The Chemical Form and Bodily Distribution of Mercury in Marine Fish

by

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The chemical form of mercury in fish has been controversial. Whereas the mercury in the muscle of fish has been reported to be mostly methylmercury by the workers in Sweden (WESTÖÖ 1966; WESTÖÖ and RYDÄLV 1969; WESTÖÖ and RYDÄLV 1971), the United States (SMITH et al. 1971), and Canada (ZITKO et al. 1971; UTEH et al. 1972), the methylmercury percentage of total mercury content has been considerably lower in the reports from Japan (UEDA et al. 1971), and Canada (BACHE et al. 1971). The difference of analytical procedures, the kind of fish, and the chemical form of mercury in the environment have to be implicated. A comparative analysis for methylmercury and total mercury on the same fish sample by a Swedish and a Japanese laboratory, did not give a satisfactory solution, e.g., the methylmercury percentage was of the same order in some samples, but different in other samples (BERGLUND et al. 1971).

Our intention in this report is to check the percentage of inorganic mercury in fish using the selective measurement of inorganic mercury by cold atomic absorption photometry (MAGOS and CERNIK 1969).

Samples and Methods

Fish to be measured for the mercury content was caught by a ship belonging to Tokyo Fishery College in the South China Sea (A to J in Table 1) and near the coast of northwestern Australia (K) in 1970, and was preserved in a freezer until measurement. A few other fish caught near Sado Island in the Japan Sea in 1971 was also measured (L & M).

One gram of muscle, liver, brain, kidney and gastric content of fish, or about half of the organ with a weight of less than 2 g was homogenized in a phosphate-buffer solution (pH 8.6, 1/16 M), and analyzed for inorganic mercury. For the measurement of total mercury, the same amount of each organ was oxidized with a solution of conc. H_2SO_4 and conc. HNO_3 in a reflux condenser. The homogenates and the

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TABLE 1

Fishes Measured for the Mercury Content

Kind of Fish		Weight (g)
(A) A kind of frigate mackerel	Auxis tapeinosoma Bleeker	100
(B) Hairtail	Trichiurus lepturus Linné	700
(C) A kind of jack mackerel	Trachurus japonicus (Temminck & Schlegel)	135
(D) A kind of jack mackerel	Caranx sexfasciatus Quoy & Gaimard 1)* 2)	165 140
(E) Blue runner	Caranx equula Temminck & Schlegel	50
(F) Yellowtail	Seriola quinqueradiata Temminck & Schlegel	900
(G) Catalufa	Priacanthus macracanthus Cuvier 1) 2)	260 180
(H) A kind of grouper	Epinephelus septemfasciatus (Thunberg)	150
(I) Nemipterid	Nemipterus virgatus (Houttuyn)	70
(J) Crimson Sea Bream	Evyunis japonica Tanaka	400
(K) Lethrinid	Lethrinus choerorhyncus (Block & Schneider)	530
(L) A kind of rockfish	Sebastes inermis Cuvier	50
(M) A kind of greenling	Hexagrammos otakii Jordan & Stards 1) 2) 3)	70 70 50

* Numbers indicate different specimens.

oxidized solutions were analyzed by mercury vapor photometry (MAGOS and CERNIK 1969). These analytical procedures for the selective measurement of inorganic mercury have previously been tested on human autopsy specimens and have yielded satisfactory results (SUZUKI et al. in press).

Results

(1) The Chemical Form and the Level of Mercury in Organs and Tissues.

Total and inorganic mercury content, and inorganic mercury percentages in the muscle, liver, brain, and gastric content are summarized in Table 2.

In the muscle, the highest inorganic mercury percentage is 17.6 in a catalufa. Weight of fish and the level of total mercury are independent of the inorganic mercury percentage. Except for fish caught in the Japan Sea, weight of fish shows a significant correlation with the total mercury content ($r=0.558$, $p < 0.05$), whereas the correlation coefficient calculated on all the fishes is not significant ($r=0.465$, $p > 0.05$). In other words, the fishes from the Japan Sea have a high mercury content for their small bodies.

Comparing the values in the muscle, both inorganic mercury percentages and levels of total mercury are elevated in the liver and kidney. The total mercury content in the liver, excluding the data on fish from the Japan Sea, is also significantly correlated with body weight ($r=0.582$, $p < 0.05$), but this is not seen in the case of the kidney.

The data on the brain are difficult to interpret both regarding the level of total mercury and the inorganic mercury percentage. In the case of a single hairtail, the level of total mercury is quite high (7.36 μg/g) and all the mercury is organic. The correlation of body weight to the level of total mercury is not significant.

The gastric content of fish was obtained from 10 fishes. The level of total mercury is markedly higher in greenlings from the Japan Sea than in other fishes from the South China Sea, which means a difference in the mercury level in the habitat, and may contribute to the differing relationships between body weight and the total mercury level in the muscle and liver. The inorganic mercury percentages scatters from 0 to 100, but the number of samples is too small to warrant discussion of the variability.

TABLE 2

Total Mercury Content and Inorganic Mercury Percentages in Organs and Gastric Content of Fish

Organ and gastric content	No. of specimens	Total Hg µg/g Mean	Total Hg µg/g S.D.	Specimens not measured or not obtained
Muscle	17	0.197	0.321	None
Liver	17	0.862	1.084	None
Brain	15	0.662	1.877	(A) (M-3)
Kidney	11	0.720	0.792	(A) (E) (H) (G-2) (L) (M-3)
Gastric content	10	0.072	0.060	(B) (C) (D-1) (F) (H) (K) (L)

Organ and gastric content	No. of specimens	Inorganic Hg µg/g Mean	Inorganic Hg µg/g S.D.	Inorganic Hg % total Hg Mean	Inorganic Hg % total Hg S.D.	Specimens not measured or not obtained
Muscle	16	0.006	0.010	5.2	6.1	(C)
Liver	17	0.267	0.495	34.8	27.0	None
Brain	13	0.015	0.019	15.7	22.8	(A) (C) (M-2) (M-3)
Kidney	11	0.247	0.357	32.7	27.6	(A) (E) (H) (G-2) (L) (M-3)
Gastric content	10	0.030	0.035	43.7	37.3	(B) (C) (D-1) (F) (H) (K) (L)

(2) The Bodily Distribution of Mercury.

The pattern of bodily distribution changes depending on the chemical form of mercury. In the case of inorganic mercury, three significant correlation coefficients are found, that is, "kidney" vs. "muscle" and "liver", and "liver" vs. "brain". There is a different pattern for organic mercury (calculated by deducting inorganic mercury from total mercury). Here the significantly correlated pairs are "muscle" vs.

TABLE 3

Correlation between Organ Mercury Content and Inorganic Mercury Percentages

		Brain	Liver	Kidney
Muscle	a) Total Hg	0.732**	0.528*	0.831**
	b) Inorganic Hg	0.155	0.207	0.737*
	c) Organic Hg	0.776**	0.618*	0.863**
	d) Inorganic Hg %	0.748**	0.316	-0.313
Brain	a) Total Hg	-	0.063	0.498
	b) Inorganic Hg	-	0.603*	0.319
	c) Organic Hg	-	0.373	0.752*
	d) Inorganic Hg %	-	0.365	-0.136
Liver	a) Total Hg	-	-	0.210
	b) Inorganic Hg	-	-	0.846**
	c) Organic Hg	-	-	0.090
	d) Inorganic Hg %	-	-	0.104

Notes: 1) For the calculation of correlation coefficients, inorganic mercury percentages are converted to the arc sine.

2) The number of data used for the calculation differs according to the pair of organs; muscle vs. brain a): 15, b)-d): 13; muscle vs. liver a): 17, b)-d): 16; muscle vs. kidney a): 11, b)-d): 10; brain vs. liver a): 15, b)-d): 13; brain vs. kidney a): 11, b)-d): 9; liver vs. kidney a)-d): 11.

* $p < 0.05$, ** $p < 0.01$

"brain", "liver", and "kidney", and "brain" vs. "kidney" (Table 3). The distribution pattern of total mercury is similar to that of organic mercury.

(3) Mercury in the Gastric Content of Fish and Its Relation to Organ Mercury Content.

In connection with the biological concentration of mercury through food chains, the relationship between the mercury content in the gastric content and that in the organ should have particularly significant meaning, if the gastric content sampled reflect the usual condition of food for fishes. Because of the small number of samples, we, at this time, only draw attention to several significant correlations between

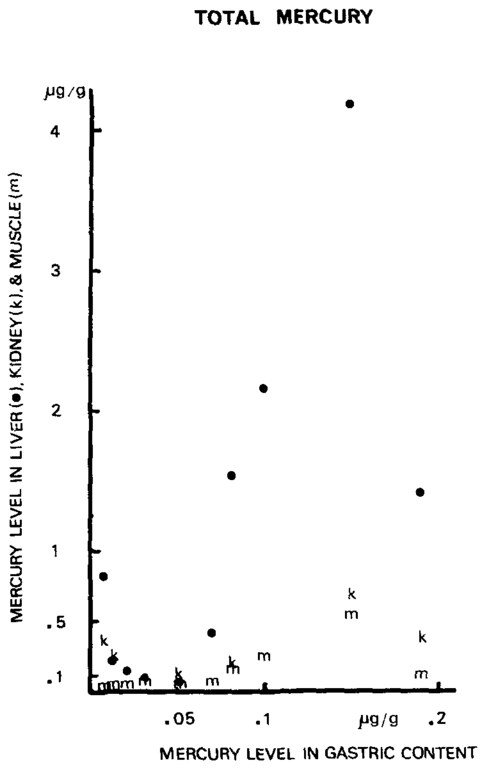

FIGURE 1. Total mercury content in organs and gastric content.

the level of total mercury in the gastric content and the liver or the muscle ($r=0.693$ and 0.655, respectively, $p < 0.05$); and between the levels of inorganic mercury in the gastric content and the liver, the kidney or the muscle ($r=0.898$, 0.939, and 0.717, respectively; in the former two, $p < 0.01$, and in the last, $p < 0.05$). As far as the level of total mercury is concerned, the extent of biological concentration, which can be shown as the ratio of mercury level in the muscle to that in the gastric content, is less than double, on the average, while the level of inorganic mercury in the muscle is one tenth of that in the gastric content (Figures 1 & 2). The liver, of which total and inorganic mercury levels are 15 times more than those in the gastric content, on the average, seems to have accumulated the mercury from the

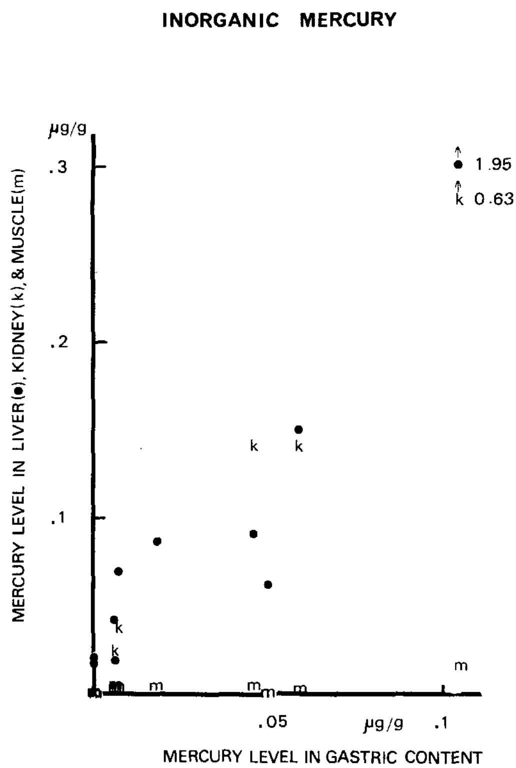

FIGURE 2. Inorganic mercury content in organs and gastric content.

food regardless of chemical form, but the kidney seems to work selectively for the inorganic mercury.

Discussion

As to the central purpose of this report, which was to know quantitatively the chemical form of mercury in fish, the results on the muscle are in agreement with the reports having shown the highest methylmercury percentage as about 15% (WESTÖÖ 1966; WESTÖÖ and RYDÄLV 1969; WESTÖÖ and RYDÄLV 1971; SMITH et al. 1971; ZITKO et al. 1971; UTEH et al. 1972). It is quite obvious from the present results that the methylmercury percentage would lessen when the whole fish is measured as in the case reported by BACHE et al. (1971).

Findings by OHMOMO et al. (1969) and BÄCKSTRÖM (1969) on the bodily distribution of mercury in freshwater fish after administration of inorganic mercury, phenylmercury or methylmercury are helpful in interpreting the present results. Though there were variations through time after administration, the three mercurials gave different patterns of distribution. Inorganic mercury concentrated in the kidney, gill, and pseudobranch with a moderate uptake in the liver, bile, spleen, but not in the nervous system and muscle. Methylmercury was rather uniformly distributed, and some of organs, i.e., the liver, spleen, kidney, gill, and pseudobranch, showed a relatively high accumulation. Phenylmercury could be said to have an intermediate pattern between these other patterns.

Based on these observations, the present results that the bodily distribution pattern of mercury differed according to the chemical form of mercury can be simply interpreted as a natural consequence of eating foods containing both inorganic mercury and methylmercury. However, the possibility of biotransformation of mercury in fish should be kept in mind. For example, UKITA and IMURA (1971) reported that only the liver of yellowfin tuna among several species of marine fishes and mammals studied had methylated inorganic mercury under the experimental conditions employed. Besides methylation, the breakage of C-Hg bond in fish is also possible. In the experiment by BÄCKSTRÖM (1969), the distribution patterns of parenterally administered mercurials changed gradually and the differences among three mercurials became less 30 days after the injection. This may have resulted from the biotransformation of mercury in fish. From the viewpoint of pharmacokinetics, because the fish is slow in many aspects of kinetics of mercury, we need long-term experimental observations to tie the experimental data to the actual living condition.

As for the 10 fishes examined, the biological concentration of mercury by the muscle is eventually related to organic mercury and the extent of concentration is not large. But, if the liver and the kidney of fish are involved, the biological concentration of mercury, both inorganic and organic, becomes remarkably dominant. A hairtail and a lethrinid, which showed the highest total mercury content in the muscle (0.98 μg/g) had no gastric contents for analysis. As UKITA and IMURA (1971) have suggested that there may be particular species of fishes which are capable in methylating inorganic mercury, these two species have to be listed for further studies in addition to the tuna and the swordfish groups.

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