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Total Mercury Content in Yellowfin and Bigeye Tuna

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Prompted by the Minimata and Nuiigata poisonings in Japan which were responsible for neurological damage and death to scores of people who had consumed fish and shellfish contaminated with an alkylmercury compound, monomethylmercury (KURLAND et al. 1960), and later by bird kills in Sweden (BORG et al. 1966) and fish contamination in Canada and the United States (FRIBERG & VOSTAL 1972), considerable attention has been directed toward quantifying and toxicological interpretation of mercury present in fish. Most, if not all, of these early contamination reports were traced to industrial and agricultural sources. However, in the early 1970's, open-ocean, pelagic species such as tuna, shark, swordfish, and marlin were often found that exceeded the current provisional maximum permissible level of 1.0 mg Hg/kg established by the U.S. Food and Drug Administration (1979). In the absence of obvious industrial sources, naturally-occurring sources of mercury are under scrutiny, as is the ability and extent of certain bacteria to transform and methylate inorganic mercury (JENSEN & JERNELOV 1969).

The object of this paper is to report and attempt to interpret the analytical findings relating to total mercury levels in samples of bigeye (Thunnus obesus) and yellowfin (T. albacares) tuna.

METHODS AND MATERIALS

Tuna samples were secured from the commercial public fish auction in Honolulu, Hawaii during the months of March and April 1971, and were caught by long-line in coastal waters of Hawaii. Each sample, representing an individual fish, was taken from the caudal peduncle area (base of tail) and consisted of 500 to 1500 g muscle (cleaned of skin and bones). A total of 100 samples of bigeye and 104 samples of yellowfin tuna were analyzed. The samples were maintained frozen until sample preparation for analysis. They were then thawed and homogenized (250 g) with a Waring blender. No water was added to the tissue.

Total mercury levels were analyzed by first using determination with cold vapor atomic absorption developed by HATCH & OTT (1968). Five grams of homogenate from each sample were digested in concentrated sulfuric-nitric acid (4 to 1) and further oxidized with concentrated nitric acid if charring occurred. Following digestion, excess oxidizing agents were reduced with a sodium chloride-hydroxylamine solution. Mercury ions (Hg^{++}) were then reduced to elemental mercury (Hg°) with stannous sulfate solution, and the mercury vaporized in the flameless atomic absorption apparatus. Depending on mercury content, a 5 to 25 mL aliquot of the digest was placed in the sample reservoir of the "closed-loop" mercury analysis system together with 50 mL of reducing solution plus sufficient distilled water to bring the total volume to 100 mL. The system was then immediately closed, and the mixture was stirred for 1 min after which airflow was initiated and maximum absorption at 2537Å noted. Standard operating conditions were employed, and mercury content calculated by comparison of sample responses to a standard curve derived from a series of $HgCl_2$ standards. Subsamples were submitted for analysis to the WARF Laboratory, Madison, Wisconsin and to the Laboratory of Hygiene of the Wisconsin State Board of Health, Madison, Wisconsin for comparison purposes. Our results showed good agreement with values obtained by the two independent laboratories. Maximum deviation was 0.05 ppm, which represented a difference of 6%.

RESULTS AND DISCUSSION

Yellowfin Tuna:

One hundred samples were analyzed for total mercury concentration. In each case the weight of the fish from which the sample was taken was recorded. A data summary is shown in Table 1.

Table 1. Data Summary: Mercury levels in 100 samples of yellowfin tuna muscle tissue.

Total No. Samples	Weights of Fish Sampled (lb)			Concentrations of Total Mercury (ppm)		
Analyzed	Range	Mean	Std. Dev.	Range	Mean	Std. Dev.
100	22-186	101	23	0.09-0.39	0.22	0.06

In order to determine if there was correlation between mercury concentration and fish weight, a linear regression analysis of the former (in ppm) on the latter (in pounds) was performed. The 'least-squares' method to best fit a straight line to a series of data pairs was employed (STEEL & TORRIE 1960). The regression curve is shown in Figure 1, and a summary of the results is presented in Table 2. In general, these data are in close agreement with the findings of RIVERS et al. (1972).

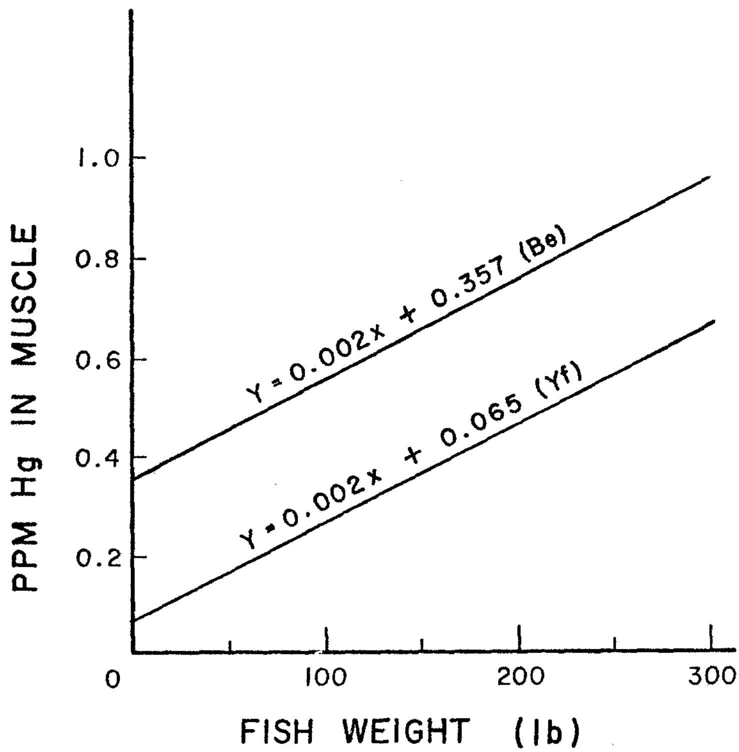

Figure 1. Regression curves describing the relationship of total mercury concentration (wet weight) in muscle tissue of yellowfin (Yf) and bigeye (Be) tuna to fish weight.

Table 2. Data Summary: Regression of mercury concentration (ppm) on weight (lb) for yellowfin tuna.

No. of Data Pairs	Correlation Coefficient	Slope	Y - Intercept
100	0.540**	0.002	0.065

** p 0.01 Highly Significant

A positive correlation between fish weight and mercury concentration is indicated. An approximate increase in mercury concentration of 0.02 ppm is observed for each 10 pounds of additional fish weight.

Bigeye Tuna:

One hundred and four samples of bigeye tuna muscle tissue were analyzed for total mercury concentration. The weight of the fish from which each sample was taken was recorded. A data summary is presented in Table 3.

Table 3. Data Summary: Mercury levels in 104 samples of bigeye tuna muscle tissue.

Total No. Samples	Weights of Fish Sampled (lb)			Concentrations of Total Mercury (ppm)		
Analyzed	Range	Mean	Std. Dev.	Range	Mean	Std. Dev.
104	47-224	126	40	0.30-0.87	0.58	0.13

The linear regression procedure of STEEL & TORRIE (1960) was again performed to determine if there was any correlation between mercury concentration and fish weight in the bigeye population. The regression curve is shown in Figure 1. A summary of the results is presented in Table 4.

Table 4. Data Summary: Regression of mercury concentration (ppm) on fish weight (lb) for bigeye tuna.

No. of Data Pairs	Correlation Coefficient	Slope	Y - Intercept
104	0.557**	0.002	0.357

** p 0.01 Highly Significant

A positive correlation between fish weight and mercury concentration is evident. As in the case of the yellowfin data, a slope of the regression line of 0.002 indicates that an increase in mercury concentration of approximately 0.02 ppm is observed with each addition of 10 pounds in fish weight. The level of contamination in the bigeye population, however, is found to be markedly higher than in yellowfin tuna. A comparison between the two species of the mercury concentrations expected in individual fish of a given weight based on the equations of the regression curves is presented in Table 5.

Table 5. Comparison of expected levels of total mercury contamination in bigeye and yellowfin tuna of various weights. Based on the results of linear regression analyses previously reported.

Fish Weight (lb)	Expected Level of Total Mercury Contamination (ppm) Yellowfin	Bigeye
50	0.17	0.46
75	0.22	0.51
100	0.27	0.56
150	0.37	0.66
200	0.47	0.76
250	0.57	0.86

Another method was employed to compare the level of contamination in the two species of tuna. A comparison of the average of the level of contamination per pound of fish in each of the individuals representing the two species was conducted. This was done by dividing the concentration of mercury (ppm) in each fish by the fish weight (lb) and then calculating the arithmetic mean of the resulting quotients.

A one-factor analysis of variance (STEEL & TORRIE 1960) was performed to determine the level of significance of the resulting difference. The results are presented in Table 6.

Table 6. Comparison of the level of total mercury contamination observed in the muscle tissues of yellowfin and bigeye tuna. The mercury level in each fish (ppm) was divided by the fish weight; the mean of the quotients within each group was calculated.

	No. Samples Analyzed	Hg Conc. (ppm) Divided by Fish Weight (lb) x 1000 Range	Mean	Std. Dev.	F-Ratio
Yellowfin	100	1.02-10.90	2.26	1.11	233.5**
Bigeye	104	2.14-9.84	4.91	1.36	

**p 0.005 Highly Significant

In the samples of yellowfin muscle tissue analyzed, a range in total mercury concentration of 0.09-0.03 ppm was observed; the mean concentration was 0.22 ppm. In the samples of bigeye muscle tissue the range was 0.30-0.87 ppm; the mean concentration 0.58 ppm. A positive correlation between mercury level and fish weight was observed in each of the two species. The level of total mercury in bigeye tuna was significantly higher than in yellowfin tuna. This was determined by comparing the mercury concentration per pound of fish in each individual sampled.

In discussing food habits and behavior of bigeye and yellowfin tuna, WHEELER (1975) says that yellowfin are migratory, seasonal in movement, swim near the surface, and feed on surface-living fish especially Scomberesox ("skippers"), Exocoetidae (flying fish), squid, and pelagic crustaceans. He describes bigeye tuna as strongly migratory, with schools suspected of travelling hundreds, possibly thousands of miles, that they range into deep water, especially during daylight, and feed on crustaceans (mainly prawns), squid, and locally common fish. The two species appear to have much in common, yet bigeye contain, on a weight-for-weight basis, approximately twice the level of mercury as does yellowfin tuna. Perhaps toxicant intake through diet is equal but the fish differ as regards protein binding.

With respect to mercury contamination of marine species with no known or apparent ties to industrial or agricultural anthropogenic pollution, several questions remain an enigma. We still do not know the source(s) of mercury, nor do we know that this is a new problem.

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