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New Insights Into Legacy Phosphorus From Fractionation of Streambed Sediment

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Abstract Streambed and lake sediment was studied in Sävjaån, a eutrophic mesoscale catchment (722 km^2) in central Sweden. Triplicate sediment cores from five lakes and nine streams, ranging from headwater to fourth order, were sampled. The sediment was analyzed with a sequential extraction method, where six different phosphorus (P) fractions were measured. The results showed that streambed sediments store considerable amounts of P and in some cases have comparable P contents (g/kg DW) to lake sediment. Land use, soil type, and drainage area (location in the catchment) had a significant effect on the different P fractions found in surficial sediments. Sediment from lakes and forested headwater streams generally had high proportions of organic P and iron bound P. In larger streams located in agricultural areas on clay soils closer to the catchment outlet, with dense sediment and a relatively low proportion of organic matter, P was to a larger extent bound to calcium. Streambed sediment may be an important catchment scale P store and should be considered when modeling catchment P dynamics. The large stores of streambed legacy P should also be considered when performing ditch maintenance to avoid unnecessary mobilization of bioavailable P.

1. Introduction

Cultural eutrophication is responsible for deleterious environmental, social, and economic problems on both local and regional levels (Smith & Schindler, 2009; Withers & Jarvie, 2008). Major advances in understanding and managing eutrophication have been made during the last half century (Schindler, 2006; Smith & Schindler, 2009). While reductions in phosphorus (P) inputs have successfully controlled eutrophication in some cases (Smith & Schindler, 2009), it is not always enough for recovery of water bodies (Reitzel et al., 2005). Phosphorus stored in the catchment land-water continuum (legacy P) can delay improvements in water quality despite reductions in P input (Jeppesen et al., 2007; Sharpley et al., 2013; Søndergaard et al., 1996). Legacy P (Kleinman et al., 2011) accumulates in ditches (Shore et al., 2016), wetlands (Geranmayeh et al., 2018), riparian soils (Fox et al., 2016), lake sediment (Søndergaard et al., 2003), and streambed sediment (Withers & Jarvie, 2008). These P stores can, under certain conditions, be released and contribute to internal loading by biogeochemical release from the sediment or physical processes mobilizing material for transport further downstream (Palmer-Felgate et al., 2009; Søndergaard et al., 2003; Withers & Jarvie, 2008).

The importance of internal loading of P from lake sediment is well established (Mortimer, 1942; Schindler, 2006; Søndergaard et al., 2003). Yet, on a catchment level, streams are also important for transport and fate of particulate P (Noll et al., 2009; Owens & Walling, 2002; Sandström et al., 2020) and as P stores (Jarvie et al., 2005; Taube et al., 2019). Studies looking at the sediment Equilibrium Phosphorus Concentration (EPC$_0$) in streambed sediment conclude that the P stored is likely available as a P source (Hongthanat et al., 2016; Weigelhofer et al., 2018), for example, when concentrations in the sediment are higher than the dissolved P in the water column and during certain times of the year (Hongthanat et al., 2016). Phosphorus stored in streambed sediment may be a missing link between diffuse P sources and subsequent impacts from excess P at ecologically sensitive times, for example, summer low flows (Jarvie et al., 2005; Svendsen et al., 1995). Any additional mobilization of streambed sediment P caused by disturbances (e.g., dredging) during these ecologically sensitive times could have disproportionate effects on aquatic biota. Despite this, the legal system in Sweden maintains old "drainage permits," often approved 100 years ago. These permits give the land owner the right and the responsibility to maintain the same depth and position of the ditch over time. In practical terms, this translates to repeatedly digging out the newly

deposited bed sediment, usually with heavy machinery (The Swedish EPA, 2009). According to the Swedish Environmental Code the permit owner does not have to notify the authorities before dredging streams/ditches, except for when the works can be detrimental to fishing (Government Offices of Sweden, 2020).

Sediment P stores have a profound effect on P transport through catchments (Svendsen et al., 1995) and the P exchange between the sediment and overlying water column is a dynamic process (Syers et al., 1973). Sediment interactions are highly important for P dynamics, for example, sorption/desorption and mineral precipitation/dissolution (House, 2003). The availability of P stored in the sediment depends on, for example, if it is sorbed to hydroxides or minerals or contained in molecules (organic or mineral lattices) combined with physiochemical properties in the surrounding environment (Reddy et al., 1996; Søndergaard et al., 1996; Syers et al., 1973).

The sediment composition, and what P is associated with, affects the P dynamics. In sediments, P can be sorbed to iron (Fe) and aluminum (Al) (hydr)oxides, as well as calcium carbonates depending on the pH (Reddy & DeLaune, 2008). Dynamics of loosely bound P can be affected by sediment pore water and water column P concentrations (Reddy et al., 1996), as well as turbulence and discharge (Fuchs et al., 2009; Reddy et al., 1996). When P is associated with Fe, organic matter mediated redox processes can be important for bioavailability (Records et al., 2016). Aluminum bound P is considered stable within the pH range of surface water (Stumm & Morgan, 1995). However, availability of Al bound P depends on if it is associated with amorphous molecules (i.e., active Al (hydr)oxides) or present in an occluded form (crystalline Al minerals) (de Vicente et al., 2008; Jan et al., 2015; Lijklema, 1980). Phosphorus is also present in organic molecules of living and dead biota (Lukkari et al., 2007). The size and structure of organic molecules affect their resistance to hydrolysis and degradation; microbial activity and pH will also influence the bioavailability of organic P (Lukkari et al., 2007). Phosphorus can also be contained in a variety of mineral lattices, for example, apatite $(Ca_5(PO_4)_3(F,Cl,OH))$, variscite $(AlPO_4 \cdot 2\ H_2O)$, and strengite $(FePO_4 \cdot 2\ H_2O)$ (Stumm & Morgan, 1995), all characterized by low bioavailability (Lukkari et al., 2007).

Internal loading from lake sediments is often controlled by the combination of Fe bound P (Christophoridis & Fytianos, 2006) and more or less labile organic material (Søndergaard et al., 2003) with prevailing environmental conditions. Sediment pore water and water column chemistry, including porewater P concentration (Reddy et al., 1996), oxygen conditions (Moosmann et al., 2006), and pH (Boström et al., 1988; Christophoridis & Fytianos, 2006; Zhou et al., 2005) are important for sediment water P exchange. In streambed sediment, P storage is primarily influenced by hydrological and physical processes that drive mobilization and transport of P in the catchment (Dorioz et al., 1998; Palmer-Felgate et al., 2009; Søndergaard et al., 1992). Even so, biogeochemical processes like sorption and precipitation can be important also in streams (Records et al., 2016).

To gain a better understanding about the magnitude and potential mobility of P stored in sediments, sequential chemical extraction methods (i.e., P fractionation) are often used. The Psenner and Pucsko (1988) P fractionation scheme identifies loosely bound P (MQ-P), redox sensitive P bound to, for example, Fe/Mn (BD1, BD2), nonreducible P bound to, for example, Al (hydr)oxides $(NaOH_{RP})$, organic P $(NaOH_{NRP})$, and calcium (Ca) bound P (HCl-P). Results from sediment P fractionation are time-integrated, where effects from year to year and seasonal variation are minimized (Søndergaard et al., 1996). Sequential extraction has been widely used to identify P fractions, and their potential mobility, in lake sediments (Boström et al., 1988; Rydin, 2000; Søndergaard et al., 1996; Zhang et al., 2013). In a few cases, sequential extraction has also been used for streambed sediment (Audette et al., 2018; Noll et al., 2009; SanClements et al., 2009).

The aim of this study was to determine the magnitude and potential mobility of legacy P stored in lake and stream sediments in a nutrient-impacted, 722 km^2 mesoscale catchment in central Sweden. Both the distribution of different P fractions as well as variation by depth in individual sediment profiles were determined to gain an understanding about the availability and potential mobility of legacy P. The study included sediment from lakes and headwater to fourth-order streams with different land use and soil types. The three research questions addressed in this study aimed to (i) quantify the TP content and concentrations stored in lake and streambed sediments in the catchment, (ii) measure and compare the P fractions present in lake and streambed sediments, and (iii) identify factors controlling both the amount and fractional composition of sediment P. We hypothesized that (i) similar amounts of legacy P are stored in lake and streambed
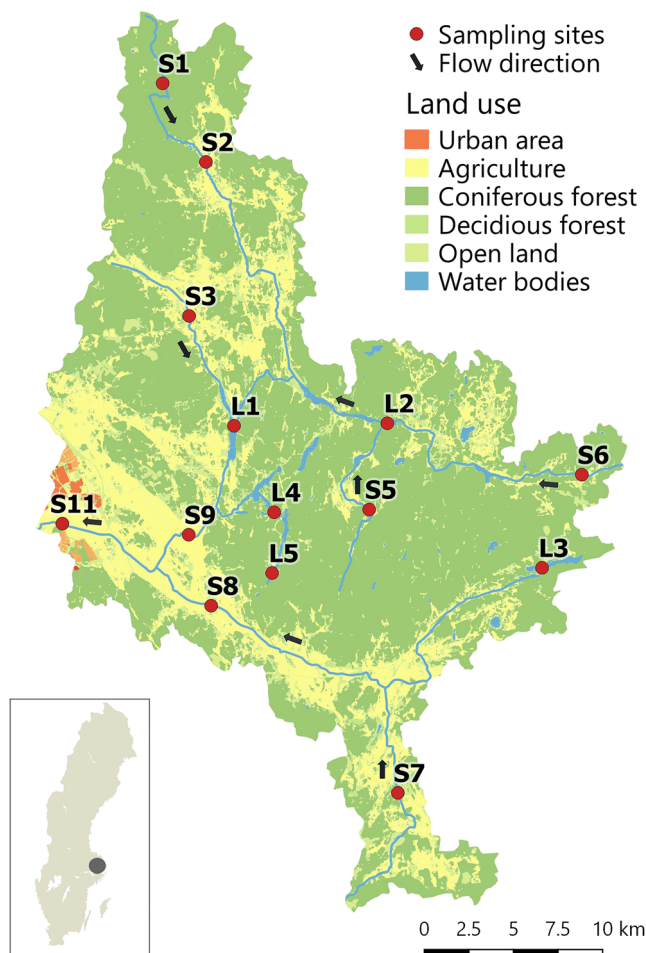
Figure 1. Sävjaån catchment with sampling sites of streams (S) and lakes (L). Flow direction in the catchment is indicated by arrows and different land use by different colors. ©The National Land Survey.

sediment and (ii) P stores would accumulate in greater concentrations downstream in the catchment and that accumulation rates would be affected by land use.

2. Materials and Methods

2.1. Study Site

Lakes and streams in Sävjaån, a mixed land use catchment in east-central Sweden, were studied. Nearly 50% of the lakes and streams are regarded as eutrophic in the area; hence excessive nutrients are a major environmental problem (Vattenmyndigheten Norra Östersjön, 2016). The Sävjaån catchment covers 722 km^2 with forest (70%), agriculture (28%), urban area (1%), and lakes (1%) (Vattenwebb - Mätningar, 2019). In agricultural areas the most common crops are winter wheat (17%) and spring barley (11%); some land is also used for ley (14%) and fallow (6%) (Hansson et al., 2019). The catchment is generally flat, and the difference between the highest and lowest points is 70 m (The National Land Survey, 2020). Surficial geology in the catchment is dominated by sandy till, postglacial/glacial clay, and bare rock. The underlying bedrock consists mostly of granitoids and syenitoids (Geographical Survey of Sweden, 2020) (Maps of soil type and bedrock are found in the supporting information Figures S1 and S2). In the area, the generally Ca rich glacial clay can contain 10–30% calcite (CaCO$_3$) (Möller, 1993), and glacial/postglacial clay soils have a clay content between 40–70% (Möller, 1993). Agriculture dominates on clay soils while forests are more common on the slightly higher elevation outwash till soils (Lannergård et al., 2019). Average annual precipitation is 639 mm, and average annual runoff is 189 mm (1981–2010) (Vattenwebb - Modelldata per område, 2019). Flows are not managed and are generally flashier during spring. In most years there is a spring runoff peak associated with snowmelt. Winter flows are sustained by groundwater and increasingly common winter rainfall and snowmelt events. During summer the flow is low, while the autumn and winter can generate higher and flashier flows (Figure S3). The streams are often covered with ice for a few months each year. In the 12 months prior to sampling (June 2016–May 2017), discharge was lower than the long term average (1981–2019), the spring flow peak occurred in March 2017. Mean annual temperature is 6°C, with a range of average daily temperatures between −27°C and +26°C (1949–2017). In the catchment, 640 km streams and ditches are managed and have a "drainage permit" allowing drainage according to Swedish legislation (County Administrative Board of Uppsala, 2020; Figure S4). For the sampled streams (Figure 1) 63% of the total stream length is associated with a drainage permit.

Five lakes and nine streams in the catchment were chosen based on the following criteria: location in the catchment, stream order (first to fourth), draining of different parts of the catchment, representativeness of land use in the nearby area (detailed presentation in Table S1), and accessibility (Figure 1, Tables 1 and 2).

2.2. Sediment and Surface Water Sampling

Sampling of all sites was conducted in May 2017. Field protocols are presented in Table S2. All equipment was washed in water from the sampling location before use. Water samples were collected with a Ruttner water sampler in lakes and a Fyris water sampler in streams, consisting of a bottle attached to a rod. Visibility was estimated with a secchi disk, and profiles for temperature and oxygen (YSI ProODO) were recorded at the time of sediment sampling in both lakes and streams. Triplicate sediment cores were collected from each location. Cores were sectioned on site and stored cold (8°C) and dark in airtight cans until laboratory analysis.

Table 1
Site Information for Studied Streams (S1–S11)

Site	Site name	Stream order	Stream width (m)	Stream depth (m)	Status WFD	Drainage area (km^2)
S1	Lejstaån	1st	2.0	0.5	Moderate (N, HM)	1.1
S2	Ingvastra	1st	1.5	0.9	Moderate (N, HM)	46
S3	Tomtaån	1st	2.5	0.8	Moderate (N, HM)	30
S5	Dammtorpet	1st	2.0	0.5	N.D	16
S6	Stora hopan	1st	2.0	0.5	Moderate (HM)	7.3
S7	Vidboån	2nd	3.0	0.5	Moderate (HM)	32
S8	Storån	3rd	6.0	1.0	Moderate (N, HM)	180
S9	Funboån	3rd	10	1.0	Moderate (N, HM)	460
S11	Sävjaån Kuggebro	4th	12	2.0	Moderate (N, HM)	700

Note. Stream width and stream depth are an approximation from field studies, and status according to the Water Framework Directive was given from Water Information System Sweden (2019) (N = excessive nutrients, HM = hydromorphological problems, N.D = no data).

2.2.1. Lakes

Triplicate sediment cores were taken from the deepest area of the lake by boat, with a Willner gravity corer. Some distance was left between replicates to make sure the top sediment was not disturbed. Cores were sectioned at 1 cm intervals between 0–2 cm and at 2 cm intervals thereafter.

2.2.2. Streams

Sampling locations were chosen based on U.S. Geological Survey guidelines for sampling streambed sediment (Shelton & Capel, 1994). Streams were perennial, and sampling stations were at least 10 m away from bridges or roads to avoid direct contamination. At the time of sampling, streams had a water depth of at least 0.5 m. Sediment cores were collected from areas without stones or rooted vegetation. Cores were collected midchannel to avoid collapsed side banks. Thus, samples represented the streambed rather than riparian accumulation zones. Triplicate samples were taken within 2 m of each other, in most locations wading upstream so as not to disturb the sediment. While wading, the samples were taken with a Willner gravity sediment-coring device on a rod or by simply pushing the tube in manually with two plugs creating a vacuum. All samples were taken before extensive growth of rooted vegetation occurred in the streams. Discharge conditions were low at the time of sampling for all locations (1.28–1.78 m^3/day at outlet, lower 25th percentile 1.02 m^3/day 1979–2019). The samples were sectioned on site at 1 cm intervals.

2.3. Laboratory Analysis

All water chemistry analyses were performed by the accredited geochemistry laboratory at the Swedish University of Agricultural Sciences (SLU) (ISO/IEC 17025). Unfiltered orthophosphate (PO$_4$-P) was analyzed according to ISO 15923-1:2013. Unfiltered total P was analyzed using a potassium peroxidisulfate digestion followed by the ammonium molybdate blue spectrometric method (SS-EN ISO 6878:2005). In this study, definitions of P fractions follow Haygarth and Sharpley (2000) and unfiltered orthophosphate (PO$_4$-P) will hereafter be called reactive P (RP).

Table 2
Site Information for Studied Lakes (L1–L5)

Site	Site name	Mean, max depth (m)	Volume (mm^3)	Residence time (days)	Status (WFD)	Drainage area (km^2)	Lake area (km^2)	D/L ratio
L1	Funbosjön	1.7/5.5	3.5	114	Poor (N)	380	2.07	184
L2	Östra Långsjön	3.4/8.2	2.2	24	Moderate (N, HM)	109	0.64	170
L3	Ubby Långsjön	3.0/6.3	2.0	N.D.	Moderate (N)	19	1.10	17
L4	Trehörningen	3.2/6.2	4.7	295	Moderate (N)	21	1.46	14
L5	Edasjön	3.0/4.8	0.5	186	Moderate (N, HM)	2.6	0.17	15

Note. Mean, max depth, volume and residence time and lake area were collected from Brunberg and Blomqvist (1998) (N.D = no data). Status was according to the Water Framework Directive from Water Information System Sweden (2019) (N = excessive nutrients, HM = hydromorphological problems). Drainage area was determined by a GIS analysis from the Cadastrial map (The National Land Survey, 2020); D/L ratio represents the drainage to lake area ratio.

Table 3
Scheme for Sequential Fractionation of P

Fraction	Extractant	Duration	Expected P fractions	Hereafter called
MQ-P	H_2O, double de-ionized water	2 hr	P in pore water, loosely bound to surfaces (e.g., Fe (hydr)oxides, $CaCO_3$).[a]	H_2O-P
BD1-P	$Na_2SO_3/NaHCO_3$ (0.11 mol/L), pH 7.2	10 min	Redox sensitive P mainly bound to amorphous Fe (hydr)oxides.[b]	Fe_1-P
BD2-P	$Na_2SO_3/NaHCO_3$ (0.11 mol/L), pH 7.2	1 hr	Redox sensitive P mainly bound to more crystalline Fe (hydr) oxides.[b]	Fe_2-P
NaOH$_{RP}$	NaOH (1 mol/L)	16 hr	Phosphorus exchangeable against OH^- ions (e.g., P sorbed to Al (hydr)oxides), P compounds soluble in bases.[a]	Al-P
NaOH$_{NRP}$	Addition of potassium persulfate ($K_2S_2O_8$) and autoclaved. Difference between digested NaOH extract and NaOH$_{RP}$	(see NaOH$_{RP}$)	Phosphorus in microorganisms and organic matter (poly-P, organic P in detritus, P bound in humic compounds).[a]	Org-P
HCl-P	HCl (0.5 mol/L)	16 hr	Calcium phosphate minerals, acid-soluble organic P.[a]	Ca-P

[a]Adapted from Hupfer et al. (2009). [b]Adapted from Jan et al. (2015).

A sequential chemical extraction method for wet sediment based on Psenner and Pucsko (1988), modified by Hupfer et al. (1995, 2009) and Jan et al. (2015) was used to quantify sediment P fractions (Table 3). Because fractions are defined by the extracting agent and extraction conditions, they are designated as "operational" phases (Hupfer et al., 2009). All procedures were performed at room temperature.

After homogenization and sieving (1 mm), a known amount of wet sediment (0.1–0.2 g) from each sediment section was placed into a centrifuge tube (15 ml, VWR) and sequentially extracted. For each extraction step, 10 ml of extractant (Table 3) was allowed to react with the sediment for a given time. After extraction, samples were centrifuged (3,000 rpm, 10 min), and RP content in the supernatant was determined with a spectrophotometer (Hach Lange DR2800) at 880 nm using the ammonium molybdate blue method (Murphy & Riley, 1962). In each sample batch three blank samples were run for quality control. A reference sample was also included in every analysis batch to identify possible discrepancies between runs. Compared to the original Psenner and Pucsko (1988) method, we employed a set of minor modifications (all of them described in Text S1). The most important modification was that the BD extraction was divided into two steps: a short 10 min (BD1) extraction followed by a second 1-hr extraction (BD2) (Jan et al., 2015). This modification was made according to Jan et al. (2015), who showed that the longer step could dissolve more crystalline forms of Fe-(hydr)oxides in addition to the redox sensitive and more easily available P (Table 3).

In the field of lake eutrophication management, P from the H_2O-P, Fe_1-P, and labile Org-P-step is commonly referred to as "labile P," which can contribute to internal loading (Huser et al., 2016; Reitzel et al., 2005; Rydin, 2000; Schütz et al., 2017). Thus, these fractions will therefore be referred to as such in this paper. However, processes affecting internal P loading from lake (Orihel et al., 2017) and streambed sediment (Records et al., 2016) are difficult to quantify for different environments as the processes affecting internal loading are dynamic and varying in time and space. Labile Org-P was estimated by subtracting background Org-P concentrations measured deeper in the sediment. These deeper Org-P concentrations were assumed to represent recalcitrant forms of organically bound P (Huser & Pilgrim, 2014; Reitzel et al., 2005; Rydin, 2000). Earlier studies have shown that the sum of P fractions (Table 3) is within $\pm10\%$ of total sediment P (for Swedish lakes, Huser & Rydin, 2005; Psenner & Pucsko, 1988) and is hereafter termed sediment TP.

Water content was determined by freezing the samples ($-20°C$, 12 hr) followed by freeze drying ($-40°C$, 96 hr). Sediment bulk density and percent organic matter were determined after loss on ignition (550°C, 2 hr) (Håkanson & Jansson, 2011). Bulk density was calculated from Equation 1, where W is the percent water content and IG is the percent loss on ignition (building on Håkanson & Jansson, 2011, Pajunen, 2000).

$$\rho = \frac{260}{100 + 1.6 \times \left(W + \left(\left(\frac{100 - W}{100} \right) \times IG \right) \right)} \tag{1}$$

2.4. Data Analysis

All univariate statistical analyses were performed with JMP Pro 13.2.1 (SAS Institute, 2013). Multivariate statistical analyses were performed using CANOCO 5 (ter Braak & Šmilauer, 2012). Summaries of sediment P

contents (g/kg DW) and concentrations (mg/cm^3) as well as fractions were based on the average of triplicate cores collected at each site. Average contents and concentrations in the surficial layer (top 0–4 cm) of the sediment were used for most analyses. Delineation of surficial layer depth was based on Reynolds and Davies (2001), who suggested that the top 4–5 cm of sediment can be regarded as active. However, the active layer of sediment can be greater, but 4 cm was used as the active/surficial layer for all sites to standardize comparisons.

First, it was important to explore if the actual results differed between lakes and streams as well as sites. A mixed model nested ANOVA was used to explore the influence of waterbody type (lake/stream: fixed effect) and site (nested random effect) on variations in TP contents and concentrations in the sediment (g/kg and mg/cm^3). Lakes and streams were treated as fixed effects as they define all relevant waterbody types, while individual lake and stream sites were treated as random effects as each site is a sample from a larger population (Weiss, 2005).

There are a number possible sources of variation among P fractions. Thus, it is important to explore if there are statistical differences in the sediment content of different P fractions between waterbody types, sites, and triplicate samples. Components of variability in sediment P fractions were quantified using a hierarchical partitioning of variation following a protocol described by Šmilauer and Lepš (2014). The total variation in sediment content of different P fractions was partitioned into three categories: (i) between waterbody type (lakes vs. stream), (ii) within waterbody type (i.e. among lakes or streams), and (iii) within triplicate replicate samples. For these analyses, the P fraction data were Hellinger transformed to produce meaningful ordination results derived from proportional data (Kothawala et al., 2015). Variation due to between waterbody differences (i) was quantified using an RDA (redundancy analysis, Legendre & Legendre, 2012) with a dichotomous variable for lakes and streams as the sole predictor. The unassigned variation from this analysis was assumed to be equal to the total remaining variation. The within site variation associated with triplicate samples (iii) was quantified using a partial RDA with replicate as the explanatory variable and site as the block in which replicates were permuted. Within waterbody variation (ii) was identified as the variance remaining after between water body type (i) and within triplicate (iii) variation was accounted for. The RDA analysis was conducted using CANOCO 5.

To estimate the mass of P in the catchment a GIS analysis was performed to compare the amount of P stored in lakes with P stored in streams. To extrapolate the results from the sampling campaign, stream sections were categorized according to Water Information System Sweden. Each stream section was assumed to be the same width and have the same sediment TP concentration (cumulative 0–4 cm, mg/cm^3) as the geographically closest sampling point. Mass of TP was estimated by multiplying the length of each stream section by stream width and TP concentration. The estimated lake sediment area for the sampled lakes in the catchment was delineated using the lake area from the Cadastral map (The National Land Survey, 2020). The lake sediment was assumed to have the same TP concentration (cumulative 0–4 cm, mg/cm^3) over the whole lake area. Lake area and TP concentration were multiplied to estimate the total TP mass in the lake sediment. All stream section and lake TP mass estimates were summarized and compared.

To determine if there were any relationships between the distribution of P fractions at the different sites and the surrounding land use/soil type, a principal component analysis (PCA) was performed using CANOCO 5. A PCA is a way to graphically represent multivariate data for examination of clusters, outliers, and structures (Zuur et al., 2007). The land use and soil type data were extracted from the Cadastral map (The National Land Survey, 2020) and Soil map (Geographical Survey of Sweden, 2020) for the area draining the sampling location (Tables 1 and 2). The spatial data were later used in a PCA together with the relative fractions of P (g/kg DW) from 0–4 cm. A Hellinger transformation suitable for relative abundance data was used (Kothawala et al., 2015).

3. Results

3.1. Phosphorus Content in the Sediment

Generally higher TP contents (g/kg DW) were found in lake as compared to stream sediment, both in the surficial layers (0–4 cm) and deeper in the cores (Figure 2, Table 4). The average surficial TP content for lakes varied between 1.31–2.00 g/kg DW (Table 4, standard deviation reported in Table S3). A few streams showed
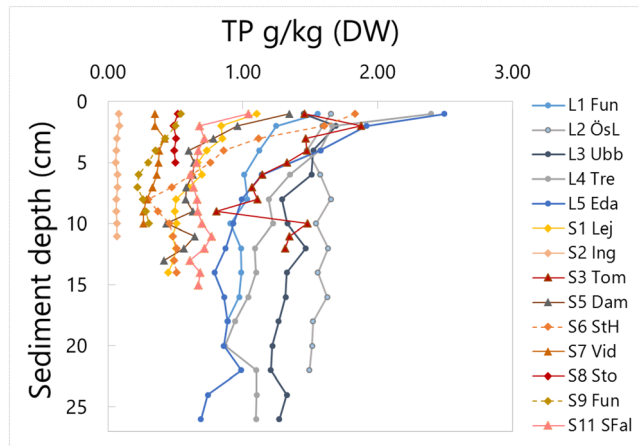
Figure 2. Total phosphorus content (g/kg) by depth in lake (L1–L5) and stream (S1–S11) sediment.

comparable TP contents to lakes (e.g., S3 1.57 g/kg DW, S6 1.35 g/kg DW, Figure 2, Table 4), while some streams had considerably less TP stored in the bed sediment (e.g., S2 0.073 g/kg DW, Figure 2, Table 4). The variation between the different sites was large, especially between the different streams (e.g., 22 times higher TP contents in S3 compared to S2). The difference in relative amounts of variation between sites was confirmed by a nested ANOVA which showed a weak overall effect on TP content (g/kg DW) depending on whether a site was a stream or a lake ($p = 0.0027$) but a strong site effect. The variance was mostly attributed to differences between sites (78%), while the replicate variation was 22% of the total variance (Table S4).

Total P content decreased with sediment depth for all lakes, whereas streambed sediment had more variable depth profiles (Figure 2). Some stream sites showed similar contents in the top and bottom of the core (e.g., S2, difference 0.017 g/kg DW) compared to some of the lakes where the difference between the top sediment and bottom sediment was as high as 1.81 g/kg DW (L5) (Table 4).

Water content, organic matter, and density varied greatly between different streams (in the surficial layer), showing a pronounced difference in type of bed sediment (Table 4). Streams with sediments having a high proportion of clay sized particles (visual assessment, S2, S7, S8, S9, and S11) had a low water content (39–51%), low organic matter content (4–7%), and high density (1.41–1.59 g/cm^3). Sediments from streams located in mostly forested areas (S1, S5, and S6) had high water content (80–91%), high organic matter content (31–61%), and lower density (1.02–1.10 g/cm^3). The lakes had high water content in the surface sediment (87–96%), likewise high organic matter content (22–31%) and densities ranging between 1.02 and 1.07 g/cm^3.

Total P concentration (mg/cm^3) in surficial sediment varied greatly across sites (0.21–1.96 mg/cm^3) (Table 4, Figure S5). Stream sites close to the catchment outlet (S8, S9, and S11) generally had the largest sediment TP concentrations (1.72–1.96 mg/cm^3). Lakes appeared to have relatively homogeneous TP concentrations in surficial sediments (0.22–0.57 mg/cm^3). This was confirmed in a nested ANOVA where there was again a weak but statistically significant overall effect on TP concentration depending on whether a site was a stream or a lake ($p = 0.0002$) but a strong site effect. The variance was mostly attributed to differences between sites (88%), while the replicate variation was 12% of the total variance (Table S4).

Table 4
Observed Data From the Study

Site	Sediment Total P (g/kg DW)	Sediment P fractions (content g/kg DW 0–4 cm)						Labile P content (g/kg)	Sediment total P (mg/cm^3) 0–4 cm	Sediment water content (%)	Sediment organic matter (%)	Sediment density (g/cm^3)	Water RP (µg/L)	Water TP (µg/L)
		H$_2$O-P	Fe$_1$-P	Fe$_2$-P	Al-P	Org-P	Ca-P							
S1 Lej	0.882	0.006	0.084	0.027	0.040	0.524	0.202	0.57	0.298	91	61	1.02	3	21.5
S2 Ing	0.073	0.005	0.008	0.003	0.006	0.010	0.042	0.02	0.210	49	7	1.42	4	37.4
S3 Tom	1.568	0.029	0.220	0.106	0.483	0.395	0.335	0.46	1.415	79	19	1.12	13	43.1
S5 Dam	0.922	0.011	0.070	0.037	0.055	0.376	0.372	0.29	0.721	80	31	1.10	3	15.8
S6 StH	1.352	0.013	0.457	0.077	0.052	0.544	0.209	0.99	0.645	87	39	1.06	5	27.5
S7 Vid	0.375	0.007	0.022	0.011	0.079	0.073	0.182	0.07	1.215	45	7	1.47	5	27.5
S8 Sto	0.498	0.037	0.023	0.017	0.018	0.005	0.399	0.06	1.719	43	5	1.51	6	28.9
S9 Fun	0.454	0.037	0.079	0.017	0.013	0.025	0.283	0.14	1.744	39	4	1.59	6	46.7
S11 Sfal	0.775	0.027	0.129	0.040	0.084	0.083	0.412	0.22	1.955	51	5	1.41	6	47.6
L1 Fun	1.309	0.023	0.398	0.091	0.127	0.359	0.312	0.56	0.538	87	23	1.07	3	42.5
L2 ÖsL	1.574	0.017	0.455	0.097	0.331	0.467	0.207	0.60	0.572	88	22	1.06	5	43.6
L3 Ubb	1.555	0.013	0.421	0.053	0.074	0.812	0.184	0.75	0.263	94	31	1.02	1	19.9
L4 Tre	1.876	0.010	0.244	0.041	0.041	1.407	0.133	1.33	0.221	96	31	1.02	1	48.4
L5 Eda	1.995	0.015	0.473	0.056	0.135	1.075	0.242	1.30	0.239	96	27	1.02	3	35.2

Note. Measured contents (g/kg DW) of TP in the sediment samples, measured fraction of RP (g/kg DW), calculated content of labile P (g/kg), TP concentration (mg/cm^3), water content (%), organic matter (%), and surface water RP (µg/L) and TP (µg/L) for each site (S indicates stream and L lake).
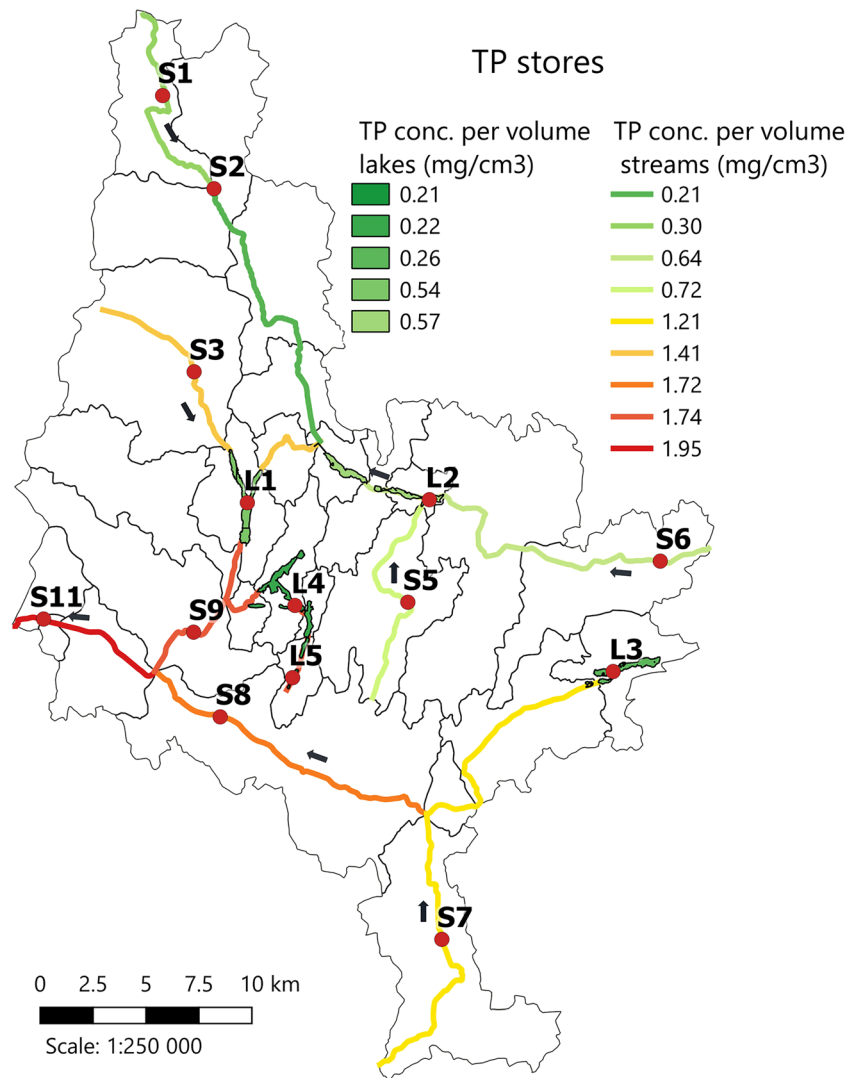
**Figure 3.** Estimated TP storage in lakes/streams in the catchment.

When calculating the TP concentration (mg/cm³), dry matter content and density of the sediment were used. A linear regression between percentage of clay soils in the drainage area of the sampling site and sediment TP concentrations (mg/cm³) resulted in a good positive correlation ($r^2 = 0.70$, $p = 0.0002$, $n = 14$). This may be attributed to the higher proportion of clay sized particles, which allow lower water content and therefore enhancing TP concentration. TP concentrations increase by depth for both lakes and streams (Figure S6), which is unsurprising given that deeper sediments are often more consolidated and have a lower water content (Håkanson & Jansson, 2011).

The surficial sediment in all lakes was estimated to contain 6.0 tonnes of P and all streams 2.1 tonnes (estimated according to Figure 3). This indicates that lake sediments, with a larger overall area, contain about 2.8 times more P than the streambed sediments, despite the generally higher concentrations in streambed sediment (Figure 3).

3.2. Phosphorus Fractions in the Sediment

The absolute and relative amounts of the different sediment P fractions varied substantially between sites (Table 4, Figure 4). Lakes generally had a larger fraction of Fe-P, (15–37%), while one of the forested headwaters streams (S6) had comparable levels (39%). An agricultural headwater stream (S3), which generally had high TP contents in the streambed sediment (Table 4), had a large proportion of Al-P (31%). Some
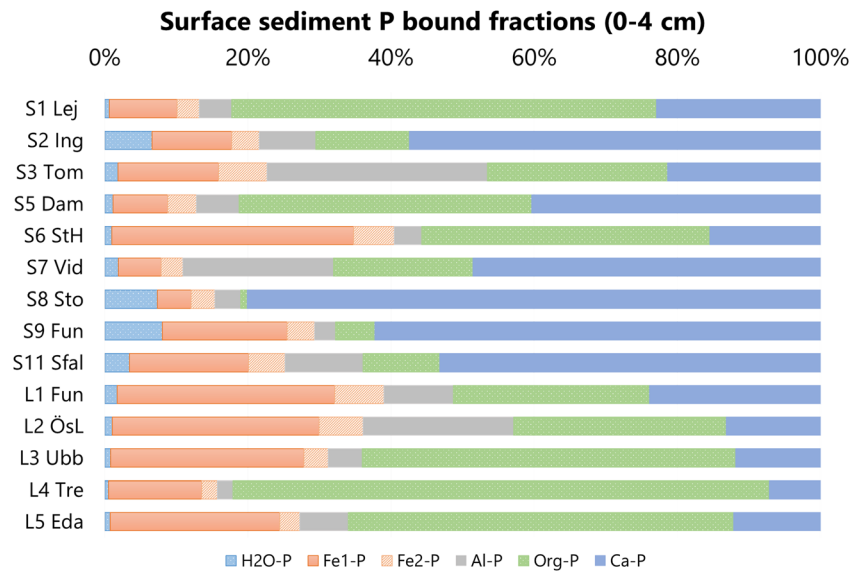
Figure 4. The relative amounts of P stored in each fraction in the surficial sediment (0–4 cm) of the stream (S1–S11) and lake (L1–L5) sites.

streams located in the agricultural parts of the catchment had comparably higher proportions of loosely bound P (H_2O-P) and Ca-P, for example, S2 (7% and 57%), S8 (7% and 80%), and S9 (8% and 62%). One forested headwater stream (S1) and one lake (L4) had a very high proportion of Org-P (S1: 59% and L4: 75%).

In some of the streams (e.g., S1, S6, and S11) H_2O-P and Ca-P fractions increased by depth in the sediment (Table S5). In addition, three other streams (S2, S5, and S9) and all lakes had an increase in only Ca-P by depth. Generally Org-P decreased by depth for all sites, likely due to degradation over time. In the lakes the Al-P increased by depth and in all the streams the Al-P bound fraction decreased by depth for all sites.

Labile P fractions (H_2O-P, Fe_1-P, and labile Org-P) varied between the sites (Figure 5). Lake sediments generally had higher labile P contents than stream sediments (lakes: 0.56–1.33, streams: 0.02–0.99 g/kg DW) (Figure 5). However, three streams (S1, S3, and S6) had comparable labile P amounts to lakes.

The RDA showed that approximately 10% of the variation in surficial layer P fraction composition was associated with replicates collected at the same site and 23% of the variation could be attributed to differences between lakes and streams (Table S6). The remaining 68% of the variation was due to differences in sediment P fraction composition between sites.
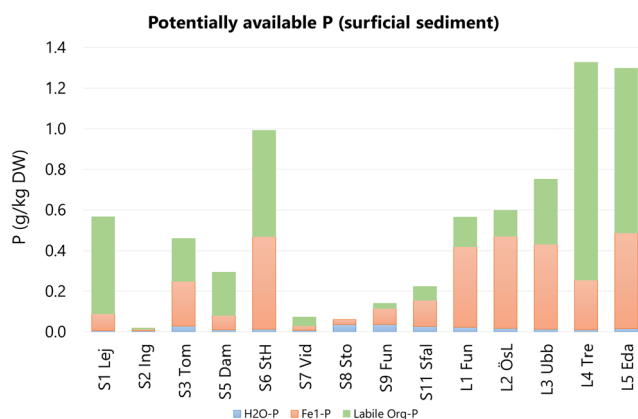

Figure 5. Potentially available P content (H_2O-P, Fe_1-P, and labile org-P) in the surficial sediment (0–4 cm) of stream (S1–S11) and lake (L1–L5) sites.

The results from the snapshot water chemistry campaign show no strong correlation between labile P (H_2O-P, Fe_1-P, and labile Org-P) in the surficial sediment and measured RP or TP in the overlaying water. Also no other individual fraction was significantly correlated to RP or TP. On the other hand, surficial sediment TP concentration (mg/cm^3) was weakly correlated to RP concentration ($r^2 = 0.43$, $p = 0.01$, $n = 14$). Hence, during the conditions sampled, water chemistry could not be attributed to the P fractions in the sediment, but possibly there could be a connection to TP concentration.

3.2.1. Effect of Land Use and Soil Type

A PCA (Figure 6) showed relationships between sediment content of different P fractions, land uses, and soil types. Sampling sites with forest, rock, and water land cover in the drainage area (S1, S5, S6, L3, L4, and L5), as well as lakes in general had surficial sediment containing high proportions of Org-P and Fe_1-P. In areas of the catchment with clay soils, agriculture, and open land the dominant fractions were H_2O-P and Ca-P
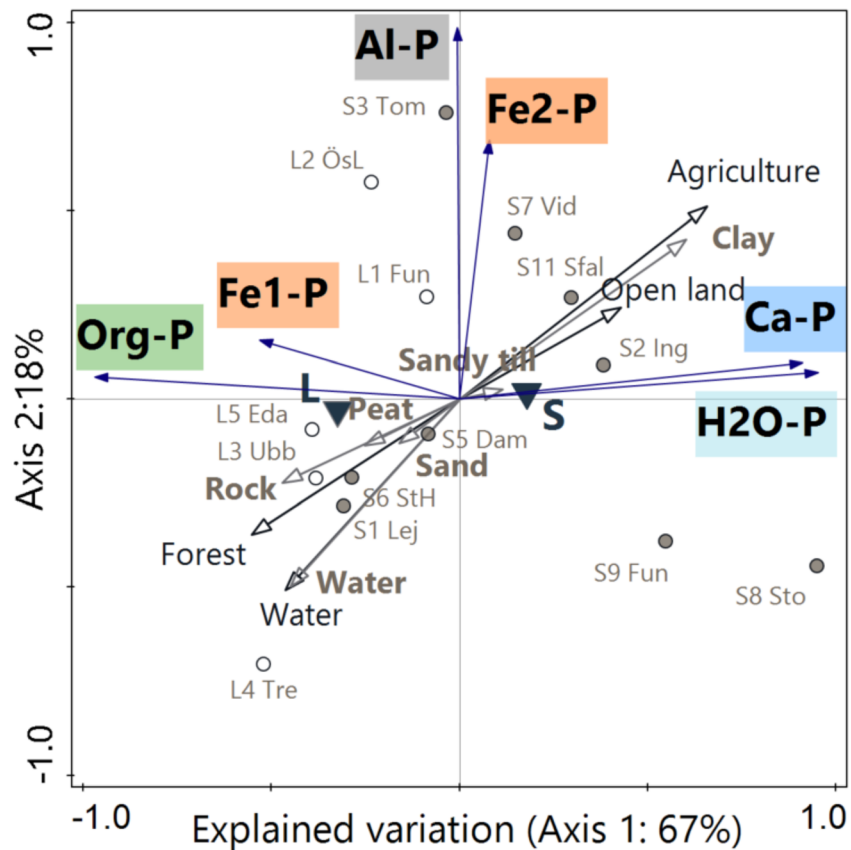
Figure 6. Biplot from a principal component analysis on Hellinger-transformed P fractions in surficial sediments of lakes and streams, where the distributions of P fractions in surficial sediment (g/kg 0–4 cm), soil type, land use, and lake/streams (L/S) were included.

(Figure 6). Streams in general correlated with these P fractions as well. The fractions Al-P and Fe$_2$-P correlated with a few sampling sites, both streams and lakes. The PCA explained 85% of the total variation in surficial layer sediment P fractions on the first two axes.

4. Discussion

4.1. Phosphorus Storage in the Catchment

The observed TP content (g/kg DW) in this study (0.073–2.00) was within previously reported ranges for both lake sediment (0.66–8.5 g/kg DW; Boström et al., 1988; Rydin, 2000; Søndergaard et al., 1996; Zhang et al., 2013) and streambed sediment (0.155–2.00 g/kg DW, Audette et al., 2018; Dorioz et al., 1998; Noll et al., 2009; Palmer-Felgate et al., 2009; SanClements et al., 2009). Most of the variation in TP content was between sites, with no clear separation between streams and lakes. Stream sites were more variable than lake sites (Figure 2, Table 4), with some streams having comparable TP contents to lakes. For sediment TP concentration, seven out of nine stream sites had higher TP concentrations than the lake with highest concentration (i.e., >0.57 mg/cm^3). Especially the sites close to the outlet of the catchment had large stores of P. Owens and Walling (2002) reported that TP streambed sediment content increased downstream in two of the three catchments they studied. Némery and Garnier (2007) observed that the particulate P content of suspended sediment was 4 times higher at the catchment outlet than in the headwaters. The explanation for this would be that the outlet of the catchment is receiving an integrated signal from both point sources and diffuse agricultural sources from a larger contributing area. Despite the generally higher TP concentrations in streams compared to lakes, lakes were estimated to contain about 3 times as much TP in surficial sediments due to their larger area (Figure 3).

Under certain conditions (e.g., low oxygen conditions combined with Fe bound P, substantially higher P concentration in bed sediment pore water than in the water column, decomposition of organically bound P), lake sediment can release P to the water column that is later transported through the catchment. For release to occur, a potentially mobile P store in the sediment is needed. If there are similar TP contents (g/kg) and concentrations (mg/cm^3) (and in some cases P fractions) in streambed sediment, it would be highly unlikely that these stores did not contribute to the transported P in the catchment, as has been shown in earlier studies (e.g., Hongthanat et al., 2016; Jarvie et al., 2005; Svendsen et al., 1995; Taube et al., 2019; Weigelhofer et al., 2018). The results confirm the likely significance of streams as important stores of legacy P and potential P sources, as well as lakes which corroborates our first hypothesis. Moreover, it highlights the importance of considering P stored in streams when conducting drainage management activities or catchment scale water quality modeling for decision support.

4.2. Stream Order and Land Use Effects on Sediment Phosphorus

4.2.1. Forested Headwater Streams and Lakes

Phosphorus fractions varied between lakes and streams but also with catchment location and land use, which strengthens our second hypothesis. Headwater streams (with mostly forest and on sandy till soils) had similar fractional P composition as the lakes (Figure 4), where the dominating fractions were generally Org-P and Fe_1-P. In lakes, both allochthonous and autochthonous organic material are accumulated over time in the bed sediment; hence Org-P was also the predominating fraction (L3–L5) (Figure 4). The forested headwater stream sites (S1, S5, and S6) had a layer corresponding to about 8 cm of brown sediment with lower density compared to the deeper layers (Table S2). This accumulation of "fresh sediment" could be explained by lower discharge in the first-order streams and a steady supply of organic material consisting of degrading debris and litter from the boreal landscape. Small headwater streams have a greater potential for physical, chemical, and biological P exchange because they have lower water volume to bed sediment area ratios than higher-order streams and rivers (Withers & Jarvie, 2008). The fact that these streams generally had high TP contents (g/kg DW, Table 4), as well as a larger proportion of labile P (Figure 5) compared to the other streams makes them relevant as a potential source of P. This supports the argument made in Bol et al. (2018), where headwater catchments are suggested as especially interesting regarding P processes and pathways for future research.

4.2.2. Agricultural Streams

For streams located in agricultural areas (S2, S7, S8, S9, and S11) Ca-P dominated (Figure 4), which we assume is closely linked to the proportion of clay particles in the sample. These streams had considerably less organic matter (4–7%), lower water content (39–51%), and higher density (0.41–1.59 g/cm^3) compared to lake sediments (Table 4). Here the fresh sediment was estimated to extend to a depth of 1–4 cm with more stones and less visible organic matter (Table S2). Stream order and the consequent increase in discharge (S8, S9, and S11) probably mobilize organic matter and lower density particles. What remains is an aggregated clay layer with comparably high density and a greater TP concentration than the low density sediment. In the PCA agriculture and clay soils correlated with the distribution of P fractions in most of the streams (S2, S7, S8, S9, and S11) (Figure 6). That is likely due to the catchment land cover organization where agriculture is located on clay soils while the forest generally is located on sandy till soils (Figure 1, Figure S1). Consequently these streams (S8, S9, and S11) have limited organic matter input from the environment, which is reflected in the streambed sediment.

Anthropogenic activities that influence input of P, suspended sediment and organic matter, water residence time, and the riparian zone also affect P cycling (Gramlich et al., 2018; Palmer-Felgate et al., 2009). For example, the higher TP concentrations in the streambed sediment in the agricultural areas might depend on input of fertilizer from the surrounding fields. Generally, agriculture reduces the organic carbon in streams (Stutter et al., 2018), which could be connected to the lower Org-P observed relative to forest sites. Artificial drainage affects hydrological pathways which may enhance the total water flow and flow velocity (Gramlich et al., 2018) which could be connected to the limited accumulation of "fresh sediment" at the time of sampling. The results suggest that the P stores in the larger agricultural streams are more transitory, which would suggest a need to further investigate suspended sediment as a vector for P transport in these streams.

The high TP concentrations found in the sediment of the headwater agricultural stream (S3), corroborated by the high TP concentration in the water sample from the snapshot survey, could likely be explained by

anthropogenic influence. The stream has been classified as having moderate ecological status and bad chemical status, significantly affected by a landfill, agriculture, and discharge from a private sewage treatment system (Water Information System Sweden, 2019).

4.3. Geological Effect on Sediment Phosphorus

Catchment geology may influence sediment P storage capacity, for example, concentrations of Fe and Ca in bed sediment have shown a positive correlation with TP concentrations (House & Denison, 2002; SanClements et al., 2009; van der Perk et al., 2006). The glacial clay in the catchment likely contains between 10–30% $CaCO_3$. This can explain the high levels of H_2O-P and Ca-P in the samples (up to 80% of the total concentration in S8). Since the Ca-P increases by depth for all sites it is likely that this fraction is connected to the parent material in the area and may be the more stable fraction over time (Noll et al., 2009).

4.4. Availability of Phosphorus: Influence on Internal Loading

Sediment P release depends on potential mobility in combination with prevailing conditions in the surrounding environment. Different P fractions are considered to have different potential to contribute to internal loading. Two forested headwater streams (S1 and S6) and an agricultural headwater stream (S3) show similar levels of labile P to the studied lakes (Figure 5). The streams S3 and S6 also have comparable TP contents (g/kg DW, Figure 2, Table 4) as lakes. Hence, not only the TP contents of lake and stream sediments are in some cases comparable but also the labile fraction. There is however a need to question whether the same fractions ought to be considered as labile for lakes and streams. Lake and stream internal loading processes have many similarities but also some differences.

In streams, discharge variation causing turbulence/hyporheic exchange is of high importance for the internal loading, mobilizing particulate P, activating sorption/desorption mechanisms and releasing dissolved P from pore waters (Withers & Jarvie, 2008). The loosely bound fraction (H_2O-P) as well as P fractions that are susceptible to sorption/desorption ought to be affected (Fe-P, Al-P, and Ca-P in high pH systems) could all be affected by turbulence. However, less is known about the dynamics of Al bound P (Rydin, 2000). When the turbulence decreases, new sorption equilibria need to be established influencing chemical diffusion in the sediment. Wind mixing and resuspension have been reported as being two of the most important factors for internal loading in shallow lakes (Fan et al., 2001; Koski-Vähälä & Hartikainen, 2001; Jones & Welch, 1990). While turbulence in lakes might occur occasionally (e.g., induced by increased wind), the stream bed sediment is expected to be exposed to changes in discharge continuously.

Processes that affect sediment oxygen levels in lakes are of high importance for P dynamics, due to the redox mediated release of P. The redox sensitive P fraction (Fe_1-P and Fe_2-P) is considered to be potentially available because P release can occur under anoxic conditions that can develop during lake stratification (e.g., Søndergaard et al., 2003) and during less turbulent conditions or night time when only respiration occurs in shallow water bodies and streams (Bamforth, 1962; Whitney, 1942). If the whole water column is oxic, which means that the top part of the sediment is oxic as well, this microlayer can inhibit sediment release and might retain P (Penn et al., 2000). When conditions become anoxic, P can be released into the water column again. The hyporheic zone in the streambed sediment is generally oxic; hence the Fe-P fraction would not be considered as prone to be released as in lakes. However, streambed sediment can undergo anoxic periods during intrusion of reduced groundwater (Krause et al., 2011; Soulsby et al., 2009) which would affect the Fe-P fraction.

Low order streams are more responsive than higher-order streams and lakes to temperature increase that reflect biological processes (e.g., mineralization) (Withers & Jarvie, 2008). An increased organic matter loading (and subsequent higher mineralization rates) can lower sediment oxygen concentrations, which in turn affect redox processes (Søndergaard et al., 2003; Withers & Jarvie, 2008). Biological processes could also contribute to degradation of labile organic P, which would make this fraction important in streams. Labile organic P may be potentially available due to natural degradation of more labile forms of organic matter and subsequent release of P in lakes (Ahlgren et al., 2011; Rydin, 2000; Søndergaard, 1989).

The conclusions to be drawn from the data set available regarding internal loading of P in different types of water bodies are highly limited. Depending on the relative importance of drivers in the stream and lake environments different fractions might be of interest from an internal loading perspective. The availability of the different fractions is highly dependent on environmental conditions which are variable in time and

space. For future studies it would be valuable to explore the bioavailability of P in streams further and look into what fractions are potentially available over the year. It would also be relevant to couple the P cycle to other elements present, for example, sulfate (Roden & Edmonds, 1997), as well as to determine the lability of the organic matter.

Additional sampling over the year would likely provide further insights into sediment P dynamics. Ballantine et al. (2009) showed that P concentrations and fractions in the streambed sediment varied over the year with a maximum concentration in the late summer/early autumn. The variation depends on plant uptake, which affects the inorganic and organic sediment P stores. In lakes, changes in redox status, sediment mobile P content (Pilgrim et al., 2007), and P concentration gradients also vary seasonally and affect sediment P release (Penn et al., 2000).

4.5. Implications for Policy

Our results suggest that 2.1 tonnes of P are stored in stream sediments in the Sävjaån catchment (Figure 3). A proportion of this P store is readily available, and another potentially available during certain conditions. Legacy P is an important component of the P transferred from fields to receiving waters and contributes to slowing the recovery of eutrophic aquatic systems (Søndergaard et al., 2003). Of the streams studied here, 63% are covered by drainage permits that give the land owner the right and duty to maintain the depth and position of water over time (County Administrative Board of Uppsala, 2020). Dredging has been shown to remove freshly deposited sediment (including organic matter) as well as biotic communities (aquatic plants, algal mats, and algae) leading to a decreasing capability to retain P in the ditch (Smith & Pappas, 2007). Consequently, these streams periodically turn into sources of P increasing the speed of P transport to downstream waterbodies and the sea (Smith et al., 2006). Maintenance actions for compliance with drainage permits, for example, dredging, harvesting vegetation, and deepening stream channels accelerate and potentially increase the transport during a short time (Smith et al., 2006; Smith & Pappas, 2007). Well maintained ditches/streams are a widely used tool to ensure efficient removal of water from agricultural fields, but dredging and other maintenance should be done during times of the year when nutrient loads are expected to be low and does not coincide with fertilizer application (Smith & Pappas, 2007). In Sweden the permit owner does not have to notify the authorities before dredging streams/ditches, except for when the works can be detrimental to fishing (Government Offices of Sweden, 2020). Accordingly timing and environmental consideration generally cannot be suggested or requested by authorities. It is not yet known how much P is actually mobilized because of these actions; hence further studies are needed to evaluate the effect of ditch maintenance on P transport and sediment P release. With the results from this study, we show that maintenance of many of the studied streams should be done with caution, especially during ecologically sensitive times of the year to avoid mobilization of bioavailable P.

5. Conclusions

Streambed sediment can store considerable amounts of P, in some cases comparable to P stored in lake sediment. Land use and soil type have significant effects on the different P fractions in the sediment. Sediment in lakes and forested headwater streams had large proportions of Org-P, indicating that the input of organic matter may be important for P storage. Ca-P was the predominating fraction in sediments in the larger streams situated close to the outlet of the catchment in agricultural areas with clay soils and higher discharge. Larger streams in agricultural areas had high concentrations of sediment TP in the surficial layer (mg/cm^3), but the total storage of P in lake sediment was 2.8 times higher than in streambed sediment due to a larger lake area in the catchment. However, the stream environment with continuous turbulence creates a prevailing risk of internal loading—if the bed sediment P contents are high. We suggest that it is important to consider streambed sediment P stores when modeling P transport in catchments and to evaluate the potential for mobilizing bioavailable P during maintenance of streams/ditches. This is moreover relevant as water from Sävjaån and most other Swedish agricultural catchments eventually reaches the Baltic Sea, where eutrophication is a serious problem.

Conflict of Interest

No conflicts of interest are relevant for this study.

Data Availability Statement

Data sets for this research are available at Figshare (https://doi.org/10.6084/m9.figshare.12052890, https://doi.org/10.6084/m9.figshare.12833639, and https://doi.org/10.6084/m9.figshare.12833630), as well as described in this paper and supporting information.

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