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Indicator PCBs in foodstuffs: occurrence and dietary intake in The Netherlands at the end of the 20th century

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Abstract

The report presents a survey of the most recent (1998/1999) information on the occurrence of indicator-PCBs in foodstuffs in the Netherlands. The data on occurrence collected during measurement programmes on occurrence were combined with food consumption data to assess the dietary intake of the seven indicator-PCBs (polychlorinated biphenyls, congeners 28, 52, 101, 118, 138, 153 and 180). The estimated median lifelong-averaged intake of indicator-PCBs in the population is 5.6 ng per kg bw per day. The 95th percentile of intake in the population is estimated at 11.9 ng per kg bw per day. The contribution of different food groups to the total intake of indicator-PCBs is fairly uniformly distributed over the foods consumed: meat products (27%), dairy products (17%), fish (26%), eggs (5%), vegetable products (7%), and industrial oils and fats (18%). Compared with earlier intake estimations the present estimation shows a considerable reduction in intake of indicator-PCBs, albeit that this reduction flattened out during the last decade. This substantial reduction is related to the decrease in the concentration of PCBs in the majority of foodstuffs. However, a small part of the population still has a rather high intake. If this high intake only occurs for a limited period of time, it is not expected to result in adverse health effects. To provide regulators with a health-based guideline to prevent health effects of exposure to indicator PCBs, the derivation of a TDI, preferably by international bodies, is recommended. Monitoring the dietary intake of PCBs is just as important as monitoring the intake of dioxins, and attempts to decrease the exposure to both compound classes need continuous attention.

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Samenvatting

Inleiding

Dit rapport beschrijft de aanwezigheid van indicator PCB's (polychloorbifenylen; IUPAC congener nummers #28, #52, #101, #118, #138, #153 en #180) in Nederlandse voedingsmiddelen. Deze werden geanalyseerd in verschillende categorieën voedingsmiddelen die werden verzameld in een in 1998/1999 uitgevoerd onderzoek naar de inname van dioxinen (PCDD's en PCDF's) en dioxine-achtige PCB's via de voeding (zie Freijer et al., 2001). De concentraties in voedingsmiddelen die op deze wijze werden gemeten vormden de basis voor innameberekeningen van de indicator-PCB's via de voeding. Deze inname werd berekend aan de hand van gegevens betreffende de consumptie van voedingsmiddelen die verkregen waren in de Voedselconsumptiepeiling van 1997/1998. Evenals de dioxinen en de dioxine-achtige PCB's zijn de indicator-PCB's persistente milieucontaminanten die neigen tot accumulatie in het lichaam, in het bijzonder in lichaamsvet. De grootste belangstelling gaat daarom uit naar de inname op lange termijn.

Methoden

Voor de selectie van voedingsmiddelen werd gebruik gemaakt van de derde Nationale Voedselconsumptiepeiling (VCP), uitgevoerd in 1997/1998. Basis voor de selectie was het relatieve aandeel van voedingsmiddelen in de totale vetconsumptie. In verschillende gebieden van Nederland werden monsters verzameld, waarvan in het laboratorium representatieve mengmonsters werden gemaakt. Deze werden vervolgens geanalyseerd op hun gehalte aan indicator-PCB's, en de resultaten daarvan dienden als startpunt voor de innameberekeningen, wederom gebruikmakend van de consumptiegegevens van de derde VCP. Dit resulteerde in 6250 individuele innamegegevens van indicator-PCB's. Door middel van statistische analyse kon de innameverdeling voor de Nederlandse bevolking worden geschat.

Om tenslotte de inname op lange termijn te berekenen werd een twee-staps procedure toegepast. Eerst werden aan de hand van de VCP de 6250 persoonlijke dag-gemiddelde innamegegevens berekend voor twee opeenvolgende dagen, hetgeen resulteerde in 12500 datapunten. Daarna werd de relatie van de lange-termijn inname met de leeftijd vastgesteld gebruikmakend van regressie-analyse en geneste variantie-analyse. Via de regressie-analyse werd de inname gekwantificeerd als functie van de leeftijd (70 jaar), zodat de levenslange inname kon worden berekend. Met behulp van de variantie-analyse kon onderscheid worden gemaakt tussen de inter- en de intra-individuele componenten van de totale variatie in de daggemiddelde inname van de populatie.

Resultaten

De gemiddelde concentratie van de som van de zeven indicator-PCB's in dierlijke vetten varieert van 4 tot 32 ng per g vet. Deze concentraties zijn een factor 10^4 tot 10^5 hoger dan de concentraties van de som van de dioxinen en de dioxine-achtige PCB's. Op basis van producten zijn de gehalten indicator-PCB's in vis hoger (1 tot 32 $\mu\text{g}/\text{kg}$) dan in vlees (0,2 tot 3 $\mu\text{g}/\text{kg}$). De mediaan van de inname zoals gemodelleerd met behulp van regressie-analyse, varieert van 12,1 ng per kg lichaamsgewicht (lg) per dag voor kinderen van 2 jaar

tot 4,8 ng/kg lg/dag voor volwassenen van 40 jaar. De mediane levenslange inname van de gehele populatie bedraagt 5,6 ng/kg lg/dag, het 95^{ste} percentiel is 11,9 ng/kg lg/dag. De bijdrage van de verschillende groepen voedingsmiddelen aan de gemiddelde PCB-inname is tamelijk evenwichtig verdeeld: vlees en vleesproducten (27%), zuivelproducten (17%), vis en visproducten (26%), eieren (5%), groenten en fruit (7%), en industriële oliën en vetten (18%). Vijfenzeventig procent van de totale inname komt dus voor rekening van dierlijke producten.

Discussie

Om een beeld te krijgen van het verloop in de tijd van de PCB-inname werd de huidige inname vergeleken met de innamen zoals berekend uit eerdere 24-uurs duplicaat voedingsstudies uitgevoerd door het RIVM. Daaruit blijkt dat de gemiddelde inname gedurende de afgelopen decennia enorm is gedaald: van 83 ng/kg lg/dag in 1978 tot 39 ng/kg lg/dag in 1984/1985 en 10 ng/kg lg/dag in 1994.

De toxiciteit van de PCB's komt vooral tot expressie in het centraal zenuwstelsel, de schildklier en het endocriene systeem, maar pas bij incidentele zeer hoge blootstelling dan wel na bioaccumulatie in het lichaam als gevolg van langdurige inname. Hoewel een klein deel van de bevolking een relatief hoge inname van PCB's heeft, wordt daarvan geen schadelijke klinische gevolgen voor de gezondheid verwacht, mits deze hoge inname zich beperkt tot een relatief kortdurende periode.

Conclusies

In de afgelopen decennia is de inname van indicator-PCB's via de voeding aanzienlijk gedaald. Niettemin heeft een klein deel van de bevolking nog altijd een relatief hoge inname, maar indien dit tot een korte periode beperkt blijft worden daarvan geen directe schadelijke gevolgen voor de gezondheid verwacht.

De bijdrage van de verschillende voedingmiddelen aan de inname van indicator-PCB's is tamelijk evenwichtig. Voor zowel de PCB's als de dioxinen vormen dierlijke producten (inclusief zuivelproducten en vis) en oliën en vetten bijna 90% van de inname van de bevolking.

Om de autoriteiten de beschikking te geven over richtlijnen betreffende de inname van indicator-PCB's gebaseerd op gezondheidscriteria, wordt de afleiding van een TDI voor de indicator-PCB's aanbevolen. Dit dient bij voorkeur door internationale organisaties te worden uitgevoerd.

Gezien de toxiciteit van PCB's en het gegeven dat ze veelal samen met dioxinen in dezelfde voedingsmiddelen aanwezig zijn, is het belang van de inname van dioxinen via de voeding niet los te zien van het belang van de inname van PCB's. Maatregelen om de blootstelling aan beide categorieën stoffen verder terug te dringen verdienen voortdurende aandacht.

Summary

Introduction

The occurrence of indicator PCBs (polychlorinated biphenyls; IUPAC congener numbers #28, #52, #101, #118, #138, #153 and #180) in foodstuffs in The Netherlands was investigated. These compounds were measured in composite consumer food categories, which were sampled in a survey carried out in 1998/1999 for the study on dietary intake of dioxins (PCDDs and PCDFs) and dioxin-like PCBs (non-ortho PCBs and mono-ortho PCBs) [see Freijer et al., 2001]. The concentrations in foodstuffs obtained in this way formed the basis for the assessment of the dietary intake of indicator PCBs in the general population. The dietary intake was estimated using the food consumption levels in the population as obtained in the 1997/1998 food consumption survey. Just as dioxins and dioxin-like PCBs, indicator PCBs are persistent contaminants that tend to accumulate in the body, particularly in body fat. Hence, the main interest is in the long-term intake.

Methods

The database of food consumption from the Dutch National Food Consumption Survey (DNFCS) performed in 1997/1998 was consulted for the selection of foods based on their relative importance in the total fat consumption. Next, samples were collected in different regions in the Netherlands. In the laboratory, national representative test samples were prepared and chemically analysed. The results from these chemical analyses were used as input in the database of the DNFCS. The combination of data on levels in the selected food categories and food consumption records included in the DNFCS database resulted in dietary intakes for 6250 individuals. From a statistical analysis of the dietary intake data, the intake distribution was estimated.

A two-step approach was used to estimate the long-term intake in the population. Firstly, for 6250 individuals the personal daily-averaged intake was calculated for two consecutive days, using the food consumption data and concentrations in consumed products (12500 data points). Next, the relationship of the long-term intake with age in the population was determined using regression analysis and nested variance analysis. The regression analysis was used to quantify the intake as a function of age. From this relationship the lifelong-averaged (70 yrs) intake could be calculated. The nested variance analysis served to unravel the between-subject and the within-subject components of the total variation of daily-averaged intake in the population.

Results

The measured average concentrations of the sum of the seven indicator PCBs in animal fats range from 4 to 32 ng/g fat. These concentrations are a factor 10^4 - 10^5 higher than the concentrations of the sum of dioxins and dioxin-like PCBs. On a product basis, concentrations of the indicator PCBs in fish are higher (1 to 32 µg/kg) than in meat products (0.2-3 µg/kg). The median intake, modelled by the regression analysis, ranges from 12.1 ng/day/kg bw for two-year old children to 4.8 ng/day/kg bw for adults at the age of 40.

The regression analysis was also used to calculate the distribution of the intake variation of the indicator PCBs. The median lifelong-averaged intake is estimated to be 5.6 ng/kg bw/day, the 95th percentile in the population is 11.9 ng/kg bw/day.

The contribution of different food groups to the average intake is rather evenly distributed over these groups: meat products (27%), dairy products (17%), fish (26%), eggs (5%), vegetable products (7%), and industrial oils and fats (18%). Thus, 75% of the intake originates from animal products.

Discussion

To obtain a time trend, the average intake of indicator PCBs was compared to the average intakes as measured in earlier 24-h duplicate diet studies performed by our institute. These studies showed that the intake decreased enormously over the last decades: from 83 ng/kg bw/day in 1978 to 39 ng/kg bw/day in 1984/85 and to 10 ng/kg bw/day in 1994. PCBs express their toxicity on the central nervous system, the thyroid and the endocrine system, but only after a very high incidental intake or after bioaccumulation in the body upon long term intake. Although a small fraction of the population is exposed to relatively high intake levels, a relatively short period of such a high intake is not expected to result in any clinical adverse health effects.

Conclusions

In the past decades, the dietary intake of indicator PCBs in the population has decreased considerably. However, a small part of the population still has a rather high intake, but for a limited period of time this is not expected to have immediate adverse health effects.

The contribution of different food groups to the average intake is rather evenly distributed over these groups. For both the PCBs and the dioxins animal products (including dairy products and fish), and oils and fats constitute almost 90% of the intake of the general population.

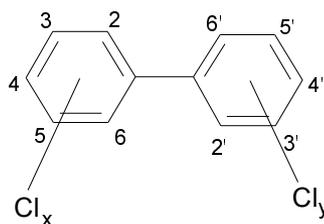
To provide regulators with a health-based guideline to prevent health effects of exposure to indicator PCBs, the derivation of a TDI, preferably by international bodies, is recommended.

In view of the toxicity of PCBs and taking into account that they are generally present in foodstuffs that also contain dioxins, the intake of dioxins is closely associated with the intake of PCBs. Attempts to further decrease the dietary exposure to both compound classes need continuous attention.

1 Introduction

1.1 PCBs

Polychlorobiphenyls (PCBs) have been produced commercially for some five decades starting about 1920, by direct chlorination of biphenyl. This chlorination occurs with one to ten chlorine atoms, resulting in 209 possible PCB congeners. The general formula is shown in Fig. 1.



*Figure 1.1. Polychlorinated biphenyls
($x = 0 - 5$; $y = 1 - 5$)*

PCBs have been produced as mixtures; individual congeners are hardly synthesised. The various (commercial) technical PCB-mixtures are characterised by their chlorine content; the brand names are known as 'Aroclor' (produced in the USA), 'Clophen' (produced in Germany), 'Phenoclor' (produced in France), 'Fenclor' (produced in Italy), and 'Kanechlor' (produced in Japan).

PCB mixtures were used in a wide scale of applications, such as coatings, inks, flame retardants and paints. Its major uses, however, were in electronic appliances, heat transfer systems, and hydraulic fluids. For the different applications many different technical mixtures were being used. The total world production is estimated at 1-2 million tonnes. Due to the persistent nature of PCBs in the environment it was decided by many countries some decades ago to ban the use of PCBs in open applications. PCBs may, however, still be in use in closed systems such as capacitors and transformers. The use in these applications will decrease in time. Waste disposal, both of households and industrial waste, is the major source of PCBs emissions into the environment (ATSDR, 2000; Baars et al., 2001). Since most PCBs congeners are very lipophilic and persistent, PCBs tend to accumulate in soils, sediments and the food chain. As the various congeners differ in their physiochemical properties, they demonstrate different behaviour in the environment. Some congeners are easily being degraded by light, and others by microbial processes. Other congeners are very persistent. Consequently, the composition of the mixtures of PCBs that can be found in the food chain may substantially deviate from the technical mixtures (ATSDR, 2000; Baars et al., 2001).

The chlorination pattern of the PCB is important for the toxicity of the substance. A number of PCB congeners show 'dioxin-like' toxicity. These PCBs have no or only one chlorine atom at the ortho position. The phenyl rings of these molecules can rotate and can adopt a coplanar structure, which leads to the same toxicity as the polychlorinated dibenzo-*p*-dioxins (PCDDs) and the polychlorinated dibenzofurans (PCDFs). These PCBs are assigned with a Toxic Equivalency Factor (TEF) that relates their toxicity to that of TCDD (2,3,7,8-tetrachlorodibenzo-*p*-dioxin; Van den Berg et al., 1998), and are to be evaluated as dioxins. If, however, two of the ortho positions in a PCB molecule are occupied, the two phenyl rings are not in the same plane and the PCB expresses toxicity which is non-dioxin-like. These PCBs act on the central nervous system, the thyroid and the endocrine system, but only after a relatively high incidental intake or after bioaccumulation in the body upon long term intake (ATSDR, 2000).

Mixtures of PCBs are generally assessed on the basis of a chemical analysis of the (sum of the) seven so-called 'indicator PCBs'. The IUPAC numbers of these indicator PCBs are #28, 52, 101, 118, 138, 153, and 180, and are listed in table 1.1. The amount of chlorine atoms of these PCBs ranges from three to seven. PCBs #28 and #118 are mono-ortho PCBs, the others have two chlorine atoms on the ortho-positions.

Table 1.1. Indicator-PCBs

IUPAC number	IUPAC name
28 *	2,4,4'-trichlorobiphenyl
52	2,2',5,5'-tetrachlorobiphenyl
101	2,2',4,5,5'-pentachlorobiphenyl
118 *	2,3',4,4',5-pentachlorobiphenyl
138	2,2',3,4,4',5'-hexachlorobiphenyl
153	2,2',4,4',5,5'-hexachlorobiphenyl
180	2,2',3,4,4',5,5'-heptachlorobiphenyl

* Congeners #28 and #118 are dioxin-like PCBs.

The indicator PCBs are known to be persistent in the environment and to bioaccumulate in the food chain, and are assumed to be a suitable representative for all PCBs. Since these are the predominant congeners in biotic and abiotic matrices, this group was chosen for the present dietary intake assessment.

1.2 Dietary intake of PCBs

PCBs and other halogenated organic compounds, such as dioxins and furans are ubiquitously present in the environment, and because they are persistent and accumulate in the food chain also in the human diet. The need to conduct a survey on the levels in food products was strongly increased when elevated dioxin levels were reported for cow's milk collected near a municipal waste incinerator in the 1980s. The dietary intake of PCBs has been studied in 24-h duplicate diet studies in 1978, 1984 and 1994. The intake of the sum of the indicator PCBs decreased in this time period from 83 ng/kg bw/day in 1978 to 39 ng/kg bw/day in 1984/85, and to 10 ng/kg bw/day in 1994 (Liem and Theelen, 1997). Another method to estimate the dietary intake is to combine food analyses with consumption data. This was done for the first time in 1990-1991, for PCDD/Fs (Liem et al., 1991). This procedure was repeated in 2001, using the third Dutch National Food Consumption Survey of 1997/1998, and analytical data on PCDD/Fs and dioxin-like PCBs in foodstuffs sampled in 1999 by a joint monitoring programme of RIVM and RIKILT (Freijer et al., 2001). The dioxin study of 2001 was the basis for the present study into the dietary intake of indicator PCBs.

1.3 Objectives

The current study investigates the dietary intake of the seven indicator PCBs. This is an extension of the study into dioxins and dioxin-like PCBs (Freijer et al., 2001), in which the dietary exposure was estimated for dioxins, based on results of chemical analyses of collected foods and food consumption data. The collected samples were also analysed for indicator PCBs.

The main objectives are:

- a) to obtain representative data on levels of PCBs in foods consumed by the general population in the Netherlands.
- b) to estimate dietary intake in the population and the relative importance of specific food groups to the total intake of indicator PCBs.
- c) to compare the results of the current survey with previous duplicate diet studies.

2 Sampling strategies

The samples that were primarily taken for the study into the dietary intake of dioxins and dioxin-like PCBs (Freijer et al., 2001) were analysed for the indicator PCBs in the current study. PCBs, just as dioxins, accumulate in fat. Consequently, the sampling strategy that was the basis of the study of Freijer et al. is also applicable for the indicator PCBs. Therefore, Chapter 2 of Freijer et al. will be summarized here.

2.1 Consumer foodstuffs

2.1.1 Introduction

A sampling programme was designed to obtain representative data on levels of lipophilic components like dioxins and PCBs in foods consumed by the general population in the Netherlands. The sampling strategy is based on the assumption that these substances are almost entirely present in the fat fraction of the foodstuffs. Using the results of the third Dutch National Food Consumption Survey 1997/1998 (DNFCS 3), 24 food categories were defined to cover most of the fat containing foodstuffs. In order to meet the needs to improve the estimate of the contribution of other foods one food category containing vegetables and three categories containing complex dishes and mixed products were recognised.

In co-operation with the Dutch Inspectorate for Health Protection and Veterinary Public Health (Regionale Keuringsdiensten van Waren) a sampling protocol was formulated. For the defined food categories relevant foods were purchased and pooled, to measure the contaminant levels in composite samples. The choice of food products in each category was based on their portion in the total (fat) intake of these products as derived from DNFCS 3.

2.1.2 Materials and methods

Selection of foods

For the selection of foods, the database of the DNFCS 3 was used. The survey has been described in detail elsewhere (Voedingscentrum, 1998; Kistemaker et al., 1998). The food consumption of 6250 individuals (2770 households) was assessed by a 2-day dietary record method, equally distributed over the seven days of the week and over a whole year. For each subject, data on age, sex, body weight and a series of other characteristics were available. The population ranged from 1 to 97 years in age. For each person, the quantities of various ingested food items over the day were recorded. This resulted in consumption data of 1209 different food products. Of each food product, a comprehensive description of the food items, including percentage total fat, was available from the Netherlands Nutrient databank (NEVO, 1996). The descriptions in this databank were used to investigate

the type(s) of fat or oil in the 1209 food products included in the DNFCS 3 database. Food products not expected to contain dioxins or PCBs were not considered in the selection procedure (Kistemaker et al., 1998). This screening procedure resulted in a reduction of 1209 to 807 food products ranked into food categories according to type of fat or oil. The database of the DNFCS 3 was also consulted to perform a secondary screening. This screening aimed at identifying the food categories most significantly contributing to total fat consumption. This resulted in a selection of 18 food categories with differing types of fats and oils. For each of these food categories, a set of food products was defined covering at least 95% of the total fat intake of the respective category.

For the category vegetables the fat based approach was assumed to be impractical. The 21 most popular vegetables were needed to cover 95% of the average intake of the vegetables by the Dutch population. Sampling of the following food categories was assumed to be not necessary or was abandoned in a later stage: (a) consumer milk, since data from a specific monitoring programme were available; (b) the categories liver, game and horses, because of their relatively small contribution to the average diet; and (c) mixed categories like mixed products, complex dishes, pastries and sweets, since these average levels can be calculated from other categories by the CPAP (Conversion of Primary Agricultural Products) programme (Van Dooren et al., 1995). Summarising, 24 food categories were defined for which levels of dioxins and dioxin-like PCBs had to be established, either by direct chemical analysis or by use of the CPAP programme. These food categories are shown in Appendix 1.

Collection and composition of samples

Samples were collected from all selected food products belonging to each food category. During sampling, possible differences between geographical areas and seasons and the proportional contributions of the various food products have been taken into account. To reduce the number of measurements, a general sampling scheme was followed as illustrated in Figure 2.1. The composition of the samples for chemical analysis was aimed at the highest attainable degree of national representativity for each of the selected food categories.

Almost all categories were sampled using a proportional technique. The samples were prepared from a mixture of different food products, with each item added in weight proportional to its average consumption, as determined in the DNFCS 3. Appendix 1 lists the individual amounts of the food products combined to a (regional) representative sample for each of the respective food categories. The selection of food items to be mixed had to cover at least 95% of the fat intake of the respective category according to the DNFCS 3. All these composite samples held at least a total of 50 g of fat. For the category vegetables the fat based approach was not used. Instead, the 21 most popular vegetables were mixed to cover the required 95% of the average intake of the vegetables by the Dutch population. The vegetables were not washed or cleaned before adding them to the composite samples. The items themselves were collected in five different regions in the Netherlands. In each region, two different inspectors of a Regional Inspectorate for Health Protection and Vet-

erinary Public Health collected independently from each other the complete set of requested food items at stores of their own choice. A protocol containing general instructions and a detailed account of the amounts of each food product to be collected were provided to the respective inspectors. The samples were collected in March 1999. Then all samples were transported to the regional laboratory 'East' of the Inspectorate for Health Protection and Veterinary Public Health (Keuringsdienst van Waren Oost). The preparation of the regional and national composites (Figure 2.1) was carried out at the Laboratory of the Regional Inspectorate in Nijmegen and at the Laboratory for Organic-analytical Chemistry of RIVM. All samples were stored frozen at -20°C until chemical analysis. Some food categories were not sampled/analysed. For details see Freijer et al. (2001).

Consumer milk

Nowadays the milk that reaches the majority of consumers in The Netherlands is produced by only two major manufacturers that have factories distributed all over the country. Milk produced by these factories (further referred to as 'consumer milk') represent pooled samples of milk produced by dairy farms evenly distributed over the country. Only minor fractions are brought directly on the market by individual dairy farms. A monitoring programme on consumer milk is active since October 1997. Every few days cartons of milk containing 1 litre each are bought at local stores. By mixing equal weight amounts of the temporal samples, a monthly representative pooled sample was obtained. After the merging of some of the manufacturers, the sampling campaign was reduced to the North-Eastern and South-Western region as of January 1999. The results from the monthly samples of March 1999 were used as input in the intake calculations. The original study design of the monitoring programme consisted of chemical analysis of PCDDs and PCDFs only. In the framework of this dietary intake study, the regional composites from March 1999 were also analysed for contents of indicator PCBs.

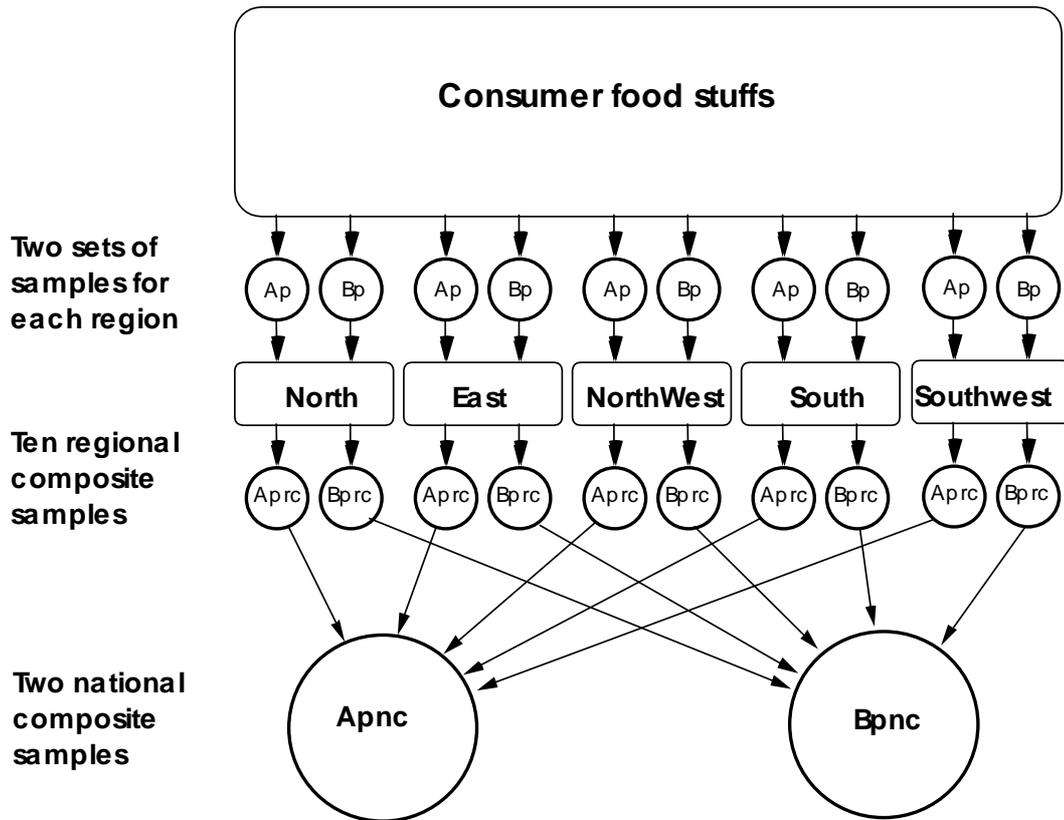


Figure 2.1. Schematic presentation of the applied strategy for sample collection and preparation of national representative composite samples to determine the levels of PCDDs, PCDFs and PCBs in almost all categories of food consumed in the Netherlands

In each of the five regions, two sets of samples ('Ap' and 'Bp') were collected by two different inspectors independently, consisting of various food products. Next, regionally collected food items were mixed either proportionally, when composed of different types of food items (covering 95% of the fat intake of the respective food category), or by weight (vegetables). In preparing the regional composite samples ('Aprc' and 'Bprc'), of each food product either the Ap or Bp sample was used. Next, national representative composite samples ('Apnc' and 'Bpnc') were composed by mixing equal weights of five Aprc and Bprc mixtures, respectively.

3 Measurements of concentrations of indicator PCBs in foodstuffs

3.1 Consumer foodstuffs

3.1.1 General principle of the chemical analysis

Samples were extracted with organic solvent(s) in order to isolate the fat fraction containing the PCDDs/PCDFs and PCBs. Before fat clean-up, $^{13}\text{C}_{12}$ -labeled standards of the indicator PCBs were added to the samples in order to quantify and identify the compounds according to the isotope dilution technique. An aliquot of extracted fat was dissolved in hexane and purified on a silica chromatographic column in a normal phase LC system. The eluting fraction containing the compounds of interest was collected and concentrated. Analysis was performed by injecting aliquots of the concentrated eluates into a GC/MS system.

In the following sections, the various extraction steps and additional refinements applied in the analysis of the different food samples will be described.

Sample extraction

After homogenisation, an amount of sample was weighed for extraction. The further preparation and extraction procedures carried out for the different food items are outlined in detail in Freijer et al. (2001). After refluxing, all extracts were evaporated to dryness and the amount of (extracted) fat was weighed to determine the fat content of the original sample.

Clean-up

An aliquot of the extracted fat was dissolved in hexane at a concentration of 45 mg/ml, while 200 μl PCB $^{13}\text{C}_{12}$ labels were added. A volume of 400-600 μl of this extract was injected onto a normal phase HPLC system, equipped with a silica column. A fraction of 4 ml containing the compounds of interest was collected, evaporated to dryness and redissolved in 50 μl toluene containing $^{13}\text{C}_{12}$ -labeled PCB 80 as internal sensitivity standard.

Gas chromatography - mass spectrometry

GC/MS analyses were performed on a VG70SQ or AutoSpec (Micromass, Manchester, UK) mass spectrometer coupled to a HP 5890 (Hewlett Packard, Palo Alto, MA, USA) gas chromatograph. GC separations were carried out on a non-polar column (60 m DB-5MS ; J&W Scientific, Folsom, USA; 0.25 mm ID, 0.10 μm film thickness). The temperature programme consisted of an isothermal period (100°C, 1 min), a rise at 15°C/min to 175°C, then at 4°C/min to 290°C and finally a second isothermal period of 2 min at 290°C. Samples for the PCBs analysis were injected using a CTC-A200s (CTC-Analytics,

Zwinger, CH) autosampler, and helium was used as carrier gas at a linear velocity of 30 cm/s.

The GC/MS interface was maintained at 275°C in all cases. Ionization of samples was performed in the electron impact mode (EI) with 31 eV electrons. Instruments were operated at increased resolution. The resolving power (RP) was typically between (static) 3000 and 5000. Detection was performed by selected ion recording.

Quality Control

In general a Relative Standard Deviation of 10-15 percent is observed in a recent WHO intercalibration study on human milk and blood plasma as for quality control samples as analysed in the recent Dutch human milk study (WHO, 2000).

The total concentrations have been calculated in agreement with the 1991 intake study (Liem et al., 1991), assuming non-detects equal zero (the lower bound estimates).

3.1.2 Results and Discussion

The results of the analytical chemical analysis are summarised in Table 3.1 for each of the food categories. In this table the results from meat and dairy products are expressed as ng/kg fat, while the concentrations in fish and vegetable categories are expressed as ng/kg whole product. This difference is often made (EU-SCOOP, 2000) since the latter products might have very low or less informative fat contents.

For most of the categories comparison with SCOOP data is impossible since the composition of most of the samples is tailor made for an intake study of the Dutch population.

Average concentrations

The data in Table 3.1 present the average concentration in 16 food categories. In summary, the following may be observed:

- (a) Meat, dairy products and eggs: Concentrations of indicator PCBs in several types of meat, dairy products and eggs vary between 4 and 32 ng/g fat. The highest values were measured in chicken, the lowest in milk.
- (b) Fish: On a product basis, the average concentrations of indicator PCBs in fish are, just as the concentrations of PCDD/Fs, higher than in meat products. The highest average concentration is found in fatty fish: 31.8 ng/kg product. The lowest value is found in the category 'Fish, prepared' (1.04 ng/kg product).
- (c) Vegetable products: in the category 'Vegetables' the average concentration is 0.02 ng/kg product. In contrast with the dioxins, which showed levels in most vegetable products (on a product basis) comparable to, or lower than those in low-fat animal products (poultry, beef), the indicator PCBs have much lower concentrations in vegetables than in low-fat animal products.

Variation of measurement data

Table 3.1 shows the observed levels in each composite sample, as well as the average and the Relative Standard Deviation (RSD) between the two national composite samples. The RSD is the standard deviation of the two numbers divided by the average and multiplied by 100% to express the number as a percentage. From these individual RSDs a pooled RSD was calculated as the square root of the average of the squares of the RSDs. This pooled RSD, 47.8 %, is much larger than the RSD of 10-15 % that is observed in multiple analyses of a single sample. This suggests that the analytical variation is a minor part of the total variation indicating that the latter is dominated by the difference between the samples.

In contrast to most food categories, four categories show more pronounced differences between the two composite samples: eggs, margarine, nuts and fatty fish.

One of the main objectives of this study was to calculate the intake of indicator PCBs based on the average levels in the food categories reported. For this purpose it is important to note that for the general population the intake is not dominated by a single food category but is due to contributions of many food items. In that case, the influence of the random uncertainties in the pooled samples will have a large tendency to level out.

Table 3.1. Results of the chemical analysis of indicator PCBs in different categories of foodstuffs

Food category	Units	Average	RSD (%) *)
Milk	ng/g fat	4.0	30
Beef	ng/g fat	10.6	44
Pig	ng/g fat	6.7	7
Poultry	ng/g fat	32.2	25
Butter	ng/g fat	5.8	10
Egg	ng/g fat	15.7	77
Fried snacks	ng/g fat	3.1	12
Vegetable oil	ng/g fat	1.3	54
Margarine	ng/g fat	2.5	104
Cheese	ng/g fat	4.7	9
Vegetables	ng/g product	0.02	5
Nuts	ng/g product	0.9	61
Fish, prepared	ng/g product	1.0	4
Fish, fatty	ng/g product	31.8	97
Fish, lean	ng/g product	2.4	6
Crustaceans	ng/g product	8.3	1

*) Relative standard deviation.

The pooled standard deviation (all samples together) is 47.8 %.

4 Dietary intake of indicator PCBs

In this chapter the dietary intake of the sum of indicator PCBs is assessed by combining data on concentrations of indicator PCBs in different food products and the consumption of these products.

4.1 Method

4.1.1 Method outline

Figure 4.1 displays the principal flow scheme which has been employed to estimate human dietary intake of dioxins and dioxin-like PCBs (Freijer et al., 2001). This flow scheme, which shows the relationships between different submodels and databases, was also used in the current study. In calculating the human exposure to indicator PCBs we use a two-step approach. In the first step, concentrations in consumed products are combined with the consumption rate of the products. Step one of the calculation yields two estimates of daily personal intakes for all individuals included in the survey. These estimates contain the basic information for the second step, which consists of the evaluation of the intake distribution for the population with the statistical exposure model STEM (Slob, 1993a; Slob, 1993b).

4.1.2 Dutch National Food Consumption Survey

The consumption of the Dutch population was examined with the third Dutch National Food Consumption Survey (DNFCS 3). This survey describes the consumption of the Dutch population and includes information on the daily consumption over two consecutive days and a record of age, sex and body weight of 6250 individuals (Kistemaker et al., 1998).

4.1.3 Concentrations in individual products

To calculate the intake, concentrations of indicator PCBs in the consumed products are needed. In the joint monitoring program of RIVM and RIKILT for the 'dioxin intake study' (Freijer et al., 2001) Dutch consumer products were collected and analysed. The programme on consumer products yielded concentrations of composite samples of 24 food categories (see Appendix 1). Usage of the DNFCS 3 in calculating dietary intakes requires the concentration to be known in much more detail, i.e. at the level of the individual products of the Dutch Nutrient Databank ('NEVO-products'; NEVO, 1996). The CPAP (Conversion of Primary Agricultural Products) programme (Van Dooren et al.,

1995) was used to translate the concentrations found in the measurement programme for each food category to those in the NEVO products.

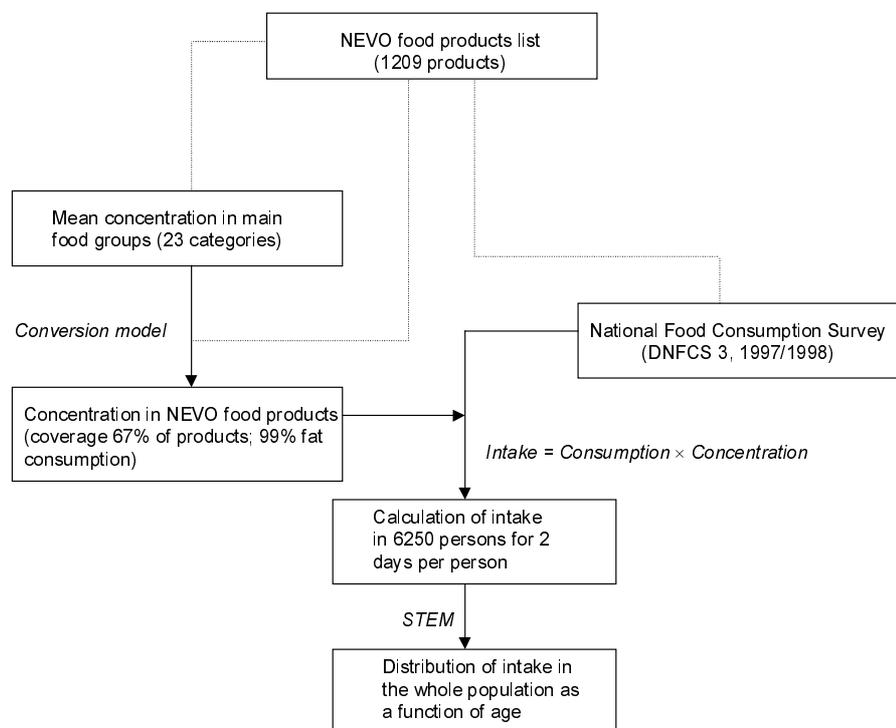


Figure 4.1. Overview of databases and submodels used in analysing human exposure to indicator PCBs

Three different procedures were used to assign concentrations to NEVO products:

- Concentrations in main food categories measured on a weight basis (i.e. in ng/g product) were directly assigned to each NEVO product belonging to that category.
- Concentrations in main food categories measured on a fat basis (in ng/g fat) were assigned to that NEVO product, using the fat content of the NEVO product considered to convert to a weight basis.
- The PCB concentrations of four main mixed categories (mixed products, complex dishes, pastries and sweets) were not determined in the measurement programme (Chapter 2). The concentrations of the NEVO-items belonging to these categories were estimated by calculating the weighted sum of two or more of the other categories, of which concentration levels were measured. Weights were assigned by dietitians. This procedure is part of the CPAP conversion model for Primary Agricultural Products (Van Dooren et al., 1995)

The conversion covered 67% of the NEVO items that were recorded in the DNFCS 3. The fat consumption via the selected NEVO products includes 99% of the fat consumption summed over all recorded NEVO products. The selected NEVO items thus represent the largest part of the products that are relevant for dioxin and PCB intake.

4.1.4 Statistical analysis

In the first step of the calculation, for each participant of the DNFCS the intake was computed for the two consecutive days considered in the survey. A frequency distribution of these intakes yields information on the variability of daily intakes in the population. Such a distribution shows the variation in short-term intake, but is unsuitable for an assessment of the long-term intake, which is required to assess the possible health risks of this intake. A distribution of life-long averaged intakes would be considerably narrower than the distribution of daily intakes, because within-subject variations level out. Another drawback of looking at just the frequency distribution of daily averaged intakes of all individuals in the survey is that no insight is gained in the relationship between age and intake in individuals. As a consequence, we performed a second step in calculating the dietary intake, namely a statistical analysis of the data.

Slob (1993a; 1993b) developed a statistical model for the description of dietary intake of chemicals with long-term effects (like dioxins and PCBs) for the population: the Statistical Exposure Model (STEM). STEM is intended to model the mean dietary intake as a function of age. It combines regression analysis on age by fitting a polynomial curve to the data, and nested variance analysis to separate within-subject variance from between-subject variance. The within-subject variance is estimated by analysing the differences between the intake during the two consecutive days for each person. By subtraction of the within-subject variance from the total variance an estimate can be made of the long term between-subject variance. The basic assumptions of STEM are as follows:

- (a) Intake in the population is lognormally distributed.
- (b) The within-subject variance is homogeneous throughout the population.
- (c) Intakes of the two consecutive days at which food consumption was recorded are not correlated.
- (d) The integral of temporal variations in concentrations of contaminants in consumed foodstuffs approximates the average concentration in these products as measured in this study.

Further details on the procedure and an extensive evaluation of the assumptions can be found in Slob (1993a; 1993b). The above assumptions limit the use of STEM to contaminants that can be found throughout the diet of the general population. Many environmental contaminants comply to this condition.

4.2 Results and discussion

4.2.1 Intake of DNFCS participants

Figure 4.2 shows a frequency distribution of the calculated intake of indicator PCBs, obtained by the direct combination of concentration data and food consumption patterns of 6248 individuals on two consecutive days. The average intake is 7.6 ng/kg bw/day, the 95th percentile is 19.9 ng/kg bw/day.

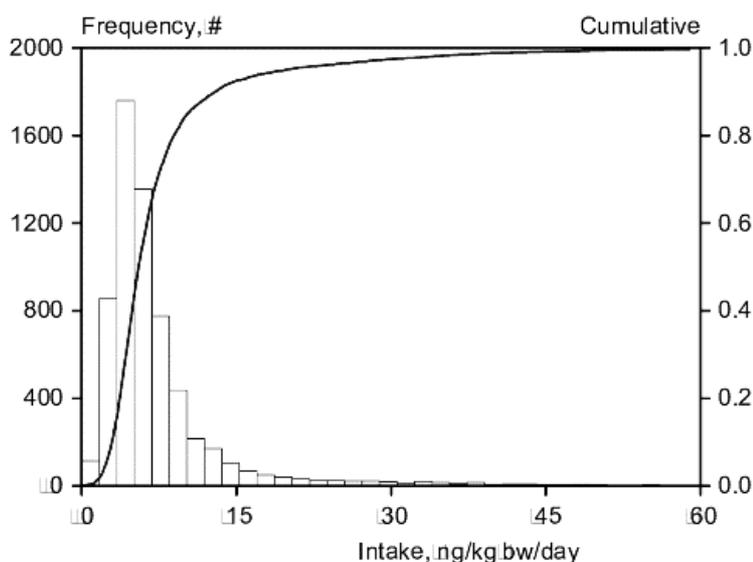


Figure 4.2. Frequency distribution of daily-averaged intake (in ng/kg bw/day) of indicator PCBs in the Dutch population. Data represent 12496 days of 6248 persons

4.2.2 Intake of the population

Processing of the intake data displayed in Fig. 4.2 with the statistical exposure model (STEM) yields the results presented in Fig. 4.3. This figure displays the relationship between intake and age. The upper panel (a) depicts the individual data points included in the frequency distribution of Fig. 4.2. Also, the median of each age class is indicated. These data are input to the STEM model, which estimates the median relationship with age as shown in the lower panel (b) of Fig. 4.3. The regression line (heavy line in Fig. 4.3b) shows that the fitted relative intake corresponds to the median value for each age class. The percentiles in the lower panel (b) of Fig. 4.3 represent the variation within the population (between-subjects variance, equal to 0.02627), which is obtained after subtracting the within-subject variance (0.06315) from the total variance (0.08942).

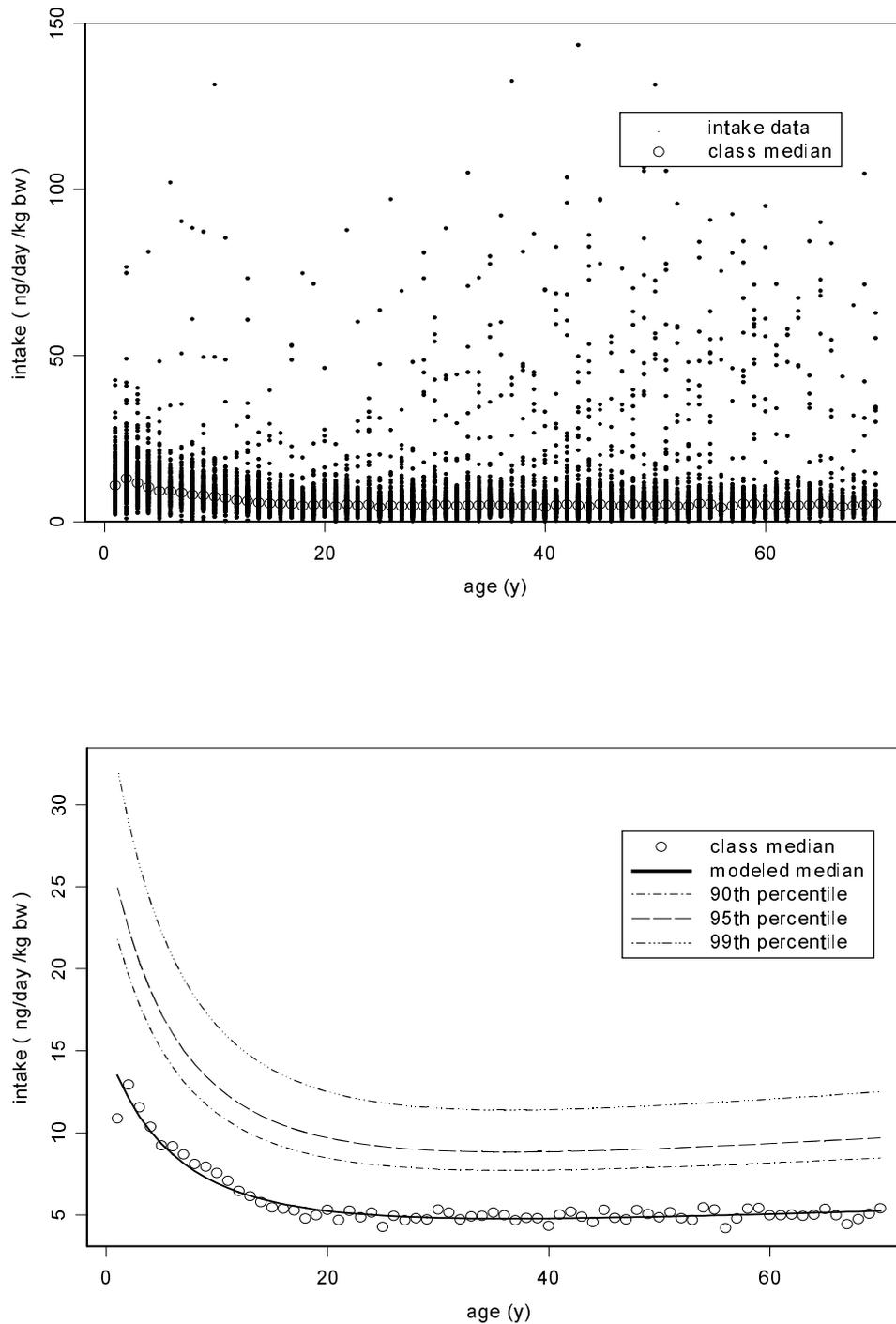


Figure 4.3. Relationship between intake of indicator PCBs and age. The upper panel (a) shows all the raw data (see also Fig 4.2) and the calculated median intake for each age class. The lower panel (b) depicts the intake distribution for the population after performing regression and nested variance analysis (Slob, 1993a) on the data of the upper panel. Percentiles refer to the between-subject variation.

Accordingly, the variation indicated by the percentiles is much less than the variation in the raw data in the upper panel (Fig. 4.3a).

From the lower panel (Fig. 4.3b) we can now deduce parametric intake distributions for each age class. Figure 4.4a-b displays two of these cross-sections for ages 2 and 40 yrs. Obviously, as dictated by the regression result, the median for young children is higher than for adults.

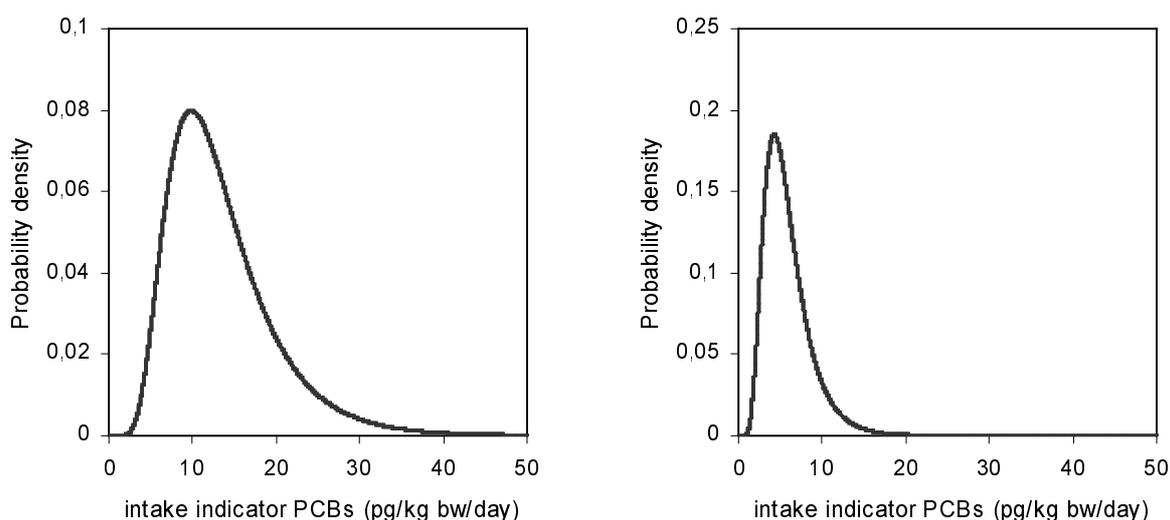


Figure 4.4. Modelled distribution of age-depending yearly-averaged intake of indicator PCBs (ng/ kg bw/day) in the population. Probability density refers to variation in each age class. Left panel: for age 2 yrs, right panel: for age 40 yrs. Note the difference in the probability density scales.

The results for age groups 2, 10 and 40 years are summarised in Table 4.1. The higher intake of children is mainly due to the high intake of food relative to their bodyweight, and not to the concentrations of contaminants in the food products eaten by children.

Table 4.1. Key statistics of intake distributions as calculated by STEM

	Intake (ng/kg bw/day)		
	2 years	10 years	40 years
median	12.1	6.9	4.8
average	15.3	8.7	6.0
90 th percentile	21.9	12.5	8.6
95 th percentile	25.7	14.7	10.1

The intake-age relationship established in the above described fashion is the starting point for the calculation of lifelong-averaged intake. The assessment of lifelong-averaged intake as well as the assessment with respect to possible adverse health effects are discussed in Chapter 5.

4.2.3 Contribution of different food groups

The intake of indicator PCBs is rather evenly distributed over the various food groups (Figure 4.5), just as it is for dioxins and dioxin-like PCBs (Table 4.2). A remarkable difference between the two compound classes can be seen for fish. While fish is an important route for indicator PCBs (26%), for dioxins (PCDDs/Fs) fish modestly contributes to the total intake (16%). For dairy products the opposite is true: dairy contributes for 17% to the intake of indicator PCBs and 27% to that of dioxins and dioxin-like PCBs.

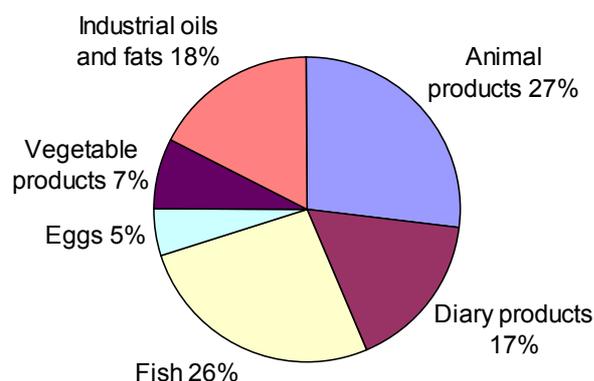


Figure 4.5. Estimated average contribution of food groups (%) to the intake of indicator PCBs in the Dutch population in 1998/1999

Table 4.2. Estimated average contribution of food groups to the intake of indicator PCBs, and total dioxins (PCDDs and PCDFs) and dioxin-like PCBs in the Dutch population in 1998/99. Absolute values are expressed per person

Food group	Contribution to total intake			
	Σ indicator PCBs		Σ dioxins + PCBs ^{a)}	
	ng/day	%	pg/day ^{b)}	%
Animal products	125	27	21	23
Dairy products	79	17	25	27
Fish	124	26	14	16
Eggs	23	5	3	4
Vegetable products	35	7	12	13
Industrial oils and fats	82	18	15	17
Total	468	100	90	100

^{a)} PCDDs, PCDFs and dioxin-like PCBs.

^{b)} WHO-TEQ.

5 Implications, trends and uncertainties

Section 5.1 assesses the dietary intake of indicator PCBs in the Dutch population with respect to possible adverse health effects. The comparison with earlier studies on the intake of indicator PCBs and the intake of dioxins and dioxin-like PCBs is presented in section 5.2. An overview of uncertainties that affect the results in this study is given in section 5.3.

5.1 Fraction of the population at risk for adverse health effects

In order to protect the general population against the adverse health effects of exposure to environmental contaminants, health safety objectives such as TDIs (Tolerable Daily Intake) and ADIs (Acceptable Daily Intake) have been derived. Since a TDI (or ADI) is established for lifelong intake, the exposure measure needs to have a matching time-frame. The lifelong-averaged intake is such a measure and can be derived from the intake-age relationship presented in Fig. 4.3. For the median of the population, this is done by integrating the age dependent median intake from age 1 to 70 yrs, and expressing the result on a daily basis. As such, in this scenario it is assumed that exposure concentrations in food remain unchanged throughout one's life. This means that the potential effect of the current exposure conditions is evaluated as if it would be effective on a lifelong period. The median lifelong-averaged intake is estimated to be 5.6 ng/kg bw/day, the 95th percentile in the population is 11.9 ng/kg bw/day (Fig. 5.1).

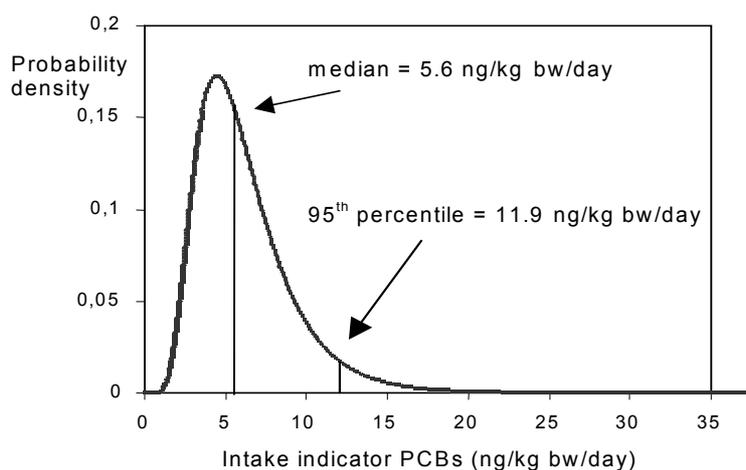


Figure 5.1. Modelled distribution of lifelong-averaged intakes (1-70 years; ng/kg bw/day) of indicator PCBs in the Dutch population

In 2001 RIVM estimated a MPR (Maximum Permissible Risk level, TDI) for the sum of the seven indicator PCBs of 10 ng/kg bw/day (Baars et al., 2001). This MPR for PCBs was derived from a long-term toxicity experiment with rhesus monkeys orally exposed to Aroclor 1254 for 55 months, resulting in decreased specific and non-specific immune parameters (no NOAEL, LOAEL was 5 µg/kg bw/day; Tryphonas et al., 1991). Applying an uncertainty factor of 250 a MPR for Aroclor 1254 of 20 ng/kg bw/day was derived, which was converted to a MPR of 10 ng/kg bw/day for the sum of the seven indicator PCBs (assuming that approximately half of Aroclor 1254 consists of indicator-PCBs; Baars et al., 2001).

It must be noted, however, that this MPR was derived in the framework of the 'Dutch Intervention Values' for soil contamination (Lijzen et al., 2001), which seriously limits its applicability. It has also to be appreciated as provisional, just because this particular application and because it was estimated on the basis of an experiment with a commercial PCB mixture which consisted of both dioxin-like and non-dioxin-like PCBs. As yet it is quite difficult – if not impossible – to distinguish between the toxicity caused by either type of PCBs.

Toxicity studies with individual congeners have been published though, but these studies are limited in number, they cover a limited number of congeners, they are limited in scope, and only evaluate a few specific toxicological endpoints (ATSDR, 2000). Hence the derivation of a TDI based on the toxicity of individual congeners is as yet not possible.

However, some conclusions can be drawn from an evaluation of the composition of Aroclor 1254, the PCB mixture that was used in the pivotal animal experiment.

Reviewing the average composition of Aroclor 1254 as reported by the ATSDR (2000), it can be concluded that one gram of Aroclor 1254 may contain 17–51 µg TEQ resulting from the amount of dioxin-like PCBs in Aroclor 1254. In other words, the 'dioxin-like' toxicity of 1 gram of Aroclor 1254 due to dioxin-like PCBs equals the toxicity of 17 to 51 µg TCDD. The latter value is taken as being representative for a worst case situation.

In addition, most PCB-mixtures contain PCDDs and PCDFs as impurities formed during production. Based on the amount of these impurities in Aroclor 1254 as reported by the ATSDR (2000), an additional TEQ of 1.7 µg per gram Aroclor 1254 can be calculated.

In conclusion, 1 gram of Aroclor 1254 might thus contain maximally 51 µg TEQ due to dioxin-like PCBs, and 2 (rounded figure) µg TEQ due to PCDD and PCDF impurities present, in total approximately 53 µg TEQ per gram.

The pivotal study on the toxicity of PCBs using Aroclor 1254 resulted in a LOAEL of 5 µg/kg bw/day. Theoretically this dose might thus have contained in total 265 pg TEQ. This is about 100 times the TDI for dioxins of 2 pg/kg bw/day as recently established by the Scientific Committee on Food of the European Commission as tolerable weekly intake of 14 pg/kg bw/week (SCF, 2001).

From this estimation it seems that the results of the chronic monkey study with Aroclor 1254 might have been considerably influenced by the presence of dioxins (dioxin-like PCBs, PCDDs and PCDFs) in this specific Aroclor mixture. It should be noted, however,

that the time frame and the endpoints of the studies for PCBs and for dioxins were totally different.

This particular monkey study was also used by the ATSDR to derive a MRL (Minimum Risk Level, TDI) of 20 ng/kg bw/day for chronic exposure to PCBs (ATSDR, 2000).

From the intake distribution as illustrated in Fig. 5.1 it can be concluded that approximately 10% of the population has an intake of more than 10 ng/kg bw/day and 5% has an intake of more than 12 ng/kg bw/day.

Because PCBs express their toxicity on the central nervous system, the thyroid gland and other endocrine organs only after either a very high incidental intake or after bioaccumulation in the body upon long term intake, it is not expected that the intake of people in the upper 5th percentile of the distribution curve (> 12 ng PCBs per kg bw per day) for a relatively short period will result in any clinical adverse health effects.

However, people consuming relatively large amounts of fish and fish products and/or meat and meat products are not only at risk of exceeding the TDI of dioxins (approximately 8% of the general population has an intake of dioxins and dioxin-like PCBs that is higher than the TDI of dioxins; Freijer et al., 2001), but may also be at risk for toxic effects caused by PCBs. As a consequence, it must be concluded that monitoring of the dietary intake of indicator PCBs is just as important as monitoring of dioxins and dioxin-like PCBs. Likewise, attempts to decrease the exposure to PCBs should deserve the same attention of risk management authorities as the attempted decrease of exposure to dioxins and dioxin-like PCBs.

5.2 Time trend

To obtain a time trend, the average intake of indicator PCBs was compared to the average intakes as measured in previous 24-h duplicate-diet studies performed at RIVM (Liem and Theelen, 1997): from 83 ng/kg bw/day in 1978 to 39 ng/kg bw/day in 1984/85 and 10 ng/kg bw/day in 1994. This considerable decrease is illustrated in Fig. 5.2, but shows also that the decrease is currently flattening out. The relative decrease in the intake of dioxins and dioxin-like PCBs is comparable to that of the indicator PCBs (Fig. 5.2).

Comparison of duplicate diet studies with the outcome of model calculations may not be completely fair, because both methods have different sources of error and uncertainty. For the duplicate diet study underreporting may be a source of error. This phenomenon may also occur in the Food Consumption Survey used in the model calculations. Another error in the latter type of study may be that the calculated average concentrations in the foodstuffs may not be representative. However, the smoothness of the curve for the dioxins as presented in Fig. 5.2, composed of measurements of both methods, suggests a fair comparability of both types of studies.

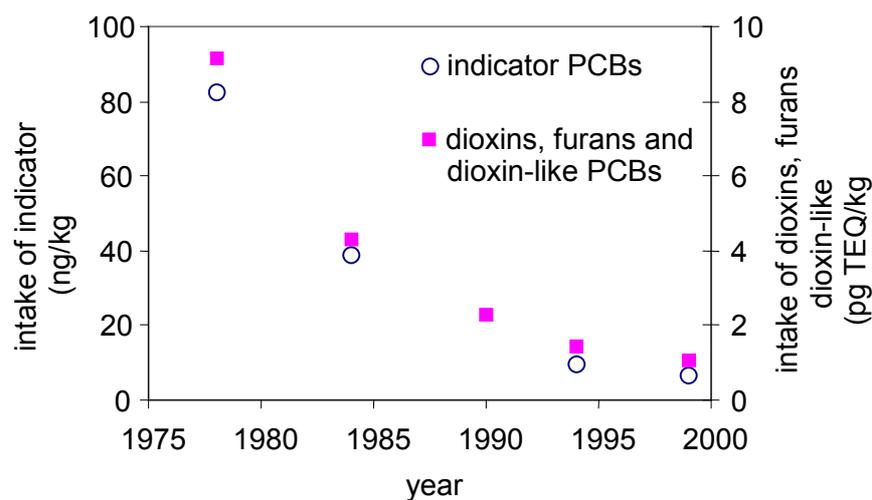


Figure 5.2. Time trend of dietary intake of the sum of indicator PCBs (circles, primary y-axis) and the sum of dioxins, furans and dioxin-like PCBs (squares, secondary y-axis)

The studies of 1978, 1984 and 1994 were 24-h duplicate diet studies (Liem and Theelen, 1997). The studies of 1990 (Liem et al., 1991) and 1998/1999 (present study) were intake calculations based on measured concentrations in food samples.

5.3 Uncertainty in data and methods

For a full discussion of the uncertainties in the analytical data and methods the reader is referred to Freijer et al. (2001). Of course, the present report addresses indicator PCBs and not dioxins, and hence for the former substances a short discussion is presented below.

The differences between the concentrations in the two series of samples, expressed as a pooled relative standard deviation (RSD) for indicator PCBs is 47.8 %. This is higher than the RSD for the dioxins and dioxin-like PCBs (34 %). Whereas for most food categories the RSD is comparable for the two compound classes, for the categories margarine, nuts and fatty fish the RSD for indicator PCBs is much larger than that for the dioxins and furans. An explanation for this observation may be that the two compound classes stem from different sources. While dioxins and furans are mainly emitted via air, the main source of PCBs is soil. It can be expected that the distribution of compounds in soil is usually more heterogeneous than that of compounds in air, consequently leading to larger differences between the concentrations of different samples.

6 Conclusions

In the past decades, the dietary intake of indicator PCBs in the general population has decreased considerably. However, a small part of the population still has a rather high intake. If this high intake only occurs for a limited period of time, it is not expected to result in adverse health effects.

To provide regulators with a health-based guideline to prevent adverse health effects of exposure to indicator PCBs, the derivation of a TDI, preferably by international bodies, is recommended.

The contribution of different food groups to the average intake of indicator PCBs is rather evenly distributed. For both the PCBs and the dioxins animal products (including dairy products and fish), and oils and fats constitute almost 90% of the intake of the general population.

Since a high intake of PCBs (with as yet not quantifiable adverse health effects) is usually accompanied by a high intake of dioxins, monitoring of the dietary intake of PCBs is just as important as that of dioxins. Attempts to decrease the exposure to both groups of compound deserves therefore continuing attention.

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Appendix

Composition of the food categories. The composition of the food categories represents at least 95% of the fat intake of the respective category. However, for vegetables the composition represents at least 95% of the product intake of the respective category. See also the text in chapter 2.

Category	Foodstuff	Foodstuff (in Dutch)	Mass g	Contribution %
Animal products				
Beef	minced cows meat	gehakt runder-	211	59.1%
	hamburger	hamburger	64	17.9%
	braising steak	rundersucadelappen	43	12.0%
	stewing steak	runderriblappen	20	5.6%
	ground cows meat	rundertartaar	19	5.3%
		Σ	357	
Pig	pig sausage	varkensbraadworst	61	29.3%
	bacon (lean)	speklap, mager, zonder zwoerd	55	26.4%
	chop	varkenshals/schouderkarbonade	71	34.1%
	pork	spek vers, vet rauw	9	4.3%
	kromenski	slavink	12	5.8%
		Σ	208	
Poultry	chicken skinless	kip/bout zonder vel	335	31.3%
	chicken with skin	kip/bout met vel	147	13.7%
	chicken meat	kipfilet	547	51.1%
	turkey	kalkoen	31	2.9%
	chicken liver	kippenlever	11	1.0%
		Σ	1071	
Dairy products				
Butter	butter unsalted	boter ongezoeten	43	69.4%
	butter salted	boter gezouten	16	25.8%
	butter (half)	boter halfvolle kuip (Linera of ander)	3	4.8%
		Σ	62	
Cheese	cheese Gouda	kaas Goudse 48+	136	84.0%
	cheese Edam	kaas Edammer 40+	15	9.3%
	cheese "Maaslander"	kaas Maaslander 48+	5	3.1%
	brie 50+	kaas Brie 50+	3	1.9%
	brie 60+	kaas Brie 60+	3	1.9%
		Σ	162	
Fish				
Lean fish	cod	kabeljauw	2337	66.4%
	plaice	schol	797	22.6%
	coal-fish	koolvis	385	10.9%
		Σ	3519	
Fatty fish	herring (salted)	haring gezouten	188	57.5%
	eel (smoked)	paling gerookt	34	10.4%
	mackerel (steamed/raw)	makreel gestoomd/rauw	34	10.4%
	mackerel (smoked)	makreelfilet gerookt	17	5.2%
	salmon (raw)	zalm rauw	28	8.6%
	herring (marinated)	haring gemarineerd	26	8.0%
		Σ	327	
Crustaceans	mussels	mosselen gekookt	627	34.7%
	shrimps	garnalen gepeld	1079	59.7%
	crab	krab	58	3.2%
	lobster	kreeft	44	2.4%
		Σ	1808	
Fish, prepared	fried fish filet	lekkerbekje gebakken	257	59.9%
	fish fingers	vissticks	172	40.1%
		Σ	429	

continued

Appendix, continued. Composition of the food categories. The composition of the food categories represents at least 95% of the fat intake of the respective category. However, for vegetables the composition represents at least 95% of the product intake of the respective category. See also the text in chapter 2.

Category	Foodstuff	Foodstuff (in Dutch)	Mass g	Contribution %
Eggs				
Eggs	chicken eggs (cooked)	ei kippen- gekookt	472	100.0%
Vegetable products				
Nuts	peanut butter	pindakaas	33	30.8%
	peanuts (salted/unsalted)	noten pinda's gezouten/ongezouten	40	37.4%
	nuts (to go with cocktails)	noten borrel-	17	15.9%
	nuts mixture	noten gemengd gezouten	7	6.5%
	peanut sauce	saus saté- bereid	10	9.3%
			Σ 107	
Vegetable oil	margarine	margarine, kuipje/pak	34	41.0%
	low-fat margarine	halvarine	11	13.3%
	cooking-fat	vet bak- en braad-	4	4.8%
	potato crisps	chips	26	31.3%
	mayonnaise (80 % oil)	mayonaise 80% olie	8	9.6%
			Σ 83	
Vegetables	cauliflower	bloemkool	22	11.7%
	onion	ui	20	10.6%
	cucumber	komkommer	14	7.4%
	butter-bean	sperziebonen	14	7.4%
	carrot	wortelen	14	7.4%
	tomato	tomaat	12	6.4%
	chicory	witlof	12	6.4%
	leek	prei	10	5.3%
	Brussels sprouts	spruitjes	8	4.3%
	lettuce	sla	6	3.2%
	sauerkraut	zuurkool (k&k)	6	3.2%
	endive	andijvie	8	4.3%
	butter-bean (tin/glass)	sperziebonen uit blik/glas (k&k)	6	3.2%
	lettuce Iceberg	ijsbergsla	6	3.2%
	beetroot	bieten	4	2.1%
	French bean	snijbonen	4	2.1%
	mushrooms	champignons	6	3.2%
	tomato (cooked)	tomaat gekookt (k&k)	4	2.1%
	spinach (frozen)	spinazie diepvries (k&k)	4	2.1%
	spinach with cream (frozen)	spinazie à la creme (k&k)	4	2.1%
	broccoli	broccoli	4	2.1%
			Σ 188	
Industrial oils/fats				
Margarine (indus-	margarine	margarine, pakje	22	15.3%
	low-fat margarine	halvarine	11	7.6%
	cooking-fat	vet bak- en braad-	25	17.4%
	frying-fat	vet frituur-	4	2.8%
	French fries	frites bereid	82	56.9%
			Σ 144	
Fried snacks	minced meat hot dog	frikandel bereid	114	38.8%
	croquette	kroket bereid	98	33.3%
	sausage roll	broodje saucijze-	24	8.2%
	prawn crackers	kroepoek bereid	15	5.1%
	egg-roll (Chinese)	loempia bereid	43	14.6%
			Σ 294	

Appendix 2

Mailing list

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