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Research Note

Content of Toxic and Essential Metals in Canned Mussels Commonly Consumed in Tenerife, Canary Islands, Spain

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ABSTRACT

Concentrations of three toxic heavy metals (Hg, Pb, Cd) and six essential heavy metals (Fe, Zn, Mn, Cr, Cu, Ni) were determined in mussel conserves (*Mytilus galloprovincialis, Bivalvia, Mollusca*) consumed habitually by individuals in Tenerife (Canary Islands, Spain). A total of 600 samples were analyzed, corresponding to six different commercial brands and four different processing types: pickled sauce (mixture of olive oil, vinegar, red pepper, laurel, and salt), coquille St. Jacques sauce (coquille St. Jacques broth), nature (water and salt), and bionature (water, salt, and soluble vegetal fiber). Samples were collected weekly from markets in Santa Cruz de Tenerife during a 12-month period. All values for toxic metals were lower than the permitted maximum for human consumption as proscribed in European Community Directive 2001/22/CE (1,000 μg/kg wet weight for Pb and Cd) and European Community Decision 93/351/EEC (500 μg/kg wet weight for Hg). For the six essential heavy metals, mussels are a very good source, contributing high percentages of the recommended daily allowance.

The tendancy of mussels to accumulate heavy metals is related to their weight *(8)* and can reflect the local concentration of various metals and other toxins *(4)*. Mussels are often used as bioindicators because they have the capacity to concentrate contaminants from seawater by several orders of magnitude.

However, these molluscs are an important component of aquaculture programs (Spain produces nearly 300,000 tons yearly) and make up part of the Spanish traditional diet. They can be used in their natural state or they can be commercialy processed in many ways, the most popular of which is canning. Because of their popularity, mussels are a nutritionally important component of the Spanish diet.

In the Canary Islands (central-east Atlantic Ocean, 27 to 30°N and 13 to 19°W), there are no natural or cultured populations of mussels of the genus *Mytilus* (e.g., *Mytilus edulis* and *Mytilus galloprovincialis*), which are typical to the Iberian Peninsula. However, because demand for these molluscs is high they are imported and are one of the most common marine foods in the island diet. The material chosen for study was canned mussels of Galician origin (northwestern Spain), which are always available in Santa Cruz de Tenerife supply markets.

Heavy metals can accumulate in aquatic organisms from exposure to toxic substances in water and sediments or in the food chain *(38)*. Because consumption of contaminated foodstuffs is the main route of exposure to trace metals for most humans, information about dietary intake is important to assess risks to human health *(21)*. In this study, we analyzed concentrations of toxic metals (Hg, Pb, and Cd) and essential metals (Fe, Zn, Mn, Cr, Cu, and Ni), which can become toxic at high concentrations.

Mercury is well known for its neurotoxicity, but it also affects the gastrointestinal tract and renal function *(31)*. In aquatic environments, inorganic Hg is microbiologically transformed into the lipophilic organic compound methylmercury. This transformation makes Hg more prone to biomagnification in aquatic food chains *(17)*. The presence of metals in marine sediments is a consequence of both natural and anthropogenic contamination, but the presence of toxic metals in some species of fish and their products indicates extreme contamination of the aquatic environment by those metals *(29)*. Consequently, human populations with a traditionally high dietary intake of food originating from either freshwater or marine environments have the highest potential for exposure to methylmercury *(17)*.

Cadmium in the human diet constitutes a potential chronic health hazard. In the nonsmoking general population, diet is the major source of Cd exposure. In 1968, the Japanese Ministry of Health and Welfare admitted that ''Itai-itai'' disease was caused primarily by Cd. The WHO International Programme on Chemical Safety established that important sources of Cd contamination include phosphate fertilizer, sewage sludge, plated or galvanized equipment, enamels, and glazes. Although molluscs and crustaceans accumulate Cd, fishes generally have Cd levels of <0.2 μg/kg *(15)*.

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TABLE 1. *Reference materials for recuperation study*

Metal	Reference material	Mean concentrations (μg/kg wet weight)[a]	Certified values (μg/kg wet weight)	Method[b]
Pb	NBS SRH 1577a bovine liver	135 ± 15	135 ± 15	ET-AAS
Pb	BCR-278 R mussel tissue	2,000 ± 40	1,970 ± 70	ET-AAS
Cd	BCR-278 R mussel tissue	348 ± 7	347 ± 8	FT-AAS
Cd	BCR CRM 184 bovine muscle	12 ± 1	13 ± 2	ET-AAS
Hg	BCR-278 R mussel tissue	196 ± 9	195 ± 10	CV-AAS
Cr	BCR-278 R mussel tissue	780 ± 60	760 ± 40	ICP-AES
Cu	BCR-278 R mussel tissue	9,450 ± 130	9,390 ± 100	ICP-AES
Cu	BCR-442 cod muscle	1,050 ± 70	1,050 ± 80	ICP-AES
Fe	BCR-278 R mussel tissue	5,460 ± 300	5,460 ± 300	ICP-AES
Mn	BCR-278 R mussel tissue	7,690 ± 230	7,690 ± 220	ICP-AES
Zn	BCR-278 R mussel tissue	8,310 ± 1,700	8,280 ± 1,000	ICP-AES
Ni	BCR-414 plankton	18,800 ± 800	18,800 ± 600	ICP-AES

[a] $n = 11$.
[b] ET-AAS, electrothermal atomic absorption spectrophotometry; CV-AAS, cold vapor atomic absorption spectrophotometry; ICP-AES, inductively coupled plasma atomic emission spectrophotometry.

Lead is a nonessential heavy metal that can occur in high concentrations in animals, especially mammals *(32)*. High concentrations of Pb have been linked to human health problems, including nervous system dysfunction in fetuses and infants and hematotoxicity, reproductive dysfunction, and Alzheimer's disease in adults. Even low Pb concentrations can be associated with asthma, anemia, and learning disabilities *(16, 20)*.

Iron and zinc are essential minerals for humans. Nevertheless, high Fe concentrations can produce toxic cardiovascular, neurological, and gastrointestinal effects *(3)*. Zinc concentrations in bivalve molluscs usually exceed the 50% lethal dose during embryogenesis *(33)*. Manganese is the least toxic of the metals analyzed in this study, but it can be toxic to the nervous system, resulting in Parkinsonian signs *(32)*. Chromium compounds are often environmental pollutants and are known for their mutagenic, carcinogenic, and toxic effects *(28)*. Copper concentrations of 63.6 μg/liter reduce the growth of bivalve molluscs and can be fatal at >86.5 μg/liter *(19)*. Aquatic Ni concentrations are usually approximately 6 μg/liter, but higher concentrations have been reported *(14)*.

The main goal of this research was to assess heavy metal contamination in canned mussels normally consumed in Tenerife and to use these data to evaluate nutritional patterns and toxicological risk.

MATERIALS AND METHODS

Samples for study (five cans per week) were collected over the course of 12 months from various stores in Santa Cruz de Tenerife. A total of 600 samples were analyzed, corresponding to six different commercial brands (A through F) and four different processing types: pickled sauce (mixture of olive oil, vinegar, red pepper, laurel, and salt), coquille St. Jacques sauce (coquille St.

TABLE 2. *General composition of mussels by brand*

Brand	*n*	Dry weight (g)	% moisture	Ash weight (g)
A	150	3.25 ± 0.04	67.51 ± 0.34	0.17 ± 0.01
B	90	3.30 ± 0.001	67.03 ± 0.15	0.19 ± 0.01
C	51	3.21 ± 0.04	67.92 ± 0.27	0.19 ± 0.02
D	129	3.16 ± 0.07	67.95 ± 0.17	0.22 ± 0.02
E	93	3.23 ± 0.04	67.68 ± 0.24	0.18 ± 0.02
F	57	3.35 ± 0.03	66.53 ± 0.22	0.21 ± 0.02

TABLE 3. *General composition of mussels by processing type*

Process	*n*	Dry weight (g)	% moisture	Ash weight (g)
Coquille St. Jacques sauce	213	3.28 ± 0.04	66.8 ± 0.40	0.25 ± 0.01
Pickled sauce	240	3.30 ± 0.02	67.02 ± 0.15	0.18 ± 0.007
Nature	75	2.99 ± 0.08	69.80 ± 0.40	0.16 ± 0.02
Bionature	41	3.14 ± 0.05	68.62 ± 0.47	0.22 ± 0.03

TABLE 4. *Heavy metal concentration (μg/kg wet weight) with respect to brand and processing type*

Metal	Brand	Coquille St. Jacques sauce	Pickled sauce	Nature	Bionature	Mean ± SD
Pb	A	4.47 ± 0.68	7.75 ± 0.68	7.73 ± 0.82		6.96 ± 0.48
	B	7.12 ± 0.86	7.87 ± 0.75			7.57 ± 0.25
	C		7.94 ± 1.09			7.94 ± 1.09
	D	5.28 ± 0.84	5.90 ± 0.92	5.58 ± 0.96	8.16 ± 1.10	6.38 ± 0.36
	E	6.21 ± 0.80	7.96 ± 0.65			7.17 ± 0.20
	F		8.09 ± 0.85			8.09 ± 0.85
Cd	A	5.13 ± 0.46	4.49 ± 0.18	4.52 ± 0.32		4.65 ± 0.08
	B	4.74 ± 0.35	4.94 ± 0.44			4.86 ± 0.11
	C		5.71 ± 0.87			5.71 ± 0.87
	D	6.00 ± 0.59	5.55 ± 0.29	4.87 ± 0.53	4.35 ± 0.24	5.01 ± 0.09
	E	4.33 ± 0.36	3.98 ± 0.15			4.14 ± 0.10
	F		4.60 ± 0.34			4.60 ± 0.34
Cu	A	1,380 ± 70	1,870 ± 110	1,880 ± 110		1,760 ± 70
	B	1,170 ± 120	1,560 ± 90			1,400 ± 80
	C		1,580 ± 130			1,580 ± 130
	D	1,120 ± 110	1,790 ± 190	1,490 ± 120	1,580 ± 100	1,460 ± 80
	E	1,600 ± 150	1,790 ± 120			1,710 ± 90
	F		1,550 ± 90			1,550 ± 90
Ni	A	5,110 ± 270	5,570 ± 250	4,920 ± 400		5,220 ± 190
	B	7,920 ± 100	6,360 ± 710			6,980 ± 250
	C		7,760 ± 260			7,760 ± 260
	D	8,300 ± 850	6,780 ± 290	7,660 ± 720	8,870 ± 350	8,030 ± 350
	E	4,580 ± 270	4,950 ± 290			4,780 ± 200
	F		5,830 ± 250			5,830 ± 250
Fe	A	18,310 ± 2,920	42,180 ± 5,960	33,270 ± 2,240		33,240 ± 3,010
	B	32,350 ± 3,070	26,650 ± 1,700			28,330 ± 1,730
	C		44,620 ± 4,700			44,620 ± 4,700
	D	28,260 ± 3,140	31,870 ± 3,800	32,070 ± 4,220	35,520 ± 3,510	31,920 ± 1,880
	E	33,320 ± 5,130	42,550 ± 3,990			38,380 ± 3,310
	F		26,220 ± 2,350			26,220 ± 2,350
Zn	A	51,530 ± 2,530	62,370 ± 3,430	74,620 ± 4,400		64,180 ± 2,510
	B	63,040 ± 2,630	56,680 ± 2,950			59,230 ± 2,150
	C		69,830 ± 2,810			69,830 ± 2,810
	D	60,880 ± 3,080	59,430 ± 3,600	71,060 ± 7,230	58,890 ± 2,730	61,620 ± 2,020
	E	60,550 ± 3,620	53,770 ± 2,040			56,680 ± 2,080
	F		61,190 ± 2,870			61,190 ± 2,870
Cr	A	1,420 ± 100	1,480 ± 70	1,290 ± 80		1,400 ± 50
	B	1,770 ± 80	1,740 ± 80			1,750 ± 40
	C		2,120 ± 100			2,120 ± 100
	D	1,760 ± 100	1,800 ± 100	1,390 ± 140	1,750 ± 90	1,700 ± 60
	E	1,250 ± 90	1,270 ± 60			1,260 ± 50
	F		1,690 ± 100			1,690 ± 100
Mn	A	1,610 ± 180	1,740 ± 70	1,350 ± 110		1,570 ± 80
	B	2,040 ± 180	1,370 ± 70			1,630 ± 110
	C		1,800 ± 190			1,800 ± 190
	D	1,820 ± 180	1,490 ± 170	1,510 ± 160	1,800 ± 150	1,700 ± 90
	E	1,950 ± 160	1,640 ± 100			1,780 ± 100
	F		1,430 ± 470			1,430 ± 470

TABLE 5. *Daily consumption of mussels necessary to exceed the admissible daily intake of Pb, with respect to brands and processing types*

	Amount (g wet weight) by processing type			
Brand	Coquille St. Jacques sauce	Pickled sauce	Nature	Bionature
A	194.933	222.717	221.238	
B	210.970	202.429		
C		175.131		
D	166.666	180.180	205.338	229.885
E	230.946	251.256		
F		217.391		

TABLE 6. *Daily consumption of mussels necessary to exceed the admissible daily intake of Cd, with respect to brands and processing types*

	Amount (g wet weight) by processing type			
Brand	Coquille St. Jacques sauce	Pickled sauce	Nature	Bionature
A	212.765	129.032	129.366	
B	140.449	127.064		
C		125.944		
D	189.393	169.491	179.211	122.549
E	161.030	125.628		
F		123.609		

Jacques broth), nature (water and salt), and bionature (water, salt, and soluble vegetal fiber). Whatever processed products were available at the time were collected. The individual mussels in each can (all the same size and coming from the same place) were homogenized to obtain the required amount of total sample (≈31.5 g) for analysis. Taking into account the peculiarities of Hg compared with the other metals analyzed, two different sample preparation protocols were followed. For Hg analysis, three 0.5-g subsamples (each weighed in Teflon reactors) were taken from the homogeneized portions, 10 ml of concentrated HNO_3 was added, and the mixture was heated in a microwave oven long enough for total digestion to take place (≈20 min). The mixture was allowed to cool to room temperature, and the volume was brought to 50 ml by the addition of double-distilled deionized water.

For the remaining metals, three 10-g subsamples were weighed into porcelain crucibles and heated on a hot plate to 80 to 105°C until a constant dry weight was obtained. The crucibles were then placed in an oven furnace and the temperature was raised gradually (≈50°C/h) until 450 ± 25°C was reached. Approximately 18 to 24 h elapsed from the beginning of incineration until the formation of white ash. Crucibles were then left to cool to room temperature, and ash was disolved with 1 ml of 0.6% HNO_3. Sample volume was brought to 50 ml by the addition of double-distilled deionized water.

For toxic heavy metal analysis, an atomic absorption spectrophotometer (4100 ZL Zeeman, Perkin Elmer, Foster City, Calif.) was used with a graphite 4100 ZL oven (Perkin Elmer) and an automatic AS 70 sampler for Pb and Cd *(29)* and with a hydrures generator, FIAS 400 injection system, and AS 90 automatic sampler (Perkin Elmer) for Hg *(19, 28)*. For essential heavy metal analysis, an atomic emission spectrophotometer (Atomscan 25, Thermo Jarell Ash, Ventura, Calif.) of inductively coupled plasma was used. Table 1 lists the values for the recuperation study carried out using the corresponding reference materials to demonstrate the performance of the method.

Results were analyzed for normality with the Kolmogorov-Smirnov model *(36)* and for homogeneity of variance with a Levene test *(27)*. When data were not normally distributed, the nonparametric Kruskal-Wallis test and Mann-Whitney U test *(11)* were used. Normally distributed data were analyzed using an analysis of variance and post hoc tests *(24)*.

RESULTS AND DISCUSSION

Results regarding the general composition of the mussels are presented in Tables 2 and 3, and results from the heavy metal analyses are presented in Table 4. Dry weight, moisture percentage, and ash weight obtained were not significantly different among the different brands and processing types, mainly because the different brands market the same type of mussel in cans. The moisture percentage in the samples was less than that obtained by Moreiras et al. *(22)*. In their study, 85.4% moisture was obtained, which is 20% higher than that obtained in the present study, but this higher moisture content could be due to the comparison between fresh and canned food; despite being immersed in liquid, canned mussels have been boiled and thus have lost part of their water content.

The Pb concentration found in samples was always much less than the maximum allowed by European Community Directive 2001/22/CE *(13)* (1,000 μg/kg wet weight; all concentrations reported here are on a wet weight basis unless otherwise indicated). The highest Pb concentrations were found in brand F mussels (8.09 ± 0.85 μg/kg) and in mussels processed with pickled sauce (8.16 ± 1.10 μg/kg) (Fig. 1). Other authors have found higher concentrations of Pb in other bivalves; for oysters, 1,250 to 14,000 μg/kg was found in areas of urban contamination *(2)* and 440 to 640 μg/kg *(7)* was found in zones considered normal.

The Cd concentration was also always much less than that permitted by Directive 2001/22/CE (1,000 μg/kg wet weight). The highest concentrations were found in brand C (5.71 ± 0.87 μg/kg) and in mussels processed with coquille St. Jacques sauce (6.0 ± 0.59 μg/kg) (Fig. 2). Several authors have studied Cd content in molluscs, obtaining concentrations in *Mytilus* spp. of 1,780 μg/kg in the Looe estuary (Cornwall, UK) and concentrations in *M. galloprovincialis* of 4,000 ± 1,500 μg/kg in Cape Ghir and 800 ±

TABLE 7. *Nutritional importance of 150 g of canned mussels (approximately one can) in reference to heavy metals with recommended daily allowance (RDA)*

Metal	RDA (mg/day)	Metal concentration (mg/150 g mussels)	% RDA
Mn	2–5.0	0.27	13.5
Fe	15	6.60	44
Cu	1.3–3	0.24	18.46
Zn	15 or 12	10.47	15

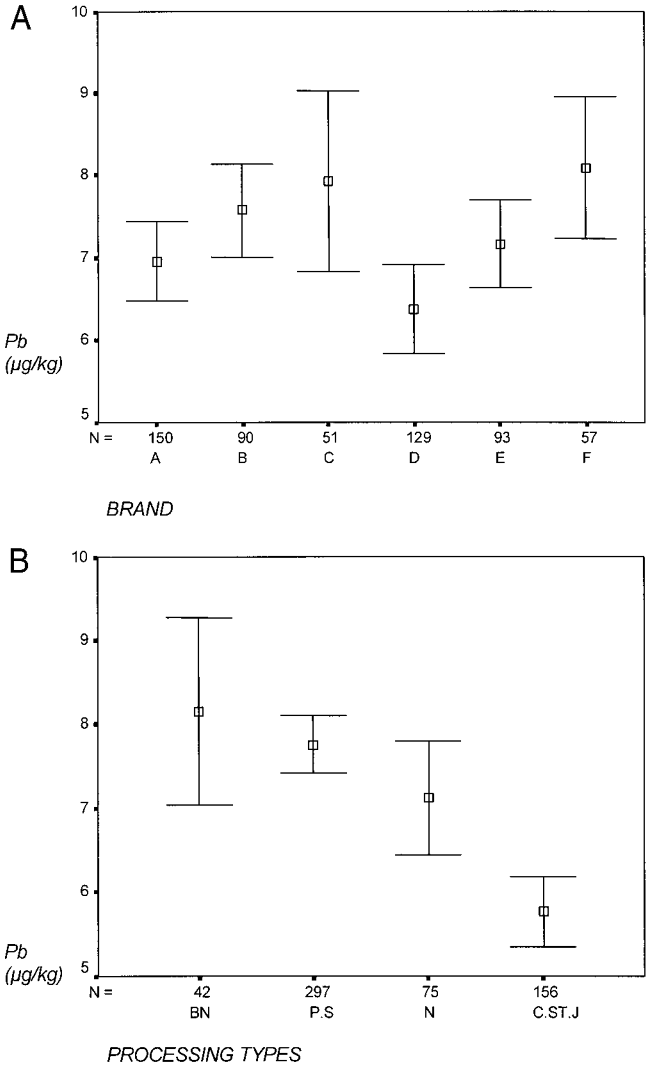

FIGURE 1. *Pb concentration in canned mussels (µg/kg wet weight) with respect to brand (A) and processing type (B).*

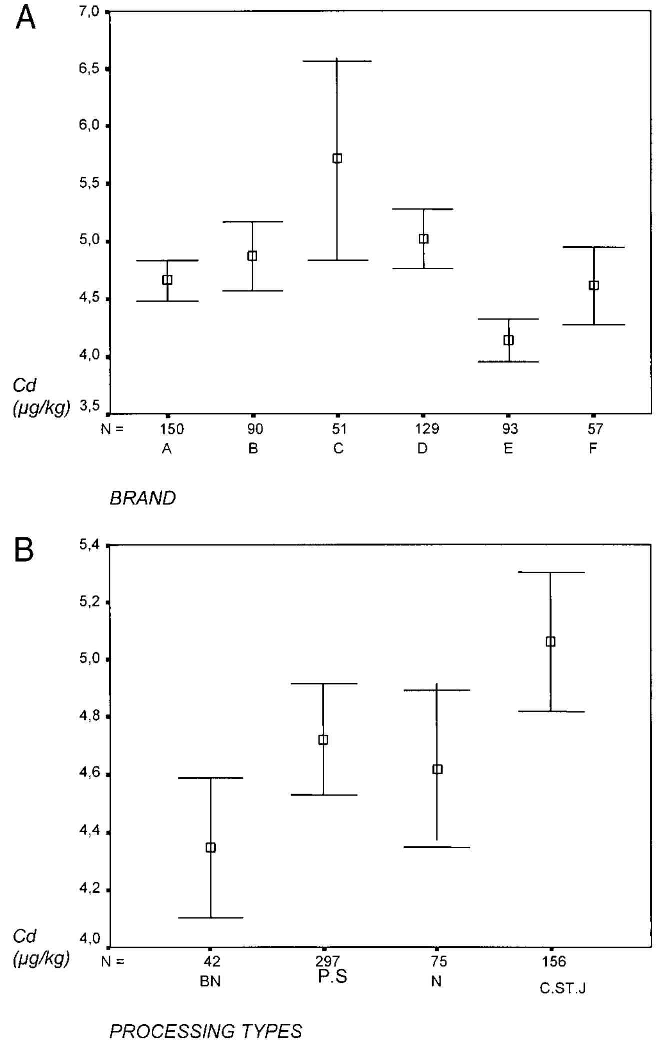

FIGURE 2. *Cd concentration in canned mussels (µg/kg wet weight) with respect to brand (A) and processing type (B).*

300 µg/kg in Anza (Saharian coast) *(18, 23).* For *Venus verrucosa,* 3,930 ± 2,840 µg/kg was obtained in Cansado Bay and 11,860 ± 4,820 µg/kg was obtained in Levrier Bay *(30).* These concentrations are higher than those found by us because these researchers carried out their analyses in areas susceptible to patent contamination.

All sample concentrations of Hg were below the limit of detection for the analysis equipment, which was 5 µg/kg. Because of the small amount of sample processed (0.5 g maximum), the signal was never higher than background, so we could not determine the exact concentration of this metal in these samples. This limit of detection is much lower than the maximum concentration set by European Community Decision 93/351/EEC *(12),* which is 500 µg/kg wet weight. Other authors have obtained Hg concentrations ranging from 10 to 210 µg/kg (mean, 60 µg/kg) in fish, including prawns, and from 10 to 130 µg/kg in shellfish, with means of 40 µg/kg *(25, 26)* and 54 µg/kg *(37).*

Brand C mussels contained the highest concentrations of Cr (2,120 ± 100 µg/kg), Fe (44,620 ± 4,700 µg/kg), Mn (1,800 ± 190 µg/kg), and Zn (69,830 ± 2,810 µg/kg) (Fig. 3). Zinc concentrations between 38,000 and 61,700 µg/kg have been reported in mussels and oysters *(18)* and concentrations between 35,100 and 69,400 µg/kg have been found in *Homarus americanus* digestive glands *(10).*

Brand C mussels were small, with a mean size between 18 and 24 mm, whereas in cans of the other brands the mean sizes were larger, between 24.5 and 42.4 mm. This finding indicates that the assimilation of essential metals is higher in less mature organisms, as has been hypothesized by others *(6, 34).* Brand D mussels had the highest concentrations of Ni (8,030 ± 350 µg/kg). Others have reported Ni concentrations ranging from 200 µg/kg *(2)* to 7,080 ± 1,570 µg/kg *(1),* in general agreement with the results of the present study.

Brand A mussels had the highest concentrations of Cu (1,760 ± 70 µg/kg). Cu concentrations are highly variable in bivalve molluscs, with reports of 3,900 to 49,800 µg/kg in mussels and oysters *(18)* and 5 and 20 µg/kg for *M. galloprovincialis (5).* Data collected from brand C mussels confirm that Pb, Cu, Zn, and Fe concentrations in *Mytilus* decrease with increasing mussel weight, whereas Ni and Cd concentrations remain constant *(8, 9).*

Tables 5 and 6 present the daily consumption of mussels (expressed in grams) necessary to exceed the admis-

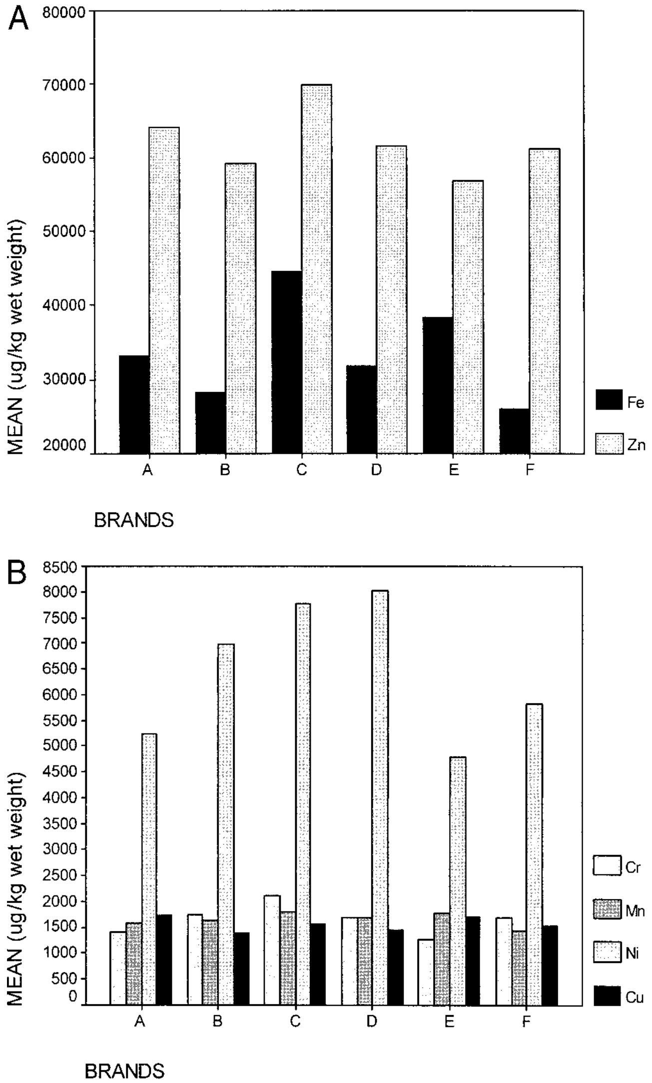

FIGURE 3. *Concentrations of essential heavy metals Zn and Fe (A) and Cr, Mn, Ni, and Cu (B) with respect to brand of canned mussels.*

sible daily allowance of Pb and Cd in humans. Because the amounts of mussels normally consumed are much lower than these thresholds, the mussel brands and preparations studied can be considered safe to consume.

We were unable to accurately assess Hg concentrations with the procedures used in this study because all concentrations were below the detection limits of the equipment. Thus, we concluded that Hg concentrations in these products are minimal with respect to the maximum legal limit for human consumption.

The nutritional significance of mussels, based on consumption of 150 g of canned mussels (approximately one can) for an adult, for the essential heavy metals that have recommended daily allowances (Mn, Fe, Cu, and Zn) is presented in Table 7. These data confirm the suitability of this food for human consumption. Because of low concentrations of toxic metals and high concentrations of essential metals, mussels have an important place among healthy high-quality foods according to recommendations of the World Health Orgaization published in 1988 *(35)*.

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