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Dating of mine waste in lacustrine sediments using cesium-137

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Abstract For over a century Medicine Lake in northern Idaho has received heavy-metal-laden tailings from the Coeur d'Alene mining district. Establishing the depositional chronology of the lake bottom sediments provides information on the source and rate of deposition of the tailings. Cesium-137, an isotope produced in the atmosphere by nuclear bomb tests, was virtually absent in the environment prior to 1951, but reached its apex in 1964. Our analysis of cesium-137 in the sediments of Medicine Lake revealed that 14 cm of fine-grained tailings were deposited in the lake from 1951 to 1964 and tailing deposition downstream was greatly reduced by the installation of tailings dams in the district in 1968. Cesium-137 analysis is accomplished by a fairly simple gamma-ray counting technique and should be a valuable tool for analyzing sedimentation in any lacustrine environment that was active during the 1950s and 1960s.

Key words Cesium — Mine waste — Mine tailings

Introduction

Medicine Lake, one of 11 lateral lakes of the Coeur d'Alene River valley in northern Idaho, contains heavy-metal-laden sediments. The lakes lie downstream of a major base-metal mining district that has been active for over a century (Fig. 1). From 1885 until 1968, mill tailings and smelter wastes from the Coeur d'Alene district were dumped directly into tributaries of the Coeur d'Alene River. In 1968, tailing impoundment ponds were constructed to prevent further contamination of the river valley. In 1983, a 54 sq km area in the district was designated an Environmental Protection Agency Superfund Site (Fig. 1), and today there is considerable interest in the environmental effects of transported tailings downstream.

During floods over the past century, Medicine Lake has received heavy-metal contamination in the form of mine tailings deposited as well-stratified heavy-metal-contaminated silts on the lake bottom. To characterize the heavy-metal-laden sediments of Medicine Lake for future remediation, the relationship of these sediments to current and past mining practices in the district needs to be determined.

For over 40 years, the lateral lakes and their upstream catchment areas have been exposed to a variable flux of cesium-137 nuclides in rainfall and dryfall as a result of the atmospheric testing of distant nuclear weapons. Cesium-137 fallout began in February 1951 with Operation Ranger at the Nevada Nuclear Test Site, and global fallout reached a maximum in the northern hemisphere in 1963 or 1964 (Pennington and others 1973; Krey and others 1990).

Cesium-137 is commonly used to study erosion, transportation, and deposition of sediments. The mobility of cesium-137 appears to be low in soils. Past soil sampling studies have shown that the vast majority of cesium-137 is found in the top 5 cm of stable soils (Eakins and others 1984; Lance and others 1986). Cesium-137 is believed to be retained in soils by ion exchange on clay minerals (Schulz and others 1960; Francis and Brinkley 1976) and hence should be at higher concentrations in water when erosion occurs during rising floods (Eakins and others 1984) and is a useful marker of the movement and redistribution of fine-grained materials. Studies of cesium-137 in reservoir sediments suggest that much of the deposited cesium-137 is allochthonous, having been eroded from the watershed and transported to the reservoirs (Krey and others 1990). These reservoir studies also demonstrate that freshwater bodies with sedimentation rates greater than 1 cm/yr can provide reasonably precise histories of radioactive fallout in watersheds.

We have found that cesium-137 provides excellent time-markers within the heavy-metal-contaminated sediments of Medicine Lake. Using cesium-137, we have been able to

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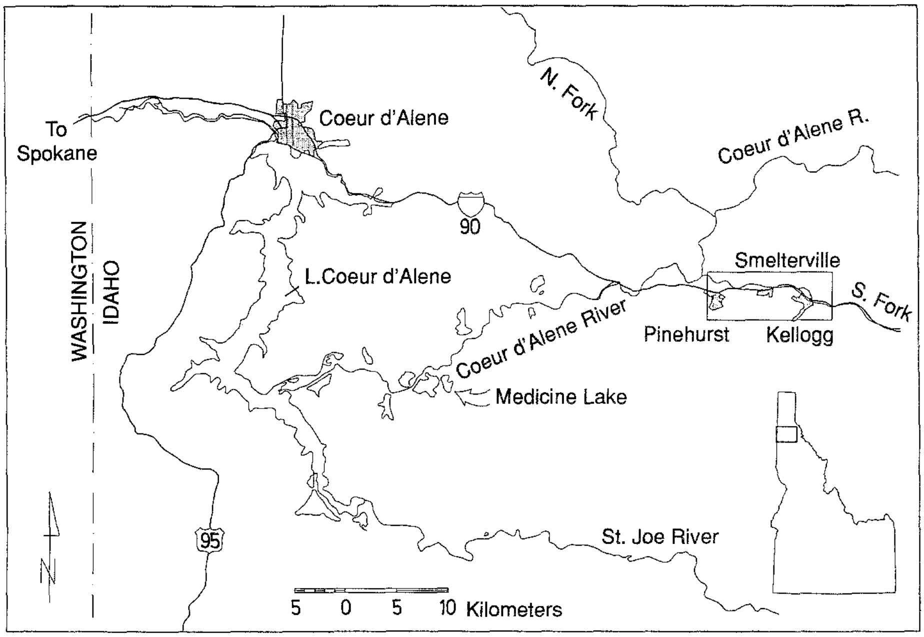

Fig. 1. Location map. Medicine Lake is one of 11 lateral lakes of the Coeur d'Alene River valley that lie upstream of Lake Coeur d'Alene and downstream of the Bunker Hill Superfund Site (boxed area)

calculate the rate of deposition of tailings and to evaluate the effectiveness of tailings dams that were installed 25 years ago in the mining district to mitigate downstream transportation of tailings. In this paper, we first discuss our sampling and analysis procedure and then interpret the cesium profile from Medicine Lake in terms of past mining practices.

Sampling

Our sampling in Medicine Lake was performed entirely through the use of a Huttenen freeze box. The freeze box was originally developed in Finland by Quaternary geologists to obtain undisturbed in-situ lake bottom sediments for varve studies (Huttenen and Merilainen 1978). The sampling device consists of a hollow stainless-steel box that is beveled at the bottom on the nonsampling side to ensure the sediment is not disturbed when the box is placed into position.

The sampling procedure was carried out in the winter when the lake surface was frozen. This made for a larger working area and greater stability at the surface than working from rafts. The depth of water was first measured; the box was then filled with Dry Ice and alcohol, and the cover secured. A steel pipe, of length greater than the depth of water, was attached to the top of the box, and ropes were

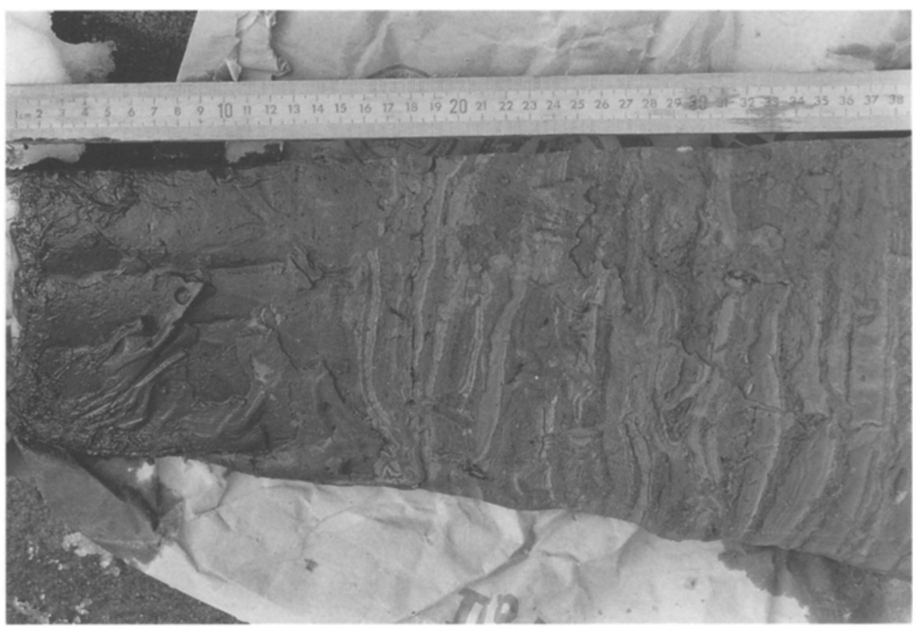

Fig. 2. Undisturbed lake bottom sample from Medicine Lake obtained with a freeze box sampler

secured to the box handles. The box was then lowered to the bottom of the lake and sunk into the lake bottom sediment. The entire apparatus was then immobilized at the ice surface with a pipe vise and left undisturbed for about 30 min. Using the ropes and the steel pipe, the sampler was withdrawn. Then the sample was removed from the sampler by emptying the alcohol and filling the box with lake water, thus allowing the sample surface to thaw slightly and so become detached from the box. The result was a stratigraphically undisturbed slab sample measuring about 2 cm thick × 55 cm long × 25 cm wide (Fig. 2). The slab was then placed in a labelled plastic bag, sealed, and stored in a cooler until it could be transferred to a freezer. For subsampling in the laboratory, the frozen slab was cut at 2-cm depth intervals. These subsamples were then dried in a low temperature oven for 24 h.

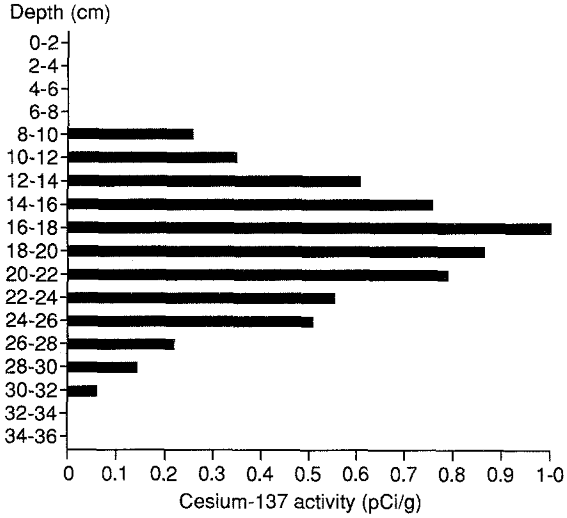

Fig. 3. Cesium-137 distribution in the Medicine Lake sample

Analysis

Cesium-137 was determined in the dried samples by gamma-ray spectroscopy using a lithium-drifted germanium gamma-ray spectrometer (Nuclear Data ND6700) at Washington State University's Nuclear Radiation Center. The dried samples, ranging in weight from 7 to 260 g, were placed in Marinelli beakers and counted on the above detector for 40,000 s. We then followed a procedure adapted from Cook (1990). The detector was calibrated using a point source of cesium-137 (at 662 keV) and a point source of bromine-82, which has gamma-ray energies that bracket the cesium-137 peak (619 keV and 698 keV). By doing this, the detector was calibrated to both point sources and the activity was measured for the bromine-82 solution. A Marinelli beaker with a bromine-82 solution of known activity and a volume approximating that of the samples was also counted. The ratio of the bromine-82 point source and the bromine-82 solution to that of the cesium-137 point source permitted the calculation of the detector efficiency of the cesium-137 in the Marinelli beaker configuration, thus resulting in a quantitative determination of cesium-137 in the samples.

Results

The Medicine Lake sample that we analyzed for cesium-137 is shown in Fig. 2. The upper 10 cm of the sample is fresh highly saturated organic material that constitutes the presentday lake bottom surface. Below 10 cm, the sample consists of dense sediments with varve-like features typical of heavy-metal-laden sediments in the lateral lakes. These sediments have concentrations up to several percent lead and zinc as well as anomalous amounts of copper, cadmium, arsenic, and silver. The dark laminates in the sediments represent autochthonous organic material from the lake environment; the light bands represent allochthonous fine-grained mine tailings deposited during floods.

The distribution of cesium-137 with depth for the sample is shown in Fig. 3. The cesium is confined to the depth interval between 8 cm and 32 cm in the sample. A distinct maximum is apparent at 16–18 cm.

The varve-like nature of the sediments shows that the deposition of the mine tailings in the lake occurred only during flood periods. However, based on the number of varve-like cycles present, floods were more or less an annual occurrence and no great periods of hiatus are suspected in the stratigraphy. The 14-cm-interval thickness between the maximum cesium-137 (1963–1964) and the onset of cesium-137 (1951) suggests an average sedimentation rate of 1.1 cm/yr during that time. Extrapolation to the top of the varve-like sediments (10–12 cm) yields an estimated date of 1969 for the uppermost tailings, a date consistent with the 1968 installation of the tailing ponds in the mining district. Thus, the tailings impoundments have drastically decreased contamination of Medicine Lake.

Conclusion

Using cesium-137 concentrations in the bottom sediments of Medicine Lake, we have found that mine tailings were deposited at the rate of 1.1 cm/yr prior to the installation of tailings dams in the mining district. Since the installation of the tailings dams, deposition of tailings in Medicine Lake appears to have been greatly reduced. Our findings suggest that cesium-137 can be very useful to characterize pollution problems in lakes, ponds, and reservoirs that involve continuous sedimentation through the 1951–1964 time interval.

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