

THE GERMAN ENVIRONMENTAL SPECIMEN BANK - MONITORING OF PENTACHLOROPHENOL AND HEXACHLOROBENZENE IN HUMAN BLOOD

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Introduction

Several chlorinated hydrocarbons as e.g. pentachlorophenol (PCP) and hexachlorobenzene (HCB) are widely restricted in industrialized countries because of known adverse effects to humans and to the environment. Many regulative and legal measures have been taken to reduce the release of this class of compounds. The success of these measures can be demonstrated by biomonitoring.

Pentachlorophenol was widely used as a pesticide and wood preservative. Production, marketing of PCP and PCP compounds have been banned in Germany by the Pentachlorophenol Prohibition Ordinance since 1989¹. However, exposure of humans might continue from the uncontrolled import of PCP treated goods (leather, textiles, wood) and from treated products still in use. PCP has been also a major source of dioxins in the past. Although the half life of PCP in the body is very short (10 – 20 days), PCP is still measurable in blood plasma.

Hexachlorobenzene (HCB) was used as a fungicide to protect the seeds of plants and as a chemical intermediate in the manufacture of solvents and dyes. As a pesticide it has been widely banned (e.g. 1977 in Germany), but large quantities of HCB continue to be produced and released into the environment as a waste by-product or impurity in the manufacture of pesticides and plastics. HCB is one of the 12 chemicals treated by the Stockholm POP convention². Due to its lipophilic and persistent character, food is the main source of HCB for humans. HCB accumulates in the food chain and in the human body with a half life of 2 to 3 years. HCB has some properties that are typical for dioxin like compounds. Therefore including of HCB in the TEF concept is under discussion^{3,4}.

The German Environmental Specimen bank (ESB) was established in 1985 as a permanent institution for the systematic collection, processing, characterization and storage of environmental samples from marine, limnic and terrestrial ecosystems as well as human samples⁵. Blood and other human specimens have been collected since 1981 from unexposed persons in defined peripheral conditions. The subjects have to complete standard questionnaires about family and health status, occupational exposure, nutrition, smoking and drinking habits and the use of medicine. Every step in the procedure from sampling to transport, preparation, chemical analysis and long-term storage is carried out according to obligatory Standard Operating Procedures⁶ (SOP).

The aim of the present study is to report current levels and time trends of PCP and HCB in human plasma over the past twenty years (1985-2005) in Germany.

Material and Methods

Study group. The study sample set is a subset of routine Real-Time-Monitoring (RTM) sampling campaigns of the German ESB. Since 1997 annual sampling campaigns consist of acquiring a total of about 500 students (about half of them female) in four German cities (Muenster, Halle, Greifswald, Ulm) and are accomplished during the same season of one year, i.e. in the late winter or early spring (Table 1). To avoid variation of body burden with age the study was restricted to subjects born in Germany in the age range 20-29 years.

Sample preparation and laboratory analysis. Details of the sampling procedures and laboratory measurements are described in SOPs³. Briefly, blood samples are prepared (plasma extraction) and portioned immediately after withdrawal and stored under nitrogen atmosphere (-150°C). The analysis of

Body burdens: pattern, levels and trends

PCP and HCB concentrations in blood plasma is done via gaschromatographic mass spectrometric detection (GC-MS-system Finnigan SSQ 7000) after extraction with hexane+acetone (4+1, pH 2.0) and derivatization with CH_2N_2 . In addition to POP concentrations other important clinical parameters (protein, cholesterol, tryglycerides) are determined too, using common clinical methods.

Table 1 Study groups in the framework of the German ESB

Sampling place	Start of sampling	Number median (range)
Münster	1985	122 (77 – 143)
Greifswald	1992	122 (51 – 128)
Halle	1995	116 (111 – 121)
Ulm	1997	119 (102 – 127)

Results and Discussion

No significant differences between sample locations was apparent, so PCP and HCB concentrations in human blood plasma were evaluated without local differentiation.

Pentachlorophenol: In the observation period 1985 to 2005 median PCP concentrations in plasma samples decreased by about 95% from 2.64 to 0.13 $\mu\text{g/L}$ (Table 2). Recent years show slower but still significantly decreasing time trends. The success of PCP banning became obvious in the period 1991 to 1992 by a drastically decline in levels of about 44 % within one year. PCP concentrations in male and female species differ hardly, no significant correlation between gender and concentration were detectable.(Figure 1). Due to the short half life of PCP in the human body, it has to be assumed that decreasing body burden is directly related to continuously decreasing releases of PCP into the environment.

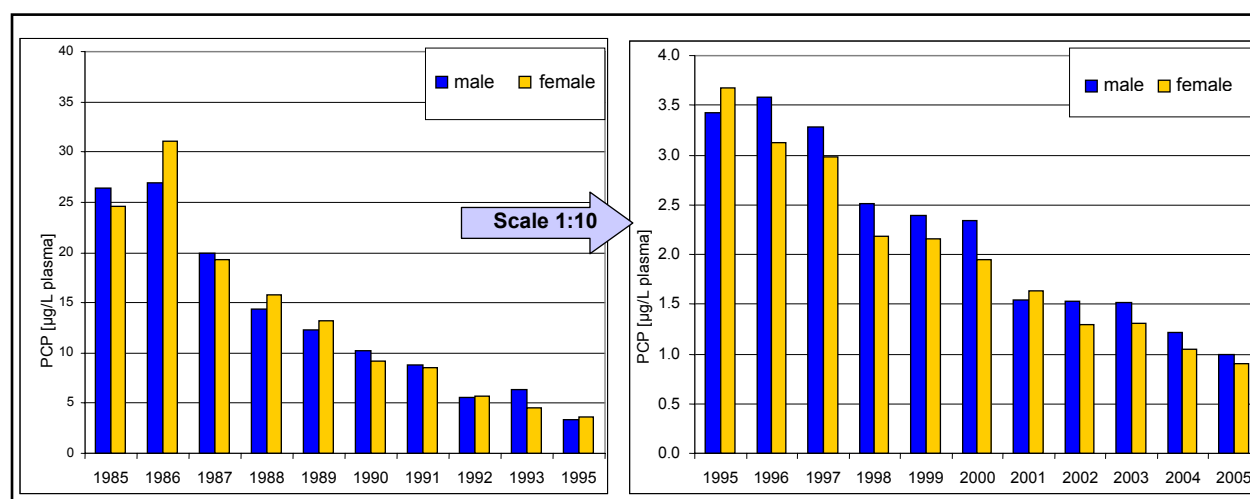


Figure 1. Median concentrations ($\mu\text{g/L}$) of pentachlorophenol in plasma samples from 1985 to 2005. Note: change in scale in 1995.

Table 2 Basic statistical data on PCP ($\mu\text{g/L}$) in human plasma samples

Year	Number	Mean	Minimum	Maximum	Median	90. Percentile	95. Percentile
1985	129	30.6	8.9	128.3	25.8	49.8	62.8
1986	85	36.5	13.7	153.1	29.2	59.2	86.8
1987	68	22.9	7.5	97.5	19.2	32.5	38.5
1988	233	18.6	2.1	204.7	14.9	28.8	36.8
1989	209	16.6	3.4	213.5	12.4	25.6	33.7
1990	165	11.8	3.5	73.4	9.9	20.7	23.0
1991	71	12.4	3.3	98.3	8.5	22.6	29.2
1992	130	7.4	1.4	83.6	5.6	12.2	15.8
1993	63	6.2	1.5	26.0	4.8	9.9	14.5
1995	428	4.9	0.1	46.4	3.5	10.0	14.9
1996	565	5.8	0.7	160.8	3.3	10.8	17.3
1997	370	4.7	0.4	36.8	3.1	9.5	14.0
1998	361	3.8	0.7	32.8	2.3	7.0	10.8
1999	398	3.5	0.5	51.8	2.3	6.3	9.3
2000	397	3.1	0.6	55.5	2.0	5.4	8.3
2001	387	2.3	0.3	51.4	1.6	3.5	4.5
2002	448	2.0	0.4	17.2	1.4	3.3	5.6
2003	443	2.4	0.3	74.2	1.4	4.8	7.7
2004	455	1.6	0.3	18.4	1.2	2.7	3.8
2005	449	1.6	0.3	52.0	0.9	2.4	3.6

Hexachlorobenzene: Between 1985 and 2005 median HCB concentrations in human blood plasma decreased from 25.8 to 0.95 $\mu\text{g/L}$. This is a reduction of more than 96 % in 20 years (Table 3). A strong correlation between gender and concentration became evident. In general, women have higher HCB concentrations in blood plasma (Figure 2). This finding is in accordance with the results of the German Environmental Survey 1998⁷. Explanations for gender-related differences due to a different lipid metabolism are ambiguous. PCBs are chemically akin to HCB but for men PCB concentrations in plasma are usually higher than for women⁸.

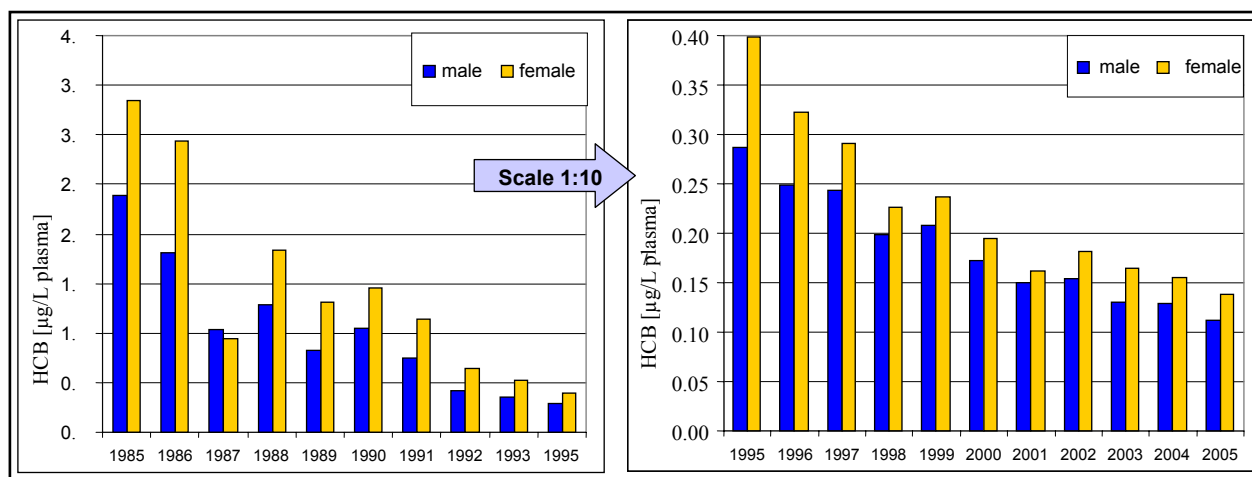


Figure 2. Median concentrations ($\mu\text{g/L}$) of hexachlorobenzene in plasma samples from 1985 to 2005. Note: change in scale in 1995.

Table 3 Basic statistical data on HCB ($\mu\text{g/L}$) in human plasma samples

Year	Number	Mean	Minimum	Maximum	Median	90. Percentile	95. Percentile
1985	129	3.15	0.49	18.86	2.64	5.20	5.93
1986	85	2.62	0.62	6.72	2.26	4.40	5.37
1987	68	1.16	0.23	6.33	0.97	2.14	2.59
1988	233	1.81	0.25	6.62	1.59	2.96	4.09
1989	209	1.17	0.25	5.67	0.94	2.10	2.75
1990	165	1.27	0.28	4.14	1.18	2.04	2.22
1991	71	1.24	0.28	4.75	0.97	2.17	2.90
1992	130	0.61	0.16	2.25	0.53	0.96	1.14
1993	64	0.52	0.18	1.83	0.45	0.90	1.00
1995	428	0.42	0.11	3.56	0.34	0.74	0.95
1996	565	0.36	0.09	5.80	0.28	0.54	0.69
1997	370	0.30	0.10	1.02	0.27	0.48	0.62
1998	361	0.28	0.06	7.49	0.21	0.37	0.49
1999	398	0.25	0.09	4.00	0.23	0.36	0.40
2000	397	0.20	0.06	1.45	0.19	0.28	0.37
2001	387	0.22	0.04	4.38	0.16	0.27	0.39
2002	448	0.18	0.07	0.53	0.17	0.25	0.28
2003	443	0.16	0.04	0.60	0.15	0.23	0.26
2004	455	0.16	0.05	2.35	0.15	0.23	0.26
2005	449	0.14	0.02	0.47	0.13	0.21	0.24

The real-time-monitoring of HCB and PCP demonstrates impressively the effectiveness and success of the implemented measures. Furthermore, as PCP is a major source for dioxin exposure, the drastically reduction had also a declining effect on dioxin in humans⁹.

References

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