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**Petersen, Annette; Jensen, Bodil Hamborg; Löbl, Nathalie Michelle; Nielsen, Elsa Ebbesen; Bredsdorff, Lea; Fagt, Sisse; Christensen, Tue; Boberg, Julie**

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## The impact of dietary habits on contaminant exposures

**Authors:** Annette Petersen, Bodil Hamborg Jensen, Nathalie Michelle Löbl, Elsa Nielsen, Lea Bredsdorff, Sisse Fagt, Tue Christensen, Julie Boberg

All authors: National Food Institute, Technical University of Denmark, Denmark

Corresponding author: Annette Petersen, [annp@food.dtu.dk](mailto:annp@food.dtu.dk)

### Abstract

This study shows that dietary habits have an impact on contaminant exposures. A tool was developed to calculate chemical exposures for different Danish population groups. First, the tool divided the individuals into quartiles using a previously developed scoring system for how well their diet complies with the Danish dietary guidelines. Then the exposure was calculated for several contaminants for both children and adults within the quartiles. Comparisons were then performed between the exposures for the lowest and highest quartiles. The individuals having a diet more in compliance with the dietary guidelines have a higher exposure to contaminants than individuals having a diet less in compliance with the dietary guidelines. Standard deviations for the mean exposure were in general large indicating that the consumption patterns can be very different within each population group. A risk characterisation for each contaminant and population group was performed by calculation of Hazard Quotients (HQs). For dioxins and dioxin-like polychlorinated biphenyls (PCDD/F+DL-PCBs), inorganic arsenic, and lead all HQs were above 1 indicating a potential risk for all groups. For hexabromocyclododecane (HBCDD) and nickel, a potential risk was identified for at least one group. For all other contaminants the HQs were well below 1 for all groups.

Keywords: contaminant, exposure, dietary habits, risk assessment

## Highlights

- Dietary habits have an impact on contaminant exposures.
- A diet more in compliance with the Danish dietary guidelines results in higher contaminant exposures.
- A potential risk of adverse health effects was identified for dioxins, inorganic arsenic, and lead for all population groups.
- For a few population groups a potential risk was identified for HBCDD and nickel.
- No appreciable risk was identified for the remaining contaminants included in this study.

## 1. Introduction

It is well known that eating a healthy diet may lead to improved quality of life and increased longevity (Knudsen et al., 2012). However, some foods that are considered healthy from a nutritional point of view can have high concentrations of contaminants. For example, some vegetables have high concentrations of nitrate, fatty fish can have high concentrations of dioxins and dioxin-like polychlorinated biphenyls (PCDD/F+DL-PCBs) and methyl mercury (Me-Hg), and

bread from wholemeal flour has higher concentrations of cadmium than bread from white flour (Petersen et al., 2013).

The difference in contaminant exposure of people eating a “healthy” versus an “unhealthy” diet has, however, not been quantified. Such a quantitative analysis can be performed by combining the information on dietary habits of individuals with information on chemical content in foods.

In Denmark, the Danish Veterinary and Food Administration publishes dietary guidelines in an attempt to improve eating habits and thereby the public health of the Danish population. These guidelines are based on the Nordic Nutrition Recommendations (Nordic Council of Ministers 2014) and on literature studies regarding food intake and health (e.g. Knudsen et al., 2012).

Information on how well the adult Danish population (age 18-75 years) complied with the Danish dietary guidelines was collected by Knudsen et al. (2012) who presented a “diet quality index”, i.e. a scoring system where high scores reflect a high degree of compliance with six selected dietary guidelines. This scoring system can be applied to define population groups that comply with the dietary guidelines to different degrees. Each person was awarded with a score according to how well they complied with the criteria for each of the selected dietary guidelines. The selected guidelines, which are also included in the current ten dietary guidelines (Danish Veterinary and Food Administration, 2019) were chosen because it could be quantified whether a person comply with these dietary guidelines or not. Persons with the lowest scores generally had a high intake of red meat, processed meat, and white bread whereas persons with the highest scores had the highest intake of fish, poultry, and rye- and wholemeal bread. In addition, they had a higher consumption of water and wine.

The Danish Veterinary and Food Administration has collected information on certain contaminant levels in Danish foods for several years. The most recent report on contaminant exposure in Denmark covers analyses from the period 2004-2011 (Petersen et al., 2013). These data have been combined with information on food intake from the Danish Dietary Survey 2011-13 (Pedersen et al., 2015) to calculate the contaminant exposures for the Danish population.

The contaminants included in this paper are two mycotoxins, nitrate, some industrial pollutants, process contaminants, as well as some elements.

The two mycotoxins are ochratoxin A (OTA) and deoxynivalenol (DON). OTA is produced by several fungal species of the genera *Penicillium* and *Aspergillus* and occurs in several different food commodities, e.g. cereals. DON is primarily produced by *Fusarium* fungi occurring predominantly in cereal grains.

Nitrate occurs naturally in plants and is of low toxicity; however, it can be reduced to nitrite in food and the gastrointestinal tract and nitrite can form nitrosamines that are considered to be carcinogenic. Nitrate occurs in high amounts in leafy vegetables.

Polycyclic aromatic hydrocarbons (PAHs) are process contaminants formed during e.g. certain cooking practices. In this paper the concentration data and exposure calculations are for the sum of the four PAHs benzo[*a*]pyrene, benzo[*a*]anthracene, chrysene, and benzo[*b*]fluoranthene and is denoted PAH-4.

As industrial pollutants are emitted into the environment they can be found in many foods. The industrial pollutants included in this paper are PCDD/F+DL-PCBs, PCB-6, hexabromocyclododecane (HBCDD), and perfluorooctanyl sulfonate (PFOS). PCB-6 is used as an abbreviation for the sum of six indicator congeners for non-dioxin like PCBs, namely PCB-28, 52, 101, 138, 153 and 180. DDT has previously been used as a pesticide, but is now considered as a pollutant.

The elements included in this paper are arsenic, cadmium, lead, mercury and nickel. They are occurring naturally and some of them are also or has been used in industrial activities. Arsenic can be found in many foods and as different species; in this paper inorganic arsenic is included as being the most toxic arsenic species. Cadmium, lead and nickel are widespread in the environment and can therefore be found in many different foods. Me-Hg, which is the most toxic species of mercury, is formed in the aquatic environment and can therefore be found in e.g. fish.

In this paper, the aim is to investigate the hypothesis that individuals who comply with the dietary guidelines have higher exposures to the above mentioned contaminants than individuals who do not comply with the dietary guidelines. By combining the dietary intake and contaminant databases, it is possible to compare contaminant exposures for individuals having the highest scores of compliance with the dietary guidelines with contaminant exposures for individuals having the lowest scores. Additionally, Hazard Quotients (HQs) are calculated to estimate the risk associated with exposure to each contaminant.

## 2. Methods

### 2.1 Developing a database for comparing chemical exposures of population groups

A quantitative analysis of contaminant exposures in two different population groups was performed by combining information on dietary habits of individuals with information on the contents of contaminants in foods. The population groups were defined as Children (4-14 years of age) and Adults (15-75 years of age). The exposure calculations were performed using an Excel tool that was developed to enable the comparison of contaminant exposure of different age groups participating in the Danish Dietary Survey for 2011-2013 (Pedersen et al., 2015) using concentration data from measurements of contaminants in Danish foods in the period 2004-2011 (Petersen et al., 2013). Additionally, all individuals were ranked according to how well their eating habits complied with the Danish dietary guidelines (Knudsen et al., 2012), and divided into quartiles. Means and 95<sup>th</sup> percentiles for exposure were then calculated within each quartile for both consumer groups.

## 2.2 Scores for the compliance with the dietary guidelines

Table 1 lists the six dietary guidelines used for the scoring of how well individuals complied with the dietary guidelines (Knudsen et al., 2012). These six guidelines were chosen among the ten Danish dietary guidelines because they are quantifiable.

Dietary guideline	Food/nutrient included in the scores for the compliance with dietary guidelines
Eat less fat – particularly fats from meat and dairy products	1. Max 30 energy% total fat 2. Max 10 energy% from saturated fat
Eat potatoes, rice or pasta and wholemeal bread – every day	3. Min 500 g/day
Eat fruit and vegetables every day – 6 portions/pieces per day	4. Min 600 g fruits and vegetables per day <sup>a</sup>
Eat fish and fish products several times a week	5. Min 350 g fish/week
Limit intake of sugar – particularly from soft drinks, confectionary and cakes	6. Max 10 energy% from added sugar <sup>b</sup>

<sup>a</sup>Including up to 100 ml juice per day

<sup>b</sup>Added sugar consists of industrially added sugar and sugar added in cooking or at the table and includes sugar in candy, bakery and soft drinks.

Table 1 Selected food based Dietary Guidelines used for calculating scores for the compliance with the Danish dietary guidelines (from Knudsen et al., 2012).

For each individual, a score (“dietary quality index”) was calculated for each of the six dietary guidelines as the ratio between the actual intake and the recommended intake of the specific nutrient or food (Knudsen et al., 2012). The score for each of the guidelines included ranged from 0 to 1, with 0 assigned to the intake of a food that is furthest away to comply with the dietary guideline, and 1 assigned to the intake complying best with the guideline. For example, if an individual eats 200 g fruits and vegetables per day and the recommendation is to eat 600 g per day, the score is  $200/600 = 0.33$ . When a food or nutrient has an upper limit of recommended intake, the score was calculated as  $1 - [(intake - recommended) / recommended]$ . For an individual with an intake

of 40% energy from total fat where the recommendation is 30%, the score was calculated as  $1 - [40 - 30 / 30] = 0.70$ . For individuals with intakes higher than the cut-off values, or lower for fat and added sugar, the individual was assigned the maximum score of 1. For each individual, the score for each of the six guidelines were summed to a total score with 0 as the minimum score and 6 as the maximum score (Knudsen et al., 2012).

### 2.3. Consumption data

The consumption data were generated based on the Danish Dietary Survey for 2011-2013 and the data were collected as a part of DANSDA (DANish National Survey of Diet and physical Activity) in 2011-2013. The data are a subset of the data reported in “Danskernes kostvaner 2011-13” (Pedersen et al., 2015) and was chosen as they match the period for chemical analysis best and is the most recently published dataset. The dataset covers intake of food and beverages collected among 3946 Danish consumers aged 4 to 75 years. The participants in the survey were drawn as a simple random sample from the civil population registration system and considered representative for the Danish population. The participants in the survey recorded their food intakes for seven consecutive days in a pre-coded (semi-closed) food diary with answering categories for the most commonly consumed foods and dishes in the Danish diet. The food diary was organised according to the typical Danish daily meal pattern. For food items not found in the pre-coded categories it was possible to note type and amount eaten. The amounts of food eaten were given in household measures and estimated from photos of different portion sizes (44 photo series of different foods with 4-6 portion sizes depending on food item). The participants record e.g. how many cups of coffee or glasses of water consumed or how many small or bigger portions they consume of different vegetables. For fruits participants record amounts eaten in pieces of oranges, bananas, apples etc. while other fruits are measured in slices (melons) or handfuls (nuts and dried fruits). The information collected represents information about the current dietary intake in the Danish

population. In the survey each participant self-reported their bodyweight and this is used in the exposure calculations.

#### 2.4. Concentration data

Information on contaminant concentrations in Danish foods was available from analyses of contaminants in food on the Danish market in the period 2004-2011 (Petersen et al., 2013). Data from other sources using other classification systems were not included since consumption and concentration data were linked according to the Danish food classification system (National Food Institute, Technical University of Denmark, 2019).

As mentioned previously the contaminants include OTA, DON, nitrate, industrial pollutants (PFOS, HBCDD, PCDD/F+DL-PCBs, PCB-6, DDT), process contaminants (PAH-4), as well as Me-Hg, lead, inorganic arsenic, cadmium, and nickel.

For OTA and DON information on concentrations in foods are only available for cereals. The concentration in bread is re-calculated from flour based on recipes from the Danish Dietary Survey. For OTA mainly samples of rye flour were analysed together with a few samples of wheat bran, so only rye bread and wheat bran are included in the current exposure calculations. Open rye bread sandwiches are the most typical meal eaten at lunchtime in Denmark. It is well known that OTA can be found in other foods, e.g. raisins and red wine, but no data were available for the current exposure assessment. For DON, several types of cereals, as well as more samples of each cereal type were analysed compared to OTA, e.g. wheat flour and rolled oats. This means that wheat bread and rye bread, as well as prepared foods such as biscuits and crispbread are included.

Nitrate was analysed in lettuce, potatoes, spinach, rocket salad, and Chinese cabbage in the period 2004-2011, and only these foods are included in the exposure assessments. Drinking water was not

a part of the analysis program and is therefore not included, even though this can have a great impact on the exposure of nitrate.

Only samples of fish were analysed for HBCDD and the species included were cod, eel, herring, mackerel, plaice, salmon, and trout. The concentration used is the sum of the isomers  $\alpha$ -,  $\beta$ -, and  $\gamma$ -HBCDD.

Samples of meat (beef, pork, and poultry) and fish (cod, flounder, herring, plaice, and trout) were analysed for PFOS, but except for pork only six or less samples per commodity were analysed.

For PCBs, six indicator congeners (PCB-28, 52, 101, 138, 153 and 180) were measured, primarily in fish and seafood and products of animal origin, and the sum was presented as PCB-6 (Petersen et al., 2013).

For PAH-4 there are no Danish concentration data in the food groups dairy products, bread and cereals, and fresh vegetables so results from the EFSA opinion from 2008 (EFSA, 2008) have been used for the exposure calculations.

As mentioned previously Me-Hg is the most toxic mercury species and therefore, exposures were only calculated and shown for Me-Hg. Only Me-Hg in fish and fish products is included in this assessment.

As the most toxic arsenic species is inorganic arsenic exposures were only calculated for this species. For rice and seafood, data on inorganic arsenic were available and used in the exposure calculations (Petersen et al., 2013). All other foods were analysed for total arsenic, but as no

information on species is available it was assumed that the total arsenic is present as inorganic arsenic, which is considered as a worst-case estimate.

## 2.5 Exposure calculations

The contaminant levels in foods on the Danish market were combined with the consumption data from the Danish dietary survey in the developed Excel tool to calculate the exposure to each contaminant for individuals in the Danish population. The exposure was expressed per kg bodyweight (bw) using each individual's self-reported bodyweight. In this way the impact on how much people eat were taken into consideration. Before calculation of the exposure, each individual was ranked according to their score for the compliance with the dietary guidelines.

All the individuals were distributed into four groups with 25% in each group named Q1, Q2, Q3, and Q4, with Q4 having the highest scores of compliance with the dietary guidelines and Q1 having the lowest scores. Quartiles were made separately for children 4-14 years of age (n=189-190 per quartile) and adults 15-75 years of age (n=797 per quartile). These population sizes were considered sufficient for comparison of chemical exposures. The Excel tool includes the option to divide individuals according to gender and age, but this was not done in this paper due to the risk of over-interpretation of results on small populations. For each quartile, the mean and 95<sup>th</sup> percentile of contaminant exposure were calculated.

## 2.6 Risk assessment

Risk assessment for a single compound is generally performed by estimation of an HQ, which is calculated by dividing the exposure estimate with a human health-based reference point, generally a

health-based guidance value (HBGV, e.g. a tolerable daily intake (TDI) or a tolerable weekly intake (TWI)) for that particular compound.

The HQ calculations for the compounds in this paper are based on the HBGVs predominantly established by EFSA as the reference point (see Table 2). The critical effect on which the reference point is based is also listed in Table 2. For those compounds where the HBGV is a TWI, the TWI is divided by a factor of 7 in order to express the reference point on a daily basis.

For some of the compounds, i.e. HBCDD, PAH, inorganic arsenic, and lead an HBGV has not been set by EFSA. For these compounds, EFSA applied the margin of exposure (MoE) approach for the risk characterisation where the MoE is the ratio between the reference point and the estimated dietary exposure. The BMDL (Benchmark dose lower confidence limit) is generally used by EFSA as the reference point for the MoE approach. In this paper, the reference point for the HQ calculations is the BMDL divided by an appropriate factor that is in accordance with the MoEs in the EFSA opinions for the respective compounds. For inorganic arsenic and PAH-4 that are genotoxic carcinogens the appropriate MoE is 10,000 (EFSA, 2009a, EFSA, 2008). For HBCDD the BMDL<sub>10</sub> is divided by a factor of 8 because the CONTAM Panel concluded that a MoE larger than 8 might indicate that there is no health concern (EFSA, 2011b). For lead, the CONTAM Panel concluded that a MoE of 10 or greater should be sufficient to ensure that there was no appreciable risk for the critical endpoint (EFSA, 2010). For PCB-6, the TDI of 10 ng/kg bw/day set by the French Food Safety Agency (AFSSA) for the congeners most commonly found in food matrices is used for the risk characterisation (AFSSA, 2007).

Contaminant	Reference point (Critical effect)	Type (Value if not TDI or ADI)	Reference
OTA	17 ng/kg bw/d (kidney toxicity)	TWI (120 ng/kg bw/week)	EFSA, 2006
DON	1.0 µg/kg bw/d (reduced body weight gain)	TDI	EFSA, 2017b

PFOS	1.9 ng/kg bw/d (increased serum cholesterol as a risk factor for cardio-vascular disease)	TWI (13 ng/kg bw/week)	EFSA 2018b
HBCDD	0.38 µg/kg bw/d (neurodevelopmental effects)	BMDL <sub>10</sub> (3 µg/kg bw/d) /8	EFSA, 2011b
PAH-4	0.034 µg/kg bw/d (carcinogenicity mediated via a genotoxic mode of action)	BMDL <sub>10</sub> (0.34 mg/kg bw/d) /10,000	EFSA, 2008
PCDD/F+DL-PCBs	0.29 pg/kg bw/d (effects on semen quality)	TWI (2 pg/kg bw/week)	EFSA , 2018a
PCB-6	10 ng/kg bw/d (neuro-behavioural effects in children)	TDI	AFSSA, 2007
DDT	0.33 µg/kg bw/d (liver toxicity)	TDI	Beltoft et al., 2000
Nitrate	3.7 mg/kg bw/d (to protect for toxicity of nitrite)	ADI (as food additive)	EFSA, 2017a
Cadmium	0.36 µg Cd/kg bw/d (kidney toxicity)	TWI (2.5 µg Cd/kg bw/week)	EFSA, 2009b
Inorganic arsenic	0.03 ng As/kg bw/d (carcinogenicity mediated via a genotoxic mode of action)	BMDL <sub>01</sub> (0.3 µg As/kg bw/d) /10,000	EFSA, 2009a
Nickel	2.8 µg Ni/kg bw/d (reproductive and developmental toxicity)	TDI	EFSA, 2015
Me-Hg	0.19 µg Hg/kg bw/d (neurodevelopmental effects)	TWI (1.3 µg Hg/kg bw/week)	EFSA, 2012a
Lead	0.05 µg Pb/kg bw/d (neurodevelopmental effects)	BMDL <sub>01</sub> (0.5 µg Pb/kg bw/d) /10	EFSA, 2010

Table 2. HBGVs and BMDLs used in the calculation of the HQ

### 3. Results and discussions

In brief, the data showed higher contaminant exposures for individuals with high scores for of compliance with the dietary guidelines than for those with low scores, yet variations were large within each population group. In general, children had higher exposures than adults.

#### 3.1 Scores for the compliance with dietary guidelines

Table 3 shows the minimum and maximum scores for Q1, Q2, Q3, and Q4 for children and adults, respectively. The highest score for any individual is 5.0, meaning that no individual in the dietary survey complies with all the dietary guidelines, as the highest achievable score is 6. The lowest value in Q1 is less than 1 for both adults and children indicating that some individuals do not comply with any of the selected dietary guidelines. As shown in Table 3, 50% of the population (Q2 and Q3) fall within a relatively narrow range of scores.

It is especially the guideline on fat and saturated fat that only a few individuals comply with (personal communication senior adviser Sisse Fagt). If less broad groups, e.g. deciles instead of quartiles had been used it is assumed that the differences in scores between the lowest and highest groups would have been larger. On the other hand, the number of individuals in each group would have been lower and this could have resulted in less certain calculations. Using broad groups gives a higher number of individuals in each group, but also gives groups where the individuals have more diverse dietary habits.

	Q1	Q2	Q3	Q4
Children (4-14 years of age), n= 189-190 per quartile; total =758	0.73-2.7	2.7-3.2	3.3-3.7	3.7-5.0
Adults (15-75 years of age), n= 797 per quartile; total = 3188	0.94-2.8	2.8-3.4	3.4-4.0	4.0-5.0

Table 3. Minimum and maximum scores in each quartile for children and adults

### 3.2 Contaminant exposures for children and adults, Q1 and Q4.

Figure 1 and 2 show the calculated exposures for the contaminants included in this paper for Adults and Children, respectively. These data are also presented in Table 4, which also includes the 95<sup>th</sup> percentiles.

Substances	Adult, Q1	Adult, Q4	Children, Q1	Children, Q4
OTA (ng/kg bw)	0.23±0.2 (0.60)	0.34±0.2 (0.74)	0.35±0.3 (0.94)	0.79±0.80 (1.9)
DON (µg/kg bw)	0.13±0.06 (0.26)	0.15±0.06 (0.26)	0.27±0.14 (0.50)	0.39±0.17 (0.68)
PFOS (ng/kg bw)	0.27±0.59 (1.1)	0.99±0.79 (2.5)	0.20±0.45 (0.77)	1.5±1.6 (4.1)
HBCDD (ng/kg bw)	0.10±0.20 (0.50)	0.62±0.61 (1.7)	0.06±0.22 (0.33)	0.62±0.61 (2.0)
PAH-4 (µg/kg bw)	0.013±0.006 (0.024)	0.017±0.007 (0.030)	0.021±0.010 (0.037)	0.033±0.014 (0.056)
PCDD/F+PCB (pg/kg bw)	0.53±0.32 (1.1)	0.82±0.81 (1.6)	0.73±0.40 (1.4)	1.3±0.85 (3.1)
PCB-6 (ng/kg bw)	1.7±1.2 (4.0)	3.9±4.6 (8.0)	2.2±1.4 (4.7)	5.3±4.0 (13)
DDT (ng/kg bw)	3.1±2.3 (7.4)	7.1±6.8 (17)	4.1±3.0 (9.1)	9.5±7.5 (24)
Nitrate (mg/kg bw)	0.53±0.37 (1.2)	0.75±0.55 (1.7)	0.52±0.41 (1.2)	0.80±0.55 (1.7)
Cadmium (µg/kg bw)	0.13±0.06 (0.23)	0.15±0.06 (0.26)	0.24±0.11 (0.44)	0.34±0.12 (0.53)
Inorganic arsenic (µg/kg bw)	0.086±0.037 (0.152)	0.101±0.036 (0.163)	0.124±0.048 (0.208)	0.180±0.074 (0.293)
Nickel (µg/kg bw)	1.5±0.81 (3.0)	1.9±0.89 (3.4)	2.7±1.7 (5.0)	4.0±1.9 (6.9)
Me-Hg (µg/kg bw)	0.011±0.019 (0.041)	0.041±0.041 (0.11)	0.011±0.020 (0.059)	0.061±0.070 (0.20)
Lead (µg/kg bw)	0.20±0.11 (0.39)	0.23±0.09 (0.38)	0.30±0.22 (0.63)	0.33±0.15 (0.59)

Table 4. Contaminant exposures for children (4-14 years of age) and adults (15-75 years of age). Mean exposure ± standard deviation (95<sup>th</sup> percentile in parentheses). Units are not the same for the individual contaminants.

As expected, children in general have a higher mean contaminant exposure per kg bw than adults (Fig. 1 and 2), probably due to a higher consumption of food per kg bw.

The individuals in Q4, i.e. those that comply best with the dietary guidelines, have a higher mean contaminant exposure than the individuals in Q1. This also reflects the dietary habits described by Knudsen et al. (2012) who found that individuals with high scores of compliance with the dietary guidelines have a higher consumption of fish, fruits, vegetables, and wholemeal bread compared to individuals with low scores. Some of these food items contain high levels of contaminants, e.g. wholemeal flour has higher concentrations of mycotoxins compared to white flour and high

concentrations of Me-Hg in fish (Petersen et al., 2013). However, the standard deviation for exposure to most of the substances is very high (Fig. 1 and 2), indicating a high degree of variation in food consumption, and by that the exposure, within each population group.

The highest relative standard deviations are seen for contaminants such as Me-Hg, PFOS, OTA, and HBCDD where only few food items are included in the exposure calculations. These results indicate that the consumption of the included foods is unevenly distributed within the population groups. Other substances, e.g. lead and cadmium are analysed in many different food items. For these contaminants, the high standard deviations for exposure are not only due to high variations in consumption of specific food items, but may also reflect the many different exposure sources for these contaminants. Within the same population groups, e.g. Q4 children, some individuals could have a high exposure of a contaminant from eating a lot of fruit and vegetables, whereas other individuals could have a high contaminant exposure from beverages or from eating a lot of fish.

For many substances, the 95<sup>th</sup> percentile exposure is more than twice the mean exposure within the same population group (see Table 4). It should also be noted that because the number of individuals is lower than 60 at the 95<sup>th</sup> percentile the results should be interpreted with caution (EFSA, 2011a)

### 3.3 Sources of exposure to individual chemicals

For Me-Hg and HBCDD, only results for fish are included in the exposure calculations; therefore, the large differences shown for both children and adults between Q1 and Q4 are due to differences in consumption of fish (see Fig 1 and 2, Table 4). A large difference between Q1 and Q4 for both children and adults is also observed for PFOS where only contents in fish and meat were included in the exposure calculations. According to Knudsen et al. (2012), adults with high scores of compliance with the dietary guidelines eat more fish than those with low scores. For HBCDD the difference between Q1 and Q4 is larger for children than adults, which indicates that differences in

fish consumption are larger for children than for adults. High standard deviations are seen in all population groups indicating high variation in fish consumption within each group. For all three contaminants, the 95<sup>th</sup> percentile exposures are 4-10 times higher than the mean exposures indicating a very high fish consumption in a small part of the population.

Fish was also the main contributor to the exposure for PCDD/F+DL-PCBs, PCB-6, and DDT, with additional contribution from fats and fat products as well as meat and meat products.

Even though fish contribute to a high degree to the exposure and thereby to the risk of adverse health effects from many of the contaminants, fish consumption has also many positive health effects, for example due to the contents of long chain polyunsaturated fatty acids and micronutrients (Thomsen et al., 2018). A quantitative risk-benefit assessment was performed on the substitution of red and processed meat with fish in the diet of Danish adults (>15 years) (Thomsen et al., 2018). The assessment showed a positive health impact if meat is substituted with fish at the recommended amount of 350 g fish (fatty or mix of fatty and lean fish) per week. The analysis included PCDD/F+DL-PCBs, Me-Hg, and two (n-3) long chain polyunsaturated fatty acids (docosahexaenoic acid and eicosapentaenoic acid).

Lead, cadmium, and inorganic arsenic are present in many foods. There is only a little difference between the mean exposures for Q1 and Q4, for both adults and children (Fig 1 and 2, Table 4). For cadmium, the main contributor to the exposure is cereals and cereal products while for lead and inorganic arsenic the food group “beverages” is the main contributor to the exposure (Petersen et al., 2013). Beverages include drinking water, tea, coffee, wine, soft drinks etc. and it is assumed that the consumption of these beverages is more equally distributed across population groups than for other food groups. This may explain why the difference between Q1 and Q4 with regard to lead and inorganic arsenic exposure is less than the difference between Q1 and Q4 for cadmium.

## Adults

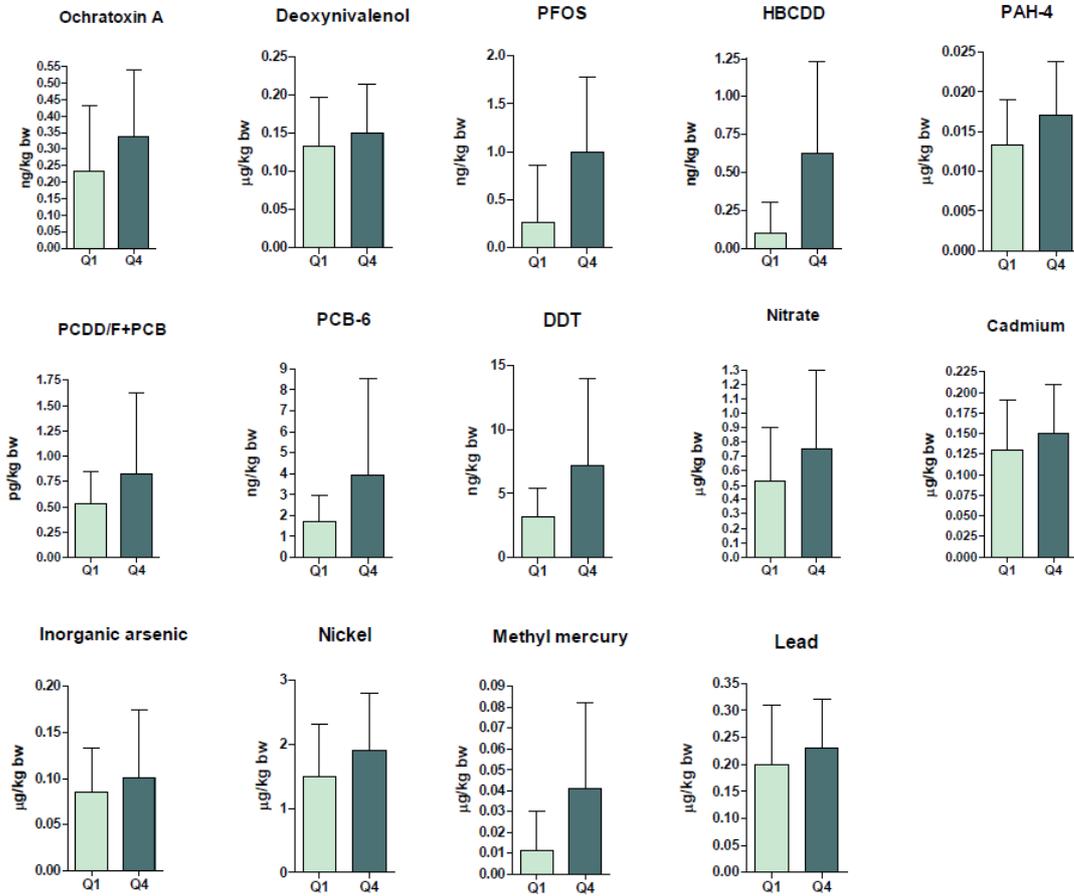


Figure 1. Exposure for Danish adults (age 15-75 years) in Q1 and Q4

## Children

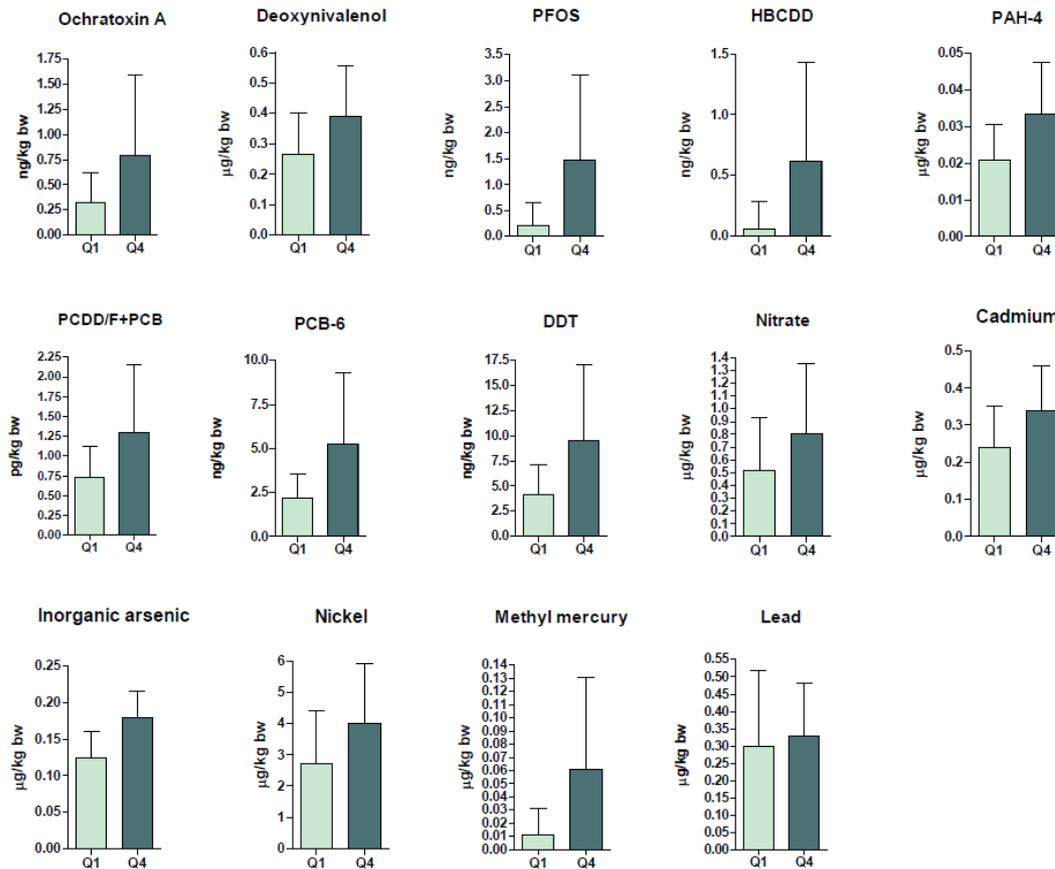


Figure 2. Exposure for Danish children (age 4-14 years) in Q1 and Q4

For mycotoxins, different patterns are seen for OTA and DON. Exposure to OTA is very different between Q1 and Q4 for both adults and children, and this difference is likely due to differences in the consumption of rye bread. For DON the exposure does not differ much between Q1 and Q4, and this may be related to the fact that exposure includes more sources e.g. wheat and rye bread, rolled oats, biscuits, and crispbread.

Potatoes and lettuce are the main exposure source of naturally occurring nitrate (Petersen et al., 2013), and only small differences in exposure are seen both between Q1 and Q4 and between adults and children.

PAHs are produced during food processing, and mainly milk and milk products, cereals and cereal products, and beverages contribute to exposure of the Danish population (Petersen et al., 2013). As for many other substances, children are more exposed than adults are, even though rather high amounts of PAHs can be found in coffee and tea, of which children consume less than adults. Coffee consumption is supposed to be almost the same in Q1 and Q4, and may explain why the exposure is somewhat more similar in Q1 and Q4 for adults compared to children.

### 3.4 Comparison of Hazard Quotients for risk characterisation

HQs were calculated for adults and children, and as expected from the exposure calculations, these were higher for the Q4 populations than for the Q1 populations, for both children and adults (Table 5).

If the exposure to a specific contaminant exceeds the reference point for that contaminant, i.e. the HQ is above 1 a potential risk for adverse health effects is indicated. The HQ was above 1 for all population groups for PCDD/F+DL-PCBs, inorganic arsenic, and lead. This indicates a potential risk of adverse health effects from exposure to these contaminants and is in line with the results in the report for 2004-2011 (Petersen et al., 2013). Especially for inorganic arsenic, the HQs are extremely high ranging between 2900 and 6000. Both in the report from 2013 and in the current paper the same BMDL is used for the risk characterisation (see Table 2). The highest exposures for inorganic arsenic are for cereals (rice) and beverages (Petersen et al., 2013).

For HBCDD and nickel, the HQ was above 1 for at least one population group while for cadmium and PAH-4 the HQ for Q4 children was above 0.9, i.e. very close to 1. For PFOS and PCB-6 the HQ for Q4 children was above 0.5. For OTA and DDT the lowest HQs were below 0.1 for all population groups.

It should be noted that a contaminant with an HQ less than 1 may be of concern when considering combined exposure to several contaminants even if a low risk is identified for that individual contaminant.

Contaminant	HQ for Q1 adults	HQ for Q4 adults	HQ for Q1 children	HQ for Q4 children
OTA	0.014	0.020	0.019	0.046
DON	0.13	0.15	0.27	0.39
PFOS	0.14	0.53	0.11	0.79
HBCDD	0.27	1.7	0.17	1.7
PAH- 4	0.39	0.50	0.61	0.98
PCDD/F+DL-PCBs	1.9	2.9	2.5	4.6
PCB-6	0.17	0.39	0.22	0.53
DDT	0.0094	0.0022	0.012	0.029
Nitrate	0.14	0.20	0.14	0.22
Cadmium	0.36	0.42	0.67	0.95
Inorganic arsenic	2900	3400	4100	6000
Nickel	0.54	0.68	0.96	1.4
Me-Hg	0.050	0.22	0.059	0.32
Lead	4.0	4.6	6.0	6.6

Table 5. Hazard Quotients for persons with low (Q1) or high (Q4) scores for the compliance with the dietary guidelines. Hazard Quotients are calculated as the mean exposure divided by the reference point (see Table 2).

For PCDD/F+DL-PCBs the HQs presented here (Table 5) are higher than previously reported (Petersen et al., 2013), as EFSA in 2018 has set a lower TWI (see Table 2), i.e. this TWI is a factor of seven lower than the former (EFSA, 2018). For PFOS, EFSA has also (in 2018) revised the

HBGV, i.e. instead of the previous TDI of 150 ng/kg bw a TWI of 13 ng/kg bw (corresponding to a TDI about 2 ng/kg bw per day) has been established and therefore, the HQs calculated here are higher than previously reported (Petersen et al., 2013).

The main risks identified here can be compared to findings in a Dutch study (Mengelers et al., 2017), where the exposure to all harmful substances in the diet was analysed including both chemicals and microorganisms. As in the current paper, a Risk Quotient (RQ, identical to the HQ in this paper) was calculated for the general population, 7-69 years of age and for children 2-6 years of age. The highest risk was seen for lead and cadmium with RQs above 1 for all children while for OTA, DON, Me-Hg, and PCDD/F+DL-PCBs the RQs were less than 1 for all children. For the general population no RQs were above 1 for all consumers, but for some consumers the RQ was above 1 for OTA and cadmium. It should be noted that in the Dutch report exposures were not calculated for all the contaminants included in the current paper. As in the current paper, the exposures and thus the RQs were highest for children except for OTA. In both studies, lead is of concern with RQs or HQs above 1. For PCDD/F+DL-PCBs the TWI used in the Dutch report is the former EFSA TWI, which is seven times higher than the revised EFSA TWI used in this paper. Using the revised TWI the RQs for the Dutch population would also be above 1.

Even though it can be concluded that individuals who comply most with the dietary guidelines in general have the highest exposures to contaminants it is still considered healthier to eat according to the dietary guidelines. This is because foods contributing to the contaminant exposure also contain beneficial substances, e.g. essential fatty acids in fish, fibres in wholemeal, vitamins and minerals in fruits and vegetables. Moreover, individuals who comply less with the dietary guidelines eat generally more sugar and fat and are therefore at a higher risk to develop diabetes and obesity, which both are risk factors for cardio-vascular diseases.

The exposure assessments include uncertainties in concentration data as well as in consumption data. Uncertainties in concentration data can be due to e.g. analytical methods or to the number of samples analysed. Uncertainties in the consumption data can be due to e.g. underreporting of the amount of candy eaten or over-reporting of the amount of fruit and vegetables eaten. In this report all the contaminants are analysed in the major food items contributing to the exposure, but not analysed in all foods included in the Danish Dietary Survey. On the other hand, consumption is determined for all foods analysed. These uncertainties can have an impact on the calculated exposures for the individual persons, but it is considered that they have no or little impact on the distributions of the quartiles and no impact on the conclusions in this paper.

#### 4. Conclusion and future perspectives

The results reported in the current paper support the hypothesis that populations with a diet more in compliance with the dietary guidelines have a higher exposure to contaminants than populations with a diet less in compliance with the dietary guidelines. However, the large standard deviations in the exposure calculations in the current paper indicate that the consumption patterns can be very different within each population group.

The main potential health risks were identified for PCDD/F+DL-PCBs, inorganic arsenic, and lead while for HBCDD and nickel, a potential health risk was identified for at least one population group. For all other contaminants, a negligible health risk was identified; however, for cadmium and PAH-4 the HQs for Q4 children was very close to 1 indicating a potential health risk.

The developed database is very useful for exposure calculations for selected population groups. It can be used not only for total exposure calculations, but also for calculations for specific foods or food groups.

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