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# Occurrence of selected halogenated flame retardants in Belgian foodstuff

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**Abstract**

This paper reports on the occurrence of halogenated flame retardants (HFRs), namely PBDEs, HBCDs, TBBPA, brominated phenols (BrPhs), dechlorane plus (DP) and emerging FRs in a variety of Belgian foodstuffs. A total of 183 composite food samples were analyzed by GC-MS and LC-MS/MS techniques for the presence of HFRs. The analyses revealed that 72% of the samples was contaminated with HFRs to some extent. The highest number of contaminated samples was observed within the group 'Potatoes and derived products', 'Fish and fish products' and 'Meat and meat products', while the least contaminated group was 'Food for infants and small children'. The total HFR content ranged from <LOQ to 35.4 ng/g ww with an average content of 1.2 ng/g ww and median of 0.25 ng/g ww. The samples with the highest total HFR levels were canned king crab, fresh mackerel, Emmental cheese, fresh eel and plaice. The most frequently detected HFRs were PBDEs and BrPhs being present in almost all food groups, and among the individual HFRs, the most frequently found compounds were BDE-47 (53%), BDE-209 (46%) and 246-TBP (40%). TBBPA, DPs, TBPH and γ-HBCD occurred with a frequency of less than 5%. TBBPS, 26-DBP, HBB, TBB and BTBPE were not detected in any of the analyzed food samples.

**Keywords**

Brominated flame retardants; Dechloranes; PBDEs; HBCDs; food analysis; occurrence

## 1. Introduction

Flame retardants (FRs) are a diverse group of chemicals added to a wide range of consumer products, including plastics, polymers, textiles, building materials, and electric and electronic equipment, to prevent or delay the propagation of fire (Kodavanti and Loganathan 2016). Currently, there are four major groups of flame retardants on the market: inorganic, halogenated organic, organophosphorus, and nitrogen based compounds. Brominated flame retardants (BFRs; a subgroup of the halogenated organic class) are currently the largest market group of flame retardants due to their low cost and high-performance efficiency. The main BFRs are polybrominated diphenyl ethers (PBDEs), hexabromocyclododecane (HBCDs), tetrabromobisphenol A (TBBPA) and brominated phenols (BrPhs).

PBDEs have become widespread environmental pollutants because they can leach out of the consumer products (Covaci et al. 2011). Due to concerns about their persistent, bioaccumulative and toxic properties (Birnbaum & Staskal 2004; De Jourdan et al. 2013), legal restrictions have been imposed upon their use and production (Directive 2003/11/EC; Directive 2002/95/EC; Commission Decision 2005/717/EC). However, despite the restrictive measurements on their use, PBDEs have been detected in various food products for human consumption, such as fish (Covaci et al. 2005; Eljarrat et al. 2007; Voorspoels et al. 2007; Roosens et al. 2008), meat products (Darnerud et al. 2006; Voorspoels et al. 2007) and eggs (Pirard & De Pauw 2007; Voorspoels et al. 2007; Covaci et al. 2009). The technical mixture/commercial formulation of HBCD contains three isomers, α-HBCD, β-HBCD and γ-HBCD, and is used as additive flame retardant in polystyrene, textiles, electronics and plastic materials. Due to its wide use and chemical persistence, it occurs in different environmental matrices and may accumulate in the food web (Janak et al. 2005; Janak et al., 2008; Leslie et al., 2011; Li et al., 2011; Koch et al. 2015; Letcher et al. 2015; Lyche et al. 2015). HBCDs have been detected in wild-caught and farmed fish, seafood and eggs (Janak et al. 2005; Covaci et al. 2009; Rawn et al. 2009; Schechter et al. 2010; Goemans et al. 2007). Exposure to HBCDs can have effects on liver hormones and might induce neurobehavioral alterations (Deng

et al. 2009; Feng et al. 2013; Cruz et al. 2015). TBBPA, the most commercially relevant BFR (Abdallah 2014; Cruz et al. 2015), is used as a reactive BFR and is covalently bonded to plastic, thus limiting its release to the environment compared to the additive BFRs (de Wit 2002). However, several studies have reported its presence in the indoor and outdoor environment (Sjödin et al. 2001; Tollbäck et al. 2006; Abdallah et al. 2008; Harrad et al. 2010; Abafe & Martincigh 2016). Effects of TBBPA on thyroid hormones, neurological function, and reproduction have been shown (Chan & Chan 2012; Huang et al. 2013). BrPhs, such as 2,4-dibromophenol (24-DBP) and 2,4,6-tribromophenol (246-TBP), are used as reactive FRs in epoxy, phenolic and polyester resins, polyolefins and vinyl-aromatic polymers (EFSA 2012a). Furthermore, it has been shown that a number of BrPhs occur naturally in different marine organisms (Chung et al. 2003a,b; EFSA, 2012a; Vetter & Janussen, 2005), BrPhs are not generally readily biodegradable and will persist in the environment (EFSA 2012a; Howe et al. 2005).

The reduction in the use of PBDEs and HBCDs has consequently opened the way for emerging flame retardants (EFRs) to reach the market (Covaci et al. 2011). However, several of these EFRs raised concerns about their persistence, bioaccumulation, long-range transport and toxicity (DiGangi et al. 2010), and almost no information is available on their occurrence in food. It is known that human exposure to BFRs varies widely throughout the world as it depends on their country-related usage, production and legislation (Roosens et al. 2010). Therefore, since dietary intake is known to be one of the main routes of human exposure to BFRs, together with inhalation of indoor air and ingestion of (indoor) dust (Bakker et al. 2008; Harrad et al. 2008; Roosens et al. 2010; Domingo 2012), the lack of data on the presence of BFRs in food can lead to a consequent incorrect estimation of the health risks. In several reports dedicated to different classes of BFRs, the European Food Safety Authority (EFSA 2011a,b; EFSA 2012a,b) has indicated that it is not possible to perform an accurate risk assessment due to the lack of data on the occurrence in food and consequently on the exposure to BFRs *via* diet. For this reason, the EU prepared a

Recommendation on the monitoring of BFRs in foodstuffs (Commission Recommendation 2014/118/EU).

The main objective of this study was to respond to this Recommendation, to contribute filling the gap on the occurrence data of BFRs in food. Fish and seafood, and to a lesser extent food of animal origin (meat and dairy products), are estimated as the main contributors to the dietary intake of PBDEs and HBCDs for the Belgian population (Voorspoels et al. 2007; Roosens et al. 2010; Goscinny et al. 2011). Thus, the present study aimed to assess the presence and levels of PBDEs, EFRs (including bis(2-ethylhexyl) tetrabromophthalate (TBPH), 2-ethylhexyl 2,3,4,5-tetrabromobenzoate (TBB), bis-tribromophenoxy-ethane (BTBPE), hexabromobenzene (HBB)), HBCDs (α-HBCD, β-HBCD, γ-HBCD), TBBPA, BrPhs and derivatives (including, 4-bromophenol (4-BP), 24-DBP, 2,6-dibromophenol (26-DBP), 246-TBP, 2,4,6-tribromoanisole (TBA), tetrabromobisphenol S (TBBPS)) and dechlorane plus (DPs, *syn*- and *anti*- isomers) in 183 composite food samples belonging to the above-mentioned food groups and to other relevant food categories purchased in Belgium (e.g., eggs, grains, fats and oils, vegetables and potatoes, food for infants, etc.). Several of these BFRs have persistent organic pollutant (POP)-like properties and are known for environmental and health issues.

## 2. Materials and Methods

### *2.1 Sampling and pooling of the food items*

Individual food items (1,289 samples) were purchased in Belgian supermarkets, discount retailers and some specialized meat and fish stores during 2015-2016. Only the edible parts were kept (*e.g.*, no bones or fish heads) and marinated sauces and other condiments were discarded from the prepared samples (*e.g.*, vinegar for marinated fishes and oils from canned foods). The individual samples were ground and homogenized, and equal amounts (±75 g) of the food items belonging to the same food category were pooled to create a unique composite sample. Fish-based food supplements in gelatin capsules were purchased and mixed to generate one composite sample

without the gelatin coating. The composite samples were either freeze-dried, analyzed fresh (*e.g.* for samples with high fat content, or containing emulsifiers), or directly aliquoted for analysis (dry foodstuffs such as pasta and dry mashed potatoes). In total, 183 composite food samples were prepared and categorized as 'Fish and fish products' (n=61 samples, including fish (FC), crustaceans (CC) and molluscs (MC)), 'Meat and meat products' (MEC; n=35), 'Milk and dairy products' (n=38, including liquid milk (LC), desserts (DC) and cheese (CHC)), 'Food for infants and small children' (BC; n=18), 'Animal and vegetable fat' (OC/FAT-C; n=9), 'Grains and grain-based products' (GRAC; n=7), 'Eggs and egg products' (EGC; n=4), 'Potatoes and derived products' (POC; n=4), 'Other food' (OTC: stock: n=4, food supplements: n=1; vegetables (VEC): n=2). Pending analysis, the samples were stored at -20°C in Falcon tubes (50 mL) previously tested and found to be free from the targeted HFRs.

Further details on the composite samples are given in Supporting Information, Table S1. Additionally, the table reports the type of preservation (frozen, canned, etc.), the number of individual food items pooled into the corresponding composite samples, and the lipid content (%) of the composite samples. For simplicity and to enhance readability, we will refer to samples as composite samples in the text. No distinction between locally produced and imported food was made in this study.

*2.2 Sample preparation and instrumental analysis*

Due to the diverse nature of the targeted HFRs, two analytical methods, based on gas chromatography in combination with electron capture negative ionization mass spectrometry (GC-ECNI/MS) and ultra-high performance liquid chromatography-tandem mass spectrometry (UHPLC-MS/MS), were developed, in-house validated (Poma et al. 2016, Malysheva et al. 2017), and applied to the analysis of food samples. Information on the chemicals and materials used in the study is reported in Supplementary Data, section SD-1.

*2.2.1 GC-ECNI/MS analysis*

Sample preparation and instrumental details are fully reported in Poma et al. (2016). In brief, depending on the type of tissue and the lipid content of the food samples, 2 g, 1 g or 0.2 g (for non-fatty matrices, low fatty matrices, and high fatty matrices, respectively) was weighed, spiked with 50 µL of internal standard (IS) mixture (including BDE-103, BDE-128, $^{13}C_{12}$-BDE-209, $^{13}$C-TBPH, $^{13}$C-TBB, $^{13}$C-DP-s, and $^{13}$C-DP-a), and added with 5 mL of acetonitrile (ACN):toluene (9:1, *v:v*). After vortexing, the sample was loaded on empty SPE cartridges (25 mL) with a frit at the bottom and the eluting solvent was collected in 15 mL pre-cleaned glass tubes. The extract was evaporated to dryness, reconstituted in 0.5 mL hexane (Hex), loaded on a 3 mL Florisil® cartridge (pre-conditioned with 6 mL of Hex), and eluted with 6 mL of Hex:dichloromethane (DCM) (4:1, *v:v*) and 6 mL of Hex:DCM (1:1, *v:v*). The extract was evaporated to dryness, reconstituted in 0.5 mL Hex, loaded on a 6 mL cartridge containing 1.5 g of acidified silica (AS, 5%, *w/w*, prewashed with 6 mL Hex) for further clean-up, and eluted with 12 mL of Hex:DCM (1:1, *v:v*). The final extract was evaporated to near dryness, reconstituted in 100 µL of the recovery standard (RS) (CB-207 in *iso*-octane:toluene; 9:1, *v/v*) and transferred to amber injection vials for GC-ECNI/MS analysis.

The analysis of GC amenable compounds was performed with an Agilent 6890 GC (Palo Alto, CA, USA) coupled to an Agilent 5973 MS operated in ECNI mode and equipped with a DB-5 capillary column (15 m × 0.25 mm × 0.10 µm) (J&W Scientific, Folsom, CA, USA), electronic pressure control, and a programmable-temperature vaporizer (PTV) inlet. The mass spectrometer was operated in selected ion monitoring (SIM) for the quantification of all the analytes of interest (BDE-28, -47, -49, -99, -100, -138, -153, -154, -183, -209, TBPH, TBB, BTBPE, HBB, TBA, DPs). BDE-103 and BDE-128 were used as IS for all PBDE congeners (except BDE-209), TBA, HBB, and BTBPE. $^{13}$C-BDE-209 was used as IS for BDE-209. $^{13}$C-TBB, $^{13}$C-TBPH, $^{13}$C-DP-s, and $^{13}$C-DP-a, were used as IS for TBB, TBPH, DP-s, and DP-a, respectively.

*2.2.2 UHPLC-MS/MS analysis*

Details on sample preparation and instrumental settings are fully reported in Malysheva et al. (2017). In brief, for the analysis of TBBPS, 2.5 g of a lyophilized sample or oil was extracted with 15 mL of ACN:HCOOH (9:1, v/v), and after centrifugation 5 mL of supernatant was transferred into a tube containing 1 g EMR-Lipid activated beforehand with 5 mL $H_2O$. The tube was vortexed for 1 min and centrifuged for 10 min at 4500 g. The supernatant was transferred into a tube containing 1 g EMR-Lipid Polish salts and 25 mg carbon. After centrifugation, 4 mL of the upper phase was evaporated till dryness under a stream of nitrogen at 40 °C. The residue was reconstituted in 120 µL ACN. For the analysis of BrPhs (except 246-TBA), TBBPA and HBCDs, 2.5 g lyophilized sample (or 1 g oil) was extracted with 15 mL of Hex:DCM (1:1, v/v) and after centrifugation 6 mL of supernatant was reduced till approx. 2 mL and cleaned-up using 8 g AS (44%, *w:w*) packed in an empty SPE cartridge. The cartridge was conditioned with 15 mL Hex and the BFRs were eluted with 25 mL DCM. The eluate was evaporated till near dryness under a stream of nitrogen at 40 °C. The solvent was exchanged to ACN until a total volume of 100 µL.

The UHPLC-MS/MS system consisted of an ACQUITY UPLC system coupled to a Xevo-TQ-S mass spectrometer (Waters, Milford, MA, USA) operated in negative electrospray ionization ESI(-) mode. Data acquisition was performed in Multiple Reaction Monitoring (MRM) mode. Chromatographic separation was achieved using an ACQUITY UPLC™ BEH C18 column (2.1 × 100 mm, 1.7 µm) with an ACQUITY UPLC™ BEH C18 VanGuard pre-column (2.1 × 5 mm, 1.7 µm) (Waters) at 45 °C. A mobile phase consisting of eluents A [$H_2O$:ACN (95:5, *v:v*) containing 0.1% $CH_3COOH$] and B [ACN:$H_2O$ (95:5, *v:v*) containing 0.1% $CH_3COOH$] was used at a flow rate of 0.3 mL/min. A gradient elution was applied as follows: 0–1.5 min, 50%; 1.5–2.0 min, 10% A; 2.0–5.5 min, 10% A; 5.5–5.6 min, 50% A; 5.6–8.5 min, 50% A.

*2.3 Quality assurance and quality control (QA/QC)*

Several quality assurance/control measures were applied to evaluate the accuracy and reliability of the measurements. Two procedural blanks were analyzed in each analytical batch of ten composite

food samples. Blank contribution was considered constant (RSD <30%) and therefore results are blank corrected by subtraction of mean blank values from the values found in the samples. In addition, a value equal to 3*SD of the blank measurement was used as cut-off before reporting results, hereby assuring 99% confidence that any reported value was not confounded by a blank contribution. Limits of quantification (LOQs) were calculated as three times the standard deviation of blank values and divided by the amount of sample used for analysis. For compounds not detected in the blanks, the LOQ was calculated based on the signal to noise ratio of 10/1, considering also the chromatogram's characteristics for the respective retention times (e.g. co-elution, noisy baseline). The following LOQs were achieved: 5 pg/g ww for PBDEs (except for BDE-209), 100 pg/g ww for BDE-209, 200 pg/g ww for TBPH and TBB, 50 pg/g ww for TBA, and 20 pg/g ww for HBB and DPs, 10 pg/g ww for BTBPE, 0.01-8 ng/g ww for the other BrPhs, 0.2-1.8 ng/g ww for TBBPS, 0.01-0.25 ng/g ww for TBBPA, and 0.004-1 ng/g ww for HBCDs. The mean recoveries of the spiked IS ranged from 68 to 123%, while the mean expanded uncertainty of the methods ranged between 3 and 37%. Further details on the methods' performance can be found in Poma et al. (2016) and Malysheva et al. (2017).

*2.4. Statistical analysis*

For data treatment, statistical software IBM SPSS Statistics 24 was used. Because of the high frequency of non-detects, the mean and median levels of FRs were calculated with the non-detects set to zero (<LOQ = 0, i.e. lower bound, LB) in order to avoid overestimation.

## 3. Results and discussion

### *3.1. Distribution of the total HFR content and frequency of contamination of the food groups*

The GC-MS and LC-MS/MS analyses revealed that 72% of food composites were contaminated with at least one HFR. Fig. 1 provides an overview on the frequency of contamination for each food group. The highest number of contaminated samples (100%) was observed in the group 'Potatoes

and derived products', followed by 'Fish and fish products' (90%), 'Meat and meat products' (89%) and 'Other food' (86%). However, the total number of samples per group has to be taken into account in evaluation of this frequency. For instance, compared to the group 'Fish and fish products' (n=61), the group 'Potatoes and derived products' was comprised of considerably lower number of samples (n=4). The lowest number of contaminated samples were found in the 'Food for infants and small children' group with frequency of 11% (2 out of 18 samples containing $HFR_{total}$ of <290 pg/g ww).

The total content of the targeted HFRs in the investigated samples ranged from <LOQ to 35.4 ng/g ww with an average content of 1.2 ng/g ww and a median of 0.25 ng/g ww. Considering the sum of all targeted HRFs in this study, the samples with the highest levels were the following: canned king crab (CC-01; 35.4 ng/g ww) > fresh mackerel (FC-52; 22.8 ng/g ww) > Emmental cheese (CHC-20; 17.2 ng/g ww) > fresh eel (FC-56; 11.5 ng/g ww) > plaice (FC-55; 11.4 ng/g ww). Overall, the highest number of the targeted HFRs (regardless the level) was observed in fresh oysters (MC-01) > marinated trout (FC-10) = frozen sole (FC-41) > smoked mackerel (FC-16) = fresh mackerel (FC-52) = fresh eel (FC-56).

The food group 'Fish and fish products' was characterized by the presence of samples with the highest maximum levels of all groups (Fig. 2). The $HFR_{total}$ content in 'Meat and meat products' ranged from <LOQ to 9.8 ng/g ww with an LB mean of 0.8 ng/g ww and a median of 0.6 ng/g ww. The most contaminated sample of this group was a composite sample of salami (MEC-04), mostly due to the abundant presence of BDE-209 (9.2 ng/g ww). A number of HFRs (mostly BDE-47, BDE-209, TBA and 246-TBP) were quantifiable in samples from the group 'Milk and dairy products' being mostly detected in cheese and desserts. The content ranged from <LOQ to 17.2 ng/g ww with an LB mean of 0.7 ng/g ww and a median of 6 pg/g ww. Interestingly, the second highest LB mean (1.4 ng/g ww) was noted in 'Eggs and egg products' which was mainly attributed to the presence of α-HBCD (3.8 ng/g ww) in one of the samples. The lowest average $HFR_{total}$ content was observed in 'Food for infants and small children' (0.02 ng/g ww) and 'Animal and

vegetable fat' (0.2 ng/g ww). The median for the different food groups was not higher than 0.7 ng/g ww with the exception of 'Food for infants and small children' and 'Animal and vegetable fat' with a median of <LOQ.

Contribution of each HFR to the contaminated food composites is presented in Fig. 1. From this figure it is obvious that the most frequently detected HFR groups were PBDEs and BrPhs being present in all food groups, except for 'Animal and vegetable fat', in which PBDEs were not found. 'Meat and meat products', 'Fish and fish products' and 'Milk and dairy' were the groups with all or most of the targeted HFR groups being represented. Among the individual HFRs, the most frequently detected compound was BDE-47 (53%), followed by BDE-209 (46%) and 246-TBP (40%) (data not shown). TBBPA, DPs, TBPH and γ-HBCD were detected with a frequency of less than 5%. TBBPS, 26-DBP, HBB, TBB and BTBPE were not detected in any of the analyzed food samples.

In the following sections the quantitative data are discussed with regards to occurrence of the different HFR groups in the analyzed food composites, and overview of the descriptive statistics is provided in Table 1.

### *3.2. Occurrence of the different HFR groups*

#### *3.2.1. Occurrence of PBDEs*

The measured levels of individual PBDE congeners for all composite food samples are reported in Supplementary Data, Table S2. PBDEs were the most frequently detected compounds in most composite food samples (detection frequency 57%) (Fig. 1), with lower bound (LB) based mean levels for sum PBDEs ranging from 17 ± 61 pg/g ww in 'Food for infants and small children' to 1,029 ± 3,564 pg/g ww in the cheese category, and with total values for sum PBDEs up to 16,888 pg/g ww (firm-cheese emmental) (Table 1, Table S2).

The FC category was characterized by LB mean levels of 433 ± 928 pg/g ww. Surimi (FC-07) was the least contaminated seafood item (5 pg/g ww), while fresh eel (FC-56) had the highest total PBDE content (5,727 pg/g ww) (Table S2). The higher levels of PBDEs in eel samples (equal to 31 ng/g lipid weight (lw)) are similar to those observed in eels from the Elbe river, Germany (34 ± 13 ng/g lw) and the Saint Lawrence River, Canada (23 ng/g lw) (Sühring et al. 2014), and generally lower than the PBDE levels observed in eels from Belgium (1.2 – 140 ng/g lw) (Malarvannan et al. 2014), Lake Ontario, Canada (77 ng/g lw) (Sühring et al. 2014), and from the Gironde estuary system (24 – 220 ng/g lw) (Tapie et al. 2011). BDE-47 was the most representative congener (41%) of the mean contamination in the composite fish/seafood samples (mean LB level of 179 pg/g ww), followed by BDE-209 (32%), BDE-100 (12%), BDE-49 (7%), BDE-28 (4%), BDE-154 (2%), and BDE-153 and -183 (1%). The PBDE levels measured in FC samples are comparable with other studies in Europe (Kiviranta et al. 2004; Darnerud et al. 2006; Voorspoels et al. 2007; Törnkvist et al. 2011; Xu et al. 2015), Asia (Nguyen et al. 2014), USA (Schecter et al. 2008; Schecter et al. 2010a; Schecter, et al. 2010b), and worldwide (Domingo 2012). It was not possible to provide reliable quantitative results for BDE-99 due to the co-elution with $^{13}$C-TBB.

Differently from the FC category, the CHC, MEC, and POC samples were mainly affected by the presence of BDE-209 (between 95 and 100%) as the dominant congener in these samples, possibly due to a contamination during the production chain, sample handling (before and/or after purchasing), or extraction process in the laboratory. Noteworthy is the highest level of BDE-209 in firm-cheese emmental (CHC-20, 16,839 pg/g ww), and the presence of BDE-28 (from 6 to 265 pg/g ww) in 54% of the samples in MEC category. For the latter case, this is probably due to an interfering compound co-eluting with BDE-28. BDE-209 was also found in all potato samples (POC), with levels between 119 and 1213 pg/g ww, while BDE-47 was found (14 pg/g ww) only in paprika chips (POC-02). BDE-47 and BDE-209 were the only congeners found in OTC samples, ranging from 9 to 26 pg/g ww and from 209 to 557 pg/g ww, respectively.

Regarding the other analyzed food categories, BDE-47 and BDE-209 were the only congeners detected in food for infants and small children, eggs, grains, and other food. In 'Food for infants and small children' only two samples showed PBDE levels above the LOQ. In baby biscuits (BC-05), 254 pg/g ww BDE-209 was measured (maybe due to the use of contaminated fats during their production), and in powdered soy formula (BC-07) 56 pg/g ww BDE-47 was found. All other BC products had values <LOQ for PBDEs.

The low levels of PBDEs found in baby food is assuring, as infants are the most vulnerable subgroup of the population to contaminants like PBDEs (Lyche et al. 2015; Linares et al. 2015). These values are similar to those measured by Liu et al. (2014), who found total PBDE levels in baby food to be <100 pg/g ww in most analyzed samples, with BDE-47 and BDE-209 as the most frequently detected congeners.

In the 'Eggs and egg products', BDE-47 (21 pg/g ww) and BDE-209 (267 pg/g ww) were measured in quail eggs (EGC-01), while only BDE-209 (559 pg/g ww) was found in organic eggs (EGC-02). In 'Grains and grain products', BDE-47 and BDE-209 were measured in croissant and similar pastry (GRAC-02) with values of 81 pg/g ww and 1,603 pg/g ww, respectively, and only BDE-209 was measured in loaf bread (GRAC-03, 157 pg/g ww) and fresh white bread (GRAC-5, 403 pg/g ww). The prevalence of BDE-47 and BDE-209, among the considered PBDE congeners, in several food samples (such as fish/seafood, meat, eggs) resembles the composition of the commercial mixtures, even though the manufacture of these chemicals has ceased in many countries, underlining the persistence of these contaminants.

The CC category did not contain any BDE congeners above the LOQ, mostly due to the low lipid content of these aquatic organisms and their low trophic level in the food chain. Low levels of PBDEs were measured also in shrimps (19.6 pg/g w; Domingo et al. 2008) and crustaceans (20 pg/g ww; Bodin et al. 2007), whereas a more recent study in Korean foodstuff reported higher levels in (cooked) shrimps (116 pg/g ww; Nguyen et al. 2014). This can be due to differences in sample

origins (e.g. geographical location and collection sites) and proximity to eventual contamination sources, with varying levels of environmental PBDE contamination.

No PBDEs were detected in OC/FAT-C, VEC, and LC categories either.

Recently, action limits for ∑PBDEs in different foodstuffs were proposed by the Scientific Committee of the Belgian Federal Agency for the Safety of the Food Chain (FASFC) (Scientific Committee of the FASFC 2017). The proposed action limits, calculated by dividing the toxicological reference value of the compounds by the 97.5$^{th}$ percentile of consumption of the concerned foodstuffs (Scientific Committee of the FASFC 2017), are listed in Table 2. The ∑PBDEs levels found in the current study were therefore compared to the FASFC action limits and were found to be much lower than the proposed values in all cases. It has to be noted that our study was not designed for market control since composite samples were used. Nevertheless, the measured level of 64 ng/g lw for ∑PBDEs in a composite sample of Emmental cheese (CHC-20) was above the action limit for this food group (40 ng/g lw), but the measurement uncertainty associated with the result (50%) should be taken into account.

*3.2.2. Occurrence of EFRs and DPs*

The measured levels of the EFRs for all composite food samples are given in Supplementary Data, Table S2. TBPH was only found in two samples from the vegetable oil category, with values of 295 pg/g ww in mixed oils (OC-04) and 350 pg/g ww in olive oil (OC-01), and in one sample from the 'Meat and meat products' (MEC-11), with value of 202 pg/g ww in duck foie gras. In this case, as TBPH was present in detectable amounts in the procedural blanks, it cannot be excluded that these values resulted from activities related to the laboratory analysis, rather than from a real contamination of the samples.

The other targeted EFRs (HBB, TBB, BTBPE) were <LOQ in all analyzed food items. Liquid milk, desserts, 'Food for infants and small children', crustaceans, and 'Potatoes and derived products' composite samples showed non-quantifiable levels of EFRs.

344 DPs were measured only in firm cheese emmental (CHC-20, 68 pg/g ww *syn*-DP and 271 pg/g ww
345 *anti*-DP), quail eggs (EGC-01, 130 pg/g ww *syn*-DP and 507 pg/g ww *anti*-DP), and boudin noir
346 pork (MEC-16, 62 pg/g ww *syn*-DP and 269 pg/g ww *anti*-DP). Existing literature data on the
347 levels of DPs in wild bird eggs showed higher (1.5 - 15 ng/g ww, Gauthier et al. 2007; 2.2 – 21 ng/g
348 ww, Su et al. 2015) or similar (0.2 – 1.8 ng/g ww, Chen et al. 2012) values of DPs than/to our total
349 DP measurements in quail eggs. In addition, it is reported in literature that the DP isomers in the
350 technical product are present in a ratio (*syn*/*anti*) of about 0.3, with the *anti*-isomer representing
351 about 75% of the total (Sverko et al. 2008; Sverko et al. 2011). In the analyzed food samples, the
352 ratio *syn*/*anti*-isomers was 0.25, 0.26 and 0.21 for cheese, egg and meat samples respectively, with
353 the *anti*-isomer representing about 80% of the total, comparable to the reported literature data.
354
355 *3.2.3. Occurrence of HBCDs and TBBPA*
356 The levels of HBCDs and TBBPA measured in the composite food samples are reported in
357 Supplementary Data, Table S2. In all food samples, HBCDs (the sum) were detected in the range
358 from <LOQ to 5.5 ng/g ww with an average content of 76 pg/g ww and median of <LOQ. Of the
359 three HBCD isomers, α-HBCD was the most frequently detected and was most predominant in
360 'Fish and fish products', with the highest level of 5.3 ng/g ww in fresh eel (FC-56). The prevalence
361 of α-HBCD over the other HBCDs in food was also observed in most of other studies (EFSA
362 2011b; Fernandes et al. 2016; Goscinny et al. 2011; Schecter et al. 2012) and the possible reasons
363 for that were summarized in Malarvannan et al. (2014). The current results for HBCDs in this food
364 group are comparable to those obtained in Belgium in 2008 except for eel, which was not sampled
365 at that time (Goscinny et al. 2011). The HBCDs' levels in eel found in this study were similar to
366 those measured in pooled eel samples originating from sites located in rural areas along the river
367 Scheldt in Belgium (Malarvannan et al. 2014). Among the different categories in the group 'Fish
368 and fish products', HBCDs were detected only in one CC (frozen scampi: γ-HBCD at 30 pg/g ww)
369 and one MC sample (fresh oysters: all three HBCDs detected in the range 13-239 pg/g ww). As for

370 FC, the frequency of detection was 22% (of all FC samples positive for HFRs) and the range for
371 positive samples for ∑HBCDs was from 66 pg/g ww (frozen sole) to 5.5 ng/g ww (fresh eel).
372 The total HBCD content in 'Meat and meat products' ranged from <LOQ to 613 pg/g ww with an
373 LB mean of 43 pg/g ww (Table 1). The highest levels for ∑HBCDs were found in chorizo (613
374 pg/g ww) and salami (403 pg/g ww).
375 Only one sample of 'Milk and dairy products' contained HBCDs, namely β-HBCD at 5 pg/g ww
376 (LC-07: fresh low-fat cow milk). Contrary to these results, in the Belgian study in 2008 the only
377 HBCD detected in dairy products was γ-HBCD (Goscinny et al. 2011).
378 Interestingly, a relatively high level of α-HBCD (3.8 ng/g ww) was measured in organic eggs,
379 bringing the LB mean level for this compound 8 to 23 times higher than in 'Fish and fish products'
380 (124 pg/g ww) and 'Meat and meat products' (42 pg/g ww), respectively. The remaining egg
381 samples (non-organic chicken and quail eggs) did not contain HBCDs above LOQ. Such difference
382 in contamination can probably be because animals raised in organic farms usually spend more time
383 outside compared to animals raised in other systems and consequently their exposure to
384 environmental contaminants is higher. Few years ago, similar HBCD levels were found in home-
385 produced eggs from free-foraging chickens of Belgian private owners (Covaci et al. 2009).
386 However, no HBCDs were detected in eggs in another study in Belgium in 2008 (Goscinny et al.
387 2011). Remarkably, α-HBCD (487 pg/g ww) was detected in the sample of fish-based food
388 supplement (OTC-05 from the group 'Other food') and the fact that this product is derived from
389 fish, which often contains this BFR, can possibly explain this result. The HBCDs were previously
390 reported in fish-based food supplements from Canadian, Japanese and British markets (Kakimoto et
391 al. 2008; Rawn et al. 2010; UK Food Standards Agency 2006). The levels of HBCDs in 'Food for
392 infants and small children', 'Grains and grain products', 'Potatoes and derived products' and
393 'Animal and vegetable fat' were below LOQ (Table 1, Table S2).
394 As regards co-occurrence of the HBCD isomers, α-HBCD co-occurred with β-HBCD only in 15%
395 of cases, and these composites belonged to FC, MC, MEC and EGC categories. The co-occurrence

396 of all three HBCDs was observed only in samples of eel (FC-56), trout (FC-57) and oysters (MC-
397 01).
398 As for ∑PBDEs, action limits for ∑HBCDs in different foodstuffs were proposed by the Scientific
399 Committee of the Belgian FASFC (Scientific Committee of the FASFC 2017) (Table 2). In this
400 study, the ∑HBCDs content did not exceed the proposed values.
401 TBBPA had low frequency of occurrence (2%) in the composite food samples which is consistent
402 with the previously reported results, and was associated with the lower bioaccumulation potential of
403 TBBPA (Morris et al. 2004; de Boer & Wells 2006; Fernandes et al. 2016). TBBPA was detected
404 only in 'Meat and meat products' with an LB mean of 3 pg/g ww (Table 1), and the contaminated
405 samples were turkey (96 pg/g ww), fish sticks (39 pg/g ww) and canned king crabs (8 pg/g ww).
406
407 *3.2.4. Occurrence of BrPhs and derivatives*
408 The levels of BrPhs and derivatives measured in the composite food samples are provided in
409 Supplementary Data, Table S2. Total content of BrPhs and their derivatives varied by food group,
410 ranging from <LOQ to 35.4 ng/g ww with an LB mean of 730 pg/g ww and median of <LOQ. The
411 most contaminated sample was canned king crab (CC-01), followed by fresh grey shrimps (FC-52;
412 22.2 ng/g ww) and fresh plaice (FC-55; 11.3 ng/g ww). Both 26-DBP and TBBPS were not detected
413 in any of the analyzed food groups.
414 246-TBP was the most frequently detected BrPh (63% of samples positive for BrPhs) with some
415 food groups being contaminated only with this BrPh, namely 'Food for infants and small children'
416 (BC-05, baby biscuits: 27 pg/g ww), 'Potatoes and derived products' (POC-02, paprika chips: 156
417 pg/g ww) and 'Animal and vegetable fat' (up to 318 pg/g ww). Following 246-TBP, TBA occurred
418 in 43% of samples positive for BrPhs, with LB mean values ranging from 15 ± 52 pg/g ww (MEC)
419 to 358 ± 865 pg/g ww (FC). Among the different food categories, TBA was most frequently
420 occurring in FC and MC samples. The presence of high levels of TBA in composite fish/seafood
421 and mussel samples (up to 4.2 ng/g ww in fresh salmon (FC-01) and 255 pg/g ww in fresh mussels

(MC-02)) can be explained by the fact that this compound is naturally produced by algae, bacteria, fungi and sponges in the marine environment (Moore et al. 2002; Vetter et al. 2005; Bidleman et al. 2016). In addition, as fresh salmon (FC-01) and smoked salmon from Ireland (FC-02) contained more than 4 ng/g ww of TBA (4.2 and 4.0 ng/g ww, respectively) (Table S2), this fish species showed to be highly affected by the presence of TBA. However, because lower TBA levels were measured in smoked salmon from Norway (FC-03, 508 pg/g ww) and canned salmon (FC-04, 457 pg/g ww), and no specific information related to the area/country of origin of FC-01 and FC-04 is available, it is difficult to speculate further.

Although detection frequency of 4-BP was lower, this BrPh occurred overall at much higher levels (LB mean: 441 ng/g ww) compared to 246-TBP (LB mean: 63 ng/g ww) (data not shown). The levels of 4-BP in positive samples varied from 164 pg/g ww (FC-18, canned tuna) to 23 ng/g ww (CC-01, canned king crab). Only 23% of the samples in group ‘Meat and meat products’ contained BrPhs, and the content ranged from <LOQ to 826 pg/g ww with a LB mean of 77 pg/g ww and median <LOQ (Table 1). The detected BrPhs were TBA, 246-TBP and 4-BP. Overall, the BrPhs were mostly detected in processed meat products and liver with maximum levels reaching 710 pg/g ww in duck foie gras (MEC-11) and 440 pg/g ww in fresh pork chipolata sausage (MEC-18), both for 4-BP. Within the group ‘Milk and dairy products’, the CHC category contained only TBA, 246-TBP and 24-DBP at levels up to 373 pg/g ww, while no BrPhs were measured in the LC category. The groups ‘Grains and grain products’ and ‘Other food’ contained only 246-TBP and TBA at levels <200 pg/g ww. TBA was the only BrPh found in the samples of the group ‘Eggs and egg products’ at levels <100 pg/g ww in special Omega-3 eggs and organic chicken eggs. The lower levels of TBA measured in cheese, meat stocks, eggs, cereals, and meat (up to 252 pg/g ww in salami) could be likely resulted from activities related to the sample processing prior analysis, industrial production, and/or laboratory analysis (sample handling, its presence in the laboratory environment), rather than from a real contamination of the samples. It is worth mentioning that a number of EFRs might degrade into more bioavailable compounds such as 246-TBP, therefore

some BrPhs found in the environment could, in part, result from degradation processes (EFSA 2012a). Furthermore, several brominated aromatic compounds, such as 4-BP, 24-DBP, 26-DBP and 246-TBP, occur naturally in a number of different marine organisms (Chung et al. 2003a,b; EFSA 2012a; Gribble 2009; Vetter & Janussen 2005). Thus, contamination of food with these BrPhs can be of natural origin and not related to their industrial use of as FRs.

## 4. Conclusions

This study reports on occurrence of the different HFRs, including those specified in the Commission Recommendation 2014/118/EU, in Belgian foodstuffs. The study contributes to providing HFR occurrence data for, among others, the less investigated food groups as well as providing occurrence figures not only for the more frequently analyzed HFRs, as PBDEs and HBCDs, but also for EFRs, for which data are scarce.

The HFR levels that were measured in this work were generally consistent with the available literature data for food. The high frequency of contamination and the highest maximum HFR levels were found in 'Fish and fish products' which is consistent with previously reported data, and can be explained by the physico-chemical properties of the HFRs targeted in this study, their environmental behavior and their bioaccumulation potential. It is worth mentioning that 'Food for infants and small children' has very low frequency of contamination, and only few HFRs were detected in this group at low levels. This is a positive observation, as infants are the most vulnerable subgroup of the population. The most frequently detected HFRs were PBDEs and BrPhs, the latter result can possibly be affected by natural production of BrPhs by the aquatic organisms. HBCDs were detected less frequently.

Overall, these results demonstrate that certain Belgian foodstuffs remain contaminated with such HFRs as PBDEs that were banned in the EU for several years, and HBCDs, on the use of which restrictions have been applied. However, the PBDE and HBCD content measured in the study were below the action limits proposed by the Belgian FASFC for these contaminants. While this study

expands the database on HFRs in food, it is reasonable to continue surveillance on their occurrence in this matrix, since over the last years no declining trend in the PBDE and HBCD levels could be clearly observed. Further it is advisable to conduct further research to assess differences in HFR contamination of food farmed or produced in different regions of the world, as well as the effect of food packaging on HFR levels.

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**Conflicts of interest**

The authors have no conflicts of interest to declare.

**Supplementary data**

Supplementary data can be found in the online version of the article.

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756

**List of abbreviations**

| | |
|---|---|
| 246-TBP | 2,4,6-Tribromophenol |
| 24-DBP | 2,4-Dibromophenol |
| 26-DBP | 2,6-Dibromophenol |
| 4-BP | 4-Bromophenol |
| ACN | Acetonitrile |
| AL | Action limit |
| AS | Acidified silica |
| BC | Food for infants and small children |
| BFRs | Brominated flame retardants |
| BrPhs | Brominated phenols |
| BTBPE | Bis-tribromophenoxy-ethane |
| CC | Crustaceans |
| CHC | Cheese |
| DC | Desserts |
| DCM | Dichloromethane |
| DP | Dechlorane plus |
| DP-a | Dechlorane plus *anti*-isomer |
| DP-s | Dechlorane plus *syn*-isomer |
| EFRs | Emerging flame retardants |
| EFSA | European Food Safety Authority |
| EGC | Eggs and egg products |
| EMR | Enhanced Matrix Removal |
| ESI | Electrospray ionization |
| EU | European Union |
| FASFC | Federal Agency for the Safety of the Food Chain |
| FC | Fish category |
| FRs | Flame retardants |
| GC-ECNI/MS | Electron capture negative ionization mass spectrometry |
| GRAC | Grains and grain-based products |
| HBB | Hexabromobenzene |
| HBCDs | Hexabromocyclododecane |
| Hex | Hexane |
| HFRs | Halogenated flame retardants |
| IS | Internal standard |
| IQ | Interquartile |
| LB | Lower bound |
| LC | Liquid milk |
| LOQ | Limit of quantification |
| MC | Molluscs |
| MEC | Meat and meat products |
| MRM | Multiple Reaction Monitoring |
| OC/FAT-C | Animal and vegetable fat |
| OTC | Other food |
| PBDEs | Polybrominated diphenyl ethers |
| POC | Potatoes and derived products |
| POP | Persistent organic pollutant |
| PTV | Programmable-temperature vaporizer |
| QA | Quality assurance |
| QC | Quality control |

| | |
|---|---|
| RS | Recovery standard |
| RSD | Relative standard deviation |
| SD | Standard deviation |
| SIM | Selected ion monitoring |
| SPE | Solid Phase Extraction |
| TBA | 2,4,6-Tribromoanisole |
| TBB | 2-Ethylhexyl 2,3,4,5-tetrabromobenzoate |
| TBBPA | Tetrabromobisphenol A |
| TBBPS | Tetrabromobisphenol S |
| TBPH | Bis(2-ethylhexyl) tetrabromophthalate |
| UHPLC-MS/MS | Ultra-high performance liquid chromatography-tandem mass spectrometry |
| VEC | Vegetables |
| ww | Wet weight |

**Figure captions**

**Fig. 1.** Contribution of each HFR to the contaminated samples (a) and frequency of contamination of the analyzed composite food samples (b).

**Fig. 2**. Distribution of the total HFR content in the different food groups.

*For visibility reasons an exponent-transformed scale (exponent=0.5) is applied to the y-axis of both graphs. Outliers with values between 1.5 and 3 times the interquartile (IQ) range are indicated with a circle, while extremes with values more than 3 times the IQ range are indicated with a star.*

**Table 1.** Descriptive statistics for the HFR content in composite food samples. Concentrations are expressed in pg/g ww[a].

| HFRs[b] | All samples | Food group | | | | | | | | |
|---|---|---|---|---|---|---|---|---|---|---|
| | | Fish and fish products | Meat and meat products | Milk and dairy products | Eggs and egg products | Food for infants and small children | Animal and vegetable fat | Grains and grain products | Potatoes and derived products | Other food |
| **∑PBDEs** | | | | | | | | | | |
| Mean | 76 | 370 | 689 | 601 | 212 | 17 | ND | 321 | 591 | 182 |
| SD[c] | 650 | 861 | 1536 | 2733 | 268 | 61 | | 620 | 494 | 250 |
| Median | 0 | 91 | 347 | 0 | 144 | 0 | | 0 | 515 | 0 |
| Maximum | 16889 | 5727 | 9171 | 16889 | 559 | 254 | | 1684 | 1213 | 567 |
| Minimum | 0 | 0 | 0 | 0 | 0 | 0 | | 0 | 119 | 0 |
| 25th percentile | 0 | 2.5 | 23 | 0 | 0 | 0 | | 0 | 160 | 0 |
| 75th percentile | 0 | 366 | 651 | 226 | 491 | 0 | | 403 | 1097 | 487 |
| 90th percentile | 56 | 726 | 1165 | 790 | * | 76 | | * | * | * |
| **TBPH** | | | | | | | | | | |
| Mean | 2 | ND[d] | 6 | ND | ND | ND | 72 | ND | ND | ND |
| SD | 23 | | 34 | | | | 143 | | | |
| Median | 0 | | 0 | | | | 0 | | | |
| Maximum | 350 | | 202 | | | | 350 | | | |
| Minimum | 0 | | 0 | | | | 0 | | | |
| 75th percentile | 0 | | 0 | | | | 148 | | | |
| 90th percentile | 0 | | 0 | | | | * | | | |
| **∑DPs** | | | | | | | | | | |
| Mean | 5 | ND | 10 | 9 | 159 | ND | ND | ND | ND | ND |
| SD | 44 | | 61 | 55 | 319 | | | | | |
| Median | 0 | | 0 | 0 | 0 | | | | | |
| Maximum | 637 | | 358 | 339 | 637 | | | | | |
| Minimum | 0 | | 0 | 0 | 0 | | | | | |
| 25th percentile | 0 | | 0 | 0 | 0 | | | | | |

| | | | | | | | | | | |
|---|---|---|---|---|---|---|---|---|---|---|
| 75th percentile | 0 | | 0 | 0 | 478 | | | | | |
| 90th percentile | 0 | | 0 | 0 | * | | | | | |
| **∑HBCDs** | | | | | | | | | | |
| Mean | 38 | 131 | 43 | 0.1 | 971 | | | | | 70 |
| SD | 352 | 707 | 128 | 0.8 | 1943 | | | | | 184 |
| Median | 0 | 0 | 0 | 0 | 0 | | | | | 0 |
| Maximum | 5516 | 5516 | 613 | 5 | 3885 | ND | ND | ND | ND | 487 |
| Minimum | 0 | 0 | 0 | 0 | 0 | | | | | 0 |
| 25th percentile | 0 | 0 | 0 | 0 | 0 | | | | | 0 |
| 75th percentile | 0 | 0 | 0 | 0 | 2914 | | | | | 0 |
| 90th percentile | 0 | 204 | 184 | 0 | * | | | | | * |
| **TBBPA** | | | | | | | | | | |
| Mean | 0.8 | | 3 | | | | | | | |
| SD | 8 | | 16 | | | | | | | |
| Median | 0 | | 0 | | | | | | | |
| Maximum | 96 | ND | 96 | ND | ND | ND | ND | ND | ND | ND |
| Minimum | 0 | | 0 | | | | | | | |
| 25th percentile | 0 | | 0 | | | | | | | |
| 75th percentile | 0 | | 0 | | | | | | | |
| 90th percentile | 0 | | 0 | | | | | | | |
| **∑BrPhs and derivatives** | | | | | | | | | | |
| Mean | 208 | 2070 | 77 | 62 | 33 | 1.5 | 118 | 45 | 39 | 72 |
| SD | 1595 | 5457 | 184 | 134 | 39 | 6.4 | 142 | 77 | 78 | 75 |
| Median | 0 | 402 | 0 | 0 | 28 | 0 | 0 | 0 | 0 | 52 |
| Maximum | 35390 | 35390 | 826 | 662 | 76 | 27 | 318 | 175 | 156 | 197 |
| Minimum | 0 | 0 | 0 | 0 | 0 | 0 | 0 | 0 | 0 | 0 |
| 25th percentile | 0 | 8.5 | 0 | 0 | 0 | 0 | 0 | 0 | 0 | 0 |
| 75th percentile | 0 | 1603 | 0 | 72 | 71 | 0 | 259 | 138 | 117 | 138 |
| 90th percentile | 156 | 4162 | 399 | 254 | * | 2.7 | * | * | * | * |

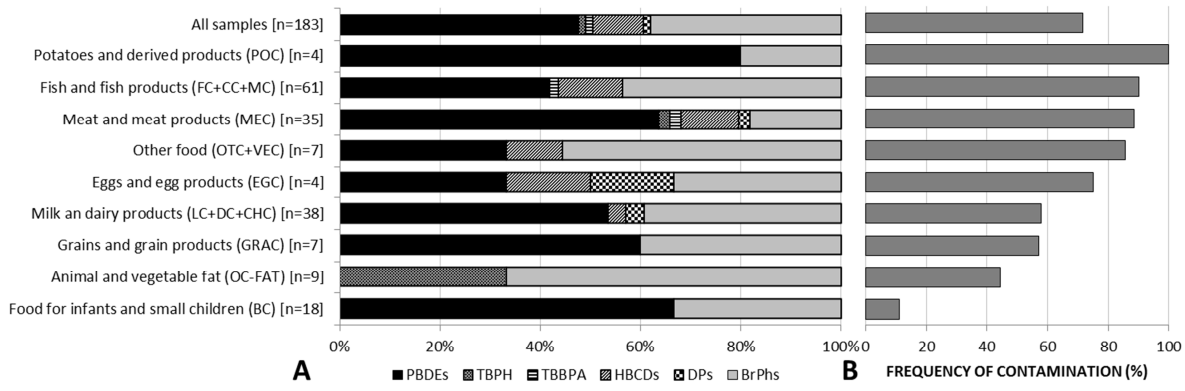
[a]Samples >LOQ as well as samples <LOQ were included
[b]HFR groups were made as follows: ∑PBDEs includes BDE-28, -47, -49, -99, -100, -138, -153, -154, -183, -209; ∑DPs includes syn-DP and anti-DP; ∑HBCDs includes α-HBCD, β-HBCD, γ-HBCD; ∑BrPhs and derivatives includes 4-BP, 24-DBP, 26-DBP, 246-TBP, 246-TBA, TBBPS
[c]SD: standard deviation
[d]ND: not detected
*not reported

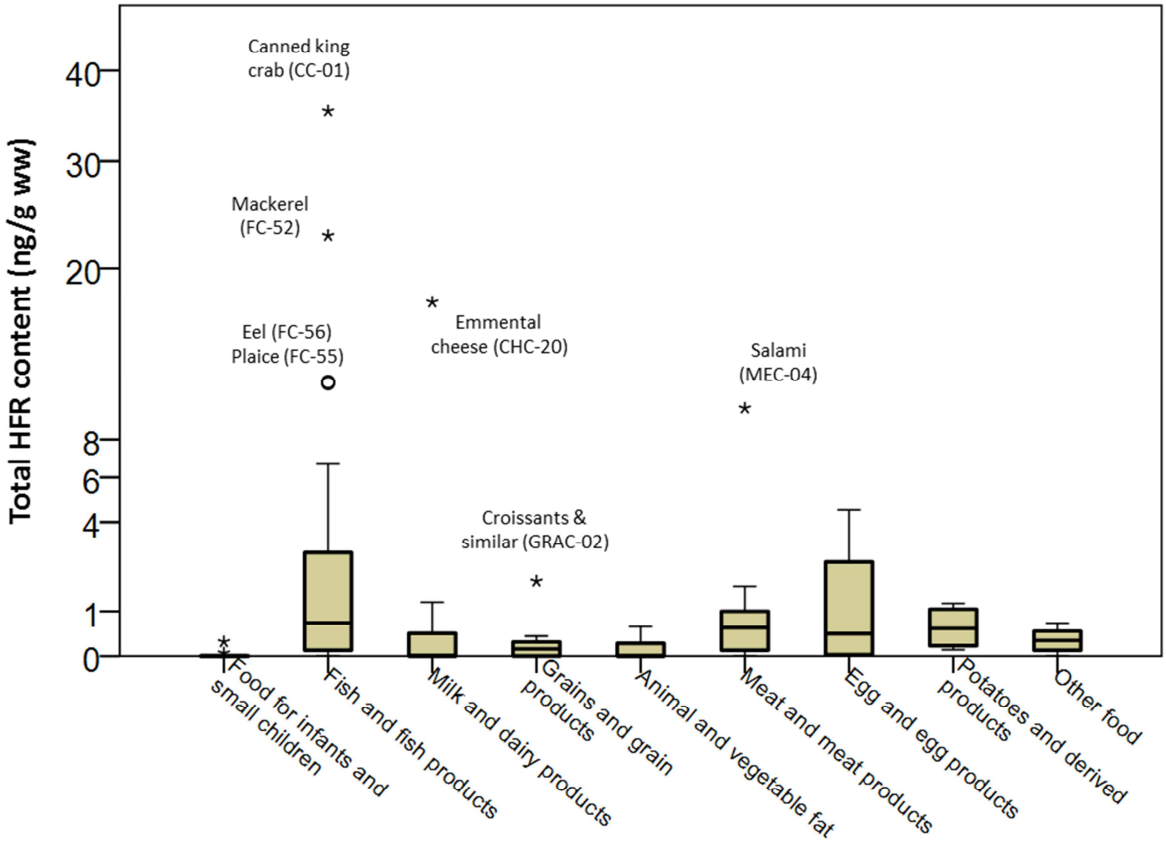
**Table 2.** Comparison of the maximum PBDE and HBCD levels measured in the current study with the proposed action limits (AL) in Belgium (Scientific Committee of the FASFC 2017).

| **Foodstuff** | **Proposed AL for ∑PBDEs[a]** | **Max ∑PBDEs in this study** | **Proposed AL for ∑HBCDs[b]** | **Max ∑HBCDs in this study** |
|---|---|---|---|---|
| Dairy products (cheese, …) (ng/g lw) | 40 | 64 | 500 | <LOQ |
| Milk (ng/g lw) | 30 | <LOQ | 400 | 0.5 |
| Eggs (ng/g lw) | 200 | 7.8 | 3000 | 54 |
| Vegetable oils and butter (ng/g lw) | 60 | <LOQ | 900 | <LOQ |
| Meat (ng/g lw) | 80 | 24 | 1000 | 2.3 |
| Fish oil based food supplement (ng/g lw) | 100 | <LOQ | 2000 | 0.6 |
| Food for infants (ng/g ww) | 0.7 | 0.3 | 10 | <LOQ |
| Fish (ng/g ww) | 30 | 5.7 | 400 | 5.5 |

[a]The sum includes BDE-28, BDE-47, BDE-99, BDE-100, BDE-153, BDE-154, BDE-183, BDE-209

[b]The sum includes α-HBCD, β-HBCD, γ-HBCD




Total HFR content (ng/g ww)
40
30
20
8
6
4
1
0
Canned king crab (CC-01)
Mackerel (FC-52)
Eel (FC-56)
Plaice (FC-55)
Emmental cheese (CHC-20)
Croissants & similar (GRAC-02)
Salami (MEC-04)
Food for infants and small children
Fish and fish products
Milk and dairy products
Grains and grain products
Animal and vegetable fat
Meat and meat products
Egg and egg products
Potatoes and derived products
Other food

## Highlights

- Analysis of 183 composite foodstuff by GC-MS and LC-MS/MS
- Occurrence data for halogenated flame retardants (HFRs) in Belgian foodstuff
- Occurrence data for emerging flame retardants and less studied food groups for which data are scarce
- PBDEs and brominated phenols were the most frequently detected HFRs
- TBBPS, 26-DBP, HBB, TBB and BTBPE were not detected in any of the analyzed foods