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HEAVY METALS ACCUMULATION IN BLACK SEA ECOSYSTEMS: SEAWATER, SEDIMENT, ALGAE, BENTHIC ORGANISMS

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ABSTRACT

The aim of the current review study was to present data on the accumulation of various heavy metals in the Black Sea ecosystems. Subject of study were Pb, Cd, As, Hg, Mn, Ni, Cu, Zn, Fe etc. and their content in seawater, sediment, algae and various benthic organisms. Available data from the Bulgarian coast and also from different Black Sea areas were presented.

Key words: heavy metals, Black sea, algae, mussels, benthos

Introduction

The Black Sea is the largest semi-enclosed sea in the world and widely perceived to be heavily polluted. Together with Azov Sea, it covers an area of 462000 km^2. Its east to west dimension is 1150 km and from north to south is 610 km. The depth of water approaches 2200 m and is virtually isolated from other seas (Readman et al., 2002). The Black Sea is surrounded by six countries located in Europe and Asia: Bulgaria, Georgia, Romania, Russia, Turkey and Ukraine (Fig. 1). To the south and southwest, the Strait of Bosphorus connects the Black Sea to the Sea of Marmara, which in turn, is connected to the Aegean Sea and Mediterranean Sea through the Strait of Dardanelles. To the north it is open to the Azov Sea through Kerch (Strezov, 2012). In fact, the Black Sea is influenced by 17 countries, 13 capital cities and some 160 million people. The distinguishing feature of the Black Sea is the shallow (150–200 m deep), oxic and biologically active top layer. The remaining 90% of its volume is anoxic because of the lack of water exchange (Tuncer et al., 1998; Readman et al., 2002).

The annual supply of sedimentary material to the Black Sea by all debouching rivers amounts to ~ 150x10^6 t. (Degens et al., 1980). Based on the salinity and temperature distribution, the Black Sea water column can be divided into three distinct water masses: (1) an upper layer (0–150 m, 17–18.5-ppt salinity); (2) a cold intermediate layer (100–200 m, 6.5–8.0°C), part of a larger intermediate layer (200–1000 m, > 21-ppt salinity, > 8.5 ° C); and (3) a bottom/deep convective layer (> 1000 m; 22.2–22.3-ppt salinity; 9-9.2 ° C) (Ovchinnikov and Popov, 1986). There exists a permanent lack of vertical mixing, and thus, anoxic conditions are prevailing in the Black Sea (Emery and Hunt, 1974).

Black Sea pollution with heavy metals is one of the major environmental challenges surpassing in importance the contamination with radionuclides (Strezov, 2008). Heavy metals are introduces through natural geological processes, rivers or by direct discharge of industrial waste (Mitryasova et al., 2020). Oil products contamination of seawater is another major source of such metals.

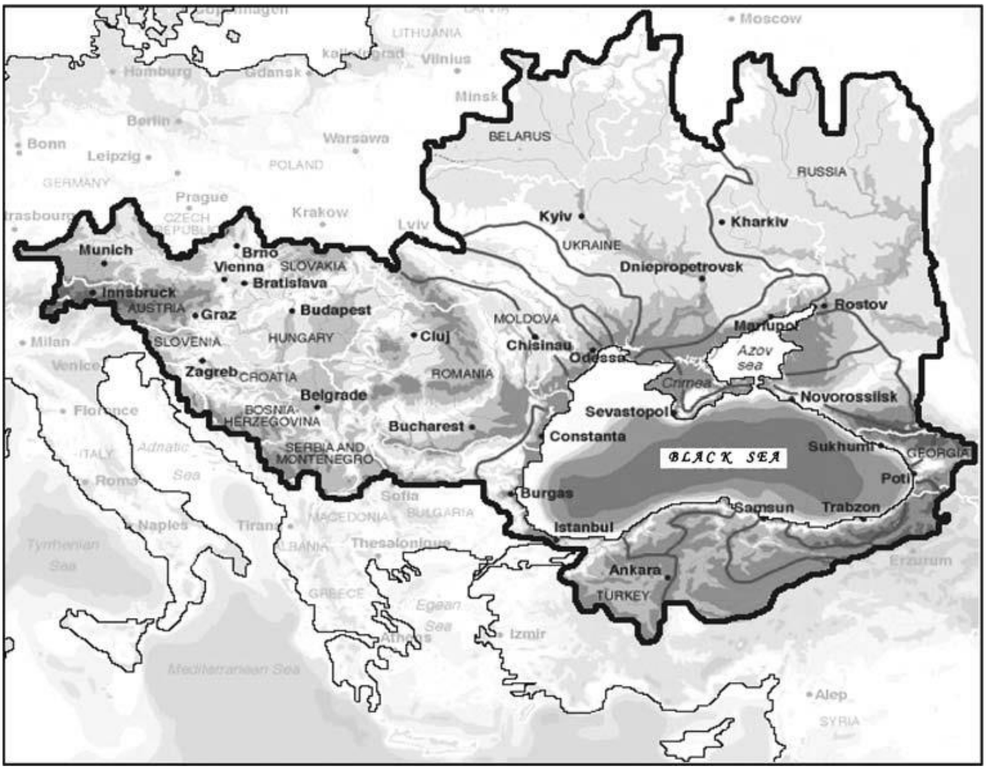

Figure 1: Black sea basin (by Altas and Buyukgungor, 2007).

Significant sources of pollution in the Danube River and in Black Sea are untreated wastewater (main impact – Bulgaria) and industrial sources (main impact – Romania) which are potential sources of heavy metals (Danube Basin Analysis, 2004).

The aim of the current review survey was to present the main data for heavy metals accumulation in water, sediment as well as in important bioindicators of enviromental pollution like algae and benthic species from various areas of Black sea in the last decades (1974–2020).

Heavy metals pollution in Black Sea water and sediment

Heavy metals pollution level in seawater is not constant and depends to a large extent on the quality of the drainage basins (Altas et al., 2001; Ariman and Bakan, 2008).

Sediments are an important repository for various pollutants such as pesticides and heavy metals and also play a significant role as sensitive indicators for monitoring contaminants in aquatic environment (Simpson and Batley, 2016). Seawater and sediment represent a major abiotic factor for sea ecosystems. Heavy metals are regarded as serious pollutants of aquatic ecosystems and a major environmental problem in the Black Sea (Bakan and Böke Özkoç, 2007).

The presence of such elements in sediments from different geographical areas is characterized by a high degree of variability, depending on the element, sediment type, distance from shore and the influence of anthropogenic sources (Coatu et al., 2013).

Heavy metal pollution study of Peycheva et al. (2017b) of seawater and sediment in Varna and Bourgas areas in 2013 estimated the following highest concentrations: Zn (15-22 μg/L), As (1.1–

1.2 μg/L) and Pb (0.7–0.8 μg/L) in water and Zn (31–52 μg/g), Pb (21-29 μg/g), Cu (20–34 μg/g) in sediment. Water and sediments showed similar spatial distribution patterns of various metals.

The range of heavy metal concentrations in water of the Romanian part of Black sea coast during 2006–2011 framed within the following limits: Cu between 0.01 to 93.51 mg/L, Cd between 0.01 to 18.32 mg/L, Pb between 0.01 to 51.97 mg/L, Ni between 0.01 to 30.59 mg/L, Cr between 0.01 to 59.74 mg/L. In sediment, heavy metals concentrations varied within the following limits: copper between 0.53 to 147.84 mg/g, cadmium between 0.01 to 9.63 mg/g, lead between 0.10 to 300.78 mg/g, nickel between 0.40 to 211.73 mg/g, chrome between 1.34 to 231 mg/g (Coatu et al., 2013).

A survey of heavy metal pollution of Romanian Black sea coast water in 2017 reported elements concentrations as follow (μg/L): Cd – 14.34–18.7, Co – 90.12–119.58, Zn – 32.58–48.88 and Pb – 13.14–24.22 (Mititelu et al., 2018). The comparison with the results of Coatu et al. shows significantly narrower boundaries of variation in values in the recent study.

Sea water pollution analysis in the Asian vicinity of Istanbul shown Ni concentrations below the recommended marine recreational water quality criteria but the lead in seawater were above the limit (Bozkurt et al., 2014).

Contamination by heavy metals should be extensively studied in the Black Sea coast, even though these are subjected to intense discharges of pollutants. It is important, therefore, that sediment by heavy metals be assessed in order to facilitate better management and protection of these valuable coastal ecosystems. This is especially the case, since Turkish Black Sea coast represents a prominent area for fishing, industrial development and urban extension and tourism activities (Bat and Özkan, 2015).

Sediments constitued up to 39% $CaCO_3$, mainly of biogenic origin from the shell remains of benthic organisms. Relatively little is known about the heavy-metal geochemistry of superficial Black Sea sediments adjacent to the North Anatolian coasts. The concentrations of Cr, Ni, Cu, Zn and Pb in the southern Black Sea sediments are markedly higher than those found in average sedimentary rocks (Yucesoy and Ergin, 1992).

A sediment pollution assessment study was carried out using the enrichment factor and geo-accumulation index which displayed that the surface sediment of the Southern Black Sea was not contaminated with respect to Cu, Pb, Zn, Mn, Fe, As, Sr, Bi, V, Cr, Mg, Zr, Y, Sb, Sn, and Ni, but is moderately contaminated with respect to Hg and Cd, and moderately to strongly polluted with respect to S and Mo due to agricultural activities and industrialization. (Ozkan and Buyukisik, 2012).

The results of Bat et al., (2015a) indicated that contamination of surface sediments in Sinop coast was dominated by As (10.2–7.4 mg/kg^{-1}), Cr (67–374 mg/kg^{-1}) and Hg (0.07–0.03 mg/kg^{-1}) and to a lesser extent Cu (7.24–5.09 mg/kg^{-1}), Fe (1.76–1.12 mg/kg^{-1}), Zn (19.3–13.8 mg/kg^{-1}), Ni (16.2–12.5 mg/kg^{-1}), Cd (0.06–0.04 mg/kg^{-1}), Pb (7.12–6.32 mg/kg^{-1}), Mn (470–227 mg/kg^{-1}) and Co (9.5–5.9 mg/kg^{-1}). Although high trace metal concentrations were found in some regions of the Black Sea coast of Turkey, the level of most of them were not extremely enriched in these surface sediments of Sinop coast and did not present a serious threat to the local fauna and flora.

Available data concerning the northern Black Sea area were insufficient. A three years surveillance of heavy metals pollution in water and sediments of Odessa region of the Black Sea reported annual variations in the content of Cu, Zn, Ni, Cd in water with mean content of 1.86, 10.30, 1.14, 0.67 mcg/dm^{-3} respectively. Corresponding concentrations in sediment were 3.03, 14.76, 2.25, 0.32 mcg/dm^{-3}. The conclusions were that most of the copper and nickel was brought into the sea from the catchment area, and zinc and cadmium were mainly of the autochthonous origin. The observation was that after heavy rains, sediments contamination by copper and nickel was increased (Dyatlov, 2015).

Significant pollution of the Azov Sea by petroleum products and heavy metals has been proven (Mazygula et al., 2019).

Heavy metals accumulation in Black sea algae

Analysis of sediments suffers from limitations. The concentration of a metal in sediment depends amongst other factors, on rates of deposition and the nature of particles and as before does not reflect bioavailability (Villares et al., 2001). That is why living organisms like various sea macroalgae could be more useful for assessment of bioavailability (Philips, 1990; Topcuoglu et al., 2001).

Brown algae *Cystoseira barbata* and *C. crinita* can accumulate trace elements in concentrations that exceed their concentrations in the Black Sea water by 3–4 orders which makes them an efficient bioindicator for heavy metal pollution. Algae samples from Sevastopol region (SW Crimea, Ukraine) showed higher levels in summer than in the spring. Among the most important abiotic factor is the concentration of elements in the environment, e.g sea water. The reported concentration for some of the tested elements were as following (in μg/g): Al – 253, Mn – 24, Fe – 188, Co – 0.42, Ni – 4.98, Zn – 35, As – 30 (Kravtsova et al., 2014).

The study of Dotsenko and Mikhailenko (2019) examined the importance of phytoplankton for the bioaccumulation of microelements in the ecosystem of the Azov Sea. The fact that algal biomass during the periods of blooming in the sea reaches 1,400 g/l makes phytoplankton essential part of marine ecosystems. Due to their ability to develop quickly in large quantities, phytoplankton plays active role in the biogeochemical cycling of elements. Most of the organic matter produced by algae and elements connected with these are destroyed and dissolved directly within the water column reentering the biogeochemical cycling.

The algae *Cystoseira barbata* and *Ceramium rubrum,* sampled from Constanta coast (Romania) were tested for Pb, Cd, Cu and Zn. Reported results were 2.51, 0.201, 4.19, 1.93 mg/kg and 5.04, 0.231, 6.58, 6.72 mg/kg respectively. The results indicated higher concentration of heavy metals in red algae probably because it lives at higher depths, where solar radiation cannot reach and part of the metals cannot enter the biogeochemic circuit and used for photosynthesis (Cadar et al., 2016).

Levels of Hg, Cd, Pb, Ni, Cr, Mn, Zn, and Cu were determined in five macroalgae (*Enteromorpha intestinalis*, *Cladophora vagabunda*, *Cystoseira barbata*, *Ceramium rubrum* and *Phyllophora pseudoceranoides*) collected in 2014 from the Roumanian Black Sea coastline (Trifan et al., 2015). Metal concentrations decreased in the following order Mn > Zn > Cu > Ni > Pb > Cr > Cd > Hg. A wide range of metal uptake ability among the different species was observed: *C.rubrum* showed a high capacity to accumulate Cu and Cr, *P.pseudoceranoides* – Mn and Pb, *C. vagabunda* – Hg, Zn and Cd, and *E. intestinalis* – Ni; hence, these species could be used as bioindicators for the respective metals. In the cited study, red algae displayed the highest capacity to concentrate heavy metals from surrounding water and sediment, followed by green and brown algae.

Another widespread alga in Black Sea is *Ulva lactuca*. The following lowest and highest concentrations (mg/kg; dw) were measured in *Ulva* samples from Istanbul Coast of the Sea of Marmara: 0.45–3.22 for Cd, 6.67–18.32 for Cu, 553.32–989.33 for Fe, 8.27–25.32 for Mn, 4.93–19.32 for Pb, and 15.16–41.30 for Zn (Ozyigit et al., 2017). Overall, the comparison of heavy metal contents in seawater and sediment with samples of the *Ulva* species showed that *U. lactuca* is a suitable plant for biomonitoring studies.

Zooplankton plays a pivotal role in shaping ecosystem structure. Concentrations of Al, As, Cu, Zn, Hg, Fe, Cd and Pb were determined in the total zooplankton samples from Sinop coast of Black Sea. Zooplankton mainly *Copepods* are the most abundant forms in the Black Sea. They filter suspended matter and have important role in transferring the organic matter from primary producers to the higher trophic levels. It is also one of the most preferred foods for the fish. The average concentration of heavy metals followed the order: Fe > Al >Zn > Pb > Cu > As > Cd > Hg. Estimated Hg was below the detection limit among all heavy metals in all samples. Fe was the most common heavy metal in total zooplankton samples present in all, Al was the other heavy metal commonly present in the samples. Pb showed high value (210 mg/kg^{-1}) in total zooplankton. Similar to Pb, a high average concentration of As (40 mg/kg^{-1}) was observed in total zooplankton. Cd was 1.1 mg/kg^{-1} (Bat et al., 2016b).

Heavy metals accumulation in benthic organisms

Marine macro benthic organisms are widely accepted to be suitable biological indicator for water pollution degree, including heavy metal (Azizi et al., 2018). Mussels from genus *Mytilus* and especially *Mytilus galloprovincialis* are worldwide recognized as pollution bioindicators and used in Mussel Watch programs, because they accumulate pollutants in their tissues at elevated levels in relation to pollutant biological availability in the marine environment (Catsiki and Florou, 2006). As filtering organisms, large volumes of water enter in contact with their body surface, causing the accumulation of pollutants. Furthermore, it is one of the main commercially important Black Sea mollusc. Frequent consumption may leads to chronic exposure and health problems for humans, because of which toxic chemicals must be periodically and carefully monitored.

Available recent data for accumulation of heavy metals in benthic species from Bulgarian Sea coast is quite scarce. The study of Peycheva et al. (2017a) included wild and farmed Black Sea mussel (*M. galloprovincialis)* and rapa whelks (*R. venosa*) which were collected during 2016 from six sites on the northern part of the coast. The mussels and rapa whelks soft tissue was analyzed for ten elements (Cd, Cr, Cu, total Hg, Ni, Zn, Pb, Mn, Fe). Concentrations of these metals, in mg/kg dry weight, ranged from 0.73–3.45 for As, 0.005–0.640 for Cd, 0.040–0.382 for Cr, 0.86–7.70 for Cu, 4.2–112.9 for Fe, 0–0.121 for total Hg, 0.260–3.190 for Mn, 0.023–0.642 for Ni, 0–0.332 for Pb and 7.5–38.2 for Zn. The concentration of these elements was below the maximum residual levels prescribed by different local and international regulation for seafood.

Values of metal accumulation in 4 species of molluscs encountered along Romanian coast (filtrate bivalves *Mytilus galloprovincialis*, *Scapharca inaequivalvis*, *Mya arenaria* and predator gastropod *Rapana venosa*) were compared by Oros and Gomoiu (2010). They established significant interspecific differences, due to the trophic and ecological features of each species. Bivalve molluscs showed an increased potential to accumulate toxic metals compared to gastropods (Oros and Gomoiu, 2012).

The study of Mititelu et al. (2018) carried out assessment of bioaccumulation of the main heavy metals in mussel from Romanian Black sea coast. The availability in flesh and shell were comparable and varied according to the area of sampling. Coast parts with more intense industrial activity resulted in increased heavy metal pollution degree. The content in the mussel flesh was in (µg/g) –

Cd – 16.64-19.79, Co – 138.35-256.21, Zn – 142.29 – 149.61, 73.56- 104.68 and for mussel shell – 16.19-19.45, 122.45-196.12, 133.65 – 148.11, 76.63-102.19 respectively.

Established concentrations of some of the heavy metals in *Mytilus galloprovincialis* from Turkish area of Black Sea were (in μg/g f.w.): 1.03–1.65 for Cu, 0.02–0.92 for Cd, 0.05–0.37 for Pb, 0.57–3.44 for Ni and 0.06–1.08 for Cr (Topcuoglu et al., 2002).

Trace element content in the Mediterranean mussel (*Mytilus galloprovincialis),* caught from Sinop coast of the Black Sea, Turkey in 2010 were estimated to be lower than in other regions and previous studies (Bat et al., 2012). Metal concentrations were in the range (in $\mu g/g^{-1}$ dry wt): Zn – 79–163, Cu – 2.41–4.82, Pb – 2.10–4.10, Cd – 0.27–0.98.

A recent assessment of the levels of Fe, Zn, Mn, Cu, Pb, Cd and Hg in the edible tissues of Mediterranean mussel (*Mytilus galloprovincialis)* picked up from the Black Sea coasts of Turkey indicated no chronic systemic risk since total hazard index (0.521) were quite below critical value 1, and the carcinogenic risk for heavy metals did not exceed the tolerable values (Bat et al., 2018a). The concentrations (mg metal kg^{-1} wet wt.) of metals ranged from 18–35 for Fe, 8–27 for Zn, 2.8–4.5 for Mn, 0.5–1.8 for Cu, 0.06–0.31 for Pb, 0.04–0.10 for Cd and 0.03–0.07 for Hg.

Furthermore, the experiments of Bat et al. (2013b) reported that the Mediterranean mussel was more sensitive to Cu than Pb and Zn, and Pb is less toxic to the mussels than Zn and Cu. Small sizes of the Mediterranean mussel were relatively resistant to the metals compared to medium or larger ones.

Another study from the same region in 2013 estimated the metal content in different benthic organisms – Mediterranean mussel (*Mytilus galloprovincialis*), veined Rapa whelk (*Rapana venosa*), and the warty crab (*Eriphia verrucosa*). Hg showed the least concentrations in all organisms, and was not detected in both *M. galloprovincialis* and *R. venosa*. Pb was also not detected in organisms, except in *M. galloprovincialis*. The highest values of Cu, Zn, and Fe were determined in *E. verrucosa*. Al was at the limit of detection in *R. venosa*, but was measurable in *E. verrucosa* (1.4 $\mu g/g^{-1}$ dry wt.) and *M. galloprovincialis* (1.4 $\mu g/g^{-1}$ dry wt.). As, Cu, Zn, Fe, and Cd levels in macrobenthic organisms ranged from 2.3 $\mu g/g^{-1}$ dry wt. in *M. galloprovincialis* to 6.7 $\mu g/g^{-1}$ dry wt. in *R. venosa*, 0.8 $\mu g/g^{-1}$ dry wt. in *M. galloprovincialis* to 53.9 $\mu g/g^{-1}$ dry wt. in *E. verrucosa*, 21.6 $\mu g/g^{-1}$ dry wt. in *R. venosa* to 54.8 $\mu g/g^{-1}$ dry wt. in *E. verrucosa*, 29.3 $\mu g/g^{-1}$ dry wt. in *M. galloprovincialis* to 86.5 $\mu g/g^{-1}$ dry wt. in *E. verrucosa*, and 0.32 $\mu g/g^{-1}$ dry wt. in *E. verrucosa* to 4.4 $\mu g/g^{-1}$ dry wt. in *R. venosa*, respectively. All elements were below the permissible level defined by international organizations, with the exception of the highest Cd level in *R. venosa* (Bat and Öztekin, 2016).

The elements concentration in Mediterranean mussel from Giresun coasts (east of Sinop) estimated that the descending order was as follow: Fe > Zn > Ni > Mn > As > Pb > Cu > Co >Se > Cr. The tested metals were with concentration of (in μg/g ww) 0.56 for Cr, 6.23 for Mn, 12.7 for Ni, 69.06 for Zn, 1.97 for Co, 2.65 for Cu, 161.08 for Fe, 3.16 for Pb, 3.16 for As, 0.62 for Se. The mean Pb and Zn contents were higher than the acceptable limit proposed by international standards (Tepe and Suer, 2016).

The comparison of metal content (Zn, Cu, Cd, Hg, and Pb) from the neighbored Marmara Sea established higher level of pollution in latter one. Mercury was not detected (<0.15 ppb) in any of the samples. The highest concentrations of Cu and Cd were 3.473 and 0.740 mg/kg^{-1} (wet weight), respectively, well below the maximum permissible levels. All samples contained Zn higher than 50 mg/kg^{-1}, while Pb was above the limits in some of the samples. Mussels from Marmara Sea were safe regarding Cu, Cd, and Hg but contained Zn and Pb above the permissible limits (Mol and Alakavuk, 2011).

Heavy metal monitoring in wild mussels (*Mytilus galloprovincialis)* from the Marmara Sea Coast of Tekirdag, Turkey resulted in assessment of eight heavy metals (Cd, Cr, Cu, Hg, Ni, As, Pb, and Zn). The concentrations (mg/kg d.w.) of these metals ranged from 1.20 to 2.79 for arsenic (As), 0.13 to 0.75 for cadmium (Cd), 0.42 to 2.46 for chromium (Cr), 1.55 to 3.5 for copper (Cu), 1.01 to 2.46 for nickel (Ni), 76.8 to 88.98 for zinc (Zn), and 2.67 to 9.2 for lead (Pb). These levels were lower than the permissible limits set by the European Commission (Dokmeci, 2017).

The content of Hg, Cd, Pb, Cu, Zn and Fe in the whole soft tissues of the limpet, *Patella caerulea*, from the Sinop region of Black Sea was evaluated (Bat et al. 2015b). Mercury was not detected and iron was seen to be the most abundant of the metals examined. The results showed that the monthly mean levels of all other metal levels in July were lower than those in May and June. In general, the heavy metal levels in the limpet *P. caerulea* were below the tolerance levels. Confirmation was made that the limpet *P. caerulea* is good bioindicator for monitoring of metal pollution in the coastal area.

The findings from the study of Bat et al. (2016) revealed that Cu, Co, Pb, Zn, Cd, Mn, Ni and Fe concentrations in the edible tissues of *Liocarcinus depurator* (decapoda), *Rapana venosa* (gastropoda) and *Mytilus galloprovincilais* (mollusca) decreased in the order: Fe > Zn > Mn > Cu > Ni > Pb > Co > Cd; Fe >Zn > Cu > Pb > Mn > Cd > Ni and Fe > Zn > Cu > Pb > Mn > Co > Ni > Cd, respectively and were lower than the maximum permissible limit as recommended by the Commission Regulation (EC, 2006). Furthermore, these benthic species are not only important food source but also significant bioindicators for heavy metal and organic pollution.

The comparison of metals accumulation in *Mytilus galloprovincilais* from the Black, Marmara, Aegean and Mediterranean Sea was able to formulate the level of heavy metals pollution in neighbored seas. In all tested mollusk samples As was with highest concentration in all areas – 4.23, 2.85, 1.96 and 1.38 mg/kg respectively. The estimated concentration of Cd was 0.097, 0.087, 0.143, 0.122 mg/kg; Hg – 0.405, 0.341, 0.079, 0.203 mg/kg and Pb in samples was found to be 0.375, 0.267, 0.405, 0.366 mg/kg. The reported results clearly indicated that mussel from Black Sea were with highest content of As and Hg from all seas (Kuplulu et al., 2018).

Again from Samsun area there were available data for different benthic organisms – *Capitella capitata, Nereis zonata, Platynereis dumerilii and Perinereis cultrifera* and the estimated metals content were compared to water and sediment pollution (Bat et al., 2019). Metals concentrations in water were found in the following order: Zn > Cu > Pb > Cd > Hg, whereas the order of abundance of these metals in sediments were as follow Zn > Cu > Pb> Hg >Cd. The results showed that the heavy metal levels in *C. capitata* were much higher than those in *N. zonata, P. dumerilii* and *P. cultrifera* and at the same time Zn showed the highest concentration in all species followed by Cu. *C. Capitata* is not edible species but significant metals deposition makes it a credible bioindicator. The Pb, Cu and Zn, Hg and Cd levels in water were somewhat seasonally dependent with highest concentration in summer. This study also showed that sedimentary concentrations of Hg, Cd, Cu and Zn collected in different seasons were with statistically not significant differences. Zn was found to be the highest and Cd was the lowest. The detected seasonal variations could be related to the hydrological parameters of the ecosystem. The measured metals concentration was higher in sediment than in water because of the organic matter on the bottom of the sea.

The study of Tan and Kizilkaya (2019) determined monthly changes of the heavy metal composition of tellina **(***Donax trunculus)* in Kefken (east of Istanbul) and the effects of seasonal temperatures on accumulation. The average amount of Pb was determined as 12.97 µg/g and of Cd, Cr, Co and Ni was 45 µg/g, 4.19 µg/g, 0.91 µg/g and 4.46 µg/g, respectively, so tellina can be conveniently consumed between April and October depending on the territory.

The concentrations of Cd, Hg and Pb in the whole parts of the worm *Nereis diversicolor* collected from the Turkish Black Sea coast have been measured for monitoring metal pollution in 2015 (Bat et al., 2018b). Amounts of Pb were found to be higher in sediment than those of *N. diversicolor*. On the other hand, Cd and Hg amounts in the worms were higher than the sediment while seasonal differences in the amounts of Cd, Hg and Pb in the sediment were not significant. However, there were differences in toxic metal levels between sizes of *N. diversicolor.* Larger specimen accumulated less toxic metal. It is concluded that the worm *N. diversicolor* are suitable biomonitors to assess changes in metal pollution in this coastal area of the Black Sea.

Heavy metals concentration in edible tissues of the brown shrimp *Crangon crangon* (Linnaeus, 1758) from the Southern Black Sea, Turkey was reported to be below the permitted levels for human consumption according to Commission Regulation (EC). Cadmium and Pb concentrations in *C. crangon* were the lowest heavy metal concentrations, while Fe concentrations were highest level observed in all seasons. The concentrations of Cd, Pb, Cu, Zn, Co, Mn, Ni and Fe were in the range of 0.228–0.481, 0.291–0.491, 5.85–14.77, 18–36, 0.24–0.61, 6–15, 2–6 and 30-58 µg. g^{-1} wet weight, respectively (Bat et al., 2013a).

Some other important and widespread benthic species in Black sea like warty crab (E*riphia verrucosa*) and brown shrimp (C*rangon crangon*) can be used as biomonitors for heavy metals pollution. The study of Erdem and al. (2015) established that the P, Ca, Cu, Zn, Ni, Cr and Mn contents in the shrimp were less than crab while Fe, Ni, and Mg contents in crab were less than shrimp ($P<0,05$). Except for Cr, trace element contents were higher in crab than in the shrimp. The trace element contents of the crab and shrimp were within food safety limits recommended by FAO/WHO and European Union. However, Pb contents of crab (1.27 µg g^{-1}) were over of the acceptability limit values.

Monitoring of heavy metal pollution in marine molluscs and crustaceans should be considered essential not only because of the ecological status assessment but also in terms of determining the potential human health risk (Jovic et al., 2012). The Bulgarian legislation related to heavy metal pollution of marine hydrobionts was fully synchronized with the European regulatory framework (Tabl. 1) (EC, 2006, SG, 2004; 2008).

Table 1: Maximum levels for certain contaminants in foodstuffs determined by (EC) No 1881/2006 of 19 December 2006

	LEAD	**Maximum level (mg/kg wet weight)**
1.	Crustaceans, excluding brown meat of crab and excluding head and thorax meat of lobster and similar large crustaceans (NephropidaeandPalinuridae)	0,50
2.	Bivalve molluscs	1,5
	CADMIUM	
1.	Crustaceans, excluding brown meat of crab and excluding headand thorax meat of lobster and similar large crustaceans (Nephropidae and Palinuridae)	0,50
2.	Bivalve molluscs	1.0

Conclusion

- Heavy metals pollution in seawater varied in different sampling areas but the detected concentrations did not present a serious threat to the local fauna and flora.
- Sediments are an important reservoir of heavy metals and also play a significant role as sensitive indicators for monitoring contaminants in aquatic environment.
- Macro algae from genus *Ulva* and *Ceramium* can facilitate the monitoring of heavy metals pollution in marine environment.
- *Mytilus galloprovincialis* is a worldwide accepted as a sensitive bioindicator of trace elements concentration dynamics.
- It should be accepted that mussels are among the main potential sources of heavy metals for the consumer, which requires strict and constant monitoring in the various Black Sea regions.

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