

Mercury Content in Tinned Molluscs (Mussel, Cockle, Variegated Scallop, and Razor Shell) Normally Consumed in Spain, 2005

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ABSTRACT

Concentrations of mercury were determined for tinned molluscs (Mollusca, Bivalvia), i.e., mussels (*Mytilus* spp.), cockles (*Cerastoderma edule*), variegated scallops (*Chlamys varia*), and razor shells (*Ensis* spp.), consumed in Spain. A total of 220 samples were analyzed: 120 mussels, 40 cockles, 24 variegated scallops, and 36 razor shells. Samples were obtained weekly from markets in Santa Cruz de Tenerife (Canary Islands) over a period of 12 months. All observed concentrations of mercury were below the maximum permitted for human consumption (0.5 mg/kg) as defined by European Community Decision 93/351/CE. Mercury concentrations were 27.28 ± 12.43 $\mu\text{g}/\text{kg}$ for mussels, 66.59 ± 23.53 $\mu\text{g}/\text{kg}$ for cockles, 33.68 ± 15.76 $\mu\text{g}/\text{kg}$ for variegated scallops, and 21.26 ± 12.24 $\mu\text{g}/\text{kg}$ for razor shells. To evaluate the importance of mercury as a food contaminant, the percentage ingested daily, the weight of mercury in the diet, and the calculated percentage of this metal in the diet contributed by reference species were estimated.

Microfiltering bivalve molluscs tend to accumulate high concentrations of metals in relation to their size and weight (2, 15). Because of their capacity to concentrate contaminants by several orders of magnitude over the corresponding concentration in sea water, these molluscs are used as bioindicators of marine pollution. They are also considered edible shellfish, and both wild and aquaculture products contribute to the traditional diet of the Spanish population. These shellfish are consumed in their natural form and as various commercial products, the most popular of which is the tinned form. Therefore, this product is of great interest to health control agencies.

No natural or aquaculture populations of these species exist in the Canary Islands (eastern central Atlantic Ocean; 27 to 30°N, 13 to 19°W), but their importation and demand are significant, and they are the most common marine foods in the island diet. The products chosen for study were tinned molluscs of Galician origin and tinned razor shells from Chile.

Heavy metals are absorbed by aquatic organisms as toxic substances in water and sediments or in the food chain (34). Mercury is well known for its neurotoxicity, but it also affects the gastrointestinal tract and renal functions (16, 31). Mercury is an important contaminant in many types of seafood (fish and shellfish) (20, 29).

Bivalve molluscs do not normally contain high concentrations of mercury because they are low on the food chain and have a short life cycle. The LD₅₀ (dose lethal to 50% of specimens) for mercury is 10.4 $\mu\text{g}/\text{liter}$ for the oyster *Crassostrea gigas* and 10.1 $\mu\text{g}/\text{liter}$ for *Mytilus* spp. (17). Mercury causes increased consumption of oxygen in *Crassostrea virginica* larvae at 5 days of life after 24 h of exposure to a concentration of 1 to 100 $\mu\text{g}/\text{liter}$ (6). Mercury also can act as a genotoxin, producing changes in sister chromatids in fertilized bivalve eggs after exposure to HgCl₂ at 30 $\mu\text{g}/\text{liter}$ (3).

MATERIALS AND METHODS

Samples were collected for a 12-month period at a large commercial center in Santa Cruz de Tenerife on a weekly basis (five

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TABLE 1. Recovery of mercury in reference material according to the national measurement system valid analytical measurement program (BCR, European Commission)

Metal	Material	No. of samples	Recovered (mg kg ⁻¹) ^a		Procedure
			Sample	Certified	
Hg	BCR-278 R mussel tissue	11	0.196 ± 0.009	0.195 ± 0.010	Cold-vapor atomic absorption spectrometry

^a Mean \pm standard deviation.

TABLE 2. Composition of various mollusc species

Species	Sample weight (g)	Dry weight (g) ^a	Ash weight (g) ^a
Mussel	5	3.24 ± 0.24	0.19 ± 0.07
Cockle	5	2.24 ± 0.14	0.33 ± 0.08
Variogated scallop	5	3.05 ± 0.25	0.21 ± 0.09
Razor shell	5	2.67 ± 0.18	0.21 ± 0.09

^a Mean ± standard deviation.

tins per week). The samples originated from Galician conserve industries (northwestern Spain) or Chile.

Because of the low concentrations of this metal per individual, all specimens in each tin (all of same size and source) were homogenized and pooled to obtain a 10-g sample. Samples were digested with an electric heater (Foss Tecator, Hoeganaes, Sweden) in digestion tubes in which 5 g of homogenized sample was mixed with 10 ml of HNO₃, 0.05 g of V₂O₅ as a reaction catalyst, and 5 ml of antilather (AP 31 Antilather Extram, Merck, Darmstadt, Germany). The sample was incubated for 15 min and then left to cool.

After addition of 15 ml of cold H₂SO₄, samples were reheated for 15 min until digestion was complete and then were cooled at room temperature. Digested samples were mixed 100 ml of double-distilled water and analyzed with an atomic absorption spectrophotometer (4100 ZL Zeeman, Perkin Elmer, Foster City, Calif.) with a hydride generator, FIAS 400 injection system, and automatic sampler (AS 90, Perkin Elmer). Table 1 gives an example of mercury recovery from reference material to demonstrate the suitability of the methodology.

The statistical analysis of the results consisted of a Kolmogorov-Smirnov test for normality (33) and a test for homogeneity of variance using the Levene statistic (25). When the data were not normally distributed, the nonparametric Kruskal-Wallis and Mann-Whitney U tests (4) were used.

RESULTS AND DISCUSSION

Study of general composition. Data on dry weight and wet weight of the bivalve samples are presented in Table 2. There were no significant differences in composition of the different species. Cockles had the highest moisture percentage (76 ± 1.4%) and ash weight (0.33 ± 0.08 g), and mussels had the highest dry weight (3.24 ± 0.24 g).

Mercury content analysis. Data on the mercury content of the different species are presented in Table 3. Mercury concentrations for all analyzed species were substantially below the maximum stipulated for molluscs in the current regulations of the European Community (500 µg

TABLE 4. Comparative mercury concentrations in molluscs within the European Community

Country	Species	Mercury concn (µg kg ⁻¹)	Source
France	Molluscs	24	11
Germany	Molluscs	29	11
Greece	Molluscs	23.5	11
Ireland	Molluscs	20	11
Portugal	Molluscs	79.5	11
United Kingdom	Molluscs	43	11
Spain	Mussel	27.83	This study
	Variogated scallop	33.68	This study
	Cockle	66.59	This study

kg⁻¹, Decision 93/351/EC) (9). Statistical analysis indicated that the cockles contained the highest concentration of mercury (66.59 ± 23.53 µg kg⁻¹). This finding was unexpected because this species was the smallest of those studied (between 13 and 20 mm).

About 80% of dietary mercury is of seafood origin (22, 27). A comparison of the results obtained in this study with those of the European Union Experts Committee, Directorate—General Health and Consumer Protection (11) revealed that the mercury concentrations in these seafood samples are within the European Community guidelines (Table 4).

Previous study and data from different geographical areas indicate substantial variability in concentrations of mercury (between 7 and 900 µg kg) (10). In *Crassostrea angulata*, low mercury concentrations were found in the Spanish Atlantic area: 190 µg kg⁻¹ on the Asturian coast, 160 µg kg⁻¹ on the Pontevedra coast, and 170 µg kg⁻¹ on the Cadiz coasts (30). López Artíguez et al. (21) found lower mercury concentrations for the same species in the coastal areas of Huelva (90 µg kg⁻¹). For *Mytilus edulis*, concentrations of 110 to 250 µg kg⁻¹ were found in wild mussels in the Bayona and Vigo areas (Galicia), respectively (8). In the Pontevedra Sea lock, concentrations in aquaculture mussels were 90 to 280 µg kg⁻¹ and those in wild mussels were 80 to 180 µg kg⁻¹, and in the Arosa Sea lock, concentrations in wild mussels were 60 to 110 µg kg⁻¹ (8). In the Sudbury River Reserve (Canada), 100 µg kg⁻¹ mercury and 41 µg kg⁻¹ methylmercury were found (28). In the second reserve on this river, 59 µg kg⁻¹ total mercury and 54 µg kg⁻¹ methylmercury were found (1).

TABLE 3. Statistical analysis of mercury content in molluscs

Species	n	Mean ± SD ^a	Mercury concn (µg kg ⁻¹)		
			Maximum	Minimum	Variance
Mussel	120	27.83 ± 12.43	11.4	63.60	154.66
Cockle	40	66.59 ± 23.53	25.60	102.60	553.81
Variogated scallop	24	33.68 ± 15.76	8.60	44.40	248.369
Razor shell	36	21.26 ± 12.24	1.60	57.80	149.772

^a SD, standard deviation.

TABLE 5. Mercury concentrations in different species of molluscs in different countries

Species	Place	Mercury concn ($\mu\text{g kg}^{-1}$)	Reference
Cockles			
<i>Anadara selenis</i>	Ghana	190	Otchere et al. (24)
<i>A. tuberculosa</i>	Costa Rica	160	De la Cruz (7)
<i>A. granosa</i>	Malaysian	30–60	Jothy et al. (19)
<i>A. selenis</i>	Nigeria	180	Joiris et al. (18)
<i>Cerastoderma glaucum</i>	France	550	Szefer et al. (32)
<i>Venerupis galactites</i>	Australia	710	Francesconi and Lenanton (12)
<i>C. edule</i>	Spain	66.59	This study
Oysters			
<i>Crassostrea tulipa</i>	Ghana	170	Otchere et al. (24)
<i>C. virginica</i>	United States	1,600	Gardner et al. (13)
<i>Macoma phenax</i>	United States	170	Gardner et al. (13)
Mussels			
<i>Perna perna</i>	Ghana	260	Otchere et al. (24)
<i>Mytilus galloprovincialis</i>	Adriatic Sea	2,000	Mikac et al. (22)
<i>M. edulis</i>	Belgium	2,000	Gurney (14)
<i>M. edulis</i>	Canada	170–230	Cossa and Rondeau (5)
<i>M. edulis</i>	W. Denmark	750	Riisgard et al. (26)
<i>M. galloprovincialis</i>	Spain	27.83	This study
Clams			
<i>Mercenaria mercenaria</i>	United States	800	Gardner et al (13)
Razor shell			
<i>Ensis ensis</i>	Spain	21.26	This study
Scallop			
<i>Chlamys varia</i>	Spain	331.68	This study

Bivalves are used as indicators of contaminants in the sea because of their filter feeding behavior and because they are constantly exposed to contaminants and pollutants in the water column and sediments (23). In *Mytilus* spp. in Spain, concentrations of 77 to 139 $\mu\text{g kg}^{-1}$ were found, and other authors have found concentrations of 100 $\mu\text{g kg}^{-1}$ (28) and 59 $\mu\text{g kg}^{-1}$ (1). Our results are slightly lower, possibly because of the purification process applied to the molluscs before canning. The places where molluscs are cultured also are strictly controlled, and the time spent in culture is never more than 1 to 2 years. In Table 5 the total mercury levels found by others in various bivalve species from different countries are detailed and compared with our data, indicating the large variation observed.

Table 6 provides information on the amount of mercury ingested for different portions of each bivalve species

studied and the percentage of the admissible daily intake (ADI) according to Decision 93/351/EC (9), which established the ADI at 500 mg kg^{-1} . The contribution of the studied bivalves to the ADI for mercury is minimum and, for normal portions, is never over the ADI. A person would have to ingest 17,966.22 g of mussels, 7,508.63 g of cockles, 14,846.61 g of variegated scallops, and 23,518.30 g of razor shells to reach the ADI.

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TABLE 6. Contribution of various molluscs to admissible daily intake (ADI) of mercury based on different portion sizes

Species	Mercury concn ($\mu\text{g kg}^{-1}$) ^a	50-g portion		100-g portion		200-g portion	
		Mercury (μg)	ADI (%)	Mercury (μg)	ADI (%)	Mercury (μg)	ADI (%)
Mussel	27.83 \pm 12.43	1.39	0.28	2.78	0.56	5.57	1.11
Cockle	66.59 \pm 23.53	3.33	0.66	6.66	1.33	13.3	2.66
Scallop	33.68 \pm 15.76	1.68	0.34	3.37	0.67	6.74	1.35
Razor-shell	21.26 \pm 12.24	1.06	0.21	2.13	0.44	4.25	0.85

^a Mean \pm standard deviation.

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