*Sakrapport till Naturvårdsverkets Miljöövervakning:*

# Intag av bromerade flamskyddsmedel hos barn i Sverige

| | |
|---|---|
| **Avtalsnummer:** | **215 0615** |
| **Utförare:** | **Livsmedelsverket** |
| **Programområde:** | **Hälsorelaterad miljöövervakning** |
| **Delprogram:** | **Exponering via livsmedel** |
| **Undersökningar/uppdrag:** | **Intagsberäkningar av bromerade flamskyddsmedel hos barn i Sverige** |

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**SAMMANFATTNING**

Human exponering för PBDE och HBCD sker främst via födan och speciellt via animaliska produkter då dessa ämnen är lipofila, bioackumulerande och ofta biomagnifierande. Studier från bland annat Sverige och Finland visar att fisk och skaldjur är den största källan till intag av PBDE. I den här studien har därför intaget av PBDE (9 kongener) och HBCD beräknats för barn i Sverige. I en rikstäckande kostundersökning utförd 2003 deltog barn i åldrarna 4, 8-9 och 11-12 år. De fick i en matdagbok ange sin konsumtion under fyra på varandra följande dagar. Data från denna undersökning kombinerades sedan med haltdata från olika livsmedel för att räkna ut intaget av PBDE och HBCD på individbasis. Undersökningen innefattade animaliska livsmedel såsom fisk och skaldjur, mejeriprodukter, köttprodukter, ägg, animaliskt och vegetabiliskt fett och fett från övriga livsmedel.

Resultaten visar att intaget av PBDE var 23 ng/dag, 31 ng/dag och 28 ng/dag för 4, 8-9 respektive 11-12 åringar. Intaget av HBCD beräknades till 7,9 ng/dag, 11 ng/dag och 9,5 ng/dag för 4, 8-9 respektive 11-12 åringar. Detta visar att 8-9 åringar har det högsta dagliga intaget av PBDE och HBCD. När intaget beräknas på kroppsviktsbasis har däremot de yngsta barnen det högsta intaget, som sedan sjunker med åldern. Fisk och skaldjur var den största källan till intaget av PBDE och HBCD, trots att konsumtionen av dessa livsmedel var relativt låg. Jämförelser med de fåtal studier som gjorts i andra länder tycks visa att svenska barn har ett lägre intag av PBDE och HBCD. Undersökningen tyder på att intaget av PBDE och HBCD hos svenska barn via livsmedel, utifrån de kunskaper vi har idag, inte utgör någon risk grundat på de effekter av PBDE och HBCD som observerats i toxikologiska studier.

*Report to the Swedish Environmental Protection Agency, 2008-10-06*

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# Dietary intake estimations of brominated flame retardants for Swedish children

## Introduction

The past use of BFRs has led to release into the environment and since these pollutants are persistent, they will remain in the environment for a long time (de Wit, 2002). Even though the development towards a more regulated market for PBDEs is positive, there is a concern regarding the debromination of the not yet restricted higher brominated BFRs to lower and possible more toxic BFRs (Stapleton et al., 2004). Furthermore, many of the products containing the now restricted flame retardants are still in use and often have a long lifetime. This will lead to a continued release of contaminants to the environment and further release in the future when these products are being disposed. Taken together, human exposure and the potential risk of BFRs are important issues to study.

Due to the lipophilic and persistent nature of the brominated flame retardants they can be detected in the environment, primarily in the aquatic environment (de Wit, 2002), where samples from biota, e.g. fish, and sediments have been shown to contain PBDE and HBCD (Ikonomou et al., 2002; Kierkegaard et al., 2004; Remberger et al., 2004).
The main exposure routes for human non-occupational exposure to PBDE and HBCD are through the diet and via ingestion of indoor dust and air (Covaci et al., 2006; Wijesekera, 2002). Children have a higher consumption of food relative their body weight and consequently have a higher intake of potential pollutants (Schecter et al., 2006). In Europe, the major contributor to the dietary intake of PBDE is generally fish (Lind et al., 2002). It has been established that a high intake of fish increases the levels of PBDEs in human breast milk and blood serum (Ohta et al., 2002; Sjodin et al., 2000).

Two market basket studies calculating the dietary intake of PBDEs for the adult population have been performed in Sweden in the last 10 years (Ankarberg et al., 2006; Darnerud et al., 2006). Darnerud et al. (2006) based their estimations on trade statistics from 1999 and the study included five PBDE congeners detected in fish, meat products, dairy products, eggs, fats and pastry. The medium-bound (MB) daily intake of PBDEs was estimated to 50.9 ng/day

(Darnerud et al., 2006). The second study, performed in 2005, included analysis of 9 congeners and the same food items as the first study with the exception of pastry. Although the intake was estimated on a larger number of congeners the result is almost identical as for the previous study; 50.6 ng/day (0.7 ng/kg bw/day, assuming a bodyweight of 73.7 kg). The five congeners included in the study from 1999 made up 79% of the sumPBDE in the study from 2005, indicating a decrease in the levels of PBDEs found in the food (Ankarberg et al., 2006). Fish was the largest contributor to the total intake in both studies making up 47% and 38% of the total intake, respectively. Market basket studies made in the adult populations in Belgium and Finland estimated the dietary intake of PBDE to 35 ng/day and 44 ng/day (MB) respectively (Kiviranta et al., 2004; Voorspoels et al., 2007). Also, these studies show that the food item contributing the most to the overall PBDE intake is fish and shellfish.

Lind et al. (2002) estimated the median daily intake of PBDEs in Sweden based on dietary surveys to 22.6 ng/day for women and 28.8 ng/day for men (MB) aged 21-30 years. The study included 5 congeners and was calculated on the consumption of fish, meat products, dairy products, fats and eggs. The major source for PBDE was fish with 55-60% of the total intake and the fatty fishes were the type of fish contributing the most (Lind et al., 2002).
In the UK, the weight-adjusted dietary intake of PBDEs have been estimated to 5.8 ng/kg bw/day (LB), based on dietary surveys (FSA, 2006). Even though the estimates are based on lower-bound data they are considerably higher than for the participants mentioned above in the Swedish studies, of which the men had a medium-bound estimated intake of 0.38 ng/kg bw/day (Lind et al., 2002). The UK study included 17 congeners and apart from the food items included in the Swedish studies, it also included cereals, vegetables, fruits, nuts, bread, sugars and preserves. Among the congeners included in the UK study, PBDE-209, which was not included in the Swedish studies, had the highest concentration in most of the food analyzed and was also the congener contributing the most to the total intake of PBDE. The food item contributing the most to the intake was meat, which also had, by far, the highest concentrations of PBDE (FSA, 2006).

There are only a few studies made on dietary intake of brominated flame retardants in children. In an American market basket study, the estimated intake for children between 2 and 11 years old was 1.8-2.7 ng/kg/day (sum of 13 congeners, MB) (Schecter et al., 2006). For Spanish children between 4 and 9 years old, the intake was approximately 2.6 ng/kg/day (sum of tetra-octaBDE) (LB) (Bocio et al., 2003). The congeners contributing the most to the

overall intake of PBDE was tetra- and pentaBDE in both studies. In addition to this, the American study found a high intake of decaBDE; this congener was not analyzed in the Spanish study. The food items included in these studies were fish, dairy products, meat products, eggs and fats. In addition, the Spanish study also included cereals, vegetables and fruits. The food group contributing the most to the PBDE intake was fish (33%) followed by fats (25%) and meat products (25%) for the Spanish children, and meat products (69%) followed by dairy products (16%) for the American children. In a study from the UK, the lower-bound intake of PBDEs was estimated to 11 ng/kg/day, 8.9 ng/kg/day and 6.7 ng/kg/day for the age groups 4-6, 7-11 and 11-14 years respectively. PBDE-209 was the congener contributing the most to the total intake of PBDE (FSA, 2006). The intake estimation from the UK is considerably higher than the estimations for American and Spanish children. This may, to some extent, be explained by the fact that the UK study analyzed a few more congeners than the other studies and it also included more food items. However, the additional congeners in the UK study were often below the quantification limit and therefore should not be the only reason for the high intake estimations. Among the food items not included in neither the American nor the Spanish study, sugars and preserves had the highest total concentration of PBDE and had the third highest contribution of PBDEs next after meat and fish in the UK study.

Only a few studies on the dietary intake of HBCD have been performed and the results indicate that fish, dairy products and fruit (!) are large contributors to the total dietary intake (Ankarberg et al., 2006; FSA, 2006). For adults, the intake has been estimated in a market basket study from Sweden and on individually based data from the Netherlands and the UK. In the Swedish market basket, fish, meat products, dairy products, eggs and fats were included. In the study from the Netherlands only fish were included and the UK study included fish, meat products, dairy products, eggs, fats, cereals, vegetables, fruits, nuts, bread, sugars and preserves. The intakes were estimated to 10.2 ng/day (MB), 8.3 ng/day (MB) and 5.9 ng/kg bw/day (UB) in Sweden, the Netherlands and the UK, respectively. In the Swedish study, fish was the food item that contributed the most to the overall intake (65%), the study from the Netherlands shows that the highest HBCD intake comes from herring and the UK study showed a high intake from fruit and dairy products (Ankarberg et al., 2006; van Leeuwen and de Boer, 2008; FSA, 2006).

The Food Standards Agency in the UK has, to date, done the only dietary estimations of

HBCD for children. This was done on the same set of food items as the UK estimations of PBDE and HBCD done for adults mentioned above. The age groups 4-6, 7-10 and 11-14 had an intake of 14 ng/kg bw/day, 9.4 ng/kg bw/day and 6.0 ng/kg bw/day (UB) respectively. Fruit and vegetables were the food items with the highest levels of HBCD (FSA, 2006).

The objective of this study

PBDEs and HBCD are detected in the environment and in food items; consequently it is evident that humans are exposed to these substances via the food. There are only a few estimations made on the dietary intake of PBDEs and HBCD in adults and reports on the dietary intake in children are even more scarce. There is consequently a lack of knowledge considering the magnitude of exposure, the kind of food contributing the most to the intake and the possible age and gender differences of dietary exposure to these BFRs, and especially HBCD. The objective of this study is therefore to estimate the dietary intake of PBDEs and HBCD in Swedish children and to compare the findings with those found in other studies.

**Materials and Methods**

The consumption data was derived from a national diet survey on children carried out in Sweden in 2003 (Riksmaten – barn 2003). It included 4 year olds (n = 590), 8-9 years olds (n = 909) and 11-12 year olds (n = 1036). The children's food intakes were registered in a food diary extending over four consecutive days distributed over the whole week (Enghardt Barbieri et al., 2006). The concentrations of contaminants in fish were obtained from the National Food Administration (NFA) Monitoring Programme (2001-2002), a fish survey (2000-2003) and from a market basket from 1999 (Ankarberg et al., 2007; Bjerselius et al., 2004; Darnerud et al., 2006). The concentrations in other food items were obtained from the NFA Monitoring Programme 2003-2007 and a market basket from 2005 (Ankarberg et al., 2006). The food items analyzed, number of samples and actual levels of PBDEs and HBCD used in this study are presented in table 2 and 3.

The food items included in this study are divided into 6 sub-groups: fish and shellfish, dairy products, meat products, eggs, animal and vegetable fats and fats from miscellaneous food products. The food items in the respective food groups are presented in table 1.

Table 1. The food groups and food items included in the study.

| **Food group** | **Food items** |
|---|---|
| Fish | fish-sticks, farmed salmon, wild salmon from the Baltic Sea, lean marine fish (e.g. cod, saithe, haddock), lean fish from freshwater lakes (e.g. perch, pike, zander, burbot), flatfish (e.g. turbot, plaice, flounder), canned tuna, herring/mackerel, Baltic herring/smoked herring, caviar and shellfish |
| Dairy products | Milk, yoghurt, sour milk, cream and cheese |
| Meat products | Meat from pork, beef and game, minced meat, sausages, liver pâté, offals and poultry |
| Eggs | Whole eggs and egg dishes |
| Animal and vegetable fats | Butter, margarine and ice-cream |
| Fat from miscellaneous products | Pizza, pie, pancakes, crêpes, pasty and waffles |

The analyses of the PBDE congeners (PBDE-28, PBDE-47, PBDE-66, PBDE-99, PBDE-100, PBDE-138, PBDE-153, PBDE-154 and PBDE-183) and HBCD were carried out at the NFA and described elsewhere (Atuma et al, 2000). Levels of PBDEs and HBCD in different food items are presented in table 2 and 3.

Table 2. Type of food analyzed, type of sample, levels of PBDEs and HBCD in the food samples and sampling year for the food items included in the survey. Values are given in µg/kg lw, except for sausages and eggs which are presented as µg/kg fresh weight. n = number of analyzed samples. Calculations were done assuming ND = ½ LOQ (MB).

| **Food item in the diet survey** | **Food sample analyzed** | **Single or pooled sample** (n) | **PBDE** (µg/kg lw) | | | | **HBCD** (µg/kg lw) | **Sample year** |
|---|---|---|---|---|---|---|---|---|
| | | | **PBDE-47** | **PBDE-99** | **PBDE-100** | **sumPBDE*** | | |
| Dairy products | Milk | P (9) | 0.10 | 0.05 | 0.04 | 0.42 | 0.11 | 2004-2005 |
| Pork | Pork | P (4) | 0.15 | 0.14 | 0.03 | 0.46 | 0.21 | 2003-2004 |
| Beef | Beef | P (4) | 0.06 | 0.03 | 0.03 | 0.26 | 0.11 | 2003-2004 |
| Game | Roe deer | P (2) | 0.39 | 0.61 | 0.08 | 1.47 | 0.39 | 2005 |
| Poultry | Chicken | P (6) | 0.18 | 0.15 | 0.03 | 0.51 | 0.13 | 2004 |
| Liver pâté and offals | Bovine liver | P (5) | 0.19 | 0.10 | 0.10 | 1.01 | 0.19 | 2005 |
| Fats | Butter, oil and margarine | P (13) | 0.06 | 0.07 | 0.03 | 0.31 | 0.07 | 2005 |
| Ice-cream | Butter and milk | P (15) | 0.09 | 0.05 | 0.03 | 0.37 | 0.09 | 2005 |
| Miscellaneous fats | Oil and margarine | P (7) | 0.06 | 0.09 | 0.02 | 0.32 | 0.09 | 2005 |
| | | | µg/kg fw | | | | | |
| Sausage | Meat mix | P (8) | 0.01 | 0.01 | 0.003 | 0.04 | 0.005 | 2005 |
| Eggs | Eggs | P (23) | 0.003 | 0.003 | 0.001 | 0.01 | 0.002 | 2004-2005 |

* sumPBDE include PBDE-28, PBDE-47, PBDE-66, PBDE-99, PBDE-100, PBDE-138, PBDE-153, PBDE-154 and PBDE-183.

Table 3. Type of fish analyzed, type of sample, levels of PBDEs and HBCD in the fish samples and sampling year for the fishes included in the survey. Values are given in µg/kg fw. n = number of analyzed samples. Calculations were done assuming ND = ½ LOQ (MB).

| **Fish in the diet survey** | **Fish sample analyzed** | **Single or pooled sample** (n) | **PBDE** (µg/kg fw) | | | | **HBCD** (µg/kg fw) | **Sample year** |
|---|---|---|---|---|---|---|---|---|
| | | | **PBDE-47** | **PBDE-99** | **PBDE-100** | **sumPBDE*** | | |
| Fish-sticks | Cod | P (1) | 0.11 | 0.004 | 0.02 | 0.16 | 0.04 | 2002 |
| Farmed salmon | Farmed salmon | S and P (3) | 1.15 | 0.21 | 0.19 | 2.00 | 0.66 | 2003 |
| Wild salmon | Wild salmon and trout from the Baltic Sea | S and P (32) | 2.00 | 0.38 | 0.40 | 3.14 | 1.89 | 2001-2002 |
| Lean marine fish | Cod | P (1) | 0.11 | 0.004 | 0.02 | 0.16 | 0.04 | 2002 |
| Flatfish | Turbot | P (5) | 0.25 | 0.03 | 0.05 | 0.41 | 0.06 | 2002 |
| Canned tuna | Cod and mackerel | P (2) | 0.80 | 0.21 | 0.15 | 1.43 | 0.55 | 2002 |
| Herring/mackerel | Herring and mackerel | P (12) | 1.21 | 0.29 | 0.22 | 2.01 | 1.65 | 2001-2002 |
| Baltic herring/ smoked herring | Baltic herring | P (26) | 1.80 | 0.25 | 0.40 | 2.71 | 1.23 | 2001-2002 |
| Caviar | Cod and herring | P (12) | 1.09 | 0.25 | 0.20 | 1.80 | 1.56 | 2001-2002 |
| Shellfish | Shrimp, crab and mussel | P (5) | 0.24 | 0.05 | 0.06 | 0.44 | 0.08 | 2002-2003 |
| Fish from freshwater lakes | Pike | S (1) | 0.35 | 0.05 | 0.08 | 0.59 | - | 1999 |

* sumPBDE include PBDE-28, PBDE-47, PBDE-66, PBDE-99, PBDE-100, PBDE-138, PBDE-153, PBDE-154 and PBDE-183 except for pike for which sumPBDE includes PBDE-47, PBDE-99, PBDE-100, PBDE-153, and PBDE-154.

The daily intake of PBDE-47, PBDE-99, PBDE-100, sumPBDE (9 congeners) and HBCD from each food item were calculated by combining the concentration of the contaminants in the food with the individual consumption data. When a congener were below the detection limit (not detected = ND) the concentration was assumed to be equal to zero (LB), one half of the quantification limit (MB) or equal to the concentration of the quantification limit (UB). Since the food diary did not specify what kind of meat, with the exception of poultry, the children consumed, it was assumed to be pork, beef and game, and to be equal to the meat consumption per capita in Sweden during 2003 (SJV, 2005). In the questionnaire it was not specified what kind of shellfish the children consumed. Therefore, statistics on the amount and type of shellfish consumed by the Swedish population were used to calculate the mean

concentration of the pollutants in shellfish (SJV, 1999). Of the eggs consumed in Sweden, 6% are organically produced and the levels in eggs have therefore been weighed. The concentrations of PBDEs and HBCD in Baltic herring varies considerably depending on where it is caught. In addition, it is often transported all over Sweden before it is consumed. Therefore the levels of contaminants were weighed according to the amount caught in different areas of the Baltic Sea. For a detailed description of the calculations mentioned, see Appendix 1.

**Results**

The results presented here are based on the median value and the medium-bound estimations of the daily intake of the pollutants. The daily consumption of food per kg bw decreased with age, being the highest for 4 year olds. Among the food groups studied, dairy products were the food group most consumed by children of all ages and meat the second most consumed, with a median consumption of 8.5-19 g/kg bw/day and 3.2-6.3 g/kg bw/day respectively. Eggs and fish/shellfish were the least consumed food groups, with a median consumption of 0 g/kg bw/day and 0.39-0.80 g/kg bw/day respectively (Fig. 1).

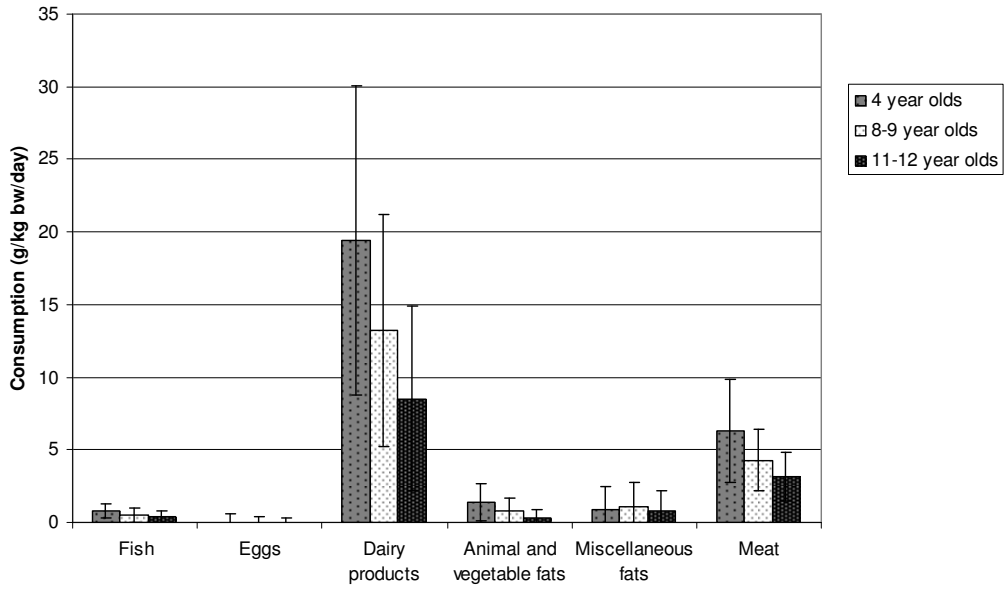


Figure 1. Daily food consumption (median ± SD) per kg bw for children in different age groups.

The most consumed fish was the group including lean marine fish, fish-sticks, flat fish and fish from freshwater lakes (lean fish), in which the two first mentioned were the most consumed within the group. The second most consumed fish group was non-Baltic fatty fish, of which farmed salmon was the most consumed (Fig. 2).

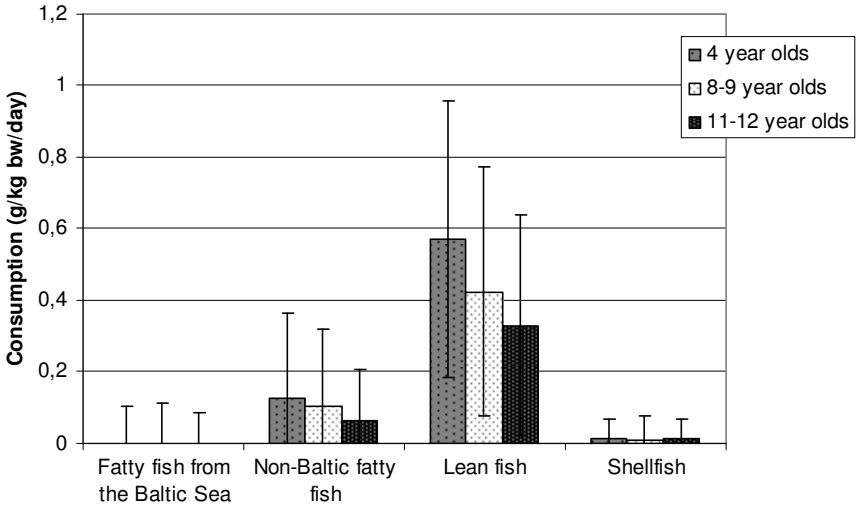


Figure 2. Daily consumption of fish (median ± SD) per kg bw for children in different age groups.

The estimated medium bound-intake of sumPBDE was 23 ng/day, 31 ng/day and 28 ng/day and for HBCD it was 7.9 ng/day, 11 ng/day and 9.5 ng/day for 4, 8-9 and 11-12 year olds respectively. 8-9 year olds have the highest individual daily intake of PBDE and HBCD compared to the other age groups (Kruskal-Wallis, Dunn´s; $p< 0.0001$). However, when comparing the intake based on estimated daily intake per kg bw and day, the intake decrease with age (Table 4 & 5 and Fig. 3) giving the highest intake in 4 year olds (Kruskal-Wallis, Dunn´s; $p< 0.0001$).

Table 4. Estimated daily intake of sumPBDE (9), per individual or per kg bw (in brackets) in ng/day and ng/kg bw/day.

| | Lower-bound | Medium-bound | Upper-bound |
|---|---|---|---|
| 4 years | 11.6 (0.63) | 23.0 (1.28[b]) | 36.7 (2.03) |
| Girls | 11.5 (0.62) | 22.3 (1.28) | 35.8 (2.02) |
| Boys | 11.7 (0.63) | 23.9 (1.29) | 38.2 (2.05) |
| | | | |
| 8-9 years | 17.0 (0.56) | 30.9[a] (1.00) | 48.1 (1.47) |
| Girls | 17.0 (0.55) | 29.8 (1.00) | 45.8 (1.42) |
| Boys | 17.0 (0.56) | 32.5 (1.04) | 51.0 (1.50) |
| | | | |
| 11-12 years | 14.8 (0.36) | 27.7 (0.68) | 44.2 (1.05) |
| Girls | 13.8 (0.33) | 25.5 (0.60) | 40.1 (0.93) |
| Boys | 16.0 (0.39) | 29.5 (0.73) | 47.6 (1.12) |

[a]Statistically different from 4-year-old and 11-12-year-old children.
[b]Statistically different from 8-9-year-old and 11-12-year-old children.

Table 5. Estimated daily intake of HBCD, per individual or per kg bw (in brackets) in ng/day and ng/kg bw/day.

| | Lower-bound | Medium-bound | Upper-bound |
|---|---|---|---|
| 4 years | 5.78 (0.31) | 7.94 (0.44[b]) | 9.76 (0.54) |
| Girls | 5.66 (0.31) | 7.59 (0.42) | 9.33 (0.52) |
| Boys | 5.91 (0.32) | 8.22 (0.45) | 10.1 (0.56) |
| | | | |
| 8-9 years | 8.12 (0.26) | 10.7[a] (0.35) | 13.1 (0.39) |
| Girls | 7.91 (0.26) | 10.6 (0.34) | 12.4 (0.38) |
| Boys | 8.27 (0.27) | 11.0 (0.36) | 13.6 (0.40) |
| | | | |
| 11-12 years | 7.40 (0.18) | 9.46 (0.23) | 11.4 (0.27) |
| Girls | 6.63 (0.16) | 8.74 (0.21) | 10.4 (0.24) |
| Boys | 7.77 (0.19) | 10.3 (0.25) | 12.3 (0.29) |

[a]Statistically different from 4-year-old and 11-12-year-old children.
[b]Statistically different from 8-9-year-old and 11-12-year-old children.

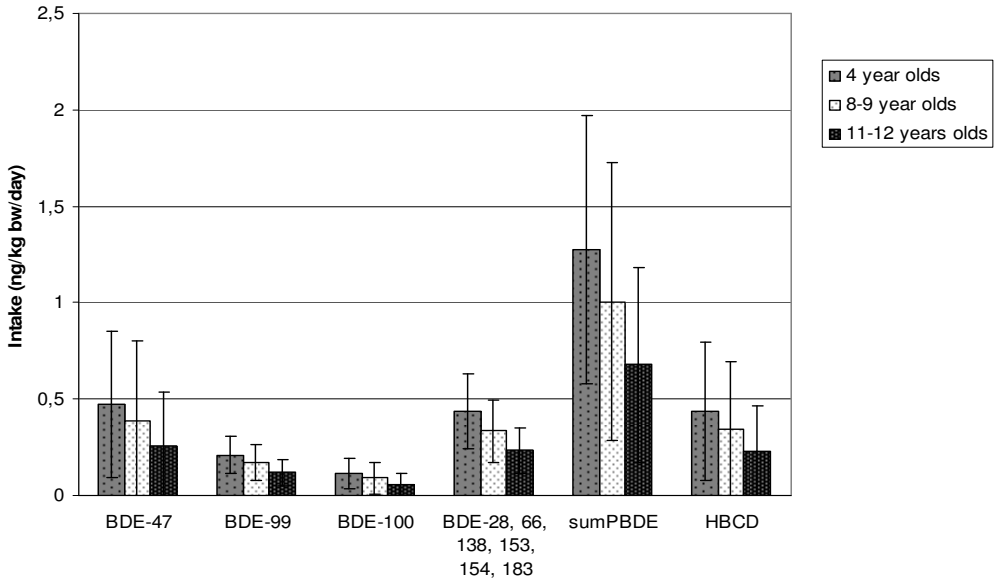


Figure 3. Daily dietary intake (median ± SD) of PBDEs and HBCD per kg bw for children in different age groups.

PBDE-47 contributed the most to sumPBDE intake in all age groups; this was also the congener most common in the analyzed food items. Boys often had a somewhat higher intake of sumPBDE and HBCD compared to girls, and these gender differences in intake where significant in the 8 year-old and 11 year-old groups (Mann-Whitney, $p< 0.01$ for sumPBDE despite of age group; $p<0,05$ for HBCD in 8 year olds and $p< 0.0001$ for 11 year olds). The contribution from different food groups to the total intake of PBDEs and HBCD was similar between the age groups. Fish contributed the most to the overall sumPBDE and HBCD intake

in all ages, with a 37-41% contribution for sumPBDE and 45-49% contribution for HBCD. There was also a large contribution from meat, which increased with age from 31% to 36% and 28% to 36% of the intake of PBDEs and HBCD respectively. The contribution from dairy products, which were the third largest contributor, was higher for 4 year olds compared with other age groups.

Among the fish consumed, the highest intake of sumPBDE and HBCD came from non-Baltic fatty fish, of which farmed salmon was the largest contributor. The contribution from non-Baltic fatty fish to the total intake from fish was 71-74% and 82-86% of sumPBDE and HBCD respectively. Lean fish (lean marine fish, fish from freshwater lakes, flatfish and fish-sticks) was the second largest contributor to the total fish intake with 24-25% and 13-16% of sumPBDE and HBCD respectively. Based on median intakes, there was no contribution from Baltic fatty fish in any age group, even though a few of the children had a large intake of PBDEs and HBCD from these fishes.

**Discussion**

PBDEs and HBCD are persistent, bioaccumulative and magnify through the food chains. Consequently, there is a risk for human exposure through the food. PBDEs and HBCD are considered to have low acute toxicity (Darnerud, 2003), but effects after long-term or repeated exposure are not that well explored. Extensive dietary intake estimations of PBDEs are scarce, and even more so regarding HBCD. In addition, there is a lack of studies including children, one of the most sensitive groups in the population. It is therefore urgent to examine the extent of the exposure and the potential risks with dietary intake of PBDE and HBCD in children. When comparing with other countries, such as Spain, the United Kingdom and the United States, the Swedish children have the lowest intake of both PBDEs and HBCD. One reason for the lower intake in Sweden might be the well established dietary advice, without which the Swedish population probably would have a higher intake of PBDEs and HBCD, as well as other pollutants, than they have now.

Dairy products were the food items most consumed by the children, yet it was only the third largest contributor to the intake of PBDEs and HBCD. Fish was the food group contributing the most to the intake of these contaminants even though it was the second least consumed food group. The most consumed fish was the group of “lean fish” (lean marine fish, fish from freshwater lakes, flatfish and fish-sticks), of which the contribution to the total intake of

PBDEs and HBCD from fish was approximately 25% and 14%. Non-Baltic fatty fish was, by far, the fish contributing the most to the intake of PBDEs and HBCD among the different fish types. Within this group, farmed salmon was both the most consumed and the fish contributing the most to the intake of PBDEs and HBCD. Fatty fish as a dominating contributor to the intake of PBDEs and HBCD is consistent with the lipophilic properties of the pollutants.

Based on median levels there were no contribution to the intake of PBDEs and HBCD from fish from the Baltic Sea (mean consumption: 0.7-1.0 g/day). However, there are children with a high consumption of this type of fish and it often leads to a high intake of PBDEs and HBCD. A few of the children in the study have a high consumption of salmon from the Baltic Sea. This fish is very rare on the market and it is unlikely that all the Baltic salmon the children reported to have consumed actually is this type of fish. The 5% of the children with the highest intake of PBDEs and HBCD often stated a high consumption of fish in the food questionnaire and many of them often had a high consumption of fish from the Baltic Sea. However, a high intake of PBDEs and HBCD was not always explained by a high consumption of fish from the Baltic Sea. Children with a high intake of PBDEs and HBCD often had a high consumption of other fish and/or meat, as well. The contribution of fish to the total intake of PBDEs and HBCD is consistent with other Swedish and European intake estimations who also found fish to be the largest contributor, in both children and adults (e.g. (Ankarberg et al., 2006; Bocio et al., 2003; Kiviranta et al., 2004)). However, a study from the UK reached other conclusions on the contribution of food groups to the intake of PBDE and HBCD. They found meat to be the largest contributor to the intake of PBDE and milk and fruits to be the largest contributor to the intake of HBCD in adults (FSA, 2006). Unfortunately the UK study does not state the food contributing the most to the total intake of PBDE or HBCD in children. However, the concentrations of PBDE in meat is higher than in any of the other food items, which explains the UK results. The same argument applies for the concentration of HBCD in milk and fruit. In the present study, dairy products is the third largest food group contributing to the total intake of HBCD.

A large proportion of the food items analyzed in the UK study had a considerable contribution of PBDE-209, which was the congener contributing the most to the total intake of PBDEs. Meat had especially high levels of PBDE-209 and the including of this congener in the UK

study may be an explanation for both the overall high intake of PBDE and the fact that meat was the largest contributor (FSA, 2006).

In other European countries, PBDE-47 is the congener contributing the most to the total intake of PBDEs (e.g. (Ankarberg et al., 2006; Kiviranta et al., 2004; Voorspoels et al., 2007). This was also the case in this study, where fish is the main reason for the high contribution of PBDE-47. PBDE-209 is seldom included in dietary intake estimations, probably because of difficulties when analyzing and detecting it. This makes it difficult to asses how reliable the contribution of this particular congener is in e.g. the UK study. Nevertheless, it is becoming more important to include this congener since it is the only congener not legally restricted in the EU and consequently the environmental levels of the commercial mixture including PBDE-209 may not follow the trends of the restricted commercial mixtures. Moreover, as the UK results show, PBDE-209 may have a great impact on the total intake of PBDEs.

The children in the present study have a higher intake of PBDEs and HBCD estimated per kg bw/day than the adults participating in earlier Swedish studies (Fig. 7). These results were expected since the children have a higher consumption of food relative their body weight compared to adults, leading to a higher intake of pollutants. It is also evident that the intake of PBDEs is lower at childbearing ages and increases slightly in the Swedish population after the age of 50. One reason for this may be the less strict dietary advice for women over child-bearing age and men, or because of differences in food habits between generations.

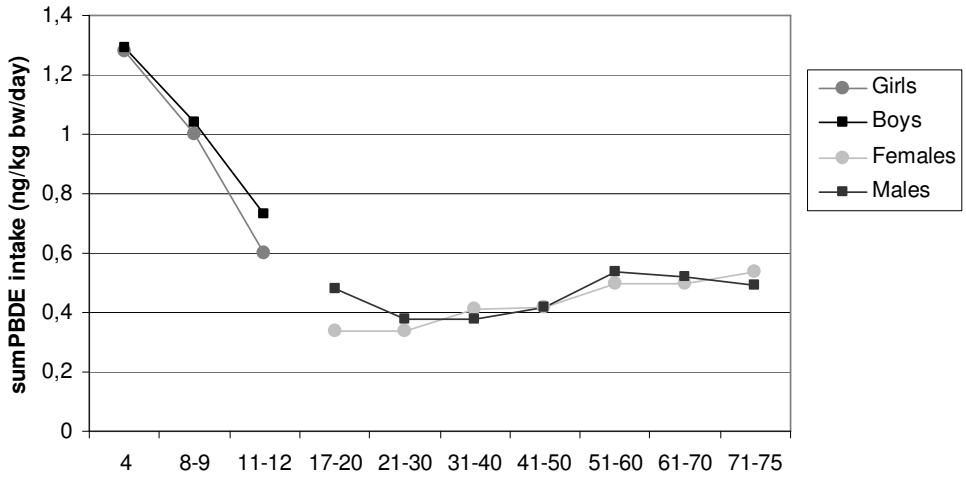


Fig 7. The median dietary intake (ng/kg bw/day) of ΣPBDE in men and women at different ages. Calculations were done assuming ND = 0,5 (MB). Results from two studies (present study and intake estimations for the adult population (Lind et al. 2002)).

There is a lack of epidemiology information regarding the effects of PBDE and HBCD, making it difficult to asses the possible risk with exposure of these substances. The information available is mainly the results of animal studies. The most sensitive end-point after exposure to PBDE are reductions in sperm count detected in rats at 60 μg/kg bw and neurodevelopmental effects, which occured at 60 μg/kg bw in rats and at 0.8 mg/kg bw in mice (Eriksson et al., 2001; Kuriyama et al., 2005). After exposure to HBCD, the most sensitive end-point is an increase in thyroid weight, visible at a BMDL of 3.4 mg/kg bw/day (van der Ven et al.,2006).

In this intake estimation, the 4-year-old children had the highest median intake of PBDEs and HBCD per kg bw and day, 1.28 ng/kg bw/day and 0.44 ng/kg bw/day respectively, which is lower than the doses mentioned for the effects on the nervous and reproductive systems. Even the children with the highest intake of PBDEs and HBCD are well below the levels where toxic effects have been shown. The intake of PBDEs is however in the same range as the levels found in breast milk from mothers to newborns with cryptorchidism, which was 3.03 ng/g lipids (Main et al., 2007).

The results of this dietary intake estimation indicate that the intake of PBDE and HBCD is not high enough to concern any risk, at least not when considering the exposure of one of these substances alone. However, we are exposed to more than one pollutant at a time which render a high total intake of pollutants and may lower the levels at which toxic effects of PBDEs and HBCD are seen. One end-point that might concern the children in this study is the time of entering puberty. Toxicity tests have shown a delay of the onset of puberty in rats (Stoker et al., 2004). Although the effects were shown at high doses of PBDEs and the risk is very low in children with the intake of PBDEs presented in this study, it might be a risk when exposed to several pollutants at the same time during repeated exposure.

Also, the highly brominated decaBDE product is not yet restricted and it is likely to debrominate in the environment causing the formation of lower brominated products (Stapleton et al., 2004). It is therefore necessary to continue with the monitoring of PBDEs in the environment, in humans and in food. To refine the dietary intake estimations in the future they would preferably include higher brominated PBDE congeners, such as BDE-209, and other food items not often included in these types of estimations, such as fruits and vegetables.

The lack of studies including HBCD, not only dietary estimations but also toxicity studies and the monitoring of levels in humans, makes it difficult to asses the extent of exposure and toxicity of HBCD. There is a need for further studies on the effects of HBCD and of the magnitude of exposure.

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Appendix 1

**Weighing of data for Baltic herring**

The levels of the pollutants in herring from different areas in the Baltic Sea were multiplied with the amount caught fish in the respective areas. These were then added up and divided by the total amount caught in the entire Baltic Sea (see equation and the following example).

$$\frac{(\textit{concentration x amount caught fish in area } 1) + (\textit{concentration x amount caught fish in area } 2)...}{\textit{total amount caught in the Baltic Sea}}$$

sumPBDE in Baltic herring:

$$\frac{(1{,}11 \times 140) + (4{,}05 \times 1800) + (3{,}25 \times 3700) + (1{,}31 \times 3000)}{8640} = 2{,}71\,\mu\text{g/kg fw}$$

**Weighing of data for shellfish**

The concentrations of the pollutants in different shellfish species were multiplied with the daily intake of respective species. These were added up and then divided by the daily intake of all shellfish (see equation and the following example).

$$\frac{(\textit{concentration in spicies } 1 \times \textit{amount consumed}) + (\textit{concentration in spicies } 2 \times \textit{amount consumed})...}{\textit{total amount consumed}}$$

sumPBDE in shellfish:

$$\frac{(0{,}22 \times 5{,}8) + (3{,}01 \times 0{,}6) + (0{,}16 \times 1{,}1)}{7{,}5} = 0{,}43\,\mu\text{g/kg fw}$$

**Weighing of data for egg**

The concentrations of the pollutants in organic eggs were multiplied with percentage organic eggs consumed and the pollutants in conventional eggs were multiplied with the percentage conventional eggs consumed. These were added and then divided by 2 (see the following example).

sumPBDE in egg:

$$\frac{(0{,}19 \times 0{,}06) + (0{,}11 \times 0{,}94)}{2} = 0{,}06\,\mu\text{g/kg fw}$$