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ABSTRACT

Four factors influence the concentrations of radionuclides in Dardanelle Lake water: injections due to fallout and discharge from Nuclear I coupled with losses due to decay, to dilution and to sedimentation. It is possible to estimate the first three factors and to measure monthly changes in the concentrations of ^{89}Sr, ^{141}Ce, ^{137}Cs, ^{58}Co, ^{144}Ce, and ^{90}Sr - ^{90}Y during periods when the concentrations of these nuclides are abnormally high (after large releases or the Chinese weapons tests) or abnormally low (during reactor refueling).

INTRODUCTION

The fate of radionuclides released by a nuclear power plant is an important factor in determining the water quality in the area affected by reactor operation. It is important that the radionuclides produced by the reactor be removed from the area of release by methods other than sedimentation, that is, by dilution or decay.

Radionuclides that are co-precipitated with sediments either can remain harmlessly adsorbed by the sediment or take one of two pathways which are deleterious to the environment:

1) since the radionuclides are concentrated in the sediment, it is likely that some quantity of radioactive material will enter the food chain through microorganisms indigenous to bottom sediment; or
2) the co-precipitated radionuclides may also be released back into the water and cause a contamination problem long after reactor operation has ceased.

The rate at which radionuclides co-precipitate with sediment can only be inferred, in rough measure, from the analysis of bottom sediment samples taken semiannually by the Technical Analysis staff at Arkansas Power and Light Company. The radionuclide concentrations are probably the average concentrations of several inches of sediment and thus are only crude measures of the quantity of radionuclides deposited over a short period of time, such as a month.

A closer approximation might be possible if an activity balance can be done on water samples taken at monthly intervals. Three mechanisms for the removal of radionuclides from Dardanelle Lake water will be considered: sedimentation, decay of the radionuclide, and dilution by "uncontaminated" water from upstream as water from the Arkansas River and the Illinois Bayou moves through the reservoir. The latter two are much more preferable from the ecological point of view.

For any radionuclide being removed from lake water

$$\text{Total Decrease} = \text{Decrease due to decay (A)} + \text{Decrease due to dilution (B)} + \text{Decrease due sedimentation (C)} - \text{Increase due to injection (D)}$$

Three occurrences during the period from June, 1976 to March, 1977 offered opportunities to estimate factors (A) and (B) in the above equation. Factor (D) was estimated from fallout data supplied by the radiochemistry group at the University of Arkansas-Fayetteville and from data supplied by AP&L. These occurrences were

1. the release into Dardanelle Lake of relatively large amounts of ^{137}Cs and ^{58}Co on June 21, 1976,
2. the Chinese nuclear tests of the autumn of 1976, which resulted in the injection of measurable amounts of ^{89}Sr and ^{141}Ce, short lived nuclides not usually found in Lake Dardanelle water, and
3. the shutdown of Nuclear I for refueling for the period from January 27, 1977 to March 26, 1977.

By measuring the decrease in the concentrations of the affected nuclides immediately following each of these occasions, it was possible to get an estimate of the amount of each radionuclide that was removed by the process of sedimentation.

MATERIALS AND METHODS

The concentrations of ^{89}Sr, ^{141}Ce, ^{137}Cs, ^{58}Co, ^{144}Ce, and ^{90}Sr - ^{90}Y were measured monthly from June, 1976 to August, 1977. The results of these measurements can be found in Chittenden (1978), and a summary found in Chittenden and McFadden (1979).

RESULTS

A. ^{89}Sr and ^{141}Ce: 10/29/76 - 2/18/77

The concentrations of ^{89}Sr and ^{141}Ce from the Chinese nuclear weapons tests, introduced into the water mainly by rainfall, are the simplest to treat. Although these nuclides are not found in reactor effluent, they provide a model to estimate the amount of long lived ^{89}Sr and ^{141}Ce that co-precipitate with sediment. Since the introduction of these nuclides into watercourses occurs over a wide area, we can assume them to be in the same concentration no matter what the source of the water. Thus, Factor (B) in the above equations = 0. The arithmetic becomes quite simple, and only concentrations need be considered. Table 1 summarizes the remaining factors for samples taken from October 29, 1976, to February 18, 1977.

Table 1. Fate of ^{89}Sr and ^{141}Ce Injected by Fallout.

Nuclide	Time Period	Station Number	Measured Concentrations of Radionuclides (pCi/l) Initial	Final	Decrease	Concentration Decay (A) (Calc.)	Concentration of Injection (D)	Concentration Residual* Sedimentation* (C)	% Decrease due to (C)
^{89}Sr	10/29/76-12/4/76	#1	0.49	0.31	0.18	0.18	0.06	0.05	11
		#2	0.45	0.34	0.11	0.17	0.06	0.00	0
		#4	0.40	0.29	0.11	0.16	0.06	0.01	4
	12/4/76-1/23/77	#1	0.31	0.20	0.11	0.15	0.04	0.00	0
		#2	0.36	0.21	0.15	0.16	0.04	0.01	5
	1/23/77-2/18/77	#1	0.20	0.075	0.13	0.06	0.01	0.08	17
		#2	0.21	0.04	0.17	0.06	0.01	0.12	67
	10/29/76-1/23/77	#6	0.58	0.12	0.46	0.42	0.30	0.35	17
^{141}Ce	10/29/76-2/18/77	#1	0.10	0.021	0.08	0.09	0.01	0.00	0
		#2	0.088	0.022	0.066	0.09	0.01	-0.01	0
		#4	0.13	0.088	0.04	0.11	0.01	-0.04	0

*Errors not specified in this and following tables are estimated to be ±30%.

B. ^{90}Sr — ^{90}Y and ^{137}Cs: 1/23/77 - 3/27/77

A similar trend is exhibited by the ^{90}Sr — ^{90}Y and ^{137}Cs concentrations after the reactor was shut down for refueling on January 27, 1977. For these nuclides, we will assume that Factor (A) = 0, since ^{90}Sr has a half-life of 28 years, and ^{137}Cs a half-life of 30 years. For ^{90}Sr, Factor (D) = 0 since there were no significant releases of this nuclide during this period.

From an analysis of the data in Chittenden (1978), it may be assumed that the $^{90}Sr - ^{90}Y$ from sources other than reactor operation (i.e. fallout) had a concentration of 0.45 ± 0.15 pCi/1 which will be referred to as "base-line concentration." This is generally the range of concentration of $^{90}Sr - ^{90}Y$ after two months of shutdown during which there were no significant releases of this pair.

After shutdown, water containing $^{90}Sr - ^{90}Y$ from both fallout and reactor effluent was diluted by water containing only fallout. Thus, in the time between two successive monthly collections, the concentrations of this nuclide should decrease, approaching the base-line concentration. ^{137}Cs was being continually injected into Dardanelle Lake. Assuming a near equilibrium mixing of water in the lake with the water flowing into the lake from the west and from Illinois Bayou, a simplified expression for the concentration of the nuclides in the second of two subsequent monthly samples can be derived.

$$C = C_b + (C_o - C_b)\exp[(-V_{flow}/V_{lake}) - \lambda t] \qquad (1)$$

where C = concentration of the nuclide in the second of two monthly samples (pCi/1)
C_b = base-line concentration of the nuclide (see above)
C_o = concentration of the nuclide in the first of two monthly samples
V_{lake} = volume of water in Dardanelle Lake = 4.86×10^5 acre feet
V_{flow} = volume of water which flowed through Dardanelle Lake
λ = decay constant; = 0 for ^{137}Cs, ^{90}Sr
= 0.01 day^{-1}for ^{58}Co
t = time interval between the two samplings (days)

The values for V_{flow} for the months of July, 1976 to March, 1977 provided by the office of the Corps of Engineers at the hydroelectric power station are presented in Table 2. The volume used to calculate the dilution factor was a weighted average of the two months through which the period between collections ran.

The values for the amount of activity injected that appear in Table 3 and 4 were derived from data on planned releases supplied by Technical Analysis, Arkansas Power and Light Company. It is assumed that extensive mixing takes place rapidly. It is also assumed, with reasonable justification, that releases of ^{137}Cs and ^{58}Co were spread out over the whole month rather than completed in a day or less and that there was minimal variation of radionuclide concentration in the effluent from day to day. Thus the following model can be proposed for the fate of ^{137}Cs, ^{58}Co and ^{90}Sr present in Dardanelle Lake.

1) The nuclides released by the Nuclear I facility are quickly mixed with lake water, C_b = Injection (Ci)/V_{lake} for ^{137}Cs and ^{58}Co. Fallout contribution of ^{137}Cs appears to be insignificant compared to injection from Nuclear I.
2) A fraction of the activity present in lake water was adsorbed onto sediment shortly after injection until the concentration reached $C_b < C_b$.

C_b was substituted for C_b in the equation (1) and calculated for each time period and station. The activity precipitated along with the sediment, A_{sed}, can then be calculated in the following manner:

$$A_{sed} = (C_o - C)V_{lake} + \text{Injection} - A_l$$

where A_l is the amount of each nuclide leaving the lake,

$$A_l = C_b V_{flow} - (C_o - C_b)(V_{lake})[\{\exp(-V_{flow}/V_{lake}) - \lambda t\} - 1]$$

Table 3 summarizes the factors contributing to the decrease in the concentrations of $^{90}Sr - ^{90}Y$ and ^{137}Cs during the period of refueling.

To estimate the maximum error inherent in these assumptions, A_{sed} was calculated assuming all injected activity was released immediately after the initial collection. This extreme value of A_{sed} was within 38% of the values of A_{sed} that appear in the Tables 1, 3 and 4. In Tables 3 and 4, percent activity removed by sediment

$$= A_{sed}/(A_l + A_{sed}) \text{ if } A_{sed} > 0 \text{ or}$$

$$= -A_{sed}/A_l \text{ if } A_{sed} < 0.$$

C. ^{137}Cs and ^{58}Co: 7/23/76 - 10/29/76

The release of ^{137}Cs and ^{58}Co on June 21, 1976, gave rise to abnormally high concentrations of these nuclides for several months after the release. Table 4 summarizes the factors which cause the decrease in the ^{137}Cs and ^{58}Co concentrations for the period of high concentrations.

Table 2. Total Monthly Release of Water from Dardanelle Lake.

Month	Volume of Water Released (Acre Feet)
July, 1976	2,850,800
August, 1976	730,540
September, 1976	365,060
October, 1976	420,020
November, 1976	336,360
December, 1976	394,020
January, 1977	505,500
February, 1977	435,760
March, 1977	1,831,040

Table 3. Fate of $^{90}Sr - ^{90}Y$ and ^{137}CS after Shutdown for Refueling.

Nuclide	Time Period	Station Number	Measured Concentrations of Radionuclides (pCi/1) Initial	Final	Injection (I) (Ci)	C_b' (pCi/1)	Residual* Activity (Ci) Removed by Sediment	% Activity Removed by Sediment
$^{90}Sr - ^{90}Y$	1/23/77 - 2/18/77	#1	0.67	0.63	ND	0.60	-0.07±0.07	-29±28
		#2	0.95	0.64	ND	0.42	-0.08±0.08	-19±19
	2/18/77 - 3/26/77	#1	0.63	0.46	ND	0.46	-0.01±0.11	-1±18
		#2	0.64	0.31	ND	0.31	0.21±0.33	26±28
		#6	0.88	0.62	ND	0.62	-0.38±0.32	-24±21
^{137}Cs	2/18/77 - 3/27/77	#1	0.14	0.094	0.0093	0.082	-0.17	-79
		#2	0.099	0.078	0.0093	0.077	-0.16	-87

ND = not detectable

Table 4. Fate of ^{137}Cs and ^{58}Co Injected into Dardanelle Lake on June 21, 1976.

Nuclide	Time Period	Station Number	Measured Concentrations (pCi/1) Initial	Final	Injection (I) (Ci)	C_b' (pCi/1)	Residual* Activity (Ci) Removed by Sediment	% Activity Removed by Sediment
^{137}Cs	7/23/76 - 8/19/76	#1	1.20	0.67	0.21	0.64	-0.33	-57
		#2	0.26	0.20	0.21	0.20	0.04	12
		#4	0.33	0.19	0.21	0.17	-0.08	-18
	8/19/76 - 9/24/76	#1	0.67	0.29	0.31	0.11	0.57	34
		#4	0.19	0.078	0.30	0.029	0.64	83
	9/24/76 - 10/29/76	#1	0.28	0.18	0.12	0.12	0.07	33
^{58}Co	6/21/76 - 7/23/76	#1	4.30	0.13	0.38	0.13	-0.16	-5
		#4	1.35	0.08	0.38	0.08	0.03	1

DISCUSSION

For the most part, the process of sedimentation removes only a small fraction of the radionuclides present in the water of Dardanelle Lake. In many cases the value for the activity removed by sedimentation is negative, indicating that activity was de-adsorbed and reentered solution.

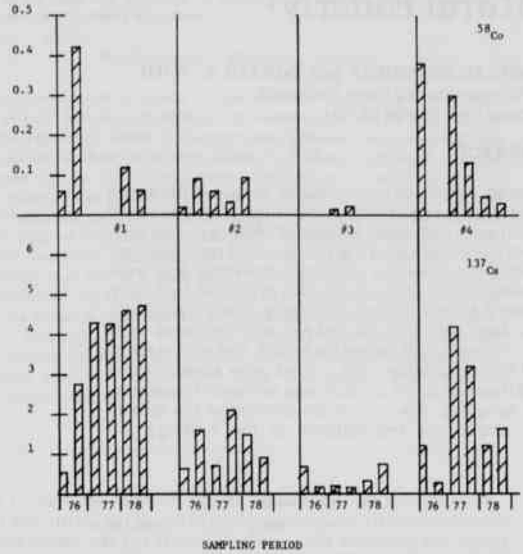

Figure 1. Concentrations of ^{137}Cs (Lower Histogram) and ^{58}Co (Upper Histogram) in sediment samples taken semi-annually in the Spring and Autumn, 1976-1978.

The two situations showing large percentages of removal by sedimentation (Table 1, 1/23/77 - 2/18/77; and Table 4, 8/19/76 - 9/24/76) are those in which the concentration of the activities of ^{90}Sr and ^{137}Cs, respectively, are not very large. For larger concentration, the percent decrease due to sedimentation becomes much smaller. This indicates that there may be a limited capacity (in Ci/g) of the sediment for co-precipitating these nuclides, particularly in the presence of significant amounts of aqueous Ca^{2+} and Na^{+}.

In many cases, the amount calculated for removal by sedimentation is a negative value. This would indicate activity is leaving the sediment. This is most likely to occur in the months following an unusually large injection (e.g. July, 1976 for ^{137}Cs and ^{58}Co) or when the activity of the nuclides in the water is abnormally low (e.g. ^{90}Sr — ^{90}Y and ^{137}Cs during shutdown, January - March, 1977).

Other than the exceptions noted above, the percentage of activity removed by sedimentation is usually less than 10% and never exceeds 26% for any of the radionuclides considered. We can thus conclude that sedimentation was not a major "sink" for radionuclides in Dardanelle Lake during the period of this study.

Data supplied by Dr. Dale Swindle of the Arkansas Power and Light Company Technical Analysis Laboratory on the concentrations of ^{137}Cs and ^{58}Co in sediment samples collected semiannually, summarized in Figure 1, confirms that there has been no significant accumulation of these nuclides in sediment except for ^{137}Cs at the mouth of the discharge canal (near the author's Station 1) where its concentration in the effluent water is at its greatest. The process of deposition at this point is probably not sedimentation but rather an exchange of ions between water and sediment.

Concentrations of these nuclides in sediment have generally been on the decline everywhere else during 1977 and 1978. This decline could be due either to a transfer of radionuclides back into the water or to the deposition of sediments with low specific activity.

It is not unreasonable to generalize these conclusions to include the rest of the radionuclides discussed herein. It is, thus, safe to conclude that a great percentage of the radionuclide load injected into Dardanelle Lake as a result of the operation of Arkansas Nuclear I is removed from the lake area in solution or suspension rather than being deposited with sediment.

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