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SELENIUM AND MERCURY CONTENTS IN NORTHERN PIKE *(ESOX LUCIUS, L.)* OF FINNISH MAN-MADE AND NATURAL LAKES

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The selenium contents in liver and muscle of 107 Northern pike from Finnish man-made and natural lakes were analyzed and the relationship between selenium and mercury was investigated. The mean selenium concentrations in liver and muscle were in man-made lakes 1.07 mg kg^{-1} (w.w.) and 0.14 mg kg^{-1}, respectively and in natural lakes 1.11 and 0.30 mg kg^{-1}. The selenium concentration of liver or muscle did not correlate with the weight, age, growth rate or dry matter content of muscle or with the water quality data of lakes or the estimated selenium content of the drainage basin. The selenium content was the highest (1.84 mg kg^{-1}) in the livers of pikes from Lake Seinäjärvi, in contrast to the percentage of methyl mercury in the livers, which was the lowest (74 % of total mercury, compared with about 90 % in other lakes).

The selenium content did not explain the mercury contents in pike within the individual lakes, although in man-made lakes the selenium levels were lower and the mercury levels higher than in natural lakes. For both selenium and mercury concentrations the muscle:liver ratio was lower in man-made lake (Lake Kalajärvi, 0.14:1 and 0.87:1) than in natural lakes (0.30:1 and 1.44:1).

Differences between the levels and distributions of selenium and mercury in pikes of man-made and natural lakes could be partly due to interactions between these elements.

Index words: Selenium, mercury, fish, man-made lake, accumulation.

1. INTRODUCTION

When high mercury concentrations in predatory fish of Finnish man-made lakes have been investigated, the question of the possible role of selenium in the occurence of mercury has arisen. Both in laboratory and field experiments selenium has been shown to reduce mercury accumulation in fish and other aquatic organisms (Klaverkamp et al. 1983, Turner and Rudd 1983, Turner and Swick 1983, see also the results of Speyer 1980), despite the fact that Heisinger et al. (1979) reported the opposite with toxic concentrations of selenium. The ability of selenium to reduce the toxicity of heavy metals, e.g. mercury, is also well-known (Magos and Webb 1980).

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Selenium is an essential micronutrient of fish. However, in concentrations slightly higher than the optimum it is toxic when intake is from either water or food (Poston et al. 1976, Gissel-Nielsen and Gissel-Nielsen 1978, Hilton et al. 1980, Hudson et al. 1980). Food seems to be the dominant source of selenium for fish (Sandholm et al. 1973, Turner and Swick 1983), the assimilation effeciency in pike being about 30 percent (Turner and Swick 1983). The highest levels are found in the liver and kidney of fish (rainbow trout, Salmo gairdneri) (Hilton et al. 1982).

The aim of this study was to evaluate the hypothesis that low intake of selenium may be one reason for high concentrations of mercury in fish. Selenium levels in pikes of Finnish man-made and natural lakes were measured and the possible interaction between mercury and selenium was evaluated.

2. MATERIAL AND METHODS

The location of the lakes studied is shown in Figure 1. The physical and chemical characteristics of these lakes have been presented by Mannio et al. (1986). Fish for this study (Northern pike, Esox lucius L.) were caught by gill-nets and drum-nets mainly in the open water season. Fish were frozen after catching and stored at -20°C. After thawing, a cut of the muscle under the dorsal fin and the whole liver of each fish were taken. Each liver was homogenitzed. The samples were stored frozen before analysis.

Sediment samples were taken from the deepest parts of the lakes with a crust-freeze sampler (Renberg 1981). The top 2 cm of the profile was dried at room temperature and analyzed.

The total number of fish analyzed was 107. Fish from different lakes were chosen on the

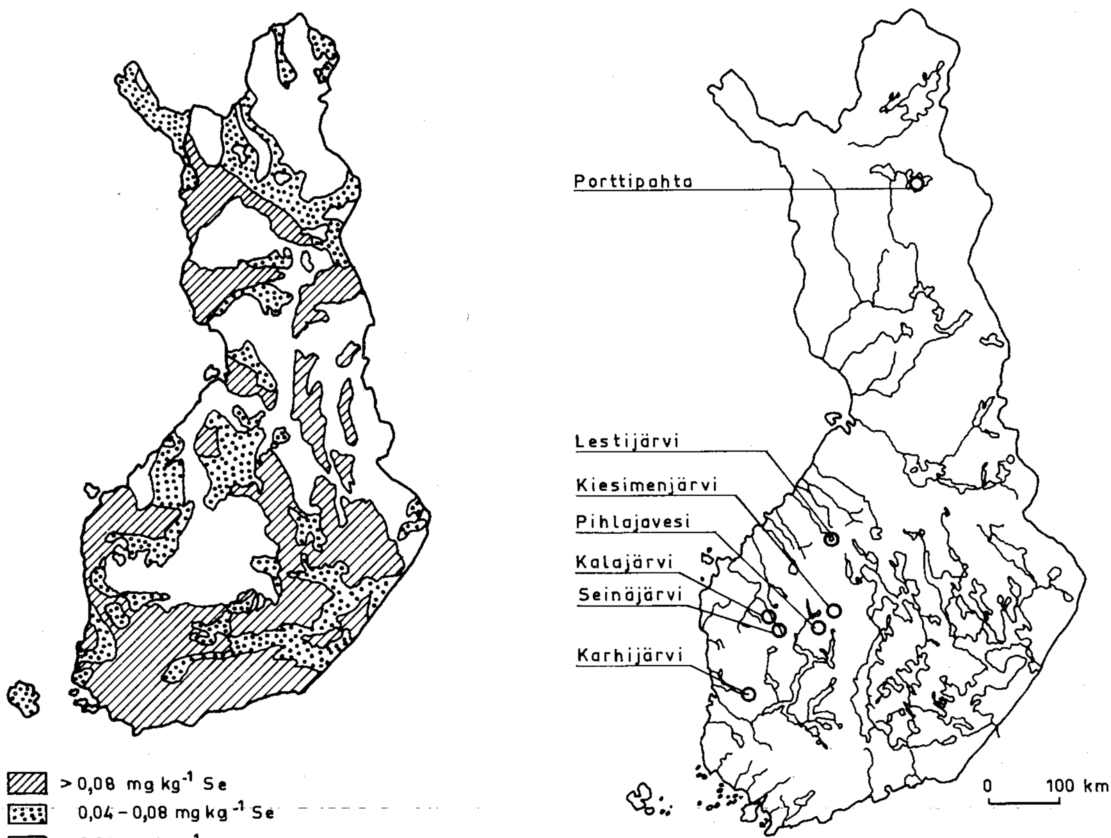

Fig. 1. The location of the lakes studied and the selenium content of Finnish soils (Koljonen 1975).

basis of similar size (mean weight varied between 520 g from Lake Seinäjärvi in 1982 and 1070 g from Lake Kalajärvi in 1980). In addition, fish with wide weight ranges from Lake Kalajärvi in 1983 were analyzed in order to study variation due to body weight (mean weight 2640 g). The number of samples was 5–19 from each lake and in each year.

Mercury samples were analyzed in the Department of Chemistry at the University of Jyväskylä and selenium contents in the Department of Food Chemistry and Technology at the University of Helsinki.

Fish muscle cuts of 1–4 g (wet weight), liver homogenates of 0.5–2 g (wet weight) or sediment samples of 0.3–1 g (dry weight) were transferred into 250 ml digestion tubes (Tecator). The samples were digested by boiling overnight in 10 ml of a 6:2:2 mixture of concentrated nitric, perchloric and sulphuric acids. Digestion was then completed with 5 ml of 30 % H_2O_2. To reduce any selenium(VI) to selenium(IV) 10 ml of 10 M HCl was added. After chelation with ammonium pyrrolidine dithiocarbamate (APDC) and extraction into methylisobutylketone, selenium was determined using a Perkin-Elmer 5000 atomic absorption spectrophotometer equipped with an HGA-500 graphite furnace, a model AS-40 autosampler and a model PRS-10 printer. The samples were analyzed in duplicate.

The accuracy of this method was tested throughout the analytical period using the National Bureau of Standards Reference Material Bovine Liver (SRM 1577). The validity of the method for biological samples has been thoroughly tested (Kamada and Yamamoto 1980, Kumpulainen et al. 1983) and a detailed method description is available elsewhere (Kumpulainen et al. 1983).

3. RESULTS

Mercury concentrations in both muscle and liver of pike were higher in man-made lakes (Lake Kalajärvi and Lake Porttipahta) than in natural lakes (others). By contrast the selenium concentration was higher in natural lakes, the difference being clear-cut in the muscle but rather small in the liver (Table 1). Differences were also observed between natural lakes in the average mercury and selenium concentrations of the liver and muscle of pike. A clear negative correlation between mercury and selenium concentrations in muscle was revealed when samples from man-made and natural lakes were plotted on the same figure (Fig. 2). However, the same relationship did not exist in the case of the liver samples. Among the samples from individual lakes the same correlation was not significant either in liver or in muscle. Mercury concentarations in Lake Kalajärvi pike varied from year to year, in contrast to the stable selenium level. The high mercury level of Lake Kalajärvi pike in 1983 was due to the fact that heavier fish were analyzed in this year than in earlier years (the positive correlation between weight and mercury concentration in muscle was very strong). The calculated values for muscle samples of »1 kg standard pike» in 1980, 1982 and 1983 were 1.9, 1.4 and 1.7 mg kg^{-1}, respectively.

Particularly in the case of liver samples, the highest selenium concentrations were in pikes from Lake Seinäjärvi (1.84 mg kg^{-1}, n=5). In contrast, only 74 percent of the mercury in the liver was methylated (n=8). In other lakes the corresponding value was about 90 percent as also in muscle samples from all the lakes (Table 1). A significant positive correlation existed between liver and muscle selenium concentrations (Fig. 3). In man-made lake (Lake Kalajärvi) the muscle:liver ratios of both selenium and mercury concentrations were lower than in natural lakes. In Lake Kalajärvi the muscle:liver ratio of mercury was 0.87:1 and in natural lakes 1.44:1 (significance of difference $p < 0.001$) and the selenium muscle:liver ratios were 0.14:1 and 0.30:1 in Lake Kalajärvi and natural lakes, respectively (significance of difference $p < 0.001$). The molar ratio between mercury and selenium contents in the muscle of pike was much higher in man-made lakes (Lake Kalajärvi and Lake Porttipahta, 4.72: 1) than in natural lakes (0.98:1).

According to the results of this study weight, age, growth rate or dry matter content in muscle did not explain either muscle or liver selenium concentrations. Likewise, seasonal variation could not be shown. Neither the water quality data nor the estimated selenium contents of the drainage basins (Fig. 1, Koljonen 1975) of the lakes studied correlated with the selenium contents of fish from these lakes.

Some sediment samples were also analyzed during this study (Table 2). Neither mercury nor selenium concentrations of the sediments had any correlation with those of pike from the same

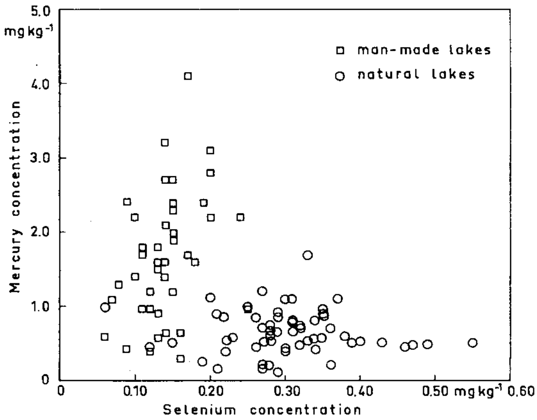

Fig. 2. Correlation between mercury and selenium concentrations of pike, whole material (r = −0.45, P < 0.001, n = 105).

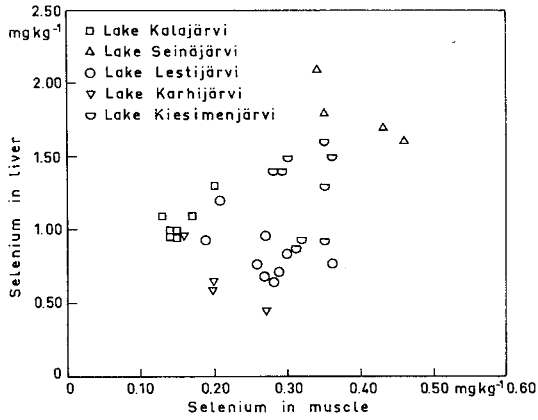

Fig. 3. Correlation between selenium concentrations in the liver and muscle of pike (r = 0.43, P < 0.01, n = 33).

Table 1. Mercury and selenium concentrations (wet weight) in muscle and liver of pikes from Finnish man-made and natural lakes. Signifigance of major differences: Between man-made and natural lakes, student's t-test, Hg-muscle, Se-muscle, Met-Hg%-muscle, Hg-liver P < 0.001, Se-liver and Met-Hg%-liver N.S. Between natural lakes, ANOVA Hg and Se in liver and muscle P<0.001.

	Total mercury mg kg^{-1} (ww)		Methyl mercury %		Total selenium mg kg^{-1} (ww)	
	muscle	liver	muscle	liver	muscle	liver
MAN-MADE LAKES						
Lake Kalajärvi 1980	2.34	..	..	..	0.16	..
Lake Kalajärvi 1982	1.47	..	89.3	..	0.14	..
Lake Kalajärvi 1983	2.60	3.18	95.2	89.5	0.16	1.07
Lake Porttipahta 1982	0.63	..	95.6	..	0.12	..
mean	1.66	3.18	92.3	89.5	0.14	1.07
S.D.	0.83	0.92	6.3	6.6	0.04	0.13
n	46	7	37	7	46	7
NATURAL LAKES						
Lake Seinäjärvi 1982	0.51	..	98.7	..	0.31	..
Lake Seinäjärvi 1983	0.65	0.42	93.6	73.8	0.37	1.84
Lake Pihlajavesi 1982	0.88	..	95.1	..	0.27	..
Lake Lestijärvi 1983	0.24	0.24	97.6	92.9	0.27	0.82
Lake Karhijärvi 1983	..	0.10	..	90.6	0.21	0.66
Lake Kiesemenjärvi 1983	0.76	0.45	93.3	91.6	0.31	1.26
mean	0.64	0.33	95.6	87.5	0.30	1.11
S.D.	0.30	0.17	3.5	9.5	0.09	0.46
n	61	33	61	33	65	29

Table 2. Mercury and selenium concentrations (dry weight) in the sediments from Finnish man-made and natural lakes in 1982. Differences between lake types are not significant.

	Total mercury (dw)			Total selenium (dw)		
	mg kg^{-1}	S.D.	n	mg kg^{-1}	S.D.	n
MAN-MADE LAKES						
Lake Kalajärvi	0.27	0.10	4	0.22	0.09	4
Lake Porttipahta	0.23	0.10	3	0.26	0.06	3
NATURAL LAKES						
Lake Seinäjärvi	0.24	0.01	3	0.39	0.07	3
Lake Pihlajavesi	0.38	0.07	2	0.41	0.16	2

lake. The number of sediment samples analyzed was, however, too small to allow definite conclusions.

4. DISCUSSION

According to Koljonen (1975), Finnish rocks and soils contain 0.03–0.10 mg kg^{-1} selenium (see Fig. 1), which is less than usually reported elsewhere. This study showed that the selenium contents of sediments are lower in Finland (Table 2) than in North America. In Lake Erie, concentrations varied between 0.10 and 0.75 mg kg^{-1}, (Adams and Johnson 1977) and in 11 natural lakes between 1.3 and 3.5 mg kg^{-1}, while in one man-made lake a concentration of 1.5 mg kg^{-1} was measured by Wiersma and Lee (1977). The concentration of 0.41 mg kg^{-1} observed in Clay Lake (Rudd et al. 1980) was about the same as in our samples from natural lakes.

The selenium concentration of pike found in this study was rather low. In North America, selenium concentrations in the muscle of 3 pikes were 0.42–0.58 mg kg^{-1} (Cappon and Smith 1981). In one lake rich in selenium (7.2 mg kg^{-1} in sediment), selenium concentrations were 1.1–3.0 mg kg^{-1} and in a lake poor in selenium (0.5 mg kg^{-1} in sediment) they were 0.20–0.62 mg kg^{-1} (Speyer 1980), i.e. at the same level as in natural Finnish lakes. Several fish species have been studied in Lake Erie, the selenium concentration in muscle being between 0.44 and 1.51 mg kg^{-1} (Adams and Johnson 1977). On the orher hand in the mercury contaminated Clay Lake in Canada, pike muscle contained only 0.18 mg kg^{-1} selenium (Rudd et al. 1980). In the Netherlands a selenium concentration of 0.13 mg kg^{-1} has been measured in pike (Luten et al. 1980), which is about the same as that observed in man-made lakes in our material. In Finland Kari and Kauranen (1978) measured 0.24 mg kg^{-1} selenium in the muscle of pike from the Bay of Bothnia, and Nuurtamo et al. (1980) the average of 0.22 mg kg^{-1} in freshwater pike.

In the liver, selenium concentrations were clearly higher than in muscle, as has also been reported in other studies (Tamura et al. 1975, Hilton et al. 1980, Turner and Swick 1983).

The lower value of the mercury muscle:liver ratio in natural lakes could be due to the higher selenium level in these lakes. Turner and Swick (1983) observed that addition of selenium to the diet of pike reduced the proportion of mercury in muscle.

The molar ratio of mercury to selenium (4.72:1) was high in man-made lakes. In the mercury-polluted Clay Lake the ratio was even higher (10:1, Rudd et al. 1980). In natural lakes the ratio (0.98:1) was nearly on the same level as elsewhere: Cappon and Smith (1981) reported 1.03–1.97:1, Luten et al. (1980) 1.97:1 and Kari and Kauranen (1978) 0.38:1.

Selenium concentrations in liver and muscle did not correlate with the weight or age of the fish, as Speyer (1980) also observed with pike and Takeda and Ueda (1978) with tuna (*Thunnus albacares*). Adam and Johnson (1977) reported a positive correlation between body weight and muscle selenium content in perch (*Perca flavescens*), but not in other fish species investigated.

The positive correlation between mercury and selenium concentrations found in some marine mammals does not appear to exist in fish (Tamura et al. 1975, Freeman et al. 1978, Kari and Kauranen 1978, Takeda and Ueda 1978, Luten et al. 1980, Cappon and Smith 1981). However, Luten et al. (1980) found a weak positive correlation between methyl mercury and selenium in the liver of perch. In this study, no relationship between the concentrations of these elements was found within individual lakes. Because the pikes of man-made lakes had high concentrations of mercury and low concentrations of selenium, whereas the opposite was true in natural lakes, a very clear negative correlation between the two elements could be calculated from the whole material. The reason for this may be a true biochemical interrelation in fish or a chemical interrelation in aquatic ecosystems, or alternatively the correlation may depend on some third factor. According to Rudd et al. (1980), selenium does not move through the ecosystem by either chemical or biological interaction with mercury. On the other hand Canadian experiments have clearly indicated that addition of dietary selenium reduces mercury accumulation in pike (Turner and Swick 1983). In our study the water quality parameters of natural lakes did not differ much from those of man-made lakes, whereas the possible differences in selenium contents (as indicated by this study) or in other sediment parameters could explain the variation of mercury and selenium concentrations in pike between man-made and natural lakes.

The high content of selenium and the low percentage of methyl mercury in pikes of Lake Seinäjärvi indicate the existence of a possible interaction between these elements in the fish liver.

According to results from striped mullet (Mullus barbatus) from the Northern Tyrrhenian Sea and The Black Sea, Leonzio et al. (1982) concluded that the total molar concentration of selenium and mercury has a linear correlation with the age of the fish, which implies the existence of Hg-Se -receptors, the number of which increases with the age of the fish. Our material did not support this conclusion in pike, because the sum of molar concentrations of selenium and mercury did not correlate better with weight or age of the fish than did mercury concentration.

The present investigation did not provide a conclusive answer to the question of the role of selenium in the high concentrations of mercury in pikes of Finnish man-made lakes. The high mercury concentration could be associated with low selenium level, but the inverse relationship between these elements in the two types of lakes could also be explained by the action of some third factor on both mercury and selenium.

However, the high concentration of selenium and the low percentage of methyl mercury in liver on the one hand and the difference between man-made and natural lakes in the mercury distribution between muscle and liver on the other indicate the existence of some kind of true biochemical interaction between these two elements. Thus higher selenium concentrations could be the reason for the higher muscle:liver ratio of mercury in natural lakes than in man-made lakes in which the selenium concentration was lower.

Addition of selenium to the ecosystem as a possible treatment for watercources in which the high mercury concentration in predatory fish is a problem has been suggested by Canadian research group (Rudd et al. 1983). However, as the Canadian group also concluded, more research must be carried out to investigate mercury-selenium relationships in fish and the aquatic cycle and the possible toxicity of selenium before practical preparations could be started. According to our results, the method could be worth studing also for the treatment of Finnish man-made lakes.

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TIIVISTELMÄ

Tässä tutkimuksessa on selvitetty suomalaisten tekoaltaiden ja luonnonjärvien haukien seleenipitoisuuksia sekä seleenin ja elohopean välisiä mahdollisia vuorovaikutussuhteita. Työ liittyy osana Vesihallituksen koordinoimaan laajempaan kalojen korkeiden elohopeapitoisuuksien syitä selvittävään tutkimukseen.

Haukien seleenipitoisuudet ovat merkitsevästi alhaisempia tekoaltailla kuin luonnonjärvillä. Tekoaltaiden kaloissa seleeniä on tuorepainoa kohden lihaksessa 0,14 mg kg^{-1} ja maksassa 1,07 mg kg^{-1} ja luonnonjärvissä vastaavasti 0,30 ja 1,11 mg/kg. Myös luonnonjärvien välillä keskimääräisissä seleenipitoisuuksissa oli eroja. Kalojen seleenipitoisuuksien vaihtelua ei selittänyt kalan paino, ikä, kasvunopeus tai lihaksen kuiva-ainepitoisuus eikä toisaalta järvien vedenlaatuparametrit tai valunta-alueen maaperän arvioitu seleenipitoisuus. Kalojen seleeni- ja elohopeapitoisuuden välillä ei voitu osoittaa korrelaatiota eri järvien sisällä. Sen sijaan verrattaessa eri järviä muodostuu selvä käänteinen suhde elohopean ja seleenin välille, sillä tekoaltailla on paljon elohopeaa ja vähän seleeniä, mutta luonnonvesissä toisinpäin. Samoin elohopean ja seleenin jakaantumisessa maksaan ja lihakseen on selvä ero tekoaltaiden ja luonnonjärvien välillä. Pitoisuussuhde lihas:maksa oli tekoaltaissa elohopealla 0,87:1 ja seleenillä 0,14:1 sekä luonnonvesissä vastaavasti 1,44:1 ja 0,30:1. Havainnot viittaavat seleenin ja elohopean välisen todellisen interaktion olemassaoloon. Kuitenkin todetut erot voivat johtua myös jonkun muun tekijän erisuuntaisesta vaikutuksesta tutkittuihin aineisiin. Maksassa tapahtuvaan vuorovaikutukseen viittaa Seinäjärvellä todettu maksan korkeampi seleenipitoisuus ja alhaisempi metyylielohopean prosenttiosuus kuin muilla järvillä.

Tutkimus ei anna lopullista vastausta seleenin merkityksestä suomalaisten tekoaltaiden kalojen elohopeapitoisuuksille. Kanadalaisissa tutkimuksissa on todettu vesistöön lisätyn seleenin vähentävän elohopean kerääntymistä kaloihin. Toisaalta seleenin kyky vähentää elohopean myrkyllisyyttä on yleisesti tunnettu. Niinpä Kanadassa onkin seleenin lisäystä ehdotettu erääksi elohopean vaivaamien vesistöjen hoitokeinoksi.

Tämän tutkimuksen pohjalta voidaan todeta, että menetelmän käyttökelpoisuutta suomalaisilla tekoaltailla kannattaisi ryhtyä tutkimaan.

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