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1 **Human mercury exposure levels and fish consumption at the French**

2 **Riviera**

3 **Mariia V. Petrova^{a*}, Mélanie Ourgaud^a, Joana R. H. Boavida^{a,b}, Aurèlie**

4 **Dufour^a, Javier A. Tesán Onrubia^a, Aurelia Lozingot^a, Lars-Eric**

5 **Heimbürger-Boavida^{a*}**

6 a. Aix Marseille Université, CNRS/INSU, Université de Toulon, IRD, Mediterranean
7 Institute of Oceanography (MIO) UM 110, 13288 Marseille, France

8 b. Center of Marine Sciences (CCMAR), University of Algarve, 8005-139 Faro,
9 Portugal

10 *Corresponding author: lars-eric.heimburger@mio.osupytheas.fr

11

12 **Abstract**

13 Humans are exposed to methylmercury (MeHg), a bioaccumulative neurotoxin,
14 mainly through the consumption of marine fish. Several studies showed that
15 high MeHg exposure can lead to neurological damage. This is particularly
16 relevant for pregnant women, because MeHg exposure negatively impacts foetal
17 development. Populations living near the sea are generally at increased
18 exposure risk due to higher consumption of fish and seafood. Here, we present
19 the first study of MeHg exposure levels of the population living at the French
20 Riviera, using mercury (Hg) concentrations in hair as a proxy for MeHg
21 exposure. We found that older people that consume more fish presented the
22 highest hair Hg concentrations. Compared to other Mediterranean bordering
23 countries and other European countries, the southern France population is
24 among those with high MeHg exposure (median for women of childbearing age
25 is $0.56 \mu\text{g g}^{-1}$). A global implementation of the Minamata Convention is
26 necessary to lower MeHg exposure of the population.

27 ***Key words:*** *hair, methylmercury, Southern France*

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34 **1. Introduction**

35 Methylmercury (MeHg) is a bioaccumulative neurotoxin, which is more
36 toxic than inorganic mercury (NRC/NAS, 2000). It has been consistently
37 observed that MeHg is biomagnifying to harmful levels along the marine food
38 webs with increasingly greater MeHg proportion (close to 100%) in top-
39 predators (Lavoie et al., 2013, Chen et al., 2018, Ourgaud et al., 2018).

40 Humans are exposed to MeHg mainly through their diet, especially through
41 the consumption of fish and other products from aquatic environment
42 (Schoeman et al., 2009, Castaño et al., 2015, Pérez et al., 2019, Sunderland et
43 al., 2018). There is a global trend of increased fish consumption, which may
44 increase the human risk of exposure to trace elements, including Hg (FAO,
45 2016). Worldwide, annual per capita fish consumption grew from 9.9 to over
46 20 kg between 1960 and 2015, respectively (FAO, 2016). In France, the
47 consumption of fish is high (annual per capita fish consumption ranges from 30
48 to 60 kg, FAO, 2012, Vieira et al., 2015) and seafood has always been an
49 important part of the Mediterranean population diet (Castaño et al., 2015; Faget,
50 2009; Metro et al., 2017).

51 The neurotoxicity of MeHg was first described for the Minamata incident, in
52 Japan, in 1956 (Kurland et al., 1960). High-level human MeHg exposure after
53 release of MeHg from a chemical factory led to fatalities and devastating
54 neurological damage in Minamata (Harada, 1995). Particularly, foetuses are
55 considered much more sensitive to MeHg exposure (NRC/NAS, 2000).

56 Consumption of fish contaminated with MeHg by pregnant women has resulted
57 in serious damage of the central nervous system such as mental retardation,
58 cerebral palsy, blindness and deafness (NRC/NAS, 2000). Since 1970, several
59 large-scale epidemiological studies showed that even low-level chronic MeHg
60 exposure by pregnant women leads to poor neurodevelopment of children
61 (decreasing of fine motor-adaptive function, reduction in IQ, attention deficit
62 disorder) later in life (Kjellstrom et al., 1986, Julshamn et al., 1987, Myers et
63 al., 2003, Deroma et al., 2013). Several studies suggested that MeHg exposure
64 by adults may be associated with an increased risk of cardiovascular diseases,
65 such as hypertension and myocardial infarction (Guallar et al., 2002, Kim et al.,
66 2014). Despite the risks of MeHg exposure, fish consumption has many benefits
67 and should not be avoided altogether from human diets (Egeland and Middaugh,
68 1997, Salvo et al., 2016, Cammilleri et al., 2017, Metro et al, 2017). Fish is a
69 valuable source of proteins, vitamin D, selenium, and other essential elements
70 (Fox et al., 2004, Holden et al., 2008, Roos et al., 2007). Fish provide
71 significant amounts of polyunsaturated fatty acids, which prevent
72 cardiovascular diseases (Simopoulos, 2008, Mozaffarian and Wu, 2011). Thus it
73 is important to balance the risks and benefits of fish consumption (Sonke et al.,
74 2013).

75 Several countries and international organisations have established a
76 reference MeHg dose estimated to be safe or without appreciable risk to health
77 (Tab. 1). Generally, all organisations advise pregnant women, women of

78 childbearing age and young children to limit their consumption of fish known to
79 have high levels of Hg (high trophic level fish such as tuna, shark, swordfish) to
80 1-2 per week. Since October 2013, the Minamata Convention under the
81 auspices of the United Nations Environment Program (UNEP), a global action
82 to protect human health and environment from anthropogenic and mercury
83 emissions, was signed (#Web1). The Minamata Convention is implemented
84 through the control of specific human activities that contribute to widespread
85 Hg pollution. The majority of the European countries ratified the Minamata
86 Convention, while others countries, especially from Northern Africa, were not
87 yet parties to the convention (#Web1).

88 Mercury (Hg) concentrations in hair, urine, tissue, toenails and blood have
89 been widely used as biomarkers for human MeHg exposure (for example,
90 WHO, 2008, Shao et al., 2013, Bonsignore et al., 2015). About 95% of MeHg in
91 fish ingested by humans is absorbed in the human gut (Aberg et al., 1969,
92 Miettinen et al., 1971). After absorption into the bloodstream, MeHg enters the
93 red blood cells and it is distributed throughout the body within hours (Aberg et
94 al., 1969, Clarkson, 1997). Hair incorporates MeHg present in circulating blood
95 during hair formation in the hair follicle (Clarkson et al., 1983).

96 Approximately 95% of measurable Hg in the blood is in methylated form
97 (Sherlock et al., 1984), making MeHg also the dominant species in hair (80%,
98 Cernichiari et al., 1995, Phelps et al., 1980). There are several advantages of
99 using hair: it is non-invasive, samples are easier to obtain without medical

100 support, to transport and store. Despite all advantages, hair analysis is suspected
101 of having several limitations, such as: inter-individual variability in the
102 pharmacokinetics of Hg uptake from blood to hair shaft (NRC/NAS, 2000);
103 inter- and intra-individual variability in hair growth rate, density, colour, and
104 waving (Sakamoto et al., 2004, Ohba et al., 2008); external contamination in the
105 areas where ambient air Hg concentrations are elevated.

106 A correlation between consumption of fish or marine products and Hg levels
107 in hair is typically observed (*e.g.*, Castaño et al., 2015, Den Hond et al., 2015).
108 With the funding of the Consortium to Perform Human Biomonitoring on a
109 European Scale (COPHES) and its demonstration project DEMOCOPHES
110 (DEMONstration of a study to COordinate and Perform Human biomonitoring
111 on a European Scale) 17 European countries provided the baseline information
112 on selected contaminants levels including Hg (Den Hond et al., 2015).
113 However, France did not participate in this project. Thus, the aim of the present
114 study was to provide a first baseline information on the Hg concentrations in
115 human hair at the French Riviera and to complete the European assessment. The
116 main investigations addressed are: (i) the influence of age, gender and fish
117 consumption on Hg concentrations; (ii) a comparison of hair Hg concentrations
118 for women of childbearing age (19 – 45 years old) in French Atlantic *vs*
119 Mediterranean coastal areas; (iii) a comparison of Hg concentrations in hair of
120 women of childbearing age and annual per capita fish consumption along
121 Mediterranean and some European countries (depending if a country is party of

122 the Minamata Convention or not); and (iv) we used this assessment to illustrate
123 the mismatch between the MeHg exposure and a lack of commitment or
124 political interest to sign and implement the Minamata Convention.

125 **2. Materials and methods**

126 **2.1. Sampling design**

127 We organized a public awareness campaign: “Arctic Mediterranean
128 Mercury - AM²” for societal Hg exposure in September 2017 (Fig. 3b). During
129 the outreach events we informed the participants about Hg in the environment
130 and exposure. The participants were then given the opportunity to join our
131 study, and to be informed about their individual MeHg exposure level. A total
132 404 volunteers along the French Mediterranean coastline between Marseille and
133 Villefranche-sur-Mer joined the study (58 % female and 42 % male with ages
134 from 1 to 84 years old). No effort was made in selecting the volunteers
135 according to age, gender, diet, etc.

136 **2.2. Data collection**

137 Hair samples were collected according to the Standard Operation Procedure
138 (SOP) established within the framework of COPHES/DEMOCOPHES (Esteban
139 et al., 2015). Briefly, hair strands (~10 – 30 mm) from each volunteers were
140 collected from the nape of the neck, close to the occipital region of the scalp
141 using titanium scissors, wearing powder free single use nitrile gloves. The grey
142 hairs were not removed for the adult group (Pozebon et al., 2017). Given that

143 hair is a stable material, samples were placed into polyethylene bags until
144 chemical analysis (Pozebon et al., 2017).

145 A standard questionnaire was used to collect data on the age ("Adults": >18
146 years old and "Child": ≤ 18 years old), gender ("Female", "Male") and
147 frequency of fish consumption ('Almost never': ≤ 0.5 fish meals per week,
148 "Once": 1 meal per week, "Twice": 2 meals per week, and 'Often' ≥ 3 meals per
149 week). We also subdivided all participants in smaller age groups (1 – 10; 11 –
150 18; 19 – 24; 25 – 29; 30 – 39; 40 – 49, ≥ 50 years old) to compare with Shao et
151 al. (2013). Volunteers gave their consent to participate to the study.

152 **2.3. Chemical analysis**

153 Mercury measurements were performed on a Hg analyser (atomic
154 adsorption spectrometer, a Leco-AMA 254). A subsample of approximately
155 0.03 – 0.05 g of hair was loaded in nickel boats and placed inside a combustion
156 tube, where the subsample was thermally decomposed (~750°C) into a gaseous
157 form. The evolved gases were then cleaned up from all interfering gases passing
158 through the catalytic compounds. Mercury vapour was then transported to the
159 amalgamator, where all Hg was trapped by gold-plated ceramics. The
160 amalgamator was heated to ~900°C essentially releasing all Hg vapour to the
161 detection system of the atomic absorption spectrometer.

162 The method's detection limit, estimated as three times the standard
163 deviation of the blank samples, was 0.005 µg g⁻¹. The certified reference
164 material (IAEA-086, human hair) was run several times per analytical batch and

165 constantly before starting the measurements, to check the accuracy of the
166 measurements. The measured values were on average, within $\pm 5\%$ of the
167 recommended values. Hair Hg concentrations are given on a fresh weight basis
168 in $\mu\text{g g}^{-1}$.

169 **2.4. Statistical analysis**

170 Differences in hair Hg concentrations were tested with non-parametric tests
171 (Shapiro-Wilk and Figner-Killeen tests indicated departures from normality and
172 homogeneity of variances, respectively, except for gender). Independent 2-
173 group Mann-Whitney U tests were used for age groups and gender. A Kruskal
174 Wallis test One Way Anova by Ranks, followed by a Dunn's Multiple
175 Comparisons test, was used to test for differences in hair Hg concentrations
176 among fish consumption groups. P-values were adjusted with the Benjamini &
177 Hochberg method for Multiple Comparisons ($\alpha = 0.05$). Statistical analyses
178 were performed in R version 3.5.0 (The R Foundation for Statistical Computing,
179 2018).

180

181 **3. Results and discussion**

182 A total of 404 hair samples from participants including children, women of
183 childbearing and non-childbearing age and men have been analysed (Tab. 2).
184 Hair Hg concentrations of participants ranged from 0.01 to $3.98 \mu\text{g g}^{-1}$ with a
185 median of $0.51 \mu\text{g g}^{-1}$ (interquartile range $0.86 - 0.28 \mu\text{g g}^{-1}$). Only one man and
186 one woman of childbearing age exceeded the French ANSES level of $3.2 \mu\text{g g}^{-1}$,

187 while about 5% of women and 4% of men exceeded the US EPA level of 1.4 μg
188 g^{-1} . None of the data exceeded the WHO level of 7 μg g^{-1} . There was no specific
189 Hg contamination source such as gold mining, coal combustion and fluorescent
190 light factories located around the study area, which should exclude elevation of
191 hair Hg concentration through direct MeHg exposure. The measured hair Hg
192 concentrations may be linked to age, gender or diet of the sampled population
193 (Liu et al., 2008, Schoeman et al., 2009, Castaño et al., 2015, Sunderland et al.,
194 2018).

195 **3.1. Influence of age and gender on Hg concentration**

196 Mean Hg concentrations increased significantly with age (Fig. 1). The
197 highest mean concentrations of hair Hg (0.91 ± 0.62 and 0.88 ± 0.66 μg g^{-1})
198 were observed in the 40 – 49 and ≥ 50 age groups, respectively; the lowest hair
199 Hg concentrations were found on the 11 – 18 age group (0.36 ± 0.34 μg g^{-1}).
200 Adult group samples (>18 years old) had significantly higher hair Hg
201 concentration than child group samples (≤ 18 years old) ($W = 28691$, $P\text{-value} <$
202 $2.2\text{e-}16$). It seems apparent that the body burden of Hg increased with age due
203 to regular accumulation. A similar increase with age was observed by Liu et al.
204 (2008), who determined that the hair Hg of a local population in Southern China
205 increased between their twenties and forties. Shao et al. (2013) also showed
206 increasing of mean Hg concentration for Chinese people living at the coast of
207 up to 49 years of age. Moreover, Liu et al. (2008) and Shao et al. (2013) also
208 noted a gradual decreasing of Hg concentrations in people > 50 years old,

209 suggesting zero Hg content in the grey hair, which resulted from the role of
210 sulphur-containing chemicals in the formation of hair pigment (Bou-Olayan and
211 Al-Yakoob, 1994; Al-Majed et al., 2000). Unlike Wakisaka et al. (1990), no
212 significant difference in Hg concentrations in hair samples between men and
213 women was observed ($W = 21429$, $P\text{-value} = 0.01937$).

214 **3.2. Fish consumption rate and Hg concentrations**

215 The lowest mean hair Hg concentration ($0.42 \pm 0.45 \mu\text{g g}^{-1}$, range: 0.02 –
216 $1.41 \mu\text{g g}^{-1}$) was observed on participants which ‘Almost never’ consume fish,
217 corresponding to 23% of the study participants, intermediate (0.59 ± 0.42 and
218 $0.87 \pm 0.66 \mu\text{g g}^{-1}$) concentrations were measured on participants eating fish
219 meals ‘Once’ (46% of participants) and ‘Twice’ (22%) per week, respectively.
220 The highest mean hair Hg concentration ($1.32 \pm 0.89 \mu\text{g g}^{-1}$) was measured for
221 participants that ‘Often’ eat fish (9%; Fig. 2a). Hair Hg concentrations were
222 significantly different among all fish consumption groups (Kruskal-Wallis chi-
223 squared = 73.169, $df = 3$, $P\text{-value} = 8.941\text{e-}16$; multiple comparisons $P\text{-values}$
224 between $1.49\text{e-}3$ to $1.34\text{e-}13$).

225 Previous studies reported that fish consumption rates affect Hg
226 concentrations in human hair. For example, significantly higher Hg
227 concentrations were found for people living in coastal areas from Sicily and
228 China, who often eat fish (4-7 times per week), compared to those who eat fish
229 less frequently (1 – 2 times per week; Giangrosso et al., 2015, Shao et al., 2013).
230 Miklavčič et al., 2014, also noted that the highest exposure levels to Hg were

231 found in coastal populations of Europe, which consume more fish than inland
232 populations.

233 Despite a statistically significant difference between hair Hg concentration
234 for children and adults overall ($W = 28691$, $P\text{-value} < 2.2e-16$), such a
235 difference was not evident between children and adults who consume fish with
236 the same frequency (Fig. 2b). Mean Hg concentrations for children and adults
237 who consume fish 'Almost never' and 'Once' were equivalent, while for a
238 weekly consumption of 'Twice' and 'Often', mean Hg concentrations were
239 lower for children. Such a variation of individual mean Hg concentration has
240 been observed even within families, where children and their mothers with the
241 same fish consumption rate showed different Hg concentrations in hair (Den
242 Hond, 2015). Children are generally characterized by lower Hg concentrations
243 than their mothers, however, Castaño et al. (2015) showed that there is strong
244 correlation between the mother and child in the same family. We are aware that
245 questionnaire answers about individual fish consumption rates do not reflect a
246 precise estimation but rather a "best guess", especially for children. Our broader
247 fish consumption categories were chosen to incorporate some of those
248 uncertainties.

249 **4. Regional (and coastal) variations in hair Hg concentration and fish** 250 **consumption in Europe**

251 Mean Hg concentration in hair for women of childbearing age in the French
252 Mediterranean coast was slightly lower (geometric mean $0.52 \mu\text{g g}^{-1}$) than in the

253 French Atlantic coast (Brittany and Nantes, geometric mean 0.62 and 0.67 $\mu\text{g g}^{-1}$
254 ¹, respectively, Pichery et al., 2012). Previous studies have shown that fish from
255 Mediterranean waters show higher Hg levels than Atlantic fish. For example,
256 bluefin tuna (*Thunnus Thynnus*), which is commonly found in the
257 Mediterranean Sea, was reported to be nearly five times as much concentrated
258 in Hg compared with yellowfin tuna (*Thunnus albacares*), which is absent in
259 Mediterranean Sea (Cammilleri et al., 2018). Significant higher Hg
260 concentrations in Mediterranean fish might lead to higher MeHg exposure by
261 population. Comparison of hair Hg concentrations of the Mediterranean and
262 Atlantic coasts is not obvious in this study due to the absence of quantitative
263 data about fish consumption by populations in the French Atlantic coast.

264 Geometric mean Hg levels in hair for women of childbearing age in the
265 French Mediterranean coast ('Once' per week - 0.62 $\mu\text{g g}^{-1}$, and 'Often' - 1.91
266 $\mu\text{g g}^{-1}$) were higher than for women of childbearing age with similar fish
267 consumption rates sampled inland (Toulouse, South-West of France; 0.44 $\mu\text{g g}^{-1}$
268 and 1.32 $\mu\text{g g}^{-1}$, respectively; Sonke et al., 2013). The French Mediterranean
269 coast compared to inland Toulouse may be related to higher fish consumption
270 from the Mediterranean Sea (Vieira et al., 2015; Faget, 2009), but we cannot
271 draw any conclusion based on our study.

272 There is a widespread difference in MeHg exposure in the European
273 population and the difference is very likely related to consumption of fish and
274 other products from marine environment (Fig. 3a, right panel). Comparing with

275 other countries along the Mediterranean Sea, we found that the Southern France
276 hair Hg concentrations were similar to Hg concentrations found in Italy, Croatia
277 and Albania, but lower than in Spain, Morocco and Greece (Fig. 3c; Miklavčič
278 et al., 2013, Babi et al., 1999, Den Hond et al., 2015, Elhamri et al., 2007, Pérez
279 et al., 2019). According to the European Market Observatory for fisheries and
280 aquaculture products (2017), the annual fish consumption in France is about 34
281 kg per capita, while it is about 45 kg in Spain. Annual fish consumption in
282 Greece and Morocco is much lower than in France, 17.3 kg and 12.5 kg per
283 capita, respectively. Higher MeHg exposure by Greek and Moroccan
284 populations could be explained by Hg contamination of local fishes. For
285 example, Elhamri et al. (2007) found up to $1.2 \mu\text{g g}^{-1}$ of Hg in hair for women of
286 childbearing age in the Tetouan province, Morocco. This was related to Hg
287 release into the environment from several chloralkali plants which used Hg cell
288 technology. The released Hg contributed to contamination of local fishes and
289 lead to higher MeHg exposure for local populations, including in women of
290 childbearing age.

291 To our knowledge, there is no published data of MeHg exposure levels for
292 African Mediterranean countries, except Morocco (Elhamri et al., 2007) and
293 Egypt (El-baz et al., 2009). These countries do not present high fish
294 consumption rates but we found that there are potential health risks connected
295 with the release of industrial Hg to the environment. For example, the Mercurial
296 Complex of Azzaba, in Algeria, was an area of very active Hg mining until

297 2005 (200-500 t y⁻¹; Hylander and Meili, 2003). This site alone accounts for 1
298 million tons of Hg waste (Ministry of land planning, environment and the city,
299 Algeria, 2012). Moreover, the cement industry potentially remains an important
300 source for Hg emissions in Northern African countries (#Web2).

301 Our data showed that France exhibits greater Hg concentrations in hair for
302 women of childbearing age than other European countries (Fig. 3c) with one
303 exception for Portugal (where Hg concentrations are up to 1.2 µg g⁻¹, Den Hond
304 et al.,2015). This trend is in accordance with the higher annual fish consumption
305 per capita in Portugal (56 kg; European Market Observatory for fisheries and
306 aquaculture products, 2017).

307 We noted that the highest hair Hg concentrations (related to high MeHg
308 exposure) were found along the Mediterranean Sea on countries that are not
309 parties to the Minamata Convention (Fig. 3c). For instance, Spain, which has
310 among the highest annual fish consumption per capita of 45.2 kg, and the
311 highest Hg hair concentration in this study of 1.49 µg g⁻¹ (Den Hond et al.,
312 2015), and Greece, with an annual fish consumption of 17.3 kg per capita and
313 Hg level of 1.20 µg g⁻¹ (Miklavčič et al., 2013), signed the convention on 10
314 October 2013 but did not ratify it. We also noted the lack of data on MeHg
315 exposure levels for Northern African and Middle East countries surrounding the
316 Mediterranean Sea. Taking in account that most of these countries did not ratify
317 the Minamata Convention (Fig. 3c), we propose further research of MeHg
318 exposure for Northern African and Middle East countries.

319 We hypothesize that populations of non-party countries are not informed
320 about the potential health risks of MeHg exposure. Moreover, published data
321 (Elhamri et al., 2007, Miklavčič et al., 2013) showed that, relatively to the
322 Minamata Convention, non-party countries have elevated MeHg exposure even
323 if they have a low annual fish consumption (*i.e.*, Greece and Morocco). In this
324 case, high MeHg exposure may be explained by Hg industrial contamination of
325 local or imported fishes. Non-party countries are characterized by relatively
326 significant Hg emissions to the atmosphere, according to the Data Visualisation
327 on Global Mercury Emissions by Country and Sector, (2018). For example,
328 Greece accounts for more than 6.5 Ton y⁻¹, which is 1.5 Ton y⁻¹ higher than Hg
329 emissions in France (Fig. 3a, left panel). The implementation of the Minamata
330 Convention can benefit party nations in terms of human health and the
331 environment, by banning Hg from industrial processes, using the best available
332 Hg emission-control technologies and wastewater treatment in new plants.

333 The implementation of the Minamata Convention may lead to some
334 economic losses if we consider the transfer from Hg-based to less efficient or
335 costlier industrial applications. For some of the more industrialized European
336 countries, the impact may be more important. For instance, most of Germany's
337 energy demand is supplied by coal-fired power plants, which have now to be
338 modernized to meet the standards according to the Minamata Convention
339 (UNEP GMA, 2018). However, Germany did implement the Minamata
340 Convention early on, since 15 September 2017. Surprisingly, some of the

341 countries with low anthropogenic Hg emissions (lower than 1 Ton y^{-1} , *e.g.*,
342 Albania and Cyprus, Fig. 3a) are not yet a party the Minamata Convention. The
343 adoption of the Minamata Convention for these countries would have little
344 impact on industry and largely be outweighed by the health benefits and
345 associated economics gains. Pichery et al. (2012) highlighted that prenatal
346 MeHg exposure has serious impacts on the life-time productivity and on society
347 due to adverse cognitive and associated economic consequences.

348 **5. Conclusion**

349 Humans are mainly exposed to MeHg when consuming marine fish, in
350 simple words: the more and the bigger fish we eat the more MeHg we take up.
351 In France, in Europe and in most Western countries, protection guidelines have
352 been developed following the Minamata Convention. However, protection
353 guidelines are often too technical, and people tend to either avoid fish
354 completely, at the risk of a polyunsaturated fatty acid-deficiency, or simply not
355 respect them, at the risk of an elevated MeHg exposure.

356 We examined MeHg exposure of people from the French Mediterranean
357 coast, based on Hg concentrations in their hair. We showed that older people
358 that consumed more fish presented higher hair Hg concentrations. We
359 highlighted that very few study participants presented values above the French
360 reference dose and none above the WHO reference dose. Comparison with
361 Mediterranean bordering and European countries showed that the southern
362 France population is among those with higher MeHg exposure.

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