Dietary exposure and human body burden to Organochlorine Pesticides and PCBs in children and women in Northern Germany

Gudrun Petzold, Maike Schäfer, Christiane Benthe, Guido Ostendorp, Gabriele Schade*, Michael Wilhelm** and Birger Heinzow

Department of Environmental Toxicology, Agency for Nature and Environment, Hamburger Chaussee 25, D - 24220 Flintbek, Germany; **Dept. Hygiene, Ruhr-University Bochum, *Ministerium für Arbeit, Gesundheit und Soziales, Adolf-Westphal-Str. 4, D - 24143 Kiel

Introduction

It is common opinion that food , especially animal fat, is the main source for exposure to persistent organic pollutants in the general population. Inhalation exposure is assumed as less important being in the range of 10 % of oral intake. These assumption however are not based on actual diet studies. It was the aim of this study to measure the concentrations of organochlorine pesticides and PCBs in duplikate diet samples to estimate the range of dietary intake and compare the data with measured concentrations in serumfat and breast milk of women from Northern Germany.

Materials and Methods

Individual duplicate diet samples of all food and beverages were collected 1995 daily for one week in PE-containers in seven male and seven female children at the age of 1.5 to 5.3 years living on an island in Northern Germany and in 1997 in seven women, aged 20 - 35 years, living in a city in Northern Germany. Blood samples (10 ml) were taken from 6 women and serum samples obtained. Questionnaires on individual characteristics and 24 hour dietary protocols were applied. From 1986 to 1997 more than 3500 milk samples were analyzed from the Northern German region. Informed consent was obtained in all cases.

Analytical Determination

Duplicate samples were homogenized, freeze dried, a 3 g aliquot was further processed by solid-liquid-phase extraction (Florisil) and silica gel-clean-up and analyzed by GC-ECD using a modified method described by Stivje and Cardinale (1) including hexachlorobenze (HCB), hexachlorocyclohexane-isomers(α-, β-, γ- HCH), Dichlorodiphenytrichloroethane DDT (p`,p-DDE and p`,p-DDT) and polychlorinated biphenyls congeners (PCBs 101, 138, 153, 180). The lipid content of the dried matter was determined by gravimetry, the lipid content of the serum and breast milk samples was determined by means of the photometric Merckotest 3221 (Merck) for total lipids. Analyses were performed by high-resolution gas chromatography on a Varian 3400 gaschromatograph equipped with an autosampler (Varian 8100), a 30m DB-5 capillary column and electron capture detector. The specific congeners were identified and quantified by comparison with response and retention times of calibration standards for each analyte of interest. The detection limit for the presented substances was 0.005 mg/kg fat. For each 8 analyte-runs one blind sample and one standard-sample was included for internal quality control. Total PCBs were calculated as 1.64 x (PCB 138 + 153 +180) (2). A similar procedure was applied for analysis of serum samples and breast milk samples. Daily intake (DI)[µg/kgbodyweight/day] is calculated as : concentration in food [µg/kg] ∗ daily food intake [kg/day]/bodyweight[kg].

Results and Discussion

a) Duplicate Diet Studies : Full reports on these studies can be obtained from the Agency for Nature and Environment (LANU Schleswig-Holstein). Results on 98 duplicate diet samples for children and 49 for women are given in table 1 respectively. From the questionnaires no specific dietary habits (e. g. vegetarian) are obvious. The food samples reflect a standard western european diet pattern. The daily average intake of food is 950 g/day /person (range 477 - 1855 g), the mean lipid content of the fresh food was 9.1 % (5.4 - 11.6 %). PCBs and DDT were the main contaminants. Comparison of the DI values for children and adults with present guidelines values for dietary intake (Acceptable Daily Intake, ADI) can be taken from table 2. The reference contamination (95. percentile) for PCBs amounts to 5 % and 2.6 % of the ADI, due to a higher food intake/bodyweight, childrens DI are in average doubled.

b) Biomonitoring : Serum samples contained an average 6.8 μg fat/l (5.4 - 7.9 μg/l). DDE and PCBs were the main contaminats as depicted in table 3. Lindane (γ-HCH) and DDT was below the limit of detection in all samples. Results are comparable to other recently published data (3). Analysis of the questionnaires and the dietary protocols revealed that the amount of dairy products, fish and meat correlated with higher contaminat concentrations in the food samples. Women with diets rich in dairy products, eggs, fatty fish and meat had higher lipid serum levels and higher DDE, HCB and PCBs concentrations in serum. Breast-milk: For comparison reasons milk subsamples were selected by using narrow limits for the parameters : age of mother (27 - 31 years), parity (primaparae) and previous nursing time (16 - 24 weeks).

The investigation of milk samples from 1986 to 1997 (4). indicated a considerable decrease of the levels of pesticides and PCBs. The median levels, expressed on a fat basis, found in the breast milk samples from 1986 to 1997 are shown in Figure 1. The levels of HCB, DDT, β-HCH and PCB concentration found in milk samples analyzed 1995 to 1997 were in the same range as in industrialized countries from other parts of the world (5, 6, 7, 8, 9). Trend for all samples (n = 3660) was similar to the results shown for selected subsamples.

The mean daily intake from previous dietary intake studies in comparisons to our study ranges from 0.03 - 0.1 μg/kgbw (10, 11, 12, 13). Together with the data from the biomonitoring studies this indicates clearly the declining environmental pollution and carry-over in the food chain. When compared with data obtained 10 - 12 years ago, HCB and β-HCH median levels indicate a more than 80 % decrease, and PCB median levels a 60 % decrease. In the case of PCB, this decline can be attributed to the prohibition of these compounds in Germany 1989. The even greater decrease in HCB may depend on the replacement of HCB as an insecticide for grain as well on an improvement in the industrial processes producing HCB as a by-product. DDT is reduced by 80 % during the same period, demonstrating the effectiveness of the government-imposed ban (1972) on the use of DDT in Germany. A similar decline in residue has been observed also in Sweden (14, 15), Norway (16), Spain (17) and Canada (6). However from the view of environmental pollution and toxicology reduction of POPs emission into the environment will remain a major public health task for the future.

Tab. 1: Concentration of POPs in foodstuff (duplicate diet samples) of adult women and of children in [μg/kg wetweight]

POP	woman (n = 49)			children (n = 98)		
	median	95. percentile	maximum	median	95. percentile	maximum

HCB	0.17	0.35	0.44	0.062	0.15	0.57
α-HCH	0.07	0.26	0.46	0.026	0.067	0.129
β-HCH	0.04	0.13	0.17	0.016	0.05	0.23
γ-HCH	0.24	0.65	0.79	0.13	0.41	1.3
DDE-4	0.35	0.98	3.59	0.16	0.4	1.4
DDT-4	0.06	0.24	1.23	0.05	0.18	4.3
Σ-DDT	0.44	1.16	4.82	0.22	0.5	5.63
PCB 101	0.04	0.19	0.79	0.015	0.08	0.16
PCB 153	0.14	0.48	1.82	0.08	0.18	0.31
PCB 138	0.13	0.47	1.66	0.08	0.18	0.34
PCB 180	0.07	0.17	0.5	0.03	0.1	0.23
Σ-PCB	0.54	1.73	6.53	0.31	0.75	1.43

Tab. 2: Dietary intake (95. percentile) of POPs in relation to recommended acceptable daily intake (ADI) in children and women

Substance [μg/kg FW]	ADI [μg/(kg $_{bw}$ d)]	DI (95 perc.) children [μg/(kg $_{bw}$ d)]	DI fractile ~of ADI %	DI (95.perc) women [μg/(kg $_{bw}$ d)]	DI fractile ~ of ADI %
α-HCH	5	0.0035	0.07	0.003	0.06
ß-HCB	1	0.0025	0.25	0.002	0.2
γ-HCH	10	0.02	0.2	0.007	0.1
HCB	0.6	0.008	1.3	0.004	0.7
Σ-DDT #	5	0.03	0.6	0.015	0.3
Σ -PCB ##	1	0.05	5	0.026	2.6

Σ-PCB = (sum of congeners 138+154+180)* 1.64
Σ-DDT = (DDE +DDT)

Tab. 3: Concentration of POPs [μg/l] in serum of 6 adult women participating in the duplicate diet study

women	Fat [g/l]	HCB	β-HCH	DDE	PCB 153	PCB 138	PCB 180	Σ-PCB *)
Mean	6.8	1.0	0.3	1.3	0.8	0.6	0.7	3.4
Median	6.9	0.9	0.3	1.3	0.8	0.7	0.7	3.5
Minimum	5.4	0.6	0.1	0.3	0.5	0.4	0.5	2.3
Maximum	7.9	1.7	0.4	2	1.0	0.8	0.9	4.3

*) Σ-PCB (138+ 153 + 180) x 1.64

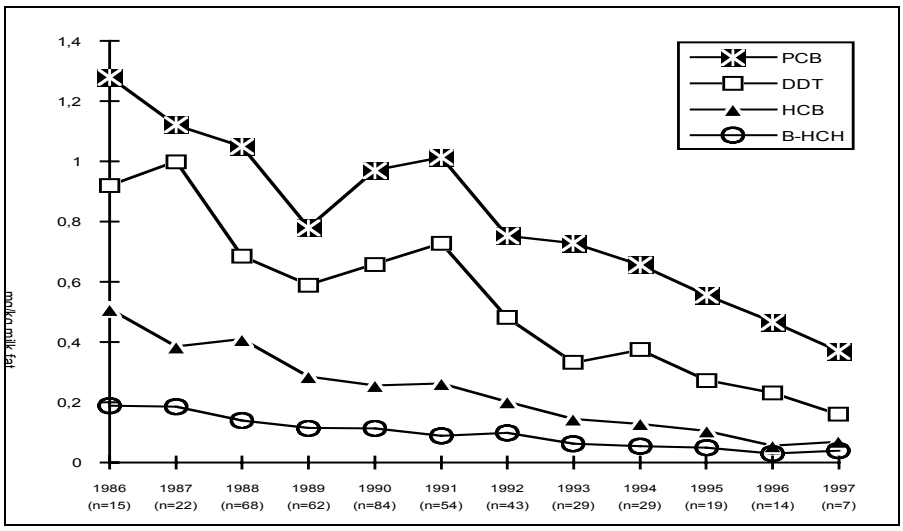

Figure 1: Median levels of PCB, DDT, HCB and β-HCH concentration in homogenous sub-samples between 1986 and 1997 (n = number of selected subsamples); [4]

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