Technical Reports   Arkansas Water Resources Center

12-1-1981

Radionuclides in Dardanelle Lake in the Area of the Nuclear I Facility: 1979-1981

D. M. Chittenden II
University of Arkansas, Fayetteville

Recommended Citation

Chittenden, D. M. II. 1981. Radionuclides in Dardanelle Lake in the Area of the Nuclear I Facility: 1979-1981. Arkansas Water Resources Center, Fayetteville, AR. PUB081. 42

RADIONUCLIDES IN DARDANELLE LAKE IN THE AREA OF THE NUCLEAR I FACILITY: 1979-1981

By
D.M. Chittenden II

AWRRC

Arkansas Water Resources Research Center

PROJECT COMPLETION REPORT

PROJECT NO: A-050-ARK   AGREEMENT NO: 14-34-0001-0104

Starting Date: October 1, 1979   Ending Date: December 31, 1981

Radionuclides in Dardanelle Lake in the
Area of the Nuclear I Facility: 1979-1981

by

D. M. Chittenden II

Arkansas Water Resources Research Center
University of Arkansas
Fayetteville, Arkansas 72701

December, 1981

Publication Number 81

ABSTRACT

RADIONUCLIDES IN DARDANELLE LAKE IN THE
AREA OF THE NUCLEAR I FACILITY: 1979-1981

The variations of the concentrations of ^{90}Sr and ^{137}Cs at four stations
in Dardanelle Reservoir were analyzed as functions of two parameters:
concentration of ionic species and the activity released, A_r, from the

two 900 Mw reactors which use the reservoir as a source of cooling water.
Multiple regression analyses were performed on the radionuclide concen-
trations using the two parameters as predictors. The analyses indicated
that ^{90}Sr is in a state of equilibrium between the solution and the sus-
pended sediment. The position of the equilibrium was found to be quite
sensitive to changes in the concentration of alkaline earth cations,
M(II). Thus, the concentration of M(II) has a substantial effect on the

removal of ^{90}Sr from the water column. The ^{137}Cs concentration varied
as A_r, indicating that solution-sediment equilibrium has not been attained

since the time necessary for newly released ^{137}Cs to come to equilibrium
is greater than the mean residence time of water in Dardanelle Reservoir.

The occurrence of ^{144}Ce-^{144}Pr in the Reservoir seems to be a consequence

of refueling or maintenance of the reactors; the ^{141}Ce as well as ^{144}Ce,
it appears, were originally airborne and find their way to the reservoir
through runoff.

TABLE OF CONTENTS

INTRODUCTION

This report summarizes the results of a program designed to measure low levels of some common radionuclides found in Dardanelle Reservoir, Arkansas and to correlate those concentrations with geochemical processes and/or with man-initiated events, particularly at Arkansas Nuclear One facility operated by Arkansas Power and Light Company. The facility houses two 900 Mw reactors; the second unit came on line in the summer of 1980.

The project running from May, 1979 to September, 1981 is a continuation of a similar project (1) conducted during the period from November, 1975 to September, 1977. It had been observed, upon a re-examination of ^{90}Sr data from 1975-1977, that the concentration of this nuclide varied as the value of the total dissolved solids (TDS). To define the geochemical parameters which control this relationship, it was necessary to determine the concentration of ions similar to Sr(II), i.e., Ca(II) + Mg(II) [hereinafter referred to as M(II)], in addition to the concentrations of ^{90}Sr-^{90}Y, ^{89}Sr, ^{144}Ce-^{144}Pr, ^{141}Ce, ^{58}Co, ^{137}Cs, and the gross beta activity in the water of Dardanelle Reservoir. As the project developed, the concentrations of all the major ions found in Lake Dardanelle were determined. Added to the list of stable species were K^+, HCO_3^-, Cl^-, SO_4^{2-}, and NO_3^-. These were added as a check on the M(II) and Na^+ determinations, since the number of equivalents of these anions and cations should be roughly equal.

Later still in the project, bottom sediments and suspended sediments were analyzed for ^{90}Sr and ^{137}Cs.

Phenomena involving equilibrium exchange of ions between the natural aqueous medium and the surface of suspended particles have been observed.

Noshkin and Bowen (2) have shown that while Pr appears to be removed from sea water by association with sinking particles, ^{137}Cs and ^{90}Sr remain in the water phase. Since some of these nuclides entered the marine environment associated with particulate matter, desorption due to increased ionic strength must be possible. The reversibility of the adsorption of ^{65}Zn and ^{54}Mn has been demonstrated by Evans and Cutshall (3). It was found that these nuclides were desorbed from suspended particles when the fresh water of the Columbia River was mixed with sea water in the river's estuary. The desorption of ^{65}Zn occurred from suspended particles but not from bottom sediment; on the other hand, ^{54}Mn was desorbed from both (4). In sea water, there is essentially no equilibrium established between adsorbed and desorbed radionuclides because the large concentrations of species like Ca(II) force the equilibrium all the way to the side of the dissolved radionuclides.

Studies of the sediment-solution interaction in fresh water, using both reactor-produced and fallout radionuclides as tracers, have led to a better understanding of the means by which dissolved species are removed from a body of water. The major routes for the removal of radionuclides from lakes have been described by Lerman and Lietzke (5).

The route of most immediate interest in the study of ^{137}Cs and ^{90}Sr is adsorption by suspended and deposited sediment. In Lake Erie the most common mode of removal of ^{137}Cs is flux to the sediment but sedimentation is a poor sink for ^{90}Sr. Lerman and Taniguchi (6) estimate that only 2-6% of the ^{90}Sr introduced into the Laurentian Great Lakes is removed by interaction with the sediment while sedimentation removes more than 80% of the ^{137}Cs. These findings have been confirmed by Alberts and Wahlgren

(7) in their comparison of the 1976 and 1979 concentrations of these

nuclides in these lakes. In the twenty years covered by these two studies,

there has been little variation observed in the ^{90}Sr values. This is to

be expected because the mean residence times in the Great Lakes range from

2.6 to 31 years. Thus no significant change in the concentrations of

either fallout radionuclides or stable species such as Ca(II) would be

expected. Even if an equilibrium had been established between dissolved

and adsorbed species, it would be difficult to detect.

In contrast to the static conditions to be found in the Great Lakes

and in sea water, the concentrations of ^{90}Sr, ^{137}Cs, and stable species in

Dardanelle Reservoir, Arkansas vary widely over the period of a year.

Dardanelle Reservoir, formed by the impoundment of the Arkansas River

southwest of Russellville, has a volume of 486,200 acre feet. The monthly

flow through ranges from 200,000 to 20,000,000 acre feet except in periods

of severe drought; thus, the mean residence time is usually less than two

months and often on the order of weeks. The concentration of dissolved

species can change quite rapidly; the total dissolved solids (TDS) ranges

from 200 to 730 mg/L (8).

The major source of ^{90}Sr in Dardanelle Reservoir is fallout carried

by the inflowing water. Only minor amounts are released from the two 900 Mw

reactors, operated by Arkansas Power and Light Company Plant (Arkansas

Nuclear One), which use the reservoir as a source of cooling water. Vari-

ations in the ^{90}Sr concentration should only be linked to the natural

removal routes of the lake environment. Conversely, the bulk of the ^{137}Cs

is reactor produced and the variation in its concentration should be

strongly linked to reactor releases and less strongly linked to the

removal processes if equilibrium is not rapidly achieved.

The factors affecting the concentrations of the other nuclides will also be analyzed. The bulk of these species was, during this time of the study, reactor produced.

EXPERIMENTAL METHODS

The concentrations of gross beta activity, $^{90}Sr-^{90}Y$, ^{89}Sr, $^{144}Ce-^{144}Pr$, ^{58}Ce, ^{58}Co, and ^{137}Cs were measured in samples collected in May and August of 1979 and monthly from October 1979 to August 1981. Samples of approximately 20 L in volume were collected from the surface at four stations (see Figure 1) and analyzed for the radionuclides. At the same time, a 1.0 L sample was collected for the stable ion analysis. The stations were: (1) 400 m downstream from the reactor's cooling water discharge canal; (2) 400 m upstream from the reactor's cooling water intake; (3) 15 km upstream from the reactor facility; (4) 8 km downstream from the reactor facility at the Dardanelle Lock and Dam. The samples were taken from the surface since, at Station 1, it was found that the heated water discharged from the facility remained in a surface layer two feet in depth.

The radiochemical procedures for the isolation of the Sr, Ce, Co, and Cs fractions have been fully described in the report for the 1975-77 study (1). A brief outline of the chemical procedures follows.

The 20 L samples were acidified and filtered through Whatman 42 paper, the necessary carriers (20-25 mg of cation) were added, and the sample was passed through a column containing ∿ 100 g of Dowex-50x8, 100-200 mesh, at a rate of 1 L/hr. The water passing through the column was discarded.
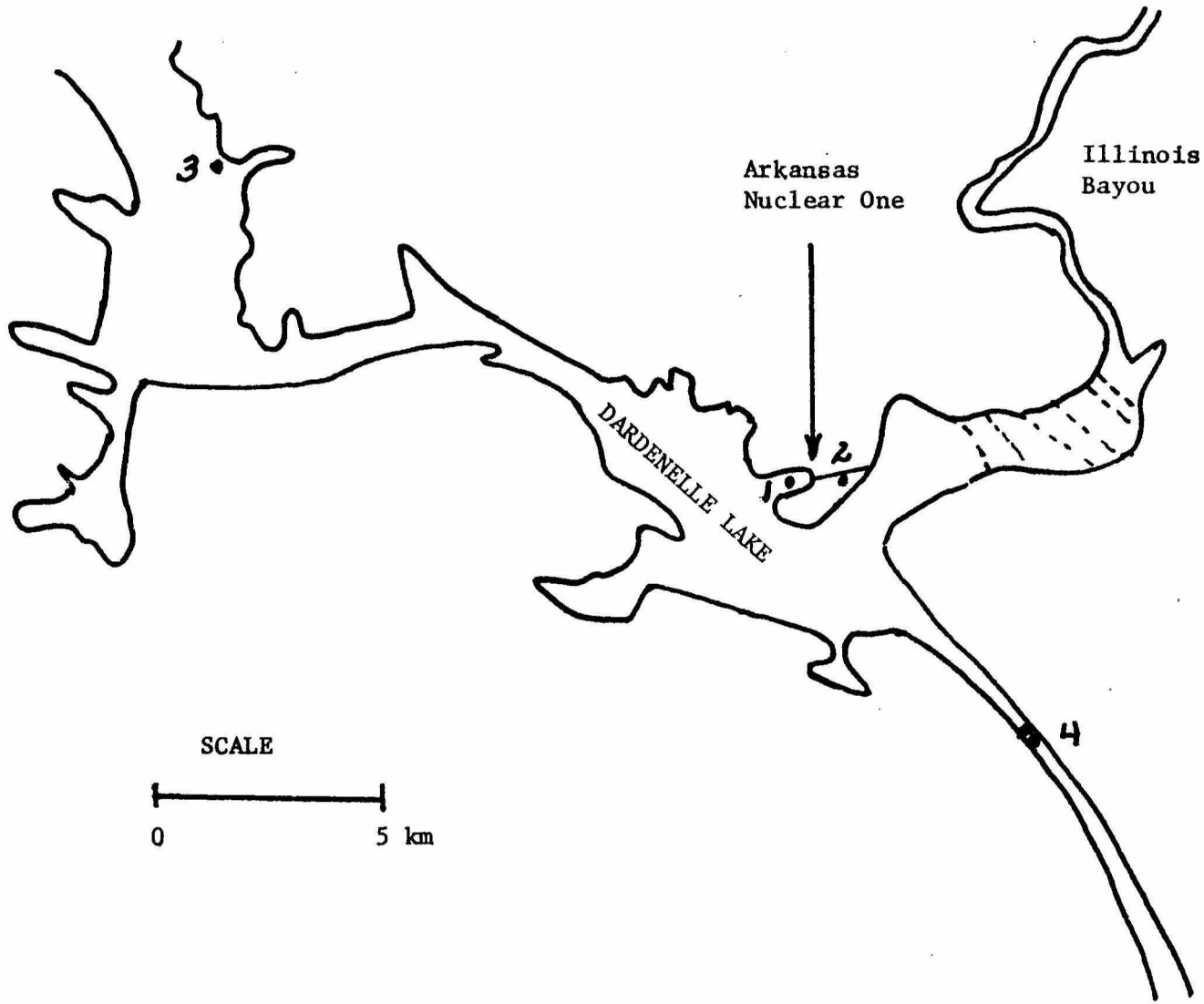
Figure 1: Locations of Sampling Stations

-5-

The cations were eluted from the column with 1 L of 6N HCl. The eluate was evaporated to dryness and the solids were redissolved in dilute acid and the solution was then made basic with NH_4OH. The Ce fraction was coprecipitated with Fe as the hydroxide. After redissolving, the Ce was precipitated as the oxalate. The Ce was further purified by extraction of Ce(IV) into methyl isobutyl ketone and finally precipitated as the oxalate (9). The insoluble sulfide fraction was precipitated from basic solution by the addition of thioacetamide. The cobalt fraction was puri- fied by precipitation twice as $K_3Co(NO_2)_6$ and the purified Co^{2+} was precipitated as CoS (10). The insoluble carbonates were then precipitated and processed for the strontium fraction. The resulting decantate was processed to isolate the cesium fraction as a $CsClO_4$ precipitate from ethanol (11). The procedure for the purification of the strontium was essentially that of Hodges (12), except that the strontium was finally precipitated as $SrSO_4$. The final precipitates were filtered onto a paper disc, dried, weighed, mounted on a stainless steel planchet, and covered with a mylar film of 1-4 mg/cm^2 thickness.

Gross beta samples were obtained by evaporating 125-250 mL of filtered water. The solids were transferred quantitatively to cupped aluminum planchets and dried at 105°C to constant weight and stored in a dessicator until counting. TDS was calculated from this weight.

The suspended solids were removed from the filter paper and leached of Sr(II) and Cs(I) by stirring for two hours in 400 mL of 6 N HNO_3 spiked with stable Sr(II) and Cs(I) carriers. The slurry was filtered and the filtrate was processed in the same manner as the eluate from the ion- exchange column described above.

Bottom sediments were collected from the upper one centimeter of the river bed by an Eckmann dredge. Only Stations 1 and 3 were sampled since the bottom of the intake canal is scoured and the bottom at the dam is at such a depth as to make dredging very difficult.

The samples were dried at 105°C and ground in a ball mill to pass a 40 mesh sieve. The fine mesh was necessary for the removal of shards of shell found in the sediment at Station 1. A 40-70 g sample was slurried with 400 mL of 6 N HCl containing carriers and stirred for an hour. The slurry was filtered through two thicknesses of Whatman 42 paper. The solids were resuspended in 400 mL of 6 N HCl, stirred for another hour, and filtered. The combined filtrates were passed through $\sim$ 100 g of Dowex 1 to remove Fe(III). The eluate was evaporated to dryness and processed as the eluate described above.

All samples were beta counted in either a Tennelec LB-1000 low background (0.97 cpm) counting system or a Tracerlab Omni-Guard system (0.60 cpm background). After correction for self-absorption and self-scattering (13) and for chemical yield, the activities of the samples were obtained from comparison with a standard.

The concentration of the major ions except sulfate and nitrate (14) were determined by standard methods (15).

All statistical computations were performed using the package MINITAB routine (16) available on the University's Harris/7 computer.

RESULTS AND DISCUSSION

The concentrations of the radionuclides and the stable ionic species is presented in the appendices.

A. <u>Gross</u> <u>Beta</u> <u>Activity</u>.

The gross beta activity A_g is a good measure of the activity released by the reactor, A_r. Since there is usually an appreciable quantity of K^+ in the water, there is a "natural" contribution to the gross beta activity. If this contribution and the contribution from fallout $^{90}Sr-^{90}Y$ is subtracted from the gross beta activity, the result will be A_r.

$$A_r = A_g - (\text{activities of } {}^{40}K + {}^{90}Sr-{}^{90}Y) \qquad (1)$$

The second term can be obtained from analysis of a plot of gross beta activity vs. TDS (Figure 2). The line drawn through the points of minimum measured activity for given TDS values is a function of TDS only and can be safely assumed to represent the second term; A_r is then equal to the deviation of a measured point from this line

$$A_r = A_g - 0.0108(\text{TDS}) \qquad (2)$$

The same analysis can be done on a plot of gross beta activity vs. concentration of K^+. Ideally, the relationship should be

$$A_r = A_g - [C(K) + \text{activity of } {}^{90}Sr-{}^{90}Y] \qquad (3)$$

where K = the concentration of K^+ in mg/L

and C = the specific activty of ^{40}K = 0.75 pCi/mg K

The slope of the line through the points of minimum activity leads to an empirical value of C = 0.85 pCi/mg K, a reasonably good agreement.

B. <u>Sr-90</u> <u>and</u> <u>Cs-137</u>.

The dependence of the ^{90}Sr concentration upon TDS in the water taken from Station 1 (Figure 3) was first noticed when the complete body of data from the 1976-1977 period was analyzed. Statistical analysis of the data indicated that a reasonably good fit of the data could be

Figure 2: The Variation of Gross Beta Activity
as a Function of TDS
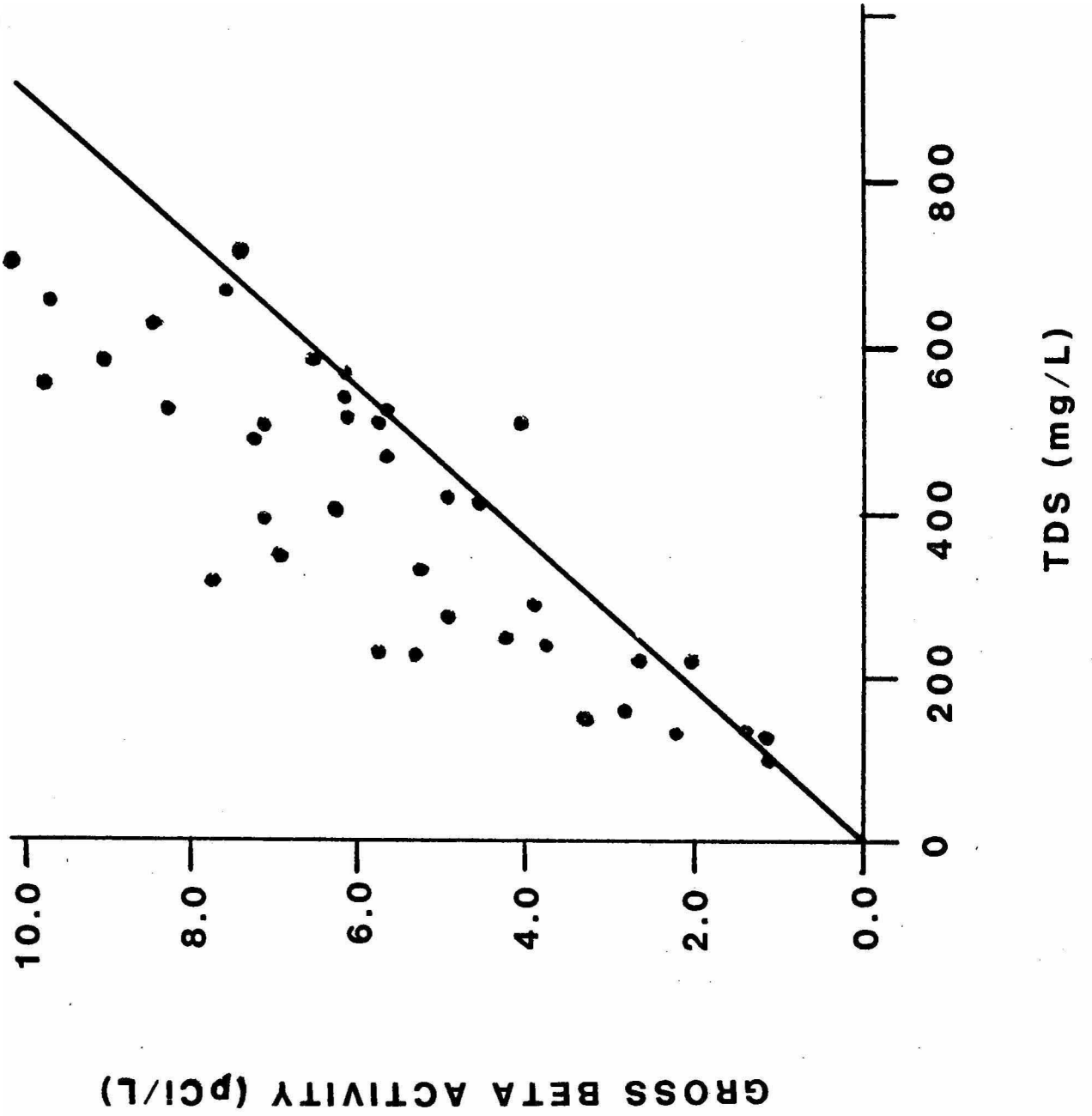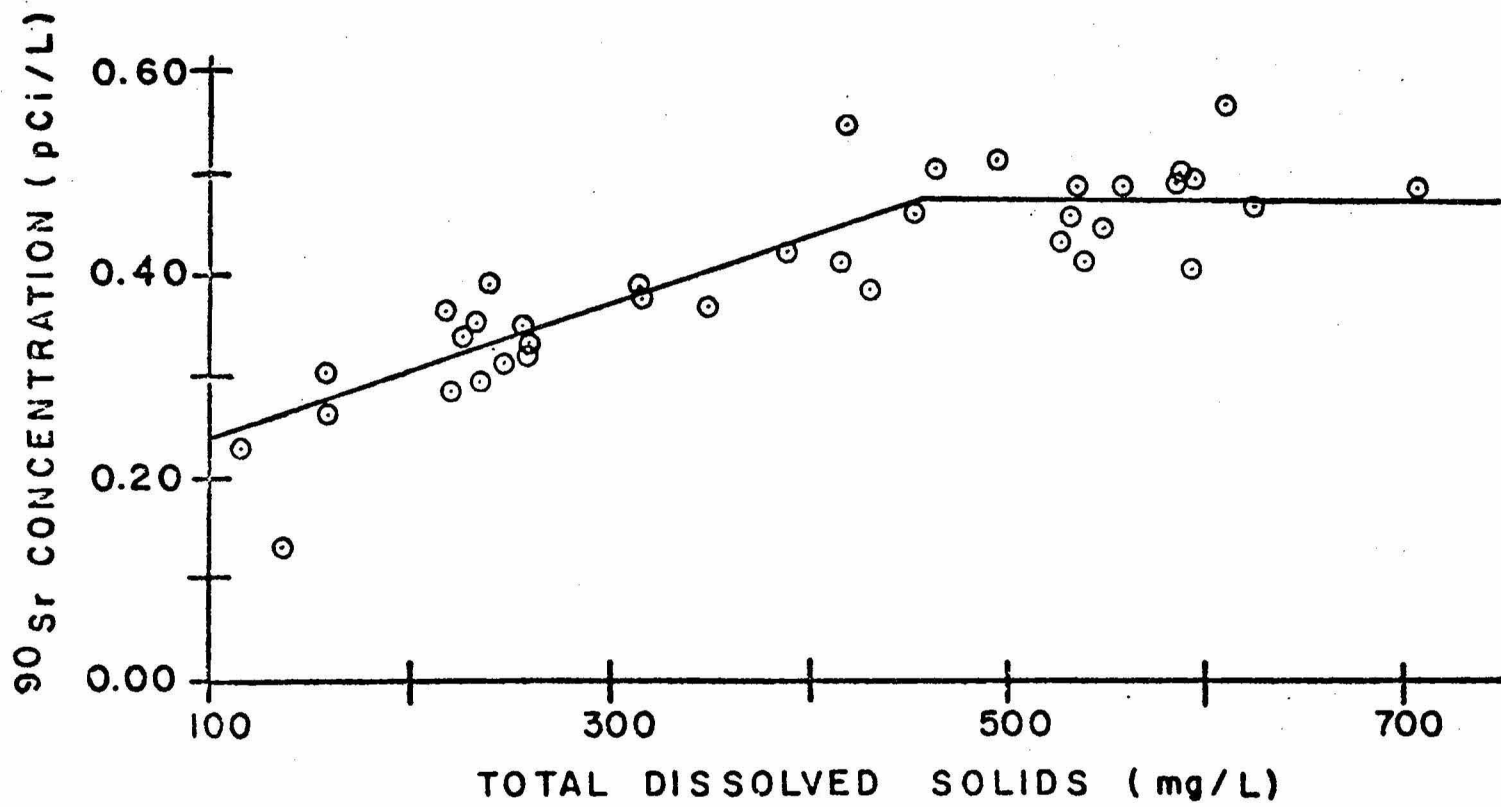
Figure 3: Dependence of ^{90}Sr Activity upon TDS: Station 1

obtained by assuming that the points described two straight lines joined by a small region of steadily decreasing slope; an equally good fit was provided by an exponential curve asymptotically approaching a maximum ^{90}Sr concentration. The second option was discarded in the light of later data. Since the region of changing slope is quite small, it will be ignored in this and subsequent figures and discussion. For the sake of simplicity, the data will be treated as if they described two intersecting straight lines.

The correlation coefficient for ^{90}Sr concentration and TDS for TDS values less than 450 mg/L was $\sim$ 0.75, a remarkably high value considering the number of variables that could affect the ^{90}Sr concentration. At TDS values greater than 450 mg/L, the concentration was a constant 0.48 ± 0.04 pCi/L.

To eliminate the second option it was necessary to determine the dependence of the ^{90}Sr concentration upon the concentration of M(II). The concentration of these ions should be a better predictor of the ^{90}Sr concentration because of their chemical similarity.

The dependence of dissolved ^{90}Sr upon the concentration of M(II) at Station 1 can be seen in Figure 4. It is obvious that the best fit is to the two straight solid lines rather than an exponential curve. At M(II) concentrations less than 2.5 meq/L the straight line was assumed to have the equation

$$^{90}\text{Sr} = a + bC_{M(II)} \qquad (4)$$

where ^{90}Sr = the concentration of ^{90}Sr in pCi/L

$C_{M(II)}$ = the concentration of M(II) in meq/L

a,b = constants

Figure 4: Dependence of ^{90}Sr Activity upon
M(II) Concentration: Station I
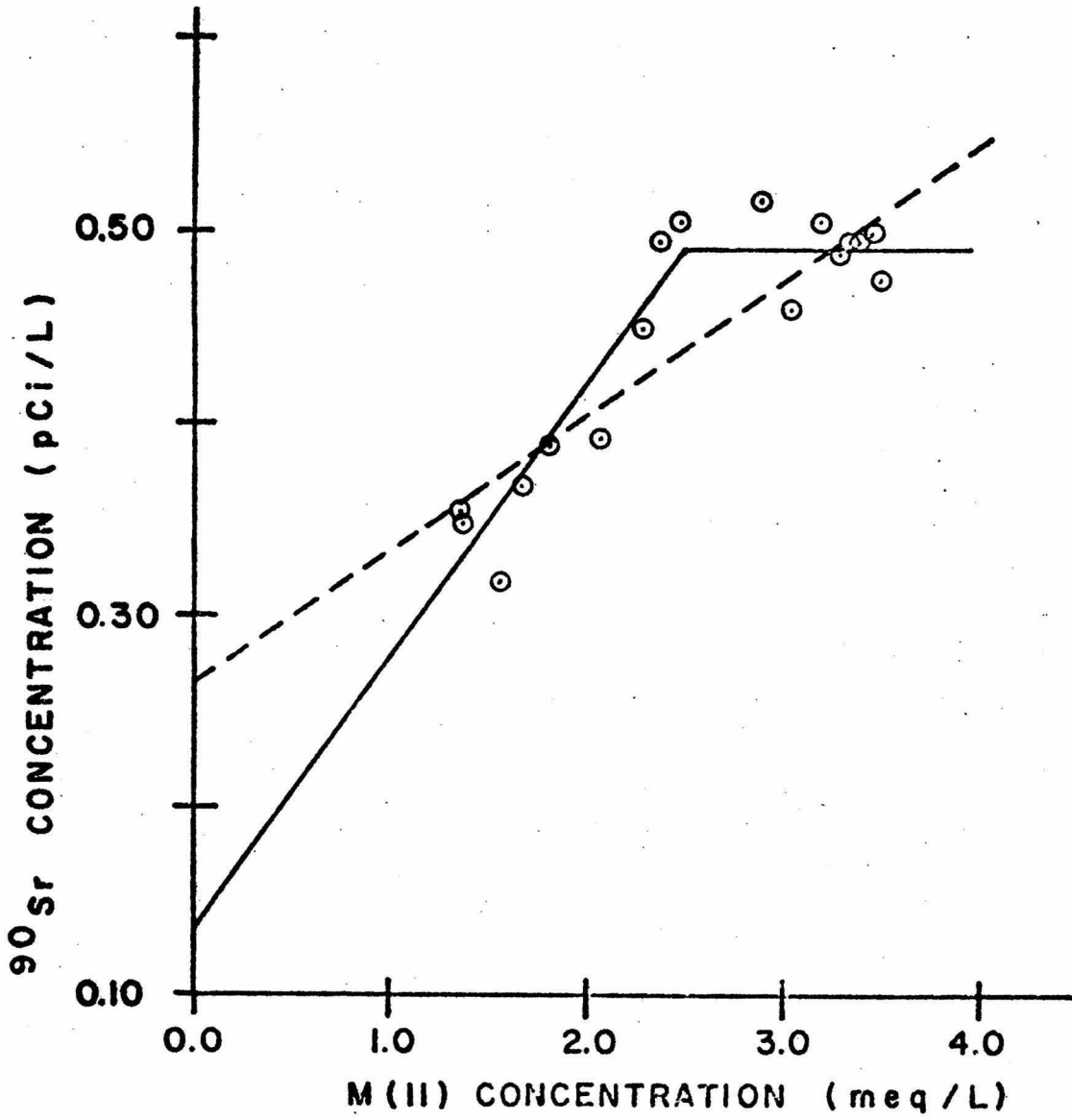
This equation can be used on the assumption that the solution has reached equilibrium with respect to ^{90}Sr, M(II), and any processes that would result in the addition or removal of ^{90}Sr from solution.

If ^{90}Sr had been newly injected into solution by release from the reactor facility, a better fit might be obtained from

$$^{90}Sr = a + b_{M(II)} + cAr \qquad (5)$$

where   c = constant

    A_r = activity of all nuclides newly injected in pCi/L

Two methods were available to determine A_r. If the K(I) concentration is known, as in the 1979-1981 period, equation (3) was used. Equation (2) was used when points from both the 1975-1977 study and the 1979-1981 study were considered. There is very little difference in their utility as predictors. The constants a, b, and c were determined by fitting the data to a straight line using linear and multiple regression analyses.

For M(II) concentrations greater than 2.5 meq/L, the value for the ^{90}Sr concentration is simply the mean of all the values of the ^{90}Sr associated with these high M(II) values.

Table I is a tabulation of the values of a, b, and c obtained by regression as well as the R^2 for the regression line. R^2 is the percentage of the variance in ^{90}Sr concentration that can be predicted from the values of the parameters that have been chosen as predictors, i.e., the concentration of M(II) below 2.5 meq/L and A_r. It is a

Table I: Constants for Equation (2) and R^2 Resulting from Multiple Regression and the Maximum Activity at Stations 1, 2, and 4.

	$C_{M(II)} \leq 2.5$ meq/L			R^2(%)			$C_{M(II)} > 2.5$ meq/L
Station	a	b	c	Due to M(II)	Due to A_r	Total	^{90}Sr(pCi/L)
1	0.135± 0.041	0.144± 0.022	--	86.5	0.0	86.5	0.48 ± 0.03
2	0.168± 0.046	0.094± 0.029	0.0117± 0.0062	81.4	7.8	89.2	0.46 ± 0.04
4	0.107± 0.094	0.098± 0.048	0.0453± 0.0200	48.6	26.1	74.7	0.47 ± 0.03

Errors listed are ± 1σ

measure of the validity of the prediction equation. Also included is

the maximum and constant value of the ^{90}Sr concentration that occurs

when the concentration of M(II) exceeds 2.5 meq/L. The contribution

of each parameter to the total R^2 is also included. It can be seen

that the M(II) concentration is by far the most important predictor;

only at Station 4 is the contribution of the A_r term important.

Also included in Figure 4 is a single dashed line derived from

the linear regression assuming only one straight line. The R^2 value

for this single line is 74.2%, a respectable value. Upon inspection

of Figure 4, however, it can be seen that we can ignore this possibility.

The deviations of the experimental points from the solid regression lines

are random, while the deviations from the single dashed line are not.

At low and high values of M(II) the deviations are uniformly negative

while in the middle range, they are uniformly positive. This situation

is typical of a poor fit of data regardless of the R^2 value. In addi-

tion, the TDS vs. ^{90}Sr curve is undoubtedly of the two-line type.

Statistically, the assumption of two intersecting straight lines is to

be preferred over a single straight line.

That the M(II) concentration is a better predictor of the ^{90}Sr

concentration than TDS is not surprising but the high value of the R^2

was quite unexpected. It approached the range (>90%) expected of

laboratory experiments in which the extraneous variables are well con-

trolled.

This straight line rise of the ^{90}Sr concentration to a maximum

and essentially constant value is due to the increasing fraction of

exchange sites on the sediment particles being occupied by M(II) rather

than ^{90}Sr(II) as the concentration of aqueous M(II) increases. At

M(II) concentrations greater than 2.5 meq/L, M(II) occupies 100% of

the exchange sites, i.e., saturation conditions, and the ^{90}Sr concen-

tration remains constant at the ∿ 0.5 pCi/L that is characteristic of

the rainfall plus runoff of this geographical area.

The relationship between M(II) and ^{90}Sr is atypical at Station 3.

The data fits well none of the possibilities considered above. The

best fit is to the case of a single straight line (Figure 5). The R^2

value for this regression line is 48.5%; the contribution of the A_r

term is negligible which is to be expected so far upstream. This

anomaly may well be due to the difference in the flow conditions at

Station 3. Station 3 is in a littoral region where the water is

quiescent. Stations 1 and 2 have flow conditions that approach tur-

bulence. On numerous occasions, the analyses here have differed

significantly from those at other stations. Nonetheless, there is

still a strong indication that the ^{90}Sr concentration is dependent on

the M(II) concentration.

The concentration of ^{90}Sr in suspended sediments should exhibit

an inverse relationship to the concentration of M(II) in solution.

There are indications of such a trend, summarized in Table II. The

uncertainty in the ^{90}Sr values is large because the samples were small,

as little as 0.18 g, and the count rate for the Sr fraction was only

slightly above background. In January, 1981, the samples were so small

that they were combined for a single analysis. Consequently, a weighted

average for each month is included in Table II. These averages indicate

that there is an inverse relationship between adsorbed ^{90}Sr and aqueous

Table II: Concentrations of ^{90}Sr(pCi/L) in Sediments as Function of Aqueous M(II) Concentration

(meq/L)

Type of Sediment	Date	Species	Station #1	#2	#3	#4	Combined Sample (a)	(b)
Suspended	7/81	^{90}Sr	0.14±0.11	0.20±0.20	0.00±0.15	0.14±0.18	0.12±0.10	
		M(II)	2.88	2.81	3.11	2.93	2.93	
Suspended	5/81	^{90}Sr	0.24±0.07	0.18±0.07	0.10±0.03	0.14±0.09	0.21±0.07	0.11±0.07
		M(II)	1.58	1.46	2.04	1.95	1.52	2.00
Suspended	3/81	^{90}Sr	0.15±0.12	0.16±0.10	0.08±0.06	0.09±0.09	0.11±0.05	
		M(II)	2.03	1.94	1.88	2.11	1.99	
Suspended	1/81	^{90}Sr	--	--	--	--	0.10±0.04	
		M(II)	--	—	--	--	3.04	
Bottom	3/81	^{90}Sr	0.32±0.04	—	0.34±0.04	—	—	

(a) Stations 1 & 2 combined

(b) Stations 3 & 4 combined

Figure 5: Dependence of ^{90}Sr Activity upon
M(II) Concentration: Station 3
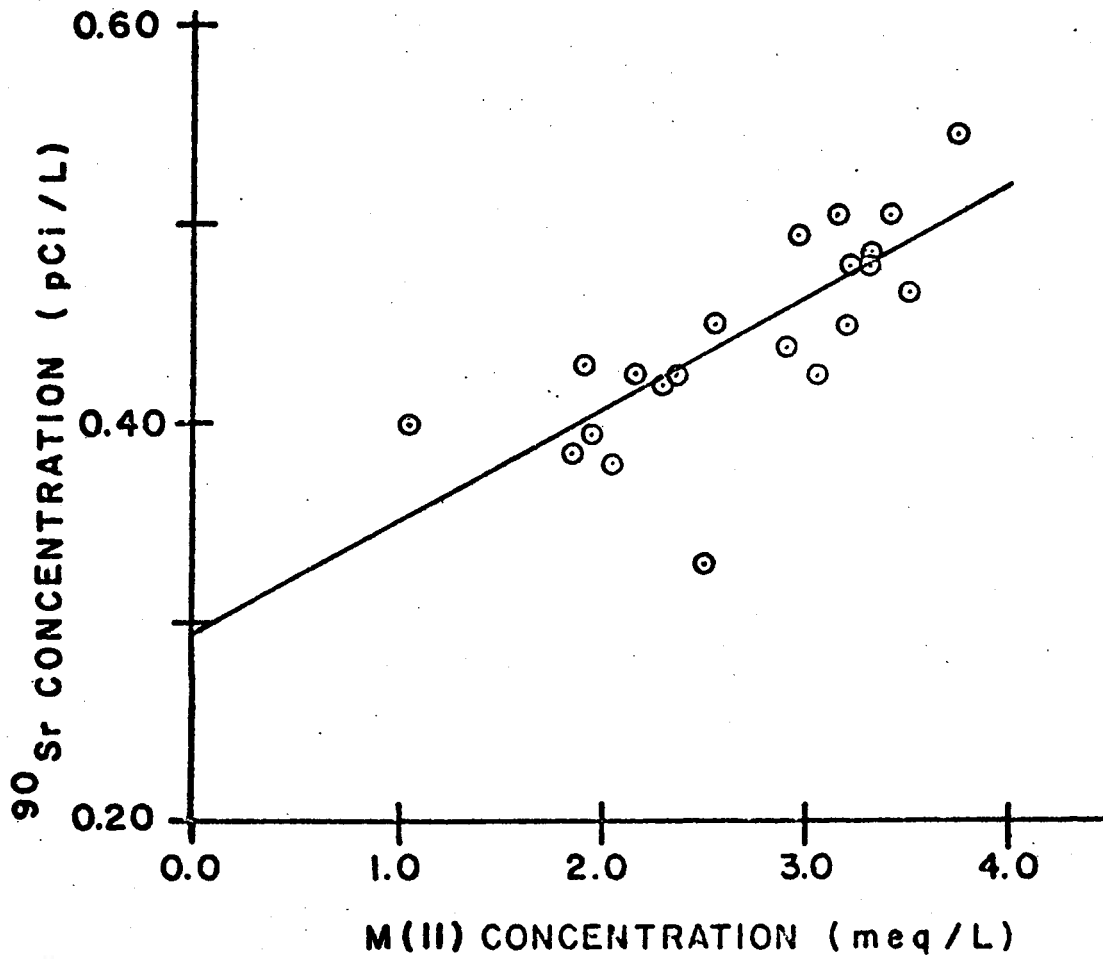
M(II).

There seems to be a minimum concentration, $\sim$ 0.1 pCi/g, below which the ^{90}Sr will not fall. This may be due to a component trapped by internal adsorption and thus not available for exchange with the aqueous phase. Studies of larger suspended sediment samples are anticipated in the future.

The parameter that is the best predictor of the ^{137}Cs is A_r; the concentration of dissolved species contributes less than 10% to the value of R^2. The best multiple regression line has the form

$$^{137}Cs = b(TDS) + cA_r \tag{6}$$

where ^{137}Cs = the concentration of ^{137}Cs in pCi/L.

TDS = total dissolved solids in g/L

Since data from both the 1976-77 and 1979-81 periods was used, A_r was calculated using equation (1) and TDS was used as the measure of the concentration of dissolved species. Statistical analysis of the data from 1979-81 indicated that M(II), Na(I) and TDS were predictors of almost equal weight. Table III is a summary of the values of the coefficients b and c, and of the contribution of each parameter to the R^2 value; the constant, a, was found to be zero.

It is not surprising that the TDS term is unimportant at Station 1 since the "fresh" aqueous ^{137}Cs is not at equilibrium with the sediment so soon after injection. It would appear that equilibrium has not been established at any of the four stations. The magnitude of the constant, c, decreases as distance from the injection point increases as does the contribution of the A_r term to the value of R^2, indicating that the ^{137}Cs is being scavenged from the water column as the water proceeds downstream.

Table III: Constants for Equation (5) and R^2 Resulting from Multiple Regression Analysis of Aqueous ^{137}Cs Concentrations.

Station	b	c	R^2(%) Due to TDS	R^2(%) Due to A_r	R^2(%) Total
1	−0.428± 0.124	0.188± 0.018	5.6	77.6	83.2
2	0.122± 0.041	0.0292± 0.0097	7.7	62.3	70.0
3	0.0842± 0.0441	0.0156± 0.0107	4.8	56.0	60.8
4	0.0	0.0552± 0.0258	0.0	40.7	40.7

It is doubtful that this removal is a surface phenomenon such as ion-exchange or adsorption. The observed D value ranges from 10^4 to 10^5 L/kg at Stations 1, 2, and 4, much higher than the value of $\sim 10^3$ (the value at Station 3) predicted from Shiao, et al. (17), an indication that there is a removal process other than surface adsorption at work. This is consistent with the findings of Duursma (18) that there is very little exchange of ^{137}Cs with stable Cs in some marine sediments. The TDS seems to be a suppressor variable at Station 1. Perhaps a higher ionic strength promotes the coagulation, precipitation, and the consequent internal adsorption of ^{137}Cs as the water slows from its turbulent flow in the outlet canal. Thus, the sediment is capable of holding more ^{137}Cs than would be possible by surface adsorption alone.

In any case, it is certain that equilibrium between ^{137}Cs in solution and in the sediment has not been reached prior to the time the water leaves the reservoir in periods of normal flow. The concentration of the newly injected nuclide remaining unprecipitated is greater than the equilibrium concentration because the time necessary to establish equilibrium between solution and sediment is significantly longer than the normal mean residence time of the water in Dardanelle Reservoir.

In the Arkansas River, with its considerable load of suspended sediment, fallout ^{90}Sr(II) has resided long enough in the water to have attained equilibrium with the suspended solid via an exchange process involving the surface of the suspended matter and a solution containing M(II) and ^{90}Sr(II). The equilibrium concentration of ^{90}Sr is quite sensitive to changes in the concentration of M(II) as long as the concentration is less than 2.5 meq/L. No other variable, such as flow

rate, suspended load, or input by fallout can account for the variations of ^{90}Sr concentrations over the period of this study. There is no doubt that the concentration of alkaline earth cations can significantly affect the efficiency of the removal (by flux to the sediment) of ^{90}Sr from the fresh water column.

No conclusion can be reached about the effect of dissolved species on the ^{137}Cs concentration because equilibrium conditions do not exist in Dardanelle Reservoir since the reactor facility is a constant source of this radionuclide. Studies of ^{137}Cs much further downstream from Arkansas Nuclear One will be necessary for a study of equilibrium conditions.

C. $\underline{^{144}Ce-^{144}Pr}$.

The concentration of ^{144}Ce-^{144}Pr is summarized in Figure 6. It will be noted that significant quantities of this pair occur usually in the first half of the year (e.g., 1977,80,81) and in the second half, the concentration is usually negligible (e.g., 1979-81). These periods of increased activity coincide with periods of refueling or extensive maintenance at ANO; the nuclide also remains for a few months after the refueling or maintenance has ceased. These activities are spread throughout the reservoir. This fact would indicate that the activity was originally airborne and is injected into the lake with runoff.

The presence of these activities in small quantities may be a good indicator of minor breaks in containment.

D. $\underline{^{141}Ce}$.

There seems to be no pattern to the appearance of this nuclide except that it appears throughout the lake, again giving the impression

Figure 6: Temporal Variation of ^{144}Ce-^{144}Pr Activity

● — Not detected.

▶ — Not determined during sampling period.

CONCENTRATION OF Ce-144 — Pr-144 (pCi/L)

MONTH/YEAR
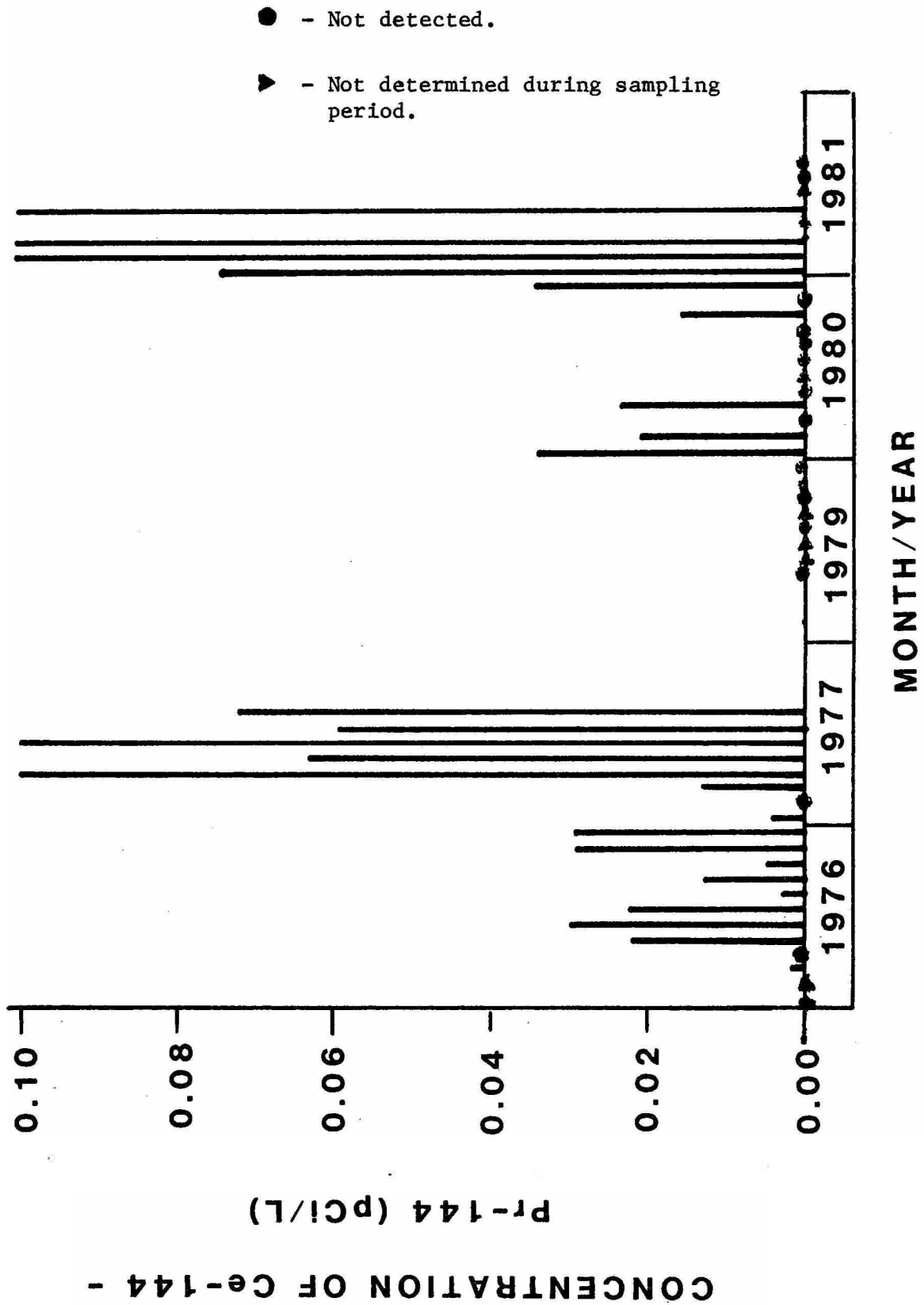
of being initially airborne. The concentrations measured during the
1979-81 period are, as a rule, higher than those found in 1976-77, a
period when the major source seemed to be fallout from the Chinese tests.
The only significant source during 1979-81 was ANO.

E. ^{89}Sr.

The only occurence of this nuclide was in April-May of 1981. No
reason for its appearance can be offered.

F. ^{58}Co.

The appearance of this nuclide is also intermittent. The appear-
ance of significant quantities of this nuclide usually accompanies
enhanced levels of ^{137}Cs.

G. Stable Species.

There is a recurring seasonal pattern of the TDS values (Figure 7).
The TDS is generally low in the winter months, rises to a maximum in
the summer months, followed by a gradual decrease in the last 4-5
months of the year. This pattern was interrupted by the drought of
late 1980.

There are long periods when there is none of the negative
correlation between TDS and flow rate which is expected of saline rivers
such as the Colorado. Season rather than flow rate seems to be the
controlling factor for TDS. This relationship is worth further study.

It can be seen from the tables in the appendix that the variations
in the Na(I) and Cl(-I) are more volatile than the other major ions.
If Na(I) and Cl(-I) in the ratio found in NaCl (1.54 mg Cl/mg Na) is
substracted out, the percentages of the other ions remain quite con-
stant at

Figure 7:   Seasonal Variation of TDS
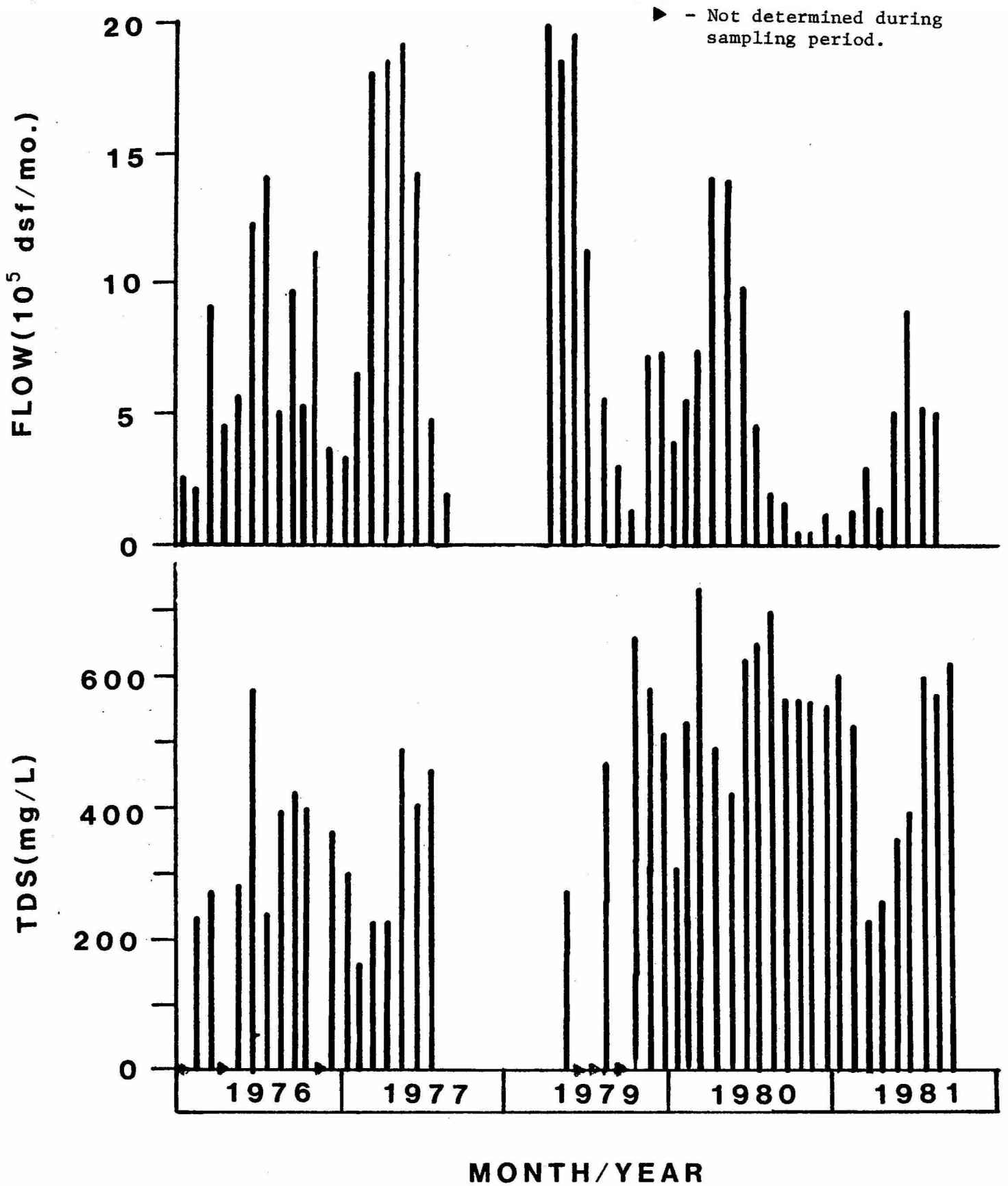
M(II) [expressed
as Ca(II)]: $19\pm2\%$ HCO_3^- : $39\pm3\%$

K(I): $1.4\pm0.1\%$ SO_4^{2-} : $19\pm1\%$

The nitrate ion remains relatively constant year round.

ACKNOWLEDGEMENTS

APPENDICES A–D

Concentrations of Radionuclides

and Stable Species

I. A Note on Errors

The estimated errors in the determination of radionuclide con-
centrations have not been included in the following tables because
of space considerations. The error in the concentration was esti-
mated by considering the standard deviation in the count rate of each
nuclide and the maximum estimated error in f_{SSA} and the error in the
activity of the standard. Together, the latter two sources of error
were found to be ±10%. Table IV summarizes the estimated error for
the nuclides in question as a function of concentration. There will
be occasional deviations from the values presented for the Ce activi-
ties when only one of the activities is present in the Ce fraction.

Table IV: Estimated Errors in the Radionuclide Concentrations

Error Range (%)	Concentration (pCi/L) of		
	^{58}Co	Cerium Activities	^{137}Cs, ^{90}Sr–^{90}Y
±50 to ±100	0.01 – 0.05	–	–
±25 to ±50	0.05 – 0.10	0.01 – 0.02	0.01 – 0.02
±15 to ±25	0.10 – 0.20	0.02 – 0.10	0.02 – 0.04
±11 to ±15	0.20 – 0.70	0.10 – 0.30	0.04 – 0.20
±10	>0.70	>0.30	>0.20

II. A Note on Standards

1. During the 1975-1977 period and early in the 1979-1981 period, the performance of the detector was checked periodically using a tritium standard as suggested by the manufacturer. Mishandling of the standard caused variations in the standard's counting rate which lead to slightly erroneous values for the concentrations of the radionuclides during the 1979-1981 period. These were included in the annual report for that fiscal year. Thus the ^{90}Sr standard became also the standard for the check of detector performance and for the calculation of the activities of the other radionuclides, and the erroneous values of the radionuclide concentrations were corrected.

2. In the summer of 1980, it was found that the distilled water supply was being contaminated through pinpoint leaks in the condenser. The potassium standards contained significant amounts of extra potassium from this source. New standards were made up from K-free water, the old standards recalibrated, and the potassium concentrations recalculated.

III. Symbols

ND   - Activity not detected.

*** - Activity not determined during that sampling period.

Appendix A -- Station 1

CONCENTRATION OF RADIONUCLIDES (PCI/L)

MONTH	GROSS BETA	PR-144-CE-144	CE-141	CO-58	CS-137	Y-90-SR-90	SR-89
579	3.9	ND	0.110	0.094	0.190	0.68	ND
879	4.9	0.014	0.036	0.060	0.072	1.10	ND
1079	9.8	ND	0.021	0.240	0.091	1.15	ND
1179	6.3	0.016	ND	0.740	0.063	1.03	ND
1279	8.4	ND	0.045	0.110	0.044	0.99	ND
180	4.2	0.053	ND	1.140	0.450	0.70	ND
280	4.2	0.022	ND	0.470	0.210	0.78	ND
380	8.4	0.029	ND	0.260	0.260	0.90	ND
480	4.6	ND	0.079	0.030	0.090	0.74	ND
580	6.7	ND	0.065	0.076	0.089	1.01	ND
680	7.8	★★★	★★★	★★★	★★★	4.91	ND
780	8.5	ND	ND	0.013	0.091	0.82	ND
880	10.1	ND	0.024	0.050	0.220	0.99	ND
980	9.5	ND	ND	0.130	0.270	1.02	ND
1080	10.0	ND	0.028	0.005	0.140	1.00	ND
1180	8.6	ND	ND	ND	0.130	0.98	ND
1280	10.5	0.067	ND	0.910	0.150	0.99	ND
181	18.8	0.170	ND	0.520	2.140	1.01	ND
201	7.8	ND	0.200	0.050	0.190	0.92	ND
381	5.8	0.180	ND	0.110	0.660	0.79	ND
481	4.5	★★★	★★★	★★★	0.160	0.76	ND
481	4.4	★★★	★★★	★★★	1.240	0.85	ND
581	4.4	0.092	0.094	ND	0.230	0.61	0.210
681	4.3	★★★	★★★	★★★	0.170	0.97	ND
781	12.6	ND	0.062	0.370	3.520	0.89	ND
881	8.0	ND	0.160	0.130	0.140	0.88	ND

CONCENTRATION OF NON-RADIOACTIVE SPECIES(MG/L)

DATE	TDS	CA+MG AS CA	NA	K	HCO3	SO4	CL	NO3
790524	227	20	***	***	***	***	***	***
790817	418	41	***	***	***	***	***	***
791019	582	56	110	***	***	***	***	***
791117	496	57	116	5.8	***	***	***	***
791220	584	47	94	5.8	***	***	***	***
800118	257	27	40	3.9	***	***	***	***
800215	313	36	53	4.5	***	***	***	***
800315	548	45	107	3.2	***	***	***	***
800411	349	33	66	2.8	***	32	***	***
800519	514	49	104	3.4	***	51	***	***
800615	587	47	145	3.7	***	57	***	***
800718	594	61	108	4.9	***	68	154	***
800815	708	67	121	5.0	138	78	173	***
800919	624	69	103	5.0	134	66	143	***
801017	594	68	93	4.8	136	61	120	7
801122	534	65	104	5.3	137	70	113	8
801218	558	66	88	4.8	141	67	119	9
810116	586	63	94	4.7	138	70	111	9
810220	530	60	78	4.2	122	63	106	8
810321	248	41	44	4.2	82	47	61	8
810411	218	29	***	***	***	28	***	***
810424	232	27	27	2.6	57	32	41	8
810520	258	31	34	3.1	63	32	38	7
810619	259	29	41	3.0	***	33	71	8
810718	591	58	137	5.1	103	63	204	9
810828	548	58	103	5.1	112	58	154	9
810918	660	65	116	5.3	123	68	172	8

Appendix B -- Station 2

CONCENTRATION OF RADIONUCLIDES (PCI/L)

MONTH	GROSS BETA	PR-144-CE-144	CE-141	CO-58	CS-137	Y-90-SR-90	SR-89
579	3.3	ND	0.082	ND	0.057	0.63	ND
879	4.5	ND	0.110	0.003	0.030	0.96	NU
1079	8.8	ND	0.053	0.049	0.049	0.99	NU
1179	5.7	ND	0.026	0.010	0.034	0.96	ND
1279	11.2	ND	0.064	0.041	0.051	0.87	NU
180	3.5	0.034	ND	0.019	0.055	0.58	ND
280	4.9	0.021	ND	0.110	0.047	0.73	ND
380	5.6	ND	0.030	0.130	0.061	0.76	ND
480	4.6	0.023	ND	0.067	0.084	0.70	ND
580	7.3	ND	0.068	0.016	0.070	0.89	ND
680	6.1	★★★	★★★	★★★	★★★	2.68	ND
780	8.5	ND	0.022	0.050	0.087	0.84	ND
880	9.0	ND	ND	0.062	0.130	0.99	ND
980	8.4	ND	0.016	0.059	0.110	1.03	ND
1080	8.0	0.015	ND	0.045	0.095	0.98	NU
1180	7.3	ND	0.047	0.120	0.110	1.02	ND
1280	7.7	0.034	ND	0.029	0.170	1.00	NU
181	6.5	0.073	ND	0.033	0.140	0.90	ND
281	8.3	0.110	ND	0.056	0.094	0.87	ND
381	5.4	0.120	0.190	0.050	0.150	0.77	ND
481	5.3	★★★	★★★	★★★	0.140	0.84	NU
581	3.2	0.073	0.082	0.033	0.120	0.48	0.420
681	4.0	★★★	★★★	★★★	0.110	1.23	ND
781	9.0	ND	0.026	0.067	0.130	0.84	ND
881	6.1	ND	0.320	0.059	0.310	0.77	ND

CONCENTRATION OF NON-RADIOACTIVE SPECIES(MG/L)

DATE	TDS	CA+MG AS CA	NA	K	HCO3	SO4	CL	NO3
790524	149	16	***	***	***	***	***	***
790817	413	41	50	***	***	***	***	***
791019	605	60	91	***	***	***	***	***
791117	508	58	114	5.8	***	***	***	***
791220	663	47	94	5.8	***	***	***	***
800118	244	27	37	3.7	***	***	***	***
800215	400	40	112	4.7	***	***	***	***
800315	530	53	101	2.6	***	***	***	***
800411	350	33	66	2.8	***	32	***	***
800519	454	47	98	3.3	***	48	***	***
800615	540	48	132	3.5	***	52	***	***
800718	585	61	104	4.9	***	67	149	***
800815	702	66	120	4.9	142	79	171	***
800919	616	68	105	5.0	134	68	142	***
801017	549	68	95	4.6	135	64	130	6
801122	583	64	96	4.7	137	68	112	8
801218	565	64	93	4.8	138	66	124	9
810116	592	62	93	4.7	140	73	109	9
810220	539	60	68	4.5	122	63	104	8
810321	237	39	44	4.1	85	43	64	8
810424	228	27	24	2.5	57	32	39	8
810520	244	29	34	2.8	60	32	34	6
810619	235	27	38	2.9	***	33	64	8
810718	590	56	129	5.1	101	63	207	8
810828	542	57	100	5.1	113	56	153	9
810918	625	64	110	5.3	124	67	169	8

Appendix C -- Station 3

CONCENTRATION OF RADIONUCLIDES (PCI/L)

MONTH	GROSS BETA	PR-144- CE-144	CE-141	CO-58	CS-137	Y-90- SR-90	SR-89
579	4.5	0.015	0.170	0.270	0.048	0.80	ND
879	6.0	ND	0.035	ND	0.037	0.99	ND
1079	12.0	ND	0.056	ND	0.051	1.01	ND
1179	6.6	ND	0.059	0.043	0.038	0.85	ND
1279	7.1	ND	0.044	0.410	0.019	0.84	ND
180	4.3	0.027	ND	0.015	0.048	0.79	ND
280	8.0	0.038	ND	0.260	0.050	0.90	ND
380	7.5	ND	0.074	0.190	0.025	0.90	ND
480	7.0	ND	0.110	0.220	0.015	0.66	ND
580	6.4	ND	0.065	0.170	0.064	0.87	ND
680	8.3	★★★	★★★	★★★	★★★	2.20	ND
780	9.9	ND	ND	0.090	0.071	1.09	ND
880	9.9	0.013	ND	0.084	0.087	0.93	ND
980	7.5	ND	ND	0.004	0.051	0.97	ND
1080	8.8	ND	ND	ND	0.063	1.01	ND
1180	7.4	ND	0.056	0.028	0.081	0.96	ND
1280	7.9	0.017	ND	0.056	0.073	0.96	ND
181	7.2	0.044	ND	0.045	0.270	0.88	ND
281	5.9	0.060	0.110	0.056	0.044	0.85	ND
381	5.6	0.130	0.400	0.033	0.067	0.71	0.590
481	5.3	★★★	★★★	★★★	0.092	0.74	0.570
581	5.7	0.250	ND	ND	0.046	1.05	0.510
681	5.8	★★★	★★★	★★★	0.071	1.11	ND
781	9.0	ND	0.098	ND	0.049	0.85	ND
881	6.4	ND	0.130	0.190	0.063	0.89	ND

CONCENTRATION OF NON-RADIOACTIVE SPECIES(MG/L)

DATE	TDS	CA+MG AS CA	NA	K	HCO3	SO4	CL	NO3
790524	268	21	***	***	***	***	***	***
790817	516	59	65	***	***	***	***	***
791019	710	63	123	***	***	***	***	***
791117	477	61	109	5.8	***	***	***	***
791220	501	46	80	5.5	***	***	***	***
800118	316	39	44	4.5	***	***	***	***
800215	600	51	112	3.4	***	***	***	***
800315	671	64	162	3.5	***	***	***	***
800411	506	50	93	3.6	***	52	***	***
800519	464	43	96	3.5	***	47	***	***
800615	669	57	162	4.2	***	66	***	***
800718	739	75	127	5.1	***	82	199	***
800815	628	70	104	5.0	144	67	148	***
800919	547	66	89	4.9	138	63	114	***
801017	569	68	92	5.2	138	62	126	6
801122	577	66	104	5.4	142	67	111	8
801218	542	64	84	4.8	141	64	119	7
810116	539	58	83	4.5	132	63	103	10
810220	402	47	56	3.7	99	50	80	8
810320	247	38	38	4.1	81	45	60	11
810424	286	37	32	3.4	74	39	52	7
810520	379	41	55	4.1	80	44	58	8
810619	440	43	93	4.3	***	51	152	10
810718	600	62	121	5.5	120	66	180	8
810828	584	65	119	5.1	127	65	176	10
810918	663	65	100	5.3	130	66	160	7

Appendix D — Station 4

CONCENTRATION OF RADIONUCLIDES (PCI/L)

MONTH	GROSS BETA	PR-144-CE-144	CE-141	CO-58	CS-137	Y-90-SR-90	SR-89
579	5.1	0.021	0.150	ND	0.051	1.09	ND
879	5.6	0.005	0.120	ND	0.030	0.98	ND
1079	9.6	ND	0.190	0.500	0.053	0.98	ND
1179	9.0	ND	0.110	0.053	0.031	1.28	ND
1279	7.1	ND	0.045	0.240	0.033	0.79	ND
180	4.9	0.047	ND	ND	0.043	0.69	ND
280	6.2	ND	0.120	0.170	0.043	0.98	ND
380	7.3	ND	0.210	0.140	0.057	0.87	ND
480	6.2	ND	0.090	0.078	0.064	0.91	ND
580	7.0	ND	0.120	0.200	0.049	0.98	ND
680	8.4	***	***	***	***	1.84	ND
780	8.1	ND	0.042	0.067	0.099	0.97	ND
880	8.9	ND	0.085	0.038	0.120	0.95	ND
980	9.7	ND	ND	0.019	0.110	0.97	ND
1080	8.0	0.029	ND	0.070	0.110	0.94	ND
1180	8.3	ND	0.059	0.067	0.140	0.99	ND
1280	8.0	ND	0.044	0.052	0.190	1.01	ND
181	8.4	0.047	ND	ND	1.000	0.92	ND
281	8.2	0.093	0.120	ND	0.110	0.86	ND
381	5.6	0.320	ND	0.110	0.230	0.71	ND
481	4.6	***	***	***	0.280	0.91	ND
581	5.2	0.180	ND	0.060	0.140	0.76	0.330
681	5.4	***	***	***	0.150	0.96	ND
781	7.5	0.054	ND	0.038	0.120	0.87	ND
881	6.6	ND	0.280	0.037	0.063	0.79	ND

CONCENTRATION OF NON-RADIOACTIVE SPECIES(MG/L)

DATE	TDS	CA+MG AS CA	NA	K	HCO3	SO4	CL	NO3
790524	273	44	***	***	***	***	***	***
790817	470	56	56	***	***	***	***	***
791019	650	58	116	***	***	***	***	***
791117	584	62	140	6.0	***	***	***	***
791220	512	47	82	5.6	***	***	***	***
800118	326	36	46	4.5	***	***	***	***
800215	525	50	88	4.3	***	***	***	***
800315	723	58	180	3.9	***	60	***	***
800411	491	49	86	3.5	***	47	***	***
800519	422	40	90	3.3	***	45	***	***
800615	626	56	161	4.2	***	60	***	***
800718	648	67	116	4.9	***	73	175	***
800815	688	71	117	5.0	142	77	167	***
800919	565	69	99	5.1	136	67	140	***
801017	562	68	90	4.8	137	67	129	5
801122	563	66	98	5.4	138	68	113	8
801218	546	67	92	5.1	141	69	125	9
810116	594	61	93	4.7	140	73	109	10
810220	528	60	68	4.2	125	62	101	8
810320	234	42	42	4.0	65	46	65	11
810424	259	31	29	3.0	65	34	47	9
810520	347	39	49	3.8	80	46	56	9
810619	389	40	77	4.0	***	47	135	9
810718	598	59	125	5.6	110	63	192	9
810828	571	62	114	5.2	122	63	171	10
810918	621	64	108	5.2	126	70	167	9

REFERENCES

(1) Chittenden, D. M., II. "Radionuclide Concentrations in the Arkansas River Upstream and Downstream from the Nuclear I Power Generating Facility"; NTIS, Report PB-281 808/6GA, 1978.

(2) Noshkin, V. E.; Bowen, V. T. In "Radioactive Contamination of the Marine Environment"; *IAEA*; Vienna, Austria, 1973; pp 671-686.

(3) Evans, D. W.; Cutshall, N. H. ibid; pp 125-140.

(4) Johnson, V.; Cutshall, N.; Osterberg, C. *Water Resources Res.* 1967, 3,99.

(5) Lerman, A.; Lietzke, T. A. *Limnol. Oceanogr.* 1975, 20, 497.

(6) Lerman, A.; Taniguchi, H. Proc. 3rd Natl. Symp. Radioecol. NTIS, Alexandria, Va., CONF-710501-P1, 1971; pp 460-464.

(7) Alberts, James J.; Wahlgren, Morris A. *Environ. Sci. Technol.* 1981, 15, 94.

(8) Chittenden, D. M., II. *Proc. Ark. Acad. Sci.* 1980, 34, 104.

(9) Glendennin, L. E.; Flynn, K. F.; Buchanan, R. F.; Steinberg, E. D. *Anal. Chem.* 1955, 27, 59.

(10) Burgus, W. W. In "The Radiochemistry of Cobalt"; Bates, L. C. and Leddicott, G. W., editors; National Academy of Science-National Research Council, Washington, D.C., NAS-NS 3041, 1961.

(11) Kahn, Berne; Smith, David K.; Staub, Conrad. *Anal. Chem.* 1957, 29, 1210.

(12) Hodges, H. L. Annual Progress Report to U. S. Atomic Energy Commission for Contract At-(40-1)-2529, 1963; p 46.

(13) Nervik, W. E.; Stevenson, P. C. *Nucleonics* 1952, 10, 18.

(14) "Water Analysis Handbook," Hach Chemical Company, 1973,1974.

(15) "Standard Methods for the Examination of Water and Wastewater,"

      15th ed.; APHA, AWWA, and WPCF, Washington, D.C., 1980;

      pp 195-199, 211-222, 231-234, 253-257, 270-271.

(16) Copyright, Pennsylvania State University, Waisman Research

      Computing Facility, 1980.

(17) Shiao, S.-Y.; Rafferty, P.; Meyer, R. E.; Rogers, W. J.

      "Radioactive Waste in Geologic Storage"; Fried, Sherman, Ed.;

      American Chemical Society: Washington, D.C., Symposium Series

      100, 1979; pp 297-324.

(18) Duursma, E. K.   In "Radioactive Contamination of the Marine

      Environment"; pp 57-71.