Journal of Food Protection, Vol. 68, No. 3, 2005, Pages 577–580

Cadmium in Cephalopod Molluscs: Implications for Public Health

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MS 04-316: Received 9 July 2004/Accepted 18 November 2004

ABSTRACT

Cadmium concentrations were measured in the flesh and hepatopancreas (digestive gland) of 1,392 specimens of different species of cephalopod molluscs (broadtail squid, spider octopus, curled octopus, horned octopus, elegant cuttlefish, and pink cuttlefish) to determine whether maximum levels fixed by the European Commission were exceeded. In all species, mean cadmium concentrations were higher in hepatopancreas than in flesh. Large differences among the different species were also observed. Pink cuttlefish and spider octopus had the highest concentrations for both flesh (spider octopus, 0.77 $\mu g\ g^{-1}$; pink cuttlefish, 0.87 $\mu g\ g^{-1}$) and hepatopancreas (spider octopus, 9.65 $\mu g\ g^{-1}$; pink cuttlefish, 18.03 $\mu g\ g^{-1}$), and the lowest concentrations were encountered in broadtail squid (flesh, 0.13 $\mu g\ g^{-1}$; hepatopancreas, 2.48 $\mu g\ g^{-1}$). The other species had intermediate concentrations of 0.20 to 0.30 $\mu g\ g^{-1}$ in flesh and 5.46 to 8.01 $\mu g\ g^{-1}$ in hepatopancreas. Concentrations exceeding the limit proposed by the European Commission (1.00 $\mu g\ g^{-1}$) were observed in 44.4 and 40.0% of flesh samples of spider octopus and pink cuttlefish, respectively. The estimated weekly intake, 0.09 to 0.66 μg/kg body weigh, was below the provisional tolerable weekly intake set by the World Health Organization.

In 1972 the World Health Organization of the United Nations (WHO) *(25)* issued a provisional tolerable weekly intake (PTWI) for cadmium of 400 to 500 μg per person. Following toxicological reevaluations, this PTWI was expressed in terms of cadmium intake per kg of body weight (7 μg/kg of body weight) *(27, 28)*. These cadmium advisories are necessary because this metal has a long half-life in the human body *(26)* and is responsible for a wide variety of adverse effects. Occupational cadmium exposure is associated with lung cancer *(11, 22)*, and in some studies occupational or environmental exposure to this metal has been associated with cancer of the prostate, kidney, liver, hematopoietic system, and stomach *(11, 23)*. Among nonsmoking and nonoccupationally exposed individuals, food is the main source of cadmium exposure *(12, 26)*. Evidence for the increased levels of cadmium in human foodstuffs has been obtained from studies of the levels of cadmium in human urine and blood samples from individuals who had not been exposed to this metal in the workplace *(18)*. Vegetables and cereals are the most significant sources of cadmium in the diet in the majority of countries *(7, 8, 10, 15, 24)*. Recent surveys have revealed that boiled rice contributes about 40% of the total cadmium intake in the Japanese general population *(10, 24)* and about 23% in the general population in Korea *(15)*. Among seafood, cephalopod molluscs are known for their ability to accumulate high levels of cadmium and may constitute another significant source of the human body burden of cadmium. In cephalopods, the hepatopancreas (digestive gland) is the major storage site of this element *(17, 19)*, containing up to 98% of the total body burden of cadmium in these animals *(2)*. This bioconcentration issue is of special interest for public health because cephalopod hepatopancreas is considered a delicacy. In southern Italy, cephalopods are eaten whole, and in Spain the whole animal is macerated and used as squid sauce. Consequently, assessment of cadmium content in the mantle and arms and in the hepatopancreas is important. The present work was undertaken to measure cadmium concentrations in the flesh and hepatopancreas of different species of cephalopod molluscs to evaluate the quality of these seafood products. Weekly human intake of cadmium deriving from the ingestion of cephalopods was then estimated and compared with the PTWI recommended for humans by the WHO *(28)*.

MATERIALS AND METHODS

In June through August 2003, 1,392 specimens of cephalopod molluscs belonging to different species (broadtail squid, spider octopus, curled octopus, horned octopus, elegant cuttlefish, and pink cuttlefish) were caught from the Adriatic Sea during several trawl surveys (Fig. 1). Specimen were grouped into pools according to their size and species. Table 1 lists the different species, the number of specimens, the weight range, and the number of pools for each species. From the specimens in each pool, flesh and hepatopancreas samples were obtained, homogenized, and analyzed. The analytical method for the determination of cadmium has been previously described *(19)*. Samples (1 to 2 g) were digested in a reaction flask with 11 ml of HNO_3-$HClO_4$ (8:3) using a hot plate at 150°C. More nitric acid was added until a completely colorless solution was obtained. After concentration, the residue was dissolved in 10 ml of deionized water. The solution was then made up to 25 ml with distilled water. The quantitative analysis

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FIGURE 1. *Cepaholpod sampling areas in Italy.*

was performed using a graphite furnace atomic absorption spectrophotometer (Analyst 800-THGA-800, Perkin Elmer, Foster City, Calif.). Three reagent blanks were similarly processed and used to determine detection limits of the analytical procedure. Each sample was analyzed in duplicate. Analyses of replicate samples indicated that the error did not exceed 7%. The recovery tests, which involved the addition of a known amount of inorganic cadmium before digestion, yielded a mean recovery of 96 ± 4%. The analytical procedure was validated using certified reference material (DORM-1, National Research Council, Ottawa, Ontario, Canada). The results of these cadmium analyses (0.080 ± 0.010 μg g^{-1} dry weight) were in close agreement with the reference values (0.086 ± 0.012 μg g^{-1} dry weight). All values were reported as micrograms per gram on a wet weight basis.

RESULTS AND DISCUSSION

Cadmium concentrations in the flesh and hepatopancreas of the different species analyzed are presented in Table 2. Cephalopod cadmium concentrations were substantially higher in hepatopancreas (2.48 to 9.65 μg g^{-1}) than in flesh (0.09 to 0.92 μg g^{-1}), which supports the hypothesis that cadmium accumulates in the digestive gland of cephalopods.

The hepatopancreas/flesh cadmium concentration ratio indicated that cadmium in the digestive gland was present at remarkably high concentrations, based on the criteria proposed by Miramand and Bentley *(14)*: <10, poorly concentrated; 10 to 50, moderately concentrated; >50, highly concentrated. The capacity of hepatopancreas to accumulate high quantities of cadmium by sequestration into organelles of hepatopancreas cells and by binding to cytosolic proteins has been reported *(1, 14, 19)*, and high concentrations have been found in organisms from unpolluted waters *(1)*. However, independent from the cadmium distribution pattern in these two tissues, cadmium concentrations were high variability among the different species. Pink cuttlefish and spider octopus had the highest concentrations in flesh (spider octopus, 0.77 μg g^{-1}; pink cuttlefish, 0.87 μg g^{-1}) and hepatopancreas (spider octopus, 9.65 μg g^{-1}; pink cuttlefish, 18.03 μg g^{-1}), whereas the lowest concentrations were found in broadtail squid (flesh, 0.13 μg g^{-1}; hepatopancreas, 2.48 μg g^{-1}). The other species had intermediate concentrations between 0.20 and 0.30 μg g^{-1} in flesh and between 5.46 and 8.01 μg g^{-1} in hepatopancreas. These large differences in cadmium concentrations are not unexpected. Cephalopods are known to be widely variable in cadmium concentrations with respect to families, geographical origin, and feeding habits *(1, 6, 17, 19, 20)*. Benthic cephalopods (octopii [Octopodidae] and cuttlefish [Sepiidae]) feeding mainly on bottom invertebrates *(13)* should have higher cadmium concentrations than the neritic and pelagic squids (Loliginidae), which prey mainly on fish and other cephalopods *(5)*. Our findings fit this general picture, with the lowest concentrations in Loliginidae, and the highest in Octopodidae and Sepiidae. Our findings also support those reported in literature, although in comparison to other marine groups, studies on cephalopods are sparse. Our results for broadtail squid were close to those reported by Storelli and Marcotrigiano *(19)* (flesh, 0.07 μg g^{-1}; hepatopancreas, 2.88 μg g^{-1}). Concentrations reported by Miramand and Bentley *(14)* for hepatopancreas of octopii (11.5 μg g^{-1}) and cuttlefish (5.34 μg g^{-1}) were comparable with our values. Bustamante et al. *(1)* reported concentrations in whole

TABLE 1. *Number of specimens and weights for different species of cephalopod molluscs*

Species	No. of specimens	Weight (g)[a]	No. of pools
Squid			
Illex coindeti (broadtail squid)	161	14.0–1,879.5 (417.7 ± 558.9)	14
Octopus			
Octopus salutii (spider octopus)	152	37.4–954.8 (221.5 ± 295.3)	9
Eledone cirrhosa (curled octopus)	122	44.2–247.0 (143.5 ± 111.4)	4
E. moschata (horned octopus)	200	15.0–476.7 (208.9 ± 133.7)	31
Cuttlefish			
Sepia elegans (elegant cuttlefish)	119	54.6–104.5 (77.3 ± 25.2)	4
S. orbignyana (pink cuttlefish)	638	6.8–99.9 (48.7 ± 35.2)	8

[a] Range (mean ± standard deviation).

TABLE 2. *Cadmium concentrations in the flesh and hepatopancreas of different species of cephalopod molluscs*

Species	Cadmium (μg g^{-1} wet wt)[a]	
	Flesh	Hepatopancreas
Squid		
Illex coindeti (broadtail squid)	0.06–0.26 (0.13 ± 0.09)	1.46–3.80 (2.48 ± 1.20)
Octopus		
Octopus salutii (spider octopus)	0.08–1.45 (0.77 ± 0.47)	4.57–16.61 (9.65 ± 3.93)
Eledone cirrhosa (curled octopus)	0.03–0.40 (0.23 ± 0.15)	6.05–11.81 (8.01 ± 2.88)
E. moschata (horned octopus)	0.01–0.73 (0.20 ± 0.19)	1.37–11.33 (5.46 ± 2.62)
Cuttlefish		
Sepia elegans (elegant cuttlefish)	0.15–0.47 (0.30 ± 0.10)	1.46–7.64 (4.08 ± 1.57)
S. orbignyana (pink cuttlefish)	0.33–1.48 (0.87 ± 0.63)	5.29–27.56 (18.03 ± 9.30)

[a] Range (mean ± standard deviation).

curled octopus (9.06 μg g^{-1}) of the same order of magnitude as those found in the present study.

Metal content in organisms suitable for human consumption is an important aspect in the evaluation of human toxicity. Food safety is of vital importance and requires special attention from competent authorities to prevent undesirable effects on human health. Concentration standards for cadmium, which when violated make seafood unsuitable for human consumption, have been established in various countries. Saudi Arabia has set a maximum limit of cadmium in fish and shellfish of 0.5 μg g^{-1}, in Croatia the limit is fixed at 0.1 μg g^{-1} wet weight, and the Italian legislature established solely for the flesh of cephalopod molluscs a maximum cadmium limit of 2 μg g^{-1} wet weight *(4)*. In Europe, the limit for cadmium in cephalopod molluscs recently proposed by the European Commission *(9)* is 1.00 μg g^{-1} wet weight. Using this value as a guideline, concentrations exceeding it were found in the present study in only 44.4 and 40% of flesh samples of spider octopus and pink cuttlefish, respectively, whereas results for all other species were below this limit. These findings seem to be reassuring, although the value set by the European Commission refers only to the flesh of these animals.

Estimates of the dietary intake of potentially harmful chemicals in food are important in assessing risks to human health. For cadmium, the WHO has established as safe a PTWI of 7 μg/kg of body weight. *(28)*. The weekly intake was calculated using the mean concentrations of cadmium in the flesh and hepatopancreas of cephalopods and assuming a weekly average consumption of 26 g per person *(19)*. The consumption of flesh only (25 g) resulted in an estimated weekly intake of 0.05 to 0.36 μg/kg of body weight, whereas the consumption of whole animal (26 g) yielded a weekly intake of 0.09 to 0.66 μg/kg of body weight (Table 3). Although these estimates based on cephalopod consumption are below the PTWI set by the WHO *(28)*, they do not take into account exposure from other foods. The type of diet and the quantity of food consumed can strongly increase cadmium exposure levels. Cadmium intakes were higher for persons consuming shellfish one or more times per week than for those eating a mixed diet with a low shellfish content *(21)*. Similarly, differences in the quantity of food consumed can impact metal intake. Based on our findings, a serving of 100 g of cephalopods with hepatopancreas (4 g) can result in a high weekly intake: 0.36 to 2.64 μg/kg of body weight. Weekly consumption at this level may not be rare. Countries near the sea probably have many frequent and heavy consumers of seafood. Therefore, recently the market demand for cephalopods has increased *(3)* and ''in response to decreases of traditional finfish stocks due to overfishing, cephalopods have gained increasing attention as alternative to the traditional marine harvest

TABLE 3. *Estimated human weekly intake of cadmium derived from consumption of cephalopod molluscs*

Species	Cadmium intake (μg/kg of body wt) from:		
	Flesh	Hepatopancreas	Whole animal
Squid			
Illex coindeti (broadtail squid)	0.05	0.04	0.09
Octopus			
Octopus salutii (spider octopus)	0.32	0.16	0.48
Eledone cirrhosa (curled octopus)	0.09	0.13	0.22
E. moschata (horned octopus)	0.08	0.09	0.17
Cuttlefish			
Sepia elegans (elegant cuttlefish)	0.12	0.07	0.19
S. orbignyana (pink cuttlefish)	0.36	0.30	0.66

and will gain a much larger importance in the future to supply mankind with marine living resources'' *(16).*

Does cadmium in these marine organisms pose a danger to human health? Although the estimated cadmium intake from this source is below the PTWI set by the WHO *(28)*, it is appropriate to consider that other foods (e.g., rice and shellfish), various activities (e.g., smoking), chronic diseases (e.g., diabetes and hypertension), and nutritional factors (e.g., iron deficiency) can influence exposure to and uptake of cadmium. The cadmium concentrations reported in this study are an important part of overall exposure in some persons, and thus continuous monitoring is needed to determine trends of cadmium contamination in marine organisms.

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