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Radionuclide Concentrations in the Arkansas River Upstream and Downstream from the Nuclear I Power Generating Facility

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RADIONUCLIDE CONCENTRATIONS IN THE ARKANSAS RIVER UPSTREAM AND DOWNSTREAM FROM THE NUCLEAR I POWER GENERATING FACILITY

Technical Completion Report
1978

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INTRODUCTION

This report summarizes results obtained from a program designed to measure very low levels of some commonly produced radionuclides in the Dardenelle 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 increase in the concentration of the radionuclides as a result of reactor operation as a function of their distance from the source. It was hoped to extend this study to include the effects of these emissions on the uptake of radionuclides into biological systems and their deposition in sediments.

Most studies of this kind are done by the isotopic gamma method of analyzing the gamma ray spectrum of a 0.5 - 3.5 liter sample of water using a high-efficiency Ge(Li) detector. In many cases, especially in the areas of recently constructed reactors, the concentration of most radionuclides is below the detection limits for this system.

To increase the sensitivity of the determination of these nuclides, it was decided to perform radiochemical separations 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/l, low background NaI(Tl) detectors and multichannel pulse height analysis were used for identification and confirmation.

The radionuclides whose concentrations were measured were ^{137}Cs, ^{144}Ce-^{144}Pr, ^{90}Sr-^{90}Y, ^{58}Co and ^{110m}Ag (see Table 1). 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.

Table 1: Nuclides Measured in Dardenelle Lake Water

Nuclide	Means of Production	Mode of Decay	Particle Energies (8)	Half Life (8)
^{137}Cs	Fission	β^-	1.176 Mev(6.5%) 0.514 Mev (93.5%)	30.0 yr.
^{144}Ce -	Fission	β^-	0.309 Mev (76%) 0.175 Mev (24%)	284 d
^{144}Pr		β^-	2.996 Mev (97.8%) 2.30 Mev (1.2%) 0.807 Mev (1.0%)	17.3 min
^{141}Ce	Fission	β^-	0.582 Mev (30%) 0.444 Mev (70%)	33 d
^{90}Sr -	Fission	β^-	0.546 Mev	28.1 yr
^{90}Y			2.268 Mev	64 hr
^{89}Sr	Fission	β^-	1.463 Mev	52 d
^{58}Co	^{58}Ni (n,p)	β^+(15%)	0.474 Mev	71.3 d
		EC (85%)	6.92, 6.93, 7.1 Kev x-rays	
^{110m}Ag	^{110}Cd (n,p)	β^-(98%)	0.530 Mev (34%) 0.085 Mev (64%)	253 d
		γ with IT (2%)		

RELEASES

The radioactive effluent from Arkansas Nuclear I consists of a low level, continuous discharge plus 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 provide some information about short-term and long-term mixing in Lake Dardenelle.

EXPERIMENTAL PROCEDURES

A. Water Sampling

Samples varying in volume from 18 - 22 liters were taken from the surface at four points each month (see Figure 1) 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 samples 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.(1)

B. 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+}

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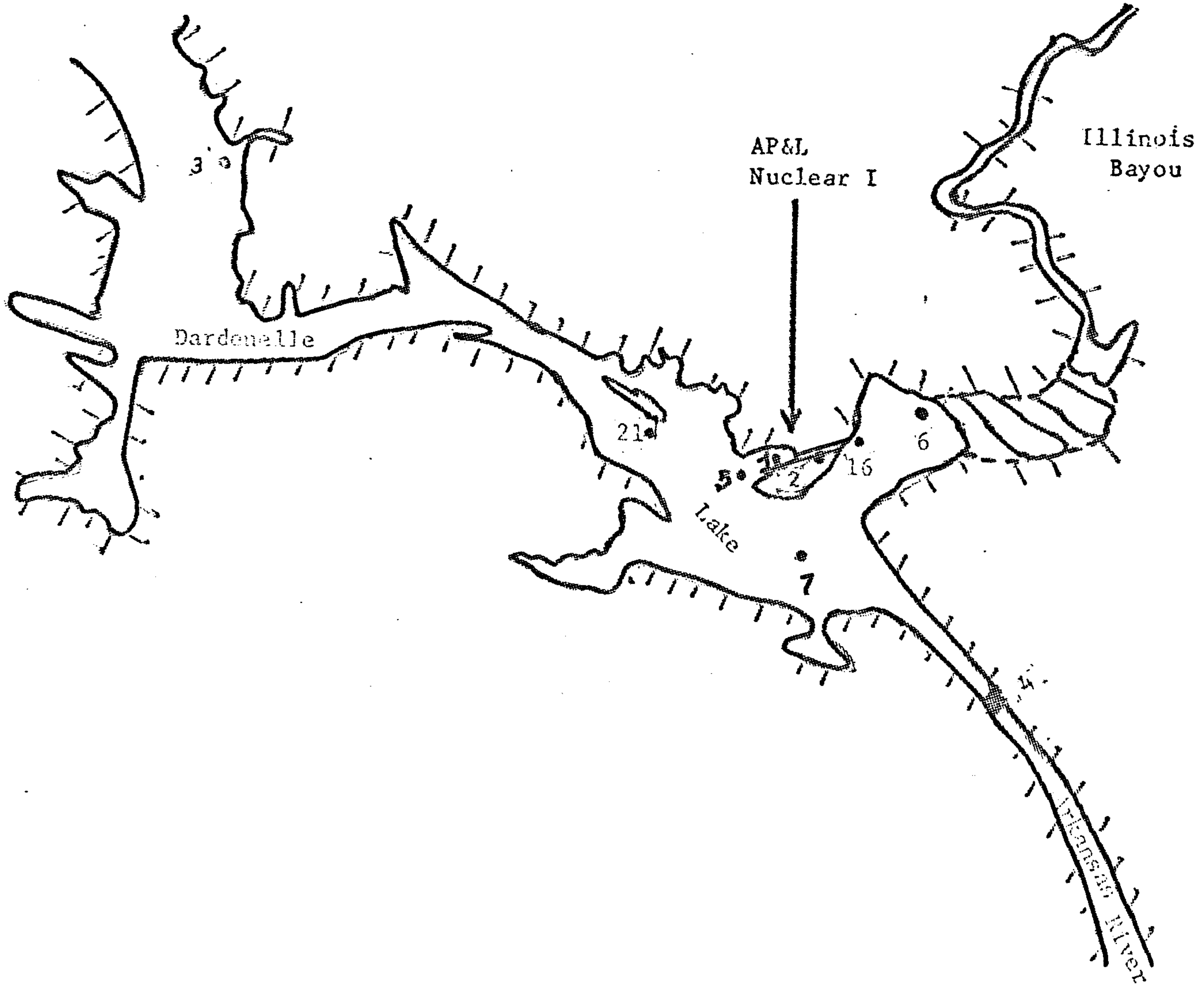
(1) Alternate stations were used in November and December of 1975. Station 5 and 7 were used instead of 1, 21 instead of 3, and 16 instead of 2.

Figure 1: Locations of Sampling Stations



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 HCl and 200 ml of water. The eluate was then evaporated to dryness.
5. The residue was dissolved in a minimum of conc. HNO_3 and the following fractions were separated:
   (a) Ce^{3+} as $Ce(OH)_3$ by the addition of conc. NH_3;
   (b) Co^{2+} as CoS by the addition of 1M thioacetamide;
   (c) Sr^{2+} as $SrCO_3$ by the addition of $(NH_4)_2CO_3$;
   (d) Cs^+ remains in solution, which is evaporated and the residue is heated to remove NH_4^+.
6. The Ce fraction, whose major impurity is Fe^{3+}, was purified by the precipitation of Ce^{3+} as the oxalate, the oxidation of Ce(III) to Ce(IV) followed by solvent extraction from the 9M HNO_3 phase into methyl isobutyl ketone. The Ce(IV) was reduced to Ce(III) and back-extracted into water. The purified Ce^{3+} was precipitated as the oxalate and counted in that form (1).
7. The Co fraction, with numerous impurities, was purified by dissolving the sulfide in conc. HNO_3, adding a Ni^{2+} holdback carrier and precipitating the mixed hydroxides with 10M KOH. After dissolving the hydroxides in glacial acetic acid, the Co^{2+} was precipitated as $K_3Co(NO_2)_6$. The Co^{2+} is redissolved by the addition of conc. HCl and precipitated for counting as CoS (2).
8. The Sr fraction, whose major impurity is Ca^{2+}, was dissolved in 9M HNO_3

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and the Sr^{2+} was precipitated as $Sr(NO_3)_2$ by the addition of fuming HNO_3. The precipitate was dissolved in water and the Sr^{2+} was precipitated as the carbonate (5,6). To assure that there was no Ca^{2+} contamination, the precipitate was again dissolved in a minimum amount of 1M HNO_3 and the Sr^{2+} was precipitated for counting as $SrSO_4$ by the addition of H_2SO_4.

9. The Cs fraction was separated by coprecipitation with ammonium phosphomolybdate. After the ammonium, phosphate and molybdate ions were removed, the Cs^+ was precipitated as the perchlorate and counted in that form (3).
10. The 2.00 liter aliquot was filtered and Ag^+ carrier was added. Sufficient conc. NH_3 was added to redissolve the resulting chloride. After a short mixing period, conc. HCl was added to precipitate AgCl. The AgCl was dissolved in 1M NH_3 and the solution was scavenged twice with $Fe(OH)_3$. The solution was again acidified with conc. HCl and the resulting AgCl precipitate was filtered and counted.
11. The 250 ml aliquot was filtered and evaporated to dryness. The unfiltrable solids were slurried and quantitatively transferred to aluminum planchets. After they were dried without baking, they were stored in a dessicator until they were counted (4).

C. 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 2 1/4" Beckman anti-coincidence 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.

D. Analysis of Data

All beta counting data were corrected for systematic errors by using the expression:

$$A_0(\text{pCi}) = \frac{R_0}{G \cdot f_c \cdot f_w \cdot f_b \cdot f_{ssa} \cdot 2.22} \qquad \text{(i)}$$

where A_0 = the activity (pCi) on the day of collection;

R_0 = the count rate (cpm) on the day of collection;

G = the geometry of the detector;

f_c = the correction factor for absorption of radiation by the sample cover;

f_w = the correction factor for absorption of radiation by the detector window;

f_b = correction factor for backscattering;

f_{ssa} = correction factor for sample self-absorption and scattering;

2.22 = dpm/pCi

The geometry factor was measured by determining the count rate of standardized samples of ^{3}H, ^{63}Ni and ^{90}Sr. The factors, f_c and f_w were estimated from standard beta absorption curves and are accurate to ±2%. The factor, f_b, was determined for aluminum and stainless steel planchets and it is accurate to ±1%. The values for f_{ssa} were derived from the data of Nervik and Stevenson (7) and are accurate to ±5%. The product of all the corrections factors is accurate to within ±7%.

^{58}Co had to be treated somewhat differently since 85% of its decays are by electron capture and 15% are by positron emission (8). To obtain a self-absorption correction factor for the x-rays, (I/I_0), the following expression was used:

$$\left(\frac{I}{I_0}\right) = \frac{\omega_K \left(\frac{K}{K + L}\right)}{(\rho x)(\mu/\rho)} \left[1 - e^{-(\mu/\rho)(\rho x)}\right]$$

where ρx = the CoS sample thickness in mg/cm^2;

(μ/ρ) = mass attenuation coefficient = 0.106 cm^2/mg (2);

$\left(\frac{K}{K + L}\right)$ = ratio of the number of K x-rays to the total number of x-rays emitted = 0.90 (8)

ω_K = fluorescence yield for Fe x-rays = 0.293 for K; 0.0 for L (8)

Auger electrons will not leave the sample or penetrate the cover since their range is $<$0.2 mg/cm^2. Thus the "f_{ssa}" factor for ^{58}Co can be calculated by "f_{ssa}" $= 0.15(f_{ssa})_{\beta^+} + 0.85(I/I_0)$.

This "f_{ssa}" value is then used in equation (i) to calculate the total activity of ^{58}Co.

To obtain the corrections factors for the gross beta samples, it is standard to treat them as though they were ^{137}Cs samples (4) even though they are a mixture of many activities.

The detection limits for this system, assuming a 50% yield of each fraction, is summarized in Table 2.

EXPERIMENTAL RESULTS

The concentrations of the various nuclides in Dardenelle Lake water samples are listed in Tables 3 - 10.

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(2) This value is obtained by averaging the (μ/ρ) values of Co (0.80) and S (0.173) (9)

Table 2: Detection Limits for the Selected Nuclides

^{137}Cs	0.03 pCi
^{144}Ce - ^{144}Pr	0.03 pCi
^{141}Ce	0.16 pCi
^{90}Sr - ^{90}Y	0.03 pCi
^{89}Sr	0.17 pCi
^{58}Co	0.10 pCi
^{110m}Ag	0.08 pCi

Tables 3 - 10: Concentrations
of the
Selected Nuclides

NOTES:
(a) Sample collected at Station 5
(b) Sample collected at Station 16
(c) Sample collected at Station 21
(d) Sample collected at Station 7
(e) Sample of very low chemical yield

Table 3: ^{144}Ce - ^{144}Pr Concentrations (pCi/l x 10^2)

Date / Sta. Number	11-22-75	12-17-75	1-23-76	2-28-76	3-27-76	5-25-76	6-21-76	7-23-76	8-19-76	9-24-76
1	(a) 6.3±0.6	(d) 0.3±1.0	0.4±0.2	1.0±1.0	0.9±0.4	0.0±0.5	2.4±0.3	1.4±0.3	2.9±0.4	6.1±1.7
2	(b) 6.9±0.7	(b) 4.6±1.3	0.1±0.4	0.1±0.9	0.0±0.6	3.0±0.8	2.3±0.4	0.3±1.2	1.3±0.4	0.5±0.4
3	(c) 0.0±0.1		1.1±0.7	0.5±0.5		3.9±0.6		1.9±0.8		
4	0.9±0.9		1.5±1.2	0.0±1.0	0.2±0.4	3.3±0.8	1.4±0.3	1.2±0.6	1.5±0.6	1.3±0.5
6		9.7±2.0			1.3±2.0		1.8±0.5		0.6±0.4	

Date / Sta. Number	10-29-76	12-4-76	1-23-77	2-18-77	3-26-77	4-27-77	5-18-77	6-14-77	7-16-77	8-20-77
1	3.6±1.3	1.2±0.7	0.0±1.0	0.6±0.6	2.1±0.8	7.1±1.0	6.5±0.9	4.2±0.7	8.1±1.2	1.8±0.7
2	2.3±1.0	2.9±1.4	0.4±0.7	0.0±1.7	1.3±1.0	11±2	6.3±1.1	10±2	5.9±1.2	7.2±0.9
3		0.0±1.3		0.0±0.6		15±2	5.9±1.1	9.5±1.1	7.5±1.0	8.5±1.0
4	2.0±1.0	0.0±1.1		0.4±0.6	2.0±0.6	9.1±1.2	5.7±0.9	6.7±0.9	3.4±0.9	5.8±0.6
6	1.7±1.2		0.5±1.0		2.8±0.7					

Table 4: ^{141}Ce Concentrations (pCi/l x 10^2)

Date / Sta. Number	10-29-76	12-4-76	1-23-77	2-18-77	3-26-77	4-27-77	5-18-77	6-14-77	7-16-77	8-20-77
1	10±2	2.4±2.4	3.7±1.1	2.2±1.5	2.1±1.8	0.0±1.0	3.7±2.1	0.6±1.3	0.0±2.6	4.5±1.0
2	8.8±2.0	8.0±5.3	1.6±0.9	2.2±1.7	4.5±1.7	0.8±2.0	4.3±2.9	0.0±6.1	0.0±3.2	1.5±1.5
3		3.4±1.3		3.2±2.1		0.0±3.8	4.6±4.0	0.0±3.5	4.7±2.7	0.0±4.9
4	15±2	6.4±3.0		6.9±1.8	2.8±1.2	5.3±3.1	0.0±2.5	0.0±2.4	1.2±1.7	3.2±1.1
6	14±3		6.8±0.8		0.0±2.2					

Table 5: ^{58}Co Concentrations (pCi/l x 10^2)

Date / Sta. Number	11-22-75	12-17-75	1-23-76	2-28-76	3-27-76	5-25-76	6-21-76	7-23-76	8-19-76	9-24-76
1	20±4 (a)	8.6±3.4 (d)	17±3	9.5±2.4	0.0±2.5	4.3±2.2	430±31	13±2	4.1±1.7	1.5±1.9
2	0.0±6.3 (b)	0.0±3.2 (b)	2.4±1.9	0.0±1.9	33±7	0.0±3.2	3.7±1.5	9.0±1.9	6.1±1.8	1.3±0.5
3	0±12 (c)		0.0±5.1	2.1±2.5		84±46 (e)		0.0±2.1		6.5±1.3
4	0±14	8.8±1.8	3.7±3.7	5.5±3.3	0.0±2.6	5.1±1.7	115±8	8.0±1.3	5.8±1.7	1.8±1.1
6		88±13			41±5		20±2		17±4	

Date / Sta. Number	10-29-76	12-4-76	1-23-77	2-18-77	3-26-77	4-27-77	5-18-77	6-14-77	7-16-77	8-20-77
1	0.0±1.7	0.8±1.6	15±2	0.0±3.7	4.4±2.5	0.0±1.7	0.0±2.6	9.4±3.8	8.0±2.1	8.4±1.6
2	0.0±2.0	0.0±1.7	5.5±3.0	3.4±1.5	0.9±3.6	1.6±2.2	0±14	0.0±6.3	1.3±2.5	0.0±9.2
3		0.0±2.0		3.4±6.8		1.5±3.0	0.0±2.7	5.2±3.5	1.6±3.1	6.4±3.2
4	2.2±1.7	7.2±1.7		3.2±3.2	25±3	0.0±2.0	3.1±2.5	11±3	7.8±2.3	1.7±2.6
6	4.7±1.9		1.8±2.2		0.0±2.3					

Table 6: ^{137}Cs Concentrations (pCi/l x 10^2)

Date / Sta. Number	11-22-75	12-17-75	1-23-76	2-28-76	3-27-76	5-25-76	6-21-76	7-23-76	8-19-76	9-24-76
1	8.8±1.5[a]	10±2 (d)	8.7±2.4	3.9±1.0	19±2	15±9	3300± 250	120±10	67±5	28±3
2	1.2±0.6[b]	1.6±0.8[b]	5.7±1.0	5.6±3.0	5.5±1.1	4.7±1.6	17±2	26±3	10±1	11±1
3	3.1±1.3[c]		5.2±0.9	4.4±0.9		8.8±1.2		14±2		6.1±0.5
4	10±2	0.0±1.0	7.8±1.0	9.7±1.5	3.0±0.8	4.0±1.3	240±20	53±4	19±2	7.8±0.7
6		0.3±1.2			5.3±2.0		130±10		24±3	

Date / Sta. Number	10-29-76	12-4-76	1-23-77	2-18-77	3-26-77	4-27-77	5-18-77	6-14-77	7-16-77	8-20-77
1	18±2	7.8±0.6	84±6	14±2	8.4±0.7	9.4±1.1	4.1±0.4	10±1	16±2	12±1
2	14±3	8.1±1.1	22±3	9.9±5.0	7.8±0.7	24±5	11±1	8.6±0.7	13±2	15±2
3		3.6±0.7		—		8.9±0.9	7.3±0.7	6.6±0.7	9.7±0.9	19±2
4	11±1	8.1±0.9		5.8±0.7	6.2±0.5	9.5±0.9	5.8±0.5	6.8±0.6	7.9±0.8	7.4±0.8
6	8.1±0.7				1.6±0.3					

Table 7: ^{90}Sr - ^{90}Y Concentrations (pCi/l x 10^{2})

Sta. Number \ Date	11-22-75	12-17-75	1-23-76	2-28-76	3-27-76	5-25-76	6-21-76	7-23-76	8-19-76	9-24-76
1	70±5 (a)	85±6 (d)	109±9	26±2	71±6	73±5	86±6	59±5	77±6	87±7
2	17±2 (b)	31±3 (b)	81±6	—	68±5	72±5	45±4	83±6	77±6	86±7
3	13±2 (c)		80±6	89±7		134±10		67±5		84±7
4	70±6	29±3	92±8	60±5	69±5	73±6	74±6	73±5	100±8	76±6
6					69±6		103±8		46±8 (e)	

Sta. Number \ Date	10-29-76	12-4-76	1-23-77	2-18-77	3-26-77	4-27-77	5-18-77	6-14-77	7-16-77	8-20-77
1	85±6	83±6	67±5	63±5	46±4	53±4	77±7	84±7	93±8	78±8
2	82±6	92±7	95±8	64±5	31±3	165±21	78±2	38±5	150±14	88±11
3		83±6		99±8		78±7	172±16	101±8	95±8	93±7
4	86±6	86±7		88±7	62±5	75±6	70±6	84±7	117±10	86±6
6	88±7		53±5		15±2					

Table 8: ^{89}Sr Concentrations (pCi/l x 10^2)

Date / Sta. Number	10-29-76	12-4-76	1-23-77	2-18-77	3-26-77	4-27-77	5-18-77	6-14-77	7-16-77	8-20-77
1	49±5	31±5	20±3	7.5±2.8	4.3±4.0	14±3	11±4	24±5	5.5±5.1	50±26
2	45±6	34±6	21±6	0.0±3.5	0.9±3.9	59±20	14±6	0.0±8.6	46±14	90±40
3		32±7		0.0±7.0		1.9±4.7	28±20	12±8	16±5	0±12
4	40±4	29±7		0.0±6.1	3.9±2.6	15±4	14±6	1.8±4.6	6.2±5.8	0±11
6	58±5		12±4		0.0±2.9					

Table 9: ^{110m}Ag Concentration (pCi/1 x 10^2)

Date / Sta. Number	11-22-75	12-17-75	1-23-76	2-28-76	3-27-76	5-25-76	6-21-76	7-23-76	8-19-76	9-24-76
1	223±31(a)	145±11(d)	38±14	41±20	22±13	59±35	158±30	11±22	36±20	27±19
2	43±16(b)	0±27(b)	21±21	0±20	14±17	32±20	17±6	27±17	0±18	0±19
3	39±28(c)		0±27	23±23				0±15		61±13
4	165±110	9±34	18±18	65±30	0±20	9±18	0±26	4±16	37±15	34±13
6		105±16			70±50		0±21		61±50	

Date / Sta. Number	10-29-76	12-4-76	1-23-77	2-18-77	3-26-77	4-27-77	5-18-77	6-14-77	7-16-77	8-20-77
1	39±31	0±23	13±13	0±13	3±13	32±18	14±19	0±13	27±15	0±28
2	28±12	10±38	26±13	2±12	3±12	18±18	0±10	0±12	6±13	59±12
3		0±15		19±11		0±13	0±11	0±15	21±14	0±12
4	0±10	23±12		0±46	10±25	0±12	0±16	0±18	0±11	33±20
6	0±10		8±10		66±28					

Table 10: Gross Beta Concentration (pCi/1)

Date / Sta. Number	11-22-75	12-17-75	1-23-76	2-28-76	3-27-76	5-25-76	6-21-76	7-23-76	8-19-76	9-24-76
1	(a)	(d)		1.3±0.2	2.5±0.2	2.7±0.3	37±3	5.3±0.5	6.3±0.5	5.8±0.5
2	(b)	(b)		1.8±0.3	1.9±0.2	2.1±0.2	3.3±0.3	2.2±0.3	6.1±0.5	4.5±0.4
3	(c)			1.0±0.2		4.3±0.4		2.2±0.3		4.7±0.4
4				2.2±0.3	4.2±0.4	3.3±0.3	7.2±0.6	4.2±0.3	5.8±0.5	5.1±0.5
6					1.1±0.2		3.5±0.3		5.0±0.4	

Date / Sta. Number	10-29-76	12-4-76	1-23-77	2-18-77	3-26-77	4-27-77	5-18-77	6-14-77	7-16-77	8-20-77
1	4.8±0.5	5.9±0.5	4.7±0.4	3.5±0.3	2.4±0.3	2.9±0.4	3.2±0.3	6.0±0.5	5.0±0.5	5.8±0.6
2	5.2±0.5	5.2±0.5	4.5±0.4	3.2±0.4	0.84±0.18	3.1±0.3	3.3±0.3	5.0±0.5	5.3±0.5	5.9±0.5
3		5.6±0.5		3.0±0.3		4.0±0.5	3.3±0.4	5.3±0.5	6.0±0.5	5.8±0.5
4	5.2±0.5	5.5±0.5		3.2±0.4	2.1±0.3	3.7±0.4	3.4±0.4	5.8±0.5	5.0±0.4	6.2±0.5
6	5.1±0.2		1.8±0.2		1.8±0.3					

CONCLUSIONS

1. The ^{144}Ce - ^{144}Pr concentrations seem little affected by reactor operation, although concentrations increased at every station sampled after the reactor had commenced full operation after refueling in the late winter of 1977. There are two possible explanations for this increase: (a) a natural increase at all stations due to fallout; (b) since the flow rate of the water through the dam and hydroelectric power station was generally quite low during most of the project period, release from Nuclear I followed by good mixing in the lake, could cause this general increase. Even so, the concentrations of the nuclides were consistently very low.
2. The ^{141}Ce and ^{90}Sr found in the ten samples were formed predominantly by Chinese nuclear testing in the atmosphere. The first of the three tests in the autumn of 1976 seems to be the dirtiest. There seems to be two significant injections of these nuclides into the lake water, one in the autumn of 1976 and the other in April-May, 1977 (see 6 below).
3. Amounts of ^{58}Co in excess of 0.12 pCi/1 appear only at station 6 between December, 1975 and August, 1976 and at most stations in the summer of 1976. There is no natural explanation for this very systematic long term appearance of relatively large amounts of ^{58}Co at station 6. It is interesting to note that the concentration decreases with a half-life of 72 days. In some way, this sampling area was contaminated with ^{58}Co and the contamination did not move with time.
4. In June, July and August, 1976, as the result of a planned release, unusually high concentrations of ^{58}Co were found in many samples. It will be noted that the concentration decreases slowly with time. This is

a result of the slow turnover of water during the dry spell of 1976 and 1977.

It was also noted that the dilution between stations 1 and 4 (some 2-3 miles distant) was only ~3-fold. This indicates very little mixing on the sampling day, but, over the long range, there seems to be significant mixing in the reservoir as a whole. This is borne out by the ^{137}Cs data.

5. The ^{137}Cs concentrations are somewhat affected by day-to-day reactor operation but the only significant injections of ^{137}Cs were in June, 1976 and in January, 1977. The trends mentioned in 3, above, can be seen also with ^{137}Cs. This dilution on release day was only ~14-fold and the effects of the release were seen for several months.

6. The ^{90}Sr-^{90}Y concentrations remained fairly constant, with a few exceptions, until January, 1977. At that time Nuclear I was shut down for refueling. During the down period, the majority of samples showed a marked decrease in ^{90}Sr-^{90}Y activity. Refueling was completed in late March and the activity in the following months showed a dramatic and regular increase. Unusually high activities began to sppear erratically at Stations 2 and 3 (the upstream stations). The ^{89}Sr concentrations also followed this erratic pattern, so it was tempting to ascribe the high concentrations in the spring of 1977 to fallout.

   Analysis of the increases in ^{89}Sr and ^{90}Sr, though, showed that this cannot be the case. ^{89}Sr began to appear with the fallout from the Chinese tests but there was very little change in the ^{90}Sr, since $A(^{90}Sr) < A(^{89}Sr)$ in the newly produced fallout even a year after the tests.

If the source of contamination is cooling water, the low $^{89}Sr/^{90}Sr$ ratio indicated that the contaminated water was stored for some time before release.

The Sr contamination could have also come from the outgassing of volatile precursors (^{89}Kr and ^{90}Kr) into the atmosphere. This could account for the widespread appearance of increased activities, both upstream and downstream.

These increases in Sr activity coincide with increases in ^{137}Cs in many samples during the spring of 1977. ^{137}Cs also has a volatile precursor, ^{137}Xe.

Reactor operation is causing an increase in the ^{90}Sr-^{90}Y activity in Lake Dardenelle by a factor of 1.5-3. Reactor operation may also have been the cause of the increase of ^{89}Sr activity in the spring of 1977.

7. There is no discernible pattern to the ^{110m}Ag concentration except to note its appearance in appreciable amounts in the planned release of June 21, 1976 and in a few samples in the winter of 1975-1976.

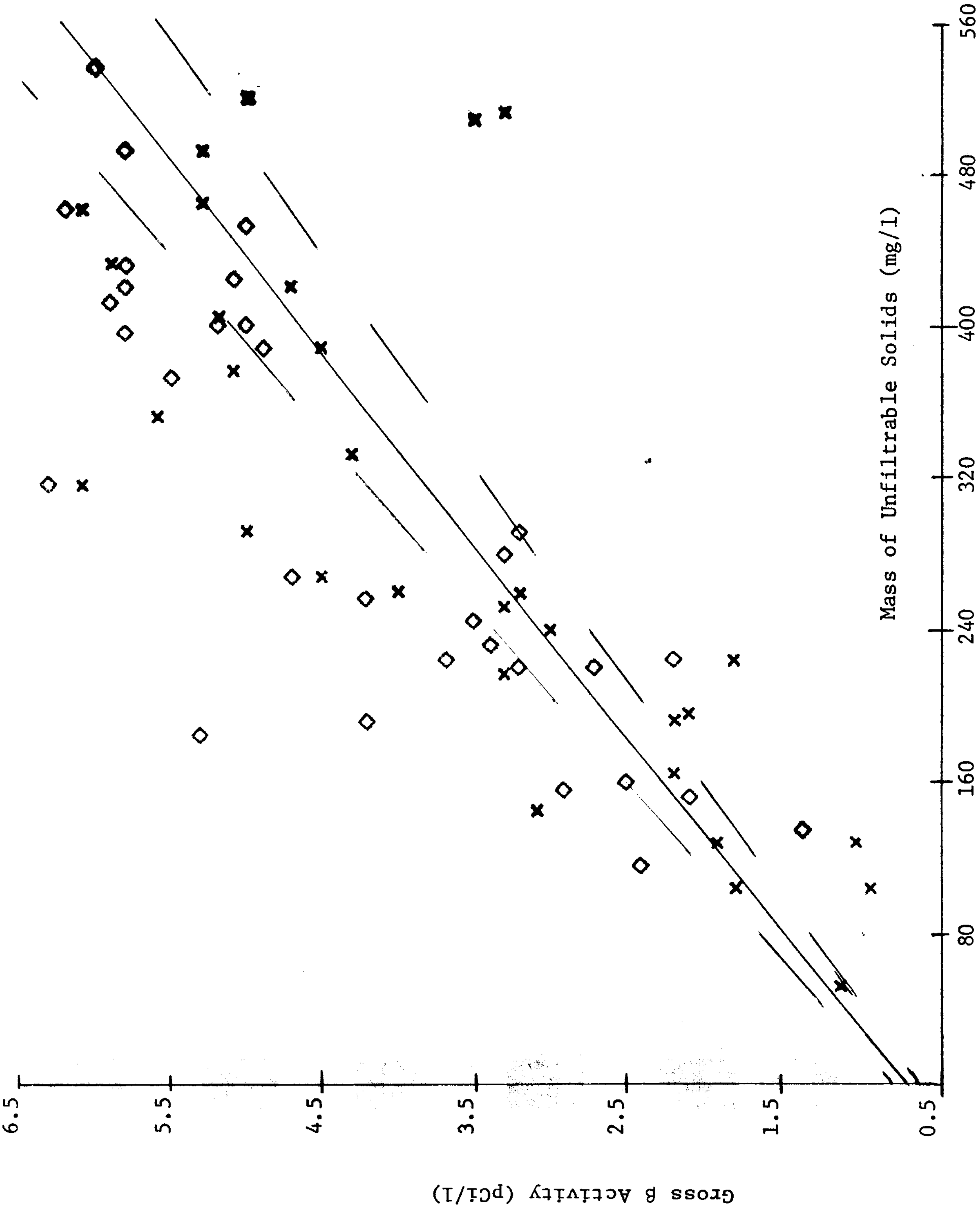
8. It was a good deal more difficult to analyze the gross β data. Winter samples usually contain smaller amounts of solids and thus lower activities. Sample sizes were also more variable than in summer. Graphical analysis lends itself to these data. Figure 2 is a plot of the gross β activity (pCi/l) vs. the mass of unfiltrable solids per liter of water. It can be seen that many points fall on or near the solid line. The dashed lines are the ±10% limits which

Figure 2: Total Gross β Activity Per Liter of Water as a Function of the Mass of the Unfiltrable Solids

x - upstream stations (2,36,16,21)

◇ - downstream stations (1,4,5,7)

Two points for the samples of 6/21/76 have been omitted because they are off the activity scale on graph.


Gross β Activity (pCi/l)
0.5
1.5
2.5
3.5
4.5
5.5
6.5
Mass of Unfiltrable Solids (mg/l)
80
160
240
320
400
480
560

approximate the standard deviation. The equation for this line can be expressed as:

$$A = (\text{sp. act.})m + b$$

where A = activity of the gross β sample (pCi/l)

(sp. act.) = the specific activity of naturally occurring radioactivity (pCi/mg)

m = mass of unfiltrable solids per liter of water (mg/l)

b = activity of other radio active nuclides that did not result from reactor operation (e.g. fallout)

Any point above the solid line represents a sample which, in addition to natural radioactivity (mostly ^{40}K) and radioactivity from fallout, contains some radioactive nuclides that are reactor produced.

If this approach is valid, the slope (sp. act.) should agree with the specific activity of ^{40}K in river water and the intercept (b) should correspond to the total activity of the fallout. The value for the slope was determined to be $\simeq 1.0 \times 10^{-2}$ pCi/mg and the intercept to be $\sim 0.7 - 0.9$ pCi/l.

If it is assumed that all the naturally occurring activity is due to the decay of ^{40}K, the unfiltrable solids would contain 1.1% K by weight.

Mason (10) cites a value of ~2% for the K content of dissolved solids in river water, twice the percentage found in Dardenelle Lake water. The low value may be explained by the fact that the Ca content of the Arkansas River is much higher than the value for Ca quoted by Mason

The intercept is also of the right magnitude. ^{90}Sr - ^{90}Y is the major radionuclide in samples unaffected by reactor effluent and its concentration is normally in the range 0.7 - 0.9 pCi/1.

It is safe to assume that points above the A vs. mass line indicate the presence of reactor generated nuclides. An appreciable number of samples, from both upstream and downstream stations show this excess activity.

It will be noticed that some points lie significantly below the line. The percentage of K must be lower here than in the average sample. These points are all for upstream samples, most of them taken from station 2, where the water is moving quite swiftly and perhaps carrying a larger colloidal load than at other sampling sites.

9. Overall, it can be concluded that the day-to-day operation of Nuclear I has very little effect on the radionuclide concentration in Dardenelle Lake water. The fallout from the Chinese tests was more evident. Only planned releases caused significantly increased concentrations. Dilution by river water may not be as effective as could be wished but this may be a seasonal effect. The slow turnover of lake water during dry periods also poses a problem because of the spread of reactor produced radionuclides throughout the lake.

10. It must be pointed out that, at no time, did the concentrations of the measured nuclides exceed the limits set by the Arkansas State Department

of Health. Concentrations were so low that it was decided not to pursue a study of the uptake of radionuclides in biological systems or in their deposition with sediments.

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