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Dietary exposure to dioxins and dioxin-like PCBs in The Netherlands anno 2004

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ABSTRACT

In this study, representative occurrence data for PCDD/Fs and dioxin-like PCBs in food were obtained and used to estimate dietary exposure of the Dutch population. Food composite samples were analyzed as well as single fish and vegetables samples. Total dioxin concentrations in animal products ranged from 0.05 pg TEQ/g product in poultry to 2.5 pg TEQ/g product (using TEF_{2006}) in fish (shrimp), with 0.12 pg TEQ/g product being the lowest concentrations measured in fish (tuna). In vegetable products, concentrations ranged from 0.00002 pg TEQ/g product (white kale) to 0.19 pg TEQ/g (oils and fats). A long-term dietary exposure distribution was calculated using Monte Carlo Risk Assessment software. The lower bound median exposure of the Dutch population to PCDD/Fs and dioxin-like PCBs was estimated at 0.8 pg WHO-TEQ/kg bw/d, half of which were dioxin-like PCBs. Dairy was the main source (38%) due to its high consumption. Time-trend analysis shows that the exposure to dioxins has further decreased by 35% over the past five years. This is due to lower levels of dioxin-like compounds in most of the foods, mainly influenced by lower levels in meat and milk. The use of the new TEFs gives an exposure reduction of 10% with respect to TEF_{1998}. Still, 4% of the Dutch population exceeds the exposure limit of 14 pg/kg bw/week as set by the EU.

1. Introduction

Dietary exposure to polychlorinated dibenzo-*p*-dioxins (PCDDs)[1], polychlorinated dibenzofurans (PCDFs) and dioxin-like PCBs (polychlorinated mono-ortho (mo-PCBs), and non-ortho biphenyls (no-PCBs) has been studied in The Netherlands on a regular basis since 1978. PCDD/Fs are emitted via incomplete combustion processes from both anthropogenic and natural sources like waste incineration, chemical production, metal industry and to a small extent by forest fires and volcanic eruptions. In contrast, PCBs are released to the environment mainly via waste disposal (EC, 2006). Produced commercially since the 1920s, PCBs were used in various applications, namely the manufacture of electronic appliances. Many countries have restricted the use of PCBs in open systems since the 1970s. The use in closed systems, like transformers, is only still allowed in some countries (SCF, 2001; Baars et al., 2004; EC, 2006).

Since dioxins are lipophilic compounds, they accumulate in the food chain. Contamination of foods occurs through deposition of contaminated emissions on farmland, while fish accumulate dioxins through contamination of the aquatic environment. Exposure through food is the main source of dioxin exposure for humans, estimated at over 95% of the total intake for non-occupationally exposed persons (Parzefall, 2002). To study the effect of measures taken to decrease the exposure to these compounds, dioxin concentrations in food have been analyzed and the dietary exposure to dioxins has been calculated for the Dutch population on a regular basis (every 5–10 years) since 1978 (Liem et al., 1991; Liem and Theelen, 1997; Baars et al., 2004). In addition, dioxin concentrations in several products like milk, eggs and animal fat are analyzed for the EU monitoring program on a yearly basis (EC, 2006).

The EU Scientific Committee on Food (SCF) set a provisional tolerable weekly intake (pTWI) for dioxins at 14 pg TEQ/kg body weight (bw)/week, analogous to 2 pg TEQ/kg bw/d (SCF, 2001). To add up the concentrations of the different congeners, the different dioxins are assigned a toxic equivalency factor (TEF), which

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[1] *Abbreviations used:* PCDD, polychlorinated dibenzo-*p*-dioxin; PCDF, polychlorinated dibenzofuran; mo/no-PCB, polychlorinated mono/non-ortho biphenyl ether; SCF, Scientific Committee on Food of European union; pTWI, provisional tolerable weekly intake; TEF, toxic equivalency factor; TEQ, toxic equivalent factor; PBDE, polybrominated diphenylether; RIVO, Netherlands Institute for Fisheries Research; RIKILT, RIKILT-Institute of Food Safety; RIVM, National Institute for Public Health and the Environment; DNFCS, Dutch National Food Consumption Survey; CPAP, food conversion program; MCRA, Monte Carlo Risk Assessment software; LOQ, limit of quantification; ISU, Iowa State University (model); STEM, Statistical Exposure Model; pTDI, tolerable provisional daily intake.

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represents their relative toxic potency compared to 2,3,7,8-TCDD, the most toxic dioxin congener. TEFs can be assigned on the basis of congeners' similar modes of action as agonists of the aryl hydrocarbon receptor. After multiplying the dioxin concentration with its TEF, the concentrations of dioxins and dioxin-like PCBs can be summed and a total WHO-Toxic Equivalent (TEQ) concentration of a product can be calculated (Van Den Berg et al., 1998, 2006).

In the present study, we calculated the dietary intake of dioxins and dioxin-like compounds (referred to hereafter as "dioxins") based on new analyses of composite food samples and samples of different vegetables and fish performed in 2004. Part of these food samples have also been analyzed for polybrominated diphenylethers (PBDEs) for the calculation of dietary intake of PBDEs (De Mul et al., 2005; Bakker et al., 2008). The aim of the current study was to obtain representative occurrence data for dioxins in food in order to estimate dietary exposure and to determine the contribution of different food groups to the exposure. In addition, we compared the calculated exposure to previous exposure estimations of dioxins. Exposure was calculated with the recently re-evaluated TEFs (van den Berg et al., 2006), and compared to the result obtained using the old TEFs (1998).

2. Materials and methods

2.1. Food sampling

Food items were sampled to obtain representative samples of the consumed foods that were expected to contain dioxins based on previous studies. The majority of the samples were composite samples of different products representing a food category (Table 1). The selection of food items for these categories was based on a sampling program designed to obtain representative concentrations of dioxins in foods consumed by the general Dutch population. For fish and vegetables, samples from individual products were available instead of composite samples.

2.2. Analyzed food categories

2.2.1. Fish

Fish and shellfish (Category 3) were sampled by The Netherlands Institute for Fisheries Research (RIVO, currently called IMARES) in 2004 and the Dutch Consumer Association (Consumentenbond) in 2005. These datasets were combined to obtain a representative sample of fish consumed in The Netherlands. The RIVO sample collection was directed by the EU monitoring obligations for The Netherlands, meaning that 7 farmed fish species and 14 wild-caught fish species were sampled. The RIVO samples were obtained through fishermen, the fish auction, wholesale traders or through the research vessel "Tridens" (IMARES). Each sample consisted of 5–25 individual samples of the same fish species, except for shellfish. Shrimp samples were composed of 500 g uncooked shrimp and mussel samples of 100 g mussels, which were briefly cooked prior to collection of the meat from the shells. The number of analyzed samples per fish type is indicated in the footnotes to Table 1. The fat of the samples was collected according to an adapted method of Bligh and Dyer (de Boer, 1988). The fat was sent to the RIKILT-Institute of Food Safety for analysis of dioxins and dioxin-like PCBs.

In 2004, the Dutch Consumer Association sampled 15 individual fish from each of the most consumed fish species from supermarkets, fish shops and markets. The fish samples were frozen (−18 °C) until they were sent to RIKILT for analysis.

2.2.2. Vegetables

Data on dioxin concentrations in vegetables (category 4) was extracted from the report on the analysis of vegetables from The Netherlands sampled in 2001–2002 (Hoogerbrugge et al., 2004). Eight samples from each vegetable type (Table 3) were purchased at different locations across The Netherlands. The majority of the vegetables were grown in The Netherlands. From each sample, 30 g were selected after removal of non-edible parts and visual soil and dirt. Vegetables were analyzed at the National Institute for Public Health and the Environment (RIVM).

2.2.3. Meat, dairy, eggs, bread, fruits, fats and oils

Composite samples for the food categories 1 and 2 were analyzed as listed in Table 1.The sampling strategy was based on the assumption that dioxins are almost entirely present in the fat fraction of foodstuffs. Hence, food items were selected which together accounted for 95% of the fat consumption within that food group, typically three to five food items. Selection of the food items per food group was based on consumption data from the third Dutch National Food Consumption Survey (DNFCS 3), the same food survey used for the dietary intake calculations as described below. For the food groups cheese, oils and fats, beef, pork and poultry, processed foods such as minced meat and hamburger were sampled; for the other food groups, primary agricultural products such as eggs, milk, apple were sampled. Within Food Category 1, eleven units of each food item per food group and within Food Category 2, four units of each food item per food group were purchased in different supermarkets across The Netherlands in 2003. Per food group, the foods were mixed proportional to their average consumption and stored at −20 °C until analysis at RIKILT. To obtain a larger dataset for milk and eggs, we used additional samples from the EU monitoring program. As part of this program, laboratories analyze several foods every year.

2.2.4. Chemical analysis

The concentration of PCDD/Fs and dioxin-like PCBs in biological samples is typically very low, on the order of pg/g expressed as toxic equivalents (TEQ) to 2,3,7,8-TCDD. Therefore, highly sensitive and specific methods are required. The method used in this study is described in detail by Tuinstra et al. (1994). In short: First the fat is extracted quantitatively, then an extensive clean-up of the sample is performed to facilitate PCDD, PCDF and co-planar PCB analysis, and finally the dioxin concentration is quantitated with high-resolution gas chromatography–high-resolution-mass spectrometry (HRGC–HRMS). Separation between dioxins and fat, for example, is carried out using gel permeation chromatography. Vegetables were analyzed at RIVM using a similar method as described by Hoogerbrugge et al. (2004) and Baars et al. (2004). The data are stored in the Quality Agricultural Products Database (KAP) (Van Klaveren et al., 2006).

2.3. Dietary intake

2.3.1. Consumption data

Consumption data was collected in the third DNFCS conducted in 1997–1998, as described in detail by Kistemaker et al. (1998). Briefly, 6250 individuals, aged 1–97 years, from 2564 households were selected. Respondents weighed and recorded their food consumption over two consecutive days. The resulting consump-

Table 1
Sampling strategy for relevant food categories

Food groups per category	Number of samples within each composite sample	Sampling date, institute and sampling method
Category 1: butter, cheese, eggs[a], vegetable oils and fats, industrial oils and fats, bread, fruit	11	June 2004, RIVM, purchase of set of food products covering 95% of fat intake, for cereals and fruit 95% of product intake, in supermarkets
Category 2: beef, pork, poultry, milk[a]	4	Sept 2004, RIVM, purchase of set of food products covering 95% of fat intake, in supermarkets
Category 3: fish, crustaceans	5–25[b]	May–Sept 2004, RIVO, samples from research vessel, fish auction, fisherman, and 2004, Consumentenbond, supermarkets
Category 4: vegetables	8	2001–2002, RIVM, supermarkets
Category 5: complex dishes, bakery products, sweets	—	Concentrations of categories in group 5 were not measured but estimated with the food conversion program CPAP (Van Dooren et al., 1995)

[a] Additional milk and egg samples analyzed for the EU monitoring programme, were used in the respective categories. The additional samples contained 16 milk samples and 50 egg samples.

[b] Every fish sample consisted of 5–25 individual fishes. The composite sample of mussels consisted of a homogenate of 100 g mussel meat (mussels were briefly cooked to open the shells for collection of the meat). Shrimps (500 g) were homogenised and analysed as whole organisms (uncooked and unpeeled). For all fish one or more fish samples were available (mussels (100 g), plaice, sole, pollack, Victoria perch, oyster and cod $n = 1$, shrimp and tilapia $n = 2$; eel, mackerel and tuna $n = 3$; salmon $n = 4$; herring $n = 10$).