# MERCURY IN FISH AND SHELLFISH OF THE NORTHEAST PACIFIC. I. PACIFIC HALIBUT, *HIPPOGLOSSUS STENOLEPIS*

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#### ABSTRACT

A total of 1,227 Pacific halibut, *Hippoglossus stenolepis*, were analyzed for mercury content in the edible muscle tissue. These fish were obtained from five geographical areas within the species range: the Bering Sea, Gulf of Alaska, southeast Alaska, British Columbia, and Washington-Oregon. Mercury was found to be uniformly distributed from nape to tail in the edible muscle tissue. Within each geographical area the mercury concentration increased as the size of the fish increased. The mercury concentration also increased in fish of the same size from the northern to the southern part of the species range.

In the past few years, numerous investigators have examined the distribution and levels of mercury in food, including aquatic food animals, because of the potential health hazards involved. The U.S. Food and Drug Administration established an administrative guideline of 0.50 ppm mercury in fish and shellfish in 1969. Since that time, the guideline has been the subject of several reviews and recently has been proposed as a formal action level (Schmidt 1974).

Since 1970, the Pacific Utilization Research Center (PURC) and the Southeast Utilization Research Center (SEURC) at College Park, Md., have been conducting extensive studies of fish and shellfish taken from marine and inland waters of the United States to determine the extent to which mercury exceeds the guideline in our aquatic resources. This paper reports our findings on mercury in the edible tissue of the Pacific halibut, *Hippoglossus stenolepis* Schmidt.

### EXPERIMENTAL PROCEDURE AND METHODS

Halibut were obtained from commercial fishing vessels, fish processing companies, and research vessels of the International Pacific Halibut Commission (IPHC). Data were obtained on area and date of catch, and weight or length of each fish analyzed. Data were also obtained on age and sex when possible.

The five areas of catch were: Washington-Oregon, British Columbia, southeast Alaska, Gulf of Alaska, and the Bering Sea (Figure 1). Commercial halibut are eviscerated at sea, landed as a heads-on eviscerated product, and then beheaded for marketing as fresh or frozen fish. Weights reported here are in pounds for heads-off eviscerated fish because this is the standard practice of the halibut industry. For convenience of some readers who do not normally use our measurement system, approximate metric equivalents in kilograms are given in the tables and figures. When actual weights were impractical to obtain, the lengths of the heads-on fish were used, and headsoff eviscerated weights were estimated using length-weight conversion tables of the IPHC. Age was determined, as described by Hardman and Southward (1965), from otoliths collected at the landing site when circumstances permitted and on all halibut taken by IPHC research vessels.

Before setting up sampling procedures, experiments were carried out to determine the uniformity of distribution of mercury in the muscle of individual fish. No significant differences in concentration of mercury (deviation did not exceed  $\pm 0.03$  ppm) were noted in muscle tissue taken from nape, midbody, or tail sections.

Analytical samples consisted of skinned and deboned edible muscle tissue that was normally taken from the nape section just behind the head. Some samples, however, were in the form of steaks and a few consisted of the entire fillets of small fish. Portions, usually about 400 g, taken from the

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FIGURE 1.-Mean mercury levels in Pacific halibut by area of catch.

nape section were ground in a Hobart grinder<sup>3</sup> equipped with a <sup>1</sup>/<sub>8</sub>-inch (3.2-mm) hole stainless steel plate. Larger steaks and fillets were ground in a Hobart Silent Food Cutter (Model 84181). The comminuted flesh was mixed thoroughly before subsampling for analysis. Because samples were often collected more rapidly than they could be analyzed, they were stored at -29°C until analysis. No change in mercury content was observed in halibut that were analyzed immediately or that had been held in frozen storage in either glass vials or aluminum containers if dehydration was prevented. A halibut sample stored in the above manner and used as an analytical control showed a mean mercury content of  $0.88 \pm 0.02$  ppm over a 2-yr period. This control was analyzed routinely to verify both accuracy and precision of the method.

Total mercury was determined at the PURC by either the method of Munns and Holland (1971) or Malaiyandi and Barrette (1970) as modified by Munns (1972). The former method uses sulfuric, nitric, and perchloric acids for digestion with sodium molybdate as a catalyst, while the Munns' modification utilizes nitric and sulfuric acids for digestion and vanadium pentoxide as a catalyst. Some samples were analyzed at the SEURC by the method of Hatch and Ott (1968) as modified by Uthe et al. (1970). This method uses sulfuric acid for digestion and potassium permanganate as an oxidizing agent. Final quantitation was by flameless spectroscopy using a Perkin-Elmer Model 403 Atomic Absorption Spectrophotometer at the PURC and by a Varian Techtron Model AA5 at the SEURC. In a collaborative study, the mean deviation between laboratories and methods did not exceed  $\pm 0.02$  ppm Hg. All samples were analyzed in duplicate or triplicate, depending upon the method of analysis used. We consider  $\pm 0.05$  ppm a significant deviation; therefore, when differences between replicates exceeded this level the samples were reanalyzed. Results are stated in parts per million wet weight.

## **RESULTS AND DISCUSSION**

A total of 1,227 halibut were analyzed for mercury content. Results indicated a relationship between mercury levels and area of catch, age, and size of fish. The results are broken down by the previously described catch areas (Figure 1). The fish taken from each area were separated by weight classes that approximate those used in the halibut industry; the low, high, and mean mercury values for each weight class are given with a frequency distribution of the fish by increasing mercury concentration (Tables 1 through 5). Because we thought that large fish would be more likely to exhibit higher concentrations of mercury, we attempted to obtain as many large fish as was practicable. For this reason our sampling contains a greater percentage of large fish than do the commercial catches from most of the areas dis-

<sup>&</sup>lt;sup>3</sup>Reference to trade names does not imply endorsement by the National Marine Fisheries Service, NOAA.

cussed here. Therefore, these data cannot be interpreted to indicate the approximate percentage of the commercial catch that is likely to contain mercury in concentrations over the guideline.

In 152 halibut taken from the Bering Sea, the mercury level in the muscle of 7 fish (5% of the sample) was over the guideline (Table 1). The incidence (percentage over the guideline of the total number of fish within a weight range) was highest among fish weighing more than 80 pounds.

Most of our samples, 761 fish, were taken from the Gulf of Alaska. We found that mercury in the muscle of 38 fish (5% of the sample) exceeded the guideline (Table 2). The highest incidence occurred in fish weighing more than 80 pounds. The weight ranges contributing most to the incidence were those of 126 to 150 pounds and those of more than 150 pounds. These two weight ranges contribute 21% and 32%, respectively, in contrast with only 3% in each of the weight ranges 81 to 100 pounds and 101 to 125 pounds.

The analytical data on 70 fish taken from south-

east Alaska area showed that mercury in the muscle of 9 fish (13% of the sample) was 0.50 ppm or higher (Table 3). The small number of fish in the larger weight ranges makes it impossible to be definitive, but it is reasonably clear that in this group, too, the incidence of mercury levels over the guideline was greatest among the largest fish.

Analyses on 163 fish from the British Columbia area showed that 44 of these (27% of the sample) were over the guideline (Table 4). In addition to this relatively high incidence, we saw for the first time the presence of significant numbers of highmercury-level fish in all weight groups, i.e., 10% of the fish were over the guideline in the 5- to 60-pound range, 75% in the 61- to 80-pound range, 73% in the 81- to 100-pound range, 100% in the 101to 125-pound range, and 67% in the 126- to 150pound range. We also saw that the concentration of mercury tended to increase with an increase in the incidence of fish that were over the guideline.

The analytical results on 81 fish taken from the Washington-Oregon area, the most southerly area of the range of the Pacific halibut, showed 29 fish

TABLE 1Mercury concentration in heads-off eviscerated Pacific halibut from the Bering Sea.	TABLE 1Mercur	y concentration in head	ls-off eviscerated Pac	ific halibut from t	he Bering Sea.
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Veight range	No.	Mercury (ppm) in edible muscle tissue											
Pounds (kg)	of fish	Low	High	Mean	< 0.25	0.25- 0.39	0.40- 0.49	- 0.50- 0.59	0.60- 0.69	0.70- 0.79	0.80- 0.89	0.90- 0.99	
			Number of tish,										
5-60 (2-27)	88	0.02	0.78	0.11	82	0	2	2	1	1	0	0	0
61-80 (28-36)	33	0.06	0.42	0.15	30	2	1	0	0	0	0	0	0
81-100 (37-45)	16	0.09	0.55	0.19	13	1	1	1	0	0	0	0	0
101-125 (46-57)	10	0.08	1.00	0.32	7	0	1	1	0	0	0	0	1
126-150 (57-68)	5	0.22	0.35	0.27	2	3	0	0	0	0	0	0	0
Total	152	0.02	1.00	0.15	134	6	5	4	1	1	0	0	1

TABLE 2.-Mercury concentration in heads-off eviscerated Pacific halibut from the Gulf of Alaska.

Weight range	No.			M	ercury (pp	om) in e	dible r	nuscle	tissue				
Pounds (kg)	of fish	Low	High	Mean	<0.25	0.25- 0.39	· 0.40 0.49	- 0.50- 0.59	- 0.60 <i>-</i> 0.69	0.70- 0.79	- 0.80- 0.89	0.90 0.99	-1.00- 1.49
				Number of tish									
5-60 (2-27)	378	0.01	0.50	0.11	371	4	2	1	0	0	0	0	0
61-80 (28-36)	92	0.05	0.47	0.18	77	13	2	0	0	0	0	0	0
81-100 (37-45)	76	0.05	1.10	0.25	49	15	10	1	0	0	0	0	1
101-125 (46-57)	92	0.03	0.74	0.29	37	36	16	2	0	1	0	0	0
126-150 (57-68)	67	0.12	1.28	0.38	19	23	11	6	3	3	0	1	1
Över 151 (68)	56	0.14	1.05	0.45	8	16	14	6	5	4	2	0	1
Total	761	0.01	1.28	0.20	561	107	55	16	8	8	2	1	3

Veight range Pounds (kg)	No.			м	ercury (pp	m) in e	dible r	nuscle	tissue				
	of fish	Low	High	Mean	<0.25	0.25- 0.39	0.40- 0.49	- 0.50 0.59	0.60 0.69	0.70- 0.79	· 0.80 0.89	0.90 0.99	-1.00- 1.49
		Number of fish											
5-60 (2-27)	33	0.04	0.34	0.12	30	3	0	0	0	0	0	0	0
61-80 (28-36)	10	0.09	1.30	0.33	7	1	1	0	0	0	0	0	1
81-100 (37-45)	9	0.09	0.59	0.28	4	4	0	1	0	0	0	0	0
101-125 (46-57)	13	0.22	0.95	0.46	1	6	1	3	0	0	1	1	0
126-150 (57-68)	3	0.26	0.36	0.31	0	3	0	0	0	0	0	0	0
Over 151 (68)	2	0.50	1.10	0.80	0	0	0	1	0	0	0	0	1
Total	70	0.04	1.30	0.26	42	17	2	5	0	0	1	1	2

TABLE 3.-Mercury concentration in heads-off eviscerated Pacific halibut from southeast Alaska.

TABLE 4.-Mercury concentration in heads-off eviscerated Pacific halibut from British Columbia.

Weight range Pounds (kg)	Weight range No. Mercury (ppm) in edible muscle tissue													
	of fish	Low	High	Mean	< 0.25	0.25- 0.39	0.40- 0.49	0.50- 0.59	0.60 0.69	0.70- 0.79	0.80- 0.89	0.90- 0.99	-1.00- 1.49	
					Number of tish									
5-60	122	0.04	1.04	0.19	99	7	4	5	3	2	0	1	1	
(2-27)														
61-80	20	0.12	1.23	0.69	2	2	1	1	4	3	2	з	2	
(28-36)														
81-100	11	0.10	1.22	0.66	1	2	0	1	2	2	0	0	3	
(37-45)														
101-125	7	0.50	1.46	0.96	0	0	0	2	0	1	0	1	3	
(46-57)														
126-150	3	0.25	0.77	0.52	0	1	0	1	0	1	0	0	0	
(57-68)														
Total	163	0.04	1.46	0.32	102	12	5	10	9	9	2	5	9	

TABLE 5.-Mercury concentration in heads-off eviscerated Pacific halibut from Washington-Oregon.

Veight range Pounds. (kg)	No.	Mercury (ppm) in edible muscle tissue											
	of	Low	High	Mean	< 0.25	0.25- 0.39	0.40- 0.49	0.50 0.59	0.60- 0.69	0.70- 0.79	0.80- 0.89	0.90- 0.99	
			Number of fish										
5-60 (2-27)	75	0.10	1.43	0.42	23	20	9	5	8	3	4	0	3
61-80 (28-36)	6	0.70	1.13	0.88	0	0	0	0	0	2	2	0	2
Total	81	0.10	1.43	0.45	23	20	9	5	8	5	6	0	5

(36% of the sample) were over the guideline (Table 5). None of these fish weighed more than 80 pounds, and only six weighed more than 60 pounds; 31% of the 5- to 60-pound fish and all of the 61- to 80-pound fish were over the guideline. In fish from this area, as in those from British Columbia, the concentrations of mercury increased with the incidence of fish over the guideline.

It is apparent that the mean level of mercury in the edible tissue and the incidence of fish over the guideline increases from the northern to the southern part of the range of the Pacific halibut (Figure 1, Table 6). There is also a relationship between the size of fish and the level of mercury in the muscle. Because of the sex-size relationship of halibut, i.e., males rarely exceed 80 pounds regardless of age, the correlation of mercury to age should be closer than that of mercury to size. However, age data were collected on only 76% of the total sampling, whereas weight was obtained on all samples. For this reason, and as a guide to industry, we have worked mostly with the mercury-size relationship. Evaluation of the data by regression analyses showed that the data are well described by the exponential function  $(y = ax^b)$ . Comparisons of the weights of halibut against

TABLE 6.-Summary of mercury concentration in Pacific halibut.

Number	Mean	weight	Me	Percent of samples exceeding		
fish	lb	kg	Low	High	Mean	0.50 ppm
152	54.6	24.8	0.02	1.00	0.15	4.6
761	71.8	32.6	0.01	1.28	0.20	5.0
70	67.6	30.7	0.04	1.30	0.26	12.8
163	39.3	17.8	0.04	1.46	0.32	27.0
81	30.3	13.8	0.10	1.43	0.45	35.8
	of fish 152 761 70 163	of Mean fish Ib 152 54.6 761 71.8 70 67.6 163 39.3	of fish Mean weight Ib kg   152 54.6 24.8   761 71.8 32.6   70 67.6 30.7   163 39.3 17.8	of fish Mean weight ib Me Low   152 54.6 24.8 0.02   761 71.8 32.6 0.01   70 67.6 30.7 0.04   163 39.3 17.8 0.04	of fish Mean weight ib Mercury (p Low Mercury (p High   152 54.6 24.8 0.02 1.00   761 71.8 32.6 0.01 1.28   70 67.6 30.7 0.04 1.30   163 39.3 17.8 0.04 1.46	of fish Mean weight Ib Mercury (ppm)   152 54.6 24.8 0.02 1.00 0.15   761 71.8 32.6 0.01 1.28 0.20   70 67.6 30.7 0.04 1.30 0.26   163 39.3 17.8 0.04 1.46 0.32



FIGURE 2.-Relationship between heads-off eviscerated weight and mercury concentration in the edible muscle tissue of Pacific halibut from the Bering Sea.



FIGURE 3.-Relationship between heads-off eviscerated weight and mercury concentration in the edible muscle tissue of Pacific halibut from the Gulf of Alaska.

mercury concentrations in the edible tissue for each area are shown in Figures 2 through 6. Correlation coefficients (r values) are shown on



FIGURE 4.—Relationship between heads-off eviscerated weight and mercury concentration in the edible muscle tissue of Pacific halibut from southeast Alaska.

each plot and are significant at the 0.1% level. Correlation coefficients between length and mercury were also significant at the 0.1% level within each area and were essentially identical to the correlation coefficients between weight and mercury. This would be expected from the weightlength relationship. Correlation between age and mercury was higher than between weight or length and mercury for fish from the Bering Sea, the Gulf of Alaska, and southeast Alaska; the same for fish from British Columbia; and lower for fish from Washington-Oregon. These correlation coefficients between age and mercury were also significant at the 0.1% level in all areas.

In evaluating the data, areas were used that are either the same as the fishery management areas defined by the International Pacific Halibut Commission (1974) or subdivisions of a management area. This was both logical and practical for the purpose of providing useful information to the halibut industry. The plots of mercury concentration in the edible muscle against weight of fish taken from both the Bering Sea and the Gulf of



FIGURE 5.-Relationship between heads-off eviscerated weight and mercury concentration in the edible muscle tissue of Pacific halibut from British Columbia.



FIGURE 6.-Relationship between heads-off eviscerated weight and mercury concentration in the edible muscle tissue of Pacific halibut from Washington-Oregon.

Alaska (Figures 2, 3) are so similar as to suggest that the environmental and biological factors that determine the rate and extent of deposition of mercury in the muscle are the same in both areas. In any case, the mean level of mercury and the incidence of fish exceeding the guideline increases, while the size of the fish decreases, from north to south. Increasing concentrations of mercury have been noted in other marine animals as one moves south from the Bering Sea. Anas (1974) pointed out that the harbor seal, *Phoca vitulina richardi*, which is a nonmigratory, inshore carnivore that feeds principally on fish, provides geographical information on local concentrations of contaminants. The livers of harbor seals taken from the Bering Sea contained lower levels of mercury than did those from Washington and Oregon, and those from southern California contained the highest levels. Sablefish, *Anoplopoma fimbria* (Pallas), also shows a similar pattern and will be the subject of another paper in this series.

These observations suggest that the total mercury contamination in the ocean environment (natural plus man-made) increases in a north-tosouth direction. Unfortunately, conclusive data to substantiate this hypothesis are not available. Eggerman and Mar (1972), in a review of the research that has been conducted on the various aspects of mercury transport, state that there is a paucity of available data, especially on the biological transport of mercury in marine waters.

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### LITERATURE CITED

- Anas, R. E.
  - 1974. Heavy metals in the northern fur seal, Callorhinus ursinus, and the harbor seal, Phoca vitulina richardi. Fish. Bull., U.S. 72:133-137.
- Eggerman, T., and B. Mar.
  - 1972. Mercury in the North Pacific-NORFISH NF01. Cent. Quant. Sci., Univ. Wash., Seattle, 15 p.

HARDMAN, W. H., AND G. M. SOUTHWARD.

1965. Sampling the commercial catch and use of calculated lengths in stock composition studies of Pacific halibut. Int. Pac. Halibut Comm. Rep. 37, 32 p.

HATCH, W. R., AND W. L. OTT.

1968. Determination of sub-microgram quantities of mercury by atomic absorption spectrophotometry. Anal. Chem. 40:2085-2087.

INTERNATIONAL PACIFIC HALIBUT COMMISSION.

1974. Pacific Halibut Fishery Regulations 1974. Int. Pac. Halibut Comm., Seattle, Wash., 5 p.

MALAIYANDI, M., AND J. P. BARRETTE.

1970. Determination of submicro quantities of mercury in biological materials. Anal. Lett. 3:579-584.

MUNNS, R. K.

1972. Mercury in fish by cold vapor AA using sulfuric-nitric acid/ $V_2O_5$  digestion. FDA (Food Drug Admin.) Lab. Inf. Bull. 1500, 8 p.

MUNNS, R. K. AND D. C. HOLLAND.

1971. Determination of mercury in fish by flameless atomic

absorption: A collaborative study. J. Assoc. Off. Anal. Chem. 54:202-205.

SCHMIDT, A. M.

1974. Action level for mercury in fish and shellfish. Fed. Regist., 39 (236) Part II: 42738-42740.

UTHE, J. F., F. A. J. ARNSTRONG, AND M. P. STAINTON.

1970. Mercury determination in fish samples by wet digestion and flameless atomic absorption spectrophotometry. J. Fish. Res. Board Can. 27:805-811.