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Selenium and Mercury in Foodstuff from a Locality with Elevated Intake of Methylmercury

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Elevated intake of methylmercury via fish-consumption has been reported in the islanders living on the Tokara Islands, Kagoshima Prefecture, Japan; hair organomercury levels were very close to, or exceeded, the lowest toxic value, 50 μg/g, of the Niigata epidemic cases in some islanders (SUZUKI et al. 1976, 1979). However, neurological examinations with a multivariate statistical evaluation did not reveal methylmercury poisoning in the islanders (HAMADA & IGATA 1976). A possible explanation for the lack of clinical manifestation of methylmercury poisoning in spite of the increased intake of methylmercury may be concomitant intake of selenium, a modifying factor against methylmercury toxicity in animal experiments (GANTHER et al. 1972, IWATA et al. 1973, JOHNSON & POND 1974, POTTER & MATRONE 1974, STOEWSAND et al. 1974, UEDA et al. 1975, OHI et al. 1975a, b, 1976, HIROTA et al. 1977).

We have measured the mercury and selenium concentrations in foodstuffs, mainly in fish, obtained on the Takarajima Island of the Tokara Islands and the surrounding sea. The selenium consumption will be evaluated in comparison with mercurial consumption.

MATERIALS AND METHODS

Sampling. In April 1978, two of the authors visited Takarajima and Amami Islands; the latter is located 90 km south of the former. They caught fish by rod-line in a coral sea surrounding Takarajima Is., and bought fish in a fish-market on Amami Is. Shellfish, *Purpura armigera*, was also collected from the coral sea, and other foodstuffs (rice, peanut and lobster meat) were sampled from households on Takarajima Island.

Analysis. Samples of fish and shellfish were transported to the laboratory in an ice-box with solid car-

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bon dioxide and ice and then stored in a freezer until analyzed. Preparation of samples involved cutting fish into fillets after removing the scale and selecting of the part close to the gill at the dorsal half of the fillet. Foodstuffs other than fish and shellfish were obtained in dried state. The samples of fish fillet and other foodstuffs were minced or crushed with a food-processor.

For Hg measurement, 1-2 g of samples were homogenized in a solution of 1 N HCl and separated into the supernatant and the precipitate. The precipitate containing most of the inorganic Hg was digested in 1 mL of 1% cysteine-HCl solution and 4 mL of 22.5% NaOH solution for 2-4 h at room temperature to avoid interference due to possibly coexisting Se (NAGANUMA et al. 1979, YAMAMOTO et al. 1980). The supernatant was measured without digestion. Inorganic and total Hg were measured by the MAGOS (1971) method. The sensitivity of this method in our laboratory was 2.5 pmol of Hg.

Se was measured by the fluorometric method (WATKINSON 1966) after ashing the sample using a Uniseal digestion vessel (Uniseal Decomposition Vessels, Ltd., Israel) : To 1 g of sample, 2 mL of conc. HNO_3 were added, and the resulting mixture was heated at 140°C for 2 h. This procedure recovered 97% of Se (as Na_2SeO_3) added. The fluorometry was sensitive to 10 pmol of Se.

RESULTS

Mercury and Selenium in Fish Fillet. Among fishes of the coral sea, the total Hg concentration was different depending upon the species (Table 1). No statistically significant correlation was found between the total Hg level and the body weight of fish for any species with adequate numbers for correlation analysis. The species-dependent difference was also found as to the percentage of inorganic to total Hg, but the other two variables, feed habit and body weight, did not show any association to this percentage.

The oceanic fish had distinctly elevated total Hg concentrations compared to the fish inhabiting coral sea areas (Table 2). Nevertheless, the percentage of inorganic Hg did not differ from the values in the coral sea fish.

The level of Se was rather uniform in all the species studied, even if occasional high values were noted (Tables 1&2). Thus, the molar ratio of Se to Hg in fish fillets was as high as 6 to 88 in coral sea fish, in

TABLE 1

Mercury and Selenium in the Fillet of Fish caught in the Coral Sea surrounding the Takarajima Island

Scientific Name (local name)	Feed[a] Habit	No. of samples	Weight(g) max.	Weight(g) min.	Hg(nmol/g) total	Hg(nmol/g) %inorganic[b]	Se(nmol/g)
Epinephelus fario (chota)	c	12	410	20	0.23±0.09[c]	9.1± 7.2	5.1±4.5
Cirrhitus pinnulatus (gabu)	c	3	230	81	0.41±0.01	9.1± 9.1	3.3±1.2
Hapalogenys nigripinnis (higedai)	o	9	138	30	0.24±0.08	2.8± 1.4	3.4±2.1
Thalassoma spp. (kusabi)	o	7	96	48	0.09±0.03	13 ± 9	3.4±0.7
Xanthichthys spp. (fukurobe)	c	3	380	144	0.14±0.07	23 ±16	5.1±2.3
Holocentrus spp. (akaio)	c	1	58		0.58	8.1	10.2
Girella punctata (subatsu)	o	1	1.400		0.04	18.4	2.1

a. Feed habit of fishes (c, carnivore; o, ominivore); habitat was coral areas: Both feed habit and habitat of fishes were referred to the report by MASUDA et al. (1975).
b. "% inorganic" means the percentage of inorganic mercury to total mercury.
c. Figures show mean±standard deviation.

TABLE 2

Mercury and Selenium in the Fillet of Fish purchased in the Market on Amami Island

Scientific name (local name)	Feed Habit	Habitat[a]	No. of samples	Hg (nmol/g)		Se (nmol/g)
				total	%inorganic	
Parathunnus sibi (shibi)	c	o	1	2.3	8.7	7.9
Coryphaena hippurus (manbiki)	c	o	1	1.1	8.1	12
Acanthocybium solandri (sawara)	c	o	1	3.7	8.4	1.2
Scombrops spp. (kuromatsu)	c	o	1	11	12	2.9
Scaridae family (erabuchi)	o	c	1	0.07	35	2.2

a. Habitat (c, coral area; o, ocean).

TABLE 3

Mercury and Selenium in Some Foodstuffs on Takarajima Island

Foodstuff	No. of sample	Hg(nmol/g) total	%inorganic	Se (nmol/g)
dried lobster meat[a]	3	0.39±0.06 [b]	3.1±2.7	8.7±4.9
meat of shellfish[c]	5	0.19±0.07	87 ± 9	8.9±1.5
rice 1[d]	-	0.015	100	6.6
2	-	0.009	100	1.8
peanut	-	0.058	71	1.5

a, Only the dried meat was available, and the most popular species in this locality is *Panulirus longipes*.
b, mean ± standard deviation
c, Shellfish is called "agarinishi", generic name is *Purpura armigera*.
d, Kernel of rice was analysed. Rice 1, non-glutinous; rice 2, glutinous.

contrast to values as low as 0.3 to 11 in the oceanic fish.

Mercury and Selenium in Other Foodstuffs. Dried lobster meat and meat of shellfish had similar total Hg levels to and slightly higher Se levels than those found in the fillet of coral sea fish. The percentage of inorganic Hg in the lobster meat was very low (3%) and that in the shellfish meat was very high (87%). Rice and peanut were very low in total Hg with very high percentage of inorganic Hg and comparable to marine products in the Se levels (Table 3).

DISCUSSION

In the foodstuffs analysed, ubiquity of Se has made a marked contrast to selective accumulation of Hg. Elevated levels of Hg in the form of organomercury, presumably methylmercury, were common in the oceanic fish, and the levels found were comparable with those already reported in the similar species caught in the sea surrounding the Tokara Islands (SUZUKI 1971, SUZUKI et al. 1976).

Since this is a first report on Se concentration in foodstuffs in this locality, there is no reference for comparison. However, the levels of Se found are in the range of values reported in domestic (YASUMOTO et al. 1976, NISHIGAKI et al. 1977) and foreign (SCOTT 1972) literature. Among the samples of foodstuffs which have been conventionally consumed in Japan, the marine products have had the highest concentration of Se and the next highest has been found in cereals, especially rice and pulses (soya bean and others). It was for these reasons that we sampled marine fish as well as rice and peanut in this locality. Considering the overall consumption, the most contributing foodstuff is rice for the intake of Se in this locality.

Thus, the molar ratio of Se to Hg in the total food and the amount of Hg ingested is variable according to the kind of fish consumed, and even in the case when only oceanic fish is consumed, the intake of Se is several times greater than that of Hg on a molar basis.

No accurate estimation for Se intake is available for the Niigata epidemic cases. If we apply the value, 200 μg/day (2.5 μmol/day), an estimation based on a standardized composition of foodstuffs for the average Japanese man (YASUMOTO et al. 1976), to these cases, and compare it with the estimated lowest toxic daily intake of methylmercury, 300 μg Hg/day (1.5 μmol/day), which was proposed as corresponding to the lowest toxic hair mercury value in the Niigata epidemic cases (SWEDISH EXPERT GROUP 1971), the molar ratio of Se to Hg will be 1.7:1. Hair mercury values distributed in a range from 50 to 570 μg/g (median: ca. 230 μg/g) in the patients of Niigata epidemic (TSUBAKI & IRUKAYAMA 1977). Providing the daily intake of Se was invariable in these patients, the molar ratio of Se to Hg would have been variable and in most of the patients, excess of Hg should have been common.

Relative efficacy of Se in fish is small compared to selenite in modifying the methylmercury toxicity in rats (OHI et al. 1976), and both selenite and natural Se in wheat prevent acute toxic effects of methylmercury in Japanese quail, whereas only in selenite, but not natural Se, fed group, methylmercury feeding depresses egg production and enhances eggshell thinning (STOEWSAND et al. 1977). Except for the above-mentioned results, little has been elucidated on modification of methylmercury toxicity by Se contained in foodstuffs. Therefore, the molar ratio of Se to Hg in the total food and relative efficacy of Se in foodstuffs are both indispensable in discussing the role of Se against methylmercury toxicity. The present results only suggest a possible role of Se as preventing the occurrence of methylmercury poisoning in the Tokara islanders.

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