Environmental levels of PTS in Estonia

Ott Roots

LEVELS IN HUMAN MILK

Concentrations of Persistent Organic Pollutants (POPs) in humans are not as well researched as far as number of other organism. Breast milk is interesting for two reasons.
Results can be used to describe the mother`s exposure, and to assess the risk to the infant as a consumer.

The POPs content of mother`s milk, collected from mothers in the Tallinn region, has been studied by Roots (1986, 1996) in 1984. A comparison of the results of this study with those obtained in Estonia between 1971 – 1974 has indicated that DDT concent-rations tions in breast milk are 5 – 20 times lower than those between 1971 – 1974 (Table 1)

Table 1. Concentrations of persistent organic pollutants in Estonian human milk (Uibo,et. al., 1975; Roots, 1986,1996)

Year	mg/kg wet weight				DDEx100 / sDDT %
	DDT	DDE	sDDT	PCB	
1971	0,026 0.004-0,050	0.099 0,021-0,230	0,125		79
1974	0,021 0,002-0,080	0,063 0,008-0,180	0,084		75
1984			0,006 0,003-0,011	0,012 0,006-0,017	95

During the years 1974 – 1984, decomposition of DDT into its main metabolite DDE was increased in the breast milk from 79% to 95%.

The mean concentrations of total DDT and polychlorinated biphenyls (PCB) in human milk in Estonia in 1984 were relatively low compared with those in other countries .

It was found that an average daily intake of total DDT and PCB by children in the Estonia didn`t exceed the ADI (acceptable daily intake) value of 0,005 mg/kg pro-posed by the WHO for DDT and for PCB concentration 0,07 mg/kg, which caused " Yusho " disease in Japan in 1968 and in Taivan in 1979.

The dietary intake of DDT in 1969 was 36 mikrogr/day (Lutsoja, et. al.,1969). Six years after the total ban of DDT in Estonia in 1968, the content of its metabolite DDE in human milk rose up to 75 – 79%. The amounts of DDT and DDE consumed by newborn child-ren on the first week after birth were 5,6 mikrogram of DDT and 15,9 mikrogram of DDE per day. During the period of II – IV weeks after birth, the amounts were respectively 11,2 mikrogram of DDT and 35,4 mikrogram of DDE per day (Uibo, et. al., 1975).

In the early eighties those were 0,5 – 1,5 mikrogr/day (DDT) and 1,1 – 3,0 mikrogr/day (PCB) respectively.

PCDDs and PCDFs have been found in human milk samples in all over the world. It has been suggested that one of the largest sources of PCDDs and PCDFs in the environment is waste incineration. Other sources are eg. Automobiles, copper smelters, scrap reclama- tion in the iron and steel industry. Analysis of fish and dairy products indicates that the major exposure is via food (Mussalo, Lindström, 1995; Roots, 1986; 1996, 1999).

The sources of PCDDs and PCDFs vary in different countries. For example, in Estonia there are power plants which use oil shale as a fuel. It is mained in Northeastern Estonia. Industry in Narva and Kohtla - Järve as well as oil shale mines in the vicinity generates a lot of waste. In Finland, three municipal waste incineration centres have been in operation, but presently only one of them is in use. There are about twenty municipal waste incineration centres in use in Sweden, and in Norway municipal waste incineration cent- res are located in three places Estonia has no such centres (Mussalo, Lindström, 1995).

The data given in Table 2 indicate that the levels of PCDDs and PCDFs in human milk are similar in Estonia, Finland, Sweden and Norway. Results also show that the same isomers are found in the same proportions. Similarly, the average concentrations of PCBs in human milk in these countries have been reported to be at the same level .

Table 2. Levels of International Toxic Equivivalents (TEQ) in human milk in 1986 –1991 (pg/g on a fat weight basis) (Mussalo-Lindström, 1995)

	Country			
Compound	Estonia	Finland	Sweden	Norway
	1991	1986-87	1987	1987
I – TEQ	13,5 – 21,4	16,0 – 17,9	20,8 – 23,8	14,9 – 20,4

Exposure of the average population in the East Baltic region (Estonia, Latvia, Lithuania and St. Petersburg region of Russia) and current pesticide use is very low to chlorinated compounds seems lower than in most of western Europe,The low exposure of the general population is indicated by low concentrations of polychlorinated dibenzo-p-dioxins, dibenzofurans and biphenyls in milk fat (Table 3). Thus the overall risk caused by pesticide residues and persistent organic compounds in the Baltic countries and northwestern Russia is low, but local sites of concern exist (Tuomisto and Hagmar, 1999).

Table 3. Polychlorinated dibenzo-p-dioxins/dibenzofurans (PCDD/F) and the marker polychlorinated biphenyl (PCB) (sum of IUPAC 28, 52, 101, 138, 153 and 180) concentrations in human milk in 1993 (WHO, 1996; Tuomisto and Hagmar, 1999)

Country	PCDD/F (pg TEQ/g fat)		Marker PCB (ng/g fat)	
	Urban area	Rural area	Urban area	Rural area
Austria	10,7	10,9	381	303
Belgium	26,6	20,8	261	276
Canada	13,4	10,8	137	86
Croatia	13,5	8,4	220	218
Czech Republic	18,4	12,1	1069	532
Denmark	15,2	-	209	-
Finland	21,5	12,0	189	134
Germany	16,5	-	375	-
Hungary	8,5	7,8	61	45
Lithuania	13,3	14,4	322	287
The Netherlands	22,4	-	253	-
Norway	10,1	9,3	273	265
Pakistan	3,9	-	19	-
Russia	15,2	5,9	197	102
Slovakia	15,1	12,6	1015	489
Spain	25,5	19,4	452	461
Ukraine	13,3	-	191	-
United Kingdom	17,9	15,2	130	131
St. Petersburg Region of Russia*	16,4	-	190	-
Estonia*	14,4	12,4	103	136

- Unpublished data analyzed simultaneously with WHO study (courtesy of Professor Terttu Vartiainen)

Estonia: Tallinn (Urban area) and Rakvere (Rural area).

References

Holoubek, I., Kocan, A., Holoubkova, I., Hilscherova, K., Kohoutek, J., Falandysz, J. and Roots, O. 2001. Persistent, Bioaccumulative and Toxic Compounds in the Central and Eastern European Countries – The – State – Of –The – Art Report – Human Exposure. Arh. Hig. Rada Toksikol., v. 52, pp. 181 – 215

Lutsoja, H. and Tamm, O. 1969. Pestitsiidid ja sanitaarne olukord Eestis. Nõukogude Eesti Tervishoid, No. 3, pp. 171 – 173 (in Estonian).

Roots, O. 1986. PCB and chlororganic pesticides detected in human milk in Estonia in 1984. Estonian Health, No. 6, pp. 419 – 422 (in Estonian, summary in English and Russian).

Roots, O. 1996. Toxic Chlororganic Compounds in the Ecosystem of the Baltic Sea, Ministry of the Environment of Estonia, Environment Information Centre, 144 p. (ISBN 9985 – 9072 – 0 –)
Uibo, M., Kask, V., Ilmoja, K. and Ivask, M. 1975. The excrection og chlorinated hydrocarbons in human milk, its Specific character and dynamics in the Estonian SSR. Proc. Of the Tartu University, XXX, v. 373, pp. 107 – 111 (in Russian, summary in English).

POLYCHLORINATED BIPHENYLS AND CHLORORGANIC PESTICIDES. ASSESSMENT OF HEALTH RISK ASSOCIATED WITH THE CONSUMPTION OF FOOD

The main reason for analysing toxic compounds from our surrounding (including food) is the will to determine how dangerous they are to people and their living environment.

The Baltic Sea is a sink for a multitude of environmentally hazardous substances. The potential effects of these substances on both humans and the environment are still rather poorly known and only a small faction of man-made substances entering the sea are properly monitored.

Risk assessment is essentially the process of decision how dangerous a substance can be. The first step in the process of risk assessment is to identify and qualitatively describe the hazard to be assessed. Next, the level of exposure to the hazardous entity is estimated, along with the response of the organisms in question (usually humans, but sometimes, as when evaluating ecosystems, other species) to different dose levels, using the best scientific data available.

At the international level, the need for a global assessment of levels of chemicals in food and in the total diet led to the establishment of the Global Environment Monitoring System – Food Contamination Monitoring and Assessment Programme (GEMS/Food) in 1976.

Food contamination monitoring is an essential component of ensuring the safety of food supplies and managing health and environmental resources, since it provides information on the levels and sources of contaminants of foods, on the amounts of contaminants ingested by humans and on trends in contamination levels (Roots, 1996; 1999).

There is mounting evidence, that human exposures and health risks of toxic chemicals as Dioxins/Furans, PCB, lead, cadmium, mercury, etc., have been brought under control in many developed countries, but generally not in the developing world as a whole.

Polychlorinated biphenyls and chlororganic pesticides were used as tracers for nonpolar organic contaminants cycling in the Baltic Sea. Environmentally hazardous substances, such as polychlorinated biphenyls (PCB) and chlororganic pesticides (COP), are acutely toxic, persistent, and bioaccumulative (i.e. they become concentrated in food chains to reach toxic levels).

The species chosen as test organisms and the sampling procedures recommended for monitoring harmful substances in biota are intended to provide a picture of the levels of harmful substances in the studied organisms and to determine trends in their levels over time. Helsinki Commission (HELCOM) marine monitoring programme " COMBINE " has selected perch (Perca fluviatilis) and flounder (Platichtys flesus) as study organisms.:

It is known that marine food is not the only route of human exposure and any overall risk assessment must take this into account .
The following questions need a proper answer:
- what is the amount of fish consumed per day that could be taken as basis for our calculations?
- from which standards shall we proceed in evaluating the risk poised by toxicants?

When considering the possible effects of consuming seafood, it is necessary to characterize communities and individuals according to the amounts they consume, since communities in different parts of the world are likely to show big differences in their consumption of seafood. For example, based on FAO food balance data, the European regional average consumption of seafood (fish and shellfish) is 60 g per person per day and in East Asia 79 g per person per day. The figure used to represent high consumption is 150 g per person per day (Reports and Studies, 1991).

The joint FAO/WHO Expert Committee on Food Additives (JECFA) generally sets the ADI (the acceptable daily intake) of a food additive on the basis of the highest "no-observed-effect" (NOEL) level in animal studies.

ADI is the daily dosage of a chemical, which, during an entire lifetime, appears to be without appreciable risk on the basis of all the facts known at the time. Where an ADI has been established for a particular pollutant it will be noted whether the intake from seafood is likely to be a substantial part of the ADI (Reports and Studies, 1991).

NOEL is the risk that may be incurred by especially susceptible groups, such as children and old people. ADI and NOEL values used in our work are based upon the literature data (FAO/WHO, 1971, 1978, 1985; Reports and Studies, 1991).

For each level of seafood consumption, the health (carcinogenic) risk from marine pollutants varies according to their concentration. This is turn varies from place to place and from time to time. We took as base for assessing the health risk organochlorine concentrations in the muscle tissue of fish caught in Estonian coastal areas (Roots, 1996;1999, 2001; Roots & Kakum,1999)..

DDT and its metabolites are some of the most frequently reported organochlorine contaminants in the environment. An overall NOEL for the toxicity of DDT of 0.25 mg kg^{-1} body weight per day in humans and ADI of 0.02 mg kg^{-1} body weight was set by the Joint Meeting on Pesticide Residues (JMPR) in 1984 (FAO/WHO, 1985; Reports and Studies, 1991).

If a person weighing 60 kg consumes 150 g d^{-1} of fish containing 20 μg kg^{-1} of DDT and its metabolites, the daily intake will be 0.05 μg kg^{-1} body weight. This would constitute 1/400 of the ADI (Roots, 1996).

If we look at the amounts of DDT and its metabolites, obtained with food (fish), we can see **that even the maximum levels of organochlorines** , **which we took as baseline**, found in the organism of Baltic fish (at the coastal area of Estonia) do not represent any health risk, as they are lower than the standards set by the WHO (table 1) . Besides, our calculations were made on the ground of 150 g of edible fish per day, compared with the European average of 60 g per day (Roots, 1999).

Hexachlorocyclohexane (HCH). Technical HCH contains a number of isomers of which gamma-HCH (lindane) is commonly used as an insecticide. In the Baltic Sea the concentrations of all three isomers (alpha, beta, and gamma) are similar (Roots, 1996).

If a 60 kg person eats 150 g d^{-1} fish containing 60 μg kg^{-1} (alpha-, beta-, and gamma-HCH), this will constitute 1/53 of the ADI (for gamma-HCH 0.008 mg kg^{-1} body weight was set by the JMPR). No ADI was established for the other isomers or for technical HCH.

Aldrin+Dieldrin. These insecticides are not a problem for the Baltic Sea. If a 60 kg person consumes 150 g d^{-1} fish containing 0.5 μg kg^{-1} (max) of aldrin plus dieldrin, the daily intake will constitute 1/80 of the ADI (0.0001 mg kg^{-1} body weight) (Roots, 1996).

Chlordane. The main components are alpha- and gamma-chlordane and the most toxicologically significant metabolite is oxy-chlordane.

If 150 g of fish containing 4 μg kg^{-1} gamma-chlordane plus oxy-chlordane (Roots & Aps, 1993; Roots, 1996) a 60 kg person is consumed daily by eats, this will constitute 1/50 of the ADI (0.005 mg kg^{-1} body weight).

Polychlorinated biphenyls (PCB). Human health studies of "Yusho" disease in Japan (1968) and in Taiwan (1979) showed that contamination of food with PCB from an industrial source can be a cause of severe human diseases.

If a 60 kg person eats 150 g d^{-1} of fish containing 20 µg kg^{-1} PCB, this will amount to 0.05 µgkg^{-1} body weight per day. This is about 1/3000 of the estimated average daily intake in the accident in Japan in 1968 (Reports and Studies, 1991), which involved about 1000 persons, and 1/800 of the NOEL of 0.04 mg kg^{-1} body weight per day for monkeys.

The comparison has so far been based upon the standards of the World Health Organisation, but ever since the year 2000 we have been able to apply **Estonian permissible limiting standards.**

On the 12th of January 2000 the Estonian Government passed a regulation number 14 (Enu-meration of permitted pollution substances in food and enactment of limiting standards by food groups), which set the permissible limiting standards for sea products. Sea products include seaweed, cancroids, molluscs and other invertebrates, amphibians and reptiles living in the water as well as the food that contain them.

As in the rest of the world, higher demands are set on the food of infants in Estonia. In other words, the food of infants should contain less toxicants than the food of adults.

These are the Estonian permissible limiting standards for fish products:

Polychlorinated biphenyls (PCB) content (sum of isomers IUPAC 28, 52, 101, 118, 138, 153 and 180) **should not exceed 2,0 mg/kg per fish.**

The content of DDT and its metabolites **should not exceed 0,2 mg/kg per fish (in case of sea fish) and 2,0 mg/kg per fish (in case of sturgeon, salmon, fatty herring).**

The content of hexachlorocyclohexane (sum of three isomers) **should not exceed 0,2 mg/kg (in sea fish as well as in meat of sea animals).**

All the above-mentioned standards are set for the food of adults.

The fish-food of **infants** should not contain residues of any plant protection products that exceed the level of 0,001 mg/kg per fish. The permissible limiting standard for PCBs is 2,0 mg/kg per fish.

This work does not deal with the permissible limiting standards set for the liver of fish.

SUMMARY

At present time the contents of toxic chlororganic compounds (DDT; PCB and HCH) analyzed in the perch and flounder of Estonian coastal waters remain below standards established by FAO/WHO and Estonian permissible limiting standards in food, in which case the content of toxicants in the food does not cause symptoms of illness in case of people (table 4).

REFERENCES

FAO/WHO. 1971. Evaluations of Some Pesticide Residues in Food. WHO/Food Add/71. 42. Rome.

FAO/WHO. 1978. Pesticide Residues in Food – 1977 Evaluations. FAO Plant Production and Protection Paper 10 Sup. Rome.

FAO/WHO. 1985. Pesticide residues in Food – 1984 Evaluations. FAO Plant Production and Protection Paper 67. Rome.

Reports and Studies. 1991. Review of potentially harmful substances: Carcinogens. – WHO, 46.

Roots, O. & Aps, R. 1993. Polychlorinated biphenyls and organochlorine pesticides in Baltic herring and sprat. – Toxicol. Environ. Chem., **37**, 3–4, 195–205.

Roots, O. 1996. Toxic Chlororganic Compounds in the Ecosystem of the Baltic Sea. Estonian Environment Information Centre, Tallinn, 144p. (ISBN 9985-9072-0-5).

Roots, O. & Kakum, T. 1999. PCB and Chlororganic Pesticides in the Ecosystem of the Matsalu Bay. In.: Loodusvaatlusi 1997 – 1999 (Eds. E. Mägi & K. Kaisel), Matsalu Looduskaitseala, Lihula, 142-154 (in Estonian).

Roots, O. 1999. The Effect of Environmental Pollution on Human Health in the Baltic States (Assessment and Regional Differences), Tallinn, 120p. (ISBN 9985-884-13-3).

Roots, O. 2001. Halogenated environmental contaminants in fish from Estonian coastal areas. – Chemosphere, v.43, 623 – 632.

POLYCHLORINATED BIPHENYLS AND CHLORORGANIC PESTICIDES IN BUTTER ORIGINATING FROM ESTONIA

The main reason for analysing toxic compounds from our surroundings (including food) is the will to determine how danderous they are to people and their living environment.

Concentrations in food (fish, butter, etc.) reflected differences in the propensity of PCB congeners to undergo long range atmospheric transport from global source regions to Estonian West-Estonian Archipelago Biosphere Reserve (Agrell, et al., 2001).

In the studies (Katantzi, et al., 2001; Weiss, et al., 2001) authors explosed the use of butter as a sampling matrix to reflect the regional and global scale distribution of chlororganic compounds in food.

Four butter samples from Estonia were collected, during the period November, 2001. All samples were kept in freezer at –20 C , until investigated . The samples consisted of 200 gram and the butter was bought in ordinary stores (Table 1).

Quantitative determination of chloorganic compounds was conducted by accreditated laboratory. The accreditations cover PCB analyses of samples from water and fats of animal or plant origin. By the (Weiss, et al., 2001) one butter sample cannot be said to be representative for butter concentrations from the country.

Table 5. Polychlorinated biphenyls and chlororganic pesticides (ng/g fat) concentrations in butter (Roots, 2002++)

Compounds	Number of samples			
	S 1	S 2	R 1	R 2
a-HCH	0,9	1,2	1,1	0,8
g-HCH	0,9	2,3	0,8	1,6
HCB	5,2	5,0	4,7	3,7
p,p`DDE	3,4	3,2	3,4	2,5
p,p`DDD	0,4	0,2	0,2	0,8
p,p`DDT	4,3	2,6	1,7	1,7
sumDDT	8,5	6,4	5,7	5,4

IUPAC 28	2,56	2,33	2,11	0,71
IUPAC 52	0,64	0,67	0,48	0,29
IUPAC 101	0,99	0,63	0,97	0,37
IUPAC 118	1,32	1,10	0,79	0,42
IUPAC 138	0,21	1,37	0,52	0,19
IUPAC 153	2,20	1,64	0,42	2,90
IUPAC 180	0,85	0,67	0,20	0 ,27
SumPCB – 7 isomers	8,77	8,41	5,49	5,15
IUPAC 126	0,23	0,20	0,31	0,41
IUPAC 105	0,50	1,22	0,30	0,57
IUPAC 170	0,95	0,88	0,22	0,35

Three samples of butter from the Baltic region of the Russian Federation were forwarded the Greenpeace Research Laboratory for analysis of dioxins/furans and dioxin-like (ortho and non-ortho PCBs). The highest concentration was found in one of the two butter samples drawn from Leningradskaja area (St. Petersburg). The contribution to overall TEQs from the non-ortho PCBs was greater than that from dioxins in all three Russian butters. The range of total TEQs (1.18-3.25 pg/g lipid) was again similar to the range previously reported for European butter, nothing however that only the butter previously analysed from Spain contained higher concentrations than the most contaminated butter from Leningradskaja (St. Peters-
Burg). Inclusion of the contribution from ortho PCBs in the calculation would increase total WHO-TEQs for the Russian (Baltic Region- St.Petersburg, Gatchina and Poleski areas) butters by 15-25% (to the range 1.40-3.74 pg/g lipid)(Santillo, et al., 2001).

References

Agrell, C., Larsson, P., Okla, L., Bremle, G., Johansson, N., Klavins, M., Roots, O.& Zelechowska, A. 2001. Atmospheric and River Input of PCBs, DDTs and HCHs to the Baltic Sea. A System Analysis of the Baltic Sea (Ed. Wulff, F., Rahm, L. & Larsson, P.). Ecological Studies, Springer Verlag , v. 148, 149-175.

Kalantzi, O., Alcock, R., Johnston, P., Santillo, D., Stringer, R., Thomas, G. and Jones, K. 2001. The Global Distribution of PCBs and Organochlorine Pesticides in Butter. ENVIRON. SCI. & TECHNOL, v…., No. …, pp. ….

Roots, O. 2002.++.The project was financially supported by the John D. and Catherine T. MacArthur Foundation.

Santillo, D., Stringer, R. & Johnston, P. 2001. Concentrations of PCDDs, PCDFs and dioxin-like PCBs in three samples of butter from the Baltic region of the Russian Federation – Greenpeace Research Laboratories Technical Note 08/2001, 6p.

Weiss, J., Päpke, O. & Bergman, A. 2001. PCDDs/PCDFs related contaminants in butter originating from 39 countries world wide. DIOXIN2001, South-Korea.

Table 4: Comparison with ADI and NOEL.

Väinameri region 1997 (Roots and Kakum, 1999 **)**			
Perch (Perca fluviatilis)	sumDDT	3x10-3	part of ADI
(n= 14)	sumDDT	0,3x10-3	part of NOEL
Length 10,3-11,4cm	sumHCH	0,6x10-3	part of ADI
	sumPCB	4x10-3	part of NOEL
Väinameri region 1999	*sumDDT*	*0,07x10-3*	*part of ADI*
Perch *(Perca fluviatilis)*	*sumDDT*	*0,007x10-3*	*part of NOEL*
(n= 20)	sumHCH	0,9x10-3	part of ADI
Length 24,5-39,0cm	sumPCB	0,3x10-3	part of NOEL
Western coast of Estonia 1998	sumDDT	0,2x10-3	part of ADI
Vilsandi National Park (Roots, 2001)	sumDDT	0,01x10-3	part of NOEL
Perch (Perca fluviatilis)			
(n = 10)	sumHCH	0,1x10-3	part of ADI
Length 19,2-27,2cm	sumPCB	1x10-3	part of NOEL
Flounder *(Platichtys flesus)*	*sumDDT*	*3x10-3*	*part of ADI*
(n = 10)	sumDDT	0,2x10-3	part of NOEL
Length 19,1-28,8	sumHCH	0,4x10-3	part of ADI
	sumPCB	0,8x10-3	part of NOEL
Eastern coast of the Gulf of Finland 2000	*sumDDT*	*0,3x10-3*	*part of ADI*
Perch (Perca fluviatilis) +	sumDDT	0,02x10-3	part of NOEL

(n = 15)	sumHCH	0,6x10-3	part of ADI
Length 23,5-270cm	sumPCB	0,8x10-3	part of NOEL
Western coast of the Gulf of Finland 2001	sumDDT	1,3x10-3	part of ADI
Perch (Perca fluviatilis) ++	sumDDT	0,1x10-3	part of NOEL
(n = 14)	sumHCH	0,6x10-3	part of ADI
Length 19,6-28,3	sumPCB	0,6x10-3	part of NOEL

SumDDT- p,p`DDE; p,p`DDD; p,p`DDT ; sumHCH- alpha-HCH; gamma-HCH;

SumPCB- IUPAC No. 28, 31, 52, 101, 105, 118, 138, 153, 158, 163 and 180.

Roots, O. 2001. Halogenated environmental contaminants in fish from Estonian coastal areas. – Chemosphere, v.43, pp. 623 – 632.

Roots, O & Kakum, T. 1999. PCB and Chlororganic Pesticides in the Ecosystem of the Matsalu Bay. In.: Loodusvaatlusi 1997 – 1999. (Eds. E. Mägi & K. Kaisel), Matsalu Looduskaitseala , Lihula, pp.142 – 154 (in Estonian).

+ Keskkonnaelementide uuring ja analüüs. 2000. Eesti Keskkonnauuringute Keskus, Tallinn, 40 lk (in Estonian).

++ Kloororgaaniliste ühendite ja polüaromaatsete süsivesinike määramine rannikumere ökosüsteemis. 2001. Eesti Keskkonnauuringute Keskus, Tallinn, 32 lk (in Estonian).