14. Mercury in lower trophic levels of the Clear Lake Aquatic Ecosystem, California: T.H. Suchanek et al. 2000

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14 Mercury in Lower Trophic Levels of the Clear Lake Aquatic Ecosystem, California

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ABSTRACT

Total and methyl mercury were analyzed in plankton and benthic invertebrates from Clear Lake, CA, an aquatic ecosystem contaminated from mining at the Sulphur Bank Mercury Mine over an 84-year period. Sediment total (primarily inorganic) mercury concentrations exceed 180,000 ng/g near the mine. Total mercury in Clear Lake biota (up to 855 ng/g in plankton, 41,671 ng/g in oligochaetes, and 27,686 ng/g in chironomids) was found to reflect the concentration of mercury in the organisms' surroundings (water or sediment). Methyl mercury, however (up to 67 ng/g in plankton, 19.9 ng/g in oligochaetes, and 61.9 ng/g in chironomids), typically was not correlated with inorganic mercury concentrations in water and

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sediment. Anomalously high concentrations of methyl mercury in biota at great distances from the point source of inorganic mercury suggests either (1) methyl mercury is produced *in situ* at sites with low inorganic mercury or (2) methyl mercury is produced in regions with high inorganic mercury and transported to other regions of Clear Lake by wind-driven currents. An increasing ratio of methyl to total mercury with increasing trophic level supports a bioaccumulation model for methyl mercury dynamics in Clear Lake biota and suggests that bioavailable mercury increases as a function of distance from the mine. Compared with other contaminated sites, Clear Lake's plankton are relatively low in both total mercury and methyl mercury, despite the fact that sediments in this system are some of the most highly contaminated in the world. Benthic invertebrates at Clear Lake exhibit the highest total mercury values (by one order of magnitude higher than any other reported sites), yet have the lowest methyl mercury concentrations of any known contaminated sites.

Keywords: *Clear Lake, mercury, mining, invertebrates, aquatic*

INTRODUCTION

Concern over mercury (Hg) bioaccumulation has stimulated substantial interest in Hg contamination in natural ecosystems and the relative efficiency of methyl Hg production and transport through trophic pathways.[1] While there has been considerable research on Hg in higher trophic level species such as predatory fishes and birds, little information exists on Hg in lower trophic levels. This information is critical to understanding the dynamics of Hg bioaccumulation, predicting the flow of Hg through various trophic levels, modeling systems contaminated with Hg, and providing potential solutions to problems associated with natural and anthropogenic sources of Hg. In addition, since lower trophic level organisms are more easily sampled than individuals at higher trophic levels, and since such collections have little impact on the populations of these organisms, it seems logical to use them as indicators of Hg contamination in the ecosystem as a whole. Here we present data on total and methyl Hg in plankton and benthic invertebrates from a highly contaminated aquatic ecosystem in Clear Lake, CA.

Clear Lake is a shallow, polymictic lake in the Northern California Coast Range. Mining of Hg ore from the Sulphur Bank Mercury Mine (periodically from about 1873 to 1957) resulted in the deposition of an estimated 100 metric tons of Hg into the Clear Lake aquatic ecosystem.[2] See Suchanek et al.[3-5,35] and Richerson et al.[6] for a more complete description of Clear Lake. Clear Lake is highly eutrophic with abundant phytoplankton (especially scum-forming cyanobacteria), zooplankton, and benthic invertebrates. In a preliminary study by the U.S. Environmental Protection Agency (USEPA), Hg levels in wet phytoplankton ranged from 0.6 to 160 ng/g, or about 160 to 700 ng/g on a dry weight basis.[7] To our knowledge, no previous study has assessed Hg levels in Clear Lake zooplankton or benthic invertebrates. Fishes from Clear Lake, specifically largemouth bass and channel catfish, also have been shown to contain Hg levels that exceed California state and federal health standards for safe consumption (i.e., ≥0.5 mg/kg).[8,9]

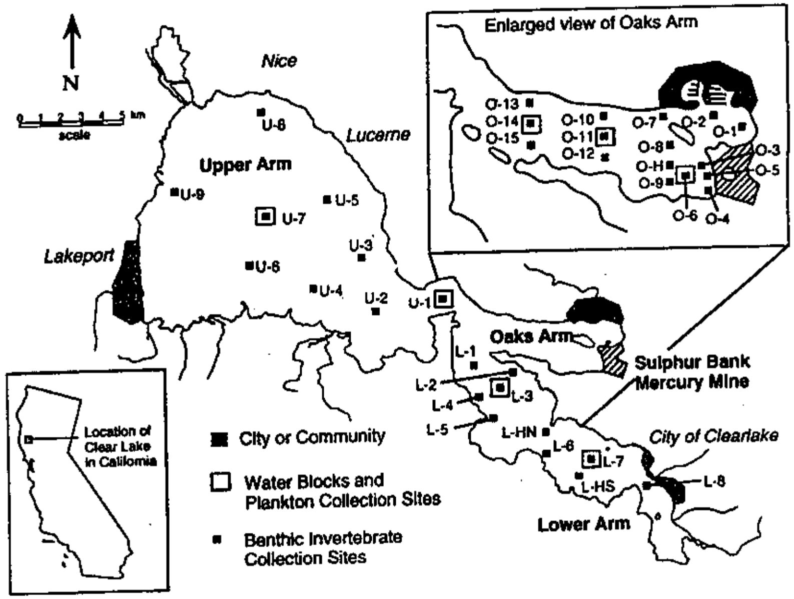

FIGURE 1 Map of Clear Lake and sampling sites.

The primary objective of this chapter is to describe the distribution of total and methyl Hg within the lower trophic levels, primarily phytoplankton, zooplankton, and benthic invertebrates (oligochaetes and chironomids) of the Clear Lake aquatic ecosystem. These results represent only a portion of a baseline study conducted as a part of an Ecological Assessment of the USEPA Region IX Sulphur Bank Mercury Mine Superfund Site.[3,35] Concentrations of Hg within the abiotic components (i.e., sediments and water) of this ecosystem are given in Suchanek et al.,[5] while Slotton et al.[9] present Hg concentrations in Clear Lake fishes. Effects of Hg on invertebrate populations and community structure are presented in Suchanek et al.[4] Taken collectively, these data represent a snapshot in time of the distribution of Hg in both abiotic and biotic components.

METHODS

All collections were made during July to November 1992. Plankton were collected from 7 water block stations throughout Clear Lake, while benthic invertebrates were collected from 35 benthic stations (Figure 1). Descriptive statistics (distance from the mine, depth, sediment grain size, total organic carbon, total Hg, methyl Hg, methyl to total ratio in sediments and water, plus standard limnological parameters) on these same sampling stations are provided in Suchanek et al.[3,5,35]

Plankton were collected (mesh size = 160 μm) over a 0.5-km² area centered on each of the seven stations until approximately 100 g was obtained. The resulting

samples consisted of a mixture of approximately 90% phytoplankton (mostly blue-green "algae" = cyanobacteria) and 10% zooplankton. A subsample of this "total plankton" (the mixture of phytoplankton and zooplankton) was analyzed at each site. Zooplankton samples for Hg analysis were obtained by separating out zooplankton from the remainder of the total plankton samples by washing the total plankton samples through a 300-μm mesh screen using deionized water. During the plankton sampling periods, phytoplankton consisted primarily of *Anabeana*, *Gleotrichia*, and *Microcystis*, whereas the zooplankton consisted primarily of *Daphnia*, *Bosmina*, *Ceriodaphnia*, and *Diaphanosoma* (Norm Anderson, Lake County Mosquito and Vector Control, personal communication). Both total plankton and zooplankton samples were transferred to acid-washed bottles, dried in an evacuating drying oven at <60°C, and weighed before and after drying. Dried samples were homogenized with Teflon™ utensils, resulting in uniform powders.

Benthic invertebrates (oligochaetes and chironomids) were collected using 6-in. Ekman grab samples and sieved with a 0.5-mm mesh screen. Samples were sorted and placed in acid-washed glass containers with water and shipped live (on ice) for Hg analysis. Because of taxonomic complexities, oligochaetes were not identified to species, but previously collected specimens from Clear Lake are known to include numerous genera: *Aulodrilus*, *Bothrioneurum*, *Branchiura*, *Dero*, *Ilyodrilus*, *Limnodrilus*, *Potamothrix*, *Tubifex*, and *Varichaetadrilus* (Art Colwell, Lake County Mosquito and Vector Control, personal communication). Chironomid larvae consisted primarily of several species of the genus *Chironomus* (*C. plumosus*, *C. frommeri*, and *C. decorus*).

For comparisons of Hg concentrations in biota with water from various locations within the lake, raw (unfiltered) water included all particulates. Filtered water was passed through a 0.45-μm filter (see Suchanek et al.[5] for details).

Analytical Laboratory Procedures

Plankton were analyzed for total Hg at all seven water block stations; methyl Hg was only analyzed for a subset of four stations. Total Hg in benthic invertebrates was analyzed at all 35 stations; methyl Hg was analyzed for a subset of 10 stations. Total Hg and methyl Hg for all tissues were analyzed by Brooks Rand Ltd. (Seattle, WA). Total Hg in plankton and benthic invertebrates was analyzed by dual amalgamation/cold vapor atomic fluorescence spectrometry.[10] Methyl Hg was analyzed in biotic samples utilizing aqueous phase ethylation, followed by cryogenic gas chromatography with cold vapor atomic fluorescence detection.[19]

Field collection and analytical methods for Hg in unfiltered and filtered water are described in detail in Suchanek et al.[5] All raw data for abiotic and biotic components are presented in Suchanek et al.[3]

Data Reduction and Statistical Analyses

All data analysis was performed on an Apple Macintosh® platform. Preliminary plotting and calculation of simple regression statistics for curve-fitting routines were accomplished using the data management and statistical visualization program JMP® (SAS Institute, Inc.). Curve-fitting and R^2 values were calculated using the presentation graphics program Delta Graph® (DeltaPoint, Inc.). Step-wise multiple linear regression analyses were performed using the statistical package DataDesk® (Data Description, Inc.).

Data for independent variables on physical environmental parameters (such as concentrations of Hg in water and sediments) used in multiple regression analyses (see Table 1) were taken from Suchanek et al.[5]

RESULTS

Plankton

Plankton exhibited total Hg concentrations (69 to 855 ng/g for total plankton and 80 to 661 ng/g for zooplankton) about four to five orders of magnitude higher than the water from which they were collected (Figure 2A). Total Hg in both types of plankton declined with distance from the mine, though concentrations at the site most distant from the mine were slightly higher than expected. Using curve-fitting routines, this decline was best described by second order polynomials, which explained about 73% of the variability in the zooplankton data and about 93% of the variability in the total plankton data. Multiple regression analyses revealed that for total Hg in both total plankton and zooplankton, a positive relationship was observed with methyl Hg in unfiltered surface water (Table 1). This relationship was much stronger in the total plankton (p <0.001) than in zooplankton (p <0.05). Zooplankton total Hg concentrations were also found to be positively correlated to surface water conductivity values (p <0.05).

Methyl Hg concentrations in plankton (10 to 29 ng/g for total plankton and 27 to 67 ng/g for zooplankton) were five to six orders of magnitude higher than the unfiltered water from which they were collected (Figure 2B). These data show only a slight decline with increasing distance from the mine, with the site furthest from the mine showing slightly elevated methyl Hg concentrations as compared to those sites at moderate distances, a trend similar to that of the total Hg data. As a result of this trend, plankton methyl Hg data was best described by second-order polynomials, explaining about 93% of the variability in zooplankton and 98% of the variability in total plankton. Multiple regression analysis on methyl Hg in total plankton yielded no significant relationship to any of the independent variables tested (Table 1). Total Hg in zooplankton, however, was found to have a moderately negative correlation with total suspended solids in surface waters (p <0.01) and a weak positive correlation with surface conductivity.

Although total Hg concentrations in zooplankton and total plankton were comparable, methyl Hg levels were considerably higher in zooplankton, likely a reflection of their higher trophic status. The ratio of methyl to total Hg ranged from 0.04 to 0.18 (4 to 18%) in total plankton and from 0.09 to 0.46 (9 to 46%) in zooplankton (Figures 2C and 4). Both sets of ratios were several orders of magnitude higher than the ratios of methyl to total Hg in surface waters (0.004 to 0.017 ng/L in filtered water and 0.003 to 0.007 ng/L in unfiltered water), and both increased significantly with distance from the mine. Results of the multiple regression analysis showed a moderately negative correlation (p <0.01) of the ratio of methyl to total Hg in total plankton with total Hg in filtered surface water (Table 1). Zooplankton Hg ratios were found to be most significantly (and negatively) correlated with the depth of a site (p >0.01).

TABLE 1
Results of Multiple Regression Analyses

					Independent Variables																		
					Site		Sediment					Water											
Dependent Variables	Number of Samples Collected	Number of Samples in Model	Whole Model Significance	Whole Model R^2	Distance from Mine	Depth	Total Hg	Methyl Hg #	Methyl/Total Hg Ratio	Grain Size	TOC	Surface Total Hg — Unfiltered	Surface Total Hg — Filtered	Surface Methyl Hg — Unfiltered	Surface Methyl Hg — Filtered	Surface TOC — Unfiltered	Surface TOC — Filtered	Surface TSS	Surface Temperature	Surface pH	Surface Conductivity	Surface D.O.	Surface Eh
Total plankton-total Hg	7	7	***	0.98	ns	ns	°	°	°	°	°	ns	ns	***/+	ns	ns	ns	ns	ns	ns	ns	ns	ns
Total plankton-methyl Hg	4	4	ns	0.73	ns	ns	°	°	°	°	°	ns	ns	ns	ns	ns	ns	ns	ns	ns	ns	ns	ns
Total plankton-methyl/total Hg ratio ◊	4	4	**	0.99	ns	ns	°	°	°	°	°	ns	**/–	ns	ns	ns	ns	ns	ns	ns	ns	ns	ns
Zooplankton-total Hg	7	7	*	0.75	ns	ns	°	°	°	°	°	ns	ns	*/+	ns	ns	ns	ns	ns	ns	*/+	ns	ns
Zooplankton-methyl Hg	4	4	*	1.00	ns	ns	°	°	°	°	°	ns	ns	ns	ns	ns	ns	**/–	ns	ns	*/+	ns	ns
Zooplankton-methyl/total Hg ratio ◊	4	4	**	0.99	ns	**/–	°	°	°	°	°	ns	ns	ns	ns	ns	ns	ns	ns	ns	ns	ns	ns
Chironomids-total Hg #	35	31	***	0.56	ns	ns	***/+	ns	°	ns	ns	°	°	°	°	°	°	°	°	°	°	°	°
Chironomids-methyl Hg	10	10	ns	0.26	ns	ns	ns	ns	°	ns	ns	°	°	°	°	°	°	°	°	°	°	°	°
Chironomids-methyl/total Hg ratio ◊	10	7	**	0.97	ns	†/+	ns	*/+	**/+	ns	ns	°	°	°	°	°	°	°	°	°	°	°	°
Oligochaetes-total Hg	35	33	***	0.37	ns	ns	***/+	ns	°	ns	ns	°	°	°	°	°	°	°	°	°	°	°	°
Oligochaetes-methyl Hg #	10	7	*	0.58	ns	ns	ns	*/+	°	ns	ns	°	°	°	°	°	°	°	°	°	°	°	°
Oligochaetes-methyl/total Hg Ratio ◊	10	7	**	0.68	ns	ns	**/+	ns	ns	ns	ns	°	°	°	°	°	°	°	°	°	°	°	°

Note: Significance levels from multiple regression analysis where † = $0.05 < p < 0.10$; * = $0.01 < p < 0.05$; ** = $0.001 < p < 0.01$; *** = $p < 0.001$; ns = not significant; ° = not tested; + = positive correlation; – = negative correlation; ◊ = variable logit transformed; # = variable log transformed.

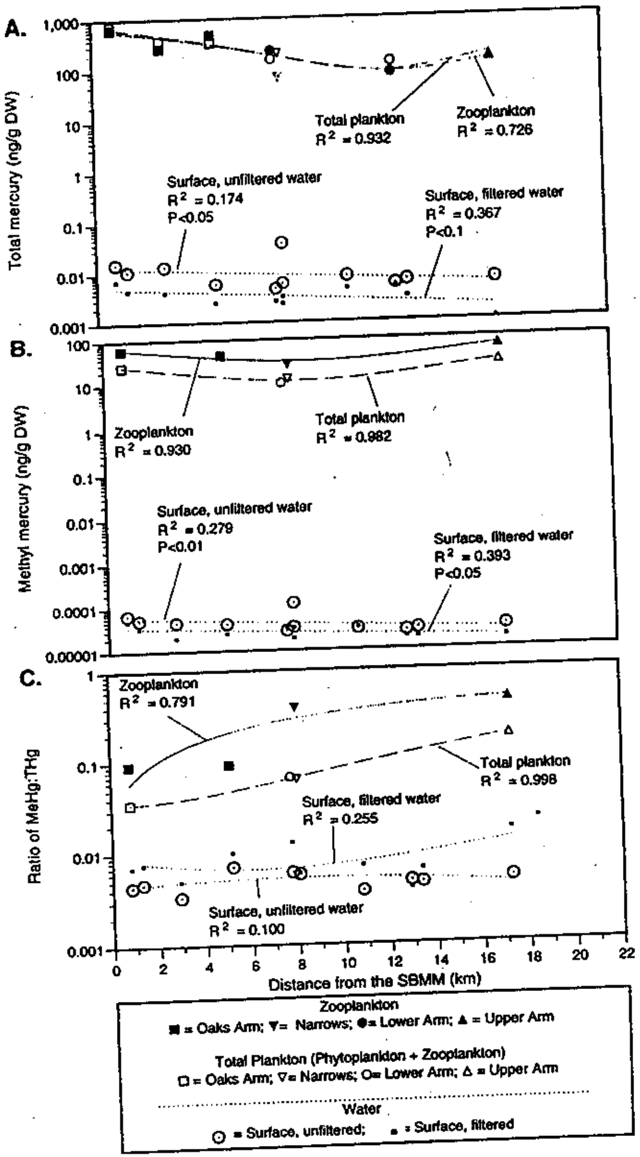

FIGURE 2 Plankton results: (A) total mercury; (B) methyl mercury; and (C) ratio of methyl to total mercury in zooplankton, total plankton, and raw and filtered surface water plotted against distance from the Sulphur Bank Mercury Mine. Note log scales. Curve fits are second-order polynomials for all. Water Hg data from Suchanek et al.[5]

BENTHIC INVERTEBRATES

Total Hg concentrations in oligochaetes and chironomids were statistically indistinguishable from each other, each exhibiting a significant exponential decline as a function of distance from the mine (Figure 3A). These values spanned over two orders of magnitude, from 316 ng/g at distant sites to 42,000 ng/g for sites less than 1 km from the mine, and were about one order of magnitude lower than the sediments from which these organisms were obtained. Because individual specimens were not purged before analysis, some of the total Hg could have been the result of sediments remaining in their guts at the time of analysis. However, the entire body burden of Hg, including gut contents, is probably a more realistic value to use when evaluating bioaccumulation pathways. Using multiple regression analysis, concentrations of total Hg in both chironomids and oligochaetes were best explained by the concentrations of total Hg in the sediments from which they were derived ($p < 0.001$), and no other factors tested were significant.

Methyl Hg ranged from 5.3 to 19.9 ng/g in oligochaetes and from 5.2 to 61.9 ng/g in chironomids (Figure 3B). In oligochaetes, this trend was best explained by an exponential decline as a function of distance from the mine, but it was not significant and exhibited a very low R^2 (0.186). In contrast, methyl Hg in chironomids was best fit with a second order polynomial curve ($R^2 = 0.677$), though this fit depends greatly upon the single datum in the Upper Arm at U-7, the furthest sampling station from the mine (Figure 3B). Unfortunately, this was the only sampling site for methyl Hg in the Upper Arm. Invertebrate methyl Hg concentrations were, at most, only about one order of magnitude higher than the methyl Hg values in the sediments from which they were derived. Multiple regression analysis on methyl Hg in chironomids yielded no significant relationship to any of the independent variables tested. Oligochaete methyl Hg concentrations showed only a weak positive correlation with sediment methyl Hg concentrations.

The percentage of total Hg as methyl in both invertebrate taxa were considerably lower than those for plankton and not much different than those values for water and sediment (see Suchanek et al.[5]), with values ranging from 0.3 to 1.4% for oligochaetes and from 0.6 to 6.0% for chironomids (Figures 3C and 4). Interestingly, the highest methyl to total Hg ratio was observed at U-7, a site over 17 km from the mine. Since no attempts were made to void oligochaete guts before Hg analyses, it is unclear whether the ratio of methyl to total Hg in these species was significantly influenced by the total Hg levels of sediment remaining in their guts. Chironomid larvae, benthic filter feeders, also reflected sediment levels of total Hg in their environment. The percentage of methyl Hg in chironomids, however, was generally higher (Figures 3C and 4) than in oligochaetes. This is probably a reflection of (1) their higher trophic position and (2) the fact that Hg levels in oligochaetes may have been influenced by residual sediments in their guts. Results of multiple regression analyses on the ratio of methyl to total Hg in chironomids revealed a moderately positive correlation ($p > 0.01$) with the ratio of methyl to total Hg in sediment and a weaker positive correlation ($p < 0.05$) with sediment methyl Hg (Table 1). The ratio of methyl to total Hg in oligochaetes was found to have a moderately positive correlation ($p < 0.01$) with sediment total Hg concentrations.

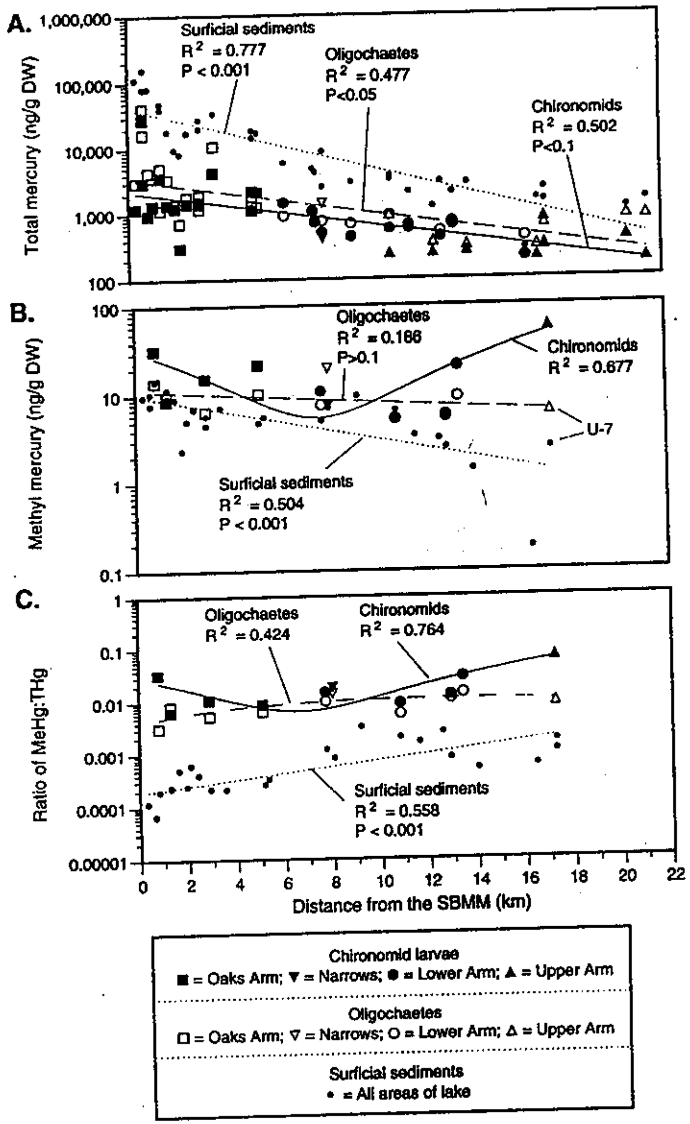

FIGURE 3 Benthic invertebrate results: (A) total mercury; (B) methyl mercury; and (C) ratio of methyl to total mercury in chironomids, oligochaetes, and surficial sediments. Note log scales in A and B. Curve fits are as follows: (A) all are exponential; (B) sediment and oligochaete data are fit with exponential fits, and chironomid data are second-order polynomial; and (C) sediment data are fit with an exponential fit and oligochaete and chironomid data with second-order polynomial. Sediment data from Suchanek et al.[5]

DISCUSSION

The declines of total Hg with distance from the mine in Clear Lake plankton and benthic invertebrates probably result from comparable declines in sediment and water concentrations of inorganic Hg. As shown in Suchanek et al.[5] abiotic components of the ecosystem (sediment and water) from Clear Lake exhibited classic point source distributions of total and methyl Hg with maximum concentrations occurring at sites closest to the mine. The correlation of biotic total Hg levels with total Hg levels in the abiotic environment is particularly evident in the results of the multiple regression analyses presented in Table 1. Methyl Hg burdens in Clear Lake biota, on the other hand, were not correlated with inorganic Hg concentrations in the abiotic environment. The relatively high methyl Hg concentrations in biota collected at sites distant from the mine indicate that methyl Hg burdens may be better explained by considering factors relating to bioavailability or methylation potential. Further evidence that methyl Hg is more readily available at sites further from the mine was seen in the studies of the abiotic components of Clear Lake.[5] That study found that the ratio of methyl to total Hg in sediment and the particulate fraction of water increased with distance from the mine. The conclusions of Suchanek et al.[5] that there was either (1) differential downgradient transport of methyl Hg or (2) preferential production of methyl Hg at sites distant from the mine are consistently reflected in the biotic data presented here.

When comparing Hg concentrations in lower trophic biota from Clear Lake to those from other systems throughout the world, we note some interesting trends (Table 2). Total Hg in Clear Lake zooplankton, which ranged from 80 to 661 ng/g, was relatively low compared to other lake systems where total Hg ranged over 3000 ng/g.[14,15] It should be noted that, like plankton, Hg concentrations (0.005 to 0.05 µg/L) in Clear Lake water, in comparison with other studies (0.0003 to 0.39 µg/L), are also at the low end of the range.[5] Though fewer studies have measured methyl Hg in plankton, Clear Lake plankton, with methyl Hg concentrations of 10 to 29 ng/g in total plankton and 27 to 67 ng/g in zooplankton, are typically much lower than those values reported in most other studies, documenting 32 to 220 ng/g in phytoplankton[16,17] and 260 ng/g in zooplankton.[17] Clear Lake plankton typically ranged from 3 to 18% methyl Hg for total plankton and 4 to 83% methyl Hg for zooplankton, much lower than those values typically reported from other contaminated and uncontaminated sites worldwide which often ranged from 70 to 100% methyl Hg.[20,24,32]

In contrast to the plankton, total Hg in Clear Lake oligochaetes (416 to 41,671 ng/g) and chironomids (194 to 27,686 ng/g) were much higher than the typical range (71 to 4900 ng/g) found in other studies, both freshwater and marine.[12,16-20] The elevated inorganic Hg in these organisms could be a reflection of the higher sediment Hg concentrations in Clear Lake (270 to 183,000 ng/g) relative to the other study sites (0 to 30,000 ng/g). Methyl Hg concentrations in Clear Lake oligochaetes (5.3 to 19.9 ng/g) and chironomids (5.2 to 61.9) were about one order of magnitude lower than the typical range (52 to 1000 ng/g) for benthic invertebrates from other sites.[16,17,29] Interestingly, Clear Lake sediments contain the highest reported sediment

TABLE 2
Comparative Data on Total Hg, Methyl Hg, and Percent of Methyl Hg in This Study with Those Reported from the Literature

			Organisms			Sediment	Raw Water	
	Location	Contamination Source	Total Hg Range (Avg) ng/g = ppb	Methyl Hg Range (Avg) ng/g = ppb	Methyl Hg as a Percent of Total Hg	Total Hg Range (Avg) ng/g = ppb	Total Hg Range (Avg) mg/L = ppb	Reference
			Plankton					
freshwater								
lake phytoplankton	Onondaga L., NY	Wastewater	—	32	24	—	0.0003	17
lake phytoplankton	Onondaga L., NY	Wastewater	1830@	220*	12	n.d.–30,000	—	16
lake phytoplankton	El-Temsah L., Egypt	Wastewater	876–2,170 (904)	—	—	1,060–4,360 (3,150)	0.11–0.39 (0.13)	14
lake zooplankton	Clear Lake, CA	Hg mine	80–661	27–67	9–83	270–183,000	0.005–0.05	This study and 5
lake zooplankton	Onondaga L., NY	Wastewater	650@	260	40	—	0.0003	17
lake zooplankton	Little Rock Lake, WI	Atmospheric	45	—	—	<0.1	0.00012	1
lake zooplankton	Lakes in Sweden	Atmospheric	—	—	45–85	90–300	—	20
lake zooplankton	Davis Creek Reservoir, CA	Gold mining	830–4,100	—	—	500–150,000	0.006	15
lake zooplankton	El-Temsah L., Egypt	Wastewater	1,060–3,170 (1,230)	—	—	1,060–4,360 (3,150)	0.11–0.39 (0.13)	14
lake zooplankton	Lakes in Finland	Atmospheric	—	—	45–100 (80)	—	—	24
lake zooplankton	Lakes in Ontario, Canada	Atmospheric	—	19–448	—	—	—	30
lake zooplankton	Natural Lakes in Quebec, Canada	Atmospheric	~75–350 (193)	~55–175	15–50	—	0.2	32,38
lake zooplankton	New reservoirs in Quebec, Canada	Atmospheric	110–300 (185)	150–850	45–85	—	0.2	32,38
lake zooplankton	Lakes in Ontario and Quebec, Canada	Atmospheric	26–377 (108)	—	—	3–267 (80)	—	28
stream zooplankton	Mountain streams, TN	Atmospheric	—	—	23–100 (75)	—	—	25
total lake plankton	Clear Lake, CA	Hg mine	69–855	10–29	3–18	270–183,000	0.005–0.05	This study and 5
lake plankton (>10 µm)	Churchill R., Canada	New reservoir	<10–2,110	—	—	6,000–14,000	0.022–0.036	12
lake plankton (>73 µm)	Churchill R., Canada	New reservoir	<10–1,230	—	—	6,000–14,000	0.022–0.036	12
marine								
marine phytoplankton	Pelagic ocean	Atmospheric	410	—	—	—	—	
marine phytoplankton	Kastela Bay, Yugoslavia	Chlor-alkali	—	—	—	90–1,370	—	13
marine zooplankton	Kastela Bay, Yugoslavia	Chlor-alkali	—	—	—	90–1,370	—	13
marine zooplankton	Lavaca Bay, TX	Chlor-alkali	100–200	—	—	—	—	26
marine zooplankton	Pelagic ocean	Atmospheric	55–388 (191)	—	—	—	—	36
marine zooplankton	Pelagic ocean	Atmospheric	39–448 (134)	—	—	—	—	37

TABLE 2 (continued)
Comparative Data on Total Hg, Methyl Hg, and Percent of Methyl Hg in This Study with Those Reported from the Literature

	Location	Contamination Source	Organisms			Sediment	Raw Water	Reference
			Total Hg Range (Avg) ng/g = ppb	Methyl Hg Range (Avg) ng/g = ppb	Methyl Hg as a Percent of Total Hg	Total Hg Range (Avg) ng/g = ppb	Total Hg Range (Avg) mg/L = ppb	
			Benthic Invertebrates					
freshwater								
oligochaetes	Clear Lake, CA	Hg mine	416–41,671 (3,000)	5–20 (9)	0.3–4.8	270–183,000	0.005–0.05	This study and 5
chironomids	Clear Lake, CA	Hg mine	194–27,686 (2,000)	5–62 (20)	0.6–5.8	270–183,000	0.005–0.05	This study and 5
amphipods	Onondaga L., NY	Wastewater	200–1,300 (397)	52–338 (103)	ca. 27	—	0.0003	17
chironomids	Onondaga L., NY	Wastewater	300–1,800 (790)	108–486	ca. 27	—	0.0003	17
amphipods	Onondaga L., NY	Wastewater	210–300	100–150#	0.5	0–30,000	—	16
chironomids	Onondaga L., NY	Wastewater	350–1,900	170–1,000#	0.5	0–30,000	—	16
insects (detritivores/ grazers)	Duncan Lake, Quebec, Canada	Atmospheric	120–143 (129)	14–61 (40)	10–50	36–59	—	29
insects (grazers/ predators)	Duncan Lake, Quebec, Canada	Atmospheric	136–256 (189)	102–124 (111)	49–75	36–59	—	29
insects (detritivores/ grazers)	LaGrand complex, Quebec, Canada	New reservoir	139–1,020 (417)	64–106 (76)	6.3–76	33–275	—	29
chironomids	LaGrand complex, Quebec, Canada	New reservoir	285–1,075 (946)	64–76 (71)	6–26	33–275	—	29
chironomids	Lakes in Ontario, Canada	Atmospheric	97–177 (158)	—	—	28–311 (134)	0.0015–0.0022	33
oligochaetes	Lakes in Ontario, Canada	Atmospheric	164	—	—	28–311 (134)	0.0015–0.0022	33
amphipods	Lakes in Ontario, Canada	Atmospheric	98–158 (128)	—	—	28–311 (134)	0.0015–0.0022	33
Sergentia (detritivore)	Lakes in Sweden	Atmospheric	71–4,680	—	45–85	90–300	—	20
Chironomus (detritivore)	Lakes in Sweden	Atmospheric	115–1,350	—	45–85	90–300	—	20
Cryptochironomus (predator)	Lakes in Sweden	Atmospheric	45	—	45–85	90–300	—	20
Procladius (predator)	Lakes in Sweden	Atmospheric	42–173	—	45–85	90–300	—	20
chironomids	Churchill R., Canada	New reservoir	1,400–1,500*	—	—	6,000–14,000	0.022–0.036	12
oligochaetes	Churchill R., Canada	New reservoir	nd–4,900*	—	—	6,000–14,000	0.022–0.036	12
annelid	Lakes	Atmospheric	500–1,500	20–180*	4–36	—	—	27
marine								
nereis (polychaete)	Nissum Broad, Denmark	Chlor-alkali	225–1,350	—	—	100–22,000	—	18,19
arenicola (polychaete)	Nissum Broad, Denmark	Chlor-alkali	335–760	—	—	100–22,000	—	18,19
maldanes (polychaete)	Novaya Zemlya, Barents Sea	Atmospheric?	130–450	80–340	18–75	—	0.002–0.015	31,34

Note: All values presented as dry weight unless specified otherwise. — = data not available; * = value calculated from wet weight data with a 7:1 wet/dry ratio; @ = value derived from methyl Hg and ratio values; # = value derived from total Hg and ratio values; nd = not detected.

inorganic Hg contamination (over 183,000 ng/g), yet methyl Hg concentrations in benthic invertebrates were typically one to two orders of magnitude lower than all other reported values for contaminated and uncontaminated sites alike (Table 2). The fact that methyl Hg concentrations in Clear Lake benthic invertebrates are low despite high total Hg concentrations may be due to several factors that may inhibit either methylation or bioaccumulation. Such factors might include (1) the high level of productivity of Clear Lake, (2) the alkaline nature of the lake, or (3) the high concentration of sulfides in water and sediments.

Among nonvertebrate biota in Clear Lake, inorganic Hg was found to dominate the total Hg burden, accounting for 75 to >99% of the total, as determined from the methyl to total Hg ratios (Figure 4). A trend of increasing proportion of methyl Hg with increasing trophic rank is consistent with many previous reports (e.g., References 16, 21, 22, and 29). Comparing methyl Hg in biota to environmental concentrations, especially in contrast to total Hg, highlights the influence of bioaccumulation. Each biotic group had methyl Hg burdens at the level of their surrounding medium or higher. Plankton had the highest differential, with methyl Hg concentrations five to six orders of magnitude higher than those in water; benthic invertebrates (with methyl Hg levels only slightly higher than that of the sediment) had the lowest.

Unlike the results from Clear Lake, studies of Hg in total plankton (zooplankton and phytoplankton combined) in other aquatic systems have not found direct positive correlations with Hg levels in water.[12,23] Studies of phytoplankton alone seem to have more varied results. Jackson[12] reported no relation between Hg in phytoplankton vs. water, yet he found inverse relationships of Hg in phytoplankton compared to environmental concentrations of total Hg, methyl Hg, and Hg methylation rate. Moreover, Hg levels in plankton correlated positively with concentrations of sulfides and suspended organic matter, which are thought to influence the bioavailability of Hg. Jackson[12] concluded that Hg concentrations in plankton reflect the bioavailability of Hg rather than the total concentration of Hg in the environment. Westcott and Kalff[30] showed that zooplankton methyl Hg was best predicted by water color and pH, being positively correlated with water color and negatively correlated with pH.

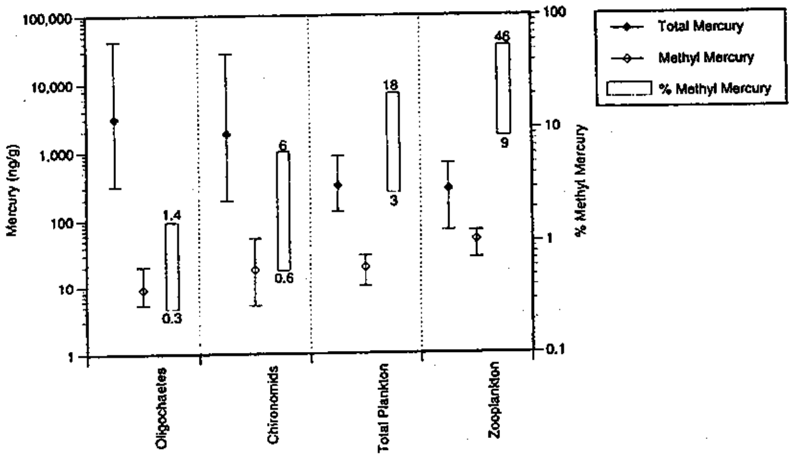

FIGURE 4 Means and ranges of total Hg, methyl Hg, and percent methyl Hg in oligochaetes, chironomids, total plankton, and zooplankton in Clear Lake.

In other field studies where Hg concentrations in benthic invertebrates were assessed, some found that chironomid Hg body burdens correlated with sediment Hg levels,[16,17] whereas others did not.[12,20] Like Clear Lake, the Lake Onondaga system studied by Becker et al.[16] and Becker and Bigham[17] had fairly high surficial sediment Hg levels (up to 30,000 ng/g), yet rather low chironomid Hg concentrations (350 to 1900 ng/g). Jackson,[12] working in the Churchill River, a system with only moderately contaminated sediments (6000 to 14,000 ng/g), found chironomid Hg burdens to have strongest correlations with sediment total organic carbon (TOC) and iron (Fe), which influence the bioavailability of Hg. In Swedish lakes with sediments of relatively little contamination (90 to 300 ng/g), Parkman and Meili[20] showed that feeding habits had the strongest influence upon chironomid Hg. In these systems, profundal detritivores (including *Chironomus* sp.), as opposed to predators, had the highest concentrations of Hg. They also found correlations between chironomid Hg and pH (negative), water color (positive), and seasonal changes, all of which are factors that may influence Hg bioavailability.

SUMMARY

In comparison with other studies worldwide, Clear Lake plankton have relatively low total (primarily inorganic) and methyl Hg concentrations. In contrast, Clear Lake benthic invertebrates exhibit some of the highest concentrations of total Hg found at any previously studied aquatic ecosystems, yet some of the lowest comparable methyl Hg values. In general, total Hg in Clear Lake biota exhibited a clear signature of a point source pollutant originating from the Sulphur Bank Mercury Mine, but methyl Hg did not. While there were high methyl Hg levels in biota near the mine, there were also high levels in the far reaches of the Upper Arm, some 15 to 20 km from the mine. These elevated methyl Hg concentrations could be derived from methyl Hg that is (1) produced *in situ* at these distant sites (from relatively low concentrations of inorganic Hg) or (2) produced near the mine (at sites with very high inorganic Hg) and transported via water and/or particulate matter by wind-driven currents to sites distant from the mine.

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