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Changes in Mercury Levels in Harbor Porpoises from the Bay of Fundy, Canada, and Adjacent Waters During 1969-1977

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Abstract. Mercury levels were studied in a sample of 146 harbor porpoises taken in the Bay of Fundy, Canada, and adjacent waters during 1969-77. Mean concentrations of total mercury recorded in 5-year old males (exemplifying sexually mature adults) were 1.12 ppm in axial muscle, 0.38 in cerebrum, 0.31 in cerebellum, 2.00 in kidney, and 15.7 in liver. Respective means in tissues of 5-year old females were 1.46, 0.42, 0.21 2.77 and 16.2 ppm. There was a clear correlation of increase in total Hg with age in the tissues examined. Except in the liver, mercury levels were less well correlated with weight and body length than with age. Hg levels were not significantly correlated ($p > 0.05$) with weight and length *within* age classes, except marginally in the case of liver ($P \geqslant 0.10$). Hg in muscle was virtually all in the methylated form, but in liver only about 17% was methylated. The proportions in other tissues were intermediate (kidney about 41%, mammary gland about 46%, brain about 8 to 52%). Levels in liver were thought to be those giving the best indication of changes in background levels of mercury in the food chain. Significant changes in Hg tissue levels appear to have occurred during the 9-year study period. Liver Hg levels in both sexes decreased from 1970 to 1971, remained low for 3 years, increased again in 1974, and continued to do so in the years following. Data are presented suggesting that this also occurred in other tissues. Since there is no evidence of a change in diet in this period, we speculate that the decline in Hg levels could be correlated with a dominance of relatively mercury-poor Gulf Stream water in the Bay of Fundy approaches in 1971-73, as opposed to the usual dominance by waters of the Nova Scotia current.

The harbor porpoise *Phocoena phocoena* (L.) is a small toothed cetacean, which is an abundant and important component of the upper part of the North Atlantic inshore food web, feeding predominantly on clupeoid and gadoid fishes (Smith and Gaskin 1974). Porpoise meat is eaten in a number of east coast villages in Maine and New Brunswick. Furthermore, Holden (1972) suggested that the relatively long-lived marine mammals might be used as "in-

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dicator species'' to monitor accumulation levels of contaminants in the marine environment. For these reasons, information is required on the quantitative states of potentially toxic compounds in this species.

Some total mercury levels in selected tissues of harbor porpoises from the western North Atlantic were published in a preliminary report some years ago (Gaskin *et al.* 1972); specimens were only collected during a 2 1/2 year period, and interpretation of the results was limited further by our inability to determine the age of specimens at that time.

In this paper we present results from a much larger number of specimens of known age collected during the nine year period 1969-77. Epaxial muscle and liver were taken from the majority of specimens as field conditions permitted. Smaller numbers of samples of kidney, brain and other organs were also collected.

We were interested in total levels of mercury in the major organs of the body, the fraction of this which was in methylated form, and the variation of mercury levels with respect to body organ, sex, age, body weight and size, geographical location, and time of year, during the period 1969-77.

Materials

One hundred and forty six harbor porpoises (84♂, 62♀) were collected alive from fish traps or by shooting at sea during a population study in the Bay of Fundy and adjacent waters by the University of Guelph between June 1969 and September 1977. Because the population is migratory in habit, the specimens were taken between May and September in each year. All specimens appeared to be in good health, with no macroscopic pathological symptoms. All but 12 were taken in the approaches to the Bay of Fundy; the remainder were from Newfoundland (1), Prince Edward Island (1), the Atlantic coast of Nova Scotia (7), southern Maine (1), and southern Rhode Island (2). Canadian specimens were taken under permit from the Fisheries and Marine Service of Environment Canada; US specimens were collected prior to the Marine Mammals Act becoming law, with the assistance of the Department of Sea and Shore Fisheries in Boothbay Harbor, Me, and the Graduate School of Oceanography, University of Rhode Island. No permit was required to collect harbor porpoises in U.S. waters prior to 1973.

Length, sex and basic morphometric data were recorded for all specimens; body weight and axial muscle weight were recorded when circumstances permitted. Sex-specific length/weight regression equations were applied to unweighed specimens, as described by Gaskin *et al.* (1972). Mandibular teeth were taken from each animal; specimens were aged from dentinal growth layers following the method of Gaskin and Blair (1977).

Tissue samples were chilled at 10°C and subsequently frozen to −20°C as soon as field conditions permitted (2 to 8 hr). Muscle samples were taken from a standard site about 10 cm behind and above the right flipper. Liver samples were excised from the medio-lateral right lobe; kidney samples from the medio-lateral region of the right kidney. Samples of cerebrum were from the superficial medial region, including tissue from both hemispheres. Cerebellar samples were invariably whole-organ, because of its relatively small size.

Analytical Methods

Total Hg Determinations

One gram of tissue from each sample was solubilized with a 4:1 sulphuric-nitric acid mixture at 63-65°C, following the method outlined by Gaskin *et al.* (1972), after Hatch and Ott (1968).

Methyl Hg Determinations (Adapted From the Method of Westöö (1968)[3]

Two to five grams of sample were homogenized in 15 ml water, then mixed with 10 g NaCl, 14 ml concentrated HCl and a further 40 ml water. This mixture was then agitated for 5 min with 70 ml benzene, and centrifuged for 10 min at 2,000 r.p.m.; after which, 50 ml of the benzene layer was transferred to a 125 ml separatory funnel, mixed with 12 ml cysteine solution (prepared fresh each day), agitated for 2 min and let stand for 10 min. The aqueous layer was drained into a 15 ml centrifuge tube, and spun for 5 min at 2,000 r.p.m. This was repeated until the aqueous layer was clear, and no trace of an emulsion remained. By pipet, 8.5 ml of this layer was transferred to a 50 ml centrifuge tube, mixed with 3 ml 6N HCl and 20 ml benzene and agitated for two min. The aqueous phase was aspirated and discarded. Some anhydrous sodium sulfate was added, and the tube gently agitated. The dry benzene extract was used directly for analysis by gas chromatography (GLC); the final sample concentration was equal to 1/2 of the original sample weight in 20 ml of benzene. GLC conditions were as follows: A Varian 1400-H^3 detector; column: ⅛ × 5 in. heavy wall pyrex, packed with 6% Carbowax on Varaport 30; Oven temp. 140°C; Detector temp. 210°C; Carrier gas N_2 at 40 ml/min. Prior to sample analysis, the GLC column was conditioned by repeated injections of high concentrations of HgCl in benzene and CH_2HgCl standard solutions. Samples were quantitated against a CH_2HgCl standard curve, and a factor of 0.8 was applied to equate to CH_3Hg.

Statistical Treatments

The harbor porpoise is a relatively large mammal, living up to 13 years of age (Gaskin and Blair 1977), which has to be taken under special scientific permits. There is, therefore, a limit to the number of specimens which it is desirable (and practical) to collect. While the present sample is quite large by large mammal standards, division by sex, age class, date, and locality reduces sub-sample sizes considerably. Relatively small sample sizes render these data unsuitable for multiple regression analysis, since there are unavoidable data gaps. Body weights of many specimens could not be taken in the field, but only calculated indirectly from standard body lengths using the preliminary equations of Gaskin *et al.* (1972).

There is obvious convenience in being able to compare mean Hg levels of the different annual and regional sub-samples directly. Since Hg levels are known to vary with age in marine mammals, and the sub-samples are composed of specimens of different ages, this could only be done safely after determining that the age structures of the sub-samples were comparable statistically. Bailey (1959, p. 50) presented a modified t-test (variances unknown and not assumed equal) which seemed appropriate for the present data. To check that this test was giving acceptable results, the non-parametric Mann-Whitney Rank Sum Test (Sokal and Rohlf 1969, p. 391) was also applied independently to those pairs of samples which were suspected to be non-comparable. Significance levels calculated from both tests were similar in all cases, so it was concluded that Bailey's test could safely be used to test comparability of the different annual, regional, and male and female sub-samples. Since a complete range of tissues could not always be taken for each specimen, it was also necessary to check the comparability of age structure organ by organ. In virtually all cases we determined, despite some variation in mean values of ages, that age structures were comparable statistically; amalgamation of certain annual samples when necessary concelled remaining differences.

Results

Mean Levels of Total Mercury in Selected Tissues of Harbor Porpoises From the Bay of Fundy and Adjacent Regions

Table 1 shows mean concentrations of total mercury (parts per million, wet

[3] Differs principally from Westöö's method in having a higher centrifuging speed, and in some other relatively minor details (see Frank *et al.* 1974).

Table 1. Total mercury in major target organs of 61 male harbor porpoises taken near Deer Island, New Brunswick, Canada, 1969-77

Year	Mean age (years)	Mean length[a] (cm)	Mean concentrations of Hg (ppm) with standard deviation									
			n	muscle	n	liver	n	kidney	n	cerebrum	n	cerebellum
1969	5.7 ± 3.9[b]	134.0 ± 14.4[b]	8	0.97 ± 0.54	4	9.70 ± 9.70		—		—		—
1970	3.3 ± 2.7	133.5 ± 31.8	5	0.51 ± 0.18		—		—		—		—
1971	3.2 ± 2.6	129.5 ± 14.6	11	0.74 ± 0.33	6	5.63 ± 7.08	11	1.30 ± 0.86	4	0.39 ± 0.22	4	0.22 ± 0.10
1972	4.3 ± 2.2	134.0 ± 13.5	6	0.54 ± 0.17	6	5.30 ± 4.56	4	1.09 ± 0.24	5	0.26 ± 0.09	5	0.16 ± 0.04
1973	3.4 ± 3.4	134.0 ± 24.4	7	0.93 ± 0.58	2	0.82 ± 0.12		—	7	0.43 ± 0.34	7	0.41 ± 0.34
1974	3.2 ± 2.1	135.0 ± 15.5	2	0.77 ± 0.12	2	2.83 ± 0.46	2	0.94 ± 0.15	2	0.21 ± 0.04		—
1975	4.1 ± 2.6	132.5 ± 12.7	7	1.09 ± 0.50	7	8.33 ± 6.06		—		—		—
1976	4.3 ± 1.7	143.0 ± 10.5	6	0.88 ± 0.17	6	13.7 ± 8.59	3	2.40 ± 0.70	1	0.33		—
1977	4.5 ± 1.9	135.5 ± 14.7	8	1.69 ± 0.76	8	25.7 ± 14.8	3	4.71 ± 1.12	2	0.59 ± 0.24	2	0.48 ± 0.12

[a] Refers to largest sample (n) used; measured to nearest 0.5 cm
[b] Standard deviation

Table 2. Total mercury in major target organs of 43 female harbor porpoises taken near Deer Island, New Brunswick, Canada, 1969-1977

Year	Mean age (years)	Mean length[a] (cm)	Mean concentrations of Hg (ppm) with standard deviation									
			n	muscle	n	liver	n	kidney	n	cerebrum	n	cerebellum
1969	2.6 ± 1.7[b]	135.5 ± 15.9[b]	11	0.86 ± 0.56[b]	8	15.2 ± 30.8		—		—		—
1970	1.0 ± 0.9	101.5 ± 20.9	3	0.52 ± 0.48		—		—		—		—
1971	2.7 ± 1.9	131.0 ± 27.8	10	0.97 ± 0.66	3	40.1 ± 62.1	10	1.65 ± 1.32	5	0.33 ± 0.29[c]	4	0.18 ± 0.02[c]
1972	2.0 ± 1.8	127.0 ± 14.8	5	0.63 ± 0.51	4	10.2 ± 17.5	3	1.24 ± 1.00	5	0.52 ± 0.98	1	0.13
1973	3.1 ± 2.4	130.0 ± 26.9	6	1.17 ± 0.49	1	3.90		—	4	0.43 ± 0.14	3	0.34 ± 0.08
1974	—	—		—		—		—		—		—
1975	2.6 ± 2.1	136.0 ± 21.3	3	0.89 ± 0.43	3	5.39 ± 4.74		—		—		—
1976	2	129.5 ± 4.5	2	1.46 ± 0.66	1	8.33		2.02		—		—
1977	4.0 ± 1.4	146 ± 8.5	2	1.55 ± 0.29	1	30.3	4	3.15		—		—

[a] Refers to largest sample used (n): measured to nearest 0.5 cm
[b] Standard deviations
[c] One anomalous specimen with 9.05 ppm in cerebrum and 5.58 ppm in cerebellum omitted from this series

weight of pureed thawed tissue) in epaxial muscle, liver, kidney, cerebrum, and cerebellum of 61 male harbor porpoises taken in waters off Deer Island, New Brunswick, Canada, during the period 1969-77. Levels of mercury were considerably higher in livers (maximum of 40.4 ppm) than in other tissues. In kidney, a maximum mercury level of 5.55 ppm was noted, and in muscle a maximum of 2.53 ppm. Corresponding maxima for cerebrum and cerebellum were 1.10 ppm and 1.17 ppm, respectively. The apparent general (but not completely consistent) rise in total Hg levels between 1969-77 is discussed in a later section.

In Table 2 the equivalent data for 43 females taken in the same area are presented. Maximum Hg levels in muscle are 2.06 to 2.50 ppm, similar to the Deer Island region males, but the maximum in liver is much higher (112 ppm). Maximum kidney Hg is comparable (4.59 ppm) to that of males, as is that in cerebrum and cerebellum tissue, except that a single female taken in 1971 had 9.05 ppm in the cerebrum and 5.58 ppm in the cerebellum, vastly more than any other specimen. This same specimen contained the highest levels of liver and third highest kidney Hg.

Table 3 summarizes the mean concentrations of total mercury in the same tissues from the other seven localities from which specimens were collected in this study. Grand Manan Island is only about 40 km from Deer Island, and the results are closely comparable to those from the latter location. In other sections of this paper, they sometimes have been combined with those from Deer Island; no statistical anomaly is involved. Total Hg levels in tissues of animals taken off Digby, Nova Scotia were in the same ranges as those recorded for Deer Island specimens, as were those from the Atlantic coast of that province. Total muscle Hg in Digby, Nova Scotia specimens was significantly higher in females than males (calculated $f = 8$, $t = 2.04$, $P > 0.05$: also, $P > 0.05$ by Mann Whitney Rank Sum method). Since other localities are represented only by one or two specimens, detailed comparisons are precluded. Larger sample sizes might reveal significant differences, because of this, these data have not been amalgamated with those from Deer Island.

Limited numbers of additional samples were analyzed from male and female reproductive organs, milk, pancreas, and intestine; these data are presented in Table 4. Single observations of total mercury levels in stomach, spleen, heart, lung, and a 7 mm foetus are given in a footnote to Table 4.

Fraction of Total Hg Which was in the Form of Methyl Mercury

In 6 males and 8 females for which total Hg determinations had already been made, sub-samples were analyzed to determine the ratios of methyl to total mercury in major target organs. These results are summarized in Table 5, expressed both as numerical ratios, and as percentages. From 76 to 100% of mercury in muscle was present in the methylated form. The next highest levels were in mammary gland and kidney, 46.5 ± 0.5 and 41.7 ± 3.8%, respectively. The methylated fraction in cerebellar tissue was 5% and 52% in the two samples analyzed. The methylated fraction of mercury in the liver varied from 2 to 41%, but the mean value was relatively low, 17.3 ± 6.1%. Absolute liver values,

Table 3. Total mercury in major target organs of 41 harbor porpoises taken at other localities in eastern North America, 1970-1973

Sex	Locality, years	Mean age (years)	Mean length[a] (cm)	Mean concentrations of Hg (ppm) with standard deviation									
				n	muscle	n	liver	n	kidney	n	cerebrum	n	cerebellum
♂	Grand Manan I, New Brunswick, 1971-73	3.1 ± 1.7[b]	133.0 ± 9.1[b]	8	0.66 ± 0.25[b]	4	3.29 ± 3.30	4	1.14 ± 0.36	7	0.39 ± 0.26	7	0.28 ± 0.13
♀	Grand Manan I, New Brunswick, 1972-73	5.0 ± 2.6	155.0 ± 16.9	3	1.56 ± 0.86	3	11.3 ± 9.18	1	2.40	3	0.79 ± 0.36	2	0.53 ± 0.13
♂	Digby, Nova Scotia 1970-73	3.9 ± 2.9	130.0 ± 17.9	11	0.86 ± 0.56	8	3.96 ± 4.58	7	1.27 ± 0.90	8	0.25 ± 0.11		—
♀	Digby, Nova Scotia 1970-73	5.2 ± 2.7	152.0 ± 15.6	9	1.48 ± 0.69	4	30.7 ± 26.2	2	1.81 ± 0.97	7	1.42 ± 2.09	2	0.98 ± 0.72
2♂, 3♀	Atlantic coast of Nova Scotia 1970-72	4.6 ± 2.2	148.5 ± 13.7	5	0.99 ± 0.47	5	19.6 ± 1.99	4	1.49 ± 0.66	4	1.33 ± 1.74	3	0.41 ± 0.33
♀	Boothbay Harbor, Maine, 1971	1	103.5		—	1	1.13	1	0.68	1	0.34	1	0.23
♀	Naragansett Bay, R.I., 1971-72	1	118.5 ± 0.35	2	0.71 ± 0.60	1	2.54	1	2.74		—		—
♂	Souris, P.E.I.	5	147.5	1	1.12		—		—	1	0.47	1	0.36
♀	St. John's Newfoundland, 1973	0.5	95.0	1	0.25		—		—		—		—

[a] Refers to largest sample used (n); measured to nearest 0.5 cm

[b] Standard deviation

Table 4. Total mercury in selected visceral organs and milk and harbor porpoises from the Bay of Fundy and adjacent waters

Sex	Age class	Standard length	Collection code no.	Hg ppm in visceral organs and milk					
				Mammary gland	Milk	Uterus	Testis	Pancreas	Upper intestine
♂	0.5	86.0	8 A 73	—	—	—	0.20	—	—
♂	1	114.0	22 A 71	—	—	—	0.13	—	0.15
♂	1	123.0	6 A 71	—	—	—	0.17	—	—
♂	5	138.0	11 A 71	1.00	—	—	—	—	—
♂[a]	7	146.0	2 A 71	—	—	—	0.59	0.70	0.45
♀	0.5	90.0	7 A 73	0.26	—	—	—	—	—
♀	0.5	103.5	15 A 71	0.22	—	—	—	—	0.18
♀	1	110.0	23 A 71	0.18	—	—	—	0.13	0.14
♀	1	116.0	3 A 72	0.37	—	—	—	—	—
♀[b]	1	118.0	1 R1 71	—	—	0.47	—	0.80	—
♀	1	123.0	1 A 72	0.09	—	—	—	—	—
♀	1	132.0	13 A 71	0.36	—	—	—	—	—
♀	2	119.0	3 A 71	0.43	—	—	—	1.06	—
♀	3	127.0	12 A 71	1.00	—	—	—	—	—
♀	3	143.5	24 A 71	0.74	—	—	—	—	0.45
♀[c]	4	162.0	1 GM 72	1.33	—	—	—	—	—
♀	5	153.0	9 A 72	1.05	0.24	—	—	—	—
♀[d]	5	155.0	14 A 71	1.44	—	1.11	—	15.4	0.95
♀	5	157.0	9 A 71	1.66	—	—	—	—	—
♀	5	157.0	2 A 75	0.66	—	—	—	—	—
♀	5	163.0	10 A 73	0.58	—	—	—	—	—
♀	5	175.0	4 A 71	0.38	—	—	—	—	—
♀	6	160.5	1 C 71	0.61	—	—	—	—	—
♀	7	150.0	11 A 73	0.64	—	—	—	—	—
♀	7	161.0	2 C 72	0.83	—	—	—	—	—

[a] 1st stomach compartment 0.82, second 0.81, third 0.65
[b] Heart 0.79, Lung 0.98;
[c] 7 mm foetus 0.09
[d] Spleen 4.38

Table 5. Estimations of the fractions of methyl mercury of total mercury in tissues of harbor porpoises from the Bay of Fundy, Canada and adjacent waters

				Ratios of methyl to total mercury, and percentage of methyl Hg, in six tissues											
Sex	Age class (yr)	Standard length (cm)	Date captured	Ratio in muscle	%	Ratio in liver	%	Ratio in kidney	%	Ratio in cerebrum	%	Ratio in cerebellum	%	Ratio in mammary gland	%
♂	1	123.0	28 July		—	0.56/1.89	30	—	—	—	—	—	—	—	—
♂	2	116.5	5 July			0.32/2.24	14	0.69/1.28	54	—	—	—	—	—	—
♂	4	140.0	21 Aug.			—	—	0.62/2.20	28	—	—	—	—	—	—
♂	5	138.0	17 Aug.	0.99/1.02	97	—	—	0.69/2.20	31	—	—	—	—	—	—
♂	7	132.0	28 July	1.03/1.06	97	—	—	0.77/1.65	47	—	—	—	—	—	—
♂	7	146.0	6 July	1.18/1.18	100	1.68/18.3	9	0.83/1.73	48	—	—	—	—	—	—
♀	1	118.0	30 May	0.86/1.13	76	—	—	—	—	—	—	—	—	—	—
♀	2	119.0	21 July	—	—	—	—	0.81/1.67	48	—	—	—	—	—	—
♀	3	127.0	17 Aug.	—	—	—	—	0.76/2.90	26	—	—	—	—	0.41/1.00	47
♀	5	155.0	21 Aug.	1.93/2.50	77	2.8 /112	2.5	1.16/4.59	25	0.75/9.05	8	0.26/5.58	5	0.66/1.44	46
♀	5	157.0	10 Aug.	1.29/1.34	96	—	—	0.76/1.66	46	—	—	—	—	—	—
♀	5	175.0	22 July	0.93/1.11	84	1.60/3.86	41	0.88/2.07	43	—	—	—	—	—	—
♀	6	160.5	2 July	0.84/1.08	78	—	—	0.53/1.70	31	—	—	—	—	—	—
♀	8	161.0	2 July	2.18/2.18	100	4.72/63.9	7	—	—	0.81/5.99	13	0.77/1.49	52	—	—
Mean percentages of methyl mercury in tissues, based on all samples				89.4 ± 3.5		17.3 ± 6.1		41.7 ± 3.8		10.5 ± 2.5		28.5 ± 23.5		46.5 ± 0.5	

however, were frequently much greater than in muscle, because total mercury levels were far higher. The maximum CH_3Hg level was 4.72 ppm in the liver of an 8-year old female (2.18 ppm in muscle of the same specimen, and 1.16 ppm in kidney from a 5-year old female). The methylated fractions found in the two cerebral samples were among the lowest, 8 to 13%.

Possible Differences in Total Hg Levels With Sex and Age

Total Hg levels in muscle, kidney, cerebrum and cerebellum were compared in male and female samples for different years and various combinations of years. No significant differences in muscle levels were found between Deer Island, Grand Manan and Atlantic coast of Nova Scotia males and females (P values all >0.10). That females from Digby had higher muscle levels than males was indicated in the first section of Results. Such comparisons were difficult in the case of other tissues because of a) the large variance of total Hg levels in liver, and b) the relatively small number of samples from other organs (Table 4). In the one case where numbers were large enough to permit between-sex testing, no significance difference was found (for cerebellum of ♂ and ♀, Deer Island 1971-73 combined; calculated $f = 20$, $t = 1.05$, $P > 0.20$). Mean total Hg levels by age class for epaxial muscle, liver, kidney, cerebrum and cerebellum are presented in Tables 6 and 7 for male and female animals taken in southern New Brunswick waters, 1969-77. Possible year-to-year variation is not considered in these tables[4] (see also later). The relationships between total Hg levels in males and females with increasing age are presented graphically in Figures 1 through 4. For muscle, the fitted regressions were:

$$♂: y = 0.38 + 0.17\,x \quad (b = 0.173 \pm 0.04;\ t = 4.320,\ p < 0.001)$$
$$♀: y = 0.29 + 0.23\,x \quad (b = 0.230 \pm 0.03;\ t = 7.660,\ p < 0.001)$$

Where y = total mercury (ppm), x is age in years, b is the coefficient of slope, and t is the unit deviation measurement.

The two regression slopes were compared, using the method of Bailey (1959, p. 97-99); the comparison yielded a unit deviation of 1.78; indicating that while the regressions were not significantly different at the 0.05 level, they approached significance at the 0.10 level. Had larger sample sizes been available, the 0.05 level might have been attained. The possible *biological* significance of this result should not be ignored (Gilbert 1973, p. 63-64), and consequently data for males and females have not been amalgamated in this paper unless it is absolutely unavoidable.

Fitted regressions for liver were:

$$♂: y = -2.13 + 4.03\,x \quad (b = 4.03 \pm 0.92,\ d = 4.38,\ p < 0.001)$$
$$♀: y = -1.81 + 3.70\,x \quad (b = 3.70 \pm 1.07,\ d = 3.46,\ p < 0.001)$$

[4] Since the age structures are almost all statistically comparable within narrow limits, the effect of this amalgamation is to potentially increase the variance at each value, but not to affect the slope as long as the annual samples are comparable.

Table 6. Mean levels of total mercury in five major target organs of male harbor porpoises from southern New Brunswick waters, presented by age classes, summed for 1969-77 (ignoring possible year to year variation)

	Muscle		Liver		Kidney		Cerebrum		Cerebellum	
Age class	n	ppm	n	ppm	n	ppm	n	ppm	n	ppm
0.5	5	0.40 ± 0.06	3	0.75 ± 0.08	1	0.48	3	0.25 ± 0.01	3	0.23 ± 0.03
1	15	0.56 ± 0.09	9	2.29 ± 0.84	5	0.73 ± 0.17	4	0.17 ± 0.02	5	0.23 ± 0.05
2	5	0.68 ± 0.07	4	2.22 ± 0.19	4	1.00 ± 0.10	4	0.24 ± 0.02	3	0.15 ± 0.03
3	3	0.61 ± 0.08	2	7.16 ± 0.34		—	1	0.42	1	0.40
4	5	1.27 ± 0.30	4	17.1 ± 7.80	4	2.11 ± 0.50	2	0.59 ± 0.07	1	0.30
5	17	1.12 ± 0.15	11	15.7 ± 3.79	6	2.00 ± 0.66	7	0.38 ± 0.10	5	0.31 ± 0.08
6	6	0.94 ± 0.18	6	16.4 ± 5.41	3	1.91 ± 0.63	4	0.42 ± 0.12	4	0.28 ± 0.10
7	6	1.08 ± 0.12	3	13.5 ± 4.03	1	1.73	1	0.64	1	0.28
8	5	1.52 ± 0.32	2	17.2 ± 7.28	2	4.34 ± 1.21	2	0.79 ± 0.31	2	0.76 ± 0.42
13	1	0.83		—		—		—		—

Table 7. Mean levels of total mercury in five major target organs of female harbor porpoises from southern New Brunswick waters, presented by age classes, summed for 1969-77 (ignoring possible year to year variation)

	Muscle		Liver		Kidney		Cerebrum		Cerebellum	
Age class	n	ppm	n	ppm	n	ppm	n	ppm	n	ppm
0.5	4	0.35 ± 0.07	1	0.55	1	0.45	2	0.20 ± 0.02	2	0.20 ± 0.04
1	11	0.53 ± 0.06	6	2.83 ± 1.03	5	0.59 ± 0.10	4	0.18 ± 0.05	1	0.10
2	11	0.82 ± 0.14	6	4.85 ± 1.02	2	1.85 ± 0.18	3	0.34 ± 0.10	2	0.21 ± 0.08
3	5	1.14 ± 0.08	2	5.97 ± 2.08	2	2.22 ± 0.68	2	0.44 ± 0.05	1	0.38
4	1	1.28	1	4.10	1	2.40	1	0.89	1	0.43
5	11	1.46 ± 0.16	7	41.5 ± 16.3	5	2.77 ± 0.52	4	3.02 ± 2.03	2	2.99 ± 2.59
7	1	1.85		—		—		—		—
8	1	2.53	1	21.6		—	1	1.09	1	0.62

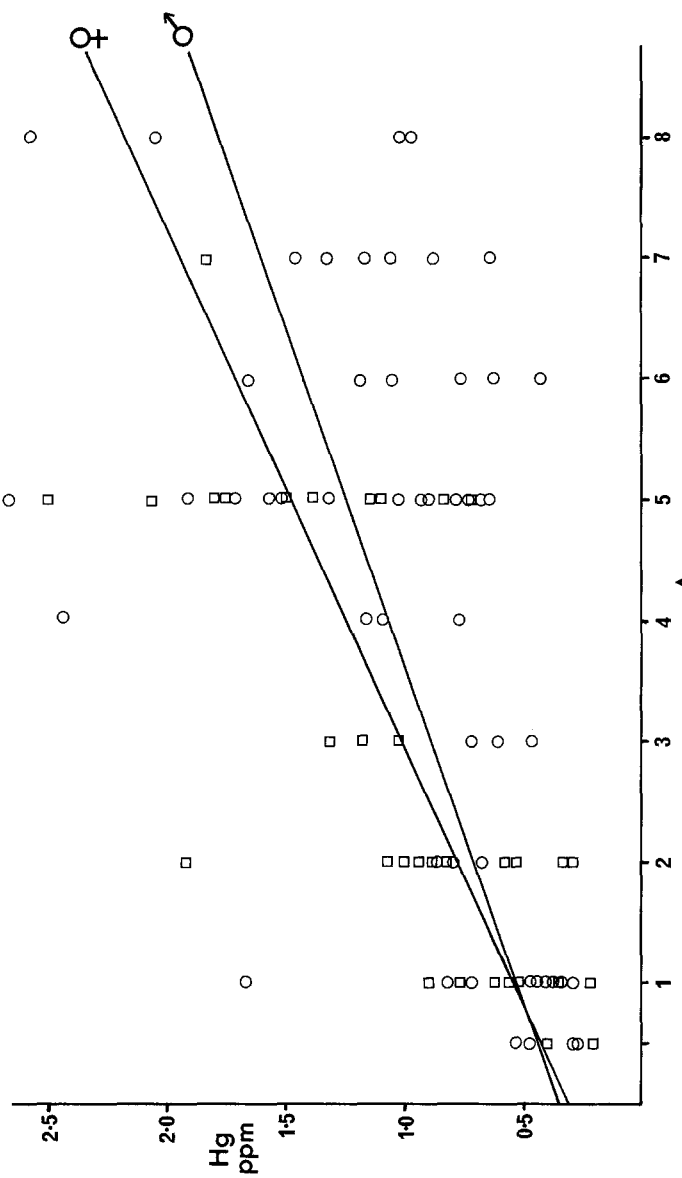

Fig. 1. Concentrations of total Hg in muscle (ppm in thawed tissue puree) and age in years of harbor porpoises in southern New Brunswick waters, 1969-77: Males (open circles), females (open squares). Respective regressions of Hg (y) on age (x) are described by $y = 0.38 + 0.17x$, and $y = 0.29 + 0.23x$

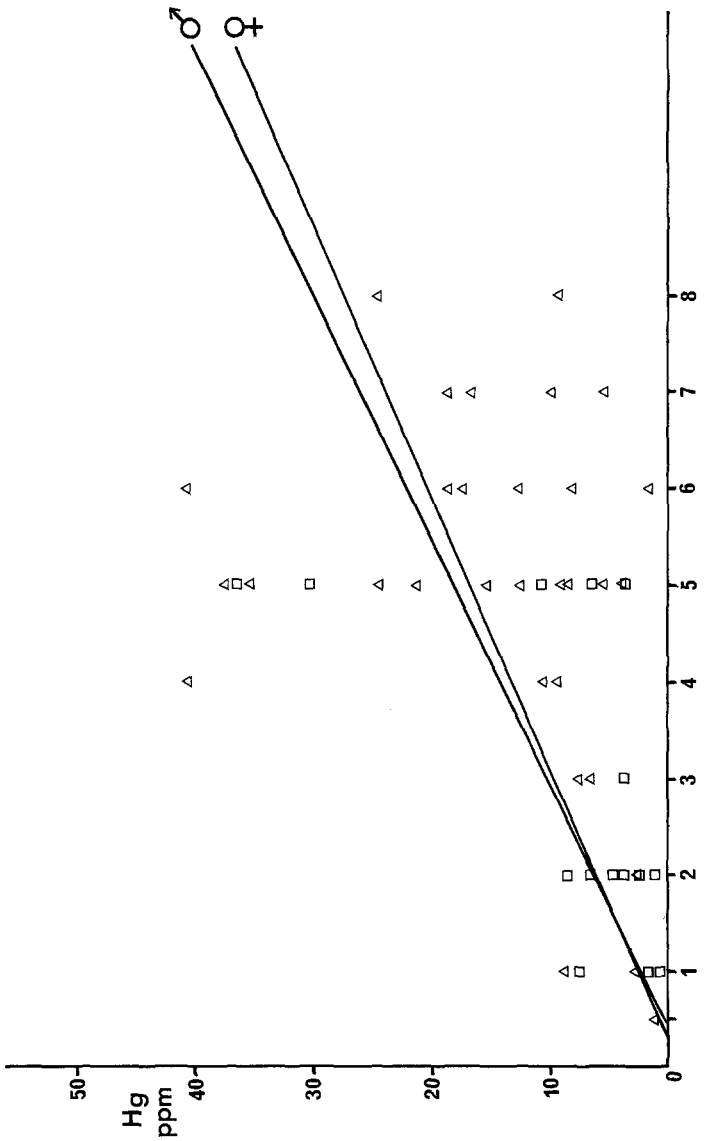

Fig. 2. Concentrations of total Hg in liver (ppm) and age in years of harbor porpoises in southern New Brunswick waters, 1969-77: Males (open triangles), females (open squares). Respective regressions of Hg (y) on age (x) are described by $y = -2.13 + 4.03x$, and $y = -1.81 + 3.70x$

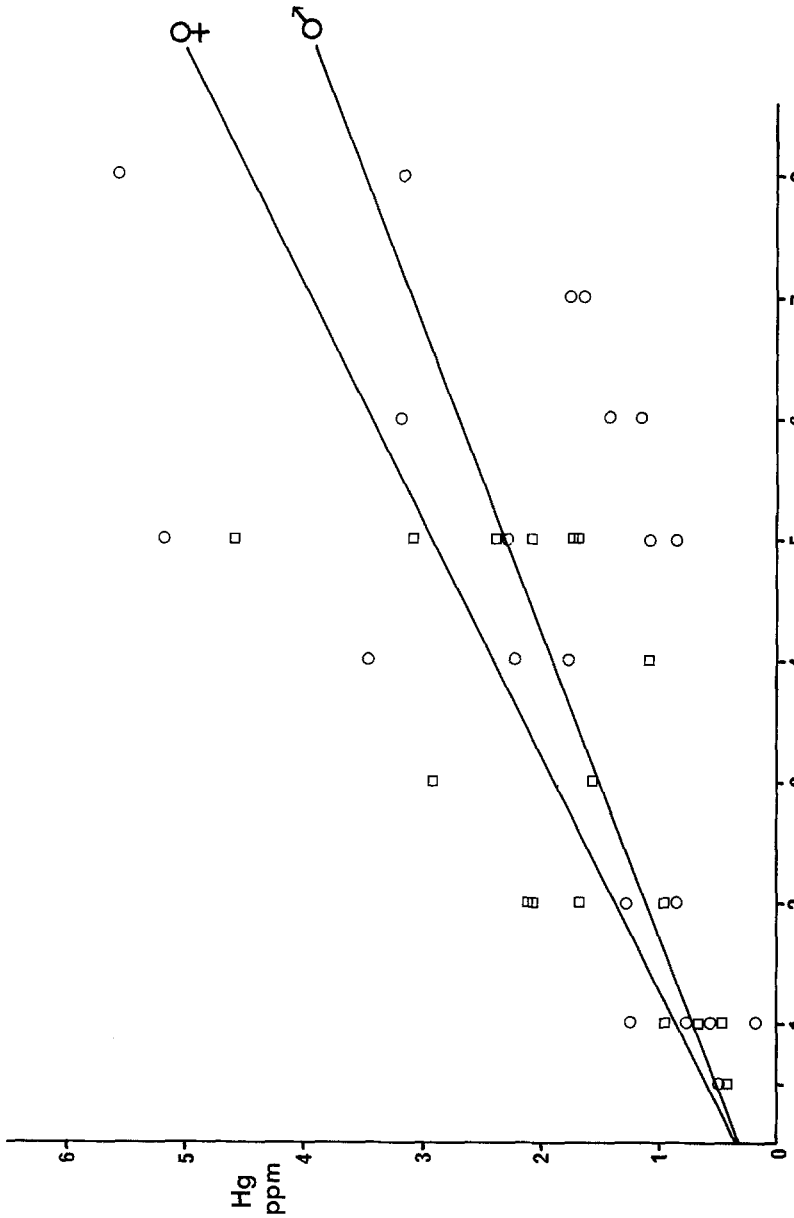

Fig. 3. Concentrations of total Hg in kidney (ppm) and age in years of harbor porpoises in southern New Brunswick waters, 1969-77: Males (open circles), females (open squares). Respective regressions of Hg (y) on age (x) are described by $y = 0.32 + 0.40x$, and $y = 0.34 + 0.51x$

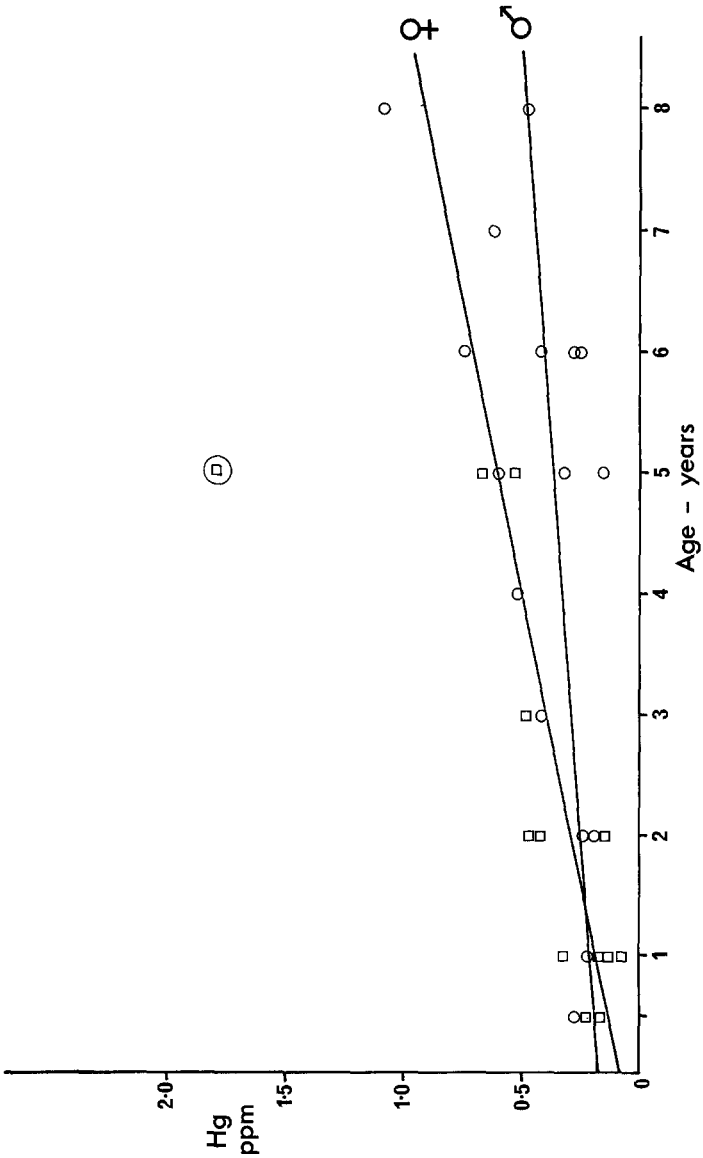

Fig. 4. Concentrations of total Hg in cerebrum (ppm) and age in years of harbor porpoises in southern New Brunswick waters, 1969-77: Males (open circles), females (open squares). Respective regressions of Hg (y) on age (x) are described by $y = 0.19 + 0.04x$, and $y = 0.12 + 0.10x$. Circled anomalous value not included in calculations (see Discussion)

Comparison of the regression slopes in this case (using the small sample techniques of Bailey (1959, p. 99) since both n values <30), yielded a low t value of 0.209, indicating no significant difference, even at the 0.25 level.

Similar regressions were obtained for kidney and cerebrum of males and females (Fig. 3, 4: equations in the captions); the divergences of the slopes can be seen by inspection to be of the same magnitude as that for muscle.

These data show that total Hg levels increase with age in all organs examined in both sexes. Similar results (not shown) are obtained if annual samples are plotted individually.

Possible Influence on Total Hg of Body Weight and Size at Any Given Age

Lacking a method of determining the age of our specimens at the time of the preliminary study (Gaskin *et al.* 1972), we showed that total Hg (ppm) in muscle and liver increased with increase in body length (L), and presumably therefore with weight (W), since the relationship of W in lbs or Kg and L in cm can be described by the linear equations

$$\text{W lbs or } 0.45 \text{ Kg} = -93.4 + 1.3 \text{ L for males,}$$
$$\text{and W lbs or } 0.45 \text{ Kg} = -80.7 + 1.3 \text{ L for females,}$$

in this species, which has slight but detectable sexual dimorphism (Møhl-Hansen 1954, Fisher and Harrison 1970). These equations describe the normal linear relationships: a study in progress (Yasui 1978), is attempting to generate slightly more complex equations which will take into account animals at the extremes of distribution.

Increments of length and weight become nearly asymptotic in this species at about 5 to 6 years of age (Gaskin and Blair 1977). The relationship between age (A) and body length (L) is relatively complex:

$$A = [L/(-1.30 \text{ L} + 209.35)] - 1 \text{ for males,}$$
$$\text{and } A = [L/(-0.84 \text{ L} + 156.15)] - 1 \text{ for females.}$$

Aging is best done from dentinal layers, since variation in both length (and hence weight) is considerable, especially in older age groups. One would therefore expect total Hg levels, if accumulating with age, to show a correlation also with weight and body length, but with increasing variance in the older year classes. This appears to be reflected in our data (see Tables 6 & 7).

Actual weights were available for only a relatively few specimens (26 in all, primarily from 1971 and 1972), because of the difficulty of using any form of scale or balance on a small boat (specimens were usually worked up at sea to obtain tissues for several projects where short post mortem periods were essential). Ignoring the relatively small differences between the sexes, and the possible influence of mixing data from two to three different years or from different times in the same year (see later) we examined the correlations between total Hg concentration in ppm in muscle, liver, kidney, and cerebrum, against age, weight, and length, respectively, in those specimens for which accurate body weights were known; these are summarized in Table 8. Our data indicate that in cerebrum (albeit with the reservation that this was the smallest

Table 8. Relationship of total mercury (ppm) in major target organs of harbor porpoises from southern New Brunswick, 1969-77, to age (in years), body weight (kg), and standard body length (cm)

Tissue	f(= n−2)	Parameter (x)	Calculated r value	Least squares fit against ΣHg (y)	Estimated p value
	26	Age	0.73	0.28 + 0.20 x	$p < 0.001$
Muscle	26	Weight	0.66	0.33 + 0.03 x	$p < 0.001$
	26	Length	0.57	−1.38 + 0.02 x	$0.01 < p < 0.001$
	7	Age	0.75	0.15 + 0.06 x	$0.02 > p > 0.01$
Cerebrum	7	Weight	0.44	Not significant	$p > 0.20$
	7	Length	0.48	Not significant	$p > 0.20$
	15	Age	0.71[a]	3.36 + 3.40 x	$0.01 > p > 0.001$
Liver	15	Weight	0.82	−25.45 + 0.93 x	$p < 0.001$
	15	Length	0.71[b]	−54.3 + 0.50 x	$0.01 > p > 0.001$
	14	Age	0.71	0.43 + 0.48 x	$0.01 > p > 0.001$
Kidney	14	Weight	0.67	−1.64 + 0.09 x	$0.01 > p > 0.001$
	14	Length	0.46	Not significant	$p \geq 0.10$

[a] 0.711 when calculated to three decimal places
[b] 0.714 when calculated to three decimal places

sample size) total Hg concentrations were significantly correlated with age only (with very low correlations with weight and body length). For muscle, significant correlations with all three parameters were found, but the highest correlation was with age. For kidney, the highest correlation was again with age, but only slightly more so than with body weight. Correlation of mercury concentration in kidney with body length was only statistically significant when plotted logarithmically (not shown). In the case of liver, on the other hand, the highest correlation of concentration levels was with body weight, then body length. The lowest (although still highly significant) correlation was with age.

Comparison of Total Hg Levels in Different Organs

We compared the levels of total Hg in muscle and liver (and for mature males, liver and kidney) within samples of yearling and mature porpoises. To increase the sample size in this case, all specimens which had reached sexual maturity (i.e. those of 4+ years of age) were considered instead of just 5 year-olds. The results, males and females treated separately, are summarized in Table 9[5]. Statistically significant relationships were found between total Hg levels in muscle and liver, and between kidney and liver. If total Hg was relatively high in one of these tissues, it could be reasonably expected to be high in the other. The correlation was much higher, however, in males than females in both age

[5] In this case, males and females were segregated, since we had evidence to suspect different mobility of Hg in breeding females and, therefore, possible differences in organ:organ ratios from males.

Table 9. Relationship between total mercury levels in different organs of yearling (1-year-old) and sexually mature (4+ years old) harbor porpoises from southern New Brunswick waters, 1969-77

Category	f(= n−2)	Tissues compared (y, x)	Calculated r value	Least squares fit (a + bx)	Estimated p value
Yearling ♂	8	liver & muscle	0.92	−0.99 + 5.27 x	$p < 0.001$
Yearling ♀	5	liver & muscle	0.67	−1.85 + 7.51 x	$p \geq 0.05$
Mature ♂	23	liver & muscle	0.64	2.61 + 11.9 x	$0.01 > p > 0.001$
Mature ♀	10	liver & muscle	0.53	−34.1 + 39.42 x	$p \geq 0.05$
Mature ♂	11	liver & kidney	0.61	−5.5 + 11.3 x	$0.05 > p > 0.02$

groups. No significant differences were found between total mercury levels in cerebrum and cerebellum samples of 23 Deer Island specimens (f = 21, t = 0.888, P = 0.20 for both modified t test and Mann-Whitney Rank Sum Test).

Possible Changes in Total Hg Levels in Tissues During the Year

Since there is so much variation of total Hg with age, we decided that possible changes in mercury levels during the year could only be detected by a study of animals of the best represented age classes. The examination was further complicated by all 5-year olds having been taken between the beginning of the third week in July, and the end of the first week of September, while five of the yearlings were taken earlier in the season. Weather conditions and the nature of the annual migration make it virtually impossible to obtain a significant number of specimens before or after these times.

Samples containing 24 yearlings and 27 five-year olds from southern New Brunswick were examined. Plotting total muscle, liver and kidney Hg in ppm for both against a linear time scale (in weeks), revealed no trends even when reproductively active females were plotted separately. Week by week means for total Hg all lay within 0.75 of one standard deviation of the overall mean in both age classes. While not strictly appropriate statistically, least squares fits gave some idea of association in such cases; three test r values calculated were very low, +.24 (=27) for muscle of 5-year olds, −.36 (=17) for kidney of yearlings, and =.04 (=32) for liver of 5-year olds. We concluded that there was no evidence of a change in kidney or liver mercury levels during the 10 week period in which most of our specimens were collected.

Possible Annual or Long-Term Changes in Total Mercury Levels in Tissues of Harbor Porpoises from the Bay of Fundy Between 1969 and 1977

The data presented in Tables 1 and 2 give the impression of a general, but not completely consistent, increase in average annual levels of total mercury in harbor porpoise tissues from 1973 to 1977. Mean levels in muscle, liver, kidney and cerebrum for all males and females are plotted in Figures 5 through 9. To further examine this aspect, we utilized all available individual data for males and females from the Deer Island region 1969-77 and calculated least square fits of

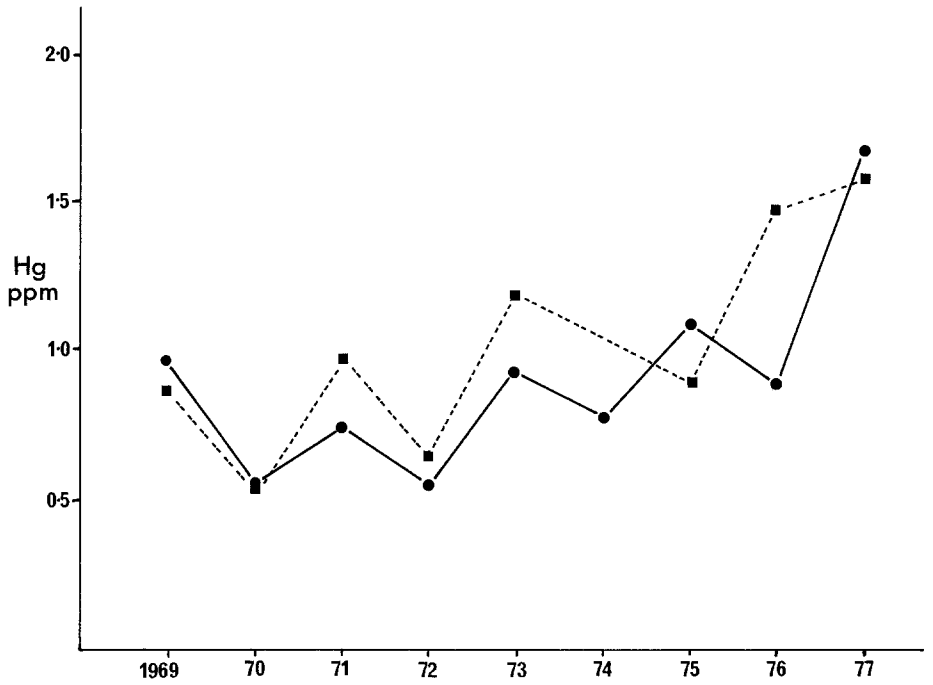

Fig. 5. Average levels of total Hg (ppm) in muscle of harbor porpoises from southern New Brunswick waters 1969-77: All sampled males (closed circles), all sampled females (closed squares). Respective regressions of Hg (y) on sequential years (x) are described by y = 0.50 + 0.082x and y = 0.53 + 0.097x

total mercury in muscle (♂ & ♀) and liver (♂ only) (data were insufficient in the case of kidney, cerebrum, cerebellum and liver of ♀♀) against chronological age. To reduce the effects of intra-year variation within the relatively small annual subsamples they were combined into threes and the sliding means were calculated, centered in turn on each year from 1970 to 1976. From the regressions so obtained (Table 10) we extracted the estimated mean values for 1- and 5-year old animals for each year, and compared these with actual levels recorded in 1- and 5-year old specimens and also the distribution of annual means of all males and all females.

Figure 5 displays graphically the mean total Hg levels in muscle of males and females from the Deer Island region. A statistically significant increase in mercury levels during the study period appeared when the data were plotted in this way. The fitted regressions were:

$$♂: y = 0.50 + 0.082\,x \ (b = 0.082 \pm 0.034;\ t = 2.412;\ 0.05 > p > 0.02)$$
$$♀: y = 0.53 + 0.097\,x \ (b = 0.097 \pm 0.030,\ t = 3.233;\ 0.02 > p > 0.01)$$

where y = total mercury (ppm), x is age in years, b is the coefficient of slope, and t is the unit deviation measurement.

In Figures 6 and 7, the results for only 1- and 5-year olds are plotted separately for each sex. Trends for the period are however, quite different in

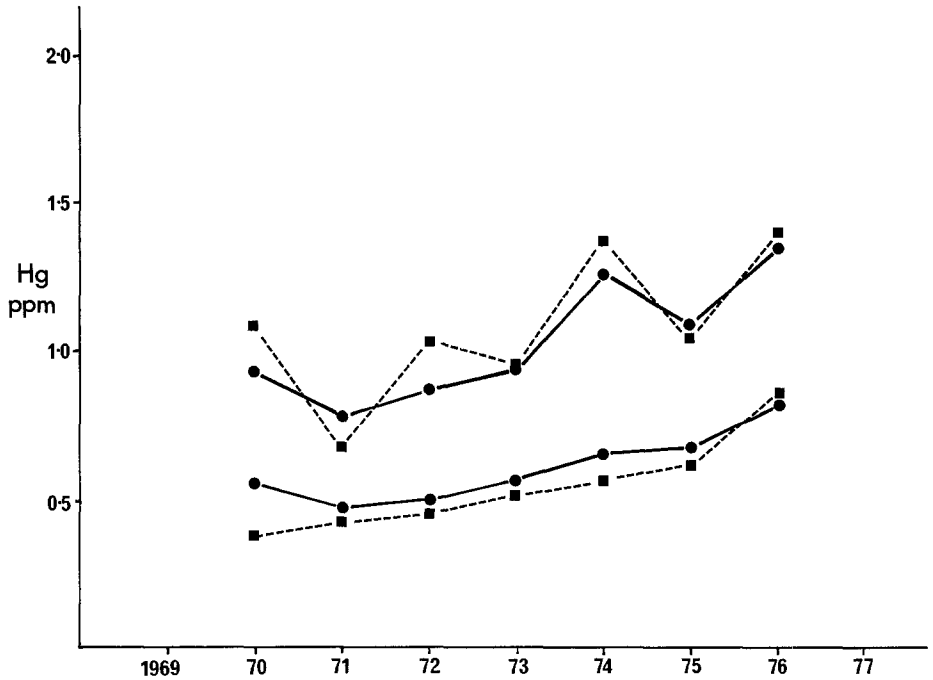

Fig. 6. Average levels of total Hg (ppm) in muscle of male harbor porpoises from southern New Brunswick waters 1969-77. Upper group: 5-year olds, values derived from least squares fits (closed circles), and from 3-year sliding average calculations based on actual 5-year old specimens (closed squares). Lower group: Similarly derived data (closed circles and closed squares respectively) for one year old males

the sexually mature males and females, a phenomenon masked by plotting all data as in Figure 5.

Data for Hg levels in liver of 1- and 5-year old males are similarly treated, and compared with annual means calculated from all levels in all males and all females (Figure 8). The more limited results for levels in kidney and cerebrum are shown in Figure 9. In all cases, the pattern seems to be of relatively low levels in 1971-73 (perhaps resulting from an earlier decline), increasing sharply during 1974-76.

Discussion

The literature concerning mercury in the aquatic environment is very extensive (Taylor 1977), but information on levels, uptake, effects, and specific and regional variation in marine mammals is much more limited, largely because of the inherent difficulties of obtaining specimens in significant numbers. To the best of our knowledge, the present study is the first to examine mercury levels in the same marine mammal population over an extended period of time.

Initial studies during the late 1960's and early 1970's found significant

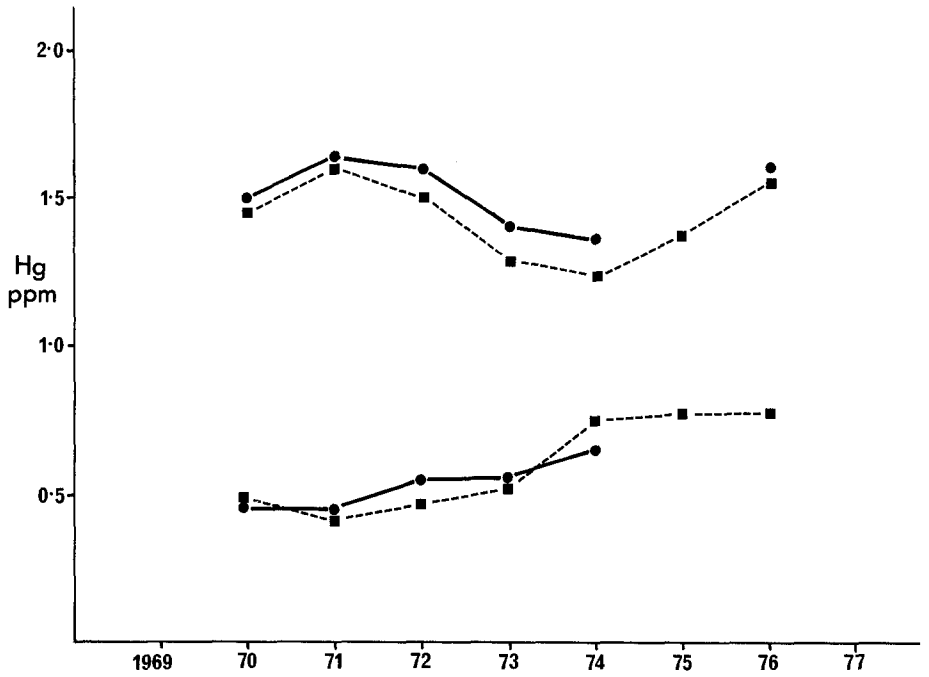

Fig. 7. Average levels of total Hg in muscle (ppm) of female harbor porpoises from southern New Brunswick waters 1969-77. Data derived and plotted as described in Figure 6

levels of mercury, particularly methyl mercury, in certain tissues (especially the liver) of northern hemisphere seals and porpoises (Helminen *et al.* 1968; Henriksson *et al.* 1969; Bligh and Armstrong 1971 and Gaskin *et al.* 1972, 1973). Some of these authors implied that the major source of such mercury was likely to be agricultural or industrial in origin. High mercury levels were also found in the tissues of some tropical Cetacea in the eastern Caribbean (Gaskin *et al.* 1974), but these authors concluded that this mercury was probably natural, and associated with the intense cyclic tectonic activity present in the southeastern Caribbean fringe. Preliminary studies on harbor seals, *Phoca vitulina,* indicated significantly higher levels in adult seals taken from ledges within southern New Brunswick waters than from the coast of southern Maine outside the Bay of Fundy (Gaskin *et al.* 1973), which suggested that regional differences could be recognized; the implications of this are discussed later.

Increase in total mercury levels in harbor porpoise with increase in body size (as illustrated by standard length) was demonstrated by Gaskin *et al.* (1972) using specimens in which age had not been determined (see Introduction). Studies by Olsson (1976) on northern pike, *Esox lucius* L., are of interest since he found that fish of the same size but of different ages (as a measure of exposure time) had similar mercury levels. He found also that males had significantly greater levels than females but that within any one sex . . . "the metabolic turnover (directly correlated with parameters of size) is more important than age or exposure time in determining mercury levels in fish."

We found clear correlations of mercury levels with chronological age in

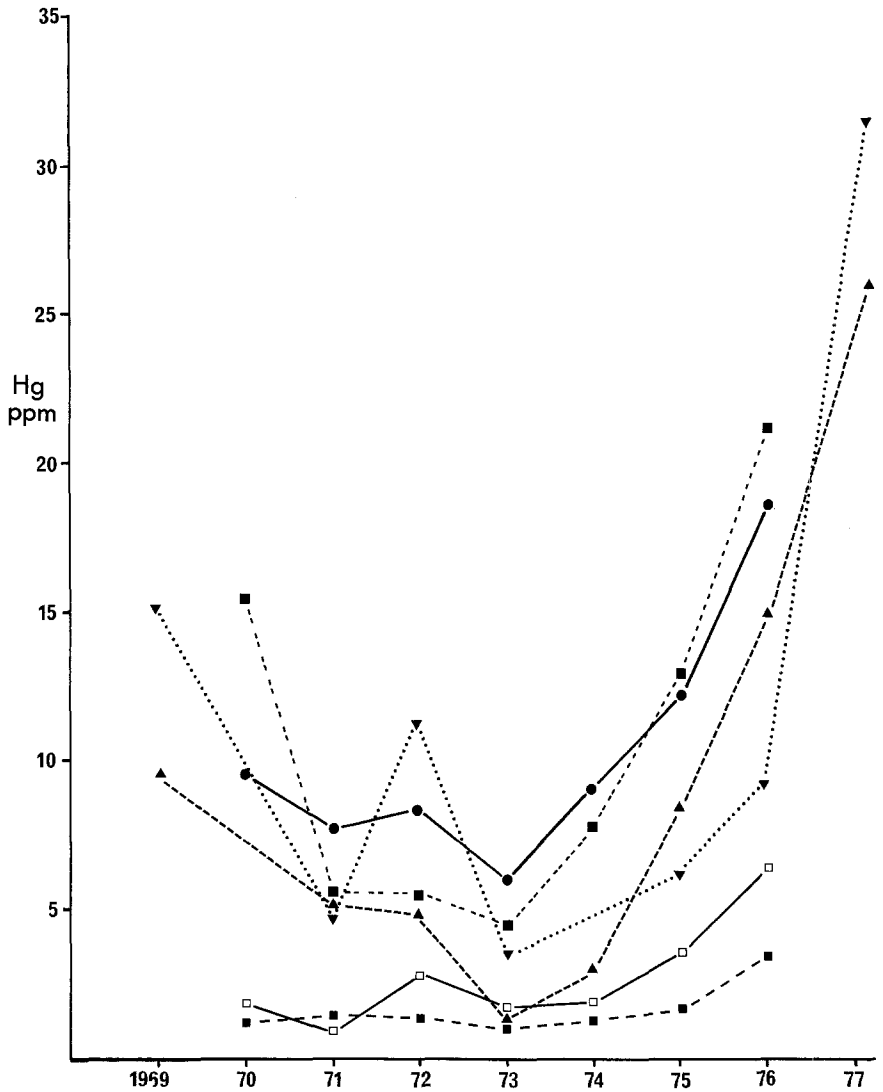

Fig. 8. Average levels of total Hg in liver (ppm) of harbor porpoises from southern New Brunswick waters 1969-77. Legend: Small closed squares—values for one-year old males based on 3-year sliding averages. Small open squares—values for one year old males based on least squares regressions. Large closed squares—values for 5-year old males based on 3-year sliding averages. Large closed circles—values for 5-year old males based on least squares regressions. Closed triangles and inverted closed triangles respectively—values for all males and all females in the sample, calculated through 3-year sliding averages

muscle, kidney, cerebrum, and liver of harbor porpoises. In the absence of experimental data on metabolic turnover of mercury in this species, we examined correlation coefficients obtained for increase of mercury levels against age, and compared them with those calculated for mercury against weight and length. Changes in mercury levels with weight and length were also examined

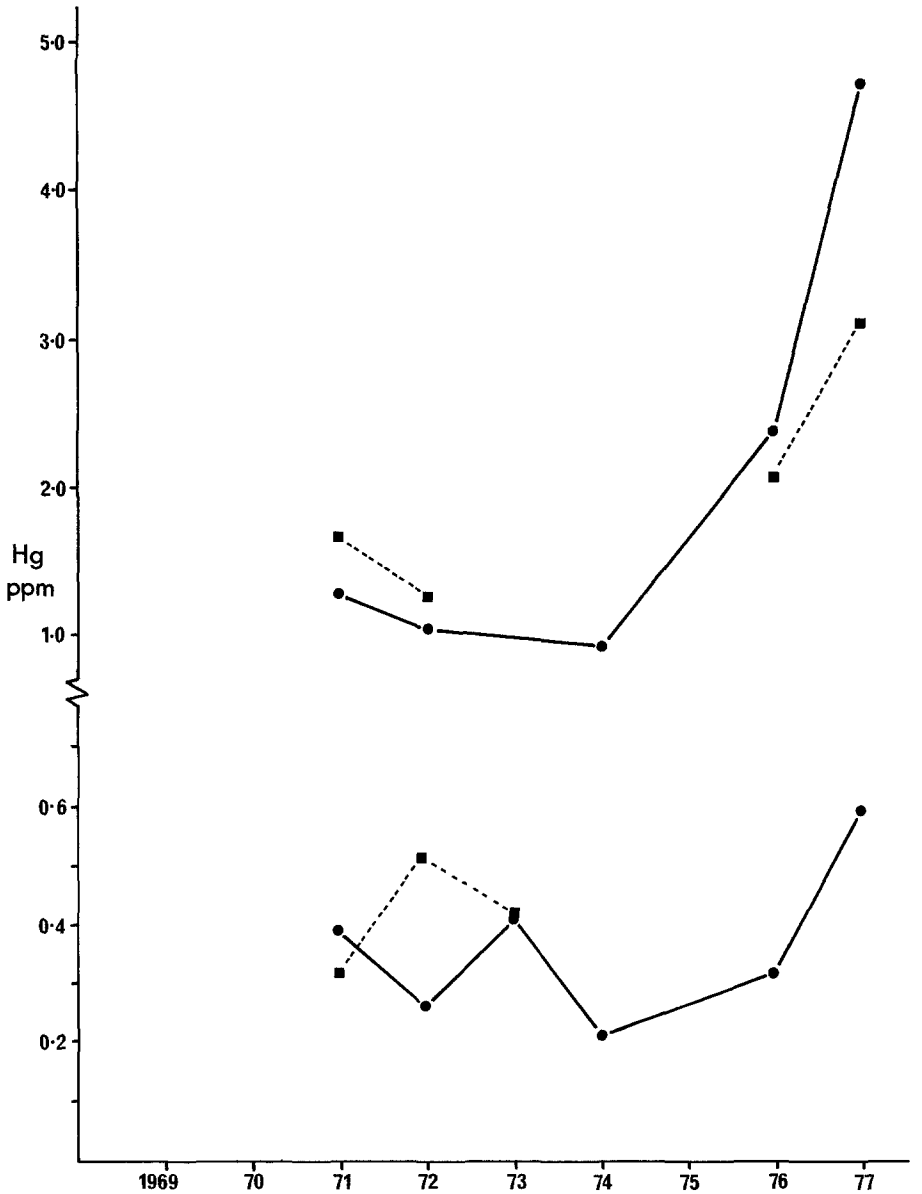

Fig. 9. Average levels of total Hg in kidney and cerebrum (ppm) of harbor porpoises from southern New Brunswick waters 1969-77. Upper group: levels in kidney of all males in sample (closed circles) and all females (closed squares), based on 3-year sliding average. Lower group: Corresponding data on total Hg in cerebrum of males and females, similarly plotted

within single age classes. Only for liver were results obtained which paralleled those of Olsson, with the highest correlation of mercury levels being with body weight, and even that with length slightly higher than that with age. In muscle, cerebrum and kidney on the other hand the correlations between mercury

Table 10. Least squares relationships between ΣHg (ppm) in muscle and liver and chronological age of harbor porpoises from southern New Brunswick waters, centered on 1970-76 successively, and calculated using three years' data each time, 1969-77, to reduce effects of intra-year variation

Tissue	Sex	f(= n−2)	Centered year	Calculated r value	Least squares fit of ppm (y) against age (x)	Estimated p value
		22	1970	0.61	0.44 + 0.08 x	$0.01 > p > 0.001$
		20	1971	0.64	0.38 + 0.07 x	$0.01 > p > 0.001$
		22	1972	0.65	0.39 + 0.10 x	$p = 0.001$
Muscle	♂	13	1973	0.52	0.43 + 0.09 x	$p = 0.05$
		14	1974	0.72	0.49 + 0.14 x	$0.01 > p > 0.001$
		13	1975	0.55	0.56 + 0.99 x	$0.05 > p > 0.02$
		19	1976	0.40	0.71 + 0.13 x	$0.10 > p > 0.05$
		22	1970	0.77	0.22 + 0.26 x	$p < 0.001$
		16	1971	0.85	0.15 + 0.85 x	$p < 0.001$
		18	1972	0.83	0.27 + 0.25 x	$p < 0.001$
Muscle	♀	9	1973	0.89	0.32 + 0.21 x	$p < 0.001$
		7	1974	0.91	0.48 + 0.18 x	$p < 0.001$
		5	1975	0.33	Not significant	$p > 0.20$
		5	1976	0.51	Not significant	$p > 0.20$
		8	1970	0.82	−0.38 + 1.98 x	$0.01 > p > 0.001$
		10	1971	0.79	−0.87 + 1.71 x	$0.01 > p > 0.001$
		12	1972	0.82	−0.59 + 1.66 x	$p < 0.001$
Liver	♂	8	1973	0.70	0.03 + 1.14 x	$0.05 > p > 0.02$
		9	1974	0.78	0.01 + 1.80 x	$0.01 > p > 0.001$
		13	1975	0.65	1.70 + 2.32 x	$0.01 > p > 0.001$
		19	1976	0.50	3.10 + 3.08 x	$0.05 > p > 0.02$
Liver	♀	Insufficient data				

levels and age consistently exceeded those for weight and length in that order (Table 7). The relationships of mercury in kidney to increasing body length, and of levels in cerebrum with increasing weight or length, were not statistically significant.

We believe that these results differ from those obtained by Olsson (with the proviso that we are after all comparing a poikilotherm and a homeotherm) principally because mercury levels in fish can change quite rapidly; not only is there a flux via the alimentary canal, but also across the gills and skin (Järvenpää *et al.* 1970). There is to the contrary no evidence that significant exchange occurs through the skin of cetaceans, which have a cornified epidermal layer 50-300 μ thick (Ling 1974) and since the harbor porpoise is several times the size of the pike, and has a larger volume to surface area ratio (the functional surface area in the case of the fish is also greatly increased by the presence of gills), then if such loss did occur a measurable change in tissue levels would take proportionately much longer. We might expect significant changes to occur within a relatively short time in the organs of active dietary metabolism, and the liver seems to be the site where the mercury, largely taken up as methyl-mercury from fish muscle (Berglund *et al.* 1971), is being metabolized and de-methylated or bound to proteins, apparently in the presence of selenium (Koeman *et al.* 1972, 1973).

The relationships between mercury levels in different organs of females of reproductive age showed consistently lower correlations than those in males of the same age class. This probably reflects greater mobility, and variation in metabolism of mercury in the former, associated with the reproductive load.

We could find no significant differences in mercury levels between cerebrum and cerebellum in either sex although such were reported in young pigs by Platonow (1968).

Koeman *et al.* (1973) presented data concerning specimens in which inordinately high mercury levels had been recorded, and suggested that in these cases the demethylation or bonding mechanism had been overloaded. A sign of this was considered to be the appearance of high mercury levels in the brain. A few such "anomalous" specimens occur in our sample; three females with respectively 63, 91, and 112 ppm of total in their liver. In the case of the first and third of these specimens, levels in cerebrum were also analyzed; they were 5.99 and 9.05 ppm respectively = levels at which some mammals begin to exhibit neurological symptoms of mercury poisoning (Berglund *et al.* 1971, Table 9.4, summarizing data of several authors). The third specimen (14 A 71), also had over 11 ppm in the uterus, and 4.38 ppm in its spleen, probably indicative of elevated blood mercury levels. The routine field autopsy report revealed nothing unusual. Unfortunately, the mercury analysis was not carried out until long after the remains had been discarded. These three specimens were omitted from certain calculations in the Results to avoid gross distortion of the arithmetic sample means.

It was no surprise to find little evidence of regional differences in mercury levels since the main areas of comparison (Deer Island region, Grand Manan and Digby) are only about 60 km apart at most, and even collecting locations on the Atlantic coast of Nova Scotia are washed by the same cool Nova Scotia current which turns into and westwards across the mouth of the Bay of Fundy (Bumpus 1960). Specimens were too few in number to permit broad conclusions to be drawn about levels in other areas. There is no sure explanation for the significantly higher levels in females from the Digby region; they could be artifactual. The average age was within acceptable limits for statistical comparison, but there was somewhat more representation of older females than in some other samples. If the difference was valid, it might be explicable in terms of Digby males (and both sexes of southern New Brunswick animals) moving out in winter into areas of lower mercury background than Digby females. It is known that some porpoises in this population maintain local distributions while others wander lengthy distances (Watson 1976, Gaskin, *et al.* 1975), and that some degree of sexual segregation occurs within this population.

The relationship between total mercury and the methylated fraction present in muscle and liver of this species was initially described by Gaskin *et al.* (1972), based on 10 determinations. More analyses were carried out for the present study, but they served only to confirm earlier results, namely that in muscle the methylated fraction approaches 100% (minimum 76%), and that in liver the percentage is much lower (2.5 to 41%), with a progressively lower methylated fraction in liver as the total concentration increases. These are very similar to results reported by Gaskin *et al.* (1973, 1974) for other species of marine mammal.

Koeman *et al.* (1973) stated that "It has been suggested (by Parizek *et al.* (1969) that mercury and selenium occur together in animal tissues and are associated with proteins by means of sulphur. Our observation that most of the mercury in seal liver and brain was tighly bound and could not be recovered in the form of methyl mercury may support this suggestion". Subsequent work indicates that the majority of mercury in liver is indeed not in the methylated form. It would seem, however, a mistake to assume that the "tight bonding" referred to by Koeman and his co-workers necessarily refers to anything other than the bonding being resistant to the particular type of chemical attack used in the analysis; data indicate that the mercury can be readily transferred from one molecule to another under appropriate circumstances in the living organism, as outlined below.

The presence of mercury in liver and kidney stimulates biosynthesis of a specific type of protein (metallothioneins) (Piotrowski *et al.* 1974), and atoms of heavy metals such as mercury and cadmium become bound to these proteins (Piotrowski *et al.* 1977). The formation of metallothioneins may render the mercury less toxic to the organism, (Wisniewska *et al.* 1970), but in the presence of selenium, mercury can be transferred from kidney to liver, and from metallothionein to soluble and non-histone protein fractions (Komsta-Szumska and Chmielnicka 1977). When the mercury is so transferred, it may once again become active as a toxin. The readiness with which mercury is transferred from site to site and from one protein to another seems to be sensitive to changes in tissue levels of selenium. These studies clearly established that mercury which is protein-bound is mobile in some degree, and presumably has to travel through the blood stream if moving from one organ to another. It is most likely to be eliminated from the body in transition; the biological half-life of mercury in blood is barely one third of that in the body as a whole (Nordberg *et al.* 1970). The work of Nomiyama and Foulkes (1977) appears to preclude heavy metals being eliminated via the urine in protein-attached form, although mercury can be readily eliminated from the kidney when in ionic form according to Magos and Butler (1976), who indicated that the usual route of elimination in mammals is through the feces.

There is reason to believe that mercury is in dynamic association in the liver and kidney, so levels in these organs might change more readily (albeit slowly) in response to alterations of background level in the food, than those in some other tissues. Retention of mercury in non-living tissue such as hair, is well known (Berglund *et al.* 1971). Mercury has been studied much less in muscle than in other major organs, but preliminary evidence seems to indicate that mercury replaces calcium in muscle (Dr. J. C. George, University of Guelph, pers. comm.), and is bound through carbon-mercury and sulphydryl linkages. In this condition, it would seem to be bonded in a rather stable fashion (Berglund *et al.* 1971); we offer our own data as additional circumstantial evidence—fluctuations in mercury levels during the study period were proportionately much less in muscle than in liver.

The biological half-life of mercury in marine mammals seems to be long in comparison with man (Clarkson 1977); up to 700+ days instead of 70-80. Over a study period as long as nine years, any changes taking place should be evident. We suggest that exchange rates for mercury in the muscle of cetaceans appear

to be slower than those in major organs of metabolism such as the liver. Even the passage of mercury in and out of the brain seems to be retarded to some extent by the blood-brain barrier (Berglund *et al.* 1971).

If changes in total mercury levels in the liver represent (even crudely) changes in the background level of mercury to which harbor porpoises were being exposed during 1969-77, then this population experienced a reduced Hg background during 1971-73 compared to 1974-77, and probably in comparison to 1969-70. There is no evidence suggesting that the diet of this population (largely herring, mackerel and small gadoids) has changed during 1969-77 (Smith and Gaskin 1974, and subsequent sampling). Mean liver levels in 1-year olds remained relatively low, and more or less constant from 1970-74, and only began to rise steeply in 1975. During this period of its life, the harbor porpoise grows very rapidly (Gaskin and Blair 1977), with a high food intake-to-growth conversion sustained into the second year. The reduced Hg background level would not necessarily be evident under such conditions. Liver levels in 5-year olds of both sexes declined drastically in 1971, remained low for three years, and increased steadily again from 1974 onwards (Figure 8). Very similar curves were obtained when mean values for males and females of all age groups were plotted.

The limited data for cerebrum and kidney are more difficult to interpret, but they also seem to reveal relatively low levels from 1970-73, followed by a rather rapid rise in 1974.

The pattern for mercury levels in muscle of mature males (Figure 6) is nearly identical to that plotted for all males (Figure 5). On the other hand, the mean levels in muscle of mature females showed an average decrease from 1971 to 1974, rising again in 1974-76 (Figure 7). This is consistent with expected differences in mercury mobility in mature males and females because of the reproductive load of the latter. Assuming that loss of mercury from muscle is relatively slow, the decline over the 1971-73 period might be relatively small in males or even non-existent, with perhaps a slow down of the rate of accumulation. Because variability is normally quite extensive, either condition is hard to detect in limited data. In mature females bearing young[6], loss of mercury is likely to be more extensive (Skerfving 1974, Clarkson 1977), and during conditions of reduced environmental mercury background, a moderate net loss could be expected. The curves in Figure 7 may also reveal a time lag of about one year before the drop becomes significant. This would also be expected, because of the long biological half-time in marine mammals (Clarkson 1977). The pattern of levels in muscle of yearlings was similar in both sexes, mercury concentration rising more steeply after 1974 (Figures 6, 7).

There is some evidence of an environmental change in 1971 in the waters off Deer Island, from water temperatures recorded between 1970-78 (except for a 1974 instrument failure). The values for 1970-78 are shown in Figure 10.

Those obtained (with standard errors) were:

1970: 11.56 ± 0.18; 1971: 12.07 ± 0.07; 1972: 11.83 ± 0.14;
1973: 11.89 ± 0.09; 1974: (records unusable); 1975: 11.49 ± 0.13;
1976: 11.36 ± 0.09; 1977: 10.96 ± 0.09; 1978: 10.90 ± 0.08.

[6] The full-term foetus is also proportionately very large in this species (Fisher and Harrison 1970).

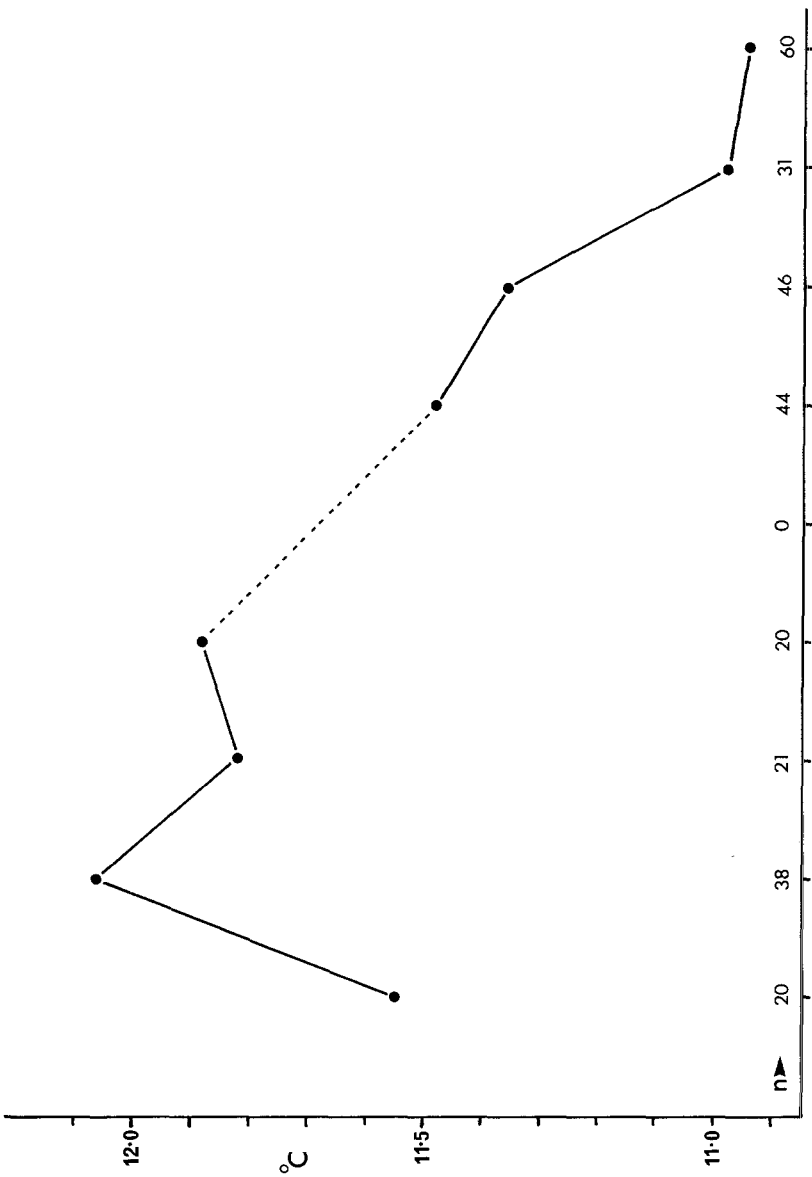

Fig. 10. Average sea surface temperatures (during second half of August) off Deer Island, New Brunswick, 1970-78, in °C. Total number of observations by Beckmann RS-5 salinometer/thermistor indicated for each season by n. Dotted line represents extrapolation only since no data were available for August 1974

A rise in average sea surface temperature of 0.5°C occurred in or by the second half of August 1971 in comparison with 1970; temperatures in August 1972, 1973 were also higher than in the latter year. By 1975, they declined quite steeply to a level even lower than in 1970. There is, therefore, a strong indication that a significant intrusion from the Gulf of Maine of warmer and, judging by comparative studies of mercury levels in harbor seals (Gaskin *et al.* 1973), relatively mercury-poor water, occurred during 1971-73. The food web in the Bay of Fundy was presumably exposed to lower than normal concentration levels (with 1970 as the arbitrary standard for "normal") for 2 to 3 years until the influence of this was counteracted by retreat of the southern warm salient and/or complete mixing with Bay water. Changes of this nature and magnitude have been documented in previous years (Bailey *et al.* 1954). We do not imply that the sea temperature *per se* is necessarily a direct operative factor, but rather that the fall in 1971-73 was indicative of a prevalence of Gulf Stream-origin rather than Scotian Current-origin water dominating the study area for a time. In August 1971, right whales *Eubalaena glacialis* penetrated as far as the Deer Island-Campobello Island region of the Bay of Fundy; usually they come no further into the Bay than the southern end of Grand Manan Island. This was indicative of unusual conditions.

A recent paper by Sutcliffe *et al.* (1976) attempted to show that the Nova Scotia current water is affected by discharge from the St. Lawrence at certain times of the year. Significant levels of mercury occur in fish and seabirds in the Gulf of St. Lawrence (Pearce *et al.* 1973), and up to 88 ppm have been found in gray seals *Halichoerus grypus* from Sable Island off the Atlantic coast of Nova Scotia. Annual levels of mercury at each trophic level in the Bay of Fundy may therefore be greatly influenced by the relative dominance of Nova Scotia current water versus a Gulf Stream salient each season. A summer season (about 120 days) should be ample time for a significant change in mercury levels in the tissues of the fish species on which harbor porpoises prey, on the basis of data presented for fish by Järvenpää *et al.* (1970).

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