Trends in Dietary Exposure to Dioxins and PCBs in The Netherlands. Results from a Duplicate Diet Study.

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Abstract

Levels of PCDDs, PCDFs and PCBs have been determined in pooled samples of lyophilized duplicate portions of 24-hour diets collected in the years 1984/85 and 1994 to study temporal trends in the average dietary intake of a Dutch population aged 18 years and older. The study revealed a significant decline in the mean dietary intake of PCDDs, PCDFs and PCBs in the period between 1984/85 and 1994. From chemical analyses of four composites of archived duplicate diet samples from 1978, the mean dietary intake of the PCDDs, PCDFs and the higher chlorinated PCBs appeared to be much higher than in 1984/85. Although there are certain factors that need further verification, the latter observation may suggest that the onset of the decline in the dietary exposure to dioxins and PCBs already occurred at the end of the 1970s.

Introduction

In 1993, we reported on the intake of 2,3,7,8-chlorine substituted dioxins, furans and non-*ortho* PCBs from food in The Netherlands [1]. The study included measurements in national composite samples of different types of food groups. By combining the concentrations of the compounds of interest with the original data from a Dutch National Food Consumption Survey [2], a large database was obtained containing dietary intake data of 5898 individuals, aged 1 to 85 years. A statistical analysis of this database revealed a median daily intake for persons of 20 years and over of 1 pg (i)-TEQ per kg body weight, with a 95 percentile of 2 pg (i)-TEQ/kg.day. For the dietary intake of non-*ortho* PCBs, a comparable median and 95 percentile were found on TEQ basis. In addition, higher intakes were calculated for younger children from 1 to 6 years of age. Fats from cow's milk, butter, cheese and beef were found to be responsible for 50% of the exposure from food [1].

In the study, only the non-*ortho* substituted PCB congeners were included in the measurement programme. Hence, information on the dietary exposure to other dioxin-like PCBs were lacking. Furthermore, there were no data available to assess trends in the dietary exposure in the past 10-15 years. To investigate these trends, we determined the concentrations of the seventeen toxic PCDDs and

PCDFs, and a selection of 29 PCB congeners in lyophilized samples of duplicate 24-hour diets, collected in the years 1994, 1984/85 and 1978.

Materials and methods

Study design. The main objective of this study was to investigate trends in the mean dietary exposure to dioxins and PCBs. Relevant information on the contribution of different types of food groups to the total TEQ intake, and the distribution of dietary intakes in relation to age was already available [1]. For this reason it was chosen to use the 'duplicate diet' approach to estimate the temporal trend in the average total daily intakes (per kg body weight) for groups of consumers reflecting the Dutch adult population.

Study population. For this study, we had access to lyophilized duplicate portions of 24-hour diets (i.e. homogenized duplicate of all foods consumed, including drinks and drinking water, during a 24-hour period) collected by adult volunteers in the periods January-March 1978, October 1984 and March 1985, and March and October 1994. A comparable sampling strategy was applied in 1984/85 and 1994. In these studies, volunteers were recruited among the adult population living in an area of 30 km around Bilthoven. The sampling strategy of the 1978 survey was slightly different from the later ones. In this survey, volunteers had been recruited at the former National Institute of Public Health in Bilthoven and the resulting group differed from those of the 1984/85 and 1994 survey with respect to the male/female ratio (relatively more males) and the distribution of age. Some characteristics of the surveyed populations are summarized in table 1.

Sample preparation. Chemical analysis was performed on pooled samples. To this end, aliquots of the 93 individual samples collected in 1978 were aggregated into four pooled samples, each composed of 23-25 samples. In this regard, the size of the aliquots to be aggregated into the same pool was standardized to the total amount of ingested food per kg body weight for the respective volunteer. The proportional pooling technique was also used in the preparation of the pooled samples for the 1984/85 and 1994 survey. For each of the latter two surveys, ten pooled samples were composed out of 10-12 samples each.

Chemical analysis. Aliquots of 50 g of each pooled sample were fortified with a mixture of $^{13}C_{12}$-labeled reference standards of PCDDs, PCDFs and non-*ortho* PCBs 77, 126 and 169, and subsequently refluxed using 250 ml of dichloromethane for 16h. After centrifugation, the extract was evaporated to dryness to constant weight. The residue, representing the fat content, was redissolved in 100 ml of hexane. An aliquot of this mixture, corresponding to 125-150 mg of extracted fat and mixed with a solution containing $^{13}C_{12}$-labeled reference standards of PCBs 28, 52, 101, 118, 138, 153, 156, 180 and 189, was subsequently used for the determination of mono-, di- and other (non-planar) PCBs with an analytical procedure based on the use of SFE and GC/HRMS as described by Van der Velde *et al.* [3].The remainder was used for the determination of PCDDs, PCDFs and non-*ortho* PCBs 77, 126 and 169 according to the Carbosphere method as extensively described elsewhere [4,5].

Results and Discussion

Table 1 shows the dietary intakes of the PCDDs, PCDFs and dioxin-like PCBs expressed in TEQs using the TEFs according to NATO/CCMS [6] and WHO [7]. Figure 1 shows the relative change in the dietary intake (mean ± SD of mean) of selected congeners calculated for the years 1978, 1984/85 (set at 100%) and 1994.

The mean intake of PCDDs and PCDFs decreased from 1.8 pg (i)-TEQ/kg.day in 1984/85 to 0.48 pg (i)-TEQ/kg.day in 1994. If the contribution of dioxin-like PCBs is included as well, the intake was on average 4.2 pg ΣTEQ/kg.day in 1984/85 and 1.4 pg ΣTEQ/kg.day in 1994. In the period between 1984 to 1994, the mean intake of almost all congeners decreased by a factor of 2 to 4. Results from the period 1978 are more difficult to compare, because there were slight differences between the surveyed populations and in the sample preparation at the time of collection. Despite these differences, a pronounced difference can be noted between the mean dietary intakes established for 1978 and 1984/85 for most of the PCDDs, PCDFs and the higher chlorinated PCBs. These data suggest that the onset of the decline in the dietary exposure to dioxins and these PCBs already occurred at the end of the 1970s. For the less chlorinated PCBs (up to pentachlorobiphenyl), the intakes seem to be comparable with those found for 1984/85.

Table 1
Characteristcs of surveyed populations and estimated dietary intakes.

Surveyed population		**1978**	**1984/85**	**1994**
Characteristics (mean)				
No. of indiv.samples		93	109	121
Fraction male/female		0.76	0.52	0.50
Body weight (kg)		71.6	69.9	74.8
Age (yr)		37.6	42.6	44.5
Food ingested (g/d)		2386	2235	2609
Fat intake (g/d)		0.94	0.90	0.94
No. of pools analysed		4	10	10
No. of samples in pools		23-24	10-11	12-13
Dietary intake (mean±SD of mean)				
PCDDs and PCDFs	pg TEQ/kg.d	4.2±0.2	1.8±0.3	0.53±0.07
Non-ortho PCBs *	pg TEQ/kg.d	2.3±0.2	1.6±0.1	0.66±0.05
Mono-ortho PCBs *	pg TEQ/kg.d	2.5±0.3	0.9±0.1	0.21±0.02
Di-ortho PCBs *	pg TEQ/kg.d	2.0±0.2	0.14±0.01	0.045±0.003
Σ TEQ	pg TEQ/kg.d	11±1	4.2±0.3	1.5±0.1
PCB 138	ng/kg.d	19±2	5.1±0.4	1.4±0.1
PCB 153	ng/kg.d	18±1	6.2±0.5	1.8±0.2
PCB 180	ng/kg.d	26±3	2.6±0.1	0.75±0.05
Σ 29 PCBs *	ng/kg.d	149±14	73±8	20±1

* Selected PCB congeners: non-*ortho* PCBs 126 and 169, mono-*ortho* PCBs 105, 114, 118, 123, 156, 157, 167 and 189, di-*ortho* PCBs 170 and 180, and 'other' PCBs 18, 28, 47, 49, 52, 60, 66, 74, 99, 101, 128, 138, 153, 163, 183, 187, and 194.

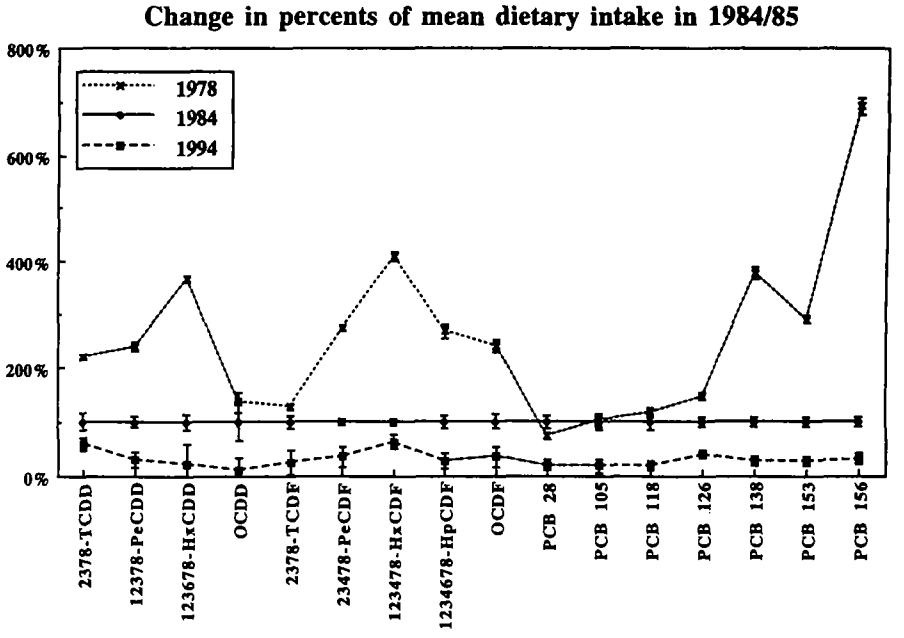

Figure 1

Mean dietary intakes of selected PCDD. PCDF and PCB congeners by an adult Dutch population in 1978 and 1994 relative to the mean dietary intakes estimated for 1984/85. Error bars denote the standard deviation of the mean.

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