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Total and Organic Mercury in Benthic Organisms Near a Major Submarine Wastewater Outfall System

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This paper reports levels of mercury measured in tissues of six benthic animals representing four phyla from the vicinity of the Los Angeles County municipal wastewater outfalls off Palos Verdes Peninsula, Calif. (Fig. 1). Past studies (EGANHOUSE, et al. 1976) have shown that surface sediment concentrations in this area reach levels 10 to 100 times the measured background values found for sediments in deeper portions of cores (approximately 0.05 mg/dry kg). We made corresponding measurements on five of the same species collected from an offshore control site (Santa Catalina Island), where sediment concentrations of mercury range from 0.03 to 0.18 mg/dry kg (CHEN and LU 1974). Choice of these study areas was based on the fact that the waters off Palos Verdes Peninsula receive roughly 350 million gallons of municipal wastewater daily. The discharge of this effluent has led to a localized buildup in sediment heavy metal levels over the years (GALLOWAY 1972; SCCWRP 1973; BRULAND, et al. 1974) such that almost four metric tons of anthropogenic mercury are now contained in the upper 30 cm (YOUNG, et al. 1975). In view of the potential for microbially-mediated methylation of mercury in these sediments and subsequent mobilization to benthic organisms, we included total organic mercury measurements in this survey.

Briefly stated, our objectives were to determine: 1) if the relatively high surface sediment (0.14 to 5.5 mg/dry kg) and wastewater particulate (4-5 mg/dry kg) concentrations of mercury were reflected in the tissues of the local biota, 2) how total and organic mercury were distributed among the tissues of these animals, and 3) whether the levels observed in outfall specimens represented enrichment when compared with concentrations found in control specimens.

Methods and Materials

Specimens were obtained by otter trawl at depths of 8 m, 61 m, and 137 m off Palos Verdes Peninsula and at 76 m, 91 m, 137 m, 152 m, and 183 m off Santa Catalina Island. All animals were rinsed with fresh

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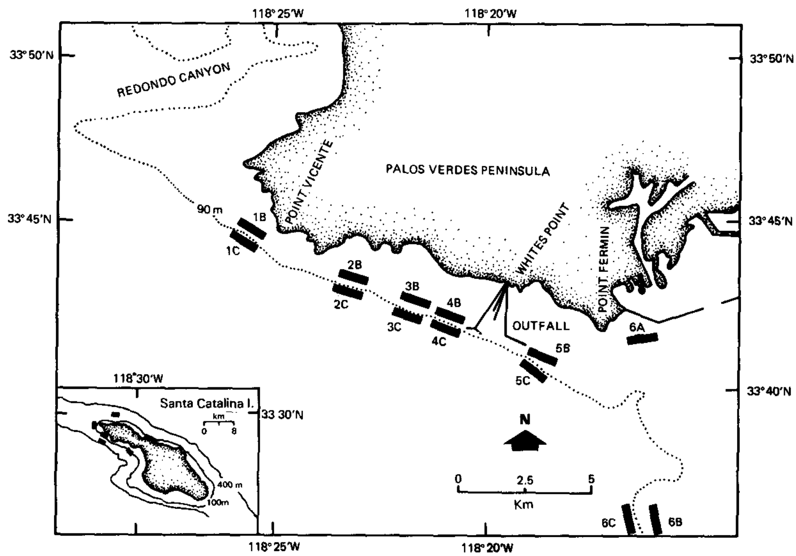
FIGURE 1

Trawl stations for benthic animal survey.



seawater and frozen in polyethylene bags until dissection could be performed. Once excised, each tissue sample was homogenized as a 50/50 mixture with deionized distilled water and analyzed for total mercury and total organic mercury. Total mercury content was determined by wet digestion (3:1 H_2SO_4/HNO_3) followed by a variation of the cold vapor atomic absorption (CVAA) technique described by HATCH and OTT (1968). Total organic mercury was measured using a modification of the method developed by RIVERS, et al. (1972) which employs liquid-liquid extractions for isolation of organomercurials and analysis by CVAA. The essential details of these techniques and a discussion of analytical precision and accuracy have previously been presented (EGANHOUSE 1975). It is acknowledged that "total" organic mercury is not necessarily synonymous with methylmercury; however, past studies (WESTÖÖ 1966; JOHANSSEN, et al. 1970; ZITKO, et al. 1971) have shown that the major portion of the mercury contained in fish is methylmercury.

Results and Discussion

Palos Verdes Survey

A summary of the results for total and organic mercury analyses of the Palos Verdes specimens is shown in Table 1. At least one species was obtained

TABLE 1

Levels of total and organic mercury in tissues of marine animals collected from the Palos Verdes shelf, 1975.

Species and Tissue	No. of Speci- mens	Total Mercury (mg/wet kg) Mean	Std. Error	Range	Organic Mercury (mg/wet kg) Mean	Std. Error	Range	Average Percent Organic Mercury
Dover sole, *Microstomus pacificus*								
Muscle	16	0.057	0.006	0.021-0.122	0.041	0.007	0.020-0.103	70.8±5.2
Liver	16	0.124	0.018	0.050-0.296	0.009*	0.002	0.002-0.018	9.6±1.7
Kidney	16	0.053	0.007	0.034-0.125	-	-	-	-
Gills	16	0.024	0.003	0.006-0.047	0.010	0.002	0.003-0.023	31.4±7.8
Crab, *Mursia gaudichaudii*								
Muscle	11	0.021	0.003	0.008-0.037	0.017	0.003	0.014-0.022	87.1±6.8
Digestive gland	11	0.030	0.003	0.011-0.042	0.005	0.001	0.001-0.010	16.0±2.9
Prawn, *Sycionia ingentis*								
Muscle	24	0.038	0.002	0.017-0.057	0.029	0.002	0.010-0.043	70.0±4.1
Snail, *Callinaticina oldroydi*								
Foot	8	0.005	0.002	0.002-0.012	-	-	-	-
Viscera	8	0.071	0.014	0.026-0.124	0.016	0.007	0.003-0.043	35.9±20.2
Urchin, *Allocentrotus fragilis*								
Gonad	3	0.021	0.001	0.020-0.024	0.003	-	-	15.8
Sea slug, *Pleurobranchaea californica*								
Whole body	23	0.015	0.002	0.002-0.030	0.007	0.001	0.003-0.017	49.6±5.1

*Only six liver samples were large enough to show detectable quantities of organic mercury.

at each trawl station although no single species was found at every station. Inspection of the mean values for all tissues reveals that in no case was the U.S. Food and Drug Administration 0.5 ppm guideline for edible seafood exceeded. While muscle tissue for most of the species tested showed high percentages of organic mercury, the mean levels were below 0.05 mg/wet kg.

The mean value of 0.041 mg/wet kg (organic mercury) observed for Dover sole muscle fell within the range of values for methylmercury found by ZITKO, et al. (1971) in marine fish from Nova Scotia Banks. Similarly, the mean level of total mercury in Dover sole muscle was statistically indistinguishable from data presented by CHILDS and GAFFKE (1973) for Dover sole caught off the Oregon coast (0.122 ± 0.076 mg/wet kg). DE GOEIJ, et al. (1974) found the livers of Dover sole collected during 1971-72 from the Palos Verdes shelf to contain 0.11 to 0.19 mg/wet kg total mercury, in good agreement with the value of 0.12 mg/wet kg reported here. Apparently, then, Dover sole from the Palos Verdes area contain similar levels of mercury in their tissues when compared to fish caught in other coastal regions. These data also indicate that liver mercury concentrations for the 1975 specimens were essentially the same as those measured in 1971-72 specimens by DE GOEIJ, et al. (1974).

It was of particular interest to know what, if any, effect sample location might have on the levels of mercury in benthic animals around the outfalls. Studies of the geochemical phase partitioning of mercury in the sediments have shown a progressive change in the distribution patterns of mercury with distance from the outfalls (unpublished results). Also, the concentrations of total mercury in sediments from transects 1 and 6 are an order of magnitude lower than those from transects 4 and 5, nearest the outfalls. As Figures 2a-2d and 3a, 3b illustrate, tissue concentrations fluctuated randomly about the mean values showing no systematic trend related to sampling locations.

Overall, these results indicate that, in spite of the marked contamination of sediments in which the animals live, tissue concentrations of total and organic mercury in benthic outfall organisms are low and apparently similar to those found for related animals from other parts of the world.

FIGURE 2

Mean total (unshaded) and organic (shaded) mercury content (mg/wet kg) in Palos Verdes Dover sole.

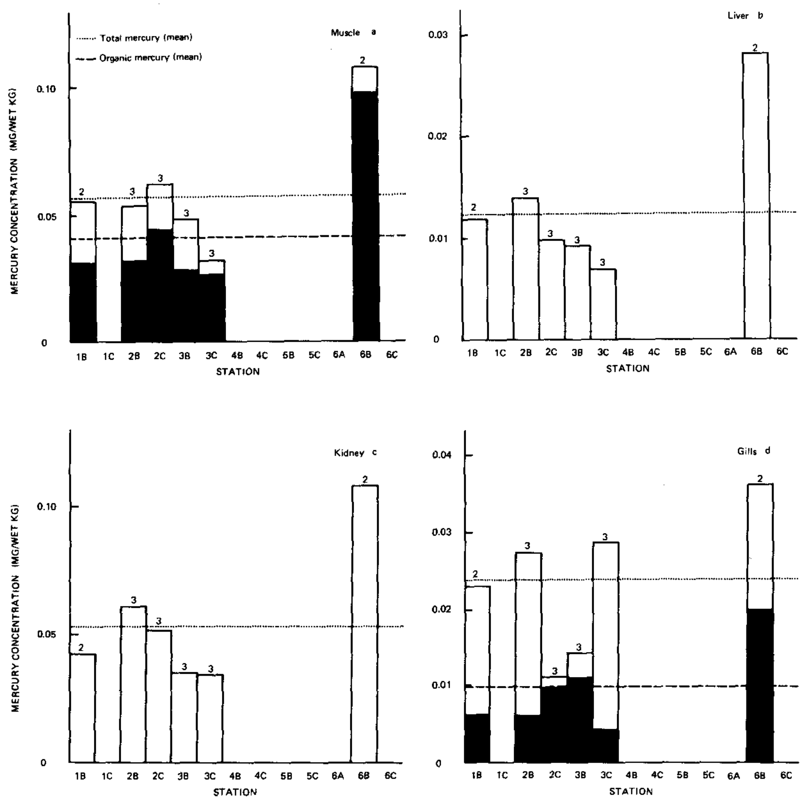
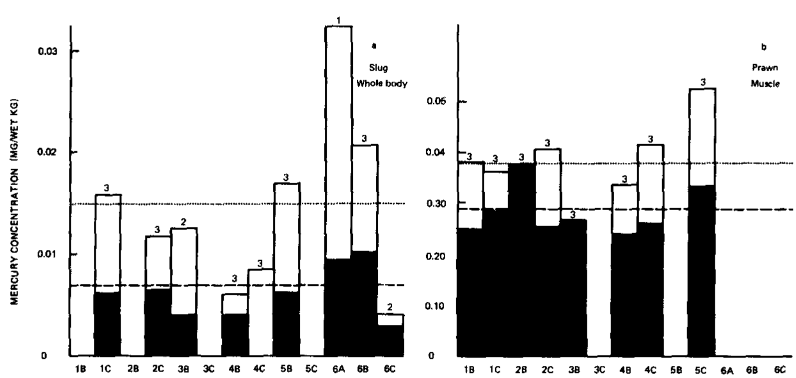


FIGURE 3

Mean total (unshaded) and organic (shaded) mercury content (mg/wet kg) in Palos Verdes sea slug and prawn.



Distribution of Mercury in Tissues

With the exception of the gills, tissues of the Dover sole showed high inter-tissue correlations with respect to total mercury (Table 2). Correlations between total mercury and organic mercury for various tissues of Dover sole also yielded high coefficients in some cases (Table 3). Significant correlations between tissue concentrations of mercury (total and organic) in Dover sole and body length and weight were observed also (Table 4). The coefficients for muscle were slightly higher than those found by RIVERS, et al. (1972). These results suggest that Palos Verdes Dover sole experience a chronic uptake of mercury expressed by the correlation of total mercury in muscle, liver, and kidney tissues with body weight and standard length. Total mercury shows a consistent distribution among the three tissues (Table 2); however, the diverse correlations between total and organic mercury (Table 3) probably results from dissimilar behavior of the various forms of mercury.

TABLE 2

Correlation coefficients for total mercury in tissues of Dover sole, Palos Verdes.

	Muscle	Liver	Kidney	Gills
Muscle	1	0.88*	0.82*	0.30
Liver	-	1	0.89*	0.40
Kidney	-	-	1	0.37
Gills	-	-	-	1

*p < 0.001

TABLE 3

Correlation of total mercury with organic mercury for Palos Verdes Dover sole.

		Organic Mercury			
		Muscle	Liver	Kidney	Gills
Total Mercury	Muscle	0.86*	0.70**	-	0.85*
	Liver	0.91*	0.51	-	0.75**
	Kidney	0.73**	0.13	-	0.69**
	Gills	0.47	-0.82	-	0.12

* p < 0.001
** p < 0.01

TABLE 4

Correlation of total and organic mercury in tissues of Palos Verdes Dover sole with body weight and standard length.

	Body Weight	Standard Length
Muscle		
Total mercury	0.55 †	0.67*
Organic mercury	0.71*	0.58†
Liver		
Total mercury	0.55†	0.66*
Organic mercury	-0.11	0.26
Kidney		
Total mercury	0.61**	0.63*
Gills		
Total mercury	0.20	0.22
Organic mercury	0.62**	0.64**

* $p < 0.01$
** $p < 0.02$
† $p < 0.05$

Note: Range in standard lengths of specimens was 193 mm to 260 mm.

Comparison of Outfall and Control Specimens

Table 5 presents a comparison of data obtained for the total mercury content of tissues for specimens from Palos Verdes and Santa Catalina Island(SCI). In this comparison median values were used because the distributions of values determined for SCI specimens were, in some cases, extremely skewed. For the Palos Verdes specimens there was excellent agreement between mean and median values.

It is noteworthy that in only three of nine cases did the median total mercury levels in tissues of Palos Verdes specimens exceed those found for the SCI control organisms: Dover sole kidneys and gills, and the sea slug. Thus, instead of showing enhanced uptake and accumulation, the outfall organisms tested generally had similar, and sometimes apparently depressed, tissue concentrations of mercury when compared to control specimens. Similar to the findings of DE GOEIJ, et al. (1974), these results are contraindicative of a direct relation between the bulk

TABLE 5

Total mercury (mg/wet kg) in tissues of five benthic animals from off Palos Verdes Peninsula (June 1975) and Catalina Island (January 1976).

Species &	Palos Verdes			S. Catalina Island		
Tissue	N	Median	Range	N	Median	Range
M. pacificus						
Muscle	16	0.052	.021-.122	8	0.157	.050-3.17
Liver	16	0.099	.050-.296	7	0.141	.078-.329
Kidney	16	0.041	.034-.125	8	0.030	.005-.036
Gills	16	0.024	.006-.047	7	0.019	.007-.027
M. gaudichaudii						
Muscle	11	0.018	.008-.037	6	0.158	.067-.516
Digestive gl.	11	0.033	.011-.042	6	0.081	.070-.282
S. ingentis						
Muscle	24	0.038	.017-.057	11	0.049	.022-.089
A. fragilis						
Gonad	3	0.020	.020-.024	6	0.034	.016-.051
P. californica						
Whole body	9	0.016	.002-.030	4	0.011	.006-.016

sediment concentrations of mercury and levels found in tissues of the indigenous epifauna. This may be explained by recent findings in this laboratory that mercury in Palos Verdes sediments is largely refractory. Hence, the transfer of mercury from these sediments to benthic organisms appears to be effectively prevented by fixation of mercury in the non-degradable fraction of the sediments.

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