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Determination of Methyl- and Ethylmercury in Natural Waters at Sub-nanogram per Liter Using SCF-Adsorbent Preconcentration Procedure

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An analytical procedure for determining methyl- and ethylmercury (MeHg/EtHg) in natural waters is described. MeHg/EtHg was preconcentrated from water on a sulfhydryl cotton fiber (SCF) adsorbent and eluted with a small volume of a mixture of 1M hydrochloric acid and 2M sodium chloride. The eluate was extracted with benzene. The measurements of MeHg/EtHg in benzene extract were determined by gas chromatography with electron capture detector. The detection limit for MeHg/EtHg was about 0.04 ng L^{-1} using a 20L water sample. The precision was about 20%. The application of the proposed method to one snow and four freshwater samples varying in humus content is described. The MeHg concentrations found in different freshwater samples were ranged from 0.09 to 0.22 ng L^{-1} and the recoveries of spiked MeHg were ranged from 42 to 68% which were strongly correlated to the content of humic substances.

The MeHg concentration found in snow was 0.28 ng L^{-1} and the recovery was 79%.

The analytical results of MeHg concentration in freshwater samples are discussed in relation to the pH used in the preconcentration, the humus content, the fraction of methylmercury in organic bound mercury and mercury in fish.

KEY WORDS: Methylmercury, alkylmercury, fresh water, preconcentration, adsorbent.

1. INTRODUCTION

Alkylmercury, especially methylmercury (MeHg), is one of the most important mercury species in natural water and is of primary concern because of its extremely toxic nature and its capability of accumulating in fish.

MeHg can be synthesized from inorganic mercury by microorganisms within sediments,¹ and by humic substances in soil² and in humus-rich water.^{3,4}

In recent years, a relatively high concentration of MeHg has been found in fish in remote Swedish lakes subject only to airborne deposition of Hg. It is therefore desirable and important to know the actual concentrations of dissolved MeHg in Swedish aquatic environments and to study both the pathways by which MeHg has been transported in aquatic and terrestrial systems and the mechanism of MeHg accumulation in fish.

Because of the extremely low concentrations of MeHg in natural water ($<1 \text{ ng L}^{-1}$), which are below the sensitivities of recent analytical techniques, there is a need to develop a sensitive and selective analytical method for determining extremely low concentrations of MeHg at sub-ppt level.

The analytical methods applied in previous work on the determination of MeHg in natural waters in general require the MeHg to be preconcentrated. The most accepted preconcentration procedure involves extraction of methylmercury chloride (MeHgCl) from an aqueous solution into an aromatic solvent (benzene, toluene). Because the distribution ratio of MeHgCl between these organic solvents and water solution was not very high,⁵ about 5 to 10, there were difficulties in concentrating MeHg from natural water by a factor more than ca. 5000.

Recently, several preconcentration methods⁵⁻⁸ using resins/adsorbent for concentrating MeHg from water have been reported. Yu & Liu (1981)⁷ reported that sulfhydryl cotton fiber (SCF) adsorbent was a promising one for concentrating MeHg from natural waters, as it can quantitatively adsorb MeHg and inorganic mercury in water solution, and the adsorbed MeHg can be eluted from the adsorbent with a small volume of hydrochloric acid and separated from the inorganic mercury.

As the humus content varies in Swedish freshwater lakes and the

MeHg concentration is likely to be lower than 0.5 ng L^{-1} , we have tested the preconcentration method using SCF-adsorbent and a modification of the method was then developed and assessed. A gas chromatograph with electron capture detector (GC/ECD) has been applied to determine methyl- and ethylmercury after 1000 to 20000-fold preconcentration.

2. EXPERIMENTAL SECTION

2.1 Reagents and solutions

All chemicals used were of a certified grade. The stock solutions of methylmercury chloride (MeHgCl) and ethylmercury chloride (EtHgCl) were prepared by dissolving them in benzene or deionized water. The analytical standard aqueous solutions of MeHgCl and EtHgCl (about 10 to 30 ng/mL) were freshly prepared for each set of analyses by appropriate dilution from the stock solutions before use and were calibrated using helium dc plasma atomic emission spectroscopy (plasma-AES)/sodium borohydride method.^{9,10} The stock solutions of sodium chloride and hydrochloric acid were washed with benzene before use.

2.2 Preparation

Preparation of adsorbent (sulfhydryl cotton fiber SCF).⁷ About 10 g cotton was placed in a pyrex bottle with ground-glass stopper containing a mixed solution prepared by adding the following solution in sequence: 50 ml of thioglycolic acid, 35 mL of acetic anhydride, 16 mL of acetic acid (36%), 0.15 mL of concentrated sulfuric acid and 5 mL of deionized water. The cotton was soaked in the mixed solution for 4–5 days at 40 to 45°C. The product was collected by suction filtration and washed thoroughly with deionized water until the filtrate was neutral. The SCF-adsorbent obtained was dried at 40 to 45°C (for about six hours) and then kept in a brown glass bottle with ground-glass stopper and was stored in a refrigerator (with no significant changes within 2 to 3 months).

2.3 Cleaning of utensils

Pyrex sampling bottles (10 L), teflon tubes and glass columns were

leached in 2% HCl for at least 24 hours. Other Pyrex glassware used in experiments was previously treated with aqua regia and carefully rinsed with deionized water.

2.4 Apparatus

An HP 5710A gas chromatograph equipped with an electron capture detector was employed for analysis of MeHgCl and EtHgCl. The gas chromatographic columns and operating conditions are summarized in Table I.

Table I Operating conditions for the gas chromatograph

Column	Pyrex, 1.8 m × 2 mm i.d.
Column packing	7% PDEAS ^a on DMCS-treated 60/80 mesh Chromosorb
Column temp., °C	160
Injector temp., °C	200
Detector oven temp., °C	300
Carrier gas	Argon (5% methane)
Carrier gas flow rate, mL/min	30

^aPDEAS = phenyldiethanol amine succinate.

2.5 Procedure

A number of samples of artificial water containing 0.05 to 20 ng L⁻¹ MeHg/EtHg as Hg were first tested, and the improved method was then applied to determine MeHg/EtHg in samples of melted snow and fresh water. The blank was also checked concerning the additional contribution of MeHg/EtHg from reagents and the method used.

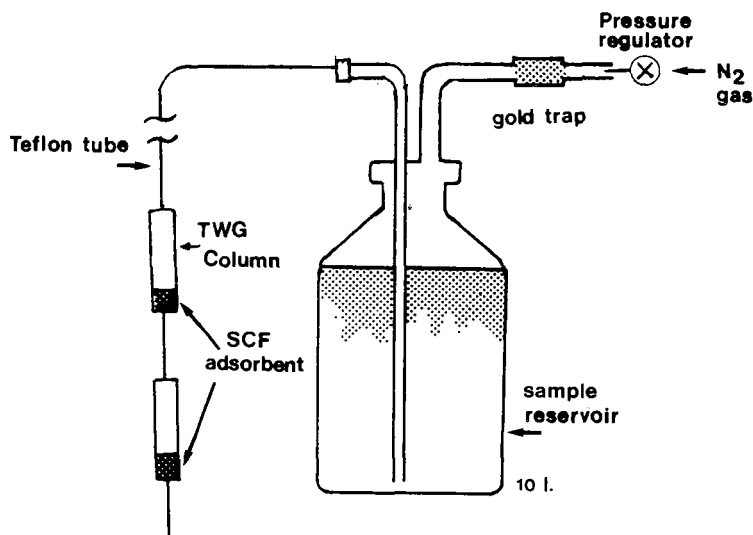
2.5.1. Sampling Fresh water samples were collected in Pyrex bottles with ground-glass stoppers, which were previously acid-washed and rinsed with lake water at the sampling sites. Three lake water samples and one run-off water sample from remote sites, each with a different humus content, were chosen for the present study. The available data of pH, color, KMnO₄ value and MeHg concentration in pike are given in Table II. The snow sample was collected from Lake Mjörn district on March 3, 1986.

Table II Data on pH, color, KMnO_4 value and Hg concentration in pike

	pH	Color mg L^{-1} Pt	KMnO_4^a mg L^{-1} O	Hg in 1-kg pike ^a ppm
Lake St. Skärsjön	4.39	90	65–75	0.9
Ladebacksviken (run-off)	4.12	32	—	—
Lake Klintsjön	5.12	6	10	1.5
Lake St. Övattnet	6.93	32	—	—

^aData provided by Marcus Meili.

2.5.2 Determination of MeHg and EtHg from water samples About 0.2–0.4 g of SCF-adsorbent was packed into a Pyrex glass column (Werner TWG column), ca. 10 mm in inner diameter. The top of the column was connected to the sample reservoir (Figure 1). After pH adjustment all samples were processed through the column at a suitable flow rate controlled by nitrogen gas pressure. 3 mL of 2N HCl/(1 M HCl + 2 M NaCl) divided into three portions were pipetted onto the adsorbent surface to elute the MeHgCl and EtHgCl

**Figure 1** Diagram of the preconcentration arrangements.

from the column. 1 mL of benzene was added to the eluate and shaken vigorously for 20 min. The MeHg/EtHg in the benzene phase was analyzed by gas chromatography using an electron capture detector (GC/ECD).

In the case of fresh water and snow samples, two SCF-adsorbent columns connected in series were used in order to check whether MeHg/EtHg was completely adsorbed onto the first column. The eluates from two columns were then combined for subsequent benzene extraction. Water samples were analyzed both with and without a spike of MeHgCl/EtHgCl in order to evaluate the recovery of MeHgCl/EtHgCl. A flow rate of less than 20 mL/min · cm³ was used in the preconcentration of water samples.

The extraction efficiencies (E) of extracting MeHgCl/EtHgCl from eluate into benzene were determined experimentally using 3 mL/6 mL eluant containing known concentrations of MeHgCl/EtHgCl (3 to 20 ng/mL), corresponding to the concentrations of MeHg/EtHg found in eluate after preconcentration. The value of extraction efficiency was also checked for every new set of analyses.

2.5.3 Determination of organic bound mercury To provide information on the relative abundance of the MeHg/EtHg compared with the other forms of organic bound mercury present in fresh water, and to determine the efficiency of the SCF-adsorbent preconcentration procedure for organic bound mercury, including MeHg/EtHg, the organic bound mercury in fresh water samples was estimated before and after preconcentration by subtracting the value of "reactive mercury" from the total mercury value. The analyses of the "reactive mercury" and the total mercury were achieved through reduction with SnCl₂ and NaBH₄ respectively, and using helium dc plasma-AES method.^{9,10}

3. RESULTS AND DISCUSSION

3.1 GC-analysis

A typical gas chromatograph obtained under the conditions used (see Table I) is shown in Figure 2. The retention time of MeHgCl and EtHgCl was 1.8 min and 3.2 min, respectively. The peaks of

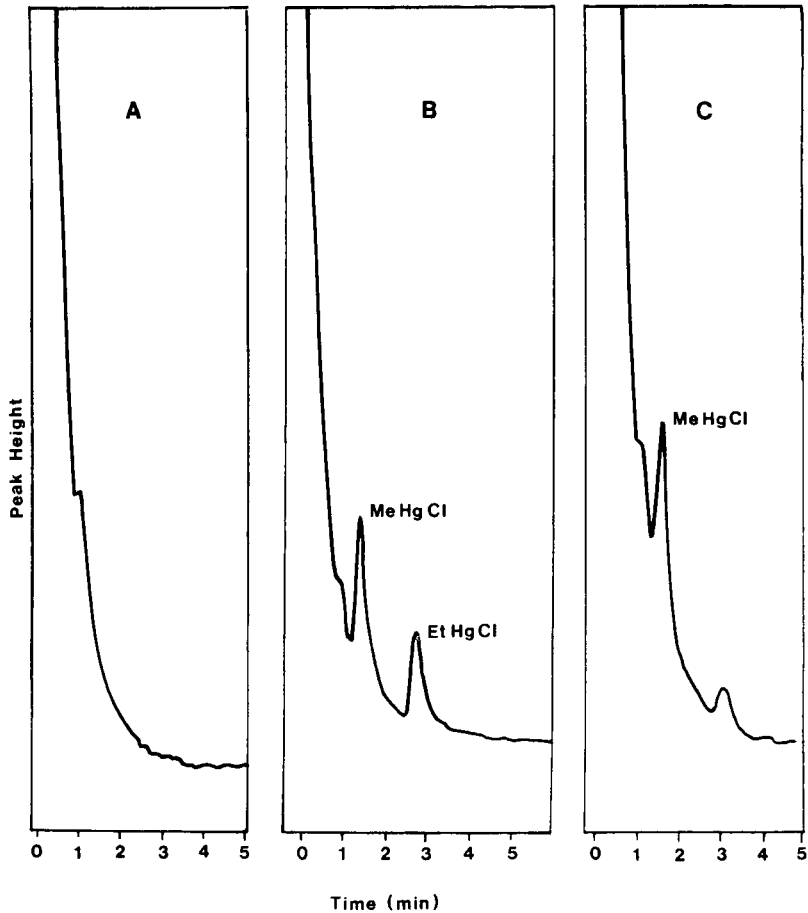


Figure 2 Gas chromatograms (A) benzene (B) 5.0 μL standard benzene solution: 2.5 ng mL^{-1} MeHgCl, 2.5 ng mL^{-1} EtHgCl (C) 6.5 μL benzene-extract after 20000-fold pre-concentration in fresh water (Lake Klintsjön).

MeHgCl and EtHgCl are adequately separated from each other. The absolute detection limit of GC for MeHgCl/EtHgCl is about 2 to 3 μg using 6 μL injection-volume. The uncertainty of GC measurement was 5 to 10% of the measured value. The precision is generally lower when the concentration approaches the detection limit.

3.2. Preconcentration treatment

3.2.1 Effects of pH, flow rate and interference of inorganic mercury The effects of pH, flow rate and interference of inorganic mercury on the preconcentration procedure are summarized in Table III. It can be seen that MeHg/EtHg was quantitatively adsorbed onto the adsorbent above pH 2.5. At a higher pH (5 to 8), as reported by Yu and Liu (1981), the adsorption of MeHg was also complete, but other heavy metal ions, such as Cu^{2+} , Pb^{2+} , Zn^{2+} , Cd^{2+} , could also be adsorbed to compete with MeHg/EtHg. It is therefore preferable to use a lower pH for preconcentrating MeHgCl/EtHgCl. Table III also shows that a flow rate of less than ca. $44 \text{ mL}/\text{min} \cdot \text{cm}^2$ allowed complete collection of MeHg/EtHg from artificial water, and the inorganic mercury did not interfere with the determination of MeHg/EtHg and that no detectable MeHg/EtHg was found in the blank.

Table III Effects of pH, flow rate and the interference of inorganic mercury on the procedure results

pH	Flow rate ml min^{-1} approx.	Hg(NO ₃) ₂	MeHgCl	EtHgCl	MeHgCl	EtHgCl
		spiked, ngL^{-1}	added,	ngL^{-1}	recovery	%
4	10–35	—	0.05–20	0.05–0.9	98.8 ± 7.7 (<i>n</i> = 10)	101 ± 7 (<i>n</i> = 6)
4	7	—		blank	(<i>n</i> = 2)	—
4	< 3	4	0.2–0.5		91.5 ± 4.7 (<i>n</i> = 2)	—
2.5–4	20	—	0.5		101 ± 3 (<i>n</i> = 4)	—
2.2	20	—	0.5		70	—
1.8	20	—	0.5		40	—
1.5	20	—	0.5		9	—

3.2.2 Desorption of MeHg/EtHg from the adsorbent 2 M hydrochloric acid and a mixture of 1 M hydrochloric acid and 2 M sodium chloride were tested to elute MeHg/EtHg adsorbed onto the adsorbent. Both eluents gave ($100 \pm 7\%$) recovery. A mixture of 1 M hydrochloric acid and 2 M sodium chloride was chosen for determining MeHg in the freshwater samples, because strong acid may

partly destroy SCF-adsorbent and leave unknown compounds which can be dissolved in benzene during solvent extraction and subsequently interfere with GC measurements. Besides, the addition of 2 M sodium chloride may increase the desorption of MeHg/EtHg from the adsorbent and decrease the emulsion formed in the extraction.

3.2.3 Extraction coefficient of MeHg/EtHg from the eluate The extraction efficiencies of 1 mL benzene extracting MeHgCl/EtHgCl from 3 mL and 6 mL of the mixture of 1 M HCl and 2 M NaCl were 0.74 ± 0.04 (20) and 0.64 ± 0.04 (17), respectively. Using 3 mL 2 M HCl the extraction efficiency was about 0.80.

The extraction procedure for MeHg/EtHg in natural water also generally consists of a back-extraction of MeHg/EtHg from the benzene by cysteine solution in order to eliminate the interference of some substances with the GC measurement. We found that back-extraction made no essential difference to the GC measurement compared with the extraction of MeHg/EtHg direct from eluate. The back-extraction was therefore omitted. The detection limit for MeHg/EtHg with the present method, including the extraction treatment, was about 0.04 ng L^{-1} using a 20 L water sample. The precision was about 20% and became lower when the concentration approached the detection limit. The main sources of error were: the uncertainty of GC analysis, the estimate of extraction efficiency, and the analytical concentrations of MeHg/EtHg in the stock water and benzene solution.

3.2.4 Amount of adsorbent for adsorption of MeHg/EtHg Yu and Liu (1981) reported that SCF-adsorbent contained 1–2% of the sulfhydryl group and the saturated capacity of methylmercury adsorption was 55 to 65 mg/g. The results of determining MeHg/EtHg in artificial water (Table III) have shown that 0.2 to 0.4 g adsorbent was enough to adsorb MeHg/EtHg from water at ppt concentration level. However, it was found that in the case of fresh water, the adsorption of MeHg was not complete in the first column and that the use of a second column was therefore necessary. The degree of adsorption on the first column depended on the composition of the fresh water and the pH used in the preconcentration. The other metal ions such as Zn^{2+} , Cd^{2+} , Pb^{2+} , Cu^{2+} , which are less strongly adsorbed on the SCF-adsorbent than MeHg is, may exist in natural

water at a concentration of about 10^{-5} to 10^{-7} mole L^{-1} , and they can also be adsorbed on the adsorbent at $pH > 3$. If the concentration of these ions is 10^{-6} mole L^{-1} , 10 L natural water will contain 0.01 to 0.04 mmole of such metal ions. The total amount of these ions will then be close to the saturated capacity of adsorption of 0.1–0.2 g SCF-adsorbent. The saturated capacity of adsorption of Zn^{2+} , Pb^{2+} , Cd^{2+} , etc., on the adsorbent was in general 0.15–0.18 meq/g adsorbent.⁷ Therefore, at $pH > 3$, these metal ions probably compete with MeHg for adsorption on the adsorbent.

3.3 Determination of MeHg in some freshwater samples and in snow

The analytical results of MeHg concentration in four freshwater samples under various pH conditions used in the preconcentration procedure are presented in Table IV. It can be seen from Table IV that the MeHg concentrations found in different freshwater samples were ranged from 0.05 to 0.26 $ng L^{-1}$ and the recoveries of MeHg were ranged from 39 to 74%.

Figure 3 shows a strong correlation between the recoveries of spiked MeHgCl in water samples and the value of color ($r^2 = 0.94$). The water samples from Lake Klintsjön with a very low content of humic substances (color 6 $mg L^{-1}$ Pt) had the highest recovery (68%), while water from Lake Skärsjön with a high content of humic substances (color 90 $mg L^{-1}$ Pt) had the lowest recovery (about 40%). Freshwater samples from the other two sites, which had the intermediate recovery (50 to 60%) had also a moderate color value (30 to 40 $mg L^{-1}$ Pt).

The main reason why the recovery of spiked MeHgCl was correlated with the concentration of humic substances may be that the adsorption of MeHg onto adsorbent depends on the competition of complexation ability between the humic substances and sulfhydryl group of SCF-adsorbent to MeHg. At constant pH, the complexation between MeHg and the humic substances was stronger at higher concentration of humic substances, and as a result less MeHg was adsorbed. Another explanation may be that there are two main fractions of MeHg in fresh water: that is weakly/moderately bound with humic substances and one that is tightly bound with humic substances (probably organic-sulfide MeHg). The MeHg concen-

Table IV Analytical results of methylmercury concentration in some fresh waters under various pH conditions used in the preconcentration procedure

Sample	MeHg (ng L ⁻¹)		Recovery (%)	Organic Hg _{adsorb.} ^a (organic Hg) _{total} (%)		Color ng L ⁻¹ Pt	pH in the Pt preconc.	Sampling data
	found ^a (±20%)	spiked found (±20%)		found (±20%)	(%)			
Lake Klimsjön	0.26	0.41	68	69	6	4.4	860417	
	0.19	0.41	63 (68)		6	3.6	860417	
		0.27	74		6			
Lake St. Skårsjön	0.10	0.41	39 (40)		90	4.5	860123	
		0.27	41					
	0.12	0.41	39 (43)	45	90	3.5	860123	
		0.27	48					
Lake Stora Övattnet	0.09	0.30	53			3.2	850923 ^b	
	0.05	0.30	53		32	6.9	850918 ^b	
	0.10	0.50	60			4.0	850422	
Ladebacksviken (run-off)	0.08	0.41	56		44	4.5	860513	
	0.11	0.36	69 (65)		32	3.5	851128	
		0.23	61					
	0.07	0.25	60	47		2.6	851212	

^aMeHg found at the condition without a spike of MeHgCl.^bThe lake was limed since this spring.^cAnalyzed by Dr A. Iverfeldt using the helium dc plasma AES method.

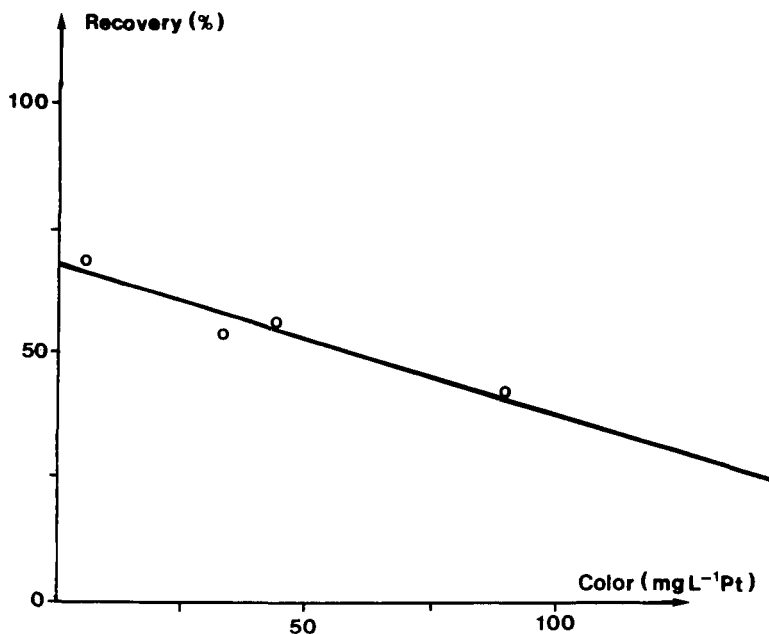


Figure 3 Relationship between recovery (%) of methylmercury and color in fresh water ($r^2=0.94$).

tration which was found with the present method may be the weakly/moderately bound MeHg fraction and correlated to the concentration of humic substances.

It can be seen from Table IV that at pH 2.5 to 4.5 no major influence on the recovery was found. However, the concentration of MeHg found in water from Stora Övattnet without a spike of MeHg at pH 6.9 was very low (0.05 ng L^{-1}) compared with that (0.09 ng L^{-1}) found at pH = 3.2. This may be related to the fact that the complexation between MeHg and the humic substances was stronger at neutral pH than that at low pH. Table IV also shows that the agreement between the values of recovery and the efficiency of SCF-adsorbent, $(\text{organic Hg})_{\text{adsorb}}/(\text{organic Hg})_{\text{total}}$ estimated using the AES method was good considering the two different methods used. It also suggests that the fraction of organic mercury which failed to be adsorbed on adsorbent may be tightly bound to the sulfhydryl group in the humic substances.

The mean of MeHg concentrations found in water samples without spike of MeHgCl and that corrected by comparison with the value of recovery are given in Table V together with the values of organic bound mercury. It can be seen from Table V that ca. one half of the total organic bound mercury in clear water from Lake Klintsjön was composed of MeHg, but in the waters containing moderate and high concentrations of humic substances, MeHg accounted for only 9 to 16 percent of the total dissolved organic mercury.

Comparing the mercury concentration in pike from Table II with the MeHg concentration in Lake Klintsjön and Lake St. Skärsjön from Table V it was found that the mercury concentration in pike was correlated, more closely, to the analytical concentration of MeHg in lake waters than to the MeHg concentration corrected by comparison with the value of recovery. An interesting finding is that the analytical concentration of MeHg found with the present method as suggested above may represent that of MeHg weakly/moderately bound with humic substances, and such a form would be easily available for biotic accumulation. The formation of more tightly bound MeHg as discussed above may involve complexation between MeHg and the sulfhydryl group in the humic substances, and this fraction of MeHg may not accumulate in fish so easily.

About 0.1 ng L^{-1} MeHg [$(\text{MeHg})_{\text{corr}} = 0.15 \text{ ng L}^{-1}$] was found in runoff water collected from the catchment area of Gårdsjön in Bohuslän, southwestern Sweden. It implies that the drainage water is a source of MeHg in receiving waters.

Table V Fraction of methylmercury concentrations found and corrected by comparison with the value of recovery in organic bound mercury in fresh water

Sample	$(\text{MeHg})_{\text{found}}$	$(\text{MeHg})_{\text{corr}}$	Org.Hg	$(\text{MeHg})_{\text{found}}$	$(\text{MeHg})_{\text{corr}}$
	ng L^{-1} ($\pm 20\%$)	ng L^{-1}	ng L^{-1}	Org.Hg (%)	Org.Hg (%)
Lake Klintsjön	0.22	0.32	0.68	32	47
Lake St. Skärsjön	0.11	0.27	3.14	3.5	8.6
Lake St. Övattnet	0.09	0.17	1.07	8.4	16
Ladebacksviken (run-off)	0.09	0.15	1.18	7.6	13

The MeHg concentrations in snow was 0.28 ng L^{-1} and the recovery was 79%.

An attempt was made to estimate the concentration of EtHg in fresh water. There was a small and poorly-formed peak in GC diagram from water samples of Lake Klintsjön and Lake St. Övattnet. Its retention time was 0.1 to 0.2 min. longer than that of EtHg from standard solution. Since ECD used in GC measurement was not selective, the finding of this peak provided no strong evidence of the presence of EtHg in water samples. Therefore, no estimated value of EtHg concentration will be reported.

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