Does forest harvest increase the mercury concentrations in fish? Evidence from Swedish lakes

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HIGHLIGHTS

• Small Swedish forest lakes subjected to commercial forest harvest within catchments
• Extensive fish sampling in small forest lakes with and without harvest
• Fish Hg bioaccumulation increased by forest harvest, but with large variation
• Fish Hg concentration varies between lakes, years, treatment and fish size.

GRAPHICAL ABSTRACT

A number of studies have evaluated the effects of forest harvest on mercury (Hg) concentrations and exports in surface waters, but few studies have tested the effect from forest harvest on the change in fish Hg concentrations over the course of several years after harvest. To address this question, mercury (Hg) concentrations in perch (Perca fluviatilis) muscle tissue from five lakes were analyzed for two years before (2010–2011) and three years after (2013–2015) forest harvest conducted in 2012. Fish Hg concentrations in the clear-cut catchments (n = 1373 fish specimens) were related to temporal changes of fish Hg in reference lakes (n = 1099 fish specimens) from 19 lakes in the Swedish National Environmental Monitoring Programme. Small (length < 100 mm) and large perch (length ≥ 100 mm) were analyzed separately, due to changing feeding habitats of fish over growing size. There was considerable year-to-year and lake-to-lake variation in fish Hg concentrations (−14%–121%) after forest harvest in the clearcut lakes, according to our first statistical model that count for fish Hg changes. While the effect ascribed to forest harvest varied between years, after three years (in 2015), a significant increase of 26% (p < 0.0001) in Hg concentrations of large fish was identified in our second statistical model that pooled all 5 clearcut lakes. The large fish Hg concentrations in the 19 reference lakes also varied, and in 2015 had decreased by 7% (p = 0.03) relative to the concentrations in 2010–2011. The majority of the annual changes in fish Hg concentrations in the clearcut lakes after harvest were in the lower range of earlier predictions for high-latitude lakes extrapolated primarily from the effects of forest harvest operations on Hg concentrations in water. Since the risk
of forest harvest impacts on Hg extends to fish and not just surface water concentrations, there is even more reason to consider Hg effects in forestry planning, alongside other ecosystem effects.

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1. Introduction

The raw materials provided by forest harvest are of great socio-economic value. There are, however, concerns about how forestry operations influence water quality (Futter et al., 2016; Roberge et al., 2016). One of these concerns is the possible effects on mercury (Hg) concentrations in freshwater biota in boreal waters that already pose a risk to human and wildlife health (Åkerblom et al., 2014). A number of studies have evaluated the effects of forest harvest on Hg concentrations and exports in surface waters, but few studies have tested the direct effect from forest harvest on fish Hg concentrations. Since there is increasing evidence that the export of total mercury (THg) and the bioaccumulative methylmercury (MeHg) is affected by forest operations (Eklöf et al., 2016), the limited information on the biotic effects of forest harvest on fish Hg concentration is a concern in itself. Further investigations on the connection between forestry and Hg concentrations in biota, especially fish, are thus needed to provide a better basis for managing the many trade-offs inherent in forest management.

The concentrations of fish Hg in all Swedish freshwater exceed the EU criterion for good chemical status (0.02 mg kg\(^{-1}\) wet weight (w.w.)) (EC, 2012) and half of the lakes exceed the WHO standard for safe fish consumption (0.5 mg kg\(^{-1}\) w.w.) (Åkerblom et al., 2014; FAO/WHO, 2015). Any disturbance that may worsen this situation needs to be recognized and measures should be applied to minimize effects from it. Forest harvest is one such disturbance that may be followed by increasing Hg concentrations in fish of streams and lakes. One reason for this is that forest harvest can influence the bioavailability of Hg through increasing net Hg methylation which can respond to changes in the soil environment, e.g. sulphur, temperature, seasonality, all of which can change in a harvested forest environment (Eklöf et al., 2015). In high latitude regions, forestry effects on fish Hg concentrations were identified by pioneering studies around Scandinavia (Rask et al., 1998) and Canada (Garcia and Carignan, 2000). Porvari et al. (2003) observed significant increases in THg and MeHg concentrations, up to 48% and 133%, respectively, in runoff from a small forest catchment during three years after clearcutting and site preparation. Munthe and Hultberg (2004) found even larger increases (over 300%) in MeHg after a skidder road respect initiation of forest harvest impacts on Hg extends to fish and not just surface water concentrations, there is even more reason to consider Hg effects in forestry planning, alongside other ecosystem effects.

Two as pointed out by Eklöf et al. (2016), most of the studies since those pioneering works (pre-2006) have focused on Hg concentrations in water (and aquatic loadings), rather than the biota, with some notable exceptions e.g. Desrosiers et al. (2006); Garcia et al. (2007) and de Wit et al. (2014). Desrosiers et al. (2006) and Garcia et al. (2007) both observed a decrease in biomass and a concurrent increase in Hg concentrations in periphyton and zooplankton. de Wit et al. (2014) reported no significant impacts of forest harvest on Hg concentrations in macroinvertebrates (and stream water) from a catchment with about 30% forest harvest although the forestry operations resulted in considerable disturbance of the soils in the harvested area. So far, there has been no study on the direct connection between different areal extents of forest clear-cutting across several catchments over a period of several years to examine how fish Hg concentrations respond to commercial forestry operations in a boreal setting.

The forestry effects on Hg in runoff water and biota have been attributed to the generally wetter and warmer conditions in soils when trees are harvested and transpiration decreases in combination with more snow accumulation in clear-cut areas (Shanley and Bishop, 2012). This in turn increases runoff and changes preferential hydrologic groundwater flow paths that may increase the mobilization of Hg (Eklöf et al., 2016). Furthermore, sub-oxic conditions form in waterlogged and disturbed areas that promote net methylation of inorganic Hg (Kronberg et al., 2016). Simultaneously, enhanced levels of dissolved organic carbon (DOC) in first-order stream runoff after forest harvest (Schelker et al., 2014) likely increase the transport of methylated Hg to aquatic ecosystems further downstream. How altered water chemistry conditions influence Hg bioaccumulation in food webs is well studied: lowered pH enhances both Hg concentrations in the food web base and further biomagnification factors, but not Hg biomagnification slopes (Jardine et al., 2013), while lowered TOC influenced Hg delivery to aquatic systems with enhanced Hg bioavailability to organisms at the base of food webs, and consequently higher Hg biomagnification slopes (Clayden et al., 2013). However, the impact of TOC on fish Hg bioaccumulation still has many uncertainties, some of which relate to interactions with changing atmospheric sulphur deposition, e.g. Lawrence et al. (2016) stated on increased Hg in forest soil resulted from elevated organic carbon after liming acidified lakes with decreased sulphur deposition.

It is important to mention the environmental benefits of utilizing forests (such as reducing the use of fossil fuels, and other substitution effects on the greenhouse gas balances), especially since the ultimate source of the Hg released by forestry practices is often anthropogenic emissions in other parts of the world (Lundmark et al., 2016). In the boreal area, most of the Hg deposited to forests is retained in the soils (Johansson et al., 2001). Forest soils thus constitute the largest pool of Hg in the catchment that eventually become mobilized and methylated after forest harvest.

Given the concern about the impact of forestry operations on Hg concentrations in Swedish lakes, where Hg levels already pose a threat to the health of humans and wildlife, the aim of this study was to determine whether forestry impacts the fish Hg concentrations in the years after forest harvest. To do this we investigated Hg concentrations in perch (Perca fluviatilis) in 5 Swedish lakes over the course of two pre-harvest (2010–2011) and three post-harvest (2013–2015) years. Between 6% and 40% of the lake catchments were subjected to commercial forest harvest in 2012. Fish Hg concentrations in the harvested lakes...
were related to one unharvested reference lake sampled during the same time period. The lakes are spread across Sweden with 400 km separating the most northerly and southerly lakes. The hypothesis is that forest harvest impacts the temporal change in fish Hg concentrations.

To account for the fish size-Hg relationship, fish weight was included in the statistical analysis. To provide a larger pool of references with which to compare the temporal changes observed in this study, data on perch Hg concentrations from 19 Swedish National Environmental Monitoring Programme (EMP) lakes were also used as references (Fig. 1). We also identified the changes in the natural abundance of $^{13}$C and $^{15}$N in fish muscle samples to better understand trophic shifts over time (McCutchan et al., 2005; Lavoie et al., 2013). Signatures for trophic ecology in perch provide insight into the trophic position ($\delta^{15}$N) and food sources ($\delta^{13}$C) of fish (McCutchan et al., 2005). $\delta^{13}$C values increase with terrestrial input in the diet and $\delta^{15}$N values increase along the trophic position. More negative $\delta^{13}$C values in aquatic biota and consequently increased Hg biomagnification, has been attributed to increased pelagic feeding of periphyton (Pouilly et al., 2013).

2. Materials and methods

2.1. Study area and sampling

Six small headwater lakes were selected for the study. The five lake catchments that were subject to conventional forest harvesting are referred to as clearcut lakes (Björntjärn (BJ), Brobo-Kroktjärn (BK), Gårdsjön (GA), Kroktjärn (KR), Långtjärn (LA)). One small catchment was not harvested (Svultentjärn (SV), and this is referred to as the clearcut reference lake (Table 1). The lakes are all situated in managed forest catchments across Sweden (Fig. 1). The lake catchments are 10–100 ha, and are not downstream from other rivers or lakes. These forest lakes are all oligotrophic and small. They have catchments underlain by slow weathering bedrocks of igneous origin overlain by glacial tills on which the predominant soil type are podzols, with histosols in lower lying areas (Sonesten, 2003b). Commercial forest harvest (affecting 6 to 40% of the catchment area) was conducted in the catchments during the autumn of 2012 according to information from the Swedish Forest Agency (www.skogsstyrelsen.se).
Fish from a number of freshwater lakes in Sweden have been sampled regularly for Hg concentrations, some since 1965, in what is now a part of the Swedish National Environmental Monitoring Programme (EMP) commissioned by the Swedish Environmental Protection Agency (www.sweishetsa.se). The EMP lakes selected for this study represent small, forested, headwater, remote, and relatively pristine freshwaters across the nation (Gotland excluded), with sufficient samples (> 10 fish sampled between Aug-Sep from each lake every year since 2010). These criteria were met by 19 EMP lakes. Data from the EMP lakes were retrieved from the Swedish national data host for Hg in fish, the Swedish Environmental Research Institute (https://dvsb.viw.se, latest accessed in Apr 17, 2017).

Specimens of perch (Perca fluviatilis), were sampled from the lakes using gill nets composed of different mesh sizes to target a broad variation of fish sizes following the European standard (Lundgrens Fiskredskap AB) (CEN, 2005; Champ et al., 2009). The sampling was done annually (2010, 2011, and 2013, 2014, 2015) in August–September. Water samples were taken concurrently with fish sampling events. Two benthic multi-mesh gillnets were applied to each lake and left standing in the water overnight (≈ 12 h). The nets were then taken and rinsed as soon as possible, usually within ~6 h. The captured perch were measured for their weight and total length (from the tip of the snout to the tip of the caudal fin) and archived individually at ~20 °C. Fish muscle samples were obtained later by dissecting the dorsal fin muscle from the thawed frozen whole-fish samples. Small muscle pieces (~1 g wet weight) were sectioned into 2 ml polypropylene plastic vials and stored (~20 °C) until they were freeze-dried (~72 h). For the determination of fish sample wet content both the wet and dried weights of each muscle sample were measured. Freeze-dried samples were kept frozen at ~20 °C until total Hg analysis. One pair of fish gill covers, or operculum, were taken for age determination.

2.2. Sample analysis

2.2.1. Total mercury

Total Hg concentrations ([Hg]) in fish muscle samples were analyzed with a SMS 10 Total Mercury Analyzer (PerkinElmer Inc.) through thermal decomposition (750 °C) followed by amalgamation on a gold-trap, thermal desorption, and analysis of Hg vapor by Atomic Absorption Spectroscopy according to EPA method 7473 (USEPA, 2007). The amount of muscle tissue used for Hg analysis was between 5 and 10 mg. Blanks and standards of certified reference material (either DOLT-4 ([Hg] = 2.58 ± 0.22 mg kg⁻¹) or DORM-4 ([Hg] = 0.410 ± 0.055 mg kg⁻¹)) were analyzed at least once per every 10 samples to assess accuracy of the analysis (Blanks and standards covariance <10%). Precision was assessed by analyzing duplicates of at least every 10th sample. The coefficient of variation from duplicate analyses was under 3%.

2.2.2. Stable isotope analysis

A subset of fish muscle samples was selected for stable isotope analysis to identify the natural abundance of 13C and 13N. The amount of muscle tissue used for stable isotope analysis was approximately 1 mg. The analysis was carried out by Stable Isotope Facility at UC Davis, United States. Stable isotope values are expressed in δ notation as parts per mille deviation from a standard reference:

δ¹³C or δ¹⁵N = [(Rsample/Rstandard) − 1] × 1000

where R is ¹³C/¹²C, or ¹⁵N/¹⁴N.

2.2.3. Water chemistry

Water samples were shipped for analysis to the Department of Aquatic Sciences and Assessment (Swedish University of Agricultural Sciences, Uppsala). Analysis was done within 7 days after sampling and data were collected for pH, total nitrogen (totN), total phosphorus (totP), iron (Fe), total organic carbon (TOC), chlorophyll a (Chl a), alkalinity, sulphate, and conductivity. Analyses were conducted using analytical methods accredited by the Swedish Board for Accreditation/Conformity Assessment (SWEDAC). Details of the water chemistry analytical methods are included in supporting information (Supporting Information (SI) Table S2).

2.2.4. Age determination

Among the fishes analyzed for [Hg] around 20 fish samples were randomly selected from each lake to be analyzed for age (except from lake Kroktjärn where only 14 perch samples were obtained after forest harvest). The fish operculums were cleansed using warm water and dried at room temperature. Age rings of the operculum were then counted underneath a stereoscope (Appelberg et al., 1995).

2.2.5. Statistical analysis

The statistical analysis of the data was performed using the software JMP 10 (© SAS Institute Inc.). The level of significance for all tests was set at 3 levels (weak: 0.1 > p > 0.05, moderate: 0.05 > p > 0.01, and strong: p < 0.01). Fish [Hg] and fish size data were log-transformed in the statistical models to achieve data normality for the residuals according to the Shapiro-Wilk Goodness-of-Fit test (Shapiro and Wilk, 1965). To take into account the covariation between log-transformed fish [Hg] (log[Hg]) and fish size, log transformed fish weight was used as the covariate to adjust for differences in fish size within and between lakes (Sonesten, 2003a). Due to the complexity of the dataset where fish samples were not always available from each lake every year during the investigation period, especially for the small fishes, the effect of forest clear-cutting on fish [Hg] was estimated by general linear modelling (GLM) with log[Hg] used as the response variable for every one year after the forest harvest period during 2013 and 2015. Special cases of GLM, analysis of variance (ANOVA) and analysis of covariance (ANCOVA) for log[Hg], δ¹³C and δ¹⁵N were used to test for statistical differences in the fish [Hg] and stable isotopes at different time periods. Years 2010–2011 were combined as the reference period, since each of the clearcut lakes were sampled during these two years prior to forest harvest.

Considering the highly variable feeding habits of perch, particularly in the boreal landscape (Rask, 1986), with young perch (age < 2) feeding on zooplankton and small macroinvertebrates, while bigger perch
are mostly piscivorous (Thorpe, 1977), we divided the fish into two size groups, small (length < 100 mm) and large (length ≥ 100 mm) fishes. Details on the size classification, which takes account of fish age and length-weight ratio (analogous to fish growth rates) (Le Cren, 1951; Rocha et al., 2015), are presented in SI 1a. The same statistical models were applied to each size group separately.

The δ13C values before and after forest harvest were compared across lakes. For the δ15N values, we derived trophic magnification slopes (TMS) from the regression of log[(Hg)fi] vs δ15N. This was done for each lake and compared with each other, regardless of the baseline (a) (Lavoie et al., 2013). The calculation of TMS is the slope (b) in Eq. 1:

$$\log[Hg] = b\delta^{15}N + a$$

(1)

B > 0 indicates biomagnification along the trophic levels, while b < 0 is a likely result from trophic dilution, and b = 0 indicates that there is no change (Gobas et al., 2009).

To distinguish temporal trends in fish [Hg] between periods before and after forest harvest, two types of GLM modelling were applied. In the first type a temporal trend was analyzed only for individual clearcut lakes and the single clearcut reference (GLM1). In the second type, the temporal trend in the clearcut lakes was tested against a general temporal trend in Swedish lakes represented by the EMP lakes (GLM2).

The modelling of temporal change in fish [Hg] for individual clearcut lakes and clearcut reference (GLM1, Eq. 2):

$$\log[Hg] = \alpha + \beta \times I_{after \cc} + \delta \log[W] + \gamma \times I_{after \cc} \log[W] + \epsilon$$

(2)

included estimates of the intercept (α) and the estimated treatment effect (β), indicating the mean difference before and after the clearcut event. Log-transformed fish weight was included in the model as a covariate to correct for individual fish size (SI 1b). This is estimated as a linear relationship between log-transformed fish weight and log [Hg] which is quantified as the slope parameter (δ). Since the relation between log fish weight and log [Hg] could change after the clearcut event, the interaction between time point and log fish weight is also determined (γ). This model was fitted for each individual lake, separately for small fish (length < 100 mm) and large fish (length ≥ 100 mm). To compare a common temporal trend in the clearcut (CC) lakes against Swedish lakes in general, a similar model (GLM2) was applied, one for clearcut lakes and one for EMP lakes (Eq. 3):

$$\log[Hg] = \alpha + \beta \times I_{after \cc} + \delta \log[W] + \gamma \times I_{after \cc} \log[W] + \epsilon$$

(3)

This model is quite similar to the GLM1 model in Eq. 2 above, except that several lakes were combined in the analysis (5 lakes in the CC group and 19 in the EMP group, excluding Sultentjärn). Therefore, a random factor \(X_{lake}\) is included in the model to account for the resulting hierarchical data structure. The interaction effect (γ) between time point and log fish weight can be removed when it turns out to be insignificant in model output.

The relative change in fish [Hg] (Relative change (%)) after clearcutting was calculated (Eq. 4):

$$\text{Relative change(%) = } 100 \times \frac{\text{Fish[Hg]}_{\text{fi}} - \text{Fish[Hg]}_{\text{fi}}}{\text{Fish[Hg]}_{\text{fi}}_{\text{A}}}$$

(4)

using the least square mean estimate (LS Mean) of fish [Hg] before (Fish [Hg]fi) and after (Fish[Hg]fi) forest harvest from the GLM output. This was done for all models using GLM1 and GLM2.

3. Results

3.1. Water chemistry

The six lakes sampled in this study (clearcut and clearcut reference) were oligotrophic (totP: 3–18 μg L−1, totN: 310–1100 μg L−1) with relatively high dissolved organic carbon (DOC) concentrations (>5 mg L−1) (Vollenweider and Kerekes, 1980) (SI Table S3). Water chemistry data for EMP lakes were retrieved from the Miljodata-MVM database (Folster et al., 2014) (https://miljodata.slu.se) and listed in SI Table S3. All of the EMP lakes are also oligotrophic according to the same criteria from Vollenweider and Kerekes (1980) (SI Table S4). No significant differences in water chemistry parameters (pH, Fe, tot P, tot N, TOC, conductivity, alkalinity, and sulphate) were found between the period before and after harvest occurred in 2012 (paired t-test, p > 0.05).

3.2. Fish sizes and total Hg

Over the sampling period 1373 fish specimens were captured and analyzed for [Hg] in the five clearcut lakes. Fewer fish specimens were collected in the period before forest harvest (n = 559) compared to the period after forest harvest (n = 814) in the clearcut lakes (Table 2). The clearcut reference lake (Svultentjärn) had in total 430 fish specimens with 282 fishes captured before forest harvest and 148 after forest harvest. The captured fish ranged in length (62–390 mm) and age (1–13 years). The [Hg] was correlated with both fish length and weight across all clearcut lakes and Svultentjärn (r < 0.41, p < 0.001). Generally, both fish length and weight were significantly higher in the period after forest harvest compared to the period before forest harvest in the clearcut lakes (length ANOVA: F ratio = 69 (degrees of freedom = 4), p < 0.0001; weight ANOVA: 29(4), p < 0.0001), except lake Björntjärn and Brobo-Kroktjärn. There was a decrease in fish length (ANOVA: 6(415), p = 0.012) and weight (ANOVA: 9(415), p = 0.0025) between the two periods (2010–2011 and 2013–2015) in the clearcut reference lake Sultentjärn. Collected small fishes were relatively insufficient compared to large fishes: Data on small fish from the clearcut lakes were only available until 2013, and for the EMP lakes until 2014 (SI Table S7 and S8).

A general temporal change of fish [Hg] in Swedish lakes was represented by 19 lakes that have been sampled in the Swedish EMP. A total of 1099 fish specimens were collected from these lakes between 2010 and 2015 (Table 2). Mean fish [Hg] of large perch in the six lakes sampled in this study was higher compared to the EMP lakes that were used for comparison of temporal trends (Table 2). Length and weight of large fishes in the EMP lakes were lower in the period 2013–2015 (length: 170 ± 22 mm, weight: 52 ± 17 g) compared to the period 2010–2011 (length: 176 ± 20 mm, weight: 56 ± 17 g) (ANOVA: 5(1072), p = 0.03), in contrast to the clearcut lakes (but similar to the clearcut reference lake Sultentjärn). However, this simple comparison does not take fish weight variation over time in specific lakes overtime into account as in the GLM1 and GLM2 models presented later in this paper. Detailed fish [Hg] from the 19 EMP lakes can be found in SI Table S5.

None of the clearcut lakes annual fish [Hg] was statistically correlated with any of the 9 water chemistry parameters presented in SI Table 3. The one exception was Lake Sultentjärn fish [Hg] which was significantly correlated with TOC on yearly average (log[Hg] vs. TOC: n = 5, p = 0.006, r = 0.97). When pooling both clearcut lakes and Sultentjärn together, totP, was negatively correlated with annual fish [Hg] (log[Hg] vs. totP: n = 30, p = 0.03, r = –0.42). Please note that the lake water chemistry in the clearcut lakes was sampled only once per year, which is not enough for a good characterization of the annual lake water chemistry.

On the other hand, when looking at the 19 EMP lakes dataset where average annual chemistry was comprised of 3 to 24 samples from the
years fishes were sampled in the clearcut lakes, there were a number of significant correlations with annual fish [Hg] (in log[Hg]) when pooling all the lakes together: totN ($n = 95, p = 0.0001, r = 0.4$), totP ($n = 95, p = 0.003, r = 0.3$), TOC ($n = 95, p = 0.001, r = 0.53$), Fe ($n = 95, p = 0.005, r = 0.31$), and Chl a ($n = 95, p = 0.003, r = 0.22$) (Details of the correlations can be found in SI Table S6).

### 3.3. Stable isotopes and total Hg

The $\delta^{13}C$ and $\delta^{15}N$ of fish samples ranged from $-34$ to $-19.6\%$, and from $3.2$ to $13.8\%$, respectively. The variation of stable isotope values within each lake was much smaller (Table 3). There is a significant and positive correlation between $\delta^{15}N$ and fish weight (expressed as logW, $r > 0.51$) across all clearcut and reference lakes.

According to Eq. 3 (excluding the lake random effect), we tested the differences of the $\delta^{13}C$ and $\delta^{15}N$ values in the clearcut lakes and clearcut reference lakes before and after forest harvest. The $\delta^{13}C$ decreased by 1.16% after forest harvest in the clearcut lakes ($p < 0.0001$), while this value increased by 0.36% in the clearcut reference lake Svultentjärn ($p < 0.0001$) (Table 3). On the contrary, the $\delta^{15}N$ values increased significantly (0.78%, $p < 0.0001$) in the clearcut lakes after forest harvest, but remained stable in the clearcut reference lake Svultentjärn ($p > 0.9$) (Table 3).

The trophic magnification slopes (TMS) of fish [Hg], or the slope of log[Hg] vs. $\delta^{15}N$ regression (Eq. 1) for all clearcut lakes combined was 0.05, while the slope was 0.08 for the clearcut reference lake. The slope of log[Hg] vs. $\delta^{15}N$ regression varied between the lakes in large fish with length ≥ 100 mm. The difference in the slope before and after forest harvest is statistically significant (ANOVA: Estimate ± SE: 0.19 ± 0.09, $p = 0.03$) (Fig. 2). TMS calculation for fish muscle samples with length < 100 mm was not possible due to the limited number of samples ($N < 5$) tested for the $\delta^{13}C$ and $\delta^{15}N$ values.

### 3.4. Temporal change in total Hg

#### 3.4.1. Clearcut lakes versus clearcut reference lakes

The change in fish [Hg] before and after forest clearcut in the clearcut lakes were identified by using GLM1 (Eq. 2) and compared to the change in the clearcut reference lake Svultentjärn. In 2013, one year after the forest harvest, there was an increase in large fish [Hg] (length ≥ 100 mm) in lake Brobo-Kroktjärn (22%, $p < 0.0001$), Kroktjärn (43%, $p = 0.02$), and Långtjärn (121%, $p < 0.0001$) when compared with fish [Hg] in 2010–2011 (Fig. 3, SI Table S6). There was no significant

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**Table 2**

Summary of fish sizes and Hg concentrations from the lakes before and after forest harvest (Mean ± SD classified in small (length < 100 mm) and large (length ≥ 100 mm) fishes. The age is based on a subset of ~20 random fish samples from each lake.

| Lake                      | Before forest harvest (2010–2011) | After forest harvest (2013–2015) |
|---------------------------|----------------------------------|----------------------------------|
|                           | N (Length (mm)) | Weight (g) | Age (Years) | [Hg] (mg/kg ww) | N (Length (mm)) | Weight (g) | Age (Years) | [Hg] (mg/kg ww) |
| Clearcut lakes            |                   |             |             |                |                   |             |             |                |
| BJ                        | 22                | 135 ± 20    | 37 ± 12     | 3.6 ± 1.3      | 258               | 146 ± 15    | 28 ± 7      | 3.7 ± 1.2     | 0.34 ± 0.14   |
| BK                        | 88                | 189 ± 17    | 73 ± 20     | 5 ± 1.9        | 259               | 179 ± 34    | 61 ± 36     | 4.1 ± 2.7     | 0.23 ± 0.1    |
| GA                        | 185               | 162 ± 43    | 59 ± 66     | 3 ± 1.2        | 213               | 197 ± 28    | 80 ± 39     | 3.7 ± 1.3     | 0.3 ± 0.13    |
| KR                        | 118               | 130 ± 25    | 25 ± 16     | 3.8 ± 1.1      | 14                 | 140 ± 18    | 30 ± 6      | 3.9 ± 1.6     | 0.4 ± 0.15    |
| LA                        | 34                | 167 ± 32    | 56 ± 56     | 3.4 ± 1.2      | 51                 | 163 ± 50    | 59 ± 12     | 4.1 ± 2.5     | 0.36 ± 0.27   |
| Sum                       | 447               | 159 ± 39    | 51 ± 50     | 3.2 ± 1.3      | 795               | 171 ± 36    | 55 ± 45     | 3.7 ± 1.5     | 0.31 ± 0.16   |
| Clearcut reference lakes  |                   |             |             |                |                   |             |             |                |
| SV                        | 268               | 150 ± 25    | 37 ± 17     | 4 ± 1.7        | 144               | 142 ± 18    | 31 ± 14     | 5.4 ± 2.9     | 0.28 ± 0.09   |
| EMP lakes                 | 403               | 176 ± 20    | 56 ± 17     | 5 ± 1.9        | 616               | 170 ± 22    | 52 ± 17     | 5.3 ± 1.9     | 0.21 ± 0.12   |
| EMP Total                 |                   |             |             |                |                   |             |             |                |

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**Table 3**

Summary of measured stable isotopes of fish muscle samples (length ≥ 100 mm) from the investigated lakes before and after forest harvest (Mean ± SE).

| Lake                      | Before forest harvest (2010–2011) | After forest harvest (2013–2015) |
|---------------------------|----------------------------------|----------------------------------|
|                           | N | $\delta^{13}C$ | $\delta^{15}N$ | N | $\delta^{13}C$ | $\delta^{15}N$ |
| Clearcut lakes            | 21 | -31.43 ± 0.29 | 7.62 ± 0.13 | 160 | -30.53 ± 0.06 | 7.37 ± 0.03 |
| BK                        | 9  | -26.62 ± 0.49 | 8.1 ± 0.22  | 119 | -27.87 ± 0.23 | 8.54 ± 0.15 |
| GA                        | 115 | -27.5 ± 0.11  | 5.1 ± 0.09  | 195 | -29.31 ± 0.11 | 6.45 ± 0.09 |
| KR                        | 64  | -30.09 ± 0.07 | 7.67 ± 0.05 | 14  | -30.05 ± 0.04 | 7.81 ± 0.05 |
| LA                        | 10 | -31.79 ± 0.09 | 7.5 ± 0.16  | 42  | -32.22 ± 0.06 | 7.49 ± 0.09 |
| Modelling total           | 219 | -28.69 ± 0.12 | 6.39 ± 0.09 | 530 | -29.82 ± 0.08 | 7.17 ± 0.06 |
| Clearcut reference lakes  | 84  | -31 ± 0.04    | 7.03 ± 0.09 | 112 | -30.62 ± 0.07 | 6.85 ± 0.06 |
| Modelled                 | 84  | -31 ± 0.07    | 6.87 ± 0.07 | 112 | -30.64 ± 0.06 | 6.86 ± 0.06 |
change in large fish in Björntjärn (−2%, \( p > 0.95 \)) or Gårdsjön (−2%, \( p > 0.99 \)). In the clearcut reference lakes Svultentjärn there was a slight increase (5%, \( p = 0.05 \)). The change in fish [Hg] from Lake Långtjärn was dependent on fish size, with increases in the [Hg] of large fish, but a decrease in small fish (SI Table S7).

In 2014, two years after the forest harvest, the situation was different: there was a weak significant increase in large fish [Hg] from Lake Björntjärn (16%, \( p = 0.1 \)), and a much more significant increase in Lake Brobo-Kroktjärn (15%, \( p = 0.02 \)) compared to before forest harvest (Fig. 3, SI Table S7). On the contrary, there was a significant decrease in large fish [Hg] from Lake Gårdsjön (−14%, \( p = 0.004 \)) and no significant change in Lake Långtjärn (19%, \( p > 0.8 \)) (Fig. 3, SI Table S7). At the same time, the clearcut reference lake Svultentjärn's large fish [Hg] increased by 49% in 2014 compared to before forest harvest (\( p < 0.0001 \)). No fish were successfully sampled from Lake Kroktjärn after 2013.

When we sampled again in 2015, three years after forest harvest, the [Hg] of large fish were significantly higher in Lake Björntjärn (29%, \( p = 0.007 \)) and Gårdsjön (15%, \( p < 0.05 \)), compared to before forest harvest (Fig. 3, SI Table S7). The fishing failed to collect sufficient samples from both Lake Brobo-Kroktjärn and Lake Kroktjärn in 2015. An increase of 21% in large fish [Hg] from Lake Långtjärn was noticed when compared to 2010–2011, yet it was insignificant (\( p > 0.45 \)). Meanwhile, there was no significant change (6%, \( p > 0.15 \)) in large fish [Hg] from the clearcut reference lake Svultentjärn (Fig. 3, SI Table S7).

The small fish (length < 100 mm) [Hg] data from both before and after forest harvest was only available in three clearcut lakes (SI Table S7). Of these, only Lake Långtjärn showed a significant change, a 38% decrease after harvest when compared to small fish [Hg] 2010–2011, even though the large fish in Långtjärn increased by 121% that same year.

### 3.4.2. Clearcut lakes versus EMP lakes

Using GLM2 (Eq. 3), the temporal change in fish from lakes subject to forest clearcut in this study was further compared to the general temporal change in small headwater lakes without forest harvest in their catchments that are sampled by nationally coordinated monitoring programs (EMP lakes). GLM2 proved no significant interaction effect (\( \gamma \)) between time point (before/after forest harvest) and log fish weight, thus we removed the \( \gamma \) term from further modelling.

In 2013, one year after the forest harvest, both clearcut lakes and unharvested EMP lakes had significant [Hg] increases in the large fish (length ≥ 100 mm) [Hg] (Clearcut lakes: 16%, \( p = 0.0001 \); EMP lakes: 8%, \( p = 0.002 \)) (Fig. 4, SI Table S8). Two years after the 2010–2011 period, there was 7% higher [Hg] (\( p = 0.1 \)) in large fish [Hg] from the clearcut lakes in 2014, while there was no significant changes for the EMP lakes (5%, \( p > 0.35 \)) (Fig. 4, SI Table S8). The 2015 large fish [Hg] were 26% (\( p < 0.0001 \)) higher in the clearcut lakes compared to the pre-harvest period (Fig. 4, SI Table S8). The large perch [Hg] within EMP lakes decreased 9% over the same period, while there were no significant change in large fish [Hg] in the clearcut reference lake Svultentjärn. Though the large fish [Hg] increased in EMP lakes during the first year after 2012, the fish [Hg] returned to the earlier level a

![Fig. 2. The trophic magnification slope (TMS) or slopes (b) of ln[Hg] vs. \( ^{13} \text{C} \)N regression in fish samples with length over 100 mm during the investigated years (2010–2011 indicated that the lakes were sampled either one or two years prior to scheduled forestry activities).](image)

![Fig. 3. Predictions of fish [Hg] in clearcut lakes (includes BJ, BK, GA, KR, and LA) and clearcut reference lake (SV) before (2010–2011; in blue pillars) and in the individual years after forest harvest. Dashed lines indicate the 2010–2011 levels of fish [Hg] concentrations in corresponding lake groups. Predicted fish [Hg] was estimated based on least-square means and standard errors from GLM1 (Eq. 1) for each lake for large (length ≥ 100 mm) fishes, in the following order from left to right for each lake: Before forest harvest in 2010–2011; One year after forest harvest in 2013; Two years after forest harvest in 2014; Three years after forest harvest in 2015. Numbers in brackets indicate sample numbers.](image)

![Fig. 4. Boxplots with median and ±1.5 interquartile ranges of predicted fish [Hg] (predicted from GLM2) in clearcut lakes (red boxplots) and EMP lakes (blue boxplots) in the time before forest harvest (2010–2011) and in each individual year afterwards. Predictions of the temporal changes were done for large (length ≥ 100 mm) fishes, in the following order from left to right for each lake: Before forest harvest in 2010–2011; One year after forest harvest in 2013; Two years after forest harvest in 2014; Three years after forest harvest in 2015. Red dashed line and blue solid line are linear interpolations of the responses of fish [Hg] in clearcut and EMP lakes, respectively.](image)
year later, i.e. in 2014 the [Hg] of large fish in the EMP lakes was not signifi-
cantly different from the 2010–2011 level. In the small fish (length < 100 mm) [Hg] there was no significant change in either the clearcut or the EMP lakes (Clearcut lakes: −12%, p > 0.2; EMP lakes: − 4%, p > 0.8) (SI Table S8).

4. Discussion

The overall trend detected in the GLM2 model was a significant 16% increase in the [Hg] of large perch in the first year after the harvest, a weak but yet significant increase of 6% in the second year, and then another highly significant increase of 26% three years after harvest, all compared with before harvest [Hg] levels (Fig. 4). The annual changes in perch [Hg] during the three-year period (across lakes: −14–121% in large fish, and −38–23% in small fish, according to GLM1) after forest harvest were often outside the range of an earlier regional estimate sug-
gest that 10–25% of Hg in Swedish freshwater fish is a consequence of forest harvest (Bishop et al., 2009). The values provided by Bishop et al. (2009), however, are regional, long-term averages, based primarily on observed water chemistry. Additionally, the estimation from Bishop et al. (2009) was based on two assumptions: harvest effects persisted for a decade and that about 10% of the landscape at any given time was affected by a clearcut within the past decade. While the present study did not cover a decade after harvest, the extent of clearcut in the catchments of the impacted lakes was >10% in four of the five study lakes.

The degree of perch [Hg] fluctuation in lakes subject to forest clearcut was consistent with a review of studies on how forestry affects [Hg] in runoff (Eklof et al., 2016). That review found that for responses to forest harvest in runoff concentrations, there was a large degree of variation with a number of “no” or “low” forestry effects being reported along-side reports of larger effects for both MeHg and total Hg. In our clearcut lakes, there was a low degree of response in some catchments, and a larger response in some, at some times, most notably in the large perch [Hg] in the first year after the forest harvest. This inter-lake and inter-year variability is likely due to both spatial and temporal variation in weather, runoff, and input of nutrient and contaminants from forest streams to lakes (Allan et al., 2009; de Wit et al., 2014).

The significant increase in fish [Hg] in the clearcut lakes for several years after the forest harvest, which contrasted with the general trend in the clearcut reference and EMP lakes, implies that we cannot reject the hypothesis of forest harvest impact on fish [Hg] (Fig. 4). Three years after forest harvest in 2012, the fish [Hg] in the clearcut lakes had increased by 26% for large fishes, compared to the period before forest harvest (GLM2, SI Table S8, Fig. 4). Our study suggests that the area of the catchment subject to forestry operations may influence the degree of response in the accumulation of Hg in fish. According to model (GLM1) prediction, the two largest increases in large perch [Hg] one year after the forest harvest occurred in Lake Långtjärn (121%) and Kroktjärn (43%), which were also the lakes with the largest proportion of harvested area in the lake catchments (40% and 36%, respectively). However, while the [Hg] of large fish in Lake Långtjärn during the sub-
sequent two years (2014 and 2015) remained larger than before harvest, the relative increase was much smaller. The variation seen in the response in fish [Hg] levels over time in Lake Långtjärn strengthens the view that effects from forest clearcut needs to be followed over long periods.

No increase in the [Hg] of small fish after forest harvest was observed in this study, possibly due to changes in food-web structure after forest harvest. Increased total phosphorus and DOC (Carignan et al., 2000; Winkler et al., 2009), as well as higher zooplankton abundance (Leclerc et al., 2011) have been observed in forestry impacted lakes. For small perch, which feed primarily on zooplankton (Rask, 1986), no significant change in fish [Hg] was found within our clearcut lakes dur-
ing the first year after harvest in 2012 according to GLM1 and GLM2. Leclerc et al. (2011) demonstrated that higher prey availability of zooplankton abundance elevated growth rate of age-0 yellow perch in Canadian Shield Lakes two years after forest harvest. We therefore suspect that the absence of a significant increase in smaller fish [Hg] shortly after forest harvest might be linked to increased feeding success and fish growth rate, a scenario similar to biodilution (Rypel, 2010). As a result, the expected elevation in fish [Hg] was mitigated in small fish. The in-
creased Hg in large fish could also be influenced by processes besides harvest-related mobilization of Hg from soils to surface waters, particu-
larly changes in the food web structure. This is yet another contribution to the complexity of studies that aim at a functional connection between catchment impacts, runoff outputs, and Hg accumulation in aquatic biota.

The decreased δ13C values in fish muscle samples from the clearcut lakes, together with an increase in fish sizes (length and weight) is an indication of a change in fish feeding habits, e.g. fish might have mi-
greated more offshore to the lake pelagic zone as compared to before for-
est harvest, resulting in increased autochthonous carbon input from zooplankton (Fry, 1983; Syväranta et al., 2008) or lower-trophic-level organisms of periphyton (Pouilly et al., 2013). The increasing δ13C values in the clearcut reference lake Svultentjärn that were accompa-
nied by a decrease in fish size, contrasted with the response in the clearcut lakes, implied that fish were likely feeding more on autochtho-
nous carbon with increasing Hg exposure from terrestrial sources. The increased δ15N values in fish from the clearcut lakes after forest harvest also indicated a possible trophic shift in the lakes. Changes of Hg TMS in our studied lakes derived from the δ15N values, implied a connection be-
tween trophic shifts and related fish [Hg] change (Lavoie et al., 2013). Lake Långtjärn had the most drastic change in terms of TMS (shifting from negative to positive) in large fishes (up to 200% increase in TMS from 2013 as compared to 2010–2011, Fig. 2), one year after forest har-
vest. This indicated a large shift of Hg bioaccumulation efficiency, which is consistent with the 66% increase of fish Hg in Långtjärn during that same period. While a tempting speculation, we still lack direct evidence from zooplankton stable isotope shifts proving that this change in the trophic structure is related to forest harvest activities.

Our study did not show any drastic changes in water chemistry after forest harvest, but with one water sample each year, there may have been seasonal changes in water chemistry that were not captured by only having one sampling each year. Many uncertainties remain regarding water chemistry changes after forest harvest related to various hy-
drological patterns: it takes at least 11% forest clearcut in catchment area for Schelker et al. (2014) to be able to detect significant changes in TOC observed from first-order streams; and there is often a several-
year delay before observing increasing nitrate effects in post-harvest catchments (Futter et al., 2010). There were however, evidence of water chemistry and fish [Hg] linkages from the reference lake Svultentjärn and our 19 EMP lakes dataset, with lake TOC and fish [Hg] as the most significant positive correlation (SI Table S6). It has been suggested that higher TOC concentrations promote formation of MeHg and facilitate more efficient Hg bioaccumulation (Driscoll et al., 1995). Långtjärn had the lowest pH among all clearcut lakes, and the highest TOC, both of which may have enhanced Hg bioavailability from water to biota. Another possibility is that increased Hg loadings in runoff that contributed to Hg methylation and bioaccumulation, com-
bined with increasing water yield and therefore total flux of Hg from the catchment to lake after forest harvest (Allan et al., 2009). On the other hand, the positive correlations of nutrients (totP, totN, and Chl a) with fish [Hg] in the EMP lakes were puzzling (SI Table S6), for this contrasted with previous findings on Hg biodilution due to increased nutrient input (Karimi et al., 2007; Liu et al., 2012). This indicated a sce-
nario of enhanced Hg bioaccumulation from food web base under im-
proved nutrient status in the boreal oligotrophic lakes, as demonstrated by MacMillan et al. (2015) and Soerensen et al. (2016).

It is likely that the changes in fish [Hg] in clearcut lakes could not be attributed entirely to an effect from forest harvest on the concentration and loading of Hg in runoff from the catchment. While clearcuts and/or
subsequent forest harvest activities (e.g. site preparation) often do exacerbate Hg concentrations and loadings in runoff from the terrestrial to the aquatic environment, the degree of change in Hg loadings and aqueous concentration varies considerably, and is sometimes not present at all (Eklöf et al., 2016). Even when there is increased loading of Hg to lakes following forestry practices, the biomagnification of Hg in biota are determined by numerous other factors, including indirect effects of the forest harvest on feeding behavior and food web structure, but also weather induced changes on both water quality and food webs (Rask et al., 1998; Rask et al., 2007).

5. Conclusions

This study detected a change in perch [Hg] in lakes subject to forest harvest (with 6–40% of the catchment harvested) that resulted in varying year to year responses during the first three years post-harvest. Generally, there was an overall increase of perch [Hg] in lakes that were subject to forest clearcut in the lake catchment. These responses of perch [Hg] change were much more varied than the previous prediction for the region. The high variation in [Hg] over time and between lakes in this study suggest three findings that are important for the evaluation of forest effects on Hg accumulation in aquatic biota: First, the complexity of understanding how forest harvest activities impact fish [Hg] is related to changes in the food web. Secondly, the effects from forest harvest on fish [Hg] need to be evaluated over a long period to account for the large natural year to year variation in fish [Hg]. And thirdly, trophic shifts of fish communities towards more autochthonous feeding triggered by forest harvest activities can be identified through stable isotope measurements. This study adds to the evidence that forestry operations not only mobilize and transform Hg from forest soils, but also adds to the Hg in fish. There are, however, many factors that need to be weighed when balancing between production and environment in forestry. The effect of forestry on fish Hg concentrations is one of the considerations in this balancing act. Further work on linking catchment characteristics and pollutant concentrations in the environment, including more long-term investigations of the biota in response to perturbations would help increase our understanding and ability to assess forestry, as well as land-use and climatic impacts on the Hg in fish.

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Appendix A. Supplementary data

Testing fish size classification (SI 1a, Fig. S1, S2, S3 and S4, SI-2); Testing fish length-weight relationship (SI 1b, Fig. S5 and S6, SI-4); Summary of catchment size and coordinates of 19 EMP lakes (Table S1, SI-5); Analytical methods, uncertainty, detection range and instruments used for the analyses of the chemical variables included in the data evaluation (Table S2, SI-6); Summary of water chemistry parameters in clearcut and clearcut reference lakes (Table S3, SI-7); Summary of water chemistry parameters in 19 EMP lakes (Table S4, SI-8 and SI-9); Summary of EMP lakes fish Hg and water chemistry correlation coefficients (Table S6, SI-10); GLM1 model predictions on temporal change in fish [Hg] (Table S7, SI-11) and GLM2 model predictions in both large and small fishes (Table S8, SI-12); Supplementary data associated with this article can be found in the online version, at https://doi.org/10.1016/j.scitotenv.2017.12.075.

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