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MERCURY CONCENTRATIONS IN AN AQUATIC ECOSYSTEM DURING TWENTY YEARS FOLLOWING ABATEMENT THE POLLUTION SOURCE

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ABSTRACT. Phenyl Hg was widely used as a slimicide in Finnish pulp industry until the end of 1967. The use of Hg caused a significant increase of Hg levels in fish in several areas. High concentrations were measured in Lake Kirkkojärvi in Hämeenkyrö, SW Finland. Vast amounts of Hg are still present in the lake sediments. Since 1968 uncontaminated fibres have partly covered the contaminated layers. Since 1971 Hg has been monitored in fish, sediments and aquatic plants in the water course downstream from the pulp and paper factory. The Hg concentration of a 1-kg pike (*Esox lucius*) has decreased from 1.5 $\mu g\ g^{-1}$ in the years 1971-74 to 0.8 $\mu g\ g^{-1}$ in 1990.

1. INTRODUCTION

Phenyl Hg was widely used as a slimicide in Finnish pulp industry until the end of 1967. The use of Hg caused a significantly increased level of Hg in fish in several areas. The highest concentration ever measured in Finland (6.9 $\mu g\ g^{-1}$) was recorded in 1975 from Lake Kirkkojärvi in Hämeenkyrö, SW Finland. During the first years after the ban of phenyl Hg no fish samples were analyzed for Hg. Starting in 1971 the Hg pollution has been monitored for 20 yr in the water course downstream from the paper and cardboard factory.

2. MATERIAL AND METHODS

2.1 Study area

The Hämeenkyrö water course is situated in SW Finland (Figure 1). The paper and cardboard factory is situated downstream from the unpolluted Lake

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Kyrösjärvi on the shore of River Pappilanjoki. The effluents of the factory contained O_2 consuming compounds (fibres) nutrients and phenyl Hg. The exact amount of Hg released is unknown. Since 1968 uncontaminated fibres have partly covered the Hg contaminated layers.

The mean flow of River Pappilanjoki is 24 $m^3 s^{-1}$ and most of the fibres discharged from the factory are deposited in the sediments of Lake Kirkkojärvi. This lake is eutrophic with periodically occurring O_2 depletion in the hypolimnion. The color values are high: 80 to 100 mg Pt L^{-1}.

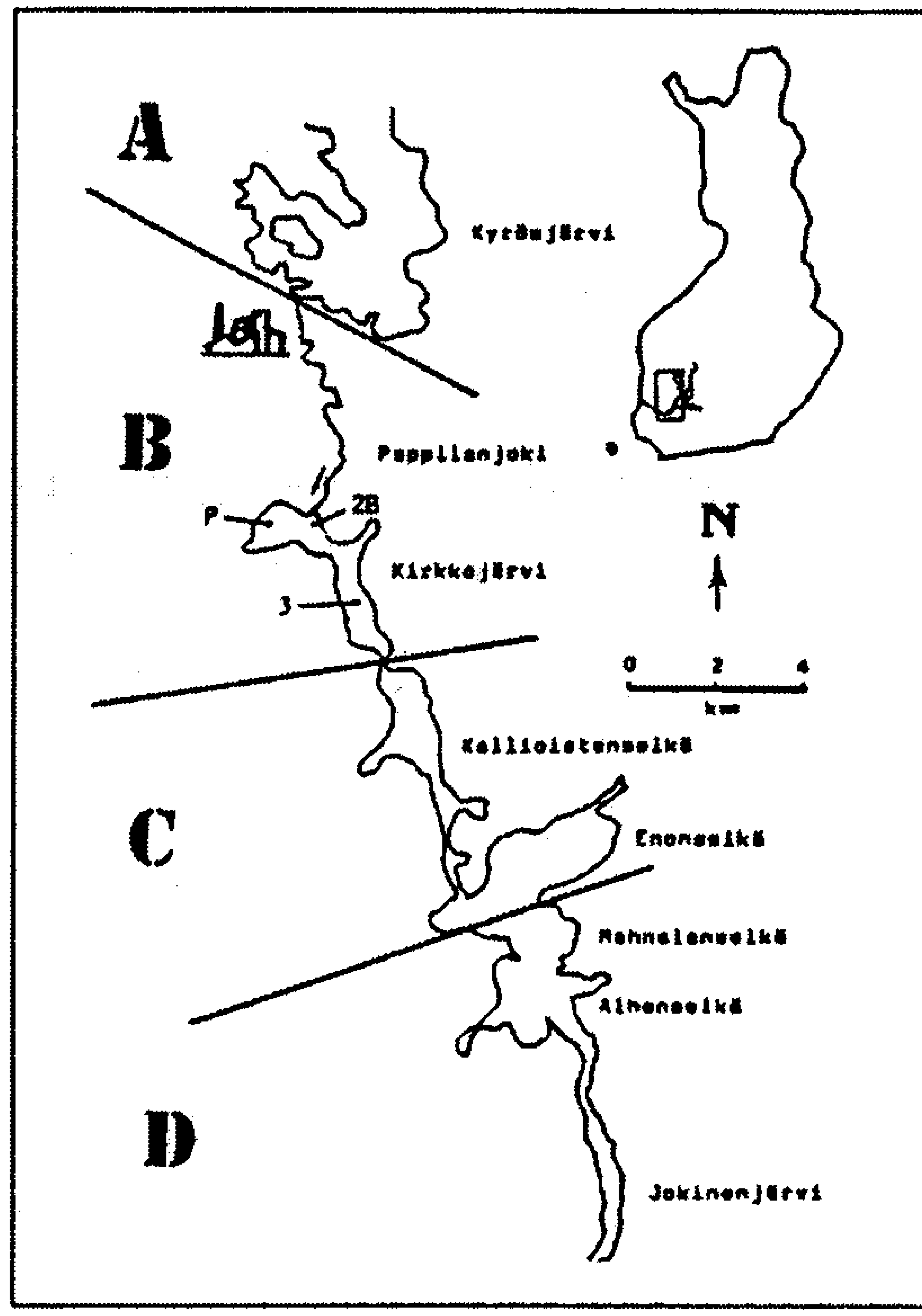


Fig. 1. The study area divided into four subareas (A, B, C, D) and the sampling sites for sediments and aquatic plants (2B, P, 3).

2.2 Samples of sediments, aquatic plants and fish

Sediment samples have been collected regularly since 1972 (unpublished reports by the Kokemäenjoki River Water Protection Association). A sediment profile was collected from the deepest site (site 3) of Lake Kirkkojärvi in August 1988 using a sediment corer (ø 6.9 cm). Two additional samples were collected from the same lake from shallow water (sites 2B and P). The sediment cores were sliced into 1 to 4 cm thick samples.

Samples of aquatic plants were collected in 1976 (Suominen *et al.,* 1977), in 1978 (Lodenius, 1980) and in 1988 (from three sites). The plants samples were dried (+40°C) and homogenized.

The fish material consists of literature data (Nuorteva *et al.*, 1975; Anon., 1976; Mankki, 1982, 1989; Mankki and Näsi, 1985) and fish sent to our laboratory for Hg analyses. Part of the data have been published earlier (Nuorteva *et al.*, 1989; Lodenius, 1988). Muscle Hg data are available for 294 specimens of Northern Pike (*Esox lucius*). Since 1979 samples of liver (N = 129), kidney (N = 123) and gonads

(N = 72) have been collected in addition to muscle samples. The fish samples were stored frozen before analysis.

2.3 Mercury analyses

The samples were digested in concentrated H_2SO_4 and HNO_3 acid in an aluminium hot block (+85°C). The concentrations of Hg were measured using cold vapor atomic absorption spectrometry (Coleman MAS-50).

3. RESULTS

3.1 Sediments

The Hg concentrations of the surficial sediment of Lake Kirkkojärvi have decreased steadily since the beginning of the 1970's. From 2.1 $\mu g\ g^{-1}$ (dw) in 1972,

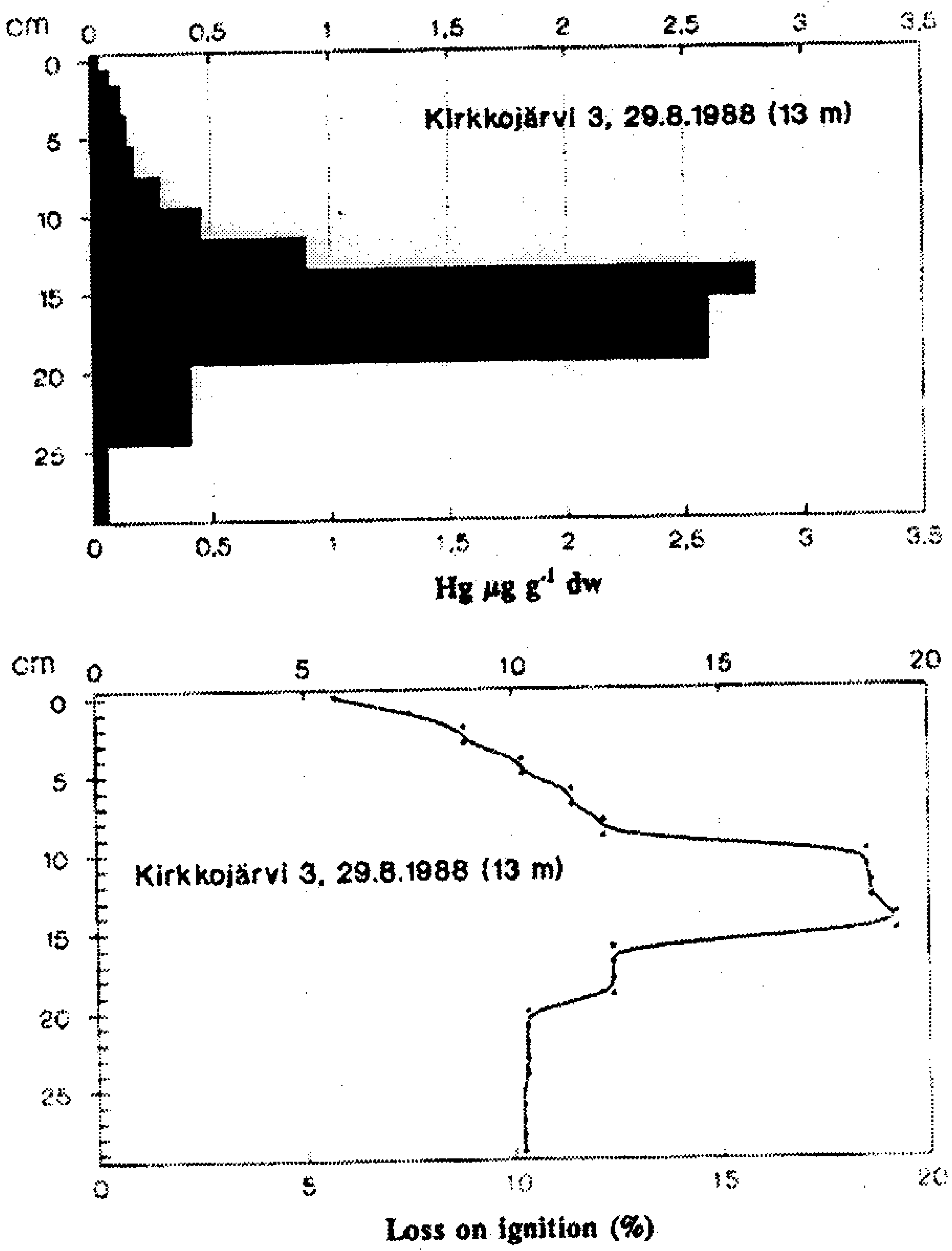


Fig. 2. Hg concentrations (μg g-1 dry wt) and loss on ignition (%) of sediments from Lake Kirkkojärvi (site 3, depth 13 m) collected in August 1988.

it decreased to 0.4 $\mu g\ g^{-1}$ in 1981 (Kokemäki River Water Protection Association, unpublished results).

The sediment profile from the deepest site (13 m) of Lake Kirkkojärvi collected in 1988 confirm that the old, highly polluted sediment layers are covered by more recent, unpolluted (0.02 $\mu g\ g^{-1}$) sediments (Figure 2). Near the mouth of River Pappilanjoki, where the current is strong, the surficial sediment contained 0.04 $\mu g\ g^{-1}$ while the surficial Hg concentration was somewhat higher (0.12 $\mu g\ g^{-1}$) in the semi-enclosed bay at site P (Figures 3 and 4).

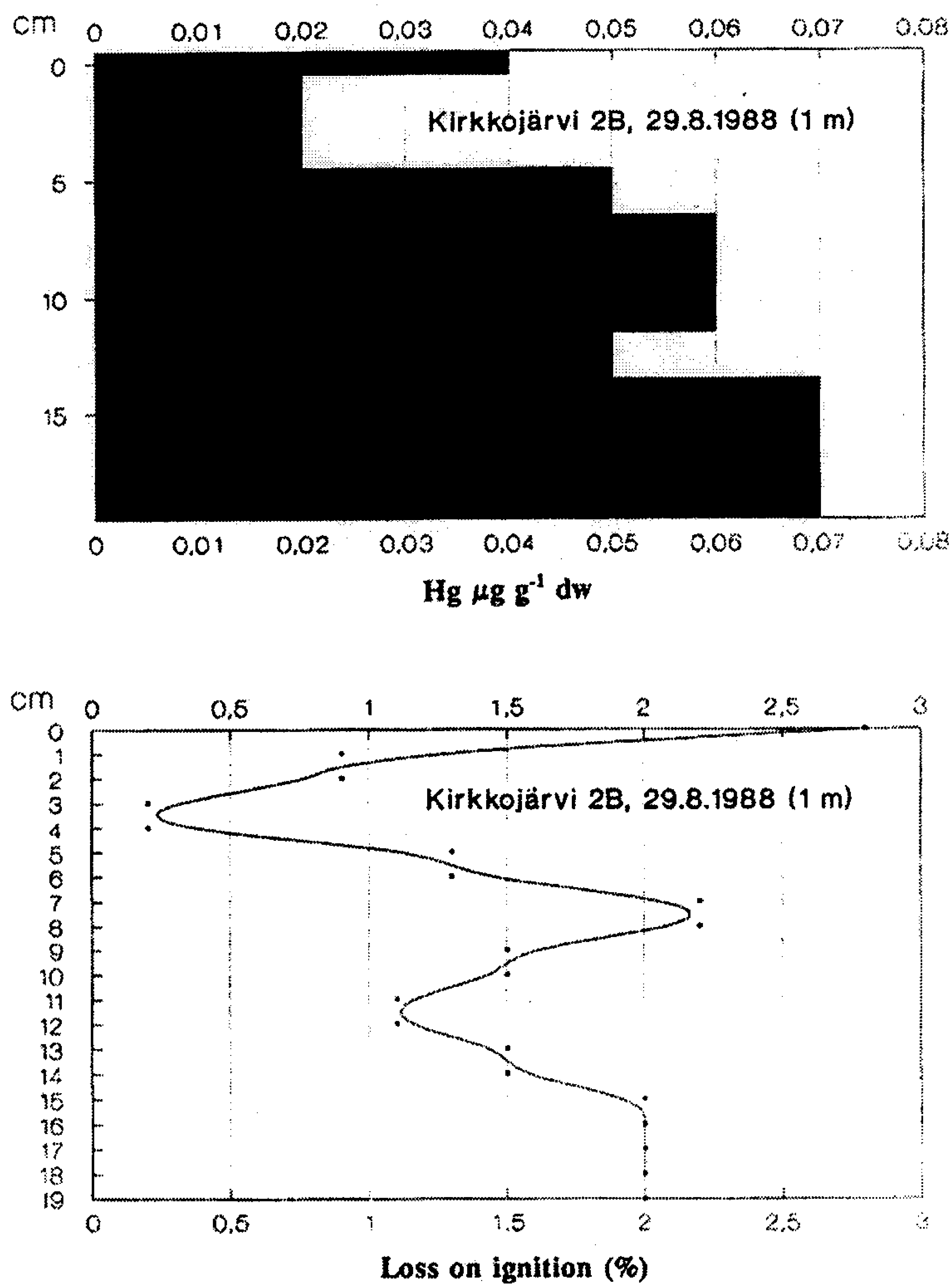
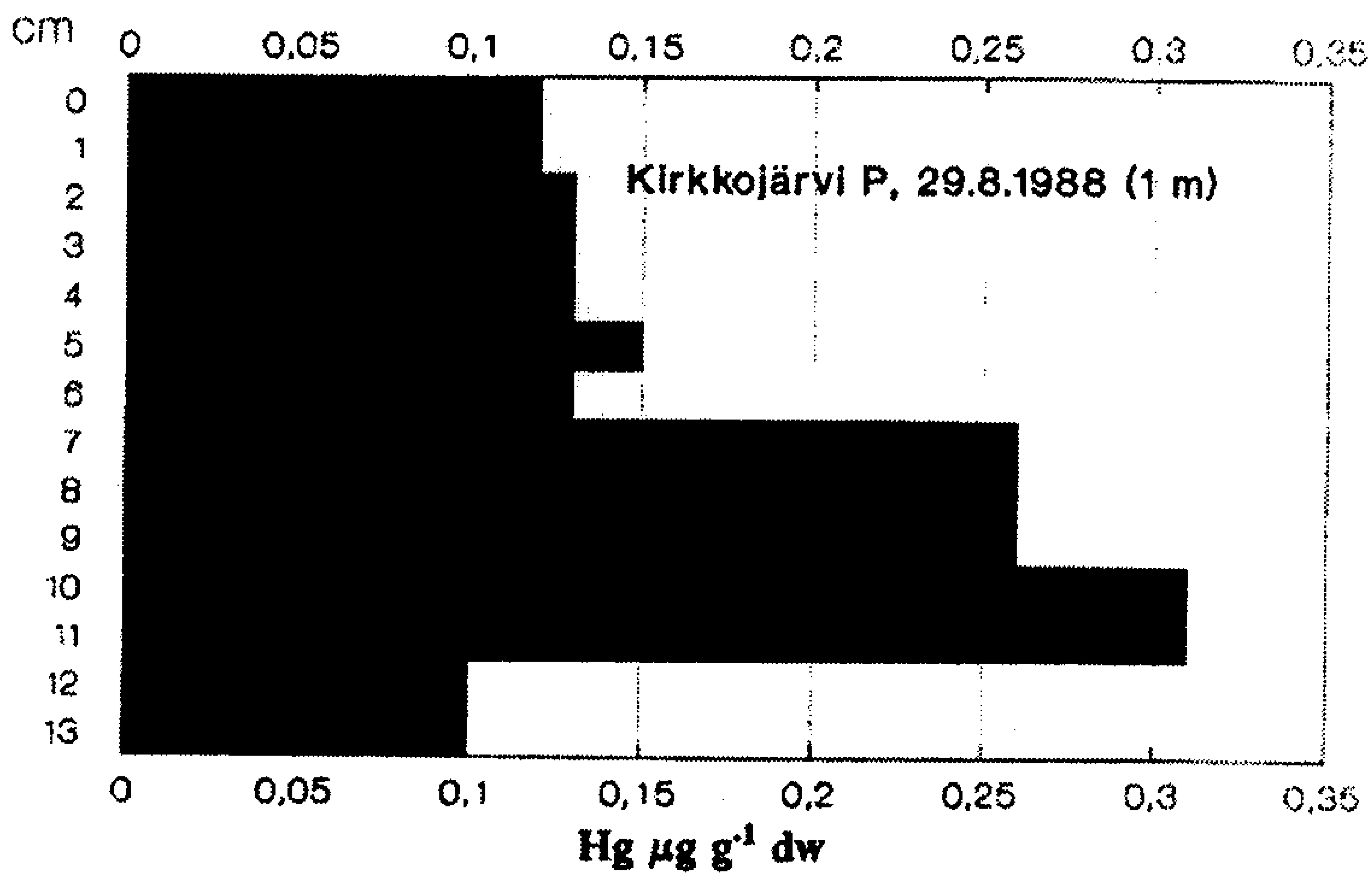


Fig. 3. Hg concentrations ($\mu g\ g^{-1}$ dry wt) and loss on ignition (%) of sediments from Lake Kirkkojärvi (site 2B, depth 1 m) collected in August 1988.



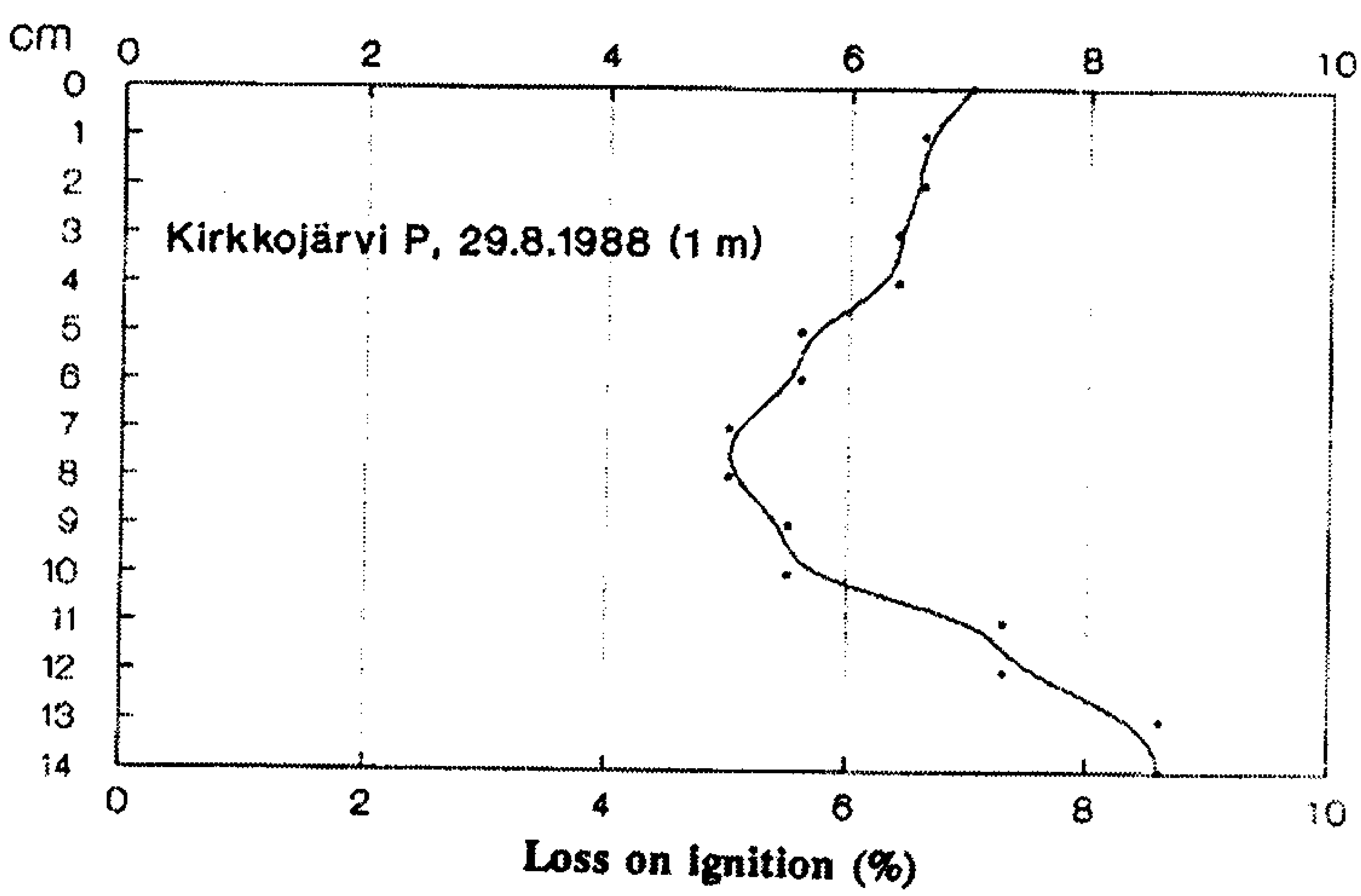


Fig. 4. Hg concentrations (µg g-1 dry wt) and loss on ignition (%) of sediments from Lake Kirkkojärvi (site P, depth 1 m) collected in August 1988.

3.2 Aquatic plants

Aquatic plants collected in the 1970's (Table I) were slightly elevated in Hg content as compared to unpolluted sites. In 1988 the Hg level was at the background level (*cf.* Lodenius, 1980). No differences were detected between the three sampling sites.

Table I. Hg concentrations (μg g-1 dry wt) of aquatic plants in Kirkkojärvi, Hämeenkyrö. The samples are collected in 1976 (Suominen *et al.*, 1977, 1978; Lodenius, 1980) and from three different sites in 1988.

	1976	**1978**	**1988** 2B	**1988** P	**1988** 3
Sparganium friesii	0.07		0.05	0.03	0.02
Potamogeton perfoliatus	0.07	0.03	0.05		0.04
Potamogeton obtusifolius	0.18			0.04	
Nuphar lutea, leaves		0.04	0.02	0.02	0.01
stems			0.02	0.01	0.01

3.3 Northern pike (*Esox lucius*)

Mercury concentrations differed between the pikes collected from the unpolluted Lake Kyrösjärvi and the lakes downstream. There was a slight decreasing tendency in Hg concentrations when moving downstream from the factory. This tendency was similar in the 1970's and 1980's (Figure 4). In the unpolluted lake the correlation coefficient for muscle Hg and fish weight was lower than in the polluted watercourses.

In the whole material the fish Hg concentrations (muscle, liver, kidney and gonads) correlated with both fish weight and length. There were no significant differences (except fish size) between males and females. Nor was there any significant difference in the size - Hg correlation between big and small specimens.

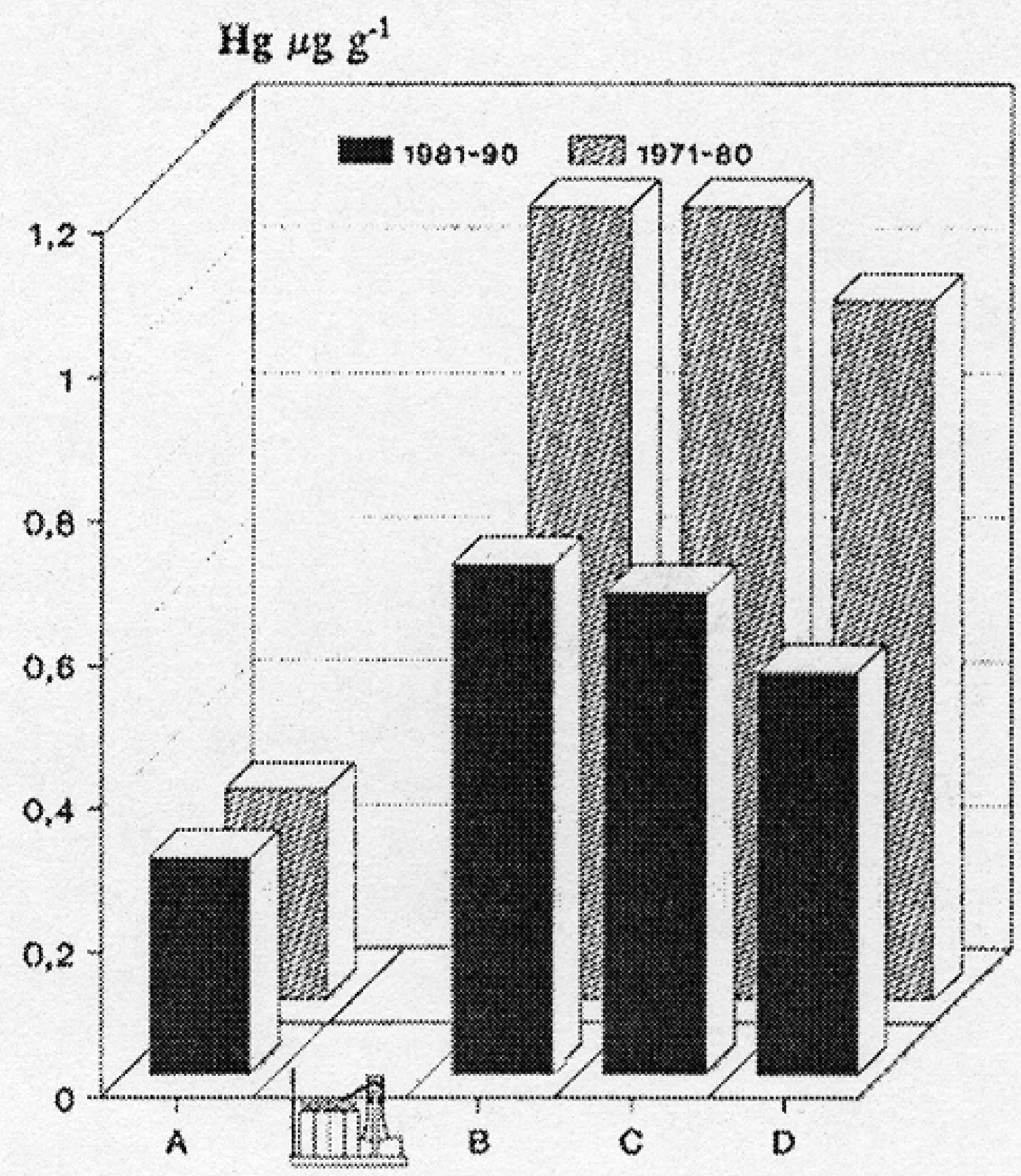


Fig. 5. Hg concentrations of a 1 kg pike in the 1970's and in the 1980's in the four subareas.

The Hg concentrations in pike normalized for fish size (1 or 4 kg) decreased during the 1970's, while the concentrations almost remained unchanged during the 1980's (Figure 6).

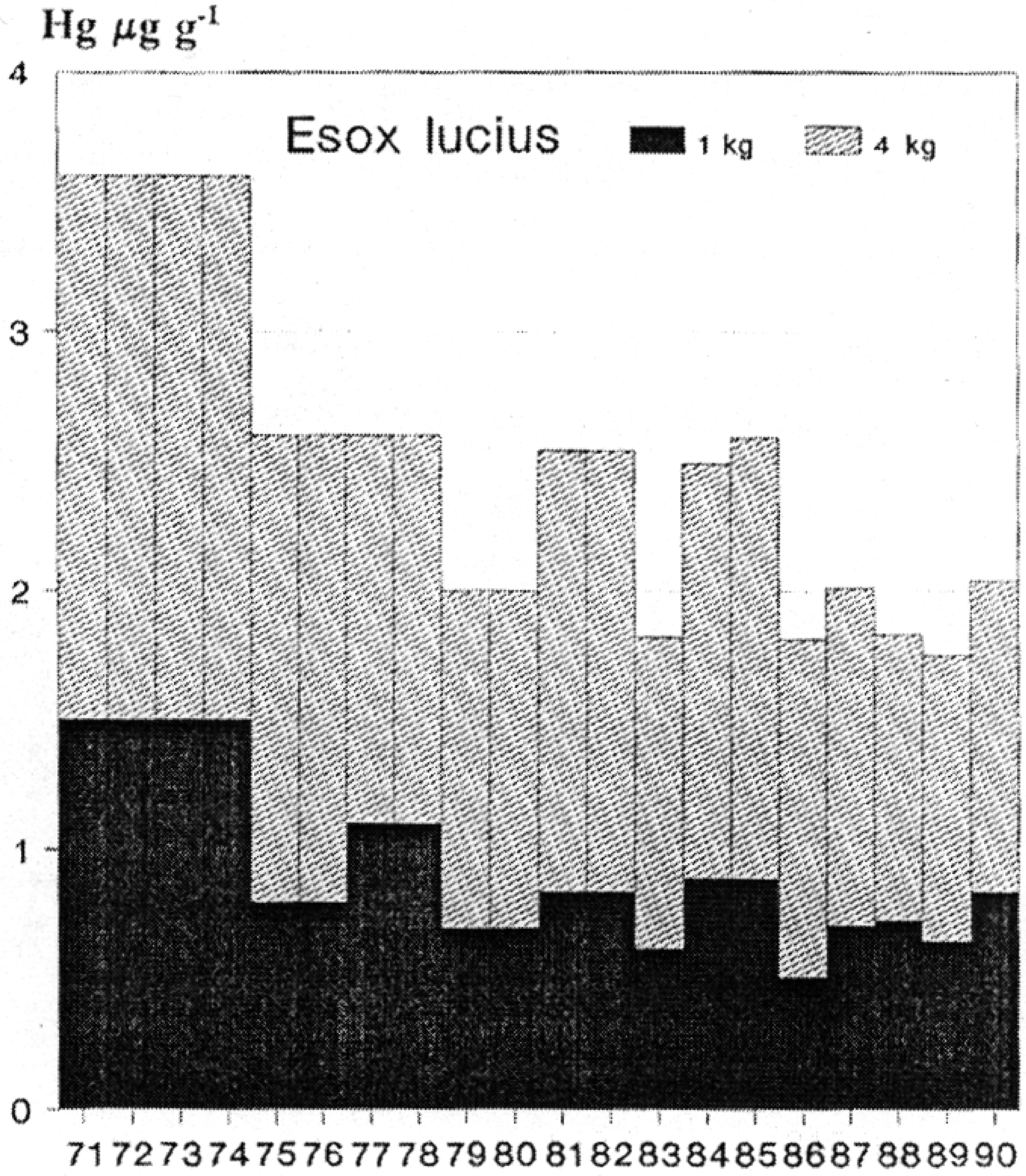


Fig. 6. Hg concentrations ($\mu g\ g^{-1}$ ww) of pike (1 and 4 kg) downstream from the factory (subareas B to D). Mean values were calculated for the 4-yr period 1971-74, the 2-yr periods 1975-76, 77-78, 79-80 and 81-82 and for single years 1983-90.

The Hg concentrations in pike usually decreased in the order: muscle > liver = kidney >> gonads (Figure 7).

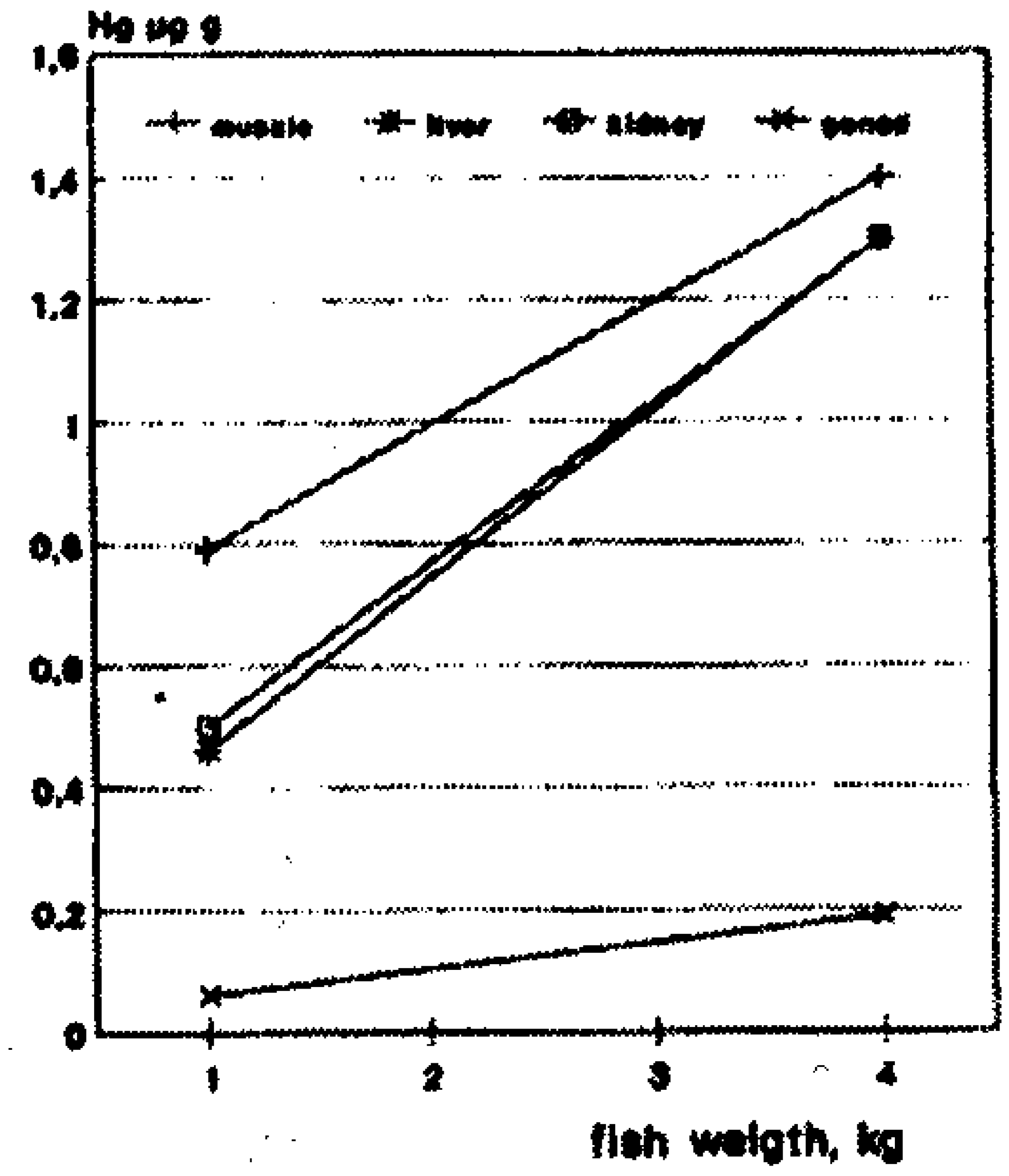


Fig. 7. Hg concentrations ($\mu g\ g^{-1}$ ww) of pike muscle, liver, kidney and gonads in relation to the fish weight.

Table II. Correlation matrix for some parameters studied.

	fish weight	fish length	muscle Hg	liver Hg	kidney Hg
fish length	0.929***				
N	234				
muscle Hg	0.666***	0.686***			
N	294	234			
muscle Hg ♂♂	0.520*	0.538*			
N	21	21			
muscle Hg ♀♀	0.806***	0.796***			
N	76	76			
liver Hg	0.750***	0.758***	0.913***		
N	129	129	129		
kidney Hg	0.686***	0.687***	0.771***	0.736***	
N	123	123	123	123	
gonad Hg	0.612***	0.644***	0.762***	0.669***	0.565***
N	72	72	72	72	71

5. DISCUSSION

The present background concentration of a 1 kg pike in Finnish lakes other than humic forest lakes has been estimated to 0.44 $\mu g\ g^{-1}$, which is significantly higher than the estimated preindustrial background value 0.18 to 0.33 $\mu g\ g^{-1}$ (Verta *et al.*, 1986; Verta, 1990).

The Hg level in fish in Hämeenkyrö still exceeds the background level. The clear decrease in the 1970's has slowed down or stopped. Sediment Hg concentrations are normally analyzed from deep water sedimentation bottoms while less attention is paid the littoral zone. No Hg should be released from the deep water sediment in Lake Kirkkojärvi. The littoral sediments are in more intensive contact with mixing forces in the lake and contain, at least in some parts of the lake, more Hg. These sediments may consequently be a factor contributing to the elevated Hg level in the watercourses downstream the factory.

Humic substances occurring in water and sediments may increase the release of Hg from the sediments in these polyhumic lakes (Miller, 1975). The Hämeenkyrö watercourse is rather eutrophic with a dense fish population. Intensive fishing may be one method to reduce the Hg level in the aquatic ecosystem.

From the development of Hg concentrations in pike it is impossible to make any exact predictions concerning the future development of the concentrations. It is however evident that no essential decrease in the Hg level will occur in the near future. A periodic monitoring of the situation is needed.

Acknowledgments

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