**Figure 19.** Plots of suspended sediment concentration (*A*) and load (*B*) versus stream flow at Freeport, California, July 1995 through June 1997.

100 compared with loads in May–June 1997. These differences were caused by high flows from Spring Creek Reservoir during the January 1997 floods. A contributing factor to this effect was the fact that a diversion of Upper Spring Creek above the Boulder Creek confluence into Flat Creek (fig. 2) was not operative during early January 1997, because the floods had washed out a bridge on Iron Mountain Road. Therefore, the flows to Spring Creek Reservoir were increased by several hundred cubic feet per second, contributing to a spill condition. On January 2, 1997 the daily average flow from the Spring Creek Debris Dam (SCDD) was 750 $ft^3$/s, including releases from the gates within the dam plus the flow over the spillway.

Water from Whiskeytown Lake, which is diverted into the Spring Creek arm of Keswick Reservoir through the Spring Creek Power Plant (SCPP, fig. 2), was sampled on two occasions for this study (December 1996 and May–June 1997). Although no samples for colloidal material were collected, total recoverable analyses of whole-water samples collected at these times can serve as proxies for colloidal plus dissolved analyses. The trends seen throughout the rest of the watershed concerning the

dominance in loads during December 1996 over May–June 1997 are not observed in the water samples from Whiskeytown Lake; May–June 1997 loads for aluminum and iron were considerably higher than December 1996 loads (Appendix 1), despite the fact that discharge on the sampling date of May 29, 1997 was 2,720 $ft^3$/s versus 3,900 $ft^3$/s on December 11, 1996 (Alpers and others, 2000). Trace metal loads from Whiskeytown Reservoir generally were much lower than the metal loads from Spring Creek, as discussed in a later section of this report (Mass Balance and Metal Transport in Keswick Reservoir).

Temporal variations in the relative proportion of dissolved and colloidal loads were markedly different for Spring Creek compared with the mainstem Sacramento River sites. During two of the three sampling periods at Spring Creek (December 1996 and January 1997), the dissolved load was greater than 50 percent of the total for lead, aluminum, and iron; during the May–June 1997 trip, the loads in Spring Creek were less than 50 percent dissolved for these metals. For cadmium, copper, and zinc, the proportion of the load in Spring Creek that was dissolved was greater than 95 percent for all three sampling periods. In contrast, at the Shasta Dam and Keswick Dam

sampling sites, the proportions of the metal load from dissolved copper were approximately 50 (±25) percent for all six sampling periods (fig. 12), although the site below Shasta Dam was not sampled in January 1997.

During the January 1997 sampling period, almost all of the Al, Cd, Cu, Fe, Pb, Hg, and Zn loads were associated with colloids at all of the downstream mainstem Sacramento River sites (from Bend Bridge to Freeport) (figs. 4–15; David Roth, U.S. Geological Survey, written commun., 1999). These results were due to the much higher total colloidal concentrations combined with the higher discharge during this extremely high-flow period. The only site for which more than 10 percent of the cadmium, copper, and zinc loads was dissolved during the January 1997 sampling period was the site below Keswick Dam (the site below Shasta Dam was not sampled during January 1997). In contrast, cadmium, copper, and zinc had considerably higher proportions of dissolved loads during other sampling periods (including December 1996) at the downstream sites (figs. 5, 6, and 9). This was not observed for lead, iron, and aluminum, for which the dissolved proportion of the load was relatively small regardless of the time of year (figs. 4, 7, and 8).

### Cadmium, Copper, and Zinc

As noted earlier, the highest cadmium loads observed in this study were during the January 1997 sampling period, followed by the December 1996 period (fig. 5). Cadmium loads at the upstream sites, including Shasta Dam and Keswick Dam and, to a lesser extent, Bend Bridge, had a large dissolved component; for all six sampling periods, more than 50 percent of the cadmium load below Keswick Dam was in dissolved form (figs. 5 and 11). Downstream mainstem sampling sites invariably showed a decline in the proportion of dissolved cadmium.

With regard to cadmium loads, there is a clear impact on the Sacramento River system from Spring Creek during certain flow regimes (figs. 5 and 11). Spring Creek carries the acid drainage from the mines at Iron Mountain and enters Keswick Reservoir about 2 mi upstream of Keswick Dam (fig. 2). Depending on the flow regime, a significant proportion of the cadmium load may come from Shasta Lake and its tributaries. During the July, September, and December 1996 sampling periods, the cadmium load below

Shasta Dam was more than 50 percent of the cadmium load below Keswick Dam. (There are no data from this study regarding colloidal cadmium transport from Shasta Lake for either the September 1996 or January 1997 sampling periods.) Finally, except for one sampling point during one sampling period (below Keswick Dam, May–June 1997), total cadmium loads uniformly increased between the Shasta Dam site and the Colusa site; during the July 1996, September 1996, and January 1997 sampling periods, cadmium loads continued to increase downriver to Freeport. In summary, there appear to be inputs of cadmium to the Sacramento River system both above Shasta Dam and above Keswick Dam that are primarily related to mine drainage; these were apparently the dominant inputs during the sampling periods in November and December 1996 and May–June 1997. During the sampling period of January 1997, the flood conditions caused transport of abundant colloidal material below Keswick Dam that dominated cadmium transport.

The decreases in cadmium transport observed along the flow path between the Colusa and Freeport sites during the November 1996, December 1996, and May–June 1997 sampling periods are likely related to large decreases in overall colloidal concentration (fig. 11). Figures 16 through 18 demonstrate that greater than 50 percent of the colloidal cadmium occurs in a reducible phase such as hydrous iron and manganese oxides, and that this proportion does not change in a systematic way either spatially or temporally.

Generally, the patterns described above for cadmium are similar to those for zinc (figs. 9 and 15). The greatest zinc loads occurred during the December 1996 and January 1997 sampling periods; upriver sites (especially Keswick Dam) showed zinc transport to have a large dissolved component, usually greater than 50 percent (fig. 15). Zinc loads at Shasta Dam were greater than 50 percent of the loads at Keswick Dam for July 1996 and December 1996 (no data for Shasta Dam in September 1996 or January 1997). Total zinc loads tended to increase uniformly between Shasta Dam and Colusa. As with cadmium, zinc transport increased continually between Keswick Dam and Freeport during the September 1996 and January 1997 sampling periods. Decreases in loads of zinc (and most other metals) between Colusa and Freeport for some sampling periods are apparently related to the anomalously high colloidal concentrations observed consistently at Colusa (figs. 10–15).

Processes responsible for the consistent increase in the concentrations of colloids at Colusa relative to sampling sites immediately upstream and downstream remain unknown. There are no anthropogenic causes of fine-grained sediment, such as construction activity, that are known to have been active during the study period, nor is there much activity from industrial sites, agricultural drainage, or municipal sewage treatment plants in the vicinity. Also, there are no major tributary streams in the reach of the Sacramento River immediately upstream of Colusa. A longitudinal profile of riverbed elevation in the Sacramento River (fig. 20) indicates that the gradient near Colusa is significantly lower than at Bend Bridge, the closest upstream sampling station. This change in slope could have led, over many years, to deposition of fine-grained sediments on the streambed in the area upstream of Colusa. Levees for flood control regulate the river in the immediate vicinity of Colusa. A linear, engineered streambed would have higher water velocities, leading to increased sheer stresses that would cause the resuspension of fine-grained sediments from the river channel.

Some differences between zinc and cadmium speciation in colloids are evident in the data for samples collected during December 1996 and January 1997 (figs. 16A, 16D, 17A, and 17D). Colloidal zinc during high flow tended to have a much higher proportion in the residual phase than in the reducible phase, especially at downriver sites. Inspection of the zinc/cadmium ratios for dissolved and colloidal analyses in which both elements were detected (fig. 21A) reveals that dissolved zinc/cadmium ratios are consistently lower than colloidal zinc/cadmium ratios for sites both upstream and downstream of Redding. The zinc/cadmium ratios in dissolved samples range from 40 to 210. Box illustrations defining the probability distribution of 25 to 75 percent of the data for dissolved samples (fig. 21A) range from about 50 to 100 for sites downstream of Redding and from about 100 to 410 for sites upstream of Redding, with most (19 of 27 samples) in the range of 70 to 150. In contrast, the zinc/cadmium ratios in colloidal samples had an overall range from about 100 to 410. The probability distribution for 25 to 75 percent of the data fall within the range of 150 to 260 for downstream sites and about 190 to 320 for upstream sites. Thus, there is a clear and consistent pattern of higher zinc/cadmium ratios in colloids relative to the dissolved component. The pattern holds true at individual sample sites as well. These differences are consistent with laboratory data that indicate that zinc will adsorb to sediment preferentially to cadmium at a given pH (Dzombak and
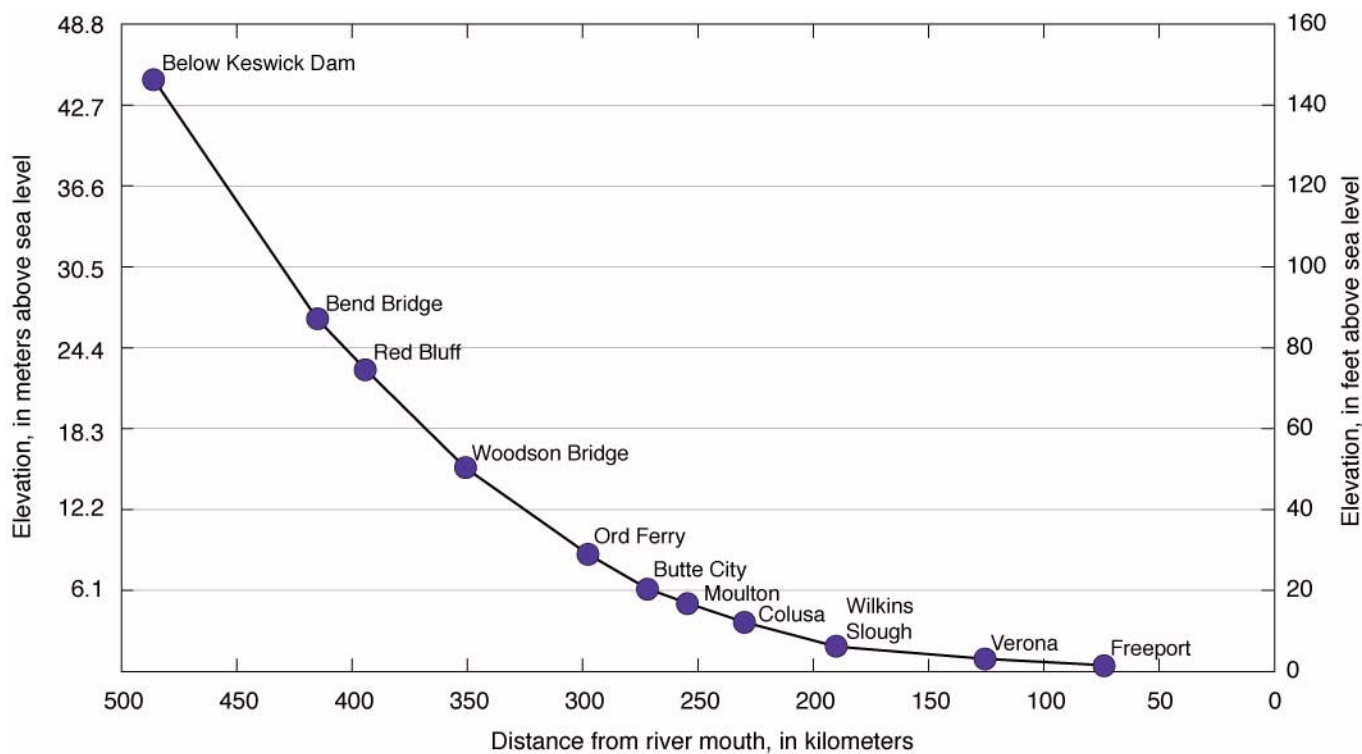


**Figure 20.** Graph showing elevation of streambed in relation to distance from river mouth, Keswick Dam to Freeport, Sacramento River, California.

Morel, 1990; Smith, 1999). Although there are some apparent trends of zinc/cadmium ratios in colloids and dissolved components with distance, there are not a sufficient number of dissolved cadmium determinations above the detection limit to warrant quantitative analysis.

The zinc/cadmium ratios observed in colloids from the Sacramento River are consistent with massive sulfide deposits as a source (Hamlin and Alpers, 1996). Seal and others (in press) have compiled data for mine drainage and natural acid rock drainage from six different types of massive sulfide deposits, showing that the zinc/cadmium ratio does not change appreciably among these types of deposits in various geologic settings. Also, the zinc/cadmium ratio in average crustal rocks is in a similar range (approximately 50 to 200; Clark, 1924), therefore, this ratio is not a definitive tracer for mining-related metal sources.

Temporal and spatial trends in copper loads show some similarities to those for zinc and cadmium loads. For example, the December 1996 and January 1997 sampling periods during high flow account for the highest copper loads described at all sites on the river. Also, the proportion of dissolved copper was always greatest at upriver sites, however the proportions did not decrease with distance downriver in the same ways as they did for cadmium and zinc loads (compare the pie graphs in fig. 12 with those in figs. 11 and 15). Finally, as with zinc, the reducible fraction of copper generally accounted for about 50 percent of the total colloidal copper (figs. 16B, 17B, and 18B), but this proportion decreased as copper in the residual phase increased downstream during the January 1997 sampling period (fig. 17B).

Variations in zinc/copper ratios for dissolved and colloidal components show contrasting patterns at sites upstream and downstream of Redding (fig. 21B). At the upstream sites, zinc/copper ratios in the dissolved component are consistently higher than those in the colloidal phase. The 25 to 75 percent probability range for dissolved zinc/copper in upstream sites is from about 2.3 to 4.2, whereas the corresponding box range for colloidal zinc/copper in upstream sites is from about 1.1 to 2.2 (fig. 21B). This relation is consistent with preferential adsorption of copper relative to zinc onto suspended matter at a given pH (Dzomback and Morel, 1990; Smith, 1999). However, at the sampling sites downstream of Redding, the relation of the zinc/copper ratio between dissolved and colloidal components is reversed from

that observed upstream. The 25 to 75 percent probability range for zinc/copper in the dissolved component for downstream sites is about 0.5 to 1.3, whereas the corresponding range for zinc/copper in the colloidal component is 1.5 to 2.1. The contrasting behavior of the zinc/copper ratio at the downstream sites may be caused by aqueous complexation of copper by dissolved organic carbon, a process that does not affect zinc to the same degree (Stumm and Morgan, 1996). Concentrations of dissolved organic carbon tend to increase with distance downstream in the Sacramento River (Alpers and others, 2000), which may play a role in the partitioning of copper and zinc between the dissolved and colloidal components.

The adsorption and desorption behavior of zinc and copper were investigated in laboratory studies using sediment samples from the Spring Creek arm of Keswick Reservoir (Coston and others, 1998; Nordstrom and others, 1999). The presence of schwertmannite, a sulfate-bearing ferric oxyhydroxide, likely causes a shift to adsorption at lower pH for a given amount of available surface area, relative to synthetic sulfate-free hydrous ferric oxides such as ferrihydrite (Coston and others, 1998; Webster and others, 1998). In the Sacramento River and Keswick Reservoir systems, the partitioning of cadmium, copper, and zinc between dissolved and colloidal phases appears to show some consistent relations between ambient field data and laboratory studies. The investigation of these relations is the focus of ongoing research by the USGS.

The ranges of zinc/copper ratios for the dissolved and colloidal components from the Sacramento River are consistent with an important source of these metals being primarily from the mineralized areas of the West Shasta mining district. The compilation by Seal and others (in press) indicates that drainage from different types of massive sulfide deposits exhibits a wide range of zinc/copper ratios. The deposits in the East Shasta and West Shasta mining districts are considered to be of the Noranda-type (Franklin and others, 1998), which typically have zinc/copper ratios between 1 and about 20 (Seal and others, in press).

## Lead, Aluminum, and Iron

The transport behavior of lead is distinct from that of cadmium, copper, and zinc. The proportion of dissolved lead load is uniformly negligible, except for occasional observations at the sampling sites below
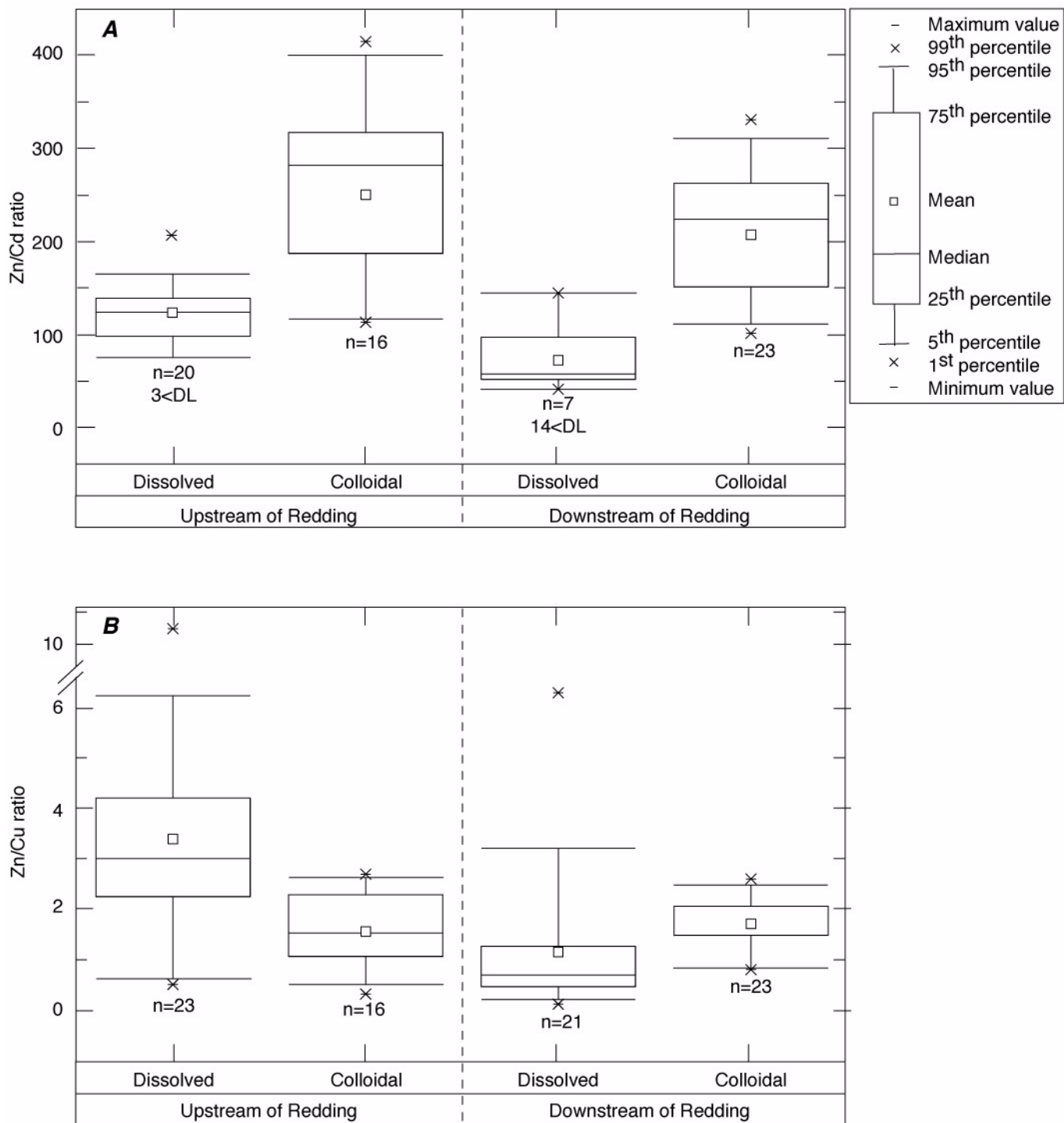
**Figure 21.** Probability box illustrations showing distribution of (*A*) Zn/Cd and (*B*) Zn/Cu data in dissolved and colloidal samples taken July 1996 through June 1997, Sacramento River, California. <DL, less than detection limit; n, number of observations greater than the detection limit. Cd, cadmium; Cu, copper; Zn, zinc.

Keswick Dam and Shasta Dam. Almost all of the lead carried in the river is associated with suspended (colloidal) sediments (figs. 8 and 14). Also, in comparison with cadmium, copper, and zinc, the ratio of the transport below Keswick Dam to that at Freeport (plus the Yolo Bypass) is much smaller (compare the upriver pie charts for lead with those for cadmium, copper, or zinc in figures 16–18, and see Appendix 1, table A1-5 and Appendix 2, table A2-5.). This smaller ratio indicates that there are probably other, more significant sources of lead in the Sacramento River watershed besides the mined areas upstream of Keswick Dam. Lead isotope data (Alpers and others, 2000) also are consistent with this interpretation.

The proportion of lead in the reducible phase of the colloids was almost always greater than or equal to about 50 percent, as was the case for copper and zinc (figs. 16–18). The reducible lead fraction in colloids was occasionally as high as 75 percent; for example at the Colusa and Verona sites during May–June 1997. The only time at which lead associated with the reducible phase was less than 50 percent of the total colloidal lead was during the January 1997 sampling period (at the mainstem sites below Keswick Dam and at Tower Bridge).

The quantities of transported aluminum and iron were similar in magnitude to each other, and the spatial and temporal variations of these two metals had similar characteristics. As with lead, practically all the aluminum transported in the Sacramento River was colloidal, except below Shasta and Keswick dams during certain sampling periods (fig. 10); a similar pattern was obtained for iron (fig. 13). A major difference between lead and aluminum speciation is that practically all of the colloidal aluminum is associated with the residual phase, whereas colloidal lead tends to be associated with the reducible phase, suggesting a close affinity for hydrous iron oxides.

Iron tended to behave similarly to aluminum at most sampling sites, with the majority of the colloidal iron (80 to 90 percent) in the residual phase. As expected, the colloid samples collected at Spring Creek, in the Spring Creek arm of Keswick Reservoir, and below Keswick Dam were exceptions to this behavior in that they had a substantial proportion of iron in the reducible phase. The iron speciation and loading data appear to indicate that Iron Mountain mine and Spring Creek are not the predominant sources of iron to the Sacramento River. The transformation of dissolved iron in Spring Creek into particulate forms of ferric iron occurs by two related mechanisms, the oxidation of ferrous iron to ferric iron and the subsequent hydrolysis of the ferric iron (Nordstrom and Alpers, 1999a). Ferrous iron actively oxidizes in the Spring Creek Reservoir, contributing abundant hydrous ferric oxide colloids to Spring Creek. The acidic water of Spring Creek carries some soluble ferric iron, which precipitates by hydrolysis caused by neutralization on mixing with dilute waters in Keswick Reservoir from both Whiskeytown Lake and Shasta Lake.

## Discussion of Transport and Fate of Metals from Various Sources

The purpose of this section is to discuss the spatial variation of metal loads in the context of known and suspected metal sources. The discussion is divided into both known sources from abandoned and inactive mines in the Shasta region, mostly upstream of Keswick Dam (Nordstrom and others, 1977), and less known sources downstream of Keswick Dam. The section on Keswick Reservoir includes some analysis of metal mass balance in the Spring Creek arm of the reservoir and in the reservoir as a whole.

### Copper-Zinc-Lead Mines in the Shasta Region

Volcanogenic massive sulfide deposits of the East Shasta and West Shasta mining districts are important sources of trace metals to the Sacramento River system and its tributaries. This section describes these sources and discusses available data from other studies on associated metal loads, for comparison with data generated in this study.

#### Mines Draining into Shasta Lake and Its Tributaries

The East Shasta mining district includes the abandoned Bully Hill and Rising Star mines, which drain into Town Creek and Horse Creek, respectively. Mineralization in the East Shasta mining district is hosted by Permian and Triassic strata, with an age of approximately 200 to 300 million years (Albers and Bain, 1985). Town Creek and Horse Creek are relatively small tributaries to Shasta Lake in its northeast corner. Copper loads were estimated to be 1.8 kg/d from the Bully Hill mine and 2.3 kg/d from the Rising Star on an annualized average basis, from periodic monitoring (Dennis Heiman, Regional Water

Quality Control Board, Central Valley Region, written commun., 1998).

Acidic drainage from several of the copper-zinc mines of the West Shasta mining district flows into Shasta Lake by way of two tributaries: Little Backbone Creek and West Squaw Creek (fig. 2). Mineralization in the West Shasta mining district is Devonian age (about 400 million years old), and consists of massive sulfides hosted by hydrothermally altered rhyolite and greenstone with minimal neutralizing capacity. The Mammoth, Golinsky, and Sutro mines in the Little Backbone Creek watershed are estimated to contribute copper loads of 32, 0.5, and 0.05 kg/d, respectively, on an annual basis, as indicated by periodic monitoring (Dennis Heiman, Regional Water Quality Control Board, Central Valley Region, written commun., 1998). In the West Squaw Creek drainage, the contribution of copper loads from the Balaklala, Keystone, Shasta King, and Early Bird mines are estimated to be 9, 1.1, 0.5, and 0.05 kg/d, respectively (Dennis Heiman, Regional Water Quality Control Board, Central Valley Region, written commun., 1998).

The total copper loads to Shasta Lake from the mines mentioned above is estimated to be about 50 kg/d, averaged throughout the year (Dennis Heiman, Regional Water Quality Control Board, Central Valley Region, written commun., 1998). The Regional Water Board advises that "* * * these are estimated loading rates based on periodic monitoring and * * * the actual discharge rate varies seasonally within any one year and between years based on annual precipitation" (Dennis Heiman, Regional Water Quality Control Board, Central Valley Region, written commun., 1998).

At the sampling site below Shasta Dam, the transport rate of total copper that is based on the sum of dissolved and colloidal loads (Appendix 1, table A1-3) ranged from 7 to greater than or equal to 180 kg/d. The comparable copper loads that are based on total recoverable analyses of unfiltered waters samples were approximately twice as high, ranging from 14 to 320 kg/d (Appendix 1, table A1-3). However, no sample was collected from the site below Shasta Dam for the present study during the period of highest flow in January 1997. Sampling by the Regional Water Board during January 1997 indicates a total recoverable copper concentration of 8.2 μg/L. (Total recoverable concentrations are based on a partial digestion of unfiltered water samples using

nitric and hydrochloric acids at pH < 2.) Combining this value with an average discharge for January 1997 of 41,600 $ft^3$/s results in an estimated average total recoverable copper load of 830 kg/d during this month of high discharge.

The data from this study, from the previous study by Nordstrom and others (1977), and from the monitoring data of the Regional Water Board indicate that abandoned mines are the source of a significant proportion of the copper loads to Shasta Lake. By analogy with data gathered for this study at Spring Creek, it is expected that copper, cadmium, zinc, and other trace metals are transported predominantly in dissolved form in acidic waters, for example, with pH values less than 5, such as the drainage from the other inactive and abandoned mines in the Shasta region. The relatively high proportions of dissolved cadmium, copper, and zinc in water samples taken during this study below Shasta Dam are consistent with the interpretation that the abandoned mines are a significant source of these metals.

### Mines Draining into Keswick Reservoir and Its Tributaries

The copper–zinc massive sulfide deposits at Iron Mountain (fig. 2) are the largest in the East Shasta and West Shasta mining districts. The overall premining size of the massive sulfide deposits at Iron Mountain is estimated at 23 million metric tons, occurring in five segments of an originally continuous lens that was displaced by normal faulting (Albers, 1985). Between the 1880s and the 1960s, about half of this tonnage was mined by underground and surface methods. Several adits remain open within Iron Mountain, serving as drains that have kept most of the unmined massive sulfide deposits in the unsaturated zone. Ready access of atmospheric oxygen and infiltrating meteoric water has resulted in optimal conditions for generation of acid mine drainage (Nordstrom and Alpers, 1995), producing some of the highest metal concentrations ever recorded in mine effluent (Nordstrom and others 1991; U.S. Environmental Protection Agency, 1992; Nordstrom and Alpers, 1995; 1999a,b; Nordstrom and others, 2000).

Prior to 1989, treatment of the metal-rich, acid drainage from Iron Mountain consisted solely of copper cementation plants operated intermittently on Boulder and Slickrock creeks. These plants, which were of variable effectiveness, used scrap iron to precipitate copper (U.S. Environmental Protection

Agency, 1992). Average metal loads in Spring Creek, downstream of the Spring Creek Debris Dam (SCDD) for the nine-year period between 1977 and 1985, were estimated as follows from Bureau of Reclamation data; copper, 350 kg/d and zinc, 1,400 kg/d (Dennis Heiman, Regional Water Quality Control Board, Central Valley Region, written commun., 1998). In perspective, these metal loads were more than twice the combined loads from the 28 next largest inactive mines in northern California and made Iron Mountain the largest discharger of metals to surface waters in the nation (U.S. Environmental Protection Agency, 1992).

Between 1989 and 1994, lime neutralization of the most concentrated adit discharges at Iron Mountain was carried out during three to four months of each wet season. Beginning in November 1994, year-round operation of a lime-neutralization plant has been removing about 85 percent of the copper loads and about 90 percent of the cadmium and zinc loads. The average metal loads leaving Iron Mountain between 1994 and 1997 were estimated to have been 52 kg copper per day and 45 kg zinc per day (Dennis Heiman, Regional Water Quality Control Board, Central Valley Region, written commun., 1998). This average copper load is roughly equivalent to the total load of copper from mine discharges into Shasta Lake, as described earlier.

The EPA has issued a Record of Decision (ROD) that mandates additional water treatment at the Iron Mountain site (U.S. Environmental Protection Agency, 1997). Specifically, the ROD calls for construction of a dam along Slickrock Creek, which drains the southwestern slope of Iron Mountain (fig. 2). Estimates by the EPA indicate that the successful implementation of the remediation measures in the 1997 ROD will result in significant additional reduction of copper and zinc loads draining from Iron Mountain in Spring Creek.

### Mass Balance and Metal Transport in Keswick Reservoir

Two different formulations of mass balance are computed using data from this study to evaluate metal transport processes in Keswick Reservoir. The first formulation involves comparison of the sum of two major inputs to the Spring Creek arm of Keswick Reservoir (Spring Creek and Whiskeytown Lake at the Spring Creek Power Plant [SCPP]) with a width- and depth-integrated sample of the Spring Creek arm. The second approach compares inputs and outputs from Keswick Reservoir, using the Spring Creek arm

sample to represent the combined contribution of Spring Creek and the SCPP. The various metal loads from Spring Creek arm are combined with the loads from the site below Shasta Dam and Flat Creek (when available) for comparison with the loads from the outfall from Keswick Reservoir below Keswick Dam.

### Mass Balance in the Spring Creek Arm of Keswick Reservoir

A width- and depth-integrated sample was taken in the Spring Creek arm during five of the six sampling periods for this study (all periods except January 1997). Spring Creek itself was sampled during three periods: December 1996, January 1997, and May–June 1997. Whiskeytown Lake was sampled only on two occasions for this study (December 1996 and May–June 1997), and on neither occasion was a colloid sample taken. Nevertheless, a mass balance for the Spring Creek arm was attempted for both the December 1996 and May–June 1997 periods.

Appendix 3 displays seven tables (one for each element of interest—Al, Cd, Cu, Fe, Pb, Hg, and Zn) that document input loads to the Spring Creek arm of Keswick Reservoir in terms of dissolved, colloidal, dissolved plus colloidal, and whole-water (total recoverable) loads. The sum of Spring Creek and Whiskeytown Lake loads is then compared with the actual loads observed in the Spring Creek arm of Keswick Reservoir. The comparisons at the bottom of tables A3-1 through A3-7 in the section "Mass balance, Spring Creek arm" are expressed as percentages of the loads in the Spring Creek arm of Keswick Reservoir. These data indicate that the balance is closer for the May–June 1997 data set than for the December 1996 data set. The balances for iron and aluminum total recoverable whole-water loads using the May–June 1997 data set are approximately 100 percent, suggesting conservative transport of these elements from the mixing zone (where the acid water from Spring Creek mixes into Keswick Reservoir) to the Spring Creek arm sampling point, located only a few hundred meters downstream (see Alpers and others, 2000). However, for reasons outlined earlier in this report, total recoverable concentration data determined from whole-water samples often under-represent the total load for elements such as iron and aluminum that occur in phases that do not dissolve completely during the total recoverable extraction procedure. The lack of colloid data for the Spring Creek Power Plant input precludes the quantitative analysis of colloid mass balance in this part of the system.

The "dissolved + colloid" category shows balance factors less than 100 percent for the seven metals considered in detail, on both sets of Spring Creek arm samples (December 1996 and May–June 1997), indicating more output than input on these occasions (Appendix 3). Several explanations for these data are possible: (1) possible additional, unsampled input sources of metals to Keswick Reservoir, (2) remobilization of fine-grained sediments, (3) desorption of metals from fine-grained sediments, (4) sampling under nonsteady-state conditions, and (5) errors in flow measurements. Of these, item 2 is a strong possibility. More than 250,000 yd$^3$ of fine-grained, metal-rich sediment have been shown to occur in the Spring Creek arm of Keswick Reservoir (Nordstrom and others, 1999). These sediments have been observed to be mobilized under conditions of fluctuating water levels in Keswick Reservoir and may be scoured by flows from the Spring Creek Power Plant (John Spitzley, CH2M Hill, oral commun., 1998).

### Overall Mass Balance in Keswick Reservoir

Appendix 4 compares metal load data for inputs to Keswick Reservoir and the outfall from the reservoir at Keswick Dam, with the Spring Creek arm sample used as a proxy for the contributions from Spring Creek and the Spring Creek Power Plant. Aluminum and iron balances are nearly 100 percent for the May–June data set for both whole-water and "dissolved + colloid" loads (tables A4-1 and A4-4). Balances for trace metals of most environmental concern in Keswick Reservoir (cadmium, copper, and zinc) indicate a range of values both above and below 100 percent. These results indicate the difficulty and uncertainty in computing mass balances from single instantaneous measurements of concentration and discharge. A much more sound approach would be to integrate samples over longer time intervals, such as the duration of a storm event or on a monthly basis. Three of the four sampling periods with adequate data for the instantaneous calculations show apparent copper attenuation (balance values > 100 percent) despite the fact that iron and aluminum transport for the same periods indicate little to no precipitation of these major elements. The apparent copper attenuation effect could be caused by increased sorption of copper during transport through Keswick Reservoir.

Zinc and cadmium loads for the mass balance behaved quite similarly to each other, the one exception being some apparent attenuation of cadmium more than zinc during November 1996. Both cadmium and zinc were attenuated less than copper, which is consistent with overall patterns of metal transport (figs. 5, 6, 9, 11, 12, and 15–18) and their chemical properties (Dzombak and Morel, 1990; Smith, 1999).

An overall hypothesis that is broadly consistent with the data from this study is that the cadmium, copper, and zinc loads at Shasta Dam are roughly half of the corresponding values at Keswick Dam throughout all sampling periods. The contribution from Spring Creek was minimal in July and September 1996, so the Shasta Dam contribution was proportionally higher during these periods; however, the overall pattern of metal loads is consistent with the hypothesis. The instantaneous total (dissolved plus colloidal) copper loads at Shasta Dam ranged from 7 kg/d (November 1996) to 180 kg/d (December 1996), with no data available for January 1997. Total recoverable (whole-water) loads ranged from 14 kg/d to 320 kg/d. These data are in an appropriate range to be consistent with the Regional Water Board's estimate of 50 kg/d for an annualized average copper contribution from the mines above Shasta Lake (Dennis Heiman, Regional Water Quality Control Board, Central Valley Region, written commun., 1998).

## Metal Sources Downstream of Keswick Dam

In this section, spatial variations in metal loads in the Sacramento River are described to identify river reaches with likely metal sources. The discussion is organized into two subsections. The first describes tributaries in the reach between Keswick Dam and Colusa, and the second describes the reach between Colusa and Freeport (including the Yolo Bypass). The second subsection is further subdivided into sections on metal contributions from agricultural drainage and urban runoff.

### Tributaries Between Keswick Dam and Colusa

Although no water samples were collected for this study from tributaries between Keswick Dam and Colusa, the data collected from the mainstem Sacramento River samples clearly indicate that such tributaries and(or) other pollution sources make significant contributions to metal loads in the watershed. Copper and lead loads both increased consistently and substantially between Keswick Dam

and Colusa during each sampling period; a single exception was an apparent decrease in copper load between Bend Bridge and Colusa during November 1996 (fig.12). In addition, lead isotope data (Alpers and others, 2000) indicate that sources of lead other than the massive sulfide mineralization upstream of Keswick Dam tend to dominate lead transport in the Sacramento River downstream of Bend Bridge, near Red Bluff (fig. 1).

During some sampling periods, the load increases were more dramatic between Keswick Dam and Bend Bridge, and during others, between Bend Bridge and Colusa, suggesting that multiple tributaries are involved and that the nature of the contributions is highly variable and perhaps seasonal in nature. In all cases, the increased copper and lead loads are associated exclusively with colloidal forms as opposed to dissolved metals (figs. 12 and 14).

A tributary that may be responsible for increasing metal loads between Keswick Dam and Bend Bridge is Cottonwood Creek (fig. 1). Sampling of Cottonwood Creek (at Interstate 5) by the Regional Water Board revealed the following elevated concentrations of total recoverable copper: 16 and 25 µg/L on April 2, 1996 and June 3, 1996, respectively, with less than 1 to 2 µg/L "dissolved" copper in 0.45-µm filtrates (Regional Water Quality Control Board, Central Valley Region, written commun., 1998). Total recoverable zinc was also elevated in the Cottonwood Creek samples, with concentrations of 24 and 52 µg/L, respectively, on the two sampling dates mentioned earlier. Some other tributaries draining the west side of the Sacramento Valley between Redding and Colusa are Clear Creek, Elder Creek, Thomes Creek, and Stoney Creek (some are shown on fig. 1). Very limited water-quality data are available for these creeks, especially during high flow. Additional monitoring on some of these tributaries is planned by the California Department of Water Resources.

Another tributary in this area for which elevated copper concentrations have been reported is Cow Creek, one of several tributaries draining the east side of the Sacramento Valley between Redding and Red Bluff. Samples taken from Cow Creek (at Dersch Road) during April and June, 1996 showed 5.2 and 6 µg/L of copper, respectively (Regional Water Quality Control Board–Central Valley Region, written commun., 1998). The same samples contained 16 to about 170 µg/L of zinc. Some of the massive sulfide mineralization and inactive/abandoned mines in the East Shasta mining district (for example, near the town of Ingot, California) are in the Little Cow Creek drainage, a tributary to Cow Creek (Nordstrom and others, 1977; Alpers and others, 2000).

### Tributaries Between Colusa and Freeport

The principal tributaries to the Sacramento River between Colusa and Freeport are agricultural drains such as the Colusa Basin Drain and the Sacramento Slough, and major rivers including the Feather and American rivers. In addition, there are several smaller tributaries that convey urban runoff.

#### Agricultural Drainage

Sampling periods in July and September 1996 and in May–June 1997 showed increases of dissolved and colloidal copper loads, and also in colloidal zinc loads in the Sacramento River between Colusa and Verona. This is the reach in which the Colusa Basin Drain and the Sacramento Slough and other agricultural return flows enter the river (fig. 1). Copper transport from rice fields may be significant because large quantities of copper sulfate and other forms of copper are applied from May through June each year to control algae in the flooded fields.

Metal loads from agricultural drainage were evaluated for this study by means of a single sample from the Colusa Basin Drain during rice-field draining in early June 1997. The Colusa Basin Drain is estimated to represent about one-third of the agricultural drainage in the Sacramento Valley. Concentrations of copper in whole-water samples from the Colusa Basin Drain were 16–17 µg/L; 0.45-µm filtrates had about 3 µg/L, and 0.005-µg-equivalent ultrafiltrates had 1.3 µg/L (Alpers and others, 2000). Dissolved organic carbon was relatively high (4.8 mg/L) in the water sample from the Colusa Basin Drain, suggesting that the dissolved copper in the ultrafiltrate may be organically complexed. The sequential extraction data indicate that about half of the colloidal copper from the Colusa Basin Drain sample was extracted in the reducible fraction, similar to other samples from the watershed (fig. 18B) and that only a small fraction is oxidizable, suggesting that any organically complexed copper is not manifested to a great extent in the colloids. However, it is possible that some copper associated with organic matter is removed in the first (reducible) step of the sequential extraction procedure. Additional work would be needed to resolve this point.

Biweekly monitoring of the Colusa Basin Drain by the NAWQA Program using 0.45-μm filtrates showed increased copper concentrations during the growing season relative to the rest of the year, reaching 6 μg/L in May 1996 and 3 to 4 μg/L in June 1997. Copper concentrations in 0.45-μm filtrates from the Sacramento Slough reached a maximum of 4 μg/L in December 1996. However, more studies of copper in agricultural drainage are needed before any significant conclusions can be reached.

The total (dissolved plus colloidal) loads of copper from the Colusa Basin Drain in the May–June 1997 sampling period were 18 kg/d, representing 19 percent of the total copper load at Freeport during that sampling period (tables A1-3 and A2-3). For comparison, the copper loads in the Spring Creek arm of Keswick Reservoir were 20 kg/d during the same sampling period, and the total (dissolved plus colloidal) copper load below Keswick Dam was 45 kg/d, representing 42 percent of the load at Freeport (tables A1-3 and A2-3). In contrast, the copper load of Spring Creek during the January 1997 flood was about 1,100 kg/d, and the load in the Yolo Bypass was about 7,700 kg/d (table A1-3). On an annual basis, most of copper and zinc loads appear to enter the Sacramento River upriver of Colusa, which is upstream of the influence of most intense agricultural drainage return flows in the Sacramento River watershed. This apparent increase in metal loading is primarily related to the consistent increase in colloid concentration between Bend Bridge and Colusa (figs. 12 and 15), which remains unexplained.

### Urban Runoff

Urban runoff can be an important source of metals to surface waters. For example, the Rhine River at the Dutch-German border contained elevated concentrations of As, Cd, Cr, Cu, Pb, Hg, Ni, and Zn that were reduced dramatically between 1975 and 1985 (Holland and Petersen, 1995). Awareness is increasing of nonpoint source problems associated with metals in urban settings, such as the use of copper in brake pads.

Although no sampling sites in this study were dedicated to urban runoff, the USGS's NAWQA Program sampled Arcade Creek in the City of Sacramento on a monthly basis for 2 years as an indicator site for urban runoff. Somewhat elevated concentrations of copper, lead, nickel, and zinc were observed in 0.45-μm filtrates, however metals were not analyzed in unfiltered samples.

Concentration profiles of lead in sediment and lead in colloids shown in the Volume 1 report from this study (Alpers and others, 2000) indicate an increase in lead concentrations downstream of Colusa. The most likely source of this increase is urban runoff, possibly related to atmospheric (wet and dry) deposition. Lead isotope data (Alpers and others, 2000) indicate a distinctly radiogenic source for the colloidal lead sampled from the Sacramento River at Tower Bridge in January 1997. During that high-flow condition, most of the water in the mainstem Sacramento River was derived from the American River, and the majority of the flow from the Sacramento River upstream of Colusa was diverted into the Yolo Bypass. Therefore, the anomalously radiogenic lead isotopes are probably related to granitic source rocks in the American River drainage, although anthropogenic sources for this lead have not been ruled out.

## Summary and Conclusions

The transport of metals in the Sacramento River from Shasta Dam to Freeport was evaluated from July 1996 through June 1997 using an approach that quantified the dissolved and colloidal concentrations and corresponding loads of metals at six sites on the Sacramento River during six sampling periods. Although the water year corresponding to this study was unusual in that most precipitation fell during December and January, a major flood occurred in January, and it was very dry from February to May, so the overall amount of precipitation was close to that expected for a normal year. Most discharge in the river occurred during the 3-month period of December 1996 through February 1997, and a higher proportion of the total annual metal fluxes or loads took place during the same time frame because of the increased transport of suspended sediment and associated higher metal concentrations associated with high flow. In a more typical wet season, the increased discharge and metal fluxes would occur over a longer period, for example December through April or May.

The mineralized area upstream of Keswick Dam is an important source of various metals, especially cadmium, to the lower Sacramento River and the Bay–Delta. This was determined by comparing the metal loads at the sampling site below Keswick Dam with those measured in the Sacramento River at Freeport and the Yolo Bypass. However, it was also determined that the loads, during the period of this

study, were highly dependent on the flow regime. For example, the proportions of mineralization-related trace-metal loads (percentages representing dissolved plus colloidal loads at Keswick Dam divided by the sum of dissolved and colloidal loads at Freeport and the Yolo Bypass, when flowing) observed during moderately high flows in December 1996 were: cadmium, 87 percent; copper, 35 percent; lead, 10 percent; and zinc, 51 percent. During the flood conditions of early January 1997, the percentages were: cadmium, 22 percent; copper, 11 percent; lead, 2 percent; and zinc, 15 percent. During the irrigation drainage season of May through June 1997, the percentages were: cadmium, 53 percent; copper, 42 percent; lead, 20 percent; and zinc, 75 percent. These estimates must be qualified by the following factors. First, metal loads at Colusa in December 1996 and at Verona in May–June 1997 generally exceeded those determined at Freeport at that time; therefore, the above percentages represent maximum estimates of the proportion of metals from mineralized areas upstream of Keswick Dam. Second, for logistics reasons, the Sacramento River was sampled at Tower Bridge instead of Freeport during January 1997. However, no other significant input of water to the Sacramento River, other than irrigation return flows, occurs between Tower Bridge and Freeport. The results indicate the importance of the mineralized areas for the transport of cadmium, copper, and zinc, but also show that lead must enter the Sacramento River mainly from other areas, as confirmed by lead isotope analyses. The results also indicate that the mineralized areas upstream of Keswick Dam are the source of a significant amount of metals to the lower Sacramento River; however, other areas along the river also must contribute significant amounts especially during extreme high-flow conditions.

The available data suggest that trace-metal loads from agricultural drainage may be significant during the growing season, but that more studies are needed before a definitive conclusion can be reached. Copper transport from rice fields may be significant because large amounts of copper sulfate and other forms of copper are applied from May through June each year to control algae in the flooded fields. Metal loads for sampling periods in July and September 1996 and in May–June 1997 showed increases of dissolved and colloidal copper and also in colloidal zinc in the Sacramento River between Colusa and Verona, the river reach along which the Colusa Basin Drain and the Sacramento Slough and other agricultural return flows enter the river. Also, water-quality sampling by the NAWQA Program shows that copper concentrations in 0.45 $\mu$m-filtered water samples from the Colusa Basin Drain are elevated during the period of copper application to the fields relative to the rest of the year. To put the copper loads associated with agricultural drainage in perspective, the total (dissolved plus colloidal) loads of copper from the Colusa Basin Drain in June 1997 were 18 kg/d, representing about 19 percent of the total copper load at Freeport, whereas the copper loads from Iron Mountain by way of the Spring Creek arm of Keswick Reservoir were 20 kg/d during the same sampling period, and the copper load below Keswick Dam represented about 42 percent of the total (dissolved plus colloidal) copper load at Freeport. In contrast to these relatively low-flow conditions, the copper load of Spring Creek during the January 1997 flood was about 1,100 kg/d and the copper load in the Yolo Bypass was about 7,700 kg/d. On an annual basis, most of copper and zinc loads appear to enter the Sacramento River upriver of Colusa, which is upstream of the influence of most intense agricultural drainage return flows in the Sacramento River watershed. Some of the metal loads are clearly from well known, mining-related sources of cadmium, copper, and zinc above Keswick Dam; however, large increases in metal loads associated with elevated colloid concentrations at Colusa remain unexplained.

This study has demonstrated that some trace metals of environmental significance (cadmium, copper, and zinc) in the Sacramento River are transported largely in dissolved form at upstream sites (below Shasta Dam, below Keswick Dam, and at Bend Bridge) proximal to the mineralized areas of the West Shasta and East Shasta mining districts. Despite continuous water treatment that has removed 85 to 90 percent of the cadmium, copper, and zinc from the mine drainage at Iron Mountain since 1994, Spring Creek remains an important source of these metals to the Sacramento River system. In the acidic water of Spring Creek, cadmium, copper, and zinc are transported almost exclusively in dissolved form. In contrast, these trace metals are transported largely in colloidal form at downstream sites (Colusa, Verona,

Freeport, and Yolo Bypass). Aluminum, iron, and lead were observed to be transported predominantly in the colloidal phase at all mainstem Sacramento River sampling sites during all sampling periods in this study.

Speciation of metals on the colloid particles, which is based on sequential chemical extractions, showed that metals are variably distributed among reducible, oxidizable, and residual phases. Further research is needed to address the biological significance of metals associated with these phases in the Sacramento River system.

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