Levels of PCDDs, PCDFs and PCBs in International Fast Food Samples

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Introduction

Following the so called 'Dioxin crisis', which took place in spring 1999 in Belgium due to the introduction of some Aroclor 1260 oil in recycling centers used as fat collecting units for animal feeding-stuffs processing[1], several monitoring campaigns have been carried out in order to implement a continuous control of food-stuffs quality. Many different matrices, such as milk, eggs, meat, fishes, and also animal feeding-stuffs, have been investigated. Data concerning background level of polychlorinated dibenzo-*p*-dioxins (PCDDs), dibenzofurans (PCDFs) and polychlorinated biphenyls (PCBs) present in these matrices are now available and can be used to make estimation of typical dietary intakes for general population[2].

Dietary habits can, however, be significantly different depending on which part of the population is considered. As fast food type meals become more and more important in industrialized countries, especially for the younger part of the population, there is some interest in establishing PCDD/F and PCB background levels for such types of food. Very few studies actually have reported data in this area and, as far as we know, no data are available for Belgium and surrounding countries. For these reasons, we carried out a survey of PCDD, PCDF and PCB levels some well distributed fast food items. In addition to national samples, international sampling was carried out to allow comparison of values issued from a same laboratory performing in well defined conditions.

Materials and methods

Samples. Fast food samples were purchased at outlets between August and December 2001. They were cooled down at room temperature, then frozen in their commercial package and separately stored in labeled plastic bags. When international shipping was required, frozen samples were placed in thermo boxes containing dry ice in order to remain frozen until delivery. Sampling sites were Sydney (Australia), Atlanta (GA), Ithaca (NY), Bratislava (Czech Republic), Zurich (Switzerland), Waterloo (Belgium), Bruges (Belgium) and Liege (Belgium). The following items were collected: McDonald's Big Mac®, McDonald's Crispy Chicken Deluxe®, McDonald's Fish Filet Deluxe® and Pizza Hut's Personal Pan Pizza Supreme® (no anchovies).

Sample preparation. Each separate samples (20-30 g portions) were entirely carved out and homogenized using dissecting and/or mortar equipment and then frozen in liquid nitrogen before freeze-drying. The freeze-dried products were ground in order to obtain a fine powder. 3 g of sodium sulfate were added to 10-15 g (dry weight) samples and the mixture was extracted by pressurized liquid extraction (PLE) using a Dionex (Sunnyvale, CA, USA) ASE 200 extractor. Samples were extracted with 20 ml of hexane per cycle, 5 min cycle time, 2 cycles per extraction, pressure of 1500 psi. Fat extracts were dried on sodium sulfate prior gravimetric determination of the lipid content. As for our validated routine analyses of food-stuffs, aliquots of about 4 g of fat were used for the clean-up step. Automated clean-up was carried out on the Power-Prep™ system

(Fluid Management Systems, Waltham, MA, USA) using disposable columns[3]. The system operated using high capacity disposable multi-layer silica columns (acidic, basic and neutral), basic alumina columns and PX-21 carbon columns. Fractionation allowed isolation of a first fraction containing 7 marker (Aroclor 1260) and 8 mono-*ortho* PCBs as well as a second fraction containing 17 PCDD/F and 4 non-*ortho* PCB (coplanar, cPCBs)[4].

Analyses. Physico-chemical analyses of PCDD/F and cPCBs were performed by gas chromatography coupled to high resolution mass spectrometry (GC-HRMS) using a MAT95XL high-resolution mass spectrometer (Finnigan, Bremen, Germany) operating under accreditation control. Analyses of the fractions containing marker and mono-*ortho* PCBs were carried out using gas chromatography-tandem mass spectrometry (GC-MS-MS) on a Saturn 2000 GC-MS-MS mass spectrometer (Varian, Walmut Creek, KS, USA) (Pirard et al., 2002). A RTX-5SIL-MS (30 m x 0.25 mm I.D., 0.25 µm film thickness) capillary column (Restek, Evry, France) was used in both systems.

Results and Discussion

Levels: Table 1 shows the PCDD, PCDF as well as non- and mono-ortho PCB concentrations expressed in WHO-TEQ and lipid-corrected. Due to the very low background levels present in samples and, following recommendations concerning the report of dioxin concentrations in food-stuffs, we applied the lower-, middle- and upper bound approach to present results[5]. As it appears in Table 1, PCDD/F and PCB concentrations for the four investigated types of meals were very low. For both fish and chicken-based meal, lower bound values were usually lower than the ones accounting for the other two meals. The low levels recorded for our chicken-based samples were quite different from those reported in a Korean study in which levels in fast food chicken were more than forty-times higher than those in hamburgers[6].

Major contributors to the PCDD/F TEQ were 1,2,3,4,7,8-HxCDD, 1,2,3,6,7,8-HxCDD, 2,3,7,8-TCDF and 2,3,4,7,8-PeCDF. Although OCDD was sometimes present in significant concentration, other congeners were usually not present and 2,3,7,8-TCDD was never recorded in any sample. No major differences appeared among cities where samples were collected. One can however notice that samples originating from Belgium were some of the lower ones. Lower and upper bound values for coplanar PCBs (PCB-77, PCB-81, PCB-126, PCB-169) were respectively 0.00 pg WHO-TEQ/g fat and 0.65 pg WHO-TEQ/g fat considering all types of samples. PCB-77 and PCB-81 concentrations were always below LOQs. In very few samples, PCB-126 and PCB-169 were recorded at low levels (<5 pg/g fat). Considering all types of meals, the relative contributions of PCDD/Fs and PCBs to the TEQ were 31% and 69%, respectively.

Measured concentrations of marker (Aroclor 1260 congeners) PCBs and mono-*ortho* PCBs were very low and it was not possible to extract any significant congener distribution or characteristic pattern. It was not neither any significant differences between samples issued from different countries. In average, considering the sum of PCB concentrations, hamburger and chicken based meals appeared to have slightly higher levels than fish based and pizza meals. Those values ranged between 30 and 84 ng/g fat.

For the present study, we performed analyses using sample sizes (on a fat content basis) that are routinely used for analysis of dioxins and PCBs in food stuffs with validated methods. It seems that such fast food matrices would require more than 4 g of lipids to allow more comfortable identification of very low level compounds. If performing analysis on much higher sample amounts is not really a problem for such easily available samples, the sample preparation procedure (extraction and clean-up) and the analytical procedure itself would be complicated.

Table 1: PCDD/F and PCB concentrations (pg WHO-TEQ/g fat) found in international fast food samples.

	PCDD/F and PCB TEQ concentrations (pg WHO-TEQ/g fat)											
	McDonald's Big Mac®			McDonald's Crispy Chicken Deluxe®			McDonald's Fish Filet Deluxe®			Pizza Hut's Personal Pan Pizza Supreme®		
	Lower Bound	Middle Bound	Upper Bound	Lower Bound	Middle Bound	Upper Bound	Lower Bound	Middle Bound	Upper Bound	Lower Bound	Middle Bound	Upper Bound
Sydney	0.08	1.50	2.91	0.00	1.43	2.85	0.31	1.67	3.04	0.02	1.44	2.86
Atlanta	0.26	1.61	2.96	-	-	-	0.00	1.42	2.85	0.03	1.45	2.86
Ithaca	0.15	1.55	2.96	0.20	0.86	1.53	0.05	1.47	2.90	2.07	3.43	4.79
Zurich	1.04	2.10	3.17	0.09	1.50	2.91	0.38	1.80	3.23	1.94	3.34	4.74
Bratislava	0.90	2.11	3.33	0.33	1.71	3.09	0.06	1.47	2.89	0.14	1.51	2.89
Bruges	-	-	-	-	-	-	-	-	-	0.05	1.47	2.89
Waterloo	0.07	1.49	2.90	0.05	1.47	2.88	0.02	1.45	2.88	0.01	1.44	2.87
Liege	0.05	1.46	2.88	0.83	1.88	2.92	0.09	1.50	2.92	0.02	1.45	2.87

Lower bound values: assuming values for all ND congeners and values <LOQ = 0. Congeners were ND when S/N was < 3, LODs were evaluated using method blank (mBC) values or values of smaller added concentration giving a S/N > 3, LODs were defined as this S/N > 3 + 3 SD. LOQs were defined as this S/N > 3 + 10 SD. *Middle bound values:* using 1/2 LOD values for ND congeners. *Upper bound values:* using LOD values for ND congeners

Table 2: Estimated contribution of selected fast food items to the PCDD/Fs, non-*ortho* PCBs and, mono-*ortho* PCBs dietary intake.

Mean values (SD)	McDonald's Big Mac®	McDonald's Crispy Chicken Deluxe®	McDonald's Fish Filet Deluxe®	Pizza Hut's Personal Pan Pizza Supreme®
Middle Bound (pg WHO-TEQ/g fat)	1.69 (0.66)	1.47 (0.74)	1.54 (0.56)	1.94 (0.89)
Lipid Content (%)	12.9 (1.1)	10.8 (0.9)	12.9 (2.4)	11.8 (1.3)
Middle Bound (pg WHO-TEQ/g whole weight)	0.22 (0.09)	0.16 (0.08)	0.20 (0.08)	0.23 (0.09)
Serving Size (g)	199.3 (8.1)	174.6 (27.5)	148.1 (9.8)	333.1 (47.4)
Middle Bound (pg WHO-TEQ/serving)	44.55 (9.67)	26.88 (4.57)	29.59 (7.23)	73.39 (24.16)
Dietary Intake (pg WHO-TEQ/kg bw/month)[1]	6.70 (1.49)	4.14 (0.70)	4.55 (1.11)	11.29 (3.72)
Dietary Intake (pg WHO-TEQ/kg bw/month)[2]	14.52 (3.22)	8.96 (1.52)	9.86 (2.41)	24.46 (8.05)
% PTMI[3] for adults	9.6	5.9	6.5	16.1
% PTMI[3] for a 10 years-old child	20.7	12.8	14.1	34.9

[1]Adult, assuming a 65 kg body weight and a consumption rate of 10 servings per month

[2]10 years-old child, 30 kg , assuming a 10 kg body weight and a consumption rate of 10 servings per month

[3]FAO/WHO proposed provisional tolerable monthly intake of 70 pg WHO-TEQ/kg bw/month

Actually, increasing the sample size will also require larger quantities of solvent and glassware, resulting in potentially higher blank levels with negative effect on LOQs. Reduction of the gap between lower and upper bound values is consequently not easily attainable for such non-common type of samples.

Estimated Dietary Intake (EDI): Estimation of the intake due to consumption of fast food items is not trivial since consumption habits can vary significantly between, not only age groups but also inside selected population group depending on occupational parameters. As non significant differences arose between countries, we used average values for each items (Table 2). Calculations were based on middle bound concentration values since it is believed to be the more representative[7] as well as the more suited approach for risk assessment and intake estimations. Dietary intakes were calculated for adults but also for ten years-old child for which consumption of this type of food is quite popular. For adults, an average estimated intake was 6.7 pg WHO-TEQ/kg bw/month, including consumption of all types of analyzed meals, representing 9.5% of the PTMI (provisional tolerable monthly intake)[8]. For child, a value of 14.5 pg WHO-TEQ/kg bw/month was obtained, representing 20.6% of the PTMI. Similar results were reported in a study carried out in the U.S. few years ago[9].

Conclusions

Background concentrations of PCDDs, PCDFs, non-*ortho* PCBs, and mono-*ortho* PCBs in fast food samples collected in different countries appeared to be low. Most of congeners were non-detected or below LOQs. No major differences were recorded from country to country. For analyses of such food, larger sample sizes should be considered regarding classical foodstuffs in order to increase the number of detected compounds. Such approach may have significant impact on the sample preparation steps. Estimated intakes shown to be in the range of what is currently expected for usual foodstuffs suggesting that no unusual contamination occurs in the fast food branch.

Acknowledgements

This research was supported by the *Fonds pour la Formation à la Recherche dans l'Industrie et l'Agriculture (F.R.I.A.),* the *Fonds National de la Recherche Scientifique (F.N.R.S.),* the *Region Wallonne* and the University of Liege. Authors deeply acknowledge Dr. Kocan, Mr. Hughes and Mr. Piro for their kind assistance in international sampling.

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