Journal of the Arkansas Academy of Science

Volume 32 Article 11

1978

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Recommended Citation

Chittenden, David M. II and McFadden, Larry (1978) "Concentration of Radionuclides in Dardanelle Lake, Arkansas," *Journal of the Arkansas Academy of Science*: Vol. 32, Article 11.
Available at: https://scholarworks.uark.edu/jaas/vol32/iss1/11

The Concentration of Radionuclides in Dardanelle Lake, Arkansas

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ABSTRACT

Concentrations of the nuclides ^{90}Sr-^{90}Y, ^{144}Ce-^{144}Pr, ^{137}Cs, ^{58}Co, ^{110m}Ag, ^{141}Ce and ^{89}Sr have been measured monthly since November, 1975. The results from the period September, 1976, to August, 1977, depend on the relative intensities of the sources of the radionuclides; emissions from Nuclear I, the Chinese nuclear tests of Fall, 1976, and fallout from older atmospheric tests.

INTRODUCTION

Very low concentrations of some commonly produced radionuclides have been measured in the Dardanelle Lake area of the Arkansas River near the Arkansas Nuclear I Power Station operated by Arkansas Power and Light Company. The main thrust of this program was to determine the changes in the concentrations of the radionuclides, as a result of reactor operation, as a function of their distance from the source. The radionuclides whose concentrations were measured were ^{137}Cs, $^{144}Ce-^{144}Pr$, $^{90}Sr-^{90}Y$, ^{58}Co, and ^{110m}Ag. After the Chinese nuclear test explosions in the autumn of 1976, ^{89}Sr and ^{141}Ce were added to the list. A gross beta activity measurement was also made for each sample.

The radioactive effluent from Arkansas Nuclear I consists of a low level, continuous discharge and occasional planned releases of high activity waste that seemed to contain only ^{137}Cs and ^{58}Co in relatively large quantities along with small quantities of ^{110m}Ag, $^{90}Sr-^{90}Y$, ^{134}Cs and ^{60}Co. Only in these planned releases were the latter two nuclides observed.

Samples taken after a planned release provided some information about short-term and long-term mixing in Lake Dardanelle.

MATERIAL AND METHODS

To assure high sensitivity in the determination of these nuclides, radiochemical separations were performed on a sample of approximately 20 liters in volume. Counting was done with a low-background, anti-coincidence, gas-flow proportional counter. In the case of ^{58}Co and ^{137}Cs concentrations of greater than 0.5 pCi/1, low background NaI(T1) detectors and multichannel pulse height analysis were used for identification and confirmation.

Water Sampling

Samples varying in volume from 18 - 22 liters were taken from the surface at four points each month (see Figure 1) from November, 1975, to August, 1977, except during periods of inclement weather. It was found that, in the outlet bay, water discharged from the cooling tower remained in a surface layer approximately two feet in depth. For all sampling periods of 1976 and 1977, samples were taken monthly at Stations 1, 2 and 4 and bimonthly, alternately, at Stations 3 and 6. Sampling at Station 6 was discontinued after March, 1977. Alternate stations were used in November and December of 1975 (Stations 5 and 7 were used instead of 1, 21 instead of 3, and 16 instead of 2).

Chemical Separations

1. An aliquot of 2.00 liters of river water was taken to analyze for ^{110m}Ag. An aliquot of 250 - 500 ml was taken to prepare the gross beta sample.
2. The remaining sample was acidified and carriers of Cs^+, Co^{2+}, Sr^{2+} and Ce^{3+} were added. The sample was filtered through Whatman 42 paper after a settling period of two to seven days.
3. The sample was passed through a column of 100 g of Dowex-50 X8 at the rate of ~1 liter/hr.
4. The column was eluted with 500 ml of 6M HC1 and 200 ml of water. The eluate was then evaporated to dryness.
5. The Ce^{3+} was separated as $Ce(OH)_3$ and purified by solvent extraction. The purified Ce^{3+} was precipitated, filtered and counted as the oxalate after the method of Glendennin et al (1955).
6. The Co^{2+} was separated as CoS and purified by precipitation as $K_3Co(NO_2)_6$. The purified Co^{2+} was precipitated, filtered and counted as CoS after the method of Burgus (1961).
7. The Sr^{2+} was separated as $SrCO_3$ and purified by precipitation of $Sr(NO_3)_2$ from fuming HNO_3 after the method of Hodges as summarized by Beck (1975).
8. The Cs^+ was separated by coprecipitation with ammonium phosphomolybdate, purified by precipitation of $CsC10_4$ from absolute ethanol solution after the method of Kahn et al (1957). The Cs fraction was counted in this form.
9. Ag^+ carrier was added to the 2.00 1 aliquot. The Ag^+ was separated as AgC1. The precipitate was dissolved in conc. NH_3 and the $Ag(NH_3)_2^+$ solution was scavenged by $Fe(OH)_3$. The purified Ag^+ was precipitated, filtered and counted as AgC1.
10. The 250 ml aliquot was filtered and evaporated to dryness. The unfiltrable solids were slurried and quantitatively transferred to

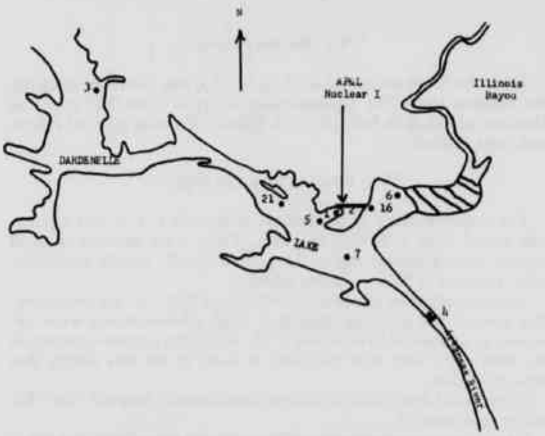

Figure 1. Sampling Stations, Dardanelle Lake Impoundment Area of the Arkansas River, Arkansas

aluminum planchets. After they were dried without baking, they were stored in a dessicator until they were counted after the method of Krieger (1975).

Counting Systems and Procedures

All the samples, except the gross beta samples, were wrapped in Mylar film with a thickness of 1-4 mg/cm^2. These samples were counted in either a Tracerlab Omni-Guard system with a 1" sample detector (G=0.209) or a Tennelec System equipped with a 2¼" Beckman anticoincidence detector (G=0.283).

The Cs and Co fractions from June and July, 1976, were also counted with a 3" x 3" NaI(Tl) detector housed in a 4" (minimum) lead shield.

Analysis of Data

All beta and X-ray (in the case of ^{58}Co only) counting data were corrected for systematic errors, i.e. the geometry of the detector, backscattering, absorption of radiation by sample cover and detector window, and self-absorption and self-scattering. Correction factors for the last two errors are from Nervik and Stevenson (1952).

Chronology of Significant Events

1. June 21, 1976 - planned release of stored cooling water into Dardanelle Lake as stations were being sampled.
2. September 26, October 17, and November 17, 1976 - Chinese test nuclear devices in the atmosphere.
3. January 27, 1977 - Nuclear I shut down for refueling.
4. March 26, 1977 - refueling completed. Start-up followed closely.

RESULTS AND DISCUSSION

^{144}Ce - ^{144}Pr - Fission Products

The concentration of this pair varied only slightly, from 0.00 to 0.05 pCi/1 for the period from 11/75 to 3/77, except for the two months following the Chinese nuclear tests.

Figure 2 summarizes the concentration data for this pair from 12/76 to 8/77. Immediately before and during refueling the concentration was quite low. After resumption of operation, the concentration rose to 0.05 - 0.15 pCi/1, higher than before refueling. There is little doubt that this increase was due entirely to reactor operation.

^{141}Ce - Fission Product

This short-lived radionuclide ($T_{1/2}$ = 33 d) was produced solely by the Chinese tests. The concentration dropped from 0.15 pCi/1 in October to less than 0.05 pCi/1 in March. A spring peak of fallout was quite evident.

^{58}Co - Neutron Reaction Product

The concentrations of this nuclide at Stations 1, 2, 3, and 4 generally varied from 0.00 to 0.10 pCi/1. There were isolated cases of higher concentrations (up to 0.8 pCi/1). Small, usually unmeasurable, amounts of ^{60}Co were also released.

Immediately after the planned release of 6/21/76, the concentration rose to 4.3 pCi/1 at Station 1. High concentrations were observed at all stations in 6/76 and 7/76, indicating extensive mixing in the lake and a very slow turnover of water in the lake during that drought period.

It was found from the 6/76 sample from Station 1 that the ^{58}Co/^{60}Co was greater than 10.

One of the most interesting and inexplicable sets of data was the concentration data for ^{58}Co at Station 6. In 12/75, the concentration was found to be an unusually high 0.88 pCi/1. The concentration in each sampling period thereafter was lower than in the preceding period. The concentration fell off monotonically with a half-life of 71 days, which is the half-life of this radionuclide.

The only explanation of this coincidence is not readily acceptable. One must conclude that the ^{58}Co contamination at this station remained undiluted by the waters of the Illinois Bayou and did not move with the current. The probability of this is quite small, but no alternative explanation presents itself.

^{110m}Ag - Neutron Reaction Product

The concentration of this nuclide was erratic but generally stayed between 0.00 and 0.40 pCi/1. In late 1975, though, levels up to 1.6 pCi/1 were observed. Since then, only on 6/21/76 has the level been abnormally high (1.2 pCi/1).

^{89}Sr - Fission Product

The trends in the concentration of this nuclide are the same as those seen in the ^{141}Ce data. ^{89}Sr is also a short-lived nuclide ($T_{1/2}$ = 53 d). Only at Station 2 in April, July and August, 1977 and at Station 1 in August were levels of ^{89}Sr anomalously high. At these times and locations the concentrations were higher than in the months immediately following the bomb tests.

^{90}Sr - ^{90}Y - Fission Products

In 1975 and early 1976, the concentration of this pair varied only slightly, from 0.6 - 0.8 pCi/1. Station 1 showed an unusually high concentration on 6/21/76 as expected.

After the Chinese tests, the levels at all stations rose to 1.1 - 1.2 pCi/1. The concentration dropped to very low levels (0.4 - 0.6 pCi/1) during refueling (see Figure 3).

After operation was recommenced, the concentration of the pair became very erratic, particularly at Stations 2 and 3 (the upstream stations) during the spring and summer, rising as high as 1.73 pCi/1. By August, concentrations at all stations had returned to the neighborhood of a more normal 0.9 pCi/1.

^{137}Cs - Fission Product

This nuclide was the most common in releases, planned or unplanned. Concentrations generally ran from 0.00 to 0.30 pCi/1.

During the release of 6/21/76, the concentration rose to 33 pCi/1 at Station 1. It is interesting to note that even before the release was finished, high levels of ^{137}Cs had already accumulated at every station. This effect was least noticeable at Station 2. High levels were observed at all stations in July and August, 1976. Assuming that there were no more releases in these two months, it can be assumed that there was a very slow turnover of the water in Lake Dardanelle during this period.

It was also found that the dilution of the released ^{137}Cs as it moved down river was not as great as expected. At the dam, Station 4, the concentration was still 2.4 pCi/1, only a 14-fold dilution. The same effect was noticed in the dilution of ^{58}Co. In the period before the next sampling was done, thorough mixing of the ^{137}Cs over the whole lake had taken place. In December, 1976, the concentration rose to 0.84 pCi/1. This may have been the remnant of a release in the preceding month.

Unusually high levels of ^{137}Cs were observed at Station 2 in April, July and August, 1977, somewhat paralleling the behavior of ^{90}Sr - ^{90}Y.

Gross Beta Activity

Gross beta activity consisted of three components; (1) fallout carried from upstream, (2) natural radioactivity (^{232}Th, ^{40}K, ^{235}U and

^{238}U) carried as unfiltrable solids and (3) release from Arkansas I. Many of the trends mentioned above are masked by components (1) and (2). A complete discussion of gross beta activity data may be found in Chittenden (1978).

Table 1 lists the maximum permissible concentrations of the various nuclides discussed in this work. It can be seen that the waterborne emissions from Nuclear I are very low level in all cases.

Only ^{90}Sr$-^{90}$Y stays near 1 pCi/1. Although the level of this nuclide is <1% of the allowable level, the increase during 1977 is cause for concern. If these unusually high levels had occurred at all stations, abnormally high fallout could have been assumed to be the cause. It is unusual, though, that the high levels should occur only upstream from the reactor. One must resort to rather unusual mechanisms to attempt an explanation of these anomalies. Upstream currents have been observed in Lake Dardanelle. These caused the good mixing observed with ^{137}Cs and ^{58}Co in 6/76. But high levels of ^{90}Sr $-$ ^{90}Y were not observed at Station 1 which should be the starting point for any liquid effluent. An alternative would be the release of volatile precursors into the atmosphere. Both ^{89}Sr and ^{90}Sr have krypton precursors of short half-life. Once they are released into the atmosphere, they and their daughters could drift up river carried by the wind where they eventually would precipitate out of the atmosphere, but not equally over all stations. Neither of these explanations is wholly satisfactory.

It is significant, though, that unusually high levels of a number of nuclides were found in Dardanelle Lake water immediately upon start up of Nuclear I after refueling. The correlation of the ^{90}Sr $-$ ^{90}Y data with that of ^{144}Ce $-$ ^{144}Pr, ^{89}Sr and ^{137}Cs definitely hints at a containment problem immediately after start up in March, 1977. However, the data of August, 1977, indicate that the problem may have been only temporary.

ACKNOWLEDGEMENTS

Partial financial support for this study was provided by the Office of Water Research and Technology through the Arkansas Water Resources Research Center, project number A-037-ARK.

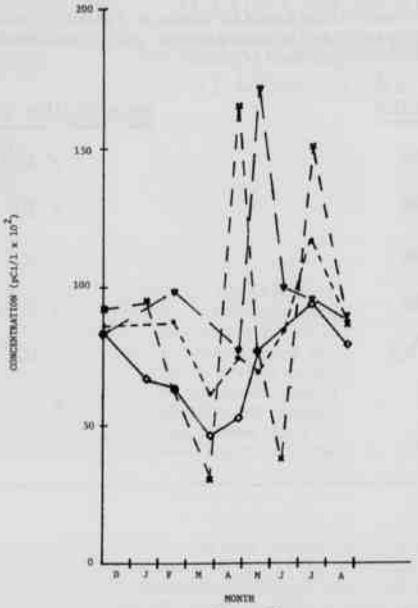

Figure 3. Concentration of ^{90}Sr $-$ ^{90}Y: 12/76 - 8/77

◇——◇ Station #1
∇- - -∇ Station #3
×- - -× Station #2
●- - - -● Station #4

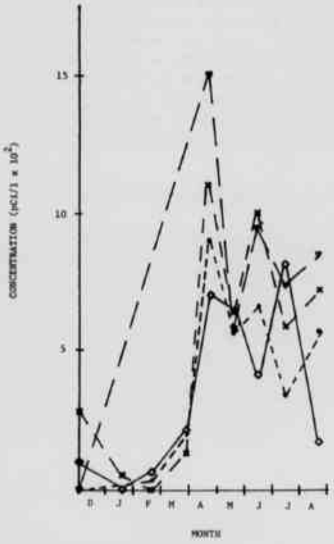

Figure 2. Concentration of ^{144}Ce $-$ ^{144}Pr: 12/76 - 8/77

◇——◇ Station #1
∇- - -∇ Station #3
×- —× Station #2
●- - - -● Station #4

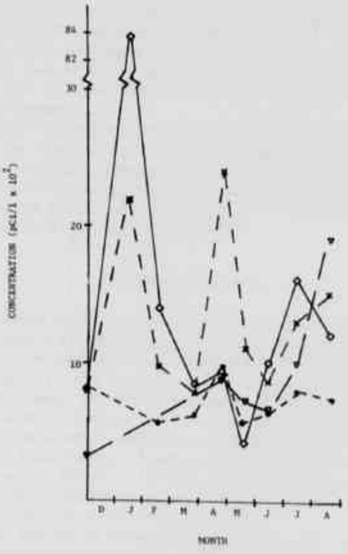

Figure 4. Concentration of ^{137}Cs: 12/76 - 8/77

◇——◇ Station #1
∇- - -∇ Station #3
×- —× Station #2
●- - - -● Station #4

Table 1. Concentration Limits for Release of Radioactive Material in Liquid Effluent to an Uncontrolled Area (pCi/1) established by the Arkansas State Department of Health (1974).

NUCLIDE	PERMISSIBLE LEVEL
^{144}Ce	1×10^4
^{141}Ce	9×10^4
^{137}Cs	2×10^4
^{58m}Co	3×10^6
^{110m}Ag	3×10^4
^{89}Sr	3×10^3
^{90}Sr	3×10^2

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