The Accumulation of Organic and Inorganic Mercury Compounds by the Eastern Oyster (*Crassostrea virginica*)

by

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The problem of mercury concentrating in the food chain was discovered and reported first in Japan in 1959, later in Scandinavia in 1967 and more recently in the United States and Canada, in 1969. Use of mercury world-wide is estimated at about 5000 tons per year, of which 2200 tons per year are used in the United States (MIETTINEN). This increased load on natural mechanisms for detoxification and disposal of mercury has allowed the element to accumulate in the near-shore environment around municipal and industrial disposal areas.

IRUKAYAMA (1966) reported that water-insoluble inorganic mercury compounds found in the mud in Minamata Bay, Japan, were scarcely accumulated by shellfish (*Venus japonica*) cultured experimentally in seawater containing the mud. He also reported that when the shellfish were exposed to inorganic and organic mercury compounds, alkylmercury compounds were concentrated whereas other organic and inorganic forms were not. In another report, IRUKAYAMA (1968) stated that mercuric chloride was concentrated by these shellfish.

Recent reports (MIETTINEN, HANNERZ, JERNELOV) show that metallic, inorganic, and phenylmercury are coverted to methylmercury by the hydrospheric microflora so that the mercury is available for concentration by the biota regardless of the chemical form of mercury introduced.

The eastern oyster is known to concentrate trace metals from the environment. The presence of mercury contamination in some coastal waters of the United States prompted this investigation into the extent of concentration of various mercury compounds by shellfish in those coastal waters. This study was designed to investigate the differences in mercury uptake when oysters were exposed to specific environmental levels of inorganic, phenyl- and methylmercury compounds.

Experimental

Commercial size oysters dredged from a public reef in Mobile Bay, Alabama, were transported to the U.S. Environmental Protection Agency, Gulf Coast Water Supply Research Laboratory on Dauphin Island, where mussels and barnacles adhering to the shells were removed. The oysters were then placed in flowing seawater

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for two weeks for acclimation. The oysters were then placed in single layers in baskets made of fiber glass dowels. The baskets were placed in 75-liter flow-through aquaria. Aqueous stock solutions of the mercury compounds, introduced into the system with Beckman* solution metering pumps, were mixed with the incoming water by flowing down baffled plastic troughs into each aquarium. Concentrations of mercury in the experimental system were verified by flameless atomic adsorption spectrophotometry. The mercury compounds evaluated were mercuric chloride, Fisher certified reagent; methylmercuric chloride, Columbia Organic Chemicals Company; and phenylmercuric acetate, Aldrich Chemical Company.

To decrease any chance of estuary contamination by mercury from the experimental system, all effluents were passed into the soil through two drums containing gravel, sand and activated carbon. This system was shown to remove more than 95% of the mercury from the effluent at the 50 µg/l concentration and an average of 75% at the 1 ppb level. To further verify this removal, water samples from five locations in the estuary around the laboratory were monitored weekly for mercury. No significant increases in mercury levels were observed during the course of the experiments in these environmental samples.

Two experiments were conducted with three groups of 100 oysters each. In the first experiment, mercury levels were maintained at 50 µg/l. The temperature of the water fluctuated between 0 and 10°C; flow rate was one l/oyster/hr. In the second experiment, the mercury levels were reduced to 1 µg/l, the temperature of the water varied between 25 and 35°C, and flow rates were maintained at two l/oyster/hr because of the increased temperature. Control oysters were treated in an identical manner but no mercury was added to the water.

At irregular intervals, 10 oysters from each aquarium were removed, shucked and the pooled meats homogenized. Duplicate aliquots of each homogenate were digested by the method of MAYER and analyzed for mercury by flameless atomic absorption spectrophotometry. Analyses of other trace metals were made by conventional atomic absorption after digestion of the tissue in nitric acid.

RESULTS

First Experiment

Mercury Accumulation

In the first experiment, the administration of organic mercury compounds was terminated after 19 days because many of

*The use of trade names of products in this publication are example only and do not imply endorsement by the Environmental Protection Agency.

the oysters in the groups receiving either methylmercury or phenylmercury were observed to be dead or moribund. Oysters classified as moribund exhibited slow, incomplete valve closure when disturbed. When the oysters which survived 19 days of exposure to methylmercury and phenylmercury were placed in flowing seawater, about half of the oysters in each group died within a week and all oysters in both groups were dead within 14 days. Oysters exposed to mercuric chloride suffered no apparent ill effects over a 42-day period of exposure.

Table 1 presents the increases in mercury concentrations in the three groups of oysters during exposure and shows that the oysters concentrated the mercury so rapidly that detection of differences in the rates of accumulation was not possible. The average concentration of mercury in the experimental animals after one week was more than a thousand times greater than that of the controls.

TABLE 1.

The concentration of mercury in oysters exposed to 50 µg mercury/l water.

Mercury compound	Mercury concentration in oysters (mg/kg wet weight) Days exposure				
	7	14	19	28	42
Mercuric chloride	25	35	40	25	60
Methylmercuric chloride	21	26	33	-	-
Phenylmercuric acetate	15	30	23	-	-
Control	<0.02	<0.02	0.02	0.02	<0.02

Six oysters from each experimental group were dissected after 19 days of exposure. The mantles, gills, palps, adductor muscles, and the digestive diverticula of the oysters in each group were pooled separately, homogenized and analyzed for mercury (Table 2). The only result that is significant is the extremely high mercury content of the gills from oysters exposed to mercuric chloride. IRUKAYAMA (1968) reported that shellfish (*Venus japonica*) cultured in a medium containing $HgCl_2$ also exhibited the highest mercury content in the gill. This may indicate that much of the inorganic mercury is bound to particulate matter adhering to the gills. A preliminary experiment, which would confirm this result, did indicate that $HgCl_2$ was sorbed from the estuarine water by the particulate matter to a greater extent than methylmercury or phenylmercury.

TABLE 2. Concentration of mercury in selected tissues of oysters exposed to 50 µg mercury/l water for 19 days.

Mercury compound	Mercury concentration in tissue (mg/kg wet weight)				
	Mantle	Gill	Palps	Adductor	Digest. Divert.
Mercuric chloride	11	180	12	4	8
Methylmercuric chloride	14	13	11	3	6
Phenylmercuric acetate	9	3	4	5	6

Effects on Other Metals

The samples were also analyzed for cadmium, chromium, copper, iron, lead, manganese and zinc to determine if exposure to mercury affected their concentrations in the oysters. The only differences observed between the experimental and control oysters were the copper and zinc concentrations, as shown in Table 3.

TABLE 3. Effect of exposure to mercury on the copper and zinc content of oysters.

Form of mercury (50 µg/l) in water		Concentrations in tissue (mg/kg wet weight) Days exposure				
		7	14	19	28	42
Control	Cu	24	25	25	22	14
	Zn	700	700	735	745	500
Mercuric chloride	Cu	16	6	18	20	13
	Zn	525	175	210	600	450
Methylmercuric chloride	Cu	6	5	8	-	-
	Zn	180	175	140	-	-
Phenylmercuric acetate	Cu	8	6	7	-	-
	Zn	300	175	123	-	-

The concentrations of copper and zinc were depressed in all three groups of oysters after one week exposure to mercury. In the two groups exposed to organomercury the concentrations declined over the 19-day exposure. Copper and zinc levels in the oysters exposed to mercuric chloride began to increase during the third week and continued to do so until they were essentially the same as the concentrations in the controls.

Second Experiment

While accumulation of mercury by oysters in the second experiment was also rapid, following the accumulation rates more accurately was possible because of the lower dosing rate. Figure 1 shows that methyl- and phenylmercury were concentrated to essentially the same degree, while inorganic mercury was concentrated about four times less over a 74-day exposure.

The relative toxicities of the mercury compounds could not be calculated because oysters were removed for analysis during the experiment. The supply of oysters receiving methylmercury was exhausted after six samplings because 40 oysters had died during this interval. The supply of oysters receiving phenylmercuric acetate was exhausted after seven samplings because 30 oysters had died during this interval. When the experiment with mercuric chloride was terminated after 74 days, 22 oysters were still alive while only 8 had died. Morbidity of the type observed in the 50 µg/l exposure experiment was not apparent in

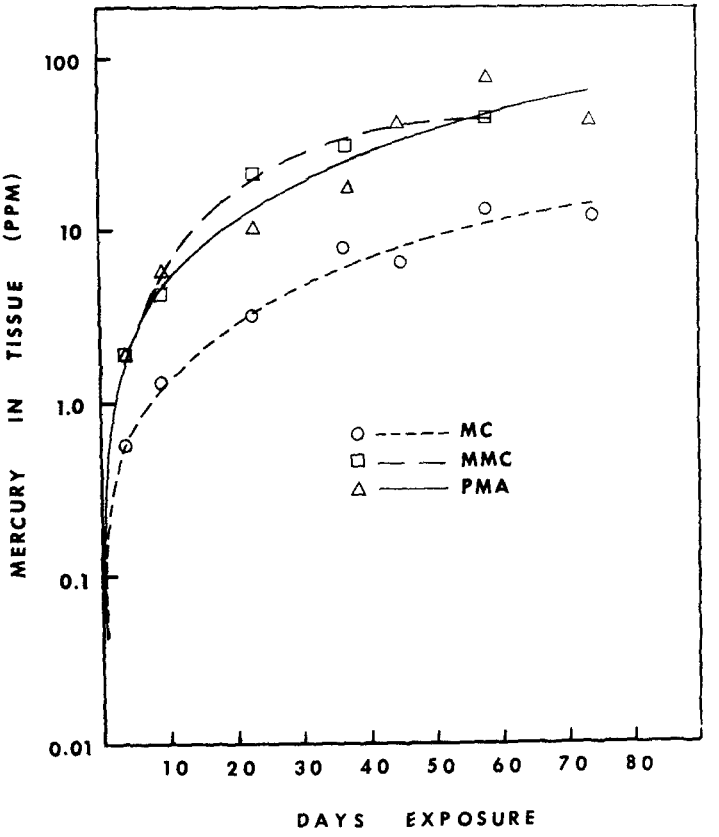

Figure 1. The accumulation of mercury by the three groups of oysters exposed to mercuric chloride (MC) methyl mercuric chloride (MMC) or phenyl mercuric acetate (PMA) at an environmental level of 1 part per billion mercury.

any of the oysters and no effect on copper and zinc levels was observed at this lower concentration.

DISCUSSION

One microgram per liter mercury is a concentration that can realistically be expected in mercury contaminated water. WERSHAW summarized the results of the analyses of more than 500 water samples representative of industrial outfalls where mercury contamination was suspected. He reported that 55% contained 0.1-5 µg/l and 17% contained in excess of 5 µg/l. HOSOHARA, *et al.* reported a concentration range of 1.6-3.6 µg/l in the waters of Minamata Bay, Japan.

IRUKAYAMA, *et al.* reported that mercury compounds were highly toxic to shellfish at concentrations of 0.5 mg/l or more mercury in seawater. Exposure of short-necked clams (*Venus japonica*) to 0.3 mg/l mercury as mercuric chloride or any of ten organomercury compounds produced 100% mortality in less than 14 days. Our experiments indicate that continuous exposure of the eastern oyster to even one µg/l mercury in any of the three mercury compounds studied, oysters rapidly concentrate mercury in their tissues far in excess of the 0.5 ppm "action guideline" established by the Food and Drug Administration (CELESTE and SHANE).

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