



Contrasting hair mercury in fishermen and workers of fish industry of Marano Lagunare (Upper Adriatic Sea), a coastal lagoon area contaminated by mining and industrial activities, against residents from the Dolomites Alps

Luca Cegolon^{a,b,*}, Stefano Covelli^c, Emilia Patriarca^a, Elisa Petranich^c, Federico Floreani^c, Donatella Sansone^a, Giuseppe Mastrangelo^d, Francesca Larese Filon^{a,e}

^a University of Trieste, Department of Medical, Surgical & Health Sciences, Trieste, Italy

^b University Health Agency Giuliano-Isontina (ASUGI), Public Health Department, Trieste, Italy

^c University of Trieste, Department of Mathematics, Informatics and Geosciences, Trieste, Italy

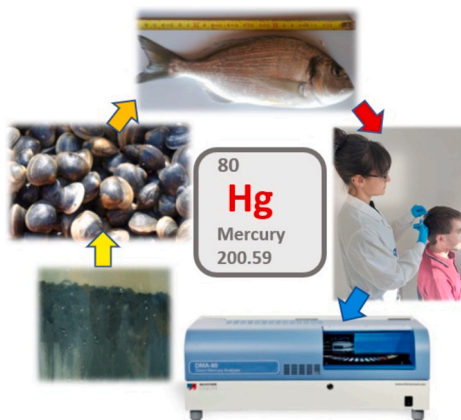
^d University of Padua, Padua, Italy

^e University Health Agency Giuliano-Isontina (ASUGI), Occupational Medicine Unit, Trieste, Italy

HIGHLIGHTS

- The Marano and Grado Lagoon is one of the coastal areas most contaminated by mercury (Hg) in the entire Mediterranean sea.
- Hair Hg concentration in fishermen and workers of fish industry of Marano Lagunare were compared with residents of the Dolomites Alps.
- Median hair Hg concentration of Alpine residents was significantly lower than fishermen and workers of fish industry of Marano Lagunare.
- Hair Hg concentration increased with fish consumption (especially >1 meal per week)
- Fishermen and fish dealers/restaurateurs exhibited significantly higher Hg concentrations

GRAPHICAL ABSTRACT



ARTICLE INFO

Editor: Xinbin Feng

Keywords:

Mercury
Methyl-mercury
Hair specimen
Fishermen

ABSTRACT

This survey aimed at estimating the concentration of hair mercury (Hg) in fishermen and workers of fish industry of Marano Lagunare (North-eastern Italy, Upper Adriatic Sea). A field investigation was conducted from 2nd of December 2023 through 18th April 2024, on 73 local fishermen, 83 workers of fish industry and 93 controls among residents (mainly farmers/herdsmen) of the Dolomites Alps. An amount of approximately 100 mg of hair was collected from all respondents, who were also asked to fill out a self-administered questionnaire collecting socio-demographic and lifestyles information. The median hair Hg concentration was 2.56 mg/kg in fishermen,

* Corresponding author at: University of Trieste, Department of Medical, Surgical & Health Sciences, Trieste, Italy.

E-mail address: luca.cegolon@units.it (L. Cegolon).

<https://doi.org/10.1016/j.scitotenv.2024.178039>

Received 20 September 2024; Received in revised form 17 November 2024; Accepted 8 December 2024

Available online 1 February 2025

0048-9697/© 2024 The Authors. Published by Elsevier B.V. This is an open access article under the CC BY-NC-ND license (<http://creativecommons.org/licenses/by-nc-nd/4.0/>).

Fish consumption
Industrial pollution

2.31 mg/kg in workers of fish industry and 0.58 mg/kg in controls. Compared with controls from the Dolomites, log-transformed hair Hg increased linearly with the amount of fish consumed (>1 meal per week), consumption of fresh fish and was significantly higher in fish dealers/fish restaurateurs and fishermen, regardless if operating on open sea or lagoon. All study groups but fish farmers and local residents involved in other business exhibited significantly higher odds of hair Hg >2 mg/kg at multiple logistic regression analysis. Whilst above the threshold background exposure recommended by WHO for the general population, the median levels of hair Hg in fishermen and workers of fish industry of Marano Lagunare were still below the cut-off of no health effects observed on human health (11.5 mg/kg). The above evidence most likely reflects contamination of lagoon bed and respective tributary river beds by sedimentary Hg from mining and industrial activities, with subsequent transfer of the metal into the aquatic trophic chain and from there to humans through consumption of local fish.

1. Background

The Gulf of Trieste (Northern Italy, Upper Adriatic Sea) and the Marano and Grado Lagoon (Fig. 1) are among the coastal areas most contaminated by mercury (Hg) across the entire Mediterranean basin [Cossa et al., 2022].

1.1. Sources

Hg contamination is primarily due to discharges from the Isonzo River, draining the mining district of Idrija (Western Slovenia), the second largest natural deposit of cinnabar (HgS) in the world after Almaden (Spain). The latter Hg environmental contamination is mainly attributable to 500 years mining activity, terminated only in 1995 [Gosar and Teršič, 2012].

In addition to the Isonzo River, industrial inputs may have also contributed to Hg contamination in coastal marine sediments of the Upper Adriatic Sea, including Trieste port with the respective industrial area and former SNIA-Viscosa complex, a large chlor-alkali plant (CAP) located in Torviscosa - in the hinterland close to the central sector of the Marano and Grado Lagoon (Fig. 1) [Acquavita et al., 2012]. The latter factory, dedicated to the production of cellulose, chlor-alkali and textile fibers since the 1940s, used Hg as a catalyst, releasing it uncontrollably into the Aussa-Corno river system until 1984 through the Banduzzi artificial tributary canal. The Aussa-Corno river system flows into the central sector of the lagoon [Piani et al., 2005], where it is estimated that approximately 186 t (up to a maximum of 20 kg/day) of Hg have been deposited since 1949, the year when SNIA Viscosa CAP started to operate [Piani et al., 2005]. Only in 1984 were industrial waste filter and control systems implemented by SNIA Viscosa [Piani et al., 2005]. Soda production by SNIA Viscosa increased from 4500 tonnes in 1950 to 20,000 tonnes in the following decade [RFVG, 1991]. Although uncontrolled discharges of Hg from SNIA Viscosa CAP definitively ceased in 2008, the Banduzzi canal, connecting the latter industrial site to the Aussa River, still serves as an active source of Hg deposited on the respective river beds [Covelli et al., 2009].

Furthermore, due to strong Hg environmental contamination, former SNIA Viscosa CAP buildings still contributes as a secondary source of atmospheric Hg for downwind environments located towards the Marano and Grado Lagoon [Acquavita et al., 2017; Floreani et al., 2020].

1.2. Hg concentration

Previous research reported continued supply of Hg-enriched sediments to the central sector of the lagoon by the Isonzo river system [Covelli et al., 2007], despite active anthropogenic sources of Hg (Idrija Hg mine and SNIA Viscosa CAP) being suppressed several years ago. Progressively lower concentrations of Hg were detected in the coastal sediments from the eastern sector of the lagoon (>11 mg/kg) – near Grado town and the mouth of the Isonzo River – towards the western sector (0.7 mg/kg), with an average concentration of ~5 mg/kg in the central lagoon area [Acquavita et al., 2012; Bettoso et al., 2023]. Mercury in the central sector of the lagoon is present both as cinnabar compounds (HgS), mainly deriving from the inputs of Idrija mining

deposits through the Isonzo River, and non-cinnabar compounds (non-HgS), likely due to the activity of SNIA Viscosa CAP, a clear mixing due to tidal dynamics [Piani et al., 2005]. Even before SNIA-Viscosa CAP stopped the production of chlor-alkali in 2008, Hg contamination in the central sector of the lagoon was mainly attributable to HgS enriched suspensions from the Isonzo River, whereas close to the mouth of Aussa-Corno River up to 98% total Hg at the bottom sediments was found to be present as non-HgS forms [Piani et al., 2005; Covelli et al., 2009]. The outflow waters of Aussa-Corno river system into the central sector of the lagoon exhibited rather high levels of Hg associated with sediment suspended particles, estimated to be 20 µg/g in the tributary Banduzzi canal compared to dissolved Hg (52.4–4.1 ng/L) [Covelli et al., 2009]. More recent investigations on the entire lagoon basin confirmed that, although varying from 1.6 to 28.7 ng/L, with a mean of 6.6 ± 5.4 ng/L, dissolved Hg concentrations in surface waters were low and never exceeded the limit set by WFD/2000/60/CE (SQA-CMA=70 ng/L) [Bettoso et al., 2023], endorsing the hypothesis of limited mobility of the metal from sediment to the upper water column.

1.3. Methylation of inorganic Hg

The detrimental effects of Hg on human health are mainly due to methyl-mercury (MeHg), an organic species forming from methylation of inorganic Hg under anoxic conditions [Compeau and Bartha, 1985; Kierfve, 1994; King et al., 2022; Lacerda et al., 1992; Gilmour et al., 1992; King et al., 1999; Hines et al., 2012]. Mercury methylation in the environment occurs mainly through biotic pathways mediated by various anaerobic and archaea bacteria characterised by *hgcA* and *hgcB* genes [Parks et al., 2013; Podar et al., 2015]. The latter gene clusters are reliable markers for presence of Hg methylating bacterial species in the environment, therefore useful to predict Hg methylation [Bravo et al., 2018; Vishnivetskaya et al., 2018; Tang et al., 2020]. However, in addition to microbial activity, Hg methylation in aquatic environments is largely influenced also by bioavailable Hg chemical species [Hsu-Kim et al., 2013].

1.4. Hg level in fish

In the Marano-Grado Lagoon, Hg levels in fishes often exceeded the commercial limits (0.5 mg/kg wet weight, ww) set for many species by the Commission of the European Communities [Eu, 2006], especially in the Grado area (Eastern sector of the lagoon), with Hg concentration varying by size and habits of fish species [Bettoso et al., 2023]. However, Bettoso et al. affirmed that the computed biota sediment accumulation factor (BSAF) points at low Hg bioavailability by transfer of Hg from sediments to biota, thereby explaining why some lagoon areas are almost certainly safe from bioaccumulation despite high Hg contamination of the respective sediments [Bettoso et al., 2023].

According to previous studies, grass goby (*Zosterisessor ophiocephalus*), a valuable and traditional resource of artisanal fisheries in this lagoon environment, exhibited critical levels of Hg in edible pulp, with average concentrations exceeding the limit set for human consumption [Acquavita and Bettoso, 2018].

Since it varies greatly, averaging around 1.0, the Se:Hg molar ratio is

unlikely to provide total protection from Hg assumed by dietary intake. The Selenium-Health Benefit Value index (HBVSe) suggests that a precautionary principle must prevail, for example, with the integration of foods rich in Se. However, according to the Provisional Tolerable Weekly Intake (PTWI), local residents of the Gulf of Trieste can comfortably have five fish meals per week, or roughly 150 g, without encountering any health risk.

More recently, Hg concentrations in seawater, sediments, plankton and benthic rays - apex predators in the Gulf of Trieste — were investigated as markers of Hg environmental sea contamination. The weight and age of benthic ray species were found to be correlated with Hg muscle concentration reaching up to 4.40 µg/g dw (dry weight) [Faganeli et al., 2018]. However, it seems likely that efficient MeHg demethylation in sediments along with water column Hg(II) reduction are relevant factors preventing extensive contamination of marine biota

in the Gulf of Trieste [Faganeli et al., 2014].

1.5. Human exposure to Hg

Marano Lagunare, a town located on the respective lagoon sector, 8 km further West from the mouth of Aussa-Corno river (Fig. 1), is a municipality whose economy largely depends on fishing, as there are 140 registered professional fishermen. While in the period April - September the predominant fishing activity is in the open sea, in autumn and early spring local boats (“batèle”) operating in the lagoon prevail [RFVG 2014].

Fishermen (and family members) represent a category at risk for Hg exposure since they tend to consume on average more fish than the general population, due to higher access to fish for their job and the area where they live [Giangrosso et al., 2016; Okati and Esmaili-Sari, 2018a;

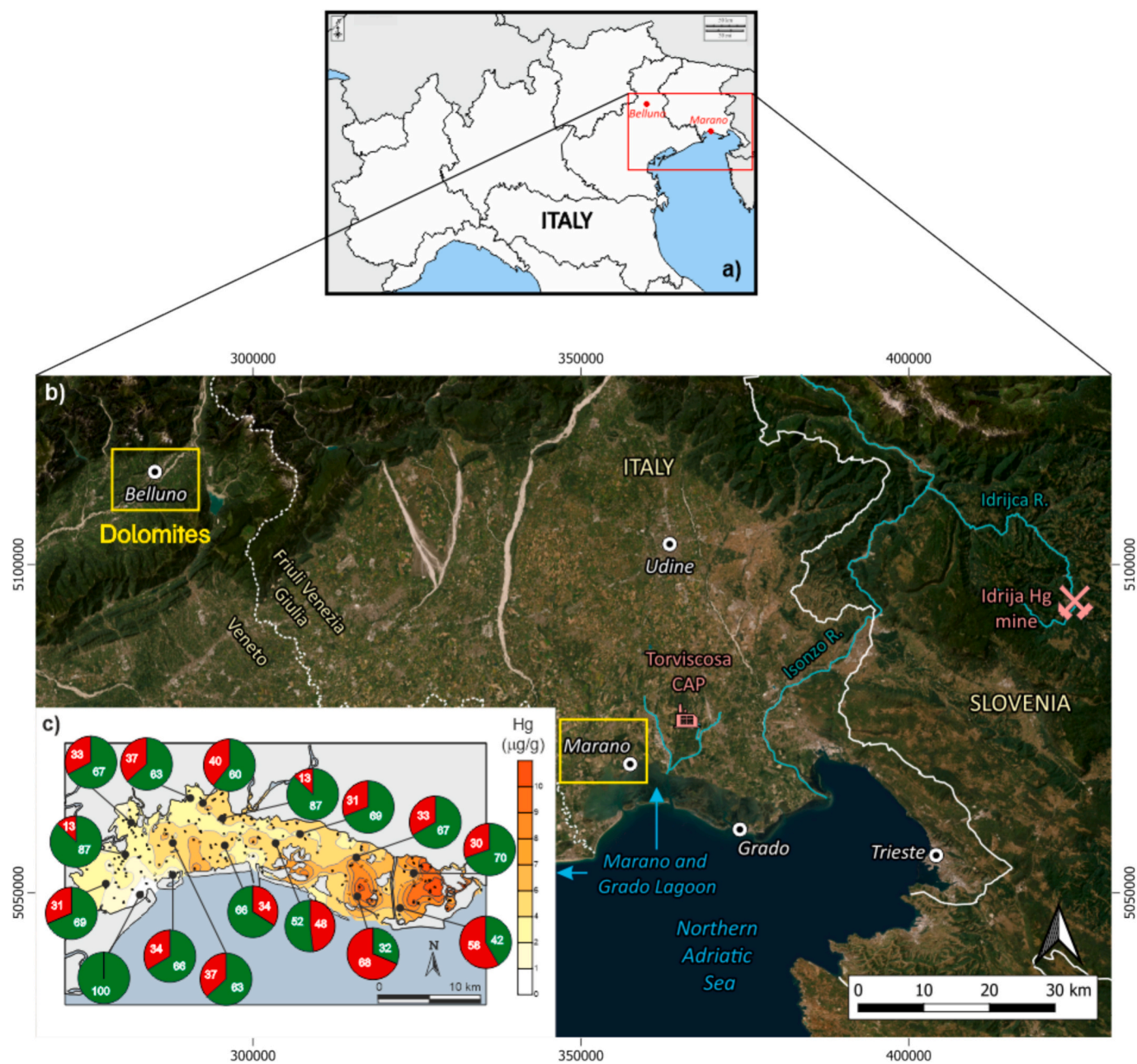


Fig. 1. a) Geographical location of Belluno (Italian Dolomites Alps) and Marano; b) Index map at regional scale of the study area that includes the Marano and Grado Lagoon and the control area of Belluno (Italian Dolomites Alps); c) Distribution of Hg concentrations along with percentage of Hg species (HgS, in red, and non-HgS, in green) in bottom sediments of the Marano and Grado Lagoon (redrawn from Acquavita et al., 2012).

Gaggi et al., 1996; Renzoni, 1992; Matthews, 1983; Bacci et al., 1976]. For example, high average concentrations of hair Hg (6.45 ± 7.03 mg/kg) were detected in 96 Sicilian fishermen [Giangrosso et al., 2016], in 58 Portuguese fishermen from Madeira (39.76 mg/kg) [Gaggi et al., 1996] and in family members of 47 Iranian fishermen (5.76 mg/kg) [Okati and Esmaili-Sari, 2018b].

1.6. Study aim

In view of the above, this cross-sectional study was planned to investigate the extent of difference in hair Hg concentration among fishermen and workers of fish industry of Marano Lagunare, a coastal lagoon area with established environmental mercury contamination due to mining and industrial activities, against residents of a low-risk area located in the Dolomites Alps as controls.

2. Methods

This study was approved by the ethical committee of FVG (CEUR n.092/2018, 23.03.2018) and the Ethic Committee of the University of Trieste (Prot. N. 11; 17/12/2024); written informed consent from each study participant.

2.1. Field investigation

The survey in Marano area was conducted from 2nd of December 2023 through 18th April 2024, by conducting multiple field investigations at the local fish market, in a local private company selling fish, in two local fish farms and through subsequent referrals among local residents connected with the fish industry.

Seventy-three out of 140 total professional fishermen registered in Marano Lagunare were recruited by convenience sampling, for a 52.1 % response rate. Fishermen were broken down by those operating exclusively in open sea ($N = 32$) against those operating in open sea in addition to lagoon (here defined as “mixed fishermen”) or exclusively in lagoon ($N = 41$). Eighty-one additional local residents were recruited, including 55 workers of the fish industry, 8 fish farmers - recruited from two local companies farming lagoon sea basses and gilt-head breams -, 4 retired fishermen, 3 household members of fishermen and 11 local residents connected with the fish industry.

Ninety-three controls were selected and surveyed among residents (mainly farmers and herdsmen) of the Dolomites Alps, during informative and training meetings on infection prevention and controls organized by the Public Health Department of the Local Health Authority N 1 “Dolomiti” of Belluno. Meetings were held on 4th April, 6th April and 10th April in Sedico and on 11th April in Pieve di Cadore (both located in the province of Belluno).

Study subjects were further broken down by 9 categories:

- Controls from the Dolomites Alps ($N = 93$);
- Open sea fishermen ($N = 32$);
- 30 lagoon fishermen and 11 fishermen operating both in lagoon as well as open sea (mixed fishermen) (Tot $N = 41$);
- Administrative staff of fish industry ($N = 18$);
- 3 fish dealers and 23 fish restaurateurs (Tot $N = 26$);
- Storers/porters/carriers/vendors referring to local fish markets ($N = 11$);
- Local residents involved in other business yet connected with the fish industry ($N = 7$);
- 5 retired (open sea) fishermen and 6 household members of fishermen (wives, partners) of fishermen (Tot $N = 10$);
- Local fish farmers ($N = 10$).

2.2. The survey

An amount of approximately 100 mg of hair was collected with

stainless steel scissors from the occipital scalp of each participant. Hair samples were then labelled and stored separately in individual polyethylene bags until analysis.

All participants also filled out a self-administered questionnaire collecting the following information: (Supplementary Table S1):

- Area of residence (urban, industrial, urban)
- Presence and type of industrial activities near the house;
- Any hobbies and free time activities;
- Number of dental amalgams present at the time of the survey
- Number of dental amalgams removed in last 2 months;
- Nasal dyspnoea (Yes vs. No);
- Bruxism (Yes vs. No),
- Daily amount of water intake (liters per day);
- Usual source of drinking water (tap, well, bottled);
- Chewing-gum consumption (Yes vs. No)
- Alcohol consumption (No vs, $\leq \frac{1}{2}$ liter/die vs. $> \frac{1}{2}$ liter/die);
- Cigarette smoking (current smoker, never smoker; ex-smoker quitting >1 year before);
- Use of dietary supplements (Yes vs, No);
- Use of skin creams (Yes vs, No),
- Use of contact lenses (Yes vs, No);
- History of kidney disease (Yes vs. No);
- Number of meals containing fish eaten per week,
- Status of fish usually consumed (fresh, frozen, canned).
- Type of fish usually consumed

Three main fish species were considered:

- **Big size fish:** swordfish, tuna, cod;
- **Small/medium size:** anchovies, sardines, sea bass, sea bream, ribbon, gilt-head bream, grey mullet, mullet, plaice, conger
- **Shellfish/crayfish/ mollusks:** scampi, prawn, shrimp, sea cicada, crab, lobster, sea crayfish, clams, mussel, squids, cuttlefish, octopus

Total Hg in hair samples was measured using a Direct Mercury Analyzer (DMA-80, Milestone, Sorisole, Italy), according to the EPA Method 7473 (U.S. EPA, 1998). The limit of detection (LOD) was equal to 0.004 mg/kg, calculated as three times the standard deviation coming from the average of 10 blanks divided by the slope of the calibration curve. The accuracy of the method for the analytical determination of Hg was verified by analysing a certified reference material (CRM) for a better representation of the results:

- ERM-DB001 (0.365 ± 0.028 mg/kg, Human Hair CRM, European Reference Materials).
- Acceptable recoveries were obtained, ranging between 98 and 106 %.

2.3. Statistical analysis

Continuous variables - presented as mean \pm standard deviation (SD), median and interquartile range (IQR) - were contrasted by Mann Whitney test. Categorical variables were presented as number and percentage and contrasted by chi square test.

Since exhibiting a skewed distribution, hair Hg was log transformed for the analysis to increase symmetry, achieving a distribution non-significantly different from normality ($p = 0.606$).

Following a backward stepwise procedure (using a $p < 0.05$ selection criteria), a multivariable linear regression model on log-transformed hair Hg was fitted onto all variables displayed in Table 1, expressing the results as adjusted regression coefficients (aRC) with 95 % confidence intervals (95%CI). Moreover, a multiple logistic regression model was fitted to investigate the odds of hair Hg concentration > 2 vs. ≤ 2 mg/kg (binary endpoint), again selecting variables by backward stepwise procedure using a $p < 0.05$ selection criterium and expressing the results as adjusted odds ratio (aOR) with 95%CI.

Table 1

Distribution of variables by study group. Number (N); column percentage (%); mean \pm standard deviation (SD); median; interquartile range (IQR); chi square test p value; Mann Whitney test p-value; M = missing values.

Terms	Strata	Total	Controls (N = 93)	Workers of fish industry (N = 83)	P-value	Fishermen (N = 73)	P-value
Sex	Female	60 (24.1)	39 (41.9)	20 (24.1)	0.012	1 (1.4)	<0.001
	Male	189 (75.9)	54 (58.1)	63 (75.9)		72 (98.6)	
Age (years)	M \pm SD	52.4 \pm 13.5	49.4 \pm 15.0	53.2 \pm 12.8	0.051	55.4 \pm 11.5	0.006
	Median (IQR)	54.4 (42.7; 62.4)	48.7 (38.2; 60.9)	54.9 (47.4; 62.4)		58.0 (48.3; 62.6)	
BMI (Kg/m ²) (M: 2)	< 43	58 (23.3)	31 (32.6)	15 (18.1)	0.101	12 (16.4)	0.067
	43–54	61 (24.5)	22 (23.2)	22 (26.5)		17 (23.3)	
	55–62	62 (24.9)	17 (17.9)	24 (28.9)		21 (28.8)	
	63+	68 (27.3)	23 (24.7)	22 (26.5)		23 (31.5)	
	M \pm SD	25.4 \pm 3.7	24.5 \pm 3.8	25.4 \pm 3.4		26.7 \pm 3.6	
	Median (IQR)	25.5 (22.5; 27.8)	23.6 (21.5; 27.4)	25.3 (23.0; 27.3)		26.5 (24.1; 28.4)	
Amalgam dental fillings (number)	< 25	114 (46.2)	54 (58.7)	36 (43.9)	0.092	24 (32.9)	0.004
	25–29	106 (42.9)	29 (31.5)	39 (47.6)		38 (52.1)	
Smoking status (M:1) (cigarettes number)	30+	27 (10.9)	9 (9.8)	7 (8.5)	0.006	11 (15.1)	0.477
	Median (IQR)	1 (0; 4)	2 (0; 4)	0 (0; 2.3)		1 (0; 4)	
Smoking status (M:1) (cigarettes number)	Never smoker	137 (55.2)	68 (73.1)	41 (49.4)	<0.001	28 (38.9)	<0.001
	Current smoker	33 (13.3)	16 (17.2)	9 (10.8)		8 (11.1)	
	< 16/ day	29 (11.7)	4 (4.3)	6 (7.2)		19 (26.4)	
	16+/ day	49 (19.8)	5 (5.4)	27 (32.5)		17 (23.6)	
Water daily intake (liters)	Ex-smoker	49 (19.8)	5 (5.4)	27 (32.5)	0.947	17 (23.6)	0.382
	M \pm SD	1.6 \pm 0.8	1.6 \pm 0.7	1.5 \pm 0.6		1.7 \pm 0.9	
Water daily intake (liters)	Median (IQR)	1.5 (1.0; 2.0)	1.5 (1.0; 2.0)	1.5 (1.0; 2.0)	0.357	1.5 (1.0; 2.0)	0.457
	Up to 1	94 (40.0)	35 (40.2)	33 (41.8)		26 (37.7)	
	1–1.5	45 (19.2)	21 (24.1)	13 (16.5)		11 (15.9)	
	1.5–2	65 (27.7)	20 (23.0)	26 (32.9)		19 (27.5)	
	>2	31 (13.2)	11 (12.6)	7 (8.9)		13 (18.8)	
Source of water intake	Bottled	No	101 (43.7)	63 (75.0)	<0.001	18 (25.4)	<0.001
	Yes	130 (56.3)	21 (25.0)	53 (73.7)		53 (74.7)	
Tap	No	122 (52.8)	15 (17.9)	58 (76.3)	<0.001	49 (69.0)	<0.001
	Yes	109 (47.2)	69 (82.1)	18 (23.7)		22 (31.0)	
Well	No	214 (92.6)	82 (97.6)	64 (84.2)	0.003	68 (95.8)	0.517
	Yes	17 (7.4)	2 (2.4)	12 (15.8)		3 (4.2)	
Daily wine intake (liters) (M:2)	0	94 (38.1)	50 (53.8)	25 (30.9)	0.002	19 (26.0)	0.001
	< 0.5	137 (55.4)	39 (41.9)	55 (67.9)		43 (58.9)	
Bruxism ^a	0.5+	16 (6.5)	4 (4.3)	1 (1.2)	0.732	11 (15.1)	0.342
	No	203 (81.5)	78 (83.9)	68 (81.9)		57 (78.1)	
Chewing gum habit (M: 2)	Yes	46 (18.5)	15 (16.1)	15 (18.1)	0.160	16 (21.9)	0.216
	No	210 (85.0)	79 (85.0)	69 (84.2)		62 (86.1)	
Occasional	No	32 (13.0)	14 (15.1)	10 (12.2)	0.158	8 (11.1)	0.125
	Usual	5 (2.0)	0	3 (3.7)		2 (2.8)	
Nasal dyspnea	No	213 (85.5)	84 (90.3)	69 (83.1)	0.158	60 (82.2)	0.125
	Yes	36 (14.5)	9 (9.7)	14 (16.9)		13 (17.8)	
Supplements	No	186 (74.7)	80 (86.0)	50 (60.2)	<0.001	56 (76.7)	0.122
	Yes	63 (25.3)	13 (14.0)	33 (39.7)		17 (23.3)	
Use of contact of lenses	No	231 (92.8)	86 (92.5)	75 (90.4)	0.616	70 (95.9)	0.358
	Yes	18 (7.2)	7 (7.5)	8 (9.6)		3 (4.1)	
Skin cream use	No	158 (63.4)	65 (69.9)	43 (51.8)	0.014	50 (68.5)	0.846
	Yes	91 (36.6)	28 (31.5)	40 (48.2)		23 (31.5)	
Weekly fish meals (Number)	M \pm SD	1.4 \pm 1.0	0.9 \pm 0.6	1.9 \pm 1.1	<0.001	1.5 \pm 1.0	<0.001
	Median (IQR)	1 (0.75; 2.0)	1 (0.5; 1)	2 (1; 2.5)		1 (0.75; 2.0)	
Status of fish consumed	< 0.75	76 (30.5)	45 (48.4)	12 (14.5)	<0.001	19 (26.0)	<0.001
	0.75–1	80 (32.1)	33 (35.5)	22 (26.5)		25 (34.3)	
	1–2	55 (22.1)	14 (15.1)	28 (33.7)		13 (17.8)	
	3+	38 (15.3)	1 (1.1)	21 (25.3)		16 (21.9)	
	No	51 (20.5)	35 (37.6)	11 (13.6)		5 (6.9)	
Fresh	Yes	198 (79.5)	58 (62.4)	72 (86.8)	0.005	68 (93.1)	<0.001
	No	195 (78.3)	57 (61.3)	67 (80.7)		71 (97.3)	
Frozen	Yes	54 (21.7)	36 (38.7)	16 (19.3)	0.003	2 (2.7)	0.106
	No	211 (84.7)	71 (76.3)	77 (92.8)		63 (86.3)	
Canned	Yes	38 (15.3)	22 (23.7)	6 (7.2)	0.996	10 (13.7)	0.004
	No	100 (40.3)	31 (33.7)	28 (33.7)		41 (56.2)	
Big size	Yes	148 (59.7)	61 (66.3)	55 (66.3)	0.004	32 (43.8)	0.005
	No	36 (14.9)	23 (25.0)	7 (8.4)		6 (8.2)	
Small/medium size	Yes	212 (85.1)	69 (75.0)	76 (91.6)	<0.001	67 (91.8)	<0.001
	No	61 (24.6)	41 (44.6)	11 (13.3)		9 (12.3)	
Shell/cray fish/ mollusks	No	61 (24.6)	41 (44.6)	11 (13.3)	<0.001	9 (12.3)	<0.001
	Yes	187 (75.4)	51 (55.4)	72 (86.8)		64 (87.7)	

^a Condition in which a person grinds, gnashes or clenches teeth unconsciously when (s)he is awake or sleeping.

In both above regression models the Benjamini-Hochberg (BH) procedure was subsequently applied to contain the role of chance, setting the false discovery rate (FDR) at 5 %.

Stata 14.1 (Stata corporation, College Station, Texas, USA) was used for the analysis.

3. Results

The distribution of variables by study group is shown in Table 1, where 73 active local fishermen were compared against 83 workers of fish industry and 93 controls from the Dolomites Alps. The group of 83 workers of fish industry included the above 11 clerks of fish industry, the 23 fish dealers/fish restaurateurs, the 11 workers of local fish markets, the 7 local residents involved in other business connected with the fish industry, the 11 retired (open sea) fishermen/fishermen household members and the 10 local fish farmers. Each *p* value provides the statistical significance of chi square test between workers of fish industry or fishermen against controls. The proportion of females was significantly higher among controls, whereas fishermen and workers of fish industry were by far composed of males (Table 1).

Controls were also younger (median age = 49.0 years) compared to workers of fish industry (54.9 years) or fishermen (58.0 years), although age difference was significant only between fishermen and controls (*p* = 0.006). The proportion of individuals younger than 43 was higher among controls (32.6 %), whereas the proportion of respondents aged >55 years was higher among workers of fish industry and fishermen of Marano, although differences were not statistically different.

From Table 1 it can be noted that the median BMI was significantly higher (*p* < 0.001) in fishermen (26.5 kg/m²) compared to controls (23.6 kg/m²) and the median number of amalgam dental fillings was significantly lower (*p* = 0.006) in workers of fish industry (median = 0) compared to controls (median = 2).

The distribution of smoking status was significantly different between study groups (*p* < 0.001), with the proportion of never smokers decreasing from 73.1 % in controls, to 49.4 % in workers of fish industry and 38.9 % among fishermen. The proportion of ex-smokers was higher among workers of fish industry (32.5 %), whereas current smokers were more prevalent in fishermen (37.5 %), with 26.4 % of them smoking 16+ cigarettes per day (Table 1).

Regarding the source of water intake, workers of fish industry (73.7 %) and fishermen (74.7 %) drank more frequently bottled water compared to controls (25.0 %), who consumed predominantly tap water (81.4 %). Moreover, workers of fish industry exhibited higher proportion of well water intake (15.8 %) (Table 1).

A significantly higher proportion of controls did not drink wine (53.8 %; *p* < 0.001), whereas the percentage of those drinking wine <0.5 L/day was higher in workers of fish industry (69.6 %) and the proportion of those drinking wine 0.5+ L/days was higher among fishermen (15.1 %).

Compared to controls (16.9 %), a significantly higher proportion of workers of fish industry used supplements (39.7 %, *p* < 0.001), or skin creams (48.2 %, *p* = 0.017).

As can be noted from Table 1, the median weekly fish intake was significantly higher in workers of fish industry (*p* < 0.001) as well as in fishermen (*p* < 0.001) compared to controls. In particular, the proportion of respondents eating fish 3+ times per week was 1.1 % among controls against 25.3 % in workers of fish industry and 21.9 % in fishermen (Table 1).

Consumption of fresh fish was significantly lower in controls (62.4 %) than workers of fish industry (86.8 %; *p* < 0.001) and fishermen (93.1 %; *p* < 0.001). By contrast, consumption of frozen fish was significantly higher in controls (38.7 %) compared to workers of fish industry (19.3 %; *p* = 0.005) and fishermen (2.7 %; *p* < 0.001). Likewise, intake of canned fish was higher controls (23.7 %) than workers of fish industry (7.2 %; *p* = 0.003) or fishermen (13.7 %; *p* = 0.106).

Consumption of big size fishes was more prevalent in controls (66.3

%) or workers of fish industry (66.3 %) compared to fishermen (43.8 %; *p* = 0.004). Conversely, intake of small/medium size fishes was significantly higher in fishermen (91.8 %; *p* = 0.005) or workers of fish industry (91.4 %; *p* = 0.004) compared to controls (75.0 %). Likewise, consumption of shell/crayfish/mollusks was significantly more prevalent in fishermen (87.7 %; *p* < 0.001) or workers of industry (88.9 %; *p* < 0.001) than controls (54.4 %) (Table 1).

Table 2 displays the distribution of hair Hg by study group and explanatory variables of Table 1. The total median hair Hg in the entire sample was 1.44 (IQR: 0.61; 2.97) mg/kg, with a mean of 2.56 ± 3.16 mg/kg. The median hair Hg concentration was 2.56 (IQR: 1.45; 4.55) mg/kg in fishermen (mean = 3.82 ± 3.97 mg/kg), and 2.32 (IQR: 1.39; 4.76) mg/kg in workers of fish industry (3.51 ± 3.13 mg/kg), against 0.58 (IQR: 0.32; 0.98) mg/kg in controls from the Dolomites Alps (mean = 0.71 ± 0.55 mg/kg). Considering the entire Marano sample (*N* = 156), comprising both fishermen as well as workers of industry, the median hair Hg was 2.39 (IQR: 1.44; 4.66) mg/kg (mean = 3.66 ± 3.54 mg/kg). The median hair level in controls was significantly lower than fishermen (*p* < 0.001) and workers of fish industry of Marano (*p* < 0.001). With few stratum specific exceptions, the latter pattern was consistently confirmed across all explanatory factors (Table 2).

Table 3, Fig. 2a-2b and Fig. 3 show the median and IQR of hair Hg by study group, while the distribution of weekly fish meals by study group can be viewed in Table 3 and Fig. 4. As can be noted, the median hair Hg concentration was higher in open sea fishermen (median 2.33; IQR: 1.41; 3.89), lagoon/mixed fishermen (median = 2.92; IQR: 1.81; 4.93) and fish dealer/fish restaurateurs (Median = 5.59; IQR: 2.66; 7.85) (Table 3). However, lagoon/mixed fishermen exhibited more extreme range values (min = 0.42; max = 20.44) compared to fish dealers/fish restaurateurs (min = 0.75; max = 13.97) or open sea fishermen (min = 0.18; max = 7.47) (Table 3 and Fig. 3).

The median number of weekly fish meals was higher in fish dealers/fish restaurateurs (median = 2.75; IQR: 1.5; 3), followed by local residents involved in other business (median = 2; IQR: 1; 3), fish farmers (median = 2; IQR: 1; 2) and storers/porters/carriers/vendors of local fish markets (median = 2; IQR: 1; 2) (Table 3 and Fig. 4). Likewise, the proportion of respondents eating fish >2 times per week was higher among fish dealers/fish restaurateurs (53.9 %) and local residents involved in other business yet connected with the fish industry (42.9 %), followed by lagoon/mixed (24.4 %) or open sea fishermen (18.8 %) (Table 3 and Fig. 4).

Extremely high hair Hg concentrations were found in lagoon fishermen (max value: 20.44 mg/kg), fish dealers/fish restaurateurs (max value: 13.97 mg/kg), followed by administrative staff of fish industry (max value: 11.15 mg/kg) (Table 3 and Fig. 3). Six lagoon/mixed fishermen exhibited hair Hg concentrations >10 mg/kg, five of whom reportedly having an average of 2 fish meals per week and one 3 weekly fish meals. Among fish dealers/fish restaurateurs, four respondents exhibited hair Hg >10 mg/kg, one having 4 fish meals per week, one 3, one 2 and one 1.5 weekly fish meals. The only administrative clerk with a concentration of hair Hg >10 mg/kg consumed 2 weekly fish meals (data not shown in tables).

The vast majority of study participants usually ate fresh fish, although the proportion of those eating frozen fish was higher among controls (38.7 %) and administrative staff of fish industry (38.9 %) (Table 4). The proportion of study subjects eating usually big fishes was lower in lagoon/mixed fishermen (36.6 %), retired fishermen/household members of fishermen (27.3 %) and fish farmers (37.5 %). By contrast, a lower proportion of controls usually ate small/medium size fishes (75.0 %) or shell-crayfish/mollusks (55.4 %) (Table 4). For a better visualization of data, bar charts for the usual consumption of fresh, frozen or canned fish by detailed study groups are reported in Supplementary files S2a-S2c. Likewise, bar charts for the usual intake of big size fishes, small/medium size fishes and shell/crayfish/mollusks are displayed in Supplementary files S3a-S3c.

A scatterplot for the distribution of log transformed hair Hg

Table 2

Concentration of hair mercury (mg/Kg) by explanatory factors. Median with interquartile range (IQR). Mann-Whitney test p value. Mean (M) ± standard deviation (SD).

Terms	Strata	All sample (N = 249)	Controls (N = 93)	Workers of fish industry (N = 83)	P value	Fishermen (N = 73)	P value
Median hair mercury		1.44 (0.61; 2.97)	0.58 (0.32; 0.98)	2.32 (1.39; 4.76)	<0.001	2.56 (1.45; 4.55)	<0.001
M ± SD		2.56 ± 3.16	0.71 ± 0.55	3.51 ± 3.13		3.82 ± 3.97	
Sex	Females	0.80 (0.40; 1.60)	0.52 (0.30; 0.93)	1.74 (1.33; 2.44)	<0.001	4.31	0.091
	Males	1.82 (0.73; 3.53)	0.59 (0.32; 1.00)	2.65 (1.44; 5.53)	<0.001	2.51 (1.43; 4.58)	<0.001
Age (years)	< 43	1.04 (0.59; 2.20)	0.62 (0.36; 1.04)	1.91 (0.87; 4.74)	<0.001	2.48 (1.96; 3.19)	<0.001
	43–54	1.55 (0.55; 3.05)	0.55 (0.30; 0.94)	2.04 (1.32; 5.16)	<0.001	2.92 (1.80; 5.78)	<0.001
	55–62	1.78 (0.78; 3.07)	0.68 (0.29; 0.98)	2.62 (1.56; 4.08)	<0.001	2.61 (1.41; 4.31)	<0.001
	63+	1.57 (0.51; 4.27)	0.42 (0.32; 1.02)	2.43 (1.45; 5.44)	<0.001	2.46 (1.35; 5.37)	<0.001
BMI (Kg/m ²) (M: 2)	< 25	1.25 (0.59; 2.31)	0.62 (0.32; 1.04)	2.04 (1.35; 4.21)	<0.001	2.15 (1.35; 4.12)	<0.001
	25–29	2.11 (0.71; 3.51)	0.42 (0.32; 0.73)	2.38 (1.26; 5.53)	<0.001	2.94 (2.16; 5.04)	<0.001
	30+	1.23 (0.47; 4.27)	0.52 (0.32; 0.71)	4.27 (2.00; 7.75)	0.002	1.35 (0.47; 4.91)	0.020
Amalgam dental fillings (number)	0	1.62 (0.63; 2.97)	0.43 (0.32; 0.84)	2.08 (1.17; 4.26)	<0.001	2.58 (1.43; 4.78)	<0.001
	1	0.92 (0.52; 2.72)	0.52 (0.48; 0.71)	2.80 (1.66; 3.49)	0.021	2.07 (1.15; 3.75)	0.006
	2	1.73 (0.92; 4.80)	1.04 (0.73; 1.47)	3.07 (1.39; 5.16)	0.047	4.06 (2.29; 6.85)	0.002
	3+	1.27 (0.49; 2.90)	0.93 (0.83; 1.31)	2.81 (1.87; 6.41)	<0.001	2.41 (1.60; 4.09)	<0.001
Smoking status	Never smoker	1.14 (0.46; 2.32)	0.55 (0.31; 0.97)	2.24 (1.45; 4.27)	<0.001	2.28 (1.51; 4.13)	<0.001
	Current smoker	0.83 (0.46; 2.28)	0.60 (0.36; 0.89)	2.16 (0.87; 3.49)	0.009	1.50 (0.45; 4.04)	0.111
	(cigarettes number)	2.07 (1.16; 3.63)	0.77 (0.20; 1.49)	2.80 (1.32; 6.68)	0.088	2.35 (1.33; 3.22)	0.029
	Ex smoker	2.85 (1.44; 4.89)	0.62 (0.52; 1.17)	2.60 (1.44; 5.16)	0.005	3.64 (2.50; 5.20)	0.001
Daily water intake (liter)	Up to 1	1.32 (0.47; 3.28)	0.43 (0.29; 0.95)	2.38 (1.10; 4.76)	<0.001	2.78 (1.35; 4.56)	<0.001
	1–1.5	1.26 (0.47; 2.15)	0.58 (0.29; 1.17)	1.91 (1.16; 2.66)	0.001	2.07 (1.43; 7.47)	<0.001
	1.5–2	1.90 (0.85; 3.23)	0.63 (0.32; 0.86)	2.50 (1.57; 5.64)	<0.001	2.46 (1.81; 3.46)	<0.001
	>2	1.67 (0.62; 3.49)	0.62 (0.48; 0.83)	2.87 (1.32; 7.29)	0.001	3.19 (1.37; 7.75)	0.003
Source of water intake	Bottled	0.93 (0.38; 2.33)	0.51 (0.31; 0.94)	2.44 (1.11; 4.15)	<0.001	3.23 (1.64; 5.01)	<0.001
		2.03 (1.09; 4.26)	0.71 (0.39; 1.17)	2.20 (1.35; 4.96)	<0.001	2.41 (1.43; 4.43)	<0.001
	Tap	2.08 (1.17; 4.28)	1.02 (0.48; 1.31)	2.20 (1.32; 5.16)	<0.001	2.33 (1.42; 4.31)	<0.001
		0.82 (0.37; 2.39)	0.51 (0.32; 0.88)	2.58 (1.09; 4.16)	<0.001	3.41 (1.77; 5.01)	<0.001
	Well	1.40 (0.60; 2.99)	0.58 (0.32; 0.98)	2.24 (1.21; 4.27)	<0.001	2.77 (1.68; 4.83)	<0.001
		2.19 (1.13; 5.54)	0.75 (0.48; 1.01)	3.23 (1.68; 6.46)	0.029	1.43 (1.09; 2.19)	0.083
Daily wine intake (liters) (M:2)	0	0.94 (0.44; 2.26)	0.50 (0.32; 0.93)	1.90 (0.87; 4.26)	<0.001	2.56 (1.72; 7.63)	<0.001
	<0.5	1.90 (0.72; 3.43)	0.59 (0.32; 1.02)	2.51 (1.62; 5.64)	<0.001	2.89 (1.43; 4.31)	<0.001
	0.5+	2.05 (1.18; 2.59)	0.89 (0.57; 1.59)	2.50	0.157	2.33 (1.33; 3.57)	0.050
Bruxism	No	1.39 (0.59; 2.92)	0.55 (0.32; 0.93)	2.36 (1.45; 5.27)	<0.001	2.56 (1.56; 4.43)	<0.001
	Yes	1.76 (0.91; 3.39)	0.91 (0.38; 1.55)	2.16 (1.17; 3.49)	0.007	2.67 (1.37; 5.00)	0.001
Chewing gum habit (M:2)	No	1.45 (0.62; 3.07)	0.57 (0.32; 0.95)	2.35 (1.39; 4.76)	<0.001	2.66 (1.71; 4.56)	<0.001
	Occasional	1.26 (0.44; 2.28)	0.58 (0.32; 1.26)	1.83 (1.25; 3.39)	0.010	1.81 (0.61; 4.42)	0.034

(continued on next page)

Table 2 (continued)

Terms	Strata		All sample (N = 249)	Controls (N = 93)	Workers of fish industry (N = 83)	P value	Fishermen (N = 73)	P value
		Usual	4.05 (2.28; 6.42)	NA	4.05 (2.65; 7.85)	NA	3.45 (1.90; 5.00)	NA
Nasal dyspnea		No	1.35 (0.58; 2.92)	0.58 (0.32; 1.00)	2.23 (1.39; 4.26)	<0.001	2.87 (1.41; 4.93)	<0.001
		Yes	1.99 (0.93; 3.55)	0.59 (0.32; 0.62)	3.44 (1.91; 6.03)	<0.001	2.24 (1.74; 3.09)	<0.001
Supplements		No	1.23 (0.53; 2.74)	0.58 (0.32; 0.97)	1.90 (1.16; 3.49)	<0.001	2.90 (1.68; 4.93)	<0.001
		Yes	2.19 (1.11; 4.26)	0.58 (0.43; 1.14)	3.07 (2.00; 5.64)	<0.001	2.07 (1.35; 3.10)	0.013
Use of contact of lenses		No	1.44 (0.59; 3.07)	0.55 (0.32; 1.00)	2.38 (1.44; 5.16)	<0.001	2.58 (1.43; 4.67)	<0.001
		Yes	1.24 (0.65; 1.93)	0.73 (0.48; 0.91)	1.63 (0.67; 2.97)	0.203	1.90 (1.81; 3.12)	0.053
Skin cream use		No	1.32 (0.52; 3.07)	0.52 (0.32; 0.98)	2.66 (1.32; 6.70)	<0.001	2.38 (1.39; 4.95)	<0.001
		Yes	1.69 (0.75; 2.93)	0.68 (0.41; 1.00)	2.16 (1.48; 3.77)	<0.001	2.61 (1.81; 3.57)	<0.001
Weekly fish meals (N)		< 0.75	0.61 (0.32; 1.40)	0.38 (0.25; 0.59)	1.35 (1.07; 2.28)	<0.001	1.90 (0.63; 3.46)	<0.001
		0.75–1	1.27 (0.59; 2.29)	0.71 (0.39; 1.14)	1.79 (0.66; 2.50)	0.001	2.24 (1.31; 3.18)	<0.001
		>1 ≤ 2	2.16 (1.17; 5.16)	0.93 (0.79; 1.17)	2.43 (1.63; 5.60)	<0.001	5.00 (2.24; 13.81)	<0.001
		3+	4.21 (2.51; 5.89)	0.61	4.76 (2.97; 7.29)	0.098	3.20 (2.40; 4.93)	0.103
Status of fish usually consumed	Fresh	No	0.58 (0.31; 1.04)	0.38 (0.29; 0.94)	0.86 (0.62; 1.39)	0.015	1.33 (0.40; 2.07)	0.075
		Yes	1.90 (0.86; 3.51)	0.64 (0.38; 1.04)	2.50 (1.65; 4.96)	<0.001	2.78 (1.68; 4.83)	<0.001
	Frozen	No	1.82 (0.81; 3.49)	0.62 (0.36; 1.04)	2.51 (1.57; 5.38)	<0.001	2.56 (1.43; 4.59)	<0.001
		Yes	0.62 (0.32; 1.20)	0.45 (0.29; 0.93)	1.37 (0.65; 2.62)	0.001	2.86 (1.90; 3.82)	0.022
	Canned	No	1.62 (0.64; 3.19)	0.59 (0.32; 0.98)	2.50 (1.44; 5.16)	<0.001	2.46 (1.45; 4.55)	<0.001
		Yes	1.01 (0.45; 1.85)	0.52 (0.32; 1.00)	1.63 (1.04; 1.78)	0.003	3.28 (1.35; 6.92)	<0.001
Type of fish usually consumed	Big size	No	1.85 (0.63; 3.19)	0.42 (0.29; 0.94)	2.50 (1.57; 4.12)	<0.001	2.33 (1.56; 4.79)	<0.001
		Yes	1.28 (0.61; 2.91)	0.62 (0.34; 1.00)	2.16 (1.26; 5.43)	<0.001	2.87 (1.37; 4.31)	<0.001
	Small/medium size	No	0.55 (0.29; 1.34)	0.38 (0.29; 0.81)	3.35 (1.04; 7.85)	<0.001	0.54 (0.22; 1.80)	0.484
		Yes	1.71 (0.73; 3.19)	0.62 (0.36; 1.00)	2.28 (1.45; 4.51)	<0.001	2.70 (1.79; 4.91)	<0.001
	Shell/cray fish/ mollusks	No	0.68 (0.32; 1.36)	0.39 (0.29; 0.93)	2.16 (0.65; 5.16)	0.008	1.72 (0.76; 2.64)	0.012
		Yes	1.85 (0.83; 3.49)	0.61 (0.39; 1.04)	2.36 (1.50; 4.75)	<0.001	2.78 (1.70; 4.83)	<0.001

concentration by number of weekly fish meals is reported in Fig. 5, whereas a plot for the linear prediction of log transformed hair Hg concentration by number of weekly fish meals is displayed in Fig. 6.

Table 5a reports the results of a multiple linear regression model on log transformed hair Hg, adjusted for weekly fish consumption, fresh fish intake, study group, BMI, smoking status, chewing gum consumption and tap water intake. Only significant estimates are displayed. Although retained with borderline significance in the final regression model by backward stepwise selection, stratum specific estimates for chewing-gum consumption (occasional or usual vs. No) were omitted from Table 5a, since they both failed to overcome the BH procedure. The level of log-transformed hair Hg increased with all levels of weekly fish intake, consumption of fresh fish (aRC = 0.56; 95%CI: 0.29; 0.82) and, compared to controls from the Dolomites Alps, was significantly higher in fish dealers/fish restaurateurs (aRC = 1.37; 95%CI: 0.95; 1.79), lagoon/mixed fishermen (aRC = 1.15; 95%CI: 0.81; 1.50) and open sea fishermen (aRC = 0.97; 95%CI: 0.58; 1.34). Following application of BH procedure set at 5% FDR, the strata corresponding to 0.75–1 weekly fish

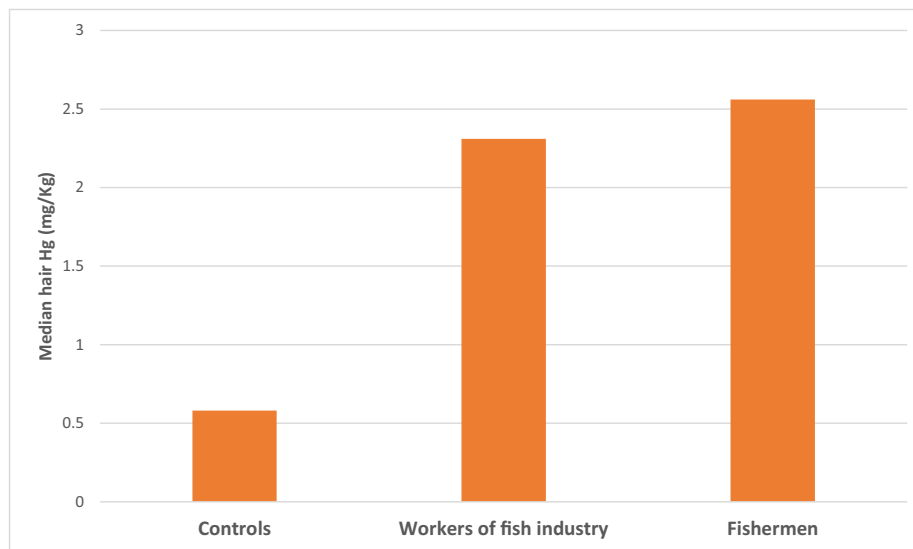
meals and administrative staff of fish industry both lost statistical significance (Table 5a). Supplementary file S4 shows the distribution of residuals of log-transformed hair Hg concentration.

Table 5b shows the results of a multiple logistic regression on odds of hair Hg > 2 mg/kg adjusted for weekly fish intake, consumption of fresh fish, study group, BMI and chewing-gum consumption. Only significant estimates are displayed. The odds of hair Hg concentration > 2 mg/Kg increased consistently with the two higher levels of fish intake, >1 ≤ 2; (aOR = 5.17; 95%CI: 1.66; 16.11) or >2 fish meals per week (aOR = 9.37; 95%CI: 2.33; 37.68), consumption of fresh fish (aOR = 9.59; 95%CI: 2.07; 44.30) and, with the exception of fish farmers and local residents involved in other business, in all study categories, especially fish dealers/fish restaurateurs (aOR = 75.88; 95%CI: 12.00; 479.84), open sea fishermen (aOR = 22.72; 95%CI: 6.34; 81.42) and lagoon/mixed fishermen (aOR = 14.87; 95%CI: 4.46; 49.54) and (Table 5b).

Table 3

Distribution of hair mercury (mg/Kg) by study group. Number (N); column percentage (%); median; interquartile range (IQR); range.

Study group	N (%)	Hair mercury level (mg/kg)		Weekly fish consumption (number of meals)				
		Median (IQR)	Range	Median (IQR)	< 0.75 N (%)	0.75 – 1 N (%)	>1 ≤2 N (%)	>2 N (%)
Controls	93 (37.5)	0.58 (0.32; 0.98)	0.02; 3.09	1 (0.5; 1)	45 (48.4)	34 (35.5)	15 (15.1)	1 (1.1)
Open sea fishermen	32 (12.9)	2.33 (1.41; 3.89)	0.18; 7.47	1 (0.75; 2)	10 (31.3)	11 (34.4)	5 (15.6)	6 (18.8)
Lagoon/mixed fishermen	41 (16.5)	2.92 (1.81; 4.93)	0.42; 20.44	1 (1; 2)	9 (22.0)	14 (34.2)	8 (19.5)	10 (24.4)
Administrative (fish industry) ^a	18 (7.2)	1.85 (0.86; 2.24)	0.46; 11.15	1 (0.75; 2)	5 (27.8)	5 (27.8)	6 (33.3)	2 (11.1)
Fish dealers/fish restaurateurs	26 (19.4)	5.59 (2.66; 7.85)	0.75; 13.97	2.75 (1.5; 3)	1 (3.9)	4 (15.4)	7 (26.9)	14 (53.9)
Storers/porters/carriers/vendors (local fish markets) ^b	11 (4.4)	1.99 (1.16; 4.05)	0.29; 5.16	2 (1; 2)	1 (9.1)	3 (27.3)	6 (54.6)	1 (9.1)
Retired fishermen/ fishermen household members	11 (4.4)	1.90 (1.44; 3.07)	0.50; 7.75	1 (0.75; 2)	3 (27.3)	5 (45.5)	3 (27.3)	0
Other business (local residents)	7 (2.8)	2.65 (1.04; 5.38)	0.48; 5.44	2 (1; 3)	1 (14.3)	2 (28.6)	1 (14.3)	3 (42.9)
Fish farmers	10 (4.0)	1.54 (1.04; 2.35)	0.61; 3.49	2 (1; 2)	1 (10.0)	3 (30.0)	5 (50.0)	1 (10.0)

^a Including a biologist and a manager of the local fish market.^b Including workers of a large fish private company.**Fig. 2a.** Distribution of median hair Hg (mg/Kg) by main study group.

4. Discussion

4.1. Key findings

The present study found a significantly higher median hair Hg concentration in fishermen (2.56 mg/kg) and in workers of fish industry (2.31 mg/kg) compared to controls from the Dolomites Alps (0.58 mg/kg) (Table 1).

The mean level of log transformed hair Hg increased with weekly fish intake (>1 meal per week), usual consumption of fresh fish and was significantly higher in fish dealers/fish restaurateurs and fishermen - regardless of whether they operated in open sea or lagoon (Table 5a).

Similarly, respondents eating >1 fish meal per week and usually consuming fresh fish were more likely to exhibit hair Hg levels >2 mg/kg. Moreover, compared to controls, all categories but fish farmers and local residents involved in other business were more likely to exhibit hair Hg concentration > 2 mg/kg, especially fish dealers/fish restaurateurs and fishermen (regardless if operating in open sea or lagoon) (Table 5b).

4.2. Interpretation of findings

4.2.1. Comparison with other studies from the Gulf of Trieste

The lower concentration of hair Hg in controls compared to all other study categories surveyed from Marano Lagunare points at consumption of fish as main risk factor of Hg exposure in this lagoon coastal area. All

participants from Marano regularly consumed locally caught fish.

The median hair Hg concentrations in fishermen (2.56 mg/kg) and workers of fish industry (2.31 mg/kg) of Marano were slightly higher than the cut-off recommended by WHO for the general population (2 mg/Kg), although well below the threshold of no health effects on humans (11.5 mg/kg) set by EFSA for chronic exposure to this heavy metal [EFSA, 2012]. Nonetheless, some outliers in Hg hair concentrations were found, especially among lagoon fishermen (maximum value: 20.44 mg/kg) and fish dealers/restaurateurs (max value: 13.97 mg/kg), likely attributable to more intense consumption of local fish caught from the lagoon. In particular, 14.6 % (=6/41) lagoon/mixed fishermen and 15.4 % (=4/26) fish dealers/restaurateurs exhibited hair Hg concentrations >10 mg/kg.

The median hair Hg concentration in controls from the Dolomites was 0.58 mg/kg, an estimate which could be used as reference, since generally every median value >0 is considered an environmental exposure in a study group.

Whilst the US Food and Drug Administration estimates that most people will have a daily food-related Hg exposure not exceeding 50 ng/kg, a dose not considered to be harmful to humans [Aprea et al., 2021, FDA, 2022], hair Hg concentration reportedly varies by amount of and type of fish intake as well as area of residence [Giangrosso et al., 2016; Okati and Esmaili-Sari, 2018a; Gaggi et al., 1996; Renzoni, 1992; Kirichuk et al., 2020]. Higher levels of hair Hg in residents of coastal areas have been in fact reported in the literature [Giangrosso et al., 2016; Okati and Esmaili-Sari, 2018a; Gaggi et al., 1996; Renzoni, 1992;

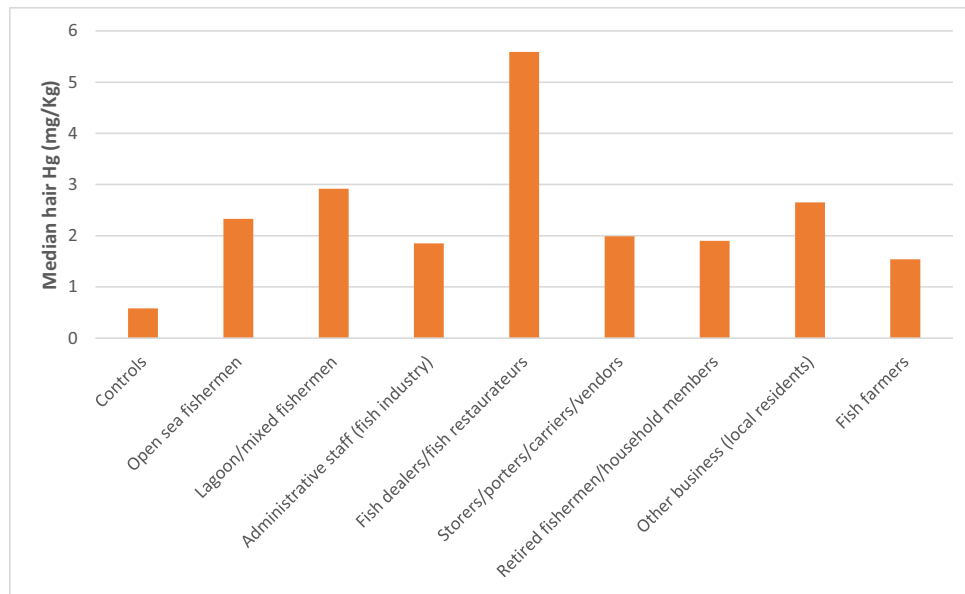


Fig. 2b. Distribution of median hair Hg (mg/Kg) by detailed study group.

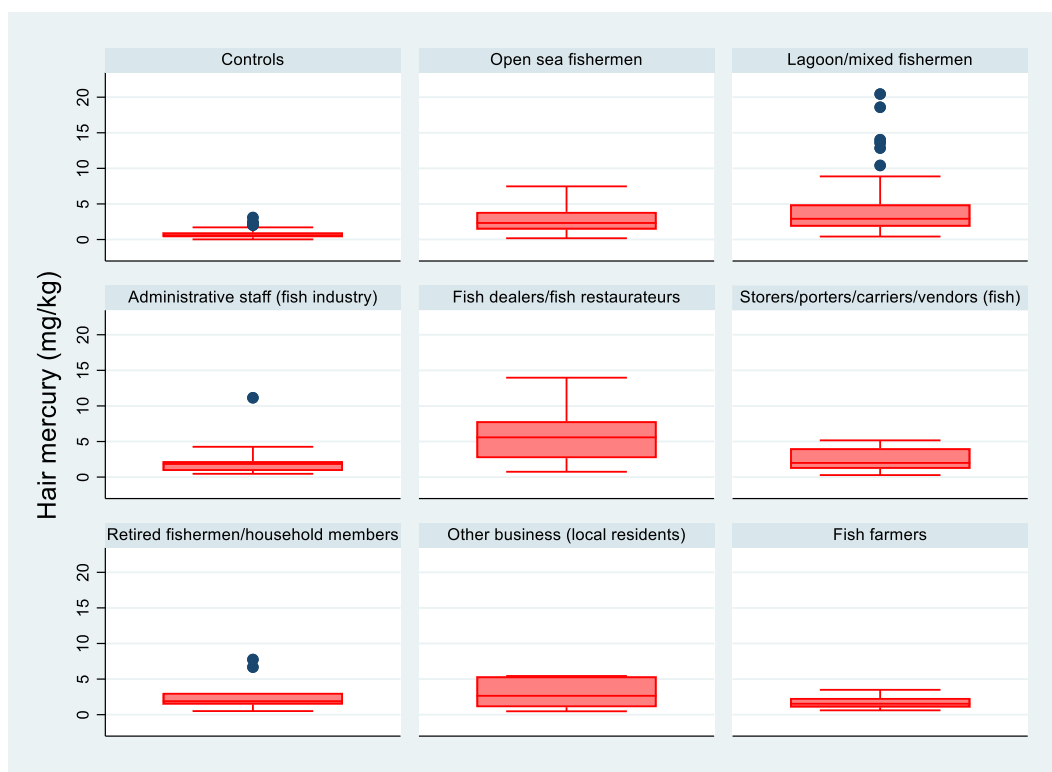


Fig. 3. Box plot displaying median and interquartile range of concentration of hair mercury (mg/Kg) by study group.

Matthews, 1983; Okati and Esmaili-Sari, 2018b; Cegolon et al., 2023; Cegolon et al., 2022; Lincoln et al., 2011; Bonsignore et al., 2016].

For instance, on the opposite site of the Adriatic Sea, in a coastal area of Croatia, substantially lower median levels of hair Hg (0.6 mg/kg; range: 0.016–8.710) were reported in 234 local women, slightly higher than 584 women from central Slovenia (median 0.298 mg/kg; range 0.015–0.439) [Trdin et al., 2019].

In a study on 606 pregnant women delivering in Trieste during 2007–2009, the mean Hg hair concentration (1.06 mg/kg) was slightly higher than the above survey from Croatia and Slovenia, yet still lower

than the present study and partially correlated with fish consumption [Valent et al., 2013]. At follow up in 2007 among mothers resident in three neighbouring municipalities facing the respective lagoon (Marano, Grado and Carlino), the median Hg hair concentration was found to be significantly higher compared to mothers living inland (1.17; IQR: 0.625; 1.950 vs. 0.871, IQR: 0.509; 1.279, respectively) [Valent et al., 2013].

Similar median estimates (1.2 mg/kg; IQR: 0.78; 1.86) were detected in another study on 301 individuals (119 males vs 182 females) from the general population recruited by convenience sampling in 2021 in the

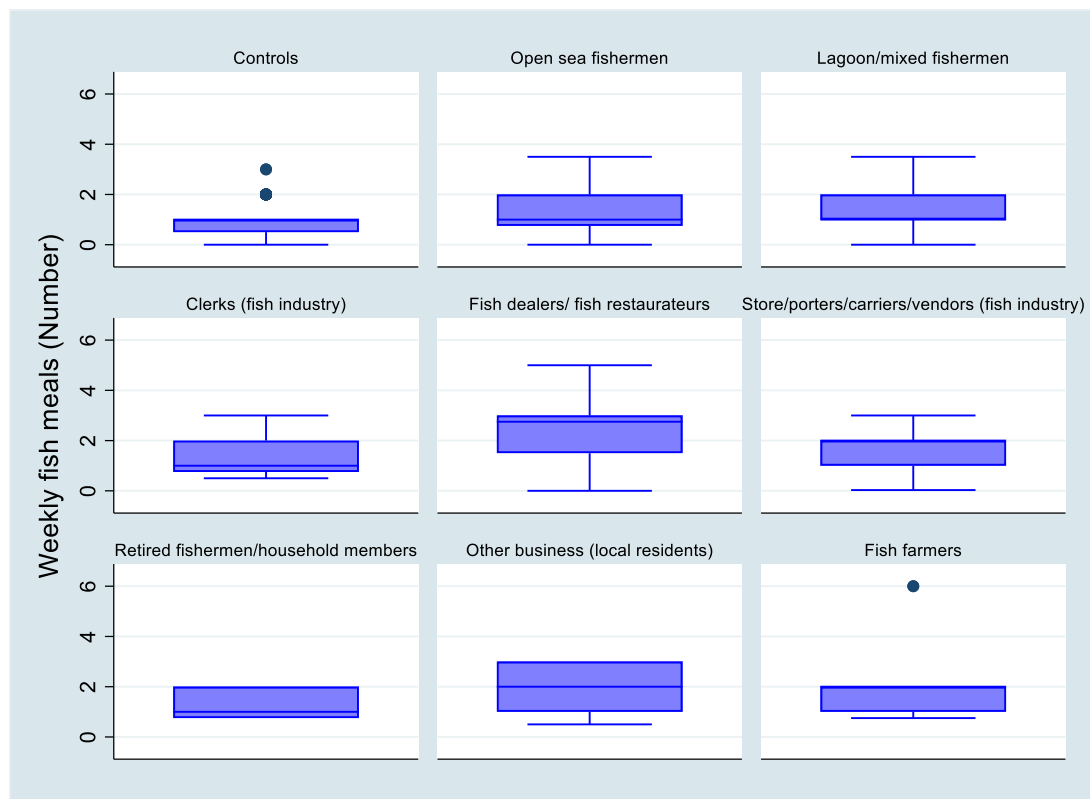


Fig. 4. Box plot displaying median and interquartile range of weekly fish meals, by study group.

Table 4

Distribution of fish type and size usually consumed, by study group and sex of respondents. Number (N) and column percentage (%).

Study group	Strata	Status of fish usually consumed N (%)			Fish type usually consumed N (%)		
		Fresh	Frozen	Canned	Big	Small/medium	Shell-crayfish/mollusks
Controls	All	58 (62.4)	36 (38.7)	22 (23.2)	61 (66.3)	69 (75.0)	51 (55.4)
	Males	31 (57.4)	22 (40.7)	16 (28.6)	35 (66.0)	39 (73.6)	31 (58.5)
	Females	27 (69.2)	14 (35.9)	6 (15.4)	26 (66.7)	30 (76.9)	20 (51.3)
Open sea fishermen	All	29 (90.6)	0	6 (18.8)	17 (53.1)	29 (90.6)	28 (87.5)
	Males	28 (90.3)	0	5 (16.1)	16 (51.6)	28 (90.3)	27 (87.1)
	Females	1	0	1 (100)	1 (100)	1 (100)	1 (100)
Lagoon/mixed fishermen	All	39 (95.1)	2 (4.9)	4 (9.8)	15 (36.6)	38 (92.7)	36 (87.8)
	Males	39 (95.1)	2 (4.9)	4 (9.8)	15 (36.6)	38 (92.7)	36 (87.8)
	Females	0	0	0	0	0	0
Administrative (fish industry)	All	15 (83.3)	7 (38.9)	3 (16.7)	15 (83.3)	17 (94.4)	12 (66.7)
	Males	10 (100)	1 (10.0)	1 (10.0)	8 (80.0)	10 (100)	7 (70.0)
	Females	5 (62.5)	6 (75.0)	2 (25.0)	7 (87.5)	7 (87.5)	5 (62.5)
Fish dealers/fish restaurateurs	All	22 (84.6)	5 (19.2)	1 (3.9)	21 (80.8)	22 (84.6)	25 (96.2)
	Males	20 (87.0)	5 (21.7)	1 (4.4)	20 (87.0)	20 (87.0)	23 (100)
	Females	2 (66.7)	0	0	1 (33.3)	2 (66.7)	2 (66.7)
Storers/porters/carriers/vendors (local fish markets)	All	9 (81.8)	2 (18.2)	0	6 (54.6)	11 (100)	10 (90.9)
	Males	8 (80.0)	2 (20.0)	0	6 (60.0)	10 (100)	10 (100)
	Females	1 (100)	0	0	0	1 (100)	0
Retired fishermen/fishermen household members	All	11 (100)	1 (9.1)	1 (9.1)	3 (27.3)	11 (100)	11 (100)
	Males	6 (100)	1 (16.7)	0	2 (33.3)	6 (100)	6 (100)
	Females	5 (100)	0	1 (20.0)	1 (20.0)	5 (100)	5 (100)
Other business (local residents)	All	5 (71.4)	1 (14.3)	1 (14.3)	5 (71.4)	6 (85.7)	4 (57.1)
	Males	4 (66.7)	1 (16.7)	1 (16.7)	4 (66.7)	5 (83.3)	4 (66.7)
	Females	1 (100)	0	0	1 (100)	1 (100)	0
Fish farmers	All	10 (100)	0	0	5 (50.0)	9 (90.0)	10 (100)
	Males	8 (100)	0	0	4 (50.0)	7 (87.5)	8 (100)
	Females	2 (100)	0	0	1 (50.0)	2 (100)	2 (100)

city of Trieste [Cegolon et al., 2023].

By contrast, lower median hair concentrations (0.70 mg/kg; IQR: 0.42; 1.34) were found in a study on 50 workers of Grado beach, a touristic seaside resort very close to the mouth of Isonzo river, at the very Eastern sector of the lagoon [Cegolon et al., 2022]. Nonetheless,

hair Hg concentrations consistently increased with fish consumption in both latter surveys [Cegolon et al., 2023; Cegolon et al., 2022].

In Grado beach, further South-West of the Isonzo river mouth, Hg concentration in sands was found to be higher than the regional background of Friuli Venezia Giulia (0.13 mg/kg) [Covelli et al., 2006],

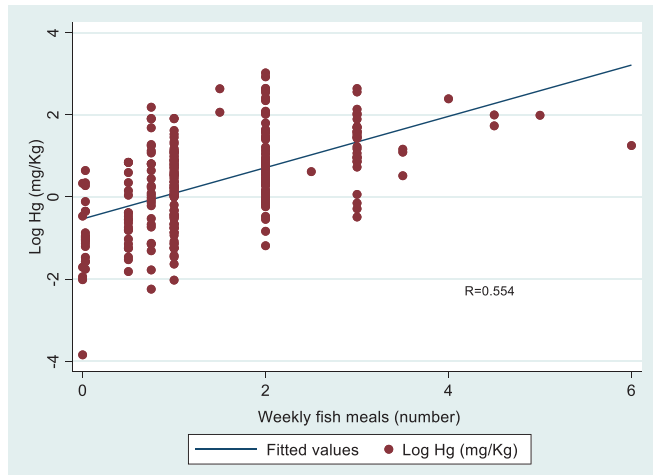


Fig. 5. Scatterplot for the distribution of log transformed hair mercury concentration (mg/Kg) By weekly fish meals.

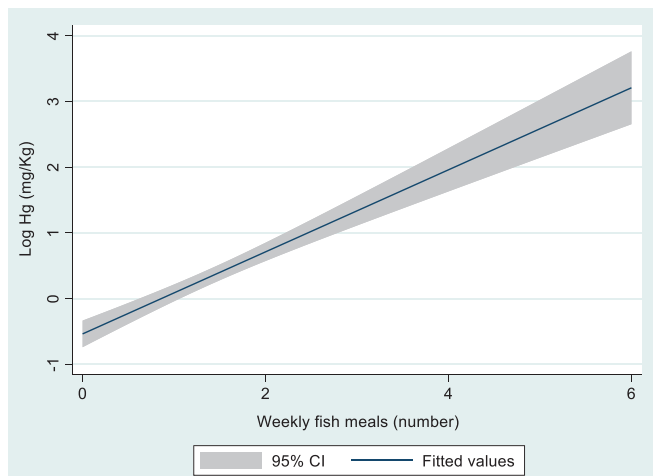


Fig. 6. Linear prediction of log transformed hair mercury concentration (mg/Kg) with 95% confidence interval (95%CI) by number of weekly fish meals.

varying between 0.17 and 6.07 (Mean = 1.99 ± 1.85) mg/kg in 15 sampling points and retrieved at different depths (50–200 cm deep) from four selected sampling spots (Covelli, unpublished data). These sediment samples also exceeded the Hg contamination limit (1 mg/kg) for soils in areas intended for resident use as established by Italian Legislative Decree n. 152/06.

Table 5a

Multivariable linear regression on the level of log transformed hair mercury (mg/Kg), adjusted for BMI, smoking status, chewing gum consumption, tap water intake, weekly fish consumption, fresh fish intake, study group. Only significant results are displayed, expressed as adjusted regression coefficients (aRC) with 95 % confidence intervals (95 %CI), Wald test p value and Benjamini-Hochberg (BH) p-value set a 5 % false discovery rate. NS = Non-significant.

TERMS	Strata	Log (hair Hg) aRC (95%CI) (227 obs.)	Wald test p-value	BH p-value (≤ 0.0150)
Weekly fish meals (Number)	< 0.75	reference		
	0.75–1	0.28 (0.02; 0.54)	0.035	0.0225 (NS)
	>1 \leq 2	0.91 (0.61; 1.21)	<0.001	0.0025
	>2	0.75 (0.39; 1.12)	<0.001	0.0050
Usual intake of fresh fish	No	reference		
	Yes	0.56 (0.29; 0.82)	<0.001	0.0075
Study group	Controls	reference		
	Open sea fishermen	0.97 (0.59; 1.34)	<0.001	0.0100
	Lagoon/mixed fishermen	1.15 (0.81; 1.50)	<0.001	0.0125
	Administrative (fish industry)	0.52 (0.08; 0.97)	0.021	0.0175 (NS)
	Fish dealers/fish restaurateurs	1.37 (0.94; 1.79)	<0.001	0.0150

Although the sand of Grado beach was found to be contaminated by cinnabar, no increased risk of exposure to the metal through skin contact with sand or ingestion of seawater was suggested. Thermo-desorption of sandy samples collected from Grado beach showed that Hg was mainly cinnabar and meta-cinnabar, two poorly soluble chemical forms, whereby inorganic Hg is not easily removable and, therefore, not bioavailable [Cegolon et al., 2022].

Differently from Grado area, the main Hg species detected in the central sector (by the mouth of Aussa-Corno River) was inorganic Hg, mostly discharged by SNIA Viscosa CAP during more than 50 years of industrial activity [Acquavita et al., 2012]. Both metal species will be ultimately converted into MeHg by anaerobic microorganisms in anoxic sediments and water, entering the food chain of aquatic systems and bio-accumulating in fishes, thereby becoming bioavailable to humans [NRC, 2000].

4.2.2. Generalizability to other Italian settings outside the Gulf of Trieste

Investigations on hair Hg concentrations in other Italian coastal areas at risk outside the Gulf of Trieste were mainly conducted in the Southern part of the country. For instance, in 237 residents of Naples (coastal Campania region, Southern Italy) – 115 females vs. 122 males –, a mean hair Hg concentration of 0.64 (range: 0.22 and 3.40) mg/kg was reported, hence much lower than the present survey [Diez et al., 2008].

Another study investigated hair and blood Hg in 224 residents of three municipalities (Augusta, Priolo, Melilli) on the Augusta Bay, a polluted costal area from Sicily. The overall median Hg level in hair was 1.47 mg/kg, broken down by 1.90 mg/kg in Augusta, 1.24 mg/kg in Melilli and 1.00 mg/kg in Priolo [Bonsignore et al., 2016]. Hg measurements positively correlated with intake of locally caught fish. In particular, the most extreme value in blood (33.6 μ g/L) was found in a subject eating local fish 3–4 times/week, whereas the lowest blood concentration was found in a resident reporting seldom ingestion of locally caught seafood (0.10 μ g/L) [Bonsignore et al., 2016].

Another investigation on six locations across Sicily (Syracuse, Messina, Trapani, Sciacca, Catania, Palermo) contrasted hair Hg of 96 fishermen against 96 controls not employed in the maritime sector, all aged between 35 and 45 years [Giangrosso et al., 2016]. Significantly higher mean hair Hg concentrations were observed in fishermen (6.45 ± 7.03 mg/kg) compared to controls (0.23 ± 0.4 mg/kg), with higher estimates in Syracuse (4.71 ± 6.79 mg/kg) and Sciacca (3.94 ± 6.38 mg/kg) - both locations hosting chemical industrial plants - with a peak concentration of 16.48 mg/kg in a fisherman from Sciacca [Giangrosso et al., 2016].

Similar mean hair Hg levels (5.64 ± 3.73 mg/kg) were found in fishermen from the Aeolian Archipelago (North of Sicily, Southern Tyrrhenian Sea) [Renzoni, 1992].

By contrast, higher mean Hg levels were found in residents of two coastal fishing villages of Northern Tyrrhenian sea, where consistently with the present study hair Hg concentrations increased with

Table 5b

Multivariable logistic regression on the level of hair mercury >2 vs. ≤2 mg/Kg, adjusted for BMI, chewing gum consumption, weekly fish intake, consumption of fresh fish; study group. Only significant results are displayed, expressed as adjusted odds ratio (aOR), with 95 % confidence interval, Wald test p value and Benjamini-Hochberg (BH) p-value set a 5 % false discovery rate. NS = Non-significant.

Terms	Strata	Hair Hg >2 mg/Kg aOR (95%CI) (245 obs.)	P value	BH p-value (≤ 0.0250)
Weekly fish meals (Number)	< 0.75	reference		
	>1 ≤ 2	5.17 (1.66; 16.11)	0.005	0.0188
	>2	9.37 (2.33; 37.68)	0.002	0.0125
Usual intake of fresh fish	No	reference		
	Yes	9.59 (2.07; 44.30)	0.004	0.0156
Study group	Controls	reference		
	Open sea fishermen	22.72 (6.34; 81.42)	<0.001	0.0031
	Lagoon/mixed fishermen	14.87 (4.46; 49.54)	<0.001	0.0063
	Administrative staff (fish industry)	5.61 (1.29; 24.35)	0.021	0.0250
	Fish dealers/fish restaurateurs	75.88 (12.00; 479.84)	<0.001	0.0094
	Retired fishermen/ fishermen household members	8.66 (1.75; 42.93)	0.008	0.0219

consumption of seafood [Renzone et al., 1998]. In particular, the mean hair Hg was 1.03 ± 0.21 mg/kg among 40 local residents eating ≤ 1 fish meals per week, 8.89 ± 5.74 among 88 eating 2–4 four weekly fish meals and 36.38 ± 21.62 mg/kg among 39 subjects reportedly having ≥ 4 fish servings per week [Renzone et al., 1998]. Likewise, in an earlier study on residents of a coastal village from the same Northern Tyrrhenian sea area (Vada, Tuscany), a mean air Hg concentration of 27.11 ± 29.30 mg/kg was found in males against 7.52 ± 8.79 mg/kg among females [Renzone et al., 1998]. In North Tyrrhenian sea most fish species (especially those living by the sea bed, as the Norwegian lobster) exhibited high levels of Hg, increasing with fish body weight [Renzone et al., 1998].

4.2.3. Generalizability outside the Mediterranean Sea

The higher accrual of Hg in bluefin tuna from the Mediterranean compared to species from the Atlantic Ocean may be partly explained by the fact that approximately half of the entire global Hg reservoir is located within the Mediterranean basin, and by the low rate of water exchange through the Gibraltar canal [Renzone et al., 1998]. Nevertheless, tuna are large predatory species at the apex of food chain, covering large parts of the North Atlantic Ocean in their annual migrations.

The average Hg value of various pelagic, benthic demersal and species from the Mediterranean reportedly was 0.381 ± 0.131 mg/kg, ranging from 0.132 ± 0.108 in Sardines (*Sardina pilchardus*) to 0.458 ± 0.059 mg/kg in Scabbard (*Lepidopus caudatus*) [Naccari et al., 2015] and this might also explain the higher levels of hair Hg in fishermen operating outside the Mediterranean area. Hg levels in benthic crustaceans and mollusks were found to be influenced by concentration of sedimentary Hg in various areas of the Mediterranean [Hornung et al., 1984; Hornung, 1986].

Very high mean Hg levels (39.76 mg/kg) were found by a survey on 80 fishermen and respective household members (58 males vs. 22 females) from Camara de Lobos village (Madeira island, Portugal) [Gaggi et al., 1996], where the most common fish type consumed by fishermen was scabbard fish (*Aphanopus carbo*), a species reportedly accumulating high levels of total Hg (0.90 ± 0.27 mg/kg) [Afonso et al., 2006]. Furthermore, Madeiran fishermen typically eat fish both at work as well as at home, whereas women eat seafood in the evening and only occasionally at lunch and this likely explains the discrepancy in concentration by sex of respondents [Gaggi et al., 1996].

Likewise, 40 male fishermen from the Seychelles exhibited a mean hair Hg level of 26.29 ± 14.51 mg/kg against 12.25 ± 6.60 mg/kg among 36 local mothers and 15.25 ± 11.50 mg/kg in 36 children [Renzone, 1992]. By contrast, 26 male fishermen from the Maldives reportedly exhibited a mean levels of total hair Hg of 10.69 ± 1.81 mg/kg against 7.82 ± 2.26 mg/kg in non-fishermen local males and 7.56 ± 1.99 in 40 local women [Renzone, 1992]. Seychellois have one of the highest fish consumption in the world (an annual 80–100 Kg per person) and in addition to Dogtooth tuna (*Gymnosarda unicolor*) local people consume also other species as Carangue balo (*Caranx gymnostethus*) and

Bonito (*Euthynnus affinis*), reportedly exceeding 1.0 mg/kg concentration [Matthews, 1983].

Although eating seafood three times per week, 8 residents from Iceland exhibited very low mean mercury levels (1.35 ± 0.25 mg/Kg) [Renzone, 1992], endorsing the hypothesis that environmental exposure to Hg is not only influenced by geographical area but also by the amount and type of fish consumed. For instance, in the Faroe islands the estimated intake of MeHg from ingestion of pilot whale (*Globicephalus meleanus*) during years 1970–1984 approached the lower value (1200 µg/person/week) of critical level for intoxication in the general population [Andersen et al., 1987]. Furtherly, mercury concentrations in liver and kidneys of Greenlanders were higher than levels of the general population of Japan, Korea and several European countries, except in the Faroe Islands, where Hg levels were 2–3 times higher. The latter findings reflected the expected dietary exposure to mercury [Johansen et al., 2007]. However, following issuance of official dietary recommendations to avoid eating pilot whales, significantly reduced hair mercury levels were found among 412 women from the Faroe islands surveyed in 1999 [Weihe et al., 2005].

Trophic transfer of MeHg through food chain reportedly determine Hg concentrations in large fish predators millions of times higher than levels detected in surface water, following bio-magnification [Lavoie et al., 2013; De Almeida et al., 2019]. The lack of association between hair Hg levels and the consumption of large fishes in the present study may be explained by ban on tuna fishing in the Gulf of Trieste. Indeed, edible tuna is imported in Friuli Venezia Giulia region from outside, and intake of local fish is limited to small/medium size fishes or shell-crayfish/mollusks. However, high Hg (605 ± 210 ng/g ww) as well as MeHg (147 ± 37 ng/g ww) contents were still reportedly found in Manila clams from the Marano and Grado Lagoon [Giani et al., 2012].

4.3. Strengths and limitations

4.3.1. Strengths

Few studies have investigated Hg concentrations in fish in this coastal marine area [Kosta et al., 1978; Horvat et al., 1999] and no published study exists on fishermen and residents of Marano, the main fishing spot of one of the largest lagoons in Europe. Moreover, recruiting workers of fish industry for this survey was particularly challenging, because of fear of detrimental repercussions on their job.

Using hair specimens to estimate Hg exposure is an established approach in surveys [Bakir et al., 1973; Davidson et al., 1998; Virtanen et al., 2005], where hair is typically used to characterize the degree of Hg exposure in a study population, a goal relatively practical and straightforward [Nuttall, 2006]. Although whole blood and urine are considered the most reliable specimens to estimate Hg exposure in humans, analysis of hair specimens presents several advantages [Nuttall, 2004]:

- longer half-life of Hg in hair, allowing to estimate exposures occurred months before;
- easiness of storage and transport of specimen before the analysis, combined with long time stability of Hg in hair,
- easy measurement of some Hg forms, especially organic MeHg, which tends to accumulate at higher concentration in hair [Bencze, 1994].

4.3.2. Limitations

A major limitation of using hair specimen is the impossibility to distinguish endogenous Hg incorporated during hair growth from exogenous supplies deposited in hair (e.g. hair dyes) [Morton et al., 2002]. Moreover, vigorous hair cleansing could potentially remove Hg incorporated during hair growth.

Hair and blood measurement of Hg increased their correlation when [Nuttall, 2006]:

- The species involved is primarily MeHg.
- Blood Hg is stable during the relevant time period, and
- No exogenous Hg contamination of hair is involved

In the present study, exogenous sources of hair Hg (as hair dyes) were assumed to be negligible and blood Hg was believed to be constant since fish consumption, the major risk factor influencing hair Hg concentration, was considered a stable habit of respondents over time.

5. Conclusions

The level of hair Hg in fishermen and workers of fish industry of Marano was higher than controls from the Dolomites area and other polluted coastal sites in Italy. Hair mercury concentration increased with weekly fish intake, consumption of fresh fish and in categories with higher access to locally caught fish as fishermen and fish dealers/fish restaurateurs.

The above evidence suggests that the environmental context linked to the dual source (industrial and mining) of Hg contamination in coastal and lagoon sediments may have influenced the transfer of the metal into the aquatic trophic chain and from there to humans through consumption of local fish.

Whilst above the threshold background exposure recommended by WHO for the general population, the median levels of hair Hg in fishermen and workers of fish industry of Marano were still below the cut-off of no health effects (11.5 mg/kg) observed on human health.

Since hair Hg concentration increased with daily fish consumption, vulnerable categories as pregnant women and children should limit the intake consumption of local fish, especially caught from the lagoon, to no more than one fish meal per week.

Supplementary data to this article can be found online at <https://doi.org/10.1016/j.scitotenv.2024.178039>.

CRedit authorship contribution statement

Luca Cegolon: Writing – original draft, Methodology, Investigation, Formal analysis, Data curation, Conceptualization. **Stefano Covelli:** Writing – original draft, Supervision, Formal analysis. **Emilia Patriarca:** Investigation, Data curation. **Elisa Petranich:** Writing – review & editing, Formal analysis, Data curation. **Federico Floreani:** Writing – review & editing, Investigation, Formal analysis. **Donatella Sansone:** Methodology, Data curation. **Giuseppe Mastrangelo:** Writing – review & editing, Supervision, Methodology. **Francesca Larese Filon:** Writing – review & editing, Supervision, Resources.

Consent to participate

Informed written consent was taken voluntarily from each eligible participant.

Consent to publish

Not applicable.

Ethical approval

This study was approved by the Regional Ethic Committee of Friuli Venezia Giulia (N. 092/2018 23.03.2018) and the Ethic Committee of the University of Trieste (Prot. N. 11; 17/12/2024).

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Acknowledgements

The authors would like to thank:

- Mr. Sandro Caporale, President of Cooperativa San Vito, for facilitating the survey and data collection from fishermen and workers of fish industry of Marano Lagunare.
- Dr. Sandro Cinquetti (Director of Public Health of AULSS 1 “Dolomiti”) and Dr. Damiano Comin (Director of Veterinary Unit of AULSS 1 “Dolomiti”), for facilitating the survey and data collection from herdsman and farmers within Belluno catchment area.

Data availability

The datasets generated during and/or analyzed for the present study are not publicly accessible but may be available from the corresponding author upon reasonable request.

References

- Acquavita, A., Bettoso, N., 2018. Mercury and selenium in the grass goby *Zosterisessor ophiocephalus* (Pisces: Gobiidae) from a mercury contaminated Mediterranean lagoon. *Mar. Pollut. Bull.* 135, 75–82.
- Acquavita, A., Covelli, S., Emili, A., Berto, D., Faganeli, J., Gianì, M., et al., 2012. Mercury in the sediments of the Marano and Grado lagoon (northern Adriatic Sea): sources, distribution and speciation. *Estuar. Coast. Shelf Sci.* 113, 20–31.
- Acquavita, A., Biasiol, S., Lizzi, D., Mattassi, G., Pasquon, M., Skert, N., Marchiol, L., 2017. Gaseous elemental mercury level and distribution in a heavily contaminated site: the ex-chlor alkali Plant in Torviscosa (northern Italy). *Water Air Soil Pollut.* 228.
- Afonso, C., Lourenc, H.M., Dias, A., Nunes, M.L., Castro, M., 2006. Contaminant metals in black scabbard fish (*Aphanopus carbo*) caught off Madeira and the Azores. *Food Chem.* 101 (1), 120–125.
- Andersen, A., Julshamn, K., Ringdal, O., Mørkøre, J., 1987. Trace elements intake in the Faroe Islands II. Intake of mercury and other elements by consumption of pilot whales (*Globicephalus meleanus*). *Sci. Total Environ.* 65, 63–68.
- Aprea, M.C., Apostoli, P., Bettinelli, M., Lovreglio, P., Negrie, S., Perbellini, L., et al., 2021. Urinary levels of metal elements in the non-smoking general population in Italy: SIVR study 2012–2015. *Toxicol. Lett.* 298, 177–185.
- Bacci, E., Angotzi, G., Bralia, A., Lampariello, L., Zanette, E., 1976. Etude sur une population humaine exposée par la consommation de poisson. *Rev. Intern. Océanogr. Med.* 41–42, 127–141.
- Bakir, F., Damluji, S.F., Amin-Zaki, L., Murtadha, M., Khalidi, A., Al-Rawi, N.Y., et al., 1973. Methylmercury poisoning in Iraq. *Science* 181, 230–241.
- Bencze, K., 1994. Determination of metals in human hair. In: *Handbook on Metals in Clinical and Analytical Chemistry* (Seiler HG, Sigel A, Sigel H, Eds). Marcel Dekker, New York, pp. 201–216.
- Bettoso, N., Pittaluga, F., Predonzani, S., Zanello, A., Acquavita, A., 2023. Mercury levels in sediment, water and selected organisms collected in a coastal contaminated environment: the Marano and Grado lagoon (northern Adriatic Sea, Italy). *Appl. Sci.* 13, 3064.
- Bonsignore, M., Andolfi, N., Barra, M., et al., 2016. Assessment of mercury exposure in human populations: A status report from Augusta bay (southern Italy). *Environ. Res.* 150, 592–599.
- Bravo, A.G., Zopfi, J., Buck, M., Xu, J., Bertilsson, S., Schaefer, J.K., Pote, J., Cosio, C., 2018. Geobacteraceae are important members of mercury-methylating microbial communities of sediments impacted by waste water releases. *ISME J.* 12 (3), 802–812.

- Cegolon, L., Mastrangelo, G., Covelli, S., Petranich, E., Pavoni, E., Larese, Filon F., 2022. Occupational exposure to mercury from cinnabar enriched sand in workers of Grado Beach, gulf of Trieste (North-Eastern Italy, upper Adriatic Sea). *Mar. Pollut. Bull.* 184, 114057.
- Cegolon, L., Petranich, E., Pavoni, E., et al., 2023. Concentration of mercury in human hair and associated factors in residents of the Gulf of Trieste (north-eastern Italy). *Environ. Sci. Pollut. Res.* 30, 21425–21437.
- Compeau, G.C., Bartha, R., 1985. Sulfate-reducing bacteria: principal methylators of mercury in anoxic estuarine sediment. *Appl. Environ. Microbiol.* 50 (2), 498–502.
- Cossa, D., Knoery, J., Banaru, D., Harmelin-Vivien, M., Sonke, J.E., Hedgecock, I.M., 2022. Mediterranean mercury assessment 2022: an updated budget, health consequences, and research perspectives. *Environ. Sci. Technol.* 56, 3840–3862.
- Covelli, S., Piani, R., Kotnik, J., Horvat, M., Faganeli, J., Brambati, A., 2006. Behaviour of hg species in a microtidal deltaic system: the Isonzo river mouth (northern Adriatic Sea). *Sci. Total Environ.* 368, 210–223.
- Covelli, S., Piani, R., Acquavita, A., Predonzani, S., Faganeli, J., 2007. Transport and dispersion of particulate hg associated to a river plume in coastal northern Adriatic environments. *Mar. Pollut. Bull.* 55, 436–450.
- Covelli, S., Acquavita, A., Piani, R., Predonzani, S., De Vittor, C., 2009. Recent contamination of mercury in an estuarine environment (Marano lagoon, northern Adriatic, Italy). *Estuar. Coast. Shelf Sci.* 82, 273–284.
- Davidson, P.W., Myers, G.J., Cox, C., Axtell, C., Shamlaye, C., Sloane-Reeves, J., et al., 1998. Effects of prenatal and postnatal methylmercury exposure from fish consumption on neurodevelopment. *JAMA* 280, 701–707.
- De Almeida, R.P., Gomes Ferrari, R., Neves dos Santos, L., Adam, Conte C., 2019. Mercury in aquatic fauna contamination: a systematic review on its dynamics and potential health risks. *J. Environ. Sci.* 84, 205–218.
- Diez, S., Montuori, P., Pagano, A., Sarnacchiaro, P., Bayona, J.M., Triassi, M., 2008. Hair mercury levels in an urban population from southern Italy: fish consumption as a determinant of exposure. *Environ. Int.* 34, 162–167.
- EFSA Panel on Contaminants in the Food Chain (CONTAM), 2012. Scientific Opinion on the risk for public health related to the presence of mercury and methylmercury in food. *EFSA J.* 10 (12), 2985.
- EU, 2006. European Community. Commission regulation no 1881/2006 of 19 December 2006 setting maximum levels for certain contaminants in foodstuffs. *Off. J. Eur. Union L* 364, 5–24.
- Faganeli, J., Hines, M., Horvat, M., Falnoga, I., Covelli, S., 2014. Methylmercury in the Gulf of Trieste (northern Adriatic Sea): from microbial sources to seafood consumers. *Food Technol. Biotechnol.* 52, 188–197.
- Faganeli, J., Falnoga, I., Horvat, M., Klun, K., Lipej, L., Mazej, D., 2018. Selenium and mercury interactions in apex predators from the Gulf of Trieste (northern Adriatic Sea). *Nutrients* 10, 278.
- Floreani, F., Barago, N., Acquavita, A., Covelli, S., Skert, N., Higuera, P., 2020. Spatial distribution and biomonitoring of atmospheric mercury concentrations over a contaminated coastal lagoon (northern Adriatic, Italy). *Atmosphere* 11, 1280.
- Food & Drug Administration (FDA), 2022. Mercury levels in commercial fish and shellfish (1990–2012). Available from: <https://www.fda.gov/food/metals-and-you-r-food/mercury-levels-commercial-fish-and-shellfish-1990-2012> (last accessed on 12 September 2024).
- Gaggi, C., Zino F., Duccini M., Renzoni A., "Levels of mercury in scalp hair of fishermen and their families from Camara de lobos-Madeira (Portugal): a preliminary study," *Bull. Environ. Contam. Toxicol.* 1996; 56 (6) : 0–865.
- Giangrosso, G., Cammilleri, G., Macaluso, A., Vella, A., D'Orazio, N., Graci, S., et al., 2016. Hair mercury levels detection in fishermen from Sicily (Italy) by ICP-MS method after microwave-assisted digestion. *Bioinorg. Chem. Appl.* 5408014.
- Giani, M., Rampazzo, F., Berto, D., Maggi, C., Mao, A., Horvat, M., Emili, A., Covelli, S., 2012. Bioaccumulation of mercury in reared and wild Ruditapes philippinarum of a Mediterranean lagoon. *Estuar. Coast. Shelf Sci.* 113, 116–125.
- Gilmour, C., Ea, Herry, Mitchell, R., 1992. Sulfate stimulation of mercury methylation in freshwater sediments. *Environ. Sci. Technol.* 26, 2281–2287.
- Gosar, M., Teršič, T., 2012. Environmental geochemistry studies in the area of Idrija mercury mine. *Slovenia Environ. Geochem. Health* 34 (Suppl. 1), 27–41.
- Hines, M., Poitras, E., Covelli, S., Faganeli, J., Emili, A., et al., 2012. Mercury methylation and demethylation in hg-contaminated lagoon sediments (Marano & Grado Lagoons, Italy). *Estuar. Coast. Shelf Sci.* 113, 85–95.
- Hornung, H., 1986. Assessment of mercury pollution in coastal marine sediments and in benthic organisms. In: In "FAO/UNEP/WHO/IOC/IAEA Meeting of the Biogeochemical Cycle of Mercury in the Mediterranean," FAO Fish. Report No. 325 (Suppl.), pp. 104–110. Rome, Italy.
- Hornung, H., Krumholz, B.S., Cohen, Y., 1984. Mercury pollution in sediments, benthic animals, fishes and sediments in Haifa Bay. *Israel Mar. Environ. Res.* 12, 191–208.
- Horvat, M., Covelli, S., Faganeli, J., Logar, M., Mandic, V., Rajar, R., et al., 1999. Mercury in contaminated coastal environments: a case study: the Gulf of Trieste. *Sci. Total Environ.* 237–238, 43–56.
- Hsu-Kim, H., Kucharczyk, K.H., Zhang, T., Deshusses, M.A., 2013. Mechanisms regulating mercury bioavailability for methylating microorganisms in the aquatic environment: a critical review. *Environ. Sci. Technol.* 47 (6), 2441–2456.
- Johansen, P., Mulvad, G., Pedersen, H.S., Hansen, J.C., Riget, F., 2007. Human accumulation of mercury in Greenland. *Sci. Total Environ.* 377 (2–3), 173–178.
- Kierfve, B., 1994. Coastal lagoons. In: Kierfve, B. (Ed.), *Coastal Lagoon Processes*. Elsevier, Amsterdam, pp. 1–8.
- King, J.K., Saunders, F.M., Lee, R.F., Jahnke, R.A., 1999. Coupling mercury methylation rates to sulfate reduction rates in marine sediments. *Environ. Toxicol. Chem.* 18, 1362–1369.
- King, J.K., Harmon, S.M., Fu, T.T., Gladden, J.B., 2022. Mercury removal, methylmercury formation, and sulfate-reducing bacteria profiles in wetland mesocosms. *Chemosphere* 46, 859–870.
- Kirichuk, A.A., Skalny, A.A.R.A.I., Tinkov, A.A., Skalny, A.V., 2020. Arsenic, cadmium, mercury, and lead levels in hair and urine in first-year RUDN university students of different geographic origins. *Environ. Sci. Pollut. Res.* 27, 34348–34356.
- Kosta, L., Ravnik, V., Byrne, A.R., Stirn, J., Dermelj, M., Stegnar, P., 1978. Some trace elements in the waters, marine organisms and sediments of the Adriatic by neutron activation analysis. *J. Radioanal. Chem.* 44, 317–332.
- Lacerda, L.D., Fernandez, M.A., Tanizaki, K.F., Calazans, C.F., 1992. Bioavailability of heavy metals in sediments of two coastal lagoon in Rio de Janeiro. *Brazil Hydrobiol.* 228, 65–70.
- Lavoie, R.A., Jardine, T.D., Chumchal, M.M., Kidd, K.A., Campbell, L.M., 2013. Biomagnification of mercury in aquatic food webs: a worldwide meta-analysis. *Environ. Sci. Technol.* 47, 23.
- Lincoln, R.A., Shine, J.P., Chesney, E.J., Vorhees, D.J., Grandjean, P., Senn, D.B., 2011. Fish consumption and mercury exposure among Louisiana recreational anglers. *Environ. Health Perspect.* 2, 245–251.
- Matthews, A.D., 1983. Mercury content of commercially important fish of the Seychelles, and hair mercury levels of a selected part of the population. *Environ. Res.* 30 (2), 305–312.
- Morton, J., Carolan, V.A., Gardiner, P.H.E., 2002. Removal of exogenously bound elements from human hair by various washing procedures and determination by inductively coupled plasma mass spectrometry. *Anal. Chim. Acta* 455, 23–34.
- Naccari, C., Vella, A., Cicero, N., et al., 2015. Toxic metals in pelagic, benthic and demersal fish species from Mediterranean FAO zone 37. *Bull. Environ. Contam. Toxicol.* 95 (5), 567–573.
- NRC - National Research Council (US), 2000. Committee on the Toxicological Effects of Methylmercury. Toxicological effects of methylmercury, Washington (DC).
- Nuttall, K.L., 2004. Interpreting mercury in blood and urine of individual patients. *Ann. Clin. Lab. Sci.* 34, 235–250.
- Nuttall, K.L., 2006. Interpreting hair mercury levels in individual patients. *Ann. Clin. Lab. Sci.* 36 (3), 248–261.
- Okati, N., Esmaili-Sari, A., 2018a. Determination of mercury daily intake and hair-to-blood mercury concentration ratio in people resident of the coast of the Persian Gulf. *Iran Arch. Environ. Contam. Toxicol.* 74 (1), 140–153.
- Okati, N., Esmaili-Sari, A., 2018b. Hair mercury and risk assessment for consumption of contaminated seafood in residents from the coast of the Persian Gulf. *Iran Environ. Sci. Pollut. Res.* 25, 639–657.
- Parks, J.M., Johs, A., Podar, M., Bridou, R., et al., 2013. The genetic basis for bacterial mercury methylation. *Science* 339 (6125), 1332–1335.
- Piani, R., Covelli, S., Biester, H., 2005. Mercury contamination in Marano lagoon (northern Adriatic Sea, Italy): source identification by analyses of hg phases. *Appl. Geochem.* 20, 1546–1559.
- Podar, M., Gilmour, C.C., Brandt, C.C., Soren, A., Brown, S.D., Crable, B.R., et al., 2015. Global prevalence and distribution of genes and microorganisms involved in mercury methylation. *Sci. Adv.* 1 (9).
- Regione, F.V.G., 2014. I Mestieri della pesca nella Laguna di Marano e Grado: Criteri e modalità di esercizio dell'attività di pesca professionale. https://www.regione.fvg.it/rafvig/export/sites/default/RAFVG/economia-imprese/pesca-acquacoltura/FOG_LIA12/allegati/Allegato_A.pdf.
- Renzoni, A., 1992. Comparative observations on levels of mercury in scalp hair of humans from different islands. *Environ. Manag.* 16 (5), 597–602.
- Renzoni, A., Zino, F., Franchi, E., 1998. Mercury levels along the food chain and risk for exposed populations. *Environ. Res.* 77, 68–72.
- RFVG, 1991. Regione Autonoma Friuli Venezia Giulia – USL n. 8 Bassa Friulana. In: La qualità delle acque della Laguna di Marano (Regional Technical Report).
- Tang, W.L., Liu, Y.R., Guan, W.Y., Zhong, H., Qu, X.M., Zhang, T., 2020. Understanding mercury methylation in the changing environment: recent advances in assessing microbial methylators and mercury bioavailability. *Sci. Total Environ.* 714, 136827.
- Trdin, A., Snoj Tratnik, J., Mazej, D., Fajon, V., Kršnik, M., et al., 2019. Mercury speciation in prenatal exposure in Slovenian and Croatian population - PHIME study. *Environ. Res.* 177, 108627.
- U.S. EPA, 1998. Method 7473 (SW-846): Mercury in Solids and Solutions by Thermal Decomposition, Amalgamation, and Atomic Absorption Spectrophotometry. Revision 0. Washington, DC.
- Valent, F., Mariuz, M., Bin, M., Little, A., Mazej, D., Tognin, V., et al., 2013. Associations of prenatal mercury exposure from maternal fish consumption and polyunsaturated fatty acids with child neurodevelopment: a prospective cohort study in Italy. *J. Epidemiol.* 23, 360–370.
- Virtanen, J.K., Vuotilainen, S., Rissanen, T.H., Mursu, J., Tuomainen, T.P., Korhonen, M. J., et al., 2005. Mercury, fish oils, and risk of acute coronary events and cardiovascular disease, coronary heart disease, and all-cause mortality in men in eastern Finland. *Arterioscler. Thromb. Vasc. Biol.* 25, 228–233.
- Vishnivetskaya, T.A., Hu, H., Van Nostrand, J.D., Wymore, A.M., Xu, X., Qiu, G., 2018. Microbial community structure with trends in methylation gene diversity and abundance in mercury contaminated rice paddy soils in Guizhou. *China Environ. Sci. Process. Impacts* 20 (4), 673–685.
- Weihe, P., Grandjean, P., Jørgensen, P.J., 2005. Application of hair-mercury analysis to determine the impact of a seafood advisory. *Environ. Res.* 97 (2), 201–208.