

LEVELS OF PCDD/PCDFs IN LUXEMBURGISH FOOD OF ANIMAL ORIGIN AND IN MOTHER'S MILK

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Introduction

Since the strong contamination of a Belgian feeding stuff with polychlorinated dibenzo(p)dioxins (PCDDs) and dibenzofurans (PCDFs) in 1999 which led to the contamination of a number of foodstuffs of animal origin all over Europe, the problem of dioxins in foodstuffs found its place in the centre of the public discussion.^{1,2} As one of the consequences of the Belgian feeding stuff scandal, the European Commission in the meantime enacted a number of directives including maximum levels for PCDD/F in feeding stuff and food.^{3,4} These limit values will have to be observed throughout Europe from July 1st, 2002 on.

In 2000/2001 the Ministry of Health of the Grand Duchy of Luxembourg conducted a study on the levels of PCDF/Ds in certain foods of animal origin and in mother's milk from different regions of the country. The impulse of this study came from a local, in the South of Luxembourg implemented association (Stop Dioxines), linked to the national Ecological Movement. It was the aim of the study to elaborate the present PCDD/F content in food and to find out whether there are regional differences. In all, 23 cow's milk samples, 16 hen's egg samples, 11 rabbit-meat samples and 22 mother's milk samples were collected and analysed for PCDD/Fs. In this paper the PCDD/F data of the study is reported and the food values are compared with those of other European countries and with the coming EC maximum levels.

Materials and Methods

The food sampling was performed by Biomonitor, Luxembourg, in behalf of the Luxembourgish Ministry of Health. With the exception of three commercial cow's milk samples, all food samples were taken directly from the producers. The mother's milk samples were collected by the association Initiativ Liewensufank, Luxembourg. All women were primiparous at the age of 22 to 40 years (mean of 29.7 years).

The samples were analysed by the GfA for PCDD/Fs by means of fat extraction, gravimetric determination of the fat fraction, chromatographic defatting of the fat extract, clean-up of the remaining fraction on different adsorbents, analysis of the purified extracts by means of capillary gas chromatography / high resolution mass spectrometry (HRGC/HRMS) and quantification via internal ¹³C₁₂-labelled standards (isotope dilution).⁵ The methods are DIN EN ISO/IEC 17025 accredited and are routinely applied by the GfA for the analysis of food, feed and human samples.

Results and Discussion

All TEQs are reported as levels in fat (pg TEQ/g fat). TEQs were calculated according to the I-TEQ and WHO-TEQ scheme (TEFs for humans). For both schemes, non-detects were included by taking the

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full detection limit. The TEQ values determined for the different numbers of cow's milk, rabbit's meat, hen's egg and mother's milk samples from different parts of Luxembourg are shown in Table 1.

Table 1. Lipid-based TEQ values of up to 23 samples from different matrices collected in 2000/2001 in the Grand Duchy of Luxembourg

Sample	Hen's eggs		Cow's milk		Rabbit's meat		Mother's milk	
	I-TEQ	WHO-TEQ	I-TEQ	WHO-TEQ	I-TEQ	WHO-TEQ	I-TEQ	WHO-TEQ
Pg TEQ/g fat basis								
1	1,76	1,99	0,81	0,95	0,42	0,49	12,0	14,0
2	5,46	6,12	0,69	0,80	0,14	0,16	9,87	11,5
3	0,87	0,98	0,77	0,91	0,33	0,37	11,3	13,4
4	1,07	1,23	0,60	0,70	0,16	0,18	15,1	17,4
5	1,00	1,13	0,61	0,72	0,61	0,67	18,2	21,0
6	1,37	1,54	0,93	1,07	4,88	5,16	19,5	22,7
7	2,36	2,65	0,48	0,54	0,39	0,43	12,8	14,9
8	2,31	2,69	0,60	0,69	0,73	0,83	14,4	16,9
9	4,93	5,38	0,57	0,66	0,36	0,43	18,2	21,4
10	1,22	1,33	0,75	0,87	0,45	0,54	13,0	15,3
11	3,16	3,43	0,65	0,73	0,22	0,25	17,9	20,8
12	1,64	1,87	1,06	1,33			9,21	10,8
13	1,27	1,41	0,56	0,66			15,0	17,4
14	12,3	10,7	0,51	0,58			10,3	11,9
15	0,85	0,97	1,08	1,28			18,1	21,1
16	0,70	0,78	0,57	0,65			17,2	19,7
17			1,14	1,28			18,9	21,9
18			0,76	0,86			17,9	20,9
19			0,28	0,32			13,5	15,8
20			0,74	0,86			17,5	20,5
21			0,50	0,57			12,1	14,1
22			0,48	0,55			21,8	25,4
23			0,39 ^a	0,48 ^a				

a cream sample

The dioxin content of the 23 Luxembourgish cow's milk samples were between 0,32 and 1,33 pg WHO-TEQ/g fat (mean of 0,79 pg WHO TEQ/g fat). This range fits well with the data found in other European countries (0,3-2,1 pg TEQ/g fat).^{6,7,8} The EC limit values for the maximum PCDD/F level of cow's milk and milk products will be 3 pg WHO-TEQ/g fat.⁴ The recommended EC action level which would entail measures to identify and reduce the source of contamination when exceeded, is 2 pg WHO-TEQ/g fat.⁹ The data of Table 1 show that all the PCDD/F-TEQs of the Luxembourgish cow's milk samples tested were below both, the coming EC maximum levels and the recommended action levels.

The situation becomes more complicated in case of the hen's eggs. Mainly values between 0.78 and 3.43 pg WHO-TEQ/g fat were found but also higher concentrations of 5.4, 6.1 and 10.7 pg WHO-TEQ/g fat were detected in three cases. While eggs from caged chicken usually show somewhat lower PCDD/F concentrations than those from foraging poultry, the two highest values of this study resulted from caged chickens. Data from other European countries usually show a range between 0.5 and 2.7 pg I-TEQ/g fat for caged chickens.⁸ The 2002 maximum tolerable EC levels will be 3 WHO-TEQ/g fat for eggs from caged chickens.⁴ For free-range and semi-intensive eggs the limit values will be valid from 10 January 2004.

Further analyses were carried out to verify the highest egg levels and to find out the sources of contamination (e.g. by feed analysis). The eggs from the producer with the former highest value were at 3.4 pg WHO-TEQ/g fat when analysing subsequent eggs. Although the reason for the contamination couldn't be cleared finally, there was a certain probability that former use of caolinite was the reason for the preceding high PCDD/F concentration in the eggs. In the second case the subsequent analysis of eggs showed even much higher values than before (18.9 pg WHO-TEQ/g fat). Fumes from a wood-burning oven were suspected to be the source of the PCDD/F contamination. The oven was operated in the hen-coop to dry and heat up feed and the fumes were not directly led to the outside. The producer was asked to stop this and not to consume the eggs. Further investigations shall verify this potential source of contamination.

With the exception of one value, the lipid-based PCDD/F-TEQs of the rabbit's meat samples were below 1 pg WHO-TEQ/g fat. The elevated sample showed a PCDD/F-TEQ of 5.16 pg/g fat and came from the producer who used an old wood-burning oven to heat up and dry feed. No reference values are available in the literature for meat of rabbits. When assessing the rabbit's meat data by means of the EC limit values for poultry and farmed game, it would be in compliance with the corresponding maximum PCDD/F level of 2 pg WHO-TEQ/g fat⁴ and with the recommended action level of 1.5 pg WHO-TEQ/g fat⁹, however, with the exception of the one sample mentioned above.

The mother's milk samples from Luxembourg showed PCDD/F concentrations between 10.8 and 25.4 pg WHO-TEQ/g fat (mean 17.7 pg TEQ/g fat). When considering the mean values for the various regions of Luxembourg, the differences are to be found of low significance. The overall range of TEQs is similar to those found in mother's milk samples from neighbouring or other industrialised countries (10-35 pg I-TEQ/g fat).^{8,10,11,12}

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