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Forest harvest effects on mercury in streams and biota in Norwegian boreal catchments

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Abstract

Forest harvesting practices can potentially increase mercury run-off from catchments. A paired catchment experiment was conducted in a boreal forest in southern Norway, to test effects of forest harvest operations on i) concentrations and fluxes of methylmercury (MeHg), total mercury (HgT), nutrients and dissolved organic matter (TOC), and on ii) MeHg bioaccumulation in stream foodwebs.

Thirty percent of a catchment was harvested in winter time with snow cover but no soil frost, resulting in wheel tracks and soil compaction. Pre-harvest differences included higher streamwater MeHg, HgT and TOC, and lower pH in the treated catchment compared to the reference.

No significant treatment effects on concentrations of MeHg, HgT and TOC were detected, whereas concentrations of nutrients (ammonium, nitrate, phosphorus (P)) increased significantly. Estimated catchment export of nitrate and ammonium increased fourfold, as a combined effect of changed discharge and concentrations. Export of MeHg and HgT increased weakly, primarily because of an increase in discharge.

Levels of MeHg in stream invertebrates mirrored differences in aquatic MeHg between the two streams, resulting in higher MeHg in biota in the harvest catchment in pre-harvest conditions. After harvest, MeHg levels in primary consumers (herbivorous stoneflies) were no longer different between the two streams, despite continued exposure to higher aqueous MeHg in the harvested catchment. Simultaneously, dietary biomarkers ($\delta^{15}N$ signature, lipid- and algal fatty acid content) in the stoneflies had changed significantly, consistent with higher nutrient loadings and associated higher diet availability in the harvested stream.

The results of our experiment do not support that forest management has a strong impact on catchment MeHg production. Catchment disturbance through forest harvesting may decrease MeHg in aquatic biota, because of higher stream productivity and associated higher quality of dietary sources, at least in the short-term. Other studies on catchment MeHg production to disturbance have shown a range in responses, from strong to none. So far, no factor has emerged to explain such range in responses. Predictions of forest management effects on MeHg in streamwater and aquatic food webs are hampered by limited understanding of catchment controls on MeHg production.

Keywords

Forest management, catchment manipulation, methylmercury, water chemistry, bioaccumulation, primary consumers, clear-cutting

1. Introduction

Mercury (Hg) is a long-range transported pollutant of great environmental concern in boreal areas across the entire northern hemisphere. Atmospheric deposition of Hg in natural ecosystems leads to long-term accumulation of Hg in soils and wetlands, where transformations of Hg to its highly toxic organic form methylmercury (MeHg) occur with subsequent transport of Hg-species to surface waters (Grigal, 2002). MeHg is a neurotoxin with a strong tendency to bioaccumulate in food webs (Morel *et al.*, 1998). Levels of MeHg in the aquatic food web are raised to levels that are potentially harmful for fish and wildlife (Scheulhammer *et al.*, 2007) and, through consumption of fish, to human health (Mergler *et al.*, 2007).

High Hg concentrations in fish are associated with brown-water streams and lakes in forested regions with a prevalence of wetlands (Nilsson and Håkanson, 1992; Driscoll *et al.*, 2007; Chasar *et al.*, 2009). Wetlands are commonly viewed as one of the main suppliers of MeHg to aquatic ecosystems, because of high groundwater levels, creating anoxia and thereby promoting conditions for methylation of Hg (Grigal, 2003). Not just wetlands, but also forests have the potential to be a significant source of MeHg to surface waters. Coniferous forests are highly effective scavengers for atmospheric Hg species, resulting in substantially higher Hg deposition in forests compared to open areas (Graydon *et al.*, 2008; Witt *et al.*, 2009), thereby enriching forest soils with Hg. Forest throughfall has been shown to be a significant input of MeHg to boreal catchments (Witt *et al.*, 2009), possibly demonstrating an additional pathway of MeHg from forest canopies to surface waters.

Recently, forest management has been suggested to be an important contributor to catchment export of MeHg, thereby increasing MeHg in the aquatic food chain (Bishop *et al.*, 2009). Forests in northern Europe (Ostlund *et al.*, 1997) and large parts of North America (Stinson *et al.*, 2011) are landscapes where forest management practices have left a strong mark. Because of increased interest in the role of forest for climate mitigation (Jackson *et al.*, 2008), especially as a source of bioenergy (Schlamadinger and Marland, 1996), forest management might intensify. Thus, further assessment of environmental impacts of forest harvest practices is needed to protect aquatic ecosystems.

77 Forest harvesting is known to have a strong impact on catchment hydrology and nutrient
78 runoff (Likens *et al.*, 1970; Kreutzweiser *et al.*, 2008). Effects of forest harvesting and soil disturbance
79 on MeHg runoff have been shown in Finland (Porvari *et al.*, 2003) and Sweden (Munthe and Hultberg,
80 2004). However, the mechanisms controlling the increased export of MeHg are not well understood.
81 Soil disturbance with associated increases in mobilization of MeHg from soil pools has been
82 hypothesized previously as controlling mechanism (Munthe and Hultberg, 2004), in addition to
83 increased discharge, changed hydrological pathways and higher soil temperatures (Porvari *et al.*, 2003;
84 Eklof *et al.*, 2013). Still, forest operations have not lead to increases in MeHg runoff in all cases. No
85 effect of harvest operation on MeHg runoff was found in catchment manipulations in Sweden, despite
86 small increases in runoff (Sorensen *et al.*, 2009a) and extensive damage to soils from forest
87 machinery (Sorensen *et al.*, 2009a; Eklof *et al.*, 2013)
88 Another type of evidence for relations between forest management and MeHg in aquatic
89 ecosystems comes from synoptic studies. Studies of lake ecosystems in Canada indicated a connection
90 between catchment disturbance and increased levels of MeHg in the aquatic food web (Garcia and
91 Carignan, 1999, 2000; Desrosiers *et al.*, 2006; Garcia *et al.*, 2007). Here, increased levels of MeHg in
92 aquatic biota and periphyton were related to catchment disturbance, while aqueous dissolved MeHg
93 was not investigated. Further, significant relations between MeHg in aquatic biota and dissolved
94 MeHg in waters were found by Hall *et al.* (2009) in Canadian flooded reservoirs, and by Chasar *et al.*
95 (2009) in a synoptic study of stream foodwebs in the United States. Thus, relations between catchment
96 disturbance and enhanced levels of MeHg in biota have been implied, but are not well-documented. In
97 addition, the limited number of studies and lack of consistent responses of forest management on
98 MeHg export indicate a strong need for a better understanding of processes underlying catchment
99 MeHg production from experimental settings.
100 In order to test the hypothesis that forest harvest i) increases streamwater MeHg and total Hg
101 concentrations and runoff, and ii) enhances MeHg concentrations in biota, we conducted a paired-
102 catchment study in a Norwegian boreal forest. Streamwater chemistry, hydrology and levels of MeHg
103 in stream invertebrates were investigated, including links between diet and bioaccumulation of MeHg.
104

2. Materials and Methods

2.1 Site description

The Langtjern study area is located in southeast Norway (60°37' N, 9°73' E) at 500 to 710 m elevation approximately 80 km northwest of Oslo (Figure 1). The Langtjern lake catchment is part of the national monitoring programme for effects of acid deposition and consists of two inlet streams and a lake outlet, where streamwater chemistry and discharge have been monitored since 1972. The eastern inlet stream catchment (LAE03) was used as the reference catchment. The treatment catchment (LAE11) is located 1.5 km southeast of LAE03, adjacent to the lake catchment, and is slightly less than one-third of the size of the LAE03 catchment (Table 1).

Mean annual discharge from the Langtjern lake outlet between 2008 and 2011 was 702 mm, while mean annual precipitation and temperature were 914 mm and 4.5 °C, respectively (nearby meteorological station Gulsvik II, 132 m elevation, 60°38' N, 9°60' E). Wet sulphur (S) deposition was 5 kg S ha^{-1} in 1990 (Larssen, 2005) and 3 kg S ha^{-1} in 2000 (De Wit *et al.*, 2007) respectively and is still declining.

The vegetation at Langtjern is dominated by low- to unproductive Scots pine forest (*Pinus sylvestris* L.), interspersed with peatlands (both forested and open Sphagnum mires) and patches of Norway spruce (*Picea abies* (L.) Karst.) forest. The stands are mature or close to maturity. The geology consists of till of felsic gneisses and granites, where thin mineral soils have developed. Deeper peaty soils are found, being most abundant close to streams. The area proportion of main forest- and vegetation types is similar in the two catchments, the most notable difference being a higher percentage of forested peatland (forest on peat soils of at least 30 cm depth) in the LAE11 catchment. In LAE11, pre-treatment volume proportions of Scots pine, Norway spruce and birch were 57%, 34% and 9%, respectively, while the corresponding numbers were 62%, 35% and 3 % in LAE03. LAE03 and LAE11 had a stocking of 78 and 62 m^3 ha^{-1}, respectively. These volumes illustrate the low site productivity of the study area.

2.2 Experimental design and harvest operation

The paired catchment experiment consisted of two small forested catchments, the reference (LAE03) and the treatment catchment (LAE11). Monitoring started in June 2008. The forest harvest operation in the LAE11 catchment was conducted from January 13 to 16 in 2009. Forest standing volume, water chemistry, discharge and aquatic biota were monitored before and after the harvesting operation. The choice for the timing of the harvest operation and thereby the length of the pre-harvest treatment was partly based on the original period of project funding, i.e. three years.

The harvesting operation was done using the 'cut to length' method (harvester and forwarder). The impacted area was confined to the lower and middle part of the LAE11 catchment, affecting about 30 % of the catchment area and with a harvest removal corresponding to 38 % of total catchment tree volume. As the forwarder would have to cross several areas with limited bearing capacity on its route between the harvested area and the landing site the harvesting operation was scheduled to take place in winter, when the soil was expected to be frozen. However, due to mild weather prior to harvest, the soil was not frozen. Snow depth was circa 20 cm during harvesting. Thus, harvesting resulted in local soil disturbance in the form of wheel ruts. This was most pronounced along the main extraction tracks and in wetter parts adjacent to the stream in the lower part of the catchment, while the upland parts of the catchment area were less affected. Norwegian guidelines for harvesting close to streams and mires required leaving a buffer zone adjacent to the mire in the central part of the catchment where the buffer zone corresponds with the stream course. With this exception, only scattered trees were retained on the impacted area. Equal volumes of Scots pine and Norway spruce were harvested, whereas birches which only occurred as scattered individuals were mostly retained.

2.3 Hydrology

V-weirs were installed in the summer of 2008 in the streams of LAE03 and LAE11 for quantification of discharge. Comparison of stream flow estimates at the Langtjern catchment lake outlet indicated that these v-profiles did not supply data of sufficient quality for quantification of stream flow, primarily due to leakage and overflow under high flow conditions. However, the v-profiles did provide information on the variation in discharge in both catchments from 2008 until 2010, and indicated

158 synchrony in high flow events and low flow periods in both catchments and thus, similar to
159 hydrographs. Discharge from the LAE03 catchment was estimated instead based on a full water
160 balance for the entire lake catchment (Figure 1), based on daily discharge in the outlet, temperature
161 data from a nearby weather station and additional hydrological measurements made during the 1970s
162 (Wright and Henriksen, 1980).

163 Water levels in Lake Langtjern may vary with around 60 cm, resulting in variation in lake
164 water storage. Because of this, the discharge of the inlet and outlet do not follow the same pattern and
165 the inlet discharge cannot be derived by simple area-scaling of the outlet discharge. A simple water
166 balance model was used to adjust the impact from lake water storage:

167 where

168 R is total runoff to the lake, calculated from an empirical relation between the change in water
169 storage (ΔS) in the lake and discharge in the outlet (Q), and corrected for precipitation directly on the
170 lake (P) and evapotranspiration from the lake (E). Details on the calculation procedure are given in
171 Wright and Henriksen (1980). In short, ΔS was calculated from an empirical relationship between lake
172 water level (available from weekly measurements between 1976 and 1978) and outlet water level (m)
173 in the stilling pond before the v-profile in the outlet. E was estimated assuming and evaporation of
174 0.15 mm day^{-1} per °C daily temperature (Lundquist, 1977). Precipitation and daily temperature were
175 derived from nearby meteorological stations. The total runoff (R, in mm) to the lake was assumed to
176 be representative for the subcatchment LAE03.

177 Discharge from the LAE11 catchment was estimated using area-corrected discharge from
178 LAE03 for the pre-harvest period. Post-harvest discharge in LAE11 was based on hydrological effects
179 of harvest in a paired-catchment experiment in in northern Sweden (Sorensen *et al.*, 2009b). The
180 catchments were covered by coniferous forest and included 3 to 18% wetland, with a slightly colder
181 and drier climate than our study site. In the Swedish study, two catchments were partially clear-cut,
182 where 30% and 71% of the catchment areas were logged. Hydrological responses after harvest
183 between the two catchments were similar. Thus, the Swedish catchments were similar in land cover

and forest harvest, compared to our study site which was also partially clear-cut. Discharge (compared to a reference, non-harvested catchment) started to increase after July and resulted in a 35% increase in mean annual runoff. The increase was +45% during base flow conditions (<1 mm day^{-1}); +27% during intermediate flow (1-5 mm day^{-1}) and +25% for high flow conditions (>5 mm day^{-1}). Daily discharge in LAE03 (in mm day^{-1}) was adjusted according to these %-wise changes in flow, from August 2009 onwards.

2.4 Stream water sampling

Streamwater grab samples for acid-base chemistry were collected biweekly or monthly, according to procedures established in the acid monitoring programme (SFT, 2009). Samples were sent to the Norwegian Institute for Water Research (NIVA) by mail and processed at the NIVA accredited laboratory. From 2008 until December 2011, streamwater grab samples for analyses of total Hg (HgT) and MeHg were collected using 125 mL acid-leached Teflon bottles. The bottles were packed in two plastic bags for ultra-clean handling (USEPA, 1996). Samples were sent to NIVA by mail and forwarded to the Swedish Environmental Research Institute (IVL) in Gothenburg. There were usually 4 to 6 days between sampling and preservation with 0.5 ml 37-38% HCl (Baker). From October 2010, streamwater samples were taken by another procedure, using 250 mL fluorinated polyethylene (FLPE) bottles and sent for analysis at NIVA. HCl (concentrated trace level grade, 1 mL) was added to the MeHg bottle to yield a 0.4 % solution during sampling. Water for HgT analysis was sampled in a separate bottle, to which BrCl (bromo monochloride) was added as oxidising agent within 2 days after arrival to the laboratory. All bottles for Hg species determination were packed in two plastic bags for ultra-clean handling. The number of samples for Hg species and acid-base chemistry taken during the pre- and post-harvest period is given in Table 2.

2.5 Sampling of biota

Streamwater biota was collected from one sampling station in each stream, at October 17 2008, May 29 2009 and on October 16 2009, thus collecting samples that reflected summer and winter conditions. Each sampling station comprised a stream reach of 30 m length, and was located in the lowermost

parts of each catchment. Biofilm was carefully removed from streambed rocks, concentrated by centrifugation and kept in separate glass vials. Macroinvertebrate species composition of the two streams was identical, and comprised two herbivorous stoneflies (nymphs of *Nemoura cinerea* and *Nemurella pictetii*) and one predatory caddisfly (larvae of *Plectrocnemia conspersa*). They were collected by kick-sampling, following the Norwegian Standard Method (EN 37828, 1994), using a hand net (25 x 25 cm opening; 250 µm mesh size). Due to small body size, stoneflies were pooled to samples of 50 to 80 individuals, to obtain enough material for chemical analysis. All samples were shock frozen (-80˚C) in the field, transported to the laboratory, freeze-dried and analyzed for MeHg, stable isotopes ($\delta^{15}N$) and fatty acids. Species identification was conducted by the biological accredited lab at the NIVA.

2.6 Analysis of water chemistry major ions

Analyses of pH, conductivity, major anions and cations, total N, total organic C (TOC), Al species, total P and UV absorbance (at 254 nm) ($UVabs_{254}$) were performed at accredited laboratories at NIVA. Total organic C was analysed by wet chemical oxidation IR-detection (EPA accredited method nr. 415.1). The samples were purged prior to analysis so only non-purgeable organic carbon was measured. Organic carbon in a sample was converted to carbon dioxide by wet chemical oxidations. The carbon dioxide formed was measured directly by an infrared detector. Total organic carbon (TOC) consisted of ca 95% DOC (filtered by 0.45µm) in the LAE03 stream. Specific absorbance at 254 nm ($SUVA_{254}$) was calculated by dividing $UVabs_{254}$ with TOC.

2.7 Mercury analysis

Two laboratories were involved in determination of HgT and MeHg; IVL (2008-2011) and NIVA (2011-2012). Both laboratories follow United States Environmental Protection Agency (USEPA) Method 1630 (USEPA, 1998) for determination of MeHg in water by distillation, aqueous ethylation, purge and trap, and cold vapor atomic fluorescence spectrometry (CVAFS). For HgT, USEPA Method 1631 for determining Hg in water by oxidation, purge and trap and CVAFS was followed (USEPA, 2002). The method detection limits (MDL) were 0.02 ng/L (NIVA) and 0.06 ng/L (IVL) for MeHg,

and 0.1 ng/L for HgT (3 standard deviations of blanks). The IVL laboratory determination of Hg species was done from one bottle (see sampling procedures). Analysis proceeded by the removal of a sample aliquot for determining MeHg, before BrCl (bromo monochloride) was added as oxidising agent and the remainder of the sample used for determination of HgT. The NIVA laboratory followed the same procedure, but samples for MeHg and HgT analysis were taken in two separate bottles.

For over a year (October 2010 to November 2011), parallel samples for several locations, including the streams in this study, were run at both laboratories. The IVL laboratory reported significantly higher ($p<0.05$) HgT concentrations than the NIVA laboratory (Braaten *et al.*, 2014). Braaten et al. (2014) show that difference in HgT is related to the removal of the aliquot for MeHg from the bottle used for both MeHg and HgT analysis, and re-dissolution of HgT species that adhered to the bottle surface. For the LAE03 and LAE11 streams, HgT from IVL was on average 12% higher than from NIVA in the parallel sampling period. Where analytical results were available from both laboratories, we used the value from the NIVA laboratory as default. We tested whether the outcome of the statistical tests (statistical methods described further below) of treatment effects on MeHg and HgT streamwater chemistry was affected by i) using IVL results instead of NIVA results for the parallel sampling of treatment effects, and ii) the change in laboratory, by downwards adjusting IVL results of HgT with 12% from 2008 to September 2010 (see Supplementary Information). The results of the statistical tests were not affected.

Biological samples were treated with hot methanolic potassium hydroxide solution for about 3-4 hours. The samples were then diluted with methanol, separated by ethylation and detected following the same procedure as described for water samples (see above). The typical detection limit was 1.5 ng g^{-1} in MeHg analysis. Reference materials NIST 2977 (Mussel Tissue) and DORM-2 (Dogfish muscle) were used, and recovery was 104% and 100%, respectively.

2.8 Stable isotope, lipids, and fatty acids analyses

Stable nitrogen isotopes ($\delta^{15}N$) of biota were analyzed after transferring freeze-dried samples (1 mg) to tin capsules and combusted in a Eurovector element analyzer. The N_2 was directly injected online to a Nu Instruments Horizon, Isotope Ratio Mass Spectrometer (Wrexham, UK) for determination of $\delta^{15}N$.

Lipids were extracted from freeze-dried (96 hours) samples using chloroform:methanol (2:1) as described elsewhere (Heissenberger *et al.*, 2010). Fatty acids were esterified from total lipid extracts to obtain fatty acid methyl esters (FAME) using toluene (1 mL) and H_2SO_4-methanol (2 mL; 1% v/v). Subsequently, FAME were analysed using a gas chromatograph (TRACE GC THERMO) equipped with flame-ionization detection, a temperature-programmable injector and an autosampler. A Supelco™ SP-2560 column (100 m, 25 mm i.d., 0.2 µm film thickness) was used for FAME separation. Excalibur 1.4™ was used for calculation and, if necessary, manual resetting of the chromatograms. Fatty acid concentrations were calculated using calibration curves based on known standard concentrations. Fatty acids were grouped to characterize bacterial fatty acids (BAFA; i.e., the sum of odd saturated and branched-chain FA: 15:0 and 17:0 and their *iso* and *anteiso* series), algal fatty acids (PUFA; i.e., the sum of polyunsaturated fatty acids) as previously presented (Sun *et al.*, 2000; Kainz *et al.*, 2002).

2.9 Calculation of catchment element and nutrient export

Catchment export of elements and nutrients was calculated by linear interpolation of streamwater concentrations of elements to daily concentrations and multiplying with daily discharge, and summed to monthly fluxes.

2.10 Statistical analysis

Random Intervention Analysis (RIA) was used to analyse treatment effects on water chemistry (Carpenter *et al.*, 1989). For a time series of any given variable, paired differences between the reference and the treated catchment were calculated. The resulting time series of *catchment differences* were used to test the effect of the intervention, by comparing *differences in catchment differences* before and after the intervention. This was done by random resampling (n=2000) values of *catchment differences* from the pre-harvest period and the post-harvest period (for one year at a time, and for the entire post-harvest period) and generating new time series. The mean values of 2000 resampled time series were calculated for the pre-harvest and selected post-harvest period, and compared to the statistical distribution of the original time series to determine whether significant treatment effect had

occurred. If the value of the mean *catchment difference* of the original time series before and after the intervention was outside a certain percentile range of the *catchment difference* of the generated time series before and after intervention, we assumed that a non-random effect had occurred as a result of the treatment. The percentiles were 5% and 95% for p=0.10, 2.5% and 97.5% for p=0.05, etcetera. We did not constrain the resampling period to take into account seasonal variation as done in a previous paired-catchment study (Lofgren *et al.*, 2009) by allowing resampling only for a limited number of months for any number of years after the given observation. A test on the effect of sampling interval showed that length of sampling interval did not affect the outcome of the analysis.

Pair-wise differences between sample locations (streams LAE03 and LAE11) were tested for concentrations of MeHg, lipid content, stable isotopes and fatty acids in invertebrates and biofilms using Student's t-test.

3 Results

3.1 Streamwater chemistry

During the pre-harvest period, both streams had a water chemistry signature typical for small inland acidified catchments with base-poor soils interspersed with peatlands, i.e. a pH below 5, low conductivity (1-3 µS cm^{-1}), high TOC (14-22 mg C L^{-1}), low base cation concentrations (Ca < 1 mg L^{-1}), low SO_4 concentrations (<1 mg L^{-1} SO_4-S) and low nutrient concentrations (inorganic N-species and total P < 10 µg L^{-1}) (Table 2, Figure 2). At almost each sampling occasion in the pre-harvest period, concentrations of MeHg, HgT, TOC, totN and TON and conductivity were higher in the LAE11 (experimental) stream than in the reference stream while pH was lower. By contrast, concentrations of base cations, SO_4, species of inorganic N and totP were similar in both streams. The pre-harvest differences in streamwater chemistry were probably related to the higher contribution of peatland in the experimental catchment which lead to more TOC and associated elements. High TOC is commonly associated with a decrease in pH as dissolved organic matter is a weak acid.

Concentrations of MeHg varied most in the experimental catchment (LAE11), with winter peak concentrations exceeding 0.5 ng L^{-1}. Interestingly, the highest MeHg concentrations in the LAE11 stream appeared each winter, although by 2011-2012 the winter peak was modest. The peak in MeHg concentration in 2009 started prior to the harvest operation. In the reference stream (LAE03), MeHg varied little from around the detection limit up to 0.2 ng /L with a tendency towards higher concentrations in summer. In both catchments, MeHg was lowest during snowmelt. Less than 10% of the variation in MeHg was explained by TOC in the LAE03 stream, while no significant relation between MeHg and TOC was found in LAE11 (LAE03: r^2= 0.08, p<0.01; LAE11: r^2= 0.0, p>0.5). This is also illustrated by the large variation in MeHg to TOC ratio (Figure 2).

Seasonal patterns in TOC concentrations were very similar in both catchments, with highest TOC in summer and lowest during snowmelt (Figure 2). Concentrations of HgT were closely correlated with TOC in both catchments (LAE03: r^2 = 0.51, p<0.0001; LAE11: r^2=0.29, p<0.0001), and HgT was usually highest in September and lowest in early winter. The HgT to TOC ratio was

within the same range (roughly 0.15 to 0.35 µg g^{-1}) in both streams and showed similar temporal variation.

Contrary to our hypothesis, no significant effects of the harvest treatment were found for any comparison of pre-harvest period and post-harvest years for streamwater concentrations of MeHg and HgT ($p>0.1$; Figure 2, Table 2). The ratio of mean MeHg in the LAE11 and LAE03 streams was 2, respectively 2.5 in the pre-harvest and post-harvest periods, respectively, possibly indicating a small but non-significant response to treatment.

Nitrate concentrations showed a strong and significant ($p<0.01$) response to harvest, with peak NO_3-N concentrations between 60 and 120 µg L^{-1} in LAE11, while peak concentrations in LAE03 were between 20 and 50 µg L^{-1} (Figure 2; Table 2). Peak concentrations of NH_4-N in the post-harvest period in LAE11 exceeded 100 µg L^{-1}, while NH_4-N in LAE03 was below 40 µg L^{-1}. Total P concentrations in LAE11 were on average almost twice as high as in LAE03 (9 and 5 µg L^{-1}, respectively) in the post-harvest period, while in the pre-harvest period total P concentrations in both streams differed less (7.0 and 5.5 µg L^{-1}, in LAE11 and LAE03 respectively). There was a significant ($p<0.005$) effect of treatment on total P in 2010 only. Organic N (TON), but not total N, showed a weak increase ($p<0.1$) after harvest in 2009 and 2011, going from 284 to >320 µg L^{-1} in LAE11, while TON was <250 µg L^{-1} and remained stable in the reference stream. A consistent and significant ($p<0.05$) treatment effect was found for the CN ratio of DOM in each year of the post-harvest period, where CN ratios in LAE11 decreased from 72 to 57, almost equal to the CN ratio in LAE03 of 54. The decrease in CN ratio suggested enrichment of N in DOM after harvest. No treatment effects were found for $UVabs_{254}$ or for specific UV-absorbance ($SUVA_{254}$). Cation concentrations that increased significantly after harvest were K ($p<0.05$) and Ca ($p<0.1$) while pH showed a weak but significant ($p<0.1$) decline in 2009 and 2010.

3.2 Hydrology

The effect of forest harvest on run-off was estimated using results from a paired-catchment study of forest harvesting in a site with comparable land cover and climate as Langtjern. Only qualitative observations of higher ground water levels and wetter soils after harvest, in the harvested catchment

compared to the reference, were available. Mean annual discharge in the reference catchment LAE03 from 2009 to 2012 was 738 mm. In the post-harvest period from August 2009 until December 2012, discharge in experimental catchment was calculated as being on average 28% higher than in the reference. No statistical tests of treatment effect were done on discharge because these results were obtained by inference and not by in-situ measurements.

3.3 Streamwater fluxes

Catchment export was calculated for MeHg, HgT, TOC, totP and inorganic N species. In the pre-harvest period, export per unit area of all elements except NO_3 was highest in the experimental catchment (Table 3). Because discharge (in mm) in LAE11 was assumed equal to LAE03 (Table 2), these differences were related to concentrations only. The most noticeable pre-harvest difference was found for MeHg export, which was 75% higher in the experimental catchment than in the reference. In the post-harvest period, export of elements increased more in the harvested catchment than in the reference. The increase in the difference between LAE11 and LAE03 ranged from 24-51% (HgT, TOC, totP), to 74-104% (totN, MeHg), to over 300% (NH_4, NO_3) (Table 3), and related to both increased discharge and increased concentrations. No statistical test was done of treatment effect as the treatment effect on discharge was estimated, not measured.

3.4 Stream biota

Biofilms covered rocky substrate in both streams and were composed of gelatinous polymers associated with the chlorophytes *Tetraspora* sp., *Microspora* sp. and various diatoms (e.g., *Eunotia* sp.). Detritus and fungi were present in low amounts. The main source of detritus in the streams was Sphagnum, while leaf litter was nearly absent, due to the low presence of deciduous trees. The macroinvertebrate fauna was species-poor and consisted of the same taxa in both streams, also after harvest. Stream water biofilm, stonefly nymphs (two closely related herbivorous Plecopterid species *Nemoura cinerea* and *Nemurella pictetii*, which together constituted the principle primary consumers) and caddisfly larvae (the carnivorous Trichopterid *Plectrocnemia conspersa*, the main predator of the stoneflies) were collected in autumn 2008, spring 2009 and autumn 2009.

There was no effect on species composition of the harvest operation. However, visual observations of the streambed indicated a strong increase in primary production in the harvested stream. In addition to a higher abundance of algae in the gelatinous biofilms, mats of green thread algae had filled substantial parts of the streambed. This was not observed in the reference stream and was interpreted as an effect of increased nutrient leaching from the catchment after the harvest operation.

Biofilms were low in MeHg (3-7 ng MeHg g^{-1} dw) (Table 4), and did not reflect differences in aqueous MeHg between the streams (Table 2, Figure 2). In the reference stream, stonefly nymphs contained 35 to 50 ng MeHg g^{-1} dw, which varied little among sampling events (Table 4, Figure 3). In LAE11, stoneflies had significantly ($p<0.0001$) higher MeHg than in the reference in autumn 2008 and spring 2009, but no differences were observed in the autumn of 2009. The differences in MeHg concentrations of the stoneflies in the first two sampling events were consistent with observed stream differences in aqueous MeHg. However, in the autumn of 2009 stream differences in aqueous MeHg were still present, while stream differences in MeHg in stoneflies had disappeared (Table 4, Figure 3). Similar patterns in stream-wise differences in MeHg levels were observed for the caddisflies, but at a lower significance level (Table 4). The $\delta^{15}N$ signatures of stoneflies in the experimental stream became significantly higher than in the reference in autumn 2009 (Figure 3, $p<0.001$), where no such differences were found at earlier sampling events. The other significant changes in chemical content of biota that occurred in the autumn of 2009 were significantly higher algal fatty acids (PUFA) (Figure 3, $p<0.05$) and total lipids (LAE03, lipid content 0.23±0.03; LAE11 lipid content 0.28±0.01; $p<0.05$) in stoneflies in LAE11, compared to the reference.

4 Discussion

4.1 Forest management effects on water chemistry

The main hypothesis guiding our paired-catchment experiment was an expected increase in MeHg concentrations as a response to the forest harvest treatment. However, no significant effect of logging was detected for streamwater MeHg concentrations, and our main hypothesis was not supported.

There was a substantial increase in MeHg export in our study – 50% more MeHg in the harvested catchment than in the reference after harvest - but this was primarily related to the estimated increased runoff after harvest (+28%). The increase in runoff was estimated based on a paired-catchment study in Balsjö in Northern Sweden, with similar climate and catchment land cover, and a similar % catchment harvest, i.e. between 30 and 40%, which documented a dominant increase of discharge during low flow (Sorensen *et al.*, 2009b). Increased water yield after harvest and other catchment disturbances is a well-known phenomenon (Hewlett and Helvey, 1970; Guillemette *et al.*, 2005; Buttle *et al.*, 2009), which is also described in catchment models (Katsuyama *et al.*, 2009). Katsuyama *et al.* (2009) simulated a 25% increase in water yield in the first six years after moderate logging operations in a forested catchment with seasonal snow cover. Porvari *et al.* (2003) reported a doubling of runoff in a Finnish paired-catchment study where a 100% clear-cut was carried out. Our estimated increase in runoff of a moderate logging disturbance appears to be in reasonable agreement with other studies.

Interestingly, only two of five published paired-catchment experiments with a focus on catchment disturbance and mercury cycling reported significant treatment effects on MeHg concentrations in surface waters. In southern Finland, the catchment manipulation included a pre-harvest period of three years, after which the catchment was clear-cut in one year and soil treatment was conducted the year after. Clear-cutting did not affect MeHg, but after the soil treatment streamwater MeHg concentrations and export rose immediately, a significant effect that lasted for three years (Porvari *et al.*, 2003) and continued for at least seven more years (Porvari, pers.comm.). The increases in streamwater MeHg were especially prominent during the growing season. In Gårdsjön in southwest Sweden, an unintended soil disturbance (wheel tracks of forest machinery)

occurred seven years after the start of the monitoring and resulted in three years of increased concentrations and export of MeHg (Munthe and Hultberg, 2004) which continued for at least another five years (J. Munthe, pers. comm.). In both studies, the increase in MeHg export was at least partly related to changes in MeHg concentrations, with peak MeHg concentrations after disturbance exceeding 1 ng L^{-1}. In the paired-catchment manipulation in Balsjö in northern Sweden, no effect of logging on MeHg concentrations and MeHg export was found after a year of pre-harvest monitoring (Sorensen *et al.*, 2009a). In Örebro in central Sweden, stump harvesting and site preparation did not affect MeHg concentrations (Eklof *et al.*, 2013). However, only logged catchments were monitored, and therefore this manipulation remains inconclusive with regard to effects of logging on aqueous MeHg.

Our study had a relatively short pre-harvest period compared to the catchment manipulations mentioned above, limiting the possibility to detect subtle responses to the treatment as intersite-variations may dominate the treatment effect (Buttle *et al.*, 2005). Nevertheless, the experimental design of our study allowed detection of a two- to fourfold increase in nitrate, ammonium and totP concentrations, suggesting that if a similarly strong response in MeHg concentration had occurred, we would have detected it. In the two experiments with long pre-disturbance periods, Munthe and Hultberg (2004) and Porvari *et al.* (2003) found a fourfold and twofold increase in mean concentrations of MeHg, respectively. Such strong responses in MeHg concentration were absent in our study. We conclude that forest management did not strongly impact catchment MeHg production in our study, similar to the results presented by Eklof *et al.* (2013) and Sorensen *et al.* (2009a). Summarizing, the conclusion that forest harvest practices may be responsible for 9 to 23% of MeHg loadings to surface waters (Bishop *et al.*, 2009) receives little support from recent catchment manipulations.

The lack of consistent responses in MeHg concentrations and export to forest harvest practices in paired-catchment studies is puzzling, partly because effects of forest harvest on streamwater MeHg have been found in synoptic studies and thus appear to be well-founded (Skyllberg *et al.*, 2009; Eklof *et al.*, 2012). Streamwater and lake MeHg are often found to correlate with the proportion of wetland

in catchments (StLouis *et al.*, 1996; Shanley *et al.*, 2005), where MeHg is thought to be produced by sulphate-reducing bacteria using labile organic matter as energy substrate (Morel *et al.*, 1998), possibly also influenced by nutrient status (Tjerngren *et al.*, 2012) . However, forest harvest operations are usually not undertaken in wetlands or organic-rich soils, and the increase in MeHg concentrations in southern Finland (Porvari *et al.*, 2003) and Gårdsjön (Munthe and Hultberg, 2004) is related to disturbance of upland, not wetland, soils. Porvari *et al.* (2003) suggested that the enhanced concentrations of MeHg were possibly related to higher soil temperatures (through increased direct solar radiation) and humidity (from higher ground water levels), favouring methylation. In all referred paired-catchment experiments where logging took place, it is reasonable to assume that increased soil temperatures and humidity in the harvested catchments did occur as this is a common effect of forest clear-cutting (Olchev *et al.*, 2009; Schelker *et al.*, 2013). However, this was clearly not sufficient for increasing streamwater MeHg in three of four cases.

Another mechanism relevant to explain forestry effects on MeHg production is through increasing loads of labile organic matter (Kainz *et al.*, 2003; Roy *et al.*, 2009), either in the form of harvest residues or from release of fresh organic matter through soil disturbance, both of which promote microbial activity and thereby Hg-methylation. Additionally, the creation of anoxic spots in the soil related to soil compaction from heavy machinery might also promote methylation. Such compaction is likely to have taken place in all catchment manipulations, but again, this was not sufficient to create increases in streamwater MeHg in all experiments. Possibly, site differences in sulphur (S) deposition could play a role for the susceptibility to logging and soil disturbance as sulphate is a limiting factor for MeHg production (Gilmour *et al.*, 1992; Akerblom *et al.*, 2013). The sites in southern Finland (Porvari *et al.*, 2003) and southwest Sweden (Munthe and Hultberg, 2004) are both located in regions that have historically received considerably higher loads of S deposition (Jenkins *et al.*, 2003; Posch *et al.*, 2012) than our study site in southeast Norway, Örebro in central Sweden (Eklof *et al.*, 2013) and Balsjö in northern Sweden (Sorensen *et al.*, 2009a).

The most distinct effect of the harvest operation was the large increase in NO_3 concentration and export, which lasted throughout the entire post-harvest period. Increased runoff of inorganic N

species after harvest is common in northern catchments (Likens *et al.*, 1970; Kreutzweiser *et al.*, 2008), but can be reduced by retaining an intact buffer zone close to the streams (Lofgren *et al.*, 2009). Following common forestry practice in Norway, the stream in our study was too small to include such buffer retention, except for a smaller part of the central catchment area where the stream course followed the border between upland forest and adjacent open mires. Another sign of changes in N cycling was the significant decrease in CN ratio of dissolved organic matter (DOM), suggesting an enrichment of DOM with nitrogen. TOC concentrations and TOC export did not respond to the forest harvest, in contrast to previous findings (Porvari *et al.*, 2003; Laudon *et al.*, 2009). Other responses were increases in total P concentrations which were only significant in the second year after logging, but effective P retention in the streambed was suggested by observations of thread algae and high concentrations of algae in the biofilm. A less distinct response of P compared to N in streamwaters after logging has also been found previously (Kreutzweiser *et al.*, 2008; Lofgren *et al.*, 2009) and could be related to strong biological retention of P in the stream (Valett *et al.*, 2002).

4.2 Forest management effects on MeHg in the stream food chain

The differences in MeHg concentrations in the streams were reflected in MeHg levels in primary consumers (herbivorous stoneflies) in the autumn of 2008 and the spring of 2009 (de Wit *et al.*, 2012). Trophic enrichment of MeHg in the biota, and the efficiency of MeHg transfer from the stream into the food chain, were similar in both streams and at both sampling occasions. Thus, the mechanisms controlling MeHg levels in aquatic biota in both streams were exactly the same. The different levels in MeHg in the primary consumers in the streams were explained by differences in exposure to aqueous MeHg, where LAE11 had higher MeHg than LAE03. Exposure to MeHg at the base of the food chain is key to the bioaccumulation in the stream food web, as studies by for instance Chasar *et al.* (2009) also indicate. We also found that fatty acids content of the invertebrates indicated that the ingestion of bacteria was likely to promote MeHg bioaccumulation, while ingestion of algae had the opposite effect. Fatty acids can be used as dietary biomarkers to indicate recent dietary success of biota (Kainz and Fisk, 2009).

The surprising observation in the autumn of 2009, compared to the first two sampling events, was that MeHg levels in primary consumers of both streams were similar in the autumn of 2009, *despite* continued differences in exposure to aqueous MeHg. That indicated that the efficiency of MeHg transfer from the water phase into the base of the food chain had declined in LAE11 compared to LAE03. Upon further inspection, this observation fitted well with the postulated importance of dietary sources for MeHg bioaccumulation in de Wit *et al.* (2012). The significant change in $\delta^{15}N$ signature in primary consumers of the harvested stream in the autumn of 2009 was interpreted as a change in baseline N availability, substantiated by the observed increase in streamwater NO_3 and NH_4. Additionally, visual inspection of the biofilm in the harvested stream indicated a much higher abundance of algae than the year before, a strong indication that nutrient access in the stream had increased. Further evidence for higher primary productivity in the harvested stream was found in the significantly higher contents of lipids and algal fatty acids in the primary consumers of LAE11, which indicate a higher dietary access to algae. Possibly, algae are a food source with relatively low contamination of MeHg - consistent with the low concentrations of MeHg in biofilms – or the larger dietary access to algae caused increased somatic growth of consumers leading to lower MeHg per unit biomass, also known as the growth dilution effect (Goedkoop *et al.*, 2007). Algal blooms have been shown to lower MeHg contamination in aquatic food webs in lakes (Pickhardt and Fisher, 2007), and we show here that a similar mechanisms may also exist in stream foodwebs.

Our results demonstrate that effects of forest harvest on MeHg in the aquatic food chain should take the following aspects into account: i) changes in MeHg in runoff, ii) changes in in-stream productivity and iii) changes of dietary sources. Previous studies on catchment disturbance effects on MeHg in aquatic biota (Garcia and Carignan, 1999, 2000, 2005; Desrosiers *et al.*, 2006; Garcia *et al.*, 2007) have indicated positive correlations between disturbance and MeHg in aquatic biota, but have paid little attention to possible confounding effects of changes in aquatic productivity and diet. Likewise, the documented increase in Hg in fish in Swedish lakes has been suggested to be related to increases in lake DOC with associated higher exposure to MeHg (Akerblom *et al.*, 2012), but our results suggest that changes in dietary sources can drive changes in MeHg levels in aquatic foodwebs.

5 Conclusion

Contrary to earlier results from paired catchment experiments and synoptic studies, we did not find an effect of forest management on catchment MeHg production. There is little understanding of crucial factors that render MeHg in surface waters sensitive to catchment disturbance, but we speculate that sulphur deposition might be important. We found a strong nutrient release to the streamwaters as a response to harvest. Such additional nutrients promote primary productivity and growth of primary consumers, inducing a decrease of MeHg in biomass in streamwater biota. Our results demonstrate that a short-term effect of forest harvest may be a reduction of MeHg in aquatic biota, because of improved dietary quality for the consumers at the bottom of the stream food webs. An assessment on the effects of forestry on Hg in northern, managed landscapes concluded that one tenth to a quarter of Hg in fish might be attributed to forest harvesting (Bishop *et al.*, 2009). Recent experimental studies, including our own, suggest that this may be an oversimplification. Paired catchment manipulations show that the impact of disturbance on MeHg in streamwaters varies strongly. Predictions of forest management effects on MeHg in streamwater and aquatic food webs are hampered by limited understanding of catchment controls on MeHg production.

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Table

Table 1 Catchment characteristics

Catchment	Area	Pine-dominated forest	Spruce-dominated forest	Forested peatland	Sphagnum mire	Other[1)]	Pre-harvest volume[2)]	Harvest removal
	ha	% of catchment area					$m^3\ ha^{-1}$	%[3]
LAE03	83	63.9	9.6	4.8	16.3	5.4	77.6	-
LAE11	24	56.8	10.5	14.7	17.9	-	62.3	38

[1)] Lakes and powerline; [2)] Overall mean for spruce- and pine dominated forest and forested peatland. [3)] % of standing volume

Table 2 Discharge (sum, in mm) and mean water chemistry of streams LAE03 (reference catchment) and LAE11 (harvested catchment) for the preharvest period (June 2008 – January 10 2009), post-harvest-1 (remainder of 2009) and postharvest 2 (2010-2012). Numbers show mean / standard deviation / number for observations. Cond, conductivity (µS cm^{-1}); labile Al (µg L^{-1}); other major ions in mg L^{-1}; TOC, total organic C; TON, total organic N; DOM, dissolved organic matter; CN_{DOM}, CN ratio of DOM; $UVabs_{254}$, absorbance of UV at 254 nm; $SUVA_{254}$, specific UV absorbance at 254 nm.

		LAE03	LAE11	LAE03	LAE11	LAE03	LAE11
		-----Preharvest-----		-----Post-harvest – 1-----		----Post-harvest – 2 -----	
Discharge (mm)		245	245	684	763	2232	2846
Hg, MeHg (ng/L)	MeHg	0.09/ 0.02 /9	0.18/ 0.11 /10	0.08/ 0.04 /28	0.22/ 0.13 /30	0.06/ 0.02 /44	0.15/ 0.10 /44
	Hg	5.0/ 2.6 /9	5.3/ 2.3 /10	3.8/ 1.1 /28	5.0/ 1.3 /30	3.1/ 0.9 /43	4.1/ 1.6 /43
	MeHg/TOC ng/g	6.8/ 3.6 /9	11.7/ 8.3 /8	6.0/ 3.1 /28	11.1/ 7.3 /30	4.7/ 1.9 /43	8.4/ 6.0 /42
	HgT/TOC ng/g	342/ 116 /9	241/ 70 /8	270/ 62 /28	245/ 53 /30	259/ 52 /42	227/ 71 /41
major ions	pH	4.74 / / 22	4.44 / / 14	4.76 / / 43	4.43 / / 44	4.83 / / 52	4.50 / / 53
	H+	18.3/ 6.5 /22	36.6/ 16.1 /14	17.4/ 10.9 /43	36.8/ 12.0 /44	14.8/ 7.8 /52	31.9/ 11.6 /53
	Cond	1.53/ 0.27 /22	2.22/ 0.59 /14	1.41/ 0.28 /15	2.18/ 0.50 /15	1.48/ 0.23 /52	2.17/ 0.38 /53
	Ca	0.75/ 0.11 /19	0.79/ 0.19 /13	0.78/ 0.18 /15	0.75/ 0.20 /15		
	Mg	0.14/ 0.02 /19	0.12/ 0.02 /13	0.14/ 0.02 /15	0.12/ 0.03 /15		
	K	0.03/ 0.01 /19	0.06/ 0.04 /13	0.06/ 0.03 /15	0.24/ 0.10 /15		
	SO_4_S	0.67/ 0.29 /19	0.61/ 0.22 /13	0.78/ 0.31 /15	0.68/ 0.23 /15	0.79/ 0.30 /27	0.67/ 0.27 /28
	Labile Al	19/ 12 /19	8/ 7 /13	23/ 9 /15	9/ 6 /15		
Nutrients µg/L	NO_3-N	5.6/ 5.9 /22	5.1/ 5.2 /14	8.3/ 6.9 /43	4.3/ 6.6 /44	10.0/ 12.3 /41	29.9/ 29.5 /42
	NH_4-N	3.6/ 1.9 /21	10.1/ 11.2 /9	6.0/ 5.2 /43	10.0/ 11.4 /44	7.2/ 6.3 /41	33.0/ 37.2 /42
	Total P	5.5/ 2.1 /22	7.0/ 2.0 /13	4.5/ 1.2 /16	8.3/ 3.9 /16	5.2/ 2.6 /47	9.4/ 5.2 /48
	Total N	258/ 50 /22	320/ 54 /14	256/ 68 /43	342/ 73 /44	246/ 43 /42	382/ 115 /43
DOM	TOC (mg L^{-1})	14.2/ 3.7 /22	21.3/ 7.3 /14	13.9/ 5.5 /43	20.3/ 5.4 /44	12.4/ 3.4 /52	18.2/ 5.4 /53
	TON (µg L^{-1})	247/ 51 /21	284/ 58 /9	241/ 68 /43	328/ 74 /44	229/ 41 /41	321/ 78 /42
	CN_{DOM} g/g	57/ 7 /21	72/ 15 /9	57/ 8 /43	62/ 7 /44	54/ 8 /41	57/ 12 /42
	$UVabs_{254}$	0.61/ 0.14 /8	0.93/ 0.18 /8	0.65/ 0.22 /43	0.97/ 0.23 /44	0.61/ 0.19 /52	0.90/ 0.27 /53
	$SUVA_{254}$	0.045/0.002/8	0.043/0.006/8	0.047/0.003/43	0.048/0.004/44	0.049/0.004/52	0.050/0.004/53

Table 3 Discharge (sum, in mm) and element fluxes (Hg, MeHg, TOC, inorganic N species, totN and totP) of catchment LAE03 (reference catchment) and LAE11 (harvested catchment) as sum for the pre-harvest period (June - December 2008) and as the mean of the post-harvest period (2009-2012). In parentheses for LAE11 are the pre- and postharvest discharge and fluxes in % of the LAE03 value.

			Preharvest	Postharvest
			June-December 2008	Annual mean 2009-2012
discharge	mm	LAE03	245	729
		LAE11	245 (100)	903 (124)
Hg	µg m^{-2}	LAE03	1.4	2.7
		LAE11	1.6 (112)	4.1 (156)
MeHg	‘’	LAE03	0.019	0.041
		LAE11	0.034 (175)	0.103 (254)
TOC	g m^{-2}	LAE03	4.1	9.7
		LAE11	6.2 (151)	17.1 (177)
NH_4-N	mg m^{-2}	LAE03	0.9	3.7
		LAE11	1.1 (128)	15.6 (424)
NO_3-N	‘’	LAE03	0.9	2.5
		LAE11	0.8 (85)	12.0 (470)
TotN	‘’	LAE03	71	176
		LAE11	85 (119)	311 (213)
TotP	‘’	LAE03	1.5	3.4
		LAE11	1.9 (124)	7.3 (176)

Table 4 Mean concentrations of MeHg (µg g^{-1} dry weight), standard deviation (std) and nr of observations (n) in biofilm, stoneflies and caddisflies, grouped by sampling event and catchment (LAE03, reference; LAE11, harvested catchment). If n=2, single values are given. Significant differences within one group, between catchments, are given as a and b when $p<0.0001$, as c and d when $p<0.005$, and as e and f when $p<0.1$ (Students t-test).

Group	catchment	Autumn '08	Spring '08	Autumn '09
			mean / std / n	
Biofilm	LAE03	5.9, 7.2 / - / 2	6.3 / 1.2 / 3	2.6 / 1.4 / 3
	LAE11	5.2 / 0.8 / 4	4.8 / 1.0 / 3	4.0 / 1.0 / 3
Stonefly	LAE03	46[a] / 7 / 5	43[a] / 5 / 3	37 / 9 / 3
	LAE11	166[b] / 29 / 4	90[b] / 5 / 3	36 / 4 / 3
Caddisfly	LAE03	90, 91[e] / - / 2	90[c] / 20 / 3	120, 130 / - / 2
	LAE11	275, 420[f] / - / 2	233[d] / 25 / 3	120 / 20 / 3

Figure 1

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Figure 2a

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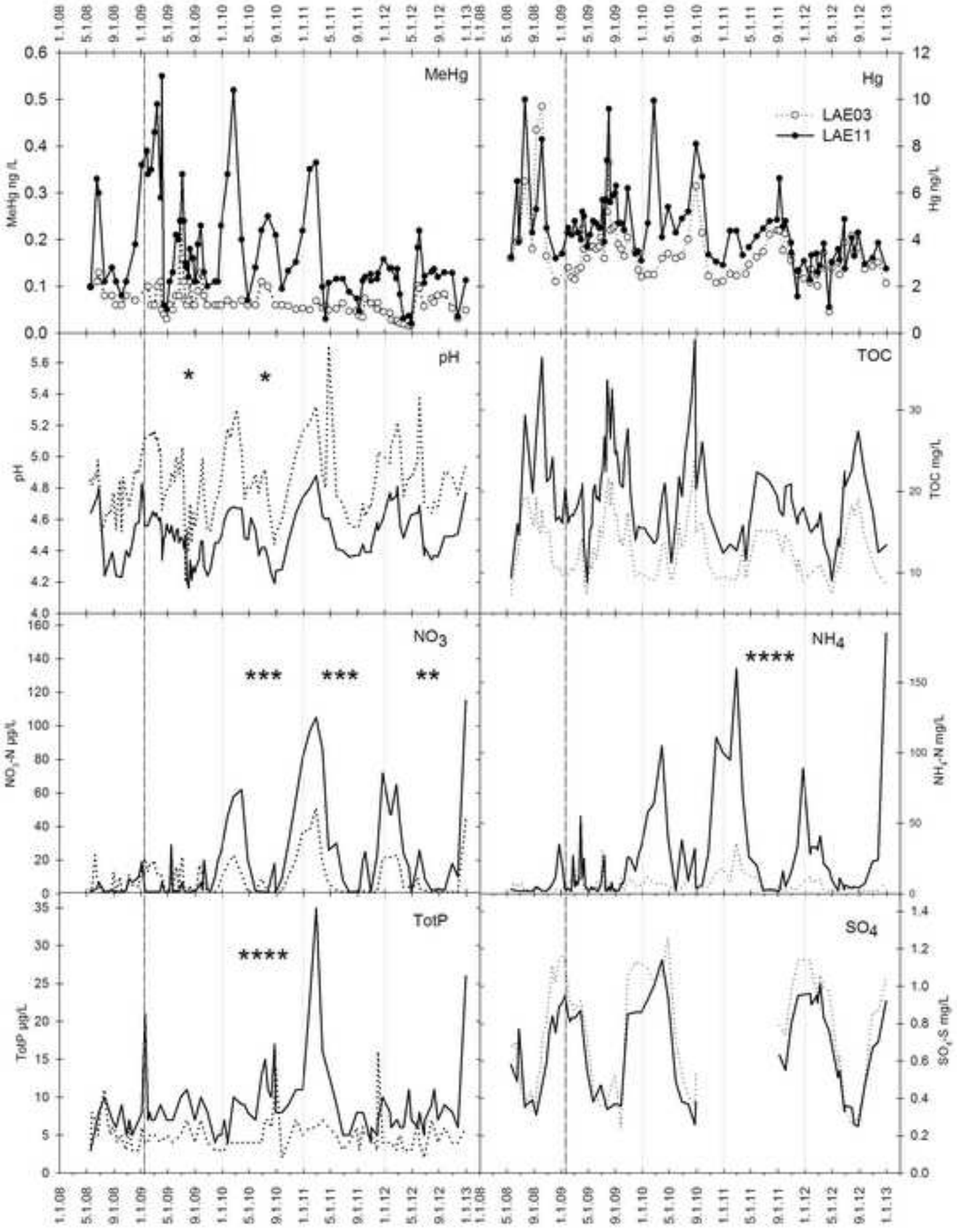

Figure 2b

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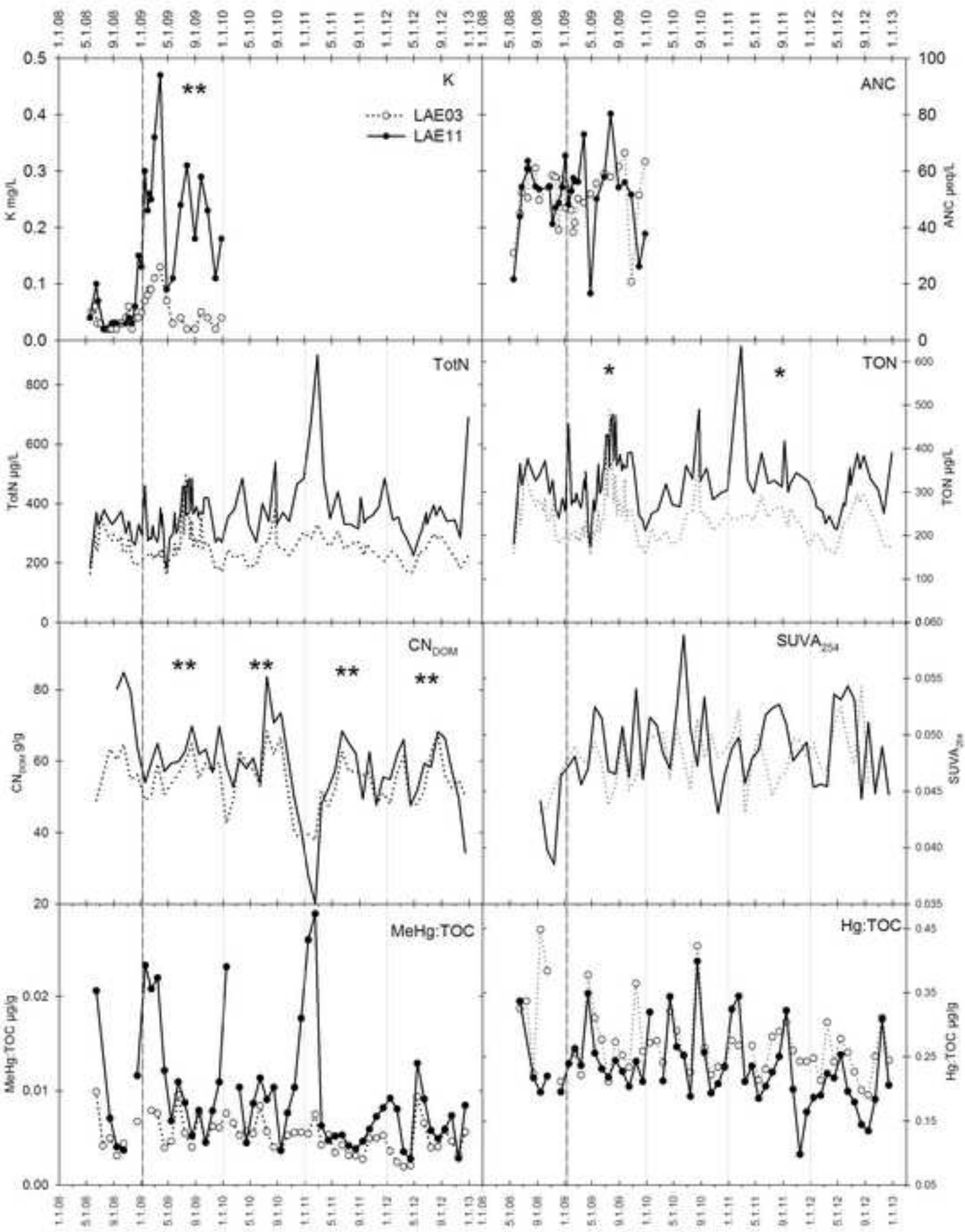

Figure 3

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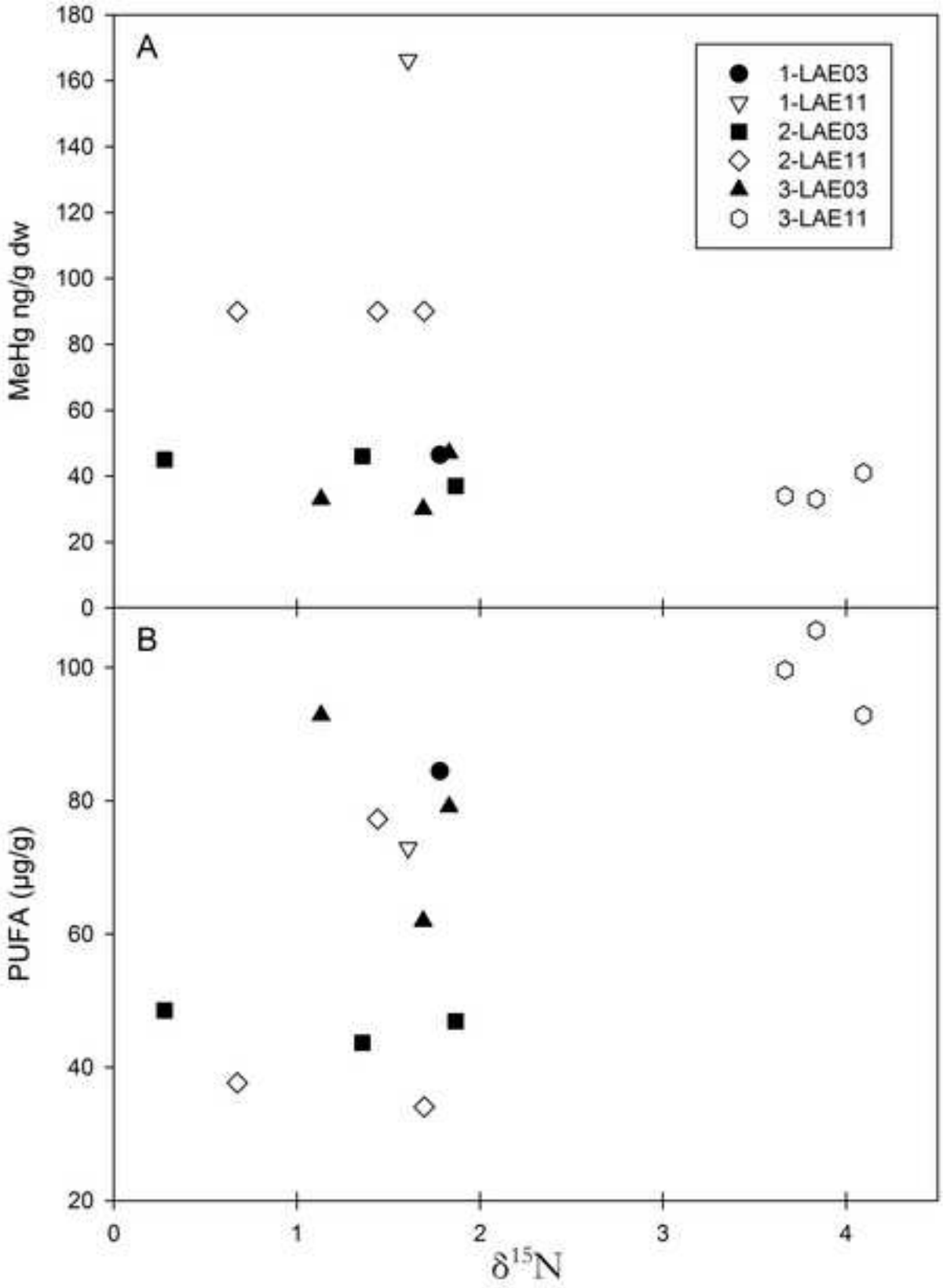

Highlights

- Forest harvest may increase MeHg in streamwaters and aquatic food chain
- Streamwater chemistry and MeHg in biota were studied in a paired-catchment design
- Concentrations of nutrients increased but not streamwater MeHg and total Hg
- MeHg in invertebrates was reduced after harvest related to improved nutrient access

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