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Widespread and persistent oligotrophication of northern rivers

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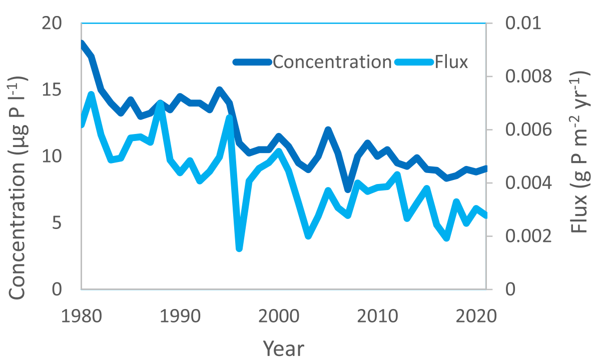
HIGHLIGHTS

- Phosphorus (P) concentrations have declined by close to half in many Swedish rivers
- P concentrations are now below reference (background) levels in many rivers
- Declining P concentrations may be related to increased forest growth
- A "less is better" paradigm is not always appropriate for nutrient management

GRAPHICAL ABSTRACT



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ABSTRACT

Phosphorus (P) is often a limiting nutrient in freshwaters and most management actions aim to reduce eutrophication associated with excess anthropogenic P inputs. Here, we report on the opposite problem, persistent and widespread oligotrophication (i.e., declining P concentrations) in northern rivers (56°-66° N) that appears unrelated to reductions in anthropogenic loading. Over the past forty years, P concentrations and fluxes in rivers draining forest dominated Swedish catchments have declined by nearly 50 %, with steeper declines in nutrient poor locations. Trends are negatively correlated with forest growth, temperature, pH and alkalinity. They are unrelated to trends in calcium, organic carbon and runoff. Declining P trends were strongest in locations draining catchments with shallow, nutrient poor soils and P concentrations in most locations are currently below estimated reference levels. These widespread and ongoing P declines highlight the need for new surface water management paradigms addressing the consequences of both nutrient scarcity and surplus.

1. Introduction

Phosphorus (P) and nitrogen (N) are the two main nutrients limiting aquatic productivity, with the former being more important in freshwaters. The vast majority of water quality policy and management activities including the UN Sustainable Development Goals (SDG 14.1.1a; UNEP, 2021), the European Water Framework Directive (WFD; EC2000/60), US Total Maximum Daily Loads (TMDLs; Boyd, 2000) and

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Chinese Surface Water Quality Standards (Su et al., 2017) are predicated upon cultural oligotrophication, or reducing nutrient concentrations in receiving waters to an acceptable level through controlling anthropogenic loading. Oligotrophication can occur due to reductions in external inputs as well as through increased retention in the aquatic continuum. Declining P concentrations associated with measures designed to reduce anthropogenic loading are well documented (Anderson et al., 2005). Increased retention in the aquatic continuum through, e.g., aluminum addition to promote sediment P retention is common, especially in eutrophied urban lakes (Huser et al., 2016). However, declining surface water P concentration trends have also been observed in high latitude northern catchments that do not have any significant changes in anthropogenic nutrient inputs, land management or land cover. Locations where declining P concentration trends have been reported include Swedish arctic lakes (Nilsson, 2022) as well as lakes in catchments where forestry is the major land use activity (Huser et al., 2018). An extensive Norwegian lake survey documented statistically significant total P (TP) declines in the most northerly lakes (de Wit et al., 2023), a finding corroborated by Huser et al. (2022). Declining P concentration trends have also been reported for headwater forest streams in Canada (Eimers et al., 2009) and Sweden (Mosquera et al., 2022).

Declines in riverine P concentrations can be driven by several mechanisms including reduced supply from the catchment, dilution due to higher flows as well as increased retention in the aquatic continuum. Reduced supply from the catchment can be related to lower external inputs (e.g., reductions in anthropogenic loading) or greater terrestrial retention. External inputs include atmospheric deposition (Tipping et al., 2014), mineral weathering (Tuyishime et al., 2022), point sources (Tong et al., 2017) and diffuse sources (McCrackin et al., 2018). There is little evidence for trends in atmospheric P deposition globally (Tipping et al., 2014) but there is some evidence for increasing P deposition in Europe in the early part of this century (Pan et al., 2021). However, deposition seems to be disconnected from observed surface water P declines (Eimers et al., 2018; Huser et al., 2018). Declines in mineral weathering inputs could lead to reduced P inputs as much of the P in forest soils is derived from in-situ mineral weathering (Tuyishime et al., 2022). Weathering rates are influenced by elemental composition and mineralogy of the parent material, with higher P concentrations increasing the potential for release. Mineral weathering rates are positively related to temperature. Warmer air temperatures will lead to warmer soils (Jungqvist et al., 2014) and warmer soils should have faster weathering rates (Kronnäs et al., 2023).

Increased in-catchment retention may cause surface water oligotrophication through a reduction in P supply to receiving waters. Retention can occur in catchment soils, vegetation or headwater ponds. Changing soil P retention may be caused by increased binding capacity associated with recovery from acidification (Kopacek et al., 2015) as well as warmer temperatures leading to more efficient P binding (Tian et al., 2023). However, there is little evidence that declining P export is associated with either of these processes. Changes in soil chemistry associated with recovery from acidification have been reported to be unrelated to P declines (Baker et al., 2015) or linked to increased P exports (Kopacek et al., 2015). Warmer soils can have higher amounts of plant available P (Vincent et al., 2014) and export more P to receiving waters (Scholz and Brahney, 2022). However, warmer soils can also bind P more tightly through increased sorption to more recalcitrant soil P fractions including iron oxyhydroxides and clays, thereby reducing bioavailable P in soil solution (Tian et al., 2023).

Increases in terrestrial plant biomass associated with forest management or a warmer climate may lead to declining P fluxes to receiving waters. The so-called “greening of the landscape” (Finstad et al., 2016), in which warmer temperatures promote longer growing seasons and higher primary productivity, may also increase P retention through increased biomass accumulation. Stoichiometric arguments suggest there should be an increase in terrestrial P retention if there is an increase in plant biomass. In addition to landscape greening, the rate of biomass accumulation in Swedish forests is increasing in all parts of the country except the far south (Fig. SI 4; Petersson et al., 2022). Increasing forest growth entails more storage of other elements including N (Lucas et al., 2016) and base cations (Karltun et al., 2022) in standing and harvested biomass. Increased water holding in the landscape due to beaver re-colonization, which is occurring in much of Sweden (Willby et al., 2018) and associated P retention due to sedimentation in the small ponds they construct is another possible factor influencing downstream oligotrophication (Crossman et al., 2016; Ecke et al., 2017). Increases in surface water iron (Fe) concentrations have been observed at multiple locations in Sweden (Temnerud et al., 2014) and across the northern hemisphere (Björnerås et al., 2017). Increasing Fe concentrations, which are correlated with increasing dissolved organic carbon (DOC) concentrations (Evans et al., 2024), may lead to increased precipitation of P (Hu and Huser, 2014; Huser et al., 2018).

At local (catchment) scales, oligotrophication can be connected to reductions in nutrient supply through, e.g., changes in land use (de Wit et al., 2020), control of point sources (Anderson et al., 2005; Tong et al., 2017), changes in internal cycling (Evans et al., 2011) and increased retention in upstream water bodies (Stockner et al., 2000). At a wider regional scale, candidate drivers of oligotrophication include climate change, recovery from acidification and surface water browning. The climate is changing. Warmer air temperatures are being reported across Europe (Twardosz et al., 2021). Since 1950, precipitation has increased in much of northern Europe, with some of the strongest trends reported for Sweden (Teuling et al., 2019). Furthermore, much of southern Sweden is recovering from the legacy of acidification (Futter et al., 2014). The geochemical consequences of this recovery may lead to increased leaching of P from the catchment due to decreased soil P binding capacity associated with increasing soil pH, increased DOC fluxes (Futter et al., 2014; Monteith et al., 2007), and decreasing in-lake immobilization as less aluminum is flushed from the catchment, and it will be less soluble as the pH of receiving waters increase (Kopacek et al., 2015). Increasing DOC fluxes have been shown to be correlated with increased Fe fluxes (Temnerud et al., 2014; Björnerås et al., 2017; Evans et al., 2024), which may lead to increased rates of P precipitation.

Changes in aquatic P retention may occur through increased rates of sedimentation (Gonzalez Rodriguez et al., 2023) that can be associated with longer water residence times as well as more effective P retention in sediment (Tammeorg et al., 2022). While sedimentation is most often associated with lentic systems, it can also be important in lotic environments (Extence et al., 2013). Aquatic P retention can also change due to geochemical factors including, e.g., recovery from acidification (Hu and Huser, 2014; Huser and Rydin, 2005) or management actions such as aluminum treatment to control eutrophication (Huser et al., 2016). Damming for hydropower production may also lead to progressive oligotrophication through changes in sedimentation rates and in-reservoir nutrient cycling (Stockner et al., 2000).

Changes in hydrology could affect P concentrations even if the mass of P transported is unchanged, e.g., changes in precipitation could alter rates of P flushing (Isles et al., 2023) and increased flows could cause dilution and concentration declines (de Wit et al., 2016). However, other studies have shown P concentration increases under increasing flows (Meyer and Likens, 1979).

Here, we analyze temporal trends and correlates of riverine TP concentrations measured between 1980 and 2021 draining forest dominated Swedish catchments with limited point source (urban) and agricultural nutrient sources (Suppl. Table 1). We assess correlations between TP concentration trends and: (i) trends in forest productivity, (ii) climate change, (iii) recovery from acidification, (iv) trends in other water quality variables and (v) underlying effects of catchment surficial geology. Finally, we discuss management and policy consequences of the observed trends and correlated factors.

2. Data and Methods

2.1. Data sources

We analysed long-term water chemistry measurement time series data (1980–2021) from the Swedish national surface water monitoring program (Fölster et al., 2014). All water chemistry measurements were made by trained samplers and analysed using SWEDAC accredited methods. Detailed method descriptions can be found at https://www.slu.se/en/departments/aquatic-sciencesassessment/laboratories/vattenlabb2/.

All candidate locations were sampled monthly. The original data set included 90 rivers and streams. Using criteria from Huser et al. (2018), any location draining a catchment with >5 % agricultural or 1 % urban land cover was excluded from the analysis. For each candidate location, the number of samples per year was enumerated and any year having ten or fewer samples was excluded to reduce potential bias associated with missing samples. In most cases, (15,661/16247 or 95 %) there was one TP measurement per month. Finally, any sampling location having five or more missing years between 1980 and 2021 was excluded.

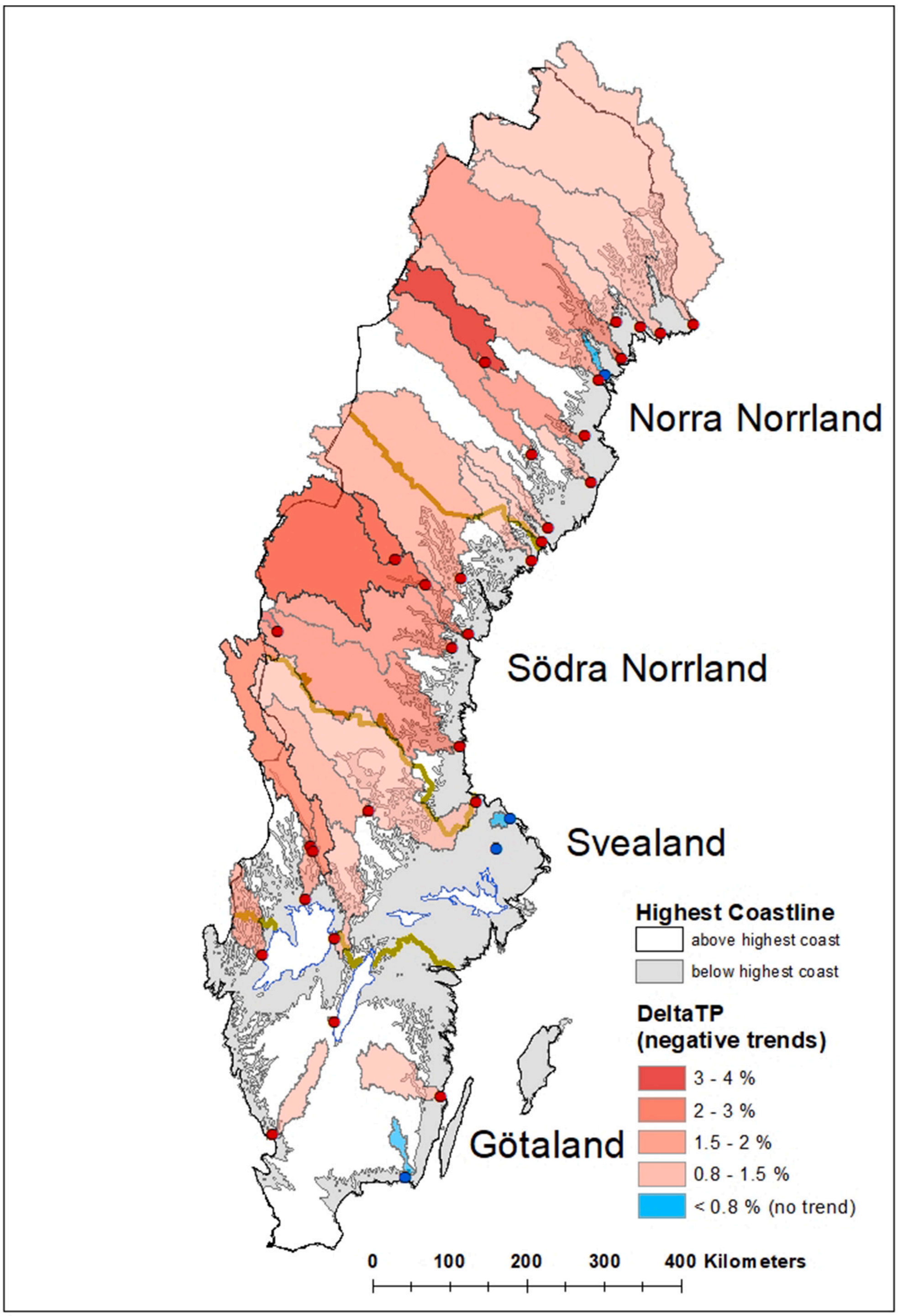

Fig. 1. Map of Sweden showing sampling stations (red or blue dots), catchments, TP trends, borders of forest regions and the highest coastline. Red represent catchments with negative TP trends and blue catchments without a significant temporal TP trend. Grey shows the areas situated below the highest coastline.

The final data set consisted of 34 river and stream locations (Fig. 1 and Suppl. Table 1). Sampling sites were located between 56.2° N and 65.9° N and 12.5° to 24.1° E (Fig. 1). Catchments are located in Köppen climate zones DfC (Subarctic), DfB (Warm summer humid continental) and CfB (Oceanic). Land cover is characterized by managed production forests in the central and southern regions and a mix of forest and bare land in the north. Sites were located in climatic regions 1 to 4 (Lindström and Alexandersson, 2004) and the nemoral to the northern boreal vegetation zones (Moen and Lillethun, 1999). Catchments ranged in size from 4 to 29,030 km^2, with a median area of 2951 km^2. Forests covered between 30 and 92 % of the catchment areas, with a median value of 72 %. Median agricultural and urban land cover were 1.0 % and 0.1 % respectively (Suppl. Table 1). The remainder of the catchment area was either bare rock or unproductive forest. Only four of the locations are on free flowing (undammed) rivers, the remainder have one or more dams for hydroelectricity generation.

We retrieved water chemistry measurements for pH, alkalinity (Alk), calcium (Ca) and total organic carbon (TOC). Alkalinity and pH are considered proxies for recovery from acidification while TOC is used a proxy for dissolved natural organic matter (DNOM), including co-transported Fe. Calcium was included as it has been suggested as an indicator of recovery from acidification, forestry impacts and weathering (Bergström et al., 2024).

Climate related variables included average daily air temperature, growing degree days (GDD), precipitation and runoff. Air temperature and precipitation were obtained from the 0.25° gridded EOBS product version 27.0e (Cornes et al., 2018; retrieved 17/8/2023). Annual average air temperature was calculated as the average of daily average temperatures for the year. GDD were calculated as annual temperature sums of the average daily EOBS temperature data. Days were included in the GDD summation when the average daily temperature was ≥ 5 °C. Annual precipitation was calculated as the sum of daily precipitation values. Monthly runoff data were obtained from the ERA5 data product (Hersbach et al., 2023; retrieved 7/7/2023). Annual runoff values were calculated as the sum of monthly values. For both EOBS and ERA 5 data, grid cells containing monitoring location coordinates were used.

Forest growth data were obtained from the Swedish National Forest Inventory (NFI; Fridman et al., 2014; retrieved 1/9/2024). Annual growth estimates, which are the sum of increment in standing volume and removals due to harvesting, were available for four regions (Fig. 1) from 1987 to 2018 (Fig. S4). Annual values reported in the NFI are five year averages centred on the reported year (e.g., 1987 is the average growth between 1985 and 1989). River monitoring locations were assigned to the forest growth region containing the majority of the catchment.

Catchment surficial geology was divided into two classes depending on whether the location was above or below the highest glacial coastline (Mörner, 1979). The highest glacial coastline demarcates the boundary between land that was never below sea level during the last glaciation and areas with a surficial geology characterized by clays derived from marine sediments. Such soils typically have high P concentrations (Skarbøvik et al., 2020) and/or lower levels of amorphous aluminum, which are correlated with high release rates and/or limited P binding ability (Møller et al., 2023). Soils above the highest glacial coastline are derived from slow weathering, alumina-silicate bedrock. These young (<13,000 year old), post-glacial soils generally contain lower amounts of P and have a relatively high P binding ability (Tuyishime et al., 2022) compared to soils from areas below the highest glacial coastline. Catchment percent area above the highest coastline was estimated by overlaying the two maps using ArcGIS.

2.2. Calculated values

Monthly P fluxes (g P m^{-2} yr^{-1}) were estimated by multiplying modelled monthly ERA5 runoff by monthly TP concentration. For years where TP concentration measurements were not available for all months, missing values were estimated as the arithmetic average of measurements for that year. Annual fluxes were calculated by summing monthly values. Flow weighted annual average TP concentrations were calculated as annual flux divided by annual runoff.

Reference condition TP concentrations (TP_{Ref}) were calculated using a relationship documented by Skarbøvik et al. (2020; their equation SI 2)

$\log10(TP_{Ref}\ (\mu g\ l^{-1})) = 1.533 + 0.240 \bullet \log10(Ca^*Mg^*\ (meq\ l^{-1})) + 0.301 \bullet \log10(AbsF) - 0.012 \bullet \mathrm{sqrt}(Altitude\ (m\ a.s.l.))$

Non-marine calcium and magnesium (Ca*Mg*) were derived from relationships developed by Henriksen and Posch (2001):

$Ca^*Mg^*\ (meq\ l^{-1}) = Ca\ (meq\ l^{-1}) + Mg\ (meq\ l^{-1}) - 0.235 \bullet Cl\ (meq\ l^{-1})$.

Where Ca, Mg and Cl are the concentrations of calcium, magnesium and chloride respectively reported as charge equivalents. AbsF is the absorbance at 420 nm over 5 cm for a filtered water sample (more information is available at https://www.slu.se/en/departments/aquatic-sciencesassessment/laboratories/vattenlabb2/). The final term in the equation is the elevation of the monitoring site, expressed as metres above sea level (m a.s.l.). In the analysis presented here, we assumed that all sites had an elevation of 100 m a.s.l.

Reference TP concentrations (TP_{Ref}) for each location were calculated using site-specific arithmetic averages of Ca, Mg, Cl and AbsF measurements made between 1995 and 1999.Ecological quality ratios (EQRs) were calculated as site-specific TP_{Ref} concentrations divided by the arithmetic average TP concentrations for the preceding three years, e.g., EQR(2018) is based on measurements made in 2016, 2017, 2018. The WFD defines the EQR boundary between good and moderate status as 0.5 and 0.7 for the boundary between good and high status (SEPA, 2007, table 10.1). Any EQR values >1.0 imply current concentrations less than those expected under reference conditions

2.3. Statistical analyses

All water quality trend analyses were conducted using annual median concentrations to avoid potential interference resulting from varying number of samples among years or rivers. Annual sums were used for assessing trends in precipitation, runoff, fluxes and GDD. All statistical analysis were executed using the R programming language v4.3.1.

Temporal trends in forest growth rates, TP, Ca, pH, Alk, TOC, precipitation, air temperature, and GDD were evaluated using the non-parametric Mann-Kendall test (Mann, 1945; Kendall, 1975). Annual rates of change (% yr^{-1}) were calculated by dividing the Theil-Sen's slope (unit yr^{-1}; Sen, 1968) by the median value (unit) for the whole time period. Both the Mann-Kendall test and the Theil-Sen's slope were calculated using the rkt-function in the rkt package in R 4.3.1 (https://cran.r-project.org/web/packages/rkt/rkt.pdf).

To elucidate potential environmental correlates to any trends in TP concentration, Pearson correlation coefficients (two-tailed) were calculated between time series of annual median TP and the following candidate explanatory series: annual forest growth, median annual Ca, Alk and TOC concentrations, annual median pH, annual precipitation, annual runoff, annual GDD and annual average air temperature.

To adjust for multiple comparisons, significance was assessed using the following heuristic. Nominal (reported) *p*-values were divided by the number of tests reported in a table, i.e., if a table contained 10 results, significance was assessed as $p < 0.005$ (0.05/10).

3. Results and discussion

3.1. Observed trends in total P and potential causal factors

Declining TP concentrations over the study period were observed at all locations (Fig. 1, Suppl. Fig. 1, Suppl. Table 2). After adjusting for multiple tests, Mann Kendall trend statistics were significant in 29 of 34 cases. Declines ranged from −4.4 % to −0.1 % yr^{-1}. The strongest, relative trends were seen at sites with lower median (1980–2021) TP concentrations (Fig. 2a). In 1980, the median TP concentration for all sites was close to 20 μg l^{-1}. From the mid-1980s to 1995, median concentration decreased to slightly <15 μg l^{-1}, and from 1996 and 2010 the median concentration was slightly >10 μg l^{-1}. Since 2010, median TP concentrations have been at or below 10 μg l^{-1} (Fig. 3), with concentrations in some rivers having reached detection limits for phosphorus (1 μg L^{-1}). Similar temporal patterns were seen with flow weighted TP concentrations (Suppl. Fig. 3). Differences in percentage declines for TP concentrations among sites mask the fact that most rivers show similar absolute declines over the study period (−0.1 to −0.2 μg L^{-1} yr^{-1}) (Fig. 2b).

The six rivers with highest median TP concentrations over the entire study period had either no significant decrease in concentration over time, or a stronger absolute decrease than rivers with a lower median concentration, potentially indicating an effect of underlying geology. Four of the aforementioned rivers had the highest proportion of their catchment below the highest glacial coastline (i.e., > 75 %, Suppl. Table 1, Fig. 1). Soils below the highest glacial coastline are of marine origin with high P concentrations (Skarbøvik et al., 2020) and/or lower levels of amorphous aluminum with limited P binding ability (Møller et al., 2023). Either of these factors would be consistent with higher surface water TP concentrations than those expected in catchments above the highest coastline where soils which have developed since the last glaciation are derived from slow weathering, alumina-silicate bedrock. These soils, which are typical for the Swedish forest landscape, generally contain lower amounts of P and have a relatively high P binding ability (Tuyishime et al., 2022).

Annual P fluxes decreased over time in all cases, ranging from −6.02 % and − 0.06 % yr^{-1}, and were significant at 22 of 34 locations after adjusting for multiple tests (Fig. 3, Suppl. Table 2). Median P-flux has declined from slightly >6 mg m^{-2} yr^{-1} in the 1980s to <3 mg m^{-2} yr^{-1}

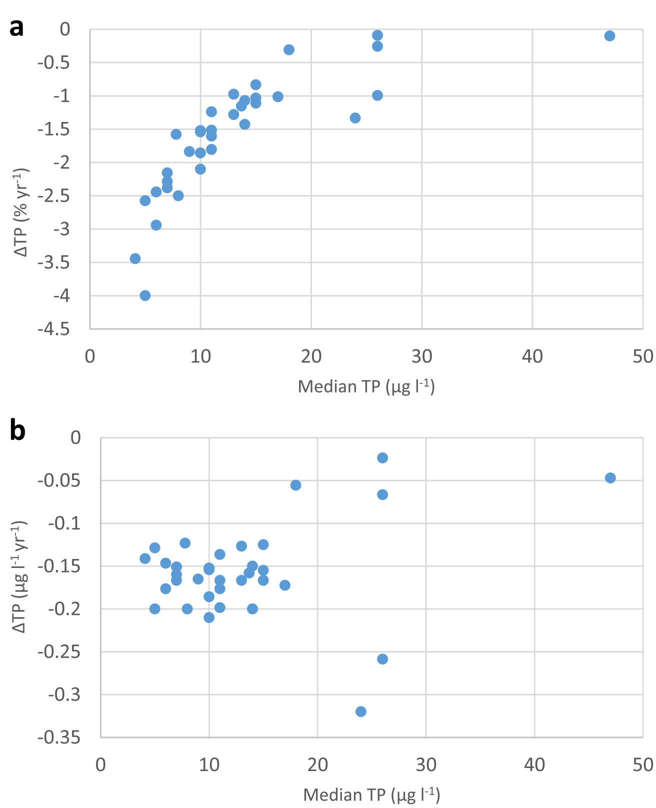

Fig. 2. The relationship between the annual rate of change in TP (ΔTP) and the overall median TP concentration for all the monitoring sites. Panel (a) shows the relative rate of change, whereas panel (b) shows the absolute rate of change.

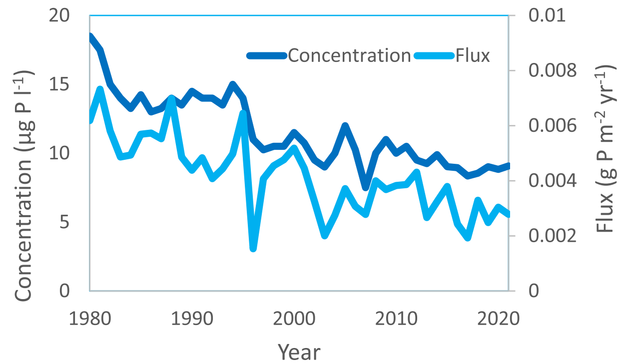

Fig. 3. Annual median Total phosphorus (TP) concentration (dark blue) and median TP-flux (light blue) over time for 34 forest-dominated Swedish river monitoring sites.

in 2021 (Fig. 3, Suppl. Fig. 2).

Land management, global and regional environmental change (i.e., forestry, climate change, browning and recovery from acidification) were evaluated as possible correlates for the observed TP declines during the study period. Forest growth rates increased in the northern and central parts of Sweden (Suppl. Fig. 4). There was no evidence of a monotonic temporal trend in southern Sweden (Götaland). Overall, growth increments were higher in central Sweden (Svealand and Södra Norrland) than in the most northerly parts of the country (Norra Norrland).

Annual average air temperature and number of GDD increased at all locations during the study period (Suppl. Table 3). There was no nominally significant ($p \leq 0.05$) temporal trend in precipitation at 29 of the 34 locations. Of the remaining five locations, precipitation increased at four and declined at one. A similar pattern was seen for runoff, which increased in two locations and displayed no trends in the remaining 32 sites between 1980 and 2021. For the 26 locations where TOC data were available, increasing temporal trends were seen at 16 sites and there were no trends at the other ten. During the study period, alkalinity and pH increased at 23 and 18 locations respectively. With the exception of one nominally significant decreasing trend in pH, there were no other significant trends for these variables.

Pearson correlations between time series of annual TP concentrations and annual values for the relevant time series (Table 1) identified possible environmental correlates for the observed temporal TP trends. Air temperature and TP concentration time series correlated negatively

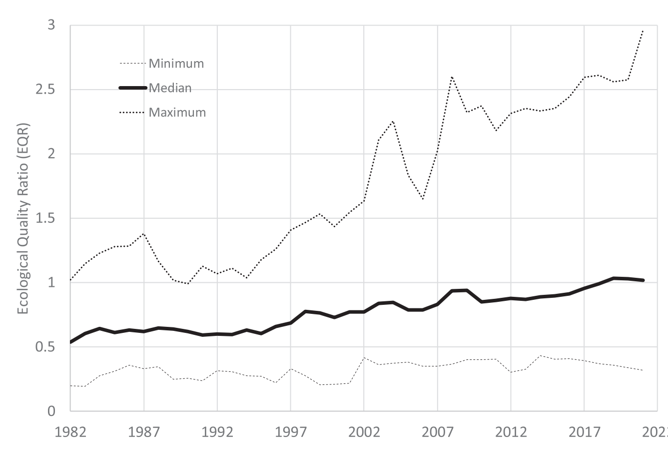

Fig. 4. Annual Minimum, median and maximum Ecological Quality Ratios (EQRs;reference TP/observed TP) for 34 forest dominated Swedish river monitoring sites. Values greater than one are indicative of lower nutrient levels than expected under background (reference) conditions.

Table 1
Pearson correlations between annual total P concentrations and candidate predictors. Candidate predictors include annual air temperature (T, °C), annual growing degree days (GDD; cumulative sum), annual precipitation (P; mm yr^{-1}), runoff (Q, m m^{-2} yr^{-1}), annual average alkalinity (Alk; meq l^{-1}), annual average pH, annual average total organic carbon (TOC; mg l^{-1}) and annual forest growth increment for the relevant region (m^3 SK ha^{-1} yr^{-1}). Correlations significant at the p<1.7E-04 (0.05/298) are highlighted in bold.

Location	T	GDD	P	Q	Alk	Ca	pH	TOC	Forest Growth
Råne älv Niemisel	−0.46	−0.37	−0.32	−0.21	0.10	0.38	0.10	−0.43	**−0.67**
Töre älv Infl.Bölträsket	−0.43	−0.24	−0.27	−0.23	0.36	**0.54**	0.03		−0.49
Torne älv Mattila	−0.28	−0.19	−0.04	−0.04	−0.13	0.01	−0.29		−0.51
Kalix älv Karlsborg	−0.46	−0.48	−0.01	0.02	−0.30	−0.30	−0.42	0.16	−0.54
Skellefte älv Slagnäs	−0.45	−0.34	−0.18	−0.12	**−0.67**	−0.15	−0.43		**−0.88**
Lule älv Luleå	**−0.55**	−0.53	−0.33	−0.14	**−0.60**	−0.43	−0.60	−0.03	**−0.87**
Alterälven Norrfjärden	−0.21	0.24	−0.32	−0.09	0.24	0.09	0.22	0.00	−0.26
Pite älv Bölebyn	−0.51	−0.31	−0.28	0.05	−0.41	−0.10	−0.16	0.09	**−0.71**
Skellefte älv. Kvistforsen	**−0.62**	−0.49	−0.11	−0.08	−0.34	−0.07	−0.46		**−0.89**
Vindelälven Maltbrännan	−0.49	−0.38	−0.06	−0.02	**−0.54**	−0.15	−0.49		**−0.89**
Rickleån Robertsfors	**−0.61**	−0.53	0.04	0.04	**−0.55**	−0.09	**−0.54**	−0.29	**−0.61**
Öre älv Torrböle	−0.38	−0.36	0.26	0.16	−0.15	0.02	−0.46	−0.11	−0.45
Lögde älv Lögdeå	−0.26	−0.11	0.15	0.18	−0.30	−0.16	−0.35	0.00	−0.43
Ammerån Skyttmon	−0.36	−0.34	0.10	0.11	−0.12	0.09	−0.47	−0.19	**−0.79**
Gide älv Gideåbacka	−0.54	**−0.58**	−0.03	0.05	−0.14	0.43	0.26	−0.41	**−0.71**
Ångermanälven Sollefteå	−0.52	−0.30	0.04	0.24	**−0.56**	0.15	−0.39	−0.01	−0.59
Indalsälven Hammarstrand	−0.44	−0.43	0.00	0.08	**−0.61**	−0.34	−0.31	−0.05	**−0.81**
Ljusnan Funäsdalen	−0.52	−0.37	−0.08	0.00	−0.02	0.29	−0.14	−0.19	**−0.87**
Indalsälven Bergeforsen	−0.48	−0.41	−0.16	0.14	−0.49	−0.18	−0.38	−0.11	**−0.83**
Ljungan Skallböleforsen	−0.46	−0.38	−0.06	0.16	**−0.71**	−0.31	−0.36	−0.04	**−0.81**
Ljusnan. Ljusne Strömmar	**−0.58**	**−0.57**	0.10	0.28	**−0.62**	−0.52	**−0.55**	−0.22	**−0.70**
Dalälven Älvkarleby	−0.26	−0.20	0.31	0.25	**−0.59**	0.27	**−0.65**	−0.29	**−0.60**
V. Dalälven Mockfjärd	−0.53	−0.47	0.05	0.20	−0.36	0.06	−0.47		**−0.61**
Forsmarksån Johannisfors	−0.40	−0.27	0.48	0.43	0.09	0.05	**−0.65**	0.19	−0.07
Klarälven Edsforsen	−0.31	−0.35	−0.01	−0.02	−0.33	−0.19	−0.36	−0.42	**−0.72**
Sävjaån Ingvasta	−0.02	−0.05	0.02	−0.01	−0.03	−0.10	−0.04	0.09	−0.16
Klarälven Norra Råda	−0.28	−0.21	−0.09	−0.07	**−0.59**	−0.47	**−0.62**	−0.44	−0.60
Klarälven Almar	−0.29	−0.30	−0.03	0.19	**−0.71**	−0.34	**−0.67**	−0.34	−0.57
Gullspångsälv. Gullspång	−0.38	−0.40	−0.16	0.16	**−0.81**	0.52	−0.48	−0.23	−0.52
Upperudsälv. Köpmannebro	−0.45	−0.53	−0.15	−0.15	−0.41	**0.74**	−0.30		**−0.69**
Svedån Sved	−0.51	**−0.55**	0.07	−0.06	−0.35	0.13	−0.10		−0.59
Emån Emsfors	−0.54	**−0.58**	0.19	0.27	**−0.64**	−0.08	−0.41	0.13	−0.05
Nissan Halmstad	−0.43	−0.50	−0.05	−0.07	−0.46	0.09	−0.60	−0.52	−0.51
Lyckebyån Lyckeby	−0.13	−0.08	0.13	**0.53**	−0.37	−0.35	−0.34	**0.63**	0.17

at all locations while total P and GDD correlated negatively at all but one location (Table 1). Precipitation time series also correlated negatively with TP concentration time series at 22 of 34 locations (Table 1). Correlations between time series of runoff volume and TP were negative at 15 locations, but were positive at the other 19. The lack of a clear connection between temporal patterns in TP concentration and runoff suggests that dilution caused by increased runoff is not likely a factor in the observed concentration decline during the study period. Together with the decreasing TP fluxes, it seems more likely that increased terrestrial retention, caused by, e.g., "greening of the landscape" (Finstad et al., 2016), and increased forest biomass (Lucas et al., 2016) could be connected to the observed oligotrophication trends during the study period.

Regional environmental changes may also cause decreasing surface water TP concentrations. Browning, which may lead to increased surface water Fe concentrations (Evans et al., 2024) as well as increased P retention in sediments (Tammeorg et al., 2022), is occurring across much of Sweden (Kritzberg et al., 2020). There is no clear evidence for a relationship between temporal trends in TOC (as a proxy for browning) and oligotrophication in Swedish rivers as TOC concentrations increased at 16 of 26 locations (Suppl. Table 3), and time series of TOC and TP concentrations were negatively correlated at 19 of 26 locations (Table 1). Much of Sweden was acidified in the 1980s (Futter et al., 2014; Fölster et al., 2014) and the subsequent recovery may have influenced riverine TP concentration trends. Geochemical changes associated with recovery from acidification such as decreasing Al concentrations should decrease aquatic P retention (Hu and Huser, 2014; Huser and Rydin, 2005) and all else being equal, lead to increasing TP concentrations. Correlations between time series of both pH and alkalinity with TP were negative at 30 of 34 locations (Suppl. Table 3). However, correlations were strongest in locations with high overall average pH or alkalinity (not shown). This, along with the lack of agreement with expected changes in TP concentrations during recovery (Kopacek et al., 2015), suggests the observed TP concentration trends are unlikely to be related to recovery from acidification (Table 1). Calcium concentrations decreased in 18 of 34 locations (Suppl. Table 3), and were negatively correlated with TP concentrations in 18 cases (Table 1). Because correlations were both positive and negative, and varied considerably in strength, it is unlikely that temporal trends in calcium concentration due to, e.g., recovery from acidification (Futter et al., 2014), intensified forestry (Bergström et al., 2024) or permafrost melting (Huser et al., 2022) are relevant proxies for environmental changes linked to ongoing oligotrophication.

To conclude, observed temporal trends in TP concentrations and fluxes are correlated most strongly with regional patterns of increased forest growth during the study period. These increases in terrestrial plant biomass are likely due to a combination of forestry practices and a warmer climate. Declines in TP correlated with both increasing trends in air temperature and growing degree days during the study period. There was no evidence to support flow dilution or increased precipitation as mechanisms to explain the observed TP declines. Increasing alkalinity and pH were negatively correlated with TP declines, there was little or no detectable relationship between temporal patterns in either calcium or TOC and TP. Effects were most pronounced in catchments located above the highest glacial coastline where soils typically have low P release rates.

3.2. Policy implications

Ongoing surface water oligotrophication has implications for water policy and management. In nearly all cases, water quality goals or guidelines for nutrients are based on a "less is better" paradigm. The possibility that phosphorus levels can be too low to support healthy aquatic ecosystems has, to the best of our knowledge, not previously been considered in management decision making. Instead, management actions focus almost exclusively on reducing nutrient levels in anthropogenically eutrophied systems. In Europe, the management goal is to move nutrient levels closer to reference conditions, which describe the situation which would be expected when there is minimal anthropogenic disturbance (Skarbøvik et al., 2020). In Europe, WFD management targets for water quality (EC2000/60) are based on ecological quality ratios (EQR). An EQR is the ratio of reference conditions values divided by present-day values. EQRs range from 0 (worst possible conditions) to 1 (best possible condition). Implicitly, there is no consideration of EQR values greater than one where observed concentrations are less than those expected in a situation of minimal anthropogenic disturbance. To the best of our knowledge, no existing management paradigms give serious consideration to the meaning or consequences of EQR values >1, which would occur as a consequence of, e.g., TP concentrations declining below reference condition values. EQR values in this study showed a monotonic increase between 1982 and 2021 for all sites (Fig. 4). In 1982, one location had an EQR >1. By 2021 this had increased to 17 of 34 sites. Fourteen locations failed to reach good status in 1982 (EQR <0.5), while this number declined to just one by 2021. From one perspective, the increase in number of locations with EQR values >0.5 is a success story, but it neglects the unknown but potentially severe impacts of excessive oligotrophication on aquatic ecosystem health.

Correlation is not causation. The fact that regional forest growth time series are the strongest correlates of declining riverine TP concentrations is not proof that forestry is causing the observed declines, but it is worthy of further consideration. Forest growth has been linked to delays in recovery from acidification (Karltun et al., 2022), declines in riverine inorganic N (Lucas et al., 2016) and has been suggested as a factor in surface water calcium declines (Bergström et al., 2024). In all these cases, the hypothesized mechanism is increasing element sequestration in forest biomass. It is noteworthy that similar TP declines have been observed in Arctic lakes with no forestry in their catchments (Huser et al., 2022; Nilsson, 2022).

The widespread and ongoing declines documented here are suggestive of increasing freshwater P limitation, which in turn will negatively impact ecosystem health and productivity, as well as freshwater ecosystem services. Biodiversity and general food web health are strongly connected to an intermediate nutrient regime (mesotrophy), suggesting that neither too much nor too little nutrients are desirable. At a number of locations reported herein, TP concentrations have already reached 1 $\mu g\ L^{-1}$ (a common analytical detection limit), and now require special methods just to detect these low levels of P that are increasingly common in the Swedish aquatic environment. Current water governance paradigms fail to adequately consider the possibility that nutrient levels can be too low, and thus offer no avenues to manage such systems, even if oligotrophication is caused by anthropogenic activity. Our results highlight the need to move beyond an exclusive "less is better" approach to water governance and develop sustainable management strategies that recognize that there can be too little as well as too much nutrients in surface waters.

Code statement

No unique code was used in this study.

Data statement

Data used in this study are available at https://doi.org/10.6084/m9.figshare.25330069.v1

CRediT authorship contribution statement

Jenny L. Nilsson: Writing – review & editing, Investigation, Formal analysis. **Sara Camiolo:** Writing – original draft, Investigation, Data curation, Conceptualization. **Brian Huser:** Writing – review & editing, Validation, Funding acquisition, Conceptualization. **Oskar Agstam-Norlin:** Writing – review & editing, Conceptualization. **Martyn Futter:** Writing – review & editing, Writing – original draft, Supervision, Funding acquisition, Conceptualization.

Declaration of competing interest

The authors declare the following financial interests/personal relationships which may be considered as potential competing interests: Martyn Futter reports financial support was provided by the Swedish Environmental Protection Agency. If there are other authors, they declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.scitotenv.2024.177261.

Data availability

https://doi.org/10.6084/m9.figshare.25330069.v1

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