# Fates of Metal Radiotracers Added to a Whole Lake: Sediment−Water Interactions

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# Fates of Metal Radiotracers Added to a Whole Lake: Sediment–Water Interactions

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HESSLEIN, R. H., W. S. BROECKER, AND D. W. SCHINDLER. 1980. Fates of metal radiotracers added to a whole lake: sediment–water interactions. Can. J. Fish. Aquat. Sci. 37: 378–386.

A whole-lake radiotracer experiment with the isotopes Se-75, Hg-203, Cs-134, Fe-59, Co-60 was carried out in Lake 224 of the Experimental Lakes Area, northwestern Ontario. The results of the analyses of isotopes in the water column show an exponential decrease in concentration with time. The rate of loss from the water column does not show dependence on the affinity of the isotope for suspended particulate material. This affinity ranged from 98% for Fe-59 to <1% for Cs-134 over the first 65 d. The major sink for isotopes lost from the water column is the sediments. Ratios of isotopes in the sediments show that those isotopes associated with suspended particulates more easily reach deep sediments than those in dissolved form which are restricted by the thermocline to adsorption to epilimnion sediments.

*Key words:* Experimental Lakes Area, metal and radioisotope budgets, environmental fate of radionuclides

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Une expérience avec radiotraceurs, couvrant un lac en entier, a été effectuée dans le lac 224 de la région des lacs expérimentaux, dans le nord-ouest de l'Ontario, utilisant les isotopes Se-75, Hg-203, Cs-134, Fe-59 et Co-60. Les analyses des isotopes dans la colonne d'eau indiquent une diminution exponentielle de concentration avec le temps. Le taux de perte depuis la colonne d'eau ne dépend pas de l'affinité de l'isotope pour la matière particulaire en suspension. Cette affinité varie de 98% pour Fe-59 à <1% pour Cs-134 pendant le premier 65 jours. Les sédiments sont le principal réceptacle des isotopes provenant de la colonne d'eau. Les rapports des isotopes dans les sédiments montrent que les isotopes associés aux particules en suspension atteignent plus facilement les sédiments de profondeur que les isotopes dissous qui, à cause de la thermocline, sont limités à l'adsorption aux sédiments de l'épilimnion.

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A program to study the fates in lakes of radioisotope tracers of heavy metals, interesting both as industrial and nuclear power effluents, has been underway since the summer of 1975. The principal objective of this study is to gain an understanding of the pathways and rates of movement of the radioisotopes in the whole-lake systems. This understanding in conjunction with work on the toxic effects of heavy metal and nuclear effluents should provide a means of estimating the potential effects of these effluents in lakes. Early work in enclosures in lakes has been presented by Kipphut (1978), Hesslein et al. (1978), and Schindler et al. (1980).

On July 29, 1976, the epilimnion of Lake 224 was spiked with six gamma emitting isotopes, Se-75 as sodium selenite, Hg-203 as mercuric chloride, Cs-134 as cesium chloride, Fe-59 as iron chloride, Zn-65 as zinc chloride, and Co-60 as cobalt chloride. The purpose of this experiment was to follow these isotopes through the chemical and biological pathways of the lake and to determine rate constants and accumulation factors for various biological and geochemical compartments. These compartments included water, settling particulates, sediments, zooplankton, periphyton, macrophytes, crayfish, lake trout, white sucker, slimy sculpin, and fathead minnows.

The purpose of this paper is to present the preliminary results of the water, sedimentation trap, and sediment analyses. The results of analyses for isotopes in the biological compartments will appear in a future paper. This separate treatment is possible because

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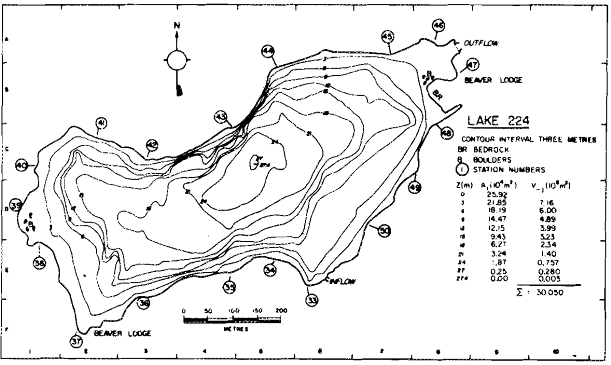

FIG. 1. Lake 224 in the Experimental Lakes Area, northwestern Ontario.

amounts of isotopes in biological compartments other than those sampled by filtering and coring (algae and periphyton) never exceeded 1% of the added amount when integrated over the whole lake.

## Methods

### STUDY SITE

Lake 224 (Fig. 1) has an area of 26 ha, a maximum depth of 27 m, and a mean depth of 12 m. Its drainage basin in Canadian Shield granite–diorite is overlain with shallow glacial sands and forested primarily by black spruce, jack pine, and birch. Lake sediments to a depth of ~10 m are mostly sandy, often overlain with a thin covering (<5 mm) of loose organic material. Clay underlies 10–20 cm of sand in some areas. There are many large boulders. At depths greater than 10 m the sediments are primarily organic ooze. The lake is oligotrophic with a secchi disk visibility of 7 m and a summer chlorophyll *a* concentration of 1.5 μg/L in the epilimnion, primarily as chrysophytes.

### SPIKE PROCEDURE

Two hundred to 500 mCi (1 Ci = 37 GBq) of each isotope as the salts previously listed were mixed into a metal barrel containing 10 L of water. The barrel, mounted on a raft, was towed around the lake by a motor boat while the solution dribbled into the lake from a tube emptying just below the surface. The rate of dribbling was slow, so this procedure required about 45 min to complete. The barrel was then refilled with water and the operation repeated. This method of addition allowed us to distribute the spike throughout the epilimnion of the lake, thus avoiding problems in interpretation which might result from a nonuniform distribution. Earlier mixing experiments (Quay 1977) with tritium as a tracer had shown that we could expect a horizontally uniform distribution within about 24 h of addition. This was borne out by measurements on the first day after the spike. All handling procedures were done directly or supervised by a health physics expert of Atomic Energy Canada Ltd. from the Whiteshell Nuclear Research Establishment. The total effect of the gamma emitting isotopes when diluted to the epilimnion of the lake was ~10% of the maximum permissible concentration (MPC) allowable in drinking water as established by the International Atomic Energy Agency.

### WATER SAMPLES

Water samples (2 L per depth) were taken in a 1-m-long Van Dorn bottle at the center of the lake and poured into linear polyethylene bottles. On returning to the laboratory the samples were filtered through a 0.45-μm membrane filter (cellulose acetate). These filters had been tested for adsorption of dissolved isotopes and this was found to be insignificant. Glass fiber filters were unsuitable as they adsorbed significant quantities of Cs-134. Because of this affinity of Cs-134 for porous glass, stainless steel filter supports were used rather than fritted glass. The membrane filters were prewashed by filtering 100 mL of distilled water to remove detergent surfactants (H. J. Simpson, Lamont Doherty Geol. Obs., personal communication).

The filtered water was then passed in sequence through columns (9 mm diam × 40 mm) of activated charcoal (Fisher Activated Coconut Charcoal 50–200 mesh) and mixed cation and anion exchange resins (Dowex 50w-x12 100–200 mesh, 1-x8 100–200 mesh). The activated charcoal is an effective adsorber of nonpolar species. Tests showed that a small percentage of some of the isotopes occasionally passed through these extractions. As a result the remaining sample was cooked at 90°C with 3.5 g/L of potassium bisulphate and 1 $cm^3$/L of concentrated sulphuric acid for 4 h. A combined ferracyanate–sulphide–hydroxide precipitation was then performed to scavenge any remaining isotope (P. Santchi, Lamont Doherty Geol. Obs., personal communication). The purpose of this procedure was to separate particulate and nonpolar and polar dissolved materials and to reduce the sample volume for increased counting efficiency.

### SEDIMENTATION TRAPS

Sedimentation traps (Fig. 2) were constructed from polyethylene bags of 22 cm diameter and 50 cm length. A floatation ring of 0.5 cm OD polyethylene tubing was attached to the open end and the closed bottom was attached by a length of rope to a weight. A loose noose of fishing line was attached at the middle of the bag and flag and cork floated the free end of the noose at the surface as a marker. The trap was retrieved by pulling on the noose, thus closing off the bag and pulling it to the surface. The water and settled particulates were filtered to remove excess water. Most traps were set with the openings just below the epilimnion at various distances from the shore.

### SEDIMENT CORES

Sediment cores were taken by two methods: divers took short (20 cm) cores in acrylic tubes (5 cm diam) in the firmer sandier sediments where sufficient light was available (0–10 m depth). Deepwater cores (10–25 m) were taken by a gravity corer (K–B corer) slowly lowered into the very soft profundal sediments. On returning to the lab, cores were sectioned into 0.5–2 cm slices depending on the character of the material. The material was then dried to 80°C and packaged for counting.

### ISOTOPE ANALYSES

The activities of gamma emitting isotopes in a sample were determined by gamma spectroscopy performed on a lithium drifted germanium (GeLi) crystal detector. The detector was supplied with samples by an automatic sample

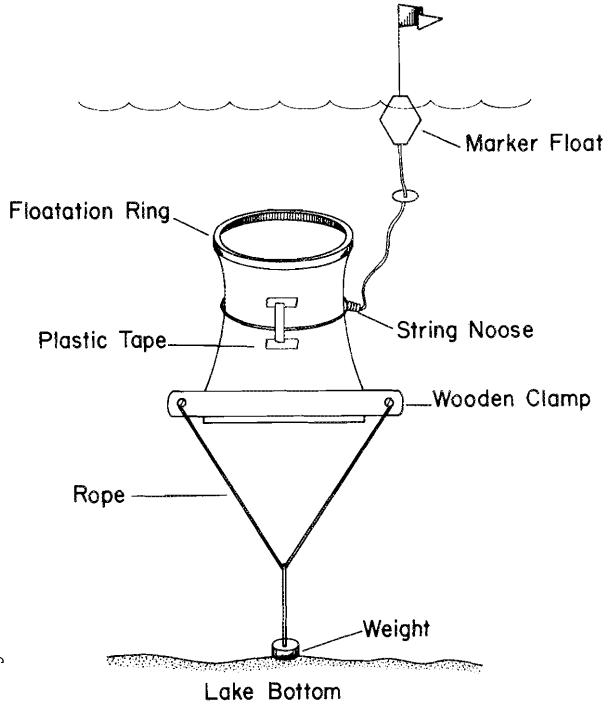

FIG. 2. Sedimentation trap of the type used to catch settling material in Lake 224.

changer. The detector was mated to a 4000 channel analyzer and spectra were processed by a programmable calculator. Spectral processing consisted of a peak find routine, integration of the peak, background subtraction based on a linear interpolation of counts bordering the peak, and estimation of the precision of the net integral based on peak and background counts. Correction for radioactive decay of the isotope back to the spike day was also performed in processing the spectra. Small petri dishes, 50 mm diam, 9 mm height were chosen as sample containers because of their nearly ideal counting geometry. All samples were counted in this format, simplifying geometry correction and allowing the use of the automatic sample changer. In all cases samples were spread uniformly in the petri dish. Corrections were made for the varying thicknesses of the samples, that of a filter to 9 mm. This correction was made by integrating an empirical curve established by measuring a filter standard at various distances from the detector. Self adsorption of gamma rays by the sample proved to be insignificant due to the low density and thinness of the samples. The overall counting efficiency of the system ranged from ~5% at 265 KeV to ~1% at 1333 KeV.

## Results

The concentrations of all gamma emitters in unfiltered epilimnion samples showed an exponential decrease with time over the first 65 d. Whole water sample data represent the sum of the fractions: particulate, charcoal extracted, ion exchange extracted, and precipitate. Shown with these data (Fig. 3) are lines represent-

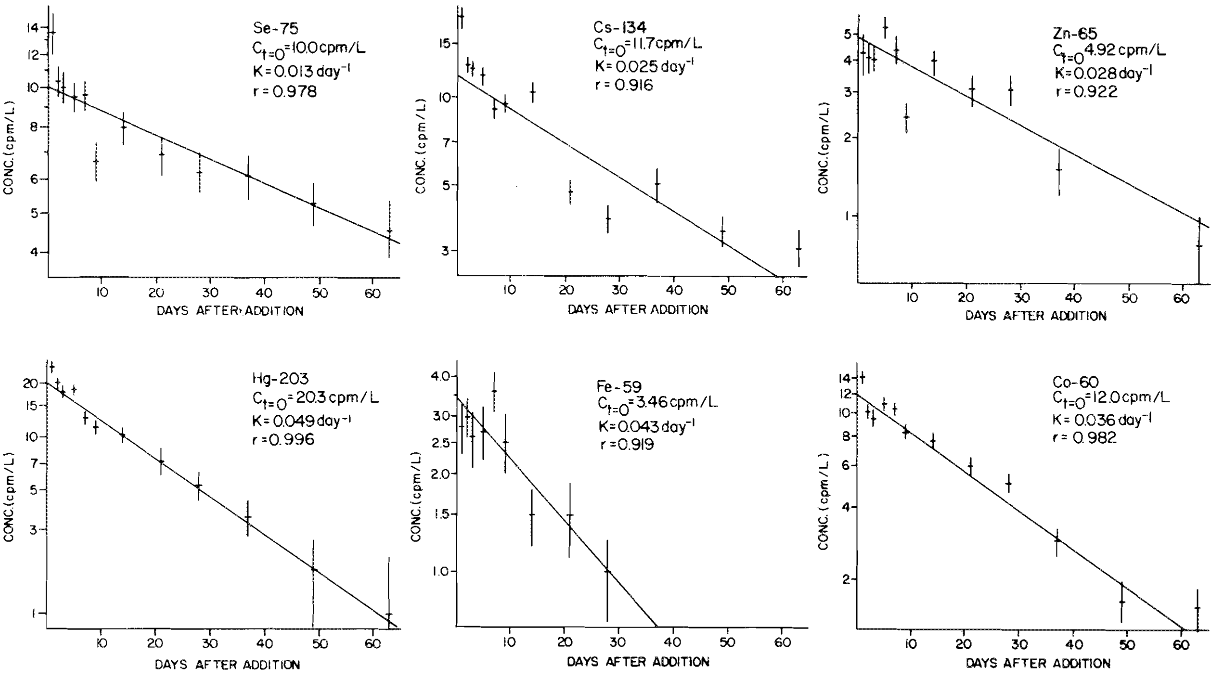

FIG. 3. Semi-logarithmic plots of the concentrations of six radioisotopes in unfiltered epilimnion water from Lake 224 over the first 63 d after addition, with linear regressions (see text for discussion of parameters).

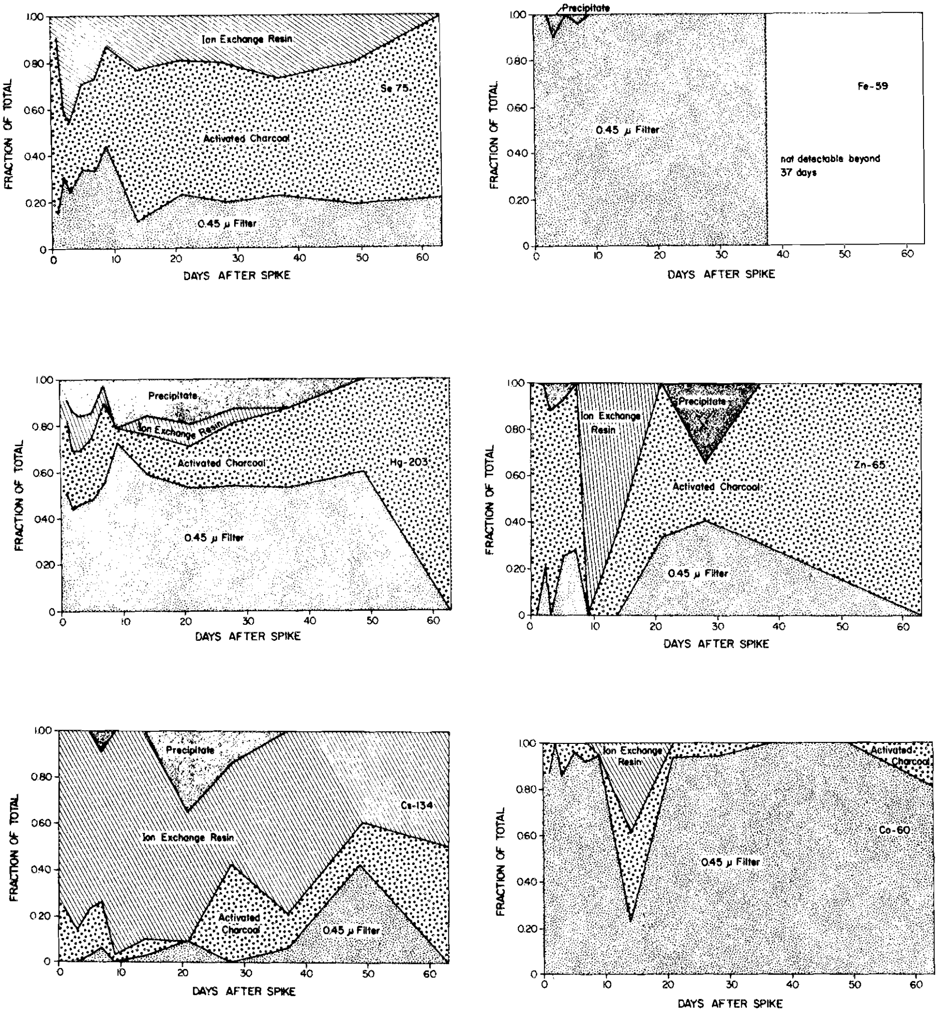

FIG. 4. The distributions of six radioisotopes among separated fractions of epilimnion water from Lake 224.

ing the linear regressions of the log of the concentration versus time. The lines represent an equation of the form: $C = C_{t=0}e^{-kt}$ where $C$ is concentration, $t$ is time, and $k$ is the rate constant for the change in concentration with time. Data from the first day after addition was not included in the linear regressions because samples indicated that some inhomogeneity remained in the epilimnion. This inhomogeneity was not present after the second day after addition.

The rate of loss of the isotopes from the epilimnion can be expressed as a half-time, that is, the time required for the concentration to decrease by a factor of two. This is just $\ln 2/K$ or $0.693/k$. The half-times for loss from the epilimnion are: Se-75, 52.0 d; Hg-203, 14.3 d; Cs-134, 28.1 d; Fe-59, 16.1 d; Zn-65, 24.7 d; and Co-60, 19.5 d.

The distribution of the isotopes between the fractions: particulate, charcoal extracted, ion exchange extracted, and precipitated (Fig. 4), remained fairly constant throughout the first 63 d. The standard deviation (with elimination of the single most aberrant value in some cases) for the fraction or particulates of all isotopes is less than 0.10 except Zn-65 which is 0.16. The loss of activity in certain fractions toward the end of the first 63-d period is probably due to low absolute concentration left in the water causing the concentration in the fraction to be below detection limits in the allotted counting time.

The areal concentrations of isotopes in cores are shown in Tables 1–4. These values are the integrated values over the length of the core (about 5–8 cm); however, in most cases the isotopes were confined to the top 1–3 cm. In many cases there is good agreement between cores at the same or similar depth, but often there are large differences, probably due to the heterogeneity of the surface sediments. Much of this spatial

TABLE 1. Activity of Cs-134 in Lake 224 cores in cpm/m$^2$ integrated over length of the core. (Dashed line represents the bottom of the epilimnion, initial activity in water was 59000 cpm/m$^2$.)

| | Days after spike (Date) | | | | | | | |
|---|---|---|---|---|---|---|---|---|
| Depth (m) | 14 (Aug. 3) | 24 (Aug. 13) | 31 (Aug. 20) | 37 (Aug. 26) | 63 (Sept. 21) | 147 (Dec. 14) | 254 (Mar. 31) | 331 (June 16) |
| 1 | 17900 | 33000 | | | | | | |
| 2 | | | | 35700 | 62900 | | | |
| 3 | 14700 | 27400 | | | | | 110000, 98400 | 238000, 148000 |
| 4 | | | 26400 | 56700 | 61700 | | | |
| 5 | 17200 | 21400 | | | | | 89800, 164000 | 117000, 94700 |
| 6 | ------ | ------ | 22800 | 37600 | 91600 | | 145000, 175000 | ------ |
| 7 | 1790 | 3000 | ------ | ------ | | | 166000 | 88700, 83400 |
| 8 | | | 4600 | 6060 | 24100 | | | |
| 10 | | | 1330 | | ------ | | | 54000, 45100 |
| 15–16 | | | | | | 43400 | 8450 | |
| 20–21 | | | | | 4760 | 17600 | | |
| 25 | | | 890 | | 2900 | 4331 | 25200 | |

TABLE 2. Activity of Co-60 in Lake 224 cores in cpm/m$^2$ integrated over length of the core. (Dashed line represents the bottom of the epilimnion, initial activity in water was 60200 cmp/m$^2$.)

| | Days after spike (Date) | | | | | | | |
|---|---|---|---|---|---|---|---|---|
| Depth (m) | 14 (Aug. 3) | 24 (Aug. 13) | 31 (Aug. 20) | 37 (Aug. 26) | 63 (Sept. 21) | 147 (Dec. 14) | 254 (Mar. 31) | 331 (June 16) |
| 1 | 7540 | 13100 | | | | | | |
| 2 | | | | 13700 | 25800 | | | |
| 3 | 8600 | 11500 | | | | | 54800, 46900 | 89900, 58500 |
| 4 | | | 13800 | 26400 | 24700 | | | |
| 5 | 5910 | 8990 | | | | | 45100, 90100 | 48400, 39600 |
| 6 | ------ | ------ | 9390 | 18400 | 44400 | | 81000, 99500 | ------ |
| 7 | 7000 | 11000 | ------ | ------ | | | 99900 | 40500, 39600 |
| 8 | | | 12700 | 31200 | 26600 | | | |
| 10 | | | 14300 | | ------ | | | 103000, 81700 |
| 15–16 | | | | | | 62200 | 10600 | |
| 20–21 | | | 760 | | 19400 | 28800 | | |
| 25 | | | 4210 | | 10600 | 13100 | 35400 | |

TABLE 3. Activity of Zn-65 in Lake 224 cores in cpm/m$^2$ integrated over length of the core. (Dashed line represents the bottom of the epilimnion, initial activity in the water was 25100 cpm/m$^2$.)

| | Days after spike (Date) | | | | | | | |
|---|---|---|---|---|---|---|---|---|
| Depth (m) | 14 (Aug. 3) | 24 (Aug. 13) | 31 (Aug. 20) | 37 (Aug. 26) | 63 (Sept. 21) | 147 (Dec. 14) | 254 (Mar. 31) | 331 (June 16) |
| 1 | 6060 | 12300 | | | | | | |
| 2 | | | | 10800 | 16400 | | | |
| 3 | 4040 | 8060 | | | | | 30400, 23400 | 59500, 32500 |
| 4 | | | 8460 | 16200 | 14200 | | | |
| 5 | 5360 | 6930 | | | | | 20500, 44000 | 31161, 24700 |
| 6 | ------ | ------ | 6810 | 10100 | 26800 | | 41500, 33500 | ------ |
| 7 | 2370 | 3190 | ------ | ------ | | | 42200 | 21500, 23100 |
| 8 | | | 2910 | 6140 | 7280 | | | |
| 10 | | | 2990 | | ------ | | | 28500, 21700 |
| 15–16 | | | | | | 16800 | 1980 | |
| 20–21 | | | — | | 3160 | 3730 | | |
| 25 | | | 770 | | 3030 | — | 9910 | |

TABLE 4. Activity of Se-75 in Lake 224 cores in cpm/m$^2$ integrated over length of the core. (Dashed line represents the bottom of the epilimnion, initial activity in the water was 50300 cpm/m$^2$.)

| | Days after spike (Date) | | | | | | | |
|---|---|---|---|---|---|---|---|---|
| Depth (m) | 14 (Aug. 3) | 24 (Aug. 13) | 31 (Aug. 20) | 37 (Aug. 26) | 63 (Sept. 21) | 147 (Dec. 14) | 254 (Mar. 31) | 331 (June 16) |
| 1 | 4660 | 6790 | | | | | | |
| 2 | | | | 8430 | 13300 | | | |
| 3 | 3290 | 4860 | | | | | 22300, 20600 | 44600, 27400 |
| 4 | | | 7060 | 19600 | 10200 | | | |
| 5 | 4040 | 4000 | | | | | 30000, 17330 | 28900, 20300 |
| 6 | ------ | ------ | 4290 | 6390 | 15700 | | 32000, 36400 | ------ |
| 7 | 3140 | 2190 | ------ | ------ | | | 36400 | 18800, 15200 |
| 8 | | | 1370 | 2570 | 2750 | | | |
| 10 | | | 1030 | | ------ | | | 25500, 10200 |
| 15–16 | | | | | | 3910 | 2480 | |
| 20–21 | | | — | | 2693 | 3330 | | |
| 25 | | | — | | 2350 | 2370 | 11000 | |

TABLE 5. Co-60/Cs-134 ratio in Lake 224 cores. (Dashed line represents the bottom of the epilimnion, Co-60/Cs-134 ratio in spike was 1.02.)

| | Days after spike (Date) | | | | | | | |
|---|---|---|---|---|---|---|---|---|
| Depth (m) | 14 (Aug. 3) | 24 (Aug. 13) | 31 (Aug. 20) | 37 (Aug. 26) | 63 (Sept. 21) | 147 (Dec. 14) | 254 (Mar. 31) | 331 (June 16) |
| 1 | 0.42 | 0.40 | | | | | | |
| 2 | | | | 0.38 | 0.41 | | | |
| 3 | 0.58 | 0.42 | | | | | 0.50, 0.48 | 0.38, 0.40 |
| 4 | | | 0.52 | 0.47 | 0.40 | | | |
| 5 | 0.34 | 0.42 | | | | | 0.50, 0.55 | 0.41, 0.42 |
| 6 | ------ | ------ | 0.41 | 0.49 | 0.48 | | 0.56, 0.57 | ------ |
| 7 | 3.9 | 3.7 | ------ | ------ | | | 0.60 | 0.46, 0.48 |
| 8 | | | 2.8 | 5.1 | 1.1 | | | |
| 10 | | | 10.7 | | ------ | | | 1.90, 1.81 |
| 15–16 | | | | | | 1.4 | 1.3 | |
| 20–21 | | | | | 4.1 | 1.6 | | |
| 25 | | | 4.8 | | 3.6 | 3.0 | 1.4 | |

variance can be removed if the data are presented as ratios of concentrations (Tables 5–7) rather than absolute amounts. Cs-134 was chosen as the denominator in these ratios as it showed almost no affinity for the particulate phase (mean of 0.03 on filters over 63 d). Only Se-75, Zn-65, Co-60, and Cs 134 were detectable in cores after 65 d, therefore only these data are presented.

Sediment trap data (Table 8) also show some highly variant values in terms of absolute concentrations. As in the case of the cores much of this variance is removed when the data are expressed as ratios. The higher variance in absolute values is likely due to variance in trapping efficiency and problems in retrieval of the traps.

## Discussion

The change in epilimnion whole water concentration of all isotopes with time shows a highly significant correlation when fitted by the first order equation $C = C_{t=0}e^{-kt}$. This suggests that the rates for removing the isotopes from solution are dependent on their concentrations. Two simple processes in the lake which would remove isotopes in this fashion are direct adsorption to sediments and settling of particulates. If these processes operated at different rates one would expect to see a difference in the removal rate of isotopes with high or low affinity for particulate material. No clear relationship is apparent between the rate constant for removal of an isotope and the average fraction of that isotope associated with particulate materials (Fig. 5), although those with a high fraction on particulate material seem to have slightly faster rates of removal. Either the rates of the two processes are similar or a more complicated process is involved.

The data in Tables 1 to 4 strongly support the theory

TABLE 6. Zn-65/Cs-134 ratio in Lake 224 cores. (Dashed line represents the bottom of the epilimnion, Zn-65/Cs-134 ratio in spike was 0.43.)

| | Days after spike (Date) | | | | | | | |
|---|---|---|---|---|---|---|---|---|
| Depth (m) | 14 (Aug. 3) | 24 (Aug. 13) | 31 (Aug. 31) | 37 (Aug. 26) | 63 (Sept. 21) | 147 (Dec. 14) | 254 (Mar. 31) | 331 (June 16) |
| 1 | 0.34 | 0.37 | | | | | | |
| 2 | | | | 0.30 | 0.26 | | | |
| 3 | 0.27 | 0.29 | | | | | 0.28, 0.24 | 0.25, 0.22 |
| 4 | | | 0.32 | 0.29 | 0.23 | | | |
| 5 | 0.31 | 0.32 | | | | | 0.23, 0.27 | 0.27, 0.26 |
| 6 | ------ | ------ | 0.30 | 0.27 | 0.29 | | 0.29, 0.19 | ------ |
| 7 | 1.3 | 1.1 | ------ | ------ | | | 0.25 | 0.24, 0.28 |
| 8 | | | 0.63 | 1.0 | 0.30 | | | |
| 10 | | | 2.3 | | ------ | | | 0.53, 0.48 |
| 15–16 | | | | | | 0.39 | 0.23 | |
| 20–21 | | | | | 0.66 | 0.21 | | |
| 25 | | | 0.90 | | 1.0 | | 0.39 | |

that the two processes of adsorption and settling are responsible for the removal of the isotopes from the water and show that a nonhomogeneous distribution of the isotopes is thereby produced in the sediments. Isotopes showing a high fraction associated with particulate, Fe-59 (0.98) and Co-60 (0.87) are not free to be readily removed by direct adsorption to sediments, but can easily settle with their associated particles. Cs-134 (0.03 on particulate) on the other hand has virtually no association with particles to allow settling, but has a large fraction in solution potentially available for adsorption. The ratios of Co-60/Cs-134 in the cores illustrate this separation due to different pathways (Table 5). Although both isotopes have similar rates of removal from the water (0.036 $d^{-1}$ and 0.025 $d^{-1}$), Cs-134 is 2.5 times more abundant than Co in the epilimnion sediments and Co-60 is 3–10 times more abundant in sediments below the epilimnion. The inability of Cs-134 to reach deep sediments is clearly shown in Table 1. Cs-134 accumulation in epilimnion sediments is 5–10 times more rapid than in sediments below the thermocline while Co-60 is accumulated at nearly equal rates above and just below the thermocline and at about half that rate in deepwater sediments. Zn-65 which is also removed from the water at a similar rate (0.028 $d^{-1}$), but has a lower association with particulate (0.21), shows a similar but less dramatic separation relative to Cs-134. Cs-134 is 1.2 times more abundant than Zn-65 in the epilimnion sediments relative to its concentrations in the water while Zn-65 is 1.6–6 times more abundant in deep water sediments. Se-75 shows the same general pattern but is more difficult to compare bcause of its much slower removal rate. Table 1 also shows clearly that, as the thermocline is eroded in the fall and Cs-134 remaining in the water comes into contact with deeper water sediments, the abundance of Cs-134 in those previously isolated sediments rises rapidly.

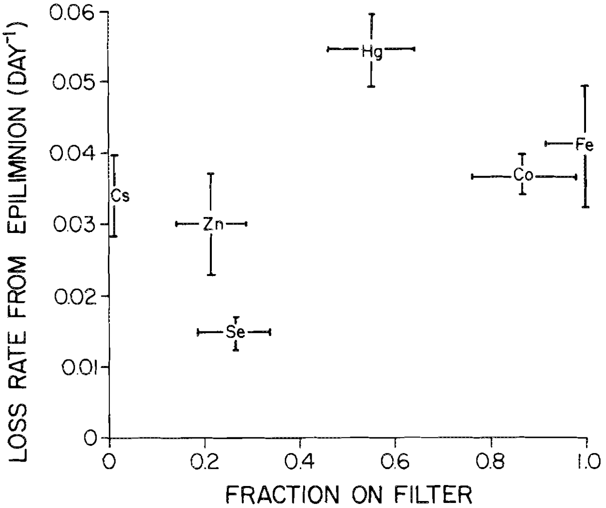

FIG. 5. Rate constant ($K$) for loss of radioisotope from the epilimnion of Lake 224 plotted against the mean fraction removed by filtration through a 0.45-$\mu$m filter over the first 37 d after addition. Error bars indicate 66% confidence interval of mean of fraction on filter for 12 samples.

In spite of the variability of activity accumulated in sediment traps (Table 8) the ratios of isotopes clearly show the greater abundance of isotopes with high affinities for particulate material such as Co-60 as compared to Cs-134. Ratios of isotopes in sediment traps hung at deeper depths are similar for ratios found in deepwater sediments (Tables 5–7). Ratios in traps hung at the bottom of the epilimnion show higher abundance of all isotopes relative to Cs-134 than epilimnion sediments but not as high as deepwater sediments and deeper sedimentation traps. Although there is no clear explanation for this discrepancy it is possible that Cs-134 in shallower traps had adsorbed to material collected from the water. This may also explain the increase in the relative abundance of Cs-134 in the sedimentation

TABLE 7. Se-75/Cs-134 ratio in Lake 224 cores. (Dashed line represents the bottom of the epilimnion, Se-75/Cs-134 ratio in spike was 0.85.)

| Depth (m) | Days after spike (Days) | | | | | | | |
|---|---|---|---|---|---|---|---|---|
| | 14 (Aug. 3) | 24 (Aug. 13) | 31 (Aug. 20) | 37 (Aug. 26) | 63 (Sept. 21) | 147 (Dec. 14) | 254 (Mar. 31) | 331 (June 16) |
| 1 | 0.26 | 0.21 | | | | | | |
| 2 | | | | 0.24 | 0.21 | | | |
| 3 | 0.22 | 0.18 | | | | | 0.20, 0.21 | 0.19, 0.19 |
| 4 | | | 0.27 | 0.35 | 0.17 | | | |
| 5 | 0.23 | 0.19 | | | | | 0.19, 0.18 | 0.25, 0.21 |
| 6 | ------ | ----- | 0.19 | 0.17 | 0.17 | | 0.22, 0.21 | ------ |
| 7 | | | ------ | ------ | | | 0.22 | 0.21, 0.18 |
| 8 | | | 0.30 | 0.42 | 0.11 | | | |
| 10 | | | 0.77 | | ------ | | | 0.47, 0.23 |
| 15–16 | | | | | | 0.09 | 0.29 | |
| 20–21 | | | | | 0.62 | 0.18 | | |
| 25 | | | | | 0.81 | 0.55 | 0.44 | |

traps with time throughout the experiment.

It is somewhat difficult to explain why Cs-134 should appear at all in the sedimentation traps, at least at deeper depths and in the deepwater sediments as it has almost no association with particulate material. It is also surprising that Cs-134, Co-60, and Zn-65 all reach the sediments at a water depth of 25 m in 31 d or less. This requires a settling velocity of at least 0.7 m/d. If one calculates the settling rate of Co-60 using its removal rate and fraction on particulates one arrives at a figure of about 0.18 m/d. One process which would accommodate both of these findings would be a significant transport of metals by fecal pellet settling. Fecal pellets would fall rapidly enough to move isotopes at a rate of 0.7 m/d or greater. In addition, because of their rapid sinking, the concentration of fecal pellets in the epilimnion would be very low thus evading detection on filters. Cs-134 would therefore appear in the sedimentation traps and deepwater sediments but not on the filters. The higher abundance of Co-60 in the epilimnion sediments relative to deepwater sediments could be due to a more or less even distribution of fecal pellet settling with an added increment of fine particles which settle more slowly to the epilimnion sediments. Fecal pellets are of major importance to sedimentation in many natural water bodies (Honjo and Roman 1978; Ferrante and Parker 1977; L. Small, U. of Michigan, personal communication) especially those of considerable depth.

TABLE 8. Activities of isotopes in sedimentation traps in cpm/m².

| Days after spike | Depth (m) | Se-75 | Hg-203 | Cs-134 | Fe-59 | Zn-65 | Co-60 |
|---|---|---|---|---|---|---|---|
| 0–14 | 7 | 227 | 836 | 410 | 454 | 300 | 737 |
| 0–14 | 7 | 1370 | 4840 | 2290 | 2090 | 1640 | 4380 |
| 0–14 | 7 | 944 | 4170 | 1950 | 1990 | 1680 | 4230 |
| 0–14 | 7 | 980 | 3360 | 1590 | 1110 | 1370 | 4810 |
| 14–28 | 5 | 282 | 1500 | 892 | 691 | 654 | 1530 |
| 14–28 | 5.5 | 256 | 1550 | 867 | 679 | 653 | 1670 |
| 14–28 | 5.5 | 476 | 2130 | 1490 | 1720 | 1190 | 3950 |
| 14–28 | 10 | 315 | 1250 | 333 | 1140 | 560 | 2200 |
| 14–28 | 12 | 59 | 334 | 84 | 231 | 103 | 460 |
| 28–42 | 8 | 448 | 1580 | 1610 | 1210 | 1160 | 3880 |
| 28–42 | 8 | 405 | 1560 | 2150 | 990 | 1200 | 3460 |
| 28–42 | 8 | 308 | 1250 | 1870 | 680 | 931 | 2060 |
| 28–42 | 8 | 541 | 2430 | 2000 | 738 | 1250 | 2320 |
| 28–42 | 10 | 368 | 1120 | 1080 | 849 | 925 | 2840 |
| 42–63 | 9 | 695 | 1670 | 3630 | 584 | 1520 | 3230 |
| 42–63 | 9 | 409 | 1220 | 2390 | 402 | 969 | 2400 |
| 42–63 | 10 | 313 | 1110 | 1300 | 602 | 933 | 2590 |

There is no evidence of major redistribution of isotopes through resuspension and settling as has been suggested by others (Davis 1968; Emerson and Hesslein 1973). The concentrations of Se-75, Zn-65, and Cs-134 in deepwater sediments remain low relative to epilimnion sediments through fall overturn (about day 80–110) and winter ice cover. If more than 10% of the epilimnion sediments with adsorbed isotopes had been resuspended and settled uniformly the deepwater sediment concentrations would have risen significantly. There is also no evidence for major "focusing" through sedimentation. Concentrations at 25-m depth are not consistently higher than those at 15- and 20-m depth.

Mass balance calculations 350 d after addition of the isotopes show 54% Se-75, 22% Cs-134, 19% Zn-65, and 23% Co-60 of the added amounts remaining in the water column. Total sediment burden based on sums weighted by area gives 50% Se-75, 112% Cs-134, 82% Zn-65, and 90% Co-60 of the added amounts. This yields sums of 104, 134, 103, and 113% of added amounts present 350 d after addition, which considering errors in integration of concentrations in sediments, is an acceptable balance. Because of the dryness of the 1976–77 season and the small fraction left in the water, outflow losses were not significant.

## Conclusion

The removal rates of the added isotopes Se-75, Hg-203, Cs-134, Fe-59, Zn-65, and Co-60 from the water column of Lake 224 were quite rapid and fairly similar ranging from 1.3 to 4.9%/d. Both settling of particulate material and direct adsorption to sediments are important mechanisms for this removal. The relative importance of the two pathways depends on the isotopes' affinity for suspended particulate material. Direct adsorption of isotopes to sediments limited the deposition of Cs-134, with no affinity for suspended particulates, primarily to epilimnion sediments; however, we believe that rapidly settling particles, perhaps fecal pellets, transported some Cs-134 to deep water sediments.

The distribution of Co-60 with a high affinity for suspended particulate material showed much less preferential deposition in the epilimnion. This distribution is probably a result of settling of both high and low velocity particles. Other isotopes were intermediate in distribution.

A relatively high percentage of isotopes (50% Se-75, and ~20% Cs-134, Zn-65, and Co-60) remained in the water column one year after addition.

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