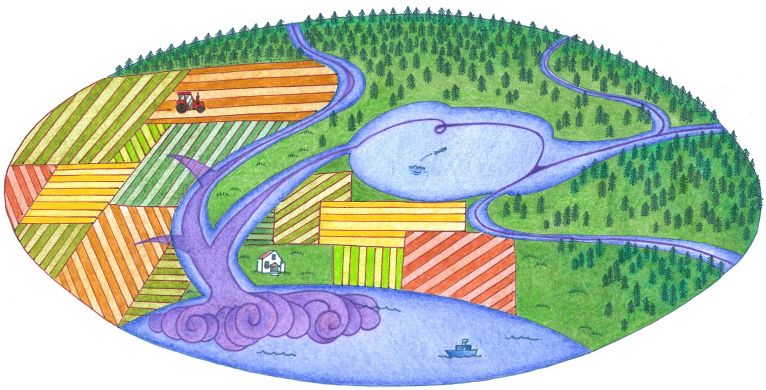
Acta Universitatis Agriculturae Sueciae

Doctoral Thesis No. 2021:84
Faculty of Natural Resources and Agricultural Sciences

Phosphorus transport in the landscape

– Integrating high-frequency monitoring, phosphorus geochemistry and modelling to improve water management

Emma E. Lannergård



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– Integrating high-frequency monitoring, phosphorus geochemistry and modelling to improve water management

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DOCTORAL THESIS
Uppsala 2021

Acta Universitatis Agriculturae Sueciae
2021:84

Cover: Phosphorus transfer in Sävjåan catchment
(illustration: B. Bailet)

ISSN 1652-6880
ISBN (print version) 978-91-7760-845-5
ISBN (electronic version) 978-91-7760-846-2

Print: SLU Service/Repro, Uppsala 2021

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Abstract

Eutrophication is one of today's most challenging water quality issues, despite considerable efforts to reduce phosphorus (P) input to surface waters. We need to explore new tools and techniques to improve the nutrient status of our waters and facilitate the management of catchment-scale P transport. This thesis includes research on P stored in lake and streambed sediment, timing and delivery of P in the system, processes influencing this transport and predictions by water quality modelling. The results show that significant amounts of P are stored in the catchment sediments, with some streams showing comparable concentrations to lakes. Phosphorus fractions are influenced by land cover and stream order. Some fractions could be important P sources during low flows when there is a significant risk of eutrophication associated with small increases in concentration. High-frequency (HF) monitoring is an important tool to increase understanding of catchment-scale P dynamics. Especially during intermediate and high flow events, the finer temporal resolution of HF data is essential for load calculations. Also, these events showed a dominant clockwise C-Q hysteresis response that suggests fast mobilisation of particles from the streambed and riparian areas. HF data was valuable in water quality modelling to describe temporal patterns but was challenging to calibrate and evaluate with standard performance statistics. Further work is needed on efficient and transferrable methods to analyse HF data. With an improved knowledge of P stores and the use of HF data, better-informed management decisions can be made to ensure water management that reduces catchment-scale P transport.

Keywords: Phosphorus transport, water management, environmental assessment, phosphorus legacy, high-frequency monitoring, proxy relations, C-Q analysis, water quality modelling, INCA-PEco

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Transport av fosfor i landskapet

Sammanfattning

Övergödning är ett av dagens stora vattenkvalitetsproblem, trots avsevärda ansträngningar att minska tillförseln av fosfor till våra ytvatten. För att förbättra näringsstatusen och förhindra vidare försämring behöver vi utforska nya metoder och verktyg. Denna avhandling är byggd på fyra studier i ett avrinningsområde med varierande markanvändning (skog och jordbruk). Vi studerade lagrad fosfor i sjö och vattendragssediment, där betydande mängder kunde konstateras. Denna fosfor är till viss del lagrad i sådan form att den kan tillgängliggöras, vilket innebär en risk för övergödning - särskilt vid låga flöden i vattendraget. Vidare användes högfrekvent data inhämtad med en in-situ sensor för att förstå kortsiktig variation och fosfortransporter bättre. Under medelhöga till höga flöden, var den högfrekventa datan särskilt värdefull då den hjälpte till att bättre kvantifiera transporten, men även skapa förståelse för hur fosforn mobiliseras i avrinningsområdet. Vid flödesökning var responsen i vattendragets grumlighet snabb, vilket indikerar att partiklarna kom från vattendraget eller den bäcknära zonen. Högfrekvent data var värdefullt i vattenkvalitetsmodellering, för att bättre beskriva förändringar i fosforns dynamik. Likväl innebar den stora variationen i datan en utmaning vid kalibrering och utvärdering av modellen med klassiska bedömningsmått. Vidare studier behövs för att ytterligare effektivisera och skapa transparenta metoder för hanteringen av högfrekvent data. Med hjälp av en ökad kunskap kring fosforns källa och användningen av högfrekvent data kan välinformerade beslut tas för en vattenförvaltning som minskar fosfortransport på avrinningsområdesnivå.

Nyckelord: fosfortransport, vattenförvaltning, lagrad fosfor, sediment, in-situ sensor, högfrekvent övervakning, vattenkvalitetsmodellering

Dedication

Till Cleo och Aron,

Jag hoppas ni hittar er plats i livet där kroppen klickar till som en mobiltelefon som sätts på laddning. Jag har fått möjligheten att vara på rätt plats i livet i fem år, då jag gjort det jobb som ligger till grund för den här boken. När nyfikenheten tillåts leva fritt, då blir även det jobbiga lätt.

”Den mätta dagen, den är aldrig störst. Den bästa dagen är en dag av törst.”

-I rörelse (*Härdarna*), Karin Boye

Contents

List of publications

This thesis is based on the work contained in the following papers, referred to by Roman numerals in the text:

I. **Lannergård, E.E.**, Agstam-Norlin, O., Huser, B.J., Sandström, S., Rakovic, J., Futter, M.N. (2020). New insights into legacy phosphorus from fractionation of streambed sediment. *Journal of Geophysical Research: Biogeosciences*, 125, e2020JG005763

II. **Lannergård, E.E.**, Ledesma, J.L.J., Fölster, J., Futter M.N., (2019). An evaluation of high frequency turbidity as a proxy for riverine total phosphorus concentrations. *Science of the Total Environment*, 651 (1), pp. 103-113.

III. **Lannergård, E.E.**, Fölster, J., Futter M.N. (2021). Turbidity-discharge hysteresis in a meso-scale catchment: the importance of intermediate scale events. *Hydrological processes* (accepted)

IV. **Lannergård, E.E.**, Crossman, J., Lewan, E., Widén Nilsson, E., Futter, M.N. High expectations: using high-frequency monitoring data for calibrating catchment scale phosphorus transport models. (manuscript)

Papers I-II are reproduced with the permission of the publishers.

The contribution of Emma E. Lannergård to the papers included in this thesis was as follows:

I. EEL designed and planned the study together with co-authors, had the main responsibility for field and lab work, analysing the data and writing the manuscript with support from co-authors.

II. EEL had responsibility for data collection from 2016 onwards, had the main responsibility for analysing the data and writing the manuscript with support from co-authors.

III. EEL had the main responsibility for analysing the data and writing the manuscript with support from co-authors.

IV. EEL planned the study together with co-authors, had the main responsibility for data collection, shared responsibility with MF regarding model calibration and writing the manuscript.

Additional papers

In addition to the papers included in the thesis, the author has contributed to the following peer-reviewed publications:

O. Agstam-Norlin, **E.E. Lannergård**, M.N. Futter, B.J. Huser (2020). Optimization of aluminium treatment efficiency to control internal phosphorus loading in eutrophic lakes, *Water Research*, 185, 116150.

S. Sandström, M.N. Futter, D.W. O'Connell, **E.E. Lannergård**, J. Rakovic, K. Kyllmar, L. Gill, F. Djodjic (2021). Variability in fluvial suspended and streambed sediment phosphorus fractions among small agricultural streams, *Journal of Environmental Quality*, 50(3), pp. 612-626

J.H. Crossman, G. Bussi, P.G. Whitehead, D. Butterfield, **E.E. Lannergård**, M.N. Futter (2021). A new, catchment-scale integrated water quality model of phosphorus, dissolved oxygen, biochemical oxygen demand and phytoplankton: INCA-Phosphorus Ecology (PEco), *Water*, 13 (723).

O. Agstam-Norlin, **E.E. Lannergård**, E. Rydin, M.N. Futter, B.J. Huser (2021). A 25-year retrospective analysis of factors influencing success of aluminium treatment for lake restoration, *Water Research*, 200, 117267

Abbreviations

ACA	Anti-clockwise – Clockwise
Al	Aluminum
Ca	Calcium
CAC	Clockwise – Anti-clockwise
C-Q	Concentration – discharge
EPC_0	Equilibrium phosphorus concentration
Fe	Iron
HF	**High-frequency**
LF	**Low-frequency**
MC	Monte Carlo
N	Nitrogen
NSE	Nash-Sutcliffe Efficiency
Org	Organic
P	**Phosphorus**
PCA	Principal component analysis
PO_4-P	Phosphate
PP	Particulate phosphorus
RDA	Redundancy analysis
RP	**Reactive phosphorus**

SD	Standard deviation
SMD	Soil moisture deficit
TP	**Total phosphorus**
TSS	**Total suspended solids**
Q	Discharge
VR	Variance ratio
WFD	Water Framework Directive

1. Introduction

We envision our common waters as healthy diverse ecosystems, a lake or stream inviting to humans and other animals to drink, bathe and use as habitat. That goal is not always easy to achieve due to present and historical pressures from a growing and increasingly urbanised population demanding energy and food.

While phosphorus (P) is an essential element for biological processes, excessive amounts can turn it into a pollutant, causing water quality deterioration in freshwaters (Schindler, 1974). Nutrient enrichment, also called eutrophication, favours the growth of algae and aquatic weeds. Eutrophication can lead to low dissolved oxygen levels, and subsequent biodiversity loss as species respond to a more nutrient-rich state (Smith et al., 1999). Among the requirements for plant growth, inorganic P and nitrogen (N) are the two principal limiting nutrients (Smith et al., 1999). Phosphorus has been identified as a key limiting nutrient in lake ecosystems, but N availability (N) and N:P ratios are also important for biomass growth (Smith, 2003). Eutrophication can affect society by creating problems in drinking water treatment, reducing recreational values (Smith et al., 1999), and toxic algal blooms that can harm the ecosystem and its visitors (Anderson et al., 2002).

Significant advances in understanding and managing cultural eutrophication have been made during the last 50 years (Schindler, 2006); despite this, it is still considered one of today's most challenging water quality issues (Cassidy & Jordan, 2011; Smith & Schindler, 2009). In Sweden, eutrophication is highly relevant, not the least due to the extensive problems with anoxia and marine "dead zones" in the Baltic Sea (Conley et al., 2009; Swedish Water and Marine Authority, 2019). In the part of central

Sweden described in this thesis, 55 % of all surface waters are regarded as eutrophic (Water Authorities, 2017).

Managing waters to reach good ecological and chemical status is the backbone of the EU Water Framework Directive (WFD) (European Commission, 2021). In a world where the conditions are rapidly changing (e.g., climate change, growth of urban areas, intensified biomass production, the green shift of agriculture), we need to search for new efficient ways to manage our waters sustainably. Haygarth et al. (2005) proposed an integrated, interdisciplinary approach to better understand P sources, mobilisation and delivery, and potential impact in receiving waters.

The main focus of this thesis, and the research behind it, was to *provide support for improved management decisions regarding phosphorus (P) transfer in a mixed land use catchment in Sweden*. With an interdisciplinary approach following areas were studied, P stored in lake and streambed sediment (**Paper I**), timing and delivery of P in the system (**Paper II**), processes influencing this transport (**Paper III**). Furthermore, water quality modelling was used to explore integrated mobilisation processes and P transport (**Paper IV**) (Figure 1).

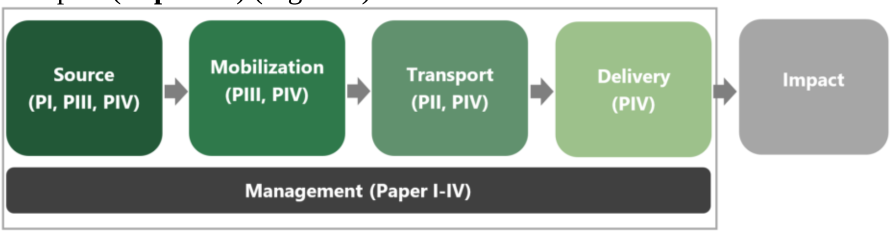

Figure 1. Inspired from the P-transfer continuum by Haygarth et al. (2005) in relation to Paper I-IV.

1.1 What are the P sources in the catchment?

To improve eutrophication management, we are interested in what sources contribute to P emissions during different conditions as well as the most influential sources (Bol et al., 2018). Often the magnitude of P stored in the landscape is unknown, and if the relative importance of P sources should be evaluated, the size of the stores must be quantified.

1.1.1 Point and diffuse sources

The origin of P decides if it is categorised as a point or nonpoint (diffuse) source. Point sources could be, e.g. sewage treatment plants, septic tanks or agricultural wastes (Schindler, 2006). Point sources pose a significant risk of contributing to eutrophication since small increases in highly bioavailable P forms (reactive P, RP[1]) can significantly increase biomass growth (Biggs, 2000) during ecological sensitive times, e.g. summer low flows (Jarvie et al., 2006). Strategies for controlling point source pollution can include legislative regulation, economic incentives and "end-of-pipe" technology applications (Armon & Starovetsky, 2014). Due to their successful control, point sources have received less attention in European countries during the latest decades (Kronvang et al., 2007; Schindler, 2006).

On the other hand, diffuse sources, e.g. nutrient runoff from agricultural soils, have been the subject of much attention and concern (Kronvang et al., 2007). Agricultural P could originate from chemical fertiliser, manure or release from the soil P pool (Haygarth & Jarvis, 1999). Phosphorus surplus on cropland is common around the world and more prominent in areas with high manure application (MacDonald et al., 2011). In Europe, agricultural production is one of the main pressures degrading surface water quality through the export of P and N (European Environment Agency, 2019). In Nordic conditions, the share of agricultural land is often positively related to the nutrient export from the catchment (De Wit et al., 2020; Tattari et al., 2017). Forested catchments can also contribute to diffuse pollution, but P export is modest compared to other land use practices (Tattari et al., 2017). Mitigation methods to limit diffuse source pollution is described in section *1.3.2 Land use mitigation measures*.

1.1.2 Legacy in the land-water continuum

While controlling point and diffuse sources of P input is crucial to counteract eutrophication, it is not always enough to ensure recovery (Reitzel et al., 2005). Historical use of P has enriched the catchment land-water continuum with P stores, often called legacy P (Kleinman et al., 2011). Legacy P has accumulated in field soils (Kleinman et al., 2011), ditches (Shore et al., 2016), riparian soils (Fox et al., 2016), wetlands (Geranmayeh et al., 2018),

[1] Laboratory measurements of unfiltered PO_4-P will hereafter be called total reactive P (RP) according to the RP (unf) definition in Haygarth and Sharpley (2000).

lake sediment (Agstam-Norlin et al., 2021) and streambed sediment (Withers & Jarvie, 2008). These P stores can, under certain conditions, contribute to internal loading, i.e. be released or suspended in the water column and spiral (go from dissolved – to particulate/organic – back to dissolved) down the system (Withers & Jarvie, 2008). Internal loading of P may mask reductions in external loading (Spears et al., 2012) and lead to questions about the effectiveness of mitigation measures (Sharpley et al., 2013). Even though streambed sediment is pointed out as an essential legacy P source (Sharpley et al., 2013), studies exploring the magnitude of the store in the landscape are few (e.g. Palmer-Felgate et al., 2009; Ballantine et al., 2009b).

Different forms of P in sediment

The bioavailability of P depends on its association with other substances. Phosphorus can be adsorbed or co-precipitated with inorganic compounds, e.g. redox-mediated co-precipitation with iron (Fe) and manganese (Mn); precipitated with aluminium (Al) or calcium (Ca); adsorbed to clays and hydroxides or associated with carbonates (Boström, 1988). Phosphorus can also be associated with or built into organic molecules of living and dead biota. Phosphorus species considered potentially mobile are redox-sensitive Fe compounds and more or less labile organic forms (Søndergaard et al., 2003; Huser et al., 2016b).

Different methods can be used to explore both total phosphorus (TP) content and different P fractions in the sediment. Sequential fractionation methods are widely used for quantification of P fractions in lake sediment (Pettersson et al., 1988; Psenner & Pucsko, 1988, Ruban et al., 1999; Rydin, 2000; Barik et al., 2016) but are rarely applied to streambed sediment (Audette et al., 2018, SanClements et al., 2009). The method is also used to explore P export potential in sediment (Kopaček et al., 2005; Rydin et al., 2000; Reitzel et al., 2005) and assess the effectiveness of lake restoration measures (Huser et al., 2016b; Agstam-Norlin et al., 2021).

1.2 Processes contributing to P transfer

To predict and manage future societal pressures, a solid understanding of the processes driving P (and N) export is crucial (De Wit et al., 2020). Therefore, mobilisation and delivery mechanisms need to be identified and understood (Bol et al., 2018).

In smaller catchments with extensive monitoring equipment, flow pathways and diffuse nutrient transfer pathways have been successfully characterised (e.g. Mellander et al., 2015). Also, studies describing parts of the different mobilisation pathways are common, e.g. erosion (Djodjic & Villa, 2015) or internal loading from streambed sediments (Smolders et al., 2017). Water quality modelling is another approach to get insight into flow pathways and potential key controls on nutrient export (Johnes & Heathwaite, 1997). Concentration-discharge (C-Q) relationships have also been used to better understand catchment processes. The difference in temporal patterns during storm events is analysed and related to environmental variables (Glover & Johnson, 1974; Bieroza & Heathwaite, 2015; Rose et al., 2018).

1.2.1 Hydrology

Water movement plays a crucial role in transporting P. Point sources, diffuse sources or legacy P all need a hydrological connection in the landscape to turn into a problem for surface waters. The transport of matter and energy (e.g. water, nutrients, and organisms) between different landscape components of the hydrologic cycle is called hydrologic connectivity (Freeman et al., 2007). The water pathway could be vertical (e.g. a moving water table), lateral (along the hillslope) or longitudinal (driven by terrain) (Bracken et al., 2013). The natural drainage network is extensively modified in managed catchments since surface and subsurface (tile) drains are common to effectively transport water from the fields downstream (Blann et al., 2009).

Hydrology is temporally variable and driven by precipitation (rainfall or snow), where intensity, duration and interval are essential factors influencing sediment and P transport (Haygarth & Jarvis, 1999). Events where precipitation causes a meaningful change in the hydrograph (often called storms) are of great importance for the total transport of sediment and phosphorus (Kronvang et al., 1997; Jordan et al., 2007).

1.2.2 Physical processes

Soil erosion and surface runoff are important pathways for P delivery to surface waters (Kronvang et al., 2007). In Sweden, soil erosion has historically not been considered a severe problem due to comparably low rain intensities, permeable soils, limited surface runoff and dense vegetation cover (Brandt, 1990). In Nordic conditions, soil erosion often occurs in autumn and during snowmelt when the soil is saturated, leading to high hydrological connectivity in the catchment (Ulén et al., 2012).

Erosion can occur laterally as sheet erosion on the soil surface, vertically transporting particles via macropores and tile drains to surface waters (Ulén et al., 2012) and longitudinally from bankside areas along streams (Djodjic & Villa, 2015). The soil erodibility depends on the internal forces holding the soil together in combination with external eroding pressures, where, e.g., silt loam and clay loam are especially vulnerable soil types (Ulén et al., 2012).

Not all parts of the catchment contribute equally to erosion. Critical source areas can cause the majority of the loss (80%), which originate from only a small proportion of the land (20%) (Sharpley et al., 2009). The explanation for these critical source areas could be, e.g. high hydrological connectivity (fast storm flow paths, surface or near-surface flow), geology with high nutrient loss potential, in combination with intense agricultural activity (Pionke et al., 2000). With successful identification and risk mapping, these areas could be targeted for mitigation measures (Djodjic & Villa, 2015).

Suspended sediments and particulate phosphorus

When dispersed particles are transported with water, they are called total suspended solids (TSS) (arbitrarily defined as inorganic and organic fine particulate matter >0.45 µm, Owens, 2007). On its own, TSS can be detrimental to aquatic biota (Bilotta & Brazier, 2008) but is also an important vector for P transport (Sandström et al., 2021). Phosphorus can be adsorbed to particles or mineral bound in primary or secondary minerals (Spivakov et al., 2007). Depending on what P is associated with, it can be more or less available to algae and instream organisms (Ballantine et al., 2009a), which is why it is necessary to study the different P fractions also in suspended sediments (Sandström et al., 2021). The smaller sized particles, i.e., colloids (clay minerals, Fe oxides and organic matter between 0.001-1 µm, Owens, 2007), can significantly contribute to the transport of P and other pollutants (Bilotta & Brazier, 2008, Gottselig et al., 2017).

1.2.3 Biogeochemical processes influencing sediment P

Sediment P release mechanisms are related to physical, biological and chemical processes. The transport in the land-water continuum is not passive, P is cycled and exchanged between different inorganic and organic pools (Sharpley et al., 2013). Phosphate occurs almost entirely as $H_2PO_4^{-}$ and HPO_4^{2-} (orthophosphates) within the pH range of natural waters (i.e. pH 5-9) (Orihel et al., 2017). Orthophosphates are reactive anions, which makes sorption processes especially important and makes a large proportion of P in the system prevail in a solid phase (Sharpley et al., 2013).

Sorption and desorption processes and precipitation with secondary minerals are important for P availability (Records et al., 2016). Depending on pH, P can be sorbed to Fe and Al(hydr)oxides as well as Ca carbonates (Reddy & DeLaune, 2008). First-order controls for sorption/desorption are (1) P content in sediment/soil compared to the surrounding water (and its flow velocity), (2) mineralogy which is connected to soil texture (higher sorption capacity of clay minerals) and (3) redox conditions (Records et al., 2016). Chemical precipitation is a key mechanism when P concentrations are higher than the soil adsorption capacity, and is dependent on specific minerals present and their solubility. Other controls include pH, organic matter and temperature (Records et al., 2016).

Redox conditions affect the Fe-bound P since anoxic conditions favour desorption of P (Smith et al., 2011; Boström et al., 1988). When oxygen is consumed by heterotrophic respiration, alternate electron acceptors will be used (NO_3^-, Mn^{4+}, Fe^{3+} and SO_4^{2-}) (Smith et al., 2011). Fe-oxide minerals are reductively dissolved by sediment microorganisms, which releases P from the sediments.

Turbulence and sediment mixing caused by bioturbation from, e.g. common carp could also increase the availability of mobile P (Huser et al., 2016a).

1.3 Management of P transfer

Kirchner et al. (2004) predicted a new era of monitoring where continuous in-situ measurements would aid in understanding temporal variation in streams and rivers. Since then, significant advances in technology and application have been achieved (Rode et al., 2016). However, “big” data pipelines and real-time processing is still a challenge (Rode et al., 2016), but essential to make the data useful to managers and authorities. In the Nordic countries, sensors are still not widely used in national monitoring programs

(Skarbøvik et al., 2021), and conditions where in-situ sensors are efficient still need exploration.

In practice, several different land management measures could be applied to hinder P export and reduce eutrophication. However, catchment specific properties matter for how efficient these measures are. Water quality models have been used to theoretically evaluate and compare the effectiveness of potential measures (Greene et al., 2011; Jin et al., 2013). One benefit of using models is that future scenarios can be explored, e.g. potential effects of climate change (Crossman et al., 2013).

1.3.1 Monitoring in running waters

By transporting nutrients, rivers link the land to coastal areas (van der Struijk & Kroeze, 2010), which is why they are crucial to monitor if we want to manage P transfer in a catchment. The transport of TSS and P is important to assess output loads, sources of contamination and evaluate the effectiveness of measures (Moatar & Meybeck, 2005).

When monitoring the status of running waters, systematic grab sampling (e.g. monthly) is an efficient way to discover changes over time (Fölster et al., 2014). Nevertheless, with monthly sampling much time is unmonitored, and for specific periods the frequency is too low to get a representative picture of the water chemistry fluctuations (Jones et al., 2012), especially regarding various forms of P and TSS that are highly variable over time (Coynel et al. 2004; Moatar & Meybeck, 2005). Unmonitored time increase uncertainty and leave knowledge gaps regarding drivers controlling particle transport, source origin (diffuse/point sources) and the proportion available P (Johnes, 2007).

One way to increase the available information about temporal variation in concentrations and fluxes is to deploy an in-situ sensor, monitoring water quality, e.g. every 15 minutes. However, in-situ sensors directly measuring RP, TP and TSS with wet chemistry techniques are still novel and face technical and management challenges (Chen & Crossman, 2021). Instead, turbidity has been used as a proxy for TSS or TP, when the correlation between parameters has been sufficiently good (Grayson et al., 1996; Jones et al., 2012; Ruzycki et al., 2014; Skarbøvik & Roseth, 2015; Koskiaho et al., 2015; Kämäri et al., 2020). The correlations between turbidity and TSS or TP are site-specific and non-transferable between catchments (Jones et al., 2012; Stutter et al., 2017). The relation can be affected by particle composition (Gippel, 1995) and particle size distribution (Stubblefield et al., 2007) which is in turn affected by season and storm events (Walling & Morehead, 1987; Bogen, 1992).

1.3.2 Land use mitigation measures

Field-scale measures

Strategies to reduce P load from agricultural practices are still a common approach to reducing P transfer in the landscape (Bergström et al., 2015). Firstly, the addition of P to cropland needs to be in balance with the removal from plants to avoid P build up in soils. Historically inorganic fertiliser P was frequently applied in large quantities to improve crop production leading to large P stores in the soil (Ulén et al., 2007; Linderholm et al., 2012). Today mineral fertiliser application has decreased and is no longer in surplus (Linderholm et al., 2012). Furthermore, appropriate manure management, amounts, distribution, timing and application technique are essential to reduce P loss (Bergström et al., 2015). Today, manure input to Swedish soils has decreased due to fewer grazing and non-grazing animals (Ulén et al., 2007). Regulations regarding livestock densities and manure application rates are also in place (Swedish Board of Agriculture, 2021c).

Soil structure is also important. In Sweden, structure liming on clay soil has been explored to avoid P leaching from fields. The addition and incorporation of quicklime or hydrated lime improves aggregate stability and has shown reduced particulate P (PP) and potentially also RP leaching (Ulén & Etana, 2014). Another field measure is to sow catch crops (grown between two main crops) to reduce the time of bare soil and decrease P export (Bergström et al., 2015). However, depending on the catch crop grown and freezing and thawing cycles, the crops might turn into P sources instead of sinks (Liu et al., 2014). Other studies report enhanced nutrient retention and less RP export during winter and spring when applying catch crops (Hanrahan et al., 2021). There are subsidies to use catch crops in some Swedish regions to reduce N leaching and P losses (Swedish Board of Agriculture, 2021a).

Tillage can affect P leaching due to increased vulnerability to soil erosion, e.g. by removing crop cover (Haygarth & Jarvis, 1999). Reduced autumn tillage is subsided in certain areas, mainly to reduce N leaching (Swedish Board of Agriculture, 2021a).

Edge of field measures

Riparian buffer zones consisting of grass, ley or other natural vegetation are commonly used and subsided as an agricultural mitigation measure to reduce surface runoff, erosion and P transport (Swedish Board of Agriculture, 2021b). The evidence base for understanding processes in the riparian buffer zones has been inconsistent due to strong site-specificity in the landscape (Stutter et al., 2021). However, regarding P transport, the riparian buffer zones have a positive effect on reduced surface runoff erosion, infiltration to increase pollutant contact with subsoils and slowing flows to increase residence time to aid biotic processes (Stutter et al., 2021). Two-stage ditches, constructed with vegetated floodplain/benches, is another method to slow down water and allow settling of PP (Hodaj et al., 2017) and plant assimilation and retention of RP (Trentman et al., 2020).

Constructed or restored wetlands is another important countermeasure to reduce P and sediment delivery to aquatic systems (O'Green et al., 2010; Kynkäänniemi et al., 2013). However, the wetlands need to be carefully located, designed and maintained to be efficient (Djojdic et al., 2020).

Channel management

Harvesting aquatic plants for phytoremediation and removal of nutrients has a long history (Reddy & Debusk, 1985). Plant nutrient allocation strategies (amongst species and during different parts of the year), e.g. storage in leaves compared to the rhizome, need to be considered to optimise nutrient removal strategies (Quilliuam et al., 2015). The method used for harvesting excessive aquatic plants will lead to varying levels of ecosystem disturbances. The harvesting can range from being done "by hand" to using large-scale cutting mechanical weed harvesters. There is also a risk of re-suspending sediment and sequestered P in combination with reduced uptake of nutrients by plants (Quilliuam et al., 2015).

Lake restoration

There are several methods to reduce P internal loading in lakes, among them treatment with Al salts that naturally bind the P (Huser et al., 2016b; Agstam-Norlin, 2021). Treatment longevity varies between lakes, where lake morphology, Al-dose and watershed to lake area ratio are important for efficiency (Huser et al., 2016b; Agstam-Norlin et al., 2021). Moreover, the Al addition method can determine the result, where Al injected into the sediment has shown greater binding efficiency than water column application (Agstam-Norlin, 2020). With the injection method, Al is distributed vertically in the sediment profile rather than precipitated in the water column (Agstam-Norlin, 2020).

1.3.3 Water quality modelling

Distributed, process-based models represent a powerful tool for understanding nonpoint pollution and the effects of land use change (Wellen et al., 2015). The goal of modelling nutrient pollution is often related to management and policy (Wellen et al., 2015), providing a predictive link between management actions and response in the studied system (Rode et al., 2010).

However, it is sometimes a challenge to get robust and reliable results due to input data requirements and weaknesses in the mathematical descriptions of landscape and biogeochemical processes (Rode et al., 2010). Even if a great fit is achieved between observed and simulated data, it is essential to get the right answer for the right reason (Kirchner, 2006). There is always a risk of equifinality, getting acceptable model calibrations based on inaccurate premises (Beven, 2006). Critical evaluations must be done to balance optimal model complexity against an acceptable level of uncertainty (Rode et al., 2010).

Different levels of confidence in the model output should be strived for depending on the purpose (e.g. regulatory, planning, exploratory) (Harmel et al., 2014). When the purpose is exploratory, with the goal of testing hypothesis against system function or exploring conceptual models, reduced confidence in predictions is acceptable (Harmel et al., 2014).

2. Objectives and research questions

Despite 50 years of research, the temporal and spatial variation in the P transfer-continuum is not fully understood. To make informed management decisions today and in the future, to effectively reduce freshwater and marine eutrophication, new tools and techniques need to be explored to measure and model P pools and fluxes.

Therefore, the primary objective of this thesis was to *provide support for improved management decisions regarding P transfer* on catchment scale. Four studies in the same lowland, mixed land use Swedish catchment were carried out to investigate legacy phosphorus in streams and lakes, study transport and processes with in-situ sensor high-frequency (HF) monitoring and perform catchment scale modelling (Figure 2). The following research questions guided this thesis work and addressed the overall objective:

I. How large are the legacy P stores in streams/lakes in the catchment, and how potentially bioavailable are they? (**Paper I**).

II. How can HF monitoring be used to better quantify P losses through the catchment outlet? (**Paper II**)

III. Does analysis of HF turbidity–discharge hysteresis patterns give meaningful information regarding processes leading to material transport in the catchment? (**Paper III**)

IV. How can HF data combined with the new insights addressed in I-III aid in improving the ability to model P transport in rivers (**Paper IV**)?

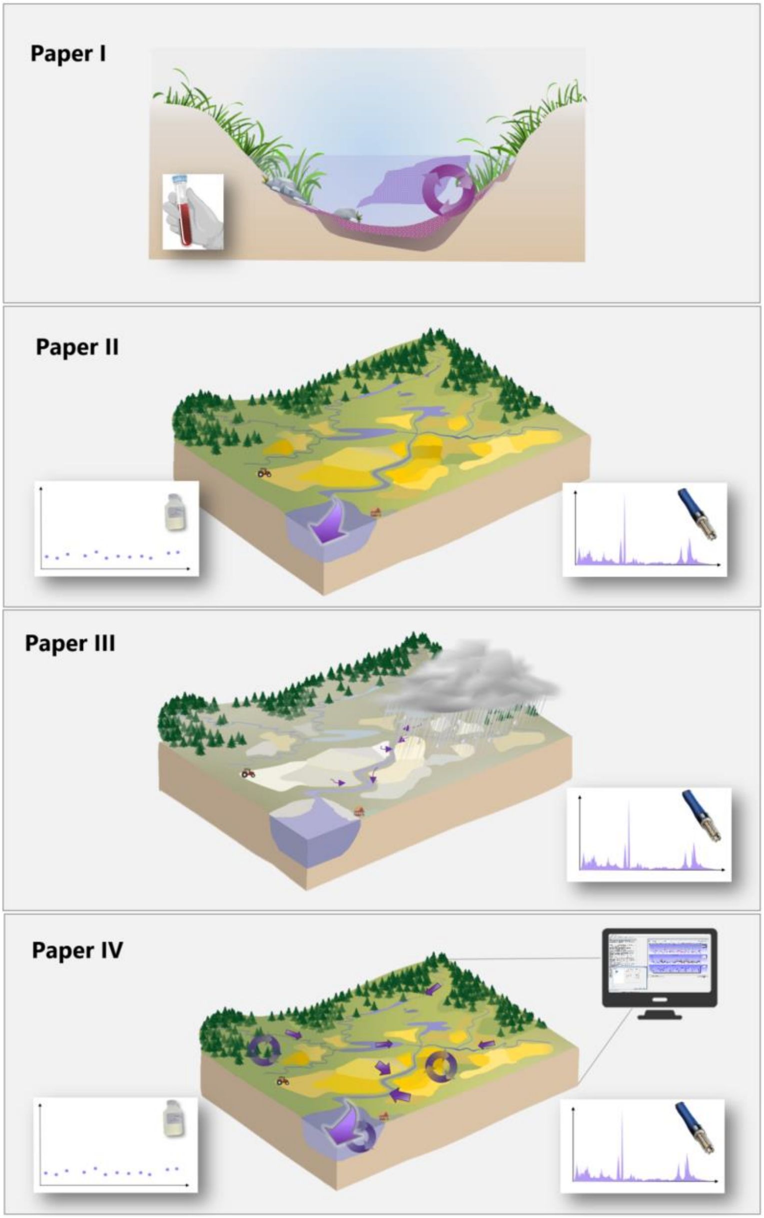

Figure 2. A conceptual representation of Paper I-IV, including scale, type of data used and most important processes in the study. P is indicated by purple colour, arrows indicate transformations and mobilisation/transport processes.

3. Methodology

The aim of this thesis was addressed by three different approaches (1) sampling of water and sediment (**Paper I and II**), (II) empirical data analysis (**Paper II and III**) and (III) process-based water quality modelling (**Paper IV**). All studies were conducted in the Sävjaån catchment (Figure 3).

3.1 Sävjaån catchment

The Sävjaån catchment is located in central-east Sweden, close to the city of Uppsala. The meso-scale catchment (722 km^2) is heterogeneous with forested headwaters in the north and east and typical low land streams in the central and south parts (Water Authorities, 2017). The area is generally flat, and the difference between the highest and lowest point is 70 m (The National Land Survey, 2020). The area was submerged with water after the last glaciation, resulting in today's catchment organisation. The dominant land cover type is forest (71%). These areas are mainly located on the slightly higher elevation outwashed till soils, while the lowland postglacial clay areas are primarily used for agriculture and pasture (24%). There are also a few centrally located lakes (3%) and a small urban area (2%) that is part of the city of Uppsala. The two largest lakes (Funbosjön and Trehörningen) are affected by eutrophication and classified as with moderate ecological status according to the WFD (Water Authorities, 2021).

The most common crops grown (in the agricultural areas) are winter wheat (24%) and spring barley (15%). Some agricultural land is also used non-intensively, e.g. growing ley and fallow (36%) (Hansson et al., 2019).

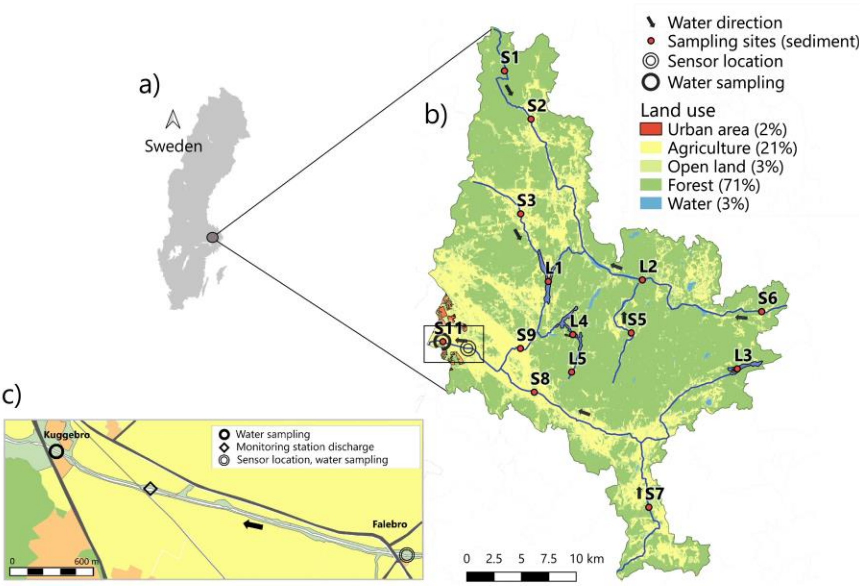

Figure 3. Sävjaån catchment, (A) location in Sweden, (B) land cover, sampling sites and in-situ sensor location, (C) location of in-situ sensor, discharge monitoring gauge and long term water monitoring site. Adapted from Paper I and II. ©The National Land Survey.

Agricultural areas in central Sweden located on clay soils are commonly tile-drained (Djodjic, 2001), affecting the water pathways in the catchment. Animal husbandry is relatively uncommon in the catchment, but grazing in riparian areas is allowed (Swedish Board of Agriculture, 2021b).

The climate is temperate continental with a mean annual temperature of 6°C, with average daily temperatures between -27°C to 26°C (1949-2017). The average annual precipitation is 639 mm, and the average annual runoff is 189 mm (1981-2010 Swedish Meteorological and Hydrological Institute SMHI, 2020). During winter, streams are often ice-covered for one or more months each year. Winter flow is sustained by groundwater and increasingly common winter rainfall and snowmelt events. Flow is generally flashier during spring and autumn, while summer flow is typically low. Until September 2020, discharge was monitored through stage height at a flow weir close to the outlet of the catchment (Station ID 2243) (SMHI, 2021).

Streams in the catchment vary between headwater (1.5-2 m wide, 0.5-0.9 m deep) and fourth-order streams (12 m wide, 2 m deep) (**Paper I**). Most streams have a WFD “moderate” status due to excessive nutrients and hydromorphological problems. In the summer, littoral vegetation in the streams is extensive. The streambed sediment varies from rich organic-based sediment (10-12 cm) to finer particulate sediment with stones (1-2 cm) (**Paper I**).

Close to the catchment outlet, an in-situ sensor is deployed (Sävjaån Falebro, Figure 3), monitoring water quality every 10-15 minutes (2012-2016 YSI 600OMS, 2016-2021 YSI EXO2). A long term monitoring station is located 2 km downstream of the in-situ sensor (Sävjaån Kuggebro S11, Figure 3), where monthly water quality has been observed since 1962. There is another long term monitoring station in the catchment, located in a headwater stream called Sävjaån Ingvastra (Figure 3, S2). Average TSS concentrations in the two sites in the catchment are similar (Table 1). The average TP and RP concentrations are slightly higher in Sävjaån Kuggebro (downstream), as is the proportion of average RP/TP. The water is also more turbid in Sävjaån Kuggebro (mean and max) than in Ingvastra.

Table 1. Summary of monthly water chemistry data from Sävjaån Ingvastra and Kuggebro 1962-2021, SD denotes standard deviation.

	Sävjaån Ingvastra (S2)			Sävjaån Kuggebro (S11)		
	Mean/SD	Min	Max	Mean/SD	Min	Max
TP (µg/l)	60.8/47.1	15	640	72.4/49.2	21	466
RP (µg/l)	23.1/27.8	2.0	408	33.6/28.0	2.0	339
TSS (mg/l)	20.6/22.8	3.4	290	19.5/22.0	1.4	242
Turbidity (FNU)	15.2/9.5	5.0	51	20.7/27.4	0.9	224

3.2 Sampling and laboratory analysis

3.2.1 Sediment sampling and analysis

Triplicate sediment cores were taken from five lakes and nine streams in the catchment in May 2017 (**Paper I**). A Willner gravity corer was used for the lake sediment. In the streams, a similar corer on a rod, or a tube, was manually pushed into the sediment and then taken up while two plugs created a vacuum. Stream sediment was sampled from areas without stones or rooted vegetation and taken from the midchannel to avoid collapsed side banks and represent the main channel (Shelton & Capel, 1994). The sediment was sectioned at 1-2 cm intervals and stored cold (8°C) and dark in airtight cans until laboratory analysis.

3.2.2 Sequential phosphorus fractionation analysis

The wet sediment was analysed with a sequential chemical extraction method based on Psenner and Pucsko (1988), modified by Hupfer et al. (1995, 2009) and Jan et al. (2015) (**Paper I**). Different P fractions were identified, specifically: loosely bound P (H_2O-P), P bound to amorphous Fe (Fe_1-P), crystalline Fe (Fe_2-P), P bound to Al (Al-P), P bound to organic matter (Org-P) and P bound to Ca (Ca-P). A known amount of wet sediment (homogenised and sieved) was placed into a centrifuge tube. Different extractants were used in a sequence to detach/dissolve the varying P fractions from the sediment, where RP was measured at every step. Since the extracting agents and conditions define the fractions, they are designated as "operational" phases (Hupfer et al., 2009). Water content was determined by freezing the samples followed by freeze-drying (-40°C, 96 h). Sediment bulk density and organic matter (%) was determined after loss on ignition (550°C, 2hr) (Håkanson & Jansson, 2011). A detailed description of the method can be found in **Paper I.** Phosphorus in the H_2O-P, Fe_1-P (amorphous Fe) and labile Org-P forms are commonly referred to as "labile P" (within the field of lake eutrophication management) and can contribute to internal loading (Huser et al., 2016b, Reitzel et al., 2005).

The method has received some criticism, including that the sample is taken from an anoxic environment and analysed in an oxic environment (Lukkari et al., 2007; Condron & Newman, 2011). Furthermore, there is a concern of P moving between fractions during the analysis (Barbanti et al., 1994). Despite this, the method has shown good reproducibility for different P fractions (Lukkari et al., 2007). Other options would be to use X-ray absorption near-edge structure spectroscopy (XANES) or nuclear magnetic resonance spectroscopy (^{31}P NMR) as a complement to sequential extraction methods (Liu et al., 2013; Werner & Prietzel, 2015). These methods are less accessible and significantly more expensive. If the availability of P is the focus of the study, streambed sediment can be explored by analysing the EPC_0-concentration (Jarvie et al., 2005; Simpson et al., 2021) or with DET-probes (diffuse equilibrium in thin films) (Zhang & Davison, 1999; Jarvie et al. 2008).

3.2.3 Water sampling and analysis

Water samples were obtained for chemical analyses at each sampling location at the time of sediment sampling (**Paper I**). The water samples were analysed at the geochemistry laboratory at the Swedish University of Agricultural Sciences (SLU), certified by the ISO/IEC 17025 standard including TP *SS-EN ISO 6878:2005* (unfiltered, digested with potassium peroxidisulfate solution and analysed with ammonium molybdate spectrometric method), RP (PO_4-P) *ISO 15923-1:2013* (unfiltered, discrete analysis, photometric), total suspended solids (TSS) *SS-EN 872:2005* (gravimetrically, 1.2 µm glass fiber filter) and turbidity *SS-EN ISO 7027:1999 ver. 3* (Turbidimeter Hach 2100AN IS, 870 nm, angle of measurement 90°).

Moreover, a water sampling campaign was conducted at Sävjaån Falebro (location of the in-situ sensor) in 2015-2017, sampling every 2nd to 4th week (**Paper II**). These samples were subsequently analysed at the same laboratory with the same methods as above for turbidity (2015), TSS and TP (2016-2017) to explore the turbidity-TP and turbidity-TSS relations.

3.3 Data treatment

3.3.1 Quality control and sensor maintenance

The sensor was manually cleaned, batteries changed, and data collected every 2nd-6th week, except when the sensor was below ice cover.

Several control examinations were performed to ensure the accuracy and quality of the in-situ sensor (**Paper II, III, IV**). Firstly, turbidity measurements reported from the in-situ sensor was compared with laboratory measurements of grab sampled turbidity to ensure representativeness of the data. A post-calibration was made for 2012-2013 since it deviated from lab measurements, suspected to be an effect of an inappropriate initial calibration. A description of the post-calibration method can be found in **Paper II**. After re-calibrating the sensor in 2014, grab sample turbidity measurements and HF turbidity data showed high concurrence (Sävjaån Falebro r^2=0.95, n=48). From 2015 the in-situ sensor was calibrated according to the manual every six months using standard turbidity solution (Hach StabCal).

Furthermore, the data set was scanned for invalid observations and outliers. Observations outside three standard deviations from the daily mean turbidity (log-transformed data) and without temporally adjacent observations in the same range were identified and evaluated (described in **Paper II**). There are numerous methods (simple to more complex) to ensure sensor data quality. Outlier removal can be done by statistical approaches, residual analysis, and with a combination of pre-set rules and statistical transformations/evaluations (Talagala et al., 2019). The method used was adapted to the data set in question and thoroughly evaluated.

3.3.2 Event identification and analysis

In **Paper III**, the first step of the analysis was to define events by analysing the hydrograph. Event definition is often based on simple criteria, e.g. a specific increase or deviation from baseflow (Eder et al., 2010; Hashemi et al., 2020; Lloyd et al., 2016; Lana-Renault et al., 2011). In Sävjaån, a simple way of determining the start and end of events was not applicable due to the multiple discharge peaks without return to baseflow (**Paper III**). Events were therefore identified by a set of criteria. The start of an event included a sequence of rising/falling subsequent observations, a percentage increase in daily discharge and a minimum discharge threshold. The end of an event was either defined by the start of a new event, or by a baseflow decay function combined with a percentage decrease in daily discharge (**Paper III**). The event definition method was designed to be objective and possible to adapt to the studied hydrograph.

Events were thereafter qualitatively and quantitatively analysed. Calculation of hysteresis indexes (Lloyd et al., 2016) facilitated comparisons between event characteristics. Hysteresis indexes were categorised into five different types (clockwise, anti-clockwise, "figure-eight" patterns – ACA and CAC, and complex loops, Figure 4) based on the categories in Haddadchi & Hicks (2020). A clockwise loop indicates a fast response of the system (eroded material near the monitoring station). In contrast, an anti-clockwise loop is caused by a slower response indicating transport from more distant sources or erosion due to soil saturation (Williams, 1989). "Figure-eight"-patterns suggest that one or more sources are active during an event (Clockwise-Anti-Clockwise CAC and Anti-clockwise-Clockwise ACA)

(Haddadchi & Hicks, 2020). Complex patterns (with one or more linked hysteresis loops) indicate a weak relation between C and Q.

The result of the hysteresis analysis was matched with prevailing environmental conditions during events, e.g. precipitation, soil moisture deficit (SMD), snowfall, snowmelt and snow depth, season, event duration and a suite of parameters describing the change in discharge and turbidity.

Other indexes can be used to describe the dynamics between C and Q, e.g. flushing indexes (Vaughan et al., 2017; Butturini et al., 2008), where the solute concentration at the point of peak discharge is compared to the discharge at the beginning of the storm. These additional indexes could potentially complement the analysis and shed more light on intermediate and high flow results.

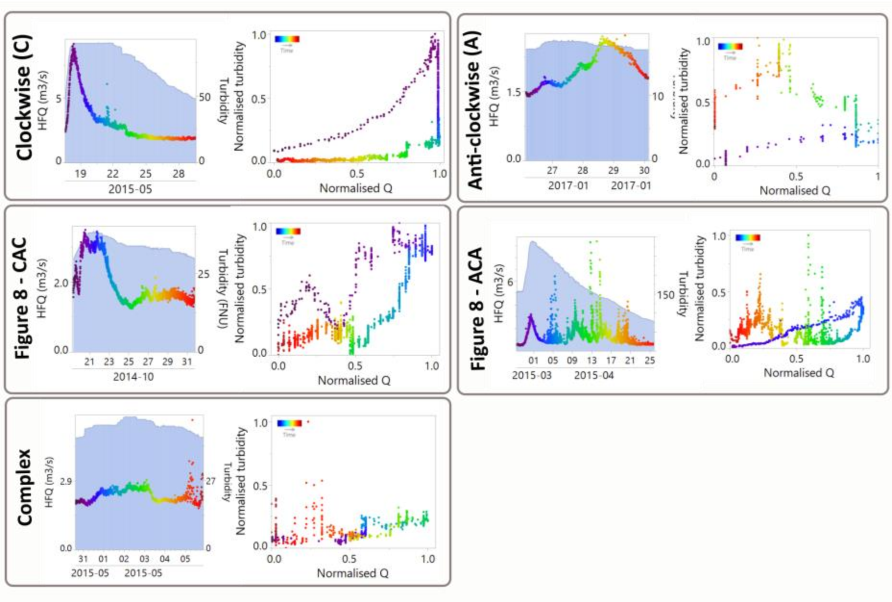

Figure 4. The five hysteresis patterns presented. The right graph describes the turbidity response (right y axis) in relation to Q increase (left y-axis) over time (x-axis). The left graph shows normalised turbidity (y-axis) vs normalised Q (x-axis), resulting in a hysteresis pattern. Time is indicated by colour where blue represents the early parts of the event and red the later parts. Adapted from Paper III.

3.4 Statistical analysis

3.4.1 Exploring variation between sites

In the lake and streambed sediment survey (**Paper I**), it was essential to explore if the results differed between lake/stream and sites. A mixed model nested ANOVA was used where lake/stream was regarded as a fixed effect (as they define all relevant waterbody types) and site a random effect (as each site is a sample from a larger population) (Weiss, 2005). The analysis was performed on TP contents (g/kg) and concentrations (mg/cm^3). Furthermore, the variations in P fractions between waterbody type, site and triplicate sample was explored. A redundancy analysis (RDA) was used (Legendre & Legendre, 2012a), where the total variation was attributed to (i) between water body type (ii) within water body type, (iii) within triplicate replicate samples and (iv) unassigned (unexplained) variation.

3.4.2 Linear regression

When using HF turbidity data as a proxy for TP or TSS, the relationship between parameters was explored with linear regression (**Paper II, Paper IV**). The linear regression model is associated with four assumptions, the data should have multivariate normality, a linear relationship, no autocorrelation and homoscedasticity of residuals (Montgomery et al., 2015). The data set did not have multivariate normality, which would support an argument for data transformation. Therefore, the effects of log transformations with back calculations were explored (Figure 5). Above turbidity of approximately 40 FNU, the log-transformed data fell outside the uncertainty intervals for the linear model, and the effect of the log-transformation became more pronounced. Given the importance of high flow/high turbidity events for flux estimation and our hypothesis of a linear relationship between TP and turbidity, the linear model for TP prediction was retained. Violating the criteria of multivariate normality was acceptable in our circumstances as very similar fits was shown even if the most extreme value was included or excluded.

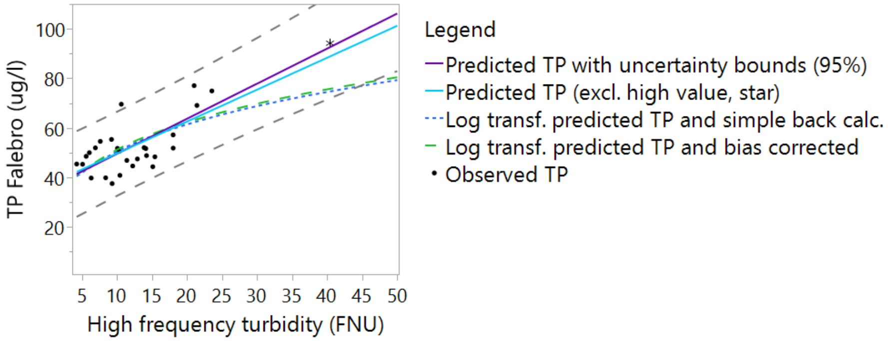

Figure 5. Relationship between HF turbidity (sensor) and TP (lab) 2016-2017, r^2=0.79, p=0.0001, n= 29 (dots). Simple linear regression analysis (solid purple line), confidence intervals for individual predicted values (grey dotted lines). Simple linear regression without one high value (star) (solid blue line). Linear regression when data was log-transformed with simple back-calculation (dotted blue line) and back-calculated and bias-corrected (dotted green line). Adapted from Paper II.

3.4.3 Principal component analysis

A principal component analysis (PCA) was used in **Paper I** and **III** to explore multivariate data graphically. A PCA is used to explore the relationship between several variables, which are in general inter-correlated (Abdi & Williams, 2010). The goal of the analysis is to reduce the dimensions of the data set and bring out the essential information by analysing the structure of observations and variables (Legendre & Legendre, 2012b). A set of new variates called principal components are generated as a product of the extracted information from the input data. Patterns of similarity are displayed in a graph that could be reduced to two dimensions (Abdi & Williams, 2010). When exploring the relative fractions of P, data was Hellinger transformed to facilitate comparison between proportions instead of their absolute magnitude (Legendre & Legendre, 2012b) (**Paper I**).

In **Paper III** the most influential variables for the PCA ordination was used and selected according to King & Jackson (1999). The ratio between observations to variables should be at least 3:1 to ensure stability and reliability in the multivariate analysis, why only the 20 most influential variables in the final ordination was kept.

3.5 Process-based modelling

3.5.1 PERSiST

PERSiST (Precipitation, Evapotranspiration and Runoff Simulator for Solute Transport) is a semi-distributed rainfall-runoff model that simulates water movement in soil and the stream on a daily time step (used in **Paper III** and **IV**) (Futter et al., 2014). Incoming precipitation is routed through different "soil boxes" (surface flow, soil water and groundwater) and calibrated to observed streamflow. Hydrological inputs are generated that can be used for the INCA-family of models, e.g. hydrologically effective rainfall and soil moisture deficit (SMD) (Futter et al., 2014). Other models could also produce these inputs, e.g. the HBV-model (Bergström, 1976). However, the newest version of PERSiST (Futter et al., in prep) was considered more suitable where updates regarding the shape of stream channel and Manning's roughness coefficients were incorporated.

The forcing time series of daily temperature and precipitation was in **Paper III** obtained from a monitoring station in central Uppsala (7 km from the sensor locations), data was collected every 10th min. In **Paper IV** gridded meteorological data was used from the E-OBS data set (daily) (Cornes et al., 2018), following the recommendations in Ledesma & Futter (2017). Different forcing data series was used in the two papers since HF precipitation data was needed to analyse sub-daily patterns in **Paper III** to enable a detailed analysis of the events. Since the same dataset was going to be used for calibration of PERSiST and analysis of the events, the monitoring data was chosen due to the higher resolution. The catchment was treated as one unit and not split into sub-catchments.

The model was calibrated analogously to the protocol described in Ledesma et al. (2012) (it was then used for the INCA-C model). The starting point was an initial manual calibration, followed by a Monte Carlo (MC) exploration of parameter space. Model performance was evaluated against Nash Sutcliffe statistics for transformed (logNSE) and untransformed data (NSE) and the ratio of variance (VR). Observed streamflow data from the SMHI station (described in *section 3.1*) was used for calibration.

3.5.2 INCA-PEco

The process-based model INCA-PEco was used to simulate TSS and TP transport in Sävjaån under varying conditions and distant points in time

(**Paper IV**). INCA-PEco (Integrated CAtchment model for Phosphorus Ecology) simulates temporal variations in the discharge and water quality dynamics both in the land and instream components of a river system (Crossman et al., 2021). The model has previously been used to understand catchment scale P dynamics and assess mitigation strategies to reduce the P load (Jin et al., 2013) and potential effects of climate change (Crossman et al., 2013).

The model is spatially adapted to the catchment of interest, which makes it possible to specify the stream network (single stem to a fully branched stream network), specify characteristics of sub-catchments, as well as parameterization of different land use classes (Crossman et al., 2021). Terrestrial simulations are based on a 1 km^2 cell (input, output, store) for the user-specified land use class, which is then up-scaled to a sub-catchment level and finally, the river network (Whitehead et al., 2011).

In the land phase hydrology, water is routed through three potential flow pathways, quick flow, soil water flow and groundwater flow (Crossman et al., 2021). Quick flow is vital for terrestrial erosion and sediment transport. Phosphorus is delivered to the stream from the different flow pathways, potentially from the storage in the soil, groundwater or eroded material. The instream representation of the model includes exchange with the streambed sediment both regarding suspended solids and RP. Phosphorus is transported downstream after accounting for point sources and instream processes, where the mass balance operates at all levels. The model produces daily estimates of discharge, TP, TSS, PP, RP concentrations and loads.

INCA-P (the precursor to INCA-PEco) has received criticism for being overly complex, with many uncertain parameters needing calibration (Jackson-Blake et al., 2017). In our case, INCA-PEco was chosen because of the processes represented, which allow us to develop our conceptual understanding of the catchment.

3.5.3 Calibration and testing

In **Paper IV**, INCA-PEco was calibrated and tested against two data sets during six years (2011-10-01 to 2017-09-30). One data set was based on monthly grab samples analysed in the laboratory (low-frequency, LF). The second was based on HF turbidity used to calculate TP and TSS (based on the relationship in the linear regression).

The model was divided into four reaches and five land use classes (urban, intensive agriculture, non-intensive agriculture, wetland and forest) based on GIS analysis.

The model was manually calibrated following the strategy proposed by Ledesma et al. (2012). First, some parameters were fixed based on literature, GIS-analysis, the previous PERSiST calibration and knowledge about the catchment. Second, some parameters and processes were excluded (mainly processes connected to biological oxygen demand and dissolved oxygen) to simplify the model. Finally, the remaining parameters were explored to guide the allowed range of variation in the automated calibration.

An MC exploration of parameter space was conducted following Futter et al. (2014). The MC exploration followed a hill-climbing algorithm and consisted of multiple iterations (100 chains, 200 model runs/chain). Parameter sensitivity was explored, and the range for exploration was updated accordingly. Calibration strategies were explored (weighting to different objective functions) with the MC analysis (**Paper IV**). A set of performance statistics was used to evaluate the modelled result against observed data, including the coefficient of determination (r^2), Nash-Sutcliffe efficiency (NSE) and the variance ratio (VR) (thoroughly described in **Paper IV**). The results were evaluated in ensembles (10 best parameter sets for LF/HF, ranked based on a combination of TSS and TP performance statistics) to avoid equifinality.

The model was tested for a period distant in time (LF monthly grab samples from 1979-1985) to explore the robustness of using the model to predict the future. A cross-testing was performed where the HF calibration was used to predict LF data and vice versa.

The parameter sensitivity analysis was also used to identify the most influential parameters for the model response (Spear & Hornberger, 1980). Parameters from the best performing parameter sets from each MC run was analysed for rectangular/non-rectangular distribution. KS-statistics were applied to each parameter to evaluate the null hypothesis (if the prior and posterior distribution differed). Parameters with a p-value <0.05 were ranked from 1-5 and kept for further analysis.

4. Results and Discussion

4.1 Legacy phosphorus in the lake and streambed sediment

Phosphorus stores in lakes are often described as a significant risk concerning internal loading and thus eutrophication (Schindler, 2006; Søndergaard et al., 2003; Agstam-Norlin et al., 2021). Even if the relevance of streambed sediment P stores has been established (Sharpley et al., 2013), they are less well studied compared to lake sediment. With an enhanced knowledge about these stores, catchment processes can be better understood and model calibrations better constrained. This chapter will present the results from **Paper I**.

4.1.1 Large amounts of TP stored in the sediment

Total P content (g/kg DW), TP concentrations (mg/cm^3) and P fractions were studied in the lake and streambed sediment in Sävjaån catchment (**Paper I**). The type of bed sediment varied greatly between sites, from dense, clay-rich sediment with low organic matter and water content (agricultural streams close to the catchment outlet) to sediment with high water and organic matter content and lower density (forested streams and lakes). Generally, higher TP contents were found in lake sediment (surficial layers 0-4 cm 1.31-2.00 g/kg DW) than the streambed sediment (0.073-1.57 g/kg DW). The ranges were comparable to previously reported levels for lake (Boström et al., 1988; Rydin, 2000; Søndergaard et al., 1996) and streambed sediment (Audette et al., 2018; Dorioz et al., 1998; Noll et al., 2009).

Concentrations of TP varied greatly between sites (0.21-1.96 mg/cm^3), where levels were high in streambed sediment close to the catchment outlet

(1.72-1.96 mg/cm^3, Figure 6) compared to lake sediment (0.22-0.57 mg/cm^3) and headwater streambed sediments (0.29-0.72 mg/cm^3). Some previous studies have shown an enrichment of TP in sediment (Owens & Walling, 2002) and P associated with TSS (Némery & Garnier, 2007) downstream in a catchment explained by the integrated signal from a larger contributing area. However, there was also a positive correlation between clay soils in the drainage area of the sampling site and TP concentration (r^2=0.71, p=0.0001), indicating that the higher proportion of clay-sized particles in the sediment combined with lower water content likely affects the TP concentration.

When evaluating the variance between the different sampling sites in TP content and concentration, most variance was attributed to site compared to waterbody type (lake/stream) and between replicates. The results confirm the significance of legacy P in streambed sediment, even if an estimation of the total amount of P stored in the studied lakes was greater (6.0 tonnes) than streams (2.1 tonnes) due to the larger lake sediment area.

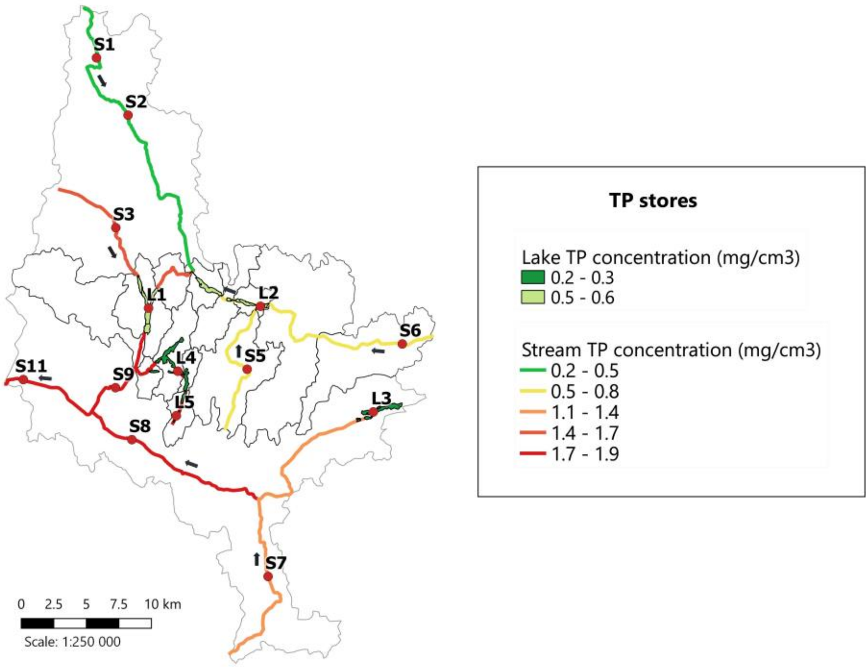

Figure 6. A map visualising the TP stores in the catchment, colours indicate TP concentrations in lakes and streams. Concentrations have been extrapolated from the sampling site to stream sections/lake areas. Adapted from Paper I.

4.1.2 Land cover effects on P fractions

Headwater streams (land cover mostly forest on sandy till soils) had similar fractional P composition as lakes, where the dominating fractions were Org-P and Fe_1-P (Figure 7) (**Paper I**). Since small headwater streams have lower water to bed sediment ratio, they have great potential for physical, chemical and biological P exchange (Withers & Jarvie, 2008). Since these streams had high TP contents and a large proportion of potentially labile P, they are suggested as especially interesting regarding sediment P processes.

In the agricultural streams, Ca-P was the dominating fraction, likely linked to the high proportion of clay particles in the samples. Less fresh sediment was accumulated in these sites, potentially due to the increase in discharge that mobilises organic matter and lower density particles.

The land cover organisation where agriculture is mainly located on clay soils, combined with limited organic carbon input in agricultural areas (Stutter et al., 2018), makes the land cover effect pronounced in sediment composition and P fractions. The results suggest that P stores in larger agricultural streams are more transitory, which indicates that TSS could be an essential vector for P transport in these streams. A study in three smaller agricultural catchments in central Sweden (6-33 km^2) has shown the importance of TSS as a vector for P. The study also showed seasonal dynamics in P fractions associated with TSS (Sandström et al., 2021), where TSS Fe-bound P was high during summer and corresponded to periods with low discharge. These findings could indicate an active redox process in the streambed sediment affecting the water column chemistry (Sandström et al., 2021).

Two forested headwater streams and one agricultural headwater stream had comparable amounts of labile P (MQ-P, Fe_1-P, Org-P) as lakes. In contrast, the streams with much Ca-P are considered less prone to release P since the fraction is considered more stable over time (Noll et al., 2009). However, the driving processes for internal loading is hypothesised to be partly different between lakes and streams. Consequently, the labile fraction might need to be reconsidered for streams.

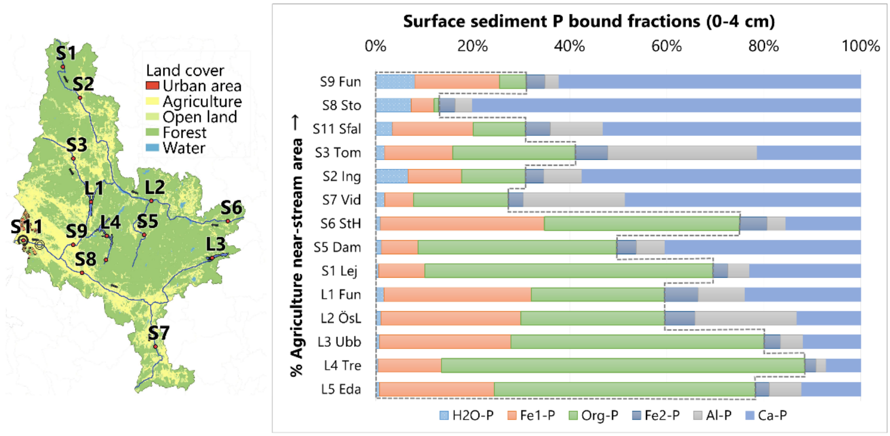

Figure 7. Fractions of P in the surface sediment (0-4cm) in all sites (expressed in %). The sites are ordered after percentage agricultural land in the near-stream area (200m on each side of the stream x 1km upstream). The dashed line separates the fractions that are considered labile. L in the site name denotes lake, and S-stream. Adapted from Paper I. ©The National Land Survey.

4.2 Resolution of monitoring

High-frequency data from in-situ sensors could aid in a better understanding of P mobilisation processes and transport. This chapter presents the benefits and challenges of HF monitoring for improved management of P transport (**Paper II and IV**).

4.2.1 Relationship between turbidity and TP or TSS

Before analysing the potential benefits of HF monitoring, the use of turbidity as a proxy for TP or TSS need to be evaluated and discussed. In Sävjaån, linear regression showed a good relationship between HF turbidity and TP (r^2=0.64, n=28) as between HF turbidity and TSS (r^2=0.68, n=29) (**Paper II**). The relationship between parameters is site-specific, and previously reported factors that could influence correlation was investigated, e.g. proportion RP/TP (Jones et al., 2012), seasonal effect (Bogen, 1992), high/low discharge (Walling & Morehead, 1987) and rising/falling limb of the hydrograph (Stutter et al., 2017).

Table 2. Results from linear regression analyses (y=α+βx) (total phosphorus: TP, total suspended solids: TSS, high frequency: HF), p<0.001 in all cases. Data from 2012-2019 for LF data and 2012-2017 for HF data. Adapted from Paper II.

Linear regression term: y	x	α	β	r^2	SE intercept	SE slope
Turbidity grab sample Falebro	HF turb	-0.5	1.0	0.95	0.7	0.04
Turbidity grab sample Kuggebro	HF turb	-1.6	1.0	0.87	1.5	0.07
TP Falebro	HF turb	35.4	1.4	0.64	3.1	0.2
TP Kuggebro	LF turb	31.2	1.9	0.86	2.1	0.08
TP Ingvastra	LF turb	18.2	2.3	0.45	4.8	0.3
TSS Falebro	HF turb	4.2	0.4	0.68	1.0	0.06
TSS Kuggebro	LF turb	-4.6	1.1	0.70	1.9	0.07
TSS Ingvastra	LF turb	-0.9	0.8	0.77	1.5	0.1

The relationship between turbidity and TP is hypothesised to get better if a large part of P is associated with particles (Jones et al., 2012) since turbidity is measured optically. Hence, a varying and sometimes large part of TP being RP in Sävjaån (7-85%, average 45%) could negatively affect the relationship. The intercept (α) in the linear regression equation (HF turbidity and TP Falebro) was high (35.4 µg/l, Table 2), indicating that at low turbidity TP concentration in the stream was high. The intercept is also remarkably close to the average RP concentration of 33.5 µg/l (Falebro, Table 1), which would suggest that RP can explain the residuals. Due to the high proportion of clay in the area close to the sensor, a hypothesis would be that the turbidity-TP relationship and the varying RP concentrations could be connected to colloids. Colloids and nanoparticles could be essential carriers of P (Gottselig et al., 2017) and could be important in the relationship between turbidity and TP. Furthermore, the HF turbidity-TP relationship was examined in terms of seasonal effect, high/low discharge and rising/falling limb of the hydrograph, but no statistically significant differences were apparent.

When comparing the turbidity-TP relationship from Falebro with the two sampling locations in Ingvastra and Kuggebro (map Figure 3), the relation was robust within the catchment. The intercept of the LF turbidity-TP relation from Ingvastra is lower than Falebro and Kuggebro further downstream in the catchment (18.2 compared to 35.4/31.2). Also here, the value of the intercept (18.2 µg/l) is close to the mean RP concentration (23.1 µg/l) in Ingvastra (Table 1, Table 2).

Further studies are needed to understand the driving mechanisms behind the turbidity-TP correlation, e.g. exploring the variation in particle composition, organic matter and its effect on both turbidity and TP. In Swedish rivers, the correlation (turbidity-TP) varies largely (r^2= 0.1-0.9, n=84) between sites (Villa et al., 2019). Hence, the relationship between turbidity-TP and turbidity-TSS needs to be examined before deploying an in-situ sensor.

4.2.2 Monthly grab sampling versus HF monitoring

Estimating annual mean and flux

In **Paper II**, annual mean and flux calculations based on HF and LF were compared. Mean turbidity did not differ much between the in-situ sensor and lab measurements (17.9 FNU versus 18.2 FNU), indicating that in Sävjaån mean concentrations are well described by monthly grab sampling (**Paper II**).

Although peak concentrations were missed with grab sampling (e.g. Figure 8, panels a and c), this did not significantly affect the flux calculations. In five out of six investigated years, TP fluxes estimated from grab sample and HF data were similar (grab sample estimates -10% to +13% P transport compared to HF estimates). The exception was in 2013 where a grab sample was taken during a 50-year spring flood, which resulted in 56 % larger flux calculated from grab samples. Previous studies have generally shown larger estimated fluxes when using HF data (Jones et al., 2012, Cassidy & Jordan, 2011; Villa et al., 2019), capturing more of the instream TP variation.

Stream discharge at the time of grab sampling and randomly capturing concentration peaks (that affected the interpolation during the nearest months, Figure 8, panel b) had a significant effect on the flux magnitude, affecting the comparison between methods. High concentrations during low discharge, had a moderate impact on flux calculations (Figure 8, panels c and

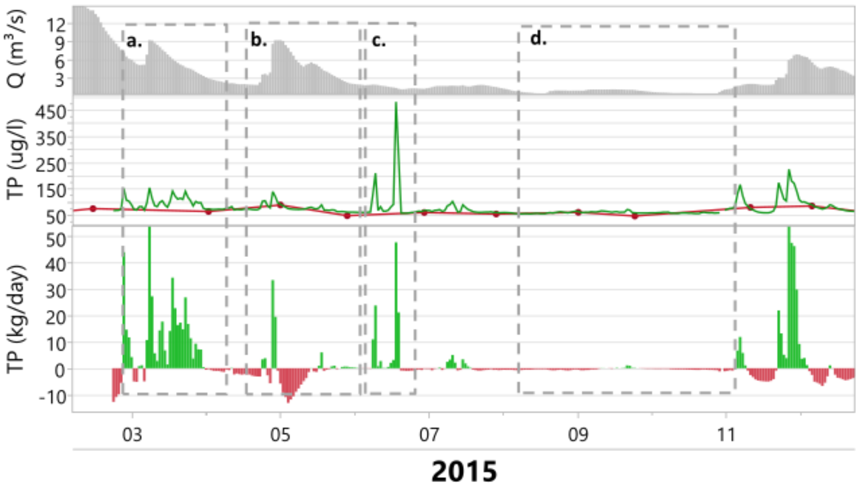

Figure 8. Three panels describing variation in Q (top), TP concentration (middle) and TP load (lower) during 2015. Grab sampling and linear interpolation (red) and TP calculated from the sensor (green). Larger loads from grab samples (lower panel) are indicated by bars below 0. Adapted from Paper II.

d). The most critical periods for increasing the resolution in data was episodes with high discharge, e.g. during winter, spring and autumn.

Water quality modelling

Calibrating the water quality model INCA-PEco to the LF and HF datasets (**Paper IV**) resulted in new insights regarding calibration strategies, model performance evaluation, and improved process understanding.

The two data sets were profoundly different regarding variation, where HF data varied more (Table 3). The NSE was shown to be hard to combine with the HF data calibration (Table 3). Since the NSE include the mean for observed values as a baseline, data sets that, e.g. have a strong seasonal effect will get a poor NSE (Schaefli & Gupta, 2007). For a HF dataset, the mean is of limited interest (and is close to the mean of LF data, **Paper II** and **IV**). The motivation for increasing the monitoring frequency would be to capture more of the instream dynamics, which is not captured by the mean of concentrations. The poor NSE could also depend on a mismatch in timing between observed and simulated concentrations. The mismatch could be related to hysteretic behaviour of TSS and TP related to discharge (**Paper III**), which is not captured in the model. Another explanation would be processes happening on a finer time scale than daily.

Generally, the LF dataset produced better model performance statistics than the HF data (present day and hindcast) (Table 3). Calibration to HF data resulted in higher concentrations of TSS and TP when evaluated against r^2 (evaluated against NSE, the results were varying). In a cross-test, the HF calibration evaluated against observed LF concentrations gave better model performance (r^2-value) than vice versa.

The HF data was more challenging to calibrate due to the significant temporal variation and a larger number of observations that needed to be reproduced in the modelled output. The benefit of the HF data was the better description of temporal patterns. With more frequent observations, temporal dynamics are better constrained, which could be the reason for the better performance of HF calibration predicting LF data in the cross-test. The result leads to the question of how well LF calibrated models predict actual water quality instream variation.

Sensitive parameters varied between the two datasets. For the HF data, stream hydrology related parameters were sensitive (Table 3), potentially related to material mobilised during events (connected to **Paper III**). For LF data, a TP related parameter and two parameters associated with terrestrial

Table 3. Differences observed from calibrating INCA-PEco to LF and HF data.

Differences LF/HF data		
	LF data	HF data
Variation in data set, CV_{TP}/ CV_{TSS}	54/88	65/157(daily average)
Performance statistics present day	Better performance statistics (r^2, NSE)	Difficult to evaluate by NSE
Hindcast	LF/HF gave an equal level of r^2, LF better NSE TSS	
Cross-testing	HF→LF, better r^2, worse NSE	
TSS and TP concentrations	HF data based on best r^2-coefficients, systematically higher TSS and TP concentrations. Evaluation based on best NSE performance: mixed response.	
Sensitive parameters	A TP related parameter (enrichment of P), land hydrology related parameters	Stream hydrology related parameters

hydrology were sensitive. Hence, different processes are more pronounced depending on the input data used for model calibration.

4.3 Discharge variation leading mobilisation processes

The hydrology of a catchment is the basis for P transfer; in **Paper III** different flow ranges showed varying responses in HF turbidity. Hence, discharge variation significantly affects P transfer and could guide us in the best ways of monitoring and mitigating during different hydrological regimes.

4.3.1 Event definition method matters

In **Paper III**, the event definition procedure influenced and constrained the subsequent analysis of hysteresis patterns. The typical approach of using deviation from baseflow as a threshold for the start and end of events (e.g. Hashemi et al., 2020) was not applicable in Sävjaån. Consequently, an event definition method was developed that was suitable for the Sävjaån hydrograph.

Potentially, HF turbidity monitoring could be used to define events in combination with discharge variation. This idea is based on the observation that there could be turbidity events without any variation in discharge (**Paper III**). These turbidity events often occurred on the falling limb of the hydrograph (5/7 cases), when SMD was increasing (5/7 cases). The events were often connected to precipitation the same day or within three previous days without a response in the hydrograph. Concentration events could then show the importance of processes regarding stream ecology, while events defined by discharge would be more critical for material transport.

4.3.2 Varying turbidity patterns during high and low flow

A qualitative analysis of events in **Paper III** (n=76) showed that change in turbidity was correlated to discharge only between the 50th and 97.5th percentiles of the flow range. Hysteresis analysis was efficient and informative for these intermediate flow events (see *4.3.3 A short distance from source to stream*). Turbidity variations within events were not strongly correlated during low (mean Q <2m^3/s) and high (max Q >15 m^3/s) flow periods. These limitations of the method have not been seen in previous literature.

Low flow events were in most cases coupled to a short time variation (with a period ranging from 8 h - 3 days) where turbidity and discharge were uncoupled (n=34). The events showed general low turbidity (average 10 FNU) and amplitude of turbidity variation (10 FNU), as well as a low mean discharge (<1 m^3/s). Complex hysteresis patterns have previously been connected to heterogeneous spatial and temporal distribution of rainfall events (Haddadchi & Hicks, 2020) or explained by biological factors (Loperfido et al., 2010). However, the short term variation could neither be connected to season or time of the day, which would indicate a biological effect.

Events with high flow (max Q >15 m^3/s) also showed a disconnection between turbidity and discharge. These events (n=4) were connected to the yearly spring flood or snowmelt. Hydrological connectivity during these events was probably high, and many processes were active in large parts of the catchment contributing to turbidity.

4.3.3 A short distance from source to stream

The analysis of HF turbidity-Q hysteresis in Sävjaån during events showed that a clockwise pattern was the most common (38% of analysed events). The hydrological driver was often precipitation (91% of analysed events), either rain or snow. The clockwise events were also connected to snowmelt (61% of analysed events) and the SMD indicative of wetter conditions. In the PCA, clockwise events were associated with the spring and winter season and discharge related parameters (maximum Q and maximum snowmelt).

Some of these events were affected by processes mobilising material at snowmelt or heavy rainfall, which is subsequently mirrored in the stream. Often the clockwise response is connected to sources near the stream (Haddadchi & Hicks, 2020; Lloyd et al., 2016), e.g. the stream channel, adjacent riparian areas (Sherriff et al., 2016), or tile drains (Bowes et al., 2005). Wet soils (low SMD) and clockwise hysteresis patterns can be related, as pre-wetted material potentially have faster erosion rates (Lawler et al., 2006). Turbidity in Sävjaån has been hypothesised to be affected by colloidal material since a large proportion of the soil near the sensor consists of clay-sized particles (**Paper II**). A dominating clockwise response in streams with a large proportion of fines has previously been observed (Rose et al., 2018). As fines are mobilised faster than larger material, the particle size fraction could also influence the fast response during events in Sävjaån. Also, when using a process-based water quality model to explore the importance of different co-existing processes in the catchment, erosion processes were highly important (**Paper IV**). Flow erosion was a central process in all different calibrations, which shows the importance of terrestrial sediment delivery as a control on instream TSS and TP concentrations.

The meso-scale of the catchment probably affected the hysteresis patterns for some of the anti-clockwise, ACA and CAC events. The travel time from the furthest part of the catchment was estimated to be between 2-5 days. Generally, the change in discharge and turbidity was small for the anti-clockwise hysteresis patterns (7/60), indicating a transport limitation. ACA (6/60) and CAC events (7/60) indicate a contribution from two or more sources of material (Haddadchi & Hicks, 2020) or increased hydrological connectivity during the event (Rose et al., 2018), leading to transport of material with different travelling times. However, one or several turbidity peaks were associated with precipitation in many (10/13) of the studied CAC and ACA events. This pattern could indicate a direct response of mobilised material from precipitation (not necessarily increase in discharge) close to the monitoring station. Complex hysteresis patterns (17/60) were often displayed during summer and low flows.

Before in-situ sensors became available, C-Q analysis was performed to explore hydrological pathways and water chemistry processes (Glover & Johnson, 1975; Evans & Davies, 1998). With the HF data, much more information can be extracted compared to LF data. However, the high volumes of data must be efficiently analysed and understood to produce decision support for environmental management, where C-Q analysis shows to be a promising technique.

4.4 P transport simulated by water quality modelling

Water quality modelling is a valuable tool to explore processes and changed conditions. INCA-PEco was used in Sävjaån catchment to test hypotheses and develop the conceptual understanding of the catchment (**Paper IV**). Model performance was good enough for these purposes and showed well described temporal dynamics. The model also performed well during an independent historical time period, giving additional confidence in predicting future conditions.

TP and labile P concentrations in the streambed sediment were used to constrain the parameters in the model (**Paper I**). These data also helped to produce plausible EPC_0 -dynamics. Conclusions from **Paper II** were used to build the input data set and interpret the results. Hysteresis patterns based on **Paper III** could not be represented in the current version of the model calibration.

5. Implications for management and transferability

5.1 Slowing down mobilisation from the source

The results from **Paper I** suggests that approximately 2 tonnes of P are stored in streambed sediment in the catchment, part of this store is regarded as labile during certain conditions. 63% of the sampled streams in the catchment are covered by drainage permits (County Administrative Board of Uppsala, 2020). These permits allow repeatedly digging out the newly deposited sediment, without notifying authorities, to maintain the depth and position in the stream/ditch over time (The Swedish EPA, 2009). Dredging of these streams could increase the speed of P transport downstream both during the digging and afterwards, as freshly deposited sediment, biotic communities and vegetation are removed (Smith & Pappas, 2007). Well maintained ditches and streams are a tool to ensure efficient removal of water from the landscape. However, dredging and other maintenance need to be done during times of the year when nutrient loads are expected to be low and not coincide with fertiliser application. The ditch cleaning masses also need to be removed from the site to avoid material and P leaking back into the water.

Furthermore, the analysis in **Paper III** showed that a fast response in the stream is typical when the discharge starts to increase. Fast material transport could be counteracted by measures like (1) riparian vegetation - especially during winter and snowmelt, (2) the use of buffer strips/set-aside areas, (3) drain discharge on vegetated areas and (4) cover crops.

5.2 Processes, monitoring and measures in different flow ranges

5.2.1 Low flows

Regarding low flows, there is an overall risk that high RP concentrations can significantly increase biomass growth and cause a profound eutrophication effect (Biggs, 2000; Jarvie et al., 2005). Sources of available P could include point sources, e.g. septic tanks or the streambed sediment. In Sävjaån, the streambed sediment has been shown to contain significant amounts of P, bound to different fractions (**Paper I**). If these low flow periods occur during summer, which is often the case in Sävjaån, the biologic activity is high. In highly productive systems, anoxic conditions could favour RP release from Fe-bound P in the sediment (Smith et al., 2011).

Reactive P concentrations need to be kept on low levels to protect the surface waters during this sensitive time (Figure 9). Dredging of ditches needs to be carefully planned to avoid these low flow periods in the summer. Furthermore, point source pollution from, e.g. private sewage, should be surveyed and mitigated. Stream channel management is an essential tool that should be further investigated, where aquatic plants could potentially be managed and used as a phytoremediation method.

5.2.2 Intermediate flows

At intermediate flows, there is a risk of fast transport of fines from the streambed sediment and near-stream areas when the flow increases during events (Figure 9). These events could potentially carry large loads of P that is transported further in the catchment.

These events were successfully explored with hysteresis analysis (**Paper III**) and have also been shown to be essential to monitor at HF (**Paper II**). Critical measures to avoid this fast transport of material to/in the stream could be to minimise the time of bare soil during the winter period (e.g. in the riparian area and winter crops in the near-field areas) and promote a good soil structure. The riparian buffer zones are critical, especially in erosion-prone areas of the landscape.

5.2.3 High flows

High flow events, e.g. spring floods, transport large loads of TP and TSS further down the system (**Paper II**). During these periods, large areas near the stream, stream banks and riparian areas can be flooded (Figure 9). Ponding surfaces can be seen in the landscape partly due to the still frozen ground. There is a high risk of overland flow and erosion.

Here, HF monitoring is of high value since it can help us analyse the timing and magnitude of the flooding events, potentially giving an early warning when water chemistry parameters change. It shows the importance of deploying in-situ sensors over the winter to capture freezing/thawing events together with the spring flood.

These events are of high relevance to soil and water managers. Erosion potential needs to be decreased during these periods, e.g. by vegetation and efficiently drained fields. The water holding capacity of the landscape needs to increase, e.g. by constructed or restored wetlands.

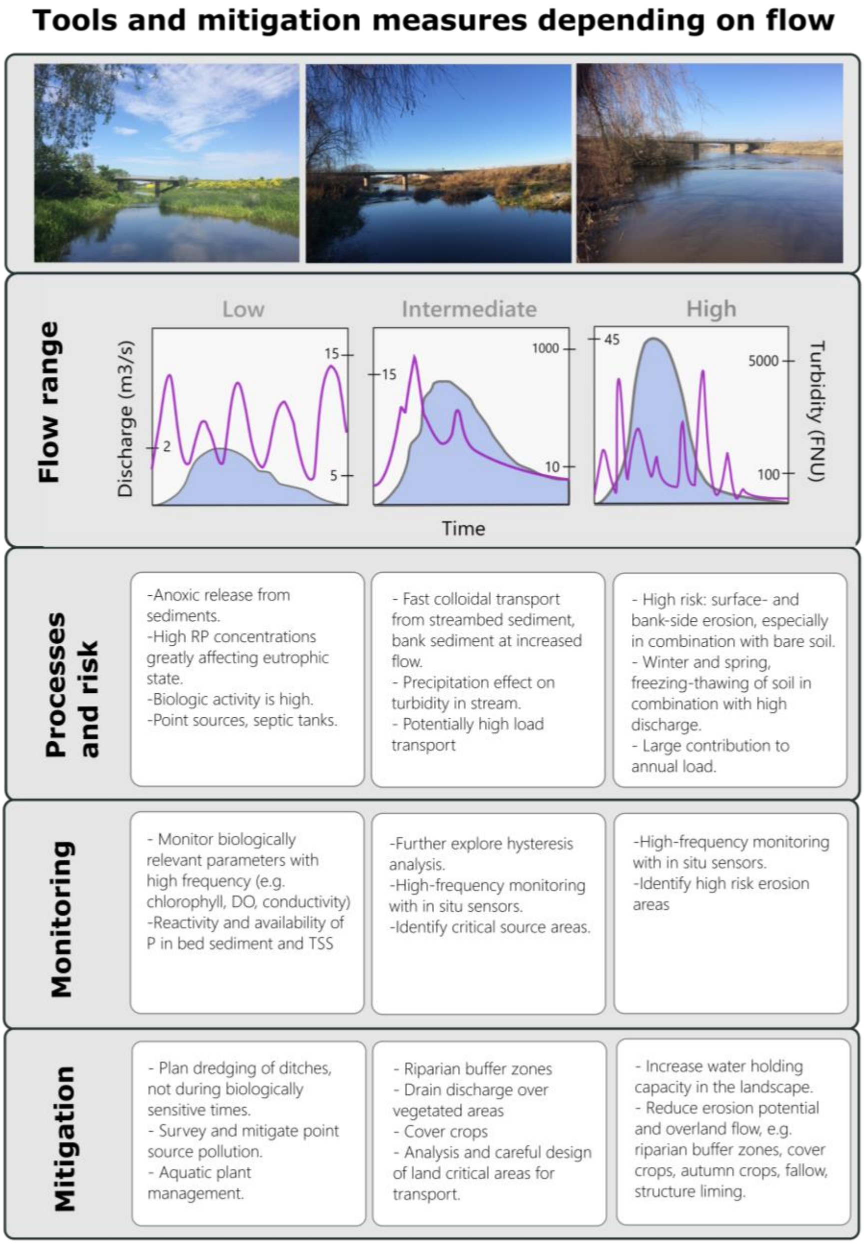

Figure 9. Summary of (1) processes and risks, (2) monitoring and (3) mitigation aspects depending on the flow range. Adapted from Paper III.

5.3 Transferability to other catchments

5.3.1 Streambed sediment

Studies of P in streambed sediments are rare in Sweden. In Sandström et al. (2021), three small agricultural catchments in central Sweden were studied regarding TP amounts and fractions. Concentrations were comparable (0.45-2.01 g/kg DW) to P stored in streambed sediments in Sävjaån (0.07-1.57 g/kg DW). It would not be unlikely that many agricultural streams are important P stores, even if it cannot be corroborated here due to a lack of data. These P stores could be released during low flow periods, alternatively transported as TSS downstream in the catchment. Further investigating the role of streambed sediment in the P transfer continuum is highly relevant, especially due to the current rules about ditch maintenance.

5.3.2 Mobilisation processes and transport

The proxy relation between turbidity-TP and turbidity-TSS is site-specific. However, it was shown to be stable within Sävjaån catchment, indicating an influence from catchment geology (Table 2). The literature has consistently shown that a new proxy relation must be formed and evaluated before using turbidity as a surrogate for any other parameter (e.g. Stutter et al., 2017; Skarbøvik & Roseth, 2015).

Monitoring frequency has previously been shown to affect flux estimations (Meybeck et al., 2003, Jones et al., 2012, Villa et al., 2019). The most benefit from increased sampling frequency is shown in flashy streams, where a lot of the material transport happens in a short time (Jones et al., 2012). The reactivity and material transport in a catchment depends on, e.g. presence of lakes, topography, soil permeability and erodibility, snowmelt runoff regimes and contributing catchment area (Meybeck et al., 2003). In Sävjaån, the general flux estimates from HF monitoring did not differ much from using monthly grab samples and linear interpolation (**Paper II**). Even if Sävjaån is not regarded as flashy, the catchment has been shown to give a fast response to discharge changes during wet conditions in the winter and spring season (**Paper III**). Moreover, these periods have been identified as necessary also for monitoring with HF in **Paper II**. These conclusions could be transferred to other meso-scale catchments, explicitly that HF monitoring in larger catchments might not give a substantial difference when calculating annual loads compared to smaller flashier catchments. However, other values can be emphasized.

Hysteresis analysis could be used to get insights into mobilisation processes and to understand larger transport events. In streams with smaller contributing areas, HF information is easier to interpret. On the other hand, the larger streams are potentially more interesting from a management perspective. Therefore, it is vital to continue investigating efficient HF data processing methods to better understand and manage larger scale catchments.

The results from **Paper III** are site-specific, but the method proves to be helpful to get insight into mobilising processes in a meso-scale catchment. A spatial dimension of HF data would be needed to understand more about the impact of the meso-scale. However, the developed event definition method can be applied to other hydrographs as a basis for further exploration of hysteresis analysis as a tool.

5.3.3 Water quality modelling

The results from **Paper IV** are site-specific, especially when it comes to process representation and sensitive parameters. The challenges experienced in evaluating HF data with current performance statistics are likely applicable to other studies. The HF data was shown to be a valuable resource to better represent TSS and TP dynamics in the stream, which would be a reason to use it more extensively in modelling in the future.

6. Conclusions and future perspectives

The main aim of this thesis was to provide support for improved management decisions based on four studies describing P transfer in a mixed land use catchment in Sweden.

With an interdisciplinary approach, using methods from lake management, analytical chemistry, soil science, aquatic science, biogeochemistry, hydrology, statistical and modelling approaches, this thesis contributes to an evidence base for better-informed management decisions.

With the studies presented here, we can conclude that it is not enough to control catchment land cover practices; we also have to control the legacy P sources as they probably mask improvements made in the fields. A significant amount of P is stored in lake and streambed sediment throughout the catchment (**Paper I**). These stores vary in immediate availability of P but are nevertheless crucial for management consideration; land cover and stream order affect sediment composition and the dominant P fractions.

Furthermore, HF data coupled with appropriate analysis methods opens the possibility of entering a new paradigm in environmental monitoring. With an improved understanding of short term variation in stream chemistry, we can (1) identify critical periods for P transport, (2) practically address mobilisation processes and (3) better parametrize our water quality models (**Paper II-IV**).

High-frequency turbidity was shown to be a good proxy for TP, and the relationship was stable throughout the catchment. Average concentrations and low flow periods were sufficiently characterised by monthly grab sampling. In contrast, high flow periods benefit from a higher temporal monitoring resolution when calculating P load from the catchment (**Paper II**). The appropriate monitoring method should be chosen depending on the management purpose, but deploying sensors year-round is vital.

Process understanding and management relevant insights were provided from the hysteresis analysis, where HF turbidity and discharge were compared and matched with prevailing environmental conditions (**Paper III**). The majority of events exhibiting a fast response during wet conditions suggest that riparian vegetation, especially during the winter, is crucial to limit material transport. Separate consideration of low, intermediate and high flow ranges could guide further studies and mitigation strategies. Low flows pose a risk of direct eutrophication in the water body, while intermediate and high flows risk transporting large quantities of P along the P transfer continuum.

Using HF data in water quality modelling improved simulations of temporal dynamics of TSS and TP concentrations in the stream. However, calibration and evaluation methods cannot be applied uncritically, as they are not adapted to the short term variation in the data. This suggests that further studies are needed to guide the handling of HF data in process-based models.

Monitoring data is the foundation for sustainable water management today and in the future. Low frequency, long term data provides the foundation for environmental management. It guides us in prioritisation and status assessments. Observations with higher temporal and chemical resolution (both regarding stores and from the in-situ sensor) can improve our conceptual understanding of the P transport in the catchment. The results from these four studies provide suggestions for (1) near and instream management considerations, (2) practical tools and techniques to use HF data and (3) a framework for flow dependent monitoring and management.

6.1 Future perspectives

The work presented here reinforces the need to understand legacy sources of P and highlights that our present conceptual models of P transport need further refinement. Specifically, the location of P stores in the catchment and the chemostatic/chemodynamic nature of material delivery in agricultural catchments must be better understood.

To assess the risk of eutrophication at low flows, we need to consider best practices of ditch management (e.g., timing and methods). To better understand the potential availability of the P stores, we need to explore the short term dynamics of P in sediment. Which fractions become available at what time? By knowing more about the availability of P fractions (in streams, lakes and wetlands), different levels of risk could be communicated to managers and authorities. Biological relevant parameters could be monitored with in-situ sensors to learn more about the active processes during low flows (e.g. chlorophyll, DO and conductivity).

A toolkit for analysis needs to be developed and standardised to interpret and make meaningful conclusions from HF data. We need to specify when it is most valuable to incorporate insights about short-term variation and how to use multiple parameters to improve our understanding of water quality dynamics in catchments. To increase the spatial coverage of HF turbidity and water level observations would be of interest here to further relate the different mobilisation mechanisms together with the travel time of the material. Here, mapping critical source areas could also be interesting to identify and target areas with a high risk of erosion that are well hydrologically connected.

The use of HF data in water quality models must be further explored. A full exploration of how the short term variation affects model calibration is needed to find the right metrics for a fair evaluation. Potentially, mathematical descriptions of landscape and biogeochemical processes in the model need to be revised to fully assimilate the data set variation. Using HF data in scenario modelling might be a great asset as we calibrate to a more detailed temporal variation that could give an improved picture of future conditions.

Swedish hydrological conditions will change in the future due to climate change. Spring floods may come earlier, high flows may become more frequent. Relative frequencies of snow-driven versus rain-driven hydrological events might change, with potential consequences for increased winter P transport. Successful management of eutrophication in our surface waters and the Sea during a period of unprecedented change requires further improvements of our methods to understand and quantify P transport in the landscape.

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Populärvetenskaplig sammanfattning

Det övergripande syftet med vårt arbete är att bidra till levande och livskraftiga sjöar och vattendrag. Vi vill skapa vattenmiljöer som inbjudande för människor och djur att dricka, bada och använda som habitat. Det målet är inte alltid lätt att uppnå, till följd av hur vi har använt och använder vår miljö, särskilt inte när våra städer växer och fler människor behöver energi och mat.

En av de stora utmaningarna vi behöver arbeta med är att för mycket näring hamnar i våra sjöar och vattendrag, vilket leder till övergödning. Övergödning innebär att alger och vattenlevande växer får tillgång till så mycket näring att de växer obehindrat. En del alger producerar ett farligt toxin som kan skapa problem vid dricksvattenproduktion, men kan också vara skadligt för djur och människor som kommer i kontakt med vattnet. Den gröna soppan av alger skapar också en obalans i ekosystemet som gör att arter försvinner, men kan även orsaka problem som syrebrist på botten av vattendraget vilket gör att livet där dör. Ett viktigt näringsämne som orsakar en stor del av de här problemen heter fosfor.

Trots 50 år av forskning kring fosfors bidrag till övergödningen vet vi inte detaljerna kring när och hur fosfor transporteras från land till vatten. För att förstå hur vi ska arbeta för att minska fosfortransporterna behöver vi förstå (1) var fosforn kommer ifrån, (2) när och hur fosfor transporteras från land till vatten och (3) hur förändrade förhållanden (exempelvis klimatförändringar) kan påverka transporten av fosfor i landskapet.

Fyra studier har genomförts i Sävjaån med omgivning, ett låglänt område med både jordbruksmark och skog i närheten av Uppsala. I den första studien undersökte vi hur mycket fosfor som fanns lagrat i sjöarna och vattendragens sediment. Vi fann betydande mängder fosfor lagrat i sjöars sediment vilket var väntat, men att stora mängder också fanns lagrat i vattendragens sediment var ny och viktig kunskap. Fosforn i sedimenten kan vara av stor betydelse vid låga vattennivåer, då endast en liten ökning av fosfor kan ha stor betydelse för övergödningen. När vi vet att det finns stora mängder fosfor lagrade i vattendragens sediment kan vi även ta bättre beslut om exempelvis när och hur diken ska rensas.

I Sverige övervakar vi vattendrag genom att ta månadsvisa vattenprover som analyseras på laboratoriet. De månadsvisa proverna lämnar luckor, då vi inte vet vad som händer i vattendraget. Fosfor som kan vara bundet till partiklar, transporteras i stora mängder exempelvis vid regn- eller snöoväder eller när vattennivån höjs kraftigt under kort tid. För att förstå när våra fosfortransporter sker behöver vi ta prover oftare, varje dag eller flera gånger om dagen. Vi har använt ett instrument, en sensor, som monterades i Sävjaån och hämtar information om vattnet var 15:e minut dygnet runt.

Vi använde datan från sensorn för att undersöka hur mycket fosfor som transporteras under året och hur våra slutsatser påverkas av hur ofta vi provtar vattnet. Vi kom fram till att det var extra viktigt med tät provtagning under perioder med höga vattennivåer. Vi utarbetade även praktiska förhållningsregler som behövs vid användandet av sensorer, bland annat att vi behöver ha dem monterade under isen vintertid för att fånga de viktigaste perioderna under året.

I nästa studie undersökte vi bara perioder med höga flöden, för att försöka förstå varifrån fosforn och partiklar som orsakar grumlighet kommer ifrån. Vi kom fram till att när vattennivån höjs så blir ofta vattnet snabbt grumligt, vilket tyder på att partiklarna kommer ifrån närliggande områden, till exempel från flodbanken eller den bäcknära-zonen. Vi behöver därför lägga extra vikt på att behålla växtlighet på dessa områden under stora delar av året, för att minska risken för att partiklar och fosfor hamnar i vattendraget.

I den sista studien använde vi en datormodell som gör att vi kan utforska fosfortransporten under olika tidsintervall, i nutid och historiskt, vilket kan ge insyn i hur väl modellen fungerar vid framtida scenarion. Även i denna studie använde vi information från sensorn, och kom fram till hur vi behöver anpassa användandet av modellen för att kunna nyttja sensordatan på bästa sätt.

Genom detta arbete har vi bidragit till kunskap om var fosforn kommer ifrån, när och hur den transporteras i landskapet. Med hjälp av denna nya information kan vi göra små förändringar i hur vi använder landskapet för att på ett effektivt sätt minska övergödningen i våra sjöar och vattendrag.

Popular science summary

The main goal of this work was to contribute to a future where lakes and streams are in balance. We want to create water environments inviting humans and animals to drink, bathe and use as habitat. That goal is not easy to achieve due to present and historical ways of using the environment. Especially not when our cities continue to grow, and more people need energy and food.

One of the challenges that we need to work with is that too much nutrients end up in our streams and lakes, which causes eutrophication. Eutrophication means that algae and aquatic plants get access to amounts of nutrients that make them grow extensively. Some algae produce a dangerous toxin that can cause problems in drinking water treatment and harm animals and people who get in contact with the water. The "algae soup" also causes an unbalanced ecosystem that makes other species vanish and affects the oxygen levels at the bottom of the stream/lake that can cause "dead zones". A critical nutrient affecting a lot of these problems is called phosphorus.

Despite 50 years of research about how phosphorus affects eutrophication, we do not know the timing and processes transporting phosphorus from land to water. To understand how we should work to reduce phosphorus transports, we need to understand (1) where the phosphorus come from, (2) how and when phosphorus is transported from land to water and (3) how changed conditions (for example, due to climate change) can change transport of phosphorus in the landscape.

Four studies have been carried out in Sävjaån catchment, a lowland area with agricultural land and forest in central-east Sweden. In the first study, we explored how much phosphorus was stored in lake and streambed sediment. We found that large amounts of phosphorus were stored in lake sediment, which was expected. Large quantities were also held in the streambed sediment, which was new and essential knowledge. Phosphorus in sediment can affect the eutrophication in the stream at low flows, where only small increases in phosphorus can be of great importance. When we know there are large quantities of phosphorus stored in streambed sediment, we can improve our management decisions, e.g. regarding ditch cleaning practices.

In Sweden, we survey changes in water quality in streams by monthly samples, which are then analysed in the laboratory. Monthly samples leave gaps of unmonitored time when we do not know what happens in the stream. Phosphorus can be bound to particles and be transported in large quantities at heavy rain or snow or when the flow significantly increases. To understand the timing of phosphorus transport, we need to increase our number of samples, e.g. to daily or sub-daily resolution. We used an instrument, an in-situ sensor, deployed in Sävjåan and collected information every 15th minute around the clock.

We used the data from the sensor to explore how much phosphorus was transported during the year and compared it to the conclusions we would have made from monthly samples. We concluded that it is crucial to sample often during high flows. We also learned how to practically use the sensor to enable efficient use, e.g. have it deployed under the ice during the winter.

The subsequent study explored only periods with increased flow, e.g. caused by heavy rain or snowfall/snowmelt. We wanted to improve the understanding of where phosphorus and particles are coming from in the catchment. At an increase of flow, the water quickly got turbid, indicating that the particles arrived from close-by areas, such as the stream bank or land areas close to the stream. Therefore, we need to make sure that these areas are vegetated during most parts of the year.

The last study used a computer-based model to explore phosphorus transport during different time periods, present day and historically, to assess the robustness of predicting the future. Also, in this study, we used data from the sensor and learned how we need to adapt the model to use the sensor data in the best possible way.

This work increased the knowledge about where phosphorus is coming from, when and how it is transported in the landscape. With the new information retrieved, we can make minor adjustments in management to efficiently decrease the eutrophication in our streams and lakes.

Acknowledgements

Can you imagine the enthusiasm of a child walking in a forest? Every little detail is there to discover and explore. That is a pretty colour! Can I dig a hole in the ground? I would like to climb that tree!

I have been given the opportunity to keep discovering new knowledge and keep my wonder for the world– and at the same time, call it my work. During these years, I have found pretty colours, dug some holes, but also climbed a few trees. Of course there were some rainy days, but most of them were sunny and bright.

First of all, I would like to thank you **Martyn,** for inviting me to the forest to wander and wonder. Thanks for letting me choose my own path, but still letting me know that you are only an e-mail away. Thanks for everything you have taught me and for the many great discussions we have had about science. Thank you also for showing me that it is possible to be warm-hearted, empathic and down-to-earth and still be a successful researcher. I look forward to working more with you in the near future.

Jens, thanks for inviting me to the world of sensors. I am so grateful that I got to be part of the knowledge development (struggle) and building the sensor infrastructure. I will always remember when we pulled that sensor up from underneath the ice, and it was *not* broken. Thanks for always being encouraging and interested! **José,** my scientific big brother, thank you for introducing PhD-life, all the structural tips and tricks, and always having an open door (also virtual). **Elin**, thank you for your genuine interest and good advice regarding work and being a woman in science. Looking forward to work more together soon. **Lisbet**, thanks for being a rock to rely on knowing

all the practical things about a PhD-education that you need to know. Thanks for always asking tricky questions in an incredibly nice way. **Brian**, thanks for the introduction to P fractionation (and the lab), thanks for sticking up for what is important. **Salle**, thanks for being there all the way and encouraging me to quit my former job– who knows where I would have been without your support.

Oskar, thanks for being my phosphorus friend from the start and all the fun that we have had. Thanks for always just being an e-mail away! **Sara**, thanks for being my "every-day-colleague" and friend. How would home-office have been without our daily "good-morning meetings"? (unbearable) Thanks for the fun field expeditions (the one with crazy snow and the one with a bit of stream swim… unforgettable)! Always a shoulder to lean on, always a good idea and solution to a problem. I will enjoy every day that we get to keep working together, and after that every day that I get to be your friend.

I have been so happy to work at our section and have colleagues that are so inspiring, knowledgeable and friendly. Thanks for all the input and interested questions about my work: **Pia, Mike, Faruk, Karin E., Marcus, Stephan, Kevin, Reinert, Lars, Emil, Stefan H., Stefan L. Andreas, Roger, Pernilla** and **Hampus**. Thank you, **Micke** and **Göran,** for being helpful, knowledgeable and nice people to spend time with. Being in the car with you is an adventure. **Ronald, Herman** and **Mats** thank you for all the happy cheering in the corridors and always being there when I need you! Thanks to **Willem** and **Lars** for being excellent head(s) of department. **Brendan** for being warm-hearted and helpful regarding all the administrative hurdles regarding the PhD-education. **Catarina, Birgitta, Erika, Tuija**, **Dorte** thank you for keeping all the details in order. **Ingrid, Thomas, Nilo, Mirsada, Sofia, Karin, Christian** and all the rest of the personnel at the biogeochemistry lab, thank you for including me in your group and always being helpful and encouraging. Thanks to all the colleagues at the Department and in the MVM-building, for making this such a nice working place.

Jelena, thanks for being there from my first day at SLU. To have shared this journey with you has been a privilege. You have been my teacher and inspiration from day one. I love to be "Piff och Puff" with you at conferences/parties/fikas, and I am so grateful to have you as a friend. **Anna**, also there from day one, thanks for being such a warm and caring friend and colleague. Thanks for all discussions and analytical thoughts about the world (and Sweden). You are my favourite soil science person in the world! **Elsa**, I am so glad to have shared this PhD-time with you, being inspired by your stubbornness (in a positive way) and grit!

Vera, the closest I have come to a soul mate when it comes to energy and positive vibes. I am so grateful to have had you by my side during the PhD journey; all the shared laughter, a tear from time to time, sports, parties, spexes, pubs and projects. I could not have wished for a better office mate and friend. **Malin** and **Claudia**, thanks for being such awesome office mates. I have really missed you during the home-office time! **Svenja,** I knew from the moment I met you that you are someone that would bring sunshine into my life. Thank you for the time we got to spend together at SLU, being my sensor colleague and fika-friend. I am happy that we stay in contact and hope to catch the night train to you as soon as possible! **Kristina**, thanks for being such a safe and positive spirit at the beginning of my PhD-time. I am so glad to have you as a friend. You inspire me with your views on life.

The old gang: **Joachim, Alex, Jakob, Mihn, Lisa, Amélie, Didier, Pianpian, Maidul, Pablo, Mirco, Oded, Wiebke, Rikard**, **Laura**. Thanks for building up such a nice atmosphere at SLU. I admire your structured fika-culture, enjoyed all the fun parties, events and inspiring discussions. You have taught me a lot about the world of science and life!

”New gang”: **Hannah, Sophia, Baolin, Mattias, Bonnie (**thanks for the fantastic front cover – you are an artist!), **James, Frank, Jenny, Jasmina, Winnie, Carlotta.** Thanks for all the fun and laughter. Thanks for fika, parties and spexes.

“Newer gang”: **Alberto, Kong, Valentina, Pilar, Sanne, Björn, Paul, Svante, Sofia.** I look forward to getting to know you and feel happy about the engagement you show to build up the culture of the new era here at SLU.

Min familj, pappa **Torbjörn**, mamma **Annelie, Fredrik, Elisabeth, Joakim, Carro,** farmor **Inger,** mormor **Mait** och morfar **Lennart**: tack för att ni uppfostrat mig med ett kritiskt tankesätt, med diskussioner och reflektioner om natur och samhälle. Tack för stöttning under doktorandtiden, peppande ord och bra reflektioner! Ni är alla en del av pusslet, men särskilt tack till pappa för naturintresset och till mamma för ambitionen och lusten att lära!

Fanny, tack för att du är min ”person” sedan för alltid. Tack för att telefontiden är dygnet-runt-öppen och för att du tror på mig i alla lägen!

Kadir, tack för att jag får vara med dig varje dag, även om det inte alltid är lätt. Tack för att du inte begränsar min ambition men samtidigt tvingar mig att sakta ned. Tack för att du stöttade mig när jag ville sluta på ett bra jobb för att plugga mer och för att du gjort det möjligt att doktorera och ha två småbarn samtidigt (låt oss leva alla drömmar på samma gång…). Tack för allt du gör för mig, stort och smått, du är min guldkant på livet.

Cleo och Aron, tack för att ni låter mig återupptäcka skogen med nya ögon. Ni får mig att utforska och expandera min hjärnas kapacitet. Tack för att ni varje dag sprider ljus och glädje ★!

I will not leave the forest, even if this wonderful and challenging chapter of my life is over. Life is not about finding yourself, it is about creating who you are. I am proud to have given myself the opportunity to live my dream and challenge myself almost every day. Thanks!

Acta Universitatis Agriculturae Sueciae

Doctoral Thesis No. 2021:84

Eutrophication is one of the most challenging water quality issues of today, where phosphorus is a key element. This thesis provides support for improved management decisions regarding phosphorus transfer in the landscape. New insights about phosphorus stored in lake and streambed sediment can guide management strategies. With an improved understanding of short term variation provided by high-frequency monitoring, we can (1) identify critical periods for phosphorus transport, (2) practically address mobilisation processes, and (3) better parametrise our water quality models.

Emma E. Lannergård received her graduate education at the Department Aquatic Sciences and Assessment at the Swedish University of Agricultural Sciences. Her M.Sc. degree in Soil and Water Management was obtained at the same university.

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Online publication of thesis summary: http://pub.epsilon.slu.se/

ISSN 1652-6880
ISBN ((print version) 978-91-7760-845-5
ISBN (electronic version) 978-91-7760-846-2