

## Chlorinated hydrocarbons, PCBs and cesium isotopes in otters (*Lutra lutra* L.) from Central Finland

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The material consists of 11 otters found dead in Central Finland in the 1980s. The concentrations of many hazardous chemicals were low in the fresh weight of liver: alfa- and oxychlordane, HCB, lindane, DDT, DDD, DDE, PCB and dieldrin. The maximum value was 2.5 ppm of PCB. However, the lipid contents were higher, being on an average  $29 \pm 14$  ppm (maximum 150) of PCB. Mirex and toxaphene were not detected using a minimum detection level of one ng/g ( $=0.001$  ppm). The effect of the Chernobyl power plant explosion in spring 1986, 1300 km SSE of the study area, can be clearly seen in increased cesium contents. The residues of chemicals were not higher in the older animals.

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### 1. Introduction

Pollution is one of the causes of the decline in European otters. Chemical residues in otters (*Lutra canadensis*, *L. lutra*) have been analysed mainly in the USA, Great Britain and Scandinavia (see Mason & Macdonald 1986 for references). Only one specimen has been studied in Finland (Skarén & Kumpulainen 1986). The present paper reports another sample from Central Finland. Heavy metal contents will be published separately.

### 2. Material and methods

The material consists of 11 otters from 9 parishes in Central Finland (Fig. 1). Most of them (8) died in accidents, while the cause of death could not be checked in the rest (Table 1).

The following chemicals were analysed at the Institute of Chemistry, University of Jyväskylä (H. Ryösa): PCB (Clophen A 60), Lindane, DDT, DDD, DDE, alfa- and oxychlordane, hexachlorbenzene (HCB), dieldrin, mirex and toxaphene. In all, the tissue examined was that of the liver, except for otter number nine, in which the thigh muscle was used.

These chemicals were analysed using gas chromatographs. Tissue samples of about ten grams were weighed exactly and ground in four times the amount of sodium sulphate. They were then dried for two days in extract stockings. 2,4,6-trichlorobiphenyl was added as an internal standard. Then the tissues were extracted for 12 hours in a mixture of petroleum-ether:acetone:hexane:diethylether (9:5.5:2.5:1). The extracts were evaporated and the fat was weighed and extracted in hexane.

The samples were then divided into two parts and the fat was removed with deactivated alumina column. Hydrocarbons were divided into two fractions with Florisil-columns (activated at 230°C and deactivated in 1.25% water). The first fraction now included PCB, the internal standard, HCB, DDT, DDD and DDE. The second fraction included the possible mirex, dieldrin and toxaphene. To quantify the three last named chemicals, the level of lindane was measured, because it was used as internal standard in the second fraction.

The minimum detection level was one ng/g ( $=0.001$  ppm) of fresh weight for toxaphene and mirex, and 0.4 ng/g for the other chemicals.

Cesium contents were determined in the thigh muscle of two otters (numbers 10 and 11, Table 1). The muscle tissues were analysed by a 23.5% germanium detector using Marinelli-geometry, according to the ANSI/IEEE 680-1978 standard (J. Honkanen, Laboratory of Physics, University of Jyväskylä).

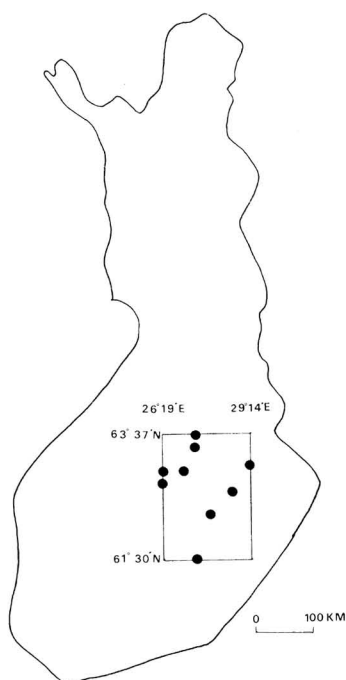


Fig. 1. Distribution of the present otter sample.

### 3. Results

#### 3.1. Cesium

Two old male otters were taken as a sample test for the cesium analysis. The first otter (number 10 in Table 1) died on 2 February 1986. The Chernobyl nuclear power plant explosion happened 26 April 1986 about 1300 km SSE from North Savo, the main study area. The second otter (number 11 in Table 1) died 28 on August 1986. Both otters were found in 'zone II', which in Finland sustained a moderate dose of radiation: the cesium-137 fallout was 3000–20 000 Bq/m<sup>2</sup> (Säteilyturvakeskus 1986a).

The cesium contents in these two otters were on 13 January 1987 as follows (Bq/kg):

	Cs-134	Cs-137
Otter no. 10	20	250
Otter no. 11	560	1250

Cesium 137 has a half-life of 30.1 years, cesium 134 2.1 years. The level of the latter isotope was low (8%) in otter no. 10. The radioactivity of this animal originated from former fallouts. But in otter no. 11 the

Table 1. The otters studied from North Savo, Central Finland.

Number	Locality	Date	Cause of death
1	Sonkajärvi	24.XII.85	car
2	Juuka	1.III.86	unknown
3	Iisalmi	13.III.85	unknown
4	Tuusniemi	2.XI.85	car
5	Pielavesi	10.VI.86	unknown
6	Tuusniemi	13.X.86	car
7	Leppävirta	19.III.85	fish trap
8	Iisalmi	10.II.86	fish trap
9	Ristiina	23.I.82	car
10	Keitele	2.II.86	fish trap
11	Vesanto	28.VIII.86	fish trap

Chernobyl effect is clear because the proportion of cesium 134 is now 45%.

#### 3.2. Chlorinated hydrocarbons

Table 2 shows the contents of the other chemicals. Almost all the levels are low, as they were in the only Finnish specimen analysed earlier (Skarén & Kumpulainen 1986). Mirex and toxaphene were not detected.

Chlordane was detected in four otters, in three of which only a trace was found. Also the concentrations of lindane and DDT were quite low. In every otter DDE, HCB and dieldrin, were detected, but in low contents, the average values being 0.07, 0.03 and 0.007 ppm (fresh weight) and 4.1, 1.2 and 0.2 ppm (lipid), respectively.

Only PCB levels in lipid were elevated in some specimens. The average was 29±14 ppm, but there were two high values. One was an old male with very little fat: 150 ppm (Vesanto). The other was a subadult female from Juuka. The liver fat percent was normal, but there was 58.1 ppm of PCB in the fat.

To summarize, it can be concluded that the otters now studied were relatively 'pure', at least concerning the chemicals now analysed. Even the oldest animals did not show increased levels (Student's *t*-test, *P* > 0.05, Table 2).

### 4. Discussion

Many variables may affect the results in residue analyses. Local differences in prey composition, the type of tissue analysed and the chemical methods used all affect the results. In seals (*Pusa hispida*), the

Table 2. Chemical contents of otters from North Savo. The animals are classed into young (1–2 years) and old (>2 years) (Skarén 1987). In all, the tissue analysed was that of the liver, except in otter no. 9 in which the thigh muscle was used. The proportion of fat was calculated from the tissue analysed. The average values concern the livers only. Oxy and alfa are chlordanes. + = a trace. DDD was found only in otters no. 5 (+) and no. 11 (8.0 ng/g). For oxy, alfa and lindane only fresh tissue values are given.

Otter no.	Sex	Age group	Weight (kg)	Fat (%)	Chemical contents (ppm in fresh tissue and lipid, respectively)									
					Oxy	Alfa	Lindane	HCB	DDE	DDT	PCB	Dieldrin		
1	f	young	2.5	3.0	+	–	0.8	0.021, 0.7	0.022, 0.7	0.0007, 0.02	0.262, 8.7	0.008, 0.3		
2	f	young	1.9	4.3	+	–	+	0.087, 2.0	0.088, 2.0	0.005, 0.1	2.50, 58.1	0.008, 0.2		
3	f	old	2.9	4.1	–	–	+	0.048, 1.2	0.135, 3.3	0.005, 0.1	0.680, 16.6	0.018, 0.4		
4	f	old	6	4.4	–	–	+	0.004, 0.1	0.071, 1.6	0.0009, 0.02	0.251, 5.7	0.003, 0.1		
5	f	old	5	1.6	–	+	–	0.022, 1.4	0.015, 1.0	+, +	0.153, 9.5	0.003, 0.2		
6	f	old	6.5	10.0	–	9.3	–	0.045, 0.5	0.111, 1.1	0.006, 0.1	0.772, 7.7	0.0006, 0.01		
7	m	young	3.5	5.1	–	–	+	0.034, 0.7	0.040, 0.8	0.003, 0.1	0.930, 18.2	0.009, 0.2		
8	m	young	4.9	1.1	–	–	+	0.012, 1.1	0.009, 0.9	–, –	0.082, 7.5	0.004, 0.3		
9	m	old	6	5.3	–	–	+	0.010, 0.2	0.026, 0.5	+, +	0.367, 6.9	0.004, 0.01		
10	m	old	7	3.6	–	–	0.9	0.020, 0.6	0.030, 0.8	+, +	0.321, 8.9	0.008, 0.2		
11	m	old	8.5	0.8	–	–	–	0.027, 3.4	0.225, 28.5	0.005, 0.7	1.20, 150	0.004, 0.5		
Mean	Young	(fresh tissue)						0.039±0.017	0.040±0.017		0.944±0.550	0.007±0.001		
±SE	Old	(fresh tissue)						0.028±0.007	0.098±0.032		0.563±0.165	0.006±0.003		
	Total	(fresh tissue)						0.032±0.007	0.075±0.022		0.715±0.230	0.007±0.002		
		(lipid)						1.2±0.3	4.1±2.7	0.2±0.1	29±14	0.2±0.04		

fat level and the organochlorine residues were highest in spring and lowest in early winter. The contents increase throughout life in males only, while females void part of the body burden when delivering and lactating (Helle 1985). More mercury seems to accumulate in the male than in the female otter (*Lutra canadensis*, O'Connor & Nielson 1980). The same was true in PCB levels in mink (*Mustela vison*) and river otter (Henny et al. 1981). The Hg-contents in *Lutra canadensis* did not correlate with age (Anderson-Bledsoe & Scanlon 1983).

#### 4.1. Cesium

When comparing results obtained in different areas, it should be kept in mind that even when using the best methods the results are accurate only within 10% (J. Honkanen, pers. comm.).

To my knowledge, the only earlier cesium-137 data from otters have been published in the USA. The maximum content of 785.1 Bq/kg (=21 219 pCi/kg) was measured in the muscle of *Lutra canadensis*, the average being 252 Bq/kg in the 'worst' area (Halbrook et al. 1981).

In Great Britain the otter scats radiated significantly more in 1986–87 than in 1985, before the Chernobyl explosion. A maximum value of 79 500 Bq/kg (dry weight) was measured after the explosion.

In 1985, the corresponding values were 0–7400 ( $M=640$ ) Bq/kg (Mason & Macdonald 1988).

In addition, radium 226 contents in the bones of *Lutra canadensis* have been measured in Canada near uranium mine sites (Wren et al. 1987a). The contents were small, mostly less than one pCi/g. The maximum value of a male was 466.6 Bq/kg (=12.6 pCi/g).

Cesium-137 accumulates especially in perch (*Perca fluviatilis*), which is one of the most important prey items of otters in the present study area (Skarén & Kumpulainen 1986, and unpublished results). Cesium-137 contents were 150–9400 Bq/kg in June–September 1986 in Finnish perches ( $n=39$ ) caught in the area which sustained a 'moderate' Chernobyl fallout (Säteilyturvakeskus 1986b). Perches in the 'worst' area had even 16 000 Bq/kg, and the consumption of small perch was totally banned (Laurila 1986). High values have been observed still in the first half of 1988: perches even 20 500 Bq/kg, pike (*Esox lucius*) 26 000 Bq/kg (Säteilyturvakeskus 1988). According to the recommendations of FAO, foods used for human consumption should not contain more than 500 Bq/kg Cs-137. In Finland the limit has been 1000 Bq/kg (Anonymous 1987a).

The average background level of radiation in Finland is about 2 mSv per year. Consuming 40 000 Bq of Cs-137 gives an additional dose of 1 mSv (Säteilyturvakeskus 1986b). Observable physical changes appear in man after obtaining 100 mSv per

year (Varteva 1986). Cesium 137 accumulates in gonads and blood-forming organs (see Halbrook et al. 1981). An acute dose of 6 Sv usually kills a man within two months (Edwards 1987). Thus in theory, if an otter consumes 400 kg of perch á 10 000 Bq/kg it would acquire 100 mSv radiation and possibly physical lesions within one year, if it consumes 1–2 kg of perch daily. In practice, otters eat other items, too. We know nothing about the risk levels of radiation in otters.

However, the Chernobyl explosion had few acute effects on otters in Finland.

#### 4.2. DDT, DDD, DDE

DDT and its metabolites were found in almost every British otter. More than 50 ppm was measured in the fat of three specimens (Mason et al. 1986), although DDT was banned in Great Britain in 1981 (Hider et al. 1982). The maximum value of 28.5 ppm of DDE in fat in the present sample is far less. The DDT contents have continuously decreased in the Baltic area in the 1980s (Pitkänen et al. 1987).

DDT was banned in the USA in 1972 (Hider et al. 1982), but even 136.7 ppm of DDE has still been found in otter adipose tissue (Halbrook et al. 1981).

#### 4.3. Chlordane

Chlordane was not measured in the British otters. Low contents have been found in the USA, for example, 0.12 ppm (wet weight) in only one otter out of 20 (Henny et al. 1981). The concentrations in fish in coastal SW Finland have been increasing (Pyysalo et al. 1984). According to Helle et al. (1985), during the last few years increasing levels of chlordanes and toxaphenes have been found in Baltic seals. However, the levels in the present study area were negligible (Table 2). Chlordane is seldom used in Finland, but air-borne fallout is possible. Restrictions in the use of chlordanes have been made in the USA since 1983 (Pyysalo et al. 1984).

#### 4.4. PCB

PCBs are among the most dangerous compounds. In a decreasing population in Oregon, USA, river otter livers contained on an average 9.3 ppm (males)

and 3.5 ppm (females) of PCBs (wet weight). The respective maximum values were 23 and 7 ppm (Henny et al. 1980).

In a healthy population in Louisiana, USA, PCB was found in only 10 (18 %) of the 57 otters in the  $\geq 3$ -year-old age group. The maximum wet weight value was 2.1 ppm in the liver of a young otter of about one year of age (Fleming et al. 1985). Likewise, in remote forest areas of Alberta, the maximum PCB concentration in the liver (wet weight) of 88 otters was 0.08 ppm of PCB (Somers et al. 1987).

In adipose tissue of otters from Georgia, USA, 0.6–66.7 ppm of PCB was found, the average being 7.4 and 9 ppm in different areas (Halbrook et al. 1981).

In the extractable fat of the liver more than 50 ppm of PCB was found in five British otters, the maximum being 232 in the liver and 300 ppm in muscle (Mason et al. 1986).

In Spain, the wet weight values of PCB were 2.4–2.5 ppm in otter livers (Hernandez et al. 1985).

Olsson et al. (1981) reported data on PCB in Scandinavian otters (in extractable muscle fat). The lowest levels were found in coastal N Norway: 1.6–30, mean 17 ppm. In northern Sweden the contents were 7.4–170 (mean = 52) ppm, and in the most polluted area of S Sweden 12–970 (mean = 220) ppm.

If we compare the present Finnish data with those mentioned above, it seems that the PCB levels here seem to be somewhere between those in N Sweden and N Norway. The two otters with higher contents (Table 2) may reflect local pollution. Thus, a factory has polluted the adjacent waters in Hämeenlinna, South Finland, so that on an average 2.85 ppm of PCB has been measured in pike in summer 1986 (Anonymous 1987b). Also the state of nourishment has an effect on the results. For example, if we imagine that the fat percents were the reverse in the two otters with the highest PCB contents in Table 2, the now 'lean' otter no. 2 from Juuka should have 316 ppm, and otter no. 11 from Vesanto 28 ppm, instead of 150 ppm of PCB. Reduction of the adipose tissue may be dangerous, because PCB can no longer 'hide' in the fat (M. Ahotupa, pers. comm.).

Wren et al. (1987b) fed minks with 1 ppm of PCB. Four months later, the liver wet weight content was 1.98 ppm in one male and the maximum value was 3.1 ppm after eating this food for eight months. It was concluded that if the wet weight levels of PCBs are 2–3 ppm in the livers of minks trapped, the viability of kits may be reduced in that population.

After consuming 3.3 ppm of PCBs for 66 days, female minks with mean PCB values of 86 ppm in extractable muscle fat had about a 50% reduction in the number of kits born per pregnant female (Jensen et al. 1977). In another test, reproduction of minks failed in animals having 1 ppm of PCB in the liver wet weight, while all the adults having an average content of  $12 \pm 11$  ppm of PCB, respectively died (Platonow & Karstad 1973).

We do not know the risk levels of PCB in otters, but it is possible that there are also some dangerous areas in central Finland, with generally viable otter populations.

#### 4.5. HCB

HCB is a petrochemical waste product, a contaminant in some pesticides, a fungicide and a breakdown product in lindane degradation. It is a global pollutant, but the effects in otters are not known. In Alberta's otters a maximum value of 0.02 ppm was found in the liver wet weight (Somers et al. 1987).

#### 4.6. Dieldrin

According to Chanin & Jefferies (1978), dieldrin was one of the most important causes of the decline in British otters, but the otter population did not recover after the voluntary restrictions in the use of cyclodienes (Mason & Macdonald 1986). The detection limit was 0.1 ppm in the report of Mason et al. (1986), and dieldrin was found in 16 otters with a maximum of 66.4 ppm in the liver fat in eastern Scotland. If the same detection limit had been used in the present sample, no dieldrin would have been found (Table 2).

In Oregon, dieldrin contents were low in fish: 0.01–0.04 ppm in the wet weight (Henny et al. 1981). Dieldrin was totally banned in 1975 in the USA and in 1981 in Great Britain (Hider et al. 1982).

#### 4.7. Mirex

Mirex was used in the control of red fire ants in southern USA for 15 years until it was banned in 1979. It caused cancer and birth defects in laboratory mice, and was also found in human tissues (Boraiko 1980). Mirex was found in the adipose tissue of about every third otter (*Lutra canadensis*) on a lowland area

in USA. The maximum value was even 169 ppm (Halbrook et al. 1981). Low levels of mirex have been detected in Baltic seals (Helle et al. 1985).

#### 4.8. Toxaphene

Toxaphene was one of the new insecticides created to replace DDT. More than 100 000 metric tons of toxaphene was produced in the USA in 1980, and as an airborne toxin it should spread over the world in increasing amounts (Pyysalo et al. 1984). This chemical has never been used in Finland, but in the 1970s and 1980s mean levels of 10–20 ppm (maximum 45 ppm) were found in the fat of Baltic seals (Helle 1985). In 1978, toxaphene contents of less than 0.01 ppm were found in pike samples from the southeastern and northernmost coastal waters of Finland (Pitkänen et al. 1987). It is not known why this chemical was not found in the present sample. A maximum of 64.5 ppm of toxaphene was found in the adipose tissue of *Lutra canadensis* (Halbrook et al. 1981).

It must be pointed out that the relatively 'good' state of the present study area does not necessarily hold true for other parts of Finland. The bulk of Finnish hazardous waste is produced in South Finland. In the year 1985, 'Ekokem' (the Finnish refining plant) received over 27 000 metric tons of hazardous waste (of which about 1300 metric tons were PCBs). Only 1.9% of this originated from Kuopio province, the main present study area (Tilastokeskus 1987). Nevertheless, the situation may be dangerous, as it has been estimated that 150 000 metric tons of hazardous waste are produced in Finland yearly. Nobody knows what happens to the bulk of this as only a fraction is brought to the 'Ekokem' (Anonymous 1987c). Possibly, it is no coincidence that the Finnish otter population seems to be the sparsest just on the southern and southwestern coast (Uhanalaisten eläinten ja kasvien suojelutoimikunnan mietintö 1986).

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