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Daily Intake of Arsenic, Cadmium, Mercury, and Lead by Consumption of Edible Marine Species

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The daily intake of arsenic (As), cadmium (Cd), mercury (Hg), and lead (Pb) through the consumption of 14 edible marine species by the general population of Catalonia, Spain, was estimated. Health risks derived from this intake were also assessed. In March−April 2005, samples of sardine, tuna, anchovy, mackerel, swordfish, salmon, hake, red mullet, sole, cuttlefish, squid, clam, mussel, and shrimp were randomly acquired in six cities of Catalonia. Concentrations of As, Cd, Hg, and Pb were determined by ICP-MS. On the basis of recent fish and seafood consumption data, the daily intake of these elements was calculated for eight age/sex groups of the population. The highest As concentrations were found in red mullet, 16.6 μg/g of fresh weight, whereas clam and mussel (0.14 and 0.13 μg/g of fresh weight, respectively) were the species with the highest Cd levels. In turn, swordfish (1.93 μg/g of fresh weight) and mussel and salmon (0.15 and 0.10 μg/g of fresh weight) showed the highest concentrations of Hg and Pb, respectively. The highest metal intake through fish and seafood consumption corresponded to As (217.7 μg/day), Cd (1.34 μg/day), and Pb (2.48 μg/day) for male seniors, whereas that of Hg was observed in male adults (9.89 μg/day). The daily intake through fish and seafood consumption of these elements was compared with the provisional tolerable weekly intakes (PTWI). The intakes of As, Cd, Pb, and total Hg by the population of Catalonia were below the respective PTWI values. However, the estimated intake of methylmercury for boys, 1.96 μg/kg/week, was over the PTWI.

KEYWORDS: Arsenic; cadmium; mercury; lead; edible marine species; daily intake; Catalonia, Spain

INTRODUCTION

For most nonoccupationally exposed individuals, diet is the main route of exposure to environmental pollutants. Recently, we performed in Catalonia (Spain) a wide survey to determine the concentrations of various environmental contaminants in a number of foodstuffs (*1*−*5*). The group comprising fish and seafood showed the highest concentrations of arsenic (As), cadmium (Cd), mercury (Hg), and lead (Pb), as well as the greatest contribution to the dietary intake of these elements (*1*). In general terms, the results were in accordance with those reported in other recent studies (*6*−*12*).

Mercury is an element of special concern. Because of the potential content of methylmercury in fish and seafood, the European Commission has recommended some restrictions concerning the consumption of marine species (especially predatory species, which accumulate Hg over a long lifetime). This is particularly important for children, pregnant women, and breast-feeding mothers.

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According to the results of our recent survey (*1*), the contribution of edible marine species to the dietary intake of As, Cd, Hg, and Pb by the population of Catalonia would be potentially important. However, the small number of fresh (sardine, hake, and mussel) and tinned (sardine, tuna) species that were analyzed in that survey (*1*) is undoubtedly an important limiting factor for the purpose of establishing recommendations about human consumption (frequency and size of meals) of fish and seafood. Therefore, to extend those data, in the current study the levels of As, Cd, Hg, and Pb were measured in 14 species of fish and seafood, species that are among the most consumed by the general Spanish population, including that of Catalonia (*13*, *14*). The contribution of this food group to the dietary intake of As, Cd, Hg, and Pb was also determined.

MATERIALS AND METHODS

Sampling. In March−April 2005, samples of fish and seafood were randomly acquired in local markets, large supermarkets, and grocery stores from six important cities (Barcelona, Tarragona, Lleida, Hospitalet de Llobregat, Terrassa, and Girona) of Catalonia, Spain. Rural areas were not included in the selection. Fish and seafood samples consisted of six species of blue fish [sardine (*Sardine pilchardus*), tuna (*Thunnus thynnus*), anchovy (*Engraulis encrasicholus*), mackerel (*Scomber scombrus* L.), swordfish (*Xiphias gladius*), and salmon (*Salmo*

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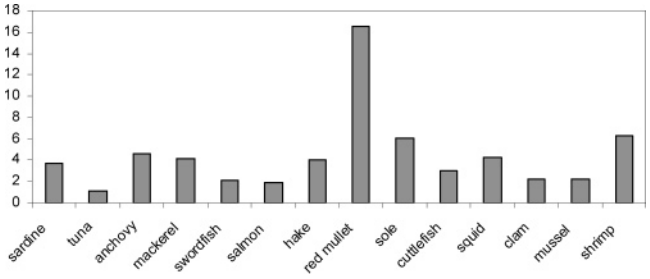

Figure 1. Arsenic concentrations (μg/g of fresh weight) in fish and seafood. Data are given as arithmetic mean values corresponding to three composite samples for each species.

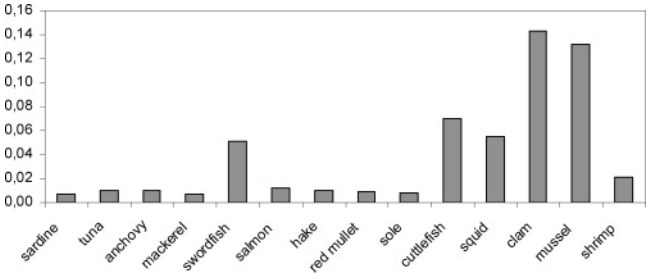

Figure 2. Cadmium concentrations (μg/g of fresh weight) in fish and seafood. Data are given as arithmetic mean values corresponding to three composite samples for each species.

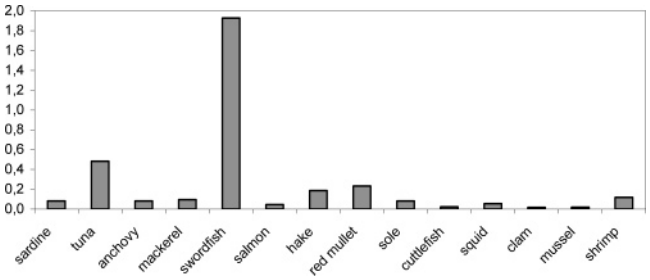

Figure 3. Mercury concentrations (μg/g of fresh weight) in fish and seafood. Data are given as arithmetic mean values corresponding to three composite samples for each species.

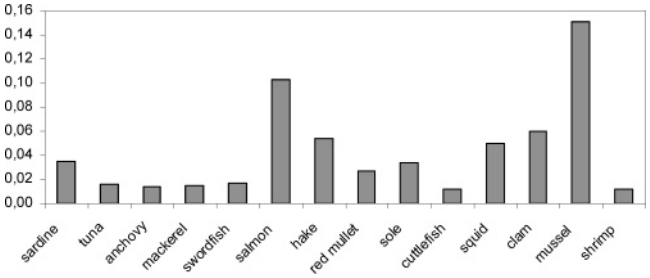

Figure 4. Lead concentrations (μg/g of fresh weight) in fish and seafood. Data are given as arithmetic mean values corresponding to three composite samples for each species.

salar L.)], three species of white fish [hake (*Merluccius merluccius* L.), red mullet (*Mullus surmuletus*), and sole (*Solea vulgaris*)], and five species of cephalopods and shellfish [cuttlefish (*Sepia esculenta*), squid (*Loligo vulgaris*), clam (*Tapes decussatus*), mussel (*Mytilus galloprovincialis* L.), and shrimp (*Penaeus setiferus*)]. For analytical purposes, composites were made up by 20 samples of individuals of each species. For small species (i.e., sardine, anchovy, clam, etc.), the entire edible part of each individual was included to prepare the composite sample. However, for bigger species (i.e., hake, swordfish, or tuna) only fillets of edible parts of each individual were collected and included in the respective composite samples. Three composite samples of each species were analyzed for As, Cd, Hg, and Pb concentrations. For all species, only edible parts were included in the composites.

Analytical Methods and Instrumentation. About 0.5 g of homogenized samples was predigested with 5 mL of 65% nitric acid (Suprapur, E. Merck, Darmstadt, Germany) in Teflon vessels for 8 h at room temperature. Subsequently, solutions were heated at 85 °C for 8 h. On completion of the digestion and after adequate cooling, solutions were filtered and made up to 25 mL with deionized water. Total concentrations of As, Cd, Hg, and Pb were determined by inductively coupled plasma−mass spectrometry (ICP-MS, Perkin-Elmer Elan 6000). Rh was used as internal standard.

Analytical grade reagents were used for blanks and calibration curves, whereas analytical quality was checked against an internationally certified reference material (*Lobster hepatopancreas*, NRC Canada, TORT 2). For sets of every 10 samples, a procedure blank and spike sample, involving all reagents, was run to check for interference and cross-contamination. Replicate measurements were performed. Limits of detection (LOD) were the following: As and Hg, 0.05 μg/g; and Cd and Pb, 0.02 μg/g. They are expressed in sample fresh weight and were limited by the blank value. Recovery rates were 84−91% for As, 86−107% for Hg, 85−93% for Cd, and 74−105% for Pb.

Dietary Exposure Estimates. Dietary intake of As, Cd, Hg, and Pb through fish and seafood consumption was calculated by multiplying the respective concentration in each marine species by the weight of that species consumed by an average individual from Catalonia (*14*). For calculations, when the level of an element was under the LOD, the concentration was assumed to be half of the respective detection limit (ND = $^1/_2$-LOD).

For health risk assessment, the provisional tolerable weekly intakes (PTWI) of As, Cd, Hg, and Pb were compared with the intake of these elements through the consumption of fish and seafood by the population of Catalonia. For this purpose, daily intake was estimated for four age groups: children (4−9 years old), adolescents (10−19 years old), adults (20−65 years old), and seniors (66−80 years old). In turn, each group was subdivided according to sex.

RESULTS

The mean concentrations of As, Cd, Hg, and Pb in the 14 species of fish and shellfish analyzed in this study are depicted in **Figures 1**−**4**. For each species, three composite samples were analyzed. The ranges of the respective concentrations are summarized in **Table 1**. The highest mean As concentration was found in red mullet (16.6 μg/g of fresh weight), followed by shrimp and sole (6.3 and 6.1 μg/g, respectively). In contrast, tuna and salmon were the species showing the lowest As levels. Mean As concentrations in sardine and mussel were similar to those found in our recent survey, whereas the current As concentration in hake was approximately twice that previously

Table 1. Arsenic, Cadmium, Mercury, and Lead Concentrations in Various Edible Marine Species Purchased in Catalonia, Spain[a]

	concn (μg/g of fresh wt)			
species	As	Cd	Hg	Pb
sardine	3.53–3.94	0.002–0.01	0.07–0.09	0.01–0.08
tuna	0.99–1.25	0.01–0.02	0.38–0.58	0.01–0.02
anchovy	3.93–5.42	0.001–0.02	0.08–0.09	0.01–0.02
mackerel	1.73–7.47	0.003–0.01	0.06–0.15	0.01–0.02
swordfish	1.78–2.44	0.02–0.10	1.59–2.22	0.01–0.02
salmon	1.60–2.37	0.01–0.01	0.04–0.05	0.01–0.25
hake	3.22–4.55	0.005–0.01	0.12.0.29	0.01–0.13
red mullet	15.39–17.77	0.001–0.01	0.14–0.36	0.002–0.07
sole	4.55–8.40	0.0004–0.01	0.04–0.13	0.01–0.08
cuttlefish	2.45–5.33	0.01–0.09	0.04–0.08	0.01–0.10
squid	1.41–4.74	0.05–0.15	0.02–0.03	0.01–0.01
clam	1.94–2.69	0.03–0.22	0.01–0.02	0.01–0.11
mussel	2.02–2.44	0.02–0.20	0.02–0.02	0.09–0.21
shrimp	3.85–8.76	0.01–0.03	0.02–0.19	0.01–0.01

[a] For each species and metal, three composite samples were analyzed.

Table 2. Intake of Arsenic, Cadmium, Mercury, and Lead through Fish and Seafood Consumption by Children (Boys) in Catalonia, Spain

species	consumption (g/day)	intake (μg/day) As	Cd	Hg	Pb
sardine	N[a]	N	N	N	N
tuna	7.2	8.1	0.07	3.5	0.10
anchovy	0.10	0.43	N	0.01	N
mackerel	N	N	N	N	N
swordfish	N	N	N	N	N
salmon	1.2	2.2	0.01	0.05	0.12
hake	18.1	74.1	0.17	3.4	0.98
red mullet	N	N	N	N	N
sole	5.2	31.9	0.04	0.43	0.18
cuttlefish	0.3	0.9	0.02	0.01	N
squid	1.9	8.1	0.10	0.11	0.10
clam	0.1	0.2	0.01	N	0.01
mussel	0.3	0.7	0.04	0.01	0.05
shrimp	N	N	N	N	N
total	**34.3**	**126.5**	**0.48**	**7.45**	**1.55**

[a] Negligible values.

Table 3. Intake of Arsenic, Cadmium, Mercury, and Lead through Fish and Seafood Consumption by Children (Girls) in Catalonia, Spain

species	consumption (g/day)	intake (μg/day) As	Cd	Hg	Pb
sardine	1.9	6.9	0.01	0.2	0.07
tuna	4.2	4.7	0.04	2.0	0.06
anchovy	N[a]	N	N	N	N
mackerel	N	N	N	N	N
swordfish	N	N	N	N	N
salmon	N	N	N	N	N
hake	16.2	67	0.16	3.0	0.88
red mullet	N	N	N	N	N
sole	2.4	14	0.02	0.2	0.08
cuttlefish	3.1	9.3	0.22	0.08	0.04
squid	0.6	2.5	0.03	0.03	0.03
clam	0.3	0.6	0.04	N	0.02
mussel	0.6	1.3	0.07	0.01	0.09
shrimp	0.7	4.5	0.01	0.08	0.01
total	**29.9**	**110.6**	**0.61**	**5.60**	**1.27**

[a] Negligible values.

observed (*1*). The highest Cd concentrations were found in clam and mussel, 0.14 and 0.13 μg/g of fresh weight, respectively. The mean Cd concentration in mussels was quite similar to the mean found in our previous survey, whereas there was an increase for sardine and hake. For total Hg (inorganic and organic), the highest concentration corresponded to swordfish. Total Hg concentration in mussel did not change in relation to our previous survey (*1*), whereas the levels of total Hg in sardine and hake increased. Finally, the highest Pb concentrations were found in mussel and salmon, 0.15 and 0.10 μg/g of fresh weight, respectively. Whereas Pb concentration in mussel did not change between the previous (*1*) and the current survey, the levels of Pb in sardine and hake were remarkably increased. However, according to the small numbers of composite samples analyzed in both surveys, the differences in metal concentrations can be considered to be merely informative rather than statistically significant.

Tables 2−**9** summarize current data concerning fish and seafood consumption, as well as mean daily intakes of As, Cd, Hg, and Pb by boys (**Table 2**) and girls (**Table 3**), male (**Table 4**) and female (**Table 5**) adolescents, male (**Table 6**) and female (**Table 7**) adults, and male (**Table 8**) and female (**Table 9**) seniors living in Catalonia. The highest As intake corresponded to the group of male seniors, 217.7 μg/day, whereas the lowest intake of this element corresponded to girls, 110.6 μg/day. For all age/sex groups, the main contributor to As intake was hake, which represented between 23.6 and 60.1% for male adolescents

Table 4. Intake of Arsenic, Cadmium, Mercury, and Lead through Fish and Seafood Consumption by Male Adolescents in Catalonia, Spain

species	consumption (g/day)	intake (μg/day) As	Cd	Hg	Pb
sardine	1.0	3.8	0.01	0.09	0.04
tuna	7.6	8.7	0.07	3.7	0.12
anchovy	2.3	10.6	0.02	0.20	0.03
mackerel	0.4	1.5	N[a]	0.03	0.01
swordfish	N	0.08	N	0.08	N
salmon	3.3	6.3	0.04	0.15	0.34
hake	8.4	34	0.08	1.6	0.46
red mullet	0.2	3.7	N	0.05	0.01
sole	6.2	38	0.05	0.51	0.21
cuttlefish	2.4	7.2	0.17	0.06	0.03
squid	1.9	8.0	0.10	0.11	0.09
clam	0.2	0.40	0.03	N	0.01
mussel	1.3	2.8	0.17	0.02	0.19
shrimp	3.2	21	0.07	0.38	0.04
total	**38.5**	**145.7**	**0.81**	**6.94**	**1.57**

[a] Negligible values.

Table 5. Intake of Arsenic, Cadmium, Mercury, and Lead through Fish and Seafood Consumption by Female Adolescents in Catalonia, Spain

species	consumption (g/day)	intake (μg/day) As	Cd	Hg	Pb
sardine	2.2	7.9	0.02	0.18	0.08
tuna	10.8	12	0.11	5.24	0.17
anchovy	1.0	4.8	0.01	0.09	0.01
mackerel	0.3	1.3	N[a]	0.03	N
swordfish	N	0.08	N	0.08	N
salmon	1.0	1.9	0.01	0.05	0.10
hake	11.2	46	0.11	2.08	0.61
red mullet	0.2	3.6	N	0.05	0.01
sole	3.7	23	0.03	0.30	0.13
cuttlefish	1.0	3.1	0.07	0.03	0.01
squid	5.2	22	0.28	0.29	0.26
clam	N	0.09	0.01	N	N
mussel	N	N	N	N	N
shrimp	3.0	19	0.06	0.35	0.04
total	**39.7**	**144.4**	**0.71**	**8.76**	**1.42**

[a] Negligible values.

Table 6. Intake of Arsenic, Cadmium, Mercury, and Lead through Fish and Seafood Consumption by Male Adults in Catalonia, Spain

species	consumption (g/day)	intake (μg/day) As	Cd	Hg	Pb
sardine	3.8	14	0.03	0.31	0.13
tuna	10.1	12	0.10	4.91	0.16
anchovy	2.0	9.5	0.02	0.17	0.03
mackerel	1.1	4.7	0.01	0.11	0.02
swordfish	0.1	0.12	N[a]	0.11	N
salmon	1.8	3.4	0.02	0.08	0.18
hake	15.8	65	0.15	2.94	0.86
red mullet	0.3	5.5	N	0.08	0.01
sole	5.5	33.4	0.05	0.45	0.19
cuttlefish	4.5	13.3	0.31	0.11	0.05
squid	3.2	13.5	0.17	0.18	0.16
clam	0.3	0.60	0.04	N	0.02
mussel	1.0	2.2	0.13	0.02	0.15
shrimp	3.5	22	0.07	0.42	0.04
total	**52.9**	**198.5**	**1.10**	**9.89**	**2.00**

[a] Negligible values.

Table 7. Intake of Arsenic, Cadmium, Mercury, and Lead through Fish and Seafood Consumption by Female Adults in Catalonia, Spain

species	consumption (g/day)	intake (μg/day) As	Cd	Hg	Pb
sardine	3.0	11	0.02	0.25	0.11
tuna	8.5	9.6	0.08	4.11	0.13
anchovy	1.9	8.8	0.02	0.16	0.03
mackerel	1.3	5.3	0.01	0.12	0.02
swordfish	0.1	0.12	N[a]	0.11	N
salmon	3.0	5.7	0.04	0.14	0.31
hake	15	60	0.14	2.73	0.80
red mullet	0.3	5.3	N	0.07	0.01
sole	5.6	34	0.05	0.46	0.19
cuttlefish	2.8	8.2	0.19	0.07	0.03
squid	3.2	14	0.17	0.18	0.16
clam	0.6	1.4	0.09	0.01	0.04
mussel	1.8	41	0.24	0.04	0.28
shrimp	3.8	24	0.08	0.45	0.05
total	**50.5**	**191.5**	**1.14**	**8.90**	**2.15**

[a] Negligible values.

Table 8. Intake of Arsenic, Cadmium, Mercury, and Lead through Fish and Seafood Consumption by Male Seniors in Catalonia, Spain

species	consumption (g/day)	intake (μg/day) As	Cd	Hg	Pb
sardine	3.5	12.9	0.02	0.29	0.12
tuna	4.9	5.6	0.05	2.38	0.08
anchovy	3.4	15.9	0.03	0.28	0.05
mackerel	0.5	2.1	N[a]	0.05	0.01
swordfish	0.1	0.13	N	0.12	N
salmon	2.2	4.2	0.03	0.10	0.23
hake	23	96	0.22	4.35	1.27
red mullet	0.4	5.9	N	0.08	0.01
sole	3.7	22	0.03	0.30	0.12
cuttlefish	6.0	18	0.41	0.14	0.07
squid	3.2	13.6	0.17	0.18	0.16
clam	0.2	0.45	0.03	N	0.01
mussel	2.1	4.6	0.27	0.04	0.31
shrimp	2.7	17	0.06	0.32	0.03
total	**56.0**	**217.7**	**1.34**	**8.64**	**2.48**

[a] Negligible values.

Table 9. Intake of Arsenic, Cadmium, Mercury, and Lead through Fish and Seafood Consumption by Female Seniors in Catalonia, Spain

species	consumption (g/day)	intake (μg/day) As	Cd	Hg	Pb
sardine	5.3	19.6	0.04	0.44	0.19
tuna	3.2	3.6	0.03	1.53	0.05
anchovy	1.2	5.6	0.01	0.10	0.02
mackerel	2.9	12	0.02	0.27	0.04
swordfish	N[a]	0.10	N	0.09	N
salmon	1.1	2.2	0.01	0.05	0.12
hake	14.6	60	0.14	2.72	0.79
red mullet	0.3	4.5	N	0.06	0.01
sole	5.2	32	0.04	0.42	0.18
cuttlefish	1.9	5.6	0.13	0.05	0.02
squid	0.8	3.3	0.04	0.04	0.04
clam	0.2	0.54	0.03	N	0.01
mussel	0.7	1.5	0.09	0.01	0.10
shrimp	1.7	10.6	0.03	0.20	0.02
total	**39.0**	**160.1**	**0.63**	**5.99**	**1.59**

[a] Negligible values.

and girls, respectively, of the total As intake through fish and seafood consumption. In turn, the contribution of sole to As intake was also notable.

For Cd, male seniors comprised the group with the highest intake, 1.34 μg/day, whereas the lowest intake corresponded to boys (0.48 μg/day). For all age/sex groups, cuttlefish was the main contributor to total Cd intake, with percentages of contribution ranging between 4.2% in boys and 35.7% in girls. In turn, male adults comprised the group showing the highest Hg intake (9.89 μg/day), whereas girls and female seniors were the groups with the lowest Hg intake through fish consumption, 5.60 and 5.99 μg/day, respectively. The main contributor to these intakes corresponded in all groups to tuna (percentages of contribution between 25.6% for female seniors and 59.8% for female adolescents). Hake was also another important contributor to Hg intake (ranging from 22.6% for male adolescents to 54.2% for girls). Finally, male seniors comprised the group with the highest Pb intake through fish and seafood consumption, 2.48 μg/day, whereas girls showed the lowest Pb intake, 1.27 μg/day. For all age/sex groups, hake was the species with the highest contribution to Pb intake, with percentages between 29.0% for male adolescents and 69.4% for girls.

DISCUSSION

In recent years, a notable number of surveys carried out in different countries have determined the concentrations of metals in various edible marine species and estimated human exposure by their consumption. However, comparison among studies is not always easy, as the dietary habits depend on each specific region or country. Moreover, fish and seafood species in the different surveys are not generally the same. Bearing this in mind, we have compared the current results with those of recent studies in which some species analyzed in the present study were also included.

Current As concentrations in mackerel and sole were higher than those reported by Amodio-Cocchieri et al. (*15*), Bordajandi et al. (*11*), and Larsen et al. (*16*), whereas hake, cuttlefish, and shrimp showed lower As concentrations than those found by Bordajandi et al. (*11*). Juresa and Blanusa (*17*) determined concentrations of total Hg, total As, Pb, and Cd in edible fresh fish and shellfish from various areas of the Adriatic Sea. The highest and lowest As concentrations were found in hake (0.37 μg/g of fresh weight) and mackerel (1.06 μg/g of fresh weight), respectively. The levels in hake were higher than those found in the present study, being lower than those detected in mackerel. Recently, Storelli et al. (*18*) reported As concentrations of 3.68 and 2.61 μg/g of fresh weight in muscle tissues of swordfish and tuna from the Mediterranean Sea.

Delgado-Andrade et al. (*19*) measured total As concentration in different foods from southeastern Spain and estimated daily As intake by the general population. These authors reported the following mean concentrations in fish and seafood: 2.72, 1.06, and 61.0 μg/g of fresh weight in fish, shellfish and molluscs, and cephalopods, respectively. The As intake through consumption of these species was 132.9 μg/day, which is into the range of the present study (110.6−217.7 μg/day). Other Spanish studies found higher As intakes: 291 μg/day in healthy adults of Valencia (*20*) and 440 μg/day for a standard adult (70 kg body weight) living in Tarragona (*12*).

Cadmium concentrations in most edible marine species were, in general terms, of the same order of magnitude as those found in the recent literature. However, the level of Cd in hake (the species showing the lowest Cd concentration) reported by Juresa and Blanusa (*17*), 0.002 μg/g of fresh weight, was lower than that of the present study, whereas the Cd concentration found in mussel (the species showing the highest Cd level), 0.14 μg/g of fresh weight (*17*), was similar. On the other hand, Licata et

al. (*21*) found a Cd concentration of 0.06 μg/g of fresh weight in the muscle of mullet of the Strait of Messina (Sicily, Italy), which was higher than the current one. In a recent study, Storelli et al. (*18*) reported Cd concentrations of 0.005 and 0.02 μg/g of fresh weight in muscle tissues of swordfish and tuna from the Mediterranean Sea. It is important to note that for the objectives of the present study, fish and seafood species were randomly purchased, without knowing their origin. Recently, we found a Cd intake from fish and seafood consumption by a 70 kg adult man of Tarragona (Spain) of 4.6 μg/day (*12*), which is considerably higher than the intake of the present study of 1.10 μg/day.

The current Hg concentration in mussels was notably lower than that found by Amodio-Cocchieri et al. (*15*), who reported a level in mussels of the Gulf of Naples, Italy, of 0.02 μg of Hg/g of fresh weight. Storelli et al. (*22, 23*) determined the levels of total Hg and methylmercury in muscle tissues of various fish species of the Mediterranean Sea. In tuna, the average total Hg concentration was 1.8 μg/g of fresh weight, whereas in red mullet it was 0.70 μg/g of fresh weight, levels higher than those found in the current study. These investigators noted that Hg was present in these fish species almost completely in the methylated form. Juresa and Blanusa (*17*) found the highest and lowest Hg concentrations in hake (0.37 μg/g of fresh weight) and mackerel (0.15 μg/g of fresh weight), respectively. Recently, Dabeka et al. (*24*) measured total Hg in edible portions of fish and shellfish purchased in Canada at the retail level. By species, average Hg concentrations ranged from 0.01 μg/g for oysters to 1.82 μg/g for swordfish. This last value is similar to that found in the present study. In another survey of the same research group (*24*), the mean total Hg concentrations of 1.08 and 0.66 μg/g were found in swordfish and fresh frozen tuna, values that are lower and higher, respectively, than the current ones. In a recent study, Storelli et al. (*25*) measured total Hg and methylmercury concentrations in the muscle tissue of striped mullet and hake from the Ionan and Adriatic Seas. Total Hg levels were 0.40−0.49 μg/g of fresh weight for mullet and 0.09−0.18 μg/g of fresh weight for hake. The Hg concentrations in mullet and hake were higher than and lower than/similar to, respectively, those of the current survey. Storelli et al. (*18*) reported Hg concentrations of 0.07 and 0.20 μg/g of fresh weight in muscle tissues of swordfish and tuna from the Mediterranean Sea.

Recently, we found a total Hg intake through fish and shellfish consumption by an adult man of Tarragona (Spain) of 5.4 μg/day (*12*). This value is slightly lower than the current intake, 6.94 μg/day. In a previous Spanish study performed in the Basque Country to estimate Hg intake associated with fish consumption in an EPIC (European Prospective Investigation into Cancer) cohort (*26*), it was found that 99.9% of the individuals had intakes below 75% of the PTWI for Hg established by the FAO/WHO (*27*). However, it was also noted that the limit dosage recommended by the U.S. EPA (oral reference dose for methylmercury, 0.1−0.3 μg/kg/day) was often exceeded. In that study, among more than 20 analyzed species, the highest Hg concentrations were found in tuna and Atlantic bonito (0.31 μg/g of fresh weight), whereas the lowest levels corresponded to mussel (0.011 μg/g of fresh weight) and squid, cuttlefish, and shrimp (0.015 μg/g of fresh weight) (*26*).

With respect to Pb, in comparison with the concentrations found in the present study, Muñoz et al. (*10*) reported a higher Pb concentration in sardine, whereas Juresa and Blanusa (*17*) reported a lower Pb concentration in hake (0.007 μg/g of fresh weight) and a similar concentration in mussel (0.15 μg/g of fresh weight), species in which these authors found the lowest and highest Pb concentrations, respectively. A Pb level similar to that of the present study was also reported by Licata et al. (*21*) in the muscle of mullet (0.03 μg/g of fresh weight). Recently, Storelli et al. (*18*) reported Pb concentrations of 0.05 and 0.10 μg/g of fresh weight in muscle tissues of swordfish and tuna from the Mediterranean Sea. In relation to human exposure to Pb through fish and seafood consumption, Bocio et al. (*12*) estimated a value of 4.2 μg/day for an adult man of Tarragona (Spain), which is twice that of the present study.

For health risk assessment, it must be noted that the European Union law (European Commission Decision 78/2005) on Hg concentrations in edible marine species sets a limit of 1 μg/g of fresh weight for tuna, swordfish, and shark and 0.5 μg/g of fresh weight for all other species. In this survey, only the levels of total Hg found in swordfish exceeded those limits. On the other hand, the current intakes concerning As, Cd, Hg, and Pb were compared with the respective PTWIs established the FAO/WHO (*27*). For inorganic As, the PTWI is 15 μg/kg of body weight per week, or 129 μg/day for a subject of 60 kg. As indicated above, in the present study all analyses were carried out for total (inorganic and organic) arsenic. However, it is well-known that most As found in fish and seafood is in the form of organic As (*10*, *28*), which is the less toxic form. In the literature, the percentage of inorganic As in fish and seafood has been reported to be between 0.02 and 11% (*10*), whereas the maximum acceptable daily load for As, set by the WHO in 1967 and unrevised in 1989, is 3000 μg for a subject of 60 kg (*29*). Taking this into account, in the current study As intake would not be of concern for any age/sex group. This intake, 110.6−217.7 μg/day, is quite similar to that found in our previous survey, 113.8−203.3 μg/day (*1*). However, it is notably higher than that recently reported for Chile, which was 44.7 μg/day (*30*). Carcinogenic risk assessment of As was done according to the slope factor established as 1.5 $(mg/kg/day)^{-1}$ (*31*). Accordingly, the carcinogenic risk for a male adult during 70 years of life would be 1.8×10^{-4}. Generally, a level of 10^{-6} or lower is considered to be safe for cancer risk in adults (*31*).

The Cd PTWI is 7 μg/kg per week. The current Cd intake represented approximately 2% of the PTWI in all age/sex groups. Cadmium intake, 0.48−1.34 μg/day through fish and seafood, is lower than the intake found in our recent survey in Catalonia, 1.86−3.33 μg/day (*1*), and in Chile, 9.2 μg/day (*30*). It is comparable to that found in Greece, 0.7−1.1 μg/day (*32*). Recently, Storelli et al. (*33*) reported weekly intakes of 0.09−0.49 and 0.05−0.24 μg/kg of body weight for Cd and Hg, respectively, from the consumption of cephalopod molluscs in Italy. This would mean contributions to the corresponding PTWIs of 1.3−7.0% for Cd and 1.0−4.8% for Hg. In the current study, all Hg intakes were under the established PTWI, 5 μg/kg/week. Boys showed the maximum Hg intake at 2.2 μg/kg/week. As for As, this amount corresponds to total (inorganic and organic) Hg. Assuming that in fish and seafood most Hg is in the methylmercury form (*18*, *22*, *23*, *25*), the intakes for most age/groups would be under the limit of safety, 1.6 μg/kg of body weight/week of methylmercury (*34*). The exception corresponds to the boys' group, 1.96 μg/kg/week, an intake that surpasses the recommended limit for methylmercury. The current intake of Hg (5.6−9.9 μg/day) through fish and seafood consumption in Catalonia is very similar to that of our previous survey, 5.0−8.9 μg/day (*1*). This intake is again notably higher than that recently reported for Chile, 1.6 μg/day (*30*). With respect to Pb, for all age/sex groups the estimated intake was

lower than the PTWI value, 25 μg/kg per week. The current Pb intakes (1.27−2.48 μg/day) are lower than those found in our recent survey (2.64−4.71 μg/day) (*1*) and comparable to that of Chile, 1.7 μg/day (*30*). However, in the Canary Islands (Spain), with an estimated fish and seafood consumption not very different from that of Catalonia, Rubio et al. (*9*) reported a remarkably higher Pb intake through marine species, 13.1−20.9 μg/day. These authors found that fish consumption even being moderate in the Canary Islands, this food group was the main contributor to Pb intake because of its high mean Pb concentration (367 μg/kg). In our study, none of the species analyzed reached that level.

In summary, comparison of the current concentrations with those found in the previously analyzed fresh species (sardine, hake, and mussel) (*1*) showed an increase in metal concentrations, especially for Hg and Pb in sardine and hake. For a standard adult man, hake represents about 30% of the total fish and seafood consumption in Catalonia. Therefore, this indicates a remarkable contribution of this species to the intake of these metals. For Hg, consumption of blue fish means a contribution of 58% of the total, whereas for Pb white fish represents 53% of the total intake. It is important to remark that Hg levels in swordfish exceeded the European Union requirements. However, the consumption of this fish species by the population of Catalonia is very low and, therefore, its contribution to daily intake of Hg is, in fact, irrelevant. Because of geographic location and sociocultural customs, in Catalonia (Mediterranean diet) fish and seafood consumption is notable. Consequently, it would be recommendable that monitoring studies are periodically performed to assess the temporal trends in human exposure to these toxic elements through fish and seafood consumption.

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