

Long-term phosphorus trends in Swedish rivers and streams

- widespread and persistent nutrient decline

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Abstract

Phosphorus (P) is often considered the limiting nutrient for freshwater productivity because of its relevant role in regulating algal production and determining community composition. The concern for increasing anthropogenic P inputs registered since the last century has shaped environmental policies around the world to limit the effects of algal bloom (also known as eutrophication) in freshwater ecosystems through the establishment of maximum threshold nutrient concentrations. In addition to water quality impairments due to eutrophication, a recent number of studies has detected critical P declines occurring in various countries of the northern hemisphere. In Sweden, the few research works within the topic have focused on lakes minimally affected by anthropogenic disturbance, showing persistent P declines across the country. The overall aim of the present study was to determine whether analogous nutrient depletion (also known as oligotrophication) could be found in Swedish streams and rivers (n=34) with comparable conditions. Time trends analyses showed that the majority of the statistically significant (Mann-Kendall test, p value <0.05) watercourses had relevant total phosphorus (Tot-P) and phosphate (PO₄P) declining concentrations between 1980 - 2020, and the most important losses were registered for river stations situated in north Sweden. Despite the failure in identifying any correlational relationship between P trends and land use and land cover variables in the present study, further research work is needed to assess the validity of the hypothesised explanatory factors (vegetation, climate change and soil) for the ongoing oligotrophication. In conclusion, the study confirmed that a significant nutrient depletion is occurring in minimally anthropogenic disturbed catchments across Sweden accordingly to previous research works on the subject matter. In view of the obtained results and the significant evidence that oligotrophication is occurring in freshwater ecosystems of the northern hemisphere, water quality policies should complement their reference values with lower nutrient concentration thresholds to assess ecosystem health and consider possible water quality impairments that might develop from nutrient depletion.

Keywords: Boreal, Depletion, Nutrient, Oligotrophication, Phosphorus, Rivers, Streams, Water Quality

Sometimes "too little" is also a problem...

Environmental issues are often related to human activities and water quality impairments are no exception. For example, it is common to link excessive nutrient levels in rivers and/or lakes to agriculture (e.g., use of fertilizers) or urbanization (e.g., sewage systems). This is particularly true for phosphorus (P), which has a crucial role in regulating freshwater productivity and can lead to algal bloom in water when it is found in excessively high concentrations. This environmental issue, known as eutrophication, is well-known by policy makers, and for this reason we can often find monitoring measures and upper concentration thresholds for P in water quality assessment mandates.

However, a number of recent studies have reported unexpected declining P trends occurring in lakes and rivers of the northern hemisphere. These studies have reported long-term decreasing P concentrations in Canada, Sweden, Finland, Russia, and China, revealing the other side of the P-related environmental issue. When such a persistent and important nutrient depletion occurs in freshwater bodies, we talk about oligotrophication. P is an essential nutrient for many important biological functions (e.g., formation of energy molecules, cellular membranes) and for the food chain, as P is used by algae to grow and therefore by all the biota that feed on them. If oligotrophication occurs, P concentrations in water can diminish to levels where lakes and rivers can lose their vital and productive functions.

Surprisingly, widespread and persistent oligotrophication has been observed in Sweden in lakes minimally disturbed by human activities, where agriculture and urbanization account for respectively <5% and <1% of the land area. These findings have raised questions on why such a nutrient depletion is occurring (*what are the drivers?*) and the extent of the problem is (*are only lakes going through this process or is surrounding catchment also affected?*).

With the present work, we intended to explore these questions by following the hypotheses presented by Huser et al. (2018) in their study of Swedish lakes. We asked three main questions:

- 1. Can we find declining P trends (oligotrophication) in rivers in the same parts of Sweden as the lakes studied by Huser et al. (2018)?
- 2. Can we relate these P trends to land use and land cover?
- 3. What possible explanations can we give to motivate these trends?

We were able to answer to all the questions, sometimes with surprising results.

1. We found that since 1980, nearly > 90% of the minimally disturbed rivers in our study had significant declining P trends, confirming that

oligotrophication is an environmental issue affecting Sweden at country level.

- 2. We could not link the P decrease to either land use or land cover in a statistically significant way, meaning that we could not with any certainty relate oligotrophication to processes operating in the catchment.
- 3. However, we were able to identify possible explanations for the observed trends based on what the results we obtained.

The trend analysis revealed that the steepest declines (specifically P losses > 2 % per year) occurred in rivers located in the mountainous north of Sweden, consistent with trends observed earlier for lakes. In this region, the observed P declines might be related to longer growing seasons and the "greening" of the mountains, which would more efficiently retain P in growing plants.

Warmer temperatures promote forest growth and the rise of the tree line in the mountain areas, with larger and older forests representing a potential storage of P. The reduced transport of P to waterbodies could therefore be explained with a greater P retention in larger and more numerous forest trees, and this driver is consistent with the increasing forest biomass and the high proportion of old forests found in northern Sweden.

On the other hand, central and southern Sweden have been historically affected by acid deposition but have started a slow recovery since the implementation of international emissions legislation in the 1980s. A return to pre-acidification conditions could potentially explain the steep and widespread P declines observed for rivers in this region, as recovery from acidification affects soil pH and can subsequently increase soil P retention. However, the phenomenon could also be partially driven by greater plant accumulation (a higher forestry biomass is reported in the area) as well as by warmer temperatures, responsible for increasing forest growth and speeding up soil processes.

As a matter of fact, our analysis was also demonstrated that Sweden has been experiencing rising temperatures since the 1980, with important increases in the length of the frost-free seasons during autumn and spring. These results are consistent with the hypotheses previously formulated, given that shorter winter would promote forest growth and P retention by trees and other forest plants, and warmer temperatures generally accelerate soil processes.

Due to some limitations present in our study, further analyses are necessary to exactly determine the drivers behind the P declines. However, it was possible to assess the magnitude of the problem (nutrient losses are occurring at catchment scale and involve all waterbodies) and validate some of the hypotheses formulated in previous studies.

As awareness of this environmental issue is spreading and becoming more present in studies across the northern hemisphere, policy makers should complement their mandates with lower concentration thresholds for nutrient and related monitoring measures. This would enable environmental scientists, policy makers and generally people who care about the environment to consider water quality impairments brought by oligotrophication in water quality assessment procedures, because sometimes "too little" phosphorus is also a problem.

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Abbreviations

Al	Aluminium	
Ca	Calcium	
CLRTAP	Convention on Long-Range Transboundary Air Pollution	
DIP	Dissolved Inorganic Phosphorus	
DOC	Dissolved Organic Carbon	
DOM	Dissolved Organic Matter	
DOP	Dissolved Organic Phosphorus	
E-OBS	European Observational dataset	
EQGs	Environmental Quality Goals	
Fe	Iron	
GDDs	Growing Degree Days	
GFS	Gränsen för Fjällnära Skog [Near Alpine Forest]	
HELCOM	Baltic Marine Environment Protection Commission-	
	Helsinki Commission	
Ν	Nitrogen	
Р	Phosphorus	
PP	Particulate Phosphorus	
POP	Particulate Organic Phosphorus	
PO ₄ P	Orthophosphate	
S	Sulphur	
SLU	Swedish University of Agricultural Sciences	
TG	Mean Temperature	
TOC	Total Organic Carbon	
Tot-P	Total Phosphorus	
WFD	Water Frame Directive	

1. Introduction

Freshwater ecosystems are closely connected to their catchments (Hynes, 1975), and so they are excellent sentinels of physical, chemical, and biological changes occurring in the surrounding terrestrial and atmospheric environment (Williamson et al., 2009). Among the number of factors affecting freshwater productivity, phosphorus (P) is considered a limiting nutrient due to its critical role in regulating community composition through algal production (Eimers et al., 2018).

Phosphorus availability in aquatic environments has been altered by human activities during the past centuries and many studies have focused on the environmental eutrophication issue caused by increasing anthropogenic P inputs (Huser et al., 2018; Smith and Schindler, 2009). The concern about rising humaninduced P concentrations and subsequent algal bloom has shaped environmental policies around the world for reducing P exports from agricultural, industrial, and urban sources, and restoring freshwater productivity. While this goal is reflected in water quality management legislation that establishes maximum nutrient concentration thresholds for waterbodies to reach good ecological status, as for instance in the European Water Framework Directive (EUWFD, 2000), no concern is found in such regulations for when waterbodies have too little nutrients (Huser et al., 2018).

Recently, a number of studies have appeared on water quality impairments of freshwater ecosystems caused by the process of nutrient depletion known as oligotrophication. Widespread and persistent P declines have been detected in waterbodies across the northern hemisphere from Canada through Fennoscandia to China, but the assessment of the potential drivers for such trends requires further investigation in the majority of cases (Arvola et al., 2011; Crossman et al., 2016; Eimers et al., 2018; Eimers et al., 2009; Huser et al., 2020; Huser et al., 2018; Hu and Huser, 2014; Stammler et al., 2017; Tong et al. 2017).

In Sweden, researchers have focused their water quality analyses on lakes extending from the Sub Arctic (CAFF, 2019; Huser et al., 2020) to the south of the country (Huser et al., 2018; Isles et al., 2018) and observed substantial and long-term P declines across the country. Specifically, a mean trend of -2.2% y^{-1} for total phosphorus (Tot-P) between 1970 – 2015 was detected for Near Arctic lakes by Huser et al. (2020), similar to the average -2.5% y^{-1} Tot-P found for 42 out 81 lakes between 1988 – 2013 by Huser et al. (2018). In the latter study, the trophic

status was also analysed, resulting in a preponderant shift to lower trophic classes for 24 lakes, of which 18 became ultra-oligotrophic (Huser et al., 2018).

The hypothesized explanatory factors for the ongoing oligotrophication of these lakes subject to minimal anthropogenic disturbance are various and diverse for the different areas of Sweden. For Tot-P declines observed in southern and central Sweden, a return to pre-acidification conditions is a plausible driver, as recovery from acidification affects soil pH and can subsequently increase soil P retention (Crossman et al., 2016; Gérard et al., 2016, Huser et al., 2018) and dissolved organic carbon (DOC) concentrations (Huser et al., 2020; Monteith et al., 2017). Huser et al. (2018) found a positive correlation between increased DOC concentrations and Tot-P trends in their study, hypothesizing that the process of P weathering in the soil, enhanced by warmer temperatures, and subsequent major external inputs of dissolved organic P might be masked by in-catchment processes such as increased nutrient uptake by terrestrial vegetation.

Differently, decreasing Tot-P in northern and alpine areas of Sweden might find explanations in changing climate and subsequent effects on forest biomass (Huser et al., 2018; Huser et al., 2020). Warmer temperatures and consequent heat accumulation, often estimated with the Growing Degree Day (GDD) indicator, lead to longer growing seasons, growing forest biomass and, thus, increased terrestrial vegetation nutrients uptake (Huser et al., 2018). Other studies have highlighted the risk of P limitation associated with growing forest biomass harvesting (Akselsson et al., 2008) and suggested that forest change might explain nitrate decline in rivers and streams (Lucas et al., 2016).

To study if long-term P variation in rivers and streams could contribute to assessing the scale of nutrient depletion and stimulate research within the topic for better understanding the catchment processes behind P declines in waterbodies of Sweden and, more broadly, the northern hemisphere. Ultimately, this work could strengthen the necessity to establish more comprehensive water quality criteria (e.g., upper and lower concentration thresholds) for waterbodies to have good ecological status, and pave the way for introducing specific management practices to restore the productivity of freshwater ecosystems.

With this project a long-term (1980 - 2020; 40 years) analysis of Tot-P and orthophosphate (PO₄P) concentrations for 34 Swedish rivers and streams characterised by minimal anthropogenic disturbance is presented. The research questions guiding the work are:

- 4. What trends can be observed for Tot-P and PO₄P concentrations in the selected (n=34) minimally disturbed rivers and streams across Sweden for the period 1980 2020?
- 5. What correlations exist between the resulting Tot-P and PO₄P concentration trends and land cover and use of the rivers' watersheds?

6. Which explanatory mechanisms may account for the detected Tot-P and PO₄P concentration trends for the rivers and streams of the study?

By answering these questions, this project will determine whether analogous P declines previously observed in Swedish lakes subject to minimal anthropogenic disturbance (Huser et al., 2018; Huser et al., 2020) can be found in Swedish streams and rivers with comparable conditions. In addition to the study of the Tot-P and PO₄P concentrations over the considered period, the work intends to explore the potential existing correlation between the detected P trends and land use and cover variables. The assessment of such a relationship might help determine the potential drivers of P concentrations for the studied Swedish waterbodies, and, thus, validate the hypothesised explanatory factors from previous literature.

2. Background

This chapter provides a general background to the main topics discussed in the present work. The following sections contain relevant information respectively on Swedish geology, climate (past and future) and land use, the role of P as Tot-P and PO₄P in freshwater ecosystems, and P behaviour in the context of historical acid deposition and recovery.

2.1. Sweden: geology, climate, and land use

Sweden belongs to the Fennoscandian Shield, a region constituted of a crystalline and metamorphic bedrock including gneiss, granite, sandstone, and marble (SGU, 2020). As a result of the numerous dramatic geological events and particularly because of the consecutive glaciation and deglaciation periods, the majority of the landscape is covered by till, a type of soil primarily made of non-sorted sediments (SGU, 2020).

Most of the country is characterised by a cold temperate climate with the southern coastal areas distinguished by a warm temperate one (SMHI, 2021a). The average temperature is subject to strong fluctuations depending on the side of the polar front zone, but it usually follows a declining gradient from north to south and from west to east, with colder air found in valleys in winter and on mountain peaks in summer (Figure 1a) (SMHI, 2021a). Precipitation (500-800 mm y^{-1} for 1960 – 1990) falls in every season and it is concentrated in summer and autumn as rain, whereas snow is common from late autumn to early spring for 6-8 months in the northern mountains and occurs more sporadically on the southern coasts (SMHI, 2021a). The wettest regions are located in the mountain areas close to the border with Norway (1500-1200 mm y^{-1} for 1960 – 1990) and in the southwestern parts of the country (1000-1200 mm y^{-1} for 1960 – 1990) because of the movement of low-pressure air masses, while the lowest precipitation (<400 mm y^{-1} for 1960 – 1990) is found on the small islands along the Swedish east coast and in some confined mountain valleys (Figure 1b) (SMHI, 2021a).

The climate of Sweden is currently facing important alterations due to the effects of climate change. Comparing the values of the normal period 1960 - 1990 with those from the 1991 - 2018 record, it is evident that the median temperature

has become warmer by about 1.0 °C overall (1.5 °C in the mountain areas of Lapland) (Figure 2a) (SMHI, 2021b). Central and north Sweden have experiences higher temperatures by >2 °C in winter, while less changes are occurring in autumn for the southwest of the country. Increases (10% wetter) have also been observed for precipitation in all seasons but autumn with some exceptions on the eastern coast where slight declines have been recorded (Figure 2b) (SMHI, 2021b).



Figure 1. Climate of Sweden for the normal period 1960-1990. Panel (a) shows the gradient of temperature and panel (b) displays the precipitation pattern across the country. Retrieved and adapted from SMHI (2021a).

Approximately 69% (27.9 million ha) of the total surface area (40.7 million ha) is covered by forest land, of which nearly 85% is used for productive purposes (Figure 3a, 3b) (SLU, 2021). The majority of productive forests (almost 42%) are located in the northern regions of Norrbotten, Västerbotten and Jämtland, together with mires, subalpine woodlands and high mountains (SLU, 2021). The percent forest cover gradually diminishes from centre to south of Sweden, where most of the pasture and arable land as well as urban areas are found (SLU, 2021). Inland water constitutes roughly 10% (4 million ha) of the total surface area (SCB, 2019) with almost 103 000 lakes (of which 30% are in Norrbotten) (SMHI, 2021c) and more



than 50 000 km of rivers and streams, running in Lapland for 20% (SLU, 2020).

Figure 3. Magnitude of climate change in Sweden. In panel (a) the annual variation in temperature where red bars indicate temperatures above the average of the normal period 1960-1990; in panel (b) the annual average precipitation where the black line shows the assessed average for ten years. Retrieved and adapted from SMHI (2021).



Figure 2. Magnitude of climate change in Sweden. In panel (a) the annual variation in temperature where red bars indicate temperatures above the average of the normal period 1960-1990; in panel (b) the annual average precipitation where the black line shows the assessed average for ten years. Retrieved and adapted from SMHI (2021)

2.2. The role of P in freshwater ecosystems

Productivity of freshwater ecosystems can be affected by several factors and among the limiting elements, P has a critical role in regulating the pace of production for aquatic plants and, ultimately, the community composition of ecosystems themselves (Eimers et al., 2018). Phosphorus is used by living organisms for the biosynthesis of genetic material (phosphates), transfer of energy (ATP) and constitution of cellular membranes (phospholipids) and it is therefore an essential nutrient for biota (Ruttenberg, 2014).

Phosphorus can be present in water in several forms with different operational definitions, but it can be broadly classified as particulate or dissolved according to the particle size, and it can be further distinguished between inorganic and organic forms (Yoshimura et al., 2007). Tot-P accounts for all these types of P, while PO₄P is included among the DIP (Dissolved Inorganic Phosphorus) forms. Availability of P in terrestrial environments (and, therefore, its export to freshwater ecosystems) is regulated by adsorption and precipitation processes between P and particles (e.g., base cations, aluminium, iron), and so the sorption mechanisms depend on variables such as pH, cation species and reaction time (Brady and Weil, 2008; Morgan, 1997). For instance, PO₄P is generally increasingly absorbed to aluminium (Al) and iron (Fe) oxides the more the pH decreases, but it precipitates together with Al and Fe cations if PO₄P is highly available at low pH or alternatively with calcium (Ca) at high pH (Gustafsson et al., 2012).

When analysing and assessing water quality, PO₄P is relevant for freshwater ecosystems as it gets directly assimilated by algae for biomass production (Ruttenberg, 2014). However, it can also be relevant to include Tot-P concentration in the analysis to account for the P forms that could potentially become bioavailable through the P cycle, and, thus, obtain a comprehensive understanding of the P content in the ecosystem. Despite its bioavailability, PO₄P usually constitutes a low fraction of Tot-P (Wetzel, 2001) and it is generally more abundant in Sweden in environments disturbed by anthropogenic activity, especially where agriculture accounts for more than half of the land use (Ulén & Jakobsson, 2005). Thus, to incorporate Tot-P in the water quality assessment becomes consistent as in some cases, \geq 70% of Tot-P is present in the natural environment as POP (Particulate Organic Phosphorus) and, so, as P bound in organic molecules of living and dead organisms (Yoshimura et al., 2007). Particulate P (PP) can contribute 6-10% of the immediately bioavailable P fraction (Uusitalo et al., 2003) and it is therefore an important source of directly usable P, specifically when found in runoff and drainage water from clay soils typically present in the central plains of Sweden (Ulén & Jakobsson, 2005; Uusitalo et al., 2003).

For these reasons, and because PP is more easily estimated than dissolved P using proxies such as turbidity (Jones et al., 2011), Tot-P is used in water quality analysis and assessment to estimate the productivity of freshwater ecosystems, not

to mention that Tot-P takes account for all the various forms P might cycle between (Jones et al., 2011).

2.3. Phosphorus and acidification in Sweden

Since the beginning of the 1920s, deposition of sulphur (S), nitrogen (N) has caused the acidification of a large number of Swedish waterbodies (Almer et al., 1974; Moldan et al., 2013) and caused severe ecological disturbances (Moldan et al., 2013; Tammi et al., 2003). With the establishment of international agreements in the 1980s, most importantly the Convention on Long-Range Transboundary Air Pollution (CLRTAP) come into effect in 1983 (UNECE, 2012), emissions of S and N have been significantly reduced with consequent notable positive effects on Fennoscandian rivers and streams (Moldan et al., 2013; Skjelkvåle et al. 2001; Wilander and Fölster, 2007). The most recent protocol (Gothenburg Protocol), discussed in 1999 and amended in 2012, welcomed new countries among its parties and set diverse international thresholds for the emission of polluting compounds until 2020 (UNECE, 2012). The same deadline was shared by the 16 Environmental Quality Goals (EQGs) adopted by Sweden in 1999, among which the target of "Natural Acidification Only" was of particular importance (SEPA, 2018).

Thanks to international and national efforts, Swedish waterbodies have started a process of recovery from acidification since the peak of 1985 (Moldan et al., 2013). As Moldan et al. (2013) observed, progress towards the pre-industrial conditions have been slower for lakes and streams compared to the decline of S deposition, addressing the lag time to a number of factors such as soil base cations replacement and their increased removal with forestry activity. Recovery from acidification is expected to continue in the future in accordance with the international policies on the subject matter; specifically, the model used by Moldan et al. (2013) predicted a further decrease in the number of anthropogenically acidified lakes ($\Delta pH > 0.4$ between 2005 – 2010) were located in the south-western and south-central regions of Sweden, but scattered and consistent pH declines were also found close to the mountain areas and on the north-eastern coast.

Recovery from acidification can alter P concentrations in terrestrial environments (soil water solution) with consequences in freshwater ecosystems. Despite the reduction of S and N emissions and the positive effects on Swedish lakes, the number of catchments presenting base cation-depleted soils is still increasing (Moldan et al., 2013). Base cations and P usually enter the soil through atmospheric deposition and mineral weathering (the process is faster in younger soils as those in Sweden) (Moldan et al., 2013; Stoddard et al., 2016; Tipping et al., 2014; Wang et al., 2015) and leave it with leaching processes and uptake mechanisms of growing vegetation; thus, the variation of soil base cations storage content can be used to assess the rate of acidification of terrestrial ecosystems (Moldan et al., 2013). As many Swedish soils continue acidifying (making them an important source of Al and Fe released in the soil water solution) and P solubility is dependent on pH with different mechanisms under various soil conditions and Al/Fe and base cations concentrations (Penn and Camberato, 2019; Huser et al., 2018), this process can affect the P concentration in the soil solution with either a positive or negative effect on external P loads and, ultimately, Tot-P surface water content (Huser et al., 2018; Huser and Rydin, 2005). Generally, potentially high external P loads from soil water solution of the catchment might result from increased dissolved organic matter (DOM) fluxes (attributed to reduced acidic deposition in previous studies) that contain dissolved organic P (DOP) and associate with Al and Fe in the soil to form soluble complexes of DOM-Fe(Al)phosphate (Huser et al., 2018; Kopáček et al., 2015; Monteith et al., 2007). On the contrary, recovery from acidification can be linked to reducing Al concentrations both in soil and surface water where soils are returning to their pre-acidification conditions (Huser et al., 2018; Huser and Rydin, 2005). This condition would increase the binding between P and Al/Fe (oxy)hydroxide metals and clay minerals (Gérard, 2016; Gustafsson et al., 2012), limiting P mobility in soil and reducing its external load from watershed to surface water (Huser et al., 2018). It can be deduced from the above-mentioned studies that pre-existing conditions of terrestrial and aquatic ecosystems (Δ pH, base cations storage, contents of P, DOM, Al/Fe...) play an important role in understanding which effects recovery from acidification might lead to regarding external P loads and Tot-P content, but it is also evident that the complexity of P cycling in acidified environments can be a limiting factor when predicting how the P concentration will change in surface waters recovering from acidification.

3. Material and Methods

The chapter presents the general relevant characteristics of the watercourses considered in the study and the methodology followed for retrieving, handling and analysing the data. Likewise, specific information regarding types of software used for data treatments and their sources are reported to ensure the reproducibility of the work.

3.1. Rivers and streams information

The study was conducted on 34 Swedish rivers and streams included in the Swedish National Monitoring Program (Fölster et al., 2014) and extending from 56.19° N to 65.87° N and 12.50° to 24.13° E (Figure 4). Since the selected watercourses cover almost the entire latitudinal length of the country (n= 21 in Norrland, n= 7 in Svealand and n= 6 in Götaland), they encompass a wide variety of climates and flora, ranging from climatic region 1 to 4 (Pechlivanidis et al., 2018) and from northern boreal to nemoral vegetation zone (Stein et al., 2021). Despite the longitudinal extent, most of the riverine stations (n=20) are located along the eastern coastal area of Sweden, while only one is situated on the western coast, and the rest are spread across the inland of the country. Because they are found in distinct Swedish ecoregions, the rivers and streams of the study present different proportions of land use and cover; however, all the watercourses fit the profile of being subject to minimal anthropogenic disturbance.



Figure 4. Map of Sweden with selected 34 stations retrieved from www.miljodata.slu.se.

3.2. Laboratory methods, data retrieval and handling

Analyses on water chemistry were performed by the staff of the Geochemistry laboratory at the Department of Aquatic Sciences and Assessment at the Swedish University of Agricultural Sciences (SLU). The laboratory uses methods accredited by SWEDAC and further information on current operational treatments can be found on the website <u>https://www.slu.se/en/departments/aquatic-sciences-assessment/laboratories/vattenlabb2/</u>.

Temporal P variation for Swedish rivers and streams subject to minimal anthropogenic disturbance was quantified by examining long-term (1980 – 2020; 40 years) Tot-P and PO₄P concentration trends for respective watercourses. The data set, originally including 90 rivers, was retrieved from the database of the Swedish National Monitoring Program (Folster et al., 2014) available at <u>https://miljodata.slu.se/MVM/</u>. For each river station, total watershed area and proportions of land use and cover (namely, agriculture, open water, forest on

mineral soil, forest on wetland, other, urbanisation, wetland) in addition to the available annual mean of Tot-P and PO₄P concentrations for the period 1980 - 2020 (Table 1) were obtained from the Department of Aquatic Sciences and Assessment at SLU (Appendix B). A preliminary cleaning of the data set was performed in Microsoft Excel and subsequent analyses were completed using RStudio 1.4.1717 (RStudio Team, 2021).

The original data set (n=90) was filtered using the software Microsoft Excel to select rivers and streams meeting the requirement of minimal anthropogenic disturbance. The selection criterion was defined as watercourses minimally affected by agriculture (<5% of watershed area) and urbanisation (<1% of watershed area) consistently with the values adopted in Huser et al. (2018). The preliminary cleaning provided 34 stations meeting the minimal anthropogenic disturbance requirement.

Variable	Unit
Tot-P	$\mu g L^{-1}$
PO ₄ P	$\mu g L^{-1}$
Total watershed area	km ²
Agriculture	% total watershed area
Open water	% total watershed area
Forest on mineral soil	% total watershed area
Forest on wetland	% total watershed area
Other	% total watershed area
Urbanisation	% total watershed area
Wetland	% total watershed area

Table 1. Retrieved variables and respective units for the riverine stations used in the project.

3.3. Data quality control and statistical analyses

RStudio 1.4.1717 (RStudio Team, 2021) was used to control the quality of the retrieved time records and perform the statistical analyses. The number of missing data was calculated for each station and rivers having \geq 5 years of missing data were removed to ensure a more complete coverage of the period 1980-2020, and avoid distinct starting and ending periods of record, reducing the final set to 34 stations (Appendix A). The time series were checked for outliers by [log] _10 transforming all Tot-P and PO₄P concentrations and excluding from the final dataset the values identified by \pm 3 standard deviations from the mean of the station's values.

In order to study the variation of Tot-P and PO₄P over the considered 40 years, the Mann-Kendall test (Kendall, 1975; Mann, 1945) was applied to the time series to determine significant monotonic trends for each station, where statistical

significance was defined at a p-value level of <0.05. The Mann-Kendall is a nonparametric and distribution-free test, representing the observations by their ranks and requiring no assumptions on the normal distribution of the data sample (Helsel et al., 2020), however, absence of serial correlation between variables and constant spread of the distribution are necessary to obtain correct p-values (Helsel and Hirsh, 1992). In the Mann-Kendall, the significance of the Kendall's rank correlation coefficient (τ) of the y variable is tested against time (x variable) to examine whether the y values are subject to a monotonic change (increase or decrease) with time (Helsel and Hirsh, 1992). To assess such a temporal trend, paired y and x data are firstly ordered by increasing time and correlation between paired y and x values estimated as a result of the following cases:

- positive correlation when *y* values are increasing more frequently with time (increasing *x* values) than decreasing;
- negative correlation when *y* values are decreasing more frequently with time (increasing *x* values) than increasing;
- no correlation when y values are increasing and decreasing approximately the same number of times (Helsel et al., 2020).

Secondly, the monotonic dependence of y and x values is calculated by difference between the number of concordant (P) and discordant (M) pairs, resulting in the Kendall's score (S) [1]:

$$S = P - M \quad [1]$$

where *P* indicates the number of paired observations having the same sign for the difference between *y* and *x* values, and *M* quantifies the number of those having opposite sign (Helsel et al., 2020). Kendall's τ measures the strength of the correlation existing between two variables and it is calculated dividing *S* by $n \cdot (n - 1)/2$, the number of possible comparisons between *n* data pairs [2] (Helsel et al., 2020).

$$\tau = \frac{S}{n \cdot (n-1)/2} \quad [2]$$

The Kendall's τ obtained has therefore a value $-1 \le \tau \le +1$, depending on whether all the *y* values decrease with the increase of *x* values ($\tau = -1$) or increase in concert with *x* values ($\tau = +1$) (Helsel et al., 2020). Finally, significance of Kendall's τ is tested and provided through calculation of the p-value on a two-sided test to determine whether the null hypothesis (H₀ $\tau=0$) should be rejected and alternative hypothesis (H₁ $\tau\neq 0$) validated (Helsel et al., 2020). *S* is commonly standardised with a large sample size (n>10), given the approximation to a normal distribution, and the standardised statistic test is applied to Kendall's τ to obtain the p-value (Helsel et al., 2020). If the p-value exceeds the established significance level, H₀ τ =0 is rejected, and a significant monotonic trend identified.

In this project, the function Kendall in the R package "Kendall" (McLeod, 2011) was used to perform the Mann-Kendall test on the Tot-P and PO₄P concentrations (*y* variables) over the period 1980-2020 (*x* variable) for the selected 34 stations. The test was computed on two numeric vectors *x* and *y* for each station, returning the following list (Table 2):

Table 2. Components of the class Kendall list returned by the Kendall package (McLeod, 2011).

Unit	Component name
tau	Kendall's tau statistic
sl	Two-sided p-value
S	Kendall's score
D	Denominator (equal to $n \cdot (n-1)/2$)
varS	Variance of S

Subsequently, the Theil-Sen's line (Sen, 1968; Theil, 1950) was calculated to estimate the magnitude (unit y^{-1}) of the trends of the assessed Tot-P and PO₄P concentration variations. The Theil-Sen is a nonparametric test used to determine the intercept (b_0) and the slope (b_1) for estimating the median of a y variable given an x variable (Helsel et al., 2020) as in the linear equation [3]:

$$\bar{y} = \overline{b_0} + \overline{b_1} \cdot x \quad [3]$$

Firstly, each data pair (x,y) of the time series is compared to all the other data pairs for a $n \cdot (n-1)/2$ number of pairwise comparisons to estimate individual slopes $(b_1 = \Delta y/\Delta x)$ for every x and y observation, and so the overall median is calculated to obtain $\overline{b_1}$ (Helsel et al., 2020). Secondly, $\overline{b_1}$ is subtracted from the median of y and multiplied by the median of x to guarantee the intercept of the Theil-Sen's line through the medians of x and y, resulting in $\overline{b_0}$ (Helsel et al., 2020). b_1 is tested for significance similarly to Kendall's τ (Ho $b_1 = 0$) and $\overline{b_1}$ is concordant with Kendall's S (Helsel et al., 2020).

The function sens.slope in the R package "trend" (Pohlert, 2020) was used in the work to compute the Theil-Sen's slope of the assessed Tot-P and PO₄P trends resulting from the Mann-Kendall test. The Theil-Sen test was applied to the individual time series with a confidence level = 0.95, resulting in the following list of components (Table 3):

Abbreviation Component name	
estimates	Sen's slope as numeric vector
data.name	Character string denoting input data
p.value	p-value
statistic	z quantile of standard normal distribution
null.value	Null hypothesis H ₀ $b_1 = 0$
conf.int	Upper and lower confidence interval
alternative	Alternative hypothesis H ₁ $b_1 \neq 0$
method	Character string denoting the test

Table 3. Components of the class htest returned by the trend package (Pohlert, 2020).

To estimate the annual rate of change (% y^{-1}) of the statistically significant trends, the Theil-Sen's slope of each watercourse was divided by the respective median Tot-P and PO₄P concentration values.

For each station, the relative trend was compared with the different retrieved variables to study the potential existing correlational relationship between Tot-P and PO₄P concentrations and land use and coordinates. The trend-parameter relationships were obtained by plotting the assessed concentration tendencies against the selected land use classes and coordinates, and visual inspection and evaluation were carried out by looking at the computed charts.

3.4. Calculation of growing degree days

For each river station, the number of growing degree days (GDDs) was calculated using the Panoply 4.12.11 (NASA, 2021) software to retrieve average temperature data (° C). The analysis of GDDs, where a GDD is defined as day with temperature > 5° C, was performed to assess variations in the length of frost-free seasons, particularly autumn and spring, and explore the validity of climate change and derived effects as a potential driver for Δ P in Swedish freshwater ecosystems as hypothesised in previous studies (Huser et al., 2018; Huser et al., 2020).

A European observational (E-OBS) dataset containing daily mean temperatures (TG) for the period 01/01/1950 - 31/12/2020 was downloaded on a 0.25° available regular grid using the 23.1e version at https://surfobs.climate.copernicus.eu/dataaccess/access eobs.php (Cornes et al., 2018). Firstly, coordinates of rivers and streams were transformed from SWEREF99 TM to WGS84 Decimal system using the coordinates conversion tool from SLU Fältforsk available at the website https://www.slu.se/fakulteter/nj/omfakulteten/centrumbildningar-och-storre-forskningsplattformar/faltforsk/ utbildning-och-teknik/dokumentation/kartkoordinater/ "Kartvisning och konvertering" (Map view and conversion). Secondly, the retrieved E-OBS dataset was opened in Panoply and the mean temperature variable was selected to create a line plot using time (days) for the vertical axis. Finally, the converted coordinates (WGS84 format) were compared with the coordinates range $(201^{\circ}N - 464^{\circ} E)$ available in Panoply 4.12.11, and closest values were chosen to obtain daily mean temperatures for the selected locations (Appendix C). Daily mean temperatures were available for all 34 stations. Retrieved array data were filtered in Microsoft Excel and only temperatures $\geq 5^{\circ}$ C were used for the sum of yearly GDDs, while graphs were obtained in R Studio 1.4.1717 to display the average daily temperature trend for the selected stations over the period 1950 – 2020 (Appendix F).

4. Results

The results obtained from the previously described analyses are presented in this chapter in accordance with the research questions guiding the work. The following sections report a general overview on the findings for P concentration trends of the 34 considered rivers during 1980 – 2020, and the individual analyses of PO₄P and Tot-P trends with the presentation of the most relevant cases. The two last sections explore the potential correlation between the produced trends and retrieved variables, and examine the results obtained in the study regarding the variation of GDDs for 1950 – 2020.

4.1. Overview on findings for PO₄P and Tot-P

A preliminary general examination of the results provided that the considered rivers and streams (n = 34) presented different median PO₄P and Tot-P concentrations as a reflection of the environmental and climatic heterogeneity they are subject to. The median PO₄P concentration ranged from 1.25 $\mu g L^{-1}$ of the northern river *Skellefte älv Slagnäs* in Norrbotten to 18.42 $\mu g L^{-1}$ of *Sävjaån Ingvasta* in Uppsala (Table 4). Likewise, the same rivers had the lower and upper limits for the median Tot-P concentration interval, which varied between 4.92 $\mu g L^{-1}$ (*Skellefte älv Slagnäs*) and 52.92 $\mu g L^{-1}$ (*Sävjaån Ingvasta*) (Table 4).

The performed analyses showed that the majority of the watercourses in the study presented declining PO₄P (n = 9) and Tot-P (n = 30) concentrations over the period 1980 – 2020 for the statistically significant (Mann-Kendall test, p value <0.05) detected trends (Table 5 and 7). For both the parameters, the stations *Ljungan Skallböleforsen* (north-eastern coast) and *Skellefte älv Slagnäs* (centrenorth) were among the rivers with the largest significant (Mann-Kendall test, p value <0.05) relative annual declines for the study period, registering respectively -2.36 % y^{-1} PO₄P and -3.11 % y^{-1} Tot-P, and -2.00 % y^{-1} PO₄P and -3.05 % y^{-1} Tot-P between 1980 – 2020 (Table 6 and 8).

No statistically relevant potentially existing correlations between Tot-P and PO₄P concentrations and other retrieved parameters resulted from the comparison of the assessed Theil-Sen's lines (unit y^{-1}) with the data on land use and cover, and coordinates.

River station	Median PO₄P	Median Tot-P
Alterälven Norrfjärden	6.75	27.72
Ammerån Skyttmon	1.75	7.20
Ångermanälven Sollefteå	1.88	7.75
Dalälven Älvkarleby	2.96	14.92
Emån Emsfors	3.46	18.17
Forsmarksån Johannisfors	4.17	19.87
Gide älv Gideåbacka	3.18	14.42
Gullspångsälv. Gullspång	3.00	11.08
Indalsälven Bergeforsen	1.50	6.17
Indalsälven Hammarstrand	1.40	6.17
Kalix älv Karlsborg	4.45	16.35
Klarälven Almar	2.75	11.75
Klarälven Edsforsen	2.50	10.46
Klarälven Norra Råda	2.65	12.75
Ljungan Skallböleforsen	2.40	10.23
Ljusnan Funäsdalen	1.75	6.53
Ljusnan, Ljusne Strömmar	2.95	11.10
Lögde älv Lögdeå	4.11	19.75
Lule älv Luleå	1.83	7.30
Lyckebyån Lyckeby	4.91	26.25
Nissan Halmstad	6.08	25.42
Öre älv Torrböle	4.00	18.00
Pite älv Bölebyn	2.92	11.52
Råne älv Niemisel	3.50	13.75
Rickleån Robertsfors	3.83	14.50
Sävjaån Ingvasta	18.42	52.92
Skellefte älv Slagnäs	1.25	4.92
Skellefte älv, Kvistforsen	1.83	6.86
Svedån Sved	3.67	11.63
Töre älv Infl. Bölträsket	7.83	29.55
Torne älv Mattila	4.45	18.17
Upperudsälv. Köpmannebro	2.75	11.00
V. Dalälven Mockfjärd	2.50	10.67
Vindelälven Maltbrännan	1.70	6.75

Table 4. Median PO₄P (μ g L⁻¹) and Tot-P (μ g L⁻¹) concentrations of the studied watercourses (n=34) for the period 1980 – 2020.

4.2. PO₄P time trends

Analysis of time trends showed that circa 26% of the studied rivers and streams presented statistically significant (Mann-Kendall test, p value <0.05) declining PO₄P trends for the period 1980 – 2020 (Table 5). Despite most of the watercourses in the study followed the general negative tendency, 1 station was found showing a significant (Mann-Kendall test, p value <0.05) positive trend for the study period (Table 5), specifically *Lögde älv Lögdeå* situated in the north-eastern coastal area of Sweden (Table 6, Figure 6d).

Table 5. Summary of the assessed PO_4P trends for the all the studied watercourses (n=34) over the period 1980 - 2020.

Trend	Number of stations
Positive (significant and non-significant)	12
Negative (significant and non-significant)	22
Significant (positive and negative)	10
Significant positive	1
Significant negative	9

For these rivers (n=10) the Theil-Sen's slope was estimated as percentage of the median value (trend/median Tot-P, % y^{-1}) to assess the magnitude of the annual rate of change, which ranged from +1.71 to -2.36 % y^{-1} and averaged -1.41 % y^{-1} (-1.75 % y^{-1} when only the negative trends were considered) (Table 6). It was evident from the comparison between the relative yearly change and the median value that stations showing negative trends (n = 9) had the lowest median PO₄P concentrations ($\leq 3.67 \ \mu g \ L^{-1}$) for the period 1980 – 2020, in contrast with the only positive trend station (*Lögde älv Lögdeå*) that presented a higher median PO₄P content (4.11 $\mu g \ L^{-1}$) (Figure 5).



Figure 5. Relative annual PO₄P rate of change versus median PO₄P concentration for the statistically significant (Mann-Kendall test, p value <0.05) watercourses (n=10) over the period 1980 – 2020.

Table 6. Summary table of the annual PO_4P rate of change (% y^{-1}) for the statistically significant (Mann-Kendall test, p value <0.05) stations (n=10) for the period 1980 – 2020.

River station	Δ PO ₄ P
Gullspångsälv. Gullspång	-2.00
Indalsälven Bergeforsen	-1.62
Indalsälven Hammarstrand	-1.62
Ljungan Skallböleforsen	-2.36
Lögde älv Lögdeå	+1.71
Lule älv Luleå	-1.52
Skellefte älv Slagnäs	-2.00
Skellefte älv, Kvistforsen	-1.76
Svedån Sved	-1.31
Upperudsälv.Köpmannebro	-1.60

At individual level, the stations that displayed the highest annual PO₄P declines (\geq 2.00 % y⁻¹) for the period 1980 – 2020 and, thus, the steepest Theil-Sen's slopes

were Ljungan Skallböleforsen (-2.36 % y^{-1}) (Figure 6a), Skellefte älv Slagnäs (-2.00 % y^{-1}) (Figure 6b) and Gullspångsälv. Gullspång (-2.00 % y^{-1}) (Figure 6c), respectively located on the northeastern coast and by the lakes Nastajaure in the centre-north and Skagen in centre-south of Sweden. Only Lögde älv Lögdeå (northeast coast) presented a substantial increase of PO₄P concentration (+1.71 % y^{-1}) over the period 1980 – 2020, but despite the postive trend, the time trend analysis provided that since 2015 the PO₄P content has dropped down significantly (Figure 6d).



Figure 6. PO₄P concentration declines and increase ($\mu g L^{-1}$) for the riverine stations Ljungan Skallböleforsen (a), Skellefte älv Slagnäs (b), Gullspångsälv. Gullspång (c) and Lögde älv Lögdeå (d) over the period 1980-2020

4.3. Tot-P time trends

Results from the analyses indicated that 97% of the watercourses in the study had statistically significant (Mann-Kendall test, p value <0.05) declining Tot-P trends between 1980 and 2020, while no statistically significant positive trends were detected for the same period (Table 7).

Table 7. Summary of the assessed Tot-P trends for all the studied watercourses (n=34) over the period 1980-2020.

Trend	Number of stations
Positive (significant and non-significant)	1
Negative (significant and non-significant)	33
Significant (positive and negative)	30
Significant positive	0
Significant negative	30

For the significant (Mann-Kendall test, p value <0.05) tendencies, the Theil-Sen's slope was calculated, and the median annual rate of change (trend/median Tot-P, $\% y^{-1}$) was later estimated. The assessed Tot-P declines were not trivial as the magnitude of decrease ranged from -3.17 to -0.53 $\% y^{-1}$, with an average yearly rate of change of >1.8 $\% y^{-1}$ (Table 8). It was observed that rivers and streams already presenting low Tot-P concentrations when the monitoring activity began showed the largest relative annual declines, as revealed from comparison of the relative annual Δ Tot-P with the median Tot-P concentration (Figure 7).

Table 8. Annual rate of change (% y^{-1}) of Tot-P for the studied watercourses (n=30) in the period 1980-2020.

River station	Δ Tot-P
Alterälven Norrfjärden	Not significant
Ammerån Skyttmon	-2.45
Ångermanälven Sollefteå	-1.98
Dalälven Älvkarleby	-1.12
Emån Emsfors	-0.85
Forsmarksån Johannisfors	-0.53
Gide älv Gideåbacka	-1.42
Gullspångsälv. Gullspång	-1.38
Indalsälven Bergeforsen	-2.82
Indalsälven Hammarstrand	-2.53
Kalix älv Karlsborg	-1.11
Klarälven Almar	-1.68
Klarälven Edsforsen	-1.93
Klarälven Norra Råda	-1.11
Ljungan Skallböleforsen	-3.11
Ljusnan Funäsdalen	-3.17
Ljusnan, Ljusne Strömmar	-2.48
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Lögde älv Lögdeå	Not significant
Lule älv Luleå	-2.28
Lyckebyån Lyckeby	Not significant
Nissan Halmstad	-1.41
Öre älv Torrböle	-0.95
Pite älv Bölebyn	-1.38
Råne älv Niemisel	-1.52
Rickleån Robertsfors	-1.50
Sävjaån Ingvasta	Not significant
Skellefte älv Slagnäs	-3.05
Skellefte älv, Kvistforsen	-2.99
Svedån Sved	-1.91
Töre älv Infl. Bölträsket	-1.60
Torne älv Mattila	-1.21
Upperudsälv.Köpmannebro	-1.44
V. Dalälven Mockfjärd	-1.48
Vindelälven Maltbrännan	-2.86



Figure 7. Relative annual Tot-P rate of change versus median Tot-P concentration for the statistically significant (Mann-Kendall test, p value <0.05) studied watercourses (n=30) over the period 1980 – 2020.

At individual level, the steepest Theil-Sen's slopes and largest relative annual Tot-P declines (>3.00 % y^{-1}) were found at the northern mountain river station *Ljusnan Funäsdalen* in Jämtland (-3.17 % y^{-1}) (Figure 8a), followed by *Ljungan* Skallböleforsen on the north-eastern coast in Sundsvall (-3.11 % y^{-1}) (Figure 8b) and Skellefte älv Slagnäs in the north central Norrbotten (-3.05 % y^{-1}) (Figure 8c). For the latter watercourse, also the station Skellefte älv, Kvistforsen situated on the coast at the river mouth displayed an important Tot-P rate of decrease (2.99 % y^{-1}) and a steep and more constant Theil-Sen's line (Figure 8d). Similar large declines occurred in the centre-south of the country, as it was found for Svedån Sved (-1.91 % y^{-1}) (Figure 9a), while moderate and milder relative Theil-Sen's slopes were observed for the stations Nissan Halmstad (-1.41 % y^{-1}) (Figure 9b) and Emån Emsfors (-0.85 % y^{-1}) (Figure 9c), respectively situated on the south-west and south-east Swedish coast.



Figure 8. Largest Tot-P concentration declines ($\mu g L^{-1}$) for the river stations Ljusnan Funäsdalen (a), Ljungan Skallböleforsen (b), Skellefte älv Slagnäs (c) and Skellefte älv, Kvistforsen (d) over the period 1980 – 2020.



Figure 9. Tot-P concentration declines ($\mu g L^{-1}$) for the south riverine stations Svedån Sved (a), Nissan Halmstad (b), and Emån Emsfors (c) over the period 1980–2020.

4.4. Tot-P and PO₄P trends correlations with other parameters

The estimated PO₄P and Tot-P time trends were compared with the retrieved variables on land use and cover and coordinates to assess potential existing correlations between P concentrations and the parameters. Δ PO₄P and Δ Tot-P concentrations over the period 1980-2020 were plotted against the land use classes and geographic position (N-E) to visualise the relationships between the variables. For neither PO₄P nor Tot-P correlational relationships were detected with any of the examined variables. As the obtained charts showed that no correlation existed between x and y axes in any of the case, further analyses were not performed.

4.5. Variations in Growing Degree Days (GDDs)

The total number of GDDs with 5° C as base temperature was calculated to assess variations in the length of frost-free for the 34 river stations between 1950 - 2020. Results showed a consistent increase in the number of days with average $\geq 5^{\circ}$ C temperature across Sweden (Appendix F) and, particularly, the comparison between the 10-year long average sum of GDDs for 1950 - 1959 and 2011 - 2020 provided a mean increase of 323 days for the 34 selected stations (Figure 10).



Figure 10. GDDs variation for the analysed river stations for the period 1950 - 2020. Blue bars represent the mean number of GDDs in 1950 - 1959, while orange bars display the mean number of GDDs in 2011 - 2020.

The most relevant increase in the number of GDDs was observed for rivers and streams of central and south Sweden (704 – 357 days), with the maximum positive increment registered for *Lyckebyån Lyckeby* (704 days) (Figure 11a) that did not account for statistically significant (Mann-Kendall test, p value <0.05) changes in Tot-P nor PO₄P. Station *Gullspångsälv*. *Gullspång* that recorded one of the largest PO₄P declines (2.00 % y^{-1}) and mid-range Δ Tot-P (-1.38 % y^{-1}) also showed the second largest increase of GDDs (514 days) (Figure 11b), likewise *Lögde älv Lögdeå*, representing the only positive trend in Δ PO₄P (+1.71 % y^{-1}) and not recording statistically significant (Mann-Kendall test, p value <0.05) variations in Tot-P, displayed an important positive increment of GDDs (357 days) (Figure 11c).



Figure 11. Increase in GDDs over 1950 – 2020. The larger increases were registered for the river stations Lyckebyån Lyckeby (5a), Gullspångsälv. Gullspång (5b) and Lögde älv Lögdeå (5c).

Among the locations characterised by the most relevant increases (704 – 357 days), two stations (*Vindelälven Maltbrännan* and *Skellefte älv, Kvistforsen*) are situated in north Sweden and showed consistent positive increment in the number of GDDs of respectively 437 and 369 days (Figure 12). *Vindelälven Maltbrännan* and *Skellefte älv, Kvistforsen* are also present in the list of rivers and streams (n= 10) registering the steepest Tot-P declines ($\geq 2.00 \% y^{-1}$), for which the comparison between the 10-year long average sum of GDDs for 1950 – 1959 and 2011 – 2020

provided a mean positive increment of 280 days, with *Vindelälven Maltbrännan* and *Skellefte älv, Kvistforsen* far exceeding 1000 days during the last decade.



Figure 12. Most relevant increases in GDDs for the northern stations of Skellefte älv, Kvistforsen (12a) and Vindelälven Maltbrännan (12b) over the period 1950 – 2020.

5. Discussion

The chapter discusses the previously presented findings by linking them to the objectives of the study and comparing them with the results of the existing literature on the topic. This work aimed at investigating whether similar P declines observed by Huser et al. (2020) and Huser et al. (2018) for minimally disturbed Swedish lakes could be found in waterbodies subject to minimal anthropogenic disturbance and, thus, revealing whether analogous oligotrophication processes are occurring at catchment scale. Additional objectives regarded the assessment of possible drivers for the detected Δ P over the period 1980 – 2020 and, so, the study of the potential correlational relationship existing between the obtained P trends and other retrieved variables (land use and land cover). Aim and objectives are therefore addressed in the following sections of the chapter, concluded by two paragraphs on the limitations affecting the present work and future potential elucidative analyses on the subject matter.

5.1. Comparison with previous studies

The obtained results indicate an evident nutrient depletion for the period 1980 – 2020 occurring in watercourses minimally affected by anthropogenic pressures across Sweden. Consistently with previous research on lakes and rivers in Sweden (Huser et al., 2020; Huser et al., 2018; Isles et al., 2018), the majority of the assessed statistically significant (Mann-Kendall test, p value <0.05) trends were negative, respectively 9 for PO₄P and 30 for Tot-P over the 34 riverine stations considered in the study. Declines in PO₄P were important as they ranged between from -1.3 to -2.3 % y^{-1} and concerned rivers and streams spreading from north to south in the country. Similar but more important declining trends were detected for the Tot-P content, which ranged from -0.5 to -3.1% y^{-1} and primarily involved the stations located in the north. As a matter of fact, despite a general negative Δ Tot-P could be observed in 97% of the selected watercourses covering the entire latitudinal length of the country, it was evident that the steepest Tot-P declines (>2.00 % v^{-1}) occurred between 65.67° (north of Slagnäs) and 61.21° N (south of Söderhamn). The assessed magnitudes of P depletion for the selected waterbodies for 1980 -2020 were consistent with those estimated for minimally disturbed Swedish lakes between 1988 – 2016 in Huser et al. (2018). In their research, the mean Theil-Sen's slope for Tot-P declines averaged >2.5% y^{-1} and the largest losses were observed for lakes already presenting low nutrients concentrations. Consistently with their analysis, the results obtained in the present study also provided a comparably important annual rate of change for Tot-P (>1.8 % y^{-1}) in the examined watercourses, and displayed that the steepest relative annual Tot-P declines were found for rivers and streams already having lowest Tot-P median concentration when the monitoring activity began. Similar declining trends were observed by Huser et al. (2020) for Swedish lakes and watercourses situated in the Near Arctic (areas outside the borders defined by the Arctic Biodiversity Assessment), where the largest Tot-P losses (almost -6 % y^{-1}) were detected compared to the other arctic zones. Despite no clear spatial pattern being distinguished, the study of Isles et al. (2018) also reported that the majority (n = 39) of the considered Swedish lakes displayed decreasing Tot-P concentration trends with similar behaviour for the Tot-P content in the 78 examined streams for 1998 - 2013. It is therefore evident that an oligotrophication process has been developing since the 1980s for waterbodies and watercourses located in Sweden. Neither in this nor previous comparable studies (Huser et al., 2020; Huser et al., 2018; Isles et al., 2018) clear and distinct spatial patterns of such a nutrient depletion were detected, but significantly noticeable P declines were found predominant in the most northern areas of Sweden.

5.2. Potential drivers for the observed trends

The analysis of the relationships between the assessed PO₄P and Tot-P trends and the other variables (land use and cover, coordinates) provided no graphically visible linear correlation. The lack of such a correlational relationship hinders the direct validation of any of the hypothesis proposed in previous studies (Huser et al., 2020; Huser et al., 2018; Isles et al., 2018) for P declines in Swedish freshwater ecosystems without the implementation of further analyses. However, because of some of the features characterising the results of the present work, it was possible to consider the hypothesised explanatory factors to assess the potential drivers for the ongoing oligotrophication process.

Phosphorus declines observed in the present study for rivers and streams located in south and central Sweden might be consistent with the hypothesis of recovery from acidification formulated by Huser et al. (2018) on the basis of previous research (Crossman et al., 2016; Gérard, 2016). As Akselsson et al. (2013), Futter et al. (2014) and Moldan et al. (2013) have described in their works, south and central Sweden were among the most acidified areas in the Fennoscandian region since 1967 and 1976 (Odén, 1976). The monitoring activity began in 1984 has shown a slow process of recovery from historic acidification for these parts of the country (Fölster et al., 2014), where recovery is intended as "a gradual process of increasing pH and improved biological conditions" (Futter et al., 2014). Such an improvement is the result of the implementation of CLRTAP in 1983 (UNECE, 2012) and subsequent declining acid deposition. As observed by Monteith et al. (2007), changes in acid deposition have been responsible for > 85% of increasing DOC concentrations (that can be used as proxy for DOM) in surface water in regions recovering from acidification, given that significative reductions of anthropogenic SO₄² have been linked to increased soil pH and decreased Al mobilisation. In addition to this, the stepwise multiple linear regression performed by Huser et al. (2018) provided that Δ TOC (Total Organic Carbon), used as a proxy for DOM, explained for 63% of the Tot-P trends for the examined lakes.

Given the findings of Monteith et al. (2007) on DOM concentrations and recovery from acidification, and the strong correlation between Δ DOM and Tot-P assessed by Huser et al. (2018), the observed Δ Tot-P of rivers and streams located in southern and central Sweden might be coherent with the hypothesis of recovery from acidification. A return to pre-disturbance conditions as a potential explanatory factor for the current oligotrophication in these areas of the country is also consistent considering that the watercourses of the present work and the lakes of Huser et al. (2018) belong to the same Swedish National Monitoring Programme, and, therefore, the watersheds and catchments under study present similar environmental and disturbance conditions.

A similar explanatory factor has been used by Crossman et al. (2016) for explaining Tot-P declines in Precambrian Shield landscapes (south-central Ontario, Canada), characterised by low pH and historical acid deposition. Their study highlighted how Δ P and, thus, P mobility and exports are linked to disturbance events, and how such effects are stronger in riparian areas given the direct hydrological connection between aquatic and terrestrial ecosystems. When ecosystems are returning to pre-disturbance conditions, P declines might be interpreted as the tail end of a period with historically high P exports and, therefore, as a sign of increased P retention in watershed soils (Crossman et al., 2016). Greater soil P retention associated with increasing pH and, thus, recovery from acidification might also explain PO₄P declines assessed in the present study for the clayey areas of south and central Sweden (Ulén & Jakobsson, 2005; Uusitalo et al., 2003). As studied by Gérard (2016), clay minerals (depending on their surface area) can have a greater PO₄P binding capacity than Al or Fe oxides, and show maximum sorption capacity when pH is 4-7, compatibly with increasing soil pH derived by recovery from acidification.

While a return to pre-disturbance conditions is a plausible driver for the observed P declines in south and central Sweden, different explanatory factors can be hypothesised for the significant oligotrophication process occurring in the areas

minimally affected by acidification in the north of the country. Previous studies (Huser et al., 2018; Huser et al., 2020) have proposed greater terrestrial vegetation control over the ecosystem nutrient budget as a potential driving mechanism for the observed declining P content. Compatibly with the growing forest biomass and productive forest land use registered in north Sweden during the last decades (SLU, 2021), Akselsson et al. (2008) have reported the risks of limitation to the soil P budget (and, thus, P declines in watersheds and catchments and, ultimately, in lakes and rivers) with intense forestry activity and harvesting. Analogous hypotheses have been used by Crossman et al. (2016) and Stammler et al. (2017) to describe the impact of disturbance events on Δ P in Precambrian Shield landscape, exacerbated by the highest P uptake of young vegetation with further limitation on soil P mobility. The mechanism of tightened nutrient cycle linked to forestry management has also been addressed in Lucas et al. (2016) for explaining the observed declines in riverine N content across northern Sweden. No linear correlation was found in the present study between the recorded ΔP (Tot-P and PO₄P) and land use and cover (forest on mineral soil and wetland), and, therefore, no direct validation of the suggested hypothesis could be determined. However, contribution to oligotrophication from climate change (that growing forestry biomass has been associated with in Huser et al., 2020; Huser et al., 2018; Lucas et al., 2016; Stammler et al., 2017) might be a consistent explanatory driver for the P declines in north of Sweden.

Most of the land use and cover in north Sweden is constituted of forests (SLU, 2021). As described in deWit et al., 2016, forest land is usually characterised by a carbon-rich topsoil layer and high terrestrial nutrient inputs with consequent high DOC concentrations (used as proxy for DOM) found in soil and soil water solution. Wetter climatic conditions as those recorded in Sweden during the last decades (SMHI, 2021c) have been correlated to increasing DOM exports from the upper forest floor to waterbodies, as the brownification process of Fennoscandian waterbodies has proven, and, therefore, different water pathways (lateral transport) in watersheds and catchments (deWit et al., 2016; Hongve et al., 2004). Given that DOM is the principal vector of P through DOP (Dissolved Organic Phosphorus) but rivers and lakes in north Sweden are showing significant oligotrophication, it is plausible that P concentrations in terrestrial DOM inputs are declining (Huser et al., 2018). As a matter of fact, Giesler et al. (2005) have demonstrated that in boreal peat soils of groundwater discharge areas the competition between P and DOC with the Al and Fe surface sites leads to high phosphate sorption and DOM desorption with low DOP releases. If P exports to waterbodies are already naturally limited by in-catchment processes, the final P budget of lakes and streams might also be altered by the effect of climate change modifying the length of seasons and, thus, shifting timing and magnitude of seasonal relevant phenomena linked to terrestrial inputs.

Spring snowmelt is the primary mechanism for supplying water and nutrients to lakes in boreal regions (Hrycik et al., 2021). The process is historically characterised by seasonal peaks in the runoff from accumulated nutrients and water in winter snow, but increased winter rains combined with early snowmelt due to the altered climate have led to changes in the final and seasonal budget of nutrients input (Hrycik et al., 2021). Previous studies (Hrycik et al., 2021 and references therein) have assessed that the nutrient load is significantly reduced in runoff occurring from a frozen or covered with snow ground compared to streamflow of thawed and saturated spring soils. Moreover, an additional loss of nutrients might result from their early access in waterbodies during winter when solar radiation and isothermal conditions are still low, leading to limitation in the nutrient uptake by phytoplankton and increased absorption to sinking particles, and, thus, removing nutrients from the water column (Hrycik et al., 2021). The loss is also exacerbated by the presence of ice and/or snow layers, which further limits light in the water column (Bolsenga and Vanderploeg, 1992) and promotes inverse stratification (Hrycik et al., 2021). Under these conditions, nutrients inputs in the form of shallow plume under the surface ice layer are prevented from mixing with the water below, leading to reduced residence time, and encouraging their horizontal passage between the waterbody outlet and ice/snow layer with subsequent outflow (Cortés et al., 2017). Under-ice snowmelt is also responsible for delivering oxygen to waterbodies, which remediates anoxic or suboxic conditions, limits the release of sediment P in the water column, and promotes the scavenge of PO₄P from the surface due to the enrichment of oxidized manganese (Mn) and Fe (Joung et al., 2017). These processes resulting from early snowmelt and, thus, altered seasonal temperatures are compatible with the observations made on the average GDD in the present study. A mean increase of 280 days (437 - 220 days) registering temperatures $\geq 5^{\circ}$ C was recorded for the northern river stations showing the most significant $\Delta P (> 2.00 \% y^{-1})$ in 2011 – 2020 compared to 1950 – 1959, indicating an important shift in the length of frost-free seasons (autumn and spring). These results are therefore consistent with the hypothesis that climate change and its effects (warmer temperatures and longer frost-free seasons) might be a potential driver for the recorded P declines and, thus, a valid explanatory factor for the ongoing oligotrophication process of minimally disturbed freshwater ecosystems in north Sweden.

5.3. Limitations

The current work presents some limitations regarding the initial design and, particularly, the meeting criterion for the selection of watercourses used in the analyses. Given the primary objective of exploring rivers and streams minimally affected by anthropogenic disturbances, only stations located in sites with <5% of

watershed area used for agriculture and <1% of watershed area utilized for urbanisation were chosen, consistently with the selection criterion of Huser et al. (2018). Such a restriction limited the final number of suitable stations with water quality records covering the period 1980 - 2020 and, specifically, reduced the number of available streams and rivers representing the different areas and environments of Sweden. This limitation is well visible in Figure 4 where 21 stations are situated in Norrland, 7 in Svealand and 6 in Götaland, and the majority of analysed waterbodies is located on the eastern coastal area of Sweden. If the heterogeneous latitudinal distribution could be partially explained by the more intensive land use characterising the centre and south of Sweden (SLU, 2021) and consequent difficulty for streams and rivers to meet the minimally disturbed criterion, the longitudinal distribution is affected by the selecting limits and, therefore, the extent of the monitored watercourses in the Swedish National Monitoring Program. As the Swedish National Monitoring Program focuses on lakes and river mouths (the latter account for the runoff from 82% of Sweden) due to the strong link with the Baltic Marine Environment Protection Commission-Helsinki Commission (HELCOM) policy (Fölster et al., 2014), limited water quality assessments and Δ P trends could be estimated for the upstream part of watercourses, where the most significant oligotrophication was observed. For instance, only 1 suitable station (Ljusnan Funäsdalen) could be found in the mountain area of Sweden, and the majority of rivers and streams that displayed the steepest P declines are located in the northern mountain and inland sites. Likewise, the lack of monitoring stations situated in the upstream and downstream parts of the same watercourses, as well as at the inlet and outlet of lakes, constraints the exact understanding of detailed catchment processes for the ongoing oligotrophication, and, therefore, limits the comprehension of the potential drivers.

A further limitation could be found in the restricted number of variables analysed in the present study, as only Tot-P and PO₄P concentration values were retrieved for the water quality assessment of the selected watercourses. Despite this being consistent with the primary objective of the project (to investigate potential P declines in Swedish rivers and streams subject to minimal anthropogenic disturbance), it limits the potential for validation of various hypothesised explanatory factors associated with other water quality variables discussed in the work, for instance DOM, DOC, and pH. Similar restrictions might be produced by the limited study of climatic variables since warmer climate seems to be of particular importance in regulating soil-water processes for P cycle in Sweden. Moreover, given the significant role that soil chemistry plays in determining the exported content of the water solution and, thus, in influencing the intimate interaction between terrestrial and aquatic ecosystem, the lack of more precise soil and vegetation (e.g., forest) data and their inclusion in the present work might further limit the exact comprehension of watershed and in-catchment processes linked to P declines and oligotrophication.

5.4. Future research

As evidenced by the work herein presented, further analyses are necessary to identify the exact explanatory factors for the ongoing nutrient depletion in Swedish freshwater ecosystems. Future research within the topic would benefit from a greater inclusion of the upstream part of watersheds in the Swedish National Monitoring Program and, therefore, a larger number of long-term monitoring stations located in the mountain and inland areas of Sweden where the most critical P declines are occurring. Given the evidence that soil, vegetation, and climate play an important role in regulating the P cycle in ecosystems and so the final P content in waterbodies, more extensive studies of these variables would be valuable to provide a more accurate interpretation of the significant environmental processes contributing to the ongoing oligotrophication.

Following the work of Giesler et al. (2005), it might be relevant to assess such a result in the catchment areas of north Sweden where the steepest Tot-P declines were detected. As this boreal region is dominated by forests (SLU, 2021) growing on peat soils (SGU, 2021) (and, thus subject to Al and Fe accumulation in the humus layer in groundwater discharge areas) (Giesler et al., 2005) subsequent increased soil P fixation and reduced DOP transport to waterbodies might contribute to the ongoing oligotrophication process. Therefore, to identify groundwater discharge areas in the catchments of interest and perform analyses regarding soil P sorption capacity and relative effects on soil variables as in Giesler at al. (2005) might be a valid direction for future research to assess the extent of the naturally limited P availability in the catchments.

In addition to this, the role of vegetation contributing to oligotrophication could be further explored in the wake of the hypotheses of Crossman et al. (2016), Huser et al. (2018) and Lucas et al. (2016). The total volume of forests has increased between 1956 and 2011 across Sweden and despite the decline registered in the last 10 years it remains well above 100 million m³ sk y^{-1} , with a mean annual positive increment following a decreasing gradient for north to south (SLU, 2021). About 6% of the total growing stock is registered above the border of near-alpine forest (also indicated as GFS, *gränsen för fjällnära skog*), an area of 8.1 million ha (circa 20% of total land) where productive forest land accounts for 37% of the surface (SLU, 2021). The region also comprises the most extensive proportion of old forests (> 120 years) as well as of large trees (diameter at breast height > 30 cm), with a mean annual increase of growing stock of 2 million m³ sk for the productive forest land (SLU, 2021). On the contrary, younger (< 40 years) and smaller trees are found below GFS, where 81% of the area is identified as productive forest (SLU, 2021).

As discussed by Crossman et al. (2016) in their study about ongoing oligotrophication in Canada, the mean annual Tot-P retention is larger in soils covered by forests or subject to forestry activity due to the high P uptake of trees compared to other vegetation types and land covers. Particularly, forestry influences long-term P exports in the catchment by removing nutrients from the ecosystems through harvesting (Akselsson et al., 2008) and by increasing P uptake with the establishment of young vegetation (Crossman et al., 2016). If so, such an explanation for reduced soil P mobility and content in the catchments below GFS might be consistent with the data presented in SLU (2021), where it is evident that forestry activity has been constantly increasing in Sweden from the 1900s, and the volume of harvest of living trees has grown in accordance with the increase of productive forest land. At the same time, given the consistent increase in total volume of forests recorded until 2010s, and the noticeable rate of growing stocks for the areas above GFS (where some of the river stations from the present work and lakes from Huser et al. (2018) registering the steepest Tot-P declines are closely located) old forest with large trees might affect P cycle from upstream analogously to the hypotheses of Huser et al. (2018) and Lucas et al. (2016). Greater volume of forest associated to larger trees might limit the vegetation control over terrestrial nutrient balances by increasing P uptake and storage in forest trees, with a subsequent decrease of P concentrations in soil and export to waterbodies (Huser et al., 2018; Lucas et al., 2018). Additionally, P limitation in upstream areas of catchments associated with increased terrestrial vegetation control might be strengthened by the variation in altitude of the tree line, which would lead to greater P uptake and storage in young trees and, thus, larger forest volume found in upstream areas. For these reasons, further investigation on the role of forests in the upstream and downstream parts of watersheds could be necessary for assessing their contribution to the oligotrophication process.

A more comprehensive study of climatic variables may also be required since and the consistency of warmer temperatures (and GDDs) with increasing growing stocks and volume of forests as well as higher altitude of the tree line, and the previously discussed influence of early snowmelt on the final P content in lakes. Particularly, further analyses on the long-term seasonal temperature variation in the catchments of interest with specific calculation of the length of frost-free seasons might help assess the time of occurrence of the spring snowmelt and evaluate its impact on the final P concentration in waterbodies.

6. Conclusions

In accordance with previous research works on the subject matter, the study confirmed that a significant nutrient depletion is occurring in minimally anthropogenic disturbed waterbodies across Sweden. The herein presented results provided that critical decreasing P trends have been affecting rivers and streams located in environments characterised by < 5 % agricultural land use and < 1 % urbanisation in the last 40 years. Particularly, for the period 1980 – 2020 statistically significant (Mann-Kendall test, p value <0.05) PO₄P losses were assessed for about 26% of the analysed watercourses, with annual slopes ranging from +1.71 to -2.36 % y^{-1} and averaging -1.41 % y^{-1} (-1.75 % y^{-1}) for the negative trends only) while statistically significant (Mann-Kendall test, p value <0.05) decreasing Tot-P trends were greater in number, as they were recorded for 97% of the studied rivers and varied from -3.17 to -0.53 % y^{-1} with an average yearly rate of change >1.8 % y^{-1} .

Despite the failure to identify any significant correlational relationship found between the detected P trends and other retrieved variables (land use and cover, coordinates) the characteristics of Δ P results were consistent with some of the explanatory factors hypothesised on previous studies. Ongoing declines in streams and rivers of south and central Sweden might be ascribed to increasing pH and clay mineral absorption capacity, and, thus, recovery from acidification; while the significant P losses of northern watercourses could depend more on the effects of climate change on the early occurrence of seasonal phenomena such as spring snowmelt. Warmer temperatures as a potential driver are consistent with the results on increasing number of GDDs across Sweden and in the locations of the north where the steepest nutrient losses were recorded.

However, further studies are necessary to exactly understand the explanatory factors behind the oligotrophication process occurring at catchment level in freshwater ecosystems of Sweden. Future research on the topic should extend the analysis to vegetation and soil variables as well as other climatic seasonal and water quality parameters (for instance pH, DOM) given their evident significant role in affecting the P cycle in terrestrial and aquatic ecosystems and, thus, the final P content in waterbodies. More promptly, in view of the obtained results in the present study, and the significant evidence that oligotrophication is taking place in various freshwater ecosystems of the northern hemisphere (Arvola et al., 2011; Crossman et al., 2016; Eimers et al., 2018; Eimers et al., 2009; Huser et al., 2020; Huser et al.,

2018; Hu and Huser, 2014; Isles et al., 2018; Stammler et al., 2017; Tong et al. 2017), water quality policies should complement the already present and used upper concentration limits with lower nutrient concentration thresholds in water quality policy mandates, as for instance in the European WFD, to assess ecosystem health and consider possible water quality impairments that might develop from nutrient depletion.

References

- Akselsson, C., H. Hultberg, P.E. Karlsson, G. Pihl Karlsson, and S. Hellsten.
 2013. Acidification trends in south Swedish forest soils 1986–2008—Slow recovery and high sensitivity to sea-salt episodes. *Science of the Total Environment*, 444, pp. 271–287. Available at: https://doi.org/10.1016/j.scitotenv.2012.11.106.
- Akselsson, C., Westling, O., Alveteg, M., Thelin, G., Fransson, A.M. & Hellsten, S. (2008). The influence of N load and harvest intensity on the risk of P limitation in Swedish forest soils. *Science of The Total Environment*, 404(2-3), pp. 284–289. Available at: https://doi.org/10.1016/j.scitotenv.2007.11.017.
- Almer, B., W. Dickson, C. Ekström & E. Hörnström. (1974). Effects of acidification on Swedish lakes. *Ambio*, 3, pp. 30–36.
- Arvola, L., Järvinen, M. & Tulonen, T. (2011). Long-term trends and regional differences of phytoplankton in large Finnish lakes. *Hydrobiologia*, 660, pp. 125–134. Available at: <u>https://doi.org/10.1007/s10750-010-0410-9</u>.
- Bolsenga, S. J. and Vanderploeg, H. A. (1992). Estimating photosynthetically available radiation into open and ice-covered freshwater lakes from surface characteristics; a high transmittance case study. *Hydrobiologia*, 243–244(1), pp. 95–104. Available at: https://doi.org/10.1007/BF00007024.
- Conservation of Arctic Flora and Fauna (CAFF) (2019). CAFF Circumpolar Biodiversity Monitoring Program State of Arctic Freshwater Biodiversity Report. Available at: <u>https://oaarchive.arcticcouncil.org/handle/11374/2315</u>.
- Cortés, A., MacIntyre, S. and Sadro, S. (2017). Flowpath and retention of snowmelt in an ice-covered arctic lake. *Limnology and Oceanography*, 62(5), pp. 2023–2044. Available at: <u>https://doi.org/10.1002/lno.10549</u>.
- Crossman, J., Eimers, M.C., Watmough, S.A., Futter, M.N., Kerr, J., Baker, S.R., & Dillon, P.J. (2016). Can recovery from disturbance explain observed declines in total phosphorus in Precambrian Shield catchments? *Canadian Journal of Fisheries and Aquatic Sciences*, 73(8), pp. 1202-1212. Available at: <u>https://doi.org/10.1139/cjfas-2015-0312</u>.

- de Wit, H.A., Valinia, S., Weyhenmeyer, G.A., Futter, M.N., Kortelainen, P., Austnes, K., Hessen, D.O., Räike, A., Laudon, H., Vuorenmaa, J., 2016. Current Browning of Surface Waters Will Be Further Promoted by Wetter Climate. *Environmental Science & Technology Letters* 3(12), pp. 430– 435. Available at: https://doi.org/10.1021/acs.estlett.6b00396.
- Eimers, M.C., Hillis, N.P. & Watmough, S.A., (2018). Phosphorus deposition in a low-Phosphorus landscape: sources, accuracy and contribution to declines in surface water P. *Ecosystems*, 21(4), pp.782–794. Available at: https://doi.org/10.1007/s10021-017-0184-2.
- Eimers, M.C., Watmough, S.A., Paterson, A.M., Dillon, P.J. & Yao, H. (2009).
 Long-term declines in phosphorus export from forested catchments in south-central Ontario. *Canadian Journal of Fisheries and Aquatic Sciences*, 66(10), pp. 1682–1692. Available at: https://doi.org/10.1139/F09-101.
- European Water Framework Directive (EUWFD) (2000). The EU Water Framework Directive: integrated river basin management for Europe. *Official Journal* L 327, pp.1-73. Available at: http://data.europa.eu/eli/dir/2000/60/oj.
- Fölster, J., Johnson, R.K., Futter, M.N. & Wilander, A. (2014). The Swedish monitoring of surface waters: 50 years of adaptive monitoring. *Ambio*, 43, pp. 3–18. Available at: <u>https://doi.org/10.1007/s13280-014-0558-z</u>.
- Futter, M.N., Valinia, S., Löfgren, S. *et al.* Long-term trends in water chemistry of acid-sensitive Swedish lakes show slow recovery from historic acidification. *Ambio*, 43, pp. 77–90 (2014). Available at: <u>https://doi.org/10.1007/s13280-014-0563-2</u>.
- Geological Survey of Sweden (SGU) (2020). Geology of Sweden. Available at: <u>https://www.sgu.se/en/geology-of-sweden/</u>.
- Gérard, F. (2016). Clay minerals, iron/aluminum oxides, and their contribution to phosphate sorption in soils a myth revisited. *Geoderma*, 262, pp. 213–226. Available at: <u>https://doi.org/10.1016/j.geoderma.2015.08.036</u>.
- Giesler, R., Andersson, T., Lövgren, L. and Persson, P. (2005). Phosphate Sorption in Aluminum- and Iron-Rich Humus Soils. Soil Sciences of American Journal, 69(1), pp. 77-86. Available at: <u>https://doi.org/10.2136/sssaj2005.0077a</u>.
- Gustafsson, J.P., Mwamila, L.B. & Kergoat, K. (2012). The pH dependence of phosphate sorption and desorption in Swedish agricultural soils. *Geoderma*, 189, pp. 304–311. Available at: https://doi.org/10.1016/j.geoderma.2012.05.014.
- Helsel, D.R. & Hirsch, R.M. (1992). Statistical methods in water resources. Techniques of Water-Resources Investigations 04-A3. U.S. Geological Survey. Available at: <u>https://doi.org/10.3133/twri04A3</u>
- Helsel, D.R., Hirsch, R.M., Ryberg, K.R., Archfield, A.S. & Gilroy, E.J. (2020). Statistical Methods in Water Resources. Techniques and Methods 3A. Available at: <u>https://doi.org/10.3133/tm4A3</u>.

- Hongve, D., Riise, G. & Kristiansen, J.F. Increased colour and organic acid concentrations in Norwegian forest lakes and drinking water – a result of increased precipitation? *Aquatic Sciences*, 66(2), pp. 231–238 (2004). Available at: https://doi.org/10.1007/s00027-004-0708-7.
- Hu, Q. & Huser, B.J. (2014). Anthropogenic oligotrophication via liming: longterm phosphorus trends in acidified, limed, and neutral reference lakes in Sweden. *Ambio*, 43, pp. 104–112. Available at: <u>https://doi.org/10.1007/s13280-014-0573-0</u>.
- Huser, B.J. & Rydin, E. (2005). Phosphorus inactivation by aluminium in Lakes Gårdsjön and Härsvatten sediment during the industrial acidification period in Sweden. *Canadian Journal of Fisheries and Aquatic Sciences*, 62, pp. 1702–1709. Available at: <u>https://doi.org/10.1139/f05-083</u>.
- Huser, B.J., Futter, M.N., Bogan, D., Brittain, J.E., Culp, J.M., Goedkoop, W., Gribovskaya, I., Karlsson, J., Lau, D.C., Rühland, K.M. & Schartau, A.K., (2020). Spatial and temporal variation in Arctic freshwater chemistry— Reflecting climate-induced landscape alterations and a changing template for biodiversity. *Freshwater Biology*. Available at: https://doi.org/10.1111/fwb.13645.
- Huser, J.B., Futter, N.M., Wang, R. & Foster, J. (2018). Persistent and widespread long-term phosphorus declines in Boreal lakes in Sweden. *Sciences of The Total Environment*, 613-614, pp. 240–249. Available at: https://doi.org/10.1016/j.scitotenv.2017.09.067
- Hynes, H.B.N. (1975) The Stream and its Valley. Internationale Vereinigung Für Theoretische Und Angewandte Limnologie. *Verhandlungen*, 19, pp. 1–15.
- Isles, P.D.F., Creed, I.F., & Bergström, A.K. (2018). Recent synchronous declines in

DIN:TP in Swedish lakes. *Global Biogeochemical Cycles*, 32, pp. 208–225. Available at: <u>https://doi.org/10.1002/2017GB005722</u>

- Jones, A.S., Stevens, D.K., Horsburgh, J.S. & Mesner, N. O. (2011). Surrogate measures for providing high frequency estimates of total suspended solids and total phosphorus concentrations. JAWRA Journal of American Water Resources Association, 47(2), pp. 239-253. Available at: <u>https://doi.org/10.1111/j.1752-1688.2010.00505.x</u>.
- Joung, D. J., Leduc, M., Ramcharitar, B., Xu, Y., Isles, P. D. F., Stockwell, J. D., Druschel, G. K., Manley, T. and Schroth, A. W. (2017). Winter weather and lake-watershed physical configuration drive phosphorus, iron, and manganese dynamics in water and sediment of ice-covered lakes. Limnology and Oceanography, 62(4), pp. 1620–1635. Available at: https://doi.org/10.1002/lno.10521.
- Kendall, M.G. (1975). Rank Correlation Methods. 4th edition. Charles Griffin, London.
- Kopáček, J., Hejzlar, J., Kaňa, J., Norton, S.A. & Stuchlík, E. (2015). Effects of acidic deposition on in-lake phosphorus availability: a lesson from lakes

recovering from acidification. *Environmental Science & Technology*, 49(5), pp. 2895–2903. Available at: <u>https://doi.org/10.1021/es5058743</u>.

- Lucas, R.W., Sponseller, R.A., Gundale, M.J., Stendahl, J., Fridman, J., Högberg,
 P. & Laudon, H. (2016). Long-term declines in stream and river inorganic nitrogen (N) export correspond to forest change. *Ecological Applications*, 26(2), pp. 545–556. Available at: <u>https://doi.org/10.1890/14-2413</u>.
- Mann, H.B. (1945). Nonparametric tests against trend. *Econometrica*, 12(3), pp. 245–259. Available at: <u>https://doi.org/10.2307/1907187</u>.
- McLeod, A.I. (2011). Kendall: Kendall rank correlation and Mann-Kendall trend test. R package version 2.2. Available at: <u>https://CRAN.R-</u> project.org/package=Kendall.
- Moldan, F., Cosby, B.J. & Wright, R.F. (2013). Modeling past and future acidification of Swedish lakes. *Ambio*, 42, pp. 577–586. Available at: <u>https://doi.org/10.1007/s13280-012-0360-8</u>.
- Monteith, D.T., Stoddard, J.L., Evans, C.D., deWit, H.A., Forsius, M., Høgåsen, T., Wilander, A., Skjelkvåle, B.L., Jeffries, D.S., Vuorenmaa, J., Keller, B., Kopáček, J. & Vesely, J. (2007). Dissolved organic carbon trends resulting from changes in atmospheric deposition chemistry. *Nature*, 450, 537–540. Available at: <u>https://doi.org/10.1038/nature06316</u>.
- Morgan, M.A. (1997). The behaviour of soil and fertilizer phosphorus, 1997. pp 137–149. CAB International. ISBN: 0851991564.
- NASA (2021). Panoply netCDF, HDF and GRIB Data Viewer. Available at: <u>https://www.giss.nasa.gov/tools/panoply/</u>.
- Odén, S. 1976. The acidity problem—An outline of concepts. *Water, Air, and Soil Pollution,* 6, pp. 137–166. Available at: https://doi.org/10.1007/BF00182862.
- Pechlivanidis, I.G., Gupta, H. & Bosshard, T. (2018). An information theory approach to identifying a representative subset of hydro-climatic simulations for impact modeling studies. *Water Resources Research*, 54, pp. 5422–5435. Available at: <u>https://doi.org/10.1029/2017WR022035</u>.
- Penn, Chad & Camberato, James. (2019). A Critical Review on Soil Chemical Processes that Control How Soil pH Affects Phosphorus Availability to Plants. Agriculture, 9(6),120. Available at: https://doi.org/10.3390/agriculture9060120.
- Pohlert, T. (2020). trend: Non-Parametric Trend Tests and Change-Point Detection. R package version 1.1.4. Available at: <u>https://CRAN.R-project.org/package=trend</u>.
- RStudio Team (2021). RStudio: Integrated Development Environment for R. RStudio, PBC, Boston, MA. Available at: <u>http://www.rstudio.com/</u>.
- Ruttenberg, K.C. (2014). 10.13 The Global Phosphorus Cycle A2 Holland, Heinrich D. In: Turekian, K. K. (Ed) *Treatise on Geochemistry*. (2nd edition). pp 499–558. Oxford: Elsevier.

- Sen, P.K. (1968). Estimates of the regression coefficient based on Kendall's tau. *Journal of the American Statistical Association*, 63(324), pp. 1379–1389. Available at: https://doi.org/10.1080/01621459.1968.10480934.
- Skjelkvåle, B. L., Mannio, J., Wilander, A., & Andersen, T. (2001). Recovery from acidification of lakes in Finland, Norway and Sweden 1990–1999, Hydrology and Earth System Sciences, 5(3), pp. 327–338. Available at: <u>https://doi.org/10.5194/hess-5-327-2001</u>.
- Smith, V.H. & Schindler, D.W. (2009). Eutrophication science: where do we go from here? *Trends in Ecology & Evolution*, 24(4), pp. 201–207. Available at: <u>https://doi.org/10.1016/j.tree.2008.11.009</u>.
- Stammler, K.L., Taylor, W.D. & Mohamed, M.N. (2017). Long-term decline in stream total phosphorus concentrations: a pervasive pattern in all watershed types in Ontario. *Journal of Great Lakes Research*, 43(5), pp.930–937. Available at: https://doi.org/10.1016/j.jglr.2017.07.005.
- Statistiska centralbyrån (SCB) (2019). Markanvändningen i Sverige, Sjunde utgåvan [Land use in Sweden, Seventh Edition]. Available at: <u>https://www.scb.se/contentassets/eaa00bda68634c1dbdec1bb4f6705557/m</u> i0803_2015a01_br_mi03br1901.pdf.
- Stein et al., 2021 Stein, R., Karlsen, K., Høgda, B., Johansen, A., Elvebakk, A. & Tømmervik, H. (2021). Use of AVHRR NDVI data to map vegetation zones in north-western Europe. Available at: <u>https://www.researchgate.net/publication/268012841_Use_of_AVHRR_N</u> DVI data to map vegetation zones in north-western Europe.
- Stoddard, J.L., Van Sickle, J., Herlihy, A.T., Brahney, J., Paulsen, S., Peck, D.V., Mitchell, R. & Pollard, A.I. (2016). Continental-scale increase in Lake and stream phosphorus: are oligotrophic systems disappearing in the United States? *Environmental science & technology*, 50(7), pp. 3409-3415. Available at: <u>https://doi.org/10.1021/acs.est.5b05950</u>.
- Sverige Lantbruksuniversitet (SLU), Artdatabanken (2020). Lakes and streams. Available at: <u>https://www.artdatabanken.se/arter-och-natur/naturtyper/sjoar-och-vattendrag/</u>.
- Sverige Lantbruksuniversitet (SLU), Institutionen för skoglig resurshushållning (2021). Skogsdata 2021: Aktuella uppgifter om de svenska skogarna från SLU Riksskogstaxeringen, Tema: Fjällskogen [Forest data 2021: current data on Swedish forests from SLU Nation Forest Assessment, Theme: Mountain forests]. Available at: https://www.slu.se/globalassets/ew/org/centrb/rt/dokument/skogsdata/skog

sdata_2021_webb.pdf.

Swedish Environmental Protection Agency (SEPA) (2018). Sweden's Environmental Objectives: an introduction. ISBN 978-91-620-8820-0.

Swedish Meteorological and Hydrological Institute (SMHI) (2021a). Sveriges klimat [Sweden's climate]. Available at: https://www.smhi.se/kunskapsbanken/klimat/sveriges-klimat. Swedish Meteorological and Hydrological Institute (SMHI) (2021b). Sveriges klimat har blivit varmare och blötare [Sweden's climate has become warmer and wetter]. Available at: https://www.smhi.se/kunskapsbanken/klimat/sveriges-klimat/sveriges-

klimat-har-blivit-varmare-och-blotare-1.21614.

- Swedish Meteorological and Hydrological Institute (SMHI) (2021c). Sveriges sjöar [Sweden's lakes]. Available at: <u>https://www.smhi.se/kunskapsbanken/hydrologi/sveriges-sjoar/sverigessjoar-1.4221.</u>
- Tammi, J., M. Appelberg, U. Beier, T. Hesthagen, A. Lappalainen, & M. Rask. 2003. Fish status survey of Nordic lakes: Effects of acidification, eutrophication and stocking activity on present fish species composition. *Ambio*, 32(2), pp. 98–105. Available at: <u>https://doi.org/10.1579/0044-7447-32.2.98</u>.
- Theil, H. (1950). A rank-invariant method of linear and polynomial regression analysis, I. Proc. Kon. Ned. Akad. v. Wetensch.A53, 386-392.
- Tipping, E., Benham, S., Boyle, J.F., Crow, P., Davies, J., Fischer, U., Guyatt, H., Helliwell, R., Jackson-Blake, L., Lawlor, A.J., Monteith, D.T., Rowe, E.C. & Toberman, H. (2014). Atmospheric deposition of phosphorus to land and freshwater. *Environmental Sciences: Processes & Impacts*, 16(7), pp.1608–1617. Available at: https://doi.org/10.1039/C3EM00641G.
- Tong, Y., Zhang, W., Wang, X., Couture, R.M., Larssen, T., Zhao, Y., Li, J., Liang, H., Liu, X., Bu, X. & He, W., (2017). Decline in Chinese lake phosphorus concentration accompanied by shift in sources since 2006. *Nature Geoscience*, 10(7), pp.507–511. Available at: https://doi.org/10.1038/ngeo2967.
- Ulén, B. & Jakobsson, C. (2005). Critical evaluation of measures to mitigate phosphorus losses from agricultural land to surface waters in Sweden. *Science of The Total Environment*, 344(1–3), pp. 37–50. Available at: <u>https://doi.org/10.1016/j.scitotenv.2005.02.004</u>.
- United Nations Economic Commission for Europe (UNECE) (2012). Adjustments under the Gothenburg Protocol to emission reduction commitments or to inventories for the purposes of comparing national total emissions with them (2012/3). Available at: <u>https://unece.org/decisions</u>.
- Uusitalo, R., Turtola, E., Puustinen, M. & Paasonen-kivekäs, M. (2003) Contribution of Particulate Phosphorus to Runoff Phosphorus Bioavailability. *Journal of Environmental Quality*, 32(6), pp. 2007-2016. Available at: <u>https://doi.org/10.2134/jeq2003.2007</u>.
- Wang, R., Balkanski, Y., Bopp, L., Aumont, O., Boucher, O., Ciais, P., Gehlen, M., Penuelas, J., Ethé, C., Hauglustaine, D., Li, B., Liu, J., Zhou, F. & Tao, S. (2015). Influence of anthropogenic aerosol deposition on the relationship between oceanic productivity and warming. *Geophysical Research Letters*, 42(24), pp. 10,745–10,754. Available at: <u>https://doi.org/10.1002/2015GL066753</u>.

- Weil, R. & Brady, N. (2017). The Nature and Properties of Soils. 15th edition. Pearson Education. ISBN: 978-0133254488.
- Wetzel, R.G. (2001). Limnology: Lake and river ecosystems. 3rd Edition. San Diego: Academic Press. ISBN 978-0-12-744760-5.
- Wilander, A., and J. Fölster. 2007. Sjöinventeringen 2005. En synoptisk vattenkemisk undersökning av Sveriges sjöar. Rapport 2007:16.
 Institutionen för miljöanalys, Sveriges Lantbruksuniversitet (SLU), Uppsala. Available at: http://info1.ma.slu.se/IMA/Publikationer/internserie/2007-16.pdf.
- Williamson, C.E., Saros, J.E., Vincent, W.F. & Smol, J.P. (2009). Lakes and reservoirs as sentinels, integrators, and regulators of climate change. *Limnology and Oceanography*, 54(6), pp. 2273 – 2282. Available at: <u>https://doi.org/10.4319/lo.2009.54.6 part 2.2273</u>.
- Yoshimura, T., Nishioka, J., Saito, H., Takeda, S., Tsuda, A. & Wells, M. L. (2007). Distributions of particulate and dissolved organic and inorganic phosphorus in North Pacific surface waters. *Marine Chemistry*, 103(1–2), pp. 112–121. Available at: https://doi.org/10.1016/j.marchem.2006.06.011.

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Appendix A

Table A1. Information on ID, coordinates (SWEREF99 TM) and area of the 34 river stations analysed in the study as retrieved from SLU National Monitoring Program available at <u>https://miljodata.slu.se/MVM/</u>.

St. J.	MVM ID	SWI	SWEREF99 TM		
Station name		Ν	Ε	Area (km²)	
Alterälven Norrfjärden	37	7270752	800624	445.1188	
Ammerån Skyttmon	169	7032026	528249	2271.068	
Ångermanälven Sollefteå	18983	7006732	613993	29029.65	
Dalälven Älvkarleby	18970	6716732	633829	27557.98	
Emån Emsfors	18974	6334048	587911	4.0005	
Forsmarksån Johannisfors	30	6694843	676810	374.286	
Gide älv Gideåbacka	18982	7030703	705895	3422.053	
Gullspångsälv. Gullspång	590	6538630	449064	4941.592	
Indalsälven Bergeforsen	28	6935006	623440	23349.01	
Indalsälven Hammarstrand	1039	6999001	568382	21416.7	
Kalix älv Karlsborg	27	7326323	872437	23229.99	
Klarälven Almar	596	6591038	411956	6218.737	
Klarälven Edsforsen	167	6659630	417949	2608.246	
Klarälven Norra Råda	602	6652237	421507	1666.833	

Ljungan Skallböleforsen	18984	6916297	601468	12062.53
Ljusnan Funäsdalen	170	6938340	374935	294.9864
Ljusnan, Ljusne Strömmar	21	6788266	611834	19802.93
Lögde älv Lögdeå	38	7054698	719250	1605.641
Lule älv Luleå	18979	7293472	822571	24462.37
Lyckebyån Lyckeby	25	6228390	541157	795.1164
Nissan Halmstad	34	6285064	369774	2676.452
Öre älv Torrböle	18973	7072014	727041	2862.695
Pite älv Bölebyn	18977	7265328	792434	6848.455
Råne älv Niemisel	18972	7339849	815475	3713.772
Rickleån Robertsfors	1063	7132319	782392	1595.343
Sävjaån Ingvasta	164	6656120	658796	24.439
Skellefte älv Slagnäs	165	7287288	644564	2097.363
Skellefte älv, Kvistforsen	18980	7191952	774344	3062.129
Svedån Sved	172	6431712	448738	39.1112
Töre älv Infl.Bölträsket	36	7334144	846887	436.8946
Torne älv Mattila	18978	7336315	915269	266.8499
Upperudsälv. Köpmannebro	583	6518055	355776	3039.953
V. Dalälven Mockfjärd	166	6705176	494341	7463.599
Vindelälven Maltbrännan	168	7168178	705435	9820.16

Appendix B

Table B1. Land use and cover for the 34 river stations analysed in the study. Wetland, agriculture, urbanisation, other, open water, forest on mineral soil and forest on wetland (expressed as % of total watershed area) were obtained from the Department of Aquatic Sciences and Assessment at SLU.

Station name	Wet.	Agri.	Urb.	Other	Open water	Forest min. soil	Forest wetland
Alterälven Norrfjärden	8.39	2.66	0.11	3.22	4.27	73.46	7.89
Ammerån Skyttmon	19.36	0.86	0.09	8.27	6.42	58.28	6.73
Ångermanälven Sollefteå	12.63	0.37	0.07	14.05	8.89	60.65	3.33
Dalälven Älvkarleby	9.84	2.52	0.34	8.32	7.29	67.43	4.26
Emån Emsfors	2.51	0.41	0.09	6.45	0.26	80.48	9.80
Forsmarksån Johannisfors	9.24	4.28	0.13	5.04	4.43	64.87	12.01
Gide älv Gideåbacka	11.19	0.75	0.11	2.85	6.21	74.30	4.59
Gullspångsälv. Gullspång	5.99	3.37	0.41	4.30	13.47	67.55	4.92
Indalsälven Bergeforsen	11.35	1.49	0.15	19.14	10.31	53.80	3.76
Indalsälven Hammarstrand	11.86	1.50	0.14	20.55	10.75	51.55	3.64
Kalix älv Karlsborg	19.52	0.14	0.37	25.54	4.62	46.99	2.81
Klarälven Almar	9.23	1.98	0.22	5.50	7.72	70.59	4.76
Klarälven Edsforsen	10.97	0.83	0.14	5.38	5.62	72.21	4.85

Klarälven Norra Råda	9.73	0.59	0.19	3.34	10.25	70.41	5.49
Ljungan Skallböleforsen	8.13	0.83	0.13	9.90	7.84	69.15	4.03
Ljusnan Funäsdalen	5.81	0.23	0.13	62.40	1.58	29.68	0.17
Ljusnan, Ljusne Strömmar	10.82	1.49	0.18	11.26	5.66	67.25	3.35
Lögde älv Lögdeå	13.39	0.41	0.07	3.10	4.73	74.88	3.43
Lule älv Luleå	10.90	0.26	0.07	39.53	9.98	37.84	1.42
Lyckebyån Lyckeby	2.95	2.79	0.55	8.48	4.35	75.87	5.01
Nissan Halmstad	7.39	4.43	0.73	8.41	5.28	60.50	13.26
Öre älv Torrböle	16.31	1.14	0.07	2.98	3.24	70.91	5.35
Pite älv Bölebyn	11.72	0.51	0.12	7.44	6.22	71.01	3.00
Råne älv Niemisel	27.22	0.05	0.04	3.23	4.10	61.40	3.97
Rickleån Robertsfors	9.09	2.09	0.12	3.20	9.86	69.85	5.78
Sävjaån Ingvasta	0.33	2.66	0.42	3.87	0.71	82.96	9.05
Skellefte älv Slagnäs	12.25	0.02	0.10	7.48	26.01	52.77	1.35
Skellefte älv, Kvistforsen	14.46	1.10	0.33	4.47	7.19	66.90	5.54
Svedån Sved	1.80	2.60	0.11	7.00	4.65	74.16	9.67
Töre älv Infl.Bölträsket	19.92	0.15	0.07	3.31	3.19	66.36	7.00
Torne älv Mattila	16.22	4.13	0.13	18.74	4.73	45.74	10.31
Upperudsälv. Köpmannebro	1.99	2.93	0.31	6.70	16.54	68.54	2.98
V. Dalälven Mockfjärd	14.32	0.60	0.16	7.95	5.54	65.97	5.46
Vindelälven Maltbrännan	12.00	0.10	0.05	29.76	6.48	49.92	1.69

Appendix C

Table C1. Coordinates in SWEREF99 TM format retrieved from <u>https://miljodata.slu.se/MVM/Search</u>, relative conversion to WGS84 Decimal system (obtained from <u>https://www.slu.se/fakulteter/nj/om-fakulteten/centrum</u> <u>bildningar-och-storre-forskningsplattformar/faltforsk/utbildning-och-teknik/doku</u> <u>mentation/kartkoordinater/</u> – "*Kartvisning och konvertering*") and selection ranges used in Panoply 4.12.11 (201° N – 464° E).

	SWEREF99 TM		WGS84	Panoply		
Station name	N E		N E		N	E
Alterälven Norrfjärden	7270752	800624	65.4199292919	21.4859302052	161	248
Ammerån Skyttmon	7032026	528249	63.4156516414	15.5657659025	153	225
Ångermanälven Sollefteå	7006732	613993	63.1717178109	17.2641453309	152	232
Dalälven Älvkarleby	6716732	633829	60.5642064128	17.4413936565	142	231
Emån Emsfors	6334048	587911	57.1412711614	16.4527180127	128	228
Forsmarksån Johannisfors	6694843	676810	60.3513731733	18.2047949723	141	235
Gide älv Gideåbacka	7030703	705895	63.3455467308	19.1156624337	153	239
Gullspångsälv. Gullspång	6538630	449064	58.9841974555	14.1138106464	135	219
Indalsälven Bergeforsen	6935006	623440	62.5254063197	17.3985487593	150	232
Indalsälven Hammarstrand	6999001	568382	63.1138985708	16.3553785019	152	228
Kalix älv Karlsborg	7326323	872437	65.8395950036	23.1731729164	163	255
Klarälven Almar	6591038	411956	59.4486435071	13.4471905346	137	216

Klarälven Edsforsen	6659630	417949	60.0655849231	13.5259636573	140	217
Klarälven Norra Råda	6652237	421507	59.9999184059	13.5926853139	140	217
Ljungan Skallböleforsen	6916297	601468	62.364245471	16.9609179698	149	230
Ljusnan Funäsdalen	6938340	374935	62.5547598135	12.5674703455	150	213
Ljusnan, Ljusne Strömmar	6788266	611834	61.2127520756	17.0818959299	144	231
Lögde älv Lögdeå	7054698	719250	63.5523248759	19.4147093902	154	240
Lule älv Luleå	7293472	822571	65.6013105523	22.0096502867	162	251
Lyckebyån Lyckeby	6228390	541157	56.1986758161	15.6633201787	124	255
Nissan Halmstad	6285064	369774	56.6914832911	12.8736564378	126	214
Öre älv Torrböle	7072014	727041	63.7023261606	19.5960070674	154	241
Pite älv Bölebyn	7265328	792434	65.3790048926	21.2988922532	161	248
Råne älv Niemisel	7339849	815475	66.0213708746	21.9681884808	164	250
Rickleån Robertsfors	7132319	782392	64.2007769325	20.8214427365	156	246
Sävjaån Ingvasta	6656120	658796	60.0115727719	17.8485268758	140	234
Skellefte älv Slagnäs	7287288	644564	65.6748618979	18.146225083	162	235
Skellefte älv, Kvistforsen	7191952	774344	64.7397345721	20.7679167116	158	246
Svedån Sved	6431712	448738	58.0240046135	14.132162419	132	219
Töre älv Infl.Bölträsket	7334144	846887	65.9378769063	22.6395412478	163	253
Torne älv Mattila	7336315	915269	65.8754685603	24.1310357591	163	259
Upperudsälv. Köpmannebro	6518055	355776	58.7783294865	12.5053229998	135	213
V. Dalälven Mockfjärd	6705176	494341	60.4827108857	14.8970400362	141	222
Vindelälven Maltbrännan	7168178	705435	64.5762116502	19.291146819	158	240

Appendix D








































































































Appendix E



































































































Appendix F




































Table F1. Variation of GDDs (descending order) as difference between the average number of GDDs for 1950 – 1959 and for 2011 – 2020. The mean number of GDDs for both reference periods was calculated on the annual summary of GDDs ($T \ge 5^{\circ}$ C). In yellow the river stations that displayed the steepest relative annual Tot-P declines ($\ge 2.00 \% y^{-1}$) between 1980 – 2020.

Station name	GDDs 1950 – 1959	GDDs 2011 – 2020	A GDDs
Lyckebyån Lyckeby	2470	3174	704
Gullspångsälv. Gullspång	2320	2834	514
Nissan Halmstad	2621	3121	501
Vindelälven Maltbrännan	718	1155	437
Emån Emsfors	2554	2965	411
Upperudsälv Köpmannebro	2403	2805	402
Svedån Sved	2353	2744	391
Skellefte älv, Kvistforsen	852	1221	369
Lögde älv Lögdeå	1676	2033	357
Klarälven Almar	2383	2729	346
Öre älv Torrböle	1719	2063	344
Gide älv Gideåbacka	1767	2104	337
Sävjaån Ingvasta	2262	2589	328
Forsmarksån Johannisfors	2234	2547	313
Rickleån Robertsfors	1773	2082	309
Kalix älv Karlsborg	1595	1902	306
Töre älv Infl.Bölträsket	1619	1919	299
Ljusnan, Ljusne Strömmar	2107	2399	292
Råne älv Niemisel	1593	1884	291
Dalälven Älvkarleby	2178	2468	290
V. Dalälven Mockfjärd	1944	2224	279
Indalsälven Hammarstrand	1584	1856	273
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