



**HAL**  
open science

## Human mercury exposure levels and fish consumption at the French Riviera

Mariia Petrova, Melanie Ourgaud, Joana R.H. Boavida, Aurélie Dufour, Javier Tesán Onrubia, Aurelia Lozingot, Lars-Eric Heimbürger-Boavida

### ► To cite this version:

Mariia Petrova, Melanie Ourgaud, Joana R.H. Boavida, Aurélie Dufour, Javier Tesán Onrubia, et al.. Human mercury exposure levels and fish consumption at the French Riviera. *Chemosphere*, 2020, 258, pp.127232. 10.1016/j.chemosphere.2020.127232 . hal-02935253

**HAL Id: hal-02935253**

**<https://hal.science/hal-02935253>**

Submitted on 13 Nov 2020

**HAL** is a multi-disciplinary open access archive for the deposit and dissemination of scientific research documents, whether they are published or not. The documents may come from teaching and research institutions in France or abroad, or from public or private research centers.

L'archive ouverte pluridisciplinaire **HAL**, est destinée au dépôt et à la diffusion de documents scientifiques de niveau recherche, publiés ou non, émanant des établissements d'enseignement et de recherche français ou étrangers, des laboratoires publics ou privés.



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*

28

29

30

31

32

33

## 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 - AM<sup>2</sup>” 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<sup>-1</sup>. 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  $\mu\text{g}$   
188  $\text{g}^{-1}$ . None of the data exceeded the WHO level of 7  $\mu\text{g}$   $\text{g}^{-1}$ . 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 ( $0.91 \pm 0.62$  and  $0.88 \pm 0.66$   $\mu\text{g}$   $\text{g}^{-1}$ )  
198 were observed in the 40 – 49 and  $\geq 50$  age groups, respectively; the lowest hair  
199 Hg concentrations were found on the 11 – 18 age group ( $0.36 \pm 0.34$   $\mu\text{g}$   $\text{g}^{-1}$ ).  
200 Adult group samples ( $>18$  years old) had significantly higher hair Hg  
201 concentration than child group samples ( $\leq 18$  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 ( $0.59 \pm 0.42$  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 <sup>1</sup>, 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<sup>-1</sup>; 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<sup>-1</sup>, 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<sup>-1</sup> (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<sup>-1</sup> (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<sup>-1</sup>, which is 1.5 Ton y<sup>-1</sup> 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.

## 363        **Acknowledgments**

364        This study was carried out as a part of The AXA Research Fund funded a  
365        postdoc fellowship project “Levering Knowledge Gaps To Understand And  
366        Anticipate Risk From Methylmercury Exposure”. A large part of this project  
367        was to investigate Hg exposure in the Arctic region. As part of the project we  
368        participated in several oceanographic cruises on the research icebreaker  
369        Polarstern to the central Arctic Ocean and Greenland. The AXA Research Fund  
370        funded then a follow up outreach, the “AM<sup>2</sup>” project. With the support of the  
371        communication teams of Mediterranean Institute of Oceanography (MIO), Aix-  
372        Marseille-University (AMU), Sciences of the Universe Observatory Pytheas  
373        (OSU Pytheas) and the French National Center for Scientific Research (CNRS),  
374        we shared impressions from those arctic expeditions, in concert with an  
375        awareness rising campaign for societal Hg exposure with the broad public  
376        during the event of *Septembre en Mer 2017*. This study forms part of the  
377        CONTAM project of MIO. The authors are grateful to the expedition sailing  
378        boat François and Sophie Cleuxis; the team of Expé 2M and the ports and the  
379        cities of Marseille, Les Embiez Islands, Port-Cros and Porquerolles national  
380        Park, Cannes, Saint Tropez, Villefranche-sur-Mer. The project leading to this  
381        publication has received funding from European Regional Development Fund  
382        under project 1166-39417.

## 383 6. References

- 384 1. Aberg B., Ekman L., Falk R., Greitz U., Persson G., Snihs J.O. (1969)  
385 Metabolism of methyl mercury ( $^{203}\text{Hg}$ ) compounds in man. *Archives of*  
386 *Environmental Health: an International Journal*. 19(4), 478-484.
- 387 2. Al-Majed N.B., Preston M.R. (2000) Factors influencing the total  
388 mercury and methyl mercury in the hair of the fishermen of Kuwait.  
389 *Environmental Pollution*. 109(2), 239-250.
- 390 3. Babi D., Vasjari M., Celo V., Korovesi M. (1999) Some results on Hg  
391 content in hair in different populations in Albania. *Science of the Total*  
392 *Environment*. 259(1-3), 55-60.
- 393 4. Bonsignore M., Andolfi N., Barra M., Madeddu A., Tisano F.,  
394 Ingallinella V., Castorina M., Sprovieri M. (2015) Assessment of mercury  
395 exposure in human populations: A status report from Augusta Bay (southern  
396 Italy). *Environmental Research*. 150, 592-599.
- 397 5. Bou- Olayan A.H., Al- Yakoob S.N. (1994) Mercury in human hair: A  
398 study of residents in Kuwait. *Journal of Environmental Science & Health*  
399 *Part A*. 29(8), 1541-1551.
- 400 6. Cammilleri, G., Vazzana, M., Arizza, V., Giunta, F., Vella, A., Lo Dico,  
401 G., Giaccone V., Giofrè S. V., Giangrosso G., Cicero N., Ferrantelli, V.  
402 (2017) Mercury in fish products: what's the best for consumers between  
403 bluefin tuna and yellowfin tuna? *Natural Product Research*, 32(4), 457-462.
- 404 7. Castaño A., Cutanda F., Esteban M., Pärt P., Navarro C., Gómez S.,  
405 Rosado M., López A., López E., Exley K., Schindler B.K., Govarts E.,  
406 Casteleyn L., Kolossa-Gehring M., Fiddicke U., Koch H.M., Angerer J.,  
407 Hond E.M., Schoeters G.E., Sepai O., Horvat M., Knudsen L.E., Aerts D.,  
408 Joas A., Biot P., Joas R., Jiménez-Guerrero J.A., Díaz G.C., Pirard C.,  
409 Katsonouri A., Černá M., Gutleb A.C., Ligočka D., Reis F.M., Berglund M.,  
410 Lupsa I., Halzlová K., Charlier C.J., Cullen E.A., Hadjipanayis A., Krsková

411 A., Jensen J.F., Nielsen J.K., Schwedler G., Wilhelm M., Rudnai P.,  
412 Kőzépessy S., Davidson F., Fischer M.E., Janasik B., Namorado S., Gurzău  
413 A.E., Jajcaj M., Mazej D., Tratnik J.S., Larsson K., Lehmann A.D., Crettaz  
414 P., Lavranos G., Posada M. (2015). Fish consumption patterns and hair  
415 mercury levels in children and their mothers in 17 EU countries.  
416 *Environmental Research*. 141, 58-68.

417 8. Cernichiari E., Brewer R., Myers G.J., Marsh D.O., Lapham L.W., Cox  
418 C., Shamlaye C.F., Berlin M., Davidson P.W., Clarkson T.W. (1995)  
419 Monitoring methylmercury during pregnancy: maternal hair predicts fetal  
420 brain exposure. *Neurotoxicology*. 16, 705-710.

421 9. Chen M.M., Lopez L., Bhavsar S. P., Sharma S. (2018) What's hot about  
422 mercury? Examining the influence of climate on mercury levels in Ontario  
423 top predator fishes. *Environmental Research*. 162, 63-73.

424 10. Clarkson T.W. (1983) Mercury. *Annual Review of Public Health*. 4, 375-  
425 380.

426 11. Clarkson T.W. (1997) The toxicology of mercury. *Clinical Laboratory  
427 Science Journal*. 34(4), 369-403.

428 12. Cossa D., Harmelin-Vivien M., Mellon-Duval C., Loizeau V., Averty B.,  
429 Crochet S., Chou L., Cadiou J.F. (2012) Influences of Bioavailability,  
430 Trophic Position, and Growth on Methylmercury in Hakes (*Merluccius  
431 merluccius*) from Northwestern Mediterranean and Northeastern Atlantic.  
432 *Environmental Science & Technology*. 46(9), 4885-4893.

433 13. Den Hond E., Govarts E., Willems H., Smolders R., Casteleyn L.,  
434 Kolossa-Gehring M., Schwedler G., Seiwert M., Fiddicke U., Castaño A.,  
435 Esteban M., Angerer J., Koch H.M., Schindler B.K., Sepai O., Exley K.,  
436 Bloemen L., Horvat M., Knudsen L.E., Joas A., Joas R., Biot P., Aerts D.,  
437 Koppen G., Katsonouri A., Hadjipanayis A., Krskova A., Maly M., Mørck  
438 T.A., Rudnai P., Kozepessy S., Mulcahy M., Mannion R., Gutleb A.C.,  
439 Fischer M.E., Ligocka D., Jakubowski M., Reis M.F., Namorado S.,

440 Gurzau A.E., Lupsa I., Halzlova K., Jajcaj M., Mazej D. , López A., Lopez  
441 E., Berglund M., Larsson K., Lehmann A., Crettaz P., Schoeters G. (2015)  
442 First Steps toward Harmonized Human Biomonitoring in Europe:  
443 Demonstration Project to Perform Human Biomonitoring on a European  
444 Scale. *Environ Health Perspect.* 123(3), 255-263.

445 14. Deroma L., Parpinel M., Tognin V., Channoufi L., Tratnik J., Horvat M.,  
446 Valent F., Barbone F. (2013) Neuropsychological assessment at school-age  
447 and prenatal low-level exposure to mercury through fish consumption in an  
448 Italian birth cohort living near a contaminated site. *International Journal of*  
449 *Hygiene and Environmental Health.* 216(4), 486-493.

450 15. Egeland, G.M., Middaugh, J.P. (1997) Balancing Fish Consumption  
451 Benefits with Mercury Exposure. *Science.* 278 (5345), 1904-1905.

452 16. El-baz F., Elhossiny R.M., Elsayed A.B., Gaber G.M. (2009) Hair  
453 mercury measurement in Egyptian autistic children. *The Egyptian Journal of*  
454 *Medical Human Genetics.* 11(2), 135-141.

455 17. Elhamri H., Idrissi L., Coquery M., Azemard S., Abidi A.E., Benlemlih  
456 M., Saghi M., Cubadda F. (2007) Hair mercury levels in relation to fish  
457 consumption in a community of the Moroccan Mediterranean coast. *Food*  
458 *Additives & Contaminants.* 24(11), 1236-1246.

459 18. Esteban M., Schindler B.K., Jiménez J.A., Koch H.M., Angerer J.,  
460 Rosado M., Gómez S., Casteleyn L., Kolossa-Gehring M., Becker K.,  
461 Bloemen L., Schoeters G, Den Hond E., Sepai O., Exley K., Horvat M.,  
462 Knudsen L.E., Joas A., Joas R., Aerts D., Biot P., Borošová D., Davidson F.,  
463 Dumitrascu I., Fischer M.E., Grander M., Janasik B., Jones K., Kašparová L.,  
464 Larssen T., Naray M., Nielsen F., Hohenblum P., Pinto R., Pirard C., Plateel  
465 G., Snoj Tratnik J., Wittsiepe J., Castaño A., EQUAS Reference  
466 Laboratories. (2015) Mercury analysis in hair: Comparability and quality  
467 assessment within the transnational COPHES/DEMOCOPHES project.  
468 *Environmental Research.* 141, 24-30.

- 469 19. Faget D. (2009) Le milieu marin méditerranéen: conflits, usages et  
470 représentations: le cas du golfe de Marseille (début XVIIIe-début XXe  
471 siècles). Thèse de doctorat, Université Aix Marseille 1, 560p.
- 472 20. FOOD AND AGRICULTURE ORGANIZATION OF THE UNITED  
473 NATIONS (FAO). (2012) The state of world fisheries and aquaculture.  
474 Rome. 209p.
- 475 21. FOOD AND AGRICULTURE ORGANIZATION OF THE UNITED  
476 NATIONS (FAO). (2016) The state of world fisheries and aquaculture.  
477 Contribution to food security and nutrition for all. Rome. 200p.
- 478 22. Fox T.E., Van den Heuvel E.G.H.M., Atherton C.A., Dainty J.R., Lewis  
479 D.J., Langford N.J., Crews H.M., Luten J.B., Lorentzen M., Sieling F.W., van  
480 Aken-Schneyder P., Hoek M., Kotterman M.J.J., van Dael P., Fairweather-  
481 Tait S.J. (2004) Bioavailability of selenium from fish, yeast and selenate: a  
482 comparative study in humans using stable isotopes. *European Journal of*  
483 *Clinical Nutrition*. 58(2), 343.
- 484 23. Giangrosso G., Cammilleri G., Macaluso A., Vella A., D'Orazio N.,  
485 Graci S., Lo Dico G.M., Galvano F., Giangrosso M, and Ferrantelli V.  
486 (2015) Hair mercury levels detection in fisherman from Sicily (Italy) by ICP-  
487 MS method after Microwave\_Assisted digestion. Hindawi Publishing  
488 Corporation. *Bioinorganic Chemistry and Applications*, Volume 2016,  
489 Article ID 5408014, 5.
- 490 24. Guallar, E., Sanz-Gallardo, M., Veer, P., Bode, P., Aro, A., Gomez-  
491 Aracena, J., Kark, J., Riemersma, R., Martin-Moreno, J. M., Kok, F.J. (2002)  
492 Mercury, Fish Oils, and the Risk of Myocardial Infarction. *The New England*  
493 *journal of medicine*. 347. 1747-54.
- 494 25. Harada M. (1995) Minamata disease: methylmercury poisoning in Japan  
495 caused by environmental pollution. *Critical Reviews in Toxicology*. 25(1), 1-  
496 24.

- 497 26. Holden, J. M., Lemar, L. E., & Exler, J. (2008) Vitamin D in foods:  
498 development of the US Department of Agriculture database. *The American*  
499 *Journal of Clinical Nutrition*. 87(4), 1092S-1096S.
- 500 27. Hylander L.D., Meili M. (2003) 500 years of mercury production: global  
501 annual inventory by region until 2000 and associated emissions. *Science of*  
502 *the Total Environment*. 304(13), 13-27.
- 503 28. Julshamn K., Andersen A., Ringdal O., Mørkøre J. (1987) Trace elements  
504 intake in the Faroe Islands. I. Element levels in edible parts of pilot whales  
505 (*Globicephalus meleanus*) *Science of the Total Environment*. 65, 53-62.
- 506 29. Kjellstrom T., Kennedy P., Wallis S. and Mantell, C. (1986) Physical and  
507 mental development of children with prenatal exposure to mercury from fish.  
508 Stage 1 preliminary tests at age 4. National Swedish Environmental Board,  
509 Solna. Report number 3080.
- 510 30. Kurland L.T., Faro S.N., Siedler H. (1960) Minamata disease: The  
511 outbreak of a neurologic disorder in Minamata, Japan, and its relationship to  
512 the ingestion of seafood contaminated by mercuric com- pounds. *World*  
513 *Neurology*, 1(5), 370-395.
- 514 31. Lavoie R.A., Jardine T.D., Chumchal M.M., Kidd K.A., Campbell L.M.,  
515 (2013) Biomagnification of Mercury in Aquatic Food Webs: A Worldwide  
516 Meta-Analysis. *Environmental Science & Technology*. 47(23), 13385-13394.
- 517 32. Liu X., Cheng J., Yuling S., Honda S., Wang L., Liu Z., Sakamoto M.,  
518 Liu Y. (2008) Mercury concentration in hair samples from Chinese people in  
519 coastal cities. *Journal of Environmental Sciences*. 20(10), 1258-1262.
- 520 33. Metro, D., Tardugno, R., Papa, M., Bisignano, C., Manasseri, L.,  
521 Calabrese, G., Gervasi T., Dugo G., Cicero, N. (2017) Adherence to the  
522 Mediterranean diet in a Sicilian student population. *Natural Product*  
523 *Research*. 32(15), 1775-1781.
- 524 34. Miklavčič A., Casetta A., Snoj Tratnik J., Mazej D., Krsnik M., Mariuz  
525 M., Sofianou K., Spirić Z., Barbone F., Horvat M. (2013) Mercury, arsenic



526 and selenium exposure levels in relation to fish consumption in the  
527 Mediterranean area. *Environmental Research*. 120, 7-17.

528 35. Miklavčič A., Kocman, D., Horvat, M. (2014) Human mercury exposure  
529 and effects in Europe. *Environmental Toxicology and Chemistry*. 33(6),  
530 1259-1270.

531 36. Miettinen J.K., Rahola T., Hattula T., Rissanen K., Tillander M. (1971)  
532 Elimination of <sup>203</sup>Hg-methylmercury in man. *Annals of Clinical Research*, 3,  
533 116-122.

534 37. Myers G. J., Davidson P. W., Cox C., Shamlaye C., Palumbo D.,  
535 Cernichiari J., Sloane-Reeves E., Wilding G.E., Kost J., Haung Li-S.,  
536 Clarkson T.W. (2003) Prenatal methylmercury exposure from ocean fish  
537 consumption in the Seychelles child development study. *The Lancet*.  
538 361(9370), 1686-1692.

539 38. Mozaffarian D., Wu J. H. (2011) Omega-3 fatty acids and cardiovascular  
540 disease: effects on risk factors, molecular pathways, and clinical  
541 events. *Journal of the American College of Cardiology*. 58(20), 2047-2067.

542 39. National Research Council (NRC). (2000) Toxicological effects of  
543 methylmercury. Committee on the Toxicological Effects of Methylmercury,  
544 Board on Environmental Studies and Toxicology, Commission on Life  
545 Sciences, National Research Council. The National Academies Press.  
546 Washington, DC. 344p.

547 40. Ohba T., Kurokawa N., Nakai K., Shimada M., Suzuki K., Sugawara N.,  
548 Kameo S., Satoh C., Satoh H. (2008) Permanent waving does not change  
549 mercury concentration in the proximal segment of hair close to scalp. *The*  
550 *Tohoku Journal of Experimental Medicine*, 214(1), 69-78.

551 41. Ourgaud M., Ruitton S., Bourgoigne H., Bustamante P., Churlaud C.,  
552 Guillou G., Lebreton B., Harmelin-Vivien M. L. (2018) Trace elements in a  
553 Mediterranean scorpaenid fish: Bioaccumulation processes and spatial  
554 variations. *Progress in Oceanography*. 163, 184-195.

- 555 42. Pérez R., Suelves T., Molina Y., Corpas-Burgos F., Yusà V. (2019)  
556 Biomonitoring of mercury in hair of children living in the Valencian Region  
557 (Spain). Exposure and risk assessment. *Chemosphere*. 217, 558-566.
- 558 43. Phelps R.W., Clarkson T.W., Kershaw T.G., Wheatley B. (1980)  
559 Interrelationships of blood and hair mercury concentrations in a North  
560 American population exposed to methylmercury. *Archives of Environmental*  
561 *Health: An International Journal*. 35(3), 161-168.
- 562 44. Pichery C., Bellanger M., Zmirou-Navier D., Fréry N., Cordier S., Roue-  
563 LeGall A., Hartemann P., Grandjean P. (2012) Economic evaluation of health  
564 consequences of prenatal methylmercury exposure in France. *Environmental*  
565 *Health*. 11(1), 53.
- 566 45. Pozebon D., Scheffler G., Dressler V. (2017) Elemental Hair analysis: A  
567 review of procedures and application. *Analytica Chimica Acta*. 992, 1-23.
- 568 46. Roos N., Wahab M.A., Chamnan C., Thilsted S.H. (2007) The role of fish  
569 in food-based strategies to combat vitamin A and mineral deficiencies in  
570 developing countries. *The journal of Nutrition*. 137(4), 1106-1109.
- 571 47. Sakamoto M., Kubota M., Liu X.J., Murata K., Nakai K., Satoh, H.  
572 (2004) Maternal and fetal mercury and n-3 polyunsaturated fatty acids as a  
573 risk and benefit of fish consumption to fetus. *Environmental Science and*  
574 *Technology*. 38 (14): 3860-3863.
- 575 48. Salvo A, Cicero N, Vadalà R, Mottese AF, Bua D, Mallamace D,  
576 Giannetto C, Dugo G. (2016) Toxic and essential metals determination in  
577 commercial seafood: *Paracentrotus lividus* by ICP-MS. *Natural Product*  
578 *Research*, 30(6):657–664.
- 579 49. Schoeman K., Bend J. R., Hill J., Nash K., Koren G. (2009) Defining a  
580 lowest observable adverse effect hair concentrations of mercury for  
581 neurodevelopmental effects of prenatal methylmercury exposure through  
582 maternal fish consumption: a systematic review. *Therapeutic Drug*  
583 *Monitoring*. 31(6), 670-682.

- 584 50. Shao D., Cheng Z., Kang Y., Wong M. H. (2013) Hair mercury levels  
585 and food consumption in residents from the Pearl River Delta: South China.  
586 *Food Chemistry*. 136(2), 682-688.
- 587 51. Sherlock J, Hislop J, Newton D, Topping G, Whittle K. (1984) Elevation  
588 of mercury in human blood from controlled chronic ingestion of  
589 methylmercury in fish. *Human Toxicology*. 3(2), 117-131.
- 590 52. Simopoulos A.P. (2008) The importance of the omega-6/omega-3 fatty  
591 acid ratio in cardiovascular disease and other chronic diseases. *Experimental*  
592 *Biology and Medicine*. 233(6), 674-688.
- 593 53. Sonke J.E., Heimbürger L.E., Dommergue A. (2013) Mercury  
594 biogeochemistry: Paradigm shifts, outstanding issues and research needs.  
595 *Comptes Rendus Geoscience*. 345(5-6), 213-224.
- 596 54. Sunderland E.M., Li M., Bullard K. (2018) Decadal Changes in the  
597 Edible Supply of Seafood and Methylmercury Exposure in the United States.  
598 *Environmental Health Perspectives*. 126(1), 017006.
- 599 55. UNEP (United Nations Environment Programme) - WHO (World Health  
600 Organization). (2008) Guidance for identifying populations at risk from  
601 mercury exposure. UNEP DTIE Chemicals Branch and WHO Department of  
602 Food Safety, Zoonoses and Foodborne Diseases, Inter-Organization  
603 Programme for the Sound Management of Chemicals (IOMC). Geneva,  
604 Switzerland, August 2008, DTI/1131/GE. 170p.
- 605 56. UNEP (United Nations Environment Programme). (2019) Global  
606 Mercury Assessment -2018. Geneva, Switzerland. 400p.
- 607 57. Vieira, H.C., Morgado, F., Soares, A. M.V.M., Abreu, S.N. (2015) Fish  
608 consumption recommendations to conform to current advice in regard to  
609 mercury intake. *Environmental Science and Pollution Research*. 22(13),  
610 9595-9602.

- 611 58. Wakisaka I., Yanagihashi T., Sato M., Nakano A. (1990) Factors  
612 contributing to the difference of hair mercury concentrations between the  
613 sexes. *Nihon Eiseigaku Zasshi. Japanese Journal of Hygiene.* 45(2), 654-664.  
614 59. #Web1: Minamata Convention on Mercury, 2020.  
615 [www.mercuryconvention.org](http://www.mercuryconvention.org)  
616 60. #Web2: Pro Global Media Ltd, 2020. [www.globalcement.com](http://www.globalcement.com)