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Total dietary intake of mercury in the Canary Islands, Spain

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Abstract

Estimating the risk associated with dietary intake of heavy metals by consumers is a vital and integral part of regulatory processes. The assessment of exposure to mercury shown in this paper has been performed by means of a study on the whole diet. Total mercury (Hg) levels were determined by cold vapour atomic absorption spectrometry (AAS) in 420 samples of regularly-consumed food and drink. The total Hg concentrations measured in the different groups of food ranged from non-detectable to 119 µg/kg w/w. The fish group had the highest concentrations of total Hg. All groups of food with regulated Hg content showed levels that were lower than the legally set values. The food consumption data used in our analysis were taken from the latest nutritional survey made in the Canary Islands. The estimated total Hg intake of local population (5.7 µg/person/day) did not exceed the PTWI (Provisional Tolerable Weekly Intake) limit of 0.3 mg/week of total mercury (43 µg/person/day) fixed by the Joint FAO/WHO Expert Committee on Food Additives. Fishery products contributed 96% of the total Hg intake. The mean Hg intake for each Island in this

archipelago, formed by seven, has also been calculated. Fuerteventura, Lanzarote and El Hierro are the ones with the highest level of Hg intake (7.0, 7.0 and 6.1 $\mu\text{g}/\text{person}/\text{day}$, respectively). La Palma Island, due to its low fish consumption, had the lowest level of Hg intake (4.5 $\mu\text{g}/\text{person}/\text{day}$), followed by La Gomera (5.4 $\mu\text{g}/\text{person}/\text{day}$), Tenerife (5.5 $\mu\text{g}/\text{person}/\text{day}$) and Gran Canaria (5.6 $\mu\text{g}/\text{person}/\text{day}$). A comparison has been made of the results obtained in this study with those found for other national and international communities.

Keywords: Mercury, AAS-cold vapour, Dietary Intake, Canary Islands.

Introduction

Environmental contamination through heavy metals is a world-wide recognized public health hazard. Among the different environmental sources of Hg, those of industrial origin (chlorine-alkaline plants, paper factories, organic-mercurial fungicides, gold purification processes) stand out as the most dangerous ones, because in the areas destined to industrial spilling there can be found the highest levels of Hg, almost all atmospheric and of great volatility (Hardisson, 1981; Kurasaki et al., 2000). In Brazil, for example, gold mining activities, like *Manaus* and *Mato Grosso*, provide a mercury dietary intake that is very close to the upper limit set by WHO (Favaro et al., 1997). In a general population, mercury dietary intake is mainly influenced by fish consumption (Wilhelm et al., 2005). The WHO International Programme on Chemical Safety established that, during 1980-1988, the contribution of Hg from fishery products varied from 20 to 85% (Galal-Gorchev, 1993). Recently, the increased popularity of fish as a source of dietary proteins and omega-3 fatty acids is expected to raise the risk of methyl mercury induced health problems in the US population (Knobeloch et al., 2006).

The presence of metals in marine sediments and some fish species and their products is a consequence of both, natural and anthropogenic contamination (Herawati et al., 2000; Schuler et al., 2000). Fish and crustaceans, due to their high accumulation of Hg, are often used as indicators of this environmental pollution. Even though an accumulation of mercury in liver for marine organisms like dolphins, seals and porpoises has been reported by many researchers (Rodríguez-López et al., 2001; Vega et al. 2001), a recent study considered the

netted whelk (*Nassarius reticulatus*) as a poor indicator of total Hg contamination (Coelho et al., 2006).

In aquatic environments, inorganic Hg is microbiologically transformed into methyl-mercury (MeHg), a toxic lipophilic organic compound. Consequently, populations with a traditionally high dietary intake of food originating from either fresh water or marine environments, like coastal population, show the highest dietary exposures to mercury and methyl-mercury. The total Hg level is a function of the methyl mercury content, and the Hg content in fish is mainly MeHg. This is, for example, the case of the Adriatic Sea, where industrial pollution has affected both total Hg and methyl-mercury content in seafood (Buzina et al., 1989) as well as the Baffin Inuit living in the Canadian Arctic (Chan et al., 1995). Even in an industrially non-polluted area of the Adriatic Sea (Buzina et al., 1995) it has been observed that 20% of the consumers had a total weekly dietary mercury intake above the PTWI. It has been calculated that people consuming, more than 6 times/week, fish and other seafood products absorb 300 µg of total mercury for every 1559 g of food and 200 µg of methyl-mercury, for every 1365g. In the Songhua River, China (Lin, 1990), 90% of the mercury that has been measured in the fishermen's body is supposed to be provided by polluted fish. Recent studies describe mercury poisonings in Indonesia (Kurasaki et al., 2000) and in India (Ali et al., 2000), where many aquatic resources have been reported to be contaminated by this metal.

Environmental, dietary and occupational exposure to inorganic mercury may cause both clinical and sub-clinical effects (Soleo et al., 2002). It is well known

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3 86 that mercury is a neurotoxic poison that causes neurobehavioral effects
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5 87 (especially on psycho-motor coordination), neuroendocrine and renal damage
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8 88 and gastrointestinal toxicity. Daily intake of methyl mercury is significantly
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10 89 correlated with hair mercury concentrations (Iwasaki et al., 2003).
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15 91 It has been observed that total level of Hg in hair, in toenails and methyl-
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17 92 mercury in blood, often used as biomarkers of the human exposure to this
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19 93 heavy metal, raise when total fish consumption increases (Yoshizawa et al.,
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21 94 2002; Bjornberg et al., 2005). Moreover, a significant dose-effect relationship
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23 95 has been associated with long-term exposure to low doses of organic mercury
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25 96 from usual consumption of marine fish in Italy (Carta et al. (2002) In Canada,
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27 97 (Innis et al., 2006), concluded that children from various population subgroups,
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29 98 like Chinese, initially included in a population not considered at risk, had
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31 99 increased risk levels of neurotoxicity caused by Hg exposure to fish.
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38 101 A high Hg intake coming from non-fatty freshwater fish, its consequent Hg
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40 102 accumulation in the body and the promotion of lipid peroxidation by mercury
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42 103 have been associated with excessive risk of acute myocardial infarction and
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44 104 with death from coronary heart disease in eastern Finnish men (Meltzer et al.
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46 105 1994; Salonen et al., 1995). Although, a recent study has not found any
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48 106 connection between total mercury exposure and the risk of coronary heart
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50 107 disease, a slight relation cannot be ruled out (Yoshizawa et al., 2002).
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57 109 As 80% of human intake of mercury comes from ingestion of marine foods, the
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59 110 Spanish Health Ministry has established the obligation to control levels of
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mercury in national and imported fish and fishery products (Rodríguez-López et al., 2001). In Europe, Hg concentrations in foods are ruled by the European Regulation 466/2001 and its modification, 78/2005. The following tolerable maximum limits have been established: 0.5 mg/kg for fishery products and fish meat and 1.0 mg/kg for certain species like tuna fish, etc.

Taking into account that the population in the Canary Islands reaches 2.000.000 people and is distributed as follows: 10.477 in El Hierro, 85.252 in La Palma, 21.746 in La Gomera, 85.2945 in Tenerife, 80.7049 in Gran Canaria, 89.680 in Fuerteventura and 12.3039 in Lanzarote, in the present work we make a study on the estimated provision to the dietary intake of mercury through consumption of fish, to know the contribution to tolerable provisional weekly intake of Hg in our population. We have made a study of total intake of mercury, with bigger toxicological importance of metil mercury.

Material and Methods

Samples: Sample preparation is a major concern in AAS as it basically governs the analytical performance characteristics, namely, detection limit, accuracy, precision, speed, throughput and cost (Subramanian, 1996). In this study, samples were randomly bought at five different points (shopping major areas present in the Canary Islands, in the Islands with bigger groups of population (Tenerife, Gran Canaria, La Palma, Lanzarote and Fuerteventura). Areas for collection of simples were selected, because these where the places with higher number of buyers and more variety of brands. The total diet has been divided into groups (milk, cheese, yogurt, other dairy products, fish, eggs, red meat,

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3 137 cold meat, viscera, poultry and rabbit, fats-oils, cereals, bakery, legumes, fruits,
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5 138 nuts, vegetables, potatoes, sweets, alcoholic drinks, non-alcoholic drinks).
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8 139 These groups were those established in the latest Nutritional Survey, performed
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10 140 at the Canaries, as the groups with highest consumption in this area between
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12 141 1997 and 1998 (ENCA, 2000.). The ENCA is a survey where food intake has
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14 142 been estimated using a method of food consumption at an individual level.
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16 143 Specifically, the ENCA used two twenty-four-hour recalls – administered in two
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18 144 non consecutive days - and a comprehensive food frequency questionnaire of
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20 145 77 food items. The survey considered ages between 6 and 75 years of 2.025
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22 146 families. The number of analysed samples in each group was twenty,
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26 147 *Analytical methodology:* Total mercury determination can be performed by
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28 148 applying different analytical methods. However, the most widely used technique
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30 149 is AAS - cold vapour atomic absorption spectrometry - due to its simplicity and
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32 150 sensitivity (Hardisson and Lozano, 1985; AOAC, 1990; Hardisson et al., 1999).
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34 151 *Reagents:* Milli-Q deionised water, sulphuric, nitric and chloride Merck p.a.
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36 152 acids, lumatom, stannous chloride in 10% HCl freshly prepared from a Fisher
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38 153 product, certified ACS, NIST SRM 1577a bovine liver, mercury standard
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40 154 solution 1.000 mg/L, Fisher, certified for EAA and Acationox detergent,
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42 155 Sherwood, 2% solution for the washing of the whole glass labware.
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46 156 *Apparatus:* Pye-Unicam SP-1900 atomic absorption spectrometer, equipped
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48 157 with Hg hollow cathode lamp and Hg cold vapour system with 15 x 1 cm quartz
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50 158 cells. The detection limit was fixed, as in previous studies of our research group,
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52 159 at 0.0005 µg/g (Hardisson et al., 1999). The instrumental conditions were set at
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54 160 253.27 nm wavelength, 0.2 nm slit-width and 6 mA lamp current without
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56 161 background correction; PHAXE pressurized reactors; CEM MDS-2000
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162 microwave oven with low pressure Teflon glasses (< 200 psi), Teflon PFA®;
163 PENTA ultrasound bath.

164 *Samples treatment:* Two sample treatments a) and b) were used at first.
165 However, once the results of the recovery tests were compared with the
166 microwave oven digestion method, method b) was selected for the study.
167 Moreover, method b) was faster than method a).

168 Method a: From 0.5 to 3 g of homogenized material were introduced in
169 topaz coloured flasks and 10 mL H₂SO₄ as well as HNO₃ mL were added
170 in cold conditions. The flasks were then, placed, for 10-18 hours, in a stove
171 at 40 ± 5 °C, until total mineralization. Once cooled, degasified in an
172 ultrasound bath and filtered through Albet 240 paper, they were finally
173 adjusted at 100 mL volume with deionised water (19, 20).

174 Method b: From 1 to 2 g of homogenized material were weighed on Teflon
175 pumps. Then, 4 mL HNO₃ and 2 mL H₂SO₄ were added for the sample's
176 digestion in a microwave oven (60-150 psi) for 40 min. Later, the samples
177 were cooled in a fridge for 20-30 min and degasified in an ultrasound bath.
178 Finally, volumes were adjusted at 100 mL with deionised water.

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Total mercury determination: volumes ranging from 10 to 50 mL of the problem solution were added in the reaction flask. Deionised water was then added to obtain 70 mL volumes. From 2 to 5 mL of stannous chloride in 10% HCl were also added. When the air force pump was started and the spectrophotometer lecture became stabilized, the absorption was recorded. In the same way, a calibration plot with a linear range was prepared, with standards of 0.05, 0.10, 0.15, 0.20, 0.25 and 0.30 mg/L of mercury

Recovery Study: the recovery study with a certified material “NIST SRM 1577a bovine liver” is shown and the accuracy of the method proved (table 1) (Díaz et al., 1994; Hardisson et al., 1999). The recovery percentage obtained for the process of digestion with microwave oven (sample treatment method b) was as good as those obtained for sample treatment method a).

Results and discussion

Table 2 shows the mean total Hg contents and standard deviations measured for the different food groups. Among the groups with detectable total Hg concentrations, marine products showed the highest levels. There were eleven food groups with non-detectable Hg concentrations (fat-oils, bakery, legumes, fruits, nuts, vegetables, potatoes, sweets, alcoholic drinks, non-alcoholic drinks). Foods with regulated Hg limits presented Hg levels under the legal values, none of the fish samples went above the mercury maximum tolerance limit fixed for fishery products by European Legislation (0.5 mg/kg).

To estimate the total Hg intake (Table 3), it was necessary to use the food consumption data from the latest Nutritional Survey carried out in the Canaries

(ENCA, 2000). Fishery products are the food group that contributes most to the Hg intake (5.4 µg/day), being 95.8% of the total intake. These results coincide with those obtained by other authors in other Spanish Regions (Schuchmacher et al, 1994; Cuadrado et al, 1995; Llobet et al., 1998) .

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Comparing the intake of the population in the Canaries with the total Hg intake calculated for the population of other Spanish Regions (Table 4), it can be observed that the total Hg dietary intake in the Canary Islands is lower than in Tarragona (Schumacher et al., 1994, Llobet et al., 1998), Andalucía, Madrid, País Vasco and Galicia and almost the same as in Valencia and the last estimation trends observed in Tarragona (Moreiras et al., 1995; Cuadrado et al., 1995; Urieta et al., 1996; Bocio et al., 2005) Because the nourishing habits of every region are different and in the Canary community there is consumed food that possess less concentration of mercury that in the mentioned communities previously, due to the fact that these communities are more industrialized that the Canary community.

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Table 5 shows the Hg intakes in different international populations. Apart from those in Greenland, Baffin Inuit and Canadian Artic populations, Hg dietary intake from local marine food has traditionally exceeded the limit established by FAO/WHO (Chan et al., 1995, Johansen et al., 2000). In table 5 it can be observed that Hg intake in the Canary Islands is lower than in USA, Japan and the United Kingdom and almost the same as in Germany (Boudene, 1990) and Chile (Munoz et al., 2005). As happens in France (Noel et al., 2003), Denmark (Bro et al., 1990; Larsen et al., 2002) and in the Netherlands (Van Dokkum et

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3 230 al., 1989), the mercury intake in the Canary Islands is far below the tolerable
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5 231 limits and therefore, below the concentrations can cause bad effects on health.
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8 232 In the case of Denmark, (Bro et al., 1990) studied the intake of essential and
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10 233 toxic trace elements in random samples of Danish men and obtained a mean
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12 234 dietary intake of mercury below the detection limit. However, (Larsen et al.
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14 235 2002), obtained a mercury intake around the 11% of PTWI, a similar value to
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16 236 the one observed for Slovenia (12,3% of PTWI for mercury) (Erzen et al., 2002)
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18 237 and higher than the average Hg intake (5,6% of the PTWI) estimated for the
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20 238 urban population in Lebanon (Nasreddine et al., 2006).
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26 240 Special attention must be paid on certain populations and age groups. In
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28 241 German children the dietary intake on days of fish consumption was found to be
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30 242 123.7 ng/kg w.w./day (16.6% of PTWI) while it only amounted to 1,7% of the
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32 243 PTWI on days without fish consumption (Wilhelm et al., 1995). In Croatia,
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34 244 Dalmatians population was the only Croatian population consuming more
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36 245 mercury through seafood, approaching the PTWI defined by WHO (Blanusa and
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38 246 Juresa, 2001).
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45 248 To conclude, it is important to point out that food consumption data is essential
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47 249 in the development of risk assessment studies. Therefore, the latest nutritional
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49 250 survey made in the Canary Islands (ENCA, 2000) represents the basis of this
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51 251 dietary intake study. The evaluation of the total Hg dietary intake in the Canary
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53 252 Islands (5.684 µg/day; 39.78 µg/week) reveals that dietary mercury fulfils the
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55 253 PTWI fixed by the FAO/WHO and that there is no risk associated to Hg dietary
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57 254 intake in the Canary Islands. Nevertheless, fish is the main food group
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contributing to the total Hg dietary intake in the Canary Islands. The Island with the lowest fish consumption (La Palma) is also the one with lowest mercury intake. On the other hand, the Island of Fuerteventura shows the highest mercury dietary intake due to the high fish consumption.

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Table 1: Accuracy of the measured concentrations of mercury in NIST Standard				
Reference material (µg/g wet weight)				
Material	N ¹	Concentration		Procedure
		Measured ²	Reported ³	
NIST SRM 1577 a	11	3.6 ± 1.0	4 ± 2	a) H ₂ SO ₄ /HNO ₃
NIST SRM 1577 a	11	3.5 ± 0.8	4 ± 2	b) Microwave

1. Number of samples analysed.

2. Mean values measured in the recovery study.

3. Certified values of reference material.

Table 2: Hg concentrations obtained for the different food groups			
Foodstuff	[Hg] (µg/kg ww)	Foodstuff	[Hg] (µg/kg ww)
Milk	0.25± 0.06	Cereals	0.437 ± 0.200
Cheese	0.37 ± 0.15	Bakery	<0.001
Yogourt	0.26 ± 0.06	Legumes	<0.001
Dairy products	0.30 ± 0.12	Fruit	<0.001
Fishery products	118.9 ± 115.5	Dried Fruits (nuts)	<0.001
Eggs	0.45 ± 0.16	Vegetables	<0.001
Red meat	0.54 ± 0.19	Potatoes	<0.001
Cold Meat	1.45 ± 0.55	Sweets	<0.001
Viscera	1.53 ± 0.63	Alcoholic drinks	<0.001
Poultry and rabbit	0.39 ± 0.15	Non alcoholic drinks	<0.001
Fats and oils	<0.001	Water	<0.001

Detection limit= 0.0005 µg/g

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Table 3: Hg intake from the food groups with detectable [Hg] µg/day in the Canary Islands.

Foodstuff	Canary Islands	Gran-Canaria	Lanzarote	Fuerteventura	Tenerife	La-Palma	La-Gomera	El-Hierro
Milk	0.08	0.08	0.07	0.05	0.07	0.08	0.06	0.07
Cheese	0.01	0.01	0.01	0.01	0.01	0.01	0.01	0.01
Yogurt	0.01	0.01	0.01	0.02	0.01	0.01	0.01	0.01
Other dairy products	0.01	0.01	0.01	0.003	0.01	0.01	0.004	0.01
Fishery products	5.45	5.31	6.50	6.78	5.24	4.26	5.17	5.86
Eggs	0.01	0.01	0.01	0.01	0.01	0.01	0.05	0.02
Red Meat	0.02	0.03	0.03	0.02	0.03	0.03	0.02	0.03
Sausages	0.04	0.04	0.03	0.04	0.04	0.04	0.04	0.04
Viscera	0.001	0.001	0	0.01	0.002	0.003	0	0.001
Poultry and rabbit	0.01	0.01	0.01	0.01	0.01	0.01	0.01	0.01
Cereals	0.05	0.06	0.68	0.04	0.05	0.06	0.05	0.07
Total Hg intake	5.68	5.57	6.74	6.98	5.49	4.50	5.42	6.133

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Table 4: Comparison of this study with results from other Spanish Communities		
Spanish Community	Reference	Hg Intake (µg/day)
Canary Islands	This study	5.69
Madrid	Cuadrado et al., 1995	10
Madrid	Moreiras et al., 1995	9.8
Galicia	Cuadrado et al., 1995	9
Galicia	Moreiras et al., 1995	8.7
Valencia	Cuadrado et al., 1995; Moreiras et al., 1995	4
Andalucía	Moreiras et al., 1995	12.8
Andalucía	Cuadrado et al., 1995	13
Cataluña (Tarragona)	Schuhmacher et al., 1994	16 (only fish)
Cataluña (Tarragona)	Llobet et al., 1998	4.8
Cataluña (Tarragona)		
Male adults	Llobet et al., 2003	21.2
Cataluña (Tarragona)	Bocio et al., 2005	5.3
Basque Country	Urieta et al., 1996	18
Spain	Bosnir et al., 1999	7.26

Table 5: Comparison of Hg Dietary Intake in the Canary Islands and other Countries

Country	Reference	Hg Intake ($\mu\text{g/day}$)
Canary Islands	This study	5.57
USA	Boudene et al., 1990	20
United Kingdom		10
Japan		14-80
Germany		5
Poland		8.71 – 22
The Netherlands	Marzec and Bulinski, 1990	4.9
	Van Dokkum et al., 1989	(18-year-old male)
	Ellen et al., 1990	7
Denmark (34 year old men)	Bro et al., 1990	< Detection limit
France	Scianna, 2000	17.43
	Noel et al., 2003	63
Sweden	Becker and Kumpulainen, 1991	12.6
Croatia	Blanusa and Juresa, 2001	19.0
Canada	Chan et al., 1995	16
Baffin Inuit (women)		435
Canadian Arctic (men)		469
Canada (Grand Manan, Bay of Fundy)	Legrand et al., 2005	0.05 ± 0.04
Canada (St Andrews/St Stephen, Bay of Fundy)		0.03 ± 0.04
Greenland	Johansen et al., 2002	846
La Habana (Cuba)*	Vega et al, 2001	31.8 =
China Songhua River Fishermen	Lin, 1990	1190-2380
China	Yang et al., 1994	50.75
Chile (Santiago)	Munoz et al., 2005	5 $\mu\text{g/day}$

* Methyl-mercury intake for a Cuban adult of 60 kg of body weight when considering a 40 g/day daily fish ingestion.