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Assessment of mercury exposure in human populations: A status report from Augusta Bay (southern Italy)

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ABSTRACT

Here we investigate mercury concentrations in the blood (HgB), urine (HgU) and human hair (HgH) of 224 individuals from a coastal area (Eastern Sicily, SE Italy) strongly affected by Hg contamination from one of the largest chlor-alkali plants in Europe. The factors affecting the distribution of Hg and the extent of the exposure of individuals have been explored with a multidisciplinary approach. Multiple regression analyses, together with evidence of high levels of HgB (exceeding the HBMI recommended levels in 50% of cases) and HgH (exceeding the EPA reference dose in 70% of cases), primarily suggest that the consumption of local fish is the main source of Hg for humans. no. significant exposure to inorganic mercury was identified. Toxicokinetic calculations produced a provisional tolerable weekly intake (PTWI) level that, in most cases, exceeds international recommendations, particularly for residents in the studied area.

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1. Introduction

Due to its high versatility, mercury (Hg) is used intensively in a number of human activities such as industry, pharmacology, gold mining and agriculture. Among the anthropogenic contributions, chlor-alkali plants contribute up to 15% of total European Hg anthropogenic emissions (Pacyna et al., 2001). Unfortunately, the use of Hg is often connected to straight environmental damage that could be possibly associated with episodes of human intoxication (De Flora et al., 1994; Sanfeliu et al., 2003; Baird and Cann 2004). Organometallic methylmercury (MHg), which is primarily ingested through a seafood diet, is the most dangerous Hg form and, due to its high solubility in lipids, can have adverse effects on the liver, reproductive organs (JOINT FAO/WHO, 2003), and central and peripheral nervous systems (Sanfeliu et al., 2003; Pinheiro et al., 2006, 2007; Crespo-Lopez et al., 2007). Furthermore, the capacity of MHg to cross the placental barrier represents a dangerous risk for proper fetal development (Guzzi, La Porta, 2008). On the other hand, exposure to metallic mercury (Hg°) and inorganic mercury (IHg), which mainly occurs in occupational environments or by releases from amalgam fillings (WHO, 2003), is usually associated with brain and kidney diseases (WHO, 1991; Clarkson and Magos, 2006). Over the past century, increasing awareness of the risks to human health associated with environmental Hg exposure (Guzzi and La Porta, 2008) was the stimulus behind intensive and specific human biomonitoring programs aimed at assessing levels of Hg exposure and environmental risks for groups of individuals living in close proximity to highly contaminated sites (Angerer et al., 2007). Once ingested, MHg is rapidly absorbed into the red blood cells, bound to hemoglobin and distributed to the tissues and brain, where it is slowly converted to IHg, probably at a rate of about 1% of the body burden per day (Clarkson and Magos, 2006; Guzzi and La Porta, 2008). As a consequence, Hg concentrations in blood (HgB) usually increase with the frequency of fish consumption, and are widely used as a tracer exposure to MHg (Wilhelm et al., 2004). Together with HgB, investigations of mercury in human hair (HgH) can also reflect MHg exposure (Airey, 1983; Matsubara and Machida, 1985; Shao et al., 2013). However, due to the high stability of the Hg incorporated in scalp hair (with a nearly stable growth rate of 1 mm/month; Phelps et al., 1980; WHO, 1990), it is generally adopted as a specific tracer in the assessment of Hg exposure on a time-scale ranging from weeks to months (JECFA, 2003). Finally, Hg in urine (HgU) is used extensively as a biological marker to assess chronic exposure to inorganic Hg, mainly in the form of Hg°, in humans (WHO, 2003; Barregard et al., 2006). This is because, once inhaled, the Hg° vapor is absorbed by the lungs and eliminated through urine and fecal excretions (Guzzi and La Porta, 2008). The petrochemical district of Priolo (SE Italy), which is delimited by the municipalities of Augusta, Melilli and Priolo, is a highly polluted area affected by the

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uncontrolled discharge of chemical pollutants from industry (Ausili et al., 2008). In particular, since the early 1960s, Augusta has hosted one of the largest chlor-alkali plant in Europe (Le Donne and Ciafani, 2008) which has discharged huge amounts of Hg into the environment. Although discharge activities were definitively stopped in 2005 (Colombo et al., 2005), the Hg contamination from the chlor-alkali plant is by far the most important environmental issue. Indeed recent investigations have denounced a severe state of degradation in the Augusta's sediment (up to 788 mg kg^{-1}) (ICRAM, 2008; Bellucci et al., 2012; Sprovieri et al., 2011; Orecchio and Polizzotto, 2013) and evasional fluxes into atmosphere (9.7 ± 0.1 g d^{-1}; Bagnato et al., 2013). Ecotoxicological investigations in the area have revealed crucial DNA damage in mussels and red mullet (Ausili et al., 2008; ICRAM, 2008) and genotoxic harm in *Coris julis* (Tomasello et al., 2012). Recently, Bonsignore et al. (2013) have reported high levels of Hg in different species of fish collected from both inside and outside Augusta Bay (up to 2.7 and 9.7 $\mu g\ g^{-1}$ in muscles and liver respectively) and the relative values of target hazard quotient (THQ) and estimated weekly intake (EWI) have advised that the consumption of fish from this area could represent a serious risk to the health of local human populations. Moreover, using Hg isotopic signatures from sediment, fish and human hair, Bonsignore et al. (2015) traced the sources and processes that transfer this contaminant from the sediment to the human population in the Augusta environment. Furthermore, significant human health concerns, including an alarming increase in spontaneous abortions, neonatal malformations and mortality rate have been denounced in this area, especially in Augusta (Madeddu et al., 2001, 2003; Bianchi et al., 2004).

The objective of this study is to assess and quantify the human exposure to Hg in the Augusta area and the key factors that influence the distribution and extent of the contamination. The Hg content in blood, urine and hair were discussed with regard to the frequency of local fish consumption, gender, age, body mass index (BMI) and education level. A toxicokinetic model was used in order to determine the effective dietary MHg intake. Moreover the estimated dietary intake previously reported for fish collected in the area (Bonsignore et al., 2013) was used to predict the expected Hg concentration in blood in individuals with exclusive local fish-based diet.

2. Materials and methods

2.1. Sample design

Randomly selected individuals living in the Augusta, Melilli and Priolo municipalities (Fig. 1) were contacted by phone and asked to participate in the biomonitoring study. A sample (~1% of the total population for each town), stratified according to the age and gender of the participants, was used to represent the entire municipality (Table 1). At the time of recruitment, 400 participants filled an extensive questionnaire to help provide a quick assessment of demographic information, health status, BMI, the frequency of seafood consumption and education level (see the details reported in the questionnaire, Supplementary material). Obesity, loss of weight (> 10 kg during the last year), cancer, and housing (for more than 10 years) in the study area were adopted as exclusion criteria for the biological sampling to prevent bias in the analysis. The frequency of fish consumption was evaluated by distinguishing between fish and shellfish origins (local vs. non-local markets). Among individuals who provided the consent for sample taking, a total of 224 individuals in the age classes 20–24, 25–29, 30–34, 35–39 and 40–44 were selected for the biological sampling (Table 1).

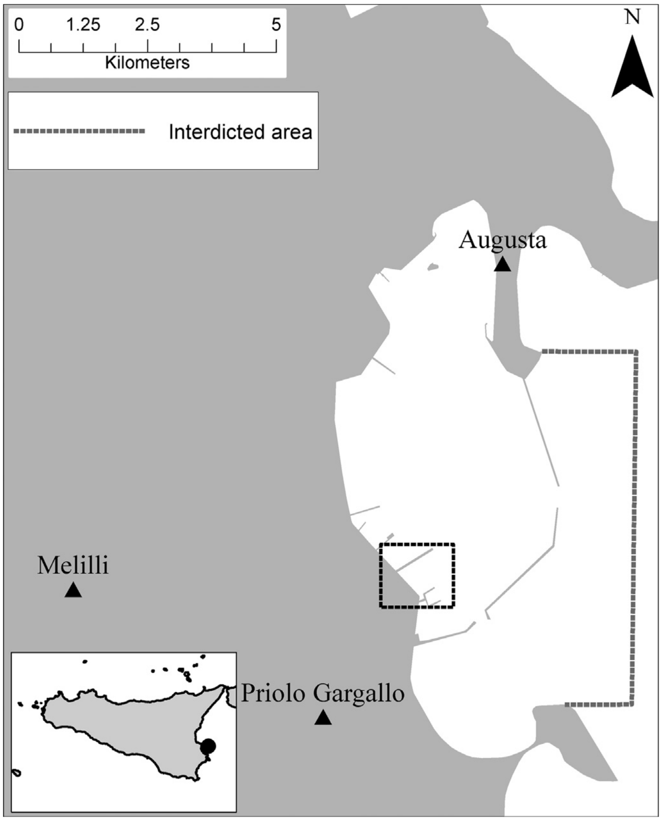

Fig. 1. Study area showing the municipalities of Augusta, Melilli and Priolo. The black dashed square indicates the chlor-alkali plant's location. The area's fishing ban is also reported with the gray dashed line.

2.2. Mercury measurements

Collection of biological samples was performed by the paramedical staff of the Public Hygiene Service (ASP N°8) of Augusta during the period within October 2012 and April 2013. Blood samples were collected in tubes containing lithium heparinate, while the first morning urine samples were deep frozen (−20 °C) until the analyses. Finally, using stainless steel scissors, a ~1 g hair sample was taken from the nape of the neck, close to the occipital region of the scalp, and kept in a clean polyethylene bag until analyses. The total Hg in the untreated blood, human hair and urine was measured by the Laboratory of Public Health of Syracuse (ASP N°8) by a Direct Mercury Analyzer (DMA80 atomic absorption spectrophotometer, Milestone, Wesleyan University, Middletown, CT, USA) according to the US EPA 7473 method (US EPA 2007).

The limit of detection of the method (LOD), estimated as three times standard deviations of the blank samples, was 0.020 μg/L blood, 0.015 μg/L urine and 0.003 μg/g hair, respectively. Accuracy was checked by running replicates of the reference materials (RM) NCS ZC 81002b (1.06 ± 0.28 μg/g), Seronorm™ Trace Elements Urine L-2 (39.8 ± 0.8 μg/L) and Seronorm™ Trace Elements Whole Blood L-2 (15.2 ± 0.8 μg/L) and L-3 (31.4 ± 1.7 μg/L). Bench quality control material was measured at the start of each analytical run (set of 20 samples) for quality assurance and control.

The measured values were, on average, within ±5% of the recommended values. In order to check the reproducibility of the analysis, about 20% of the samples were analyzed in triplicate. The coefficient of variation was between 2.4% and 3.2%.

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Table 1
Number of participants answering the questionnaire by age, gender and municipality.

	Men					Women				
Age class	20–24	25–29	30–34	35–39	40–46	20–24	25–29	30–34	35–39	40–45
Augusta	2	5	12	14	14	2	13	19	22	22
Melilli	4	5	6	3	4	5	5	5	8	3
Priolo	4	9	3	5	4	5	5	5	4	7
Total	10	19	21	22	22	12	23	29	34	32

2.3. Toxicokinetic model

We used a one-compartment toxicokinetic model (US EPA, 2001) (Eq. (1)) to calculate both the daily dietary intake at the measured levels of blood and the expected Hg concentrations in blood (μg/L) in individuals with a local fish-based diet using the estimated dietary intake of Hg previously reported by Bonsignore et al. (2013) for the same area:

$$c = \frac{d \times A \times F \times \mathrm{bw}}{b \times V} \qquad (1)$$

where:

$\boldsymbol{c}$ = average concentration in blood (μg/L).
$\boldsymbol{d}$ = daily dietary intake (μg/kg/day).
$\boldsymbol{b}$ = elimination constant (0.014 days^{-1}).
$\boldsymbol{V}$ = volume of blood in the body (5 L).
$\boldsymbol{A}$ = absorption factor (0.95).
$\boldsymbol{F}$ = fraction of absorbed dose taken up by blood (0.059).
bw = body weight (67 kg).

2.4. Statistical analysis

In a first step, specific descriptors (maximum, minimum, mean, median, 75th and 25th quartiles) were calculated to characterize the Hg levels in individuals from each municipality (Table 2). Then, because the data were right-skewed, statistical differences in the Hg levels in blood, urine and hair between the municipalities were assessed using non-parametric methods (Kruskal Wallis ANOVA). Secondly, multiple linear regression analyses were applied to the entire data set to assess the effects of diet, age, BMI, gender and level of education on the Hg concentrations in the collected blood, urine and hair samples. Specifically, factors related to food consumption were coded according to the frequency of consumption ("never/rarely" = 0; "1–2 times per week" = 1; "3–4 times per week" = 2; and "> 4 times per week" = 3), while elements related to education were ranked based on the level of instruction. Factors assuming only a double state (gender) were coded as binary factors (1: male, 0: female). Working on "raw" Hg concentrations, diagnostic regression plots evidenced the presence of trends in the "residuals vs. fitted values", as well as a skewed distribution of residuals (probably due to non-linear relationships between the response variable and predictors). The Hg concentrations in blood and hair were cubic root transformed, while log transformation was used for the urine. The significance level was set at $p < 0.05$ for all the analyses. All the statistical analyses were run in the *R* statistical environment (*R* Project for Statistical Computing; ⟨http://www.r-project.org/⟩).

3. Results

3.1. Levels of mercury in Blood, hair and urine

Table 2
Concentrations of HgB (μg/L), HgH (μg/g) and HgU (μg/L) in samples from: (a) Augusta; (b) Melilli; and (c) Priolo.

(a) Augusta						
	Mean	**Median**	**25%**	**75%**	**Min.**	**Max.**
Blood	10.1	7.30	3.80	15.3	0.20	39.7
Urine	1.67	0.95	0.50	2.1	0.10	8.40
Hair	2.61	1.90	1.08	3.44	0.04	10.5
(b) Melilli						
	Mean	Median	25%	75%	Min.	Max.
Blood	4.34	2.90	1.85	5.25	0.30	23.9
Urine	1.35	0.55	0.38	1.23	0.10	2.80
Hair	1.56	1.24	0.70	1.83	0.10	6.26
(c) Priolo						
	Mean	Median	25%	75%	Min.	Max.
Blood	4.77	3.7	1.55	6.85	0.10	22.2
Urine	1.18	0.70	0.35	1.5	0.10	6.70
Hair	1.37	1.00	0.63	1.54	0.04	7.53

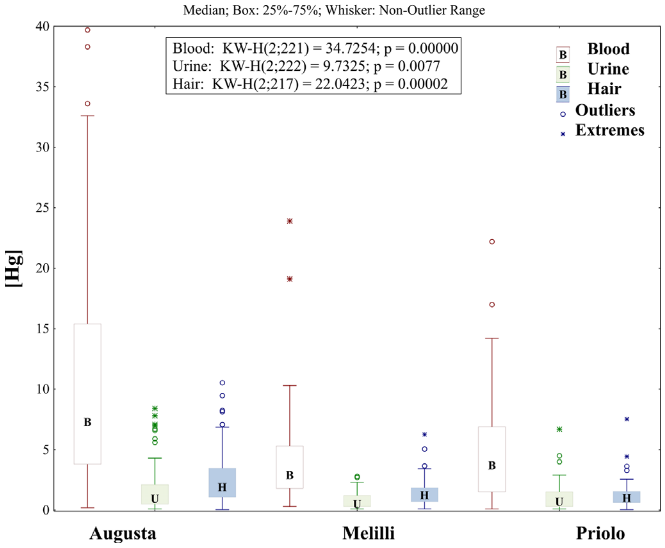

Fig. 2. Box plot of HgB (μg/L), HgU (μg/L) and HgH (H) (μg/g) for the Augusta, Melilli and Priolo samples.

The median HgB content in the entire studied population was 4.90 μg/L. Extremely high values were measured in the sample from the Augusta area (median 7.30 μg/L), while the median values were lower for those from both Melilli and Priolo (2.90 μg/L and 3.70 μg/L, respectively) (Table 2; Fig. 2). The median HgH value measured for the whole population was 1.47 μg/g. The highest concentrations were found in the Augusta residents (median 1.90 μg/g), while relatively lower values were documented in the Melilli and Priolo samples (median 1.24 and 1.00 μg/g, respectively). The median HgU (0.80 μg/L in the whole population) level was higher in the Augusta (0.95 μg/L) and lower in the Melilli (0.55 μg/L) and Priolo participants (0.70 μg/L) (Table 2; Fig. 2).

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Table 3
Amounts in percentages of local fish and shellfish consumed in the three municipalities.

		Local seafood consumption *(times a week)*			
		Rarely	**1–2**	**3–4**	**>4**
Local fish	**Augusta**	22%	58%	18%	2%
	Melilli	38%	58%	4%	0%
	Priolo	28%	63%	9%	0%
Local shellfish	**Augusta**	68%	28%	3%	1%
	Melilli	88%	12%	0%	0%
	Priolo	64%	34%	2%	0%

3.2. Information on local seafood consumption

Most of the participants (58–63%) declared to eat “local fish” (namely organisms fished from Augusta Bay) once or twice a week, with a significant no. (2–18%) doing so more than three times a week (Table 3). The consumption of local shellfish was generally less, and most of those interviewed (64–88%) rarely consumed it. Among the three municipalities, the Augusta citizens consum local fish more frequently than those from Melilli and Priolo, and just a small percentage (22%) of them declared to consume local fish rarely (Table 3).

4. Discussion

4.1. Contamination status

The measured HgB values were much higher than the reference values (RV) of the Italian population (1.68–2.40 µg/L; Human Biomonitoring Commission, 1999; Wilhelm et al., 2004; Alimonti et al., 2011; Miklavčič et al., 2013) (Fig. 3a). In ~50% of cases, the HgB levels exceeded the alert value fixed by the Human Biomonitoring Commission (1999) (HBMI = 5 µg/L), which represents the concentration below which no risk of adverse health effects occurs. In ~16% of the studied individuals, the HgB levels were higher than the action value HBMII (15 µg/L), which has been demonstrated to be the threshold of relevant risks for health consequences (Fig. 3a). Statistically significant differences ($p < 0.05$; K-W, ANOVA) were found between the levels of Hg measured in the blood of the Augusta individuals compared to those from Melilli and Priolo, suggesting a preferential exposure to Hg in the first area. Indeed, considering the distribution modes in the three municipalities, the participants from Augusta had HgB exceeding: (i) the RV in 85% of cases, (ii) the HBMI in 64% of cases and (iii) the HBMII threshold in 26% of cases, while the HgB levels in individuals from Melilli and Priolo exceeded the RV in 72% of cases, the HBMI thresholds in 31% of cases and the HBMII level in just a few situations (4%) (Fig. 3a). As for the blood samples, the HgH levels exceeded in most cases (~60%) the Italian RV (mean: 1.14 µg/g; Madeddu et al., 2004) (Fig. 3b). Based on Faroe Island and Seychelles' studies, the US Environmental Protection Agency (US EPA) adopted a reference dose (RfD) for MHg of 0.1 µg/kg of body weight/day (US EPA, 2001, 2010), which was established on the basis of the neurodevelopmental effects associated with in utero exposure and a maternal hair mercury concentration of 1.0 µg/g (NRC, 2000). More than 70% of the analyzed samples herein had HgH levels above the US EPA RfD, 35% of them exceeded the WHO “normal level” (2 µg/g), but the tolerance threshold of 10 µg/g declared by WHO (WHO 1990) was rarely overtaken (1%) (Fig. 3b). However Hg-related neuropsychological dysfunction has been observed in children even at Hg levels below 10 µg/g dry wt in maternal head hair samples from the Tapajos River basin in Brazil, the Faroe Islands, and Denmark (Grandjean

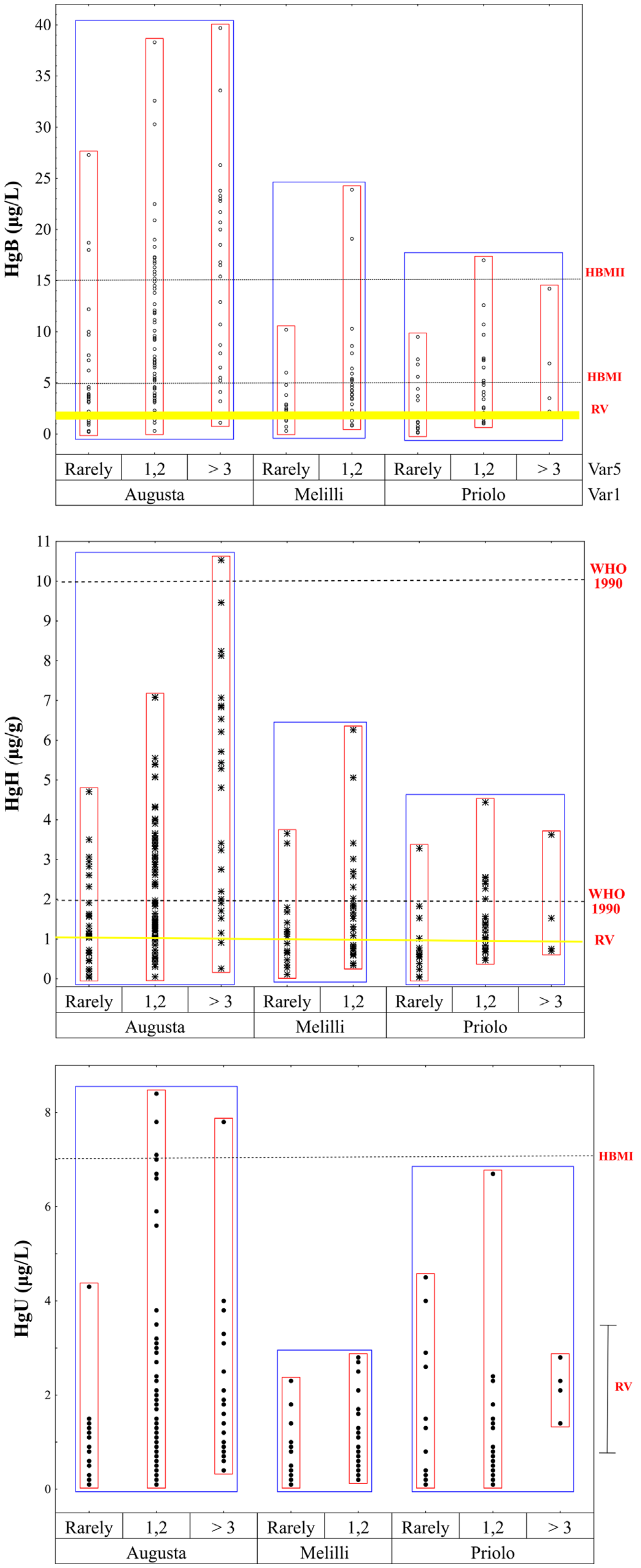

Fig. 3. Distribution of [Hg] in blood (a), human hair (b) and urine (c) from the Augusta, Melilli and Priolo samples according to the frequency of fish consumption. Reference values (RV), HBMI (the concentration below which there is no risk of adverse health effects), HBMII (the concentration above which there are increased risks of adverse health effects) and other regulation thresholds are also reported. The frequencies of local fish consumption “3–4” and “>4” times a week have been included in category >3.

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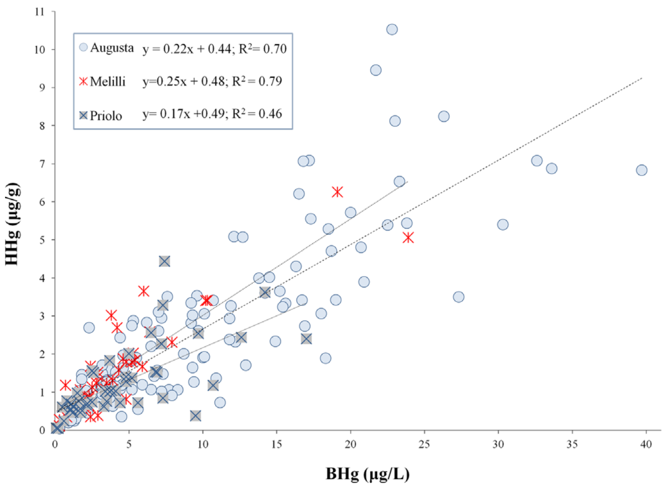

Fig. 4. Hg content in blood and human hair in the Augusta (–), Melilli (…) and Priolo (---) samples.

et al., 1997, 1999). Once again in this study the Augusta individuals contained the highest Hg levels in hair ($p < 0.05$; K-W, ANOVA), exceeding by 81% the EPA RfD and by 47% the WHO "normal level" (Fig. 3b). No remarkable anomalies were recorded for the HgU values, which were generally comparable or sometimes lower than the Italian RV (0.8–3.5 µg/L; Minoia et al., 1990; Apostoli et al., 2002a; Soleo et al., 2003; Barregard et al., 2006). The HBM I value for urine (7 µg/L) was surpassed just in five individuals from Augusta (Fig. 3c).

A statistically significant relationship was observed between the HgB and HgH values of Augusta ($r^2 = 0.70$), Melilli ($r^2 = 0.79$) and Priolo ($r^2 = 0.46$) individuals (Fig. 4). This analogous response significantly revealed that both biomarkers documented the exposure to MHg via fish consumption. Magos and Clarkson (2008) have demonstrated that human scalp hair proportionally concentrates the MHg in blood by forming a MeHg–cysteine complex. The average hair–blood ratio in humans has been estimated to be about 250:1 µg/g-mgHg/L (WHO, 1990), but in the absence of acute exposure, the concentrations of mercury are much higher in hair than in blood (estimated ratios of 370:1; Phelps et al., 1980; Shrestha and Fornerino, 1982). A calculated hair/blood ratio of 350:1 µg/g-mgHg/L suggested that the studied population suffers from a chronic exposure rather than acute effects. As mentioned above, HgU is generally used as a reliable bio-indicator of chronic exposure to inorganic Hg, which is usually associated with occupational atmospheric exposure or dental amalgam fillings.

The lack of significant anomalies in the urine samples demonstrated the absence of additional source of Hg in the form of IHg. This is in agreement with the moderate levels of atmospheric Hg measured at downtown Augusta (1.1 ± 0.3 ng m^{-3}; Bagnato et al., 2013).

4.2. Risk assessment of mercury exposure

The Fig. 3(a and b) evidenced that the highest levels of Hg in both blood and hair samples were found in individuals that consume higher amount of local fish. Specifically, the highest measured HgB value (33.6 µg/L) refers to an Augusta individual (A20; Table S1, Supplementary material) who declared to consume local fish three-four times a week, while the lowest value (0.10 µg/L) refers to a Priolo citizen who declared to "rarely" consume local fish (P37; Table S1, Supplementary material). The effect of local fish consumption on the levels of Hg in the biological samples was more evident in the Augusta dataset where individuals who frequently consume local fish ($n = 24$) have mean blood Hg contents more than twice than those doing it rarely ($n = 28$) (HgB: 14.68 ± 8.86 and 6.10 ± 6.31 µg/L respectively) (Table S1, Supplementary material). Similar considerations can be done for Hg content in hair samples, where the differences are even greater (mean HgB: 4.46 ± 2.96 and 1.49 ± 1.14 µg/L for the categories of local fish eaters "3–4 times a week" and "never/rarely") (Table S1, Supplementary material). Noteworthy, the highest HgH value (10.5 µg/L) was found in an Augusta citizen (A93; Table S1, Supplementary material) who declared to consume local fish three-four times a week, while the lowest value (0.04 µg/L) refers to subjects who "rarely" do it (A116 and P22; Table S1, Supplementary material). Averagely lower values of both HgB and HgH were measured in Melilli and Priolo subjects where the consumption of local fish is less common (Table 3). In Augusta, the effect of local fish consumption is clearly evidenced by the extreme levels of Hg measured in the blood of individuals who declared to eat local fish during the three days before samples collection (mean: 18.59 ± 9.49 µg/L; $n = 22$). In order to investigate the contribution of specific factors to the mercury levels in the blood, urine and hair samples multiple regression analysis was performed. Regression analyses were run on the overall data set without specific discrimination between the three municipalities. In fact, we were primarily interested to the macro-scale driver of Hg exposure for the residents living around Augusta Bay, and assumed that micro-scale effects at the level of a single municipality could, at this stage of the work, be minimized and were not statistically suggestive. The results showed that local fish consumption and education level were significantly associated with HgB, HgH and HgU levels, while age effects appeared to influence exclusively the level of Hg in the blood (Table 4). A significant association between gender and Hg concentrations only emerged for HgH. No significant relationship was found with BMI. Looking at standardized coefficients, it was also evident that, among the considered factors, the consumption of local fish has a major influence, suggesting that

Table 4
Output of multiple regression analyses. The standardized coefficients and p-values are reported for each model and considered factor. Significant associations ($p < 0.05$) are marked in bold.

	Blood		Urine		Hair	
	Std. coeff.	p	Std. coeff.	p	Std. coeff.	p
Fish (local market)	**0.37**	**1.44E – 07**	**0.27**	**2.85E–04**	**0.41**	**1.10E – 08**
Fish (non-local market)	0.07	0.31	– 0.12	0.11	0.05	0.43
Shellfish (local market)	0.13	0.06	0.11	0.12	0.08	0.24
Shellfish (non-local market)	0.05	0.49	**0.15**	**0.04**	0.06	0.42
Age	**0.19**	**2.93E – 03**	– 0.13	0.06	0.12	0.07
Gender	0.10	0.12	– 0.05	0.48	**0.13**	**0.05**
Educational level	**0.14**	**0.04**	**0.19**	**0.01**	**0.14**	**0.03**
BMI	– 0.10	0.12	– 0.02	0.76	– 0.10	0.14
Adj. R^2	0.285		0.1463		0.269	

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ingestion of seafood is a key and the primary driver of the increased Hg levels in blood and hair. Furthermore, a significant effect of education level is evident in all the models (Table 4). However, contrary to expectations, the relationship is positive. Indeed, a higher educational level is generally related to a greater awareness of environmental/health problems. On the other hand, taking into account that education level is usually associated with higher socio-economic conditions, as well the result suggested that a better economic status, together with limited information about the risks associated with the consumption of polluted fish, could be the most reasonable explanation for this result. Moreover, a significant influence of the age factor on blood Hg levels was found. Although this evidence has already been observed in several other studies (Wennberg et al., 2006; Schulz et al., 2007; Caldwell et al., 2009; Jenssen et al., 2012), the reasons for an age-related increase in Hg in blood are unclear and cannot be explained exclusively by diet. On the other hand, the effect of gender was evidenced in the Hg content in hair (Table 4), with men consuming slightly higher amounts of local fish than women. However, this effect was not confirmed by the Hg content measured in blood. We speculate that the effect of gender may only be appreciable over a longer time-scale, although intra-specific variations in metabolism, such as absorption rates or excretion, might also explain the observed sex-related differences (Canuel et al., 2006; Mergler et al., 2007). The adjusted R^2 were quite low in all cases (Table 4), particularly for HgU, highlighting that only part of the measured variation in the Hg content could be directly explained by the considered factors. This is in agreement with the results reported by Golding et al. (2013) who tried to quantify the contribution of the components of the maternal diet to prenatal blood Hg levels by way of a multiple regression analysis. The results obtained emphasized the complexity and generally non-linear mechanisms involved in mercury absorption. Specific diets could have a specific effect in terms of limiting Hg absorption and accumulation (Passos et al., 2003, Golding et al., 2013). Moreover, the interaction between the different factors plays an important role in determining the Hg concentrations in different tissues. Such interactions are difficult to model when limited specific factors are considered and without any prior knowledge of the biological processes involved.

4.2.1. Source of Hg exposure

Recently Bonsignore et al. (2015), using Hg isotopic signatures, suggested active mechanisms of Hg transfer from the polluted sediment of Augusta Bay to the biota compartment and, subsequently, to the local fish consumers. The application of the toxicokinetic model provided the opportunity to investigate the effective intake of Hg through fish consumption at known concentrations of Hg in the blood. On the basis of this calculation, the EWI calculated for the Augusta individuals (0.04–5.18 μg/kg of body weight) exceeded in most cases (~63%) the PTWI of 0.7 μg/kg of body weight indicated by the US EPA (2004). This figure was 33% with respect to the PTWI of 1.6 μg/kg of body weight fixed by the FAO/WHO (2006), which represents a safe value for the human population over a lifetime. The EWI in Melilli participants (0.04–3.12 μg/kg of body weight) exceeded the US EPA (2004) recommendation by ~36% and the FAO/WHO (2006) threshold by ~4. As regards the EWI of Priolo subjects (0.01–2.22 μg/kg of body weight), the US EPA (2004) and FAO/WHO (2006) limits were exceeded in ~44 and 4% of cases, respectively. Recently Bonsignore et al. (2013) calculated the THQ and EWI for fish collected inside and outside Augusta Bay, and demonstrated that a diet based on fish from the area could represent a serious risk to the health of local human populations (THQ > 1). Using the values of EWI calculated in this previous study, individuals consuming exclusively fish collected from inside the bay (range of THg: 0.25–2.64 μg/g; EWI: 1.06–11.0 μg Hg kg bw^{-1} d^{-1}) (Fig. 5) should have an Hg concentration in their blood of ~28 μg/L, which is higher than the level actually found in this research. On the other hand, an individual consuming exclusively fish from outside the bay (range of THg: 0.05–0.93 μg/g; EWI: 0.22–2.91 μg Hg kg bw^{-1} d^{-1}) (Fig. 5) should have an HgB value of ~6 μg /L, which is close to that found in Priolo and Melilli subjects, but lower than the HgB content measured in the Augusta participants. In conclusion, the levels of Hg in the blood and hair samples of the Augusta individuals probably derived from the consumption of fish from both inside and outside the bay, while those from Priolo and Melilli have a more limited exposure to mercury. The strong relationship between the levels of Hg measured in biological samples and the consumption of "local fish" (defined as organisms fished from the Augusta Bay) call for more appropriate social action to respect and possibly enlarge the fishing ban in the area.

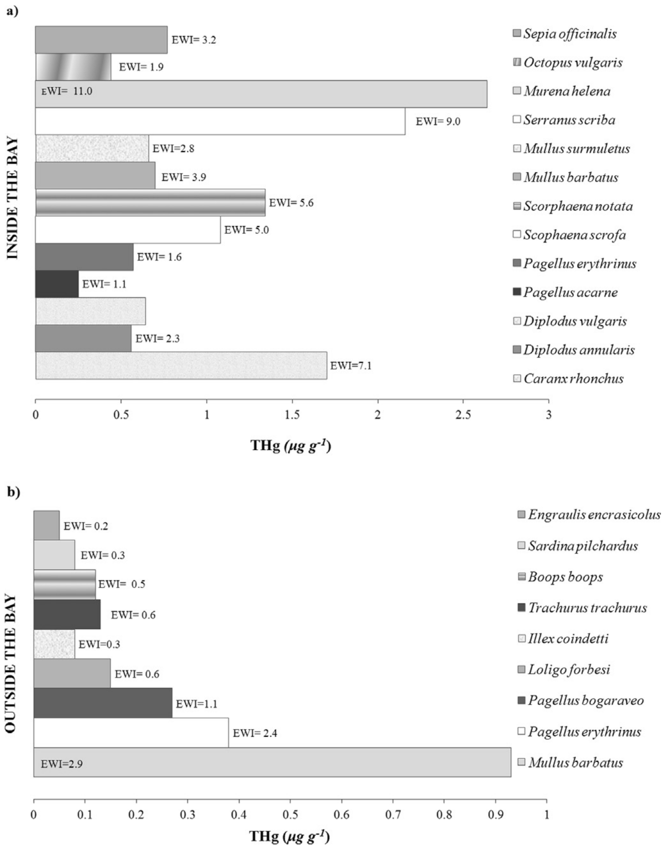

Fig. 5. Hg content measured in fishes from inside (a) and outside Augusta Bay (b) and relative values of estimated weekly intake (EWI) calculated by Bonsignore et al. (2013).

A further comparative investigation with a control group (e.g. in mountain area) could help to improve the evaluation of the human exposure to Hg in the Augusta area.

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Appendix A. Supporting information

Supplementary data associated with this article can be found in the online version at http://dx.doi.org/10.1016/j.envres.2016.01.016.

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