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EVALUATION OF HEAVY METALS CONCENTRATION DYNAMICS IN FISH FROM THE BLACK SEA COASTAL AREA: AN OVERVIEW

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Abstract

Contamination of the marine environment induced by metals has risen in recent years due to the global population increase and it affects the aquatic ecosystem via food chain. The current paper aims to present information regarding the heavy metals accumulation in fish from the Black Sea West, South and South-East coastal areas. Therefore, various databases were revised and the relevant information was centralized, in order to obtain a clear view on the concentration dynamics and accumulation tendency of several heavy metals, as follows: Pb, Cd, Fe, Zn, Cu, Hg, Ni, Cr and As. As a conclusion to this research, we can state that the highest concentrations of heavy metals in fish meat was encountered in case of red mullet (*Mullus barbatus,* Linnaeus, 1758), followed by bluefish (*Pomatomus saltatrix,* Linnaeus, 1766). The upwarding scale of heavy metals concentration, reported in the analysed studies was found as follows: $Cr < Ni < Hg < Cd < Pb < As < Cu < Fe < Zn$. Both West part (Romanian and Bulgarian marine coastal areas) and Central South part of the Black Sea (Turkish marine coastal area, represented by the perimeter situated between the Kizilirmak and Yeşilirmak rivers) have proven to be the most polluted in terms of heavy metals accumulation in fish meat.

Key words: Black Sea, heavy metals, water pollution

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1. Introduction

Metals are natural elements and they are found everywhere in nature (air, soil, water), in low concentrations. The toxic concentrations of metals in the environment are always related to human activities (Burada, 2014a; Labianca et al., 2017; Robu et al., 2003; Zazouli et al., 2014). In order to be considered heavy metals, metals and metalloids must record atomic density values over 4g/cm³ (Duruibe et al., 2007). Also, if those elements have a five times higher density value, comparing to water density, they are also considered heavy metals (Duruibe et al., 2007).

According to Heath (1995) and Pal et al. (2018) the distribution of heavy metals in the aquatic environment is as follows: dissolved in water, precipitated in sediments, or combined complexes with other organic elements, making them easy to assimilate by fish. There are different pathways that make metals enter the aquatic environment. The main paths are through natural sources (geological erosion) and anthropic sources (industrial activities, transportation, tourism, agricultural activities etc.). Metals such as Hg, Cd, Pb and Se are transported into the aquatic environment mainly through atmospheric cycle (Depledge and Rainbow, 1990; Hoaghia et al., 2017; Pavel et al., 2013). Essential heavy metals (Fe, Zn, Cu, Mg, Se, Co, Vn) have higher impact on fish, considering their important role in physiological and biological processes, compared with non-essential heavy metals (Al, As, Cd, Sb, Sn, Pt, Hg, Pb, Bi), which are considered potentially toxic trace elements

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due to the competition for binding sites with essential elements (Munoz-Olivas and Camara, 2001; Predescu et al., 2017). As water pollutants, heavy metals are of great importance because of their inability to decompose, long persistence, bio-accumulation and bio-magnification in the food chain (Brinza et al., 2007; Hlihor et al., 2009; Tuzen, 2009). Burada (2014b) suggested that some metals have a greater toxic impact on the environment, compared with others such as Cd, Cr, Cu, Pb, Ni, and Zn. According to Bat et al. (2015), heavy metals tend to accumulate in organisms situated in the top of the food chain (such as fish in aquatic environment), through bio-magnification effects. In human nutrition, fish are a great source of proteins and fatty acids and fish meat is appreciated for its high nutritional value. Thus, metals may enter the human body and accumulate in the human tissues to cause chronic toxicity. Chronic assimilation of heavy metals is a known cause of cancer (Bat et al., 2015). According to Abdulali et al. (2012), food is the main route by which heavy metals enter the human body and bound irreversibly to body tissues (Cd bounds to kidneys and Pb bounds to bones). The fish available on the market for human consumption has two sources: wild fishing and aquaculture. There are several beliefs that wild fish are healthier, compared with the ones raised in aquaculture. However, wild fish stocks grow in an uncontrolled environment, so they tend to accumulate different pollutants released into the waters. According to Martins et al. (2011), recirculating aquaculture systems may be an alternative to reduce the risk of consuming contaminated fish from the wild or from other production systems, more susceptible to environmental pollution.

The Black Sea is a semi-enclosed sea, surrounded by 6 countries (Bulgaria, Romania, Ukraine, Georgia, Russia, and Turkey), being in the past one of the most biologically productive seas in the world (Jitar et al., 2014). The percentage of coastal Black Sea distribution is: 36% Ukraine, 35% Turkey, 8.64% Russia, 8.5% Bulgaria, 6.61 Georgia and 5.25% Romania (Stanchev et al., 2011).

Between the '60 -'70, in the Black Sea there were 26 commercially fish species, which generated a production of hundreds of thousands of tones. This number decreased gradually. Therefore, nowadays there are approximately 10 commercial important pelagic fish species, all small sized: sprat (*Sprattus sprattus)*, whiting (*Merlangius merlangus euxinus)*, anchovy (*Engraulis encrasicolus)*, horse mackerel (*Trachurus mediterraneus ponticus)*, toad goby (*Mesogobius batrachocephalus)*, round goby (*Neogobius melanostomus)*, bluefish (*Pomatomus saltatrix)*, grey mullet (*Mugil cephalus)*, red mullet (*Mullus barbatus ponticus)*, sand smelt (*Atherina boyeri*) and 3 benthic species: turbot (*Psetta maxima maeotica)*, blackhand sole (*Solea nasuta)*, European flounder (*Platichtys flesus luscus)* (Patras, 2009).

The Black Sea is considered as one of the most polluted seas, fact directly related with the input of Danube, Dnieper and Dniester rivers. Annually, large amounts of organic and inorganic substances reach the sea (Table 1), both by river and through discharges of sewage and industrial wastewaters (Oros and Gomoiu, 2010; Stancheva et al., 2014).

In 1992, Zaitsev conducted a study estimating therefore the annually discharging rate of certain elements (mineral nitrogen, mineral phosphorus, organic phosphorus, iron, oil and oil products, detergents, zinc, lead, mercury, copper, arsenic, chromium, phenol), into the Black Sea (Table 1). Fish are often used as bio-indicators to assess the concentration and accumulation rate of heavy metals, as they are selective on the type of metal they concentrate (Depledge and Rainbow, 1990). Essential metals are found in many enzymes and respiratory pigments, therefore having an important role in metabolic processes (Oros and Gomoiu, 2010). Given the fact that carboxy-peptidase contains zinc, hemoglobin iron, hemocyanin contains copper and pyruvate carboxylase manganese, fish will accumulate these metals better than others (Oros and Gomoiu, 2010).

Table 1. Estimated annual quantity of elements discharged into the Black Sea (Zaitsev, 1992)

Elements	***Tones/year***
Mineral Nitrogen	575,000
Mineral Phosphorus	55,000
Organic Phosphorus	30,000
Iron	90,000
Oil and Oil Products	206,000
Detergents	48,000
Zinc	12,000
Lead	45,000
Mercury	80
Copper	2,800
Arsenic	1,700
Chromium	1,500
Phenol	2,200

Besides the type of metal, other factors influence the heavy metals accumulation in fish such as: fish size, sex or reproductive cycle and, the bioavailability of the metal in the water and environmental hydrodynamics (Burada et al., 2017; Petisleam et al., 2007).

Heavy metals use different paths to enter the fish body, like gills, skin, food or drinking water (Saidi et al., 2013). The uptake efficiency of a heavy metal through gills and food depends on the availability of the element (if it's dissolved in the water or concentrated in the food). The accumulation thorough the skin depends on the fish size and the toxicant (Heath, 1995). For example, small fish tend to accumulate a higher quantity of heavy metals because of the large surface to the volume ratio (Heath, 1995). This study is an overview regarding heavy metals uptake efficiency, which aims to present centralized data analysis, reported on Black Sea coastal area, for various species of fish. Marine fish drink about 0.5% of their body weight per hour, compared to freshwater fish, which ingest only a small amount of water, making them more exposed in accumulating metals than freshwater fish (Heath, 1995).

2. Heavy metals concentrations in different fish species

2.1. Strategies for paper retrieval and selection

The heavy metals concentration in aquatic organisms is a popular topic nowadays, fact that generates a large number of scientific publications. Therefore, it was necessary to narrow the searching domain in order to gain an appropriate number of articles for review. As discussed above, the fingerprinting is a hot topic which generated a large number of articles after the initial retrieval. It was necessary to narrow the searching domain in order to gain an appropriate number of articles for review. The searching was considering to be best achieved by using a variety of search methods (electronic and manual) and by searching multiple, possibly overlapping resources. Most of the searching took place at the beginning of the review with an update search towards the end. The following retrieval strategies were adopted:

- the journals were selected through the analysis of previous studies that classified and ranked the most significant key journals
- the papers published period ranged from the years 2003 to 2016, because the papers published earlier had lost their steering effects to the work, since heavy metals detection technologies and innovation methods have a very quick upgrading tendency.

Although the retrieval strategies adopted, 103 articles were retrieved, some filtration rules were further developed to identify the paper quality and value as follows:

- the latest papers on heavy metals topic, published in a certain journal, are prioritized in order to be used in this overview, during the scientific sources selection process.
- the researches which have the highest visibility are priority

Limited factors in the process of scientific papers selection:

- the extent of searching is determined by the research question and the resources available to the research team.
- the existence of a bibliographic software in order to manage references would help the documenting the process, streamline document management and make easier the production of reference lists for this journal papers.

2.2. Global data

The Marine Strategy Framework Directive (EC, 2008) establishes a framework for the development of marine strategies designed to achieve Good Ecological Status in the marine environment, by the year 2020, using 11 qualitative descriptors (EC, 2008). The concentration of contaminants includes heavy metals in the marine environment and their effects need to be assessed taking into account the impacts and threats to the ecosystem (Bat et al., 2015).

Several studies have been conducted (Bat et al., 2012, 2015; Culha et al., 2016; Jitar et al., 2014; Makedonski et al., 2015; Mendil et al., 2010; Stancheva et al., 2010, 2013, 2014; Turan et al., 2009; Tuzen, 2003, 2009; Uluozlu et al., 2007) in order to establish the heavy metal level in fish from the Black Sea. The sampling locations of those studies are presented in Fig. 1 and the data reported in those studies are centralized in Table 3. For the Romanian sector, the author has chosen the specific locations due to the identified pollution sources which are four municipal wastewater treatment plants with potential impacts on the sampling sites (Jitar et al., 2014). In the Bulgarian sector the authors (Makedonski et al., 2015; Stancheva et al., 2010, 2013a, 2013b, 2014) have chosen the specific locations due to the limited data about heavy metals pollution of the Bulgarian Black Sea coast for the last twenty year and that recently agricultural, industrial developments and population increase have substantially generated the contamination of Bulgarian region of Black Sea. For the Turkish sector, the authors (Bat et al., 2012, 2015; Culha et al., 2016; Mendil et al., 2010; Turan et al., 2009; Tuzen, 2003, 2009; Uluozlu et al., 2007) have chosen the sampling locations based on the lack of information about the real level of heavy metal pollution in Turkey region and the content of heavy metals in fish is extremely important for human health.

Tuzen (2009) explained that several analytical techniques are available for trace element determination in fish samples such as inductively coupled plasma optical emission spectrometry (ICP-OES), inductively coupled plasma mass spectrometry (ICP-MS), graphite furnace atomic absorption spectrometry (GFAAS), flame atomic absorption spectrometry (FAAS). For mercury determination at low concentrations, different analytical methods have been developed but the most commonly used is cold vapor atomic absorption spectrometry (CVAAS) The technique and apparatus used by each group of authors is as follows: Culha et al. (2016) used ICP-MS technique and Agilent 7700 apparatus; Makedonski et al. (2015) and Stancheva et al. (2010, 2013a, 2013b, 2014) used ICP-OES, ETAAS, FAAS technique and Perkin Elmer Zeeman 3030, Perkin Elmer Zeeman 1100B, Milestone DMA-80, Varian AA-240 apparatus; Bat et al. (2015) used ICP-MS technique and m-AOAC 999.10 method; Jitar et al. (2014) used GFAAS technique and GBC Avanta 3000 apparatus; Bat et al. (2012) used the ICP-MS technique but did not specified the utilized apparatus; Mendil et al. (2010), Turan et al. (2009), Tuzen (2009, 2003), Uluozlu et al. (2007) used ICP-OES, FAAS, GFAAS, CVAAS technique and Varian Model Liberty Series II, Perkin Elmer Analyst 700 apparatus.

Metal accumulation in fish depends on several factors such as: level of exposure, physiological factors (age, metabolic activity) and environmental factors (temperature, physico-chemical parameters and the presence of other metals) (Kim et al., 2004).

Table 3. Data regarding heavy metals concentrations in different fish species according to different authors

Fish Species	***Reference***	***Heavy Metals (μg/g)***								
		Pb	***Cd***	***Fe***	***Zn***	***Cu***	***Hg***	***Ni***	***Cr***	***As***
Scorpaena porcus	Culha et al. (2016)	0.04*	-	-	-	0.07*	0.01*	0.01*	-	0.14*
Pomatomus saltatrix	Makedonski et al. (2015)	0.03*	0.008*	-	10*	0.8*	0.09*	0.009*	-	0.77*
Mugil cephalus		0.05*	0.012*	-	5.2*	0.34*	0.05*	0.009*	-	1.1*
Trachurus mediterraneus ponticus		0.06**	0.008**	-	8.5**	0.56**	0.16**	0.008**	-	0.73**
Alosa pontica		0.05*	0.007*	-	9.0*	0.45*	0.08*	0.07*	-	0.38*
Sprattus sprattus		0.08**	0.005**	-	11**	1.4**	0.12**	0.028**	-	0.73**
Neogobius melanostomus		0.03*	0.006*	-	9.00*	0.76*	0.05*	0.008*	-	0.660*
Sarda sarda		0.06*	0.015*	-	10.0*	0.660*	0.130*	0.11*	-	0.410*
Mullus barbatus	Bat et al. (2015)	<0.05*	<0.02*	2.3*	3.2*	<0.5*	<0.05*	-	-	1.3*
Merlangius merlangus		<0.05*	<0.02*	0.87*	3.4*	<0.5*	<0.05*	-	-	1.24*
Liza aurata		<0.05*	<0.02*	3.2*	2.9*	<0.5*	<0.05*	-	-	0.25*
Trachurus mediterraneus		<0.05*	<0.02*	2.2*	24.7*	0.67*	<0.05*	-	-	0.39*
Pomatomus saltatrix	Stancheva et al. (2014)	0.03*	0.008*	5.0*	10*	0.8*	0.09*	0.009*	0.06*	0.77*
Mugil cephalus		0.05*	0.012*	2.2*	5.2*	0.34*	0.05*	0.009*	0.07*	1.1*
Trachurus mediterraneus ponticus		0.06**	0.008**	4.2**	8.5**	0.56**	0.16**	0.008**	0.03**	0.73**
Alosa pontica		0.05*	0.007*	9.00*	9.00*	0.45*	0.08*	0.07*	0.05*	0.38*
Sprattus sprattus		0.08**	0.005**	9.0**	11.0**	1.40**	0.12**	0.028**	0.04**	0.73**
Alosa pontica	Jitar et al. (2014)	0.20 *	0.02*	-	-	2.25*	-	0.06*	0.11*	-
Sprattus sprattus		0.07*	0.06*	-	-	1.91*	-	0.25*	0.05*	-
Mullus barbatus		0.32*	0.02*	-	-	3.48*	-	0.27*	0.02*	-
Atherina boyeri		0.16*	0.03*	-	-	2.92*	-	0.07*	0.03*	-
Sprattus sprattus	Stancheva et al. (2013a, 2013b)	0.08*	0.005*	-	-	-	0.12*	-	-	0.73*
Neogobius melanostomus		0.03*	0.006*	-	-	-	0.05*	-	-	0.66**
Mugil cephalus		0.07*	0.024*	-	-	-	0.08*	-	-	0.90*
Trachurus trachurus mediterraneus	Bat et al. (2012)	0.23*	0.048*	-	32.38*	6.21*	-	-	-	-
Sprattus sprattus		0.28*	0.09*	-	45.35*	7.77*	-	-	-	-
Mullus surmelutus		0.10*	0.035*	-	19.71*	5.39*	-	-	-	-
Sarda sarda		0.19*	0.028*	-	17.56*	4.12*	-	-	-	-
Mugil cephalus		0.19*	0.03*	-	42.65*	4.61*	-	-	-	-
Scorpaena porcus		0.07*	0.023*	-	12.3*	1.70*	-	-	-	-
Pomatomus saltatrix	Stancheva et al. (2010)	0.07*	0.07*	6.51*	-	1.34*	-	-	-	-
Merlangius merlangus	Mendil et al. (2010)	0.46***	0.18***		27.7***	1.8***	-	-	-	-
Trachurus trachurus mediterraneus		0.64***	0.22***	-	25.7***	2.4***	-	-	-	-
Sarda sarda		0.28***	0.35***	-	21.0***	1.9***	-	-	-	-
Mullus barbatus		0.40***	0.23***	-	17.8***	1.4***	-	-	-	-
Engraulis encrasicholus	Tuzen (2009)	0.30***	0.27***	-	38.8***	1.96***	0.055***	-	-	-
Merlangius merlangus		0.53***	0.21***	-	65.4***	1.32***	0.084***	-	-	-
Trachurus trachurus mediterraneus		0.82***	0.32***	-	52.7***	0.65***	0.078***	-	-	-
Sarda sarda		0.61***	0.13***	-	64.9***	1.43***	0.025***	-	-	-
Mullus barbatus		0.36***	0.17***	-	75.5***	0.96***	0.084***	-	-	-
Mugil cephalus		0.68***	0.35***	-	86.2***	2.14***	0.070***	-	-	-
Psetta maxima		0.28***	0.10***	-	45.2***	0.75***	0.045***	-	-	-
Pomatomus saltatrix		0.87***	0.23***	-	93.4***	2.78***	0.062***	-	-	-
Engraulis encrasicholus	Turan et al. (2009)	0.329*	0.124*	-	25.416*	-	-	-	-	-
Merlangius merlangus		0.502*	0.192*	-	6.029*	-	-	-	-	-
Mullus barbatus		0.727*	0.208*	-	7.573*	-	-	-	-	-
Engraulis encrasicholus	Uluozlu et al. (2007)	0.33***	0.65***	-	40.2***	0.95***	-	-	-	-
Merlangius merlangus		0.93***	0.55***	-	48.6***	1.25***	-	-	-	-
Trachurus trachurus mediterraneus		0.68***	0.50***	-	37.4***	0.95***	-	-	-	-

Fish Species	*Reference*	*Heavy Metals (µg/g)*								
		Pb	*Cd*	*Fe*	*Zn*	*Cu*	*Hg*	*Ni*	*Cr*	*As*
Sarda sarda		0.76***	0.90***	-	48.7***	0.84***	-	-	-	-
Mullus barbatus		0.84***	0.45***	-	106.0***	0.98***	-	-	-	-
Mugil cephalus		0.61***	0.45***	-	40.2***	1.26***	-	-	-	-
Pomatomus saltatrix		0.38***	0.60***	-	35.4***	1.83***	-	-	-	-
Engraulis encrasicholus	Tuzen (2003)	0.39*	0.18*	-	18.85*	1.96*	-	-	-	-
Trachurus trachurus mediterraneus		0.83*	0.48*	-	11.41*	1.55*	-	-	-	-
Sarda sarda		0.26*	0.10*	-	13.72*	1.29*	-	-	-	-

*Muscle tissue. ** Whole fish. *** Unspecified anatomical fish part. Original centralized table which uses data obtained by the specified authors.

Fig. 1. Sampling locations in different studies of heavy metals concentration in the Black Sea

Differences in heavy metals concentration were related to diet and feeding habits of benthic and pelagic fish species. Bustamante et al. (2003) show that benthic fish generally accumulates higher concentrations of heavy metals than pelagic fish. According to Elnabris et al. (2013), fish muscle is not an active tissue in heavy metals accumulation process and different fish species contained different concentrations of heavy metal in their muscles.

According to Boran and Altinok (2010), significant differences regarding heavy metals concentration were found between various species. Therefore, high levels of heavy metals in *Mugil cephalus* were attributed to habitat and feeding behavior of this species (Elnabris et al., 2013), while in case of *Engraulis encrasicholus*, the same phenomenon is explained by the differences related to diet, feeding habits and migration routes (Gilmartin and Revelante, 1975). In her study, Martins et al. (2009) suggested that the accumulation of heavy metals affects in different ways, small and large individuals.

In aquatic environment, metal toxicity and bioavailability can be influenced by various abiotic factors, such as pH, water hardness, alkalinity and accumulation of humic substances (Martins et al., 2011). The toxicity of metals increases with alkalinity, pH, salinity, temperature and conductivity (Radulescu et al., 2015). For many metals, alkalinity is a cofactor much more important than hardness (Burada, 2014b). The contaminant uptake rate is positively linked to the metabolic rate in marine organisms. Thus it can be supposed that metal accumulation would be higher (Kojadinovic et al., 2007). Duran et al. (2014) suggested that the origin of metals in the fish is highly related to the different chemical form of the metal in the aquatic environment. Kojadinovic et al. (2007) suggested that fish muscle impregnation is the result of heavy metal accumulation from preys, which contained high level of heavy metals concentration, living in different environments, other than where the fish were caught.

According to Elnabris et al. (2013), the following factors can influence heavy metals concentration, in different fish species: water, temperature, pH, dissolved oxygen and water transparency. He also stated that the geographical location and season of catch could lead to different metal concentrations even in the same fish species (Elnabris et al., 2013).

In his study, Burada (2014b) revealed important interspecific differences due to trophic and ecological features, as well as intraspecific variation, depending on the type of analyzed tissue or sampling location. Also, he observed an upward tendency of heavy metals in water, in the spring season, explained by the phenomenon of snow melting and spring rains, which induces important changes (both qualitative and quantitative) at the water level, due to the washing of the ground surface (Burada, 2014b). Thus, this trains different pollutants in the water, changing the physico-chemical characteristics of the water. In the summer season, he observed a decrease of heavy metals concentration in the water, explained by their tendency to fix in sediments and plants (Burada, 2014b). In his study, Coban et al. (2009) suggested that the heavy metals concentration, along the Turkish Black Sea coast, is higher than in other coastal areas and that solubility of heavy metals in seawater varies depending on the elements, however Gorur et al. (2012) concluded in their study that the concentrations of As, Mn, Fe, Cr, Ni, Zn, Cu and Pb were far below the established maximum values by the European Community regulations.

2.3. Lead concentration in various fish species from the Black Sea area

The European legislation (EC, 2006) established the maximum level of Pb permitted in the muscle tissue of fish of 0.30 mg/kg wet weight. The Joint FAO/WHO recommend a provisional tolerable weekly intake (PTWI) of 0.025 mg/kg body weight, for human adults (World Health Organization, 2007). The Turkish Food Codex, established the maximum lead level of 0.3 mg/kg fresh weight for sea fish (Turkish Food Codex, 2008). The highest concentration of Pb was registered in *Merlangius merlangus* (0.93 µg/g f.w.), in the Samsun area, Turkish coast (Fig. 2). This is the only reported Pb concentration which exceeds the maximum limit established by both Turkish Food Codex (Turkish Food Codex, 2008) and European legislation (EC, 2006). In 2008, Turkmen et al. found in their study that in Bartin coastal area (Turkey), the highest concentrations of trace elements were represented by Pb and Cu.

Merlangius merlangus is a benthic fish and according to Oros and Gomoiu (2012) benthic fish generally accumulates higher concentrations of heavy metals than pelagic fish, fact explained by its association with sediments substrate. Thus, it can directly uptake metals by ingesting the sediment particles (Oros and Gomoiu, 2012). The lowest concentration of Pb was registered in *Pomatomus saltatrix* and *Neogobius Melanostomus* (0.03 µg/g f.w) in Nessebar area, Bulgarian coast (Fig. 2).

In water, Pb is present as ion Pb^{2+} or as insoluble Pb complexes (Burada et al., 2014). The solubility of Pb is 10 ppb at pH value of 8 and at pH value of 6.5, the solubility reaches even over 100 ppb (Burada et al., 2014). A lot of factors influence Pb absorption, including water physico-chemical parameters, exposure rate and surface, water pH and water hardness. Pb and Cd have no biological role in fish organism and hence, they are harmful to fish organism, even at considerably low concentrations (Elnabris et al., 2013). The pathogenic effect of lead is multifactorial since it directly interrupts the activity of enzymes, competitively inhibits absorption of important trace minerals, and deactivates antioxidant sulphydryl pools (Saidi et al., 2013).

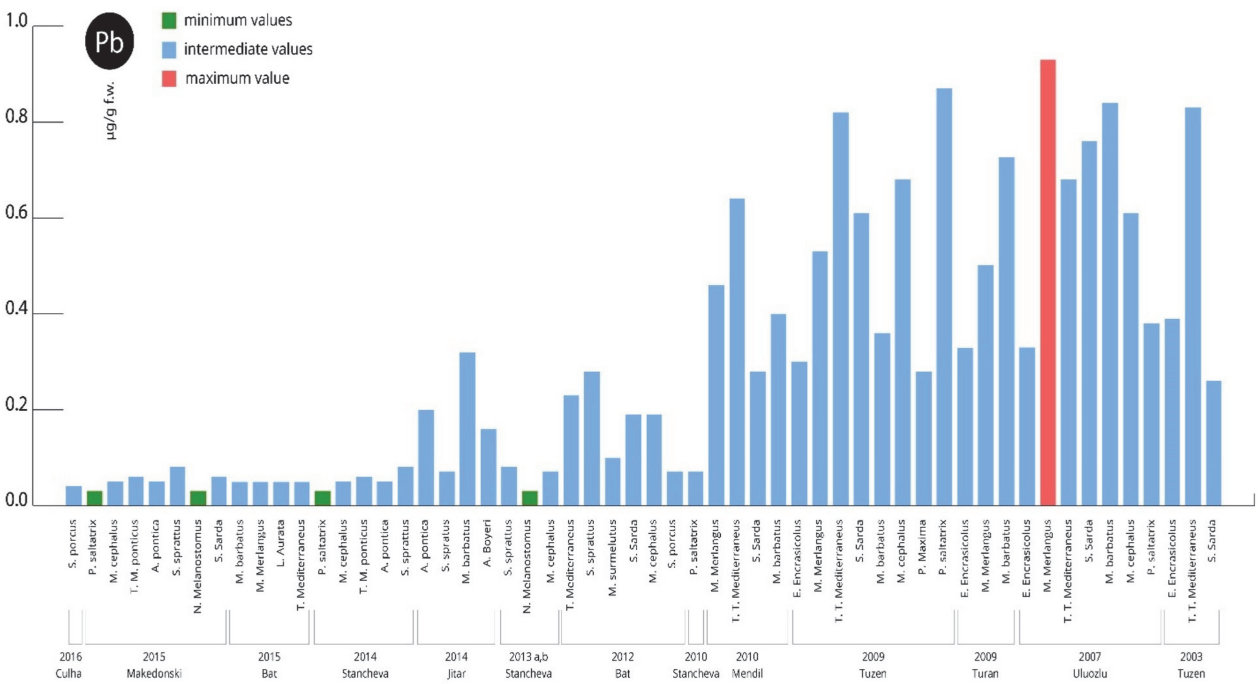

Fig. 2. The Pb concentration in different fish species in the Black Sea area according to different authors*.
*Original graphic which uses data obtained by the specified authors

2.4. Cadmium concentration in various fish species from the Black Sea area

The present European legislation, (EC, 2006), established the maximum level permitted for Cd in the fish muscle as 0.050 mg/kg wet weight, except for *Engraulis species*, *Sarda sarda, Mugil labrosus labrosus* and *Trachurus species*, in which the maximum level permitted is 0.10 mg/kg wet weight. FAO and WHO recommend the provisional tolerable weekly intake (PTWI) as 0.007 mg/kg body weight for human adults (World Health Organization, 2007). The Turkish Food Codex established the maximum level at 0.10 mg/kg for sea fish (Turkish Food Codex, 2008).

As we can observe (Fig. 3), the highest concentration of Cd was registered in *Sarda sarda* (0.90 µg/g f.w.) in Samsun area, Turkish coast. This is the only reported Cd concentration which exceeds the maximum limit established by both Turkish Food Codex (Turkish Food Codex, 2008) and European legislation (EC, 2006).

The lowest concentration of Cd was registered in *Sprattus sprattus* (0.005 µg/g f.w.) in Nessebar area, Bulgarian coast.

According to Jitar (2015), along the Turkish Black Sea coast, more exactly in Samsun area, 2 rivers flow into the sea: Kizilirmak and Yeşilirmak. Investigations revealed that the river Yeşilirmak discharges the highest quantity of pollutants (Jitar, 2015). Thus, in this area, the total concentration of Cd and Cr registered higher values due to the discharge rate (Jitar, 2015). In Oros and Gomoiu study (2012), conducted along the Romanian Black Sea coast, Cd registered the highest average values in case of sprat, anchovy, goby and flounder (1.06 – 1.40 µg/g f.w. Cd) and the lowest values for bluefish and horse marckerel (0.31 – 0.49 µg/g f.w. Cd). Another explanation of the high Cd concentration can be related to cephalopod-based diet (Bustamante et al., 2003).

Studies revealed that increasing salinity will also increase Cd absorption by submerse plants, and if salinity increases, it determines the formation of cadmium chloride, which cannot be absorbed by plants. If salinity concentration increases even more, the Cd present in the chlorides suspended in the water will be replaced by Na, generating therefore an increase of Cd concentration (Prasad, 2001).

In case of low salinity water, Cd is present as free ion Cd^{2+}, as $Cd(OH)_2$ and the organic complexes, depend on the pH and the quantity of soluble organic material (Burada, 2014a). The easiest way for the aquatic organisms to absorb Cd is in its free form, as Cd^{2+}, raising the salinity and therefore reducing the bio-accumulation (Burada, 2014a). The mobility and bioavailability of Cd in the aquatic environments is amplified by low pH value, low hardness value, low level of suspended solids, high redox potential and low salinity value (Burada, 2014a). Cd does not bind to organics as fast as copper, fact that makes it much more available for the fish to absorb (Heath, 1995).

2.5. Zinc concentration in various fish species from the Black Sea area

The Turkish Food Codex established the maximum Zn level permitted of 50 mg/kg for fish (Turkish Food Codex, 2008). The Joint FAO/WHO established the Zn provisional tolerable weekly intake (PTWI) for 7 mg/kg body weight (World Health Organization, 2007). The highest concentration of Zn (Fig. 4) was registered in *Mullus barbatus* (106 µg/g f.w.), in Samsun area, Turkish Black Sea coast. This is the only reported Zn concentration which exceeds the maximum limit established by Turkish Food Codex (Turkish Food Codex, 2008).

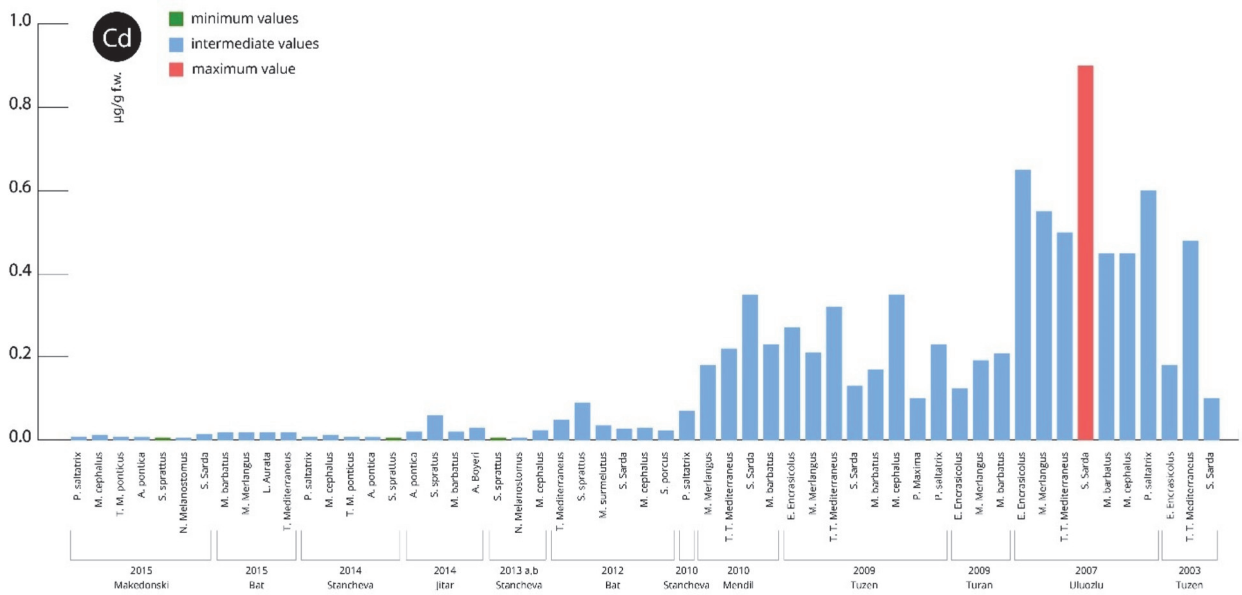

Fig. 3. The Cd concentration in different fish species in the Black Sea area according to different authors*.
*Original graphic which uses data obtained by the specified authors

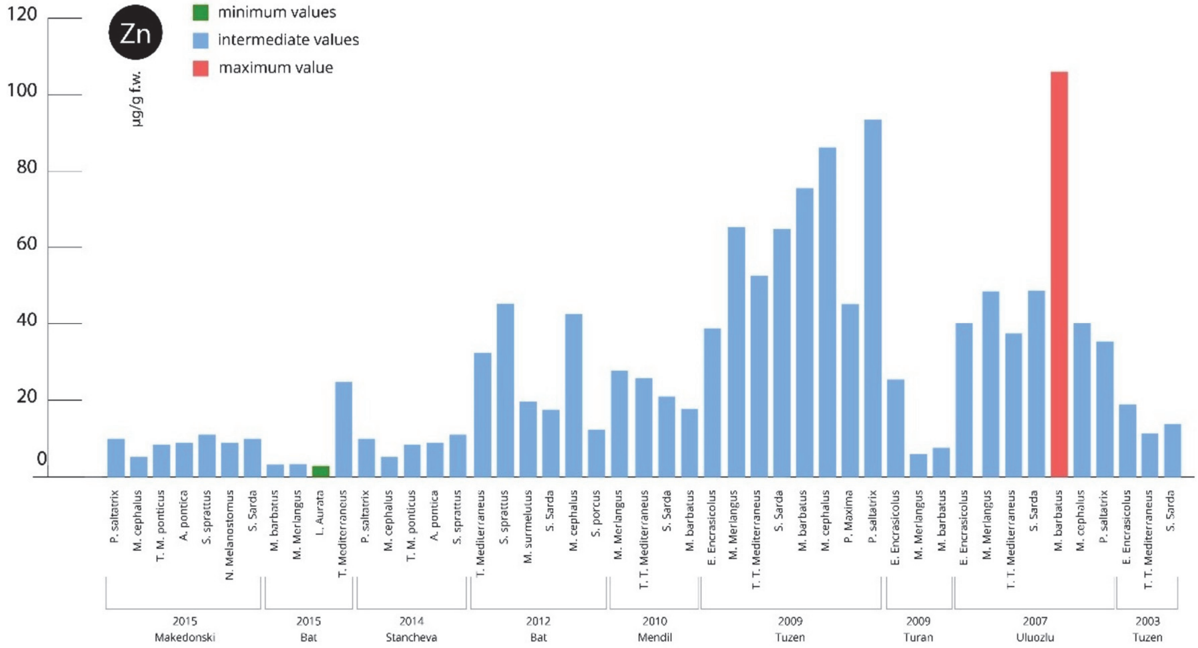

Fig. 4. The Zn concentration in different fish species in the Black Sea area according to different authors*
*Original graphic which uses data obtained by the specified authors

According to Aygun and Abanoz (2011), Zn concentrations were found to be highest in Samsun. The lowest concentration of Zn (Fig. 4) was registered in *Liza aurata* (2.9 µg/g f.w.) in Sinop area, Turkish coast. Acording to Jitar (2015), Zn has a higher remanence time in the water suspension than other metals, making it more available for the fish to upake. In his study, Burada (2014b) suggested that Zn is more toxic for fish at high temperatures, compared with low temperatures.

2.6. Iron concentration in various fish species from the Black Sea area

In case of Fe (Fig. 5), the highest concentration was registered in *Alosa pontica* and *Sprattus sprattus* (9±1 µg/g f.w.) in Burgas, Bulgarian coast and the lowest concentration in *Merlangius merlangus* (0.87 µg/g f.w.) in Sinop area, Turkish coast. Fe, Cu and Zn are essential metals and they play an important role in biological systems (Turkmen et al., 2009). According to Heath (1995) and Elanbris et al. (2013), essential metals will tend to accumulate in a higher concentration than non-essential metals, fact that can be explained by their involvement in metabolism function. In case of fish, the existing mechanism for metals uptake from the environment implies proteins and amino acids interaction (Heath, 1995). Fe availability may be enhanced by consumption of foods containing ascorbic acid (Stancheva et al., 2010).

2.7. Copper concentration in various fish species from the Black Sea area

The Turkish Food Codex established the maximum Cu level permitted of 20 mg/kg for sea fish (Turkish Food Codex, 2008). The Joint FAO/WHO established the provisional tolerable weekly intake (PTWI) of 3.5 mg/kg body weight (World Health Organization, 2007). Ocean fish and sea vegetables contain Cu (Duran et al., 2009). The highest concentration of Cu (Fig. 6) was registered in *Sprattus sprattus* (7.77 µg/g f.w.) in Sinop area, Turkish coast. The lowest concentration (Fig. 6) was found in *Scorpaena porcus* (0.07 µg/g f.w.) in as well Sinop area, Turkish coast. In their study, Oros and Gomoiu (2010) showed that Cu did not register major interspecific differences.

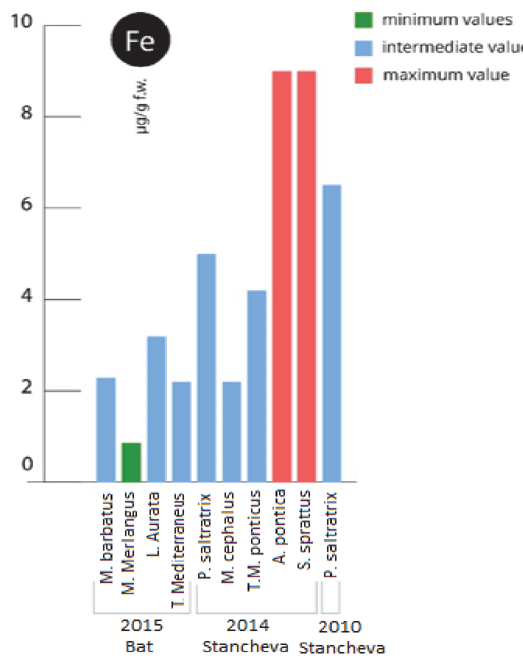

Fig. 5. The Fe concentration in different fish species in the Black Sea area according to different authors* (*Original graphic which uses data obtained by the specified authors)

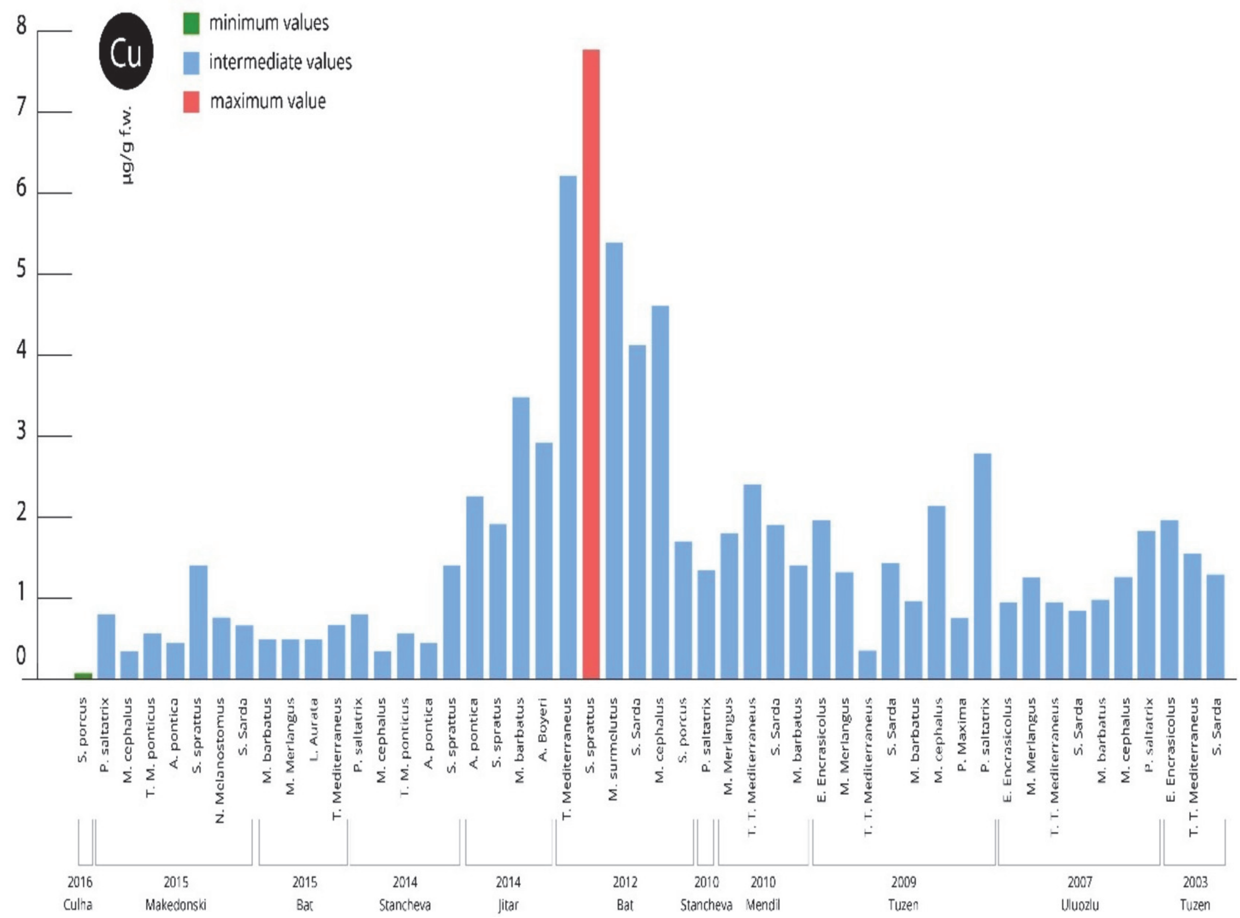

Fig. 6. The Cu concentration in different fish species in the Black Sea area according to different authors*.
*Original graphic which uses data obtained by the specified authors

According to El-Moselhy et al. (2014), the accumulation of essential metals is probably linked to their role in metabolism, so high levels of Cu and Zn in fish tissues are usually related to a natural binding protein such as metallothioneins. As well, they mentioned that benthic fish, such as *Mullus barbatus,* are more likely to have higher heavy metals concentration than fish inhabiting the upper water column (El-Moselhy et al., 2014). The richest source of Cu in fish diet is organ meat, especially liver (Stancheva et al., 2010). Additional contributions to intake of Cu may come from adventitious sources, such as copper-containing fungicides sprayed on agricultural products (RDA, 1989). Wurts and Durborow (1992) suggested that a lower water alkalinity value generates more toxic and soluble forms of Cu.

In his study Duran et al. (2014) registered significant higher values (1.79±0.09 µg/g f.w.) of Cu concentration in *Sarda sarda,* compared with other fish species. The pattern of Cu concentration in the muscles tissues, according to Duran is as follows: *M. barbatus>D. labrax=E.encrasicholus>P.maxima=P. saltatrix>Soleidae>E. aenus>Salmothymus=B. belone*.

2.8. Mercury concentration in various fish species from the Black Sea area

The European legislation established the maximum level of admitted Hg concentration in fish muscle tissue of 0.5 mg/kg wet weight, except for *Mullus* species and *Sarda sarda*, which have the maximum permitted level of 1.0 mg/kg wet weight (EC, 2006). The Turkish Food Codex established the maximum level permitted for fish meat of 0.5 mg/kg wet weight (Turkish Food Codex, 2008). WHO established the provisional tolerable weekly intake (PTWI) of 0.0016 mg/kg body weight for human adults (World Health Organization, 2007).The highest concentration of Hg was registered in *Trachurus mediterraneus ponticus* (0.16 ± 0.02 µg/g f.w.), which is a pelagic fish, in Nessebar area, Bulgarian coast (Fig. 7). The lowest concentration of Hg was noted in *Scorpaena porcus* (0.01 µg/g f.w.), which is a predacious fish, in the Sinop area of the Black Sea (Fig. 7). The higher levels of accumulation in planctonivorous fish (anchovy, sprat) compared with predators fish may be explained on the basis of a higher efficiency of metals assimilation from food (Oros and Gomoiu, 2010).

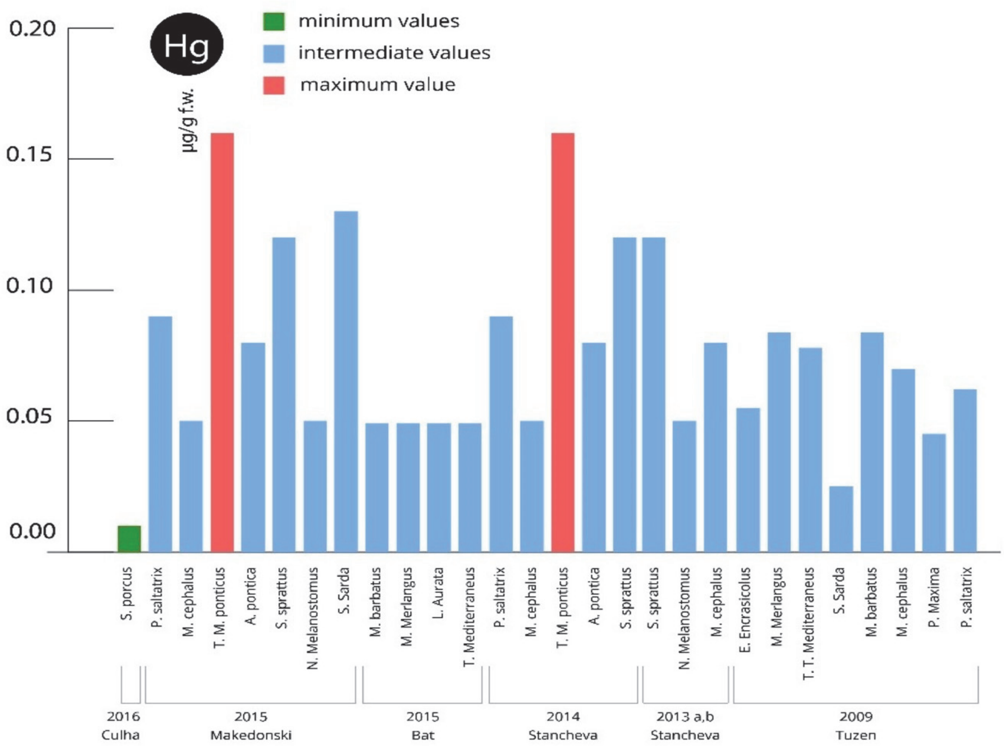

Fig. 7. The Hg concentration in different fish species in the Black Sea area according to different authors*.
*Original graphic which uses data obtained by the specified authors

2.9. Nickel concentration in various fish species from the Black Sea area

WHO (World Health Organization, 1992) recommends 100-300 μg Ni for daily intake for human adults. The highest concentration of Ni was found in *Mullus barbatus* (0.27 ± 0.13 μg/g f.w.) in Constanta area, Romanian coast (Fig. 8).

The lowest concentration was noted in *Trachurus mediterraneus ponticus* and *Neogobius melanostomus* (0.008 ± 0.001 μg/g f.w.) in Nessebar area, Bulgarian coast (Fig. 8). In Oros and Gomoiu study (2012), Ni showed higher average concentrations in turbot, anchovy, bluefish and sprat. A lot of the Ni complexes dissolve easily in water, fact that rises their availability of being absorbed by fish (Burada, 2014b).

It must be pointed out that Ni is an essential element that is required by a wide variety of enzymes and other cell components. Thus, it has vital functions in fish, although very high intake can cause adverse health problems (Elnabris et al., 2013).

2.10. Chromium concentration in various fish species from the Black Sea area

According to the Bulgarian Food Codex, the maximum Cr level permitted in fish tissue is established at 0.3 mg/kg (Regulation, 2004). The highest concentration of Cr was registered in *Alosa pontica* (0.11 ± 0.03 μg/g f.w.) in Constanta area, Romanian coast (Fig. 9).

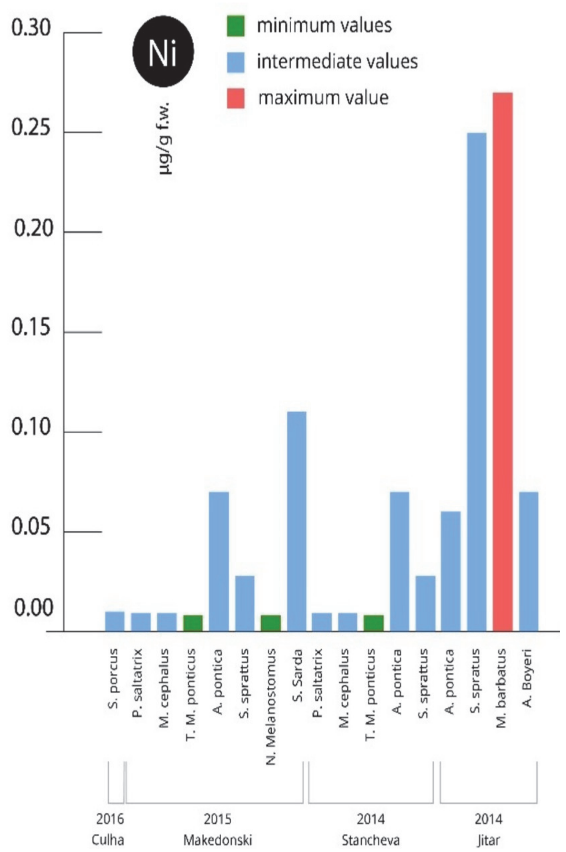

Fig. 8. The Ni concentration in different fish species in the Black Sea area according to different authors*
*Original graphic which uses data obtained by the specified authors

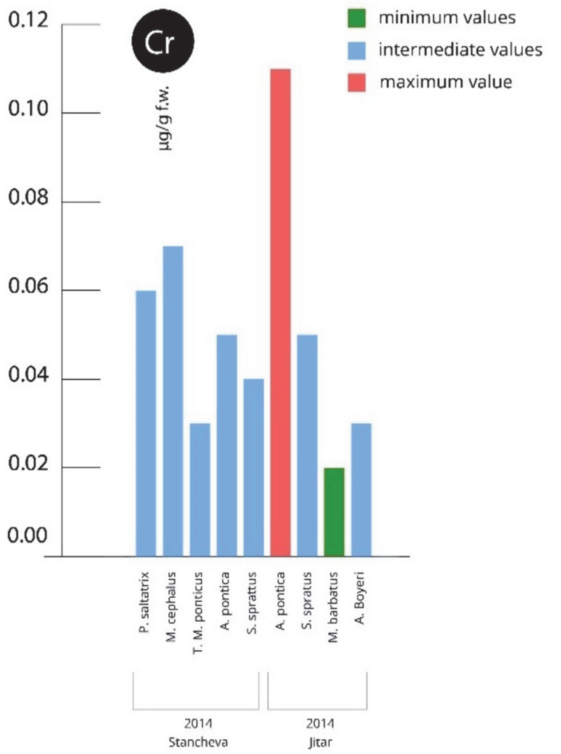

Fig. 9. The Cr concentration in different fish species in the Black Sea area according to different authors*. *Original graphic which uses data obtained by the specified authors

The lowest concentration was noted in *Mullus barbatus* (0.02 ± 0.002 µg/g f.w.), as well in Constanta area (Fig. 9). Cr is involved in insulin function and lipid metabolism (Stancheva et al., 2014). In water, Cr is normally precipitated as insoluble chromium hydroxide, formed by the reaction of trivalent Cr with hidroxilic ions (Burada, 2014b). Cr absorption is dependent on the water salinity (Jitar, 2015).

2.11. Arsenic concentration in various fish species from the Black Sea area

The Joint FAO/WHO established a provisional tolerable daily intake (PTDI) of 2.1 µg/kg body weight per day, for As (FAO/WHO, 2011). The Turkish Food Codex, published in 2002, stated that the maximum level of As for human consumption must not exceed 1 mg/kg body weight (Boran and Altinok, 2010). The toxicity resulting from As exposure is considered to be linked to an imbalance between pro-oxidant and anti-oxidant homeostasis, that results in oxidative stress (Ventura-Lima et al., 2011).

The highest concentration of As was noted in *Mullus barbatus* (1.3 µg/g f.w.) in Sinop area, Turkish coast (Fig. 10). The lowest concentration was reported in *Scorpaena porcus* (0.14 µg/g f.w.) as well, in Sinop area (Fig. 10). It was pointed out by Ventura-Lima et al. (2011) that marine fish have a higher capacity for As accumulation, compared to freshwater fish, due to water salinity.

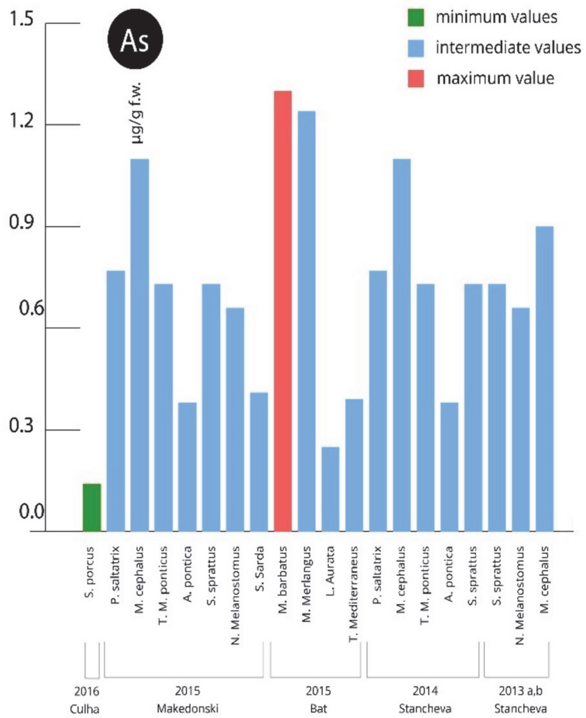

Fig. 10. The As concentration in different fish species in the Black Sea area according to different authors*. *Original graphic which uses data obtained by the specified authors

3. Conclusions

The main conclusion of the present research reveals the highest concentrations of heavy metals in fish tissues associated to red mullet (*Mullus barbatus*), followed by bluefish (*Pomatomus saltatrix*). Also, among all studied metals, Zn had the highest concentrations in all fish species, followed by Fe, Cu and As.

The maximum concentration of heavy metals reported in most of the revised studies did not exceed the maximum permitted levels established by the present European and Turkish legislation. The only exceptions are found in Samsun area (Turkey), in case of Cd from *Sarda sarda,* lead from *Merlangius merlangus* and zinc from *Mullus barbatus*.

Another conclusion that must be pointed out from the current review is that benthonic fish are more exposed to heavy metals accumulation, compared with pelagic ones. Also, the existence of both interspecific and intraspecific differences regarding heavy metals concentration in fish tissues is confirmed.

The Central South part of the Black Sea (Turkish marine coastal area, represented by the perimeter situated between the Kizilirmak and Yeşilirmak rivers) has proven to be more polluted in terms of heavy metals accumulation in fish meat, compared with South-Eastern part (Turkish coastal area) and the Western part (Romanian and Bulgarian coastal areas).

The information from this current study are meant to be used in order to evaluate the heavy metals concentration and distribution among several fish species with commercial importance, along both West and South parts of the Black Sea coastal areas.

The data presented in this research can provide support in order to identify the most polluted areas, fact that contributes to the development of various sustainable regeneration programs.

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