U.S. DEPARTMENT OF COMMERCE
National Oceanic and Atmospheric Administration
NATIONAL MARINE FISHERIES SERVICE
Southwest Fisheries Center
Honolulu and La Jolla Laboratories
P. O. Box 3830
Honolulu, Hawaii 96812

STATUS REPORT

Mercury in the Pacific Blue Marlin, *Makaira nigricans*

By

Cynthia S. Unninayar and Bernard M. Ito

INTRODUCTION

In 1971, nine species of pelagic and inshore fish from Hawaiian waters were analyzed for total and organic mercury (Rivers, Pearson, and Shultz, 1972). In eight of these species the organic mercury was greater than 80% of the total; a finding consistent with other mercury values reported in the literature (Westöö, 1967; Kamps, Carr, and Miller, 1972). In the Pacific blue marlin, however, this predominance of organic mercury was not found, but rather most of the mercury seemed to be either inorganic or an organic form which was not extractable by the organic mercury method. Because of the uniqueness of the marlin, additional samples were collected in 1972, 1973, and 1974 during the Hawaiian International Billfish Tournament in Kailua-Kona, Hawaii, to continue study of the mercury levels in this fish. This report deals primarily with our findings of 46 marlin obtained in July 1974.

METHODS

Total, organic, and inorganic mercury were determined using the flameless atomic absorption apparatus with a Perkin-Elmer 303 Atomic Absorption Spectrophotometer. Total Mercury: The homogenized sample was digested with concentrated nitric acid over low heat and further oxidized with potassium permanganate solution; excess oxidizing agents were reduced with hydroxylamine solution; mercury ions were then reduced to elemental mercury with stannous chloride and vaporized into the flameless atomic absorption apparatus. (Rivers et al., 1972). Organic Mercury: The homogenized sample was extracted with benzene in a separatory funnel containing HCl, NaCl, and water; the benzene layer was then transferred to another separatory funnel and extracted with 1% cysteine solution; a portion of the cysteine solution was transferred to a flask and oxidized with permanganate and persulfate; following reduction with hydroxylamine and stannous ion, the mercury was vaporized into the flameless apparatus (Rivers et al., 1972). Inorganic Mercury: One gram of a 1:1 tissue:water homogenate was digested with 3 ml of concentrated nitric acid in a flask for 1 hour at room temperature then diluted to an appropriate volume with water, following reduction with stannous ion, the mercury was vaporized into the flameless apparatus (Iverson and Hierliky, 1974). Inorganic analyses were performed on samples of muscle and liver to confirm that the difference between total and organic was indeed inorganic mercury. The data are not reported here but does confirm that the residual mercury is inorganic in nature.

RESULTS AND DISCUSSION

Table 1 gives the results of total and organic mercury in muscle, liver, kidney, spleen, pyloric caecum, stomach, gill, gonad, and whole blood for 14 female (weighing 39-342 kg) and 32 males (weighing 58-112 kg) Pacific blue marlin. Total mercury levels were lowest in blood (0.01-0.61, $\bar{x}$ = 0.18 ppm) and highest in kidney (0.18-77.00, $\bar{x}$ = 26.33 ppm). In muscle the total values ranged from 0.09 to 10.00, $\bar{x}$ = 3.12 ppm. Organic mercury levels were lowest in blood (0.00-0.11, $\bar{x}$ = 0.04 ppm) and highest in muscle (0.02-1.02, $\bar{x}$ = 0.40 ppm). In all tissues the organic fraction was less than 1.02 ppm.

A general analysis of variance for the 1974 samples revealed significant differences between concentrations of total mercury in males and females and between body weight and tissues although these differences were not significant for organic mercury. Additional statistical analyses considered the two sexes separately for total mercury. Table 2 gives correlation coefficients between body weight and tissues. In most cases, the relationship is highly significant. The contrast of males and females shown in Figures 1 and 2 may be due to age and/or metabolic dissimilarities since females mature at a different rate than males. To see if this difference was the same for the previous years, an analysis of covariance was performed on all samples for muscle and liver beginning with 1971. The results indicated that there is no statistical dissimilarity between either males and females or between years. As was expected, however, there was a definite difference between muscle and liver tissues. In previous years, the number of females has been smaller than in 1974 perhaps resulting in a lack of a representative sample for comparison (Rivers et al., 1972; Shultz et al., in press).

The data clearly demonstrate that the organic mercury levels are quite low relative to the total mercury. Westöö (pers. comm.) has identified the organic fraction as methyl mercury in freeze-dried samples of the marlin. Both the Swedish Laboratory and the Tokyo Metropolitan Research Laboratory of Public Health (Nishigaki, pers. comm.) have confirmed the low percentages of organic mercury in samples collected in 1971 and 1974, respectively.

The ratio of total organic mercury for gonad, blood, gill, pyloric caecum, muscle, and stomach tissues range from about 4:1 to 11:1. The ratios in spleen, liver, and kidneys, however (38:1, 45:1, 120:1, respectively), reflect an unusually high proportion of inorganic to organic mercury in these organs. Many organisms have been reported capable of transforming methyl and other types of organic mercury to an inorganic form (Norseth and Clarkson, 1970a, 1970b; Takeda and Ukita, 1970; Burrows and Krenkel, 1973; Fang, 1973; Iverson and Hierliky, 1974). In most of these cases, the conversion was thought to occur or be associated with the kidney and liver.

Although the above demonstrations occurred under artificial laboratory conditions not necessarily applicable to the natural environment, they nevertheless offer convincing evidence that the conversion of methyl mercury to inorganic mercury in the Pacific blue marlin is an adaptive response to environmental mercury contamination.

LITERATURE CITED

Burrows, W. D., and P. A. Krenkel. Environ. Sci. Tech. 7, 1127 (1973).

Fang, S. C. Arch. Environ. Cont. Tox. 1, 18 (1973).

Iverson, F., and S. L. Hierliky. Cont. Tox. 11, 85 (1974).

Kamps, L. R., R. Carr, and H. Miller. Bull. Environ. Cont. Tox. 8, 273 (1972).

Norseth, T., and T. W. Clarkson. Arch. Environ. Health. 21, 717 (1970a).

Norseth, T., and T. W. Clarkson. Biochem. Pharmacol. 19, 2775 (1970b).

Rivers, J. B., J. E. Pearson, and C. D. Shultz. Bull. Environ. Cont. Tox. 8, 257 (1972).

Shultz, C. D., D. Crear, J. E. Pearson, and J. B. Rivers. Bull. Environ. Cont. Tox. (in press).

Takeda, T., and T. Ukita. Tox. Appl. Pharmacol. 17, 181 (1970).

Westöö, G. Acta Chem. Scand. 21, 1790 (1967).

February 21, 1975

Table 1.--Total and organic mercury in tissues of 46 Pacific blue marlin, Makaira nigricans (milligram/kilogram wet weight; mercury calculated as inorganic mercury).

WT KG	SEX	TOTL MUSC	ORGN MUSC	TOTL LIVR	ORGN LIVR	TOTL KIDN	ORGN KIDN	TOTL SPLN	ORGN SPLN	TOTL STOM	ORGN STOM	TOTL P.C.	ORGN P.C.	TOTL GILL	ORGN GILL	TOTL GOND	ORGN GOND	TOTL BLD	ORGN BLD
39	F	0.09	0.02	0.13	0.13	0.18	0.04	0.08	0.02	0.06	0.06	0.17	0.08	0.21	0.02	--	--	0.02	0.02
58	M	0.33	0.16	0.22	0.11	0.78	0.09	0.19	0.05	0.09	0.06	0.20	0.11	0.19	0.02	0.03	0.03	0.03	0.03
60	M	0.52	0.40	0.78	0.39	0.77	0.20	0.26	0.16	0.18	0.03	0.36	0.17	0.19	0.05	--	--	0.11	0.03
60	M	0.98	0.32	3.60	0.25	7.10	0.12	1.78	0.13	0.48	0.08	0.61	0.23	0.31	0.05	0.10	0.06	--	--
63	M	1.68	0.18	4.65	0.19	8.45	0.16	1.80	0.10	0.60	0.06	0.80	0.26	0.13	0.03	0.27	0.09	0.11	0.05
64	M	0.72	0.15	3.35	0.19	8.35	0.16	2.12	0.12	0.43	0.08	0.61	0.21	0.12	0.03	0.12	0.05	0.06	0.03
64	M	1.96	0.14	7.00	0.09	17.80	0.09	2.60	0.03	0.86	0.06	1.12	0.13	0.24	<0.01	0.22	0.03	0.03	0.02
68	M	1.32	0.24	5.44	0.13	10.20	0.10	0.94	0.02	0.50	0.06	1.02	0.12	0.14	0.02	0.13	0.03	0.06	0.02
68	M	0.36	0.12	2.25	0.10	4.60	0.07	0.88	0.10	0.36	0.04	0.42	0.13	0.08	0.01	--	--	0.08	<0.01
68	M	2.20	0.42	3.00	0.24	16.10	0.19	4.00	0.06	0.83	0.11	0.96	0.31	0.18	0.03	0.16	0.13	--	--
69	M	0.66	0.32	1.30	0.23	3.10	0.14	0.64	0.05	0.17	0.11	0.42	0.18	0.08	0.03	0.08	0.07	0.05	0.04
72	M	2.00	0.32	11.45	0.13	19.60	0.15	4.05	0.05	1.05	0.08	1.07	0.14	0.22	0.02	0.18	0.05	0.04	0.02
72	M	1.64	0.16	8.90	0.16	15.40	0.10	4.85	0.16	1.06	0.03	--	--	0.25	0.01	--	--	0.06	0.02
73	M	2.72	0.24	13.40	0.11	29.40	0.07	--	--	1.12	0.04	1.39	0.15	0.20	0.01	0.10	0.07	0.04	0.03
75	F	3.44	0.56	9.70	0.28	16.20	0.26	5.75	0.11	0.90	0.09	1.02	0.32	0.31	0.05	--	--	0.05	0.02
75	M	3.60	0.32	19.80	0.19	30.80	0.16	6.95	0.13	2.10	0.15	3.04	0.17	0.25	0.03	0.40	0.05	0.07	0.03
75	M	1.88	0.26	13.60	0.16	16.00	0.15	4.70	0.06	1.72	0.04	1.24	0.15	0.28	0.02	0.40	0.05	--	--
75	M	1.80	0.18	15.60	0.11	18.00	0.06	3.90	0.10	--	--	1.30	0.08	0.25	0.02	0.26	0.03	--	--
76	M	3.04	0.31	15.00	0.14	21.00	0.12	--	--	1.30	0.04	1.70	0.14	0.26	0.02	0.26	0.04	0.39	0.04
79	M	3.60	0.36	13.40	0.39	18.20	0.19	6.95	0.22	1.84	0.15	2.10	0.40	0.39	0.08	0.40	0.13	0.58	0.09
79	M	2.16	0.36	9.60	0.36	21.40	0.14	4.70	0.14	1.14	0.11	1.60	0.33	0.21	0.05	0.25	0.05	0.63	0.05
79	M	3.60	0.38	11.80	0.15	24.00	0.15	5.40	0.12	1.18	0.06	2.50	0.21	0.27	0.02	0.27	0.05	--	--
82	M	3.32	0.20	14.00	0.10	46.40	0.09	12.80	0.03	2.10	0.05	1.60	0.20	0.35	0.01	0.64	0.03	0.07	0.01
85	M	3.60	0.33	24.80	0.20	42.50	0.15	11.50	0.08	2.72	0.09	2.65	0.23	0.33	0.03	0.30	0.05	0.08	0.02
86	M	4.12	0.41	15.40	0.33	36.50	0.13	10.60	0.19	--	--	2.40	0.29	0.24	0.05	0.31	0.12	0.06	0.05
86	M	3.80	0.58	10.60	0.47	29.00	0.34	9.50	0.20	1.72	0.16	2.00	0.35	0.32	0.08	0.26	0.10	0.12	0.05
93	M	5.50	0.35	20.40	0.11	40.00	0.20	11.40	0.11	2.28	0.06	2.80	0.17	0.53	0.02	0.37	0.07	0.61	0.04
96	M	3.00	0.32	9.60	0.22	27.00	0.19	7.00	0.12	1.54	0.02	1.65	0.27	0.32	0.02	0.31	0.05	0.05	0.04
97	M	3.90	0.56	23.20	0.33	41.00	0.28	10.00	0.23	1.54	0.12	3.00	0.29	0.32	0.04	--	--	--	--
101	M	4.80	0.39	30.60	0.24	77.00	0.19	13.80	0.16	2.04	0.14	3.05	0.28	0.30	0.05	0.52	0.05	0.16	0.05
102	M	5.80	0.50	39.20	0.27	77.00	0.22	11.40	0.12	3.00	0.11	3.95	0.27	0.38	0.06	0.70	0.08	0.57	0.05
104	M	3.90	0.39	--	--	39.50	0.39	11.40	0.12	2.10	0.09	2.40	0.27	0.34	0.03	0.64	0.05	0.14	0.04
112	M	5.65	0.35	15.60	0.15	56.00	0.15	12.80	0.12	1.74	0.05	3.95	0.16	0.44	0.03	--	--	0.24	0.04
112	M	6.50	0.58	21.00	0.28	37.00	0.26	11.20	0.19	2.00	0.15	2.10	0.28	0.32	0.03	0.41	0.08	0.12	0.05
115	F	0.80	0.46	4.80	0.30	6.00	0.19	1.40	0.12	0.44	0.11	0.60	0.23	0.15	0.03	0.15	0.10	0.05	0.04
128	F	1.18	0.66	5.60	0.21	9.00	0.28	4.10	0.18	0.56	0.10	0.60	0.19	0.20	0.07	0.35	0.29	0.05	0.04
135	F	1.68	0.41	3.40	0.22	9.00	0.26	3.60	0.22	0.88	0.17	1.25	0.30	0.35	0.07	0.33	0.20	0.10	0.05
140	F	1.82	0.65	5.20	0.35	7.50	0.37	6.20	0.39	0.84	0.30	1.55	0.58	0.35	0.15	0.24	0.11	0.13	0.11
149	F	0.84	0.36	2.80	0.34	4.00	0.25	2.90	0.29	0.80	0.21	1.10	0.41	0.38	0.09	0.23	0.19	0.09	0.06
155	F	7.20	0.86	20.40	0.42	57.00	0.37	17.60	0.46	2.24	0.39	3.85	0.65	0.59	0.14	0.84	0.43	0.40	0.09
181	F	4.00	0.67	5.60	0.68	26.00	0.58	9.70	0.58	1.46	0.28	3.45	0.84	0.35	0.15	0.39	0.17	0.53	0.09
188	F	4.00	0.65	15.40	0.42	48.50	0.26	7.30	0.30	0.84	0.21	2.30	0.38	0.50	0.09	--	--	0.45	0.02
190	F	3.90	0.42	8.60	0.23	24.50	0.24	9.50	0.25	0.96	0.15	1.75	0.31	0.46	0.08	0.48	0.10	0.11	0.05
222	F	5.30	0.51	16.00	0.46	51.00	0.35	17.40	0.21	1.54	0.21	3.25	0.74	0.66	0.08	--	--	0.19	0.02
260	F	8.40	0.92	16.00	0.58	36.50	0.56	13.20	0.45	1.96	0.24	3.05	0.82	0.69	0.22	1.60	0.25	--	--
342	F	10.00	1.02	18.80	0.76	76.00	0.86	16.20	0.66	2.72	0.47	4.50	0.98	0.96	0.34	2.15	0.65	--	--
Mean 106		3.12	0.40	11.58	0.26	26.33	0.22	6.93	0.18	1.27	0.12	1.83	0.30	0.32	0.06	0.40	0.11	0.18	0.04

Table 2.--Correlation coefficients for body weight and tissue for males and females for total and organic mercury.

Muscle	Liver	Kidney	Spleen	Stomach	Pyloric caecum	Gill	Gonad	Blood
Body weight/male--Total mercury								
0.898**	0.765**	0.830**	0.884**	0.773**	0.828**	0.696**	0.751**	0.318
Body weight/female--Total mercury								
0.834**	0.694**	0.795**	0.759**	0.822**	0.820**	0.897**	0.947**	0.575**
Body weight/male and female--Organic mercury								
0.794**	0.762**	0.865**	0.837**	0.812**	0.859**	0.877**	0.823**	0.358*

*P<0.05
**P<0.01

Figure 1 Total and organic Hg in muscle vs Body weight

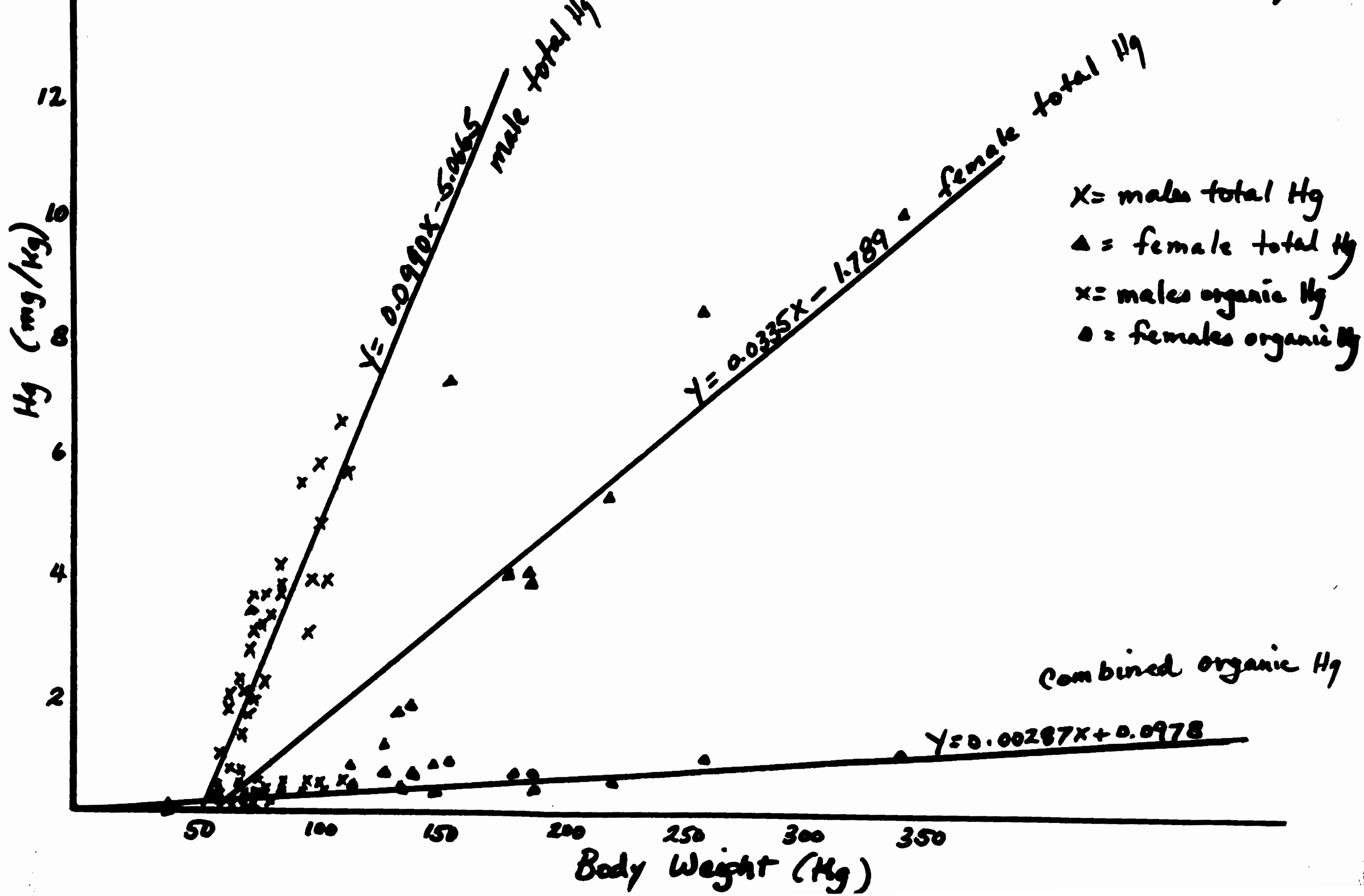

Figure 2 Total and organic Hg in liver vs Weight