Temporal Trends and Drivers of Mercury Pollution in a Boreal Hydropower Reservoir

Three decades of Mercury in Water and Fish

Karin Halldin

Temporal Trends and Potential Drivers of Mercury Pollution in a Boreal Hydropower Reservoir: Three Decades of Mercury in Water and Fish

Temporala trender och potentiella drivare av kvicksilverföroreningar i en boreal vattenkraftsdam: Tre årtionden av kvicksilver i vatten och fisk.

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Abstract

Methylmercury (MeHg) is a widespread toxic pollutant of major concern in boreal aquatic ecosystems. The inundation caused by hydropower impoundments is well-known to elevate the concentrations of MeHg in water and biota. This study will investigate how a specific hydropower impoundment in northern Sweden has affected mercury (Hg) levels in surface waters and perch during a period of 30 to 32 years. The study focuses on the evolution of Hg following the initial, well-known elevation of MeHg. Temporal and spatial analyses are used to specify how the reservoir is affecting Hg levels through time, for levels both inside the reservoir as well as upstream and downstream. An analysis of potential drivers of the Hg concentrations was used to relate the concentration trends to environmental processes. From the results, the main potential driver for all three Hg variables in the reservoir is water colour, however, several potential drivers, such as oxygen content, have not been tested. The results from temporal and spatial analyses suggest that the elevated MeHg concentrations caused by inundation subsequently decreased for about two decades before the concentrations stabilised at a concentration two times higher than those in an adjacent reference site. Total Hg (THg) concentrations in surface water and THg concentrations in perch are also stabilised at a concentration two times higher than the reference lake three decades after inundation. These continuously elevated levels are suggested to be caused by either the hydropower management, the impact the hydropower impoundment has on the water regime, or potential variations in catchment Hg and organic matter discharge between the hydropower sites and the reference site.

Keywords: Mercury, methylmercury, boreal landscape, hydropower reservoir, inundation, impoundment, fish, time series, atmospheric deposition

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Abbreviations

SLU	Swedish University of Agricultural Sciences
IVL	IVL Swedish Environmental Research Institute
OM	Organic Matter
DOC	Dissolved Organic Carbon
DOM	Dissolved Organic Matter
Hg	Mercury
THg	Total Mercury
MeHg	Methylmercury
Environmental monitoring variables	pH, Colour, Alkalinity, Conductivity, Total Phosphorus
Abs_{420}	Absorbance at 420 nm
THg_{aq}	Total Mercury in surface water samples
$MeHg_{aq}$	Methylmercury in surface water samples
THg_{fish}	Total Mercury in fish muscle samples (of young perch)
CP analysis	Change Point Analysis
LOQ	Limit of Quantification
ANOVA	Analysis of Variance
PCA	Principal Component Analysis
SKM (the reservoir)	Skinnmuddselet
OVR (upstream 1)	Övre Selet
LAN (upstream 2)	Lanaträsk
LAV (lake upstream)	Lavsjön
STT (lake downstream)	St. Tällvattnet
VIS (the reference lake)	Viskasjön

1. Introduction

Hydropower is an important energy source with both positive and negative impacts. It accounts for about 30% of the European Union's (EU's) renewable electricity consumption and about 45% of Sweden's electricity production (Swedish Energy Agency 2022; Eurostat 2024). Man-made reservoirs are created for several purposes, one of which is to generate hydropower through water regulation. Over 30 000 reservoirs have been constructed during the last half-century, altering roughly 20% of global annual river runoff (Zhou et al. 2016). Inundation following dam construction has several negative impacts, one of these is a significant increase of mono-methylmercury (CH_3Hg, known as MeHg) in water and aquatic biota (De Bonville et al. 2020). MeHg, an organic species of Hg, is a neurotoxin that can cause damage to humans' central nervous system. Fish consumption is the primary pathway for human exposure to MeHg, particularly through fish at high trophic levels (National Research Council [US] Committee on the Toxicological Effects of Methylmercury (2000). High MeHg concentrations in fish are hence a great concern in most of the northern hemisphere countries (Eklöf et al. 2012).

Inundation releases inorganic Hg and organic matter (OM) into the water. This is particularly concerning in organic-rich soils, such as those in forest and peat landscapes, as OM and nutrients stimulate the transformation of inorganic mercury into MeHg (Ullrich et al. 2001). The concentrations and composition of Hg and OM are not only dependent on the environmental conditions in the immediate surroundings but also on processes occurring upstream (De Bonville et al. 2020). Hydrology influences the Hg and OM dynamics and concentrations as well as the existing redox conditions (Ullrich et al. 2001), such as retention times (De Bonville et al. 2020) and run-off from the catchment area (Eklöf et al. 2012). Activities in the catchment area, such as forest harvest, do increase the MeHg levels (Kronberg et al. 2016). An impoundment does affect the MeHg levels in other parts of the stream as well (Levanoni et al. 2015).

This master's thesis will assess the long-term impact of hydropower reservoirs in boreal landscapes on MeHg concentrations in the waterway and its accumulation in fish. This will be addressed by disentangling the dynamics of Hg pollution in water and fish samples following the establishment of a large hydropower reservoir (Skinnmuddselet). Hg pollution in the reservoir was assessed by monitoring water samples of MeHg and THg (total Hg) as well as THg in fish (European perch, *Perca*

fluviatilis L.). The reservoir is located in northern Sweden, representing a typical hydropower reservoir in a boreal landscape characterised by abundant forests and wetlands in the catchment area. A 30-year-long monitoring dataset enables a unique analysis of long-term temporal trends and changes in Hg levels. To this writer's knowledge, this is one of the first studies of such a long-term dataset of a hydropower regulated water system. The dataset covers the reservoir, a reference lake as well as lakes upstream and downstream of the reservoir, which enables the study to investigate how the impoundment affects the Hg dynamics upstream and downstream as well. An analysis of environmental variables within the catchment area, and in the aquatic environment will also be implemented, to enable an understanding of underlying drivers that control the dynamics of Hg pollution in the artificial water system. This study is valuable since few long-term studies of surface water Hg concentrations exist (Eklöf et al. 2012), especially long-term studies of Hg levels in reservoirs (Eckley et al. 2015; Zhou et al. 2016).

1.1 Objectives

This study aims to enhance the understanding of how the establishment of a boreal hydropower reservoir influences Hg concentrations in water and fish, both downstream and upstream of the reservoir. Additionally, it seeks to identify potential drivers that control the long-term evolution of Hg pollution in water and fish over 30 years following the formation of the reservoir Skinnmuddselet.

This study aims to answer the following questions:
  (1) How has environmental Hg pollution evolved temporally and spatially in the Skinnmuddselet hydropower reservoir system?
  (2) What drives Hg concentration evolution in the hydropower-impacted aquatic system?

These objectives will be pursued using 32 years of data obtained from the Skinnmuddselet monitoring programme, conducted by the hydropower company Statkraft, as well as data such as temperature collected from open online sources, for instance, the Swedish Meteorological and Hydrological Institute (SMHI).

1.2 Background

Insights into the involved processes are essential to be able to reach the study's objectives, which this section will cover.

1.2.1 Long-Term Changes in Swedish Fish and Water

Mercury

Hg is a naturally occurring element whose origin in ecosystems is derived from both natural and anthropogenic activities. Forest fires and volcanic eruptions are among the natural processes which release Hg (Driscoll et al. 2013). The presence of Hg in terrestrial and aquatic ecosystems has increased greatly due to anthropogenic activities, such as mining, coal combustion and its use in commercial products (Munthe et al. 2001; Driscoll et al. 2013; Zhang et al. 2016). Hg levels in fish have also increased during the last century (Johansson et al. 2001). Most of the Hg in fish is in the form of neurotoxic MeHg (Bloom 1992).

The risk posed by MeHg to human health has led to several criteria regarding allowed concentrations in edible fish and drinking water to be set by various regulatory bodies (The Swedish Food Agency 2015; EU 2023). This risk has additionally caused the creation of several treaties, directives, and regulations aimed at reducing emissions and release of Hg. One such treaty is the Minamata Convention on Mercury (Minamata Convention on Mercury 2021). Another measure to keep track and increase comprehension of Hg is monitoring programmes in water systems.

There has been a substantial decrease in atmospheric Hg concentrations in Europe since the 1990s. This is derived from reduced Hg emissions within the EU (Zhang et al. 2016). This declining trend can be elucidated by the gradual removal of Hg from commercial products (caused by the implementation of the treaties, directives and regulations aimed at reducing emissions) and by changes in emissions from coal-fired utilities following the introduction of emission control measures on SO_2 and NO_x (Zhang et al. 2016).

Despite the notable decline in atmospheric Hg, levels of Hg in sediments, water, ground, and biota remain high. This persistence is due to that Hg^{II} form strong ionic bonds to OM. These strong Hg-OM complexes result in substantial amounts of legacy Hg being stored in organic-rich soils, such as those in boreal forests and peatlands. This legacy Hg continues to cycle through the environment and is continuously discharged to downstream waters via runoff (Johansson et al. 2001; Hintelmann et al. 2002; Ravichandran 2004; Driscoll et al. 2013; Bishop et al. 2020).

Browning of Surface Waters and Recovery of Acidification

In recent decades there has been a significant increase of OM in waters in Europe and North America. There exist several potential explanations for this increase such as higher runoff due to climate change, temperature, and recovery from acidification (Eklöf et al. 2012). That the browning trend is a result of water systems' recovery from acidification is probable for some areas. There has been a substantial reduction in atmospheric sulphur deposition in recent decades for some areas which probably increased the mobility of dissolved organic carbons (DOC) since the solubility of DOC is affected by the water acidity (Evans et al. 2006). It seems that THg does not follow the trend of DOC increase, even though it's strongly correlated to OM (Eklöf et al. 2012). An increasing trend of Hg in fish in recent years has however been found which possibly could be explained by the increased solubility of DOC (Rask et al. 2024).

1.2.2 Mercury in Boreal Freshwater Ecosystems

The Mercury Biogeochemical Cycle

Hg exists naturally in three forms: elemental (Hg^0), inorganic salts (Hg^I, Hg^{II}) and organic (e.g., MeHg) form (Rask et al. 2024). The major inorganic species in water are Hg^0 and Hg^{II} since the redox transition state of Hg^I is unstable in natural water (Ullrich et al. 2001). Anthropogenic Hg emission to the atmosphere consists of elemental Hg^0 and Hg^{II}, while dominated by Hg^0 (Médieu et al. 2022). The atmospheric residence time of Hg^0 ranges from several months to a year, which enables it to disperse over long distances across the globe (Driscoll et al. 2013). This means that Hg can end up far away from its original emission source. Atmospheric Hg deposition consists of dry (vegetation uptake of atmospheric Hg^0) and wet (rainfall deposition of atmospheric Hg^{II}) deposition (Bishop et al. 2020). Once deposited, Hg^{II} can be reduced back to Hg^0, some of which evades into the atmosphere (Ullrich et al. 2001; Paranjape & Hall 2017). MeHg is formed through methylation of inorganic Hg^{II}, primarily in anoxic and sub-oxic aquatic environments (Zhu et al. 2018). MeHg can be formed in boreal landscapes, e.g., peatlands and forest soils, and be transported to aquatic environments via stream runoff (Bishop et al. 2020).

Hg and OM (e.g., dissolved organic carbon, total soil organic carbon) do often exhibit a positive correlation (Eklöf et al. 2012; Lavoie et al. 2019), due to the strong binding between Hg^{II} and reduced sulphur functional groups in OM (Ravichandran 2004). OM is hence a main part of the transportation of Hg. When Hg^{II} is complexed with OM it is less bioavailable for methylation (Bishop et al. 2020). OM also affect the Hg cycle by involving processes related to microbial methylation, photodemethylation and bioaccumulation (Braaten et al. 2019).

Methylmercury

Hg methylation is considered to be primarily a microbially mediated process. Microbial methylation is where CH_3^- is transferred onto Hg^{II+} mainly by sulfate-reducing bacteria and to some extent iron-reducing bacteria in anoxic and sub-oxic conditions (Ullrich et al. 2001). The methylation rate is controlled by microbial productivity and availability of inorganic Hg (Paranjape & Hall 2017). These factors are influenced by temperature, pH, redox potential, available electron donors (e.g., SO_4^{2-}), suitable electron acceptors (often organic carbon), the presence of complexing agents, Hg species, and DOC. MeHg concentrations are affected by the dynamic processes of Hg^{II} methylation and MeHg demethylation. Demethylation occurs through two pathways: microbial metabolic oxidation and abiotic photodegradation. Photodemethylation is considered the main sink of MeHg in lakes (Ullrich et al. 2001; Levanoni et al. 2015; Paranjape & Hall 2017). Measured MeHg concentrations hence reflect the *net* methylation and demethylation.

Environmental factors affect MeHg concentrations in various ways:
- o Temperature increases microbial activity, increasing the rate microbes perform methylation (Ullrich et al. 2001).
- o pH influences which complexes exist. In a low pH, fewer MeHg bindings to humic and fulvic acids occur, meaning that MeHg is released from sediments and can be accumulated in biota to a higher extent than in a neutral environment (ibid.).
- o Which redox conditions exist determine which chemical reactions (reduction, oxidation) can occur (ibid.)
- o DOC and OM have a strong correlation to MeHg. OM-Hg complexation determines the solubility and mobility of Hg in water. Methylation and demethylation are affected since OM provides the microbes with nutrients. (ibid.)

Mercury in Fish

It's presumed that about 95% of the Hg present in the muscle tissue of adult predatory fish is MeHg (Bloom 1992). Recent studies, however, have found variability in the %MeHg of THg in fish (Moslemi-Aqdam et al. 2023). The levels of THg in fish are hence strongly influenced by the level of bioavailable MeHg in the aquatic environment. THg water concentrations on the other hand do not have the same influence on fish THg concentrations (Moslemi-Aqdam et al. 2023). Hg is bioaccumulated in biota. Fish at high tropic levels are known to have accumulated high levels of Hg due to biomagnification in food webs. The northern pike *Esox lucius* L. is a top predatory fish that preys upon other fishes such as European perch *Perca fluviatilis* L. (Rask et al. 2024). In general, pike has higher Hg concentrations than perch. Perch has higher concentrations than lower predatory fish and non-

predatory fish (Åkerblom et al. 2014). Hg concentrations are strongly linked to the length and growth of the fish, whereas larger fish generally have higher Hg concentrations (e.g., Moslemi-Aqdam et al. 2023; Rask et al. 2024).

Hg concentrations in fish are affected by changes in the ecosystem that increase the bioavailable MeHg in the water system. Such as forest harvest, lake browning, inundation, and surface runoff, which all increase the Hg and DOC load to the water system (Munthe et al. 2007; Rask et al. 2024).

Among Lake Variability in Mercury

The factors mentioned above have major impacts on where and at what level Hg and MeHg exist. Other natural factors which affect the levels of Hg and MeHg in water are surface runoff, which can be a major source of both THg and MeHg and provide an increase in OM which increases methylation. What the runoff contains is dependent on catchment hydrology such as if the flow path goes through organic and Hg-rich soils and the flow rate (Eklöf et al. 2012). The variability among lakes is influenced by catchment characteristics, such as land cover composition and surface area (Munthe et al. 2007; Eklöf et al. 2012; Moslemi-Aqdam et al. 2023).

1.2.3 Hydropower Reservoirs

When a hydropower reservoir is constructed a large area of terrestrial land is inundated by an impoundment. This impoundment alters the water regime of the area (Levanoni et al. 2015). It's well established that this inundation results in a drastic increase in MeHg concentrations in water and biota (Hall et al. 2005; Roy et al. 2009; De Bonville et al. 2020) These increased concentrations come from increased methylation rates, which are caused by newly formed anoxic environments and stimulation by an increase of fresh humic substances released from the inundated soils (Hall et al. 2005; Roy et al. 2009; Kasper et al. 2014; Herrero Ortega et al. 2018). The methylation decreased after a couple of years, probably due to less readily decomposed OM (Hall et al. 2005; Roy et al. 2009). At least one study has found that with time the Hg concentrations in predatory fish can return to background levels (Bodaly et al. 2007).

Hydropower management can affect the MeHg levels in the reservoir. In a North American study, a 75-year-old reservoir which is inundated 8 months per year shows that the sediment and porewater MeHg concentrations are 3 times higher in areas with water level fluctuations compared to permanently inundated sediments (Eckley et al. 2015, 2017). Fluctuating water levels have been shown to impact MeHg production (Singer et al. 2016).

1.3 Area Description

1.3.1 Management of Skinnmuddselet, 1986 - 2024

The artificial hydropower reservoir Skinnmuddselet is located in boreal northern Sweden at coordinates N 7106783, E 662812 in SWEREF 99 TM. In 1989, river Gideälven was impounded, creating the hydropower reservoir used by the Stennäs power station (Fig. 1).

To reduce the negative impacts of inundation on water quality preparatory measures were implemented before impoundment (Fig. 2). First, the area was limed by helicopter to increase pH. Secondly, between 1986-1989, all forest in the area was removed to reduce carbon levels. This deforested area was 55% of the area flooded by the dam. Branches thicker than 1 cm were incinerated on-site (Åberg et al. 2004; Jutterström et al. 2023). The rest of the flooded land was primarily peatland, agricultural land and Gideälven.
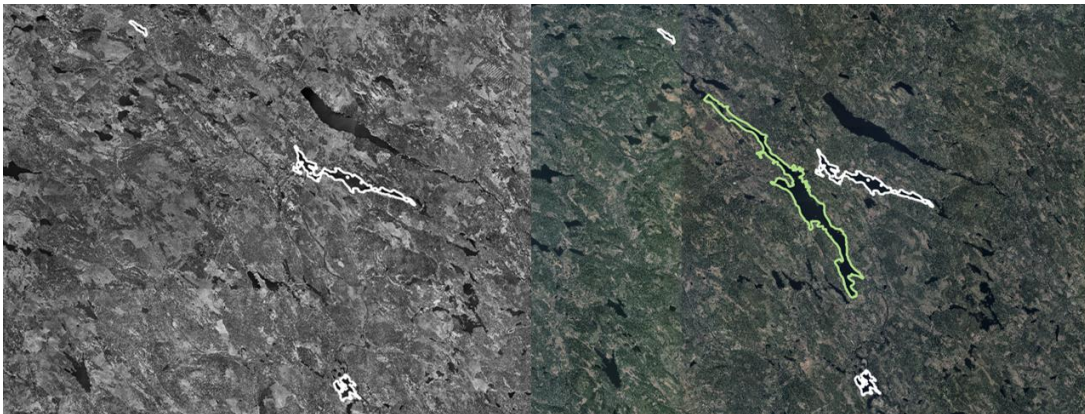
Figure 1. The area before (left) and after (right) the creation of the reservoir Skinnmuddselet. Where the white areas represent lakes investigated in this study that existed before the impoundment, and the green represents those that were created. Modified from Historiska ortofoton 1960 © Lantmäteriet 2024 (left), Ortofoto RGB present day (right) © Lantmäteriet 2024.
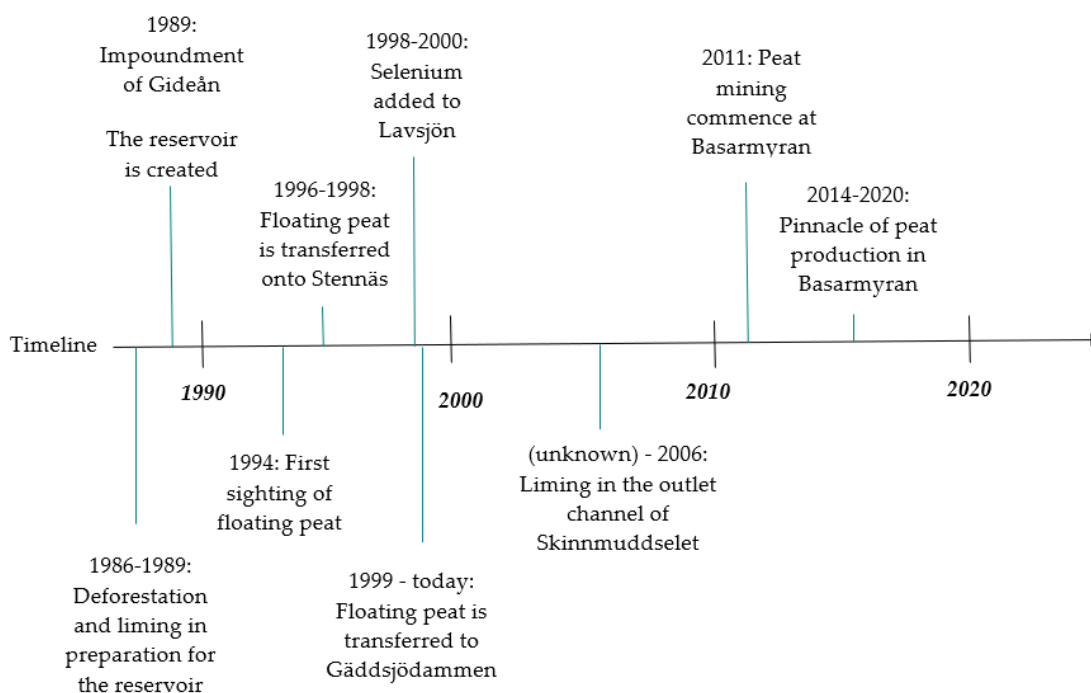
Figure 2. Timeline of recorded events in and adjacent to the reservoir during 1986 to 2024.

During the monitoring period, which ranged from 1984 to today, several events have occurred in and around the monitored waterbodies (Fig. 2).

- o Peat mining, i.e., peat extraction, began in Basarmyran in 2011 to extract peat for energy purposes. The mining had a pinnacle in extraction from 2014 until 2020 (Umeå Energi 2023) (Fig. 2). This mining site is located close to Skinnmuddselet with river outlets upstream of the reservoir, between the two sampling sites Lanaträsk (LAN) and Övre Selet (OVR) (Fig. 4).
- o Statkraft installed a liming dispenser in the outlet channel of the reservoir as a preventative measure to reduce the impact of acidification on Hg downstream by increasing pH. This liming was discontinued in 2006[1] (Fig. 2), since it was assessed to have no benefit, as the pH was already relatively high.

In 1994 floating peat was sighted in the reservoir (Fig. 3; Fig. 2), this has since then been a consistent element in the reservoir. This peat is believed to have come loose from adjacent peatlands. The presence of floating peat has increased with the years, possibly due to climate change. The first floating peat appeared after a warm and dry period hence it's assumed that high temperature or low water levels are the cause for it becoming loose. A bridge and the hydropower sluice gates are examples of areas which have on several occasions become blocked by floating peat (Fig. 3, middle). Measures were implemented to deal with the floating peat in 1996, where

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[1] Statkraft, the hydropower plant operators. Personal communication. 2024-04-22.

it was moved onto land at Stennäs. This was observed to elevate Hg levels, which caused the measures to change. From 1999 and onwards problematic floating peat is instead transferred into Gäddsjödammen (Fig. 2), where the peat has less effect on the Hg levels in the reservoir (Jutterström et al. 2023). To be able to transport the peat it's sawed into smaller segments (Fig. 3, right) which releases Hg and carbon into the water.

- Between 1998 and 2000 a low dose of selenium was added to the lake upstream of Skinnmuddselet (Fig. 2) as a measure to decrease the Hg levels in fish in the water system. The result of the measure was a reduction of MeHg in fish, attributed to decreased bioaccumulation of MeHg in the food web. This was due to the diet of zooplankton, exhibiting lower MeHg concentrations following the addition of selenium (Hultberg 2002).

A change in land use is assumed during the monitoring period, for example, wind power parks, but since most of the area is forest and peat, this is deemed minuscule considering, for example, the amount of forest harvest. Other land management might have occurred within the catchment sites without this study's knowledge.

Figure 3. Floating peat in one of the sample lakes (left). A small floating peat in front of the hydropower station's sluice gate (middle). Management of floating peat by sawing it into fragments (right) (Persson n.d a; Persson n.d b; Persson n.d c)

1.3.2 Sample Sites

The hydropower station is operated by Statkraft Sverige AB, formerly known as Graninge kraft AB. They are the financer of Skinnmuddselet Hg monitoring programme, from which this study's main dataset originates. The monitoring programme aims to oversee the progression of Skinnmuddselet hydropower dam's effect on Hg pollution in the artificial reservoir system.

This study will investigate data collected at six of the monitoring programmes sampling sites, which are (Fig. 4):

- A reference lake (Viskasjön [VIS])
- The reservoir (Skinnmuddselet [SKM])
- The upper part of the reservoir (Övre Selet [OVR])
- Upstream the reservoir in a swamp/riverlike area (Lanaträsk [LAN])

       o   The first lake upstream of the reservoir (Lavsjön [LAV])

       o   The first lake downstream of the reservoir (St. Tällvattnet [STT])

Sampling has occurred within the indicated areas in Fig. 4. These specific sites have been monitored to enlighten how the reservoir impacts the Hg levels. VIS was selected to be a reference site since it represents the same area as the other sites yet is not affected by the hydropower inundation. SKM was selected to represent the conditions within the reservoir. STT was selected to represent the conditions downstream of the reservoir, to be able to investigate how the reservoir is affecting that water quality. LAV was selected to represent the lake conditions upstream, to be able to investigate how the reservoir affects the conditions upstream. OVR and LAN were added to the monitoring in 2013, 20 years after the monitoring programme began, their purpose was to determine how activities in the catchment area and inflow between LAV and SKM could affect the Hg concentrations in SKM.
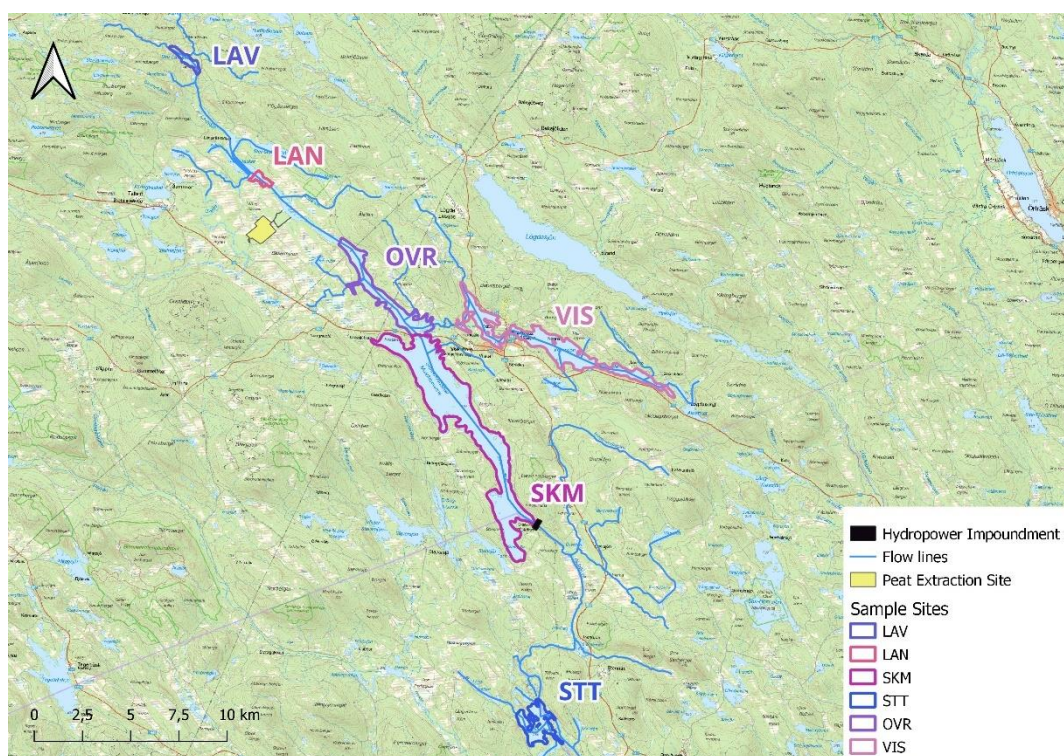
Figure 4. Map illustrating how the sampling sites are situated and relate to each other in the environment. Background map: Topografisk webbkarta Visning, Översiktlig - v1 © Lantmäteriet 2024. Sample sites are modified from SMHI (2016a). The flow lines are modified from SMHI (2016b).

The sampled surface waters and related catchment areas are illustrated in Fig. 4 and Fig. 5, respectively. The reference lake (VIS) has, for example, the smallest area of 80 km^2, and the longest turnover time (Table 1). The lake most downstream (STT) has the largest catchment area and hence the fastest turnover time. The lake most

upstream in the water system (LAV) is the shallowest (Table 1). SKM is larger than OVR, and the site which is closest to the hydropower impoundment (Table 1; Fig. 4), which is indicated by "hydropower impoundment" in Fig. 4. SKM and OVR were estimated to have the same catchment area (Table 1; Fig. 5). The site's catchment areas consist mainly of forest and peatland. The soil type is mostly till and peat. The forest is between 61 – 71%, and peatland is between 16–22% of all the land coverage. The soil type is between 55 – 65% till, between 6 – 9% thin soil and barren rock, and between 20 – 28 % peat (SMHI 2016e).

Table 1. Physical and hydrological characteristics of the sample sites. Since SKM and OVR have the same catchment area and are in the same lake, they have some identical characteristics. The catchment areas are derived from SMHI (2016c, 2016d). The water bodies' lake area and height above the sea are extracted from SMHI (2016e) and their average depth and turnover time are collected from SMHI (2016f). Some lake areas which weren't covered in these datasets have been calculated in QGIS. In SMHI (2016f) they have modelled SKM, OVR and LAN as one single lake.

	SKM	OVR	LAN	STT	LAV	VIS
Catchment area (km^2)	1420	1420	1225	1558	155	80
Lake area (km^2)	18	4.3	0.43	2.5	0.6	8.1
Height above the sea of the waterbody (m)	275	275	283	235	288	290
Lake turnover time (years)	0.77	0.77	0.77	0.032	0.056	1.8
Average depth (m)	16	16	16	6.6	4.2	5.6
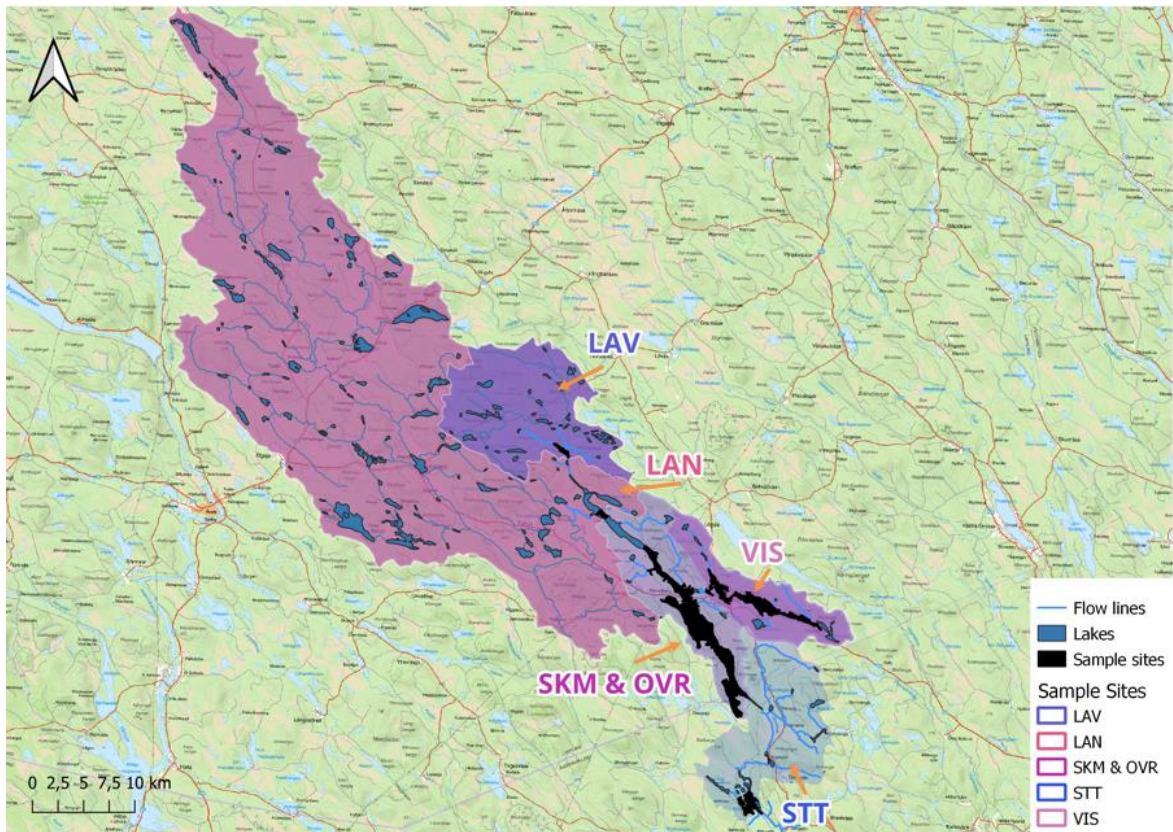
Figure 5. Map of the six sample sites' catchment areas. The arrows indicate which name belongs to which catchment area. STT has the largest catchment area while VIS has the smallest. SKM and OVR has the same. Background map: Topografisk webbkarta Visning, Översiktlig - v1 © Lantmäteriet 2024. Catchment areas are modified from SMHI (2016c) and SMHI (2016d). The lakes are modified from SMHI (2016a). The flow lines are modified from SMHI (2016b). The black sample sites are the same as in Fig. 4.

2. Method

The method consists of data collection, selection, quality assessment and calculations. Most of the raw data originates from the monitoring programme, with the rest (e.g., temperature, precipitation) obtained from open sources and Statkraft. The selection and calculation from online sources and Statkraft are accounted for in Appendix A. Appendix A contains the method for collection and calculation of temperature, precipitation, catchment areas, runoff per catchment area and forest harvest per catchment area. The selection of monitoring data has been based on the study's scope and objectives. The quality assessment of monitoring data has been conducted visually by plotting the raw data and with an established outlier exclusion method. The quality assessment also entails normalisation of THg perch concentration with the length of each perch. The calculations were made in the statistical computing and graphics programming language R, version 4.3.3; in Minitab Statistical Software 21, version 21.3.1; in Microsoft Excel version 2302; in MATLAB (R2020b, The MathWorks, Inc.) and QGIS Desktop, version 3.28.15. A result is considered statistically significant if the p-value is < 0.05. The calculations conducted on the monitoring data were: (1) temporal trend analyses on Hg variables with (a) change point analysis, (b) linear regression and (c) analysis of variance (ANOVA), (2) spatial trend analysis with one-way ANOVA and, (3) identification of potential major drivers of the potential spatial and temporal trends with principal component analysis (PCA). The environmental variables in Appendix A have been compiled to be part of the PCA analysis or/and build on the understanding of what drives the possible trends in the results.

2.1 Within the Monitoring Programme

2.1.1 Data Collection

Three different Hg variables have been measured within the monitoring programme. These are THg concentration in surface water (THg$_{aq}$, ng/L), MeHg concentration in surface water (MeHg$_{aq}$, ng/L) and THg in fish muscle for (THg$_{fish}$) for three types of fish. In this study, the only focus will be on THg$_{fish}$ for perch around 10 cm in length (mg/kg), for a motivation why the other fish types were excluded, see Appendix B. The samples have been collected with the same

methodology for most of the duration of the monitoring programme. The sampling began primarily after the impoundment occurred, in general, two to four years after the impoundment, which occurred in 1989. Measurements of Hg in surface water began in 1992 and have since 1993 been sampled for 6 to 11 months per year. Most years have been sampled for 10 months, from February to November. From 2019 the sampling only occurred between May and October. All sample sites did not become active in 1992, see Appendix C, Table C2. Collection of THg for perch around 10 cm in length began for most sites in 1991, and for other sites see Appendix C, Table C3. Until 1995 the recorded samples were of individual fishes and constituted roughly 15 fishes. From 1996 onwards about 15 perches nearly 10 cm in length were pooled into random 3 samples, where each sample consisted of a pool of 5 perches.

Besides Hg, additional environmental variables were measured in the monitoring programme. These are pH, colour, alkalinity, conductivity, and total phosphorus. The measurement of these variables began in 1984, in some sites the collection began later, see Appendix C, Table C2. These variables are as with water Hg measurements collected monthly, with a variability of which months were measured. From 2005 and onwards the trend is measurements between February - October, with more than one measurement in April and May. Usually, 1-2 measurements in April and 2-4 measurements in May.

Mercury

After collection, the water and fish samples were analysed by the IVL Swedish Environmental Research Institute laboratory. Within this monitoring period, the laboratory procedures have evolved. In 2020 the analysis methods used were the ones in Appendix E, Table E1.

Environmental Monitoring Variables

The surface water samples of the environmental monitoring variables (colour, pH, conductivity, alkalinity and total phosphorus) were analysed by the Swedish ISO 17025 accredited laboratory MoRe Research. Within this monitoring period, the laboratory procedures have evolved. In 2020 the methods used by MoRe Research were the ones in Appendix E, Table E2.

The colour variable was obtained by filtering the water samples through a membrane filter with a pore size of 0.45 µm. Then the filtered water in a 5 cm cuvette was analysed for absorbance at 420 nm (abs/5 cm), this method is described in Table E2. After this, the result was recalculated into water colour (abs*500) by MoRe Research with the equation:

$$Water\ colour \approx Abs_{420\ nm} \times 500$$

Absorbance at 420 nm portrays the quantity and quality of OM (Eklöf et al. 2012).

2.1.2 Data Quality Assessment and Selection

Suspected Outlier Removal

There are some definite outliers in the Hg water dataset, see Appendix B, Fi B1; B2. Several studies found clear variations of methylation between summer and winter periods (Levanoni et al. 2015). In this dataset, no such trend was related to the outliers. It was hence deemed relevant to use an outlier removal method for this dataset. Standard outlier tests pool the whole dataset together. For the $MeHg_{aq}$ dataset, there is a sharp decrease in Hg levels. Pooling the levels before and after the decrease together could wrongly deem "true" values to be outliers because they were measured before the decrease. This is what occurred for some of the MeHg sites, mainly for the MeHg values in LAV. The removal of outliers with the pooling method (described below) was continued despite this and the "cleaned" data is what the analysis is made of. In total the outlier removal method resulted in the removal of 31 $MeHg_{aq}$ outliers and 6 THg_{aq} outliers. 2 additional $MeHg_{aq}$ outliers were removed which were assumed to be notetaking errors. For figures of the datasets before and after outlier removal, see Appendix B, Fig. B1; B2.

The method of x × Median Absolute Deviation (MAD) for outlier removal has been used for several time series analyses (Leys et al. 2013; Ammar et al. 2024). For our dataset, 3×MAD is considered to be the most suitable since that method targets the visually estimated outliers and keeps the number of removed values to a minimum. See Appendix B for a more extensive motivation for the choice of method.

The method:

$$MAD = median\ (|x_i - median(x)|)$$

MAD is the median of the absolute difference of each residual x_i and the *median(x)* value of each site's log-transformed concentrations (either THg or MeHg). A sample is considered an outlier if its corresponding residual surpasses or fails the:

$$T_{max} = median(x) + (\alpha \times MAD)$$
$$T_{min} = median(x) - (\alpha \times MAD)$$

where α in this case corresponds to 4.4478. α is obtained from 3 times the constant 1.4826, which aids the MAD to be consistent with the normal distribution (Adapted from Ammar et al. 2024).

Under Limit of Quantification

The detection limit (LOD) and limit of quantification (LOQ) used in the IVL lab changed during the 30-year sampling period. In 2009 the lab switched the documenting system from reporting LOD to reporting LOQ. When a concentration is under LOQ it can't be reliably determined. What is known is that the concentration is somewhere between zero and LOQ. To be able to include the concentrations below LOQ in analyses standard procedure is to either divide it in

half or divide it by the square root of 2. In the dataset the current THg_{aq} LOQ limit of 0.2 ng/L where never reached. The $MeHg_{aq}$ LOQ limit of 0.06 ng/L where on the other hand reached 143 times. The concentrations under LOQ were substituted with LOQ/√2, as in (Haque et al. 2023).

Normalisation of Fish Concentrations

Fish Hg concentration is frequently observed as a function of fish size (e.g., length, weight), because of bioaccumulation (Braaten et al. 2019; Médieu et al. 2021). A power relationship between the fish length and the Log10 (observed Hg) concentration was fitted in the reference lake (VIS) (Fig. 6).

The equation shown in the figure:

$$LogHg = 0.42 \times FL^{0.30} - 2.79$$
$$R^2 = 0.58$$
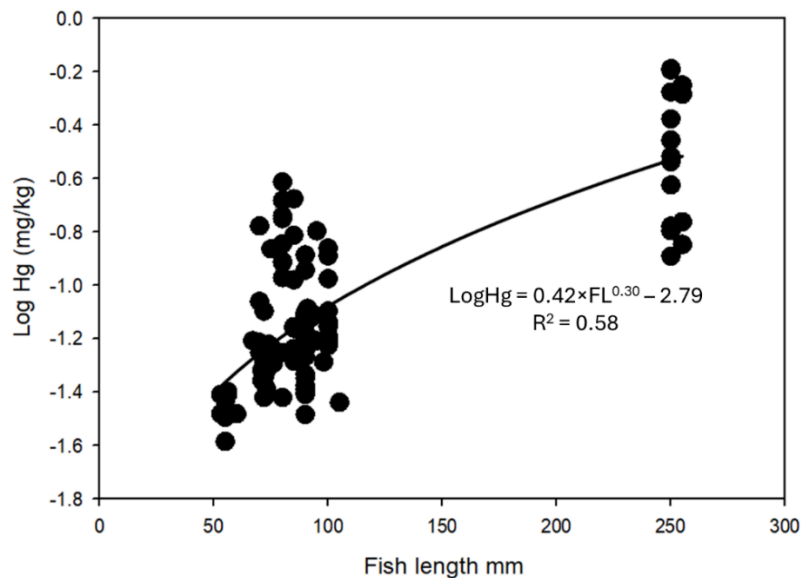
Figure 6. Normalisation equation for normalisation of concentrations of THg in perch to the length of 10 cm.

This was accomplished by first log-transforming the perch THg concentrations. Log-transformation was performed to enhance the homogeneity of variance, thereby increasing the robustness of the statistical analysis (Zuur et al. 2010; Médieu et al. 2021). Subsequently, a power relationship between the fish length of perch samples and the responding THg concentrations was determined using SigmaPlot 14, where FL in the equation represents the fish length. This method is similar to that described by Médieu et al. (2021).

As VIS has undergone minimised anthropogenic perturbation in comparison to the hydropower reservoir system, the power relationship was therefore only fitted on samples from VIS. We use the power relationship derived from VIS to standardize fish Hg concentration from both the VIS and hydropower reservoir system (SKM, OVR, LAN, STT and LAV). The initial fish sampling programme was designed to target two groups of perches that are around 100mm and 250mm. The group of perches around 250 mm were however excluded for further analysis in this study, see Appendix B for motivation. The individual and pooled perch

concentrations in the 100 mm cluster were normalised into 100 mm concentrations. The standardized fish Hg concentration is used for further analysis.

Yearly averages

To determine if yearly averages were applicable for this analysis the "monthly" $MeHg_{aq}$ concentrations, which are known to be affected by temperature, were compared with air temperature, seasons, and snow-covered/non-snow-covered months. No significant correlation between these factors and the $MeHg_{aq}$ concentrations where found, which resulted in yearly averages being deemed suitable for this study's analyses.

Raw data support parameters

The raw data for the support parameters were only available from 2005 and onwards, for earlier years only the yearly average was procurable. From 2005 the yearly arithmetic mean was calculated from the dataset to enable comparison with earlier years.

Limitations

The monitoring programme has collected samples from eleven sites; six of these sites are included in the analysis (Fig. 4) and five have been excluded, see Appendix B and C. There are some gaps in the sampling, see Appendix C, for example for colour and the reference lake, which limits the results and understanding of the trends, see results 3. All data sampled within the monitoring programme was not available for this study. The raw data available for analysis is portrayed in Appendix D, Table D1. The quality and amount of a variable, as well as how it related to this study's scope and objectives resulted in the exclusion of some of the monitoring programme variables, see Appendix B.

2.1.3 Calculations

Temporal Trends

Change Point (CP) analysis of Hg concentration was achieved by fitting a two-piece linear regression to the Hg concentrations by using the R package "lmcp", available at https://github.com/mskoldSU/lmcp. This method was developed by Haqeu et al. (2023) to identify presence of temporal trend changes. Yearly geometric means of Hg (THg_{aq} $MeHg_{aq}$ and THg_{fish}) were used as the input of the CP calculation. These means were calculated in R from "monthly" water samples and from multiple yearly normalised fish samples. The purpose of using geometric means was to further minimise the influence of extreme values.

After CP analysis the temporal trends were further investigated through linear regression. For the time series where no significant CP was detected the linear

regression was fitted from start to end year. Where there was a gap in sampling, such as for VIS, two regressions were fitted, one for each consecutive sampling series. Where a significant CP was detected two linear regressions were fitted, one from the start until the CP and one from the CP to the end of the period. The linear regression was calculated in R with the "lm"-function. A slope coefficient (β_1) and a coefficient of determination (R^2) were extracted from each regression.

To gain a greater understanding of the temporal changes the difference in yearly geometric means between the beginning and end of the monitoring period (1993-1999, 2017-2023) was compared with a one-way ANOVA. Tukey Pairwise Comparisons were used for the comparison of the beginning and end averages with a 95% confidence interval.

Spatial Trends

To investigate the spatial difference between the sites. The Hg values of the different sites were compared to each other with a one-way ANOVA using Tukey Pairwise Comparisons. The comparison was made for periods of high importance, such as the beginning period, the CP in SKM and STT and the end period. The start and end year of a period vary between variables because of the limits of that specific dataset, see Appendix D, Table D1.

Potential Drivers

Principal Component Analysis (PCA) was carried out to determine some potential drivers for this study's evolution of Hg concentrations. The yearly geometric means of $MeHg_{aq}$ and THg_{aq} concentrations in the sites SKM, STT, LAV and VIS were compared to the yearly arithmetic average of the environmental variables: colour (abs_{420}*500), pH, conductivity (mS/m), alkalinity (mmol/L HCO_3), total phosphorus (µg/L) and air temperature (°C). A yearly sum of precipitation (mm), a yearly amount of forest area logged within the catchment area (km^2), and yearly catchment runoff (km^3) calculated from yearly air temperature and yearly sum of precipitation was also included in the PCA. See Appendix A for how the environmental variables which are not included in the monitoring programme were collected and calculated, see 2.1.1. under "Environmental monitoring variables" for how the environmental monitoring variables were collected and calculated. THg_{fish} was not included in the PCA since it did not have enough data to perform a PCA on. The specific environmental variables were analysed since they were accessible for this study, varied during the monitoring period, and could have a possible impact on the Hg concentrations. Colour has for example been identified in many studies to be correlated to both THg_{aq} and $MeHg_{aq}$ (see 1.2.2).

The PCA was carried out in MATLAB. Firstly, the data was normalised by the "zscore" function. Secondly, the "pca" function was used to calculate the PCA results, visualised as loading plots in Fig. 9. Lastly, the correlation coefficient and *p*-values between the components were calculated with the "corrcoef" function, visualised in Pearson correlation matrices Fig. F1, Appendix F.

3. Results

The results consist of 3 different parts: temporal trends, spatial trends, and analysis of potential drivers. For a trend to be viewed as significant the p-value should be less than 0.05.

3.1 Temporal Trends

The results of temporal trends look at trends beginning two (1991) to four (1993) years after impoundment. It has been determined by data not included in these results that the increasing trend from impoundment occurred in the years before this analysis. The focus on the results is hence to determine the subsequent decline in concentration, after the primary drastic increase.

Within the reservoir (SKM) the concentrations of $MeHg_{aq}$ and THg_{fish} are 60% respectively 70% lower in present-day (2017-2023; 2016-2023) than they were in the years right after the impoundment (1993-1999; 1991-1996) (ANOVA, $p < 0.05$, Table 2). The high $MeHg_{aq}$ and THg_{fish} concentrations after impoundment sharply declined to 0.018 ng/L per year respectively 0.09 mg/kg per year until they stabilised (Fig. 7, $p < 0.05$). The $MeHg_{aq}$ levels in SKM reached stabilisation in 2006, 17 years after impoundment (Fig. 7, CP, $p < 0.05$). THg_{aq} did on the other hand not change significantly over the 30 investigated years (ANOVA, $p > 0.05$, Table 2; Fig. 7).

In the lake downstream of the reservoir (STT), the temporal trends were similar to those in SKM. The THg_{aq} concentration in STT had no significant change between 1993 (four years after impoundment) and present-day (ANOVA, $p > 0.05$, Table 2; Fig. 7). In STT the concentrations were 70% lower for $MeHg_{aq}$ and 80% lower for THg_{fish} in the present day than they were after impoundment (ANOVA, $p < 0.05$, Table 2). From the high levels straight after impoundment the concentrations had a sharp decrease of 0.012 ng/L $MeHg_{aq}$ and 0.083 mg/kg THg_{fish} per year until they reached stabilisation (Fig. 7, $p < 0.05$). The $MeHg_{aq}$ concentration was stabilised in 2016 (Fig. 7, CP, $p < 0.05$), 27 years after impoundment.

In the lake upstream of the reservoir (LAV), the $MeHg_{aq}$ concentration also decreases with time. Here the decline was 0.003 ng/L per year (Fig. 7, $p < 0.05$),

which is 6 times smaller than the decline in the reservoir. The decrease in LAV results in 35% lower levels of $MeHg_{aq}$ concentration in 2012-2018 compared to those in 1996-2002 (ANOVA, $p < 0.05$, Table 2). There is no CP in $MeHg_{aq}$ concentration within the LAV site (CP, $p > 0.05$). There was not enough data to be able to determine possible trends of THg_{fish} in LAV. The THg_{aq} concentrations at the beginning have no significant change just in SKM and STT. In 2011 a significant CP (Fig. 7, CP, $p < 0.05$) does however imply a decreasing trend in the THg_{aq} concentrations of 0.075 ng/L until its sampling end in 2018 (Fig. 7, $p < 0.05$), ending 5 years earlier than the rest of the sites. Looking at the whole time series there is no significant decrease in THg_{aq} concentration in LAV (ANOVA, $p > 0.05$, Table 2).

Between LAV and SKM two sites (OVR and LAN) have been sampled during a shorter period (the last 10 years). For these smaller datasets, there was no change in $MeHg_{aq}$ or THg_{fish} concentration ($p > 0.05$, Fig. 7; Table 2). The THg_{aq} concentration did however increase by 30% in OVR and 20% in LAV between 2013-2015 and 2021-2023 (ANOVA, $p < 0.05$, Table 2). THg_{aq} increased with 0.09 ng/L per year in OVR and 0.07 ng/L per year in LAN ($p < 0.05$, Fig. 7).

For the reference site (VIS) the $MeHg_{aq}$ and THg_{fish} concentrations decreased by 40% respectively 45% during these 30 years (ANOVA, $p < 0.05$, Table 2). The rate of this decrease was not able to be determined because of a gap in the dataset (Fig. 7). The THg_{aq} concentrations in VIS had no significant change during the monitoring period (ANOVA, $p > 0.05$, Table 2).

*Table 2. ANOVA results of temporal trends in $MeHg_{aq}$, THg_{aq} and THg_{fish} in the sites SKM, STT, LAV, OVR, LAN, and VIS. A comparison of the difference in means at the beginning and end of the measuring period. The means and standard deviation (st.d.) are calculated in ANOVA from $3 - 7$ yearly geometric means. "n" specifies the number of samples. p-value and R2 are statistics of the test, where * indicates a significant temporal trend.*

Site	Response variable	n	Time Interval		Mean ± St.d		p-value	R^2 (%)
	(unit)		Beginning End		Beginning End			
SKM	THg_{aq} (ng/L)	7	1993 – 1999		2.4 ± 0.28		0.64	1.9
		7	2017 – 2023		2.5 ± 0.33			
	$MeHg_{aq}$ (ng/L)	7	1993 – 1999		0.26 ± 0.066		0*	75
		7	2017 – 2023		0.096 ± 0.030			
	THg_{fish} (mg/kg)	6	1991-1996		0.39 ± 0.18		0.003*	60
		6	2016, 2018-2019, 2021-2023		0.11 ± 0.017			
STT	THg_{aq} (ng/L)	7	1993 – 1999		2.5 ± 0.31		0.47	4.4
		7	2017 – 2023		2.4 ± 0.24			
	$MeHg_{aq}$ (ng/L)	7	1993 – 1999		0.29 ± 0.072		0*	82
		7	2017 – 2023		0.081 ± 0.023			
	THg_{fish} (mg/kg)	6	1991-1996		0.38 ± 0.16		0.001*	67

		6	2016, 2018-2019, 2021-2023	0.086 ± 0.018		
LAV	THg_{aq} (ng/L)	7	1996 – 2002	2.3 ± 0.33	0.10	21
		7	2012 - 2018	2.0 ± 0.23		
	$MeHg_{aq}$ (ng/L)	7	1996 – 2002	0.15 ± 0.020	0*	79
		7	2012 - 2018	0.095 ± 0.0063		
	THg_{fish} (mg/kg)	2	1994, 1996	0.11 ± 0.0046	Not enough	data
		2	2016, 2018	0.075 ± 0.011		
OVR	THg_{aq} (ng/L)	3	2013 – 2015	2.0 ± 0.20	0.017*	79
		3	2021 - 2023	2.6 ± 0.22		
	$MeHg_{aq}$ (ng/L)	3	2013 – 2015	0.11 ± 0.031	0.93	0.22
		3	2021 - 2023	0.10 ± 0.059		
	THg_{fish} (mg/kg)	2	2015 – 2016	0.13 ± 0.0064	Not enough	data
		2	2022 - 2023	0.14 ± 0.017		
LAN	THg_{aq} (ng/L)	3	2013 – 2015	2.1 ± 0.17	0.046*	67
		3	2021 – 2023	2.5 ± 0.21		
	$MeHg_{aq}$ (ng/L)	3	2013 – 2015	0.092 ± 0.016	0.87	0.82
		3	2021 – 2023	0.089 ± 0.022		
	THg_{fish} (mg/kg)	2	2015 – 2016	0.10 ± 0.0091	Not enough	data
		2	2022 - 2023	0.12 ± 0.023		
VIS	THg_{aq} (ng/L)	7	1993 – 1999	1.3 ± 0.22	0.44	5.0
		7	2017 – 2023	1.4 ± 0.24		
	$MeHg_{aq}$ (ng/L)	7	1993 – 1999	0.085 ± 0.012	0*	70
		7	2017 - 2023	0.052 ± 0.012		
	THg_{fish} (mg/kg)	6	1991-1996	0.098 ± 0.047	0.046*	34
		6	2016, 2018-2019, 2021-2023	0.054 ± 0.010		

SKM

$R^2 = 0.010$
$\beta_1 = -0.0036$
$p = 0.60$

OVR

$R^2 = 0.60$
$\beta_1 = 0.094$
$p = 0.0050*$

LAN

$R^2 = 0.47$
$\beta_1 = 0.070$
$p = 0.021*$

THg$_{aq}$ (ng/L)

STT

$R^2 = 0.051$
$\beta_1 = -0.0079$
$p = 0.22$

LAV

$R^2_a = 0.0074$
$\beta_{1ia} = 0.0044$
$p_{ia} = 0.75$

CP = 2011

$R^2_b = 0.63$
$\beta_{1b} = -0.075$
$p_b = 0.019*$

VIS

$R^2_a = 0.15$
$\beta_{1ia} = -0.039$
$p_a = 0.39$

$R^2_b = 0.017$
$\beta_{1b} = -0.010$
$p_b = 0.70$

THg$_{aq}$ (ng/L)

SKM

CP = 2006

$R^2_a = 0.84$
$\beta_{1ia} = -0.018$
$p_{ia} = 4.8e-06*$

$R^2_b = 0.016$
$\beta_{1b} = -0.00054$
$p_b = 0.62$

OVR

$R^2 = 0.00093$
$\beta_1 = 0.00030$
$p = 0.93$

LAN

$R^2 = 0.0058$
$\beta_1 = 0.00044$
$p = 0.82$

MeHg$_{aq}$ (ng/L)

STT

$R^2_a = 0.79$
$\beta_{1ia} = -0.012$
$p_a = 6.1e-09*$

CP = 2016

$R^2_b = 0.083$
$\beta_{1b} = 0.0027$
$pb = 0.49$

LAV

$R^2 = 0.62$
$\beta_1 = -0.0030$
$p = 6.6e-06*$

VIS

$R^2_a = 0.16$
$\beta_{1ia} = -0.0021$
$p_{ia} = 0.36$

$R^2_b = 0.0026$
$\beta_{1b} = -0.00019$
$p_b = 0.88$

MeHg$_{aq}$ (ng/L)

29

Figure 7. Time series of THg$_{aq}$, MeHg$_{aq}$ and THg$_{fish}$ at the 6 sites SKM, OVR, LAN, STT, LAV and VIS. The dots represent the data after quality assessment and selection, i.e., outliers have been removed (for scatterplots with outliers included, see Appendix B, Fig. B1; B2), and the perch values have been normalised with length. The straight dark lines and shadow illustrate the linear regression on geometric yearly averages and its confidence interval. β$_1$ is the slope coefficient of the linear regression, and p is the p-value of β$_1$. A significant β$_1$ is indicated with *. R^2 provides the coefficient of determination of the regression model. The vertical lines indicate which year a significant CP occurs. For those series with a CP, there are two regressions.

3.2 Spatial Trends

During the monitoring period, the concentrations of THg$_{aq}$ were constantly significantly lower in the reference site (VIS) than in all other sites (Fig. 8 a). Right after inundation (period 1) SKM and STT had 2 times higher THg$_{aq}$ concentration than VIS had. 30-34 years after inundation (period 4) the sites SKM and STT had still 2 times higher THg$_{aq}$ concentrations than VIS (Fig. 8 a).

The MeHg$_{aq}$ concentrations in SKM and STT were significantly higher than the concentrations in both VIS and LAV during period 1 (1993-1999) (Fig. 8 b). The MeHg$_{aq}$ concentrations in SKM were 3 times higher and in STT they were 3.5 times higher than that in VIS. The mean concentration of MeHg$_{aq}$ in LAV is during this period 1.6 times higher than that in VIS, but LAV does not differ significantly from the concentration in VIS (Fig. 8 b). In period 2 (2006-2010), when the MeHg$_{aq}$ concentrations in SKM had stabilised (according to Fig. 7), the levels in SKM had decreased to be comparable to those in LAV. The MeHg$_{aq}$ concentration in STT was significantly higher than the concentrations in both SKM and LAV (Fig. 8 b). After STT was stabilised in 2016 (according to Fig. 7) (period 3) there were no significantly determined differences between the MeHg$_{aq}$ concentrations in either STT, SKM and VIS (Fig. 8 b). In period 3 the MeHg$_{aq}$ concentrations in LAV and OVR were instead the only sites with a determined significantly higher concentration than that in VIS (Fig. 8 b). In recent years, 30-34 years after impoundment (period 4), the mean MeHg$_{aq}$ concentrations exhibited were around 2 times higher than those in VIS. SKM is 1.9 times higher, STT is 1.6 times higher, OVR is 1.8 times higher, and LAN is 1.6 higher than VIS. There is however no determined statistical difference among them (Fig. 8 b).

For THg$_{fish}$ the concentrations in SKM and STT were as well significantly higher after inundation (Fig. 8 c). The THg$_{fish}$ concentrations in SKM and STT were 4 times higher than those in the reference lake. In recent years (30-34 years after impoundment) the THg concentrations in perch around 10 cm were still significantly higher in all sites, except STT, than those in the reference. SKM has in "present day" 2 times higher THg$_{fish}$ concentration than VIS (Fig. 8 c).
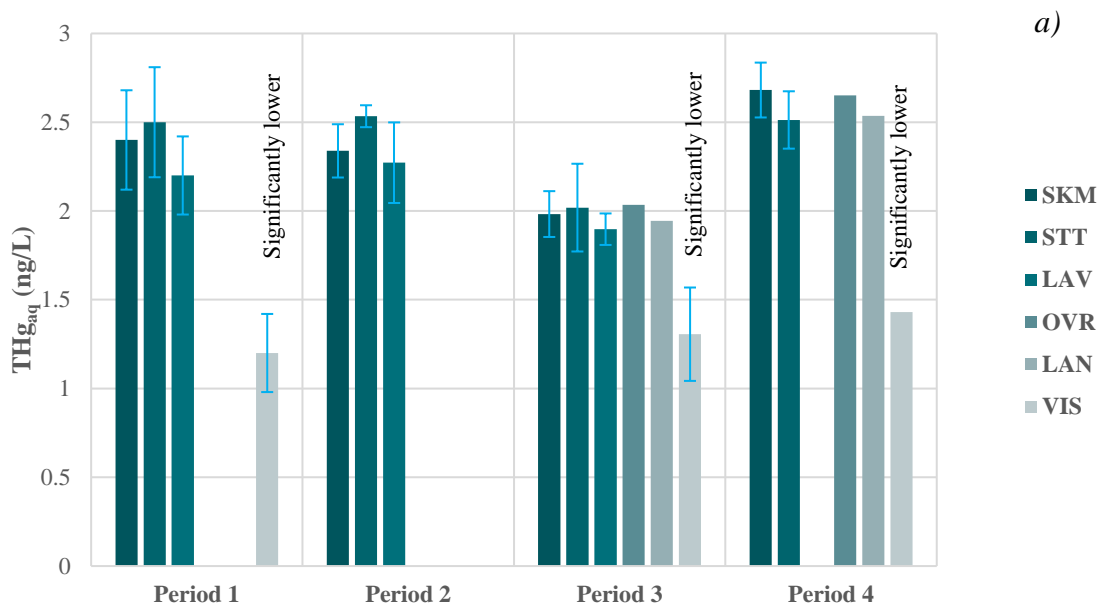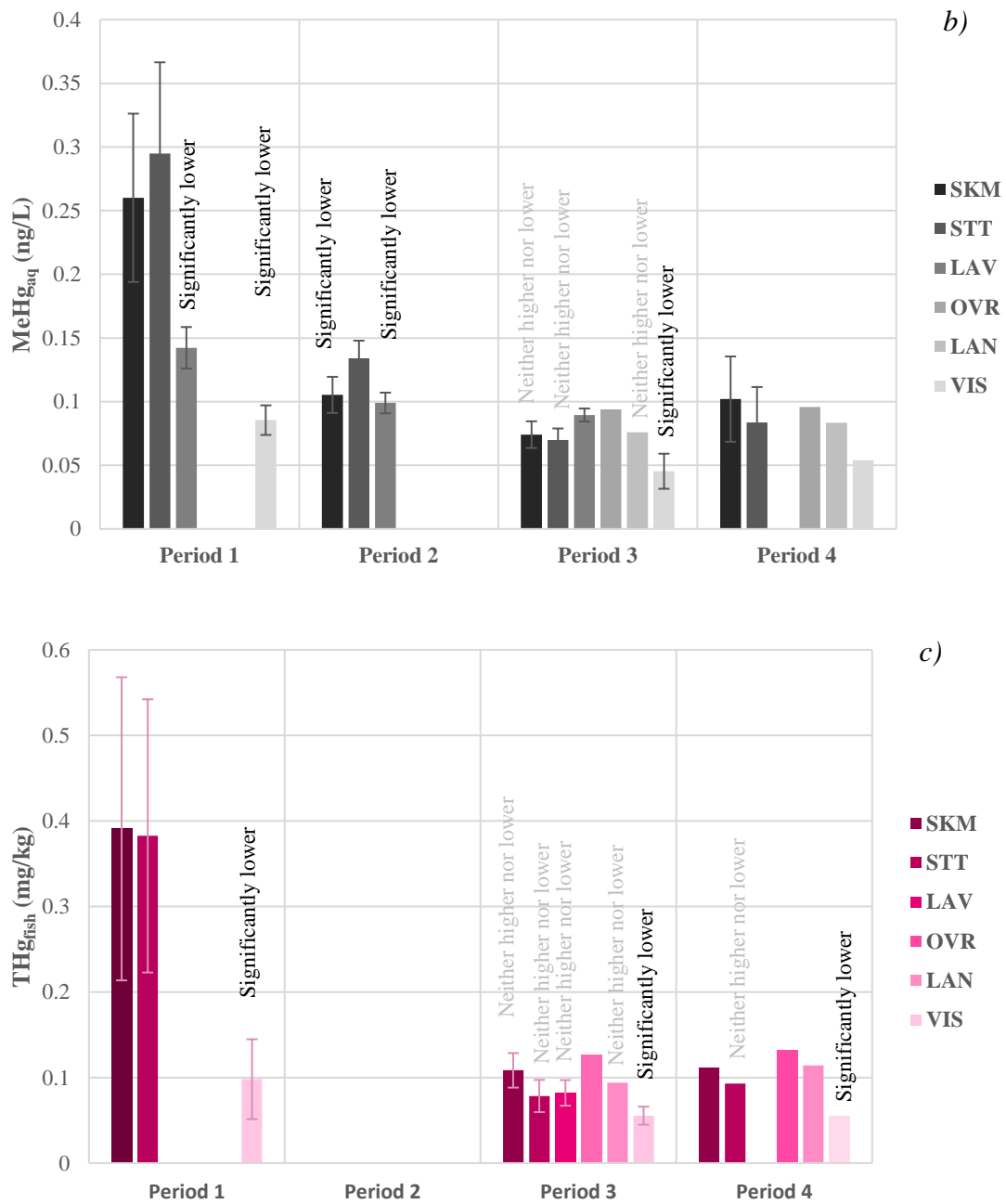
Figure 8. Spatial trend results from ANOVA where the means in sites SKM, STT, LAV, OVR, LAN, and VIS have been compared to each other. The means in question are yearly geometric means of THg_{aq} (a), $MeHg_{aq}$ (b) and THg_{fish} (c) during the monitoring period (1993-1999 [for $MeHg_{aq}$ and THg_{aq}];1991-1996 [for THg_{fish}]), the first CP 2006 (2006-2010), the third CP 2016 (2016-2018), and the "present" day (2019-2023). Which sites and years are examined vary, because of the variability of available data, exact years used for the analysis can be found in Appendix F, Table F1. The statistic test according to one-way ANOVA followed by Tukey multiple comparisons test is portrayed with the vertical texts*. For other information such as the number of geometric means which compose the period mean analysed by the ANOVA, or the exact values which the bar charts are made from, see Appendix F, Table F1.

*Bars with "significantly lower" have a significantly lower (ANOVA p-value < 0.05) concentration than at least one other site's concentration during that period. The bars with no text are significantly higher than the other sites' concentrations during that period. Bars with "neither higher nor lower" have not been determined to be significantly higher or lower than the other sites concentrations during that period by Tukey's multiple comparisons test.

3.3 Potential Drivers

An analysis of potential drivers of the variation in $MeHg_{aq}$ and THg_{aq} concentrations has been conducted through a PCA. The PCA has used yearly averages and sums for an extent of 30 years (1993-2023). The results are presented with loading plots (Fig. 9 a, d, g, j), one for each investigated site (SKM, STT, LAV and VIS), and Pearson correlation matrices (Fig. 9 b, c, e, f, h, i, k, l). The loading plots portray how the variables in the PCA relate to each other through two principal components (PC1 and PC2). Variables which are close to each other have a close relation to each other. The matrices are a selected visualisation of the Pearson correlation matrices (see Appendix F, Fig. F1 for the complete matrices). The selection of the matrices was made to simplify it by focusing on the results relevant to the analysis aim. The red values in the matrices are significant ($p < 0.05$) correlations. The middle row with matrices (Fig. 9 b, e, h, k) visualise correlations between THg_{aq} and the potential environmental drivers as well as $MeHg_{aq}$. The farthest right row contains matrices (Fig. 9 c, f, i, l) visualise the correlations between $MeHg_{aq}$ and the potential environmental drivers as well as THg_{aq}. The correlation between two variables can be either negative or positive. Variables have a positive relationship when they move together in the same direction. They have a negative relationship when they move in opposite directions, for example when one variable decreases in concentration the other variable increases.

From the loading plots (Fig. 9 a, d, g, j) it's observable that how the variables relate to each other varies substantially in the four sites. This implies that there could be different underlying conditions in the different sites.

The main factors co-varying with the $MeHg_{aq}$ variation in these study sites were THg_{aq}, colour, and pH (Fig. 9 c, f, i, l). The relation between the environmental variables and $MeHg_{aq}$ concentrations varies between sites (Fig. 9 a, c, d, f, g, i, j, l), implying that there could be different underlying reasons for changes in concentration.

Inside the reservoir (SKM) the $MeHg_{aq}$ concentration did not correlate with THg_{aq} concentrations (Fig. 9 c), colour did however significantly positively correlate to them both (0.57 respectively 0.73) (Fig. 9 b, c). In SKM the concentration of THg_{aq} had a 20% greater correlation to colour than $MeHg_{aq}$ (Fig. 9 b, c).

In the lake downstream of the reservoir (STT) the MeHg$_{aq}$ concentrations did not correlate to colour (Fig. 9 e), instead they were only significantly positively correlated to THg$_{aq}$ and significantly negatively correlated to pH (Fig. 9 f). THg$_{aq}$ was also negatively correlated to pH (Fig. 9 e). The THg$_{aq}$ concentrations in STT were additionally positively correlated to colour and negatively correlated to alkalinity (Fig. 9 e).

In the lake upstream of the reservoir (LAV) the yearly average MeHg$_{aq}$ and THg$_{aq}$ concentrations were again positively correlated (0.72) (Fig. 9 h, i). MeHg$_{aq}$ was also significantly correlated to pH and colour, negatively respectively positively (Fig. 9 i). MeHg$_{aq}$ correlated similarly to all these three factors (Fig. 9 i). The THg$_{aq}$ concentrations correlated to the same variables as in STT (Fig. 9 h).

In the reference lake (VIS) the MeHg$_{aq}$ and THg$_{aq}$ concentration were positively correlated (Fig. 9 k, l). MeHg$_{aq}$ was also correlated to pH, colour (0.89) and precipitation (Fig. 9 l). THg$_{aq}$ was also correlated to colour (0.93) (Fig. 9 k).

Loading Plot	**Correlations of THg$_{aq}$**	**Correlations of MeHg$_{aq}$**
a)	b)	c)

d)

STT

e)

	THg
MeHg	0.60
THg	1.00
pH	-0.72
Color	0.77
Alkalinity	-0.70
Conductivity	-0.28
TP	0.15
Temp	-0.24
Prec.	0.18
Flow	0.26
Deforest	-0.07

f)

	MeHg
MeHg	1.00
THg	0.60
pH	-0.69
Color	0.25
Alkalinity	-0.14
Conductivity	0.17
TP	-0.10
Temp	-0.16
Prec.	0.17
Flow	0.23
Deforest	0.10

g)

LAV

h)

	THg
MeHg	0.72
THg	1.00
pH	-0.81
Color	0.81
Alkalinity	-0.62
Conductivity	-0.16
TP	-0.27
Temp	-0.29
Prec.	0.41
Flow	0.49
Deforest	-0.43

i)

	MeHg
MeHg	1.00
THg	0.72
pH	-0.58
Color	0.70
Alkalinity	-0.43
Conductivity	-0.00
TP	-0.06
Temp	-0.47
Prec.	0.22
Flow	0.38
Deforest	-0.54

Figure 9. PCA results of MeHg$_{aq}$, THg$_{aq}$ and potential environmental drivers* presented by loading plots (left) and the significant correlations from the Pearson correlation matrices portrayed (middle and right). Significant (p < 0.05) correlations in the matrices are indicated with red font colour. The middle row of matrices visualises correlations between THg$_{aq}$ and the potential environmental drivers* + MeHg$_{aq}$. The right row of matrices portrays the correlations between MeHg$_{aq}$ and the potential environmental drivers* + THg$_{aq}$. The PCA has been performed on the sites SKM, STT, LAV and VIS.

* *Colour* → yearly average water colour (abs$_{420}$*500), *conductivity* → yearly average conductivity (mS/m), *alkalinity* → yearly average alkalinity (mmol/L HCO$_3$), *pH* → pH, *TP* → yearly average total phosphorus (µg/L), *prec.* → yearly sum of precipitation (mm), *temp.* → yearly average air temperature (°C), *flow* → yearly catchment runoff (km^3), *deforest* → yearly area logged in the catchment area (km^2)

4. Discussion

The discussion aims to answer the study objectives.

4.1 Temporal and Spatial Trends

4.1.1 THg$_{aq}$

The surface water THg$_{aq}$ concentrations in the reservoir (SKM), downstream (STT) and in the reference site (VIS) have had no significant change between 1993 to 2023 (Table 2; Fig. 7). All spatial THg$_{aq}$ trends are constant during the 30 years of monitoring (Fig. 8 a). A similar trend has been observed in De Bonville et al. (2020).

The conditions in the reference site are assumed to be representative of a natural boreal lake. Similar results as those in the reference site have been recorded in two other studies. Pettersson and Haglund (2023) found no significant change in runoff THg during the last 30 years in two northern Sweden catchments. Eklöf et al. (2012) did not observe any significant temporal changes of THg in surface water runoff between 2000-2010 from 19 boreal catchments across Sweden. During the monitoring period (1993-2023) two changes in aquatic environments have been recorded throughout Europe. None of these changes seems to have had a significant impact on the THga$_q$ surface water concentrations in this study nor on the runoff in the studies Eklöf et al. (2012) and Pettersson and Haglund (2023). One of the changes is that there has been a general increase in freshwater DOC across Europe (Evans et al. 2006). This trend is observable in this study as well (see Appendix F, Fig. F2. for the temporal trends of DOM in the sample sites.) The other change is that the atmospheric deposition of Hg^{II} over Europe has declined (Zhang et al. 2016). That the surface water THg$_{aq}$ has had no significant change implies that the amount of Hg which enters and leaves the surface water of the aquatic system is similar throughout the period. One of the main ways that Hg is presumed to enter ecosystems is with runoff from its catchment area. The Hg in runoff mainly originates from the Hg legacy in organic-rich soils (Eklöf et al. 2012; Bishop et al. 2020). The highly consistent stable THg$_{aq}$ concentrations suggest that the surface water concentrations are controlled by the catchment discharge of legacy Hg that is

stored in the forest and peat soils, rather than the contemporary atmospheric rainfall or DOM increase. These results highlight that the current concentration of THg_{aq} in the reference may be sustained for a long time. The consistently stable THg_{aq} concentrations in the reservoir and the lake downstream of the reservoir are assumed to be controlled similarly by catchment discharge.

The temporal and spatial analyses are conducted on THg_{aq} concentrations 4 years after the reservoir creation; hence the analyses cannot say what transpired during those first 4 years. Measurements before the construction are not available to compare with. With the inundation following hydropower construction organic-rich soils containing Hg complexes were flooded. These Hg-OM complexes likely leach into the water and some of them can have relocated to the surface water. During these first 4 years there hence could have been a fast incline and subsequently a stabilisation of THg_{aq} concentrations. But since the measurements are on surface waters there might only have been a slight spike in concentrations.

All sites within the hydropower system which are deemed impacted by the reservoir construction have significantly higher THg_{aq} concentrations than the reference site right after construction, this difference is constant and still exists 34 years after construction (Fig. 8 a). Right after inundation, the reservoir (SKM) and the lake downstream the reservoir (STT) both have 2 times higher THg_{aq} concentration than the reference lake (VIS). 30 years after inundation they are still 2 times higher than VIS (Fig. 8 a). These spatial trends do not correlate to the findings of De Bonville et al. (2020). De Bonville et al. (2020) investigated the variation of Hg functional pools in natural and newly impounded parts of the aquatic network of a watershed in Northern Quebec, Canada. They found that THg concentrations in surface waters were relatively constant along the network. They also found that THg was related to pools of DOC.

There are three hypothesises on why the THg_{aq} concentrations in the hydropower-affected system are continuously 2 times higher than the reference site:

- o With hydropower impoundment comes a change in the water regime. With a slower water velocity and increased retention times (Rosell et al. 2005 see Levanoni et al. 2015; Meentemeyer et al. 1999 see Levanoni et al. 2015). The THg_{aq} concentrations might have increased during the 4 first years after impoundment, such as in Hall et al. (2005). Possibly are the potentially increased retention times in the hydropower system keeping the concentrations elevated.
- o The inundated soil that before inundation was organic-rich peat and forest soil might still be leaching OM-Hg complexes into the water. If this is the case the input of these Hg complexes will eventually stop and a subsequent reduction in THg_{aq} concentrations will occur.
- o A hypothesis which would agree with the De Bonville et al. (2020) results is that hydropower impoundments do not have a significant long-

term impact on the THg concentrations in the water system. In Figure 4 in Hall et al. (2005) the THg concentrations after impoundment have a fast increase and a subsequent increase. Possibly the THg_{aq} concentrations had a significant impact during the first 4 unrecorded years and had time to return to background levels before the monitoring period began. That the hydropower system has significantly elevated concentrations compared to the reference site might instead be caused by lake variability. THg_{aq} concentrations are well-known to be controlled by catchment discharge (Munthe et al. 2007; Eklöf et al. 2012; Moslemi-Aqdam et al. 2023). The reference site has the smallest catchment area of the sample sites (Table 1; Fig. 5). The hypothesis would be that the difference in catchment discharge between the reference site and the hydropower water system could be why the concentrations in the reference site are lower.

This study is not able to determine if any of these hypotheses are correct. Further sampling and analyses would be needed to achieve this. To determine the representation of the reference site as a reference other lakes which could be considered references inside the catchment areas should be sampled and compared.

For the temporal trends in the lake upstream (LAV) and in the sites between the reservoir and the lake upstream, here written as upstream 1 (OVR) and upstream 2 (LAN), other results are observed (Table 2; Fig. 7). These 3 sites have shorter datasets, the lake upstream was sampled between 1996 – 2018 while upstream 1 and upstream 2 were sampled between 2013 – 2023. It could be argued that the increasing and decreasing trends in these 3 sites are significant since they cover a smaller timeframe. A smaller timeframe could find significant trends in what is observable as natural fluctuations in the longer timeframes in the reservoir and downstream (Fig. 7). What has caused these trends is unclear. An increase in THg could originate from increased OM-Hg complexes in the catchment runoff (Eklöf et al. 2012; Bishop et al. 2020), however, no such comparative data were available for verification.

4.1.2 MeHg$_{aq}$

The reference site (VIS) has had a 40% decrease in surface water MeHg concentration during the 32-year monitoring period (Table 2). The 40% decrease is calculated from a comparison of the average MeHg concentration between the beginning and end period in the reference site (1993-1999 and 2017-2023) (Table 2). This decline could be an effect of the reduced atmospheric Hg deposition in Europe during this period. The reduction of Hg deposition has not reduced THg levels, as mentioned above in 4.1.1. More than 90% of the THg in freshwater ecosystems is bound into complexes with OM, which hinders its methylation (Ullrich et al. 2001; Ravichandran 2004). With atmospheric deposition freshly

formed reactive bioavailable Hg^{II} enters the ecosystem (Driscoll et al. 2013; Saiz-Lopez et al. 2020). The bioavailability of Hg^{II} is one of the controlling factors of methylation (Paranjape & Hall 2017). This argument can explain why the $MeHg_{aq}$ concentrations are declined by reduced atmospheric Hg deposition while the THg_{aq} concentrations are not significantly affected.

The $MeHg_{aq}$ concentration decline in the lake upstream of the reservoir (LAV) is similar to that of the reservoir. There is a constant decrease in LAV during the monitoring of 0.003 ng/L per year. This decrease in LAV results in 35% lower levels at the end of sampling (2012-2018), compared to an average of the first 6 years of sampling in the upstream lake (1996-2002) (Table 2; Fig. 7). That the lake upstream had a smaller overall percentual change than the reference site in the analysis most likely not representative of the actual conditions in the sites. The dataset for the lake upstream has two attributes which can have caused misrepresentative results that are smaller/less than the actual conditions in the lake: The measuring period in the lake upstream began 3 years after and ended 5 years before the sampling in the reference site, the reservoir and the lake downstream did. Time series analysis on this dataset is hence not fully comparable with the longer datasets of the reservoir, the reference, and the downstream lake. The other attribute is the outlier removal. With the outlier removal method, a greater amount of the extreme values in the upstream lake dataset was removed than for the other sites datasets (see Appendix B). Most of the removed values are concentrated in the first few sampling years (see Appendix B, Figure B2). These misrepresentations are likely also one of the reasons that the $MeHg_{aq}$ concentrations in the lake upstream are not significantly larger than VIS during period 1 even though it has about a 1.6 times higher average (Fig. 8 b).

A phenomenon covered extensively by other studies (Hall et al. 2005; Roy et al. 2009; Kasper et al. 2014; Levanoni et al. 2015; Herrero Ortega et al. 2018) is that reservoirs have an immediate response of enhanced methylation after inundation from impoundment. The changes in hydrologic regime (generally decreased velocity) have shown to result in conditions beneficial for microbial methylations, such as sub-oxic and anoxic conditions, increased temperature, and increased retention times (Kasper et al. 2014; Rosell et al. 2005 see Levanoni et al. 2015; Meentemeyer et al. 1999 see Levanoni et al. 2015). The flooded vegetation and organic-rich soil in boreal areas, such as the one in this study, have been shown to have a major impact on the stimulation of methylating microbes (Hall et al. 2005; Roy et al. 2009; Herrero Ortega et al. 2018). Net MeHg production seems to be the most pronounced in the first years after inundation and gradually decreases in the following years likely as the bioavailability of the OM is reduced (Hall et al. 2005; Herrero Ortega et al. 2018).

Our dataset begins 4 years after inundation when the subsequent decline in MeHg concentrations has begun. There is a drastic 75% decline in $MeHg_{aq}$

concentrations in the reservoir (SKM) from 4 years after inundation until the concentration stabilises 17 years after impoundment (Fig. 7). There is no significant change in MeHg$_{aq}$ concentrations after the stabilisation is reached (Fig. 7). In the lake downstream of the reservoir (STT), the same trends are found (Table 2; Fig. 7). Here the MeHg$_{aq}$ concentrations decrease to a greater extent with 85% from 4 years after inundation until the concentration stabilise 27 years after inundation. That the MeHg$_{aq}$ levels downstream the reservoir are affected to a greater extent than in the reservoir itself has been found in several studies (Kasper et al. 2014; De Bonville et al. 2020). The significantly elevated MeHg$_{aq}$ concentrations and their decline are also observable in Fig. 8 b. 30-34 years after impoundment the results of the spatial one-way ANOVA found no significant difference between the means of the reference lake and the hydropower impacted sites (Fig. 8 b). The reliability of the ANOVA results for this period is however weak with a low R^2 of 28% and a high standard deviation (Fig. 8 b, Appendix F, Table F1). Further continuous monitoring is recommended to determine if there is a significant difference between the MeHg$_{aq}$ concentrations in the reference site and the hydropower-affected sites. Since a comparison of the means of the period (2019-2023) shows that all hydropower-affected sites have between 1.5-2 times higher concentrations than the reference ((Fig. 8 b), this study will assume that the concentrations are still higher than the reference site.

That the MeHg$_{aq}$ concentrations become stable 20-30 years after hydropower creation is believed to be caused by an analogous decline in bioavailable OM. This effect of bioavailable OM on MeHg$_{aq}$ concentration has been found in several studies (Hall et al. 2005; Herrero Ortega et al. 2018). The relationship between OM and MeHg has also been found in this study, see 3.3 and 4.2. When the MeHg$_{aq}$ concentrations are stable the effect of the inundation soil has hence most likely ended, and the methylation is likely instead controlled by the influx of new bioavailable OM from catchment discharge. That the MeHg$_{aq}$ concentrations in the reservoir are even after stabilisation 2 times higher than in the reference site can be motivated by the same arguments as for THg$_{aq}$ in 4.1.1. The MeHg$_{aq}$ concentrations could also be affected by another activity not mentioned in 4.1.1, namely water level fluctuations by the hydropower management. Two North American studies on a 75-year-old reservoir which is inundated 8 months per year showed that the sediment and porewater MeHg concentrations were 3 times higher in areas with water level fluctuations compared to permanently inundated sediments. They argue that these fluctuations could lead to sustained elevated MeHg concentrations in old reservoirs even after the initial impact of landscape inundation diminished (Eckley et al. 2015, 2017).

In the reservoir (SKM) the water level fluctuated from 283 m to 273 m (in monthly averages) between 2000 to 2023 (see Appendix A, under: "The Water Level in the Reservoir"). The Eckley et al. (2015, 2017) argument could because of

these ongoing water fluctuations be applicable in this reservoir (SKM). Whether these water level fluctuations are enough to cause an increased MeHg concentration in the whole hydropower water system or not would have to be further assessed.

4.1.3 THg$_{fish}$

The temporal trends of THg$_{fish}$ are generally analogous to the trends of MeHg$_{aq}$ (Table 2; Fig. 7). This is supported by the widely recognised theory that most Hg in fish exist in MeHg form (Bloom 1992).

Comparable trends are reported in various research articles. The declining trend in the reference site is consistent with findings from studies such as Braaten et al. (2019) which present that there have been consistent declines in perch THg concentrations in boreal Fennoscandia during recent decades following the reduction in atmospheric Hg deposition. Braaten et al. (2019) analysed about 50 000 freshwater fish muscle tissue THg records in boreal and subarctic Fennoscandia from 1965 until 2015. The temporal THg$_{fish}$ trends in the hydropower system (including the reservoir, downstream the reservoir, upstream the reservoir, upstream 1 and upstream 2) indicate that the levels sharply decline after the initial increase until they eventually stabilise (Fig. 7; Table 2). The THg$_{fish}$ analysis is conducted on measurements taken 2 years after impoundment. Therefore, the initial increase is not included in the analysis; instead, it's depicted in Appendix F, Fig. F3. One of several studies with similar decline and stabilisation results is Bodaly et al. (2007). Bodaly et al. (2007) determined the evolution of Hg concentration in three fish species in 14 boreal reservoirs in northern Manitoba, Canada for up to 35 years after impoundment. They found that the predatory fish species took 10 to 23 years to return to background Hg concentrations. That the fish Hg concentrations returned to background levels are different from the results from this study. In this study, the THg$_{fish}$ concentrations in young perch are 30-34 years after impoundment still significantly higher than the concentrations in the reference site (Fig. 8 c). The THg$_{fish}$ concentrations in the reservoir are here, just as for THg$_{aq}$ and MeHg$_{aq}$ 2 times higher than the reference lake (Fig. 8 c).

4.2 Potential Drivers Controlling the Concentration Evolution

The THg$_{aq}$ and MeHg$_{aq}$ concentrations have during the monitoring period generally strong correlations to colour (Fig. 9). Colour in this study comes from measurements of absorbance at 420 nm on membrane-filtered water samples. Abs$_{420}$ portray the quantity and quality of OM (Eklöf et al. 2012). A strong relationship between THg and colour/OM/DOC and MeHg and colour/DOC/OM is

well-documented (Eklöf et al. 2012; Lavoie et al. 2019). MeHg is recorded as having a variable and weaker correlation to DOC than THg (Bishop et al. 2020), which is also found here. In the reservoir THg has for example a 20% greater correlation to colour than MeHg (Fig. 9). THg_{aq} and colour are strongly correlated since DOC affects the speciation, solubility and mobility of Hg (Ravichandran 2004), for instance, the more OM in the water, the higher THg concentration there is. Much of THg is complexed with reduced sulphur functional groups in OM (Ravichandran 2004). $MeHg_{aq}$ and colour have a strong relationship since it is generally also complexed to OM, but usually to a lesser extent than THg (Ullrich et al. 2001). OM also control MeHg by stimulating methylation by providing nutrients for the methylating microbes (and demethylating microbes), providing electron acceptors, and can increase the anoxic conditions in which methylation occurs by increasing microbial activity which consumes oxygen (Ullrich et al. 2001; Levanoni et al. 2015; Paranjape & Hall 2017).

The main factors impacting $MeHg_{aq}$ variation in these study sites according to the PCA results are THg_{aq}, colour, and pH (Fig. 9). THg_{aq} affects $MeHg_{aq}$ concentration by controlling the bioavailability of Hg^{II} to methylate (Paranjape & Hall 2017). pH is negatively covarying with both THg_{aq} and $MeHg_{aq}$ (Fig. 9). When pH decreases the THg/MeHg levels increase. The correlation with pH is found in other studies as well (Roy et al. 2009; Eklöf et al. 2012). One possible motivation for this could be that low pH values could stimulate methylation and increase the availability of HgII by dissolving OM-Hg complex bindings (Roy et al. 2009). In one site (the reference lake) precipitation was only found to negatively correlate to $MeHg_{aq}$. The negative correlation means that with higher precipitation amounts the MeHg levels are lowered. This has been found in Huang et al. (2013) as well. Several variables which were not included in this study have well-documented methylation-driving abilities. Such variables are microbial community structure, iron, sulphur, and oxygen content (Levanoni et al. 2015; Bishop et al. 2020; De Bonville et al. 2020).

What drives the $MeHg_{aq}$ concentrations vary between the sites (Fig. 9), implying that there could be different conditions within the different sites. In the reservoir (SKM) the only tested variable that was identified to significantly drive the $MeHg_{aq}$ and the THg_{aq} concentration was colour (Fig. 9). This implies that the variation in OM per year between 1993 – 2023 (the years that the PCA was conducted upon) is likely a major driver of the $MeHg_{aq}$ and THg_{aq} trends in the reservoir. That OM has a great impact on the methylation in reservoirs is confirmed by other studies (Hall et al. 2005; Roy et al. 2009; Herrero Ortega et al. 2018). In the lake downstream the reservoir (STT) MeHg concentrations are not majorly driven by colour (no significant correlation). The MeHg concentrations are instead driven by pH and THg concentrations (Fig. 9). When looking at the dataset this study has of colour the measurements are on a similar level as those of SKM throughout the monitoring

period. All site's MeHg concentrations correlate with colour but STT. The THg concentration in STT does however correlate with colour. What differentiates this site from the others is that it's the farthest downstream and has the largest catchment area (see Table 1; Fig. 5). It also has the fastest turnover time (Table 1). One speculation is that much of the MeHg which enters the lake can have been formed before entering the lake, such as mentioned in Bishop et al. (2020), because of its large catchment area and downstream position. That part of the MeHg in the lake isn't created in the lake would mean that the methylation-OM correlation doesn't apply at the same level as for other sites where most MeHg in the lake is created through the methylation-OM relationship. In the lake upstream the reservoir (LAV) $MeHg_{aq}$ is covarying with both colour, THg_{aq} and pH (Fig. 9). In the reference lake (VIS) $MeHg_{aq}$ concentration is significantly correlated to both colour, THg_{aq}, pH and precipitation (Fig. 9).

The sample size of THg_{fish} was too small to conduct a PCA on. We will make the rough assumption that most of the perch Hg trends are dependent on the same variables which affect $MeHg_{aq}$ since a majority of the Hg in perch is believed to be MeHg (Bloom 1992). This assumption agrees with the similar temporal trends of MeHg and THg in Fig. 7 and Table 2. In Rask et al. (2024) pike THg is positively correlated to colour, iron, nutrients and maximum lake depth. Which most likely are factors that would affect perch in these waters as well.

4.3 Additional Discussion

4.3.1 Other Environmental Activities

There is a lot of other activity occurring in the catchment areas such as forestry, forest fires, peat mining, as well as occurrences of floating peat and floating peat management. These are all activities which is known to increase the levels of OM, and MeHg in waters. From this study, we cannot draw any conclusions regarding these variable's effect on Hg concentrations in and around the hydropower reservoir waters.

It's well established that forest harvest, especially clear-cutting leads to elevated MeHg levels in nearby waters (Kronberg et al. 2016). The PCA did however not detect any major correlation between $MeHg_{aq}$ and the forest harvest dataset (Fig. 9). A speculative motivation for this is that there might be a dissonance between forest harvest and for the affected OM-Hg complexes to enter the sample sites. This dissonance would make the PCA unable to detect the actual relationship.

The catchment area runoff had neither any correlation in the PCA (Fig. 9). There is however a well-established relationship between catchment runoff and $MeHg_{aq}$, THg_{aq} concentrations (Munthe et al. 2007; Eklöf et al. 2012; Moslemi-Aqdam et al.

2023). This is possibly because the analysis was made on yearly averages and this variation might be more observable on monthly averages.

It's known that peat contains high levels of OM and MeHg which leach into adjacent aquatic environments (Li et al. 2023; Wang et al. 2023). Speculations that can be made, without any statistical data to back them up, are that all types of peat management and where peat is in increased contact with water (as for the floating peat) could result in a release of OM-Hg complexes in the aquatic ecosystem. The occurrence of floating peat would probably increase the leaching of MeHg to the water, compared to if the peat was part of a peat land. How much these floating peats are affecting the Hg concentrations cannot be determined with this study. The site OVR is situated downstream of the peat mining site of Basarmyran (Fig. 4). When the peat mining occurred and was the most intense in Basarmyran (Fig. 2) the levels of MeHg and THg_{fish} were highest in OVR of all sample sites (Fig. 8). These findings indicate that the hydropower station was not the cause of the extra elevated levels in OVR specifically during that period.

Other studies have found a clear effect of land cover in the catchment area and the Hg levels (Moslemi-Aqdam et al. 2023). These static measures were not tested in the PCA. There is no great variance in land cover between the different sites (see 1.3.2), the great difference between the sites is instead the catchment areas and lake characteristics (see Table 1; Fig. 4; Fig. 5). These variances generate different aquatic conditions which presumably will lead to differences in Hg levels. Some of the potential effects these conditions have had on the Hg levels are brought up in the above discussion. The main attribute to the Hg concentrations in all sites within the hydropower system is however assumed to be the hydropower impoundment.

4.3.2 Future studies

Several future studies on this dataset have been suggested earlier in the discussion to elaborate the understanding of the results and confirm hypotheses. Some of these suggestions are to explore the effect of catchment discharge and how it's affecting the sample lakes. To sample more reference lakes to be able to investigate if it's the hydropower impoundment or the location of the reference lake that is resulting in the still elevated Hg concentrations. To examine if the fluctuation zone in the reservoir is continuously causing higher methylation rates. To continue the monitoring is highly recommended since it would enable future analyses on the temporal trends.

Since the monitoring dataset was large and comprehensive several variables were excluded from this analysis which would have a value to investigate further. An example of this is perch around 25 cm in length. A study on how and why the 25 cm perch samples vary in comparison to the 10 cm perch samples.

Another proposed future study is to publish these results in a paper, ensuring they reach a wider audience.

5. Conclusions

- o Skinnmuddselet hydropower impoundment has likely a significant impact on surface water THg and MeHg concentrations, as well THg concentrations in young perch.
- o With time, the impact on surface water MeHg concentrations and perch THg concentrations has subsided. It took 17 years for the MeHg concentrations within the Skinnmuddselet reservoir to stabilise and 27 years for the lake downstream the reservoir.
- o 34 years post-impoundment, the Hg concentrations in Skinnmuddselet remain twice as high as those in the reference lake.
- o Potential major drivers or co-variates of MeHg surface water concentrations include surface water colour, THg and pH.

These conclusions demonstrate long-term ecological impacts of hydropower impoundment on Hg cycling and emphasise the importance of monitoring and managing these ecosystems.

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gate.* [Photograph]. [2024-05-13]

Persson, Signe. (n.d, c). *Management of floating peat by sawing it into fragments.*
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Popular science summary

When a hydropower station is constructed a reservoir is created. The reservoir is created by impounding a river which floods a large area. The reservoir has elevated levels of methylmercury in water and fish. With time these elevated levels decline. This study has investigated what happens with this decline through time. The study has many interesting results. The two major takeaways regarding how hydropower impacts mercury long-term are: It can take about 20 years for the methylmercury concentrations in reservoirs to stop declining. For lakes downstream reservoirs it can take about 30 years for the methylmercury concentrations to come to a stable level. This decline can possibly come from a reduction in fresh organic matter with time which subsequently affects the process where methylmercury is created. Since organic matter is food for the bacteria which makes mercury into methylmercury. The other major takeaway is that the concentrations of methylmercury, and mercury in young fish and the total amount of mercury is higher in the reservoir than in an unimpacted lake. They are higher than the unimpacted lake even 30 years after the hydropower reservoir was created. The concentrations could be elevated because of how the hydropower station impacts the water speed or varies the water level or perhaps are the unaffected lake not comparable to the area in which the reservoir is located.

Methylmercury is a type of mercury which is toxic. If humans or animals consume large amounts of methylmercury the central nervous system gets damaged, which lessens for example neurological development. There are several human activities which elevate the methylmercury concentrations in fish. Flooding from hydropower construction is one of these activities. To understand how the construction is affecting mercury long-term is of great importance. Since an increased knowledge of pollutants assists us with knowledge of what preventative measures are needed.

The primary impact new reservoirs have on methylmercury is a thoroughly investigated research subject. The long-term impact has not been covered to the same extent. Making this study rare with its long qualitative dataset.

Acknowledgements

Appendix A – Methods of Data Outside of The Monitoring Programme

Data Collection, Selection, Quality Assessment and Calculations

Temperature and Precipitation

Monthly average air temperature and monthly sum of precipitation were extracted from the dataset CRU TS (Climatic Research Unit gridded Time Series) version 4.07 Google Earth Interface from the University of East Anglia Climatic Research Unit (2008). This is a climate dataset on a 0.5° by 0.5° grid over most of the world's land surfaces, acquired by interpolating measurements from a comprehensive set of weather stations (Harris et al. 2020). 6 of these half-cells were deemed to cover the land surface of all this study's catchment areas (64.25 N, 18.25 E; 63.75 N, 18.25 E; 64.25 N, 18.75 E; 64.25 N, 17.75 E; 64.75, 17.25; 64.25 N, 17.25 E). From these half-cells, a monthly average general for the catchment area was calculated for air temperature and precipitation. These monthly values were calculated into yearly mean (temperature) and sum (precipitation) for parts of the analysis.

Our motivation for using this dataset instead of a single local weather station is an increased confidence in the results because of less reliability and sensitivity on only one specific weather station. The CRU TS temperature values were compared to those of the closest weather station *Fredrika*, which did not differ considerably.

Catchment Areas

Two map layers (SMHI 2016c; SMHI 2016d) were transferred into QGIS. Each specific site's catchment area was estimated from the catchment model in SMHI (2016e).

There is some uncertainty about the extent of the catchment areas (and lake areas) since we did not have access to the exact location where the sampling occurred. The lake areas and catchment areas were modelled in consultation with Statkraft and are considered reasonable and probable.

Runoff per Catchment Area

Runoff in mm/year was calculated in R by using CRU TS annual air temperature (*T*) average and annual precipitation (*P*) sum in the water balance equation. This method is derived from Fischer et al. (2020):

$$P = ET_a + R + \Delta S$$

where ET_a is actual annual evapotranspiration, *R* is annual runoff and *ΔS* is the change in storage. If assuming that *ΔS* is zero over each year, then *R* is calculated with:

$$R = P - ET_a$$

ET_a was determined with Turc's equation (Fischer et al. 2020):

$$ET_a = \frac{P}{\sqrt{0.9 + \frac{P^2}{ET_p}}}$$

where ET_p, the potential evapotranspiration, was obtained from Langbein's equation (Fischer et al. 2020):

$$ET_p = 325 + 21T + 0.9T^2$$

Annual *R* in mm was recalculated into m and used to derive runoff per catchment area (Q_A) in km^3/year with:

$$Q_A = R \times A$$

where *A* is the catchment area in m^2.

Amount of Forest Harvest per Catchment Area

The Swedish Forest Agency produced a shapefile in SWEREF-99 TM which was transferred into QGIS. This contains the specific areas of Swedish forest harvest estimated from satellite images available at the Swedish Forest Agency (2024), described in Swedish Forest Agency (2022).

To get the amount of forest harvest per catchment area an intersect layer was created with QGIS, where the forest harvest area was intersected by the catchment area. The resulting intersect layer contained all forest harvest between 2000 and 2020 specifically for each catchment area. A sum of all areas of forest harvest within the catchment area where then calculated.

The Water Level in the Reservoir

The water level in m per day was gathered from Statkraft. The water level of the reservoir was measured at the east corner of the impoundment. This measurement is deemed representative of the reservoir since there is no sloping of the water level

caused by the water release from the hydropower. There is however some speculation regarding that high winds might create a temporary slope.[2]

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[2] Statkraft, the hydropower plant operators. Personal communication. 2024-05-22.

Appendix B – Data Exclusion

This appendix contains different aspects of the data exclusion part of the method. The different aspects are divided into bullet points.

- o Short motivations for why some of the monitoring data was excluded:
    - Pike samples were removed because of the low amount of raw data.
    - The trend for 25 cm perch does not follow the same trend as 10 cm perch. A motivation for this might be a different response or a different degree of response to ecosystem perturbation. For example, because of biodilution. We will not be able to go into the underlying reasons for this in this study. Because of this, we will not go into 25 cm perch in the result.
    - The excluded sites (Gigån, Outlet Channel, Gäddsjödammen, Kroknorforsen and Björna kvarn) were not deemed to have the same relevance to the objectives as the sites which were used for the analyses.
    - Environmental variables such as Mg, and Ca were not included in the PCA calculation, since preparatory analyses had depicted that there was no existing trend with Hg, and because of the scope of the PCA.
- o The analyses are conducted on yearly averages, for some of the water concentration measurements only one sampling occurred for the whole year (1992 MeHg$_{aq}$ for SKM, STT, VIS; 1994-1995 MeHg and THg$_{aq}$ in LAV)
- o Since this would skew the representation these were removed. Besides water Hg concentrations one sample of total phosphorus and one sample of alkalinity were removed, as they were considered notetaking errors.
- o Ammar et al. (2024) used the method of 5×Median Absolute Deviation (MAD) for outlier removal for time series analysis. With this method none of our values are outliers. Leys et al. (2013) recommend 2.5MAD to be a preferable method instead of the generally used mean ± 3×standard deviation. In absolute normal distribution 3MAD = 3×st.d, but datasets are generally not normally distributed, hence 3MAD gives a more robust result than 3×st.d. 3MAD results in a more conservative removal of outliers, than 2.5MAD, which is why Leys recommends it (Leys et al. 2013). (See the thesis reference list for these two references).

o Several outliers were removed from the Hg datasets. The 3×MAD outlier test resulted in the exclusion of 14 LAV, 2 VIS, 2 LAN, 2 SKM, 1 OVR and 1 STT T_{max} MeHg$_{aq}$ values, 9 LAV T_{min} MeHg$_{aq}$ values, 2 STT, 1 SKM and 1 LAN T_{max} THg$_{aq}$ values as well as 1 VIS and 1 LAV T_{min} THg$_{aq}$ values. 2 MeHg$_{aq}$ values in STT which are not included in the outlier test were also removed, considered to be notetaking errors. How the datasets before and after these outlier removals occurred are portrayed in Fig. B1 and B2.

With potential outliers included

SKM

STT

LAV

VIS
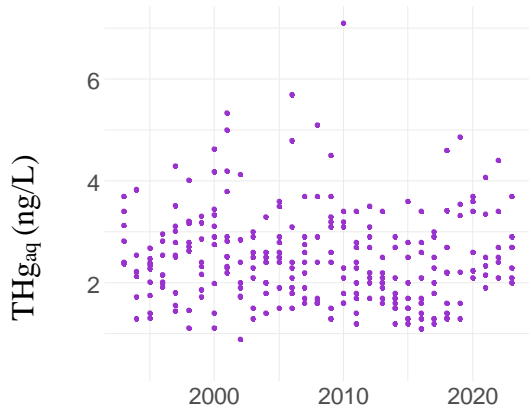
With potential outliers excluded

SKM

STT

LAV
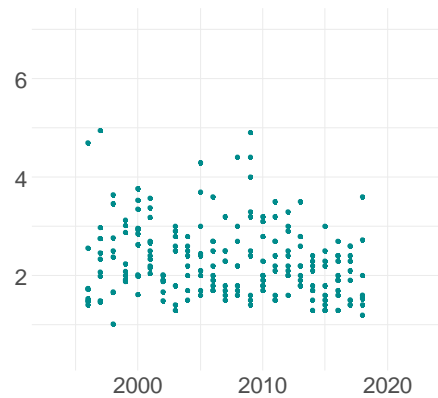
VIS

Figure B1. Difference between THg$_{aq}$ raw data (left) and THg$_{aq}$ after outlier removal by the 3MAD method and assumed notetaking errors (right).
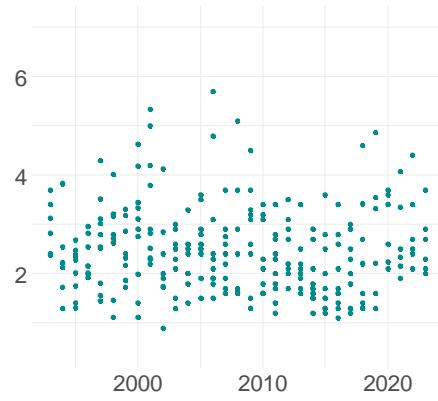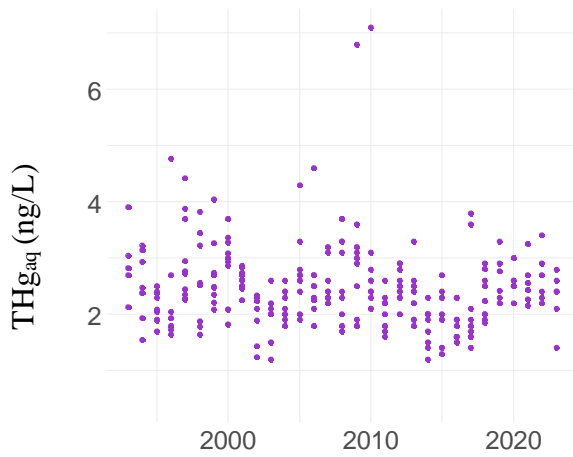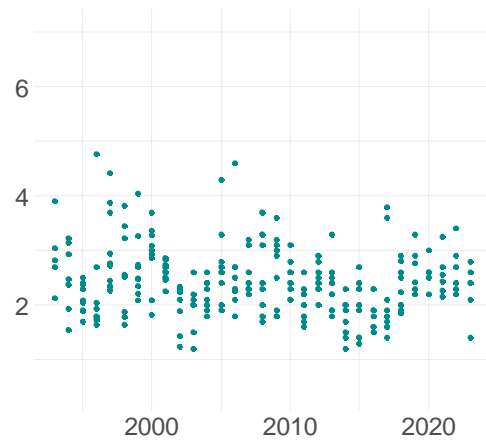
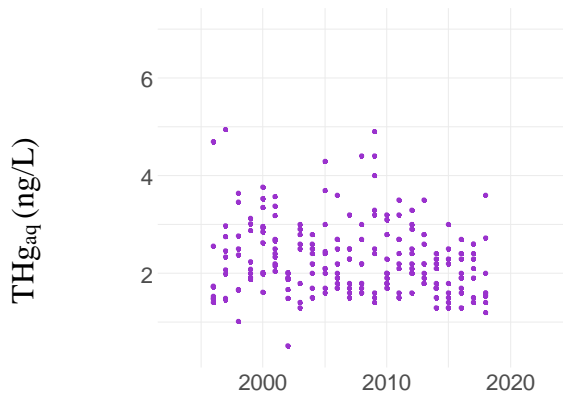**With potential outliers included**

With potential outliers excluded

SKM
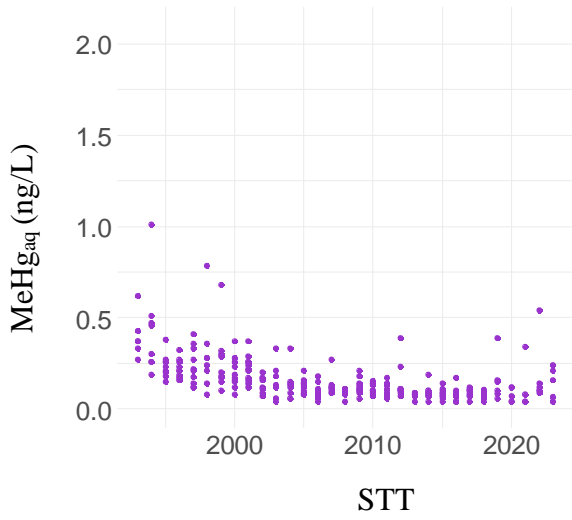
SKM
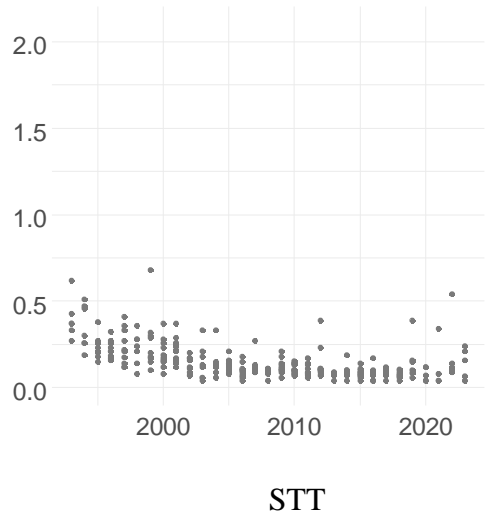
STT

STT
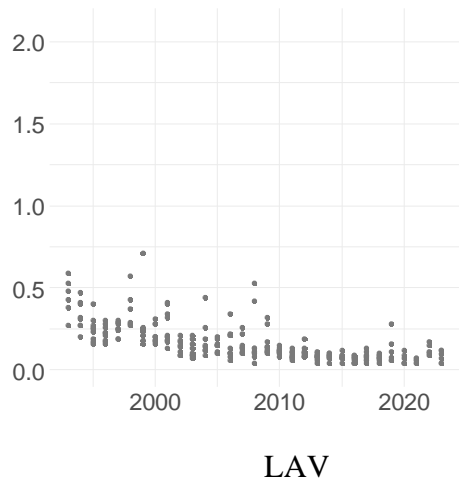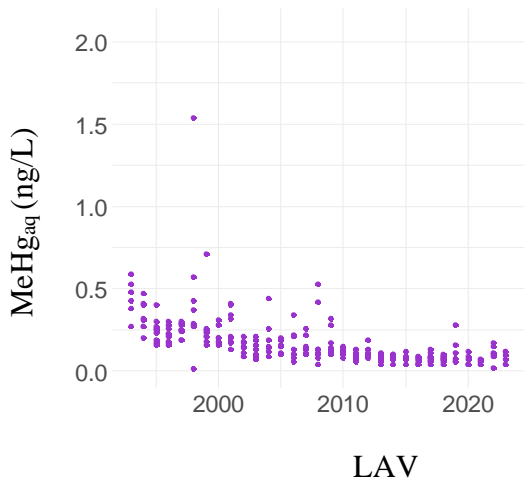
LAV

LAV
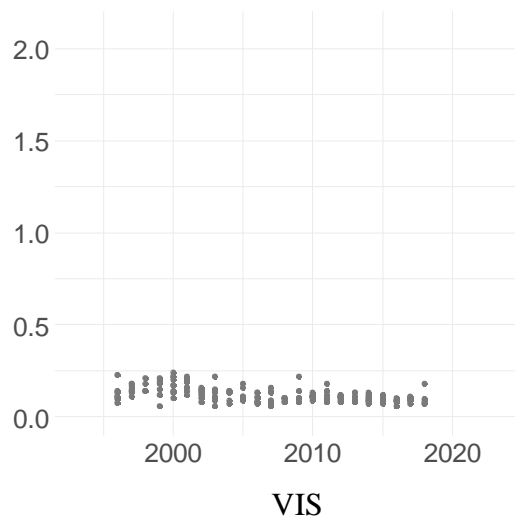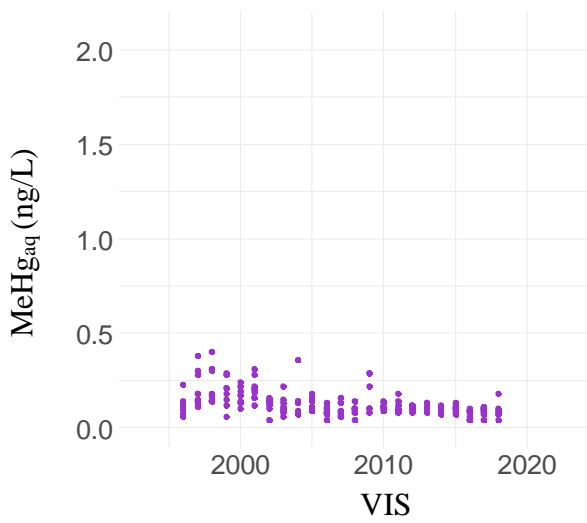
VIS

VIS
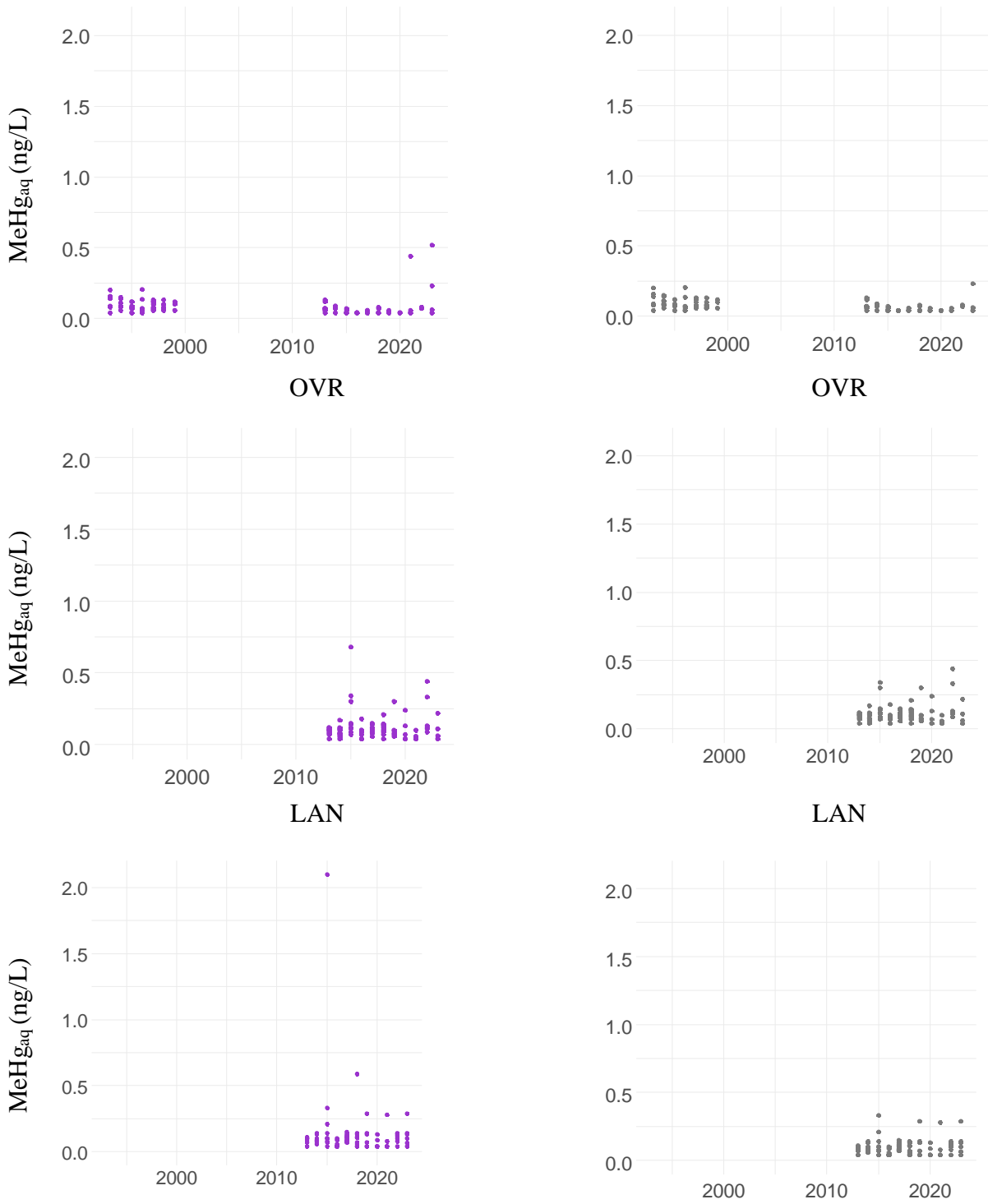
Figure B2. Difference between MeHg$_{aq}$ raw data (left) and MeHg$_{aq}$ after outlier removal by the 3MAD method and assumed notetaking errors (right).

Appendix C – Sampling Periods

Tables C1 and C2 present the periods where sampling occurred within the monitoring programme for each of the monitoring programmes sites and variables.

Table C1. Periods of sampling in various fish within the monitoring programme. The variables sampled are THg and Selenium. The table is divided into sampling site and sampling variable. Modified from Jutterström et al. (2023). Gray sites and variables are not included in this study.

Sampling sites/ Sampling variable	THg in 10 cm perch	THg in 25 cm perch	THg in 1 kg pike	Selenium in 10 cm perch	Selenium in 25 cm perch	Selenium in 1 kg pike
SKM	1989-today	1990-2018	1990-today	1994-today	2007-2018	1999-today
OVR	2013-today	2013-2018	2013-today	2013-today	2013-2018	2013-today
LAN	1991-today	2010-2018	1989-today	2000-today	2010-2018	2004-today
STT	1991-today	1991-2018	1987-today	1995, 2000-today	2007-2018	1999-today
VIS	1991-today	2011-2018	1992-today	1995-today	2011-2018	1999-today
LAV	1994-2018	1994-2018	1993-2018	1994-2018	2010-2018	2006-2018
Gigån	None	None	None	None	None	None
Gäddsjödammen	2000-2012	2000-2004	None	None	None	None
Outlet Channel	1999-2004	None	None	2000-2002	None	None

Table C2. Periods of sampling in surface water within the monitoring programme. The variables sampled are MeHg, THg, selenium, calcium, magnesium, and support parameters (pH, colour, alkalinity, conductivity, and TP). For colour, the sampling period has a pause from 1998 until 2004. Modified from Jutterström et al. (2023). Gray sites and variables are not included in this study.

Sampling sites/ Sampling variable	MeHg in surface water	THg in surface water	Selenium in surface water	Support Parameters in surface water	Calcium and Magnesium in surface water
SKM	1992-today	1993-today	1994-1996	1984-today	2005-2012
OVR	2013-today	2013-today	None	2013-today	None
LAN	2013-today	2013-today	None	2013-today	None
STT	1992-today	1993-today	1994-1996	1985-today	2005-2012
VIS	1992–1999, 2013-today	1993–1999, 2013-today	1994-1996	2013-today	None
LAV	1994–2018	1994–2018	1994-2001	1984-2018	2005-2012
Gigån	2013–2018	2013–2018	None	1984-2018	2005-2012
Gäddsjödammen	2000–2012	2000–2012	None		

Outlet Channel	1999–2012	1999–2012	None		
Kroknosforsen				1987-2012	2005-2012
Björna station				1984-2012	2005-2012

Appendix D – Periods with Available Raw Data

Table D1 visualise the raw data, alternatively the rawest available data from the monitoring programmes used for analysis by this study.

D1. Periods with available data used by the study. If not other is indicated the period had accessible raw data. When "(average)" is below the period it indicates that the available data of that period consisted of yearly averages.

	SKM	STT	VIS	LAV	OVR	LAN
THg_{fish} in perch about 10 cm	1989 – 1996, 2015 – 2016, 2018 – 2019, 2021 - 2023	1991 – 1996, 2015 – 2016, 2018 – 2019, 2021 - 2023	1991 – 1996, 2015 – 2016, 2018 – 2019, 2021 - 2023	1994, 1996, 2015 – 2016, 2018	2015 – 2016, 2018 – 2019, 2021 - 2023	2015 – 2016, 2018 – 2019, 2021 - 2023
THg_{aq} and $MeHg_{aq}$	2013 – 2023	2013 – 2023	1993 – 1999, 2013 - 2023	1996 - 2018	2013 – 2023	2013 – 2023
pH	1991 – 2004 (average), 2005 – 2023	1991 – 2004 (average), 2005 – 2023	1991 – 2004 (average), 2005 – 2023	1991 – 2004 (average), 2005 – 2018	2013 – 2023	2013 – 2023
Colour	1991 – 1997 (average), 2005 - 2023	1991 – 1997 (average), 2005 - 2023	2013 – 2023	1991 – 1997 (average), 2005 - 2018	2013 – 2023	2013 – 2023
Alkalinity	1991 – 2004 (average), 2005 - 2023	1991 – 2004 (average), 2005 - 2023	2013 – 2023	1991 – 2004 (average), 2005 - 2018	2013 – 2023	2013 – 2023
Conductivity	1991 – 1995, (average) 1998 – 2004 (average), 2005-2023	1991 – 1995, (average) 1998 – 2004 (average), 2005-2023	2013 – 2023	1991 – 1995, (average) 1998 – 2004 (average), 2005-2018	2013 – 2023	2013 – 2023
Total Phosphorus	1991 – 2004 (average), 2005 – 2023	1991 – 2004 (average), 2005 – 2023	2013 – 2023	1991 – 2004 (average), 2005 – 2023	2013 – 2023	2013 – 2023

Appendix E – Laboratory Methods

Tables E1 and E2 display which methods were used to analyse monitoring parameters by the IVL laboratory (Table E1) and the MoRe laboratory (Table E2).

Table E1. Hg parameters from the monitoring programme were analysed by the IVL laboratory with these methods in 2020. With measurement of uncertainty for the analysis methods and limit of quantification (LOQ).

Parameter	Analysis method	Measurement uncertainty	LOQ
THg_{aq}	IVL A9 – manually, based upon EPA 1631 version E	<0.25: 14% >0.25: 8%	0.05 ng/L
	Alternatively: IVL A9 – with a sample changer, based upon EPA 1631 version E	8%	0.2 ng/L
$MeHg_{aq}$	IVL A10 Based upon EPA-1630	12%	0.06 ng/L
THg_{fish}	Microwave digested sample SS-EN15763 analysed with IVL A44, based upon EPA 1631 version E	15%	0.2 ng/g

Table E2. Methods and units for analysis by MoRe Research of surface water samples of the support parameters in 2020.

Parameter	Unit	Method
pH	None	SS-EN ISO 10523:2012
Absorbance at 420 nm	Abs_{420}/5 cm	SS-EN ISO 7887:2012, method C mod.
Alkalinity	mmol/L HCO_3	SS-EN 9963-2, edition 1
Conductivity	mS/m	SS-EN 27888, edition 1
Total Phosphorus	μg/L	SS-EN ISO 6878:2005, part 7

Appendix F – Additional Results

Appendix F contains additional results not included in the main thesis text:

- A table with the spatial trend results from ANOVA (Table F1). Table F1 contain more detailed information than Fig. 8.
- The complete Pearson correlation matrices (Fig. F1). Highlights of relevant parts of the matrices are available in Fig. 9.
- Fig. F2 contain the yearly average colour measurements from the monitoring programme between 1993 and 2023.
- Fig. F3

*Table F1. Spatial trend results from ANOVA. Where the means in sites SKM, STT, LAV, OVR, LAN, and VIS have been compared to each other. The means in question are yearly geometric means of THg_{aq}, $MeHg_{aq}$ and THg_{fish} during periods of interest. These periods are the beginning of the monitoring period (1993-1999/19991-1996), the first CP 2006 (2006-2010), the third CP 2016 (2016-2018), and the "present" day (2019-2023). Which sites are examined vary, since the variability of available data. The years studied vary between THg_{fish} and the two other variables for the same reason. Black borderlines indicate a change of period. A rectangular border with coloured lines specifies the statistic test according to one-way ANOVA followed by Tukey multiple comparisons tests. An ANOVA p-value < 0.05, indicated with *, signifies that not all means in the calculation are equal. The grouping originates from Tukey's multiple comparisons test. If sites do not share the same letter (A, B, C), they are significantly different. A is the group with the highest means. Column n portrays the number of geometric mean samples per site.*

Response variable	Periods in years	Sites	n	Mean (st.d.)	Grouping*	p-value	R²
THg_{aq} (ng/L)	1993-1999	SKM	7	2.4 (0.28)	A	0*	81
		STT	7	2.5 (0.31)	A		
		LAV	4	2.2 (0.22)	A		
		VIS	7	1.2 (0.22)	B		
MeHg_{aq} (ng/L)	1993-1999	SKM	7	0.26 (0.066)	A	0*	77
		STT	7	0.29 (0.072)	A		
		LAV	4	0.14 (0.016)	B		
		VIS	7	0.085 (0.012)	B		
THg_{fish} (mg/kg)	1991-1996	SKM	6	0.39 (0.18)	A	0.003*	53

Parameter	Years	Site	n	Mean (SE)	Group	p	%
		STT	6	0.38 (0.16)	A		
		VIS	6	0.098 (0.047)	B		
THg_{aq} (ng/L)	2006-2010	SKM	5	2.3 (0.15)	A	0.061	37
		STT	5	2.5 (0.062)	A		
		LAV	5	2.3 (0.23)	A		
$MeHg_{aq}$ (ng/L)	2006-2010	SKM	5	0.11 (0.014)	B	0.002*	66
		STT	5	0.13 (0.013)	A		
		LAV	5	0.099 (0.0081)	B		
THg_{aq} (ng/L)	2016-2018	SKM	3	2.0 (0.13)	A	0.004*	73
		STT	3	2.0 (0.24)	A		
		LAV	3	1.9 (0.089)	A		
		OVR	3	2.0 (0.27)	A		
		LAN	3	1.9 (0.16)	A		
		VIS	3	1.3 (0.18)	B		
$MeHg_{aq}$ (ng/L)	2016-2018	SKM	3	0.074 (0.010)	A B	0.011*	67
		STT	3	0.070 (0.0080)	A B		
		LAV	3	0.090 (0.0051)	A		
		OVR	3	0.094 (0.018)	A		
		LAN	3	0.076 (0.023)	A B		
		VIS	3	0.045 (0.0029)	B B		
THg_{fish} (mg/kg)	2015-2016, 2018	SKM	3	0.11 (0.011)	A B	0*	84
		STT	3	0.078 (0.0086)	B C		
		LAV	3	0.082 (0.015)	B C		
		OVR	3	0.13 (0.0066)	A		
		LAN	3	0.094 (0.016)	A B		
		VIS	3	0.055 (0.012)	C		
THg_{aq} (ng/L)	2019-2023	SKM	5	2.68 (0.15)	A	0*	85
		STT	5	2.5 (0.16)	A		
		OVR	5	2.7 (0.24)	A		
		LAN	5	2.5 (0.26)	A		
		VIS	5	1.4 (0.26)	B		

Response variable	Period	Site	n	Mean (SE)	Group	p-value	Count
MeHg_aq (ng/L)	2019-2023	SKM	5	0.10 (0.033)	A	0.14	28
		STT	5	0.084 (0.028)	A		
		OVR	5	0.096 (0.043)	A		
		LAN	5	0.084 (0.021)	A		
		VIS	5	0.054 (0.014)	A		
THg_fish (mg/kg)	2019-2023	SKM	4	0.11 (0.02)	A	0.001*	69
		STT	4	0.093 (0.019)	A		
		OVR	4	0.13 (0.030)	A	B	
		LAN	4	0.11 (0.017)	A		
		VIS	4	0.055 (0.011)	B		

*Each group is significantly different from the other groups. The sites in group A have significantly the same mean, but they are significantly different from the means in groups B and C. The concentrations in group A are always significantly higher than those in B and C. The one with two letters means this group of data cannot be classified as different to either letter. In most cases, this is because of either a large variability of data or two fewer replicates. Observe that there is one "grouping" per response variable and period.

Pearson correlation matrix for SKM

Skinnmuddselet

	MeHg	THg	pH	Color	Alkalinity	Conductivity	TP	Temp	Prec.	Flow	Deforest
MeHg	1.00	0.46	-0.22	0.57	-0.34	-0.43	0.09	-0.29	0.21	0.32	0.18
THg	0.46	1.00	-0.17	0.73	-0.40	-0.39	0.27	-0.04	0.22	0.22	-0.10
pH	-0.22	-0.17	1.00	-0.46	0.68	0.48	0.58	0.20	-0.23	-0.31	-0.07
Color	0.57	0.73	-0.46	1.00	-0.67	-0.53	0.06	-0.25	0.17	0.25	-0.12
Alkalinity	-0.34	-0.40	0.68	-0.67	1.00	0.89	-0.08	0.02	-0.32	-0.31	-0.45
Conductivity	-0.43	-0.39	0.48	-0.53	0.89	1.00	-0.19	0.12	-0.34	-0.37	-0.49
TP	0.09	0.27	0.58	0.06	-0.08	-0.19	1.00	0.08	-0.11	-0.14	0.38
Temp	-0.29	-0.04	0.20	-0.25	0.02	0.12	0.08	1.00	0.05	-0.36	-0.05
Prec.	0.21	0.22	-0.23	0.17	-0.32	-0.34	-0.11	0.05	1.00	0.91	0.10
Flow	0.32	0.22	-0.31	0.25	-0.31	-0.37	-0.14	-0.36	0.91	1.00	0.12
Deforest	0.18	-0.10	-0.07	-0.12	-0.45	-0.49	0.38	-0.05	0.10	0.12	1.00

Pearson correlation matrix for STT

St.Tällvattnet

	MeHg	THg	pH	Color	Alkalinity	Conductivity	TP	Temp	Prec.	Flow	Deforest
MeHg	1.00	0.60	-0.69	0.25	-0.14	0.17	-0.10	-0.16	0.17	0.23	0.10
THg	0.60	1.00	-0.72	0.77	-0.70	-0.28	0.15	-0.24	0.18	0.26	-0.07
pH	-0.69	-0.72	1.00	-0.62	0.58	0.26	0.13	0.29	-0.21	-0.32	-0.33
Color	0.25	0.77	-0.62	1.00	-0.77	-0.55	0.22	-0.41	0.07	0.23	0.08
Alkalinity	-0.14	-0.70	0.58	-0.77	1.00	0.82	-0.23	0.12	-0.12	-0.16	-0.09
Conductivity	0.17	-0.28	0.26	-0.55	0.82	1.00	-0.29	0.14	-0.10	-0.16	-0.31
TP	-0.10	0.15	0.13	0.22	-0.23	-0.29	1.00	0.06	0.03	-0.00	0.01
Temp	-0.16	-0.24	0.29	-0.41	0.12	0.14	0.06	1.00	0.05	-0.36	0.02
Prec.	0.17	0.18	-0.21	0.07	-0.12	-0.10	0.03	0.05	1.00	0.91	0.52
Flow	0.23	0.26	-0.32	0.23	-0.16	-0.16	-0.00	-0.36	0.91	1.00	0.48
Deforest	0.10	-0.07	-0.33	0.08	-0.09	-0.31	0.01	0.02	0.52	0.48	1.00

Pearson correlation matrix for LAV

Lavsjön

	MeHg	THg	pH	Color	Alkalinity	Conductivity	TP	Temp	Prec.	Flow	Deforest
MeHg	1.00	0.72	-0.56	0.70	-0.43	-0.00	-0.06	-0.47	0.22	0.38	-0.54
THg	0.72	1.00	-0.81	0.81	-0.62	-0.16	-0.27	-0.29	0.41	0.49	-0.43
pH	-0.56	-0.81	1.00	-0.85	0.94	0.64	0.28	0.39	-0.58	-0.68	0.37
Color	0.70	0.81	-0.85	1.00	-0.81	-0.42	-0.10	-0.46	0.63	0.75	-0.42
Alkalinity	-0.43	-0.62	0.94	-0.81	1.00	0.79	0.29	0.39	-0.53	-0.54	0.32
Conductivity	-0.00	-0.16	0.64	-0.42	0.79	1.00	0.05	0.43	-0.38	-0.52	-0.13
TP	-0.06	-0.27	0.28	-0.10	0.29	0.05	1.00	-0.42	-0.19	-0.02	0.18
Temp	-0.47	-0.29	0.39	-0.46	0.39	0.43	-0.42	1.00	-0.00	-0.38	0.24
Prec.	0.22	0.41	-0.58	0.63	-0.53	-0.38	-0.19	-0.00	1.00	0.93	-0.15
Flow	0.38	0.49	-0.68	0.75	-0.54	-0.52	-0.02	-0.38	0.93	1.00	-0.23
Deforest	-0.54	-0.43	0.37	-0.42	0.32	-0.13	0.18	0.24	-0.15	-0.23	1.00

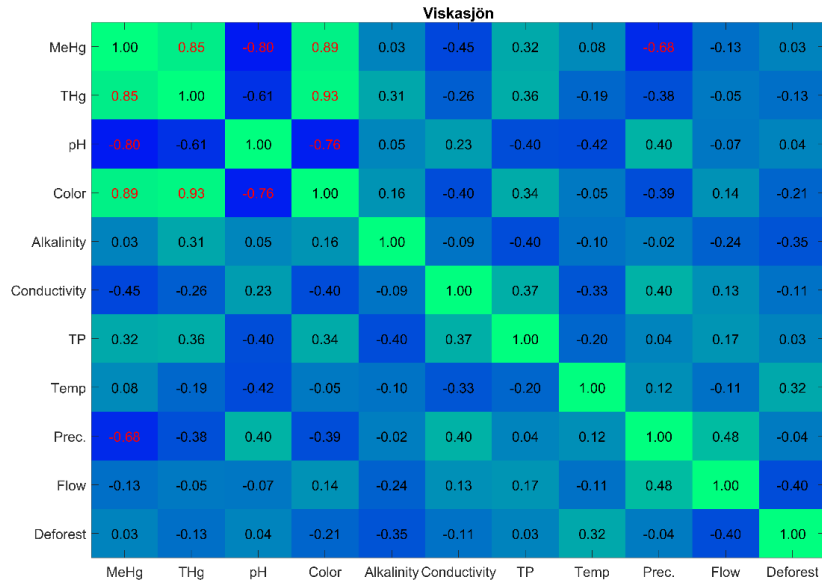Pearson correlation matrix for VIS

Figure F1. Complete Pearson correlation matrix from the PCA results of MeHg$_{aq}$, THg$_{aq}$ and potential environmental drivers presented by Pearson correlation matrices of the sites SKM, STT, LAV and VIS. Values in the correlation matrix are the correlation coefficients. Those with red fonts have significant correlations (p < 0.05).
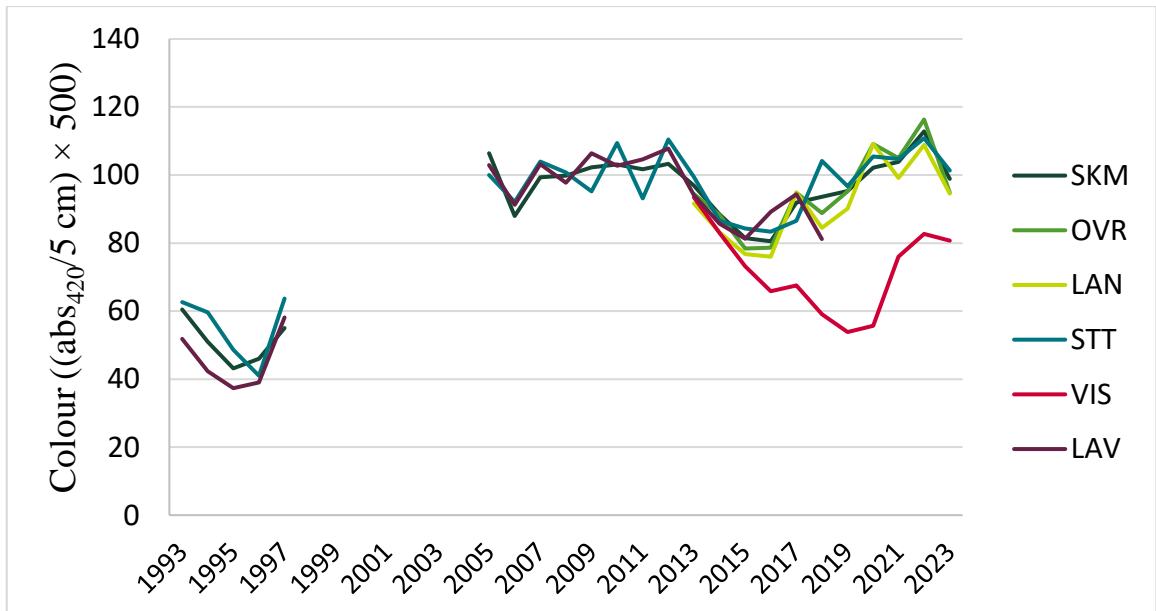

Figure F2. Yearly average of colour measurements between 1993 and 2023 within the monitoring programme. The reference site (VIS) has different trends and concentrations than the hydropower-affected sites after 2015.
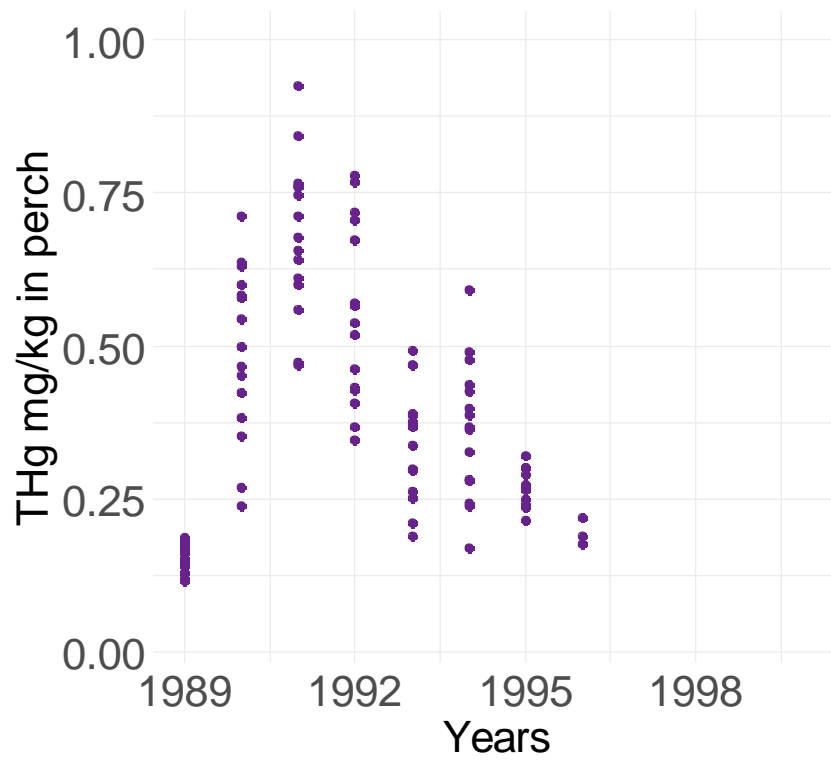
Figure F3. Portray the normalised THg$_{fish}$ concentrations for the year of hydropower impoundment (1989) and the subsequent incline and decline in concentration in the reservoir site (SKM).

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