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The abundance and speciation of mercury in the Adriatic plankton, bivalves and fish – a review

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This article presents an overview of available research on mercury speciation in the most studied biota of the Adriatic Sea as a specific biogeochemical subunit of the Mediterranean. We present current knowledge about mercury concentration, speciation, spatial distribution and temporal changes in plankton, bivalves and fish from the Adriatic Sea. Results from mercury speciation studies in marine organisms are used to describe the extent of mercury bioaccumulation in the Adriatic Sea. Mercury biogeochemical cycle in the Adriatic is characterised by increased mercury concentrations from the water column, through plankton, bivalves and smaller fish to predator fish species. Although the Adriatic Sea has the highest mercury concentration in the whole Mediterranean Sea, fish species at the higher trophic levels have similar mercury concentrations everywhere in the Mediterranean, indicating incomplete understanding of the transfer mechanisms of mercury from seawater to upper trophic levels. As consumption of (contaminated) food is the main route of human exposure to mercury, it is of great importance to understand the influence of mercury contamination in the Adriatic Sea.

Key words: mercury, methylmercury, plankton, fish, mussel

INTRODUCTION

Mercury (Hg) is a toxic environmental pollutant due to its transport and transformation properties (DRISCOLL *et al.,* 2013; UNEP, 2013). Reactive chemistry of mercury drives its cycling through atmospheric, terrestrial, marine and biological compartment (FITZGERALD *et al.* 2007; MASON *et al.,* 2012). Methylmercury (MeHg) is a mercury species of the greatest concern because it readily bioaccumulates and biomagnifies through marine food web (MASON *et al.,* 2012; UNEP, 2013).

Phytoplankton and zooplankton

Although plankton represents the link between Hg marine cycle and higher trophic levels, this point of Hg entry into ocean food webs has not been extensively investigated (GOSNELL & MASON, 2015). This is due to low oceanic MeHg concentrations which impede measurements of MeHg uptake and trophic transfer under natural conditions (LAMBORG *et al.,* 2014; MASON *et al.,* 2012). Trace metal speciation in the marine environment is affected by microbial sequestration, composition and abundance of microorganisms, presence of natural complexing agents and remineralisation of organic matter (GOSNELL & MASON, 2015). Bioaccumulation factor (BAF) for MeHg ranges between 10^4-10^6 for phytoplankton (FITZGERALD *et al.,* 2007; LEE & FISHER, 2016; PICKHARDT & FISHER, 2007). BAF for MeHg increases by approximately $10^{0.5}$ units with the next trophic level (WATRAS & BLOOM, 1992), suggesting that each higher trophic level derives MeHg from the previous trophic level. MeHg bioaccumulation mechanisms in plankton and subsequent trophic transfer are still not well understood. Previous studies reported passive diffusion (LEE & FISHER, 2016; MASON *et al.,* 1996) and active transport of MeHg across the cell membrane (PICKHARDT & FISHER, 2007; WATRAS *et al.,* 1998). MeHg could be also co-transferred alongside the accumulation of strong MeHg ligands (e.g. cysteine, thiols) (MOYE *et al.,* 2002), or act as a surrogate for actively transported compounds (CHEN *et al.,* 2008). It seems that deep-sea sediments are not an important source of MeHg in the ocean plankton; HAMMERSCHMIDT & BOWMAN (2012) suggest that most of MeHg in phytoplankton (and subsequently in higher trophic levels) originates from production in the mixed layer of the open ocean.

Bivalves

Molluscs are one of the most important types of commercial shellfish with a variety of species cultured globally (STANKOVIĆ *et al.,* 2012). Bivalve molluscs are suspension feeders that filter phytoplankton, zooplankton and inorganic matter from the surrounding water through their gills (BARKER JØRGENSEN, 1990). The presence of trace metals in bivalves depends on environmental (location, metal concentrations in surrounding water, solubility, temperature, dissolved oxygen, pH, salinity) and biological factors (size, age, sex, species, feeding habits) (LEE *et al*., 1998; LOBEL *et al*., 1991; PHILLIPS, 1990). Mussels and oysters are convenient bioindicators of toxic metals in the marine environment due to their wide geographical distribution, sedentary filter-feeding and ability to accumulate high concentrations of contaminants relative to seawater (RAINBOW, 1995). An important mussel species for biomonitoring of environmental pollution in coastal areas is the Mediterranean mussel (*Mytilus galloprovincialis*). Mediterranean mussels are more extensively used than any other mollusc species such as oyster and clam, although their feeding habits are similar. In general, all shellfish are reliable pollution indicators due to their ability to bioaccumulate contaminants in their tissues, which can be used for the estimation of pollution with toxic metals in marine environment (EISLER, 1981; RAINBOW, 1995).

Fish

Fish are an important component of a well-balanced diet, providing a healthy source of energy, high-quality proteins, vitamins, essential metals and ω-3 polyunsaturated fatty acids (GRIBBLE *et al.,* 2016). However, consumption of carnivorous fish is the main pathway for human exposure to MeHg that can cause adverse health effect. Despite extensive research, exact reasons for elevated Hg concentrations in some fish species are still unknown (CHOY *et al.,* 2009). Nutrition is the dominant Hg uptake pathway (HALL *et al.,* 1997); fish at higher trophic levels are more likely to accumulate more Hg concentrations (STORELLI *et al.* 2002a). Benthic species have higher Hg concentrations due to their predatory lifestyle and longer lifespan (CHOY *et al.,* 2009; KOENIG *et al.,* 2013). Generally, Hg levels in fish increase with increasing size and age (ENDO *et al.,* 2008; HORVAT *et al.,* 2014). MeHg accounts for 75–100% of total Hg present in most fish species (BOSCH *et al.,* 2016; STORELLI *et al.,* 1998, 2005a). In

the fish muscles, it is bound to the protein fraction through sulfhydryl complexes (MASON *et al.,* 2006). Mercury distribution in fish tissues is governed by the uptake route connected with its chemical speciation (HORVAT *et al.,* 2014). Combined effects of external (physical and chemical properties of the marine environment) and internal factors (physiological and biochemical properties of fish) influence Hg uptake through biological membranes and its accumulation in tissues (CHOY *et al.,* 2009). These effects can lead to variations in metal accumulation within and among fish species, locations and seasons. This makes fish a good bioindicators for measuring Hg bioaccumulation in the marine environment (HORVAT *et al.,* 2014).

Objective

The objective of this paper is to present Hg bioaccumulation and biomagnification in selected marine organisms (plankton, bivalves and fish) and to emphasise the importance of Hg speciation in the Adriatic Sea as a specific biogeochemical subunit of the Mediterranean Sea. We present Hg concentrations in these organisms and statistically compare them to values obtained in different parts of the Adriatic and Mediterranean Sea in order to evaluate the current state of spatial Hg distribution. When possible, we also present temporal changes in Hg concentrations and corresponding statistical analysis in order to assess the effects of reduced anthropogenic Hg releases on its bioaccumulation in these organisms.

MATERIALS AND METHODS

Study area

The Mediterranean Sea is an elongated basin situated between Africa, Asia and Europe. The length of the basin along the east-west axis is 4000 km with a maximum width of 800 km and an average water depth of 1.5 km. The Strait of Sicily separates the Eastern and Western basins. The Mediterranean Sea exchanges water with the Atlantic Ocean through the Strait of Gibraltar, and with the Black Sea through the Bosporus Strait. Cinnabar deposits are the main natural Hg source – 65% of world's Hg reserves are located under the Mediterranean (ŽAGAR *et al.,* 2014). Main anthropogenic Hg sources are mining, cement production, coal combustion, and chlor-alkali plants (KOCMAN *et al.,* 2013; ŽAGAR *et al.,* 2014).

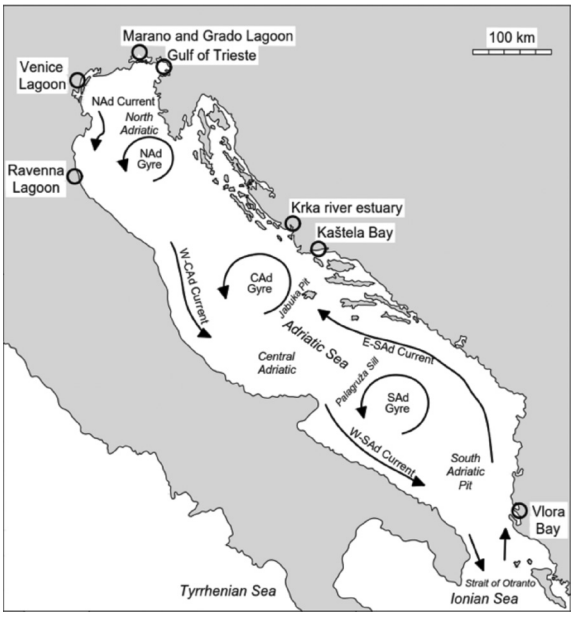

Fig. 1. Surface water circulation and geographical division of the Adriatic Sea. Adapted from KOTNIK et al. (2015).

The Adriatic Sea (Fig. 1) is an elongated basin situated in the northernmost part of the Mediterranean Sea. It is connected with the rest of the Mediterranean Sea through the Strait of Otranto. Based on different biogeochemistry, latitude and bathymetry, it is divided into northern, central and southern Adriatic (UNEP/MAP, 2012). The southern Adriatic is the deepest (~1250 m) and the northern is the shallowest with an average depth of 35 m. Central Adriatic is on average 140 m deep. Eastern and western South Adriatic currents (E-SAd and W-SAd), Central and Southern Adriatic gyres (CAd and SAd), and North Adriatic current (N-Ad) are the main water currents in the Adriatic Sea (Fig. 1).

Conversion of measurement units, analytical methods and statistical analysis

Mercury concentrations in different marine biota cover a wide range and are reported in lit-

erature on either wet weight (ww) or dry weight basis (dw). We converted all concentrations to ng g^{-1} or µg g^{-1} ww so that we could compare values with the EU maximal allowed levels in foodstuffs (EUROPEAN COMMISSION, 2006). We used literature data for transformations of dw to ww values, as follows. Water content in phytoplankton/microseston and zooplankton fractions is assumed to be 90 and 95%, respectively (KNAUER & MARTIN, 1972). Hg concentrations in bivalves and fish are converted using the average dw/ww conversion factors of six and five, respectively (COSSA *et al.,* 2012; CRESSON *et al.,* 2014; JOVIĆ & STANKOVIĆ, 2014; KLJAKOVIĆ-GAŠPIĆ *et al.,* 2007; NASCI *et al.,* 1998). We used these conversion factors throughout this review, unless otherwise stated in the literature.

The choice of analytical methods for the determination of total mercury (THg) and MeHg in environmental samples depends on sample matrix and expected mercury concentrations. The most currently used method for the determination of THg in biological samples is acid digestion (or microwave digestion) followed by reduction and suitable detection (atomic absorbance spectrometry, atomic fluorescence spectrometry, inductively coupled plasma mass spectrometry) (e.g. ANDRAL *et al.,* 2011; BOGDANOVIĆ *et al.,* 2014; MIEDICO *et al.,* 2013). Determination of MeHg requires previous separation from inorganic Hg compounds. From the sample, MeHg can be extracted with suitable organic solvent or leached with diluted nitric acid. Derivatisation of extracted/leached MeHg (usually ethylation) is followed by MeHg adsorption on trap, separation from other Hg compounds by gas chromatography and suitable detection after pyrolysis (e.g. STORELLI *et al*., 2002b; HAMMERSCHMIDT *et al.,* 2013).

Statistical analysis was performed using Stata 12.0 software. Differences between groups were determined using Mann-Whitney U test (two groups) and Kruskal-Wallis test (three or more groups), after testing normality of original and transformed data (Wilk-Shapiro test). In figures, the distribution of Hg concentrations is presented with box-and-whisker plot (median with 5, 10, 25, 75, 90 and 95th percentiles).

RESULTS AND DISCUSSION

Microbial role in trophic transfer of contaminants

The microbial community plays a critical role in the biogeochemical Hg cycling, influencing its chemical speciation, reactivity, and bioavailability through mediation of processes such as methylation, demethylation, oxidation, and reduction. The key process that drives Hg bioaccumulation is methylation. HINES *et al.* (2017) found high Hg methylation potential in the sediments from the Gulf of Trieste, where sulphate-reducing bacteria are often involved in the transformation of inorganic Hg compounds. PARKS *et al.* (2013) showed that the number of bacteria capable of methylating Hg is much higher. Oxic ocean water can also host biotic Hg methylation (HEIMBÜRGER *et al.,* 2010). The highest seawater MeHg concentrations are related to the presence of small algae (nano- and picophytoplankton), as shown for Western Mediterranean (HEIMBÜRGER *et al.,* 2010). Dissolved MeHg, which is the most biologically available organic Hg form, is bioaccumulated up to a million times in microscopic particles, including phytoplankton and bacteria, via adsorption to cell surfaces in the water column (MASON *et al.,* 1996). These MeHg-enriched particles are then consumed by zooplankton, which in turn are a primary food source for larval, juvenile, and some adult fish (HALL *et al.,* 1997).

Heterotrophic bacteria and autotrophic picoplankton (APP) (*Prochlorococcus*, *Synechococcus* and picoeukaryotes) represent the major components of the marine picoplankton community, especially in oligotrophic areas such as the Adriatic Sea (ŠOLIĆ *et al.,* 2010, 2015, 2016). These organisms are consumed by heterotrophic nanoflagellate grazers, which are consumed in turn by larger ciliated protozoa, forming a link ('microbial loop') to higher trophic levels (AZAM *et al.,* 1983) (Fig. 2). The reason for domination of these microorganisms in oligotrophic conditions is due to the number of advantages these organisms have for living in nutrient-poor environments: lower energetic costs (because of simple biomass composition), rapid metabolism and

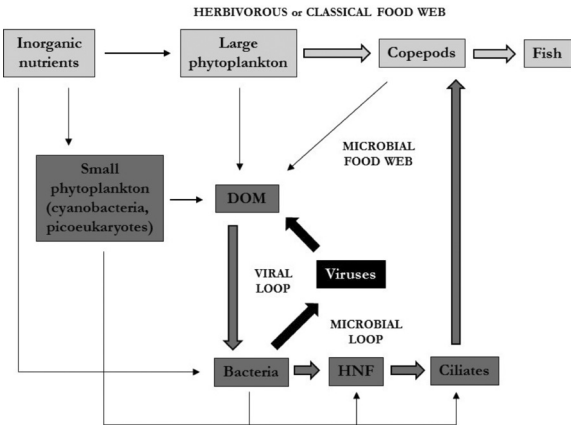

Fig. 2. Herbivorous (classical) food web versus microbial food web and role of "microbial loop" and "viral loop" within microbial food web. Redrawn from ŠOLIĆ et al. (2016).

small size which means less internal machinery and large surface/volume ratio (higher efficiency in nutrients uptake). The same factors that enable scavenging nutrients at low concentrations also facilitate contaminant accumulation in their biomass for subsequent transfer to other components in the food web. Prokaryotic heterotrophs accumulate contaminants more efficiently than phytoplankton (AXELMAN *et al.,* 1997), suggesting that prokaryotes may be a critical component of the food web that can promote our understanding of the behaviour of contaminants in marine systems.

FENCHEL (2008) suggested that autotrophic and heterotrophic organisms in the microbial loop play key roles in Hg transfer into marine food webs, influencing its biogeochemical cycling, as well as being major contributors to elemental cycling and vertical fluxes. One important impact of the microbial loop on Hg cycling in the water column is the acceleration of organic matter mineralization and the regeneration of nutrients to fuel primary production (FENCHEL, 2008). Furthermore, in comparison to classical herbivorous food web (large phytoplankton – large zooplankton – fish), microbial loop involves a higher number of trophic steps (bacteria – heterotrophic nanoplankton – microzooplankton – large zooplankton – fish), which implies greater concentration of contaminants along the food chain through biomagnification. Moreover, in eutrophic areas, where algal biomass increases, the concentration of MeHg per cell decreases, resulting in a lower dietary input to the herbivorous food web and actually reducing bioaccumulation in algal-rich eutrophic systems (PICKHARDT *et al.,* 2002; CHEN & FOLT, 2005). Further strengthening of the role of microbial food web could be expected in mainly oligotrophic Adriatic Sea if predicted rise of temperature in the near future results with increase of bacterial production (ŠOLIĆ *et al.,* 2017).

MERCURY IN PLANKTON

Mercury in phytoplankton/microseston

Mercury content in phytoplankton/microseston fraction is not often measured in the Adriatic or Mediterranean Sea. Therefore, available data do not allow systematic analysis of spatial and temporal trends. Seston can sometimes consist mainly of biological material other than phytoplankton. Analysis of material obtained by towing net (pore size 20 μm) in the Venice Lagoon showed that it was mostly composed of seagrass debris (DOMINIK *et al.,* 2014).

THg in phytoplankton (mostly dinoflagellates, diatoms and coccolithophorids) in Kaštela Bay (Fig. 1) was on average 315 ng g^{-1} (ZVONARIĆ *et al*., 1991). However, these samples were obtained during the operation of chlor-alkali plant that was releasing Hg-containing effluents directly into nearby sea. In the 1970s, FOWLER (1986) performed the most comprehensive analysis of THg in Mediterranean microseston. The overall average THg concentration was 5.40 ng g^{-1}. Similar to THg in zooplankton, western sub-basin had slightly lower THg content in microseston than eastern sub-basin (4.41 and 6.62 ng g^{-1}, respectively). To our knowledge, Hg speciation data in the Adriatic and Mediterranean phytoplankton are not available.

Average THg concentration in the Mediterranean seston is higher than in other world seas: 3.88 ng g^{-1} in NW Atlantic (HAMMERSCHMIDT *et al.,* 2013), 0.77 ng g^{-1} in Long Island Sound (GOSNELL *et al.,* 2017), 3.00 ng g^{-1} in the Baffin Bay (CAMPBELL *et al.,* 2005), 2.00 ng g^{-1} in the Baltic Sea (NFON *et al.,* 2009), and 1.95 ng g^{-1} in

the Ross Sea (BARGAGLI *et al.,* 1998). The higher average value found in the central Pacific Ocean (6.89 ng g^{-1}) shows that phytoplankton community concentrates more Hg from the water in the oligotrophic open ocean (GOSNELL & MASON, 2015). Direct comparison of these values should be taken with reserve however, due to differences in sampling methods, sampling years, and environmental conditions that influence Hg distribution in the sea. Furthermore, differences could also be attributed to changes in dominant organisms within the phytoplankton community (GOSNELL *et al.,* 2017).

Mercury concentrations in zooplankton from different seas

Mercury concentrations in marine plankton have been determined in the Mediterranean Sea since 1960s to provide an insight in Hg contamination at the lowest marine trophic levels. Comparison of recent data for THg in zooplankton from the Adriatic, Mediterranean, Atlantic and Pacific shows inverse relations (Fig. 3A) with statistically significant differences among locations (Kruskal-Wallis, $P < 0.001$). Although the highest THg concentrations in seawater are found in the Adriatic Sea (KOTNIK *et al.,* 2015), it seems that specific conditions that influence Hg uptake in biota are considerably different. Copepods account on average for 77% of mesozooplankton from the Adriatic and Mediterranean Sea, while long-lived copepods and euphausiids are the major zooplankton genera in the Atlantic Ocean (HAMMERSCHMIDT *et al.,* 2013). Even though the composition of zooplankton community is similar, the observed differences in THg concentrations in zooplankton from different seas (Fig. 3A) are probably due to different

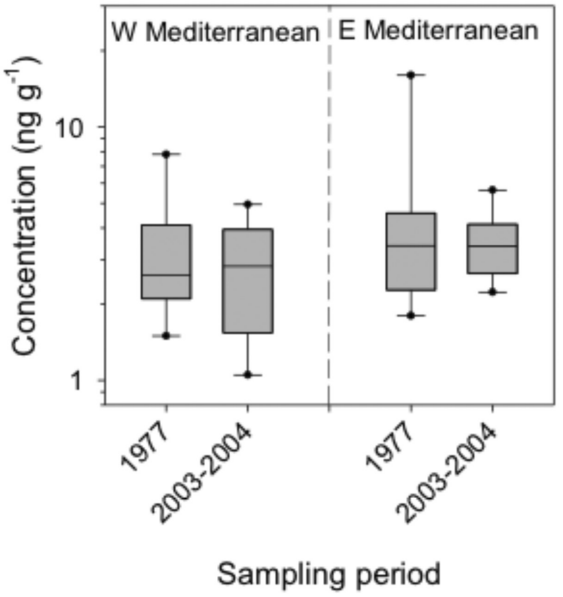

Fig. 4. Box-and-whiskers plots of the geographical distribution of THg in zooplankton from the Mediterranean Sea. Data from FOWLER, 1986; and M. Horvat (unpublished data).

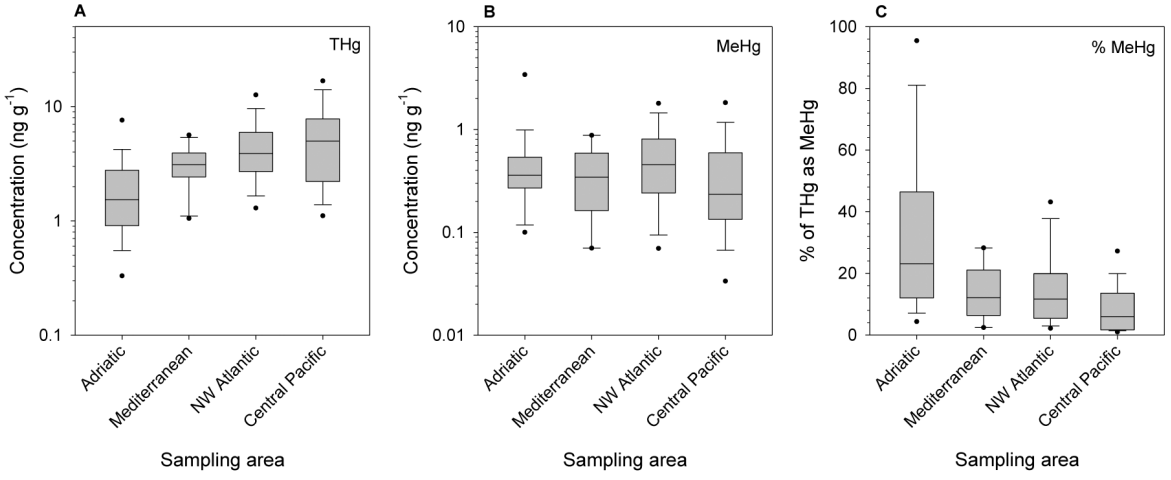

Fig. 3. Comparison of recent THg and MeHg concentrations and percentage of THg as MeHg in zooplankton from different world seas. Data from GOSNELL & MASON, 2015; GOSNELL et al., 2017; HAMMERSCHMIDT et al., 2013; and M. Horvat (unpublished data).

sampling methods and/or sampling periods. Another explanation might be significant difference in zooplankton water content, which causes error during transformation of concentrations expressed on a dry weight basis.

For MeHg, there are no significant differences between the Adriatic, Mediterranean and Atlantic zooplankton (Kruskal-Wallis, $P = 0.335$). Lower MeHg concentrations are found in the Pacific Ocean (Fig. 3B). Due to lower THg content, percentage of MeHg in zooplankton is the highest in the Adriatic Sea (33.1% on average; Fig. 3C).

In the Mediterranean Sea, the western sub-basin has slightly lower THg content in zooplankton than the eastern sub-basin (2.85 and 3.51 ng g^{-1}, respectively). It seems that these concentrations have not changed in the past 30 years (Fig. 4) indicating that localised Hg inputs do not have a considerable influence on THg concentrations in zooplankton in the open Mediterranean Sea as they do in the small enclosed basin such as Adriatic Sea (Fig. 5). Mercury speciation is rarely performed in zooplankton. Average MeHg concentrations in zooplankton are 0.25 and 0.28 ng g^{-1} in the western and eastern Mediterranean, which account for 9.16 and 9.68% of THg in Mediterranean zooplankton, respectively (M. Horvat, unpublished data).

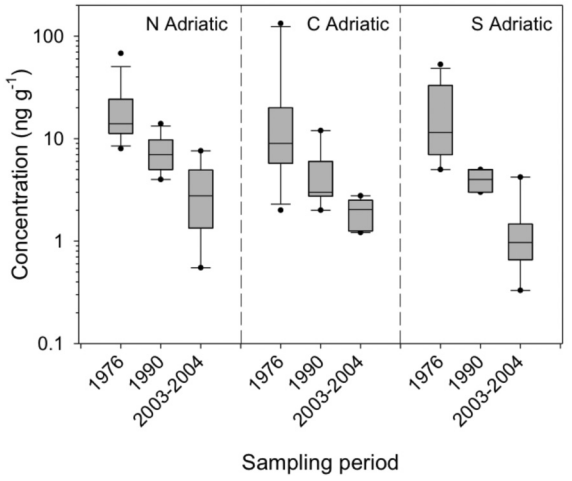

Fig. 5. Box-and-whiskers plots of the geographical distribution of THg in zooplankton from the Adriatic Sea. Data from FERRARA & MASERTI, 1992; KOSTA et al., 1978; and M. Horvat (unpublished data).

Mercury in Adriatic zooplankton

Changes in THg concentrations with time in the Adriatic zooplankton are significant (Fig. 5) (Kruskal-Wallis, $P < 0.01$ for all three sub-basins), contrary to trend observed in the Mediterranean Sea. In the Adriatic Sea, decrease in localised Hg input (mainly due to closure of Idrija mine and chlor-alkali plants in the northern Adriatic) is followed by up to 10-fold decrease in zooplankton THg. The highest natural THg concentrations (up to 53 ng g^{-1}) are found in the Gulf of Trieste, which have been affected by the polluted Soča/Isonzo River and its centuries-long draining of cinnabar-rich deposits from the Idrija mine (FAGANELI *et al.,* 2003). Temporal variability of zooplankton Hg is the consequence of its biomass, species composition and grazing. The speed of this variability is observed through a 25-fold decrease during a nine month period (HORVAT *et al.,* 1999). The highest THg concentrations in zooplankton are related to industrial mercury sources; VUČETIĆ *et al*. (1974) measured up to 1.63 μg g^{-1} of THg in zooplankton at the nearshore station in the central Adriatic Sea, and attributed this value to industrial pollution originating near Split. As in the Mediterranean Sea, MeHg measurements in the Adriatic zooplankton are scarce. In the Gulf of Trieste, MeHg ranges from 0.21-6.57 ng g^{-1}, with an average of 2.16 ng g^{-1} (HORVAT *et al.,* 1999) which represents 12.4% of THg in zooplankton. In the open Adriatic waters, MeHg in zooplankton shows a north-south concentration gradient (0.40, 0.41 and 0.80 ng g^{-1} in the northern, central and southern Adriatic, respectively; M. Horvat, unpublished data). The highest values coincide with the area of the highest MeHg in the Adriatic seawater (KOTNIK *et al.,* 2015), which indicates possible methylation of inorganic Hg by Adriatic microorganisms.

MERCURY IN BIVALVES

Mercury in mussels from different seas

The MEDPOL Programme recommended use of Mediterranean mussels as biomonitors of chemical contaminants in the Mediterranean Sea

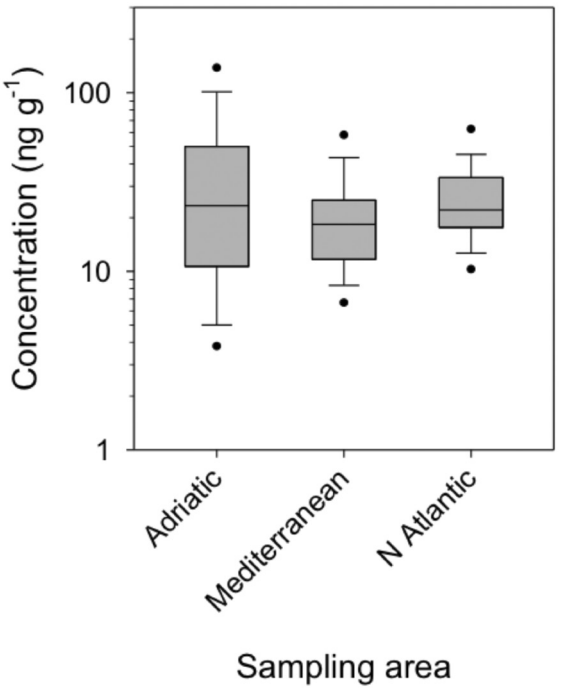

Fig. 6. Comparison of THg concentrations in mussels (Mytilus galloprovincialis) from different world seas. Data from: BENEDICTO et al., 2011; BILANDŽIĆ et al., 2016; BRIANT et al., 2017; CASTELLANI et al., 2015; CHEN et al., 2009; CLAISSE et al., 2001; CUBADDA et al., 2006; CULLAJ et al., 2006; FABBRI et al., 2006; FATTORINI et al., 2008; GALGANI et al., 2014; GORBI et al., 2008; KLJAKOVIĆ-GAŠPIĆ et al., 2006; KWON et al., 2014; LOCATELLI, 2003; LOCATELLI & MELUCCI, 2010; MARKOVIĆ et al., 2012; MOSCHINO et al., 2016; MZOUGHI & CHOUBA, 2012; SPADA et al., 2013; STANKOVIĆ et al., 2011; TSANGARIS et al., 2013.

(BENEDICTO *et al.,* 2011). Since this project was conducted in 1970s, several other projects (e.g. MYTILOS and MYTIOR) have been established to assess the state of trace metal contamination in the marine environment (ANDRAL *et al.,* 2011; GALGANI *et al.,* 2014). Recent data show that THg values in mussels are slightly, but significantly lower (Kruskal-Wallis, $P < 0.01$) in the Mediterranean Sea than in the Adriatic Sea and Atlantic Ocean (Fig. 6). The Adriatic THg values are obtained from both low-Hg and contaminated locations (BILANDŽIĆ *et al.,* 2016; KLJAKOVIĆ-GAŠPIĆ *et al.,* 2006) resulting in a wider box-and-whiskers plot. In the Mediterranean Sea, THg concentrations are characterised by approximately 10-fold decrease in 30 years period (Fig. 7). On average, THg in mussels from the western Mediterranean is higher than in the eastern sub-basin (23.0±19.2 and 17.9±11.5 ng g^{-1}, respectively), but without statistically significant difference (Mann-Whitney, $P = 0.124$). Mercury speciation data in the Mediterranean mussels are scarce; therefore, only THg concentrations are available as a proxy for environmental contamination. The most comprehensive Hg speciation in mussels from the western Mediterranean (French coast) was recently published by BRIANT *et al.* (2017). MeHg concentrations in these samples range from 3.00-28.7 ng g^{-1} (average 10.5 ng g^{-1}) and account for 11.4-57.0% of THg (average 30.3%). Almost the same percentage (29.0%) is found in the Ionian Sea (SPADA *et al.,* 2012). A similar mussel species (Blue mussel, *Mytilus edulis*) from the NW Atlantic, has an average MeHg concentration of 20.6 ng g^{-1} which accounts for 37.4% of THg (CHEN *et al.,* 2009). Slightly higher percentage (40.8%) is found in the same species from the French

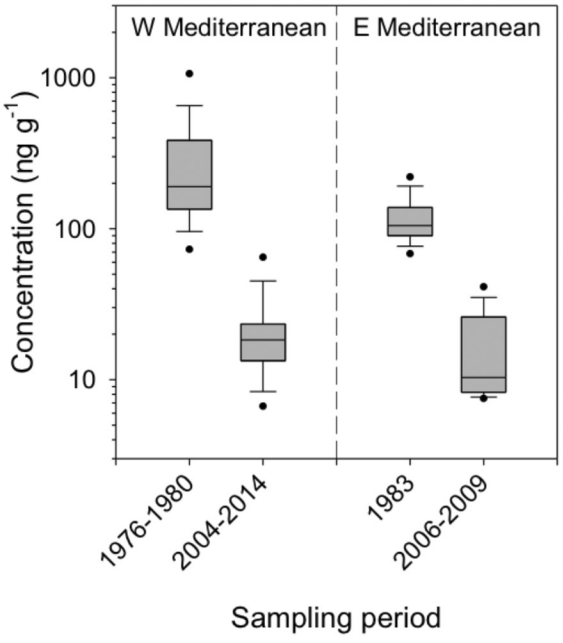

Fig. 7. Box-and-whiskers plots of the geographical distribution of THg in mussels (Mytilus galloprovincialis) from the Mediterranean Sea. Data from BENEDICTO et al., 2011; BRIANT et al., 2017; CASTAGNA et al., 1985; GALGANI et al., 2014; LEONZIO et al., 1981; MZOUGHI & CHOUBA, 2012; TSANGARIS et al., 2013.

Atlantic coast, but with almost 50% lower average MeHg concentration (11.4 ng g^{-1}) (BRIANT *et al.,* 2017). Bioaccumulation of MeHg in mussels can be observed through the 3-fold higher percentage of MeHg compared to Mediterranean zooplankton. However, filter-feeding bivalves accumulate less MeHg than fish and present a lower risk to humans (STANKOVIĆ *et al.,* 2012).

Mercury in Adriatic mussels

In the Adriatic Sea, relative changes in median THg concentrations with time (Fig. 8) are not as great as in the Mediterranean Sea. In the northern Adriatic Sea, an approximately 3-fold decrease in median mussel THg can be observed. This is probably due to decrease in environmental concentrations caused by closure of Idrija mine and chlor-alkali plants in the northern Adriatic (FAGANELI *et al.,* 2014). Interesting to note is the increase in mussel THg in the southern Adriatic, but this might be due to limited data prior to 1990s. When low-Hg environment and contaminated locations are separately compared (Fig. 9A), statistically significant decreases in mussel THg values with time are observed in both environments (Kruskal-Wallis, $P < 0.001$ for Kaštela Bay; Mann-Whitney, $0.002 < P < 0.05$ for Krka River estuary, Venice Lagoon and Ravenna Lagoon). Kaštela Bay, which has been contaminated by industrial effluents from a chlor-alkali plant (KLJAKOVIĆ-GAŠPIĆ *et al.,* 2010), has a 70-fold decrease in average mussel THg after plant closure in 1990. On the contrary, in the low-Hg environment of the Krka River estuary (Fig. 1), only a slight decrease can be observed (~26% during five years). Although water currents along the eastern Adriatic flow northwards (Fig. 1), it seems that anthropogenic Hg contamination is localised at its source. This effect is best observed in the shallow northern Adriatic Sea where counter-clockwise water currents carry cinnabar-rich particles from the Soča/Isonzo River delta to the adjacent Marano and Grado Lagoon. The average THg concentration in mussels from this lagoon (220±60 ng g^{-1}; MAJORI *et al*., 1991) was higher than in other parts of the northern Adriatic Sea (Fig. 8). The uncontrolled Hg discharges from the Torviscosa

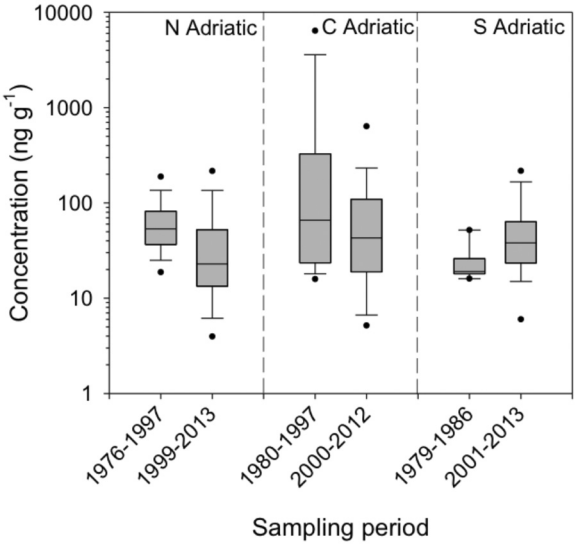

Fig. 8. Box-and-whiskers plots of the geographical distribution of THg in mussels (Mytilus galloprovincialis) from the Adriatic Sea. Data from BILANDŽIĆ et al., 2016; BLOOM et al., 2004; BOGDANOVIĆ et al., 2014; CASTELLANI et al., 2015; CATTANI et al., 1999; CORSI et al., 2011; CUBADDA et al., 2006; CULLAJ et al., 2006; DA ROS et al., 2000; DOMINIK et al., 2014; FABBRI et al., 2006; FATTORINI et al., 2008; FAVRETTO et al., 1997; GIORDANO et al., 1991; GORBI et al., 2008; JOVIĆ & STANKOVIĆ, 2014; KLJAKOVIĆ-GAŠPIĆ et al., 2006; KOSTA et al., 1978; LOCATELLI, 2003; LOCATELLI & MELUCCI, 2010; LOWE et al., 1995; MAJORI et al., 1991; MARKOVIĆ et al., 2012; MARTINČIĆ et al., 1987; MIEDICO et al., 2013; MIKAC et al., 1985, 1989, 1996; MOSCHINO et al., 2016; NAJDEK & SAPUNAR, 1987; NASCI et al., 1998; ODŽAK et al., 2000; RAMŠAK et al., 2012; SPADA et al., 2013; STANKOVIĆ et al., 2011; STORELLI & MARCOTRIGIANO, 2001; TURK et al., 2007; WIDDOWS et al., 1997; ZATTA et al., 1992; ZVONARIĆ, 1991.

chlor-alkali plant (1949-1984) were additional Hg sources in this lagoon (PIANI *et al*., 2005). Elemental Hg from chlor-alkali plant is the main sediment fraction in the Marano sector and sulphide-bound Hg from Soča/Isonzo River in the Grado sector (ACQUAVITA *et al*., 2012). Venice and Ravenna lagoons were also contaminated by Hg discharges from chlor-alkali and acetaldehyde plants in the Marghera industrial zone and Ravenna, respectively (BLOOM *et al*., 2004; COVELLI *et al*., 2011). Following the closure of these plants, average THg concentrations in mussels

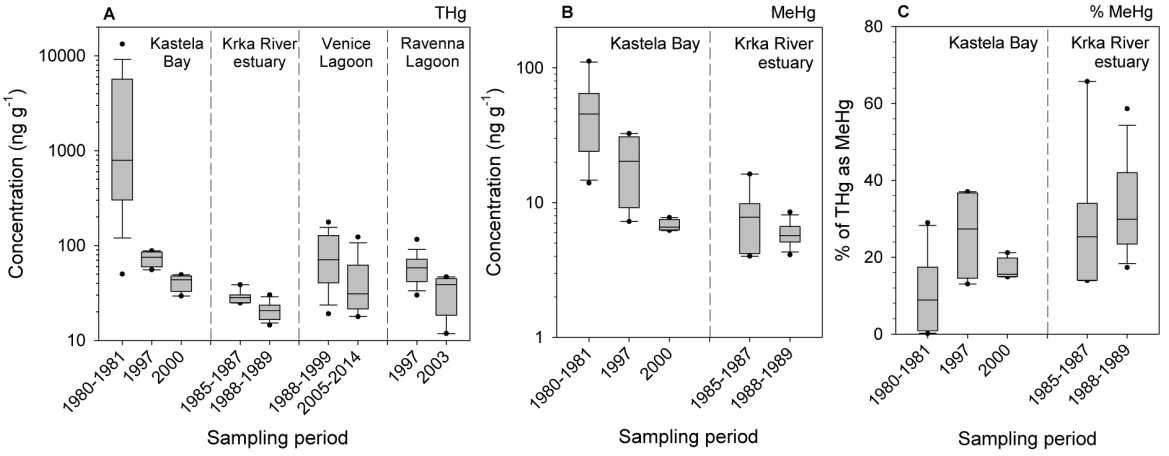

Fig. 9. Temporal changes of THg and MeHg concentrations and percentage of THg as MeHg in mussels from case studies in the Adriatic Sea. Data from BILLE et al., 2015; CASTELLANI et al., 2015; CATTANI et al., 1999; CUBADDA et al., 2006; DA ROS et al., 2000; DOMINIK et al., 2014; FABBRI et al., 2006; KLJAKOVIĆ-GAŠPIĆ et al., 2006; LOWE et al., 1995; MIKAC et al., 1985, 1989, 1996; NASCI et al., 1998; ODŽAK et al., 2000; WIDDOWS et al., 1997; ZATTA et al., 1992.

are characterised by an approximate 2-fold decrease (Fig. 9A). In addition, there is no statistically significant differences in THg concentrations in mussels after 2000 from Kaštela Bay, Venice and Ravenna Lagoons (Kruskal-Wallis, $P = 0.778$).

Mercury speciation data in mussels are scarce. However, some locations are more studied providing an insight in MeHg bioaccumulation in the Adriatic mussels. In both Kaštela Bay and Krka River estuary, we can observe a decrease in MeHg concentrations with time (Fig. 9B), but this decrease is not as large as for THg. The average MeHg values are similar (6.79 ng g^{-1} in Kaštela Bay in 2000, and 5.91 ng g^{-1} in Krka River estuary in 1988-1989), but are lower than observed in the western Mediterranean during 1987-2014 (10.5 ng g^{-1}; BRIANT *et al.,* 2017). Percentage of mercury as MeHg increases with time, to average values of 16.8 and 32.7% in Kaštela Bay and Krka River estuary, respectively (Fig. 9C). The percentage in the Krka River estuary is similar to that in the Mediterranean, and the low value in Kaštela Bay is in agreement with the observation that in this region conditions do not promote Hg methylation (KWOKAL *et al.,* 2002). In the Venice Lagoon, average MeHg concentration (52.2 ng g^{-1}) accounts for 57.4% of THg (DOMINIK *et al.,* 2014). Previous researches in this lagoon found lower percentage (37.9%), but exceptionally higher average MeHg concentration (138 ng g^{-1}) (BLOOM *et al.,* 2004).

Mercury speciation in tissues from Adriatic mussels

Only a few papers describe Hg speciation in mussel organs and tissues. In Kaštela Bay, average THg concentrations are the highest in the digestive gland (189 ng g^{-1}), followed by gills (109 ng g^{-1}), while the whole soft tissue has the lowest THg concentrations (70.6 ng g^{-1}) (ODŽAK *et al.,* 2000). In the Venice Lagoon, average THg concentration in digestive gland is much lower (105 ng g^{-1}) (LOWE & FOSSATO, 2000). THg values in digestive gland are approximately four times higher than in the gills in mussels from the Gulf of Trieste (ŽNIDARIČ *et al.,* 2006). Contrary to THg, MeHg has a different distribution in tissues: the highest average values are found in the whole soft tissue (20.0 ng g^{-1}), while gills and digestive gland have similar MeHg values (11.7 ng g^{-1}). KLJAKOVIĆ-GAŠPIĆ *et al.* (2006) found similar THg distribution, but almost uniform MeHg concentrations in all tissues. The lowest percentage of THg as MeHg was found in the digestive gland (8.31%), compared to whole soft tissue (21.3%) (KLJAKOVIĆ-GAŠPIĆ *et al.,* 2006).

Mercury speciation in tissues indicates different distribution pathways in the mussels. It appears that MeHg is transported to other tissues after uptake through digestive gland and gills (ODŽAK *et al.,* 2000), while inorganic Hg is accumulated in the digestive gland where it is probably transformed to some less toxic form before its excretion or storage (KLJAKOVIĆ-GAŠPIĆ *et al.,* 2006).

Mercury in other bivalves

Although mussels are usually used as bioindicators, THg concentrations in other bivalves can also be used to assess Hg contamination and distribution in the Adriatic Sea. Table 1 shows THg concentrations in whole shellfish tissue of Adriatic bivalves from recent studies. A decreasing southward THg gradient can be observed in European flat oyster (*Ostrea edulis*) and Manila clam (*Ruditapes philippinarum*) (Table 1). Observed average THg concentrations in these bivalves generally follow THg gradient in sediments and seawater, which is a consequence of either natural or anthropogenic Hg contamination (ŽIVKOVIĆ *et al*., 2017).

Table 1. Total mercury concentrations (ng g^{-1} wet weight) in Adriatic bivalves.

Bivalve specie	Location	Range	Average	Reference
Callista chione	Veneto region	2.00-60.0	30.0	BILLE *et al.,* 2015
Cerastoderma edule	Veneto region	2.00-230	60.0	BILLE *et al.,* 2015
Cerastoderma glaucum	Venice Lagoon		166	DOMINIK *et al.,* 2014
Chamelea gallina	Apulia	6.00-25.0	17.0	MIEDICO *et al.,* 2013
Chamelea gallina	Veneto region	2.00-250	30.0	BILLE *et al.,* 2015
Mimachlamys varia	Istria/Gulf of Trieste	26.0-512	252	BOGDANOVIĆ *et al.,* 2014
Mimachlamys varia	Veneto region	2.00-120	20.0	BILLE *et al.,* 2015
Crassostrea gigas	Veneto region	2.00-120	60.0	BILLE *et al.,* 2015
Crassostrea gigas	Venice Lagoon		105	DOMINIK *et al.,* 2014
Crassostrea gigas	northern Adriatic	10.0-340	92.5	BURIOLI *et al*., 2017
Donax trunculus	Veneto region	2.00-80.0	30.0	BILLE *et al.,* 2015
Ensis minor	Veneto region	10.0-130	60.0	BILLE *et al.,* 2015
Mactra stultorum	Veneto region	10.0-60.0	30.0	BILLE *et al.,* 2015
Ostrea edulis	Apulia	6.00-64.0	15.3	MIEDICO *et al.,* 2013
Ostrea edulis	Istria	30.0-100	60.0	BILANDŽIĆ *et al.,* 2015
Ostrea edulis	northern Adriatic	15.0-109	60.0	BILANDŽIĆ *et al.,* 2016
Ostrea edulis	southern Adriatic	13.0-70.0	39.0	BILANDŽIĆ *et al.,* 2016
Ostrea edulis	Veneto region	30.0-60.0	40.0	BILLE *et al.,* 2015
Pecten jacobaeus	Istria	10.0-30.0	20.0	BILANDŽIĆ *et al.,* 2015
Pecten jacobaeus	Veneto region	2.00-30.0	10.0	BILLE *et al.,* 2015
Ruditapes decussatus	Veneto region	20.0-190	70.0	BILLE *et al.,* 2015
Ruditapes philippinarum	Marano Grado Lagoon	54.0-985	308	GIANI *et al.,* 2012
Ruditapes philippinarum	Marano Grado Lagoon	225-338	272	SFRISO *et al*., 2008
Ruditapes philippinarum	Ravenna Lagoon	42.0-92.0	77.4	TROMBINI *et al.,* 2003
Ruditapes philippinarum	Veneto region	2.00-190	40.0	BILLE *et al.,* 2015
Ruditapes philippinarum	Venice Lagoon		121	DOMINIK *et al.,* 2014
Ruditapes philippinarum	Venice Lagoon	30.0-70.0	52.2	SFRISO *et al*., 2008
Venus verrucosa	Istria	0.00-50.0	20.0	BILANDŽIĆ *et al.,* 2015
Venus verrucosa	Kaštela Bay	49.0-378	169	BOGDANOVIĆ *et al.,* 2014
Venus verrucosa	Veneto region	40.0-80.0	60.0	BILLE *et al.,* 2015

Therefore, these organisms could also be used as environmental bioindicators of Hg contamination. Besides ambient THg concentrations, differences in bivalve THg also arise due to biological diversity between the species. For example, European flat oyster in the Adriatic has 2-3 times higher average THg concentrations than mussels (BILANDŽIĆ *et al.,* 2015, 2016). This could be due to a larger gill area and two-fold greater filtration rate in *Ostrea* sp. than in *Mytilus* sp. (WALNE, 1972).

Mercury speciation data in the other Adriatic bivalves are limited. However, these data show that MeHg accounts for approximately 40-50% of THg in *Cerastoderma glaucum*, *Ruditapes philippinarum* and *Crassostrea gigas* (DOMINIK *et al.,* 2014; GIANI *et al.,* 2012). These values are higher than in mussels, showing that biological parameters, alongside environmental ones, have significant influences on Hg bioaccumulation in bivalves. The highest average percentage of MeHg (81.2%) and the highest average of MeHg concentrations (85.7 ng g^{-1}) are found in *Ruditapes philippinarum* from Ravenna Lagoon (TROMBINI *et al.,* 2003).

Importance of coastal lagoons in mercury transformations and bioaccumulation

Shallow coastal lagoons are particularly complex environments, often subjected to strong anthropogenic impacts. Lagoon environments are often ideal for mariculture due to their high productivity and their shallow and protected surroundings (COVELLI *et al*., 2008). However, these coastal lagoons accumulate labile organic matter in anoxic sediments that drives microbial processes responsible for rapid Hg methylation (HINES *et al*., 2012). In addition, the redox potential has values that are usually more negative along the coast due to reduced hydrodynamics (BRAMBATI, 2001). For example, Venice lagoon is a shallow lagoon bordered by highly productive intertidal wetlands, contains high nutrient loading from agricultural runoff and urban sewage, and therefore represents an ideal site for Hg methylation (BLOOM *et al*., 2004). The water flow through the Venice lagoon represents the net source of THg and MeHg to the open Adriatic Sea (BLOOM *et al*., 2004), which could be later accumulated by the Adriatic biota.

In addition to bacterial methylation processes in the sediments, MeHg transfer to organisms in the Marano and Grado Lagoon depends on intake of already enriched organic substances (micro and macro epiphytes, bits of plant tissue, etc.) originating from the lagoon and/or the Isonzo River (BRAMBATI, 2001). The abundance of plant and animal species in lagoons increases the risk of Hg remobilization from sediments (by bioturbation) to the water column and subsequent bioaccumulation (COVELLI *et al*., 2008), especially in benthic species like bivalves. The increased bioaccumulation is observed through the higher THg values in mussels from the Venice Lagoon and the Marano and Grado Lagoon, compared to those from low-Hg environment of the Krka River estuary (Fig. 9A). In addition, high THg and MeHg values in clams from the northern Adriatic lagoons (e.g. *Ruditapes philippinarum*) could be due to high bioaccumulation and methylation potentials in these ecosystems.

Role of metallothioneins in mussels in mercury detoxification

The synthesis of cysteine-rich metallothioneins (MTs) is induced in mussels when exposed to certain metals, including Hg. They are biomarkers of response to metal contamination and have important roles in the regulation, storage and detoxification of metals in mussels (ŽNIDARIČ *et al.,* 2006). Concentrations of MTs in mussels from the Hg contaminated Vlora Bay (Fig. 1) are up to 50% higher than in mussels from the background station (CULLAJ *et al*., 2006). Field study in the Gulf of Trieste showed that MTs and metal content in mussels are not well correlated (RAMŠAK *et al.,* 2012). However, other biological and environmental factors may influence MTs concentrations. Although Hg is not a strong inducer of MTs synthesis, it has a strong binding affinity to MTs and can displace other metals bound to MTs (RAMŠAK *et al.,* 2012). ŽNIDARIČ *et al.* (2006) showed that Hg binds to MTs in gills and digestive glands in mussels exposed to inorganic Hg, but only in digestive glands in wild mussels from industrial site. Hg

binding to MTs in digestive gland could explain the observed higher THg values in digestive gland than in other tissues.

MERCURY IN FISH

Comparison of mercury in fish from different seas

In the 1970s, THg concentrations in Mediterranean fish were higher than in the same fish species caught in the adjacent Atlantic Ocean (BERNHARD & RENZONI, 1977). Although this trend can also be observed today by comparing literature data, only few recent studies have simultaneously measured THg concentrations in both basins. Average THg concentrations in European hake are up to 5.5 times higher in the Mediterranean Sea than in the Atlantic Ocean (Table 2). These observations raise questions about specificity of Mediterranean mercury biogeochemistry, but definite conclusions have not yet been found.

Due to their ubiquitous distribution in the Mediterranean, red mullets (*Mullus barbatus*

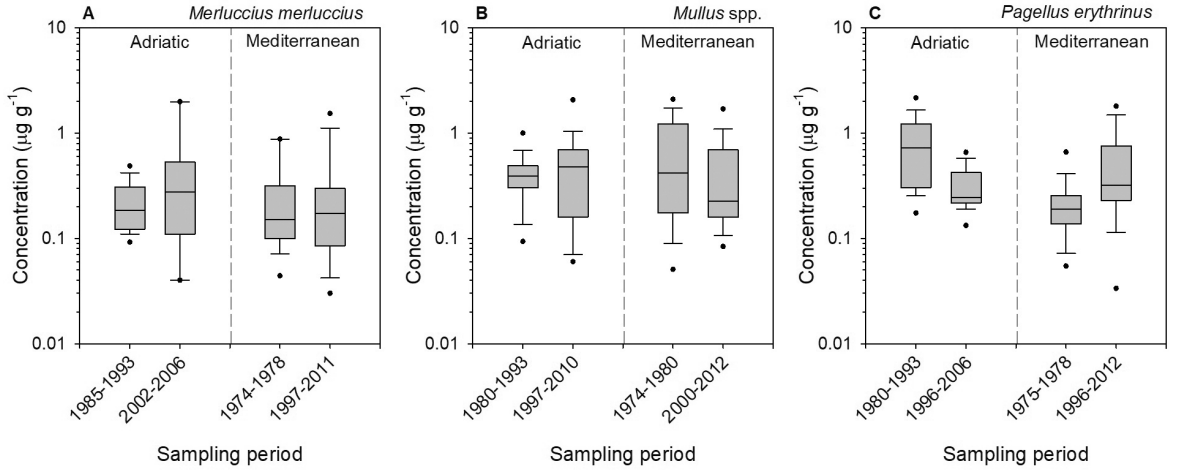

Fig. 10. Box-and-whiskers plots of the geographical distribution of THg in the three most studied fish species (A – Merluccius merluccius, B – Mullus spp., C – Pagellus erythrinus) from the Adriatic and Mediterranean Sea. Data obtained from BERNHARD & RENZONI, 1977; BILANDŽIĆ et al., 2011; BONSIGNORE et al., 2013; BRAMBILLA et al., 2013; BUZINA et al., 1989, 1995; COPAT et al., 2012; COSSA et al., 2012; ČULIN & ZVONARIĆ, 1995; DURAL et al., 2010; FALCÓ et al., 2006; HARAKEH et al., 1985; HARMELIN-VIVIEN et al., 2009, 2012; HORNUNG et al., 1980; HORVAT et al., 1999; JUREŠA & BLANUŠA, 2003; KONTAS, 2006; LAHAYE et al., 2006; LEONZIO et al., 1981; MIKAC et al., 1985; MINIERO et al., 2014; NAUEN et al., 1980; PERUGINI et al., 2009; STORELLI, 2008; STORELLI & MARCOTRIGIANO, 2000, 2005; STORELLI et al., 2003, 2005a, 2007; STORELLI & BARONE, 2013; TUNÇEL et al., 1980; ULUTURHAN & KUCUKSEZGIN, 2007; VUKADIN et al., 1995; YANNAI & SACHS, 1978; ZVONARIĆ, 1991.

Table 2. Comparison of THg concentrations expressed as µg g^{-1} wet weight in various fish species from the Mediterranean Sea and the Atlantic Ocean.

Fish specie	Mediterranean		Atlantic		Reference
	Range	Average	Range	Average	
Merluccius merluccius	0.034-0.058	0.045	0.018-0.046	0.033	LAHAYE *et al.,* 2006
Merluccius merluccius	0.022-2.862	0.334	0.008-0.132	0.060	COSSA *et al.,* 2012
Sardina pilchardus	0.067-0.127	0.094	0.019-0.034	0.023	LAHAYE *et al.,* 2006
Engraulis encrasicolus	0.043-0.061	0.050	0.026-0.032	0.028	LAHAYE *et al.,* 2006
Micromesistius poutassou	0.020-0.023	0.021	0.016-0.017	0.016	LAHAYE *et al.,* 2006
Xiphias gladius		1.678		0.773	DAMIANO *et al.,* 2011

and *Mullus surmuletus*) are probably the most analysed fish species, beside the commercially important species European hake (*Merluccius merluccius*) and common pandora (*Pagellus erythrinus*). Fig. 10 shows the comparison of THg in these fish species from the Adriatic and Mediterranean Seas through time. Recent data shows no statistically significant differences in fish THg concentrations from the two seas (Mann-Whitney, $P = 0.493$ for European hake and $P = 0.430$ for red mullets). In addition, there are no significant changes in THg concentrations in Adriatic fish with time (Mann-Whitney, $P = 0.358$ for European hake and $P = 0.565$ for red mullets). For common pandora, there is a significant decrease in Adriatic THg with time and significant difference between concentrations in two basins during past measurements (Mann-Whitney, $P < 0.001$). Specific reasons for these observations in common pandora are unknown, especially when taking into account the similar habitats, eating habits and trophic position of red mullets and common pandora in food web. However, recent results (Fig. 10C) show no statistically significant difference between THg concentrations in common pandora from the Adriatic and Mediterranean Seas (Mann-Whitney, $P = 0.167$).

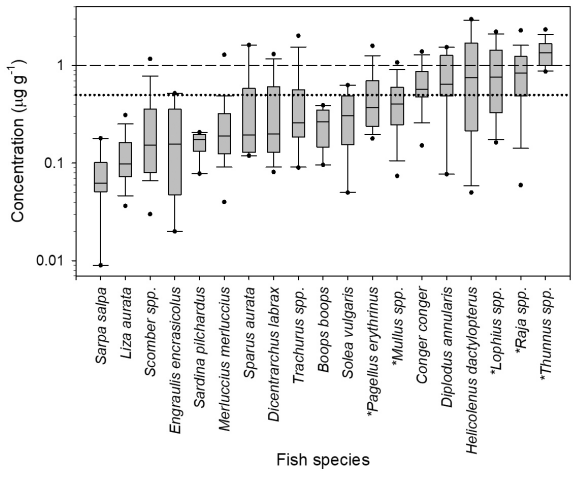

Fig. 11. Box-and-whiskers plots of THg concentrations in the most studied fish species from the Adriatic Sea. Dotted and dashed lines correspond to the EU maximal allowed THg levels of 0.5 and 1.0 µg g^{-1} (on graph), respectively. Data obtained from: BRAMBATI, 2001; BILANDŽIĆ et al., 2011; BUZINA et al., 1989, 1995; ČULIN & ZVONARIĆ, 1995; DELLA TORRE et al., 2010; DOMINIK et al., 2014; HORVAT et al., 1999, 2014; JUREŠA & BLANUŠA, 2003; MIKAC et al., 1985; NAJDEK & BAŽULIĆ, 1983; PERUGINI et al., 2009; STORELLI, 2008; STORELLI & BARONE, 2013; STORELLI & MARCOTRIGIANO, 2000, 2004; STORELLI et al., 1998, 2002b, 2003, 2005a, 2006, 2007; VUKADIN et al., 1995; ZVONARIĆ, 1991; ŽVAB ROŽIČ et al., 2014.*

Total mercury in fish

Mercury content in the Adriatic fish was determined to observe effects of industrial effluents (from chlor-alkali plants) and elevated natural seawater levels (from the Idrija River) to marine biota. THg concentrations in Adriatic fish span several orders of magnitude. THg in Adriatic fish for which box-and-whisker graphs could be constructed from literature data, are presented in Fig. 11. This figure emphasises the importance of diet for mercury biomagnification; Hg in fish generally increases with increasing trophic position in food web of its prey (COSSA *et al.,* 2012; HORVAT *et al.,* 2014). The lowest THg concentrations are found in demersal herbivorous fish like salema porgy (*Sarpa salpa*) (STORELLI *et al*., 1998), followed by planktivorous fish (e.g. European anchovy, *Engraulis encrasicolus*) (BILANDŽIĆ *et al.,* 2011), omnivorous (e.g. bogue, *Boops boops*) and carnivorous fish (e.g. annular seabream, *Diplodus annularis*) (BUZINA *et al.,* 1989). The highest THg values are found in top predator species (HORVAT *et al.,* 2014). For example, STORELLI *et al.* (2002a) measured 18.3 µg g^{-1} in hammerhead (*Sphyrna zygaena*) from the Ionian Sea. Exceptionally high average THg concentrations were also determined in gulper shark (*Centrophorus granulosus*) from the Adriatic Sea (9.66 µg g^{-1}) (STORELLI *et al.,* 2002a). Average THg concentrations higher than 2.0 µg g^{-1} in the top predator species can be found in ghostshark (*Chimaera monstrosa*), blackmouth catshark (*Galeus melastomus*), Mediterranean moray (*Muraena helena*), electric ray (*Torpedo nobiliana*) and European barracuda (*Sphyraena sphyraena*). In addition, high THg values can be found in European seabass (*Dicentrarchus labrax*) from the Artalina and Francamela

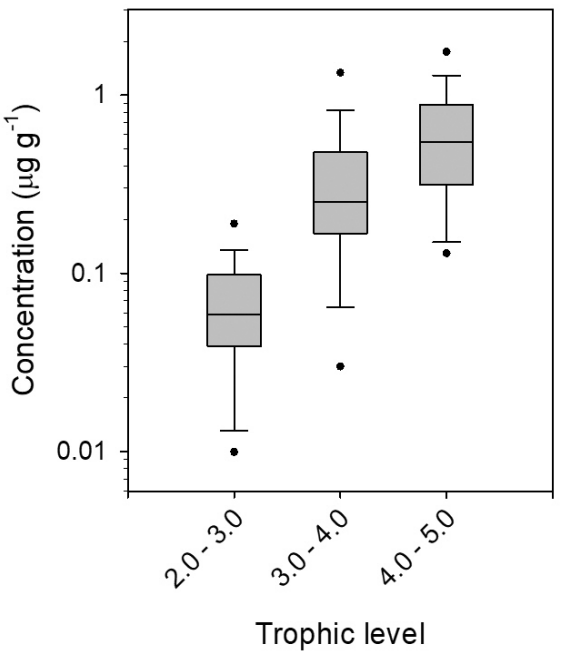

Fig. 12. Box-and-whisker plots of MeHg concentrations in 41 Adriatic fish species sorted in three trophic level ranges. Data obtained from: BRAMBATI, 2001; BUZINA et al., 1989, 1995; ČULIN & ZVONARIĆ, 1995; DOMINIK et al., 2014; HORVAT et al., 1999, 2014; MIKAC & PICER, 1985; NAJDEK & BAŽULIĆ, 1983; STORELLI et al., 2002a, 2002b, 2002c, 2003, 2005a, 2005b; VUKADIN et al., 1995; ZVONARIĆ, 1991.

fish farms (Grado Lagoon). These THg values indicate that mercury is readily biomagnified through the Mediterranean and Adriatic trophic food webs.

Methylmercury in fish

Methylmercury concentrations in the Adriatic fish are not as extensively measured as THg. MeHg concentrations were summarised from available papers by trophic position of fish species in food web (Fig. 12). Trophic level data were obtained from BRAMBILLA *et al.* (2013) or from internet database (www.fishbase.org). These values differ at most by 0.3 trophic level from the values calculated by DOMINIK *et al.* (2014) based on $\delta^{15}N$ measurements in the Venice Lagoon, and can therefore be used approximately as Adriatic trophic levels. From MeHg concentrations in 41 Adriatic fish species sorted in three trophic level ranges, it is evident that the lowest and highest MeHg concentrations are found at the lowest and highest trophic levels, respectively (Fig. 12). Differences in MeHg concentrations between trophic levels are statistically significant (Kruskal-Wallis, $P < 0.001$). MeHg biomagnifies in organisms, as indicated by its concentration increase with each subsequent trophic level (COSSA *et al.,* 2012; HORVAT *et al.,* 2014). MeHg can account for up to 100% of THg in the Adriatic fish. In the Gulf of Trieste, the percentage of THg present as MeHg increases from 57-77 % in small benthic fish to 38-71% in small pelagic fish and 90-100% in larger benthic fish to 100% in larger pelagic fish (FAGANELI *et al.,* 2014; HORVAT *et al.,* 2014). However, these percentages depend on specific biological and environmental factors. For example, MeHg percentage in fish species from Kaštela Bay was on average 30% lower than in the same species from the Island of Vis (BUZINA *et al.,* 1995). It seems that Hg originating from the chlor-alkali plant in Kaštela Bay is not readily available for methylation. Specific environmental conditions drive transformation of inorganic Hg to MeHg in the Adriatic sediment (BRATKIČ *et al.,* 2017) and water column (KOTNIK *et al.,* 2015), and therefore result in different MeHg content in fish.

Influence of environmental and biological parameters on mercury concentrations

Besides trophic position of the fish in food web, higher Hg concentrations have also been related to the living environment. Demersal fish usually have comparatively higher THg levels than pelagic fish (STORELLI *et al.,* 1998). Furthermore, fish that feed on benthic organisms accumulate Hg to a higher degree that other species (STORELLI *et al.,* 2002a). This is probably due to mercury absorption from Hg-rich porewaters and solid phase by benthic algae, and its consequent transfer to the benthic fauna and food web (FAGANELI *et al.,* 2014). Another possibility is active Hg-release mechanism from sediments to the water column and its transfer to benthic biota (BONSIGNORE *et al.,* 2013). Depth is also an important factor influencing Hg concentrations in fish.

Deep-sea species usually have higher THg values than fish inhabiting shallow waters (CHOY *et al.,* 2009; KOENIG *et al.,* 2013). Values above 1.0 μg g^{-1} have been reported for various fish species from the western Mediterranean waters deeper than 1000 m, even for the small planktivorous Mediterranean spiderfish (*Bathypterois mediterraneus*) (KOENIG *et al.,* 2013). To explain these observations, CRESSON *et al.* (2014) have recently proposed that food quality and quantity decreases with depth, affecting organisms by slowing their growth. Deep-water fish would appear older and would be able to accumulate Hg during a longer time. Second, most MeHg in water column is produced in oxygen-depleted waters (COSSA *et al.,* 2009), and can be readily adsorbed on sinking organic particles which represent food for deep-water animals (CRESSON *et al.,* 2014).

In the Adriatic Sea, variation of Hg with fish length or weight can be observed, as well. Various authors have described linear and exponential correlations between THg (MeHg) in fish muscle and length (STORELLI *et al.,* 2007; STORELLI & BARONE, 2013; HORVAT *et al.,* 2014) or weight (STORELLI *et al.,* 2002a, 2006; HORVAT *et al.,* 2014). This shows that the highest Hg concentrations are found in large older fish, which are usually predators. The positive correlation between fish size (which is a proxy for fish age) and Hg concentrations suggests that consumers who eat larger fish might have higher exposure to mercury than those who eat smaller fish (STORELLI *et al.,* 2006, 2007). Frequent consumption of fish with high Hg content can exceed the maximum tolerable weekly intake (1.6 μg kg^{-1} body weight). They should therefore be eaten in moderation (BOSCH *et al.,* 2016; STORELLI & BARONE, 2013). In the European Union, maximum levels for Hg in foodstuffs are set to 0.5 μg g^{-1} for most fish species, and 1.0 μg g^{-1} for benthic fish and top predator species (Fig. 11) (EUROPEAN COMMISSION, 2006). Although most of the Adriatic fish species have some values above these limits, some fish species exceed this limit in about 75% cases, such as European conger (*Conger conger*), annular seabream (*Diplodus annularis*) and tuna (*Thunnus* spp.). Although MeHg represents the greatest part of THg, EU legislation does not set maximum MeHg levels in fish.

Mercury in different tissues of Adriatic fish

Mercury concentrations are usually determined only in fish muscle used for human consumption. However, Hg content in other fish tissues shows variability, similarly as in mussels. Biomagnification studies should measure Hg concentrations in different organs and tissues of fish body, and not only in muscle, due to different concentrations in various organs (HAMMERSCHMIDT & FITZGERALD, 2006). In rays from the Gulf of Trieste HORVAT *et al.* (2014) found the highest THg and MeHg in muscle tissue followed by gills and liver. It seems that high values in the gills suggest the capability to accumulate high quantities of Hg from water, especially in inorganic form, as seen by the lowest percentage of THg present as MeHg. Negative correlation between the percentage of MeHg in the liver and weight of bull ray (*Pteromylaeus bovinus*) indicates a demethylation processes in the liver of older specimens. Demethylated MeHg in the liver shows weak mobility, probably due to formation of weakly soluble salts of inorganic Hg, such as mercury(II) selenide (HORVAT *et al.,* 2014). Contrary to rays, albacore (*Thunnus alalunga*) and European eel (*Anguilla anguilla*) from the Adriatic Sea have up to 2.5-fold and 6-fold higher THg concentrations in liver than in muscle, respectively (RENZI *et al.,* 2012; STORELLI & MARCOTRIGIANO, 2004). Different bioaccumulation in various tissues among species is due to their feeding behaviour (HORVAT *et al.,* 2014), while different accumulation in the liver is probably due to different metabolic activities (STORELLI & MARCOTRIGIANO, 2004).

Role of selenium in fish on mercury detoxification

Selenium can increase Hg elimination from organisms, and growing evidence suggests Se-mediated mercury toxicokinetics (GRIBBLE *et al.,* 2016). Several studies measured Se:Hg molar ratios in various fish species from the Adriatic Sea (MIKAC *et al.,* 1985; PERUGINI *et al.,* 2014;

STORELLI & MARCOTRIGIANO, 2002). This ratio is usually higher than 1.0 in Adriatic fish, and the highest ratios (>20) are found in smaller fish (PERUGINI *et al.,* 2014). Equimolar Se:Hg ratios, which can be found in marine animals with high THg concentrations (above 1 μg g^{-1}) (STORELLI & MARCOTRIGIANO, 2002), represent a threshold associated with Hg toxicity. Generally, Se:Hg molar ratios show stronger correlation with Hg concentrations than with Se concentrations, which means that Hg has greater biomagnification potential than Se (GRIBBLE *et al.,* 2016). Metallothioneins are rarely significantly correlated with Hg in the Mediterranean fish (SISCAR *et al.,* 2014), indicating that they are not as important as Se regarding Hg detoxification. Generally, glutathiones represent the primary defence against Hg toxicity, followed by metallothionein induction, and as the most important formation of Hg-Se-protein complexes (CUVIN-ARALAR & FURNESS, 1991; HORVAT *et al.,* 2014).

CONCLUSIONS

Complexity of biogeochemical Hg cycle in the Adriatic Sea is observed through variability of mercury concentrations along trophic levels. Although the highest Hg concentrations are found in the Adriatic Sea, THg in zooplankton does not appear to follow this pattern and has significantly lower values in Adriatic than in the Mediterranean and other world seas. However, methylation potential in the Adriatic Sea appears to be the highest, as observed from the highest percentage of THg present as MeHg. THg in zooplankton from all three Adriatic sub-basins shows considerable decrease with time.

THg concentrations in the Adriatic mussels are slightly higher compared to ones from the Mediterranean Sea, but content of THg in mussels from both seas declines by approximately 10-fold in the last 30 years. Mussels highly reflect localised influence of anthropogenic contamination. Percentage of mercury as MeHg is similar in the Mediterranean and the Adriatic low-Hg environments. However, previously contaminated locations can have lower MeHg percentages. This indicates low methylation potential and/or detoxification mechanisms.

European hake and red mullet have similar THg concentrations in the Adriatic and Mediterranean seas without clear seasonal variability. THg and MeHg increase with the position of fish in the food web from herbivorous organisms to the top predator species. Percentage of mercury present as MeHg also increases in the same manner through the trophic level. Mercury concentrations are usually positively correlated with length (age) and weight of fish.

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Zastupljenost i specijacija žive u jadranskom planktonu, školjkašima i ribi – pregled

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SAŽETAK

Ovaj članak predstavlja pregled dostupnih istraživanja o specijaciji žive u najproučavanijoj bioti Jadranskog mora kao posebne biogeokemijske podjedinice Mediterana. Predstavljamo postojeće znanje o koncentraciji žive, specijaciji, prostornoj raspodjeli i vremenskim promjenama u planktonu, školjkašima i ribi Jadranskog mora. Rezultati studija o specijaciji žive u morskim organizmima su korišteni za opis razdiobe bioakumulacije žive u Jadranskom moru. Biogeokemijski ciklus žive u Jadranu karakterizira povećanje koncentracije žive od vodenog stupca, preko planktona, školjkaša i manjih riba do predatorskih ribljih vrsta. Iako Jadransko more sadrži najviše koncentracije žive u cijelom Mediteranu, riblje vrste na višim trofičkim razinama imaju slične koncentracije žive posvuda u Mediteranu, ukazujući na nepotpuno razumijevanje prijenosnih mehanizama žive iz morske vode na više trofičke razine. Budući da je konzumiranje (kontaminirane) hrane glavni put ljudske izloženosti živi, od iznimne je važnosti razumijevanje utjecaja kontaminacije živom u Jadranskom moru.

Ključne riječi: živa, metilživa, plankton, riba, dagnja