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Review article

Organochlorine pesticides, brominated flame retardants, synthetic musks and polycyclic aromatic hydrocarbons in shrimps. An overview of occurrence and its implication on human exposure

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ABSTRACT

Shrimps are widely distributed in coastal areas, estuaries and rivers. Although this shellfish is a good source of nutrients, it can also accumulate environmental contaminants, such as organochlorine pesticides (OCPs), brominated flame retardants (BFRs), synthetic musks (SMs) and polycyclic aromatic hydrocarbons (PAHs). Due to their bioaccumulative properties, these pollutants are endocrine disruptors. In this review, an overview of the world's shrimp market, pollutants legislation and values found in shrimp samples will be discussed. Shrimps analysed from all continents showed the presence of contaminants, Asia being the continent with the highest values reported. The concentration values reached a maximum of 26100 ng/g wet weight (ww) for OCPs, of 226.45 ng/g ww for BFRs, of 12.1 ng/g ww for SMs and of 50650 ng/g ww for PAHs. Exposure data and risk, taken from different studies, are very variable and indicate that shrimp's consumption may represent a risk especially in certain geographic areas.

1. Introduction

Seafood is consumed worldwide and considered high quality food. The seafood market has experienced a constant growth in the last century, making it an important source of nutrients and energy worldwide. Shrimps are detritivores contributing to the breakdown of organic matter. They feed on phytoplankton and are a food source for larger animals. To the human population they are considered a delicacy [1, 2]. Shrimp is one of the most popular seafood in the world and they can be a healthy addition to our diet. Shrimps are low in fat and calories. They are composed of a larger amount of water (three-fourths of the edible portion), and primarily made of protein (80% of the remaining portion, dry matter). In fact, the average protein content of fresh shrimp is 19.4 g/100 g and it contributes to 87% of the total energy [3]. Shrimps have low levels of lipids, containing 65–70% phospholipids, 15–20% cholesterol and 10–20% total acyl glycerol. 32 % of the total lipid composition is made up of polyunsaturated fatty acids, a term usually associated with high-quality seafood. These lipids are rich in omega-3 fatty acids, especially eicosapentaenoic acid and docosahexaenoic acid [3, 4]. This shellfish is also a good source of key nutrients such as phosphorus, choline, copper, zinc, iodine, B-complex vitamins, vitamin A and E [3, 5, 6]. While the majority of the antioxidants we ingest originated from vegetables and fruits, shrimp is a good source of compounds with antioxidant properties such as selenium and astaxanthin. Astaxanthin has been found to be a potent natural antioxidant [3].

Nonetheless, seafood can also be a source of environmental contaminants due to the pollution of many coastal areas that affect marine ecosystems, biodiversity and fisheries. Shrimp is widespread and can be found near the seafloor of most coasts and estuaries, as well as in rivers and lakes. Hence, they live in the aquatic environments affected by lipophilic pollutants that have been accumulating over time. This raises a concern about their safety for human consumption. Organochlorine pesticides (OCPs) used in agriculture, brominated flame retardants (BFRs) routinely added to products in order to reduce their flammability, synthetic musks (SMs) used in personal care product fragrances and polycyclic aromatic hydrocarbons (PAHs) generated primarily during the

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incomplete combustion of organic materials are major chemical pollutants. The release of chemical pollutants in aquatic environments raises awareness about the health and environmental impact of these pollutants on seafood.

This study aims to review the world's shrimp market, legislation on OCPs, BFRs, SMs and PAHs, studies published from 2004 to 2020, reporting the occurrence of OCPs, BFRs, SMs and PAHs in shrimps, the possible effects of these on human health and an evaluation of risk assessment.

2. The status of world's shrimp production, importation, exportation and consumption

Regarding shrimp importation, the top five importers in 2017 were the United States (US), the European Union (EU), Vietnam, China and the Republic of Korea [7]. In terms of production and exportation, China remained the largest producer of farmed shrimp in the world. However, most of China's harvest remains in the domestic market. India's shrimp industry, on the other hand, mainly exports and is the world's second largest producer of farmed shrimp. The top two world exporters in the first half of 2017 continue to be India and Ecuador, with market values increasing in supplies 35% and 18%, respectively. Among the other top exporters, Vietnam and China reported higher shipments January–June 2017, whereas exports declined in Thailand [7].

The average annual *per capita* worldwide consumption of shrimp in 2013 was 1.3 kg. According to the FAO database and trace statistics a total of 9,129,021 tons of shrimp was consumed worldwide in 2013. Asia is the largest consumer of shrimp in the world, with China and Japan accounting for 4,035,409 tons and 661,624 tons, respectively. The America continent is in second place, responsible for 19.6% of the world consumption, the USA being the country with the highest shrimp consumption (1,287,094 tons). Although, considering the amount of shrimp consumed *per capita* in 2013 (Figure 1) Australia & New Zeeland surpassed Asia with 2.29 kg/*per capita* consumption versus 1.45 kg/*per capita*. In second place, once more, is the America continent with 1.85 kg/*per capita*. Interestingly, Norway was in 2013 the country with highest *per capita* consumption, achieving 9.38 kg/*per capita*, followed by Japan with 5.20 kg/*per capita* and USA with 4.02 kg/*per capita*. Shrimp consumption *per capita* was rising in the Netherlands, the Republic of Korea and China between 2000 and 2013, it has been declining in Japan since 2005 and in Spain since 2006. In Norway the consumption oscillated until 2008, when it continually increased. In Portugal the consumption in 2013 reached 1.4 kg/*per capita*, values similar to Belgium and Italy. All this reported data emphasizes the importance of shrimp in the global economy [8, 9].

3. Pollutants

The rapid industrialization and urbanization have led to severe pollution of environmental matrices. New products are being developed without any regard to their long-term potential risk to humans and the environment. Coastal regions are developing rapidly due to commercial, agricultural, and industrial activities, leading to an increase in the contamination of the aquatic environment, affecting those locations and the biota living there. Shrimps are wildly consumed all over the world as described above, with the increase in coastal pollution, the impact on human health is an ever-increasing concern.

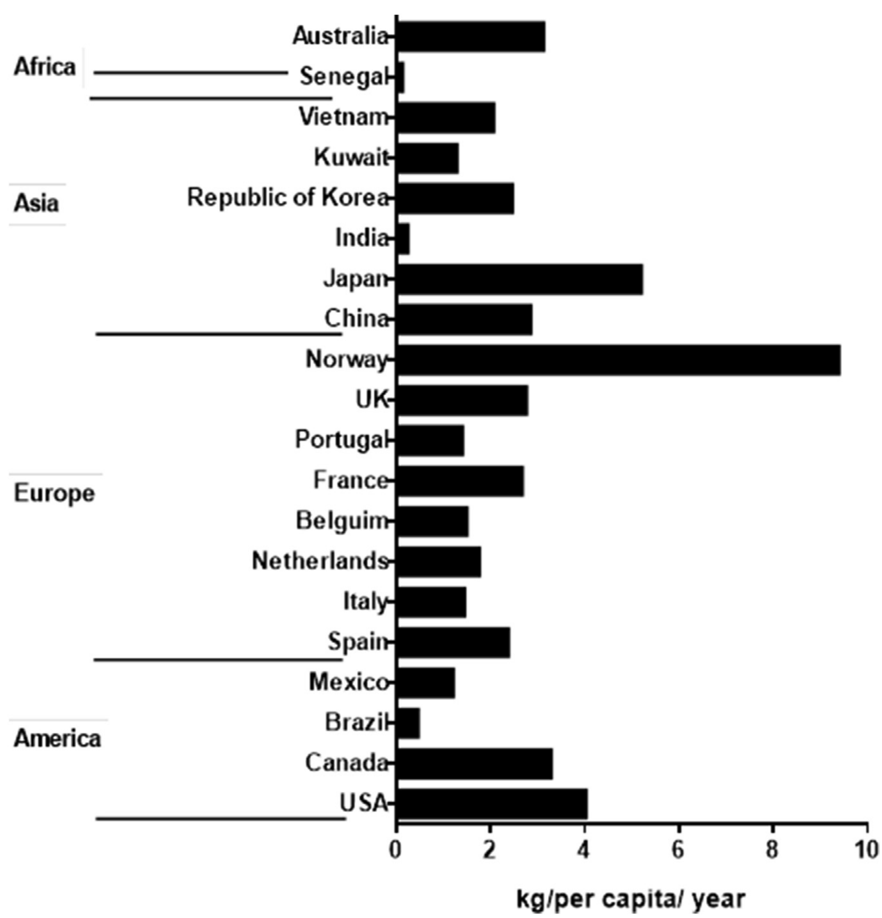

Figure 1. Shrimp estimated consumption in 2013 divided by countries adapted from [8, 9].

3.1. Organochlorine pesticides

OCPs have been used around the world as pest and insect control for more than half a century. The agricultural usage of most OCPs have been banned worldwide. However, these chemicals are hard to degrade, and hence capable of remaining in the environment for decades [10]. OCPs are chemical persistent, lipophilic, and hydrophobic compounds that can accumulate in biota, become biomagnified through the food chain, meaning that the concentrations in biota increase as the trophic level increases [11, 12]. In aquatic environments, hydrophobic compounds such as OCPs can enter shrimps, mainly via two pathways: bioconcentration, directly through the water environment and/or biomagnification, through food web preys. Lipid content, depuration rates, size and time of exposure of the organism, as well as the structure of the food and the environmental chemical concentrations are all potential factors influencing bioaccumulation of these compounds in aquatic organisms [10]. These compounds have clearly demonstrated to induce metabolic alterations in human cell lines and be harmful to humans. In fact, continuous exposure to them can cause several adverse health effects, such as, endocrine disrupting effects, cancers, neurodegenerative disorders, respiratory disorders, reproductive disorders [13], thyroid dysfunction, immunological disorders [14], increase risk for obesity and type 2 diabetes [15].

3.2. Brominated flame retardants

BFRs are applied in high quantities to reduce the flammability of polymers used in indoor applications and products including electronics, vehicles, plastics and textiles. This group of chemicals consists of tetrabromobisphenol A (TBBPA), polybrominated diphenyl ethers (PBDEs) that comprises 209 congeners, polybrominated biphenyls (PBBs), and hexabromocyclododecane (HBCDD). BFRs is a huge and complex chemical family, with molecular weights varying from 249 for Mono-BDEs to 959 for decabromodiphenyl (DecaBDE). These compounds are also, lipophilic and persistent in the environment, they have the capacity to accumulate in animal fats including aquatic species. The harmful health effects of these chemicals can be related to their persistency, bioaccumulation and biomagnification potential through the food chain [16, 17]. PBDE is one of the most studied group of BFRs, this group is widely detected in human tissues, such as blood and breast milk [17]. BFRs can act as endocrine disruptors and the continuous human exposure to them is associated with several disorders, including diabetes [16], cancers, neurological effects, thyroid disorders [17] and reproductive disorders [18].

3.3. Synthetic musks

SMs have been used over the years. Nowadays large quantities of SMs are manufactured and used as fragrance additives and fixative in a great variety of personal care products and household products, such as perfumes, shampoos, lotions, deodorants, soaps, and detergents. SMs can be divided into four groups: nitro, polycyclic, macrocyclic and alicyclic, among them polycyclic and nitro musks are dominant in terms of production volume; comprising 61% and 35%, respectively, of the total amount of SMs produced in the world. Due to their highly lipophilic nature, SMs tend to accumulate in sediments, sludge, and biological tissues, such as blood, breast milk and adipose tissue [19, 20, 21]. Little is known about the biological effects of SMs in humans after a prolonged exposure. The health concerns associated with SMs are endocrine disruptor effects, cancers and neurological disorders [22].

3.4. Polycyclic aromatic hydrocarbons

PAHs have at least two fused benzene rings in linear, angular, or cluster arrangements. They are a group of chemicals that usually occur naturally and generally appear as complex mixtures, not as single compounds. PAHs are primarily derived from incomplete combustion or pyrolysis of organic materials, for instance when coal, oil, gas, wood, garbage or tobacco are burned [23]. PAHs enter the marine environment through several mechanisms including atmospheric deposition, discharge of industrial sewage, marine transport, terrestrial runoff, and petroleum spills [24]. PAHs are distributed among different trophic levels through bioaccumulation processes. They start to accumulate in sediment, a contamination source to the surrounding water and aquatic biota. Aquatic organisms such as shrimp, are capable of concentrate pollutants directly from sediments and water and transfer these pollutants through the food web [25]. In terms of human exposure, PAHs have been reported to be carcinogenic, mutagenic and teratogenic [24]. The most significant endpoint of PAH toxicity after long term exposure is cancer, they can lead to proliferation of mutated cells resulting in cancer growth [26]. Furthermore, they have been reported to have endocrine disruptor capacity [27]. PAHs are also linked to adverse neurobehavioral effects, reproductive disorders and the prenatal exposure to them is associated with adverse health effects on children [28, 29]. Recent studies regarding the interaction of PAHs and obesity, reported that obesity may enhance the effect of PAHs exposure contributing to a greater risk for diabetes [30].

4. Legislation

The Stockholm Convention concerning persistent organic pollutants was adopted on the 22nd of May 2001 in Stockholm, Sweden [31]. OCPs such as, aldrin, endrin, chlordane, dichlorodiphenyltrichloroethane (DDT), heptachlor, mirex, toxaphene and hexachlorobenzene (HCB), have been banned for agricultural or domestic usage in many European, North American and South American countries since 1970s–1980s [32]. However, some specific OCPs are still allowed to be used in some countries. One example is DDT that is still used to prevent spreading of malaria and other vector-borne diseases such as dengue, leishmaniosis and Japanese encephalitis through the prevention of mosquito growth. Another example of widely used OCP is lindane (γ-HCH), this OCP had been used to treat head lice in children [33].

As for BFRs, some of them are already restricted in the EU in order to protect the health of the environment and the human population. However, due to their persistence in the environment there are still concerns about the risks that these chemicals pose to public health. The EU has directives to control, reduce or stop the sale and use of some BFRs. Directive 2003/11/EC, amends Directive 76/769/EEC on the marketing and use of certain dangerous substances and preparations, bans the sale of two commercial mixtures of PBDEs, known as PentaBDE and OctaBDE, in concentrations higher than 0.1% by mass. As of July 2006, under Directive 2002/95/EC, all new electrical and electronic equipment can no longer contain PBBs and PBDEs in any concentration. In July 2008, DecaBDE, originally exempted from the restrictions, was also banned by the European Court of Justice [34]. In the US, Washington State's law ESHB 2545 mandates that after July 1, 2017, no children's products may be sold within the state containing more than 1,000 ppm of DecaBDE, HBCDD and TBBPA [35].

In the case of SMs the Registration, Evaluation, Authorization and Restriction of Chemicals (REACH) regulation, has classified in 2008, musk xylene as a substance of high concern with a very persistent and very bioaccumulative designation. A restricted use warning was placed on musk ketone. Nitro musk compounds do not degrade easily, being highly stable and ubiquitous in the environment. Therefore, nitro musks have been banned in several countries and replaced by polycyclic musks. Nevertheless, nitro musks are still being produced in China and India and used in non-cosmetic compounds. The US has no restrictions to the use of SMs [21].

Lastly, PAHs have restricted regulations too. In this group more than 200 PAHs are known, in which, 16 PAHs were classified as priority pollutants by the EU and US Environmental Protection Agency (EPA) [36]. The EU has regulations regarding the levels of PAHs in food.

Regulation (EU) No 835/2011 amending Regulation (EC) No 1881/2006 regards maximum levels for PAHs in foodstuffs. Regulation (EU) No 1327/2014 amending Regulation (EC) No 1881/2006 as regards maximum levels of PAHs in traditionally smoked meat and meat products and traditionally smoked fish and fishery products. Regulation (EU) 2015/1125 amending Regulation (EC) No 1881/2006 as regards maximum levels for PAHs in Katsuobushi (dried bonito) and certain smoked Baltic herring. Regulation (EU) 2015/1933 amending Regulation (EC) No 1881/2006 as regards maximum levels for PAHs in cocoa fiber, banana chips, food supplements, dried herbs and dried spices [37].

Several guidelines have been implemented by different countries and continents to protect water of natural resources from pollutants. Water protection has been considered a top priority for some time for human consumption. However, the protection of the environment, the water resources and consequently the biota that lives in aquatic environment have been a priority as well. Table 1 resumes guidelines of quality standards for some pollutants in natural waters from different geographical areas, such as EU [38], US [39], Australia [40], South Africa [41] and Vietnam [42]. Vietnam has guidelines with the highest values of OCPs allowed. This data can explain some of the differences in the pollutant's concentrations found in shrimp from different geographic areas.

5. Occurrence of pollutants in shrimps

For this review article, a thorough search on Web of Science database from 2004 to 2020 was performed. In this search a total of 33 original research articles were found for OCPs, 37 for BFRs, 3 for SMs and 24 for PAHs. When possible, the reported concentrations were converted to the same unit (ng/g ww), allowing fair comparison.

5.1. Occurrence of OCPs

Table S1 (supplementary data) summarizes published studies from 2004 to 2020 regarding shrimp contamination with OCPs [43, 44, 45, 46, 47, 48, 49, 50, 51, 52, 53, 54, 55, 56, 57, 58, 59, 60, 61, 62, 63, 64, 65, 66, 67, 68, 69, 70, 71, 72, 73, 74, 75, 76]. The most studied OCPs in these publications are hexachlorocyclohexanes (HCHs), DDT, dichlorodiphenyldichloroethylene (DDE), and dichlorodiphenyldichloroethane (DDD). HCHs reached values as high as 17.84 ng/g wet weight (ww) in shrimp species from Cauvery River, India. In this location the large number of industries and agriculture activities can contribute to the high pollution [46]. An exceptionally high value, 26100 ng/g ww, was found in *Penaeus monodon* from Kolleru Lake, India [71]. For DDE the values ranged from ND to 143 ng/g ww, the maximum value was detected in gei wai shrimp (*Metapenaeus sp.*) from Mai Po (a Ramsar site) in Hong Kong, China. Domestic and industrial effluent discharged in the Pearl River Delta, have been a principal pollution source of this Ramsar site [54]. For DDD values ranged from ND to 14.83 ng/g ww, maximum value was detected in *Exopalaemon modestus* from Qiantang River, China. Upstream of the river, in Lanxi, a pesticide factory still produces OCPs which is a potential pollution source in the region [76]. As for DDT values were between ND and 16 ng/g ww in Chinese prawn from Yellow Sea, China. The Yellow Sea has been suffering with the rapid industrial and agricultural development [48]. An exceptionally high value was found in *P. monodon* (9800 ng/g ww) from Kolleru Lake, India [71]. The Σendosulfan, reached values as high as 7.31 ng/g ww, this value was detected in a *Litopenaeus spp.* sample from Pozo-Rey estuary. Agricultural practices of the region and sanitary campaigns for mosquito control may contribute to the pollution in this estuary [66]. An exceptionally high value was detected in *P. monodon* sample (27800 ng/g ww) from Kolleru Lake, India. As potential sources of pollution in this lake are the direct discharges of industrial effluents, sewage and agricultural wastes. The concentration of OCPs found in shrimps from Kolleru Lake were higher than the allowable limits for human consumption recommended by FAO/WHO [71]. A total of 31 OCPs were analysed in shrimp samples from the studies shown in Table S1 (supplementary data). The number of samples analysed in these studies vary between 2 [74] and 205 [64]. Some studies indicated the individuals collected varying between 9 [63] and 180 [70], and the specimens pooled per analyses ranged from 5 [68, 76] to 30 [67]. Various authors collected the samples by purchasing them at the local markets [47, 48, 50, 52, 59, 61, 65, 75]. Other samples were collected directly from their natural habitat, in some cases collected by

Table 1. Allowed values of quality standards (μg/L) for some pollutants in natural waters in different geographical areas.

Pollutants		European Union	United States		Australia		South Africa*	Vietnam
		Surface waters	Salt water	Fresh water	Salt water	Fresh water	Coastal and marine waters	Fresh Water
OCPs	Aldrin		1.3	3		0.01	0.003	
	Chlordane		0.09	2.4	0.004	0.01		
	DDT		0.13	1.1		0.0015	0.025	42
	Dieldrin		0.71	2.5		0.005	0.003	
	Endosulfan	0.004	0.034	0.22	0.001	0.003	0.0005	
	Endrin		0.037	0.18		0.002		
	HCB	0.05						
	Heptachlor	0.00003	0.053	0.52		0.005		18
	Lindane		0.16	2	0.004	0.01		56
	Methoxychlor					0.03		
BFRs	HBCDD	0.05						
	BDEs	0.014						
PAHs	Acenaphthene						20	
	Anthracene	0.1					0.1	
	Benzo(a)pyrene	0.027					0.00017	
	Benzo(b)fluoranthene	0.017						
	Benzo(k)fluoranthene	0.017						
	Benzo(g,h,i)perylene	0.00082						
	Fluoranthene	0.12					0.0063	
	Naphthalene	130					2	
Ref.		[38]	[39]		[40]		[41]	[42]

* In the case of South Africa, the values presented are chronic values, for the other geographical areas the value presented, represents the maximum allowed value.

professional fishermen [63, 68, 70, 73]. The sample catchment methods applied were trawl fishing [43, 49, 51, 62, 64] or fishing with the use of different nets, such as scoop net [71]. It is worth noting that Asia generally has higher levels of OCPs in shrimps than other continents (Figure 2). Although, we must consider that there are more studies available in this continent, which increase the chances to find contaminants. China is the country with the most studies conducted [48, 54, 57, 58, 60, 61, 64, 67, 68, 70, 74, 75, 76] contributing extensively to the larger number of papers in Asia and making the public aware of the risk of shrimp consumption. The next highest values found for ΣOCPs in shrimp were Egypt [65] and Mexico [66], followed by some European countries [43, 47, 49, 51, 73]. As possible contamination sources of the sampling areas the authors indicated industrial effluents and sewage [43, 45, 57, 71, 75, 76], agricultural activities [44, 45, 56, 60, 66, 71, 72], intensive shipping and fishing [60, 64] and sanitary campaigns for mosquito control [66]. Even in recent studies, shrimps analysed from several continents contain OCPs residues despite the limited usage imposed since the Stockholm Convention.

5.2. Occurrence of BFRs

Table S2 (supplementary data) summarizes published studies from 2004 to 2020 regarding shrimp contamination with BFRs [47, 49, 51, 55, 59, 62, 64, 68, 69, 70, 74, 75, 77, 78, 79, 80, 81, 82, 83, 84, 85, 86, 87, 88, 89, 90, 91, 92, 93, 94, 95, 96, 97, 98, 99, 100, 101, 102]. As shown in Table S2, 30 different PBDEs were analysed. This group has the most number of compounds. BDE209 was the one with the highest values, with concentrations ranging between ND and 226.45 ng/g ww, the highest value reported was in *Neocaridina denticulate* from Qingyuan city, China [81]. For TBBPA the values reached were 0.10 ng/g ww, being the highest value detected in *Neomysis integer* from Scheldt estuary (Schaar van Waarde), Netherlands [100]. Regarding ΣHBCDD values ranged from ND to 8 ng/g ww, with the highest value found in *Neomysis integer* from Scheldt estuary (Bath), Netherlands [100], the same estuary where the highest value of TBBPA were reported however at a different site. Regarding the number of samples analysed per study, the number varies between 2 [74,99] and 205 [64]. Being the number of individuals collected between 2 [96] and 180 [70], and the number of specimens per pool analysed between 7 [84–86] and 50 [97]. Concerning the sample collection, some authors reported that they purchased the samples at local markets [47, 59, 75, 78, 79, 82, 89, 91, 93, 95, 96] or directly from local fishermen [68, 70, 94]. The sample catchment methods applied were trawl fishing [49, 51, 62, 64, 77, 97, 101] and electric pulse fishing [68, 84, 85, 86]. As possible pollution sources near the sampling locations the authors indicated highly urbanized and industrialized areas [49, 51, 68, 70, 75, 78, 83, 87, 88, 89, 91, 93, 94, 97, 101], e-waste recycling areas [59, 70, 75, 81, 84, 85, 86] and intensive shipping and fishing [64]. Concerning ΣBFRs as we can see in Figure 3, the location with the highest value detected was in Qingyuan city, China [81]. Similar to what happened with OCPs, Asia still is the continent with the greatest amounts of studies performed in this field. In Europe some studies were performed [47, 49, 51, 95, 96, 97, 98, 99, 100, 101, 102] and the highest value was detected in the Netherlands [100]. These findings reveal that BFRs are already widespread and the legislators must be aware of them.

5.3. Occurrence of SMs in shrimp

Table S3 (supplementary data) summarizes published studies from 2004-2020 regarding shrimp contamination with SMs [20, 103, 104]. These are emergent pollutants that have been a cause for concern. Regardless of the few papers available in the last years, we can see that some SMs, such as Galaxolide (HHCB) and Tonalide (AHTN) are present in the shrimp samples in the three studies published. Emphasising the need for more studies on the existence and concentrations of SMs in shrimps worldwide. Regarding the number of samples collected the authors indicated 25 [104] and 33 specimens purchased at fish markets [20]. Sapozhnikova et al. suggested wastewater and sewage as possible contamination sources of SMs in the aquatic environment.

5.4. Occurrence of PAHs in shrimp

Table S4 (supplementary data) summarizes published studies from 2004-2020 regarding shrimp contamination with PAHs [24, 25, 36, 52, 54, 60, 69, 71, 105, 106, 107, 108, 109, 110, 111, 112, 113, 114, 115, 116, 117, 118, 119, 120]. As demonstrated in Table S4 the 16 PAHs that EPA listed as priority pollutants are the most studied ones. Among these

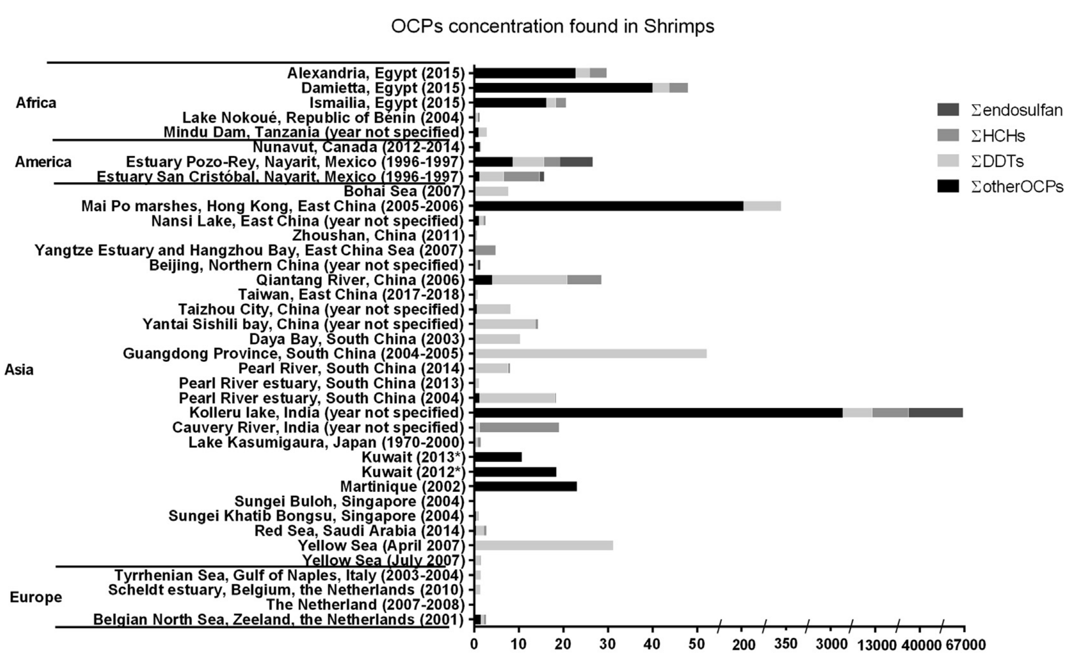

Figure 2. Reported concentrations of ΣOCPs found in shrimp samples, from 2004 to 2020. * studies performed by the same author. Year of sampling not specified. Presented year of publication.

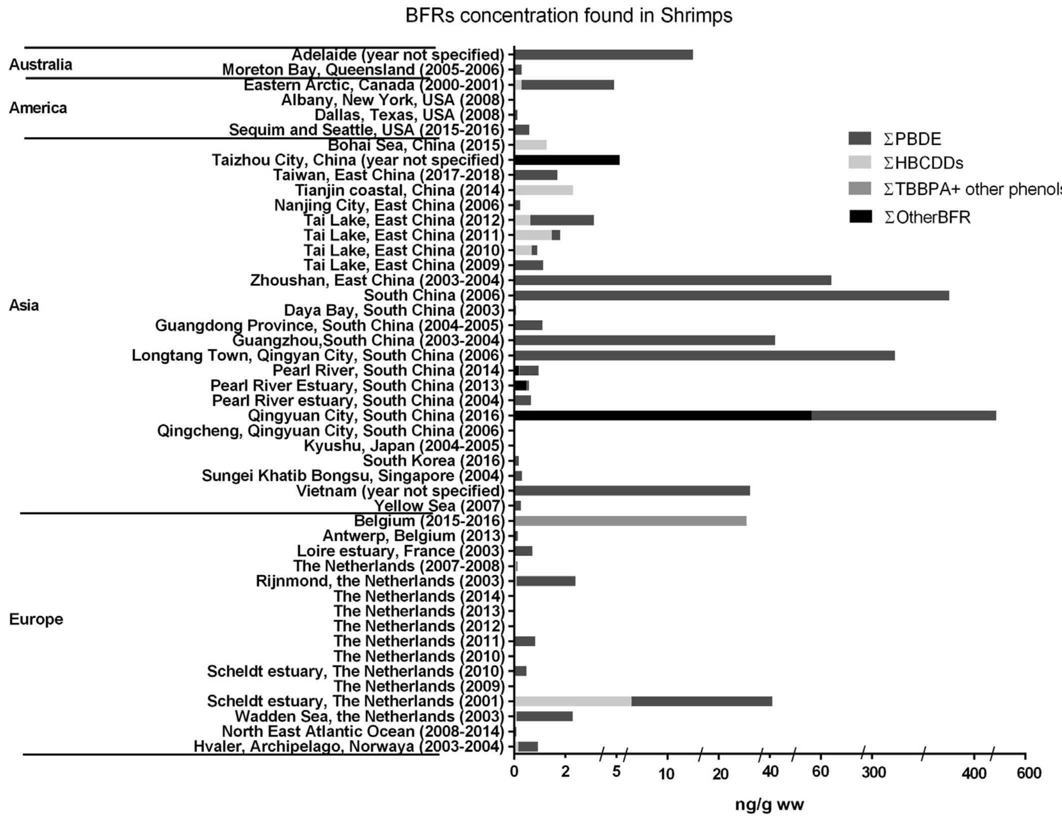

Figure 3. Reported concentrations of ΣBFRs found in shrimp samples, from 2004 to 2020.

PAHs, with a maximum value of (50650 ± 21880 ng/g ww) were found in *Penaeus notialis,* for naphthalene, in Lagos, Nigeria [115]. The most detected PAHs were from petrogenic origin, indicating that anthropogenic activities were influencing PAH concentrations in this area. With the exception of the values reported in Lagos, Nigeria [115], Asia is the continent with more studies and the highest values reported, as shown in Figure 4. The comparison between the concentrations found from two different places of the same species (*Penaeus monodon*), India [71] and Africa [117] shows that the habitat seems to be more important to PAHs accumulation in shrimp then the species itself. Moreover, the shrimps

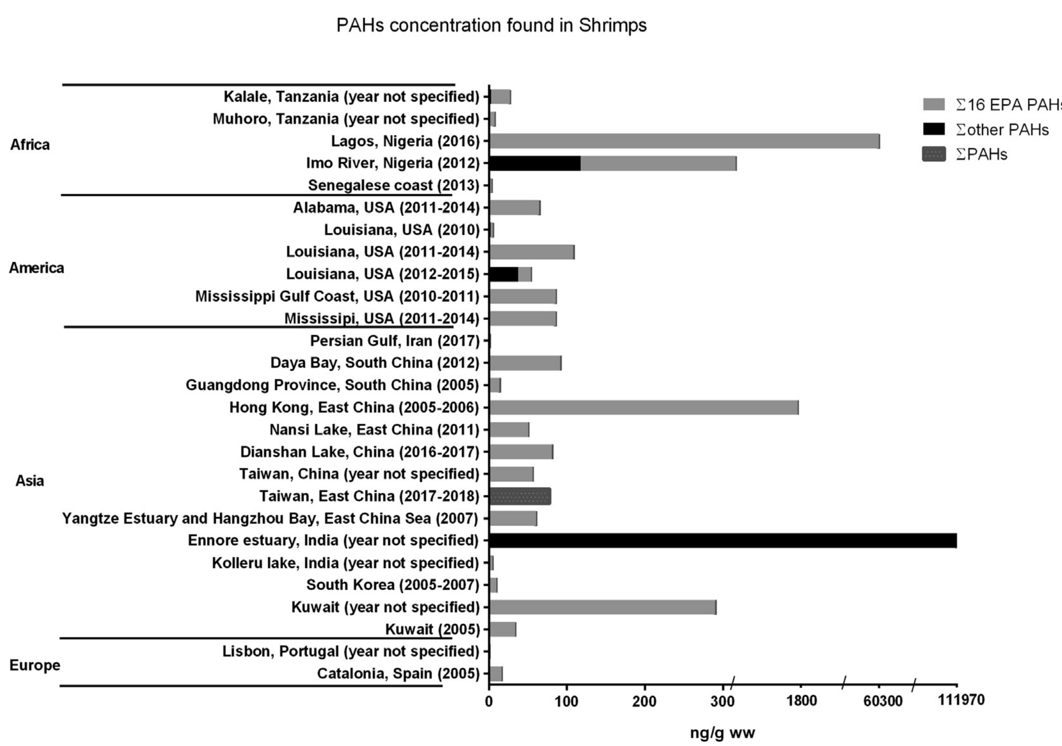

Figure 4. Reported concentrations of Σ16 priority PAHs found in shrimp samples, from 2004 to 2020.

sampled in estuary regions seem to be the mo contaminated ones [24, 36, 54, 60, 106, 109, 117]. For the PAHs evaluation the number of samples analysed varied between 3 [54,116,117,119,120] and 10 [24], the number of individuals collected ranged between 10 [117] and 300 [24]. The number of specimens pooled per sample ranged between 7 [109] and 200 [120]. Regarding the sample collection, some authors reported that the samples were purchased at local markets or directly from local fishermen [52, 105, 107, 108, 111, 114, 115, 119, 120]. The most applied sample catchment method was trawl fishing [24, 113, 116, 118] and one author reported the use of a scoop net [71]. As possible pollution sources of PAHs near the sampling locations the authors indicated highly urbanized and industrialized areas [54, 60, 71, 110, 117, 118], more specifically industry, such as petrochemical refineries, nuclear power stations and thermal power stations [24, 106, 109, 113], oil spills [105, 112, 115, 120], and specifically the deepwater horizon oil spill [36, 114, 116], another possible source of PAHs formation can be the grilling of food [108, 111].

6. Risk assessment

Until now only a few studies that reported levels of these lipophilic pollutants in shrimps included a risk assessment [56, 58, 59, 75, 82, 104, 107, 109].

Concerning the OCPs, a study conducted in Africa (Republic of Bénin) reported levels in shrimps and risk assessment data [56]. Calculated daily intakes (from shrimp) for ∑DDT and for α-endosulfan were of 0.65 and 0.15 ng/kg bw/day respectively. A toxic unit (TU) was also calculated by dividing the estimated daily intake of each pesticide by its tolerable daily intake. The sum of TU for the analysed pesticides was 0.00009, thus far below the value of 1 that would indicate possible health risks. The authors concluded that the risk for humans consuming shrimp from the lake or lagoon, was low [56]. Labunska et al. estimated adult and child exposure to selected organohalogen contaminants via different types of food at e-waste recycling sites in Taizhou, China. The daily intake of ∑DDTs for children and adults was 0.81 and 3.16 ng/kg bw/day, respectively. For HCB the calculated DIs were 0.06 and 0.22 ng/kg bw/day for adults and children. These exposure values were well below the tolerable daily intakes [59]. Some studies also presented a carcinogenic risk assessment for OCPs [58, 75]. To assess the potential health risk related to dietary exposure to DDTs, authors applied a method based on the benchmark concentration. In this method the hazard ratios (HRs) from the 50th and the 95th percentile assessed concentrations were considered. For the calculation of Benchmark concentration for carcinogenic effect it was used the USEPA cancer slope factor by setting cancer risk to one in one million due to lifetime exposure. The results showed that the 50th percentile cancer HRs for all age groups were significantly higher than one without significant differences amongst the different age groups. Therefore, dietary intake of DDTs via fish consumption were considered to cause a possible serious concern with a lifetime cancer risk. However, for shrimps, authors concluded that the risk was lower than one [75]. In another study, that applied the same calculation method to assess cancer risk, the HRs of DDTs and HCHs were 0.43 and 1.55, respectively. For the HCHs, results pointed that daily exposure to OCPs had a lifetime cancer risk of greater than one in one million [58].

For the BFRs, Miyake et al. [82] published results for two Chinese coastal cities, Guangzhou and Zhoushan. In their study, the authors determined daily intakes of 132 and 80.9 pg/kg bw/day for ∑PBDEs, of 1.20 or 0.483 9 pg/kg bw/day for ∑polybrominated dibenzo-p-dioxins (PBDDs) and of 0.934 or 0.619 for 9 pg/kg bw/day ∑polybrominated dibenzofurans (PBDFs), depending on the sampling location. The authors concluded that the results of the performed risk assessment indicated that exposure to lower brominated PBDEs and BDE 209 was unlikely to result in significant health risks. However, for the PBDDs/DFs it was considered that the determined levels could pose some health risks to the local population [82].

A study concerning SMs fragrances in seafood commercialized in several countries from EU was undertaken [104]. For the selected contaminants, no established health-based guidance values were available, therefore the authors calculated a Tolerable Weekly Intake (TWI_cal) value based on the available no-observed-adverse-effect level values. In these calculations, it was applied an uncertaint factor of 100 (to account for species differences and human variability). Considering all the studied countries, a mean value of 0.0024 μg/kg bw/week for HHCB and of 0.013 μg/kg bw/week for AHTN was estimated. These values were below the TWI_cal, which was also true even under a worst-case scenario (percentile 99). However, authors highlighted that southern European countries such as Spain and Portugal, due to a higher seafood consumption, presented higher exposure levels.

Hong-Gang Ni et al. reported risk assessment data for parent PAHs and halogenated polycyclic aromatic hydrocarbons (HPAHs) [107]. Shrimp was one of the major contributors for the total calculated intakes. Considering that the toxic equivalency quotient (TEQ) can be regarded as a better index for the potent toxicity than the concentration, authors also calculated the TEQ of PAHs and HPAHs. The excess cancer risk (ECR) induced by dietary exposure to PAHs and HPAHs via seafood consumption was also determined. The calculated mean TEQ of Σ_{16}PAHs was of 69.2 pg TEQ/g ww for shrimp. DahA (dibenzo[a,h]anthrancene) and B[a]P were the major contributors to the total TEQ of 16 PAHs. The median values of ECRs induced by 16PAHs for all subgroups (according to age and gender) were lower than the acceptable risk level. Thus, authors concluded that the results showed no significant cancer risk related to seafood consumption for people in South China [107]. Also, for PAHs cancer risk associated with consumption of shrimps in Nigeria was assessed through comparison of estimated daily intakes (EDI) and references values from US EPA and EFSA. The EDI for naphthalene and BaP were of 3 and 62.53, and of 4 and 84.25 ng/kg bw/day for adults and children, respectively. Therefore, the determined values were lower than the US EPA benchmarks and EFSA levels of concern values for adults and children population, suggesting a low probability of cancer development [109].

Even though, the results of the available studies pointed to levels of exposure that were below benchmark levels (or calculated), more studies are needed to better explain associated risks from pollutants exposure through shrimp consumption. This being particularly relevant for countries that have higher seafood consumption and consequently probable higher exposure levels for the respective population.

Considering the maximum values found in shrimps (Tables S1 from to and Table S4 supplementary data), respective average world shrimp consumption and average weight from an adult, a conservative risk assessment was performed. Consequently, results are presented by each group of compounds in Tables 2, 3, and 4.

For OCPs EFSA acceptable daily intakes (ADI) [121] were considered for risk assessment. Following our procedure, applying the maximum reported concentration values (worst scenario) the HQ values for a few pesticides exceeded the value of 1 (Tables 2, 3, and 4). Particularly, considering the extremely high concentration reported by Amaraneni et al [71].

To assess potential public health risks concerning PBDEs, maximum exposure concentrations were compared to minimal risk derived by Agency for Toxic Substances and Disease Registry following the same procedure as applied by Miyake et al [82]. A value of 0.5 was obtained, thus lower than 1. However, in one particular study exceptional high exposure values were reported in South China [84, 85] and if considered those values for risk assessment evaluation a value of 3.12 (Table 3) would be obtained. This value would indicate reasons for concern.

For SMs two compounds were considered HCBB and AHTN. Even considering the maximum reported values in the literature, risk assessment revealed no reasons for concern at the present moment. According to EPA a no-observed-effect level for oral dose of 10 000 μg/kg bw/day, with an uncertainty factor of 100, for HCBB was established. Considering the maximum reported values, a maximum of 0.00041 μg/kg bw/day

Table 2. Organochlorine pesticides hazard quotient.

Compound	Max. Value (ng/g ww)	Daily consumption (in 2013)	Average weight (kg) [123]	Calculated daily intake	Hazard risk#	References	EU ADI (μg/kgbw/day)
Chlordane	182*	3.58	62	10.509	**21.018**	[54]	0.5
Chlordane	1.05	3.58	62	0.061	0.121	[74]	
DDT/DDE	143	3.58	62	8.257	0.826	[54]	10
	9800*	3.58	62	565.871	**56.587**	[71]	
Heptachlor	4.53*	3.58	62	0.262	**2.616**	[66]	0.1
	2.97	3.58	62	0.171	**1.715**	[76]	0.1
Aldrin	2.03	3.58	62	0.117	**1.172**	[76]	0.1
Dieldrin	0.86	3.58	62	0.050	0.497	[76]	0.1
	3100*	3.58	62	179	**1790**	[71]	0.1
Endrin	2.94	3.58	62	0.170	0.849	[66]	0.2
	3.22*	3.58	62	0.186	0.930	[76]	0.2
Endosulfan	5.15	3.58	62	0.297	0.005	[66]	60
Endosulfan	27800*	3.58	62	1605.226	**26.754**	[71]	60

* Extemporaneous value, # bold values: Hazard risk ≥ 1.

Table 3. PBDEs hazard quotient.

	Compound	Max. Value (ng/g ww)	Daily consumption (in 2013)	Average weight (kg) [123]	Calculated daily intake	Hazard risk#	Reference
World	PBDEs	61.15	3.58	62	3.531	0.50	[82]
		378*	3.58	62	21.827	**3.12**	[84]

* Extemporaneous value, # bold values: Hazard risk ≥ 1.

Table 4. PAHs hazard quotient.

Compound	Max. Value (ng/g ww)	Daily consumption (in 2013)	Average weight (kg) [123]	Calculated daily intake	Hazard risk#	Reference	EPA RfD
acenaphthene	11.52	3.58	62	0.665		[24]	nd
naphthalene	55.5	3.58	62	3.205	0.160	[108]	20
	650*	3.58	62	37.532	**1.877**	[54]	20
fluorene	15.29	3.58	62	0.883	0.022	[25]	40
	63.7*	3.58	62	3.678	0.092	[54]	40
phenanthrene	37.92	3.58	62	2.190		[25]	n.a
	318.5*	3.58	62	18.391		[54]	nd
anthracene	9.84	3.58	62	0.568	0.019	[24]	30
fluoranthene	8.15	3.58	62	0.471	0.012	[36]	40
	188.5*	3.58	62	10.884	0.272	[54]	40
pyrene	17.38	3.58	62	1.004	0.033	[120]	30
	448.5*	3.58	62	25.897	0.863	[54]	30
benzo[α]anthracene	0.76	3.58	62	0.044		[25]	n.a
	39*	3.58	62	2.252		[54]	n.a
chrysene	2.08	3.58	62	0.120		[36]	n.a
	71.5*	3.58	62	4.129		[54]	n.a
benzo[*b*]fluoranthene	2.65	3.58	62	0.153		[36]	n.a
benzo[α]pyrene	6.35	3.58	62	0.367	**1.222**	[36]	0.3
benzo[*k*]fluoranthene	3.6	3.58	62	0.208		[36]	n.a
dibenz[α,*h*]anthracene	8.06	3.58	62	0.465		[36]	n.a
benzo[*g*,*h*,*i*]perylene	1.1	3.58	62	0.064		[25]	n.a
indeno[1,2,3-*c*,*d*]pyrene	3.71	3.58	62	0.214		[36]	n.a

* Extemporaneous value, # bold values: Hazard risk ≥ 1.

was calculated considering the average world daily consumption. The hazard quotient (HQ), obtained dividing the calculated DI by the NOAEL of 10 000 μg/kg bw/day was well below 1, which would indicate reasons for concern.

The non-cancer risk for PAHs associated with consumption of shrimps was assessed through comparison of estimated daily intakes based on maximum reported values and reference values from US EPA [122]. For the majority 16 EPA PAHs the calculated hazard risk values were below

1, thus not indicating reason for concern. However, for B[a]P Xia et al. reported a value of 6.35 ng/g in shrimp wet weight (6.35 μg/kg) [36] which resulted in a HQ of 1.22 (Table 4).

The risk assessment reported in this section, as mentioned, was based upon the highest reported values which can be regarded as the worst-case scenario. If median values are considered, generally no HQ values of ≥1 are achieved, which is in accordance with what has been reported by other authors [56, 59, 82, 104, 107, 109]. Average weight of 62 kg were considered for the general population [123].

For SMs no reference values are available thus values from [104] were taken into account. For PAHs risk assessment was performed as described by Dosunmu et al. [109] applying EPA reference values. For OCPs EU ADI reference values were taken into account. Hazard risk was calculated dividing the calculated daily intake by reference daily intake values; values equal or higher than 1 can indicate possible associated health risks.

7. Conclusion

This work is an overview on reported contamination levels of some lipophilic pollutants in shrimp samples from all over the world. A vast amount of published data on these pollutants' presence in shrimp species has been reported in recent years. Asia is the continent with more publications in this field and perhaps that's why this is the continent with the highest values of contamination in shrimp reported, in conjunction with the fact that this seems to be where the legislation regarding natural waters is not as restrictive. Guidelines of quality standards for natural waters have different values depending on the geographical area and government policies. These differences in guidelines and restriction laws can reflect different pollution values in waters and consequently in biota from different geographical areas of the planet. Pollutants were found in concentrations ranging from ND to 26100 ng/g ww (exceptional value in *Penaeus monodon* from India) for HCHs, OCP; from ND to 226.45 ng/g ww (found in *Neocaridina denticulate* from Qingyuan city, China) for BDE209, BFR; from ND to 12.1 ng/g ww (found in Asia) for HHCB-Lactone, SM; and from ND to 50650 ng/g ww (exceptional value found in *Penaeus notialis* in Lagos, Nigeria) for naphthalene, PAH. The pollutants deposited in the environment can enter the food chain and ultimately affect the human population. Although contamination levels were generally relatively low, the risk for accumulation of pollutants could pose risks to human health through the consumption of seafood such as shrimp. Public concern about the health effects of foodborne diseases related to pollutants, highlight the need of carrying out studies in this field. These pollutants have been associated with endocrine disruptive effects, cancer growth, neurobehavioral and reproductive disorders and diabetes. This review highlights some significant concerns regarding human health associated with hazard risks. Future studies on shrimp are needed in order to detect levels that could represent a risk for human health and to fully understand the contamination of biota such as, shrimps.

Declarations

Author contribution statement

All authors listed have significantly contributed to the development and the writing of this article.

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Competing interest statement

The authors declare no conflict of interest.

Additional information

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