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ORGANIC AND INORGANIC MERCURY IN THE FOOD CHAIN OF SOME LAKES AND RESERVOIRS IN FINLAND

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Contents of organic and inorganic mercury in food chain specimens, as well as sedimentation in two natural Finnish lakes and three impounded reservoirs, were studied. The proportion of organic mercury of total mercury varied in individual specimens from 32.7 to 100 %. Sedimentation (settleable solid) with very high contents of organic matter had ratios of organic to total mercury ranging from 2.4 to 87.3 %. These variations were similar in each of five water ecosystems studied. Benthic invertebrates had higher ratios of organic to total mercury than reported earlier. Total mercury concentrations in fish, zoobenthos and zooplankton of young impounded reservoirs were significantly higher than those of natural lakes. To explain this it is suggested that humic materials transfer mercury to the water and thence into the food chain.

Index words: Impounded reservoir, methyl mercury, total mercury, settleable solids, sedimentation, zooplankton, zoobenthos, fish, aquatic birds.

1. INTRODUCTION

Very recently a new environmental contamination problem has arisen, namely high concentrations of mercury in fish in young reservoirs. This phenomenon was first observed in the USA and Canada (Potter et al. 1975, Abernathy and Cumbie 1977, Cox et al. 1979, Meister et al. 1979, Bodaly and Hecky 1979) and soon also in Finland (Verta 1981, Lodenius et al. 1981, Alfthan et al. 1983). Lately, high mercury levels have also been found in fish in natural, unpolluted lakes with humic brown water (Hultberg and Hasselrot 1981, Björklund 1982, Paasivirta et al. 1983, Verta et al. 1986a). It was suggested

for fish in reservoirs that the higher the concentration of organic material in the water, the higher is the accumulation of mercury through the gills (Alfthan et al. 1983). People eating fish from young reservoirs had elevated mercury concentrations in their hair (Lodenius et al. 1981, Alfthan et al. 1983). Organic mercury in fish tissues is almost entirely in the form of monomethyl mercury (Huckabee et al. 1979), which is a very hazardous substance for all living things.

The percentage ratio of organic mercury to total mercury is generally very high in fish (Noren and Westöo 1967) but lower in benthic invertebrates (Jernelöv and Lann 1971). The variation of this ratio in zoobenthos between

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areas, species and individuals has been reported to be very great (Huckabee and Hildenbrand 1974, Cox et al. 1975, Hildenbrand et al. 1976, Trudel et al. 1977).

The goal of this study was to monitor the enrichment of mercury in food chain of reservoirs and natural lakes by analysing inorganic and organic mercury concentrations in food chain specimens from these areas. This study is part of a wider program, »The cycle of mercury in water environments», coordinated by the National Board of Waters and executed in the Water Research Institute of the National Board of Waters and in the universities of Jyväskylä, Helsinki and Kuopio.

2. MATERIALS AND METHODS

2.1 The study areas

The lakes and reservoirs sampled are illustrated in Figure 1. Pihlajavesi (L1) and Seinäjärvi (L2) are natural lakes. Kalajärvi (R1) is a reservoir filled in 1977, while Kyrkösjärvi (R2) was filled in 1981 and Porttipahta (R3) in 1970.

L2, R1 and R2 are situated in the same river basin. General and water quality characteristics of the lakes and reservoirs are presented in Table 1.

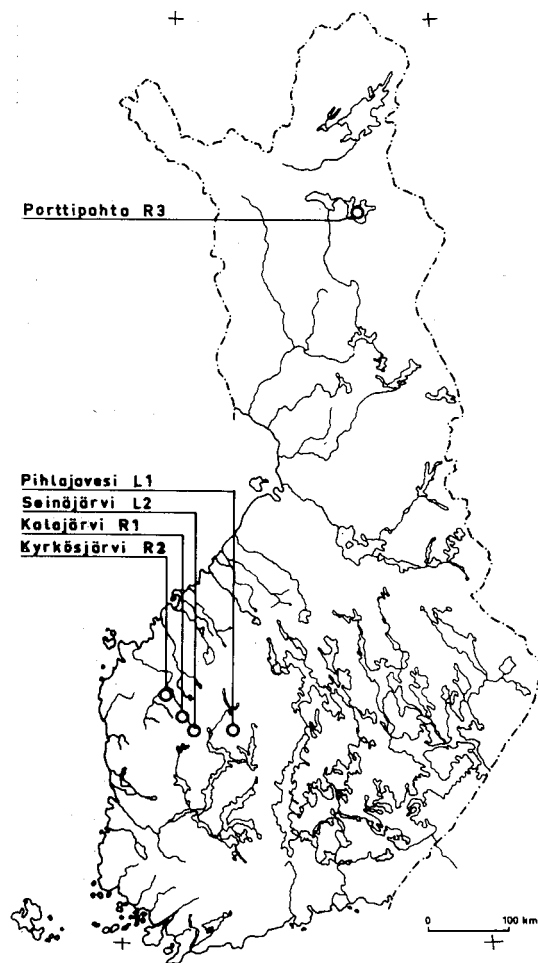


Fig. 1. The study areas.

Table 1. General and water quality characteristics of the study lakes (L1, L2) and reservoirs (R1–R3).

Parameter	L1 Pihlajavesi	L2 Seinäjärvi	R1 Kalajärvi	R2 Kyrkösjärvi	R3 Porttipahta
Year of filling	—	—	1977	1981	1970
Area km ²	19.9	8.5	11.3	6.4	214
Volume Mm ³	103	11	43	16	1350
Drainage area km ²	370	97	500	820	2460
Mean depth m	5.2	1.3	3.8	2.5	6.3
Maximum amplitude of regulation m	—	1	6	2	11
Conductivity mS m ⁻¹	3.4	3.4	3.8	4.0	5.7
Alkalinity mmol dm ⁻³	0.05	0.04	0.03	0.24	0.16
pH	6.1	5.9	5.7	6.7	5.8
Colour Pt mg dm ⁻³	110	190	200	130	330
COD (Mn) mg dm ⁻³	15	18	28	11	39
Total P μg dm ⁻³	16	22	53	38	190
Total Fe μg dm ⁻³	660	1300	1500	2100	4300
Total Ca μg dm ⁻³	2400	1900	2300	2600	3200

2.2 Sampling methods

Samples were collected during the years 1980–1984. The fish (pike, *Esox Lucius L.*; perch, *Perca fluviatilis L.*; roach, *Rutilus rutilus L.*) were caught with fykes and nets and frozen immediately in aluminium foil and stored at -20°C . Fresh tissues were used for the analyses. Tissue samples were taken under the last dorsal fin immediately above the horizontal septum and a few millimeter below the skin.

The sampling of zoobenthos was performed manually from littoral areas. Most samples consisted of trichoptera larvae but some dragonfly larvae were also collected. The samples were kept in water for 4–10 hours before removing the larvae and freezing. Samples were freeze-dried under reduced pressure before analysing.

The sampling of zooplankton was carried out mainly from littoral areas using $400\ \mu\text{m}$ mesh plankton nets. In addition some samples were collected from pelagial areas. Some of the samples were frozen immediately in the field with solid carbon dioxide (-79°C). Zooplankton samples were freeze-dried under reduced pressure.

Settleable solids (sedimentation samples) were collected in 5 cm (diameter) glass funnels which were placed in different sites in the lake about one meter above the bottom. The funnels were emptied at two-month intervals and samples were collected through out the year. In addition to settleable solids the funnels also gathered some zooplankton of large size. For the analyses the samples were centrifuged and air-dried.

Sediment samples were taken by crust-freeze sampling (Renberg 1981) from lakes depths. The sediment columns were cut into 1–2 cm layers and air-dried to constant weight.

Aquatic birds were shot and frozen in aluminium foil. Fresh tissues of the chest muscles were used for the analyses. The eggs of goldeneyes (*Bucephala clangula L.*) were collected from several nests and frozen and freeze-dried before determination of mercury. In the case of eggs of which the yolk and white could be separated, these were analysed separately. The shells of the eggs were not analysed.

2.3 Analysis methods

The analyses of mercury were performed with a Perkin Elmer Coleman MAS-50 mercury ana-

lyzer. The reduction vessel was a $100\ \text{cm}^3$ gas washing bottle and the carrier gas was nitrogen with a flow rate of $1.2\ \text{dm}^3\ \text{min}^{-1}$. The gas was dried with sulfuric acid. Peak heights were recorded with a Kipp and Zonen recorder.

Using this method for the determination it is possible to analyse inorganic and organic mercury from the same sample. The NaOH digestion procedure employed was a modification of that used by Oda and Ingle (1981). Magos (1971) reagents were used for the reduction of mercury compounds.

Reagents:

- Sodium hydroxide, NaOH, Merck p.a., 45% (w/v) aqueous solution.
- Nitric acid, HNO_3 , Merck p.a., 69.6%.
- Sodium chloride, NaCl, Merck p.a., 1% (w/v) aqueous solution.
- The reductant, 10% (w/v) tin (II) chloride (SnCl_2 , Merck p.a.), 2% (w/v) cadmium (II) chloride (CdCl_2 , Merck p.a.) and 5% sulfuric acid (H_2SO_4) in an aqueous solution. The bubbling of the reductant removed mercury that may have been present.
- n-Octanol, $\text{CH}_3(\text{CH}_2)_7\text{OH}$, Merck p.a., anti-foam.
- Organic mercury standard: about 310 mg of methyl mercury (II) chloride (CH_3HgCl , Merck z.a.) was dissolved in $25\ \text{cm}^3$ of ethanol and further dilution to $250\ \text{cm}^3$ was made with deionized water.
- Inorganic mercury standard: Coleman's 1000 mg Hg dm^{-3} mercury standard was diluted to $1\ \text{mg Hg dm}^{-3}$ with water.

Procedure:

A portion of 0.3–0.4 g wet weight or 0.1–0.2 g dry weight of the sample was carefully weighed in a $50\ \text{cm}^3$ test-tube and $3\ \text{cm}^3$ of NaOH was added. The test-tube was tightly capped and heated in boiling water for 15–30 min. After cooling $3\ \text{cm}^3$ of NaCl and $8\ \text{cm}^3$ of nitric acid were added. The solution was then cooled for at least one hour (or overnight), transferred to the reduction vessel and diluted to $30\ \text{cm}^3$ with deionized water. Two drops of n-octanol and $2\ \text{cm}^3$ of the reductant were added. The reduction vessel was then immediately connected to the gas flow and the maximum absorbance, due to inorganic mercury (+ phenyl mercury, Campe et al. 1978, Campe et al. 1982), was recorded. After the peak had returned to the base line $9\ \text{cm}^3$ of NaOH was added and again the maximum ab-

sorbance, now due to organic mercury, was recorded. Serial amounts of the standard solutions and the reagent blank were analysed in the same way.

This method for the determination of mercury species (SEP-ALK) was compared with the method of the Research Laboratory of the National Board of Waters, in which a commonly accepted method for the determination of total mercury (TOT-ACD), involving acid digestion, oxidation with potassium permanganate and reduction with tin(II)chloride, is used. Seventy-seven fish samples were analysed with these two methods (Fig. 2.) and the results of total mercury were compared using the paired t-test (Tables 2 and 3).

According to the t-test the results obtained with the TOT-ACD method were significantly higher than those obtained with the SEP-ALK method.

Earlier, Kacprzach and Chvojka (1976) carried out a similar study in which the selective method yielded the same results as the »permanganate» method when the content of mercury was below

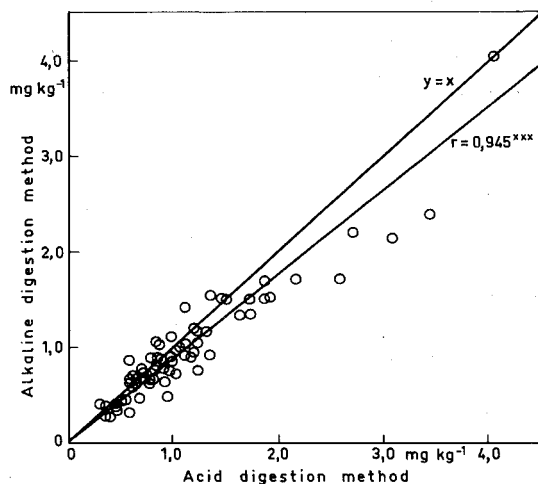


Fig. 2. Results of the determination of total mercury by the TOT-ACD (acid digestion) and SEP-ALK (alkaline digestion) methods.

Table 2. Statistical analysis of the results of total mercury obtained with the SEP-ACD methods.

	SEP-ALK	TOT-ACD
Mean (\bar{x}), mg kg ⁻¹	0.992	1.095
Standard deviation (s), mg kg ⁻¹	0.571	0.686
Number of cases (N)	77	77
t-Test		p < 0.001
Mean of the differences		0.10
Standard deviation of the differences		0.24
Regression line	TOT-ACD = 1.137 SEP-ALK - 0.0325	
Correlation coefficient	r = 0.945	

Table 3. Statistical analysis of the results below 1 mg kg⁻¹ obtained with the SEP-ALK and TOT-ACD methods.

	SEP-ALK	TOT-ACD
Mean (\bar{x}), mg kg ⁻¹	0.651	0.679
Standard deviation (s), mg kg ⁻¹	0.187	0.187
Number of cases (N)	41	41
t-Test		p = 0.157
Mean of the differences		0.028
Standard deviation of the differences		0.13

1 mg kg⁻¹. The differences between the results obtained in the present study were also found to be insignificant in the case of results below 1 mg kg⁻¹ (Table 3).

the different areas were found only in fish and aquatic birds. The contents of mercury increased considerably when ascending the food chain in all the study areas.

3. RESULTS AND DISCUSSION

3.1 Mercury concentrations in the study areas

The highest mercury concentrations in most organisms were found in the Kalajärvi and Kyrkösjärvi reservoirs (Fig. 3.) and the lowest concentrations in lake Seinäjärvi. However, remarkable differences between mercury concentrations in

Aquatic birds

The highest level of total mercury was always found in fish-eating birds, the maximum concentration being 1.95 mg kg⁻¹ in a black-throated diver (*Gavia arctica* L.) found dead in lake Pihlajavesi.

Goldeneyes had lower mercury levels than the fish-eating birds, as was found in the mercury-polluted Lake Päijänne (Särkkä et al. 1978c, Paasivirta et al. 1981b). As a rule mercury concentrations in the eggs of the goldeneyes were higher (Fig. 3.) than those in the muscle of juvenile or adult goldeneyes (Table 4.), indicating that fe-

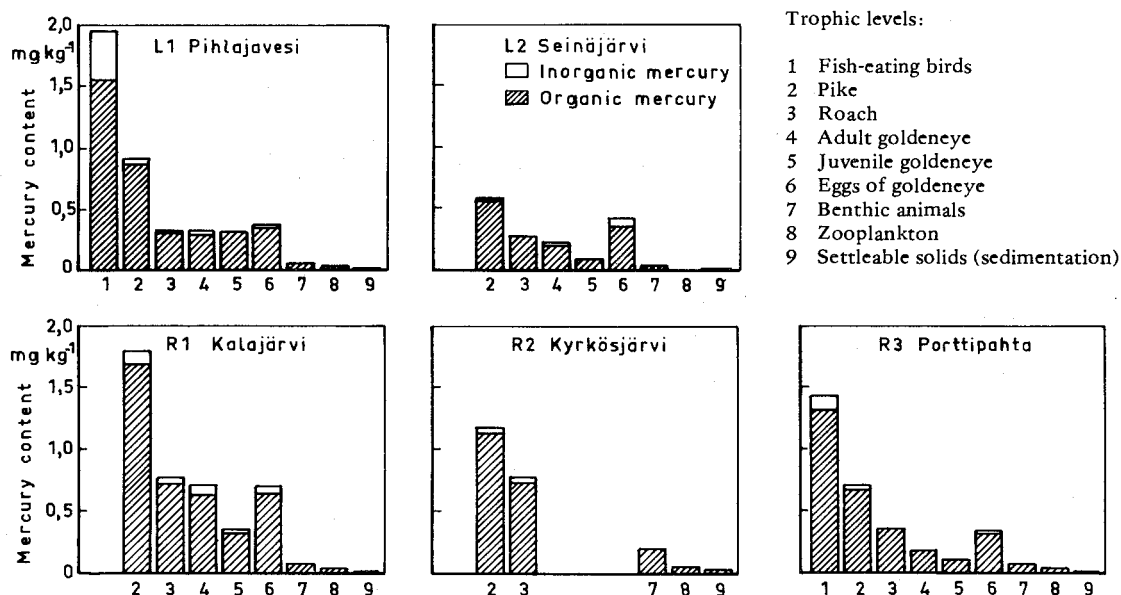


Fig. 3. Inorganic and organic mercury concentrations (mg kg⁻¹ fresh weight) in different trophic levels at five study areas.

Table 4. Number of cases (N), mean values (\bar{x}) and range of total mercury and organic mercury contents (mg kg⁻¹) in fresh tissue of chest muscle of juvenile and adult goldeneyes from four areas.

Area	Total mercury (mg kg ⁻¹)			Organic mercury (mg kg ⁻¹)	
	N	\bar{x}	range	\bar{x}	range
L1 Pihlajavesi	6	0.33	0.19–0.52	0.30	0.18–0.42
L2 Seinäjärvi	8	0.14	0.01–0.31	0.13	0.01–0.30
R1 Kalajärvi	10	0.43	0.15–1.06	0.38	0.14–0.95
R3 Porttipahta	5	0.11	0.08–0.17	0.10	0.08–0.16

male birds excrete large quantities of mercury into the eggs.

If it is assumed that the mercury content of the muscle represents the average mercury concentration in the bird, the overall amount of mercury in goldeneyes can be calculated. The results of this calculation clearly indicate that the amount of mercury is considerably greater in juveniles in those study areas (Kalajärvi and lake Pihlajavesi) where the concentrations in adult goldeneyes are also highest (Fig. 4.). Thus, the method of total amount comparison (Paasivirta et al. 1981b) proved to be applicable to the comparison of regional differences in pollution. The mercury content in zoobenthos (trichoptera) was also significantly higher in these study areas indicating (Fig. 3) that dietary mercury intake by goldeneyes has a significant role in these areas.

Fish

The concentration of mercury in both roach and pike was considerably higher in the two young reservoirs than in the other study areas (Tables 5 and 6), exceeding 1.0 mg kg^{-1} in pike, which is the limit for edible fish laid down by the National Board of Health in Finland. In the non-regulated natural lake Pihlajavesi the mean mercury concentration of pike (0.92 mg kg^{-1} , wet weight) was among the highest in comparison with other unpolluted Finnish lakes (Paasivirta et al. 1983, Verta et al. 1986a).

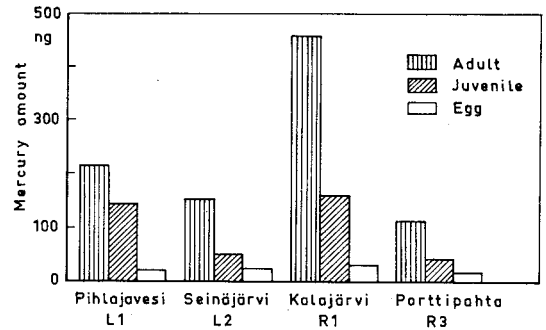


Fig. 4. The overall amount of mercury (ng) in adults (650 g), juveniles (500 g) and eggs (50 g) of goldeneyes from four areas.

Zoobenthos

Total mercury concentrations in zoobenthos ranged between 0.01 and 0.47 mg kg^{-1} (wet weight) in reservoirs and between 0.003 and 0.16 mg kg^{-1} in natural lakes (Table 7). The corresponding values on a dry weight basis were 0.07 – 1.6 mg kg^{-1} and 0.02 – 0.62 mg kg^{-1} , respectively. The values from natural lakes were in good agreement with earlier reports from uncontaminated environments (Johnels et al. 1967, Hasselrot and Göthberg 1974, Huckabee and Hildenbrand 1974, Grahn et al. 1976, Göthberg 1983).

Table 5. Number of cases (N), mean values (\bar{x}) and range of total mercury and organic mercury contents (mg kg^{-1}) in fresh muscle of pike from five areas.

Area	Total mercury (mg kg^{-1})			Organic mercury (mg kg^{-1})	
	N	\bar{x}	range	\bar{x}	range
L1 Pihlajavesi	35	0.92	0.50–1.74	0.87	0.47–1.59
L2 Seinäjärvi	47	0.60	0.23–1.51	0.58	0.23–1.38
R1 Kalajärvi	70	1.80	0.85–4.06	1.69	0.68–3.90
R2 Kyrkösjärvi	17	1.19	0.63–1.85	1.14	0.59–1.81
R3 Porttipahta	46	0.70	0.30–1.71	0.67	0.25–1.63

Table 6. Number of cases (N), mean values (\bar{x}) and range of total mercury and organic mercury contents (mg kg^{-1}) in fresh muscle of roach from five areas.

Area	Total mercury (mg kg^{-1})			Organic mercury (mg kg^{-1})	
	N	\bar{x}	range	\bar{x}	range
L1 Pihlajavesi	11	0.33	0.22–0.41	0.31	0.20–0.40
L2 Seinäjärvi	15	0.29	0.16–0.40	0.29	0.15–0.39
R1 Kalajärvi	13	0.77	0.43–1.17	0.72	0.41–1.15
R2 Kyrkösjärvi	7	0.78	0.63–1.20	0.74	0.62–1.06
R3 Porttipahta	5	0.35	0.24–0.49	0.34	0.23–0.47

Concentrations in young reservoirs were, however, significantly higher than those in other study areas and the mean and maximum values exceeded all those reported in uncontaminated natural areas.

Using the same analytical method Verta et al. (1986a) reported for total mercury a range of 0.04–0.76 mg kg⁻¹ (dry weight) which corresponds 0.009–0.114 mg kg⁻¹ on a wet weight basis in zoobenthos of Finnish forest lakes.

Zooplankton

Total mercury concentrations in zooplankton ranged from 0.05 to 0.73 mg kg⁻¹ (dry weight) in reservoirs and from 0.11 to 0.55 mg kg⁻¹ (dry weight) in natural lakes (Table 8). The corresponding values on a wet weight basis were 0.002–0.135 mg kg⁻¹ and 0.001–0.154 mg kg⁻¹, respectively. Only a slight difference was found between young reservoirs and the other study areas in 1982–1983 although very high concentrations (max. 2.1 mg kg⁻¹, dry weight) were found in the youngest reservoir in 1981 (Verta et al. 1986b).

Reported values from uncontaminated small forest lakes are rare. Mercury concentrations ranged from 0.03 to 0.18 mg kg⁻¹ (dry weight) total mercury in one study (Grahn et al. 1976, Cajander 1980) and were less than 0.25 mg kg⁻¹ (dry weight) methyl mercury in another (Hultberg and Hasselrot 1981). Verta et al. (1986a)

found that the concentrations in zooplankton ranged from 0.02 to 0.55 mg kg⁻¹ (dry weight) in Finnish forest lakes.

It seems possible that impounded reservoirs and brown-water lakes have higher concentrations of mercury in zooplankton than clear-water lakes since the lakes in the present study are considerably more humic than those studied by Grahn et al. (1976), Cajander (1980) and Hultberg and Hasselrot (1981). Hultberg and Hasselrot (1981) also found that water in Swedish »high level lakes», in which the concentrations of methyl mercury in zooplankton and fish were higher than in other lakes with the same pH, usually had a high colour value.

Settleable solids

No differences were found between the mercury concentrations of settleable solids (sedimentation samples) in the study areas (Table 9). Mercury content did not correlate with the concentration of organic matter. The mean organic matter concentration (ignition loss) was 41.1%.

3.2 Correlation of mercury content of pike with weight, length and age

Both inorganic and organic mercury concentrations in muscle of pike correlated positively

Table 7. Number of cases (N), mean values (\bar{x}) and range of total mercury and organic mercury contents (mg kg⁻¹) in zoobenthos (wet weight) from five areas.

Area	Total mercury (mg kg ⁻¹)			Organic mercury (mg kg ⁻¹)	
	N	\bar{x}	range	\bar{x}	range
L1 Pihlajavesi	13	0.055	0.008–0.155	0.045	0.007–0.138
L2 Seinäjärvi	26	0.045	0.003–0.112	0.040	0.002–0.089
R1 Kalajärvi	22	0.083	0.020–0.231	0.078	0.010–0.221
R2 Kyrkösjärvi	12	0.205	0.022–0.472	0.186	0.021–0.446
R3 Porttipahta	24	0.056	0.010–0.139	0.047	0.007–0.106

Table 8. Number of cases (N), mean values (\bar{x}) and range of total mercury and organic mercury contents (mg kg⁻¹) in zooplankton (dry weight) from five areas.

Area	Total mercury (mg kg ⁻¹)			Organic mercury (mg kg ⁻¹)	
	N	\bar{x}	range	\bar{x}	range
L1 Pihlajavesi	9	0.22	0.12–0.39	0.19	0.11–0.33
L2 Seinäjärvi	8	0.28	0.11–0.55	0.22	0.09–0.49
R1 Kalajärvi	8	0.30	0.13–0.73	0.26	0.12–0.62
R2 Kyrkösjärvi	7	0.42	0.19–0.65	0.38	0.19–0.63
R3 Porttipahta	26	0.24	0.05–0.43	0.18	0.04–0.41

Table 9. Number of cases (N), mean values (\bar{x}) and range of total mercury and organic mercury contents (mg kg^{-1}) in settleable solids (dry weight) from five areas.

Area	Total mercury (mg kg^{-1})			Organic mercury (mg kg^{-1})	
	N	\bar{x}	range	\bar{x}	range
L1 Pihlajavesi	14	0.30	0.11–0.49	0.14	0.01–0.40
L2 Seinäjoki	13	0.22	0.12–0.37	0.10	0.04–0.16
R1 Kalajärvi	12	0.18	0.06–0.38	0.09	0.04–0.23
R2 Kyrkösjärvi	1	0.42		0.21	
R3 Porttipahta	5	0.23	0.17–0.28	0.16	0.14–0.19

with body weight of fish in all the areas except the Kyrkösjärvi reservoir (Fig. 5). The Kyrkösjärvi reservoir is very young and the mercury concentrations in fish are rapidly increasing (Fig. 6). The variation of weight was very limited in the samples from Kyrkösjärvi. In other areas the correlation coefficients of mercury content with age and length were of the same magnitude as those with weight. Several authors have reported similar positive size-age relationships for total mercury concentration in predatory fish (Johnels et al. 1967, Scott and Armstrong 1972, Kelly et al. 1975, Annett et al. 1975, Paasivirta et al. 1981a).

3.3 Proportion of organic mercury of total mercury

The proportion of organic mercury of total mercury had a range of 72.7–98.0% in individual birds, 75.5–100% in fish, 35.6–100% in zoobenthos, 45.1–100% in zooplankton and 2.4–87.3% in settleable solid samples. The mean proportions for different materials were:

fish	95.1%
birds	90.8%
goldeneye eggs	86.4%
zoobenthos	86.0%
zooplankton	79.9%
settleable solids	51.3%

The organic mercury in fish and in other biological material can be assumed to be present almost entirely as monomethyl mercury, since no other organic mercury compound has been reported (Huckabee et al. 1979). The methyl mercury fraction measured in fish is of the same magnitude as those reported earlier (Noren and Westö 1967, Bache et al. 1971, Zitko et al. 1971, Kamps et al. 1972, Lockhart et al. 1972, Hucka-

bee et al. 1974, Kacprzak and Chovjak 1976, Manthey and Berge 1980). A considerably lower fraction of methyl mercury in zoobenthos has been reported, although the range was 0–100% (Jernelöv and Lann 1971, Huckabee and Hildenbrand 1974, Cox et al. 1975, Trudel et al. 1977). The great variation of methyl mercury in zooben-

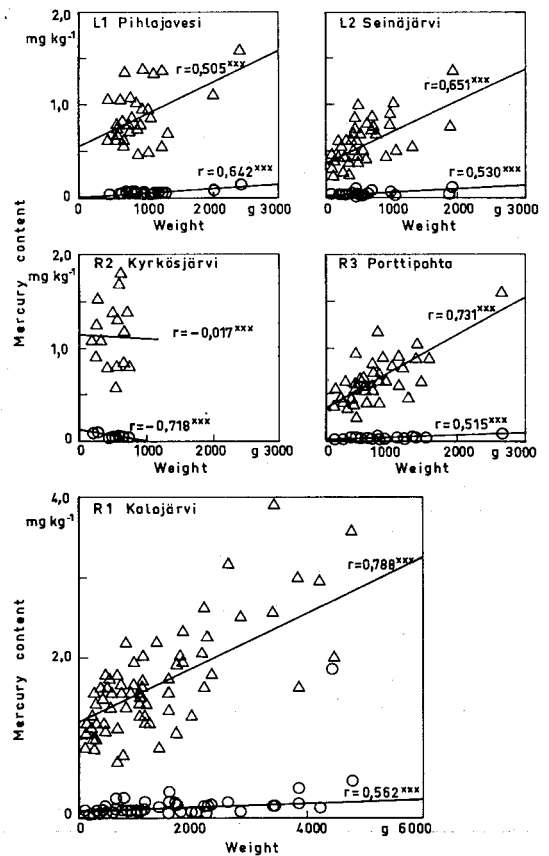


Fig. 5. Relations of organic (Δ) and inorganic (O) mercury concentrations (mg kg^{-1}) to body weight of fish (g) for individual samples of pike.

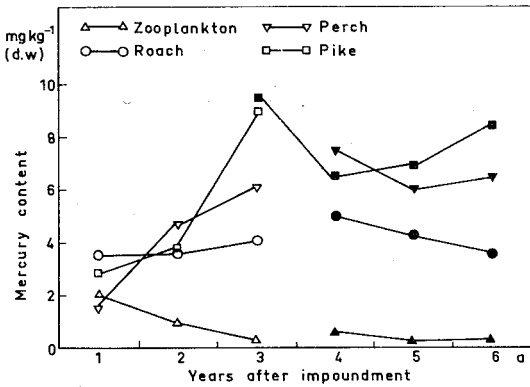


Fig. 6. Total mercury contents in the Kyrkösjärvi (R2; light spots) and Kalajärvi (R1; dark spots) reservoirs after impoundment.

thos may be explained both by the different species and classes of zoobenthos studied and by analytical differences.

No statistical differences in the methyl mercury fraction were found between the study areas, with the exception of the Kyrkösjärvi reservoir, which had somewhat higher methyl mercury fractions in zooplankton than did the other study areas.

3.4 The effect of the soil and humic material on mercury concentrations

Methylated mercury is known to be produced in the sediment and water phase of aquatic environments (Westö 1966, Jensen and Jernelöv 1969, Jernelöv 1970) and in soils (Van Faassen 1970, Yamada and Tonomura 1972). Humic substances have been found to methylate mercury abiotically (Rogers 1977, Nagase et al. 1982, Nagase et al. 1984). Furthermore, it has recently been reported that in polyhumic waters up to more than 90% of the total carbon in zooplankton may be allochthonous (Salonen and Hammar 1984).

In Kyrkösjärvi very high mercury concentrations in zooplankton (max. 2.1 mg kg⁻¹, dry weight, Verta et al. 1986b) were observed during the first summer after impoundment (Fig. 6). The measured concentration is five times higher than the mean concentration found in the same area two years later, and nearly ten times that found in other areas in the present study and

likewise tenfold compared to the mean concentration in a study of 35 Finnish forest lakes (Verta et al. 1986a).

Higher mercury contents in zoobenthos of the young reservoirs than in the other study areas is evident, although the difference is not as clear as in the case of zooplankton. However, it seems possible that immediately after impoundment large amounts of already methylated mercury, or mercury that can very easily be methylated are dissolved to the water phase and accumulated particularly into zooplankton.

It should be noted that during the first years of impoundment very low oxygen concentrations (down to 0%) and very dark colour (up to 1000 Pt mg dm⁻³) were recorded in the water of the Kyrkösjärvi reservoir. In the second winter after impoundment a mercury concentration of 40 ng dm⁻³ was measured in an oxygen free water sample 1 m above the bottom of the Kyrkösjärvi reservoir (Verta et al. 1986b), which also indicates the dissolving of mercury from the immersed soils.

The mercury concentrations in pike in the study areas cannot be explained by the mercury concentration in sediments, water pH and trophic level as Håkanson (1980) has proposed (Table 10). The observed values for 1 kg pike exceed those calculated with the formula of Håkanson (1980) by a factor of 5–8 in impounded reservoirs and by a factor of 1.5–2 in natural lakes.

Only one thorough study of mercury contents in food chains has carried out earlier in Finland (Hattula et al. 1978, Särkkä et al. 1978 a, b, c). When comparing the results of the present study with those in lake Päijänne (Table 11), a clear difference in the accumulation pattern of mercury was observed. The mercury content in biota were of the same level or even higher in the young reservoirs than in lake Päijänne, although the concentrations in the flooded land were significantly lower than in the sediments of lake Päijänne. These data further indicate that methylation or accumulation of mercury is more effective in lakes and impoundments containing large quantities of humic material than in clear-water lakes or lakes with inorganic mercury pollution.

3.5 Development of mercury content in reservoirs

In the Kyrkösjärvi reservoir the maximum concentration of mercury in zooplankton was

Table 10. Comparison of the mercury concentrations found by determination of methyl mercury and by calculation using the formula of Håkanson (1980) in pike from five study areas.

F(Hg) = the content of methyl mercury in 1 kg pike in mg kg^{-1} wet weight.

Hg50 = the weighted mean mercury content of surface sediments, 0–1 cm, in ng kg^{-1} dry weight.

pH = the mean pH of the water system.

BPI = the bioproduction index.

Area	Hg50 ng kg^{-1}	pH	BPI	F(Hg) calculated mg kg^{-1}	F(Hg) observed mg kg^{-1}
L1 Pihlajavesi	220	6.1	4.2	0.61	0.95
L2 Seinäjärvi	100	5.6	5.2	0.33	0.71
R1 Kalajärvi	130	5.6	6.3	0.36	1.61
R2 Kyrkösjärvi	70	5.8	8	0.24	1.79
R3 Porttipahta	50	6.7	5.2	0.14	0.76

Table 11. Comparison of mercury concentrations of young reservoirs (Kyrkösjärvi and Kalajärvi), natural lakes (Pihlajavesi and Seinäjärvi), and the mercury-polluted lake Päijänne. (Särkkä et al. 1978 a,b). (dw = dry weight, ww = wet weight).

Trophic level	Mercury concentration (mg kg^{-1})		
	Lake Päijänne	Young reservoirs	Natural lakes
Sediment (dw)	0.31	0.10	0.20
Zoobenthos (ww)	0.08	0.14	0.05
Zooplankton (ww)	0.01	0.04	0.04
Roach (ww)	0.50	0.78	0.31
Pike (ww)	1.51	1.50	0.76
Goldeneye (ww)	0.24	0.43	0.29

reached during the first summer after impoundment. After this, the concentrations decreased smoothly (Fig. 6). In fish the maximum concentrations are probably reached within 3–5 years of impoundment, after which the concentrations might be expected to decrease slightly because of the smaller load of mercury from the soil. However, in the Kalajärvi reservoir the mercury concentration in pike again increased 5–6 years from impoundment (Fig. 6). This may be a consequence of the fact that the pikes are forced to change their main food source from invertebrates to fish. In many reservoirs it has been found that pikes use zoobenthos as the main food source at least for part of the year (Bodaly and Lesack 1984, Koskenniemi 1984 pers. comm.). According to several studies the maximum of the total biomass of zoobenthos is reached within 1–5 years of impoundment (Nursall 1952, Petr 1969, 1972, Jankovic 1972, Paterson and Fernando 1970, Armitage 1977).

4. CONCLUSION

The analysis results are consistent with the assumption that humic substances transfer mercury from soil/sediment to the water phase and further to biota mainly in the form of organic mercury.

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TIIVISTELMÄ

Elohopean rikastumista ravintoketjuun tutkittiin kolmessa tekojärvässä ja kahdessa luonnonjärvässä vuosina 1980–1984 analysoimalla epäorgaanisen ja orgaanisen (metyyli) elohopean pitoisuuksia ravintoketjun eri osissa. Tutkitut trofiatasot olivat sedimentoituva kiintoaines, eläinplankton, pohjaeläimet, telkän munat, telkän poikanen, aikainen telkkä, särki, hauki ja kaloja syövät linnut.

Nuorissa tekojärvissä (Kalajärvi ja Kyrkösjärvi) todettiin yleensä kaikilla trofiatasoilla suurimmat elohopeapitoisuudet kuin luonnonjärvissä (Seinäjärvi, Pihlajavesi) tai tutkitussa vanhassa tekojärvässä (Porttipahta). Sedimentoituvassa kiintoaineessa ei havaittu eroja eri alueiden välillä.

Sekä epäorgaaninen että metyylielohopea koreloivat yleensä positiivisesti hauen painon ja pituuden kanssa. Poikkeuksena oli nuorin Kyrkösjärven tekojärvi, jossa pitoisuudet kalastossa olivat nousussa tutkimusjakson aikana.

Metyylielohopean osuus kokonaiselohopeasta oli keskimäärin kaloissa 95,1%, linnuissa 90,8%, lintujen munissa 86,4%, pohjaeläimissä 86,0%, eläinplanktonissa 79,9% ja laskeutuvassa kiintoaineessa 51,3%. Metyylielohopean prosenttiosuudessa ei esiintynyt merkittäviä eroja eri tutkimusalueiden välillä.

Elohopeapitoisuuksien kehittyminen nuorten tekojärvien eliöstössä ja vedessä viittasivat siihen, että välittömästi tekojärven rakentamisen jälkeen suuri määrä jo metyloitunutta tai nopeasti metyloituvaa elohopeaa liukenee maaperästä veteen ja rikastuu ensi vaiheessa erityisesti eläinplanktoniin.

Vertailu kirjallisuudessa esitettyyn malliin ja Päijänteen eliöstön elohopeapitoisuuksiin viittasi siihen, että elohopean metyloityminen tai rikastuminen on tehokasta nuorissa tekojärvissä. Syyksi esitettiin humusyhdisteiden toimimista elohopean välittäjinä maaperästä veteen ja eliöstöön.

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