

INTAKE OF 2,3,7,8 CHLORINE SUBSTITUTED DIOXINS, FURANS, AND PLANAR PCBs FROM FOOD IN THE NETHERLANDS: MEDIAN AND DISTRIBUTION

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ABSTRACT

To establish a standard for dioxin and related compounds in cow's milk fat, a study was carried out to determine the intake of the seventeen 2,3,7,8-chlorine substituted dioxins and furans, and three planar PCBs from food in the Netherlands. In this study were 26 different food samples, or mixtures of food products analyzed. Sampling strategy was set up using information of the Dutch food consumption survey from 1987 to 1988. By combining the concentrations of the compounds of interest with the original data from the food consumption survey, a large database was acquired, that contained intake figures of 12,000 individuals for various food categories. With these data, a statistical model was developed, that was used to calculate the median and distribution of the daily intake, in relation to age and body weight. Besides the contribution of the various food categories to the total median intake was determined. The model was used for a scenario analysis, to calculate the percentage of the population that exceeds the TDI of dioxin and related compounds after lifetime exposure from food, with increasing concentrations of TEQ in cow's milk fat and beef.

The results showed a median daily intake for adults of 1 pg (i)-TEQ per kg body weight, with a 95 percentile of 2 pg (i)-TEQ/kg, lognormal distributed. This median is lower than the results reported by authors from other countries. For the planar PCBs the figures were 1.4 and 2.5 pg (PCB)-TEQ respectively. Younger children showed a higher intake, due to their lower body weight. Fat from cow's milk, butter and cheese, and associated beef was responsible for 50% of the exposure from food. Second important were food items with a mixture of vegetable and animal fats and oil, as added by the food industry. Fish oil was responsible for that contribution, but it showed a large variation in concentrations of 2,3,7,8-chlorine substituted dioxins and furans, and three planar PCBs, in comparison with the other food categories. Finally, the model predicts an exceeding of the TDI of 10 pg TEQ/kg.day for less than 1 percent of the population at the Dutch standard of 6 pg TEQ/g milkfat.

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KEY WORDS

PCDD; PCDF; Planar PCB; Standard for cow's milk; Exposure

INTRODUCTION

In July 1989 elevated levels of dioxins and furans in cow's milk and cheese were detected in the vicinity of a municipal solid waste incinerator (MSWI) in the Netherlands (Olie, pers.comm. 1989). For a health risk assessment of these concentrations, an estimation of the average total exposure to dioxins and furans for the Dutch population was made. The results estimated that an exceeding of the Dutch tolerable daily intake (TDI), which was 4 pg TEQ/kg body weight.day at that time, was likely at concentrations in cow's milk above 6 pg TEQ/g milkfat (Liem *et al.*, 1989). Accordingly, a Dutch (temporary) standard of 6 pg TEQ/g milkfat was set by law in July, 1989.

In 1990 there was a call for international harmonization of the TDI for dioxin and related compounds, as it varied from 1 to 10 pg/kg bw.day for different European countries. An expert meeting was organized by the European office of the World Health Organization (WHO) on behalf of the Dutch Ministry of Welfare, Public Health and Cultural Affairs. It proposed a TDI of 10 pg dioxin or related compounds per kg body weight in December 1990 (WHO/EURO, 1991). The Dutch government accepted the proposal. The new TDI implied a re-evaluation of the temporary standard for TEQ in cow's milk fat.

As a consequence of using average food consumption figures, exceeding of the TDI is possible for 50 percent of the local population that consumes milk and cheese with 6 pg TEQ/g fat. So, there was a need to know the distribution of individual intakes of dioxin and related compounds in the Netherlands. With this one could set a more accurate standard in cow's milk. As 90 to 98 percent of the total daily exposure to chlorinated dioxins and furans by the Dutch population is coming from food (Theelen, 1991), it was decided to analyze the seventeen 2,3,7,8-chlorine substituted dioxins and furans in selected food items. As planar PCBs are considered to be dioxin related compounds, these chemicals were included. The sampling strategy had to focus on the most relevant routes of contamination. By combining the results of the chemical analysis with individual food consumption patterns, both information on the distribution of exposure from food, and on the contribution of the various food items, such as cow's milk fat, could be determined.

Using a scenario with increasing concentrations TEQ in food items with cow's milk fat, one could calculate intake figures in relation to dioxin in cow's milk fat. With these intake figures, in combination with the TDI of 10 pg/kg bw.day, an adjustment of the standard for TEQ in cow's milk can be made.

MATERIAL AND METHODS

Food consumption survey

Data on individual food consumption habits were obtained from the Dutch food consumption survey (VCP, 1988). This survey includes a description of the daily consumption of 5898 persons over two consecutive days, in 1987 or 1988. For each individual in the survey both age, sex, body weight and a series of other characteristics are recorded. The persons ranged from 1 to 70 years in age. Sampling days were equally distributed over time, except for the Dutch holidays. Per person, the quantities of various ingested food items over the day were recorded. A comprehensive description of the food items, including percentage total fat was available (NEVO, 1988).

Sampling strategy

In the consumption survey were 1350 food items recorded. Food samples that were to be analyzed, were selected as representatives for the various different routes of contamination. In the first step, about 800 food items were rejected after consulting a dietary specialist, as these items were not expected to be contaminated with dioxin and related compounds. The remaining items were divided into 26 categories, according to the description of the composition of the food item (Kistemaker, 1989).

For the categories with one type of animal fat, such as beef and pork, fat samples were collected in slaughter houses from different regions in the Netherlands. Samples were taken by regulatory veterinarian inspectors.

Categories such as fish, mixed meat, and cheese were sampled using a proportional technique. These samples were prepared from a mixture of different food items, with each item added in weight proportional to its average consumption, as determined by the food inquiry. All these mixtures held at least a total of 50 g fat. The items themselves were gathered over the Netherlands in different stores by inspectors of the food inspection departments.

A third type of sampling was planned for the food items with fat, available from the food industry. These items, such as margarine and sauces, contain a mixture of vegetable oils and fats. Other items from the food industry do have a mixture of vegetable and animals oils and fats. Examples are pastry, (canned) pre-processed food, sweets and candy bars. The types of oils and fats in these products vary in global origin, depending on the world market prices. They are added to the product after a complex refinery process. In collaboration with the Dutch co-ordinate organization for margarine, fats and oil, the food industry provided purified oils and fats of the types, that are added to the food items. A mixture of these oils and fats, proportional to its annual use was said to represent the categories food items from the food industry.

Chemical analysis

Analysis of the food samples was based on isolation of the fat fraction containing the compounds of interest, purification of the fat extract and isomer-specific analysis of the seventeen 2,3,7,8-chlorine substituted PCDDs and PCDFs and three planar PCBs (e.g. 3,3',4,4'-TeCB, 3,3',4,4',5-PeCB and 3,3',4,4',5,5'-HxCB) by gas chromatography-mass spectrometry. Prior to extraction, laboratory samples were fortified with mixtures of carbon-13 labeled analogues of the congeners to be analyzed (¹³C₁₂-labeled standards, from Cambridge Isotope Laboratories, Woburn, MA, USA). Clean-up methods consisted of consecutive column chromatography separations on active carbon (Carbosphere activated carbon, 80-100 mesh, surface area 1000 m²/g from Chrompack, Middelburg, The Netherlands) and alumina (basic, activity super I, from ICN Biomedicals GmbH, Eschwege, Germany), developed for the analysis of PCDDs and PCDFs in milk samples (Liem *et al.*, 1990), based on the carbon method according to Smith *et al.* (1984). GC-MS analyses were performed on a VG 70SQ or AutoSpec (Fisons Instruments, Manchester, UK) mass spectrometer coupled to a HP 5890 (Hewlett Packard, Palo Alto, MA, USA) gas chromatograph, or on a Finnigan MAT95 (Finnigan, Bremen, Germany) mass spectrometer coupled to a Varian 3400 gas chromatograph. GC separation between PCDD and PCDF isomers was carried out by using an apolar column (Ultra-II of DB-5 type columns), except for analysis of PCDDs and PCDFs in fish oils, where a polar CPSil88 column was used. Ionization of samples was performed in electron impact mode (EI) with 30-70 eV electrons. Instruments were operated at increased resolution. The resolving power (RP) was typically between (static) 3000 and 5000. Limits of determination ranged from 0.1 to 1 pg/g fat for the lower to higher chlorinated congeners, respectively, resulting in limits of determination of approximately 0.3 pg (i)-TEQ/g and of about 0.01 pg (PCB)-TEQ/g of fat, when expressed in toxic equivalents. Based on intralaboratory quality control procedures, an analytical coefficient of variation was determined of 100% on TEQ-basis for vegetable oils and fats and of approximately 5% on TEQ-basis for the other matrices. A full description of the applied analytical chemical methodology is in preparation.

Statistical analysis.

Individual intake data were subjected to statistical evaluation, to determine the median intake and the distribution within the Dutch population, in relation to age and body weight. The statistical model is described elsewhere (Slob, 1993).

RESULTS

Concentration of dioxin and related compounds in various food samples. The concentrations of the individual congeners in the food items are extensively reported elsewhere (Liem *et al.*, 1991). In this paper, the

concentrations are expressed in TEQ, using (i)-TEFs (Van Zorge *et al.*, 1988), and (PCB)-TEFs from the Dutch TEF working group, assigning a TEF of 0.01 for PCB #77, 0.1 for PCB #126, and 0.005 for PCB #169.

The concentration of dioxin and related compounds for the category food products from the food industry was calculated in proportion to the quantity of the various types of oil and fat added to the food items in the Netherlands in 1988 (MVO, 1989). The results are summarized in table 1.

table 1. Concentrations dioxin and furans, and planar PCBs in food categories (average value from duplicates)

food category		PCDD/PCDF [pg TEQ/g fat]	planar PCB [pg TEQ/g fat]	
beef		1.75	2.45	
cow's liver		5.7	3.95	
pork		0.43	0.16	
pig's liver		15.3	2.05	
poultry		1.65	2.0	
chicken's liver		3.25	2.8	
mutton		1.85	2.0	
horse meat		13.85	25.2	
game	M	16.8	18.0	(N=4)
butter		1.8	2.1	
cheese	M	1.4	2.05	
nuts	M	0.2	0.35	
cereals	M	0.34	1.0	(N=3)
eggs		2.0	2.25	
fatty sea fish	M	6.65	12.8	
lean fish	M	48.65	146.75	
eel		28.0		(N=6)
fresh water fish	M	2.4	6.05	
mixed-meat product	M	0.67	0.5	
dairy products		1.58	1.45	(N=3)
soy bean oil		0.025	0.015	(N=4)
rape-seed oil		0.006	0.015	
palm-oil		0.030	0.030	
sunflower-oil		0.006	0.010	(N=4)
coco-nut fat		0.024	0.030	
palm-fat		0.010	0.020	
fish-oil		2.2	4.2	(N=10)
items with vegetable oil	M	0.02	0.02	
items from food industry	M	0.41	0.76	

(M indicates a proportional mixture of food items)

To calculate the concentration of dioxin and related compounds in a food item on a total product basis, the percentage total fat in the food item, as recorded in the NEVO-list (NEVO, 1988), was multiplied with the concentration TEQ per g fat in the sample from the category, representing the food item.

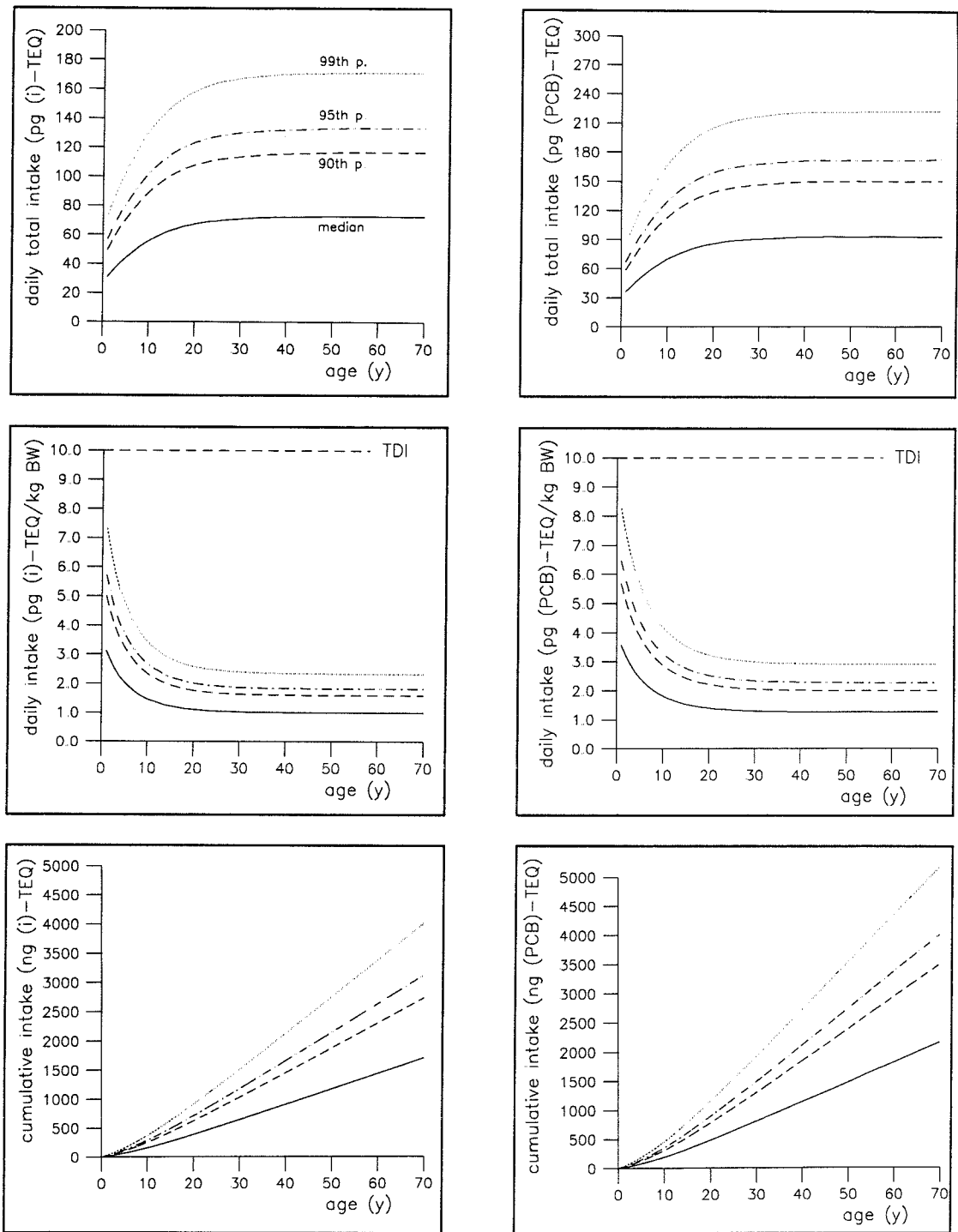


Fig 1. Daily intake of the seventeen 2,3,7,8-chlorine substituted dioxins and furans (figures on the left), and planar PCBs (figures on the right), from food in the Netherlands, in relation to age: total, per kg body weight and cumulative total (as median, 90th, 95th, and 99th percentile)

Calculation of individual human daily intake

Individual intake. For each individual in the food consumption survey the daily intake was calculated by multiplying the amount of consumed product (gram) with the concentration TEQ (per gram) on product basis, and adding up the TEQs of the different food items consumed. The daily intake per kg body weight for each individual was obtained using the body weight of the individual, as reported in the database.

Distribution of intake

With the statistical model the median daily intake and the distribution of (i)-TEQ from the seventeen 2,3,7,8-chlorine substituted dioxins and furans in relation to age was calculated. The daily intake per kg bodyweight in relation to age was calculated the same model, as was the distribution of intake of TEQ from the three planar PCBs. Using the results from the statistical model, the cumulative lifetime intakes of TEQ from dioxins and furans, and planar PCBs were computed. The results are shown in figure 1.

Contribution of the categories to the total intake

To establish the contribution of a food category to the total daily intake from food, the statistical analysis was performed with the individual intake data, excluding the category addressed. The contribution of that food category was then calculated as the difference in median intakes for all categories compared to all minus that category. Figure 2 shows the contribution of the different categories to the lifetime intake of TEQ from both dioxins, furans, and planar PCBs.

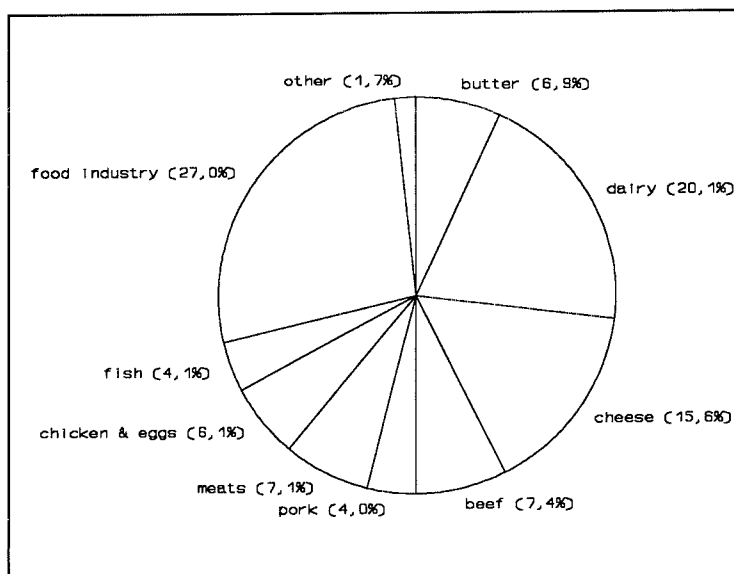


Fig 2. Contribution of categories food products to lifetime exposure to (i)-TEQ and (PCB)-TEQ in the Netherlands

Scenario analysis of TEQ in cow's milk fat

To evaluate the consequences of elevated levels of dioxins and furans in cow's milk fat, a scenario analysis at several TEQ levels in cow's milk was performed. As shown by Olling *et al.* (1990), concentration in beef fat and cow's milk fat for the various congeners are similar. By varying concentrations TEQ in beef fat, cow's milk, cheese and butter simultaneously, a series of intake models was obtained. A maximal tolerable lifetime human exposure of 10 pg TEQ per kg per day (TDI) is equivalent with 255.5 ng TEQ per kg bodyweight over 70 years. The percentage of the Dutch population, that would exceed that maximal tolerable exposure was calculated with the statistical model, for TEQ in cow's milk products and beef in a range from 0 to 14 pg TEQ per gram fat. The result is presented in figure 3.

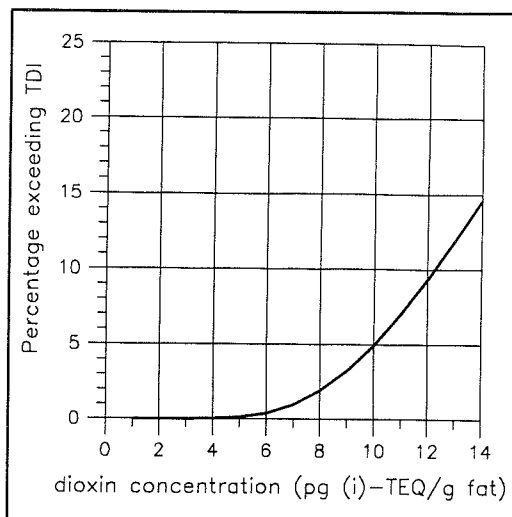


Fig 3. Percentage of the Dutch population, exceeding the TDI of 10 pg TEQ/kg.day after lifetime exposure to (i)-TEQ and (PCB)-TEQ, in relation to the TEQ in cow's milk fat and beef

DISCUSSION

Travis and Hattemer-Frey (1987) estimated a daily intake of 50 pg 2,3,7,8-TCDD for the general population of the USA, using a Mackay model. A Swedish study of food basket samples reported a daily intake of 14 to 55 pg (Nordic)-TEQ (National Environmental Protection Board, 1988). In a Danish study an intake of 2 pg (Nordic)-TEQ/kg.week was calculated (Büchert, 1988). Fish was reported to be responsible for half of total exposure. Birmingham *et al.* (1989^a) calculated a daily intake of 92 pg (i)-TEQ from food for Canadian adults, while in an other study from Birmingham *et al.* (1989^b) a daily intake of 140 pg (i)-TEQ was reported. In both studies dairy products and beef were the major contributors. Beck *et al.* (1989) estimated the German exposure from food products at 93.5 pg (FHO)-TEQ, and Fürst *et al.* (1990) estimated the exposure from food in Germany at 85 pg (FHO)-TEQ, one third from milk, one third from fish and one third from meat. Expressed in (i)-TEFs these German results are corresponding to approximately 160 pg (i)-TEQ. In Italy average daily intake from food was reported to be about 260 to 480 pg (i)-TEQ (Di Domenico, 1990). In that study a significant exposure from vegetables was calculated. In Norway, a provisional intake of 625 pg (Nordic)-TEQ/week was calculated (Faeden, 1991), primarily from milk products. In a previous Dutch study (Theelen, 1991), the estimated daily intake was 115 pg (i)-TEQ from food. In the UK a daily intake of 125 pg (i)-TEQ was reported, mainly from milk, meat, oil, and fat (Ministry of Agriculture, Fisheries and Food, 1992). In all

these studies, intake was calculated by multiplying average consumption figures with the concentrations of the seventeen 2,3,7,8-chlorine substituted dioxins and furans in samples of the food products.

This present Dutch study shows a considerable lower median intake of (i)-TEQs than the other countries, except for the Swedish food basket survey. As in this present study the sampling strategy was derived from the Dutch food consumption survey, this study includes more food items, relevant for actual consumption than other studies. As concentrations (i)-TEQ in mother's milk and adipose for West European countries are comparable (Jensen, 1989), intake of (i)-TEQ must be similar for the different countries. So, using average consumption figures overestimates median intake from food.

The intake data show a lognormal distribution for all ages. The daily intake of the three planar PCBs is somewhat higher than the seventeen dioxins and furans, expressed as TEQ. When summing up (i)-TEQ and (PCB)-TEQ, a few percent of the children from 1 to 6 years of age did have an intake per kg body weight, higher than the TDI of 10 pg TEQ/kg.day from food, due to their lower body weight. It must be emphasized, that exposure of breast fed babies from mother's milk was excluded in this study. As the actual concentration TEQ from dioxins, furans, and planar PCBs in milkfat is about 3 pg/g, it can be seen in figure 3, that for the general population after lifetime exposure the TDI is not exceeded. So, the higher exposure from food for the younger children is compensated later on.

Milk and milk products are the major route for human dioxin exposure in the Netherlands. That conclusion was also drawn by most other studies, previously mentioned. Consequently, contamination of cow's milk will have a substantial impact on exposure to dioxin and related compounds. Therefore, setting a standard for dioxin and related compounds in cow's milk fat, will be an effective means to control human exposure.

A second important route are the food items from food industry. The fat is a mixture of various types of oil and fats. The concentration of the seventeen dioxins and furans, and planar PCBs in this mixture was calculated in a way, that matches the proportional contribution of all different types of fat. Although usage of fish oil was only about 18% of all oil and fat used by the food industry in 1988 in the Netherlands (MVO, 1989), it was noticed that fish oil determined about 90 to 95% of the quantity of dioxins and furans, and PCBs in the industrial mixture of fats. The concentrations in fish oil from different global origin ranged from 0.24 to 4.4 pg (i)-TEQ, and from 0.42 to 9.7 pg (PCB)-TEQ per gram oil (Liem *et al.*, 1991). It was reported by Rappe *et al.* (1989), that unprocessed fish oils show substantially higher concentrations, and refinery by the food industry will decrease these levels a tenfold. The fish oils in our study were obtained from the food industry as purified product. It can be concluded, that fish oils in items from food industry show large variations in its contribution to exposure to dioxin and related compounds from food. The variation will be due to the global origin of the fish oils, and methods of refinery and purification by the food industry.

In summary, it can be concluded, that daily exposure for the general population to dioxin and related compounds from food in the industrial countries is in the order of 1 pg (i)-TEQ/kg. body weight, and that planar PCBs do increase that exposure in the same order of magnitude. Intake of animal fat is responsible for that exposure. In the Netherlands it can be noticed, that local contamination of animal fat with these compounds will occur near sources of emission, and that by control of these emissions an additional exposure can

be prevented. But it is also shown, that "hidden" fats do contribute into a large extend to that exposure. As these "hidden" fats and oils are coming from the global market, their sources of contamination can not be easily controlled. On the other hand, as local emissions are reduced, and "hidden" fats carefully monitored, one can reduce exposure to dioxin and related compounds in a very substantial way.

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