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Dietary exposure to non-dioxin-like PCBs of different population groups in Austria

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HIGHLIGHTS

- Levels of ndl-PCBs were determined in foods from the Austrian market.
- Dietary exposure was assessed for children, women and men.
- Dietary intake was well below the tolerable daily intake proposed by WHO.
- The health risk from the dietary intake of ndl-PCBs in Austria appears to be low.

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ABSTRACT

The dietary exposure to the sum of the six indicator PCBs (Σ_6 PCBs; PCB 28, 52, 101, 138, 153, and 180) across different Austrian population groups was assessed in this study by combining data on occurrence from food of the Austrian market (n = 157) analysed during 2006–2011 with national food consumption data. The most contaminated food group was meat, poultry, game and offal with average levels of ndl-PCBs of 5.20 ng g^{-1} fat. In fish and fish products and eggs, mean concentrations of 3.89 ng g^{-1} fresh weight (fw) and 4.00 ng g^{-1} fat, respectively, were found. In milk and dairy products average concentrations ranged from 3.07 to 4.44 ng g^{-1} fat. The mean dietary intake of Σ_6 PCBs was estimated to be 3.37 ng kg^{-1} bw d^{-1} for children (6–15 years old), 3.19 ng kg^{-1} bw d^{-1} for women (19–65 years) and 2.64 ng kg^{-1} bw d^{-1} for men (19–65 years). In all three population groups, milk and dairy products was the major contributing food group to the total dietary intake (50–55%) followed by fish and fish products (23–27%). The exposure of all Austrian population groups is well below the tolerable daily intake (TDI) of 10 ng kg^{-1} bw d^{-1} proposed by WHO, accounting for 34% in children, 32% in women and 26% in men.

1. Introduction

Polychlorinated biphenyls (PCBs) are persistent organic pollutants comprising a group of 209 congeners, which can be divided into two different groups according to their toxicological properties. Dioxin-like (dl) PCBs including 12 congeners exhibit a toxicological profile similar to that of polychlorinated dibenzo-p-dioxins and benzofurans (PCDD/Fs). The toxic and biological effects of these compounds are mediated via activation of the aryl hydrocarbon receptor (AhR) (Safe et al., 1985; Van den Berg et al., 2006). Non-dioxin-like (ndl) PCBs refer to the remaining congeners working through different mechanisms, not involving the AhR (US-EPA, 2003).

Historically, PCBs were widely used as technical mixtures in industry, notably as dielectric fluids in capacitors and transformers, heat transfer and hydraulic fluids, and as plasticizers in paints, flame retardants, pesticide extenders, and adhesives (ATSDR, 2000; US-EPA, 2003; EFSA, 2010). Although the production and use of PCBs has been banned in almost all industrial countries since decades, PCBs are still present and can be released into the environment. Improper disposal practices or leaks in electrical equipment and hydraulic systems are continuing sources of PCBs (WHO, 2002; EFSA, 2005). Due to their lipophilic properties PCBs accumulate in the food chain. Therefore, the main route of exposure to ndl-PCBs with a contribution of more than 90% in the general population is via food (EFSA, 2005).

Numerous toxicological effects have been reported for PCB mixtures and/or individual congeners in the literature. PCBs are associated with a number of adverse effects, such as effects on liver,

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thyroid, immune function, and behaviour, as well as reproduction function (EFSA, 2005). The International Agency for Research on Cancer (IARC) has classified PCBs in Group 1 as carcinogenic to humans (IARC, 2014).

Due to the inability to differentiate between toxicological effects of dl-PCBs and ndl-PCBs, the Scientific Panel on Contaminants in the Food Chain of EFSA did not establish a health-based guidance value for ndl-PCBs (EFSA, 2005).

However, for all 209 congeners a tolerable daily intake (TDI) of 0.02 μg kg^{-1} bw d^{-1} has been derived for mixtures of PCBs (WHO, 2003). Assuming that the sum of the so called indicator PCBs (Σ_6 PCBs; PCB 28, 52, 101, 138, 153, and 180) represents about 50% of all PCB congeners in food, a TDI of 10 ng kg^{-1} bw d^{-1} was initially proposed by the WHO at the "2nd PCB workshop" in Brno (Czech Republic, May 2002), but has already been used as a reference value by others (RIVM, 2001; AFFSA, 2007; VKM, 2008).

Many studies have reported dietary exposure to PCDD/Fs and dl-PBCs or PCBs in general (Arisawa et al., 2008; Windal et al., 2010; Kilic et al., 2011; Marin et al., 2011; Song et al., 2011; Tornkvist et al., 2011; Perello et al., 2012; Sirot et al., 2012), however, only a few focussed on the exposure assessment to ndl- PCBs. Dietary intake estimates to ndl-PCBs were published in France (Arnich et al., 2009), Belgium (Cimenci et al., 2013), Italy (Fattore et al., 2008), Norway (VKM, 2008) and Germany (Fromme et al., 2009).

To our best knowledge, this is the first time that dietary exposure to ndl-PCBs has been estimated for the Austrian population. In addition to previously published results for PCDD/Fs and dl-PCBs (Rauscher-Gabernig et al., 2013), the aim of the present study was to estimate the dietary exposure to Σ_6 PCBs in order to assess the risk for different Austrian population groups. The exposures were calculated by combining concentrations found within this study with national consumption data (Elmadfa et al., 2009).

2. Material and methods

2.1. Sampling

In total 157 food samples were collected and analysed during 2006–2011. The food samples were collected by official food inspectors at retail covering all nine provinces of Austria taking into account the main production areas of the respective food group (e.g. pork mainly from Upper Austria and Styria). Because food of plant origin does not contribute significantly to human exposure, only food of animal origin was taken into account in this study. Analysed food samples included: meat, poultry, game and offal; fish and fish products; milk and dairy products; and eggs and egg products.

2.2. Chemical analysis

Chemical analysis was performed by the Environment Agency Austria using the methodology for PCDD/F and PCB analysis in food based on EC Regulation 1883/2006 (EC, 2006). Although ndl-PCB were not included in the above mentioned regulation at the time of analysis, the herein described method was appropriate by adding additional ^{13}C-labelled standards for the 6 ndl-PCB congeners. The food samples, except eggs, were chopped, freeze-dried and ground. The fat was extracted in a Soxhlet apparatus for at least 8 h with mixtures of toluene/ethanol (2:1) for fish and eggs and *n*-hexane/dichloromethane (1:1) for all other food matrices (VWR, Leuven, Belgium). The egg yolks were separated from the egg white. Samples were homogenised and a subsample of 50 g was taken and mixed with 50 g ASE-Prep (Thermo Scientific, Sunnyvale, USA). Extraction was performed with a mixture of *n*-hexane/dichloromethane (1:1) using the Accelerated Solvent Extraction System ASE 300 from Dionex Corporation (Sunnyvale, USA). After determination of the fat content, a portion of the extract containing 5 g of fat was spiked with 6 ^{13}C-labelled ndl-PCB congeners (Cambridge Isotope Laboratories, Andover, USA). The fat was removed by treatment of the sample with concentrated sulphuric acid adsorbed on Celite (Roth, Karlsruhe, Germany). Further clean-up comprised a multilayer silica column and AlO*x* column, both used in a self-built semiautomatic system supplied from Fluid Management Systems (Watertown, USA). GC/HRMS was carried out on a Finnigan MAT 95 (Bremen, Germany) coupled to a HP 6890 gas chromatograph (Hewlett–Packard, Waldbronn, Germany) equipped with a cool injection system by Gerstel (Mühlheim/Ruhr, Germany). The columns used were J&W DB5MS and DBDIOXIN (Agilent Technologies, Santa Clara, USA) depending on the available capacity of one of the two existing GC/HRMS systems. The results from both columns are equal as confirmed by validation data and participations in proficiency tests. The mass spectrometer was operated in the multiple ion detection mode at a mass resolution of more than 8000. Detection and quantification limits for ndl-PCBs were in the range of 0.001–0.005 μg g^{-1} fat and 0.002–0.01 μg kg^{-1} fat, respectively. The measurement uncertainty of the method was calculated to be 20% and blanks have been detected in the range of 0.15–0.9 μg kg^{-1} fat. Participation in proficiency studies organised by the European Union Reference Laboratory for Dioxins and PCBs in Feed and Food ensure the quality of the method.

2.3. Food consumption data

Consumption data of the different population groups were obtained from a survey conducted within the scope of the Austrian Nutrition Report 2008 (Elmadfa et al., 2009). Food consumption data of women and men were collected by a 24-h recall; for children, the dietary assessment method of a 3-day food record was applied. The sample included 1345 females aged 19–65 years, 778 males aged 19–65 years and 757 children aged 6–15 years. Mean body weights were 63.6 kg for women, 81.5 kg for men and 39.7 kg for children. Gender differences in consumption habits of children are mainly reported for fruits and vegetables (Perez-Rodrigo et al., 2003; Wardle et al., 2004). Because these food groups are not included in the current study, no differentiation between male and female children was made in the exposure assessment. Dietary surveys are often biased because participants tend to report fewer and healthier foods. Although a method to exclude the so called "under-reporters" was applied, an underestimation of the actual exposure cannot be excluded.

2.4. Exposure assessment

For dietary exposure assessment, a deterministic approach was chosen which has already been described elsewhere (Rauscher-Gabernig et al., 2013). Briefly, the estimated dietary intakes were calculated by multiplying the mean food consumption data by mean concentrations of the individual PCBs in each food group. Dietary exposure to Σ_6 PCBs was assessed according to the following formula:

$$E = OxC/W$$

where E is the dietary exposure of Σ_6 PCBs in a particular food group (in ng kg^{-1} bw d^{-1}); O is the mean ndl-PCBs concentration in a particular food group (occurrence) expressed in ng g^{-1} fresh weight (fw) or ng g^{-1} fat; C is the mean consumption of the food group by different population groups in g day^{-1}; and W is the mean body weight of different population groups in kg.