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Research article

Water quality in a large complex catchment: Significant effects of land use and soil type but limited ability to detect trends

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ABSTRACT

Globally, significant societal resources are devoted to mitigating negative effects of eutrophication from excessive phosphorus (P) and nitrogen (N) loading. Potential effectiveness of mitigation measures and possible confounding factors are often assessed using studies conducted in headwater catchments. However, success is often evaluated based on trends in river mouth water chemistry. It is not clear how transferrable insights from headwater catchments are to larger rivers. Here, relationships between P and suspended solids (SS) identified in small agricultural headwater catchments were applied to 30 larger, mixed land use catchments draining into Mälaren, a Swedish great lake. Relationships identified in headwater streams between SS concentration, catchment agricultural land percentage and arable land clay content were corroborated for the larger catchments ($R^2 = 0.59$, p-value<0.001. The same was true for connections between SS and particulate P ($R^2 = 0.74$, p-value<0.001). This study highlights the importance of agricultural land, clay content and SS for P transport, on both smaller headwater as well as larger catchment scales, supporting the use of headwater findings on larger, management relevant scales. Consequently, these relationships should be used to target mitigation measures to reduce SS and P losses. To explore the effectiveness of mitigation measures on water quality, we assessed long-term (20 year) trends in tributary water quality and compared these trends to the amount of mitigation measures implemented in the catchment. Overall improving trends were detected using regional Mann Kendall tests, but few decreasing trends in nutrient concentrations were found for individual sites using Generalized Additive Models (GAM). The lack of significant trends and identifiable connections to amount of mitigation measures implemented could be due to several reasons, e.g. insufficient time for recently implemented measures to have an effect, ongoing release of legacy P as well as low areal coverage and poor spatial placement of implemented measures. In addition, trend detection requires large amounts of data and the results should be carefully interpreted and communicated.

1. Introduction

Eutrophication associated with exceedance of the planetary boundaries for biogeochemical flows of both phosphorus (P) and nitrogen (N) is an urgent global problem (Steffen et al., 2015). To avoid further acceleration of eutrophication, anthropogenic P and N exports need to be controlled (Steffen et al., 2015). Consequences of eutrophication including, e.g., toxic algal blooms, oxygen deficits and dead bottom waters (Smith and Schindler, 2009) have negative impacts on surface water quality. Good surface water quality is important for recreational values, drinking water quality and the health of aquatic ecosystems. Erosion and transport of soil particles can also contribute to impaired water quality by e.g. causing decreased light penetration (Bilotta and Brazier, 2008) and carrying other pollutants including pesticides (Boardman and Poesen, 2006), heavy metals (Kronvang et al., 2003) and P (Haygarth et al., 2006). Thus, actors from local to global scales need to work towards improving surface water quality. Significant resources have been and are devoted to implementing mitigation measures to achieve national and international water quality goals. However, local monitoring of mitigation measure efficiency is limited and improvements in water quality associated with mitigation efforts can be difficult to quantify.

Processes, trends and mitigation measure effectiveness are often studied on small headwater catchments and findings are later up-scaled

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to larger management-relevant scales (Kronvang et al., 2009; Sharpley et al., 2009; Kyllmar et al., 2014) but often without testing applicability. Headwaters are usually considered as the first influence on the surface water system (Bol et al., 2018) and typically show higher variability in nutrient concentrations than larger catchments (Dupas et al., 2023). Using more easily studied systems as proxies for other, more complex systems can be a way forward for management, but findings from headwater catchment studies must be tested in larger, multifaceted systems.

Previous studies have explored relationships between water quality and catchment characteristics. For example, Sandström et al. (2020) established a linear relationship between suspended solids (SS) losses and the product of clay content and the proportion of agricultural land based on monitoring data from 11 small (5.7–33.1 km^2) agricultural headwater catchments. They also showed a power-law relationship between particulate P (PP) and SS (Sandström et al., 2020). Similarly, Djodjic et al. (2021) showed a strong connection between the proportion of agricultural land in the catchment and P and N concentrations as well as decreased P and N concentrations with increasing forest and wetland coverage in 235 small (area < 50 km^2) headwater catchments. If specific land use and soil type characteristics can be associated with an elevated risk for P and N transport, these areas can be targeted for monitoring and mitigation measures.

Mitigation measures to limit nutrient export from Swedish agricultural land include mandatory (e.g., manure and fertilizer management, Swedish Board of Agriculture, 2019) and voluntary actions (e.g., agro-environmental schemes for wetlands, structural liming, two-stage ditches, buffer zones, etc., Bergström et al., 2015). Systematic monitoring and reporting on voluntary measures before, during and after implementation is rare, leading to limited management-relevant information on measure efficiency or effects on receiving waters nutrient concentrations. Quantification of measure effectiveness is typically made at a local scale (e.g., Weisner et al., 2016; Geranmayeh et al., 2018; Stutter et al., 2012). These assessments must then be up-scaled to the relevant management unit (e.g., headwater or larger catchment). However, a recent study by Tomczyk et al. (2023) found no evidence for significant effects of state-level policy implementation on decadal trends in nutrient concentrations.

Firstly, we explore whether headwater water quality relationships are applicable to larger rivers and if proportion of agricultural land, soil clay content and SS are driving factors for P transport on a larger catchment scale. Second, we evaluated long-term monitoring data collected from tributary rivers draining the catchment of Mälaren, a Central Swedish great lake where diffuse losses from agriculture now account for 64% of the total P load to the lake (Mälarens vattenvårdsförbund, 2021). Temporal trends in relevant water quality parameters over the past 20–25 years, three decades after the substantial positive effects of the large point source reductions were achieved (Persson, 2001), were assessed using the same monitoring data. Third, all existing reports on mitigation measures implemented on arable land in the Mälaren catchment were collected to explore possible relationships with water quality. The combination of these three approaches gives unique insights into transferability between scales and water chemistry trends in the area.

2. Methods

2.1. Catchment properties

All catchments are located in the Norrström basin (22,600 km^2, 59°30′N, 17°12′E), with 14 main tributaries discharging into Mälaren, a lake with great economic and recreational value (Sonesten et al., 2013). Tributary catchments have varying land use and soil type distributions, from forests with nutrient poor soils and peatland in the northwest to intensive agriculture on nutrient rich clay soils in the northeast (Table S1, Fig. 1). This study includes all main tributaries, along with 16

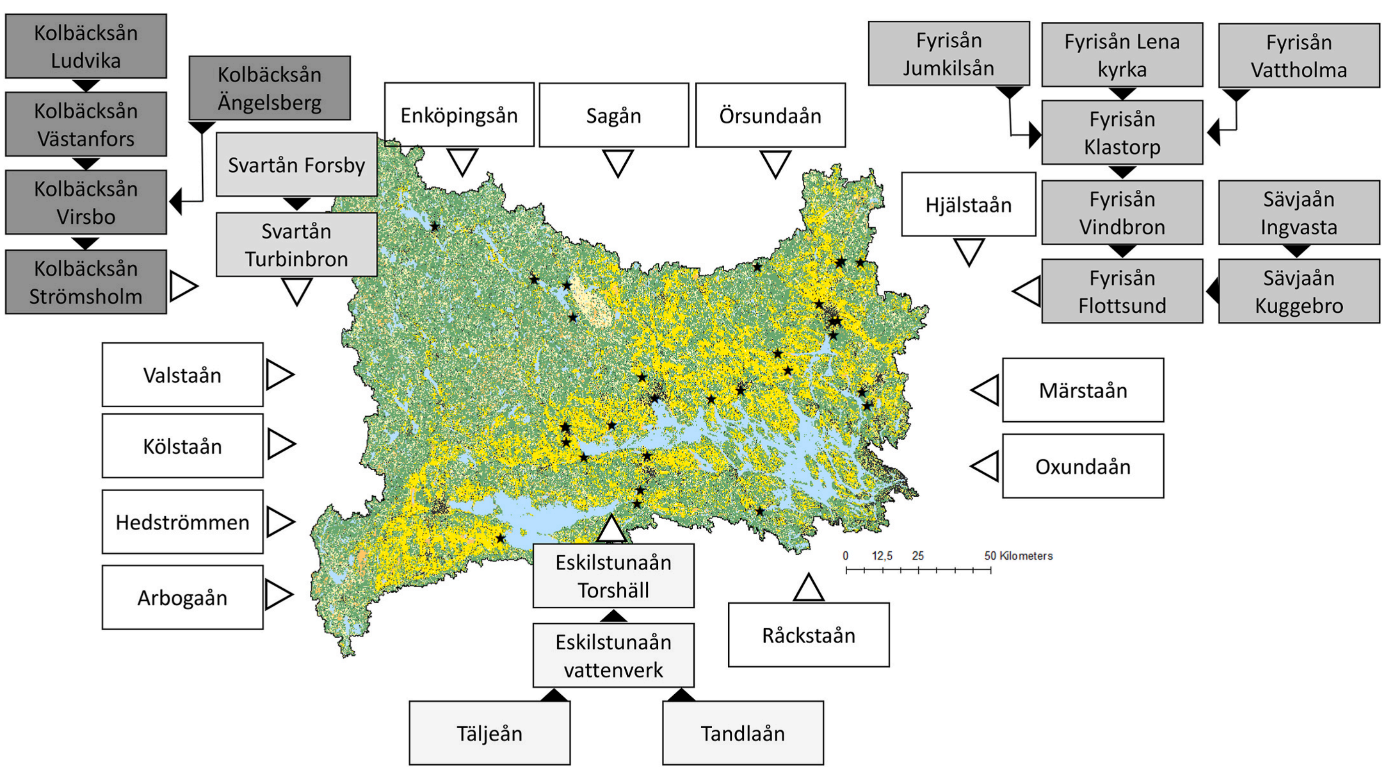

Fig. 1. Map of the Mälaren catchment, sampling stations and connectivity between different rivers and stations. The background map displays land use, and the stars identify sampling locations. White boxes represents streams with only one sampling location. Same shade of grey indicates sampling locations in the same river with arrows indicating connectivity between streams and sampling locations. (For interpretation of the references to colour in this figure legend, the reader is referred to the Web version of this article.)

additional stations, i.e., all sites in the Norrström basin included in the Swedish National Monitoring Programme (30 sites, Fig. 1; Tables S1 and S2) representing both smaller streams, and larger river mouths discharging directly into Mälaren (Fölster et al., 2014). Water chemistry has been monitored in Mälaren tributary mouths since 1965 (Persson, 2001).

In the 1970s wastewater treatment was improved and point source emissions were considerably reduced, resulting in significant decreasing total P (TP) trends in most larger streams discharging into Mälaren (Sonesten et al., 2013). Mitigation measures to reduce P and N transport from agricultural land are widely used in the catchment. However, eutrophication is an ongoing problem in Mälaren, partly due to incoming nutrient-rich water from agricultural areas, with many lakes and streams in the catchment failing to reach good ecological status based on current water chemistry (WISS, 2022).

2.1.1. Water chemistry, discharge and catchment data

Water quality parameters with monthly (or every other month) measurements between 1997 and 2020 were used. These included SS, TP, total N (TN), nitrate/nitrite (NO_2/NO_3–N hereafter called NO_3–N), ammonium (NH_4–N) and total organic carbon (TOC) (Miljödata-MVM, 2022). Water samples were analysed at the Swedish University of Agricultural Sciences (SLU) ISO/IEC 17025 accredited geochemistry laboratory.

Modelled flow from Swedish Meteorological and Hydrological Institute (SMHI) was used for load calculations since discharge was not monitored at all stations (SMHI, 2022a). Modelled flows were available from 2004, dictating the use of 2004–2020 for analysis of loads and flow weighted (fw) concentrations (see chapter 2.2). Measurements from 1997 to 2020 were used for analysis of raw concentrations. See Supplementary Fig. S1 for flow chart of methodology.

2.2. Data handling

Water quality observations below the reporting limit were set to half the reporting limit. Flow weighted (fw) mean values (2004–2020) were calculated following Linefur et al. (2019), where each raw sampled concentration was used to represent daily concentrations backwards to the previous sampling occasion, but for a maximum of two months. When there was more than two months between sampling, values were treated as missing and excluded from subsequent analyses. Estimated raw daily concentrations were multiplied with modelled daily flows to get daily loads (mg day^{-1}). Monthly fw concentrations (mg L^{-1}) were derived from monthly loads and flows and an overall mean fw concentration was calculated.

2.3. Statistical and trend analysis

Overall mean fw concentrations were used to evaluate applicability of headwater scale water quality relationships (Sandström et al., 2020) to larger, mixed land use catchments. A Principal Components Analysis (PCA) was performed to visualize relationships between water quality parameters and catchment characteristics. Multiple regressions for TP and SS (forward selection) were performed where parameters identified from the PCA were evaluated. Due to differences in analytic methods between Sandström et al. (2020) and this study, no further investigations of PP were possible. A factor of variation (FV) between different tributaries was calculated for all parameters as the ratio between the maximum total fw mean and minimum total fw mean value.

Trends were analysed in raw concentrations (1997–2020), monthly fw concentrations and monthly loads (2004–2020). We used R scripts from von Brömssen et al. (2021) to run general additive models (GAM) to detect overall increasing or decreasing trends as well as identifying periods with increasing or decreasing trends. A Regional Mann-Kendall (RMK; Helsel and Frans, 2006) test was performed using the R package rkt (Marchetto, 2021) to see if any cross-site trends could be detected.

RMK tests weighted by land cover were also performed. Trends in TP, SS, TN, NH_4–N and NO_3–N were weighted by amount of agricultural land. A similar analysis was performed where trends in TOC were weighted by share of forest cover and wetlands. Weights were assigned in the following manner. Individual Mann-Kendall tests for all stations were performed using the same R package, followed by weighting was done on the Kendall's score (S) statistics and the variance (σ) from the individual tests.

$$S_{WR} = \frac{1}{\overline{\overline{W}}} \sum_{L=1}^{m} w_L S_L \tag{1.}$$

$$\sigma_{WR} = \sqrt{\frac{1}{\overline{\overline{W}}} \sum_{L=1}^{m} w_L \sigma_L} \tag{2.}$$

Where S_{WR} is the weighted regional S, L is the site number, m the total number of sites, w is the fraction of land use, and $\overline{w}$ is the mean land use (either agricultural land or forest and wetland). A p-value <0.05 was determined as statistically significant for all statistical and trend analyses. All statistical analyses were performed using R 4.0.3 (R Core Team, 2020).

2.4. Summary of mitigation measures to reduce diffuse losses from agriculture

All available data on mitigation measures implemented to reduce agricultural diffuse nutrient losses in the Norrström basin were downloaded on June 22, 2022 from Water Information System Sweden (WISS, 2022). Only existing and fully implemented measures valid for the water cycle 2017–2021 were retained for further analysis. For constructed wetlands, the SMHI wetland database (SMHI, 2022b) was used as it is more complete than WISS for the study region. Each measure was assigned to a catchment based on reported geographic coordinates. Six main mitigation measures appeared in the search results: buffer zones, adjusted buffer zones, structural liming, spring tillage, catch crops and constructed wetlands. Measure validity was assessed based on information from WISS for starting implementation year and measure life span. Structural liming has a life span of 15 years, constructed wetlands 30 years and all other measures 5 years. Areas of all mitigation measures were summed for each tributary with consideration of existing upstream catchments. These areas were then related to total arable land area within the catchment and expressed as a percentage. Spatial analyses were performed using ArcGIS Desktop 10.8.

Both buffer zones and adjusted buffer zones have permanent vegetation, often grass, between the field and the stream that is meant to trap particles and nutrients (primarily P) in overland flow, and thereby decrease losses to receiving waters (Uusi-Kämppä et al., 2000). Adjusted buffer zones may also be placed within a field, e.g., around a surface runoff inlet well. Structural liming is used mainly on clay soils to stabilise soil structure and decrease erosion risk, thereby limiting particle and P loss (Ulén et al., 2012). Constructed wetlands can decrease losses of both N and P (Kynkäänniemi, 2014). Spring tillage primarily decreases N mineralisation and losses but also protects soil from erosion and thereby decreases particle and P losses (WISS, 2022). Catch crops are planted to avoid bare soils after harvesting, to reduce erosion and to take up remaining N, thereby reducing N losses (Aronsson et al., 2016).

Statistics on manure and mineral fertilizer use were obtained from Statistics Sweden (SCB, 2021; production area 4, "Svealands slättbygder"). Winter wheat (*Triticum aestivum*) yields for four counties covering the Norrström basin were downloaded from Sweden Statistics database (SCB, 2022).

3. Results & discussion

3.1. Water quality relations and catchment characteristics – transferability from headwaters to larger catchments

Mälaren tributary catchments cover a range of physico-chemical conditions, hydrological regimes, land use and soil types (Table S1). Concentrations for water quality parameters are highly variable (Table S2). Differences in nutrient levels between sites dominated by forest versus arable land were apparent, e.g., fw TP concentrations in forest dominated catchments (>65%) (Arbogaån, all Kolbäcksån stations, Kölstaån, Hedströmmen, Valstaån and Svartån) were low, while catchments with higher percentages of arable land (Enköpingsån, Hjälstaån and Sagån) had higher fw TP concentrations (Tables S1 and S2). Catchments with a larger percentage of water have lower nutrient and SS concentrations, which is consistent with other studies (Saunders and Kalff, 2001; Land et al., 2016; Alexander et al., 2008).

The PCA showed that percentage of water (mainly lakes and wider parts of streams) within the catchment and upstream catchment area were negatively correlated with both SS and P losses (Fig. S2). Similar to Sandström et al. (2020), there were linear relationships between both TP and SS and TP and soil clay content (Fig. S3).

Despite the varying land uses across tributary catchments, SS and soil clay content are still the best predictors of TP (eq. (3)) ($p < 0.001$, $R^2 = 0.80$, $N = 30$):

$$TP = -0.01 + 0.04 * SS + 0.0009 * MedianClayContent \quad (3.)$$

Catchment percentage water area is also a significant predictor of SS (eq. (4)) ($p < 0.001$, $R^2 = 0.76$, $N = 30$):

$$SS = 3.5 - 0.42 * Waterarea + 0.13 * MedianClayContent + 0.22 * Arableland \quad (4.)$$

Previously identified relations (Sandström et al., 2020) were also corroborated for these larger mixed land use catchments (Figs. S4 and S5). This demonstrates transferability of findings based on studies of smaller headwater catchments to larger, more complex ones, but also points out the importance of arable land and clay content for SS and P concentrations across spatial scales. The R^2-values are slightly lower than those reported by Sandström et al. (2020) (0.59 and 0.75 (Figs. S4 and S5) vs. 0.75 and 0.95 previously), but the equations are very similar. Tributary catchments are within the lower range of SS and P losses and are larger with a more mixed land use in comparison to those studied by Sandström et al. (2020). Thus it is reasonable that other factors also influence PP and SS concentrations, and regression models have a lower amount of explained variance.

The PCA indicated that larger upstream catchment areas were correlated with lower SS and PP fw concentrations (Fig. S2). This finding corroborates Milliman and Syvitski (1992) who studied 280 rivers discharging to the ocean across the world and highlighted the inverse relationship between drainage basin area and areal SS yield. Low relief landscapes in larger catchments tend to promote sediment deposition and storage, resulting in lower SS yields and concentrations (Woodward and Foster, 1997). Using 36 Swedish stations, Brandt (1990) concluded that concentrations of suspended material in small catchments were more than twice the mean value of those for nearby larger basins where suspended material could settle in lakes and reservoirs, on floodplains and in river channels. Catchments studied by Sandström et al. (2020) were all smaller, dominated by arable land and without lakes. Since lakes retain SS and slow down material transport, they should have influence on SS exports in rivers and streams. However, the influence of clay content and percent agricultural land seen in smaller headwater catchments is still apparent at the larger catchment scale. Previous studies have not been able to demonstrate this transferability across spatial scales. Bol et al. (2018) suggested that patterns visible at a larger catchment scale are not visible at a headwater scale. Dupas et al. (2023) suggest that this is due to larger variability between catchments. Haygarth et al. (2012) tested upscaling of P signals from plot scale to headwater scale. They found difficulties detecting the same signals at a headwater scale, that they observed on a plot scale. Taken together, these findings highlight the importance of (1) studying relationships between catchment properties and water quality across a range of spatial scales and (2) the importance of agricultural land, clay content and SS for P transport.

Some N species also correlate with P (Fig. S2). TOC is not related to other water quality variables, but is positively correlated to amount of forest and wetlands (Fig. 2). High TOC levels are common in more forested catchments. The site with the greatest forest (Hjälstaån) also had the highest total mean TOC concentration (Tables S1 and S2). However, all Kolbäcksån sites have a large proportion of forest cover, but some of the lowest mean fw TOC concentrations.

3.2. Water quality trends

Soil clay content and percentage of agricultural land are generally stable over time. Thus, mitigation measures placed on agricultural land are one way to focus efforts for decreasing nutrient losses to surface waters, and water quality trend analyses can be used to detect potential effects and changes. No significant trends in modelled water discharge over the studied period (2004–2020) were detected (data not shown). GAM modelling of raw concentrations (Fig. 2) identified a larger number of significant trends than for fw concentrations and loads (Fig. S6). TOC had the largest number of detected trends for both raw (17/30) and fw concentrations (7/30, Fig. S6) of all analysed parameters, with most trends occurring approximately simultaneously (Fig. 2). Individual MK trend tests also identified the largest amount of trends in TOC, but the same amount for NH_4–N (20/30 sites) (Table S3).

3.2.1. Phosphorus and suspended solids

There were six significant decreasing GAM trends for raw TP concentrations, with two lasting for the entire studied period (Kolbäcksån Västanfors and Tandlaån). MK trend tests identified 11 significant trends for raw TP concentrations, three increasing and eight decreasing (Table S3). For SS raw concentrations, three significant decreasing GAM trends that lasted the entire study period were found (Fyrisån Flottsund, Kolbäcksån Strömsholm and Oxundaån; Fig. 3 and Fig. S6). Valstaån showed an overall increasing raw SS concentration trend. Both GAM and MK trend tests identified significant trends at 12/30 sites for raw SS concentrations (Table S3, Fig. 2). Significant decreasing RMK trends for raw TP concentrations and raw SS concentrations were detected, both with the unweighted RMK and when the analysis was weighted for percentage agricultural land (Table S4).

The small number of significant decreasing TP trends could depend on several factors. Internal loading where P is released the streambed could sustain TP concentrations even when external inputs are reduced (Sonesten et al., 2013; Withers and Jarvie, 2008). Most decreasing trends in TP were found in forest dominated catchments (Kolbäcksån Strömsholm, Kolbäcksån Västanfors and Kolbäcksån Virsbo). Decreases were also seen at Sävjaån Kuggebro and Tandlaån; both these catchments have considerable amounts of arable land (>20%). Dupas et al. (2018) concluded that multidecadal P trends are influenced by decreasing point source emissions. In Mälaren, the largest point source reductions occurred three decades ago, before the start of the present study. Furthermore, the studied time period might be too short to detect changes using GAM analyses, since large amounts of data are needed to detect changes (Wellen et al., 2020). In the RMK analyses all data in the larger Norrström basin were used simultaneously, increasing the data amount compared to GAM analyses of individual sites. A 20% reduction of TP load and fw mean concentrations requires decades of data to be detected (Wellen et al., 2020). Wellen et al. (2020) use a “before and after” change approach, while in this study there is no clear time of implementation as measures are consistently being implemented,

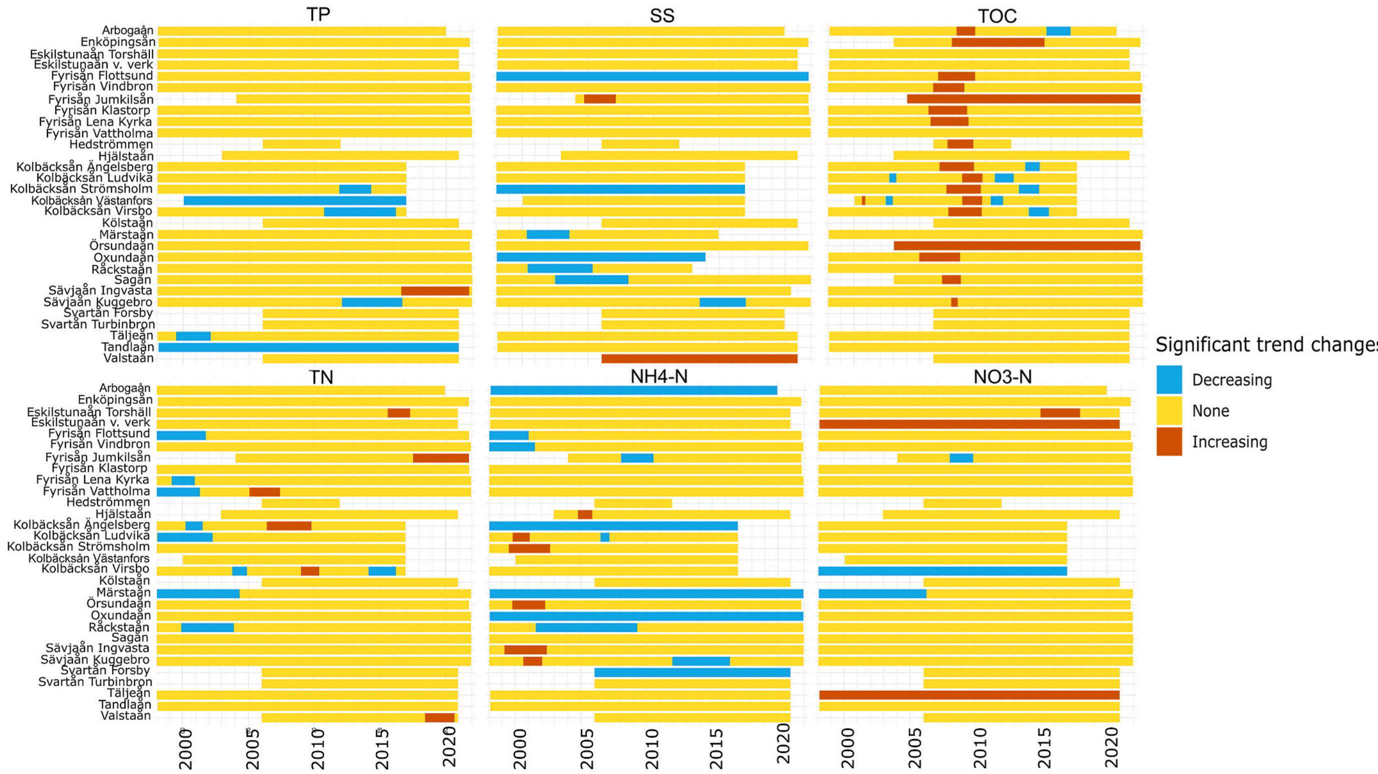

Fig. 2. Trends in raw water quality (from grab samples) for total phosphorus (TP), suspended solids (SS), total nitrogen (TN), ammonium-N (NH4–N), nitrate-N (NO3–N) and total organic carbon (TOC). Blue indicates a period with a significant decreasing trend, yellow no significant trend and red a significant increasing trend. (For interpretation of the references to colour in this figure legend, the reader is referred to the Web version of this article.)

resulting in a constant change. The difference in results between GAM and RMK highlights the need for large data sets and long term monitoring to actually detect trends. There are several additional possible explanations for the lack of strong significant downwards trends. In many cases there are simply not enough mitigation measures implemented or correctly placed to make an actual difference in nutrient loads (Djodjic et al., 2020). The decreasing trends in several tributaries recorded at the end of 1960s and early 1970s (Persson, 2001) were all due to improved wastewater treatment. For instance, TP loads from Eskilstunaån, Fyrisån, Märstaån and Oxundaån to Mälaren decreased by 44, 68, 94 and 76%, respectively before (1966–1970) and after (1981–1985) introduction of waste water treatment (Persson et al., 1990). Such tremendous reductions in P inputs within a short period of time and usually very close to the measurement station resulted in a rapid response of riverine concentrations with clear decreasing trends. Since then, most P inputs to these tributaries are likely from diffuse sources (Mälarens vattenvårdsförbund, 2021). There are a number of possible reasons why effects of measures on water quality were not detected. In many cases, there is a lack of accurate information on exact location of the implemented measures. Compared to point sources, reductions from diffuse sources are usually considerably lower, less certain, more variable and prolonged in time, and by definition occurring very often far away from the measurement point, i.e. before the natural retention in the system occurred (Alexander et al., 2000). Similar results have been seen on a larger European scale. For example, (Haase et al., 2023) studied biodiversity recovery trends in rivers, which should be strongly connected to water quality. Recovery of freshwater biodiversity came to a halt after 2010, suggesting that current mitigation measures are not resulting in the desired effect.

In general, lower nutrient concentrations were observed in the more forested dominated catchments (e.g. Kolbäcksån), especially for P (Table S1). One plausible explanation reported from several Swedish forested lakes with minimal anthropogenic impact is an ongoing oligotrophication (Huser et al., 2018; Camiolo, 2022), where P concentrations in already nutrient poor systems keep decreasing.

3.2.2. Nitrogen

Few GAM trends were detected for TN and NO_3–N. However, constant decreasing raw NH_4–N concentration trends were noted for five stations using GAM analyses (Arbogaån, Kolbäcksån Ängelsberg, Märstaån, Oxundaån and Svartån Forsby) and 18 stations using MK tests. There were also decreasing GAM trends for NH_4–N loads at five stations, and at seven stations for fw concentrations (Fig. S6). However, different tributaries had significant trends for raw concentrations, fw concentrations and loads. Fyrisån Flottsund, Hjälstaån, Örsundaån and Sagån showed constant decreasing trends for fw concentrations and loads but not for raw NH_4–N concentrations, despite no detected trends for water flow. Both N fertilizer use and yields have increased during recent years (Fig. S7) and spring tillage (Fig. 3) is used in several catchments to decrease N losses. These counter-acting factors in combination with increasing yields in recent years may balance the increased fertilization, resulting in no or decreasing trends. RMK tests for raw TN, NH_4–N and NO_3–N all resulted in significant decreasing trends whether or not trends were weighted by proportion of agricultural land (Table S4).

When fw concentrations and loads were used, fewer GAM trends were detected (Fig. S6), perhaps due to the shorter time period used for analysis (2004–2020) (Wellen et al., 2020). NH_4–N showed many decreasing trends for raw concentrations, fw concentrations and loads. Tributaries with significant GAM trends for several parameters included Fyrisån Jumkilsån, Kolbäcksån Strömsholm and Kolbäcksån Virsbo. At Eskilstuna vattenverk only one trend (raw NO_3–N concentrations) was detected during the studied time.

3.2.3. Total organic carbon (TOC)

Increasing TOC GAM raw concentrations occurred at several stations

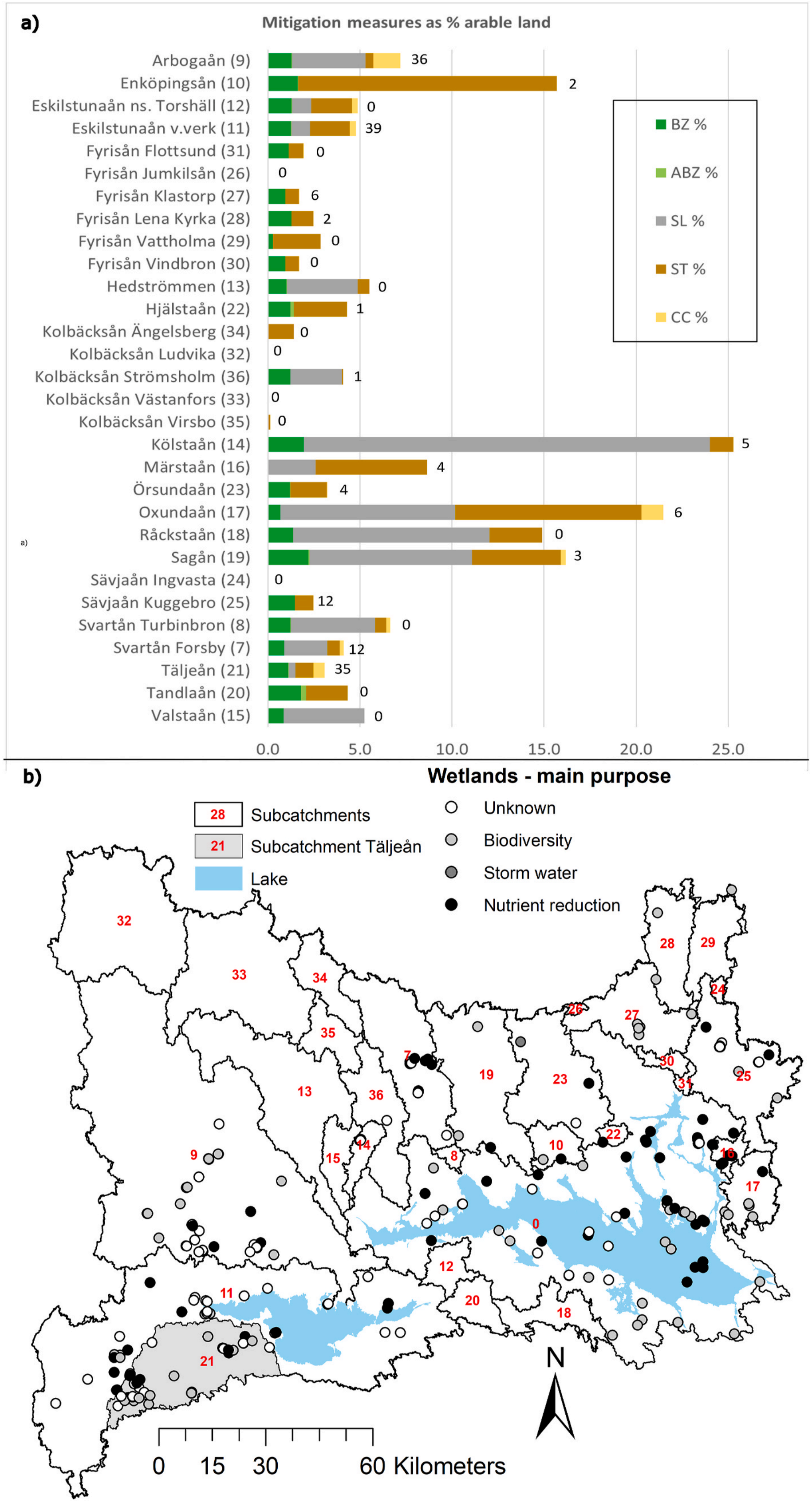

Fig. 3. a) Implemented mitigation measures for nutrient losses in all Mälaren catchments expressed as a percentage of arable land. BZ = buffer zones, ABZ = adjusted buffer zones, SL = structure liming, ST = spring tillage, CC = catch crops. The number after the bar represents the number of constructed wetlands within the catchment. The number in brackets after the name corresponds to the catchment number in the map in b). b) Map over implemented wetlands in the different

catchments and their main purpose. The red numbers correspond to catchment number in a). (For interpretation of the references to colour in this figure legend, the reader is referred to the Web version of this article.)

(Fig. 2). In all Fyrisån tributaries except Vattholma, all Kolbäcksån tributaries and five more locations (Arbogaån, Enköpingsån, Hedströmmen, Oxundaån, Sävjaån Kuggebro) temporarily increasing trends for raw TOC concentrations occurred around 2006–2009 (Fig. 2). Overall increasing trends occurred at Fyrisån Jumkilsån and Örsundaån. A similar pattern in TOC trends was seen by Eklöf et al. (2021) who studied temporal changes of TOC and coloured dissolved organic matter (CDOM) in 164 watercourses across Sweden, where a similar partial increase around 2007 was seen in several stations. They concluded that there has been a brownification with increasing TOC, which seem to have ceased. This could be what we are detecting here as well as their findings correspond with observations from individual stations (Fig. S8).

For all Kolbäcksån sites, the increasing TOC raw concentration trend was followed by a decreasing trend (Fig. 2), with a peak around 2010 followed by a decrease, that seems to be continuing (e.g. Fig. S8a). A spatial pattern in TOC trends (Figs. 1 and 2) is visible, with more trends in the western part of the Mälaren basin. The RMK tests indicated significant increasing trends for both the unweighted and areal weighted analyses (Table S4).

3.3. Mitigation measures

The lack of strong decreasing trends in nutrient concentrations is most likely due to several factors including low sampling frequency, limitation of the statistical tools, a need for larger data sets, limited implementation of measures and poor targeting of existing measures.

Mitigation measures were implemented in almost all study catchments (Fig. 3). Structural liming and spring tillage are the most common measures followed by buffer zones (Fig. 3). However, the percentage of arable land subject to mitigation measures is generally low in all catchments, with the highest amount in Kölstaån (25% of arable land) and no mitigation measures implemented in the forest dominated Fyrisån Jumkilsån, Kolbäcksån Ludvika, Kolbäcksån Västanfors and Sävjaån Ingvasta catchments (Fig. 3). This is not surprising due to the low amount of arable land in these catchments. Some mitigation measures (e.g. structural liming and spring tillage) can overlap and cover the same area. In total, 239 wetlands were found in Norrström. However, 70 of these were situated in the vicinity of Mälaren itself and therefore not assigned to any tributary. Eskilstunaån has the highest number of implemented constructed wetlands, while Täljeån has the highest wetland density (Fig. 3).

The Täljeån tributary was therefore studied more in detail to estimate potential nutrient reductions associated with implemented measures. Following Djodjic et al. (2020), we calculated potential nutrient reductions associated with all 36 constructed wetlands in the Täljeån tributary catchment. Long-term annual average nutrient loads transported by Täljeån are 748 t N and 14 t P. The total modelled N and P reduction for all wetlands in this catchment amounted to 3.8 t N and 0.7 t P, accounting only for 0.5 and 5 % of total annual load for N and P, respectively (Table S5). The low effect was mainly due to the inappropriate wetland placement. Wetlands were placed in areas with low shares of arable land, thus nutrient loads and retention were both low (Djodjic et al., 2022). Such low reductions might be difficult to identify by trend analysis (Table S5). Optimizing placement within the catchment for most efficient removal of nutrients would decrease the needed area of the constructed wetland and increase P retention (Djodjic et al., 2020). Similarly placing buffer strips around erosion prone areas would increase their efficiency (Djodjic and Villa, 2015; Djodjic et al., 2018; Piniewski et al., 2021). Furthermore nutrient removal efficiency of wetlands and some other measures is highly dependent on loading rate (Land et al., 2016), meaning that the effect of implemented measures at the river outlet might be masked by natural variations in flow and nutrient concentrations. In other words, the high removal efficiency in wetlands during years with high nutrient loadings might not be detected as a significant change in river concentrations due to a higher loading from all other sources. This highlights the importance of continuous high frequency monitoring, to see if implemented mitigation measures will have an effect later on.

Despite national and international commitments to reduce eutrophication and funding programs for mitigation measures, there are few detectable decreasing nutrient trends in the Mälaren tributaries. This is either due to low implementation levels or the limitations of trend analyses. A lack of coordinated reporting of mitigation measure implementation makes it even more difficult to evaluate effectiveness (Swedish Institute for Marine Environment, 2022). Even catchments with high reported intensity of measure implementation, e.g., Kölstaån, Sagån, Enköpingsån and Oxundaån (>15%) had no detectable nutrient trends using GAM analyses (Fig. 2 and S5). Some mitigation measures, e.g., buffer zones (Hoffmann et al., 2009) and structural liming (Blomquist, 2021) can reduce erosion and surface runoff, and constructed wetlands slow water flows allowing SS and PP to settle. Since a large proportion of P loads in these catchments are from agriculture (Mälarens vattenvårdsförbund, 2021), mitigation measures implemented on arable land should have the most detectable effects. In addition, the strong signal of amount agricultural land affecting SS and P concentrations in these streams and rivers suggest that this is where we need to focus management efforts to decrease loads, since reducing the amount of agricultural land in most cases is not a viable solution. Hence, significantly decreasing SS and TP trends were expected in catchments with a large number of implemented measures. On the other hand, the spatial extent of implemented mitigation measures is generally low (<5% of arable land) (Fig. 3).

Legacy stores of e.g. P in lakes and streams (e.g. Lannergård et al., 2020, (Sandström et al., 2021)) could also affect the situation where a reduced external input will not result in immediate decreasing concentrations in the stream with considerable time lags before the effects can be detected (Meals et al., 2010; Sharpley et al., 2013), the same is true for recently implemented measures (Van Meter and Basu, 2017). Van Meter et al. (2018) concluded that even if agricultural N use became 100% efficient, it would take decades to meet target N loads due to legacy N within the Mississippi River basin. Fertilizer application strategies should consider the amount of nutrients accumulated in the soil. Goyette et al. (2018) calculated historical P accumulation in 23 Canadian watersheds, and could define a threshold, where after a certain mass of P (2.1 t P km^{-2}) accumulated in the soil, losses to nearby watercourses increased linearly. Current P accumulation in Swedish agricultural soils is an average of 65 t km^{-2} (Andersson et al., 1998). Applying this value to the proportion of arable land in the Mälaren tributaries results in 25/30 catchments exceeding the critical threshold (Table S6). The forested Kolbäcksån catchments do not exceed this threshold or are just on the limit (2.1 t km^{-2} for Kolbäcksån Strömsholm). Enköpingsån and Täljeån have the largest amounts of accumulated P (27.1 t km^{-2} and 29.3 t km^{-2} respectively). In these sites barely any P trends were detected except for a partial decreasing trend in TP raw concentrations at the start of the time period in Täljeån. Recently, the Swedish government identified 20 pilot study catchments across the country Catchment managers were appointed in each catchment with a mandate to promote and facilitate the implementation of cost-efficient mitigation measures to reduce eutrophication (Swedish Agency for Marine and Water Management, 2021). This could be a way forward to increase implementation of mitigation measures.

Increasing variability in rain intensity and more freeze/thaw cycles in a changing climate might lead to an increased variability in amount and timing in nutrient losses. Increases in precipitation and higher winter temperatures have already been noted in the studied area (Sonesten et al., 2013). On the one hand, effectiveness of currently implemented mitigation measures could be reduced by increases in precipitation and runoff. On the other hand, increased precipitation and runoff could instead prevent any increase in concentrations (Crossman et al., 2013; Räike et al., 2020). Catchments with higher percentages of recently implemented measures (Kölstaån, Oxundaån, Råckstaån, Sagån) should be carefully monitored in coming years to follow up on measure effectiveness. Given the variability in climate and including the uncertainty in sampling methodology and lab analyses, there is a need for better evaluation of measure effectiveness and trend detection.

There is an ongoing transgression of the planetary boundaries, including biogeochemical flows of P and N (Steffen et al., 2015), and the lack of trends seen in these parameters in the tributaries to Mälaren does not suggest a step in the right direction. To decrease these flows and improve the status of Mälaren tributaries, Lake Mälaren and ultimately the Baltic Sea, the final recipient, there is a need to increase the efforts for decreased P and N losses. Using the findings from Mälaren, a great lake, implying that higher implementation rate of mitigation measures is possible and necessary, indicates that similar conditions might be valid for the Baltic Sea.

4. Conclusions

Water quality in agricultural catchments and elsewhere will be under increasing pressure in the near future, due to climate change, land use changes and the limited implementation of mitigation measures.

Our specific findings are as follows:

- The relationship between share of arable land, soil clay content and SS and P concentrations observed at the headwater scale is applicable in larger, mixed land use catchments. This relationship should be recognised when targeting mitigation measures, to achieve higher cost-efficiency.
- Few significant water chemistry trends were identified using GAM. However, RMK tests identified significant regional trends of all parameters. This emphasizes the importance of long-term, regional monitoring programs to document trends in water quality under a changing climate.
- No connection between implemented mitigation measures on arable land and water quality trends could be seen, even though arable land is the main source of nutrients in the majority of the catchments. This may be due to insufficient implementation of mitigation measures or intrinsic limitations in the monitoring programme, emphasizing the need for further research to address effects of mitigation measures in different scales, ranging from fields and headwaters to river basins.
- Careful evaluation and communication of results is imperative as the lack of significant trends might discourage from further mitigation. Absence of evidence is not evidence of absence.

This study shows that the work done so far is not enough to improve the water quality in the studied catchments. More work needs to be done to explore the reasons behind this, e.g. follow up on implemented mitigation measures to determine if a change could be expected. As proven in this study, the effect of arable land and clay content on SS in small streams is strong enough to also show in larger rivers, indicating a possibility for further up-scaling, to use findings from great lakes on enclosed seas, e.g. the Baltic Sea. Future studies should explore this possibility further to develop policy actions to avoid crossing planetary boundaries.

CRediT author statement

Sara Sandström: Conceptualization, Formal analysis, Investigation, Methodology, Software, Validation, Visualizations, Writing: original draft and review & editing. **Emma E. Lannergård:** Conceptualization, Formal analysis, Data curation, Methodology, Visualizations, Writing: review & editing. **Martyn N. Futter:** Conceptualization, Methodology, Formal analysis, Writing: review & editing. **Faruk Djodjic:** Conceptualization, Methodology, Data curation, Visualizations, Formal analysis, Writing: review & editing.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Data availability

The data is open environmental monitoring data, and has been referenced in the methods section with a link in the reference list.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.jenvman.2023.119500.

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