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Global PBDE contamination in cetaceans. A critical review☆

Alice Bartalini [a,b], Juan Muñoz-Arnanz [a,*], Natalia García-Álvarez [b], Antonio Fernández [b], Begoña Jiménez [a]

[a] Department of Instrumental Analysis and Environmental Chemistry, Institute of Organic Chemistry (IQOG-CSIC), Juan de la Cierva 3, 28006, Madrid, Spain
[b] Unit of Histology and Pathology, Institute of Animal Health (IUSA), Veterinary School, University of Las Palmas, 35413 Arucas, Las Palmas de Gran Canaria, Spain

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ABSTRACT

This review summarizes the most relevant information on PBDEs' occurrence and their impacts in cetaceans at global scale, with special attention on the species with the highest reported levels and therefore the most potentially impacted by the current and continuous release of these substances. This review also emphasizes the anthropogenic and environmental factors that could increase concentrations and associated risks for these species in the next future. High PBDE concentrations above the toxicity threshold and stationary trends have been related to continuous import of PBDE-containing products in cetaceans of Brazil and Australia, where PBDEs have never been produced. Non-decreasing levels documented in cetaceans from the Northwest Pacific Ocean might be linked to the increased e-waste import and ongoing production and use of deca-BDE that is still allowed in China. Moreover, high levels of PBDEs in some endangered species such as beluga whales (*Delphinapterus leucas*) in St. Lawrence Estuary and Southern Resident killer whales (*Orcinus Orca*) are influenced by the discharge of contaminated waters deriving from wastewater treatment plants.

Climate change related processes such as enhanced long-range transport, re-emissions from secondary sources and shifts in migration habits could lead to greater exposure and accumulation of PBDEs in cetaceans, above all in those species living in the Arctic. In addition, increased rainfall could carry greater amount of contaminants to the marine environment, thereby, enhancing the exposure and accumulation especially for coastal species.

Synergic effects of all these factors and ongoing emissions of PBDEs, expected to continue at least until 2050, could increase the degree of exposure and menace for cetacean populations. In this regard, it is necessary to improve current regulations on PBDEs and broader the knowledge about their toxicological effects, in order to assess health risks and support regulatory protection for cetacean species.

1. Introduction

In recent years there has been growing concern pertaining the stable and high levels of some legacy persistent organic pollutants (POPs) continuously reported in the environment (Jepson et al., 2016; Jones, 2021; Stuart-Smith and Jepson, 2017). At the same time, recent studies have drawn attention to the impact that climate change could have on POPs' cycling and distribution and consequently on exposure and accumulation in living organisms (Borgå et al., 2022; Hung et al., 2022; Nadal et al., 2015). Evidence suggested that climate change-driven processes could enhance long-range transport of POPs (Dalla Valle et al., 2007; Li et al., 2021) and their remobilization from secondary sources, especially in the Arctic zone (Borgå et al., 2022; Chen et al., 2019; Ma et al., 2011; Potapowicz et al., 2019). Undoubtedly, this could translate into enhanced POP exposure and accumulation in cetaceans living in this area. Additionally, some cetacean species inhabiting industrialized regions such as killer whales (*Orcinus orca*) are already facing important anthropogenic pressures and ultimately population depletion, which has been linked to their high body burdens of POPs (Desforges et al., 2018; McGuire et al., 2020).

In this regard, two individual-based models have described the possible negative influence of polychlorinated biphenyls (PCBs) and structurally similar compounds on the potential annual growth rate in the bottlenose dolphin (*Tursiops truncatus*) population of Sarasota Bay (Hall et al., 2006), and on the long-term viability of the world's killer whale population (Desforges et al., 2018). Despite the evidence of the detrimental impact of POPs on marine mammals' health, a comprehensive understanding of toxicological consequences has not yet been

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* Corresponding author.
E-mail address: juan.ma@iqog.csic.es (J. Muñoz-Arnanz).

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achieved. In consequence, species-specific threshold values are not well established.

High and stable concentrations, enhanced exposure due to climate change processes and population collapse related to high body burdens for some cetacean species, have led to questioning about the effectiveness of current global efforts aimed at the mitigation and elimination of POPs (Stuart-Smith and Jepson, 2017; Wang et al., 2021; García-Alvarez et al., 2014). Thus, revision of existing multilateral environmental agreements (MEAs), implementation of national legislations and more holistic approaches have become necessary. Special attention should be given to polybrominated diphenyl ethers (PBDEs) for which emissions from in use and waste stocks are expected to continue until 2050 (Abbasi et al., 2019). It is plausible that prohibitions of production and use are not steadily lowering the long-term environmental impact of PBDEs, but shifting instead the issue from more to less industrialized regions where the demand of PBDE-containing products and e-waste import have increased since the late 1990s (Li et al., 2016).

PBDEs are a family of brominated-flame retardants (BFRs) comprising 209 congeners that are divided into 10 groups, from mono-to deca-BDE, according to their different degree of halogenation. The estimated global production from 1970 to 2005 was around 1.3 and 1.5 million of tons (UNEP, 2010), comparable to that of PCBs (Breivik et al., 2002). United States and Israel were the two main manufacturers prior to 1995, while China started its production in the 1980s, principally focused on deca-BDE, and became one of the main producers 20 years later (Abbasi et al., 2019). PBDEs have been commercialized as three technical mixtures, i.e., penta-, octa-, and deca-brominated diphenyl ether, named so after the major congeners present in each mixture. They were primarily used as flame retardants in polyurethane (PUR) foams, computers, housing appliances, electrical and electronic products.

By the beginning of the 21st century, environmental authorities (e.g., the European REACH regulation; the Stockholm Convention; the United States Environmental Protection Agency; Environment and Climate Change Canada) became aware of the harmful effects of PBDEs based on the available scientific evidence, and nations started to take measures. In 2004 the United States and Europe voluntarily phased out the production of penta- and octa-BDE mixtures (Hites, 2006), while in 2009 they were globally regulated with their introduction in the annex A (elimination) of the Stockholm Convention (UNEP, 2009). Accordingly, most reported PBDEs started to decline or level off in the early 2000s in environmental media around the world (Law et al., 2014), with notorious exceptions such as some top predators in Northern Europe (McKinney et al., 2006; Vorkamp et al., 2005) and beluga whales from the Gulf of St. Lawrence (Canada) (Raach et al., 2011) and Alaska (Hoguet et al., 2013). Importantly, the ban on these two mixtures lead to an increased production of deca-BDE formulation, made up of 96.8% BDE-209, until its more recent inclusion in the annex A of the Stockholm Convention in 2019 (UNEP, 2022a). The extensive use of deca-BDE shifted the environmental congener pattern distribution of PBDEs with increasing levels of higher brominated congeners in different abiotic and biotic matrices (Olofsson et al., 2012; Su et al., 2015; Verreault et al., 2018). Thus, Olofsson et al. (2012) reported decreasing concentrations (ca. 20% each year) of BDE-154 and BDE-183 (representative of penta-mix and octa-mix, respectively) in sewage sludge from Sweden during 2004–2011, while BDE-209 increased (ca. 16%) over the same period. Heightened levels of deca-BDE were observed in herring gulls (*Larus argentatus*) from the Laurentian Great Lakes (Su et al., 2015) and top predator birds from the Arctic (Verreault et al., 2018), sampled during 2012–2013. Even if BDE-209 is highly hydrophobic and has low water solubility, making it hardly bioavailable, evidence suggests that its biomagnification may take place in aquatic food webs (Castro-Jiménez et al., 2021; Johnson-Restrepo et al., 2005). In addition, its affinity to abiotic matrices such as sediments generates a large contaminant reservoir that put at risk low-trophic level species and their feeders. Toxicological relevance of higher brominated BDEs has not yet been fully investigated, but their debromination to lighter, more toxic and bioaccumulative congeners have been demonstrated (Eriksson et al., 2004; La Guardia et al., 2007; Rayne et al., 2003; Zhu et al., 2019). This fact is worthy of attention due to the large historical production of deca-BDE estimated to be almost 10 times higher than those of penta- and octa-BDE mixtures (Abbasi et al., 2019).

Even though PBDEs are mostly prohibited today on a global scale, their production and use is still ongoing in some world areas (Sharkey et al., 2020). Several countries have not yet ratified some amendments of the Stockholm Convention and in many cases also lack national regulations for both production and use. For example, in the United States only 13 states have applied limitations on the use/presence of PBDEs in certain goods entering the market, but no federal restrictions are in place. Other examples are those of Brazil and Indonesia where there are no regulations or limits defined for marketable goods or wastes (Sharkey et al., 2020). China has set concentration limits in newly manufactured goods for penta- and octa-BDE, but not for deca-BDE (Sharkey et al., 2020). Moreover, specific exemptions under the Stockholm Convention permit the use of the three commercial BDE mixtures for certain specific applications (UNEP, 2022b; 2022a). For instance, deca-BDE counts on specific exemptions allowing its production up to 2036 in the European Union, Republic of Korea and Switzerland (Sharkey et al., 2020).

Today, in use PBDE-containing products and waste stocks, accentuated by improper disposal in both developed and developing countries, represent important sources of PBDE contamination (Zhang et al., 2021). Due to the lack of chemical bonds and susceptibility to physical, chemical and biological factors, PBDEs can be easily released during waste disposal and therefore contaminate the surrounding environment or reach water masses through leaching and runoff (Danon-Schaffer et al., 2013; Zhang et al., 2021). Additionally, the high temperatures (80°C–90 °C) that can be reached in landfills enhance volatilization of lower brominated congeners (Stubbings and Harrad, 2014). Once in the atmosphere and thanks to their ability for long-range transport, these substances can reach remote areas where they have never been produced (Stubbings and Harrad, 2014; Wang et al., 2017; Zhang et al., 2021). Although recycling infrastructures have been implemented in many countries, older landfills without proper engineered facilities continue to be in use in Europe and United States (Propp et al., 2021), being likely responsible for inadvertent release of PBDEs into the environment (Ohajinwa et al., 2019).

As reported by Abbasi et al. (2019), emissions of PBDEs will continue for the next 30 years. This means that without appropriate control an additional enormous amount of BDE-209 (40 kilotons) may be subject to improper disposal. On top of that, higher temperatures and increased rainfall-runoff expected over the next few years as consequences of global warming could enhance PBDE volatilization and transportation from landfills and dumpsites. These all are likely to increase the amount of these semi-volatile contaminants reaching remote areas and entering the marine environment (Nadal et al., 2015).

Indeed, PBDEs have been detected in various environmental matrices for the last 40 years and since 10 years ago they have been recognized as ubiquitous contaminants (Abbasi et al., 2019). These substances enter the coastal waters through municipal and industrial wastewater outfalls, landfill leachates and atmospheric deposition (De Wit, 2002). Once in the marine environment, thanks to their hydrophobicity and persistency, PBDEs accumulate in biological tissues and biomagnify along the food web (Aznar-Alemany et al., 2019; De Wit, 2002; Noël et al., 2009). Species with large lipid reservoirs, high trophic levels and long-life spans, such as those in the cetacean suborder of odontocetes, tend to accumulate great concentrations of these substances and thus are regarded as bioindicators of POP contamination (Tanabe and Ramu, 2012). In addition, their reduced metabolic ability to eliminate POPs in comparison with carnivorous predators (polar bears, seals and walruses) make toothed whales especially vulnerable to this pollution (McKinney et al., 2011; Sonne et al., 2018). Even if POPs accumulate to a lesser extent in mysticetes, mobilization and redistribution of sequestered contaminants resulting from fasting and associated lipid fluctuations seem to cause a "reexposure" of target tissues (Bengtson Nash, 2018) increasing the likelihood and/or intensity of toxic effects (Aguilar et al., 1999; Bossart, 2011) in this suborder as well.

It has been demonstrated that upon continue exposure, marine mammals could reach potential toxic levels of PBDEs in their tissues. In the last 30 years many studies have reported PBDE levels in species around the world surpassing the threshold value for alteration in thyroid hormone levels in blubber of grey seals (Alonso et al., 2014). Moreover, stable or increasing concentrations of higher brominated congeners have been reported for different species and areas, in some cases in populations already impacted by other anthropogenic threats (Lebeuf et al., 2014; Simond et al., 2017; Jeong et al., 2020; Rotander et al., 2012).

Unlike others contaminants such as PCBs, PBDE effects in marine mammals have not been deeply investigated so far. In spite of the limited information available in literature in this regard, however, PBDEs in marine mammals seem to be associated with different harmful health effects. Particularly, they are responsible for toxicity responses involving the endocrine and immune systems (Simond et al., 2019; Beineke et al., 2005; Huang et al., 2020) as well as for cytotoxicity and genotoxicity to some extent (Rajput et al., 2021).

This review aims to compile the most relevant information on PBDE occurrence and associated health effects in cetaceans. A major objective would be drawing attention on the anthropogenic and environmental factors that could increase PBDE concentrations and associated risks in the next future. The discussion of PBDE levels and trends will be focused on the species with the highest reported levels and therefore the most potentially impacted by the current and continuous release of these substances.

The studies analyzed in this article are performed in blubber or liver of worldwide cetaceans covering a temporal range of about 30 years, since 1992 to 2021. Data are discussed when possible as mean concentration and range, and expressed as ng/g on lipid weight (l.w.) basis. All species investigated and discussed in this work are listed in Fig. 1.

1.1. Three decades of PBDEs in cetaceans: highest levels reported to date and trends

Despite their prohibition dating back more than 10 years ago, penta-BDE and octa-BDE congeners are still globally reported in marine mammals (Alonso et al., 2014; Aznar-Alemany et al., 2021; Kratofil et al., 2020; Simond et al., 2017). This can easily be linked to different factors, e.g. the persistence of these substances in the environment, the in use PBDE-containing products, the still ongoing production of these flame retardants in some geographical areas as well as the degradation of higher brominated (mainly deca-BDE) to lighter, bioavailable and more toxic congeners.

Kuehl et al. (1991) published the first study on PBDEs in cetaceans in 1991 reporting a mean value of 200 ng/g l.w. of total tetra-to hexa-BDEs in blubber of bottlenose dolphins collected during 1987–1988 along the Atlantic coast of the U.S. Since then, several studies have been conducted on different tissues and species, most of them focused on blubber of odontocetes inhabiting the Northern Hemisphere (Alonso et al., 2014) and reporting levels one order of magnitude higher than those initially documented in 1991 (Boer et al., 1998; Fair et al., 2007; Lindström et al., 1999; Moon et al., 2010). Values between 1060 and 7900 ng/g l.w., have been reported in more recent years (Kratofil et al., 2020; Noël et al., 2018; Simond et al., 2017) when, *a priori*, it would be expected a greater decrease as a consequence of the ban on production and use of penta- and octa-BDE. The only known threshold value for PBDEs is that of 1500 ng/g l.w., associated to the concentration in blubber of grey seals (*Halichoerus grypus*) eliciting alterations in thyroid hormone levels (Hall et al., 2003). Several studies have documented levels of PBDEs above this threshold (Boer et al., 1998; Fair et al., 2007; Kratofil et al., 2020; Lam et al., 2009; Lindström et al., 1999; Moon et al., 2010; Tuerk et al., 2004), pointing out the hazard these substances pose to cetacean health. Interestingly, some cetacean populations showing the highest concentrations of PBDEs are facing population depletion and lack of recovery. Most of them are classified as endangered species (Kratofil et al., 2020; Noël et al., 2018; Simond et al., 2017), which is likely consequence of

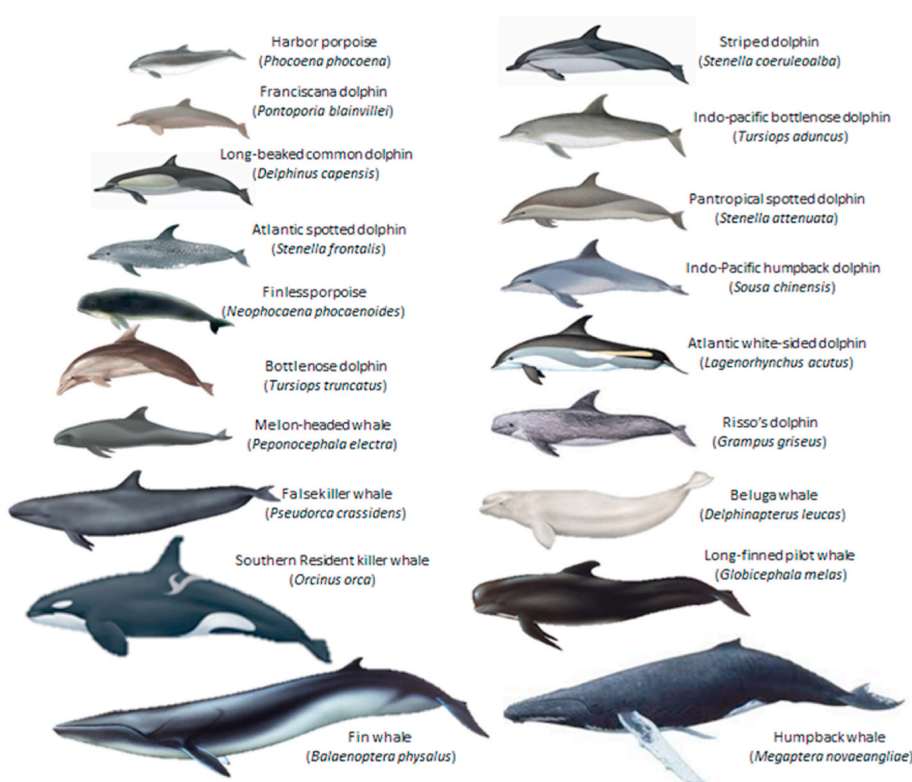


Fig. 1. Cetacean species investigated in this review.

the synergic effects of multiple anthropogenic threats including chemical pollution. Cetacean species living in urbanized coastal regions also deserve special attention due to the constant release of PBDEs in these areas and the subsequent high degree of contamination in their habitats.

To date, the highest concentrations of PBDEs in cetaceans' blubber have been reported for Southern Resident killer whales from Canada (mean: 7900 ng/g l.w.) (Noël et al., 2018), Main Hawaiian Island false killer whales (*Pseudorca crassidens*) (mean: 2900 ng/g l.w.) (Kratofil et al., 2020), Atlantic white-side dolphins (*Leucopleurus acutus*) in the North Sea (mean 7777 ng/g l.w.) (Boer et al., 1998), Indo-pacific humpback dolphin (*Sousa chinensis*) from Hong Kong waters (mean: 3590 ng/g l.w; range: 280-51,100 ng/g l.w.) (Lam et al., 2009) and bottlenose dolphins in the Northwestern Atlantic Ocean (mean: 7850 ng/g l.w.; range: 2680–22,800 ng/g l.w.) (Fair et al., 2007) (Fig. 2). High concentrations of PBDEs, often surpassing the threshold mentioned previously, have also been reported for long-finned pilot whales (*Globicephala melas*) from Faroe Islands (NW Atlantic) (mean: 1939 ng/g l.w.) (Lindström et al., 1999), Atlantic white-sided dolphins of Massachusetts coast (NW Atlantic) (mean: 1607 ng/g l.w.) (Tuerk et al., 2004), long-beaked common dolphins (*Delphinus capensis*) from Korea (NW Pacific) (mean: 1650 ng/g l.w.; range: 140-3100 ng/g l.w.) (Moon et al., 2010) and beluga whales from St. Lawrence Estuary (mean: 1068 ng/g l.w.; range: 666-1287 ng/g l.w) (Simond et al., 2017) (Fig. 2). These high values are to be linked to feeding habits and longer stays in urbanized and industrialized areas such as the California coastline – a feeding area for killer whales (Hanson et al., 2021) - and St. Lawrence Estuary where belugas live.

Different studies revealed the presence of PBDEs in Pacific salmon (*Oncorhynchus* spp) (O'Neill and West, 2009; Shaw et al., 2008; Veldhoen et al., 2010) with higher levels in coastal species, such as Chinook salmon (*Oncorhynchus tshawytscha*) the main prey of southern resident killer whale, than in those species with more oceanic distributions (O'Neill et al., 2006). As reported by the authors, this fact could contribute to the higher PBDE levels found in Southern Resident killer whales in comparison to those reported for other killer whale populations.

PBDE burdens in California's coastline have been found in sediments (Dodder et al., 2012) and wildlife, such as seabirds (Clatterbuck et al., 2018), fish (Maruya et al., 2016) and marine mammals (Shaul et al., 2015; Cossaboon et al., 2019), principally deriving from wastewater and stormwater discharges (Kimbrough, 2009). At the same time, different studies reported the presence of PBDEs in seal species from the Estuary and Gulf of St. Lawrence (Frouin et al., 2011; Soulen et al., 2018) and in fish and invertebrates from the St. Lawrence River, heavily impacted by discharge of treated wastewater, with the Montreal effluent being the most important PBDE source (Marcogliese et al., 2015).

Additionally, important concentrations of PBDEs in Main Hawaiian Island false killer whales have been related to the discharge of contaminated wastewater effluents in the coastal waters of Hawaii (Ylitalo et al., 2009). It is noteworthy that Canadian Southern Resident killer whale and Main Hawaiian Island false killer whale populations are experiencing a population depletion and lack of recovery due to multiple anthropogenic threats, among which chemical contamination may be playing an important role (Bradford et al., 2020; Desforges et al., 2018; Foltz et al., 2014; Hall et al., 2018). Hence, Southern resident killer whales and St. Lawrence beluga whales have been classified as endangered in the United States (Department of commerce. National Oceanic and Atmospheric, 2005) and Canada (SARA, 2017). The Main Hawaiian Island false killer whale stock was listed as endangered under the U.S. Endangered Species Act (ESA) in 2012 as a consequence of a rapid population decline during 1990s (GovInfo Endangered and Threatened Wildlife and Plants, 2012). Thus, the last census counted only 167 individuals, three time less than a previous estimation from 1998 (Bradford et al., 2018). Among the various causes, such as bycatch, decreased prey biomass and size and reduced genetic diversity, exposure to POPs and its related adverse health effects may contribute to the reduction of this population (Foltz et al., 2014; Kratofil et al., 2020; Ylitalo et al., 2009).

Among mysticetes, humpback whales (*Megaptera novaeangliae*) from the Gulf of Maine seem to be the most PBDE contaminated species, with a mean level around 900 ng/g l.w. in specimens sampled during 2005–2006 (Elfes et al., 2010). As suggested by the authors, this is likely a consequence of the great industrialization and human population density of the east coast of the United States.

Liver is the second most investigated tissue for PBDE contamination in cetaceans. PBDE levels in liver at which biological responses or toxicity effects occur have not yet been elucidated. Bearing that in mind, in this work we have also adopted as proxy for a toxicity reference the

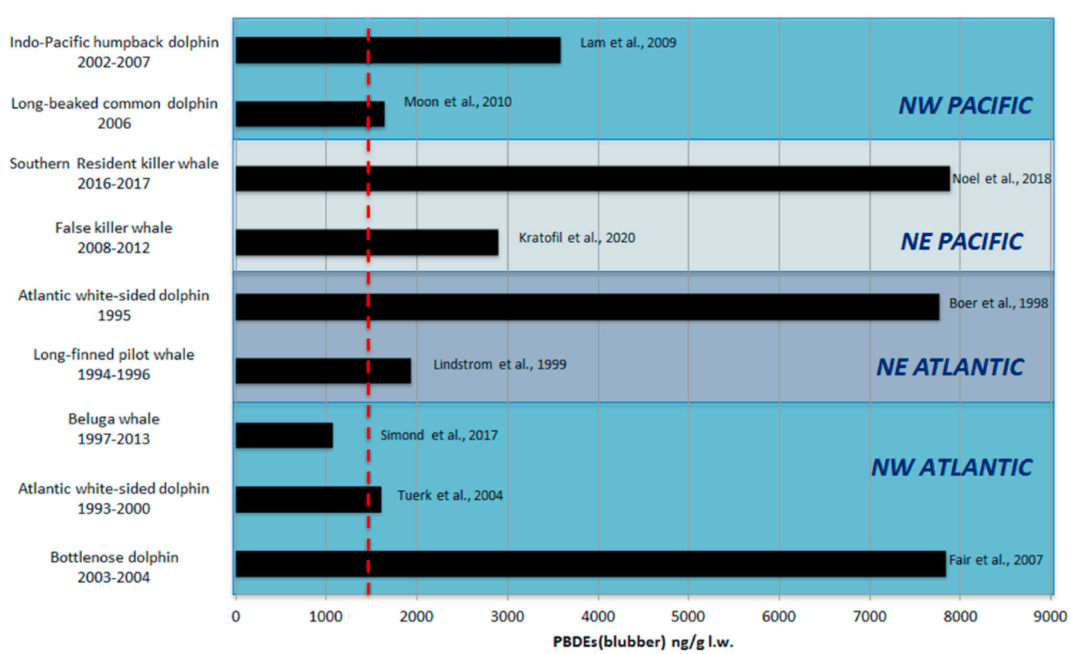


Fig. 2. PBDE highest mean concentrations (ng/g l.w.) in cetaceans' blubber around the world. Dotted line represents the threshold value for alteration of thyroid hormone levels in grey seals (Hall et al., 2003).

threshold established for alteration of thyroid hormone levels in grey seals (Hall et al., 2003).

Notable concentrations surpassing this threshold value of 1500 ng/g l.w., have been reported in liver of striped dolphin (*Stenella coeruleoalba*) (mean: 3624 ng/g l.w.; range: 129-8133 ng/g l.w.) and risso's dolphin (*Grampus griseus*) (mean: 2564 ng/g l.w.; range: 1778–2998 ng/g l.w.) from the Mediterranean Sea (Pettersson et al., 2004), and beluga whale in St. Lawrence Estuary (mean: 2210 ng/g l.w.; range: 246-3030 ng/g l.w.) (McKinney et al., 2006) (Fig. 3). Significant values of PBDEs have been also found in cetaceans inhabiting the highly industrialized and urbanized south-eastern coast of Brazil that represents a feeding and breeding ground for several coastal species such as the Guiana dolphin (*Sotalia guianensis*) (Ribeiro-Campos et al., 2021) and the highly threatened franciscana dolphin (*Pontoporia blainvillei*) (Secchi et al., 2021). Dorneles et al. (2010) found mean PBDE levels in liver of Atlantic spotted dolphins (*Stenella frontalis*), pantropical spotted dolphins (*Stenella attenuata*) and false killer whales of 1150 ng/g l.w., 1215 ng/g l.w. and 3600 ng/g l.w., respectively, which are in the same order of magnitude as those in Mediterranean dolphins (Pettersson et al., 2004) and Canadian Belugas (McKinney et al., 2006) (Fig. 3), dwellers of two well-known contaminated areas. This is in line with the results from a recent study performed in four distinct groups of Atlantic spotted dolphins, stranded between 2005 and 2015 in the Canary Islands, Azores, Caribbean Sea and São Paulo (Méndez-Fernandez et al., 2018). In this study among the four different populations, the highest concentrations of PBDEs in blubber were reported in specimens stranded along the coast of Brazil.

As shown in Fig. 4 the highest PBDE levels have been described for species living in the North Hemisphere and in many cases inhabiting or frequenting recognized contaminated areas such as the Mediterranean Sea, the North American East Coast or the North West Pacific zone. Cetacean species living in Southern Hemisphere waters, on the other hand, appear to be less investigated. The highest PBDE loads in that case have been reported for species inhabiting the Eastern coast of Brazil and Australia, but basically no information exists for the African Coast or the Indian Ocean.

In general, PBDE temporal trends in cetaceans showed remarkable increased concentrations from the late 1980s and earlier 2000s (Hoguet et al., 2013; Kajiwara et al., 2008, 2006; Lebeuf et al., 2004; Leonel et al., 2014; Ramu et al., 2006; Rotander et al., 2012) followed by a slight decrease or level off in subsequent years (Isobe et al., 2011, 2009; Jeong et al., 2020; Kunisue et al., 2021; Rotander et al., 2012; Simond et al., 2017). At the same time, an increased proportion of higher brominated compounds, such as BDE-154 and BDE-153, have been documented in more recent years (Jeong et al., 2020; Kunisue et al., 2021). This tendency probably reflects the regulatory measures implemented in 2009 for penta- and octa-BDE that lead to increased use and production of deca-BDE, only banned in 2019 (UNEP, 2022a, 2009). All of this translated into ongoing inputs of deca-BDE in the environment and a greater bioavailability of higher brominated congeners deriving from debromination processes of BDE-209 (La Guardia et al., 2007; Zhu et al., 2019).

As shown in Table 1, upward trends prior to 1990s and earlier 2000s have been reported in the Northwest Pacific Ocean in finless porpoises (*Neophocaena phocaenoides*) (South China) and melon headed-whales (*Peponocephala electra*) (Japan) (Kajiwara et al., 2008, 2006; Ramu et al., 2006); in the Northeast Pacific and Northwest Atlantic Ocean in beluga whales (Canada, Alaska) (Hoguet et al., 2013; Lebeuf et al., 2004); in the Northeast Atlantic Ocean in long-finned pilot whales (Faroe Islands), white-sided dolphins (Faroe Islands) and fin whales (*Balenoptera physalus*) (Iceland) (Rotander et al., 2012); in the Southwest Atlantic Ocean in franciscana dolphins (*Pontoporia blainvillei*) (Brazil) (Leonel et al., 2014); in Indo-Pacific bottlenose dolphin from South Australia (Weijs et al., 2020). On the other hand, only one study showed clearly downward trends during the 1990s and 2018s in Mediterranean striped dolphins (Aznar-Alemany et al., 2021), whereas Law et al. (2010) reported a decreased trend in harbor porpoises (*Phocoena phocoena*) from United Kingdom between 1998 and 2008. It is worth noting that the majority of these studies did not include BDE-209. This is, however, the dominant component of the deca-BDE mixture and in some cases one of the main BDE congeners of total PBDE contents (Koenig et al., 2013; Muñoz-Arnanz et al., 2011). As mentioned before, while lower brominated congeners such as those predominantly present in the commercial penta-BDE mixture or deriving from debromination processes are decreasing in some cetacean species (Aznar-Alemany et al., 2021; Rotander et al., 2012), increasing concentrations of higher brominated congeners have been reported for different species and areas (Jeong et al., 2020; Kunisue et al., 2021).

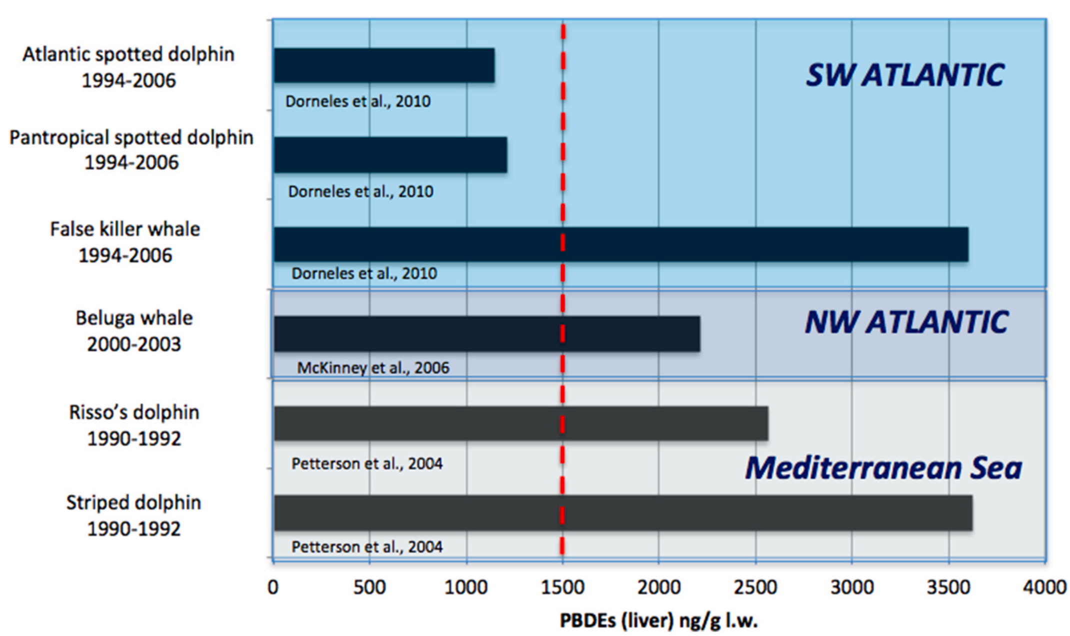
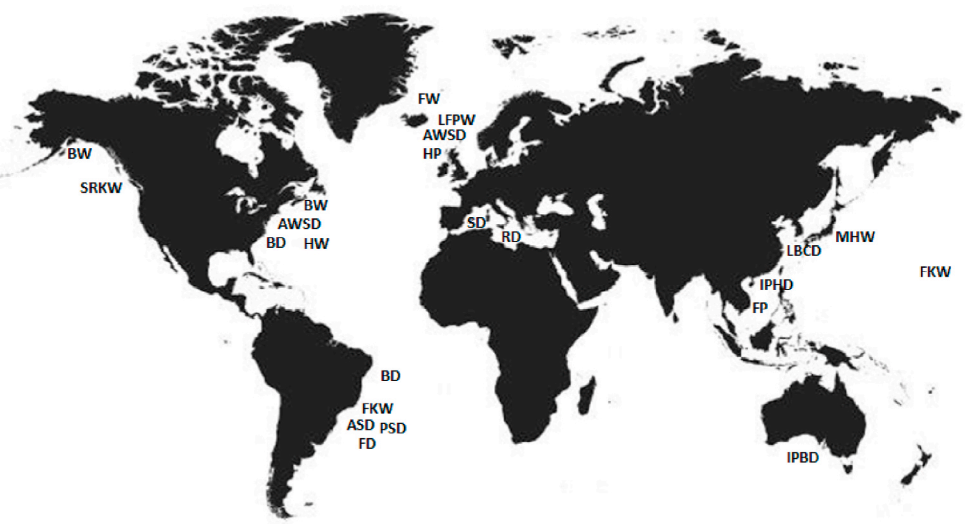


Fig. 3. PBDE highest mean concentrations (ng/g l.w.) in cetaceans' liver around the world. Dotted line represents the threshold value for alteration of thyroid hormone levels in grey seals (Hall et al., 2003).



ASD Atlantic spotted dolphin; **AWSD** Atlantic white-sided dolphin; **BD** Bottlenose dolphin; **BW** Beluga whale; **FD** Franciscana dolphin; **FKW** False killer whale; **FP** Finless porpoise; **FW** Fin whale; **HP** Harbor porpoise; **HW** Humpback whale; **IPBD** Indo-Pacific bottlenose dolphin; **IPHD** Indo-Pacific humpback dolphin; **LBCD** Long-beaked common dolphin; **LFPW** Long-finned pilot whale; **MHW** Melon-headed whale; **PSD** Pantropical spotted dolphin; **RD** Risso's dolphin; **SD** Striped dolphin; **SRKW** Southern Resident killer whale.

Fig. 4. Map showing locations of cetacean species analyzed in the studies discussed in this review.

Table 1
Most relevant PBDE trends in cetaceans worldwide, according to the levels found in blubber or *liver samples.

Species	Locations	Sampling years	Trends	References	
Finless porpoise	South China	1989–2006	↗ increased	Kajiwara et al., 2006[a]; Ramu et al., 2006[b]	NW Pacific
Finless porpoise	Korea	2003–2010	↘ decreased 2003–2010 ↔ stable 2010–2015	Jeong et al., 2020[c]	NW Pacific
Melon headed-whale	Japan	1982–2015	↗ increased 1982–2011 ↔ stable 2001–2015	Kunisue et al. (2021)[d]	NW Pacific
Melon headed-whale	Japan	1982, 2001, 2002, 2006	↗ increased	Kajiwara et al. (2008)[e]	NW Pacific
Beluga whale	Alaska	1989–2006	↗ increased	Hoguet et al. (2013)[f]	NE Pacific
Beluga whale	St. Lawrence Bay	1987–2013	↗ increased 1987–1997 ↔ stable 1997–2013	Lebeuf et al. (2014)[g]; Simond et al. (2017)[h]	NW Atlantic
Fin whale	Iceland	1986–1989, 2006, 2009	↗ increased	Rotander et al. (2012)[i]	NE Atlantic
Long-finned pilot whale	Faroe Islands	1986, 1997, 2006, 2007	↗ increased	Rotander et al. (2012)[i]	NE Atlantic
White-sided dolphin	Faroe Islands	1997, 2001, 2006	↗ increased 1997–2001/2002 ↘ decreased 2001/2002-2006	Rotander et al. (2012)[i]	NE Atlantic
Harbor porpoise	United Kingdom	1992–2008	↗ peakead around 1998 ↘ decreased until 2008	Law et al. (2010)[j]	NE Atlantic
Franciscana dolphin	Brazil	1994, 1996, 1998, 2000, 2002, 2004	↗ increased	Leonel et al. (2014)[k]	SW Atlantic
Bottlenose dolphin*	Brazil	1994–2012	↔ stable	Dorneles et al. (2010)[l]; Lavandier et al. (2019)[m]	SW Atlantic
Indo-Pacific bottlenose dolphin	South Australia	1989–2014	↗ increased 1989–2005 ↗ increased 2009–2014	Weijs et al. (2020)[n]	SE Indian
Stripped dolphin	Catalan coast	1990, 2004–2009, 2014–2018	↘ decreased trend	Aznar-Alemany et al. (2021)[o]	Mediterranean Sea

PBDE congeners investigated in each study.
[a,b,e] n. 3, 15, 28, 47, 99, 100, 153, 154, 183, 209.
[c] n. 17, 28, 47, 66, 71, 85, 99, 100, 119, 126, 138, 153, 154, 183, 184, 191, 196, 197, 206, 207, 209.
[d] n. 17, 28, 30, 47, 49, 66, 71, 77, 85, 99, 100, 119, 126, 138, 139, 140, 153, 154, 156/169, 171, 180, 183, 184, 191.
[f] n. 28, 47, 99, 100, 138, 153, 154, 155, 156, 181, 183, 190, 191, 203, 205, 206, 209.
[g]n. 28, 47, 49, 66, 99, 100, 153, 154, 155, 183.
[h] n. 7, 10, 15, 17, 28, 47, 49, 66, 71, 77, 85, 99, 100, 119, 126, 138, 139, 140, 153, 154, 171, 180, 183, 184, 191, 196, 197, 201, 203, 204, 205, 206, 207, 208, 209.
[i] n. 28, 47, 66, 85, 99, 100, 138, 154, 153,183.
[j] n. 28, 47, 66, 85, 99, 100, 138, 153, 154.
[k] n. 1, 2, 3, 7, 8, 11, 10, 12, 13, 15, 17, 25, 28, 30, 32, 33, 35, 37, 47, 49, 66, 71, 75, 77, 85, 99, 100, 116, 118, 119, 126, 138, 153, 154, 155, 166, 181, 183, 190.
[l] n. 28, 47, 66, 85, 99, 100, 153, 154, 183.
[m] n. 47, 100, 99, 154, 153.
[n] n. 47, 99, 100.
[o] n. 28, 47, 99, 100,154, 153, 183, 209.

As reported in Table 1, stable trends of PBDEs have been documented in cetaceans from Northwest Pacific, Northwest Atlantic, and Southwest Atlantic. In addition, increasing proportions of higher brominated congeners have been recently reported in cetaceans from Northwest Pacific. Jeong et al. (2020) investigated blubber of finless porpoises from Korean coastal waters during 2010–2015 and reported a significant decrease of lower brominated congeners (BDE-28 and BDE-47). At the same time, this study documented an increase of more brominated congeners (BDE-153 and BDE-154) during the study period, which is likely related to the greater consumption of deca-BDE in recent years. No significant variation on PBDE concentrations and relatively higher proportions of BDE-154 (equivalent to those of BDE-47) have been reported during 2011–2015 in melon-headed whales of the Japanese coast (Kunisue et al., 2021). Considering that technical penta- and octa- BDE products were banned by the early 2000s, the high proportion of BDE-154 – a minor component of these two mixtures - could be related as well to the increased discharge into the environment of BDE-209 and its debromination processes (La Guardia et al., 2007; Stapleton et al., 2006; Zhu et al., 2019). It is worth noting that China, one of the biggest countries in the Northwest Pacific area and one of the main producers and suppliers of deca-BDE mixture in the last years (Zhang et al., 2017), started the production of this higher brominated product later than Europe and the United States. This could have resulted in increased emissions of this mixture in recent years, and likely in a delay in the effectiveness of global restrictions (An et al., 2022). Even if China has established concentration limits for penta- and octa-BDE in electronics, it has not yet ratified the Stockholm Convention for deca-BDE (Sharkey et al., 2020) and its production, use, import and export will not be banned until 2025 (MEE, 2021). Additionally, China's great degree of industrialization and economic growth as well as e-waste import and inappropriate waste management might have contributed to elevated concentrations of these contaminants in its area, as reported by different studies (Li and Achal, 2020; Luo et al., 2021; Ma et al., 2011; Peng et al., 2019; Zhang et al., 2019). All these facts draw attention to the continuous inputs of PBDEs in the Northwest Pacific zone and the potential impacts on endangered cetaceans inhabiting this area for which information about PBDE contamination is scarce. To date, no studies have investigated PBDEs in North Pacific right whale (*Eubalaena japonica*), Western grey whale (*Eschrichtius robustus*), Northern sei whale (*Balaenoptera borealis*) and Northern blue whale (*Balaenoptera musculus*), all living or frequenting the Northwest Pacific Ocean and classified as endangered by the International Union of Conservation of Nature (IUCN) Red List of Threatened Species (Cooke, 2018a, 2018b; Cooke et al., 2018; Cooke and Clapham, 2018). In Yangtze finless porpoises (*Neophocaena asiaeorientalis asiaeorientalis*), an endemic species of the Yangtze River (China) listed as critically endangered by the IUCN (Wang et al., 2013), a declining population trend has been associated to a high risk of pollution particularly by pesticides. Yet, no information is present about the potential detrimental impact of PBDEs in this species (Zhao et al., 2008).

Continuous exposure to PBDEs has been also reported for St. Lawrence estuary belugas. This population seems to be exposed to continuous inputs of PBDEs as indicated by the high and stable levels of penta-, octa- and deca-BDE from 1997 to 2013, preceded by an increasing trend from 1987 to 1997 (Lebeuf et al., 2014; Simond et al., 2017) (Table 1). Elevated concentrations of PBDEs reported in different abiotic and biotic matrices in this area are to be related to the proximity of North America and the discharge of Montreal municipal effluents - the major source of PBDEs in the St. Lawrence River - along with the Great Lakes water mass (Marcogliese et al., 2015). In consequence, high values of PBDEs continue to be detected in different aquatic species and in a similar fashion as those of Canadian belugas (Travis et al., 2020; Zhou et al., 2019). Zhou et al. (2019) described an increase in PBDE concentrations in predator fishes of the Laurentian Great Lakes from 1990 to 2000, rapidly decreasing concentrations from 2000 to 2007, and unchanged or slightly decreasing levels up to 2015. This tendency probably reflects the voluntary phase out of penta- and octa-BDE commercial mixtures in 2004 and the extensive use of deca-BDE in the following years resulting in increasing concentrations of brominated congeners such as BDE-100 and BDE-154, which possibly stem from debromination processes of BDE-209. This thesis is supported by the increased proportion of BDE-209 - from 72% to 95% - of the total PBDE burdens after 2013 (Zhou et al., 2019).

An increase of PBDE levels was observed from 1989 to 1995 (range: 80–91 ng/g l.w. blubber) and from 2009 to 2014 (range: 23-1851 ng/g l.w. blubber) in Indo-Pacific bottlenose dolphins (*Tursiops aduncus*) from Spencer Gulf and Gulf St Vincent (South Australia), with a difference of at least one order of magnitude between the two study periods (Weijs et al., 2020). Concentrations were in general higher than those reported for other odontocetes from Australia, normally ranging between 4.3 and 440 ng/g l.w. in blubber (Symons et al., 2004; Weijs et al., 2016). These data are not consistent with the results reported for human serum blood in the Australian population, which showed a constant lowering of PBDE levels during 10 years (Drage et al., 2019; Toms et al., 2018). Yet sediments from the Sidney estuary confirmed increased concentrations of these substances in the marine environment from 1990 to 2014, with BDE-209 reaching 97% of the total PBDE concentrations in more recent years (Drage et al., 2015). Prohibition of penta- and octa-BDE and the consequent increment in the use of deca-BDE could be responsible for the rapid increase in the average contribution of BDE-209 to the total PBDE content in sediment. Australia has never manufactured penta- and octa-BDE mixtures and its importation was banned in 2005 (Toms et al., 2009). Despite this, import of PBDE-containing products manufactured elsewhere continues to take place in the country (Drage et al., 2019).

Two different studies on male bottlenose dolphins stranded in Southeast Brazilian coast during 1994–2006 and 2007–2012 reported mean PBDE levels in liver of 982 ng/g l.w. and 960 ng/g l.w., respectively (Dorneles et al., 2010; Lavandier et al., 2019). Even if comparison and interpretation of data generated from different studies is not straightforward, owing to the different variables involved (sampling procedures, analytical methods employed, biological factors, etc.), it suggests that levels of PBDEs in bottlenose dolphins of Southeast Brazil have remained unchanged for nearly 20 years (1994–2012). PBDEs have never been manufactured in Brazil; nevertheless, this country is one of the major global electronic and textile waste producers (Baldé et al., 2017), commercial sectors where large amounts of these substances are employed. In this region, almost 215 thousand tons of municipal solid waste is generated daily and in a great proportion (40%) inadequately disposed (ABRELPE, 2017). This, together with the absence of specific legislation limiting the concentrations of PBDEs in commercial waste goods and a lack of regulation on the import/export of these BFRs (Rodrigues et al., 2015) could favor constant releases of these substances. Moreover, the existing exemptions for the use of penta-/octa-BDEs and deca-BDE in certain applications in Brazil, which will expire in 2030 and 2036, respectively, are likely to delay future decreases in the environmental concentrations of these contaminants (Sharkey et al., 2020).

1.2. Does climate change have an impact on PBDE accumulation in cetaceans?

In recent years, different studies have focused on the influence that climate change-driven processes could have on POPs' fate and distribution and the consequent impact on the global environmental pollution and living organisms (Alava et al., 2017; Chen et al., 2019; de Wit et al., 2022; Gong and Wang, 2022; Hung et al., 2022; Li et al., 2021; Nadal et al., 2015; Potapowicz et al., 2019). It is documented that changes in environmental variables and meteorological conditions brought about by global warming may affect POPs' behavior in the environment and may alter the "grasshopper effect" that is strictly dependent on temperature (Chen et al., 2019; Dalla Valle et al., 2007; Li et al., 2021; Ma et al., 2011; Wania and MacKay, 1996). Enhanced volatilization of POPs between primary and secondary sources, faster degradation in the aquatic ecosystem and distorted partitioning between geochemical spheres, are some processes caused by climate change and potentially determining POPs' fate and distribution (Cai et al., 2014; Macdonald et al., 2005; Nadal et al., 2015; Noyes et al., 2009). A recent study by Gong and Wang (2022) has drawn attention to the increased contribution of "secondary sources" to global contamination and the accelerated cycling of POPs under climate change conditions. It is plausible that increased temperatures are forcing the re-volatilization of POPs stored in pools (glaciers, vegetation and soils), thus converted in secondary sources responsible for the re-emissions of these contaminants into the global cycle (Dalla Valle et al., 2007). Secondary sources represent a threat above all for cold-climate ecosystems, more sensitive to climate change and where increased long-range transport might contribute to accumulation of POPs (IPCC, 2021; Li et al., 2021; Wania and MacKay, 1996). In addition, cryosphere degradation observed both in the Northern and Southern Hemispheres during the last decades could represent another important secondary source of POPs, previously trapped into it (Chadburn et al., 2017; Grannas et al., 2013; Siegert et al., 2019; Stroeve et al., 2012; the IMBIE team, 2018).

As sentinels of marine ecosystem pollution, cetaceans could be affected by changes in POPs' distribution, revealing alterations in the concentration and accumulation pattern of these substances over time. On the other hand, some studies demonstrated alterations in cetaceans' distributions, habitat and migration due to climate change that in some cases could result in increased residence times in more polluted areas (Ramp et al., 2015; Storrie et al., 2018; van Weelden et al., 2021).

Global warming may have an important impact on POPs' occurrence especially to those related to sediments and soils such as BDE-209, which accounts approximately for 75% of the worldwide use of PBDEs (Shi et al., 2015). Increased rainfalls may enhance POPs' deposition into soil, while more intense precipitations and land runoffs could raise the quantity of eroded soil particles reaching the ocean (Dalla Valle et al., 2007). Thus, increased amounts of these toxic and persistent substances could reach the marine environment entering aquatic food webs throughout benthonic organisms feeding on surface sediments or within them. At the same time, increased precipitation and atmospheric aerosols could enhance POPs' deposition to aquatic ecosystems (Castro-Jiménez et al., 2017; Macdonald et al., 2003; Noyes et al., 2009). A high contribution of BDE-209 (70–75% of the detected ΣPBDEs) has been documented in zoo- and phytoplankton as a possible consequence of the constant atmospheric input through dry deposition (Castro-Jiménez et al., 2021; 2017).

Even if BDE-209 is highly hydrophobic with low water solubility that makes it hardly bioavailable, studies have documented accumulation and biomagnification of this congener in cetaceans' tissues (Aznar-Alemany et al., 2019; Noël et al., 2009). It has been also demonstrated that an increase in environmental temperatures and atmospheric concentrations of ozone driven by global warming could enhance PBDE degradation and generation of more toxic and persistent lower brominated congeners (Niu et al., 2015). Keeping in mind the huge historical production and its recent prohibition, deca-BDE is worthy of attention in the next future, above all in those countries where production and use continue, and in urbanized and industrialized coastal areas where great amounts of higher brominated congeners and associated metabolites could be continuously released. It could be argued that the quantity of BDE-209 and other brominated congeners entering the marine food web could increase in the next years due to meteorological changes. This fact could lead to an increase of concentrations in marine top predators such as odontocetes living in coastal areas. On the other hand, as mentioned above, global warming and changes in meteorological conditions have been related to shifts in migration habits of some cetaceans. Thus, some of them began to spend more time in more contaminated lower latitude regions and to act as vectors of contamination to less polluted areas (Borgå et al., 2022; Ramp et al., 2015; Storrie et al., 2018). Species that spend longer periods than usual feeding at southern latitudes are probably increasing their exposure to these substances, as it has been already reported for some seabird species (Bustnes et al., 2010; Elliott et al., 2021; Mora et al., 2016).

A study by Ramp et al. (2015) assigns the earlier ice break-up, caused by global warming, as the plausible cause for fin whales anticipating their date of arrival and increasing their residence time (in about 16 days) in the summer feeding area at St. Lawrence Estuary. This is a well-known contaminated area situated in the North Atlantic Ocean and where the St. Lawrence River pours its waters after receiving PBDE contaminated urban effluents from the Metropolitan Community of Montreal (3.8 million inhabitants) (Marcogliese et al., 2015). Cumulative effects of changes in migrations habits, more intense precipitations and continuous emissions of PBDEs above all in industrialized coastal areas, can result in greater PBDE exposures for fin whales (Ramp et al., 2015). Moreover, higher brominated BDE congeners accumulate preferentially in lower trophic levels such as zooplankton, compartment that includes the main prey of this species and other baleen whales (Castro-Jiménez et al., 2021).

The Cook Inlet Alaskan beluga whale population has increased its residence time in the more industrialized and urbanized upper Cook Inlet during warmer years, suggesting a risk of toxicological implications for this endangered population that is already facing a growth rate reduction and difficult recovery (Ezer et al., 2013). Furthermore, if long-term climatic changes occur in the future, Cook Inlet belugas whale could stay longer in the northernmost part of the inlet facing higher levels of contamination. Hoguet et al. (2013) documented a different degree of PBDE exposure between two distinct populations of Alaskan belugas with the Cook Inlet beluga population being more contaminated than the Bering Sea population. As explained by the authors, the different degree of contamination is probably the consequence of distinct geographic localizations. Cook Inlet is an urbanized and industrialized area where localized inputs of contamination occur, for instance stemming from wastewaters from Anchorage's treatment plant serving more than 200,000 people.

Some species of baleen whales such as humpback whale, fin whale, common minke whale (*Balaenoptera acutorostrata*) and blue whale have increased their annual maximum latitudes between 1° and 4°, apparently due to the reduction of sea ice extent as a consequence of climate warming (Storrie et al., 2018). As cetaceans could act as biovectors of POPs, enhanced ice melt and the subsequent habitat expansion of these species might rise the transportation of persistent and toxic pollutants into polar zones. Furthermore, increasing temperature and sea ice melting could result in remobilization of organic contaminants, such as PBDEs, and increased concentrations in the marine environment (Ma et al., 2011).

1.3. *Effects of PBDEs in cetaceans' health: what is already there and what is lacking*

A limiting factor in risk assessment of PBDEs in cetaceans is the lack of species-specific toxicity thresholds that can be used to assess potential health risks of these substances. To the best of our knowledge the single upper limit threshold for PBDEs in marine mammals (1500 ng/g l.w.) has been established for endocrine disruption in blubber of grey seals (Hall et al., 2003). To date this threshold is employed for a wide range of species with some degree of approximation (Lam et al., 2009; Lindström et al., 1999; Moon et al., 2010; Tuerk et al., 2004). Toxicokinetic differences as well as distinct physiological characteristics lead to a high level of uncertainty when extrapolating a threshold value for different species, even if among marine mammals. Hence, there is a need for toxicological studies focused on calculating species-specific and tissue-specific threshold values, as well as to assess different endpoints, already settled for other POPs such as PCBs and dioxins (Desforges et al., 2016; Kannan et al., 2000; Ross et al., 1995; Schwacke et al., 2012). PBDEs lack of a unifying methodology like that of the Toxic Equivalent (TEQ) approach used in risk assessment for dioxins, and others dioxin-like compounds. Moreover, while a group of seven indicator congeners exists for PCBs, there is not a similar counterpart yet for PBDEs (Boalt et al., 2013). Despite the lesser attention given to PBDEs, it has been demonstrated that these substances could alter the normal activity of biological systems in the same way as PCBs and in some cases with a higher degree of interference. Studies in rats suggested agonist potency of PBDEs on the aryl hydrocarbon receptor (AHR) comparable to those of some mono-*ortho* PCBs (Meerts et al., 1998). Hooper and McDonald (2000) reported greater ethoxyresorufin-o-deethylase (EROD) induction capacity of commercial penta-BDEs compared to that of Aroclor1254. Potential additive reduction on circulating thyroxin hormone level (T4) induced by coexposure of PCBs and PBDEs have been described by Miller et al. (2012).

Besides being endocrine disruptors (ECD) (Hall et al., 2003; Zhou et al., 2001), PBDEs have been associated to immunotoxicity in harbor porpoises, pantropical spotted dolphins, sperm whales (*Physeter macrocephalus*), striped dolphins, fin whales, Bryde's whales (*Balaenoptera edeni*), killer whales, long-beaked common dolphins, and furthermore, genotoxicity and cytotoxicity in pantropic spotted dolphins (Beineke et al., 2005; Villanger, 2011; Hall et al., 2003; Huang et al., 2020; Marsili et al., 2019; Rajput et al., 2021; Simond et al., 2019). Thymic atrophy and splenic depletion have been significantly correlated with high PBDE concentrations in harbor porpoises (Beineke et al., 2005). Stimulation of innate immune response has been reported in fibroblast cells of pantropical spotted dolphins exposed to BDE-47, BDE-100 and BDE-209 (Huang et al., 2020). A recent study on fibroblast cells of pantropic spotted dolphin described alterations on reactive oxygen species (ROS) production, mitochondrial membrane potential, cellular calcium levels, mitochondrial structure, cell membrane structure and apoptosis, after exposure to BDE-47, BDE-100 and BDE-209 (Rajput et al., 2021). Also, alterations on a polymorphic protein that is induced upon stress, damage or transformation of cells, has been described in fibroblast cells of killer whale, long-beaked common dolphin, Bryde's whale, sperm whale, striped dolphin and fin whale, after treatment with PBDEs (Marsili et al., 2019). Besides, disruptive capacity to thyroid hormones (Villanger, 2011) and alteration of gene transcription involved on the regulation of thyroid and/or steroid hormones (Simond et al., 2019) have been documented in beluga and minke whales.

All these *in vitro* studies, performed mainly in fibroblast cells, have demonstrated the capacity of these contaminants to interfere with hormone production and the immune system response, to negatively affect the structure of the cells and their functions as well as to alter production of reactive oxygen species (ROS) resulting in oxidative stress. Furthermore, it has been shown that the hormone, cellular and immune systems respond to PBDE exposure in a specific way according to different types of congeners, species or tissue analyzed. For example, BDE-47, BDE-99 and BDE-100 have been reported to differently influence oxidative stress, mitochondrial membrane potential (MMP) response, cellular calcium levels and protein expression of apoptosis-associated genes, being BDE-47 the most cytotoxic one (Rajput et al., 2021). At the same time, Marsili et al. (2019) showed differences in the expression of polymorphic proteins in fibroblast cells exposed to a mixture containing 27 PBDEs in different marine mammal species; downregulation has been documented for killer whale and Bryde's whale, while upregulation has been reported for striped dolphin.

Toxicological sensitivity, mechanisms and pathways may vary between species and each congener could interfere with vital systems in multiple ways and at different levels. Therefore, species-specific investigations as well as a broader knowledge about toxicity effects triggered by different substances and congeners result crucial. However, if on one hand it is essential to study negative effects caused by mixtures, as cetaceans are normally exposed to different class of pollutants and various congeners simultaneously, on the other hand, studies on specific substances such as PBDEs could play a key role in management and conservation policies.

2. Conclusions

Despite all regulatory measures taken, PBDEs continue to impact cetaceans at global scale. Climate change and the constant economic growth together with the increase of e-waste trading could lead to higher exposure of cetaceans to PBDEs in the next years, especially in certain areas of the world such as the Arctic and the Northeast Pacific Ocean. Information about negative effects on the health status is still unknown for many species for which toxicological studies have not yet been performed. All of that represents a limiting factor for understanding the real impact exerted by PBDEs on these species both, at individual and population levels.

Canadian Southern Resident killer whales, Atlantic white-sided dolphins in the North Sea, bottlenose dolphins in the Northwest Atlantic Ocean, Indo-Pacific humpback dolphins inhabiting Hong Kong waters, Mediterranean striped dolphins and false killer whales from Hawaii and Brazil showed the highest tissue concentrations of PBDEs. Significant levels and stable trends of PBDEs recently reported in cetaceans of the Southern Hemisphere, where PBDEs have never been produced, highlight the environmental impact of the existing trade of PBDE-containing products. It is noticeable the urgent need for more carefully investigations and monitoring, especially in those countries that have not yet established concentration limits for imported products. The highest levels recently found in species living in urbanized coastal regions have been related to discharge of contaminated waters deriving from wastewater treatment plants, which should improve treatment processes and removal efficiencies of PBDEs. The synergic effects of climate change and ongoing emissions of PBDEs, expected to continue at least until 2050, may result in increased contamination exposure for some species, in particular cetaceans inhabiting the Arctic for which global warming seems to have a greater impact.

In light of this, a greater knowledge about effects both at species and population levels as well as more specific threshold values are strongly needed for cetaceans, in order to assess health risks and support regulatory protection for these species. The lack of generalized decreasing trends in total PBDE concentrations along with the reported changes in congener profiles underpin the need to reevaluate the efficacy of current regulations on PBDEs and to improve waste management, both in developed and developing countries.

Credit author statement

Alice Bartalini: Investigation, Writing - original draft, Juan Muñoz-Arnanz: Conceptualization, Supervision, Writing - review & editing, Natalia García-Álvarez: Writing - review & editing, Antonio Jesús Fernández: Writing - review & editing, Funding, Begoña Jiménez: Supervision, Writing - review & editing, Funding.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Data availability

All data are public and properly cited

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