

RESEARCH ARTICLE

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Key Points:

- Lake browning can promote carbon and mercury burial in boreal lake sediment
- Increase in Hg burial corresponded to the magnitude of browning
- Respiration mode strongly affects OC mineralization and Hg methylation

Supporting Information:

- Supporting Information S1

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The effect of lake browning and respiration mode on the burial and fate of carbon and mercury in the sediment of two boreal lakes

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Abstract In many northern temperate regions, the water color of lakes has increased over the past decades ("lake browning"), probably caused by an increased export of dissolved organic matter from soils. We investigated if the increase in water color in two lakes in Norway has resulted in increased burial of organic carbon (OC) and mercury (Hg) in the sediments and if the Hg was prone to methylation. Lake Solbergvann experienced a threefold water color increase, and OC burial increased approximately twofold concomitant to the water color increase. This lake had prolonged periods of anoxic bottom water, and anoxic OC mineralization rates were only about half of the oxic OC mineralization rates (7.7 and 17.5 g C m⁻² yr⁻¹, respectively), contributing to an efficient OC burial. In Lake Elvåga, where water color increase was only approximately twofold and bottom water was oxygenated, no recent increase in OC burial could be observed. Hg burial increased strongly in both lakes (threefold and 1.6-fold in Lake Solbergvann and Lake Elvåga, respectively), again concomitant to the recent water color increase. The proportion of methylated Hg (MeHg) in surficial sediment was 1 order of magnitude higher in Lake Elvåga (up to 6% MeHg) than in Lake Solbergvann (0.2–0.6% MeHg), probably related to the different oxygenation regimes. We conclude that lake browning can result in increased OC and Hg burial in lake sediments, but the extent of browning and the dominating mode of sediment respiration (aerobic or anaerobic) strongly affect burial and fate of OC and Hg in sediments.

1. Introduction

The important role of inland waters in the carbon cycle is increasingly being recognized. Estimates of carbon dioxide (CO₂) and methane (CH₄) emissions from inland waters point toward a pivotal role of inland waters in the carbon balance of the continents [Bastviken et al., 2011; Raymond et al., 2013]. However, inland waters not only emit greenhouse gases but also bury carbon in their sediments. Globally, the sediments of lakes and reservoirs are estimated to bury more OC than the entire ocean [Tranvik et al., 2009]. The role of detrital organic matter (OM) in lake ecosystem functioning has early been recognized [Lindeman, 1942] and has been shown to be an important carbon and energy source for aquatic secondary production [Tranvik, 1988]. Accordingly, the organic carbon (OC) that reaches the lake sediment surface will partly be mineralized to CO₂ or CH₄ by heterotrophic organisms, and the remainder will be buried in the sediments. Important regulators of the ratio between organic carbon mineralization and burial are temperature, oxygen exposure time, and the reactivity of organic matter [Gudasz et al., 2010; Sobek et al., 2009]. Lake sediments generally contain a mixture of OC derived from either internal production or from terrestrial sources, but while OC derived from lake-internal primary production to a large extent is mineralized and assimilated by sediment bacteria [Gudasz et al., 2012; Sobek et al., 2014], terrestrial OC is rather resistant to degradation and therefore prone to long-term burial in lake sediments [Chmiel et al., 2015; Sobek et al., 2009].

An increase in water color, usually called "water browning," has been reported from many studies across the Northern Hemisphere [Hongve et al., 2004; Monteith et al., 2007]. It has been ascribed to an increased export of colored dissolved organic carbon (DOC) from catchment soils [Forsberg and Petersen, 1990], although other factors such as residence time, pH, and iron may also affect water color [Köhler et al., 2013]. Since most of the OC export from boreal catchments is in the dissolved form [Mattsson et al., 2005], particulate export is generally regarded less important, although erosion rates and particulate OC transport may change in response to changes in climate. There are several potential explanations for the browning of lakes in the

boreal zone. First, it has been explained by climate changes, such as an increase in temperature, precipitation, and CO₂ concentration in the air [Freeman *et al.*, 2001; Hongve *et al.*, 2004]. Second, it has been related to interannual variation in discharge [Erlandsson *et al.*, 2008]. Third, browning might also be attributable to catchment recovery from acidification (i.e., reduction of sulfate in precipitation), lowering the ionic strength of soil pore water and thereby mobilizing DOC [Haaland *et al.*, 2010; Monteith *et al.*, 2007]. These different potential causes of browning all result in an increased loading of colored material from catchments to lakes.

Processes that contribute to a retention or mobilization of DOC in lakes are still not fully understood. Land-derived DOC is prone to flocculation and sedimentation [von Wachenfeldt and Tranvik, 2008]; in fact, a large share of the OC in boreal lake sediments has been shown to originate from flocculation of DOC [von Wachenfeldt and Tranvik, 2008]. Iron (Fe) has been shown to coflocculate and settle out with DOC [von Wachenfeldt *et al.*, 2008], and a large fraction of the land-derived colored OC has been shown to have a high molecular mass, which is associated with Fe [Riise, 1999]. This fraction increased during high flow and probably settled during low flow periods [Riise, 1999]. Also, photochemical reactions and microbial activity have been shown to be related to DOC flocculation rates [von Wachenfeldt *et al.*, 2008; Wachenfeldt *et al.*, 2009]. Even though one study has indicated that increased lake browning may result in increased flocculation and sedimentation in the water column [von Wachenfeldt *et al.*, 2008], it is presently not known if the increased carbon load to lakes also leads to an increased burial of carbon in sediments.

Dissolved organic matter (DOM) is known to interact very strongly with mercury (Hg), affecting its biogeochemical cycling in aquatic environments [Ravichandran, 2004]. DOM is the main vector for mercury and methylmercury (MeHg) from catchment soils to surface waters in boreal catchments [Grigal, 2002]. Total mercury (THg) and OC concentrations therefore often correlate in streams and water columns [Braaten *et al.*, 2014; Meili *et al.*, 1991; Watras *et al.*, 1995]. Soils are therefore not only an important source of OC but also Hg to lakes, particularly since soils have accumulated a large pool of Hg due to atmospheric deposition during the industrial era [Johansson *et al.*, 2001], even if OC and Hg may not always be sourced from the same soil type or horizon [Dittman *et al.*, 2010]. Hg binds strongly to sulfur species such as inorganic sulfides and organic thiols [Skylberg, 2008]. The decrease in soil ionic strength, due to the decline of atmospheric acid deposition, can mobilize soil OC [Monteith *et al.*, 2007] but might also enhance the transport of organically bound Hg to lakes [Hongve *et al.*, 2012]. An enhanced terrestrial Hg load due to recent lake browning has been linked to increased Hg concentrations in fish [Hongve *et al.*, 2012]. A fraction (0.1–15%) of inorganic Hg (IHg) can be transformed to MeHg under suboxic and anoxic conditions in stratified water columns and/or lake sediments during anaerobic respiration of OM by bacteria [Fleming *et al.*, 2006; Gilmour *et al.*, 2011; King *et al.*, 2000]. MeHg is readily taken up by aquatic organisms at the base of food webs and is thereafter biomagnified, providing the major pathway of human Hg exposure via fish consumption [United Nations Environment Programme, 2013]. Similarly to the questions regarding the fate of OC deposited on lake sediment, there is a need to examine if the enhanced Hg load to lakes due to lake browning may result in an enhanced Hg burial and/or a higher MeHg production, since the latter could potentially trigger an increased Hg concentrations in fish as suggested by Hongve *et al.* [2012].

To the best of our knowledge, detailed studies linking the recent increase of terrestrial OM load (i.e., browning) to the burial and fate of OC and Hg in lake sediments are lacking. We, therefore, conducted a study on the burial and fate of OC and Hg in the sediments of two contrasting boreal lakes that have, to a different degree, experienced a recent increase in water color. With the term “fate,” we refer to the fact that OC can either remain in the solid phase of the sediment (i.e., be buried) or be transferred to more mobile species like CO₂ and CH₄ by respiration; these mobile species can then either leave the sediment by, e.g., diffusion or be taken up by organisms. With fate of Hg we similarly refer to either burial of Hg in sediment or methylation to MeHg, which implies a greater possibility for uptake by organisms. We hypothesized that (1) an increase in water color will be followed by an increase in OC and Hg burial in sediments and (2) the mineralization rates of recently deposited OC in the sediment will affect the extent of OC burial as well as Hg methylation in sediments.

2. Materials and Methods

2.1. Site Description and Sampling

We tested our hypotheses in two lakes close to the city of Oslo, Norway, where an increase in water color has been evidenced [Haaland *et al.*, 2010; Hongve *et al.*, 2004]. The two studied lakes, Lake Solbergvann and Lake Elvåga, have experienced different extents of water color increase (see section 3) and may therefore be

Table 1. Description of Lake Solbergvann and Lake Elvåga^a

	Lake Solbergvann	Lake Elvåga
Coordinates	59°53.999'N 10°51.982'E	59°51.485'N 10°55.182'E
Altitude (m)	235	195
Surface area (ha)	1.25	19.6
Maximum depth (m)	5	23
Catchment area (km ²)	0.13	11.8
Catchment-to-lake area	10	60
Land use	90.4% coniferous forest 9.6% Lake Solbergvann	86.1% coniferous forest 8.4% wetlands 3.8% other lakes 1.7% Lake Elvåga

^aCatchment characteristics presented here are for the southern basin of Lake Elvåga.

regarded as examples of boreal lakes that experience moderate to severe browning. The lakes are situated in a protected boreal forest area, Østmarka, located 7–9 km from Oslo city center (Table 1). The lakes are rather pristine, as severe restrictions are put on human activities in the area. Lake Elvåga is a drinking water source for Oslo municipality. The major part of the Lake Elvåga catchment (64%) drains into a southern basin. Hence, the southern basin receives the largest part of the terrestrial input to the lake and was therefore chosen for sampling. The southern and northern basins of Lake Elvåga were previously separate basins but were connected with each other in 1964, as a dam was built at the end of the northern basin to enlarge the drinking water reservoir, and the water level was raised. The present water level is regulated for drinking water purposes. Lake Elvåga is a rather nutrient-poor lake, moderately rich in dissolved humic matter (Table 2). The other lake, Lake Solbergvann, is a small forest headwater lake, highly colored and with low ionic strength (Tables 1 and 2). The catchment to lake area ratio is 6 times lower in Lake Solbergvann than in Lake Elvåga (Table 1). More information about land uses, surface area, altitude, and chemical parameters are presented in Tables 1 and 2. The time series of the water color (Figure 1) are based on our data for Lake Solbergvann and the Oslo Water and Sewerage Works for the northern basin of Lake Elvåga.

Lake Solbergvann and the southern basin of Lake Elvåga were sampled twice in their deepest part, once in the beginning of September 2012 and once in the middle of May 2013. This way, the samplings include seasonal variability in stratification and thus bottom water oxygenation. Profiles of oxygen, conductivity, and temperature in the water column were measured in situ with an HQ40d portable multiparameter meter (HACH). pH was measured with a glass electrode (Metrohm) in the water overlying the sediment after the cores had been brought to the shore. Unfiltered water samples were collected with a Ruttner water sampler for THg and MeHg measurements. Water samples were collected in the oxic part of the water column of both lakes and in the case of Lake Solbergvann also in the oxygen-depleted hypolimnion (5 m depth). About 1 L of water per depth was placed in Teflon® bottles and immediately acidified (1 % vol/vol final concentration; HCl Ultrex II, J. T. Baker®) on the boat.

Table 2. Lake Solbergvann and Lake Elvåga Physical and Chemical Parameters^a

	Lake Solbergvann		Lake Elvåga	
	September	May	September	May
Bottom water temperature (°C)	6.8	4.3	5.5	4.0
Secchi depth (m)	1.6	1.5	2.4	2.5
Conductivity (μS cm ⁻¹)	25	22	24	21
Water color (mg Pt L ⁻¹)	142	-	67	-
TOC (mg L ⁻¹)	15.4	11.8	8.3	6.5
TotP (μg L ⁻¹)	12	-	6	-
pH	5.9	5.6	6.2	5.9
Oxygen penetration depth (mm)	0	0	3.1 ± 1.0	2.5 ± 0.5

^aLake parameters were measured on 5 Sept 2012 and 14 May 2013.

Sediment cores were taken with a UWITEC gravity corer at the deepest spot of each lake basin. The deepest part was chosen for sediment coring in order to sample sediments that experience the maximal sediment accumulation rate and therefore allow for detection of potential recent trends in accumulation. Thereby, the observed rates of sediment accumulation are not representative for the entire lake basin. Samples for dissolved methane in sediment pore water were taken from core liners equipped with side ports every 1 cm of the core. Pore water from three cores from each lake was subsampled at 1 cm depth intervals for the upper 10 cm and at 2 cm intervals for the next 20 cm of the sediment within an hour after obtaining the cores. The samples were fixed following *Sobek et al.* [2009]. In order to measure total carbon, nitrogen, and sulfur in the dry sediment, one core from each lake was sliced into 0.5 cm intervals for the first 10 cm and 1 cm intervals for the next 30 cm, immediately after sampling. Additional cores were stored cold and dark until arrival at the laboratory of the Norwegian University of Life Sciences. One of those was used for THg and MeHg analyses, and six were used for the measurement of mineralization rates.

2.2. Sediment Mineralization Rates

Respiration rates in the sediments were determined by oxic incubations of the sediment cores and anoxic incubations of the sediment cores (only in Lake Solbergvann).

For the incubation experiments, about 5 cm of the sediment in the sampling core was carefully transferred into incubation cores (54 mm in diameter and 400 mm high). These cores were then filled with bottom water from the corresponding lake according to *Gudas et al.* [2010]. Prior to oxic incubation, the experiment cores were aerated until the water above the sediments was fully oxygenated. The cores were tightly closed without headspace. Dissolved inorganic carbon (DIC) samples were taken from each core through tubing mounted on the stoppers. Oxygen concentrations in the overlying water were measured noninvasively in each core at 5 cm above the sediment surface with optical sensors (Presens, Germany). The cores were incubated at in situ temperatures of the corresponding lake bottom water at the sampling day (Table 2). During the incubation, oxygen concentration was measured every hour in one core from each lake to avoid the establishment of anoxic conditions. The water above the sediment was gently and continuously mixed with a magnet-based stirring device to avoid stratification of the water column. The cores were incubated in the dark for 1 week. Before stopping the incubation, oxygen and DIC were measured in every core.

Due to anoxia in Lake Solbergvann during both sampling occasions, anoxic respiration rates were assessed in Lake Solbergvann sediments. Anoxic incubations were performed similarly to the oxic incubations, but the overlying waters were carefully bubbled with nitrogen until the oxygen level in the cores was below the detection limit of $<0.015 \text{ mg L}^{-1}$. DIC and oxygen were measured as described for the oxic experiments. For methane measurements, 40 mL of water was taken in a 60 mL syringe and 10 mL of nitrogen was added. The syringe was shaken for 1 min, then the equilibrated headspace was transferred into a gas chromatography (GC) vial filled with a saturated salt solution and closed with a 12 mm thick butyl rubber stopper for storage until measurement. The cores were incubated for 1 week in the dark. Oxygen levels were continuously monitored throughout the experiment, and samples for DIC and methane were collected at the end of the experiment.

2.3. Laboratory Analyses

The water samples were brought to the laboratory immediately after their collection and stored dark and cold until further preparation or analysis. Water color was measured as absorbance at 410 nm of filtered samples ($<0.45 \mu\text{m}$) in a 5 cm cuvette and calibrated against a Pt standard the day after sampling. Total organic carbon (TOC) was analyzed with a Shimadzu TOC-V_{CPN} instrument. Sulfate concentration was measured in water samples by ion exchange chromatography (Lachat IC5000 instrument equipped with a Dionex-AC22 column). Total iron concentration in the water samples was measured by inductively coupled plasma mass spectrometry after acid decomposition (69 % (wt/wt) nitric acid 260°C) in a Milestone UltraClave III microwave oven. Accuracy in the analysis and traceability were ensured by a concurrent analysis of certified reference materials (inductively coupled plasma-mass spectrometry (ICP-MS)-Agilent 8800). DIC was analyzed with a Sievers 900 TOC analyzer. All DIC samples were kept cold and were analyzed within a few hours after sampling. Methane concentrations in the samples were measured using headspace equilibration and a gas chromatograph (Agilent Technologies 7890A) equipped with flame ionization detector.

Oxygen penetration depth was measured in eight sediment cores of Lake Elvåga with a microoptode (Presens, Germany) fixed to a micromanipulator. Oxygen measurements were taken in 50 or 100 μm steps in the overlying water and the sediment until the oxygen concentration was below the detection limit. In total, 20 profiles were recorded. In Lake Solbergvann, oxygen penetration depth could not be measured since the bottom waters were anoxic at both sampling occasions.

For measurement of total carbon, nitrogen, and sulfur concentrations in the sediment, the samples were freeze-dried and ground by hand in a mortar. The dry samples were stored in desiccators to avoid moisture. The wet and dry weight of the samples was measured in order to calculate the water content. Between 7 and 10 mg of samples were packed tightly in pressed tin capsules (Elemental Microanalysis, 6 \times 4 mm) and analyzed by high-temperature catalytic oxidation with a COTECH elemental combustion system 4010 elemental analyzer calibrated with a sulfanilamide standard (C 41.84%, N 16.27%, H 4.68%, O 18.58%, and S 18.62%). The analytical precision was $<\pm 0.3\%$ for C, $\pm 1.5\%$ for N, and $\pm 3.5\%$ for S.

For dating of the sediment cores, the radioisotopes ^{210}Pb and ^{137}Cs were measured in the ground dry samples by gamma spectroscopy at Eawag, Switzerland. The age and sedimentation rates of the sediment were determined from Pb-210 concentrations in the sediment samples using a constant rate of supply model [Appleby, 2001; Appleby and Oldfield, 1978].

For THg and MeHg measurements, the cores were stored cold and dark until they were delivered to the laboratory. They were then immediately sectioned into 0.5 cm intervals for the upper 10 cm and 1 cm intervals for the underlying 10 cm under an N_2 atmosphere in a glove bag. The samples were frozen immediately after slicing and then freeze-dried and ground using mortars. The THg concentration in the sediment samples was determined using thermal decomposition atomic absorption spectrometry (Perkin Elmer SMS 100). The analytical precision of the THg measurements, determined by replicate analysis, was $<\pm 6\%$. The accuracy of the THg analyses was checked by analyzing the lake sediment reference material WQB-3 (National Water Research Institute, Canada). The measured values were within the certified range for all analyses, with a $96 \pm 8\%$ ($n=4$) recovery of THg of the certified reference material. The extraction of inorganic Hg (IHg) and MeHg species was achieved with a focused microwave method (4 min, 80°C) from 200 mg of sediment in 5 mL of 6 N HCl [Rodríguez Martin-Doimeadios *et al.*, 2003]. IHg and MeHg in sediment extracts and unfiltered water samples were measured by species-specific isotope dilution and capillary gas chromatography-inductively coupled plasma–mass spectrometry (GC-ICP-MS, Agilent 6890–7500a) to correct for potential Hg species' losses and conversion reactions [Monperrus *et al.*, 2008; Rodríguez Martin-Doimeadios *et al.*, 2003]. Recovery of the speciation analysis in sediment samples was $99 \pm 6\%$ ($n=54$) (i.e., inorganic divalent Hg (IHg) + MeHg measured by GC-ICPMS versus THg measured by SMS 100).

2.4. Calculations and Statistics

Dry sediment mass accumulation rates ($\text{g m}^{-2} \text{yr}^{-1}$) were calculated from the chronologies and used for calculation of C, Hg, and S mass accumulation rates. Relationships between the downcore trends of C, Hg, and S were tested by generalized least squares linear models, accounting for an autocorrelation with the generalized least squares function in R. We tested the autocorrelation and included for both lakes a first-order autoregression model AR(1), accounting only for the previous layer. Statistical significance was set at a $p < 0.05$. This procedure could not be used to analyze the most recent trends in the sediment mass accumulation rates due to the low number of dated sediment strata during the period of lake browning, particularly in Lake Solbergvann.

Organic carbon burial efficiency was calculated as the ratio between OC burial and OC deposition onto the sediment, the latter calculated as the sum of organic carbon mineralization and organic carbon burial. Carbon burial of sediment younger than 10 years was corrected for the expected mass loss due to continuing sediment degradation after deposition according to Galman *et al.* [2008]. For Lake Elvåga, oxic conditions over the whole year were assumed, while for Lake Solbergvann, anoxic conditions over the whole year were assumed. These assumptions are based on observations of stratification and oxygenation properties of the two lakes in late summer and spring.

The profiles of methane concentration in the sediment pore water were used in order to identify methanogenic conditions and suboxic/anoxic layers in the sediment.

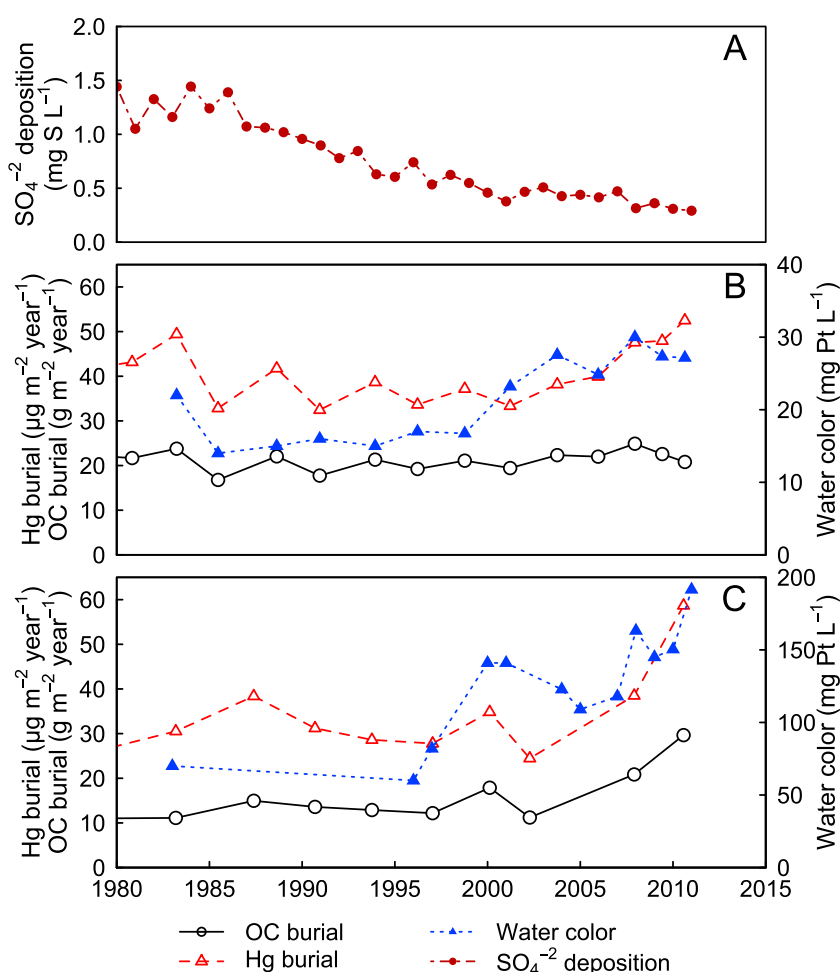


Figure 1. Water color, sediment OC, and THg burial rates in (b) Elvåga and (c) Solbergvann for the period of browning and decreasing (a) SO_4^{2-} deposition in the forest area of Oslo, Norway.

3. Results

3.1. Lake Properties

Pronounced increases in water color have been observed in both lakes over the past 15 years, particularly in Lake Solbergvann. During the period from year 1996 to 2011, water color in Lake Solbergvann has increased more than 3 times, from 60 to 192 mg Pt L^{-1} ($130 \pm 35 \text{ mg Pt L}^{-1}$, $n = 12$), whereas in the northern basin of Lake Elvåga, the increase was less than twofold from 17 to 27 mg Pt L^{-1} ($23 \pm 7 \text{ mg Pt L}^{-1}$, $n = 57$) (Figure 1). It is likely that the magnitude of water color change was larger in the southern basin of Lake Elvåga as compared to the northern basin, as the southern basin receives drainage from 64% of the entire lake catchment and therefore receives the majority of terrestrial DOC input to the lake. Accordingly, at the time of sampling, the southern basin exhibited a higher water color than the northern basin (64 and 33 mg Pt L^{-1} , respectively). Similar to the difference in water color between the lakes, we found that TOC concentration in the surface water of Lake Solbergvann (15.4 mg L^{-1}) was higher than in Lake Elvåga (8.3 mg L^{-1}).

The stratification properties were very different between the two lakes. Lake Solbergvann was strongly stratified at both sampling occasions (Figure S1 in the supporting information) as indicated by the temperature profile, anoxic bottom water, and strongly elevated conductivity in the bottom water. This indicates the absence of complete mixing during spring, and thus that anoxic conditions predominate in the bottom water of Lake Solbergvann. In contrast, Lake Elvåga was thermally stratified, but the bottom water oxygen was at 20–30% saturation at the two sampling occasions, indicating regular holomixis.

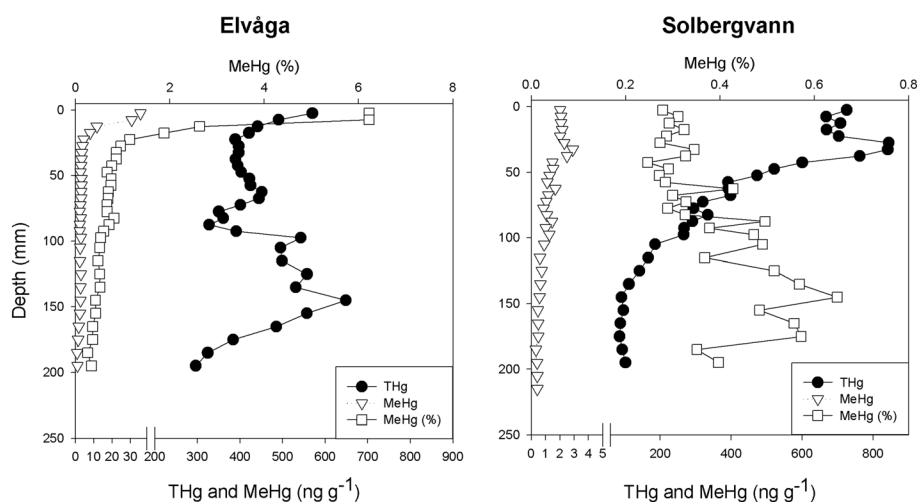


Figure 2. Sediment profiles of total mercury (THg) and methylmercury (MeHg). Note the difference in scales.

THg concentrations in the surface water were slightly higher in Lake Elvåga ($6.4 \pm 0.5 \text{ ng L}^{-1}$) than in Lake Solbergvann ($5.1 \pm 0.9 \text{ ng L}^{-1}$), whereas similar surface water MeHg concentrations were measured in both lakes (0.32 ± 0.005 and $0.34 \pm 0.015 \text{ ng L}^{-1}$, respectively). In Lake Solbergvann only, we observed a distinct peak of MeHg ($1.1 \pm 0.013 \text{ ng L}^{-1}$) in anoxic bottom water at 5 m depth, corresponding to 26% MeHg of THg. This MeHg peak in Lake Solbergvann coincided with an increase of total iron concentration and a decrease of the sulfate concentration toward the sediment (Figure S2).

3.2. Sediment Properties

In general, both lakes had soft, organic-rich sediments. Mean OC concentration (\pm standard deviation) was very high in Lake Solbergvann ($361 \pm 27 \text{ g kg}^{-1}$) and varied only slightly over depth (Figure S3). The OC concentration in Lake Elvåga decreased downcore from 226 g kg^{-1} at the surface to 197 g kg^{-1} at 400 mm depth, with a mean concentration of $213 \pm 9 \text{ g kg}^{-1}$. Nitrogen concentrations followed similar patterns over depth as the OC concentration in Lake Solbergvann, indicating that nitrogen to a large extent is organically bound (Figure S3). In Lake Elvåga, N concentrations were invariant over depth. The C/N ratio in Lake Solbergvann increased from about 12 to almost 14 with depth but was around 10 in the topmost layers (Figure S4). In Lake Elvåga, the C/N ratio varied only slightly between 13 at the surface and 12 at the bottom of the core. Sulfur concentrations varied between 6 and 52 g kg^{-1} in Lake Solbergvann, with a peak at 42.5 mm depth, while in Lake Elvåga, sulfur concentrations were lower and less variable over depth (range, $1\text{--}7 \text{ g kg}^{-1}$) (Figure S3).

THg displayed different patterns in the two lakes (Figure 2). In Lake Solbergvann, THg concentrations increased from 96 ng g^{-1} at the bottom of the core (\sim A.D. 1850) to 844 ng g^{-1} at 27.5 mm depth (\sim A.D. 1990) and decreased again toward the surface (725 ng g^{-1}). Previous sediment studies from Lake Solbergvann have shown similar concentration gradients for THg ($90\text{--}1000 \text{ } \mu\text{g L}^{-1}$) [Sæther, 2009]. In Lake Elvåga, in contrast, the THg concentration showed less variability over depth, averaging at $440 \pm 83 \text{ ng g}^{-1}$, with higher values at the sediment surface (570 ng g^{-1}) and between 10 and 15 cm depth ($500\text{--}600 \text{ ng g}^{-1}$), which correspond to the end of the 1960s when the dam was built. In the Lake Solbergvann sediment, despite generally high THg levels in the upper 5 cm of the sediment, the percentage of THg present as MeHg, often considered as a proxy of the net MeHg formation rate [Drott *et al.*, 2008], was low (0.2–0.6%) and relatively constant over depth (Figure 2). The Lake Elvåga sediment, however, displayed a strong peak in MeHg at the sediment surface, reaching values of 6% MeHg, over 1 order of magnitude higher than in Lake Solbergvann (Figure 2). In Lake Elvåga, the peak in MeHg coincides with high THg concentrations in the most recently deposited sediment layers. Over the whole core, the MeHg concentration was not coupled to THg in Lake Elvåga, whereas in Lake Solbergvann THg and MeHg concentrations were significantly correlated ($p < 0.01$).

3.3. Sediment Burial

The sediment dry mass accumulation rate in Lake Solbergvann was on average $39 \pm 14 \text{ g m}^{-2} \text{ yr}^{-1}$ but increased toward the sediment surface to $84 \text{ g m}^{-2} \text{ yr}^{-1}$ (Figure S5). The sediment dry mass accumulation

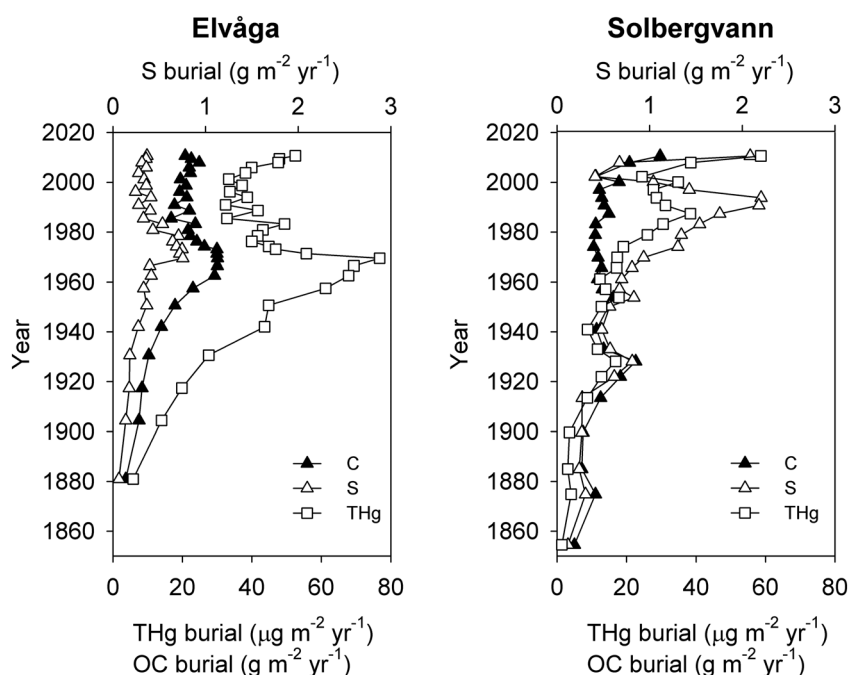


Figure 3. Burial rates of OC, S, and THg over time for the two study lakes.

rate in Lake Elvåga was higher, averaging at $95 \pm 32 \text{ g m}^{-2} \text{ yr}^{-1}$ with the highest values ($143 \text{ g m}^{-2} \text{ yr}^{-1}$) following the construction of the dam in A.D. 1964. In Lake Solbergvann, average OC burial was $13.4 \pm 4.5 \text{ g m}^{-2} \text{ yr}^{-1}$, peaking at $24.8 \text{ g m}^{-2} \text{ yr}^{-1}$ in the top sediment layer as the result of a pronounced increase in OC burial during the past 20 years (Figure 3). S burial in Lake Solbergvann followed the OC burial until about A.D. 1960. Between \sim A.D. 1960 and \sim A.D. 1990, the S burial increased from 0.7 to $2.2 \text{ g m}^{-2} \text{ yr}^{-1}$, whereas the OC burial did not vary. The S burial increase was interrupted by an abrupt decline at \sim A.D. 1990 (Figure 3). However, since \sim A.D. 2000, S and OC burial concomitantly increased again. In Lake Elvåga, the higher sediment dry mass accumulation rate as compared to Lake Solbergvann resulted in high OC burial rates, in spite of a comparatively lower OC concentration. The OC burial in Lake Elvåga was on average $20.8 \pm 6.8 \text{ g m}^{-2} \text{ yr}^{-1}$ with a peak of $30.1 \text{ g m}^{-2} \text{ yr}^{-1}$ \sim A.D. 1969 ± 1 , after the dam was built. There was no distinct increase in OC burial since the onset of lake browning at \sim A.D. 2000 in Lake Elvåga (Figure 1). The S burial in Lake Elvåga followed the trends of the OC burial and was on average $0.4 \pm 0.2 \text{ g m}^{-2} \text{ yr}^{-1}$ with a peak of $0.8 \text{ g m}^{-2} \text{ yr}^{-1}$ \sim A.D. 1969 ± 1 .

THg burial displayed a strong variability over time in both lakes (Figure 3), starting at similarly low background levels in the late 1800s and increasing by about 1 order of magnitude from the preindustrial period to recent times. In Lake Solbergvann, the THg burial increased from the bottom of the core ($1.4 \mu\text{g Hg m}^{-2} \text{ yr}^{-1}$) toward the surface ($59 \mu\text{g Hg m}^{-2} \text{ yr}^{-1}$), where it reached the maximum (Figure 3). The THg burial followed similar patterns as the OC and S burial until \sim A.D. 1960, when the THg and S burial started to increase until \sim A.D. 1990, while the OC burial remained low. Since the onset of lake browning of Lake Solbergvann however, a concomitant increase in the burial of both Hg and OC can be observed, while S burial followed a similar trend, but less tightly linked to OC and Hg burial due to a comparatively low S burial around A.D. 2000 (Figure 1). In Lake Elvåga, the lowest THg burial was found in the bottom of the core ($5.8 \mu\text{g Hg m}^{-2} \text{ yr}^{-1}$). The THg burial increased upcore and reached its maximum after the dam construction ($77 \mu\text{g Hg m}^{-2} \text{ yr}^{-1}$) and then slightly decreased upcore. The THg burial historically followed the trends of the OC and S burial, including the peak after the construction of the dam. Since \sim A.D. 1990, however, the THg burial has increased from ~ 30 to $\sim 50 \mu\text{g Hg m}^{-2} \text{ yr}^{-1}$, without a concomitant increase in the OC and S burial (Figure 3).

3.4. Sediment Mineralization and OC Burial Efficiency

The differences in oxygenation of the bottom waters of the lakes strongly influenced OC mineralization rates. When incubated with oxygenated overlying water, the OC mineralization rates of the Lake Solbergvann sediments

were 2 times higher than when incubated with anoxic overlying water (17.5 ± 4.1 and 7.7 ± 0.9 g C m⁻² yr⁻¹, respectively). In Lake Elvåga, the bottom water was oxygenated at both sampling occasions. The OC mineralization rates of the Lake Elvåga sediments incubated with oxygenated water (15.5 ± 4.1 g C m⁻² yr⁻¹) were very similar to the oxygenated sediment OC mineralization rates in Lake Solbergvann.

To calculate recent OC burial efficiency, representative for the period of lake browning, we used the anoxic mineralization rate of Lake Solbergvann and the oxic mineralization rate of Lake Elvåga, given that Lake Solbergvann was found to have anoxic bottom water and Lake Elvåga to have oxygenated bottom water at both sampling occasions. Also, for calculation of the recent OC burial efficiency we used OC burial during the past 5 years only, in order to reflect the recent peaks in both water color and OC burial. OC in young sediment layers is still degrading, and degradation is exponentially related to sediment age [Middelburg *et al.*, 1993]. To account for incomplete OC mineralization in these relatively young sediment layers, we subtracted the expected C mass loss during continued mineralization based on the observed C mass loss of a Swedish lake sediment [Galman *et al.*, 2008]. Accordingly, OC burial during the past 5 years was calculated to 22.6 ± 3.1 g C m⁻² yr⁻¹ in Lake Solbergvann and 21.2 ± 2.5 g C m⁻² yr⁻¹ in Lake Elvåga, respectively. These calculations return an OC burial efficiency of $72 \pm 12\%$ in Lake Solbergvann and $57 \pm 9\%$ in Lake Elvåga. It should be noted that these values are not representative for the entire lake basins but only for the coring sites at the deepest spots.

4. Discussion

4.1. Effects of Lake Browning on OC Burial

The results of this study are consistent with other studies that suggest that an increase in lake water color in boreal lakes, which has been ascribed to an increase of organic matter input from land [Hongve *et al.*, 2004], can result in an increased delivery of organic matter to the sediment [von Wachenfeldt *et al.*, 2008]. This study adds to existing knowledge by showing that in some boreal lakes, increasing water color may also be related to increased OC burial in the sediment. In Lake Solbergvann, we observed that OC burial rates peaked at the most recently deposited sediment, which corresponds to the period with the highest water color (Figures 1 and 3), indicating that the increased delivery of OM to the sediment due to lake browning is relatively well conserved in the sediment record. While both lakes experienced water color increase over the past ~15 years, the change in water color was about threefold in Lake Solbergvann but only twofold in Lake Elvåga. The smaller change in water color may partly explain the absence of a measureable response of sediment OC burial to lake browning in Lake Elvåga (Figure 1). These results suggest that the increasing water color and DOC concentration observed in many boreal lakes [Monteith *et al.*, 2007] can, in some lakes, result in increased OC burial in the sediments. This interpretation is supported by an observed increase in OC sedimentation from the water column of a boreal lake that has experienced browning [von Wachenfeldt *et al.*, 2008]. Similarly, a recent palaeolimnological study of two boreal lakes [Bragee *et al.*, 2013] demonstrated that trends in past lake water TOC concentrations, a proxy of water color, and terrestrial OM input were related to the OC concentration of the sediment, particularly in a small, highly humic lake. Apparently, water color and terrestrial DOC export is linked to the carbon budget of lake sediments. This conclusion is drawn from recent OC burial rates that were corrected for future OC mass loss during ongoing mineralization [Galman and Rydberg, 2008]; hence, the increase of OC burial in surficial layers cannot be explained by incomplete mineralization.

4.2. Influence of the Dominating Mode of Respiration on OC Burial

Our data demonstrate that OC burial is affected by the dominating mode of respiration, i.e., aerobic or anaerobic. In Lake Solbergvann, a lake with prolonged periods of bottom water anoxia, sediment OC is mainly degraded via anaerobic respiration. We found that anaerobic respiration reduced overall OC mineralization by about half compared to aerobic respiration (7.7 ± 0.9 g C m⁻² yr⁻¹ and 17.5 ± 4.1 g C m⁻² yr⁻¹, respectively), resulting in a high OC burial efficiency ($72 \pm 12\%$) in Lake Solbergvann. In Lake Elvåga, in contrast, the bottom water remained oxygenated over the year, resulting in twice as high sediment OC mineralization rates as compared to the anoxic Lake Solbergvann. Consequently, the OC burial efficiency was lower in Lake Elvåga ($57 \pm 9\%$), and hence, any potential increase in OC delivery to the sediment is not as well conserved in the sediment record of Lake Elvåga as compared to Lake Solbergvann (Figure 1). We suggest that this is the second reason, apart from the greater increase in water color, why OC burial increase in response to lake browning could be observed in Lake Solbergvann, but not in Lake Elvåga. While two sampling campaigns

cannot account for all seasonal and interannual variability in stratification and bottom water oxygenation, the clear differences between the two lakes at both occasions (Figure S1) likely illustrate the typical situations predominating in those lakes.

Stratification properties are closely linked to the degree of bottom water oxygenation and thus to the resulting mode of respiration, which in turn affects the OC burial in the sediment. This effect is probably strongest in lake sediments that primarily receive OC inputs from terrestrial sources, since the degradation of OC derived from land plants has been shown to be much more inhibited by the absence of oxygen as compared to the degradation of OC derived from aquatic primary producers [Sobek *et al.*, 2009; Zehnder and Svensson, 1986]. The lakes studied here receive sizeable terrestrial OM inputs, as indicated by C/N ratios, generally >12 in both lakes (Figure S4) [Meyers and Ishiwatari, 1993], explaining the observed strong effect of oxygen on OC mineralization. Most boreal lakes receive high inputs of terrestrial OC [Sobek *et al.*, 2007], and the resulting dark water color inhibits lake-internal primary production [Algesten *et al.*, 2004; Karlsson *et al.*, 2009], such that terrestrial OC has been shown to be the major source of OC to boreal lake sediments [Chmiel *et al.*, 2015; von Wachenfeldt and Tranvik, 2008], even if temporal variability in source contribution can be expected (e.g., Figure S4). In addition, boreal lakes generally stratify strongly due to highly colored water and wind protection by surrounding forests [Fee *et al.*, 1996]. Lake browning therefore not only implies a higher loading of terrestrial OC to lakes and an increased sedimentation of terrestrial OC in lakes [von Wachenfeldt *et al.*, 2008] but also stronger and longer stratification periods, which further promote OC burial due to decreased oxygenation of bottom waters [Jankowski *et al.*, 2006]. On the other hand, as the concentration of terrestrial DOC has been shown to be positively related to CO₂ emission from boreal lakes [Sobek *et al.*, 2003], lake browning may also increase aquatic CO₂ emissions to the atmosphere. However, we can only speculate to which degree the increasingly mobilized DOC is either emitted or buried. Estimates of the ratio of lake emission to burial, ranging in scales from individual lakes to the catchment and global scale, point toward a dominance of emission over burial [Algesten *et al.*, 2004; Kortelainen *et al.*, 2013; Tranvik *et al.*, 2009].

4.3. Effects of Lake Browning on Hg and MeHg in Sediments

In both lakes, the extent of the recent increase in Hg burial rates equaled the increase in water color, i.e., 1.6 and 3 times in Lake Elvåga and Lake Solbergvann, respectively (Figure 1), even if the statistical significance of this trend cannot be ascertained due to the small number of sediment layers exposing recently increased Hg burial. Hg and OC burial were significantly correlated in both lakes since A.D. 1990 ($p < 0.001$). A relationship between water color and Hg concentration has been previously reported [Dittman *et al.*, 2009; Eklöf *et al.*, 2012; Nilsson and Hakanson, 1992] and recently a relationship between water color and Hg concentrations in sediment and fish [Hongve *et al.*, 2012]. Here we demonstrate that increasing water color was paralleled by increased Hg burial rates in the sediment of two boreal lakes that experienced different degrees of browning. In lakes that are similar in terms of limnological characteristics and geographical location as the lakes in this study, lake browning could therefore potentially offset the declining trends of Hg concentrations in sediments that have been lately reported consecutive to the reduction of Hg emissions [Engstrom *et al.*, 2007]. It should be noted that the reports of declining sediment Hg concentrations [Engstrom *et al.*, 2007] only cover the time period before the year 2000, which was prior to the major increase in THg and OC burial observed in our study.

Our results are consistent with the concept that the terrestrial OM, mobilized from the catchment by decreased ionic strength of soils as a result of reduced acid precipitation [Haaland *et al.*, 2010; Monteith *et al.*, 2007], may serve as a vector of both OC and Hg from soils to surface waters [Hongve *et al.*, 2012] and may leave an imprint in the sediment record [Teisserenc *et al.*, 2011] (Figure 3). If so, these results may carry further environmental implications because despite the decrease of Hg atmospheric emission, the mobilization of Hg by increased mobilization of terrestrial OM due to lake browning might lead to increased Hg concentrations in boreal lakes of similar characteristics and geographical location.

The remobilized Hg can eventually be methylated. The percentage of MeHg in sediments, used as a proxy of the net Hg methylation [Drott *et al.*, 2008], was higher in Lake Elvåga (up to 6%) than in Lake Solbergvann ($\leq 0.6\%$) (Figure 2). Sulfate- and iron-reducing bacteria are considered the major contributors to MeHg formation in anaerobic sediments even if methanogens might also contribute [Gilmour *et al.*, 2013]. Therefore, the greatest methylation activity is often found just below the oxic/anoxic transition zone in sediments underlying oxygenated water [Bravo *et al.*, 2014; Taylor *et al.*, 2001; Watras *et al.*, 1995], where both sulfate and iron

reduction rates can be high. Conversely, low percentages of MeHg are commonly observed in completely anoxic, methanogenic environments where sulfate and iron reduction is limited. In Lake Elvåga, the oxic/anoxic boundary was located in the surficial sediment (mean oxygen penetration depth into the sediment, 2.8 ± 0.9 mm; Table S2), implying that methylation takes place in surficial sediments, where IHg availability to methylating organisms was high. In Lake Solbergvann, water column profiles of oxygen and sulfate concentrations indicate the occurrence of sulfate reduction in the water column (Figure S2). Furthermore, methane concentrations in pore waters were higher in Lake Solbergvann than in Lake Elvåga (Figure S6), which together with the absence of oxygen indicates that sulfate and iron reduction was low in Lake Solbergvann sediments. The low percentage of MeHg found in the surface sediments and the presence of a peak of MeHg in the anoxic water column (at 5 m depth) suggest that methylation likely occurs in the anoxic layer of the water column of Lake Solbergvann. The methylation is likely limited under methanogenic conditions that appear dominant in Lake Solbergvann sediments. Hence, our study suggests that lake browning will likely increase Hg concentrations in lake sediments but that the vertical position of the oxycline, and as a consequence the dominant mode of respiration in the sediment, will regulate the extent of MeHg formation in the sediments, where the major IHg pool resides. Therefore, relationships between water color and Hg levels in fish of any given lake [Hongve *et al.*, 2012] are affected by factors that affect net methylation rates in sediments and bottom waters, such as stratification and oxygenation properties, making generalized conclusions about the influence of water color on Hg in fish difficult to draw.

4.4. Temporal Trends of OC, Hg, and S Burial Prior To Lake Browning

The pattern of the sediment OC and Hg burial varied over time between the two lakes. In Lake Elvåga, the rates of Hg burial were significantly correlated to the rates of OC burial throughout the entire sediment profile ($p < 0.001$; $n = 29$) (Figure 3). Despite the fact that the OC and S burial were correlated ($p = 0.0027$; $n = 29$), no significant correlation was observed between the Hg and S burial ($p = 0.58$; $n = 29$). The increase of Hg was not followed by an increase of S, indicating that S and Hg sources to Lake Elvåga are not the same. This is probably related to the high catchment to lake area ratio for Lake Elvåga, implying that a higher share of Hg atmospheric deposition occurs on catchment soils as compared to the deposition directly on the lake. The sediments of Lake Elvåga therefore mainly record the secondary loading of organically bound Hg and C from the catchment.

In Lake Solbergvann, in contrast, a different pattern was found in OC, Hg, and S burial. The Hg burial was significantly correlated to the OC burial ($p = 0.001$, $n = 13$) and S burial ($p < 0.001$, $n = 13$) from A.D. 1860 to about A.D. 1960 (Figure 3). During A.D. 1960–1990, however, we observed a correlation between the Hg burial and the OC burial ($p = 0.047$, $n = 8$), but neither the OC ($p = 0.89$, $n = 8$) nor the Hg ($p = 0.79$, $n = 8$) burial correlated with the S burial, indicating a high atmospheric deposition of S during this period. Lake Solbergvann has a 6 times lower catchment to lake area ratio compared to Lake Elvåga; hence, its sediments will, to a much larger degree, reflect direct atmospheric deposition onto the lake surface. Furthermore, this decoupling of S and OC burial fits well with the inverse relationship between acid deposition and DOC, which is regarded as an important driver for the recent increase in DOC [Monteith *et al.*, 2007]. Since ~A.D. 1990, the Hg burial was largely unaffected by the strong dip in S burial around ~A.D. 2000 (Figure 3). Both atmospheric S and Hg deposition have decreased lately (Figure 1) [Johansson *et al.*, 2001], and the concomitant decrease in S burial but unaffected Hg burial could imply that the catchment was the main source of OC and Hg to the lake since A.D. 1990. The observed increases in water color have likely resulted in stronger stratification and more pronounced bottom water anoxia [Fee *et al.*, 1996; Jankowski *et al.*, 2006] in the water column (Figure S2). As anoxia also reduces OC mineralization rates, OC-Hg complexes are relatively stable in sediments, explaining the tight coupling of OC and Hg burial since the past 20 years. Patterns of OC, S, and Hg burial in these lake sediments therefore result from a combination of inputs from atmospheric deposition and catchment exports but also from stratification properties, which modulate the records through the mode of respiration.

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Erratum

In the originally published version of this article, the article title was “Lake browning and respiration mode affect the burial and fate of carbon and mercury in the sediment of two boreal lakes”. The title has since been corrected and this version may be considered the authoritative version of record.