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RIVM Letter report 2016-0037
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Colophon

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De inname van polybroomdifenylethers in Nederland via voedsel

Polybroomdifenylethers (PBDE's) zijn stoffen die als vlamvertrager in allerlei producten worden gebruikt. Deze stoffen zijn via het milieu in voedsel terechtgekomen, waardoor mensen ze via hun voeding kunnen binnenkrijgen. Uit berekeningen van het RIVM blijkt dat de inname van drie PBDE's (-47, -99 en -153) zodanig laag is dat het risico op schadelijke gezondheidseffecten verwaarloosbaar is. Voor twee andere (PBDE-100 en -183) ontbreken richtwaarden om hier een uitspraak over te kunnen doen.

Aangezien PBDE's oplosbaar zijn in vet, komen ze vooral voor in dierlijke voedselproducten, zoals vis, schelpdieren, melk, eieren, vlees, oliën en vetten. Daarnaast komen ze in plantaardige oliën en vetten voor. In totaal kunnen 209 PBDE's worden gemaakt, maar zijn er acht aantoonbaar aanwezig in het milieu.

Om conclusies te kunnen trekken over effecten op de gezondheid van de inname van PBDE's via voeding, zijn meerdere soorten gegevens nodig. Naast gegevens over de hoeveelheden die mensen van een product eten, betreft dit gegevens over de concentraties van PBDE's in de geconsumeerde producten en gegevens over de zogeheten richtwaarden. Richtwaarden geven aan hoeveel van een stof mensen langdurig binnen kunnen krijgen zonder dat dit op termijn nadelige gevolgen heeft voor de gezondheid. Drie van de acht in het milieu voorkomende PBDE's zijn niet meegenomen in deze studie (PBDE-28, -154, en -209), omdat de beschikbare concentratiegegevens niet bruikbaar of betrouwbaar bleken te zijn; voor PBDE-28 en -154 ontbreken tevens richtwaarden.

Voor de innameberekeningen zijn voedselconsumptiegegevens van de Voedselconsumptiepeiling (VCP) gecombineerd met concentratiegegevens van deze groep stoffen in producten.

Kernwoorden: Polybroomdifenylethers, PBDEs, jonge kinderen, kinderen, volwassenen, langetermijninname, statistisch modelleren

Abstract

Dietary exposure to polybrominated diphenyl ethers in the Netherlands

Polybrominated Diphenyl Ethers (PBDEs) are organobromine compounds that are used as flame retardant in a wide range of products. PBDEs have ended up in food products via environmental contamination, and can therefore be ingested via the diet. Calculations performed by the Dutch National Institute for Public Health and the Environment (RIVM) show that the intake of three PBDEs (-47, -99 and -153) is so low that the risk to health is negligible. No conclusion could be drawn about the health effects of two other PBDEs (-100 and -183) due to the absence of guidance values.

PBDEs are fat-soluble and hence predominantly found in food products of animal origin, including fish, shellfish, milk, eggs, meat, oils and fats. They are also found in vegetable oils and fats. In total, 209 PBDEs can be produced, but in practice only eight are detectable in the environment.

Conclusions about the health effect of dietary PBDE intake can only be drawn when the required data are available. In addition to data on the consumption of food products, information is also needed on PBDE concentrations in consumed food products, and on so-called guidance values. These values quantify the amount of a compound to which a person may be exposed on average over a long period without detrimental consequences for health. Three of the eight PBDEs present in the environment (PBDE-28, -154 and -209) were not considered in this study. The concentration data available for these compounds proved to be unsuitable or unreliable; in addition, no guidance values are available for PBDE-28 and -154.

To calculate the dietary exposure, food consumption data derived from the Dutch National Food Consumption Survey were combined with PBDE concentration data in food products.

Keywords: Polybrominated Diphenyl Ethers, PBDEs, young children, children, adults, long-term exposure, statistical modelling

Contents

1 Introduction

In 2011, the Panel on Contaminants in the Food Chain (CONTAM Panel) of the European Food Safety Authority (EFSA) published a scientific opinion on Polybrominated Diphenyl Ethers (PBDEs)[1] (a group of flame retardants) in food (EFSA, 2011). PBDEs are a class of brominated aromatic compounds with a basic structure consisting of two phenyl rings linked by an ether bond. The position and number of the bromine atoms results in 209 possible compounds, referred to as PBDE congeners. In the CONTAM Panel opinion, the dietary exposure to nine PBDE congeners was reported. Additionally, benchmark doses (BMDs) and their corresponding lower 95% confidence limits for a 10% response ($BMDL_{10}$s) were derived for effects on neurodevelopment. These $BMDL_{10}$s were derived for four PBDE congeners: PBDE-47, -99, -153 and -209 (EFSA, 2011). It was concluded that the dietary exposure to PBDE-47, -153 and -209 did not raise a health concern in the different Member States, whereas for PBDE-99 a potential health concern could not be excluded (EFSA, 2011).

The most recent study into the dietary exposure to PBDE congeners in the Netherlands was based on food consumption of the third Dutch National Food Consumption Survey conducted in 1997/1998 (DNFCS-3) and PBDE congener concentrations in food collected in 2004 and 2008 (Zeilmaker et al., 2014). In this study, the dietary exposure to PBDE-47, -99, -100 and -209 was calculated. It was observed that the exposure to PBDE-47 and -209 via food did not pose a health concern, whereas the exposure to PBDE-99 did. No $BMDL_{10}$ was available for PBDE-100 (EFSA, 2011), making a conclusion about a possible health risk for this congener not possible. In the Dutch study, it was also observed that the PBDE-47, -99 and -100 intakes were higher in 2008 compared to 2004 (Zeilmaker et al., 2014).

An updated intake assessment with recent concentrations and consumption data is desirable given changes in dietary habits over time (Geurts et al., 2013; van Rossum et al., 2011) and possible changes in PBDE congener concentrations in foods. In 2011, food consumption data of a survey conducted in 2007 to 2010 among persons aged 7 to 69 living in the Netherlands (DNFCS 2007-2010) were released (van Rossum et al., 2011). Furthermore, recent PBDE congener concentrations in different products of animal origin are available from two Dutch monitoring programmes, as well as PDBE congener concentrations in different products of vegetable origin which are not covered in both monitoring programmes. All these points allow a revision of earlier performed exposure assessments.

The objective of the current study was to estimate the dietary exposure to five PBDE congeners (PBDE-47, -99, -100, -153 and -183) in the Dutch population aged 7 to 69 using recent information on food

[1] PBDEs are flame retardants which are applied to plastics, textiles, electronic castings and circuitry. PBDEs are ubiquitously present in the environment and likewise in food and feed (EFSA, 2011).

consumption and concentration. To cover as many ages as possible, the dietary exposure in children aged 2 to 6 was also estimated using food consumption data of the DNFCS-Young Children 2005/2006 (Ocké et al., 2008). Other potential sources of exposure (e.g. house dust) were not considered.

2 Intake calculations

2.1 Food consumption data

Exposure calculations for young children were performed using food consumption data of the DNFCS-Young children (Ocké et al., 2008). This survey covers the dietary habits of young children aged 2 to 6 and was conducted in 2005 and 2006. Calculations for the population aged 7 to 69 were performed using food consumption data of the DNFCS 2007-2010 (van Rossum et al., 2011). For a more detailed description of both surveys, see Appendix A.

2.2 Concentration data

Concentration data of the five PBDE congeners (PBDE-47, -99, -100, -153 and -183) in products of animal origin, including cow's milk, eggs and meat (beef, pork, sheep, horse, poultry, deer and rabbit) were obtained from the Dutch monitoring programme on dioxins, dioxin-like polychlorinated biphenyls (PCBs), non-dioxin-like PCBs and flame retardants in primary agricultural products[2]. As part of this programme, samples are taken at farms and slaughterhouses, and analysed on a yearly basis. These samples are reported to the European Union (EU) within the framework of the EU monitoring of background concentrations on dioxins, dioxin-like PCBs, non-dioxin-like PCBs and flame retardants in foodstuffs, and were used in the exposure assessment reported here. Concentration data on wild and farmed fish, and shellfish were obtained from the Dutch monitoring programme on contaminants in Dutch fish and fishery products[3]. Both monitoring programmes are performed on behalf of the Ministry of Economic Affairs. Samples analysed in 2011-2013 were included in the exposure assessment. All these concentrations were stored in the Quality Programme of Agricultural Products (KAP) database[4]. The number of samples varied between products, ranging from one for meat of rabbit and liver to 59 for pork meat and meat of chicken. In general, the products most relevant in view of consumption (e.g. eggs, fish, milk, and meat of cow) were analysed more frequently than those consumed less (e.g. meat of deer, rabbit, and goat, and liver). For the analytical method used to analyse the monitoring samples for PBDE congeners, see Appendix B1.

Additional PBDE congener concentrations in foods were obtained from RIKILT Wageningen UR to cover also possible exposure contributions from products of vegetable origin. These included cereal products, fruits, (low-fat) margarine, pasta and rice as prepared, nuts, potato products, seeds, vegetable oil, vegetables and wine. Samples of these products were obtained from a mycotoxin-dedicated total diet study (mTDS) performed in 2013 (Sprong et al., 2016). In this mTDS study, individual products were collected from Dutch supermarkets, prepared as consumed based on information available in DNFC-Young children and

[2] www.wageningenur.nl/en/Expertise-Services/Research-Institutes/rikilt/Research/Chemical-contamination/Contaminants/Dioxin-analysis/Monitoring-dioxins-PCBs-and-flame-retardants.htm
[3] www.wageningenur.nl/en/project/Monitoring-contaminants-in-Dutch-fish-and-fishery-products.htm
[4] chemkap.rivm.nl

Table 2-1. Overview of concentration data[a] used to assess dietary exposure to the five PBDE congeners considered in this study

Product	Years	Source
Cereal products[b], fruits, (low-fat) margarine, nuts and seeds, prepared pasta, prepared rice, potato products[c], vegetable oils, vegetables and wine	2013	Mycotoxin-dedicated total diet study (Sprong et al., 2016)
Cow's milk, eggs, meat	2011 - 2013	Dutch monitoring programme on dioxins, dioxin-like PCBs, indicator PCBs and flame retardants in primary agricultural products
Fish and shellfish	2011 - 2013	Dutch monitoring programme on contaminants in Dutch fish and fishery products

[a] Concentration below which PBDE congener concentrations were reported as "less than" was a limit of quantification (LOQ).
[b] These products were covered by seven mycotoxin-dedicated total diet study (mTDS) samples: bread (n=2), rye and corn products (n=2), breakfast cereals (n=2), and biscuits and cookies (n=1).
[c] These products were covered by one mTDS sample consisting of weighted amounts of cooked potato, fried potato and potato crisps.

DNFCS 2007-2010, and subsequently pooled to represent a certain product. For example, the sample of product vegetable oils consisted of a weighted pool of different types of vegetable oils based on consumption levels recorded in both DNFCs. The mTDS samples were analysed for the five PBDE congeners using a modified analytical approach (Appendix B2).

The PBDE congener concentrations belonging to cow's milk, meat and eggs were expressed per gram fat, whereas the concentrations in samples belonging to the remaining products were expressed per gram product. Concentrations in processed food products, such as cheese, butter, minced meat, etc., were included in the exposure assessment via a food conversion model, except for (low-fat) margarine, rice and pasta as prepared, and wine (section 2.3).

A short overview of the concentration used to assess the exposure to the five PBDE congeners is presented in Table 2-1. For more details on sample numbers and the concentration data per product and congener, see Appendix C.

2.3 Food mapping

PBDE congeners were analysed in raw products of animal origin, also called raw agricultural commodities (RACs), and in products of vegetable origin at retail level (section 2.2). The products of vegetable origin analysed at retail level included also RACs, except for cereal products, (low-fat) margarine, rice and pasta as prepared, potato products, and wine. Note that vegetable oil was considered a RAC.

Mapping is the process of matching the products analysed to the foods recorded in food consumption databases. For the exposure assessment to the five PBDE congeners two types of food mapping were used:

1. Mapping via RAC
2. Direct mapping between a product analysed and a food recorded in the food consumption database;

The mapping was predominantly performed via RAC, because the majority of analysed products were RACs (Table 2-1). Furthermore, mapping via RACs gives the possibility, via a food conversion model, to include also processed foods containing the RAC as ingredient but which are not analysed in the exposure assessment (section 2.3.1). For four products of vegetable origin, direct mapping was used (section 2.3.2).

2.3.1 Food mapping via RAC

Mapping via RAC was performed for (shell)fish, cow's milk, eggs, meat, fruits, vegetables, nuts and seeds, and vegetable oil. Furthermore, PBDE congener concentrations in cereal products, covering mTDS samples of bread (n=2), rye and corn products (n=2), breakfast cereals (n=2), and biscuits and cookies (n=1), were assigned to cereals at RAC level, including wheat, oat, rye, maize, rice, spelt, millet, buckwheat and barley, and considered as such in the exposure assessment. Congener concentrations in rice at RAC level were mapped to rice consumptions recorded as unprepared and as part of a composite food, such as rice flour, rice crackers and chicken curry, in the DNFCSs. See section 2.3.2 for the mapping to 'rice as prepared' as recorded in the DNFCs. The PBDE congener concentrations analysed in the mTDS sample potato products, covering cooked potato, fried potato and potato crisps, were assigned to potato at RAC level, and considered as such in the exposure assessment.

To model the dietary exposure, the concentrations in RACs need to be converted to concentrations in foods as recorded in the DNFCSs. For this, it is important to realise that foods recorded in food consumption databases include foods consisting of one ingredient (e.g. fruits, vegetables, vegetable oil, full-fat milk and eggs) and composite foods consisting of more than one ingredient (e.g. pizza and salads). Furthermore, since PBDE congeners are lipophilic compounds, the fat content of a food may be important for determining its PBDE congener concentration. This is relevant for the RACs in which the concentrations were reported per gram fat: cow's milk, eggs and meat. For example, congener concentrations will be lower in semi-skimmed milk compared to full-fat milk when reported per gram product.

PBDE congener concentrations in RACs were converted to concentrations in foods as recorded in both food consumption databases as described below.

Consumed foods consisting of one RAC ingredient

Concentrations in RACs were directly assigned to single RAC ingredient foods as recorded in the food consumption databases. This was possible for all analysed RACs, except those analysed per gram fat with a

different fat percentage than the consumed single RAC ingredient food (see below).

For the mapping of fish, consumed and analysed fish species were divided in two groups based on their fat content: lean (<10% fat) and fatty fish (≥10% fat). Consumed fish species were assigned the relevant PBDE congener concentrations at group level. Because PBDE congener concentrations in eel were approximately three times higher than those in other fatty fish species (Appendix C) and since eel is consumed less than these fatty fish species, such as herring or salmon[5], eel was excluded from the fatty fish group, and mapped directly to eel as recorded in the food consumption databases. Crab (legs), mussels, lobster, oyster, squid and shrimps were all assigned a pooled concentration for shellfish. See Appendix D for the individual (shell)fish species belonging to the groups lean fish, fatty fish and shellfish.

Composite foods
To include exposure via the consumption of composite foods in the assessment, a food conversion model was used. In this model, chemical concentrations per RAC are converted to equivalent concentrations in composite foods (Boon et al., 2009b; Geraets et al., 2011; van Dooren, et al., 1995). This model first converts composite foods to their corresponding RAC ingredients (including their weight fractions) based on recipe data and conversion factors of processed ingredients to their raw counterparts. For example, pizza is split first into equivalent amounts of its ingredients like flour, cheese and tomato. These ingredients are subsequently converted to their raw counterparts (wheat, milk and tomato, respectively) using conversion factors. Then, the chemical concentrations analysed in these RAC ingredients are attributed to these fractions and summed to result in the chemical concentration in pizza. This approach was used to assign PBDE congener concentrations to composite foods in the present assessment. For RAC ingredients with a different fat percentage than the analysed RAC, an extra correction was applied (see below).

Mapping corrected for fat percentage
In various cases, the mapping between the analysed RAC and consumed single RAC ingredient food or RAC ingredient of a composite food was corrected for fat percentage. This was relevant for RACs analysed per gram fat. For example, concentrations of PBDE congeners analysed in raw (full-fat) milk were mapped to semi-skimmed or skimmed milk, either consumed as such or as an ingredient of a composite food (e.g. yoghurt, cheese), taking into account their fat percentage. The food conversion model was used for this. This model contains the fat weight fractions per RAC ingredient.

2.3.2 *Direct food mapping*

Direct mapping was used in the present exposure assessment for the products (low-fat) margarine, prepared pasta and rice, and wine. Via direct mapping the analysed products are mapped as much as possible

[5] The mean consumption of eel in DNFCS 2007-2010 is 0.1 g (consumption on 4 out of 7630 consumption days (0.1%)). Corresponding numbers for herring and salmon are 1.6 g (88 consumption days (1.2%)) and 3.0 g (245 consumption days (3.2%)).

to identical foods or to appropriately similar foods recorded in the food consumption database. Mapping via the relevant RACs (vegetable oil, cereals and wine grapes, respectively) was not considered for these products. For (low-fat) margarine, PBDE congener concentrations analysed were lower than those analysed in vegetable oil (Appendix C). Combining these concentrations with those analysed in vegetable oils would thus have resulted in an underestimation of the exposure via the consumption of vegetable oil, but overestimation of the exposure via the consumption of (low-fat) margarine. To avoid this uncertainty, PBDE congener concentrations in (low-fat) margarine were mapped directly to those recorded in both DNFCs. For the samples of prepared pasta and rice, the PBDE concentrations analysed were potentially diluted by use of water in the preparation, and were therefore not included in the concentrations for cereals at RAC level (section 2.3.1), but directly mapped to the recorded consumption of prepared rice and pasta in the DNFCs. Also wine was mapped directly to the consumption of wine, because this could easily be done, and resulted in the most optimal mapping.

2.4 Long-term dietary exposure assessment

The long-term (or usual) dietary exposure to PBDE congeners was assessed, because for consumers repeated exposure to these compounds is most relevant. Repeated exposure may result in an elevated body burden and thus in adverse health effects (EFSA, 2011). To assess the exposure, the Monte Carlo Risk Assessment (MCRA) software, release 8.1 was used (de Boer et al., 2015). This software contains the Observed Individual Means (OIM) model and the LogNormal-Normal (LNN) model, which were both used in the current long-term exposure assessment to PBDE congeners.

In both models, daily consumption patterns of individuals were multiplied with the mean PBDE congener concentration per consumed food, and summed over foods per day per individual and PBDE-congener. All daily estimated exposures were adjusted for individual body weight, resulting in a distribution of mean daily exposures per individual. Exposures were expressed in "ng/kg body weight (bw) per day", and were weighted for small deviances in socio-demographic factors[6] and season[7], and additionally for day of the week for persons aged 7 to 69, to make the results representative for the relevant Dutch population and for all days of the week and all seasons (Ocké et al., 2008; van Rossum et al., 2011).

With OIM, this distribution of mean daily exposures per individual is used as a proxy for the long-term exposure distribution. With LNN, the daily exposure distribution is subsequently corrected for the day-to-day variation in exposure to estimate the long-term exposure distribution. See Appendix E for a description of LNN. The reported percentiles of the long-term exposure distribution were P50, P95 and P99.

[6] Include age, gender, educational level of the head of the household, region and urbanization.
[7] To correct for a higher representation of winter and autumn than spring and summer.

LNN is the preferred model of choice to assess the long-term exposure, since this model corrects for the within-person's variation in intake (Boon and van der Voet, 2016). This approach is known to result in more realistic exposure estimates than when this correction is not performed (Dodd et al., 2006; Hoffman et al., 2002; Slob, 1993). However, the removal of the within-person's variation within LNN can only be performed when the daily positive exposure distribution is normally distributed after transformation (Appendix E). If this condition is not met, the use of LNN to assess the long-term exposure might be debatable. Normality can be checked by using the normal quantile–quantile (q–q) plot, a graphical display of observed vs. theoretical residuals (de Boer et al., 2009). Examination of the q-q plots showed that the daily positive exposure distributions of three PBDE congeners (-99, -153 and -183) in both age groups could be considered close to normal (Appendix F), justifying the use of LNN[8]. However, for PBDE-47 and -100 the fit was not acceptable (Appendix F). For these two congeners, the long-term exposure was therefore estimated using OIM, a model with no model assumptions, but which is known to overestimate the upper tails of the long-term exposure distribution. Furthermore, OIM does not allow the inclusion of covariables in the exposure assessment. Therefore, the exposure in young children was only reported per age when using LNN. For reasons of comparison, also with the use of LNN the exposure was reported for the whole age group of 2 to 6. The exposure estimates in persons aged 7 to 69 were reported independent of age for both models.

In order to evaluate the uncertainty in the dietary exposure assessment due to the sampling size of concentration and food consumption database, the bootstrap approach was used. The uncertainty is reported as the 95% confidence interval around the percentiles of exposure. See Appendix G for a description of the bootstrap.

Samples with congener concentrations below LOQ
A number of samples analysed for PBDE congeners were reported to contain congeners below the limit of quantification (LOQ)[9]. In the intake calculations, we assigned ½LOQ to these samples, the so-called medium bound (MB) scenario. To study the sensitivity of the intake calculations to the concentration assigned to samples with a congener concentration below LOQ, two other scenarios were performed in which either zero (lower bound (LB) scenario) or LOQ (upper bound (UB) scenario) was assigned to these concentrations. Appendix H lists the results of these scenarios.

Effect of processing and migration from packaging material
PBDE congeners are chemically stable lipophilic compounds (EFSA, 2011). Based on a study into the limited information available on the effects of processing on PBDE congener concentrations in food performed by the CONTAM panel, it was concluded that due to processing PBDE congener concentrations may be reduced in processed foods. This possible reduction is mainly caused by loss of fat during

[8] The positive exposure distribution can be considered close to normal when the observed (in red) vs. theoretical residuals (in black) follow approximately a straight line.
[9] The LOQ is the lowest quantity of a substance that can be quantified as a positive concentration.

preparation, rather than degradation (EFSA, 2011). Due to lack of information, this possible effect was not included in the present study resulting possibly in an overestimation of the exposure.

Furthermore, the CONTAM panel noted that PBDE congener concentrations in processed foods may be increased due to migration of these compounds from PBDE containing packaging material. This effect was also not addressed in the current study and may have resulted in an underestimation of the exposure.

Both effects were also not considered in Zeilmaker et al. (2014) and by the CONTAM panel (EFSA, 2011).

2.5 Exposure versus Guidance Values for long-term intake

In 2011, the CONTAM Panel derived benchmark dose lower confidence limits corresponding with a 10% change in response ($BMDL_{10}$, external dose) for neurodevelopmental toxicity[10], i.e. a 10% reduction in response in exposed vs untreated animals, for several PBDE congeners, including PBDE-47, -99 and -153 (EFSA, 2011). These $BMDL_{10}$s equalled 309, 12 and 83 µg/kg bw, respectively. No $BMDL_{10}$s were derived for PBDE-100 and -183.

To use these $BMDL_{10}$s to assess if long-term exposures to PBDE congeners pose a health concern, the $BMDL_{10}$s were translated to Guidance Values (GVs) for long-term intake as described in EFSA (2011) and Zeilmaker et al. (2014). In short, the animal $BMDL_{10}$s were first translated to their corresponding animal body burden (total amount of PBDE congener per kg bw). Based on this animal body burden, and taking into account the half-lives in humans, the daily human intake which leads to this burden in the human body was estimated. According to the CONTAM panel (EFSA, 2011), a Margin of Exposure of 2.5 (or higher) between this human intake value and its corresponding actual intakes does not indicate a potential health concern. Dividing the derived intake value by this factor results therefore in GVs for long-term intake that, when exceeded, are indicative for a potential health concern. This reasoning resulted in the derivation of the following GVs for long-term intake for PBDE-47, -99 and -153: 69, 1.7 and 3.8 ng/kg bw per day, respectively. Apart from this, a GV based on reproductive toxicity[11] has been derived for PBDE-99, namely 0.23 ng/kg bw per day (Bakker et al., 2008).

To assess if there is a possible health risk related to the exposure to the three PBDE congeners for which GVs for long-term intake were available, the P99 of exposures were compared to their GVs as listed above (Boon et al., 2009a). The percentages of persons with an estimated long-term dietary exposure to the PBDE congeners exceeding the GVs were also calculated.

[10] Disturbance of pre- and postnatal growth of the central nervous system
[11] Disturbance of spermatogenesis after intrauterine exposure

3 Results

3.1 Long-term dietary exposure assessment

The median exposure[12] to the five PBDE congeners ranged from 0.02 ng/kg bw per day for PBDE-100 to 0.10 ng/kg bw per day for PBDE-47 and -99 in 2- to 6-year olds, and from 0.01 ng/kg bw per day for PBDE-100 to 0.05 ng/kg bw per day for PBDE-47 and -99 in 7 to 69-year olds (Table 3-1). Corresponding estimates for P99 of exposure[13] were 0.07 ng/kg bw per day for PBDE-153 and 0.43 ng/kg bw per day for PBDE-47, and 0.03 ng/kg bw per day for PBDE-153 and 0.28 ng/kg bw per day for PBDE-47, respectively. Given the uncertainty around the exposure estimates due to the sampling size of the concentration and consumption database (section 2.4), P99 exposure for PBDE-99, -153 and -183 could be as high as 0.22, 0.10, and 0.17 ng/kg bw per day in 2-year olds, and for PBDE-47 and -100 as high as 0.67 and 0.18 ng/kg bw per day in 2- to 6-year olds (Table 3-1).

3.2 Contribution of products

The products that contributed at least 5% to the total long-term exposure distribution to PBDE congeners in 2- to 6-year olds and in persons aged 7 to 69 are presented in Figure 3-1. In both age groups and for all congeners, cow's milk, and fruits and vegetables contributed largely to the exposure (≥ 10%). For PBDE-47 and -100, also fish was an important contributor, for PBDE-99 also potato and cereal products, for PBDE–183 also pork meat, and for PBDE-153 also pork liver.

Examining the upper 5% of the exposure distribution showed that the contribution of fruits and vegetables to the exposure to all PBDE congeners decreased in both age groups. For PBDE-47 and -100, fish became the predominant source of exposure, up to 84% for PBDE-100 in 7- to 69-year olds. For PBDE-153, the contribution of pork liver became largest in both age groups (48% in 2- to 6-year olds and 20% in 7- to 69-year olds), whereas for PBDE-183, cow's milk and pork meat remained the most important contributors, however about 10% higher. In the upper 5% of the exposure distribution of PBDE-99, only slight shifts in contributions were observed. The contribution of potato products was slightly higher (19% in both age groups), as well of as of meat of cow (beef) in 7- to 69-year olds (10%), and pork meat and fish in 2- to 6-year olds (13 and 10%, respectively).

3.3 Exposure versus Guidance Values for long-term intake

The P99 of exposures of PBDE-47, -99 and -153 were compared to the relevant GVs for long-term intake to assess if there is a possible health risk related to the dietary exposure to these compounds. The results show that the percentage of persons with an exposure exceeding the GVs for neurodevelopmental toxicity, as well as for reproductive toxicity for PBDE-99, did not exceed 1% at any age (Table 3-2).

[12] Best (point) estimate of the median exposure within 95% confidence interval (Table 3-1)
[13] Best (point) estimate of the P99 of exposure within 95% confidence interval (Table 3-1)

Table 3-1. Percentiles of long-term dietary exposure to five PBDE congeners in persons aged 2 to 69 in the Netherlands in which samples with PBDE concentrations below limit of quantification (LOQ) equalled ½LOQ (medium bound scenario).

PBDE congener and percentiles of exposure	Age (years) and percentiles of exposure(ng/kg bw per day)						
	2	3	4	5	6	2-6	7-69
PBDE-47							
P50						0.10 (0.09-0.12)	0.05 (0.05-0.06)
P95						0.25 (0.20-0.32)	0.19 (0.14-0.24)
P99						0.43 (0.30-0.67)	0.28 (0.21-0.44)
PBDE-99							
P50	0.12 (0.10-0.13)	0.11 (0.10-0.12)	0.10 (0.09-0.11)	0.10 (0.09-0.11)	0.09 (0.08-0.10)	0.10 (0.10-0.11)	0.05 (0.04-0.05)
P95	0.17 (0.16-0.19)	0.16 (0.14-0.17)	0.15 (0.13-0.16)	0.14 (0.12-0.15)	0.13 (0.12-0.14)	0.15 (0.14-0.17)	0.09 (0.08-0.10)
P99	0.19 (0.18-0.22)	0.18 0.16-0.20)	0.17 (0.15-0.19)	0.16 (0.14-0.18)	0.15 (0.13-0.17)	0.18 (0.16-0.20)	0.12 (0.11-0.13)
PBDE-100							
P50						0.02 (0.02-0.02)	0.01 (0.01-0.01)
P95						0.06 (0.05-0.08)	0.05 (0.04-0.07)
P99						0.11 (0.08-0.18)	0.08 (0.06-0.14)
PBDE-153							
P50	0.04 (0.03-0.05)	0.03 (0.03-0.04)	0.03 (0.02-0.04)	0.03 (0.02-0.03)	0.03 (0.02-0.03)	0.03 (0.02-0.04)	0.02 (0.01-0.02)
P95	0.06 (0.04-0.08)	0.06 (0.04-0.07)	0.05 (0.04-0.06)	0.05 (0.03-0.06)	0.04 0.03-0.06)	0.05 (0.04-0.07)	0.03 (0.02-0.03)

PBDE congener and percentiles of exposure	Age (years) and percentiles of exposure(ng/kg bw per day)						
	2	3	4	5	6	2-6	7-69
P99	0.08 (0.05-0.10)	0.07 (0.04-0.09)	0.06 (0.04-0.08)	0.06 (0.04-0.08)	0.05 (0.04-0.07)	0.07 (0.04-0.09)	0.03 (0.03-0.04)
PBDE-183							
P50	0.05 (0.04-0.09)	0.05 (0.03-0.08)	0.05 (0.03-0.07)	0.04 (0.03-0.07)	0.04 (0.03-0.06)	0.04 (0.03-0.07)	0.02 (0.02-0.03)
P95	0.08 (0.05-0.14)	0.08 (0.05-0.13)	0.07 (0.04-0.11)	0.07 (0.04-0.11)	0.06 (0.04-0.10)	0.08 (0.05-0.12)	0.04 (0.03-0.06)
P99	0.10 (0.06-0.17)	0.09 (0.06-0.16)	0.09 (0.05-0.14)	0.08 (0.05-0.14)	0.08 (0.05-0.13)	0.09 (0.06-0.16)	0.05 (0.04-0.07)

Note: 2.5% lower-97.5% upper confidence limits of the percentiles of exposure are reported between brackets.

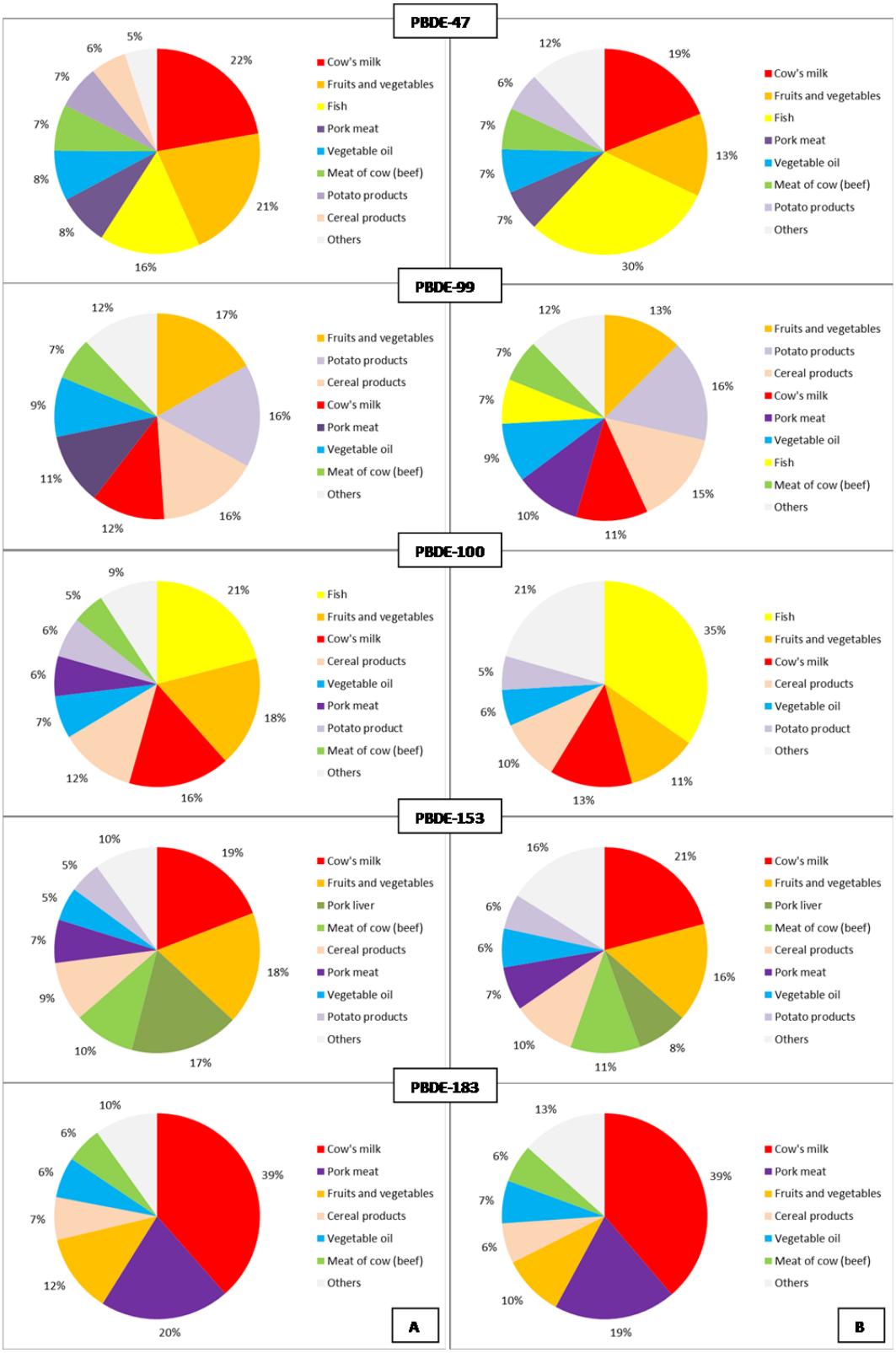

Figure 3-1. Contribution (%) of products contributing at least 5% to the total long-term dietary exposure distribution to five PBDE congeners in children aged 2 to 6 (A) and persons aged 7 to 69 (B) in the Netherlands in which samples with PBDE congener concentrations below limit of quantification (LOQ) equalled ½LOQ (medium bound scenario).

Table 3-2. Percentages of persons aged 2 to 69 in the Netherlands with a long-term dietary exposure to three PBDE congeners above the corresponding guidance value[a] for long-term intake in which samples with congener concentrations below limit of quantification (LOQ) equalled ½LOQ (medium bound scenario).

Age (years)	Percentages exceeding the corresponding guidance value per PBDE			
	PBDE-47	PBDE-99		PBDE-153
Guidance value (ng/kg bw)	69	0.23	1.7	3.8
2		0.10 (0.01-0.51)	0.00 (0.00-0.00)	0.00 (0.00-0.00)
3		0.02 (0.00-0.12)	0.00 (0.00-0.00)	0.00 (0.00-0.00)
4		0.01 (0.00-0.07)	0.00 (0.00-0.00)	0.00 (0.00-0.00)
5		0.00 (0.00-0.06)	0.00 (0.00-0.00)	0.00 (0.00-0.00)
6		0.00 (0.00-0.00)	0.00 (0.00-0.00)	0.00 (0.00-0.00)
2-6	0.00 (0.00-0.00)	0.00 (0.00-0.12)	0.00 (0.00-0.00)	0.00 (0.00-0.00)
7-69	0.00 (0.00-0.00)	0.01 (0.00-0.02)	0.00 (0.00-0.00)	0.00 (0.00-0.00)

Note: 2.5% lower-97.5% upper confidence limits of the percentiles of exposure are reported between brackets.
[a] See for more details, section 2.5.

Given the uncertainty due to the sampling size of the concentration and consumption database (section 2.4), the percentage could be as high as 0.51% in 2-year olds.

4 Discussion

The present study describes the dietary exposure to five PBDE congeners in the Netherlands. Guidance values (GVs) for long-term intake to determine if the long-term exposure to these congeners may pose a health concern were available for three of these compounds. Below, the results are discussed in relation to earlier studies into the dietary exposure to PBDE congeners in the Netherlands, and to the methodology and input data used.

4.1 Comparison with earlier studies into PBDE congener intake

The most recent exposure study of PBDE congeners in the Netherlands was published in 2014 (Zeilmaker et al., 2014). In this study, food consumption data of the DNFCS of 1997-1998 of persons aged 1 to 97 were combined with PBDE congener concentrations analysed in products sampled in 2004 and in 2008. Table 4-1 lists the levels of exposure as reported in this study for the PBDE congeners also addressed in the present study (PBDE-47, -99 and -100) based on the 2008 concentrations. For reasons of comparison, only the results of the persons aged 7 to 69 were reported from the present study. The percentiles of exposure estimated in the present study were lower compared to those reported by Zeilmaker et al. (2014). This may have been partly due to the presence of children aged 1 to 6 in the population included in the earlier Dutch study. However, this very likely does not completely explain the differences in exposure. Other explanation was the higher PBDE congener concentrations in the majority of products used in the earlier study (Appendix I). Other reasons that may explain the differences in exposure were differences in food consumption data and long-term exposure model used, and differences in food mapping.

The CONTAM panel has also published PBDE congener intake estimates based on food consumption data from the Netherlands combined with PBDE congener concentrations derived from products sampled in

Table 4-1. Median (P50) long-term dietary exposure to three PBDE congeners in persons aged 7 to 69 (present study) or 1 to 97 (Zeilmaker et al., 2014) in the Netherlands in which samples with congener concentrations below limit of detection (LOD) (Zeilmaker et al., 2014) or LOQ (present study) equalled ½LOD or ½LOQ (medium bound scenario), respectively

PBDE congener	Percentiles of exposure to PBDE per study (ng/kg bw per day)	
	Present study	Zeilmaker et al.[a]
PBDE-47	0.05 (0.05-0.06))	0.78
PBDE-99	0.12 (0.11-0.13)	0.28
PBDE-100	0.08 (0.06-0.14)	0.13

Note: 2.5% lower-97.5% upper confidence limits of the percentiles of exposure are reported between brackets.
[a] PBDE congener concentrations of 2008

Table 4-2. Dietary exposure to the five PBDE congeners considered in the present study as reported by the CONTAM Panel (EFSA, 2011) and the present study.

Age (in years) and study	Lower and upper bound P95 estimate of dietary exposure (ng/kg bw per day)				
	PBDE-47	PBDE-99	PBDE-100	PBDE-153	PBDE-183
CONTAM Panel					
1-3[a]	4.44-5.09	1.36-2.24	0.76-1.69	0.22-1.87	0.07-1.90
3-6[a]	3.36-3.94	0.96-1.71	0.76-1.43	0.19-1.51	0.06-1.53
Adults[b]	1.54–1.72	0.30-0.58	0.39-0.58	0.07-0.50	0.03-0.46
Present study					
2-6	0.15-0.34	0.11-0.19	0.04-0.08	0.03-0.18	0.03-0.12
7-69	0.14-0.32	0.07-0.11	0.04-0.08	0.01-0.09	0.02-0.06

[a] Based on consumption data of DNFCS-Young children 2005-2006
[b] Based on consumption data of DNFCS Young adults 2003, including persons aged 19 to 32 years (Ocké et al., 2005)

different Member States (EFSA, 2011). Also compared to this study, the exposures reported here were lower (Table 4-2). Earlier studies comparing national estimations of exposure to food contaminants to those reported by EFSA, also showed that national estimations were lower (Boon et al., 2012; Sprong and Boon, 2015). Explanations for this were methodological, such as long-term model used and a more refined food mapping used in the Dutch studies. Also the use of specific, lower, concentrations could explain the observed differences in exposure. For PBDE, these three factors very likely also resulted in lower exposure estimates than reported in EFSA (2011). For example, in the food mapping used by EFSA differences in fat content between consumed and analysed foods such as milk were not addressed, and an examination of the concentrations used by the CONTAM Panel for the different products showed that also the concentrations were higher for all five PBDE congeners compared to those used in the present study.

The present assessment is performed with the most up-to-date information on food consumption and concentrations currently available, using a food conversion model to link the analysed products to processed foods. Additionally, for three PBDE congeners, a statistical model was used to assess long-term exposure. The reported exposures present therefore the current state-of-art regarding dietary exposure to five PBDE congeners in the Netherlands.

4.2 Methodological issues

The exposure estimates of PBDE congeners presented in this report are influenced by different sources of uncertainty. The most important sources are summarized in Table 4-3, including the direction and magnitude of the uncertainty relative to the exposure estimate, using the format as proposed by EFSA (2006). These uncertainties are discussed in more detail below, except those already addressed in section 2.4: model uncertainty, uncertainty regarding the effect of processing and migration of PBDE congeners from packaging material, and the uncertainty related to sampling size of the concentration and food consumption database, which was quantified via a 95% confidence interval around the percentiles of exposure.

Table 4-3. Main sources, direction and magnitude of uncertainty in dietary exposure assessment to PBDE congeners using the medium bound scenario.

Source of uncertainty[a]	Direction & magnitude[b]	Section[c]
Concentrations		
½LOQ[d] to fruit, vegetable, cereal and potato product samples with a PBDE congener concentration < LOQ	+	4.2.2.
Food mapping		
Calculation via RACs[e]	--/++	4.2.4
Mapping cereal and potato product samples via RAC	+	4.2.4
Effect of processing was not considered	•/+	2.4
Migration of PBDE congeners from packaging material	•/-	2.4
Model uncertainty		
OIM (used for PBDE-47 and – 100)[f]	++[9]	2.4
LNN (used for PBDE-99, -153 and 183)[h]	•	2.4
Other sources of exposure		
Not addressing other sources of exposure, such as house dust	•/-	4.2.5
Overall assessment: Based on the qualitative evaluation of different uncertainty sources, it was concluded that the estimated dietary exposure to the five PBDE congeners is likely to be overestimated due to the use of ½LOQ in fruit, vegetable, and cereal and potato product samples with a congener concentration below LOQ, and food mapping of cereal and potato product samples via RAC. Additionally, the high level exposure (P95 and P99) of PBDE-47 and -100 may be further overestimated due to the use of OIM to assess the long-term exposure.	+	

[a] Apart from the listed sources of uncertainty, also the uncertainty due to the sampling size of concentration and food consumption data was quantified via a bootstrap analysis (Appendix G). This uncertainty is quantified as the 95% confidence interval around the percentiles of exposure (section 2.4).
[b] Key to direction and magnitude
+, ++, +++ = uncertainty likely to cause small, medium or large overestimation of exposure
-, --, --- = uncertainty likely to cause small, medium or large underestimation of exposure
• = uncertainty likely to cause a negligible effect on exposure
[c] Section in which the uncertainty source is addressed.
[d] LOQ = limit of quantification
[e] RAC = Raw agricultural commodity
[f] OIM = Observed Individual Mean (section 2.4)
[9] Overestimation of the upper tail of the long-term exposure distribution
[h] LNN = LogNormal-Normal (Appendix E)

4.2.1 Food consumption data

The food consumption data used in this assessment were the most recent food consumption data available for the Netherlands. Due to the use of correction factors for small deviances in socio-demographic factors and season for both populations, and additionally for day of the week for persons aged 7 to 69 the exposure results are regarded to be representative for the Dutch population aged 2 to 69.

4.2.2 Concentration data

PBDE congener concentrations in products of animal origin were the most recent data available in the Netherlands. These samples are

collected on a yearly basis. By using the results that are reported to the EU within the framework of the EU monitoring of background concentrations on dioxins, dioxin-like PCBs, non- dioxin-like PCBs and flame retardants in foodstuffs, it was ensured that the data were non-targeted. Samples of fish and shellfish were also non-targeted and included both farmed and wild types. The results of legs of (North Sea) crab were used in the assessment, since mainly meat present in these parts are consumed.

In the exposure assessment, the PBDE congener concentrations of the three most recent years available (2011-2013) were used to increase the number of concentrations included in the assessment, as well as to address variation in PBDE congener concentrations between years. During these years no trend in lower or higher concentrations were observed (data not shown).

Additional PBDE congener concentrations in product groups of vegetable origin were included in the exposure assessment to avoid possible underestimation of the exposure (Table 2-1). PBDE congeners are lipophilic and are therefore expected to be present in only very low concentrations, if even present, in the analysed mTDS samples of fruits and vegetables. Of the analysed samples, all had a PBDE congener concentration below LOQ, except for PBDE-99 in two vegetable samples. Due to this and to avoid possible underestimation of the exposure, all fruit and vegetable samples were assigned a PBDE-congener concentration equal to ½LOQ in the medium bound (MB) and LOQ in the upper bound (UB) scenario. Given the relative high consumption of these products, this may have resulted in an overestimation of the exposure in both scenarios, as well as in an overestimation of the contribution of fruits and vegetables to the total exposure per PBDE congener as shown in Figure 3-2. In earlier exposure assessments to dioxins and dioxin-like PCBs, also lipophilic compounds, fruits and vegetables were considered to contain no dioxins and dioxin-like PCBs in the MB and UB scenario (Boon et al., 2009a, 2014). This was also true for cereals and potato (Boon et al., 2009a, 2014). In the current assessment, these products were also assumed to contain PBDE congener concentrations in the MB and UB scenario (see also section 4.2.4), which may also have resulted in an overestimation of the exposure and of their contribution to the total exposure per PBDE congener.

4.2.3 Samples with congener concentrations below LOQ

To assess the uncertainty related to the concentrations assigned to PBDE congener concentrations analysed below LOQ, the exposure was also calculated assigning either 0 mg/kg (lower bound (LB) scenario), or LOQ (UB scenario) to all products (section 2.4). The results are presented in Appendix H. Considering the uncertainty around the estimated exposure percentiles, the largest differences between the exposure percentiles of the LB and UB scenario were observed for PBDE-47, -99, and -183. For example, the P99 of exposure in 2- to 6-year olds for PBDE-47 ranged from 0.34 ng/kg bw per day in the LB scenario to 0.53 ng/kg bw per day in the UB scenario (Appendix H). Corresponding figures for PBDE-99 and -183 were 0.11 and 0.19 ng/kg bw per day, and 0.04 and 0.14 ng/kg bw per day, respectively. The main

contributors to this uncertainty were cow's milk for PBDE-47 and -183, and cereal products for PBDE-47 and -99. For these products, a relatively large difference in concentration between the LB and UB scenario were observed for the respective congeners (Appendix C). Together with high consumption levels, this resulted in significant differences in exposure between the LB and UB scenarios for these congeners. Use of an analytical method with a lower LOQ could reduce this uncertainty.

4.2.4 Food mapping

The majority of PBDE congener concentrations used in the exposure assessment was analysed in raw commodities (RACs). To use these concentrations, a food conversion model was used (see section 2.3). Linking concentrations analysed in RACs to consumed amounts of foods with this model has the advantage that processed foods are included in the assessment without the need for analysing them separately. Furthermore, analyses in RACs are performed as part of different monitoring obligations prescribed in legislation and therefore available every year. However, a disadvantage of this approach is that there is no direct link between analysed and consumed foods. As a result, there is always an uncertainty whether the calculated concentrations in foods via the food conversion model are representative for the concentrations in the foods actually consumed. In addition, recipes may change over time. These recipes are presently not updated and may therefore not be representative of the foods currently on the market. Furthermore, in the food conversion model variation in recipes and conversion factors is not addressed. Because of these different factors, the use of the food conversion model can result in over- or underestimation of the exposure. It is not possible to indicate which direction is most likely or whether these uncertainties level out in the final exposure estimate.

Products of vegetable origin (33 samples) were analysed in foods as consumed collected in the mTDS (Sprong et al., 2016). Except for (low-fat) margarine, prepared rice and pasta and wine, PBDE congener concentrations analysed in these products were also mapped via RAC (section 2.3). This approach was obvious for fruits, vegetables, nuts and seeds, and vegetable oil since these can be considered as RACs. For cereal and potato products, this choice was less obvious, and was made from a pragmatic point of view, introducing an additional uncertainty in the exposure assessment. The PBDE congener concentrations analysed in cereal products, especially bread, breakfast cereals, and biscuits and cookies, may have been (partly) derived from the fat present in these products. Assigning these concentrations to cereal at RAC level in the food conversion model, in which the exposure via fat is mapped separately, may thus have resulted in an overestimation of the exposure and the contribution of cereal products to the overall exposure to PBDE congeners. The same applies to the mTDS potato product sample, which was a composite sample of cooked potato, fries and potato crisps. Given the very low exposure levels to PBDE congeners in relation to the relevant GVs (Table 3-2), no attempt was however made to refine the food mapping of these mTDS samples.

4.2.5 *Other sources of exposure*

Apart from via food, people may also be exposed to PBDE congeners via dust, inhalation and dermal contact (Bakker et al., 2008). The CONTAM Panel reviewed the literature regarding these sources of exposure and concluded that dust may be a substantial source of exposure in children (EFSA, 2011). This was particularly true for PBDE-209 and less for the other congeners.

4.2.6 *Summary*

The different sources contributing to the uncertainty of the exposure estimates are summarized in Table 4-3. Overall, the estimated dietary exposure to the five PBDE congeners is likely to be overestimated due to the use of ½LOQ in fruit, vegetable, and cereal and potato product samples with a congener concentration below LOQ, and food mapping of cereal and potato product samples via RAC. Additionally, the high level exposure (P95 and P99) of PBDE-47 and -100 may be further overestimated due to the use of the observed individual mean (OIM) model to assess the long-term exposure.

4.3 Guidance values for long-term intake

The P99 of exposures of PBDE-47, -99 and -100 were compared to GVs for long-term intake to asses if there is a possible health concern related to the exposure. We used the GVs based on the $BMDL_{10}$s derived by the CONTAM Panel based on neurodevelopmental toxicity for all three PBDE congeners (EFSA, 2011) and a GV based on reproductive toxicity for PBDE-99 (Bakker et al., 2008) (section 2.5). No GVs for long-term intake were available for PBDE-153 and -183.

In persons aged 2 to 69, the exposure to none of the PBDE congeners exceeded the GVs for neurodevelopmental toxicity and reproductive toxicity at the P99 of exposure, including the upper confidence limit of the 95% confidence interval (Table 3-2). The percentage of persons exceeding this value ranged from 0.10% in 2-year olds to 0% in persons aged 7 to 69. Given the uncertainty due to sampling size of the food consumption and concentration data, the percentage in 2-year olds could be as high as 0.51%.

Overall, we conclude that health risk related to the intake of PBDE-47, -99 and -100 in the Dutch population is negligible.

4.4 Conclusion

The long-term dietary exposure to PBDE-47, -99 and -100 is so low that the risk to health is negligible in the Netherlands. No conclusion could be drawn about the risk to health of PBDE-153 and PBDE-183 due to a lack of GVs.

In the present study, the exposure to five PBDE congeners was assessed. The CONTAM Panel identified eight PBDE congeners to be of primary concern (EFSA, 2011). Apart from the five congeners considered in this report, these included PBDE-28, -154 and -209. These congeners are also analysed in the two Dutch monitoring programmes. However, the available concentrations were not suitable for a meaningful exposure assessment: PBDE-154 concentrations were

predominantly below LOQ and those of PBDE-28 were only analysed at quantifiable levels in some fish species. The concentrations of PBDE-209 were influenced by background contamination making them unsuitable for exposure assessment purposes. PBDE-209 is the only PBDE congener that was not addressed for which a $BMDL_{10}$ has been established and which may therefore be of toxicological concern (EFSA, 2011). Reliable concentration data of PBDE-209 are needed to determine whether the intake of this congener is of health concern in the Netherlands.

Despite the fact that toxicity studies showed that different PBDE congeners induce a similar type of toxicity (neurodevelopmental toxicity), the present study evaluated the exposure to individual PBDE congeners, i.e. no cumulative exposure by dose-addition was applied. The reason for this is that the latter presumes a common mode of action of the various congeners in inducing toxicity. Though ignoring dose-addition may underestimate toxic risk, such dose addition has yet to be established in proper (animal) experiments.

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Appendix A Description of consumption data used in the exposure assessment to five PBDE congeners

DNFCS-Young Children 2005/2006 (Ocké et al., 2008)
The target population of the DNFCS-Young Children 2005/2006 consisted of boys and girls aged 2 to 6 living in the Netherlands. Respondents were selected from representative consumer panels of Market Research Agency GfK. Panel characteristics, such as socio-demographic characteristics, are known to GfK. Persons in these panels participate in all types of surveys and were not specially selected on nutritional characteristics. Institutionalised persons were excluded, as well as children whose parents/carers did not have sufficient knowledge of the Dutch language. Per family, only one child was included to avoid correlations in dietary consumption patterns between children of the same family. In total, 1,634 children were invited to participate in the study, of which 1,279 consented (net response of 78%). During recruitment, the representativeness of the study population was monitored and, if necessary, the recruitment was adjusted for age and sex, education of the head of the household, level of urbanisation, place of residence and region. The study population was representative regarding socio-demographic characteristics (including region and education of the head of the household), but densely populated areas were slightly underrepresented.

The food consumption data were collected in the period October 2005 to November 2006 via a food diary on two non-consecutive days (separated by about 8 to 13 days). Parents/carers were visited at home by a trained employee of GfK. During the home visit survey materials were presented and overall instructions were given.

Portion size of the foods and meals were estimated by using photographs, domestic measures (a small and a large spoon were supplied to standardise estimates), standard units, weight and/or volume. The usual volume of cups and glasses used was measured by the carer. All days of the week were equally represented, but the winter and autumn period were slightly overrepresented compared to the spring and summer period. National and/or religious holidays or holidays of the participants were not included in the survey.

DNFCS 2007-2010 (van Rossum et al., 2011)
The target population of the DNFCS 2007-2010 consisted of people aged 7 to 69 living in the Netherlands. Pregnant and breast-feeding women, as well as institutionalised people were not included. Respondents were selected from representative consumer panels of GfK. A maximum of one person per household was included in the survey to avoid correlations in dietary consumption patterns between members of the same family. In addition, the panels only included people with sufficient knowledge of the Dutch language. In total, 5,502 individuals were invited to participate in the study, of which 3,819 consented (net response of 69%). Children were overrepresented in the study population and adults underrepresented.

The food consumption data were collected over a 3-year period from March 2007 to April 2010 via two non-consecutive 24-hour dietary recalls (separated by 2 to 6 weeks). Children aged 7 to 15 were interviewed face to face during home visits in the presence of at least one of the child's parents or carers. Participants aged 16 and over were interviewed by telephone, at dates and times unannounced to the participants.

Portion sizes of the foods consumed were quantified in several ways: by means of quantities as shown on photos in a provided picture booklet, or in household measures, standard units, by weight and/or volume. The survey covered all days of the weeks and all four seasons. National and/or religious holidays or holidays of the participants were not included in the survey.

Appendix B Analytical methods used to analyse PBDE congeners

B1. PBDE analysis of the Dutch monitoring samples.

PBDE congeners were analysed in the fat fraction of the samples. Therefore, the fish samples were homogenized via cryogenic grinding and the fat was extracted using Smedes method for samples with an estimated fat content of <5% or Automated Solvent Extraction (ASE) for fat extraction from fatty fish (fat content ≥ 5%). After filtration over anhydrous Na_2SO_4 and drying at 60°C, the fat content was determined quantitatively. Fat from meat samples was obtained by melting out the fat of the product using a microwave, followed by filtering over Na_2SO_4. For the raw cow's milk samples, the fat was isolated by centrifugation and collection of the upper layer (cream), followed by mixing with anhydrous Na_2SO_4 and subsequent extraction of the fat by n-hexane. From the eggs, only the yolks were analysed and the fat was extracted from the yolks by mixing the product with anhydrous Na_2SO_4 and extraction with pentane.

For the analysis of the PBDE congeners, a defined amount of internal standards was added to the fat sample, and the sample was dissolved in 30 ml of hexane. The sample was then purified with a PowerPrep system (FMS). First, the fat moves through an acid silica column to oxidize the fat. Next the eluate is led through a combined silica column, where any remaining fat is removed and the eluate is neutralized. The third column is an alumina column, which is used to remove any interfering components from the eluate. The last column is a carbon column. The eluate from the carbon column, being the cleaned extract, contains the PBDE congeners. The cleaned extract is concentrated to a final volume of 0.5 ml. The recovery standard (PCB^{14}-209) was added to the eluate and 10 µl of the sample extract was introduced into the GC-MS (Trace GC, Thermo Finnigan), equipped with a 30-meter RTX Cl-pesticide capillary column (ID=0.25mm) for separation of the PBDE congeners. The ionisation of these contaminants was carried out at 70 eV via negative chemical ionisation (NCI) using methane as the reaction gas, monitoring m/z 79 and 81. The PBDE congeners concentrations were calculated on the basis of the PCB 198 internal standard and calibration standards.

The PBDE congener concentrations per gram fat in fish samples were converted to concentrations per kg product using the fat content per fish species as determined during the analysis of the PBDE congener concentrations. The concentrations in the other products, including egg, cow's milk and meat, were expressed per gram fat.

[14] polychlorinated biphenyl

B2. PBDE analysis of the total diet study samples.
An amount of sample of 1-13 gram was taken for the analysis of PBDE congeners. Prior to fat extraction, the samples were spiked with a mix of ^{13}C-labeled internal standards (including PBDE-28, -47, -77, -99, -100, -138, -153, -154, -183 and -209). The samples were extracted using 10 mL ethyl acetate. For dried samples, additional Milli-Q water (up to a volume of 13 mL) was added to facilitate the extraction. For fresh samples (i.e. with moisture), no additional water was added. After extraction, a mixture of sodium sulphate (4 g) and sodium chloride (2 g) was added and thoroughly shaken. Phase separation was obtained by centrifugation (10 min, 1500 rpm). The supernatant (ethyl acetate) was collected and blown down to < 1 mL. This was dissolved in 10 mL n-hexane. For lipid-rich samples, such as vegetable oils, a pre-cleaning was performed by dispersive silica clean-up by addition of acidified silica (70-230 mesh, high-purity grade, mixed with concentrated sulphuric acid) and leaving it overnight. The supernatant was collected after centrifugation (10 min, 1500 rpm), and the residual silica was washed again with n-hexane, which was collected again and combined with the 1st supernatant. The pre-cleaned samples and the other samples were cleaned by silica column chromatography (1 gram, 3% deactivated silica and 8 gram acidified silica). The sample extract (in n-hexane) was applied on top of the column, and the PBDE congeners were eluted using 18 ml n-hexane, followed by 12 mL dichloromethane. The sample was concentrated and 50 µl ^{13}C-PCB 209 solution was added as a syringe standard. The final extract was made up to a final volume of 250 µL.

The samples were injected on an Agilent (HP 6890+, Avondale, USA) gas chromatograph (GC) coupled to a Waters (Machaster, UK) Autospec Ultima magnetic sector mass spectrometer (HRMS). The GC was equipped with a PAL autosampler (CTC Analytics AG, Zwingen, Zwitserland) and a CIS 3 PTV injector (Gerstel, Mülheim an der Ruhr, Duitsland). A Rtx-ClPesticides analytical column was used (Restek) of 30 m*0.25 mm i.d. and 0.25 mm, film thickness. The GC was operated at constant flow (helium, 2.0 ml/min). 10 µl of extract was injected using a solvent split flow of 200 ml/min in stop flow mode. The HRMS was operated in Electron Impact (EI) mode at an electron energy of approx. 35 eV. The source temp was 260 °C and the resolution was set at 10000 (+ 10%). Multiplier gain was set at 350 - 400 V. Identification of the PBDE congeners was done at selective ion recording (SIR).

The PBDE congener concentrations were calculated on the basis of the mass labelled internal standards and calibration standards, and expressed per gram product.

Appendix C Total number of samples analysed and the mean PBDE congener concentrations (ng/g product or fat) following three scenarios of assigning concentrations to congener concentrations below limit of quantification (LOQ)

PBDE-47

Product (group)	Nr of samples	Mean concentration[a] (ng/g product or fat)		
		LB scenario	MB scenario	UB scenario
Cow's milk[b] (per gram fat)	28	0.017	0.046	0.075
Cereal products[c] (per gram product)	7	0.0007	0.0016	0.0024
Egg[b] (per gram fat)				
Egg yolk	42	0.02	0.032	0.044
Fish[b] (per gram product)				
Eel	6	0.644	0.644	0.644
Fatty fish	17	0.159	0.163	0.166
Lean fish	28	0.063	0.075	0.087
Shellfish	11	0.058	0.063	0.068
Fruits (per gram product)	9	0	0.0015	0.003
Margarine and low-fat margarine[c] (per gram product)	2	0	0.0005	0.001
Meat[b] (per gram fat)				
Meat of cow (beef)	49	0.034	0.049	0.063
Liver of cow	2	0.018	0.026	0.034
Pork meat	59	0.039	0.05	0.06
Pork liver	2	0.032	0.032	0.032
Meat of sheep	30	0.031	0.04	0.05
Meat of horse	2	0.095	0.098	0.1
Meat of veal (calf)	29	0.052	0.052	0.052
Meat of chicken	59	0.014	0.025	0.036
Meat of other poultry	3	0.132	0.132	0.132
Meat of goat	3	0.032	0.032	0.032
Meat of deer (farmed)	6	0.096	0.096	0.096
Meat of rabbit (tame)	1	0.065	0.065	0.065
Nuts and seeds (per gram product)	3	0	0.0012	0.0023
Pasta, prepared (per gram product)	1	0	0.0015	0.003
Potato products[d] (per gram product)	1	0	0.0025	0.005
Rice, prepared (per gram product)	2	0	0.0025	0.005

Product (group)	Nr of samples	Mean concentration[a] (ng/g product or fat)		
		LB scenario	MB scenario	UB scenario
Vegetable oil (per gram product)	3	0.0105	0.0105	0.0105
Vegetables (per gram product)	12	0	0.002	0.004
Wine (per gram product)	1	0	0.0015	0.003

PBDE-99

Product (group)	Nr of samples	Mean concentration[a] (ng/g product or fat)		
		LB scenario	MB scenario	UB scenario
Cow's milk[b] (per gram fat)	28	0.016	0.021	0.026
Cereal products[c] (per gram product)	7	0.0029	0.0039	0.0048
Egg[b] (per gram product)				
Egg yolk	42	0.059	0.061	0.062
Fish[b] (per gram product)				
Eel	6	0.033	0.039	0.046
Fatty fish	17	0.022	0.028	0.035
Lean fish	28	0.003	0.013	0.024
Shellfish	11	0.012	0.019	0.025
Fruits (per gram product)	9	0	0.001	0.002
Margarine and low-fat margarine (per gram product)	2	0.0033	0.0045	0.0058
Meat[b] (per gram fat)				
Meat of cow (beef)	49	0.039	0.039	0.04
Liver of cow	2	0	0.008	0.015
Pork meat	59	0.062	0.062	0.062
Pork liver	2	0.043	0.043	0.043
Meat of sheep	30	0.104	0.104	0.104
Meat of horse	2	0	0.006	0.012
Meat of veal (calf)	29	0.04	0.04	0.041
Meat of chicken	59	0.039	0.039	0.039
Meat of other poultry	3	0.185	0.185	0.185
Meat of goat	3	0.049	0.049	0.049
Meat of deer (farmed)	6	0.067	0.067	0.067
Meat of rabbit (tame)	1	0.037	0.037	0.037
Nuts and seeds (per gram product)	3	0	0.0022	0.0043
Pasta, prepared (per gram product)	1	0	0.001	0.002
Potato products[d] (per gram product)	1	0.0051	0.0051	0.0051
Rice, prepared (per gram product)	1	0	0.0015	0.003

Product (group)	Nr of samples	Mean concentration[a] (ng/g product or fat)		
		LB scenario	MB scenario	UB scenario
Vegetable oil (per gram product)	3	0.0075	0.008	0.0085
Vegetables (per gram product)	12	0.0103	0.0103	0.0103
Wine (per gram product)	1	0	0.001	0.002

PBDE-100

Product (group)	Nr of Samples	Mean concentration[a] (ng/g product or fat)		
		LB scenario	MB scenario	UB scenario
Cow's milk[b] (per gram fat)	28	0	0.007	0.014
Cereal products[c] (per gram product)	7	0	0.0008	0.0015
Egg[b] (per gram product)				
Egg yolk	42	0.017	0.019	0.021
Fish[b] (per gram product)				
Eel	6	0.244	0.244	0.244
Fatty fish	17	0.046	0.047	0.047
Lean fish	28	0.018	0.019	0.021
Shellfish	11	0.011	0.013	0.015
Fruits (per gram product)	9	0	0.00025	0.0005
Margarine and low-fat margarine (per gram product)	2	0	0.001	0.002
Meat[b] (per gram fat)				
Meat of cow (beef)	49	0.004	0.008	0.012
Liver of cow	2	0	0.005	0.01
Pork meat	59	0.004	0.008	0.012
Pork liver	2	0.01	0.012	0.015
Meat of sheep	30	0.045	0.046	0.046
Meat of horse	2	0	0.005	0.01
Meat of veal (calf)	29	0.002	0.006	0.01
Meat of chicken	59	0.003	0.007	0.011
Meat of other poultry	3	0.031	0.032	0.034
Meat of goat	3	0.013	0.015	0.017
Meat of deer (farmed)	6	0.016	0.018	0.02
Meat of rabbit (tame)	1	0.017	0.017	0.017
Nuts and seeds (per gram product)	3	0	0.0008	0.0017
Pasta, prepared (per gram product)	1	0	0.00025	0.0005
Potato products[d] (per gram product)	1	0	0.0005	0.001
Rice, prepared (per gram product)	1	0	0.0005	0.001

Product (group)	Nr of Samples	Mean concentration[a] (ng/g product or fat)		
		LB scenario	MB scenario	UB scenario
Vegetable oil (per gram product)	3	0	0.002	0.004
Vegetables (per gram product)	12	0.00005	0.0005	0.0009
Wine (per gram product)	1	0	0.00025	0.0005

PBDE-153

Product (group)	Nr of samples	Mean concentration[a] (ng/g product or fat)		
		LB scenario	MB scenario	UB scenario
Cow's milk[b] (per gram fat)	28	0.005	0.011	0.017
Cereal products[c] (per gram product)	7	0	0.00075	0.0015
Egg[b] (per gram product)				
Egg yolk	42	0.023	0.024	0.025
Fish[b] (per gram product)				
Eel	6	0.029	0.029	0.029
Fatty fish	17	0.005	0.005	0.006
Lean fish	28	0.001	0.002	0.002
Shellfish	11	0.005	0.006	0.006
Fruits (per gram product)	9	0	0.0003	0.0007
Margarine and low-fat margarine (per gram product)	2	0	0.001	0.002
Meat[b] (per gram fat)				
Meat of cow (beef)	49	0.016	0.018	0.02
Liver of cow	2	0	0.005	0.01
Pork meat	59	0.009	0.012	0.016
Pork liver	2	1.625	1.628	1.63
Meat of sheep	30	0.08	0.08	0.081
Meat of horse	2	0	0.005	0.01
Meat of veal (calf)	29	0.014	0.016	0.017
Meat of chicken	59	0.004	0.008	0.011
Meat of other poultry	3	0.022	0.024	0.025
Meat of goat	3	0.049	0.049	0.049
Meat of deer (farmed)	6	0.108	0.108	0.108
Meat of rabbit (tame)	1	0	0.005	0.01
Nuts and seeds (per gram product)	3	0	0.0008	0.0017
Pasta, prepared (per gram product)	1	0	0.0005	0.001
Potato products[d] (per gram product)	1	0	0.0005	0.001
Rice, prepared	1	0	0.00025	0.005

Product (group)	Nr of samples	Mean concentration[a] (ng/g product or fat)		
		LB scenario	MB scenario	UB scenario
(per gram product)				
Vegetable oil (per gram product)	3	0	0.002	0.004
Vegetables (per gram product)	12	0	0.00056	0.0011
Wine (per gram product)	1	0	0.00025	0.0005

PBDE-183

Product (group)	Nr of samples	Mean concentration[a] (ng/g product or fat)		
		LB scenario	MB scenario	UB scenario
Cow's milk[b] (per gram fat)	28	0.015	0.03	0.045
Cereal products[c] (per gram product)	7	0	0.00075	0.0015
Egg[b] (per gram product)				
Egg yolk	42	0.01	0.02	0.029
Fish[b] (per gram product)				
Eel	6	0.003	0.005	0.007
Fatty fish	17	0	0.002	0.005
Lean fish	28	0	0.003	0.005
Shellfish	11	0	0.003	0.006
Fruits (per gram product)	9	0	0.0003	0.0007
Margarine and low-fat margarine (per gram product)	2	0	0.001	0.002
Meat[b] (per gram fat)				
Meat of cow (beef)	49	0.005	0.015	0.026
Liver of cow	2	0	0.012	0.025
Pork meat	59	0.033	0.043	0.054
Pork liver	2	0	0.012	0.025
Meat of sheep	30	0.03	0.035	0.04
Meat of horse	2	0	0.012	0.025
Meat of veal (calf)	29	0	0.012	0.023
Meat of chicken	59	0.001	0.013	0.025
Meat of other poultry	4	0	0.012	0.025
Meat of goat	3	0.038	0.042	0.047
Meat of deer (farmed)	6	0.022	0.032	0.042
Meat of rabbit (tame)	1	0	0.005	0.01
Nuts and seeds (per gram product)	3	0	0.0008	0.0017
Pasta, prepared (per gram product)	1	0	0.00025	0.0005
Potato products[d] (per gram product)	1	0	0.0005	0.001

Product (group)	Nr of samples	Mean concentration[a] (ng/g product or fat)		
		LB scenario	MB scenario	UB scenario
Rice, prepared (per gram product)	1	0	0.0005	0.001
Vegetable oil (per gram product)	3	0.0021	0.0035	0.0048
Vegetables (per gram product)	12	0	0.0006	0.0011
Wine (per gram product)	1	0	0.00025	0.0005

[a] LB = lower bound, congener concentrations below LOQ were assigned a concentration of 0 ng/kg fat or product; MB = medium bound, congener concentrations below LOQ equalled ½LOQ; UB = upper bound, congener concentrations below LOQ equalled LOQ.
[b] Each egg sample consisted of 20 eggs; each fish sample contained a mixture of 25 fish species or 1-2 kg in the case of mussel and shrimp; each meat sample consisted of 1 animals; each cow's milk sample was taken from a milk ('Rijdende Melk Ontvangst') truck (a composite sample of 2-3 farms)
[c] These products included the mycotoxin-dedicated total diet study (mTDS) samples bread, rye and corn products, breakfast cereals, and biscuits and cookies (Sprong et al., 2016).
[d] These products were covered by one mTDS sample consisting of weighted amounts of cooked potato, fried potato and potato crisps (Sprong et al., 2016).

Appendix D Individual (shell)fish species assigned to the groups lean fish, fatty fish and shellfish

(Shell)fish species	Group[a]		
	Lean fish	Fatty fish	Shellfish
Bass	A/C		
Cod	A/C		
Crab (leg)			A/C
Dab	A/C		
Eel		A/C	
Flounder	A/C		
Gunard	C		
Haddock	A		
Herring		A/C	
Lobster			C
Mackerel		A/C	
Mussels			A/C
Oyster			C
Pangasius	A/C		
Plaice	A/C		
Pollack	C		
Salmon		A/C	
Sea wolf	C		
Sheatfish	A		
Shrimp			A/C
Sole	A/C		
Sprat		C	
Squid			C
Tilapia	C		
Trout	A/C		
Tuna	C		
Turbot	A		
Whiting	A		

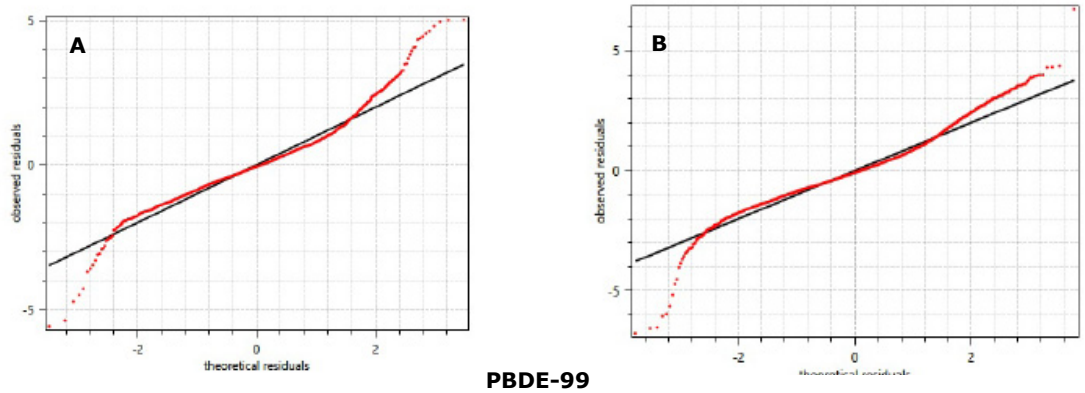
[a] A = analysed, C = recorded in the food consumption databases

Appendix E Modelling of long-term exposure using LNN

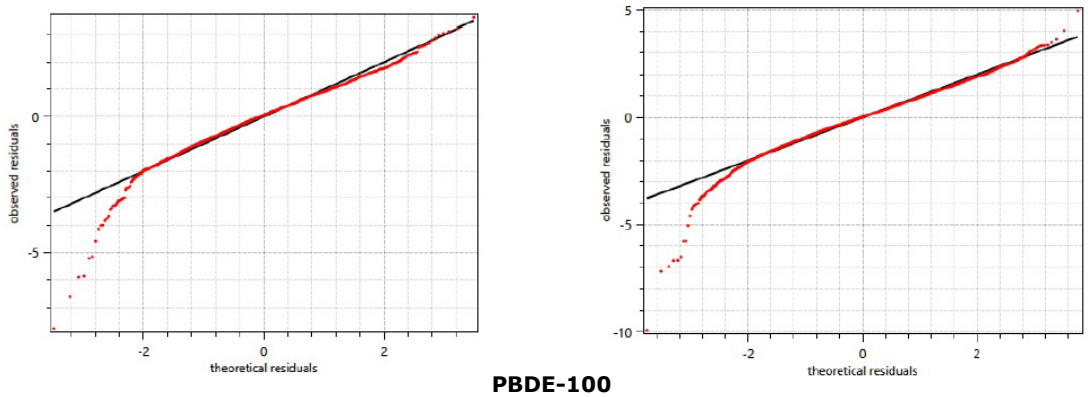
LNN models exposure frequencies and exposure amounts separately, followed by an integration step (Goedhart et al., 2012). For the consumption frequencies, LNN fits a logistic regression model to the number of days with consumption per individual, providing both an estimate of the mean consumption frequency and of the variation between individuals in this frequency (dispersion factor). For the modelling of the positive amounts, LNN first transforms the positive daily exposure distribution into a more normal distribution using a logarithmic or power function. Then, a normal-distribution based variance components model is fitted to remove the within-person's variation. The resulting between-person normal distribution is then back-transformed and combined with the exposure frequency distribution to estimate the long-term dietary exposure distribution. This is achieved by sampling a large number of times from both the exposure frequency distribution and the back-transformed positive exposure distribution (Monte Carlo integration). In this report a logarithmic transformation for the positive daily exposure distribution was used. The correlation between intake frequency and amount was assumed zero.

Appendix F Observed vs. theoretical residuals of the positive daily exposure distributions to five PBDE congeners in children aged 2 to 6 (A) and persons aged 7 to 69 (B) in the Netherlands in which congener concentrations below limit of quantification (LOQ) equalled ½LOQ (medium bound scenario)

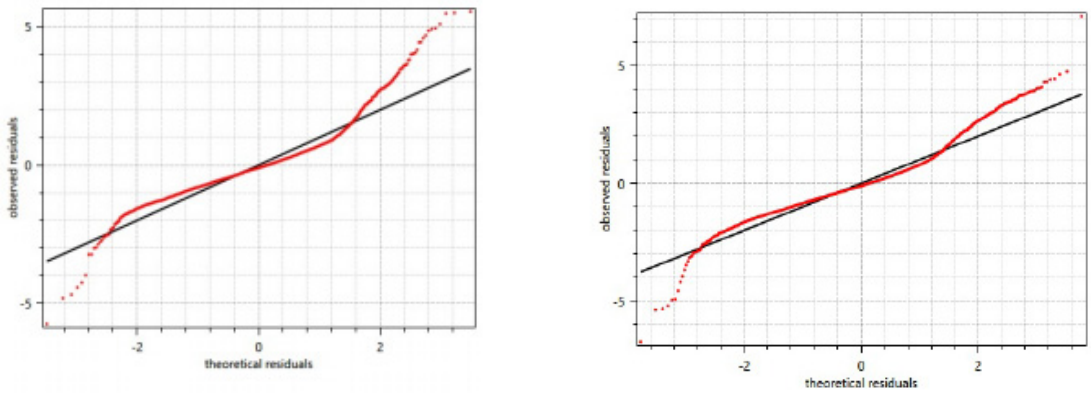
PBDE-47



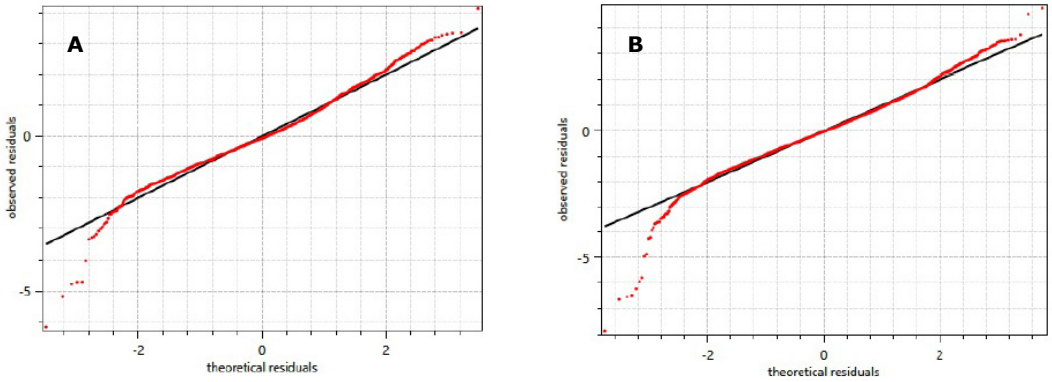
PBDE-99



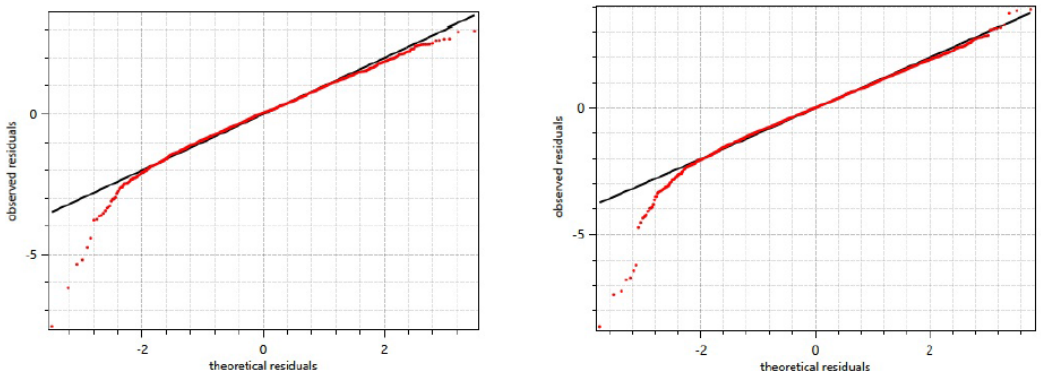
PBDE-100



PBDE-153



PBDE-183



Appendix G Description of the bootstrap

There are different sources of uncertainty in dietary exposure assessments. One of these sources is the uncertainty due to the limited size of the dataset. The smaller the dataset, the more uncertain the data are. This uncertainty can be quantified by using the bootstrap method (Efron, 1979; Efron and Tibshirani, 1993).

With this method a bootstrap database is generated of the same size as the original database for both the food consumption and concentration database by sampling with replacement from the original datasets. These bootstrap databases are considered as databases that could have been obtained from the original population if another sample was randomly drawn. These two bootstrap databases are then used for the exposure calculations and derivation of the relevant percentiles. Repeating this process many times results in a bootstrap distribution for each percentile that allows for the derivation of confidence intervals around it. The bootstrap approach was used in this report by generating 100 food consumption and 100 concentration bootstrap databases and calculating the chronic or acute (with at least 10,000 iterations each) dietary exposure. Of the resulting bootstrap distributions per percentile a 95% uncertainty interval was calculated by computing the 2.5% and 97.5% points of the empirical distribution.

Note that by bootstrapping both the consumption and concentration database in one analysis it is not possible to quantify which part of the uncertainty was due to a limited number of consumption or concentration data.

Appendix H Percentiles of long-term dietary exposure to PBDE congeners in persons aged 2 to 69 in the Netherlands following two scenarios of assigning concentrations to congener concentrations below limit of quantification (LOQ)

Age (years)	Percentiles of exposure (ng /kg bw per day)					
	LB scenario[a]			UB scenario[b]		
	P50	P95	P99	P50	P95	P99
PBDE-47[c]						
2-6	0.04 (0.03-0.05)	0.15 (0.10-0.23)	0.34 (0.21-0.60)	0.17 (0.15-0.19)	0.34 (0.30-0.42)	0.53 (0.43-0.77)
7-69	0.02 (0.02-0.03)	0.14 (0.10-0.18)	0.23 (0.16-0.37)	0.08 (0.07-0.09)	0.23 (0.20-0.27)	0.33 (0.28-0.43)
PBDE-99[d]						
2	0.08 (0.07-0.10)	0.12 (0.11-0.15)	0.15 (0.13-0.17)	0.15 (0.14-0.17)	0.22 (0.20-0.24)	0.25 (0.23-0.28)
3	0.08 (0.07-0.09)	0.12 (0.10-0.14)	0.14 (0.12-0.16)	0.14 (0.13-0.15)	0.20 (0.19-0.22)	0.23 (0.21-0.25)
4	0.08 (0.06-0.09)	0.11 (0.10-0.13)	0.13 (0.11-0.16)	0.13 (0.12-0.14)	0.19 (0.17-0.20)	0.22 (0.20-0.23)
5	0.07 (0.06-0.09)	0.11 (0.09-0.13)	0.12 (0.11-0.15)	0.12 (0.11-0.13)	0.17 (0.16-0.19)	0.20 (0.19-0.22)
6	0.07 (0.06-0.08)	0.10 (0.09-0.12)	0.12 (0.10-0.14)	0.11 (0.10-0.12)	0.16 (0.15-0.17)	0.19 (0.17-0.20)
2-6	0.08 (0.07-0.09)	0.11 (0.10-0.13)	0.13 (0.12-0.16)	0.13 (0.12-0.14)	0.19 (0.18-0.21)	0.23 (0.21-0.25)
7-69	0.04	0.07	0.09	0.07	0.11	0.14

Age (years)	Percentiles of exposure (ng /kg bw per day)					
	LB scenario[a]			UB scenario[b]		
	P50	P95	P99	P50	P95	P99
	(0.03-0.04)	(0.06-0.08)	(0.08-0.10)	(0.06-0.07)	(0.10-0.12)	(0.13-0.15)
PBDE-100[c]						
2-6	0.003 (0.002-0.004)	0.04 (0.02-0.05)	0.09 (0.05-0.13)	0.04 (0.04-0.04)	0.08 (0.07-0.10)	0.14 (0.11-0.22)
7-69	0.002 (0.001-0.002)	0.04 (0.03-0.05)	0.07 (0.05-0.10)	0.02 (0.02-0.02)	0.06 (0.05-0.07)	0.09 (0.07-0.13)
PBDE-153[d]						
2	0.02 (0.008-0.03)	0.04 (0.02-0.06)	0.05 (0.02-0.09)	0.06 (0.05-0.07)	0.09 (0.07-0.12)	0.11 (0.08-0.14)
3	0.02 (0.007-0.02)	0.03 (0.01-0.06)	0.05 (0.02-0.08)	0.06 (0.05-0.06)	0.08 (0.07-0.10)	0.10 (0.08-0.12)
4	0.01 (0.006-0.02)	0.03 (0.01-0.05)	0.04 (0.02-0.07)	0.05 (0.04-0.06)	0.08 (0.06-0.09)	0.09 (0.07-0.11)
5	0.01 (0.006-0.02)	0.03 (0.01-0.05)	0.04 (0.02-0.07)	0.05 (0.04-0.06)	0.07 (0.06-0.09)	0.08 (0.07-0.11)
6	0.01 (0.006-0.02)	0.03 (0.01-0.04)	0.04 (0.02-0.06)	0.04 (0.04-0.05)	0.07 (0.05-0.08)	0.08 (0.06-0.10)
2-6	0.01 (0.007-0.02)	0.03 (0.01-0.05)	0.04 (0.02-0.07)	0.05 (0.04-0.06)	0.08 (0.06-0.10)	0.10 (0.07-0.14)
7-69	0.006 (0.004-0.008)	0.01 (0.009-0.02)	0.02 (0.01-0.02)	0.02 (0.02-0.03)	0.04 (0.04-0.05)	0.05 (0.05-0.06)
PBDE-183[d]						
2	0.02 (0.005-0.05)	0.04 (0.009-0.10)	0.05 (0.01-0.13)	0.09 (0.07-0.11)	0.13 (0.10-0.18)	0.16 (0.12-0.21)
3	0.02 (0.005-0.04)	0.04 (0.009-0.09)	0.05 (0.01-0.12)	0.08 (0.06-0.10)	0.12 (0.09-0.16)	0.14 (0.11-0.18)

Age (years)	Percentiles of exposure (ng /kg bw per day)					
	LB scenario[a]			UB scenario[b]		
	P50	P95	P99	P50	P95	P99
4	0.02 (0.005-0.04)	0.04 (0.008-0.08)	0.05 (0.01-0.11)	0.07 (0.06-0.09)	0.11 (0.08-0.14)	0.13 (0.10-0.17)
5	0.02 (0.005-0.04)	0.03 (0.008-0.07)	0.04 (0.01-0.10)	0.07 (0.05-0.09)	0.10 (0.08-0.13)	0.12 (0.09-0.16)
6	0.02 (0.004-0.04)	0.03 (0.008-0.07)	0.04 (0.009-0.10)	0.06 (0.05-0.08)	0.10 (0.07-0.13)	0.12 (0.09-0.15)
2-6	0.02 (0.005-0.04)	0.03 (0.008-0.07)	0.04 (0.01-0.09)	0.07 (0.06-0.09)	0.12 (0.09-0.15)	0.14 (0.11-0.19)
7-69	0.01 (0.002-0.02)	0.02 (0.005-0.05)	0.03 (0.007-0.07)	0.04 (0.03-0.06)	0.06 (0.05-0.11)	0.08 (0.06-0.14)

Note: 2.5% lower - 97.5% upper confidence limits of the percentiles of exposure are reported between brackets.
[a] LB = lower bound, PBDE congener concentrations below LOQ were equalled to 0 ng/kg fat or product.
[b] UB = upper bound, PBDE congener concentrations below LOQ were equalled to LOQ.
[c] Exposure was calculated using the Observed Individual Mean (OIM) model. For details, see section 2.4.
[d] Exposure was calculated using the LogNormal-Normal (LNN) model. For details, see section 2.4 and Appendix E.

Appendix I Mean concentrations of three PBDE congeners in products of animal and vegetable origin used in the present study and an earlier Dutch study using concentration data of 2008 (Zeilmaker et al., 2014) in which samples with PBDE congener concentrations below a limit value equalled half this limit value (medium bound scenario)

Product[a]	Concentrations in ng/g fat or product[b, c]					
	PBDE-47		PBDE-99		PBDE-100	
	Present study	Zeilmaker et al. (2014)	Present study	Zeilmaker et al. (2014)	Present study	Zeilmaker et al. (2014)
Cereal products (product)[d]	0.0016	0.025	0.0039	0.004	0.0008	0.002
Cow's milk (fat)	0.046	1.700	0.021	0.500	0.007	0.300
Egg yolk (fat)	0.032	0.622	0.061	0.233	0.019	0.144
Fish, lean (product)[e]	0.075	0.074	0.013	0.013	0.019	0.016
Fish, fatty (product)[e]	0.163	0.694	0.028	0.146	0.047	0.179
Fruits (product)	0.0015	0.014	0.001	0.003	0.0003	0.002
Meat of chicken and other poultry (fat)	0.030	0.811	0.050	0.433	0.008	0.200
Meat of cow (beef) (fat)	0.049	0.531	0.039	0.192	0.008	0.108
Nuts and seeds (product)	0.0012	-	0.0022	-	0.0008	-
Pork meat (fat)	0.050	0.348	0.062	0.190	0.008	0.071
Potato products (product)	0.0025	0.014	0.0051	0.003	0.0005	0.003
Shellfish (product)	0.063	0.189	0.019	0.089	0.013	0.052
Vegetables (product)	0.002	0.014	0.0016	0.003	0.0005	0.002
Vegetable oil (product)	0.0105	0.145	0.0103	0.061	0.0020	0.012
Wine (product)	0.0015	-	0.001	-	0.0003	-

[a] Between brackets, per product (group) is indicated whether the congener concentrations are reported per gram product (product) or fat (fat).

[b] PBDE congener concentrations in products of animal origin used in the present study and in Zeilmaker et al. (2014) were derived from samples analysed in 2011-2013 and 2008, respectively. Congener concentrations reported in Zeilmaker et al. (2014) per gram product for meat of chicken and other poultry, pork meat, meat of cow (beef), egg yolk and milk were converted to concentrations per gram fat based on the fat concentrations reported in that publication for reasons of comparison.
[c] Concentration below which PBDE congener concentrations were reported as "less than" is limit of quantification in the present study and limit of detection in Zeilmaker et al. (2014).
[d] PBDE congener concentrations for cereal products were analysed in flour by Zeilmaker et al. (2014), and were allocated to cereals (wheat, oat, rye and barley) for the exposure assessment. In the present study, these concentrations were obtained from mycotoxin-dedicated total diet study (mTDS) samples bread, rye and corn products, breakfast cereals, and biscuits and cookies (Sprong et al., 2016). PBDE congener concentrations analysed in cereal products in the present study were also allocated to cereals (wheat, oat, rye, maize, sweet corn, rice, spelt, millet, buckwheat and barley) for the exposure assessment.
[e] See Appendix D for the fish species included in these two groups.

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