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Review

Detection of Arsenic, Chromium, Cadmium, Lead, and Mercury in Fish: Effects on the Sustainable and Healthy Development of Aquatic Life and Human Consumers

Athanasia K. Tolkou [1,*], Dimitra K. Toubanaki [2] and George Z. Kyzas [1]

[1] Hephaestus Laboratory, Department of Chemistry, International Hellenic University, GR-65404 Kavala, Greece; kyzas@chem.ihu.gr
[2] Immunology of Infection Group, Department of Microbiology, Hellenic Pasteur Institute, GR-11521 Athens, Greece; dtouban@pasteur.gr
* Correspondence: tolkatha@chem.ihu.gr; Tel.: +30-2510-462-218

Abstract: Heavy metals are among the most important pollutants that threaten the aquatic environment when their concentrations exceed certain limits. Some of these metals and metalloids are beneficial and necessary for fish, but others, such as arsenic (As), chromium (Cr), cadmium (Cd), lead (Pb) and mercury (Hg), are non-essential and toxic. In reviewing the recent relevant literature, 4 different continents, 13 different countries, and more than 50 different fish species were analyzed in terms of As, Cr, Cd, Pb and Hg concentrations. According to the comparative results, it was found that in Tercan Dam Lake, Turkey, the highest concentration of Cr was detected in *Capoeta umbla* (2.455 mg/kg), and of As in *Ctenopharyngodon idella* (0.774 mg/kg) species. Greater values than the permissible limits of FAO/WHO in terms of As were also found in Andalusia, Southern Spain, in *Mullus surmuletus* (0.427 mg/kg), and *Sardina pilchardus* (0.561 mg/kg) and in *Sprattus sprattus* (0.636 mg/kg) in the Baltic Sea, but a remarkably high content of As (8.48 mg/kg) was determined in *Penaeus notialis*, found in Guinea, Africa. Moreover, Cd concentration was low to nil in almost all cases, with the exception of *Amblyceps mangois* species collected from the Dhaleshwari River in Bangladesh, which showed the highest value (0.063 mg/kg). Finally, extremely high levels of Pb were found in *Plectropomus pessuliferus* (5.05 mg/kg) and *Epinephelus summana* (2.80 mg/kg) in Jeddah, Saudi Arabia. The Hg content in fish was under the permissible limit in almost all cases, with megrim and red mullet from the Andalusian Sea exhibiting a relatively higher content (0.091 and 0.067 mg/kg). In general, the sequence of accumulation of toxic elements in fish was As > Cr > Pb > Cd > Hg.

Keywords: aquatic environment; heavy metals; limits; pollution; toxicity; contamination source; biomarker

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1. Introduction

Heavy metals are one of the most important pollutants that threaten the aquatic environment and the living organisms in it when their concentrations exceed the permissible limits [1]. Although these metals are not always harmful or toxic to aquatic life, they can affect human health through the food chain [2]. The growing pollution of water resources by heavy metals poses a threat both to public water supplies and to consumers of fish and seafood in general [3]. Furthermore, heavy metals and metalloids have been shown to cause fish toxicity due to their non-biodegradability and long-term existence in the environment [4]. The study of the bioaccumulation of contaminants in fish is significant in order to determine the content of trace elements in different fish species, which are due both to their metabolic capacity and to their eating habits [5].

The early developmental stages of fish, namely embryos and larvae, are more sensitive to contaminants such as heavy metals than juvenile and adult fish, and are widely used

as model organisms for biomarkers that define the toxicity of such chemicals to water bodies [3].

Although some heavy metals such as zinc (Zn), iron (Fe), cobalt (Co), and copper (Cu) are necessary for fish at very low levels (for their enzyme activity and other biological processes), they can become toxic when they exceed certain limits. On the contrary, other metals such as lead (Pb), cadmium (Cd), chromium (Cr), mercury (Hg), and arsenic (As) do not have an essential role in living organisms and are toxic even in very low concentrations, which is why they are dangerous both for fish and for humans (through the food chain) [2].

In this short review, five of these toxic heavy metals and metalloids, As, Cr, Cd, Pb and Hg, were selected to further analyze their effect on various fish species and therefore on humans through a review of the recent relevant literature. Fish bodies mainly consist of muscle tissues, which is their most-consumed part [6]; for this reason, in this review, the values of heavy metals and metalloids contained and reported were found mostly in muscle tissues (Figure 1). The particularity of this short review is that it includes different case studies from a large proportion of the planet, summarizing the main values of heavy metals found, and comparing the quality of water environments.

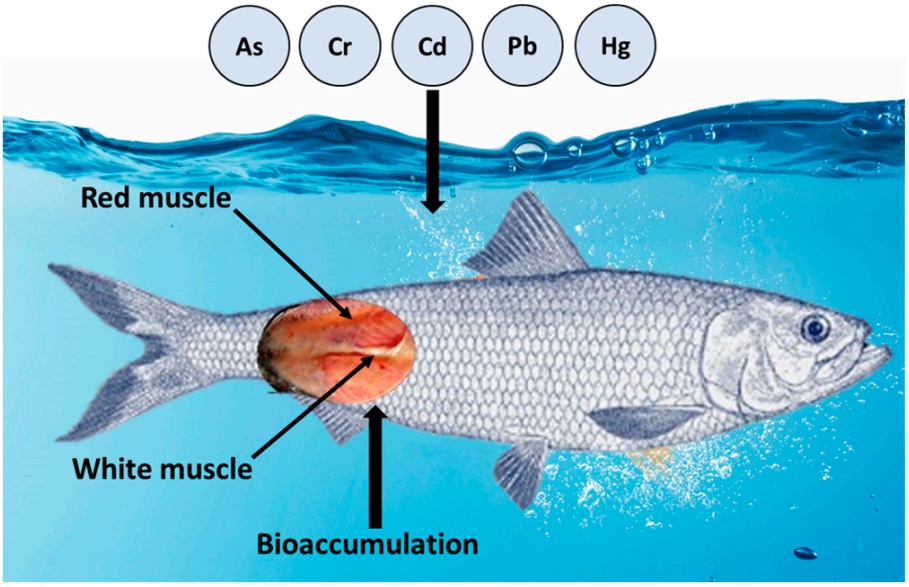


Figure 1. Bioaccumulation of heavy metal and metalloid toxicity in muscle tissues of fish (source: authors).

The heavy metals and metalloids evaluated in this commentary are elaborated upon below:

- Arsenic (As): As is released into the aquatic environment from various anthropogenic and agricultural sources, and has two oxidation stages; As(III) (arsenites) is more toxic than As(V) (arsenates) [7]. Arsenic from contaminated water and food is rapidly absorbed into fish tissues through the gills and skin, and can contaminate them. Exposure of freshwater fish to arsenic results in its bioaccumulation, mainly in liver and kidney tissues, showing a histopathological modification in the gills and liver tissues of these fish [4]. In addition, the presence of As affects fish reproduction by inhibiting spermatogenesis and oogenesis, thus reducing the fertilization rate [3].
- Chromium (Cr): Cr appears in water as Cr(III) and Cr(VI,), with the latter being more toxic and more soluble in water [8]. The concentration of Cr in water bodies, such as in rivers and lakes, usually ranges from 1 to 10 mg/L, but in seawater, the values are much lower, i.e., <0.1, and in some cases can reach 5 mg/L. Cr(VI) prevails in well-oxygenated marine waters, while Cr(III) appears mainly in coastal areas [9]. In addition, Cr(III) is an essential metal for humans and animals, and plays an important role in improving glucose tolerance [10], but Cr(VI) causes acute and chronic effects in humans [11].

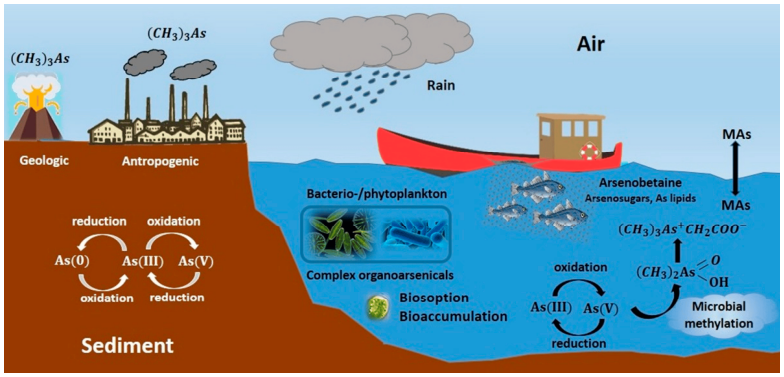
- Cadmium (Cd): Cd is a non-essential but very toxic metal with a chronic effect on human health due to its accumulation mainly in the liver, but also in the blood, bones, muscles, and kidneys [12]. Numerous studies carried out in several countries have shown that most foods have a Cd content between 0.005–0.1 mg/kg, but some foods like seafood may contain higher concentrations. Industrial waste, some fertilizers, etc., are responsible for about 50% of the Cd that reaches the sea. Mollusks tend to accumulate Cd in a higher amount than other organisms, as it is necessary for the responsiveness of metallothionein and hemocyanin genes [13].
- Lead (Pb): Pb is one of the most highly toxic metals that appears and threatens aquatic environments [14]. Fish are the most sensitive to the toxic effects of Pb exposure, which affects physiological and biochemical functions and is mainly caused by bioaccumulation [15]. Chronic exposure to Pb causes high accumulation of it, mainly in liver and kidney tissues [14]. The relative FAO and WHO limits for Pb are 2 and 0.5 mg/kg, respectively [16].
- Mercury (Hg): Hg belongs to the class of heavy metals that are harmful to all organisms even in extremely low concentrations, and can be found in both freshwater and marine ecosystems [17]. According to the FAO/WHO, the permissible concentration for Hg is 0.5 mg/kg for humans [16]. Hg is present as Hg^0, Hg^+, Hg^{2+}, and harmful organic mercury, especially MeHg (methyl) [18]. Moreover, the levels of Hg in fish brain are generally higher after MeHg exposure than after the Hg^{2+} exposure. Thus, MeHg in fish is the main source of mercury contamination, and there is high potential risk for consumers [19].

In Table 1 are listed the foremost sources of heavy metals and metalloids in aquatic environments. Industrial and geogenic sources as well as anthropogenic activities are the main sources of the contamination of water with these elements.

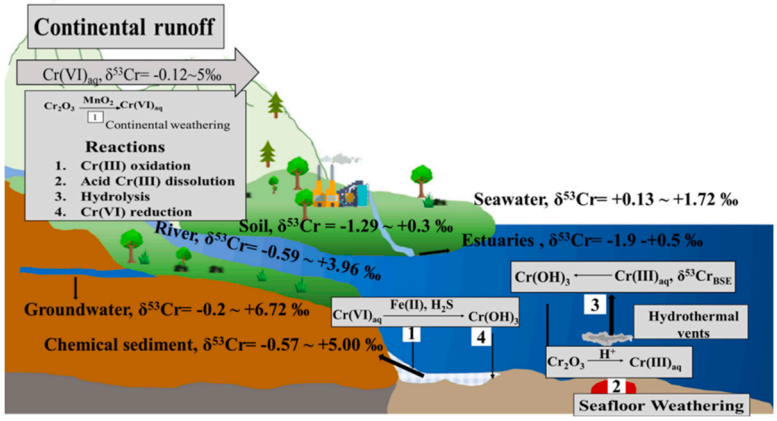
Table 1. Sources of As, Cr, Cd, Pb, and Hg contamination in the aquatic environment [20].

Heavy Metal (Loid)	Sources
As	Wood preservatives, mining and smelting, coal power plants, herbicides, volcanoes, petroleum refining, animal feed additives
Cr	Electroplating industry, sludge, solid waste, tanneries
Cd	Geogenic sources, metal smelting and refining, fossil fuel burning, sewage sludge
Pb	Mining and smelting of metalliferous ores, burning of leaded gasoline, municipal sewage, industrial waste enriched with Pb, paints
Hg	Volcano eruptions, forest fire, emissions from industries producing caustic soda, coal, peat, and wood burning

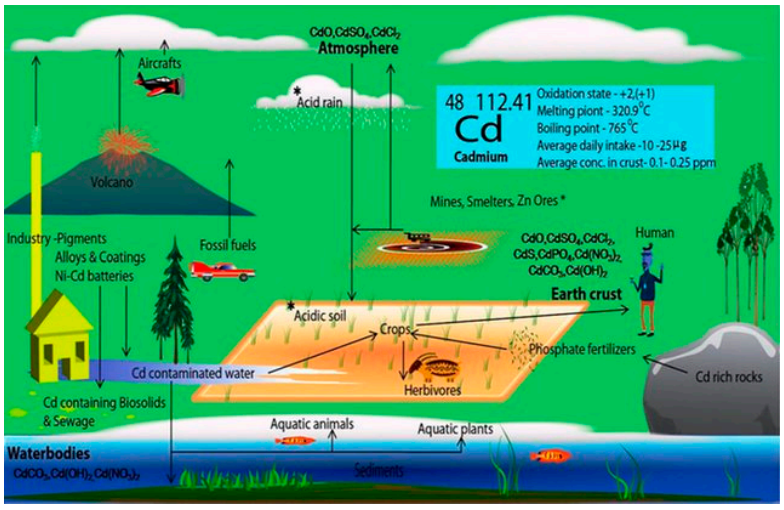
In aquatic environments, element concentrations within a biogeochemical area may differ between the water and land due to the nature of the environment. Therefore, the effect of the biogeochemical properties of the toxic elements studied in this review on the marine environment is crucial. Thus, the biogeochemical cycles of As, Cr, Cd, Pb and Hg are influenced by natural and anthropogenic processes that transform these elements through multiple chemical forms and environments, and are presented in Figure 2. As illustrated, these compounds originate in the environment, mainly from anthropogenic sources, in plants, soil and in water, and a chemical equilibrium between their speciation forms can exist.



(**a**)

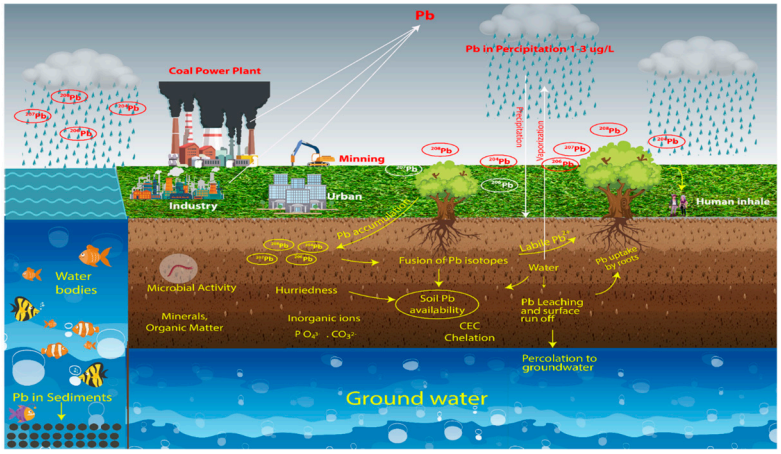


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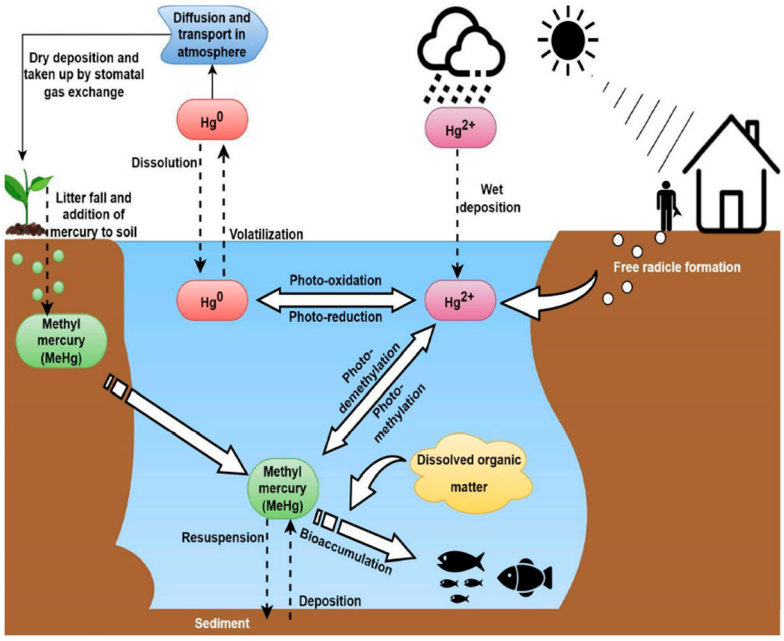


(**c**)

Figure 2. *Cont.*



(**d**)



(**e**)

Figure 2. Biogeochemical processes of As, Cr, Cd, Pb and Hg. Reproduced with permission. (**a**) As [21], (**b**) Cr [22], (**c**) Cd [23], (**d**) Pb [22], and (**e**) Hg [24].

Furthermore, as shown in Figure 3, the release of heavy metals in aquatic systems from natural or anthropogenic sources can cause the initial pollution of these systems by affecting the sustained and healthy development of aquatic life. Then, marine bodies, and specifically the fishes that are presented in this manuscript, can uptake these heavy metals, leading to their bioaccumulation. The bioaccumulation of heavy metals may pose great hazards to the health of humans and animals that depend on water bodies [1]. Fish not only constitute an important trophic level in aquatic ecosystems, but also serve as an important source of protein, vitamins, minerals, and polyunsaturated fatty acids for human beings [25]. Thus, seafood safety is a critical requirement for sustainable global quantitative and qualitative development. However, fish contaminated with heavy metals are capable of disrupting this balance in the food chain and causing many serious problems for humans through their consumption.

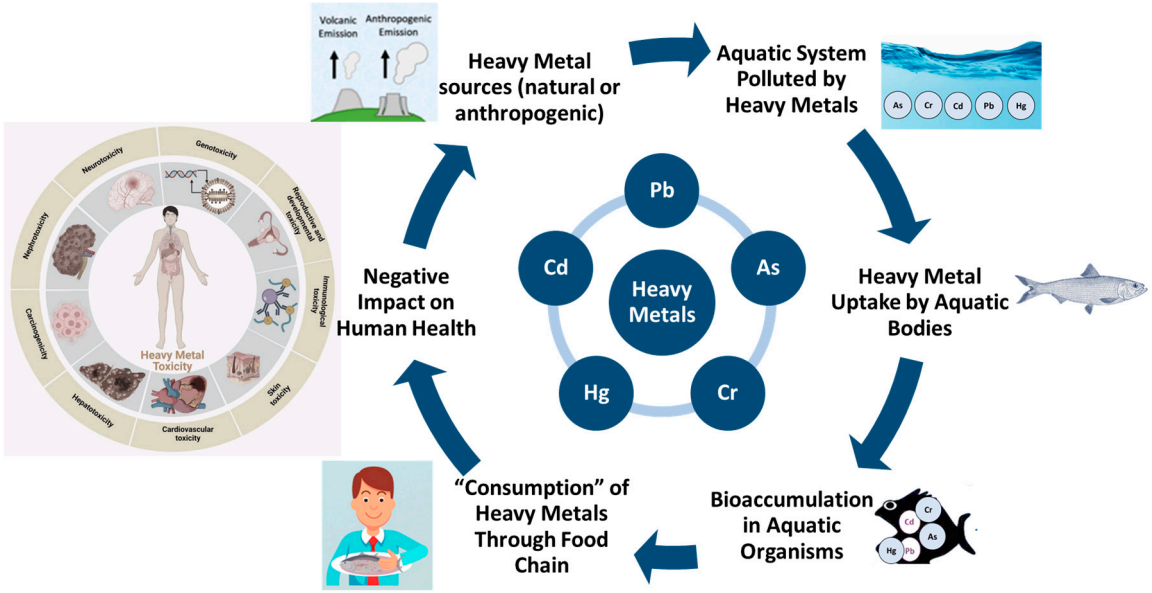


Figure 3. Life cycle of heavy metal pollution (source: authors).

To be more specific, in Figure 4 are illustrated the serious hazardous impacts that heavy metals can have when they enter the human body, causing biological and physiological problems because they cannot be decomposed and are non-biodegradable.

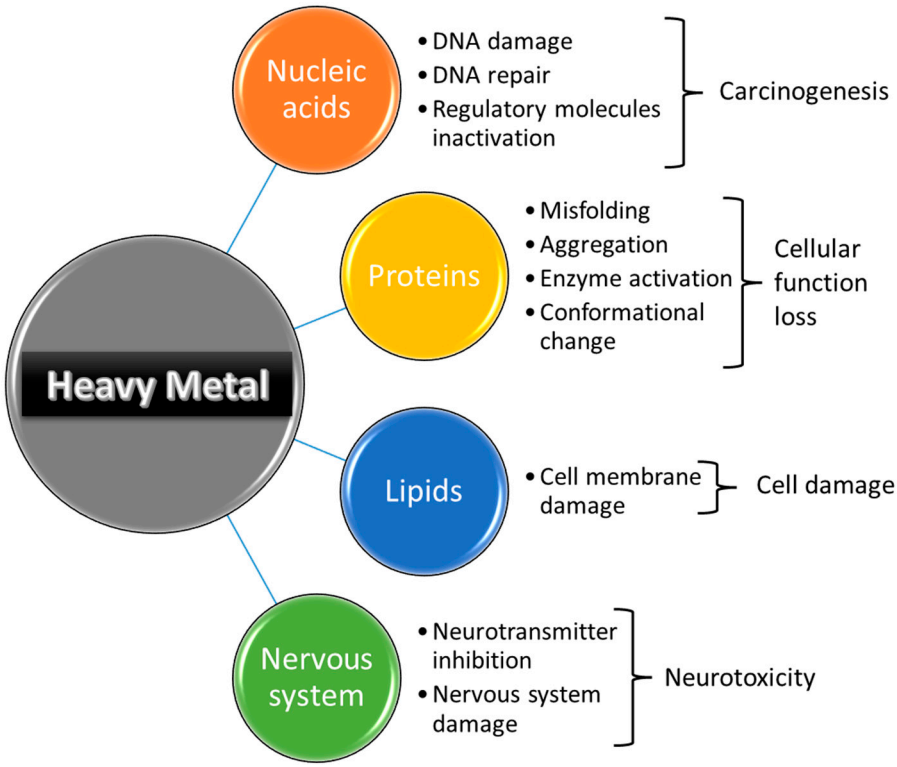


Figure 4. Effects of heavy metals on human health (source: authors).

2. Analytical Methods for the Determination of Heavy Metals and Metalloids

Several methods with digestion protocols were assessed for fish samples' preparation for heavy metal and metalloid analysis. The methods mentioned in the literature, which were optimized to achieve the best results, are as follows:

- Digestion of fish tissue with a combination of hydrogen peroxide/hydrochloric acid, coupled with solid-phase purification of the digested solid [26].
- Microwave-assisted digestion and extraction, which showed good recovery and extraction efficiency [27].
- Addition of hydrogen peroxide (30%) and subsequent digestion using a microwave digestion unit [28].

Milli-Q water was used for the dilution of the digest for the following analysis.

In general, various advanced analytical techniques can be used to detect heavy metals and metalloids in environmental and biological samples. These include inductively coupled plasma coupled with optical emission spectrometry or mass spectrometry, X-ray absorption spectroscopy, and molecular spectroscopy [29]. However, it is worth noting that the appropriateness of the applied technique also depends on the specific sample and the research question, and the disadvantages that may exist must also be taken into account. For instance, expensive equipment and operational costs, complex sample pretreatment, and long detection time, which make in situ measurement difficult, are among the drawbacks [30]. Compared with the above heavy metal(loid) detection methods, electrochemical techniques are a promising point-of-care testing method due to their advantages of low cost, simple equipment, fast speed analysis, and dependability, and can offer transferability and fast responses for on-site analysis [31].

In most of the studies referenced in this short review, Cr, As and Cd in fish were analyzed using inductively coupled plasma–mass spectrometry (ICP-MS), high-performance liquid chromatography–inductively coupled plasma–dynamic reaction cell–mass spectrometry (hPlC/ICP-DRC-MS), inductively coupled plasma–optical emission spectrometry (ICP-OES) and atomic absorption spectrophotometry (AAS) equipped with a graphite furnace (GFAAS), or with direct flow injection analysis through a hydride generation system (FIAS) [28,32–34]. Pb content was measured using electrothermal atomic absorption spectrometry (ETAAS) [35] or atomic absorption spectrophotometry (AAS) [36], and Hg concentration was determined using a single-purpose atomic absorption spectrometer AMA-254 [35].

3. Biomarkers of Heavy Metal and Metalloid Toxicity in Fishes

Biomarkers are measurable indicators of a biological condition. There are numerous biomarkers of heavy metal poisoning in freshwater fishes at several research levels [37].

Among the most important biomarkers, stress proteins such as metallothionein have shown delayed expression when exposed to sublethal chromium concentrations. Moreover, heat shock protein 70 (HSP-70) has been reported to be overstimulated in different fish species after exposure to excessive amounts of chromium [38]. In addition, integrated biomarker response index (IBRv2) has been applied to shorten biomarker response interpretation [39], as IBRv2 could differentiate fish organs according to biomarker responses. According to this index, a higher IBRv2 value indicates that an organ is affected, possibly due to some association with the level of accumulation of heavy metals and metalloids. Oxidative stress biomarkers reflecting the presence of heavy metals and metalloids have also been reported as indicators of environmental pollution due to the modification of antioxidant enzymes, the glutathione system, and the induction of lipid peroxidation in fish organs [40].

Furthermore, the handling of fish as a heavy metal assessment tool (according to behavioral responses, hepatocyte variation, and enzymatic reactions) has proved to be very valuable in environmental pollution monitoring [41]. The monitoring of hematological and biochemical biomarkers can provide information on fish health [42]. Some of the biomarkers associated with ecotoxicology in the aquatic environment apart from the most common heat shock protein (HSP 70), metallothionein, and oxidative stress are triglyceride level, vitellogenin (yolk proteins), cytochrome P4501A (CYP1A), circulating hormone levels, 17 beta estradiol hormone, etc. [37].

4. Discussion

There are many case studies in the literature examining existing concentrations of heavy metals and metalloids, such as As, Cr, Cd, Pb and Hg, and these are analyzed in this review. Tables 2–5 summarizes these cases by listing the water body in which they were detected, as well as the fish species tested and the maximum concentrations of these metals found. In addition, in Figure 5 are illustrated some of the sampling regions of fish that are analyzed in this review.

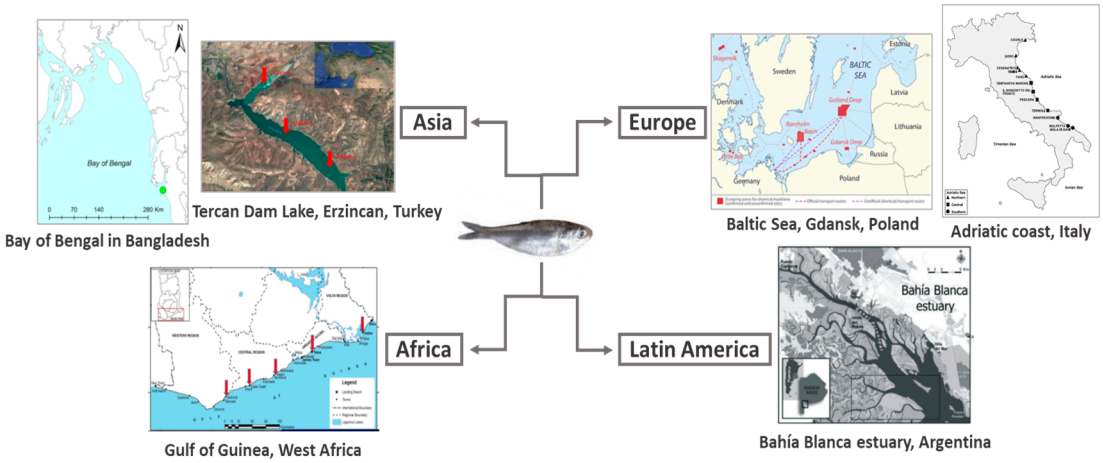


Figure 5. Sampling sites of fish in several continents (source: authors).

4.1. Case Studies in Asia

In brief, in the case of Dhaleshwari River in Tangail, Bangladesh [43], six fish species were examined, and the relative concentrations of Cd, Cr, As, Pb and Hg were found to vary between 0.002–0.019, 0.046–0.159, 0.016–0.128, 0.091–0.234 and 0.004–0.012 mg/kg, respectively. The sequence of accumulation of heavy metals and metalloids in fish was Pb > As > Cr > Cd > Hg, but in almost all species, the concentrations were found to be below permissible limits [43], proving that there is practically no heavy metal pollution in this river, and the fish can be consumed. Some exceptions are the concentrations of Cd in *Amblyceps mangois*, of Cr in *Mastacembelus armatus*, and of As, Pb, and Hg in *Metapenaeus tenuipes (syn. Spinulatus)*, which were found to be higher than the limits, possibly because of the agricultural use of various insecticides and pesticides in the area. In *Puntius puntio*, the As content was found to be 0.061 mg/kg; in *Amblyceps mangois*, the relative Cd concentration was 0.063 mg/kg; and in *Mastacembelus armatus*, the Cr content was found to be 0.159 mg/kg, as these were the most contaminated species found [43]. Furthermore, in another river of Bangladesh, Buriganga River in Dhaka, which is the most polluted river in the country, fish samples such as *Heteropneustes fossilis* were collected to evaluate the presence of Cd, Pb and Cr. According to the results of this study [44], higher values than those permitted by the FAO/WHO of Cd (0.040 mg/kg), Pb (1.040 mg/kg), and Cr (0.33 mg/kg) were observed, making this river hazardous to human health. Determination of As, Cr, Cd, and Pb contents in fish from Shitalakshya River, located also in Dhaka, Bangladesh, and the potential threat to human health as a result of their consumption were also studied. *Systomus sarana* (Olive barb), *Pethia ticto* (Ticto barb), and *Mastacembelus armatus* (Tire-track Spiny eel) were the three species that were used for examination. In the study of Hasan et al. [45], in order to quantify the impact of public health risk, the target hazard quotient (THQ) and carcinogenic risk (TCR) were calculated. According to their results, As, Cr, Cd, and Pb concentrations were over the considered allowable limits, and the found TCR values demonstrated an increased risk of cancer from eating the fish selected for study; for this reason, continuous monitoring of these polluted rivers is suggested. Moreover, in the study of Resma et al. [33], three different fish species highly consumed by the people of the southern region of Bangladesh were evaluated for their content of toxic metals As,

Cr, and Cd. According to these results, the order in which the highest concentrations appear is Cr > As > Cd. Maximum concentrations of Cr were recorded in *Labeo rohita* (0.623 mg/kg), and *Pangasius pangasius* contained the slightly higher concentrations of As and Cd (0.045 and 0.006 mg/kg, respectively). In the muscle tissues of *Coilia dussumieri* and *Sardinella fimbriata*, in the Bay of Bengal in Bangladesh [46], concentrations of Cr, As, and Cd were also determined. The relative values were 0.63, 1.33 and 0.13 mg/kg for *Coilia dussumieri*, and 1.57, 3.93, 0.06 mg/kg, respectively, for *Sardinella fimbriata;* the As content in *Sardinella fimbriata* was found to be very high.

Furthermore, three categories of fish from the Tibetan Autonomous Region of China were also examined [28]: i.e., big-headed carp, grass carp, and tilapia. As concluded, arsenic concentration in the tilapia (0.286 mg/kg) was higher than that in big-headed and grass carp (~0.034 mg/kg), and exceeded more than twice the maximum established limit. Another case study took place in Tercan Dam Lake, Erzincan, Turkey [5], and aimed to determine As, Cr, and Pb contents in some fish species. The highest concentrations of As and Cr were detected in the gill tissues and the liver of the fish, and were significantly higher than acceptable levels. In Table 2, the mean values of mg/kg determined in the liver of the fish are reported; apparently, the *Capoeta umbla* species far exceeded the limits of Cr and Pb, with found values of 2.455 and 2.074 mg/kg, respectively, followed by *Cyprinus carpio* (0.918 and 0.534 mg/kg, respectively). In the case of As, *Ctenopharyngodon idella* (0.774 mg/kg) and *Cyprinus carpio* (0.383 mg/kg) exhibited higher values. Thus, *Cyprinus carpio* is considered to be the most contaminated species, as it contains concentrations of the metals that exceed permissible limits [47].

Moreover, in China, and particularly in Hangzhou, a recent study [48] assessed the bioaccumulation of metals in commercially important fishes (six different species), and calculated the potential hazard to human health resulting from their consumption. Each species has a precise bioaccumulative susceptibility to various metals. Thus, the decreasing order of metal concentration in organisms was as As > Cr > Cd > Pb > Hg. Apart from the Cd (with *Harpadon nehereus*, at 0.070 mg/kg, having the highest concentration) and As (*Cynoglossus joyneri*, at 0.350 mg/kg, having the highest concentration) levels found in all tested fish, the metal concentrations did not exceed national and international guideline values. The carcinogenic risk (CR) index values indicated that children were vulnerable to carcinogenic risk from As and Cd consumed through fish. Therefore, thorough examination of all aquatic organisms consumed by humans is highly recommended, including metal concentrations in various organs [48].

In the Coastal Area (Tam Hung and Minh Duc) of Hai Phong City in Northern Vietnam, two species, snake-headed fish (*Channa argus*) and catfish (*Pangasianodon hypophthalmus)*, were selected in order to evaluate the concentrations of four heavy metals and metalloids (As, Cr, Cd and Pb) [49]. As found in this study, the concentration of Cr in catfish (2.250 mg/kg) exceeded the levels suggested by the FAO/WHO. In addition, the concentration of Cd (2.300 mg/kg) was higher in snake-headed fish, and in particular, the order was Cd > Cr > As > Pb. The level of As was similar in both fish. These results indicate that consumption of these fish may endanger human health.

Moreover, a study contacted in Jeddah, Saudi Arabia, which examined five fish samples [50], showed that the values of most identified metals were within permissible limits, as shown in Table 2. An exception is the high As values found in *Lethrinus nebulosus* (0.23–0.26 mg/kg), *Plectropomus pessuliferus* (0.21–0.26 mg/kg), and *Epinephelus summana* (0.21–0.23 mg/kg). On the other hand, Cd was not found in these species. In addition, higher Pb levels were found in *Plectropomus pessuliferus* (5.050 mg/kg) and *Epinephelus summana* (2.800 mg/kg) compared to the permissible standard limits. Therefore, in this study, it is proposed that the residual concentrations of heavy metals and metalloids should be detected and recorded before these fish are available for consumption, in order to ensure public health.

Pakistan is a country facing many environmental issues, mainly due to increasing pollution from human activities. One of the most critical environmental issues in Pakistan is heavy metal pollution, due to which the Karachi Coast in Pakistan is a degraded aquatic

environment [20]. The effect of heavy metals on two fish on the Karachi Coast, focusing on those discussed in this brief review, were as follows: Cd > Cr > Pb > Hg > As for *Thunnus spp* and Cr > Pb > Cd > Hg > As for *Rastrelliger kanagurta*. In this coastal area, fish are used as a biomarker for assessing the health of aquatic ecosystems, particularly around industrial, agricultural, municipal, and domestic contaminated sites [20].

In addition, in the East Kolkata Wetlands, India, the presence of toxic metals such as Cr, Pb, Cd and Hg was investigated in the muscle tissues of fishes [51]. Three different species were examined, i.e., Rohu (*Labeo rohita*), Tilapia (*Oreochromis niloticus*), and Catla (*Catla catla*), and according to the results, the bioaccumulation of toxic heavy metals in fish follows the order of Tilapia > Rohu > Catla; the pattern of bioaccumulation of toxic metals shows the trend Pb > Cd > Cr > Hg in all seasons and years.

Table 2. Bioaccumulation of As, Cr, Cd, Pb, and Hg in fish species in Asia, according to the literature.

	Location	Fish Species		As * (mg/kg)	Cr * (mg/kg)	Cd * (mg/kg)	Pb * (mg/kg)	Hg * (mg/kg)	Ref.
		Scientific Name	Common Name						
Asia	Dhaleshwari River, Tangail, Bangladesh	*Channa punctata*	Spotted snakehead	0.016	0.046	0.019	0.133	0.005	[43]
		Mastacembelus armatus	Zig-zag eel	0.027	0.159	0.010	0.091	0.011	
		Mystus vittatus	striped dwarf catfish	0.033	0.116	0.012	0.234	0.005	
		Puntius puntio	Puntio barb	0.061	0.124	0.002	0.174	0.006	
		Amblyceps mangois	Indian torrent catfish	0.035	0.103	0.063	0.183	0.004	
		Metapenaeus tenuipes (syn. Spinulatus)	Shrimp	0.128	0.112	0.005	0.231	0.012	
	Southern region, Bangladesh	*Oreochromis niloticus*	Nile tilapia	0.042	0.590 ***	0.004	nd **	nd **	[33]
		Pangasius pangasius	Pangas catfish	0.045	0.577 ***	0.006	nd **	nd **	
		Labeo rohita	Rohu	0.035	0.623 ***	0.004	nd **	nd **	
	Buriganga River, Dhaka, Bangladesh	*Heteropneustes fossilis*	Asian stinging catfish	nd **	0.330	0.040	1.040	nd **	[44]
	Shitalakshya River, Dhaka, Bangladesh	*Systomus sarana*	Olive barb	0.001	0.004	0.001	0.001	nd **	[45]
		Pethia ticto	Ticto barb	0.001	0.003	0.001	0.003	nd **	
		Mastacembelus armatus	Zig-zag eel	0.004	0.001	0.001	0.005	nd **	
	Bay of Bengal, Bangladesh	*Coilia dussumieri*	Gold-spotted grenadier anchovy	1.330	0.630	0.130	0.230	nd **	[46]
		Sardinella fimbriata	Fringescale sardinella	3.930	1.570	0.060	0.930	nd **	
	Coastal Area (Tam Hung and Minh Duc) Hai Phong, N. Vietnam	*Channa argus*	Northern snakehead	1.180	2.120	2.300	0.080	nd **	[49]
		Pangasianodon hypophthalmus	Iridescent shark catfish	1.660	2.250	1.060	0.100	nd **	
	Jeddah, Saudi Arabia	*Lethrinus nebulosus*	Spangled Emperor	0.245	nd **	0.000	1.510	nd **	[50]
		Scomberomorus commerson	Narrow-barred Spanish mackerel	0.170	nd **	0.000	0.890	nd **	
		Plectropomus pessuliferus	Roving coral grouper	0.235	nd **	0.000	5.050	nd **	
		Pampus argenteus	Silver pomfret	0.165	nd **	0.000	1.690	nd **	
		Epinephelus summana	Summan grouper	0.220	nd **	0.000	2.800	nd **	
	Lhasa, Tibetan Autonomous Region, China	*(Mean values of five species)*	Tilapia	0.286	0.003	0.004	0.021	nd **	[28]
		(Mean values of six species)	Big-head carp	0.034	0.018	0.000	0.004	nd **	
		(Mean values of five species)	Grass carp	0.033	0.005	0.003	0.024	nd **	
	Hangzhou Bay, China	*Coilia nasus*	Japanese grenadier anchovy	0.220	0.090	0.020	0.060	0.020	[48]
		Collichthys lucidus	Bighead croaker	0.210	0.070	0.060	0.060	0.010	
		Cynoglossus joyneri	Red tongue sole	0.350	0.050	0.010	0.060	0.003	
		Harpadon nehereus	Bombay duck	0.150	0.070	0.060	0.070	0.010	
		Lophiogobius ocellicauda	Lophiogobius	0.200	0.060	0.040	0.050	0.010	
		Miichthys miiuy	Brown croaker	0.190	0.090	0.040	0.030	0.010	

Table 2. *Cont.*

Location		Fish Species		As * (mg/kg)	Cr * (mg/kg)	Cd * (mg/kg)	Pb * (mg/kg)	Hg * (mg/kg)	Ref.
		Scientific Name	Common Name						
Asia	Tercan Dam Lake, Erzincan, Turkey	*Luciobarbus capito*	Bulatmai barbel	0.124	0.398	nd **	0.090	nd **	[5]
		Capoeta umbla	Tigris scraper	0.186	2.455	nd **	2.074	nd **	
		Ctenopharyngodon idella	Grass carp	0.774	0.049	nd **	0.455	nd **	
		Cyprinus carpio	Common carp	0.383	0.918	nd **	0.534	nd **	
	Karachi Coast, Pakistan	*Thunnus*	Tuna	nd **	0.350	0.710	0.270	nd **	[20]
		Rastrelliger kanagurta	Indian mackerel	nd **	0.370	0.310	0.320	nd **	

* WHO/FAO standard values: As 0.1; Cr 0.15; Cd 0.05; Pb 0.5/2.0; Hg 0.5 mg/kg [38,47]. ** nd = not determined. *** Cr 1.0 mg/kg in drinking water [33,52].

4.2. Case Studies in Europe

In Europe, across the coast of the Adriatic Sea [9], six fish species were collected and examined for Cd, Cr, and Pb content. The highest values were observed for *Engraulis encrasicolus* (Cd: 0.020, Cr: 0.083 and Pb: 0.046 mg/kg), for *Mullus barbatus* (Cd: 0.003, Cr: 0.031 and Pb: 0.036 mg/kg), and for *Scomber scombrus* (Cd: 0.008, Cr: 0.028 and Pb: 0.012 mg/kg), but the determined values were lower than FAO/WHO limits. Moreover, in Slovakia, Cd, Pb, and Hg contents were analyzed in a sample of common carp fish (*Cyprinus carpio*) from Žitava River, Nitra, Slovakia, where only 0.06 mg/kg Cd, 0.14 mg/kg Pb and 0.02 mg/kg Hg were found [6]. Further north, specifically in the Baltic Sea (area of Gdansk) [53], the As concentration in the muscle tissues of *Gadus morhua callarias L* (0.390 mg/kg), *Sprattus sprattus* (0.636 mg/kg), *Clupea harengus membras* (0.460 mg/kg), and *Platichthys flesus* (0.588 mg/kg) was determined. European sprat muscle (*Sprattus sprattus*) had the highest content of total As.

In addition, fresh fish species in Andalusia, Southern Spain, were examined for their toxic As, Cd, Pb, and Hg contents [54]. As presented in Table 3, the Cd concentration was low to nil, in contrast to the concentration of As, which was relatively higher. Particularly, sardines, with 0.561 mg/kg, and red mullet, with 0.427 mg/kg, showed the highest contents of As. Cuttlefish provided the highest Pb concentration (0.117 mg/kg), and megrim and red mullet the relative highest Hg content (0.091 and 0.067 mg/kg).

Table 3. Bioaccumulation of As, Cr, Cd, Pb, and Hg in fish species in Europe, according to the literature.

Location		Fish Species		As * (mg/kg)	Cr * (mg/kg)	Cd * (mg/kg)	Pb * (mg/kg)	Hg * (mg/kg)	Ref.
		Scientific Name	Common Name						
Europe	Andalusia, Southern Spain	*Solea solea*	Common sole	0.233	nd **	0.001	0.052	0.019	[54]
		Sepia officinalis	Common cuttlefish	0.172	nd **	0.000	0.117	0.032	
		Scomber scombrus	Atlantic mackerel	0.190	nd **	0.001	0.004	0.022	
		Lepidorhombus boscii	Four-spotted megrim	0.066	nd **	0.000	0.004	0.091	
		Mullus surmuletus	Striped red mullet	0.427	nd **	0.000	0.004	0.067	
		Sardina pilchardus	European pilchard	0.561	nd **	0.002	0.004	0.034	
	Adriatic Sea coast, Italy	*Engraulis encrasicolus*	European anchovy	nd **	0.083	0.020	0.046	nd **	[9]
		Lophius piscatorius	European angler	nd **	0.007	0.001	0.011	nd **	
		Merluccius merluccius	European hake,	nd **	0.010	0.006	0.005	nd **	
		Scomber scombrus	Atlantic mackerel	nd **	0.028	0.008	0.012	nd **	
		Mullus barbatus	Red mullet	nd **	0.031	0.003	0.036	nd **	
		Solea solea	Common sole	nd **	0.062	0.001	0.015	nd **	

Table 3. *Cont.*

Location		Fish Species		As * (mg/kg)	Cr * (mg/kg)	Cd * (mg/kg)	Pb * (mg/kg)	Hg * (mg/kg)	Ref.
		Scientific Name	Common Name						
Europe	Žitava River, Nitra, Slovakia	*Cyprinus carpio*	Common carp	nd **	nd **	0.060	0.140	0.020	[6]
	Baltic Sea, Gdansk, Poland	*Gadus morhua callarias L*	Baltic cod	0.390	nd **	nd **	nd **	nd **	[55]
		Sprattus sprattus	European sprat	0.636	nd **	nd **	nd **	nd **	
		Clupea harengus membras	Baltic herring	0.460	nd **	nd **	nd **	nd **	
		Platichthys flesus	European flounder	0.588	nd **	nd **	nd **	nd **	

* WHO/FAO standard values: As 0.1; Cr 0.15; Cd 0.05; Pb 0.5/2.0; Hg 0.5 mg/kg [38,47]. ** nd = not determined.

4.3. Case Studies in Africa

In Africa (Table 4) and in particular the Gulf of Guinea, West Africa, recently, Nyarko et al. [56] detected (among others) the levels of Cd As, Pb and Hg in the muscles of four different fish: *Sardinella maderensis*, *Dentex angolensis*, *Sphyraena sphyraena*, and *Penaeus notialis*. Higher concentrations of both metals (As 8.48 and Cd 0.03 mg/kg) were found in *Penaeus notialis*, except Pb, which was below the detection limit in all species examined. Regarding Hg concentration, this was higher in *Dentex angolensis* (0.137 mg/kg) and *Sphyraena sphyraena* (0.063 mg/kg). The remarkably high content of As determined in this fish increases the risk of future health problems, and therefore, it should be consumed with caution.

Table 4. Bioaccumulation of As, Cr, Cd, Pb and Hg in fish species in Africa, according to the literature.

Location		Fish Species		As * (mg/kg)	Cr * (mg/kg)	Cd * (mg/kg)	Pb * (mg/kg)	Hg * (mg/kg)	Ref.
		Scientific Name	Common Name						
Africa	Gulf of Guinea, West Africa	*Sardinella maderensis*	Madeiran Sardinella	1.560	nd **	0.005	nd **	0.021	[56]
		Dentex angolensis	Angola dentex	1.870	nd **	0.006	nd **	0.137	
		Sphyraena sphyraena	European barracuda	0.820	nd **	0.002	nd **	0.063	
		Penaeus notialis	Southern pink shrimp	8.480	nd **	0.027	nd **	0.026	

* WHO/FAO standard values: As 0.1; Cr 0.15; Cd 0.05; Pb 0.5/2.0; Hg 0.5 mg/kg [38,47]. ** nd = not determined.

4.4. Case Studies of Latin America

In Latin America, the Bahía Blanca estuary is a coastal environment in Argentina that has one of the largest petrochemical centers, resulting in large amounts of wastewater discharge. For this reason, a recent study [34] focused on the determination of Cr and Pb in six commercial fish species, as detailed in Table 5. According to the results of this study, the species *Cynoscion guatucupa* and *Odontesthes argentinensis* showed at least one sample containing high concentrations of Cr (0.450 and 0.250 mg/kg, respectively) above permissible levels for human consumption. In contrast, the Pb concentration in all species was below the method's detection limits.

4.5. Statistical Comparison

Considering the analysis made in Tables 2–5 and the concentrations of the various toxic substances studied in this manuscript, Figure 6 compares the percentage (%) of concentrations exceeding permissible limits, depending on continent. As can be observed, Asia presents a major problem in As, Cr, Cd and Pb, which in the case of As exceeds the permitted limits in more than 50% of studies. On the contrary, in the case of Europe and Africa, the main problem appears in increased As contents, in relation to toxic heavy metals, with the continent of Africa accounting for 100% of the detected contamination. On the

other hand, Latin America seems mainly to have excessive levels of Cr, which are beyond permissible values. Finally, in all continents and case studies, the levels of Hg were found not to exceed permissible limits, and this is the reason why its bar does not appear in Figure 6 (green bar).

Table 5. Bioaccumulation of As, Cr, Cd, Pb and Hg in fish species in Latin America, according to the literature.

Location		Fish Species		As * (mg/kg)	Cr * (mg/kg)	Cd * (mg/kg)	Pb * (mg/kg)	Hg * (mg/kg)	Ref.
		Scientific Name	Common Name						
Latin America	Bahía Blanca estuary, Argentina	*Cynoscion guatucupa*	Stripped weakfish	nd **	0.450	nd **	<mdl ***	nd **	[34]
		Micropogonias furnieri	Whitemouth croaker	nd **	0.095	nd **	<mdl ***	nd **	
		Mustelus schmitti	Narrow-nose smooth-hound	nd **	0.047	nd **	<mdl ***	nd **	
		Brevoortia aurea	Brazilian Menhaden	nd **	0.010	nd **	<mdl ***	nd **	
		Odontesthes argentinensis	-	nd **	0.250	nd **	<mdl ***	nd **	
		Paralichthys orbignyanus	Black flounder	nd **	0.030	nd **	<mdl ***	nd **	

* WHO/FAO standard values: As 0.1; Cr 0.15; Cd 0.05; Pb 0.5/2.0; Hg 0.5 mg/kg [38,47]. ** nd = not determined. *** <mdl = below the method detection limits.

Furthermore, in Figure 7, a comparison is made between the percentage (%) of cases found to exceed permissible limits for As depending on the water type of the study area. Specifically, an evaluation of excesses in fresh and marine water was carried out, and as it turned out, the fish species studied for the determination of As in Europe and Africa, and that exceed permissible levels, are seawater fish. In Asia, As appears slightly more increased in freshwater fish.

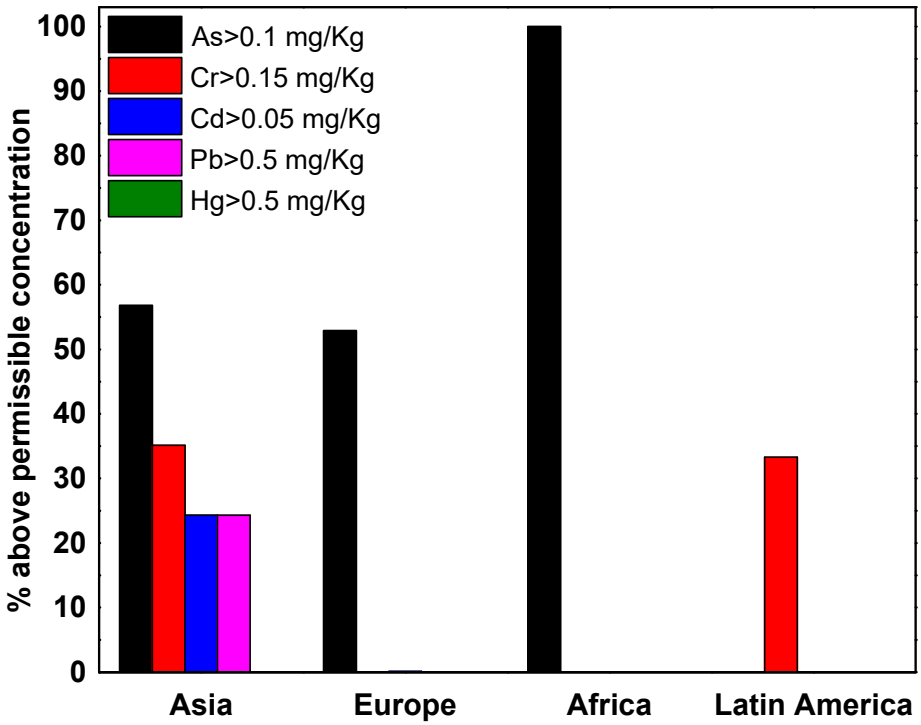


Figure 6. Percentage (%) of concentrations that exceed permissible limits of toxic heavy metals, depending on continent.

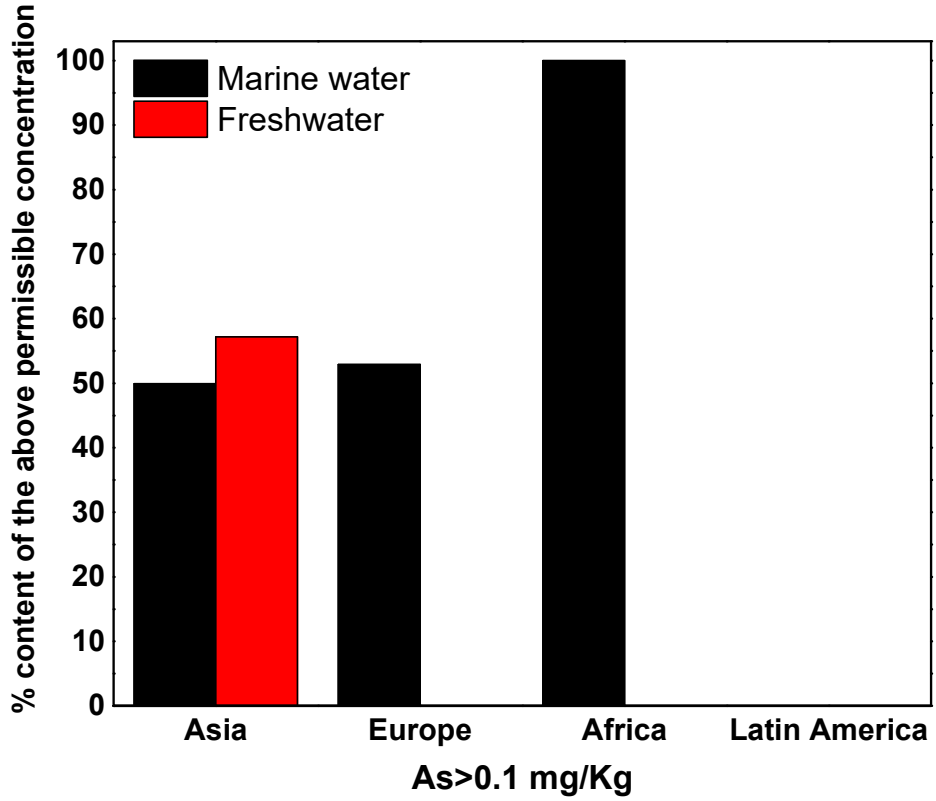


Figure 7. Percentage (%) of concentrations that exceed the permissible limits of As, based on the type of water (fresh water or marine water) across each continent.

5. Conclusions

The objective of this commentary was to review heavy metal and metalloid concentrations of elements such as arsenic (As), chromium (Cr), cadmium (Cd), lead (Pb), and mercury (Hg) in the tissues of freshwater and marine fish, according to recent relevant literature. Therefore, 4 different continents (Europe, Asia, Africa, and Latin America), 13 different countries (Spain, Italy, Slovakia, Poland, China, India, Pakistan, Turkey, Bangladesh, Vietnam, Saudi Arabia, Guinea and Argentina), and 50 different fish species were discussed in terms of Cd, As, Cr, Pb and Hg concentrations. As observed, in Tercan Dam Lake, Turkey, the highest concentration of Cr and Pb was detected in the *Capoeta umbla*-Tigris scraper (2.455 and 2.074 mg/kg, respectively) and of As in *Ctenopharyngodon idella*-Grass carp (0.774 mg/kg) species. The highest content of As was determined in Gulf of Guinea in Africa, providing 8.48 mg/kg for *Penaeus notialis*. Values above the permissible levels of FAO/WHO for human consumption were also found in Andalusia, Southern Spain, in the species *Mullus surmuletus* (striped red mullet) (0.427 mg/kg), *Sardina pilchardus* (European pilchard (0.561 mg/kg), *Sprattus sprattus* (European sprat) (0.636 mg/kg), and *Platichthys flesus* (European flounder) (0.588 mg/kg) in the Baltic Sea. For Cd concentration, *Amblyceps mangois* (Indian catfish), collected from the Dhaleshwari River in Bangladesh, showed the highest value of 0.063 mg/kg; a Pb concentration of 5.050 mg/kg was found in *Plectropomus pessuliferus*, and of 2.800 mg/kg in *Epinephelus summana* in Jeddah, Saudi Arabia. On the other hand, Hg content in fish was under the permissible limit in almost all cases. The order of accumulation of heavy metals and metalloids in fish was As > Cr > Pb > Cd > Hg. According to statistical comparisons, Asia presents the major problem regarding As, Cr, Cd and Pb; in the case of As, levels exceeded the permitted limits in more than 50% of the studies reviewed. In addition, the fish species with increased levels of As in Asia are freshwater fish, and in Europe and Africa, the fish that exceed permissible levels of As are seawater fish. In most of the considered studies, the analytical methods used for the determination of As, Cr, and Cd were ICP-MS, HPLC/ICP-DRC-MS,

ICP/OES, GFAAS, and FIAS, and several digestion protocols were considered for fish samples' preparation for heavy metals analysis.

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