# **HUM I**

# PCBs and chlorinated pesticides in blood from Swedish men

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### 1. Introduction

PCBs and chlorinated pesticides (e.g. DDT, chlordane, HCB, HCH) are man-made chemicals that have the potential to negatively affect human health owing to their persistence and toxicity. The production and use of this large group of chlorinated substances have been banned in Sweden for many years. However, due to their persistence in the environment, exposure to PCBs and chlorinated pesticides via food will continue for decades. We report here preliminary results on levels of PCB and chlorinated pesticides in blood from Swedish males of ages 40 to 75 years, as a part of a project on food as a source of exposure to PCBs and chlorinated pesticides.

#### 2. Methods

The original sample was composed of 800 men from Uppsala and Knytby, selected as representatives of the general male population in urban and rural areas of Sweden. Of these individuals, 286 agreed to participate in an extensive survey of food habits and to donate blood samples. The blood sera donated by the first 120 individuals (age 40-75 years) were analysed for PCB and chlorinated pesticides. The PCB congeners analysed were IUPAC no 28, 52, 101, 105, 118, 128, 138, 153, 156, 167 and 180. The chlorinated pesticides hexachlorbenzene (HCB), hexachlorocyclohexane ( $\alpha$ -,  $\beta$ - and  $\gamma$ -HCH), chlordane (oxychlordane and transnonachlor) and DDT (*p*,*p*-DDT, *p*,*p*-DDD, *p*,*p*-DDE, *o*,*p*-DDE and *o*,*p*-DDT) were also analysed.

The analytical method was based on an extraction method originally described by Burse *et al.* (1) and an analytical technique described by Atuma *et al.* (2). Four grams of serum was mixed with 4 ml methanol and the mixture was vortexed for 1 min. The serum was extracted three times with n-hexane/diethyl ether mixture (1:1). After evaporating the combined extracts, the fat content was determined gravimetrically.

The fat extract (dissolved in hexane) was treated with concentrated sulphuric acid followed by elution through a silica gel column (4.5 g of 3% water-deactivated silica gel) with ca 30 ml hexane (fraction 1). Fraction 2, which contained the chlorinated pesticides (except DDE and HCB), was eluted with 40 ml of a hexane/diethyl ether mixture (3:1). After appropriate reduction in volume, fraction 1 was injected into a Gilson HPLC system, equipped with an hypercarb column. The first two fractions of 0-4 ml and 4-20 ml, respectively, collected with hexane as mobile phase, contained mainly the di-*ortho* and mono-*ortho* PCBs respectively. The third fraction, which contained the planar PCBs, was obtained by back flushing the column with DCM (100%). After each complete run, the HPLC column performance was restored by flushing with n-hexane at a flow rate of 1-2 ml/min for at least 30 min, to re-establish equilibrium. The mono-*ortho* and di-*ortho* PCB congeners and the chlorinated pesticides were finally quantified on a dual-column gas chromatograph with dual-electron capture detectors.

#### 3. Results and Discussion

The concentrations of total PCB and total DDT were generally higher than those of HCB, total HCH and total chlordane (Table 1). PCB 153 was strongly correlated to the total PCB concentration (r=0.98, p<0.001) (Fig. 1a). Furthermore, PCB 153 contributed 38% ( $\pm$ 4%) to the total PCB concentration. This relationship was constant in the age interval studied (Fig. 1a), showing that PCB 153 can be used as an indicator substance for total PCB in monitoring studies of men of different ages. A recent study has also shown that PCB 153 can be used as an indicator for total PCB in serum from Swedish females (3).

*p*,*p*-DDE contributed 96% (±3%) to the total DDT concentration in blood serum. As for PCB 153, this percentage was constant over the age interval studied. The levels of *p*,*p*-DDD, *o*,*p*-DDT and *o*,*p*-DDE were low and for *o*,*p*-DDT and *o*,*p*-DDE generally below the detection limit (<4 ng/g fat), showing that recent direct exposure to technical DDT mixtures had not occurred.  $\beta$ -HCH dominated among the HCH isomers (93±5% of the total HCH level), whereas the concentration of  $\gamma$ -HCH (lindane) was low (<2-12 ng/g fat). The concentration of the chlordane isomer, transnonachlor, was about twice as high as that of oxychlordane.

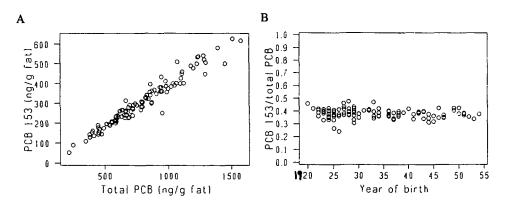


Fig. 1. (A) Correlation between the concentrations of PCB 153 and total PCB in sera from Swedish men (r=0.98, p<0.001). (B) The ratio between the concentration of PCB 153 and total PCB among Swedish men of different ages.

Table 1. Concentrations	of PCB	and	chlorinated	pesticides	in	blood	of	Swedish	men	(mean±SD,
N=119-120)										

PCB <sup>a</sup> (ng/g fat)	PCB-TEQ <sup>b</sup> (pg/g fat)	HCB (ng/g fat)	HCH <sup>c</sup> (ng/g fat)	Chlordane <sup>d</sup> (ng/g fat)	p,p-DDE (ng/g fat)	Sum-DDT <sup>e</sup> (ng/g fat)
765±289	19±6	71±40	52±30	45±22	811±705	834±715
apCB conge	eners 28, 52, 101,	105, 118, 128, 1	38, 153, 156, 16	7 and 180		

aPCB congeners 28, 52, 101, 105, 118, 128, 138, 153, 156, 167 and bI-TEQs for PCB congeners 105, 118, 156, 167 and 180  $c_{\alpha}$ -HCH,  $\beta$ -HCH and  $\gamma$ -HCH dOxychlordane and transnonachlor

ep,p-DDT, p,p-DDD and p,p-DDE

Substance							
	1920-24 (n=22)	1925-29 (n=34)	<b>1930-34</b> (n=16)	1935-39 (n=14)	<b>1940-44</b> (n=12)	1 <b>945-49</b> (n=9)	<b>1950-54</b> (n=9)
Total PCB	822±313	806±285	841±258	713±223	748±401	693±250	502±82
Total DDT	1024±892	861±718	1053±707	661±391	745±662	760±762	393±439
HCB	80±39	69±35	62±27	61±16	89±70	67±26	72±71
Total HCH	52±25	55±33	49±26	49±19	49±32	64±50	40±25
Chlordanc	48±18	51±22	52±25	39±19	37±28	36±12	22±6

Table 2. Concentrations of PCB and chlorinated pesticides in different age classes of Swedish men

The levels of PCB congeners and chlorinated pesticides increased slightly with increasing age, although the spread in the results was large (Table 2). Men born between 1920 and 1934 had total PCB and total DDT levels ranging from 171 to 1510 ng/g fat and 138 to 4117 ng/g fat, respectively. The concentrations of total PCB and total DDT were positively correlated (r=0.61, p<0.001), indicating a common source of exposure. This was also the case for PCB and chlordane (r=0.64, p<0.001). The correlation between PCB and the other chlorinated pesticides was lower (HCB: r=0.35, p<0.001; HCH: r=0.39, p<0.001), indicating differences in the sources of exposure between PCB and the chlorinated pesticides.

The mean serum concentrations of PCB 153 (299 ng/g lipid), p,p-DDT (20 ng/g lipid) and p,p-DDE (811 ng/g lipid) in males in the present study are somewhat lower than in blood plasma from males with a moderate consumption of fish from the Baltic Sea (580, 45 and 1200 ng/g lipid) (4). Males with a high consumption of fish from the Baltic Sea had mean concentrations of PCB 153, p,p-DDT and p,p-DDE that were about 5 times higher than in the males in the present study (4). The relatively low levels of PCB and DDT in the present study are probably due to a low consumption of fish from the Baltic Sea among the males from Uppsala and Knutby.

Mono-ortho PCB congeners (IUPAC no 105, 118, 156 and 167) and one di-ortho PCB congeners (IUPAC no 180) analysed in the present study have dioxin-like activity, and 2,3,7,8-tetrachloro-pdioxin toxic equivalency factors have been proposed for these congeners by an international expert group (5). A calculation of the concentration of TCDD toxicity equivalents (TEQs) from the concentrations of mono- and di-ortho PCBs resulted in a mean TEQ level of  $19\pm 6$  pg/g serum fat. As a comparison, a previous study on dioxin and dibenzofuran levels in Swedish males indicated a mean dioxin/dibenzofuran TEQ level ranging from 18 to 77 pg/g serum fat, depending on the consumption rate of fish from the Baltic Sea (4). Taken together, the results indicate that the mono- and di-ortho PCBs give a significant contribution to the total TEQ concentration.

#### 4. Conclusions

All of the males in the present study had measurable levels of PCB and chlorinated pesticides in the blood. The levels were low in comparison to some accidentally or occupationally exposed individuals (6-8). Nevertheless, it is shown that contamination of the human body by PCBs and chlorinated pesticides is still evident although the production and use of many of the chlorinated substances have been severely restricted for decades.

#### 5. References

- 1) Burse V.W., D.F.Groce, M.P Korver, P.C. McClure, S.L. Head, L.L. Needham, C.R. Lapeza and A.L. Smrek (1990). Use of reference pools to compare the qualitative and quantitative determination of PCBs by packed and capillary gas chromatography with ECD. Part I. Analyst 115, 243-251.
- 2) Atuma S. S and L. Hansson (1994): Using porous graphitic carbon column for HPLC separation and isolation of potentially toxic PCB congeners in fish. Dioxin '94 Extended Abstract: Organohalogen Compounds 19, 81-84.
- Grimvall E. (1995). Biological and environmental monitoring of polychlorinated biphenyls. Ph. D. Thesis. Department of Analytical Chemistry, Stockholm University, Stockholm, Sweden.
- Asplund L., B.-G. Svensson, A. Nilsson, U. Eriksson, B. Jansson, S. Jensen, U. Wideqvist and S. Skerfving (1994). Polychlorinated biphenyls, 1,1,1-trichloro-2,2-bis(p-chlorophenyl)ethane (p,p'-DDT) and 1,1-dichloro-2,2-bis(p-chlorophenyl)-ethylene (p,p'-DDE) in human plasma related to fish consumption. Arch. Environ. Health 49, 477-486.
- Ahlborg, U.G., G.C. Becking, L.S. Birnbaum, et al. (1994). Toxic equivalence factors for dioxin-like PCBs. Report on a WHO-ECEH and ICPS consultation, December, 1993. Chemosphere 28, 1049-1067.
- 6) Kolmodin-Hedman, B., L. Palmér, P. Götell and S Skerfving (1973). Plasma levels of lindane, p,p-DDE and p,p-DDT in occupationally exposed persons in Sweden. Scand. Work Environ. Health 10-2, 100-106.
- Jitunari, P., F. Asakawa, N. Takeda, S. Suna, Y. Manabe (1995). Chlordane compounds and metabolite residues in termite control workers' blood. Bull. Environ. Contam. Toxicol. 54, 855-862.
- 8) Luotamo, M., J. Järvisalo and A. Aitio (1991). Assessment of exposure to polychlorinated biphenyls: analysis of selected isomers in blood and adipose tissue. Environ. Res. 54, 121-134.