Ecological Applications, 18(8) Supplement, 2008, pp. A12–A28

THE LEGACY OF MERCURY CYCLING FROM MINING SOURCES IN AN AQUATIC ECOSYSTEM: FROM ORE TO ORGANISM

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Abstract. Clear Lake is the site of an abandoned mercury (Hg) mine (active intermittently from 1873 to 1957), now a U.S. Environmental Protection Agency Superfund Site. Mining activities, including bulldozing waste rock and tailings into the lake, resulted in ~100 Mg of Hg entering the lake's ecosystem. This series of papers represents the culmination of ~15 years of Hg-related studies on this ecosystem, following Hg from the ore body to the highest trophic levels.

A series of physical, chemical, biological, and limnological studies elucidate how ongoing Hg loading to the lake is influenced by acid mine drainage and how wind-driven currents and baroclinic circulation patterns redistribute Hg throughout the lake. Methylmercury (MeHg) production in this system is controlled by both sulfate-reducing bacteria as well as newly identified iron-reducing bacteria. Sediment cores (dated with dichlorodiphenyldichlorethane [DDD], ^{210}Pb, and ^{14}C) to ~250 cm depth (representing up to ~3000 years before present) elucidate a record of total Hg (TotHg) loading to the lake from natural sources and mining and demonstrate how MeHg remains stable at depth within the sediment column for decades to millenia. Core data also identify other stresses that have influenced the Clear Lake Basin especially over the past 150 years.

Although Clear Lake is one of the most Hg-contaminated lakes in the world, biota do not exhibit MeHg concentrations as high as would be predicted based on the gross level of Hg loading. We compare Clear Lake's TotHg and MeHg concentrations with other sites worldwide and suggest several hypotheses to explain why this discrepancy exists. Based on our data, together with state and federal water and sediment quality criteria, we predict potential resulting environmental and human health effects and provide data that can assist remediation efforts.

Key words: benthic invertebrates; birds; Clear Lake, California, USA; coring; fish; mercury; mining; plankton; remediation; sediment; water.

Introduction

This contribution represents a foundation and synthesis paper for the Special Issue of *Ecological Applications* on mercury cycling in the Clear Lake aquatic ecosystem and serves several purposes: (1) it identifies the problem of mercury (Hg) contamination derived specifically from mining sources, its cycling and bioaccumulation in relation to the Clear Lake system; (2) it provides common background information about the Clear Lake aquatic ecosystem and watershed for the other contributions so that similar information need not be repeated; (3) it provides an historic background for Clear Lake watershed environments and processes, including biotic and abiotic components, providing a framework for a more holistic view of Hg cycling in this series of studies; (4) it provides a summary of previous Hg-related studies that have been published on Clear Lake, especially those that influence our understanding of Hg cycling; (5) it describes the regulatory process and how it relates to the history of Hg in Clear Lake and remediation attempts; and (6) it provides a synthesis of this collection of studies on Hg cycling in Clear Lake, from the ore to the organism.

This series of papers represents an integrated and holistic ecosystem approach to understanding the origin,

Manuscript received 20 February 2008; accepted 17 March 2008. Corresponding Editor: J. S. Baron. For reprints of this Special Issue, see footnote 1, p. A1.

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transformation, transport, bioaccumulation, and fate of Hg in a mine-dominated aquatic ecosystem (Clear Lake, California) that can serve as a model for other Hg-contaminated systems that are influenced primarily by mining.

Mercury (Hg) bioaccumulation and associated concerns for human and environmental health currently drive significant research efforts throughout the world (Wiener et al. 2003, Mergler et al. 2007, Scheuhammer et al. 2007, Swain et al. 2007). While most of the Hg-contaminated systems being studied are associated with Hg loading derived, to a large degree, from atmospheric deposition (e.g., Ebinghaus et al. 1999, Hintelmann et al. 2002, Sandilands et al. 2005, Lindberg et al. 2007), few studies have described the magnitude and spatial and temporal trends of Hg contamination and bioaccumulation resulting from local/regional mining sources. The U.S. Environmental Protection Agency (U.S. EPA) *Mercury Report to Congress* (U.S. EPA 1997) did not even identify mining as a significant source of Hg to the environment. Yet, in the western United States, historic Hg, gold, and silver mines continue to play a major role in the release of Hg to the environment, and some progress is being made toward understanding these processes (Domagalski et al. 2004; J. G. Wiener, C. C. Gilmour, and D. P. Krabbenhoft, *unpublished manuscript*). These include studies of the Carson River and Lahontan Reservoir system in Nevada (e.g., Gustin et al. 1994, Bonzongo et al. 1996, Lechler et al. 1997), several California rivers (Rytuba 2000), two Oregon lakes (Ambers and Hygelund 2000), and San Francisco Bay (Thomas et al. 2002). Others (e.g., Lockhart et al. 1995) have utilized known contamination histories from mining and other industrial sites as proxies to analyze natural vs. anthropogenic Hg loading to lake sediments.

California is unique, with nearly 300 abandoned Hg mines and prospects, mostly along the Coast Range (California Division of Mines 1950, Churchill 2000). Over the past 150 years, Hg contamination from Coast Range mines has been, and continues to be, deposited into streams that join either directly or indirectly with the Sacramento River, the San Joaquin River, and the San Francisco Bay-Delta estuary (Domagalski et al. 2004). Elemental Hg (primarily from Coast Range mines) was typically transported to and used in the Sierra Nevada Range (eastern California) to extract gold or silver from parent ore by amalgamation (Alpers et al. 2005). Losses of elemental Hg from mining in the Sierra Nevada Range waterways are estimated at $2.4–4.8 \times 10^6$ kg (Churchill 2000) and continue to move downstream to the San Francisco Bay-Delta estuary. The distribution of Hg, gold, and silver mines in California (see map in Wiener and Suchanek 2008) illustrates the extent of this problem. A significant research effort (funded by the California Bay-Delta Authority) is underway to evaluate the magnitude and significance of Hg contamination in the San Francisco Bay-Delta estuary, including the relationship to upstream loading from abandoned Hg, gold, and silver mines (J. G. Wiener, C. C. Gilmour, and D. P. Krabbenhoft, *unpublished manuscript*).

Clear Lake (39°00′ N, 122°45′ W; Fig. 1) is 402 m above sea level, with surrounding ridge tops to 1500 m. The lake has a surface area of 155 km^2 and a watershed area of 1370 km^2. The lake supports active commercial and sport fisheries and is the site of one of the most significant abandoned Hg mines in the United States (Suchanek et al. 2003). (Currently, there are no active Hg mines in the United States; the last mine closed in 1990 [Jasinski 1995].) Deep cores collected in the Upper Arm of Clear Lake in 1980 by the U.S. Geological Survey (USGS) showed that Clear Lake is at least 450 000 years old and possibly as old as ~2 million years, making it one of the oldest lakes in North America (Sims et al. 1988). The lake is shallow (average depth ~6.5 m), alkaline (~pH 8), polymictic (typically well mixed), and eutrophic. It is located within an active volcanic region, and abundant geothermal springs release both fluids and gases (primarily CO_2, H_2S, and CH_4) from the lake bottom. Because of its high productivity, it supports large populations of fish, waterfowl, and avian and mammalian fish predators (Horne 1975, Richerson et al. 1994, Moyle 2002, Suchanek et al. 2003). The Clear Lake area is considered one of 158 "Important Bird Areas" in California, based on a high number of sensitive species (14 listed species by the U.S. Fish and Wildlife Service) and often extraordinarily high wintering and breeding populations of migratory and resident waterbirds (Cooper 2004).

Clear Lake has characteristics that make it a useful ecosystem for scientific investigation. It is enclosed within a well-defined watershed, with inputs and outputs that can be reasonably well quantified. In addition, the distribution of Hg from a point source, the Sulphur Bank Mercury Mine, creates a steep concentration gradient of Hg that can be used to monitor the effects of Hg on natural populations and can be used to test hypotheses involving the influence of varying Hg concentrations on biotic systems.

A long-term (50-year) record of population fluctuations in the benthic invertebrates *Chaoborus* and Chironomidae from 1954 to 2001 as well as planktonic invertebrates from 1988 to 2002 is provided in Suchanek et al. (2008*c*). Long-term trends in fish population abundance are provided in Eagles-Smith et al. (2008*b*) and population fluctuations in two dominant bird species (grebes and Ospreys) since 1992 are documented in Anderson et al. (2008).

Suchanek et al. (2003) identified the major geological, climatological, ecological, political, and economic factors that influence the outcome of management and policy-level decisions on the health and well-being of the Clear Lake ecosystem, especially the lake proper.

The Historical Landscape

Clear Lake and its associated watershed have been subjected to multiple stresses over the past 150 years

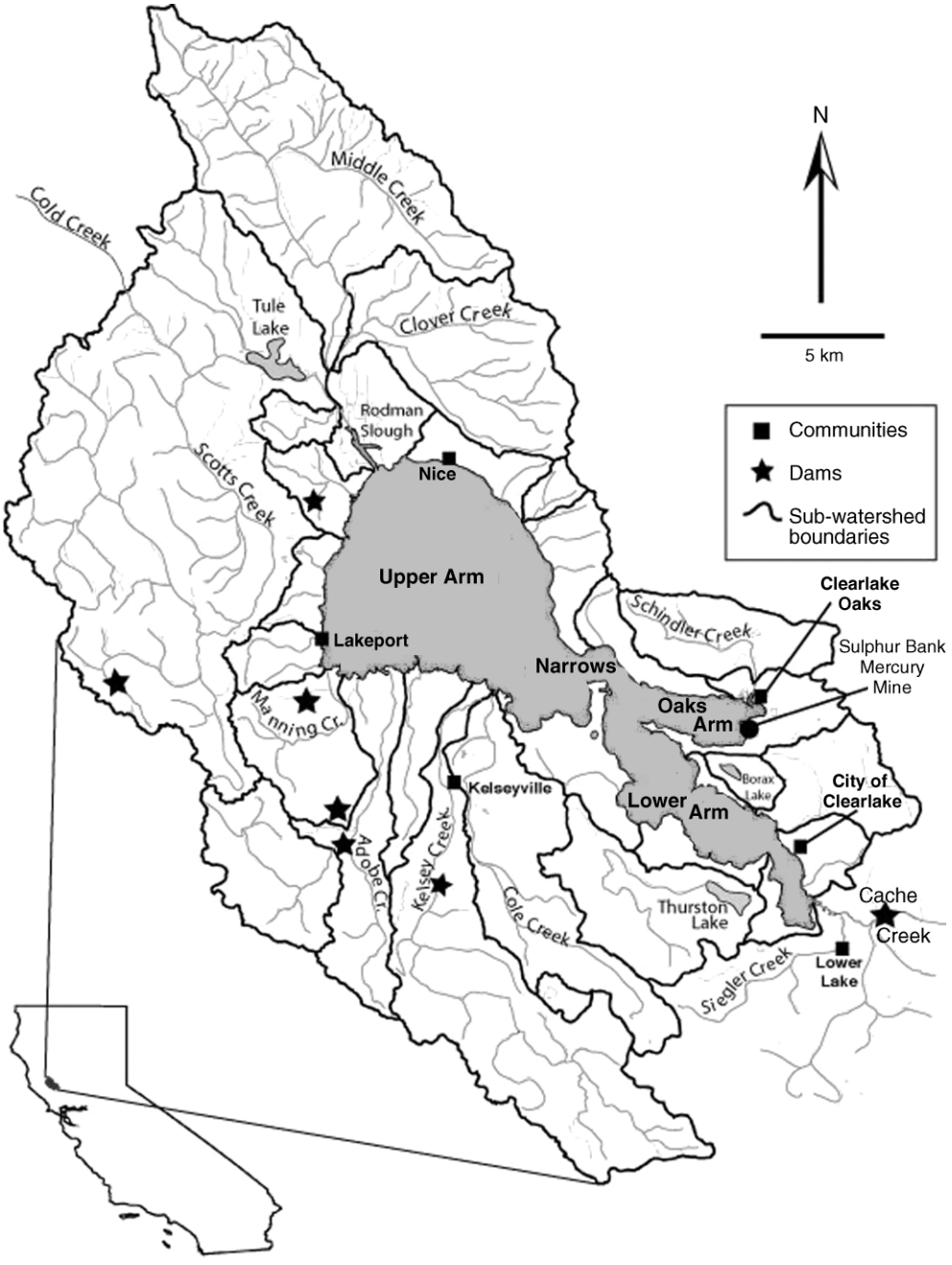

FIG. 1. Clear Lake, California, USA, and surrounding watershed, with subwatershed boundaries.

(Suchanek et al. 2003, Richerson et al. 2008). While some stresses are natural, the vast majority are anthropogenic, including: (1) modifications of the landscape that have increased fire frequency, (2) logging, deforestation, and stream bed gravel mining that promoted higher erosion rates and nutrient loading, (3) levee construction to convert wetlands to agricultural production (also resulting in enhanced nutrient loading), (4) shoreline modifications such as marinas, gunnite, or riprap zones, (5) lake level manipulations for crop irrigation, (6) intentional (to enhance fisheries) and unintentional species introductions, (7) intentional addition of aquatic and terrestrial pesticides for the control of insect pests and aquatic weeds, and (8) mining for numerous minerals and products (Suchanek et al. 2003). Approximately 90% of the county's 64 000 residents live in several towns and small cities immediately adjacent to the lake (data current as of May 2006). Clear Lake is a dominant feature in Lake County, which depends for much of its economy, either directly or indirectly, on lake-based tourism and recreation. Clear Lake is famous as one of the prime bass tournament fishing destinations in

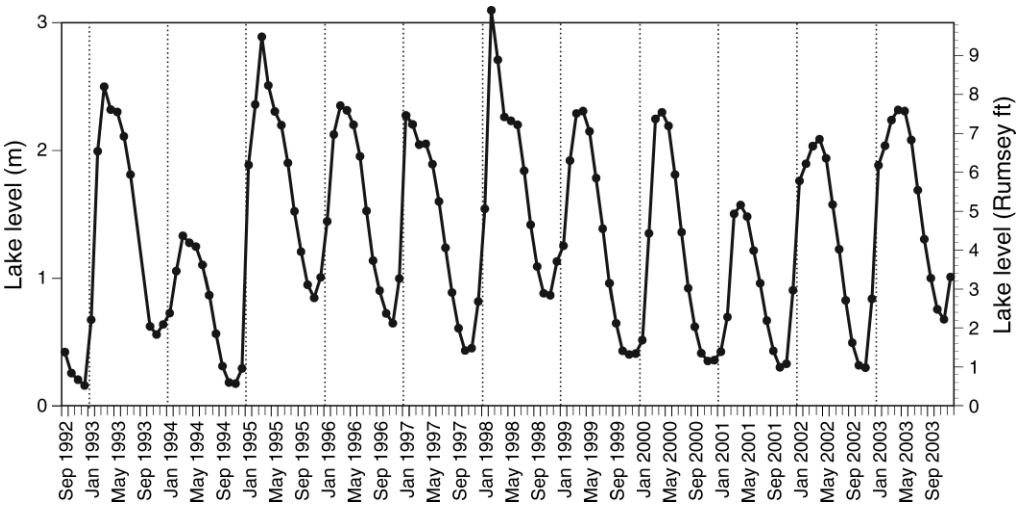

FIG. 2. Monthly mean water levels (see the local Rumsey scale in feet; 0 feet Rumsey = 402 m elevation) for Clear Lake during this study (1992–2004).

California. Based on studies related to massive additions of dichlorodiphenyldichloroethane (DDD) to the lake in the late 1940s and early 1950s to control the Clear Lake gnat (a non-biting mosquito-like midge, *Chaoborus astictopus*), with devastating effects on breeding Western/Clark's Grebe (*Aechmophorus* sp.) populations, Clear Lake became the first site where the process of pesticide bioaccumulation in food webs was identified and subsequently popularized in Rachel Carson's book *Silent Spring* (Carson 1962, Rudd 1964; see Suchanek et al. 2003 for details). Actual and perceived Hg contamination in this system has also impacted the socioeconomic vitality of the region, with a decade-old estimate of $US 7–10 million annual loss in revenue (Goldstein and Tolsdorf 1994). Additionally, at least four Native American tribal reservations that surround Clear Lake no longer utilize historic natural resources (especially fishes) because of Hg contamination.

Because it is used to store irrigation water, Clear Lake experiences significant water level fluctuations (up to 4 m/yr) that can affect aquatic ecosystem processes and may significantly influence the transport of Hg within the lake and downstream into Cache Creek (the outlet for Clear Lake), a tributary of the Sacramento River in the Central Valley of California. Mean monthly lake level fluctuations (1992–2003), mean monthly historic rainfall (1955–2004), and annual cumulative rainfall (1886–2004) are provided in Figs. 2, 3, and 4, respectively. Annual minimum and maximum lake levels from 1850 to 2000 are given in Suchanek et al. (2003) and Richerson et al. (2008).

Although it is likely that Holocene Clear Lake has always been eutrophic, observations by lakeside resi-

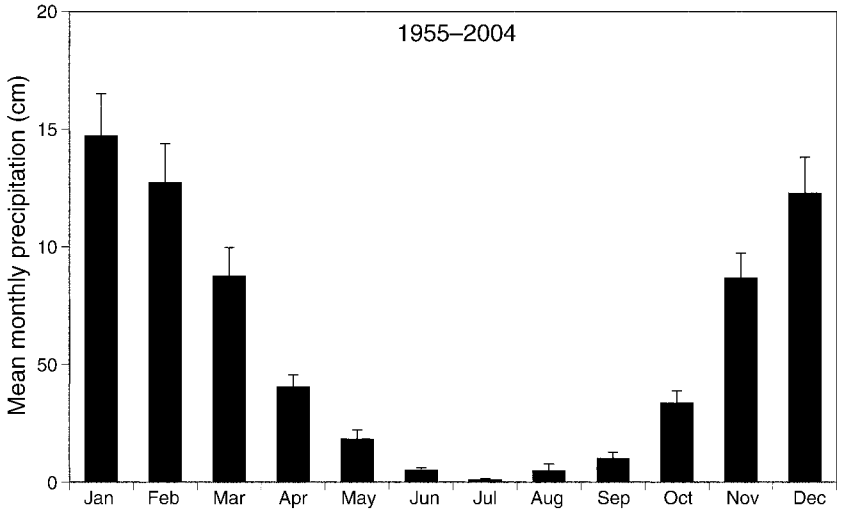

FIG. 3. Historic monthly precipitation (mean + SE; calendar year data) for Clear Lake Basin from 1955 to 2004.

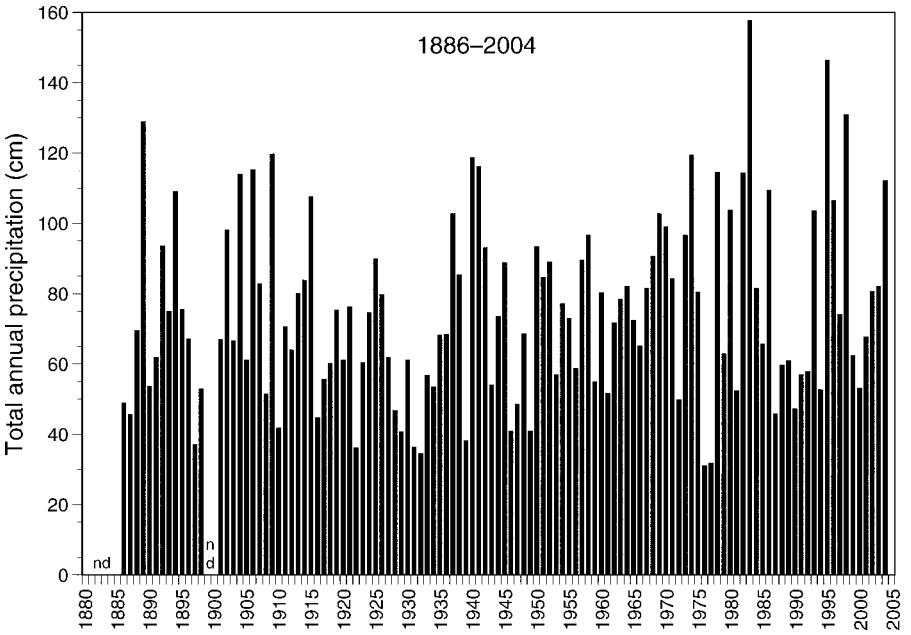

FIG. 4. Historic total annual precipitation (calendar year data; "nd" indicates no data) for Clear Lake Basin from 1886 to 2004. Data are from the University of California Statewide Integrated Pest Management Project, Weather Database and the Lake County Air Quality Management District, Lakeport Station.

dents suggest that sediment and nutrient loading from the Clear Lake watershed probably increased significantly in the early part of the 20th century (Richerson et al. 1994). Our coring work confirms these observations (Osleger et al. 2008). This eutrophication occurred primarily from elevated sediment transport into the lake as a result of the elimination of up to 85% of the wetlands for conversion to agricultural production (Richerson et al. 1994, Suchanek et al. 2003). This led to large blooms of scum-forming cyanobacteria primarily during summer months, causing extremely low water clarity (e.g., Secchi depth <2 m). Although the reasons are as yet unclear, cyanobacterial blooms have declined significantly since ca. 1992, increasing the water clarity considerably and leading to heavy growth of rooted aquatic vegetation in shallow water. During years when cyanobacterial blooms are present, they contribute significant organic detritus to the lake when they decompose, typically at the end of summer. This, in turn, can stimulate sulfate-reducing bacteria that can methylate Hg. Secchi disk depths from 1969 to 2004 (Fig. 5) illustrate the dramatic changes in water clarity experienced since ca. 1991/1992.

Winds play an important role in the movement of contaminants within Clear Lake (Pálmarsson and Schladow 2008, Rueda et al. 2008). Although episodic storms may produce winds from any direction, wind in the Clear Lake basin blows predominantly from the west-northwest toward the Sacramento Valley (Fig. 6). Due to the extreme topographical variation adjacent to the lake (Mount Konocti, along the south shore of the lake, rises 900 m above lake level and the ridges defining the watershed are generally 400 m or more above lake level), there is considerable spatial variability in the wind field that exerts an influence on lake circulation patterns (Rueda et al. 2005). These circulations have the potential to remobilize and transport sediment and associated contaminants to both the Upper Arm and the Lower Arm (Rueda and Schladow 2003, Rueda et al. 2003), which is evidenced by the present distribution of Hg in lake bed sediments (Suchanek et al. 2008*b*).

Historic Land Use Changes

Dramatic land use changes took place soon after European contact, although there were already about 3000 Native American inhabitants of the Clear Lake basin in the 1850s (Simoons 1952). Abundant volcanic activity provided heat to drive hydrothermal systems that created rich mineral deposits in the region, and exploitation of these minerals (including Hg) began almost immediately after European contact.

Through the use of sediment cores from Clear Lake, with ages constrained by ^{14}C, ^{210}Pb, and DDD dating, Richerson et al. (2008) and Osleger et al. (2008) analyzed the textural, chemical, and magnetic properties of sediments to ~250 cm depth (equivalent to ~3000 years before present [ybp]) to assess the depositional response to a series of land use changes that occurred in the watershed since European settlement. Interestingly, evidence from the sediment cores suggests that the first century of settlement by Europeans left scant impacts on the watershed and lake. However, clear and abrupt shifts in grain size and magnetic concentration/mineralogy in all three arms of Clear Lake occur concurrently with a

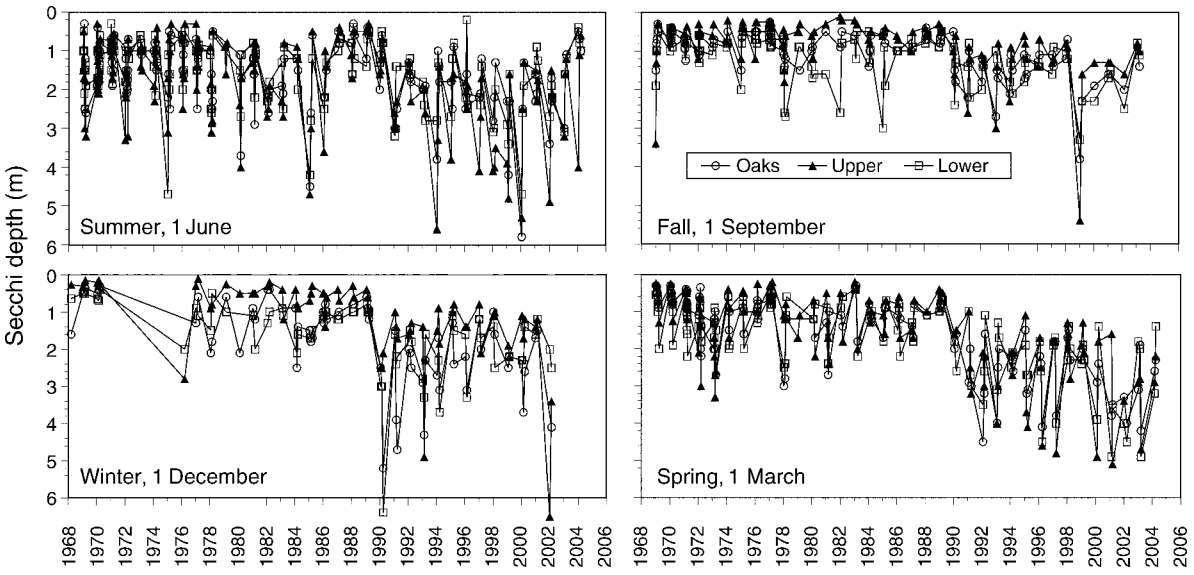

FIG. 5. Secchi disk depths in Clear Lake during summer (June, July, August), fall (September, October, November), winter (December, January, February), and spring (March, April, May) from 1969 to 2004. Data are from the California Department of Water Resources, Sacramento, California, USA.

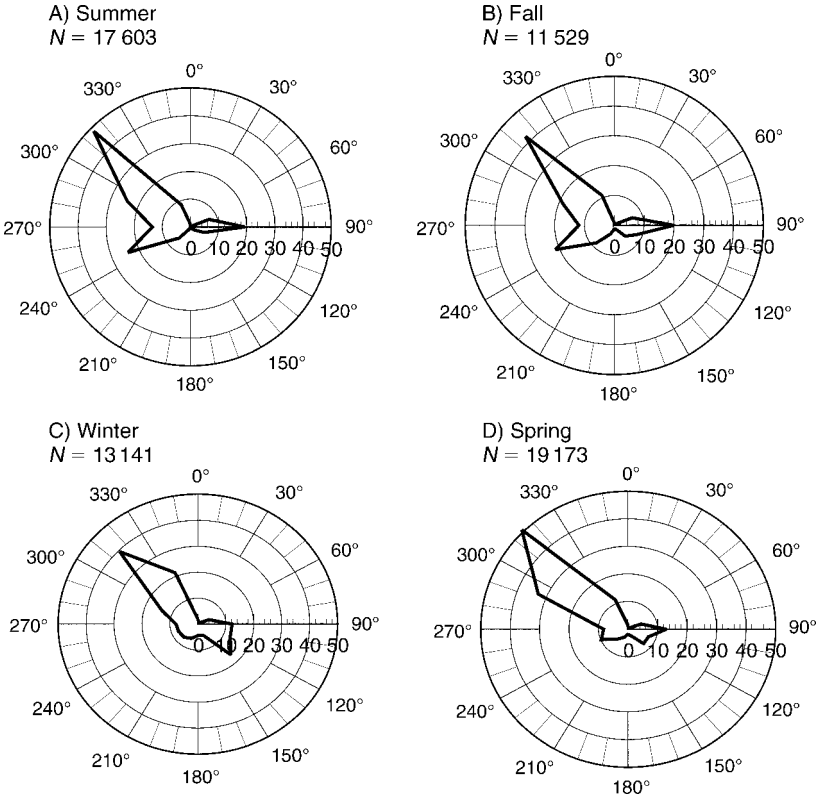

FIG. 6. Wind rose data representing wind direction and velocity (km/h) from a weather station in Lakeport, averaged over four seasons: (A) summer (June, July, August), (B) fall (September, October, November), (C) winter (December, January, February), and (D) spring (May, June, July). Data are from the Air Quality Management District weather station in Lakeport, California, USA.

variety of ecological and chemical changes in sediments beginning around 1927. The use of heavy earthmoving equipment increased sedimentation rates and facilitated open-pit operations at the Sulphur Bank Mercury Mine that resulted in the contamination of the lake with Hg. Furthermore, the eutrophication caused by an enhanced supply of phosphorus from increased sediment loading caused a complex array of additional changes (Richerson et al. 1994, 2008, Suchanek et al. 2003).

Sulphur Bank Mine

One of the most influential events in Clear Lake's history was the development of a sulfur- and Hg-mining operation located on the eastern shoreline of the Oaks Arm (Chamberlin et al. 1990). The Sulphur Bank ore deposit was first discovered in 1857 by Dr. John Veatch of the California Borax Company, which was mining borax in the region (Veatch 1883). In 1865, California's first sulfur mine (the Sulphur Bank Mine) was established to exploit a surface deposit of elemental sulfur (California Division of Mines 1950). Deeper deposits of sulfur from this site were contaminated with cinnabar (a form of mercury sulfide [HgS]), and in 1873 the site was converted to Hg mining and renamed the Sulphur Bank Mercury Mine. The ore body at the Sulphur Bank Mine was described by White and Roberson (1962:397–398) as "... probably the outstanding example in the world of a major ore deposit that is clearly related to hot springs." At that time California accounted for 89% of the nation's Hg production and the Sulphur Bank Mercury Mine yielded ~10% of California's total Hg production (Simoons 1952). Cumulative production from the mine was documented at 129 418 flasks of Hg (a flask = 33.9–34.6 kg, depending on the mining era). The total original content of the deposit was estimated at ~233 000 flasks, or ~7000 Mg (White and Roberson 1962).

Mercury was mined at first using surface scraping, and then shafts and underground mining were used until about 1900. The advent of mechanized earthmoving equipment such as dump trucks, bulldozers, and steam shovels enabled a large open-pit mining operation to begin in 1927, resulting in an 8-ha, 30 m deep excavation (the Herman Pit; see Suchanek et al. 2008*b*). Mining continued intermittently until 1957, yielding ~5000 Mg of commercial-grade elemental Hg (Suchanek et al. 2008*e*), which was used primarily for gold and silver mining in the Sierra Nevada (Alpers et al. 2005). More than 1 250 000 Mg of material were removed, processed, and disposed of during nearly a century of intermittent mining activity. Retort operations were conducted on site during several different time periods, yielding some volatile Hg emissions from the site that likely caused additional localized contamination. As of 2007, Herman Pit is filled with fluids of pH ~ 3 and is separated from Clear Lake by a waste rock berm ~200 m wide (see the cover photo of this Special Issue and Suchanek et al. [2008*b*]). In 1988, Herman Pit was reported to contain Hg concentrations in water and sediment of 800 ng/L and 26 mg/kg, respectively (Columbia Geoscience 1988). Based on furnace losses and residual Hg in tailings and waste rock, it is estimated that ~4400–7000 Mg of elemental Hg were extracted from the site (see Suchanek et al. [2008*e*] for mining history).

Mining operations included deliberate dumping or bulldozing of waste rock and tailings directly into the nearshore regions of the lake, especially when significant amounts of ore were extracted by open-pit mining beginning in 1927. In later years, waste rock removed from the Herman Pit typically was bulldozed toward the lakeshore and allowed to slump into the lake at the angle of repose. As a result of these processes, an estimated 100 Mg of Hg were deposited into the Clear Lake aquatic ecosystem (Chamberlin et al. 1990). Elevated inorganic Hg concentrations have been documented in surface (>400 mg/kg) and near-surface (nearly 1200 mg/kg) sediments and decline exponentially with distance from the mine (Suchanek et al. 1998, 2000*a*, 2008*b*).

A USGS sediment core (to ~28 m depth) collected from the approximate center of the Oaks Arm in 1973 was analyzed for Hg (Sims and White 1981). The Hg profile from this core, reconstructed in Suchanek et al. (2008*e*), exhibits a number of episodes of natural elevated Hg at the following estimated sediment ages (with approximate Hg concentrations in parentheses): 3600 yr (35 mg/kg), 7400 yr (65 mg/kg), 9500 yr (10 mg/kg), and 18 000 yr (15 mg/kg). However, the profile also indicates that the Hg concentration increases exponentially (approaching 80 mg/kg) within the top 1 m of the core, although the very top of their core was not recovered, comparable to the findings reported by Richerson et al. (2008). Shorter historic cores (as deep as 100 cm), collected throughout the lake in 1986 by the Central Valley Regional Water Quality Control Board (CVRWQCB 1987), show Hg concentrations at nearly 1200 mg/kg at lake bed sediment sites in front of the mine at 40 cm sediment depth, but no dating was performed on these cores (see Chamberlin et al. 1990, Suchanek et al. 2008*e*). The most recent Clear Lake cores, collected in 1996 and 2000, exhibited an approximate 10-fold increase in Hg associated with the peak of the open-pit mining era at an estimated date of 1933 using ^{210}Pb dating (Richerson et al. 2008).

Mercury toxicosis (Minamata disease) was first discovered in the 1950s in Minamata Bay, Japan, and resulted from the release of MeHg from a chemical plant with subsequent bioaccumulation into higher trophic level fish and consumption by humans, especially the fishermen from the bay (Tsubaki and Irukayama 1977, Hamada and Osame 1996). With the publicity and knowledge that human consumption of Hg-contaminated seafood was the cause of Minamata disease and because of concern over potentially elevated Hg concentrations in fishes from Clear Lake first documented by Curtis (1977), the California Department of Health Services issued the first fish consumption advisories for Clear Lake (Stratton et al. 1987), with a revised set of advisories in January 2005

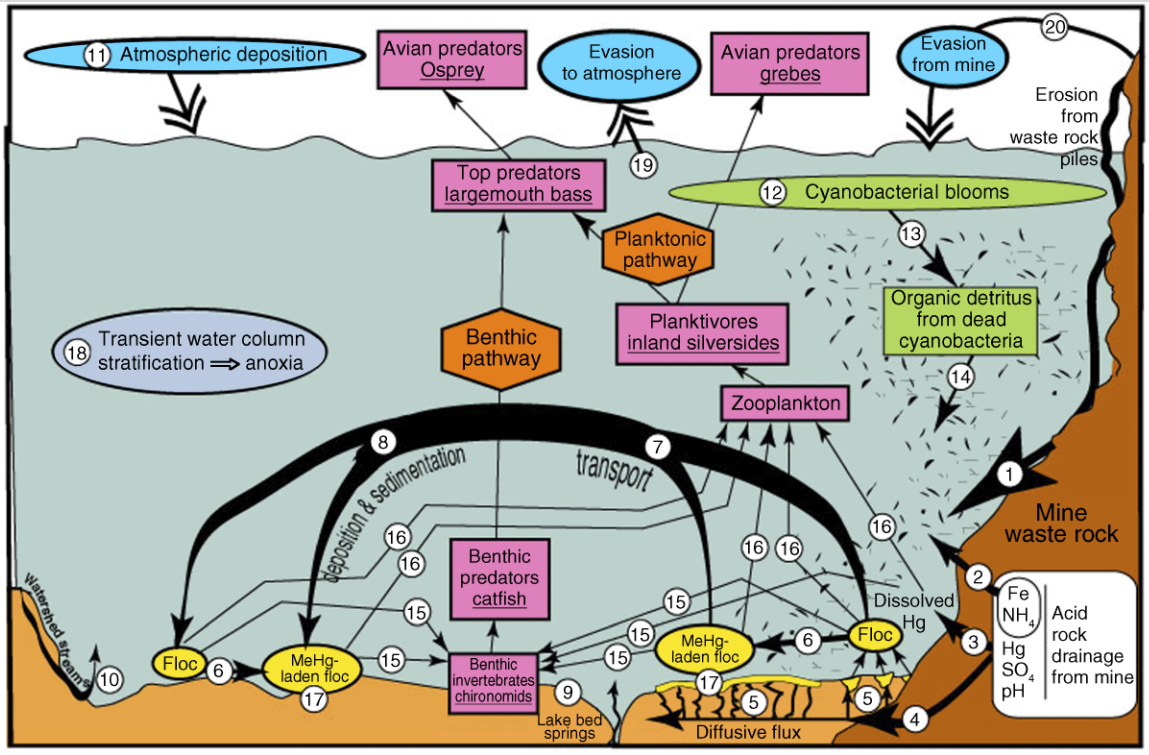

FIG. 7. Conceptual model of pathways and processes involved in mercury cycling in Clear Lake. Key to numbered circles: 1, Surface erosion of acid rock drainage (ARD), producing particulate and dissolved Hg from waste rock and tailings; 2, subsurface ARD flow into the water column yielding particulate Hg, Fe, and NH_4 to the near-shore environment; 3, subsurface ARD flow into the water column yielding dissolved Hg, SO_4, and acidic fluids to the near-shore environment; 4, sub-sediment lateral diffusive flow that moves ARD far out into the Oaks Arm of Clear Lake; 5, upward advection of ARD to the sediment–water interface and production of floc; 6, transformation of inorganic Hg to MeHg; 7, transport of inorganic Hg and MeHg on floc particles to other regions of Clear Lake; 8, deposition of products from pathway 7 onto sediments in other regions of Clear Lake; 9, input of fluids from lake bed springs; 10, input of Hg from watershed streams; 11, input of Hg from wet and dry atmospheric deposition onto the lake surface; 12, seasonal production of cyanobacterial blooms; 13, degradation of cyanobacterial blooms to produce organic detritus source; 14, utilization of cyanobacterial organic detritus by SO_4^- and Fe^- bacteria that methylate Hg; 15, bioaccumulation of Hg via benthic pathway; 16, bioaccumulation of Hg via planktonic pathway; 17, long-term burial of Hg in sediments; 18, transient water column stratification drives anoxic conditions stimulating methylation; 19, evasion of Hg from the lake surface to the atmosphere; 20, evasion of Hg from mine site and redeposition onto the lake surface.

(OEHHA 2005). Currently, 27 other water bodies in California have fish consumption advisories because of mining-associated Hg contamination (OEHHA 2006). In accordance with the Clean Water Act, Clear Lake was placed on California's 303(d) list of impaired water bodies in 1988 because of elevated Hg concentrations in fish tissues. Subsequently, the Sulphur Bank Mercury Mine was placed on the U.S. EPA's National Priority List and was designated as a Superfund Site in August 1990.

In the summer of 1992, under an emergency removal action to reduce erosion of Hg-laden waste rock into the lake, the U.S. EPA cut back the angle of the waste rock pile and added riprap along ~400 m of shoreline to protect the remaining mine site soils. Although this may have significantly reduced the erosion rate of Hg-laden soils, it does not appear to have slowed the flow of acid mine drainage from the mine to the lake.

In 1994 the U.S. EPA issued a Remedial Investigation/Feasibility Study (RI/FS) Report that addressed the state of the knowledge of Hg in surface water and waste rock associated with the mine and the lake (U.S. EPA 1994). However, subsequent studies by Suchanek et al. (1997) revealed additional sources of Hg input to Clear Lake via groundwater and surface water resulting in the reopening of the Remedial Investigation of the mine to address these other loading sources. As of this writing (in 2007) the U.S. EPA is in the process of revising a draft RI/FS Report (U.S. EPA 1994) for only the terrestrial portion of the mine site (termed Operable Unit 1).

MERCURY CYCLING IN CLEAR LAKE

The title of this paper indicates our attempt to trace Hg through the various geochemical and biological pathways that result in elevated Hg concentrations in high trophic level fishes and birds in the Clear Lake ecosystem. The pathways and processes important to the origin, transport, transformation, bioaccumulation, and export of Hg in the Clear Lake aquatic ecosystem are represented in Fig. 7, which also represents a guide and summary of the studies represented in this Special Issue.

Mercury ore body

Mercury is a trace element in the Earth's crust with a mean concentration of 0.08 mg/kg (Krauskopf 1967). However, background soils in Lake County are reported to have 0.10–0.22 mg/kg Hg (Bradford et al. 1996). Most Hg deposits require grades in the range of 0.1% (1000 mg/kg) to be economically viable. This requires a 10^4-fold enrichment over typical worldwide natural abundance levels. Economically viable Hg deposits are thus quite rare. However, the central California Coast Range contains hundreds of areas mineralized with Hg, including the world's fifth and sixth largest historic Hg producers, the New Almadén Mine in Santa Clara County and the New Idria Mine in San Benito County (Pickthorn 1993). The Sulphur Bank Mercury Mine was the fifth largest producer of Hg in California. Before discussing the origin of this deposit, it is instructive to consider the geochemistry and non-anthropogenic cycling of Hg in this system.

The mineralogy of the ore is relatively simple. Mercury ore was deposited at and below the groundwater table in the form of cinnabar (HgS, hex) accompanied by marcasite and pyrite (different crystalline forms of FeS_2). Pyrite and cinnabar are the dominant sulfide minerals deeper in the deposits, and metacinnabar (HgS, cub) and stibnite (Sb_2S_3) were reported as minor phases. Concentrations of sulfide in an excess of dissolved Hg resulted in near-quantitative precipitation of Hg below the groundwater table. This implies that the pre-mining hot springs and fumaroles at the site likely contributed little Hg to the environment, even though oxidation of the excess sulfide resulted in oxidizing acidic fluids capable of transporting high concentrations of dissolved Hg. Open-pit mining, however, exposed the deposits of marcasite, pyrite, and cinnabar to oxidation. Acid mine drainage began to deliver highly soluble and reactive Hg^{2+} to the lake, initiating the ore-to-organism transfer of Hg into the Clear Lake aquatic ecosystem.

Pathways of mercury transport into Clear Lake

The most critical outcomes of this study are the input, transport, output, and storage of Hg in this system, as well as the subsequent bioaccumulation and impacts on biota. This section will provide an overview of the pathways of Hg movement into and out of Clear Lake and identify storage components. Based on all identified sources, Suchanek et al. (*in press*) developed a mass balance Hg budget that identified the known inputs, outputs, and storage of TotHg in the Clear Lake system. A measurable load of Hg derives from stream flow into the lake from the 1370 km^2 watershed (Fig. 7, pathway 10). Included in this component is all wet and dry atmospheric deposition that falls on the landscape that is drained by these streams. Mercury loading from the Clear Lake watershed is estimated at 0.9–48.4 kg/yr (Suchanek et al., *in press*). Both wet and dry atmospheric deposition directly onto the lake surface have diverse sources (Fig. 7, pathways 11 and 20), but no formal studies at Clear Lake have quantified these to date. However, based on known atmospheric deposition at other sites in the region, an estimate of total atmospheric Hg contribution to the Clear Lake surface was calculated at 0.99–2.76 kg/yr.

Locally, Hg-rich soils and waste rock at the Sulphur Bank Mercury Mine volatilize a significant amount of Hg from the open-pit area and the waste rock area (Fig. 7, pathway 20). Mercury efflux data from Gustin et al. (2003) and Nacht et al. (2004), together with knowledge of local wind field conditions, yield estimates of ~0.55–3.40 kg of Hg/yr being redeposited from the mine surface back into the lake.

Some (Verekamp and Waibel 1987) have suggested that the origin of much of Clear Lake's Hg contamination is from natural lake bed springs (Fig. 7, pathway 9). While significant gases and fluids do emanate from the lake bottom, no elevated source of Hg has been found to be associated with lake bed springs (Suchanek et al. 1993, 1997, 2008*b*; Suchanek et al., *in press*).

The mine has varied as a source of Hg to Clear Lake, depending on the degree of mining activity and the types of mining practices. In the 1800s, Hg was mined primarily using shafts, which produced a relatively small amount of ore and rock that was moved to the surface and heated in retort furnaces; the remaining tailings and calcines (retort waste solids) were discarded on site. Little Hg moved from the mine to the lake until the 1920s, when earthmoving equipment changed the nature of mining at Sulphur Bank. Open-pit mining practices begun ca. 1927 produced a large increase in Hg in lake bed sediments (Richerson et al. 2008). Now, ~50 years after the cessation of mining (1957), Hg still flows from the mine to the lake through several pathways (Fig. 7, pathways 1–4). Surface and near-surface runoff from precipitation, which becomes highly acidic upon contact with mine soils, can leach Hg from tailings, calcines, and waste rock, entering the near-shore region as particulate and/or dissolved Hg. Precipitation, seeping down through tailings, calcines, or the 20 m high waste rock pile, creates acid mine drainage that flows through subsurface channels, fracture zones, or into the groundwater table, eventually surfacing in Clear Lake some distance offshore (Shipp and Zierenberg 2008). However, these subsurface flows are very difficult to measure directly.

Herman Pit (pH ~ 3) is another source of acidic fluids that percolate through and leach Hg from the waste rock pile. An early estimate by White and Roberson (1962) estimated 0.0032–0.0060 m^3/s (reported as ~50–95 gallons per minute [gpm] in the original) flowed from Herman Pit to Clear Lake. To evaluate a portion of this pathway, three independent tracer experiments were conducted in Herman Pit during April 1997 (using rhodamine WT dye), June 1998 (using sulfur hexafluoride [SF_6]), and September 1998 (using a combination of SF_6 and ^{22}neon gas), to test the potential flow rate of

acidic fluids out of Herman Pit and thus potentially into Clear Lake (Oton 2000, Schladow and Clark 2008). Tracer concentrations were followed in Herman Pit, mine site monitoring wells, and in Clear Lake proper. The flow rate estimates out of Herman Pit were 0.4417–0.5679 m^3/s (7000–9000 gpm), 0.6373 m^3/s (10 100 gpm), and 0.6310–1.096 m^3/s (10 000–16 000 gpm), respectively, for the three experiments. Flow rate measurements into Clear Lake itself were not taken. While these estimates only project the amount of fluids leaving Herman Pit, they also represent comparable estimates of the potential volume that could be reaching Clear Lake.

Interestingly, based on a modeling approach assuming a homogeneous subsurface environment and using monitoring well data (collected by Tetra Tech EMI, Rancho Cordova, California, USA), from February and April 2000, the U.S. EPA estimated the flow rate of fluids passing from the entire mine site to Clear Lake at 0.0014 m^3/s (22.2 gpm) during their defined "dry season" (February 2000) and 0.0010 m^3/s (16.3 gpm) during their defined "wet season"(April 2000, Jewett et al. 2000*a*). This translates into Hg-loading values of 1.47 kg/yr and 2.41 kg/yr during their "dry season" and "wet season," respectively (Jewett et al. 2000*b*; Tetra Tech EMI). However, in terms of the effects of "seasonality" on flow rates and groundwater levels, in general February and April are both winter wet-season months (see seasonal and historic precipitation data in Figs. 3 and 4).

Total mercury concentrations in unfiltered mine site pit waters (~300–1700 ng/L) and Clear Lake waters (~5–500 ng/L) are higher than most known contaminated sites worldwide (Suchanek et al. 2008*b*). This is surprising because most of the Hg that enters the Herman Pit and Basalt Pit is precipitated out by sulfides due to the active flow of hydrothermal fluids and gases containing H_2S into these pits. However, as acidic water percolates through subterranean pathways down-gradient toward the lake, other Hg is leached out of the surrounding waste rock. Mercury concentrations have been documented in mine site monitoring wells (that are in a direct line between Herman Pit and Clear Lake) at 10 000–340 000 ng/L (~300 times higher than the highest known other site, Carson River/Lahontan Reservoir, Nevada) before flowing into Clear Lake's nearshore zone (see Suchanek et al. 2008*b*).

From the Herman Pit and mine site, Hg-laden acid mine drainage can flow directly to the nearshore region or into the Oaks Arm along identified under-lake, subsediment conduits (Fig. 7, pathway 4; Shipp and Zierenberg 2008). Fluids having chemical and isotopic compositions of Herman Pit fluid can be detected at least 80 m away from the mine, but the spatial extent of this contaminated groundwater plume is as yet undetermined.

When acidic fluids (as low as pH ~ 3.0), high in dissolved Hg, mix with lake water pH ~ 8.0 (Fig. 7, pathways 1, 3, and 5), a white flocculent precipitate (floc) composed of alumino-silicate (halloysite) clay forms that is high in TotHg, is close to neutral density in water, and can be easily mobilized and transported by wind-driven currents to other regions in Clear Lake (Suchanek et al. 2000*a*, *b*; Fig. 7, pathways 7 and 8).

In-situ mercury methylation

During calm periods, floc accumulates in large masses at the sediment–water interface in front of the mine and becomes a site for elevated sulfate reduction and MeHg production (Fig. 7, pathway 6; Mack 1998, Suchanek et al. 2000*a*, *b*). This occurs in an environment that is influenced heavily by the low-pH hot spots identified above, providing an environment conducive to elevated Hg methylation. In addition, because of the configuration of the lake's bathymetry and direction of wind-driven currents (Rueda et al. 2008) large masses of blue-green "algae" (cyanobacteria; Fig. 7, pathway 12) die, sink, and accumulate at the eastern end of the Oaks Arm near the mine, especially in late summer/early fall (Richerson et al. 1994; Fig. 7, pathways 13 and 14). This rich organic loading provides energy resources for bacterial sulfate reduction. The presence of floc, a low-pH environment, organic loading from cyanobacterial blooms, and elevated summer temperatures act in concert to yield elevated MeHg production on particulate organic matter and remobilized sediments near the mine.

Mine-derived floc appears to be an important driver of Hg methylation in this system, although the precise mechanism for this stimulation has not been fully elucidated. A series of laboratory core tube experiments demonstrated that the presence of mine-derived floc in or on sediments increased the production of MeHg by 5–20 times above treatments without floc (Mack 1998; T. H. Suchanek, *unpublished data*; Fig. 7, pathway 6). In Clear Lake, sulfate reduction and Hg methylation are greatest in the top 0–2 cm of sediment and peak in the late summer/early fall (Mack 1998). Annual rates of sulfate reduction summed over the surface area of the lake have been estimated at: Oaks Arm (area $= 2.0 \times 10^7$ m^2), 1.6×10^8 mol/yr; Narrows (area $= 2.2 \times 10^6$ m^2), 1.4×10^7 mol/yr; Upper Arm (area $= 9.4 \times 10^7$ m^2), 3.9×10^8 mol/yr; Lower Arm (area $= 2.4 \times 10^7$ m^2), 2.4×10^8 mol/yr; total $= 8.2 \times 10^8$ mol/yr (Mack 1998).

Based on molybdate inhibition studies, sulfate-reducing bacteria have been widely judged to be the dominant methylators of Hg in marine, estuarine, and freshwater sediments (e.g., Compeau and Bartha 1985, Gilmour and Henry 1991, Kerry et al. 1991, Gilmour et al. 1992, King et al. 2001; see Fleming et al. 2006 for further discussion). However, in a series of molybdate inhibition studies on Clear Lake sediments, using increasing concentrations of molybdate to inhibit the activities of sulfate-reducing bacteria, Mack (1998) showed that while sulfate reduction could be reduced to negligible levels, MeHg production was only reduced by an average of ~40–50%, suggesting a significant additional source of Hg methylation not attributable to sulfate-reducing bacteria (Mack 1998). Further studies have

identified iron-reducing bacteria (*Geobacter* sp.) that may play a previously unappreciated dominant methylating role in these iron-rich sediments (Fleming et al. 2006).

Total mercury and methylmercury transport throughout Clear Lake

Particulate material, containing both TotHg and MeHg on detritus or floc, enters the lake from the mine and/or becomes resuspended from the lake bed and is acted upon by wind-induced currents (Fig. 7, pathways 7 and 8). Clear Lake currents are dominated by the diurnal cycle of wind, interacting with the lake's stratification and the earth's rotation (Rueda et al. 2008). Afternoon and evening winds generate horizontal temperature gradients both along and across the Oaks Arm. Negligible wind forcing during the night and early morning yield baroclinic pressure gradients (along with Coriolis forces) that drive currents up to 10–15 cm/s westward at the surface and eastward near the bottom (Rueda and Schladow 2003). Such current velocities are sufficient to resuspend Hg-laden detritus and floc from the sediments. The change of longitudinal circulation patterns from being driven by wind to being driven by baroclinic forces produces conditions that allow surface currents to converge and bottom currents to diverge, creating a front with significant vertical motions. Such conditions promote the exchange of bottom and surface waters, even under thermally stratified conditions.

The exchange of water and Hg-contaminated particles between the main body of water in each basin and the littoral regions is augmented by a different set of processes. Here, differential heating and cooling between the shallow and deep regions leads to the formation of offshore surface currents and onshore bottom return flows during the day and the reverse at night (Pálmarsson and Schladow 2008, Rueda et al. 2008). This process provides a mechanism whereby suspended material from the pelagic zone can be transported directly to the shore. At times of high lake level, this process would allow the surrounding marshes to act as an effective trap for contaminated particulates.

Distribution of mercury in Clear Lake

Mercury in water and sediments.—Total mercury and MeHg in sediments exhibit a classic point source distribution and an exponential decline with distance from the mine (Suchanek et al. 1998, 2008*b*). Because mining wastes were bulldozed or dumped into the nearshore environment, sediment Hg concentrations in the Oaks Arm near the mine are extremely patchy (see Shipp and Zierenberg 2008). Wind-driven currents transport Hg-laden particles, first from the Oaks Arm to the Upper Arm, then to the Lower Arm, yielding sediment Hg concentrations that are higher in the Upper Arm than in the Lower Arm at comparable distances (Suchanek et al. 2008*b*). Total mercury in unfiltered water (especially near the sediment–water interface) mirrors the pattern of exponential decline with distance exhibited by sediments, but TotHg in filtered water (without Hg-laden particles remobilized from the sediment) does not, indicating that the primary load of TotHg is in the particulate fraction. Lake bed springs do not appear to be a detectable source of TotHg to Clear Lake. From 1992 to 1998, TotHg in sediments remained relatively constant, but MeHg exhibited seasonal minima in winter/spring and maxima in summer/fall when sulfate-reducing bacteria and other methylators (e.g., iron-reducing bacteria; Fleming et al. 2006) exhibit the greatest production of MeHg (Mack 1998). At all sites studied, the percentage of TotHg in the dissolved fraction of water samples exhibited a dramatic seasonal maximum in winter (Suchanek et al. 2008*b*), which is likely related to the ongoing input of dissolved Hg in acid mine drainage entering Clear Lake from the mine during winter rainy periods. Thus, although the 1992 remediation by the U.S. EPA likely reduced the erosion of the waste rock piles and loading of Hg-laden soils from the mine site into Clear Lake (Fig. 7, pathway 1), a significant load of dissolved Hg is still entering the system dissolved in acid mine drainage, especially during winter periods (Fig. 7, pathways 3–5).

During and subsequent to transport throughout the lake and mixing with organic detritus, Hg becomes permanently buried in the sediments (Fig. 7, pathway 17). In 2003, an estimated loading of 320–379 kg Hg dry mass (DM) was added to Clear Lake's surficial sediments (Suchanek et al., *in press*). With a modern lake-wide average sedimentation rate of ~3.5 mm/yr (Richerson et al. 2008), a significant portion of this Hg load becomes buried and represents a stable in situ record of historic events. Some have hypothesized that MeHg is not stable within sediments and undergoes diagenetic upward mobilization through the sediment column (see Suchanek et al. 2008*e* for discussion). Contrary to these previous hypotheses, cores raised during this study show that a significant amount of Hg (both TotHg and MeHg) remains fixed at depth, the most significant peaks being intimately associated with the open-pit mining era (1927–1957). However, elevated TotHg and MeHg also remain stable deep within sediments that are as old as ~3000 ybp.

Mercury in biota.—Mercury, especially bound to particles, is consumed by myriad organisms from plankton to benthic invertebrates to fishes and passed on to higher trophic levels (e.g., birds and humans) either through the benthic pathway (Fig. 7, pathway 15) or the planktonic/pelagic pathway (Fig. 7, pathway 16; Suchanek et al. 2008*c*). Spatially, benthic invertebrates (e.g., chironomid insect larvae and oligochaete worms) and plankton exhibit TotHg (but not MeHg) concentrations that reflect the same trends as water and sediments, a significant exponentially decreasing relationship with distance from the mine. There were no seasonal or long-term trends in TotHg in benthic and planktonic biota, but in both groups MeHg exhibited

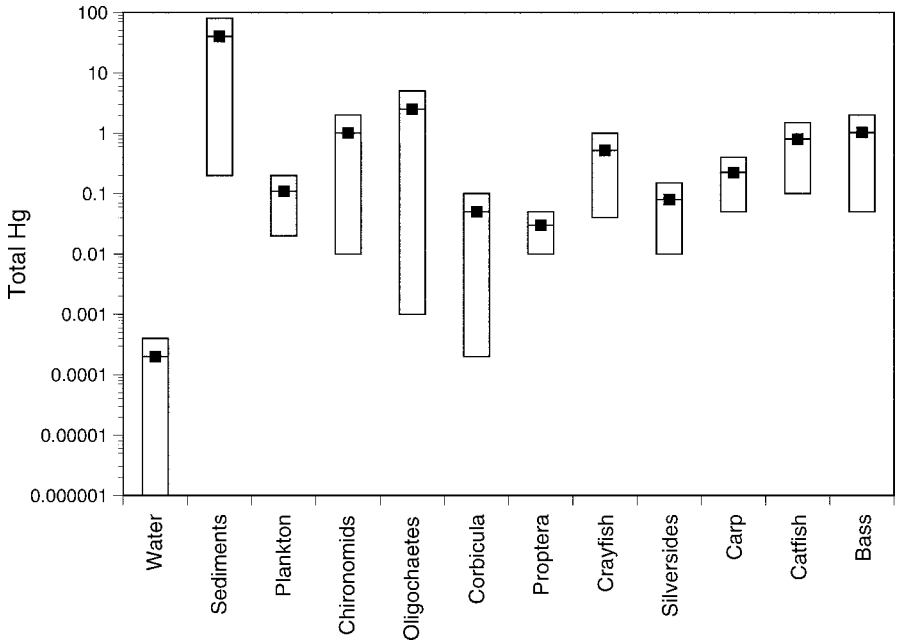

FIG. 8. Range and mean of total mercury (ppm, wet mass) in the abiotic and biotic matrices of the Clear Lake aquatic ecosystem.

maxima in late summer/early fall and minima in winter, comparable to those trends in water and sediments. In comparison with other sites worldwide, Clear Lake's lower trophic species exhibit significantly higher TotHg concentrations, but much lower MeHg (Suchanek et al. 2008*c*). Even at sediment TotHg concentrations of 1–1200 mg/kg wet mass (WM; which exceed known ecological health criteria by up to 7000 times), we could demonstrate no clear, strong toxic effect on Clear Lake's invertebrates, although correlations between Hg concentrations and declines in population numbers or diversity indices for some species were statistically significant (Suchanek et al. 1995).

Clear Lake fishes are represented by several different adult trophic guilds that consume a wide diversity of Hg-contaminated food resources (Suchanek et al. 2008*d*). Maximum muscle Hg concentrations observed for species representing different trophic guilds were (with their prey in parentheses): inland silversides (0.41 mg/kg WM, plankton), common carp (0.66 mg/kg WM, organic detritus), channel catfish (1.50 mg/kg WM, benthic invertebrates), and largemouth bass (1.94 mg/kg WM, other fish species). Interestingly, 11 out of 18 fish species surveyed showed significant population declines with increasing proximity to the mine, but this cannot be attributed conclusively to the influence of Hg or the mine itself (Suchanek et al. 2008*d*).

Analyses of stable isotopes, diet data, and trophic transfer coefficients indicate that Hg bioaccumulation in Clear Lake increases with the degree of benthic foraging and trophic position (Eagles-Smith et al. 2008*a*). Chironomids, which account for the majority of profundal prey, act as an especially important primary conduit for Hg transfer to fishes in Clear Lake. In Clear Lake, fish Hg concentrations also appear to be influenced significantly by the boom and bust cycles of an introduced planktivorous fish species, the threadfin shad (*Dorosoma petenense*). When abundant, this species significantly depletes zooplankton populations, causing other typically planktonic feeding fish species to shift from a 60–80% planktivorous diet to a diet almost entirely composed of zoobenthos, feeding more on chironomids. With this shift to a benthic diet, Hg concentrations increased in these species by ~50% (Eagles-Smith et al. 2008*b*).

Relative mercury contamination, bioaccumulation, and toxicity

Although concentrations of inorganic Hg in the sediments of Clear Lake are very high and there is ongoing loading of dissolved Hg from mine-derived acid mine drainage, this does not appear to have translated into high levels of MeHg production, bioaccumulation, or toxicity that might be expected given the level of Hg contamination present (Suchanek et al. 2008*a*). The lack of connection between TotHg in sediments and water and the concentrations of bioaccumulated MeHg is surprising. The ratio of MeHg:TotHg in sediment and water, often used as a proxy for bioavailability of Hg, decreases with proximity to the mine (Suchanek et al. 2008*a*, *b*). As a consequence, biota in Clear Lake (a highly contaminated system) have proportionately less MeHg in their tissues in comparison to other systems that are only mildly contaminated. The ranges of TotHg in the major Clear Lake compartments analyzed are provided in Fig. 8. The possible reasons for the relatively

low Hg contamination of the Clear Lake food web are discussed in Suchanek et al. (2008*a*).

Fate and effects of mercury in higher vertebrates

In addition to being lethal at high concentrations, Hg is a known mutagen, teratogen, and carcinogen and causes embryocidal, cytochemical, and histopathological effects (Eisler 2000). In higher vertebrates, even at relatively low concentrations, Hg can negatively affect reproduction, growth, development, behavior, blood and serum chemistry, motor coordination, vision, hearing, histology, and metabolism (Heinz 1996, Thompson 1996, Wolfe et al. 1998, Eisler 2000, Ackerman et al. 2007).

Preliminary studies of Hg in Clear Lake mammals and birds conducted in 1993 were reported by Wolfe and Norman (1998). In contrast to Hg in lower trophic invertebrate species and fishes in the lake proper, raccoon and mink Hg body burdens did not correlate with body mass or distance from the mine. Brain Hg concentrations were all below "lowest observed adverse effects level" (LOAEL) and "no observed adverse effects level" (NOAEL) values reported for wild mammals (Wobeser et al. 1976*a*, *b*, Wren 1986). Brain tissues of raccoons and mink did not exhibit signs of histopathologic Hg-induced lesions, nor did they exhibit changes in erythrocyte morphology. A few opportunistic tissue samples of birds from various locations around Clear Lake also were evaluated for tissue Hg concentrations in 1993/1994 by Wolfe and Norman (1998). Although sample sizes were small, brain and liver Hg concentrations from all bird species were below those typically associated with toxicity in young birds, which are 1–10 mg/kg in brain and 5–20 mg/kg in liver (Heinz 1996, Wolfe et al. 1998).

A 10-year study of population status, reproduction, and Hg burdens of two Clear Lake piscivorous birds, Osprey (*Pandion haliaetus*) and Western/Clark's Grebes (*Aechmophorus* sp.), was conducted from 1992 to 2001 (Anderson et al. 2008). In 1992, feather and tissue residue data from local populations of both Ospreys and grebes suggested that Hg residues were high enough to predict ecotoxicological effects (mean Osprey feather Hg = 20.0 mg/kg DM). Interestingly, Hg residues in tissues of grebes from museum specimens from 1967–1969 (closer to the period of active mining) ranged up to 42 mg/kg DM, indicating that Hg reaching picivorous birds was already in a declining phase at the beginning of our studies in 1992. Osprey nests in the Clear Lake system increased from seven in 1992 to more than 30 nests in 2005; during 1992–2005, reproductive success at Clear Lake was comparable to Eagle Lake, a California site not contaminated with Hg. Grebes exhibited a similar pattern of increase in both locations. However, the increase at Clear Lake was heavily influenced by improved management practices that protected nesting areas on the lake from boat traffic. Nonetheless, results from a post-study survey in 2003–2004 show that Hg residues in Osprey feathers again increased to concentrations that were even higher than those during 1992. Significant fluctuations in Hg concentrations in Osprey feathers may reflect concomitant fluctuations in prey fish (Suchanek et al. 2008*d*), which in Clear Lake are in turn likely being driven by the alien threadfin shad, *Dorosoma petenense* (see Anderson et al. 2008, Eagles-Smith et al. 2008*b*).

Human health

Human health studies of Hg contamination at Clear Lake have not addressed the lake's population as a whole. Many individuals residing in the Clear Lake area are retired or live on other sources of fixed income, and some supplement their diets with fish caught from the lake. Additionally, individuals who work in the fish-related industries at Clear Lake (tourism and commercial fishing), and the families of these individuals, may also consume Clear Lake fish in larger quantities than other citizens. Only one human health survey has been conducted on residents to evaluate Hg levels in blood, urine, and hair, done in 1992 (ATSDR 1992, CDHS 1996). (Interestingly, 1992 was a year when fish Hg [as evidenced by juvenile largemouth bass Hg burdens] exhibited some of the lowest concentrations over the past 30 years. On the other hand, in 2004, juvenile largemouth bass exhibited the highest Hg concentrations during this 30-year period [Suchanek et al. 2008*d*, Eagles-Smith et al. 2008*b*].) This survey was conducted on 63 individuals, including primarily Elem Pomo Indians who reside in the Elem Rancheria adjacent to the mine and other individuals that frequented the Rancheria. Results of this study showed that Hg in hair ranged from 0.3 to 2.3 μg/g (ppm), Hg in urine ranged from 0.2 to 6.1 μg/g (adjusted for creatinine; the typical range for a non-contaminated population was listed as 0–20 μg/g), inorganic Hg in blood ranged from 0.7 to 4.7 μg/L (= ppb), and organic Hg in blood ranged from 2.5 to 38.8 μg/L (the typical range for a fish-eating population was listed as 0–20 μg/L). The mean concentration of organic Hg in blood in tribal members at 15.6 μg/L, with a maximum of 38.8 μg/L, is significantly higher than the average U.S. population (Schober et al. 2003). Although several Elem residents exceeded typical ranges for blood Hg concentrations, these concentrations did not reach levels known to be associated with clinical symptoms (CDHS 1996). However, the National Research Council and the U.S. EPA established a reference dose level of blood Hg corresponding to a concentration of 5.8 μg/L in cord blood to prevent developmental effects from in utero MeHg exposure (NAS 2000, Rice et al. 2003). The Elem study also found a correlation between blood organic Hg and consumption of fish by Elem residents and indicated that an estimated 10% of the adults exceeded the state fish consumption advisory guidelines established in 1987. Elem residents were well aware of studies documenting elevated levels of Hg in fish and had

reduced their consumption of fish considerably by the time the Department of Health Services study was conducted (J. Brown, Tribal Leader, *personal communication*). Their body burdens during the time when they consumed larger quantities of fish were, unfortunately, undocumented. One recommendation of the reevaluation of the 1992 study was to evaluate the possibility of conducting additional biological testing for blood Hg among Lake County residents who frequently consume Clear Lake fish in excess of fish consumption guidelines (CDHS 1996). To our knowledge, this recommendation has not been implemented.

Acknowledgments

Deep appreciation is extended to the many staff from the UC–Davis Clear Lake Environmental Research Center (CLERC) at Clear Lake who contributed greatly to the collection and processing of data and samples, analyses, and discussions over the entire 15-year period of this study. Particular thanks go to Jesse Becker, Amanda Bern, Linnie Brister, J. R. Flanders, Brenda Johnson, Brad Lamphere, Anne Maxson, Laurent Meillier, Lauri Mullen, Mark Sexton, Mike Shaver, Brian Swisher, Lori Webber, Steve Why, and Cat Woodmansee. Their long hours of dedicated efforts have produced a wealth of long-term data from which this series of papers is derived. Steve Hill, Tom Smythe, Robert Reynolds, and Sue Arterburn smoothed collaboration with Lake County on the fieldwork for this project, and Robert Lossius provided support during the latter stages. The Lake County Board of Supervisors was highly supportive of our work, with special thanks to Karan Mackey and Louise Talley. Secchi disk data were provided by Jerry Bowles, Department of Water Resources, Northern District. Wind data were provided by the County of Lake Air Quality Management District. We thank Monica Choi, Ann Houck, and Robert Kaufman for useful discussions and Karen Phillips and Julie Yee for constructive comments on earlier drafts. This work was supported by the U.S. EPA-funded (R819658 and R825433) Center for Ecological Health Research at UC Davis, U.S. EPA Region IX Superfund Program (68-S2-9005), and UC Davis. Thanks to Carolyn d'Almeida and Jeri Simmons of U.S. EPA Region IX Superfund and to Karen Moorehouse of EPA-ORD for assistance in connection with this funding. We are extremely grateful to Dennis Rolston and Cheryl Smith who helped make the final compilation of studies in this issue a reality. Finally, special appreciation goes to Anne Fairbrother and Betsy Henry, guest editors, who did the heavy lifting of making sure the Special Issue manuscripts were reviewed in a timely and efficient manner by well-qualified reviewers. Although the information in this document has been funded wholly or in part by the U.S. EPA, it may not necessarily reflect the views of the Agency, and no official endorsement should be inferred. Any use of trade, product, or firm names in this publication is for descriptive purposes only and does not imply endorsement by the U.S. government.

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