from deposited and
739 resuspended sediment to the Lake of the Woods. *Lake and Reservoir Management*, 34(4),
740 347-359. <https://doi.org/10.1080/10402381.2017.1312647>
- 741 26. James, W.F. (2017b). Diffusive phosphorus fluxes in relation to the sediment phosphorus
742 profile in Big Traverse Bay, Lake of the Woods. *Lake and Reservoir Management*, 33(4),
743 360-368. <https://doi.org/10.1080/10402381.2017.1346010>
- 744 27. Jensen, H.S., & Andersen, F.Ø. (1992). Importance of temperature, nitrate, and pH for
745 phosphate release from aerobic sediments of four shallow, eutrophic lakes. *Limnology*
746 *and Oceanography*, 37(3), 577-598. <https://doi.org/10.4319/lo.1992.37.3.0577>
- 747 28. Jeppesen, E., Søndergaard, M., Jensen, J.P., Havens, K.E., Anneville, O., Carvalho, L., et al.
748 (2005). Lake responses to reduced nutrient loading — an analysis of contemporary long-
749 term data from 35 case studies. *Freshwater Biology*, 50(10), 1747–1771.
750 <https://doi.org/10.1111/j.1365-2427.2005.01415.x>
- 751 29. Jiang, X.Y., Gao, G., Hu, Y., Shao, K.Q., Tang, X.M., Cheng, H., and Li, J. (2022). The shift
752 from macrophytic to algal particulate organic matter favours dissimilatory nitrate
753 reduction to ammonium over denitrification in a eutrophic lake. *Freshwater Biology*,
754 67(3): 564-576. <https://doi.org/10.1111/fwb.13863>

- 755 30. Joshi, S.R., Kukkadapu, R.K., Burdige, D.J., Bowden, M.E., Sparks, D.L., & Jaisi, D.P.
756 (2015). Organic matter remineralization predominates phosphorous cycling in the mid-
757 bay sediments in the Chesapeake Bay. *Environmental Science and Technology*, 49, 5887-
758 5896. <https://doi.org/10.1021/es5059617>
- 759 31. Katsev, S., Tsandev, I., L'Heureux, I., and Rancourt, D.G. (2006). Factors controlling long-
760 term phosphorus efflux from lake sediments: Exploratory reactive-transport modeling.
761 *Chemical Geology*, 234(1-2): 127-147. <https://doi.org/10.1016/j.chemgeo.2006.05.001>
- 762 32. Kelton, N., & Chow-Fraser, P. (2005). A simplified assessment of factors controlling
763 phosphorus loading from oxygenated sediments in a very shallow eutrophic lake. *Lake*
764 *and Reservoir Management*, 21(3): 223-230.
765 <https://doi.org/10.1080/07438140509354432>
- 766 33. Kerr, J.G., Burford, M.A., Olley, J.M., Bunn, S.E., & Udy, J. (2011). Examining the link
767 between terrestrial and aquatic phosphorus speciation in a subtropical catchment: The
768 role of selective erosion and transport of fine sediments during storm events. *Water*
769 *Research*, 45, 3331-3340. <https://doi.org/10.1016/j.watres.2011.03.048>
- 770 34. Kowalczywska-Madura, K., Gołdyn, R., Bogucka, J., & Strzelczyk, K. (2019a). Impact of
771 environmental variables on spatial and seasonal internal phosphorus loading in a
772 mesoeutrophic lake. *International Journal of Sediment Research*, 34(1),14–26.
773 <https://doi.org/10.1016/j.ijsrc.2018.08.008>
- 774 35. Kowalczywska-Madura, K., Kozak, A., Dera, M., & Gołdyn, R. (2019b). Internal loading of
775 phosphorus from bottom sediments of two meso-eutrophic lakes. *International Journal of*
776 *Environmental Research*, 13: 325-251. <https://doi.org/10.1007/s41742-019-00167-y>

- 777 36. Kuznetsova, A., Brockhoff, P.B., & Christensen, R.H.B. (2017). lmerTest package: Tests in
778 linear mixed effects models. *Journal of Statistical Software*, 82(13), 1-26.
779 <https://doi.org/10.18637/jss.v082.i13>
- 780 37. Lewis, W.M. (1983). A revised classification of lakes based on mixing. *Canadian Journal of*
781 *Fisheries and Aquatic Sciences*, 40(10): 1779-1787. <https://doi.org/10.1139/f83-207>
- 782 38. Lin, Q., Liu, E., Zhang, E., Bindler, R., Nath, B., Zhang, K., et al. (2022). Spatial variation of
783 organic carbon sequestration in large lakes and implications for carbon stock
784 quantification. *CATENA*, 208: 105768. <https://doi.org/10.1016/j.catena.2021.105768>
- 785 39. Lukkari, K., Hartikainen, H., & Leivuori, M. (2007). Fractionation of sediment phosphorus
786 revisited. I: Fractionation steps and their biogeochemical basis. *Limnology and*
787 *Oceanography: Methods*, 5: 433-444. <https://doi.org/10.4319/lom.2007.5.433>
- 788 40. Mackay, E.B., Jones, I.D., Folkard, A.M., & Barker, P. (2012). Contribution of sediment
789 focussing to heterogeneity of organic carbon and phosphorus burial in small lakes.
790 *Freshwater Biology*, 57(2): 290-304. <https://doi.org/10.1111/j.1365-2427.2011.02616.x>
- 791 41. Madsen, J.D., Chambers, P.A., James, W.F., Koch, E.W., & Westlake, D.F. (2001). The
792 interactions between water movement, sediment dynamics and submersed macrophytes.
793 *Hydrobiologia*, 444, 71-84. <https://doi.org/10.1023/A:1017520800568>
- 794 42. Mortimer, C. (1941). The exchange of dissolved substances between mud and water in lakes.
795 *Journal of Ecology*, 29, 280-239. <https://doi.org/10.2307/2256691>
- 796 43. North, R.L., Johansson, J., Vandergucht, D.M., Doig, L.E., Liber, K., Lindenschmidt, K., et
797 al. (2015) . Evidence for internal phosphorus loading in a large prairie reservoir (Lake
798 Diefenbaker, Saskatchewan). *Journal of Great Lakes Research* 41(S2): 91-99.
799 <https://doi.org/10.1016/j.jglr.2015.07.003>

- 800 44. Nowlin, W.H., Evarts, J.L., & Vanni, M.J. (2005). Release rates and potential fates of
801 nitrogen and phosphorus from sediments in a eutrophic reservoir. *Freshwater Biology*
802 *50*(2), 301–322. <https://doi.org/10.1111/j.1365-2427.2004.01316.x>
- 803 45. Nürnberg, G.K. (1988). Prediction of phosphorus release rates from total and reductant-
804 soluble phosphorus in anoxic lake sediments. *Canadian Journal of Fisheries and Aquatic*
805 *Science*, *45*, 453–462. <https://doi.org/10.1139/f88-054>
- 806 46. Oksanen, J., Blanchet, F.G., Friendly, M., Kindt, R., Legendre, P. McGlinn, D., et al. (2019).
807 vegan: Community Ecology Package. R package version 2.5-6. [https://CRAN.R-](https://CRAN.R-project.org/package=vegan)
808 [project.org/package=vegan](https://CRAN.R-project.org/package=vegan)
- 809 47. Orihel, D.M., Baulch, H.M., Casson, N.J., North, R.L., Parsons, C.T., Seckar, D.C.M, et al.
810 (2017). Internal phosphorus loading in Canadian fresh waters: a critical review and data
811 analysis. *Canadian Journal of Fisheries and Aquatic Science*, *74*, 2005-2029.
812 <https://doi.org/10.1139/cjfas-2016-0500>
- 813 48. Orihel, D. M., Schindler, D. W., Ballard, N. C., Graham, M. D., O'Connell, D. W., Wilson,
814 L. R., et al. (2015). The "nutrient pump:" Iron-poor sediments fuel low nitrogen-to-
815 phosphorus ratios and cyanobacterial blooms in polymictic lakes. *Limnology and*
816 *Oceanography*, *60*(3): 856-871. <https://doi.org/10.1002/lno.10076>
- 817 49. Ortiz, D.A., Wilkinson, G.M. (2021). Capturing the spatial variability of algal bloom
818 development in a shallow temperate lake. *Freshwater Biology*, *66*(11): 2064-2075.
819 <https://doi.org/10.1111/fwb.13814>
- 820 50. Ostrofsky, M.L. (2012). Differential post-depositional mobility of phosphorus species in lake
821 sediments. *Journal of Paleolimnology*, *48*, 559-569. [https://doi.org/10.1007/s10933-012-](https://doi.org/10.1007/s10933-012-9631-4)
822 [9631-4](https://doi.org/10.1007/s10933-012-9631-4)

- 823 51. Papera, J., Araújo, F., & Becker, V. (2021). Sediment phosphorus fractionation and flux in a
824 tropical shallow lake. *Acta Limnologica Brasiliensia*, 33: e5.
825 <https://doi.org/10.1590/S2179-975X9020>
- 826 52. Pease, L.A., King, K.W., Williams, M.R., LaBarge, G.A., Duncan, E.W., & Fausey, N.R.
827 (2018). Phosphorus export from artificially drained fields across the Eastern Corn Belt.
828 *Journal of Great Lakes Research*, 44(1), 43-53. <https://doi.org/10.1016/j.jglr.2017.11.009>
- 829 53. Psenner, R., & Puckso, R. (1988). Phosphorus fractionation: advantages and limits of the
830 method for the study of sediment P origins and interactions. *Archiv fur Hydrobiologie*,
831 30, 43–59. ISSN: 0071-1128.
- 832 54. R Core Team (2019). R: A language and environment for statistical computing. R
833 Foundation for Statistical Computing, Vienna, Austria. <https://www.R-project.org/>
- 834 55. Randall, M.C., Carling, G.T., Dastrup, D.B., Miller, T., Nelson, S.T., Rey, K.A., et al.
835 (2019). Sediment potentially controls in-lake phosphorus cycling and harmful
836 cyanobacteria in shallow, eutrophic Utah Lake. *PLoS ONE*, 14(2): e0212238.
837 <https://doi.org/10.1371/journal.pone.0212238>
- 838 56. Rippey, B., Anderson, N.J., Renberg, I., & Korsman, T. (2008). The accuracy of methods
839 used to estimate the whole-lake accumulation rate of organic carbon, major cations,
840 phosphorus and heavy metals in sediment. *Journal of Paleolimnology*, 39: 83–99.
841 <https://doi.org/10.1007/s10933-007-9098-x>
- 842 57. Schindler, D.W., Carpenter, S.R., Chapra, S.C., Hecky, R.E., & Orihel, D.M. (2016).
843 Reducing phosphorus to curb lake eutrophication is a success. *Environmental Science &*
844 *Technology*, 50(17), 8923-8929. <https://doi.org/10.1021/acs.est.6b02204>

- 845 58. Schindler, D.W., & Vallentyne, J.R. (2008). *The Algal Bowl: Overfertilization of the World's*
846 *Freshwaters and Estuaries*. Edmonton, AB: The University of Alberta Press.
- 847 59. Song, K., & Burgin, A.J. (2017). Perpetual phosphorus cycling: Eutrophication amplifies
848 biological control on internal phosphorus loading in agricultural reservoirs. *Ecosystems*,
849 20, 1483-1493. <https://doi.org/10.1007/s10021-017-0126-z>
- 850 60. *Standard Methods for the Examination of Water and Wastewater*, 20th Edition, Method 4500-
851 P B.5 (1998).
- 852 61. *Standard Methods for the Examination of Water and Wastewater*, 20th Edition, Method 4500-
853 P E (1998).
- 854 62. *Standard Methods for the Examination of Water and Wastewater*, 20th Edition, Method
855 10200-H (1998).
- 856 63. Stone, M., & English, M.C. (1993). Geochemical composition, phosphorus speciation and
857 mass transport of fine-grained sediment in two Lake Erie tributaries. *Hydrobiologia* 253,
858 17-29. <http://doi.org/10.1007/BF00050719>
- 859 64. Søndergaard, M., Jensen, J.P., & Jeppesen, E. (1999). Internal phosphorus loading in shallow
860 Danish lakes. *Hydrobiologia*, 408/409, 145-152.
861 <https://doi.org/10.1023/A:1017063431437>
- 862 65. Søndergaard, M., Jensen, J.P., & Jeppesen, E. (2003). Role of sediment and internal loading
863 of phosphorus in shallow lakes. *Hydrobiologia*, 506-509, 135-145.
864 <https://doi.org/10.1023/B:HYDR.0000008611.12704.dd>
- 865 66. Søndergaard, M., Bjerring, R., & Jensen, J.P. (2013). Persistent internal phosphorus loading
866 during summer in shallow eutrophic systems. *Hydrobiologia*, 710 (1), 95-107.
867 <http://doi.org/10.1007/s10750-012-1091-3>

- 868 67. Tammeorg, O., Möls, T., Niemistö, J., Holmroos, H., & Horppila, J. (2017). The actual role
869 of oxygen deficit in the linkage of the water quality and benthic phosphorus release:
870 Potential implications for lake restoration. *Science of the Total Environment*, 599-600:
871 732-738. <http://dx.doi.org/10.1016/j.scitotenv.2017.04.244>
- 872 68. Tang, Q., Peng, L., Yang, Y., Lin, Q., Qian, S.S., & Han, B. (2019). Total phosphorus-
873 precipitation and chlorophyll α -phosphorus relationships of lakes and reservoirs mediated
874 by soil iron at regional scale. *Water Research*, 154: 136-143.
875 <http://doi.org/10.1016/j.watres.2019.01.038>
- 876 69. Tao, Y.Q., & Lu, J. (2020). Occurrence of total phosphorus in surface sediments of Chinese
877 lakes and its driving factors and implications. *Journal of Hydrology*, 580: 124345.
878 <https://doi.org/10.1016/j.jhydrol.2019.124345>
- 879 70. Trolle, D., Zhu, G., Hamilton, D., Luo, L., McBride, C., and Zhang, L. (2009). The influence
880 of water quality and sediment geochemistry on the horizontal and vertical distribution of
881 phosphorus and nitrogen in sediments of a large, shallow lake. *Hydrobiologia* 627:31-44.
882 <https://doi.org/10.1007/s10750-009-9713-0>
- 883 71. Twilley, R.R., Ejdung, G., Romare, P., & Kemp, W.M. (1986). A comparative study of
884 decomposition, oxygen consumption and nutrient release for selected aquatic plants
885 occurring in an estuarine environment. *Oikos*, 47, 190-198.
886 <https://doi.org/10.2307/3566045>
- 887 72. Welch, E.B., & Cooke, G.D. (1995). Internal phosphorus loading in shallow lakes:
888 Importance and control. *Lake and Reservoir Management*, 11(3), 273-281.
889 <http://doi.org/10.1080/07438149509354208>

- 890 73. White, A., & Stone, M. (1996). Spatial variation and distribution of phosphorus forms in
891 bottom sediments of two Canadian Shield Lakes. *Canadian Geographer*, 40(3): 258-265.
892 <https://doi.org/10.1111/j.1541-0064.1996.tb00452.x>
- 893 74. Wickham, H., Averick, M., Bryan, J., Chang, W., D'Agostino McGowan, L., François, R., et
894 al. (2019). Welcome to the tidyverse. *Journal of Open Source Software*, 4(43), 1686.
895 <https://doi.org/10.21105/joss.01686>
- 896 75. Xing, X.G., Ding, S.M., Lui, L., Chen, M.S., Yan, W.M., Zhao, L.P., et al. (2018). Direct
897 evidence for the enhanced acquisition of phosphorus in the rhizosphere of aquatic plants:
898 A case study on *Vallisneria natans*. *Science of the Total Environment*, 616, 386-396.
899 <https://doi.org/10.1016/j.scitotenv.2017.10.304>
- 900 76. Yu, Z., & Lu, C. (2018). Historical cropland expansion and abandonment in the continental
901 U.S. during 1850 to 2016. *Global Ecology and Biogeography*, 27(3), 322-333.
902 <https://doi.org/10.1111/geb.12697>

903 **Supporting References**

- 904 77. Allan, R., & Williams, J. D. H. (1978). Trophic status related to sediment chemistry of
905 Canadian prairie lakes. *Journal of Environmental Quality*, 7: 99–106.
906 <https://doi.org/10.2134/jeq1978.00472425000700010020x>
- 907 78. Allan, R. J., Williams, J. D. H., Joshi, S. R., & Warwick, W. F. (1980). Historical changes
908 and relationship to internal loading of sediment phosphorus forms in hypertrophic prairie
909 lakes. *Journal of Environment Quality*, 9: 199–206.
910 <https://doi.org/10.2134/jeq1980.00472425000900020007x>
- 911 79. Lucas, B. T., Karsten, L., & Doig, L. E. (2015). Spatial and temporal trends in reservoir
912 physicochemistry and phosphorus speciation within Lake Diefenbaker, a Great Plains

- 913 reservoir, as inferred from depositional sediments. *Journal of Great Lakes Research*,
914 41(Suppl. 2): 67–80. <https://doi.org/10.1016/j.jglr.2015.07.009>
- 915 80. Lucas, B. T., Liber, K., & Doig, L. E. (2015). Reconstructing diatom and chironomid
916 assemblages to infer environmental spatiotemporal trends within Lake Diefenbaker, a
917 narrow river valley reservoir on the Canadian Prairies. *Journal of Great Lakes Research*,
918 41(Suppl. 2): 45–55. <https://doi.org/10.1016/j.jglr.2015.09.003>
- 919 81. Mayer, T., Simpson, S. L., Thorleifson, L. H., Lockhart, W. L., & Wilkinson, P. (2006).
920 Phosphorus geochemistry of recent sediments in the south basin of Lake Winnipeg.
921 *Aquatic Ecosystem Health & Management*, 9: 307–318.
922 <https://doi.org/10.1080/14634980600876039>
- 923 82. Mikulyuk, A., Sharma, S., Van Egeren, S., Erdmann, E., Nault, M.E., & Hauxwell, J. (2011).
924 The relative role of environmental, spatial, and land-use patterns in explaining aquatic
925 macrophyte community composition. *Canadian Journal of Fisheries and Aquatic
926 Sciences*, 68(10), 1778-1789. <https://doi.org/10.1139/f2011-095>

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Supporting Information for

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

932

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942

Introduction

943

The supporting information contains detailed explanations of the equations used to determine

944

the sediment physical characteristics and the concentrations of sediment phosphorus (P)

945

fractions (Text S1) as well as a description of the point-intercept method used to sample aquatic

946

macrophytes (Text S2). Table S1 details the frequency of thermal stratification in the study lakes

947

as well as dissolved oxygen conditions at the sediment-water interface. Tables S2 and S3 contain

948

more detailed information on watershed soil series and landcover respectively. Table S4

949

compares the total and loosely-bound P concentrations at the deep site of each study lake to

950

the whole-lake mean values. Table S5 summarizes how sediment total P concentrations in our

951

study lakes compare to other values in the literature. Figure S1 illustrates 13-19 years of TN:TP

952

ratios for the study lakes to demonstrate that the lakes are weakly to strongly phosphorus-

953

limited on average. We have also included a visualization of the relationship between sediment

954

P pools and macrophytes across Swan Lake (Figure S2).

955 **Text S1.**956 *Eq. 1 Moisture Content (MC)*

957
$$\text{Moisture Content (\%)} = \left[\frac{(W_w - W_t) - (W_d - W_t)}{W_w - W_t} \right] \times 100 \quad (1)$$

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

960

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

962
$$\text{LOI Organic Matter Content (\%)} = \left[\frac{(W_d - W_t) - (W_a - W_t)}{W_d - W_t} \right] \times 100 \quad (2)$$

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

964

965 *Eq. 3 – Bulk Density*

966
$$\text{Bulk Density (g/cm}^3\text{)} = \frac{260}{100 + 1.6 \times \left[\text{MC} + \left(\frac{\text{LOI}}{100 \times (100 - \text{MC})} \right) \right]} \quad (3)$$

967

968 *Eq. 4 – Dry Mass Equivalent of Fresh Sediment Used*

969
$$\text{Dry Mass Equivalent (g)} = \text{Mass Fresh Sediment (g)} \times (100 - \text{MC}) \quad (4)$$

970

971 *Eq. 5 – Loosely sorbed and pore water P*

972
$$\text{Loosely-Bound P (mg P/g dry sediment)} = \frac{\text{Concentration SRP (mg/L)} \times \text{Extractant Volume (L)}}{\text{Dry Mass Equivalent of Sediment Used (g)}} \quad (5)$$

973 The concentration of SRP used should reflect the average of lab duplicate measures. The
974 extractant volume should equal the total volume of 1M NH_4Cl used across both extractions,
975 (0.05 L). The dry mass equivalent of the fresh sediment used is estimated based on MC (Eq. 4)
976 and will be the same for the calculations of each subsequent extraction (Eq. 6-9).

977

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

979
$$\text{Redox-Sensitive P (mg P/g dry sediment)} = \frac{\text{Concentration SRP (mg/L)} \times \text{Extractant Volume (L)}}{\text{Dry Mass Equivalent of Sediment Used (g)}} \quad (6)$$

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

983

984 *Eq. 7 – Aluminum-Bound P*

985
$$\text{Al-Bound P (mg P/g dry sed.)} = \frac{\left[\text{Concentration SRP} \left(\frac{\text{mg}}{\text{L}} \right) \times \frac{(\text{Post pH (g)} - \text{Tare (g)})}{(\text{Pre pH (g)} - \text{Tare (g)})} \right] \times \text{Extractant Volume (L)}}{1.00152 \times \text{Dry Mass Equivalent of Sediment Used (g)}} \quad (7)$$

986 The concentration of SRP used should reflect the average of lab duplicate measures and must
987 be corrected for the pH adjustment. Tare is the mass of the I-chem jar in which the adjustment is
988 performed; Pre pH is the mass of the jar and the extraction supernatant; and Post pH is the mass
989 of the jar, supernatant, and titrant used for the pH adjustment. The constant 1.00152 is used for
990 0.1 M HCl as the titrant. The corrected SRP concentration can then be corrected for the
991 extractant volume (0.025 L) and sediment mass.

992

993 *Eq. 8 – Labile Organic P*

994
$$\text{Labile Organic P (mg P/g dry sed.)} = \left[\frac{\text{Concentration SRP, digested (mg/L)} \times \text{Extractant Volume (L)}}{\text{Dry Mass Equivalent of Sediment Used (g)}} \right] - \text{Al-Bound P} \quad (8)$$

995 The concentration of SRP from the *digested* supernatant should first be corrected for the volume
 996 of NaOH used (0.05 L) and the sediment mass. This value represents the concentration of
 997 aluminum-bound and labile organic P in the sediment pellet. The concentration of labile organic
 998 P is calculated as the difference between this value and the Al-bound P concentration (Eq. 7).
 999

1000 *Eq. 9 – Calcium-Bound P*

1001
$$\text{Ca-Bound P (mg P/g dry sed.)} = \frac{\left[\text{Concentration SRP} \left(\frac{\text{mg}}{\text{L}} \right) \times \frac{(\text{Post pH (g)} - \text{Tare (g)})}{(\text{Pre pH (g)} - \text{Tare (g)})} \right] \times \text{Extractant Volume (L)}}{1.00452 \times \text{Dry Mass Equivalent of Sediment Used (g)}} \quad (9)$$

1002 The concentration of SRP used should reflect the average of lab duplicate measures and must
 1003 be corrected for the pH adjustment. The constant 1.00452 is used for 0.1 M NaOH as the titrant.
 1004 The corrected SRP concentration can then be corrected for the extractant volume (0.025 L) and
 1005 sediment mass.
 1006

1007 *Eq. 10 – Total P*

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

1009 The concentration of TP used should reflect the average of lab duplicate measures and must be
 1010 corrected for the pH adjustment. The constant 1.00452 is used for the 0.1 M NaOH as the titrant.
 1011 This corrected SRP concentration can then be corrected for the volume to which the sample was
 1012 diluted after boiling (0.05 L) and the mass of dry sediment used.
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1014 **Text S2.**

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1016 ***Point intercept method for sampling aquatic macrophytes***

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1018 We followed a grid of 98 sampling sites, evenly-spaced 65 m apart across the lakebed (Ortiz &
1019 Wilkinson, 2021). At each sampling point, a two-side rake was lowered to the bottom and used
1020 to collect submersed and floating-leaf macrophytes from an approximately 0.4 m² area
1021 (Mikulyuk et al., 2011). Species presence and a qualitative estimate of density were recorded at
1022 each sampling site. We scored bed density on a scale of 0-3 based on whether the site yielded
1023 no plants on the sampling rake or in sight of the boat (score of 0), a few plants on the rake or
1024 visible within 2 m of the boat (score of 1), enough plants to fill the rake but leave the tines
1025 visible (score of 2), or enough plants to completely fill the rake leaving no part of the metal rake
1026 head visible (score of 3).

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1028 ***References***

1029 Mikulyuk, A., Sharma, S., Van Egeren, S., Erdmann, E., Nault, M.E., & Hauxwell, J. (2011). The
1030 relative role of environmental, spatial, and land-use patterns in explaining aquatic
1031 macrophyte community composition. *Canadian Journal of Fisheries and Aquatic Sciences*,
1032 68(10), 1778-1789. <https://doi.org/10.1139/f2011-095>

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

Lake Name	Frequency of Summer Thermal Stratification (%)		Dissolved Oxygen at Sediment-Water Interface (mg L ⁻¹)
	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

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* 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.

1070 **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

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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

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Table S4. RMSE of deep site P concentrations

Lake	Total P ($\mu\text{g P g dw}^{-1}$)				Loosely-Bound P ($\mu\text{g P g dw}^{-1}$)			
	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

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1078 **Table S5.** Literature comparison of sediment total P concentrations

			Whole-lake Total P across study lakes ($\mu\text{g P g}^{-1}\text{ dw}$)	
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
Kowalczywska-Madura et al., 2019b	2 meso-eutrophic lakes	Poland	840 - 1,300	1,072

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1080 * Includes results from five other studies:

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1082 Allan, R., & Williams, J. D. H. (1978). Trophic status related to sediment chemistry of Canadian
1083 prairie lakes. *Journal of Environmental Quality*, 7: 99–106.1084 <https://doi.org/10.2134/jeq1978.00472425000700010020x>

1085

1086 Allan, R. J., Williams, J. D. H., Joshi, S. R., & Warwick, W. F. (1980). Historical changes and
1087 relationship to internal loading of sediment phosphorus forms in hypertrophic prairie
1088 lakes. *Journal of Environment Quality*, 9: 199–206.1089 <https://doi.org/10.2134/jeq1980.00472425000900020007x>

1090

1091 Lucas, B. T., Karsten, L., & Doig, L. E. (2015). Spatial and temporal trends in reservoir
1092 physicochemistry and phosphorus speciation within Lake Diefenbaker, a Great Plains
1093 reservoir, as inferred from depositional sediments. *Journal of Great Lakes Research*,
1094 41(Suppl. 2): 67–80. <https://doi.org/10.1016/j.jglr.2015.07.009>

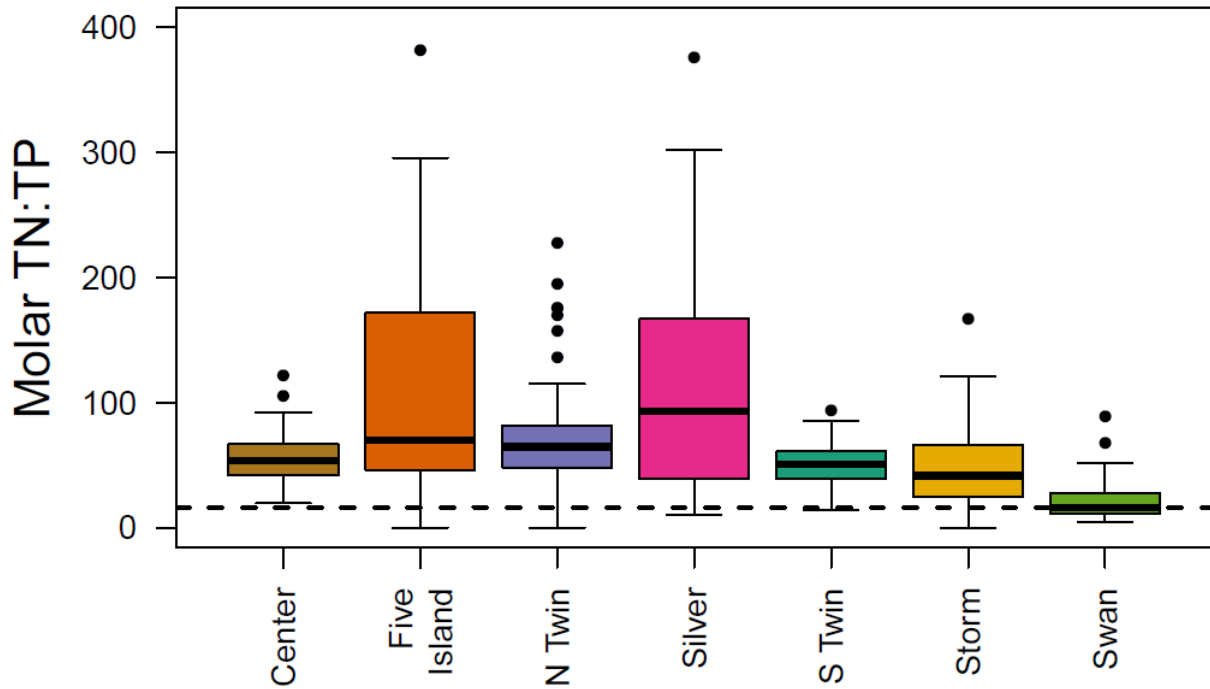
1095

1096 Lucas, B. T., Liber, K., & Doig, L. E. (2015). Reconstructing diatom and chironomid
1097 assemblages to infer environmental spatiotemporal trends within Lake Diefenbaker, a
1098 narrow river valley reservoir on the Canadian Prairies. *Journal of Great Lakes Research*,
1099 41(Suppl. 2): 45–55. <https://doi.org/10.1016/j.jglr.2015.09.003>

1100

1101 Mayer, T., Simpson, S. L., Thorleifson, L. H., Lockhart, W. L., & Wilkinson, P. (2006).
1102 Phosphorus geochemistry of recent sediments in the south basin of Lake Winnipeg.
1103 *Aquatic Ecosystem Health & Management*, 9: 307–318.1104 <https://doi.org/10.1080/14634980600876039>

1105



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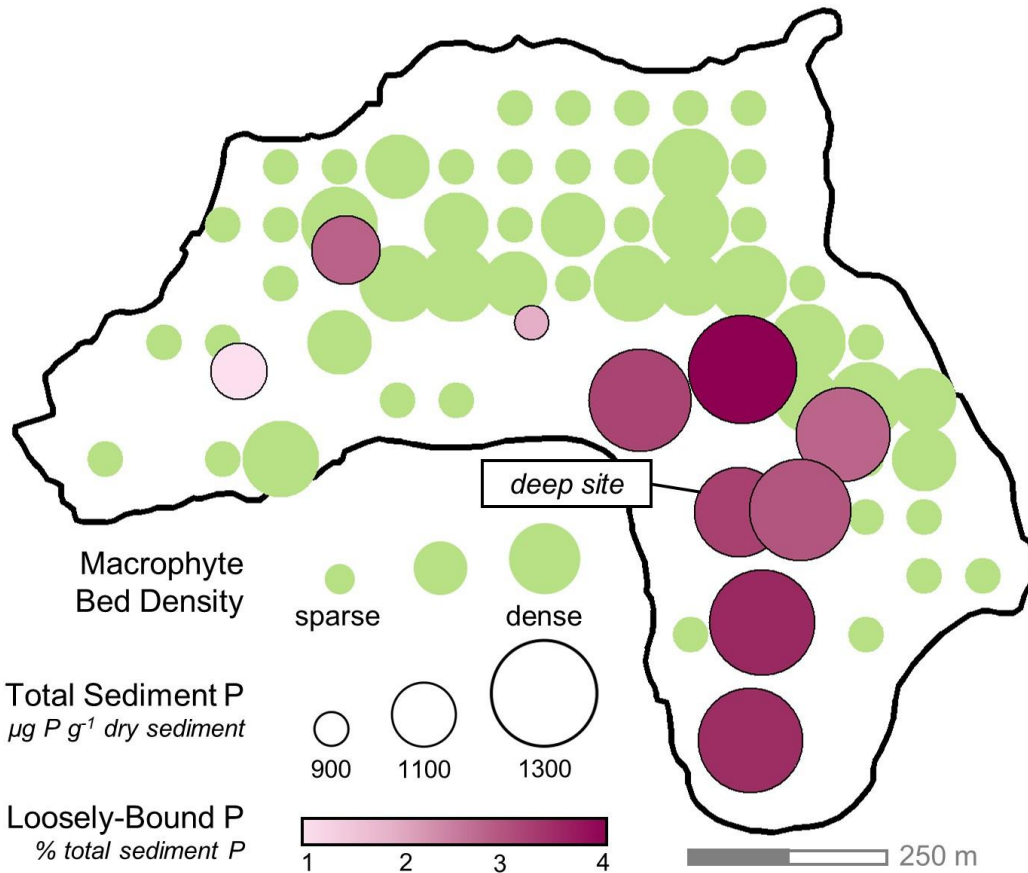
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Figure S1. Long-term molar TN:TP ratios across the study lakes. The values represent 19 (2000-2018) years of data during the ice-free season, with the exception of South Twin Lake, which represents 13 years of measurements. Swan Lake is weakly phosphorus-limited on average. Center, Storm, North Twin, and South Twin Lakes are moderately phosphorus-limited, and Five Island and Silver Lakes are strongly phosphorus-limited.



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Figure S2. 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*.