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Contamination of two Oregon reservoirs by cinnabar mining and mercury amalgamation

R.K.R. Ambers · B.N. Hygelund

Abstract Two reservoirs in western Oregon contain mercury-contaminated sediment and fish as a result of historic mercury mining in the Cottage Grove Lake watershed and mercury amalgamation used in gold mining in the Dorena Lake watershed. On average, sediment in Cottage Grove Lake contains ten times as much mercury as sediment from Dorena Lake (2.720 versus 0.242 ppm). Mercury content in Cottage Grove Lake sediment shows a sharp initial decrease and leveling off with time that reflects the end of the major cinnabar mining phase; deposition of other heavy metals appears to be linked to the clay content of sediment. Mercury input to Dorena Lake has remained fairly constant with time, but small increases in mercury are associated with the deposits of large floods. Copper, lead, and zinc input to Dorena Lake exhibits a marked decrease and leveling off related to the end of commercial mining for these metals.

Keywords Cinnabar mining · Gold mining · Lake sediment · Mercury

Introduction

Mercury contamination of lakes and rivers is widespread around the world and poses a serious threat to the health of ecosystems and human populations. Although natural sources of mercury exist, contamination has frequently been associated with (1) liquid effluent from factories, such as pulp mills and chloro-alkali plants, (2) atmos-

pheric deposition related to coal-burning power plants, waste incinerators, etc., and (3) mining of mercury and gold deposits (Fergusson 1982; Lacerda and Marins 1997). In the case of gold mining, mercury is used to amalgamate with and remove gold from primary and secondary ores, a technique that has been widely utilized for over 2000 years. Many regions have experienced serious mercury contamination problems as a result of this practice, including the Amazon region of South America and historic gold mining areas in the United States (Miller and others 1995; Leigh 1997; Villas Bôas 1997; Lacerda and Salomons 1998). Inorganic forms of mercury in the environment are problematic because they tend to be converted to methylmercury, a highly toxic, fat-soluble compound that bioaccumulates rapidly and often concentrates in fish. Factors found to encourage methylation of mercury in freshwater lakes and rivers include high concentrations of organic matter, slightly acidic water, and high levels of microbial activity, particularly sulfate-reducing bacteria (Gilmour and others 1992; Lacerda and Salomons 1998).

Oregon is one of 35 states with fish consumption advisories for mercury, and these are in effect on four man-made reservoirs, as well as natural water bodies (Oregon Health Division 1996). Many mercury sources in Oregon are natural deposits associated with volcanic rocks. In some cases, however, historic and modern-day mining operations have spread mercury-rich mine waste or elemental mercury across the landscape, converting areas with naturally high background concentrations into full-scale pollution problems.

Two of Oregon's mercury-contaminated reservoirs, Dorena Lake (DL) and Cottage Grove Lake (CGL), are located in the west-central part of the state within 25 km of each other (Fig. 1). Fish in the two lakes, as well as bass and squawfish from the entire Willamette River downstream of the dams (Oregon Health Division 1997a), contain elevated levels of mercury. In CGL, 0.3 to >1.0 ppm mercury has been observed in sport fish tissue, and an average of 0.37 ppm was found in DL fish (a consumption advisory is issued when the concentration is ≥ 0.35 ppm; Oregon Health Division 1993; Oregon Health Division 1997b; Park and Curtis 1997). The watersheds of both reservoirs are impacted by relatively small-scale mines that opened during the late 19th century. In the CGL watershed, cinnabar (HgS) has been mined intermittently since 1890 at Black Butte to produce quicksilver

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(elemental mercury). In the DL watershed, quicksilver was used extensively for over 60 years for gold and silver recovery from lode deposits in the Bohemia Mining District.

This study was undertaken to characterize and compare the contamination problems in these two similar reservoir systems and to gain insight into the impact of mining activity and flood events on mercury accumulation. To address these points, the acid-extractable, trace-metal concentrations of several sediment cores from each reservoir were examined. This work expands upon previous studies (Park 1996; Park and Curtis 1997) and forms a significant basis for further research.

Study area

Reservoirs

Both Cottage Grove and Dorena Lakes are US Army Corps of Engineers (USACE) projects constructed as part of the Willamette Valley flood-control plan. Cottage Grove Lake, completed in 1942, is 10 km south of the town of Cottage Grove, and it regulates the headwaters of the Coast Fork of the Willamette River. Dorena Lake, completed in 1949, regulates a Coast Fork tributary named the Row River and is 10 km east of Cottage Grove. DL is deeper and larger than CGL and has a larger watershed (Table 1); but climate, land use, and vegetation in the two watersheds are quite similar. Although flood control during the rainy season (November to May) is the main function of the reservoirs, they also conserve water during the summer for recreation and for possible use in irrigation and pollution dilution on the Willamette River. Both lakes are very popular with local residents, together having more than 760,000 recreational visits annually (Oregon Water Resources Department 1999). Management of these reservoirs follows a distinctive annual cycle. As Fig. 2 illustrates, lake level is kept at summer high-pool elevation from late May to early September. Drawdown then begins so that winter low-pool elevation is reached by the end of October. When a storm occurs, water is held back in the reservoir to keep outflow below a certain discharge, and the lake fills partially or completely, depending on the size of the event. After the storm, water is drained over a period of days or weeks to reach the appropriate lake level. From February to May, the lakes are slowly filled to their summer high-

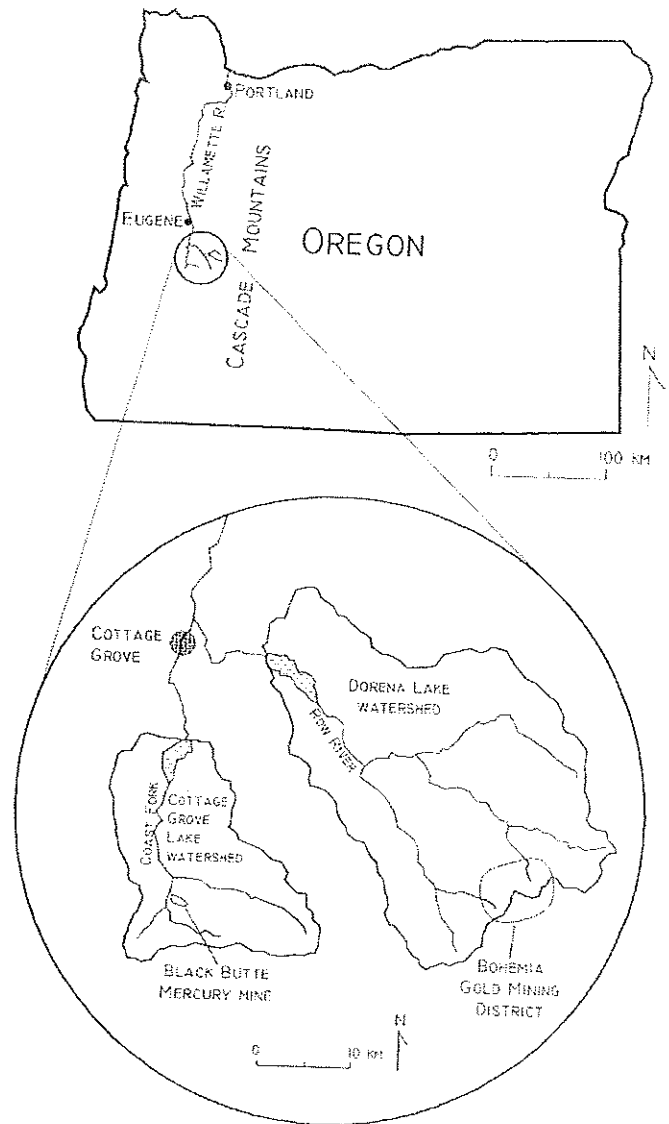


Fig. 1 Map showing the location of Dorena and Cottage Grove Lakes in Oregon. Watershed boundaries, major rivers and creeks, and mining areas are also shown

pool elevations again. As a result of these management practices, the lake bottom experiences annual and, during the rainy season, storm-related transgression-regression events. Sediment influx is closely tied to flood events, so it is also seasonal. A large proportion of the lake bottoms are exposed during much of the rainy season, allowing some plant growth. As the exposed sediments drain, a vadose zone develops, causing oxidization in the upper layers of the otherwise reduced sediment.

Mines

Black Butte, Oregon's fourth largest quicksilver-producing mine, is approximately 13 km upstream of Cottage Grove Lake (Fig. 1). Cinnabar was mined and milled on-site there during 1890-1908, 1916-1919, 1927-1942,

Table 1

Characteristics of the study reservoirs

Reservoir	Capacity (m ³)	Area at Full Pool (km ²)	Watershed size (km ²)
Cottage Grove Lake	40.6 × 10 ⁶	4.9	269
Dorena Lake	95.6 × 10 ⁶	7.4	686

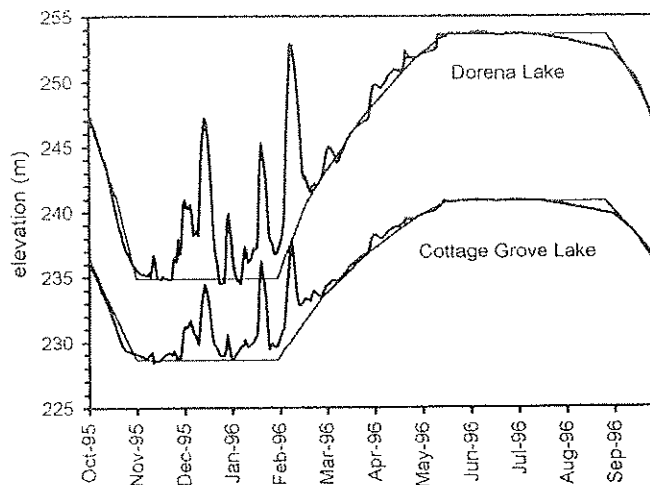


Fig. 2

Lake levels at Dorena and Cottage Grove Lakes during Water Year 1996 (*thick lines*). Rule curves (*thin lines*) are also shown to illustrate the daily lake elevations USACE tries to maintain. Peaks in elevation during winter and spring months represent storm flows temporarily held in the reservoir to prevent flooding downstream

1956–1957, and 1967–1968 (Brooks 1963; Brooks 1971). Evidence of the level of pollution already emanating from Black Butte after only a few years of production can be found in a University of Oregon senior thesis written near the turn of the century (Hardin 1909). Although Hardin was speaking of economic rather than environmental problems, he recognized that “the prevailing trouble at the mine lies principally in its [ore furnace] condensers. ... This was shown to be true by small globules of mercury settling on the fir trees farther up the mountain” (p. 22). He also noted that the tailings and waste rock piles served as a supply of aggregate for surfacing the 2.4-km road leading up to the mine (Hardin 1909). According to Black Butte production records from the US Bureau of Mines (Brooks 1963) and estimates of furnace/condenser efficiency provided by Hardin (33% during 1900–1907 and $\leq 87\%$ during 1908–1909), by 1910, approximately 26 tons of mercury had been lost to the atmosphere and dumped on tailings piles. Estimates of furnace and condenser efficiency are unavailable for the later years of mine operation, but if the average rate were 95–98% (assuming a considerable improvement in condenser technology with time), 10–27 additional tons of mercury would have been lost by 1958. At least 35 to 50 tons of mercury were thus likely lost in processing, and an unknown quantity was dumped in mine waste-rock piles.

The Bohemia Mining District, the largest and most productive gold and silver mining area in the Western Cascades (Brooks and Ramp 1968), is ~ 30 km upstream of Dorena Lake (Fig. 1). Analyses of watershed stream sediments and mine waste rock/tailings indicate that the mining district is the source of mercury contamination in the DL watershed (Hygelund 2000). Many different mine

and mill sites have been developed in the district since its discovery in 1858. The area was exploited first for gold and silver, both from placer and underground deposits, and later for its copper and lead resources; zinc is also present in abundance. The bulk of mining activity in the district occurred before World War II, and large-scale commercial mining has not resumed since that time (Brooks and Ramp 1968), although hobby mining is popular.

Mercury mineralization has not been reported within the Bohemia District, but elemental mercury was used in stamp mills from 1872 to the late 1930s to extract gold and silver from ore rock (Honey 1980). These mills crushed the ore and ran it over mercury-coated plates to capture gold and silver by amalgamation. The amalgam was later collected and heated to vaporize the mercury and recover the precious metals. Although attempts were made to re-condense and reuse mercury in this method, losses to the atmosphere, spills, and dumping of mercury were inevitable given the technology available at the time. Exactly how much mercury was lost is unknown, however, and the extent of soil contamination around former stamp mill sites has never been determined. A rough estimate of mercury loss can be obtained by applying a mercury emission factor to precious metal production. Studies of amalgamation methods have found that 1.0–1.7 kg of mercury is usually lost during production of 1 kg of gold or silver, and 65–83% of that mercury is lost to the atmosphere (Lacerda and Salomons 1998). The Bohemia District produced slightly more than 1 ton of gold and silver from 1880 to 1930 (Brooks and Ramp 1968), so approximately 1.0–1.7 tons of mercury may have been emitted during that time, most of it to the atmosphere. The origin of much of the mercury used in the stamp mills was the nearby Black Butte Mine (Prospectors Club 1993), so, ironically, the contamination in both the DL and CGL watersheds has the same geological source.

Methods

A total of seven lake-sediment cores were taken from the two reservoirs (Fig. 3). A set of two, meter-long cores from each lake (CGL-1, CGL-2, DL-1, and DL-2) was taken from the lake-bottom areas that are exposed during the winter, and selected samples were also analyzed from a 2-m core from DL (DL-4). Coring was performed with an Eijkelkamp gouge auger, which produces a 6-cm diameter sediment core. An additional core from each lake (DL-3 and CGL-3, both slightly more than 0.5 m long) was taken by boat near the delta that has formed where the main river meets the winter low pool. To obtain these subaqueous cores, a homemade corer with a detachable, 6-cm diameter PVC barrel was used. All the cores represent the complete thickness of lake sediment at the sampling location, as verified by the presence of pre-lake soil or river gravel, in case of DL-1 and DL-2, at the base of the cores.

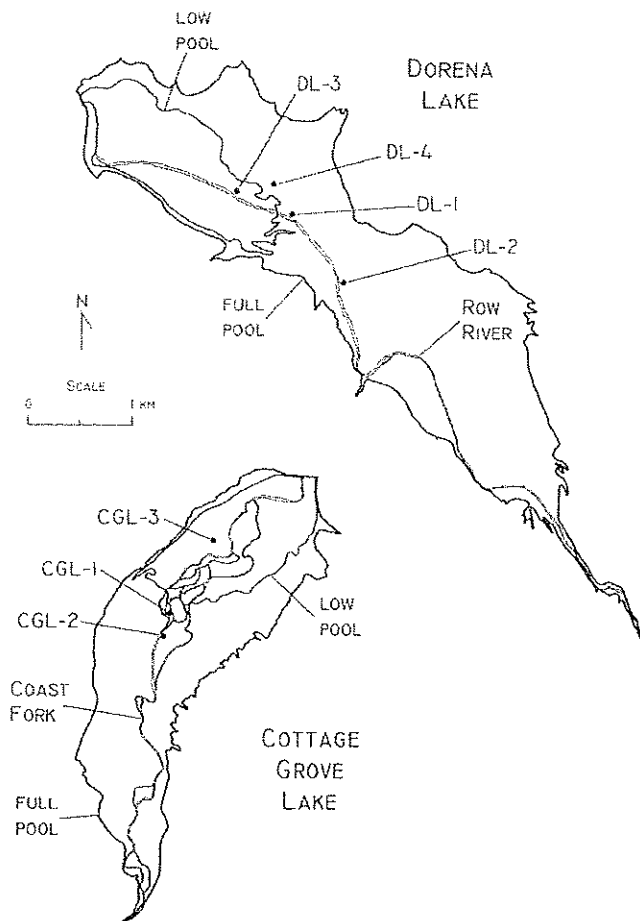


Fig. 3

Core locations in Dorena and Cottage Grove Lakes. The outlines of the full pool and winter low pool are shown for each reservoir, as well as the original channels of the Row and Coast Fork Rivers that feed the lakes. Both rivers flow in a northerly direction

Once a core was obtained, it was immediately cut into 5- to 7-cm-long sections, which were placed in plastic, reclosable freezer bags. Back in the lab, the bags were opened and placed on a tray above a warm furnace to allow the sediment to dry (maximum temperature did not exceed 50 °C). Dry samples were broken up in their bags with a rubber mallet, and the relatively fine-grained sediments were sieved to remove organic material coarser than 0.5 mm (1.0 ϕ or medium sand). Splits of the samples were then lightly ground in a tungsten carbide shatter box. The ground splits were sent to an analytical testing company for elemental analyses. Gold analyses (only done on DL-1 and DL-2 samples) were performed by fire assay pre-concentration followed by atomic absorption (lower detection limit = 5 ppb). For other metals (Al, Sb, As, Ba, Bi, Cd, Ca, Cr, Co, Cu, Fe, La, Pb, Li, Mg, Mn, Mo, Ni, Nb, K, Sc, Ag, Na, Sr, Ta, Te, Sn, Ti, W, V, Y, Zn, and Zr), the samples were first digested in aqua regia (3:1 HCl:HNO₃) then analyzed by inductively coupled

plasma atomic emission spectroscopy (lower detection limits vary from 0.2 to 100 ppm depending on the element). For mercury, analyses were performed by cold vapor atomic absorption after aqua regia digestion (lower detection limit = 0.010 ppm). Mercury was the only element analyzed in DL-4 samples.

With the use of aqua regia digestion, carbonates, hydroxides, most sulfide and oxide minerals, and base metals are readily dissolved, but silicates may be only slightly or moderately digested. For this reason, results for metals other than Au, Ag, As, Bi, Cd, Cu, Hg, Pb, and Zn represent only a portion of the actual amounts present in the samples and so cannot be directly compared with data obtained by different analytical methods.

Particle-size analyses to determine the percentages of sand, silt, and clay were performed on un-ground splits of the core samples using the hydrometer method described by Gee and Bauder (1986).

Results

Mercury

Mercury concentrations in the three cores from CGL show a consistent pattern between cores and through time (Fig. 4a). Mercury values range from 5.143 to 11.605 ppm at the bottom of the lake sediment, decrease sharply, and then level off between 0.835 and 2.677 ppm in the top half of the cores. Mercury shows a slight increase in the top two CGL-3 samples, but values in the other cores remain constant. The average mercury content of all the lake sediment samples is 2.720 ± 2.475 ppm ($n=39$); but if only the samples from the top 60% of the cores are averaged, the mean is 1.452 ± 0.526 ppm ($n=26$). Pre-lake soil samples at the base of CGL-2 and CGL-3 have the highest mercury concentrations of each core (20.440 and 9.099 ppm, respectively). In contrast, the pre-lake soil in CGL-1 has the lowest concentration of any CGL sample (0.760 ppm).

On average, mercury concentrations in the DL cores are an order of magnitude lower than those from CGL and show a much different pattern (Fig. 4b). Pre-lake soil values from DL-3 and DL-4 (0.042 and 0.052 ppm, respectively) are less than one-third the lowest mercury value in the rest of the DL cores. All the mercury concentrations in the lake sediment fall within the range of 0.155 to 0.362 ppm, with the exception of the sample at the base of DL-2 that contains 0.723 ppm. The mean value for lake sediment is 0.242 ± 0.085 ppm Hg ($n=49$). Unlike the CGL cores, no simple, consistent trend is evident in mercury concentration (Fig. 4b). There is an initial decrease in mercury in the bottom third of DL-1, DL-2, and DL-3, followed by a moderate mercury peak in all the cores at around 40–50% thickness. Additional peaks in mercury concentration occur at 73–77% and 93–95% thickness in DL-2 and DL-3. Stratigraphically, these higher-mercury samples are found within sediment layers from 1964–1971, 1981–1986, and 1996–1999, which con-

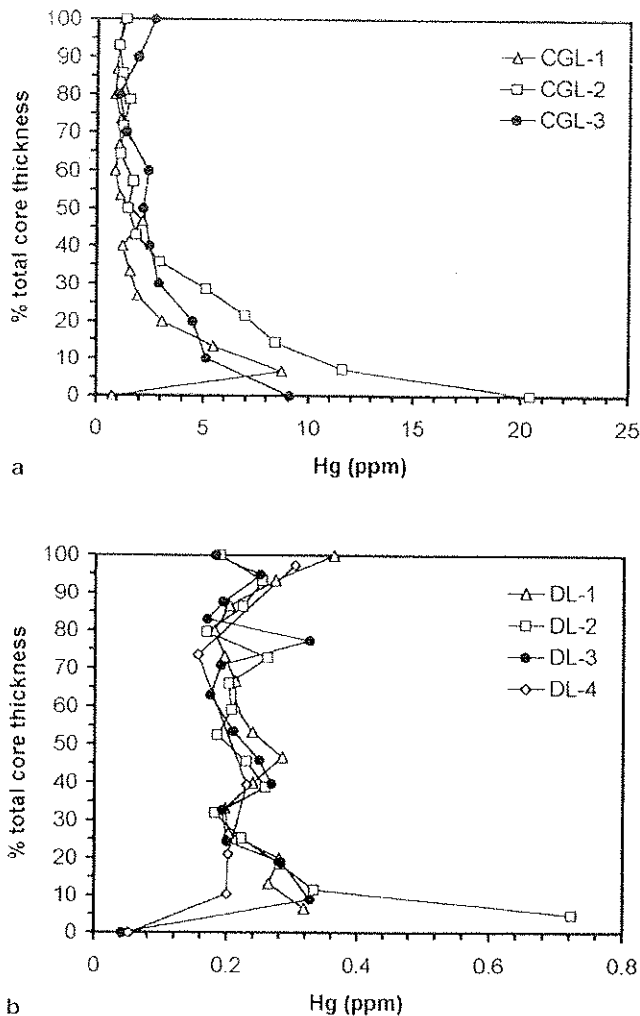


Fig. 4

Mercury concentration in samples through a three CGI cores and b four DL cores. Pre-lake soil samples plot at 0% total core thickness, and the tops of the cores are at 100%. *Open symbols* represent cores taken above the low pool elevation; *filled symbols* designate subaqueous cores

tain deposits from the three largest floods in the history of the reservoir (>150-year flood of December 1964, 25-year flood of December 1981, and 100-year flood of November 1996; Ambers 2000). The relatively high mercury samples at the top of DL-1 and DL-4 were deposited during 1996–1999, so these cores have higher mercury concentrations only in the deposits of the two largest events.

Other metals

All the cores except DL-4 were analyzed for 33 elements in addition to mercury; DL-1 and DL-2 were also analyzed for gold. Only a few of the metals show any significant variation through the cores, so the present discussion will be limited to those elements. The complete data set is available in the electronic material supplementary to this article.

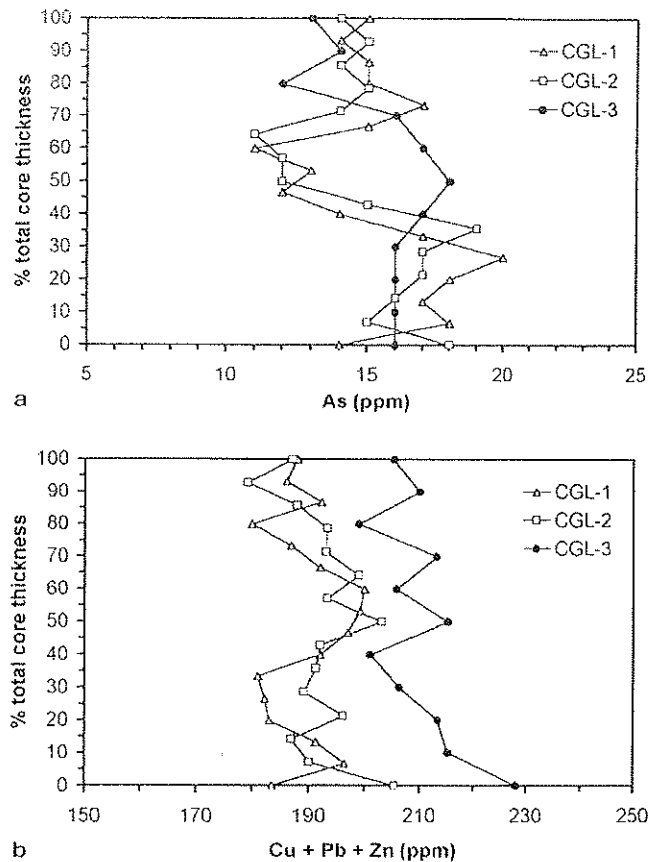


Fig. 5

Variation in a arsenic concentration and b copper + lead + zinc concentration through the CGI cores. CGL-3 shows higher values than CGL-1 and CGL-2 in b mainly because its copper concentrations are higher than in the other two cores

In the CGI watershed, the only significant mining area beside Black Butte is a mine on Hobart Butte that has produced high-alumina clay and is known to contain a small amount of realgar (AsS) (Brooks 1963). Hydrothermal alteration at Black Butte produced mainly cinnabar, iron carbonates, chalcedony, and small amounts of oxidized sulfides (Brooks 1963). In CGI sediment, arsenic concentration varies by nearly 10 ppm through the cores, with CGL-3 showing a very different pattern compared with CGL-1 and CGL-2, particularly between 30–70% thickness (Fig. 5a). The sum of copper, lead, and zinc oscillates around 190 ppm in CGL-1 and CGL-2, and around 210 ppm in CGL-3 (Fig. 5b). CGI sediment contains equivalent arsenic (11–20 ppm), less zinc (94–107 ppm) and lead (0–9 ppm), but more copper (85–101 ppm in CGL-1 and CGL-2, 100–124 ppm in CGL-3) than DL sediment. No analyzed element in the lake sediment correlates strongly with mercury, although zinc and arsenic are moderately correlated with mercury ($r^2=0.17$ and 0.15 , respectively). Zinc also correlates moderately with copper ($r^2=0.23$).

In the DL watershed system, the source of mercury is the Bohemia Mining District, which is known to contain high

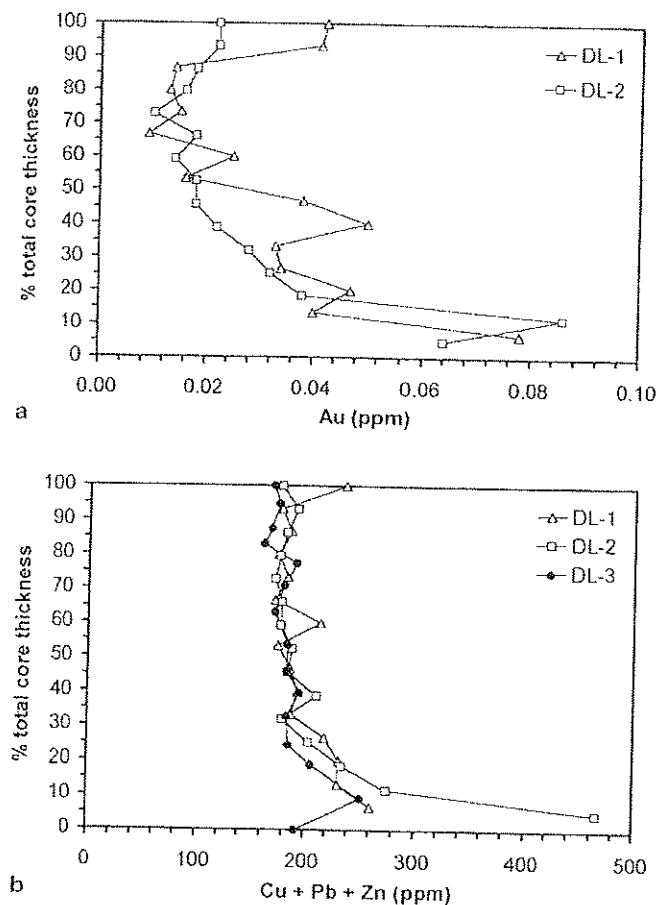


Fig. 6

Variation in a gold concentration and b copper + lead + zinc concentration through the DL cores

levels of gold, silver, copper, lead, and zinc from epithermal to mesothermal mineralization (Brooks and Ramp 1968; Hygelund 2000). Gold values generally decrease upward in the cores with a slight increase at the top, although the pattern in DL-2 is much smoother than that of DL-1 (Fig. 6a). The sum of copper, lead, and zinc concentrations behaves similarly in all three cores, decreasing upward in the bottom third and then remaining relatively constant in the top two-thirds (Fig. 6b). Excluding the sample at the base of DL-2, zinc concentrations are the highest of the mining-district metals (109–158 ppm), followed by copper (36–67 ppm), lead (6–54 ppm), and gold (0.009–0.086 ppm). Silver was detected in only two samples from DL-1 (0.3 ppm) and three samples from DL-2 (0.2–0.7 ppm; lower limit of detection = 0.2 ppm). Compared with other cores, the top sample of DL-1 contains relatively high concentrations of most of the heavy metals; but the base of DL-2 contains more silver, zinc, copper, and lead than any other sample (Ag = 0.7 ppm, Zn = 210 ppm, Cu = 112 ppm, and Pb = 144 ppm). The pre-lake soil of DL-3 is unusually high in cobalt, chromium, nickel, manganese, titanium, and iron and low in lead compared with all the other DL samples. When all

the DL sediment data are considered together, copper, lead, and zinc values correlate strongly with each other ($r^2 = 0.70$ – 0.75). The sum of copper, lead, and zinc correlates strongly with mercury and with gold ($r^2 = 0.84$ and 0.45 respectively), and gold correlates moderately with mercury ($r^2 = 0.36$). When the unusual sample at the base of DL-2 is excluded from the latter two regressions, the correlations improve markedly ($r^2 = 0.70$ for Au and Cu + Pb + Zn; $r^2 = 0.50$ for Au and Hg).

Sediment particle size

The texture of CGL-1 and CGL-2 samples are very similar, mostly loams and silt loams, with clay content ranging from 17–30%, silt from 40–57%, and sand from 14–43%. In the subaqueous CGL-3 core, the samples are finer, silty clays and silty clay loams containing 29–49% clay, 50–71% silt, and 0–7% sand. None of the pre-lake soils are texturally distinct from the lake sediments. Clay content has a weak correlation with mercury concentrations when all the data points are regressed ($r^2 = 0.039$); but if high mercury values from the base of the cores are excluded, the correlation becomes moderate ($r^2 = 0.39$). Correlation between clay content and the sum of copper, lead, and zinc concentrations is strong ($r^2 = 0.69$). The particle-size of DL sediment does not vary a great deal. Most samples are either loams or silt loams with a clay content of 9–42%, silt of 42–67%, and sand of 3–45%. In contrast, the pre-lake soil of DL-3 is a clay, and the base of the 1964 flood deposit is a sandy loam. On the whole, DL-3, the subaqueous core, is somewhat finer than the other cores, and DL-2 contains slightly more clay than DL-1. Clay content has very weak correlation with mercury ($r^2 = 0.0002$) and with the sum of copper, lead, and zinc concentrations ($r^2 = 0.005$).

Discussion

Heavy metal contamination

Guidelines developed by the US Geological Survey for the Willamette Basin confirm that mercury concentrations in lake sediment from both CGL and DL are enriched above background levels for the region (breakpoint value = 0.11 ppm Hg for $< 62 \mu\text{m}$ sediments; Rickert and others 1977; Rinella 1998). Copper in CGL and arsenic in both lakes are also enriched (> 50 ppm Cu and > 10 ppm As), presumably by natural causes, as suggested by other workers (Hinkle and Polette 1999). In some DL samples, copper, lead, and cadmium concentrations are also above background values (> 30 ppm Pb and > 0.5 ppm Cd). The amount of mercury in the pre-lake soils of CGL and DL varies considerably. In CGL-2 and CGL-3 soils, mercury values appear to follow the general trend in those cores; but the pre-lake soils of CGL-1, DL-3, and DL-1 contain significantly less mercury than the associated lake sediments. Concentrations in the DL pre-lake soils are well below the Willamette Basin breakpoint value for mercury, although the CGL-1 soil still appears enriched.

The explanation for this phenomenon (and why DL-1 and DL-2 have no pre-lake soils at all) lies in the activities of USACE. During construction of the two dams, large, shallow, borrow pits were dug in the valleys just upstream of the dam sites to supply earthfill (primarily gravel and clay). In DL, a diversion channel was also dug to re-route the Row River during the final phase of dam construction (DL-4 was taken in this channel). As a result, some parts of the lake bottoms had their topsoil and most recent floodplain deposits stripped away, leaving only deeper subsoils (or, in the case of DL-1 and DL-2, ancient river gravels) that formed before the onset of mining and subsequent contamination of river sediment. The above-background mercury values in the CGL pre-lake soil appear to reflect the presence of a natural mercury deposit upstream; but, consistent with the study of Hygelund (2000), mercury levels in DL soils imply no such source in the DL watershed.

The high concentrations of elements such as cobalt, chromium, nickel, and iron in the pre-lake soil of DL-3 appear to indicate contamination by steel. This core is located adjacent to both an old railroad line and an old lumber mill site, so some scrap metal may have been dumped in the area and incorporated into the soil.

Comparison of the DL and CGL systems reveals important differences in the influx rates of mercury and other metals. Most striking is the order-of-magnitude difference in average mercury concentrations in sediment from the two reservoirs. The total mass of mercury in DL and CGL can be estimated by multiplying the average mercury concentration by the mass of sediment contained in each lake. From an early USACE estimate of sedimentation rates in CGL (US Army Corps of Engineers 1953), the lake probably contains ~1.5 million tons of sediment. With an average mercury concentration of 1.452 to 2.720 ppm, the total mass of mercury in CGL is ~2–4 tons. In DL, a sedimentation survey performed in 1999 after 50 years of operation found ~2.4 million tons of sediment (Ambers 2000), so with an average mercury concentration of 0.242 ppm, the lake contains approximately 0.6 tons of mercury. CGL thus contains three to seven times as much mercury as DL even though it is only a few years older and its watershed is less than half the size (Table 1).

The distribution of mercury through the sediment cores is also different in the two reservoirs. Mercury influx to CGL decreased initially and then stabilized over time (Fig. 4a). This pattern is consistent with fact that the major cinnabar mining phase at Black Butte ended soon after the reservoir was constructed (Brooks 1963). Cessation of mining and milling activities probably caused the input of mercury-rich sediment to local streams to decrease until a relatively steady state was attained. Given the cinnabar deposits and inefficient milling history at Black Butte (Hardin 1909) and the correlation between clay content and mercury in more recent lake sediment, mercury is likely present in CGL as both cinnabar and elemental/ionic mercury adsorbed onto clays and organic matter. The influx of other heavy metals to the lake does

not appear to be mining-related and, with the strong correlation to clay content, may be associated mainly with the clay fraction of the sediment.

In DL, the general pattern of mercury variation is one of relatively constant input over time (Fig. 4b); but the values are slightly oscillatory, and small increases in concentration are associated with the deposits of very large flood events. Like mercury in CGL, the concentrations of gold, copper, lead, and zinc in the DL cores generally show an initial decrease and leveling off (Fig. 6), but the trend is better defined in some elements and cores than in others. The use of mercury for gold mining in the Bohemia Mining District tapered off by the end of the 1930s, but commercial metal mining continued until World War II (Brooks and Ramp 1968), and the reservoir was not completed until the fall of 1949. The initial decrease in heavy metal content may thus be related to the end of large-scale mining activities. If this were the case, the timing of the trend in mercury would be offset from that of the other metals, and the major input of mercury-contaminated sediment may have occurred well before the dam was constructed to trap it. Examination of pre-lake floodplain deposits would be necessary to confirm this hypothesis.

The Bohemia Mining District, with its numerous old stamp mill sites and tailings piles, could also act as a diffuse source of mercury that has provided a relatively constant, low-level input to the river (and lake) system since contamination began. In any case, mobilization of mercury appears to be linked to the occurrence of large floods, and heavy metal distribution may be controlled somewhat by depositional processes on the lake bottom. Higher velocity events and environments tend to increase the heavy mineral (and therefore heavy metal) content of the sediment. In the case of some of the cores, the depositional environment has changed over time as lake sediment thickness has increased, which may explain the higher metal content at the top of DL-1 located near the low pool.

The form in which mercury is being transported to DL (probably Hg^0 , Hg^{2+} adsorbed onto clays and organic matter, and/or gold-mercury amalgam particles) cannot be determined definitively from the current data set, but some inferences can be made. The very weak correlation between clay content and mercury indicates that mercury is not being transported primarily in the clay fraction of the sediment. Modern-day, recreational placer miners in the DL watershed have reported panning elemental mercury and gold-mercury amalgam from stream sediments (K. Smith, US Forest Service, personal communication 1999), so both are present in the river system near the mining district. Liquid mercury has not been found to be highly mobile in riverine environments, however, because of its high density and low chemical reactivity under surface conditions (Lacerda and Salomons 1998). In DL-1 and DL-2, the moderate to strong correlation between gold and mercury implies that at least a portion of the mercury is being transported in amalgam particles. The strong correlation between mercury and the sum of cop-

per, lead, and zinc in all the cores also suggests that mercury travels with these heavy metals. Because much of the coarser and heavier mercury-rich sediment in transit between mine and lake is likely residing in temporary storage within stream channels and banks (Miller 1997; Hygelund 2000), it is primarily re-mobilized during periodic, high-flow events.

Implications

The relatively constant input of mercury to both CGL and DL in recent years implies that current rates of mercury influx to the lakes will probably continue for many years, perhaps many decades, before a significant decrease is observed. Even if no more mercury escaped from Black Butte or the Bohemia Mining District, the contaminated sediment already in the stream systems would take some time to be flushed out. Remediation of the mine sites may therefore be of limited use in controlling lake contamination problems in the short term, although there would certainly be long-term benefits. From the standpoint of lake management, the problem does not appear to be getting worse with time, which is good news. Provided that bioaccumulation rates parallel sediment-mercury input rates, fish contamination should remain at manageable, although admittedly not ideal, levels.

The relationship between the concentration of mercury in sediment and fish from these lakes has important implications about the bioavailability of mercury in the two systems. Compared with DL, mercury concentration at CGL is roughly ten times higher in sediment but only three times higher in fish (Park and Curtis 1997). CGL fish contain only slightly less mercury than is present in the upper layers of CGL sediment; but in DL, fish actually contain more mercury, on average, than the lake sediment does. If significant amounts of mercury methylation were taking place upstream of DL and transported in with water, sediment, or biota, the unexpectedly high mercury content of its fish could be explained without regard to internal lake processes. Alternatively, the form(s) of mercury present in lake sediment can affect methylation rates because forms other than Hg^{2+} must undergo one or more reactions before methylation can take place (Lacerda and Salomons 1998). Perhaps the proportion of mercury bound as cinnabar in CGL results in slower production of methylmercury than occurs with the elemental and/or ionic forms found in DL. A study of the speciation of mercury in the two lakes would be needed to resolve this issue.

In both lakes, methylation may be enhanced by the annual drawdown and refilling cycle (Fig. 2). These seasonal changes in lake level result in a transition from oxidizing to reducing conditions in the pore fluids of lake sediments, which could significantly enhance methylation rates by creating conditions that are favorable for methylating bacteria (Regnell 1994; Rudd 1995; Croston and others 1996). When full-pool elevation is attained in late May, rapid growth of macrophytes and phytoplankton occurs in broad, shallow, upstream areas of the lakes,

creating an environment that may also be amenable to methylation and rapid bioaccumulation of mercury. Clearly, further study of these dynamics is required to determine where and how methylation is taking place and what role reservoir management practices may play in influencing the fish contamination problem. Since input of mercury to CGL and DL is not likely to decrease any time soon, the only cost-effective way to decrease levels of fish contamination might be to change the way the reservoirs are managed. Given the relatively low levels of contamination, the lack of evidence for any negative impacts on public health, and the desire for both flood control and recreational opportunities, however, management changes may not be considered worthwhile if only small improvements in fish contamination were likely to be achieved.

Conclusions

Dorena and Cottage Grove Lakes are impacted by very different kinds of mining activity. Cinnabar mining and milling in the CGL watershed resulted in relatively high levels of contamination in lake sediment that have decreased and then stabilized over time, apparently in response to the cessation of activity at Black Butte just after the reservoir was completed. Some of the mercury appears to reside in the clay fraction of the sediment, as do other heavy metals; but some mercury is likely present as HgS particles, too. In the DL watershed, historic use of mercury for amalgamation with gold and silver in stamp mills has resulted in lower levels of contamination in lake sediment. The mercury input to the lake has been relatively constant over time, but minor increases are associated with the deposits of very large floods. Input of heavy metals, such as copper, lead, and zinc, generally decreased and leveled off with time, probably in relation to the end of commercial mining in the Bohemia District a few years before the reservoir was constructed. Mercury in DL sediment is associated with other heavy metals and may be partially contained in amalgam particles. In both lakes, mercury input now appears to be relatively constant, so no significant decreases are expected in the foreseeable future unless remediation of the mining areas is undertaken. Further study of mercury methylation processes is needed in the context of current reservoir management practices in order to determine how management affects methylation rates and what changes, if any, could be implemented to improve the fish contamination problems.

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