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Mercury Geochemistry of Gold Placer Tailings, Sediments, Bedrock, and Waters in the Lower Clear Creek Area, Shasta County, California–Report of Investigations, 2001-2003

By Roger P. Ashley and James J. Rytuba

Open-file Report 2008-1122

Cover photo description:
Wetlands ponds constructed on an alluvial terrace previously mined for placer gold, Old Mill area, lower Clear Creek. R.P. Ashley, January 2003.

U.S. Department of the Interior
DIRK KEMPTHORNE, Secretary

U.S. Geological Survey
Mark D. Myers, Director

U.S. Geological Survey, Reston, Virginia 2008

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Conversion Factors

Inch/Pound to SI

Multiply	By	To obtain
Length		
inch (in.)	2.54	centimeter (cm)
inch (in.)	25.4	millimeter (mm)
foot (ft)	0.3048	meter (m)
mile (mi)	1.609	kilometer (km)
Area		
acre	4,047	square meter (m^2)
acre	0.4047	hectare (ha)
acre	0.4047	square hectometer (hm^2)
acre	0.004047	square kilometer (km^2)
section (640 acres or 1 square mile)	259.0	square hectometer (hm^2)
square mile (mi^2)	259.0	hectare (ha)
square mile (mi^2)	2.590	square kilometer (km^2)
Volume		
ounce, fluid (fl. oz)	0.02957	liter (L)
pint (pt)	0.4732	liter (L)
quart (qt)	0.9464	liter (L)
gallon (gal)	3.785	liter (L)
gallon (gal)	0.003785	cubic meter (m^3)
gallon (gal)	3.785	cubic decimeter (dm^3)
cubic foot (ft^3)	28.32	cubic decimeter (dm^3)
cubic foot (ft^3)	0.02832	cubic meter (m^3)
Flow rate		
cubic foot per second (ft^3/s)	0.02832	cubic meter per second (m^3/s)
Mass		
ounce, avoirdupois (oz)	28.35	gram (g)
pound, avoirdupois (lb)	0.4536	kilogram (kg)
ton, short (2,000 lb)	0.9072	megagram (Mg)

SI to Inch/Pound

Multiply	By	To obtain
Length		
centimeter (cm)	0.3937	inch (in.)
millimeter (mm)	0.03937	inch (in.)
meter (m)	3.281	foot (ft)
kilometer (km)	0.6214	mile (mi)
meter (m)	1.094	yard (yd)
Area		
square meter (m^2)	0.0002471	acre
hectare (ha)	2.471	acre
square kilometer (km^2)	247.1	acre
square kilometer (km^2)	0.3861	square mile (mi^2)
Volume		
liter (L)	33.82	ounce, fluid (fl. oz)
liter (L)	2.113	pint (pt)
liter (L)	1.057	quart (qt)
liter (L)	0.2642	gallon (gal)
liter (L)	61.02	cubic inch (in^3)
Flow rate		
cubic meter per second (m^3/s)	35.31	cubic foot per second (ft^3/s)
Mass		
gram (g)	0.03527	ounce, avoirdupois (oz)
kilogram (kg)	2.205	pound avoirdupois (lb)
megagram (Mg)	1.102	ton, short (2,000 lb)

Temperature in degrees Celsius (°C) may be converted to degrees Fahrenheit (°F) as follows:
°F=(1.8×°C)+32
Temperature in degrees Fahrenheit (°F) may be converted to degrees Celsius (°C) as follows:
°C=(°F-32)/1.8
Vertical coordinate information is referenced to the National Geodetic Vertical Datum of 1929.
Horizontal coordinate information is referenced to the 1927 North American Datum (NAD 27)."
Altitude, as used in this report, refers to distance above the vertical datum.
Specific conductance is given in microsiemens per centimeter at 25 degrees Celsius (µS/cm at 25 °C).
Concentrations of chemical constituents in water are given either in milligrams per liter (mg/L), micrograms per liter (µg/L), or nanograms per liter (ng/L).

Mercury Geochemistry of Gold Placer Tailings, Sediments, Bedrock, and Waters in the Lower Clear Creek Area, Shasta County, California–Report of Investigations, 2001-2003

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Abstract

Clear Creek, one of the major tributaries of the upper Sacramento River, drains the eastern Trinity Mountains. Alluvial plain and terrace gravels of lower Clear Creek, at the northwest edge of the Sacramento Valley, contain placer gold that has been mined since the Gold Rush by various methods including hydraulic mining and dredging. In addition, from the 1950s to the 1980s aggregate-mining operations removed gravel from the lower Clear Creek flood plain.

Since Clear Creek is an important stream for salmon production, a habitat restoration program is underway to repair damage from mining and improve conditions for spawning. This program includes moving dredge tailings to increase the area of spawning gravel and to fill gravel pits in the flood plain, raising the concern that mercury lost to these tailings in the gold recovery process may be released and become available to biota. The purposes of our study are to identify sources, transport, and dispersal of mercury in the lower Clear Creek area and identify environments in which bioavailable methylmercury is produced. Analytical data acquired include total mercury and methylmercury concentrations in sediments, tailings, and water.

Mercury concentrations in bedrock and unmined gravels in and around the mined area are low and are taken to represent background concentrations. Bulk mercury values in placer mining tailings range from near-background in coarse dry materials to more than 40 times background in sands and silts exposed to mercury in sluices. Tailings are entrained in flood-plain sediments and active stream sediments; consequently, mercury concentrations in these materials range from background to about two to three times background. Mercury in sediments and tailings is associated with fine size fractions. The source of most of this mercury is historical gold mining in the Clear Creek watershed. Although methylmercury levels are low in most of these tailings and sediments, flood-plain sediment in shallow flood-plain ponds, tailings in a dredge pond, and active stream sediment in a Clear Creek backwater have elevated levels of methylmercury.

Stream waters in the area show low mercury levels during both summer and winter base-flow conditions. During winter high flows total mercury increases by about one order of magnitude; this additional mercury is associated with suspended particulate material. Methylmercury is low in stream waters.

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Ponds in various environments generally have higher total mercury levels in waters than Clear Creek under base-flow conditions and higher methylmercury levels in both sediments and waters. Ponds are probably the main source of bioavailable mercury in the lower Clear Creek area.

Several saline springs occur in the area. The saline waters are enriched in lithium, boron, and mercury, similar to connate waters that are expelled along thrust faults to the south on the west side of the Sacramento Valley. Saline springs may locally contribute some mercury to pond and drainage waters.

Introduction

The Klamath Mountains of northern California and southwestern Oregon have produced significant amounts of gold, both from placer and lode deposits (fig. 1). The most important placer deposits occur along the major rivers, including Clear Creek and the Trinity, Klamath, and Smith Rivers, and their tributaries (Clark, 1970). The placers of lower Clear Creek have been mined intermittently by various methods since the 1850s (Clark, 1970; Averill, 1933), with the result that all the alluvial gravel forming the flood plain of Clear Creek and most of the gravel capping adjacent terraces has been disturbed. In addition, in recent decades gravel has been removed from the lower Clear Creek alluvial system for aggregate.

Through most of the placer mining period, mercury was used for recovery of gold by amalgamation, resulting in contamination of the sediment that was processed. Elevated levels of mercury have been recognized as a water quality problem throughout the Sacramento River basin (Domagalski, 1998; Domagalski and others, 2000a,b). Sources of this mercury include mercury mines in the Coast Ranges and mercury losses from gold mining operations, both placer and lode, in the Klamath Mountains and Sierra Nevada.

Habitat restoration for migratory salmon has been underway in the lower Clear Creek basin since 1998. Mercury contents of the sediments impounded behind Saeltzer Dam, in the western part of the lower Clear Creek area, were examined as part of a floodway rehabilitation project involving removal of the dam in the summer of 2000 (Yahnke, 2001; URS Corporation, 2000). Sediments that have been exposed to mercury have been used for flood plain restoration, and will likely be used in the future. The main purpose of this study is to determine how much mercury has been added to the sediments, how it is distributed within them, and whether unusual amounts of methylmercury are associated with sediments under any conditions. We have looked at mercury and methylmercury concentrations in waters of Clear Creek and various ponds in the lower Clear Creek area to determine whether waters in contact with mercury-bearing sediments have elevated concentrations of mercury, and whether conditions favor methylation in any aquatic environments. We have sampled Clear Creek within and above the restoration area during summer and winter base flow and winter high flow conditions to see how much mercury is being transported, and under what conditions.

Fine-grained sediment that has been exposed to mercury as a result of gold-mining activities is likely the primary source of mercury in aquatic environments in the lower Clear Creek area. However, concentrations of total mercury in sediment are often not closely correlated with mercury levels in aquatic organisms (Beckvar and others, 1996). Mercury concentrations in organisms, especially at higher trophic levels, ultimately provide the best measure of mercury bioavailability in an area.

Therefore a study of mercury levels in biota was carried out during the period of this study (Hothem and others, 2004). Preliminary results of this study, based on some of the data reported here, were presented in Ashley and others (2002).

Mining History

Following the initial discovery of gold in gravels of the Trinity River in 1848, placer deposits were the main source of gold in the Klamath Mountains region until the 1880s (Clark, 1970). Gold was discovered in Clear Creek in 1849. For the first few years, crude methods were used to mine the gravels in the active channel and adjacent flood plain; mercury was not available to use for gold recovery.

In the mid-1850s hydraulic mining was developed. In this mining method, hydraulic monitors were used to excavate off-stream gravels, especially in paleochannels or on benches. Extensive systems of sluices charged with mercury were used to recover the gold. Terrace deposits in the lower Clear Creek area were probably mined locally by hydraulic methods, mainly before 1884, at which time most hydraulic mining in tributaries of the Sacramento and San Joaquin Rivers was shut down by judicial decree (Sawyer Decision). Hydraulic mining was done in the Igo district, located on the divide to the west of the lower Clear Creek area (fig. 1), from the 1860s to the 1880s (Clark, 1970).

From the 1880s until World War I, lode and placer mines produced roughly equal amounts of gold in the Klamath Mountains. Gold production declined in the 1920s but revived in the 1930s as many dredges were put in operation, returning placer deposits to prominence. The placer deposits of lower Clear Creek supported numerous dredging operations (Averill, 1933). Large areas in the lower Clear Creek valley and some tributary drainages were dredged during this period, using both drag-line and bucket-line dredges. Most mining operations were forced to close in 1942 in response to War Production Board Order L-208, and few were able to reopen after World War II.

Bedrock in the area hosts some narrow and shallow gold-quartz veins that yielded small but rich pockets. The most productive of these was the Yankee John mine, located about 3 km north of the center of the study area (fig. 1). The French Gulch district, the largest lode gold district in the Klamath Mountains, lies in the Clear Creek drainage basin north of Whiskeytown, above the main placer-mining areas.

Aggregate mining began in the lower Clear Creek basin in the 1950s. Gravel has been obtained both from in-stream and off-stream pits. Mining in-stream and in the adjacent flood plain ceased in the 1980s, but off-stream mining continues.

Geology

The Klamath Mountains in northern California consist of a series of northwest- to north-trending terranes, or belts of deformed and metamorphosed sedimentary and volcanic rocks ranging in age from Ordovician to Jurassic (fig. 1; Irwin, 1972, 1981, 1985). These belts represent a stack of east-dipping thrust plates; from east to west the belts comprise progressively younger rocks. The thrust-

fault zones that bound the plates contain ultramafic bodies that are mostly serpentinized. Numerous granitic plutons ranging in age from Devonian through Cretaceous intrude the terranes. Groups of plutons of similar age form belts that parallel the trends of their host rocks (Irwin, 1985).

The Clear Creek drainage basin lies entirely within the Eastern Klamath Terrane, the easternmost and oldest of the belts recognized in the Klamath Mountains (fig. 1). The upper part of the basin, in the French Gulch area and northward, is dominated by slates of the Bragdon Formation of Mississippian age. The lower part of the basin is underlain by metavolcanic rocks of Devonian or older age, including the Copley Greenstone and the Balaklala Rhyolite, and granitic rocks including the Mule Mountain stock of Devonian age, and the Shasta Bally batholith of Early Cretaceous age (Strand, 1962; Albers, 1964; Kinkel and others, 1956; Fraticelli and others, 1987).

The oldest rocks in the Great Valley province are nearly unmetamorphosed Late Jurassic and Cretaceous marine sedimentary rocks of the Great Valley sequence, which crop out along the west side of the Sacramento Valley. Lithologies include mudstone, graywacke, conglomerate, and shale. There are exposure areas of both Lower and Upper Cretaceous rocks in the lower Clear Creek area, too small to show on figure 1 (Strand, 1962).

Formations of Tertiary and Quaternary age occupy most of the area of the Great Valley province, including lower Clear Creek. Tertiary rocks in the lower Clear Creek area are included in the Tehama Formation of Pliocene age (Helley and Harwood, 1985). It consists of sandstone and siltstone with lenses of conglomerate derived from the Coast Ranges and Klamath Mountains to the west and north. The Tehama grades eastward into the Tuscan Formation, which consists of volcanic and volcaniclastic rocks erupted and transported from volcanic vents in the Cascades volcanic province to the east.

The Nomlaki Tuff Member of the Tehama Formation is locally exposed in bluffs along Clear Creek and gulches incised into the terrace on the north side of Clear Creek. Some of the best exposures are in drainage cuts and tunnels related to placer mining. The Nomlaki is a dacitic ash-flow tuff, water-reworked in some areas (Helley and Harwood, 1985). In the vicinity of lower Clear Creek it is typically a white or pale-gray, massive, non-welded pumice lapilli tuff. Its stratigraphic position is at or near the base of the Tehama Formation, and it has been dated at 3.4 Ma (Evernden and others, 1964).

The Tehama Formation is overlain unconformably by the Red Bluff Formation of Quaternary age, which forms a thin veneer of red, weathered gravels. Helley and Harwood (1985) interpret the Red Bluff as a sedimentary cover on a widespread pediment surface that formed in the Sacramento Valley between 450 Ka and about 1.08 Ma. It occupies the broad flat divide between lower Clear Creek and Dry Creek to the south, and scattered patches remain on the north side of Clear Creek. It probably covered the terrace on the north side of Clear Creek that was extensively mined by various methods.

The flood plain of Clear Creek, including low terraces adjacent to the active stream channel, is underlain by alluvium of Holocene age. The bulk of this material is probably gravel and sand. As a result of restricted sediment supply in the current hydrologic regime, stream erosion has locally exposed the substrate beneath the gravel. This substrate is described as a hard-pan clay layer (McBain & Trush, 2001). Where we have observed the clayey substrate, however, it is weathered Nomlaki Tuff, or in some cases relatively clay-rich weathered Tehama Formation.

How Gold Mining Practices Introduce Mercury into Tailings and Sediments

Recovery of gold from placer (alluvial) deposits, in which gold is present as free particles, is mainly by gravity methods, which take advantage of the high density of gold (specific gravity of pure gold is 19.3). Miners during the Gold Rush used various devices including the gold pan, rocker, and various types of sluices to recover gold (Averill, 1946; Silva, 1986). Mercury quickly became available from mercury mines in California as gold mining created a demand for it, but the amount used in early small-scale mining of surficial placer deposits is unknown (Churchill, 2000). During the 1850s, as the easily accessible high-grade placer deposits were depleted, mining operations expanded in scale, complexity, and sophistication to exploit larger and lower grade deposits, and the use of mercury increased. By the early 1860s bulk mining by the hydraulic method was common, and provided over half of California gold production between 1853 and 1884 (Clark, 1970; Maurer, 1950; Lindgren, 1911). The hydraulic method used high-pressure water cannons (monitors) to dislodge and move auriferous gravel. The sluice was the most practical device for recovering gold in the large-scale placer operations of the later 19th century, especially in hydraulic mines, which produced huge volumes of slurry (Hunerlach and Alpers, 2003). Large operations employed complex sluice systems that included size classification and treatment tailored to specific particle size ranges. Only the basic operation of sluices is described here.

Sluices are troughs, usually made of wood, set at an angle of a few degrees so that material can be washed through them (McClelland, 1918; Averill, 1946; Silva, 1986). They are always fitted with riffles, which provide settling traps for gold and other heavy minerals. Although there are many styles of riffles, a common type consists of wood strips mounted transversely in the bottom of the trough. Recovery of fine gold can be improved greatly by pouring mercury into the sluice, so that small beads of mercury are lodged behind the riffles (McClelland, 1918; Anonymous, 1963). When fine gold comes in contact with the mercury it forms amalgam, which adheres to the mercury. Periodically the sluice is cleaned up, at which time the amalgam is removed and retorted to recover the gold. Mercury loss into tailings is inevitable and is probably caused by the impact of particles on the mercury beads, and agitation by the flowing water, forming mercury droplets so small that they can be washed away. The tendency of mercury to form very small globules when agitated is referred to as "flouring." Sluices are still in use in small mining operations throughout the world.

The trend toward larger-scale operations to profitably exploit large low-grade placer deposits spawned numerous attempts to develop large-capacity digging machines (dredges) for bulk mining of such deposits (Winston and Janin, 1910). The first successful dredge began operation in 1898 on the lower Feather River, in what later became the Oroville dredge field (Clark, 1970; Winston and Janin, 1910). The largest capacity dredges used buckets on a continuous belt (bucket lines) for digging, delivering the material to a floating wash plant. Smaller operations used cable systems to drag buckets to wash plants either on land or in ponds. Such "drag-line" dredges, which could exploit small deposits, were popular in the 1930s.

Since gold in alluvial deposits amenable to dredging is fine-grained, all dredges employ size-classification equipment, such as screens or rotating trommels, to remove barren coarse material (usually larger than about 3/8 inch or 10 mm) and save finer gold-bearing material for gold recovery (Janin, 1918). Thus only fine materials are directly exposed to mercury. The coarse material is discharged through conveyor systems called stackers. As was the case in other California gold operations employing bulk-mining methods, sluices were commonly used for gold recovery. Some

dredges, however, used tables instead of sluices. These are flat surfaces with riffles installed over more than half their areas, the riffle coverage and arrangement varying with different designs. The table is elevated on one side or corner, where fine-grained material (usually sand size and smaller) and water are introduced. The table is attached to a small motor with an eccentric drive to provide a mechanized shaking motion, usually parallel to the riffles. Low-density particles tend to be washed across the riffles, while high-density particles lodge behind the riffles and work their way to the down-slope end of the riffles. Although tables often recover gold efficiently without the use of mercury and are in use in mercury-free gold recovery systems today, before World War II it was common practice on dredges to add mercury to tables to improve the recovery of fine gold (Janin, 1918). Mercury is used on tables in essentially the same way as it is in sluices.

Sand and silt discharged from sluice boxes or tables on the dredge are usually deposited on the excavated floor of the pond and covered by later stacker tailings as the dredge moves forward. As a result, sand lenses containing more or less silt appear at or near the base of the stacker tailings. Both dredge sluice sands and stacker tailings contain some fine silt and clay, because fine sediment particles settle out of the dredge pond water and are incorporated into materials at the bottom of the pond. Less common are silt-clay layers that likely represent fine material that settled in undisturbed parts of the dredge pond away from the active digging or tailings discharge areas. Dredge pond water may also infiltrate materials around the pond, especially porous stacker tailings, carrying fine silt and clay particles into them.

Recent studies of mercury speciation in tailings and sediments have shown that most of the mercury lost by dispersal of fine particles from sluices is converted to forms other than metallic mercury, including organically bound mercury, mercury adsorbed to mineral particles (mainly iron oxyhydroxides), and mercury sulfide (Slowey and others, 2005; Bloom and others, 2003; Kim and others, 2004). Mechanisms of transformation, and how quickly mercury is transformed after release, are not known.

Mercury was also used in lode gold mine mills. Amalgamation was the main method used for gold recovery in stamp mills before the 1890s, when cyanidation was introduced, and mercury loss rates during that period were substantial, resulting in more than 14 g/g or more in tailings (Churchill, 2000). Mercury use in California quartz mills continued until World War II, but losses were generally much lower because mercury use was limited to high grade ores, or to only one part of the mill circuit. Some smaller mills continued to use amalgamation as their main method of recovery. The French Gulch district, which is the most productive lode gold district in the Klamath Mountains, lies mostly within the Clear Creek drainage basin. On the basis of total production and average ore grade (Albers, 1965), about 2,000,000 tons of gold ore have been milled in the French Gulch district, most of that amount before 1900. Since tailings were not usually impounded in California before about 1910, it is inferred that these tailings were washed away, and contribute mercury to Clear Creek stream sediments.

Sample Sites

We collected samples to characterize mercury species and associated elements in gold placer tailings, bedrock, sediments, and stream and pond waters in the lower Clear Creek area. Samples reported here were collected on several occasions between August 27, 2001 and July 29, 2003, during both summer and winter low stream-flow conditions and higher-flow winter conditions. The winter

sampling event was carried out during quiet weather, but included a short period of moderately high flow produced by a controlled release from Whiskeytown Reservoir. Field sampling sites are clustered in four parts of the area (fig. 2). Site locations are shown on figures 3A-D and the sites and samples are listed and described in table 1. The geographic coordinates in table 1 were obtained in the field using a hand-held Global Positioning System (GPS) unit.

The eastern group of sites (CC1, 2, 3, 4, 5, 6, 7, 25, 26, 30, 36; fig. 3A) provided samples of sediments and waters from the main channel of Clear Creek and three ponds in the floodplain. South Pond (fig. 4) was an in-stream aggregate pit. East Pond (fig. 5) occupies a meander channel that receives overflow from the main channel of Clear Creek during winter high flows. Pond 3 occupied a meander scar possibly modified by aggregate mining. South Pond and Pond 3 were filled in summer 2001 as part of the restoration program. Site CC25 is at China Garden, where a tract of unmined gravel provided background data. Site CC26 is at Restoration Grove, where the flood plain was reconstructed using local dredge tailings and tailings from a borrow area upstream at Reading Bar (CC24, 29; see fig. 3C). The easternmost site (site CC30) provided sediment and water from Clear Creek at the downstream end of the restoration project area.

The group of sites in the center of the area (CC8, 9, 10, 11, 31, 32, 33, 34; fig. 3B) are located on or near an extensive terrace that was covered by auriferous gravels. A lumber mill was formerly located in the northeastern part of this area, and it is therefore sometimes referred to as the "Old Mill" area. The gravels were probably mined using a variety of methods, possibly including drag-line and hydraulic methods. The western edge of the extensive dredge field that covers the lower Clear Creek flood plain and adjacent terraces forms the eastern boundary of this mined area; apparently gravels here were too thin to support a dredging-type operation. Site CC8 provided samples of placer tailings. A system of sluice cuts and a tunnel drained the placer mining operations. Site CC31 is in the sluice cut (a bedrock drainage cut once lined with sluices) above the tunnel, and site CC10 is at the discharge end of the tunnel. One sluice cut is blocked, forming a small seasonal pond (Beaver Pond, site CC34). In addition, seasonal wetlands ponds have recently been constructed on the eastern part of the terrace, using local tailings to construct the impoundment berms. Sites CC32 and 33 provided water and mud from the constructed wetlands ponds. The Nomlaki Tuff forms the bedrock immediately beneath the terrace gravels (CC9). In addition, sediments and water from Clear Creek were sampled (CC11). This Clear Creek site is about 1.5 km downstream from the former Saeltzer Dam site and about 3 km above the flood plain restoration project area.

The large cluster of sites toward the west end of the area (CC12, 13, 14, 15, 16, 17, 20, 21, 22, 23, 24, 27, 28, 29, 35; fig. 3C) comprises mainly dredge tailings. These sites include drag-line dredge stacker piles along Spring Creek (CC14, 20: fig. 6), an intermittent tributary of Clear Creek, and probable bucket-line dredge stacker piles on a Clear Creek terrace, the site of a former rifle range known as the "Shooting Gallery" (CC15, 16, 17, 21, 22, 23). Two backhoe trenches provided samples at the Shooting Gallery (CC15, 16, fig. 7; CC22, 23). Also in the Spring Creek drainage is an impoundment remaining from dredging known as "Red Pond" (CC 12, 35; fig. 8), which receives water from a saline spring at site CC27. Adjacent to Red Pond are exposures of Tehama Formation gravels beneath the Nomlaki Tuff that contain short prospect tunnels (CC13, fig. 9). We sampled Red Pond water and sediment, saline spring water, and unmined gravel from a prospect tunnel. Spring Creek also receives saline water from an artesian well (CC28) and probably other saline springs and seeps in the vicinity. Clear Creek sediment and water were sampled upstream from Spring Creek at Reading Bar (CC 29). Dredge tailings from the restored borrow area at Reading Bar were also sampled (CC24).

The westernmost samples (sites CC18, 19; fig. 3D) are located at the U.S. Geological Survey stream gaging station known as the "Igo Gage." This site provided water and sediment upstream from the mined area. This site is not pristine, however, because considerable lode and placer mining took place upstream, mainly in the French Gulch district (fig. 1; Albers, 1964).

Field Sampling Methods

Dry Sediments and Tailings

At undisturbed sites we dug a shallow pit 10-30 cm deep and removed approximately 5 kg of material from the bottom with a stainless steel trowel. We separated material coarser than 2 mm from material finer than 2 mm (the lower size limit for gravel and the upper size limit for sand in the Wentworth classification) at the site, using a stainless steel wire-mesh sieve. Material was delivered directly to the sieve, which was shaken by hand, and the material smaller than 2mm collected in a plastic gold pan. Each fraction was placed in a pre-cleaned and certified borosilicate glass jar with a Teflon^{TM} seal (I-CHEM^{TM} Series 300), using the stainless steel trowel or a plastic scoop. If the material contained abundant fines that adhered to the plastic pan, a steel pan was used. If the volume of a fraction exceeded one liter, the material was roughly quartered in the pan and opposite quarters transferred to the storage jar.

Channel samples were collected from the walls of trenches and pits, using a stainless steel trowel and a plastic pan to catch the material, which was delivered to the sieve and processed as above to yield +2 mm and –2 mm size fractions.

At each site we retained 1-2 kg of bulk material for both analysis and archive purposes. The bulk material comprised separate grab samples dug from the same holes or cut from the same channels on trench or pit walls.

Equipment was cleaned between sites using a nylon bristle brush. If adhering fines could not be removed satisfactorily with the brush, equipment (especially plastic items that collect dust from static cling) was rinsed with de-ionized water, or in some cases swabbed with paper towels moistened with de-ionized water. Throughout the sampling process, all personnel handling sampling equipment and storage jars wore powder-free latex gloves. Glove sets were dedicated to each site, and disposed of after sampling. Gloves were immediately changed following any accidental contact with metal objects other than those used directly for sampling. Large sampling equipment items were stored and transported in plastic bags, and small items and gloves were stored and transported in Ziploc® bags. The sieves used are dedicated to handling of materials expected to have low levels of mercury and other metals.

We did not attempt to separate any fractions smaller than 2 mm in the field, because dry sieving done by hand is ineffective in separating fines from coarser particles. Because the dredge tailings sampled here have relatively low levels of mercury contamination, we did not consider it necessary to acid-rinse equipment between sites. Equipment previously deployed at other sites, however, was rinsed with 5 percent trace-metal grade HCl and de-ionized water before redeployment in the Clear Creek area.

Wet Sediments and Tailings

Wet sediments and tailings were wet-sieved in the field using stainless steel sieves and 1-2 liters of ambient water. Sieves used at all wet sites included 2 mm (at the gravel/sand break in the Wentworth classification) and 63 m (at the sand/silt break in the Wentworth classification). Sieves with other mesh sizes were employed at some sites, but the size fractions analyzed and reported here are >2 mm, between 2 mm and 63 m, and <63 m. Sediment at and immediately below the sediment-water interface was removed with a stainless steel trowel and delivered directly to stacked sieves and to a jar for a bulk sample. Fines (<63 m) washed through were captured in a stainless steel pan and transferred to a plastic gold pan, to allow washing of the coarser material with several volumes of ambient water. Fractions retained on the sieves (>2 mm and <2 mm>63 m) were delivered to glass jars using the stainless steel trowel. The slurry of fines (<63 m) retained in the gold pan was allowed to settle for approximately 10 minutes, then the suspension was decanted into a storage jar leaving approximately 50 ml of head space. The remaining suspension was discarded, and the settled material in the pan transferred to the jar by washing with ambient water using a polyethylene wash bottle.

Certified Teflon^{TM}-sealed I-CHEM^{TM} glass jars (the same as for dry sediments) were used for subsamples analyzed for metals. Polycarbonate jars (125 or 250 ml capacity) were used for mercury speciation subsamples (usually only <2 mm>63 m and <63 m size fractions). These were frozen with dry ice immediately after collection (freezing time approximately 20-30 minutes) and kept frozen until analysis.

Sampling equipment used and equipment handling protocol were the same as for dry sediment and tailings samples, described above.

The sampling procedure used here allows us to determine metals concentrations in various size fractions, but cannot provide quantitative estimates of amounts of metals residing in the size fractions, owing to unavoidable losses of finer materials. During removal of material at the sediment-water interface, some material escapes into the water column. In quiet water this is mainly silt- and clay-size particles, but in flowing water, some sand-size particles may be lost as well. The sieving, settling, and decanting procedure used for collecting size fractions is designed to produce coarser fractions (sand and gravel size) that are relatively clean with respect to finer particles, but some finer material, especially suspended clay-size particles, is lost. Analyses of bulk material, however, compared with analyses for all size fractions, give some idea of the relative importance of silt- and clay-size material in accounting for the metals content.

Waters

We followed ultra-clean sampling and handling protocols for field sampling of waters for mercury analysis (Bloom, 1995, Gill and Fitzgerald, 1987). The mercury subsamples were grab samples collected in 1-liter borosilicate bottles (I-CHEM^{TM} Series 300, spot-checked for mercury levels by

Frontier Geosciences) with excluding Teflon^{TM} caps (to eliminate head space), and kept chilled to <4°C until analysis. Bottles were triple rinsed with ambient water at each site before filling.

For all other components, we filled a 1-liter HDPE bottle, double rinsed in ambient water; all subsamples for analysis of metals (other than mercury) and anions were removed from this 1-liter bottle. Since these were grab samples, results are not suitable for quantitative estimates of metals concentrations or fluxes. Otherwise water sampling protocols used here, including bottle preparation and sample preservation, generally follow those of Water Resources Discipline of the U.S. Geological Survey for trace metals (Horowitz, 1994). Field filtrations were done with disposable 25 mm-diameter sterile cellulose acetate filters (0.45 m openings) and disposable syringes.

For metal determinations, both filtered and unfiltered subsamples were preserved with Ultrex-grade HNO_3, acidified to pH<2, and stored in HDPE (high-density polyethylene) bottles pre-rinsed with similar trace-metals grade acid. Subsamples for anion determinations were filtered, stored in HDPE bottles, and chilled to <4°C until analysis. Subsamples for alkalinity determinations were treated similarly.

During most sampling events, a field blank was collected by processing de-ionized water and collecting the same subsamples (except for alkalinity) by the same procedures as used for the field samples. Laboratory blanks and acid blanks were analyzed periodically to determine whether our equipment, containers, reagents, and procedures introduced significant contamination.

In 2001, water parameters including pH, conductivity, and temperature were measured in the field with a battery-powered pH meter (Orion Model 290) with low-maintenance sealed gel triode electrode, and a specific conductivity meter (Orion Model 120). The pH triode, which has automatic temperature compensation, was also used for temperature measurements. Dissolved oxygen concentrations were determined with a CHEMets® colorimetric field test kit. In 2002 and 2003, pH, conductivity, temperature, oxidation-reduction potential, and dissolved oxygen concentrations were determined with a Hydrolab Minisonde, Model 4a.

Analytical Methods

Sediments, Tailings, and Rocks

Multi-element analyses for all dry tailings, sediments, and rocks were performed in the laboratories of ALS Chemex. Bulk samples were ground in a zirconia ring mill and subjected to a near-total four-acid digestion. Major elements were determined by inductively coupled plasma-atomic absorption spectroscopy (ICP-AES). Minor elements other than mercury were determined by inductively coupled plasma-mass spectrometry (ICP-MS). Mercury was determined by cold vapor atomic absorption spectroscopy (CVAAS) following methods similar to those described by Crock (1996) and O'Leary and others (1996).

Mercury and methylmercury analyses for all wet sediments were carried out at Frontier Geosciences, Inc. Some dry materials analyzed by Chemex were also analyzed by Frontier. For total mercury, the sediment was leached with cold aqua regia, followed by stannous chloride ($SnCl_2$) reduction, two-

stage gold amalgamation, and cold vapor atomic fluorescence spectroscopy (CVAFS) detection. Methylmercury was obtained by acid bromide/methyl chloride extraction followed by aqueous phase ethylation, isothermal gas chromatographic (GC) separation, and CVAFS detection (Horvat, Bloom, and Liang, 1993). Results were reported on both a wet- and dry-weight basis.

Chemex and Frontier sample preparation steps are shown for comparison in table 2. Chemex preparation involves fine crushing of the entire sample, and utilizes a 250 g split, which is pulverized, for analytical portions. The Chemex 4-acid digestion dissolves all but a few resistant minerals such as zircon and some rare-earth minerals. It is important to note that our samples were treated as rocks rather than soils, and no sieving was performed in the laboratory; all material including clasts as large as 1-2 cm diameter was processed. Frontier's preparation avoids handling and uses a strong leach (aqua regia), which removes elemental mercury, adsorbed mercury, mercury in amalgams, and combined mercury including mercury sulfide, but does not affect silicate minerals. Material coarser than sand size, however, is not included in Frontier's leach.

Waters

Alkalinity as $CaCO_3$ was determined in the field or in the laboratory by titration with H_2SO_4 using Gran's technique (Stumm and Morgan, 1996; Rounds, 2006), within 2-4 days after sample collection. Sulfate, chloride, nitrate, and fluoride concentrations were determined in U.S.G.S. laboratories by ion chromatography (Fishman and Friedman, 1989). Total organic carbon was determined in U.S.G.S. laboratories by wet oxidation to carbon dioxide, which was measured by nondispersive infrared spectrometry (Wershaw and others, 1987). Cations were analyzed by inductively coupled plasma-atomic emission spectrometry (ICP-AES) and inductively coupled plasma-mass spectrometry (ICP-MS). Ion chromatography and laboratory alkalinity analyses were performed in U.S.G.S. laboratories by Peter M. Theodorakos. The ICP-AES analyses were determined in U.S.G.S. laboratories by Paul Briggs. The ICP-MS analyses were determined in U.S.G.S. laboratories by Allen L. Meier. Duplicate water samples, blank samples, and U.S.G.S. Water Resources standard reference waters were analyzed with the data set.

At Frontier Geosciences, samples were handled in a Class-100 clean air station monitored routinely for low levels of total gaseous mercury. An ultra-clean mercury trace metal protocol was followed, including the use of rigorously cleaned and tested Teflon^{TM} equipment and sample bottles and pre-screened and purified reagents. Laboratory atmosphere and water supply are also routinely monitored for low levels of mercury. Primary standards used in the laboratory were NIST-certified or traceable to NIST-certified materials. Monomethylmercury (MMHg) standards were made from pure powder and calibrated against NBS-3133, and cross verified by daily analysis of Certified Reference Material DORM-2 (National Research Council of Canada Institute for National Measurement Standards, 1999).

After filtration under ultra-clean laboratory conditions, both filtered and unfiltered fractions were preserved with 0.4 volume percent hydrochloric acid (HCl). Chloride-rich samples (identified by high specific conductivity) were preserved using 0.2 volume percent sulfuric acid (H_2SO_4) instead of HCl. After analysis for methylmercury, the samples were further preserved with 2.0 volume percent bromine monochloride (BrCl).

EPA Method 1631 was used. Total mercury was determined by BrCl oxidation followed by $SnCl_2$ reduction, two-stage gold amalgamation, and detection by cold vapor atomic fluorescence spectroscopy (CVAFS) (Bloom, Crecelius, and Fitzgerald, 1988). Methylmercury was liberated from water using an all-Teflon^{TM} distillation system. Distilled samples were analyzed using aqueous phase ethylation with purging onto Carbotrap, isothermal gas chromatographic (GC) separation, and CVAFS detection (Bloom, 1989). To address accuracy and precision, quality assurance measures were employed with the following minimum frequency: laboratory duplicates, one per ten samples; method blanks, three per analytical batch; filtration blanks, one per ten samples; and spike recovery or standard reference material, one per ten samples.

Results For Tailings and Sediments

Mercury Concentrations in Background Samples

Figure 10 shows results for materials in the lower Clear Creek area that have not been mineralized or exposed to mercury as a result of human activities; analytical data are shown in table 3. The metamorphic and granitic pebbles categories consist of pebble composites (6-10 pebbles each). Metamorphic rock types dominate the Clear Creek sediments. Granitic rock clasts are common but subordinate to metamorphic types. The pebbles came from the gravel used to fill South Pond, which was from the flood plain in the vicinity of that pond. The Nomlaki Tuff samples came from an exposure in a mine drainage cut near Beaver Pond (site CC9; fig. 3B) and from an exposure at Red Pond (site CC13; fig. 3C). These materials all show relatively low mercury values (<10-30 ng/g). All subsamples of unmined gravel from the Tehama Formation (>2 mm, <2 mm, and bulk; fig. 10) did not have detectable levels of mercury at a 10 ng/g detection threshold, and both coarse and fine subsamples from China Garden had 10 ng/g.

Black sulfide mud deposited around the collar of the well at site CC28 has 67 ng/g of mercury, the highest mercury concentration seen in any naturally-occurring material in the area. The volume of this precipitate, however, is extremely small (probably no more that a few cubic meters), so it is not representative of the geologic units that dominate the area.

The limited data suggest that background mercury values in the area are relatively low, and a background range of <10-30 ng/g is used here for comparison purposes.

Placer Mine Tailings, Old Mill Area

Figure 11 shows mercury determinations for placer mine tailings and materials derived from placer tailings in a tract near the site of a former lumber mill ("Old Mill," Anonymous, 1999; fig. 3B) that was mined by methods other than dredging. The dominant geographic feature is a broad terrace cut on Nomlaki Tuff, covered with alluvium and possibly older gravels of the Red Bluff Formation, which comprised the auriferous deposit (fig. 12). Water was diverted from Clear Creek at Saeltzer Dam and delivered to this mining area through a ditch system. Gold was recovered in sluices, and wastewater and tailings removed through drainage ditches that likely also served as sluice-ways.

Piles of coarse tailings (mainly cobbles) remain on the terrace surface; it was common practice to put only finer-grained material through sluices. Mining and moving material into the sluices was probably done by various methods including small-scale hydraulicking, ground sluicing, and possibly mechanized earth-moving equipment such as drag lines and scrapers. Extensive placer mining was done throughout the lower Clear Creek area, first by hand methods and then by methods such as those used in the Old Mill area. In many places evidence of this early mining has been destroyed by 20th-century dredging, because tailings from earlier mining were reworked. The Old Mill area is bounded on the east side by the large lower Clear Creek dredged tract (see figs. 2 and 3B).

The system of drainage ditches in the Old Mill area was developed at two or more levels. The lowest ditch was cut into Cretaceous shale bedrock and discharges to Clear Creek through a tunnel cut in Nomlaki Tuff (fig. 12). It was probably lined with sluices throughout its length, including the tunnel section. The first three sets of columns in figure 11 represent sediment collected from the discharge portal of this tunnel in 2001 and again in 2002 and 2003 (site CC10). In 2001 only bulk sediment was analyzed, whereas in 2003 bulk, sand size (<2 mm>63 m), and silt-clay size (<63 m) fractions were analyzed, and in 2002, bulk as well as gravel, sand, and silt-clay fractions were analyzed. The results consistently show that mercury concentrations in bulk material are at least four times background, and that most of this mercury is located in the silt-clay fraction, where concentrations are at least 10 times background levels. Water flows in the drainage ditch that feeds into the drainage tunnel only during the winter months. The fourth category in figure 11 (labeled "sluice cut," site CC31) represents sand and silt-clay fractions collected in the ditch just above the tunnel inlet in January 2003. Mercury and gold amalgam were panned from the sediment in the ditch at that time, corroborating the high mercury values found analytically in both size fractions.

As part of the restoration program, berms have been constructed on the terrace, to capture winter run-off and form seasonal wetlands (fig. 3B; cover photo). Local placer mine tailings were used for this purpose. The ponds drain over a rip-rapped spillway into a sump that in turn overflows into the ditch described above. During periods of heavy winter rains, the ponds overflow, carrying fine mud into the sump and at times into the ditch. The next two categories in figure 11 show mercury values for bulk mud from the sump and from one of the constructed ponds. Mercury enrichments are around 10 times background, similar to the fine fraction of sediments in the drainage ditch and tunnel.

On the west side of the terrace is an older seasonal pond, known as the "Beaver Pond," formed where a mining drainage cut is blocked by road fill. As shown in figure 11, the mercury concentration in mud from the Beaver Pond is similar to fine materials elsewhere in the Old Mill mining area. Coarse dry tailings collected from the terrace surface immediately adjacent to the Beaver Pond, however, show a bulk mercury value in the background range and a value for fines (sand-silt-clay) only slightly elevated above background.

On figure 11 and subsequent figures a reference line is shown at a mercury value of 150 ng/g. This is a benchmark value for mercury in sediments used by California Department of Toxic Substances Control, and used in characterization of sediment done prior to removing Saeltzer Dam (URS Corporation, 2000). It is an "effects range-low" (ERL) value, below which adverse effects on aquatic organisms attributable to mercury are highly unlikely, as discussed in a National Oceanic and Atmospheric Administration (NOAA) report addressing mercury as a contaminant in aquatic habitats (Beckvar and others, 1996). The 150 ng/g value was originally proposed by Long and McDonald (1992). This value is close to the threshold effects level (TEL), 130 ng/g, which was used as a screening value for sediment contamination by the U.S. Environmental Protection Agency (USEPA)

in the first National Sediment Quality Survey (U.S. Environmental Protection Agency, 1997). In the second National Sediment Quality Survey (U.S. Environmental Protection Agency, 2004), USEPA used a screening value of 230 ng/g, which was derived from a logistic regression model (Field and others, 1999, 2002). This value (T_{25}) represents the concentration at which the probability of observing a toxic effect in aquatic organisms is 25 percent. A reference line for this value is also shown on figure 11 and subsequent figures. The authors of the NOAA and EPA reports point out that although sediment is the primary source of mercury in most aquatic systems, concentrations of total mercury in sediment may not be useful in predicting the bioavailability of mercury.

Dredge Tailings

Dredging began on lower Clear Creek in 1906 (Winston and Janin, 1910), and dredge tailings represent the most voluminous mining waste in the area. The largest dredging tract comprises about 2000 acres including the flood plain and a broad terrace on the north side of Clear Creek in Sections 26, 27, 28, 33, and 34 (fig. 2). A smaller tract including the Clear Creek floodway and adjacent terraces comprises about 500 acres in Sections 31 and 36. The latter area includes Reading Bar and the Shooting Gallery, a former rifle range, and is adjacent to the site of the former mining community of Horsetown, which can no longer be precisely located. Throughout these tracts the poorly sorted pebbles, cobbles, and occasional boulders typical of stacker tailings are seen; however, undisturbed stacker piles remain at only a few locations, owing to extensive gravel mining and other development that has taken place since dredging ended in World War II. Dredge sluice sands are seen only occasionally in excavations.

The categories in figure 13A represent results for dredge stacker tailings in the Shooting Gallery and Spring Creek areas. The Shooting Gallery tailings are probably from a bucket-line dredge and the Spring Creek tailings from a drag-line dredge. Tailings sampled at these sites are located near the tops of stacker piles; they were deposited above the level of the dredge pond. Bulk values for the Spring Creek tailings are about 2-3 times background. Coarse material in the Shooting Gallery tailings shows background levels of mercury, whereas mercury levels in fine materials (sand size and smaller) are at background or are slightly elevated above background. Fine material shows higher values than coarse materials at three of four sites, but at two of the three Spring Creek sites the fine size fraction is lower than the bulk value. Either sampling error or analytical error or both may account for this apparently anomalous relationship between values for bulk versus coarse and fine fractions. The proportion of fine material (<2 mm) in stacker tailings is small, as observed while sieving the samples in the field. The silt- and clay-size material present may include mercury-contaminated wind-blown dust, and particles derived from abrasion of clasts with handling. Although these materials were probably deposited above the water level of the dredge pond, they may have been submerged later owing to water-level fluctuations, at which time suspended material from the dredge pond was transported into the tailings by infiltration.

Figure 13B shows results for sluice sand, silt-clay, and coarser layers that were probably deposited below water level in the dredge pond during mining. The first two categories show results for a lens of dredge sluice sand and overlying pebbly sand (mixed sluice/stacker, site CC22, fig. 3C) exposed in a trench at the west end of the Shooting Gallery. Values exceed 10 times background. Even the coarse fraction of the pebbly sand, which represents material transitional from sluice sand to overlying stacker tailings, shows significant mercury contamination. The next four categories in figure 13B show results for a trench at the eastern end of the Shooting Gallery (sites CC15 and

CC16; fig. 7). Here several layers were sampled, including a silt-clay layer, coarse and fine sluice sands, and overlying stacker tailings. Bulk values range from at least twice to more than five times background, with higher values associated with smaller grain sizes. Fines in the stacker tailings (sand, silt, and clay) show a mercury value comparable to those of the fine-grained layers. The figure 7 photograph shows the wall of the Shooting Gallery backhoe trench that yielded these four samples. It shows that the coarse tailings at the top of the section are very poorly sorted; the bulk mercury value and values for the size fractions from this layer are indicated on the photo. The layer below contains the finest material in the section (silt-clay), which shows the highest mercury value (160 ng/g), whereas sands interbedded with and underlying the silt-clay both show an intermediate value of 90 ng/g. Bulk values for the >2 mm portion of the coarse upper layer approach background (40 ng/g).

The last two categories in figure 13B show mercury concentrations in bulk mud from Red Pond, an abandoned dredge pond remaining at the upstream end of a small tributary to Spring Creek (fig. 8). This mud, composed predominantly of silt and clay, presumably settled in the pond after dredging ceased. Its high mercury content is similar to the sluice sand at the west Shooting Gallery site (CC23). Clay eroded from Tehama Formation and Nomlaki Tuff bedrock in the vicinity probably has also contributed to the sediment in this pond. Site CC35 is a small pond occupying a sink hole located only 10-15 m from Red Pond, in the same muddy tailings materials.

In the course of restoration activities in the lower Clear Creek area, dredge tailings have been reconfigured and moved. As tailings are excavated, any contaminated sluice sand and silt layers present are likely to be mixed with coarse stacker material. Figure 13C shows results for tailings that have been excavated and moved or otherwise disturbed. The South Pond categories represent results for samples collected in late August 2001 from the north and south sides of this pond (sites CC1 and CC3, respectively; table 1), which was an abandoned in-stream gravel mining pit, while it was being filled with tailings mixed with some local stream alluvium (site CC4). Although the sample of dry bulk fill material showed a mercury value near background levels, the two samples of fill material from the pond showed relatively high mercury values in both bulk and silt-clay size material. Although it is impossible to determine what proportion of these fines came from alluvium versus tailings, the results show that construction activities can release fine materials with relatively high mercury values.

Figure 13C also shows mercury values for Restoration Grove and Reading Bar. Restoration Grove is a reconstructed and revegetated segment of the flood plain of lower Clear Creek (McBain & Trush, 2001). Dredge tailings from Reading Bar were used as borrow material for this project. Mercury values for one site each in Restoration Grove and Reading Bar are similar. The modest mercury enrichments, especially in the fines (sand-silt-clay), are consistent with probable mixing of coarse and fine tailings materials, as described above.

Stream and Flood-Plain Pond Sediments

Since most of the gravel inventory of lower Clear Creek has been exposed to gold mining and recovery processes, active alluvium in the Clear Creek channel and on the adjacent flood plain is largely reworked tailings. Therefore mercury concentrations in stream sediments are of interest. To examine stream sediments, we repeatedly sampled three sites in the active channel of lower Clear Creek: a site immediately upstream from the SR 273 bridge (site CC30, fig. 3A), a site about 1.5 km

downstream from the former site of Saeltzer Dam (site CC11, fig. 3B), and a site at Reading Bar (site CC29, fig. 3C). A fourth site is at the U.S. Geological Survey gaging station just above the Placer Road crossing (Igo Gage, site CC18, fig. 3D), which is near the downstream end of the gorge reach of Clear Creek, above the lower reach (using Clear Creek reaches defined by Moore, 2002). A site in the active channel at Restoration Grove was added in summer 2003 (site CC36, fig. 3A). This site was disrupted by construction activities related to restoration in 2001. Flood-plain ponds are closely related to the active channel because they occupy parts of channels that are abandoned as the main active channel migrates across the flood plain, but continue to serve as overflow channels. They are flushed by winter high flows one or two years out of three. Mercury concentration data for stream and flood-plain pond sediments are presented in figures 14 and 15. Stream sediment data are presented in geographic order, downstream from the Igo Gage (figs. 14A and 14B).

The site at the Igo Gage (CC18) is in a small embayment at the edge of the active channel. Bulk mercury values for gravel range from near background to several times background (fig. 14A). In 2002 we analyzed bulk gravel and three size fractions; although sand-size and coarser-than-sand-size fractions show background mercury levels, mercury in fines is significantly above background, and the value for the bulk sample shows the mercury contribution from these fines. Results for 2003 were roughly similar. This confirms that sediments upstream from the lower Clear Creek area contain mercury above background levels, presumably from historic gold-mining activities in the upper reaches of the Clear Creek basin. The last category for the Igo Gage area (site CC19) represents a pebbly sand from a flood bar deposit, which shows a near-background mercury value.

Bulk sediment mercury values for sites on lower Clear Creek (CC29, CC11, CC36, CC30) are variable, ranging from near background to several times background (figs. 14A, B). The pattern throughout the reach is that the coarse fraction (>2 mm) shows a lower mercury concentration than the bulk material, the sand-size fraction shows about the same or somewhat higher, but occasionally significantly lower mercury concentrations than the bulk material, and the silt/clay size fraction is always enriched in mercury. Bulk values are influenced mainly by the amount of mercury in fines, being a function of the proportion of fines and the concentration of mercury in them. The results for site CC30, at the Highway 273 bridge (fig. 14B), suggest that mercury concentration in fines may decline downstream. The University of Montana study, however, which presents trace metals data for the silt/clay fraction in sediments at more than 20 sites on lower Clear Creek, showed no clear downstream trend within the reach (Moore, 2002; Moore and Hughes, 2003).

The flood-plain ponds fill low spots in channels that carry water during high-flow periods. Although the channels are cut in alluvial gravels of the flood plain, they contain fine mud deposited as flood flows slacken. Bulk sediment samples from two flood-plain ponds show modest enrichment of mercury above background levels (fig. 15). The fines (silt-clay), however, show mercury enrichments similar to fines in Clear Creek stream gravels. The two 2002 East Pond samples are replicates from site CC2. The sample having the relatively high bulk mercury value contained abundant mercury-enriched fine mud.

Methylmercury in Tailings and Sediments

All forms of mercury are more or less toxic to humans and biota (Agency for Toxic Substances and Disease Registry, 1999). The most toxic are organic mercury compounds. All forms of mercury, including metallic mercury (Hg^0), mercurous ion (Hg^+) and mercuric ion (Hg^{2+}) can be converted to

organic mercury compounds in aquatic systems. The most toxic organic mercury compound formed in significant amounts is methylmercury. Monomethylmercury (CH_3Hg^+) is the dominant form; dimethylmercury may occur in small amounts. In the analytical procedure for methylmercury and mercury (see earlier section) dimethylmercury is converted to monomethylmercury, so it cannot be determined separately (Horvat and others, 1993). Methylmercury is formed by microbial action and is favored by warm temperatures, low dissolved oxygen and redox potential, and presence of dissolved organic carbon and moderate concentrations of sulfate (Wiener and others, 2003; Benoit and others, 1998). Once formed, methylmercury tends to be concentrated in tissues of aquatic organisms (bioaccumulation) and increase in concentration relative to ambient water in organisms at progressively higher trophic levels (biomagnification). As a result of biomagnification, ingestion of mercury-contaminated fish is the most common mercury exposure pathway for humans (Mergler and others, 2007).

Figure 16 shows results of methylmercury analyses for a subset of the same wet sites and subsamples as shown in figures 11, 13, 14, and 15, and also for some dry sites (data in table 3). Note that both axes of the plot are logarithmic, so equal-spaced diagonal lines differ by a factor of ten. Methylmercury values for dry tailings and sediments are all very low, ranging from <0.015 ng/g to 0.387 ng/g, accounting for less than one percent of total mercury present, indicating that no extraordinary conditions favorable for methylation of mercury occur in these materials in the area. In wet materials, methylmercury shows the same tendency as total mercury to be enriched in fine size fractions relative to bulk material. Methylmercury generally accounts for one percent or less of total mercury present in sediments and tailings, which indicates normal rates of conversion of mercury to methylmercury. Sediments from Red Pond, the flood-plain ponds, and the Clear Creek backwater at the Igo Gage, however, showed methylmercury in the range of 2.5 to 5 percent. Red Pond mud showed a low proportion of methylmercury early in the summer of 2002, whereas the proportion of methylmercury was high (about 20 percent) in late summer of 2001 and relatively low again (about 2 percent) in mid-summer 2003.

Atmospheric Mercury Deposition and Mercury Vapor Fluxes

We have not attempted to quantitatively evaluate atmospheric deposition of mercury, which could add to total mercury in sediments and tailings in the area (Fitzgerald and others, 1998). Notable increases in mercury concentrations in soils and sediments from atmospheric deposition, however, are likely to be restricted to within a few centimeters of the surface (see, e.g., Rood and others, 1995). Dry stacker piles that have been exposed undisturbed for more than 60 years typically show background mercury levels, including the fine size fraction, indicating that atmospheric deposition is insufficient to increase mercury concentrations above background levels. Generally finer grain-size materials show the highest mercury concentrations, which is anticipated based on the gold recovery processes used, suggesting that gold mining is the main source of mercury in the area. However, some atmospheric contribution to fine grain size fractions of stream and pond sediments cannot be ruled out.

Mercury vapor emission could reduce or redistribute mercury in tailings. Vapor emission has been measured over dry tailings at Reading Bar, Restoration Grove, and at several sites along Spring Creek and at the Shooting Gallery. Mercury vapor flux was also measured over undisturbed gravels at China Garden. Although bulk mercury values within 10-50 cm of the surface at these sites range from background levels (in stacker tailings) to more than 1 mg/kg (in dredge sluice sands), preliminary results of the flux measurements show that mercury vapor emissions are not significantly above background levels at any site (Richard Zehner and Mae Gustin, unpub. data, 2002). This suggests that metallic mercury added to the sediments for gold recovery has been converted to non-volatile mercury compounds.

Effect of Sample Treatment on Analytical Results for Sediments and Tailings, and Implications

About 60 percent of the sediment and tailings samples, both wet and dry, were analyzed by both Chemex and Frontier laboratories using different methods of preparation and analysis (see section on analytical methods for sediments and rocks, and table 2).

Results are compared in figure 17. Identical results fall on the 1:1 line in the figure. The lines on either side of the 1:1 line show a two-standard deviation analytical error envelope. The analytical error is not rigorously determined from replicate analysis, but rather is estimated from laboratory experience. The comparison plot demonstrates that samples with low mercury concentrations (less than 30 ng/g) show similar results by both methods. At higher mercury concentrations, most samples whose median grain size is sand size or smaller also yield similar results by both methods. Results that fall outside the envelope, considered likely to be significantly different, are mostly for samples with large median grain size, and in all cases the Frontier analyses show the higher mercury values. Because the Frontier analysis method does not include material from the silicate clasts, which are likely low in mercury, this confirms that much of the mercury present is associated with sand or smaller grain sizes and localized in aqua regia-leachable mineral grains or coatings on silicate grains. The mercury in leachable grains and coatings is also incorporated in the Chemex analyses, but the pulverization and digestion includes low-mercury material from clasts, which dilutes the leachable mercury. Although some mercury losses from the Chemex drying step cannot be ruled out, the similarity between Chemex and Frontier results for other samples, especially those having relatively high mercury concentrations and relatively small grain size, suggest that such losses are not significant.

Mercury Concentrations in Waters

General Comments

Figure 18 shows total mercury results for waters, most of which were sampled at the same sites as wet sediments (figs. 11, 13, 14, 15). The plot shows the total (unfiltered) mercury concentration (x axis), versus dissolved (filtered) mercury (y axis). The diagonal lines indicate equal proportions of dissolved versus total mercury. "Dissolved" mercury is here operationally defined as the concentration of mercury measured in an aliquot passing a 0.45 m filter. The filtration was done in the laboratory under ultra-clean mercury-free conditions (see section on analytical methods for waters). Note that both axes of the plot are logarithmic, so equal-spaced diagonal lines differ by a factor of ten. The results are discussed in detail in the sections that follow.

Proposed aquatic life criteria for total mercury in water span a wide range. An aquatic life criterion is a level above which there is evidence of potential for observable effects in biota, not necessarily including bioaccumulation and biomagnification. The lowest proposed criterion for chronic effects in fresh-water aquatic life (12 ng/L total recoverable mercury) is shown on figure 18 for reference. It was first recommended by EPA in 1986, and is the level recommended by BLM's National Applied Resource Sciences Center (Barkow, 1999). In April 1999 EPA issued a guideline of 770 ng/L (dissolved mercury) for chronic effects in fresh-water aquatic life. The 50 ng/L level shown on figure 16 is the California Water Quality Standard. This number, proposed in 1997, refers to total recoverable mercury and is a human health standard. Most samples are below the 12 ng/L level.

Mercury Concentrations in Clear Creek and Flood-Plain Ponds

Total mercury values for Clear Creek stream waters are generally low (less than about 3 ng/L; fig. 18), and similar to many values for waters obtained elsewhere in the Sacramento River Basin (Domagalski, 1998; Domagalski and others, 2000a,b). The proportion of dissolved mercury ranges greatly, from about 12 to 65 percent. The values that cluster below 3 ng/L represent samples collected during the summer low-flow and winter base-flow periods. The cluster of three samples having relatively high total mercury (around 10 ng/L) and low percentages of dissolved mercury were collected during a winter high-flow period.

Figure 19 shows the relationship between unfiltered and filtered (total and dissolved) mercury and turbidity for samples from Clear Creek during low-flow and high-flow conditions. Data for several ponds in the Old Mill area (discussed in detail in a later section) are shown for comparison. Results for filtered and unfiltered aliquots for each sample are connected by a vertical tie line. The cluster of Clear Creek samples with low total mercury (less than 3 ng/L) and turbidity less than 5 nephelometric turbidity units (NTU) represent samples taken at Igo Gauge (site CC18), Reading Bar (site CC29), below Saeltzer Dam (site CC11), and at Highway 273 (site CC30) during winter base flow conditions (200 ft^3/s). Under these conditions total mercury values are similar throughout the lower reach of Clear Creek, and proportions of dissolved mercury fall in a relatively narrow range of about 20-50 percent. The Clear Creek samples having turbidity values of 8 NTU or more were collected at the same locations during a controlled release from Whiskeytown Reservoir (fig. 1) on 27 January 2003 (peak flow 1200 ft^3/s). Under these flow conditions total mercury concentrations increase and are roughly correlated with turbidity, whereas concentrations of dissolved mercury remain the same, indicating that increased amounts of mercury present during high flow conditions represent mercury associated with increased numbers of suspended sediment particles. In contrast, the wetlands ponds at the time of sampling, during quiet winter weather conditions, showed consistently low turbidities and no relationship between turbidity and suspended mercury.

The data points for flood-plain ponds (fig. 18) include samples from South Pond and Pond 3, collected in summer 2001, and East Pond, collected in summer 2001 and again in the summers of 2002 and 2003. In 2001 South Pond and Pond 3 were filled in as part of the flood-plain restoration program. The range of total mercury values in the ponds is within the range of total mercury values in Clear Creek waters, and the range of proportion of dissolved mercury (12 to 42 percent) is within the range of dissolved mercury seen in Clear Creek waters. The average total mercury value in the ponds, however, is higher than the average value in Clear Creek waters, and the average percentage of dissolved mercury is lower. This may reflect the fact that the ponds are intermittently flushed by Clear Creek during winter high flow events, at which time they receive water with relatively high total mercury concentrations and low percentages of dissolved mercury.

Chloride concentrations (fig. 20) in stream waters and some pond waters, which fall between about 1.5 and 5 mg/L, are in the normal range for surface waters near the Pacific Ocean (Hem and others, 1990) and similar to chloride concentrations found in most parts of the Sacramento River basin (Domagalski and others, 2000a). The higher concentrations in East Pond and Pond 3 (about 10-15 mg/L) likely reflect effects of evaporative concentration.

Sulfate concentrations (fig. 21) fall in two groups. Those below 5 mg/L are in the normal range seen in West Coast rivers (Hem and others, 1990). Values for Clear Creek waters and most samples from flood-plain ponds are in this range. An exception is the elevated sulfate level seen in East Pond in 2001. This sulfate value is accompanied by elevated chloride, which suggests that significant evaporative concentration occurred during the summer months, and could explain the elevated sulfate.

In 2002, however, chloride was similarly elevated in East Pond while sulfate was not, suggesting that sulfate may vary for other reasons. Pyrite-bearing reduced sediments of the Tehama Formation are exposed in the bed of Clear Creek near the site of Pond 3. Where exposed to oxidation, pyritic reduced zones in the Tehama Formation could be sources of sulfate in waters. The group of waters with sulfate values above 5 mg/L include saline spring waters, and ponds in the Old Mill area, which are described in the following section. Figure 21 shows that total mercury is not correlated with sulfate in either the low-sulfate or the high-sulfate group.

Iron concentrations vary greatly in the waters sampled (fig. 22). Flood-plain ponds show surprisingly high iron, although no iron mineral precipitates have been observed. The source of iron is not obvious, but oxidizing sulfides in nearby bedrock are a possible source, as well as a source of sulfate in the case of East Pond. In all cases where we have iron analyses for both unfiltered and filtered subsamples, the unfiltered values are several times the filtered values. This indicates that the majority of iron in the waters is present in suspension, probably as iron oxyhydroxide particles or coatings on other particles, a condition that is commonly seen in natural waters.

Mercury Concentrations in Saline Waters—Saline Springs and Red Pond

In addition to the dilute surface waters described above, highly saline waters occur locally in the lower Clear Creek area. Red Pond, an abandoned dredge pond (site CC12, figs. 2, 3C), shows chloride concentrations from 3600 to 13,000 mg/L. It captures drainage from a saline spring (site CC27, fig. 3C), which had a chloride concentration of 32,000 mg/L in June 2002, and 45,000 mg/L in late July 2003. Saline water at this site discharges from a set of north-trending fractures in an outcrop of Cretaceous granodiorite. The chloride concentration measured for this spring water early in the summer of 2002 is similar to that of seawater (about 30,000 mg/L), whereas later in the summer, in 2003, it exceeded that of seawater, owing to evaporative concentration. At site CC28, saline water discharges from a corroded 6-inch-diameter pipe that is blocked at a depth of about 0.5 m. The discharge point is located near the bottom of a valley, suggesting that this occurrence is another saline spring. It is possible, however, that the pipe is the head of a well that taps a deeper artesian source. The water from this spring also has a chloride concentration (29,000 mg/L) similar to that of seawater.

Red Pond shows moderately elevated levels of total mercury (10-40 ng/L; fig. 18), both early in the summer when the pond level is high and late in the summer when it is low. The saline spring water that drains into Red Pond also shows elevated total mercury concentrations (the value near 50 ng/L measured in 2003 is more reliable than the lower value determined in 2002, which is a rough estimate; see table 5). Both saline spring waters and Red Pond waters show considerable variation in the proportion of mercury that is dissolved (fig. 18). In the case of Red Pond, this variation may result from an interplay between leaching of mercury from fine-grained mercury-enriched sediments and adsorption of mercury on iron oxyhydroxide particles during periods in which iron is being precipitated. It may also be to some extent an artifact of sampling; bulk subsamples collected for mercury determinations may have incorporated various amounts of iron oxyhydroxide floc. The saline spring water collected in June 2002 showed a high proportion of dissolved mercury, as expected. The saline spring water collected in July 2003, on the other hand, showed a low proportion of dissolved mercury. Although precipitation of iron or aluminum minerals was not obvious, salts were being deposited at that time, so possibly mercury was adsorbed to suspended particles.

The water discharging from the saline well (site CC28) precipitates sulfide that is enriched in mercury (67 ng/g), which explains the relatively low total mercury value for this water (fig. 18). The mercury detected in the water sample was probably associated mainly with suspended sulfide particles, resulting in the low percentage of dissolved mercury.

Figure 20 shows that in the lower Clear Creek area high-salinity waters generally contain more mercury than low-salinity waters, but within the high salinity group, mercury and chloride are not correlated.

Sulfate levels measured in saline waters were elevated relative to Clear Creek waters, with the exception of Red Pond in summer 2001 (fig. 20). Note that the high salinity reduces the analytical precision that can be achieved for other anions by the IC method, so sulfate values for high-salinity samples are determined by ICP-MS. Three samples from Red Pond showed about the same or higher sulfate concentrations as the saline spring that feeds the pond; however, chloride concentrations in Red Pond indicate that saline water has been diluted by surface water at a ratio of 2:1 or more. The excess sulfate is probably derived from the Tehama Formation. Prospect tunnels in gravels of the Tehama Formation exposed above Red Pond contain fine-grained pyrite that is now being oxidized. The Red Pond sample with a relatively low sulfate concentration may reflect depletion of sulfate by reduction to sulfide near and below the sediment-water interface. Black mud, indicating reducing conditions, appears 1-2 cm below the sediment-water interface in Red Pond. Figure 21 shows that total mercury is not correlated with sulfate in saline waters.

Iron values are high in Red Pond, and are anticipated from the observation that iron oxyhydroxide precipitates on the bottom and on pond vegetation during the summer months (figs. 8 and 22). The relatively high iron values in flood-plain ponds were discussed in the previous section.

Mercury Concentrations in Wetlands and Drainage, Old Mill Area

The drainage system utilized for placer mining in the Old Mill area is partly preserved. The most prominent feature is the southwest-trending drainage ditch cut in bedrock (Cretaceous shale and Nomlaki Tuff) at the southern edge of the mined area, separated from Clear Creek by a low narrow ridge of Nomlaki Tuff bedrock (fig. 12). This ditch drains to Clear Creek through the tunnel cut through the bedrock ridge. The ditch was lined with sluices, and sluices were probably also installed in the tunnel. Although the sluices have been removed, sediment in the ditch contains elemental mercury and gold amalgam (see previous section on tailings in the Old Mill area). Water flows in the ditch and through the tunnel during the winter rainy season. Total mercury values for waters in the ditch and the tunnel (fig. 18) are somewhat elevated, and most samples show a relatively high proportion of dissolved mercury, probably as a result of contact with the mercury metal in the bed load. The tunnel discharge sample collected in late summer 2001 showed relatively high total mercury and very low dissolved mercury. At the time of sampling the water level at the tunnel portal was very low (about 10 cm) and flow was imperceptible. As a result, fine mercury-rich floc may have been dislodged from the bottom during sampling and incorporated into the water sample. Subsequent samples collected in summer 2002 and winter 2003 did not show similar high total and low dissolved mercury.

In the eastern part of the Old Mill area, the placer mining drainage system and the terrace surface that was mined have been modified; drainages have been blocked and berms added to form seasonal

wetlands (fig. 12; cover photo). The ditch described above receives water from the dredge tailings tract to the east and northeast and from an unidentified groundwater source (discussed below), but under normal conditions does not receive runoff directly from the constructed ponds. Such conditions prevailed at the times that waters in the ditch and tunnel were sampled. During heavy rains the ponds overflow and drain over a spillway into a small catchment pond (sump) and then into the ditch.

We sampled three ponds in the Old Mill area in winter of 2003, including one of the constructed ponds, the sump pond, and a pond in tailings impounded where a drainage cut is blocked by road fill, known as the Beaver Pond. The total mercury values for these ponds overlap values for the flood-plain ponds but include higher values (fig. 18). The proportion of mercury present as dissolved mercury is more than 50 percent in all three. The ponds contain mercury-enriched fine sediment derived from placer mine tailings (160-310 ng/g; see fig. 11). When the ponds fill in the winter, soluble mercury species are probably extracted from the fine sediments, resulting in the relatively high total mercury values and consistently high percentages of dissolved mercury seen in the ponds.

Waters in the drainage ditch and tunnel show chloride concentrations that are relatively high, but not as high as saline spring waters or Red Pond, which contains diluted saline spring water (fig. 20). In summer, discharge from the tunnel has chloride concentrations about two orders of magnitude higher than typical surface waters. In winter, when there is water in the drainage ditch, chloride concentrations in drainage waters, both in the ditch and at the tunnel portal, drop to about one order of magnitude higher than surface waters, owing to dilution from surface runoff. In contrast, the constructed wetlands ponds show chloride concentrations in the range of Clear Creek waters or slightly higher, but within the range for flood-plain ponds.

Connate waters derived from the Cretaceous sedimentary rocks of the Great Valley Sequence are high in boron, chloride, iodide, bromide, and lithium (Rytuba and others, 2000; Donnelly-Nolan and others, 1993). Such waters appear in springs on the west side of the Sacramento Valley, to the south of the Clear Creek area, and contribute to fluids in The Geysers geothermal field and to ore-forming fluids and groundwaters at the McLaughlin gold deposit. As figure 23A shows, high concentrations of boron and lithium accompany the chloride in saline spring waters in the Clear Creek area, and indicate that the saline springs probably represent similar connate fluids from sedimentary rocks at depth (bromide and iodide were not determined in this study). The somewhat lower boron and lithium values seen in Red Pond, which fall on saline spring-surface water mixing lines for boron and lithium (fig. 23B), indicate that Red Pond waters consist of mixtures of connate water and surface water, with the proportion of surface water varying seasonally.

In drainage waters from the Old Mill area, elevated boron and lithium values accompany elevated chloride concentrations (fig. 23A), indicating that they are probably also mixtures of surface waters and connate waters similar to those near Red Pond. In the Old Mill area no saline springs have been identified; apparently a small amount of saline ground water mixes with meteoric water (surface water that has percolated down to the water table). The composition of water discharged from the tunnel in the summer gives an indication of the composition of this mixed ground water. The compositions of drainage waters in the winter indicate that run-off seasonally dilutes the ground water by a factor of about 10.

Most flood-plain pond waters have boron concentrations slightly higher than Clear Creek, and lithium concentrations that are slightly lower than Clear Creek in spite of modestly elevated chloride in some ponds (fig. 23A). In the constructed wetlands of the Old Mill area, on the other hand, boron is

distinctly higher than in most Clear Creek waters, even though chloride is low, and lithium concentrations are the same as most Clear Creek waters. The Beaver Pond has lower boron and higher lithium than other ponds in the Old Mill area. Although it is not clear why boron-lithium-chloride relationships in the two types of ponds differ from those of surface waters and from each other, neither shows patterns that suggest connate waters are responsible for the compositional variations.

Some connate waters are enriched in mercury relative to surface waters, so waters of deep-seated origin may locally contribute some mercury to the surface environment in the Clear Creek area, in addition to mercury introduced during gold mining.

In the Old Mill area, summertime sulfate values in the drainage tunnel discharge are among the highest seen so far in the Clear Creek area (fig. 21). Sulfate is also relatively high in the ponds, and similar to values for the nearby drainage during the wintertime, when the ponds contain water. Sources for this sulfate have not been identified. Possibilities include the saline groundwater that contributes chloride to the drainage (but apparently does not affect the ponds), sulfide-bearing Tehama Formation, and possibly sulfide-bearing pre-Tertiary bedrock.

Methylmercury Concentrations in Waters

Figure 24 shows amounts of methylmercury versus total mercury for unfiltered waters. Lines of equal proportions of methylmercury are shown for reference (note that as for figure 18, the scales for both axes and the intervals between the percentage lines are logarithmic). Most of the methylmercury values measured fall within the range seen elsewhere in the Sacramento River Basin (0.02-0.85 ng/L; Domagalski, 1998, 2000b). In stream waters between 1 and about 5 percent of total mercury is methylmercury, which is within the range of values commonly seen in relatively uncontaminated oxic surface waters (0.1-5 percent; Wiener and others, 2003; Kelly and others, 1995). Only one South Pond sample had a methylmercury percentage in this range, otherwise proportions of methylmercury were relatively high in Pond 3 and East Pond in 2002 and 2003, showing that conditions in shallow ponds on the flood plain may favor methylation of mercury.

Proportions of methylmercury in all Red Pond samples were high, reaching 65 percent of total mercury present in water (fig. 24) and 20 percent of total mercury present in sediment (fig. 16). Because total mercury concentrations are also relatively high (figs. 13B and 18), concentrations of methylmercury in both water and sediment are large (figs. 24 and 16).

All ponds with high proportions of methylmercury are shallow, contain decaying organic matter, and were warm at the time of sampling (24-29°C). Whether the unusual high chloride content of Red Pond waters plays a role in the production and preservation of methylmercury (or high methylation versus demethylation rates) there is unclear. Somewhat elevated chloride concentrations under some conditions may enhance mercury bioavailability, whereas higher concentrations may reduce bioavailability (Wiener and others, 2003). The constructed wetlands ponds were not tested for methylmercury.

The extraordinarily low methylmercury level measured in water in the hydraulic drainage tunnel in 2001 (0.042 ng/L, representing 0.12 percent of total mercury) was not confirmed in 2002. The value obtained in 2002 is within the 1-10 percent range for methylmercury common in the lower Clear

Creek area. Mercury-rich fine particulate material (sediment or biogenic floc) may have been incorporated in the 2001 sample, leading to an erroneously high total mercury value, as described in an earlier section. The filtered subsample collected in 2001 showed a proportion of methylmercury in the normal range; total mercury in the filtered subsample was 2.94 ng/L (versus 50.7 ng/L in the unfiltered sample), 1.4 percent of which was methylmercury (see table 5), suggesting that fine particulate material was responsible not only for the high total mercury value but also for the anomalously low methylmercury value. It is not clear, however, why incorporating mercury-rich particulate material into the sample would result in increased total mercury without accompanying increased methylmercury.

Saline spring waters show low methylmercury values, which is not surprising because these waters were collected at discharge points and therefore were exposed to surface conditions for only short periods of time.

Detailed studies of mercury speciation and cycling in wetlands, estuaries, and other aquatic environments have shown that mercury methylation occurs where sulfate-reducing bacteria (SRBs) are active, and that most methylation takes place at the oxic/anoxic interface in sediments, which is usually near the sediment/water interface (Wiener and others, 2003; Gilmour and others, 1998). Figure 25A shows sulfate versus percentage of total mercury present as methylmercury in waters. Clear Creek waters, saline spring waters, and Old Mill area drainage waters have relatively low percentages of methylmercury, and there is no relation between methylmercury percentage and sulfate content. High methylmercury percentages were restricted to ponds, and sulfate values in these ponds ranged from 2 to 11 mg/l; however, there is no correlation between methylmercury percentage and sulfate concentration. At the same sites with high methylmercury percentages in water the percentage of methylmercury in fine-grained sediments (<63 m sieved fraction or bulk mud) also tends to be relatively high (fig. 25B). The relatively high methylmercury value in sediment at the Igo Gage is associated with a sulfate value in the 2-11 mg/L range as well (2.1 mg/L by IC method, 3 mg/L by ICP-MS method). Studies of mercury speciation in sediment pore waters near the sediment surface have shown high methylmercury percentages associated with a similar range of sulfate concentrations (about 2-8 mg/L; Benoit and others, 1998). Although some sulfate must be present to support SRBs, sulfate-rich environments (with more than about 10 mg/L sulfate) are not favorable owing to accumulation of sulfide, which apparently reduces the bioavailability of mercury to SRBs, possibly by reducing the abundance of the HgS^0 neutral complex (Benoit and others, 1999), and thus reduces mercury methylation. Although our sulfate data are for superjacent waters rather than sediment pore waters, a range of sulfate values optimal for mercury methylation appears to exist here as it does elsewhere.

Figure 26 shows that moderate to relatively high total organic carbon (TOC) concentrations are associated with the high proportions of methylmercury in Red Pond and East Pond waters. The elevated TOC values result from abundant vegetation and decaying organic matter in these ponds. Water collected from the Igo Gauge site in 2002 showed the highest TOC concentration seen in Clear Creek waters; the fine-grained sediment collected at the same time showed the unusually high percentage of methylmercury noted earlier.

Conclusions

Mercury analyses for materials unaffected by anthropogenic mercury inputs, including bedrock, clasts in gravels, and unmined auriferous gravels indicate that background mercury concentrations are low, generally 10 ng/g or less. The highest value measured for an abundant rock type in the area was 30 ng/g, in metamorphic rock clasts from stream gravels. High-salinity connate waters enter the surface environment in the area through springs and seeps, and these waters may be an additional, but probably relatively minor, source of mercury. Sulfide mineral precipitates in discharge aprons around some saline springs contain more that 50 ng/g mercury, but volumes of such materials are insignificantly small. Mercury from atmospheric sources is probably also relatively minor.

In materials enriched in mercury, including tailings and sediments derived from tailings, most of the mercury is associated with fine grain-size fractions. Consequently mercury concentrations in sand-size or smaller fractions in tailings are commonly above the NOAA ERL of 150 ng/g. The highest values are seen in lenses of finer-grained material, mainly sand but including silt and clay, that was exposed to mercury in sluice boxes or on separating tables. The lowest values, near background, are seen in stacker tailings that were deposited above the dredge pond water level during mining.

Bulk mercury values in tailings and sediments reflect the concentrations of mercury in the fine grain-size components and the proportion of fines in the bulk material. Bulk mercury values are lowest in stacker tailings that were deposited above the dredge pond water level, and highest in dredge sluice sands, as is the case for the fine fractions alone. Bulk mercury values in tailings generally exceed background concentrations. Bulk mercury values in stream and flood-plain sediments, which include reworked tailings, are modestly elevated above background values. Most of the mercury load in fine-grained tailings and sediments is anthropogenic.

Although low-level mercury contamination is ubiquitous in sediments in the lower Clear Creek area, mercury levels remain relatively low in Clear Creek stream waters under base-flow conditions. Elevated mercury concentrations are associated with increased amounts of particulate material entrained during flood events. Thus periods of significant mercury transport are episodic and transitory. The proportion of mercury converted to methylmercury in Clear Creek is in the normal range for fluvial environments, indicating that little methylation occurs and little methylmercury persists in flowing waters. Increased methylation may occur in quiet backwater areas, but because total mercury levels in stream waters are low, amounts of methylmercury produced are small.

Relative to Clear Creek, average mercury values are somewhat higher in ponds on the Clear Creek flood plain and proportions of dissolved mercury somewhat lower, probably reflecting intermittent flushing of these ponds by Clear Creek during winter high-flow events. Variations in chloride and sulfate contents of pond waters suggest that the ponds are also affected by groundwater inputs and evaporative losses. Ponds in the constructed wetlands of the Old Mill placer mining area show generally higher total mercury concentrations and higher proportions of dissolved mercury than the flood-plain ponds, probably reflecting leaching of soluble mercury from fine-grained tailings. The implication is that mercury in flood-plain ponds is dominantly associated with suspended particulate material that was transported into the ponds by Clear Creek, whereas mercury in the constructed wetlands was predominantly derived from leaching of fine-grained tailings.

Flood-plain ponds commonly show elevated percentages of methylmercury. In Red Pond, an abandoned dredge pond, a relatively large proportion of the mercury present is methylmercury, which coupled with relatively high total mercury values in water and sediment results in high methylmercury

concentrations. In the ponds where notable methylation occurs, sulfate values in water range from 2 to 11 mg/L and total organic carbon concentrations are elevated. Sulfate sources have not been positively identified, but saline springs and weathering of sulfide-bearing bedrock probably augment the sulfate normally present at low levels in surface waters. The ponds support vegetation and contain abundant organic debris, and are relatively warm, with winter-time lows around 10°C and summer temperatures ranging from 24°C to 32°C. The chemistry of Red Pond is unusual owing to input of saline spring water, but whether the high salinity there promotes methylation or suppresses demethylation and thus plays a role in producing high methylmercury concentrations, is unknown.

Although mercury and methylmercury concentrations vary greatly from pond to pond and vary over time in individual ponds, ponds in various environments are clearly the main sources of bioavailable methylmercury in the lower Clear Creek area. Since fine-grained sediment contaminated with mercury is present throughout the lower Clear Creek area, mercury supply is probably not a controlling factor in methylmercury production. Presence of sulfate in a favorable concentration range, organic carbon, warm summer temperatures, and possibly elevated salinity are factors that may promote methylation of mercury in lower Clear Creek area ponds.

Acknowledgements

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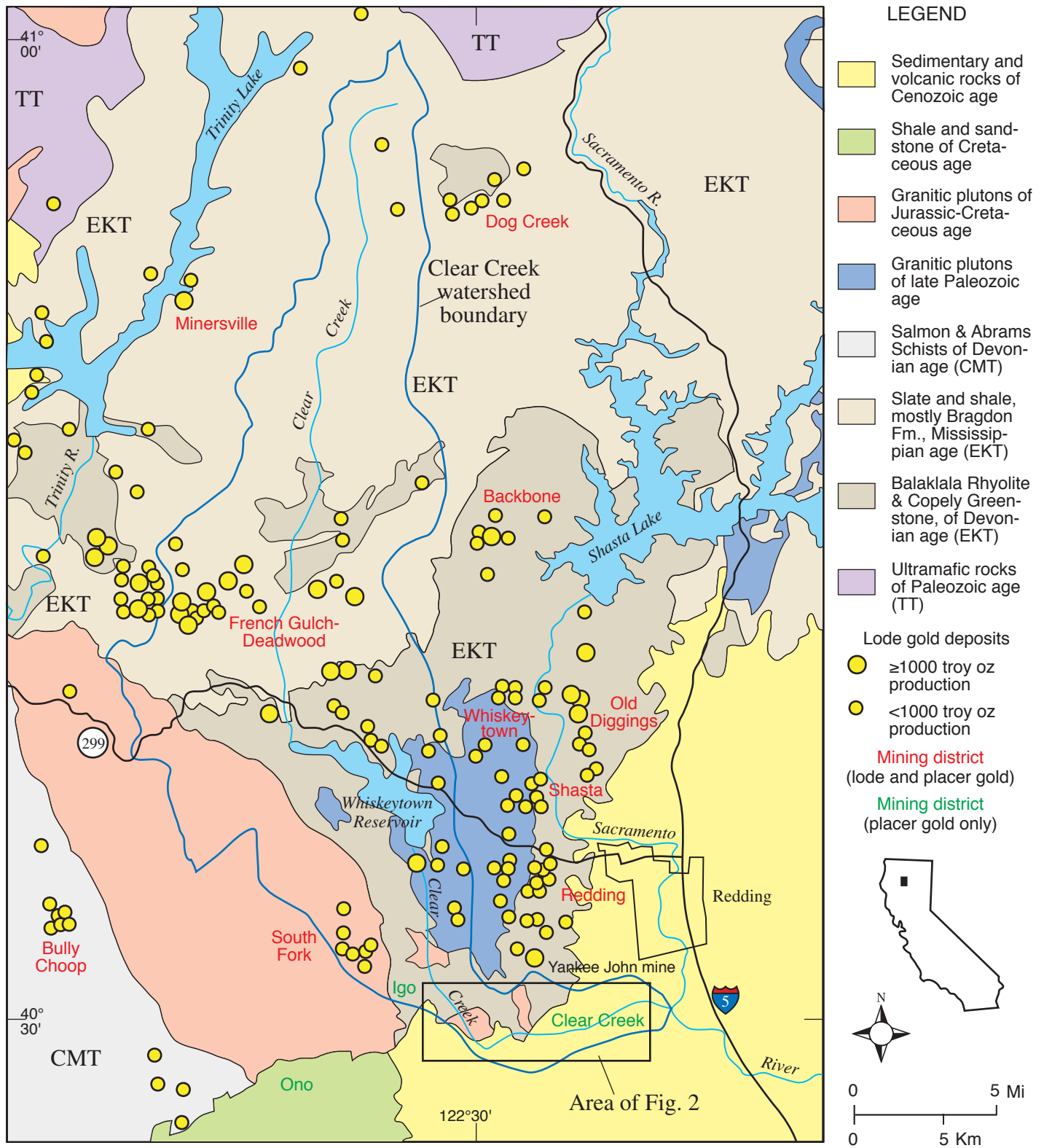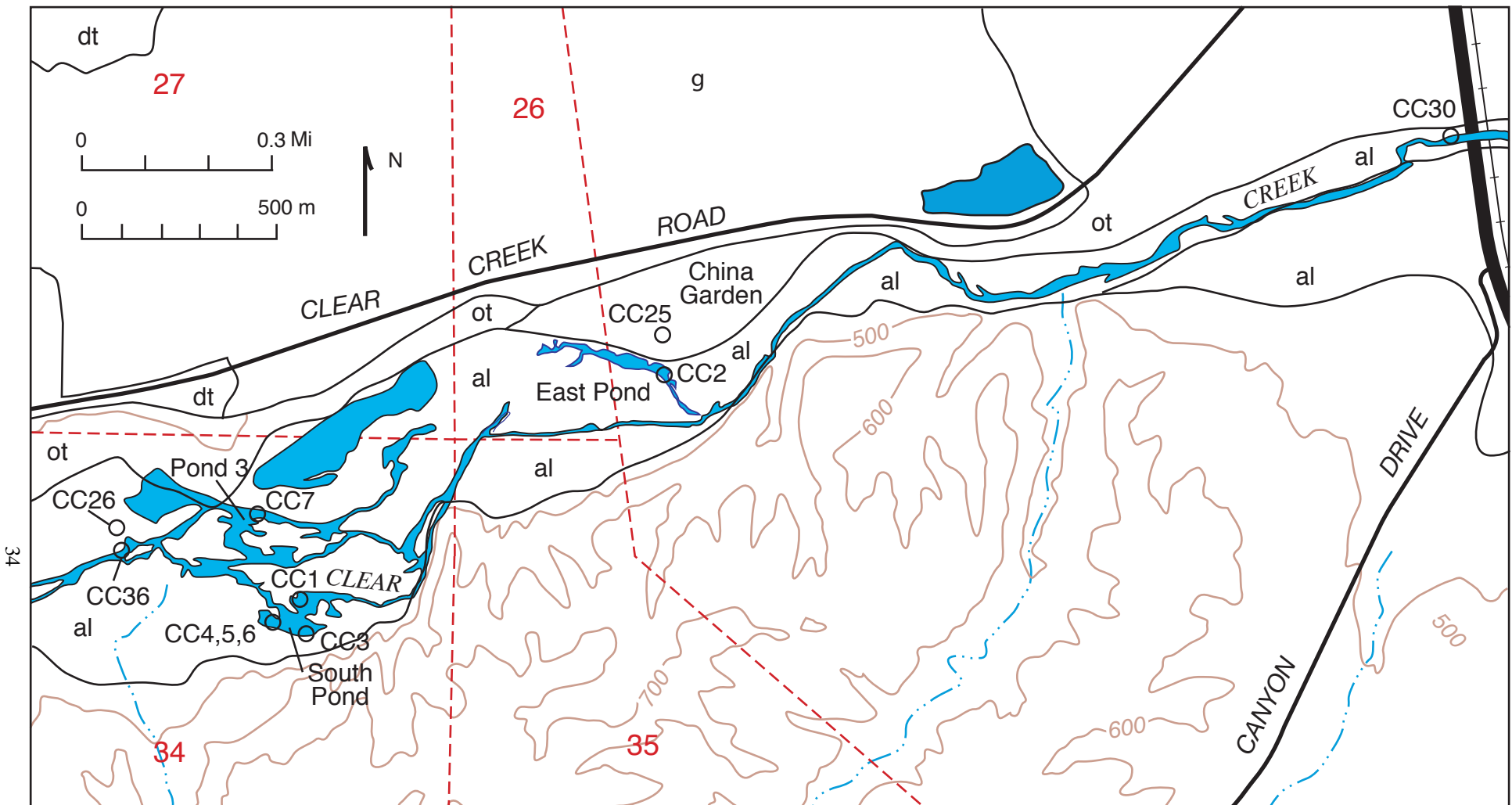
LEGEND

Sedimentary and volcanic rocks of Cenozoic age

Shale and sand-stone of Cretaceous age

Granitic plutons of Jurassic-Cretaceous age

Granitic plutons of late Paleozoic age

Salmon & Abrams Schists of Devonian age (CMT)

Slate and shale, mostly Bragdon Fm., Mississippian age (EKT)

Balaklala Rhyolite & Copely Greenstone, of Devonian age (EKT)

Ultramafic rocks of Paleozoic age (TT)

Lode gold deposits

≥1000 troy oz production

<1000 troy oz production

Mining district (lode and placer gold)

Mining district (placer gold only)

N

0   5 Mi

0   5 Km

41° 00'

TT

TT

EKT

Trinity Lake

Minersville

Clear Creek

Clear Creek watershed boundary

EKT

Dog Creek

Sacramento R.

EKT

EKT

Trinity R.

Backbone

Shasta Lake

EKT

French Gulch-Deadwood

EKT

Whiskey-town

Old Diggings

299

Whiskeytown Reservoir

Shasta

Sacramento

Redding

Redding

Clear

Bully Choop

South Fork

Igo

Creek

Yankee John mine

Clear Creek

I-5

40° 30'

CMT

Ono

River

Area of Fig. 2

122°30'

Figure 1. Map showing bedrock geology of the southeastern Klamath Mountains and location of the Clear Creek watershed, northern California. Geology from Hotz (1971) and Fraticelli and others (1987). Gold-quartz lode mine locations and sizes from Hotz (1971) and Clark (1970). Tectonostratigraphic terranes: EKT=Eastern Klamath; CMT=Central Metamorphic; TT=Trinity, from Irwin (1981, 1985). Mining districts have indefinite boundaries, so locations are approximate.

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For base information see figure 2

Compiled by R. P. Ashley, 2004-2006

EXPLANATION OF MAP SYMBOLS

g   Areas of dredge tailings removed by post-dredging gravel mining operations

al   Active stream alluvium

dt   Dredge tailings

ot   Pre-dredging placer-mined areas. Includes placer tailings, bedrock surfaces exposed by mining, and other mining-related features including drainage cuts and impoundments

Unassigned areas include alluvial terraces of Quaternary age and bedrock units of Tertiary and older age

Figure 3A. Eastern part of lower Clear Creek area, showing locations of sampling sites.
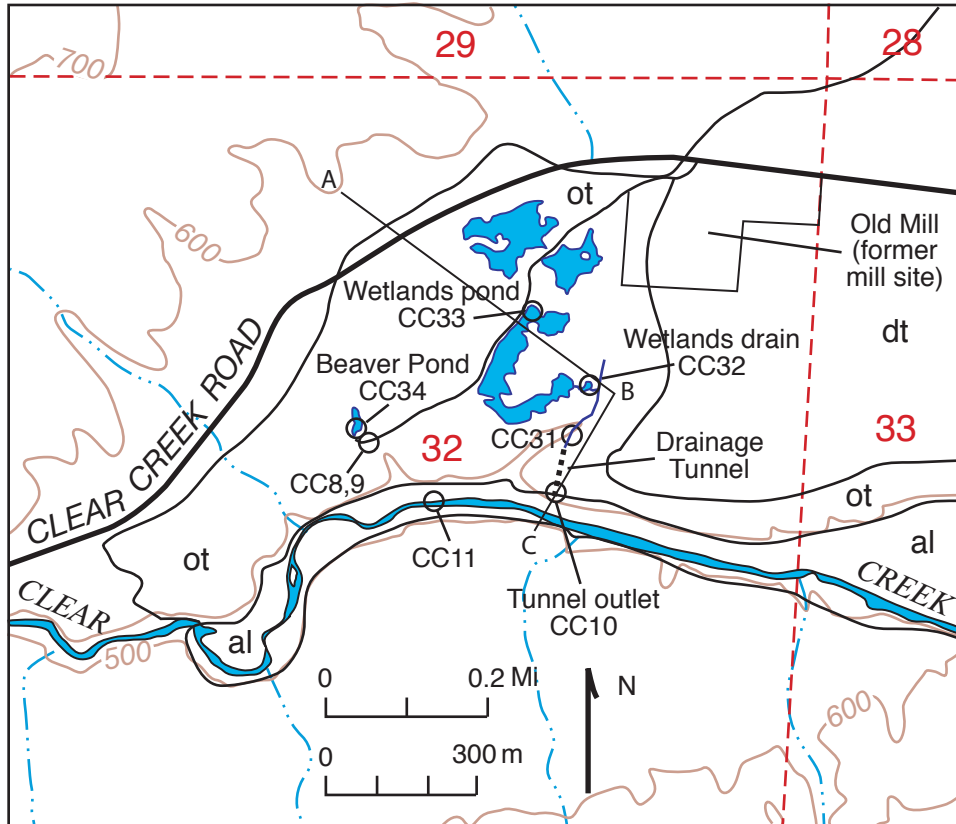
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related features including drainage cuts
and impoundments

Unassigned areas include alluvial terraces
of Quaternary age and bedrock units of
Tertiary and older age

Figure 3B. Central part of lower Clear Creek area, also known as the "Old Mill" area, showing constructed wetlands ponds and locations of sampling sites. Lines of schematic cross section (Fig. 12) shown by A-B-C.
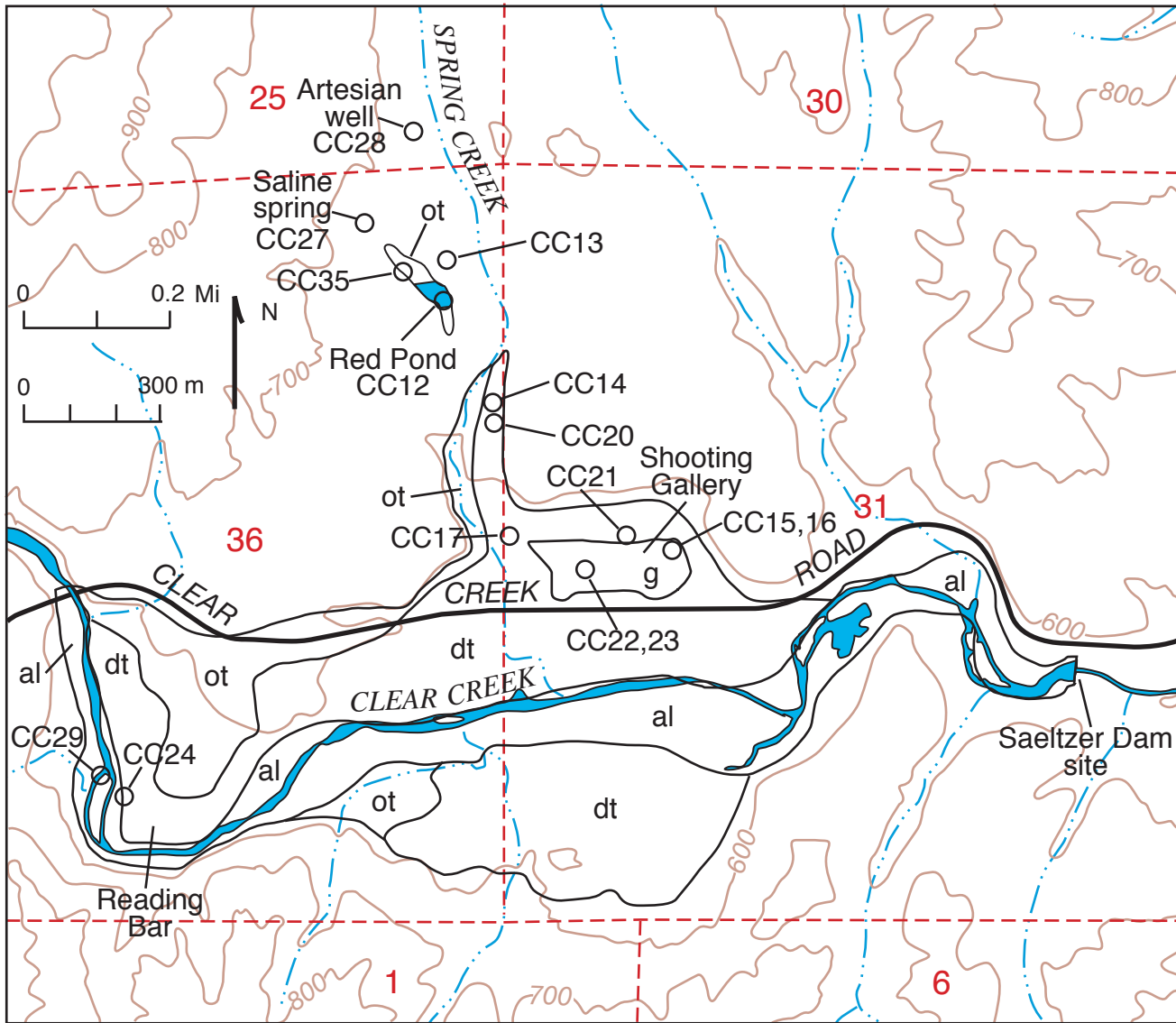
For base information see figure 2

Compiled by R. P. Ashley, 2004-2006

EXPLANATION OF MAP SYMBOLS

g   Areas of dredge tailings removed by
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related features including drainage cuts
and impoundments

       Unassigned areas include alluvial terraces
of Quaternary age and bedrock units of
Tertiary and older age

Figure 3C. Western part of lower Clear Creek area, also known as the Reading Bar area, showing locations of sampling sites.
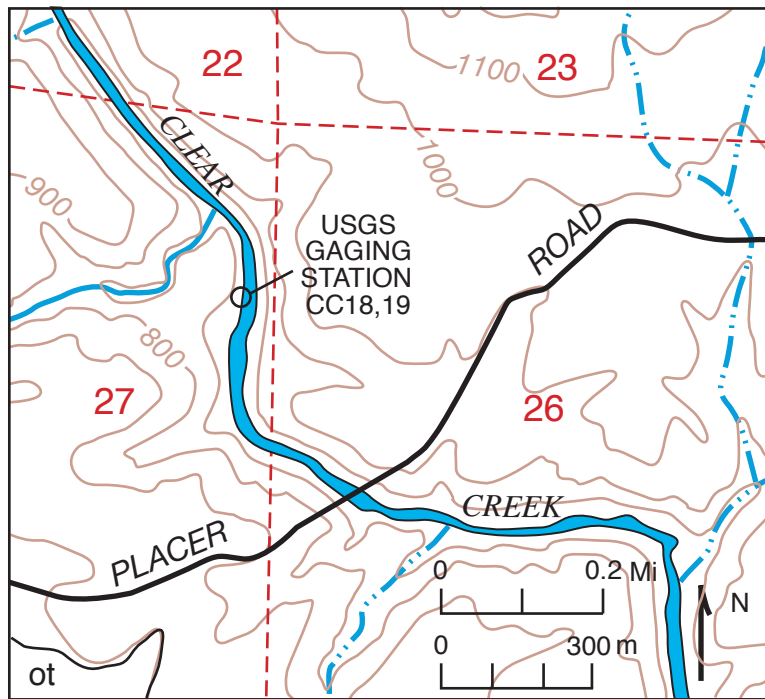
36

EXPLANATION OF MAP SYMBOLS

ot   Pre-dredging placer-mined areas.
        Includes placer tailings, bedrock surfaces
        exposed by mining, and other mining-
        related features including drainage cuts
        and impoundments

        Unassigned areas include alluvial terraces
        of Quaternary age and bedrock units of
        Tertiary and older age

Figure 3D. Clear Creek in the vicinity of U.S. Geological Igo Gaging Station, in the gorge reach, showing locations of sampling sites.

Figure 4. South Pond (site CC3), showing clouds of fine sediment from tailings used to fill the pond. At time of photo, tailings were being injected at a point immediately out of view to the left.

Figure 5. East Pond (site CC2), a marshy pond occupying a meander scar channel on the flood plain of lower Clear Creek.

Figure 6. Dragline dredge stacker tailings that were deposited in absence of a dredge pond or above the level of the dredge pond, Spring Creek (site CC14).
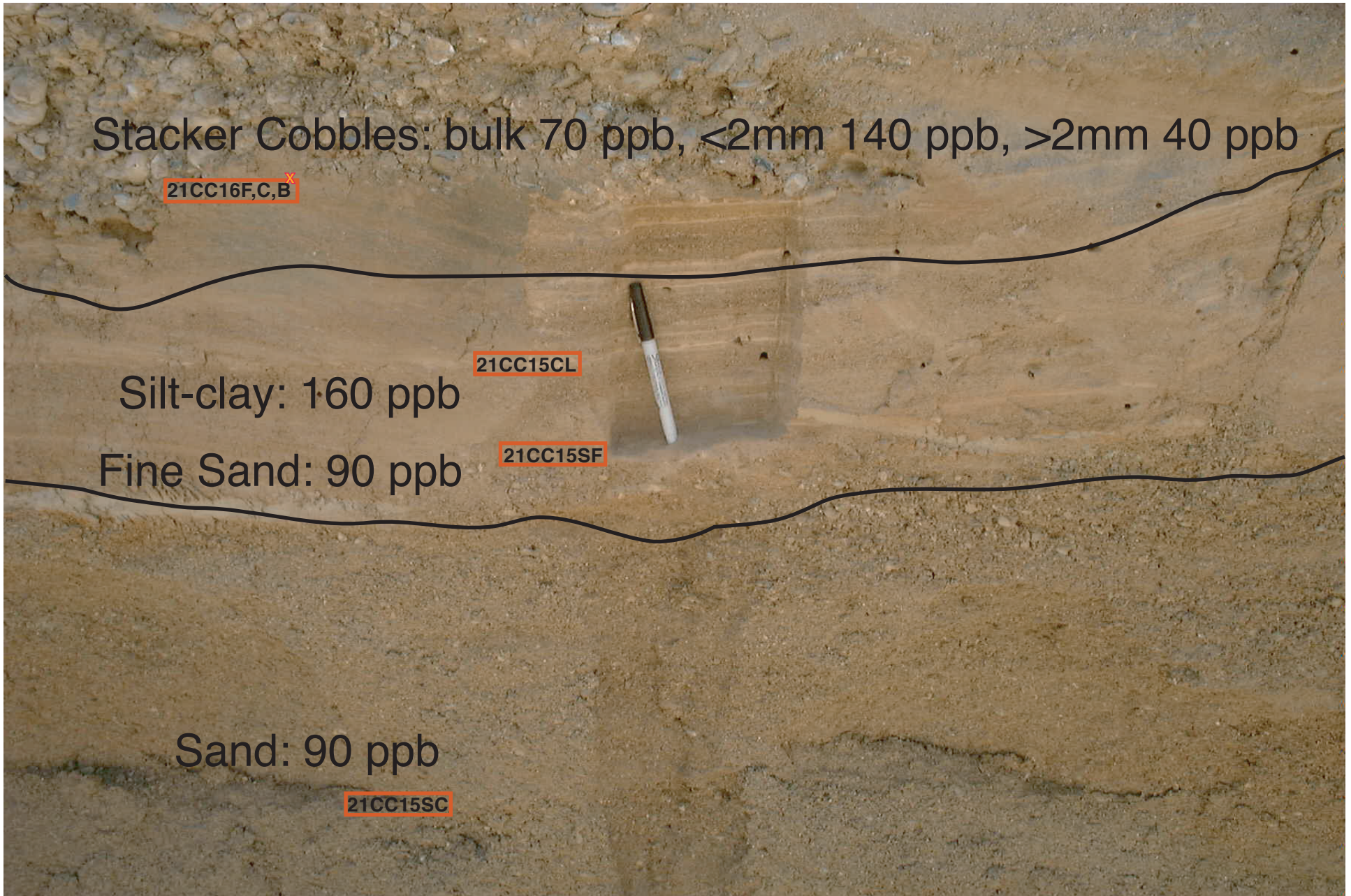
Figure 7. Dredge sluice tailings at site CC15-16 (sand, silt, and clay; 21CC15 samples), overlain by stacker tailings (pebbles and cobbles; sample 21CC16). Annotations show mercury concentrations of layers having various grain sizes, as noted; sample numbers shown in red boxes. Samples were cut from channel under the pen. These deposits represent tailings materials that were released into a dredge pond.

Figure 8. Red Pond (site CC12), an abandoned dredge pond fed by a saline spring. Iron oxyhydroxides precipitate on pond vegetation and bottom mud during summer evaporation, staining banks and vegetation below high-water line red.

Figure 9. Conglomerate of the Tehama Formation, exposed in prospect tunnel beneath Nomlaki Tuff, adjacent to Red Pond (site CC13).
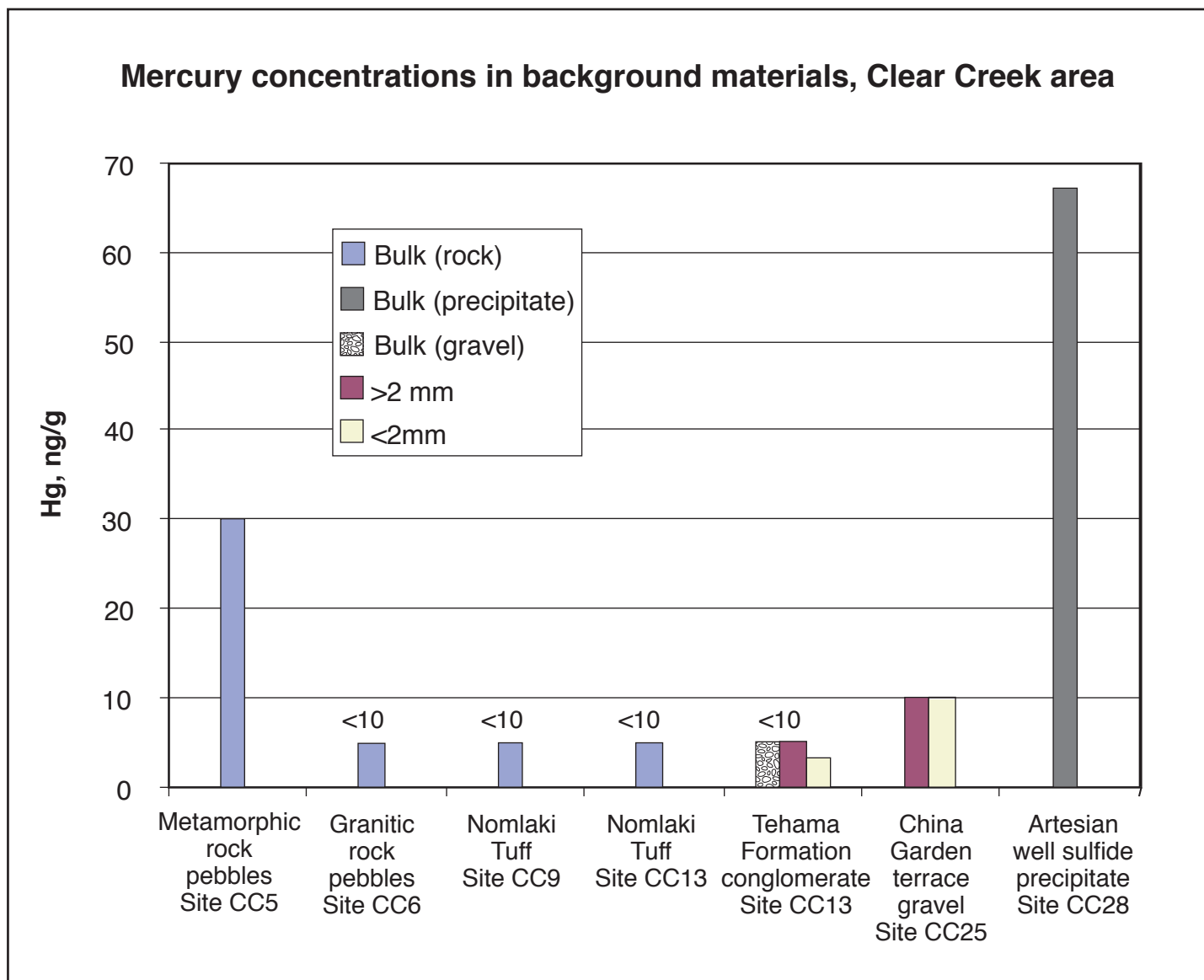
Figure 10. Total mercury concentrations in various geologic materials and units in the lower Clear Creek area. Tehama Formation and China Garden gravels are locally gold-bearing, and represent unmined auriferous materials.

Figure 11. Total mercury concentrations in placer mine tailings and sediments derived from placer tailings. The NOAA ERL shown at 150 ng/g Hg is a benchmark value below which there should be no noticeable effects on aquatic organisms, and the T_{25} at 230 ng/g is a value at which the probability of observing a toxic effect in aquatic organisms is 25 percent, determined from a logistic regression model; see text for further explanation.
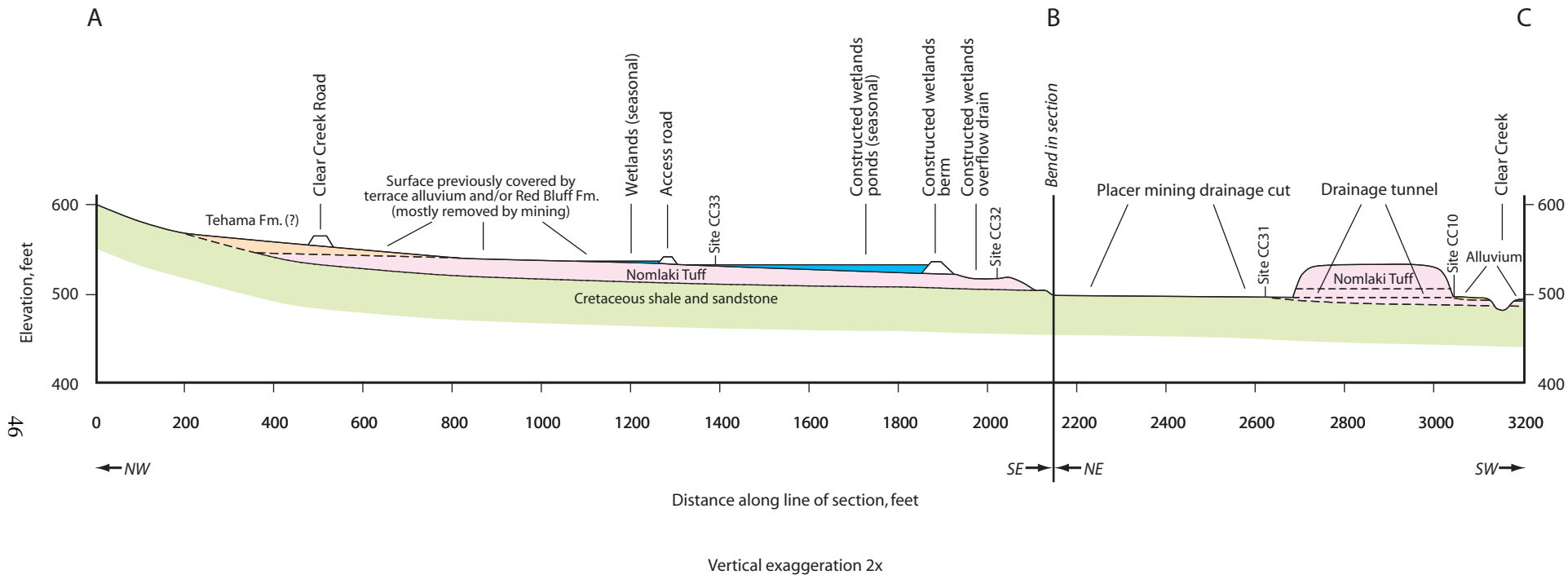
Figure 12. Schematic cross section of Old Mill placer mining area, showing geology and features related to placer mining and restoration.
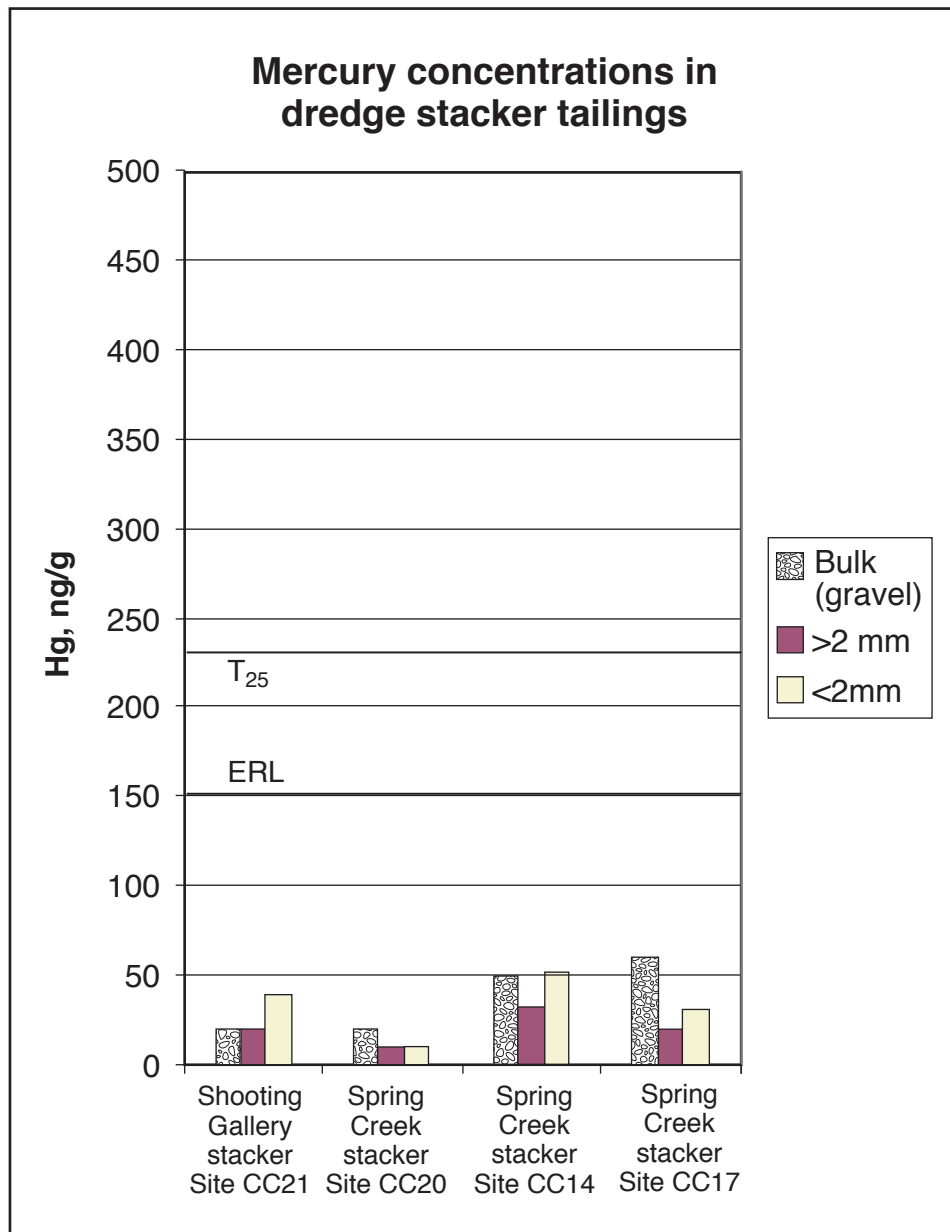
Figure 13A. Total mercury concentrations in dredge stacker tailings deposited above dredge pond water level, Shooting Gallery and Spring Creek areas.

Mercury concentrations in tailings deposited in dredge ponds
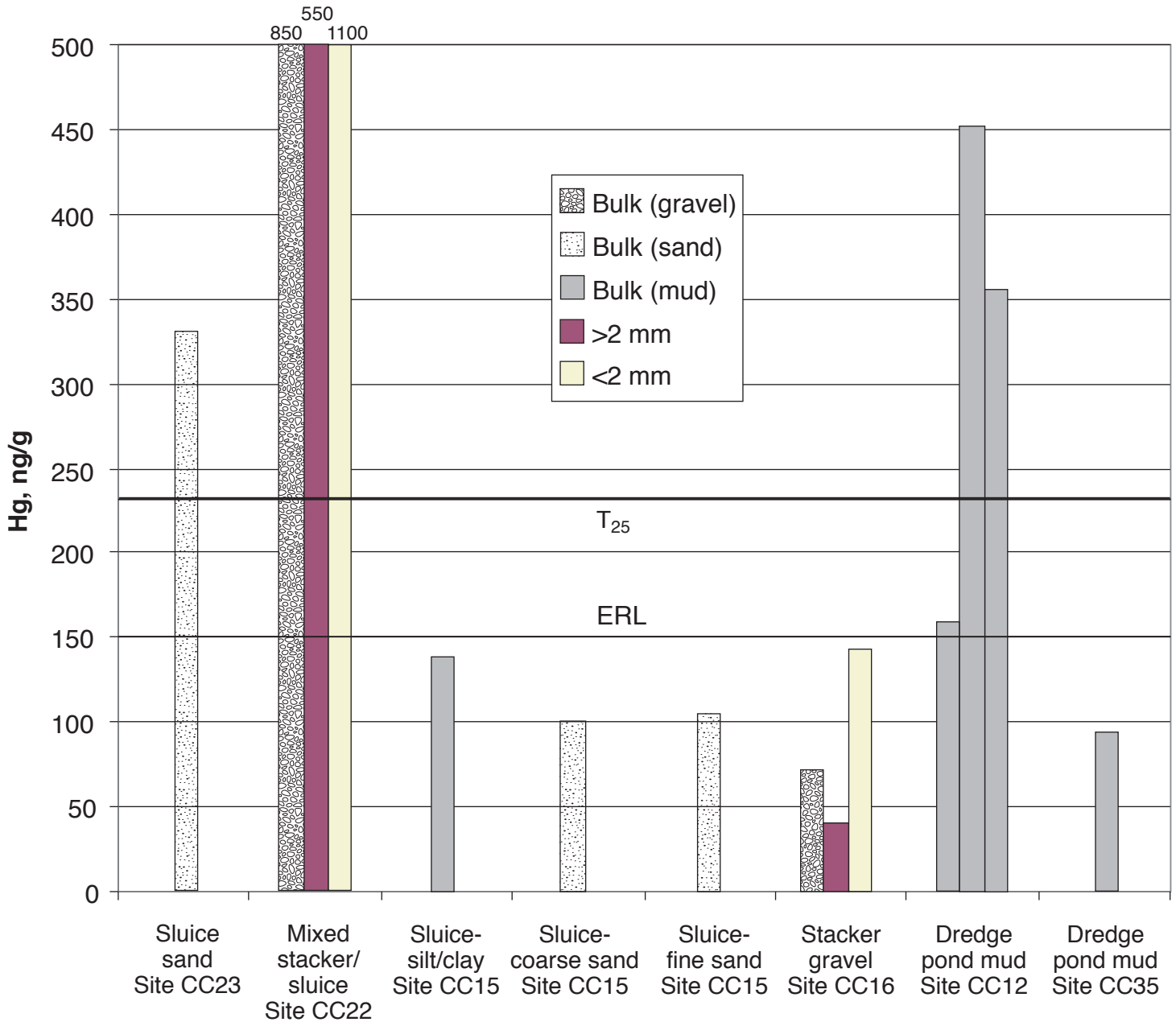
Figure 13B. Total mercury concentrations in dredge sluice sands, mixed stacker gravels and sluice sands, stacker gravels deposited below water level, and dredge pond muds (silt- and clay-size particles that settled out of the dredge pond water column). All sites are in the Shooting Callery area except sites CC12 and CC35, which are at Red Pond. The cluster of three site CC12 values represent samples taken in 2001, 2002, and 2003.
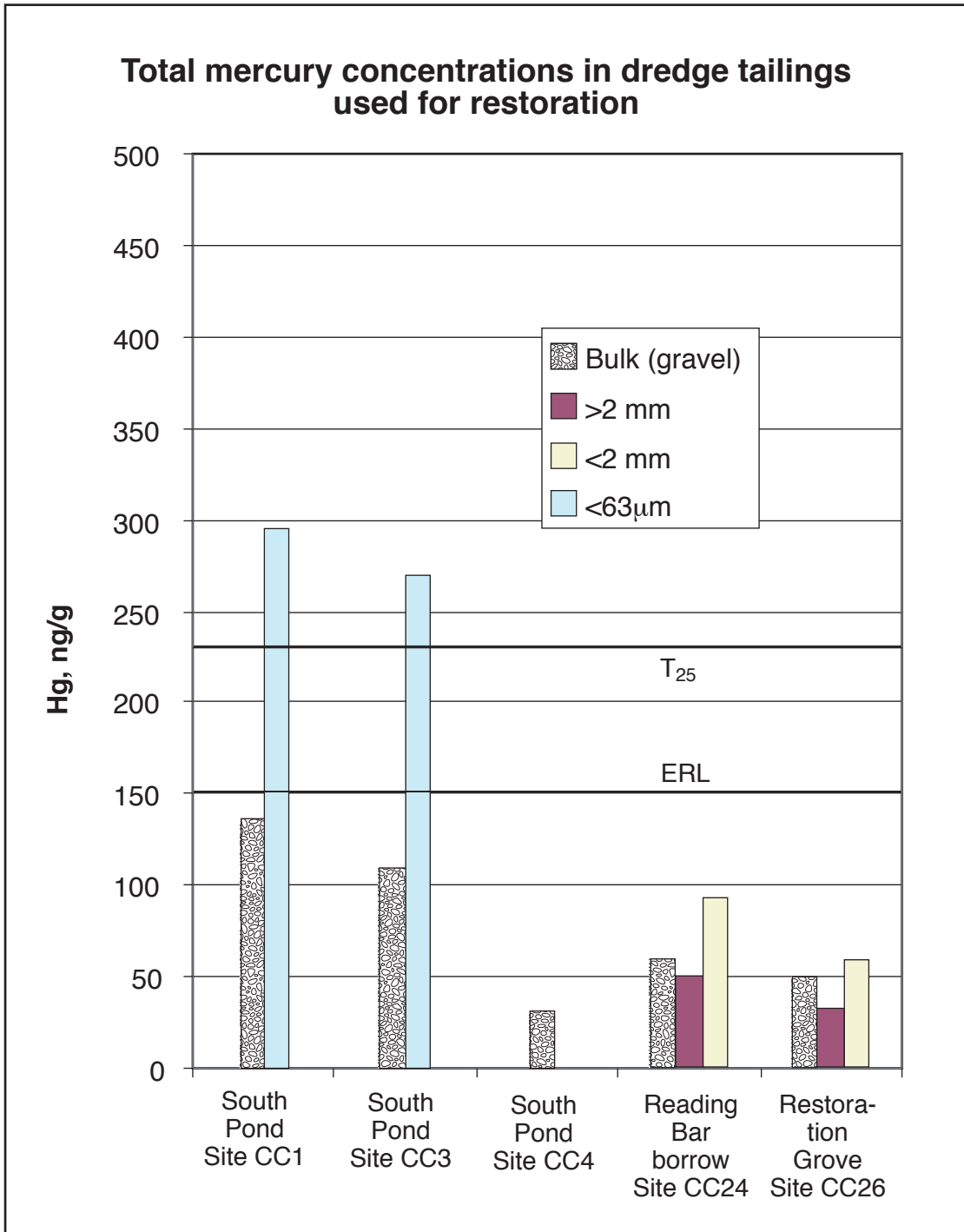
Figure 13C. Total mercury concentrations in dredge tailings used for restoration in the lower Clear Creek area.

Mercury concentrations in stream sediments
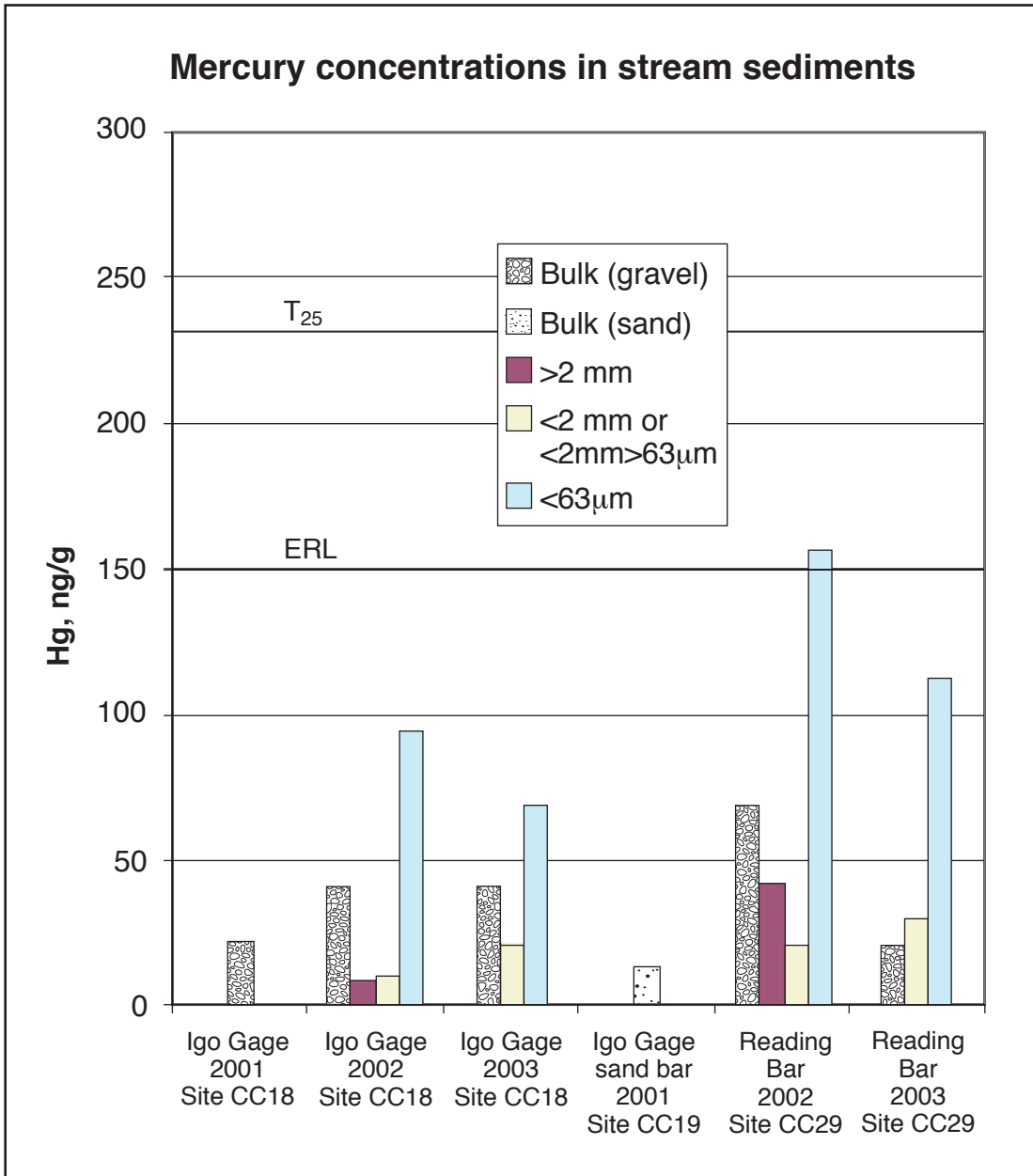
Figure 14A. Total mercury concentrations in Clear Creek stream sediments, western part of lower Clear Creek area.
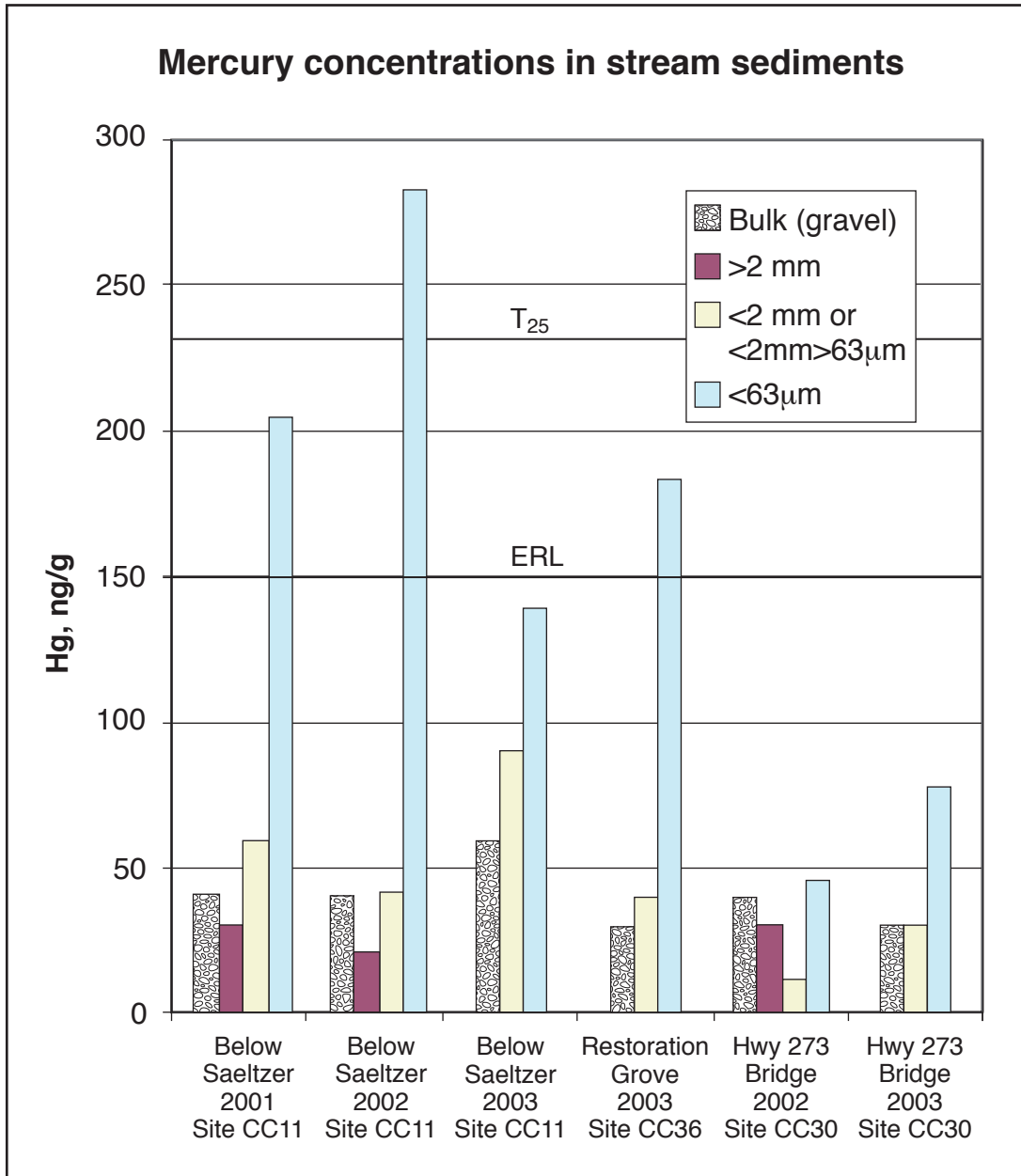
Figure 14B. Total mercury concentrations in Clear Creek stream sediments, central and eastern parts of lower Clear Creek area. Site CC11 is about 1.5 km downstream from the site of Saeltzer Dam, removed in 2000.

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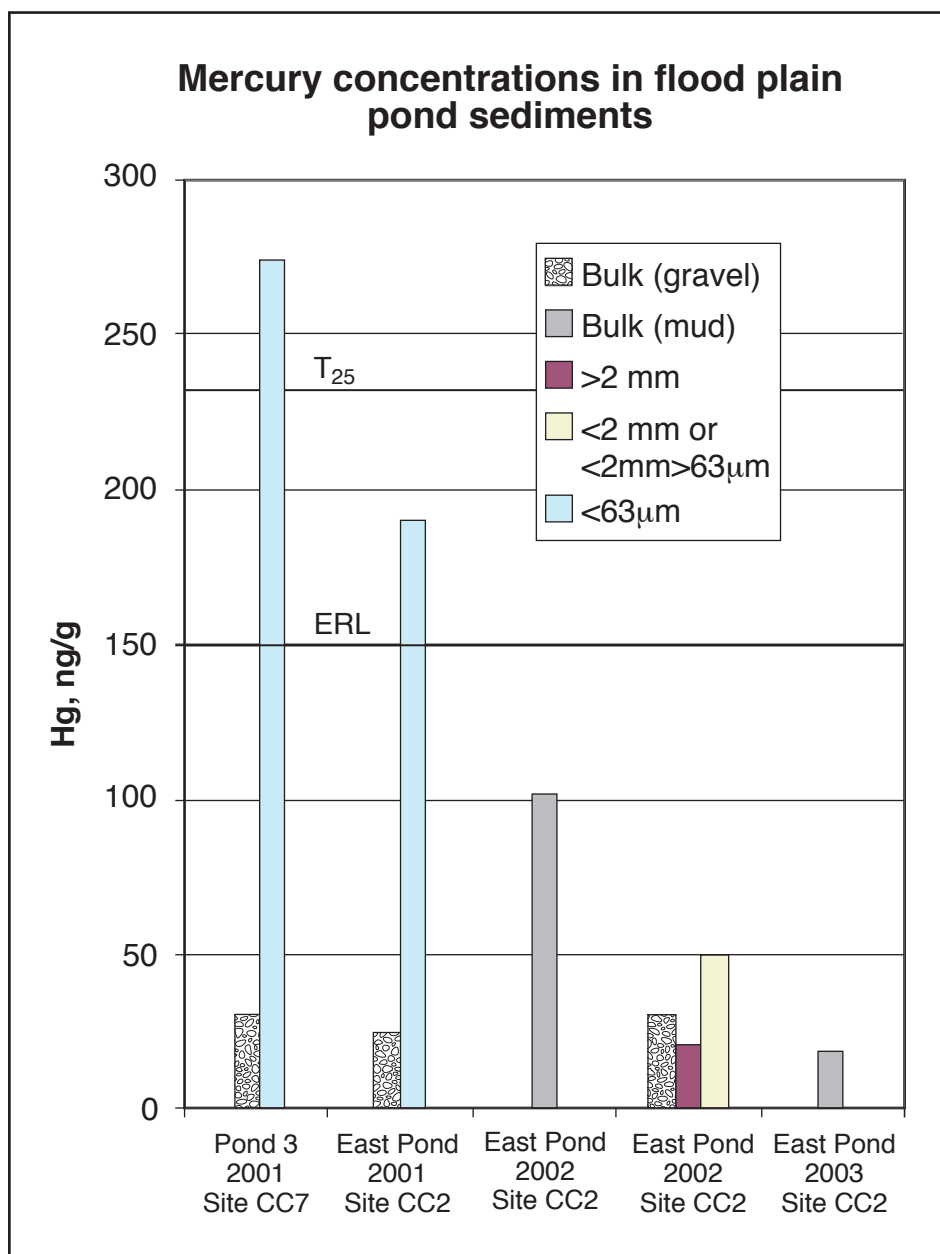
Figure 15. Total mercury concentrations in Clear Creek flood plain pond sediments.

Figure 16. Total mercury versus methylmercury concentrations in wet and dry sediments and tailings. Symbols with downward arrows represent maximum values (determinations below detection limits).

Total mercury, Chemex vs. Frontier results
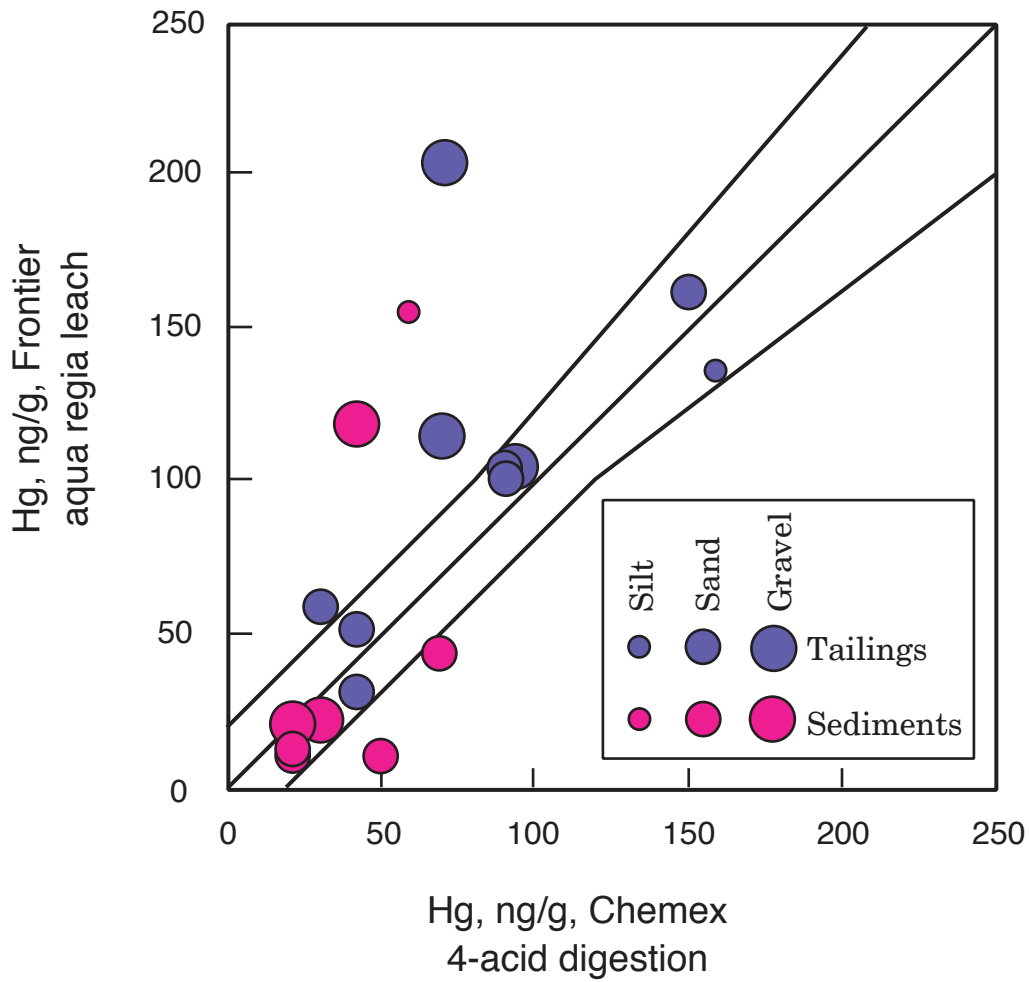
Figure 17. Concentrations of total mercury in sediments and tailings, comparison of results obtained by two different methods. See text for details.
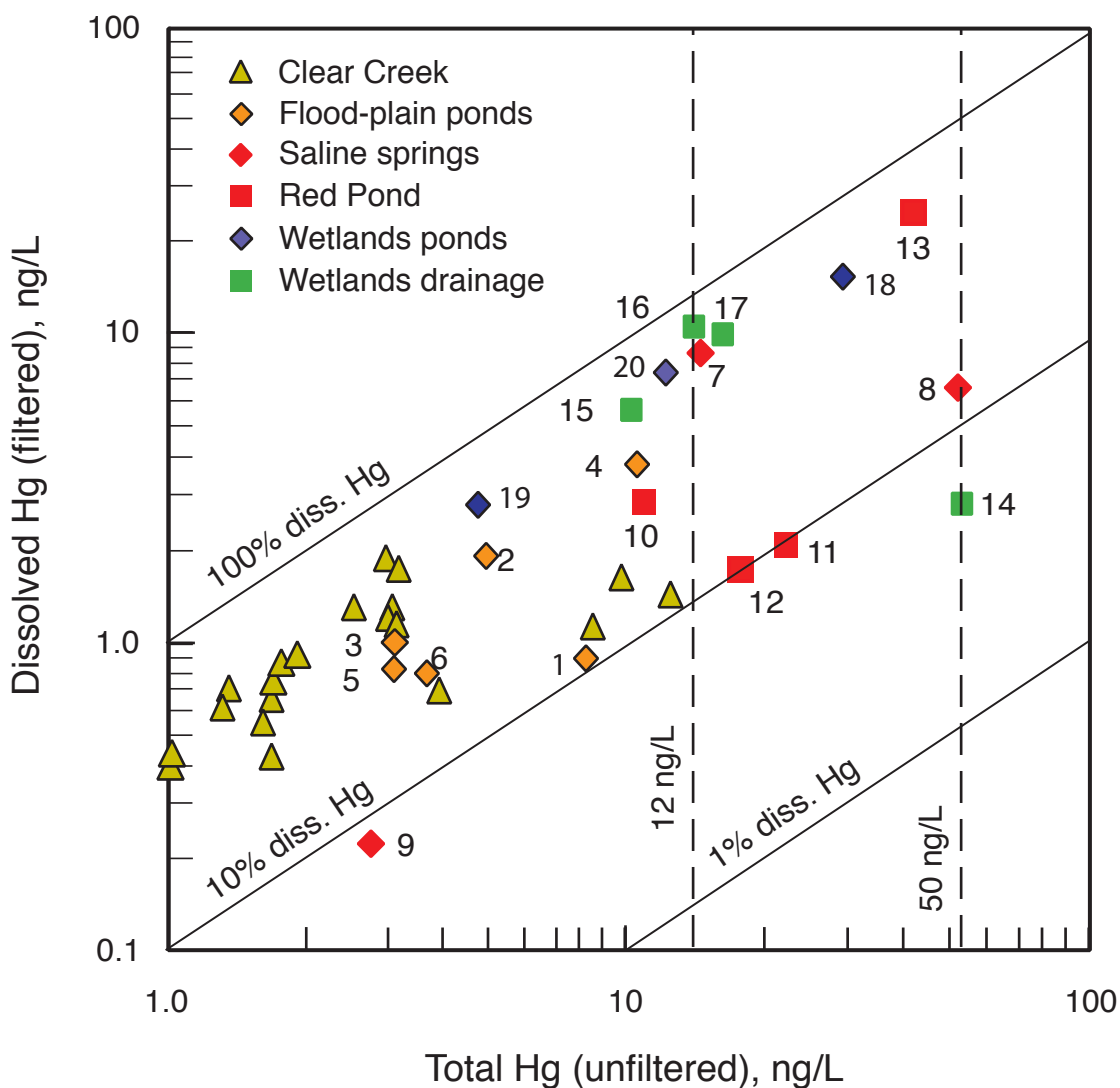
Figure 18. Total mercury versus dissolved mercury concentrations in waters. 1. East Pond, 8/27/01. 2. East Pond, 6/5/02. 3. East Pond, 7/29/03. 4. Pond 3, 8/28/01. 5. South Pond, 8/27/01. 6. South Pond, 8/28/01. 7. Saline spring near Red Pond, 6/4/02. 8. Saline spring near Red Pond, 7/28/03. 9. Saline well in upper Spring Creek drainage, 6/4/02. 10. Red Pond, 8/29/01. 11. Red Pond, 6/4/02. 12. Red Pond, 7/28/03. 13. Sinkhole near north end of Red Pond, 7/28/03. 14. Discharge at tunnel portal, Old Mill area, 8/28/01. 15. Discharge at tunnel portal, 6/5/02. 16. Discharge at tunnel portal, 1/29/03. 17. Drainage ditch above tunnel, 1/29/03. 18. Constructed wetlands overflow catchment (sump), 1/29/03. 19. Constructed wetlands pond, 1/29/03. 20. Beaver Pond, 1/29/03.

Turbidity vs. total and dissolved mercury
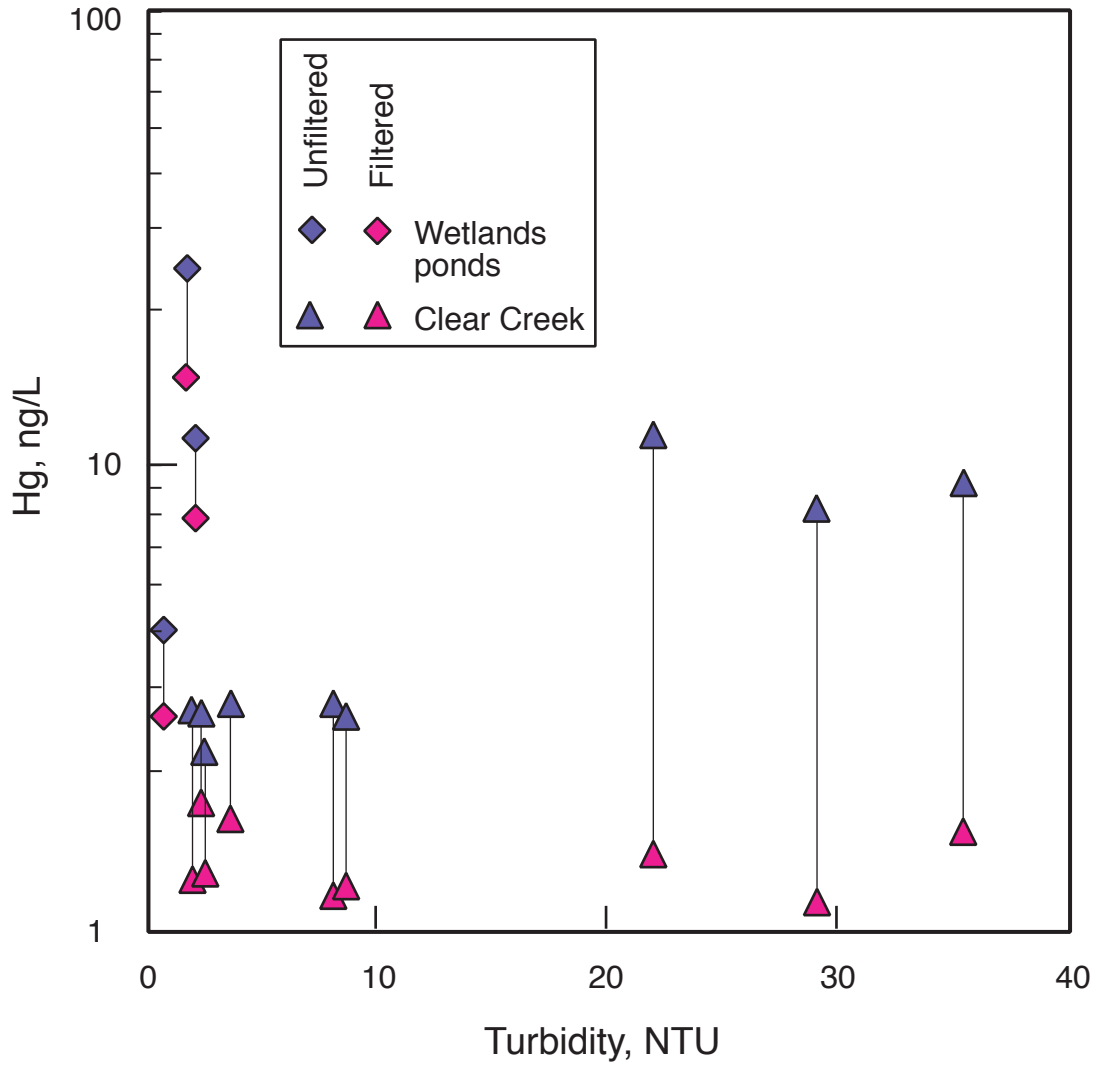
Figure 19. Turbidity in Nephelometric Turbidity Units (NTU) versus unfiltered (total) and filtered (dissolved) mercury concentrations for Clear Creek and constructed wetlands ponds. The tie lines connect values for unfiltered and filtered subsamples for each sample.
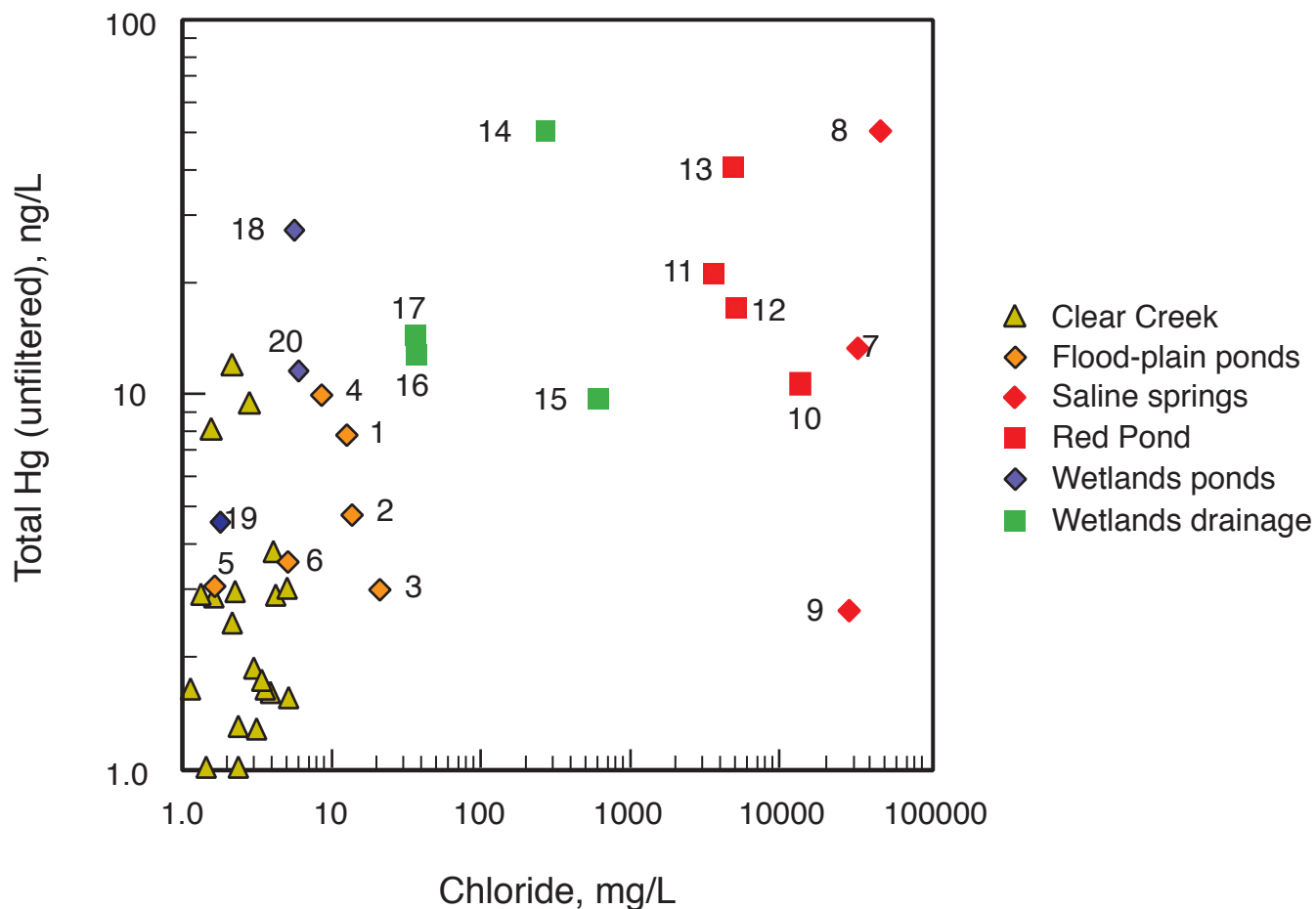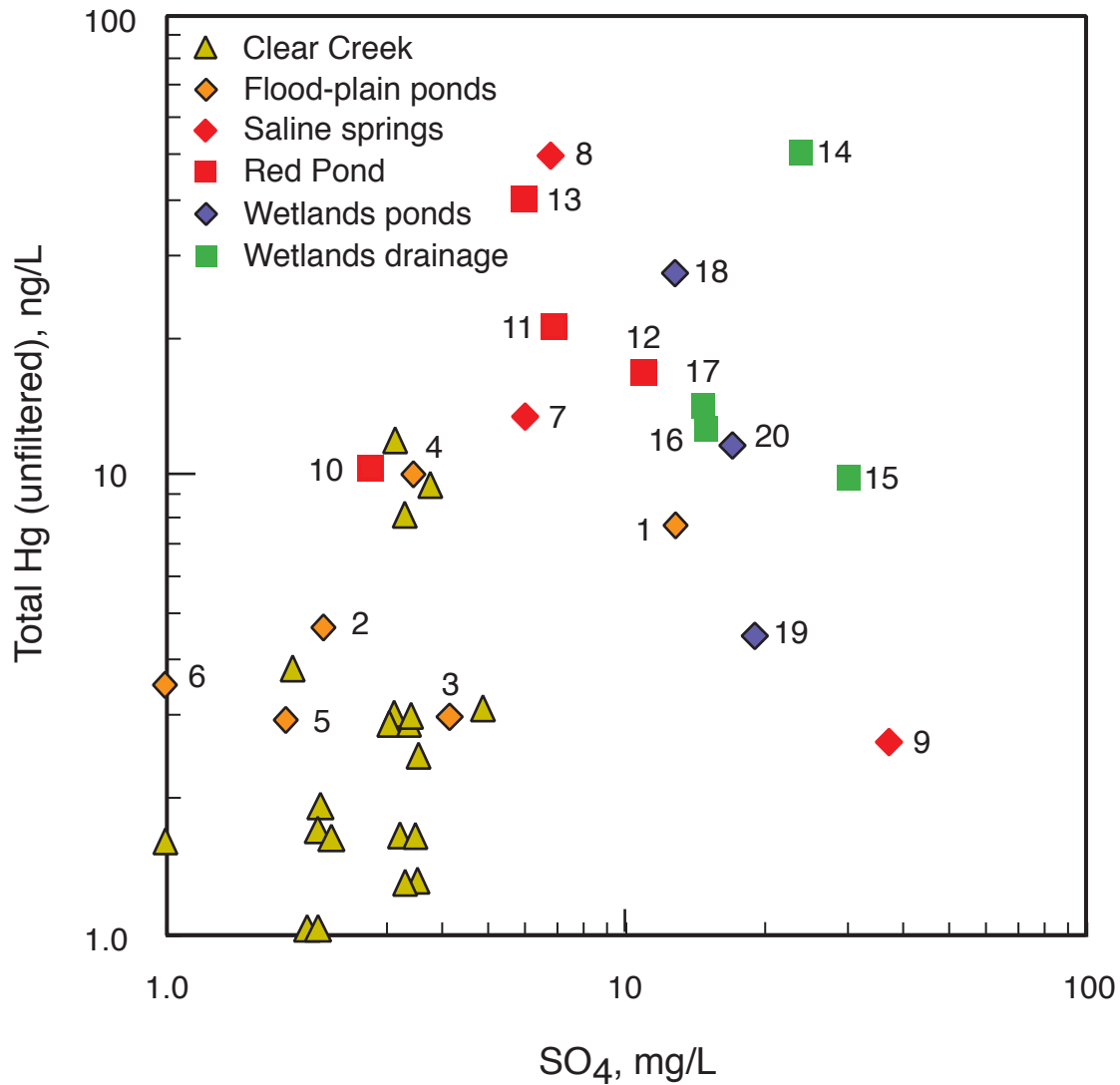
Figure 20. Chloride versus total mercury concentrations in waters. 1. East Pond, 8/27/01. 2. East Pond, 6/5/02. 3. East Pond, 7/29/03. 4. Pond 3, 8/28/01. 5. South Pond, 8/27/01. 6. South Pond, 8/28/01. 7. Saline spring near Red Pond, 6/4/02. 8. Saline spring near Red Pond, 7/28/03. 9. Saline well in upper Spring Creek drainage, 6/4/02. 10. Red Pond, 8/29/01. 11. Red Pond, 6/4/02. 12. Red Pond, 7/28/03. 13. Sinkhole near north end of Red Pond, 7/28/03. 14. Discharge at tunnel portal, Old Mill area, 8/28/01. 15. Discharge at tunnel portal, 6/5/02. 16. Discharge at tunnel portal, 1/29/03. 17. Drainage ditch above tunnel, 1/29/03. 18. Constructed wetlands overflow catchment (sump), 1/29/03. 19. Constructed wetlands pond, 1/29/03. 20. Beaver Pond, 1/29/03.
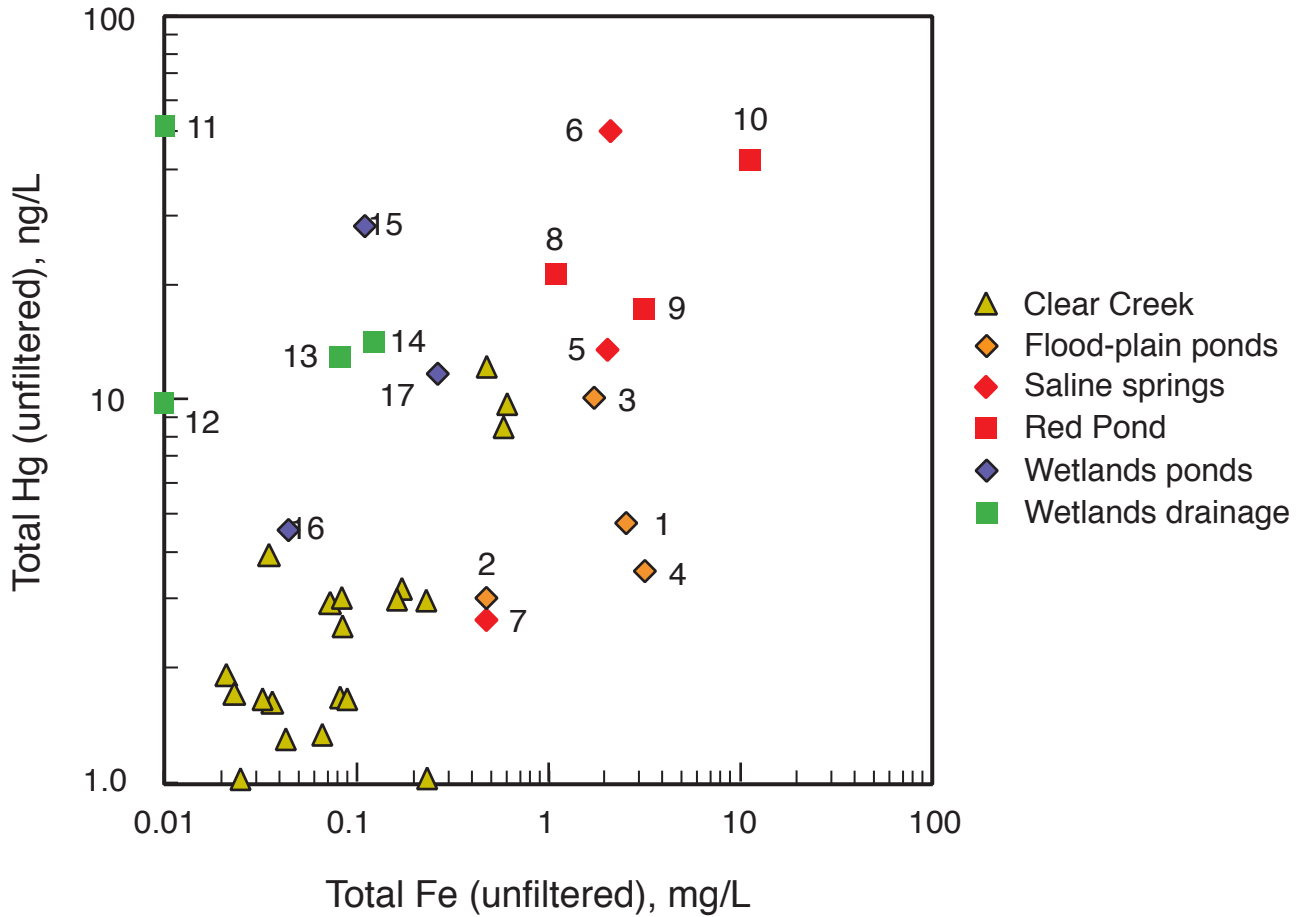
Figure 21. Sulfate versus total mercury concentrations in waters. 1. East Pond, 8/27/01. 2. East Pond, 6/5/02. 3. East Pond, 7/29/03. 4. Pond 3, 8/28/01. 5. South Pond, 8/27/01. 6. South Pond, 8/28/01. 7. Saline spring near Red Pond, 6/4/02. 8. Saline spring near Red Pond, 7/28/03. 9. Saline well in upper Spring Creek drainage, 6/4/02. 10. Red Pond, 8/29/01. 11. Red Pond, 6/4/02. 12. Red Pond, 7/28/03. 13. Sinkhole near north end of Red Pond, 7/28/03. 14. Discharge at tunnel portal, Old Mill area, 8/28/01. 15. Discharge at tunnel portal, 6/5/02. 16. Discharge at tunnel portal, 1/29/03. 17. Drainage ditch above tunnel, 1/29/03. 18. Constructed wetlands overflow catchment (sump), 1/29/03. 19. Constructed wetlands pond, 1/29/03. 20. Beaver Pond, 1/29/03.

Figure 22. Total iron (unfiltered, by ICP-AES) versus total mercury concentrations in waters. 1. East Pond, 6/5/02. 2. East Pond, 7/29/03. 3. Pond 3, 8/28/01. 4. South Pond, 8/28/01. 5. Saline spring near Red Pond, 6/4/02. 6. Saline spring near Red Pond, 7/28/03. 7. Saline well in upper Spring Creek drainage, 6/4/02. 8. Red Pond, 6/4/02. 9. Red Pond, 7/28/03. 10. Sinkhole near north end of Red Pond, 7/28/03. 11. Discharge at tunnel portal, Old Mill area, 8/28/01. 12. Discharge at tunnel portal, 6/5/02. 13. Discharge at tunnel portal, 1/29/03. 14. Drainage ditch above tunnel, 1/29/03. 15. Constructed wetlands overflow catchment (sump), 1/29/03. 16. Constructed wetlands pond, 1/29/03. 17. Beaver Pond, 1/29/03.

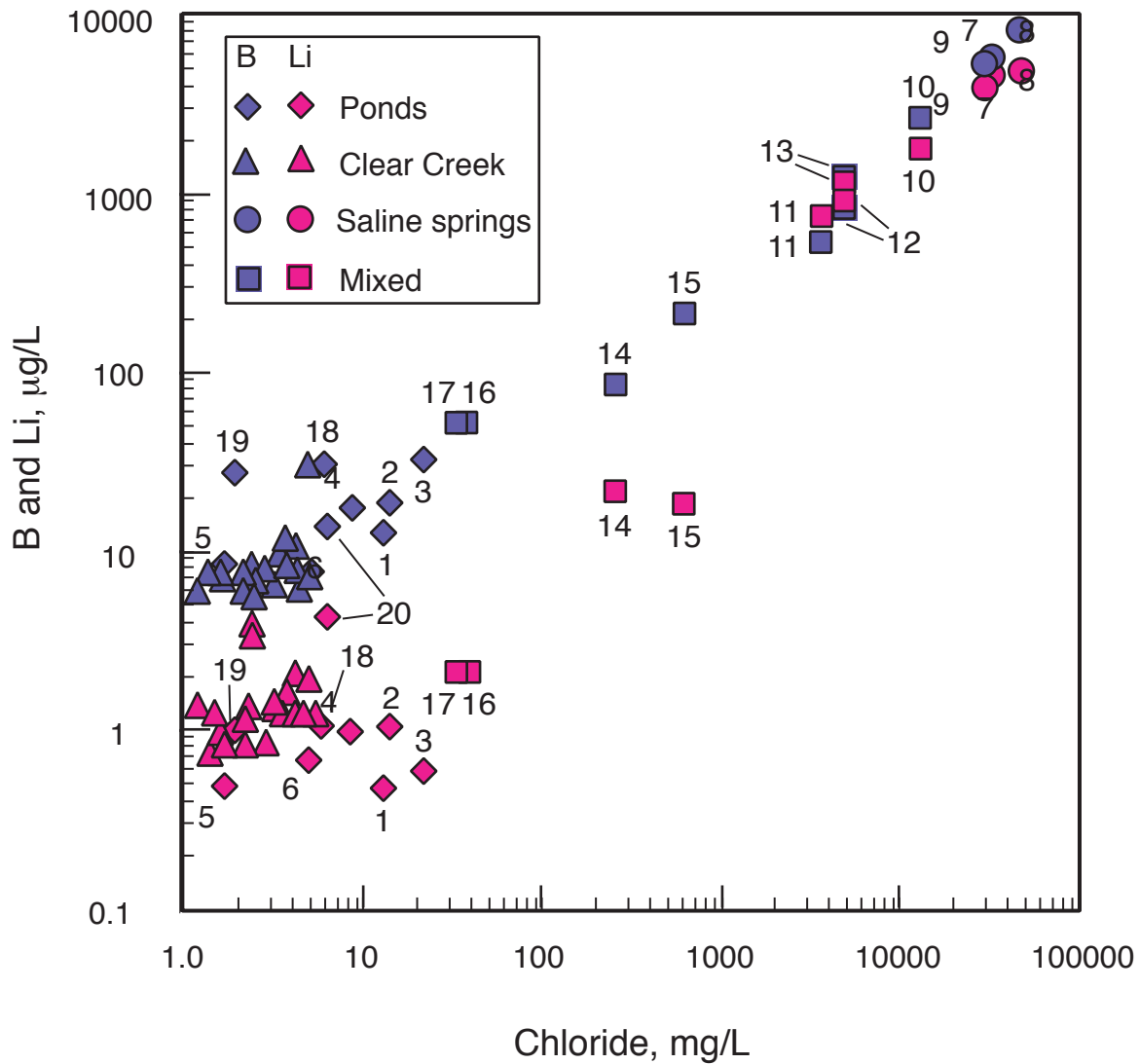# Chloride vs. lithium and boron concentrations in waters

Figure 23A. Log-log plot of chloride versus lithium and boron concentrations in waters. 1. East Pond, 8/27/01. 2. East Pond, 6/5/02. 3. East Pond, 7/29/03. 4. Pond 3, 8/28/01. 5. South Pond, 8/27/01. 6. South Pond, 8/28/01. 7. Saline spring near Red Pond, 6/4/02. 8. Saline spring near Red Pond, 7/28/03. 9. Saline well in upper Spring Creek drainage, 6/4/02. 10. Red Pond, 8/29/01. 11. Red Pond, 6/4/02. 12. Red Pond, 7/28/03. 13. Sinkhole near north end of Red Pond. 14. Discharge at tunnel portal, Old Mill area, 8/28/01. 15. Discharge at tunnel portal, 6/5/02. 16. Discharge at tunnel portal, 1/29/03. 17. Drainage ditch above tunnel, 1/29/03. 18. Constructed wetlands overflow catchment (sump), 1/29/03. 19. Constructed wetlands pond, 1/29/03. 20. Beaver Pond, 1/29/03.

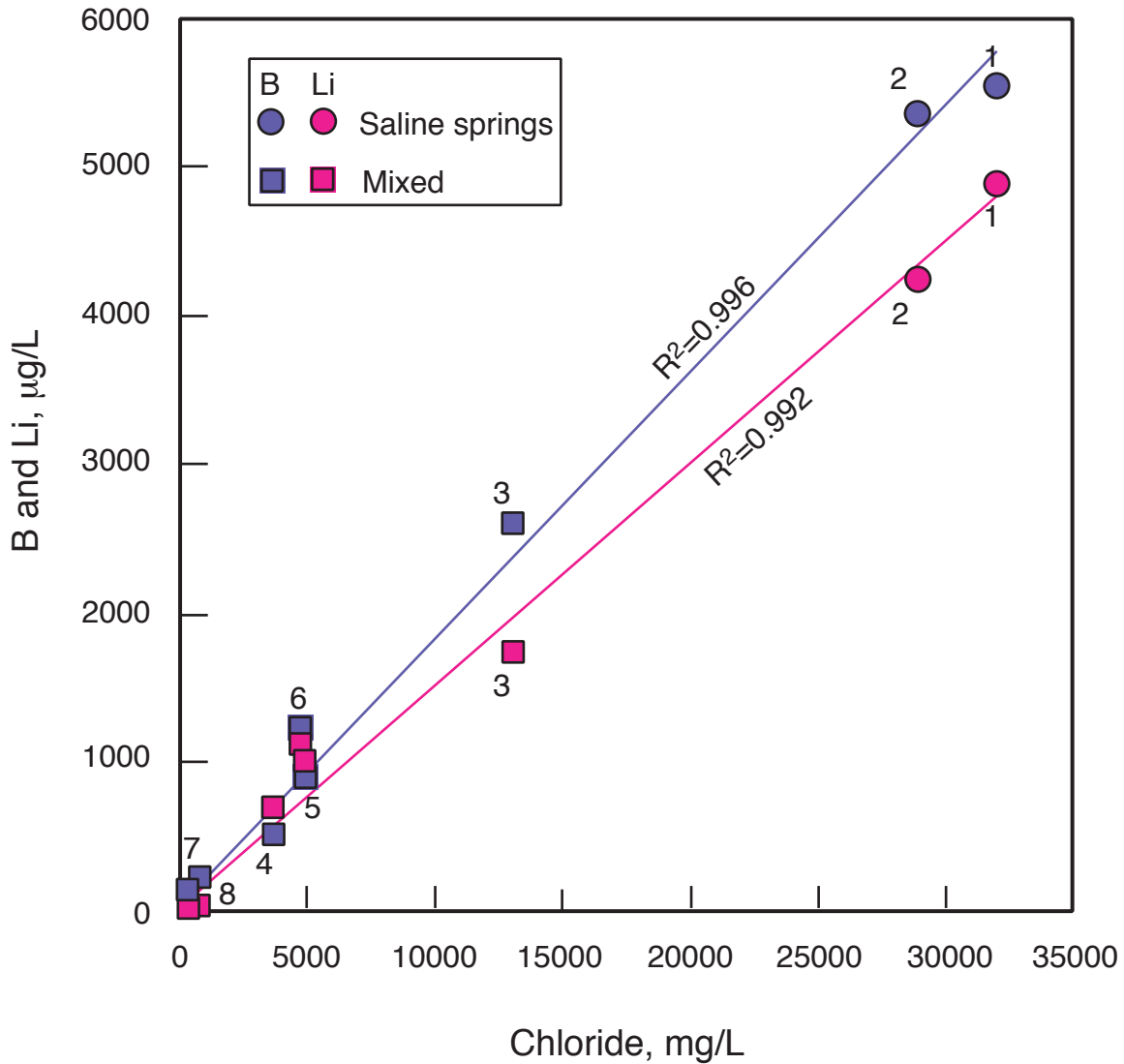## Mixing lines for boron-chloride and lithium-chloride in waters

Figure 23B. Regression lines for boron-chloride and lithium-chloride concentrations in waters. The regressions include values for Clear Creek, pond waters, and some Old Mill area drainage waters that plot near the origin (not shown). Saline spring near Red Pond on 7/28/03 is excluded because lithium was preferentially depleted owing to local precipitation of salt. 1. Saline spring near Red Pond, 6/4/02. 2. Saline well in upper Spring Creek drainage, 6/4/02. 3. Red Pond, 8/29/01. 4. Red Pond, 6/4/02. 5. Red Pond, 7/28/03. 6. Sinkhole near north end of Red Pond. 7. Discharge at tunnel portal, Old Mill area, 8/28/01. 8. Discharge at tunnel portal, 6/5/02.
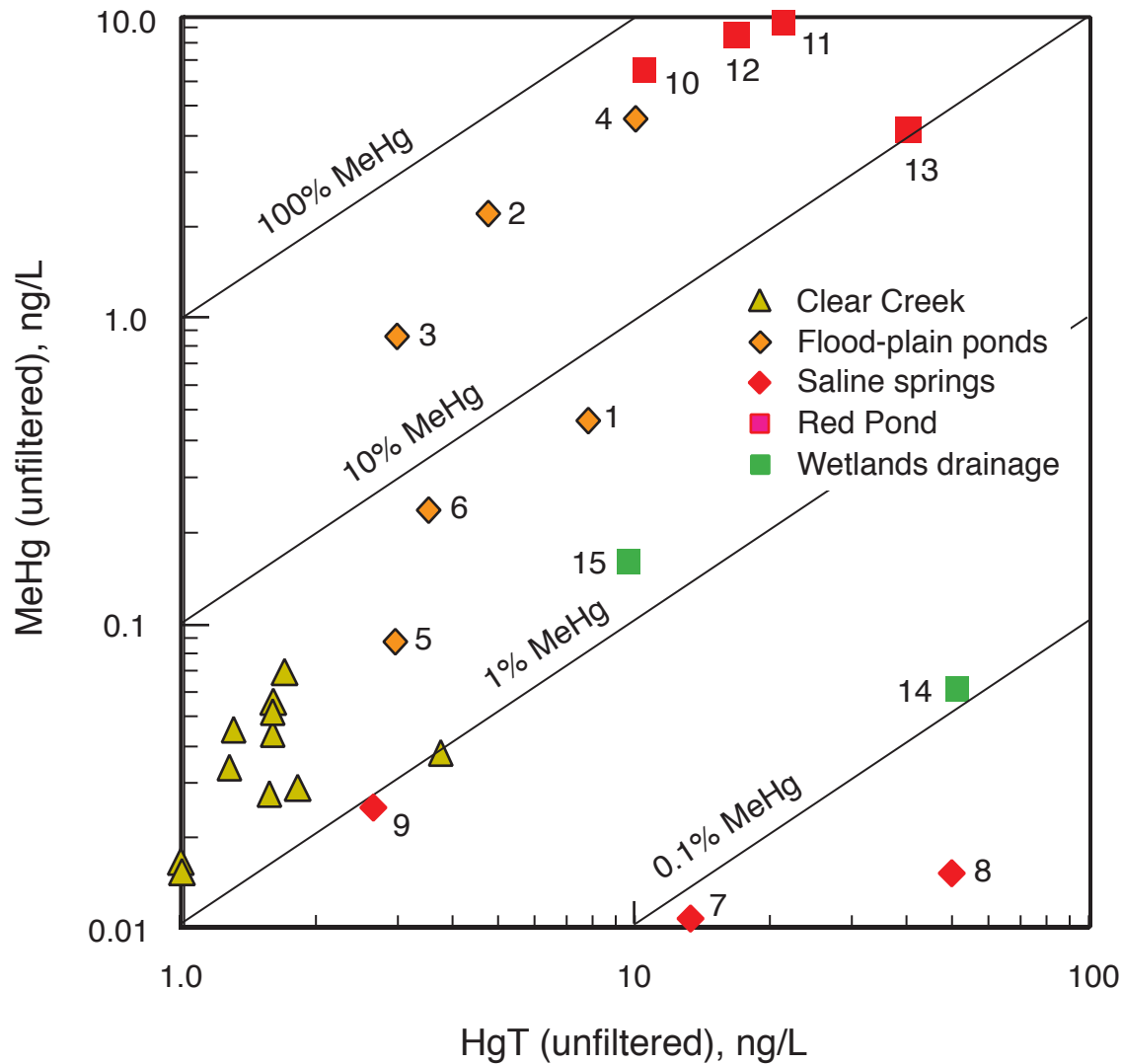
Figure 24. Total mercury versus methylmercury concentrations in waters. 1. East Pond, 8/27/01. 2. East Pond, 6/5/02. 3. East Pond, 7/29/03. 4. Pond 3, 8/28/01. 5. South Pond, 8/27/01. 6. South Pond, 8/28/01. 7. Saline spring near Red Pond, 6/4/02. 8. Saline spring near Red Pond, 7/28/03. 9. Saline well in upper Spring Creek drainage, 6/4/02. 10. Red Pond, 8/29/01. 11. Red Pond, 6/4/02. 12. Red Pond, 7/28/03. 13. Sinkhole near north end of Red Pond, 7/28/03. 14. Discharge at tunnel portal, Old Mill area, 8/28/01. 15. Discharge at tunnel portal, 6/5/02.
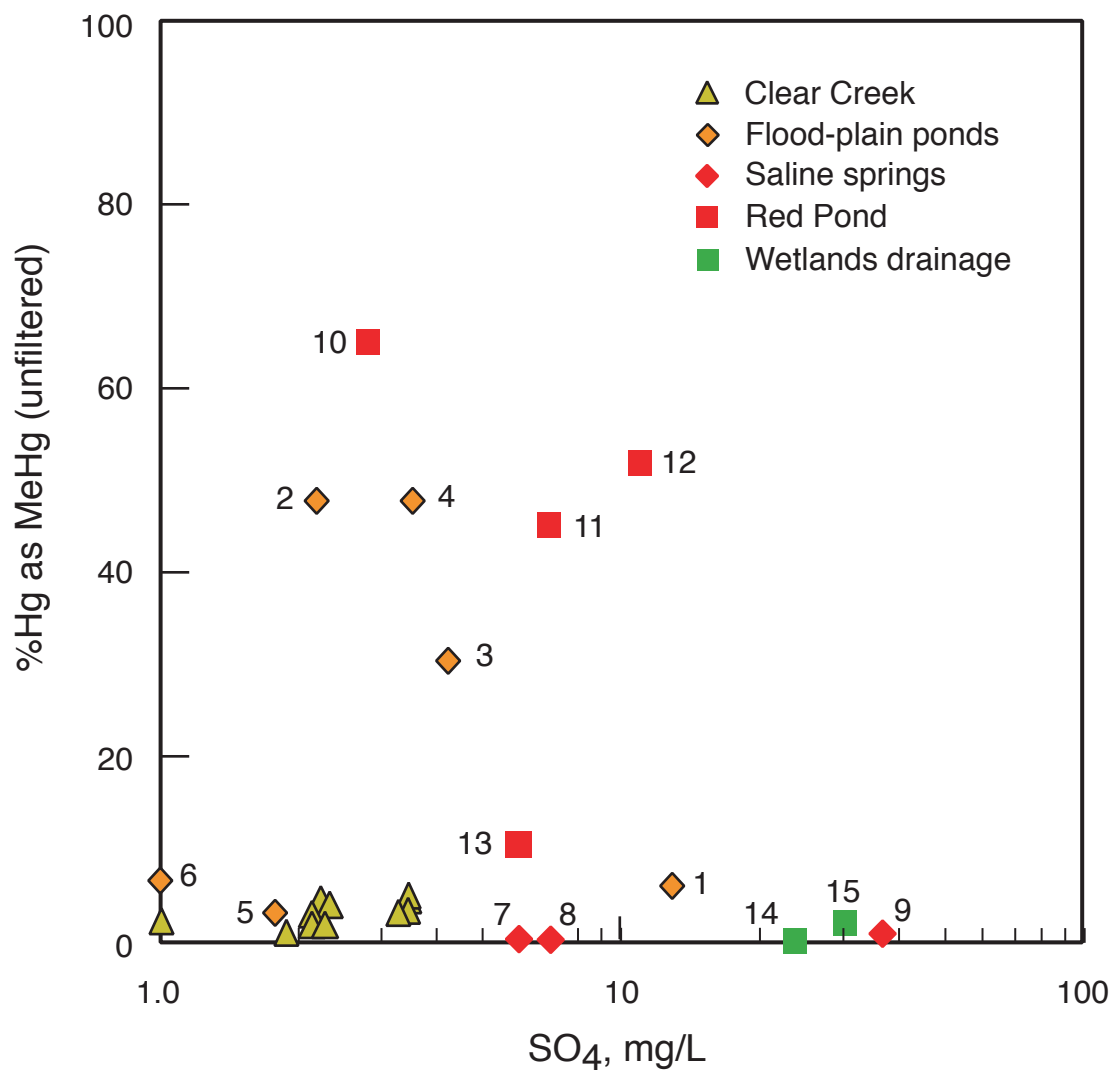
Figure 25A. Sulfate versus percentage of mercury present as methylmercury in waters. 1. East Pond, 8/27/01. 2. East Pond, 6/5/02. 3. East Pond, 7/29/03. 4. Pond 3, 8/28/01. 5. South Pond, 8/27/01. 6. South Pond, 8/28/01. 7. Saline spring near Red Pond, 6/4/02. 8. Saline spring near Red Pond, 7/28/03. 9. Saline well in upper Spring Creek drainage, 6/4/02. 10. Red Pond, 8/29/01. 11. Red Pond, 6/4/02. 12. Red Pond, 7/28/03. 13. Sinkhole near north end of Red Pond, 7/28/03. 14. Discharge at tunnel portal, Old Mill area, 8/28/01. 15. Discharge at tunnel portal, 6/5/02.

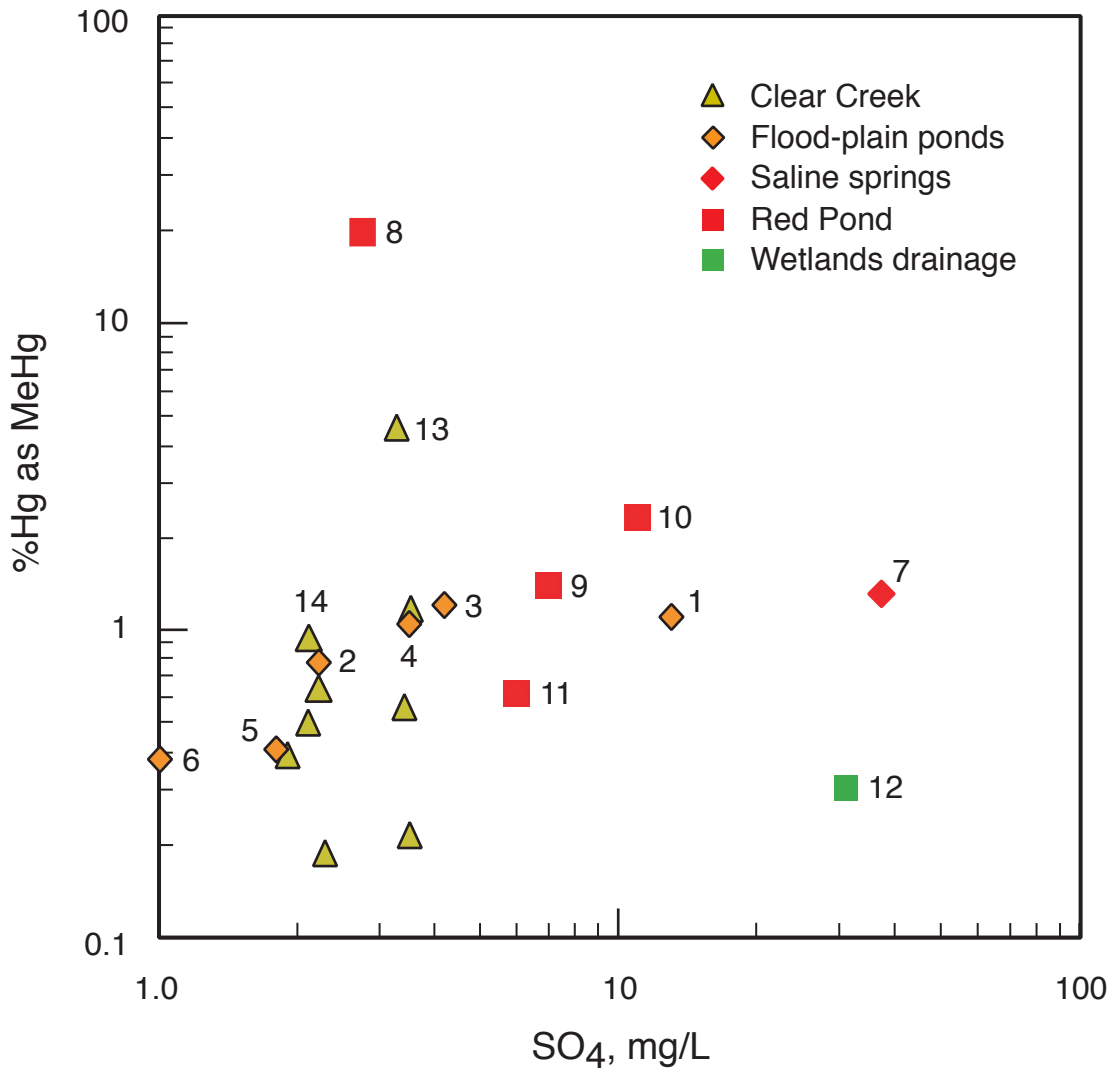**Sulfate in waters vs. percent mercury as methylmercury in sediments**

Figure 25B. Sulfate in water column versus percentage of mercury present as methylmercury in subjacent fine-grained sediments (<63μm or bulk mud). 1. East Pond, 8/27/01. 2. East Pond, 6/5/02. 3. East Pond, 7/29/03. 4. Pond 3, 8/28/01. 5. South Pond, 8/27/01. 6. South Pond, 8/28/01. 7. Saline well in upper Spring Creek drainage, 6/4/02. 8. Red Pond, 8/29/01. 9. Red Pond, 6/4/02. 10. Red Pond, 7/28/03. 11. Sinkhole near north end of Red Pond, 7/28/03. 12. Discharge at tunnel portal, 6/5/02. 13. Backwater on Clear Creek at Igo Gauge, 6/5/02. 14. Backwater on Clear Creek at Igo Gauge, 7/29/03.

Total organic carbon vs. percent mercury as methylmercury in waters
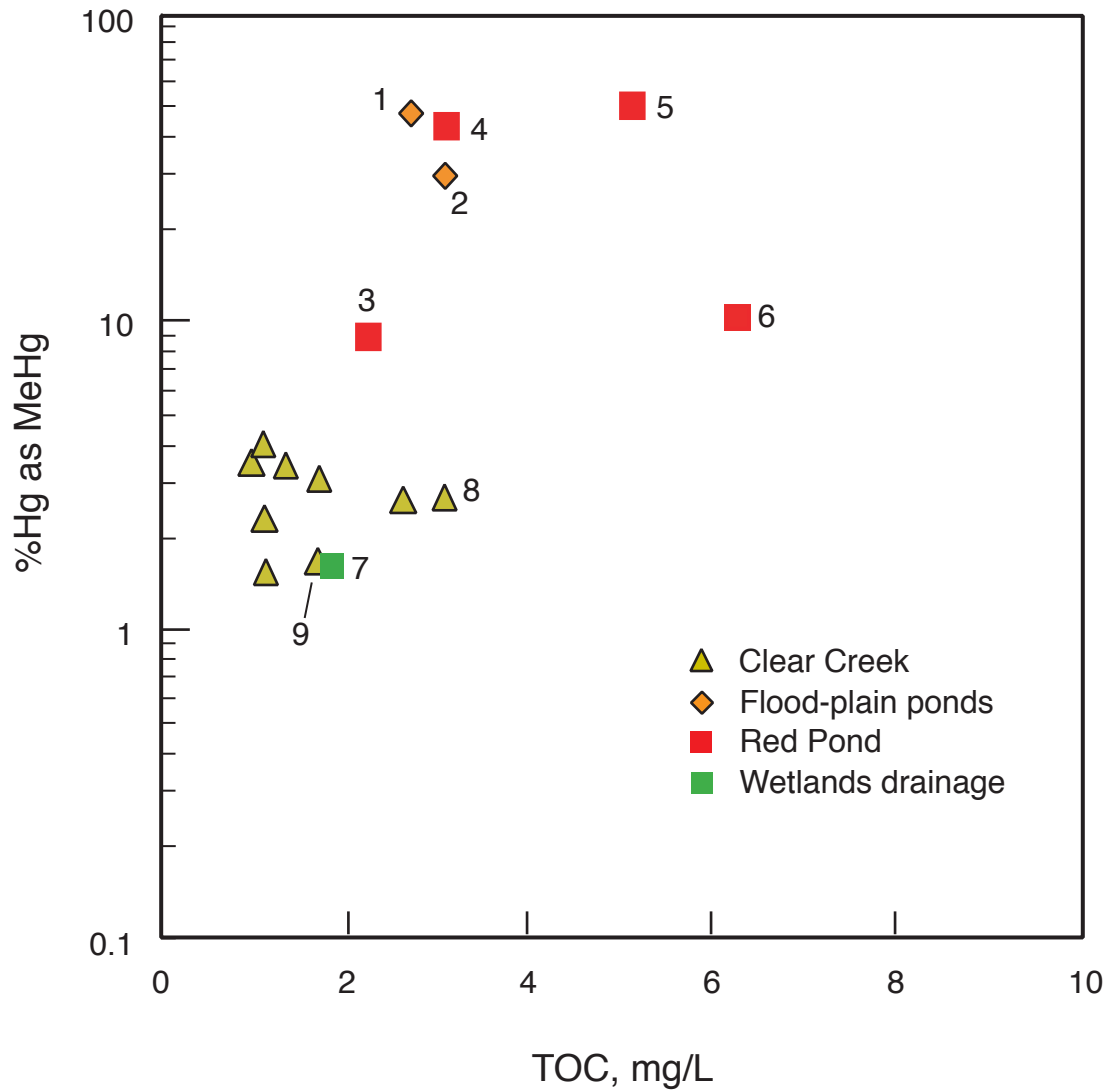
Figure 26. Total organic carbon (TOC) versus percentage of mercury present as methylmercury in waters. 1. East Pond, 6/5/02. 2. East Pond, 7/29/03. 3. Red Pond, 8/29/01. 4. Red Pond, 6/4/02. 5. Red Pond, 7/28/03. 6. Sinkhole near north end of Red Pond, 7/28/03. 7. Discharge at tunnel portal, 6/5/02. 8. Backwater on Clear Creek at Igo Gauge, 6/5/02. 9. Backwater on Clear Creek at Igo Gauge, 7/29/03.