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TEMPORALLY INTENSIVE STUDY OF TRACE METALS IN SEDIMENTS AND BIVALVES FROM A LARGE RIVER–ESTUARINE SYSTEM: SUISUN BAY/DELTA IN SAN FRANCISCO BAY

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ABSTRACT

Distributions in time and space of Ag, Cd, Cr, Cu, Pb and Zn were determined in fine-grained sediments and in the filter-feeding bivalve *Corbicula* sp. of Suisun Bay/delta at the mouth of the Sacramento and San Joaquin Rivers in North San Francisco Bay. Samples were collected from seven stations at near-monthly intervals for 3 years. Aggregated data showed little chronic contamination with Ag, Zn and Pb in the river and estuary. Substantial chronic contamination with Cd, Cu and Cr in Suisun Bay/delta occurred, especially in *Corbicula*, compared with the lower San Joaquin River. Salinity appeared to have secondary effects, if any, on metal concentrations in sediments and metal bioavailability to bivalves. Space/time distributions of Cr were controlled by releases from a local industry. Analyses of time series suggested substantial inputs of Cu might originate from the Sacramento River during high inflows to the Bay, and Cd contamination had both riverine and local sources. Concentrations of metals in sediments correlated with concentrations in *Corbicula* only in annually or 3-year aggregated data. Condition index for *Corbicula* was reduced where metal contamination was most severe. The biological availability of Cu and Cd to benthos was greater in Suisun Bay than in many other estuaries. Thus small inputs into this system could have greater impacts than might occur elsewhere; and organisms were generally more sensitive indicators of enrichment than sediments in this system.

INTRODUCTION

An array of physical, chemical and biological processes affect trace elements as large rivers enter their estuaries. Inputs from local sources may be superimposed upon metal loads entering with the river (Cutter, 1989; Maest et al., 1990). Seasonal and annual variations in river flow affect transport processes that distribute metals within the system (Nichols et al., 1986; Ackroyd et al., 1987). Adsorption/desorption, particle formation, metal speciation and metal bioavailability may be affected by salinity differences on seasonal and annual time scales (Sholkovitz, 1976; Sunda et al., 1978). The physical and chemical characteristics of particles may change, affecting adsorption processes (Luoma and Bryan, 1981; Forstner and Wittman, 1983); and the naturally variable responses of local species to metals (Cain and Luoma, 1986, 1990) may be affected by life in a zone of salinity transition. The most influential of these processes will affect the space/time distribution of metals in water, sediments and biota. Thus,

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defining temporal and spatial distributions at the estuarine mouths of large rivers may improve understanding of metal fate and aid determination of any adverse biological effects of the metals.

Sediments and many benthic biota concentrate metals and integrate temporally variable metal inputs, and thus are useful in characterizations of metal distributions (Bryan et al., 1980, 1985; Phillips, 1980; Forstner and Wittman, 1983; Salomons and Forstner, 1984). Yet intensive concurrent studies of metal distributions in sediments and biota are not common in freshwater/ seawater transition zones.

This paper characterizes the regional distribution and temporal variablity of Ag, Cd, Cr, Cu, Pb and Zn in fine-grained sediments and a benthic bivalve (*Corbicula* sp) within the broad salinity transition zone where the Sacramento and San Joaquin Rivers enter San Francisco Bay (the Suisun Bay/delta region). The goal of this case study is to demonstrate the usefulness of intensive sampling in assessing the fate and effects of trace metals in estuaries. The study successfully tests whether chronic metal contamination differs between an urbanized/industrialized estuarine area and an upstream riverine site subjected only to agricultural runoff. The influence of salinity changes on metal concentrations and bioavailability are also assessed within the estuary. Specific temporal and chronic influences of local metal inputs are demonstrated and effects of riverine metal inputs on monthly and interannual variations in metal concentrations are suggested. Aggregated data from 3 years of repeated sampling at each station are employed to characterize chronic metal distributions, the relationship of sediment metals to concentrations in *Corbicula*, and relationships between metal concentrations and the condition of *Corbicula*.

STUDY AREA

San Francisco Bay is a large, urbanized estuary (Conomos, 1979) that has been subjected to a variety of pressures from human activities throughout the last 120 years (Luoma and Cloern, 1982; Nichols et al., 1986). Trace metals are one of several possible sources of biological stress, but broad generalizations about the effects of metals (Martin et al., 1984; Smith et al., 1986; Luoma and Phillips, 1988) are difficult to substantiate. The difficulty is partly the result of poor understanding of the patchy mosaic of metal contamination that characterizes the system (Luoma and Cloern, 1982; Luoma et al., 1985; Smith et al., 1986). The space/time distribution of metals in the Suisun Bay/delta, at the mouth of the Sacramento and San Joaquin Rivers (Fig. 1), is especially poorly known.

Concentrated metal inputs from local sources, quantitatively large metal inputs from river systems, and complex estuarine processes combine in Suisun Bay/delta. This area has been characterized as one of the U.S. estuaries most susceptible to pollution effects, based upon its physical characteristics and pollutant inputs (Biggs et al., 1989). The Sacramento River carries into the

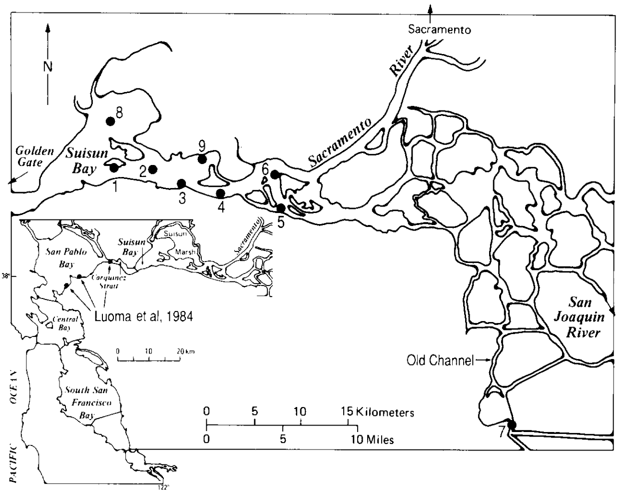

Fig. 1. Map of the Suisun Bay/delta region showing sampling sites in the present study. Inset: map of San Francisco Bay showing sites in North Bay sampled by Luoma et al. (1984).

estuary an annual mass of metals from historic mining operations upstream (Luoma and Phillips, 1988) that is equal to or in excess of total loadings from local point sources and urban runoff (Eaton, 1979; Russell et al., 1982; Gunther et al., 1987). Superimposed upon riverine discharges, nine of the largest point source dischargers in the Bay area release wastes directly into Suisun Bay/ delta (Gunther et al., 1987).

Stations 1–6, plus 8 and 9 (Fig. 1) are located along the estuarine gradient affected by the urban/industrial waste discharges and the inputs from the Sacramento River. Metal concentrations at these stations were compared with those at Station 7 in the "old channel" of the San Joaquin River, upstream from the urban/industrial area. The San Joaquin carries principally agricultural runoff. However, during most months in all years and all months in some years the flow of the San Joaquin River is less than the quantity of water diverted for human use from near its mouth (U.S. Bureau of Reclamation statistics, unpublished). The diversion pumps draw water from the Sacramento River across the delta to Station 7. Thus the Sacramento contributes to the water at this site, but the exact mix is poorly defined and variable with time.

The climate of central California is dry between May and October; thus river flow into San Francisco Bay follows a predictable seasonal cycle (Conomos, 1979). The magnitude of inflow varies from year to year, affecting estuarine circulation and the location of the salinity transition zone (Peterson et al., 1975; Conomos, 1979; Walters et al., 1985). Surface and bottom drifters move directly downstream when released into the channel of Suisun Bay at high river flows (Conomos and Peterson, 1977). In contrast, hydrodynamic models (Walters et al., 1985) suggest that metals could be redistributed upstream and across the shoals of the bay during low river flows. Patterns of Se distribution in the water column are consistent with such differences between flow regimes (Cutter, 1989).

Salinities also directly affect the fate (Sholkovitz, 1976) and bioavailability (Sunda et al., 1978; Nugegoda and Rainbow, 1989) of metals, and are affected by river flows. River flow differed among the 3 years of this study. The estuarine transition zone was located downstream from Suisun Bay throughout 1983 and salinities at all sites were < 1‰. Salinities exceeding 1‰ moved as far upstream as Station 4 in 1984 and up to Station 5 and 6 in 1985 with progressively lower river flows in those years.

METHODS

The nine stations were sampled monthly from April 1983 through September 1983, then at near-monthly intervals from February 1984 through February 1986. Sediments and clams were sampled from Stations 1–7, and sediments only from Stations 8 and 9 (Fig. 1). All samples were collected from below mean low water.

Statistics

Differences among stations and in time were determined by ANOVA comparisons of log-transformed data. To compensate for possible non-lognormal distributions the non-parametric Kruskal–Wallace test was also employed for comparisons among groups of animals. Significance ($p < 0.05$) is stated only where shown in both approaches.

Chromium discharge from the steel plant near Station 5 was monitored by plant personnel and data are from State Regional Water Quality Control Board records, compiled by Aquatic Habitat Institute, Richmond, CA (unpublished). The effluent analyses showed that a high proportion of the Cr released was in hexavalent form.

Sediments

Sediment samples were collected employing either a large diameter polycarbonate core or a polycarbonate-lined grab. Samples were raised slowly to avoid disturbing surficial material, and the sample was carefully collected from the layer of surface sediment that appeared oxidized (usually < 1 cm deep). This sediment was sieved through 100 μm mesh into water of ambient salinity. The sedimentary material that passed through the mesh was subsampled for metal analysis, and total organic carbon determination. The proportion of the sieved sediment that was composed of particles < 14 μm in diameter was estimated from the weight of unsettled particles in a sample held in a column of distilled water for 15 min (assuming the particles settled according to Stokes Law).

Coarse sand sediments, with insufficient fine material for analysis, were found on eight sample dates at Station 1, 18 sample dates at Station 2, and twice at Station 5. The particle size bias in unsieved sandy sediments resulted in low values for all sediment constituents (Luoma et al., 1989). Thus data from the sandy sediments were not considered comparable to data from sieved silt/clay sediments and were omitted from interpretive comparisons. When silt/clay sediments were present at Stations, 1, 2 and 5, constituent concentrations were typical of the rest of Suisun Bay (Table 1).

Sediment subsamples were refluxed for near-total metal analysis in concentrated nitric/sulfuric acids (2:1, respectively) (Luoma and Bryan, 1981). Although this method should remove all metal of biological significance, the near-total reflux is not expected to liberate metals from silicate lattices. However, recoveries for routinely conducted analyses of NBS reference material 1645 (River Sediment) were consistent and ranged between 79 and 97% (within the acceptable range reported by NBS for Cu, Zn, and Mn). Recoveries of Pb from standard sediments were low (56%), and thus Pb values are not comparable to other estuaries.

After digestion, sediment samples were evaporated to dryness, reconstituted in 25% hydrochloric acid, and analyzed for Cu, Pb, Zn, Fe and Mn by flame atomic absorption spectrophotometry (AAS). Concentrations of Cd and Ag in sediments were below detection by flame AAS. Concentrations of Ag were determined by flameless AAS. Cadmium was determined from separate 2 g

TABLE 1

Average physicochemical characteristics of sediments among 21 sampling dates, range, and 95% confidence interval observed at nine stations in Suisun Bay and the San Joaquin Delta

	Particle size (% < 14 μm)			TOC (%)			Fe_{HCl}			Mn_{Hxl}		
Station	$\bar{x}$	CI	Range	$\bar{x}$	CI	Range	$\bar{x}$	CI	Range	$\bar{x}$	CI	Range
1[a]	53	± 5	37–67	1.08	± 0.07	0.95–1.47	6309	± 598	4347–7707	401	± 52	267–518
2[b]	61			1.13			7583			496		
3	49	± 7	21–78	1.21	± 0.11	0.79–1.73	7894	± 815	5399–13459	363	± 48	153–614
4	54	± 6	31–82	1.21	± 0.12	0.48–1.57	6386	± 827	1630–9100	345	± 44	215–606
5[c]	54	± 5	37–58	1.10	± 0.16	0.71–1.53	5521	± 845	3129–7700	324	± 68	210–570
6	45	± 4	27–75	1.00	± 0.12	0.59–1.60	5579	± 798	2390–10547	325	± 40	171–553
7	38	± 4	19–58	0.95	± 0.11	0.48–1.39	4784	± 646	1834–7800	524	± 73	132–797
8	65	± 4	51–81	1.29	± 0.05	1.10–1.49	6674	± 714	3604–9093	505	± 66	270–829
9	45	± 3	28–56	0.96	± 0.08	0.52–1.34	5725	± 513	3909–9211	428	± 41	241–641

[a] Silt/clay sediments collected on 11 dates.
[b] Silt/clay sediments only collected once.
[c] Silt/clay sediments collected on 19 dates.

samples of dried sediments extracted with 10 ml of 0.6 *M* hydrochloric acid (Luoma and Bryan, 1981). This procedure removes 95% of the Cd extracted by the near-total digest, but has fewer analytical interferences (Luoma and Bryan, 1981). Concentrations of Cr in sediments were unusually variable (e.g. high concentrations in some sandy sediments) and recoveries were low. Thus Cr in sediments is not reported here. Deuterium arc background correction was employed in Cd, Pb and Zn analyses. The amount of amorphous iron oxide and non-residual metal was determined by extracting wet sediments (within 36 h of sediment collection) with 0.6 *N* hydrochloric acid for 2 h at room temperature (Luoma and Bryan, 1981). Concentrations of extractable Mn were determined by extracting a sediment subsample with 0.1 *N* hydroxylamine hydrochloride in 0.01 *N* nitric acid at pH 2 for 0.5 h (Chao, 1972). Total organic carbon was determined using a total carbon analyzer after inorganic carbon was removed from the samples by hydrochloric acid digestion. All sediment data are available in Luoma et al. (1990).

Metal concentrations in duplicate samples, analyzed from all sediment collections, seldom differed by more than 20% and usually less than 10%.

Clams

Corbicula is a freshwater filter feeder that can survive the salinities of the freshwater/seawater transition zone of estuaries (Evans et al., 1977). Foe and Knight (1986) describe the species of *Corbicula* found in Suisun Bay as *Corbicula fluminea*. However, a species name is not used here because uncertainties about taxonomy of the genus *Corbicula* have been raised by recent genetic studies (Belanger et al., 1986).

Corbicula sp. were raked from the surface of sediments, and depurated for 72 h in ultrapure water. Shell length was measured for each individual, then soft tissues were removed from shells, dried, weighed and digested for whole body metal analysis. Each collection from each station consisted of 15–25 animals that were sorted for analysis into 5–8 composites. Each composite was made up of individuals of similar shell length.

Composites of clam soft tissue were refluxed in concentrated nitric acid, evaporated to dryness, reconstituted in 25% hydrochloric acid and analyzed for Ag, Cd, Cu, Cr, Pb and Zn by flame AAS. National Bureau of Standards Oyster Tissue was periodically analyzed to verify full recovery by the digestion and analytical consistency with other methods. All *Corbicula* data are available in Luoma et al. (1990).

Several factors can bias interpretation of metal concentrations in *Corbicula*.

(i) Correlations between metal concentrations and animal size. During periods of contamination and at chronically contaminated stations, concentrations of Cu, Cd and Cr in *Corbicula* increased with animal size (weight and length) (data in Luoma et al., 1990). Where length and metal concentration were significantly correlated ($p < 0.10$), a representative metal concentration for the population was interpolated for the average length animal (3.0 cm shell length) in Suisun Bay. Confidence limits were then determined from the standard deviation of the fit for the value estimated from the regression.

(ii) An inadequate range of animal sizes. Occasionally, larger clams (> 3.0 cm shell length) could not be found at some stations (this happened in 20% of collections in the worst cases at Stations 5 and 6). This is a common problem when collecting bivalves from a contaminated environment (see also Strong and Luoma, 1981). Because significant correlations of metal concentration with shell length were also most common in the most contaminated environments, metal concentrations may have been underestimated in individual collections that included only small animals. Such underestimates increased the frequency and amplitude of temporal fluctuations (i.e. added noise) and could affect spatial comparisons at any one point in time. However, when data were aggregated through time such episodes had much less influence upon interpretations.

(iii) Effects of reproductive cycle. The tissue mass (and condition) of a bivalve fluctuates seasonally with the increase and loss of glycogen (energy reserves) and reproductive materials. Such increases or decreases in tissue mass are not often accompanied by changes in metal mass in bivalves, and can dilute or enhance tissue metal concentrations (respectively) (Cain and Luoma, 1986, 1990; Cain et al., 1987). Metal content of a standard shell length *Corbicula*, rather than metal concentrations, were employed in time series in order to eliminate such biases. For the calculation of content, metal concentration was normalized by the weight of a 3.5 cm *Corbicula* interpolated from the regression of shell length and tissue dry weight for that collection (as described by Cain and Luoma, 1990). The error in interpolating weight is very small, thus the confidence limits for the content values were estimated from confidence limits for population-representative concentrations.

Condition index, an indicator of well-being in *Corbicula* (Foe and Knight, 1986), was determined by regressing the mean length per individual against mean dry weight per individual for all composites from a collection, then interpolating the weight of a 3.5 cm clam at that station, on that date (Joy, 1985; Cain and Luoma, 1986, 1990). To estimate condition factors typical of a station, data were aggregated from all collections within each year, thus reducing biases caused by seasonal cycles in tissue weight.

RESULTS

Spatial distributions

Sediment

Concentrations of several metals were chronically enriched in sediments of Suisun Bay/delta compared with the lower San Joaquin River. Although Ag concentrations at all stations were always low, 3-year mean concentrations of Cu, Pb and Zn were significantly higher at Stations 1–6 than at Station 7 (Fig. 2). Concentrations of Cu in the bay/delta were two-fold higher than in the San

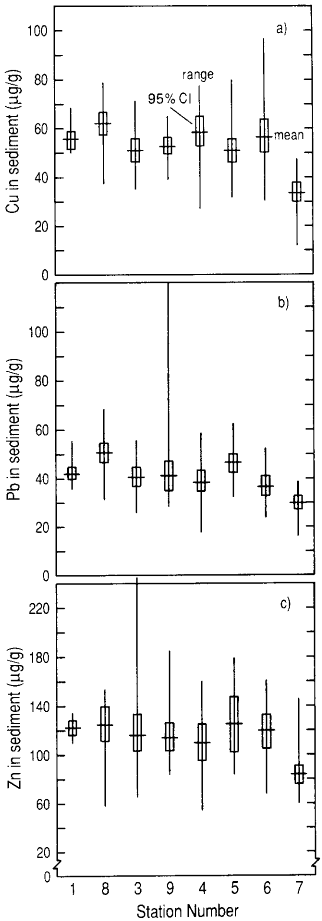

Fig. 2. Three-year means, with 95% confidence intervals and ranges of Cu (a), Pb (b), and Zn (c) concentrations in fine-grained sediments at the different stations in Suisun Bay/delta and the lower San Joaquin River.

Joaquin River; the differences for Pb and Zn were smaller. Luoma et al. (1984) reported mean Cu concentrations of 33 ± 6, 51 ± 9 and 46 ± 7 $\mu g\,g^{-1}$ in fine-grained sediments at three intertidal stations (Fig. 1) located between Suisun Bay and the mouth of San Francisco Bay (21–24 samples at each station collected in 1978–1980). These values were not significantly ($p < 0.05$) different from 3-year means from Suisun Bay/delta stations, suggesting that the general level of enrichment with Cu extends into North San Francisco Bay.

Concentrations of Cd in sediments never exceeded 1 $\mu g\,g^{-1}$ dry weight, but were frequently higher in Suisun Bay/delta than at Station 7. Concentrations were $< 0.2\,\mu g\,g^{-1}$ on all 21 sampling dates in the San Joaquin River (Fig. 3), but were $> 0.2\,\mu g\,g^{-1}$ on slightly more than half the sampling dates at Stations, 3, 4, 5 and 9. The highest Cd concentrations occurred at Station 6, and both Stations 6 and 8 had higher frequencies of elevated Cd concentrations than did other stations in Suisun Bay.

The average composition of the sediments had only a small influence on chronic sediment-metal concentrations. Differences between stations in mean sediment characteristics were small (less than two-fold) and the range of constituent concentrations overlapped among sites (Table 1). Regression analysis employing all data from all stations indicated that < 20% of the variance in Cd, Cu, Zn and Pb concentrations was explained by coincident changes in any sediment characteristic (Table 2). Correlations between 3-year mean metal concentrations in sediments and mean constituent concentrations were statistically insignificant ($p > 0.05$). The strong correlation of Cu with Cd and Zn (Table 2) may reflect some coincident inputs (see later discussion).

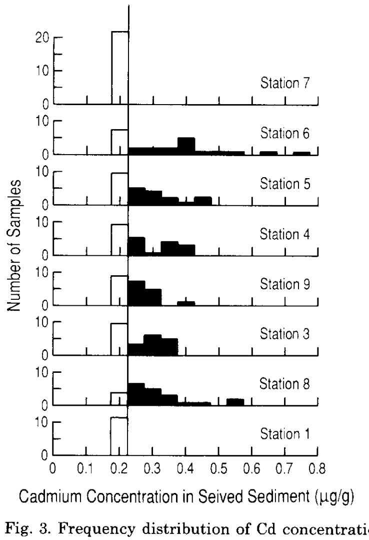

Fig. 3. Frequency distribution of Cd concentrations in fine-grained sediments observed at eight stations in Suisun Bay/delta.

TABLE 2

Matrix of correlation coefficients (r) for metals and sediment characteristics when all data from sieved sediments at all stations are combined

	Particle size	TOC	Fe_{HCl}	Mn_{Hxl}	Cu_T	Zn_T	Cd_{HCl}[a]
Particle size							
TOC	0.41*						
Fe_{HCl}	0.50*	0.48*					
Mn_{Hxl}	0.18	0.42*	0.11				
Cu_T	0.42*	0.43*	0.45*	0.13			
Zn_T	0.27*	0.31*	0.26*	0.13	0.63*		
Cd_{HCl}	0.20	0.29	0.20	−0.063	0.59*	0.42*	
Pb_T	0.42*	0.29*	0.35*	0.15	0.44*	0.38*	0.20

[a] Considers only data where Cd concentrations exceed $0.2\,\mu g\ g^{-1}$.
*$p < 0.01$.

Corbicula

Concentrations of Zn, Pb and Ag in *Corbicula* (Table 3) were low and showed little difference among stations, although Station 7 always had the lowest mean concentrations. The very small confidence intervals at each station may reflect some physiological regulation of Zn by *Corbicula* (a common observation in molluscs — Bryan, 1984). The small differences among the low Ag concentrations must be interpreted conservatively because values were near detection limits.

Distribution of Cu and Cr in *Corbicula* showed significant chronic enrichment in Suisun Bay/delta, but no consistent downstream trend in the estuary. Three-year mean concentrations of Cu and Cr were significantly greater ($p < 0.01$) at all stations in the Suisun Bay/delta than in the lower San Joaquin River (Fig. 4). Within the bay/delta, Cu concentrations were significantly higher at Stations 2, 5 and 6 than at Stations 1 and 4. The gradient in Cr distribution within the estuary was related to a local source of input.

TABLE 3

Concentrations of Zn, Pb and Ag in *Corbicula* sp. from seven sites in the Suisun Bay/delta region of San Francisco Bay

Station	Zn	Pb	Ag
7	121 ± 15	1.6 ± 0.2	0.07 ± 0.01
6	157 ± 14	2.3 ± 0.3	0.11 ± 0.02
5	177 ± 15	3.1 ± 0.5	0.10 ± 0.02
4	165 ± 21	2.1 ± 0.3	0.09 ± 0.02
3	160 ± 14	2.0 ± 0.3	0.11 ± 0.02
2	141 ± 16	2.6 ± 0.5	0.20 ± 0.05
1	140 ± 15	2.3 ± 0.4	0.13 ± 0.04

Station 5, where Cr concentrations showed significant chronic enrichment, was located near an industrial facility that is the largest single discharger of Cr to San Francisco Bay (Gunther et al., 1987). The spatial distributions suggest these discharges affected all Suisun Bay.

Annual mean concentrations of Cd in *Corbicula* were more variable than other metals, but chronic contamination throughout Suisun Bay/delta was also evident for this metal. Mean concentrations in every year at Stations 1–6 were significantly greater ($p < 0.05$) than at Station 7 (Fig. 5). Annual mean concentrations of Cd were significantly ($p < 0.05$) elevated, relative to the rest of the estuary, in 1984 at Station 2 and in 1983 at Stations 1 and 6.

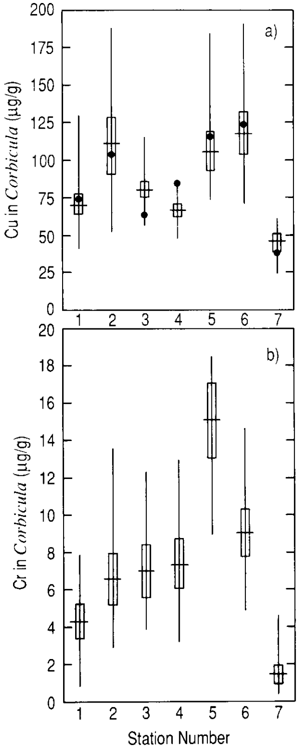

Fig. 4. Three-year means, with 95% confidence intervals and range of Cu (a) and Cr (b) concentrations in *Corbicula* sp. at Stations 1–7. (●) Mean values from interpolations for a 3.0 cm animal.

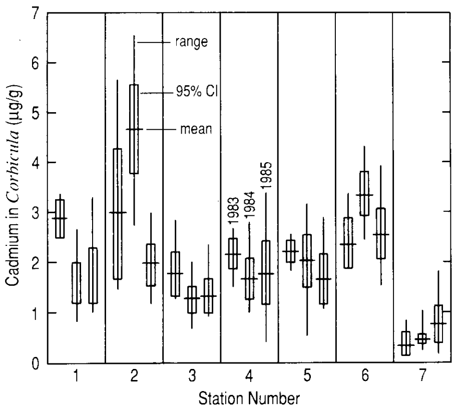

Fig. 5. Yearly means, with 95% confidence intervals and range of Cd concentrations in *Corbicula* sp. in 1983, 1984 and 1985 at Stations 1–7 in Suisun Bay/delta.

Temporal variability

Concentrations of the most enriched metals (Cu, Cd and Cr) in *Corbicula* and in sediments fluctuated two-to-four fold through the 3-year period of study (e.g. Figs 6–12). The patterns of fluctuations showed enough differences among stations to verify the importance of establishing mean conditions to define chronic contamination or a temporal context when assessing spatial distribution of contamination at any one point in time. Analyses of the time series suggested influences of several processes on metal concentrations in sediments and the clams.

Effect of sediment characteristics

Fluctuations of Cu and Zn concentrations in sediments at Stations 3, 4 and 7 (Fig. 6) were typical of fluctuations observed for all metals [see Luoma et al. (1990) for a more complete set of time series]. Patterns of fluctuation showed many differences among stations, but were often similar for different metals at the same station.

Some of the seasonal fluctuations in sediment metal concentrations coincided with fluctuations in sediment characteristics (Figs 6 and 7). For example, at Station 3, sediments were consistently most coarse in mid-summer (whatever the salinity), with low concentrations of TOC, Fe, and Mn; and most fine in Oct.–April with higher concentrations of TOC, Fe, and Mn (Fig. 7). Concentrations of Cu at Station 3 correlated more significantly with TOC than

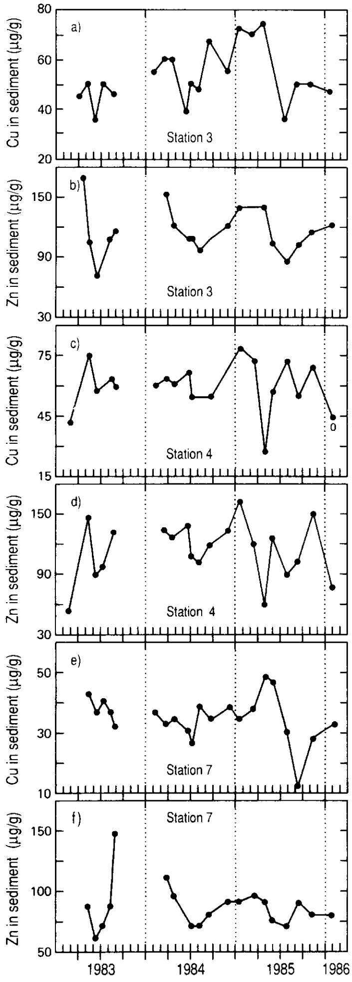

Fig. 6. Temporal variation between 1983 and 1986 in concentrations of total Cu and Zn in fine-grained sediments from Stations 3, 4, and 7 in the study area.

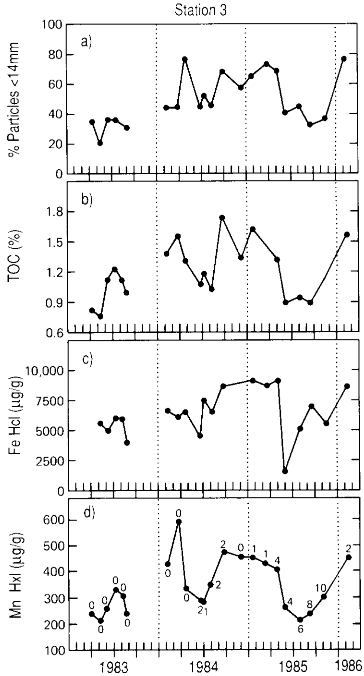

Fig. 7. Percent fine particles (a), total organic carbon (TOC) (b), extractable Fe (c), and extractable Mn (d) determined in fine-grained sediments at Station 3 in Suisun Bay between 1983 and 1986. Salinities at Station 3 are shown as numbers near data points in (d).

with other variables; fluctuations in TOC appeared to explain 35% of the Cu variance at this station ($r^2 = 0.35; p < 0.01$). At Station 8, 46% of the variation in sedimentary Cu occurred coincident with variation in concentrations of TOC ($p < 0.01$). In contrast, the temporal variability of Cu concentrations did not correlate significantly ($p > 0.05$) with variations in constituent concentrations at Stations 4, 6, 7 or 9, although coincidence was suggested in some specific events (Luoma et al., 1990). This example reinforces the suggestion from the aggregated data, that the composition of the sediments exerted some influence on metal concentrations, but was not the only important factor.

Movement of the freshwater/seawater transition zone in the estuary, reflected by changing salinity at the different stations, had no discernable effect on metal concentrations in bed sediments or on sediment composition. Metal concentrations and concentrations of sediment constituents showed no significant correlation with salinity, nor did changes in salinity coincide with any obvious consistent response in metal or sediment constituent concentrations through the 3-year time series (Figs 6 and 7). Longer time series seem necessary to illustrate any influence of salinity transitions, or their accompanying processes, on metal concentrations in the bed sediments of Suisun Bay (e.g., see Thomson-Becker and Luoma, 1985).

Seasonal changes in condition of Corbicula

Significant seasonal differences were observed in concentrations of Cu, Cr, and Cd in *Corbicula* at all stations. The weight of a 3.5 cm *Corbicula* also fluctuated seasonally at all stations (e.g. see Fig. 8 for Station 1). At least a portion of the fluctuation in metal concentrations occurred inversely with fluctuations with weight, as observed in mussels (Phillips, 1980) and in deposit-feeding clams (Cain and Luoma, 1986, 1990). When metal content was calculated to compensate for the effects of weight change (Phillips, 1980; Cain and Luoma, 1990), the significant peaks in concentration that were observed between June and September at most stations in 1983 and 1984 disappeared.

Effect of local anthropogenic inputs

As shown for other bivalve species (Cain and Luoma, 1990), metal content of *Corbicula* appeared to reflect the metal exposure history of the organism. Chronic spatial distributions indicated an internal source of biologically available Cr occurred within Suisun Bay/delta. Temporal fluctuations in Cr content of *Corbicula* at Station 5 also appeared to reflect exposures when compared in 1984 and 1985 with Cr concentrations in the effluent of the same source (a nearby steel plant, Fig. 9). The Cr content of *Corbicula* was significantly increased in December, 1984 ($p < 0.05$), compared with preceding

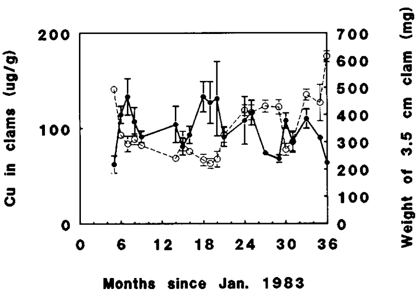

Fig. 8. Concentration of Cu in *Corbicula* and weight of a 3.5 cm clam (interpolated from tissue mass vs length in each collection) between 1983 and 1986 at Station 1 in Suisun Bay/delta. Vertical bars represent 95% confidence interval or standard deviation of the fit for each interpolated value.

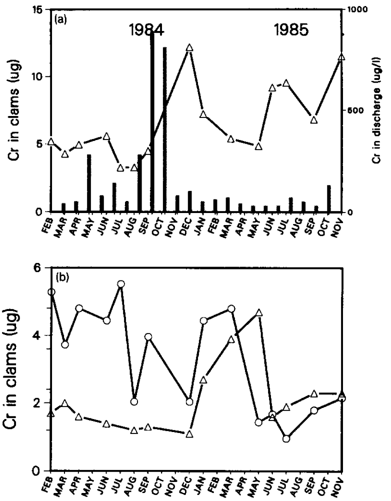

Fig. 9. Content of Cr in a 3.5 cm *Corbicula* (a) at Station 5, and (b) at Station 3 (○) and Station 1 (△) in Suisun Bay/delta in 1983–1986. Vertical bars represent Cr concentrations in effluent of an industrial plant near Station 5.

months, after an episode of Cr discharge from the plant in late 1984 (Fig. 9). An episode of elevated Cr content in *Corbicula* was also suggested at every station in Suisun Bay/delta in the months following the 1984 release (Fig. 9). A significant increase in Cr concentration was observed at Station 5 in summer 1985, coincident with smaller releases from the plant. It is possible that localized contamination of the clams during this period was enhanced by the extended period of low river inflow, but verification of such an hypothesis would require more intensive study.

Salinity and metal bioavailability

Low salinities could enhance metal bioavailability during high river flows by affecting solute metal speciation and osmoregulation processes in the animals. Increases in Cd and Zn availability at low salinity are well documented in controlled studies (Sunda et al., 1978; Nugegoda and Rainbow, 1989). However, correlations between salinity and the content of the most important metal contaminants in *Corbicula* (Cu, Cd or Cr) were statistically insignificant through time within stations and when comparisons were made among stations. Nor did multiple regression of sedimentary metal concentrations and salinity vs metal content of *Corbicula* indicate any significant in-

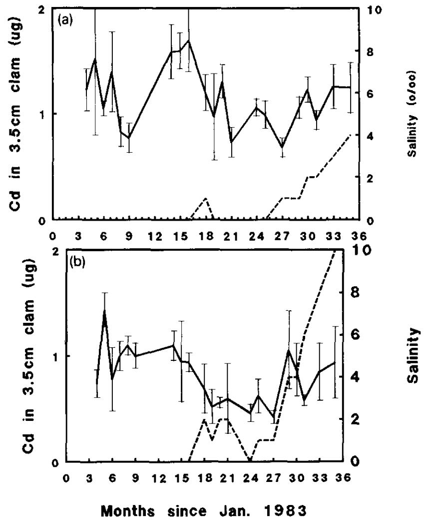

Fig. 10. Content of Cd (——) in a 3.5 cm *Corbicula* at Station 6 (a) and Station 4 (b) in Suisun Bay delta. (- - - -) Salinities determined at the time clams were collected. If no data present, salinity was < 1‰. Vertical bars represent 95% confidence intervals determined from confidence intervals for concentration values.

teractive effect on metal bioavailability. The lack of response of Cd content of *Corbicula* to changing salinities is illustrated in Fig. 10. Significantly higher Cd content occurred at Station 6 in Jan.–April 1983 and 1984 compared with summer months in those years, in the absence of changes in salinity. More important, no significant declines in Cd content occurred at any station in 1985 during the period of low flow and increasing salinity (Fig. 10). Within the range of salinities observed during this study (0–14‰), effects on speciation of Cu, Cr and Cd were apparently less significant than other processes in affecting the bioavailability of these metals to *Corbicula.*

River flow

Metal inputs to Suisun Bay/delta occur via the Sacramento River. One source of metals to the river is the area of historic metal mining in the upper watershed (Nordstrom et al., 1977). Downstream transport of such contamination over long distances has been shown in other river systems (Axtmann and Luoma, 1987). The largest metal discharges to the Sacramento River occur via Spring Creek Reservoir near Lake Shasta, and are monitored by the California

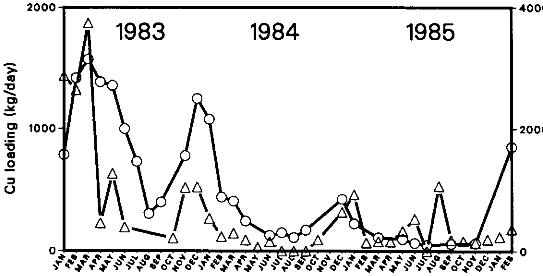

Fig. 11. Mean monthly river flow to San Francisco Bay from the Sacramento/San Joaquin Rivers (○) and Cu discharge from Spring Creek Reservoir (△) in the watershed of the Sacramento River in 1983–1986. River inflow is determined from the "total outflow index" at Chipps Island as recorded by the U.S. Bureau of Reclamation (unpublished).

Water Quality Control Board. Those data show that discharges of Cu to the river were greatest during the highest river flows in 1983–86 (Fig. 11). An anomalous small discharge episode also occurred during low flow in August 1985.

Significant increases in Cu and Cd content of *Corbicula* were especially evident at Station 6, at the mouth of the Sacramento River, during high metal discharges in 1983 and 1984 (Fig. 12) (salinities at this station were <1‰

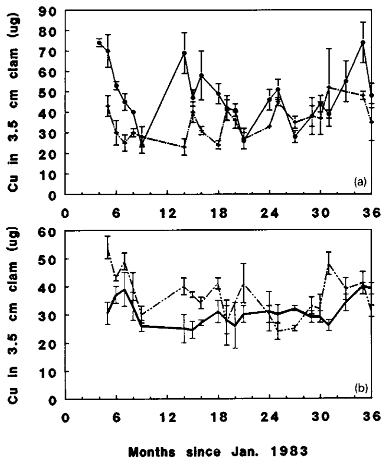

Fig. 12. (a) Content of Cu in 3.5 cm *Corbicula* from Station 6 (———) and Station 4 (–·–·–) in 1983–1986. (b) Content of Cu for Station 2 (—··—) and Station 1 (———). Vertical bars are 95% confidence intervals determined from confidence intervals for concentration values.

throughout these periods). Episodic coincidence with Cu discharges from the river were also suggested at other stations, but will require more intensive investigation for verification. The spatial distribution of Cu in *Corbicula* at high flow in 1983 and 1984 was also indicative of effects from riverine inputs. Copper content was highest at Station 6, at the mouth of the Sacramento River, lower among other stations in Suisun Bay, and lowest at Station 7, where Sacramento and San Joaquin water mix. In contrast, spatial distributions during the highest inflow years suggested Cd inputs were not exclusively from the river (Fig. 5). Cadmium content of *Corbicula* followed the order: Station 1, 6 > 2, 3 > 4, 5 > 7.

The lowest content of Cd and Cu in *Corbicula* in 1983 and 1984 occurred in summer (Figs 10 and 12). In contrast, the highest metal content in 1985 at all stations except Station 7 occurred between July and November. The August episode of metal discharge upstream in the Sacramento River may have contributed to the summer 1985 metal increases. However, the magnitude of the increase was similar at all stations and the increase lasted throughout the low flow period at most stations. This raises the possibility that contamination with bioavailable Cu and Cd was held longer and spread more widely in Suisun Bay/delta because of the extended period of low river flow that occurred during the unusually dry conditions in 1985.

Metal concentrations in sediments vs concentrations in animals

Although *Corbicula* is a filter feeder, rather than a deposit feeder, the spatial distribution of contamination in sediments was generally similar to the distribution of metals in the bivalve. Chronic levels (whether determined by 3-year or yearly mean concentrations) of Cu, Cd, Pb and Zn in *Corbicula* correlated significantly with chronic contamination in sediments. The strength of the correlation differed among metals, however. For some metals (e.g., Cu, Fig. 13) the differences between Station 7 and Stations 1–6 dominated the relationship. For others (e.g., Cd), the correlation was more consistent among stations.

Amongst all data the correlations between sediments and animals were not statistically significant ($p < 0.05$). In some cases, episodes of increased or decreased enrichment did not correspond in sediments and animals. In other cases, delays appeared to occur in animal responses to changes in environmental concentrations of metals. Such differences are common in comparisons of metal contamination in sediment and biota (Luoma, 1983, 1989; Cain and Luoma, 1990) and demonstrate the importance of aggregating data from repeated samplings at each site to demonstrate broad or general relationships.

Relationship of metal content to condition of Corbicula

The contamination of *Corbicula* with Cd, Cr and Cu in Suisun Bay/delta may affect the condition of the clams. Annual mean condition index differed significantly among populations of *Corbicula* in the study area and between years

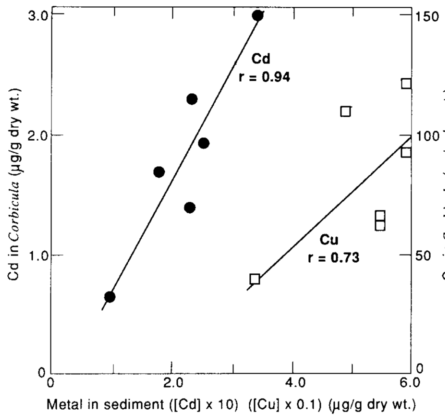

Fig. 13. Correlation of 3-year mean concentrations of Cd and Cu in sediments with mean concentrations in *Corbicula* sp. among Stations 2–7. Station 1 is not included in regression because fine-grained sediments were found there only on one sampling date.

within some populations (Table 4). Mean and maximum shell length were significantly greater at Station 7 than elsewhere, but were not significantly different among Stations 1–6 (Johns and Luoma, 1988). Food availability (i.e. phytoplankton productivity) may be greatest at Station 7, but is not likely to be different between Stations 1–6 (Cloern, 1987). Salinity might stress *Corbicula*, but no significant correlation between mean condition and salinity exposure was observed among the populations, nor were the differences in condition index consistent along the estuarine gradient (e.g. Table 4). However, the mean condition index correlated significantly with mean tissue concentration and, more importantly, content of Cd, Cu or Cr (r^2 = 0.67, 0.81 and 0.40, respectively; Fig. 14). Mean condition index also correlated strongly

TABLE 4

Annual mean condition index of *Corbicula* (mg/3.5 cm clam) and annual mean salinity (‰) at four stations in Suisun Bay/delta

	7	6	3	2
1983	805 ± 171 (0)	427 ± 74 (0)	491 ± 68 (0)	342 ± 86 (0)
1984	785 ± 158 (0)	291 ± 36 (0.01)	564 ± 113 (0.09)	255 ± 45 (2.2)
1985	645 ± 140 (0)	271 ± 45 (1.6)	539 ± 95 (4.9)	350 ± 81 (6.1)

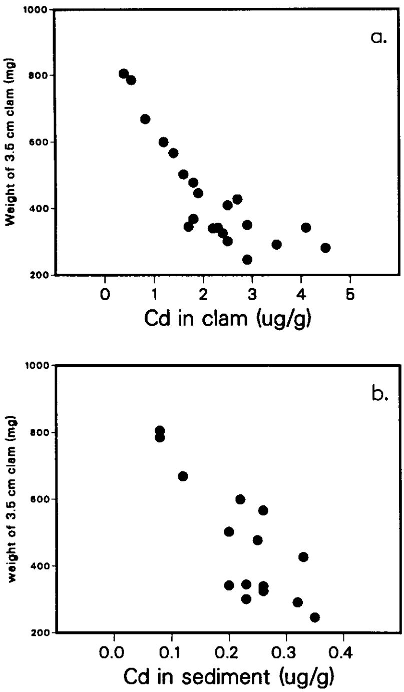

Fig. 14. Correlation of Cd concentrations (a) in *Corbicula* (r^2 = 0.67; $p < 0.001$) and (b) in sediments (r^2 = 0.67; $p < 0.001$) with condition index. Condition index is defined as dry weight of a 3.5 cm *Corbicula*.

with concentrations of Cd in sediments ($r^2 = 0.67, p < 0.001$), and significantly with sediment concentrations of Cu ($r^2 = 0.29$, $p < 0.05$). The differences between Cu and Cd in the strength of the latter correlation could reflect a greater influence of Cd on condition, or the poorer correlation of sediment-bound Cu with bioavailable Cu.

Several episodes in the time series also suggested that the condition index responded to elevated metal concentrations. For example, during the episode of Cr release in 1985, insignificant relationships between weight and length were observed where Cr concentrations were highest, in five of eight collections at Station 5 and three of nine collections at Station 6. No insignificant relationships were observed in that year at the least contaminated Stations 3, 4 or 7. The insignificant relationships resulted from low tissue weight in one or more composites (usually larger, more contaminated individuals) in a collection.

Comparisons with other systems

Trace element enrichment at Station 7 in the lower San Joaquin River was not great, compared with other systems. In the rest of Suisun Bay/delta, the Cu and Cd contamination in sediments was moderate relative to other marine and estuarine systems (Goldberg et al., 1978; Hallberg, 1979; Katz and Kaplan, 1981; Sinex and Helz, 1981), although differences in analytical methodologies and physicochemical sediment characteristics affect comparisons (Luoma, 1990). Concentrations of Zn, Pb and Ag in both sediments and *Corbicula* at Stations 1–6 were also moderate or low (Caldwell and Buhler, 1983; Elder and Mattraw, 1984; Belanger et al., 1986; Tatem, 1986), although Foe and Knight (1986) have observed some elevated Pb concentrations near Station 5.

In contrast, Cu, Cd and Cr contamination of *Corbicula* from the industrialized area of Suisun Bay/delta was substantial relative to other systems. Copper in *Corbicula* was 6–10 times higher than in unenriched systems (Fig. 15). Contamination with Cd was even more severe. Concentrations at Station

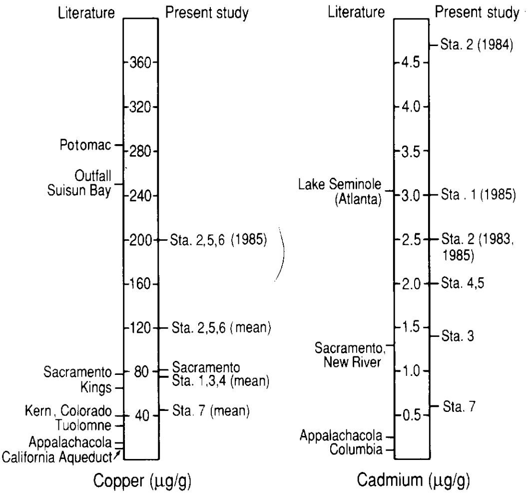

Fig. 15. A comparison of the Cd and Cu concentrations observed in *Corbicula* from Suisun Bay with concentrations observed in other studies. Sources for data from other systems are: Potomac River, Cory and Dresler, 1981; Appalachicola River and Lake Seminole, Florida, Elder and Mattraw, 1984; Sacramento, Foe and Knight, 1986, Woodard, 1979; Kings, Tuolumne, Colorado and Kern Rivers, California, Woodard, 1979; California Aquaduct, Harrison et al., 1984; New River, Virginia, Rodgers, et al., 1977; Lake Washington, Mississippi, Price and Knight, 1978; Outfall study, Suisun Bay, Foe and Knight, 1986; Columbia River, Washington, Caldwell and Buhler, 1983.

2 in 1984 and Station 1 in 1983 exceeded those for *Corbicula* in unaffected systems by 20–30-fold; and Cd found in *Corbicula* in 1984 at Station 2 (means of up to 4.5 $\mu g\ g^{-1}$ and levels of up to 10.5 $\mu g\ g^{-1}$ in larger animals) was higher than reported anywhere in nature for this species. The World Health Organization limit for Cd in food for human consumption is 10 $\mu g\ g^{-1}$ (dry weight) (Talbot et al., 1985). The highest Cr concentrations reported outside San Francisco Bay were 35–40 $\mu g\ g^{-1}$, found in the Potomac River (Cory and Dresler, 1981). Some individual samples of *Corbicula* found near Station 5 in our study and *Corbicula* collected in that area by Foe and Knight (1986) contained Cr at this high level. The severity of the Cd, Cr and Cu contamination in *Corbicula* from Suisun Bay helps explain the suggested influence of metals on the condition index of the clams.

DISCUSSION

This study illustrates the variety of processes that may influence the distribution and effects of metal contamination as large rivers enter their estuaries. Enrichment with some metals (e.g. Se, Cu, Cr, Cd), but not others (e.g. As, Hg, Ag) appears to occur coincident with urban/industrial waste discharge and Sacramento River inputs of metals to the Suisun Bay/delta (see also Johns and Luoma, 1988, 1990). Influences of specific local sources were demonstrated for Cr, and influences of episodic riverine metal inputs were indicated for Cu and Cd. Although the direct effects of salinity on metal distributions and bioavailability appeared to be secondary, natural processes were influential. Changes in sediment composition affected sediment metal concentrations. Seasonal and interannual differences in estuarine hydrodynamics may also have affected space/time metal distributions. Some data suggested that, during consecutive wet years, low inflow periods are sufficiently short that dry season increases in metal availability do not occur. Build-up of contamination may occur when low inflow periods are extended (see also Cutter, 1989), and internal or stored sources of contamination may spread through Suisun Bay/delta. Because human activities are changing river inflows to San Francisco Bay (Nichols et al., 1986), further exploration of hypotheses concerning flow-related hydrodynamic effects on contamination is important.

Copper and cadmium appeared to be highly bioavailable to *Corbicula* in Suisun Bay/delta. Although sediments were only moderately contaminated with these metals, substantial contamination occurred in *Corbicula*. Concentrations of Cu in *Corbicula* are lower than in sediments in other systems (Elder and Mattraw, 1984), but concentrations of this metal in clams in the Suisun Bay/delta were 2–3-fold higher than in sediments. Bioconcentration factors (the ratio of metal in organism to metal in solution) of (1.7–2.3) $\times 10^4$ for Cu and (0.17–0.37) $\times 10^4$ for Cd were determined for *Corbicula* by Graney et al. (1983, 1984) from model stream studies. Eaton (1979) measured Cu concentrations of 3.0–5.3 $\mu g\ g^{-1}$ and Cd concentrations of 0.12–0.24 $\mu g\ g^{-1}$ near our Stations 2 and 5 in Suisun Bay. Employing these values, Cu and Cd bioconcentration factors at Stations 2 and 5 in Suisun Bay exceeded the highest values reported by Graney et al. (1984) [the range for Cu is (2.25–6.25) × 10^4 and for Cd it is (1.04–3.92) × 10^4]. Although such calculations represent only crude estimates, their conclusions are substantiated by other observations. Thomson et al. (1984) and Luoma et al. (1985) also reported an enhanced availability of Cu to the clam *Macoma balthica* in South San Francisco Bay; and other studies describe patches of very high metal concentrations in benthic biota in several reaches of San Francisco Bay where sediments were only moderately enriched (Luoma and Cloern, 1982; Luoma and Phillips, 1988). Together these results suggest that some process enhances Cu and Cd bioavailability in at least parts of the San Francisco Bay system, making it more vulnerable to biotic enrichment with these metals than many comparable environments. The enhanced bioavailability of these metals also means that the clams were more sensitive indicators of trace metal enrichment than were sediments in the Suisun Bay/delta system.

The enhanced bioavailability of Cu and Cd, plus the large industrial inputs of Cr to Suisun Bay/delta, may affect the condition of *Corbicula*. Other studies have demonstrated the sensitivity of tissue growth and condition index to stress in this bivalve (Belanger et al., 1986; Foe and Knight, 1986). However, relationships between condition index and both tissue concentrations and sediment concentrations of trace elements have not been shown elsewhere. Other lines of investigation should be pursued to determine if *Corbicula* is stressed and/or vulnerable to additional human-induced or natural stresses in the more contaminated localities in Suisun Bay/delta.

The temporal variability of metal concentrations in estuaries and the spatial variability characteristics of urbanized estuaries pose challenging design problems for studies of contaminant distributions or trends. In the present study, results from analyses of both sediments and a benthic animal provided converging evidence to substantitate definitions of metal distributions. Annual or 3-year means from frequent, repetitive sampling provided a confidence interval for estimates of chronic contamination (Johns and Luoma, 1988). Linkages of sediment contamination with that in animals, and affects of contamination on the condition of *Corbicula* were also most evident in data aggregated from repetitive samplings. Interpretations of time series data from 3 years sampling were more tenuous, but did point toward important hypotheses about processes affecting metal fate in this complicated system. Longer-term data collection, even at a single station, has improved time series interpretations in other studies (Luoma et al., 1985; Thomson-Becker and Luoma, 1985; Cain and Luoma, 1990). Most important, these results demonstrate that strategies for monitoring estuaries and determining contaminant dynamics or effects, must recognize the spatial and temporal complexity and heterogeneity of contamination in large rivers and their estuaries.

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