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HEAVY-METAL CONTAMINATION OF *CRASSOSTREA VIRGINICA* AND ASSOCIATED SEDIMENTS OF THE CORPUS CHRISTI BAY SYSTEM, TEXAS

by

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Contents

Illustrations

Tables

ABSTRACT

In a preliminary survey, Crassostrea virginica from areas of the Corpus Christi Bay system of Texas show significant concentrations of Cd, Cu, Pb, and Zn in their tissues and shells; concentrations of these same metals in associated sediments are also high in certain areas of the bay system. Zn and Cd concentrations in tissue show a high negative correlation to each other, whereas Zn and Pb in tissue and shell show a high positive correlation to one another. Sediment contents of Pb and Zn best reflect tissue values of the heavy metals; sediment concentrations of Cd and Cu show a poor inverse correlation to tissue concentrations. Some possible factors influencing these correlations are suspended-sediment type, physiological changes, water quality, and Ca intake.

INTRODUCTION

Coastal environments are more industrialized and urbanized than in any previous time in history. As a result, the natural stability and integrity of coastal environments and processes are increasingly jeopardized, as is human health. Seacoast industrialization and urbanization have made coastal regions receptacles for numerous pollutants. Heavy metals are good indicators of anthropogenic contamination and are of particular interest to human health because of possible biomagnification.

Corpus Christi Bay (Fig. 1) is a shallow estuary less than 5 m deep; it is separated from the Gulf of Mexico, in its eastern boundary, by the Mustang Island barrier. Nueces Bay, on the western boundary, is a very shallow bay, less than 1 m deep, and adjoins Corpus Christi Bay. A dredged ship channel 15 m deep and 123 m wide almost bisects Corpus Christi Bay and terminates in a narrow, 20-km-long, landlocked harbor just south of Nueces Bay. Petroleum refineries and petrochemical plants, paint manufacturers, grain terminals, and a zinc smelter are located along the length of the harbor.

This was a preliminary survey of the concentrations and possible relationships between concentrations of selected heavy metals--cadmium (Cd), copper (Cu), lead (Pb), and zinc (Zn)--in the American oyster, Crassostrea virginica (Gmelin), and associated sediments of the Corpus Christi Bay system of Texas. Sediments within the bay system provide a trace-metal-rich environment (Holmes et al., 1974; Holmes, 1974) for marine organisms. C. virginica can concentrate high levels of heavy metals (Bryan, 1971; Eisler et al., 1972; Shuster and Pringle, 1969) and are good biological indicators of heavy-metal contamination (Kopfler and Mayer, 1969; Huggett et al., 1973; Frazier, 1975, 1976). Crassostrea populations are distributed throughout the Corpus Christi Bay system. Crassostrea is found from the Gulf of St. Lawrence to the Gulf of Mexico and the West Indies (Miner, 1950, p. 573-574; Andrews, 1977); it has been introduced into Pacific Coast waters (Morris, 1966, p. 17; Hedgpeth, 1970). Therefore, Crassostrea virginica is a potential index of contamination in North American coastal waters.

METHODS

One hundred-fifty live Crassostrea virginica were collected from seven sampling sites within the Corpus Christi Bay system during July and August, 1974 and 1975 (Fig. 1). Each oyster (whole tissue mass) was removed from

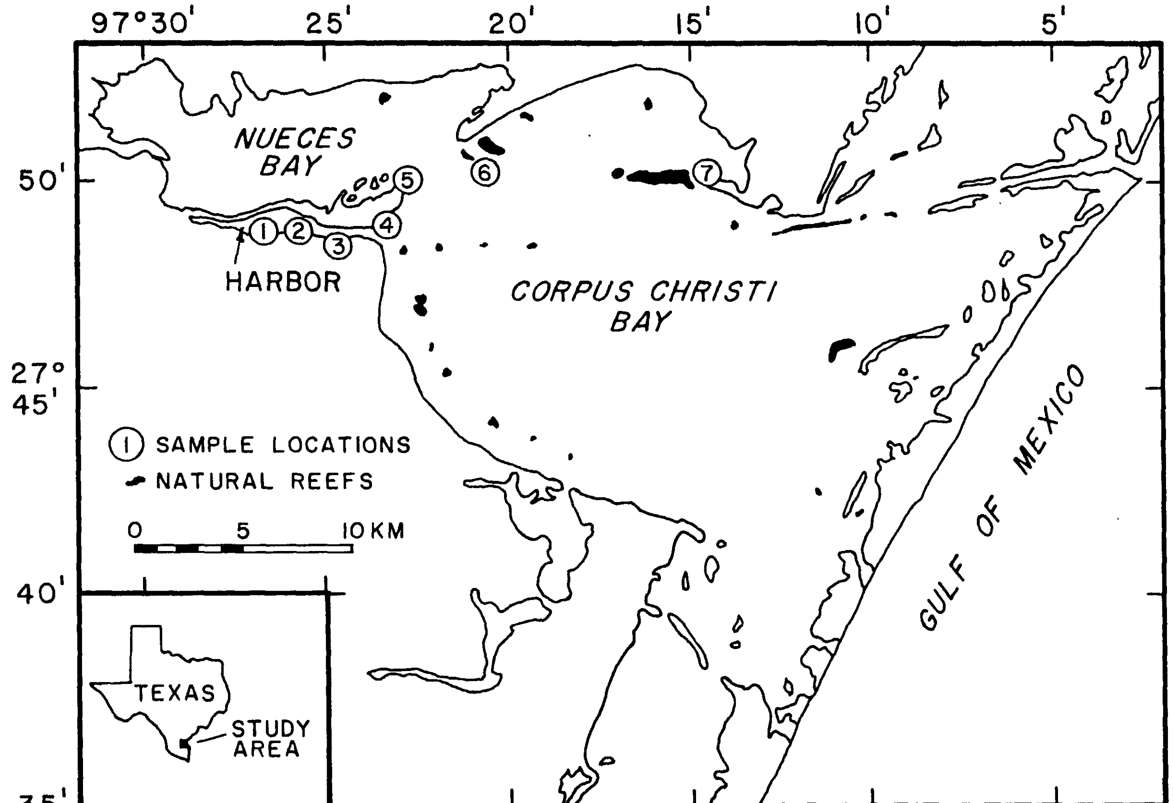

Fig. 1
Map of study area and sampling locations

its shell, placed in a preweighed Erlenmeyer flask, weighed, heated to dryness, and then reweighed.

A 2-ml aliquot of concentrated (36N), reagent grade, sulfuric acid was initially added to each tissue sample. Concentrated (16N) reagent grade, nitric acid was then added in 1-ml aliquots until digestion was complete. Each dissolved tissue sample was transferred to a volumetric flask and diluted to a known volume with deionized water. Appropriate dilutions of this final volume were analyzed for Cd, Cu, Pb, and Zn concentrations by atomic-absorption spectrophotometry. Cu, Pb, and Zn contents were determined by a flame spectrophotometer (Perkin-Elmer 303); Cd concentrations were determined on a graphite furnace spectrophotometer (Perkin-Elmer 360). The sample preparation and atomic-absorption procedure was checked for accuracy by analyzing bovine-liver standard obtained from the U.S. National Bureau of Standards (Table 1); duplicate samples were analyzed by the same methods used to analyze *Crassostrea* tissue.

Crassostrea shell samples were dried and ground to a fine powder in an acid-washed, ceramic mortar. Duplicate 1-g samples were weighed and transferred to acid-washed test tubes with approximately 10 ml of concentrated (16N), reagent-grade nitric acid. They were heated and stirred at 90°C until cessation of brown nitrogen dioxide fumes (approximately 1 hour). The samples were then transferred to 50-ml teflon beakers and evaporated to dryness under infrared heat lamps at 101°C. One ml of 16N HNO_3 and 9 ml of deionized water were added to the dried samples; samples then were transferred to acid-washed test tubes and analyzed for the same heavy metals as the tissue by atomic-absorption spectrophotometry. The deviation of replicate shell samples was approximately 4%.

To determine heavy-metal concentrations of sediment at sites 1-4, 55 surface grab samples were collected from the harbor area; for site 5, 75 grab samples from Nueces Bay were analyzed and their metal concentrations averaged to represent the sediment data. The sediment values for sites 6 and 7 are represented by data from Holmes (1974). Because *C. virginica* are epifaunal filter feeders and can be affected by pollutants in a wide area around them, the sediment trace-metal values for the oyster sampling sites were determined by averaging the concentrations of the grab samples at sites surrounding the oyster populations.

Each sediment sample was homogenized, placed in an acid-washed evaporating dish, dried at 101°C, and ground in a ceramic mortar to pass through a 200u-mesh nylon screen. Duplicate samples were then placed in a muffle furnace at 450°C for 4 hours, reweighed after cooling in a desiccator, and leached with 16N HNO_3 using the same method as for the oyster shells. The samples were analyzed for Cd, Cu, Pb, and Zn content by atomic-absorption spectrophotometry. Duplicate samples showed a deviation of less than 4%.

To correlate the results of this study, which presents metal concentrations based on the dry weights of tissues, with results of other investigators who report concentrations based on wet weights of tissues, wet weight values were multiplied by a correlation factor of 4.9 because our samples lost 79% of their weight as water; this method of comparison was also used by Shuster and Pringle (1969).

Table 1. Comparison of trace-metal values for U.S. National Bureau of Standards bovine liver.

Element	NBS value (ppm)	This study (ppm)
Cd	0.27 $\pm$.04	0.28 $\pm$.02
Cu	193 $\pm$10	208 $\pm$3.6
Zn	130 $\pm$10	129 $\pm$1.7
Pb	0.34 $\pm$.08	0.33 $\pm$.01

RESULTS

The baseline study indicates the presence of significant concentrations of heavy metals in sediment and Crassostrea virginica within certain areas of the Corpus Christi Bay system (Fig. 2, Tables 2 and 3). Heavy-metal contents of the sediments (Table 2) decrease in concentration from site 1, in the harbor, to site 7, across the bay and away from the direct industrial influence of the harbor area. Sediments from sites 1 and 2 contain the highest concentrations of heavy metals; a trend toward reduced values of sediment heavy metals begins at site 4 (located at the entrance of the harbor). The values for harbor sites 1-3 show a wide range of concentrations (Table 2). These are representative values and are not the result of sampling error; individual values were verified with replicate analyses. Variations in metal contents are probably the result of sediment mixing by ship traffic in the harbor and of variations in the sediment texture. Although these concentrations may vary as much as 300% temporally at the same site, the presence of heavy-metal-rich sediment is evident at these sample sites.

Various amounts of Cd, Cu, Pb, and Zn are being concentrated in Crassostrea virginica of the Corpus Christi Bay system (Table 3). Zn contents in tissue and shell samples show a direct relationship to one another ($r = 0.85$, Table 4). The positive relationship between tissue and sediment concentrations of Zn is represented by $r = .80$; between sediment and shell, there is also good correlation ($r = .91$) (Table 4). Pb contents in tissue and shell samples show a direct relationship ($r = .84$) except at site 4 (Fig. 2). A leveling off of values for Pb is reached in tissue and shell from sites 3-7 (Fig. 2). A corresponding line is established for the sediment; the positive relationships between sediment values for Pb, compared to tissue and shell values, are $r = .99$ and .85, respectively (Table 4). Zn/Pb in tissue and shell show a high positive correlation ($r = .79$ and .97, respectively) (Tables 5 and 6). The Cd content of the shells remains consistent at all sites whereas Cd contents of the tissues are variable (Fig. 2); Zn/Cd concentrations in tissue show a high negative correlation ($r = -.83$). Cu concentrations for shells are relatively consistent whereas Cu contents of the tissues increase significantly from the harbor to the bay; the relationship of Cu concentrations in tissues and shells compared to sediment is poor; tissue and shell Cu contents correlate poorly with all heavy-metal contents determined in this study.

DISCUSSION

The heavy-metal concentrations in sediments begin decreasing (Fig. 2) at site 4 (entrance to the harbor), except for Cd; this decrease reflects increased dilution (i.e., dispersion) by bay-water circulation, increasing distance from the point source of the heavy metals, and the difference between the reducing environment of the harbor versus the oxidizing environment of the bay (Holmes, 1974; McLerran and Holmes, 1974; Suter, 1980). During summer months, stagnation of the harbor water increases the concentration of heavy metals, allowing significant amounts to precipitate in the reducing environment of the harbor bottom water. Bacteria may play a significant role in the transport of Zn and Cd from seawater to sediment; bacterial activity and H_2S production in the harbor are at a maximum during the summer as is Zn and Cd deposition (McLerran and Holmes, 1974). During times of increased water exchange between the bay and the harbor (e.g.,

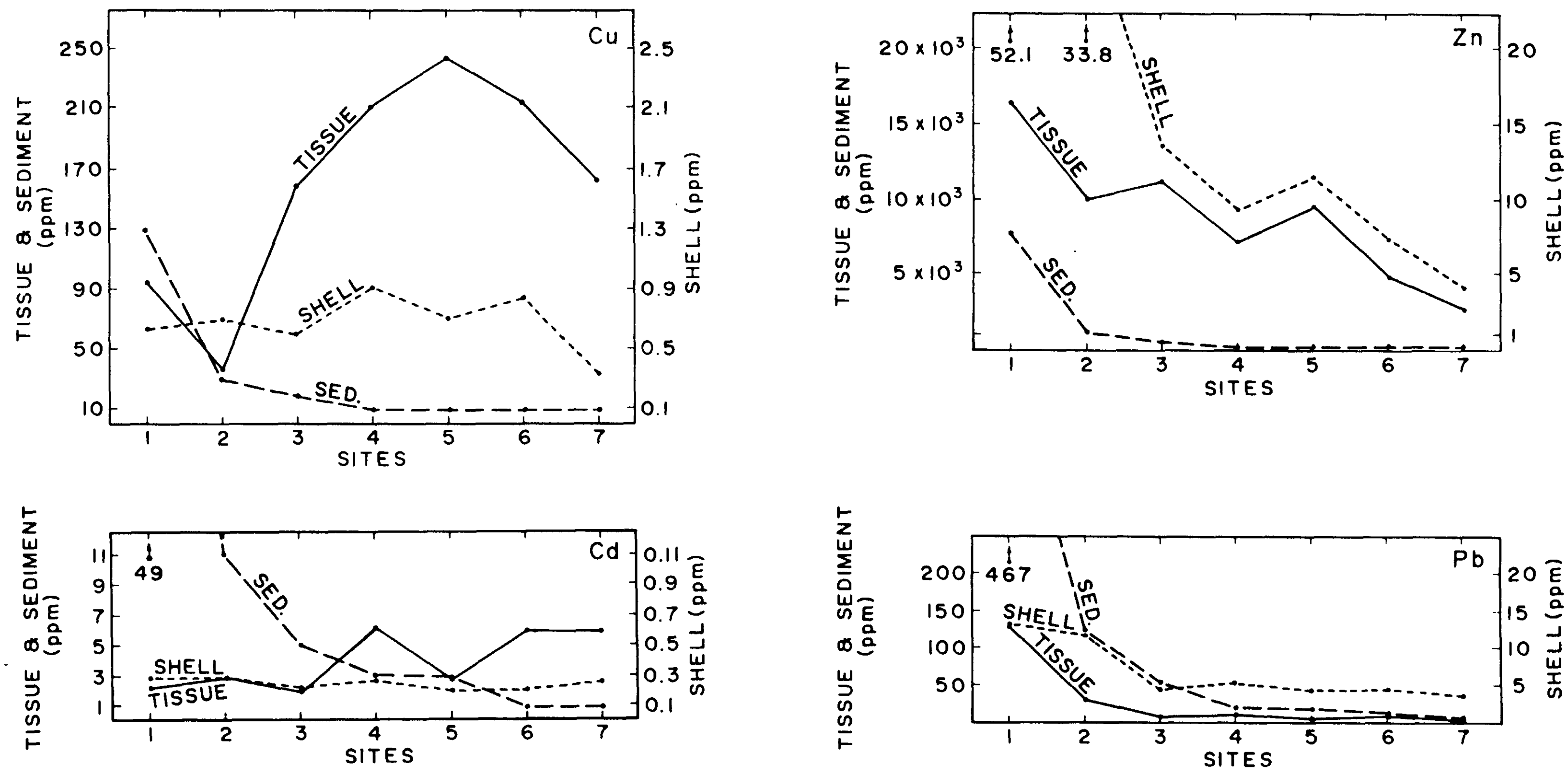

Fig. 2, Cu, Cd, Zn, Pb concentrations in tissue and shell of *Crassostrea virginica* and associated sediment

Table 2. Trace-metal concentrations (ppm) in sediment samples (data for sites 6 & 7 are from Holmes, 1974).

	Site no.	Cd Avg.	Cd Range	Cu Avg.	Cu Range	Pb Avg.	Pb Range	Zn Avg.	Zn Range
Harbor	1	49	34-64	130	36-193	467	170-975	7839	2450-14000
	2	11	3-20	31	11-45	124	30-265	1367	350-3300
	3*	5	3-9	21	16-28	58	40-90	540	300-1150
Bay	4	3	1-13	14	6-38	23	10-30	193	150-350
	5	3	1-3	8	6-12	20	16-25	169	130-190
	6	<1	<1	8	5-15	11	4-23	15	9-20
	7	<1	<1	8	5-15	4	3-8	11	9-13

*Collected from filtering screens through the cooperation of Central Power and Light Company, Corpus Christi, Texas.

Table 3. Average trace-metal values for *Crassostrea* shell and tissue (dry wt.) and sediment samples for each sampling area (ppm).

	Site	Tissue				Shell				Sediment			
		Cd	Cu	Pb	Zn	Cd	Cu	Pb	Zn	Cd	Cu	Pb	Zn
Harbor	1	2.1	94	129.0	16414	0.27	0.63	13.0	52.1	49	130	467	7839
	2	2.8	35	31.1	10007	0.28	0.69	11.6	33.8	11	31	124	1367
	3	1.7	158	11.4	11256	0.21	0.59	4.8	13.6	5	21	58	540
Bay	4	6.2	211	8.1	7305	0.26	0.90	5.7	9.4	3	14	23	193
	5	2.6	243	6.5	9650	0.18	0.68	4.3	11.5	3	8	20	169
	6	6.0	213	9.2	4713	0.20	0.83	4.4	7.4	<1	8	11	15
	7	5.9	161	2.8	2649	0.25	0.31	3.8	4.1	<1	8	4	11

Table 4. Correlation coefficients between heavy metals in tissue, shell, and sediment (*statistically significant at 0.05 level).

	Sediment/Tissue	Tissue/Shell	Sediment/Shell
Cd	-.49	.06	.48
Cu	-.54	.25	-.05
Pb	.99*	.84*	.85*
Zn	.80*	.85*	.91*

Table 5. Correlation coefficients between *Crassostrea* tissue heavy-metal concentrations (*statistically significant at 0.05 level).

	Cu	Pb	Zn
Cd	.43	-.47	-.83*
Cu	-	-.54	-.44
Pb	-	-	.79*

Table 6. Correlation coefficients between *Crassostrea* shell heavy-metal concentrations (*statistically significant at 0.05 level).

	Cu	Pb	Zn
Cd	-.10	.71*	.57
Cu	-	.10	.03
Pb	-	-	.97*

winter months), the increased flow of oxygen-rich bay water into the stagnant harbor water results in desorption of some of the precipitated heavy metals (Holmes et al., 1974). As a result of this mixing by water currents, the metals are available for adsorption onto suspended sediment and transportation into the bay (Holmes et al., 1974).

The Corpus Christi Bay system has a wind-dominated (average velocity of 13 mph) sediment-dispersal system (Shideler and Stelting, 1981), and the suspended sediment can range from fine silt (0.012 mm) to clay (0.003 mm). The suspended sediment consists primarily of inorganic silt and clay and an organic fraction dominated by diatoms (Shideler, 1980); the dominant clay species is montmorillonite (smectite). Heavy metals of the Corpus Christi Bay system, therefore, have a convenient medium and means for their dispersal throughout the bay, for adsorption of heavy metals is the greatest for small particles (e.g., clay); additionally, phytoplankton can rapidly accumulate pollutants such as heavy metals from the water (Bryan, 1971). These heavy metals are, therefore, readily available to filter feeders such as *Crassostrea*, especially in contaminated, turbid waters. In this study, the Cu, Cd, and Zn concentrations in sediment from sites 3-7 are comparatively low, but tissue concentrations fluctuate, implying that heavy-metal uptake, in this case, reflects suspended-sediment and/or water quality.

Many factors may influence an organism's ability to concentrate heavy metals; these variables include ambient metal concentrations, salinity, pH, temperature, feeding rates (Wolfe, 1974), breeding seasons (Cossa et al., 1979), and Eh. The sex of an organism can also be a factor; as *Crassostrea* can change sex, an additional variable is added. *Crassostrea* closes its shell an average of 7 hours out of each 24-hour period, and ventilation rates are affected by pH (Nicol, 1967); it has been reported to pump 4-15 liters per hour (Loosanoff, 1950), depending upon water conditions. Because of these and other variables, precise ratios between sediment contamination and organism contamination are, at times, difficult to establish, but correlations do exist. As Frazier noted (1976), sediment contents are not an absolute predictor of biological tissue concentration but may indicate relative availability of metals to biota.

Frazier's study (1975) showed a high positive correlation between Zn, Cu, and Cd in tissue; *Crassostrea* studied by Frazier showed the following correlations in tissues: Cd/Cu $r = .71$; Cd/Zn $r = .88$; Cu/Zn $r = .87$. Our results are almost the reciprocal of Frazier's: Cd/Cu $r = .43$; Cd/Zn $r = -.83$; Cu/Zn $r = -.44$. Such obvious differences for the same species can have many explanations--especially when the vastly different and distant geographical locations of each study area are considered. Frazier's study was conducted for a 12-month period; salinity range was 2-13‰ and temperature ranged from 1° to 32°C. Our study was conducted during two summer months for two consecutive years; average salinity for the Corpus Christi Bay system was 33‰, and the average temperature was 33°C for these months. Consequently, with differing physical parameters inherent to each geographical region, the organisms' physiology can also be expected to vary. Loosanoff and Nomejko (1951) considered *Crassostrea virginica* of the east coast and Gulf coast divisible into different geographical or physiological races. The basis for their conclusion is that the northern oysters breed at lower temperatures than the southern ones. Ciliary activity in the gill of *Crassostrea* differs according to region also; Menzel (1955) supported the suggestion that Crassostrea exists as several physiological races, not only in relation to the temperature threshold in spawning, but also to other physiological functions. Consequently, the same species, in different geographical regions, may have their own particular responses to heavy metals; these different responses may be an example of specialized physiological adaptive radiation. The environmental differences between areas (especially temperature and salinity) can greatly affect interaction of heavy metals with organisms and each other.

Values for tissue Cu in this study do not correlate well with values for the other heavy metals analyzed, and Cu is the only heavy metal whose content steadily increased toward and peaked at station 5 (mouth of Nueces Bay); Cu values are apparently not greatly affected by Zn, Cd, and Pb concentrations for the correlation coefficients between them and Cu are poor (Table 5). There is no apparent point source for Cu in the area where it is the highest in tissue, i.e., the bay. The reason for the high Cu contents probably lies in the fact that the solubility of Cu is dramatically decreased under reducing conditions (the harbor), unlike the solubilities of some other transition metals which form stronger solution complexes with sulfide (Leckie and Nelson, 1975); once Cu makes its way into the oxidizing bay, it should become more readily available to organisms and suspended material. Crassostrea has a propensity for concentrating Cu in its tissues (Roosenburg, 1969; Bryan, 1971). Phillips (1976) observed erratic Cu uptake in Mytilus edulis and stated that Cu uptake for Mytilus was very dependent upon salinity and temperature. Another factor for the erratic Cu concentrations in tissues, as observed in Mytilus (Phillips, 1976), Pinctada radiata (Shiber, 1980), and Crassostrea of this study, may be the influence of hemocyanin--a copper-based blood found in many molluscs (Prosser, 1973). While blood-Cu probably remains consistent, the tissue's Cu values may be influenced by the presence of hemocyanin since they are constantly exposed to it at all metabolic levels.

Tissue Cd in this study shows an increase in concentration as distance from the contaminated harbor-sediment increases. This could be explained by Cd's increased availability in the oxidizing bay waters as opposed to the reducing environment of the harbor (Holmes et al., 1974). Harris et al. (1979) found a similar condition in an Australian estuary where Cd concentration also increased with increased distance from the neighboring industrial area. In addition, the increased tissue Cd levels away from the harbor may be the result of an antagonistic relationship with Zn. Ashby et al. (1980) demonstrated that in white rats, increased Cd concentrations decreased plasma Zn concentrations; in our study, the tissue Cd/Zn is $r = -.83$ and sediment Cd/Zn is $r = .70$.

Crassostrea maintained Pb levels in tissues below sediment values; Pb was the only element studied for which this relationship was consistently observed. The correlation coefficient between tissue and sediment for Pb in this study is $r = .99$. Bryan (1971) stated that Crassostrea may be able to control excess Pb by storage in the blood cells or tissues; it may lose this excess when the water concentration is reduced to some undetermined level. Increased Ca intake may be the mechanism for this Pb regulation in Crassostrea; in humans, vitamin D, phosphorus, and Ca influence deposition of Pb in soft tissue and bone. Lowering Ca and/or phosphorus content of the diet increases bone or soft-tissue Pb content at low levels of Pb intake; Ca is considered more important physiologically than phosphate or vitamin D in Pb regulation (Six and Goyer, 1970).

Zinc shows the most dramatic concentrations in Crassostrea of the four heavy metals analyzed (Fig. 2). Zn appears to be easily accumulated by bivalves (Prosser, 1973); for example, in Pinctada radiata, Zn concentrations were highest of the metal contents determined by Shiber (1980). Zn contents were also higher than those of other metals for Crassostrea specimens from the Chesapeake Bay region (Frazier, 1975). Bryan (1971) stated that Zn in bivalves is accumulated mainly by ingestion rather than from solution. In this study, tissue Zn concentrations are high compared to sediment concentrations, and the Zn/Pb contents in tissue and shell show a high positive correlation (.79 and .97, respectively). The sediment Zn/Pb correlation coefficient in this study is .99. The reason for elevated tissue Zn and a high Zn/Pb correlation may be explained by the suspended sediment type. The clay fraction in the Corpus Chrsiti Bay system is dominated by Na^+ montmorillonite (smectite). Na^+ montmorillonite has a strong affinity for Zn and Pb (Nriagu, 1980); this results from montmorillonite's high cation exchange capacity, which is higher than those of other clay species (Van Olphen and Fripiat, 1979).

The variation in Zn content of tissues in Crassostrea may also be directly related to Ca intake; Zn in oysters has been shown to be directly associated with a high intake of Ca (McIntyre and Mills, 1975); montmorillonite also has a strong affinity for Ca (Nriagu, 1980). Deknudt and Gerber (1979) observed that a lack of Ca not only aggravates the general toxic symptoms of heavy metals in humans, but also sponsors induction of genetic damage by Zn and Pb but not by Cd; they recommend a diet rich in Ca for persons who risk intoxication by these metals. Ca may be the element that regulates and protects Crassostrea virginica from the high concentrations of certain heavy metals.

Tissue can provide an indication of heavy metal condition in the environment at the time of sampling, whereas shell analysis may provide information on past conditions. Analysis of individual growth layers of Crassostrea shell may provide a more detailed record of the heavy metal history for a given area (Hastings, 1974). Crassostrea's shell is formed year round, even during the colder winter months when the oyster is not feeding and formation of soft tissue has stopped; direct measurement of Ca deposition, using ^{45}Ca, gives a rate of 30 mg/day at 25°-26°C (Nicol, 1967). The shell could be a source of contamination chronology and has been the subject of some work using X-ray analysis (Hastings, 1974).

GENERAL IMPLICATIONS

Crassostrea virginica specimens taken from the Corpus Christi Bay system of Texas show, for some areas, significant concentrations of Cd, Cu, Pb, and Zn in their tissues and shells; sediment concentrations of the heavy metals are consistently higher in the harbor than in the bay. Zn and Cd in tissue show a high negative correlation, whereas Zn and Pb in tissues show a high positive correlation. Sediment values of Pb and Zn best reflect tissue values of the heavy metals. Crassostrea and sediment concentrations of heavy metals are probably greatly influenced by the nature of the abundant suspended sediment in the Corpus Christi Bay system.

Invertebrates' basic physiology can vary from one genera to another; these may range from subtle to major differences. Addition of various environmental factors complicates understanding of basic physiology and how it responds to such factors; introduction to pollutants, such as heavy metals, further impedes understanding the interaction of all the variables. For example, two heavy metals may act synergistically in one organism but become antagonistic in a related species (Braek et al., 1980). The physiology of the same species can vary from one geographic region to another; such physiological differences may or may not be significant, depending upon the type of research being conducted. Regarding heavy-metal analysis in general, and oysters in particular, the same species of a geographically similar area can probably be compared to one another resulting in an accurate description of that geographic species' reaction to heavy metals and the interactions of the heavy metals within that species. When comparing data on the same species from dissimilar geographic regions, conclusions must be tempered with the understanding that such variables exist; nevertheless, the ensuing correlations, or lack of, increase the understanding of heavy metals, the organisms' physiology, and the reactions each has on the other under various conditions. For this study, insight has been gained into *Crassostrea virginica* of the Corpus Christi Bay system and how it accumulates certain trace metals, and its relationship to trace-metal contents in associated sediments.

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