ARSENIC IN BENTHIC BIVALVES OF SAN FRANCISCO BAY AND THE SACRAMENTO/SAN JOAQUIN RIVER DELTA

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ABSTRACT

Arsenic concentrations were determined in fine-grained, oxidized, surface sediments and in two benthic bivalves, *Corbicula* sp. and *Macoma balthica*, within San Francisco Bay, the Sacramento/ San Joaquin River Delta, and selected rivers not influenced by urban or industrial activity. Arsenic concentrations in all samples were characteristic of values reported for uncontaminated estuaries. Small temporal fluctuations and low arsenic concentrations in bivalves and sediments suggest that most inputs of arsenic are likely to be minor and arsenic contamination is not widespread in the Bay.

INTRODUCTION

The aquatic chemistry, geochemistry, and bioaccumulation of anthropogenically mobilized arsenic have been the focus of much research because of the established toxicity of this element (Ferguson and Gavis, 1972; Crecelius, 1975; Crecelius et al., 1975; Penrose et al., 1975; Andreae, 1978; Fowler and Unlu, 1978; Waslenchuk and Windom, 1978; Unlu and Fowler, 1979; Wrench et al., 1979; Klumpp, 1980; Langston, 1980, 1983, 1984; Sanders and Windom, 1980; Phillips and Depledge, 1985, 1986). This paper reports the spatial distribution of arsenic in two species of benthic bivalves and in fine, oxidized surface sediments of San Francisco Bay. The major goal was to determine if arsenic contamination is common in bivalves and sediments in the estuary, with a particular focus on the northern reach, which shows regional enrichment with selenium (Johns et al., 1988). Strong seasonal variations of riverine trace element inputs and sediment characteristics contribute to fluctuations in levels of biologically available trace elements within the San Francisco Bay estuary (Luoma and Phillips, 1988). These fluctuations can cause trace element concentrations within benthic bivalves to fluctuate as well (Luoma et al., 1985; Luoma and Phillips, 1988). In order to separate possible natural fluctuations from anthropogenically caused enrichment of arsenic in bivalves and

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sediments, temporal variability in tissue and sediment concentrations of arsenic was also considered.

San Francisco Bay is a large, urbanized estuary which has been extensively altered by human activity (Nichols et al., 1986). The sediments and biota reflect trace element inputs from both local and regional sources (Luoma and Cain, 1979; Luoma and Cloern, 1982; Thomson et al., 1984; Johns et al., 1988; Luoma and Phillips, 1988; Luoma et al., 1990). However, data are limited for the concentrations of arsenic in sediments and bivalves. Previous studies in San Francisco Bay focused on effects of dredging a naval shipyard (Anderlini et al., 1975) and determining concentrations in sediments and several bivalve species in the more saline portions of the Bay, prior to recent increased urbanization and industrial activity (Risebrough et al., 1977). No studies have examined arsenic concentrations in bivalves or sediments in the northern reach of the estuary.

Arsenic may enter the estuary in the effluents of a number of municipal and industrial discharges (Luoma and Cloern, 1982; Moore and Ramamoorthy, 1984). Another potential source of arsenic to the estuary is the San Joaquin River. In the western San Joaquin Valley, natural weathering of soils derived from arsenic-rich marine shales has been accelerated by irrigation and artificial drainage of saline soils. Some of the agricultural drainage water enters the San Joaquin River and may reach the head of the estuary.

METHODS AND MATERIALS

Sampling design

Arsenic concentrations were determined in two species of benthic bivalves, Corbicula sp. and Macoma balthica, and in fine-grained, oxidized, surface sediments. Arsenic concentrations in bivalves and sediments from the agricultural region of the lower San Joaquin River and Delta (C7, C11; Fig. 1) were compared with arsenic concentrations at stations in the northern reach of San Francisco Bay, a heavily urbanized and industrialized area (C1-C7, C8). Stations C1-C7 were sampled repeatedly from September 1984 through September 1986 to examine temporal variability.

Several more stations were sampled once each, concurrently with C7. Arsenic concentrations in the sediments and bivalves at C7 were compared with areas known to receive agricultural drainage return water from saline soils (C12), and with two stations which did not receive significant runoff from saline soils, C9 on the Sacramento River and C10 on the Tuolumne River (Fig. 1). Station C11, also in the lower San Joaquin region, was sampled once to compare the representativeness of Station C7 in this hydrologically complex area.

A small sampling effort examined potential enrichment of arsenic in sediments and bivalves (*Macoma balthica*) in the more saline portions of San Francisco Bay. Samples were collected twice at one location in North Bay



Fig. 1. Locations of sampling stations and designated geographical areas. 'M' indicates stations where *Macoma balthica* was sampled; 'C' indicates stations where *Corbicula* sp. was sampled.

(M1), once at one location in Central Bay (M2), and twice at one station in South San Francisco Bay (M3). From February 1986 through May 1987, monthly samples were collected at a fifth South Bay location (M4) which has shown contamination of sediments and bivalves with several metals (Luoma and Cain, 1979; Thomson et al., 1984; Fig. 1). Arsenic concentrations in *Macoma* at these stations were compared with one collection from the estuary at Big River on the Northern California Coast (M5, Fig. 1). Big River has no known anthropogenic source of arsenic.

Sample collection, preparation, and analysis

Bivalves were sampled, depurated, divided into size classes, homogenized, lyophilized, and prepared for arsenic analysis by a dry ash method as previously described (Johns et al., 1988). The 4 M HCl solutions of samples were treated with an excess of potassium iodide to ensure reduction of all arsenate to arsenite prior to analysis by hydride generation atomic absorption spectroscopy, employing 3% NaBH₄ (in 1% NaOH) as the reductant.

Reagent blanks and standard reference materials (NBS SRM 1566 Oyster Tissue, RM 50 Albacore Tissue, and SRM 1572 Citrus Leaves) were analyzed at regular intervals. Recoveries of arsenic from the biological reference materials are reported in Table 1.

Sediment samples were collected and prepared for arsenic analysis by the same methods as previously described for selenium (Johns et al., 1988). Recoveries of arsenic from sediment reference materials are reported in Table 1. Aliquots of wet-sieved sediments were extracted with 0.5 M HCl as described by Langston (1980).

TABLE 1

Recoveries of total arsenic from standard reference materials

Certified value ^a	Recovered value
13.4 + 1.9	$13.0 \ (n = 13; \text{SD} = 0.90)^{\text{b}}$
$3.3 + 0.4^{\circ}$	3.1 (n = 24; SD = 0.20)
3.1 + 0.3	3.3 (n = 6; SD = 0.10)
11.6 + 1.3	10.4 (n = 6; SD = 0.4)
66 ^d	68.7 (n = 6; SD = 2.1)
115 + 10	122 (n = 2; SD = 2.1)
10.0	9.3 $(n = 3; SD = 0.6)$
	$\begin{array}{c} \text{Certified value}^{a} \\ 13.4 + 1.9 \\ 3.3 + 0.4^{c} \\ 3.1 + 0.3 \\ 11.6 + 1.3 \\ 66^{d} \\ 115 + 10 \\ 10.0 \end{array}$

* Concentrations in micrograms arsenic per gram, dry weight.

^b n = number of separate samples processed; SD is the standard deviation of the mean.

^c Not certified. Value given as the probable mean.

^d Only reported values (not certified) are obtainable.

RESULTS

Arsenic concentrations in Corbicula

Mean arsenic concentrations in *Corbicula* ranged from 5.4 to $11.5 \,\mu g g^{-1}$ among all stations. Temporal variability in mean arsenic concentrations was slight at most stations and showed no consistent seasonal trends (Fig. 2, Station C5).

Arsenic concentrations in animals from the San Joaquin River were significantly ($P \le 0.05$) lower than in animals from the Sacramento and Tuolumne (Fig. 3). Grand mean arsenic concentrations were not significantly different (P > 0.05) at C7 and C11, in the lower San Joaquin River (Fig. 3). Arsenic concentrations at C7 and C11 were also similar to those at C12 on the mid-San Joaquin River, which receives agricultural return water.

The strategy of repeatedly sampling Stations C1-C7 allowed sensitive resolution of spatial distributions of arsenic. The grand mean concentrations in *Corbicula* (aggregated data, all collections) were significantly ($P \le 0.05$) higher from stations in the estuary (C1-C6, C8) than in the lower San Joaquin River (C7, Fig. 3); but concentrations in clams from the estuary were not different than in clams from the Sacramento and Tuolumne Rivers. Some significant, but small, differences in mean arsenic concentrations in tissues occurred among Stations C1-C6 as well ($P \le 0.05$; Fig. 3). However, there was no discernible spatial trend in arsenic concentrations among these stations.

Animal size showed no consistent influence on arsenic concentrations in tissues of *Corbicula*. Significant ($P \le 0.05$) positive relationships between shell length and tissue concentration of arsenic occurred in 39% of the collections.



Fig. 2. Mean arsenic concentrations in *Corbicula* from Station C5 as observed from September 1984 through September 1986. Vertical bars represent 95% confidence intervals for the means. The mean arsenic concentration ($\mu g g^{-1}$ dry weight) is listed above each bar. The number of composited samples analyzed is designated at the bottom of the figure. Values for means and confidence limits have been back-transformed from log₁₀.



Fig. 3. Grand mean arsenic concentrations in *Corbicula* sp. from northern San Francisco Bay and the lower San Joaquin River (C1–C8) collected between September 1984 and September 1986, compared with mean concentrations in animals collected in November 1985 from the Sacramento River (C9), and in September 1986 from the Tuolumne (C10), the lower San Joaquin at Mossdale (C11), and the middle San Joaquin River (C12). Vertical bars represent 95% confidence intervals for the means. The number of composited samples analyzed for each mean is shown above the station designation. Bars are labelled by the same letter when means are not significantly different (P > 0.05; analysis of variance and T-K method for testing differences among means; Sokal and Rohlf, 1981). Two sets of ANOVA and range tests were performed; one compared grand means at C1–C8 and the other compared means for C7 and C9–C12. Tests are differentiated by locating one set of letters above and the other below the 95% confidence limits (respectively).

Slopes of these regressions and the portion of the variance in arsenic concentrations accounted for by animal size varied markedly among sampling dates at each station. Size influence did not appear to bias comparisons among stations. Mean shell lengths of *Corbicula* from Stations C7, C11, and C12 were similar to each other and significantly ($P \le 0.05$) greater than the other *Corbicula* stations (Johns et al., 1988). However, lowest mean arsenic concentrations were found at C7 and C11. Highest mean arsenic concentrations were found in animals from C1–C6 and C8, even though mean shell lengths were smaller at these stations.

Arsenic concentrations in Macoma

No significant, positive relationships between size of *Macoma* and arsenic concentration were observed within individual collections at M4. However, in a linear regression of all arsenic concentrations on shell lengths at M4, animal size accounted for a small portion of the variability in tissue arsenic concentration (P < 0.05; coefficient of determination = 0.124). Significant, positive linear relationships between shell length and tissue arsenic concentration occurred only twice among 23 collections at five *Macoma* stations (M5, r = 0.661, $P \leq 0.05$; M3, r = 0.863, $P \leq 0.01$).



Fig. 4. Mean arsenic concentrations in *Macoma balthica* from stations in San Francisco Bay at sampling dates between September 1985 and May 1987: M1 (\ominus); M2 (\odot); M3 (\blacktriangle); M4 (\bullet), and Big River: M5 (\bigcirc). Vertical bars represent 95% confidence intervals for the means. Mean arsenic concentration ($\mu g g^{-1}$) is listed beneath each bar. The number of composited samples analyzed is designated at the bottom of the figure.



Fig. 5. Grand mean arsenic concentrations in *Macoma balthica* from stations in San Francisco Bay (M1-M4) and Big River (M5). The number of composited samples analyzed for each mean is shown above the station designation. Vertical bars represent 95% confidence intervals for the means. Bars are labelled with the same letter when means are not significantly different (P > 0.05; analysis of variance and T-K method for testing differences among means; Sokal and Rohlf, 1981).

Mean arsenic concentrations in *Macoma balthica* at M4 ranged from 5.8 to 11.7 μ g g⁻¹ with a grand mean of 9.0 μ g g⁻¹ among the 17 collections. Mean arsenic levels in the M4 bivalves fluctuated among collections, but did not show consistent seasonality (Fig. 4). The grand mean arsenic concentration (7.6 μ g g⁻¹) was lowest at M2 (P \leq 0.05; Fig. 5). This station is located in the well-flushed central portion of the estuary and animals at this station probably represent a recent recolonization of the site (Luoma, unpublished data). Otherwise, grand mean arsenic concentrations in *Macoma* were similar among most stations in Central and South San Francisco Bay and did not differ from the mean arsenic concentration at M5, where no anthropogenic sources of arsenic input are known.

Arsenic concentrations in fine sediments

Total arsenic concentrations of all fine sediment samples ranged from 6 to $16 \ \mu g \ g^{-1}$ and typically averaged $10-12 \ \mu g \ g^{-1}$ at most stations (Fig. 6). Concentrations in the lower San Joaquin, the Sacramento and the Tuolumne were slightly lower than in the estuary, although the stations were similar in ancillary characteristics of the sediments (% carbon, total iron, total manganese, fine particles abundance; Luoma et al., 1990). Grand mean HCl-extractable arsenic concentrations ranged between 2.0 and 4.0 $\mu g \ g^{-1}$ and differed less among stations than did total concentrations. Slight arsenic enrichment was evident in North Bay compared with South Bay sediments. The



Fig. 6. Mean total arsenic (\odot) and mean extractable (0.5 N HC1) arsenic (\bullet) in fine (< 100 μ m) oxidized surface sediments from stations in San Francisco Bay (M1-M4, C1-C6, C8), Big River (M5), the lower San Joaquin (C7), Sacramento (C9), Tuolumne (C10), and middle San Joaquin Rivers (C12). Vertical bars represent 95% confidence intervals for the means. The number of samples analyzed is given below each bar or circle.

mean concentration of arsenic for all sediments from North Bay (C1–C8; $3.5 \pm 0.17 \ \mu g \ g^{-1}$) was higher than the mean concentration from all South Bay sediments (M1–M4; 2.2 ± 0.38 , mean \pm one standard error). Differences in sediment arsenic concentrations between C7 and C1–C6 were similar to differences in arsenic concentration in *Corbicula* at these stations.

Neither HCl-extractable arsenic nor total arsenic in sediments correlated significantly with arsenic in *Corbicula* or *Macoma*. The range of values was small and variances were similar in sediments and animals. The ratio of HCl-extractable arsenic to HCl-extractable iron has proved a useful predictor of arsenic availability to the bivalve *Scrobicularia plana* (Langston, 1980). Ratios calculated for Stations C1-C8 were very low (0.40-0.65) compared with those observed by Langston (1980), reflecting both low concentrations of total arsenic in the sediments and low concentrations of potentially biologically available arsenic.

DISCUSSION

Arsenic concentrations in whole soft tissues of both *Corbicula* and *Macoma*, and in fine sediments, appeared comparable to concentrations in bivalves and sediments from uncontaminated estuaries. *Scrobicularia plana* from United Kingdom estuaries not influenced by metalliferous mining wastes contained from 13 to $24 \ \mu g \ g^{-1}$ arsenic in whole soft tissues (Langston, 1980). Similar concentrations of arsenic, 6.6–16.6 $\mu g \ g^{-1}$, were found in *Macoma balthica* in uncontaminated estuaries (Bryan et al., 1985; Langston, 1985). In contaminated estuaries, arsenic concentrations as high as 65.5 $\mu g \ g^{-1}$ were observed in *Macoma balthica* (Langston, 1986).

Langston (1980) reported near-total arsenic concentrations ranging from 2 to 16 μ g g⁻¹, similar to observations in this study, in sediments of uncontaminated estuaries in the United Kingdom. Offshore sediments from Lake Michigan contained from 5.2 to 9.2 μ g g⁻¹ arsenic (Christensen and Chien, 1979). Similarly, total arsenic concentrations in sediments of 10 Saskatchewan lakes ranged from 2.7 to 13.2 μ g g⁻¹ (Huang and Liaw, 1978).

Studies conducted more than 10 years ago in San Francisco Bay also show arsenic concentrations similar to those found here. *Macoma balthica* near naval shipyards in North Bay contained arsenic concentrations ranging from 8.9 to 16.3 μ g g⁻¹, with most values between 10 and 13 μ g g⁻¹ (Anderlini et al., 1975). *Mytilus edulis* in central and southern San Francisco Bay contained from 5 to 12 μ g g⁻¹ arsenic in whole soft tissues (Anderlini et al., 1975; Risebrough et al., 1977). Sediment samples from several South San Francisco Bay sites contained 5–7.9 μ g g⁻¹ arsenic (Risebrough et al., 1977). Arsenic concentrations in sediments collected near naval shipyards were typically 10–14 μ g g⁻¹, but reached 26 μ g g⁻¹ (Anderlini et al., 1975; Risebrough et al., 1977). Total arsenic at C7 was similar to concentrations reported more recently for fine (< 63 μ m) bed sediments of the San Joaquin River (median value, 9.8 μ g g⁻¹; Clifton and Gilliom, 1989). These data suggest, unlike other trace elements (Luoma and Phillips, 1988), large inputs of biologically available arsenic do not appear to be common within San Francisco Bay. Slight arsenic enrichment was indicated in both sediments and animals in Suisun Bay (Fig. 1). This enrichment appears to be local in origin. Low arsenic concentrations at stations on the San Joaquin (C12), Sacramento (C9), and Tuolumne (C10) Rivers also suggest a lack of significant arsenic inputs from agricultural return water and small riverine inputs of bioavailable arsenic in general.

Temporal fluctuations in arsenic of *Corbicula* and *Macoma* were small, but showed some similarities to biologically-driven fluctuations of other metals (Cain and Luoma, 1986; Luoma et al., 1990). Animals size explained only a small portion of variance in arsenic concentrations in *Macoma*, probably reflecting a lack of significant levels of bioavailable arsenic at the *Macoma* stations. In the northern reach of the estuary where tissue arsenic concentrations in *Corbicula* suggest slight arsenic enrichment, animal size may have more importance, as reflected in the increased occurrence, although variable in magnitude, of positive relationships between *Corbicula* animal size and arsenic concentration in tissues.

A second possibility for the small temporal fluctuations relates to seasonal reproductive cycles. In *Scrobicularia*, arsenic was rapidly incorporated into gonadal tissues and shed during spawning, although in *Mytilis* gonadal development diluted total arsenic concentrations (Langston, 1984). Arsenic content of gonadal tissue as a percentage of total body arsenic in *Scrobicularia* ranged from 6.4 to 20.5% over a season. If fluctuations of the same magnitude occur in *Macoma* or *Corbicula*, this might contribute to the fluctuations of tissue arsenic concentration observed in these species.

SUMMARY

Arsenic concentrations in *Corbicula* and *Macoma balthica* in San Francisco Bay and the Sacramento/San Joaquin Delta are generally consistent with concentrations found in uncontaminated systems, despite the occurrence of arsenic-rich marine shales in the watershed. Lower arsenic concentrations occur in both *Corbicula* and fine sediments in the lower San Joaquin River stations than in the urbanized Suisun Bay/Delta. However, the generally low arsenic concentrations in bivalves and fine sediment indicate that local and regional inputs of arsenic are minor at the head of this estuary and do not measurably influence the distribution of arsenic in bivalves or fine sediments throughout the Bay and delta.

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