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### **Occurrence and transport of total mercury and methyl mercury in the Sacramento River Basin, California**

**J. Domagalski\***

U.S. Geological Survey, Placer Hall, 6000 J Street, Sacramento, CA 95819-6129, USA

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### **Abstract**

Mercury poses a water-quality problem for California's Sacramento River, a large river with a mean annual discharge of over 650 m<sup>3</sup>/s. This river discharges into the San Francisco Bay, and numerous fish species of the bay and river contain mercury levels high enough to affect human health if consumed. Two possible sources of mercury are the mercury mines in the Coast Ranges and the gold mines in the Sierra Nevada. Mercury was once mined in the Coast Ranges, west of the Sacramento River, and used to process gold in the Sierra Nevada, east of the river. The mineralogy of the Coast Ranges mercury deposits is mainly cinnabar (HgS), but elemental mercury was used to process gold in the Sierra Nevada. Residual mercury from mineral processing in the Sierra Nevada is mainly in elemental form or in association with oxide particles or organic matter and is biologically available. Recent bed-sediment sampling, at sites below large reservoirs, showed elevated levels of total mercury (median concentration 0.28 µg/g) in every large river (the Feather, Yuba, Bear, and American rivers) draining the Sierra Nevada gold region. Monthly sampling for mercury in unfiltered water shows relatively low concentrations during the nonrainy season in samples collected throughout the Sacramento River Basin, but significantly higher concentrations following storm-water runoff. Measured concentrations, following storm-water runoff, frequently exceeded the state of California standards for the protection of aquatic life. Results from the first year of a 2-year program of sampling for methyl mercury in unfiltered water showed similar median concentrations (0.1 ng/l) at all sampling locations, but with apparent high seasonal concentrations

measured during autumn and winter. Methyl mercury concentrations were not significantly higher in rice field runoff water, even though rice production involves the creation of seasonal wetlands: higher rates of methylation are known to occur in stagnant wetland environments that have high dissolved carbon.

**Author Keywords:** mercury; water pollution; water quality; Sacramento River; California

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## 1. Introduction

Water-quality problems can result from precious-metal mining activities. Runoff from abandoned or inactive mines can result in serious consequences for aquatic biological communities, as well as cause human health problems. The Sacramento River Basin (Fig. 1) in northern California has extensive mineral and mine drainage sources to the Sacramento River. Gold mining activities in the Sierra Nevada (Fig. 2) became increasingly important following the discovery of placer deposits in 1849. Mercury was mined in California in the Coast Ranges (Fig. 2), and then used in Sierra Nevada gold mining operations to recover the gold from ore-bearing minerals by the amalgamation process (Bradley, 1918). Once released to the environment, mercury can affect aquatic communities by bioaccumulation, resulting in potential human health impacts by consumption of mercury-contaminated fish. The form of mercury released to the environment affects how much bioaccumulation may occur; the organic compound methyl mercury ( $\text{CH}_3\text{Hg}^+$ ) bioaccumulates most rapidly in tissue. Mercury methylation rate depends on the form or speciation of mercury released to the environment. Methylation of  $\text{HgS}$  is slow, relative to elemental mercury, because of the insolubility of that mineral. Methylation also is dependent on various features of the aquatic environment. Higher rates are known to occur in stagnant wetland environments that have high amounts of dissolved organic carbon, compared to lower rates in swiftly flowing rivers (Zilloux et al., 1993; Rudd, 1995).



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Fig. 1. Study area.

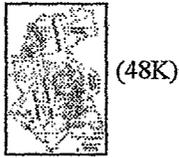


Fig. 2. Physiographic provinces and locations of gold and mercury mines in the Sacramento River Basin, California.

Mercury accumulation in fish has occurred in both the Sacramento River and the San Francisco Bay (Fig. 1), and advisories for fish consumption in these areas have been posted (Rasmussen and Blethrow, 1990; San Francisco Regional Water Quality Control Board, 1995). Although the locations of mercury mines and gold mining activities are known, the principal sources of mercury to the Sacramento River and its tributaries, the locations of mercury methylation, the forms of mercury introduced to the Sacramento River, and the potential to mitigate the mercury loads are not well understood. A study of the temporal and spatial variability in mercury and the methyl mercury concentrations in unfiltered water and associated loads was undertaken as part of the Sacramento River Basin National Water Quality Assessment (NAWQA) Program of the U.S. Geological Survey (USGS). Complete details of the NAWQA Program are given in Hirsch et al. (1988) and Leahy and Thompson (1994). The purpose of this report is to describe the design and discuss results from one complete year of sampling. The period of that sampling was from February 1996 to February 1997. Bed sediments also were studied to determine mercury concentrations. Those samples were collected during October–November 1995.

## 2. Summary of area, geology, and mineralization

The Sacramento River Basin (Fig. 1) in northern California covers nearly 70,000 km<sup>2</sup>. The Sacramento River, the largest river in California, has an average streamflow of over 650 m<sup>3</sup>/s and an annual runoff of 27,600 hm<sup>3</sup>. The Sacramento River is a major source of drinking water for residents of northern and southern California, a principal source of irrigation water for the Sacramento Valley, and the largest source of freshwater flow to the San Francisco Bay.

Physiographic provinces for the Sacramento River Basin are shown in Fig. 2. Extensive mineralization and mining activities for precious metals occur in the Klamath Mountains, the Coast Ranges, and the Sierra Nevada. Mercury deposits occur in widely scattered localities in the Klamath Mountains (Albers, 1966). These deposits are a potential source for mercury to the Sacramento River. Mercury mines are most extensive in the Coast Ranges (Fig. 1 and Fig. 2), especially near Clear Lake. The Clear Lake hydrothermal mercury deposits of Cenozoic age are the most northern part of a group of similar deposits associated with volcanism and the migration of a transform fault system (Rytuba, 1996). Cache Creek, which is in the Coast Ranges (Fig. 1 and Fig. 2), is another potential source of mercury because several mercury mines are within its drainage basin. However, Cache Creek generally is not a significant source of mercury to the Sacramento River, except during occasional high flows that reach the Sacramento River. The locations of Sierra Nevada gold mines are shown in Fig. 2. In addition, numerous locations on rivers have been exploited for placer gold deposits. Gold deposits of the Sierra Nevada have been described by Clark (1966). According to that report, gold deposits are related genetically to the intrusions of the Sierran granitic batholith and occur in belts of metamorphic rocks, including slate, greenstones, and amphibolite. The western foothills of the Sierra Nevada, known as the Mother Lode, contain linked or

en-echelon gold-quartz veins and intervening mineralized schist and greenstone (Clark, 1966). The placer deposits are of two main types: (1) older (Tertiary age), which are on interstream ridges; and (2) younger (Quaternary age), which are in or near present stream channels. The older deposits consist of gravels that were deposited in river channels several million years ago and subsequently capped by beds of andesite (Clark, 1966).

### 3. Sampling sites and collection methods

Sampling sites were chosen to determine the spatial and temporal variability of mercury in river bed sediments and water, and in the methyl mercury concentrations of water in the main stem of the Sacramento River and its major tributaries. Seventeen sites were selected for the sampling of river bed sediment (Fig. 3) on rivers draining the major physiographic zones. Bed sediments were sampled during October and November 1995 when river flow was low at all sites. A total of eleven sites were chosen for monthly sampling of total mercury in unfiltered water (Fig. 4). Four of these sites were chosen on the Sacramento River. The site on the Sacramento River above Bend Bridge near Red Bluff, located farthest upstream, was chosen because the mercury concentrations were expected to be relatively low. River flow at this site is largely the result of reservoir releases from Shasta Lake. However, local runoff from the Klamath Mountains may elevate mercury concentrations at this site, especially following winter storms. The site on the Sacramento River at Colusa was chosen because it is sufficiently downstream from mercury sources within the Klamath Mountains, and upstream of potential sources from the Sierra Nevada and most agricultural drainage. The site on the Sacramento River at Verona was chosen because it is immediately downstream of the confluence of the Feather and Sacramento rivers and directly downstream of two large agricultural drainage canals. The Feather River is a large tributary and also a large potential source of mercury from sources in the Sierra Nevada. The final site, the Sacramento River at Freeport, was chosen because of its proximity to the mouth of the river. Three of the eleven sites were chosen on large rivers draining the Sierra Nevada: the Yuba River near Marysville, the Feather River near Nicolaus, and the American River at Sacramento. One site, Cache Creek at Rumsey, was chosen to measure mercury inputs from the Coast Ranges. Two agricultural sites within the Sacramento Valley were chosen (the Colusa Basin Drain at Road 99E near Knights Landing and the Sacramento Slough near Knights Landing), each of which drain a large part of the valley: 4270 and 3370 km<sup>2</sup>, respectively. Finally, one urban site, Arcade Creek near Del Paso Heights, was chosen. These eleven sites were sampled monthly and across a range of flow conditions. An alternative site, Yolo Bypass at Interstate 80 near West Sacramento, was sampled occasionally. The Yolo Bypass is a flood control channel that is used only when the channel capacity of the Sacramento River is expected to be exceeded. During parts of the year, the land in the Yolo Bypass is used for agriculture. When necessary, water is then diverted into the Yolo Bypass from several locations along the Sacramento River between Colusa and Sacramento to prevent downstream flooding in Sacramento. At times, more water is actually flowing through the Yolo Bypass to the San Francisco Bay than through the Sacramento River at Freeport. Therefore, it is necessary to sample both waterways to understand mercury transport to the San Francisco Bay. Because the Yolo Bypass is a poorly defined channel, it is not shown on Fig. 4; only the sampling location is identified.



(64K)

Fig. 3. Locations of sites sampled for mercury in river bed sediment in the Sacramento River Basin, California.



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Fig. 4. Locations of sites sampled for mercury in water in the Sacramento River Basin, California.

Five sites were chosen for sampling of methyl mercury in unfiltered water. These sites were chosen to determine whether agricultural practices in the Sacramento Valley may be contributing to the load of methyl mercury in the Sacramento River. Rice is a major crop of the Sacramento Valley. Rice production involves flooding a field for 5 months, thus creating a temporary wetland. Because methyl mercury is known to form in natural wetlands (Zilloux et al., 1993), it is possible that mercury methylation may be occurring in these rice fields. The two agricultural sites, Colusa Basin Drain at Road 99E near Knights Landing and Sacramento Slough near Knights Landing, were sampled monthly for methyl mercury. Three sites on the Sacramento River also were sampled monthly: the Sacramento River at Colusa, which is above most of the agricultural drainage to the Sacramento River; the Sacramento River at Verona, which is directly downstream of most of the agricultural drainage to the Sacramento River; and the Sacramento River at Freeport, which is downstream of the agricultural drainage and urban runoff from the city of Sacramento. The site on the Sacramento River at Colusa was expected to have the lowest concentrations of methyl mercury and the least variability because it is above the agricultural drainage and above the confluence of the Sacramento River and the Feather River, the largest river draining the Sierra Nevada gold region. The sites on the Sacramento River at Verona and at Freeport were expected to have higher concentrations of methyl mercury because they are downstream of agricultural sources, mining sources, and, in the case of the site at Freeport, urban sources.

Samples for mercury in river bed sediment were collected by selecting a 100-m reach of river and collecting material from sediment deposition zones. The composite sample was collected with a Teflon spoon and placed in an acid-cleaned glass container. After thorough mixing, the sample was sieved through a 63- $\mu$ m screen and placed in a clean plastic jar. Samples for mercury and methyl mercury in unfiltered water were collected using dedicated equipment and clean techniques to minimize contamination; Teflon sampling equipment and bottles were used. Three-liter Teflon bottles, equipped with Teflon nozzles for the collection of isokinetic samples, were used for sample collection. Prior to sampling, the Teflon equipment was soaked in an acid bath containing 10% hydrochloric acid at a temperature of 65°C for 48 h. The equipment and bottles were rinsed with clean water, and all bottles were filled with 1% hydrochloric acid. The bottles were capped tightly with a wrench and double-wrapped in plastic bags for transport. For total mercury in unfiltered water samples, 500-ml bottles were used to hold the sample. At the field site, the bottles were rinsed three times with native water, filled with the sample, preserved with approximately 10 ml of 50% hydrochloric acid, recapped tightly with a wrench, and double-wrapped in plastic bags. The procedure for collecting methyl mercury in unfiltered water samples was similar, except that 250-ml Teflon bottles were used and, in place of the preservative acid, the samples were frozen. At all sites, a depth-integrated sample was collected at a

single point of the river, usually near the center of the channel. It was not possible to collect a sample integrated across the entire channel because the compositing equipment, normally used for that purpose, could not be cleaned in hot acid in the same manner as the Teflon bottles. At all sites, samples also were collected for dissolved and particulate organic carbon, major cations and anions, other trace metals, nutrients, pH, alkalinity, conductivity, and dissolved oxygen.

Quality assurance for sediment analyses consisted of the collection of triplicate samples at two stations to check for variability in collection procedures. Those samples had very little variability (less than 5%). Replicate samples for total mercury in unfiltered water were collected each month at three stations, and replicate samples for methyl mercury in unfiltered water were collected at one randomly selected station per month. The variability of the replicate samples was between 10 and 15%. Blank samples of unfiltered clean laboratory water also were submitted for total mercury. Total mercury in those samples was equal to, or less than, 0.2 ng/l. Replicate samples for total methyl mercury in unfiltered water consisted of replicate samples at one station per month. Fewer replicates were collected for total methyl mercury, relative to total mercury, because samples were collected at fewer sites. The variability of total methyl mercury concentrations in the replicate samples was similar to that of the total mercury samples.

#### 4. Sample preparation and analysis

Samples for mercury in bed sediment were shipped to the USGS Geologic Division for analysis. The samples were acid-digested and analyzed by cold-vapor atomic absorption spectrometry; the detection limit was 0.02  $\mu\text{g/g}$ . A detailed description of that method is given in [Fishman and Friedman \(1989\)](#). Samples for mercury and methyl mercury in unfiltered water were shipped to the USGS laboratory in Madison, Wisconsin, for analysis. The total mercury samples were prepared and analyzed according to the method of [Bloom and Fitzgerald \(1987\)](#). The method uses bromine monochloride oxidation, two-stage gold amalgamation, and cold-vapor atomic fluorescence detection; the detection limit was 0.03 ng/l. Methyl mercury was distilled and ethylated according to the method of [Horvat et al. \(1993\)](#) and [Liang et al. \(1993\)](#). The distillation process separates methyl mercury from interfering substances, such as dissolved organic carbon. The detection limit for methyl mercury was 0.02 ng/l.

#### 5. Results

A plot of mercury in river bed sediment is shown in [Fig. 5](#). Mercury concentrations are shown relative to average abundance in the earth's crustal layer ([Emsley, 1996](#)). The highest concentrations of mercury in bed sediment were measured in rivers draining the Sierra Nevada: the Feather, Yuba, Bear, and American rivers. The mercury in samples from these rivers can be attributed to mercury used historically in gold mining operations. However, it is currently known that reservoirs upstream of these sampling sites act as traps for both sediment-associated inorganic mercury and biologically available mercury ([Slotton et al., 1997](#)). This was determined by sampling biota above, within, and below gold-country reservoirs. It was found that elevated levels of mercury can be measured in the tissue of aquatic organisms, such as trout, collected above and within the reservoirs, but the levels downstream of the reservoirs are significantly lower ([Slotton et al., 1997](#)). Samples were collected from organisms across an entire food web and tested for mercury concentrations ([Slotton et al., 1997](#)). It is possible, however, that some particulate mercury is transported from reservoirs as a result of storm-water runoff. The elevated mercury levels measured below these reservoirs are probably the result of deposition prior to the construction of the reservoirs. Mercury is elevated on the main stem of the Sacramento River at the site above Bend Bridge near Red Bluff, but is relatively low at the Sacramento River at Colusa. Mercury concentrations also are elevated at

the Sacramento River at Verona, which is downstream of the confluence of the Sacramento and Feather rivers. It was expected that the highest concentrations might be found at Cache Creek at Guinda because that site is downstream of a mercury mining location. Although the mercury concentrations are elevated, relative to average crustal abundance, the level was below that of the Sierra Nevada streams and comparable to the urban site at Arcade Creek near Del Paso Heights. However, it is possible that the unexpectedly lower concentrations of mercury in Cache Creek sediments may be attributed to the method of material transport. One possibility is that the sediment-bound mercury is transported through the system to downstream locations by storm-water runoff and little is actually deposited in the creek bed, such as near the sampling location on Cache Creek at Guinda. Further investigation could test this hypothesis. Mercury in river sediment was very low at the two reference sites, McCloud River below Ladybug Creek near McCloud and Deer Creek near Vina. Mercury concentrations also were low at Jack Slough at Highway 70 near Marysville. Although the drainage for Jack Slough (Fig. 1) is mainly in the Sacramento Valley, mercury associated with gold processing in the Sierra Nevada was not detected in sediment samples from Jack Slough.

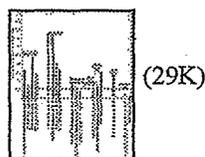


Fig. 5. Concentrations of mercury, in micrograms per gram, measured in river bed sediment in the Sacramento River Basin, California.

Total mercury concentrations in unfiltered water were expected to be partly dependent on bed sediment concentrations, and the highest concentrations in water were expected during periods of high flow when bed sediment becomes resuspended in the water column. The highest concentrations were expected to be found downstream of the Sierra Nevada gold mining region (on the Feather, Yuba, and American river sites, and on the Sacramento River at Verona and at Freeport) and on Cache Creek in the Coast Ranges, which is downstream of mercury mines.

Boxplots of total mercury concentrations in unfiltered water samples are shown in Fig. 6 for the period February 1996–February 1997. The concentrations are shown relative to a California standard for the protection of aquatic life (12 ng/l). The boxplots show that the data are positively skewed. The high river-flow periods are responsible for the skewness of the data. The greatest amount of variability in concentrations is at Cache Creek at Rumsey. Mercury concentrations exceeded 12 ng/l, the state standard for protection of aquatic life, periodically at all sites and frequently at a few sites. Mercury concentrations exceed 12 ng/l for up to 60% of the samples collected at Cache Creek at Rumsey and for up to 25% of the samples at Colusa Basin Drain at Road 99E near Knights Landing, the Sacramento Slough near Knights Landing, and at Arcade Creek near Del Paso Heights. The lowest median mercury concentration (1.56 ng/l) was measured at the American River at Sacramento (Fig. 6). The median concentration at the American River at Sacramento was significantly different from all sites, except for the Sacramento River above Bend Bridge near Red Bluff, using the nonparametric Mann–Whitney test of medians (significance level 0.05). Median mercury concentration was also low (2.04 ng/l) at the Sacramento River above Bend Bridge near Red Bluff. That median concentration was significantly different from the median concentration at the Sacramento River sites at Verona and at Freeport, and at Cache Creek at Rumsey, Arcade Creek near Del Paso Heights, Colusa Basin Drain at Road 99E near Knights Landing, and the

Sacramento Slough near Knights Landing. Median mercury concentration for the Sacramento Slough near Knights Landing was significantly higher than those at the Sacramento River at Colusa, the Yuba River near Marysville, and the Feather River near Nicolaus, as well as at the American River at Sacramento and the Sacramento River above Bend Bridge near Red Bluff.

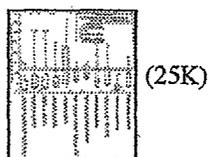


Fig. 6. Concentrations of total mercury in unfiltered water, in nanograms per liter for the period from February 1996 to February 1997 in the Sacramento River Basin, California.

The highest total mercury in unfiltered water for most of the sites occurred during runoff periods such as during December 1996 and especially during January and February 1997. Extreme flooding occurred in northern California during the first week of January 1997, following heavy rainfall on a heavy Sierra Nevada snowpack. Precipitation was widespread throughout northern California with some of the largest amounts (exceeding 50 cm) within the Feather River drainage. Fig. 7 shows the concentration of mercury in unfiltered water samples at the eleven sites in the Sacramento River Basin for January 1997.

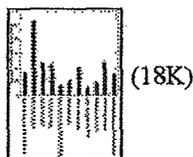


Fig. 7. Concentrations of total mercury in unfiltered water in nanograms per liter, collected during January 1997 in the Sacramento River Basin, California.

During the first week of January 1997, when river flows were greatest, mercury loads along the main stem of the Sacramento River increased from 7.2 kg/d at the northern station on the Sacramento River above Bend Bridge near Red Bluff to about 24 kg/d on the Sacramento River at Colusa, an intermediate location. The loading at the most downstream locations, where the Sacramento River flows into the San Francisco Bay, was 32 kg/d on January 7, 1997. The January 7, 1997, mercury load is the summation of the loads measured at the Sacramento River at Freeport and at the Yolo Bypass at Interstate 80 near West Sacramento. A subsequent measurement a month later showed that the loading to the San Francisco Bay decreased to 2 kg/d. By comparison, loadings to the San Francisco Bay during the dry season are on the order of 0.2 kg/d.

Mercury can be mobilized and transported by storm-water runoff to the San Francisco Bay from several large river basins in northern California, including the Sacramento River Basin. Although the increase in river flow during storm periods may result in dilution of dissolved constituents, the loading and transport of sediment-bound mercury increases because of soil erosion and suspension of river bed sediment by higher flow regimes. The majority of transport of sediment-bound mercury occurs during high-flow periods. During the January 1997 flood, the greatest increase in mercury concentration and load was measured at the sites between the Sacramento River above Bend Bridge near Red Bluff and the Sacramento River at Colusa. Although the rainfall was particularly high over the drainages of the Feather

and Yuba rivers, mercury loadings from those rivers were not as great as along the Sacramento River. This suggests that a significant source or sources of mercury are present somewhere between Red Bluff and Colusa. At present, it is not known where those sources are located.

Boxplots of methyl mercury concentrations in unfiltered water at the five sites sampled are shown in Fig. 8. Similar to the plots for total mercury, only a few samples are positively skewed. Median concentrations of methyl mercury are statistically the same at these five sites according to the nonparametric Mann-Whitney test (significance level 0.05). The median concentrations at the two agricultural sites are the same as for the Sacramento River sites, even at the Sacramento River at Colusa, which is above the agricultural drainage. The boxplots show a greater degree of variability in concentration for the two agricultural sites relative to the Sacramento River at Colusa. The agricultural drainage from rice fields is contributing methyl mercury to the Sacramento River, but the similarity in median methyl mercury concentrations suggests that the inputs are no greater than in other parts of the basin.

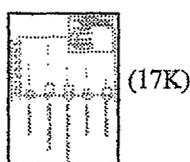


Fig. 8. Concentrations of total methyl mercury in unfiltered water, in nanograms per liter for the period from February 1996 to February 1997 in the Sacramento River Basin, California.

Methyl mercury concentrations for various types of aquatic media have been reported by Rudd (1995). Upland rivers located above wetland environments have low methyl mercury concentrations equal to or less than 0.1 ng/l. The median concentrations of methyl mercury in the Sacramento River sites are very close to 0.1 ng/l. Therefore, methyl mercury concentrations exceed this value at least 50% of the time. The highest values measured during this study were from November 1996 to February 1997, indicating a seasonal component related to higher river flows or storm-water runoff. According to Rudd (1995), runoff from flooded areas, along with discharge from wetlands and atmospheric deposition, represents an important source of methyl mercury to a watershed. There are large anthropogenic and geologic sources of mercury within the Sacramento River Basin. The increases in methyl mercury concentrations measured at several locations throughout the Sacramento River Basin during the rainy season suggest that numerous diffuse sources of methyl mercury are present. Alternatively, most of the methyl mercury may be generated during the dry season at numerous locations, but may only be mobilized and transported downstream during high flow, which occurs primarily during the winter. The highest concentrations of methyl mercury in the Sacramento River were measured after runoff periods, suggesting that diffuse nonpoint sources contribute the greatest amount of methyl mercury to the Sacramento River.

## 6. Conclusions

Sampling for total mercury and methyl mercury within the Sacramento River watershed have shown, at least for the 1-year period of record, that concentrations and loads tend to be greatest following runoff periods. Although it was suspected that the greatest concentrations in water would be measured downstream of Sierra Nevada rivers because of the historic use of mercury to recover gold from ore, the greatest increase in concentrations and loadings on the Sacramento River actually were measured upstream of the Feather River inputs from the Sierra Nevada gold mining region following a period of

extreme runoff. The locations of the upstream sources of mercury currently are not known and are the subject of ongoing investigations. Mercury concentrations at several locations exceeded California water-quality standards for the protection of aquatic life, especially following storm-water runoff. The findings of this study indicate the need to examine and identify the sources of that mercury, its bioavailability, and the potential for remediation or environmental cleanup. Methyl mercury concentrations in rice field drainage water were not found to be significantly higher than concentrations measured at any of the Sacramento River sites, including those located above the agricultural region. However, relatively high concentrations of methyl mercury can be measured in the Sacramento River following runoff periods, suggesting that numerous diffuse sources exist for methyl mercury in this river basin.

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\*Fax: +1 (916) 278 3071; E-mail: [joed@usgs.gov](mailto:joed@usgs.gov)

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