

## COMPARISON OF SEDIMENTS AND ORGANISMS IN IDENTIFYING SOURCES OF BIOLOGICALLY AVAILABLE TRACE METAL CONTAMINATION

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**Abstract**—Sediments and an indicator organism (*Macoma balthica*, a deposit-feeding bivalve) were used to assess the relative importance of secondary sewage, urban runoff, a landfill containing metal-enriched ash wastes and a yacht harbor in contributing to Ag, Cu and Zn enrichment in South San Francisco Bay. Spatial gradients in sediments and organisms showed Cu and Ag enrichment originated from sewage discharge, whereas Zn enrichment originated from both sewage and urban runoff. Elevated concentrations of Cu in the sediments of the yacht harbor resulted from a high abundance of fine particles. The biological availability of Cu, Ag and Zn did not coincide with metal enrichment in sediments. The availability of Cu and Ag was greatest nearest the sewage outfall and greater in winter and spring than in summer. The availability of Zn in urban runoff appeared to be lower than the availability of Zn associated with sewage.

### INTRODUCTION

Trace metals may enter an estuary from a variety of natural and anthropogenic sources, including industrial or domestic sewage, storm runoff, leachates from landfills, shipping and boating activities and aerial deposition. The relative importance of such sources has been assessed on the basis of mass discharge (e.g. Russell *et al.*, 1982). However, comparisons of the contributions of such sources to pollutant concentrations observed *in situ* are rare.

Sediments and organisms both have been used to monitor or identify trace metal contamination in aquatic environments. Both methods have limitations which many studies have not considered. Sediments integrate metal discharges through time, and are the most concentrated physical reservoir of metals in aquatic systems. However, in attempting to identify sources of contamination of sediment, consideration must be given to physicochemical characteristics which may control trace metal concentrations. These include grain size, iron concentration and concentrations of organic carbon, all of which may be interrelated (Bradford and Luoma, 1980). Furthermore, total metal concentrations in sediments do not necessarily reflect concentrations which are available to biota.

Organisms have been used in comparative monitoring of pollutants in different systems (Bryan, 1979; Goldberg *et al.*, 1978) and to locate sources of contaminants (Young and Jan, 1979; Widdows *et al.*, 1981; Popham *et al.*, 1980; Jensen *et al.*, 1981). However, biomonitoring also has limitations. Growth, age, seasonal physiological cycles, acclimation and genetic adaptation may influence metal concentrations or metal impacts in organisms in ways that have not been totally resolved. Nevertheless,

metal uptake by organisms remains the only method available for estimating biologically available metal concentrations in natural systems; *in situ* bio-monitoring is the only method which takes into account all changes in the receiving water quality which may affect the toxicity to the organisms studied.

Our objective in this study was to compare the relative value of sediments and organisms *in situ* in distinguishing among several potential sources of trace metal input in a study area in South San Francisco Bay.

### METHODS AND MATERIALS

#### Study area

Samples were collected along 7 km of the shoreline of South San Francisco Bay (Fig. 1) where previous studies had shown unusually high concentrations of Cu and Ag in organisms (Luoma and Cain, 1979; Bradford and Luoma, 1980). Sampling stations were chosen to permit differentiation among four potential sources of contamination within the transect: (1) a sewage treatment plant outfall; (2) a shoreline landfill; (3) a small stream which receives urban storm runoff; and (4) a small enclosed yacht harbor. Samples were collected in July 1979, January 1980 and March 1980, in order to include any influence of the substantial seasonal variability in metal concentrations in organisms shown to occur by Luoma and Cain (1979).

#### Procedures

*Macoma balthica* (a deposit-feeding bivalve) and surficial sediments (Luoma and Bryan, 1981) were collected from intertidal stations at low tide. At least 20 individual animals, representing the size distribution of the population, were collected at each site then placed in clean seawater for 48 h to depurate. Three to five similar-sized animals (within 1 mm shell length) were pooled to yield five to seven composite samples from each collection. Animals were dissected into soft tissues and shell, and the soft tissues were refluxed in concentrated nitric acid. Upon complete digestion, the acid was evaporated and the sample was reconstituted in 25% HCl (Luoma and Cain, 1979) for

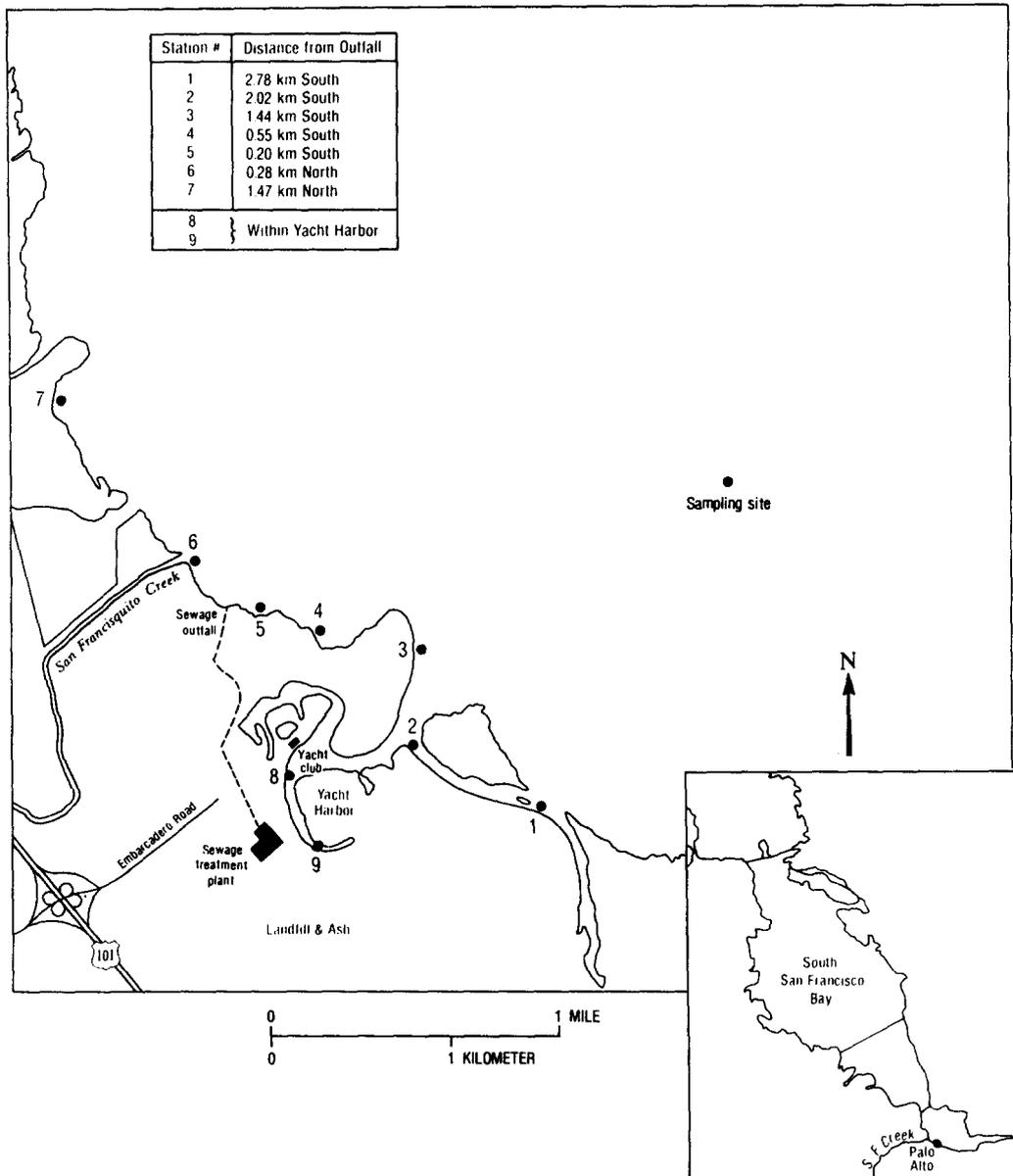


Fig. 1. Location of sampling stations.

analysis of Cu, Ag and Zn by flame atomic absorption spectrophotometry (AAS). Previous work indicated no significant matrix effects.

Sediments were scraped from the upper 5 mm of sediment exposed at low tide, sieved through 100  $\mu\text{m}$  polyethylene mesh (Luoma and Bryan, 1981; Thomson *et al.*, 1980), refluxed in 2:1 nitric-sulfuric acid, then analyzed for Cu and Zn by flame AAS; Ag analyses were done by graphite furnace AAS. These analyses are used as a measure of "total" metal. [Although this method removes more than 95% of Ag, Zn and Cu from sediments, some highly crystalline metal forms are not extracted (G. W. Bryan, The Laboratory, Plymouth, U.K., unpublished data).] Ash from the landfill was treated similarly.

Total carbon in the sediments was measured by total combustion in a carbon analyzer. Inorganic carbon was measured as the acid-soluble fraction—sediments were digested in hot phosphoric acid, the carbonates converted to  $\text{CO}_2$ , and inorganic carbon measured in a carbon analyzer.

Total organic carbon (TOC) was determined by difference. Salinity was measured, using a refractometer, in both surface pools on the mudflats and in body water from freshly collected clams.

Particle size distribution was estimated by determining the percentage of presieved (<100  $\mu\text{m}$ ) particles which remained suspended after 15 min in an Andreasen pipette apparatus (defined by Stokes' Law calculations as "percent particles less than 14  $\mu\text{m}$ "). Some sediments had a tendency to flocculate during the particle size analysis, causing rejection of several analyses in the summer samples.

#### Sources of metal

**Sewage treatment plant.** At the time of the study, the Palo Alto sewage treatment plant was employing a secondary-treatment activated-sludge process to treat wastes. The discharge from the plant was released into a channel in a nearby salt marsh from whence it flowed directly onto the mudflats of the Bay without mechanical diffusion. Flow

rates averaged 28 MGD in 1979 and 31 MGD in 1980. Metal loadings in effluent, calculated from quarterly filtered, composited metal analyses done by the Palo Alto sewage treatment plant, ranged from 6–21 kg day<sup>-1</sup> Cu, 5–8 kg day<sup>-1</sup> Zn and 0.4–1.1 kg day<sup>-1</sup> Ag during the study period. Fluctuations in loadings did not follow any definable pattern. Removal efficiencies for Cu, Zn and Ag ranged, respectively, from 68–91%, 70–83% and 50–87%, between 1978 and early 1980 (calculated from data from Doris Maez, Palo Alto sewage treatment plant, personal communication). The Cu and Zn efficiencies are comparable to other secondary plants (Brown *et al.*, 1973; Davis and Jacknow, 1975) (no Ag data were available).

**Landfill.** Ash originating from incinerated sludge at the sewage treatment plant has been buried in a shoreline landfill near the plant for about 9 years at a rate of about 1200 tons (10<sup>6</sup> kg) per year (Steve Hayashi, Palo Alto sewage treatment plant, personal communication). The ash supports a profitable metal-extraction operation for silver and gold. We measured concentrations of Ag as high as 900 µg g<sup>-1</sup>, Cu as high as 17,000 µg g<sup>-1</sup> and Zn as high as 4630 µg g<sup>-1</sup> in dry ash. However, we found no detectable leaching in experiments with freshwater and seawater. Metals also were below detection in water from a freshwater pond near the landfill.

**Urban runoff.** San Francisquito Creek carries runoff from a large expanse of urban area into the Bay. To assess solute metal contributions from this source, water samples were collected from the creek both early and late in the rainy season. The replicate 50 ml samples were filtered (0.45 µm), acidified with 0.5 ml HNO<sub>3</sub>, and analyzed for Zn and Cu by graphite furnace AAS (Ag is not expected to be enriched in urban runoff). Rainfall occurred only between December and May during this study. Concentrations of Cu were less than 2 µg l<sup>-1</sup> in samples collected in April 1980 and May 1980. The sample taken in December 1980, following the season's first two rainfalls, had 7.8 µg l<sup>-1</sup> Cu. Concentrations of Zn were less than 0.1 µg l<sup>-1</sup> in the stream in April and May 1980. In December 1980, a Zn concentration of 53 µg l<sup>-1</sup> was found in replicate samples.

**Yacht harbor.** Antifouling paint on ship and boat bottoms is high in Cu and has been associated with Cu contamination in dockyards and yacht harbors (Young *et al.*, 1979). Thus, Cu contamination outside the mouth of Palo Alto Yacht Harbor could originate from the harbor itself. However, actual mass loading to the system from this source cannot be calculated.

## RESULTS

### Sediments

Concentrations of Cu and Ag in the sediments were high at Stas 4 and 5, the first stations south of the sewage outfall (Fig. 2a, b). There was less spatial variation in zinc concentrations (Fig. 2c) than in copper or silver. Zinc concentrations were generally higher north of the outfall than south. In April, the mean Zn concentration was elevated at the site nearest the creek. However, the range of two replicate samples was larger at that site than at any other site in the study, perhaps because increased inhomogeneity is an effect of the street runoff coming from the creek. Zinc concentrations were an average of 37% higher in January (the height of the rainy season) at all stations south of the Creek than in April or July, further suggesting substantial Zn impact from urban runoff. Cu concentrations were higher in January than in April or July, although the

source of the Cu appeared to be the sewage outfall. Ag concentrations differed little with season.

Concentrations of Cu were also high in sediments from the yacht harbor (Sta. 8) and from near the landfill (Sta. 9) in January. However, assessment of the significance of these enrichments required consideration of other characteristics of the sediments.

The largest percentage of particles less than 14 µm occurred in January at all stations (55–76%). Percentages of fine particles were less in April (22–39%) and July (18–25%). Particle size also varied spatially. Fine particles were more abundant at Stas 2 and 7, and in the semi-enclosed yacht harbour (Stas 8 and 9).

A significant relationship between percent particles less than 14 µm and total organic carbon was observed in summer and spring (when flocculation did not occur in the test) (Fig. 3). The fine "winter" sediments were low in carbon relative to particle size compared to the spring (March) and summer (July) sediments. This seasonal difference is observed throughout San Francisco Bay and may be a result of terrigenous sediment input accompanying the first winter storms.

Compared to Stas 6 and 7 (north of the outfall), the stations south of the outfall all were enriched in Cu and Ag with respect to organic carbon (e.g. Fig. 4), and fine particles. Station 5 (all seasons) and Sta. 4 (summer) show the greatest enrichment. Although the yacht harbor samples (Stas 8 and 9) showed high concentrations of Cu and Ag, they were not as enriched per unit TOC, or per unit particle size as Sta. 5; they were more similar to samples from the stations further south of the outfall (Fig. 4). Concentrations of Zn per TOC were slightly enriched at San Francisquito Creek in the spring, but enrichment at the creek was not as evident as when only concentration was considered. The higher concentrations of zinc found in the winter appeared to be at least partly the result of a greater abundance of fine particles.

### Effects of body size

When clams are used as an indicator of metal enrichment, comparisons of concentrations between sites may be affected by the sizes of organisms collected. It has been shown that body size (length) generally correlates with Cu and Ag (but not Zn) concentrations in tissues within size classes of *M. balthica* (Strong and Luoma, 1981). In this study, Cu and Ag concentrations among different size classes were most similar in the winter (see Fig. 5a, b, for example). Concentrations among different size classes diverged in the spring due to "tissue dilution" of metals in the smaller animals caused by their faster growth (Strong and Luoma, 1981), and were again similar in the summer, probably due to more rapid uptake by the smaller animals compensating for the longer accumulation time of the older, larger animals (Strong and Luoma, 1981). However, all size classes generally followed similar spatial trends.

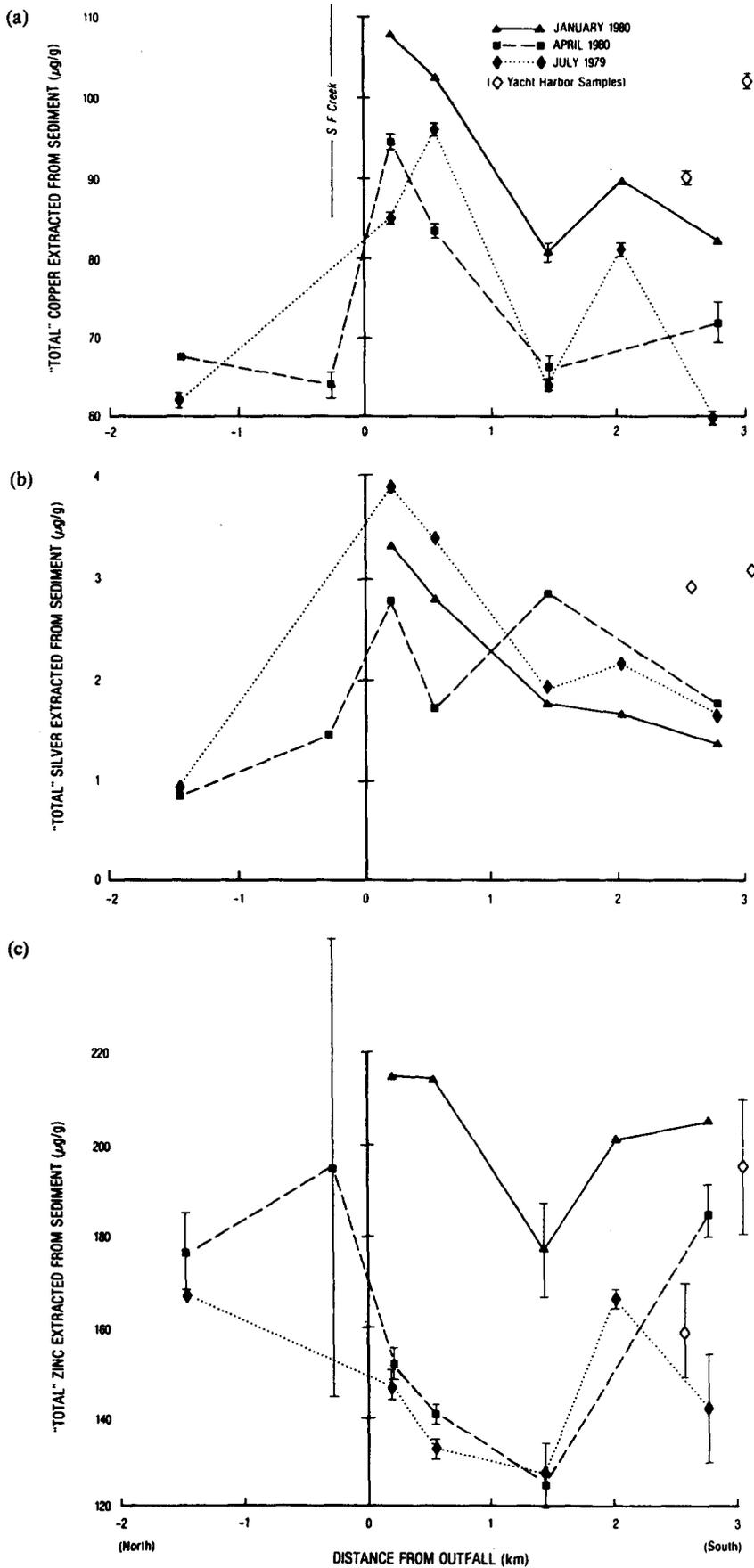


Fig. 2. Concentrations of metals in sediments at sites along three transects. Bars represent the range of two replicate samples for copper and zinc. (a) Copper; (b) silver; (c) zinc.

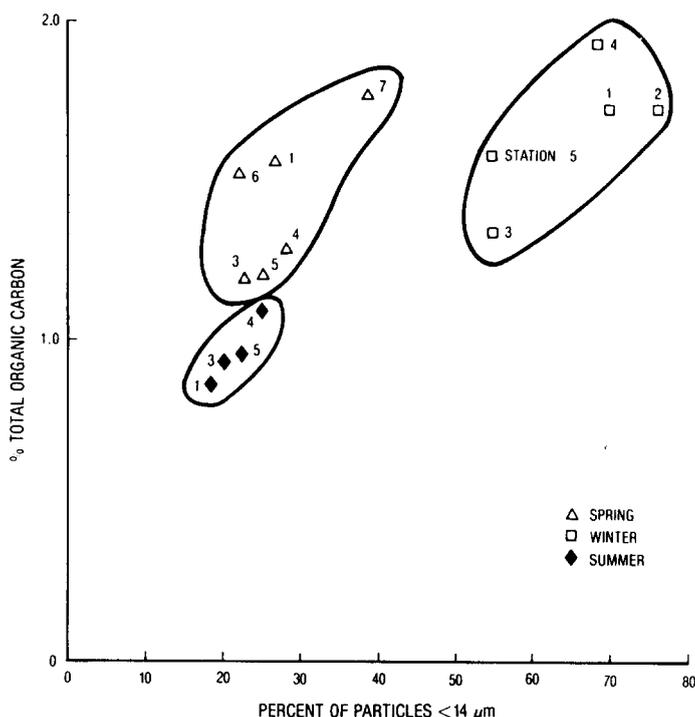


Fig. 3. Per cent of particles < 14 μm vs per cent total organic carbon for sediments from three transects. In spring and summer, there is a significant correlation ( $r = 0.778, P < 0.01$ ); in winter, the relationship is not statistically significant ( $r = 0.719, P > 0.05$ ).

*Metals in organisms—Cu and Ag*

The highest Cu and Ag values in clams were found in the winter samples (Fig. 6a, b). The highest metal concentration occurred in clams from Sta. 5. In January 0.1% of the dry body weight of these animals was copper. As occurred with sediment enrichments, Cu and Ag in clams declined much less rapidly to the south than to the north of the outfall. Clams sampled within the Palo Alto yacht harbor or at Sta. 6 showed no enrichment in Cu or Ag compared to those at the other sites.

Although patterns of temporal and spatial changes were similar in sediments and organisms, the Cu and Ag concentrations in the organisms were substantially greater than the concentrations in the sediments. For example, at Sta. 5, Cu in *Macoma balthica* was 251, 1084 and 811 μg g<sup>-1</sup> for summer, winter and spring, respectively; Cu in sediments was 85, 108 and 95 μg g<sup>-1</sup>. Typical “background” values of 30–50 μg g<sup>-1</sup> Cu were found in *M. balthica* in the Bay (Luoma, unpublished data).

At Sta. 5, Ag in *Macoma balthica* was 32, 133 and 110 μg g<sup>-1</sup> (summer, winter and spring), and Ag in sediments was 3.89, 3.33 and 2.78 μg g<sup>-1</sup>. “Background” Ag values in *M. balthica* in the Bay average below 1 μg g<sup>-1</sup> (Luoma, unpublished data).

*Zinc*

Zinc concentrations in the organisms (Fig. 6c) also were higher in winter and spring than in the summer.

Spatially, Zn was the highest in the organisms closest to the outfall (Sta. 5) and at the mouth of San Francisquito Creek (Sta. 6). At these sites, concentrations in the clams were generally double the concentration in the sediments, and 4 times “background” Zn in *M. balthica* in the Bay (Luoma, unpublished data).

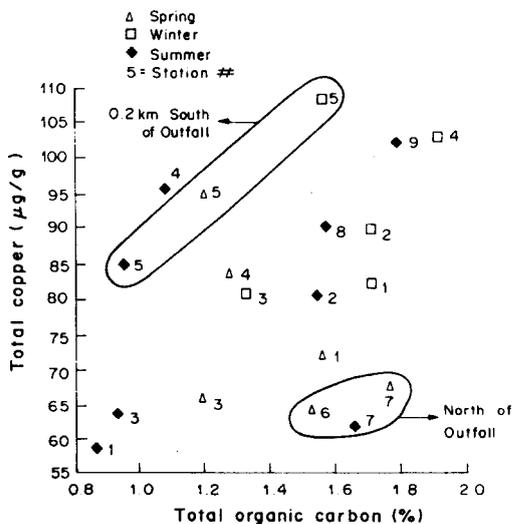


Fig. 4. Per cent total organic carbon vs total copper for sediments from three transects.

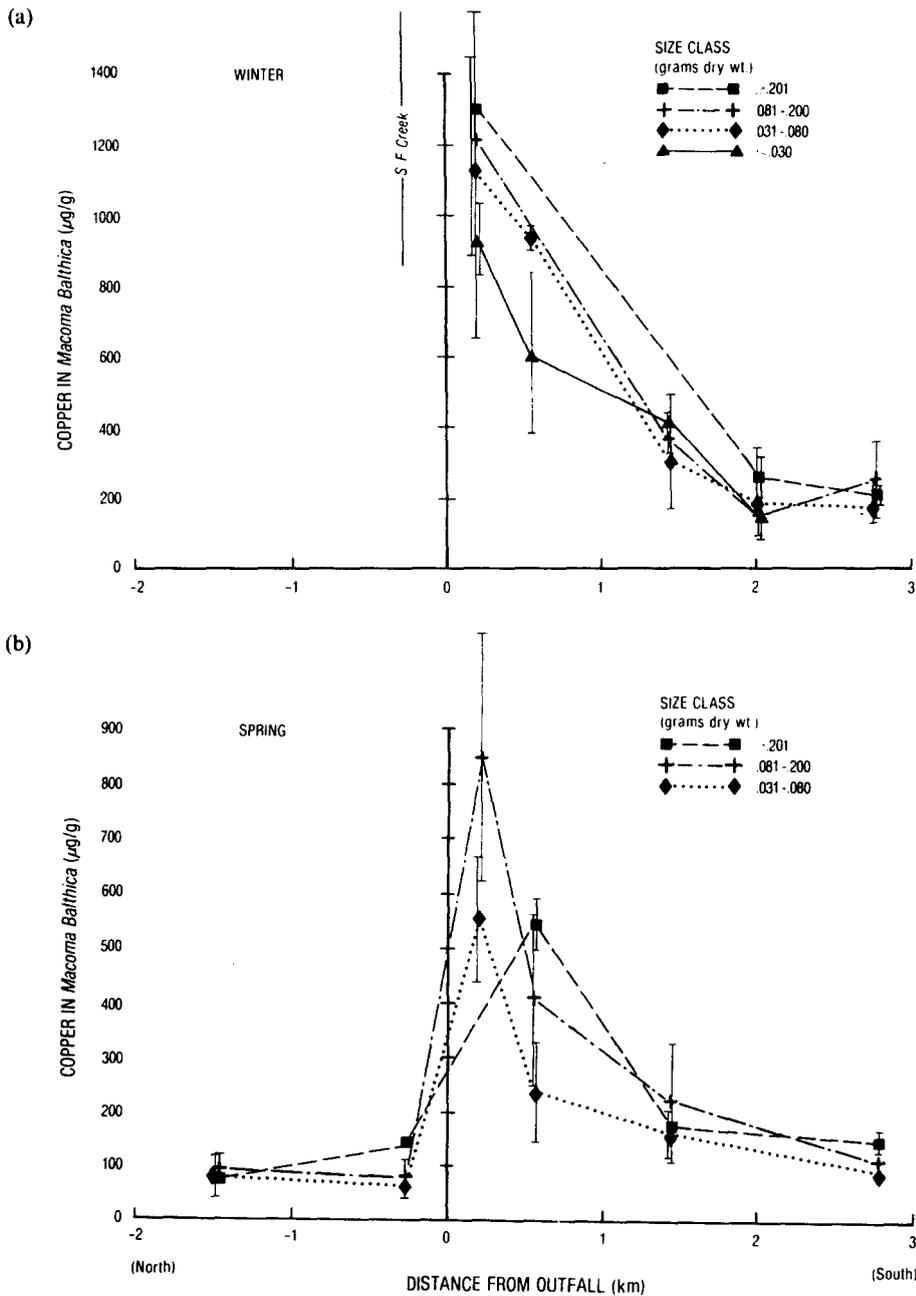


Fig. 5. Copper in *Macoma balthica* by size class for (a) winter and (b) spring transects. Error bars ( $\pm 1$  SD) are shown.

#### *Effects of discharge and sediment metal concentrations on organisms*

Total Cu in the sediment and *M. balthica* at Sta. 3 south of the outfall was measured nearly monthly for a period of 4 years. Figure 7(a) shows that Cu in the surface sediments clearly reflected periods of increased Cu input from the sewage plant. However, fluctuations in Cu in clams (Fig. 7b) did not always follow fluctuations in discharged Cu or sediment Cu, despite the strong suggestion from spatial differences that the sewage effluent was the source of Cu for *M. balthica*.

To assess the influence of processes other than sediment concentration on the availability of metals to *M. balthica* the difference between sediments and organisms (*B*) was calculated using the ratio

$$B = C_o / C_s \quad (1)$$

$C_o$  is metal concentration in the organism, and  $C_s$  is metal concentration in sediment.

Spatial changes in sediment-metal concentration were not linearly reflected in the animals. The difference between sediment and clams for all metals was greatest nearest the sewage outfall in January

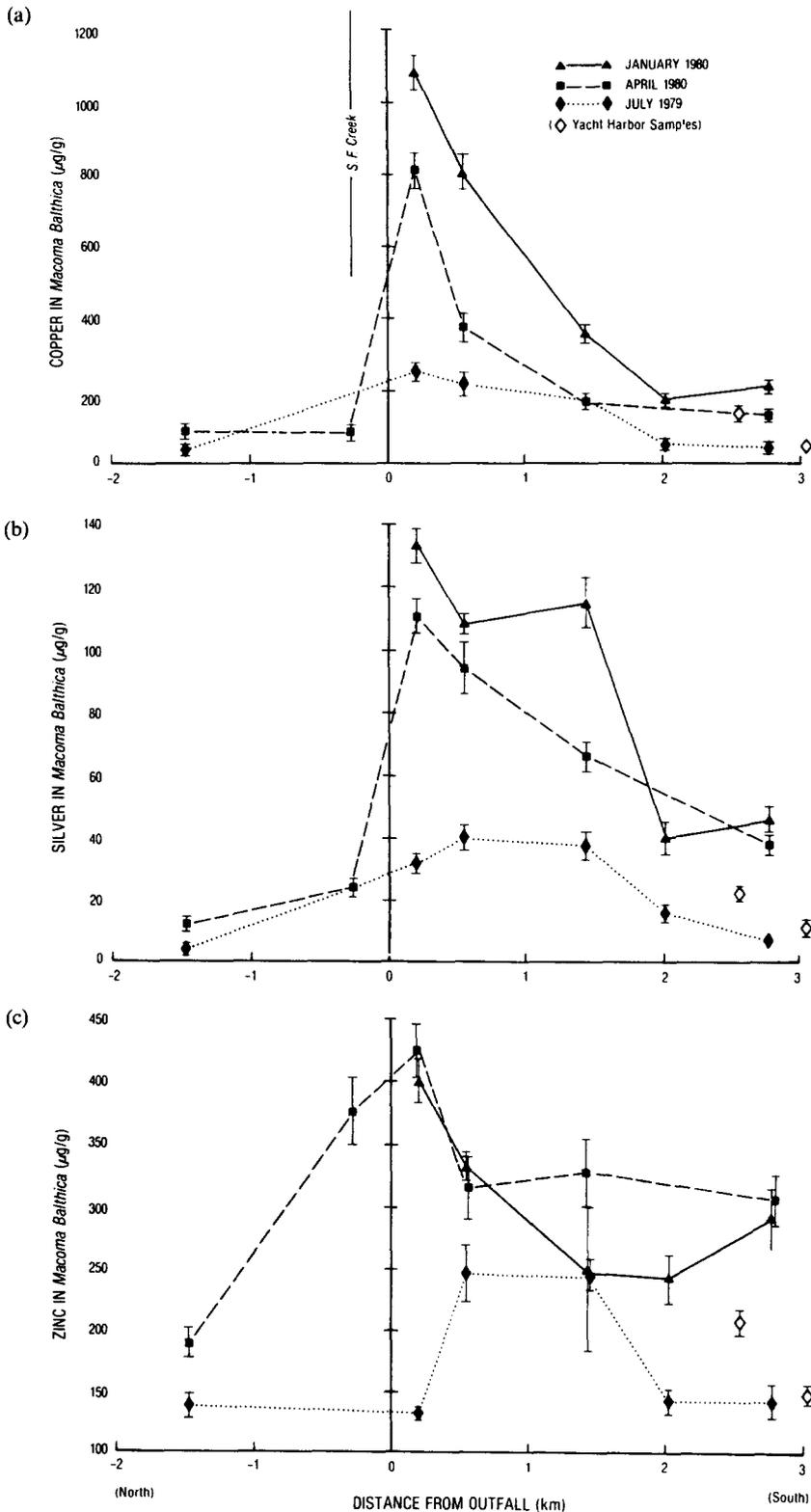


Fig. 6. Concentrations of metals in *Macoma balthica* at sites along three transects. Error bars ( $\pm 1$  SD) are shown. (a) Copper; (b) silver; (c) zinc.

and April, and was greater in winter and spring than in summer (Fig. 8). The creek appeared to be a source of Zn for both the sediment and the animals, which

would be expected from studies which show Zn to be present in urban runoff (Pitt and Amy, 1973). However, *B* was 1.93 at the creek and 2.80 near the sewage

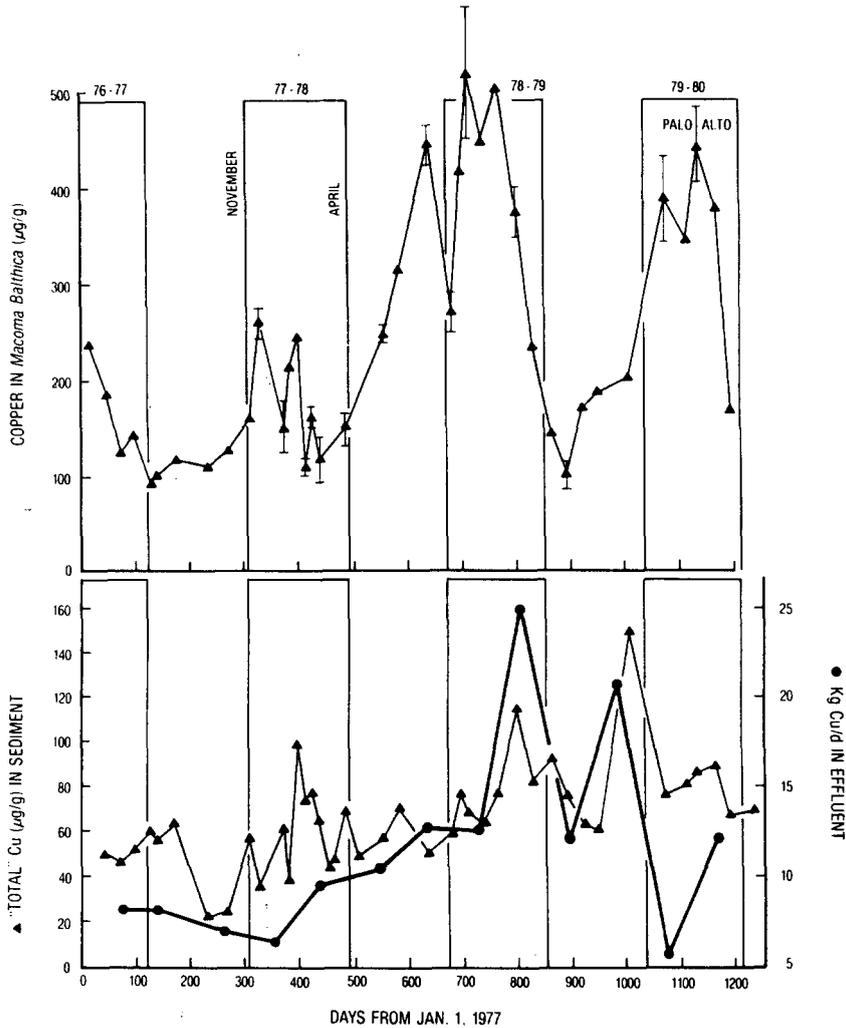


Fig. 7. Copper in sediments and organisms compared to copper in effluent. Error bars ( $\pm 1$  SD) are shown for clams.

outfall. Although the Zn concentration in the sediment was highest near the creek, Zn concentrations in the animals were similar at the creek and the outfall, indicating Zn availability to *M. balthica* was greater in sediments influenced by sewage discharge than in sediments influenced by storm runoff.

Salinity varied little among stations at any one sampling (<15%), suggesting salinity had little influence on the metal differences. Other results also show that seasonal differences in metal availability do not correspond closely with seasonal changes in salinity in this area (Luoma *et al.*, unpublished data).

#### SUMMARY AND CONCLUSIONS

Despite the use of secondary treatment, enrichment of Cu, Ag and Zn in *Macoma balthica* at Palo Alto appears to be due primarily to input from a sewage treatment plant. Yücel and Russell (1980) estimated that storm runoff contributes 33% of the "total equivalent metals loading" (based on relative tox-

icities of different metals) to the entire Bay, with higher concentrations in runoff from urban areas than from non-urban areas. Although detectable concentrations of Zn originated from urban runoff in our study area, runoff was not a detectable source of Cu or Ag to the organisms or sediments in comparison to sewage effluent. Compared to metal input from the sewage plant, the leaching of metals from the landfill also did not contribute significant amounts of sediment-bound or biologically available metals. The accumulation of fine sediments in the yacht harbor led to elevated sedimentary concentrations of Cu, but not to Cu enrichment in *M. balthica*.

Peaks of Cu discharge in sewage effluent are closely reflected in sediments, indicating surface sediments were a sensitive indicator of temporal changes in metal discharges. When interpreting spatial gradients in sediments, however, it was essential to consider sediment characteristics which affected trace metal concentrations.

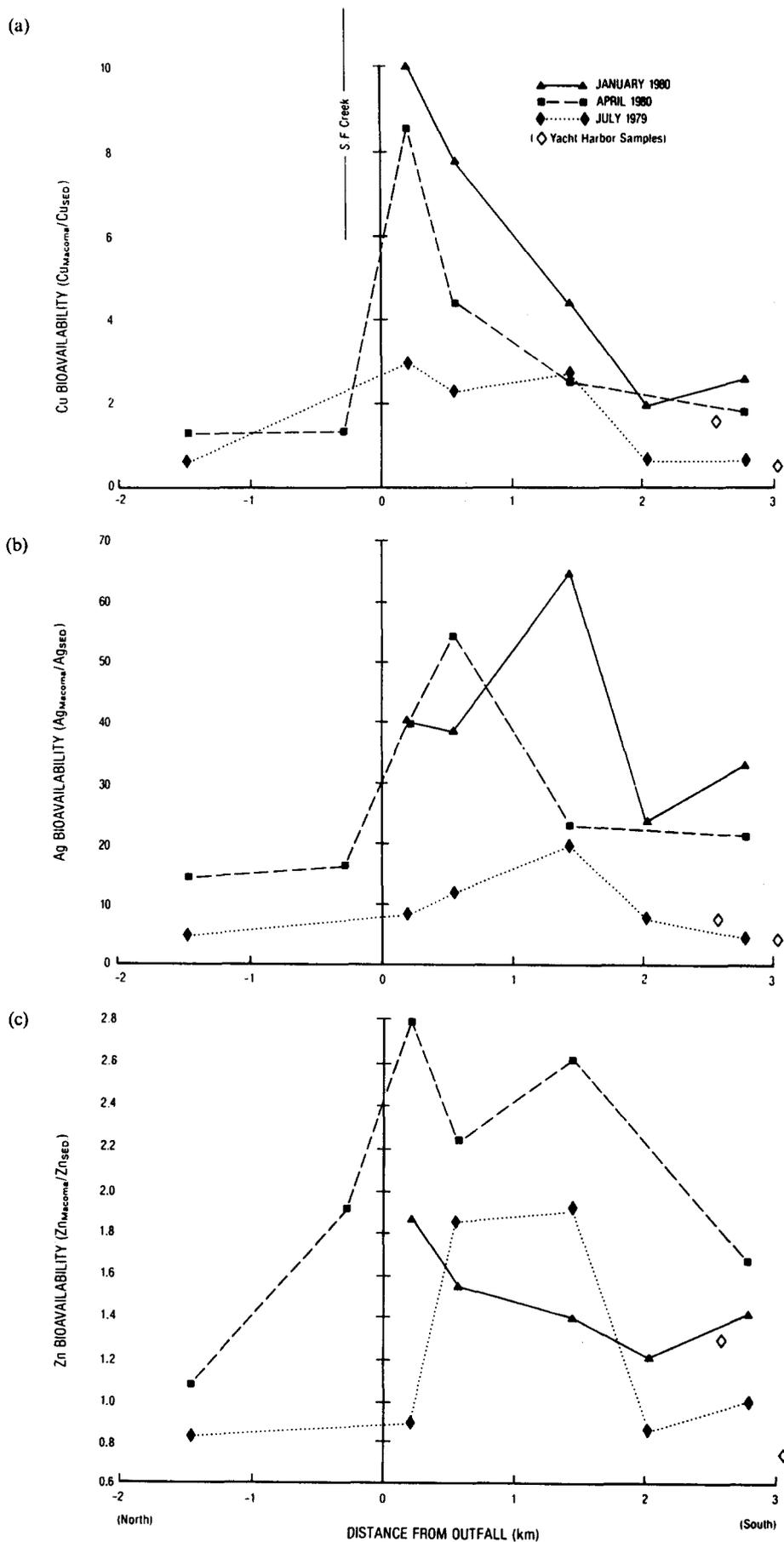


Fig. 8. Metal bioavailability to *Macoma balthica* (concentration in organisms/concentration in sediment) at sites along three transects. (a) Copper; (b) silver; (c) zinc.

Although total concentrations of some metals in sediment may be generally correlated with concentrations in organisms (Luoma and Bryan, 1978, 1979, 1982), predictive relationships are rare. Metal exposure is not the only process controlling metal bioaccumulation (Luoma and Jenne, 1977; Sunda and Guillard, 1976; Luoma, 1983). Physicochemical reactions in receiving waters (which could vary with time) can affect bioavailability, although precise influences are often poorly understood (Luoma, 1983). Neither sediment-metal concentrations nor discharge concentrations were reliable indicators of the bioavailable fraction of the metals in our study. Either the availability or the solute/sediment ratio of all those metals appeared to be greatest nearest the sewage outfall. Urban runoff was a source of Zn, but the availability of Zn in urban runoff appeared to be lower than Zn availability in areas affected by sewage. Where metal concentrations in sediment were elevated simply because of the presence of finer particles, no elevation of metal levels in clams occurred.

If organisms are to be used to detect or differentiate sources of pollution, sampling sites must be chosen carefully, especially in water bodies where circulation patterns are poorly known. Popham *et al.* (1980) found major changes in Cu, Zn and Pb concentrations in the mussel *Mytilus edulis* within 1 km of a source. McGreer (1982) reported a similar sharp gradient in metal concentrations in *M. balthica* near a small sewage outfall. Near Palo Alto, Cu and Ag concentrations in *M. balthica* declined more than six-fold within several hundred meters north of the sewage outfall, but extended for several kilometers south of the outfall. The hydrodynamics of South San Francisco Bay are not well established, but it is generally accepted that non-tidal, residual currents are not strong in the slowly circulating, semi-enclosed terminus of South Bay (Conomos, 1981). Nevertheless, the distinct, consistent pattern of metal distribution suggests an important influence of hydrodynamics, with an indication of a consistent net southward transport. This consideration is noteworthy in that a sample taken a short distance north of the Palo Alto outfall would not reflect an input from that source of contamination.

In this study, sediment analyses were useful for verifying the presence, and the source, of trace metal contaminants, proving that trends in animals were not the result of purely physiological processes. However, the biological availability of the metal contaminants was not related solely to the concentration of metal discharged or present in the sediment. Compared to sediments, organisms exhibited greater spatial sensitivity, a greater ability to concentrate metals, and were the more reliable tool for identifying sources of biological contamination. However, the most information can be obtained by considering both sediments and organisms as indicators.

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