Factors controlling temporal variation in methyl mercury levels in sediment and water in a seasonally stratified lake

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Abstract
Total mercury (total Hg) levels were low in the water (2.5-7.5 pM) and in the sediment (150-300 pmol g (DW)^-1), indicating an absence of significant local sources of Hg. During summer stratification, methyl mercury (MeHg) levels increased below the thermocline, reaching 2.5 pM in the anoxic hypolimnion, whereas in the epilimnion levels remained low (0.25-0.6 pM) throughout the study period (late April-late October 1993). On October 27 (the last sampling date), the lake was totally mixed and MeHg in the eutrophic water column had returned to low levels. Within the part of the basin turning anoxic during summer stratification, the MeHg levels in the surficial sediment (0-0.5 cm) and those in the overlying water were negatively correlated (r = -0.87, P < 0.05). Sediment MeHg decreased during summer stratification until August and then increased. In the sediment, total organic carbon was significantly correlated with total Hg (r = 0.86, P < 0.05), MeHg (r = 0.90, P < 0.01), and total Mn (r = 0.88, P < 0.01). A mechanism for the partitioning of MeHg between water and sediment is discussed, involving the Mn redox cycle. Budget calculations indicated that exchanges of a fixed amount of MeHg between the water and sediment could not explain the observed seasonal variation in MeHg levels. Other processes that may have contributed to the variation in MeHg levels in the sediment and water are methylation, demethylation, mass dilution, and uptake of MeHg in the biota.

Methyl mercury (MeHg), owing to its remarkable ability to pass biological membranes, its high chemical stability, and its slow excretion from most organisms (Lofroth 1968), is biomagnified in aquatic food chains (Cahana and Rasmussen 1994). Its toxicity makes it a potential threat to regular consumers of fish. Furthermore, epidemiological studies and experiments with various mammals suggest that the sensitivity of the human fetus to MeHg may be much higher than that of human adults (Clarkson 1990). Even in seemingly pristine lakes, the fish surprisingly often contain high enough levels of MeHg to call for fish consumption advisories (Driscoll et al. 1994). This is explained by long-range atmospheric transport and deposition of both methylated and inorganic Hg species (Lee and Hultberg 1991; Fitzgerald et al. 1991). Some of the deposited inorganic Hg(II) will be converted to MeHg in lake water, lake sediments, soils, and wetlands, which probably is the main reason for the buildup of MeHg in most lakes (St. Louis et al. 1994; Watras et al. 1995).

If the aim is to reduce MeHg levels in freshwater fish, it is of vital importance to know the relative contributions from different sources to the MeHg burden of lakes. An important question is whether most of the MeHg in a lake is produced within the lake itself or originates from external sources. It is relatively easy to quantify the MeHg that is entering a lake through direct precipitation and runoff water. The difficult part, however, is to determine the fate of MeHg within a lake. Some of it could be demethylated in the water or assimilated by the sediment, from which it may or may not be released back to the water. As long as these processes are poorly understood, it is not possible to draw any firm conclusions concerning the relative importance of internal and external sources in the supply of MeHg to lakes.

Studies of the partitioning of Hg between sediment and water in microcosms, carried out in our laboratory, have indicated MeHg release from sediments to be a redox-controlled process (Regnell and Tunlid 1991; Regnell 1994; Regnell et al. 1996). Here we report results from a lake study that support this finding. We think that this study is the first to demonstrate an inverse relationship between MeHg concentrations in surficial sediment and those in the overlying water, suggesting internal cycling of MeHg between water and sediment.

Methods

Study site—The study was performed in Lake Levrasjön, which is located in southern Sweden. Its phosphorus and nitrogen levels indicate mesotrophic to eutrophic conditions (total P of ~40 µg liter^-1, total N of ~700 µg liter^-1). Total Hg concentrations in the sediment are typically below the local background concentration for lake sediments (~500 pmol Hg g(DW)^-1). The lake has an area of 285 hectares, a drainage area of 820 hectares (mostly farmland and meadows), and a maximum depth of 18 m. The theoretical residence time has been estimated to be 11.5 yr based on specific runoff data. The amount of water at depths greater than 10 m constitutes ~25% of the entire lake volume.

Sampling strategy—Water and sediment samples were collected at approximately monthly intervals from late April to late October. At each sampling occasion temperature and dissolved oxygen profiles were established. At the July and August samplings, water was taken from five depths corresponding to 1 m below the surface, the uppermost part of
Table 1. Data on analytical quality of the total Hg and MeHg measurements in the present study.

<table>
<thead>
<tr>
<th>Sample (unit)</th>
<th>Detection limit*</th>
<th>Accuracy†</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>Total Hg (pg Hg ml⁻¹)</td>
<td>MeHg (pg Hg ml⁻¹)</td>
</tr>
<tr>
<td>Water (pM Hg)</td>
<td>0.25</td>
<td>0.1</td>
</tr>
<tr>
<td>Sediment (pmol Hg g (WW)⁻¹)</td>
<td>0.05</td>
<td>0.005</td>
</tr>
</tbody>
</table>

<table>
<thead>
<tr>
<th>CRMs/SRMs (certified/standard ref. material)</th>
<th>Total Hg (pg Hg g (DW)⁻¹)</th>
<th>MeHg (pg Hg g (DW)⁻¹)</th>
</tr>
</thead>
<tbody>
<tr>
<td>CRM (NRCC)-DORM-1, dogfish muscle (μg Hg kg⁻¹)</td>
<td>731±60</td>
<td>1.52±0.04</td>
</tr>
<tr>
<td>SRM (NIST)-1641b, water (μg Hg ml⁻¹)</td>
<td>1.52±0.04</td>
<td>1.51±0.06 (n = 10)</td>
</tr>
<tr>
<td>CRM-280, lake sediment (μg Hg g (DW)⁻¹)</td>
<td>0.670±0.019</td>
<td>0.655 (n = 1)</td>
</tr>
</tbody>
</table>

*a Valid for water samples of 30 and 50 ml (for MeHg and total Hg determinations, respectively) and for 1 g of wet sediment.
† Mean difference between duplicate samples taken in this study ±1 SD.

The thermocline, the middle of the thermocline, the lowest part of the thermocline, and 1 m above the sediment. In the other months, water was only sampled from 1 m below the surface and 1 m above the sediment. Five sampling stations 75 m apart were marked out with buoys in the central part of the lake where the depth was 16 ± 0.5 m. Water samples were taken from only one of these stations, whereas sediment samples were taken from all five stations.

**Sampling methodology**—Water samples for Hg measurements were obtained by a submersed Teflon tubing connected to a glass bottle. Water was pumped into the bottle by a hand-driven pump connected to the bottle through a gold trap, thus minimizing the risk of contamination. Teflon bottles (vol of ~120 ml), filled with ultraclean MQ water and 0.5 ml concentrated HCl were emptied and rinsed twice with the sample water from the glass bottle before being completely filled up with sample. Each Teflon bottle was then double bagged using plastic polyethylene bags and placed in a refrigerated box. In July and August additional Teflon bottles were filled with water that was filtered (0.45 μm) in the laboratory within 48 h of sampling.

Sediment was obtained with a coner of our own design equipped with plastic liners. Only the upper 0.5 cm of the sediment cores was transferred to Teflon bottles for mercury analyses and to glass jars for other analyses. The Teflon bottles were of the same kind as those used for water samples except that they were filled with unacidified MQ water during storage before sampling. When filled with sediment, they were double bagged and immediately frozen in a box with dry ice.

All equipment used to obtain and store samples for mercury analyses was acid washed (ultraclean HCl) and always handled with care. Long plastic gloves were used to avoid contamination.

**Analytical procedures**—Total Hg was determined using a double amalgamation technique and atomic fluorescence detection (AFS).

For total Hg determinations, water samples were oxidized by adding bromine chloride (BrCl) before reducing the mercury to elemental mercury by stannous chloride (SnCl₂). The elemental Hg was then bubbled off with nitrogen and collected on a gold trap. To avoid interference during measurement, the Hg was thermally desorbed from the first gold trap and collected on a second gold trap from which it was subsequently desorbed and led into the measuring cell. See Table 1 for detection limits and certified and/or standard reference materials (CRMs/SRMs).

Sediment samples were wet oxidized by adding a mixture of sulfuric acid and nitric acid. The sample was then subjected to refluxing by heating on a hot plate for ~6 h. After dilution with MQ water, the resulting solution was analyzed as described for water samples.

MeHg was quantified according to the method described by Bloom (1989). In short, MeHg in an aqueous phase was ethylated by adding sodium tetraethyl borate, resulting in volatile methylthiol Hg that was bubbled off and collected on a carbotrap containing graphitized carbon. The methylthiol Hg was then thermally desorbed from the carbotrap and collected on a gas chromatography (GC) column held in liquid nitrogen. Elution from the GC column was attained by raising the temperature to 180°C. A thermal decomposer converted the methylthiol Hg to elemental Hg vapor, which was quantified in an AFS-measuring cell. Calibration curves were established using standard solutions of CH₃HgCl(s).

To remove substances that might interfere with the ethylation process, water samples were amended with a KCl-HCl solution converting all MeHg species to MeHgCl, which was extracted with methylene chloride. After the water had been discarded, the MeHgCl was back-extracted to water by adding MQ water to the methylene chloride, which was subsequently boiled off (Bloom 1989). The resulting aqueous solution was then subjected to the ethylation procedures described above.

By using the method of Horvat et al. (1988), ~1–2 g of wet sediment was mixed with MQ water, sulfuric acid, and KCl in a Teflon tube. This Teflon tube was connected to another identical Teflon tube placed in cold water. The sample was distilled over to the cooled Teflon tube by placing
the first Teflon tube in a heating block held at a temperature of \( \sim 145^\circ C \). The distillate was then ready for the ethylation procedures and was analyzed as described for water samples.

Total Mn, Fe, and Ca were determined in water samples that were acidified with HNO\(_3\), using an ICP-ES instrument (Perkin Elmer, plasma II emission spectrometer). In order to determine these metals in sediment samples, lyophilized sediment (\( \sim 0.5 \) g) was placed in a Teflon container to which HNO\(_3\) and MQ water was added (2.5 + 5 ml). Wet oxidation was performed in a microwave oven (CEM). The wet oxidized samples were diluted to 25 ml and analyzed for Mn, Fe, and Ca as described for water samples.

The organic C content of the water was determined using a carbon analyzer (Shimadzu, TOC-2000). The total C (inorganic + organic) of the sediment was determined in \( \sim 200 \) mg of lyophilized sediment using a carbon analyzer for solid samples (LECO). Organic C (TOC) was determined after acidifying the sample with 5 ml of 5 M HCl and then drying it. Inorganic C (TIC) was calculated by subtraction.

**MeHg budget calculations**  We assumed a density of 1 for wet sediment originating from the upper 0.5 cm sediment layer because the water content was stable at 96.2 \pm 1\% (n for wet sediment originating from the upper 0.5 cm sediment layer). Inorganic C (TIC) was calculated by subtraction.

Amount of MeHg in layer (mmol)

\[
= \left[ (A_0 - Z(A_0 - A_1))Z \right] \frac{Z(C_0 - C_1)}{Z} dz
\]

where \( Z \) is the depth of layer (m), \( A_0 \) is the upper cross-section area of layer (km\(^2\)), \( A_1 \) is the lower cross-section area of layer (km\(^2\)), \( C_0 \) is the concentration of MeHg at the upper cross section (pm), and \( C_1 \) is the concentration of MeHg at the lower cross section (pm).

Similar budget calculations were performed to estimate the amount of Mn and Fe in the sediment and water.

**Incubation experiment**—Starting on 16 September, we incubated water samples with radiolabeled HgCl\(_2\) (specific activity, 2.5 \( \muCi/\mug \) Hg). We collected water from five depths in the water column representing the epilimnion (1 m), the upper thermocline (12 m), the middle of the thermocline (13 m), the lower thermocline (14 m), and the hypolimnion (15 m). Winkler bottles (120 ml) were completely filled with water to avoid air contamination. The isotope was injected at the bottom, the addition of Hg corresponding to 16 \( \mug \) Hg liter\(^{-1}\). The bottles (nine per level) were closed with a ground glass stopper, excluding all air and suspended at the same depth in the water column as the water samples originated from. After 1, 5 and 32 d, three water samples per level were assayed for total \(^{203}\text{Hg}\) and Me\(^{203}\text{Hg}\). Dissolved fractions were determined after filtration through 0.2-\( \mu m \) filters. The identificat on and quantification of Me\(^{203}\text{Hg}\) have been described elsewhere (Regnell and Tunlid 1991).

**Data analysis**—To study relationships between two variables, correlation coefficients (\( r \)) and significance probabilities (\( P \)) were computed for linear log-log regression lines. Except for total Hg and MeHg, determination of the different compounds/elements were performed on separate samples. Thus, we used mean values in the regressions. Further to illustrate relationships between MeHg and various compounds/elements, a principal component analysis (PCA) was performed using the computer program Sirius (Kvalheim and Karstang 1987).

To eliminate the effect of TOC on the variation in MeHg and total Hg levels, MeHg and total Hg were normalized (ANOVA) with respect to TOC (Herbert and Keenleyside 1995). The normalized values were based on regressions in which the variables were not log-transformed (normalized value = overall mean of variable + residual from regression).

**Results**

**Physical and chemical characteristics of the sediment and water**—After a period of warm weather in May, the summer of 1993 was unusually cold in southern Sweden, as reflected by the water temperature of Lake Levrasjön. By late May the lake was stratified. The thermocline moved downward during the summer, resulting in complete mixing of the water column in mid-October. The bottom water of the lake turned anoxic in July and stayed anoxic until October (Fig. 1). On 26 April, the first sampling date, the profundal sediment had an upper oxidized brown layer. On 25 May, it was still...
brown with a tinge of purple. From late June until late August the sediment surface was grayish-white, almost marble-like. On 29 September, a brownish floe was overlying a purple-colored layer. On 27 October, the last sampling date, the upper sediment was oxidized as evidenced by the upper brown layer.

Ranges of values for various chemical variables measured in the sediment and water are given in Table 2.

Levels of total Hg in water and sediment—The total Hg levels in both sediment and water were always low, indicating an absence of significant local sources of Hg (Table 2). Total Hg in the water, mostly below 5 pM, was uniformly distributed throughout the water column and did not show any consistent temporal trends (data not shown). However, total Hg concentrations in the upper 0.5-cm sediment layer, sampled at the five sampling stations where the water depth was 16 ± 0.5 m, decreased during summer stratification and increased thereafter (Fig. 2).

Levels of MeHg in water and sediment—MeHg levels in water below the upper boundary of the thermocline exhibited distinct seasonal variation, increasing during summer stratification to 2.6 pM in the hypolimnion, and returning to low epilimnetic values in the fall, whereas MeHg levels in epilimnetic water was rather stable at 0.5 pM (Fig. 3).

In the hypolimnetic water, MeHg were positively correlated with Mn (\( r = 0.85, P < 0.05 \)) and Fe (\( r = 0.82, P < 0.05 \)), but not with TOC and total Hg (\( r = 0.38, P > 0.05 \) and \( r = 0.37, P > 0.4 \)). Neither was there a significant correlation between total Hg and TOC (\( r = 0.65, P > 0.1 \)).

On 27 July, MeHg levels in filtered and unfiltered water increased rapidly in the thermocline and continued to increase slightly toward the sediment. On 25 August, MeHg

Table 2. Ranges of measurements of the variables indicated in unfiltered water and in surficial sediment sampled in Lake Levrasjön between 24 April and 27 October 1993. The sediment was sampled at the five stations where the water depth was 16 ± 0.5 m.

<table>
<thead>
<tr>
<th>Variable</th>
<th>Epilimnion (1 m)</th>
<th>Hypolimnion (15 m)</th>
<th>Surficial sediment (0–0.5 cm)</th>
</tr>
</thead>
<tbody>
<tr>
<td>MeHg</td>
<td>0.2–0.6 pM</td>
<td>0.2–2.6 pM</td>
<td>2.2–16 pmol g(DW)^{-1}</td>
</tr>
<tr>
<td>Total Hg</td>
<td>1.7–7.5 pM</td>
<td>2.0–6.1 pM</td>
<td>107–318 pmol g(DW)^{-1}</td>
</tr>
<tr>
<td>Mn</td>
<td>0.07–0.60 μM</td>
<td>0.16–4.50 μM</td>
<td>8.1–43.1 μmol g(DW)^{-1}</td>
</tr>
<tr>
<td>Fe</td>
<td>&lt;0.18–0.23 μM</td>
<td>&lt;0.18–1.40 μM</td>
<td>70.1–338.6 μmol g(DW)^{-1}</td>
</tr>
<tr>
<td>Ca</td>
<td>1.07–1.77 mM</td>
<td>1.76–1.32 mM</td>
<td>2.3–9.7 mmol g(DW)^{-1}</td>
</tr>
<tr>
<td>TOC</td>
<td>0.43–0.75 mM</td>
<td>0.53–0.80 mM</td>
<td>8.4–12.2 mmol g(DW)^{-1}</td>
</tr>
<tr>
<td>TIC</td>
<td>1.8–2.1 mM*</td>
<td>2.0–2.2 mM*</td>
<td>1.8–7.4 mmol g(DW)^{-1}</td>
</tr>
<tr>
<td>pH</td>
<td>8.0–8.3</td>
<td>7.1–8.0</td>
<td></td>
</tr>
</tbody>
</table>

* Not measured in the present study. Values presented were typical of 1987 and 1988.
Fig. 3. Seasonal variation in MeHg in unfiltered epilimnetic and hypolimnetic water in Lake Levrasjön during 1993. Only one sample was analyzed, except on the last three occasions (n = 2). Bars indicate range of values obtained. Some of the bars are covered by symbols.

levels increased almost linearly with depth below the thermocline (Fig. 4).

MeHg levels in the surficial sediment (0–0.5 cm), sampled at the five sampling stations where the water depth was 16 ± 0.5 m, decreased during summer stratification and increased in the fall. This result was obtained both when MeHg was expressed as an absolute concentration and as a percentage of total Hg (Fig. 5). TOC and total Hg were positively correlated with MeHg (Table 3). PCA indicated close relationships in terms of seasonal variation between MeHg, TOC, total Hg, and Mn, as these variables formed a tight cluster. This cluster was well separated from TIC, Fe, and Ca (Fig. 6).

Fig. 4. MeHg in unfiltered and filtered water, sampled in Lake Levrasjön on 27 July (n = 1) and on 24 August 1993 (n = 2). Bars indicate range of values obtained for the August measurement. Some of the bars are covered by symbols. The curves were not drawn through the epilimnetic data points since this would give the false impression of a gradient in the epilimnion.

Fig. 5. Seasonal variation in MeHg in surficial sediment (0–0.5 cm) of Lake Levrasjön during 1993, based on measurements at the five stations where the water depth was 16 ± 0.5 m. Bars are ±1 SD (n = 5, except on 26 April (the first measurement), when two samples from each station were analyzed: n = 10). *Significantly higher than the value in late July (P < 0.05); **P < 0.01; ***P < 0.005.

There was a significant negative correlation between the levels of MeHg in the sediment and those in the overlying water; r = −0.87 (P < 0.05). For Mn, this relationship was even more pronounced (r = −0.98; P < 0.005). No such correlations were found for total Hg, Fe, Ca, or TOC (data not shown).

Budgets—The seasonal change in the amount of MeHg present in the entire water mass was an order of magnitude larger than the seasonal change in the amount of MeHg present in the top 0.5 cm of the sediment. This calculation included the sediment within the part of the basin being seasonally anoxic (~60% of the entire lake area). Similar results were obtained for Mn but not for Fe (Fig. 7).

Normalized MeHg and total Hg levels in the sediment—Because both MeHg and total Hg were significantly correlated with TOC in the sediment (Table 3), it was tempting...

Table 3. Correlation matrix (linear log-log regression lines) with the variables indicated. These were measured at monthly intervals (n = 7) in the surficial sediment (0–0.5 cm) of Lake Levrasjön between 24 April and 27 October 1993. Regressions were based on mean values of 5 or 10 sediment samples, sampled at the five stations where the water depth was 16±0.5 m.

<table>
<thead>
<tr>
<th></th>
<th>TIC</th>
<th>TOC</th>
<th>Ca</th>
<th>Fe</th>
<th>Mn</th>
<th>Total Hg</th>
</tr>
</thead>
<tbody>
<tr>
<td>MeHg</td>
<td>−0.36</td>
<td>0.90*</td>
<td>0.46</td>
<td>0.63</td>
<td>0.74</td>
<td>0.80*</td>
</tr>
<tr>
<td>Total Hg</td>
<td>−0.75</td>
<td>0.86*</td>
<td>0.15</td>
<td>0.42</td>
<td>0.57</td>
<td></td>
</tr>
<tr>
<td>Mn</td>
<td>−0.44</td>
<td>0.88*</td>
<td>0.19</td>
<td>0.35</td>
<td></td>
<td></td>
</tr>
<tr>
<td>Fe</td>
<td>0.02</td>
<td>0.53</td>
<td>0.94*</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Ca</td>
<td>0.35</td>
<td>0.34</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>TOC</td>
<td>−0.52</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

* P<0.05; **P<0.01.
Fig. 6. A plot based on a PCA model, the variables being those indicated in the figure (measured in surficial sediment [0–0.5 cm] of Lake Levrassjön), and the objects (cases) being the seven dates on which measurements were made (between late April and late October 1993). The sediment was sampled at the five sampling stations where the water depth was 16 ± 0.5 m. Mean values of these samples were used in the calculation. All variables were standardized (SD = 1) prior to performing the PCA. The variance explained by each PC, expressed as percentage of total variance, is given in parentheses. The large cross indicates origin.

to normalize them with respect to TOC and study the remaining variation. When the variables were not log-transformed, correlation coefficients (r) were 0.95 (P < 0.005) for MeHg and 0.83 (P < 0.05) for total Hg. For total Hg most of the seasonal variation after normalization (ANCOVA) was explained by one elevated value in late May and one low value in late October. Normalized MeHg, however, displayed an almost sinusoidal curve between late April and late October, the trough being in mid-July (Fig. 8).

**Incubation experiment in the water column**—There was no Me$^{203}$Hg in the bottles after 1 and 5 d of incubation. After 32 d, Me$^{203}$Hg was present in all anoxic bottles. No Me$^{203}$Hg was produced in the bottles containing water from the epilimnion and the upper thermocline. The production of Me$^{203}$Hg was highest in the bottles containing water from the middle of the thermocline, the production decreasing toward the bottom. Sulfide levels increased with depth and were present in all bottles except in those containing epilimnetic water (Fig. 9).

Already after 1 d of incubation, >90% of the added $^{203}$Hg(II) was in the particulate fraction in the bottles containing water from the middle of the thermocline and below, whereas most of the MeHg that appeared after 32 d of incubation passed 0.2-μm filters (Fig. 10).

Fig. 7. Seasonal variation in the amount of (A) MeHg, (B) Mn, and (C) Fe in the entire water mass and in surficial sediment (0–0.5 cm) of Lake Levrassjön during 1993. The surficial sediment (●) is that part of the lake having a water depth of ≥10 m (~60% of the entire lake area). It was assumed that the sediment samples from the five sampling stations (water depth of 16 ± 0.5 m) were representative of this part of the basin. The sediment outside this part of the basin was not included in the calculation.
Discussion

Seasonal variation in MeHg levels—MeHg levels in metalimnetic and hypolimnetic water have been shown to vary seasonally in a number of lakes, with a typical buildup of MeHg during summer stratification and return to low MeHg levels following turnover (Bloom et al. 1991; Jacobs et al. 1995; Parks et al. 1989; Watras and Bloom 1994). However, we think that the present study is unique in demonstrating seasonal variation in MeHg levels in surficial sediment. Furthermore, MeHg levels in the surficial sediment and those in the overlying hypolimnetic water were negatively correlated.

In previous experiments with sediment cores, including sediment cores from Lake Levrasjon, we have demonstrated that oxic conditions favor sediment uptake of both total Hg (essentially inorganic Hg(II)) and MeHg, whereas anoxic conditions favor release of total Hg and MeHg from the sediment (Regnell 1994; Regnell and Tunlid 1991; Regnell et al. 1996). These results are in line with our present field observations that during anoxia there was a decrease in MeHg levels in the sediment and an increase in MeHg levels in the overlying water, and that this trend was reversed during the reoxygenation of the sediment-water interface. Thus, we conclude that redox-controlled exchanges of MeHg between the sediment and water contributed to the seasonal variation in MeHg levels in the sediment and water of Lake Levrasjon.

Another possible explanation of the fact that MeHg levels in the water and sediment were negatively correlated is that methylation in water and sediment responded differently to changing redox conditions. Decreasing redox potentials in the sediment during summer stratification, the sediment being anoxic already at the onset of stratification except for the uppermost millimeters, may have led to inhibition of Hg methylation in the sediment because of increased sulfide levels (Berman and Bartha 1986; Craig and Moreton 1985). In the water, which is well oxygenated in the spring, decreasing oxygen levels instead may have favored Hg methylation, because Hg-methylating microorganisms are found mainly (or exclusively) among anaerobic microorganisms (Compeau and Bartha 1984; Callister and Winfrey 1986; Regnell et al. 1996). In fact, our results clearly show that both methylation and partitioning of MeHg must be interred to explain the observed seasonal variation in MeHg levels in the sediment and water (see below).

In late July, a peak in MeHg may have been present near the lower boundary of the thermocline, or right below it, and further down there was only a weak increase in MeHg levels toward the sediment. This indicated that Hg methylation in the anoxic water explained most of the hypolimnetic buildup.
of MeHg. In late August, however, MeHg showed a strong gradient toward the sediment, indicating that MeHg released from the sediment had a strong influence on the MeHg depth profile (Fig. 4). The incubation experiment, starting in mid-September, provided further evidence for this, because it indicated that the potential for MeHg production in the water was highest in the middle of the thermocline (Fig. 9). Thus, increasing MeHg levels toward the sediment, as seen in late August, was probably not supported by a corresponding increase in Hg methylation in the water column. In line with these results, normalization (ANCOVA) of MeHg levels in the sediment with respect to TOC indicated decreasing MeHg production in the sediment during June and July and increasing production during August (see below). However, it cannot be excluded that MeIIg associated with particles (e.g. senescent bacteria) sedimented from sites of high methylation in the water column down to the sediment surface. There, mineralization should lead to MeHg recycling back to the water, the sediment acting as an indirect source of MeHg. Whatever caused the increase in MeIIg in the bottom water during August, this increase raised the concentration of MeHg by no more than 0.4 pM (Fig. 3). Thus, our conclusion is that, although the sediment released MeHg during summer stratification, as evidenced by a decreasing MeIIg content in the surficial sediment, water column methylation of Hg explained most of the hypolimnetic buildup of MeHg. This conclusion gain further support from our budget calculation (see below).

We think that decreasing net production of MeHg in the water below the middle of the thermocline, as indicated by the incubation experiment, was caused by the fact that sulfide levels increased toward the sediment (Fig. 9). It has been shown in several studies that sulfide inhibits Hg methylation, and that the highest Hg methylation rates are found where sulfide and dissolved oxygen levels are both low (Jackson 1988a). This may seem to preclude high Hg methylation rates in the sediment during the later stage of summer stratification, because extrapolating sulfide levels in the downward direction would result in higher sulfide levels in the sediment than in the water column. However, in laboratory experiments we observed high methylation rates in sediment even when sulfide levels in the overlying water were high (Regnell et al. 1996). Also, Furutani and Rudd (1980) found that Hg(II) was methylated in sediment despite the presence of sulfide. Possibly, methylation of Hg(II) proceeds less hindered by sulfide in sediment than in water. Available sulfide capacity (FeS formation) in the sediment (Davies-Colley et al. 1985) may suppress the levels of free sulfide (HS~ and H2S), thereby preventing the formation of inert HgS(s). It is also possible that organic compounds containing thiol groups effectively compete with sulfide in the binding of Hg(II) (Dyrrsen and Wedborg 1991; Wallschlager et al. 1996). It has been shown that Hg(II) bound to thiols like cysteine is readily methylated by microorganisms, whereas HgS(s) is not (Craig and Moreton 1985).

Vertical profiles of MeHg reported from several lakes suggest significant water column production of MeHg (Verta and Matilainen 1995; Watras and Bloom 1994; Watras et al. 1995). However, in Clay Lake, Canada, with highly Hg-contaminated sediments, it was found that MeHg levels in the water column increased toward the bottom sediment, indicating significant Hg methylation in the sediment (Furutani and Rudd 1980). Presumably, in lakes with severe Hg contamination, it is more likely that the major source of MeHg is the sediment than in less contaminated lakes.

Processes other than methylation of Hg(II) and partitioning of MeHg that may have caused seasonal variation in MeHg levels in the sediment and water of Lake Levrajsjon are mass dilution in the sediment due to settling material, demethylation, and uptake/release of MeHg by the lake biota (see below).

Budget calculations—According to our budget calculations, internal cycling of a fixed amount of MeHg between the sediment and the water could not explain the observed seasonal variation in MeHg in the water, unless MeHg was exchanged to and from sediment depths an order of magnitude deeper than 0.5 cm (Fig. 7A).

It was impossible to separate all overlying water from the sediment, but we are confident that the amount of MeIIg in the sediment was not seriously miscalculated as a result of erroneous water content determinations. Given the organic content of the sediment, and considering the fact that we only collected the upper 0.5 cm layer, our water content determinations are in reasonable agreement with values provided by Häkansson and Jansson (1983). Furthermore, based on the cross-section area of the sampler, and the dry weight of the samples, we estimated a water content of 94.7%, not far from the 96% that was used in the calculation above.

Evidently, the increase in the amount of MeHg in the water between June and August was considerably larger than the concomitant decrease in the amount of MeHg in the upper 0.5-cm sediment layer within the basin turning anoxic. This provided further evidence that the water column was a larger source of MeHg than the sediment. A high rate of Hg methylation in the sediment, where yet the rate of release of MeHg exceeded the rate of new production, is less likely considering the depth profile of MeHg in late July (Fig. 4).

Factors explaining the discrepancy between the estimated losses of MeHg from the water and the estimated gain in MeHg in the sediment from August onward may have been demethylation and uptake of MeHg by the lake biota. However, it is also possible that we underestimated the amount of MeHg deposited onto the sediment (see below).

Sediment core studies at our laboratory have indicated that downward migration of MeHg in sediment is a rather slow process (Regnell et al. 1996). Furthermore, sedimentation of new material during the study period corresponding to much more than a few millimeters of sediment does not seem likely either. Sediment trap data from Lake Levrajsjon during 1993 (obtained from colleagues; Larsson and Okla pers. commun.) and our finding that the surficial sediment had a water content of 96% indicated a monthly sedimentation corresponding to ~1 mm of surficial sediment. Thus, it is not likely that MeHg that disappeared from the water between August to November (Fig. 7A) should be found deeper down in the sediment than 0.5 cm.

Seasonal variation in total Hg levels—Although the seasonal variation in total Hg levels in the sediment (Figs. 2,
Mechanisms behind exchanges of MeHg and total Hg at the sediment–water interface—It is well known that Fe(III) and Mn(III and IV) oxides/oxyhydroxides have high capacities for adsorbing several heavy metals and a variety of organic compounds (Dyrssen and Kremling 1990; Jackson 1988b; Wang and Lee 1993). When conditions turn anoxic at the sediment–water interface, Fe and Mn are reduced to Fe(II) and Mn(II), whereby substances previously bound in the oxic surficial sediment layer may be released to the water. Upon reoxidation in conjunction with lake turnover, freshly precipitated Mn and Fe oxides/oxyhydroxides may scavenge substances previously released from the sediment during anoxia and settle out to the sediment. Given that both inorganic Hg(II) and MeHg are complexed to organic ligands in lake water (Dyrsen and Wedborg 1991; Jackson et al. 1980; Wallslauger et al. 1996; Zepp et al. 1974), it seems likely that they are adsorbed by the oxides via organic ligands or that they are exchanged between these ligands and organic coatings on the oxides.

When looking at simple linear regressions between different constituents of the sediment, it is evident that Mn, total Hg, and MeHg showed seasonal variations similar to TOC (Table 3). A PCA provided further illustration of the relationship between MeHg and the other constituents that we measured in the sediment (Fig. 6). In the reproduced space defined by the first and second PC, which explained 86% of the entire seasonal variation in the concentration of the various constituents of the sediment, there appeared a cluster consisting of Mn, TOC, total Hg, and MeHg, well separated from Fe, Ca, and TIC. This indicated that both MeHg and total Hg were associated with organic matter, and that Mn rather than Fe controlled the uptake/release of the organic fraction with which the Hg was associated. This is consistent with the fact that Mn undergoes much more profound redox cycles than Fe, Fe(III) being more stable toward reduction than Mn(III and IV) (Dyrsen and Kremling 1990; Hamilton-Taylor and Morris 1985). Furthermore, the water solubility of Mn(II) unlike that of Fe(II) is relatively insensitive to sulfide. In agreement with our findings, Jacobs et al. (1995) concluded that the Mn redox cycle contributed to the temporal and spatial variation of MeHg in the water of hypereutrophic Onondaga Lake, New York.

Mn was released from the sediment as early as May, whereas release of MeHg was not evident until the sampling in late June (Fig. 7A, B). In the bottom water, the Mn concentration increased from 0.2 to 3.1 μM between late April and late May, whereas the concentration of MeHg decreased somewhat from 0.5 to 0.2 pM. A surficial oxic sediment layer was still present in late May and evidently this layer was sufficient to prevent upward diffusion of MeHg. The ability of Mn(II) to diffuse through such a layer is in line with findings by others (Davison et al. 1982). Given the higher concentration of Fe than of Mn in the top 0.5-cm layer of the sediment, the thin oxic layer (1–2 mm) probably contained more Fe than Mn. It is thus reasonable that Fe oxyhydroxides participated in the adsorption of MeHg, slowing down the release of MeHg, unless Fe oxyhydroxide is a much less efficient scavenger of MeHg than Mn oxide/oxyhydroxide.

Gagnon et al. (1996) also concluded that a surficial oxic sediment layer (marine sediment) served as a barrier to the diffusion of MeHg into the overlying water. Because the MeHg concentration in this layer was low, compared to the MeHg concentrations deeper down in the sediment, they suggested that demethylation in the oxic layer prevented upward diffusion of MeHg. In contrast to their findings, the concentration of MeHg in the surficial sediment (0–0.5 cm) of Lake Levraessjon was highest when a surficial oxic layer was present. Thus, it does not seem likely that demethylation played a major role in preventing release of MeHg from oxic sediment in our study.

It has been demonstrated that the hydroxyl radical (OH•) is able to degrade MeHg to inorganic Hg (Suda et al. 1991). Oxidation of Fe(II) to Fe(III) is known to produce H2O2, which in turn reacts with the remaining Fe(II) to produce hydroxyl radicals (King et al. 1995). Because Fe(II) oxidation occurs mainly at the oxic–anoxic interface (King et al. 1995), it is tempting to hypothesize that MeHg is abiotically degraded by this mechanism in or near the oxic surficial sediment layer. Furthermore, during lake turnover, production of hydroxyl radicals may also take place in the water. Whether MeHg is degraded to a significant extent in connection with Fe(II) oxidation may depend on several yet unknown factors.

Precipitation of CaCO3 occurred in late summer–early fall in Lake Levraessjon, raising the CaCO3 content of the sediment significantly (data not shown). However, CaCO3 appeared to have been a poor scavenger of MeHg because MeHg levels in the hypolimnetic water was relatively unaffected by this event (Fig. 3). In line with this finding Sigg et al. (1987) concluded that CaCO3 was an inefficient scavenger of heavy metals (Hg was not included in the study) in Lake Zurich, Switzerland.

We were surprised that Fe and Ca levels in the sediment showed strong covariation (Table 3). The fact that they increased simultaneously indicated simultaneous precipitation (data not shown). However, formation of FeCO3(s) probably did not take place, because no precipitation of Mn was observed. Saturation with respect to MnCO3(s) should precede that of FeCO3(s), considering the facts that Mn concentrations were higher than Fe concentrations in the anoxic water and that the solubility product is somewhat lower for MnCO3 than for FeCO3 (Verdouw and Dekkers 1980). Rather, formation of CaCO3(s) and increased sulfide production in the hypolimnion coincided, the latter causing Fe(II) to precipitate (but not Mn(II)).
Evidence for lateral transport of MeHg—As for Mn, the amplitude of change in the amount of Mn in the water was an order of magnitude greater than in the 0–0.5-cm upper sediment layer within the part of the basin being seasonally anoxic (Fig. 7A, B). Apart from Mn probably being released by sediments outside this area (Davison et al. 1982), mobilization of Mn might have occurred much deeper down in the sediment than 0.5 cm. A study by Verdoux and Dekkers (1980) in Lake Vechten (the Netherlands) indicated that Mn released to the hypolimnion was mobilized from a depth of at least 12 cm in the sediment.

More of interest in the present study is the fact that, according to our calculations, the decrease in the amount of Mn in the water during turnover in the fall was an order of magnitude greater than the concomitant increase in the 0–0.5-cm sediment layer within the seasonally anoxic part of the basin. The missing Mn was probably partly explained by an increased settling of Mn on the sediment surface as the water depth decreased within this part of the basin. Hamilton-Taylor and Morris (1985) found that the Fe : Mn ratio in the surface sediment decreased from 27.3 at a water depth of 15.5 m to 4.5 at a water depth of 5.5 m along a midsummer transect in Esthwaite Water (U.K.). They explained this phenomenon by Mn being mobilized to the water at high rates from the anoxic sediment situated in the deepest parts of the lake. Mn thus released then precipitated once it reached higher water strata and settled on the sediment situated in the more shallow parts of the lake, where dissolved oxygen in the water overlying the sediment was sufficient to prevent redissolution. Thus, our assumption that the sediment surface below a water depth of 16 ± 0.5 m was representative of the sediment surface within the seasonally anoxic basin (water depth, ≥10 m) probably led to an underestimation of the amount of Mn that settled to the sediment within this area. Furthermore, owing to water movements during turnover, large amounts of Mn may have settled outside this area.

Given that Mn oxides will adsorb MeHg in the water, MeHg might display the same lateral movement as Mn, i.e. from sediment situated in the deepest part of the lake basin via the water column to sediment within the more shallow parts of the basin. Thus, using similar arguments for MeHg as for Mn, the missing MeHg (Fig. 7A) could be partly explained by an increased settling of MeHg onto the sediment as the water depth decreased. If a lateral movement of MeHg in the direction from profundal to littoral sediment is significant, this might have significant implications for the bioavailability of MeHg in a lake. Additionally, the losses of MeHg from the water could be explained by accumulation of MeHg in the lake biota (Slotton et al. 1995) and demethylation (Sellers et al. 1996). If the depletion of MeHg in the water was caused mainly by these processes, the lateral transport of MeHg as here described might be insignificant.

Looking at the seasonal variation in the amount of Mn and MeHg in the water (Fig. 7A, B), it is evident that the decline in Mn was delayed in comparison with the decline in MeHg. This was probably explained by the fact that Mn was mobilized continuously from the profundal sediment, as evidenced by increasing concentrations of Mn in the hypolimnetic water (data not shown), whereas mobilization and production of MeHg was insufficient to compensate for the losses of MeHg to the sediment over which the thermocline passed.

For Fe, in contrast to Mn and MeHg, the increase in the amount in the sediment at the end of summer stratification was larger than the concomitant decrease in the water (Fig. 7C). The main reason for this was probably that the lateral movement we just described for Mn was virtually nonexistent for Fe, owing to the much lower propensity for Fe to become soluble under anoxic conditions (Hamilton-Taylor and Morris 1985; Verdoux and Dekkers 1980). Thus, sediment focusing and vertical movement of Fe within the sediment could possibly account for the changes in Fe levels in the surficial sediment within the deeper parts of the basin. Again, our results indicate that the cycling of MeHg followed the redox cycle of Mn rather than that of Fe.

Variation in MeHg expressed as a proportion of total Hg—When MeHg was expressed as a proportion of total Hg, there was still a significant decline in MeHg in the surficial sediment during the initial phase of anoxia and a significant recovery during the autumn months (Fig. 5). The initial decline during anoxia could be explained by increased solubility of MeHg owing to the formation of the complex MeHgS⁻ (or MeHgSH), while the release of inorganic Hg(II) from the sediment, if affected at all by sulfide, was counteracted by the coprecipitation of Hg(II) with FeS (Dyrssen and Hedborg 1991; Zepp et al. 1974). In the incubation experiment, in accordance with this conclusion, almost all of the added inorganic Hg(II) was in the particulate fraction (>90%) in the anoxic water already after 1 d of incubation, whereas most of the MeHg produced was dissolved (passed 0.2 μm) (Fig. 10).

The increase in MeHg as a proportion of total Hg in the surficial sediment during August was probably explained by increased methylation in the sediment, or by sedimentation of particulate matter rich in MeHg, e.g. scenscent bacteria, which more than compensated for the released MeHg. Later in the season there was no indication of high methylation rates in the sediment, so the fact that MeHg as a proportion of total Hg kept increasing was probably a result of higher uptake/settling rates of MeHg than of total Hg (see below).

Seasonal variation in sediment MeHg and total Hg normalized (ANOVA) with respect to TOC—In the sediment, TOC was significantly correlated to both MeHg and total Hg, which is in line with the fact that both MeHg and inorganic Hg(II) have strong affinities for organic matter (Table 3).

Normalizing with respect to TOC, it was evident that almost all residual variance in total Hg was caused by a peak value in late May and a decline during October. For MeHg, in addition to a similar peak and decline, residual variance was caused by a trough during summer stratification (Fig. 8). Whereas the peak in late May is difficult to explain, the decline in October could be a result of dilution by freshly sedimented organic matter, less enriched in inorganic Hg and MeHg than the organic matter in the sediment, and/or a result of volatilization owing to reduction to Hg(0). The di-
lution hypothesis is supported by the fact that the organic matter content of the sediment, after a steady decrease, started to increase during October (data not shown). Increased degradation of MeHg in October is in line with the fact that several studies have demonstrated thatoxic conditions favor degradation rather than production of MeHg (e.g. Compeau and Bartha 1984; Olson and Cooper 1976; Ramjal et al. 1986). Note, however, that both total Hg and MeHg levels on a simple dry weight basis increased in the sediment during October (Figs. 2, 5). Thus, although depleted in normalized MeHg and total Hg, the settling matter that reached the sediment during October was enriched in MeHg and total Hg on a dry weight basis relative to the surficial sediment.

The decrease in normalized MeHg in the sediment during June and July was probably a result of continuous release of an organic fraction rich in MeHg, increased solubility of MeHg owing to sulfide production, and low methylation rates. The increase in normalized MeHg during August may have been caused by increased methylation, supported by the fact that MeHg levels increased both in the sediment and in the overlying water in August (Figs. 3, 5). Sedimentation of easily degradable organic matter, originating from crashed algal populations, may have stimulated methylating microorganisms in the surficial sediment during August. Alternatively, as already noted, sedimentation of MeHg-enriched particles of biological origin may have caused the increase in normalized MeHg in the sediment during August.

Representativity of the present results of lakes in general—We are aware that there could be several alternatives to our interpretation of the present data, and that further research is needed before firm conclusions can be drawn concerning the dynamics of MeHg in lakes. Furthermore, the sources of MeHg, its partitioning between sediment, water and biota, and its degradation may differ considerably between lakes of different categories. We think that our findings are representative of eutrophic dimictic lakes with high hypolimnia and possibly also for deep reservoirs. For boreal forest lakes, the most common lake type in Canada, northern U.S., Finland, and Sweden, and especially for shallow ones with rapid water renewal, processes within the watershed (e.g. in upstream wetlands) are likely to have a major influence on MeHg levels (Branfireun et al. 1996; St. Louis et al. 1994).

Conclusions

In Lake Levrasjön, most of the hypolimnetic buildup of MeHg during summer stratification was probably explained by Hg methylation in the water column and in the oxic–anoxic boundary, with only a minor contribution from the sediment. The Mn redox cycle seemed to have a strong influence on the exchange of MeHg between water and sediment. Anoxic conditions at the sediment–water interface, leading to dissolution of Mn oxides/hydroxides, allowed diffusion of MeHg from the sediment, whereas a thin oxic sediment layer containing Mn oxides/hydroxides was sufficient to prevent its release to the overlying water. During the downward migration of the thermocline, waterborne Mn(II) was reoxidized and precipitated, adsorbing MeHg while settling to the sediment. Prior to complete lake turnover, precipitated Mn probably redissolved in the anoxic bottom water in the deepest parts of the lake. This may have led to a lateral transport of both Mn and MeHg in the direction from profundal sediment to littoral sediments. However, processes other than sedimentation leading to depletion of MeHg in the water, such as demethylation and uptake of MeHg into the biota, may have reduced the importance of such a lateral transport of MeHg. Total Hg (essentially inorganic Hg(II), MeHg, and Mn were all correlated with the TOC content in the sediment. This indicated that Mn oxides/hydroxides scavenged organic matter to which inorganic Hg and MeHg were associated, so that the scavenged organic matter bound these Hg fractions in a second step. MeHg showed stronger seasonal variation than total Hg in both water and sediment, presumably because of seasonal variation in net methylation and because of reactions with sulfide, where sulfide increased the solubility of MeHg and decreased that of total Hg. Furthermore, unlike for MeHg, the atmosphere was probably a significant source of total Hg, obscuring any effect of sediment uptake/release on total Hg levels in the water. Further research is clearly needed to evaluate and understand how different processes affect MeHg levels in lake water and sediment.

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