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# ICP Waters report 132/2017
## Spatial and temporal trends of mercury in freshwater fish in Fennoscandia (1965-2015)


International Cooperative Programme on Assessment and Monitoring Effects of Air Pollution on Rivers and Lakes

Convention on Long-Range Transboundary Air Pollution

Norwegian Institute for Water Research

# REPORT


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Summary

Mercury (Hg) emissions to the atmosphere cause elevated Hg levels in fish, even in many remote regions of the world. Here we present an extensive database of more than 50 000 measurements of Hg in fish, including 2 775 individual water bodies in Fennoscandia (Norway, Sweden, Finland, Russian part of Kola Peninsula) sampled between 1965 and 2015. The data have been analysed for spatial patterns and temporal trends, on raw and weight-adjusted data. The database presents a useful reference for assessment of impacts of environmental policy on Hg in freshwater fish (i.e. *Convention on Long-Range Transboundary Air Pollution* and *The Minamata Convention on Mercury*).



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# CONVENTION OF LONG-RANGE TRANSBOUNDARY AIR POLLUTION

# INTERNATIONAL COOPERATIVE PROGRAMME ON ASSESSMENT AND MONITORING EFFECTS OF AIR POLLUTION ON RIVERS AND LAKES

## SPATIAL AND TEMPORAL TRENDS OF MERCURY IN FRESHWATER FISH IN FENNOSCANDIA (1965-2015)

Prepared at the ICP Waters Programme Centre
Norwegian Institute for Water Research
Oslo, September 2017

# Preface

*The International Cooperative Programme on Assessment and Monitoring Effects of Air Pollution on Rivers and Lakes (ICP Waters)* was established under the Executive Body of the *United Nations Economic Commission for Europe* (UNECE) *Convention on Long-Range Transboundary Air Pollution* (CLRTAP) at its third session in Helsinki in July 1985. UNECE is a catalyst in the international work aiming at reducing transboundary air pollution. Norway provides facilities for the ICP Waters Programme Centre at the Norwegian Institute for Water Research (NIVA). The Norwegian Environment Agency provides financial support and a representative who is Chair of ICP Waters. ICP Waters receives additional financial support from the UNECE Trust Fund of the CLTRAP. In the long-term strategy for the Convention it is stated that environmental effects of acidifying components, and its potential interaction with climate change and biodiversity, continue to be among the significant remaining problems with regard to air pollution effects on the environment. These can be addressed with the multi-pollutant/multi-effects approach of the Gothenburg Protocol.

The main aim of the ICP Waters is to assess, on a regional basis, the degree and geographical extent of the impact of atmospheric pollution on surface waters. The pollutant mercury (Hg) is addressed in the Aarhus Protocol under the CLRTAP. Documentation of the environmental effects of Hg is important for the CLRTAP and the UN Environment *Minamata Convention on Mercury*, which entered into force on August 16th, 2017. Hg is of particular interest in surface waters, as alarmingly high levels of the element have been reported in fish even in remote lakes. Currently, an international monitoring network for assessing distribution and effects of long-range transported Hg in freshwater ecosystems is lacking.

This report has been prepared in cooperation with the *International Cooperative Programme for Integrated Monitoring of Air Pollution Effects on Ecosystems* (ICP IM). The report presents a comprehensive database of fish Hg concentrations in freshwater ecosystems in Fennoscandia (Norway, Sweden, Finland and the Kola Peninsula in Russia) for the period 1965 to 2015, gathered from national monitoring programmes and research projects. Its main objective is to investigate temporal trends and spatial patterns of Hg in fish in the context of environmental change, especially atmospheric pollution.

We would like to thank all who have contributed with data for the database and comments on the report and to acknowledge the Norwegian project *Climer* (243644/E50), funded by the Research Council of Norway (RCN), for financial support.

*Heleen de Wit*

ICP Waters Programme Centre
Oslo, September 2017

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# Executive summary

Fish in freshwater ecosystems constitute an important exposure pathway of Hg to humans and wildlife, and are thus considered as critical receptors of long-range transboundary air pollution of Hg. Fish Hg levels, even in remote regions, commonly exceed environmental quality standards (EQS) set by the World Health Organisation (WHO) and Food and Agricultural Organisation (FAO) of the UN for protection of human health (0.3 – 1.0 parts per million, ppm, wet weight, ww). The EQS set by the European Union Water Framework Directive (WFD) to protect wildlife (0.02 ppm ww) is exceeded in the vast majority of the water bodies across Fennoscandia (i.e. Norway, Sweden, Finland, and Russian Kola Peninsula) that were investigated in this study.

In this report, our aim was to assess effects of atmospherically transported air pollutants on fish Hg levels. We have assembled fish Hg data and associated explanatory variables, including fish metrics, in a Fennoscandian database for the period between 1965 and 2015. The initiative for the project was taken by ICP Waters, and the work was done in collaboration with ICP Integrated Monitoring, both bodies under the LRTAP Convention.

A total of 54 560 Hg levels for individual fish were retrieved from Swedish (n = 34 691), Finnish (n = 14 878), Norwegian (n = 4 792), and Russian (n = 199) databases. Monitoring of Hg levels in freshwater fish in Fennoscandia has been undertaken since the 1960s, resulting in fish Hg data from lakes and rivers across a wide range of climatic, depositional, and land cover gradients typical for boreal, subarctic, and Arctic ecosystems. Some of the lakes and rivers have been impacted by local industrial emissions of Hg directly to the water, in contrast to those that were only impacted by air pollution sources. Data on the extent of such local industry impacts versus possible geogenic sources are not easily available, which presented some challenges for interpretation of the results in relation to air pollution.

The Fennoscandian fish Hg database includes a variety of species (n: pike (*Esox lucius*) > perch (*Perca fluviatilis*) >> brown trout (*Salmo trutta*), Arctic charr (*Salvelinus alpinus*), roach (*Rutilus rutilus*)) with a variation in fish species composition within and between lakes. Fish species, size and trophic level is used for the analysis, and we highlight the use of additional data on water chemistry, climate, and deposition for future testing of hypotheses on the potential effects of environmental change on Hg in fish. An example of this is effects from dissolved organic matter (DOM). High Hg levels in fish are usually associated with lakes with high concentrations of DOM, and surface waters are currently experiencing long-term increases in DOM concentrations in many boreal and subarctic Nordic ecosystems, potentially impacting Hg levels in fish. However, to understand variation in fish Hg levels, there is a need to consider the wide range of factors that can affect Hg cycling and bioaccumulation (e.g., catchment characteristics, water quality, trophic structure, and climate) in addition to atmospheric deposition.

The spatial patterns of Hg levels in Fennoscandian freshwater fish populations reveal concentrations higher than the WFD EQS in all but three examined water bodies. Median observed Hg concentration per fish species ranged from 0.16 ppm (brown trout) to 0.62 ppm (pike). Of the 54 560 fish samples included in the database for analyses, only 82 specimens showed concentrations below 0.02 ppm. According to this criterion, good chemical condition is not met for the vast majority of Fennoscandian lakes with respect to Hg concentrations in fish. We conclude that the WFD EQS for Hg in biota has limited relevance for assessing the risks of fish Hg exposure in Fennoscandia because it does not differentiate between lakes with higher and lower risk.

Of the observed and uncorrected, directly measured Hg levels in the database, 46, 36 and 20 % of the collected fish had concentrations above the 0.5 ppm FAO/WHO limit in Sweden, Finland, and Norway, respectively. The method used for size-adjustment and correction of the fish data, needed due to the strong co-variation between Hg concentration and fish age, influences the conclusions drawn from the spatial patterns and comparisons with relevant EQS. Size-adjustment of the reported data was done in two ways: *i*) to adjust for positive relationship between Hg concentrations and fish weight, and *ii*) by conversion to a standard 1-kg pike (following the ICP Modelling and Mapping, ICP M&M, manual for calculation of critical loads). The conversion to a standard 1-kg pike is done to correct for Hg accumulation differences between fish species, in order to allow for an evaluation in one standardised unit across regions. Generally, the conversion to a standard 1-kg pike generated the highest median fish population Hg concentrations.

We aimed to investigate the spatial patterns and temporal trends of Hg concentrations in fish in relation to long-range atmospherically transported Hg pollution. Thus, influences from local industry emission sources would confound relations between air pollution and Hg in fish. Therefore, we separated lakes that are influenced by local Hg pollution sources from lakes that are only impacted by atmospheric Hg deposition. Hg is released to the atmosphere through natural processes, and has also in recent centuries been released due to anthropogenic activities, leading to long-term accumulation of Hg in catchment soils. All lakes in the database were characterised as one of the following: *1*) lakes subject mainly to sources of Hg from atmospheric deposition with no known industrial local point sources of Hg; or *2*) lakes with known local industry point sources of Hg. Data from the two groups of lakes indicated that lakes that are predominantly affected by atmospherically deposited Hg (0.25 ± 0.27 ppm, n = 703) had lower mean observed fish Hg concentrations than lakes affected by local pollution sources (0.55 ± 0.38 ppm, n = 167). These differences reflect the significance of direct historical industrial releases of Hg to surface waters on present day Hg concentrations in fish, as well as the importance of separating these two groups when considering how policy aimed at reducing air pollution of Hg has its intended effect, i.e. reduced contamination of freshwater fish.

Fish Hg concentrations within lakes that were classified as being subject mainly to sources of Hg from atmospheric deposition, show decreasing Hg concentrations from south-to-north. This pattern also follows the Hg deposition gradient with decreasing Hg deposition from the south towards the north, evident through measurements of Hg in for example top layers of lake sediments. Thus, this suggests that lower atmospheric Hg deposition indeed leads to lower accumulation of Hg in fish. However, the south-to-north gradient of Hg deposition is confounded by similar gradients in DOM and temperature, both strong regulators of Hg in fish. Thus, the spatial relationship between atmospheric Hg deposition and Hg in fish cannot simply be interpreted as evidence for a direct link between atmospheric Hg and fish Hg. A better understanding of controls of, for example, DOM and temperature on Hg fish accumulation is needed in order to disentangle spatial drivers of fish Hg accumulation.

Upon more detailed analysis, we found that the temporal Hg concentration trends were not consistent across fish species, fish normalisation methods, or country/region of origin. As an example, in the selection of lakes that were identified as being impacted only by atmospheric deposition of Hg, observed perch Hg concentrations were declining throughout the study period, while weight-normalised concentrations showed no change. And while weight-adjusted perch Hg concentrations were declining in Finland (1965-2015), they were increasing in Norway (1990-2015). Additionally, it is clear from the temporal trend analyses that, despite high concentrations in the

1970s, inter-annual variation of fish Hg concentrations, even for normalised data, were larger than the long-term changes.

Despite the lack of coherent trends for individual lakes and fish species, the correction of data to a standard 1-kg pike showed a consistent and significantly decreasing trend for both the entire database and the lakes mainly influenced by atmospheric deposition of Hg. This decline is consistent with the reported decline in atmospheric Hg emissions and deposition from The European Monitoring and Evaluation Programme (EMEP). Thus, the simultaneous decline in fish Hg, standardized to 1-kg pike, and deposition of atmospheric Hg, could suggest that reduced emissions lead to lower Hg in fish. However, a better understanding of the impacts on temporal trends from various methods for standardisation of fish Hg concentrations, and also other possible confounding environmental processes, is needed prior to concluding that the two declining trends are causally linked. Based on the entire database, i.e. not separating lakes with local Hg impacts from those mainly impacted by atmospherically deposited Hg, we conclude that, for pike and perch (the most abundant fish in the database) the Hg concentrations in Fennoscandia are presently at their lowest since the first recorded measurements in the 1960s. Most likely, local and national policies aimed at reducing emissions from industry to surface waters are partly responsible for this improvement.

The considerable spatial and temporal variation in fish Hg remains poorly understood. It is likely that atmospheric deposition of Hg, foodweb dynamics, geogenic sources of Hg, legacy Hg, catchment characteristics, climate, reduced sulphur deposition and climate change all interact and control foodweb exposure to, and foodweb accumulation of, Hg. The present database provides an excellent opportunity for further investigation of environmental controls on Hg in fish, especially when the fish records can be combined with more site-specific data on these controls. Additionally, the database and reported findings are a potential useful baseline for future monitoring of Hg in the environment, particularly relevant to document the effectiveness of the global agreement *Minamata Convention on Mercury* which entered into force in August 2017. The results will be communicated to international policy bodies focusing on air pollution and Hg contamination.

# Short summary


- Fish Hg records from 2 775 lakes in Fennoscandia (Norway, Sweden, Finland and the Kola Peninsula in Russia), sampled between 1965 and 2015, were compiled with the aim of evaluating impacts of atmospheric Hg pollution on Hg contamination in fish.
- A considerable number of lakes were found to only be impacted by atmospheric sources of Hg (n = 888), while significant numbers of lakes were also impacted by local, mostly historical, industrial point sources (n = 158). Data on the remaining lakes are lacking. Data on local emission sources are not readily available and pose a challenge to evaluation of data in relation to air pollution.
- Hg in fish is positively correlated with size, and both raw data and weight-adjusted fish Hg concentrations were analysed. Data were also adjusted with regard to species through a conversion to a standard 1-kg pike (following the ICP M&M manual for calculation of critical loads).
- Median lake-specific fish Hg concentrations in the vast majority of the studied Fennoscandian lakes exceed EQS for Hg in biota (0.02 ppm) set by EU WFD.
- Where a significant decline in temporal fish Hg concentration was observed, the trend was much stronger for the entire database than for lakes only impacted by atmospheric sources of Hg. We attribute the contrast in trends primarily to declining impacts of local pollution sources.
- Significant declines in fish Hg concentrations for lakes only impacted by atmospheric sources of Hg with latitude could be a consequence of lower atmospheric deposition towards the north, but the pattern also coincides with a decline in temperature and aquatic concentrations of dissolved organic carbon. Further work is required to disentangle the effects of climate, deposition and catchment properties on fish Hg concentrations.
- The fish Hg database is a valuable source of information for continued monitoring of impacts of Hg in the environment. In particular, the lakes primarily impacted by atmospheric sources of Hg will be relevant for documentation of effects of reduced air pollution on fish Hg (CLTRAP). The entire database has a large potential for evaluation of effectiveness of past and future policy to reduce Hg in the environment, including the global *Minamata Convention on Mercury* which entered into force in August 2017.

# 1 List of contributors/acknowledgements

The lead authors of the report are Hans Fredrik Veiteberg Braaten (ICP Waters, NIVA) and Staffan Åkerblom (ICP IM, Swedish University of Agricultural Sciences, SLU), responsible for gathering and organising the database, analysing the data, and writing the main body of the text. The editorial committee consisted of Martti Rask (Natural Resources Institute Finland, Luke) and Heleen de Wit (NIVA).

The present report and the data therein is created and collected with help from a large group of institutes, universities, researchers, and others, located all over Fennoscandia (Figure 1). All the co-authors of this report have, in addition to donating their time and expertise towards the text and conclusions of the work, contributed to the database with raw data; numbers being either previously published or un-published. Additional data that are published in reports or other scientific work are referenced in the literature list at the end of this report.

National and regional monitoring and surveillance programs and research activities in Fennoscandia financed the data collection presented here. A complete list of people that have contributed to the data collection cannot be generated given the number of individuals that have been involved in for example data gathering (field work, planning and design of monitoring studies), fish analysis (fish processing), chemical analysis (determination of Hg in fish), reporting and presenting of numbers and findings over the last six decades. Such a list would include researchers, assistants, students, professors, engineers, analysts, lab technicians, local fishermen, and many, many more. We gratefully acknowledge their contributions.

Hans Fredrik Veiteberg Braaten and Staffan Åkerblom, September 2017

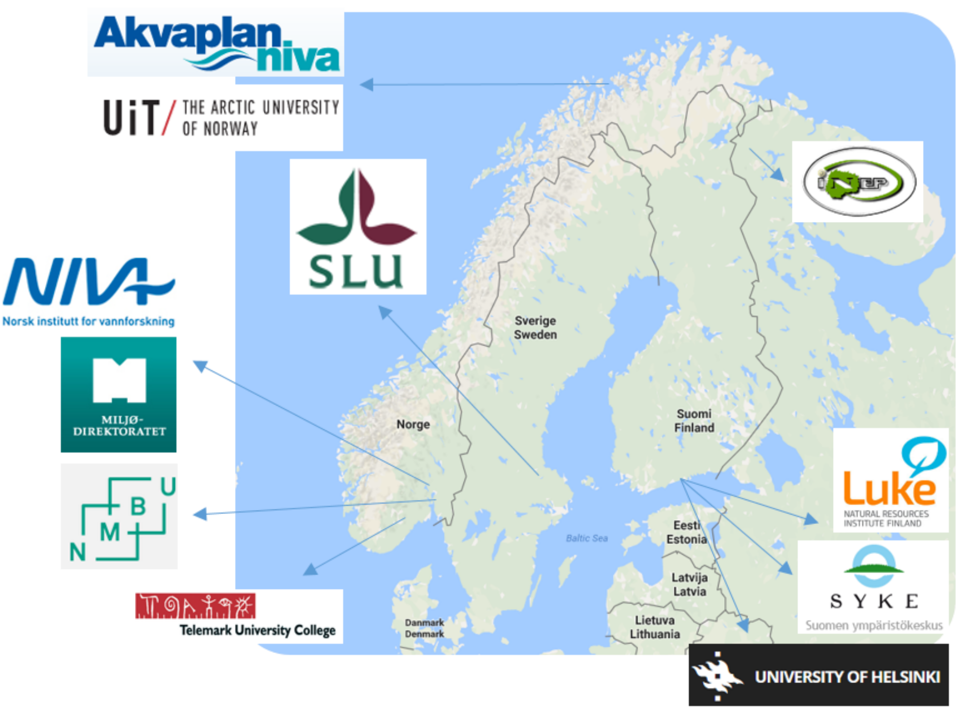


**Figure 1** An overview of Fennoscandian collaboration responsible for the report.

# 2 Introduction

## 2.1 Main report goals and objectives

Mercury (Hg) can undergo long-range atmospheric transport, and nearly two centuries of elevated Hg deposition from anthropogenic activities have led to enhanced stores of Hg in soils, even in remote locations (Fitzgerald et al., 1998). A subsequent effect is elevated Hg concentrations in the aquatic environment, demonstrated by elevated concentrations of Hg in fish in Arctic (e.g. AMAP, 2011; Riget et al., 2011) and boreal regions (e.g. Åkerblom et al., 2014; Gandhi et al., 2014). Hg concentrations in freshwater fish from these areas can be many orders of magnitude higher than the concentrations in the surrounding waters, and levels often exceed national and international dietary advisory limits, which typically fall between 0.3-1.0 ppm (FAO, 1995; UNEP, 2002).

Freshwater fish constitute an important food source for humans and wildlife but are also an important source of Hg, which can pose a threat to human and ecosystem health (Scheulhammer et al., 2007; Zahir et al., 2005). Fish is a key study organism for monitoring of the fate and exposure of Hg. Although the toxic effects of Hg have been known for more than half a century (Kurland et al., 1960), and awareness was raised concerning Hg in fish already in the 1960s (Johnels et al., 1967), a thorough understanding of the complex processes involved in the biogeochemical cycling of Hg in the environment is lacking. Despite the risks posed by Hg to the health of humans and wildlife, it is unclear whether reduced atmospheric emissions of Hg are reflected in Hg concentrations in fish, or if other factors than atmospheric emissions play more important roles.

A comprehensive collection and assessment of available data on Hg concentrations in air, precipitation, sediments and fish from Norway, Sweden, and Finland was presented by Munthe et al. (2007). Since then, studies and reports have documented temporal trends of Hg concentrations in northern ecosystems, including increasing, decreasing and unchanged fish Hg concentrations covering the last five to six decades. These studies cover different geographical regions, ranging from local (e.g. Norwegian counties) to regional (e.g. South-east Norway, Sweden etc.), and span several orders of magnitude when it comes to number of fish collected (from < 50 to thousands).

In this report, our aim was to assess effects of atmospherically transported Hg on fish Hg levels, by investigating temporal trends and spatial patterns of Hg concentrations in fish from Fennoscandia. A starting point for the work was to collect all available fish Hg data from Fennoscandia (including Norway, Sweden, Finland, and the Kola Peninsula in Russia). Following the collection of data, which included data from national monitoring programmes, peer-reviewed literature, scientific reports, and university theses, we created a database for historical data covering six decades (i.e. 1965 – 2015). The database was used to evaluate spatial patterns and temporal trends of Hg concentrations in freshwater fish from Fennoscandian freshwater ecosystems. No limitations were made for the data included in the database with respect to geographical location, natural background levels of Hg, or fish species and numbers of fish specimens per lake.

In short, this report presents:

- An overview of freshwater fish Hg concentrations for several fish species and regions across Fennoscandia;
- A comparison of different methods for normalisation of fish Hg data, used to account for effects from relationships between fish size and Hg levels;

- Spatial and temporal patterns of Hg concentrations in freshwater fish from Fennoscandia;
- Temporal trends for a subset of lakes across Fennoscandia that are being subject mainly to sources of Hg from atmospheric transport.

# 3 Background and status of knowledge

## 3.1 Mercury in Fennoscandian ecosystems

An assessment of available data on Hg concentrations in air, precipitation, sediments and fish from the Nordic countries was published in 2007, containing an analysis of 33 116 individual fish in total, collected in the period covering 1965 to 2004 (Munthe et al., 2007). Of the fish collected for the Munthe et al. (2007) report, a large portion of the data (> 60 %) was based on measurements of Hg in pike from Swedish monitoring programmes. One of the main conclusions from the report was that Hg levels in Nordic ecosystems were still, despite reduced emissions, influenced and affected by long-range atmospheric transported Hg from outside the Nordic countries, particularly central Europe. However, fish Hg concentrations were also influenced by historical emissions of Hg (as catchment stored Hg) and different ecosystem characteristics also affects the bioaccumulation of Hg (Munthe et al., 2007).

Available databases for observations of Hg concentrations in fish from freshwater ecosystems are often country specific and temporal trends are analysed based on geographic drivers rather than larger inter-regional drivers (e.g. atmospheric deposition, patterns of organic matter (OM) concentrations etc.). Efforts are needed to broaden the geographic scope of freshwater fish Hg databases, in order to allow for broader inferences to be made on controls on Hg concentrations in fish in the global north. Publications show that concentrations of Hg in many fish populations are increasing throughout the boreal forest regions, including the Nordic countries (e.g. Åkerblom et al., 2012; Braaten et al., 2014b; Miller et al., 2013) and North America (e.g. Gandhi et al., 2014; Riget et al., 2011, see details below). However, many regions and lakes also show trends of decreasing concentrations (e.g. Åkerblom et al., 2014; Åkerblom et al., 2012; Riget et al., 2011).

## 3.2 Spatial and temporal distributions

In thousands of North American and Scandinavian freshwater lakes, fish Hg concentrations exceed limits advised for human consumption (0.3 – 1.0 ppm Hg ww, UNEP, 2002). A compilation of multi-annual studies of Hg levels in terrestrial, freshwater and marine biota in polar and circumpolar areas in North America and Scandinavia, under coordination of the Arctic Council, suggests that neutral and rising trends of Hg are dominating (Riget et al., 2011). Riget et al. (2011) states that data on Hg in fish covering the past one to three decades can be used to illustrate how Hg concentrations have changed in recent times and will also yield insight into likely near-time future trends. However, only a few time series for freshwater fish were included in the review by Riget et al. (2011).

Larger parts of Norway, Sweden and Finland are part of the *boreal forest zone*, a circum-global belt in the northern hemisphere, located between the subarctic and the temperate forests. A large amount of Hg research, including analyses of several ecosystem matrices (i.e. sediments, soils, waters and biota), are available from the boreal forest zone, due to the large amount of historically deposited Hg stored in these soils, continuously affecting the adjacent lakes and their food chains (Fitzgerald et al., 1998).

### 3.2.1 Spatial patterns of mercury concentrations in freshwater fish

Surface waters of natural boreal lakes without local Hg contamination sources usually show low concentrations (ng/L, parts per trillion, ppt) of Hg (e.g. Braaten et al., 2014a; Eklof et al., 2012). In such systems, atmospheric deposition of Hg is the main source of Hg contamination (Jackson, 1997) and has led to long-term accumulation of Hg in catchments (Fitzgerald et al., 1998). In areas where the local influence of Hg contamination is minimal (e.g. typical for many regions in the boreal forest zone), there is a clear trend of increasing aquatic Hg concentrations from north-to-south (Braaten et al., 2014a). Similar increases in Hg along a north-south gradient in Scandinavia have been observed in mosses (Berg et al., 2006), surface sediments (Munthe et al., 2007) and in freshwater fish (perch: Fjeld, 2010; pike: Åkerblom et al., 2014). Patterns from Sweden are demonstrated by Åkerblom et al. (2014), covering four decades of Swedish fish Hg monitoring.

Miller et al. (2013) found no significant statistical spatial pattern in perch Hg concentrations covering Sweden and Finland. The authors suggest that the lack of a typical north-to-south increase in fish Hg concentrations is due to the influence of various biological effects (e.g. age, size and diet) on Hg bioaccumulation (Miller et al., 2013).

### 3.2.2 Temporal trends of mercury concentrations in freshwater fish

In recent years, a range of studies have emerged in the peer-reviewed literature focusing on temporal trends of Hg concentrations in fish (Table 1). Studies from the boreal ecozone are particularly abundant, with the Arctic also being present in recent years. Common for the available studies is the fact that they are representing relatively short time periods (< 20 years) or that they are restricted to a certain country or smaller region (e.g. Sweden, south-east Norway, Ontario, Canada, etc.). In Table 1 we summarise the current literature (including both reports and peer-reviewed papers) on temporal trend studies of Hg in fish from the Fennoscandic countries.

Areas where temporal trend analysis has revealed increases in fish Hg concentrations include Sweden (Åkerblom et al., 2012), Finland (Miller et al., 2013), Norway (Fjeld, 2009) and Canada (Ontario, Gandhi et al., 2014), although this rising trend is not found in all regions and for all fish species. Recent studies from lakes in Sweden (Åkerblom et al., 2014; Miller et al., 2013) in fact show declining concentrations of Hg in fish between 2005 and 2015. Given these mixed results, there is a clear need for more consideration of year-to-year variations. Gandhi et al. (2014) considered time trends for different fish species in a study from Ontario, Canada, where species-specific differences in accumulation of MeHg (as according to Bhavsar et al., 2010) were included for different time periods between 1970 and 2012. It was shown that while fish Hg concentrations from 1970 to 1990 were declining, concentrations in recent decades (time periods 1985-2005 and 1995-2012) were increasing. Overall (1970-2012), patterns were shown to be neutral or declining, depending on the fish species considered (Gandhi et al., 2014).

A similar study to the one by Gandhi et al. (2014) were the same year published by Åkerblom et al. (2014), investigating temporal trends of Hg concentrations in Swedish pike. To date, the study by Åkerblom et al. (2014) is the work relaying on the largest database when analysing historical trends, both with respect to the time period covered (1965-2012) and the number of individual fish included (n = 44 927). The authors documented an overall long-term decline in mean fish Hg concentrations of approximately 20 % during the time period 1965 – 2012, but found no consistent regional pattern that could explain this (Åkerblom et al., 2014).

In summary, there is at present not enough evidence to suggest that Hg contamination in fish is generally declining in Fennoscandia, despite temporal trends of Hg deposition having declined for years throughout Europe (EMEP, 2016).

## 3.3 Atmospheric mercury concentrations

Despite reduced Hg emissions in several regions of the world (Streets et al., 2011) and reduced or unchanged atmospheric Hg deposition in Northern Europe (Harmens et al., 2008; Torseth et al., 2012; Wangberg et al., 2007) and Canada (Cole et al., 2014), Hg budgets show that emissions to the atmosphere are increasing on a global scale (Pirrone et al., 2010). The increase is mainly related to anthropogenic activity in Asia (Streets et al., 2011), and unless emission controls are widely implemented, this trend is expected to continue. EMEP, a co-operative programme for monitoring and evaluation of the long-range transmission of air pollutants in Europe, reported in 2016 that annual Hg deposition fluxes in the EMEP region had, on average, declined with 1.2 % between 1990 and 2012 (EMEP, 2016). The decline was larger between 1990 and 2001 (1.8 % annually) than between 2002 and 2012 (0.5 % annually). The EMEP Hg data is based on measurements from 12 sites in Europe determining Hg in air and precipitation, covering an area from Finland to the north and Portugal to the south, Ireland to the west and Lithuania to the east.

**Table 1** A summary of studies (reports and peer-reviewed literature) investigating temporal trends of mercury (Hg) concentrations in fish from the Fennoscandic region. Shown for each study is species of interest, direction of temporal trend (↑ indicates significantly increasing; ↓ indicates significantly decreasing; ↔ indicates no significant trend), country/region and study years/period. The list is sorted by year of sampling of the oldest fish included in each study.

| Citation | Study period | Fish species | Country/region | Number of populations (lakes) | Direction of trend |
|---|---|---|---|---|---|
| **Munthe et al. (2007)** | 1965-2004 | Arctic charr, perch, pike, trout, whitefish | Finland, Norway and Sweden | 2758 | n.a. |
| **Åkerblom et al. (2014)** | 1965-2012 | Arctic charr, perch, pike, roach, trout + 10 others | Sweden | 2881 | ↓ |
| **Paasivirta et al. (1981)** | 1970-1980 | Pike | Finland | 1 | ↓ |
| **Åkerblom and Johansson (2008)** | 1972-2006 | Perch, pike | Sweden | 2223 | ↑↓ |
| **Paasivirta and Linko (1980)** | 1973-1978 | n.a. | Finland | 2 | ↔ |
| **Miller et al. (2013)** | 1974-2005 | Perch | Finland and Sweden | 341 | ↑↓ |
| **Fjeld et al. (2010)** | 1990-2010 | Perch | South east Norway and Northern Norway | 5 | ↑↔↓ |
| **Fjeld and Rognerud (2009)** | 1991-2008 | Perch | South east Norway | 10 | ↑↔ |
| **Rask et al. (2007)** | 1993-2003 | Perch, pike | Finland | 1 | ↑↓ |
| **Åkerblom et al. (2012)** | 1994-2006 | Pike | South to mid Sweden | 25 | ↑↔↓ |
| **Braaten et al. (2014b)** | 2010-2012 | Perch | South east Norway | 2 | ↑ |

## 3.4 Historically accumulated stores of atmospheric mercury in soil

In many boreal, subarctic and Arctic lakes in Fennoscandia, long-range atmospheric transport of Hg is the main source of Hg contamination (Jackson, 1997) and has led to long-term accumulation of Hg in lake catchments (Fitzgerald et al., 1998). Hg bound in the bedrock (geogenic Hg) could also potentially be an important source of Hg to freshwater systems. However, measurements of Hg and inorganic sediment fractions in the Nordic countries have documented that Hg is almost entirely associated with the organic fractions, typical for areas with bedrock low in Hg (Munthe et al., 2007).

Due to catchment retention, atmospheric inputs of Hg do not correlate directly to Hg in freshwaters (Larssen et al., 2008), and catchment loading of Hg to lakes can often exceed direct on-lake Hg deposition (Lee et al., 2000; Lee et al., 1998). A large manipulation study in North America (*The Mercury Experiment to Assess Atmospheric Loading in Canada and the United States*, METAALICUS), where Hg were added to the catchment as well as the lake, showed that an increase in Hg loading of approximately 7 times the ambient wet deposition gave increased concentrations in biota (30-40 %, including young of the year fish) over a three-year period (Harris et al., 2007). Harris et al. (2007) state that "*essentially all of the increase in fish MeHg concentrations came from Hg deposited directly to the lake surface. In contrast, <1% of the Hg isotope deposited to the watershed was exported to the lake.*" Based on this, the authors suggest that lakes receiving reduced input of Hg from the atmosphere due to increased emission controls, would lower their fish Hg concentrations. The decline in the Hg content of fish would be rapid, as a result of reduced direct deposition to the lake, followed by a slow (centuries-long) further decline due to re-equilibration of the catchment pools. The size of the initial response to reduced deposition will strongly depend on the catchment area to lake area ratio.

Since most Scandinavian lakes have a large catchment relative to the lake surface, the findings from the North American manipulation study would imply that only a small initial response to reduced atmospheric input can be expected, and the catchment pools of Hg will be of major importance compared to direct atmospheric deposition to the lake (Larssen et al., 2008; Lee et al., 2000). From Larssen et al. (2008) (and Lee et al., 2000) it is estimated that pristine catchments can contain pools of Hg that are 8000 (and 15500) times larger than the annual stream water output and 2000 (and 600) times larger than the input from throughfall and litterfall. The response of reduced atmospheric deposition should therefore be expected to be very slow.

## 3.5 Policy regulations

In November 2013, the Minamata Convention on Mercury was signed by 93 countries, aiming to protect human health and the environment from adverse effects of Hg at a global scale (UNEP, 2014). On May 18$^{th}$, 2017, the 50-ratification milestone required for the Minamata Convention to enter into force was reached, and on August 16$^{th}$, 2017 the Convention became legally binding for all its Parties. At present, 128 countries have signed the Convention and 74 have ratified it. All the Fennoscandian countries have signed the Convention, and Norway, Sweden and Finland have also ratified it.

An important aspect of the current work and its relevance for the Minamata Convention is that policy on Hg in the environment must acknowledge the large Hg stores in the environment, accumulated from centuries of anthropogenic Hg emissions. The accumulated Hg may be mobilized and contaminate aquatic food webs for centuries to come, and climate change may enhance both mobility of recently and historically deposited Hg. For the evaluation of the effectiveness of the Convention (Article 22), these aspects, in combination with monitoring of Hg in the environment (e.g. fish), are essential for the global success of the Minamata Convention.

# 4 Materials and methods

## 4.1 Selection of data

The database comprised data from Fennoscandia. Fennoscandia consists of Norway, Sweden, Finland, and the Russian regions of Karelia and the Kola Peninsula. No measurements of Hg concentrations in fish were found for the Karelian area (in the available literature or among partners), and as such, we have only included fish from Russian lakes located on the Kola peninsula in the current database. The data is collected from all available literature and databases, including national monitoring programmes, peer-reviewed literature, scientific reports, and university theses (MSc).

The collection of data is not restricted to the boreal zone (where a large amount of work is previously done, Table 1). We have also included lakes from further north (i.e. subarctic areas of Norway, Sweden and Finland) and from further south (i.e. southern parts of Sweden), in order to not restrict the database to typical geographical regions.

The toxicity and potential human health effects from Hg contamination and pollution, particularly for the organic form methylmercury (MeHg), became evident after the Minamata accident in Japan in the 1950s (Kurland et al., 1960). Monitoring and research activities of Hg in fish have been carried out in Fennoscandia since at least the 1960s, and we found available data covering these last six decades (1960s-2015). We acknowledge that more recent investigations of Hg concentrations in fish have also been undertaken, i.e. while the work with this report has been on-going, but we have chosen to not include data from 2016 and later for practical reasons related to quality assurance and statistical analysis of the database.

In addition to fish Hg concentrations we made an effort to collect as much metadata as possible (as a minimum, the geographical location and name for the lake or river of interest, fish size (length, weight) and fish species name). For a selection of the studies, other types of metadata for the individual fish were available as well, including stable isotopes (nitrogen and carbon), age, sex and maturity stage. Where available, this has also been added to the database (Table 2).

In the initial collection of data, no restrictions were made with respect to whether the lakes/rivers of interest had any local Hg contamination source or not (see section 3.5 below for details on this).

## 4.2 Data quality assessment and data selection

In compiling a fish Hg database of the current magnitude, the authors appreciate the fact that a thorough quality assurance of each individual measurement with respect to all data is nearly impossible. To ensure that the final data included was of the best possible quality a selection of entries were removed. This means that from the original database of 66 464 individual fish measurements, 11 904 measurements were eliminated (approximately 18 %).

To overcome obvious errors present in the database, data entries where measurements of individual fish Hg concentrations were not recorded were removed. These entries were of little interest to the current work (n = 2 801). In order to calculate adjusted fish Hg concentrations, and to test relationships between Hg concentrations and fish characteristics, weight measurements were ensured to exist for the complete data set (see description on data treatment further on) and entries were removed where weight was not included (n = 3 600). With respect to fish size, length and/or weight versus Hg concentrations, and length versus weight relationships were also tested (for a detailed presentation of the relationships we refer to section 4.3 below). Following these relationships, residual outliers (i.e. entries where length and weight did not correspond to a typical relation within a lake or fish species, n = 70) were excluded.

**Table 2** A summary of the data included for each entry in the finalised database. Note that not all the below-mentioned parameters exist for all entries in the database. Parameters written in italic with an asterisk were considered a minimum for a measurement to be included in the database.

| Database specification | Data specifications | Description |
|---|---|---|
| *Geographical location** | (latitude, longitude) | Given as WGS84 decimals |
| *Geographical name** | Lake name | Official name given in the local language |
| Fish parameters<br>• *Length**<br>• *Weight**<br>• Age<br>• Sex<br>• Maturity stage | <br>• Centimetres, cm<br>• Grams, g<br>• Years<br>• 1 = male, 2 = female<br>• 1-12 | <br>• Total fish length (not fork length)<br>• Total weight<br>• Decided by inspection of operculums, scales or otoliths<br>• Decided by inspection of reproductive glands<br>• Classified by Dahl (1917), described by Jonsson and Matzow (1979) |
| *Fish species name** | Species name | English and Latin name given |
| Fish chemical measurements<br>• *Mercury**<br>• Stable isotopes | <br>• mg/kg, ppm<br>• δN and δC | <br>• Numbers given as wet weight<br>• Dried samples, not baseline corrected |

The database was limited to the fish species with a significant geographical distribution throughout Fennoscandia (i.e. the fish is represented throughout the four countries) and fish caught in a significant number (i.e. representing at least 1 % of the total database amount). This included Northern pike (*Esox lucius*, 42.4 %), perch (*Perca fluviatilis*, 34.1 %), Arctic charr (*Salvelinus alpinus*, 1.2 %), brown trout (*Salmo trutta*, 3.1 %) and roach (*Rutilus*, 1.3 %). Burbot (*Lota lota*), although present throughout Fennoscandian waters, was only available from Finnish lakes in the present database, and the data were taken out of the final database.

To be able to calculate a representative median or mean concentration per lake or river represented in the study, a minimum of 5 measurements of fish Hg concentrations needed to be available per water body. All lakes or rivers where n < 5 measurements were collected and reported, were removed from the final database.

## 4.3 Fennoscandian fish species

Numerous freshwater fish species are represented throughout Fennoscandia, showing large variability in their Hg concentrations. The challenges related to analysing temporal trends of fish Hg concentrations are related to separation of the non-temporal variance between lakes from underlying temporal patterns. The non-temporal variance is related to differences in factors including fish growth rates, fish body sizes, food web structure, and Hg trophodynamics (Lavoie et al., 2013; Watras et al., 1998). These effects cause variation both within and between lakes. The five main fish species from the current database were:

- Pike (*Esox lucius*) is found in freshwaters throughout the Northern hemisphere (including Europe, Russia and North America) and has been studied with respect to accumulation and magnification of Hg for six decades (Johnels et al., 1967). Pike is a piscivorous fish, often shown to feed exclusively on fish throughout their life (Sharma et al., 2008) and hence, significantly accumulate Hg (Vøllestad et al., 1986). The fish feed on larger prey as they grow (Lawler, 1965) and typically inhabit shallow areas of the lake (Nursall, 1973).
- Perch (*Perca fluviatilis*), similar to pike, has been shown to accumulate significant amounts of Hg, including based on numerous studies from the Fennoscandian countries (e.g. Braaten et al., 2014b). However, its dietary preferences differ from that of pike. Perch undergo an ontogenetic (i.e. developmental) shift in diet as they age (Collette et al., 1977), shifting from a pelagic zooplankton diet as juveniles to a diet dominated by benthic invertebrates at intermediate sizes, then becoming piscivorous when large enough (typically 90 – 240 mm, Hjelm et al., 2000). Hence, perch feed at different trophic levels through their lifetime. Perch are also non-migratory (Collette et al., 1977), which makes the fish ideal for examining patterns of local Hg concentrations.
- Arctic charr (*Salvelinus alpinus*) is the northernmost freshwater fish and the only fish species that is truly “holoarctic”, being present on all land masses of the Arctic region (Reist et al., 2006). Tremendous morphological and ecological variability within this species has been reported, with its flexibility in diet and life-history considered adaptive for inhabiting Arctic regions (Power, 2002; Reist et al., 2012). Arctic charr also inhabits the boreal region, and is represented throughout Fennoscandia. Although habitats can differ between the two species, Arctic charr dietary patterns are similar to that of perch, with the fish experiencing ontogenetic dietary shifts (Eloranta et al., 2010).
- Roach (*Rutilus rutilus*) is an omnivorous fish, feeding on zooplankton, invertebrates, algae, epiphytes, macrophytes and detritus (Brabrand, 1985). Because of their diet, roach does not accumulate Hg to the same degree as higher trophic level fish such as pike and perch. Roach is commonly found all over Europe, in lakes and rivers (Kottelat and Freyhof, 2007).
- Brown trout (*Salmo trutta*) has a similar dietary pattern as that of perch and Arctic charr, although large variation in diet has been documented. Brown trout include a wide range of prey in their diet, ranging from small zooplankton to relatively large fish (Jonsson, 1989). The brown trout has been shown to become piscivorous at a length of 20-25 cm (L’Abée-Lund et al., 1992).

## 4.4 Data treatment

When Hg concentrations in fish are to be compared between lakes, years and seasons, a length and/or age adjustment is needed due to the strong co-variation between Hg concentration and fish size (i.e. length and weight; Sonesten, 2003, Chasar et al., 2009) as well as fish age (Braaten et al., 2014b). To overcome the effect of a size-Hg relationship, several attempts to normalise, adjust or correct fish Hg concentrations have been applied for comparison across lakes and regions over time. Commonly used correction techniques includes co-variance analyses creating general linear models utilising length (Fjeld, 2009), weight (Hakanson et al., 1988; Sonesten, 2003) or age (Braaten et al., 2014b) adjusted concentrations; simply dividing Hg concentrations by the fish weight (Johnels et al., 1967); to select fish within certain size ranges (Åkerblom et al., 2012), individual adjustment to standard species-specific weights (Munthe et al., 2007); and individual normalisation using non-linear species-specific transfer functions (Åkerblom et al., 2014; CLRTAP, 2016). Choice of method partly depends on the availability of supporting data.

In the present report two techniques were applied, and included in our analysis are the following presentations of fish Hg concentrations:

1. ***No adjustments***: A measurement of the Hg concentration in a fish at a given time for a specific lake is the value most accurately describing "the current situation". Based on the size of our database, a "normalisation" might not be necessary and we have tested patterns of trends without adjusting the observed Hg concentrations ($[Hg]_{obs}$).
Throughout the report, we refer to these concentrations as **observed values/concentrations**.

2. ***Weight adjustments***: As for length, weight is often used as an estimate of fish size (and hence age). Weight was recorded for all the data entries in our database, and Hg concentrations showed a significant positive correlation with weight (all data, $r^2$=0.41, $p$<0.0001, Figure 8, n = 54 563), included all the five main species pike ($r^2$=0.08, $p$<0.0001), perch ($r^2$=0.19, $p$<0.0001), Arctic charr ($r^2$=0.10, $p$<0.0001), brown trout ($r^2$=0.17, $p$<0.0001) and roach ($r^2$=0.10, $p$<0.0001, Table 3). The weight adjustment was done by "normalising" individual $[Hg]_{obs}$ to the species-specific population mean through the following function:

$$[Hg]_{weight} = ([Hg]_{obs}/ W_{obs})* W_{mean} \qquad (1)$$

where the $[Hg]_{obs}$ was divided by the individual fish weight ($W_{obs}$) and multiplied with the species-specific population mean weight ($W_{mean}$). $W_{mean}$ were 998.0 g, 77.5 g, 304.7 g, 725.1 g and 127.6 g for pike, perch, Arctic charr, brown trout and roach, respectively.
Throughout the report, we refer to these concentrations as **weight adjusted values/concentrations**.

3. ***ICP M&M manual adjustments***: As described in an ICP M&M manual (CLRTAP, 2016), a standardisation of fish Hg concentrations can also be done by calculating $[Hg]_{obs}$ to correspond to a 1-kg pike in the same lake ($[Hg]_{std}$). This is done by utilising an empirically supported transfer function applicable to any fish species at any site (CLRTAP, 2016):

$$[Hg]_{std} = [Hg]_{obs}/(f_{HgY} + f_{HgW}\, W_{obs}^{2/3}) \qquad (2)$$

In the mathematical function (2), $W_{obs}$ is the fish body size in kg (as a unitless value), $f_{HgY}$ is a parameter representing the concentration ratio between newly-hatched young fish and 1-kg pike, and $f_{HgW}$ is a species-specific empirical coefficient. We set parameters to default values,

obtained from the ICP M&M manual (CLRTAP, 2016) and used by Åkerblom et al. (2014): $f_{HgY}$ = 0.13 for all species, $f_{HgW}$ = 0.87, 1.65 and 1 for pike, perch and all other species, respectively. The robustness of the parameters used for standardisation of fish Hg concentrations between lakes and over time needs to be considered for analysis of spatial patterns and temporal trend.
Throughout the report, we refer to these concentrations as **standard 1-kg pike concentrations**.

Based on the obtained observations and the approaches for normalisation and adjustments above we ended up with three sets of Hg concentrations for each individual fish, including **observed**, **weight-adjusted**, and **standard 1-kg pike concentrations**. These data are then used to create lake-specific median values based on fish data from each lake. This reporting is done both for each species separately, and for all five main fish species combined (see below for details).

## 4.5 Classification of lakes based on potential local contamination

In order to investigate temporal trends of Hg concentrations in Fennoscandian fish and relate this to differences in atmospheric Hg, it is critical to separate lakes that are influenced by local industry point sources from lakes that are not. Industries known to have released Hg historically to the environment in the Nordic countries include pulp and paper industry, and chlor-alkali industry. Examples include Lake Vänern, where a chlor-alkali plant released from three to five tons of mercury annually before new legislations were introduced in the 1970s and 1980s (Danielsson et al., 2002). To account for differences in the Hg input to individual lakes, the lakes in the database were characterized according to the following groups:

1. *Lakes subject mainly to atmospheric deposition of Hg with no known local point sources of Hg (i.e. "un-contaminated lake", or "lake only influenced by air pollution of Hg");*
2. *Lakes where sources are possible or the situation is unclear (i.e. "lake possibly contaminated from local sources"); and*
3. *Lakes with known local point source(s) of Hg (i.e. "contaminated lake").*

It is extremely complicated to provide a complete list of Fennoscandian lakes where some sort of local Hg pollution source has been present throughout history. Hence, we did not distinguish between when the lake was potentially contaminated and when it was sampled: If a lake had a local point source (e.g. paper industry or other types of industry) in the period 1975 – 1988, and the fish was sampled in 1969 or 2014, we still characterized this as a "contaminated lake".

Throughout the report, we refer to the groups "1", "2" and "3" above as **air pollution** lakes, **unknown** lakes, and **contaminated** lakes, respectively.

## 4.6 Statistical models (spatial patterns and temporal trends)

Spatial and temporal patterns of Hg concentrations in fish populations were explored using lake median fish Hg concentration (see below for details on fish species of interest). This procedure was used for observed fish Hg concentrations, in order to reflect the *actual* fish Hg concentration in the lakes. Following this, we also did a spatial interpolation based on median concentrations per lake for the total database. After this initial spatial assessment, we used existing literature from Scandinavia, and created interpolated maps of Hg concentrations in sediments and aqueous total organic carbon

(TOC) concentrations. Spatial patterns were also presented for two subgroups of the database: 1) for air pollution lakes classified as "1" above (see section 3.5, "lakes only influenced by air pollution of Hg"); and 2) for lakes classified as "1" where data existed for at least 5 years in the database. For all these investigations, we presented data based on the three different approaches for normalisation of Hg data (see section 3.4).

Temporal trends were first investigated by linear regressions and smoother lines for the total database (including all fish species), for each of the three normalisation methods (observed Hg concentrations, weight adjusted Hg concentrations, and standard 1 kg pike concentrations). Correlation coefficients were used to statistically describe the relationship between fish Hg concentrations and latitude as well as sampling year in the database. A probability for each correlation ($p$) was used to estimate significance for the analysis. Unless otherwise mentioned, a significance level of $\alpha = 0.05$ was used.

## 4.7 Evaluation of temporal trends in subset of lakes

In order to investigate the temporal trends in more detail and to look for drivers behind the trends, we did more detailed analyses on lakes classified as "1" (lakes subject to atmospheric deposition of Hg) and where more than 5 years of data existed. After assessing the temporal trends in fish Hg concentrations for Fennoscandia throughout the study period 1965-2015, we narrowed the database following certain criteria, in order to investigate and explore drivers of the potentially changing fish Hg concentrations. We selected data from lakes mainly influenced by Hg from atmospheric deposition (n = 17 015 specimens, n = 683 lakes, see section 3.5), followed by a second selection of lakes that were sampled in at least 5 years (i.e. a median value was available from at least 5 different years) throughout the study period (n = 5 553 specimens, n = 72 lakes).

# 5 Results and discussion

## 5.1 A general description of the database

### 5.1.1 Origin of data and fish species

Data for a total of 66 464 individual fish were collected, where approximately 60 % of these were from Sweden and Swedish monitoring programmes (Figure 2). Following Sweden, Finland had the largest part of the database (29.6 %), with Norway comprising approximately 10 %, and Russia only 0.3 %.

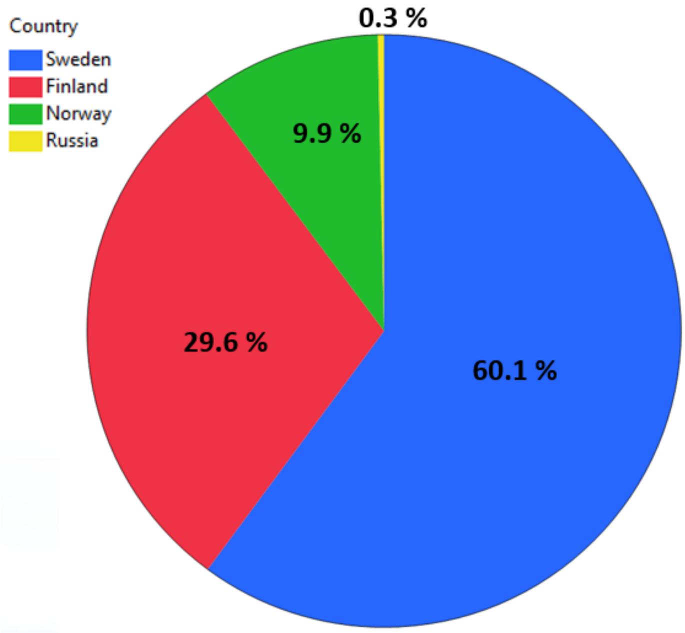


**Figure 2** The relative distribution of the source of the data collected for the database. Data for a total of 66 464 fish were included. Data from the four countries Sweden, Finland, Norway and Russia are represented by the colours blue, red, green and yellow, respectively.

Almost 90 % of data entries in the database were registrations of pike (n=33 699) and perch (n=24 960, Figure 3). The main reason for this is the focus on pike in Swedish monitoring programmes, and perch in Finnish and Norwegian monitoring. Following pike and perch, brown trout (n=2 343), roach (n=1 368), and Arctic charr (n=861) were dominant fish species, which when combined accounted for approximately 8 % of the database (Figure 3). In addition to these five species, a total of 3 233 specimens were distributed among 10 other species, including burbot (n=916), common bream (*Abramis brama*, n=543), crucian carp (*Carassius carassius*, n=13), European smelt (*Osmerus eperlanus*, n=29), grayling (*Thymallus thymallus*, n=21), Atlantic salmon (*Salmo salar*, n=14), tench (*Tinca tinca*, n=4), vendace (*Coregonus albula*, n=215), whitefish (*Coregonus lavaretus,* n=777) and zander (*Sander lucioperca*, n=160). Additionally, we were not able to assign a species for 300 fish measurements.

After removing entries with insufficient data or residual outliers (see 3.2 Data quality assessment and data selection) 54 563 specimens remained in the database. This includes the main five species of the database, representing at least five measurements from each lake included (Figure 3).

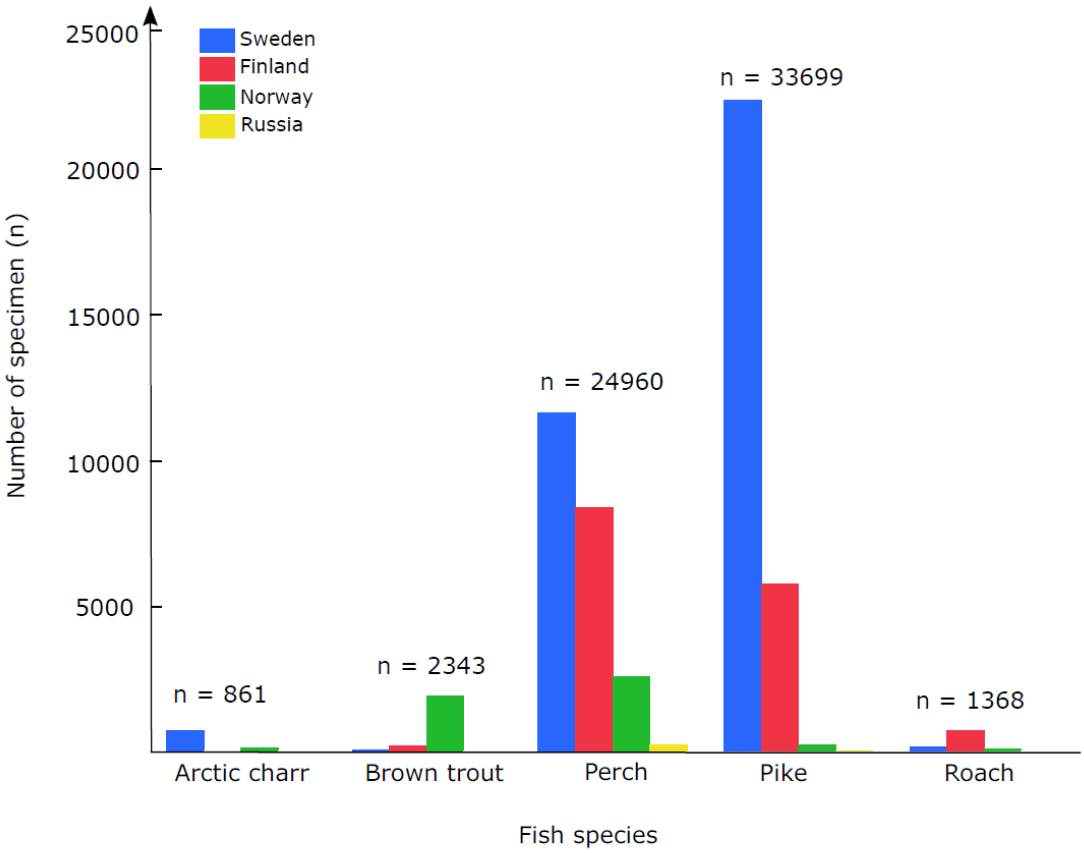


**Figure 3** The amount of fish collected for the five most intensively sampled species, including (from left to right) Arctic charr, brown trout, burbot, perch, pike and roach, per country. Data from the four countries Sweden, Finland, Norway and Russia are represented by colours blue, red, green and yellow, respectively.

The total number of lakes represented in the database is 2 775, which means that on average 22 fish was collected from each lake. Of the 2 775 lakes that were "sampled", 1 941 were Swedish, 688 were Finnish, 141 Norwegian and 15 Russian. The mean number of fish caught per lake varied between the countries: Sweden (13.9) and Russia (13.2) had similar numbers, while values from Finland (24.0) and Norway (41.8) were higher.

The earliest registered capture date of fish included in the present report is from Sweden in 1965 (Figure 4). A noteworthy effort was made from Swedish monitoring programmes particularly during the 1980s, representing the dominant fraction of the fish caught in that century (Figure 4). In Finland, monitoring for Hg in fish has been taking place for nearly as long as in Sweden, with the first measurements dating back to 1967. Finnish monitoring has been relatively stable throughout the study period, with a peak in measurements in the last 5-6 years. In Norway, the oldest gathered data is from 1983, while from Russia, the oldest data is from 2002. Norwegian data collection shows a clear peak between 2008 and 2012, related to NIVA led projects funded from NEA and RCN, respectively.

### 5.1.2 Lake locations and geography

Samples from a total of 2 775 Fennoscandic lakes were included in the database after the data quality assessment (Figure 5). The lakes span a south-north gradient from 55.495° N, 13.303° E in Southern Sweden (Börringesjöen) to 70.031° N, 25.556° E in Northern Norway (Gaerdusjavrit), and an west-east gradient from 59.037° N, 5.995° E in Western Norway (Åsvatnet) to 67.393° N, 37.374° E on the Kola Peninsula (Makarovskoye, Figure 5). The density pattern evident from the geographical locations of the lakes (Figure 5) reflects some factors that are known to influence Hg concentrations in fish:

*i)* distribution of fish species sampled during Hg monitoring efforts in Fennoscandia (see section 3.3);
*ii)* geographical location of people and where they fish (particularly relevant in Norway);
*iii)* locations of industries (see section 3.5, particularly relevant in Sweden); and
*iv)* the west-to-east and north-to-south TOC gradients in Fennoscandia (see section 4.4).

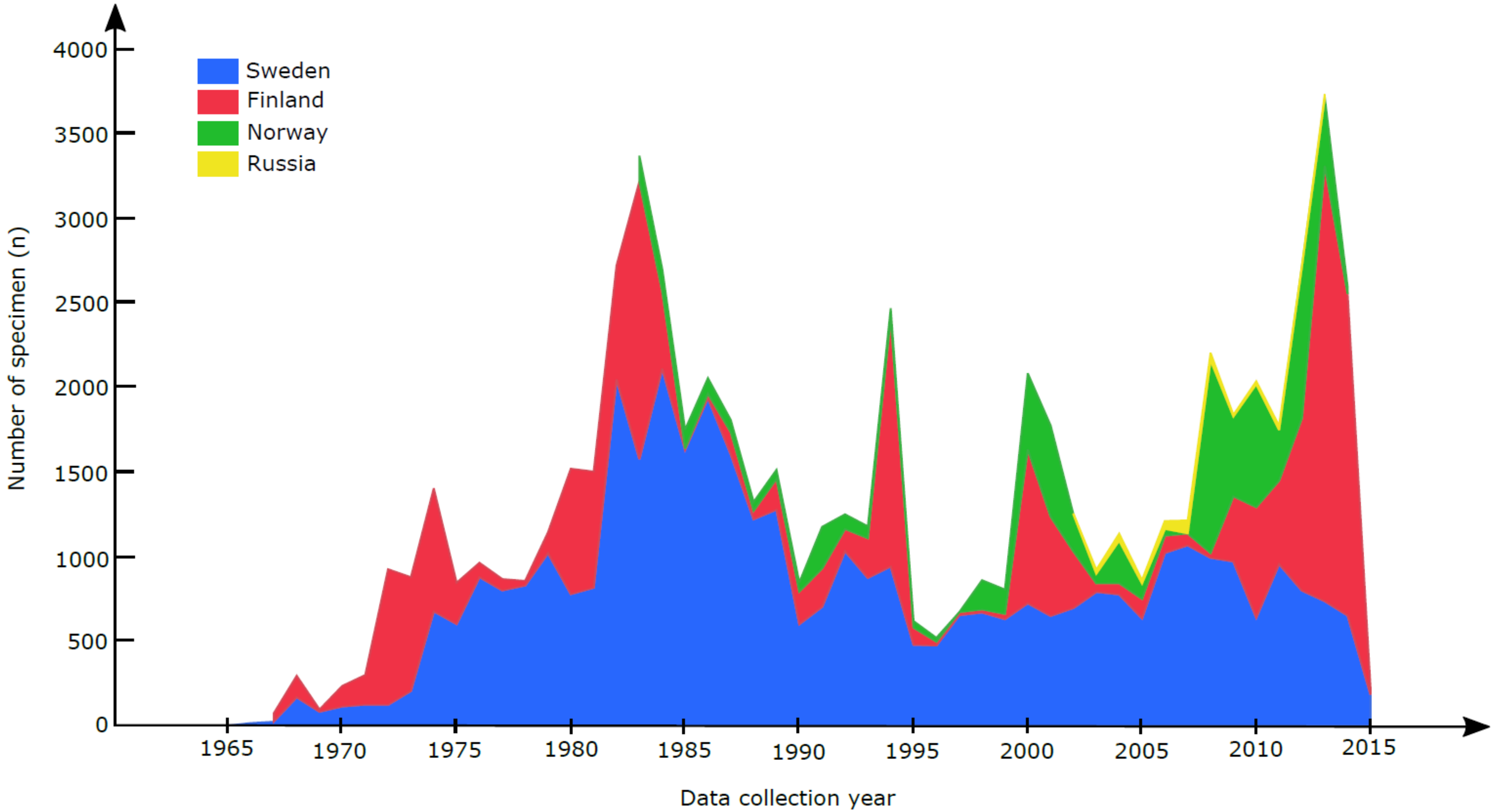


**Figure 41** The number of fish caught per year in the four Fennoscandic countries in the complete study period from 1965 to 2015. Included are data for the five main species: Arctic charr, brown trout, perch, pike and roach. Data from the four countries Sweden, Finland, Norway and Russia are represented by colours blue, red, green and yellow, respectively.

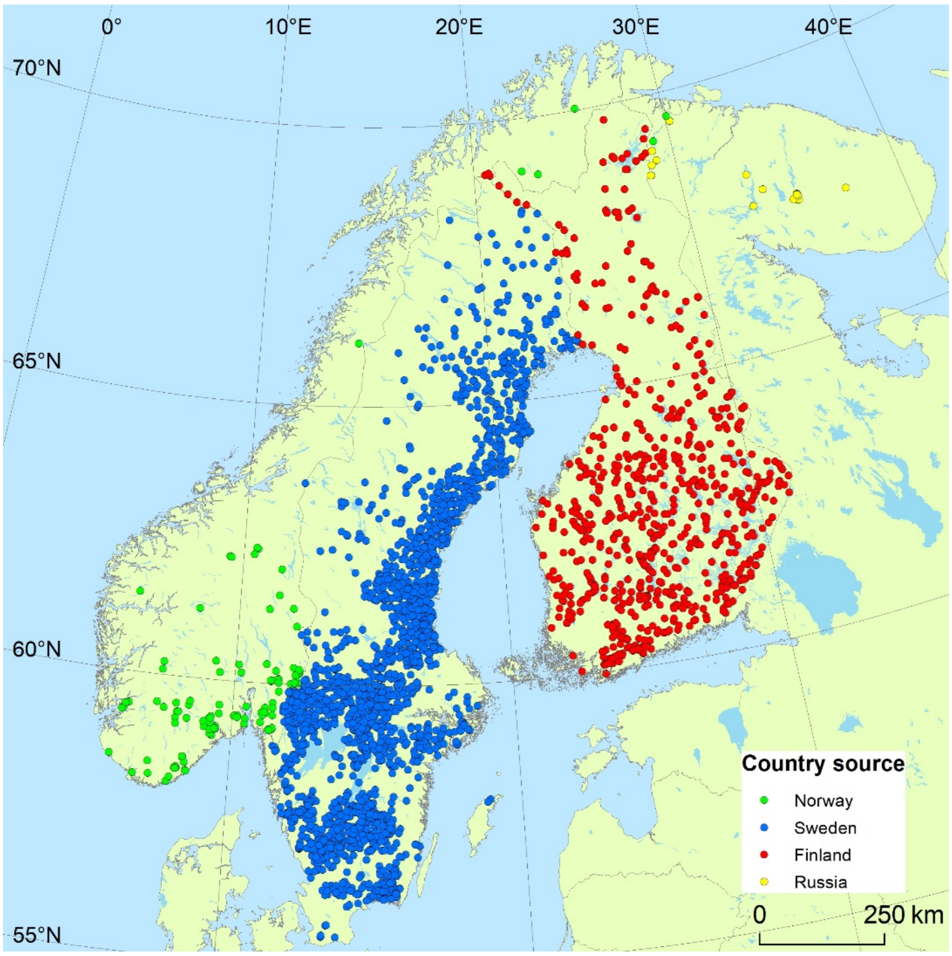


**Figure 5** The geographical location of the lakes (n = 3303) in the Fennoscandian fish Hg database. Colours represent country of data origin: Sweden (blue), Finland (red), Norway (green) and Russia (yellow). Lakes that are located in more than one country are represented with the colour from the country having the largest fraction of the lake area within its borders.

### 5.1.3 Fish morphology and bioaccumulation of mercury

The bioaccumulative properties of Hg are evident when investigating concentrations of Hg in fish muscle in relation to fish characteristics related to age (i.e. weight and length). For practical reasons (i.e. the time-consuming aspects related to age determination, typically done by microscopic inspection of otoliths or opercula), weight and length are more commonly recorded than age. For the present database, Hg concentrations were positively correlated with age (log Hg = -2.3 + 0.6*log age; $r^2$ = 0.19, $p$ < 0.0001, Figure 6), but fish that have been aged represent less than half of our database entries (n = 19 901). Figure 7 and Figure 8 show significant relationships between fish Hg concentrations and length and weight, which are often used as indicators of fish age. The fish Hg concentrations for the complete database showed a significant positive correlation with both length (Hg = 0.005 + 0.015* length; $r^2$ = 0.35, $p$ < 0.0001, n = 41 602, Figure 7) and weight (Hg = 0.312 + 0.3* weight; $r^2$ = 0.41, $p$ < 0.0001, n = 54 563, Figure 8). The significant relationships with length and weight also hold when considered on a species-specific basis for the five main fish species in the database (Table 3). As for the complete data set, the species-specific fish Hg-correlations are generally marginally stronger for weight regressions compared to the length regressions (the exception is brown trout correlations). Although the relationships between Hg concentrations and length and weight are significant (all $p < 0.05$), the amount of variation explained is generally low (all $r^2 < 0.20$), indicating that other factors also play a large role.

**Table 3** Specifications for the linear regressions between fish Hg measurements and weight and length for the five main fish species, Arctic charr, brown trout, perch, pike and roach.

| Specifications | Linear regression | $r^2$ | n | *p* |
|---|---|---|---|---|
| **Arctic charr** | | | | |
| - length | log Hg = -6.3 + 1.2*log length | 0.08 | 798 | < 0.0001 |
| - weight | log Hg = -4.4 + 0.4*log weight | 0.10 | 799 | < 0.0001 |
| **Brown trout** | | | | |
| - length | log Hg = -4.9 + 0.9*log length | 0.18 | 2041 | < 0.0001 |
| - weight | log Hg = -3.4 + 0.3*log weight | 0.17 | 2046 | < 0.0001 |
| **Perch** | | | | |
| - length | log Hg = -3.9 + 0.8*log length | 0.18 | 22369 | < 0.0001 |
| - weight | log Hg = -2.7 + 0.3*log weight | 0.19 | 22629 | < 0.0001 |
| **Pike** | | | | |
| - length | log Hg = -3.0 + 0.6*log length | 0.04 | 22112 | < 0.0001 |
| - weight | log Hg = -2.7 + 0.3*log weight | 0.08 | 28211 | < 0.0001 |
| **Roach** | | | | |
| - length | log Hg = -3.6 + 0.9*log length | 0.07 | 771 | < 0.0001 |
| - weight | log Hg = -2.6 + 0.3*log weight | 0.11 | 874 | < 0.0001 |

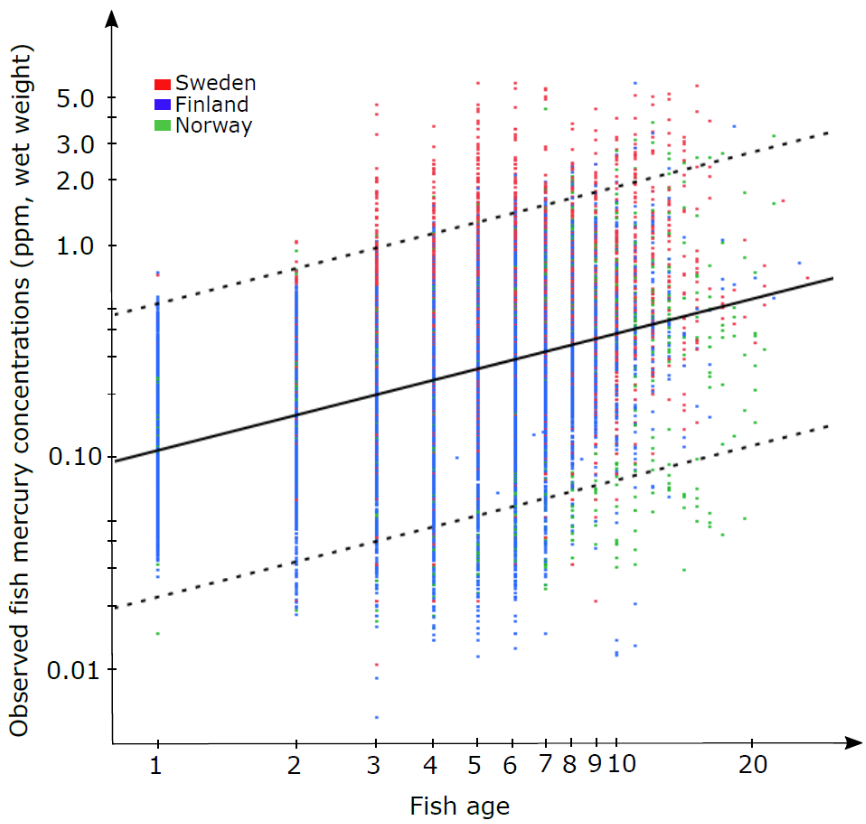


**Figure 6** A scatter plot of observed fish mercury (Hg) concentrations (ppm wet weight, y-axis) versus fish age (x-axis). The linear regression (log Hg = -2.3 + 0.6*log age; r2 = 0.19, p < 0.0001) is shown as the black solid line with dotted 95 % confidence interval lines. Colours represent country of sample origin (Finland = red, Sweden = blue and Norway = green).

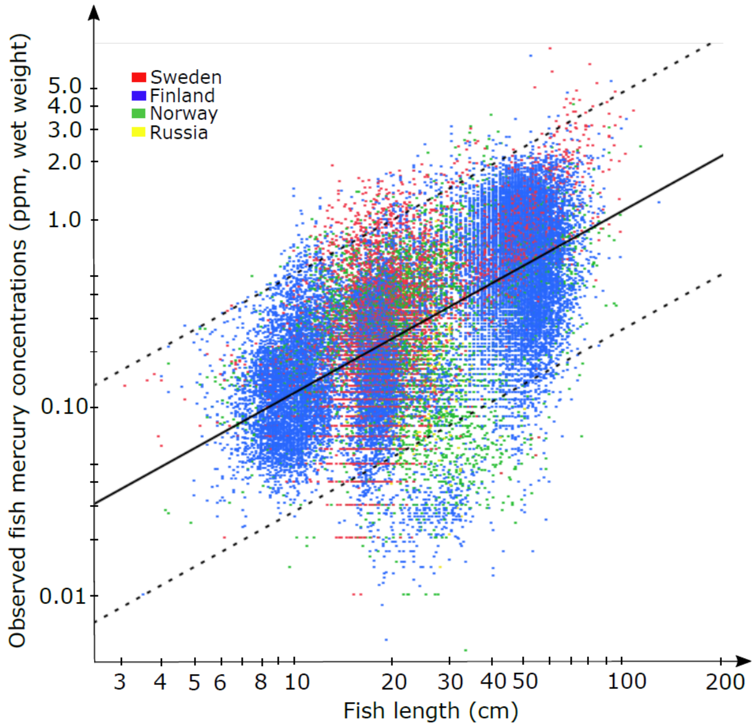


**Figure 7** A scatter plot of observed fish mercury (Hg) concentrations (ppm wet weight, y-axis) versus fish length (cm, x-axis). The linear regression (Hg = 0.005 + 0.015* length; r2 = 0.35, p < 0.0001) is shown as the black solid line with dotted 95 % confidence interval lines. Colours represent country of sample origin (Finland = red, Sweden = blue, Norway = green, and Russia = yellow).

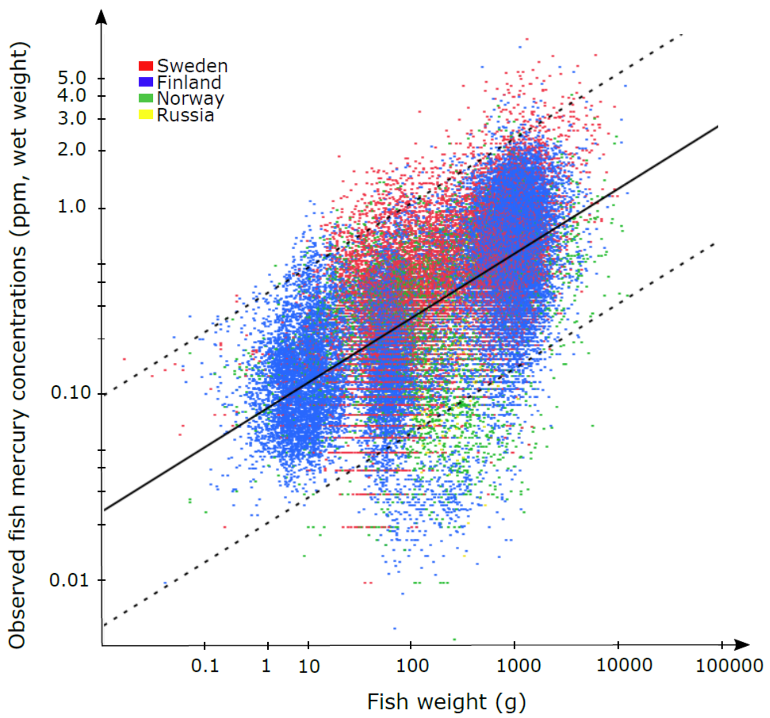


**Figure 8** A scatter plot of observed fish mercury (Hg) concentrations (ppm wet weight, y-axis) versus fish weight (g, x-axis). The linear regression (Hg = 0.312 + 0.3* weight; r2 = 0.41, p < 0.0001) is shown is the black solid line with dotted 95 % confidence interval lines. Colours represent country of sample origin (Finland = red, Sweden = blue, Norway = green, and Russia = yellow).

The relationships between concentrations of Hg in fish and age, length and weight (Figures 6-8), illustrate the biomagnification and bioaccumulation of Hg in aquatic food webs. MeHg concentrations increase with trophic position (Kidd et al., 1995), calculated from the ratio of heavier to lighter stable isotopes of nitrogen ($\delta^{15}$N, Kidd et al., 1999). However, analysis of $\delta^{15}$N increases the project budget costs related to determination of trophic transfer of Hg, and is often found to a much lesser extent than length and weight throughout literature. In our database, 3 973 entries included measurements of $\delta^{15}$N (Figure 9). The relationship between observed fish Hg concentrations and $\delta^{15}$N was significantly positive (log Hg = -2.72 + 0.12*$\delta^{15}$N, $r^2$ = 0.08, $p$ < 0.0001), and reflects the biomagnifying properties of organic Hg in these food webs. A weakness and possibly significant source of uncertainty related to these calculations would be that these data are not baseline adjusted. A common long-lived primary consumer is often used for within and between system comparisons of $\delta^{15}$N (Vander Zanden and Rasmussen, 1999). In a review by Lavoie et al. (2013), it was shown that MeHg biomagnification in aquatic food chains on a global scale is positively related to latitude. The mechanisms thought to be responsible for a possible south-north gradient mainly relate to temperature (discussed in Lavoie et al., 2013) and include growth dilution (increased in warmer regions, Simoneau et al., 2005), trophic transfer efficiency, which is reduced in warmer regions, and excretion rates of MeHg, which is reduced in colder regions (Trudel and Rasmussen, 1997). However, given the uncertainties related to data collected in this database, these mechanisms are not further investigated.

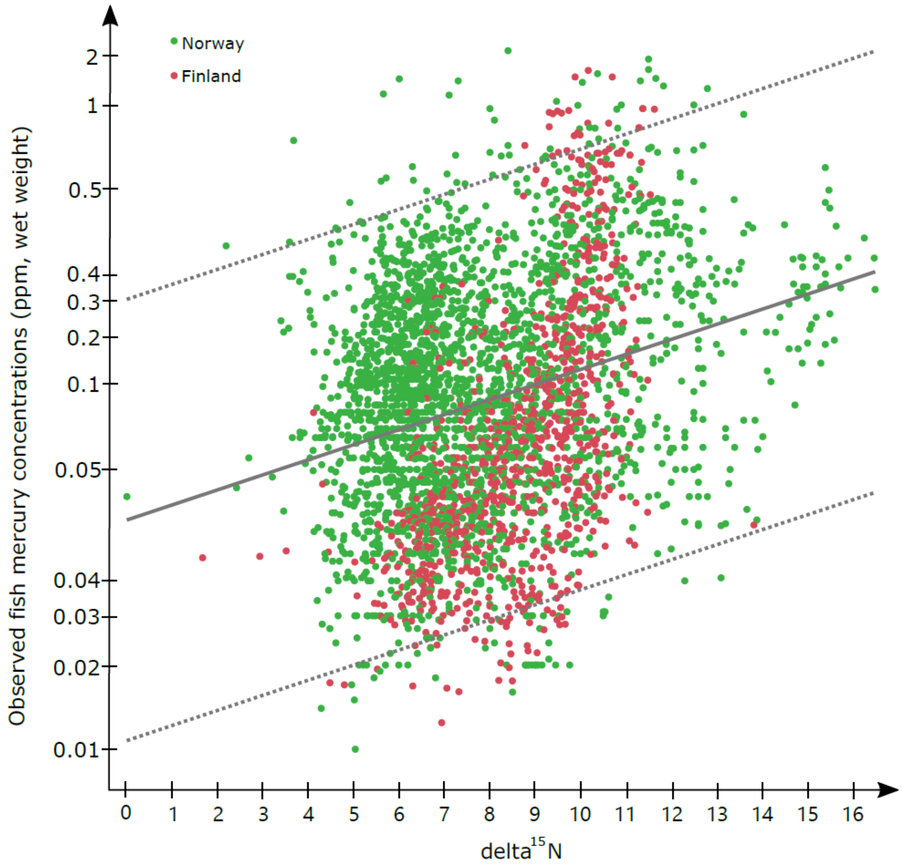


**Figure 2** A scatter plot of observed fish mercury (Hg) concentrations (ppm wet weight, y-axis) versus fish stable nitrogen isotope values (δ15N, x-axis). The linear regression (log Hg = -2.72 + 0.12*δ15N, r2 = 0.08, p < 0.0001) is shown as the grey solid line with dotted 95 % confidence interval lines. Colours represent country of sample origin (Finland = red, and Norway = green).

Organic MeHg is the form of Hg that accumulates in food webs, and at higher trophic levels Hg is often assumed to be present as > 90 % MeHg (Bloom, 1992). Since chemical analysis and determination of the total Hg fractions (organic, inorganic and elemental forms) is more cost-effective than the determination of MeHg, total Hg is mostly reported for Hg in fish. In our database, all the Hg entries are determinations of total Hg. However, for a selection of the entries from Sweden (n = 2 026), MeHg was available in addition to total Hg. The linear relationship between total Hg and MeHg for these entries is very strong (Hg [as total Hg] = 0.18 + 0.82*MeHg, $r^2 = 0.74$, $p < 0.0001$, Figure 10). Mean (± one standard deviation) and median % MeHg of total Hg for these entries are 103 ± 50 % and 98 %, respectively, justifying the use of total Hg concentrations when analysing the spatial and temporal trends for fish Hg concentrations.

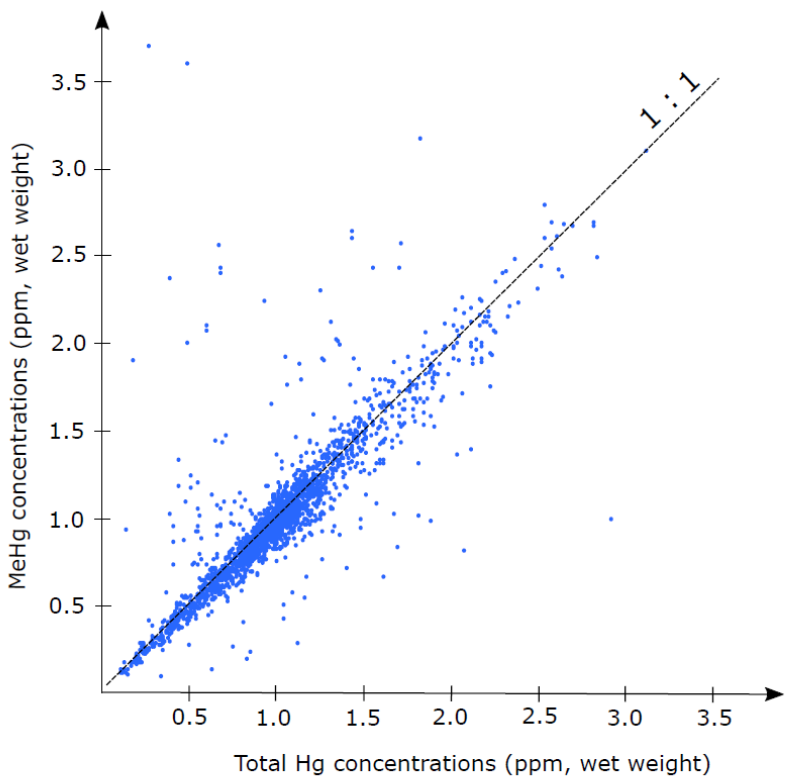


**Figure 10** A scatter plot of measurements of fish mercury (Hg) concentrations determined as total mercury (Total Hg, ppm, wet weight, x-axis) versus concentrations determined as methylmercury (MeHg, ppm, wet weight, y-axis). The linear 1:1 line is shown as the black broken line. All samples are from Sweden.

### 5.1.4 Species- and country/region-specific mercury concentrations

A general description of the database, including data from the observed and adjusted fish Hg concentrations, and the total number of fish and specifics for the five main fish species illustrate the major differences in Hg concentrations between countries and species (Table 4, Table 5 and Table 6). Data are derived as site-specific (based on lake/stream name and coordinates) median concentrations, and the tables show the median and $10^{th}/90^{th}$ percentile of the Hg concentrations for sites in the Fennoscandian region. Site specific concentrations are reported based on Hg concentrations as they were reported in the fish database (Table 4). Correction of the reported data were done in two ways, including *i*) to adjust for variation that is connected to fish size (weight, Table

5), and *ii*) by modelling according to the ICP M&M manual to obtain a value that refers to a standard 1-kg pike (Table 6). The adjustment of data relative to a 1-kg pike is done to adjust for differences in Hg concentrations between fish species within a lake and thereby provide a suitable indicator that relates to human consumption habits. A value that corresponds to a standard 1-kg pike provide a system-specific value that can be compared between lake ecosystems.

The observed fish Hg concentrations in the database (Table 4) revealed the highest median concentration for pike (0.64 ppm). Pike is a fish present at high trophic levels in the food chain and a typical carnivorous fish where high concentrations are expected. Also for weight-adjusted data (Table 5), pike showed the highest median concentrations, and the difference is relatively small between the various normalisation techniques: 0.64 ppm for observed data (Table 4); 0.71 ppm for weight-adjusted data (Table 5); and 0.62 for the standard 1-kg pike (Table 6). The numbers are also similar to the median Hg concentrations of observed pike data from Munthe et al. (2007) and Åkerblom et al. (2014): 0.65 (1965-2004) and 0.66 ppm (1966-2012), respectively. This is not surprising as most of the data is based on the same fish samples, since the larger part of the pike database comes from Swedish monitoring programmes (Figure 3).

Observed median Hg concentrations were lower for Arctic charr (0.10 ppm), perch (0.19 ppm) and brown trout (0.12 ppm) compared to the pike concentrations (Table 4). The same pattern was also found in the studies of Munthe et al. (2007) and Åkerblom et al. (2014), although the observed concentrations differed some. For perch and brown trout, observed concentrations are similar for our study (0.19 ppm and 0.12 ppm) and Åkerblom et al. (2014, 0.18 ppm and 0.16 ppm), while Munthe et al. (2007) report higher concentrations for perch (0.30 ppm) and lower concentrations for trout (0.08 ppm). Both Munthe et al. (2007) and Åkerblom et al. (2014) found similar median concentrations for Arctic charr (0.08 and 0.09 ppm, respectively), to what we found (0.10 ppm).

The lower observed concentrations in populations of brown trout, perch and Arctic charr compared to pike likely reflect the fact that they are normally present at a lower trophic level and not completely carnivorous. However, the database does not contain enough stable isotope measurements to confirm this. A deviation of this is however the observed median concentrations of Hg in roach (0.42 ppm), which is actually higher than those observed for perch, Arctic charr and brown trout. The observed roach Hg concentrations from the present study is also higher than previously reported values (0.21 ppm, Åkerblom et al., 2014). A direct comparison of Hg concentrations between fish species is not valid due to differences between lakes, that have inherently different capacity to biomagnify Hg. For a fish species that is omnivorous the high concentrations in roach are however surprising and could be related to local lake contamination rather than biological or ecological factors related to accumulation of Hg (see detailed discussions below).

For Hg concentrations in Arctic charr, the differences in levels between observed (0.10 ppm) and weight adjusted concentrations (0.13 ppm) are small. This is also the case for roach (0.42 and 0.43 ppm, respectively), while the differences are much large for perch (0.19 and 0.35 ppm, respectively) and brown trout (0.12 and 0.33 ppm). The differences between observed and adjusted concentrations are related to the size distribution of the fish populations, given that the normalisation techniques depend only (weight-adjusted concentrations) or largely (standard 1-kg pike concentrations) on the weight of the fish. A concluding remark is that it is not arbitrary what kind of technique that is chosen, and we will discuss the details and implications of this in sections 4.1.5 and Chapter 6.

For the observed concentrations (Table 4), country-specific patterns are generally similar to the ones described above for the complete database. Hence, pike showed the highest observed concentrations in Sweden, Norway and Finland. For the Kola Peninsula, only perch and pike have been collected, both with median concentrations lower than for the other countries. Compared to the other countries/regions, few samples and lakes are present in the dataset originating from the Kola Peninsula (Figure 2), thus few conclusions can be drawn for this region based on the current database.

Roach occasionally showed unexpectedly high concentrations, an example being the weight-adjusted Hg concentrations from Sweden (0.37 ppm) and Finland (0.46 ppm). These estimates are based on relatively small sample sizes, n = 144 and 681 specimens for Sweden and Finland, respectively, and caution should be made when drawing conclusions from these data.

### 5.1.5 Environmental quality standards for mercury concentrations in fish

A comparison of the approaches to derive lake-specific Hg concentrations show differences in the number of lakes that exceed both limit values set by FAO/WHO for Hg levels in fish used for human consumption (0.5 ppm ww, FAO, 1995) and the EU EQS for the WFD (0.02 ppm ww, European Commission, 2014, Figure 11). Generally, "all" fish caught in Fennoscandia over the last six decades show concentrations above the WFD limit of 0.02 ppm. Of the 54 560 fish samples included in the database for analyses, only 83 specimens had concentrations below 0.02 ppm, and good chemical condition is not met for the vast majority of Fennoscandic lakes with respect to concentrations of Hg. In fact, median concentrations of Hg in fish are higher than the WFD EQS in all but three examined water bodies.

The WFD EQS for biota has two goals for protection that are relevant for Hg, including protection from chemical accumulation in the food chain (i.e. top predators including fish and wildlife), and protection of human health from deleterious effects from consumption of food (e.g. fish). For Hg, the EQS is based on the most stringent *No Observed Effect Concentration* (NOEC) available for MeHg, and a high assessment factor (10) is applied due to the large variation in observed NOEC for MeHg. However, based on the large amount of fish that are shown to have concentrations above the 0.02 ppm limit in the current database, we conclude that the WFD EQS for Hg in biota has limited relevance for assessing the risks of fish Hg exposure in Fennoscandia because it does not differentiate between lakes with higher and lower risks.

In Swedish lakes, the median observed Hg concentration for the complete database is below the FAO/WHO EQS 0.5 ppm limit (0.45 ppm) while, the weight adjusted median concentration (0.65 ppm) and the standard 1-kg pike concentration (0.64 ppm) is above 0.5 ppm (Figure 11). For Finnish lakes, the observed (0.33 ppm) and weight adjusted median concentrations (0.40 ppm) are below the FAO/WHO EQS and only the median standard 1-kg pike concentration is above (0.60 ppm). For Norway, observed (0.24 ppm) and both normalised median concentrations (0.32 ppm and 0.49 ppm, respectively) show median concentrations below the FAO/WHO limit. For Russia, with substantially lower number of fish samples available, all three methods show levels well below the FAO/WHO limit (all < 0.22 ppm). When considering the observed concentrations of Hg from the database, 46, 33 and 20 % of the collected fish shows concentrations above the FAO/WHO limit in Sweden, Finland and Norway, respectively. The same values are 64, 59 and 49 % for standard 1-kg pike concentrations, and 62, 41, and 35 % for weight-adjusted concentrations.

The comparison of Hg concentrations with EQS values demonstrates that methods used for normalisation of fish data affects the outcome. An example is the proportion of fish with concentrations above the 0.5 ppm FAO/WHO limit from the Swedish selection of the database: By using observed concentrations, 46 % is exceeding the limit, while by using weight or standard 1-kg pike adjusted concentrations, the amount rises to 62 and 64 %, respectively. This has implications for the international policy bodies focusing on these limits, and also for local authorities working to develop advisories.

**Table 4** A general description of the database, including the total number of fish included and specifics for the five main fish species. Shown are number of specimens, number of lakes and Hg concentrations. Hg concentrations represent the observed values. Medians and percentiles (10/90) were based on median Hg concentrations values for each lake from the original dataset.

| Observed fish Hg data | | | | | | |
|---|---|---|---|---|---|---|
| **Country/region** | Species | Lake counts (n) | Fish counts (n) | Fish Hg concentrations per lake (ppm, wet weight) | | |
| | | | | **Median** | **10th percentile** | **90th percentile** |
| **Sweden** | Arctic charr | 9 | 688 | 0.140 | 0.029 | 1.300 |
| | Brown trout | 2 | 20 | 0.486 | 0.439 | 0.532 |
| | Perch | 247 | 11564 | 0.245 | 0.084 | 0.792 |
| | Pike | 2172 | 22278 | 0.630 | 0.261 | 1.149 |
| | Roach | 5 | 144 | 0.440 | 0.085 | 0.520 |
| | | | | | | |
| **Finland** | Brown trout | 9 | 160 | 0.077 | 0.026 | 0.308 |
| | Perch | 582 | 8326 | 0.190 | 0.080 | 0.500 |
| | Pike | 260 | 5711 | 0.480 | 0.206 | 1.076 |
| | Roach | 48 | 681 | 0.340 | 0.172 | 0.584 |
| | | | | | | |
| **Norway** | Arctic charr | 6 | 111 | 0.324 | 0.057 | 0.800 |
| | Brown trout | 73 | 1866 | 0.145 | 0.054 | 0.562 |
| | Perch | 62 | 2554 | 0.335 | 0.150 | 0.651 |
| | Pike | 11 | 212 | 0.510 | 0.089 | 1.200 |
| | Roach | 3 | 49 | 0.370 | 0.086 | 0.558 |
| | | | | | | |
| **Kola peninsula** | Perch | 16 | 189 | 0.120 | 0.043 | 0.567 |
| | Pike | 1 | 10 | 0.065 | 0.065 | 0.065 |
| | | | | | | |
| **Summary** | Arctic charr | 15 | 799 | 0.100 | 0.024 | 0.400 |
| | Brown trout | 84 | 2046 | 0.124 | 0.042 | 0.550 |
| | Perch | 905 | 22633 | 0.187 | 0.070 | 0.560 |
| | Pike | 2441 | 28211 | 0.640 | 0.253 | 1.130 |
| | Roach | 56 | 874 | 0.416 | 0.138 | 0.740 |

**Table 5** A general description of the database, including the total number of fish included and specifics for the five main fish species. Shown are number of specimens, number of lakes and Hg concentrations. Hg concentrations are adjusted using weight of each specimen.

| Weight-adjusted fish Hg data | | | | | | |
|---|---|---|---|---|---|---|
| **Country/region** | Species | Lake counts (n) | Fish counts (n) | Fish Hg concentrations per lake (ppm, wet weight) | | |
| | | | | **Median** | **10th percentile** | **90th percentile** |
| **Sweden** | Arctic charr | 9 | 688 | 0.331 | 0.047 | 4.308 |
| | Brown trout | 2 | 20 | 0.084 | 0.076 | 0.092 |
| | Perch | 247 | 11564 | 0.209 | 0.074 | 1.047 |
| | Pike | 2172 | 22278 | 0.671 | 0.281 | 1.325 |
| | Roach | 5 | 144 | 0.370 | 0.182 | 1.072 |
| | | | | | | |
| **Finland** | Brown trout | 9 | 160 | 0.179 | 0.074 | 0.475 |
| | Perch | 582 | 8326 | 0.252 | 0.082 | 1.001 |
| | Pike | 260 | 5711 | 0.737 | 0.268 | 2.253 |
| | Roach | 48 | 681 | 0.459 | 0.181 | 1.276 |
| | | | | | | |
| **Norway** | Arctic charr | 6 | 111 | 0.282 | 0.025 | 1.463 |
| | Brown trout | 73 | 1866 | 0.367 | 0.116 | 3.121 |
| | Perch | 62 | 2554 | 0.282 | 0.091 | 1.531 |
| | Pike | 11 | 212 | 0.636 | 0.182 | 1.882 |
| | Roach | 3 | 49 | 0.228 | 0.149 | 0.889 |
| | | | | | | |
| **Kola peninsula** | Perch | 16 | 189 | 0.068 | 0.015 | 0.219 |
| | Pike | 1 | 10 | 0.164 | 0.164 | 0.164 |
| | | | | | | |
| **Summary** | Arctic charr | 15 | 799 | 0.131 | 0.032 | 0.455 |
| | Brown trout | 84 | 2046 | 0.333 | 0.082 | 2.299 |
| | Perch | 905 | 22633 | 0.347 | 0.094 | 1.697 |
| | Pike | 2441 | 28211 | 0.714 | 0.272 | 1.603 |
| | Roach | 56 | 874 | 0.430 | 0.164 | 1.217 |

**Table 6** A general description of the database, including the total number of fish included and specifics for adjustment of data to a standard 1-kg pike. Shown are number of specimens, number of lakes and Hg concentrations. Hg concentrations are adjusted according to the ICP M&M manual.

| Fish Hg data adjusted to 1-kg pike | | | | | | |
|---|---|---|---|---|---|---|
| **Country/region** | Species | Lake counts (n) | Fish counts (n) | Fish Hg concentrations per lake (ppm, wet weight) | | |
| | | | | **Median** | **10th percentile** | **90th percentile** |
| **Sweden** | 1-kg pike | 2292 | 34691 | 0.64 | 0.25 | 1.32 |
| | | | | | | |
| **Finland** | 1-kg pike | 699 | 14905 | 0.60 | 0.21 | 1.61 |
| | | | | | | |
| **Norway** | 1-kg pike | 126 | 4765 | 0.49 | 0.13 | 1.32 |
| | | | | | | |
| **Kola peninsula** | 1-kg pike | 16 | 199 | 0.22 | 0.09 | 0.67 |
| | | | | | | |
| **Summary** | 1-kg pike | 3133 | 54560 | 0.62 | 0.22 | 1.38 |

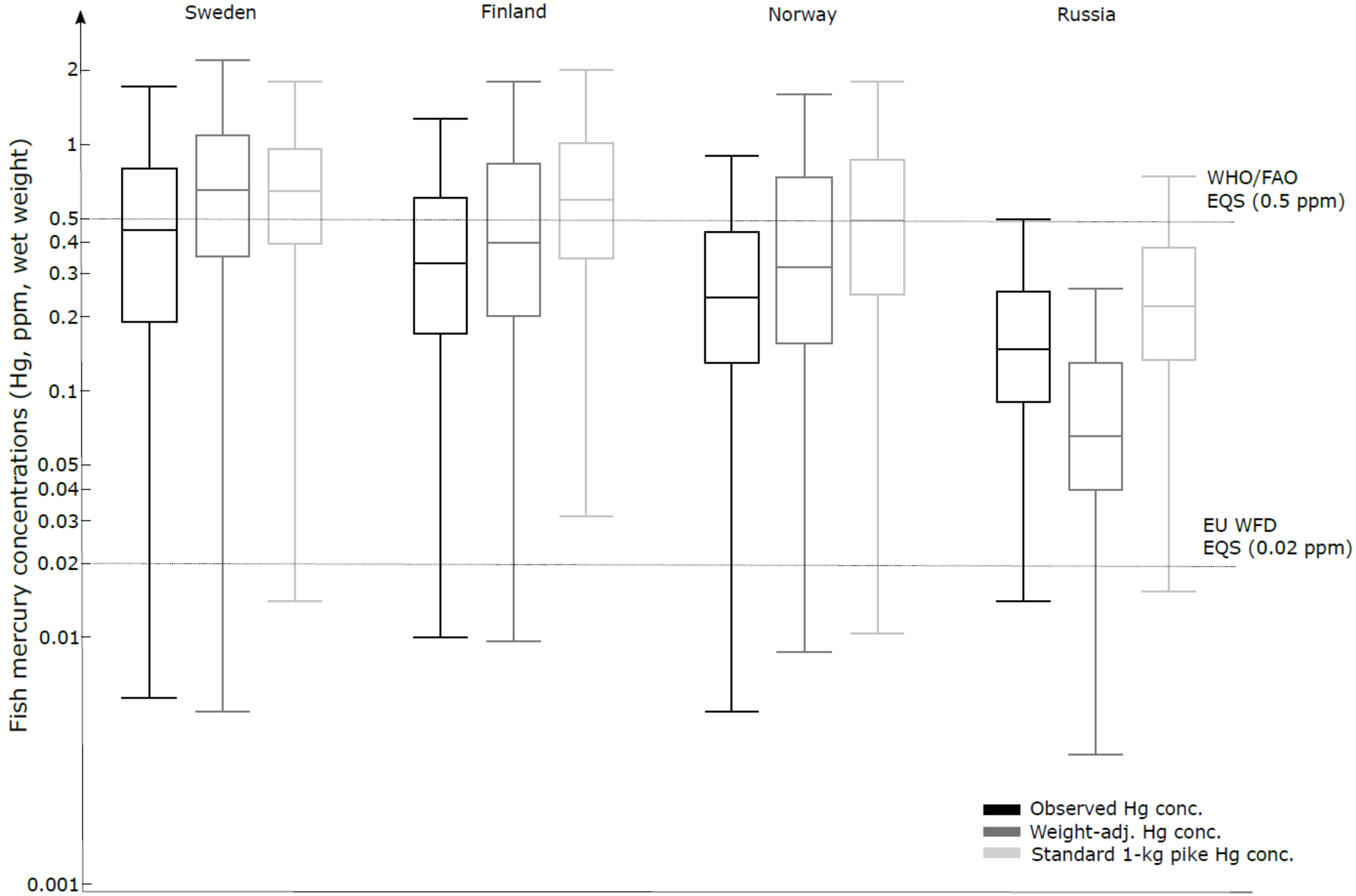


**Figure 11** Histogram displaying fish Hg concentrations (ppm ww) in lakes from Fennoscandia. Shown are observed, weight-adjusted and standard 1-kg pike Hg concentrations. Horizontal dashed lines represent environmental quality standards (EQS) set by the WHO/FAO to protect human health and the EU WFD to protect ecological health in aquatic ecosystems. Distributions of Hg concentrations are described with minimum and maximum values (bars), 25 and 75 percentiles (box), and median concentrations (horizontal line).

## 5.2 Spatial patterns and temporal trends for the complete database

### 5.2.1 Spatial patterns of mercury in fish from Fennoscandia

Fish Hg concentrations across Fennoscandia revealed a south-to-north belt of relatively high concentrations from south-west Sweden toward the north-eastern parts of the Bothnian Sea (red colours on the map, concentrations above 1 ppm, Figure 12). As the geographical location map (Figure 5), the spatial Hg concentration map is reflecting patterns of species occurrence (i.e. consumption preference), fishing interests, and monitoring traditions (related to a range of factors including suspected point sources of Hg). As pointed out and discussed by Munthe et al. (2007), this is most likely a reflection of historical emissions of Hg to air and water from local industry. However, Munthe et al. (2007) did not take any measures to avoid the influence from these potentially locally contaminated lakes on the overall conclusions related to Hg concentrations in fish. We have, in the present work, made efforts to remove these lakes, in order to assess lakes with long-range transported Hg as the main source to Hg (see section 4.3.1).

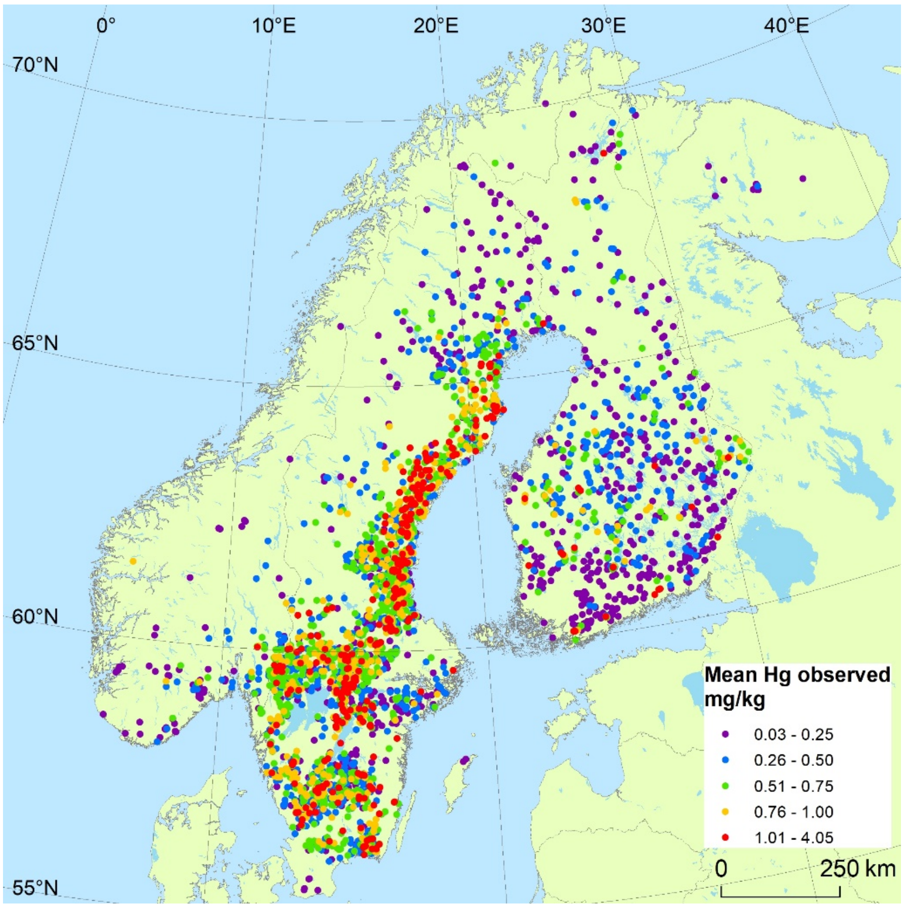


**Figure 12** Median observed fish Hg concentrations per lake in the complete data set (1965-2015). Measurements are grouped based on concentration levels and include all the five main fish species, Arctic charr, brown trout, perch, pike and roach.

The ecosystem characteristics typically shown to be important for fish Hg concentrations can be grouped into climatic, chemical, biological or physical factors. One important chemical factor is OM, as this acts as a transport vector for Hg species from catchment soils to adjacent lakes. In other words, Hg stored and accumulated in catchment soils following historical atmospheric deposition will be leached out and transported to lakes and rivers bound to organic carbon given the strong Hg affiliation for sulphur sites in OM. The strong relationships between aqueous Hg and TOC is often shown on a spatial scale (Braaten et al., 2014a), but the processes and mechanisms that follow TOC in aquatic ecosystems are complex and difficult to assess on a temporal scale.

In 1995, there were national lake surveys in several northern European countries, initiated by the environment agencies and authorities in Norway, Sweden, and Finland. In Fennoscandia, nearly 5000 lakes were sampled. We have taken TOC spot sample data from this survey and spatially-interpolated it using inverse distance weighting (IDW), aggregating results to a 25km grid (Figure 15). The grid is consistent with other spatially-distributed gridded datasets such as nitrogen and sulphur deposition and climate data, which we hope will be of future use in analysing the current fish database. The map provides an averaged snapshot of TOC concentrations across the whole of Fennoscandia in 1995.

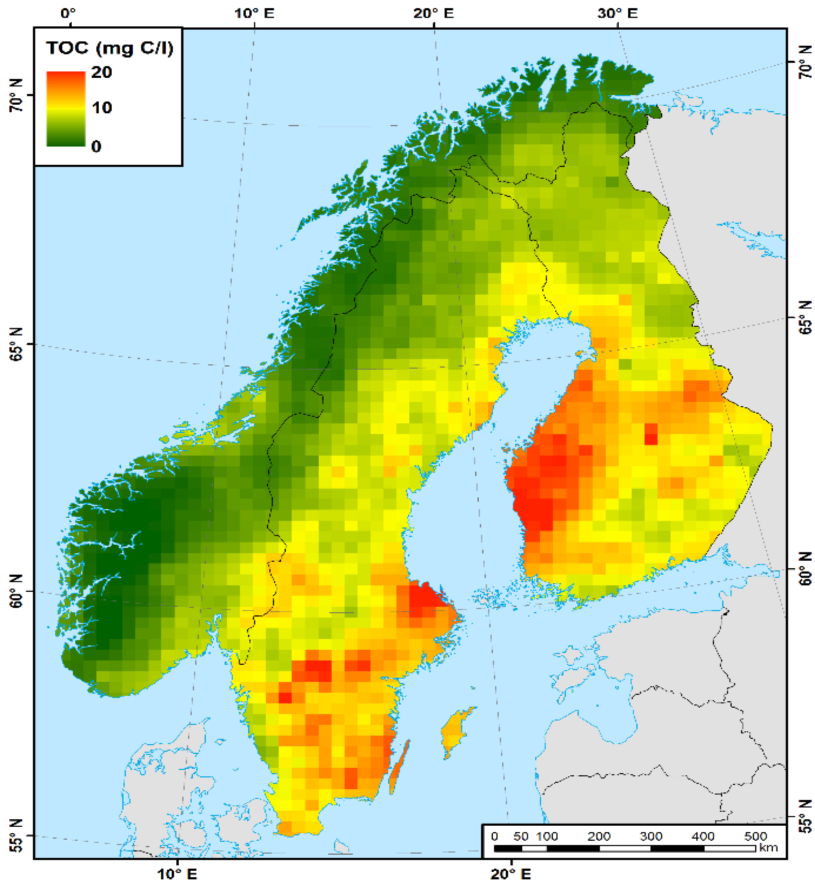


**Figure 13** A spatially-interpolated map of total organic carbon (TOC) concentrations for Norway, Sweden, and Finland. Results are aggregated to 25 km grids.

By comparing the spatial map for fish Hg concentrations (Figure 14) and the interpolated TOC map (Figure 13), it is evident that, as for surface sediment Hg concentrations (Figure 15), TOC cannot fully explain the Hg variation seen throughout Fennoscandia. However, it does show, particularly for Norwegian and Finnish lakes, that relatively high Hg concentrations tend to coincide with dark humic lakes.

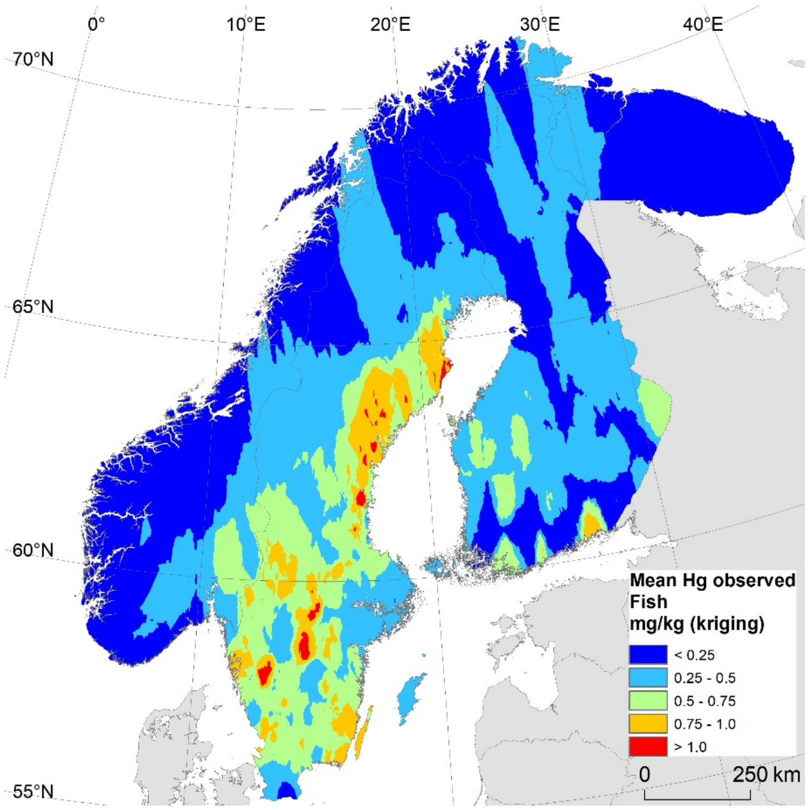


**Figure 14** Interpolated (by kriging) concentrations of Hg in fish for Fennoscandia based on the complete data set (1965-2015). Measurements are grouped based on concentration levels and include all the five main fish species, Arctic charr, brown trout, perch, pike, and roach.

In combination, our spatial data analysis confirms what has previously been shown, particularly by Munthe et al. (2007), that the Hg concentration pattern is a combination of historical atmospheric Hg deposition, local point sources and ecosystem characteristics. A very important part of this is the role of OM, which does not only influence the catchment-to-lake transport of Hg, but also the in-lake cycling of Hg. However, the in-lake cycling of Hg, including methylation and de-methylation processes of organic Hg, is beyond the scope of the present report.

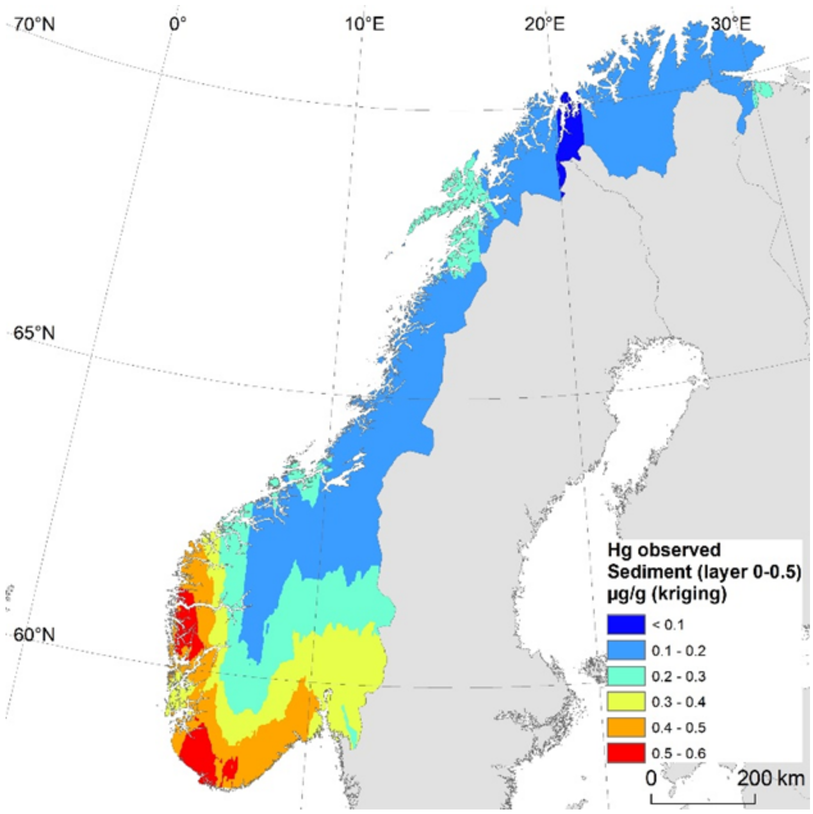


**Figure 15** Surface sediment (0-0.5 cm) Hg concentrations interpolated (by kriging), based on measurements of sediment total Hg in Norway during 2006-2008 (Skjelkvåle et al., 2008).

### 5.2.2 Temporal trends of mercury in fish from Fennoscandia

Temporal trend analyses for the complete database (median Hg concentration for each year) covering the entire study period and including all five main fish species for observed Hg concentrations (Figure 16), weight-adjusted concentrations (Figure 17) and concentrations adjusted to a standard 1-kg pike (Figure 18) showed that direction of the trends are dependent on both fish species and methods for data normalisation.

The inter-annual variation in fish Hg concentration is large (exemplified by the extent of year-to-year variations seen for all species in the fish Hg database, Figures 16-18). Correlations between time (year) and fish Hg concentrations show decreasing concentrations for perch ($r=-0.45$, $p<0.0001$), pike ($r=-0.12$, $p<0.0001$) and roach ($r=-0.21$, $p=0.03$). For Arctic charr ($r=-0.07$, $p=0.55$) and brown trout ($r=-0.09$, $p=0.36$), there is no significant temporal trend in Hg concentrations. Weight-adjustment of fish Hg concentrations (Figure 17) did not change the temporal trends from trends seen in observed fish Hg concentration for perch ($r=-0.04$, $p=0.05$) and pike ($r=-0.08$, $p<0.0001$), while the direction for roach ($r=-0.17$, $p=0.09$), Arctic charr ($r=0.21$, $p=0.06$) and brown trout ($r=0.28$, $p=0.003$) differed. For the standard 1-kg pike (Figure 18), the data show a decreasing temporal trend ($r=-0.21$, $p<0.0001$).

When the data are broken down to mean concentrations per decade, the decrease in fish Hg concentration is substantial when all fish species are grouped together (Table 7). All three techniques for presenting Hg concentrations show a decrease between 1965-75 and 2006-15, with the decrease being largest for the observed concentrations: the mean 2006-15 concentration (0.27 ppm) is only 36

% of the level in 1965-75 (0.74 ppm). For weight adjusted (77 %) and standard 1 kg pike data (74 %), the relative levels in 2006-15 are higher (i.e. the decrease in concentration not as substantial).

Pike Hg concentrations reveal a temporal trend that is remarkably similar between the three techniques of normalisation, where Hg concentrations decreased every decade between 1965-75 and 2006-15 (Table 7). For Arctic charr, brown trout, perch and roach, the variation between decades and techniques of normalisation is much less consistent. As an example, perch concentrations have decreased between 1965-75 and 2006-15 for both observed and weight adjusted concentrations, but the decrease is much smaller for weight-adjusted concentrations (31 %), compared to observed concentrations (64 %). Weight-adjusted concentrations for perch also show increased levels for 1996-2005 (120 % of 1965-75 levels), something which is not observed when considering the observed data (35 % of 1965-75 levels).

Although the overall trend for all fish species in combination is a decrease in Hg concentrations (Table 7), the pattern is not very clear when it comes to individual fish species and different techniques for data normalisation (Figures 16-18). Important aspects to consider with respect to these trends would include 1) the source of Hg contamination (air pollution versus local industry), 2) differences between fish species (predatory and non-predatory species), and 3) differences between ecosystems (e.g. catchment, lake and climate characteristics). The effects of these aspects on spatial and temporal trends are presented in more detail in chapter 6.

**Table 7** Mean concentrations of mercury for the different fish species and for the complete database per decade for the three different techniques of normalisation. Shown are mean values ± one standard deviation and the relative level of each decadal value compared to the concentration in 1965-75.

| Decade/ Fish specie | Observed | | Weight-adjusted | | Standard 1-kg pike | |
|---|---|---|---|---|---|---|
| | Hg (ppm) | Relative to 1965-75 (%) | Hg (ppm) | Relative to 1965-75 (%) | Hg (ppm) | Relative to 1965-75 (%) |
| **Arctic charr** | | | | | | |
| **1965-75** | 0.48 ± 0.15 | 100 | 0.21 ± 0.07 | 100 | - | - |
| **1976-85** | 0.18 ± 0.15 | 38 | 0.12 ± 0.07 | 57 | - | - |
| **1986-95** | 0.07 ± 0.10 | 15 | 0.11 ± 0.10 | 52 | - | - |
| **1996-05** | 0.16 ± 0.22 | 33 | 0.17 ± 0.20 | 81 | - | - |
| **2006-15** | 0.16 ± 0.20 | 33 | 0.51 ± 1.08 | 243 | - | - |
| **Brown trout** | | | | | | |
| **1965-75** | 0.13 ± 0.08 | 100 | 0.64 ± 0.49 | 100 | - | - |
| **1976-85** | 0.45 ± 0.26 | 346 | 0.25 ± 0.19 | 39 | - | - |
| **1986-95** | 0.10 ± 0.06 | 77 | 0.49 ± 0.43 | 77 | - | - |
| **1996-05** | 0.27 ± 0.40 | 208 | 0.34 ± 0.37 | 53 | - | - |
| **2006-15** | 0.19 ± 0.22 | 146 | 2.19 ± 3.96 | 342 | - | - |
| **Perch** | | | | | | |
| **1965-75** | 0.64 ± 0.49 | 100 | 0.88 ± 0.73 | 100 | - | - |
| **1976-85** | 0.62 ± 0.43 | 97 | 0.56 ± 1.72 | 64 | - | - |
| **1986-95** | 0.33 ± 0.27 | 52 | 0.52 ± 1.90 | 59 | - | - |
| **1996-05** | 0.23 ± 0.21 | 35 | 1.06 ± 1.57 | 120 | - | - |
| **2006-15** | 0.24 ± 0.23 | 36 | 0.61 ± 0.82 | 69 | - | - |
| **Pike** | | | | | | |
| **1965-75** | 0.82 ± 0.62 | 100 | 1.02 ± 0.85 | 100 | 0.88 ± 0.59 | 100 |
| **1976-85** | 0.77 ± 0.44 | 94 | 0.98 ± 2.05 | 96 | 0.82 ± 0.46 | 93 |
| **1986-95** | 0.71 ± 0.42 | 87 | 0.87 ± 0.77 | 85 | 0.77 ± 0.46 | 88 |
| **1996-05** | 0.65 ± 0.55 | 79 | 0.79 ± 1.06 | 77 | 0.67 ± 0.55 | 76 |
| **2006-15** | 0.63 ± 0.51 | 77 | 0.77 ± 0.85 | 75 | 0.65 ± 0.45 | 74 |
| **Roach** | | | | | | |
| **1965-75** | 0.51 ± 0.24 | 100 | 0.71 ± 0.56 | 100 | - | - |
| **1976-85** | 0.43 ± 0.26 | 84 | 0.59 ± 0.53 | 83 | - | - |
| **1986-95** | 0.50 ± 0.24 | 98 | 0.40 ± 0.22 | 56 | - | - |
| **1996-05** | 0.08 ± 0.03 | 16 | 0.19 ± 0.09 | 27 | - | - |
| **2006-15** | 0.27 ± 0.24 | 53 | 0.51 ± 0.53 | 72 | - | - |
| **All species summarised** | | | | | | |
| **1965-75** | 0.74 ± 0.57 | 100 | 0.94 ± 0.80 | 100 | - | - |
| **1976-85** | 0.74 ± 0.44 | 100 | 0.92 ± 1.98 | 98 | - | - |
| **1986-95** | 0.63 ± 0.43 | 85 | 0.80 ± 1.05 | 85 | - | - |
| **1996-05** | 0.40 ± 0.45 | 54 | 0.88 ± 1.32 | 94 | - | - |
| **2006-15** | 0.27 ± 0.30 | 36 | 0.72 ± 1.32 | 77 | - | - |

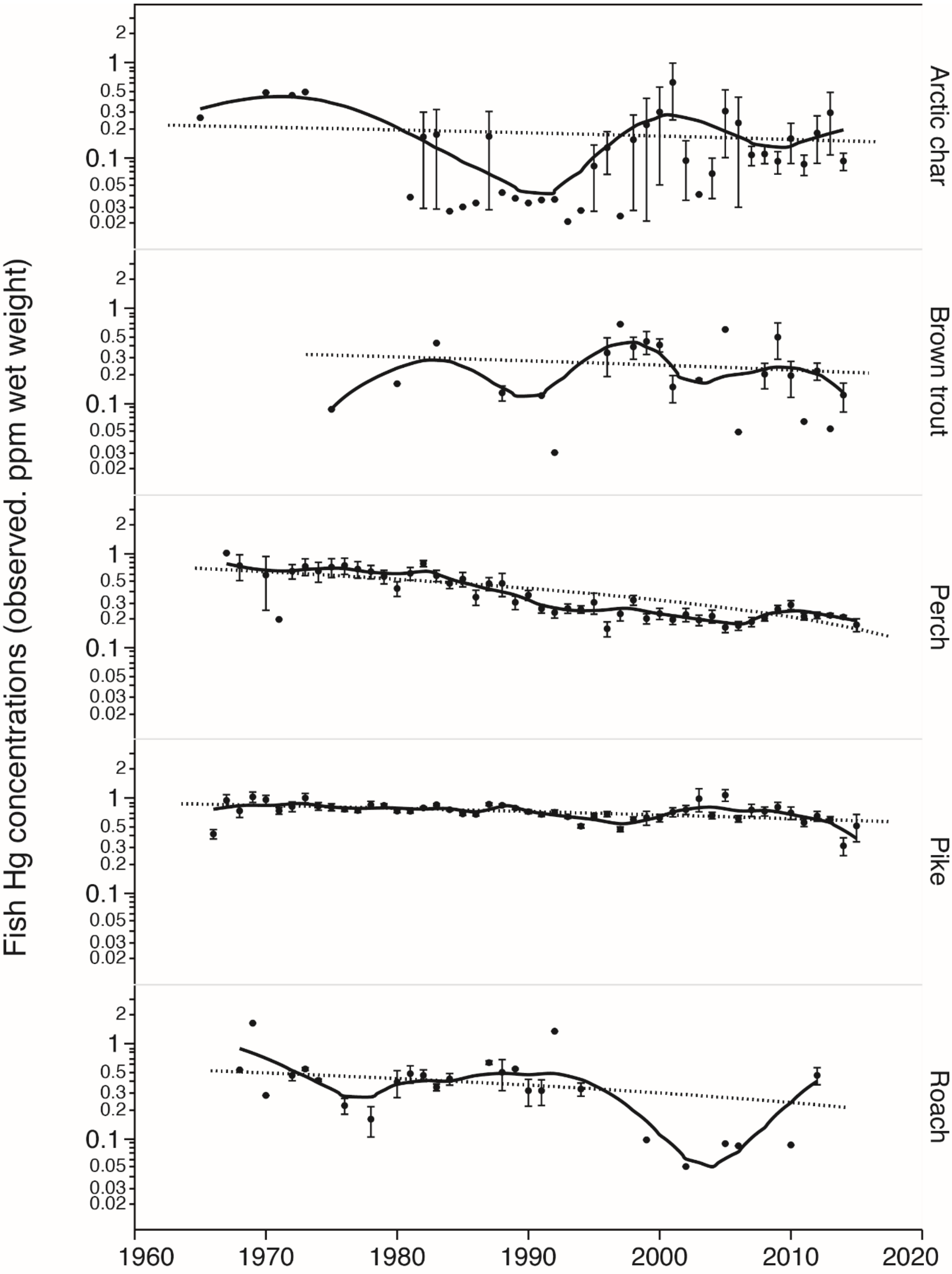


**Figure 16** Observed fish Hg concentrations for (from top to bottom, with direction and significance of temporal trend) Arctic charr (r=-0.07; p=0.55), brown trout (r=-0.09; p=0.36), perch (r=-0.45; p<0.0001), pike (r=-0.12; p<0.0001) and roach (r=-0.21; p=0.03). Each circle represents mean Hg concentration per year. N = 6 368 values included. The solid line represents a smoothed linear function and the dotted line represent a linear regression. Error bars represent ± one standard deviation. Data from all lakes (including lakes with local sources of Hg) from Fennoscandia are included.

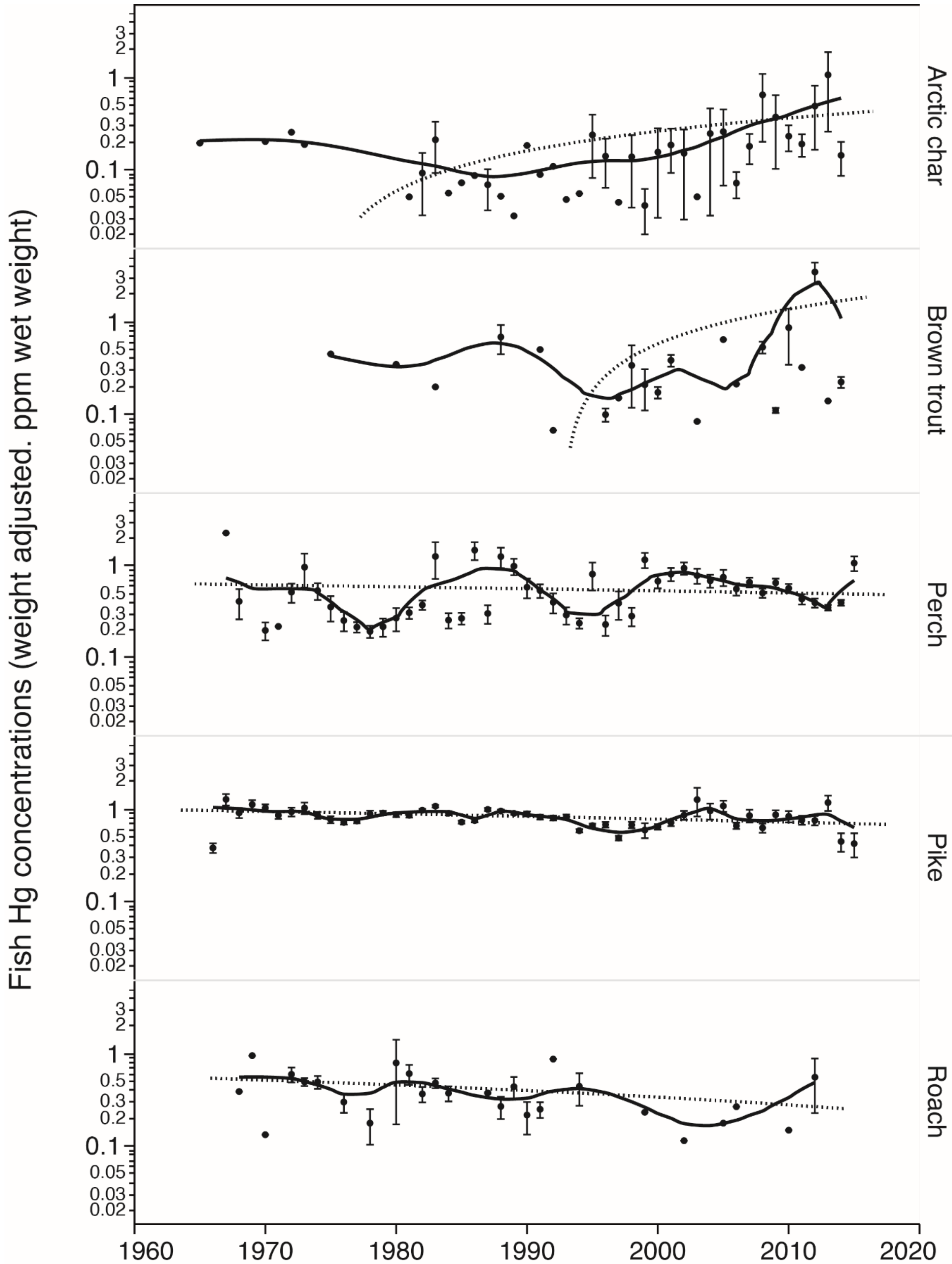


**Figure 17** Weight-adjusted fish Hg concentrations for (from top to bottom, with direction and significance of temporal trend) Arctic charr (r=0.21; p=0.06), brown trout (r=0.28; p=0.003), perch (r=-0.04; p=0.05), pike (r=-0.08; p<0.0001) and roach (r=-0.17; p=0.09). Each circle represents mean Hg concentration per year. N = 6 368 values included. The solid line represents a smoothed linear function and the dotted line represent a linear regression. Error bars represent ± one standard deviation. Data from all lakes (including lakes with local sources of Hg) from Fennoscandia are included.

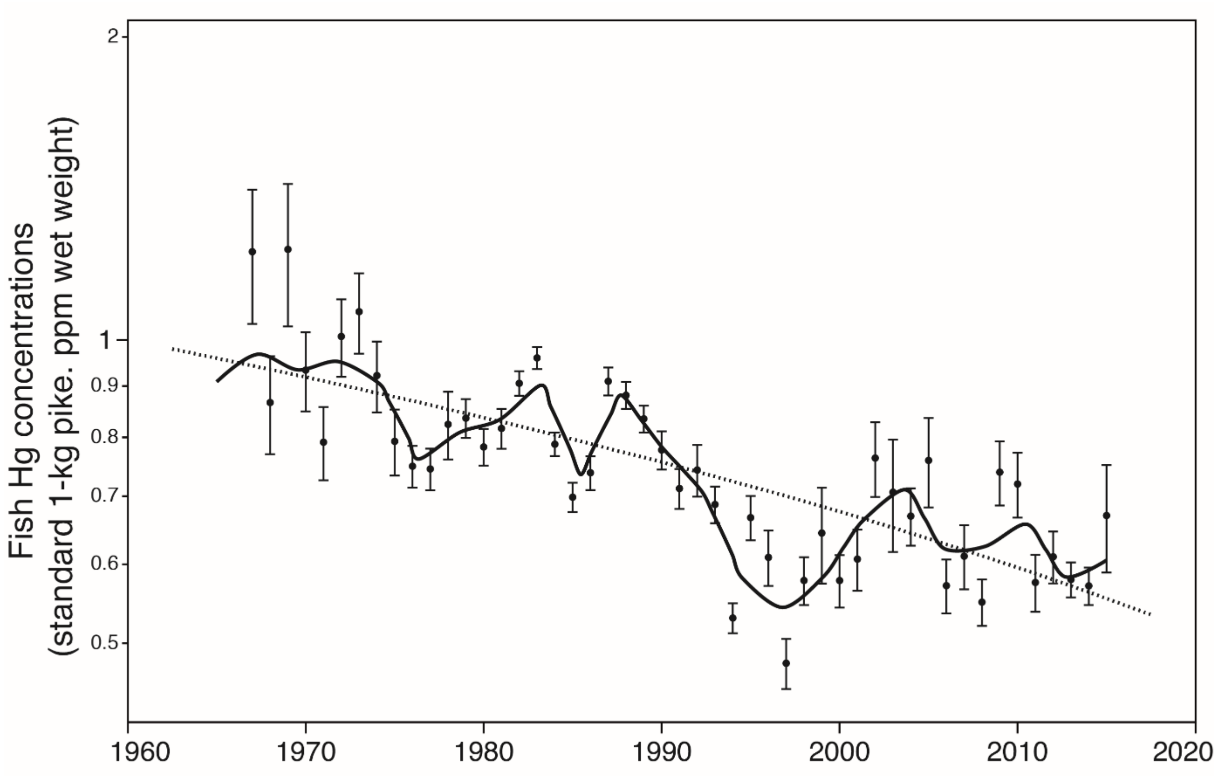


**Figure 18** Standard 1-kg pike Hg concentrations (r=-0.21; p<0.0001). Each circle represents mean Hg concentration per year. The solid line represents a smoothed linear function and the dotted line represent a linear regression. Error bars represent ± one standard deviation. Data from all lakes (including lakes with local sources of Hg) from Fennoscandia are included.

## 5.3 Spatial patterns and temporal trends for lakes mainly influenced by atmospheric mercury deposition

### 5.3.1 Air pollution versus locally contaminated lakes – a preliminary analysis

Lakes that were classified as being subject mainly to sources of Hg from atmospheric deposition show remarkable differences in the availability of data between countries, as seen in reported values per country (Table 8, Figure 19). As an example, Sweden has 23 lakes and 1 874 fish counts while Finland has 754 lakes and 11 673 fish counts. The Swedish selection of lakes within group “1” (only air pollution lakes) was limited even further and only included lakes that were sampled on more than 5 occasions while both Finland and Norway (111 lakes and 3 468 fish counts) covers lakes with no such limitation. The limitation in the Swedish database was mainly a consequence of the large amount of data recorded from locally contaminated lakes in national monitoring programmes, and the difficulties related to identifying these lakes (see section 4.1.2 for details).

From the subset of lakes that are classified as only being influenced from air pollution and atmospheric deposition of Hg (n = 17 015 specimens of fish), we have selected lakes that have been sampled on at least 5 separate years throughout the study period (1965-2015). Data from these lakes were further used to investigate temporal trends that relate more closely to changes in loads of atmospherically deposited Hg (Figure 19). By removing lakes that are influenced by local contamination sources, most of the lakes with elevated median Hg concentrations (> 0.6 ppm) are removed. However, as is evident from the spatial patterns, using adjusted Hg concentrations (by weight or according to the ICP M&M manual) provides several lakes with high Hg concentrations in fish.

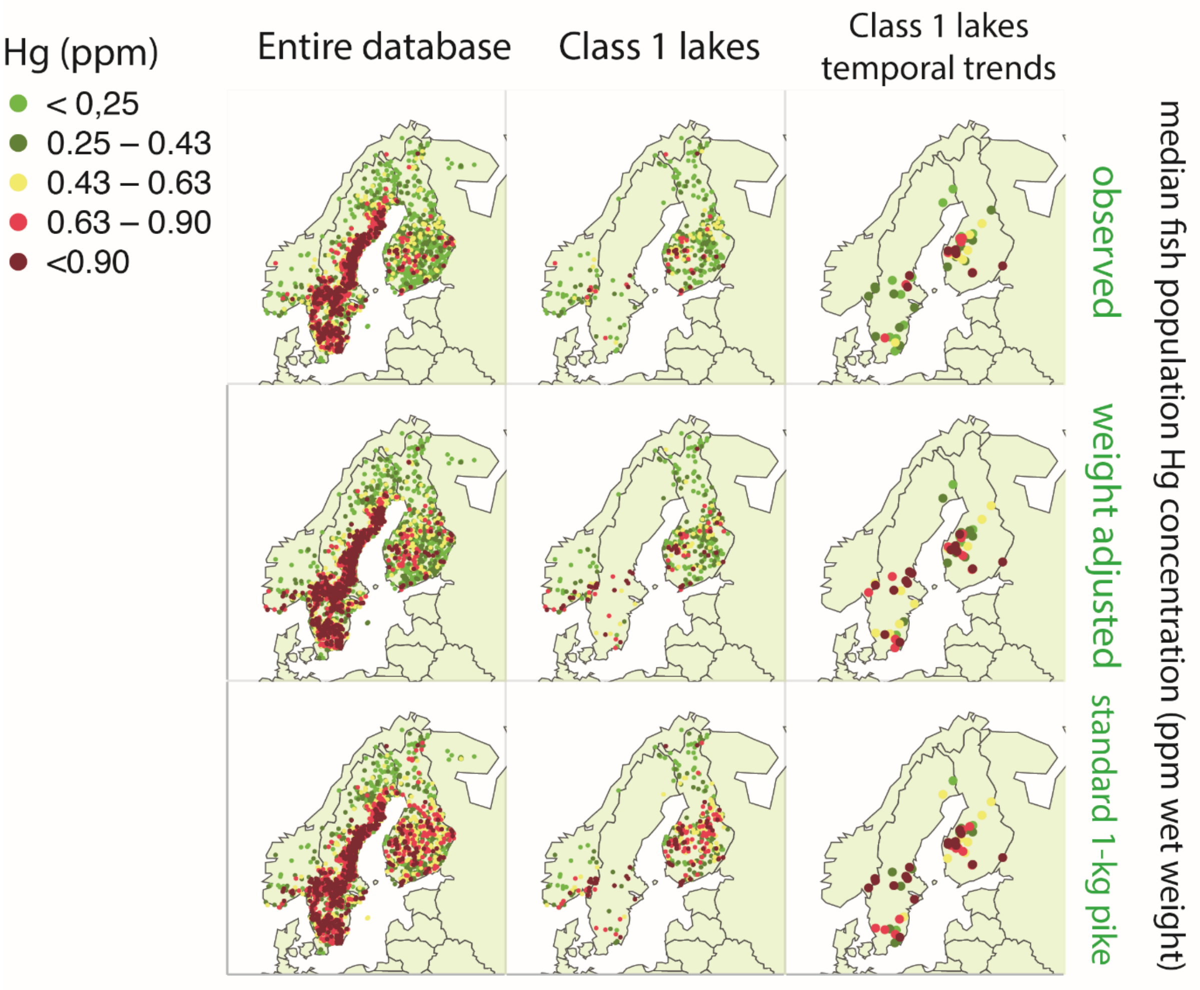


**Figure 19** Maps with lakes (dots) across Fennoscandia from the entire database (left panels), lakes subject to sources from atmospherically derived Hg (air pollution lakes, central panels, Class "1" lakes), and lakes that have been used for temporal trend analysis and being resampled at least 5 years throughout the study period (right panels, Class "1" lakes temporal trends). Shown are observed concentrations (top row), weight adjusted concentrations (central row) and standard 1 kg pike concentrations (lower row).

For these lakes normalisation of observed fish Hg data to fish size was carried out through weight normalisation or normalisation to a standard 1-kg pike for the different species (Table 8). Perch and pike were represented in the largest number of lakes (561 and 226, respectively) followed by brown trout (58 lakes), roach (41 lakes) and Arctic charr (2 lakes in Norway). Pike have the highest observed range in country median Hg concentrations, ranging from 0.48 ppm (Norway) to 0.88 ppm (Sweden). In Norway, roach had the highest lake median concentration (0.56 ppm) but the data come from one lake and are thus not directly comparable to a larger set of lakes. Lake median Hg concentrations in perch ranged between 0.20 ppm (Sweden) and 0.35 ppm (Norway). The normalisation to weight and to a 1-kg pike increased the lake median values for each fish species in all countries, and especially the 90$^{th}$ percentile of lake medians increases substantially. This effect is mainly ascribed to a skewness in the fish sizes between the selected lakes.

Differences in observed Hg concentrations between the two groups of lakes (group “1” and “3”) indicate that lakes affected predominantly by atmospheric deposition of Hg (0.25 ± 0.27 ppm, n = 703) were lower compared to lakes also affected by local pollution sources (0.55 ± 0.38 ppm, n = 167) (ANOVA: F-ratio=264(3504), $p$<0.0001, $r^2$=0.13, left panel, Figure 20, Table 8). A large degree of the between-lake variation in fish Hg levels is likely due to differences in catchment and food web characteristics, as well as by climate and external factors (Sorensen et al., 1990). This variation complicates efforts to discern the effects of atmospheric deposition of Hg on subsequent Hg concentrations in fish.

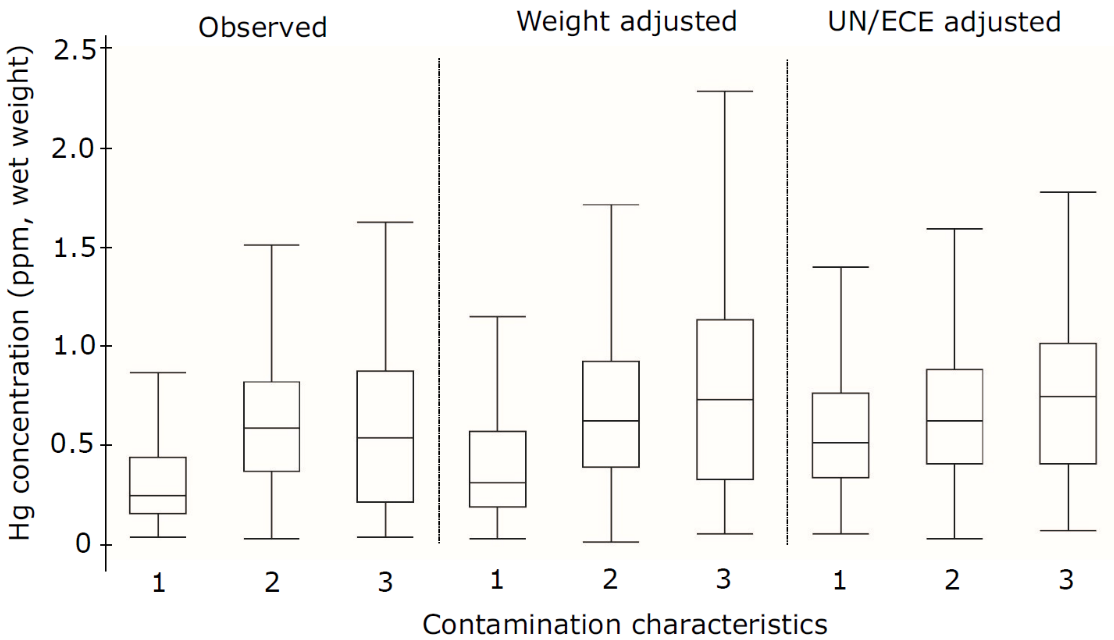


**Figure 20** Box-plots of lake medians of fish Hg concentrations from lakes only affected by air pollution (1), lakes affected by local polluting industry (3) and lakes where pollution sources are “uncertain” (2).

The regression coefficient ($b$) between fish size (weight) and fish Hg concentration is an indicator of the degree to which fish accumulate Hg as they grow (Figure 8). For the lakes influenced by atmospheric deposition of Hg, the coefficient varied considerably between countries and fish species (Figure 21). The strongest relation between fish size and Hg levels were seen in brown trout in Finnish lakes ($b$=0.70, $r^2$=0.49). For Norwegian lakes, the variation in bioaccumulation efficiency between fish species was low (brown trout $b$=0.21; perch: $b$=0.27; pike: $b$=0.52; roach: $b$=0.48), but a lot of the variation in fish Hg levels remained unexplained (i.e. low $r^2$ values) for brown trout ($r^2$=0.08) and pike ($r^2$=0.07) compared to perch ($r^2$=0.21) and roach ($r^2$=0.88). Hg concentrations in roach in Finnish lakes were not significantly related to fish weight ($b$=-0.01, $r^2$=0.0001). In Swedish lakes, the variation in fish Hg levels was only explained by weight to a minor extent in perch ($r^2$=0.01) and pike ($r^2$=0.18). Perch also showed a low bioaccumulation efficiency in Swedish lakes ($b$=0.06) while it was higher in pike ($b$=0.41). The bioaccumulation efficiency of Hg in roach was high ($b$=0.79, $r^2$=0.64). There are obvious differences in the patterns of bioaccumulation between species and countries and attention to this variation needs to be taken in the evaluation of temporal and spatial patterns of fish Hg data across the Fennoscandian shield. As an example, future monitoring would benefit from adding additional parameters than what is common today, especially $\delta^{15}N$.

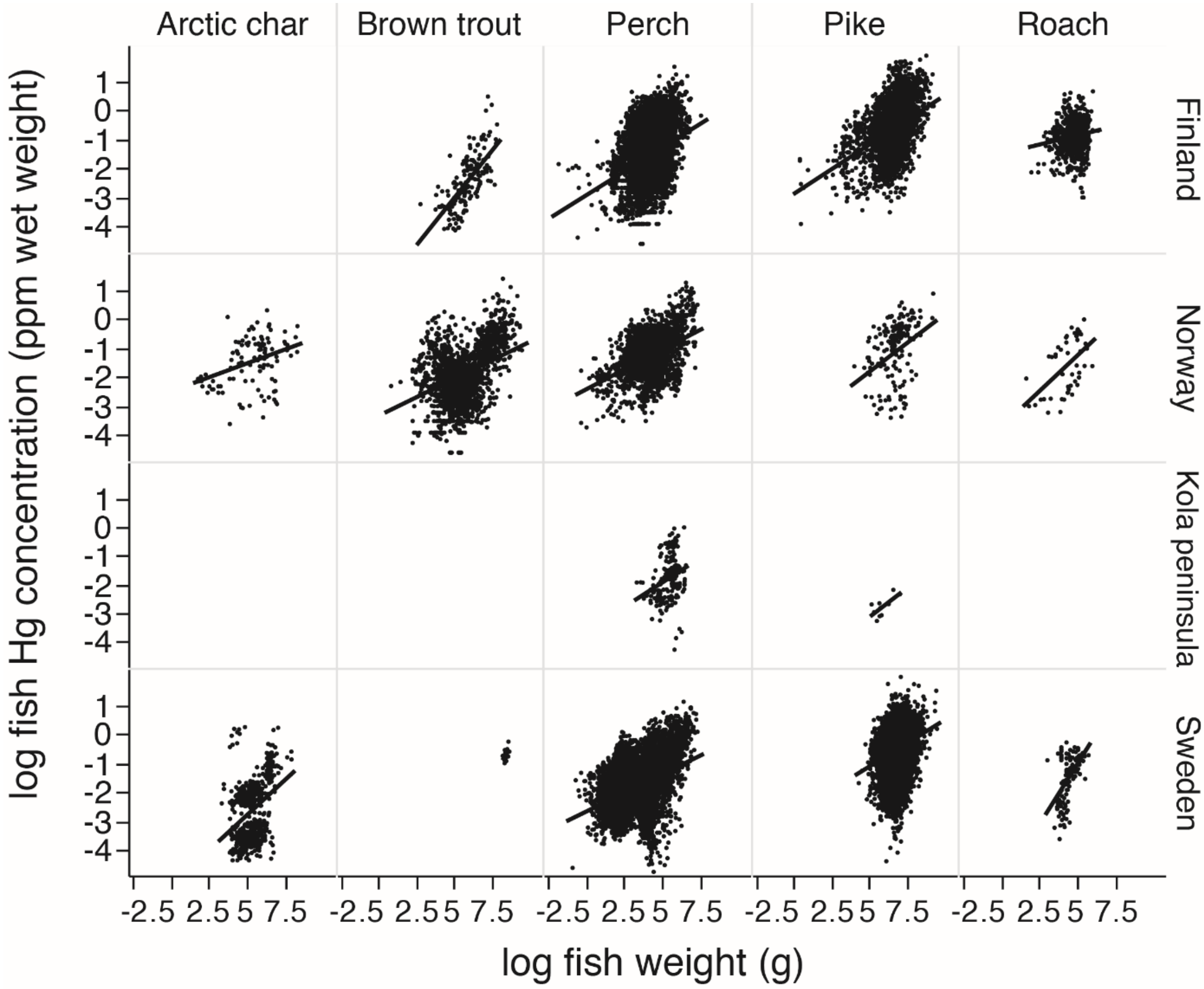


**Figure 21** Regression for the dependence of fish Hg levels (log fish Hg ppm ww) on fish size (log weight g) for different fish species (arctic char, brown trout, perch, pike, and roach) between countries across Fennoscandia.

For the weight-adjusted concentrations, lakes classified as being subject to sources of loads of Hg from local sources had higher fish Hg concentrations (0.59 ± 0.63 ppm) than lakes subject to atmospheric deposition of Hg (0.31 ± 0.58 ppm, Figure 20). For the standard 1-kg pike the differences are not as large between the different lake groups with mean concentrations for lakes subject to loads of Hg from local sources (0.61 ± 0.39 ppm) and lakes subject to atmospheric deposition of Hg (0.50 ± 0.48 ppm) having more similar levels. Again, this underlines the importance of being careful when choosing the method for presenting concentrations of Hg in fish, i.e. how data are "normalised".

**Table 8** Description of the subset of lakes that were classified as being predominantly affected by long-range atmospheric inputs of Hg, including the total number of fish and specifics for the five main groups of fish species with lake median Hg concentrations for observed Hg levels, weight-adjusted Hg levels and Hg levels adjusted according to the ICP M&M manual.

| Country/region | Species | Lake counts (n) | Fish counts (n) | Fish Hg concentrations per lake (ppm, wet weight) | | | | | | | | |
|---|---|---|---|---|---|---|---|---|---|---|---|---|
| | | | | Observed Hg | | | Weight-adjusted Hg | | | Standard 1-kg pike | | |
| **Sweden*** | | | | median | 10th | 90th | median | 10th | 90th | median | 10th | 90th |
| | Arctic char | - | - | - | - | - | - | - | - | - | - | - |
| | Brown trout | - | - | - | - | - | - | - | - | - | - | - |
| | Perch | 13 | 1473 | 0.20 | 0.08 | 0.39 | 0.56 | 0.12 | 3.27 | 0.57 | 0.24 | 1.72 |
| | Pike | 9 | 344 | 0.88 | 0.50 | 1.40 | 1.08 | 0.27 | 1.68 | 0.97 | 0.37 | 1.51 |
| | Roach | 1 | 57 | 0.28 | 0.28 | 0.28 | 0.37 | 0.37 | 0.37 | 0.83 | 0.83 | 0.83 |
| | | | | | | | | | | | | |
| **Finland** | Arctic char | - | - | - | - | - | - | - | - | - | - | - |
| | Brown trout | 9 | 160 | 0.07 | 0.03 | 0.31 | 0.22 | 0.08 | 0.43 | 0.14 | 0.05 | 0.32 |
| | Perch | 495 | 6778 | 0.21 | 0.08 | 0.57 | 0.24 | 0.09 | 0.69 | 0.51 | 0.21 | 1.26 |
| | Pike | 211 | 4407 | 0.47 | 0.18 | 1.0, | 0.62 | 0.25 | 1.72 | 0.56 | 0.22 | 1.39 |
| | Roach | 39 | 328 | 0.36 | 0.17 | 0.61 | 0.36 | 0.16 | 0.77 | 0.95 | 0.46 | 1.65 |
| | | | | | | | | | | | | |
| **Norway** | Arctic char | 2 | 18 | 0.06 | 0.06 | 0.07 | 0.06 | 0.02 | 0.09 | 0.10 | 0.07 | 0.12 |
| | Brown trout | 49 | 1040 | 0.13 | 0.05 | 0.47 | 0.32 | 0.08 | 1.47 | 0.27 | 0.08 | 0.50 |
| | Perch | 53 | 2270 | 0.35 | 0.15 | 0.69 | 0.31 | 0.12 | 1.62 | 0.77 | 0.28 | 1.43 |
| | Pike | 6 | 120 | 0.48 | 0.06 | 1.22 | 0.61 | 0.16 | 1.21 | 0.54 | 0.11 | 1.13 |
| | Roach | 1 | 20 | 0.56 | 0.56 | 0.56 | 0.89 | 0.89 | 0.89 | 1.51 | 1.51 | 1.51 |
| | | | | | | | | | | | | |
| **Kola peninsula** | Perch | - | - | - | - | - | - | - | - | - | - | - |
| | Pike | - | - | - | - | - | - | - | - | - | - | - |
| | | | | | | | | | | | | |
| **Summary** | Arctic char | 2 | 18 | 0.06 | 0.06 | 0.07 | 0.06 | 0.02 | 0.09 | 0.10 | 0.07 | 0.12 |
| | Brown trout | 58 | 1200 | 0.13 | 0.05 | 0.46 | 0.41 | 0.08 | 1.29 | 0.30 | 0.07 | 0.46 |
| | Perch | 561 | 10521 | 0.21 | 0.09 | 0.57 | 0.25 | 0.10 | 0.83 | 0.55 | 0.23 | 1.30 |
| | Pike | 226 | 4871 | 0.49 | 0.22 | 1.10 | 0.70 | 0.25 | 1.70 | 0.60 | 0.22 | 1.41 |
| | Roach | 41 | 405 | 0.36 | 0.16 | 0.60 | 0.33 | 0.16 | 0.82 | 0.95 | 0.46 | 1.64 |

* Sweden only includes lakes that have been resampled > 5 years.

### 5.3.2 Spatial patterns in subset of lakes

The spatial coverage of data for perch and pike was suitable for testing for latitudinal gradients in fish Hg concentrations for the subset of lakes subject mainly to atmospheric inputs of Hg (Figure 22). Lakes classified as being subject mainly to loads of Hg from atmospheric deposition would likely have fish Hg levels that relate to patterns in Hg deposition and hence follow a south-to-north gradient with decreasing fish Hg concentrations to the north.

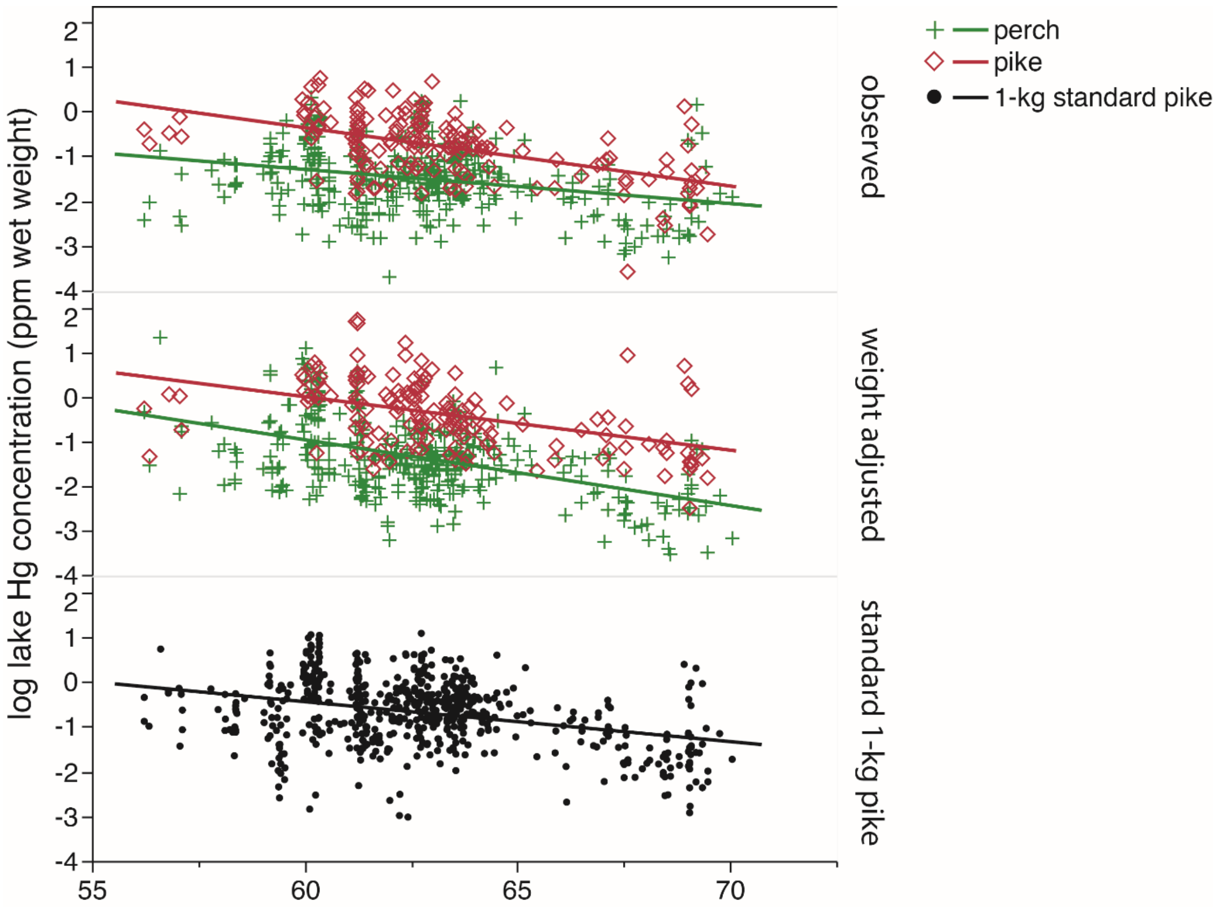


**Figure 22** Latitudinal gradient of fish Hg concentrations from countries across the Fennoscandian shield from lakes dominated by atmospherically deposited Hg. Linear regression was used for the presentation of the latitudinal gradient and were based on reported values from the database (top panel), weight normalised Hg levels (middle panel) and Hg levels in 1-kg pike (lower panel). Each symbol represent median for each lake.

Correlations between latitude and observed fish Hg concentrations showed decreasing fish Hg levels both for perch (r= -0.08, $p$<0.0001) and pike (r =-0.13, $p$<0.0001) and support the view that the Hg levels in these lakes follow patterns of Hg levels in the atmosphere and Hg deposition. However, the south-to-north gradient of Hg deposition is confounded by other gradients, including aqueous DOM, temperature, and sulphur (S) deposition, also possibly regulating Hg in fish. Thus, the spatial relationship between atmospheric Hg deposition and Hg in fish cannot simply be interpreted as evidence for a direct link between atmospheric Hg and fish Hg. A better understanding of controls of DOM, temperature and S deposition on Hg fish accumulation is necessary to be able to disentangle spatial drivers of fish Hg accumulation.

Normalised data for fish size also show negative correlations between latitude and lake medians of weight normalised fish Hg data for perch (r= -0.29, *p*<0.0001) and pike (r= -0.21, *p*<0.0001) and a standard 1-kg pike (r= -0.19, *p*<0.0001). This pattern also follows the Hg deposition gradient with decreasing Hg deposition from the south towards the north (indicated from surface sediment Hg concentrations, Figure 14). This points to the importance of separating lakes into groups based on whether their loads of Hg can be attributed to local (“3”) or atmospherically deposited (“1”) sources.

An increased strength (increasing correlation coefficient) in the Hg-latitude relation was seen when using normalised fish Hg data compared to observed fish Hg levels. The choice of normalisation method for fish Hg data is therefore a factor that contributes to the differences in the relationship between fish Hg levels and latitude.

### 5.3.3 Temporal trends in subset of lakes

The temporal trend in lakes classified as being subject to sources of Hg from mainly atmospheric sources show high inter-annual variability, but declining trends covering the whole study period were observed for perch and pike (Figure 23). The inter-annual variability of concentrations in lakes subject to atmospheric sources of Hg does not differ between the different methods of data normalisation. As an example, pike concentrations show a minimum in the mid-1970s, and three distinct peaks through time after that: early 1980s, mid 1990s and between 2005 and 2010 (Figure 23).

However, the linear regressions for the temporal trends show different directions depending on the method of data normalisation (Figure 23). This is similar to what was documented for the complete database (section 4.2.2). The observed data reveal a significant declining temporal trend for perch (r=-0.48, *p*<0.0001), while no significant temporal trend was observed for pike (r=-0.02, *p*=0.76, Figure 23). The weight adjusted concentrations show no significant temporal trend for perch (r=-0.03, *p*=0.60) or pike (-0.02, *p*=0.72). Despite the contrasting trends between species and method of normalisation, there was a significant decreasing temporal trend for normalised fish Hg data to a standard 1-kg pike (r=-0.16, *p*=0.0001).

When the temporal trends are broken down to represent each country in the database (Figure 24), it is evident that the direction of the temporal trend depend on what method is used for the normalisation of observed fish Hg data. For example, in Finland the observed Hg concentrations in perch did not change between 1967 and 2015 (r=-0.22, p=0.18), while the weight-adjusted perch Hg concentrations showed a significant increase (r=0.37, p=0.02). Also, in Norway the observed fish Hg concentrations between 1990 and 2012 showed no temporal trend for perch (r=0.001, p=0.98) or pike (r=0.73, p=0.16), while a normalisation of data with respect to fish weight revealed a significant increase in Hg levels for perch (r=0.60, p=0.002). In Sweden, the observed fish Hg concentrations in perch decreased from 1965 until 2013 (r=-0.50, p=0.003), while the perch Hg concentrations did not show any significant trend after normalisation of concentrations with respect to fish weight (r=0.007, p=0.97, Figure 24). Temporal trends of Hg concentrations in a standard 1-kg pike showed no trend in Finland (r=0.17, p=0.12), an increasing trend in Norway (r=0.52, p=0.01) and a decreasing trend in Sweden (r=-0.44, p=0.0007).

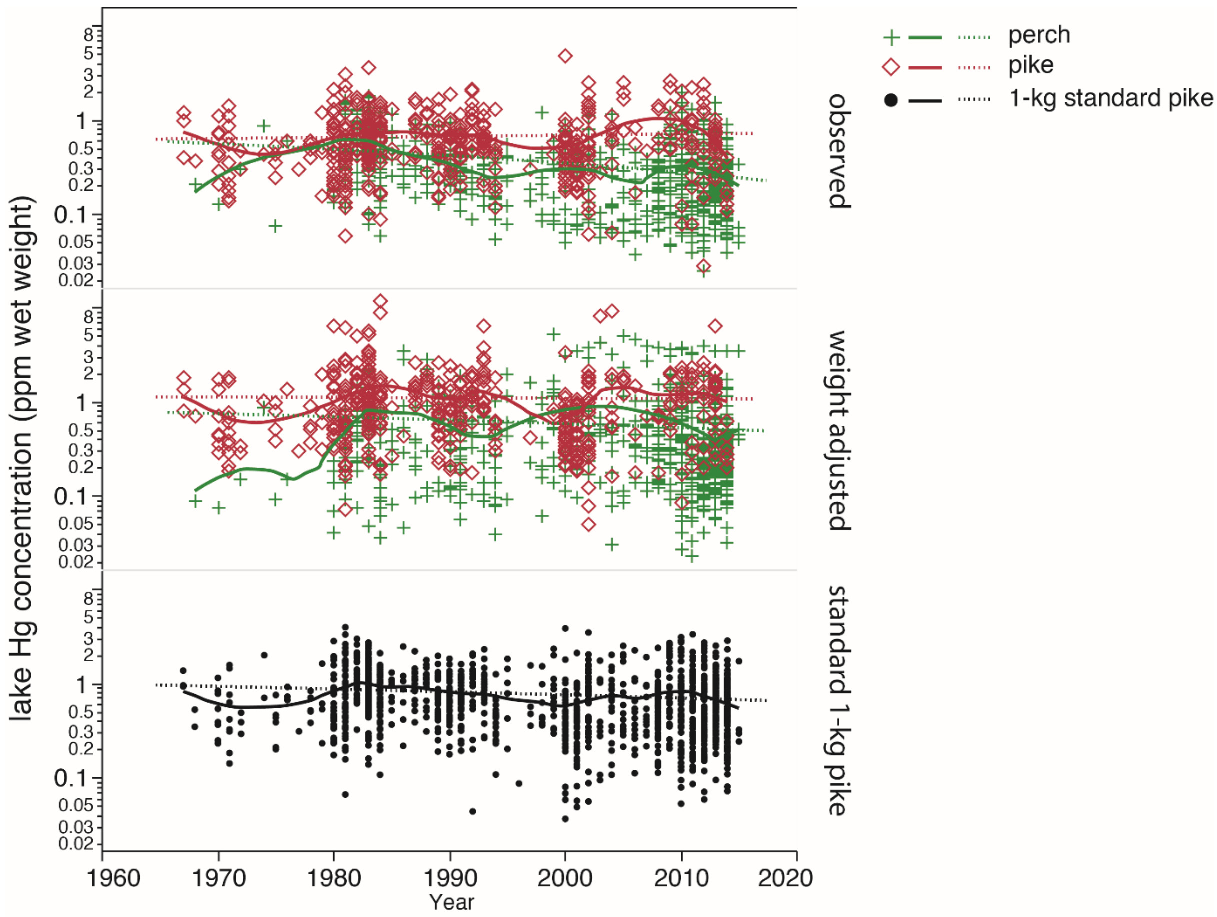


**Figure 23** Temporal trends of fish Hg concentrations from Fennoscandia from lakes subject to exclusively atmospheric deposited Hg with reported data covering more than 5 years. Data used for temporal trend analysis were based on reported values from the database (top panel), weight-adjusted Hg levels (middle panel) and Hg levels in 1-kg pike (lower panel). Each symbol represents annual median for lakes sampled each year. The temporal trend is presented both as a simple linear regression (dotted line) and a smoothed linear function (solid line) for perch and pike data individually.

Differences in temporal trends between the different methods of adjustments of the fish Hg data (observed, normalisation to fish weight, and a standard 1-kg pike) can be explained, at least partly, by a temporal trend in fish size (Figure 25), where perch size was declining ($r=-0.27$, $p<0.0001$). The normalisation of fish Hg data is needed to account for the temporal trend seen in fish size and is the main reason why there are differences in the temporal trends between observed fish Hg data and weight normalised data. Also, variation in fish species caught over time reported in the database (Figure 3 and 4) is taken into account, which adds to the differences in temporal trends between the methods for fish Hg data normalisation.

Temporal trends in the size of fish in the database also show contrasting trends between countries (Figure 25). In Finland, there was no significant change in pike weight over time ($r=-0.03$, $p=0.17$) while the temporal trend in Swedish data show that larger pike were caught over time ($r=0.27$, $p<0.0001$).

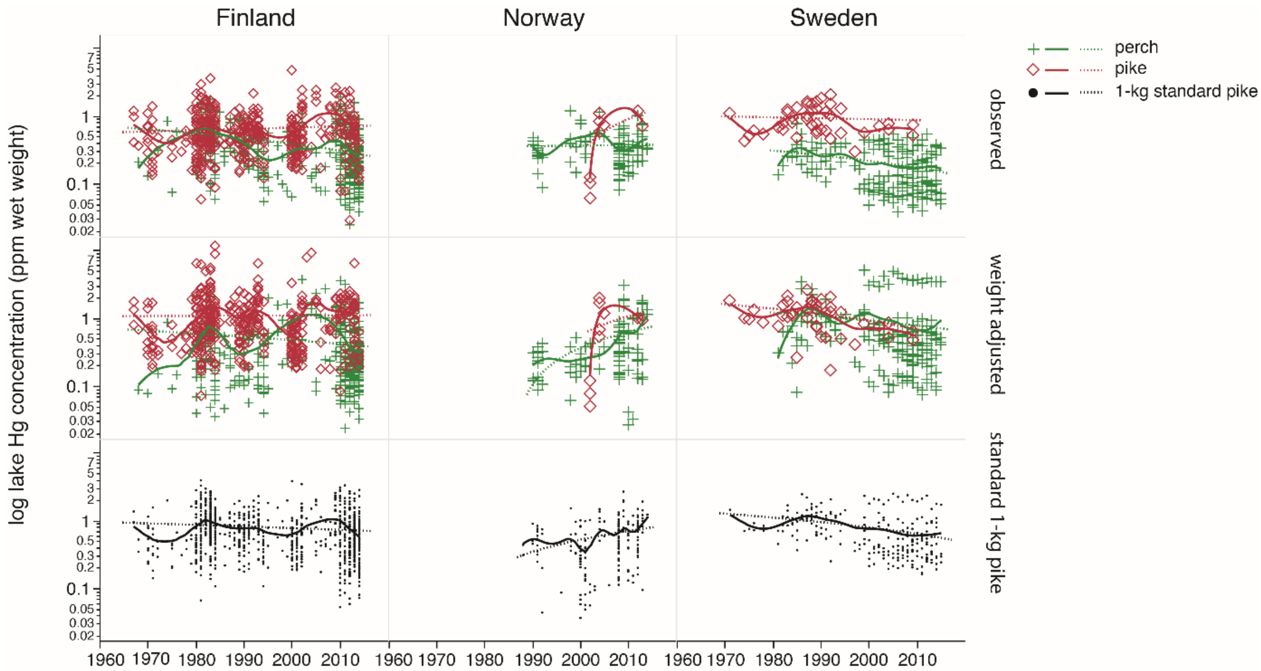


**Figure 24** Temporal trend of fish Hg concentrations from countries across Fennoscandia from lakes subject to exclusively atmospheric deposited Hg with reported data covering more than 5 years. Data used for temporal trend analysis were based on reported values from the database (top panel), weight-adjusted Hg levels (middle panel) and Hg levels in 1-kg pike (lower panel). Each symbol represents annual median for lakes sampled each year. The temporal trend is presented both as a simple linear regression (solid line) and a smoothed linear function (dotted line).

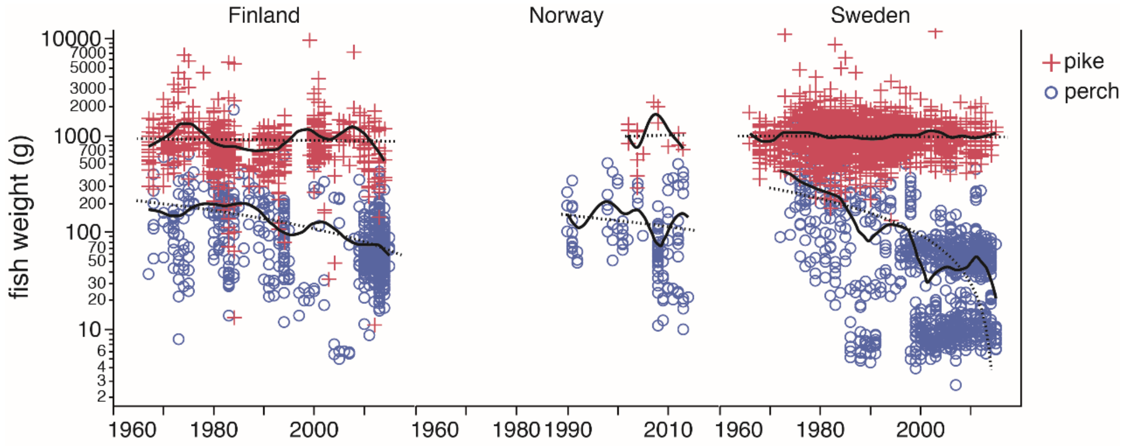


**Figure 25** Temporal trend of fish size from countries across the Fennoscandian shield from lakes subject to exclusively atmospheric deposited Hg with reported data covering more than 5 years. Each symbol represents annual median for lakes sampled each year. The temporal trend is presented both as a simple linear regression (solid line) and a smoothed linear function (dotted line)

### 5.3.4 Future work

The database compiled and presented in this report will be of great value for further analysis of drivers of fish Hg levels. Specifically, the possibility for increasing the database with similar measurements and work from North America (i.e. Canada and Alaska, US) and Russia, will be important to pursue. There is also the potential to create a circum-boreal database that can document the effects of historical atmospheric deposition of Hg. This is particularly relevant for the coming 5-20 years given that the Minamata Convention entered into force in 2017, demanding monitoring of Hg in the environment (including fish) in order to document the success (i.e. effectiveness) of the convention. This will also demand a focus on identification of legacy Hg sources (i.e. local direct industry emission) and on separating these sources from long-range atmospheric sources of Hg.

With the current database of fish Hg measurements, a range of possible drivers for changing fish Hg concentrations can potentially be tested and investigated, examples including:

- Despite the significant reductions in Hg releases (Streets et al., 2011) and deposition fluxes (EMEP, 2016) to many areas of the boreal region the last six decades, trends of Hg concentrations in fish populations do not seem to follow the same patterns, which could be due to the substantial amount of historical Hg being stored in the lake catchments and continuously being transported to the adjacent lakes;
- Temporal trends of fish Hg concentrations show inter-decadal differences with concentrations decreasing throughout the 1990s following the large reduction of Hg depositions (EMEP, 2016) to the environment, and concentrations increasing in recent years following reduced acid deposition leading to increased releases of organic matter (Monteith et al., 2007), and subsequently Hg, from catchments to lake surface waters;

Additionally, it will be possible to investigate more detailed processes from the database, examples including:

- Fish Hg concentrations from individual lakes reveal significant local variations with lake-specific temporal trends being significantly different from the regional Fennoscandian trends;
- As a response to the significantly higher atmospheric temperature raise in Arctic areas compared to boreal areas the last 50 years, there are significant differences in temporal trends between southern and northern parts of Fennoscandia;
- As a response to differences in increasing TOC rates between Arctic areas and boreal areas (Monteith et al., 2007), there are significant differences in temporal trends between regions with different increases in aqueous TOC concentrations.

# 6 Uncertainties and limitations

With the available database in hand, and the opportunities to analyse patterns and trends in fish Hg concentrations following the gathered data, the authors would like to highlight some of the uncertainties emerging from the conclusion drawn in this work. As highlighted throughout several chapters in this report, models for normalisation of fish Hg concentrations to fish size/age can and should be developed further, including validation with independent datasets. Not only do the methods presented in this work (observed data, weight-adjusted data and standard 1-kg pike adjusted data) yield different patterns on a spatial scale, they also affect the observed temporal trends in the dataset. For our detailed study of lakes that mainly received Hg from atmospheric deposition and also sampled on > 5 year throughout the study period, both unchanged and decreasing trends are observed depending on the choice of normalisation method.

To be able to relate the temporal trends of Hg concentrations in Fennoscandian fish to atmospheric deposition of Hg, an essential part of the data analysis is to classify individual lakes based on main sources of Hg input. This is particularly important in Sweden, where different industries (e.g. pulp, paper, and chlor-alkali industry) have been located close to waterways where determination of Hg levels in fish were part of environmental monitoring programmes. Due to the large number of such industries and lack of information about where, when and how they operated, it is extremely difficult to provide a complete list of Fennoscandian lakes where some sort of local Hg pollution source has been present throughout history. A great deal of uncertainty is therefore connected to the lack of knowledge on individual lakes in the present database, an example being the number of lakes (n = 2068) where we do not have enough information to classify the lake as either a lake affected predominantly by long-range transported Hg (group 1) or a locally contaminated lake (group 3, see sections 3.5 and 4.3.1). Important questions to answer in future use of this database will be how legacy Hg pollution is affecting Hg levels and cycling in the lakes.

Additional aspects of uncertainties may be related to analytical procedures. The current report and database have not recorded any information with respect to this, and although measurements of Hg in fish muscle rely on standardised analytical techniques and procedures, the methods to analyse Hg in biota have changed over the last decades and can also differ between labs.

Within each lake, fish species, fish population and year, there are substantial ranges in fish Hg concentrations and fish sizes. Fish age is more time consuming to record than fish length and weight. Following this, length and/or weight are often used as an indication of age. However, fish from different lakes may show differences in growth rates, with higher bioaccumulation expected for lakes with slower growing fish. This again will affect the Hg-weight relationships and, as such, will also affect the spatial patterns and temporal trends presented in the present report.

# 7 Conclusions and future perspectives

Efforts to decrease environmental impacts of mercury from atmospheric, anthropogenic emissions are largely coordinated through international processes such as the CLTRAP and the Minamata Convention. The main goal for this report was to assess effects of atmospherically transported air pollutants on fish Hg levels, and the results provide an overview on fish Hg levels across Fennoscandia. This is relevant for the Minamata Convention that entered into force in August 2017. Fish Hg data in the database originates from Norway, Sweden, Finland and the Kola Peninsula, including a diversity of lakes that covers a latitudinal gradient from 55°29'N to 70°1'N, and several fish species. Main conclusions from this report are as follows:

- Median lake-specific fish Hg concentrations in the vast majority of the studied Fennoscandian lakes exceed EQS for Hg in biota (0.02 ppm) set by EU WFD;
- Mean observed Hg concentrations were lower for lakes affected by dominantly atmospherically deposited Hg (0.25 ± 0.27 ppm, n = 703) compared to lakes affected also by local pollution sources (0.55 ± 0.38 ppm, n = 167), reflecting the significance of direct historical industrial releases of Hg to surface waters for present day Hg concentrations in fish;
- Fish Hg concentrations within lakes that were classified as being subject mainly to sources of Hg from atmospheric deposition, show a decreasing south-to-north concentration gradient. This suggests that lower atmospheric Hg deposition leads to lower accumulation of Hg in fish, but the Hg deposition gradient is confounded by similar gradients in aqueous DOM, S deposition and temperature, which are other strong regulators of Hg in fish;
- While a significant long-term decline in fish Hg concentrations was observed, the trend was much stronger for the entire database than for lakes only impacted by atmospheric sources of Hg. We attribute the contrast in trends primarily to declining impacts of local pollution sources;
- Temporal trends in Hg concentrations were not consistent across fish species, fish normalisation methods, or country/region of origin. Additionally, it is clear from the temporal trends analyses that, despite high concentrations in the 1970s, inter-annual variation of fish Hg concentrations, even for normalised data, were larger than the long-term changes;
- After correction of data to a standard 1-kg pike, a consistent and significantly decreasing trend for both the entire database and the lakes only influenced by atmospheric deposition of Hg was observed. This decline is consistent with the reported decline in atmospheric Hg emissions and deposition from EMEP, which is a strong indication of reduced Hg exposure to the aquatic food web.

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# 9 Reports and publications from the ICP Waters programme

All reports from the ICP Waters programme from 2000 up to present are listed below. Reports before year 2000 can be listed on request. All reports are available from the Programme Centre. Reports and recent publications are also accessible through the ICP Waters website; http://www.icp-waters.no/

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