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THE BASIS FOR ECOTOXICOLOGICAL CONCERN IN AQUATIC ECOSYSTEMS CONTAMINATED BY HISTORICAL MERCURY MINING

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Abstract. The Coast Range of California is one of five global regions that dominated historical production of mercury (Hg) until declining demand led to the economic collapse of the Hg-mining industry in the United States. Calcines, waste rock, and contaminated alluvium from inactive mine sites can release Hg (including methylmercury, MeHg) to the environment for decades to centuries after mining has ceased. Soils, water, and sediment near mines often contain high concentrations of total Hg (TotHg), and an understanding of the biogeochemical transformations, transport, and bioaccumulation of this toxic metal is needed to assess effects of these contaminated environments on humans and wildlife. We briefly review the environmental behavior and effects of Hg, providing a prelude to the subsequent papers in this Special Issue. Clear Lake is a northern California lake contaminated by wastes from the abandoned Sulphur Bank Mercury Mine, a U.S. Environmental Protection Agency Superfund Site. The primary toxicological problem with Hg in aquatic ecosystems is biotic exposure to MeHg, a highly toxic compound that readily bioaccumulates. Processes that affect the abundance of MeHg (including methylation and demethylation) strongly affect its concentration in all trophic levels of aquatic food webs. MeHg can biomagnify to high concentrations in aquatic food webs, and consumption of fish is the primary pathway for human exposure. Fish consumption advisories have been issued for many North American waters, including Clear Lake and other mine-impacted waters in California, as a means of decreasing MeHg exposure. Concerns about MeHg exposure in humans focus largely on developmental neurotoxicity to the fetus and children. Aquatic food webs are also an important pathway for MeHg exposure of wildlife, which can accumulate high, sometimes harmful, concentrations. In birds, wild mammals, and humans, MeHg readily passes to the developing egg, embryo, or fetus, life stages that are much more sensitive than the adult. The papers in this issue examine the origin, transport, transformations, bioaccumulation, and trophic transfer of Hg in Clear Lake, assess its potential effects on biota and humans, and provide information relevant to remediation of mine-impacted aquatic ecosystems.

Key words: bioaccumulation; biomagnification; Clear Lake, California, USA; environmental transport; human exposure; mercury; methylmercury; mining; toxicity.

Introduction

Mercury (Hg) has a long history of human usage, including mining for precious metals and an array of industrial, domestic, and agricultural applications (Hylander and Meili 2005). Beginning in the late 1960s, increasing awareness of the hazards of Hg exposure prompted widespread discontinuation or phased reductions in use of the metal in many applications and goods and regulation of many industrial emissions of Hg to receiving waters (Wiener et al. 2003). The rapid declines in demand and prices for Hg precipitated abrupt decreases in Hg mining and the eventual economic collapse of Hg-mining operations in the United States (Jasinski 1995) and elsewhere (Hylander and Meili 2003).

The mountainous Coast Range in the state of California (USA) was one of five mining regions that dominated the historical global production of elemental Hg (Jasinski 1995, Ferrara 1999). The other regions were the Almadén district in Spain, the Idrija district in Slovenia, the Monte Amiata district in Italy, and the Huancavelica district in Peru. Mining for gold and other precious metals was the primary use of Hg in the United States during the latter half of the 1800s, and the mining of Hg deposits (primarily cinnabar ore, HgS) in the Coast Range of California (Fig. 1) was stimulated by the demands created by gold and silver mining (Averill 1946, Jasinski 1995, Alpers et al. 2005). Approximately 100 000 Mg of Hg were mined from the Coast Range.

The mining operations, emissions, and environmental contamination associated with Hg mining at Almadén (Spain), Idrija (Slovenia), and Mt. Amiata (Italy), three

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sites that together accounted for two-thirds of the estimated total global production of Hg (i.e., ~530 000 of 800 000 Mg), have been reviewed by Ferrara (1999). Operations at these sites involved mining of cinnabar ores, which were transported to smelters, crushed, and roasted at 650°–700°C. The Hg vapors released by roasting were condensed in cooling towers and placed in flasks for transport. In mining operations before ca. 1960, from 20% to 40% of the Hg in processed ores was released to the surrounding environment (Ferrara 1999).

Thus, the air, soil, water, and sediment in the vicinity of Hg-mining and smelting operations often contain high concentrations of total Hg (TotHg) (Gosar et al. 1997, Ferrara 1999, Turner and Southworth 1999, Gray et al. 2000, Lockhart et al. 2000). Moreover, calcines (roasted ores), waste rock, and contaminated alluvium from mining sites can release Hg (including methylmercury, MeHg) to the environment for decades or centuries after mining and smelting operations have ceased (Ganguli et al. 2000, Hines et al. 2000, Rytuba 2000, Covelli et al. 2001, Domagalski et al. 2004, Gray et al. 2004, Lowry et al. 2004). Concentrations of MeHg in benthic invertebrates (Žižek et al. 2007), fish, and fish-eating birds (Gray et al. 2000, Weech et al. 2004, 2006) are substantially greater in waters affected by Hg mines than in unaffected reference waters. Concentrations of MeHg and TotHg are also substantially elevated in terrestrial plants and wildlife at sites contaminated by historic Hg-mining operations (Gnamuš and Horvat 1999).

In Pinchi Lake (British Columbia, Canada), for example, cinnabar-containing waste ore from an adjacent Hg mine was deposited into the lake during 1940–1944 (Plouffe et al. 2004), significantly contaminating sediments in Pinchi Lake and downstream Stuart Lake (Lockhart et al. 2000). Although the most Hg-contaminated sediments have been buried under subsequent deposits (Lockhart et al. 2000), fish and fish-eating birds sampled from Pinchi Lake in 2001–2002 contained substantially higher concentrations of Hg than fish and birds sampled concurrently from nearby lakes unaffected by Hg-mining activities (Weech et al. 2004, 2006). This indicates that Hg from mining operations may continue to be methylated and bioaccumulated as MeHg for decades after Hg-mining operations cease.

About 12 000 Mg of Hg^0 mined in California were used in the state, mostly in gold-mining operations in the Sierra Nevada and Klamath-Trinity Mountains (Averill 1946, Alpers et al. 2005). Substantial quantities of elemental Hg (Hg^0) were released to the environment at gold-mining sites (Fig. 1). At a typical hydraulic gold-mining site in California, for example, several hundred kilograms of Hg^0 would be added to a single sluice to recover gold through amalgamation. An estimated 10–30% of the Hg^0 used in gold mining in California was released to the environment (Averill 1946, Alpers et al. 2005). Total anthropogenic emissions of Hg in North America during 1995–2000 were ~200 Mg/yr (Pacyna et al. 2006); thus, the estimated 1200–3600 Mg of Hg^0 released to the environment of California during gold-mining operations represents a substantial anthropogenic source at the continental scale. In many developing countries, there has been a resurgence in the use of Hg in gold mining in recent decades, in small-scale (artisanal) mining operations to amalgamate gold, exposing millions of miners and their families to high concentrations of Hg^0 vapor (Swain et al. 2007).

The mining, extraction, redistribution, and widespread use of Hg, followed by decades of environmental transport and redistribution, has left California and other regions in the western United States with a legacy of Hg-contaminated streams, rivers, reservoirs, and floodplains down-gradient from historic mining sites in the Coast Ranges and Sierra Nevada extending through San Francisco Bay (Hornberger et al. 1999, Domagalski et al. 2004, Heim et al. 2007). While there are a variety of environmental disturbances from Hg, gold, and silver mines and prospects in California, very few sites have undergone extensive remediation to lessen the impacts of Hg on humans and wildlife. However, many Hg-contaminated mining sites in California and Nevada are undergoing investigation. Environments surrounding the Sulphur Bank Mercury Mine in California and the Carson River/Lahontan Reservoir (gold and silver mining) region in Nevada, both U.S. Environmental Protection Agency (EPA) Superfund Sites, contain very high concentrations of TotHg in water and sediment. Given the geographic extent and intensity of such environmental contamination, information on the cycling, transport, transformations, and bioaccumulation of Hg in environments affected by inactive Hg-, gold-, and silver-mining sites is needed to assess the potential consequences of this contamination. Results of these investigations can inform management decisions at mining sites where Hg is a contaminant of concern.

In North America, many investigations of environmental Hg pollution during recent decades have focused on ecosystems contaminated by atmospheric deposition (Lamborg et al. 2002, Grigal 2003, Branfireun et al. 2005, Orihel et al. 2006, Lindberg et al. 2007) or industrial sources (Rudd et al. 1983, Turner and Southworth 1999, Wiener and Shields 2000). Recent studies have also focused on systems with high rates of MeHg production, such as newly flooded reservoirs (Bodaly et al. 2004, St. Louis et al. 2004) and wetlands (St. Louis et al. 1996, Gilmour et al. 1998, Branfireun et al. 2005). Many areas in California (Fig. 1), the western United States, and Alaska contain abandoned mine sites (including Hg, gold, and silver mines) that continue to release significant amounts of Hg into down-gradient aquatic environments.

The transport, distribution, transformation, and bioaccumulation of Hg in mining-impacted landscapes have received increasing study in recent years, and it is evident that some aspects of the physical transport, biogeochemical transformations, uptake, and effects of

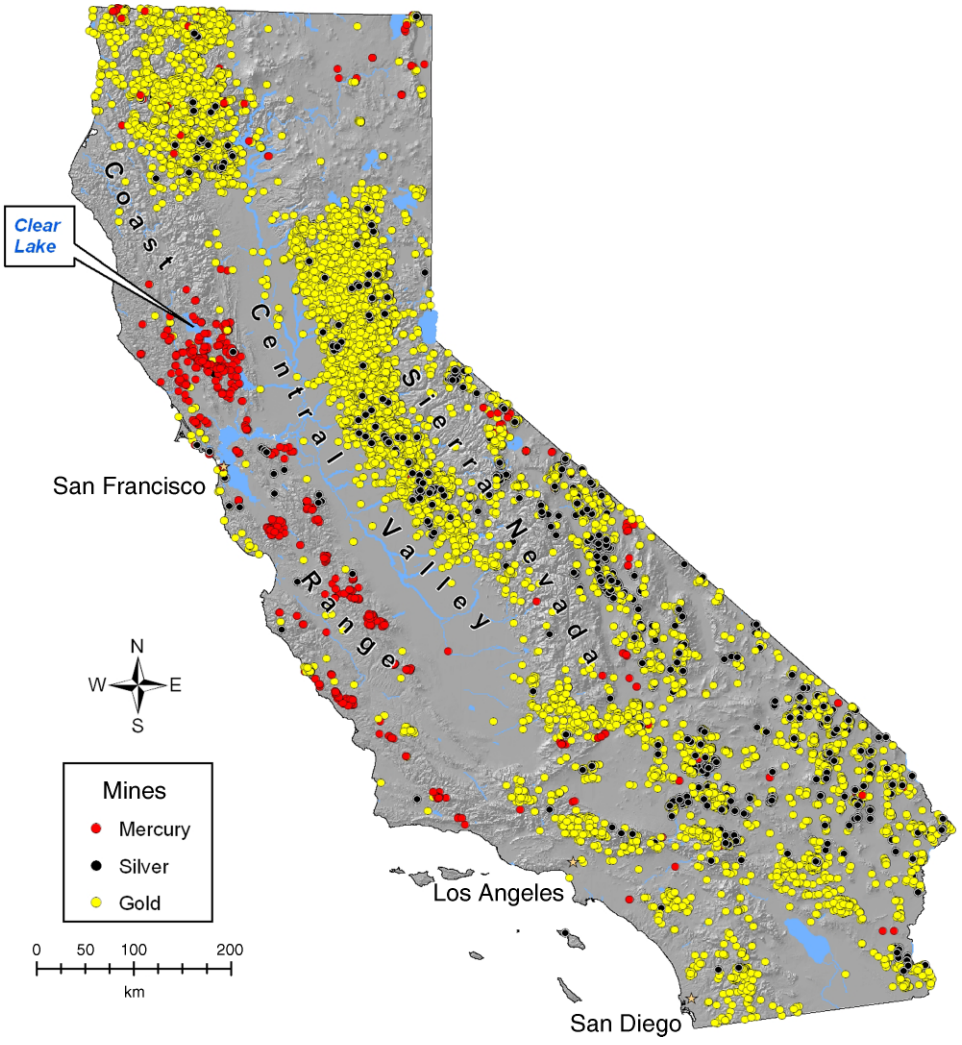

FIG. 1. Locations of Clear Lake and of known historic sites of mercury, gold, and silver mines and prospects in California, USA. Data were compiled from the Department of Conservation (California Geological Survey, Sacramento) and the U.S. Geological Survey (Sacramento).

Hg from Hg-mining sites differ substantially from that at sites dominated by Hg from atmospheric deposition, industrial sources, and gold mines. At Hg-mining sites, the total masses of Hg are large, existing mostly as particulate Hg-sulfides (cinnabar and metacinnabar; Lockhart et al. 2000, Rytuba 2000, Lowry et al. 2004). At Clear Lake, Hg from calcines and waste rock from the Sulphur Bank Mercury Mine is probably transported largely as colloidal and fine-grained cinnabar and metacinnabar (Lowry et al. 2004). Cinnabar and metacinnabar have low solubility under oxic conditions, leading one to expect that the bioavailability of Hg in these forms to methylating bacteria would be low. Yet mine wastes, stream sediments, and surface waters at the Almadén Mining District in Spain, the world's largest Hg-producing region, contain very high concentrations of MeHg (Gray et al. 2004). In anoxic, sulfidic sediments, cinnabar can dissolve and become available for methylation (Benoit et al. 2001). Organic acids from vegetation can enhance the dissolution of cinnabar (Ravichandran et al. 1998) and increase the transport of colloidal Hg from former mining sites (Slowey et al. 2005).

This issue provides a comprehensive assessment of environmental and biotic impairment of the Clear Lake ecosystem from the Sulphur Bank Mercury Mine, an abandoned Hg mine site in the Cache Creek drainage basin in the Coast Range of northern California. The primary objective of this introductory paper is to briefly review the ecotoxicological effects of Hg, providing a prelude to the subsequent research papers from the Clear Lake investigation. We do not attempt to review the biogeochemistry of Hg in mine-impacted surface waters, but instead focus on the rationale for concern about Hg pollution and its adverse effects in aquatic ecosystems. A synthesis of information on the Clear

Lake Hg investigation is provided by Suchanek et al. (2008*e*).

Mercury in Aquatic Ecosystems and Food Webs

Toxicological concerns about Hg pollution of aquatic ecosystems focus on MeHg, a highly toxic, organometallic compound that readily accumulates in exposed aquatic organisms and biomagnifies in food webs (Wiener et al. 2003). Although most of the Hg in terrestrial and aquatic environments exists as inorganic forms, nearly all of the Hg accumulated by fish and higher trophic levels is MeHg (Grieb et al. 1990, Bloom 1992, Hammerschmidt et al. 1999), even in surface waters containing unusually high concentrations of inorganic Hg (Southworth et al. 1995, Kuwabara et al. 2007). Methylmercury readily crosses the lining of the gastrointestinal tract and other internal biological membranes (Pickhardt et al. 2006), is eliminated slowly relative to its rate of uptake (Trudel and Rasmussen 1997, Van Walleghem et al. 2007), and accumulates to concentrations in aquatic organisms that vastly exceed those in the surrounding water. In fish, for example, concentrations of MeHg commonly exceed those in the water in which they reside by a factor of 10^6–10^7 or more (Wiener et al. 2003). Direct uptake from water is important for organisms, such as algae, in the lowest trophic levels (Pickhardt et al. 2002, Gorski et al. 2006), whereas aquatic organisms, such as fish, in upper trophic levels obtain MeHg almost entirely from the diet (e.g., Rodgers 1994, Hall et al. 1997, Harris and Bodaly 1998). Characteristic patterns in the biomagnification of MeHg are evident across ecosystems that differ in type of water body, Hg source, and pollution intensity (Wiener et al. 2003). For example, the concentration of MeHg increases up the food web from water and lower trophic levels to fish and piscivores, the greatest increase in concentration occurs in the trophic step between water and algae, and the fraction of TotHg present as MeHg increases with ascending trophic level from algae through fish.

In contrast to MeHg, inorganic Hg^{II} and Hg^0 in natural waters are not readily transferred through successive trophic levels and do not biomagnify in food webs (Watras et al. 1998, Kim and Burggraaf 1999, Pickhardt et al. 2002). In a toxicological sense, the primary problem with Hg in aquatic ecosystems stems from biotic exposure to, or bioaccumulation of, MeHg (Wiener et al. 2003).

Processes that affect the mass of MeHg in aquatic ecosystems or its concentration at the base of the aquatic food web strongly affect its concentration in all trophic levels, including predatory fish and wildlife (Paterson et al. 1998, Benoit et al. 2003, Wiener et al. 2003). Such processes include the production of MeHg via the microbial methylation of inorganic Hg^{II} (Benoit et al. 2003) and the destruction of MeHg by photodemethylation (Sellers et al. 1996, 2001) and microbial demethylation (Oremland et al. 1991, Marvin-DiPasquale et al. 2000). Anaerobic zones in sediments, hypolimnia, and wetlands are the most important sites of microbial methylation, and a water body can receive MeHg from both internal and external sites (Watras et al. 1994, Sellers et al. 2001). Wetlands are important sites of MeHg production and export to adjacent or downstream waters (Hurley et al. 1995, St. Louis et al. 1996, Sellers et al. 2001, Wiener et al. 2006). Concentrations of MeHg in phytoplankton, zooplankton, and higher trophic levels can also be influenced by biodilution of MeHg at the base of the food web by algal blooms or high algal biomass (Pickhardt et al. 2002, 2005, Chen and Folt 2005).

Exposure of Humans and Wildlife

Aquatic food webs are the primary pathway of MeHg exposure in most human populations, given that finfish, marine mammals, and shellfish are the principal sources of MeHg in the human diet (NRC 2000, Mahaffey et al. 2004, Clarkson and Magos 2006). Elevated MeHg exposure in human populations with high levels of fish consumption has been documented around the globe, unconstrained by geographic, social, economic, or cultural boundaries (Mergler et al. 2007). To reduce human exposure to MeHg, fish consumption advisories have been issued for many lakes, rivers, and coastal waters, providing guidance on the number of meals and species of fish that can be eaten safely (U.S. EPA 2007). The State of California first issued a fish consumption advisory for Clear Lake in 1987; this advisory was recently updated to include recommendations based on analyses of additional data for Clear Lake and nearby water bodies (Gassel et al. 2005).

Methylmercury contamination has adversely affected the benefits derived from fishery resources in many inland and coastal waters. In the United States, MeHg was responsible for 80% or 3080 of the fish consumption advisories posted in 2006, when 48 states, one territory, and two tribes had advisories attributed to MeHg (U.S. EPA 2007). The number of statewide fish consumption advisories issued for coastal waters, lakes, and rivers in the United States has increased substantially since 1993 (Wiener et al. 2003, U.S. EPA 2007). In 2006, 23 states had Hg-related, statewide fish consumption advisories for lakes, 21 had statewide advisories for rivers, and 13 had statewide advisories for coastal waters. More than 57 400 km^2 of lake area and 1 420 000 km of rivers in the United States were under advisory for Hg in 2006. In Canada, more than 97% (2572) of all fish consumption advisories listed in 1997 were attributed to Hg (U.S. EPA 2001). In California, many of the lakes, rivers, and reservoirs with fish consumption advisories for Hg are mining-impacted systems (OEHHA 2007).

The consumption of fish and aquatic organisms is also an important pathway for MeHg exposure of wildlife, including birds, mammals, and reptiles (Wiener et al. 2003). Moreover, wildlife atop aquatic food webs can bioaccumulate high concentrations of MeHg (Wolfe et

al. 1998, Wiener et al. 2003, Ackerman et al. 2007, 2008, Scheuhammer et al. 2007).

Adverse Effects

The uptake, distribution, and effects of MeHg in humans have been recently reviewed in detail (Clarkson and Magos 2006, Mergler et al. 2007). To summarize briefly, MeHg in ingested food is very efficiently absorbed across the gut, enters the bloodstream, and is rapidly transported to all tissues and organs, readily crossing both the blood–brain and placental barriers. Methylmercury is extremely neurotoxic, adversely affecting both the adult and developing brain, and damage to the central nervous system is irreversible. In adults exposed to lethal doses of MeHg, a substantial latent period (months) precedes the onset of symptoms. In lethal and severe cases of MeHg poisoning in adults, paresthesia has been the first symptom to appear, followed in rapid succession by ataxia (loss of voluntary muscular coordination), dysarthria (loss of speech), impaired hearing, constriction of the visual fields, and loss of vision. Fetal exposure occurs via the maternal diet, and the fetus is highly sensitive to MeHg because of its developmental neurotoxicity. Accordingly, toxicological concern about human exposure to MeHg has focused largely on women of childbearing age, the fetus, and children (Schober et al. 2003, Mahaffey et al. 2004, Gassel et al. 2005, Oken et al. 2005). Some recent studies suggest that exposure to MeHg could increase the risk of adverse cardiovascular effects in humans, including adult males (Mergler et al. 2007).

Present exposures to MeHg in human populations are much lower than those that caused the historic epidemics of severe Hg poisoning in Minamata, Japan, a few decades ago (Mergler et al. 2007). Yet persons who consume significant quantities of predatory fish can accumulate harmful doses of MeHg. At present exposure levels, concerns regarding health effects of MeHg exposure focus on reduced neurologic status and slower development in infants and children exposed to MeHg in the womb and during early childhood. In children, for example, in utero exposure to MeHg has been associated with lower performance on tests of language, attention, memory, visuospatial, and motor functions (Mergler et al. 2007).

The impacts of contaminated fishery resources on humans are not limited to the direct effects of MeHg exposure. In Canada, for example, some aboriginal communities that had relied on subsistence fishing have suffered adverse cultural, social, health, and economic effects as a result of industrial Hg pollution (Wheatley 1997, Wheatley et al. 1997, Wheatley and Wheatley 2000). For these communities, abandonment of subsistence fishing was followed by a change to less healthy diets, and the disruption of lifestyle led to social and cultural upheaval. These multidimensional effects have presented a more severe overall problem for the affected communities than the direct, clinical effects of exposure to MeHg via consumption of contaminated fish (Wheatley and Wheatley 2000).

In birds and mammals, MeHg in reproducing females readily passes to the developing egg or embryo, life stages that are much more sensitive than the adult to MeHg exposure (reviewed by Wolfe et al. 1998, Wiener et al. 2003, Scheuhammer et al. 2007). In birds, for example, the dietary concentrations of MeHg that significantly impair reproduction are only one-fifth of those that produce overt toxicity in the adult (Scheuhammer 1991). Reproductive impairment has been associated with high MeHg exposure in field studies of several aquatic and marsh birds (Wiener et al. 2003, Heath and Frederick 2005, Scheuhammer et al. 2007), including populations of the endangered California Clapper Rail (*Rallus longirostris obsoletus*) nesting in the San Francisco Bay-Delta estuary (Schwarzbach et al. 2006). In laboratory experiments with birds and mammals, MeHg adversely affects adult survival, reproductive success, behavior, and neurological development, reduces immune resistance to disease, and causes teratogenic effects (Wolfe et al. 1998, Spalding et al. 2000, Wiener et al. 2003, Scheuhammer et al. 2007). Recent experiments have also shown that exposure of fish to environmentally realistic concentrations of MeHg can impair foraging efficiency and adversely affect endocrine systems and reproduction (Fjeld et al. 1998, Latif et al. 2001, Hammerschmidt et al. 2002, Drevnick and Sandheinrich 2003, Scheuhammer et al. 2007). Diminished reproductive success could have adverse population-level consequences for fish and wildlife species exposed to high levels of MeHg.

The Clear Lake Study

Clear Lake is a 177-km^2 eutrophic lake in Lake County, California. The lake, which is described elsewhere (Suchanek et al. 2003, 2008*e*), was selected for an ecosystem-scale investigation of Hg cycling and effects for several reasons. First, the dominant source of Hg in the lake was (and remains) the now-inactive Sulphur Bank Mercury Mine (Suchanek et al. 2008*e*; Suchanek et al., *in press*). During operation of the mine, Hg-laden tailings and waste rock that were not of sufficient quality for processing were bulldozed into the lake for disposal (Suchanek et al. 2008*a*, *e*). Second, the cycling, transport, distribution, transformations, bioaccumulation, trophic transfer, and ecotoxicological effects of mine-derived Hg had not been studied at the ecosystem scale in a system of this type. Third, the distribution, transport, and cycling of Hg within the lake were sufficiently constrained within the basin to identify key inputs, outputs, and inventories (Rueda et al. 2008; Suchanek et al., *in press*). Fourth, the lake has received one of the highest loadings of inorganic Hg of any site worldwide (Suchanek et al. 2008*a*, *e*), and the distribution of Hg from mining sources can be characterized spatially and temporally in the physical and biotic components of the ecosystem (Anderson et al. 2008,

Suchanek et al. 2008*a*, *b*, *c*). Fifth, the pronounced spatial gradients in Hg concentrations that extend from the mine site to the furthest end of the lake provide a template for assessing the transport, cycling, and bioaccumulation of Hg from the mine through several levels of the food web. Sixth, the availability of coring data from deep (28–177 m) sedimentary strata deposited in the lake as early as 2- to 3-million years before present allows comparison of prehistoric and modern rates of Hg accumulation in the lake bottom (Sims and White 1981, Sims et al. 1988, Osleger et al. 2008, Richerson et al. 2008, Suchanek et al. 2008*d*). Lastly, Clear Lake is representative of many other aquatic ecosystems that are contaminated with Hg from mining sources, such as Pinchi and Stuart lakes in British Columbia, Canada (Lockhart et al. 2000, Weech et al. 2004, 2006).

The papers in this issue collectively provide a comprehensive assessment of environmental and biotic impairment of the Clear Lake ecosystem from the Sulphur Bank Mercury Mine. This dedicated issue examines the origin, transport, transformations, bioaccumulation, trophic transfer, and effects of this Hg on resident biota and humans in this ecosystem, providing a holistic view of the effects of the Hg mine on a lacustrine ecosystem, as well as information relevant to remediation.

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