PCDD/Fs AND PCBs IN FOOD SAMPLES FROM GERMANY, FRANCE AND SPAIN – DATA AND PROPOSALS FOR EU LEGISLATION

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INTRODUCTION

The Community Strategy [i] to reduce the presence of dioxins and PCBs in feed and food comprises legislative measures which consist of three pillars: the establishment of maximum levels at a strict but feasible level in food and feed, the establishment of action levels acting as a tool for "early warning" of higher than desirable levels of dioxin in food or feed and the establishment of target levels, over time, to bring exposure of a large part of the European population within the limits recommended by the SCF. Council Regulation (EC) No. 2375/2001 sets maximum levels for food of animal origin and oils and fats [ii]. Council Directive 2001/102/EC and Directive 2002/32/EC set maximum levels for animal feed [iii, iv]. Action levels for food and animal feed were recommended by the Commission in March 2002 [v]. So far, these regulations and recommendations do not include dioxin-like PCBs. According to Council Regulation 2375/2001 the inclusion of dioxin-like PCBs should be reviewed by 31 December 2004. According to Council Directive 2001/102/EC, target levels should be set by 31 December 2004. A recent communication to the Council by the European Commission on the implementation of the Community Strategy summarizes the main progress over the first two years (end of 2001 to end of 2003) [vi].

As part of a cooperation between selected regions in Europe, Baden-Württemberg (south-western Germany), France and Catalonia (north-eastern Spain) support a joint programme to determine the levels of PCDD/Fs, dioxin-like PCBs and marker PCBs in different categories of food and animal feed and to estimate their contribution to the daily intake. The CVUA Münster (in Northrhine-Westfalia in western Germany) joined this cooperation to broaden the database. This paper summarizes results of food analyses in different regions of Germany, France and Spain and gives an orientation for further developments of the three pillars of legislative measures.

METHODS

The samples analysed by CVUA Freiburg and CVUA Münster are samples of the official food control in the German "Länder" Baden-Württemberg and Northrhine-Westfalia. The samples analysed by CARSO were part of a control programme for the French Ministry of Agriculture, Paris. CSIC analyses for the Spanish Ministry of Agriculture, Madrid. The analytical methods and data on reliability of the results are described elsewhere. All laboratories participated in numerous interlaboratory studies, among them some to show comparability of the results between the laboratories included in this study [vii, viii, ix, x].

RESULTS AND DISCUSSION

For reasons of limitations of the size of short papers, only few selected results of milk and milk products can be presented here to show the principles of the procedure to contribute proposals for EU legislation.

CVUA Freiburg analysed 2084 samples of milk and milk products in the years 1993 to 2003 for their PCDD/F-content, from these 1322 milk samples. Figure 1 shows the time trends in cow's milk. It is obvious that there is no significant change of the median dioxin content in the mid-90s, then an increase as result of the Brazilian citrus pulp contamination in 1997/98 [xi] and a slow decrease afterwards. Thus, for the purpose of revision of legislative measures, data for samples from 2000 – 2003 are summarized in table 1 and 2. During this period, the median decreased from 0.46 to 0.36 pg WHO-PCDD/F-TEQ/g fat.

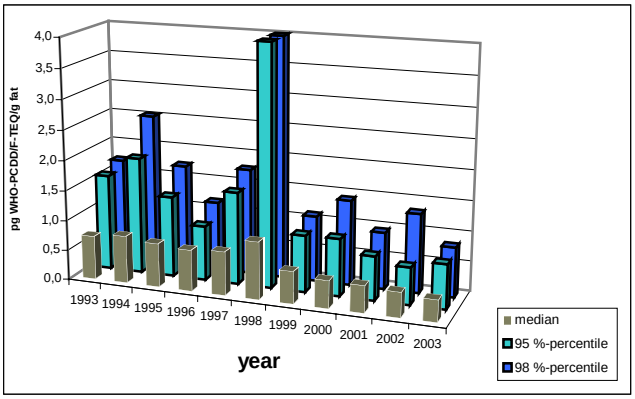

Figure 1: Time trend of WHO-PCDD/F-TEQ in cow's milk (results of CVUA Freiburg)

Council Regulation (EC) No. 2375/2001 sets a maximum level of 3 pg WHO-PCDD/F-TEQ/g fat for milk and milk products, including butter fat. The Commission has recommended an action level of 2 pg WHO-PCDD/F-TEQ/g fat. So far, these regulations and recommendations do not include dioxin-like PCBs.

Milk and milk products are good indicators for the contamination of persistent organic pollutants (POPs) in the food chain. Thus, butter has been used for comparison of the PCDD/F contamination in several countries [xii, xiii] including new EU member states [xiv]. EU Scientific Committee on Food (SCF) has considered the national figures contained in the EU SCOOP database [xv] selecting only data since 1995 and calculated frequency distributions for a number of foods [xvi]. The mean concentrations of two subgroups of milk and its products ranged approximately from 0.6

to 1.0 pg I-TEQ/g or 0.6 to 1.3 pg PCB-TEQ/g, lipid basis. The upper confidence limits were in the order of 1 pg I-TEQ/g fat and 2-10 pg PCB-TEQ/g fat for dioxin-like PCBs.

In comparison to the EU SCF data, the average dioxin content decreased to a mean of 0.40 pg WHO-PCDD/F-TEQ/g fat in Baden-Württemberg reflecting now the actual low range of background contamination (range 0.2 to 1.0; upper bound). The 95 %-percentile which was used to set the maximum limits in legislation is below 1 pg WHO-PCDD/F-TEQ/g fat in all samples of 2000 – 2003. In the years 2001 – 2003, even the 98 %-percentile is below 1 pg WHO-PCDD/F-TEQ/g fat (95 %- percentile: below 0.75 pg WHO-PCDD/F-TEQ/g fat). This shows clearly that the maximum limits and action levels valid at the moment are quite high and could be reduced considerably.

	pg WHO-PCDD/F-TEQ/g fat			
no. of samples	85	82	87	99
year	**2000**	**2001**	**2002**	**2003**
minimum	0,23	0,28	0,20	0,20
median	**0,46**	**0,45**	**0,41**	**0,36**
mean	0,54	0,49	0,47	0,40
90 %-percentile	0,82	0,64	0,58	0,55
95 %-percentile	0,95	0,73	0,62	0,74
98 %-percentile	1,42	0,94	1,32	0,83
maximum	1,61	1,54	3,17	1,09

Table 1: Results of dioxins in milk samples (years 2000 – 2003) from Baden-Württemberg

	PCB-TEQ pg/g fat	PCDD/F-TeQ pg/g fat	sum TEQ PCDD/F + PCB pg/g fat	contrib. (%) of PCB-TEQ to sum TEQ
milk 2001 - 2004				
no. of samples	28	28	28	28
min	0,36	0,26	0,85	30,3
25 %-percentile	0,77	0,36	1,13	64,2
median	0,93	0,43	1,41	69,3
mean	0,97	0,45	1,42	67,5
90 %-percentile	1,37	0,58	1,85	77,1
95 %-percentile	1,57	0,60	2,09	80,5
98 %-percentile	1,70	0,71	2,21	81,2
max	1,73	0,83	2,27	81,9
butter (2002)				
no. of samples	21	21	21	21
min	0,23	0,23	0,46	50,3
25 %-percentile	0,79	0,31	1,13	66,5
median	0,87	0,33	1,19	71,4
mean	0,88	0,35	1,24	70,3
90 %-percentile	1,04	0,47	1,49	76,6
95 %-percentile	1,23	0,47	1,75	78,9
98 %-percentile	1,46	0,50	1,86	81,9
max	1,62	0,52	1,93	84,0

Table 2: Results of dioxin-like PCBs in milk and milk products from Baden-Württemberg

In 28 milk samples and 21 butter samples, also PCB-TEQ was determined (table 2). The average contamination is about 0.9 – 1 pg WHO-PCB-TEQ/g fat (range 0.23 to 1.73; upper bound). These and previous data [xvii] are in accordance with reports that in Germany PCDD/F contribute only about 30 % of the total TEQ value whereas about 70 % are caused by PCB-TEQ when a mean of 26 dairy products 0.77 pg WHO-PCDD/F-TEQ/g fat (range 0.55 to 1.16) and of 1.83 pg WHO-PCB-TEQ/g fat (range 0.71 – 3.04) was found [xviii].

How difficult it might be for authorities to derive the usual background contamination from a data set can be shown with the French data (table 3): CARSO has analysed 683 milk samples for their PCDD/F-content between 2001 and 2003 for the French Ministry of Agriculture. These data include many samples from partly highly contaminated areas which cannot be taken into account for discussions of maximum limits. Highly contaminated milk samples, however, show the need for such harmonized maximum and action limits. Special programs in contaminated areas were run in 2001 and 2003. After elimination of these results, the frequency distribution showed considerably

lower values (in table 3: comparison of upper part for samples not marked as contaminated with lower part for all samples). However, the remaining data set still includes some highly contaminated samples not representing the usual background contamination. The usual background contamination in France can be derived more reliably from randomly taken milk samples which were analysed for PCDD/F and PCBs (also included in table 3). The 59 samples of 2002 and 95 samples of 2003 show comparable results to the Baden-Württemberg data. As a results for the legislative measures, the scientific evaluation of data sets is very complex and a big problem if scientific committees receive data without detailed knowledge of the origin of samples.

Carefully analysing the data set of the involved laboratories it can be concluded that the average background contamination in milk and milk products is now below 1.5 pg WHO-PCDD/F-TEQ/g fat (98 % percentile) respectively 1.0 pg WHO-PCDD/F-TEQ/g fat (95 % percentile) and below 2.0 pg WHO-PCB-TEQ/g fat (close agreements of maximum, 98 % percentile and 95 % percentile). This could give an orientation for future amendments in legislation.

Using the comprehensive data set of the here contributing laboratories, table 4 shows the upper end of the usual background contamination.

	PCDD/F-TEQ	**PCB-TEQ**	**sum TEQ**	**unit**
cow's milk and milk products	1,5	2,0	3,0	pg/g fat
eggs from caged chicken	1,0	1,5	2,0	pg/g fat
eggs from free-range chicken	2,0	2,0	4,0	pg/g fat
meat from pigs	0,5	0,5	1,0	pg/g fat
meat from poultry	1,0	1,0	2,0	pg/g fat
meat from ruminants	2,0	2,0	4,0	pg/g fat
muscle meat of fish < 1 % fat	0,2	1,0		pg/g fw
muscle meat of fish with 1-3 % fat	0,6	3,0		pg/g fw
muscle meat of fish with 3-5 % fat	1,0	5,0		pg/g fw
muscle meat of fish with 5-10 % fat	1,2			pg/g fw
muscle meat of fish > 10 % fat	4,0			pg/g fw

Table 4: Upper end of the usual background contamination

	PCDD/F-TEQ	PCDD/F-TEQ	PCDD/F-TEQ	PCB-TEQ	sum TEQ	% contr. PCB to sum TEQ	PCDD/F-TEQ	PCDD/F-TEQ	PCB-TEQ	sum TEQ	% contr. PCB to sum TEQ
year	2001	2002	2002	2002	2002	2002	2003	2003	2003	2003	2003
subgroup	1)	1)	2)	2)	2)		1)	2)	2)	2)	
not marked as contaminated											
No samples	160	180	59	58	59	58	155	101	95	101	95
min	0,36	0,12	0,12	0,14	0,26	42	0,12	0,12	0,18	0,30	33
25 %-percent.	0,51	0,46	0,36	0,62	0,99	57	0,41	0,37	0,63	1,00	60
median	0,67	0,66	0,46	0,71	1,18	60	0,52	0,46	0,93	1,33	66
mean	1,79	1,09	0,50	0,80	1,29	61	0,95	0,50	1,11	1,54	66
90 %-percent.	3,84	1,90	0,76	1,30	1,95	70	1,35	0,79	2,01	2,57	78
95 %-percent.	8,04	3,09	0,87	1,44	2,12	72	2,09	0,84	2,42	3,12	80
99 %-percent.	8,04	3,09	0,87	1,44	2,12	72	2,09	0,84	2,42	3,12	80
max	28,34	9,61	0,97	1,56	2,53	79	17,47	1,08	4,61	5,03	92
all samples											
No samples	216	312					155				
min	0,36	0,12					0,12				
25 %-percent.	0,57	0,58					0,41				
median	1,03	1,40					0,52				
mean	4,09	2,60					0,95				
90 %-percent.	10,28	6,11					1,35				
95 %-percent.	17,87	8,79					2,09				
99 %-percent.	17,87	8,79					2,09				
max	36,73	20,52					17,47				

Table 3: Results of milk samples from France (in pg WHO-PCDD/F-TEQ/g fat, pg WHO-PCB-TEQ/g fat or pg sum WHO-TEQ/g fat; subgroups: 1) analysed only for PCDD/F, 2) analysed for PCDD/F and PCB)

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