Occupational exposure to mercury from cinnabar enriched sand in workers of Grado Beach, Gulf of Trieste (North-eastern Italy, upper Adriatic Sea)

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ABSTRACT

Health and safety of occupations entailing extensive skin contact with cinnabar-enriched sand in beaches of Friuli-Venezia Giulia (FVG) Region (North-eastern Italy) have been questioned for possible skin absorption of mercury (Hg). One hundred mg hair was collected from the occipital scalp of 50 male workers of Grado beach and 121 males from FVG general population. Factors associated with hair Hg content were investigated by multivariable logistic (considering Hg levels >1 vs ≤1 mg/kg) and log-transformed linear regression. The median hair concentration of Hg in male beach workers was 0.70 (IQR = 0.42; 1.34) mg/kg, lower than FVG general population's [1.29 (IQR = 0.87–2.06) mg/kg ($p < 0.001$)]. In both regression models the hair Hg increased with fish consumption, both among beach workers of Grado and FVG general population. The mean Hg levels in beach workers of Grado fell within an acceptable range, not requiring restrictions of their occupational activities.

1. Background

After Almadén (Spain), Idrija (Western Slovenia) is the second largest mercury (Hg) deposit in the world, discovered back in 1490 and operating as a mine until its closure in 1995 (Spanish Ministry of Culture).

Over 12 million tons of cinnabar were excavated in almost 500 years, corresponding to 150,000 tons of Hg extracted and 38,000 tons of the element lost in the environment as Hg vapour dispersed in air or as particles in association with soils and fluvial sediments as well as mining residues (Gosar et al., 1997). The area impacted by mine's operation has been drained by the Idrijca River, tributary to the Isonzo/Soča River (Fig. 1), which is the main water and sediment supply to the Gulf of Trieste (Friuli-Venezia Giulia region, FVG), located in the north-eastern sector of the Adriatic Sea (Italy) (Covelli et al., 2001).

As a result, the concentrations of Hg in marine sediments of the Gulf of Trieste are the highest (>30 mg/kg) across the entire Mediterranean Sea (Covelli et al., 2001).

Mercury is mostly present in detrital form (cinnabar, HgS) in sandy–silty sediments close to the mouth of Isonzo River and surrounding beaches (Biester et al., 2000; Covelli et al., 2001), where sediments showed Hg concentrations around 25–30 mg/kg, exponentially decreasing towards the open sea (Covelli et al., 2001). In the nearby beach resort of Grado (Fig. 1), further South-West of the Isonzo river mouth, Hg concentration in sands was found to be higher than the local regional background (0.13 mg/kg) (Covelli et al., 2006), varying between 0.17 and 6.07 (Mean = 1.99 ± 1.85) mg/kg in 15 sampling points and retrieved at different depths (from top 50 cm to 200 cm deep) from four selected sampling points (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. For this reason, the presence of Hg in the environment of Grado beach was questioned by local authorities as a potential threat to human health. In spite of the high concentration of Hg found in the sediments of the Gulf of Trieste, as well as in the sands of Grado coastline, results from speciation studies reported that the main Hg chemical forms in the sediments – mainly cinnabar or Hg sulphides – are not easily remobilisable (Biester et al., 2000; Covelli et al., 2021).

Abbreviations: MΩ·cm, Microhm centimeter; Milli-Q Water, water purified using a Millipore Milli-Q lab water system; HCl_{conc}, Concentrated hydrochloric acid.

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Fig. 1. (a–d) The area of concern, including Idrjia (a–b), the mouth of Isonzo River (b–c), Grado beach (a–c), the Marano and Grado Lagoon (c), the location of the dismissed Snia-Viscosa chlor-alkali plant (c) and the mouth of Aussa-Corno River (c).

c

d

Fig. 1. (*continued*).

On the other hand, the main exposures to Hg for the general population are methylmercury (MeHg) from seafood, inorganic mercury (I-Hg) from food, and vapour mercury (Hg^0) from dental amalgams (Björkman et al., 2007).

Most Hg flowing to the sea from draining rivers is elemental (metallic) Hg (Hg^0), a chemical species naturally existing only in liquid form at room temperature. Although it can rapidly turn to vapour (a highly avid and toxic form for the gray matter of the cerebral and cerebellar cortices), ingestion of elemental Hg in liquid form poses little health hazard to humans, since its absorption by the human gastro-intestinal system is limited (bioavailability <0.01 %) and living organisms can get rid of it quickly (WHOI; Duck-Park and Zheng, 2012). Absorption of elemental Hg by the human body requires its oxidation into inorganic divalent ion (Hg^{2+}) (Clarkson, 1997).

Nonetheless, vapour Hg^0 can be inhaled by lungs and retained in the body from the atmosphere or dental amalgam fillings following chewing (Sallsten et al., 1996), nocturnal bruxism (Isacsson et al., 1997) or whitening compounds for teeth (Robertello et al., 1999). As far as the Gulf of Trieste is concerned, Barago et al. (2020) reported that gaseous elemental mercury (GEM) concentrations do not reach levels of concern for the local population. GEM levels varied from <LOD (2.0 ng m^{-3}) to 48.5 ng m^{-3} (mean 2.7 ng m^{-3}), with no significant differences found among sites. A clear daily pattern emerged, with maximum values reached just after sunset.

MeHg – which has a methyl group (CH_3^+) attached to the Hg atom – is the chemical form accumulating to toxic levels in fishes (WHOI). Conversion of Hg to MeHg is likely a biotic process, but little more is known about this reaction. As fish, neither phytoplankton nor zooplankton probably methylate Hg. However, some bacterial species, as those living in seafloor sediments along coasts and on continental shelves (WHOI), do produce MeHg as a byproduct of their respiration process. When converted to MeHg, Hg diffuses into phytoplankton, crossing the food chain in ever-accumulating quantities. As an example, large predator fishes such as tuna contain about 10 million times as much MeHg as the water surrounding them (WHOI). Since fish consumption is a major exposure route to MeHg through gastro-intestinal absorption, and given the Hg contamination of the northern Adriatic Sea, tuna fishing has been banned along the coast of FVG region.

Nonetheless, occupational activities entailing extensive skin contact with cinnabar-enriched beach sand along FVG coast have also been questioned for potential Hg exposure through skin absorption. Mercury exposure can in fact result from ingestion of soil through hand to mouth habits, consumption of polluted fish or water, dermal adherence due to wading/swimming activity or inhalation of volatile Hg (USEPA, 2011; Guney et al., 2013; Elumalai et al., 2022). Indeed, albeit skin absorption of Hg following exposure to Hg^0 vapour is negligible (Hursh et al., 1989), it has been reported in volunteers following application of a solution of Hg chloride 0.1 % (Silberberg, 1968) and at other concentrations (Baranowska-Dutkiewicz, 1982). Skin absorption of Hg is documented in people using lightening creams containing Hg (Chan, 2011; Ho et al., 2017, Pramanik et al., 2021) and cases of Hg poisoning associated with creams containing high amount of Hg (12,000 μg/g) as MeHg (Mudan et al., 2019) or calomel have been reported (Copan et al., 2015). The application of these creams causes increase of Hg in urine, reaching values above the biological occupational limits (20 μg/g creatinine) (Chan, 2011). However, the introduction of Hg in creams is not allowed in the European Union and USA and its limits are fixed to 1 ppm (1 mg/kg) only if its presence is unavoidable under good manufacturing practice, even though Hg containing creams can be purchased illegally and their application may have potential detrimental effects in subjects using them for long periods (Pramanik et al., 2021).

Albeit in vitro skin contact with contaminated soil caused Hg permeation of about 1.5 % (Moody et al., 2009), Sartorelli et al. (2022) did not find a significant skin absorption of Hg after exposure to Hg contaminated soil containing inorganic Hg

Furthermore, oral intake of water during swimming was the major exposure route for metals (arsenic, chromium) in a Swedish study estimating exposure to contaminated sediments during recreational activities at a public bathing place in an urban environment (outside Stockhlom), whereas skin absorption was estimated to be more important for polycyclic aromatic hydrocarbons (Filipsson et al., 2009). In the latter study, in women swimming 0.5 h/day for 16 days/year the mean water intake was estimated to be equal to 0.028 L/h and the mean sediment intake 50 mg/day, whereas the reasonable maximum exposure was 0.072 L/h for water intake and 100 mg/day for sediment intake (Filipsson et al., 2009). Considering a male working as a beach lifeguard for 5 months/year, the above estimates could increase ten times and using Hg concentrations of sands in the beaches of Grado, ranging from 0.17 to 6.17 mg/kg, the cumulative intake of Hg during five summer months in the worst scenario could be close to the permissible limits of MeHg in fishery products established by European Regulation (EC) no. 1881/2006 (Eur Lex, n.d.), fixed at 0.5 mg/kg for fish and fish muscle, and 1 mg/kg for big species as sharks, swordfishes, tunas, monkfishes, sturgeons, among other.

Lastly, according to a literature search in Google Scholar ad PubMed, Hg exposure was never investigated in beach workers.

1.1. Aims

This study aimed at assessing whether workers of Grado beach (FVG Region) may be occupationally exposed to Hg, after adjusting for confounding factors.

2. Methods

This study was approved by the ethical committee of FVG (CEUR n. 092/2018, 23.03.2018).

All male workers employed at the beach resort of Grado ($N = 50$) in

Table 1
Main characteristics of study subjects expressed as continuous variables: median (interquartile range and mean ± standard deviation in male Grado beach workers and in males from the general population of the Friuli Venezia Giulia Region. Mann-Whitney *p*-value for the comparison between columns. M = missing information.

Factors	50 male beach workers	121 males from the general population	*p*-Value
Hair mercury concentration (mg/kg) (M: 20)	0.70 (0.42; 1.34) 1.20 ± 1.66	1.29 (0.87; 2.06) 1.85 ± 1.79	<0.001
Urine Hg concentration (μg/g) (M: 1)	0.47 (0.21; 1.56) 1.20 ± 1.62		
Hg creatinuria ratio (M: 1) (μg/g)	0.37 (0.16; 1.01) 0.84 ± 1.05		
Age (years)	43 (26; 52) 40.6 ± 14.0	32 (20; 52.5) 36.2 ± 20.2	0.119
BMI (kg/m^2)	24 (22; 26) 24.0 ± 3.4	22 (20; 24) 22.4 ± 4.0	0.004
Job seniority (months) (M: 4)	32 (12; 128) 72.3 ± 84.1		
Water intake (L/day) (M: 13)	2.9 ± 1.4 3 (2; 4)	1.4 ± 0.6 1 (1; 2)	<0.001
Fish intake (number of servings/months) (M: 6)	4 (1; 4) 4.9 ± 4.3	4 (2; 4) 3.8 ± 2.4	0.625

Table 2
Main characteristics of study subjects expressed as frequency variables: strata and distribution in male Grado beach workers and in males from the general population of the Friuli-Venezia Giulia (FVG) Region. Number and column percentage (%); chi-square p-value for the comparison between columns; M = missing information.

Factors		Strata	50 male beach workers	121 males from the general population	p-Value
Hair mercury concentration (mg/fg) (M: 20)		<1.1	20 (66.7)	42 (34.7)	0.095
		1.1–2.0	6(20.0)	45 (37.2)	
		>2	4 (13.3)	34 (28.1)	
Age (years)		<26	12 (24.0)	44 (36.7)	0.291
		26–43	16 (32.0)	30 (25.0)	
		44–53	11 (22.0)	17 (14.2)	
		54/+	11 (22.0)	29 (24.2)	
BMI (kg/m^2)		<25	33 (66.0)	94 (80.3)	0.120
		25–30	15 (30.0)	19 (16.2)	
		>30	2 (4.0)	4 (3.4)	
Job task		Life guard	19 (38.0)		
		Beach workers	27 (54.0)		
		Sand therapists	4 (8.0)		
Job seniority (months) (M: 4)		<12	15 (32.6)		
		13–36	12 (21.7)		
		37–72	9 (17.4)		
		73+	13 (28.6)		
Residence area		Industrial	23 (46.0)	5 (4.2)	<0.001
		Urban	26 (52.0)	84 (70.0)	
		Rural	1 (2.0)	31 (25.8)	
Smoking		Non (ex) smoker	26 (52.0)	109 (90.8)	<0.001
		<15 cig/day	19 (38.0)	10 (8.3)	
		15+ sig/day	5 (10.0)	1 (0.8)	
Wine consumption (liter/day) (M: 1)		No	34 (69.4)	72 (60.5)	0.234
		<0.5	14 (28.6)	46 (38.7)	
		0.5+	1 (2.0)	1 (0.8)	
Water intake (liter/day) (M: 13)		<1	6 (15.8)	42 (67.7)	<0.001
		1–2	11 (29.0)	16 (25.8)	
		2–3	11 (29.0)	0	
		3+	10 (26.3)	4 (6.5)	
Supplements		No	48 (90.6)	88 (73.3)	0.019
		Yes	5 (9.4)	32 (26.7)	
Skin cream use		No	23 (46.9)	58 (48.7)	0.832
		Yes	26 (53.1)	61 (51.3)	
Contact lenses		No	44 (88.0)	97 (80.8)	0.258
		Yes	6 (12.0)	23 (19.2)	
History of kidney disease		No	49 (98.0)	115 (95.8)	0.485
		Yes	1 (2.0)	5 (4.2)	
Fish intake (number servings/months) (M: 6)		<4	23 (43.4)	56 (47.9)	0.061
		4	13 (24.5)	41 (35.0)	
		8	7 (13.2)	11 (9.4)	
		>8	10 (18.9)	9 (7.7)	
Fish type (M:9)	Fresh	No	30 (73.2)	70 (57.9)	0.081
		Yes	66 (26.)	11 (26.8)	
	Frozen	No	11 (26.8)	43 (35.5)	0.307
		Yes	126 (66.3)	30 (73.2)	
	Canned	No	13 (31.7)	27 (22.3)	0.228
		Yes	145 (76.3)	28 (68.3)	
Fish size (M:3)	Big*	No	37 (78.7)	105 (86.8)	0.195
		Yes	30 (15.2)	10 (21.3)	
	Small/medium**	No	32 (68.1)	93 (76.9)	0.242
		Yes	52 (26.3)	15 (31.9)	
	Shell/cray fish$	No	41 (87.2)	95 (78.5)	0.196
		Yes	36 (18.2)	6 (12.8)	
Bruxism		No	42 (84.0)	100 (86.2)	0.711
		Yes	8 (16.0)	16 (13.8)	
Chewing gum consumption		No	48 (96.0)	104 (88.1)	0.112
		Yes	2 (4.0)	14 (11.9)	
Dental filling amalgams		0	26 (74.3)	34 (44.7)	0.010
		1–3	6 (17.1)	20 (26.3)	
		4+	3 (8.6)	22 (29.0)	

* Sword fish, tuna, cod.
** Anchovies, sardines, sea bass, sea bream, ribon, gilt-head bream, grey mullet, mullet, plaice, conger.
$ Scampi, prawn, shrimp, sea cicada, crab, lobster, sea crayfish, clams, mussels, squids, octopus.

summer 2018 were recruited and 100 mg of their occipital scalp hair as well as a urine sample were collected in September of the same year. Scalp hair collection was limited to 30 beach workers, since 20 of them were bald. One hundred and twenty-one males from the general population living in the same costal area of FVG were recruited as a control group by convenience sampling on 24–26 September 2021, through an outdoor gazebo placed in the main square of Trieste, the major city of FVG Region, during "Trieste Next" celebrations, a traditonal annual festival dedicated to the dissemination of scientific research. All subjects were also asked to complete a self-reported questionnaire (Supplementary file S1) collecting extensive socio-demographic, life-style and medical history information. Due to logistic limitations and the recruitment approach, urinary sample could not be collected from the general population of FVG.

Moreover, four superficial samples of sand were collected from Grado beach in order to detect total Hg concentration and its chemical form.

2.1. Chemical analysis

Total Hg in hair and sandy samples was analysed by a Direct Mercury Analyzer (DMA-80, Milestone, Sorisole, Italy), according to the Environmental Protection Agency (EPA) Method 7473 (EPA, 1998). The limit of detection (LOD) was approximately 0.0015 ng. The accuracy of the methods for analytical determination of Hg was checked using three certified reference materials (CRMs) for a better representation of the results, PACS-3 (2.98 ± 0.36 mg/kg; Marine Sediment CRM, NRCC, Canada), MESS-4 (0.09 ± 0.04 mg/kg; Marine Sediment CRM, NRCC, Canada) and ERM-DB001 (0.365 ± 0.028 mg/kg; Human Hair CRM, European Reference Materials). Acceptable recoveries were obtained, ranging between 100 and 104 %, 95 and 104 % and 97 and 104 %, respectively.

In order to identify the respective Hg chemical form, an aliquot of each sandy sample was analysed by a pyrolytic technique. This technique is based on the gradual heating of solid sample (0.5 °C/s) from ambient temperature to 710 °C which releases Hg within definite temperature intervals depending on its chemical form (Petranich et al., 2022). This study used a serial RA-915 M Zeeman mercury atomic absorption spectrometer coupled with a PYRO-915+ pyrolysis attachment (Lumex Instruments), which allows for the heating of the sample and continuous monitoring of the releasing Hg (Mashyanov et al., 2017). The software RAPID provided real-time visualization and a record of the Hg release process from the sample with a response time of 1 s. The final result is a thermogram with the signal as a function of the temperature.

Mercury in urine samples was measured by means of CV-AFS (Mercury Analytic Jena) using a method without enrichment. However, to dose urinary Hg concentrations, each sample was digested with a solution composed by 0.54 g KBr into 50 mL of HCl_{conc} and 0.76 g $KBrO_3$ adding slowly. A volume of 0.1 mL of this solution was added to 1 mL of each urine sample and diluted to a final volume of 10 mL adding Milli-Q water (18.2 MΩ·cm). Thereafter, the sample was maintained for at least 40 min at room temperature. Prior to the analysis, 5 μL of $NH_2OH{\cdot}HCl$ was added to 10 mL of sample to remove free halogens. For sample preparation the limit of detection was found to be 5 ng/L. Urinary Hg values were corrected for creatinine for comparison with the occupational biological limits recommended by ACGIH American Conference of Governmental Industrial Hygenist (2018).

2.2. Statistical analysis

Mean (M) ± standard deviation (SD), median with interquartile range (IQR), number and percentage of hair Hg (expressed in mg/kg) as well as urinary Hg/creatinine ratio (expressed as μg/g) by explanatory factor was estimated. Comparison between continuous data were performed using Mann-Whitney test, as data were not normally distributed. Comparison between proportions was done by chi-square test.

A logistic regression model was employed to investigate the relationship between hair Hg concentration expressed as a binary outcome (>1 vs ≤1 mg/kg) and a group of predictors selected by backward stepwise procedure among all variables shown in Tables 1 and 2. Results were expressed as odds ratio (OR), with 95 % confidence interval (95% CI) and respective stratum specific *p*-value.

A log transformation was undertaken to make the uneven distribution of hair Hg close to normal for a linear regression model (Supplementary Files S2–S4). A multivariable linear regression was then fitted to model the relationship between log-transformed hair Hg and explanatory variables chosen by backward stepwise regression, expressing the results as regression coefficients (RC) with 95 % confidence interval (95%CI).

A *p*-value lower than 0.05 was the threshold considered for statistical significance. Missing valued were excluded and complete case analysis was performed. Stata14.2 (Stata Corporation, College Station, Texas, USA) was employed for the statistical analysis.

3. Results

3.1. Descriptive results

The main characteristics of study subjects – 50 male beach workers and 121 males selected from the general population of FVG – are shown in both Table 1 (factors expressed as continuous variables) and Table 2 (factors expressed as frequency variables).

Table 1 shows medians (with interquartile range) and means (±standard deviation) because the variables had often an uneven distribution. The hair Hg concentration was significantly ($p < 0.001$) higher in the general population than in the occupational sample (Fig. 2). The finding was strengthened, in spite of 20 missing values corresponding to the number of bald beach workers, by the lack of significant differences between bald and hairy beach workers regarding urinary Hg concentrations or any other variable shown in Tables 1 and 2 (Supplementary File 5). As can be seen from Fig. 3, the correlation coefficient between Hg/creatininuria ratio and hair Hg was rather low ($R^2 = 0.250$). As per protocol, urinary concentration of Hg and occupational seniority were not investigated in subjects from the general population. Interestingly, the two groups had the same age and amount of daily fish intake, whereas significant differences were found for body mass index, BMI ($p < 0.004$) and water intake ($p < 0.001$), both being higher in the occupational group.

Table 2 shows the strata in which the variables were broken down, the distribution of variables with number and column percentage by group, and the chi-square test *p*-value. The lower value of Hg concentration in hair of beach workers (Table 1) was confirmed in Table 2, even though the statistical significance was borderline.

The distribution of BMI was shifted towards high values among workers of Grado beach, although the level of significance was borderline. Job task and seniority were not investigated in the general population. Thirty-eight percent (=19/50) worked as beach lifeguards, 54.0 % (=27/50) as beach service workers and 4 as sand therapists. Beach workers were more likely to live in an industrial area ($p < 0.001$) and to be a smoker ($p < 0.001$). While wine consumption was similar between the two groups, among beach workers water intake was much greater (*p*

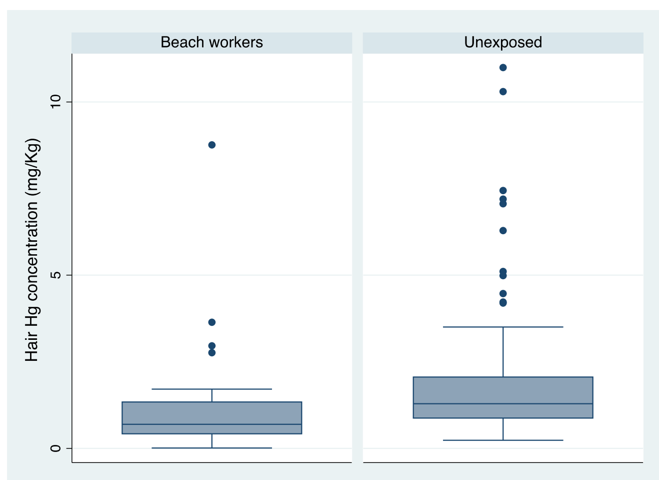

Fig. 2. Box plot displaying the distribution of 50 male beach workers of Grado examined in 2018 vs. 121 males from the general population of FriuliVenezia Giulia sampled in 2021.

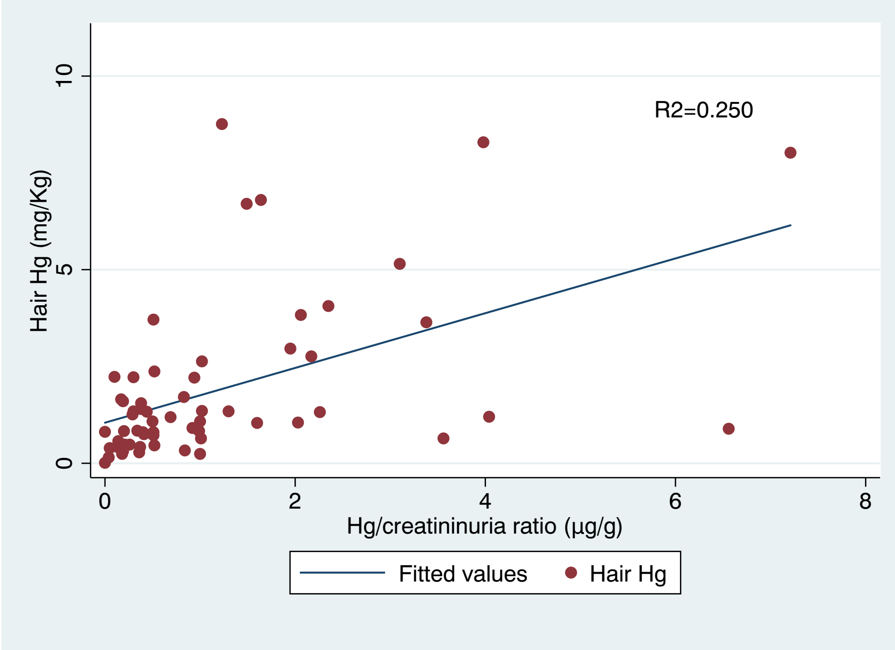

Fig. 3. Scatterplot displaying the predicted values of hair mercury (Hg) by Hg/creatininuria ratio. R^2 = correlation coefficient.

< 0.001) and use of supplements slightly larger ($p < 0.019$). No differences were found between the two groups in terms of use of skin cream, contact lenses, chewing gum consumption, history of kidney disease and dental filling amalgam. Fish ingestion was slightly lower among beach workers ($p < 0.061$); however, the distributions by fish type and fish size were very similar between the two groups.

3.2. Analytical results

As can be seen from Table 3, both age and fish intake increased the probability of hair concentration of Hg > 1 mg/kg, whereas working as beach workers decreases such risk. Consumption of about 8 monthly fish servings or employment as beach workers significantly increased or, respectively, decreased the risk ($p < 0.05$). It can also be noted that working as beach lifeguards reduced the risk, although the *p*-value was equal to 0.057; such result could be considered as borderline significant. Similarly, another borderline significant factor could be the intake ($p = 0.061$) of about 4 monthly fish servings, doubling the risk. Although being statistically significant, the OR for age was very low because it expressed the effect of a one-unit increase of the predictor, namely one year of age.

Lastly, as can be seen in Table 3, the results of the same multivariable logistic analysis showed that employment as beach workers decreased the body burden of Hg, after adjusting for the increasing effect of age

Table 3
Multivariate linear regression for Hg concentration in hair (outcome) expressed as logarithm: terms that passed the F test to enter the model; strata; regression coefficient (RC) with 95 % confidence interval (95%CI) and *p*-value (P). Multivariable model fitted onto 146 complete (analysis) observations.

Terms	Strata	RC	95%CI	P
Age (years)	Linear term	−0.01	−0.00; 0.01	0.056
Monthly fish servings (number)	<4	Reference	–	
	4	0.40	0.11; 0.69	0.008
	8	0.99	0.52; 1.47	<0.001
	>8	0.51	0.05; 0.96	0.030
Occupational status	Unexposed	Reference	–	
	Beach lifeguards	−0.88	−1.40; −0.35	0.001
	Beach workers	−0.71	−1.10; −0.32	<0.001
	Sand therapists	−1.20	−2.77; 0.37	0.133

Table 4
Multivariate logistic regression for Hg concentration in hair (outcome) expressed as dichotomous variable (>1 vs. ≤1 mg/kg): terms that passed the F test to enter the model; strata; odds ratio (OR) with 95 % confidence interval (95%CI) and *p*-value (P). Multivariable model fitted onto 145 complete (analysis) observations.

Terms	Strata	OR	95%CI	P
Age (years)	Linear term	1.02	1.00; 1.04	0.023
Monthly fish servings (number)	<4	Reference	–	
	4	2.18	0.96; 4.93	0.061
	8	8.28	1.51; 45.5	0.015
	>8	3.00	0.79; 11.4	0.107
Occupational status	Unexposed	Reference	–	
	Beach lifeguards	0.23	0.05; 1.06	0.057
	Beach workers	0.16	0.05; 0.50	0.002
	Sand therapists	1	–	1

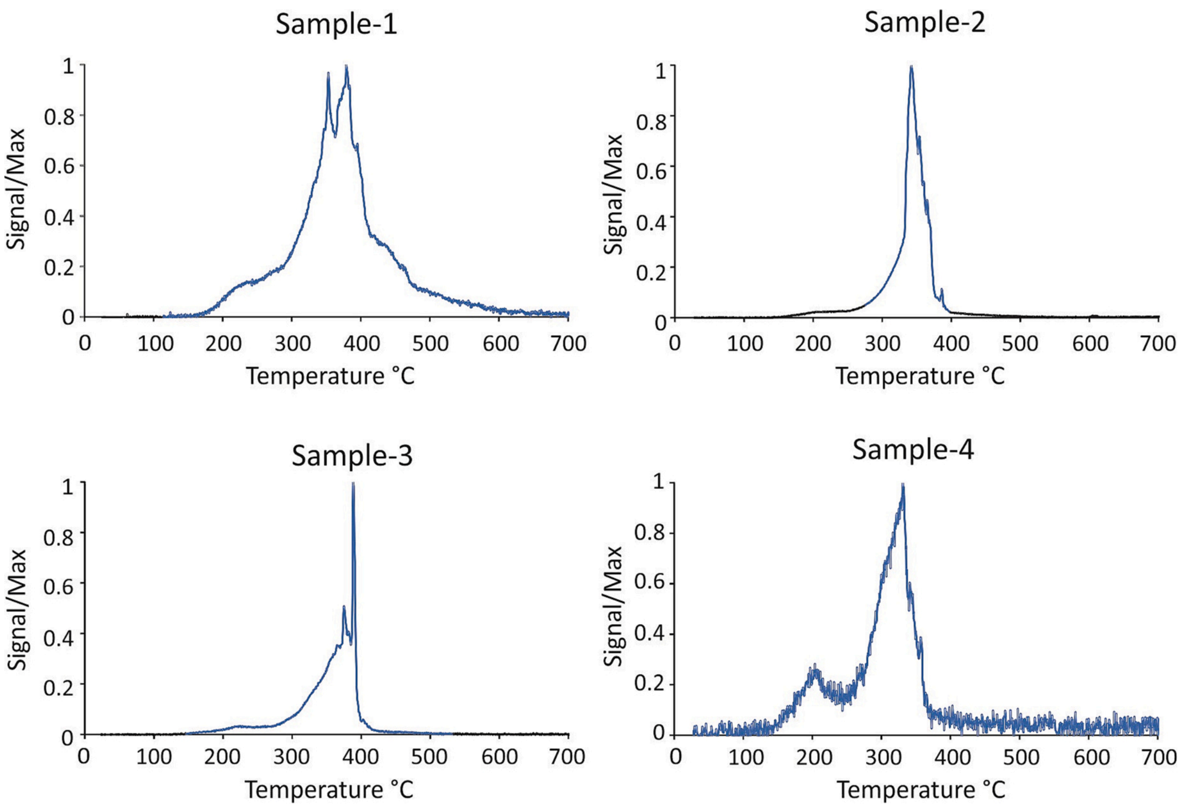

Fig. 4. Maximum temperature (Celsius degree, X-axis) for the release of the totality of Hg (Signal/Max, Y-axis) from the four sandy samples (1 to 4) collected from Grado beach.

and fish intake.

The multivariable linear regression analysis (Table 4) confirmed the results obtained by the logistic regression model, with mean hair Hg concentration increasing with fish consumption and decreasing among lifeguards and workers employed in general beach services. As can be seen from Supplementary file S6, the distribution of the residual of the outcome was fairly linear.

Mercury concentration in sandy samples ranged between 0.53 ± 0.02 (Sample-4) and 6.07 ± 0.09 mg/kg (Sample-1). The Sample-2 and Sample-3 showed central values, equal to 4.50 ± 0.08 and 2.75 ± 0.08 mg/kg, respectively (Supplementary file S7).

All thermo-desorbed sandy samples evidenced the maximum temperature for Hg release, corresponding to the majority or totality of Hg released from the sample considered, varying between 331 and 388 °C (Supplementary file S7). Only Sample-4 showed a small secondary temperature peak at 204 °C (Fig. 4).

4. Discussion

4.1. Key findings

Our study investigated hair Hg concentration in a group of workers of Grado beach (FVG Region) to assess whether absorption of the metal increased with exposure to sand contaminated by high concentrations of Hg, mainly as cinnabar (Covelli et al., 2001, 2021). Workers were investigated at the end of the summer season (September) and their data were compared to the male general population living in the same costal area. Mercury median values in hair were significantly lower in beach workers (0.70 mg/kg; IQR = 0.42–1.34) than the general population (1.29 mg/kg; IQR = 0.87; 2.06) and fish intake confirmed to be the main factor influencing Hg content in human hair.

Although urinary Hg levels are reportedly influenced also by residence area, area of dental amalgams and smoking (Apostoli et al., 2002), in the present study hair Hg content was associated only with fish intake and job tasks.

4.2. Interpretation of findings

Although elevated levels of blood as well as urinary Hg and MeHg have been linked to increased diastolic blood pressure and hypertension (Tang et al., 2022), the major health effects of Hg are on the kidney (major target organ for IHg) and the central nervous system (targeted by MeHg as well as Hg^0), where the metal can cause a range of acute and chronic consequences (Duck-Park and Zheng, 2012, Guzzi and La Porta, 2008). Following metabolisation by microbiota of the bowel, MeHg combined with L-cysteine can cross the endothelial cells of the blood–-brain barrier, accumulating in the brain, where it is rapidly converted into its inorganic form (IHg) and retained for long time (Guzzi and La Porta, 2008).

Although massive environmental releases – as those occurred in Minamata (Japan) in 1953 or Iraq in 1971–72 – are infrequent nowadays, chronic low dose exposure, mainly from ingestion of contaminated fish, can still occur (Guzzi and La Porta, 2008; Ye et al., 2016). Detrimental neurological effects of MeHg are due to loss of neuronal cells in specific anatomical regions of the brain, starting with paresthesia at low dose exposure, potentially progressing to ataxia, cerebral palsy, deafness, blindness and dysarthria. Neurological symptoms are more likely to affect fetuses during gestation, potentially evolving into mental retardation. In adults, the health effects of MeHg follow a latency period depending on dose and length of exposure to the metal (Guzzi and La Porta, 2008).

WHO recommends to maintain mean hair Hg concentrations <1.0 mg/kg in scalp hair (WHO, 2007), since higher concentrations are associated with increased risk of neurological side effects (NRC, 2000). The reference daily intake of MeHg established the US Environmental Protection Agency (USEPA) is 0.1 μg/kg (USEPA, 2011, 2021; Wong et al., 2022).

According to USEPA, Hg concentration in sediments is an indicator of health and safety on beach sands (USEPA, 2021) and poses eco-toxicological risks to the biota of an environment (Elumalai et al., 2022). The impact of Hg on human health is influenced by its chemical form (elemental, organic or inorganic), exposure dose, exposure route, individual susceptibility and environmental factors (Goldblum et al., 2006). In particular, due to low solubility, Hg released by cinnabar is very limited (product constant ksp = 10–55.9 ~ −50.9 for the reaction $HgS(s) = Hg^{2+} + S^{2-}$) (Drott et al., 2013; Jiang et al., 2016).

Mercury concentrations from 7 beaches South of Urban (South Africa) ranged between 0.62 mg/kg and 4.88 mg/kg dw and the average Hg sediment concentrations from each of those beaches ranged from 1.1 to 2.36 mg/kg dw. In the latter study only 13 % samplings were below the permissible Hg limit set by the European Union for sediments (0.8 mg/kg) (Horvart and Kotnik, 2019) and Hg concentrations of the most polluted resort were attributable to industrial pollution, wastewaters drained by rivers and illegal dumping (Elumalai et al., 2022).

The Marano and Grado Lagoon is divided into two basins, Marano and Grado, featured by different salinity and environmental as well as hydrological characteristics (Acquavita and Bettoso, 2018). Mercury input in the northern Adriatic Sea is primarily due to the Isonzo River, predominantly as cinnabar (Biester et al., 2000; Covelli et al., 2021), although it could partly be attributable also to industrial activities of a chlor-alkali plant (Snia-Viscosa) dismissed in 2008, located slightly inland and discharging its wastewaters in the central sector of the Marano and Grado Lagoon through the Aussa-Corno River (Piani et al., 2005; Covelli et al., 2009) (Fig. 1). Nevertheless, a progressively decreasing Hg gradient was reported in coastal sediments from the Eastern part (>11 mg/kg) – near Grado and the Isonzo river mouth – towards West (0.7 mg/kg), with a value of ~5 mg/kg in the central sector of the lagoon (Acquavita et al., 2012). Moreover, before the above industrial plant suppressed the production of chlor-alkali in 2008, Hg pollution in the central sector of the lagoon was mainly due to cinnabar enriched suspensions from the Isonzo River, whereas near the mouth of Aussa-Corno River up to 98 % total Hg was found to be bound in no-cinnabar forms (Piani et al., 2005).

According to the results obtained by Petranich et al. (2022) on the thermo-desorption of pure Hg compounds, the maximum temperature for Hg release from Idrija cinnabar (α-HgS), mixed with carbonate sediment, fell in the range 345–357 °C, which exactly correspond to the desorption thermograms of the four samples of carbonate sand considered (Fig. 4), especially if compared with Sample-3. Regarding the Sample-4, the temperature for Hg release detected at 204 °C could be attributed to meta-cinnabar (β-HgS), as reported by Petranich et al. (2022). Moreover, a similar finding was also obtained from marine sediments of the Gulf of Trieste (Covelli et al., 2021; Petranich et al., 2022), where Hg was mainly found in both sulphide forms (α-HgS and β-HgS) from which the metal is not easily removable in the dissolved phase.

Whilst in the above South African study a total exposure hazard index was estimated by probabilistic risk assessment combining dermal contact, ingestion and inhalation exposure route, the present study relied on Hg concentration in hair as an objective indicator of chronic exposure to the metal (Elumalai et al., 2022). Human hair is the ideal biomarker for establishing long-standing exposure to MeHg (Koenigsmark et al., 2021; Thompson et al., 2014; Esteban-López et al., 2022), taking also into account its relatively low cost and the non-invasive sampling procedure (Wang et al., 2021). In human scalps, MeHg from blood accumulates in hair by binding with sulfhydryl groups of cheratin, providing an ongoing indication of length of exposure to Hg by typical growth rate of human hair of about 1 cm per month.

By contrast, urinary Hg is a biomarker predominantly for environmental exposure to Hg^0 and IHg, albeit part of urinary Hg derives from de-methylation of MeHg in blood from food (Abe et al., 1995; CDC – NHANES, 2021; Castaño, 2015; Koenigsmark et al., 2021; WHO (World Health Organization), 1990). Albeit MeHg in the body combines with sulfur atoms of thiol ligands to form complexes soluble in water, urinary excretion of MeHg is negligible (Guzzi and La Porta, 2008) and should be considered only if hair measurement is not available. Furthermore, the combination of urine and hair does not improve the prediction provided only by hair (Esteban-López et al., 2022).

Nevertheless, hair assessment is not free from downsides, since it is impossible to differentiate between Hg incorporated during hair growth and the metal deposited from external sources (Morton et al., 2004). Mercury in hair could in fact be the result of both dietary and external exposure, which may occur via application of shampoos/cosmetics or by contact with inorganic Hg present in the environment, for example contaminated waters during swimming (Morton et al., 2004). Moreover, hair analysis can be frequently affected by measurement variability (Nuttal, 2006).

From a toxicological perspective, hair Hg concentrations in the present study were well below the 10 mg/kg threshold recommended by UNEP in 2008 as derived-non-observed-adverse-effect-level (NOAEL) (UNEP, 2008a, b). Likewise, total exposure hazard index (HI) value was <1 both in adults (0.11) and children (0.10) in the above South African study, indicating that no adverse health effects were expected (Elumalai et al., 2022). However, in the present study 33.3 % beach workers and 65.3 % subjects sampled from the general population still had hair concentrations exceeding the limit recommended by WHO for children and pregnant women (1 mg/kg). According to the open literature, residents of coastal areas tend to have elevated Hg hair concentrations, mainly due to higher fish intake (Lincoln et al., 2011; Okati and Esmaili-Sari, 2018a; Kirichuk et al., 2020; Bonsignore et al., 2016), as confirmed also in the present study. Moreover, concentration of hair Hg reportedly increased in specific categories of maritime workers more likely to consume higher amounts of fish, as Sicilian (6.45 ± 7.03 mg/kg) or Iranian (5.76 mg/Kg) fishermen (Giangrosso et al., 2016; Okati and Esmaili-sari, 2018b). Median levels of hair Hg (0.70 mg/kg) among workers of Grado beach were lower than males' from the general population of the FVG Region but rather similar to those (0.704 mg/kg) reported in another study on 1308 mother-child dyads delivering at Trieste maternity hospital (Barbone et al., 2019).

A lower level of hair Hg in beach workers with higher daily water intake (55.6 % beach workers drinking 2 L per day vs 6.5 % of general population, $p < 0.001$) may be explained by improved renal clearance.

The median value of urinary Hg corrected for creatininuria – equal to 0.37 μg/g (IQR = 0.21–1.56) – confirmed the low level of Hg in beach workers, close to the reference values for urinary Hg (0.16; 95%CI: 0.16–0.17 μg/L) reported by the most recent National Health and Nutrition Examination Survey (NHAMES) (So et al., 2021]), although well under the occupational exposure index recommended by ACGIH (20 μg/g) and the estimates for the entirety of Italy (0.79 μg/g creatinine; 5°–95° percentiles = 0.21–3.2 μg/g creatinine respectively) reported by the Italian Society of Threshold Values (Soleo et al., 2003). Of note, the median urinary values of Hg/g creatininuria in residents of Genoa (a coastal city in North-Western Italy) was 1.06 μg/g (5°–95° percentiles = 0.12–6.04 μg/g respectively) (Soleo et al., 2003).

Another aspect to be considered in the present survey is the significant lower number of dental amalgams in beach workers (25.7 %) compared to general population group (55.3 %), since it is well known that the presence of amalgams can be a potential source of inorganic Hg (Apostoli et al., 2002; Factor-Litvak et al., 2003; Bates, 2006), absorbed following oxidation of elemental Hg vapour (Hg^0) released by dental amalgams, with chewing. Although we did not find an association between Hg concentration in hair and number of dental fillings amalgams, a finding consistent with other studies (Esteban-López et al., 2022; Díez et al., 2008), an investigation on 60 children reported higher urine Hg levels in those with amalgam fillings (Levy et al., 2004).

5. Conclusions

Our study provided evidence that beach workers of Grado had Hg concentration in hair similar to males from the general population of FVG, suggesting no increased risk of absorption of the metal through skin contact with cinnabar contaminated sand or ingestion of seawater. The latter finding was further endorsed following thermo-desorption of sandy samples collected from Grado beach, where Hg was found to be present mainly as cinnabar and metacinnabar, two poorly soluble chemical forms, whereby inorganic Hg is not easily removable and, therefore, not bioavailable.

The main factor influencing Hg concentration in human hair was fish consumption, a finding consistent with the open literature).

Follow up studies on larger samples, possibly involving other nearby beach resorts along the coasts of the FVG Region are recommended to confirm our findings.

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Ethical statement

Approval to conduct this study was granted by the ethical committee of Friuli Venezia Giulia Region (CEUR n. 092/2018, 23.03.2018).

Data availability statement

The datasets generated and analyzed during the current study are not publicly available, since they were purposively collected by the authors for the present study, but are available from the corresponding author on reasonable request.

CRediT authorship contribution statement

Luca Cegolon: Methodology, Formal analysis, Data curation, Writing – original draft. **Giuseppe Mastrangelo:** Methodology, Formal analysis, Data curation, Writing – original draft. **Stefano Covelli:** Investigation, Data curation, Writing – original draft. **Elisa Petranich:** Investigation, Data curation, Formal analysis, Writing - original draft. **Elena Pavoni:** Investigation, Data curation, Writing – review & editing. **Francesca Larese Filon:** Methodology, Investigation, Data curation, Writing – original draft.

Declaration of competing interest

None to declare.

Data availability

Data will be made available on request.

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