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**Dietary intake of brominated flame  
retardants by the Dutch population**

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## Abstract

The mean dietary intake of a number of brominated flame retardants (BFRs) by the Dutch population is estimated using measurements from The Netherlands Institute for Fisheries Research (RIVO) and the consumption data from the third Dutch National Food Consumption Survey. The polybrominated diphenylether (PBDE) congeners BDE #28, #47, #99, #100, #153, #154, tetrabromobisphenol-A (TBBP-A) and hexabromocyclododecane (HBCD) were analysed by RIVO. Polybrominated biphenyls and PBDE congeners BDE #71, #77, #190, #209 were not detected in the food products. Since only a limited number of samples was measured and furthermore, the concentrations of a high percentage of the samples were below the limit of detection (LOD), the present study only gives a rough estimate of the dietary intake of BFRs. The calculated mean dietary intakes of the “middle” scenario (assuming a value of 0.5\*LOD for non-detects) and the “low” scenario (non-detects are set to zero; the values from this scenario are placed between brackets) are 3.2-3.5 (0.2) ng/kg bw/day for the sum of PBDEs, 0.04 (0.04) ng/kg bw/day for TBBP-A and 2.9 (1.5) ng/kg bw/day for HBCD. For most BFRs the differences between the results of the two scenarios are large. The attributed values to the non-detects apparently have a large influence on the result. In order to reduce the uncertainty in the estimate of the dietary intake of brominated flame retardants by the Dutch population more sensitive analysis techniques in food products as well as a larger number of analysed samples per food commodity are needed.

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## Samenvatting

De gemiddelde inname van een aantal gebromeerde vlamvertragers via de voeding door de Nederlandse bevolking is geschat op basis van metingen van het Rijks Instituut voor Visserij Onderzoek (RIVO) en gegevens van de derde Voedsel Consumptie Peiling (VCP). Van de volgende gebromeerde vlamvertragers is een inname geschat: de polybroomdifenylether (PBDE) congeneren BDE #28, #47, #99, #100, #153 en #154, tetrabroombisfenyl-A (TBBP-A) en hexabroomcyclododecaan (HBCD). Polybrombifenylen en de PBDE congeneren BDE #71, #77, #190 en #209 konden door het RIVO niet gedetecteerd worden in voedingsmiddelen. Doordat er slechts een beperkt aantal monsters is gemeten en bovendien de concentraties in de monsters in veel gevallen niet hoger waren dan de detectielimiet kan met de huidige data slechts een grove innameschatting worden gedaan. Er zijn twee scenario's gehanteerd bij de innameberekeningen. Het eerste scenario levert een "middelste" schatting van de inname, door voor de non-detects een waarde ter grootte van de helft van de opgegeven detectiegrens aan te nemen. Bij het tweede zogenaamde ondergrens-scenario zijn de non-detects op nul gesteld. De berekende gemiddelde innames volgens de twee scenario's, waarbij de resultaten van het ondergrens-scenario tussen haakjes worden weergegeven, zijn 0,01 (0,01) ng/kg lg/dag voor BDE #28, 0,7 (0,5) ng/kg lg/dag voor BDE #47, 0,5 (0,3) ng/kg lg/dag voor BDE #99, 0,2 (0,1) ng/kg lg/dag voor BDE #100, 1,0 (0,1) ng/kg lg/dag voor BDE #153, 0,5 (0,2) ng/kg lg/dag voor BDE #154, 3,2-3,5 (0,2) ng/kg lg/dag voor de som van PBDEs, 0,04 (0,04) ng/kg lg/dag voor TBBP-A en 2,9 (1,5) ng/kg lg/dag voor HBCD. Ter vergelijking: De PBDE inname door inhalatie van binnenuilucht is in het Verenigd Koninkrijk op ongeveer 33 ng/dag (0,5 ng/kg lg/dag) geschat. De verschillen tussen de resultaten van de twee innamescenario's zijn voor de meeste vlamvertragers groot. Ook zijn de procentuele bijdragen van de verschillende voedselgroepen aan de totale inname van gebromeerde vlamvertragers voor de twee scenario's vaak sterk verschillend. De toegekende waarden aan de non-detects hebben blijkbaar een grote invloed op het eindresultaat. Hieruit kan geconcludeerd worden dat gevoeligere analysetechnieken nodig zijn om een nauwkeuriger innameberekening van gebromeerde vlamvertragers via voeding door de Nederlandse bevolking te kunnen uitvoeren. Tevens zou een groter aantal monsters per voedselgroep moeten worden geanalyseerd om de verdeling binnen de populatie te kunnen berekenen.

## Summary

The mean dietary intake of a number of brominated flame retardants by the Dutch population is estimated using measurements of the Netherlands Institute for Fisheries Research (RIVO) and the consumption data from the third Dutch National Food Consumption Survey. The following brominated flame retardants were investigated: the polybrominated diphenylether (PBDE) congeners BDE #28, #47, #99, #100, #153, #154, tetrabromobisphenol-A (TBBP-A) and hexabromocyclododecane (HBCD). Polybromobiphenyls (PBBs) and PBDE congeners BDE #71, #77, #190, #209 were not detected in the food products by RIVO. Since only a limited number of samples was analysed and furthermore the concentrations of a large percentage of the samples were below the limit of detection (LOD), the present study only gives a rough estimate of the dietary intake of brominated flame retardants.

Two scenarios were applied for the intake estimations. The first scenario yields a “middle” estimate, by assuming the value of  $0.5 \times \text{LOD}$  for non-detects. In the second scenario (“low” scenario) a concentration of zero was used for non-detects. The calculated mean dietary intakes of the scenarios -the results of the second scenario are placed between brackets- are respectively 0.01 (0.01) ng/kg bw/day for BDE #28, 0.7 (0.5) ng/kg bw/day for BDE #47, 0.5 (0.3) ng/kg bw/day for BDE #99, 0.2 (0.1) ng/kg bw/day for BDE #100, 1.0 (0.1) ng/kg bw/day for BDE #153, 0.5 (0.2) ng/kg bw/day for BDE #154, 3.2-3.5 (0.2) ng/kg bw/day for the sum of PBDEs, 0.04 (0.04) ng/kg bw/day for TBBP-A and 2.9 (1.5) ng/kg bw/day for HBCD. In comparison: in the UK the intake of PBDEs via inhalation of indoor air is estimated at 33 ng/day (0.5 ng/kg bw/day).

The differences between the results of the two dietary intake scenarios is large for most BFRs. The contributions of the different food groups to the total intake are also different for the two scenarios. The attributed values to the non-detects apparently have a large influence on the result. Therefore it can be concluded that more sensitive analysis techniques for brominated flame retardants in food products are needed in order to reduce the uncertainty in the estimate of the dietary intake by the Dutch population. In addition, a larger number of samples per food commodity should be analysed to be able to estimate the intake distribution within the population.

## 1. Introduction

Brominated flame retardants (BFRs) are widely used in electronic household equipment (e.g. personal computers and television sets), plastics, textile and polyurethane foam in furniture and cars for safety reasons (Boon et al., 2002). The annual world production of flame retardants is roughly 600,000 tons, of which about 150,000 are brominated compounds. Of the brominated products, about one-third contain tetrabromobisphenol-A (TBBP-A) and derivatives, another third contain various bromines, including polybrominated biphenyls (PBBs) and hexabromo-cyclododecane (HBCD) and the last third contain PBDEs. The waste from the products mentioned above is either incinerated or deposited in landfills. Potential routes of discharging of BFRs into the environment is through incineration, sewage, leaching from landfills and through volatilisation from electrical components during their lifetime. (Darnerud et al., 2001 and references therein).

BFRs are persistent, bioaccumulative and toxic substances. PBDEs for example act on the thyroid gland and can cause immunotoxicity. The environmental fate of BFRs is similar to the fate of other persistent organic pollutants, such as PCBs and dioxins. The presence of BFRs in the environment has been shown in air, sewage sludge, sediments, fish, birds and mammals, including human breast milk and adipose tissue (De Wit, 2002; Boon et al., 2002). The levels of PBDEs seem to be increasing, and several trend analyses, including in humans, indicate that this increase may be rapid (De Wit, 2002). The intake of BFRs can occur via the food and the inhalation of (indoor) air. Since the behaviour of BFRs is similar to that of PCBs and dioxins, food products of animal origin with high fat content (fatty fish, meat and dairy products) are expected to be major contributors to dietary exposure. For nursing infants the exposure to BFRs via mother's milk is significant. An exception may be TBBP-A, for which no evidence is present to indicate a potential for bioaccumulation. The exposure to BFRs via breast milk is presented in a separate report (De Winter-Sorkina et al., 2003). The concentrations of BFRs in human food products are not well known, except for the results of a few studies on concentrations of PBDEs in chickens in the US (Huwe et al., 2000), game in Sweden (Sellström et al., 1996) and cow's milk in Germany (Krüger, 1988). In addition, concentrations of PBDEs and PBBs in fish have been reported (De Boer et al., 1998; Jacobs et al., 2002; an overview in De Wit, 2002). The intake of BFRs via the inhalation of air is caused by the emissions from electronic equipment like computers and TV sets, especially from warm devices after prolonged use. Wijesekera et al. (2002) measured PBDEs in food samples and in indoor air. It was reported that diet and inhalation contribute 73 % and 27 % respectively to an overall median daily exposure to the sum of PBDEs of 123 ng/day per person.

Recently, the Netherlands Institute for Fisheries Research (RIVO) investigated the background concentrations of BFRs in food products consumed by the Dutch population (Leonards et al., 2002). The PBDE residues in poultry measured by RIVO are lower than found in chickens from the US (Huwe et al., 2000). The objective of the present study is to calculate the dietary intake of BFRs by the Dutch population (age 1 year and older) using the results of the Netherlands Institute for Fisheries Research (RIVO). A risk assessment of the intake of BFRs is not carried out in the current study. It is recommended to perform this in a future study.

## 2. Data and methods

### 2.1 Samples

The RIVO determined the concentrations of PBDEs, HBCD, PBBs and TBBP-A in 91 samples (Leonards et al., 2002; see Appendix 1) from the food categories dairy, meat, animal fat, eggs, fish and vegetable oil (Table 1). The samples were obtained from the current investigation into dioxins and polychlorinated biphenyls of the Inspectorate for Health Protection (Keuringsdienst van Waren), region East. For each product pooled samples were made by the Inspectorate.

*Table 1. Food products analysed by RIVO.*

Category	Product	Number of (pooled) samples
Dairy and dairy products	blue-veined cheese	11
	hard cheese	2
	milk	6
	whipping cream	2
	coffee creamer	1
Eggs	egg	9
Meat and poultry	beef	7
	pork	5
	poultry	9
Animal fats	fat of cattle	4
	fat of pigs	3
Fish	plaice	1
	salmon	1
	mackerel	2
	herring	2
	eel	8*
	mussel	2
	shrimp	1
Vegetable oil	oil	2
	sunflower olive oil	6

\*smoked eel (n=1), IJsselmeer eel (n=2), hatched eel (n=3) and imported eel (n=2)

The samples were analysed by RIVO for PBDEs, PBBs, TBBP-A, Me-TBBP-A and HBCD (Appendix 2). Processed samples are analysed by gas chromatography with mass spectrometry.

### 2.2 Intake calculation

The dietary exposure to brominated flame retardants was estimated using the consumption data of the third Dutch National Food Consumption Survey (DNFCS). This survey describes the consumption pattern of the Dutch population and includes information on the daily consumption over two consecutive days and a record of age, sex and body weight of 6250 individuals (Kistemaker et al., 1998). For the calculation of the dietary intake, the intake of primary agricultural products rather than that of individual food products on the level of the Netherlands Nutrient database (NEVO, 1996) was considered. An exception was made for the food products hard cheese, soft

cheese and whipping cream, since concentrations were determined in these individual food products (which are not primary agricultural products). The products sunflower oil and olive oil were taken together as the primary product vegetable oil. The conversion of the NEVO food products to the primary agricultural products is made using the RIKILT CPAP conversion model (Van Dooren et al., 1995). The Dutch consumption of eel is assumed to consist of 90 % hatched eel, 5 % IJsselmeer eel and 5 % other eel. Therefore, concentrations in eel were calculated according to this ratio.

The average dietary intake of brominated flame retardants by the Dutch population was calculated based on mean data from the database (point estimate) by multiplication of the average compound concentration with the average consumption per food group. A flow diagram of the estimation method is presented in Figure 1. According to Boon et al. (2001) the number of positive measurements per food commodity required for a sensible calculation of an intake distribution with a Monte Carlo Risk Analysis (MCRA) should at least equal 10. Because the amount of positive concentration data in all the food commodities (except for fish) is smaller than 10, the MCRA method of RIKILT was not used for the calculation of the intake.

Two scenarios were calculated. In scenario 1, the so-called “middle” scenario, for samples with a BFR level lower than the limit of detection (LOD), a value of  $0.5 \times \text{LOD}$  was assumed (Figure 2a). The RIVO determined a LOD for every analysis separately (Leonards et al., 2002). When a compound could not be detected at all in a food group, the food group was omitted for that compound. In scenario 2 (“low” scenario) the concentrations of non-detects are set to zero (Figure 2a). The reason to perform two calculations was that for some samples RIVO reported very high detection limits. For example, one pork sample (see Appendix 1) has a detection limit for BDE #153 of 8.2 ng/g, while the maximum measured BDE #153 concentration in pork equals 0.2 ng/g. Thus, in this case the non-detect concentration dominates the calculation of the intake. In addition to the separate compounds, we analysed the sum of the PBDEs. This sum was calculated in two manners, treating the non-detects in two different ways (Figure 2b). In scenario A (“congener scenario”) the PBDE congeners having only non-detects in a food group were assumed to have a concentration of 0. If one or more samples of a congener in a food group is positive, *for this congener* the rest of the samples in this group gets a value of  $0.5 \times \text{LOD}$  for scenario 1 or zero for scenario 2 (Figure 2b). In scenario B (“food group scenario”), if one or more of the PBDE congeners has one or more positive samples in a food group, the value of  $0.5 \times \text{LOD}$  or zero was taken for the samples of *all congeners of this food group* with concentrations  $< \text{LOD}$ , even if for a congener all samples are below the limit of detection (see Figure 2). The results of scenario 1 will logically be higher than those of scenario 2. Comparing the outcome of scenarios A and B is however more complicated: For scenario 1, since in scenario B a number of “zeros” of scenario A are replaced by  $0.5 \times \text{LOD}$ , the calculated mean concentration and intake of scenario B will be higher than for scenario A (Figure 2b). In scenario 2 the number of “zeros” of scenario B is higher than that of scenario A, and the calculated mean concentration and intake of scenario B will be lower than for scenario A (Figure 2b).

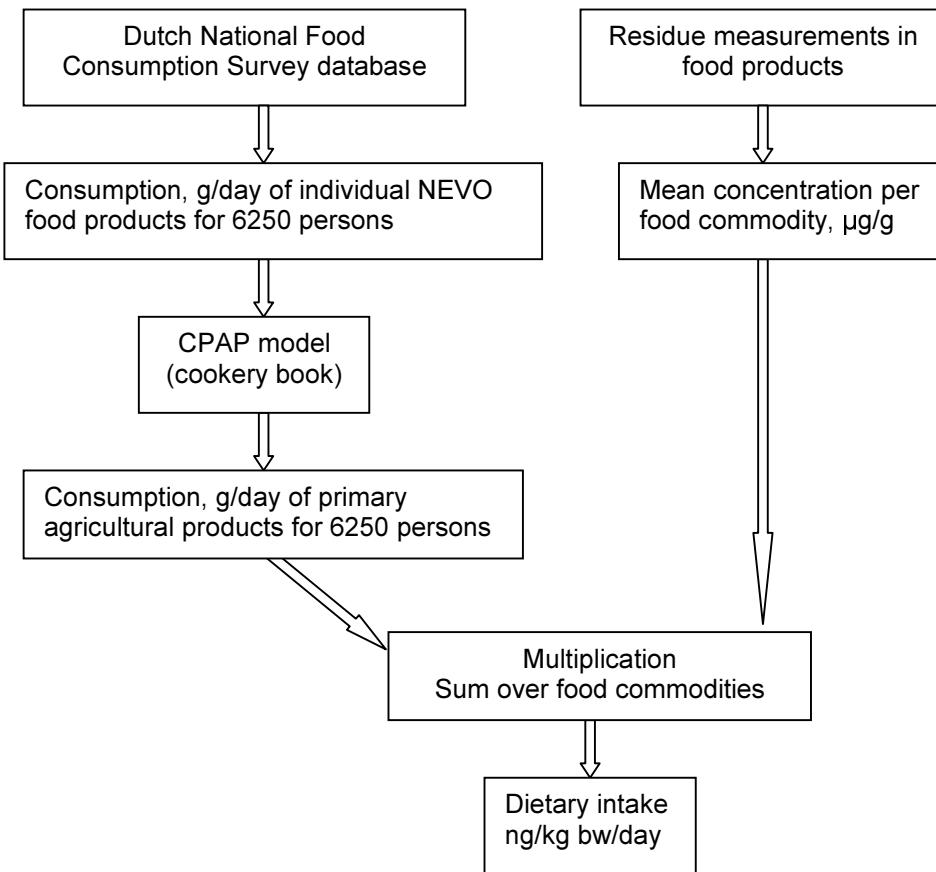


Figure 1 Flow diagram of the dietary intake estimation method.

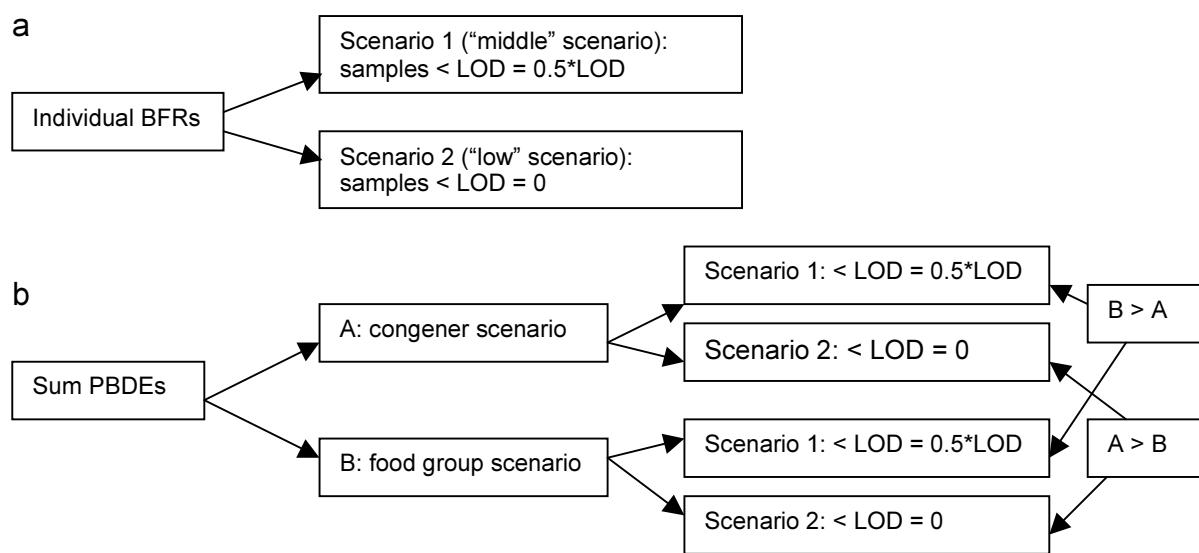


Figure 2 Scheme of scenarios used for the individual BFRs (a) and the sum of the PBDEs (b)

### 3. Results

#### 3.1 Average intake of brominated flame retardants by the Dutch population

The PBBs and PBDE congeners BDE #71, #77, #190, #209 were not detected in the food products by the analysis of RIVO. The PBDE congeners BDE #28, #47, #99, #100, #153 and #154 could be detected in some of the samples. BDE #28 was only detected in fish, while 12 out of 18 food categories showed positive samples for BDE #47, #99 and #100. The highest concentrations of these congeners were measured in herring, eel, mackerel, pork and fat of pigs. The presence of BDE #153 and #154 is proven in all categories except eggs and milk. TBBP-A was only found in fish and hard cheese, while HBCD was present in 15 out of 18 categories. However, the percentage of non-detects is high: In 60 %-91 % of the samples none of the various BFRs could be detected (Appendix 1). The high percentages of non-detects strongly influences the outcome of the intake estimates. Thus, the uncertainty is large and can only be reduced by the development of more sensitive analysis techniques. The average consumption of the different food categories for the whole population (consumers and non-consumers) and the number of consumers were obtained from the DNFCs (Table 2).

Table 2. Average consumption and a number of consumers (from DNFCs) of various food groups.

Category	Consumption (whole population) g/day	No. of consumers (two days)
blue-veined cheese*	1.5	617
cheese**	25.0	8173
Whipped cream	1.5	984
Beef	41.0	8212
Fat of cattle	1.0	1923
Pork	55.2	8943
Fat of pig	1.0	1907
Poultry	19.4	3315
Eggs	17.1	8171
Cow's milk	351.4	11655
Eel	0.1	41
Herring	1.8	201
Mackerel	0.3	53
Plaice	0.5	70
Salmon	1.1	341
Shrimp	0.4	263
Mussels	0.6	44
Vegetable oil and fats	23.7	11739

\*NEVO product codes: 556, 593, 714, 716, 719, 726, 1108, 1302, 1487.

\*\*NEVO product codes: 304, 511-517, 654, 718, 721-722, 724-725, 881-883, 927-928, 1103-1104, 1109-1110, 1112-1113, 1382, 1385-1386, 1489-1490, 1723-1727, 1809, 9644.

For the sum of PBDEs (congener scenario) the mean concentrations, the calculated intakes and the contributions to the total intake according to scenario 1 and 2 are presented in Table 3. Only those food groups which contribute more than 2 % to the total intake are

included in the table. The differences between the results of the congener scenario and the food group scenario are small, therefore only the former scenario is presented. The complete set of data can be found in Appendix 3. For HBCD a similar table is presented (Table 4). We report the sum of the PBDEs here, since it can be compared with other studies, while HBCD was chosen because of its highest incidence.

From both tables it appears that the difference between scenario 1 and 2 in the mean concentrations of the food groups is large, due to the large number of non-detects and the relative high detection limits of some samples. This leads to large differences in the mean intakes and consequently, to large differences in the relative contributions to the total intake for the two scenarios.

The food groups pork, beef and herring give the highest contributions to the total intake of the sum of PBDEs (Table 3). For HBCD, the food groups with the highest contributions to the total intake are beef, poultry and vegetable oils and fats (Table 4).

*Table 3. Mean concentrations, calculated mean intakes of the sum of PBDEs (congener scenario) and the relative contribution to the total average intake by the Dutch population per food category, according to scenario 1 (<LOD = 0.5\*LOD) and scenario 2 (<LOD=0). Only categories with contributions  $\geq 2\%$  are shown.*

Category	n > LOD	Mean conc., ng/g Scenario 1	Mean conc., ng/g Scenario 2	Mean intake ng/day Scenario 1	Mean intake ng/day Scenario 2	Contrib. % Scenario 1	Contrib. % Scenario 2
Cheese*	2	0.3	0.04	7.3	1.0	3	7
Beef	4	0.7	0.08	27.0	3.1	<b>13</b>	<b>21</b>
Pork	2	2.1	0.08	117.6	4.2	<b>55</b>	<b>28</b>
Poultry	5	0.3	0.04	6.7	0.7	3	5
Herring	2	12.9	2.14	22.7	3.8	<b>11</b>	<b>25</b>
Salmon	1	3.4	0.57	3.8	0.6	2	4

\*NEVO product codes: 304, 511-517, 654, 718, 721-722, 724-725, 881-883, 927-928, 1103-1104, 1109-1110, 1112-1113, 1382, 1385-1386, 1489-1490, 1723-1727, 1809, 9644.

*Table 4. Mean concentrations, calculated mean intakes of HBCD and the relative contribution to the total average intake by the Dutch population per food category, according to scenario 1 (<LOD = 0.5\*LOD) and scenario 2 (<LOD=0). Only categories with contributions  $\geq 2.0\%$  are shown.*

Category	n > LOD	Mean conc., ng/g Scenario 1	Mean conc., ng/g Scenario 2	Mean intake ng/day Scenario 1	Mean intake ng/day Scenario 2	Contrib. % Scenario 1	Contrib. % Scenario 2
Beef	1	1.8	0.4	75.3	18.2	<b>40</b>	<b>19</b>
Fat of cattle	2	3.8	3.0	3.8	3.0	2	3
Poultry	3	2.6	1.5	50.8	29.0	<b>27</b>	<b>30</b>
Eggs	2	0.4	0.3	6.0	5.1	3	5
Cow's milk	1	0.03	0.02	10.5	6.4	6	7
Herring	2	5.8	5.8	10.2	10.2	5	<b>11</b>
Mackerel	2	7.9	7.9	2.1	2.1	1	2
Veg. oil and fats	3	1.1	0.8	27.1	18.0	<b>14</b>	<b>19</b>

A summary of the total average intakes of the BFRs is reported in Table 5. Ranking the BFRs according to their calculated intakes the following result is obtained:

HBCD > BDE #153 > BDE #47 > BDE #99  $\approx$  BDE #154 > BDE #100 >> BDE #28.

Be aware of the fact that the uncertainty in the calculated intakes is large, due to the low number of (positive) samples.

*Table 5. Total average dietary intake of BFRs by the Dutch population, according to scenario 1 ( $<LOD=0.5*LOD$ ) and scenario 2 ( $<LOD=0$ ).*

Compound	Mean intake, ng/(kg bw day) <sup>1</sup> scenario 1	Mean intake, ng/(kg bw day) <sup>1</sup> scenario 2
BDE #28	0.01	0.01
BDE #47	0.7	0.5
BDE #99	0.5	0.3
BDE #100	0.2	0.1
BDE #153	1.0	0.1
BDE #154	0.5	0.2
Sum PBDEs	3.2 (scenario A) <sup>2</sup> 3.5 (scenario B) <sup>3</sup>	0.2 (scenario A) 0.2 (scenario B)
TBBP-A	0.04	0.04
HBCD	2.9	1.5

<sup>1</sup> bw – body weight, the mean body weight of DNFCS participants was 65.8 kg.

<sup>2</sup> scenario A: congener scenario

<sup>3</sup> scenario B: food group scenario

### 3.2 Comparison with other studies

The dietary intake of PBDEs was also studied in Canada, the UK and Sweden.

The daily dietary exposure of Canadian adults to the sum of PBDEs (#28, #47, #99, #100, #153 and #154) was estimated via a “market basket” study (pooled samples composed from different food products) at 44 ng/day per person (Ryan and Patry, 2001; Table 6). In the UK, the dietary intake of the sum of PBDEs was estimated using data on the PBDE content of 10 duplicate diet samples (pooled samples of food eaten by a person during the whole day) and food mass ingestion data for the individuals consuming each diet sample (Wijsekera et al., 2002). The estimated median value for the “low” scenario (i.e. samples  $<LOD$  are set to zero) was 90.5 ng/day per person. The data of Wijsekera et al. (2002) indicate a wide range of individual intakes, from 37 ng/day to 235 ng/day (Table 6). The Swedish dietary intake estimate (Darnerud et al., 2001) was made on the basis of PBDE levels (BDE #47, #99, #100, #153, and #154) in market basket samples from 1999. Assuming a concentration of half the detection limit for those samples which had concentrations below the detection limit (“middle” estimation), the total PBDE intake in Sweden was estimated to be 51 ng/day (Table 6). The “low” estimate of the mean intake of the sum of PBDEs in The Netherlands is lower than that in the UK and Canada, but the “middle” estimate is higher than in Sweden. However, it is difficult to make a fair comparison of the results of the present study with the ones from Canada, UK and Sweden. In the measurements from RIVO, used in the current study, samples of individual food products are analysed. The other studies used samples composed

of different food products. Firstly, when the analysis method is not very sensitive, in composed samples the residue concentration detected in a single food product can be spread out and become a non-detect. Secondly, the “market basket” method used in the Canadian and Swedish studies does not include all food products. Although measurements of RIVO are performed for a limited number of food products, the intake estimation method used in the present study includes a considerable number of composite food products containing these analysed primary products.

*Table 6. Comparison of dietary intakes of the sum of PBDEs from different studies.*

Country	Mean dietary intake of the sum of PBDEs ng/day	Method	Reference
Canada	44	“market basket”, composed samples	Ryan, 2001
UK	90.5* (range 37-235)*	“duplicate diet”, composed samples from 10 individuals	Wijesekera, 2002
Sweden	51 <sup>+</sup>	“market basket”, composed samples per food group	Darnerud, 2001
The Netherlands	213 <sup>+</sup> (13*)	Single food product samples	This study

\* “low” estimate

<sup>+</sup> “middle” estimate

## 4. Conclusions and recommendations

Using the present concentration data only a rough estimation of the intake of BFRs can be made, due to the low amount of analysed samples and high percentage of non-detects herein. The calculated mean dietary intakes of the “middle” scenario (assuming a value of 0.5\*LOD for non-detects) and the “low” scenario (non-detects are set to zero; the values from this scenario are placed between brackets) are 3.2-3.5 (0.2) ng/kg bw/day for the sum of PBDEs, 0.04 (0.04) ng/kg bw/day for TBBP-A and 2.9 (1.5) ng/kg bw/day for HBCD.

The difference between the results of the two intake scenarios (“middle” and “low”), whether looking at calculated intakes or at relative contributions per food category to the total intake, is large for most BFRs, indicating that the results of the “middle” scenario are largely determined by the levels of the detection limits. Therefore it is concluded that more sensitive detection techniques are needed to reduce the uncertainty in the calculations. In addition, a larger number of samples per food commodity should be analysed to reduce the uncertainty in the intake estimation.

Although not estimated in the present study, the exposure to BFRs via inhalation of indoor air may be significant (~one third of the intake via food) and should be taken into account.

## Acknowledgment

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## Appendix 1 Concentrations of BFRs in food samples

Concentrations (ng/g product) of dominant PBDE congeners, TBBP-A and HBCD in analysed samples (Leonards et al., 2002).

2002/0204	Coffee creamer	<0.02	<0.02	<0.02	<0.02	<0.02	<0.02	<0.02	<0.5
2002/0206	Whipped cream	<0.07	<0.08	<0.08	<0.07	<0.1	0.04	<0.1	<0.4
2002/0207	Whipped cream	<0.08	<0.09	<0.09	<0.08	<0.1	0.04	<0.2	<0.4
2002/0168	Eggs	<0.03	<0.03	<0.03	<0.03	<0.03	<0.03	<0.02	<0.14
2002/0169	Eggs	<0.02	<0.03	<0.03	<0.02	<0.02	<0.02	<0.02	<0.13
2002/0170	Eggs	<0.02	<0.03	<0.03	<0.03	<0.03	<0.03	<0.02	<0.13
2002/0171	Eggs	<0.03	<0.03	<0.03	<0.03	<0.03	<0.03	<0.03	<0.14
2002/0172	Eggs	<0.02	<0.03	<0.03	<0.02	<0.02	<0.02	<0.02	1.1
2002/0173	Eggs	<0.02	<0.02	<0.02	<0.02	<0.02	<0.02	<0.02	<0.11
2002/0174	Eggs	<0.03	<0.03	<0.03	<0.03	<0.03	<0.03	<0.02	1.6
2002/0175	Eggs	<0.02	<0.03	<0.03	<0.02	<0.02	<0.02	<0.02	<0.13
2002/0176	Eggs	<0.03	<0.04	<0.03	<0.03	<0.03	<0.03	<0.03	<0.16
2002/0199	Olive oil	<0.2	<0.2	<0.2	<0.2	<0.2	<0.2	*	<1.1
2002/0198	Olive oil	<0.6	<0.6	<0.2	<0.3	<0.2	<0.3	*	<1.3
2002/0202	Olive oil	<0.7	<0.6	<0.2	<0.3	<0.2	<0.2	*	3.0
2002/0203	Olive oil	<0.8	<0.6	<0.2	<0.3	<0.2	<0.2	*	1.7
2002/0201	Olive oil	<0.9	<0.6	<0.2	<0.3	<0.2	<0.2	*	<1.2
2002/0200	Olive oil	<0.1	<0.6	<0.2	<0.3	<0.2	<0.2	*	<1.2
2001/2023	Vegetable oil	<0.2	<0.3	<0.3	<0.2	0.4	0.1	*	1.4
2001/2025	Vegetable oil	<0.6	<0.6	<0.2	<0.3	<0.2	<0.2	*	<1.3
2001/2020	Salmon	0.1	2.1	0.4	0.3	0.3	0.2	<0.05	1.6
2002/1077	Plaice	0.007	0.3	0.01	0.06	0.009	0.03	<0.05	<0.1
2001/2017	Mackerel	0.2	4.6	2.0	0.07	0.3	0.3	<0.16	6.8
2001/2018	Mackerel	0.07	1.4	0.6	0.3	0.1	0.1	<0.19	8.9
2001/2014	Herring	0.2	6.8	1.1	1.4	0.1	0.2	0.6	6.5
2001/2015	Herring	0.3	8.7	3.9	2.5	0.2	0.3	<0.12	5.0
2001/2016	Smoked eel	0.2	9.3	1.0	1.0	0.2	0.2	<0.2	7.2
2002/1065	IJsselmeer eel	0.30	14	1.1	3.5	1.2	1.0	<0.1	12
2002/1066	IJsselmeer eel	0.05	2.0	0.1	0.7	0.3	0.30	<0.1	1.7
2002/1067	Hatched eel	<0.1	1.7	0.1	0.3	<0.1	0.1	0.2	<0.4
2002/1068	Hatched eel	0.20	4.7	0.4	0.7	<0.1	0.1	3.4	1.7
2002/1069	Hatched eel	0.20	5.4	0.3	0.9	<0.1	0.2	0.2	1.5
2002/1070	Imported eel	0.30	5.9	0.27	0.9	0.1	0.2	1.3	<0.5
2002/1071	Imported eel	0.10	11	0.4	3.3	0.6	0.8	<0.1	6.7
2002/1072	Mussel	0.02	0.2	0.06	0.05	<0.08	<0.08	<0.1	<0.1
2002/1073	Mussel	0.01	0.2	0.07	0.05	<0.1	0.01	0.001	0.6
2002/1078	Shrimp	<0.03	0.03	0.02	0.01	<0.03	0.01	<0.03	<0.1

\* bad quality measurement

## Appendix 2 Brominated flame retardants analysed by RIVO

Name	IUPAC-nr	Structure
PBDE	28	2,4,4'-tri
	47	2,4,2',4'-tetra
	66	2,3,4,4'-tetra
	71	2,6,3',4'-tetra
	75	2,4,6,4'-tetra
	77	3,4,3',4'-tetra
	85	2,3,4,2',4'-penta
	99	2,4,5,2',4'-penta
	100	2,4,6,2',4'-penta
	119	2,4,6,3',4'-penta
	138	2,3,4,2',4',5'-hexa
	153	2,4,5,2',4',5'-hexa
	154	2,4,5,2',4',6'-hexa
	190	2,3,4,5,6,3',4'-hepta
	209	2,3,4,5,6,2',3',4',5',6'-deca
PBB	15	4,4'-di
	49	2,4,2',5'-tetra
	52	2,5,2',5'-tetra
	101	2,4,5,2',5'-penta
	153	2,4,5,2',4',5'-hexa
	209	2,3,4,5,6,2',3',4',5',6'-deca
TBBP-A		tetrabromobisphenol-A
Me-TBBP-A		methyl-tetrabromobisphenol-A
HBCD		hexabromocyclododecane

## Appendix 3 Dietary intake of BFRs

The average concentrations (from RIVO data), the calculated average intakes of BFRs and the relative contribution of food categories to the total average intake by the Dutch population per food category. “Middle” estimate: concentrations of non-detects are set to 0.5\*LOD. “Low” estimate: concentrations of non-detects are set to zero.

Category	No. of samples > LOD	Mean conc., ng/g <LOD=0.5 *LOD	Mean conc., ng/g <LOD=0	Mean intake, ng/day <LOD=0.5 *LOD	Mean intake, ng/day <LOD=0	Relative contribu-tion, % <LOD=0.5*LOD	Relative contribu-tion, % <LOD=0
<b>BDE 28</b>							
Sum of blue-veined cheese*	-	-	-	-	-	-	-
Sum of cheese**	-	-	-	-	-	-	-
Whipped cream	-	-	-	-	-	-	-
Beef	-	-	-	-	-	-	-
Fat of cattle	-	-	-	-	-	-	-
Pork	-	-	-	-	-	-	-
Fat of pig	-	-	-	-	-	-	-
Poultry	-	-	-	-	-	-	-
Eggs	-	-	-	-	-	-	-
Cow's milk	-	-	-	-	-	-	-
Eel	7	0.15	0.14	0.02	0.02	3.4	3.1
Herring	2	0.25	0.25	0.44	0.44	<b>70.9</b>	<b>70.2</b>
Mackerel	2	0.14	0.14	0.04	0.04	5.8	5.8
Plaice	1	0.01	0.01	0.004	0.004	0.6	0.6
Salmon	1	0.10	0.10	0.11	0.11	<b>17.8</b>	<b>17.6</b>
Shrimp	-	-	-	-	-	-	-
Mussels	2	0.02	0.02	0.01	0.01	1.4	1.4
Vegetable oil and fats	-	-	-	-	-	-	-
<b>BDE 47</b>							
Sum of blue-veined cheese*	-	-	-	-	-	-	-
Sum of cheese**	2	0.10	0.10	2.50	2.50	5.4	8.1
Whipped cream	-	-	-	-	-	-	-
Beef	1	0.16	0.10	6.62	4.04	<b>14.3</b>	<b>13.1</b>
Fat of cattle	-	-	-	-	-	-	-
Pork	1	0.32	0.10	17.66	5.52	<b>38.2</b>	<b>17.9</b>
Fat of pig	1	0.21	0.12	0.20	0.12	0.4	0.4
Poultry	1	0.08	0.04	1.53	0.86	3.3	2.8
Eggs	-	-	-	-	-	-	-
Cow's milk	-	-	-	-	-	-	-
Eel	8	4.38	4.38	0.61	0.61	1.3	2.0
Herring	2	7.75	7.75	13.72	13.72	<b>29.6</b>	<b>44.5</b>
Mackerel	2	3.00	3.00	0.81	0.81	1.8	2.6
Plaice	1	0.30	0.30	0.16	0.16	0.4	0.5
Salmon	1	2.10	2.10	2.33	2.33	5.0	7.6
Shrimp	1	0.03	0.03	0.01	0.01	0.03	-
Mussels	2	0.20	0.20	0.11	0.11	0.2	0.4

Vegetable oil and fats	-	-	-	-	-	-	-
<b>BDE 99</b>							
Sum of blue-veined cheese*	-	-	-	-	-	-	-
Sum of cheese**	2	0.07	0.07	1.75	1.75	4.9	9.6
Whipped cream	-	-	-	-	-	-	-
Beef	1	0.18	0.11	7.21	4.34	<b>20.2</b>	<b>23.8</b>
Fat of cattle	1	0.49	0.40	0.49	0.40	1.4	2.2
Pork	1	0.35	0.10	19.32	5.52	<b>54.2</b>	<b>30.3</b>
Fat of pig	-	-	-	-	-	-	-
Poultry	1	0.08	0.04	1.53	0.86	4.3	4.7
Eggs	-	-	-	-	-	-	-
Cow's milk	-	-	-	-	-	-	-
Eel	8	0.30	0.30	0.04	0.04	0.1	0.2
Herring	2	2.50	2.50	4.43	4.43	<b>12.4</b>	<b>24.3</b>
Mackerel	2	1.30	1.30	0.35	0.35	1.0	1.9
Plaice	1	0.01	0.01	0.01	0.01	0.02	0.03
Salmon	1	0.40	0.40	0.44	0.44	1.2	2.4
Shrimp	1	0.02	0.02	0.01	0.01	0.02	0.05
Mussels	2	0.07	0.07	0.04	0.04	0.1	0.2
Vegetable oil and fats	-	-	-	-	-	-	-
<b>BDE 100</b>							
Sum of blue-veined cheese*	-	-	-	-	-	-	-
Sum of cheese**	2	0.03	0.03	0.75	0.75	4.8	8.5
Whipped cream	-	-	-	-	-	-	-
Beef	1	0.09	0.03	3.63	1.35	<b>23.1</b>	<b>15.3</b>
Fat of cattle	-	-	-	-	-	-	-
Pork	1	0.11	0.04	6.07	2.21	<b>38.7</b>	<b>25.0</b>
Fat of pig	1	0.14	0.07	0.14	0.07	0.9	0.8
Poultry	1	0.06	0.02	1.10	0.43	7.0	4.9
Eggs	-	-	-	-	-	-	-
Cow's milk	-	-	-	-	-	-	-
Eel	8	0.76	0.76	0.11	0.11	0.7	1.2
Herring	2	1.95	1.95	3.45	3.45	<b>22.0</b>	<b>39.1</b>
Mackerel	2	0.19	0.19	0.05	0.05	0.3	0.6
Plaice	1	0.06	0.06	0.03	0.03	0.2	0.4
Salmon	1	0.30	0.30	0.33	0.33	2.1	3.8
Shrimp	1	0.01	0.01	0.004	0.004	0.03	0.05
Mussels	2	0.05	0.05	0.03	0.03	0.2	0.3
Vegetable oil and fats	-	-	-	-	-	-	-
<b>BDE 153</b>							
Sum of blue-veined cheese*	2	0.05	0.03	0.08	0.05	0.1	0.6
Sum of cheese**	1	0.03	0.02	0.69	0.38	1.1	5.0
Whipped cream	-	-	-	-	-	-	-
Beef	1	0.11	0.05	4.57	1.99	7.2	<b>26.3</b>
Fat of cattle	1	0.25	0.18	0.25	0.18	0.4	2.3
Pork	1	0.94	0.04	51.89	2.21	<b>82.3</b>	<b>29.2</b>
Fat of pig	2	0.55	0.50	0.54	0.49	0.9	6.5
Poultry	1	0.06	0.02	1.13	0.43	1.8	5.7
Eggs	-	-	-	-	-	-	-

Cow's milk	-	-	-	-	-	-	-
Eel	5	0.10	0.05	0.01	0.01	0.02	0.1
Herring	2	0.15	0.15	0.27	0.27	0.4	3.5
Mackerel	2	0.20	0.20	0.05	0.05	0.1	0.7
Plaice	1	0.01	0.01	0.005	0.005	0.01	0.06
Salmon	1	0.30	0.30	0.33	0.33	0.5	4.4
Shrimp	-	-	-	-	-	-	-
Mussels	-	-	-	-	-	-	-
Vegetable oil and fats	1	0.14	0.05	3.25	1.18	5.2	<b>15.6</b>

**BDE 154**

Sum of blue-veined cheese*	2	0.05	0.003	0.08	0.005	0.2	0.04
Sum of cheese**	-	-	-	-	-	-	-
Whipped cream	2	0.04	0.04	0.06	0.06	0.2	0.5
Beef	4	0.12	0.09	4.92	3.81	<b>15.1</b>	<b>32.9</b>
Fat of cattle	-	-	-	-	-	-	-
Pork	2	0.41	0.10	22.63	5.52	<b>69.6</b>	<b>47.7</b>
Fat of pig	1	0.13	0.07	0.13	0.07	0.4	0.6
Poultry	5	0.07	0.05	1.39	1.06	4.3	9.1
Eggs	-	-	-	-	-	-	-
Cow's milk	-	-	-	-	-	-	-
Eel	8	0.17	0.17	0.02	0.02	0.1	0.2
Herring	2	0.25	0.25	0.44	0.44	1.4	3.8
Mackerel	2	0.20	0.20	0.05	0.05	0.2	0.5
Plaice	1	0.03	0.03	0.02	0.02	0.0	0.1
Salmon	1	0.20	0.20	0.22	0.22	0.7	1.9
Shrimp	1	0.01	0.01	0.004	0.004	0.01	0.04
Mussels	1	0.03	0.01	0.01	0.003	0.04	0.02
Vegetable oil and fats	1	0.11	0.01	2.51	0.30	7.7	2.6

**Sum of PBDEs "Middle" scenario (<LOD=0.5\*LOD)**

Sum of blue-veined cheese*	4	Mean2 0.30	Mean1 0.11	Mean2 0.44	Mean1 0.16	Mean2 0.2	Mean1 0.1
Sum of cheese**	2	0.29	0.29	7.32	7.32	3.2	3.4
Whipped cream	2	0.25	0.04	0.38	0.06	0.2	0.03
Beef	4	0.79	0.66	32.52	26.95	<b>14.3</b>	<b>12.6</b>
Fat of cattle	1	1.13	0.74	1.14	0.74	0.5	0.3
Pork	2	2.23	2.13	123.10	117.58	<b>54.3</b>	<b>55.1</b>
Fat of pig	2	1.25	1.03	1.22	1.01	0.5	0.5
Poultry	5	0.40	0.34	7.79	6.69	3.4	3.1
Eggs	-	-	-	-	-	-	-
Cow's milk	-	-	-	-	-	-	-
Eel	8	5.86	5.86	0.82	0.82	0.4	0.4
Herring	2	12.85	12.85	22.74	22.74	<b>10.0</b>	<b>10.7</b>
Mackerel	2	5.02	5.02	1.36	1.36	0.6	0.6
Plaice	1	0.42	0.42	0.22	0.22	0.1	0.1
Salmon	1	3.40	3.40	3.77	3.77	1.7	1.8
Shrimp	1	0.10	0.07	0.04	0.03	0.02	0.01
Mussels	2	0.40	0.36	0.23	0.20	0.1	0.1
Vegetable oil and fats	1	1.00	1.00	23.65	23.65	<b>10.4</b>	<b>11.1</b>

**Sum of PBDEs "Low" scenario (<LOD=0)**

Sum of blue-veined cheese*	4	Mean2 0.006	Mean 1 0.018	Mean2 0.009	Mean1 0.026	Mean2 0.07	Mean1 0.17
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Sum of cheese**	2	0.028	0.041	0.689	1.033	5.4	6.8
Whipped cream	2	0.003	0.020	0.005	0.030	0.0	0.2
Beef	4	0.063	0.076	2.588	3.106	<b>20.4</b>	<b>20.5</b>
Fat of cattle	1	0.096	0.230	0.097	0.232	0.8	1.5
Pork	2	0.063	0.076	3.496	4.195	<b>27.5</b>	<b>27.7</b>
Fat of pig	2	0.127	0.191	0.125	0.187	1.0	1.2
Poultry	5	0.031	0.038	0.608	0.730	4.8	4.8
Eggs	-	-	-	-	-	-	-
Cow's milk	-	-	-	-	-	-	-
Eel	8	0.967	0.967	0.135	0.135	1.1	0.9
Herring	2	2.142	2.142	3.791	3.791	<b>29.8</b>	<b>25.0</b>
Mackerel	2	0.837	0.837	0.226	0.226	1.8	1.5
Plaice	1	0.069	0.069	0.037	0.037	0.3	0.2
Salmon	1	0.567	0.567	0.629	0.629	4.9	4.2
Shrimp	1	0.012	0.018	0.005	0.007	0.04	0.05
Mussels	2	0.046	0.055	0.026	0.031	0.2	0.2
Vegetable oil and fats	1	0.010	0.031	0.246	0.739	1.9	4.9

**TBBP-A**

Sum of blue-veined cheese*	-	-	-	-	-	-	-
Sum of cheese**	1	0.08	0.08	1.88	1.88	<b>71.1</b>	<b>72.9</b>
Whipped cream	-	-	-	-	-	-	-
Beef	-	-	-	-	-	-	-
Fat of cattle	-	-	-	-	-	-	-
Pork	-	-	-	-	-	-	-
Fat of pig	-	-	-	-	-	-	-
Poultry	-	-	-	-	-	-	-
Eggs	-	-	-	-	-	-	-
Cow's milk	-	-	-	-	-	-	-
Eel	4	1.17	1.19	0.16	0.17	6.2	6.5
Herring	1	0.33	0.30	0.58	0.53	<b>22.1</b>	<b>20.6</b>
Mackerel	-	-	-	-	-	-	-
Plaice	-	-	-	-	-	-	-
Salmon	-	-	-	-	-	-	-
Shrimp	-	-	-	-	-	-	-
Mussels	1	0.026	0.0005	0.01	0.0003	0.6	0.01
Vegetable oil and fats	-	-	-	-	-	-	-

**HBCD**

Sum of blue-veined cheese*	2	0.57	0.40	0.68	0.59	0.4	0.6
Sum of cheese**	-	-	-	-	-	-	-
Whipped cream	-	-	-	-	-	-	-
Beef	1	1.84	0.44	75.30	18.17	<b>39.7</b>	<b>19.0</b>
Fat of cattle	2	3.76	2.95	3.80	2.98	2.0	3.1
Pork	-	-	-	-	-	-	-
Fat of pig	2	1.02	0.80	1.00	0.78	0.5	0.8
Poultry	3	2.62	1.49	50.82	28.93	<b>26.8</b>	<b>30.3</b>
Eggs	2	0.35	0.30	6.02	5.12	3.2	5.4
Cow's milk	1	0.03	0.02	10.54	6.44	5.6	6.7
Eel	6	1.60	1.53	0.22	0.21	0.1	0.2
Herring	2	5.75	5.75	10.18	10.18	5.4	<b>10.7</b>
Mackerel	2	7.85	7.85	2.12	2.12	1.1	2.2
Plaice	1	0.05	-	0.03	-	0.01	-
Salmon	1	1.60	1.60	1.78	1.78	0.9	1.9
Shrimp	-	-	-	-	-	-	-
Mussels	1	0.33	0.30	0.19	0.17	0.1	0.2

Vegetable oil and fats	3	1.14	0.76	27.05	18.03	<b>14.3</b>	<b>18.9</b>
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\*NEVO product codes: 556, 593, 714, 716, 719, 726, 1108, 1302, 1487.

\*\*NEVO product codes: 304, 511-517, 654, 718, 721-722, 724-725, 881-883, 927-928, 1103-1104, 1109-1110, 1112-1113, 1382, 1385-1386, 1489-1490, 1723-1727, 1809, 9644.

Mean1: Congeners with no positive samples were assumed to have a concentration of zero.

Mean2: If one of the BDE congeners is positive for that sample, the value of 0.5\*LOD was taken for those congeners with concentrations < LOD.

## Appendix 4 Mailing list

- 1 Prof. P. Peters, VWA
- 2 Ir. H. de Goeij, VWS
- 3 Dr. ir. G. Kleter, KvW
- 4 Dr. J.M.de Stoppelaar, VGB, VWS
- 5 Drs. B.W. Ooms, VWA
- 6 Dr. ir. W. de Wit, VWA
- 7 Dr. W.F. Passchier, Gezondheidsraad
- 8 Dr. J. de Boer, RIVO, IJmuiden
- 9 Dr. P.E.G. Leonards, RIVO, IJmuiden
- 10 Dr. C.D. de Gooijer, RIKILT, Wageningen
- 11 Dr. R.F.M van Gorcom, RIKILT, Wageningen
- 12 Dr. K.F.A.M. Hulshof, TNO Voeding
- 13 Dr. C.A. de Wit, University of Stockholm
- 14 Prof. dr. J.P. Boon, NIOZ, Texel
- 15 Dr. G. Ellen, NIZO, Ede
- 16 Dr. P.O. Darnerud, National Food Administration, Sweden
- 17 Dr. T.A. McDonald, California EPA, USA
- 18 Dr. J.K.Ryan, Health Canada, Canada
- 19 Dr. R. Wijesekera, University of Colombo, Sri Lanka
- 20 Dr. G. Becher, Norwegian Institute of Public Health, Norway
- 21 Dr. R. Malisch, State Institute for Chemical and Veterinary Analysis of Food, Germany
- 22 Dr. J.S. LaKind, LaKind Associates, USA
- 23 Dr. S. Harrad, University of Birmingham, UK
- 24 Dr. L. S. Birnbaum, US EPA, USA
- 25-34 Werkgroep dioxinen
- 35 Depot Nederlandse Publikaties en Nederlandse Bibliografie
- 36 Directie RIVM
- 37 Prof. dr. ir. D. Kromhout
- 38 Dr. ir. M.N. Pieters
- 39 Dr. L.A. van Ginkel
- 40 Dr. E.A. Hogendoorn
- 41 Dr. F.X.R. van Leeuwen
- 42 Dr. A.G. Oomen
- 43 Dr. ir. A.J.A.M. Sips
- 44 Dr. C.H.M. Versantvoort
- 45 Prof. dr. J.G. Vos
- 46 Ir. H.J. van de Wiel
- 47-50 authors
- 51 SBC/afdeling Communicatie
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- 53 Bibliotheek RIVM
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- 60-120 Reserve-exemplaren