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Mercury in fishes from Augusta Bay (southern Italy): Risk assessment and health implication

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ABSTRACT

Our study reports on the total mercury (HgT) concentrations measured in the muscles and livers of several benthic, demersal and pelagic fish species caught inside and outside of Augusta Bay (southern Italy), a semi-enclosed marine area, highly contaminated by the uncontrolled (since the 1950s to 1978s) discharge of the largest European petrochemical plant. Mercury levels in fish tissues are discussed with regard to specific habitat, size and/or age of the specimens and HgT distribution in the bottom sediments. Results suggest a still active Hg release mechanism from the polluted sediments to the marine environment. Also, the high HgT concentrations measured in fishes caught in the external area of the bay imply a potential role of Augusta Bay as a pollutant source for the Mediterranean ecosystem. Finally, values of hazard target quotient (THQ) and estimated weekly intake (EWI) demonstrate that consumption of fishes caught inside the bay represents a serious risk for human health. Also, data indicate that intake of fishes caught from the external area of the bay, especially for that concern demersal and benthic species, could be represent a significant component of risk for the local population.

1. Introduction

The city of Augusta, located in the SE of Sicily (southern Italy), has experienced an important industrialisation phase since the early 60s. This has led to the creation of several chemical and petrochemical plants and oil refineries resulting in a severe pollution of the surrounding environment. In particular, the petrochemical industry in Augusta Bay is one of the largest in Europe with the most important chlor-alkali plant in Italy (Le Donne and Ciafani, 2008). Its activity started in 1958 and stopped in 2005, with production of chlorine and caustic soda by electrolysis of sodium chloride aqueous solution in electrolytic cells with a graphite anode and metallic mercury cathode. Uncontrolled chemical discharge of Hg occurred in the Augusta Bay until 1978, when restrictions were imposed by the Italian legislation.

In the last decade, several studies have provided detailed information on the pollution levels and risks for human health of resident populations of Augusta Bay (ICRAM, 2005; Ausili et al., 2008; Di Leonardo et al., 2007, 2008; ENVIRON International Team, 2008; Ficco et al., 2009; Sprovieri et al., 2011). Sprovieri et al. (2011) reported high-resolution maps of HgT distribution from superficial sediments collected in 2005, highlighting extremely high concentrations (ranging between 0.1 and 527.3 mg kg^{-1}) and speculating on the key role that Augusta Bay could play in exporting Hg to the Mediterranean Sea, as an effect of the outflow intercepted by the Levantine Intermediate Waters (LIWs). Also, data recently collected by ICRAM (2008), ENVIRON International Team (2008) and Ausili et al. (2008), demonstrated HgT transfer from the abiotic system (sediments and seawater) to fishes (top predators and filter-feeders) and documented significant health risks associated with the consumption of fish caught in the area. Toxicological Hg effects were also evaluated on mussels and red mullet by micronuclei (MN) studies, which documented DNA damage (Ausili et al., 2008 and ICRAM, 2008). Finally, Tomasello et al. (2012) report on DNA genotoxic and oxidative damages in *Coris julis* specimens from Augusta Bay.

Fish food seems to constitute the main route of Hg uptake for humans (Holsbeek et al., 1996; Nakagawa et al., 1997).

Renzoni et al. (1998) demonstrated that long-term and frequent intake of fish with high Hg levels is statistically associated with a toxic risk, especially in pregnant women. A sad, famous poisoning episode occurred in the 1950s among people living around Minamata Bay (Japan), showing the irreversible neurological damage and teratogenic effects due to consummation of Hg-contaminated fish (De Flora et al., 1994). Methylmercury (MeHg) is the most toxic form, able to interfere with thiol metabolism, causing inhibition or inactivation of proteins containing thiol ligands and ultimately

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leading to mitotic disturbances (Das et al., 1982; Elhassani, 1983). Numerous recent studies indeed have concluded that the majority, if not all, of the Hg that is bioaccumulated through the food chain is as MeHg (Winfrey and Rudd, 1990; Mason and Fitzgerald, 1990, 1991; Gilmour and Henry, 1991; Horvat et al., 1999; Carbonell et al., 2009).

High mortality rates, statistical high frequency of neonatal malformations and cancerous diseases reported for resident populations around Augusta Bay (Martuzzi et al., 2006; Bianchi et al., 2004, 2006; Fano et al., 2005, 2006; Madeddu et al., 2006) definitively calls for more detailed exploration and definitive assessment of the role played by the intake of Hg-contaminated fish on the health of the consumers.

In this work, we aim to explore the effects of HgT pollution in Augusta Bay on the fish compartment, inside and outside the semi-enclosed area, and to assess the potential health risks associated with the consumption of contaminated fish.

2. Materials and methods

2.1. Sampling

Four different sampling sites were selected: two inside, and two outside of Augusta Bay (Fig. 1). Sampling outside the bay was performed during May 2001, on board of the N/O "Dallaporta", by means of a mid-water trawl-net at 50–100 m of depth in two sampling areas, in front of the Scirocco inlet (300-m wide and 13-m deep), and the Levante inlet (400-m wide and 40-m deep) (Fig. 1: C1, C2). Mainly pelagic fish specimens were caught (Table 1). Sampling inside the bay was performed during May 2012 by means of a fishing boat equipped with a gillnet wall, positioned at the bottom (mean depth = 20–25 m) (Fig. 1: C3, C4). Several specimens of benthic and demersal fishes were collected. From the two sampling activities, a total of 227 fish specimens were collected: 107 from mid-water sampling (outside the bay) and 120 from bottom-water sampling (inside the bay). Moreover, specimens of *Engraulis encrasicolus* ($n = 38$) were caught from the unpolluted marine area of Marsala (western Sicily) (Fig. 1), during July 2001, on board of a fishing boat equipped with a purse seine net. After collection, fishes were stored at −20 °C until biological and chemical analyses were performed in the laboratories of biology and biogeochemistry at the Institute for Coastal and Marine Environment (CNR) of Capo Granitola.

2.2. Biological data and tissue collection

The total length (TL) of each specimen was measured. Muscle and liver tissues were collected from each organism, using plastic materials cleaned with HNO_3 (10%) and MilliQ water, in order to avoid Hg contamination. Tissues were stored at −20 °C until analysis. Otoliths were extracted from anchovy and sardine specimens for age determination. Readings and interpretation of otolith increment growths were carried out by transmitted visible lights based on higher-resolution microscopy (20–25× magnification) (Campana et al., 1987; Nielsen, 1992). The procedure adopted for European anchovy age determination follow Uriarte et al. (2007) and La Mesa et al. (2009).

2.3. Chemical analyses

Total mercury concentrations (HgT) in tissues were measured using a direct mercury analyser (Milestone_DMA-80), atomic absorption spectrophotometer, according to analytical procedures reported in EPA 7473. Briefly, approximately 0.1 g of fresh tissue was loaded in nickel boats and transferred to the DMA-80 system. In order to minimise contamination risks, acid-cleaned laboratory materials were used during sample preparation and analyses. A Reference Standard Material (TORT-2; HgT certificate value = 0.27 ± 0.06 μg g^{-1}) was analysed to assess analytical accuracy (estimated to be ∼3%) and precision (routinely better than 4%; RSD%, $n = 3$). Finally, duplicated samples (about 20% of the total number of samples) were measured to estimate reproducibility, which resulted in better than 7%.

2.4. THQ and EWI calculation

Target hazard quotient (THQ) and estimated weekly intake (EWI) were calculated for muscles of fishes caught inside and outside the bay.

The target hazard quotient was calculated according to the US EPA (1989) method and it is described by the following equation:

$$THQ = \left(\frac{EF \times ED \times FIR \times C}{RFD \times WAB \times TA}\right) \times 10^{-3}$$

where EF is exposure frequency (365 days/year); ED is the exposure duration (70 years), equivalent to the average lifetime; FIR is the food ingestion rate (36 g/person/day) (FAO, 2005); C is the metal concentration in seafood (μg g^{-1}); RFD is the USEPA's reference dose (0.1 μg Hg kg bw^{-1} d^{-1}) (http://cfpub.epa.gov) or acceptable daily intake determined by WHO (0.23 μg Hg kg bw^{-1} d^{-1}) (http://apps.who.int); WAB is the average body weight (60 kg), and TA is the average exposure time for no carcinogens (365 days/year × ED).

The THQ was calculated for all the studied species in the Augusta Bay using the US-EPA's reference dose (THQa) and the acceptable daily intake determined by the WHO (THQb). In particular, we assumed that the measured mercury is integrally in its methylated form (Winfrey and Rudd, 1990; Mason and Fitzgerald, 1990, 1991; Gilmour and Henry, 1991; Horvat et al., 1999; Carbonell et al., 2009).

The estimated weekly intake (EWI) was calculated by multiplying the HgT concentration (C) times by the weekly dietary intakes (FIR × 7) and reporting to the average body weight (WAB).

Finally, mean THQ and EWI values were calculated for each studied species. Also, considering that fishing activity within the bay has been interdicted since 2007 (Order No. 73/07), data relative to fishes from inside and outside the bay were processed separately.

3. Results

3.1. Biological features

The fish caught from bottom-water sampling (inside the bay) consisted of 2 pelagic, 106 demersal and 16 benthic specimens, while specimens from mid-water sampling (outside the bay), consisted of 103 pelagic, 3 demersal and 1 benthic (Table 1). A total of 21 different species were recognised. The number of specimens per species and total length ranges are shown in Table 2. Almost all the caught species, in particular, *E. encrasicolus, Sardina pilchardus, Boops boops, Mullus barbatus* and *Illex coindetii*, are typical of the Mediterranean Sea and are commercially relevant to Italian fishing (Irepa, 2010). Only one specimen was found to belong to a so-called alien species, specifically *Sphyraena sphyraena*. This is a typical species of the tropical seas, today present also in the Mediterranean Sea (Streftaris and Zenetos, 2006).

3.2. Total mercury concentrations (HgT)

Total mercury concentrations measured in tissues from pelagic, demersal and benthic fishes, caught inside and outside of Augusta Bay, are graphically summarised in Fig. 2a and b. Mercury mean values calculated for each species, together with available comparative data from the literature and HgT content measured in anchovies from Marsala, are presented in Table 2.

Mercury concentrations ranged between 0.021 and 2.709 μg g^{-1} in muscles (Fig. 2a) and between 0.029 and 9.720 μg g^{-1} in livers (Fig. 2b). The HgT content in liver is from 1.5 to 6 times higher than that measured in muscles from the same specimens (Table 2). The highest HgT values were found in species caught inside the bay: 2 demersal specimens, a specimen of *Diplodus vulgaris* (HgT in liver = 4.979 μg g^{-1}) (extreme point in Fig. 2b) and a specimen of *Serranus scriba* (HgT in muscle = 2.709 μg g^{-1}) (extreme point in Fig. 2a), a large pelagic specimen of *S. sphyraena* (HgT = 9.720 and 2.269 μg g^{-1} in liver and muscle, respectively) (Table 2) and a benthic specimen of *Murena helena* (HgT = 2.638 μg g^{-1} in muscle) (Table 2). However, these very high levels represent outliers of the whole dataset (Fig. 2a and b). The highest non-outlier values refer once again to specimens caught inside the bay and specifically to benthic species (Fig. 2a and b). In particular, *Scorpanea scrofa* and *Scorpanea notata* show the highest HgT mean concentrations for both liver (1.638 and 2.339 μg g^{-1}, respectively) and muscle (1.082 and 1.341 μg g^{-1}, respectively) (Table 2). The lowest non-outlier ranges were found in pelagic specimens caught outside the bay (0.021–0.167 μg g^{-1} for muscles and 0.029–0.5708 for livers) (Fig. 2a and b), and the HgT mean values measured in the different studied species are substantially comparable (Table 2). Finally, data for demersal species from the

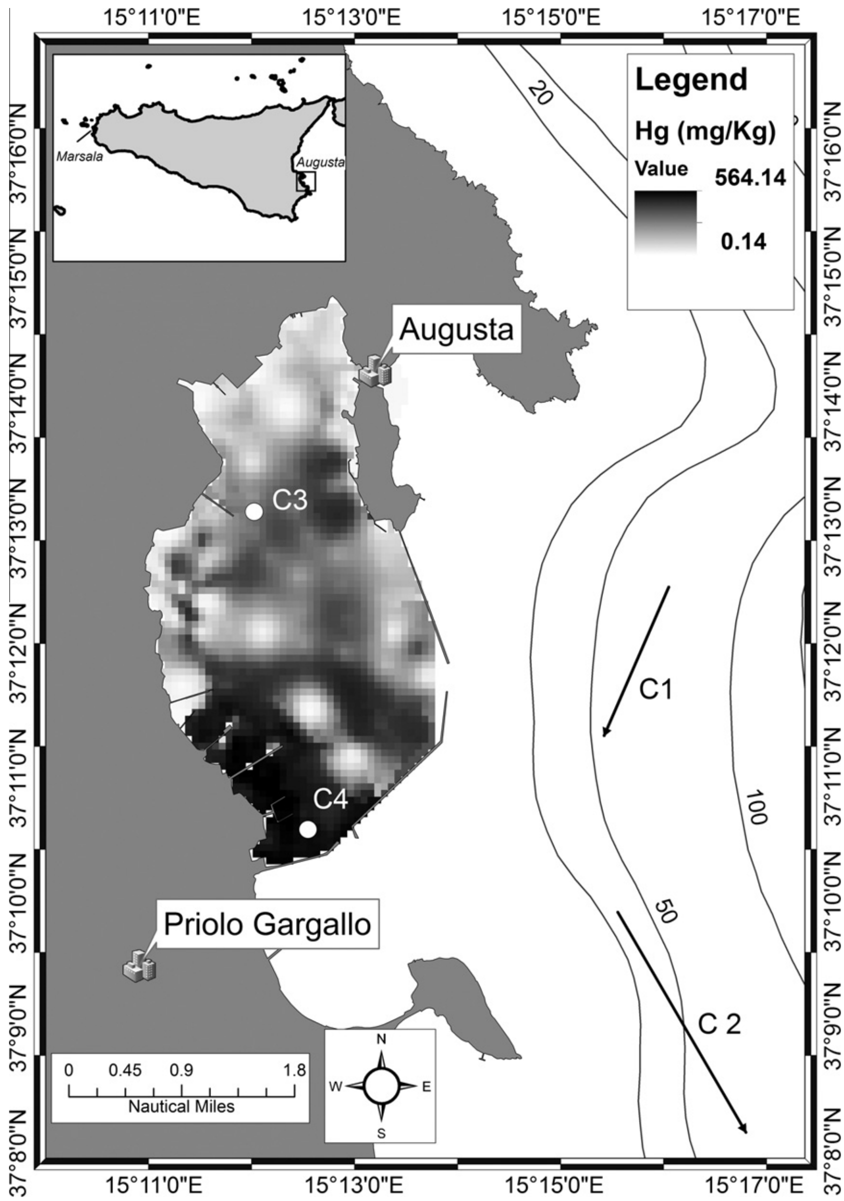

Fig. 1. Sampling sites in Augusta Bay and distribution of total mercury (HgT) in bottom sediments (data from Sprovieri et al. (2011)).

Table 1
Number of specimens *per* species caught in the sampling sites.

Mid-water sampling (outside the bay)				Bottom-water sampling (inside the bay)			
Species	C1 (no.)	C2 (no.)	Habitat	Species	C3 (no.)	C4 (no.)	Habitat
Engraulis encrasicolus	20	20	Pelagic	*Diplodus annularis*	59	15	Demersal
Sardina pilchardus	8	20	Pelagic	*Diplodus vulgaris*	–	3	Demersal
Boops boops	–	20	Pelagic	*Pagellus erythrinus*	1	6	Demersal
Trachurus trachurus	–	6	Pelagic	*Pagellus acarne*	11	1	Demersal
Illex coindetii	6	–	Pelagic	*Sepia officinalis*	2	6	Demersal
Loligo forbesi	3	–	Pelagic	*Serranus scriba*	2	–	Demersal
Pagellus erythrinus	1	–	Demersal	*Caranx rhonchus*	1	–	Pelagic
Pagellus bogaraveo	2		Demersal	*Sphyraena sphyraena*	1		Pelagic
Mullus barbatus	1	–	Benthic	*Scorpaena notata*	–	5	Benthic
				Scorpaena scrofa	3	1	Benthic
				Mullus barbatus	–	3	Benthic
				Mullus surmuletus	1	1	Benthic
				Murena helena	1	–	Benthic
				Octopus vulgaris	–	1	Benthic

Table 2
HgT means concentrations in muscle and liver of the analysed species and comparison with data for other areas.

Species	No.	Range of total length (mm)	HgT muscle (μg g^{-1})	S.D.	References	Site	HgT liver (μg g^{-1})	S.D.
Engraulis encrasicolus	40	109–138	0.052	0.019	This work	Augusta	0.204	0.147
	11	120–139	0.057	0.014	This work	Marsala	0.119	0.038
			0.040		Bilandžić et al. (2011)	Adriatic sea		
	9	121–147	0.070	0.090	Gibičar et al. (2009)	Adriatic sea[a]		
			0.030	0.030	Copat et al. (2012)	Sicily (Catania)		
			0.060	0.030	Copat et al. (2012)	Syracuse (Sicily)		
	18		0.060		Pastor et al. (1994)	Mediterranean sea (Spain)[a]		
			0.070		Martorell et al. (2011)	Mediterranean sea (Spain)		
	4		0.055	0.003	Tuzen (2009)	Black sea (Turkey)[a]		
Sardina pilchardus	28	115–150	0.082	0.035	This work	Augusta	0.196	0.157
	10	168–178	0.090	0.040	Gibičar et al. (2009)	Adriatic sea[a]		
			0.080	0.030	Copat et al. (2012)	Catania (Sicily)		
			0.180		Buzina et al. (1995)	Adriatic sea[a]		
			0.198		Buzina et al. (1995)	Adriatic sea (Kastela Bay)[a]		
	14		0.052		Wolfgang (1983)	Adriatic sea[a]		
	35	190–260	0.066		Wolfgang (1983)	Biscay Bay		
	5	157–165	0.050		Wolfgang (1983)	Mediterranean sea		
	41		0.170		Wolfgang (1983)	Ligurian sea		
	28	120–150	0.030		Wolfgang (1983)	North Africa (Ceuta)[a]		
	20	160–210	0.040		Wolfgang (1983)	Western english channel		
	38		0.105		Pastor et al. (1994)	Mediterranean sea (Spain)[a]		
			0.019		Martorell et al. (2011)	Mediterranean sea (Spain)		
	7	188–200	0.033	0.016	Harakeh et al. (1985)	Lebanon		
Boops boops	20	95–150	0.120	0.049	This work	Augusta	0.236	0.191
	11	158–198	0.196	0.204	Hornung et al. (1980)	Israel[a]		
	1		0.075		Pastor et al. (1994)	Mediterranean sea (Spain)[a]		
	2	130–160	0.190		Stoeppler and Nürnberg (1979)	Med. sea (Dubrovnik)		
			0.267		Buzina et al. (1995)	Adriatic sea		
			0.312		Buzina et al. (1995)	Adriatic sea (Kastela Bay)[a]		
	16	139–171	0.036	0.025	Harakeh et al. (1985)	Lebanon		
Trachurus trachurus	6	56–222	0.131	0.147	This work	Augusta	0.344	0.176
	2	260–285	0.170		Stoeppler and Nürnberg (1979)	North sea (German Bight)		
		170	0.170		Mikac et al. (1984)	Adriatic sea (Kastela Bay)[a]		
	37	130–236	0.122	0.101	Hornung et al. (1980)	Israel[a]		
	16	159–203	0.045	0.019	Harakeh et al. (1985)	Lebanon		
			0.053		Martorell et al. (2011)	Mediterranean sea (Spain)		
	4		0.078	0.005	Tuzen (2009)	Black Sea (Turkey)[a]		
	5		0.053	0.012	Keskin et al. (2007)	Marmara sea (Turkey)[a]		
Diplodus annularis	74	109–179	0.557	0.303	This work	Augusta	1.195	0.827
			0.653		Buzina et al. (1995)	Adriatic sea[a]		
			0.628		Buzina et al. (1995)	Adriatic sea (Kastela Bay)[a]		
Diplodus vulgaris	3	102–179	0.643	0.614	This work	Augusta	2.035	2.554
	5		0.378	0.017	Keskin et al. (2007)	Marmara sea (Turkey)[a]		
Sphyraena sphyraena	1	1190	2.269		This work	Augusta	9.727	
	14	219–295	0.167	0.068	Hornung et al. (1980)	Israel[a]		
Caranx rhonchus	1	264	1.701		This work	Augusta		
Pagellus acarne	12	149–161	0.254	0.028	This work	Augusta	0.618	0.178
	3	135–141	0.112		Hornung et al. (1980)	Israel[a]		
	15	164–182	0.032	0.014	Harakeh et al. (1985)	Lebanon		
Pagellus bogaraveo	2	178–179	0.266	0.227	This work	Augusta	1.230	0.700
Pagellus erythrinus	8	154–205	0.407	0.100	This work	Augusta	2.322	0.445
	5	110	0.341	0.025	Papetti and Rossi (2009)	Tyrrhenian sea (Lazio)		
	57	115–187	0.180	0.094	Hornung et al. (1980)	Israel[a]		
	9	89–173	0.240	0.190	Gibičar et al. (2009)	Adriatic sea[a]		

(continued on next page)

Table 2 (*continued*)

Species	No.	Range of total length (mm)	HgT muscle (μg g^{-1})	S.D.	References	Site	HgT liver (μg g^{-1})	S.D.
	28	140–152	0.042	0.023	Harakeh et al. (1985)	Lebanon		
	5		0.168		Pastor et al. (1994)	Mediterranean sea (Spain)[a]		
	5		0.290	0.044	Keskin et al. (2007)	Marmara sea (Turkey)[a]		
Serranus scriba	2	122–140	2.165	0.768	This work	Augusta	2.581	0.592
	3		1.030	0.459	Gibičar et al. (2009)	Tyrrhenian sea (Tuscany)[a]		
Mullus barbatus	4	155–202	0.815	0.777	This work	Augusta	1.518	0.582
		102–230	0.116	0.056	Hornung et al. (1980)	Israel[a]		
			0.400	0.400	Storelli et al. (2004)	Ionian sea		
			0.490	0.500	Storelli et al. (2004)	Adriatic sea[a]		
	13	117–180	0.700	0.730	Gibičar et al. (2009)	Adriatic sea[a]		
			0.370		Buzina et al. (1995)	Adriatic sea		
			0.318		Buzina et al. (1995)	Adriatic sea (Kastela Bay)[a]		
	59		0.139		Pastor et al. (1994)	Mediterranean sea (Spain)		
			0.010		Martorell et al. (2011)	Mediterranean sea (Spain)		
	30	128–166	0.054	0.025	Harakeh et al. (1985)	Lebanon		
		130–200	0.233		Stoeppler and Nürnberg (1979)	Mediterranean Sea (Sardinia)		
	4		0.036	0.002	Tuzen (2009)	Black Sea (Turkey)[a]		
	5		0.434	0.012	Keskin et al. (2007)	Marmara sea (Turkey)[a]		
Mullus surmuletus	2	200–209	0.662	0.089	This work	Augusta	1.112	
	9	120–160	0.086		Hornung et al. (1980)	Israel[a]		
	59		0.139		Pastor et al. (1994)	Mediterranean sea (Spain)[a]		
	2	185–203	0.250		Stoeppler and Nürnberg (1979)	North sea (German Bight)		
	37		0.060		Bilandžic et al. (2011)	Adr.sea (Croatian coast)		
Scorpaena scrofa	4	93–112	1.082	0.285	This work	Augusta	1.637	0.380
			0.222		Buzina et al. (1995)	Adriatic sea		
			0.390		Buzina et al. (1995)	Adr. sea (Kastela Bay)[a]		
Scorpaena notata	5	114–133	1.340	0.380	This work	Augusta	2.339	0.529
	5		0.490	0.430	Gibičar et al. (2009)	Tyrrhenian sea (Tuscany)[a]		
Illex coindetii	6	33–92	0.078	0.039	This work	Augusta		
	13	52–224	0.100	0.100	Gibičar et al. (2009)	Adriatic sea[a]		
Loligo forbesi	3	45–170	0.147	0.024	This work	Augusta	0.311	0.011
Sepia officinalis	8	108–148	0.766	0.288	This work	Augusta		
Octopus vulgaris	1	123	0.443		This work	Augusta		
Murena helena	1	800.5	2.638		This work	Augusta	3.817	

[a] Polluted site.

inner bay show the widest non-outlier ranges (0.084–1.116 μg g^{-1} for muscles, 0.109–2.747 μg g^{-1} for livers) and the most elevated number of outliers and extreme values (Fig. 2a and b). In particular, the highest HgT mean values (2.165 μg g^{-1} in liver and 2.581 μg g^{-1} in muscle) were measured in the *S. scriba* species (Table 2).

3.3. THQ and EWI values

Mean THQ and EWI values calculated for each caught species, inside and outside the bay, are reported in Table 3. Most of the fish species inside the bay show higher values (TQa = 1.53–15.8; TQb = 0.66–6.88; EWI = 1.06–11.0) than those outside the studied area (TQa = 0.31–4.20; TQb = 0.66–6.88; EWI = 0.22–2.91). In particular, the highest values were calculated for *M. helena* (TQa = 15.8; TQb = 6.88; EWI = 11.0), *S. scriba* (TQa = 13.0; TQb = 5.65; EWI = 9.02) and *Caranx rhonchus* (TQa = 10.2; TQb = 4.44; EWI = 7.09) caught inside the bay, while, the pelagic species outside the bay show the lowest values (TQa = 0.31–0.88; TQb = 0.14–0.38; EWI = 0.22–0.61). Finally, no significant differences were found between the same or similar species, collected inside (*Pagellus acarne, Pagellus erythrinus, M. barbatus, Mullus surmuletus*) and outside (*Pagellus bogaraveo, P. erythrinus* and *M. barbatus*) the bay.

4. Discussion

4.1. Mercury bioaccumulation effects: length/age vs. HgT content

The Hg accumulation in marine fish primarily depends on some important biokinetic parameters: assimilation from the ingested prey, uptake constants from the aqueous phase, de-toxification rate (Wang, 2012; Wang et al., 1997, 1998; Wang and Fisher, 1999; Dang and Wang, 2011) and environmental features (e.g., Hg concentration and speciation in seawater, dietary sources, etc.) (Wang and Wong, 2003). However, physiological and geochemical species-specific influences on Hg bioaccumulation are still not fully understood (Baines et al., 2002; Xu and Wang, 2002; Wang and Wong, 2003; Dang and Wang, 2012).

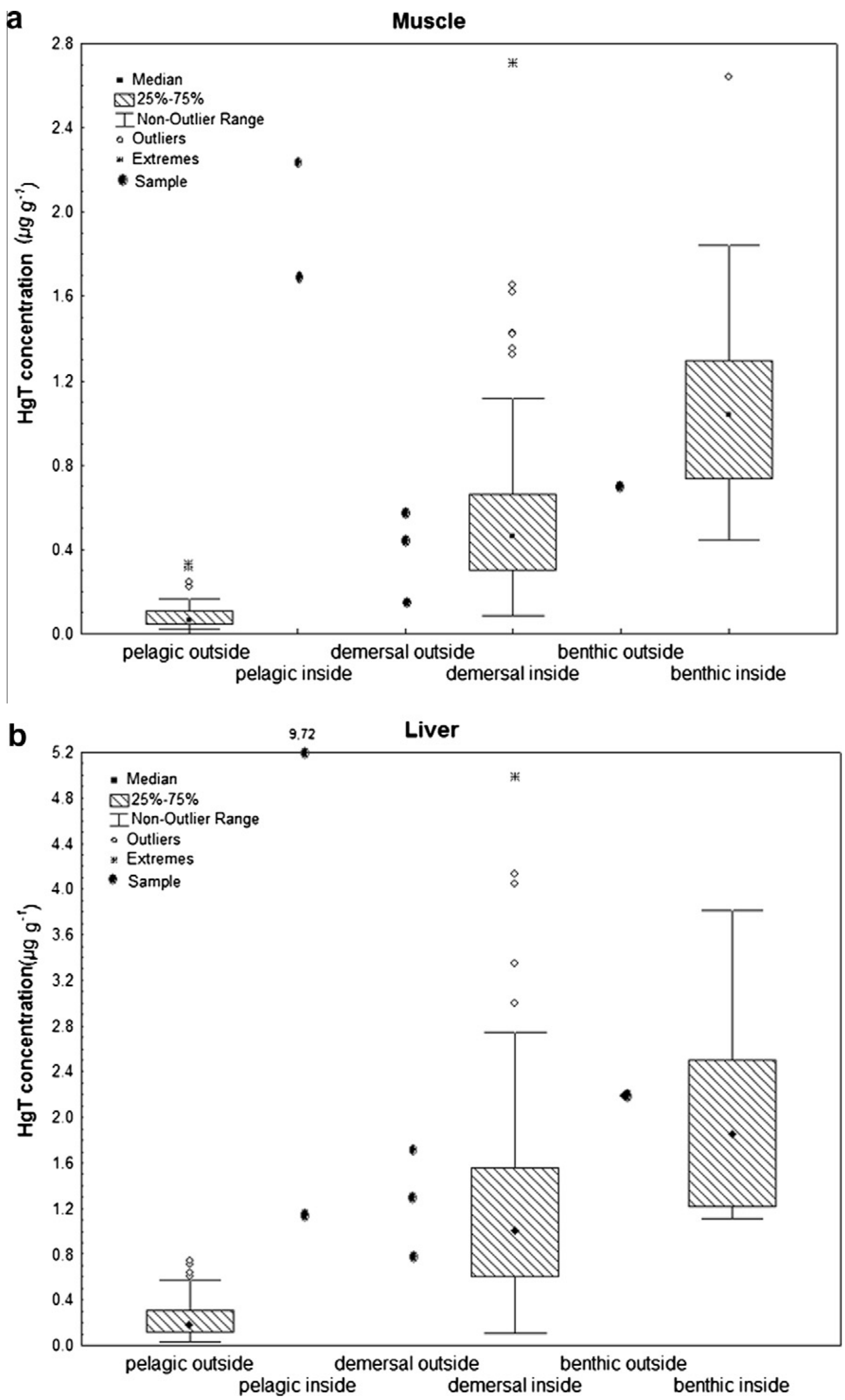

Fig. 2. Box-plots with HgT concentrations in the muscles and livers of pelagic, demersal and benthic fishes.

Several studies have demonstrated that Hg concentrations in the muscles of marine organisms proportionally increase with size and age (Lange et al., 1994; Burger et al., 2001; Green and Knutzen, 2003; Simonin et al., 2008). Moreover, Hg de-toxification rates appear negatively correlated with the fish size (Trudel and Rasmussen, 1997), supporting a potential correlation between Hg levels and size/age in the organisms. However, detailed investigations on different groups of species and on a wide range of HgT concentrations are lacking and, when available, sometimes controversial (Stafford and Haines, 2001), especially for fishes with low mercury levels (average below 0.2 ppm) (Park and Curtis, 1997; Burger and Gochfeld, 2011). Strong correlations between size and Hg levels in fish are reported for Swordfish (*Xiphias gladius*) and Bluefin Tuna (*Thunnus thynnus*) from the Mediterranean Sea (Storelli and Marcotrigiano, 2001), for several pelagic fish species from the Adriatic Sea (Storelli, 2008) and for *S. pilchardus* specimens from Tunisia (Joiris et al., 1999). Furthermore, Burger et al. (2007) found a positive correlation between size and Hg levels for 11 of 14 species of marine fishes collected in the western Aleutians (Bering Sea/North Pacific) and Luten et al. (1987) found the same positive correlation in Atlantic Cod. Moreover, Leonzio et al. (1981) report positive correlations between Hg content and weight in *M. barbatus* and a slight Hg increasing trend with size in *E. encrasicolus* from the northern Tyrrhenian Sea. Finally, Gewurtz et al. (2011) show strong

Table 3
THQ and EWI calculation for each caught species (inside and outside the Bay).

Inside				Outside			
Species	THQa	THQb	EWI	Species	THQa	THQb	EWI
Caranx rhonchus	10.2	4.44	7.09	*Engraulis encrasicolus*	0.31	0.14	0.22
Diplodus annlularis	3.34	1.45	2.32	*Sardina pilchardus*	0.49	0.21	0.34
Diplodus vulgaris	3.86	1.68	2.68	*Boops boops*	0.72	0.31	0.50
Pagellus acarne	1.53	0.66	1.06	*Trachurus trachurus*	0.79	0.34	0.55
Pagellus erythrinus	2.30	1.00	1.60	*Illex coindetti*	0.47	0.20	0.33
Scophaena scrofa	7.17	3.12	4.98	*Loligo forbesi*	0.88	0.38	0.61
Scorphaena notata	8.05	3.50	5.59	*Pagellus bogaraveo*	1.60	0.69	1.11
Mullus barbatus	5.59	2.43	3.88	*Pagellus erythrinus*	3.41	1.48	2.37
Mullus surmuletus	3.97	1.73	2.76	*Mullus barbatus*	4.20	1.82	2.91
Serranus scriba	13.0	5.65	9.02				
Murena helena	15.8	6.88	11.0				
Octopus vulgaris	2.66	1.16	1.85				
Sepia officinalis	4.60	2.00	3.19				

a: USEPA's reference dose (0.1 μg Hg kg bw^{-1} d^{-1}).
b: acceptable daily intake determined by WHO (0.23 μg Hg kg bw^{-1} d^{-1}).

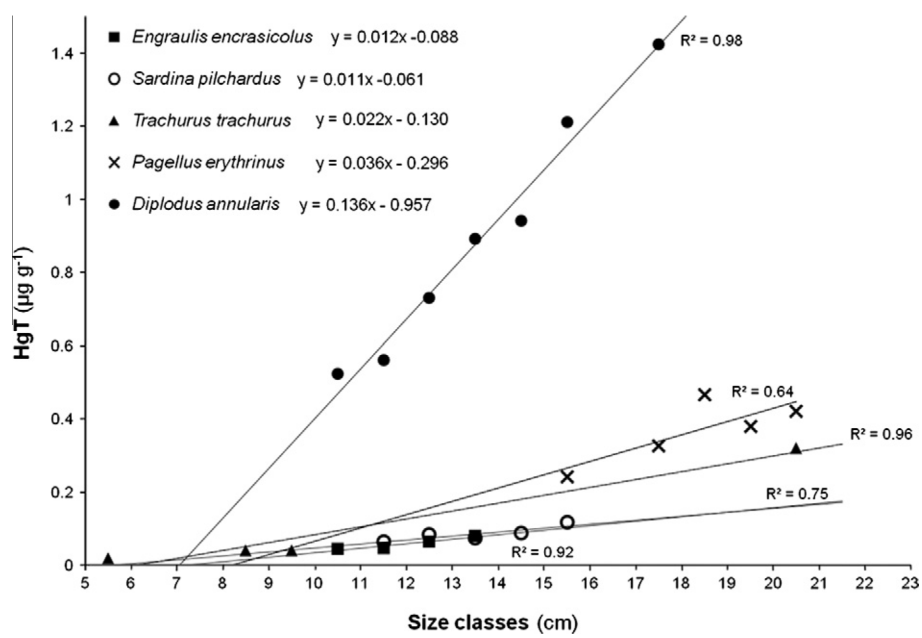

Fig. 3. Relationship between HgT concentrations and total body length for *Sardina pilchardus*, *Engraulis encrasicolus*, *T. trachurus, D. annularis* and *P. erythrinus.* Points represent the mean values for each size class.

correlation between HgT concentration and length in most freshwater fishes from the Canadian Great Lakes and Ontario (Canada).

Here, the high number of specimens available from pelagic, benthic and demersal fish species associated with a wide range of length/age and HgT variability detected in tissues offer a challenging opportunity to explore in more depth the actual bioaccumulation processes of Hg in the studied organisms. In particular, we assumed the length of fishes as a reliable parameter for age estimates (Boening, 2000; Waldron and Kerstan, 2001; Scudder et al., 2009; Panfili et al., 2010; Basilone et al., 2011; Bacha et al., 2012) and, thus, reported HgT values vs. length to assess biomagnification of that contaminant with time. Statistically reliable and robust correlations were found between HgT mean values measured in muscles for size classes and length in *S. pilchardus* ($r^2 = 0.75$), *E. encrasicolus* ($r^2 = 0.92$), *Trachurus trachurus* ($r^2 = 0.96$), *Diplodus annularis* ($r^2 = 0.98$) and *P. erythrinus* ($r^2 = 0.64$) (Fig. 3). Specifically, the calculated HgT accumulation rates for *S. pilchardus, E. encrasicolus, T. trachurus, P. erythrinus* and *D. annularis* are 0.011, 0.012, 0.022, 0.036 and 0.136 μg g^{-1} -cm^{-1}, respectively, in good agreement with data reported by Hornung et al. (1980) for *P. erythrinus* and *T. trachurus* species. This definitively supports a significant linear HgT-length relationship for the studied fish species and a species-specific accumulation effect on the studied marine organisms.

In our dataset an evident increasing trend was measured between HgT content and age in the two most abundant species, *E. encrasicolus* and *S. pilchardus* (Fig. 4) with significant differences ($p < 0.005$; ANOVA test) among age group, although, the restricted range of available age classes needs a larger data collection.

4.2. Sources of HgT and fish contamination in Augusta Bay

Muscles are the most commonly analysed tissues to monitor Hg levels in fishes because they represent the edible part of the organism associated with human health risk implications (Henry et al., 2004). Indeed, Hg accumulates over time more readily in liver than in muscle, but muscle appears to retain Hg for a much longer period (Boudou and Ribeyre, 1995). Thus, liver may provide information only on short-term exposure to Hg pollution or may bioaccumulate only when an organism is exposed to constant or increasing levels of dietary mercury (Atwell et al., 1998). This is clearly reflected in the studied dataset, where HgT concentration

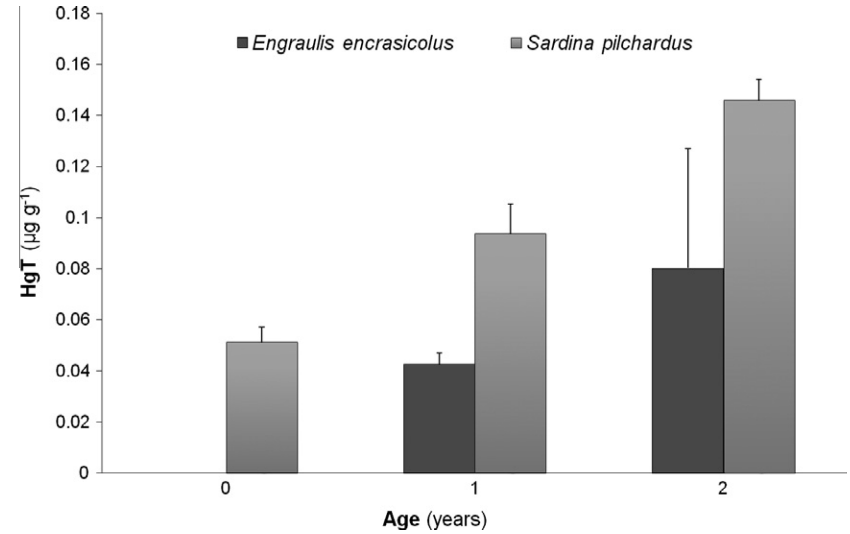

Fig. 4. Relationship between total mercury concentration (median value of HgT) in fish muscles vs. age in *E. encrasicolus* and *S. pilchardus*. Black lines = confidence interval.

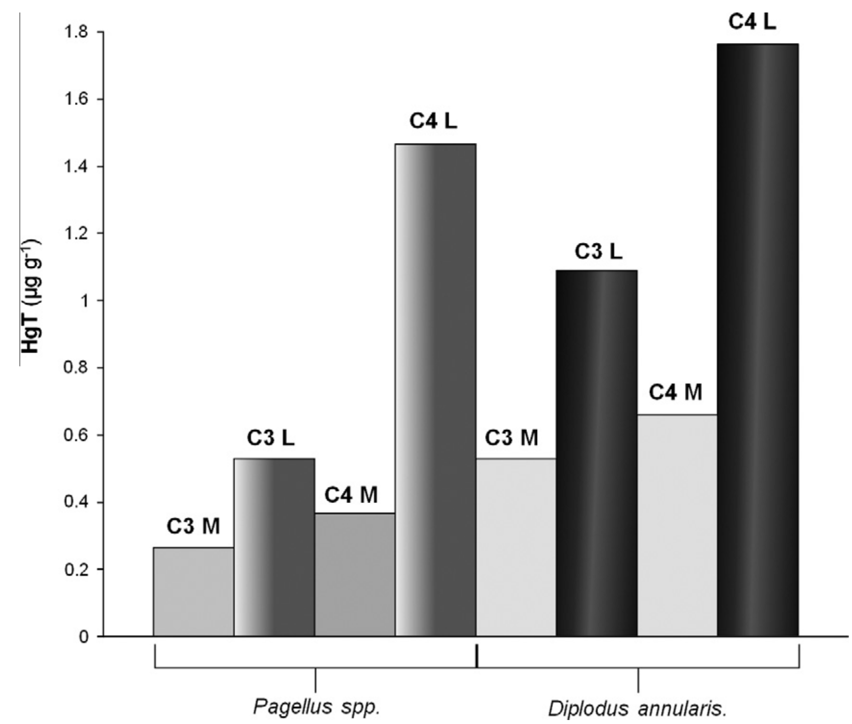

Fig. 5. Differences in muscle (M) and liver (L) HgT contents in *Pagellus* spp. *and Diplodus annularis* from the northern (C3) and the southern (C4) part of Augusta Bay.

measured in liver is up to two orders of magnitude higher than in muscles.

The HgT content measured in the tissues of fishes from Augusta Bay show an increasing trend with habitat depth, specifically, with highest values measured in benthic species with respect to the lowest levels detected in pelagic organisms (Table 2). Additionally, contamination effects show a south-north gradient evident from HgT levels measured on the ubiquitous *Pagellus* spp. and *D. annularis* specimens (Fig. 5). In particular, the highest HgT mean concentrations occur in fishes caught from southern Augusta Bay where bottom sediments show the highest concentrations of mercury (Sprovieri et al., 2011) (Fig. 1). This suggests a key role played by the highly polluted sediments as sources of Hg to the investigated marine environment. Also, measurements of HgT in seawater reported from the bottom, mid and surface waters by ENVIRON International Team (2008) with an average concentration of 0.25 nmol L^{-1} and range of 0.05–0.37 nmol L^{-1} show a crucial effect of Hg efflux from sediments of the bay to the water column with a potential direct impact on the bioaccumulation processes in the trophic web. A direct comparison of HgT content in benthic species from Augusta Bay and other Mediterranean areas affected by comparable Hg discharges by chlor-alkali plant and sewage sludge disposal, specifically Tuscany and Israel (Hornung et al., 1980; Gibičar et al., 2009), show 2–7 times higher values, thus, underlying the combined effects of high pollution levels and specific biogeochemical pathways driving mercury bioavailability in the studied system (Table 2).

Sprovieri et al. (2011) and Fantozzi et al. (2013) have argued on a potential Hg export from Augusta Bay to the Eastern Mediterranean seawater, as a result of the measured 3D circulation system.

Nonetheless, Hg contamination detected in sediments outside of Augusta Bay, by effects of dredged material from the inner bay (Di Leonardo et al., 2008; Tranchida et al., 2010), could also directly influence the state of pollution of the open sea.

Here, we extend the potential role of Augusta Bay as an Hg point source for the open ocean also considering the significant transfer of pollutants by pelagic fishes moving between the inner and external part of the bay. This implies potential effects on the food web of the surrounding area as already reported by several authors studying marine systems (Riisgard and Hansen, 1990; Futter, 1994; Jarman et al., 1996; Atwell et al., 1998). This hypothesis is supported by the high mean HgT concentrations measured in pelagic species caught outside the bay, which are similar to those reported for other sites affected by Hg pollution: the Adriatic Sea (Wolfgang, 1983; Storelli and Marcotrigiano, 2001; Storelli et al., 2002, 2004, 2007, 2010; Gibičar et al., 2009), Turkish areas (Tuzen, 2009), Spanish coastal areas (Pastor et al., 1994) and Israel area (Hornung et al., 1980) (Table 2).

The HgT mean concentrations measured in the livers of *E. encrasicolus* specimens from Augusta Bay are about twice as high ($p = 0.044$) as those measured in fishes from the unpolluted area of Marsala (Table 2), suggesting a direct, short-term effect of the bay pollution on the pelagic fishes. Moreover, the caught pelagic species prefer to inhabit warmer coastal seawaters during their first life stages (Basilone et al., 2011), but they generally move in deeper waters during the older stages (Wirszubski, 1953; Schneider, 1990; Whitehead, 1990), thus, representing a significant and potential vehicle of contaminants to the deep marine food web. This evidence definitively corroborates our hypothesis of a potential Hg export through the food web, from Augusta Bay to the surrounding area.

4.3. Target hazard quotient and weekly intake: a real health risk from fishery in the Augusta Bay?

Although estimation of the target hazard quotient (THQ) and weekly intake (EWI), do not provide a quantitative and definitive estimate on the dangerous health effects on exposed populations, these methodologies offer preliminary information on the health risk level resulting from pollutant exposure. Several authors showed that selenium (Se) offers protection against Hg toxicity (Parízek and Ostádalová, 1967; Satoh et al., 1985; Ralston, 2009; Lémire et al., 2010) that suggests to take in account Se contents in fishes to assess a real risk associated to Hg intake. Positive relationships has been found between Hg and Se contents in different seawater fishes (Burger and Gochfeld, 2011; Dang and Wang, 2011; Calatayud et al., 2012). Ralston et al. (2008) showed that Se:Hg molar ratios above 1 protect against Hg toxicity. However this ratio definitively depends o species-specific toxic-kinetics processes (Watanabe, 2002; Burger and Gochfeld, 2012). This feature leads to a wide variability of Se:Hg molar ratios and makes difficult their use in risk assessment. Accordingly, here we estimated health risk for Hg intake only on THQ and EWI parameters. These indexes are widely used to assess risk associated with fish consumption (Storelli et al., 2004, 2010; Storelli, 2008; Martorell et al., 2011; Domingo et al., 2012). In particular, for no carcinogenic effects, an HQ exceeding 1.0 indicates a potential health risk (US EPA, 1989). In our dataset, either using USEPA's reference dose (TQa) that WHO acceptable daily intake (TQb), species inside the Bay exceeded the value 1 in all cases, while fishes outside the Bay only in demersal and benthic fishes (*P. bogaraveo, P. erythrinus, M. barbatus*) (Table 3). International agencies indicate a provisional tolerable weekly intake (PTWI) of Hg, ranging from 0.7 $\mu g\,kg^{-1}$ body weight (b.w.) (US-EPA, 2004) to 1.6 $\mu g\,kg^{-1}$ b.w. (FAO/WHO, 2006). These limits represent safe values for human population over lifetime. The calculated EWI index exceed the PTWI (US-EPA, 2004; FAO/WHO) in almost all the species collected inside the Bay and in demersal and benthic fishes from outside. In summary, the calculated THQ and EWI highlight that the consumption of fish from inside the Bay represents a serious risk for human health of resident populations and confirm the importance of the current fishing ban in this area. Also, the results suggest caution in the consumption of fishes from outside the Bay, especially of demersal and benthic species, confirming that Hg contamination in this area is a serious concern that calls for appropriate and timely social actions.

5. Conclusions

The main conclusions of this work can be synthesised as follows:

– The high HgT concentrations measured in benthic species from Augusta Bay suggest an active release mechanism of mercury from polluted sediments to the water column, with consequent effects of bioaccumulation in the trophic web.
– High contamination of pelagic species measured in the external zone of the bay confirms the role of the Augusta marine environment as a potential Hg source for the surrounding area and underscores the crucial risk associated with contaminant transfer from the semi-enclosed basin to the open sea.
– The THQ and EWI values advise that consumption of fish from inside the Augusta Bay represents a serious risk for human health of the local populations, while suggest caution in consuming demersal and benthic fishes from outside the Augusta Bay definitively demanding for appropriate social actions.

Conflict of Interest

The authors declare that there are no conflicts of interest.

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