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Arsenic and mercury contamination related to historical gold mining in the Sierra Nevada, California

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Abstract: Arsenic (As) is a naturally occurring constituent in low-sulphide gold-quartz vein deposits, the dominant deposit type for lode mines in the Sierra Nevada Foothills (SNFH) gold (Au) province of California. Concentrations of naturally occurring mercury (Hg) in the SNFH Au province are low, but extensive use and loss of elemental Hg during amalgamation processing of ore from lode and placer Au deposits led to widespread contamination of Hg in the Sierra Nevada foothills and downstream areas, such as the Sacramento–San Joaquin Delta and San Francisco Bay. This review paper provides an overview of As and Hg contamination related to historical Au mining in the Sierra Nevada of California. It summarizes the geology, mineralogy, and geochemistry of the Au deposits, and provides information on specific areas where detailed studies have been done in association with past, ongoing, and planned remediation activities related to the environmental As and Hg contamination.

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Arsenic is a naturally occurring constituent in low-sulphide Au-quartz vein deposits, the dominant deposit type for lode mines in the Sierra Nevada Foothills (SNFH) Au province (Ashley 2002). Because of elevated concentrations of As in accessory iron-sulphide minerals including arsenopyrite (FeAsS) and arsenian pyrite ($Fe(S,As)_2$), As is commonly a contaminant of concern in lode Au mine waste, including waste rock and mill tailings. The principal pathways of human As exposure from mine waste include ingestion of soil or drinking water, and inhalation of dust in contaminated areas (Mitchell 2014).

Concentrations of naturally occurring Hg in the SNFH Au province are low, but extensive use and loss of elemental Hg during amalgamation processing of ore from lode and placer Au deposits (Churchill 2000) led to widespread contamination of Hg in the Sierra Nevada foothills and downstream areas, such as the Sacramento–San Joaquin Delta and San Francisco Bay (Alpers *et al.* 2005*a*). Conversion of Hg to monomethylmercury (MeHg) by sulphate-reducing and iron-reducing microbes facilitates its bioaccumulation (Wiener *et al.* 2003). The human Hg exposure pathway of main concern is ingestion of MeHg from sport (non-commercial) fish, especially higher trophic levels such as bass species (Davis *et al.* 2008). Wildlife exposure to MeHg is also a concern because of chronic and reproductive effects, for example in fish-eating and invertebrate-foraging birds (e.g. Wiener *et al.* 2003; Eagles-Smith *et al.* 2009; Ackerman *et al.* 2016).

Geological Setting

The geology of the Sierra Nevada region is dominated by a large granitic batholith, emplaced between 151 and 80 Ma (Goldfarb *et al.* 1998), which is exposed at higher elevations in the eastern part of the region (Fig. 1). In the western foothills, metamorphosed sedimentary and volcanic rocks were deformed between 155 and 123 Ma (Goldfarb *et al.* 1998). Mineralization of low-sulphide Au-quartz vein deposits took place between about 144 and 107 Ma (Böhlke & Kistler 1986; Goldfarb *et al.* 1998; Böhlke 1999).

Low-sulphide Au-quartz vein deposits in the SNFH Au province formed from upwelling metamorphic fluids rich in carbon dioxide (Böhlke 1989). In addition to quartz, common gangue minerals include carbonates such as calcite, dolomite, ankerite, and siderite-magnesite solid solution (Böhlke 1988). Host rocks for the vein deposits include the aforementioned metamorphic and igneous rocks. Typically the vein deposits are associated with steeply-dipping thrust faults including the Melones Fault Zone in the Mother Lode region, with similar deposits in the adjacent West Belt, East Belt, and Northern Mines (Fig. 1b; Tuminas 1983; Ashley 2002; Ernst *et al.* 2008*a*, *b*).

The vein deposits currently exposed at the surface were originally formed at depths of *c.* 5 – 8 km in the mesozonal environment, where As typically occurs with Au (Groves *et al.* 1998; McCuaig & Kerrich 1998; Ashley 2002). Other trace metals typically associated with lode Au deposits in the SNFH province include silver (Ag), lead (Pb), copper, and zinc (Ashley 2002); locally there may be enrichment in chromium, nickel, cobalt, vanadium, tellurium, platinum group elements, and/or scandium, which has been attributed to hydrothermal scavenging from wall rocks (Savage *et al.* 2000*a*; Ashley 2002).

Mining History

The discovery of Au in January 1848 by James Marshall at Sutter's Mill (Fig. 1c) on the South Fork American River at Coloma, California, was followed by the well-known 1849 Gold Rush (Clark 1970). Gold production in California was dominated by hydraulic mining of placer deposits from the 1850s until 1884, when the practice was largely shut down by the decision of Judge Lorenzo Sawyer in the case *Woodruff v. North Bloomfield Mining and Gravel Company* (Kelley 1959). After 1893, some hydraulic Au mining resumed under the Caminetti Act, a federal law requiring impoundment of tailings (James 2005). Gold production from underground lode mining increased during the 1890s (Averill 1946; Hagwood 1981; Kelley 1984). During the early 1900s up to World War I (1914 – 17), large-scale dredging of placer deposits became an important source of Au in California (Averill 1946). During the 1920s Au production declined, but it picked up greatly from dredges and underground mines during the Great Depression of the 1930s,

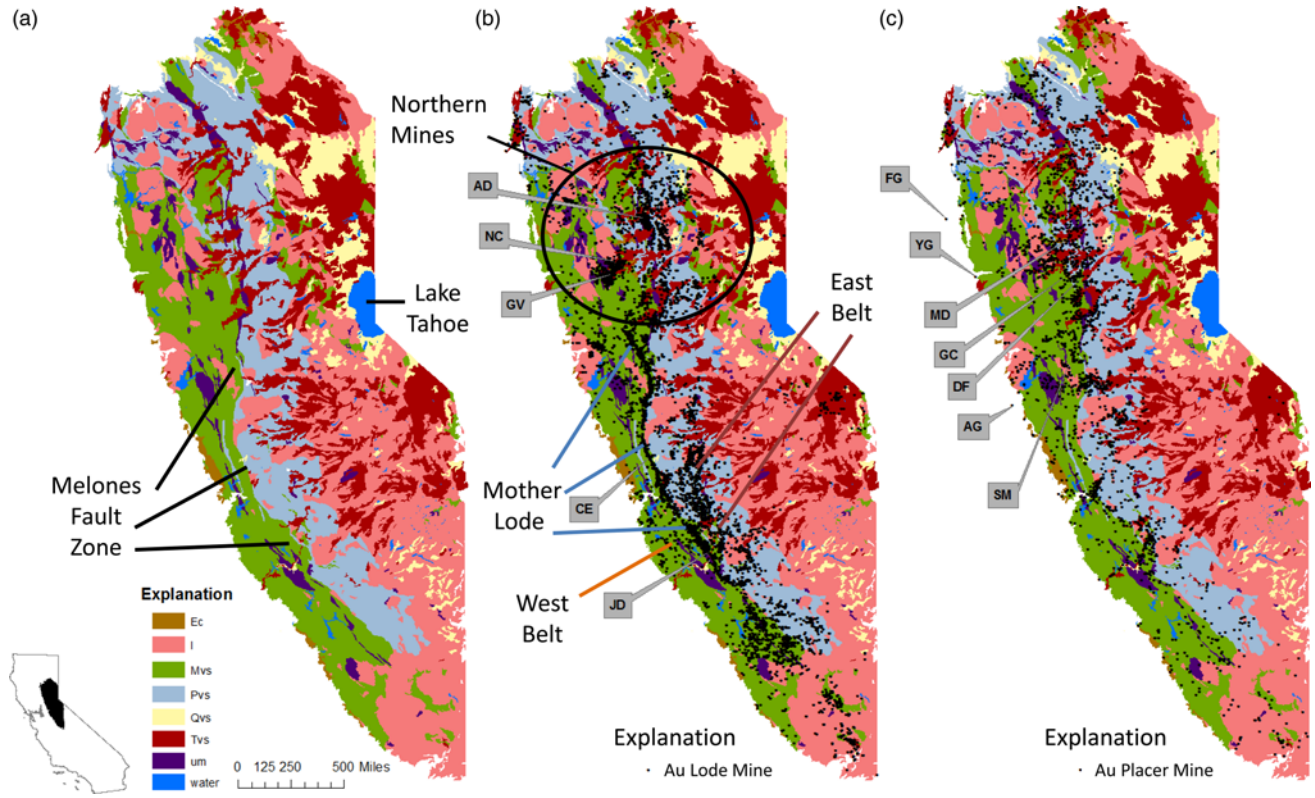

Fig. 1. Distribution of lode and placer gold mines in the Sierra Nevada. (**a**) Simplified geologic map (modified from Saucedo *et al.* 2000): Ec, Tertiary gravels; I, intrusive rocks (mostly Mesozoic); Mvs, Mesozoic (mostly) metavolcanic and metasedimentary rocks; Pvs, Palaeozoic metavolcanic and metasedimentary rocks; Qvs, Quaternary volcanic and sedimentary rocks and unconsolidated deposits; Tvs, Tertiary volcanic and sedimentary rocks; um, ultramafic rocks; (**b**) Lode gold mines and mining districts of the Sierra Nevada (USGS 2013): AD, Alleghany district; CE, Central Eureka mine; GV, Grass Valley district; JD, Jamestown district; NC, Nevada City district; (**c**) Placer gold mines of the Sierra Nevada (USGS 2013): AG, American River goldfields; DF, Dutch Flat; FG, Feather River goldfields; GC, Greenhorn Creek; MD, Malakoff Diggins; SM, Sutter's Mill/Coloma; YG, Yuba River Goldfields.

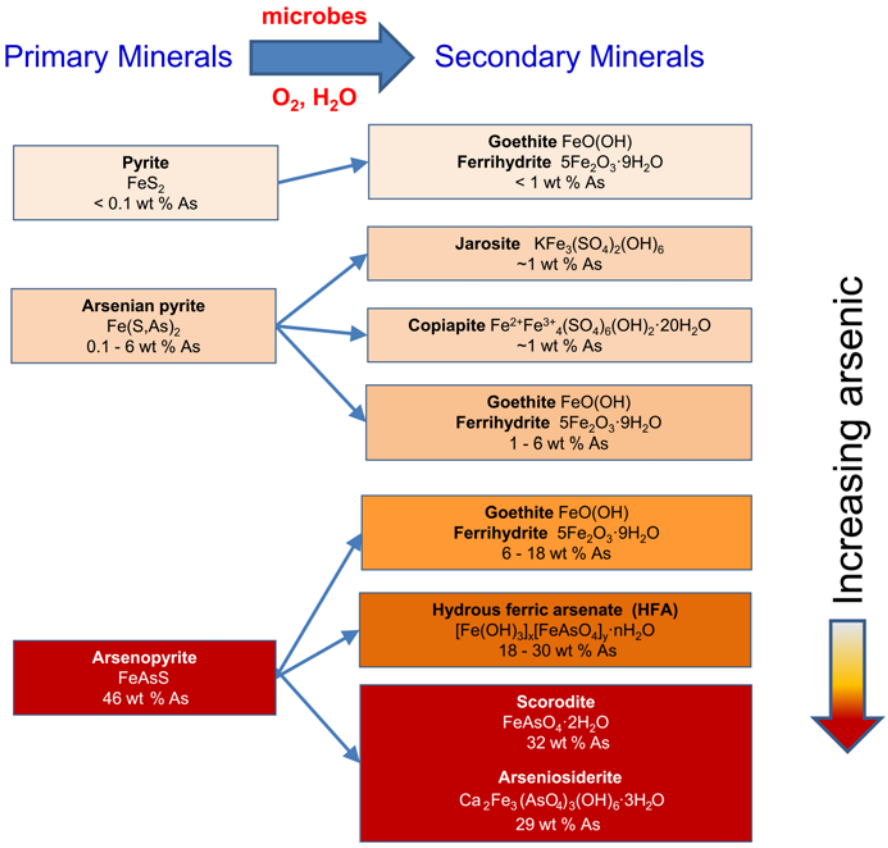

Fig. 2. Primary and secondary arsenic minerals (based on Alpers *et al.* 2014*a*).

spurred by an increase of the Au price. During World War II, most Au production was halted by Limitation Order L-208 by the War Production Board in 1942 (Clark 1970). California Au production fell during the 1950s and early 1960s because of increased costs and the fixed Au price (Clark 1970; Craig & Rimstidt 1998).

Arsenic Associated with Gold Mineralization and Mine Waste

There is a wide range of As concentration in Au ore, wall rock, and mine waste in the SNFH Au province, from less than 10 to more than 10 000 mg kg^{-1} (Ashley 2002). In the northern Sierra Nevada (e.g. Alleghany district, Fig. 1b) As is typically higher in Au ore and waste rock than in the southern part of the region (e.g. Jamestown district, Fig. 1b). Arsenopyrite is the main host of As in northern, unweathered deposits (e.g. Lindgren 1896; Ferguson & Gannett 1932), with minor arsenian pyrite (e.g. Burlak 2012), whereas As is hosted primarily by arsenian pyrite with rare arsenopyrite in the southern deposits, such as the Jamestown district and the Clio mine (Savage *et al.* 2000*a*, *b*). The concentration of As in arsenian pyrite is typically 0.1 – 2 wt% and has been observed up to 6 wt% (Savage *et al.* 2000*a*; Alpers *et al.* 2014*a*).

Recent studies of As contamination at several mining districts in the SNFH orogenic Au province have focused on the mineralogy, geochemistry, and bioavailability of secondary As minerals formed during weathering, as reviewed by Alpers *et al.* (2014*a*). Common weathering products of arsenopyrite include scorodite ($FeAsO_4$•$2H_2O$) and hydrous ferric arsenate (HFA), as shown in Figure 2. The composition of HFA is intermediate between scorodite and ferrihydrite ($5Fe_2O_3$•$9H_2O$) and likely represents a fine-grained, amorphous or nanocrystalline mixture of those two compositions (Paktunc *et al.* 2008; Paktunc 2015). The HFA material has a molar Fe:As ratio less than 3.0 (Walker *et al.* 2009), which corresponds to an As concentration greater than 18 wt%. Common weathering products of arsenian pyrite include ferrihydrite, goethite (FeO(OH)), and jarosite ($KFe_3(SO_4)_2(OH)_6$). Ferrihydrite, goethite, and jarosite formed from weathering of arsenian pyrite may contain As in concentrations up to 18 wt% (Dutrizac and Jambor 2000; Stoffregen *et al.* 2000; Savage *et al.* 2000*a*, *b*; Alpers *et al.* 2014*a*).

The *in vitro* bioaccessibility and *in vivo* bioavailability of secondary As phases as well as the mineralogy and speciation of As in weathered mine waste are areas of active research (e.g. Basta & Juhasz 2014; Foster & Kim 2014; and references therein). Results to date have shown that Ca-Fe-arsenate minerals such as arseniosiderite ($Ca_3Fe_4(AsO_4)_4(OH)_6$•$3H_2O$) and yukonite (nominally $Ca_2Fe_3(AsO_4)_3(OH)_4$•$4H_2O$) are significantly more bioaccessible (and presumably more bioavailable) than other As-bearing minerals in the human digestive tract (e.g. Meunier *et al.* 2010). In weathered samples of Au mine waste, Ca-Fe-arsenates may represent a small fraction of the total As present, but a large fraction of the bioaccessible or bioavailable As. Therefore, characterization methods with fine spatial resolution such as synchrotron-based X-ray Absorption Spectroscopy (XAS; e.g. Foster & Kim 2014), electron microprobe analysis (EMPA), scanning electron microscopy (SEM, including QEMSCAN®; e.g. Burlak 2012; Alpers *et al.* 2014*a*), or transmission electron microscopy (e.g. Walker *et al.* 2009) are needed to detect and quantify the presence of these rare but highly bioaccessible phases.

Arsenic Case Studies

The inactive Harvard mine pit in the Jamestown district (Fig. 1) has developed into a monomictic lake with As concentrations up to 1.2 mg L^{-1} (Savage *et al.* 2002*a*, *b*, 2009). Arsenic-bearing secondary minerals include goethite, jarosite, copiapite group minerals, weilite ($CaHAsO_4$), and Mg-sulphates (Savage *et al.* 2009). At the Eagle-Shawmut and Clio mines, located a few km south of Jamestown, mill tailings are submerged by Don Pedro Reservoir. Secondary iron minerals in As-rich mine waste at the Eagle-Shawmut and Clio mines include goethite and jarosite (Savage *et al.* 2000*a*; Ashley & Savage 2001). The Mesa de Oro mine site, near the Central Eureka mine (Fig. 1b) contained mill tailings with elevated As (Salocks *et al.* 1996) that was relatively high in bioavailability (Golub *et al.* 1999).

At the inactive Argonaut mine, also located near the Central Eureka mine (Fig. 1b), arsenopyrite and its weathering products were characterized by Foster *et al.* (1998). A preliminary assessment of the tailings indicated elevated concentrations of As, Pb, and Hg (USEPA 1998*a*). More recent work has focused on assessment of specific areas for possible remediation (Ecology & Environment 2014, and references therein). Areas with discarded sulphide concentrates were considered high priority for remediation because of low pH, lack of carbonate minerals with pH-buffering capacity, and elevated metal concentrations, including As.

Within the Empire Mine State Historic Park (EMSHP), in the Grass Valley mining district (Fig. 1b), several accumulations of As-bearing waste rock and mill tailings have been characterized recently (Alpers *et al.* 2014*a*, and references therein). The Red Dirt Pile, a remnant sulphide-rich stockpile of mill tailings or low-grade Au ore, had As concentrations in soil as high as 6000 mg kg^{-1} (CVRWQCB 2012). Thirteen waste-rock piles representing the largest of 138 waste dumps within the EMSHP (Selverston & Hilton 2013) had As concentrations ranging from 10.1 to 15 300 mg kg^{-1} (MFG 2009). Three of the waste-rock piles had mean arsenic concentrations >6000 mg kg^{-1}. The As concentration in arsenian pyrite in waste-rock samples from EMSHP ranged from <0.04 to 5.1 wt%, with a median value of 0.51 wt% based on more than 500 EMPA observations (Burlak 2012). Secondary oxide and arsenate minerals found in mine waste at EMSHP are summarized in Figure 2 with regard to their primary source sulphide minerals. Bulk and microbeam chemical, mineralogical, and XAS (As and Fe K-edge) datasets for waste rock and soils from the Empire Mine (Foster *et al.* 2014) were used in combination with *in vitro* bioaccessibility data (Basta *et al.* 2014) and *in vivo* (juvenile swine) bioavailability data (Casteel & Naught 2011) to determine correlative relationships (Foster *et al.* 2014). The abundance of arsenopyrite and/or arsenian pyrite (determined by x-ray diffraction, QEMSCAN® and bulk XAS) were negatively correlated with *in vitro* bioaccessibility and *in vivo* bioavailability. Positive correlations with *in vitro* bioaccessibility and *in vivo* bioavailability data were found with the relative abundances of As(V)-ferrihydrite (by bulk XAS), Fe (hydr)oxides (by bulk XRD and Fe XAS), and As(V and III) associated with Al oxyhydroxide, gibbsite, or kaolinite (by bulk As XAS), and with the As concentration of ferrihydrite (by EMPA). The quantity of ferrihydrite and/or the As concentration in ferrihydrite are two parameters which correlated strongly with *in vitro* bioaccessibility and/or *in vivo* bioavailability in Empire Mine waste-rock samples and which can be measured for relatively low cost (Foster *et al.* 2014).

At the inactive Lava Cap mine, in the Grass Valley district (Fig. 1b), a tailings dam failed in a large rain storm during January 1997. Arsenic concentrations were very high (about 8000 mg kg^{-1} dry wt) in Fe-oxyhydroxide-bearing microbial mats, whereas other mine waste, ore, and contaminated sediments had average As concentrations in the range of 500 – 1500 mg kg^{-1} (Foster & Ashley 2002; Foster *et al.* 2011). Tailings that were exposed after the dam failure became oxidized, producing As-bearing Fe-oxyhydroxides and a Ca-Fe arsenate (possibly arseniosiderite; Foster *et al.* 2011).

Arsenic Summary

Arsenic is a naturally occurring contaminant that is closely associated with mineralization in the SNFH Au province. In

general, primary As-bearing minerals (sulphide minerals including arsenian pyrite and arsenopyrite) are relatively low in bioavailability and bioaccessibility, whereas secondary arsenic-bearing minerals have a wide range of bioavailability and bioaccessibilty, so detailed characterization is needed on site-specific basis. Characterization techniques with fine spatial resolution are needed to identify some secondary minerals, such as Ca- and Ca-Fe-bearing phases, that may constitute a small proportion of sample mass but represent a large proportion of bioavailable or bioaccessible As.

Mercury Used and Lost during Gold and Silver Amalgamation

Amalgamation with Hg was the primary method of Au and Ag recovery during the 1800s and early 1900s at both lode (hardrock) and placer mines. At some of the larger lode mines, chlorination plants came into operation during the 1880s, especially for treatment of sulphide-rich ores (Ashley 2002). Widespread use of cyanide to recover Au and Ag at lode mills began during the 1890s and early 1900s. At many lode mining operations, Hg was used as a primary method of Au and Ag recovery at stamp mills and then cyanide was applied as a secondary recovery method (Churchill 2000; Ashley 2002). The use of Hg continued at placer Au mines into the mid-1900s, primarily at large-scale dredging operations, which persisted into the 1960s, such as the American River, Feather River, and Yuba River goldfields (Fig. 1c). For the period 1848 through the mid-1970s, Churchill (2000) estimated that a total of 5.8 million kg of Hg were lost in California Au processing, broken down as follows: 4.5 million kg lost in placer Au mining and mineral processing, and 1.3 million kg lost in lode Au milling.

The Hg used for amalgamation at Au and Ag mines in the Sierra Nevada was produced by retorting cinnabar (HgS) ore at Hg mines in the California Coast Range. Total California Hg production during the period 1850 – 1970 was about 100 million kg, and an additional 30 million kg of Hg were lost to the atmosphere during retorting at the Hg mines (Churchill 2000). Analysis of Hg in dated ice cores from glaciers in the Wind River Range, Wyoming revealed a distinct increase starting around 1850, attributed to Au and Hg mining activities in California (Schuster *et al.* 2002). Atmospheric Hg losses from the California Coast Range and perhaps also from retorting of amalgam in the Sierra Nevada foothills likely caused contamination of soils in the Sierra Nevada upstream of mine sites, as evidenced by enrichment of Hg in post-1850 sediments deposited in Lake Tahoe (Heyvaert *et al.* 2000). Lake Tahoe is located to the east (typically downwind) and at higher elevation than the Au mines in the SNFH province (Fig. 1a) and the Hg mines on the west side of the Sacramento Valley.

Mercury Contamination in Sediment and Biota

The first large-scale study of MeHg bioaccumulation associated with historical Au mining in the Sierra Nevada was conducted during 1993 – 1995 by scientists at the University of California, Davis (UCD; Slotton *et al.* 1997), who sampled several taxa of invertebrates as well as rainbow and brown trout, where available, at 57 locations in the northwestern Sierra Nevada. Results indicated locally elevated MeHg bioaccumulation, especially in the Cosumnes, Bear, and Yuba River watersheds, all locations of significant historical Au mining activity.

In 1998, the U.S. Geological Survey (USGS) began investigating Hg contamination in water and sediment at an inactive placer Au mine in the Dutch Flat mining district (Fig. 1c) within the Bear River watershed. Visible elemental Hg was recovered by panning sediments in abandoned sluice tunnels such as the Polar Star tunnel (Hunerlach *et al.* 1999), a site subsequently remediated by the U.S. Environmental Protection Agency (USEPA), as described below.

The USGS then began in 1999 a larger-scale study of Hg and MeHg contamination related to historical Au mining in the northern Sierra Nevada and Trinity Mountains, in cooperation with the Bureau of Land Management (BLM) and the U.S. Forest Service (USFS). The Sierra Nevada portion of the study focused on the Bear River, Deer Creek, and South Yuba watersheds, which had among the highest MeHg bioaccumulation (Slotton *et al.* 1997) and the highest intensity of past placer Au mining (Alpers & Hunerlach 2000).

Greenhorn Creek (Fig. 1c) is a tributary to the Bear River with an especially high density of historical placer Au mining activity, including the Buckeye, Sailor Flat, and Red Dog mining districts. Results from USGS sampling during 1999 – 2001 indicated several locations with elevated Hg and/or MeHg in water and sediment, and some areas with elevated MeHg in predatory invertebrates (Alpers *et al.* 2005*b*). Additional sampling during 2002 – 04 in the Greenhorn Creek drainage and other watersheds of the Sierra Nevada, as well as the Trinity Mountains of northern California, was done by USGS for the purpose of identifying 'mercury hot spots' as potential remediation targets for the BLM and USFS. Some of these 'mercury hot spot' sites have since been remediated, as described below.

The distribution of total Hg (THg) and MeHg in USGS sediment samples from historical Au mine sites and downstream areas in the Sierra Nevada is shown in Figure 3. The field panning method developed by Hunerlach *et al.* (1999) for samples with visible liquid Hg uses about one kilogram of sediment, with a detection limit of 100 mg kg^{-1} (equal to 100 parts per million, ppm). The median Hg concentration by the panning method was 1000 ppm for 31 observations in the Sierra Nevada, and the maximum observed Hg concentration was 45 000 ppm (4.5 wt%). Samples without visible Hg, analyzed by laboratory methods ($n = 114$), ranged in concentration from 0.003 to 41 ppm, with a median value of 0.15 ppm.

For comparison, a national USGS study of Hg and MeHg in stream environments (Scudder *et al.* 2009) found median total Hg concentrations of 0.049 ppm in mined basins ($n = 86$) and 0.031 ppm in unmined basins ($n = 259$). The higher concentrations in the Sierra Nevada samples compared with the national data set may be due in part to the fact that several of the Sierra Nevada samples were taken directly from mining features such as ground sluices and sluice tunnels rather than streams or rivers. A consensus Threshold Effects Concentration (TEC) for total Hg is 0.18 ppm (MacDonald *et al.* 2000). Forty-nine of 114 (43%) of the USGS mining-affected sediment samples from the Sierra Nevada exceed this TEC value for total Hg.

Methylmercury concentrations in 107 Sierra Nevada sediment samples from Au mine sites and downstream areas ranged from 0.00001 to 0.07 ppm (0.01 – 70 parts per billion, ppb) with a median value of 0.00047 ppm (0.47 ppb) (Fig. 3). The median MeHg concentration in sediment was very similar to the median values found in the USGS national study: 0.52 ppb in mined basins and 0.51 ppb in unmined basins (Scudder *et al.* 2009). The ratio MeHg/THg, a quantity referred to as Hg methylation efficiency (e.g. Krabbenhoft *et al.* 1999), ranged from 0.0005 to 29% in the Sierra Nevada samples with a median of 0.39% ($n = 104$). For comparison, the USGS national study found median MeHg/THg values in stream sediment of 1.27% in mined basins and 1.72% in unmined basins (Scudder *et al.* 2009).

Following the work by Slotton *et al.* (1997), several studies have investigated Hg bioaccumulation in sport fish in Sierra Nevada streams, lakes and reservoirs. Fish from five reservoirs and several streams in the Bear River and Yuba River watersheds had locally elevated Hg (May *et al.* 2000), which led to the first fish-consumption advisories for Hg in the Sierra Nevada (Klasing & Brodberg 2003). A collaborative effort by USGS and UCD at Lake

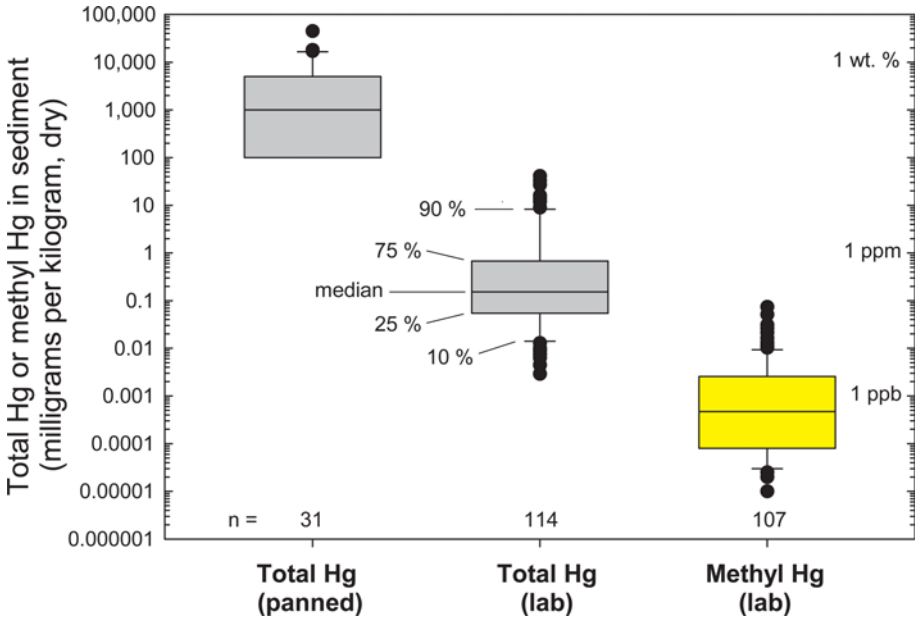

Fig. 3. Boxplots showing concentrations of total Hg and methyl Hg in sediment from Sierra Nevada gold mines and downstream environments. Data from Alpers *et al.* (2005*b*) and USGS National Water Information System http://waterdata.usgs.gov/nwis; Hg, mercury; n, number of samples; ppm, part per million; ppb, part per billion.

Natoma, a reservoir on the American River downstream of Sutter's Mill (Fig. 1c), documented elevated Hg in sport fish, including channel catfish with an average Hg concentration of 1.5 ppm wet weight (Saiki *et al.* 2004, 2005). This led to a site-specific fish consumption advisory for Lake Natoma (Klasing & Brodberg 2004).

A detailed USGS water-quality and food web study in Camp Far West Reservoir, in the Bear River watershed downstream of Greenhorn Creek (Fig. 1c) and the Grass Valley mining district (Fig. 1b), led to improved understanding of seasonal variations in Hg and MeHg concentrations in water, phytoplankton, and zooplankton and their effects on biomagnification to higher tropic levels (Alpers *et al.* 2008; Stewart *et al.* 2008; Saiki *et al.* 2009). Stable isotopes of nitrogen ($\delta^{15}N$, representing $^{15}N/^{14}N$) were used as a proxy for trophic level. Spotted bass, the top predator in the pelagic food web, had a mean mercury concentration of 0.93 µg/g (wet) (Saiki *et al.* 2009). An updated fish-consumption advisory (OEHHA (Office of Environmental Health Hazard Assessment) 2009), recommending no human consumption of spotted bass, incorporated data from the USGS study.

Other detailed studies of Hg in sediment in the Yuba River watershed were done by USGS at Daguerre Point Dam (Hunerlach *et al.* 2004) and Englebright Lake (Alpers *et al.* 2006). The Daguerre Point Dam study characterized sediment trapped behind a small (8-m high) dam on the lower Yuba River, adjacent to the Yuba Goldfields (Fig. 1c). Within Englebright Lake, silty bed sediment from shallow cores had total Hg concentrations mostly in the range of 200 – 300 ppb (dry wt). Deep cores showed more variation in grain-size distribution and Hg content. Highest median values for total Hg were found in deep drill holes near the dam (about 350 ppb); lowest median concentrations of total Hg were found in silty sand material from the three most upstream sites (about 20 – 100 ppb), and intermediate median values were found at mid-reservoir sites with intermediate grain size distribution (135 and 245 ppb). In general, sand from the Yuba River has low total Hg concentration, about 10 ppb, whereas silt and clay in areas affected by historical mining has more elevated Hg concentration in the range of 200 – 300 ppb (Hunerlach *et al.* 2004; Alpers *et al.* 2006).

During 2011 – 2012, the USGS and UCD sampled water, sediment, and sport fish, mostly rainbow and brown trout, at 24 river sites in the northern Sierra Nevada (Alpers *et al.* 2014*b*, c). The Hg and MeHg data were combined with historical data on Hg and MeHg in fish and sediment, and available spatial data on land use/land cover and the distribution of historical Au mines, to develop a predictive model for Hg in five species of sport fish in Sierra Nevada streams (Alpers *et al.* 2016). A Mining Influence Factor (MIF) was computed based on the influence of historical hydraulic mine-pit area and overall gold mine density on fish THg concentrations. Plots of THg and MeHg in sediment as a function of organic content (loss on ignition) and grain size (percent less than 0.063 mm; Fig. 4) show the strong influence of historical mining on mercury distribution.

Mercury Summary

Several publications by USGS since 1999 have documented widespread Hg contamination in the Sierra Nevada associated with historical Au mining, both placer and hardrock. Elevated concentrations of THg in sediment are typically associated with mine features where Hg amalgamation was practiced, such as ground sluices and tunnel sluices at hydraulic mine sites, and stamp mills at hardrock mine sites; streambed sediment THg is higher in mining-affected areas when controlling for grain size and organic content (Fig. 4). Methylation efficiency (as measured by the ratio MeHg/THg) is typically low in mining-affected areas of the the Sierra Nevada, perhaps because of relatively low organic carbon and sparse wetlands. Nevertheless, elevated fish tissue Hg concentrations are associated with mining-affected areas (Alpers *et al.* 2016), and waterbodies in these areas typically have fish consumption advisories for mercury (e.g. Klasing & Brodberg 2003, 2004; OEHHA 2009).

Remediation Efforts in the Sierra Nevada

Several historical Au mining areas in the Sierra Nevada have been remediated in recent years to mitigate problems with either As or Hg in mine wastes and related contamination.

Mine Sites with Arsenic as Primary Contaminant of Concern

(1) Mesa de Oro, a residential subdivision in the town of Sutter Creek near the Central Eureka mine (Fig. 1) in Amador County, became well known in the mid-1990s (e.g. Greenwald 1995) because of As contamination associated with hardrock mill tailings from a lode Au mine.

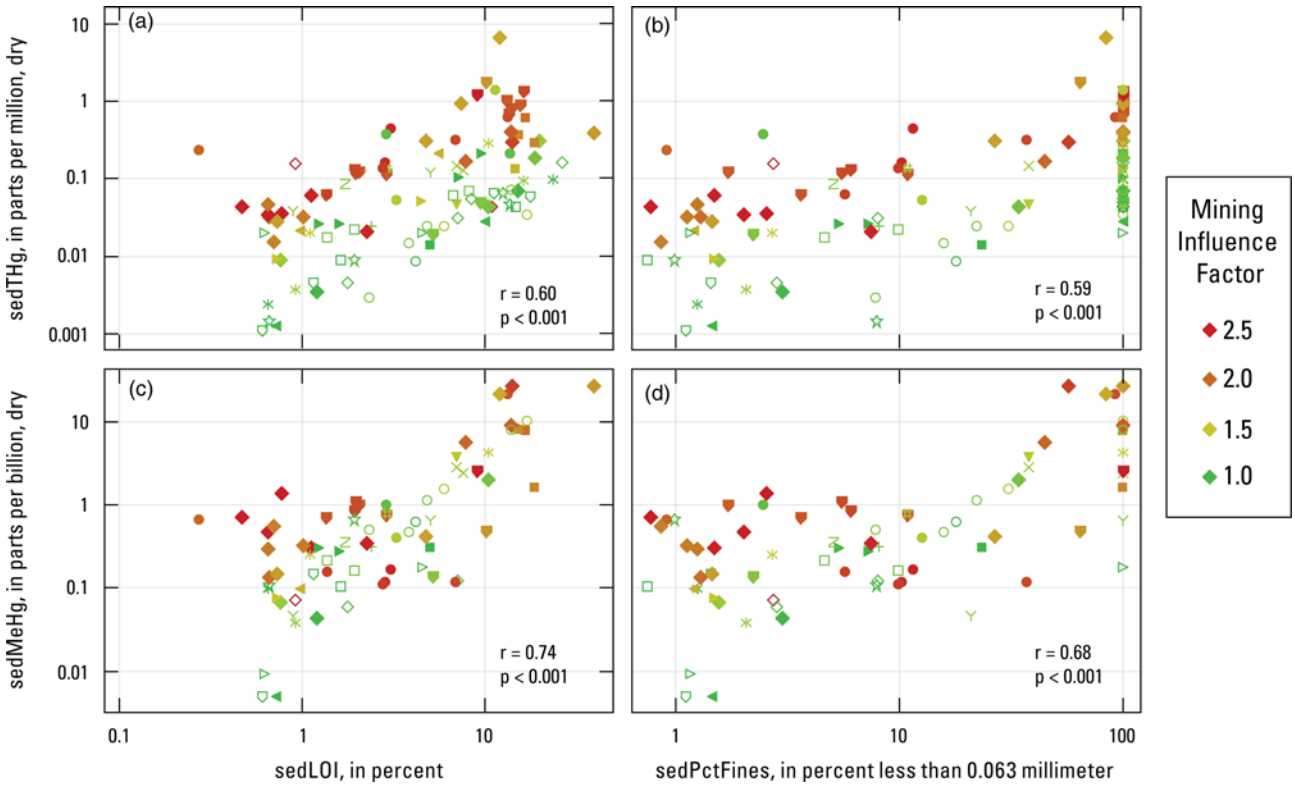

Fig. 4. Scatter plots showing concentrations of sediment total Hg (sedTHg) and sediment methyl Hg (sedMeHg) from Sierra Nevada streams as a function of organic content measured as Loss on Ignition (sedLOI), and percent fines (sedPctFines). Mining Influence Factor based on relative upstream area of historical hydraulic mine pits and gold mine density; r, Spearman rank correlation coefficient; p values indicate significant correlations. Reproduced from Alpers *et al.* (2016) with permission from Elsevier, Ltd.

Remediation comprised removal of contaminated soil and tailings and placement of clean fill (USEPA 1998*b*; DTSC 2005).

(2) Argonaut mine, also located in Amador County, has an extensive deposit of mill tailings that are impounded by a concrete multiple-arch dam built in 1916. Structural analysis of the dam by the U.S. Army Corps of Engineers (Abela & Pattermann 2015) determined that the dam does not meet minimum engineering requirements, and options to repair or retrofit the dam are being considered, along with possible removal actions in nearby contaminated areas (USEPA 2013). The site was recently added to the National Priorities List (NPL) by the USEPA under the Comprehensive Environmental Response, Compensation, and Liability Act (CERCLA, or Superfund) (USEPA 2016).

(3) The Lava Cap mine was placed on the NPL by the USEPA under CERCLA in 1999. Work at the site is divided into four operable units (OUs): the Mine Area Operable Unit (OU1); the Groundwater Operable Unit (OU2); the Lost Lake Operable Unit (OU3); and the Mine Area Residence Operable Unit (OU4) (USEPA 2006). Construction and removal of contaminated soils were done for OU1 and OU4 prior to 2011 (USEPA 2011). A 1.5-mile, 8-inch diameter pipeline is being built to bring drinking water to residents whose wells were contaminated with arsenic, as part of OU2 (USEPA 2012).

(4) At La Trinidad mine, As-rich mill tailings were removed by helicopter at this remote site in the Tahoe National Forest (R. Weaver, U.S. Forest Service, written comm., 2014).

(5) Contaminated areas within Empire Mine Historic State Park have been subdivided into 11 OUs (DTSC 2015). Work has been completed on removal of contaminated sediment from the Red Dirt Pile, and remediation of trails in the Osborne Hill area (DTSC 2006, 2008, 2015). A passive water treatment system using engineered wetlands was constructed in 2011 to treat discharge from the Magenta Drain tunnel (Gusek *et al.* 2011); during the period October 2012 through December 2013, the system typically met final effluent limits for As, Fe and manganese (Alpers *et al.* 2014*a*).

Mine Sites with Mercury as Primary Contaminant of Concern

(1) The Polar Star mine tunnel, in the Dutch Flat placer mining district (Fig. 1c) was remediated by removing Hg-contaminated sediment and lining the tunnel with concrete (USEPA 2000, 2015*a*, *b*, *c*).

(2) The Sailor Flat placer Au mine, located in the Greenhorn Creek drainage (Fig. 1c), was identified as a 'mercury hot spot' based on USGS sampling of water, sediment, and biota during 1999 – 2001 (Alpers *et al.* 2005*b*). The site was remediated during 2003 by removing a shallow sluice tunnel and restoring the hillside to a natural grade (DeGraff 2007).

(3) Alpha Diggins is an abandoned hydraulic mine located in the South Yuba River watershed between Malakoff Diggins and Greenhorn Creek (Fig. 1c). Based on results of initial sampling by the USGS in 1999 – 2001, follow-up sampling was done during 2003 (SAIC 2004) and 2004 – 05 (Tetra Tech 2006). Remediation during 2007 – 08 was focused on erosion control and revegetation (R. Weaver, U.S. Forest Service, written commun. 2014).

(4) The Boston mine sluice tunnel, in the Greenhorn Creek drainage (Fig. 1c) was identified as a 'mercury hot spot' by USGS sampling during 1999 – 2001 (Alpers *et al.* 2005*b*). The site was partially remediated by the BLM during 2005 (DOI (Department of the Interior) 2006). Mercury-

contaminated sediment was removed from the downstream part of the tunnel, but no remediation on the upstream part was performed because of its obstruction by a bulkhead.

(5) At Malakoff Diggins State Historic Park (Fig. 1c), an assessment is underway of remediation options to mitigate problems with Hg and suspended sediment discharge from the historical hydraulic mine pit (Monohan 2015). Ongoing work by USGS includes a ground-based LiDAR time-series to determine erosion rates at selected locations within the Malakoff Diggins hydraulic mine pit, using an approach similar to that used on an eroding riverbank elsewhere in the Yuba River watershed (Howle *et al.* 2016).

(6) At the Relief Hill hydraulic mine site in the South Yuba River watershed, five sluice tunnels were identified and sampled for Hg contamination (SAIC 2004; Tetra Tech 2006). Remediation was performed at the site during 2015 – 16 by the USFS (R. Weaver., USFS, written commun., 2016).

Summary of Remediation Efforts

Several mine sites in the Sierra Nevada have been remediated since the late 1990s because of contamination with As or Hg. In most cases, waste material has been excavated and either removed from the site or stabilized on site in a landfill or repository. Reducing As contamination at these sites has reduced exposure to human receptors, which is particularly important in areas accessible to the public, such as trails in state parks. The intended benefit from reducing Hg contamination at mine sites is typically reduction of Hg bioaccumulation in fish tissue, which represents a risk to human and ecological consumers. Future monitoring efforts are needed to determine if such reductions can be measured.

Concluding Remarks

Arsenic and mercury are the primary contaminants of concern at many historical Au mines in the Sierra Nevada, California. Characterization studies of As-bearing mine wastes at several lode Au mine sites in the Sierra Nevada have focused on total As concentration as well as mineralogy, speciation, *in situ* bioavailability and *in vitro* bioaccessibility. Weathering products including Fe-oxyhydroxides, Fe-arsenates and Ca-Fe-arsenates typically have higher bioavailability and bioaccessibilty than the primary As-bearing sulphide minerals arsenopyrite and arsenian pyrite.

Mercury contamination associated with historical placer Au mining has been documented and remediated at several locations in the Sierra Nevada. Typically the most Hg-contaminated areas are ground sluices and tunnel sluices where Au amalgamation was practiced during active hydraulic mining operations starting in the 1850's and continuing in the early 1900s. Mercury methylation efficiency (the ratio of MeHg to total Hg) in Hg-contaminated sediments is typically less than 1%; nevertheless there is extensive bioaccumulation of MeHg in invertebrates and fish in the Sierra Nevada region downstream of historical Au mining areas. Fish consumption is the main pathway of concern for human exposure to MeHg. Fish-consumption advisories related to Hg contamination have been issued by the state of California (e.g. OEHHA 2009) for several reservoirs and streams affected by historical Au mining in the Sierra Nevada. Additional monitoring efforts are needed to determine if remediation of Hg-contaminated mine sites has resulted in measureable reductions in Hg bioaccumulation.

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