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Impact of Beaver Pond Colonization History on Methylmercury Concentrations in Surface Water

Oded Levanoni,[*,†] Kevin Bishop,[†,‡] Brendan G. Mckie,[†] Göran Hartman,[§] Karin Eklöf,[†] and Frauke Ecke[†,∥]

[†]Department of Aquatic Sciences and Assessment, Swedish University of Agricultural Sciences, (SLU), Box 7050, SE-750 07 Uppsala, Sweden

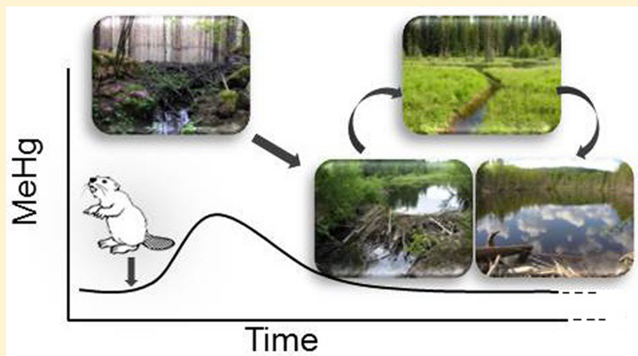
[‡]Department of Earth Sciences, Air Water and Landscape Sciences, Villavägen 16, Uppsala University, SE-752 36 Uppsala, Sweden

[§]Department of Ecology, Swedish University of Agricultural Sciences, (SLU), Box 7050, SE-750 07 Uppsala, Sweden

[∥]Department of Wildlife, Fish, and Environmental Studies, Swedish University of Agricultural Sciences, (SLU), SE-901 83 Umeå, Sweden

Supporting Information

ABSTRACT: Elevated concentrations of methylmercury (MeHg) in freshwater ecosystems are of major environmental concern in large parts of the northern hemisphere. Beaver ponds have been identified as a potentially important source of MeHg. The role of beavers might be especially pronounced in large parts of Europe, where beaver populations have expanded rapidly following near-extirpation. This study evaluates the role of the age and colonization history (encompassing patterns of use and reuse) of ponds constructed by the Eurasian beaver *Castor fiber* in regulating MeHg concentrations in Swedish streams. In 12 beaver systems located in three regions, we quantified MeHg concentrations together with other relevant parameters on five occasions per year in 2012−2013. Five were pioneer systems, inundated for the first time since beaver extirpation, and seven were recolonized, with dams reconstructed by newly recolonizing beavers. MeHg concentrations in pioneer but not in recolonized beaver systems were up to 3.5 fold higher downstream than upstream of the ponds, and varied between seasons and years. Our results show that pioneer inundation by beavers can increase MeHg concentrations in streams, but that this effect is negligible when dams are reconstructed on previously used ponds. We therefore expect that the recovery and expansion of beavers in the boreal system will only have a transitional effect on MeHg in the environment.



■ INTRODUCTION

High mercury (Hg) concentrations in soils, water and biota are a major concern in large parts of the northern hemisphere. Mercury bioaccumulates in aquatic foodwebs,[1] and can potentially impact human health through the consumption of fish. Concentrations of Hg in fish tissue higher than the WHO recommendations[2] as well as the U.S. Environmental Protection Agency criteria[3] (0.5 and 0.3 mg Hg/kg tissue, respectively) commonly occur in large parts of the hemiboreal zone. For example, the European Union threshold limit of 0.02 mg Hg/kg tissue[4] is exceeded in many water bodies in Fenoscandia.[5−7] Methylmercury (MeHg) is one of the most toxic forms of Hg and effectively bioaccumulates in the food web. Concentrations of MeHg in surface waters are the result of the dynamic balance between methylation and demethylation,[8,9] which is facilitated by both a biotic and abiotic process.[10] Methylation is predominantly a microbial process derived by sulfate reducing bacteria and other microbes, and influenced by various factors including suitable electron acceptors (such as SO_4^{2-}), electron donors (often organic carbon of appropriate quality), Hg species, redox conditions, and thermal regimes favoring sulfate reducing and other bacteria.[11−13] These conditions often occur when new wetlands are formed and in association with reservoir creation.[14,15] In summer, warmer water temperature and increased biological activity in such systems can create reducing environments that increase the activity of sulfate reducing bacteria and promote methylation, while colder seasons are associated with reduced methylation.[16,17]

By building dams, beavers convert stream stretches into systems characterized by pond complexes, creating new wetland areas and generating altered riparian conditions along former stream reaches. Dam construction alters the annual stream

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discharge regime (typically decreasing velocity), increases the retention of sediment and organic matter, and can also affect water temperature and oxygen content.[18,19] The decay of flooded vegetation and dead trees in beaver systems might result in anoxic conditions in beaver ponds.[18,20,21] All these changes contribute to the potential of beaver ponds to function as sites for increased methylation of mercury. Upon abandonment, due to low food quality/quantity for example, beaver systems are no longer active and regrowth of vegetation takes place.[22−24] Once food availability is sufficient, a new colonization might take place.[20,25−27] These recolonized systems may have characteristics closer to older ponds rather than new ponds since characteristics of the beaver systems might be preserved even when not active (i.e., dams, channels and vegetation structure).

The Eurasian beaver, *Castor fiber*, was formerly widely distributed along forested riparian habitats and water bodies across most of Europe.[28,29] However, overhunting, together with reduction of its habitat, led to the near extirpation of *C. fiber* in most of Eurasia by the middle of the 19th century, including Scandinavia. It is estimated that before conservation measures were undertaken, only a few small distinct populations survived in Europe, with roughly 1200 individuals.[28,29] This process was particularly marked in Sweden, where there were no new beaver observations after the 1870s. Since the beginning of the 20th century, protection of the species and reintroductions in many countries have led to the recovery of *C. fiber* in Sweden, and today there are estimated to be over 130,000 beavers.[30,31] The rapid recolonization of beavers has resulted in dams of varying age and colonization history, with potentially variable effects on MeHg concentrations in the stream systems.

Measurements from a beaver pond in the U.S. that had existed for at least a decade indicated methylation production that was comparable to rates in natural wetlands, and well below rates in flooded terrestrial areas.[13] In two studies of beaver ponds in Canada, methylation efficiency (percentage of MeHg out of total Hg) was higher in recently constructed beaver ponds especially in summer, and decreased with increasing pond age.[17,32] To our knowledge, these are the only studies on the effects of beavers on MeHg in streamwater that consider seasonal variation and/or pond age.[13,17,32] We are not aware of any studies on the role of colonization history, potential regional differences, or between-year differences on MeHg in beaver pond systems. A larger-scale perspective, accounting for cross-regional and longer-term variation, on the role of beavers in regulating MeHg is urgently needed, considering the continuing expansion in both population sizes and the distribution range of Eurasian beavers, and potential implications of further increases in MeHg levels in surface waters.

The main objective of this study was to investigate how colonization history and age of beaver systems in combination with geographic region and seasonality affect MeHg concentrations of downstream beaver pond systems. Due to the initial flooding of soils, and the input of organic material from decaying terrestrial vegetation and soil organic carbon as well as tree-felling by beavers, we expected newly inundated ponds to favor methylation of mercury, reflected in significantly higher MeHg concentrations downstream as compared to upstream of the pond. This effect is expected to be enhanced during summer and low flow conditions. After the initial inundation and the subsequent colonization cycle, including abandonment and reuse of sites by succeeding beavers, this downstream effect should decrease with time. Hence, we hypothesized that recolonized beaver systems would have lower downstream MeHg concentrations than systems newly inundated for the first-time. Finally, we discuss the implications of our results for future beaver management.

■ MATERIALS AND METHODS

Study Sites. Methylmercury concentrations and other supporting data were sampled from 12 beaver systems, six located north and six south of the *limes norrlandicus*, a major biogeographical discontinuity in Scandinavia seen most markedly as the natural northern limit of the English oak *Quercus robur*.[33] The southern sites are located at the border between the hemiboreal and southern boreal vegetation zone (Latitude 59.2° to 59.8°), whereas the six northern sites belong to the middle boreal vegetation zone (n = 3 in the so-called "slightly oceanic section" (Latitude 62.2° to 62.6°), and the so-called "indifferent section" of the zone (n = 3, Latitude 66.2° to 66.5°)) (Ahti et al. 1968) (Figure 1). All catchments of the study sites were dominated by coniferous forest (see SI Table S1).

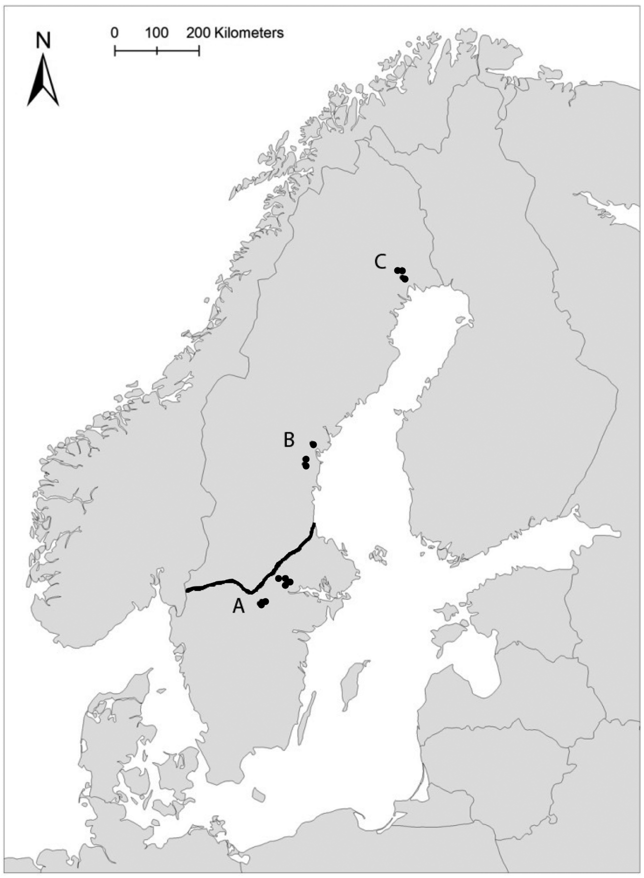

Figure 1. Map of the study sites (filled circles) in Sweden and in relation to the limes norrlandicus (solid line). The six southern sites are located at the border between the hemiboreal and southern boreal vegetation zone (A) and the six northern sites belong to the middle boreal vegetation zone; three are located in the so-called "slightly oceanic section" (B), and three in the so-called "indifferent section" of the zone (C) (see Material and Methods for definition of vegetation zones).

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Pond Age and Colonization History. To determine the age of our beaver ponds, we inspected aerial photographs for the period 1973–2012, taken by the Swedish Mapping, Cadastral and Land Registration Authority (Lantmäteriet), with 2–10 years intervals between photographs. The photos were available in black-and-white or in CIR (color infrared) taken at a flight height of either 3000 or 9600 m. All aerial photos were available digitally with a spatial resolution of at least 0.5 m. The aerial photos were rectified in ArcGIS[34] with a second order polynomial transformation resulting in a mean RMS (root-mean-square) error of 3.8.

We checked the photos for any visible signs of beaver activity, starting with the latest image from 2012 and going back in time to older images. Signs included dams, felled trees, and aggregation of snags (standing dead trees). Upper and lower limits of the system age were determined by the oldest photo with signs of beaver activity and the youngest photo without signs (e.g., signs of beaver activity in 2007 but not 2005 would yield a dam age of 5–7 years in 2012).

We combined information from the photographs with the knowledge of local farmers and residents, forestry companies and municipal documentation, as well as our own observations to classify the age of each system, and its colonization history, that is, first colonization, abandoned or recolonized. Based on this information, we divided our ponds into two categories of colonization history: pioneer and recolonized. Pioneer beaver systems are those where we could only identify one dam construction event, with no previous signs of beaver activity. Recolonized beaver systems are those where we could identify one or more periods of recolonization and abandonment after initial dam construction, before being occupied again. System colonization status (pioneer or recolonized) was further supported based on visual signs, such as number of lodges around the main pond, snags in the ponds, vegetation on the dam itself, the structure and size of the dam, the depth of the pond, remains of old fallen trees in the surroundings, and number of ponds (see SI table S2 and S3, respectively, for details on the applied criteria and characteristics of each studied beaver system). Similar factors have previously been successfully applied to characterize beaver systems.[17,32] The beaver systems were also classified to three age groups (Young <10 years, Intermediate 10–20 years, Old >18 years) following previous studies.[17,32]

Field Sampling. Water samples were first collected during November 2011, and thereafter approximately every second month from April 2012 to November 2013. We subdivided these sampling events into five "seasons"; spring (April to early June), summer (late June to July), late summer (August to September), autumn (October to November, before thick ice formation) and winter (February to March, sampled only in 2013 under maximum ice cover). At each beaver system, samples were taken from three sites: (1) An *upstream reference* site, upstream from the first pond in the system, where no beaver activity was apparent, (2) A *pond* site, representing the largest pond in the system, which was always the pond where the lodge was constructed, and (3) A *downstream* site, immediately downstream from the last pond in the system. In all cases water samples were taken at 20–30 cm depth (unless the total depth was less than 20 cm, then samples were taken 5 cm above the stream bottom). Samples for total mercury (THg) were collected in 100 mL fluorinated ethylene propylene (FEP) bottles that were precleaned with BrCl and HCl. MeHg samples were collected in new brown glass bottles. Water samples for total organic carbon (TOC) and dissolved organic carbon (DOC) were collected in 250 mL high-density polyethylene bottles. Single use plastic gloves were used when collecting samples. All bottles were rinsed three times in streamwater before the sample was collected. The samples were stored in a cooler during transport to the laboratory. In the laboratory, samples were stored in a refrigerator (0 °C for TOC/DOC and 5 ± 3 °C for THg and MeHg). Samples were sent to the laboratories analyzing MeHg and THg within 1–3 days after sampling. In the MeHg laboratory an enriched MeHg isotope standard ($MeHg^{200}$) was added to the sample within 24 h of arrival to account for any MeHg loss occurring during further storage and/or analysis.[35] In the THg laboratory samples were preserved by adding concentrated suprapur HCl as soon as possible after arrival. The TOC and DOC analyses were conducted within 1 week from sampling.

Dissolved oxygen measurements were taken using an optical dissolved oxygen sensor (HDO, resolution 0.01 mg/L or ±1% saturation) that was two-point calibrated (0 and 100% saturation) a day before measurement took place. Repeated measurements were logged every 5 s until a stable reading was obtained. Chlorophyll-*a* measurements were taken using a fluorometer sensor (resolution 0.01 μg/L accuracy ±3% of full scale). Both HDO and Chlorophyll-*a* sensors were mounted on a Eureka Manta2 multiparameter water quality recorder (Eureka Water Probes LTD). Water level and temperature were logged at all sites every 4 h using TruTrack WT-HR Water Height Data Loggers (resolution ±1 mm ±1% of full scale and repeatability ±0.1 °C).

Laboratory Analysis. The total Hg analyses were performed at the Swedish Environmental Research Institute (IVL). Total Hg concentrations were quantified with the aid of cold vapor atomic fluorescence spectroscopy (CVAFS) after oxidation by BrCl and reduction to Hg (0) with $SnCl_2$ following the US Environmental Protection Agency standards, method EPA 1631.[36] The detection limit of the analysis method was 0.04 ng/L and the limit of quantitation (LOQ) was 0.1 ng/L.

Methylmercury (MeHg) in water was analyzed by species-specific isotope dilution followed by mass spectrometry based on the procedure described in Lambertsson et al.[35] by ALS Scandinavia AB (certified by SWEDAC reg no. 2030). However, the GC and ICPMS instrument, the isotopic standard and the buffer solution differ to what is used by Lambertson et al.[35] and these are described in Baxter et al.[37] For each batch of samples, preparation blanks quality control and synthetic quality control was preformed (for detailed information see SI Appendix 2). The detection limit of the analysis method was 0.01 ng/L and the LOQ was 0.03 ng/L.

Organic carbon in water samples was analyzed at the Department of Aquatic Sciences and Assessment, Swedish University of Agricultural Sciences (SLU), Uppsala. DOC samples were filtered through 0.45 μm cellulose acetate filters (Minisart, Sartorius). TOC and DOC samples were analyzed with a Shimadzu TOC-VCPH carbon analyzer. Absorbance was measured at 254 nm (PerkinElmer Lambda 40) in a 5 cm cuvette and used to calculate SUVA (specific UV absorbance, here DOC-normalized absorbance).

Sulfate (SO_4^{2-}), pH, total nitrogen, nitrate (NO_3^-), nitrite (NO_2^-) and total phosphorus were analyzed by the Department of Aquatic Sciences and Assessment at SLU using SWEDAC accredited methods (Fölster et al. 2014 and references therein).[38]

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Data Analysis. To evaluate between year discharge differences, precipitation was calculated between June to October for each beaver system based on daily measurements averaged to 4 × 4 km horizontal grid resolution (Swedish Meteorological Survey PTHBV database).[39]

To quantify the effect of beaver activities on the concentrations of MeHg, the ratio of MeHg between the sites down- and upstream of the beaver ponds (D/U MeHg ratio) was calculated for each beaver system.[32] The effect of beaver activity on D/U MeHg ratios and the interactive effects of beaver system colonization status, geographic region, season, and year were then investigated using analysis of variance (ANOVA). Prior to analysis, response variables were natural log-transformed to satisfy parametric assumptions. Fixed factors included beaver system colonization status (pioneer and recolonized), year (2012, 2013), latitude and geographic region (north and south) as well as season (spring, summer, late-summer, autumn). The winter season was excluded since there was only one winter sampling, in 2013. Stream system was fitted as a random block factor. We tested for all fixed factors and the interaction between them (crossed and nested). Differences in the variance of D/U ratios among colonization categories were assessed with Levene's test. A Wilcoxon signed-rank test was used to determine significant differences of upstream and downstream MeHg concentrations, to determine significant differences of the D/U MeHg to the 1:1 ratio, and to compare overall MeHg concentrations between pioneer and recolonized systems.

Partial least-squares regression (PLS) was used to assess the relative importance of the different predictors for explaining variation in the D/U MeHg ratio. PLS maximizes explained covariance between variables by extracting orthogonal components from the set of variables.[40] A predictive model for the response variable is constructed using the extracted components, and variable influence on the projection (VIP) is calculated to rank the predictors in their relative importance for explaining the response variable. VIP values are calculated for each variable by summing the squares of the PLS loading weights, weighted by the amount of sum of squares explained in each model component. The sum of squares of all VIPs is equal to the number of terms in the model. Hence, the average VIP is equal to 1.[40] VIP-values larger than 1 indicate "important" predictor variables and values under 0.5 indicate "unimportant" ones. In accordance with Eriksson et al. (2006)[40] we applied a threshold VIP of 0.8 for identifying useful predictors. The response variable was the D/U MeHg ratio, which was natural log transformed to correct for skewness. Categorical predictors were the colonization history (pioneer or recolonized), year (2012 or 2013), season (four seasons, as with the ANOVA, winter was excluded due to only one year of sampling) and the stream system (n = 12). We included further continuous predictors which can explain variation in MeHg D/U ratio as a result of methylation-demethylation dynamics. These predictors included latitude (in decimal degrees), discharge (catchment mean monthly discharge m^3/sec), temperature (mean temperature for 7 days prior to sampling, °C), total Hg, TOC, SO_4^{2-}, pH, dissolved oxygen, chlorophyll-*a* in the water column, total N, NO_3^-+NO_2^-, and total P. All selected physiochemical parameters were shown in previous studies to affect MeHg concentration in water.[12,41−45] The model was validated by comparing the goodness of fit of the original model with the goodness of fit of 1000 randomly permutated Y-observations while the X-matrix has been kept intact (Permutation plot) and by analysis of variance in the cross validated residuals of the Y variable (CV-ANOVA).[40]

The ANOVA model was conducted using JMP.[46] The Wilcoxon signed-rank test and Levene's test for homogeneity of variance were conducted using R.[47] PLS was conducted using SIMCA-P.[48] If not stated otherwise, we give results as mean ±1 SE.

■ RESULTS

The Effect of Colonization History on MeHg Concentrations. The overall downstream MeHg concentrations in pioneer beaver systems (median 0.40; Interquartile range (IQR) 0.25−0.96 ng/L) were significantly higher than the upstream concentrations (median 0.13, IQR 0.08−0.24 ng/L, Wilcoxon signed-rank test, P < 0.001) during the entire study period. In these pioneer systems, the downstream MeHg concentrations during summer were up to 3.5-fold higher than the upstream ones. In contrast, overall MeHg concentrations measured downstream from recolonized beaver systems (median 0.27, IQR 0.21−0.37 ng/L) were not significantly different from upstream concentrations (median 0.27; IQR 0.20−0.42 ng/L, Wilcoxon signed-rank test P > 0.05). Overall, the D/U MeHg ratio was significantly higher in pioneer (mean ±1 SE: 2.49 ± 0.20) than in recolonized beaver systems (1.21 ± 0.10; ANOVA $F_{1,1}$ = 68.35, P < 0.001) and was significantly higher than the 1:1 ratio in pioneer systems (Wilcoxon signed-rank test, P < 0.001) but not in recolonized ones (Figure 2, see also ANOVA table in SI table S4). Surprisingly, the D/U MeHg ratio in recolonized beaver systems was similar in the three age groups (Young 1.22 ± 0.13 (n = 10), Intermediate 1.22 ± 0.15 (n = 36), Old 1.20 ± 0.21 (n = 20); ANOVA $F_{2,63}$ = 0.26, P > 0.05). Pioneer D/U ratios had a higher variance than those in recolonized systems (1.79 and 0.69, respectively; Levene's test for homogeneity of variance, P < 0.001). Detailed information on mean concentrations of Total Hg and MeHg in ponds as well as upstream and downstream sites is given in the Supporting Information (SI table S5).

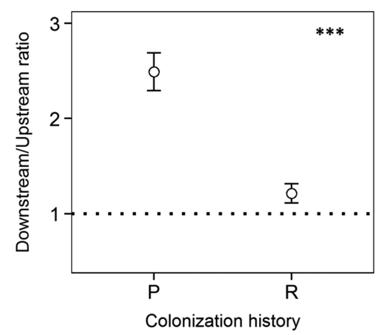

Figure 2. Mean D/U (downstream/upstream) MeHg ratios (±1 Standard Error) in pioneer (P, n = 5) and recolonized (R, n = 7) beaver systems during 2012 and 2013. The dashed line denotes the 1:1 ratio. The asterisk above whisker plots denotes a difference between pioneer and recolonized sites (*** P < 0.001).

Temporal Variation in Methylmercury. Seasonality had a strong effect on the D/U MeHg ratio (ANOVA $F_{3,3}$ = 20.02, P < 0.001), with pioneer systems showing ratios that were significantly higher than the 1:1 ratio in summer (3.52 ± 0.45), late summer (3.07 ± 0.40) and autumn (2.23 ± 0.24) (Figure 2). No significant difference from the 1:1 ratio was observed in winter and spring in pioneer beaver systems (1.66 ± 0.27 and

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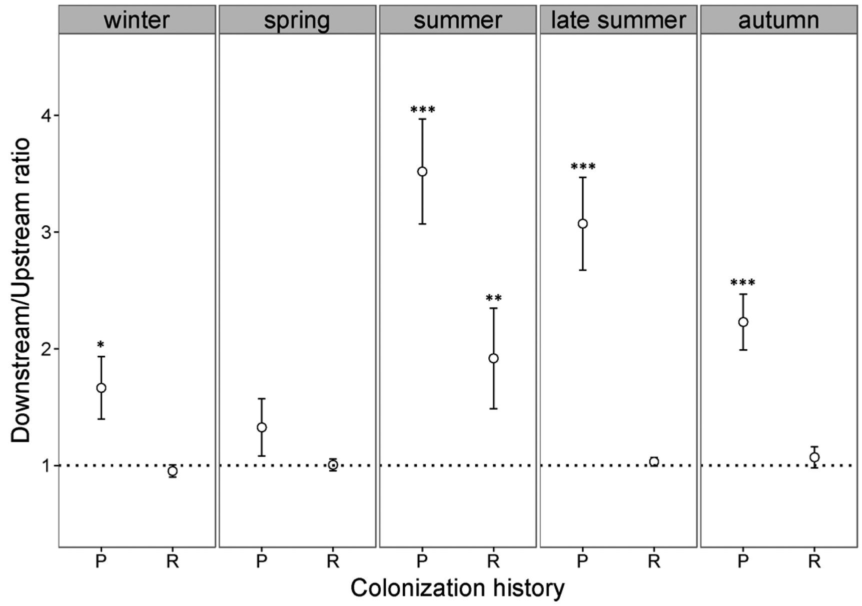

Figure 3. Mean D/U (downstream/upstream) MeHg ratios (±1 Standard Error) in pioneer (P, $n = 5$) and recolonized (R, $n = 7$) beaver systems during five seasons from November 2011 to November 2013. Please note that we only sampled one winter season (February to March 2013). The dashed line denotes the 1:1 ratio. Asterisks above whisker plots denote a D/U ratio higher than 1:1 (*** $P < 0.001$, ** $P < 0.01$, *$P < 0.05$).

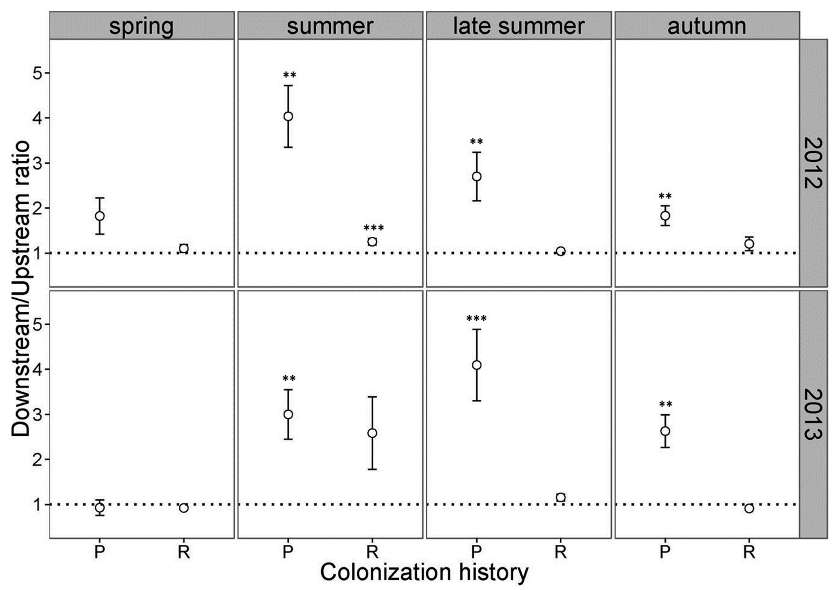

Figure 4. Mean D/U (downstream/upstream) MeHg ratios (±1 standard error) in pioneer (P, $n = 5$) and recolonized (R, $n = 7$) beaver systems during four seasons from April 2012 to November 2013. The dashed line denotes the 1:1 ratio. Asterisks above whisker plots denote a D/U ratio higher than 1:1 (*** $P < 0.001$, ** $P < 0.01$).

1.33 ± 0.25 respectively). Recolonized beaver systems showed D/U MeHg ratios significantly higher than 1:1 only in summer (1.92 ± 0.43, $P < 0.05$) (Figure 3).

The two sampling years followed strongly contrasting seasonal trajectories in terms of weather: 2012 was a wet year while 2013 was a dry year. Mean precipitation for beaver systems in 2012 and 2013, respectively, was 93 mm and 73 mm in the so-called "indifferent section" of the middle boreal zone ($n = 3$), 88 mm and 89 mm in the so-called "oceanic section" of the middle boreal zone ($n = 3$) and 110 mm and 58 mm in the south ($n = 6$). Overall, year (2012 and 2013) had no effect on the D/U MeHg ratio during spring, autumn and late summer ($P > 0.05$). However, the year had an effect in the summer (ANOVA $F_{3,3} = 5.09$, $P < 0.05$) and pioneer beaver systems had a significantly higher D/U MeHg ratio than the 1:1 ratio in both summers (2013, dry year: 3.00 ± 0.62; 2012, wet year: 4.00 ± 0.68, $P < 0.001$) (Figure 3). In contrast, recolonized systems had a significantly higher D/U ratio than 1:1 only in summer 2012 (wet year, 1.25 ± 0.08 $P < 0.01$) but not in summer 2013 (Figure 4, see also ANOVA table in SI table S4).

Pond Methylation Efficiency. Mean methylation efficiency (percentage of MeHg out of total Hg) downstream compared with upstream was significantly higher in pioneer beaver systems (14.2 ± 2.0% and 7.7 ± 1.1%, respectively, $P < 0.01$) but not in recolonized systems (9.5 ± 0.7% and 8.8 ± 0.6%, respectively, $P > 0.05$). We calculated methylation efficiency for the biggest pond of each beaver system. Overall there was a tendency for higher mean methylation efficiency in

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the pioneer than in the recolonized ponds (pioneer: 14.2 ± 2% recolonized: 9.6 ± 0.7%, $P > 0.05$). In summer (July to September), pioneer ponds had significantly higher mean methylation efficiency (19.6 ± 3.1%) than recolonized ponds (12.8 ± 0.8%, $P < 0.05$).

Other Variables Explaining MeHg Ratios. The PLS model explained 44.8% of the variance in the D/U MeHg ratio (see Table 1 for VIP and slope of the parameters in the model).

Table 1. Most Important Predictors (VIP > 0.8) of D/U (Downstream/Upstream) MeHg Ratios from the Partial Least Square regression (PLS) Analysis.[a]

variable	VIP ± SE	slope
pioneer	2.35 ± 0.16	0.12
dissolved oxygen	2.19 ± 1.04	−0.11
spring season	1.47 ± 0.70	−0.07
summer season	1.43 ± 0.85	0.08
temperature	1.37 ± 0.74	0.07
chlorophyll-a	1.02 ± 0.60	0.05
total P	0.98 ± 0.49	0.05
discharge	0.93 ± 0.66	−0.05
$NO_2^- + NO_3^-$	0.86 ± 0.61	−0.05
pH	0.84 ± 0.81	−0.04

[a]Variables are listed with their VIP's (variable importance to the projection) ±1 SE and regression slopes in descending VIP index order. The analysis explained 44.8% of the variance in D/U MeHg ratios.

The most important predictor of the D/U MeHg ratios was the colonization history, where pioneer systems were associated with higher D/U MeHg ratios and recolonized systems with lower ratios (Table 1). Dissolved oxygen in the water was also important, where D/U MeHg ratios decreased as dissolved oxygen concentrations increased. Higher D/U MeHg ratios occurred in the summer, and were associated with warmer temperatures, while spring and high discharge were related with lower ratios. Additional important predictors were concentrations of chlorophyll-*a* and total P, both associated with higher D/U MeHg ratios (Table 1). Pioneer ponds compared to recolonized ones did not have significantly higher chlorophyll-*a* concentrations (5.90 ± 1.42 μg/L and 3.34 ± 0.14 μg/L, respectively, $P > 0.05$), but total P differences were significant (pioneer: 42.08 ± 8.80 μg/L, recolonized: 14.76 ± 0.76 μg/L, $P < 0.01$). Higher $NO_2^- + NO_3^-$ concentrations and pH were associated with lower D/U MeHg ratios (Table 1). Pioneer beaver systems had lower concentrations of NO_{2^-} + NO_3^- and lower pH ($NO_2^- + NO_3^-$ pioneer: 41.00 ± 8.0 μg/L, recolonized: 90.89 ± 10.86 μg/L, $P < 0.01$; pH pioneer: 6.06 ± 0.06, recolonized: 6.50 ± 0.03, $P < 0.001$). TOC and DOC concentrations did not differ and therefore, we only used TOC in the PLS analisys. TOC, SO_4^{2-}, and Total Hg did not improve the PLS model (see also Supporting Information, SI Tables S5 and S6 for mean values and VIP's of the parameters included in the PLS analysis, respectively).

DISCUSSION

The Effect of Colonization History on MeHg Concentrations. In accordance with our hypothesis, colonization history was the most important variable explaining the observed patterns in MeHg, with MeHg elevated downstream of pioneer but not recolonized pond systems. Higher D/U MeHg ratios in young beaver ponds compared to older ones have been observed previously.[13,17,32] However, the previous studies focused solely on the age of the pond, while in this study we also considered the colonization history of the beaver systems. In pioneer systems, flooded vegetation is degraded and beavers dig in riparian soils rich in organic carbon.[49−53] Lower redox conditions in these exposed riparian and forest soils when flooded might favor increased activity by sulfur reducing bacteria. Accordingly, the elevated downstream MeHg concentrations in pioneer systems are likely to reflect the combination of anoxic or subanoxic conditions in the former riparian soils, and the high availability of degradable carbon sources for sulfur reducing bacteria and other methylators in these flooded areas. These conditions evidently do not occur to the same extent in older or recolonized ponds, with the overall result that elevated methylmercury was almost entirely associated with the pioneer systems in our study.

In the active colonization period, beavers maintain high water levels by building dams, deepening ponds and digging channels.[54] Depending on food availability, a beaver territory will be abandoned and recolonized by others or the same individuals.[20,55] Years after abandonment, dam structure and channels often continue to affect the vegetation[23,24] and hydrological pathways[22,51,54] of these abandoned systems. Upon recolonization, a beaver system may exhibit less intensive decay processes and fewer disturbances to the riparian soils and vegetation, which might result in lower MeHg concentrations in recolonized as compared to pioneer systems. This could be analogous to the finding by Tjerngren et al. (2012)[56] that restoration of wetlands (i.e., rewetting areas that have previously been wetlands) did not lead to significantly higher MeHg concentrations compared to the concentrations seen before restoration. High variation in downstream/upstream ratios in pioneer compared to recolonized systems may be the result of flooding more heterogeneous landscapes compared with recolonized ones. This aspect needs however to be further investigated. All our pioneer systems were younger than nine years old. We can therefore not infer the effect of pioneer system aging on MeHg patterns, but we might expect that older pioneer systems show similar patterns to recolonized ones. Interestingly, although our recolonized systems included the age groups "young", "intermediate" and potentially even "old" (>20 yrs) defined in Roy et al. (2009)[32] (i.e., in our study: young $n = 1$, intermediate $n = 4$ and old (>18 yrs in our study) $n = 2$), we found no evidence for variation in methylation efficiency or downstream/upstream ratio among them. Rather, it was only the pioneer beaver systems that had significantly increased pond methylation efficiency and MeHg concentrations downstream of the ponds overall. Our results therefore support the contention that it is the time since first inundation, rather than the age of any individual pond, or of the beaver system per se that primarily determines MeHg levels in beaver ponds. Reconstruction of dams on previously flooded systems, even when very recent, are thus likely to have less impact on MeHg levels than newly constructed dams on systems that have never been flooded previously.

There is increasing scientific support that photolytically driven demethylation in surface water may be the largest sink of MeHg in lakes and open water bodies.[42,57−61] Compared to recolonized beaver ponds which tend to have larger open water area, pioneer beaver systems are typically covered by snags and logs that result in less open water surface[22] and hence potentially lower rates of photolytically driven demethylation. To better understand the importance of recolonization history

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for MeHg concentrations, we need to further investigate how the processes regulating the balance between methylation and demethylation change as beaver pond systems age.

Temporal Variation in Methylmercury. Roy et al. (2009)[17] reported that MeHg concentrations in beaver ponds peak during the warmer season and that their beaver systems experienced an increase in MeHg at outlets relative to inlets. They sampled three ponds during one year from March to September; young (<10 years, n = 1), intermediate (10−20 years, n = 1) and old (>20 years, n = 1). In April and May (in the old pond) and May (in all ponds combined), MeHg concentrations did not increase from inlets to outlets.[17] In our study methylation efficiency in beaver ponds varied seasonally, generally peaking in the warmest months of the year, and more evidently in the pioneer than recolonized systems. In pioneer beaver ponds, warmer water temperature in the summer and autumn likely in combination with the availability of organic matter[18] can create the reducing environment that increases the activity of sulfate reducing bacteria and promotes methylation.[62] In contrast, older organic material exposed to inundation for many years, and larger open water bodies which promote demethylation may have resulted in lower methylation efficiency in recolonized compared to pioneer ponds during summer. Nevertheless, even in the recolonized ponds there was evidence for a slight increase in MeHg concentrations downstream of the ponds during the warmer months. This demonstrates that these systems can continue to be sites for methylation during some times of year, albeit at much lower rates than the pioneer ponds.

The PLS analysis indicated that apart from colonization history, oxygen concentration was the single most important predictor for D/U MeHg ratio, with MeHg concentrations increasing as oxygen concentrations decline. This is in agreement with other studies (reviewed by Ullrich et al. and shown for beaver ponds by Roy et al.).[17,32,42] Chlorophyll-*a*, total P and NO_2^-+NO_3^- levels were also predictors of D/U MeHg ratios, reflecting the importance of nutrient availability and biological activity which is also enhanced in the warmer period of the year. Methylation often takes place where nutrient availability supports microbial activity; often in the upper layer of the sediment and on suspended organic material (Reviewed by Ullrich et al.).[42] Although interference of DOC in the MeHg analysis might create artifacts, DOC and TOC did not improve the PLS model and was not an important predictor for MeHg concentrations. Since no increase in turbidity or particulate organic carbon could be detected downstream from beaver ponds, regardless of their history, we do not believe that particulates are involved where MeHg increases. To further elucidate temporal variation of MeHg in beaver ponds, closer investigation is required into the relationship between oxygen, temperature and nutrient availability for processes occurring in the sediment (methylation) and the upper surface layer (demethylation) in beaver systems with different colonization histories.

MeHg at the Landscape Scale and Management Implications. The boreal forest landscape developed together with the engineering activity of beavers.[63] Over the past 150 years, this link was broken, and the near extirpation of beavers combined with vast anthropogenic impacts, including forestry, has driven comprehensive changes in forest stream ecosystems.[64,65] Today, following successful conservation efforts, we may witness the "natural restoration" of these boreal stream habitats[66,67] as beavers reoccupy large areas in the boreal zone. Due to the ongoing expansion of the beaver's distribution range, limited availability of suitable pioneer habitats, and increases in population sizes, the proportion of pioneer beaver systems is expected to decrease in the future.[30,68,69] This implies that the effect of pioneer dams on MeHg at the landscape scale will decline with time, as more systems enter the older, recolonized state. Due to the similarity in the ecology, behavior (e.g., regarding the building of dams) and life history of *C. fiber* and *C. canadensis*,[70,71] we expect this effect to apply not only to Eurasia where both species occur but also to North America where only *C. Canadensis* occurs. However, allowing beaver systems to "mature" requires particular management strategies. In many European countries, *C. fiber* was initially (1992) included as an Annex II species of the Habitats Directive, that is, a species of community interest whose conservation requires the designation of special areas of conservation.[72] As a result of these conservation efforts, the species showed remarkable recovery and in 2007 was excluded as an Annex II species of the Habitats Directive in Finland, Sweden, Lithuania, Latvia, and Estonia.[73] In addition, the Eurasian beaver was declared "near threatened" in 2002 and since 2008 it is "of least concern" with an increasing population trend according to the International Union for Conservation of Nature Red List of Threatened Species.[74] In most Scandinavian countries (including Sweden), beaver hunting is limited only by season (not by quota) and often land owners use dam and lodge removal as a mitigation action if beaver ponds conflict with human interests. Intensive hunting and removal of beaver systems may prevent beaver systems from reaching the successional stage of recolonized systems which, according to our results show D/U MeHg ratios close to 1:1 or even less. Therefore, we recommend beaver management policies to consider the potential benefit of maintaining natural processes of aged and recolonized beaver systems toward reducing the environmental burden of MeHg.

■ ASSOCIATED CONTENT

Supporting Information

The Supporting Information is available free of charge on the ACS Publications website at DOI: 10.1021/acs.est.5b03146.

land cover characteristics of the watersheds of the beaver systems, tables on the classification of colonization history and age of beaver systems, respectively, tables summarizing the results of the ANOVA model and mean values as well as VIP's, SE and slopes of the parameters included in the PLS analysis, analyses on the potential interaction between colonization history and age, and MeHg quality control and method validation (PDF)

■ AUTHOR INFORMATION

Corresponding Author

*E-mail: oded.levanoni@slu.se.

Notes

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REFERENCES

(1) Clarkson, T. W.; Magos, L. The toxicology of mercury and its chemical compounds. *Crit. Rev. Toxicol.* **2006**, *36* (8), 609−662.

(2) *Evaluation of Certain Food Additives and Contaminants*, Sixty-first report of the Joint FAO/WHO Expert Committee on Food Additives; World Health Organization: Geneva, 2004, 132−139.

(3) Water Quality Criterion for the Protection of Human Health: Methylmercury. EPA-823-R-01−001, United States Environmental Protection Agency: Washington DC, 2001; http://water.epa.gov/scitech/swguidance/standards/criteria/health/upload/2009_01_15_criteria_methylmercury_mercury-criterion.pdf.

(4) EU. Directive 2008/105/EC of the European Parliament and of the Council of 16 December 2008 on environmental quality standards in the field of water policy. *Official Journal of the European Union* **2008**, 51/L348, 84−97.

(5) Johansson, K.; Bergbäck, B.; Tyler, G. Impact of atmospheric long-range transport of lead, mercury and cadmium on the Swedish forest environment. *Water, Air, Soil Pollut.: Focus* **2001**, *1*, 279−296.

(6) Åkerblom, S.; Nilsson, M.; Yu, J.; Ranneby, B.; Johansson, K. Temporal change estimation of mercury concentrations in northern pike (*Esox lucius* L.) in Swedish lakes. *Chemosphere* **2012**, *86* (5), 439−445.

(7) Munthe, J., Rognerud, S., Fjeld, E., Verta, M., Porvari, P. & Meili, M. *Mercury in Nordic Ecosystems*; IVL Swedish Environmental Research Institute, 2007.

(8) Compeau, G. C.; Bartha, R. Sulfate-reducing bacteria - principal methylators of Mercury in anoxic esturaine sediment. *Appl. Environ. Microbiol.* **1985**, *50* (2), 498−502.

(9) Gilmour, C. C.; Henry, E. A.; Mitchell, R. Sulfate stimulation of Mercury methylation in fresh-water sediments. *Environ. Sci. Technol.* **1992**, *26* (11), 2281−2287.

(10) Skyllberg, U.; Drott, A.; Lambertsson, L.; Björn, E.; Karlsson, T.; Johnson, T.; Heinemo, S-Å.; Holmström, H. Net methylmercury production as a basis for improved risk assessment of mercury-contaminated sediments. *Ambio* **2007**, *36* (6), 437−442.

(11) Benoit, J. M.; Gilmour, C. C.; Heyes, A.; Mason, R. P.; Miller, C. L., Geochemical and biological controls over methylmercury production and degradation in aquatic ecosystems. In *Biogeochemistry of Environmentally Important Trace Elements*, Cai, Y., Braids, O. C., Eds. **2003**; Vol. *835*, 262−297.10.1021/bk-2003-0835.ch019

(12) Eklöf, K.; Fölster, J.; Sonesten, L.; Bishop, K. Spatial and temporal variation of THg concentrations in run-off water from 19 boreal catchments, 2000−2010. *Environ. Pollut.* **2012**, *164*, 102−109.

(13) Driscoll, C. T.; Holsapple, J.; Schofield, C. L.; Munson, R. The chemistry and transport of mercury in a small wetland in the Adirondack region of New York, USA. *Biogeochemistry* **1998**, *40* (2−3), 137−146.

(14) Hall, B. D.; Louis, V. L. S.; Rolfhus, K. R.; Bodaly, R. A.; Beaty, K. G.; Paterson, M. J.; Cherewyk, K. A. P. Impacts of Reservoir Creation on the Biogeochemical Cycling of Methyl Mercury and Total Mercury in Boreal Upland Forests. *Ecosystems* **2005**, *8* (3), 248−266.

(15) St. Louis, V. L.; Rudd, J. W. M.; Kelly, C. A.; Bodaly, R. A.; Paterson, M. J.; Beaty, K. G.; Hesslein, R. H.; Heyes, A.; Majewski, A. R. The Rise and Fall of Mercury Methylation in an Experimental Reservoir. *Environ. Sci. Technol.* **2004**, *38* (5), 1348−1358.

(16) Selvendiran, P.; Driscoll, C. T.; Bushey, J. T.; Montesdeoca, M. R. Wetland influence on mercury fate and transport in a temperate forested watershed. *Environ. Pollut.* **2008**, *154* (1), 46−55.

(17) Roy, V.; Amyot, M.; Carignan, R. Seasonal methylmercury dynamics in water draining three beaver impoundments of varying age. *J. Geophys. Res.* **2009**, *114*, G00C06.

(18) Rosell, F.; Bozser, O.; Collen, P.; Parker, H. Ecological impact of beavers *Castor fiber* and *Castor canadensis* and their ability to modify ecosystems. *Mammal Review* **2005**, *35* (3−4), 248−276.

(19) Meentemeyer, R. K.; Butler, D. R. Hydrogeomorphic effects of beaver dams in Glacier National Park, Montana. *Physical Geography* **1999**, *20* (5), 436−446.

(20) Collen, P.; Gibson, R. J. The general ecology of beavers (*Castor* spp.), as related to their influence on stream ecosystems and riparian habitats, and the subsequent effects on fish - a review. *Reviews in Fish Biology and Fisheries* **2000**, *10* (4), 439−461.

(21) Kemp, P. S.; Worthington, T. A.; Langford, T. E. L.; Tree, A. R. J.; Gaywood, M. J. Qualitative and quantitative effects of reintroduced beavers on stream fish. *Fish and Fisheries* **2012**, *13* (2), 158−181.

(22) Naiman, R. J.; Johnston, C. A.; Kelley, J. C. Alteration of North-American streams by beaver. *BioScience* **1988**, *38* (11), 753−762.

(23) Wright, J. P. Linking populations to landscapes: richness scenarios resulting from changes in the dynamics of an ecosystem engineer. *Ecology* **2009**, *90* (12), 3418−3429.

(24) Wright, J. P.; Flecker, A. S.; Jones, C. G. Local vs. landscape controls on plant species richness in beaver meadows. *Ecology* **2003**, *84* (12), 3162−3173.

(25) Howard, R. J.; Larson, J. S. A stream habitat classification-system for beaver. *J. Wildl. Manage.* **1985**, *49* (1), 19−25.

(26) Fryxell, J. M. Habitat suitability and source-sink dynamics of beavers. *J. Anim. Ecol.* **2001**, *70* (2), 310−316.

(27) Zavyalov, N. A. Dynamics of food resources for beavers in settlements colonized and abandoned several times. *Biology Bulletin* **2013**, *40* (10), 872−878.

(28) Nolet, B. A.; Rosell, F. Comeback of the beaver *Castor fiber*: An overview of old and new conservation problems. *Biological Conservation* **1998**, *83* (2), 165−173.

(29) Ducroz, J. F.; Stubbe, M.; Saveljev, A. P.; Heidecke, D.; Samjaa, R.; Ulevicius, A.; Stubbe, A.; Durka, W. Genetic variation and population structure of the Eurasian beaver *Castor fiber* in Eastern Europe and Asia. *J. Mammal.* **2005**, *86* (6), 1059−1067.

(30) Hartman, G., The case of the Eurasian beaver in Sweden: re-introduction project carried out before the existence of re-introduction guidelines! In *Global Re-Introduction Perspectives: 2011*; Soorae, P. S., Ed.; International Union for Conservation of Nature 2011; 165−167.

(31) Hartman, G., The beaver (*Castor fiber*) in Sweden. In *Restoring the European Beaver: 50 Years of eExperience*; Sjöberg, G., Ball, J. P., Eds.; Pensoft; Sofia & Moscow, 2011; pp 13−17.

(32) Roy, V.; Amyot, M.; Carignan, R. Beaver Ponds Increase Methylmercury Concentrations in Canadian Shield Streams along Vegetation and Pond-Age Gradients. *Environ. Sci. Technol.* **2009**, *43* (15), 5605−5611.

(33) Sjörs, H. The background: Geology, climate and zonation. *Acta phytogeogr. Suec.* **1999**, *84*, 5−14.

(34) ESRI (Environmental Systems Research Institute). *Arc Map*, 10.2; 2013.

(35) Lambertsson, L.; Björn, E. Validation of a simplified field-adapted procedure for routine determinations of methyl mercury at trace levels in natural water samples using species-specific isotope dilution mass spectrometry. *Anal. Bioanal. Chem.* **2004**, *380* (7−8), 871−875.

(36) Method 1631: Measurement of Mercury in Water; Revision E. EPA-821-R-02-019, United States Environmental Protection Agency: Washington DC, 2002; http://water.epa.gov/scitech/methods/cwa/metals/mercury/upload/2007_07_10_methods_method_mercury_1631.pdf.

(37) Baxter, D. C.; Faarinen, M.; Österlund, H.; Rodushkin, I.; Christensen, M. Serum/plasma methylmercury determination by isotope dilution gas chromatography-inductively coupled plasma mass spectrometry. *Anal. Chim. Acta* **2011**, *701* (2), 134−138.

(38) Fölster, J.; Johnson, R. K.; Futter, M. N.; Wilander, A. The Swedish monitoring of surface waters: 50 years of adaptive monitoring. *Ambio* **2014**, *43*, 3−18.

(39) PTHBV temperature and precipitation grid database. Swedish Meteorological Survey http://luftweb.smhi.se/ (accessed 12 November 2014).

(40) Eriksson, L.; Johansson, E.; Kettaneh-Wold, N.; Trygg, J.; Wikström, C.; Wold, S. *Multi- and Megavariate Data Analysis Part I Basic Principles and Applications*, 2nd ed.; Umetrics AB Umeå, SE, 2006.

(41) Galloway, M. E.; Branfireun, B. A. Mercury dynamics of a temperate forested wetland. *Sci. Total Environ.* **2004**, *325* (1−3), 239−254.

DOI: 10.1021/acs.est.5b03146
Environ. Sci. Technol. 2015, 49, 12679−12687

(42) Ullrich, S. M.; Tanton, T. W.; Abdrashitova, S. A. Mercury in the aquatic environment: A review of factors affecting methylation. *Crit. Rev. Environ. Sci. Technol.* **2001**, *31* (3), 241−293.
(43) Brigham, M. E.; Wentz, D. A.; Aiken, G. R.; Krabbenhoft, D. P. Mercury Cycling in Stream Ecosystems. 1. Water Column Chemistry and Transport. *Environ. Sci. Technol.* **2009**, *43* (8), 2720−2725.
(44) Wasik, J. K. C.; Mitchell, C. P. J.; Engstrom, D. R.; Swain, E. B.; Monson, B. A.; Balogh, S. J.; Jeremiason, J. D.; Branfireun, B. A.; Eggert, S. L.; Kolka, R. K.; Almendinger, J. E. Methylmercury Declines in a Boreal Peatland When Experimental Sulfate Deposition Decreases. *Environ. Sci. Technol.* **2012**, *46* (12), 6663−6671.
(45) Tjerngren, I.; Karlsson, T.; Björn, E.; Skyllberg, U. Potential Hg methylation and MeHg demethylation rates related to the nutrient status of different boreal wetlands. *Biogeochemistry* **2012**, *108* (1−3), 335−350.
(46) SAS. *JMP*, 10.0; 2012.
(47) R Core Team. R: A language and environment for statistical computing, R foundation for statistical computation, Viena, Austria. 3.1.0; 2014. http://www.R-peoject.org/.
(48) UMETRIX. *SIMCA-P*, 13.0; 2012.
(49) Wohl, E. Landscape-scale carbon storage associated with beaver dams. *Geophys. Res. Lett.* **2013**, *40* (14), 3631−3636.
(50) Margolis, B. E.; Castro, M. S.; Raesly, R. L. The impact of beaver impoundments on the water chemistry of two Appalachian streams. *Can. J. Fish. Aquat. Sci.* **2001**, *58* (11), 2271−2283.
(51) Naiman, R. J.; Melillo, J. M.; Hobbie, J. E. Ecosystem alteration of boreal forest streams by beaver (*Castor canadensis*). *Ecology* **1986**, *67* (5), 1254−1269.
(52) Smith, M. E.; Driscoll, C. T.; Wyskowski, B. J.; Brooks, C. M.; Cosentini, C. C. Modification of stream ecosystem structure and function by beaver (*Castor canadensis*) in the Adirondack mountains, New-York. *Can. J. Zool.* **1991**, *69* (1), 55−61.
(53) Correll, D. L.; Jordan, T. E.; Weller, D. E. Beaver pond biogeochemical effects in the Maryland Coastal Plain. *Biogeochemistry* **2000**, *49* (3), 217−239.
(54) Hood, G. A.; Bayley, S. E. Beaver (*Castor canadensis*) mitigate the effects of climate on the area of open water in boreal wetlands in western Canada. *Biological Conservation* **2008**, *141* (2), 556−567.
(55) Fustec, J.; Lode, T.; Le Jacques, D.; Cormier, J. P. Colonization, riparian habitat selection and home range size in a reintroduced population of European beavers in the Loire. *Freshwater Biol.* **2001**, *46* (10), 1361−1371.
(56) Tjerngren, I.; Meili, M.; Björn, E.; Skyllberg, U. Eight Boreal Wetlands as Sources and Sinks for Methyl Mercury in Relation to Soil Acidity, C/N Ratio, and Small-Scale Flooding. *Environ. Sci. Technol.* **2012**, *46* (15), 8052−8060.
(57) Lehnherr, I.; St Louis, V. L.; Emmerton, C. A.; Barker, J. D.; Kirk, J. L. Methylmercury Cycling in High Arctic Wetland Ponds: Sources and Sinks. *Environ. Sci. Technol.* **2012**, *46* (19), 10514−10522.
(58) Sellers, P.; Kelly, C. A.; Rudd, J. W. M. Fluxes of methylmercury to the water column of a drainage lake: The relative importance of internal and external sources. *Limnol. Oceanogr.* **2001**, *46* (3), 623−631.
(59) Sellers, P.; Kelly, C. A.; Rudd, J. W. M.; MacHutchon, A. R. Photodegradation of methylmercury in lakes. *Nature* **1996**, *380* (6576), 694−697.
(60) Hammerschmidt, C. R.; Fitzgerald, W. F. Photodecomposition of methylmercury in an arctic Alaskan lake. *Environ. Sci. Technol.* **2006**, *40* (4), 1212−1216.
(61) Hammerschmidt, C. R.; Fitzgerald, W. F.; Lamborg, C. H.; Balcom, P. H.; Tseng, C. M. Biogeochemical cycling of methylmercury in lakes and tundra watersheds of Arctic Alaska. *Environ. Sci. Technol.* **2006**, *40* (4), 1204−1211.
(62) Tsui, M. T. K.; Finlay, J. C. Influence of Dissolved Organic Carbon on Methylmercury Bioavailability across Minnesota Stream Ecosystems. *Environ. Sci. Technol.* **2011**, *45* (14), 5981−5987.
(63) Rybczynski, N. Castorid phylogenetics: implications for the evolution of swimming and tree-exploitation in beavers. *Journal of Mammalian Evolution* **2007**, *14* (1), 1−35.
(64) Essen, P.-A.; Ehnstrom, B.; Ericson, L.; Sjoberg, K. *Boreal forests: The focal habitats of Fennoscandia* **1992**, *1*, 252−325.
(65) Schindler, D. W., Sustaining aquatic ecosystems in boreal regions. *Conserv. Ecol.* **1998**, *2*, (2): article 18.
(66) DeVries, P.; Fetherston, K. L.; Vitale, A.; Madsen, S. Emulating Riverine Landscape Controls of Beaver in Stream Restoration. *Fisheries* **2012**, *37* (6), 246−255.
(67) McColley, S. D.; Tyers, D. B.; Sowell, B. F. Aspen and Willow Restoration Using Beaver on the Northern Yellowstone Winter Range. *Restoration Ecology* **2012**, *20* (4), 450−455.
(68) Hartman, G. Long term population development of a reintroduced beaver (*Castor fiber*) population in Sweden. *Conservation Biology* **1994**, *8* (3), 713−717.
(69) Hartman, G. Habitat selection by European beaver (*Castor fiber*) colonizing a boreal landscape. *J. Zool.* **1996**, *240*, 317−325.
(70) Danilov, P. I.; Fyodorov, F. V. Comparative characterization of the building activity of Canadian and European beavers in northern European Russia. *Russ. J. Ecol.* **2015**, *46* (3), 272−278.
(71) Parker, H.; Nummi, P.; Hartman, G.; Rosell, F. Invasive North American beaver Castor canadensis in Eurasia: a review of potential consequences and a strategy for eradication. *Wildlife Biology* **2012**, *18* (4), 354−365.
(72) On the conservation of natural habitats and of wild fauna and flora. Council Directive 92/43/EEC, 1992; Annex II, pp 206/22−206/37. http://eur-lex.europa.eu/legal-content/EN/TXT/?uri=OJ:L:1992:206:TOC.
(73) On the Conservation of Natural Habitats and of Wild Fauna and Flora. version 1.1.2007.; Council Directive 92/43/EEC, 2007; Annex II, pp 24−48. http://eur-lex.europa.eu/legal-content/EN/TXT/PDF/?uri=CELEX:01992L0043-20070101&from=EN.
(74) Batbold, J.; Batsaikhan, N.; Shar, S.; Amori, G.; Hutterer, R.; Kryštufek, B.; Yigit, N.; Mitsain, G.; Palomo, L. J. *Castor Fiber. The IUCN Red List of Threatened Species*, Version 2014.2, 2008.

DOI: 10.1021/acs.est.5b03146
Environ. Sci. Technol. 2015, 49, 12679−12687