Dioxins and PCBs in a Swedish food market basket study - dietary intake estimations and temporal trends

Lignell S[1], Aune M[1], Cantillana T[1], Darnerud PO[1], Fridén U[1] and Glynn A[2]

[1]National Food Agency, Uppsala, Sweden, SE-751 26, sanna.lignell@slv.se, [2]Swedish University of Agricultural Sciences, Uppsala, Sweden, SE-750 07

Introduction

The Swedish National Food Agency (NFA) regularly performs market basket (MB) studies which include analysis of toxic compounds in commonly purchased foods on the Swedish market. Here we present results from analyses of polychlorinated biphenyls (PCBs), polychlorinated dibenzo-*p*-dioxins (PCDDs) and polychlorinated dibenzofurans (PCDFs) in food from a MB study performed in 2015.[1] The aim of the study was to estimate the mean per capita intake of PCBs and PCDD/Fs in the Swedish population due to background contamination. Since the study is the fourth in a series, it also gives the opportunity to study time trends. Results are important for risk assessment and for risk management activities.

Materials and methods

Sample collection and preparation

The basis for sampling of food items for the MB study was per capita-consumption data, derived from Swedish producers and trade reported by the Swedish Board of Agriculture[2]. Selected food items, representing food categories consumed on average more than 0.5 kg per person per year, were purchased in Uppsala in 2015 from five major grocery chains. From each of the five purchased food baskets, the food items were divided in 12 main food categories and, for reasons of homogenization, dairy products were further split in liquids and solids.

From each food item, one percent (by weight) of the yearly per capita consumption was taken out for homogenate preparation and subsequent analysis. For food items where wastage could be supposed, inedible parts such as bone, skin, etc. were removed prior to homogenization. The weighed amounts from every food item within a food category were subsequently mixed together and carefully blended (by using a household mixer). From these food category homogenates, samples were taken for analyses.

Analysis

PCDD/F congeners (N=17), dioxin-like PCBs (N=12) and non-dioxin-like PCBs (N=6) were analysed in the six food categories that are main contributors to exposure, i.e. egg, fish, meat, liquid dairy products, solid dairy products and fats/oils. One sample per food category and basket/grocery store was analyzed, resulting in a total of 30 samples. The samples were analysed at the NFA using a previously described method[3] partly modified for solid matrices. Briefly the samples were extracted using liquid-liquid or pressurized liquid extraction, followed by clean-up and fractionation using a PowerPrep™-system from Fluid Management Systems (MA, USA). The final analysis was performed with isotope dilution technique using gas chromatography coupled to high resolution mass spectrometry (GC-HRMS). A number of control samples were analysed together with the samples to verify the accuracy and precision of the measurements.

Intake estimations and temporal trends

The per capita average intake of PCDD/F and PCBs was calculated on the basis of analytical results and average annual per capita consumption amounts. The calculations are described in detail elsewhere.[4,5] Non-quantified contaminant levels were extrapolated to 0 (lower bound), to half the quantification limit (LOQ) (medium bound) or to the LOQ (upper bound) in the calculations. The non-dioxin-like PCB congener PCB 153 was used as a marker for total PCB since earlier market basket studies have shown strong correlations between per capita intake of PCB 153 and total PCB (sum of 28 congeners).[6] The PCDD/F and dioxin-like PCB levels are expressed as toxic equivalents (PCDD/F/PCB TEQ) using WHO 2005 toxicity equivalent factors.[7]

Calculated intakes of PCDD/F/PCB TEQ and PCB 153 from the 2015 market basket study were compared with intakes estimated in similar studies performed by the NFA in 1999, 2005 and 2010.[4,6,8] Temporal trends were investigated using log-linear regression analysis. In this case, TEQs were calculated using WHO 1998 toxicity equivalent factors[9], since this was done in the earlier studies. Medium bound levels were used in the calculations.

Table 1. Levels of PCDD/Fs and PCBs in food homogenates of selected market basket food groups. Data are presented as grocery store specific (sample 1-5) and as means of all five samples. Medium bound levels are presented (<LOQ=½LOQ) and given on fresh weight basis.

Food category	Sample[a]	Fat %	pg TEQ_{2005}/g			pg/g	
			PCDD/F[b]	PCB[c]	PCDD/F/PCB[d]	Indicator-PCB[e]	PCB 153
Eggs	1	8.7	0.029	0.0083	0.037	39	6
	2	8.7	0.031	0.022	0.053	188	83.0
	3	7.9	0.051	0.27	0.32	33	5.5
	4	8.1	0.027	0.24	0.26	70	24.6
	5	8.9	0.033	0.27	0.30	31	6
	mean	*8.4*	*0.034*	*0.16*	*0.19*	*72*	*25*
Fish	1	12	0.11	0.18	0.29	2130	804
	2	8.3	0.11	0.16	0.27	1860	672
	3	11	0.095	0.17	0.27	2040	734
	4	12	0.12	0.19	0.31	2300	803
	5	11	0.13	0.21	0.34	2210	808
	mean	*11*	*0.11*	*0.18*	*0.30*	*2108*	*764*
Meat	1	8.4	0.0084	0.011	0.02	73	30
	2	11	0.011	0.015	0.026	89	34
	3	13	0.010	0.013	0.024	79	33
	4	11	0.0085	0.021	0.03	155	64
	5	11	0.009	0.013	0.022	80	34
	mean	*11*	*0.0094*	*0.015*	*0.024*	*95*	*39*
Dairy products, liquids	1	1.8	0.0019	0.0037	0.0056	17	7.1
	2	1.6	0.0024	0.0041	0.0065	17	7.0
	3	1.6	0.0016	0.0036	0.0051	16	6.3
	4	1.7	0.0023	0.0034	0.0057	16	6.4
	5	1.5	0.0020	0.0031	0.0051	16	6.4
	mean	*1.6*	*0.0020*	*0.0036*	*0.0056*	*16*	*6.6*
Dairy products, solids	1	25	0.027	0.026	0.054	219	90
	2	25	0.035	0.048	0.083	208	84
	3	23	0.028	0.054	0.082	215	87
	4	25	0.026	0.056	0.082	251	106
	5	26	0.024	0.042	0.066	174	71
	mean	*25*	*0.028*	*0.045*	*0.073*	*213*	*88*
Fats, oils	1	62	0.036	0.031	0.066	202	63
	2	65	0.038	0.038	0.076	230	71
	3	63	0.034	0.034	0.068	201	67
	4	65	0.084	0.053	0.14	276	91
	5	62	0.046	0.046	0.092	286	96
	mean	*63*	*0.048*	*0.040*	*0.088*	*239*	*78*

[a]Sample 1-5 represents homogenates prepared with food items purchased from five different grocery chains. [b]Sum TEQ of 17 PCDD/F congeners. [c]Sum TEQ of 12 dioxin-like PCB congeners (PCB 77, 81, 105, 114, 118, 123, 126, 156, 157, 167, 169, 189) [d]Sum TEQ of 17 PCDD/F and 12 dioxin-like PCB congeners. [e]Sum of 6 non-dioxin-like PCB congeners, i.e. indicator-PCBs (PCB 28, 52, 101, 138, 153, 180).

Results and discussion

Results from the analysis of PCDD/Fs and PCBs are presented in Table 1 and calculated average per capita intakes of PCDD/F/PCB TEQ and PCB 153 are presented in Table 2. Total per capita intake of both PCDD/F/PCB TEQ and PCB 153 varied less than 2-fold between grocery chain baskets, showing a homogenous contamination pattern on the Swedish food market. For PCB 153, most of the individual food group baskets had

concentrations higher than LOQ, giving almost identical lower- and upper-bound total per capita intakes. In the case of PCDD/Fs and dioxin-like PCBs, there were some congeners with a large proportion of concentrations below LOQ. As a consequence the lower- and upper-bound total per capita intakes differed more than 30%.

The highest levels of PCDD/Fs and PCBs were found in fish (Table 1) and fish was also the food category that contributed most to the total intake of PCDD/F/PCB TEQ and PCB 153 (Figure 1). The most obvious differences in food category contribution were observed for egg and fish. Fish contributed to 62% of the mean total per capita intake of PCB 153, and only to 38% of PCDD/F/PCB TEQ. Eggs gave a small contribution (1%) to PCB 153 intake but contributed to 14% of the PCDD/F/PCB TEQ intake. This suggests differences in the contamination patterns between the two substance groups. There were large variations in levels of PCDD/F/PCB TEQ and PCB 153 between egg samples (Table 1), resulting in high variations in contribution from egg to the total intake. The contribution differed 15-fold for PCB 153 (0.29-4.4%) and 6-fold for PCDD/F/PCB TEQ (3.7-24%). The large variation in contamination level between egg samples may be due to inclusion of eggs from producing facilities with high levels of PCDD/F/PCB in hen's feed or in the environment of the hens. Also in the 2010 market basket study, a large variation in the contamination of egg samples was observed.[6]

Table 2. Calculated average per capita intakes of PCDD/F/PCB TEQ (pg/day) and PCB 153 (ng/day) based on levels of contaminants in a Swedish market basket study from 2015.

Compound	**Intake (lower-bound)** median (range)	**Intake (medium-bound)** median (range)	**Intake (upper-bound)** median (range)
PCDD/F/PCB TEQ[a]	31 (21-38)	38 (28-43)	43 (34-47)
PCB 153	55 (52-66)	56 (52-66)	56 (52-66)

[a]Sum TEQ of 17 PCDD/F and 12 dioxin-like PCB congeners.

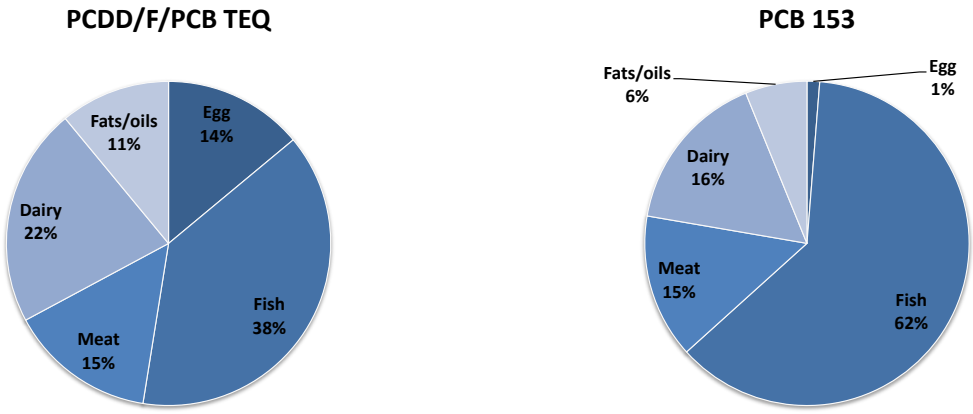

Figure 1. Relative contributions to the intake of PCDD/F/PCB TEQ and PCB153 from different food categories.

Declining trends of total per capita intake of PCDD/F/PCB TEQ_{1998} and PCB 153 were observed between 1999 and 2015, with a mean decrease of 4.5% per year for both substances (Figure 2). Declining temporal trends of non-dioxin-like PCBs and PCDD/F/PCB TEQ in mother's milk from nursing women in Sweden since the early 1970s also show that human exposure in Sweden has decreased for many decades after risk management efforts to minimize environmental pollution were introduced.[10,11]

Although exposure estimates from MB studies only refer to population means they give an overall picture of exposure levels in a cost-effective way, and can be used for preliminary risk assessments. The established tolerable daily intake (TDI) of PCDD/F and dioxin-like PCBs (2 pg TEQ/kg body weight/day)[12] and the reference dose (Rfd) published by the US EPA (0.7 pg TEQ/kg body weight/day)[13] are both relevant for girls and women of a child-bearing age that accumulate these contaminants before pregnancy. Assuming an average

body weight of 60 kg for young women, the total per capita medium-bound intake of PCDD/F/PCB TEQ based on the market basket study 2015 corresponds to 0.47-0.72 pg TEQ/kg body weight/day. This is 3-4 times lower than the TDI, but just below or at the same level as the Rfd. For younger children and adolescents with lower body weights the Rfd is exceeded. Consequently, the presence of dioxin-like compunds in food still constitutes a potential health problem, and efforts to reduce exposure should continue.

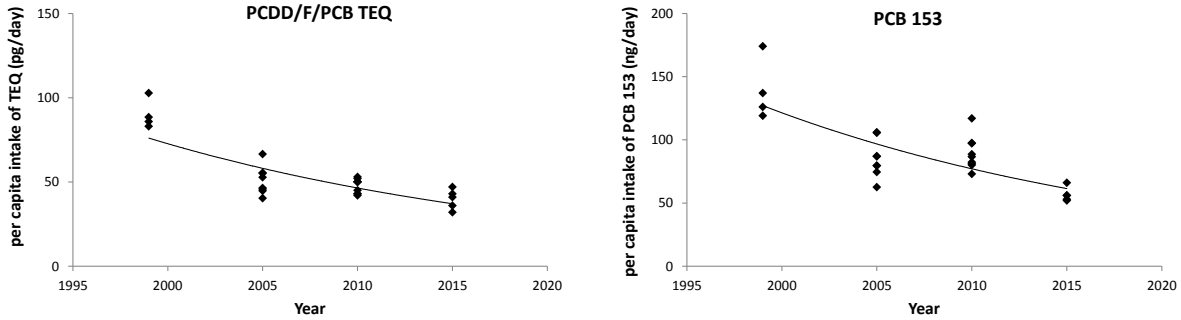

Figure 2. Per capita intake of PCDD/F/PCB TEQ_{1998} and PCB 153 (medium-bound) 1999-2015. Dots represent per capita intake estimated from levels in market baskets from four different cities (1999 and 2005, Malmoe, Gothenburg, Uppsala, Sundsvall) and from different grocery chains in Uppsala (2010 and 2015). The log-linear regression lines show that intake of PCDD/F/PCB TEQ decreased 4.5% per year (mean; standard error 0.64%, $p<0.001$) and intake of PCB 153 decreased 4.5% per year (mean; standard error 0.74%, $p<0.001$). N=26.

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