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THE ORIGIN AND LEVEL OF MERCURY IN FINNISH FOREST LAKES

Matti Verta1), Seppo Rekolainen1), Jaakko Mannio1) & Kari Surma-Aho2)


Mercury concentrations of pike in Finnish lakes affected by no known mercury pollution were analysed in 1980—1983. The effect of water quality, hydrographic and morphometric parameters, land use, mercury content of the diet and mercury content of sediments on the mercury content of pike were studied on the basis of correlation analysis and stepwise regression analysis. The roach mercury content, the terrestrial catchment area/lake volume ratio and water quality parameters describing organic matter content in the water all correlated positively with pike mercury contents, whereas the areal percentage of lakes in the catchment area was negatively correlated. Slowly growing pikes had higher mercury concentration in relation to the mercury content of the diet than did rapidly growing pikes. As much as 57% of the mercury variation in 1 kg pike from different lakes could be explained by the morphometric, chemical and biological variables studied. The mercury level in pike in southern and central Finland was estimated to have increased during the past 100 years by a factor of about 2. The most probable reason for this was concluded to be an increased load of atmospheric mercury.

Index words: Mercury, methylation, lakes, pike.

1. INTRODUCTION

Emissions of mercury to the atmosphere in Finland have been estimated at about 1 ton or more per year in recent years (Ympäristönsuojelunneuvosto 1982, Lodénius 1985). Before about 1970, however, the emissions were higher, probably of the order of 3—10 tons per year (Häsänen 1975, Ympäristönsuojelunneuvosto 1982). Present emissions in Europe are several orders of magnitude higher and range from 300 tons to 1200 tons per year (Lindqvist et al. 1984). High-level emissions from central Europe and Sweden are thought to have increased the fish mercury levels considerably in lakes in southern and central Sweden, probably due to the increased leaching of atmospheric mercury bound to humic substances in the soil (Björklund et al. 1984, Lindqvist et al. 1984).

The effect of organic material and particularly of humic substances in binding mercury in soils, sediments and freshwaters is well known (eg. Andersson 1967, Strohal and Huljev 1971, Håkanson 1974, Cheam and Gamble 1974, 1) Water Research Institute, National Board of Waters, P.O. Box 250, SF-00101 Helsinki, Finland

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Lindberg and Harris 1974, Benes et al. 1976, Lodenius and Seppänen 1984) and recent findings indicate that the methylation of mercury to monomethyl mercury may also occur abiotically in environments containing large quantities of humic material (Rogers 1977, Nagase et al. 1982, Lee et al. 1985). Alfthan et al. 1983 and Verta 1984 further hypothesized that methylation of mercury may take place through the biological degradation of humic material in polyhumic lakes or in the soil. This hypothesis was supported by findings of good correlations between fish mercury contents and organic material and oxygen depletion in the water of impounded reservoirs (Verta 1981).

An intensive programme of forestry draining (16% of the total land area) has caused a considerable load of organic and inorganic material including mercury to recipient small forest lakes in Finland (Seuna 1982, Simola and Lodenius 1982, Rekolainen et al. 1986b). This has also been proposed to have increased fish mercury levels in these lakes (Lodenius 1983).

When relatively high mercury concentrations were observed in small forest lakes subjected to no direct mercury contamination, a joint project was undertaken in 1982–1984 to study the relative impact of different anthropogenic sources of mercury on mercury concentrations in fish in these lakes. In addition, the available studies of the water and health authorities on the level of mercury in fish in 1980–1981 were collected and summarized.

2. MATERIALS AND METHODS
2.1 Study lakes

The lake material in the studies in 1980–1981 was comprised of 57 lakes situated throughout the country, mainly in southern and central Finland. The lakes were partly highland lakes (32) and partly lakes situated downstream (25), with a wide range of water quality.

In 1983, 36 lakes were chosen for a more detailed study. The lakes were highland lakes with one exception and were located in the water divide area from the western coast to the eastern border of Finland (Fig. 1). Most of the lakes were small with an area of less than 10 km² and maximum depth less than 10 m. Only six lakes had a surface area of more than 20 km².

The catchment areas of the lakes consisted mainly of forests and peatlands. Forest management operations such as construction of roads, clearcutting and peatland ditching were the most important disturbances in the areas. The largest lakes, however, also had cultivated land (up to 15%) and small municipalities in their catchment areas. In a few cases only very little or no disturbances had occurred in the catchment areas of the lakes.

The main water quality criteria in selecting the lakes in 1983 were organic matter content and pH. In the study lakes low pH values were strongly indicative of high organic matter contents (Mannio et al. 1986) and only one clear
lake with low pH (possibly acidified, Simola et al. 1985) was recorded. The lakes were characterized as of low ionic concentration, mostly oligotrophic, from neutral to acidic and from clear-water to highly coloured polyhumic water (Table 1).

### 2.2 Sampling and analysis methods

Of the lakes studied in 1980–1981 only those with mercury data from three or more pikes (Esox lucius L.) were accepted. The average sample size was six pikes per lake.

In 1983 an average of 10 pikes (Esox lucius L.) (range 4—35) and 10 roaches (Rutilus rutilus L.) were caught in spring during spawning. The fish were frozen immediately in aluminium foil and stored at −20°C. A sample of muscle tissue was taken from under the dorsal fin of half-thawed fish. Pikes were analysed separately, whereas a homogenate was made of the five smallest and five largest individual roaches.

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 from their tubes and freezing. Samples were freeze-dried under reduced pressure before analysing.

Zooplankton samples were also taken from littoral areas using 400 μm mesh plankton nets. Samples were frozen immediately in the field with solid carbon dioxide (−79°C). Zooplankton samples were freeze-dried under reduced pressure before analysing.

Sediment samples (0—2 cm) were taken with a pistonless gravity corer in a plexiglass tube with an inner diameter of 5 cm from 3—5 different places and pooled. The samples were freeze-dried and their dry weights were measured as well as their loss on ignition (550°C, 1 h).

Sediment samples were analysed from nitric-sulphuric acid digests by the cold vapour atomic adsorption spectrophotometric technique (Armstrong and Uthe 1971). Fish, zoobenthos and zooplankton were analysed for inorganic and organic (methyl) mercury (Surma-Aho et al. 1986).

### 2.3 Statistical analysis

Mercury contents in 1 kg pike were estimated by linear regression analysis. Correlation- and stepwise regression analysis (Dixon and Jennrich 1983) were used to determine the environmental variables which best explained pike mercury contents. In the final test the variables used were:

**Morphometric**
- Lake area (log scale)
- Total catchment area (log scale)
- Terrestrial drainage area/lake volume (log scale)
- Morphoedaphic index (conductivity/mean depth)
- Mean depth
- Lake percentage

**Relative depth** (mean depth/area)
- Maximum depth
- Percentage of mineral forest soils (n=19)
Percentage of peatlands (n=19)
Percentage of drained peatlands (n=19)

Chemical
Oxygen saturation
Suspended solids
Conductivity
Alkalinity
pH
Colour
Silica
Nitrogen

Biological
Pike growth rate
Hg content of roach

The analysis was first carried out separately for water quality and morphometric variables, then with the combination of these two and finally with the combination of all three sets of data. The analysis was repeated as long as the F-value of the added variable was significant (p < 0.01).

3. RESULTS
3.1 Mercury level

The mean total mercury concentration of 1 kg pikes in the 93 lakes studied was 0.53 mg kg⁻¹ (Table 2). Of the whole lake material 67 lakes could be classified as forest lakes, with forest and peatland representing 80% or more of the catchment area and no large municipalities. The mean 1 kg pike mercury content in these lakes was 0.56 mg kg⁻¹. Other lakes representing lakes in areas of intensive cultivation or downstream lakes, had a mean of 0.44 mg kg⁻¹ in 1 kg pike.

Pike mercury contents in four lakes exceeded 1.0 mg kg⁻¹, which is the highest permissible level of mercury in fish for human consumption in Finland, laid down by the National Board of Health. In half of the lakes pike mercury contents exceeded 0.5 mg kg⁻¹, the limit at which restrictions on the consumption of fish are recommended. A clear difference was found between pike Hg-contents in southern and central Finland on the one hand and northern Finland on the other in 1980—1981, the lower levels being recorded in the north. The mean pike Hg content in 8 lakes situated in Finnish Lapland was only 0.28 mg kg⁻¹ with a range from 0.09 mg kg⁻¹ to 0.58 mg kg⁻¹. The highest concentration was recorded in an impounded lake subject to intensive water level regulation. In other trophic levels clearly lower mercury levels were observed (Table 2).

3.2 Correlations

The mercury contents in pike and roach correlated positively with all water quality variables describing the humic matter content of the water (Fig. 2, Mannio et al. 1986). Phosphorus and iron, which had strong positive correlations with humic material, also correlated positively with fish mercury contents. Water pH correlated negatively with pike mercury content in the data from 1980—1981 (p < 0.01) but did not correlate in the 1983 data. The best correlation was recorded between the high molecular weight

<table>
<thead>
<tr>
<th></th>
<th>Tot. Hg mg kg⁻¹</th>
<th>Met. Hg %</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>Range</td>
<td></td>
</tr>
<tr>
<td>Pike (1 kg)</td>
<td>1980—1981</td>
<td>0.45</td>
</tr>
<tr>
<td>Pike (1 kg)</td>
<td>1983</td>
<td>0.66</td>
</tr>
<tr>
<td>Roach</td>
<td>1983</td>
<td>0.31</td>
</tr>
<tr>
<td>Zooplankton</td>
<td>1983</td>
<td>0.25</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Trichoptera</td>
<td>1983</td>
<td>0.24</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Odonata</td>
<td>1983</td>
<td>0.24</td>
</tr>
<tr>
<td>Sediment (0—2 cm)</td>
<td>1983</td>
<td>0.15</td>
</tr>
</tbody>
</table>

n.a. = not analysed
(HMW) fraction of dissolved organic material and pike (p < 0.001) and roach (p < 0.01) mercury contents in 1983 (see also Mannio et al. 1986). In the data from 1980—1981 CODMn was best correlated with pike mercury content (p < 0.001).

Of the morphometric parameters studied the areal percentage of lakes in the catchment area correlated negatively (p < 0.01) with pike mercury contents and the terrestrial catchment area/lake volume ratio correlated positively (p < 0.01) (Fig. 3). The percentage of mineral forest land in highland lakes correlated positively (p < 0.05) with the pike mercury content. The data used for this calculation was, however, rather small.

The mercury concentration in roach correlated positively with pike mercury content (p < 0.01, Fig. 4), whereas the total or methyl mercury content in surface sediments, zooplankton and in benthic animals (trichoptera) did not correlate.

In seven of the lakes the pike mercury contents were higher in relation to roach mercury content than in the other lakes (Fig. 4). To study the difference between lakes, 16 lakes with similar roach mercury contents were examined with Student’s t-test. The results showed that the lakes with high pike mercury contents had lower growth rates of the pike, were deeper and had higher organic matter contents than the lakes with similar roach mercury contents but lower pike mercury contents.

---

Fig. 2. The total mercury content of 1 kg pike as a function of (a) high molecular weight (HMW) organic matter, (b) colour and (c) chemical oxygen demand (CODMn) in the lake water in September—October.
3.3 Stepwise regression analysis

As a result of stepwise regression analysis five equations were developed to describe pike mercury contents:

\[
Hg = 0.0028 \text{COL} - 0.379 \\
R = 0.575
\] (1)

\[
Hg = -0.0165 \text{LP} - 0.126 \log \text{LA} + 0.984 \\
R = 0.584
\] (2)

\[
Hg = 0.287 \log (\text{DA/V}) - 0.131 \log \text{LA} + 0.984 \\
R = 0.584
\] (3)

\[
Hg = 0.0027 \text{COL} - 0.0827 \log \text{LA} + 0.447 \\
R = 0.622
\] (4)

\[
Hg = 1.12 \text{RO} - 0.012 \text{LP} - 0.148 \log \text{LA} - 0.429 \text{GR} + 0.756 \\
R = 0.757
\] (5)

where:

- \( Hg \) = mercury content of 1 kg pike (mg kg\(^{-1}\), ww.)
- \( \text{COL} \) = mean water colour (Pt mg \(^{-1}\))
- \( \text{LP} \) = lakes in the catchment area (%)
- \( \text{LA} \) = lake area (km\(^2\))
- \( \text{DA/V} \) = terrestrial drainage area (m\(^2\)) / lake volume (m\(^3\))
- \( \text{MD} \) = mean depth (m)
- \( \text{RO} \) = mean mercury content of roach (mg kg\(^{-1}\), ww.)
- \( \text{GR} \) = the coefficient of exponential growth rate of pike \((\mu - \rho)\), according to the equation \( \ln W = \ln W_0 + (\mu - \rho) t \), where:
  - \( W \) = weight (g) at time \( t \)
  - \( W_0 \) = weight (g) at time \( t_0 \)
  - \( \mu \) = growth rate coefficient (1/t)
  - \( \rho \) = respiration rate coefficient (1/t)
  - \( t \) = time (a)
4. DISCUSSION

4.1 Fish mercury levels

The results obtained are in good agreement with those obtained by Miettinen and Verta (1985). They found a mean mercury level of 0.48 mg kg\(^{-1}\) in 1978–1979 in pikes from Finnish lakes with no known mercury pollution. Pike mercury contents were somewhat lower than those found in Swedish forest lakes (Lindqvist et al. 1984, Björklund et al. 1984). In Sweden the areal mean values ranged from 0.57 to 0.98 mg kg\(^{-1}\) in southern and central districts and was 0.36 mg kg\(^{-1}\) in the north. The lakes studied in Sweden were somewhat smaller on average than those in the present study. According to equations 2–5 the difference in lake area should, however, have resulted in only 0.05–0.1 mg kg\(^{-1}\) higher mercury levels in the Swedish lakes and the greater difference cannot be explained by the size of the lakes. The observed lower mercury content in pikes in Finnish Lapland is consistent with the results from Swedish Lapland and is most probably a consequence of lower mercury loads in the north (Björklund et al. 1984, Lindqvist et al. 1984, Rekolainen et al. 1986a,b).

The calculations of mercury content in 1 kg pike, however, were made using different equations in the Swedish study and in the present investigation. In the Finnish material of 1983 the difference between the results of the linear regression equation and the Johnels–Westermark equation

\[
\sum \frac{\text{Hg concentration}}{\text{weight}} \times \frac{1}{n}
\]

used in the Swedish studies (Björklund et al. 1984, Lindqvist et al. 1984) was tested.

The latter yielded on average 20 % higher results for 1 kg pike mercury contents than did the linear regression model when using only pikes with weights between 0.6 and 1.4 kg. The lower was the Hg-content and the lower the mean weight of pikes, the greater was the difference between these two calculations. It is thus possible that part of the apparent difference between mercury levels in pike in Swedish and Finnish lakes originates in differences between the calculations.

4.2 The 'natural' mercury level in pike

A crucial question when trying to estimate increases in the mercury contents of fish is the natural level of mercury in fish prior to any influence of human activities. However, no mercury analyses were carried out from fish in Finnish lakes before 1966. In Sweden the natural mercury level in pike has been estimated to have been between 0.05 and 0.2 mg kg\(^{-1}\) fresh weight. This was based on analyses carried out during the 1930s and 1940s and, on the other hand during the 1960s in lakes unaffected by mercury discharge and situated in Scandinavian mountains in Norway and in Swedish Lapland (Stock and Cucuel 1934, Raeder and Snevik 1941, Johnels et al. 1967a, b). Investigations of the feathers of museum specimens of fish-eating birds were also used (Edelstam et al. 1969).

The mercury content in bedrock has not been shown to have any major influence on the mercury content of fish (Johnels et al. 1967, Johnels et al. 1979). The findings of this study from Finnish lakes situated in clayish soils indicate that pike in these waters have mercury levels very close to 0.2 mg kg\(^{-1}\).

The mean mercury level found in pike in Finnish Lapland in the present study, 0.28 mg kg\(^{-1}\), is close to the Swedish estimate of the natural level. It is, however, questionable whether this level can be regarded as 'natural' in the forest lakes of southern Finland. According to sediment data (Rekolainen et al. 1986b), the mercury content of sediments and consequently mercury accumulation in sediments in lakes in southern Finland was noticeable higher than in lakes in northern Finland already in the 18-th and 19-th centuries.

It can be assumed that in the 'natural' state fish mercury contents are also related to organic matter contents in polyhumic lakes. With this assumption an estimate of the possible natural level of mercury in pike in forest lakes with different contents of organic matter can be made as follows (Fig. 5):

- the 'natural' level of mercury in pike in clear water lakes is the level presented in the literature (0.05–0.2 mg kg\(^{-1}\))
- the maximum natural level of mercury in pike can be calculated by assuming that the relationship between pike mercury content and dissolved organic matter in the water was of the same magnitude before the onset of man's influence as in the present study (Fig. 2)
- the most probable natural level of mercury in
pike can be calculated by assuming that the relationship between pike mercury content and dissolved organic matter in the water was 45% of that in the present study. This is the same change as was observed between organic matter and mercury content in sediment profiles during the past 100 years (Rekolainen et al. 1986b). Because the mean mercury level of 1 kg pike in forest lakes in the present study was 0.56 mg kg\(^{-1}\) and ranged from 0.11 to 1.3 mg kg\(^{-1}\) in southern and central Finland, it appears that the mercury content in pike may have increased on average by 0.2—0.5 mg kg\(^{-1}\) (Fig. 5). This means that the pike mercury content may have doubled in southern and central Finland. This calculation is considerably lower than the Swedish estimate of mercury increases in pike in southern and central Sweden (Björklund et al. 1984, Lindqvist et al. 1984). In Finnish Lapland the pike mercury content, according to these assumptions, has remained at or very close to the 'natural' level as was also observed in northern Sweden.

4.3 The effect of morphometric factors and water quality on mercury concentrations in pike

The most important feature of the correlation analysis was that all the chemical variables describing allochthonous organic matter content in the water correlated positively with pike mercury content (see also Mannio et al. 1986). Consequently it was not surprising that the variable, which described the leaching of allochthonous material in relation to the lake volume (DA/V), also had a positive correlation with pike mercury content. As could be expected the ratio DA/V correlated positively with variables describing organic matter content (p < 0.001), suspended solids (p < 0.01), turbidity (p < 0.01), iron (p < 0.001) and total phosphorus (p < 0.001). The variables describing the organic matter content or load in a lake probably also describe the content or load of mercury in the lake. A strong positive correlation between fish mercury level and the drainage basin/lake volume ratio was also reported by Suns et al. (1980).

The areal percentage of lakes in the runoff area (the study lake included), on the contrary, correlated negatively with organic matter content (p < 0.01), turbidity (p < 0.01), iron (p < 0.001), total phosphorus (p < 0.01) and pike mercury content (p < 0.01) probably due to more efficient sedimentation of allochthonous material and mercury. However, only about one third of the variation in mercury content in pike could be explained with these variables (equations 1—4).

The weak positive correlation between mineral forest soils in the catchment area and pike mercury content may be an indication of the methylation of mercury in the surface humic layers of these lands. Verta et al. (1986) observed that mercury attached to humic layers of the podsol-type profile was more readily dissolved after flooding, probably due to more intensive bacterial activity, than was mercury attached to peatlands. According to Andersson (1979), microbial processes are the most important in the methylation of mercury in soils.

Pike mercury contents did not correlate with the percentage of peatland and drained peatland in the catchment area. The lakes with no or very few drainage operations in the catchment area had the same level of mercury in pike as did the lakes with 30—40% of the catchment area drained during the last 20 years. This was in spite of the fact that in most lakes drainage operations can be assumed to have caused an increase in the mercury load to the lake, as illustrated by Simola and Lodenius (1982) and Rekolainen et al. (1986b). The mercury load to these lakes, however, has mainly increased due to the mercury bound to suspended organic matter, which

![Fig. 5. The observed mercury content in pike and different estimates of the natural level as a function of water colour.](image)
sedimentates very effectively after entering the lake. Only a slight and temporary increase in dissolved organic matter has been observed in the leached water after peatland drainage (Kenttämmies and Laine 1984, Bergqvist et al. 1984). It is obvious that possible short-time effects of peatland drainage on the methyl mercury content of biota are not seen in this kind of study, but demand several years of monitoring before and after the drainage operations.

4.4 Pike growth rate and mercury content

If pike receives most of its mercury from its diet, the mercury level in the diet (roach) should correlate strongly and positively with pike mercury content. In the present data this correlation was good (p < 0.01) but no better than the correlation between pike and most variables describing the organic matter content in the water (Figs. 2 and 4).

The t-test between the seven lakes with unusually high mercury contents in pike in relation to roach mercury content revealed that the former lakes had a lower average growth rate of pike. These 'high level lakes' were the same lakes that also, in relation to organic matter content, to DA/V and to lake percentage, contained more mercury in pike than most other lakes. Elimination of these 'high level lakes' from the data did not, however, affect the correlation of these variables with pike mercury content. It is thus likely that, in addition to the factors determining the mercury load and methylation in lakes, the factors affecting the accumulation efficiency, such as the growth rate, greatly determine the level of mercury in pikes. This is also indicated by the fact that equation (5), which included two biological variables describing the accumulation efficiency of mercury, best explained pike mercury contents.

4.5 The role of atmospheric mercury

Lakes with very small disturbances in the catchment area had the same level of mercury in fish as did the lakes in areas with intensive forest management operations. However, mercury accumulation in the sediments of these lakes has been estimated to have increased by a factor of 1.4 — 6.6 during the last century (Rekolainen et al. 1986b). On the basis of similar findings in Scandinavia, several authors have concluded that the main reason for this has been the increased load of airborne mercury reaching these lakes from anthropogenic emissions to the atmosphere (Tolonen and Jaakkola 1983, Johansson 1985, Rekolainen et al. 1986b).

The present load of atmospheric mercury to the surface of the lakes < 10 μg m⁻² a⁻¹ in southern and central Finland (Rekolainen et al. 1986a) is only about one third of the amount of mercury that is annually sedimentated to the bottom sediments of the lakes (16—32 μg m⁻² a⁻¹) (Rekolainen et al. 1986b). The mercury leached from the catchment area thus represents the main load of mercury to a typical small forest lake in southern Finland. This calculation is consistent with the finding of good correlations between mercury in pike and the morphometric variables discussed earlier. It is therefore reasonable to assume that this mercury, bound to organic material, determines to a great extent the level of methyl mercury in the biota of these lakes. What proportion of atmospheric mercury and mercury derived from weathering of rocks is represented in the leached mercury is not easy to quantify. However, if anthropogenic emissions have increased the mercury load to the lakes, they have most probably to some extent caused the increase of methyl mercury in biota.

5. CONCLUSIONS

Mercury contents in fish in small forest lakes in southern and central Finland have increased during the last century. In northern Finland the mercury content is very close to or at the natural level. The most probable cause of a general increase of mercury contents in fish is the increased load of atmospheric mercury due to anthropogenic emissions to the air.

The variation of fish mercury content is wide and the environmental factors affecting mercury contents in fish are only poorly understood. The effect of human activities on the leaching of mercury from the catchment area and on the content of methyl mercury in lakes requires further research. Moreover, the relative importance of different methylation sites (soil, water, sediment)
and factors affecting methyl mercury accumulation in the biota of lakes should also be investigated.

ACKNOWLEDGEMENTS

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