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RESULTS OF THE WHO-COORDINATED EXPOSURE STUDY ON THE LEVELS OF PCBs, PCDDs AND PCDFs IN HUMAN MILK

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Introduction

Since the mid-eighties, the WHO Regional Office for Europe has coordinated a comprehensive programme on possible health risks of polychlorinated biphenyls (PCBs), polychlorinated dibenzo-p-dioxins (PCDDs) and polychlorinated dibenzofurans (PCDFs). This programme was carried out in collaboration with other international organisations and national institutions, and concentrated particularly on the health risk of infants, due to exposure through contaminated breast-milk, and aiming to prevent and control environmental exposure to these chemicals. Because breast milk contains many lipid soluble compounds that are also present in mother's adipose tissue, it can also be assumed that the levels of PCDDs, PCDFs and PCBs in breast milk are representative for those in plasma, serum lipid and adipose tissue. Therefore levels of these contaminants in human milk do reflect the body burden and can thus be used as an indicator for the overall exposure of the general population. The collection of human milk is a non-invasive sampling method, offering a great advantage over the collection of other biological samples to assess overall human exposure. In addition, the high content of fat makes the extraction method easier and the precision of the measurements higher.

WHO has carried out a series of international exposure studies on levels of these contaminants in breast milk. The first WHO-coordinated exposure study took place in 1987-1988 (1), the second round in 1992-1993 (2). In the second round, in which concentrations of PCBs, PCDDs and PCDFs were determined in milk samples collected in a total of 47 areas from 19 different countries, mean levels in industrialised countries ranged from 10-35 pg I-TEQ/g. In developing countries mean levels were usually less than 10 pg I-TEQ/g milk fat. Compared to the outcome of the first round, a trend towards declining levels of PCDDs/PCDFs and PCBs in breast milk was found, with largest declines in countries with the highest initial levels.

The 3rd round of the WHO-coordinated exposure study has been initiated in March 2000. In order to collect data on more countries, also beyond the European region, the study was organised in collaboration with the International Programme on Chemical Safety (IPCS) and the WHO Global Environment Monitoring System/Food Contamination Monitoring and Assessment (GEMS Food).

The current exposure study has the following aims: a) produce reliable and comparable data on levels of PCBs, PCDDs and PCDFs in human milk for further improvement of the health risk assessment in infants; b) determine time trends in exposure levels in the countries and areas already studied in the first and second round of the study in the period 1987-88 and 1992-1993, respectively; c) provide a better overview of exposure levels in various countries and geographical areas; d) identify highly exposed local populations in relation to their daily intake for guidance on

risk management actions, including epidemiological follow-up studies; e) promote, if necessary, additional national studies to be closely linked with the present study through the use of the same protocol.
At the Dioxin2002 conference in Barcelona, the preliminary results of the 3d round of the WHO-coordinated exposure study, comprising eighteen countries, have been presented (3). Since then results for a number of additional countries have become available. This manuscript presents the final results of the WHO exposure study on levels of PCBs, PCDDs and PCDFs, comprising a total of 26 countries from all over the world.

Methods

Protocol. For the current study a protocol was used that was only slightly modified compared to the one used in the previous rounds. This protocol to be followed by the participating countries dealt primarily with number and type of samples, selection of donors, collection, storage and pooling of samples, and shipping of samples to the reference laboratory. Milk from well-defined groups of 10 mothers was collected and pooled. It was the aim of the study to include at least two different groups from each country, if possible a high- and low-exposure group. For selection of donating mothers the following criteria were applied: a) they should be primiparae, b) healthy, c) exclusively breastfeeding one child (i.e. no twins), and d) residing in the area for about 5 years. For further details of the study protocol the reader is referred to Van Leeuwen and Malisch (3).
Analysis. To ensure reliability of exposure data and to improve comparability of analytical results from different laboratories, the WHO has co-ordinated a number of inter-laboratory quality assessment studies. The fourth round on levels of PCBs, PCDDs and PCDFs in human milk was conducted between February 1996 and April 1997, with the objective to identify laboratories, whose results could be accepted by WHO for exposure assessment studies (4). Only the State Institute for Chemical and Veterinary Analysis of Food Freiburg met all the pre-set criteria for analyses of PCDDs, PCDFs, dioxin-like PCBs, marker PCBs and fat in human milk, and was thus selected as reference laboratory for the 3rd round of the WHO exposure study. For all samples of the current WHO exposure study the analysis and the rigid quality control programme was carried out as described before (5, 6).

Results and Discussion

One country (New Zealand) submitted samples in 2000. All other samples were received by the reference laboratory between the beginning of 2001 and the beginning of 2003. At last twenty six countries (Australia, Belgium, Brazil, Bulgaria, Croatia, Czech Republic, Egypt, Fiji, Finland, Germany, Hong Kong SAR, Hungary, Ireland, Italy, Luxembourg, New Zealand, Norway, Philippines, Romania, Russia, Slovak Republic, Spain, Sweden, The Netherlands, Ukraine, United States) participated in this round of the WHO exposure study. Altogether, 100 pooled samples were collected and analysed. From these, 97 samples represent different areas or concepts (e.g. reflecting different intake situations); three samples are included to confirm unusual findings.
The results of the third round as given in Table 1 show that, in general, variation between countries is much higher that within countries. Lowest levels of PCDDs/PCDFs and dioxin-like PCBs are found in countries on the Southern hemisphere (Fiji, Brazil, Philippines, Australia, New Zealand). Also for a number of European countries (Bulgaria, Croatia, Hungary, Ireland) and for the USA levels of PCDDs/PCDFs and dioxin-like PCBs are low, whereas comparatively high levels have been found in a number of Western European countries (Italy, Spain, Germany, Luxembourg, Belgium and the Netherlands) and in Ukraine. In the latter country a very high level of dioxin-like PCBs has been found. Of all countries Egypt shows the highest WHO-TEQ level with a remarkably high contribution of PCDD/Fs (22.3 pg WHO-PCDD/F-TEQ/g fat as median).

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Table 1. Levels of PCDD/Fs, dioxin-like PCBs and indicator PCBs in human milk

Country	PCDDs/PCDFs WHO-TEQ pg/g fat		PCBs WHO-TEQ pg/g fat		Sum indicator PCBs ng/g fat		Number of pools
	Median	*Range*	*Median*	*Range*	*Median*	*Range*	
Australia	5.57	5.39 – 5.75	2.89	2.52 – 3.26	30	25 – 36	2
Belgium	16.92	14.78 – 19.07	12.6	11.22 – 13.98	191	169 – 213	2
Brazil	3.92	2.73 – 5.34	1.77	1.30 – 12.28	16	10 – 97	11
Bulgaria	6.14	5.08 – 7.11	4.21	3.74 – 4.70	42	32 – 52	3
Croatia	6.40	5.99 – 6.80	7.17	6.82 – 7.52	135	121 – 150	2
Czech Republic	7.78	7.44 – 10.73	15.24	14.32 – 28.48	502	496 – 1009	3
Egypt	22.33	14.90 – 51.50	5.48	4.41 – 8.26	106	12 – 140	9
Fiji	3.34	3.17 – 3.51	1.75	1.70 – 1.80	17	16 – 19	2
Finland	9.44	9.35 – 9.52	5.85	5.66 – 6.03	91	84 – 98	2
Germany	12.53	11.14 – 12.72	13.67	12.80 – 14.31	220	188 – 238	4
Hong Kong SAR	8.69	5.80 – 10.09	4.73	2.80 – 6.58	45	16 - 80	11
Hungary	6.79	5.26 – 7.46	2.87	2.38 – 4.24	34	29 – 59	3
Ireland	7.72	6.19 – 8.82	4.57	2.72 – 5.19	60	41 – 65	4
Italy	12.66	9.40 – 14.83	16.29	11.02 – 19.33	253	195 – 323	4
Luxembourg	14.97	13.68 – 16.25	13.67	12.98 – 14.36	217	196 – 237	2
New Zealand	6.86	6.08 – 7.00	3.92	3.50 – 4.71	37	30 – 41	3
Norway	7.30	7.16 – 7.43	8.08	6.56 – 9.61	119	106 – 132	2
Philippines	3.94	3.64 – 4.24	2.38	2.22 – 2.54	26	26 – 26	2
Romania	8.86	8.37 – 12.00	8.06	8.05 – 8.11	173	165 – 198	3
Russia	9.36	7.16 – 12.93	13.45	12.92 – 22.95	126	84 – 311	7
Slovak Republic	9.07	7.84 – 9.87	12.60	10.72 – 19.49	443	331 – 621	4
Spain	11.56	10.24 – 18.68	9.42	6.93 – 17.94	241	162 – 467	6
Sweden	9.58	–	9.71	–	146	–	1
The Netherlands	18.27	17.09 – 21.29	11.57	10.90 – 13.08	192	178 – 210	3
Ukraine	10.04	8.38 – 10.16	19.95	14.10 – 22.00	136	103 – 148	3
USA	7.18	6.22 – 8.14	4.61	3.69 – 5.52	54	43 – 64	2

Also the levels of indicator PCBs vary widely between the countries. High levels were found in the Czech Republic, the Slovak Republic and Spain (median in these countries: range 400 - 500 ng/g fat), and low levels (range of median: 16 - 26 ng/g fat) in those countries also having low levels of PCDD/Fs and dioxin-like PCBs (Brazil, Fiji and the Philippines).
It can be seen that in a number of countries (Brazil, Fiji, Philippines, Hungary, Bulgaria, Croatia) the levels for all three groups of compounds (PCDD/Fs, dioxin-like PCBs and indicator PCBs) are consistently lower than in other countries. Other countries (Italy, Spain, Luxembourg, Germany, Belgium, The Netherlands) consistently show comparatively high levels for the three groups of compounds, but still there is considerable variation. Preliminary analyses of the results reveal varying profiles of the individual congeners, indicating different sources of contamination.
In addition, for a number of countries, pooled samples (areas) were identified as being different from others obtained from the same country. Usually these samples show a higher level of contamination, as is obvious from a comparison of the median values and the ranges as given in Table 1. The most striking examples can be found for Brazil, the Czech Republic and Egypt, with areas with a high level of contamination with dioxin-like and/or indicator PCBs. Further analysis of the pattern of the various congeners and the demographic data collected in the different countries will provide a clarification for these differences. From the results of countries participating in the current as well as in one of the previous WHO exposure studies, it can be concluded that the declining trend as observed before, continues. On average, the decline between the levels found in 2nd round in 1993 and the levels found in the current study, is about 40%.

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References
1. Environmental Health Series No 34 (1989): Levels of PCBs, PCDDs, and PCDFs in breast milk, WHO Regional Office for Europe, Copenhagen, Denmark.
2. Environmental Health in Europe No. 3 (1996): Levels of PCBs, PCDDs and PCDFs in human milk: Second round of WHO-coordinated exposure study, WHO Regional Office for Europe, Copenhagen, Denmark.
3. Van Leeuwen FXR and Malisch R (2002) Results of the third round of the WHO coordinated exposure study on the levels of PCBs, PCDDs and PCDFs in human milk, Organohalogen Compounds, Vol. 56, 311-316.
4. Interlaboratory Quality Assessment of Levels of PCBs, PCDDs and PCDFs in Human Milk and Blood Plasma – fourth round of WHO-coordinated study (2000), WHO Report EUR/00/5020352, WHO Regional Office for Europe, Copenhagen, Denmark.
5. Malisch R and van Leeuwen FXR (2002) Third round of WHO-coordinated exposure study: Analysis of PCDDs, PCDFs and PCBs in human milk, Organohalogen Compounds, Vol. 56, 317-320.
6. Malisch R, Bruns-Weller E, Knoll A, Fürst P, Mayer R and Wiesmüller T (2000) Results of an „emergency quality control study“ as confirmation of a PCDD/PCDF-contamination of milk and butter samples. Chemosphere 40, 1033 - 1040.