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FATE OF MERCURY IN A RIVER-RESERVOIR SYSTEM

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APPLICATION OF ^{137}Cs AND ^{210}Pb IN TRACING THE FATE OF MERCURY IN A RIVER-RESERVOIR SYSTEM

Ralph R. Turner* and Curtis R. Olsen

ABSTRACT

The vertical distribution of ^{137}Cs and ^{210}Pb in sediment cores from the Tennessee River-Reservoir System (USA) was used to trace the fate of Hg discharged from two upstream facilities and to resolve the relative contribution from each facility. Discharges since 1943 at the Oak Ridge nuclear facilities left a clear record of releases for Hg and ^{137}Cs in undisturbed downstream sediments. High releases in the 1950s are reflected in well-defined peaks, located 30 cm or more below the sediment-water interface, which can now be used to accurately date sediment layers. Chronologies based on ^{210}Pb gave sediment ages concordant with those based on the release histories and helped to resolve mercury contributions from a chloralkali plant located 150 km downstream of the Oak Ridge facilities.

INTRODUCTION

During the 1950s, large quantities of Hg and ^{137}Cs were released to the Tennessee River-Reservoir System (USA) in association with both the production of weapons components and nuclear energy research at federal facilities in Oak Ridge, Tennessee. The history of releases of radionuclides is reasonably well documented, but quantitative information on releases of other contaminants is either absent or incomplete. Recent (1983) declassification of information concerning mercury discharged and unaccounted for from one of the facilities (ref 1) led to increasing scrutiny of downstream areas for residual mercury. The chief objectives of the work described in this paper were to use sediment cores to confirm independently the release history of mercury from the Oak Ridge facilities and to resolve contributions from a downstream chloralkali plant.

Metallic mercury was used in large quantities in Oak Ridge between 1955 and 1963 in the production of lithium enriched in ^6Li (ref 2). The release history (aquatic discharges) for Hg is shown in Fig 1a. The total aquatic release is estimated to be 110 metric tons (ref 2).

Reactor research programs began in the 1940s with the generation of fission products such as ^{137}Cs . As shown in Fig 1b, measurements of ^{137}Cs were begun in 1949 but earlier releases certainly occurred. The most striking feature of the release history is the near coincidence of the peak release year for ^{137}Cs (1956) with the peak release years for Hg (1957-58). The near coincidence in peak release years and the high affinity of both Hg and ^{137}Cs for particulate matter suggest that these contaminants should be highly covariant in downstream sediments.

Aquatic contaminants from the Oak Ridge facilities were discharged directly or indirectly into the Clinch River which joins the Tennessee River about 25 km downstream of the Oak Ridge Reservation. Dams at Tennessee River Miles (TRM) 530 and 471 form Watts Bar Lake and Chickamauga Lake, respectively. Watts Bar Dam was first closed for filling in January 1942, whereas filling of Chickamauga Dam was first begun in January 1940. Thus, undisturbed sediments in these reservoirs

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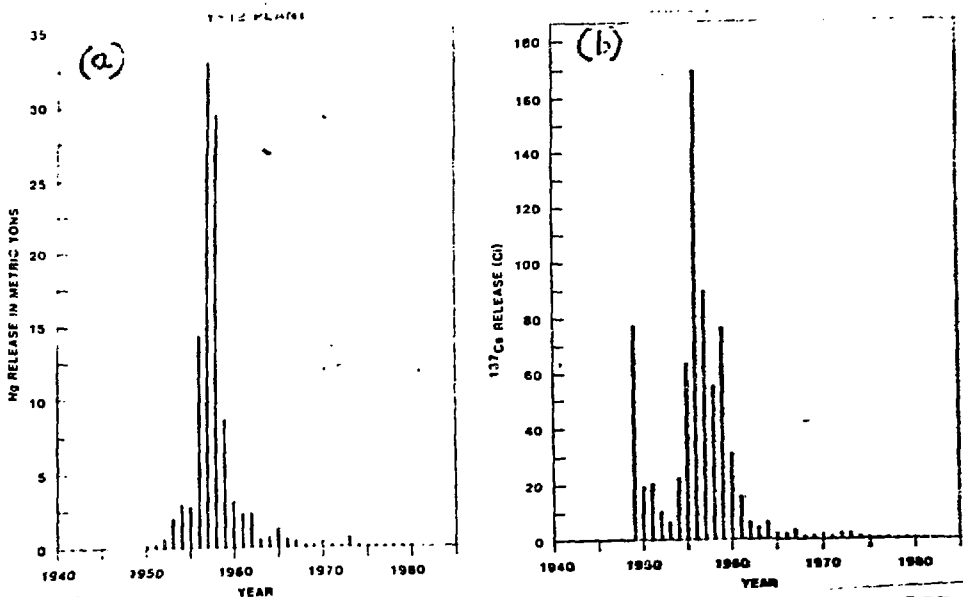


Fig 1 Release histories (aquatic discharges) of (a) Hg and (b) ¹³⁷Cs for Oak Ridge facilities.

were expected to contain a complete record of Hg and ¹³⁷Cs releases from Oak Ridge operations, which began in 1943.

MATERIALS AND METHODS

Sediment cores were obtained using a simple gravity corer with a 5-cm ID barrel. Cores were extruded and sectioned into 1-, 2-, 3-, or 5-cm intervals immediately after collection. On return to the laboratory, sections were dried for 16 h at 55°C and then analyzed for total Hg (ref 3) and for gamma emitters (ref 4). Analysis of ²¹⁰Pb in one sediment core (TRM 472) was performed by direct measurement of the 46.5-keV γ -ray. Self-absorption corrections were made as described by Cutshall et al. (ref 5).

RESULTS AND DISCUSSION

Large subsurface peaks in Hg and ¹³⁷Cs concentrations occurred in all cores. In those from the Clinch River and Watts Bar Lake (e.g., Fig 2a), Hg and ¹³⁷Cs peaks were coincident, yielding a correlation coefficient of 0.94 ($P < 0.001$) between Hg and ¹³⁷Cs. Several cores from Watts Bar Lake showed a secondary peak in ¹³⁷Cs activity, which was not associated with elevated Hg levels, below the primary peak. This lowermost peak in ¹³⁷Cs may be derived from the 1949 maximum in ¹³⁷Cs release (Fig 1).

The presence of a chloralkali plant constructed in 1963 on the Hiwassee River (tributary to Tennessee River below Watts Bar Lake) was expected to complicate interpretation of core data for lower Chickamauga Lake. Cores from this area exhibited Hg and ¹³⁷Cs profiles typified by Fig 2b. The peak in Hg concentration is not coincident with the peak activity in ¹³⁷Cs, and the Hg profile is not characterized by a single, well-defined peak. The sharp increase in Hg concentration beginning at 60 cm is coincident with the sharp increase in ¹³⁷Cs beginning at the same depth, suggesting that the first increase in Hg occurred ca. 1955-56 (peak release of ¹³⁷Cs). Using the average sedimentation rate for this core, based on deposition of 90 cm of sediment since dam closure (43 years), leads to a 1953 date for the first sharp increase in Hg concentration. These two lines of evidence suggest that the first sharp increase in Hg resulted from releases at Oak Ridge and not from the chloralkali plant, but additional proof was needed.

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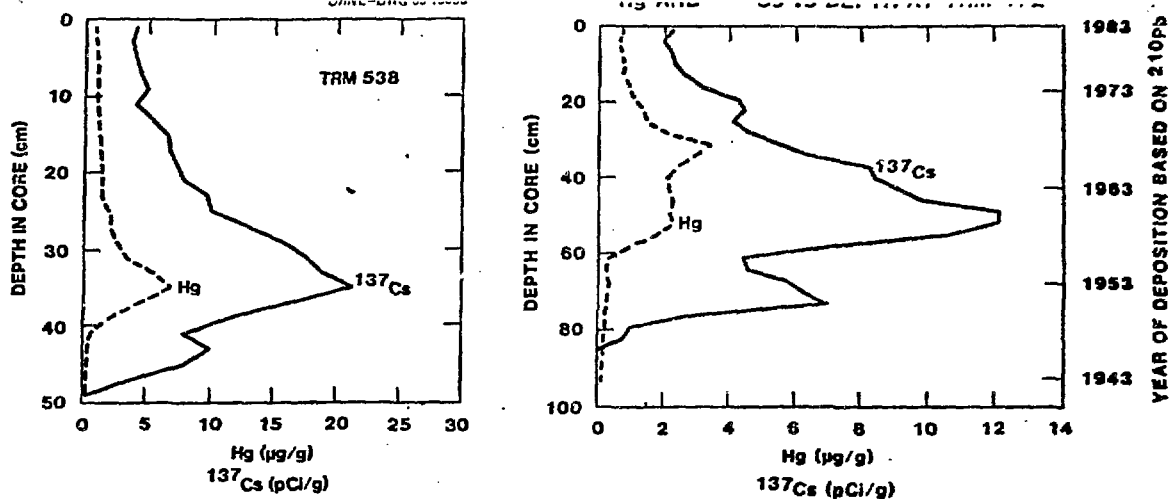


Fig 2 Mercury and ^{137}Cs versus depth in sediment cores from TRM 538 [Watts Bar Lake (a)] and TRM 472 [Lower Chickamauga Lake (2b)].

Application of the ^{210}Pb method of sediment dating provided additional proof as shown by the right-hand scale in Fig 2b. Sediment deposition dates were derived using the constant flux model as described by Robbins (ref 6).

The ^{210}Pb dates are consistent with the following conclusions: (1) The oldest sediment in the reservoir was deposited ca. 1943 and thus shortly after dam closure in 1940. (2) The lowermost peak in ^{137}Cs represents the 1949 high release (Fig 1). (3) The first increase in Hg occurred ca. 1955 and is due to the Oak Ridge facilities. (4) The second increase and major peak in ^{137}Cs occurred ca. 1956 and is due to the maximum release from Oak Ridge in 1956. (5) The peak Hg concentration at 30 cm is post-1963 and thus probably derived from the chloralkali plant. (6) A sharp decline in Hg begins in the mid- to late-1960s, perhaps in response to probable reductions in releases from the chloralkali plant.

Assuming the Hg profile at TRM 472 is composed of two intersecting and upwardly skewed peaks, it is possible to resolve approximately the relative contribution of Hg from the chloralkali plant and the Oak Ridge facilities. Based on peak areas, the lower peak (Oak Ridge) accounts for ~40% of the total area of the Hg profile. This apparent contribution from Oak Ridge can be cautiously extrapolated to the entire reservoir, but the result should be considered only a crude estimate. Overall, sediments in the two downstream reservoirs contain a substantial fraction of the mercury discharged from Oak Ridge facilities. Most of this mercury is now buried under 20 cm or more of sediment and thus not in direct contact with the biosphere.

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