

Environmental risk of plastic materials:  
**Hexabromocyclododecane Contamination in  
China's Marine Environment**

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April 30, 2023

## **ABSTRACT**

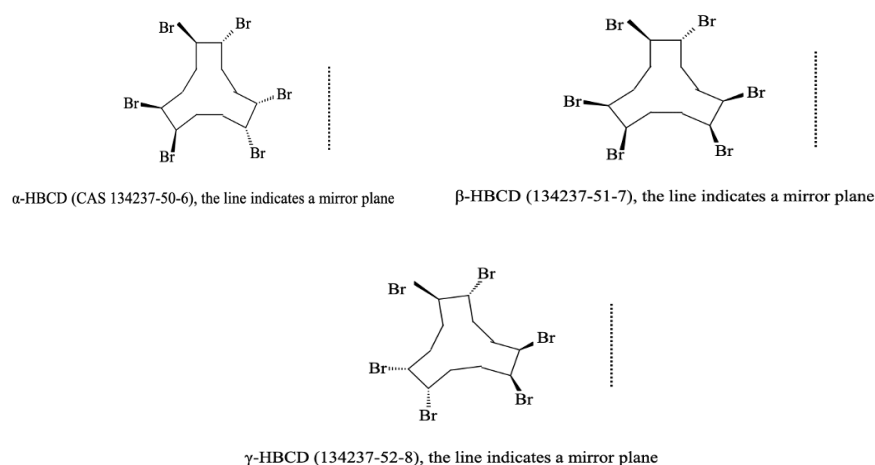
This mini-review discusses the physical-chemical properties, usage, and environmental impact of 1,2,5,6,9,10-hexabromocyclododecane (HBCD), a commonly used brominated flame retardant. The review highlights HBCD's persistence, bioaccumulation, long-range transport, and toxicity profile, as well as its potential to contaminate water and affect human health in China.

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# 1 INTRODUCTION

The compound 1,2,5,6,9,10-hexabromocyclododecane (HBCD) is a commonly used brominated flame retardant (BFR) that has been produced since the 1960s. It is primarily used in the construction industry, but also in secondary industries such as upholstered furniture, car seats, interior fabrics, and electrical and electronic devices. HBCD is mainly produced in the US, Europe, China, and Japan, and its global demand has significantly increased from 16,500 tons in 2001 to 31,000 tons in 2011 over the past decade (POPRC, 2011, POPRC, 2012a). Notably, China emerged a major producer of HBCD, with 18,000 tons being produced in 2011 alone (POPRC, 2012a). Technical HBCD (t-HBCD) is mostly composed of the diastereoisomers  $\alpha$ ,  $\beta$ , and  $\gamma$ , with  $\gamma$ -isomer making up over 70% of the total, while  $\alpha$ - and  $\beta$ -isomers make up roughly 10% and 6% of the mixture, respectively. Due to its persistence, bioaccumulation, long-range transport, and toxicity profile, HBCD was included in the US Environmental Protection Agency's List of Chemical of Concern and is being considered for listing as a persistent organic pollutant (POP) by the Persistent Organic Pollutants Review Committee (POPRC) under the Stockholm Convention (Encyclopedia of Toxicology 3rd edition, 2014). China banned the production, import, export, and use of HBCDs in December 2016 and added them to the list of Priority Chemicals under Control in December 2017. Despite this, HBCDs are still present in the environment globally and can enter the ocean via sewage outlets, surface runoff, and rivers due to its non-degradable nature, as reported by Ying Zhang et al. (2022). In China, high concentrations of HBCD were found in river water, river sediment, and soil surrounding one of the largest production facilities of HBCD, which is located approximately 8 km from the coastline (Zhang et al., 2018). The river water contaminated with HBCD flows into the sea at the estuary, potentially affecting coastal organisms. As a result, further investigation was conducted to track contamination in organisms to ensure food safety and human health. Moreover, this report is aimed at deriving some important results from other studied, which estimated the health risk for residents consuming fish in China. Likewise, an ecological risk assessment of HBCD in the marine ecosystem of China was conducted based on the collected data.



**Figure 1. The three main diastereoisomers (pairs of enantiomers) in t-HBCD (recreated from EC (2008); data from (Heeb et al. 2005)).**

## 2 PHYSICAL-CHEMICAL PROPERTIES

Two numbers are listed in the CAS Registry for undefined combinations of commercial or t-HBCD: 25637-99-4 (without numbering for the location of the bromine substitution pattern) and 3194-55-6 for 1,2,5,6,9, and 10-HBCD. HBCD has complicated stereochemistry that includes 16 stereoisomers, and it can isomerize both in the production process and outside in the environment. Most chemical characteristics and toxicity tests that have been published so far concern t-HBCD, an isomer mixture.

**Table 1. Summary of physico-chemical properties**

Reach ref Annex, §	Property	Value	Comments
VII, 7.1	Physical state at 20 °C and 101.3 kPa	White odourless solid	
VII, 7.2	Melting/ freezing point	Ranges from approximately: 172-184 °C, as an average value was used as input data in the EU risk assessment	Smith et al. (2005)
		179-181 °C $\alpha$ -HBCD 170-172 °C $\beta$ -HBCD 207-209 °C $\gamma$ -HBCD	Smith et al. (2005)
VII, 7.3	Boiling point	Decomposes at >190 °C	Peled et al. (1995)
VII, 7.5	Vapour pressure	6.3 x 10 <sup>-5</sup> Pa (21 °C)	Stenzel and Nixon (1997)
VII, 7.7	Water solubility	48.8±1.9 µg/L $\alpha$ -HBCD 14.7±0.5 µg/L $\beta$ -HBCD 2.1±0.2 µg/L $\gamma$ -HBCD 65.6 µg/L (HBCD technical product, sum of above)	MacGregor and Nixon (2004)
VII, 7.8	Partition coefficient n-octanol/water (log value)	5.07±0.09 $\alpha$ -HBCD 5.12±0.09 $\beta$ -HBCD 5.47±0.10 $\gamma$ -HBCD 5.625 (technical product)	MacGregor and Nixon (1997) Hayward et al. (2006)

### 3 ENVIRONMENTAL FATE PROPERTIES

**Degradation.** HBCD does not degrade significantly through hydrolysis due to its low water solubility, but it can degrade through biodegradation and photochemical breakdown. Two articles (Davis et al., 2003a, b and Davis et al., 2004) provide results from degradation simulation studies on HBCD in sediment and soil, where degradation was observed. HBCD was found to degrade into 1,5,9-cyclododecatriene (CDT) via a step-wise reductive dehalogenation process in both aerobic and anaerobic sediment. However, mineralization was not observed in the degradation experiments. The European Chemical Agency (ECHA, 2008) also reports that HBCD concentrations have been rising in biota, despite it degrading more slowly in sediment than predicted by modelling studies. Hence, ECHA concludes that HBCD meets the P-criteria in soil and sediment.

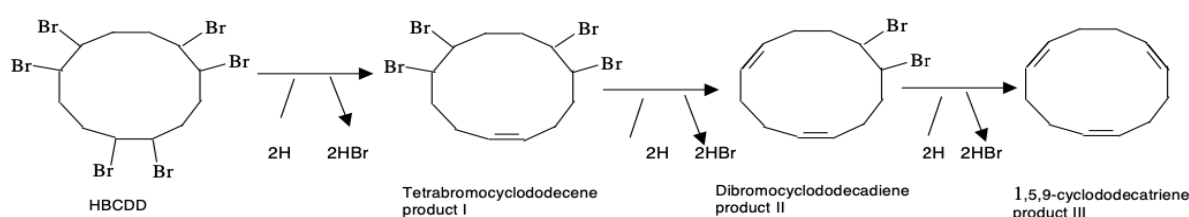


Figure 2. Stepwise dehalogenation of HBCD (Davis et al., 2004)

**Environmental distribution.** On adsorption, there are no experimental data available. The EU RAR, 2008 has derived a logK<sub>oc</sub> of 4.66, suggesting extremely high adsorption potential. It is reasonable to assume that HBCD has relatively little mobility in soil and sediment. HBCD is very slightly volatile according to the observed vapour pressure ( $6.3 \times 10^{-5}$  Pa at 20 °C). Henry's law constant at 20-25 °C is 0.75 Pa m mol based on the sum of the water solubilities of the individual diastereomers ( $66 \mu\text{g l}^{-1}$ ). Therefore, HBCD has a low potential for evaporating from aqueous surfaces. Evaporation of HBCD appears to be a less significant route of distribution because of its low volatility and high adsorption ability to suspended particles.

According to the EUSES modelling used for the EU risk assessment of HBCD (ECHA, 2008), the steady state mass fractions for HBCD at the regional level are as follows:

Freshwater	0.003%
Sea water	0.0003%
Air	0.00003%
Agricultural soil	45%
Industrial soil	0.015%
Freshwater sediment.	0.02%
Sea water sediment	0.0003%

**Distribution modelling.** Assuming equal emissions to air, water, and soil, the level III fugacity model Epiwin 3.20 derives the following distribution of HBCD:

Air	0.03%
Water	8.1%
Soil	83%
Sediment	9.1%

Despite HBCD's slow degradation in the atmosphere (half-life > 2 days), its low volatility and strong adsorption potential mean that long-distance atmospheric transport occurs primarily in aerosol form (Wania, 2003). But it should be noted the significance of HBCD's distance-to-concentration correlation were detected to be low ( $r^2 = 0.45$ ;  $p=0.33$ ), and the estimates' standard errors were high, possibly due to the limited number of sites used in the regression. However, when combined with other findings in organohalogen chemicals, the results suggest that HBCD has a strong long-range transport capability Ueno et al. (2006).

**Aquatic bioaccumulation.** The concentration of hexabromocyclododecanes (HBCDs) in mollusks from Bohai Sea and Hong Kong has been reported in Zhang et al (2022). The concentration in mollusks from the Bohai Sea was higher than in Hong Kong waters, possibly due to pollution from domestic and industrial effluents. HBCD concentrations in mollusks from China were comparable to those in South Korea and Europe, higher than those from California, and much lower than the highest concentration in Japan. The bioaccumulation of  $\alpha$ -HBCD is higher than  $\gamma$ -HBCD and  $\beta$ -HBCD, while  $\gamma$ -HBCD and  $\beta$ -HBCD are metabolized more quickly than  $\alpha$ -HBCD, resulting in an enrichment of  $\alpha$ -HBCD. The molecular response of clams exposed to HBCDs at environmentally relevant exposure levels showed differentially expressed genes involved in stress, defense response and metabolism. The toxicity of HBCDs on marine crustaceans was also evaluated, and it affected the ingestion and filtration rates of the marine copepod in a short-term sub-lethal exposure experiment. HBCDs were widely detected in fish from Chinese coastal areas, and more studies are necessary to understand the response and adaptation mechanisms of these organisms under multiple environmental stressors.

**Terrestrial bioaccumulation.** There are no BCF studies on earthworms available. Nonetheless, the content of HBCD in earthworms has been measured in a study on the survival and reproduction of earthworms (Aufderheide et al., 2003). In the study, it was stated that the bioaccumulation factors based on the wet weight concentrations of worms and soil varied from 0.03 to 0.08. In worm tissue, the  $\alpha$ -HBCD fraction accounts for around 60% of the entire concentration of HBCD, whereas the  $\alpha$ -diastereomer makes up approximately 6% of the total concentration in soil. For  $\alpha$ -HBCD (0.3- 0.8), the diastereomer specific BAF is more than an order of magnitude greater than for  $\gamma$ -HBCD (0.005-0.02). This is consistent with what has been seen in other biota, such as mammals and fish, where the  $\alpha$ -HBCD is the dominant diastereomer. It is unknown why there is this difference. It might be caused by, for instance, a greater absorption of the  $\alpha$ -diastereomer or variations in metabolism between the diastereomers.

**Biomagnification factor (BMF) and trophic magnification factor (TMF)** are commonly used to study the biomagnification behavior of pollutants in the food chain. BMF is defined as the ratio of the average lipid-normalized concentration between predator and prey, while TMF describes biomagnification and is calculated based on the correlations between average lipid-normalized concentration and trophic levels (TLs). Values greater than 1 indicate biomagnification, while values between zero and one suggest that the compound is present throughout the food web but is not being biomagnified. In several studies investigating the biomagnification properties of hexabromocyclododecanes (HBCDs) in the marine ecosystem of the Chinese coast, mantis shrimp, veinedrappa whelk, helice crab, anchovy, octopus, weever, bartial flathead, sea catfish, hairtail, clam, oyster, shrimp, conch, crab, mullet, flathead fish, tongue sole, perch, porgy, and goby were analyzed. The results showed significant positive associations between the concentrations of  $\alpha$ -HBCD and total HBCDs and trophic levels, while  $\beta$ -HBCD and  $\gamma$ -HBCD did not show any statistically significant correlation between the concentrations and the TLs. The biomagnification was observed throughout the food web, with TMF values ranging from 1.64 to 10.8. The biomagnification potential of  $\beta$ -HBCD was higher than that of  $\gamma$ -HBCD, with BMFs of 3.06-524 and 1.56-45.9 for  $\beta$ -HBCD and  $\gamma$ -HBCD, respectively. The strongest biomagnification was found in cetacean species, with BMFs of 16.8-7223 for  $\alpha$ -HBCD and TMFs of 7.9 and 10.3 for  $\Sigma$ HBCDs and  $\alpha$ -HBCD concentrations, respectively. (Zhang et al., 2022)

**Toxicity.** The ECHA indicated the following data for acute toxicity:

The minimum lethal dose in oral exposure, in rats, is greater than 20 g/kg.

The minimum lethal dose in inhalation, in rats, is greater than 200 mg/l.

The minimum lethal dose in dermal exposure, in rabbits, is greater than 20 g/kg. (ECHA, 2008)

Their conclusion: the data on acute toxicity that are currently available indicate very low acute toxicity and do not support a classification of HBCD under EU standards. (ECHA, 2008). Moreover, the substance does not cause irritation to the eye or corrosivity to skin. (ECHA, 2008). The data available on repeated dose toxicity and carcinogenicity do not suggest a classification of HBCD according to EU criteria. HBCD was negative in tests for in vitro chromosomal aberration and in vivo micronucleus, and it did not cause mutations in the Ames test. Hence, it can be said that HBCD has negligible genotoxic potential both in vitro and in vivo. The mutagenicity data that are currently available do not support the EU classification of HBCD (ECHA, 2008). HBCDs can cause adverse effects to the thyroid and liver, increase cholesterol and tibial bone mineral density, inhibit egg development and neural development in rats, and in severe cases, liver necrosis and cancer may occur in rats exposed to HBCDs. Chronic toxicity of HBCDs was also shown in aquatic life although its acute toxicity is low, such as the significant toxic effects on the reproductive development of marine invertebrates: the malformation rate increased, and survival reduced significantly in HBCD exposed zebrafish. Thus, HBCDs may be harmful to humans through the process of biomagnification. (Zhang et al., 2022)



## 4 HAZARD ASSESSMENT

**Occupational risk.** According to the literature and field research, study conducted by Shan Yi (2016) included 599 plants and identified 614-1213 potentially exposed workers to hexabromocyclododecane (HBCD) across six occupational scenarios. Personal samplers were used to collect air samples from representative workshops in each scenario, and the concentration of HBCD detected in workshop air was between 3.72-35.5  $\mu\text{g m}^{-3}$ . HBCD exposure levels for workers ranged from 0.61-840  $\mu\text{g kg bw}^{-1} \text{d}^{-1}$ , with the highest levels found under scenario I and minimal levels under scenarios V and VI. The study also found that workers exposed to HBCD had no significant risk of repeated dose toxicity, but approximately 9-11 workers (0.90-1.34% of all workers) might encounter reproductive toxicity risk, and approximately 117-226 workers (18.6-19.1% of all workers) might carry a risk of developmental neurotoxicity. Finally, approximately 453-840 workers (69.2-73.6% of all workers) might carry a risk of reduced bone mineral density in female offspring. (Shan Yi et al., 2016)

**Environmental risk.** The study conducted by Shan Yi's research group in 2016 aimed to examine the level of contamination by HBCD in the environment surrounding HBCD production and processing plants. The study found that soil and marine sediment samples collected from the study area were contaminated by HBCD, with the  $\gamma$ -HBCD isomer being the most prevalent in the sediments, accounting for over 70% of total HBCD. The HBCD concentrations in biological samples collected from the study area were 1-2 orders of magnitude higher than those observed in other non-point source studies, indicating that organisms in the area had been contaminated by HBCD production and processing activities. The study also found that the rate of contamination exceeded the rate of HBCD degradation, migration, or conversion, leading to accumulation in the environment. The concentrations of HBCD in environmental samples varied, with soil samples ranging from 0.11 to 560  $\text{ng g dw}^{-1}$ , sediment samples ranging from 20.4 to 24.2  $\text{ng g dw}^{-1}$ , and biological samples ranging from 166 to 1964  $\text{ng g lw}^{-1}$ . The HQ method was used to evaluate the environmental risk posed by HBCD, and the results indicate that it is not a significant threat to nearby soil and marine environments. However, the presence of a high proportion of  $\gamma$ -HBCD suggests that HBCD could potentially pose a threat to marine environments, which in turn could impact human health.

**Occurrence, Spatial distribution, and health risk of HBCD in Source Water in Lower Yangtze River, China.** The concentration of HBCD in the water samples collected from different sources ranged from 0.58 to 3.71  $\text{ng/L}$ , with a mean value of 1.18  $\text{ng/L}$  and a median value of 0.88  $\text{ng/L}$  in Yueqing Zhang's study in 2022. While the detection frequency for  $\alpha$ -HBCD was 86%, the detection frequency for  $\beta$ - and  $\gamma$ -HBCD was 100%. HBCD levels in natural water sources without direct emission were below 1  $\text{ng/L}$ . In contrast, wastewater resulted in higher concentrations of HBCD, such as 5080  $\text{ng/L}$  in the Mihe River of China and 180-2100  $\text{ng/L}$  in the Kuzuryu River of Japan. The spatial distribution of HBCD in water sources showed a hotspot at site S18, which had a relatively high concentration of 3.71  $\text{ng/L}$  due to the presence of industries such as textile printing/dyeing and automobile and equipment manufacturing that use HBCD. The study suggests that textile and laundry wastewater may be the source of HBCD

at site S18 and municipal wastewater may be the source at the other sites in the Yangtze River. The exposure to HBCD through drinking water was much lower than that through food. The derived reference dose (RfD) for metabolic effects was the lowest at 2 ng/kg bw/day, indicating that metabolic disorder was the most sensitive effect. (Zhang et al., 2022)

**Human dietary intake of HBCD at e-waste recycling sites in Eastern China.** Food has been reported as one of the main routes of human exposure to hexabromocyclododecane (HBCD) (Zheng et al. 2012; He et al. 2013).

**Table 2. Concentration of HBCD and its diastereomer profile in composite food samples from e-waste recycling and control sites in China.**

Sample type		Site Category	Lipid (%)	$\Sigma$ HBCD ng/g lipid weight	$\alpha$ -HBCD (%)	$\beta$ -HBCD (%)	$\gamma$ -HBCD (%)
Vegetable oil		Control	100	0.1	100	0	0
Muscle	Fish	Control	23.4	5.9	85	0	15
		E-waste	5.7	310	80	3	17
	Shrimp	Control	4.5	5.7	100	0	0
		E-waste	4.5	<0.06			
	Chicken	Control	19.0	0.09	100	0	0
		E-waste	38.6	78.7	49	2	49
	Duck	Control	18.2	2.4	95	0	5
		E-waste	48.1	26.4	84	5	11
	Pork	Control	24.6	3.1	51	0	49
		E-waste	18.4	<0.06			
Liver	Chicken	Control	38.6	78.7	47	0	53
		E-waste	20.9	42.5	71	3	22
	Duck	Control	21.5	29.9	33	0	67
		E-waste	14.5	5.7	64	0	36
Egg	Chicken	Control	29.6	1.7	100	0	0
		E-waste	28.5	47.5	100	0	0
		E-waste	26.5	37.7	100	0	0
	Duck	Control	32.7	0.5	100	0	0
		E-waste	32.0	9.3	100	0	0
		E-waste	27.0	3.0	100	0	0
		E-waste	31.0	17.5	90	5	5
		E-waste	28.8	2.2	72	22	6
		E-waste	29.6	14.7	100	0	0

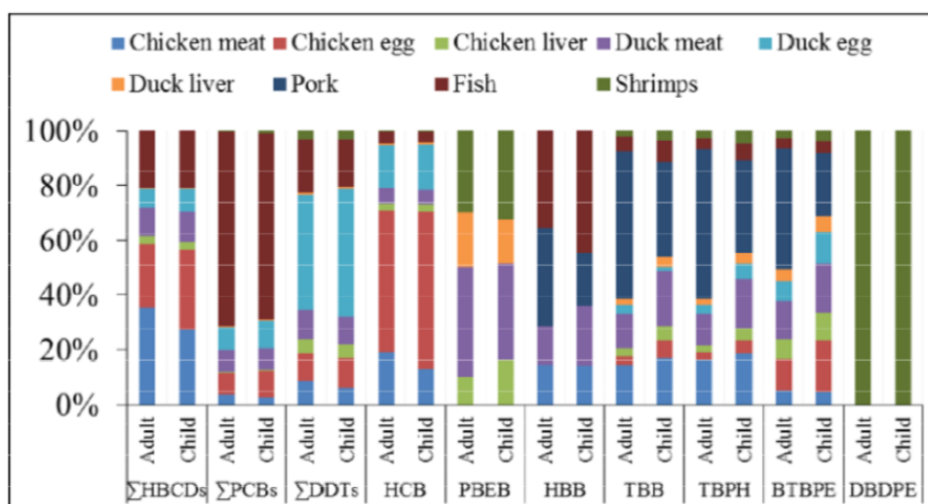
According to Table 2, fish muscle had the highest concentration of HBCD, followed by chicken muscle, chicken egg, and chicken liver. In most e-waste related samples, HBCD concentrations were much higher than those in corresponding controls, except for e-waste related pork and shrimp samples, which had lower HBCD concentrations than their corresponding controls.

Previous studies have reported higher HBCD concentrations in passerine birds from urban sites, which was attributed to higher HBCD releases from textile and construction applications in urban areas. However, e-waste-impacted samples of duck muscle, chicken muscle, and liver showed significantly higher concentrations of HBCD than corresponding controls, likely due to the use of potentially HBCD-containing recycled construction materials in the construction of chicken and duck housing at the Taizhou e-waste sites. Interestingly, the highest concentration of HBCD was found in a duck egg sample from a site where e-waste recycling had only recently ceased. The study also found that  $\alpha$ -HBCD is the predominant HBCD congener, contributing to an average of  $77\pm 31\%$   $\Sigma$ HBCDs in all samples. In e-waste related sites,  $\gamma$ -HBCD only comprised 22% and 36% of  $\Sigma$ HBCDs in chicken and duck livers, respectively, whereas in control samples of the same matrices,  $\gamma$ -HBCD contributed 53% and 67% of  $\Sigma$ HBCDs, suggesting exposure of control birds to a comparatively "fresh" HBCD source. (Labunska et al., 2015).

**Table 3. Estimated adult and child exposure to  $\Sigma$ HBCD (ng/kg bw/day) via different types of food at e-waste recycling sites in Taizhou, China. n/e – not estimated.**

	Meat					Liver		Egg		TDI
	Fish	Shrimps	Chicken	Duck	Pork	Chicken	Duck	Chicken	Duck	
<b>Adult</b>	2.18	n/e	3.67	1.1	n/e	0.28	0.03	2.44	0.71	<b>10.4</b>
<b>Child</b>	7.48	n/e	9.86	3.95	n/e	1.08	0.11	10.5	3.06	<b>36.1</b>

**Figure 3. Contributions to total dietary exposure for organic contaminants from different types of food originating from Taizhou, China**



Irina Labunska's study in 2015 found that the estimated daily intake of HBCD was significantly higher in adults and children at e-waste sites in Taizhou compared to other studies in Sweden and Belgium. Chicken muscle and eggs were the main sources of HBCD exposure, with fish also contributing to dietary exposure. Duck muscle and eggs were found to be the main sources of PBDE exposure. Liver made the lowest contribution to the estimated dietary intake of HBCD. These differences in exposure likely reflect variations in local dietary preferences and environmental contamination.

**Figure 4. Comparison of consumption of different products in China in the unit of kilogram per capita in 2015 and Contributions to total dietary exposure for HBCD from the same types of food originating from Taizhou, China (2015).**

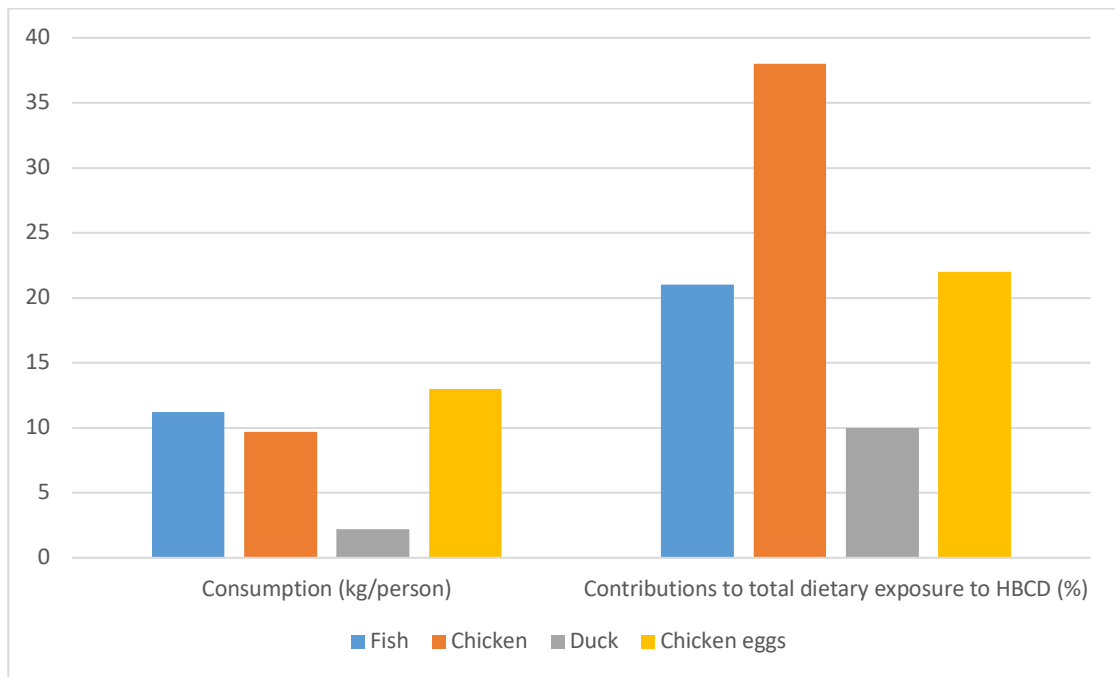


Figure 4 suggests that consumption of chicken contributed to the highest dietary exposure of HBCD in China in 2015, followed by chicken eggs and fish. The data were collected from [www.statista.com](http://www.statista.com).

## 5 ALTERNATIVE PRODUCTS

There doesn't seem to be a fire retardant that will work equally well for the main applications of HBCD, notably in Expanded Polystyrene (EPS) and Extruded Polystyrene (XPS). The primary advantage of HBCD is that it provides exceptional performance in polystyrene foams since it is efficient at low concentrations (EC RAR, 2008).

For use as building insulation, there are commercially available equivalents for EPS/XPS that are technically possible in many, but not necessarily all, situations. The fact that mineral wools are currently widely used and account for at least 30% of the building insulation industry in Europe suggests that they are both technically and financially feasible alternatives to EPS/XPS in many applications. However, in some applications, EPS/XPS's lower mass density or superior moisture resistance could be crucial. Another product that is available on the market that complies with fire safety regulations is phenolic foam. It has a low density, is moisture resistant, and is very thermally efficient. Only a small portion of the European insulation market is occupied by it, which is mostly utilized in situations where moisture resistance or thickness are crucial. This may indicate that it is relatively expensive. Although the chemicals used to make phenolic foam are dangerous to human health, these alternative insulation materials are not particularly harmful to the environment or human health. (ECHA/2008/02/SR4/ECA.226)

In High Impact Polystyrene and textile coatings, there are commercially available substitutes for HBCD that offer the same level of fire protection and functionality. These substitutes include non-halogenated flame retardants. (ECHA/2008/02/SR4/ECA.226)

## 6 CONCLUSION

In conclusion, HBCD is a widely used brominated flame retardant that is mainly produced in the US, Europe, China, and Japan. Despite being banned in China in 2016, HBCD still exists in the environment globally and can enter the ocean through various means. This poses a risk to coastal organisms and food safety, requiring further investigation. The report summarizes important results from studies that estimated the health risk for residents consuming fish in China and conducted an ecological risk assessment of HBCD in the marine ecosystem. (HBCD) does not degrade significantly through hydrolysis but can degrade through biodegradation and photochemical breakdown. HBCD has a high adsorption potential, which suggests that it has relatively little mobility in soil and sediment. Long-distance atmospheric transport occurs primarily in aerosol form due to its low volatility and strong adsorption potential. HBCD concentrations have been rising in biota, including mollusks, fish, and earthworms, indicating bioaccumulation. The  $\alpha$ -HBCD fraction is the dominant diastereomer in soil and biota. The toxicity of HBCDs on marine crustaceans and the molecular response of clams exposed to HBCDs have been evaluated, but more studies are necessary to understand the response and adaptation mechanisms of these organisms under multiple environmental stressors.

HBCD is a hazardous chemical with potential health risks for workers exposed to it and for the environment. Studies have shown that workers in HBCD production and processing plants may carry risks of reproductive toxicity, developmental neurotoxicity, and reduced bone mineral density in female offspring. HBCD has also been found to contaminate soil, sediment, and organisms in the environment, potentially posing a threat to marine environments and human health. The concentration of HBCD in water sources has been found to be relatively low, but higher concentrations have been found in wastewater. Food, especially fish and poultry, is a major route of human exposure to HBCD, with e-waste recycling sites in Eastern China showing higher concentrations of HBCD in food samples due to the use of potentially HBCD-containing recycled construction materials. Overall, measures should be taken to reduce exposure to HBCD and prevent further environmental contamination.

There are few available alternative products to replace HBCD, namely mineral wool and phenolic foam, but these products do not work equally well as a fire retardant in main applications such as EPS and XPS. However, non-halogenated flame retardants are commercially available substitute, and they provide equivalent functionality and fire safety in high impact polystyrene and textile coatings.

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