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Particulate phosphorus and suspended solids losses from small agricultural catchments: Links to stream and catchment characteristics

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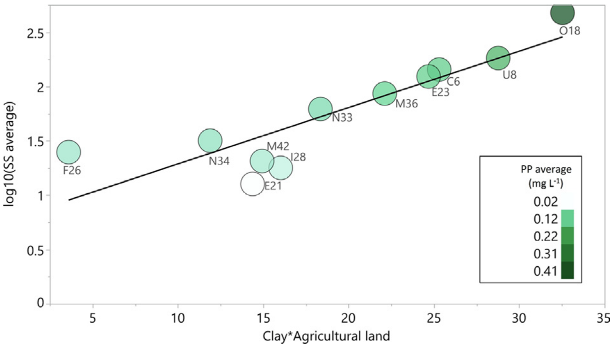
HIGHLIGHTS

- Identification of catchment and in-stream factors correlating to phosphorus losses.
- Long-term time series of flow and water quality from 11 headwater catchments.
- Principal components analysis, linear regression and trend analysis were performed.
- Soil texture (primarily clay content) strongly linked to losses of suspended solids.
- Suspended solids form a clear, monotonic relationship with particulate phosphorus.

GRAPHICAL ABSTRACT



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ABSTRACT

Excessive phosphorus (P) inputs from agriculture are well established as a contributor to freshwater eutrophication. Decreasing these inputs is an important step in improving the ecological state of impaired waters. Particulate P (PP) is a significant contributor to diffuse P inputs in agricultural catchments. Identifying the main correlates for PP losses is an important step in reducing these inputs. However, there are few studies of long term temporal and spatial dynamics of PP in agricultural streams. Here, we investigate the relative importance of hydrology, catchment characteristics and geochemistry on PP concentrations and fluxes in agricultural headwaters. We evaluate long-term monitoring data from eleven small (<35 km^2) Swedish catchments with at least seven years of measured flow and flow proportional water quality sampling. Using parametric and non-parametric regression together with principal components analysis (PCA), we identify in-stream and catchment variables relevant for predicting PP concentrations, e.g., suspended solids concentrations (SS), soil texture and average catchment soil P content, measured as ammonium lactate/acetic acid extractable P (P-AL). We show that PP is primarily correlated to SS concentrations, which in turn are correlated to average clay content and land use. However, the SS:PP relationships differ between catchments. No correlation between PP concentrations in the stream and soil P content was found. An increasing clay content decreases the slope of the relationship between SS and PP, i.e., in catchments with higher clay content, less PP is transported per unit SS. The

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PP/SS ratio increased significantly ($p < 0.05$) over time in four catchments, despite limited changes in SS or PP concentrations. Our study highlights the importance of long time series since the enrichment of P on SS in the streams is only detected when using long term monitoring data.

1. Introduction

Excessive phosphorus (P) inputs are well established as a contributor to freshwater eutrophication (Sharpley et al., 2015). Decreasing these inputs is an important step towards improving the ecological state of impaired waters, and thereby reaching goals set by, e.g., the European Water Framework Directive (WFD) (European Commission, 2000). To decrease P loading to surface waters, the headwater sources and instream processes influencing P need to be identified. Agriculture has been identified as an important non-point source of P to degraded waters in many countries, with an episodic and spatially highly variable input pattern (Sharpley et al., 2009, Ulen et al., 2010, Sharpley et al., 2013, Kronvang et al., 2005, Withers and Jarvie, 2008). This variability in P inputs to streams and lakes is one of the main issues when designing measures against P losses from agricultural land (Sharpley et al., 2011, Thomas et al., 2016, Djodjic and Markensten, 2018). In northern Europe, there is a significant focus on P control measures to mitigate Baltic Sea eutrophication. To successfully address eutrophication of the Baltic, coordinated regional efforts are needed across the Nordic and Baltic states, but many of the actual actions must take place on the smaller scale of individual farms.

Globally, a significant proportion of the P applied to crops accumulates in soils. Some of this P is subsequently transported by surface water runoff and drains into nearby lakes, rivers and streams, largely in different particulate mineral and/or organic forms (Walling et al., 1997; Bowes et al., 2003; Baulch et al., 2013; Cooper et al., 2015). This accumulated 'legacy phosphorus' gradually leaches into the water in a more biologically available soluble form, i.e., PO_4^{2-}, thereby impacting surface water quality for decades. This time lag of P release to aquatic ecosystems means that the effects of nutrient abatement measures for the reduction in P loads may be significantly delayed (Sharpley et al., 1994, 1999; Brogan et al., 2002).

An earlier compilation of water quality monitoring data from Swedish streams (71–4187 km^2) showed the high temporal variability and importance of particulate P (PP) as a vector for P transport (Persson, 2001). The importance of suspended solids (SS) for P transport was also highlighted, and it was established that land management (% arable land in the catchment) was linearly related to PP ($R^2 = 0.85$) and SS concentrations ($R^2 = 0.76$). A significant amount of TP transported in rivers, is in particulate form in the UK, Sweden and other regions (Ballantine et al., 2008, Persson, 2001, Sharpley et al., 2013) which again highlights the importance of understanding mechanisms of PP loss.

Other studies have addressed PP export from agricultural headwater catchments (Walling et al., 1997; Ballantine et al., 2008). Here, we focus on the relationships and correlations between TP-, PP loss and catchment characteristics (i.e., clay content and soil texture). Kyllmar et al. (2014) studied long-term patterns in nutrient losses (TP and total N (TN)) in small Swedish agricultural streams and established a linear relationship between TP and catchment clay content ($R^2 = 0.68$). These losses often occur via erosion and surface runoff, but also via preferential macropore flow where PP is mobilized and transported during storm events (Djodjic et al., 1999). Almost half (47%) of Sweden's agricultural fields are artificially drained (Statistics Sweden, 2017a), where P is rapidly transported through macropores to drainage pipes and thereafter transported directly to the stream. An earlier study of temporal trends in nutrient loads in Swedish agricultural streams (Kyllmar et al., 2006), based on manual fortnightly sampling, has shown significant downward trends for phosphate P (in 8 out of 24 catchments) and PP (in 2 out of 24 catchments), but also a significant upward trend for PP in two catchments. Similar studies of nutrient losses in agricultural catchments have been made elsewhere. Bechmann et al. (2008) studied losses of TN, TP and SS from eight agricultural catchments in Norway, focusing on long term trends and the effects of agricultural management practices. They found a linear relationship between SS and TP for three catchments, ($R^2 = 0.84–0.97$), with a different slope for the relationship in each catchment (Bechmann et al., 2008). Baulch et al. (2013) reported on spatio-temporal patterns of P loss from a mixed land use catchment in Canada with similar climate and soil properties as are found in Sweden. Crossman et al. (2019) evaluated the connection between catchment measures and improvements in downstream water quality. Wilcock et al. (2013) studied both TP and N in New Zealand agricultural streams and their change over time after introducing measures to reduce nutrient losses from dairy farms, highlighting the importance of long-term monitoring data for water quality assessment as well as for determining trends.

Larger rivers in Sweden are well monitored (Fölster et al., 2014). However, smaller, poorly monitored headwaters comprise around half of the stream network length (Bishop et al., 2008). Bishop et al. (2008) called these streams "aqua incognita, the unknown headwaters", where the water begins its journey towards larger streams and lakes. Bol et al. (2018) highlight the need to focus on agricultural headwater catchments, since variability in both dissolved and particulate P fluxes is higher in headwater catchments and the processes controlling P in headwaters have a large influence on the larger catchment.

The environmental monitoring program in Sweden conducts long-term monitoring on nutrient losses and water quality in several, small, contrasting agricultural catchments in southern Sweden (Kyllmar et al., 2014). Similar monitoring programs exists in other Nordic and Baltic countries (Vagstad et al., 2004; Bechmann et al., 2008; Granlund et al., 2005). Previous Nordic and Baltic studies have focused primarily on losses of TN and TP, while measurements of different P-species and their connection to catchment characteristics are rare outside the Swedish and Norwegian monitoring programs. The Swedish monitoring program has been collecting water quality data with fortnightly sampling in 21 agricultural catchments for a time period of at least seven years, and in some catchments for over 25 years.

Since only a few headwater catchments are intensively monitored, there is a need to scale up results from existing monitoring programs. Therefore identification of factors influencing levels and forms of P losses in agricultural headwaters are needed. Specifically, we hypothesize that additional factors beyond clay content, e.g., SS and percentage of agricultural land, are needed for adequate prediction of stream PP concentrations and fluxes in agricultural headwaters. Here, we investigate (i) how TP and PP concentrations, based on biweekly flow proportional water quality measurements, are affected by other in-stream and catchment factors, (ii) to identify correlates of PP concentrations in agricultural streams and to deepen the understanding of these connections.

This study aims to gain improved understanding of the processes and variables controlling P mobilization and transfer in the unknown agricultural headwaters.

2. Material & methods

2.1. Monitoring catchments

The environmental monitoring program includes 21 small agricultural catchments (area < 35 km^2), where some catchments have been monitored since the 1990 s, with focus on nutrient losses (Kyllmar et al., 2014). The main purpose of the monitoring program is to evaluate agricultural policy measures (subsidies, rules and agricultural advisory campaigns), follow the effects of these policies on farmer's practices on their fields and in turn how that affects the water quality in draining streams as well as increasing the knowledge of these connections and how they evolve over time. For this project, 11 out of the 21 catchments in the monitoring program were chosen (Fig. 1). The 11 catchments all had at least seven years of biweekly flow proportional water quality measurements and measured streamflow (Table 1).

The data used in the analysis are mainly from the same time period (Table 1). Flow-proportional sampling for three catchments started in 2007 and monitoring of N33 was discontinued in 2014. Data for crops grown in the catchments generally covers the same time period as the water quality data, except for E23, N33 and U8 where crop data was available for shorter time periods (2006–2013, 2007–2013 and 2007–2013, respectively).

2.2. Water quality sampling and laboratory analysis

The water quality data used in this project was based on automatic flow proportional water sampling (Kyllmar et al., 2014). Samples are collected and stored in a 10 L glass bottle from which

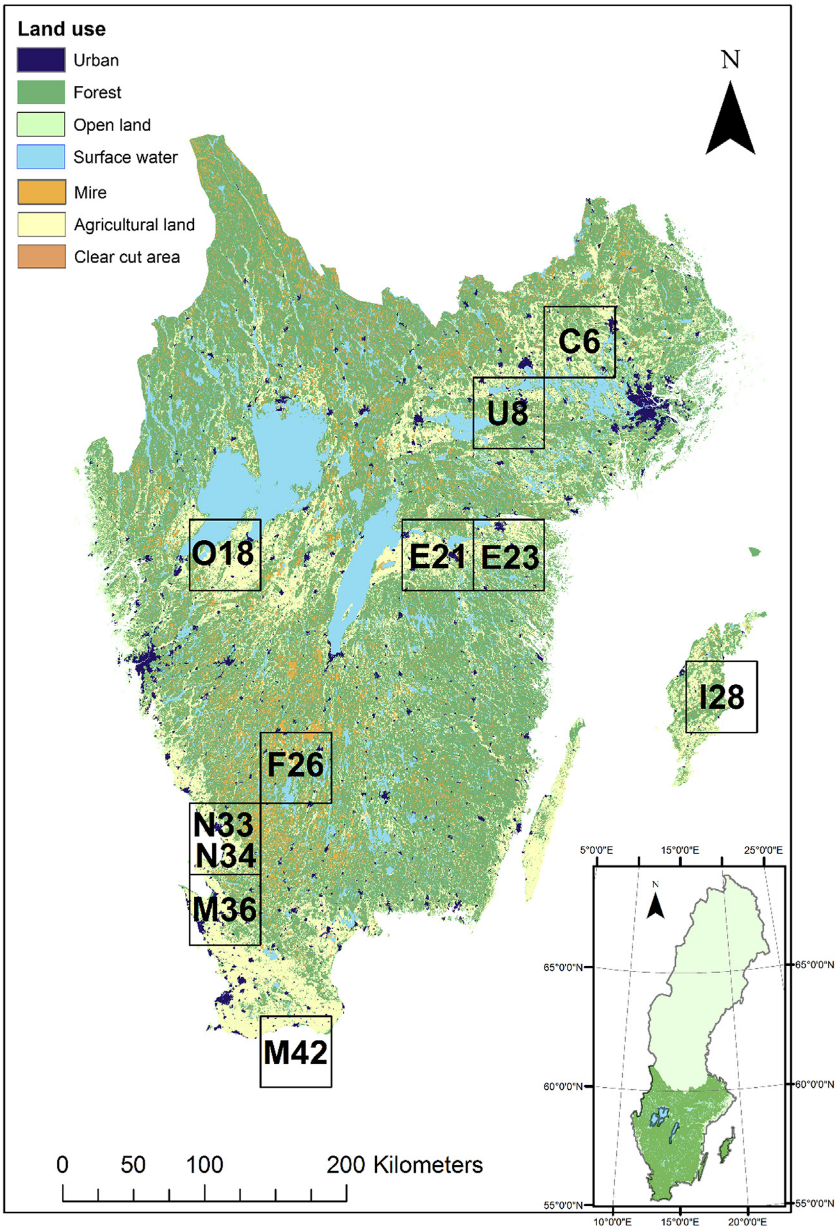

Fig. 1. Approximate location of all catchments used in the analysis, zoomed in to southern Sweden, with a land use map as a background. Latitude and longitude lines are shown in the inset figure, displaying the whole of Sweden.

Table 1
Catchment characteristics for all study catchments (Linefur et al., 2017; Kyllmar et al., 2014). Average annual runoff is given for each catchment's respectively monitoring period. Precipitation and temperature are normal values from SMHI 1961–1990 (SMHI, 2001).

Catchment ID	Area (km^2)	Arable land (%)	Stocking density (AU ha^{-1})	Scattered households (pers. km^{-2})	Monitoring period for water quality data	Dominant soil texture class (USDA)	Precipitation (mm)	Temp. (°C)	Runoff (mm)
C6	33.1	59	0.05	10	2004–2017	Clay loam	623	5.5	220
E21	16.3	89	0.05	9	2004–2017	Sandy loam	506	6.0	157
E23	7.4	54	0.6	7	2007–2017	Clay	594	6.3	181
F26	1.8	70	1.2	33	2005–2017	Loamy sand	1066	6.2	482
I28	4.7	84	0.5	11	2005–2017	Sandy loam	587	6.9	156
M36	7.9	86	0.3	37	2004–2017	Clay, sandy loam	719	7.6	277
M42	8.2	93	0.1	10	2006–2017	Sandy loam, loam	709	7.7	282
N33	6.6	87	0.1	7	2007–2014	Loam, sandy loam	823	7.1	251
N34	13.9	85	0.4	19	2004–2017	Sandy loam and silt loam	823	7.2	372
O18	7.7	92	0.05	8	2004–2017	Clay	655	6.1	332
U8	5.7	56	0.05	11	2007–2017	Clay	539	5.9	206

composite samples are taken every two weeks, followed by emptying the glass bottle (Kyllmar et al., 2014). The intensity of sub sampling is dependent on flow, with more sub samples collected during high flow periods, and time proportional sampling is used during low flow periods, with two sub samples per day. The sampler is calibrated to cover high flow periods for each specific station. A full description of the sampling protocol is presented in Kyllmar et al. (2014).

Samples are analyzed at a water laboratory certified by the Swedish Board for Accreditation and Conformity Assessment (SWEDAC), following Swedish Standard Methods (Kyllmar et al., 2014). Several different parameters are analyzed, the ones used in this study are TP, phosphate phosphorus (DP), SS and the concentration of PP which is the filtrate fraction of TP.

Sediment and P loads are calculated based on daily average values of water flow and estimated daily nutrient concentrations extrapolated backwards from the fortnightly composite samples, using the same concentration for the entire two week period between sampling occasions. Measured daily flows are multiplied by estimated daily concentrations to obtain daily loads, which are summarized to monthly and annual loads. Area specific loads (kg km^{-2}) are calculated by dividing the load by the catchment area. The flow weighted concentrations (mg L^{-1}) used in the data analysis are obtained by dividing the calculated load (kg km^{-2}) by the runoff (mm) for the relevant time period (Table 2). In some catchments, the stream is periodically dry and hence some months lack data. Average annual runoff was calculated as the average of the annual runoffs from the evaluated time period.

To explore the influence of land use and crop distribution on P losses, three crop type groups were created: 1) Ley 2) Cereals & Rape and 3) Other Crops. The three groups reflect different management intensities and P loss potential. The average percentage of each crop type over the time period was used to get a value for each group. The "Ley" group includes both ley and fallow. Ley is the most common individual crop in Sweden and has considerably lower P losses compared to cereals (Johnsson et al., 2016). The "Cereals & Rape" group covers the largest area in all studied catchments and includes crops with similar management systems and P losses. Finally, crops with higher P inputs and higher risk for P losses, e.g., potato and sugar beets, were assigned to the category

Table 2
Additional catchment data used in the PCA. PP = particulate phosphorus, SS = suspended solids, TP = total phosphorus, DP = PO4-phosphorus, P-AL = Phosphorus extracted by ammonium lactate/acetic acid, representing soil P content. The crop types Ley, Cereals & Rape and Other crops are given as a percentage of arable land. All concentrations are given as total average values with the standard deviation in parentheses. FV = Factor of variation, describes the relationship between the highest and the lowest value among the catchments.

ID	TP (mg L^{-1})	PP (mg L^{-1})	DP (mg L^{-1})	SS (mg L^{-1})	PP/SS	PP/TP	P-AL (mg/100 g)
C6	0.21 (0.06)	0.15 (0.06)	0.05 (0.01)	143.6 (65.3)	1.0E−03 (1.8 E−04)	0.61 (0.17)	6.34 (1.73)
E21	0.059 (0.02)	0.02 (0.01)	0.03 (0.02)	12.7 (5.0)	1.9 E−03 (7.8 E−04)	0.49 (0.28)	10.30 (5.08)
E23	0.28 (0.02)	0.14 (0.03)	0.12 (0.02)	123.6 (39.2)	1.1 E−03 (2.0 E−04)	0.46 (0.16)	7.41 (12.75)
F26	0.12 (0.05)	0.06 (0.03)	0.03 (0.01)	24.9 (18.0)	2.6 E−03 (1.5 E−03)	0.54 (0.14)	4.17 (5.95)
I28	0.18 (0.06)	0.04 (0.02)	0.12 (0.05)	17.8 (5.9)	2.3 E−03 (1.0 E−03)	0.26 (0.14)	9.01 (7.96)
M36	0.20 (0.04)	0.12 (0.04)	0.06 (0.009)	86.2 (21.3)	1.4 E−03 (4.2 E−04)	0.55 (0.14)	9.97 (3.26)
M42	0.15 (0.03)	0.06 (0.02)	0.08 (0.03)	20.6 (10.7)	2.8 E−03 (9.7 E−04)	0.37 (0.15)	12.60 (5.16)
N33	0.16 (0.02)	0.09 (0.02)	0.05 (0.01)	62.0 (17.4)	1.5 E−03 (2.5 E−04)	0.57 (0.18)	10.27 (5.55)
N34	0.10 (0.03)	0.07 (0.03)	0.02 (0.004)	31.8 (10.2)	2.3 E−03 (5.8 E−04)	0.70 (0.15)	14.24 (7.38)
O18	0.50 (0.16)	0.41 (0.15)	0.07 (0.02)	476.9 (220.8)	8.6 E−04 (2.7 E−04)	0.65 (0.20)	5.28 (3.12)
U8	0.26 (0.07)	0.19 (0.06)	0.06 (0.02)	182.9 (71.7)	1.0 E−03 (2.2 E−04)	0.69 (0.15)	5.90 (6.85)
FV:	8.5	17	6.3	38	3.3	2.7	3.4
ID	Regression slope PP-SS	Agricultural land (%)	Ley (%)	Cereals & rape (%)	Other crops (%)	Clay average (%)	Silt average (%)
C6	8.3 E−04	61	22	76	2	41.8	42.0
E21	1.5 E−03	90	10	78	11	16.1	32.5
E23	6.3 E−04	62	38	55	7	39.8	40.2
F26	1.6 E−03	73	72	26	2	4.9	29.9
I28	2.2 E−03	86	23	64	13	18.6	33.4
M36	8.8 E−04	87	34	54	11	25.6	28.5
M42	1.5 E−03	95	7	76	17	15.8	32.0
N33	1.2 E−03	88	24	67	9	20.9	36.8
N34	1.9 E−03	86	16	61	23	13.9	28.5
O18	6.8 E−04	93	5	88	7	35.2	51.0
U8	7.4 E−04	58	24	75	1	50.0	40.8
FV:	3.4	1.6	14	3.4	30	10	1.8

"Other crops". Crop types are reported as a percentage of arable land (Table 2). Percentage of agricultural land in the catchment is given as the sum of arable and pasture land. The majority of the catchments have a small percentage of pasture, i.e., <3%, except for E23 with 8% pasture.

Catchment arable soil characteristics include clay and silt content. Average, median and maximum values of percentages of both clay and silt in the catchment were calculated using the GIS software ArcMap (ESRI, 2015) and the tool *Zonal Statistics as Table* based on catchment digital soil maps (Söderström & Piikki, 2016).

The P content in arable soil was estimated based on P extracted by ammonium lactate/acetic acid (P-AL) at pH 3.75, the standard method to measure plant available soil P content in Sweden (Egnér et al., 1960). Average soil P-AL values (mg/100 g; Table 2) are based on top soil samples collected at one sampling occasion, but at different years for each catchment, with sample points spread out over the catchment. In a majority of catchments, the number of sampling points range from 4 to 14 samples km^{-2}, except for E23, where 63 samples km^{-2} were taken. Data are collected from several different projects, hence the spread in number of sampling points and differences in sampling years. P-AL content is assumed to be stable over time, and the difference in sampling years is not assumed to be important (Djodjic & Spännar, 2012; Mattsson, 2002).

2.3. Data analysis – Multivariate analysis

Data were evaluated in several steps. First, individual correlations between in-stream variables were explored using linear regressions of monthly flow weighted concentrations for each catchment. Regressions were performed in JMP® 13.0.0 (2019). Second, regression parameters for the relationships between PP and SS were compiled together with in-stream and in-catchment variables to be used for Principal Components Analysis (PCA) (Table 2), performed using Canoco 5. The PCA was used as a base for further analysis, to find correlations between catchment properties and in-stream properties, and their connection to P, especially PP. All variables were put in the PCA at first. However after examination of the results, several variables describing the same property were removed, and the variables having the highest impact, i.e., the highest loadings, on the principal components were kept. Variables having low/no impact, i.e., loadings close to zero, were also removed, e.g., average flow and a number of regression parameters. Multiple linear regression was performed to explore how different combinations of variables could explain the variance in PP, as well as the variance in the slope of the regression between PP and SS. This analysis was made using monthly flow weighted concentrations, as well as using total average values for the whole monitoring period in each catchment (Table 2). A factor of variation (FV) was calculated as the ratio between the highest and the lowest obtained value for each parameter (Table 2), to illustrate the variation in catchment factors and water quality parameters between catchments.

Finally, to investigate whether flow-weighted concentrations of PP, SS, DP and TP and the PP/SS ratio changed over time, a two-sided Mann-Kendall test was used. Trend analyses were also performed on monthly discharge, as well as on monthly loads.

3. Results

3.1. Variation between catchments – Concentrations, catchment characteristics and land use

There is a high variability in the catchments and their in-stream and soil characteristics (Fig. 2, Table 2). Catchments with a high average clay content generally have a lower P-AL content, while catchments with coarser textured soils (mainly sandy loam) have a higher P-AL content. However, F26 with the coarsest soil texture deviates from this pattern as it has the lowest P-AL content of all catchments (Fig. 2, Table 2). For many catchments the particulate P fraction is the main part of the TP (seven catchments having PP > 50%), especially for catchments with a high clay content (O18, U8, E23 and C6) (Fig. 2, Table 2). Concentrations of SS and PP also showed high variation between catchments (FV = 38 and 17, respectively, Table 2). The variation of DP was considerably lower, with a FV of 6.3. Clay content (FV = 10) showed much higher variation than both P-AL (FV = 3.4), silt content (FV = 1.8) and fraction of arable land (FV = 1.6).

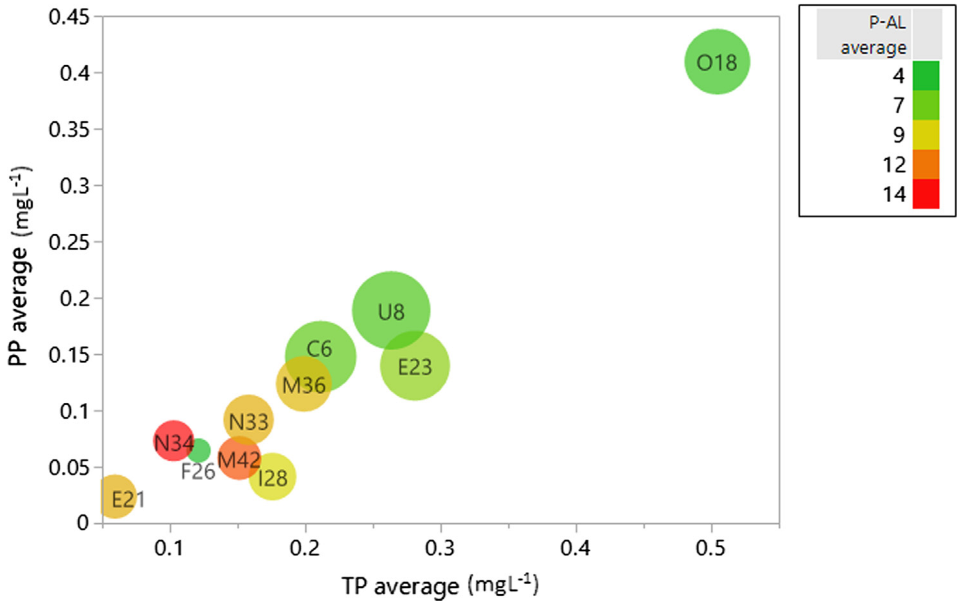

Fig. 2. Overall average values of flow-weighted concentrations of particulate phosphorus (PP) *vs.* total phosphorus (TP) for the study catchments. Symbol size is proportional to average clay content in the catchment, the bigger size, the more clay. The colors describe average phosphorus acetate lactate (P-AL, mg 100 g^{-1} of soil) content in the soil.

The average flow weighted SS concentrations ranged between 13 and 477 mg L^{-1}, and the average flow weighted TP concentrations ranged between 0.06 and 0.5 mg L^{-1}. The average flow weighted PP concentrations had approximately the same range as TP, i.e., between 0.02 and 0.4 mg L^{-1}, while DP concentrations were lower and ranged between 0.02 and 0.1 mg L^{-1} (Table 2). The sandy loam catchment E21 had the lowest SS, TP and PP concentrations while the clay catchment O18 represented the other end of the spectrum, with the highest SS, TP and PP concentrations. The R^2-value for the linear relationship between PP and SS for monthly flow-weighted concentrations for individual catchments varied between 0.26 and 0.83, with the highest value obtained for the clay catchment U8 and the smallest value for the clay/sandy loam catchment M36. In general, a higher R^2-value was obtained for the catchments with medium-high clay content. The slope of the linear relationship between PP and SS varied between 6E-04 and 2E-03, with the steepest slope for the small, sandy loam catchment I28, and the lowest slope for the clay catchment E23 (Table 2). The slopes for the catchments with a coarser soil texture are in general steeper, while the slopes for the catchments with a finer soil texture are lower.

3.2. Observed correlations between studied variables

In the PCA, PP positively correlated with SS and average clay content (Fig. 3). These three variables also have a positive correlation with average silt content, since they all line up next to each other (Fig. 3). No significant correlation between PP and P-AL was found. Different crop types do not seem to influence PP, whereas there seem to be a weak negative correlation between DP and Ley, as well as between PP/TP and P-AL (Fig. 3). The different catchments tends to group in the biplot according to their distinct characteristics. For instance, catchments with a higher clay content are grouped together (U8, C6, O18 and E23). Catchment F26, with a coarse soil texture and a large number of animals (Table 1) is instead highly correlated to Ley. Average clay content in the soil has a negative correlation with the slope of the linear relationship between PP and SS (Fig. 3).

The monthly flow weighted SS concentrations were a strong explanatory variable for the variance in monthly flow weighted PP concentrations (R^2 = 0.67, N = 1441, p-value < 0.01). Average clay content did not add any further statistical significance to this relationship in a multiple regression (R^2 = 0.67, N = 1441, p-value < 0.01). The same holds for a linear regression using total average values for PP and SS (R^2 = 0.98, N = 11, p-value < 0.001), where adding clay content gave the same results with no added significance to the regression. However, clay content did explain 62% of the variance in the slope of the relationship between PP and SS in a simple linear regression (R^2 = 0.62, N = 11, p-value < 0.05), where silt content explained 44% of the variance in the slope (R^2 = 0.44, N = 11, p-value < 0.05).

The relationships between PP, SS and average clay content identified in the PCA (Fig. 3) and the multiple linear regression, were further investigated together with average silt content (Fig. 4 and appendix Fig. A1). Average clay content is correlated with SS concentrations (R^2 = 0.36, N = 11, p-value < 0.05), with the clay catchment O18 deviating from the linear pattern (appendix Fig. A1). Average silt content of catchment soils also displays a linear relationship with SS (R^2 = 0.77, N = 11, p-value < 0.05), with O18 acting as an extreme value, but in this case, still lining up with the linear pattern (appendix Fig. A1). When fraction of agricultural land is taken into account together with the average clay content (Fig. 4), a linear relationship is found (R^2 = 0.75, N = 11, p-value < 0.01), where the product of average clay content and fraction of agricultural land is an explanatory variable for the logarithm of SS. Using the product of average silt content and fraction of agricultural land as a joint, explanatory variable for either SS or log(SS) does not result in a significant relationship (data not shown).

Multiple linear regression with SS as response variable and average clay content, average silt content and fraction of agricultural land as explanatory variables results in a significant linear relationship (R^2 = 0.81), with average silt content being the main explanatory variable (p-value < 0.05). A regression with just average silt content as the explanatory variable gives an R^2-value of 0.77, just slightly less then when adding both clay and fraction of agricultural land. Having clay content and fraction of agricultural

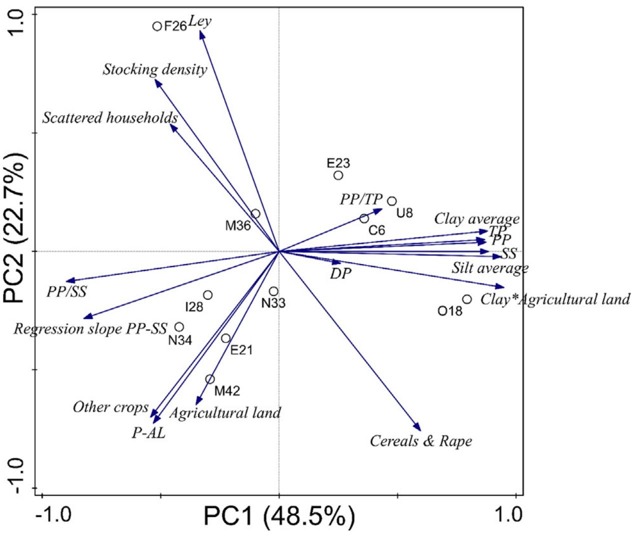

Fig. 3. Biplot from principal components analysis (PCA). Arrows represent variables, while circles represent catchments. All catchments are labelled by their abbreviation and all arrows are labelled by the variable they represent. PP = particulate phosphorus, TP = total phosphorus, SS = suspended solids and P-AL = plant available soil phosphorus content. The percentage on each axis describes the amount of variance explained by each principal component.

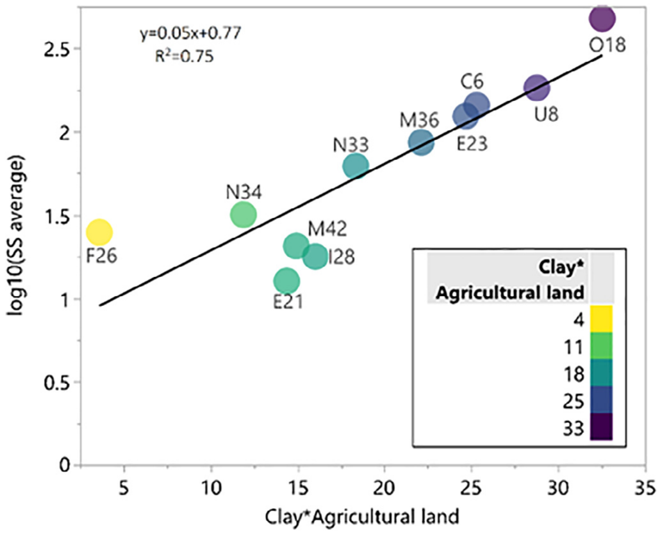

Fig. 4. The average in-stream suspended solids concentration (SS; logged values on the y-axis), *vs.* the product of average clay content and fraction of agricultural land in the catchment (x-axis; Clay*Agricultural land). The solid line represents a linear regression, with the R^2-value and regression equation shown in the figure. Symbol color represents the product of average clay content and fraction of agricultural land in the catchment.

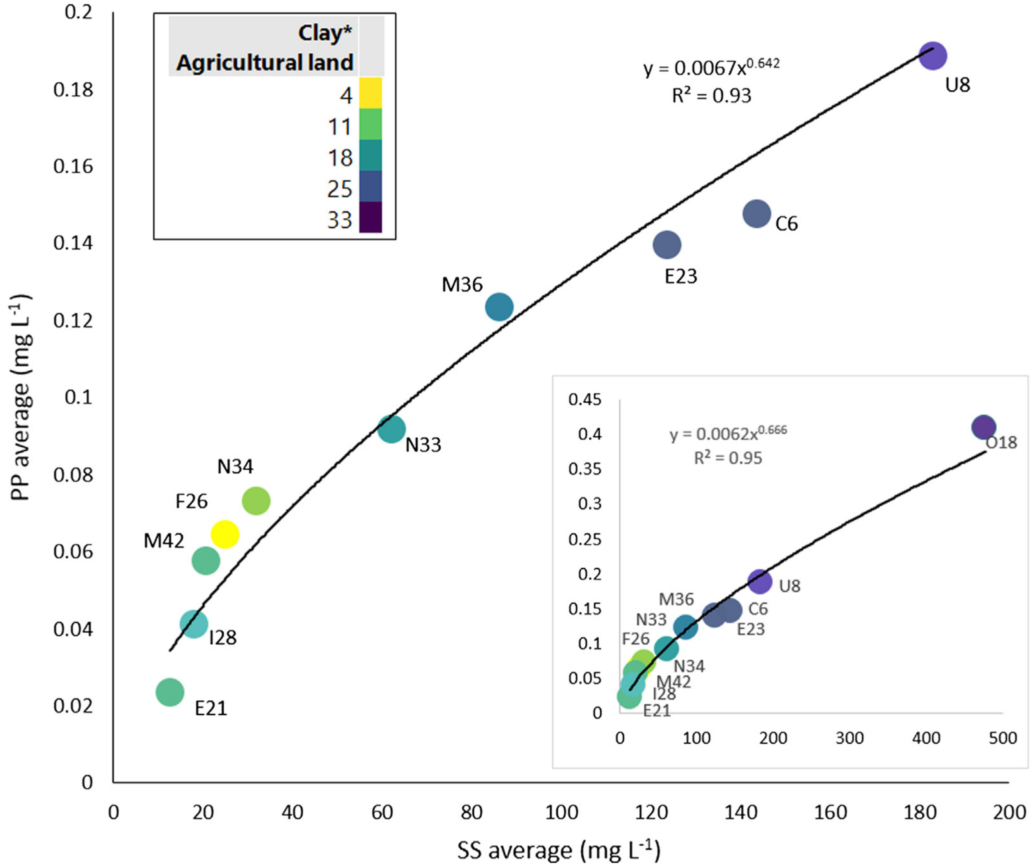

Fig. 5. Overall average flow weighted particulate phosphorus (PP) concentrations (y-axis) against overall average flow weighted suspended solids (SS) concentrations (x-axis). Symbol color represents the product of average clay content and fraction of agricultural land in the catchment. Catchment O18 is not included in the main figure. The inset figure shows a zoomed-out version of the main figure with the one extreme catchment (O18) included. The solid black lines in the main and inset figures represent fitted power law functions with the obtained equation and R^2-value shown.

land as separate explanatory variables for either SS or log(SS) in a regression, does not add the same significance as treating them as a joint variable, using the product of average clay content and fraction of agricultural land (Fig. 4).

Average clay and silt content in catchment soils correlates linearly with SS, while SS correlates with PP (Fig. 5). There is a strong, monotonic relationship between PP and SS, in both monthly flow weighted concentrations (R^2 = 0.67, N = 1441, p-value < 0.01), and total average values (Fig. 5). O18, the catchment with the highest values of TP, PP and SS, as well as a high clay content, acts as an extreme here as well (Fig. 5). There seem to be an exponential increase in the relationship between PP and SS in the lower concentrations ranges and for the catchments with a coarser soil texture, while the relationship increases more linearly for the higher concentration ranges and for catchments with a finer soil texture. The relationship can be described using a power law function (Fig. 5), where the exclusion of the more extreme catchment O18 results in only a slight decrease of the explained variance (R^2 = 0.93, Fig. 5).

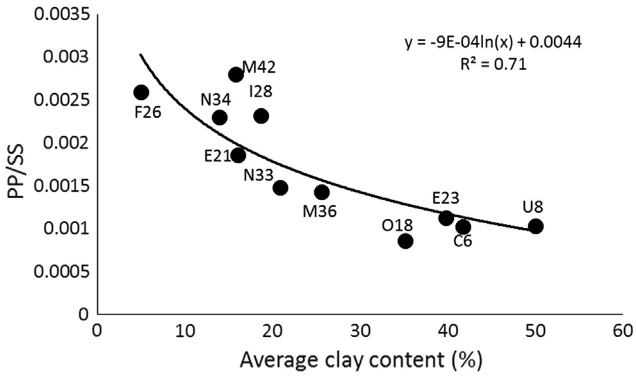

Fig. 6. The ratio between flow weighted average particulate phosphorus (PP) concentrations and flow weighted average suspended solids (SS) concentrations against average clay content. The circles describes each catchment, the solid black line describes a fitted logarithmic function. The equation and obtained R^2-value are shown.

Based on the left side of the biplot (Fig. 3), the relationship between the ratio PP/SS and average clay content was further investigated (Fig. 6). There is a decrease in the PP/SS ratio with an increase of average clay content, until around 30% average clay content, where it flattens out. The same pattern can be observed for PP/SS *vs.* average silt content, where the decrease of the ratio is observed at a silt content of approximately 42% (data not shown).

The relationships between DP, Ley, stocking density and scattered households observed in the PCA (Fig. 3) were further investigated with linear regression, but did not result in any significant relationships. The same was true for the correlation between P-AL and PP/TP.

3.3. Trend analysis of water quality parameters and discharge

A significant increase in the PP/SS ratio can be seen in 8 out of 11 catchments (Table 3). Four of the catchments, all having a medium to high clay content (E23, M36, O18 and U8), show no significant change in either PP or SS concentrations, while the PP/SS ratio is increasing. The sandy loam catchment E21 with the

Table 3
Results from the Mann-Kendall trend test of the ratio between PP/SS, discharge and flow-weighted concentrations of SS, PP, DP and TP over the whole monitoring period. The first column represents each catchment and the used time period in parenthesis. Each column represents the rate of change in the investigated variables where relevant and change in $mgL^{-1}yr^{-1}$ for flow-weighted concentrations and $Ls^{-1}yr^{-1}$ for discharge. Significant change with $p < 0.05$ is represented by * and $p < 0.01$ is represented by **. Empty cells means no significant change.

ID	PP/SS	PP ($mgL^{-1}yr^{-1}$)	SS ($mgL^{-1}yr^{-1}$)	DP ($mgL^{-1}yr^{-1}$)	TP ($mgL^{-1}yr^{-1}$)	Discharge ($Ls^{-1}yr^{-1}$)
C6 (2004–2017)	2.3 E−06**	5.0 E−04**		1.5 E−04**	5.7 E−04**	
E21 (2004–2017)	5.3 E−06*	2.1 E−04**	7.1 E−02**			
E23 (2007–2017)	4.3 E−06*				−1.5 E−03**	
F26 (2005–2017)	2.3 E−05**		−0.14**	7.0 E−05*		
I28 (2005–2017)		3.1 E−04**	4.5 E−02*	1.1 E−03**	1.4 E−03*	
M36 (2004–2017)	3.4 E−06*			1.7 E−04**	2.5 E−04*	
M42 (2006–2017)				7.0 E−04**	5.9 E−04*	
N33 (2007–2014)						
N34 (2004–2017)	7.7 E−06**	2.1 E−04*				−6.70**
O18 (2004–2017)	2.4 E−06*					−4.30**
U8 (2007–2017)	7.9 E−06**					−2.44**

lowest SS, TP and PP concentrations has a significant increase in the ratio and the two concentrations as well, however, the rate of change differs. The clay catchment C6 and the more sandy loam catchment N34 both show an increase in the ratio and the PP concentrations, but no change in SS. The loamy sand catchment with the smallest area, F26, is the only catchment showing a decrease in any of the investigated concentrations, in this case SS (Table 3). Catchment I28 has an increase in both PP and SS concentrations, but no change in the ratio.

No significant changes in loads of PP, TP, SS and DP can be seen in the majority of the catchments, except for the clay catchment U8 where there is a significant decrease in the load of PP, SS and TP. In general, there is a decreasing trend in water discharge in all catchments, however, the decrease is statistically significant in only three of the catchments: N34, O18 and U8 (Table 3).

4. Discussion

4.1. Catchment characteristics, water quality and their variability between catchments

There is a high between-catchment variability, both in catchment properties, but also in the different water quality parameters (Table 2, Fig. 2). Average flow-weighted TP, PP and SS concentrations have a high variability among the catchments, confirming the conclusions of Bol et al. (2018). However, our findings suggest that catchments with similar soil texture and land use behave similarly. The large variation in P fluxes between catchments supports the necessity of targeted measures against P losses. Additionally, different types of measures and even the appropriate placement of measures in the catchment might vary depending on catchment properties.

All catchments are clearly grouped in the biplot (Fig. 3) according to their character. Catchment F26 is strongly driven by its high and increasing stocking density and a high production of ley, and it also has one of the coarsest soil textures. This catchment deviates from the general relationship between log(SS) and the product between average clay content and fraction of agricultural land with somewhat higher SS concentrations (Fig. 4, Table 2). The concentrations of Total Organic Carbon are also highest for this catchment (Linefur et al., 2017), which may imply that some of the SS is of organic origin. Catchments N34, N33, M42, I28 and E21 all have around 90% agricultural land, with high P-AL content in the soil and quite similar sandy soil texture (Table 2). Consequently, they all are placed in the bottom left quarter of the biplot (Fig. 3). Catchments C6, U8, E23 and O18 have the highest TP, PP and SS losses and also high clay content and they all plot on the right half of the biplot (Fig. 3).

Iron (Fe), aluminum (Al) and calcium (Ca) geochemistry may explain some of the differences in results between catchments. Specifically, mineral bearing PP phases are formed by the reaction of soluble reactive P (SRP) with Fe, Al-Bierooxyhydroxide, Ca carbonate and organic matter (OM) associated with the composition of the particular catchment soils and aquatic sediments (O'Connell et al., 2015; Cooper et al., 2015). The P retention capability of fluvial sediments has been associated with various correlations of P/Al/Fe ratios (Rydin and Welch, 1999; Norton et al., 2008). Particularly, it has been demonstrated that Fe–P ratios can be useful indicators of the P buffering capacity of aquatic sediments (Jensen and Thamdrup, 1993), with greater Fe–P ratios suggesting enhanced SRP adsorption capacity onto the surfaces of Fe containing clay and sediment particles. In contrast, Al-P ratios also indicate SRP adsorption capacity to Al-oxyhydroxides, however, these are more stable under fluctuating redox potentials than ferric-(hydr)oxides (Mortimer, 1941, 1971; Reitzel et al., 2013). The lack of data regarding more detailed P speciation for here studied catchments is a limiting factor for further studies. The logical, and currently undergoing, next step is therefore collecting samples of fluvial sediment in some of the catchments for analyses of various P fractions and P pools.

4.2. Correlations between variables – SS concentrations as a main correlate for PP concentrations

Average clay and silt content in the catchments have a strong influence on SS concentrations in the streams, where a higher clay and silt content result in higher transport of soil material. These catchments are also typically known for having high SS and TP losses (Linefur et al., 2017). The main part of TP in these catchments (mainly U8, O18 and C6) consists of PP (Fig. 2, Table 2), and as we have shown, SS acts as the main in-stream correlate for PP, while the product of clay content and fraction of agricultural land acts as the main correlate for SS. These three catchments are typical examples of how PP losses are influenced by soil characteristics. A better regression is found between SS and silt content (R^2 = 0.77) than SS and clay content (R^2 = 0.36) (Section 3.2), in contrast to what we expected and what other studies have found, since clay content is the soil type that is historically mostly connected to SS and P losses e.g., (Kyllmar et al., 2014). However, both the afore mentioned relationships are strongly influenced by one catchment, i.e., O18. Excluding this catchment from the analysis decrease the correlation between silt content and SS (R^2 = 0.66) but considerably increase the correlation between clay content and SS (R^2 = 0.91). Clay soils are also typically more prone to erosion, and will therefore lose more P rich material (Ulén and Jakobsson, 2005), supporting a stronger relationship between SS and clay content instead of silt content. At the same time, the factor of variation is much lower for silt (FV = 1.8) compared to the corresponding value for clay content (FV = 10). Consequently, the slope of the relationship between silt content and SS concentrations is much higher, leading to rapid increases in predicted SS concentrations even with small increases in silt content, which might lead to large errors. Therefore, if predictions of expected SS concentrations are to be made for ungauged small catchments, choosing clay content, with possible consideration to fraction of agricultural land (Fig. 5), would be a more forgiving method. Additionally, in the regression with clay, O18 deviates the relationship from the linear pattern, while for silt, it drives the relationship towards linearity. This is a plausible explanation to why silt stands out as a more significant explanatory variable for SS in the multiple regression using both clay and silt. O18 has extreme SS and P losses, with the highest TP losses of all studied catchments (Linefur et al., 2017) and the highest average flow weighted concentrations (Table 2). However, it still represents an example of how losses in similar catchments can look, and mainly follows the same patterns as the other clay catchments, except with more extreme values.

Kyllmar et al. (2014) found a linear relationship between soil clay content and TP at the stream outlet. We argue here that transport of SS is an important step between soil clay content and, in this case, PP, instead of a direct relationship between PP and clay content since P found at the stream outlet is mainly in particulate form. Modeling studies using erosion and SS as a proxy for P losses from agricultural land have shown good agreement both on field and catchment scale (Djodjic and Markensten, 2018), in line with our results. The correlations between SS and PP that we can establish in this study also agrees with findings for larger catchments (Persson, 2001), that also stresses the importance of SS for P transport, as well as the importance of land use, i.e., fraction of arable land for SS losses. Persson (2001) uses percentage of arable land, and in this study the product of clay content and fraction of agricultural land is used, showing that these factors have a big influence on P losses in agricultural headwater catchments.

No significant relationships between DP concentrations and other variables in the dataset were found (Fig. 3). This is probably due to the fact that PP comprises the majority of the TP in these catchments (Table 2) and thus, DP does not influence the variation in the dataset as much as PP, which is often the case in small, agricultural catchments (e.g., Withers and Jarvie, 2008). Additionally, the variation in DP concentrations (FV = 6.3) between the studied catchments is much lower compared to PP (FV = 17). Previous studies have found other processes controlling the transport of DP compared to PP, with DP mainly being transported via subsurface flow (Rodríguez-Blanco et al., 2013; Dupas et al., 2015). Based on results of a lysimeter leaching study, Andersson et al. (2015) concludes that the subsoil can act as a source or a sink for P leaching depending on P content, degree of P saturation and P sorption capacity. Unfortunately there is very scarce data on subsoil properties in different catchments studied here.

The negative correlation found between average clay content and the regression slope for the PP-SS relationship indicates that less P is transported per SS particle in catchments with a higher clay content and consequently higher SS concentrations. Soil erosion is a selective process with respect to soil particle size (Sharpley, 1980) resulting in larger mobilisation and transport of smaller clay particles. Clay particles have, due to their higher specific surface area, higher P sorption potential compared to coarser soil particles (silt and sand). This results in enrichment ratios, i.e., a higher P content of eroded sediment compared to the P content of bulk soil. However, as Kleinman et al. (2011) show, "sediment P enrichment ratios tend to decline with increasing rates of erosion (and greater removal of larger soil particles and aggregates)". This statement agrees with our results (Fig. 6), where the PP/SS ratio decreases with an increasing clay content, suggesting that catchments with soil textures more prone to erosion have lower PP/SS (Villa Solís, 2014). In catchments with a high clay content, more SS is transported, but less PP is transported per unit SS. Villa Solís (2014) showed a strong decrease of P enrichment ratio (calculated as the content P in SS to that in soil), with increasing concentrations of SS. There seem to be a higher enrichment of P on the particles in catchments with a coarser soil texture as they usually have lower SS concentrations. However, we see an increase in the ratio PP/SS over time (Table 3) in several catchments, especially those with a high clay content, which that more PP per unit SS is being transported in these clay catchments over time.

4.3. The lack of correlation between soil P content and stream P concentrations

In this study we could not find evidence of a significant correlation between P in the stream and the plant available P (P-AL) content in the soil (Fig. 3). This may be a result of using an average value of P-AL in the entire catchment, and for some catchments the variation in P-AL is quite big (Table 2). Since P losses are episodic, and usually come from only a small area of the catchment, typically referred to as critical source areas (CSAs) (Pionke et al., 2000), it is possible that there is a correlation between P-AL in CSA soils and PP. Furthermore, we have shown (Fig. 5) that the impact of SS losses on PP losses and thereby TP losses is strong and therefore overrides the possible influence of other factors, including soil P content. Catchments with finer soil texture in our data set (Fig. 2, catchments O18, U8, C6, E23) prone to higher erosion losses have low P-AL content (Table 2) whereas the lowest TP and PP losses were observed from catchments with high P-AL content (Fig. 2, catchments E21, N34, M42). Withers et al. (2009) showed that the P enrichment ratio of SS declined exponentially as soil P increased. They found relatively small increases (10%) in the PP content of SS when soil P content increased from average to high fertility status. In our data set, between-catchment variation in average P-AL content (FV = 3.4, Table 2) is much lower than the variation in TP and PP concentrations (FV = 8.5 and 17, respectively, Table 2). Phosphorus load at the catchment outlet is mainly a remobilized secondary P source rather than a result of direct P delivery from the catchment sources (Jarvie et al., 2013). For example, Haygarth et al. (2012) could not establish a relationship between soil-P content estimated using Olsen-P and P export in two headwater catchments, which is in line with our results. To conclude, when considering highly variable SS concentrations and rather low variation in soil P content, it is difficult to expect a clear detectable effect of the soil P content on P losses.

4.4. Increasing trends in PP/SS ratio

There is an increase in the PP/SS ratio in eight of the eleven catchments, where five of the catchments have medium to high clay content (Table 3). Four of these catchments show no significant changes in either PP or SS concentrations. The increase of the ratio may point towards a shift in the size of particles being transported from the fields to the streams, with a higher percentage of smaller clay particles being transported, leading to transport of a higher amount of P (Kleinman et al., 2011). There could also be a small change in concentrations of both PP and SS over time, which are not significant, but when looked at together in a ratio, the change becomes significant.

The small, loamy sand catchment F26 with the highest rate of change in the PP/SS ratio is the only catchment showing a decrease in SS concentrations (Table 3). In this catchment there is a clear enrichment of P on the SS particles, where more P is being transported with a lower amount of particles. There has been an increase in number of animals in F26 over the monitoring period, as well as an increase in applied nutrients, especially from manure (Linefur et al., 2018), leading to more P being transported to the stream. Fertilizer and manure application rates are generally quite low for all catchments and in line with current recommendations from Swedish Board of Agriculture (2015) and are in agreement with average values for the surrounding production area (Statistics Sweden, 2017b). Three catchments (C6, I28 and O18) have a slightly higher application rate of fertilizers than their surrounding area (Kyllmar et al., 2014), however, the application rates of both fertilizers and manure are more or less constant with minor changes for all catchments, except F26, during the study period. Therefore it is unlikely that catchments displaying increasing trends of PP/SS, PP, DP, TP or SS are affected by fertilizer and manure application. Crop distributions in the catchments did not change significantly over the studied time period. Three catchments have an increase in percentage of catch crops, while four have a decrease and the rest are more or less the same (Linefur et al., 2017). Catch crops are used in Sweden primarily to reduce N losses and may lead to increased P losses from biomass after freezing-thawing cycles (Liu et al., 2014). However, no obvious patterns could be found between the observed trends in several catchments (Table 3) and the changes in percentage of catch crops. Structural liming has been applied in six catchments at least once during the study period. Structural liming is used as a measure to reduce P losses through improvement of soil structure and the stability of soil aggregates, especially on clay soils. However, as mentioned earlier, no decreasing trends in SS were recorded for these clay catchments (Table 3).

The increasing ratio between PP/SS in general, and especially in the catchments where no increase in the individual parameters is visible, is an indication that it is not enough to only study losses of P or SS by themselves, but to also study the ratio to detect changes and evaluate abatement efforts. The material being transported in some catchments is richer in P than before and the behavior that can be seen in some catchments where either there is only a change in the ratio, or in one of the concentrations, strengthens this statement. Catchment N33 is the only catchment that has no significant change in any of the variables. It is also the catchment with the lowest number of data points and the shortest monitored time period, which could be a reason why no change is detected.

Despite these increases that we see in the ratio between PP/SS, and in some concentrations, no significant increases in the different loads are found. One catchment (U8), in spite of having an increase in the PP/SS ratio, shows a significant decrease in the loads of PP, SS and TP (Section 3.3), probably due to a significant decreasing trend in water discharge. This is also one of the catchments participating in a project focusing on measures to decrease nutrient losses as mentioned earlier. Generally, catchment discharges are decreasing (although with a statistically significant decrease in only three catchments, including the mentioned U8) (Table 3). One reason for the lack of decreasing trends in loads despite decreasing discharge may be the increase in PP/SS ratios and some concentrations. Our results suggests a change in water quality, with approximately the same amount of P being exported from the catchments into the streams with less volume of water, leading to increasing concentrations of finer, more P rich particles being lost from the soil to the stream.

With this paper we want to highlight the importance of long-term monitoring data from headwater catchments to look at more general trends and correlates for stream P concentrations. Monitoring over long time periods with continuous sampling is needed to detect small changes in concentrations, especially as we have shown that in the majority of the catchments, there are significant changes in either the ratio PP/SS, or in one of the concentrations. The only catchment showing no change at all has the shortest time period in the analysis, which also highlights the need for having longer time periods to be able to detect trends.

5. Conclusions

Studying long-term monitoring data from these eleven small, agricultural catchments has lead us to the following conclusions:

- Clay content and fraction of agricultural land in the catchment are the main correlates for in-stream suspended solids concentrations
- The concentration of suspended solids is the main correlate for particulate phosphorus concentrations in the stream, acting as the middle step between soil texture, land use and phosphorus concentrations
- If these results are to be used for predictions of suspended solids losses, we suggest using clay content and amount agricultural land in the catchment as these variables forms a more robust relationship
- There is an increasing trend in the PP/SS ratio in several catchments, pointing towards a change in water quality in the streams

Due to the high spatial variability and wide range in soil texture in the studied catchments together with the long, consistent measurements performed in the catchments, we believe that these results are applicable to other regions with similar soil types, climate, agricultural practices and hydrological conditions.

Declaration of Competing Interest

No conflicts of interest are relevant for this study.

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Appendix

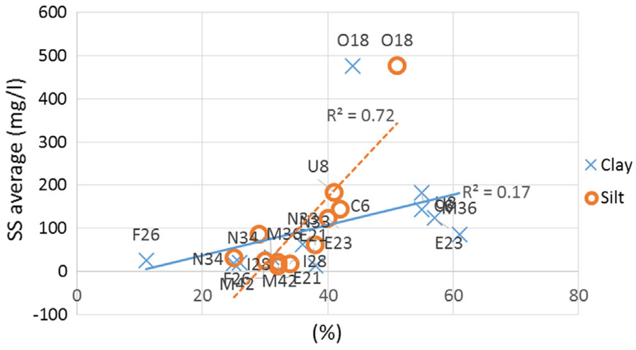

Fig. A1. The total average value of suspended solids (SS) against average clay content and average silt content for all catchments. The crosses describes clay content in each catchment, while the circles describes silt content in each catchment. Each catchment is labeled in the figure. The solid line is the regression line for the SS vs. clay relationship, and the dotted line is the regression line for the SS vs. silt relationship. Both regression's R^2-values are visible in the figure.

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