PCBs, PCDDs, PCDFs and Organochlorine Pesticides in Human milk in The Netherlands. Levels and Trends.

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Abstract

Results are presented of a survey on levels of organochlorine pesticides (OCPs), polychlorinated biphenyls (PCBs), dibenzodioxins (PCDDs) and dibenzofurans (PCDFs) in individual human milk samples from the Netherlands collected in June 1993. Observed data indicate that since 1972 levels of OCPs (except for p,p'-DDE) are declining to levels around the limits of determination (0.01-0.03 mg/kg of fat). A decline (of approximately 30%) is also observed for the PCDDs and PCDFs in the period 1988-1993. However, no significant decrease could be found for the predominant PCB congeners with IUPAC nos. 118, 138, 153 and 180 in the period 1983-1993.

1. Introduction

In the Netherlands human milk surveys are performed at five-year intervals to study trends in human exposure and to evaluate risks of subsequent exposure of breastfed infants to harmful xenobiotic compounds like organochlorine pesticides (OCPs, since 1972), polychlorinated biphenyls (PCBs, as individual compounds since 1983), dibenzodioxins and dibenzofurans (PCDDs and PCDFs, since 1988) [1-2]. In this paper, we report on the results from the 1993 survey in which about 100 individual samples have been analysed that were collected in maternity centers throughout the Netherlands in June 1993. Results are briefly discussed in terms of trends in environmental concentrations and exposure levels for the general population [3].

2. Materials and methods

Sampling strategy

The sampling strategy for June 1993 was similar to the approach used in former human milk surveys [2]. With the assistance of maternity centers, women (primiparae) planning to breastfeed and willing to cooperate were included in the survey-population. Each respondent was asked to collect a

(100 ml) milk sample and to give information on factors potentially influencing the levels of contamination of human milk (e.g. personal characteristics, pre-pregnancy dietary habits, residential factors, life style, workplace, recreative habits). To measure babies (post-natal) exposure, mothers were also asked to, retrospectively, describe their babies feeding pattern during the lactation period. Furthermore, clinical parameters of babies thyroid functioning, collected in a national screening programme, were made accessible by informed consent.

Laboratory analysis

The analytical programme consisted of compound specific determinations of ten OCPs, fifteen PCBs, and seventeen 2,3,7,8-substituted PCDDs and PCDFs. Analysis was based on fortification of the milk samples with ^{13}C-labelled internal standards (PCDDs, PCDFs, non-ortho-PCBs), extraction of the sample [4], followed by purification of the fat extract and identification and quantitation by use of gaschromatographic techniques with either electron capture detection (GC-ECD) for OCPs and mono- and di-ortho substituted PCBs [5] or high resolution mass spectrometry (GC-HRMS) for PCDDs, PCDFs [6] and non-ortho substituted PCBs [7]. To control the purity of glassware, reagents and solvents used during sample preparation, different blanks were analysed prior to analysis of a series of samples.

Analytical variation

Method performance characteristics of applied analytical methods have been published in more detail elsewhere [4-7]. Based on the results from participation in WHO coordinated interlaboratory quality control studies [8-9], our laboratory has been qualified by WHO/EURO to perform analyses of PCDDs, PCDFs and PCBs in the framework of analytical field studies on human milk.
To test long-term repeatability, quality control (QC) samples of cow's milk (N=8) and human milk (N=11) were incorporated in each series of samples during the course of this study. These tests showed relative standard deviations (RSD) of 2.3 (N=8) and 2.5% (N=11) for the determination of the fat content in the QC-sample of cow's milk and human milk, respectively. For HCB, b-HCH, p,p'-DDE and p,p'-DDT, RSDs varied between 10 and 18% for concentrations between the limits of determination (LOD, see table 1) and 1.0 mg/kg fat. For those PCDD, PCDF and PCB congeners occurring far above the LOD, RSDs were better than 10%, whereas for congeners present at levels around these LODs (i.e. TCDF, 1,2,3,7,8-PeCDF, 1,2,3,4,7,8,9-HpCDF, OCDF, PCB 52 and 189), higher RSDs between 20 and 100% should be taken into account.
When levels of PCDDs, PCDFs and dioxin-like PCBs are expressed in 2,3,7,8-TCDD equivalents using the International Toxic Equivalency Factors (I-TEF) for the PCDDs and PCDFs [10] and the Interim WHO-TEFs for the dioxin-like PCBs [11], RSDs were better than 5% for dioxin levels between 2.7 and 24.5 pg I-TEQ/g fat, about 5% for non-ortho PCB levels between 3.2 and 12.6 pg TEQ/g fat and approximately 10% for a mean TEQ-value of 12.9 pg TEQ/g fat based on levels of PCB congeners with IUPAC nos. 105, 118, 156, 157, 167, 180 and 189.
Repeatability data compared well with those found in method validation experiments [4-7]. Observed RSDs appeared to be negligible compared to the spread in the concentrations observed for the investigated samples.

3 Results and discussion

Table 1 summarizes the results of the measurements performed in this human milk survey. Levels for the OCPs, PCB congeners 28, 52, 101, 118, 138, 153 and 180 as well as for PCDDs and PCDFs can be compared with those resulting from previous human milk surveys, performed in 1972/73 (OCPs), 1983 (OCPs and PCBs) and 1988 (all). These comparisons are shown in the histograms in figures 1 to 3. Based on a preliminary interpretation of the questionnaire information (results will be published elsewhere), observable differences in these figures are presumably not caused by differences between the cohorts. The investigated cohort of 1993 appeared to be comparable with that of 1988.

Organochlorine pesticides

As can be concluded from Fig.1, median OCP levels show a downward trend in the period between 1972 and 1993. For a-HCH, g-HCH, TDE, o,p-DDT, p,p'-DDT, levels are below the limits of determination and confirm expectations based on previous surveys. With the exception of p,p'-DDE, other OCPs are declining to levels around the limits of determination of 0.01-0.03 mg/kg fat. HCB, b–HCH and p,p'-DDE are the only OCPs that are found at detectable levels in all samples investigated. It is evident that human body burdens of investigated OCPs are decreasing probably due to a reduction of concentrations in the environment and to a decreasing contamination of the human food chain.

Polychlorinated biphenyls

Figure 2 shows that, unlike the time trend for OCPs, no significant changes can be observed for the most dominating PCB congeners with IUPAC nos. 118, 138, 153 and 180. The stable levels are suggesting that human exposure levels of these particular compounds are not decreasing.
The international database to compare observed levels of the non-ortho and mono-ortho PCBs is rather limited since methods for the determination of these compounds have only recently been implemented. Observed levels of non-ortho PCBs are slightly lower than those found in a recently performed human milk study reported by Koopman-Esseboom and co-workers [12]. This may be due to analytical differences.

Polychlorinated dibenzodioxins and dibenzofurans

In the 1988 survey, a mean TEQ-level of 34.3 ± 3.4 pg I-TEQ/g fat (range: 30.6-39.6) was found for ten pooled samples of human milk. These samples were composed of nine to thirteen individual samples collected in areas with different degrees of urbanization in March 1988. Questionnaire items were equally distributed within the pooled samples (Student's T-Test). The 1993 survey shows a decrease of about 30% when compared to the average observed in 1988. This observation could be in line with the mean dioxin level of 30.2 pg I-TEQ/g fat recently found for a large set (N=176) of Dutch human milk samples collected in the period between June 1990 and June 1992 [12]. Decreasing dioxin levels have also been reported for Germany [13] suggesting that measures being undertaken in the past few years to reduce contamination of the environment and the human food chain [3] may already have resulted in a reduction of human body burdens of these compounds in these countries.
When levels of individual congeners are compared, remarkable differences can be observed for the various PCDD and PCDF congeners (see Fig. 3). This may be due to changes in exposure patterns (i.e. levels in food) and toxicokinetic factors (i.e. differences in biological half-lives). A comprehensive study to investigate the relative contribution of these factors is currently in progress.

4 References

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TABLE 1.
Levels of organochlorine pesticides, PCBs, PCDDs and PCDFs in individual samples of human milk from the Netherlands. Samples were collected in June 1993.

Component	TEF	dimension	LOD	No. of determ.	lowest	median	mean ± SD	90-PCT	highest
Fat content		wt.%	0.10	107	1.1	2.6	2.6 ± 0.9	4.0	5.8
HCB		mg/kg	0.01	89	0.01	0.04	0.05 ± 0.03	0.08	0.13
b-HCH		mg/kg	0.01	89	0.02	0.05	0.05 ± 0.03	0.07	0.26
g-HCH		mg/kg	0.01	89	n.d.	n.d.	n.d.	0.01	0.05
b-HEPO		mg/kg	0.02	89	n.d.	n.d.	n.d.	0.02	0.05
p,p'-DDE		mg/kg	0.03	89	0.13	0.33	0.50 ± 0.55	0.99	4.34
p,p'-DDT		mg/kg	0.03	89	n.d.	n.d.	0.01 ± 0.03	0.05	0.16
2378-TCDD	1	pg/g	0.6	103	0.9	2.8	3.1 ± 1.4	4.7	9.4
12378-PeCDD	0.5	pg/g	0.4	103	2.9	7.7	8.1 ± 2.9	11.4	22.4
123478-HxCDD	0.1	pg/g	0.4	103	0.8	7.7	8.6 ± 4.8	13.5	37.7
123678-HxCDD	0.1	pg/g	0.3	103	12.5	33.8	37.1 ± 16.7	52.6	119.0
123789-HxCDD	0.1	pg/g	0.5	103	1.7	6.0	6.9 ± 3.3	9.7	30.1
1234678-HpCDD	0.01	pg/g	0.5	103	8.8	39.4	44.9 ± 24.6	76.0	162.3
OCDD	0.001	pg/g	1.0	103	49.6	258.2	294.7 ± 167.7	501.8	939.5
2378-TCDF	0.1	pg/g	0.2	103	n.d.	0.4	0.4 ± 0.4	0.8	2.2
12378-PeCDF	0.05	pg/g	0.1	103	n.d.	0.2	0.2 ± 0.2	0.4	0.9
23478-PeCDF	0.5	pg/g	0.2	103	5.6	15.7	18.0 ± 7.5	27.1	45.9
123478-HxCDF	0.1	pg/g	0.3	103	n.d.	4.9	5.2 ± 2.3	7.8	16.6
123678-HxCDF	0.1	pg/g	0.3	103	n.d.	4.0	4.4 ± 2.1	6.5	16.2
234678-HxCDF	0.1	pg/g	0.3	103	0.5	2.2	2.4 ± 1.3	3.8	10.3
1234678-HpCDF	0.01	pg/g	0.5	103	n.d.	5.2	6.0 ± 3.1	9.8	22.1
1234789-HpCDF	0.01	pg/g	0.5	103	n.d.	n.d.	0.1 ± 0.1	0.2	0.8
OCDF	0.001	pg/g	0.5	103	n.d.	n.d.	0.3 ± 0.5	1.0	3.0
TEQ [PCDD/F]		pg/g		103	8.4	21.7	23.5 ± 8.9	34.5	63.1
PCB 77	0.0005	pg/g	1.0	104	n.d.	6.2	11.3 ± 21.8	20.5	197.0
PCB 126	0.1	pg/g	2.5	104	23.8	70.9	82.1 ± 36.2	129.5	207.4
PCB 169	0.01	pg/g	4.0	104	14.2	54.4	58.0 ± 24.7	90.8	158.4
TEQ [non-ortho PCBs]		pg/g		104	2.8	7.7	8.8 ± 3.8	13.7	21.7
PCB 28		ng/g	1.5	88	n.d.	2.5	3.2 ± 2.8	5.2	22.7
PCB 52		ng/g	1.0	86	n.d.	n.d.	0.2 ± 0.6	1.1	3.5
PCB 60		ng/g	0.5	89	n.d.	0.8	0.9 ± 0.7	1.6	3.9
PCB 74		ng/g	1.0	89	4.6	11.4	12.1 ± 5.2	17.3	31.8
PCB 101		ng/g	1.0	89	n.d.	1.0	0.9 ± 0.8	1.8	4.3
PCB 105	0.0001	ng/g	0.5	83	n.d.	3.5	4.0 ± 2.0	6.0	14.3
PCB 118	0.0001	ng/g	0.5	89	7.4	21.4	24.3 ± 11.4	42.0	63.6
PCB 138		ng/g	0.5	89	26.5	79.2	84.5 ± 34.8	124.7	207.2
PCB 153		ng/g	0.5	89	44.6	117.0	121.9 ± 46.2	178.6	273.7
PCB 156	0.0005	ng/g	1.0	89	4.6	13.4	14.1 ± 5.9	20.8	35.7
PCB 157	0.0005	ng/g	1.0	88	0.8	2.6	2.7 ± 1.2	4.1	6.9
PCB 167	0.00001	ng/g	0.5	89	1.5	3.4	3.8 ± 1.6	5.6	9.5
PCB 180		ng/g	0.5	89	18.2	59.9	64.6 ± 26.1	96.4	171.2
PCB 189	0.0001	ng/g	0.5	88	0.0	1.2	1.2 ± 0.9	2.1	4.5
TEQ [other PCBs]		pg/g		83	4.7	11.2	12.1 ± 4.7	18.6	27.9

Note: In at least 90% of all samples analysed, observed levels of a-HCH, dieldrin, TDE, o,p'-DDT , 123789-HxCDF and 1234789-HpCDF were below the limits of determination (LOD). These compounds were not included in the table.

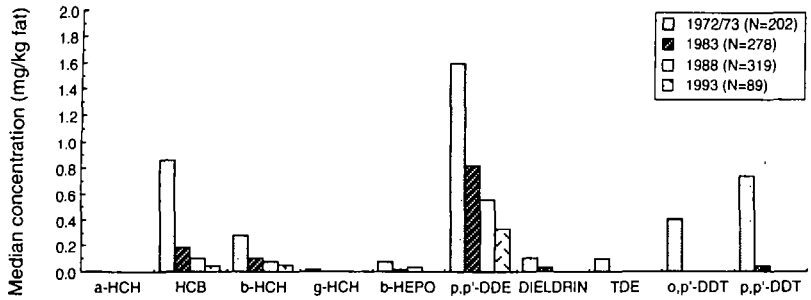

FIGURE 1 Trends in levels of organochlorine pesticides (median in mg/kg fat) in Dutch human milk.

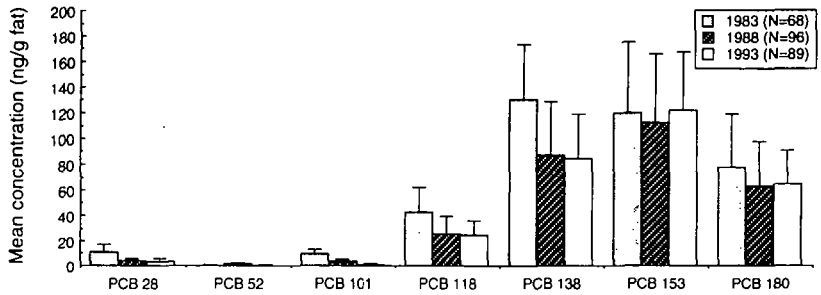

FIGURE 2 Trends in levels of PCBs (mean±SD in ng/g fat) in Dutch human milk. Y-error bars denote standard deviation of mean.

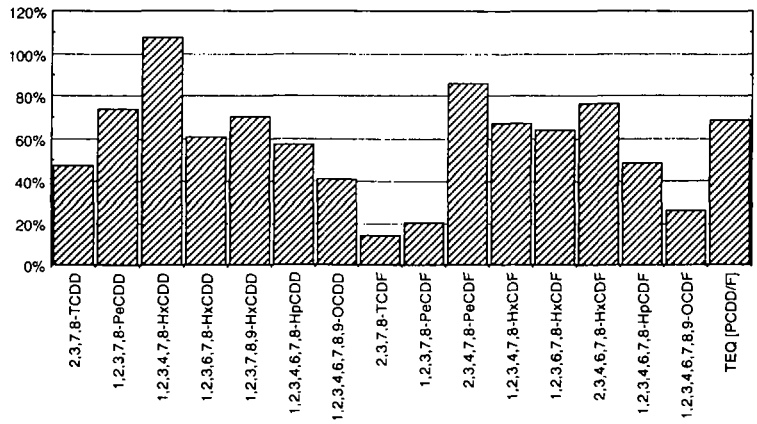

FIGURE 3 Change in mean levels of PCDDs and PCDFs (in pg/g fat) in Dutch human milk between 1988 and 1993. The figure shows observed mean levels in 1993 in percents of the mean concentrations found in 1988.