

# MONITORING INTER-ANNUAL VARIABILITY REVEALS SOURCES OF MERCURY CONTAMINATION IN CLEAR LAKE, CALIFORNIA

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**Abstract:** Mercury (Hg) in the aquatic ecosystem of Clear Lake has been documented since the 1970s when fishes were found to have elevated levels of toxic methyl mercury (meHg). Mining practices at the Sulphur Bank Mercury Mine (active intermittently from 1872–1957) along the shoreline of Clear Lake included the bulldozing of waste rock and overburden ore into the shallow nearshore regions of the lake and the creation of steeply sloped piles of waste rock at the water's edge. This process, plus erosion of the waste rock piles, resulted in the accumulation of an estimated 100 metric tons of Hg in Clear Lake. A monitoring program to assess Hg in Clear Lake was established in 1992, and conducted continuously from 1994. Drought conditions in California had persisted for ca. 6 yrs prior to 1992, when the U.S. Environmental Protection Agency (USEPA) remediated the steeply sloped eroding waste rock piles, which appeared to reduce sediment Hg concentrations significantly. In April 1995, a white flocculent material was observed in Clear Lake adjacent to the mine and has been observed every year since, leading to the discovery of ongoing acid mine drainage (AMD), low pH fluids high in Hg and extremely high in sulfate. AMD is now believed to be the most likely cause of elevated meHg in Clear Lake. The discovery of this source of meHg production in Clear Lake, which will significantly influence remedial options, was only made possible by implementation of a diligent monitoring program.

**Keywords:** mercury, mining, monitoring, aquatic, acid mine drainage, floc, Clear Lake, remediation

## 1. Introduction

Clear Lake, California is the site of the Sulphur Bank Mercury Mine, designated as a United States Environmental Protection Agency (U.S. EPA) Superfund Site in 1990, primarily as a result of elevated mercury (Hg) concentrations in fishes (e.g., channel catfish and largemouth bass) discovered in the 1970s. Clear Lake has a surface area of ca. 17,500 ha, lies in a shallow basin in the Coast Range ca. 130 km north of San Francisco, drains a watershed of ca. 1,370 km<sup>2</sup> and is the largest lake completely contained within California at about 32 km in length and 5 km in width (see Figure 1). It is shallow (average depth ca. 8 m), warm (5–25°C), alkaline (pH ~ 8.0), hyper-eutrophic (with abundant cyanobacterial blooms, especially during sum-



mer), and its circulation is influenced significantly by wind-driven currents. A nearly continuous sequence of lakebed sediments dating back 480,000 yrs was retrieved from Clear Lake by the U.S. Geological Survey (USGS), making it the oldest continuous lake in North America (Sims et al. 1988). Clear Lake is also one of the most mercury (Hg) contaminated lakes in the world, with a steep gradient of sediment total Hg (totHg) concentrations ranging from ca. 450 mg kg<sup>-1</sup> (ppm) dry weight near the mine to <1.0 ppm about 23 km away (Suchanek et al. 1997, 1998ab).

The Sulphur Bank Mercury Mine began operation as a shallow sulfur mine in 1865, with a long but intermittent history of Hg mining, active from 1872–97, 1899–1902, 1915–18, 1927–47 and 1955–57 (Chamberlin et al. 1990). The mine is located along the shoreline of the Oaks Arm of Clear Lake (Figure 1) and mining operations were responsible for extending the shoreline 60–150 m into the lake from the original 1888 position through the dumping of large volumes of waste rock and mining overburden. Before the 1920s the Sulphur Bank mine was primarily an open cut and shaft mining operation, which had little contact with, and influence on, Clear Lake.

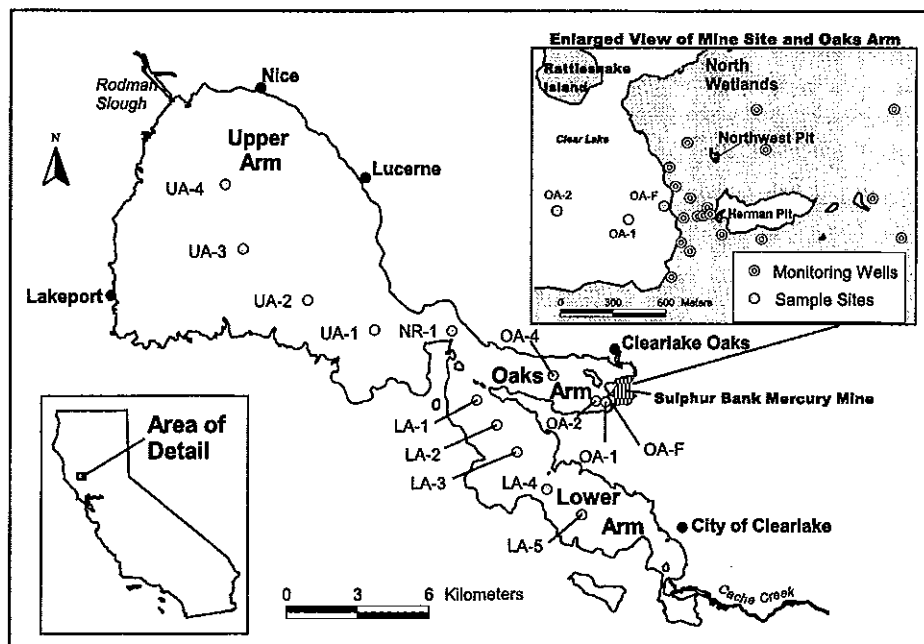


Figure 1. Monitoring locations in Clear Lake. Inset provides a close-up view of the eastern region of the Oaks Arm near the Sulphur Bank Mercury Mine showing the distribution of near-shore long-term lake monitoring stations and the location of monitoring wells on the mine site.

However, beginning in the 1920s, especially with the advent of mechanized earth moving equipment, large volumes of waste rock and overburden were removed from the central region of the mine site, creating what is known as the Herman Pit, which is 30 m deep with a volume of ca.  $1 \times 10^6 \text{ m}^3$ . Herman Pit water has a pH of ca. 3, a mean totHg concentration of  $0.54 \mu\text{g/ L}^{-1}$  ( $n=7$ ), and a mean sulfate concentration of  $2,721 \text{ mg L}^{-1}$  ( $n=14$ ). Dewatering the pit during mining was accomplished with pumps, routing water to Clear Lake, another mine site pit (Northwest Pit), and to the wetland immediately north of the mine site. The beginning of this process was also coincident with the historical period when methyl mercury (meHg) was first observed in high concentrations in sediment core profiles (Richerson et al. 2000).

A study was conducted by Humboldt State University in 1988–89 to evaluate the contribution of the Sulphur Bank Mercury Mine to Hg contamination in Clear Lake (Chamberlin et al. 1990). They concluded that sheet-wash erosion and slope failures of waste rock and overburden piles, with subsequent deposition of Hg-laden materials into near-shore regions, were the most significant processes introducing Hg into Clear Lake, with estimates of ca. 100 kg of Hg being delivered to Clear Lake annually from these processes. In addition, mining practices (such as bulldozing waste rock and overburden piles into the lake) from 1927–44 and 1955–57 also contributed significantly to the most recent accumulation of Hg within the upper layers of lakebed sediments (Chamberlin et al. 1990) which have totHg concentrations as high as  $450 \text{ mg kg}^{-1}$  (ppm) in the Oaks Arm near the mine (Suchanek et al. 1997, 1998ab).

The 1988–89 evaluation by Chamberlin et al. (1990) of the processes contributing to the input of Hg to Clear Lake was influenced significantly by the fact that California was in the midst of a six year drought through 1992. Our initial monitoring of Hg within the Clear Lake aquatic ecosystem began in the fall of 1992, and seasonal monitoring has been continuous from May 1994 through the present. The winter of 1994–95 produced extreme rainfall at Clear Lake and fluids from the Herman Pit overflowed into Clear Lake. During this overflow, a white flocculent precipitate (floc) was observed in Clear Lake immediately adjacent to the mine site. As a result of a diligent ongoing monitoring program, the discovery of floc in the spring of 1995 dramatically altered our perception of Hg loading to the Clear Lake ecosystem.

As an alternative to the view that Hg contamination in Clear Lake is primarily from the mine, Varekamp and Waibel (1987) suggest that geothermal springs in Clear Lake could represent a natural source of Hg contamination to the lake system in pre-historic (i.e., geologic) times and possibly modern times. However, Sims and White (1981) and Suchanek et al. (1997, 1998a) provide ample evidence that historic (i.e., modern) Hg input to the lake system is the result of mining activities associated with the Sulphur Bank Mercury Mine.

Data presented here address the significance of the floc discovery, its relationship to acid mine drainage (AMD) from the Sulphur Bank mine and how this has

altered our perception of Hg methylation in Clear Lake, and potential remedial actions designed to reduce Hg contamination in Clear Lake.

## 2. Materials and Methods

### 2.1 MONITORING PROGRAM

A survey of totHg and meHg within biotic and abiotic components was performed at 36 Clear Lake stations in the fall of 1992 (Suchanek et al. 1993, 1998a, 2000). In the spring of 1994, a seasonal monitoring program (ca. every 6–8 wks) was initiated to evaluate those same parameters at a reduced number of stations (see Figure 1). Limnological variables (e.g., temperature, dissolved oxygen, conductivity and pH) were also documented at one meter depth intervals at each station [see Suchanek et al. (1997, 1998a) for details] and monthly surveys using an Ekman dredge were used to identify the presence of floc.

### 2.2 CHEMICAL ANALYSES

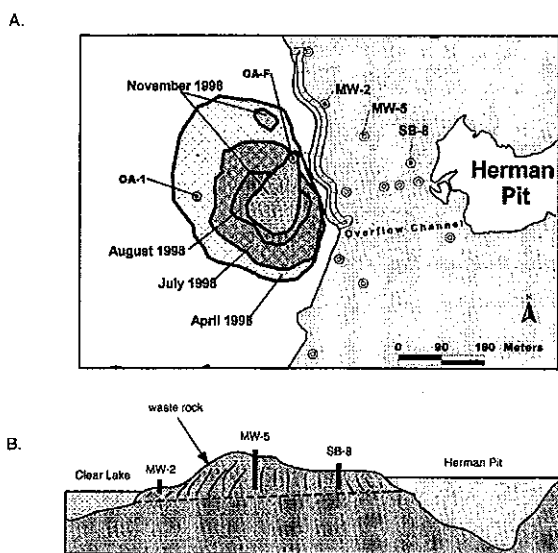
Total Hg (totHg) in surficial sediments (to 5 cm depth) and floc was determined using standard cold vapor atomic absorption methodology. TotHg in water was determined using dual amalgamation/cold vapor atomic fluorescence spectrometry. MeHg in water and sediment was determined using aqueous phase ethylation followed by cryogenic gas chromatography with cold vapor atomic fluorescence detection [see Suchanek et al. (1998a) for details]. This meHg analytical method has the potential to produce some artificially elevated meHg concentrations, especially in samples high in inorganic Hg (Liang et al. 1996, Bloom et al. 1997), and we are presently evaluating the significance of this possible analytical artifact on our results.

For floc, Inductively Coupled Plasma-Mass Spectrometry (ICP-MS) was used to determine the metal composition of the floc, and Scanning Electron Microscopy linked with an Energy Dispersive X-ray Fluorescent Spectrometer was used to determine the basic physical/chemical structure. The analysis of Acid Volatile Sulfides (AVS) was performed by Battelle Marine Science Laboratory following Cutter and Oates (1987). Briefly, this method employed selective generation of hydrogen sulfide, cryogenic trapping, gas chromatographic separation, and photoionization detection to measure AVS in sediment. Simultaneously Extractable Metals (SEM) in lakebed sediments and floc were analyzed by Battelle Marine Science Laboratory. The SEM extract was analyzed for Cd, Cu, Ni, Pb and Zn by Inductively Coupled Plasma Mass Spectrometry (ICP-MS) (EPA 1991) and for Hg by Cold Vapor Atomic Fluorescence (CVAF).

Sulfate in water, collected from 1 m above the sediment/water interface and from centrifuged porewater, was determined following EPA method 160.2.

### 3. Results and Discussion

During our seasonal monitoring program, an easily resuspended white flocculent material (floc) up to 10 cm thick was observed (in April 1995) covering a large area of the sediments in the Oaks Arm near the mine (Figure 1). This event coincided with extensive precipitation and the overflow of the Herman Pit (which has a water level about 4 m above Clear Lake). It was estimated that the floc covered ca.  $1 \times 10^6 \text{ m}^2$ , with traces extending out as far as lake sampling site OA-2 (see Figure 1). As a result of the floc discovery near the mine, we established a new monitoring site (OA-F) to evaluate seasonal fluctuations of totHg and meHg in native lakebed sediments and floc. Subsequent monitoring during 1998 (a high precipitation year) and 1999 (a low precipitation year) indicates significant seasonal variability in the extent of the floc coverage, with a more extensive distribution during times of higher precipitation (indicating greater production and delivery of floc to the nearshore region) and a more restricted distribution during time of lower precipitation (Figure 2A – 1999 data not shown). Acid mine drainage (AMD) likely flows downslope from the Herman Pit into Clear Lake through mining waste rock, which has been deposited over an andesite base (Figure 2B).



**Figure 2A.** Distribution of floc in the nearshore region of the mine during 1998, showing a significant reduction in aerial coverage coincident with a reduction in precipitation from April through November.

**Figure 2B.** Cross-sectional view of mine site including the Herman Pit, three monitoring wells in the waste rock piles and Clear Lake. Dashed line indicates bottom of waste rock pile, which rests on an andesite sill. Note: Figure 2B is not to scale.

Floc had totHg concentrations that were lower than surrounding sediments yet considerably elevated meHg concentrations, especially during the summer months (Figure 3 A&B). ICP-MS analysis of dried floc indicates a relatively high aluminum and iron content with higher levels of silica than found in Herman Pit sediments. A scanning electron microscope image of the floc crystals (Figure 4) provides a view of the background matrix and crystalline structure of the floc. An Energy Dispersive X-ray Fluorescent Spectrometric analysis indicates that the background matrix is mostly iron and aluminum, with abundant tubular-shaped hollow crystals about 40  $\mu\text{m}$  long and about 5  $\mu\text{m}$  in diameter. This structure and chemical composition is typical of kaolinitic type materials, and the floc is considered to be a halloysite precipitate from acid mine drainage (AMD). The chemical composition of these crystals (in an

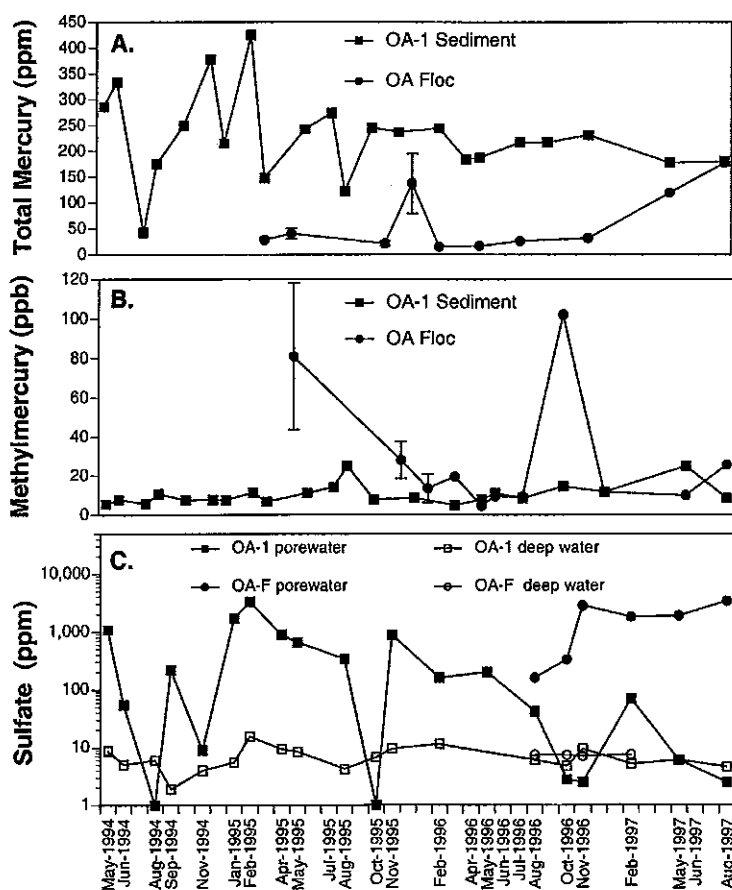
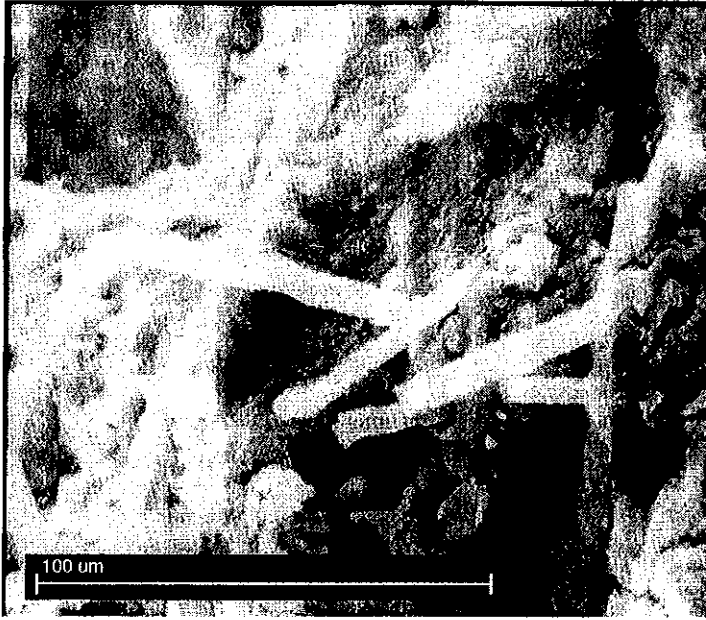


Figure 3. Long-term monitoring data for (A) total mercury [ $\text{mg kg}^{-1}$  (ppm) dry wt.] and (B) methyl mercury [ $\mu\text{g kg}^{-1}$  (ppb) dry wt.] in sediments, and (C) sulfate [ $\text{mg L}^{-1}$  (ppm) dry wt.] in lake water and sediment porewater at sites OA-1 (ambient conditions near the mine) and OA-F (floc site) from 1994–1997.



*Figure 4. Scanning Electron Micrograph of dried, natural floc, showing background matrix and aluminosilicate tubules (approximately 40  $\mu\text{m}$  long).*

approximate ratio of 2 silicon to 1 aluminum), indicates that the majority is present as  $\text{SiO}_2$  and  $\text{Al}_2\text{O}_3$ .

Mixing Clear Lake and Herman Pit waters in a beaker yielded a material ("synthetic floc") identical to that found in the lake in both appearance and chemical composition (Reynolds et al. 1997). The largest quantity of floc formed at ca. pH 8, at a ratio of about 10:1, lake water: Herman Pit water. At dilutions of 20:1 or greater, floc production was greatly reduced.

Recent tracer studies (using Rhodamine-WT and sulfur hexafluoride) show that water originating from the Herman Pit and other sources at the mine move through the waste rock and overburden piles and reach Clear Lake (Oton et al. 1998). Along this route, the chemical composition of Herman Pit fluids is modified greatly: Hg and sulfate are increased dramatically and the low pH is either maintained or sometimes decreased as AMD leaches through waste rock and native ore. Table I shows changes in the chemical composition of AMD fluids typically collected from monitoring wells between the Herman Pit and Clear Lake.

The OA-F site in the vicinity of the mine is also indicative of a site where AMD is reaching Clear Lake, as indicated by low water column pH (as low as 5.5) and high porewater sulfate concentrations (up to ca. 4,500 ppm) (Figure 3C). Sulfate leaching into the sediment adjacent to the mine is reduced by bacteria during the oxidation of organic matter and creates highly reduced sediment conditions in

Table I

Chemical characteristics of mine related fluids in Herman Pit, three monitoring wells within the waste rock piles, and typical values for Clear Lake "deep" (near the sediment/water interface) water column samples. Data given are mean values and ranges (in parentheses). Units for totHg are  $\mu\text{gL}^{-1}$  (ppb), and for sulfate are  $\text{mgL}^{-1}$  (ppm). Data collected 1994–1998. \*from Richerson et al. (1994); \*\*lowest value obtained from a survey at OA-F in 5/96.

	Clear Lake average conditions	Clear Lake OA-F sampling site	waste rock monitoring wells			typical Herman Pit
			MW-2	MW-5	SB-8shallow	
pH	7.96*	5.5**	3.2 (2.9–3.4)	3.1 (3.2–3.5)	2.9 (2.5–3.1)	3.2 (2.5–3.5)
dissolved total Hg	0.0007 (0.0002–0.012)	1.8 (0.5–5.2)	28,500 (12,800–80,100)	66,850 (54,200–84,100)	34,350 (18,000–53,400)	690 (18–1,890)
sulfate	8 (2.5–14.4)	6.7 (0–33)	3,067 (2,600–4,500)	2,867 (2,500–3,300)	2,380 (2,000–2,700)	2,358 (1,000–3,200)

the summer and fall, when water temperatures are high, creating seasonally elevated meHg concentrations (up to ca. 100  $\mu\text{g}/\text{kg}$  = ppb) in the floc (see Figure 3B).

Our Hg monitoring program began in 1992 when the U.S. EPA remediated part of the mine site by reducing the slope of waste rock piles, thereby preventing further erosion and accumulation of inorganic Hg into Oaks Arm sediments. Through microstratigraphy analysis of short sediment cores, it appears that this remediation likely reduced inorganic Hg in surficial sediments close to the mine by as much as 50–150  $\text{mg kg}^{-1}$  (ppm). However, the large input of inorganic Hg from the erosion of waste rock piles was likely not very bioavailable. The significance of the recent floc discovery, as indicative of the production and transport of AMD to Clear Lake, is that this represents an alternative source of Hg loading from the mine site to the lake. Not only is there an additional loading source of Hg, but the form of this Hg in floc is likely to be more bioavailable.

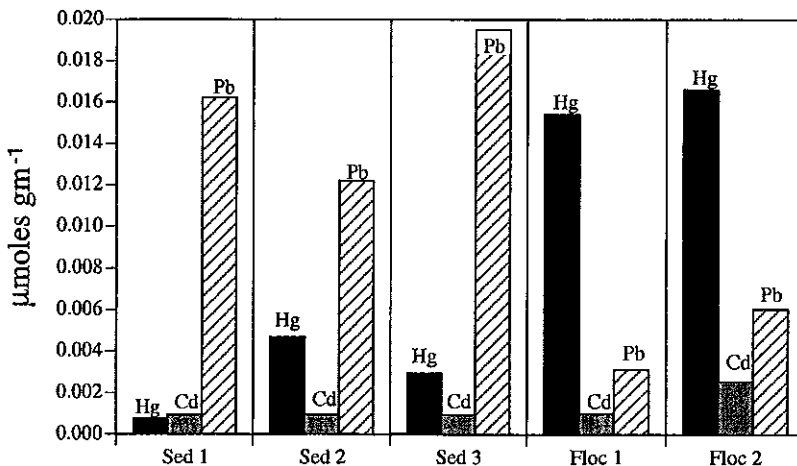
Although our discovery of the floc in 1995 has modified our interpretation of potential Hg (especially meHg) input sources to Clear Lake, it is likely that such floc has been generated from the mine since the early days when open pit mining was initiated on a large scale about 1927. Because fluids continually entered the mining pit during this process, they pumped these fluids (most likely very acidic) out into both Clear Lake and the wetland to the north of the Sulphur Bank Mine. In addition, drainage channels were likely cut through the waste rock and tailings piles that flowed into Clear Lake. Coleman (1930) reported on such a stream draining from the mine into Clear Lake indicating "This little stream ... turns the water milky white for some distance from the mouth." This may also help to ex-



plain why the concentration of not only total Hg (primarily inorganic Hg) as well as meHg, as observed in sediment cores from Clear Lake, increased exponentially at this time (Meillier et al. in press).

Because metal bioavailability can be highly dependent upon sulfur binding (DiToro et al. 1990), we analyzed Acid Volatile Sulfides (AVS) and Simultaneously Extractable Metals (SEM) to evaluate potential Hg bioavailability in typical Clear Lake sediments vs. floc. The ratio of SEM:AVS provides one measure of sulfur binding (and thus lack of bioavailability/toxicity); values  $> 1.0$  suggest weak sulfur binding and increased bioavailability, whereas values  $< 1.0$  suggest lowered bioavailability (DiToro et al. 1990). Typical Clear Lake sediments exhibit a SEM:AVS ratio much lower than 1.0 (0.007–0.11; Suchanek et al. 1997), but the ratio for floc is significantly higher (0.8–2.1; Flanders et al. 1998). Furthermore, SEM for Hg was ca. four times higher in floc than typical Clear Lake sediments, indicating a much higher potential for bioavailable Hg in floc (Figure 5). In addition, the lowered pH and increased sulfate in this region likely stimulate the production of meHg (Mack et al. 1997).

The discovery of this AMD derived floc in the spring of 1995 has dramatically altered our understanding of Hg loading to the Clear Lake aquatic ecosystem. This discovery has now fueled investigations into the Hg methylation process and its implications to Hg bioaccumulation throughout Clear Lake. It is clear that AMD from the mine, as evidenced by the production of this floc, is a mechanism for the introduction of highly bioavailable Hg into Clear Lake and its biota. This discovery was only made possible by the diligent ongoing seasonal monitoring program. Before this discovery, it was assumed that the primary Hg loading to Clear Lake was derived from sheet wash erosion and waste rock slope failures, resulting in



**Figure 5.** Concentrations of Simultaneously Extractable Metals (mercury, cadmium and lead) in three lakebed sediment samples (from station OA-1) and two floc samples (from station OA-F) collected in July 1995 showing that floc contains mercury that is much more easily extractable, which is indicative that this mercury may be more bioavailable.

high Hg concentrations in Oaks Arm sediments. One option to remediate this condition would have been to cap Oaks Arm sediments. With the discovery of floc as an additional significant source of bioavailable Hg to Clear Lake derived from AMD, alternative remedial options must be considered.

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