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Comparison of DDE and PCB Residues in the General Diet and in Human Blood—Ontario 1986–87

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In an earlier study residues of PCB and DDE in the blood of the Ontario general public were below 10 and 0.2 $\mu g.kg^{-1}$ respectively (Frank et al. 1988). Williams et al (1988) published mean PCB and DDE residues in autopsy adipose tissue of Ontarians collected in 1984 showing levels of 2.1 $\pm$ 1.5 $\mu g\ g^{-1}$ and 3.2 $\pm$ 2.6 $\mu g\ g^{-1}$, respectively. Sahl et al. (1985) reported that PCB levels in blood plasma of 738 employees taken prior to employment by the Southern California Edison Company was 5 $\pm$ 4 ppb. This level was considered to be the level expected from exposure to PCBs in the general environment. It is worth noting that in Ontario DDT was removed from major uses in 1970 and all other field and outdoor uses by 1972. PCBs were voluntarily removed from open system uses in Canada during 1971. In 1976 federal legislation restricted all uses and disposal procedures for PCBs. In March 1989, new interim controls on PCBs were introduced under the Canadian Environmental Protection Act with final regulations to further reduce uses and emissions of PCBs under preparation. In this study the residues of DDE and PCBs in the general diet were compared with those found in the blood of Ontario residents.

MATERIALS AND METHODS

Foods were collected June to August 1986 as described in a joint Ontario Ministry of Agriculture and Food and Ministry of the Environment report (Anonymous 1988). Six major animal and five major plant commodities that were readily available in June to August were selected. Raw milk samples as part of this study had previously been selected and analysed in May 1986. Samples included beef as prime steaks and hamburger, pork chops, chicken broilers, hen eggs and fresh whole milk. Fruit and vegetables included apples, peaches, potatoes and tomatoes, and cereals included wheat. Blood was obtained from concerned patients suspecting oral, dermal or inhalation exposure to PCBs either environmentally or occupationally either recently or in the recent past. Blood was collected into stoppered new test tubes and sent by courier to the laboratory where samples were analysed within 48hr. Samples were submitted by general practitioners or health clinics across the industrial areas of central southern Ontario, an area where the large population of Ontario reside.

Blood and food were extracted according to the procedure described by Holdrinet (1974). Extracts were cleaned up and fractionated on Florisil and charcoal as described by Holdrinet (1974). DDE and PCB's were determined by packed column gas chromatography as described by Frank et al. (1985).

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PCB determinations were based on a comparison with either Aroclor 1254, 1260 or a mixture most closely resembling the sample chromatograms, and were quantitated by comparison of the sum of peaks VII, VIII, and X according to the Reynolds (1969) numbering system.

Recoveries of DDE and PCBs were checked periodically by fortification of sample tissue homogenate prior to the extraction. Recovery for DDE ranged from 88 to 97% while PCB recoveries averaged 85 to 90%. The data were not corrected for recoveries. Quantitation limits, below which values were designated as not detected, approximated 0.5 ug kg^{-1} for DDE and 6 ug kg^{-1} for PCB.

Residue levels of DDE and PCBs in blood were generally too low to be confirmed by methods described earlier by Frank et al. (1985, 1988, 1989). Confirmation was therefore obtained by analysis of selected samples using capillary column gas chromatography in conjunction with ion-trap mass-spectrometric detector.

RESULTS AND DISCUSSION

The dietary levels of DDE and PCB in foods consumed by Ontario residents during 1985-87 appears in Table 1 (Ontario Ministries of Agriculture and Food and Environment 1988, Frank and Braun 1988). The levels are reported on a whole food basis as well as in the extractable fat. DDE on a whole food basis ranged from 0.05 to 0.77 ug kg^{-1} while PCB ranged from <0.1 to <3.0 ug kg^{-1}.

Table 1. Mean dietary levels of DDE and PCBs in foods consumed by Ontario residents during 1985-87.

Food Item	In Extractable Fat			In Whole Food[1]	
	Fat %	DDE (ug/kg)	PCBs (ug/kg)	DDE (ug/kg)	PCBs (ug/kg)
Fruit - raw	-	-	-	<0.5	0.7
Vegetable - raw	-	-	-	0.3	<0.1
Wheat products	-	-	-	<0.5	< 3
Eggs	10.6	0.7	< 3	0.07	<0.3
Pork	35.0	2.2	3.3	0.77	1.1
Beef	11.0	2.9	< 3	0.32	<0.3
Broiler	9.8	0.5	< 3	0.05	<0.3
Milk	3.75	8.6	15	0.32	0.6

[1]Compilation of data generated by Davies (1988) and Anonymous (1988)

During 1986-87, 750 whole blood samples were analysed for DDE and PCBs and these data appear in Table 2 and 3. The data were grouped under six large urban centres and several medium to small urban centres. Although nine centres or regions are reported, the data came from a large number of urban centres in each of the regions or areas listed. The highest mean residue of PCBs was in the Niagara region as 13.0 ug kg^{-1} and the lowest in the Durham region at 6.2 ug kg^{-1}. These were not significantly different.

Table 2. Mean residues of PCBs and DDE in whole blood from residents of large and medium to small urban centres across Ontario 1986-87.

Urban Centres	Number	PCBs (ug/kg) Mean±SD	PCBs (ug/kg) Highest	DDE (ug/kg) Mean±SD	DDE (ug/kg) Highest
Large					
Etobicoke - North York	30	7.4± 8.4	49	3.3±5.7	30
Hamilton	237	9.8±11.2	110	3.2±3.5	29
London	64	10.0±10.1	57	2.7±2.8	16
Mississauga	36	6.4± 3.7	20	4.2±4.4	16
Scarborough	84	12.2±35.5	92	3.9±6.1	51
Toronto - York	130	7.3± 5.8	41	5.1±5.2	36
Subtotal	**581**	**9.3±16.0**	**110**	**3.8±4.0**	**51**
Medium to Small					
Burlington	16	10.2± 4.7	22	4.5±4.0	14
Dundas	12	6.7± 3.1	15	2.7±2.4	7.3
Durham Region	15	6.2± 4.2	17	2.0±1.5	6.0
Niagara Region	39	13.0± 9.9	43	4.2±3.6	13
South Western Ontario	12	7.3± 4.0	16	2.7±2.7	10
Stoney Creek	28	8.2± 3.5	16	2.6±2.3	11
Wentworth Region	12	11.5± 8.1	33	3.9±2.8	9.0
York Region	19	6.9± 4.5	20	3.8±5.0	16
Other Regions	16	6.8± 4.1	15	1.8±1.9	5.3
Subtotal	**169**	**8.8± 7.0**	**43**	**3.2±3.2**	**16**
Total	**750**	**9.2±14.5**	**110**	**3.7±3.9**	**51**

If it is assumed that diet can give blood residues of up to 10 ug kg^{-1} (Sahl et al. 1985) then 79% of samples fell into that range leaving a possible 21% where other exposures resulted in levels between 10 and 110 ug kg^{-1}.

The mean residue of DDE was 3.3 ug kg^{-1} and 96% of the blood had residues between 0 and 10 ug kg^{-1} (Tables 2 and 3).

PCBs and other organochlorines have been shown to contaminate sediments, aqueous phases and biota in Ontario lakes, most notably the Great Lakes (MacCrea et al. 1985; Schmitt et al. 1990; Environment Canada et al. 1991). Great Lakes Water Quality Objectives were set in 1986 on a whole fish basis for PCBs (0.1 ppm) and DDT and its metabolites (1.0 ppm). The PCB limit is exceeded on most fish types from all the Great Lakes, and the limit for DDT and its metabolites is exceeded in Lakes Ontario and Michigan. These whole fish criteria are important in protecting wildlife, especially fish-consuming birds and mammals, as well as the restriction of commercial use of whole fish products such as for pet food and animal feed. When considering direct

human exposure, analysis based on skinless, dorsal muscle portions has been used as the basis for the relevant Ontario Sports Fish Consumption Criteria of 2.0 ppm for PCBs. Monitoring has shown that larger fish of a variety of species exceed the guideline from Lakes Ontario, Erie, Huron, Michigan and some locations in Superior (Environment Canada et al. 1991). Although the Ministry of the Environment annually publishes a guide indicating lakes, and fish species and sizes where consumption restrictions on fish are recommended, this is intended to provide information to allow sports fisherman to make informed choices (Ontario Ministries of Environment and Natural Resources 1991). The potential exists for significant dietary exposures from consumption of sport fish.

If it is assumed that the whole blood levels of PCB derived from current diets falls between 0 and 10 ug kg^{-1} then this survey shows that 79% of individuals sampled derived their blood PCB levels from domestically-produced food (Table 2 and 3). The remaining 21% that contained residues between 10 and 100 ug kg^{-1} must be considered to have derived their residues from other sources and which could be from other dietary sources such as sport fish or from occupational or accidental exposure. The dietary levels of PCB for Ontario residents fell between <0.3 and 1.1 ug kg^{-1} in whole food or <3 to 15 ug kg^{-1} in extractable fat of foods. These data suggest a tenfold magnification from whole food to whole blood with extractable fat and whole blood having similar concentrations. Frank et al. (1988) reported human adipose concentrations of PCB to range from 400 to 3600 ug kg^{-1}, a thousand fold accumulation from whole food levels and hundred fold accumulation from the fat portion in food.

If it is assumed that the whole blood levels of DDE derived from diet also fall into the same range as PCB, namely 0 to 10 ug kg^{-1}, then 96% of blood DDE levels were derived from food (Table 2 and 3). This leaves 5% of blood levels that ranged from 10 to 51 ug kg^{-1} and could have come from occupational or accidental exposure. These exposures could include the use of DDT to treat buildings for mouse or bat control or exposure during demolition of buildings treated previously with DDT. The dietary levels of DDE ranged from 0.05 to 0.77 ug kg^{-1} in whole food or 0.5 to 8.6 ug kg^{-1} in extractable fat of animal products. Similar accumulations between food and blood were observed for DDE as for PCBs. Frank et al. (1988) reported human adipose concentrations of total DDT, mainly as DDE that ranged from 60 to 3560 ug kg^{-1}. Again similar magnification of DDE accumulations can be observed as to that reported for PCBs.

PCBs are found in human breast milk and adipose tissues. Health and Welfare Canada (1978) preliminary national study of PCB in human breast milk collected in 1975-76 found levels from 1 to 68 µg kg^{-1} of whole milk, with 98% of the samples below 50 µg kg^{-1}. Measurements have shown that PCB levels in blood and milk fat are proportional with blood levels of approximately 1/10 (Kodama and Ota 1980) to 1/20 (Mes et al. 1984) of those found in milk. The latter paper also reported a similar ratio for DDE. A longitudinal study of PCBs and organochlorine pesticides in breast milk in Norway by Skaare and Polder (1990) shows a general downward population trend over time in residue levels especially PCB and HCB; as well as a decrease in an individual's residue levels during the lactation period for first children, while no pronounced changes in levels were observed during lactation for second or third children.

Table 3. Frequency distribution of PCB and DDE residues in whole blood from residents from large and medium to small urban centres across Ontario, 1986-87.

Urban Centres	PCB Numbers					DDE Numbers			
	0-10	11-20	21-30	31-50	51-110	0-10	11-20	21-40	41-60
	ug/kg								
Large									
Etobicoke - North York	28	1	0	1	0	28	1	1	0
Hamilton	182	38	9	4	4	228	7	2	0
London	51	7	5	0	1	62	2	0	0
Mississauga	32	4	0	0	0	32	4	0	0
Scarborough	66	14	3	0	1	81	1	1	1
Toronto - York	110	14	4	2	0	124	4	2	0
Subtotal	459	78	21	7	6	555	19	6	1
%	80.7	13.4	3.6	1.2	1.0	95.5	3.3	1.0	0.2
Medium to Small									
Burlington	10	5	1	0	0	15	1	0	0
Dundas	11	1	0	0	0	12	0	0	0
Durham Region	13	2	0	0	0	15	0	0	0
Niagara Region	24	8	4	3	0	35	4	0	0
South Western Ontario	9	3	0	0	0	12	0	0	0
Stoney Creek	19	9	0	0	0	27	0	0	0
Wentworth Region	9	3	0	0	0	12	0	0	0
York Region	16	3	0	0	0	17	2	0	0
Other Regions	12	4	0	0	0	16	0	0	0
Subtotal	123	38	5	3	0	16	8	0	0
%	72.8	22.5	3.0	1.8	0.0	95.3	4.7	0.0	0.0
Total	592	116	26	10	6	716	27	6	1
%	78.9	15.5	3.5	1.3	0.8	95.5	3.6	0.8	0.1

Since both PCBs and DDE are fat soluble persistent organochlorine compounds, they would be expected to accumulate to similar levels of magnitude. Over the last two decades the residues of DDE and PCBs have been observed to decline in domestically produced food in Ontario. Over the same period residues of these two components in adipose tissues has declined only very slowly and appeared to reach a steady state. These findings probably reflect that storage in tissues is very long term. While the turnover in generations of animals making up the food basket is rapid ie. 16 weeks for poultry, six to eights months for pork, one to two years for beef, hence giving a rapid cleansing of the food system, the turnover in generations of humans is slow.

The ongoing exposure to organochlorine residues in the diet from especially sports fish, and transference of PCBs and other organochlorine residues *in utero* from placental transfer in blood and from breast-feeding to infants makes the complete elimination of PCB exposure to new generations difficult in the shortrun. Since the cessation in use of DDT (1970-72), and PCBs (1971-76), time has elapsed to allow several generations of domestic food producing animals but not allowed time for the clean-up of the human system.

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