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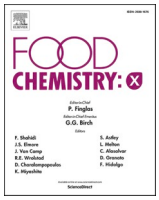

The first German total diet study (BfR MEAL Study) confirms highest levels of dioxins and dioxin-like polychlorinated biphenyls in foods of animal origin

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A R T I C L E I N F O

Keywords:
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Chemical compounds:
2,3,7,8-TCDD (PubChem CID: 15625)
1,2,3,7,8-PeCDD (PubChem CID: 38439)
1,2,3,4,7,8-HxCDD (PubChem CID: 38251)
1,2,3,6,7,8-HxCDD (PubChem CID: 42540)
1,2,3,7,8,9-HxCDD (PubChem CID: 29575)
1,2,3,4,6,7,8-HpCDD (PubChem CID: 37270)
OCDD (PubChem CID: 18636)
2,3,7,8-TCDF (PubChem CID: 39929)
1,2,3,7,8-PeCDF (PubChem CID: 42138)
2,3,4,7,8-PeCDF (PubChem CID: 42128)
1,2,3,4,7,8-HxCDF (PubChem CID: 51130)
1,2,3,6,7,8-HxCDF (PubChem CID: 42140)
1,2,3,7,8,9-HxCDF (PubChem CID: 51720)
2,3,4,6,7,8-HxCDF (PubChem CID: 43495)
1,2,3,4,6,7,8-HpCDF (PubChem CID: 38199)
1,2,3,4,7,8,9-HpCDF (PubChem CID: 41510)
OCDF (PubChem CID: 38200)
PCB 77 (PubChem CID: 36187)
PCB 81 (PubChem CID: 51043)
PCB 126 (PubChem CID: 63090)
PCB 169 (PubChem CID: 36231)
PCB 105 (PubChem CID: 36188)
PCB 114 (PubChem CID: 53036)
PCB 118 (PubChem CID: 35823)
PCB 123 (PubChem CID: 47650)
PCB 156 (PubChem CID: 38019)
PCB 157 (PubChem CID: 50891)

A B S T R A C T

The first German Total Diet Study, called the BfR MEAL Study, generated a comprehensive dataset of polychlorinated dibenzo-*p*-dioxins and -furans (PCDD/Fs) and dioxin-like polychlorinated biphenyls (DL-PCBs) in foods representative for the consumption habits in households in Germany. PCDD/Fs and DL-PCBs are persistent organic pollutants. Dietary intake is considered to be the most relevant exposure pathway for humans. Levels were examined in 300 foods that were prepared as typically consumed by the population in Germany. Highest PCDD/F and DL-PCB levels were detected in animal-based foods such as fish, butter, dairy products, liver, and meat. The comparison of conventionally and organically produced foods revealed a trend to slightly higher contents in organically produced foods. Sampling discriminated by region and season showed no major differences. Analysed occurrence data will improve future dietary exposure and food safety assessments in Germany.

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Introduction

Polychlorinated dibenzo-*p*-dioxins (PCDDs) and -furans (PCDFs), commonly referred to as dioxins, and dioxin-like polychlorinated biphenyls (DL-PCBs) are planar aromatic compounds, belonging to the persistent organic pollutants (POPs). Depending on the position of up to 8 chlorine atoms, 75 PCDD and 135 PCDF congeners occur, which often have similar chemical and toxicological properties (WHO, 2019). PCDD/Fs and DL-PCBs are lipophilic and very stable against chemical and microbiological degradation. They are found ubiquitously in the environment and easily enter and bioaccumulate in the food chain. Hence, food – especially food of animal origin – is considered to be the major source of human exposure to PCDD/Fs and DL-PCBs (greater than90%), with the exception of specific cases of accidental or occupational exposure (Beck, Dross, & Mathar, 1994; EFSA CONTAM, 2018). PCDD/Fs have never been produced on an industrial scale and have no technical use. They are formed unintentionally in a number of industrial and thermal processes as unwanted impurities or reaction by-products (EFSA CONTAM, 2018). PCBs were produced for various technical applications, e.g. as cooling liquids in transformers or as additives for hydraulic oils. Due to regulatory activities for PCDD/F emissions and the prohibition of PCBs in many countries, contamination levels decreased significantly during the last decades (Padberg, Bührer, Menzel, Weikert, Schaefer, & Abraham, 2018; Päpke, 1998). Nevertheless, exposure assessment and further monitoring of PCDD/Fs and DL-PCBs is important, as adverse effects can already occur at very low levels of exposure. Therefore, the European Food Safety Authority (EFSA) derived a tolerable weekly intake (TWI) of 2 pg TEQ/kg body weight per week which is seven times lower than the previous TWI (EFSA CONTAM, 2018).

The 17 PCDD/F congeners that are chlorinated at positions 2, 3, 7, and 8 are persistent and accumulate in fatty matrices. Their toxicity is mainly mediated by a common mode of action with binding to the aryl hydrocarbon receptor (AhR) and the subsequent regulation of genes involved in metabolism and endocrine pathways (Haws et al., 2006). Due to their coplanar structure, a subgroup of 12 out of 209 PCBs (DL-PCBs) acts via the same toxicological pathway. To express the cumulative toxicity of complex mixtures of PCDD/Fs and DL-PCBs in the same unit, the concept of the toxic equivalent (TEQ) was developed. The most toxic congener is 2,3,7,8-tetrachlorodibenzo-*p*-dioxin (TCDD) and assigned a toxic equivalency factor (TEF) of 1. TEFs of other congeners range between 0.0003 and 1 in case of 2,3,7,8-substituted PCDD/Fs, and between 0.00003 and 0.1 in case of DL-PCBs, demonstrating their relative toxicity in comparison to that of TCDD (van den Berg et al., 2006).

Numerous adverse effects following experimental PCDD/F and DL-PCB exposure have been described in laboratory animals including carcinogenic effects and effects on reproduction and development as well as on the immune system, the thyroid and the liver (EFSA CONTAM, 2018). Chloracne is the most unequivocal toxicity outcome following high exposure in humans (EFSA CONTAM, 2018). According to EFSA, effects with causal associations in humans are reproductive and developmental effects, including those on sperm quality, and effects on enamel and tooth development. The reported impact on sperm quality was regarded as the most sensitive health endpoint occurring already at low doses in rats and humans.

The availability of representative and reliable occurrence data for harmful chemical substances in the diet is crucial for derivation of assessments related to food safety. In addition to general food monitoring, Total Diet Studies (TDSs) represent a complementary and cost-effective public health tool to establish an extensive database of substances for exposure assessments (EFSA/FAO/WHO, 2011; Kolbaum, Jaeger, Ptok, Sarvan, Greiner, & Lindtner, 2022). The step of food processing is included in the design of a TDS, as levels of substances may increase, decrease or occur during industrial or at domestic food processing. This can lead to over- or underestimation in exposure assessments. The first German TDS, the BfR MEAL Study (meals for exposure assessment and analysis of foods), is one of the most comprehensive TDSs worldwide in terms of matrix-compound combinations. It was initiated in 2015 at the German Federal Institute for Risk Assessment (BfR, Berlin, Germany) following the guidelines of the EFSA, of the Food and Agriculture Organization of the United Nations (FAO), and of the World Health Organization (WHO) (EFSA/FAO/WHO, 2011). According to the three essential principles of a TDS, selected food items were: i) representative of the whole diet consumed by the respective population, ii) analysed as consumed, and iii) subsequently pooled to one sample if related.

The objective of this work was to provide novel data on PCDD/F and DL-PCB levels in foods from the German market that were bought and prepared as consumed in a representative household in Germany. Furthermore, the BfR MEAL Study intended to evaluate differences in levels between four regions of Germany, between two seasons as well as between conventional and organic type of production. The obtained occurrence data will be used for future dietary exposure and risk assessments in Germany.

Material and methods

Food sample selection and purchasing

The BfR MEAL Study fulfils the underlying principles of a TDS that are described elsewhere (Sarvan et al., 2017). Food sample selection (MEAL foods) was based on 24-hour dietary recall data of the second German National Nutrition Survey (NVS II, age 14–80 years) (Heuer, Krems, Moon, Brombach, & Hoffmann, 2015) and of a consumption survey among infants and young children (VELS, age 6 month up to 4 years) (Banasiak, Heseker, Sieke, Sommerfeld, & Vohmann, 2005).

For PCDD/Fs and DL-PCBs, the design of the BfR MEAL Study includes 300 selected MEAL foods and covers at least 90% of the average German diet for different age groups and gender. Using the FoodEx2 classification, the MEAL food pools are assigned to 17 main food groups (e.g., milk and dairy products, composite dishes) and were processed and prepared as consumed. The MEAL food list also includes food items rarely consumed by the population in Germany, but shown to have high PCDD/F and DL-PCB levels in national food monitoring reports. MEAL foods represent a variety of individual subsamples (e.g., variety, brand, production method, shopping location, and out-of-home consumption). Subsamples were selected according to German shopping behaviour based on market share data from a representative panel with 30,000 households as well as a telephone-based and online survey.

MEAL foods with no regional differentiation were purchased in the area of Berlin in line with representative German buying habits (e.g., farmers market, super market, and discounter). Regionally sampled MEAL foods were obtained in four regions of Germany (east, south, west, north) from three locations each (rural area, small city, big city) resulting in four different pools of a specific MEAL food according to regions. If differences between production types were expected, purchasing was divided into conventionally and organically produced foods. Seasonally sampled MEAL foods were purchased at two different times of the year to integrate the influence of global food supply including differences in the exporting country, climate, soil, and animal husbandry conditions. All food pools with and without differentiation (region, season, type of production) consist of 15 and 20 individual subsamples, respectively. Exceptions are the food pools edible offal (pork, beef), maize germ oil, and boletus/porcino mushroom with less

subsamples owing to the market situation. For the determination of PCDDs, PCDFs, and DL-PCBs, 300 MEAL foods were purchased between December 2016 and May 2019 including sampling for region (n = 51), type of production (n = 87), and season (n = 38) resulting in 645 food pools including 10,529 subsamples.

The details about number and differentiation of subsamples and food pools are summarised in Table S1. For PCDD/Fs and DL-PCBs, primarily foods where quantifiable results were expected were investigated, such as foods of animal origin, cucurbits, and foods prepared with animal-based ingredients, causing an adjusted, shorter food list compared to other substances analysed in the BfR MEAL Study.

MEAL food preparation and homogenisation

MEAL food samples were prepared in the study kitchen of the BfR with the objective to mimic the household behaviour of the population in Germany (e.g., sources of recipes, kitchen utensils, and browning degree). For this purpose, representative telephone ($n \geq 1000$) and online surveys ($n \geq 2000$) as well as most visited cooking homepages and most sold recipe books were considered. For ingredients that make up less than 5% of a recipe (wet weight), the most sold brand (conventional production) according to market share data was used.

After preparation, subsamples were pooled and homogenised using a knife mill (Grindomix GM200 and GM300, Retsch GmbH, Haan, Germany). Where required, ultrapure water (Milli-Q Integral 5, Merck, Darmstadt, Germany) or liquid nitrogen was added to achieve complete homogenisation. Samples were stored in amber glass vessels at −20 °C until analysis.

Analysis of PCDD/Fs and DL-PCBs

All PCDD/F and DL-PCB analyses were conducted by the accredited (DIN EN ISO/IEC 17025) Chemical and Veterinary Control Laboratory (CVUA-MEL, Münster, Germany) according to the performance criteria of the Commission Regulation (EU) No 644/2017 (EU, 2017).

Extraction and sample clean-up

Depending on the fat content, up to 100 g of a MEAL food pool homogenate was used for fat extraction. Solvents (Picograde) were purchased from Promochem (Wesel, Germany). Soxhlet extraction with toluene/acetone was applied for dry plant-based samples, cold extraction with dichloromethane/cyclohexane for pasty matrices (e.g., eggs, meat, fish, and liver) and liquid/liquid extraction with ethanol/*n*-pentane for milk samples. The extracts were evaporated to dryness with a rotary evaporator, permitting determination of the fat content. Oils and fats were used directly for further processing. After fat extraction, $^{13}C_{12}$ labelled internal standards (Wellington Laboratories, Guelph, Canada) were added.

For an automated sample preparation, the entire extract or up to 4 g of fat per sample were dissolved in 10 ml of *n*-hexane and loaded onto the sample loop of a DEXTech Plus device (LCTech GmbH, Obertaufkirchen, Germany). For extracted material with no fat, nonane was added as a keeper. The ready-to-use LCTech columns (acid silica, alumnia, and activated carbon) allowed a simultaneous clean-up of PCDD/Fs and DL-PCBs in a single run with two fractions (fraction 1: mono-*ortho* PCBs in *n*-hexane/dichloromethane, fraction 2: PCDD/Fs and non-*ortho* PCBs in toluene). After addition of 10 µl of dodecane and the recovery standards to the fractions, eluants were evaporated to dryness. The residue was dissolved in 100 µl of the recovery standard, transferred to a µ-vial and, depending on the fraction, further concentrated in a gentle stream of nitrogen to the required volume.

GC/HRMS detection and data evaluation

Samples were analysed by gas chromatography/high resolution mass spectrometry (GC/HRMS) using an Agilent 6890 GC (Agilent Technologies, Wilmington, USA)/Micromass Autospec Ultima HRMS) (Waters, Milford, USA) system with a resolution of R = 10,000. 2 µl of the PCDD/F and non-*ortho* PCB fraction were injected splitless to an Agilent J&W DB-5MS capillary column (60 m, 0.25 mm, 0.1 µm film thickness) with the following temperature program: 70 °C (2.5 min) to 170 °C (25 °C/min) to 290 °C (4 °C/min). Mono-*ortho* PCBs were analysed by injection of 2 µl splitless into an Agilent ZORBAX HT-8 capillary column (50 m, 0.22 mm, 0.25 µm film thickness) with the subsequent temperature program: 120 °C (2 min) to 180 °C (30 °C/min) to 310 °C (4 °C/min). To confirm the reproducibility of the measurement (single determination), analysed samples were repeated in regular intervals. A laboratory blank accompanied each batch of seven samples and a quality control pool was included in every fifth batch. The limit of quantification (LOQ) were determined in every sample for each of the 29 congeners according to the Commission Regulation (EU) No 644/2017. Regarding the toxic equivalent (TEQ) principle, the total toxicity of a mixture of congeners (17 PCDD/Fs, 12 DL-PCBs) in MEAL food pools was calculated by summing up each congener's detected level that was multiplied by its respective World Health Organization toxic equivalency factor (WHO_{2005}-TEF). In line with the EFSA guidance (EFSA CONTAM, 2018), results are expressed using the lower bound (LB) and the upper bound (UB) approach. At the LB and UB, the result of a congener below the LOQ was replaced by zero and by the numerical value of the LOQ, respectively. Depending on the sample and the kind of comparison, data are expressed in pg WHO_{2005}-TEQ/g fat (meat and meat products, excluding liver and edible offal; eggs and egg products; milk and dairy products, excluding buttermilk; animal and vegetable fats and oils) or in pg WHO_{2005}-TEQ/g wet weight (all the others like fish, seafood and invertebrates; composite dishes; food products for infants and toddlers). Occurrence data are shown as the sum of seven PCDDs and ten PCDFs (WHO_{2005}-PCDD/F-TEQ), the sum of twelve DL-PCBs (WHO_{2005}-PCB-TEQ) or as the sum of all 29 congeners (WHO_{2005}-PCDD/F-DL-PCB-TEQ or WHO_{2005}-TEQ). When comparing sum values of MEAL foods among the main food groups, all WHO_{2005}-TEQs were adjusted to the wet weight.

The laboratory analyses were conducted in a contract laboratory.

Statistics

If not stated otherwise, data were presented as mean UB level. Standard deviations determined on the main food group level reflect the variability between different MEAL foods in the particular group and do not consider the variation covered by single subsamples. Firstly, subsamples were aggregated to a pooled sample during sample homogenisation and secondly, data of single MEAL food pools of the same food were aggregated to the mean MEAL food result during calculation of statistical parameters. For LB and UB results of single food pools, please see Table S1.

Statistical analyses were performed using IBM SPSS Statistics for Windows, version 26.0. (IBM Corp., Armonk, USA). Testing for normal distribution was done with the Kolmogorov-Smirnov test. When comparing two datasets, statistical significance was evaluated using the Mann-Whitney *U* test for non-normally distributed data. Differences between four datasets were calculated using the Kruskal-Wallis test. Significance was accepted at p less than 0.05. As the tests only reveal differences across all MEAL foods sampled by production type or region, differences between individual MEAL foods were reported descriptively.

Results and discussion

PCDD/F and DL-PCB levels of foods in Germany

In the BfR MEAL Study, 645 food pools referring to 300 MEAL foods and 17 main food groups were analysed for PCDD/Fs and DL-PCBs. For the sum of PCDD/Fs and DL-PCBs (WHO_{2005}-TEQ) as well as for the sum of DL-PCBs (WHO_{2005}-PCB-TEQ), mean LB values were quantifiable for all food pools. The WHO_{2005}-PCDD/F-TEQ was quantifiable in 94% of

the food pools as at least one congener was quantifiable; levels for 36 food pools (33 MEAL foods) were below the limit of quantification (LOQ) (see Table S1).

Highest mean UB levels for the sum of PCDD/Fs and DL-PCBs were found in the main food groups containing products of animal origin, with the highest level measured in 'fish, seafood and invertebrates' (0.903 pg WHO_{2005}-TEQ/g wet weight) (see Table 1). Considerably lower levels were found in the categories 'animal and vegetable fats and oils', 'milk and dairy products', and 'meat and meat products' with 0.140, 0.061, and 0.053 pg WHO_{2005}-TEQ/g wet weight, respectively. Lowest mean UB levels in decreasing order were detected in the groups 'foods for infants and toddlers', 'coffee, cocoa, tea and infusions' and 'alcoholic beverages'.

Considering only food products of animal origin expressed on fat basis (see Table 1, values in parenthesis), the group 'milk and dairy products' showed the highest content (0.523 pg WHO_{2005}-TEQ/g fat) followed by 'animal fats', 'meat and meat products', and 'eggs and egg products' with 0.430, 0.258, and 0.187 pg WHO_{2005}-TEQ/g fat, respectively. On the individual food level, highest levels within these groups were observed in flavoured quark dessert, wild boar meat, butter, and hen eggs (see Table S2). Out of the samples of the main food group 'milk and dairy products', dairy products rich in fat displayed higher and most cheese products displayed slightly lower values than plain cow milk. Comparing the meat of different animal species, levels ranged from 0.128 to 1.07 pg WHO_{2005}-TEQ/g fat with highest levels in game (wild boar meat and deer) and sheep meat, followed by bovine, duck, chicken, and pork meat. Processed meat products showed the lowest levels within 'meat and meat products', ranging from salami-type sausage (pork, beef) with 0.330 pg to coarse cooked sausage (e.g., "Bierwurst", "Jagdwurst") with 0.064 pg WHO_{2005}-TEQ/g fat (see Table S2).

The 15 MEAL foods with the highest PCDD/F and DL-PCB levels are depicted in Fig. 1. MEAL foods with the highest UB levels mainly belonged to the main food group 'fish, seafood and invertebrates', with cod liver showing by far the highest value (13.7 pg WHO_{2005}-TEQ/g wet weight). The second highest level was found in smoked spiny dogfish (3.55 pg WHO_{2005}-TEQ/g wet weight), followed by eel and several herring products (see also section 3.2). Next to fish products, high UB levels were detected in sheep liver (0.644 pg WHO_{2005}-TEQ/g wet weight) and butter (0.388 pg WHO_{2005}-TEQ/g wet weight).

For most of the main food groups, DL-PCBs contributed less to the total WHO-TEQ (UB scenario) than PCDD/Fs (see Table 1). Lowest proportions were detected in the food groups 'coffee, cocoa, tea and infusions' (6%), 'starchy roots or tubers and products thereof' (11%), and 'products for non-standard diets and food imitates' (11%). A notable exception was the group 'fish, seafood and invertebrates' where DL-PCBs accounted for 77% to the total WHO-TEQ.

The congener profiles (LB approach) of selected animal-based MEAL food pools showed a similar pattern with some specific variations (see Table S3). Overall, 22, 19, 16, and 27 out of the 29 investigated congeners were found above LOQ in the food pools salmon, plain cow milk, minced meat (pork), and hen egg, respectively. Non-*ortho* substituted PCBs contributed most to the overall TEQ, with dominant contribution of PCB 126 (32.7 to 68.8%), which has the highest WHO_{2005}-TEF within this group. Except for PCB 118 and PCB 169, the other non-*ortho* and mono-*ortho* PCBs contributed less than 1.3% each to the total TEQ. Considering the PCDD and PCDF congeners, TEQ-based ratios of TCDD/Fs, PeCDD/Fs, and HxCDD/Fs were markedly higher than those of HpCDD/Fs and OCDD/Fs. With the exception of minced meat (pork), 2,3,4,7,8-PeCDF represented the most abundant congener of the PCDFs in the evaluated food pools. The congener with the highest toxicity, 2,3,7,8-TCDD, was only detected in hen eggs (7.0%).

Human exposure to PCDD/Fs and DL-PCBs occurs mainly via animal-based foods like dairy products, fish, meat, and eggs (EFSA CONTAM, 2018). In Germany, foodstuffs such as fruits, vegetables, and grain-based products were found to exhibit low levels and to generally contribute to a minor extent to the total body burden (Päpke, 1998; Schwarz, Lindtner, Blume, Heinemeyer, & Schneider, 2014). The low levels in fruits, vegetables and grain-based products were also reflected by our dataset, which is broadly consistent with TDS data from other countries. In the second French TDS, highest PCDD/F and DL-PCB levels were detected in the fish group as well, followed by butter, crustaceans and molluscs, offal, and cheese (Sirot et al., 2012). Results from the UK TDS revealed

Table 1
Mean levels of the sum of PCDD/Fs and DL-PCBs in the main food groups in pg WHO_{2005}-TEQ/g wet weight with, in parenthesis, data of products of animal origin in pg WHO_{2005}-TEQ/g fat.

Main food group	MEAL foods (n)	Mean	SD	Median	Min.	Max.	Ratio of DL-PCBs (%)
Grains and grain-based products	38	0.026	0.021	0.019	0.003	0.103	28
Vegetables and vegetable products	18	0.015	0.008	0.012	0.005	0.037	24
Starchy roots or tubers and products thereof	7	0.024	0.017	0.016	0.009	0.055	11
Legumes, nuts, oilseeds and spices	20	0.037	0.037	0.023	0.003	0.151	20
Fruit and fruit products	8	0.013	0.008	0.013	0.004	0.030	17
Meat and meat products[1] (w/o liver and edible offal)	35 (28)	0.053 (0.258)	0.107 (0.239)	0.025 (0.142)	0.005 (0.064)	0.644 (1.071)	37
Fish, seafood and invertebrates[2]	30	0.903	2.51	0.300	0.013	13.7	77
Milk and dairy products (w/o buttermilk)	23 (22)	0.061 (0.523)	0.054 (0.211)	0.047 (0.465)	0.005 (0.255)	0.206 (1.070)	59
Eggs and egg products	2 (2)	0.026 (0.187)	0.008 (0.006)	0.026 (0.187)	0.020 (0.182)	0.032 (0.191)	30
Sugar, confectionery and water-based sweet desserts	10	0.039	0.020	0.043	0.008	0.074	21
Animal and vegetable fats and oils (animal fats)	8 (2)	0.140 (0.430)	0.121 (0.019)	0.091 (0.430)	0.035 (0.416)	0.388 (0.443)	40
Coffee, cocoa, tea and infusions	7	0.007	0.011	0.001	0.001	0.031	6
Alcoholic beverages	8	0.002	0.001	0.002	0.001	0.003	12
Food products for infants and toddlers	11	0.009	0.006	0.010	0.002	0.022	22
Products for non-standard diets and food imitates	7	0.012	0.013	0.007	0.002	0.036	11
Composite dishes	52	0.021	0.012	0.022	0.006	0.051	39
Seasoning, sauces and condiments	16	0.034	0.039	0.022	0.002	0.150	55
Total/Mean	300 (54)	0.084 (0.350)	0.176 (0.119)	0.040 (0.306)			30

Left censored-data were analysed using the upper bound (UB) scenario. Results below the LOQ were set to the value reported as the LOQ. w/o: without.
[1] includes liver and edible offal (when related to wet weight).
[2] includes fish liver.

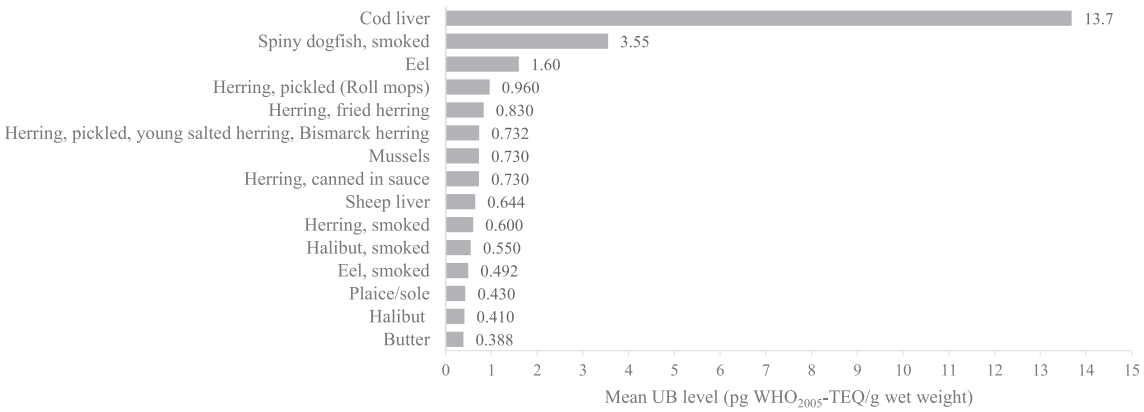

Fig. 1. Mean levels of the sum of PCDD/Fs and DL-PCBs in the 15 MEAL foods exhibiting the highest mean upper bound levels, expressed in pg WHO_{2005}-TEQ/g wet weight.

comparable values with highest levels in decreasing order in the groups 'fish and seafood', 'offal', 'milk and dairy products', and 'fats and oils' (Bramwell, Mortimer, Rose, Fernandes, Harrad, & Pless-Mulloli, 2017). When compared with collected data from 23 European countries published by the EFSA, levels measured in MEAL foods were noticeably lower (EFSA CONTAM, 2018). For instance, the sum of PCDD/Fs and DL-PCBs for 'milk and dairy products' was 1.7-fold and the one for 'eggs and egg products' 7-fold lower. Values for different meats were 1.3- to 5.0-fold lower in the present study. Occurrence data collected by the EFSA were mainly based on monitoring programs with unprepared foodstuffs. Often, TDSs reported lower levels. Manufacturing processes, transport, storage, and preparation steps at home such as washing, peeling and cooking could explain potential losses (Domingo, 2010). Another aspect is the higher analytical sensitivity that can lead to lower LOQs.

In addition, TDSs determine levels of contaminants in pooled samples that per definition result in mean values. Hence, single high values that exceed maximum levels might not be detected in pooled samples.

In the past years, several cases of PCDD/F and DL-PCB levels above the EU maximum values have been discovered. PCDD/Fs and DL-PCBs can enter the food chain via contaminated feed or ingested soil and are subsequently accumulated in lipid rich matrices such as adipose tissue, liver, and muscle-fat and are transferred to milk and eggs. Single exceedances of maximum levels are observed regularly (e.g., in cod liver, meat, sheep liver, hen eggs) (Weber, Herold, Hollert, Kamphues, Blepp, & Ballschmiter, 2018). In the present study, none of the food pools that are regulated by the Commission Regulation (EC) No 1881/2006 (as amended) exceeded the respective maximum levels for PCDD/Fs and DL-PCBs.

Sheep liver had the highest mean PCDD/F and DL-PCB level regarding meat and meat products of terrestrial animals (0.644 pg WHO_{2005}-TEQ/g wet weight). In comparison to livers originating from bovine animals, pigs, and poultry, detected levels in sheep liver were 7-, 16-, and 43-fold higher, respectively. Due to these high levels, frequent consumption of sheep liver might represent a health concern especially for children and women of child-bearing age (EFSA CONTAM, 2011). Expressed on fat basis, meat of wild boar was the animal-based food pool with the highest PCDD/F and DL-PCB level (1.07 pg WHO_{2005}-TEQ/g fat). Wild boars have a relatively high exposure as they are digging for food in the soil which is a main environmental sink for POPs (Weber et al., 2018). Nevertheless, consumption of wild boar meat in Germany does not represent a risk to consumers health (German Federal Institute for Risk Assessment (BfR), 2011).

Similar to literature data, the WHO_{2005}-TEQ of PCB 126 was found to have the highest contribution to the total TEQ (EFSA CONTAM, 2018). In salmon, 2,3,7,8-TCDF and 2,3,4,7,8-PeCDF were the dominating dioxins, corresponding well with earlier findings in fish (Karl, Ruoff, & Blüthgen, 2002). Except of OCDD, highest chlorinated dioxin compounds represented the smallest contribution to the total toxicity, similar to reports in literature (Jiménez, González, & Hernández, 1991).

Fish, seafood and invertebrates

All MEAL foods analysed in the food group 'fish, seafood and invertebrates' are compiled in Table 2. Our sampling covered more than ten different fish species, shrimps, mussels, squid as well as a large variety of fish products and smoked fish, giving a good overview of products consumed in Germany. In all 30 MEAL foods for 'fish, seafood and invertebrates', occurrence data varied considerably ranging from 0.013 to 13.7 pg WHO_{2005}-TEQ/g wet weight. Especially fish rich in fat such as eel (*Anguilla anguilla*), herring (*Clupea harengus*), and halibut (*Hippoglossus hippoglossus*) exhibited higher levels than lean fish species like pollock (*Pollachius virens*) and cod (*Gadus morhua*). Lowest PCDD/F and DL-PCB levels were detected in tuna (*Thunnus*) products, pangas catfish (*Pangasius pangasius*), and fish products from Alaska pollock (*Gadus chalcogrammus*) (fish fingers and gratinated fish filet dish). Within the sub category seafood and invertebrates, the MEAL food mussels exhibited a more than 10-fold higher mean UB level than shrimps/prawns or squid/octopus (see Table 2).

Compared to the second French TDS (Sirot et al., 2012) and the UK TDS (Bramwell et al., 2017), we observed 1.7- and 2.8-fold higher contents in the food group containing fish, respectively. The differences between countries can be partly explained by the composition of the food list for fish pools, due to varying consumption behaviour between countries and different degrees of segmentation into food pools. The French food list did not contain fish with specifically high levels such as cod liver, spiny dogfish, eel or herring products. However, comparing our mean UB levels in specific fish species with data reviewed by the EFSA (EFSA CONTAM, 2018), we observed lower PCDD/F and DL-PCB levels with factors ranging from 1.5 (carp) to 5.8 (eel).

Whereas the muscle meat of the lean fish species cod (*Gadus morhua*) exhibited very low PCDD/F and DL-PCB levels, cod liver represented the MEAL food with the highest content (see Fig. 1 and Table 2). Cod liver is a traditional part of the diet in many coastal areas and rich in vitamin A, vitamin D and ω-3-polyunsaturated fatty acids (Kołakowska, Stypko, Domiszewski, Bienkiewicz, Perkowska, & Witczak, 2002). However, it is also a dietary source for cadmium (Fechner et al., 2022), arsenic (Hackethal, Kopp, Sarvan, Schwerdtle, & Lindtner, 2021), and methylmercury (Sarvan, Kolbaum, Pabel, Buhrke, Greiner, & Lindtner, 2021). According to PCDD/F and DL-PCB levels, cod liver is, in some cases (exceedance of the maximum level), not appropriate for human consumption (Karl & Lahrssen-Wiederholt, 2009). With 13.7 pg WHO_{2005}-TEQ/g wet weight, the level in our food pool was rather low in comparison with EFSA (up to 110 pg WHO_{2005}-TEQ/g wet weight), but comparable with data from north Norway (14.1 pg WHO_{2005}-TEQ/g wet

Table 2
Mean levels of the sum of PCDD/Fs and DL-PCBs, PCDD/Fs, DL-PCBs (pg WHO2005-TEQ/g wet weight) and ratio of DL-PCBs to the total WHO-TEQ as well as fat content of MEAL foods in the main food group 'fish, seafood and invertebrates'.

Sub food group	MEAL food	PCDD/Fs and DL-PCBs	PCDD/Fs	DL-PCBs	Ratio of DL-PCBs (%)	Fat content (%)
Fish products	Cod liver (*Gadus morhua*)	13.7	2.11	11.6	84.7	65.9
	Fish fingers (*Gadus chalcogrammus*)	0.020	0.013	0.007	35.0	14.7
	Fish filet dish, gratinated (*Gadus chalcogrammus*)	0.024	0.012	0.011	45.8	6.7
	Herring (*Clupea harengus*), canned in sauce	0.730	0.370	0.360	49.3	15.4
	Herring (*Clupea harengus*), fried herring	0.830	0.450	0.380	45.8	18.2
	Herring (*Clupea harengus*), pickled, young salted herring, Bismarck herring	0.732	0.390	0.342	46.7	11.6
	Herring (*Clupea harengus*), pickled (Roll mops)	0.960	0.510	0.460	47.9	9.3
	Tuna (*Thunnus*), canned in sauce or own juice	0.013	0.004	0.009	70.0	1.3
	Tuna (*Thunnus*), canned in oil	0.021	0.010	0.011	52.4	11.0
Freshwater fish	Pangas catfish (*Pangasius pangasius*)	0.015	0.011	0.004	24.7	7.3
	Carp (*Cyprinus*)	0.280	0.123	0.156	55.8	15.5
Marine fish	Cod (*Gadus morhua*)	0.048	0.014	0.035	72.9	4.0
	Plaice (*Pleuronectes platessa*)/sole (*Solea solea*)	0.430	0.190	0.240	55.8	9.1
	Halibut (*Hippoglossus hippoglossus*)	0.410	0.140	0.270	65.9	14.1
	Halibut (*Hippoglossus hippoglossus*), smoked	0.550	0.190	0.360	65.5	12.7
	Herring (*Clupea harengus*), smoked	0.600	0.310	0.290	48.3	16.3
	Redfish, ocean perch (*Sebastes norvegicus*)	0.330	0.120	0.210	63.6	7.9
	Pollock (*Pollachius virens*)	0.075	0.032	0.042	56.0	4.5
	Spiny dogfish (*Squalus acanthias*)	3.55	0.473	3.08	86.7	24.1
	Tuna (*Thunnus*)	0.043	0.011	0.032	74.4	1.2
	Tuna (*Thunnus*), smoked	0.015	0.006	0.009	60.7	0.2
Migratory fish	Eel (*Anguilla anguilla*)	1.60	0.231	1.37	85.5	30.4
	Eel (*Anguilla anguilla*), smoked	0.492	0.117	0.376	76.3	23.6
	Salmon (*Salmo salar*)	0.320	0.098	0.220	68.8	14.4
	Salmon (*Salmo salar*), smoked	0.170	0.050	0.120	70.6	7.2
	Trout (*Salmo trutta*, *Oncorhynchus mykiss*)	0.203	0.042	0.158	77.8	10.2
	Trout (*Salmo trutta*, *Oncorhynchus mykiss*), smoked	0.106	0.024	0.083	77.9	4.8
Seafood and invertebrates	Mussels (*Mytilus edulis*, *Pecten spp.*, *Ostera edulis*)	0.730	0.270	0.450	61.6	/
	Shrimps/prawns	0.067	0.040	0.027	40.3	1.7
	Squid (*Loligo vulgaris*)/octopus (*Octopus vulgaris*)	0.042	0.023	0.019	45.2	11.9

Left censored-data were analysed using the upper bound (UB) scenario. Results below the LOQ were set to the value reported as the LOQ.

weight) (Karl, Kammann, Aust, Manthey-Karl, Lüth, & Kanisch, 2016). This can be explained by 70% of the cod liver subsamples in the BfR MEAL Study which originated from Norway.

Herring is one of the most consumed fat fish in Germany and salted and/or marinated herring products are traditionally eaten. Despite different processing techniques, herring samples exhibited similar mean UB levels (0.600–0.960 pg WHO_{2005}-TEQ/g wet weight) (see Table 2). Other studies showed a relation between PCDD/F and DL-PCB levels in herring and the fishing ground, with herring from the Baltic Sea showing the highest levels (EFSA CONTAM, 2018; Fechner, Frantzen, Lindtner, Mathisen, & Lillegaard, 2019; Karl et al., 2002).

The lean fish species plaice (*Pleuronectes platessa*) and sole *(Solea solea*) showed about one order of magnitude higher PCDD/F and DL-PCB levels than other lean fish species. As plaice and sole represent bottom-dwelling fish, this can be explained by their additional uptake of PCDD/Fs and DL-PCBs from the sediments (Knutzen & Oehme, 1989).

In the present study, mean PCDD/F and DL-PCB contents found in pools of mainly farmed fish like salmon and trout were lower compared with those analysed in wild catches like herring (see Table 2). Levels in farmed fish mainly reflect the intake via feed ingredients including fishmeal and fish oil (EFSA CONTAM, 2018). Lundebye et al. (2017) reported similar results comparing farmed salmon with wild Atlantic salmon (higher levels in wild Atlantic salmon) (Lundebye et al., 2017). Consistently, collected data submitted to the EFSA indicated that the sum of PCDD/Fs and DL-PCBs in farmed fish were about 5- to 9-fold lower compared with those of wild caught fish (EFSA CONTAM, 2018).

By comparing smoked and non-smoked fish products, notable differences were observed (see Table 2). Except for halibut, we detected 37 to 69% lower PCDD/F and DL-PCB levels in the smoked variants. Due to water loss during smoking, smoked fish might possess relatively higher fat contents compared with the raw material. Interestingly, we measured higher fat contents in the non-smoked fish. Presumably, this observation can be explained by the fact that the non-smoked food samples were prepared and analysed as consumed. For example, subsamples of trout, salmon or tuna were thermally treated in the BfR study kitchen. These processing techniques, including storing and thawing of deep frozen subsamples, could have led to relevant water losses (Skipnes, Johnsen, Skåra, Sivertsvik, & Lekang, 2011; Xu, Song, Xia, & Jiang, 2019) resulting in elevated final fat contents in the non-smoked samples. Moreover, butter used for cooking could be a factor that might have affected the fat content and PCDD/F and DL-PCB levels of the prepared fish samples. However, a comparison of the same food pool before and after smoking or preparation was not possible within this study design.

Despite the detected levels of PCDD/Fs and DL-PCBs, fish contains a variety of beneficial nutrients (e.g., ω-3-polyunsaturated fatty acids, iodine, selenium, vitamin D) and is recommended to be part of a well-balanced diet (Thomsen et al., 2021).

Type of production

Eighty-seven MEAL foods assigned to 15 main food groups were sampled according to conventional or organic type of production. Differences according to the type of production were not significant between these foods ($p = 0.368$). The largest difference of mean levels was found in 'animal and vegetable fats and oils', mainly driven by the difference between conventional and organic butter. The biggest difference in respect to the factor (factor 1.8) was found in the main food group 'meat and meat products' (see Fig. S1). Nonetheless, distinct differences between individual MEAL foods sampled conventionally produced and organically produced are possible. In single MEAL foods, the highest difference between conventional and organic samples could be observed in the composite dishes meatball and goulash (pork, beef) with 2.7 and 2.3 times higher levels in the organically produced foods, respectively (see Table S1 for raw data per wet weight). Therefore, the data suggest

that the type of production can have an impact on PCDD/F and DL-PCB levels in food. Primary food production, including animal husbandry and food preparation could be considered as possible factors. As PCDD/Fs and DL-PCBs are persistent in the environment, especially free-range animals can accumulate these substances via contaminated feed or ingested soil.

Focussing on foods of animal origin expressed on fat basis (n = 19; excluding buttermilk, edible offal, and livers), a trend towards higher PCDD/F and DL-PCB levels was found in organic compared with conventional foodstuffs (factor 1.5, $p = 0.075$). The largest differences were detected in 'meat and meat products' mainly attributed to the differences in duck and pork meat (see Fig. 2). An exception was chicken meat, displaying a lower level for PCDD/F and DL-PCBs in the organic sample. In the food group 'milk and dairy products', yoghurt (cow milk, plain) showed the biggest difference between conventional and organic type of production with 2.2-fold higher mean values in organic production. However, not all MEAL foods in this food group exhibited the same trend. Organically produced plain cream and quark had lower PCDD/F and DL-PCB levels than the conventional alternatives and plain cow milk food pools showed similar levels comparing both production types. For butter and hen eggs, higher levels were detected in the organically produced foods (1.2-fold and 1.9-fold, respectively).

Organic farming is a fast growing sector that responds to a consumer demand for sustainable food products. The EU is the second largest market of organic foods in the world with Germany representing the largest EU share (Willer & Lernoud, 2019). According to the Regulation (EU) No 2018/848, organic production guarantees food quality and stands for environmental protection and higher animal welfare along the whole food supply chain. This includes strict regulations regarding use of chemical pesticides and fertilisers, housing conditions with a permanent access to open air areas, and mainly organic feed. For livestock with outdoor access, soil is the dominant exposure pathway to environmental pollutants, followed by feed and bedding (Malisch, 2017). The design of organic farms can vary highly and could result in different exposure of the animals to PCDD/Fs and DL-PCBs (Wagner, Brinkmann, Bergschmidt, Renziehausen, & March 2021).

Laying hens and their eggs are prone to take up compounds like PCDD/Fs and DL-PCBs. Eggs from organic and home-raised free-range hens exhibited elevated levels compared with cage and barn eggs (Schoeters & Hoogenboom, 2006). This can mainly be explained by an increased uptake of soil and annelids like earthworms outside the barn. Recent studies have shown that particularly eggs from small flocks repeatedly exceeded the EU maximum level of 5 pg WHO_{2005}-TEQ/g fat. Moreover, researchers assumed that backyard burning of waste is a major contamination source (Hoogenboom et al., 2016). However, eggs and egg products from Germany sampled in this study exhibited about 27 times lower levels than the EU maximum level. They show lower levels than products from Dutch EU monitoring data (2.4-fold) (Adamse, Schoss, Theelen, & Hoogenboom, 2017) or the UK TDS (2.5-fold) (Bramwell et al., 2017), but are comparable with foods of the French TDS (Sirot et al., 2012). If soil is a dominant factor for PCDD/F and DL-PCB exposure in animal-derived foods, free-range farming might be the most relevant factor (higher exposure to soil) rather than the type of production. In our study, it was not possible to differentiate levels of

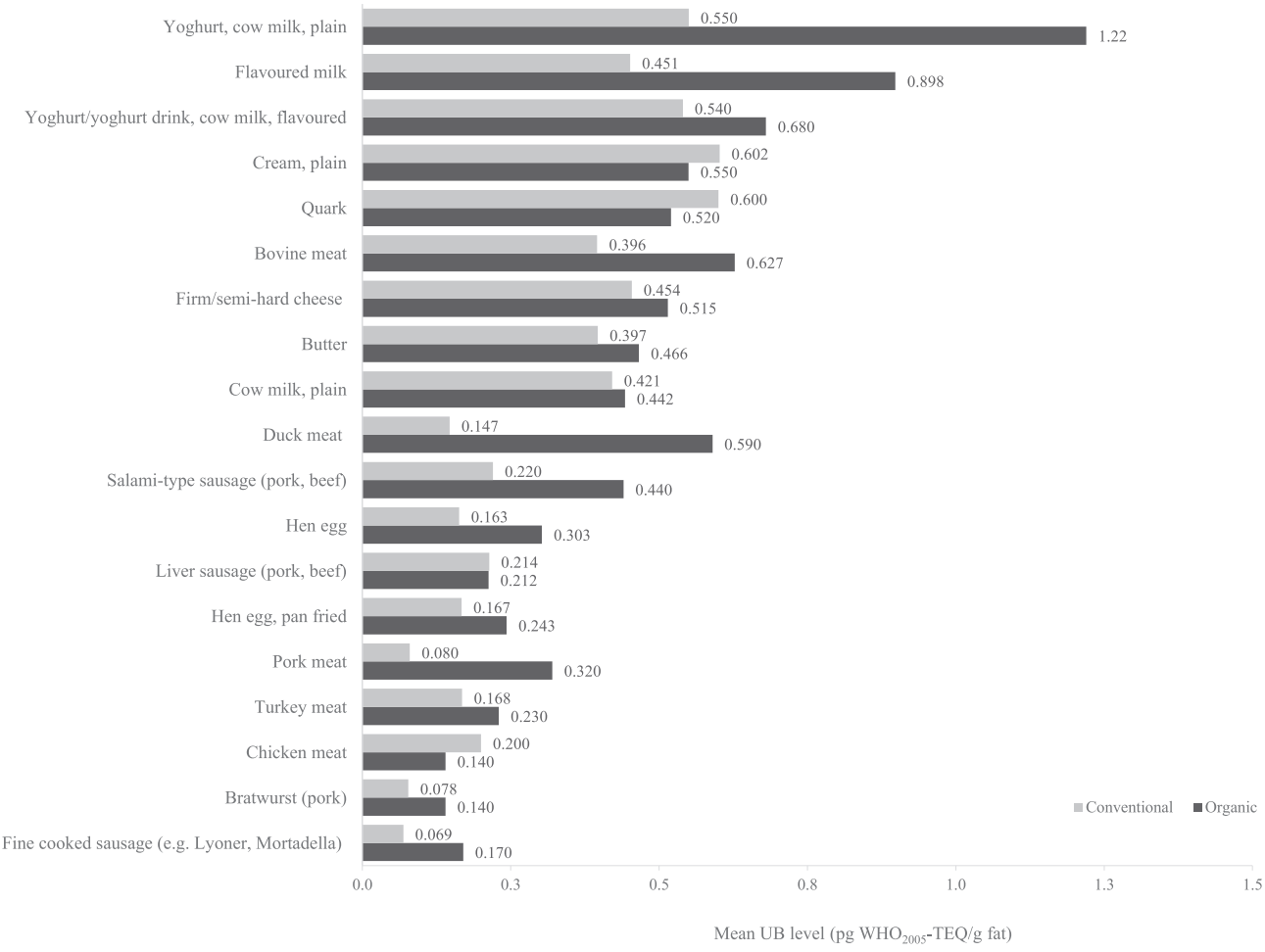

Fig. 2. Mean levels of the sum of PCDD/Fs and DL-PCBs (mean upper bound levels) in MEAL foods differentiated according to type of production, expressed in pg WHO_{2005}-TEQ/g fat.

PCDD/Fs and DL-PCBs between foods derived from free-ranged animals in conventionally or in unspecific food production.

Regionality and seasonality

In four different regions of Germany (east, south, west, and north), 51 MEAL foods assigned to six main food groups were purchased. Overall, statistical analysis revealed no significant difference in levels of PCDD/F and DL-PCBs between the four regions ($p = 0.702$). Nevertheless, distinct differences between individual MEAL foods and main food groups are possible (see Table S4). Slightly higher levels were found in the food groups 'fish, seafood and invertebrates' and 'eggs and egg products' in the region 'east' when compared with the other regions.

Thirty-eight MEAL foods assigned to seven main food groups were tested for seasonal differences. No remarkable differences in the sum of PCDD/F and DL-PCB levels were detected comparing season 1 and season 2. Please find the individual WHO-TEQs in Table S1 and Table S2.

The main reason for occurrence of PCDD/Fs and DL-PCBs in food of animal origin in Germany are contaminated soils, PCB emissions from constructions and buildings, and existing PCB point sources (Weber et al., 2018). Factors influencing the levels of PCDD/Fs and DL-PCBs in soil of different regions are previous use of PCBs, present emissions of PCDD/Fs, the use of chlorinated pesticides with PCDD/Fs as relevant impurities, the historical input of sewage sludge, landfills, open burnings, and waste dumping (Weber et al., 2018). The entry of PCDD/Fs and DL-PCBs into our food via environmental contamination can be dependent on the respective location. Populated or industrial areas often displayed higher PCDD/F and PCB contents than rural areas or grasslands. Moreover, due to their ability to filter atmospheric pollutants, forests function as a sink for PCDD/Fs and PCBs (Rotard, Christmann, & Knoth, 1994). Nonetheless, potential regional differences were not visible in our selected samples.

Limitations and uncertainties of the study

In comparison to other TDSs, the BfR MEAL Study is one of the most comprehensive TDS worldwide with a rather low food aggregation level. Nevertheless, due to pooling, information on levels in single subsamples of MEAL food pools (e.g., brand, preparation method, origin) is not available.

The MEAL food list covers more than 90% of foods consumed in German households representatively. The MEAL food list is based on consumption data from VELS (2002) and the NVS II (2006) covering data from children aged between 6 month up to 4 years and adults from 14 to 80 years, respectively. Hence, consumption habits of children in the age group 5 to 13 years were not considered. The assessment of the consumption behaviour was based on two 3-day dietary records (VELS) or two 24 h-recalls (NVS II) on non-consecutive days. Rarely consumed foods might be underrepresented as well as foods frequently consumed by specific population groups (e.g., hunters, people following a vegetarian or vegan diet). As eating behaviours might have changed during the last years, current food trends might not be considered.

Food sampling in respect to seasonality, regionality, and type of production was performed only when differences were expected. As the composition of the subsamples was dependent on market data and market availability, in some cases the number of subsamples varied between differentiations.

Usage of the LB and UB approach for left-censored data involves the possibility for under- or over-interpretation of actual levels, respectively. The applied statistics have the following limitations typical for the TDS design: (i) it does not consider that each MEAL pool is already representing a mean level of the subsamples with an unknown variability, (ii) it might not be able to detect significant differences if there are significant differences in only some foods (iii) or if the direction of the differences is not the same. Further details could be investigated in the future.

Conclusion

In the scope of the BfR MEAL Study, we established a comprehensive dataset of PCDD/Fs and DL-PCBs in the diet of the population in Germany. Contents in 300 MEAL foods, analysed as typically prepared, provide knowledge for a refined exposure and risk assessment. For the first time in a European TDS, the extensive data set differentiated sampling not only by season and region, but also by type of production resulting in 645 investigated food pools. Highest mean UB levels for the sum of PCDD/Fs and DL-PCBs were found in the main food group 'fish, seafood and invertebrates' with cod liver representing the MEAL food with the highest content. As PCDD/Fs and DL-PCBs bioaccumulate in fatty matrices, animal-based products exhibited higher values than plant-based foods. If regulated by the Commission Regulation (EC) No 1881/2006, all levels of MEAL food pools were below the respective EU maximum level. Sampling by type of production revealed a slight trend towards higher PCDD/F and DL-PCB levels in organically produced foods compared with conventionally produced products. This trend was not consistent for all main food groups. We found no significant differences over all foods sampled by type of production or region.

The first German TDS improves our knowledge about differences in PCDD/F and DL-PCB levels comparing conventional and organic production and delivers insights in levels in accordance to seasonality and regionality.

Representative occurrence data from this study are important for future exposure assessments and recommendations for consumers. This work will substantially contribute to the improvement of future food safety assessments in Germany.

CRediT authorship contribution statement

Mandy Stadion: Formal analysis, Investigation, Visualization, Writing – original draft. **Christin Hackethal:** Methodology, Validation, Writing – review & editing. **Katrin Blume:** Validation, Writing – review & editing. **Birgit Wobst:** Writing – original draft, Writing – review & editing. **Klaus Abraham:** Writing – review & editing. **Carolin Fechner:** Validation, Writing – review & editing. **Oliver Lindtner:** Conceptualization, Funding acquisition, Project administration, Writing – review & editing. **Irmela Sarvan:** Conceptualization, Project administration, Methodology, Writing – review & editing, Supervision.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Data availability

Data will be made available on request.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.fochx.2022.100459.

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