

- Seppo Rekolainen, Matti Verta & Olli Järvinen: Mercury in snow cover and rainfall in Finland 1983–1984**
Tiivistelmä: Sadeveden ja lumen elohopeapitoisuus Suomessa 1983–1984 3
- Seppo Rekolainen, Matti Verta & Anita Liehu: The effect of airborne mercury and peatland drainage on sediment mercury contents in some Finnish forest lakes**
Tiivistelmä: Ilmaveintäisen elohopean ja metsäojituksen vaikutus sedimentin elohopeapitoisuteen eräissä Suomen metsäjärvisä 11
- Matti Verta, Seppo Rekolainen, Jaakko Mannio & Kari Surma-Aho: The origin and level of mercury in Finnish forest lakes**
Tiivistelmä: Elohopean alkuperä ja pitoisuustaso Suomen metsäjärvisä 21
- Jaakko Mannio, Matti Verta, Pirkko Kortelainen & Seppo Rekolainen: The effect of water quality on the mercury concentration of northern pike (*Esox lucius*, L.) in Finnish forest lakes and reservoirs**
Tiivistelmä: Veden laadun vaikutus hauen elohopeapitoisuteen Suomen metsäjärvisä ja tekoaltaissa 32
- Matti Verta, Seppo Rekolainen & Kari Kinnunen: Causes of increased fish mercury levels in Finnish reservoirs**
Tiivistelmä: Kohonneiden elohopeapitoisuuksien syyt Suomen tekoaltaissa 44
- Kari Surma-Aho, Jaakko Paasivirta, Seppo Rekolainen & Matti Verta: Organic and inorganic mercury in the food chain of some lakes and reservoirs in Finland**
Tiivistelmä: Orgaaninen ja epäorgaaninen elohopea eräiden Suomen järvien ja tekoaltaiden ravintoketjuissa 59
- Jari Leskinen, Ossi V. Lindqvist, Jari Lehto & Pekka Koivistoinen: Selenium and mercury contents in northern pike (*Esox lucius*, L.) of Finnish man-made and natural lakes**
Tiivistelmä: Seleenin ja elohopean pitoisuus Suomen tekoaltaiden ja luonnonjärvien hauissa 72
- Vappu Pennanen, Pirkko Kortelainen & Jaakko Mannio: Comparative study on the estimation of humic matter in natural waters**
Tiivistelmä: Luonnonvesien humuspitoisuuden arviointi eri menetelmillä 80
- Pirkko Kortelainen, Jaakko Mannio & Vappu Pennanen: Characteristics of the allochthonous organic matter in Finnish forest lakes and reservoirs**
Tiivistelmä: Alloktonisen orgaanisen aineen ominaisuuksista suomalaisissa metsäjärvisä ja tekoaltaissa 88
- Tom Frisk & Vappu Pennanen: A steady-state model for two humic fractions**
Tiivistelmä: Kahden humusfraktion tasapainotilan malli 98

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THE EFFECT OF AIRBORNE MERCURY AND PEATLAND DRAINAGE ON SEDIMENT MERCURY CONTENTS IN SOME FINNISH FOREST LAKES

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Mercury contents were analysed in sediment cores taken from 13 lakes in southern, central and northern Finland. In lakes in southern Finland with no drainage operations being performed in their catchment areas, a distinct increase in mercury concentrations was observed in recent sediments. In northern Finland only a slight or zero increase was recorded. According to Pb-210 dating the most remarkable increase began in the years 1920-1930. The increase was concluded to be mainly a result of increased atmospheric mercury deposition. The sediment mercury content was found to correlate significantly with the organic matter content of the sediment and with the pH of the water. Intensive peatland drainage of the catchment area was concluded to have caused the decrease in mercury content in the topmost sediment layers in some lakes. This was probably due to the higher content of leached allochthonous material of the total sedimentating material. The mercury accumulation rate was calculated to range from 25 to 50 $\mu\text{g m}^{-2} \text{a}^{-1}$ in lakes with undisturbed or only slightly disturbed runoff areas and to reach 370 $\mu\text{g m}^{-2} \text{a}^{-1}$ in a lake affected by intensive drainage operations.

Index words: Mercury, sediment, organic matter, atmospheric deposition, peatland drainage.

1. INTRODUCTION

Lake sediments can be regarded as a function of long-term changes in several ecological variables. Alterations in allochthonous and autochthonous inputs and changes resulting from human activities can be seen in sediment profiles:

Mercury reaches lake waters and lake sediments by direct atmospheric deposition, by leaching from drainage areas and also in waste waters. The concentration of mercury has been reported to

increase towards surface layers in sediments even in lakes with no known direct mercury pollution (e.g. Vernet and Thomas 1972, Björklund and Norling 1979, Johansson 1980, 1985, Björklund et al. 1982, Simola and Lodenius 1982, Ouellet and Jones 1983, Tolonen and Jaakkola 1983). The increased anthropogenic input and atmospheric deposition of mercury, and also human activities which have increased the leaching of mercury from drainage areas, have been postulated as possible causes.

The beginning of the distinct increase in the

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mercury contents of sediments has usually been estimated at about 1910-1940 in northern Europe and Canada (Johansson 1980, 1985, Björklund et al. 1982, Ouellet and Jones 1983), but slight increases have also been observed in earlier sediments (Aston et al. 1973).

In this work the mercury concentrations in recent sediments in some Finnish forest lakes are reported. In particular the effects of atmospheric mercury fallout and of human activities in drainage basins are discussed.

2. MATERIALS AND METHODS

2.1 Study lakes

The location of the study lakes is presented in Figure 1 and the hydrographic properties of the lakes in Table 1. The bedrock in all the drainage areas consisted mainly of acidic granite, gneiss and granodioritic minerals. The content of calcium in water was rather low, from 1.1 to 6.9 mg l⁻¹. The most distinguishable feature in the study lakes is the organic matter content. Measured as the colour of water it varied from 5 to 250 mg Pt l⁻¹ (Table 2.). The lowest pH value was measured in lake Kangasjärvi near the western coast of Finland, whereas the lakes in northernmost Finland had the highest pH and the best buffering capacity.

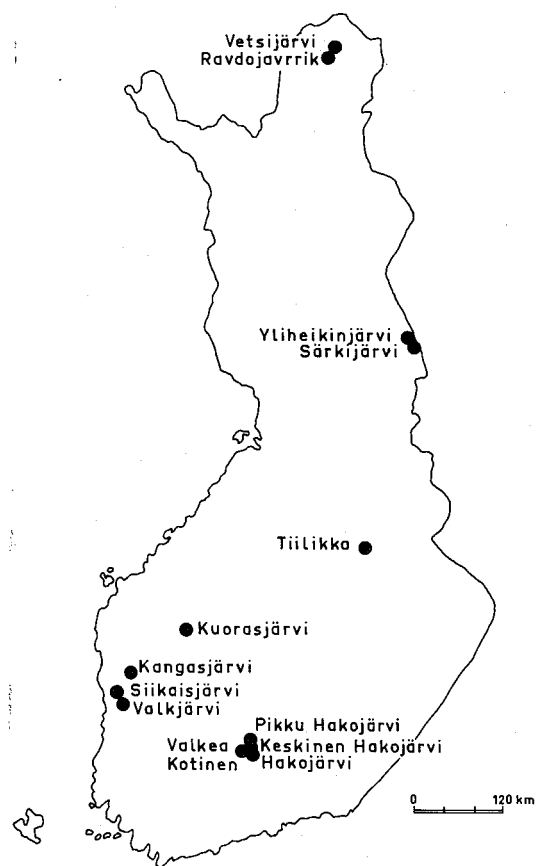


Fig. 1. The site of the study lakes.

Table 1. General characteristics of the study lakes.

Study lakes	Area	Mean depth	Volume	Area of drainage basin	Turnover time	Percentage of lakes of drainage area	Percentage of peat soils of drainage area	Percentage of drained peat soils of drainage area
	ha	m	10 ⁶ m ⁻³	km ²	a			
1 Pikku Hakojärvi	0.6	0.8	0.005	1.1	0.02	0.6	32	32
2 Keskinen Hakojärvi	1.3	1.5	0.020	1.3	0.06	1.5	30	30
3 Hakojärvi	17.0	4.7	0.800	2.0	1.40	9.5	20	20
4 Valkea Kotinen	4.0	2.0	0.100	0.7	0.46	17.0	5	0
5 Siikaisjärvi	480.0	1.4	6.500	105.0	0.20	6.0	43	
6 Valkjärvi	340.0	2.9	9.900	12.0	2.60	28.0	7	
7 Kangasjärvi	47.0	3.0	1.300	2.0	2.10	24.0	10	0
8 Kuorasjärvi	1 220.0	2.3	27.600	160.0	0.55	14.0		
9 Tiihikka	400.0	2.1	8.200	148.0	0.47	7.0	74	20
10 Yliheikinjärvi	22.0	4.1	0.900	6.5	0.45	7.0	18	12
11 Särkijärvi	24.0	2.7	0.600	27.0	0.07	13.0	15	16
12 Vetsijärvi	759.0	2.0	15.200	306.0	0.16	15.0	20	0
13 Ravdojavrrrik	56.0	2.5	2.500	13.0	0.31	12.0	10	0

Table 2. The means of some chemical water parameters in the study lakes.

Study lakes	Conductivity mS m^{-1}	Alkalinity mmol l^{-1}	pH	Colour mg Pt l^{-1}	COD Mn mg l^{-1}	Tot. P $\mu g\ l^{-1}$	Tot. Fe $\mu g\ l^{-1}$	Ca mg l^{-1}
1 Pikku Hakojärvi	5.2	0.11	5.9	210	27.0	52	1 400	6.9
2 Keskinen Hakojärvi	4.4	0.05	5.9	160	24.0	26	630	5.9
3 Hakojärvi	4.1	0.07	6.2	100	16.0	26	910	3.9
4 Valkea Kotinen	3.7	0.02	5.7	130	10.0	26	310	2.8
5 Siikaisjärvi	6.5	0.12	6.3	220	19.0	34	1 700	3.7
6 Valkjärvi	5.1	0.16	6.7	20	5.3	18	130	2.8
7 Kangasjärvi	2.8	0.01	5.3	5	2.2	12	80	1.1
8 Kuorasjärvi	5.6	0.06	5.9	260	22.0	22	1 300	2.5
9 Tiilikka	2.3	0.02	6.0	90	12.0	14	1 500	1.3
10 Yliheikinjärvi	6.3	0.30	6.6	60	8.8	7	230	5.9
11 Särkijärvi	4.7	0.31	6.4	50	9.1	13	410	5.4
12 Vetsijärvi	4.5	0.47	7.0	25	4.0	8	330	
13 Ravdojavrrik	2.0	0.07	6.7	7	3.7	6	50	

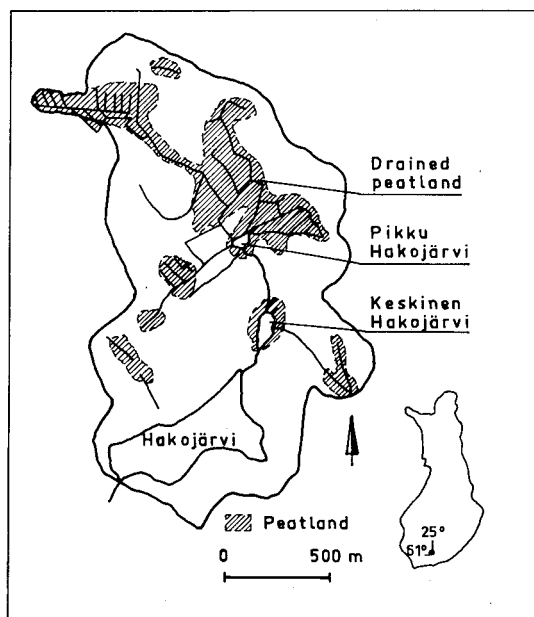


Fig. 2. Drainage basin of lake Hakojärvi.

The lakes Pikku Hakojärvi, Keskinen Hakojärvi and Hakojärvi are situated in the same runoff area and they compose a chain of lakes (Fig. 2). About 32 % of the watershed of Pikku Hakojärvi has been drained for forestry. The nearby lake Valkea Kotinen is situated in a nature conservation area, where no ditching or other forest management operations have been performed. The drainage areas of lake Kangasjärvi near the western coast

and of the two lakes Vetsijärvi and Ravdojavrrik in northernmost Finland are also unditched. However, in the drainage area of lake Kangasjärvi other forestry operations, such as clear cutting, have been performed.

2.2 Sampling and analysing methods

Sediment samples were taken with a crust-freeze sampler (Renberg 1981) at the deepest points of the lakes. Cores were cut into 1–2 cm sections and freeze-dried. The loss of ignition was measured after heating for 1 h at 550°C. Mercury was determined by the cold vapour AAS-technique after wet combustion (Armstrong and Uthe 1971).

Samples for Pb-210 dating were taken with a pistonless gravity corer (inner diameter 50 mm) and the cores were cut into 1 cm sections, freeze-dried and homogenized. The Pb-210 activity present at each depth of the sediment was determined by measuring the activity of its granddaughter Po-210. The method has been described in detail by Häsänen (1977).

In homogenous sediments the Pb-210 activity will be almost constant in the lower part of the core. This activity was assumed to correspond with the supported activity, which is the part of the total Pb-210 activity which originates from the decay of Ra-226 in the sediment. Unsupported Pb-210, on which the dating was based, was defined as the difference between total and supported activity. Calculations of age and sedimentation rate were carried out using the C.I.C. (constant initial

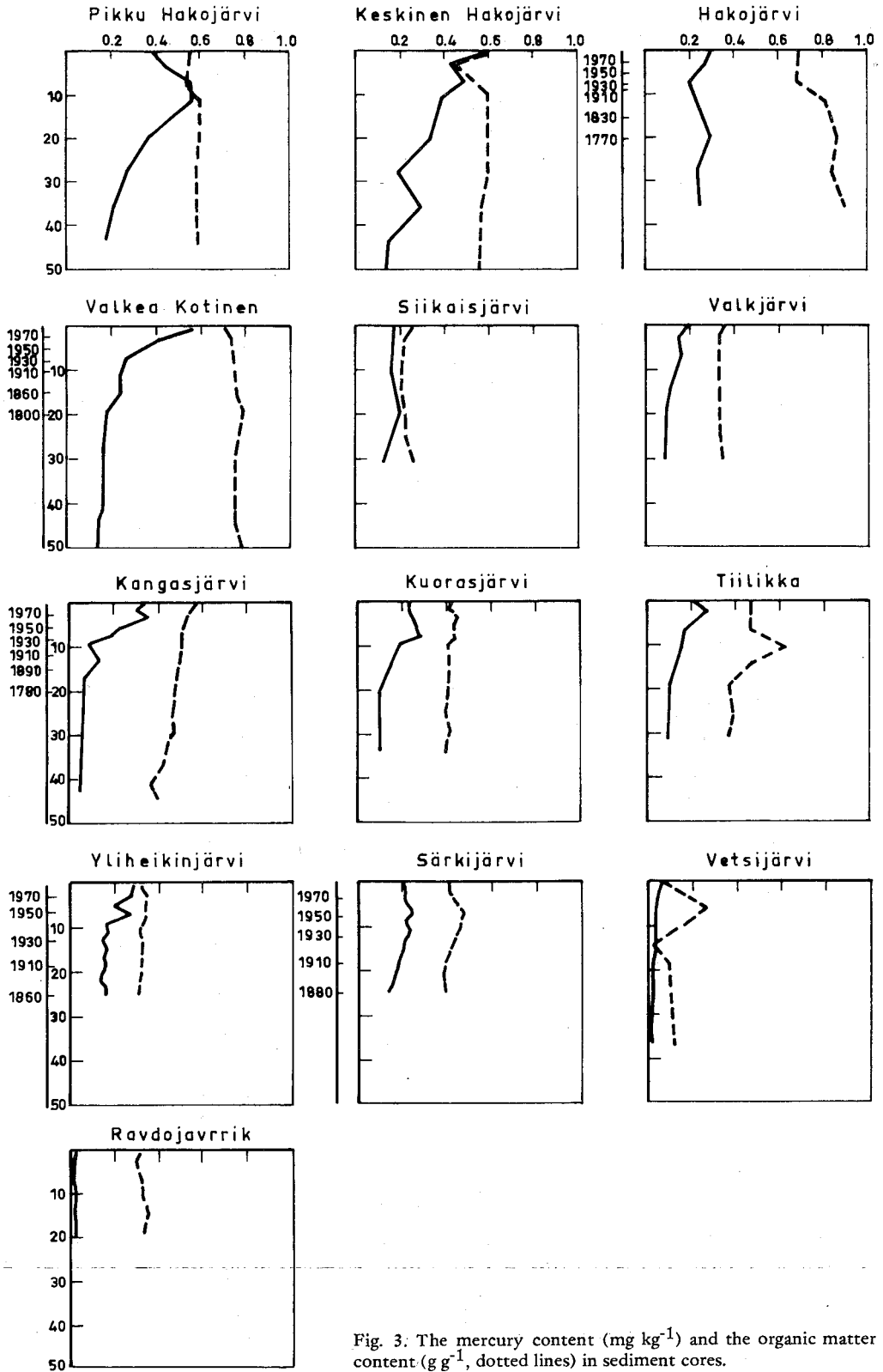


Fig. 3: The mercury content (mg kg^{-1}) and the organic matter content (g g^{-1} , dotted lines) in sediment cores.

concentration) and C.R.S. (constant rate of supply) models (Robbins and Edgington 1975, Appleby and Oldfield 1978).

3. RESULTS

3.1 Mercury and organic matter contents in sediments

The mercury contents in surface sediment layers ranged from 0.17 to 0.55 mg kg⁻¹ (dry weight) in southern and western Finland, from 0.20 to 0.24 mg kg⁻¹ in central Finland and were as low as 0.02–0.05 mg kg⁻¹ in mountain lakes in Finnish Lapland (Fig. 3). The background mercury contents in the lower sediment strata showed a similar pattern. They ranged from 0.05 to 0.25 mg kg⁻¹ (dry weight) in southern, western and central Finland and were lower in the lakes of Finnish Lapland. It is obvious that in some cases the 30–40 cm sediment core was not enough to reach the strata with the lowest mercury values.

The increase in the mercury content towards the surface sediment layers was very clear in lakes Valkea Kotinen, Keskinen Hakojärvi and Kangasjärvi. In these lakes the mercury concentration was 3.9–6.6 times higher in the surface than in the lower parts of the sediment profile. In lake Vetsijärvi in northern Finland the surface mercury content was also 5.0 times higher than the background value, although it was still very low (0.05 mg kg⁻¹). In other lakes in southern Finland the corresponding coefficient ranged from 1.4 to 2.4 and in central Finland from 1.6 to 1.7 (lakes Särkijärvi and Yliheikinjärvi).

The content of organic matter in sediment measured as loss of ignition ranged from 50 to 900 mg g⁻¹ (Fig. 3). The highest values were measured together with the highest mercury contents in sediment. In Vetsijärvi and Ravdojavrrik the organic matter contents were very low, 50 and 260 mg g⁻¹ respectively.

3.2 Sedimentation rate and mercury accumulation

The dry mass sedimentation rate was estimated by determining the Pb-210 activity in seven sediment cores (Fig. 4). In the sediment of the lakes Pikku Hakojärvi and Keskinen Hakojärvi, leaching of unsupported Pb-210 derived from the catchment

area may be regarded as a significant source of nonlinear Pb-210 activity. The assumptions for the C.I.C. and C.R.S. models are therefore not fulfilled and calculations of sedimentation rate cannot be made (Appleby and Oldfield 1978, Oldfield and Appleby 1984). In the case of the five lakes Valkea Kotinen, Hakojärvi, Kangasjärvi, Särkijärvi and Yliheikinjärvi, the direct atmospheric fallout can be regarded as the main pathway by which Pb-210 reaches the sediment.

The mean sedimentation rate ranged from 90 to 160 g m⁻² a⁻¹ (dry weight) in surface sediment layers (Fig. 5), corresponding to about 0.8–1.9 mm a⁻¹. In lakes Kangasjärvi and Hakojärvi the rate was higher in the surface layers than in the deeper layers, whereas in lakes Särkijärvi and Yliheikinjärvi the maximum sedimentation rate (up to 260 g m⁻² a⁻¹) was measured in the 10–20 cm depth. In the sediment of lake Pikku Hakojärvi some peat moss remains were observed in the 12 cm depth and the colour of the sediment above that depth was lighter. Major drainage operations (32 % of the runoff area) were carried out in 1967. If it is assumed that the moss remains have settled during the first years after ditching, an annual sedimentation of 7.5 mm can be estimated.

According to Pb-210 dating the mercury accumulation rate was calculated to range in surface sediment layers from 25 to 50 μg m⁻² a⁻¹ and in deeper layers from 5 to 27 μg m⁻² a⁻¹ (Fig. 5). In lake Pikku Hakojärvi the accumulation rate was calculated to be 370 μg m⁻² a⁻¹ from a 7.5 mm annual sedimentation.

In lakes Vetsijärvi and Ravdojavrrik in northern Finland no sediment dating was carried out, but on the basis of the very low mercury contents the mercury accumulation rate can be estimated to be much lower than the corresponding values measured in southern and central Finland. The mercury accumulation rate in the northern lakes can be estimated to be below 10 μg m⁻² a⁻¹.

4. DISCUSSION

4.1 The effect of sediment organic matter and water pH on sediment mercury contents

The mercury content was closely correlated with organic matter in both the upper and lower layers of the sediment profile (Fig. 6). The wide range in mercury contents between the lakes is partly due

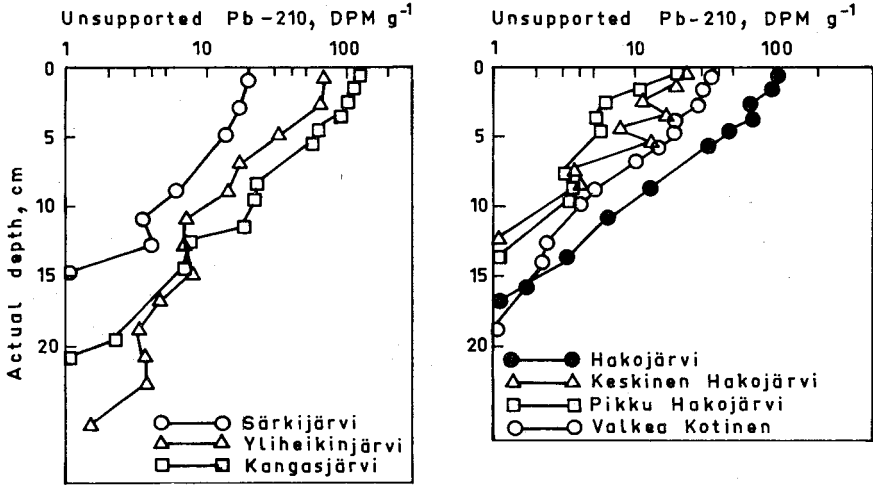


Fig. 4. The logarithm of the unsupported Pb-210 activity against the actual sediment depth.

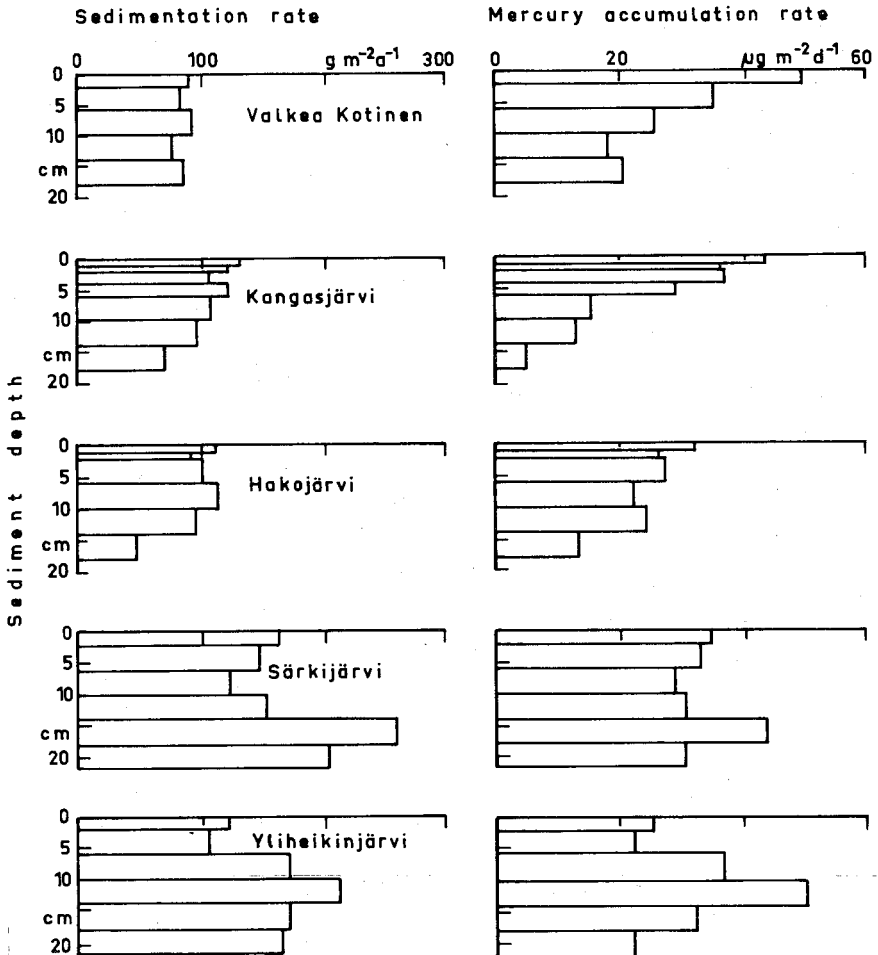


Fig. 5. The sedimentation rate and the mercury accumulation rate as a function of sediment depth.

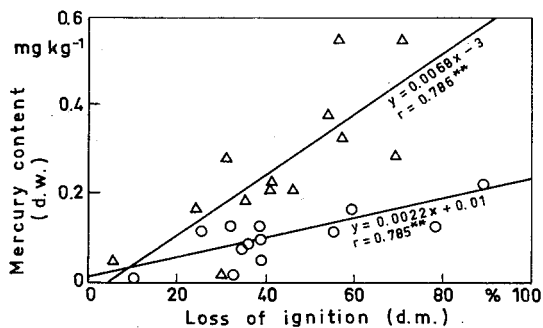


Fig. 6. The sediment mercury content as a function of the organic matter content measured as loss of ignition. Upper sediment layers (Δ), lower sediment layers (O).

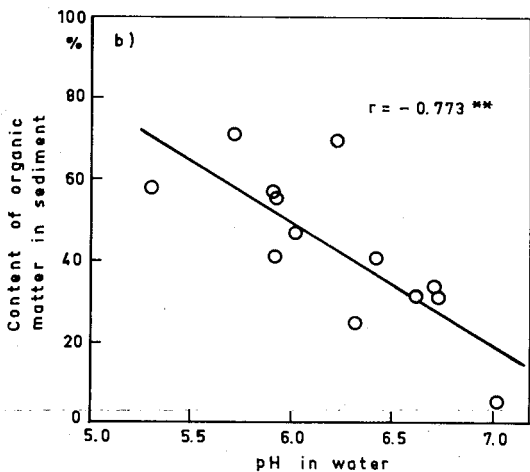
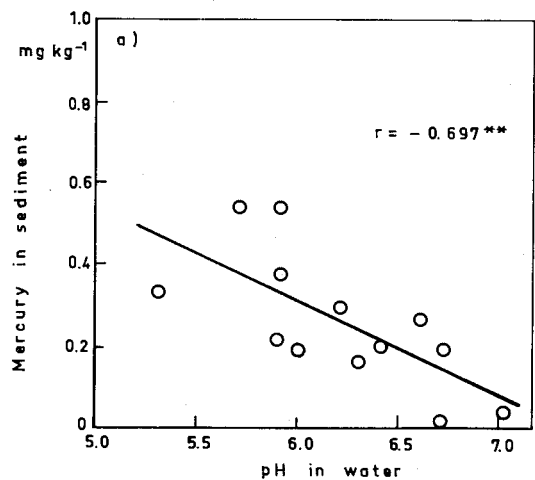


Fig. 7. The mercury content (a) and the organic matter content (b) of sediment as a function of pH in water.

to the variation in organic matter content (see also Johansson 1985). The regression coefficient between organic matter content and mercury content was smaller in the lower layers than in the surface sediment layers (Fig. 6). In spite of the increase of the mercury content the content of organic matter did not increase, as can also be seen in Fig. 3. It can therefore be concluded that the main reason for the increase in the mercury content is the increased input of inorganic mercury, most probably of atmospheric origin.

The most distinct increases in sediment mercury contents were observed in lake sediments with the highest organic matter contents (lakes Valkea Kotinen, Kangasjärvi and Valkjärvi). This could be due to their situation high in the watercourse, with long residence times compared with the other lakes in southern Finland with similar atmospheric inputs of mercury (lakes Hakojärvi, Siikaisjärvi and Kuorasjärvi). In the latter lakes the mercury derived from the catchment area has had time to settle in lakes higher in the watercourse. With longer residence times smaller particles settle out and these particles are assumed to have higher mercury contents (e.g. Cranston and Buckley 1972).

The absorption of mercury to organic matter has been reported to increase at lower pH values (e.g. Anderson 1967, Håkanson 1972, 1974, Jackson et al. 1980, Schindler et al. 1980, Lodenius et al. 1983). In our study lakes the mercury concentration in surface sediment layers was higher in lakes with low pH in the water phase (Fig. 7a), which is in agreement with the observations of Johansson (1985) in Sweden. This cannot be explained by spatial distribution. However, the content of organic matter in sediment was also high in the lakes with low pH (Fig. 7b). This could be due to the effective sedimentation of humic substances in acidic lakes or to the low rate of decomposition of organic matter in sediments with low pH values. The increased binding of mercury to organic matter at low pH values may also increase the mercury content in sediment. This implies that if the lake water is acidified, the content of mercury in the lake sediment may increase without any increase in mercury fallout or leaching to the lake.

4.2 The effect of the atmospheric fallout of mercury on sediment mercury contents and on accumulation in sediments

The mercury accumulation rate was found to range in southern Finland from 25 to 50 $\mu\text{g m}^{-2} \text{a}^{-1}$ and

in the lakes of Finnish Lapland to be lower than $1\text{C } \mu\text{g m}^{-2} \text{a}^{-1}$. Previously, the mercury accumulation rate in Finland has been reported to be from 30 to $120 \mu\text{g m}^{-2} \text{a}^{-1}$ (Simola and Lodenius 1982) and $120 \mu\text{g m}^{-2} \text{a}^{-1}$ (Tolonen and Jaakkola 1983). In Sweden the rate has been calculated to range from 10 to $40 \mu\text{g m}^{-2} \text{a}^{-1}$ (Lindqvist et al. 1984).

The most distinct increase in the mercury content, as well as in the mercury accumulation rate in sediment, was observed in lakes Valkea Kotinen and Kangasjärvi. The increase in the mercury content was observed to have accelerated considerably in the nineteen-twenties in these lakes (Fig. 3). Because no or only slight disturbances have been made in the drainage basins of these two lakes, the main reason for the increase may be the increased atmospheric fallout of mercury caused by the increased anthropogenic emissions of mercury. The pH in lake Kangasjärvi was relatively low and it is possible that this is a result of increased acid precipitation. The possible acidification of the lake water could be another reason for the increase in sediment mercury content, as discussed earlier in this paper.

The observations made in Sweden and Canada also indicate that atmospheric emissions (local or long-distance) and fallout of mercury have caused increases in the mercury contents in lake sediments (e.g. Johansson 1980, 1985, Björklund et al. 1982, Ouellet and Jones 1983). The beginning of the increase in mercury contents is also in good agreement with the results reported in Sweden and Canada. Prior to the rapid increase during the 20th century, a slight gradual increase was detectable earlier. In lake Windermere, England, the increase began as early as in the 16th century (Aston et al. 1973).

4.3 The influence of peatland drainage on sediment mercury contents

The effect of peatland drainage on the mercury content and accumulation rate in lake sediments was studied especially in lake Pikku Hakojärvi, of which 32 % of the drainage basin has been drained. The drainage of forests and peatlands has been reported to increase the leaching of suspended organic matter considerably during the first years after the drainage operations (Sallantausta and Pätilä 1983, Lundin 1984, Bergqvist et al. 1984). The mercury content of the suspended solids in lake water has been observed to be higher than that of

soil material (Bodaly et al. 1984, Surma-Aho et al. 1986, Verta et al. 1986). If the fraction of material leached from the drainage area of the total sedimenting material increases, it is therefore probable that the mercury content in the sediment will decrease. In lake Pikku Hakojärvi the sediment mercury content first increased up to a depth of about 12 cm, possibly due to the increased atmospheric fallout of mercury. Subsequently, forestry drainage operations have increased the leaching of allochthonous material and the sediment mercury content has begun to decrease. Decrease in the mercury content of the uppermost sediment layers was also reported by Simola and Lodenius (1982).

The decrease in the mercury content in the upper sediment layers of lakes Kuorasjärvi and Tiilikka may also be a consequence of drainage operations. Forestry drainage in the runoff area of lake Tiilikka has been very widespread (20 % of the drainage area). The exact percentage of the drainage area in the lake Kuorasjärvi drainage basin is not known, but it can be estimated to be more than 10 %. The beginning of the decrease in mercury content was observed in notably higher sediment layers than in lake Pikku Hakojärvi. This can be explained by the lower sedimentation rate in lakes Tiilikka and Kuorasjärvi.

The increased leaching of suspended organic matter after ditching operations causes, despite the decrease in the mercury content in sediment, an increased mercury load to the watercourse. On the basis of the peat moss remains the mercury accumulation rate was estimated to be as high as $370 \mu\text{g m}^{-2} \text{a}^{-1}$ in lake Pikku Hakojärvi. The accumulation rate in the lower sediment layers in lake Pikku Hakojärvi is not known, but by comparison with the other study lakes at the same latitude with equal atmospheric fallout, the mercury accumulation rate can be estimated to be about 7–12 times higher in the upper sediment layers than in deeper layers. Simola and Lodenius (1982) have also reported the maximum mercury accumulation rate ($120 \mu\text{g m}^{-2} \text{a}^{-1}$) to be much higher than the background value in lake Polvijärvi in eastern Finland, of which 28 % of the runoff area has been drained. In lake Nummijärvi in western Finland Tolonen (pers. comm.) observed the maximum mercury accumulation rate to be $122 \mu\text{g m}^{-2} \text{a}^{-1}$, which was 14.6 times higher than the background value in the 20 cm sediment depth. Major drainage operations have been carried out in the runoff area of lake Nummijärvi for peat production.

5. CONCLUSION

According to this study the main reason for the increase in sediment mercury contents appears to be the increased atmospheric fallout of mercury. In some lakes, coincident acidification may also have increased sediment mercury contents. Land use operations in the drainage areas may even decrease the mercury contents in sediments, but the overall mercury load caused by these operations can be very high in extreme cases, due to the increased runoff and sedimentation.

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TIIVISTELMÄ

Tutkimuksen tarkoituksena oli selvittää metsäjärvien sedimenttien elohopeapitoisuuden muutoksia sekä ilman kautta kulkeutuvan elohopean että valuma-alueella suoritettujen metsäojitusten vaikutuksia sedimenttien elohopeapitoisuuteen.

Sedimentin pintaosien elohopeapitoisuuden havaittiin vaihtelevan 0,17—0,55 mg kg⁻¹ (kuiva-ainetta) Etelä- ja Keski-Suomessa. Lapissa sijaitsevien järvien pintasedimentin elohopeapitoisuus vaihteli 0,02—0,05 mg kg⁻¹. Sedimentin syvemmissä osissa elohopeapitoisuus oli yleensä alempi. Elohopean kertymisnopeus vaihteli sedimentin pintaosissa 25—50 μg m⁻² a⁻¹ ja oli Lapin järvissä alle 10 μg m⁻² a⁻¹. Metsäojitusten erittäin voimakkaasti kuormittamassa Pikku Hakojärvässä kertymisnopeudeksi arvioitiin 370 μg m⁻² a⁻¹.

Sedimentin elohopeapitoisuuden havaittiin korreloivan positiivisesti orgaanisen aineen pitoisuuden kanssa ja negatiivisesti veden pH:n kanssa. Elohopeapitoisuuden kasvun havaittiin kiihtyneen voimakkaasti 1920-luvulla. Parhaiten kasvu ilmeni

valuma-alueeltaan luonnontilaisissa ja ojitamattomissa järvissä. Kasvun pääteltiin aiheutuneen pääasiassa ilmaväntäisen elohopean ja elohopealaskeman kasvusta. Metsäojitusten todettiin mahdollisesti alentavan sedimentin elohopeapitoisuutta, mutta lisäävän elohopean kerääntymistä sedimenttiin ääritapauksissa jopa moninkertaisesti lisääntyneen sedimentaation johdosta.

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