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Variability in fluvial suspended and streambed sediment phosphorus fractions among small agricultural streams

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Abstract

Agriculture is a major source of sediment and particulate phosphorus (P) inputs to freshwaters. Distinguishing between P fractions in sediment can aid in understanding its eutrophication risk. Although streams and rivers are important parts of the P cycle in agricultural catchments, streambed sediment and especially fluvial suspended sediment (FSS) and its P fractions are less studied. To address this knowledge gap, seasonal variations in FSS P fractions and their relation to water quality and streambed sediment were examined in three Swedish agricultural headwater catchments over 2 yr. Sequential fractionation was used to characterize P fractions in both streambed sediment and FSS. All catchments had similar annual P losses (0.4–0.8 kg ha^{-1}), suspended solids (124–183 mg L^{-1}), and FSS total P concentrations (1.15–1.19 mg g^{-1}). However, distribution of P fractions and the dominant P fractions in FSS differed among catchments ($p < .05$), which was most likely dependent on differences in catchment geology, clay content, external P sources, and flow conditions. The most prominent seasonal pattern in all catchments was found for iron-bound P, with high concentrations during low summer flows and low concentrations during winter high flows. Streambed sediment P fractions were in the same concentration ranges as in FSS, and the distribution of the fractions differed between catchments. This study highlights the need to quantify P fractions, not just total P in FSS, to obtain a more complete understanding of the eutrophication risk posed by agricultural sediment losses.

1 | INTRODUCTION

Phosphorus (P) is a primary limiting nutrient in freshwater ecosystems (Correll, 1998; Schindler, 1974). Agricultural activities generate spatially variable and episodic P inputs to surface waters (Sharpley et al., 2009). Such nonstationary dynamics can lead to challenges for understanding the primary processes governing P mobility in different catchments as well as difficulties in predicting and implementing appropriate mitigation policies and countermeasures. These issues are common in agricultural headwater catchments, which can have a considerable influence on downstream rivers and lakes (Bol et al., 2018).

Abbreviations: Al-P, aluminum-bound phosphorus; Ca-P, calcium-bound phosphorus; Fe-P, iron-bound phosphorus; FFS, fluvial suspended sediment; H_2O-P, loosely bound phosphorus; OM, organic matter; org-P, organically bound phosphorus; PP, particulate phosphorus; RP, reactive phosphorus; SS, suspended solids; TP, total phosphorus.

Historical land management practices have caused accumulation of legacy P in soils and freshwater sediments (Kleinman et al., 2011; Sharpley et al., 2013) where biogeochemical processes can transform recalcitrant P fractions to more bioavailable forms (Lannergård et al., 2020). Although streambed sediments are important sources and sinks for P cycling (Ezzati et al., 2020; Jarvie et al., 2005; Owens & Walling, 2002; Weigelhofer et al., 2018), the role of fluvial suspended sediment (FSS) in P cycling is rarely investigated despite its potential significance. Different P fractions (e.g., phosphate-P, organic P, and metal-bound P) differ in mobility and bioavailability, which governs impacts on water quality and aquatic ecosystem functioning (Kaiserli et al., 2002; Ye et al., 2017). New insights into the relative and absolute amounts of different P fractions in both streambed and FSS will support more comprehensive assessments of potential effects of agriculture on aquatic ecosystems and generate more focused and effective catchment-specific eutrophication mitigation strategies.

Several chemical sequential extraction schemes exist to quantify different P fractions in sediment (Goedkoop & Pettersson, 2000; Hupfer et al., 2009; Psenner, 1988; Ruttenberg et al., 2009; Williams et al., 1976). Most schemes define operational P fractions, including loosely sorbed P, metal-associated P (e.g., iron [Fe] and aluminum [Al]), and calcium (Ca)-bound P (Condron & Newman, 2011). Numerous studies exist on P fractions in lake sediment (Agstam-Norlin et al., 2020; Kaiserli et al., 2002; O'Connell et al., 2020; Pettersson, 1998), and there are a few studies of P fractions in streambed sediment (Audette et al., 2018; Lannergård et al., 2020; SanClements et al., 2009). However, studies of FSS are rare (Ballantine et al., 2008; Nguyen & Sukias, 2002), and knowledge about seasonal variations is needed.

The study aims were to quantify patterns in the distribution of P fractions in FSS and streambed sediment in three headwater agricultural catchments and investigate potential controlling factors (Aim 1), to investigate seasonal changes in distribution of FSS P fractions (Aim 2), and to explore relationships between water column phosphate P and the labile FSS P fraction as well as between water column and FSS total P (TP) (Aim 3). To address the study aims, FSS and streambed sediment from three well-monitored agricultural headwater catchments in southern Sweden were sampled over 2 yr and analyzed for different P fractions.

2 | MATERIALS AND METHODS

2.1 | Catchment descriptions

Three small southern Swedish agricultural catchments (C6, U8, and E23) in the Swedish Environmental Monitoring program (Kyllmar et al., 2014) with a history of high P load and suspended solids (SS) transport were selected (Figure 1; Tables 1 and 2) (Sandström et al., 2020). All three catchments are dominated by agricultural land (>50%) and clay-rich soils. The smallest catchment, U8, has the heaviest soil texture (Table 1). Streams in U8 and E23 are small and can experience extremely low flows, especially in summer. The most southern catchment, E23, has a markedly higher stocking density and higher rates of manure applications (Table 1), whereas C6 has the lowest rate (Linefur et al., 2019). The U8 outlet is surrounded by agricultural fields, and the stream channel can become overgrown with vegetation during summer. The C6 stream outlet is also surrounded by agricultural fields, and alongside the outlet there is a wetland-like terrace that is inundated during high flows. Streambed sediments at the outlet of both U8 and C6 consist mainly of clay, overlain by organic material (2–5 cm). In E23, the catchment outlet runs through a forested area, with stones and twigs in the stream. Streambed sediment in E23 is harder than in U8 and C6 and contains substantially more gravel and sand.

Core Ideas

- Similar total phosphorus losses were seen in three agricultural headwater streams.
- P fractions in suspended and bed sediment varied significantly among catchments.
- Large seasonal variations in suspended sediment P fractions were seen.
- Dominant P fractions are dependent on clay content, geology, and flow.
- Suspended and bed sediment P fractions should be included in routine monitoring.

2.2 | Fluvial suspended sediment and streambed sediment: Sampling strategy and sample preparation

Two Phillips samplers (Supplemental Figure S1) (Phillips et al., 2000) were placed at the outlet of each catchment to collect FSS. The samplers were positioned mid-stream at approximately mean depth above the streambed, with the inlet facing upstream (Phillips et al., 2000). During the first year, samplers were emptied approximately every fourth week into 10-L buckets (to collect FSS for further analyses) and then put back into the stream (Figure 1). Sampling started in C6 and U8 during autumn 2017, and E23 was added in autumn 2018. During 2018/2019, samplers were emptied every 8–10 wk to ensure enough FSS was collected for later analyses. From October 2017 to May 2018, the C6 stream was flooded, and samplers were not retrievable during this period; hence,

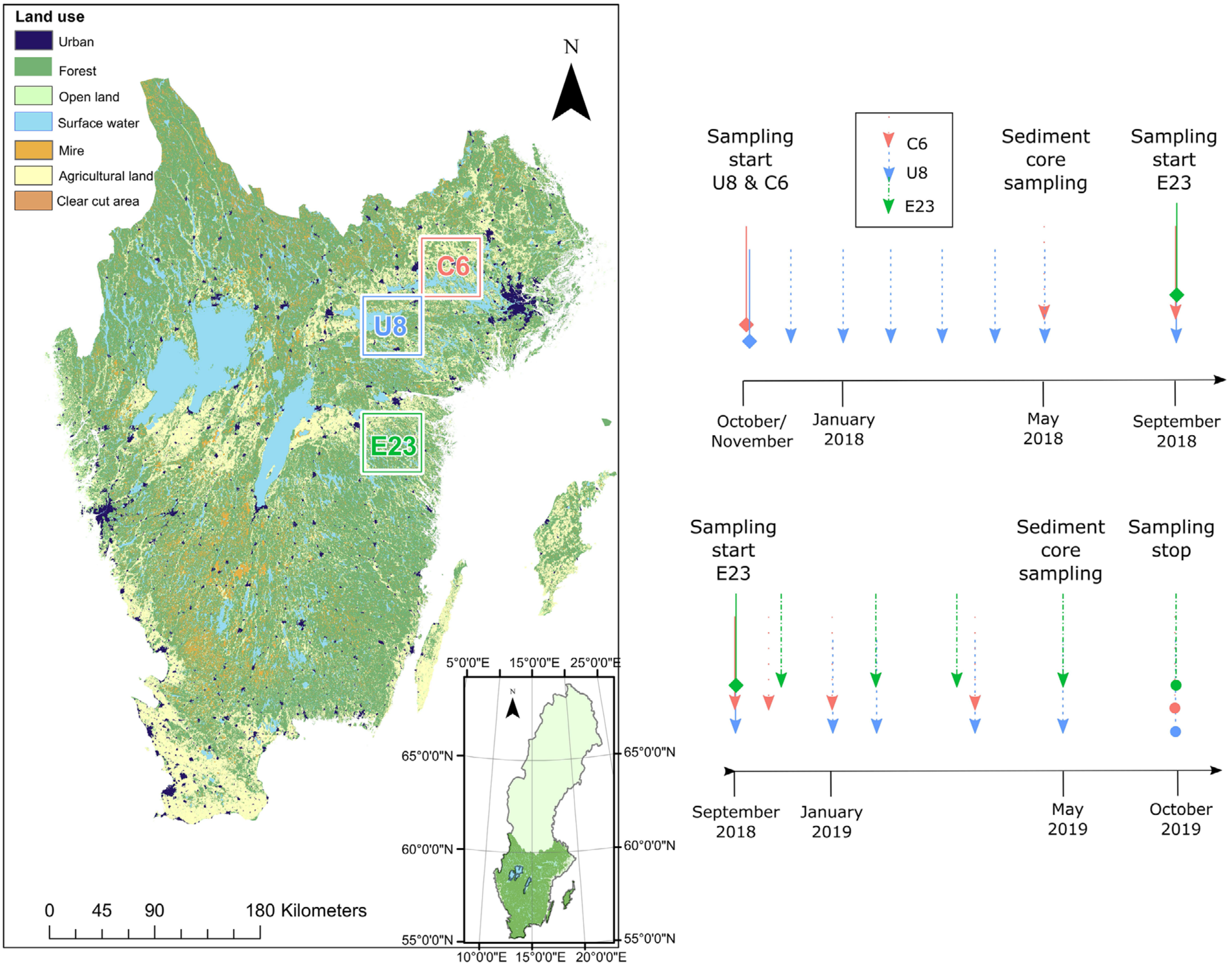

FIGURE 1 (Left) Locations of the three study catchments in southern Sweden including land use as the background map. Exact locations cannot be provided due to agreement with the farmers. The inset map displays Sweden with marked latitude and longitude lines. (Right) Illustration of sampling scheme of fluvial sediment and streambed sediment. The different colors represent the different catchments. Rhombi indicate sampling start for each individual catchment, filled circles represent the time at which sampling stopped, and colored arrows indicate sampling dates

one sample represents this entire 6-mo period. From April to May 2019 there was not enough material in the C6 samplers for analysis. All samples were brought back to the laboratory and wet sieved through a 0.063-mm nylon mesh to collect the smallest fractions and remove plant material. Samples were stored in 2-L buckets at 4 ˚C for at least 24 h to allow particles to settle. After sedimentation, as much supernatant as possible was removed through decanting and pipetting. The remaining material was transferred to a 50-cl centrifuge tube and centrifuged for 5 min at 3,500 rpm, and the supernatant was removed by decanting. Samples were stored in airtight boxes at 4 ˚C until analysis.

Streambed sediment cores were taken in each catchment at a mid-channel location as close as possible upstream from the Phillips samplers to get a representative sample from the stream rather than the streambanks. Each core was collected using a Willner gravity corer on a rod or by simply pushing the tube in manually, with two plugs creating a vacuum. In U8 and C6, cores were collected in May 2018. The E23 core was collected in May 2019. Cores were collected after the spring flood and before too much vegetation had grown to avoid plant disturbance. Due to the coarse and hard E23 streambed, this core was markedly shorter (4 cm) than cores from the other catchments (8–9 cm). All cores were sliced into 1 cm (where possible) slices in the field. Slices were stored in airtight cans at 4 ˚C until analysis (slices were held for a maximum of 6 wk).

2.3 | Water quality sampling and analysis

In each catchment, daily discharge was measured at the stream outlet by continuous water level recording at well-defined sections (i.e., a v-notch or broad-crested weir) (Kyllmar et al., 2014). Automatic flow-proportional and grab water samples were collected at the catchments' outlets within the frames of the Swedish Environmental Monitoring Program (Kyllmar et al., 2014). All samples were analyzed following Swedish Standard Methods (Kyllmar et al., 2014). Parameters reported here include total P (TP), phosphate-P (reactive P [RP], following Haygarth & Sharpley [2000]), and SS. Total P was analyzed on unfiltered water samples after digestion, whereas RP was analyzed after filtration at 0.2 µm, both using the molybdate blue method (Murphy & Riley, 1962). Particulate P (PP) was estimated as the difference between TP and RP. Estimates of flow-weighted concentrations and loads were based on Linefur et al. (2019) (see Methods in Supplemental Material).

2.4 | Sequential P fractionation

To analyze samples (suspended and streambed sediment) for P concentration and different P fractions, a sequential P fractionation method based on Psenner and Pucsko (1988), Psenner et al. (1984), and Hupfer et al. (1995), Hupfer, Zak et al. (2009) was used. The P fractions include loosely sorbed P, redox-sensitive P (iron/manganese [Fe/Mn] bound P), nonreducible and Al-hydroxide–bound P, organically bound P (org-P), Ca bound P (Ca-P), and refractory P. The main difference between the method used here and the aforementioned studies is omission of the refractory P step. All sediment samples were analyzed fresh and in triplicate. All fractions are operationally defined and will be referred to by their operational name as follows in parenthesis: extraction with Milli-Q water (H_2O-P), buffered dithionate solution (Fe-P), and NaOH (Al-P), organically bound P (org-P, defined as the difference between Al-P and extraction with NaOH following digestion), and extraction with HCl (Ca-P). All samples were analyzed at the end for RP (unfiltered) following Murphy and Riley (1962). All fractionation results are presented as mg g^{-1} in the context of FSS and streambed sediment comparisons or as mg L^{-1} for comparison to water column concentrations. The sum of all P fractions is hereafter termed "TP" because it has been found to be within ±10% of total sediment P (Huser & Rydin, 2005; Psenner, 1988). The first two steps in the sequential P fractionation method (H_2O-P and Fe-P) are commonly referred to as labile P (Huser et al., 2016; Reitzel et al., 2005, 2006) and were used for comparisons with RP in water columns.

All sediment samples were analyzed for dry matter content by freezing for at least 12 h at −20 ˚C and then freeze drying for at least 4 d. Loss on ignition to determine organic matter (OM) content of the sample was performed by igniting a freeze-dried sample for 2 h at 550 ˚C (Håkanson & Jansson, 1983).

TABLE 1 Catchment characteristics

Catchment ID	Sampling period	Area	Arable land	Stocking density[a]	Scattered households	Applied manure P (mean 2007–2010)	Three dominant soil texture classes[b]	Dominating bedrock (>50%) (main mineralogy)[c]	Precipitation[d]
		km^2	%	AU ha^{-1}	persons km^{-2}	kg ha^{-1}			mm
C6	Oct. 2017–Oct. 2019	33.1	59	<0.1	10	0.35	silty clay (77%), clay loam (14%), clay (4%)	quartz arenite (feldspar-mica-granite)	521
E23	Sept. 2018–Oct. 2019	7.4	54	0.6	7	17	clay loam (41%), silty clay (31 %), clay (19%)	granite (feldspar-mica-granite)	591
U8	Oct. 2017–Oct. 2019	5.7	56	<0.1	11	8	silty clay (85%), clay (14%), clay loam (1%)	granodiorite-granite (feldspar-mica-granite)	539

[a] Stocking density for all catchments, amount of P applied to E23, U8 (Kyllmar et al., 2014), and C6 (Linefur et al., 2017); 1 AU refers to one cow, three pigs, 10 sheep, or 100 hens.

[b] USDA data from Söderström and Piikki (2016).

[c] Bedrock information is from Geological Survey of Sweden (SGU, 2016).

[d] Precipitation and temperature are normal values (i.e., a long-term mean value, from SMHI 1961–1990; Alexandersson et al., 2001).

TABLE 2 Catchment data

Catchment ID	Temp.	pH (H_2O)[a]	TP conc.[b]	RP conc.[b]	PP conc.[b]	SS conc.[b]	TP load[c]	Discharge[d]
	˚C		mg L^{-1}				kg ha^{-1}	mm
C6	5.5	7.5	0.21	0.05	0.15	144	0.4	226
E23	6.3	7.6	0.28	0.12	0.14	124	0.4	176
U8	5.9	7.3	0.26	0.06	0.19	183	0.8	230

[a] The pH value represents a mean value for the study period.

[b] Long-term mean concentrations of total P (TP), phosphate P (RP), particulate P (PP), and suspended solids (SS) (U8 and E23: 2007–2017; C6: 2004–2017) (Sandström et al., 2020).

[c] Long-term average load of TP (U8 and E23: 1995/1996–2014/2015; C6: 2005/2006–2014/2015) (Linefur et al., 2017).

[d] Long-term (1995/1996–2016/2017) average discharge (Linefur et al., 2019).

2.5 | Statistical analysis

Two-sided Wilcoxon rank sum tests were performed, where a *p* value <.05 was defined as significant. The same significance threshold was used for linear regressions and correlations. All sequential P fractionation results are presented as a mean of three subsamples. Standard deviations for each result are reported (Supplemental Tables S1 and S2).

To compare streambed sediment P fractions to concentrations in FSS, fluvial samples collected immediately prior to sediment core collections were used. Concentrations of P fractions in the top 4 cm of sediment were used because this depth is generally considered to be active (Reynolds & Davies, 2001). All statistical analyses were performed using R 3.6.2 (R Core Team, 2019).

3 | RESULTS

3.1 | Seasonal and between-catchment variations in FSS P fractions

Small temporal variations were observed for H_2O-P in U8 and E23 (Figure 2a). Proportions of H_2O-P were low and similar for all catchments (3.6–6.6% of TP; Figure 3a). The most prominent temporal change in concentration was recorded for the Fe-P fraction (Figure 2b), with a similar pattern in all catchments. The highest Fe-P concentrations (0.40–0.52 mg g^{-1}) were observed during summer; the lowest concentrations (0.13–0.23 mg g^{-1}) were observed during winter and spring (Figure 2b; Supplemental Table S1). Variation in Fe-P concentration was highest in C6 (CV, 56%; Supplemental Table S1), where it was the dominant fraction (Figure 3b).

In U8, Al-P was the dominant fraction (36%) and was significantly ($p < .005$) higher than in the other catchments (Figures 2c and 3c). For C6, the Al-P fraction was relatively stable over the study period (Figure 2c), with low total variation (0.16–0.26 mg g^{-1}; 11–27%) (Figure 3c). In E23 there seemed to be a monotonic decrease of Al-P with low total variation (0.09–0.30 mg g^{-1}; 10–16%) (Figure 3c).

Patterns for org-P were similar for all catchments, except for one peak in E23 in autumn 2018 (Figure 2d). In U8 and E23, the org-P fraction had the highest concentration variation among the different fractions (CV, 68 and 76%, respectively) (Figure 3d; Supplemental Table S2). In E23, org-P was the dominant fraction (32%) (Figures 2d and 3d). Organically bound P and Fe-P in FSS correlated positively ($R^2 = .37$, $n = 23$, $p < .05$). No other significant correlations between FSS P fractions were observed.

There was a significant difference between U8 and both C6 and E23 for the Ca-P fraction. However, the difference for Ca-P was not as clear as for Al-P (Figure 3e). Calcium-bound P was otherwise fairly stable for all catchments and had low total variation in concentrations (U8: CV, 30%; C6: CV, 6%, E23: CV, 23%) (Figures 2e and 3e; Supplemental Table S1), except for one peak in E23 and U8. Variation in TP concentrations was similar between catchments and had a clear seasonal pattern, with higher concentrations during summer and early autumn and a decrease during late autumn and winter (Figure 2f).

For U8, fractional H_2O-P concentrations were significantly lower ($p < .001$) than all other fractions, and Fe-P was significantly lower than the Al-P fraction, which was significantly higher than both the org-P and Ca-P fractions. At C6, the H_2O-P fraction was significantly lower ($p < .005$) than all other fractions. At E23, the H_2O-P fraction was significantly lower than all other fractions except for Al-P, which was significantly lower than the org-P fraction.

3.2 | Relations between water quality data, discharge, and FSS P fractions

Flow-weighted TP and SS concentrations correlated well when all catchments were used in the same linear regression ($R^2 = .71$, $n = 24$, $p < .001$). When tested separately, TP and SS correlated in U8 ($R^2 = .83$, $n = 12$, $p < .001$) and C6 ($R^2 = .90$, $n = 7$, $p < .01$) but not in E23 ($R^2 = .11$, $n = 5$, $p > .5$). The correlation between PP

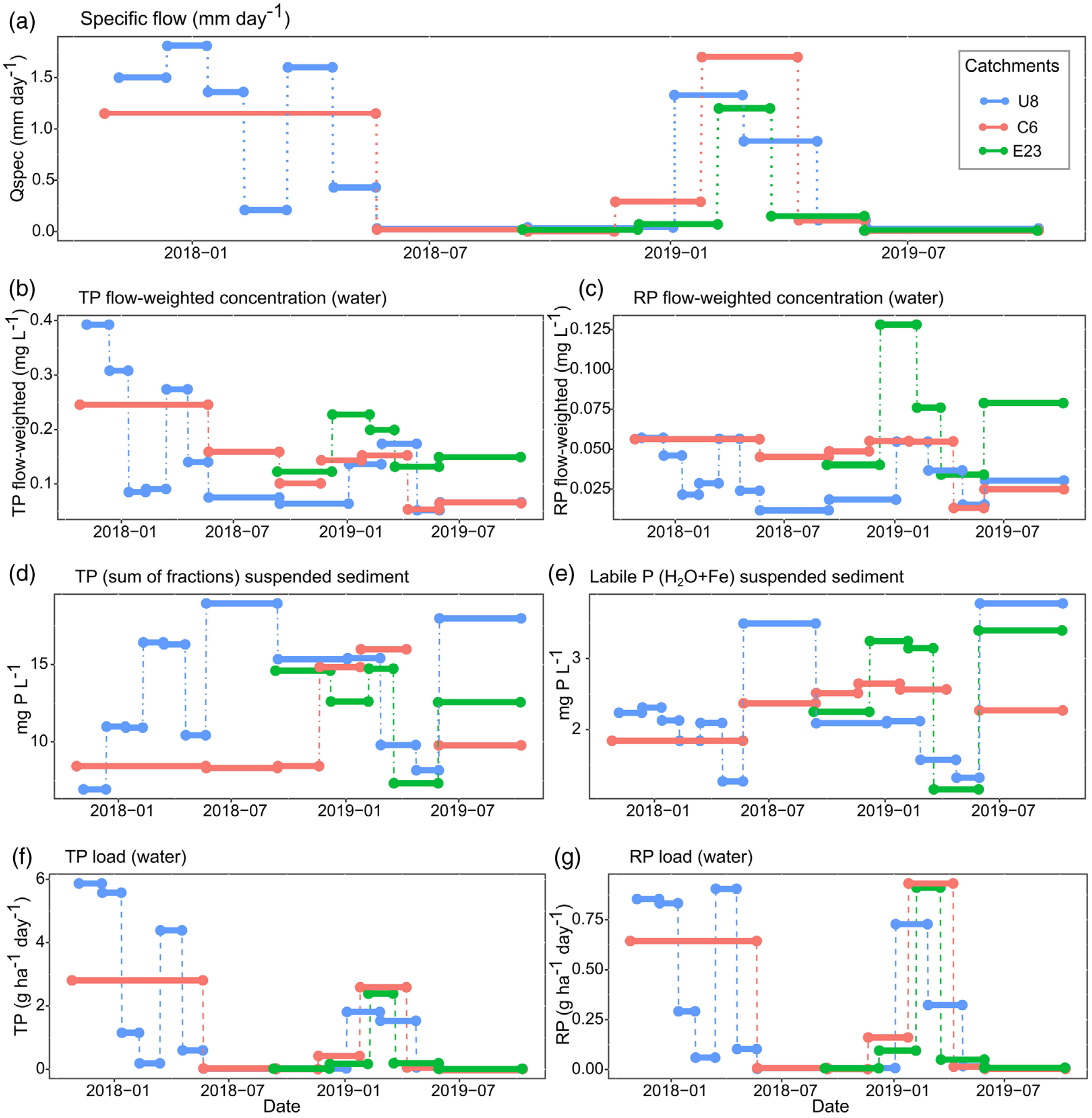

FIGURE 2 (a–e, g) Seasonal variations of the different phosphorus (P) fractions (Al-P, aluminum-bound P; Ca-P, calcium-bound P; Fe-P, iron bound P; H_2O-P, loosely bound P; org-P, organically bound P; TP, total P) over the sampling period in the three study catchments. Each bar in the graphs represents the concentration of the composite sample from the period the sampler was in the stream; different colors represent different catchments. The dotted lines are inserted to make it easier to differentiate the sampling periods. Sampling in E23 started approximately 1 yr after the other catchments, in September 2018 (green bars and dotted lines). Between April and May 2019 in catchment C6, not enough material was collected to perform the analysis, hence the tilted dotted red line. (f) Sum of the different fractions. Note that the y-axes are differently scaled for each fraction to enhance visibility of the individual changes of each fraction

and SS flow-weighted concentrations when all catchments were used in the same linear regression analysis was even higher ($R^2 = .80$, $n = 24$, $p < .001$) (Supplemental Figure S2). The only significant linear correlation between flow-weighted RP and SS concentrations was at U8 ($R^2 = .43$, $n = 12$, $p < .05$).

Annual specific discharge was well below average in all catchments in the second study year (Table 2; Supplemental Table S2; Supplemental Figure S3a). In all catchments, TP and RP loads followed the same pattern as the specific discharge (Supplemental Figure S3a,f,g). All catchments have similar flow-weighted TP concentrations, especially during

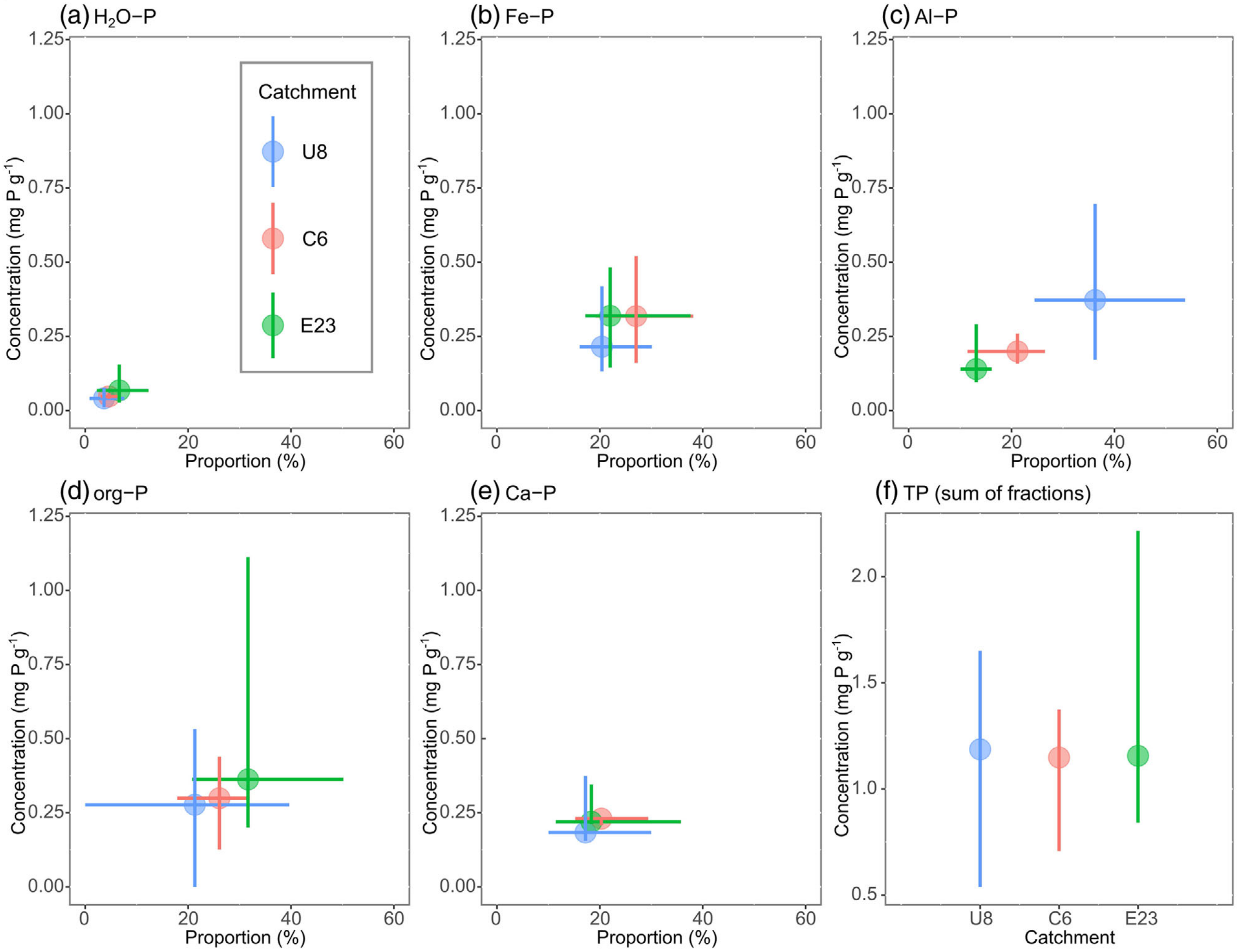

FIGURE 3 (a–e) Total variation of each phosphorus (P) fraction (Al-P, aluminum-bound P; Ca-P, calcium-bound P; Fe-P, iron bound P; H_2O-P, loosely bound P; org-P, organically bound P; TP, total P) in the fluvial suspended sediment and the sum of all fractions (f), colored by catchment. Dots represent the median value of each fraction, and the ends of the lines represent the minimum and the maximum values, respectively. The x-axis represents percentage of fraction, and the y-axis shows concentration of the fraction. In (f), only the variation in concentration of the sum of the fractions is displayed

the last study year (Supplemental Figure S3b). Flow-weighted water column and FSS TP concentrations showed no significant correlation (data not shown).

Flow-weighted water column RP concentrations seemed to follow a similar temporal pattern as FSS labile P (H_2O-P + Fe-P) (Supplemental Figure S3c,e). In C6 and E23, especially, a significant but weak correlation existed between the two variables ($R^2 = .16$, $n = 24$, $p < .05$) (Supplemental Figure S4). The RP/TP ratio was generally low for all catchments over the entire study period (Supplemental Table S2).

3.3 | Comparison of P fractions in FSS and the streambed active sediment layer

In all catchments, concentrations of different FSS P fractions immediately prior to sediment core collections generally followed concentrations in active streambed sediments (Figure 4) and were in the same concentration ranges. However, there were some clear differences.

In C6, Ca-P was higher in streambed sediment (64%) than in FSS (29%), where Ca-P was also the largest fraction (Figure 4a; Supplemental Figure S5). There were similar proportions (18 and 23%, respectively; Supplemental Figure S5) of the Fe-P fraction in the top streambed sediment and FSS. In streambed sediment, org-P was the smallest fraction, whereas H_2O-P was the smallest fraction in FSS (Figure 4a; Supplemental Figure S5).

In U8 streambed sediment, org-P, Al-P, and Fe-P concentrations and fractions were similar at 27, 30, and 30%, respectively (Figure 4b; Supplemental Figure S5). Relative fractions in FSS and streambed sediment were similar, except that org-P was the dominant fraction in FSS (40%) (Supplemental Figure S5).

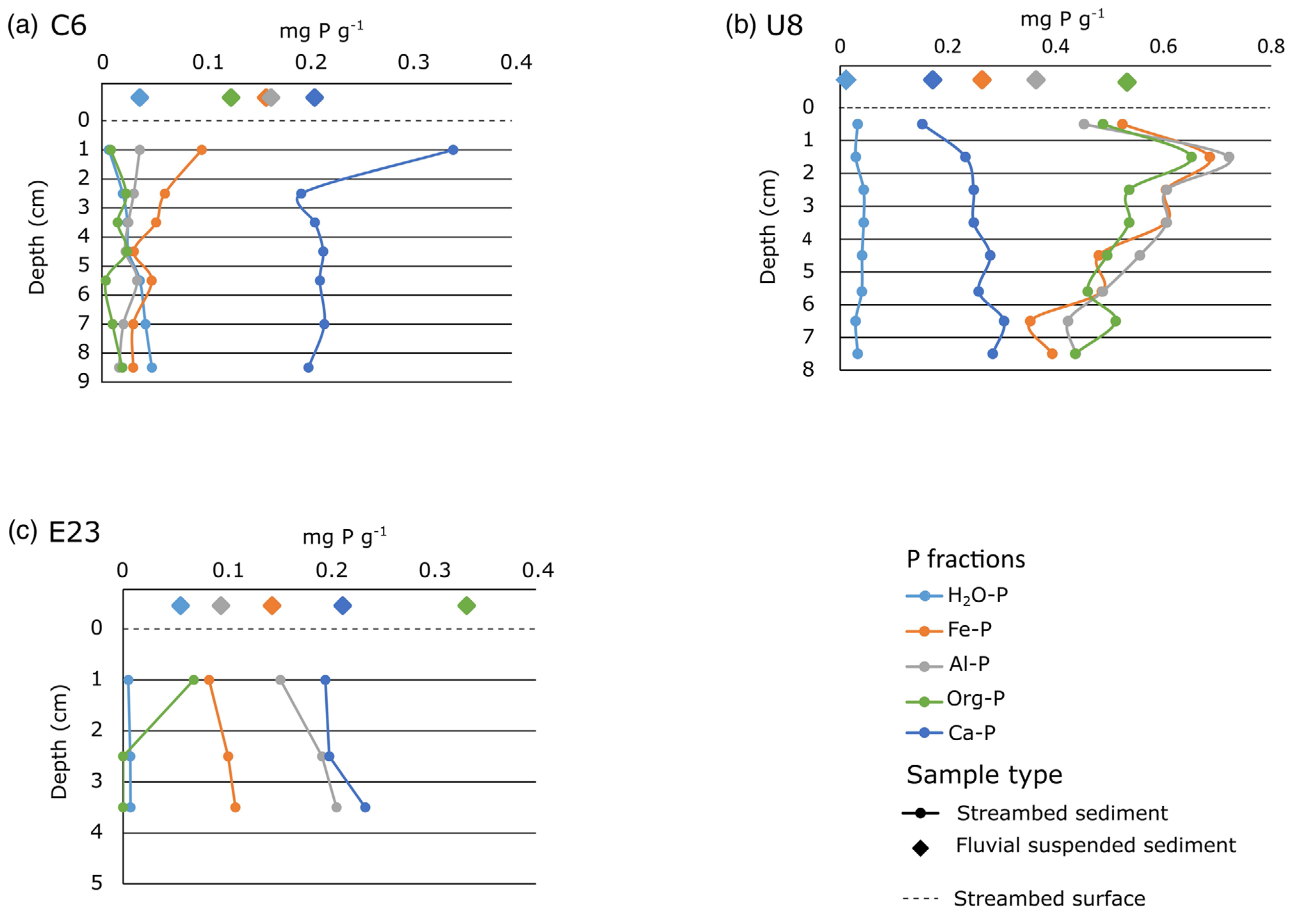

FIGURE 4 Concentrations of phosphorus (P) fractions (Al-P, aluminum-bound P; Ca-P, calcium-bound P; Fe-P, iron bound P; H_2O-P, loosely bound P; org-P, organically bound P; TP, total P) in sediment cores from the three catchments (dots and lines) and in fluvial suspended sediment (rhombi) from the period preceding sediment core collection. The different colors represent the different fractions, and a–c represent the different catchments. Each value is a mean value of triplicate samples from the same sediment sample

In E23, concentrations and fractions of org-P in FSS were clearly higher than in streambed sediment (Figure 4c), where Ca-P was dominant (Figure 4c). Calcium-bound P also contributed to a high proportion in the FSS (Supplemental Figure S5a). Concentrations and proportions of the Fe-P fraction were similar in the streambed and in FSS (Figure 4c; Supplemental Figure S5).

Organic matter content was markedly higher throughout the U8 streambed sediment profiles (20–30%) than in C6 and E23 (3 and 4%) (Supplemental Table S3). Streambed sediment profiles from U8 also had markedly higher water content (82–93%) than either C6 (40–64%) or E23 (31–35%) (Supplemental Table S3). No significant correlations between labile P in the active streambed sediment and corresponding RP flow-weighted concentrations were observed (data not shown).

4 | DISCUSSION

4.1 | Seasonal and between-catchment variations in P fractions in FSS and active streambed sediment

Although all catchments are dominated by clay soils and had similar FSS TP concentrations, the dominant P fraction in FSS differed between catchments. The identified differences between catchments are described below, and their probable explanations fullfil Aim 1 of the study. There was also a clear temporal variation in all catchments, especially for Fe-P, answering Aim 2.

4.1.1 | Labile P in FSS and streambed sediment

Bioavailability of different P fractions in sediments depends on environmental conditions, such as pH (Lukkari et al., 2007), redox conditions (Records et al., 2016; Sharpley, 1993), and turbulence (Withers & Jarvie, 2008). The labile P fraction (H_2O-P and Fe-P) together with org-P has been termed as "potentially bioavailable P" (Kaiserli et al., 2002). For H_2O-P, the most bioavailable fraction is PO_4^{3-}, which is directly available for uptake by aquatic organisms (Zhou et al., 2001). However, H_2O-P is likely to be of minor importance in these catchments due to its low concentrations. The Fe-P fraction is usually the largest pool of bioavailable P due to its high release potential from sediment undergoing changes in redox status (Li et al., 2016). Reducing conditions can occur in streambed sediment due to intrusion of anoxic groundwater (Krause et al., 2011) but also due to high microbial and biological activity in shallow, stagnant ditches during summer low flow (Sallade & Sims, 1997). Organically bound P can become bioavailable because it can be mineralized to RP (Reitzel et al., 2005; Schütz et al., 2017). Aluminum-bound P is considered stable in the pH range of surface waters (Stumm & Morgan, 1995). Calcium-bound P is usually contained in mineral lattices and hence is usually nonavailable or even considered inert (Lukkari et al., 2007). There is a need for further studies on the effect of environmental conditions on potential P bioavailability in FSS and streambed sediment in headwater streams because the environmental conditions affecting P release in headwater streams can differ from larger rivers as well as lakes (Lannergård et al., 2020).

In all catchments, the increases in FSS Fe-P during summer and early autumn corresponded to periods of lower specific discharge (Figure 2b; Supplemental Figure S3a). This decrease in discharge, along with higher temperatures and increased biological activity, could change the redox potential in streambed sediments, resulting in a reducing environment conducive to dissolution and in the release of ferric Fe and phosphate into the water column (Records et al., 2016; Sallade & Sims, 1997; Smolders et al., 2017). When sediment is mobilized from the streambed into the potentially more aerobic water column environment, Fe oxidation may occur converting Fe^{2+} to ferric Fe^{3+}, which can react with water column RP. Such Fe oxidation and RP sorption may occur on fluvial sediment particles, resulting in higher FSS concentrations and lower water column concentrations. The increase in FSS Fe-P is prominent in both summers, with a noticeable decrease during winter and spring months, when the specific discharge is higher (Figure 2b). Flow-weighted water column RP concentrations are also lower during summer, especially at the two smaller streams (U8 and E23), supporting this theory (Supplemental Figure S3c). Because the C6 stream is much larger than the other two streams, the flow rarely stagnates, staying in constant motion, with potentially less frequent anoxic conditions close to the streambed sediment surface. High amounts of FSS Fe-P during low flow periods, at times where anoxic conditions are more common, does suggest an increase in bioavailable P during the growing season. However, it is unknown whether these FSS particles would remain in suspension to settle in the streambed or be transported further downstream to receiving waters, potentially contributing to an increased P pool in receiving sediments.

The Fe-P fraction was dominant in FSS in C6 (Figure 3b), where soils around the catchment outlet are mainly silty clay loam. Catchment bedrock mineralogy (Table 1) does not imply a high Fe content, and there are no available measurements of Fe content in catchment soils or sediment. However, a probable explanation for the dominance of Fe-P in C6 is the presence of Fe hydroxides/oxides in the soil and sediment. Streambed sediment in C6 is similar to the FSS in that sense, with the Fe-P fraction being the second largest fraction, although decreasing with depth throughout the sediment profile. Earlier studies have found strong correlations between Fe and P in streambed sediment (Palmer-Felgate et al., 2009) and suspended sediment (Cooper et al., 2015), suggesting that when Fe is present in a higher amount under appropriate redox conditions, a majority of P will be bound to iron hydroxides/oxides. The national soil survey data show that Fe-P content in topsoils in this region, measured after extraction with ammonium lactate, was above the national average and the second highest among 20 studied regions of Sweden (Djodjic, 2015). Cooper et al. (2015) found strong correlations between Fe and P in suspended particulate matter in streams during base flow but not during storm flow. This supports our results with higher FSS Fe-P during low flow periods and lower Fe-P concentrations during high flow periods. Furthermore, McDowell et al. (2020) reported a decrease in streambed Fe-P with sediment depth, which was also observed in our study. Phosphorus release due to Fe hydroxides/oxides dissolution with progressively more reducing sedimentary conditions at depth may explain this pattern (Parsons et al., 2017).

The low concentrations and low variation in FSS H_2O-P in all catchments could be related to the low solubility of P because the pattern is similar in the streambed sediment. The FSS is in constant motion; thus, it is likely that any H_2O-P will either be taken up directly by biota or further transported. Similar behavior is likely to occur for H_2O-P in the active streambed sediment. Simpson et al. (2019) also found low concentrations of loosely sorbed P when studying the effects of different pretreatments on streambed sediment. McDowell et al. (2020) found low concentrations of H_2O-P in streambed sediment, with Fe-P as the general dominating fraction at their sampling sites. These low H_2O-P concentrations (or corresponding fractions in other fractionation schemes) seem to be quite consistent for streambed sediment and lake sediment (Kaiserli et al., 2002; McDowell et al., 2020; Pettersson, 1998).

4.1.2 | More stable P (Al-P and Ca-P) and geological conditions

In the smallest catchment (U8), Al-P is the dominant FSS P fraction, and the Al-P concentrations were markedly higher than in C6 and E23 (Figure 3c). In contrast, U8 had lower Ca-P concentrations than C6 and E23. Bedrock in U8 consists mainly of granodiorite-granite (Table 1) that contains plagioclase feldspar, generated from Al-containing minerals. Similarly, the Al-P fraction is high in U8 streambed sediment, which supports a geological weathering contribution to the Al-rich environment. The presence of particles such as Al-silicates (clay minerals) (House et al., 1998) and especially Fe and Al hydroxides and oxides (Zhang & Huang, 2007) will make RP react and bind to mineral or particle surfaces (Withers & Jarvie, 2008). The average clay content in U8 is approximately 10% higher than in C6 and E23 (Sandström et al., 2020), with Al as a primary element in most clay minerals. This also suggests more Al-P because clay minerals in soils having Al sheets in their structure have high P sorption capacity, depending on pH conditions (Gérard, 2016), where U8 has the lowest pH (Table 2). High P sorption to Al-containing clay minerals has also been found in lake sediments (Tonello et al., 2020). The C6 bedrock is instead mainly quartz arenite, and in E23, it is mainly granite. However, all catchments have similar mineralogy consisting of quartz, feldspar, and mica (Table 1). This implies that the bedrock might have less influence on P fractions in C6 and E23 because the mineralogy does not seem to be reflected in the P fractions as prominently as in U8.

The FSS Ca-P fraction was similar across all catchments, proving that parent soil material is quite comparable. However, there was one Ca-P peak in U8 at the end of the study period. This could be related to the practice of structural liming in the catchment. The practice, which is a popular mitigation measure to reduce P losses from clay soils in Sweden, is usually performed around that time of the year (Geranmayeh, 2017). Although no recorded data are available for structural liming during the study period in U8, structural liming was used in the catchment prior to 2013 when a product containing approximately 15% $Ca(OH)_2$ was applied at a rate of 4 t ha^{-1} (Malgeryd et al., 2015). In U8 streambed sediment profiles, Ca-P seemed to increase with sediment depth (Figure 4b), whereas in C6, the active streambed sediment had markedly higher Ca-P concentrations that then decreased and stayed more constant down through the core (Figure 4a). Calcium-bound P represents the most tightly bound P and is thus more stable deeper in the streambed sediment, where less, if any, exchange with the overlaying water column occurs.

4.1.3 | Organically bound P

The U8 catchment had a markedly higher OM content in streambed sediment (Supplemental Table S3) than C6 and E23. This was not reflected in the FSS, where OM content was more similar among the catchments (Supplemental Table S1). The low OM content in the streambed sediments compared with FSS in E23 and C6 was also reflected in the org-P fractions, with lower concentrations and proportions of org-P in streambed sediment but still moderately high concentrations of FSS (Figure 4b,c; Supplemental Figure S5). In U8, with a closer match in org-P content between the streambed and FSS, there was a lot of vegetation in the stream compared with C6 and E23. This catchment is also the smallest one (Table 1) and has periods of very low flow where the stream almost dries out. This contributes to slowing down the movement of water and FSS. In the larger C6 catchment, the stream flow was almost constant, and the FSS will likely not settle in the streambed for a longer time period. The vegetation itself in U8 is probably a contributor to the higher OM content in streambed sediment but could also contribute to trapping more OM by slowing flows, allowing it to settle in the streambed sediment (Cotton et al., 2006; Verschoren et al., 2017). In contrast, vegetation cover is much poorer in the coarser E23 streambed.

In E23, org-P was the dominant fraction in FSS, where it displayed a high variation in concentration but not as much variation in distribution (Figure 3d). The high concentration variation might be attributed to the peak in org-P at the start of the study period, although concentrations remained relatively high throughout, making the variation in distribution smaller. In comparison to the other catchments, FSS in E23 have generally higher org-P concentrations. The difference between the streambed and FSS in both org-P concentrations and OM content also suggests an external source of org-P. Differing from C6 and U8, E23 had a higher stocking density, higher manure inputs, and a horse paddock with a steep slope down to the stream located just upstream the sampling point (Ulén et al., 2011). The near-stream zone by the outlet of E23 is forested, unlike the other catchments that have mainly agricultural fields surrounding the outlet. The comparatively high FSS org-P concentration in E23 could reflect timing of manure application to the fields or direct inputs from the horse paddock, which are potential hotspots for P leaching (Parvage et al., 2015). This could have led to an increase in particulate P transport to the stream and thus to a peak in FSS org-P, which was not reflected in the streambed sediment. The weak but significant correlation between org-P and Fe-P might indicate that the org-P interacts with and contributes to the pool of bioavailable P.

4.1.4 | TP and general patterns between streambed and FSS

As stated in Section 3.3, P fractions and concentrations in FSS sampled immediately prior to streambed sediment sampling are in the same concentrations ranges as in the active streambed sediment having similar proportions of dominating fractions, especially in U8 and C6 (Figure 4). In contrast, mean values of FSS P fractions and concentrations over the whole study period did not reflect the active streambed sediment as well. Due to the marked variations in FSS P fractions and concentrations over time, a comparison with the time-aggregated mean value would not be representative. Unfortunately, due to high flow conditions, the C6 FSS sample was collected over a longer time period than in U8 and E23 and thus may not be as representative. Our study supports Palmer-Felgate et al. (2009), who found a significant correlation between TP in streambed sediment and P concentration in suspended sediment in catchments having different agricultural land use intensity, but the relationships with stream water P concentrations were not as clear.

All catchments had approximately the same median FSS TP concentrations (Figure 3f) and had similar seasonal variation. Selection of catchments was based on long-term monitoring indicative of high transport of P and SS. Interestingly, the two smallest catchments had the greatest total variation in FSS TP concentrations, even though the concentrations varied in all catchments. Notably, this study has important implications for understanding the P cycle in headwater catchments. It supports the argument of Bol et al. (2018) for a greater focus on headwaters due to their high variability in P losses (Haygarth et al., 2012). Moreover, it demonstrates that focusing solely on TP dynamics will provide an incomplete indication of P mobility and reactivity among headwaters because it neglects variation in FSS P fractions and bioavailability. The variation in FSS P fractions has been shown by increases in Fe-P during the growing season.

4.2 | Water quality and P in FSS

The observed correlations between flow-weighted PP and SS concentrations (Section 3.2; Supplemental Figure S2) support earlier work in these catchments (Sandström et al., 2020). The lack of significant relationships between SS and RP (except for in U8) and the low loads and concentrations of RP reflect the dominance of PP in these catchments.

Comparisons of water column P fractions with corresponding FSS P fractions to address Aim 3 of our study showed a weak correlation between FSS labile P and water column RP (Supplemental Figure S4). This illustrates the complexity of P circulation in small headwater catchments, where no clear relationship to flow was visible for all catchments. The U8 catchment had two outlying sampling occasions with low discharge (high FSS labile P and low water column RP concentrations; Supplemental Figure S4) and was the only catchment with a significant relationship between RP and SS. This suggests that during high discharge, some water column RP might consist of small clay particles or colloids that will pass through a filter and be identified as RP instead of PP. Indeed, Ulén (2003) showed that the colloidal P fraction was important in surface runoff, tile-drainage water, and stream water in a clay-dominated catchment in central Sweden. The significant but weak correlation between labile P and water column RP could also indicate the importance of external sources, such as manure.

In U8, where redox-independent Al-P was the dominant P fraction, FSS TP concentrations seemed to display an opposite pattern to water column TP concentrations (Supplemental Figures S3b and S5d). Here also FSS TP concentrations seemed to be highest during low flow periods, whereas water column TP was also low. This could be explained by low SS losses during low flow periods, resulting in P enrichment on SS particles (Sandström et al., 2020). It also points toward an exchange between FSS and the water column, where P could theoretically be sorbed to suspended sediment particles, increasing the concentration on the particles during periods with low concentrations of water column TP.

During the second study year there was a drought in Sweden (SMHI, 2018), resulting in low flows in all catchments (Supplemental Figure S3a), especially E23 (Supplemental Table S1). The drought affected TP loads and concentrations, with lower levels than usual in all catchments. Although the effect of the drought on FSS P fractions is difficult to establish from the 2-yr study period, effects of flow on redox conditions and temporal variability in the Fe-P fraction are visible.

5 | CONCLUSIONS

Based on time-integrated sampling of FSS in the three headwater catchments covering different seasons in combination with water quality monitoring data and streambed sediment sampling, the following conclusions can be drawn:

- There is a significant difference in the distribution of FSS P fractions in the three catchments. The differences are likely related to differences in geology, soil clay content, external P sources and flow conditions.
- There are seasonal variations, especially for Fe-P, in FSS P fractions and concentrations in all catchments that may

be due to seasonal changes in flow, temperature and redox conditions.

- Water column TP concentrations are not clearly related to FSS TP, with an almost opposite temporal pattern. There is a weak but significant positive correlation between water column RP and FSS labile P.
- The P fractions found in FSS are in the same concentration ranges as P fractions in the streambed sediment; however, the distribution of different P fractions differs. Although the dominant P fractions were the same in FSS and streambed sediment for C6 (Ca-P) and U8 (org-P, Al-P, and Fe-P), the fractional distribution in streambed sediment and FSS show no resemblance in E23.

This seasonality of FSS P fractions shows the importance of measuring different P fractions and not only TP during ecologically sensitive periods. Because the FSS may act as the interface between the streambed sediment and the water column, measuring and reporting P fractions in FSS will lead to a better understanding of the biogeochemical processes controlling P dynamics in agricultural headwaters.

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AUTHOR CONTRIBUTIONS

Sara Sandström, Conceptualization, Data curation, Formal analysis, Investigation, Methodology, Project administration, Validation, Visualization, Writing-original draft, Writing-review & editing; Martyn N. Futter, Conceptualization, Data curation, Methodology, Supervision, Writing-review & editing; David W. O'Connell, Conceptualization, Funding acquisition, Methodology, Writing-review & editing; Emma E. Lannergård, Formal analysis, Investigation, Writing-review & editing; Jelena Rakovic, Formal analysis, Writing-review & editing; Katarina Kyllmar, Conceptualization, Methodology, Supervision, Writing-review & editing; Laurence W. Gill, Funding acquisition, Writing-review & editing; Faruk Djodjic, Conceptualization, Data curation, Investigation, Methodology, Project administration, Supervision, Writing-review & editing.

CONFLICT OF INTEREST

No conflicts of interest are relevant for this study.

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