

# The Non-Toxicity of Metals in the Sea

Willard Bascom

Director, Coastal Water Research  
Southern California Coastal Water Research Project  
Long Beach, California

Inorganic metals in the sea are not a hazard to sea animals or to persons who eat those animals. This paper reviews the evidence on which that conclusion is based and gives an alternative view of the meaning of the laboratory toxicity tests now in general use that have led to a contrary opinion.

In some places it is useful to dispose of metal-containing wastes in the ocean. Sewage sludges, industrial wastes and dredge spoils are frequently named as sources of "toxic" metals, and a great effort has been made to avoid putting these where they would be "available" to sea animals.

The evidence to be cited here will show that in open coastal waters, the circumstances greatly reduce metal availability. This, combined with natural body defenses, protects animals against metal toxicity. These data come from measurements of actual environmental conditions and of animals taken from open coastal waters with highly contaminated bottoms resulting from nearby waste discharges. The effects on animals exposed at high levels in the sea are contrasted with those in laboratory experiments.

I shall discuss the nature of metals released into the sea and present three kinds of evidence: (1) the inadequacy of most toxicity data, (2) measurements of metals in the food web, and (3) natural detoxification mechanisms in animals.

## BACKGROUND

A voluminous and growing scientific literature deals with marine pollution (Reish et al., 1980). A large part of it deals with concentrations of metals in sea water, bottom sediments and sea animals; another part covers LC50 toxicity tests in which plants and animals are exposed in the laboratory to various metals and combinations thereof. Still other papers describe changes in real world animal communities that are exposed to wastes containing metals without defining the cause of change. But few papers deal with effects on specific animals under actual ocean conditions. One result of this plethora of papers is a general suspicion that the mere presence of metals in the sea or the body of an animal should be equated with some toxic effect—even though the only evidence of the latter is based on unrealistic laboratory tests, not on evidence obtained under actual environmental conditions.

This scientific suspicion has been passed on to the public. Environmental groups, unable to evaluate the risks, fear the worst and seek more rigorous regulations. For them, every drop of mercury in the ocean raises the specter of another Minamata, and every animal displaced is equated with an endangered mammal. The result has been needless concern and regulatory overkill.

The author believes that the risk of metals toxicity in the

\* Southern California Coastal Water Research Project.

sea is very small and that there is adequate data to support this position. The effort now spent in attempting to protect sea animals against metals (that already are well protected by nature) could be much better expended on more important environmental problems.

## DATA COLLECTION

The data presented here were almost entirely obtained by the Coastal Water Research Project as part of its ten-year-long investigation of the effects of man on southern California coastal waters. Samples of wastes, sediments and animals were taken, processed and analyzed with meticulous care in the manner reported in previous papers and in the SCCWRP\* annual/biennial reports (Bascom, 1975-1980, Bascom, 1978). Briefly, each sediment sample for chemistry is a composite of five 2-cm-deep cores of the surface of an undisturbed Van Veen grab; benthic infauna larger than 1 mm were screened from a similar grab. Fish and larger invertebrates were collected in standard trawls and carefully dissected under a positive-pressure hood; very large fish were obtained from fishermen but dissected by Project scientists. Trophic levels were assigned by Dr. Alan Mearns; metals analyses were made by Dr. David Young, Pat Hershelman, Tsu-Kai Jan and Robert Eganhouse using the atomic absorption method.

## METALS IN THE SEA

The metals discussed here are, with one exception, the inorganic forms of Cd, Cr, Cu, Ni, Pb, Zn, Hg, Ag. These





are present in wastes and in the environment as ions, usually attached to particles (Goldberg, 1954) or as any of several compounds, often sulfides. Table 1 shows some natural environmental levels, input levels and resulting sea floor contamination levels. Figure 1 shows the extensive region where metals levels in the sediments range from 10 to 100 times control values and from which the exposed animals were taken. Waste discharges from Los Angeles City and County continue to add about 1400 tons of metals each year to the water in the concentrations shown in Table 1. About 10 percent of the discharged metals are attached to particulates that fall to the bottom near the outfalls. Most of the remaining 90 percent of the discharged metals are attached to very tiny particles and drift off to sea in very low concentrations. By any standard, the hatched region shown in Figure 1 has high levels of metals to which the exceptionally large number of animals in the region are exposed.

Each metal has certain special characteristics, some of which reduce the toxic threat. For example, high valence chromium (Cr +6) which is very toxic in laboratory tests, finds its way (from bathrooms and chrome plating

plants) into city sewer systems. However, in passing thru even a minimal treatment plant it is reduced to Cr +3. The latter, upon release into the sea, becomes a harmless hydroxide precipitate (Oshida, 1981).

The organic compounds of metals act quite differently from the inorganic forms. They are lipophilic and they seem to move about in the environment much the same as chlorinated hydrocarbons. This means they are potentially toxic and do increase in concentration with trophic level. Fortunately, the only metal whose organic form is present in sufficient quantity to be of concern is (methyl) mercury. The mercury in sediments around the Los Angeles County outfalls (rarely exceeds a concentration of 1 ppm) is over 99 percent inorganic mercury (Eganhouse, 1978). Measurement of mercury in sea animals taken from this region, where sediments are as much as 100 times background, show no increase of mercury in body tissues. The mercury in animals, unlike that in the sediments, is organic mercury (Eganhouse, 1976; Young, 1975). Fish caught both in coastal waters and far from any likelihood of human influence have (organic) mercury in their tissues in similar amounts at similar trophic levels. Therefore, it seems evident that

TABLE 1

Metal concentrations under natural conditions, in inputs, and resultant contamination. Unites are ug/liter unless otherwise indicated.

	NATURAL			CONTAMINATED INPUTS				RESULTANT CONTAMINATION		
	Sea water Calif. coast background	Control sediments 60 m depth Calif. coast	Santa Clara River runoff	Los Angeles River runoff	Primary effluent Los Angeles County 1979	Los Angeles County Outfall plume highest level (computed)	Los Angeles City (from 7-mile sludge outfall) average	Sediments (most contaminated station at Palos Verdes)	Sediments in canyon below sludge outfall	Average conditions along 60 m contour Santa Monica Bay
	mg/kg							mg/l	mg/kg dry wt.	
Cd	0.05	0.39	11.2	8.5	27	0.27	0.96	60.8	39	1.2
Cr	0.2	23.1	251	92	260	5.7	10.0	1317	646	73
Cu	0.2	9.1	173	108	220	4.0	13.2	782	512	32
Ni	1.0	12.2	220	70	210	2.0	3.4	107	79	20
Pb	0.02	6.6	166	550	150	1.7	2.42	537	180	18
Zn	0.5	42.2	512	980	690	12.0	21.0	2096	683	67
Hg	0.03	0.03*	—	0.9	1	0.006	0.25	4.0*	5.5*	—
Ag	0.01	0.38	4.8	1.5	19	0.08	0.72	18.1	46	2.9
	(Bascom, 1976)	(Word, 1977)	(SCCWRP, 1978)	(Young, 1980)	(Schafer, 1980)		(Schafer, 1980)	(Word, 1979)	(Bascom, 1980) *(Eganhouse, 1978)	(Word, 1979)



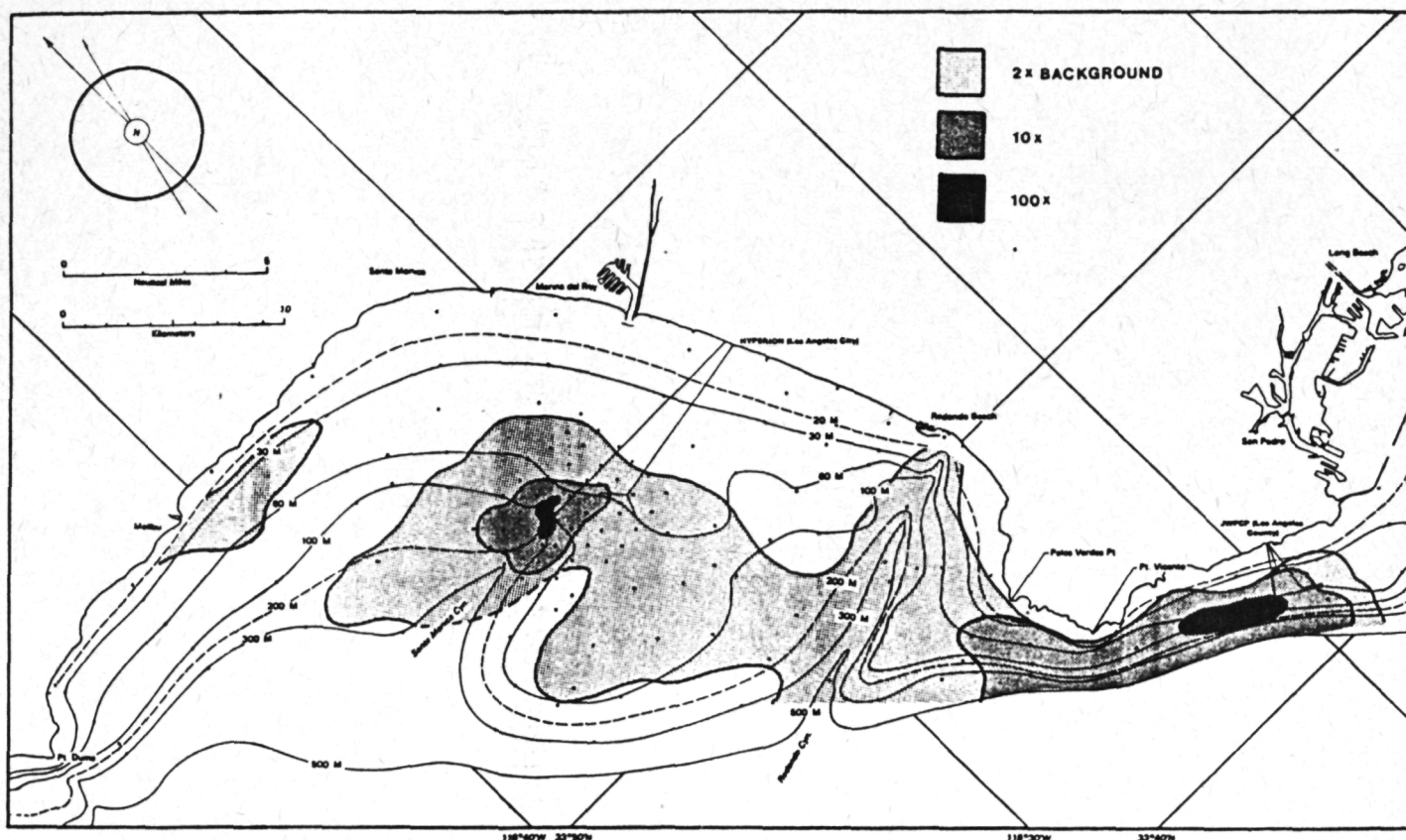


Figure 1. Concentrations of cadmium as multiples of background in the top 2 cm of sediments in Santa Monica Bay and off the Palos Verdes Peninsula showing the effect of 2 major outfall systems. Patterns for other metals were similar. All were approximately 10 to 100 times background in the hatched area from which our fish and invertebrate specimens were taken. (Hershelman, 1980)

the organic mercury measured in sea animals is normal and its presence has little or no relation to human waste disposal.

Organic mercury (methylmercury chloride) was, in the late 1950s, directly discharged into the sea at Minamata, Japan, where it caused very serious toxicity problems (D'Itri, 1972). Now the world has been warned, it seems unlikely that this relatively rare compound will be discharged again. This terrible experience of over two decades ago with very high levels of mercury that was discharged in the organic form should not prevent the use of the sea for disposal of wastes containing very low levels of inorganic mercury.

### TOXICITY TESTS

A comprehensive survey of marine toxicity data was made for the State of California that summarized data resulting from hundreds of chronic and acute toxicity tests for cadmium, chromium, copper, lead, mercury, nickel, silver and zinc (Klapow and Lewis, 1978). The acute toxicities given ranged over two to five orders of magnitude for each metal. For zinc and copper, which

are required by the enzymes of all animals, the toxicities given were very close to the natural levels in seawater. Conservative estimates based on these data were then multiplied by an "application factor" (generally 0.01) to arrive at state standards for discharge.

Most of the data given by Klapow and Lewis were concentrations reported to cause 50 percent mortality during laboratory exposure periods lasting one to four days. The authors liked the idea that the numbers were "standardized and qualitative" but noted that "sea water contains organic salts and dissolved organic matter in concentrations that may complex a significant fraction of an applied toxicant and alter its effects."

I submit that toxicity tests made in small laboratory tanks with filtered sea water and ionic solutions of metals, usually far above likely environmental levels, are invalid for the purpose of setting realistic standards. Such tests do not adequately replicate actual ocean conditions. Moreover, marine animals have evolved natural protective mechanisms to survive the variations in the amounts of metals present in sea water to which they have been subjected over geologic time. To be realistic, a test must permit animals to use the protection so developed.





## METALS IN THE FOOD WEB

Some small invertebrates at a low trophic level have slightly more (or less) metals in their bodies when they live in contaminated regions than similar animals that live in non-contaminated regions. Levels may reach to ten times that in controls, which is only a few parts per million. This increase may be caused by particles in the GI tract or by sorption of metals to external body parts rather than actual uptake into the animal's soft tissues. In any case, our laboratory has been unable to find any evidence of metals damage to these invertebrates in the environment. We do see shifts in animal assemblages for which there are many explanations, but even where there are high levels of metals in sediments near outfalls some species of benthic infauna proliferate and become very abundant, suggesting that a rise in metal body burden is not detrimental.

Table 2 compares metals levels in larger invertebrates taken in a contaminated region with those in two uncontaminated control regions (Young, 1978). As can be seen, the high and low values are randomly distributed between the three regions. We conclude from these and other such measurements that there is no rise in body burden of metals and therefore no contamination or toxicity in these animals.

For higher animals on which clean dissections of tissues can be made, the evidence is quite clear. In a very extensive investigation of contaminants in marine food webs for the National Science Foundation, it was shown that metals do not increase with trophic level (Young and Mearns, 1982; Young et al., 1980). Table 3 shows two of these food webs, one representing control conditions, the other using animals taken as close as possible to an outfall that is a substantial source of metal contamination (right side, Figure 1).

Eight metals and organic mercury were measured in numerous replicated samples of the widest possible range of trophic levels. Stomach analysis showed that the higher animals fed mainly on the ones below them in this list, confirming the trophic levels given. As the data

shows, the concentration of inorganic metals in muscle tissue of fish does not change substantially from low to high trophic levels or from control to contaminated environment. Dry weight levels are four or five times the wet weight level shown for muscle tissue. No amplification or biomagnification can be seen in these data.

The small invertebrates (copepods, mysids and decapod shrimp) have somewhat higher levels of metals than the fish. As mentioned before, it is possible that these metals are in the gut or sorbed externally and are not in soft tissues. Either way, they are consumed by the animals at the next trophic level whose muscle tissue concentrations do not increase.

Although inorganic mercury remains low at all trophic levels, organic mercury rises to levels of over one part per million in the muscle tissue of larger fish. As the data shows, there is no appreciable difference between the organic mercury in the control fish and those that live in the contaminated region. In fact, the concentrations are similar to those found in an open ocean food web several hundred kilometers west of Costa Rica in the open Pacific (Schafer, 1982).

## PROTECTIVE MECHANISMS IN SEA ANIMALS

Metals at trace levels continually enter the bodies of sea animals through the gut or gills and then, often by diffusion, become incorporated in body tissues or the blood stream. These metals are naturally present in seawater, plus additions from man's wastes. The excess metal in sea water may sometimes be in the form of free ions but it is more likely to be sorbed to tiny particles or organic materials or clays that drift far and settle very slowly. There are also metal excesses in sedimentary interstitial water and probably in the water immediately above the bottom into which metal ions escaping from sediments must diffuse. In any case, all sea animals are exposed to metals, some of which may be in ionic form. All animals studied to date are protected against this possible toxic threat in the following manner.

TABLE 2

Metals levels in invertebrates mg/kg dry edible tissues. (ND = not detected) (After Young, 1978). (— = not measured)

	Abalone Muscle			Scallop Muscle			Lobster Muscle			Sea Urchin Gonads		
	Near LA Co. outfall	Coastal control	Island control	Near LA Co. outfall	Coastal control	Island control	Near LA Co. outfall	Coastal control	Island control	Near LA Co. outfall	Coastal control	Island control
Cr	3.3	0.5	0.6	1.6	0.6	ND	ND	ND	ND	ND	<0.3	1.4
Cu	16.7	27.8	17.3	1.5	0.8	1.5	29.1	30.5	76.4	—	—	—
Ni	2.4	<	<	—	—	—	—	—	—	0.5	ND	<2.0
Pb	—	—	—	—	—	—	3.3	3.2	<2.0	1.2	ND	ND
Zn	20.5	32	24.4	49.5	60.8	38.8	28.5	36.4	52.5	—	—	—
Ag	0.4	1.7	0.4	0.5	0.5	0.8	—	—	—	0.3	0.6	0.5



TABLE 3

The non-increase of metals in muscle tissue with trophic level. Where a less than (<) is shown no signal was obtained so the detection limit of the AA machine was multiplied by the dilution (4). Inorganic mercury concentrations are total mercury less the benzene-extractable fraction shown in the organic column. (After young and Mearns, 1981).

## 3a. Open coastal Pelagic food web

	No. samples	Animals/sample	Trophic level	Organic Hg	mg/kg wet wt. Inorganic							
					Hg	Cd	Cr	Cu	Ni	Pb	Zn	Ag
Mako Shark	5	1	4.40	1.42	0	0.03	0.06	0.32	<0.04	<0.12	3.9	0.006
Thresher Shark	4	3	3.82	0.66	0	0.01	0.05	0.24	<0.07	<0.12	3.8	<0.003
Pacific Mackerel	5	10	3.54	0.09	0.02	0.07	0.03	0.37	0.12	<0.10	4.6	0.003
Bonito	3	10	3.80	0.20	0	0.01	0.04	0.34	<0.09	<0.12	3.5	<0.003
Sardine	5	10	3.01	0.01	0.04	0.02	0.04	0.25	<0.08	<0.15	3.5	<0.003
Anchovy	5	30	2.82	0.02	0.01	0.16	0.07	0.34	<0.04	<0.12	8.9	0.010
Zooplankton	3	>100	2.0	<0.01	<0.02	1.03	0.16	0.98	0.31	0.38	9.6	0.044

## 3b. Food web, within 3 km of Los Angeles County outfall

Scorpion Fish	5	2	4.53	0.22	0	<0.01	<0.02	0.18	<0.03	<0.06	2.3	<0.002
Spiny Dogfish	5	1	4.16	1.48	0.05	0.05	0.04	0.11	<0.02	<0.06	3.8	<0.002
White Croaker	5	10	3.36	0.06	0.01	<0.01	<0.01	0.24	<0.02	<0.06	1.9	<0.002
Dover Sole	5	10	3.52	0.02	0.03	<0.01	<0.01	0.15	<0.02	<0.06	2.7	<0.002
Sicyonia	5	15	3.33	0.05	0.02	0.01	0.16	5.3	<0.02	<0.06	11.8	<0.001
Mysids and Decapod Shrimp	5	>100	2.78	0.01	0.00	0.15	0.94	5.1	0.74	0.33	9.1	0.167

Zinc and copper are required by all animals for certain necessary enzymes. In order to maintain a reserve supply of these essential metals, a protein called metallothionein has evolved that sequesters excess copper and zinc entering certain tissues and holds it for future use. Thus the enzymes remain fully supplied with their metal needs. Other metals, such as cadmium, nickel, and inorganic mercury that are not essential to the animal's health also are sequestered by the metallothionein. This means that all metals, except those re-

quired immediately, are prevented by the metallothionein from reaching the "sites of toxic action" that include the enzymes and genetic materials.

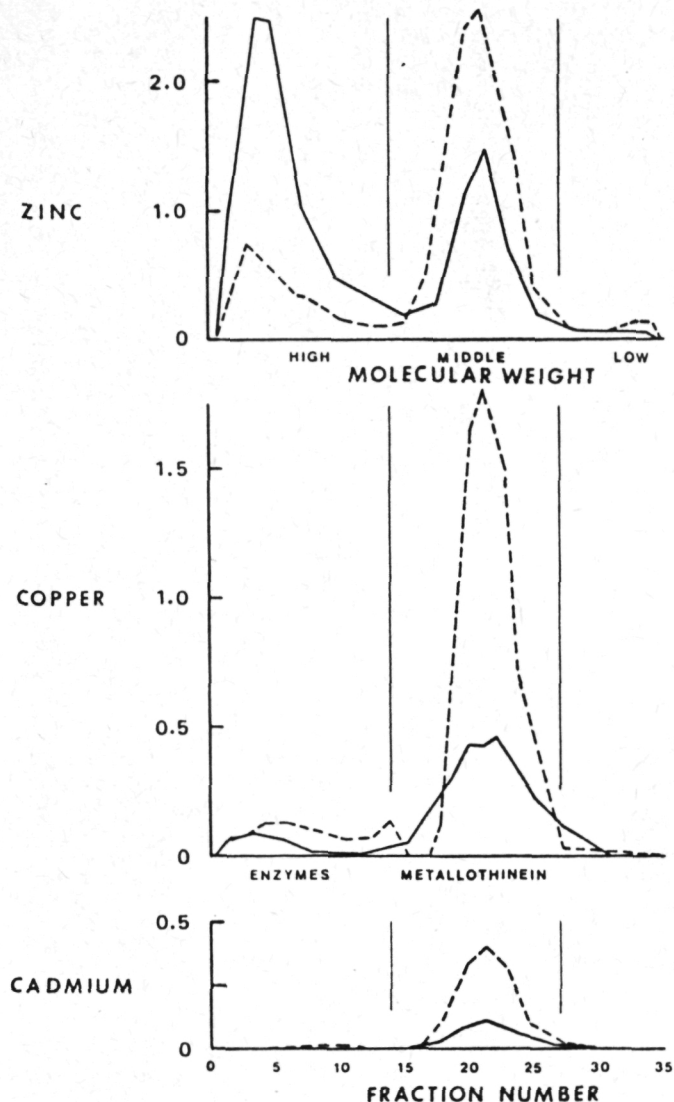
It is possible to determine whether this detoxification has operated effectively in sea animals exposed to high levels of metals by the following technique. Tissue from the animal or organ in question is homogenized, centrifuged, and run thru a 75,000 Sephadex column to separate the components into fractions (usually 35) accord-





ing to molecular weight (Brown et al., 1977). Each of these fractions is then analyzed for trace metals to determine how the metals are distributed.

Because the enzymes and some genetic materials (e.g., mRNA) fall in the high molecular weight fraction and the metallothionein is in the middle molecular weight fraction, a plot of metal levels against molecular weight fraction shows that animal is protected against excess metals. Figure 2 gives examples from the extensive investigations by David Brown into this matter. It shows



**Figure 2.** Distribution of metals in the molecular weight fractions of cytosol in animals from a contaminated area near the Los Angeles County outfall. The dashed line shows the levels in the livers of fish (10 white croaker); the solid line shows the levels in the gonads of 10 sea urchins (composites). The amounts of copper and zinc in the high molecular weight fraction are needed by the enzymes. The unneeded copper and zinc plus all the cadmium are bound to metallothionein and prevented from reaching the "sites of toxic action." Numbers on left scale are in parts per million. Note scale differences.

the location of the Zn, Cu and Cd in the gonadal tissues of a sea urchin and the liver of a white croaker taken from the most contaminated area shown in Figure 1.

It can be seen that except for the normal enzymatic requirements for zinc (about 2.5 ppm) and copper (about 0.2 ppm) all the excess metal is in the middle molecular weight fractions attached to metallothionein. The same is true of the cadmium which was used to represent other metals not needed by the body tissues (Brown et al., 1980). Once the need for the two essential metals is satisfied, other metals (e.g., Cd) are excluded from the high molecular weight fraction which contains the "sites of toxic action." This is the protective mechanism (Brown, 1978).

Metallothionein has been found in all animals so far tested, including oligochaetes, white mice and humans, as well as the sea animals named above (Jenkins, Brown et al., 1982a). It does not exist in all tissues. For example, in fish it is present in liver, kidney and intestinal lining, but not in muscle tissue. In mussels, all tissues, with the exception of the adductor muscle, have metallothionein.

If an animal is exposed to very high concentrations of metals in ionic form, as in a laboratory test, it is possible to exceed the capacity of metallothionein to sequester metals; if so, a "spillover" of metals from the middle molecular weight fraction into the high molecular weight fraction is observed. However, after analyzing many sea animals taken from coastal waters of known high-metal contamination, the Coastal Water Research Project has not seen evidence of spillover (Jenkins, Brown et al., 1982b). We theorize that spillover and toxicity is almost always the result of acute laboratory bioassays that kill or damage animals by dosing them with an instantaneous high concentration of some metal ion. If the same level of metal exposure is attained by gradual increase in concentration, so that there is time for the animal to respond by making metallothionein, the animal may survive without damage.

As indicated in Figure 2, several metals can be simultaneously sequestered by the same metallothionein pool. Other measurements by our laboratory have shown six metals in the same pool and we have no reason to believe that more would not be taken up in the same way.

## CONCLUSION

In this paper I have shown that:

- Large quantities of metals have been discharged into the open coastal waters of California where the 10 percent of them that are attached to rapidly-settling particles have formed substantial areas of contaminated sediments with levels 10 to 100 times background.

- Small invertebrates exposed to these high concentra-



tions sometimes show an apparent increase in the body burden of metals, but larger invertebrates exposed to the same sediments show no increase of metals in their tissues.

- Toxicity tests made to determine acute or chronic toxicity (using LC 50 percent as the criterion) give a wide range of answers for each metal. Such tests do not accurately replicate metal exposure in actual environmental conditions.

- Inorganic metals do not move upward through the marine foodwebs.

- Inorganic metals in fish muscle tissue do not increase above natural levels regardless of the exposure, so there is no hazard to persons who eat them.

- The toxic (methylated) form of mercury does move up through the food web but its presence (except in rare circumstances) is natural and not the result of man's ordinary wastes.

- Animal cells generate a protective protein that prevents excessive concentrations of metal from damaging their enzyme systems or genetic materials, except in the special conditions of very high free ion concentrations in the laboratory. When the amount of metal in each molecular weight fraction is examined in animals taken from highly contaminated environments it is evident that undesirable metals have not reached the sites of toxic action.

Therefore, I repeat the opening thesis: Metals in the sea, even in regions of high concentration, are not a hazard to sea animals or to persons who eat those animals.

**Note:** Original version of paper presented at IAWPR, London, September 1981; published in *Water Science Technology*, Vol. 14, pp. 41-52, London, 1981.

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