NUREG/CR-3940

Field Experiment Determinations of Distribution Coefficients of Actinide Elements in Alkaline Lake Environments

Prepared by H. J. Simpson, R. M. Trier, Y.-H. Li, R. F. Anderson, A. L. Herczeg

Lamont-Doherty Geological Observatory
Columbia University

Prepared for
U.S. Nuclear Regulatory
Commission

NOTICE

This report was prepared as an account of work sponsored by an agency of the United States Government. Neither the United States Government nor any agency thereof, or any of their employees, makes any warranty, expressed or implied, or assumes any legal liability of responsibility for any third party's use, or the results of such use, of any information, apparatus, product or process disclosed in this report, or represents that its use by such third party would not infringe privately owned rights.

NOTICE

Availability of Reference Materials Cited in NRC Publications

Most documents cited in NRC publications will be available from one of the following sources:

1. The NRC Public Document Room, 1717 H Street, N.W.
   Washington, DC 20555
2. The NRC/GPO Sales Program, U.S. Nuclear Regulatory Commission,
   Washington, DC 20555
3. The National Technical Information Service, Springfield, VA 22161

Although the listing that follows represents the majority of documents cited in NRC publications, it is not intended to be exhaustive.

Referenced documents available for inspection and copying for a fee from the NRC Public Document Room include NRC correspondence and internal NRC memoranda; NRC Office of Inspection and Enforcement bulletins, circulars, information notices, inspection and investigation notices; Licensee Event Reports; vendor reports and correspondence; Commission papers; and applicant and licensee documents and correspondence.

The following documents in the NUREG series are available for purchase from the NRC/GPO Sales Program: formal NRC staff and contractor reports, NRC-sponsored conference proceedings, and NRC booklets and brochures. Also available are Regulatory Guides, NRC regulations in the *Code of Federal Regulations*, and *Nuclear Regulatory Commission Issuances*.

Documents available from the National Technical Information Service include NUREG series reports and technical reports prepared by other federal agencies and reports prepared by the Atomic Energy Commission, forerunner agency to the Nuclear Regulatory Commission.

Documents available from public and special technical libraries include all open literature items, such as books, journal and periodical articles, and transactions. *Federal Register* notices, federal and state legislation, and congressional reports can usually be obtained from these libraries.

Documents such as theses, dissertations, foreign reports and translations, and non-NRC conference proceedings are available for purchase from the organization sponsoring the publication cited.

Single copies of NRC draft reports are available free, to the extent of supply, upon written request to the Division of Technical Information and Document Control, U.S. Nuclear Regulatory Commission, Washington, DC 20555.

Copies of industry codes and standards used in a substantive manner in the NRC regulatory process are maintained at the NRC Library, 7920 Norfolk Avenue, Bethesda, Maryland, and are available there for reference use by the public. Codes and standards are usually copyrighted and may be purchased from the originating organization or, if they are American National Standards, from the American National Standards Institute, 1430 Broadway, New York, NY 10018.

GPO Printed copy price: $5.00

NUREG/CR-3940
RW

Field Experiment Determinations of Distribution Coefficients of Actinide Elements in Alkaline Lake Environments

Manuscript Completed: March 1983
Date Published: August 1984

Prepared by
H. J. Simpson, R. M. Trier, Y.-H. Li, R. F. Anderson, A. L. Herczeg

Lamont-Doherty Geological Observatory
Columbia University
Palisades, NY 10964

Prepared for
Division of Radiation Programs and Earth Sciences
Office of Nuclear Regulatory Research
U.S. Nuclear Regulatory Commission
Washington, D.C. 20555
NRC FIN B7057

ABSTRACT

The extent to which stored radionuclides and their daughter products might be released by, and subsequently transported away from, a high-level nuclear waste repository is an essential consideration in choosing locations for storage sites. Predictions of the distances that a given radionuclide may travel over a given period of time are dependent on many factors, including numerical values assigned for solubilities and/or sorption-desorption particle affinities (K_d's). Numerical values of radionuclide distribution coefficients have considerable uncertainty due in part to the necessity of extrapolating from small scale laboratory experiments. Data from natural systems can provide important constraints on radionuclide transport calculations in the vicinity of HLW repositories because the temporal and spatial scales are generally more similar to a waste repository environment than beaker scale experiments.

Measurements of the radioisotope concentrations of a number of elements (Am, Pu, U, Pa, Th, Ac, Ra, Po, Pb, Cs, and Sr) in the water and sediments of a group of alkaline (pH = 9-10), saline lakes demonstrate greatly enhanced soluble-phase concentrations of elements with oxidation states of (III)-(VI) as the result of complexing by carbonate ion. Ratios of soluble radionuclide concentrations in Mono Lake to those in seawater ($[CO_3^{2-}]$ in Mono Lake = 200 times that of seawater) were: Pu(≈10), ^{238}U(≈150), ^{231}Pa, ^{228}Th, ^{230}Th(≈10^3), and ^{232}Th(≈10^5). Effective distribution coefficients of these radionuclides in high CO_3^{2-} environments are several orders of magnitude lower (i.e., less particle reactive) than in most other natural waters. The importance of CO_3^{2-} ion on effective K_d values was also strongly suggested by laboratory experiments in which most of the dissolved actinide elements became adsorbed to particles after a water sample normally at a pH of 10 was acidified, stripped of all CO_2, and then returned to pH 10 by adding NH_4OH. Furthermore, the effect of complexation by organic ligands is of secondary importance in the presence of appreciable carbonate ion concentration.

Neither pure phase solubility calculations nor laboratory scale K_d determinations accurately predicted the measured natural system concentrations. Therefore, measurements of the distribution of radionuclides in natural systems are essential for assessment of the likely fate of potential releases from high level waste repositories to groundwater.

TABLE OF CONTENTS

LIST OF TABLES

List of Tables (Cont.)

List of Tables (Cont.)

LIST OF FIGURES

List of Figures (Cont.)

1.0 INTRODUCTION

One of the most important factors governing the transport of radionuclides in aqueous environments is partitioning between aqueous phases and geological materials such as detrital particles. The propensity of a particular radionuclide to associate with solid phases, often expressed in terms of an effective distribution coefficient, K_d, has considerable influence on its transport rates and pathways in aquatic systems. Low values of K_d (i.e. $< 10^1$), defined here as the ratio of the Activity of Nuclide R per gram of dry solid material to the Activity of Nuclide R per ml of solution (A_R per gram solid/A_R per ml solution), imply behavior of radionuclides predominantly as soluble components with relatively little retardation of either diffusive or advective aqueous phase transport. In contrast, high values of K_d (i.e. 10^4-10^6) indicate substantial association with solid materials, and net aqueous phase transport by diffusion and advection at rates orders of magnitude slower than for low K_d radionuclides. Because the chemical composition of aqueous solutions exert a profound effect on K_d, a single non-parameterized distribution coefficient assigned for a given radionuclide is clearly not sufficient to estimate the likely mobility of an element in all natural systems of interest.

1.1 Issues addressed by this research

Some of the primary issues concerning the migration of radionuclides in the far field that we are examining include:

To what extent will radionuclides be complexed by natural inorganic and organic ligands?

How do numerical values of effective distribution coefficients of radionuclides in real natural water systems compare with those derived from small scale experiments?

How do numerical values of effective distribution coefficients of radionuclides in natural waters vary depending upon the oxidation state of a particular nuclide?

Which inorganic ions are most important in forming anion complexes with radionuclides in natural waters?

1.2 Background and Objectives

One method to help establish representative radionuclide distribution coefficients is through laboratory experiments using small amounts of solid and aqueous phases. The compositions of both phases can be readily varied in such experiments, and "end-members" such as specific clay minerals or manganese oxides and reagent NaCl solutions buffered at various pH levels can be used, as well as natural solid phases and solutions from representative environmental systems (Duursma and Bosch, 1970). Measurements of distribution coefficients in such experiments can be made rapidly and with considerable precision for a single equilibration by employing tracer activity levels of one or more radionuclides analyzed by gamma spectrometry. These types of experiments can provide valuable insights into processes which are likely to be important in regulating effective distribution coefficients, but they are not sufficient by themselves to establish representative radionuclide behavior in real aqueous systems. Laboratory experiments restrict both the time and spatial scales involved in radionuclide behavior, and may inadvertently exclude some of the processes which are important in the situations of most relevance to management of radionuclides. In addition, laboratory experiments yield numerical values of distribution coefficients with a substantial range (up to several orders of magnitude) for a single radionuclide, depending on factors such as (1) whether the experiment was conducted to yield a sorption or a desorption coefficient, (2) the total dissolved solids was typical of fresh water or much higher as for seawater, (3) the solid phases had appreciable sand components which can introduce large variations because of the very low sorption capacity of large grains of quartz and other sand grains, or (4) the pH of the solution phase is near the low end of natural solution conditions (≈ pH 4) or toward the high end (≈ pH 9-10). Extended discussion of these and a number of other factors which influence laboratory K_d measurements can be found in a NRC summary report by Schell and Sibley (1982) and previous reports by these and other authors.

The data reported here consist primarily of measurements of natural and anthropogenic (fallout) radionuclides in the water and sediments of a group of natural lakes. These lakes can be viewed as a set of large-scale natural experiments from which the effective partitioning of radionuclides between water and solid phases can be observed. The time-scales of equilibration and spacial scales represented by these natural lakes are much greater than is feasible to employ in laboratory distribution coefficient measurements, and thus can provide valuable constraints on extrapolations of results from these latter experiments to the real systems of primary interest in waste management.

1.3 Scope of Research

Our efforts are directed towards obtaining data on radionuclide behavior in natural water systems which can provide critical information for testing of nuclide concentrations computed from thermodynamic models and small scale laboratory experiments. The systems we have studied initially provide information on radionuclide complexing by carbonate ions, which appear to be of considerable importance in enhancing the potential for nuclide transport. Field and laboratory work in progress will provide information on the effects of sulfate and chloride ions on radionuclide mobility.

2.0 METHODS

The systems chosen for sampling are fairly large (most have surface areas of more than one hundred square kilometers), closed-basin, alkaline lakes located (Figure 1) in the western Great Basin physiographic province of the United States (California, Oregon and Nevada). This area is

Western U.S. Alkaline, Saline Lakes

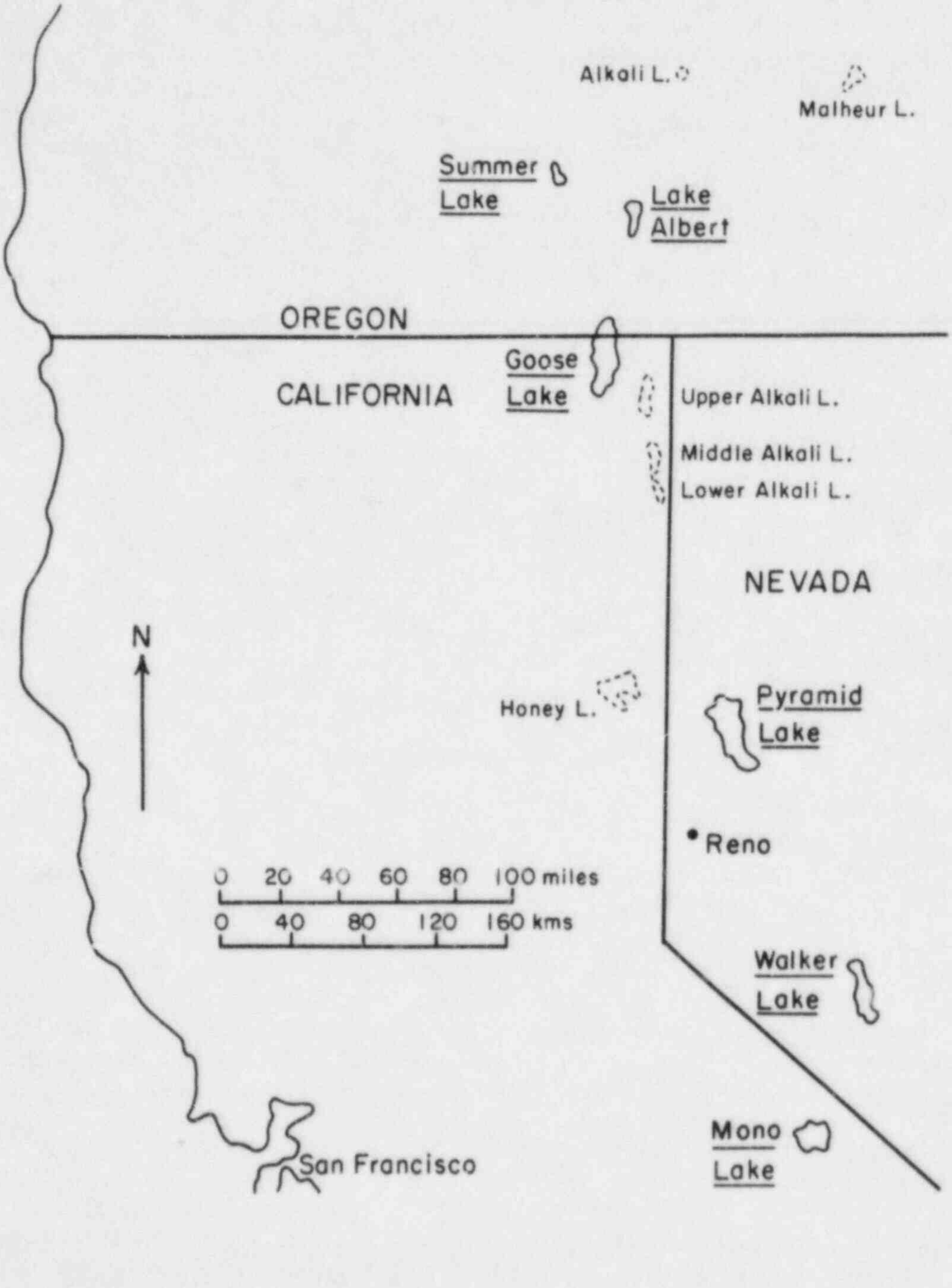


Figure 1.

characterized by a series of structural basins with no exterior drainage to the ocean. Many of these interior drainage basins contain persistent saline lakes or playas which support ephemeral lakes during unusually wet periods. Some of these lakes, such as Great Salt Lake in Utah, have relative proportions of major ions similar to seawater, with chloride accounting for a large fraction (more than 90%) of the toal anionic balance and inorganic carbon as bicarbonate and carbonate ions being minor constituents (less than 1% of total anion equivalents). Another group, which derive most of their supply of dissolved ions from weathering of silicate minerals followed by extensive evaporation of the resultant fresh water streams and springs to produce alkaline lakes (Hutchinson, 1957; Broecker and Walton, 1959; Jones, 1966; Garrels and MacKenzie, 1967; Simpson, 1970), have relative proportions of major ions quite different from seawater. This latter group, some of which have carbonate ion concentrations of more than 10^3 greater than in seawater, provides an ideal natural environment for establishing the effect of carbonate complexing on radionuclide mobilities.

We concentrated ^{239,240}Pu, ^{238}U, ^{232}Th, ^{230}Th, ^{228}Th, ^{231}Pa, ^{210}Po, and ^{210}Pb from large samples in the field (80 to 240 liters) after filtration through glass fiber filters, by coprecipitation with $Fe(OH)_3$. The hydroxide precipitates were returned to the laboratory, and analyzed by chemical preparation and alpha spectrometry procedures frequently used in the field of chemical oceanography (Ku, 1968; Wong, 1971; Kaufman et al., 1973). Measurements of ^{238}U, ^{234}U and ^{234}Th were made on small samples (1-10 liters) after filtration, laboratory chemical preparation and alpha (^{238}U, ^{234}U) or beta (^{234}Th) counting. We measured ^{226}Ra by scintillation counting of its gaseous daughter product, ^{222}Rn. Cesium-137 was determined by γ-counting of an exchange resin used to remove cesium from large samples (40-240 liters) and ^{90}Sr was determined by β-counting. Sediment samples were collected by coring, and analyzed at many depth intervals for the same nuclides discussed above.

3.0 FIELD SAMPLING LOCATIONS

The Great Basin of the western U.S. is characterized by a series of structural basins with no exterior drainage to the ocean. Many of these interior drainage basins contain persistent saline lakes or playas which support ephemeral lakes during unusually wet periods. During the last glacial period a number of the Great Basin lakes were much larger, due to greater precipitation and/or lower evaporation rates, and prominent fossil shoreline features of these pluvial lakes have been studied extensively as indicators of past climatic conditions (Russell, 1885; Russell, 1889; Broecker, 1957; Hutchinson, 1957). Some of the Great Basin saline lakes, such as Great Salt Lake in Utah, have relative proportions of major ions similar to seawater, with chloride accounting for a large fraction (more than 90%) of the total anionic balance and inorganic carbon as bicarbonate and carbonate ions being minor consituents (less than 1% of total anion equivalents). Some of these high chloride lakes derive major portions of their present dissolved ion influx from saline streams which drain areas characterized by weathering of evaporite minerals such as halite, but all of the saline lakes with high proportions of bicarbonate and carbonate derive most of their supply of dissolved ions from weathering of silicate minerals followed by extensive evaporation of the resultant fresh water streams and springs to produce alkaline lakes (Hutchinson, 1957; Broecker and Walton, 1959; Jones, 1966; Garrels and Mackenzie, 1967; Simpson, 1970). The alkaline lake for which the most chemical and geological data is available is probably Mono Lake (i.e. Mason, 1967; Scholl et al., 1967; Christensen et al., 1969; plus other references cited above).

Physical characteristics of six alkaline lakes discussed here, Mono L., L. Abert, Walker L., Pyramid L., Goose L. and Summer L. are compiled in Table 1. Our analytical results are most extensive for the first four of these lakes. Goose Lake at the time of our sampling (1981) was quite shallow and turbid, which caused considerable difficulty in obtaining particle-free samples of sufficient size to measure some of the dissolved radionuclides. Summer Lake had the same problems, only to a greater extreme. We obtained small samples of that lake by walking across soft exposed sediments for more than a mile before reaching water of a few centimeters depth. The other four lakes were sufficiently deep and accessible to allow us to obtain large volume samples. All of these lakes are currently declining in surface elevation and volume over the time scale of decades due to diversions of tributaries for irrigation water or domestic supplies. The largest ($\approx$ 20 km^3) and deepest (maximum depth $\approx$ 100 meters) of the lakes we sampled was Pyramid Lake in Nevada, which lies entirely within an Indian reservation. The smallest of the more accessible lakes was Lake Abert, with a volume in 1981 of 0.1 to 0.2 km^3 and a surface area of about 100 km^2.

Sediments from each of the six lakes were obtained by coring, either with a gravity corer, or by pushing plastic tubing into the sediments of the two very shallow lakes. Locations of the sediment cores and bottom depth contours are shown in Figures 2-7.

4.0 DISSOLVED RADIONUCLIDE CONCENTRATIONS IN ALKALINE LAKES

Concentrations of dissolved fallout plutonium in Mono Lake measured at LDGO are listed in Table 2. These large volume samples ranged in size from 80 to 240 liters, with the exception of one sample obtained in 1979 by equilibrating a large dialysis bag filled with "artificial" Mono Lake water (14 liters) free of radionuclides with surface water from the lake. The activity of ^{239,240}Pu in water inside the dialysis bag after three days equilibration was the same as for a fresh sample of water collected from the lake. Most of the 33 values obtained for ^{239,240}Pu were between 10 and 20 pCi/m^3. Samples collected prior to 1980 were unfiltered, while those collected in 1980 and 1981 were passed through large diameter glass fiber filters, unless denoted "(U)", indicating an unfiltered sample. No systematic differences between filtered and unfiltered samples were observed for dissolved plutonium in Mono Lake. Deep water samples obtained from water which is free of oxygen (below 13 meters) for about half of each year had the same plutonium concentrations as samples from oxygenated water.

Table 1

Alkaline Lake Physical Parameters

Lake	Surface Elevation(m)	Surface Area(km^2)	Mean Depth (m)	Maximum Depth(m)	Volume (km^3)	Lake Annual Precip. (cm)	Drainage Basin Annual Precip(cm)	Drainage Basin Area (km^2)	Annual Stream Input (km^3)
Mono L.-'64[a]	1948	200	19	52	3.8	~20	43	1650	~0.10
Mono L.-'78	1940	160	17	44	2.7	~20	43	1650	~0.03
L. Abert-'60[b]	1300	150	2	5	0.35	~25	30	2230	~0.07
L. Abert-'81	1299	120	1.1	4	0.13	~25	30	2230	~0.06
Walker L.-'68[c]	1210	165	24	36	4.75	15	23	9600	~0.13
Walker L.-'81	1203	132	~17	~28	2.65	15	23	9600	~0.08
Pyramid L.-'68[d]	1161	460	61	109	28	15	35	7140	~0.31
Pyramid L.-'81	1149	420	52	97	22	15	35	7140	~0.16
Goose L.-'62[b]	1432	338	0.9	2	0.36	35	35	2950	~0.20
Goose L.-'81est	1431	273	0.6	1.3	0.17	35	35	2950	~0.11
Summer L.-'63[b]	1264	100	<1	2	0.03	28	32	1010	~0.08
Summer L.-'81est	1263	~60	<0.5	<1	<0.02	28	32	1010	~0.05

a) Mason, 1967

b) Phillips and VanDenburgh, 1971

c) Rush, 1974

d) VanDenburgh et al, 1973

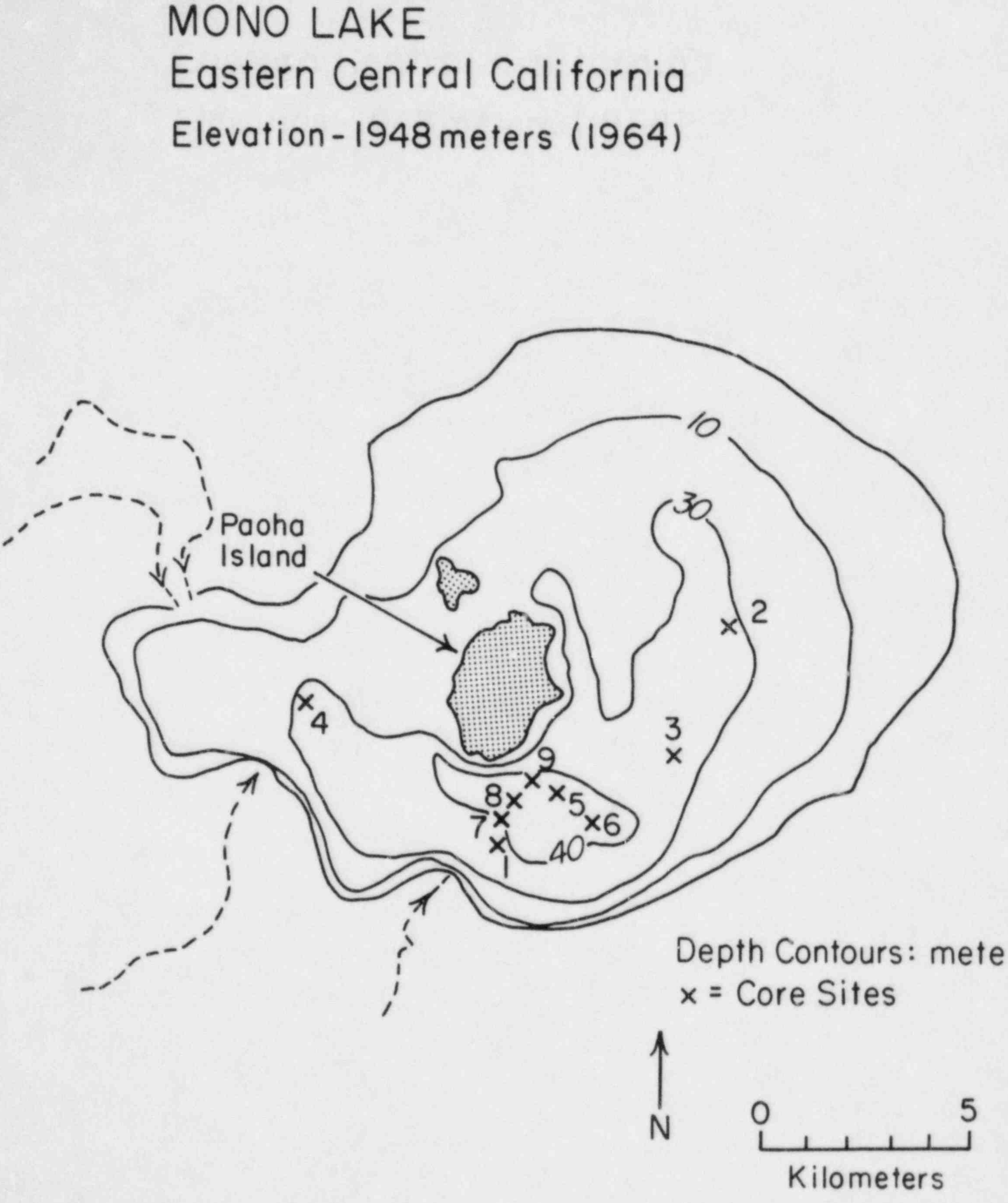

MONO LAKE
Eastern Central California
Elevation - 1948 meters (1964)
Paoha Island
10
30
40
1
2
3
4
5
6
7
8
9
Depth Contours: mete
x = Core Sites
N
0
5
Kilometers

Figure 2.

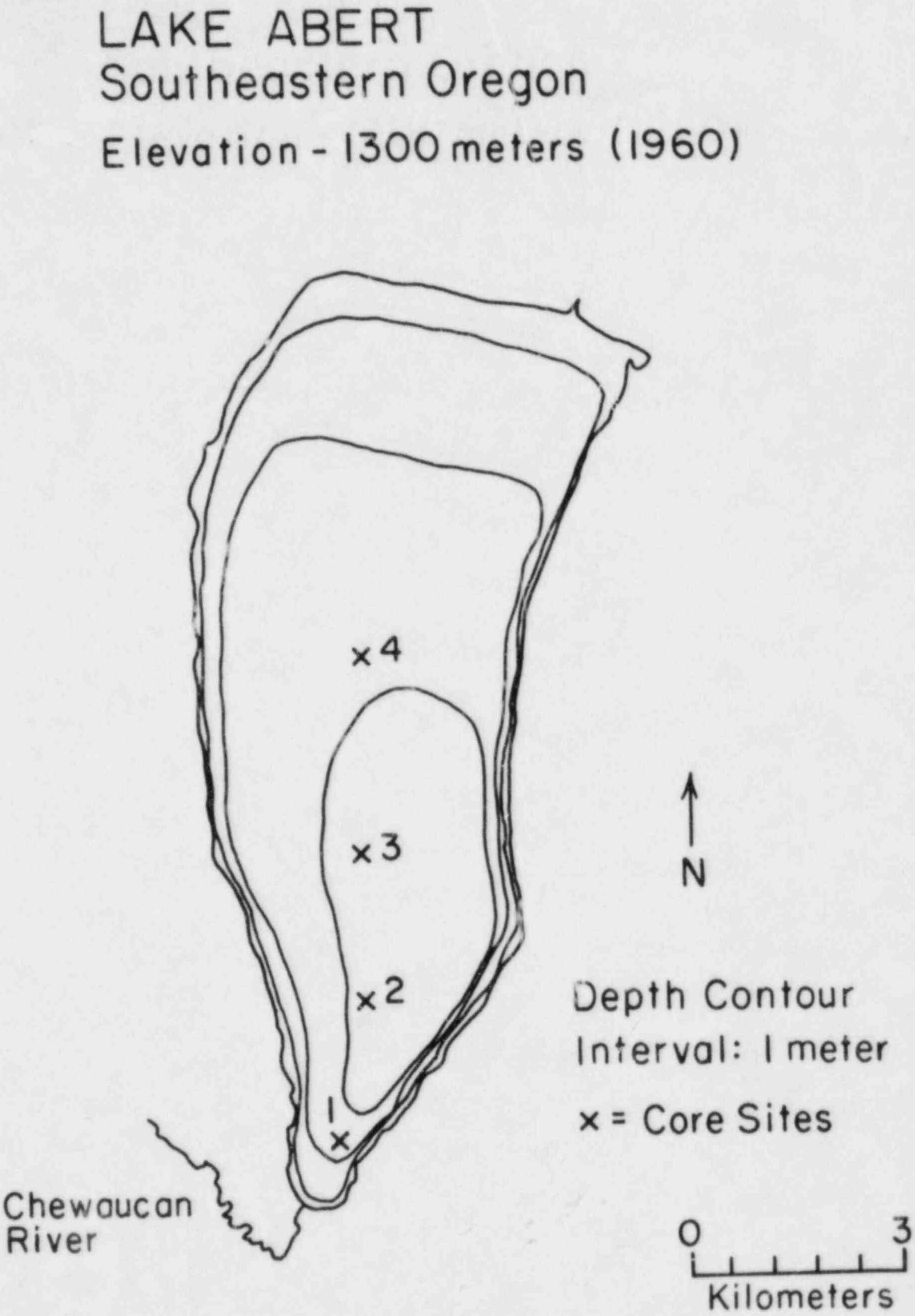

LAKE ABERT
Southeastern Oregon
Elevation - 1300 meters (1960)
x4
x3
x2
1
x
N
Depth Contour
Interval: 1 meter
x = Core Sites
Chewaucan
River
0
3
Kilometers

Figure 3.

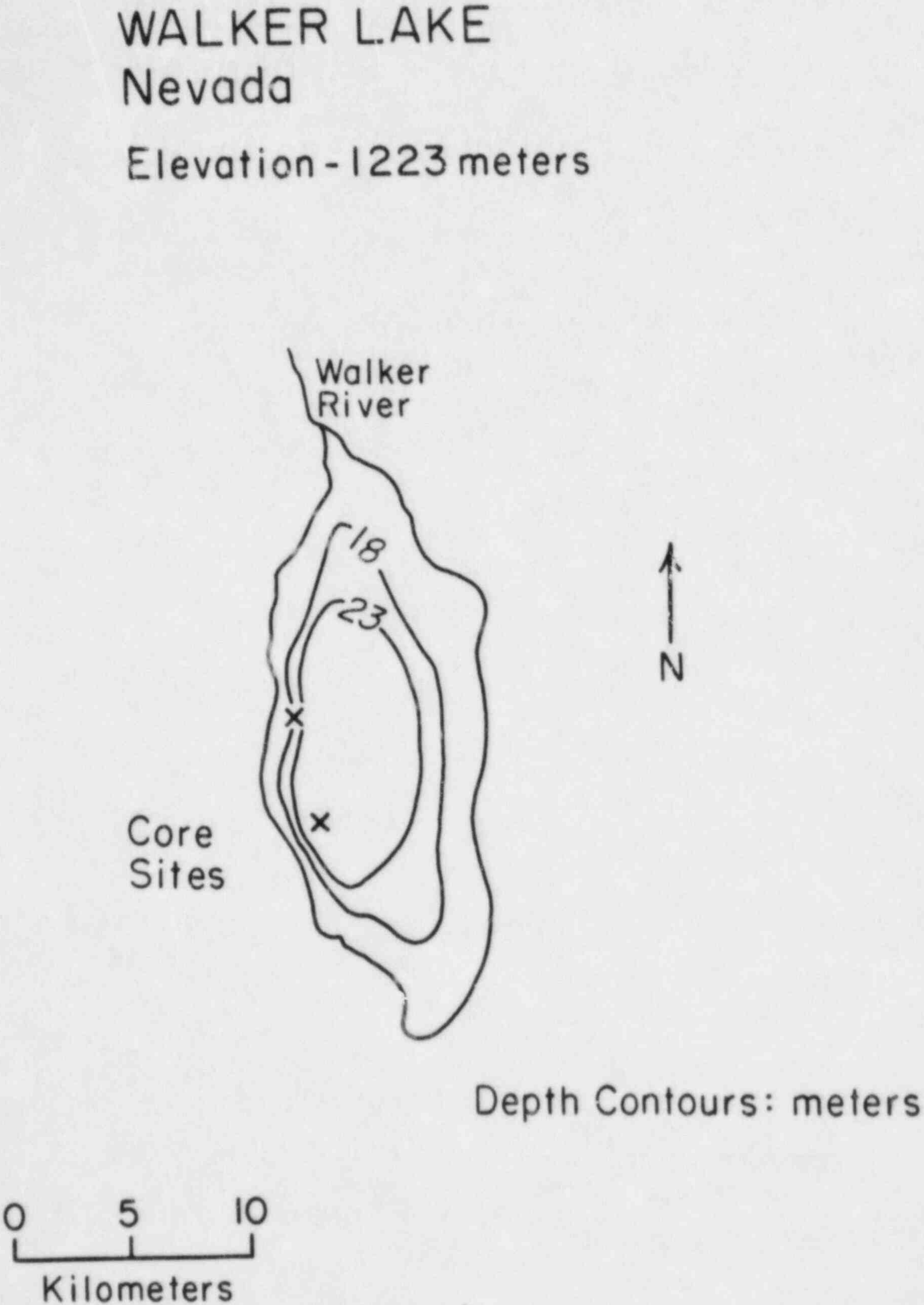
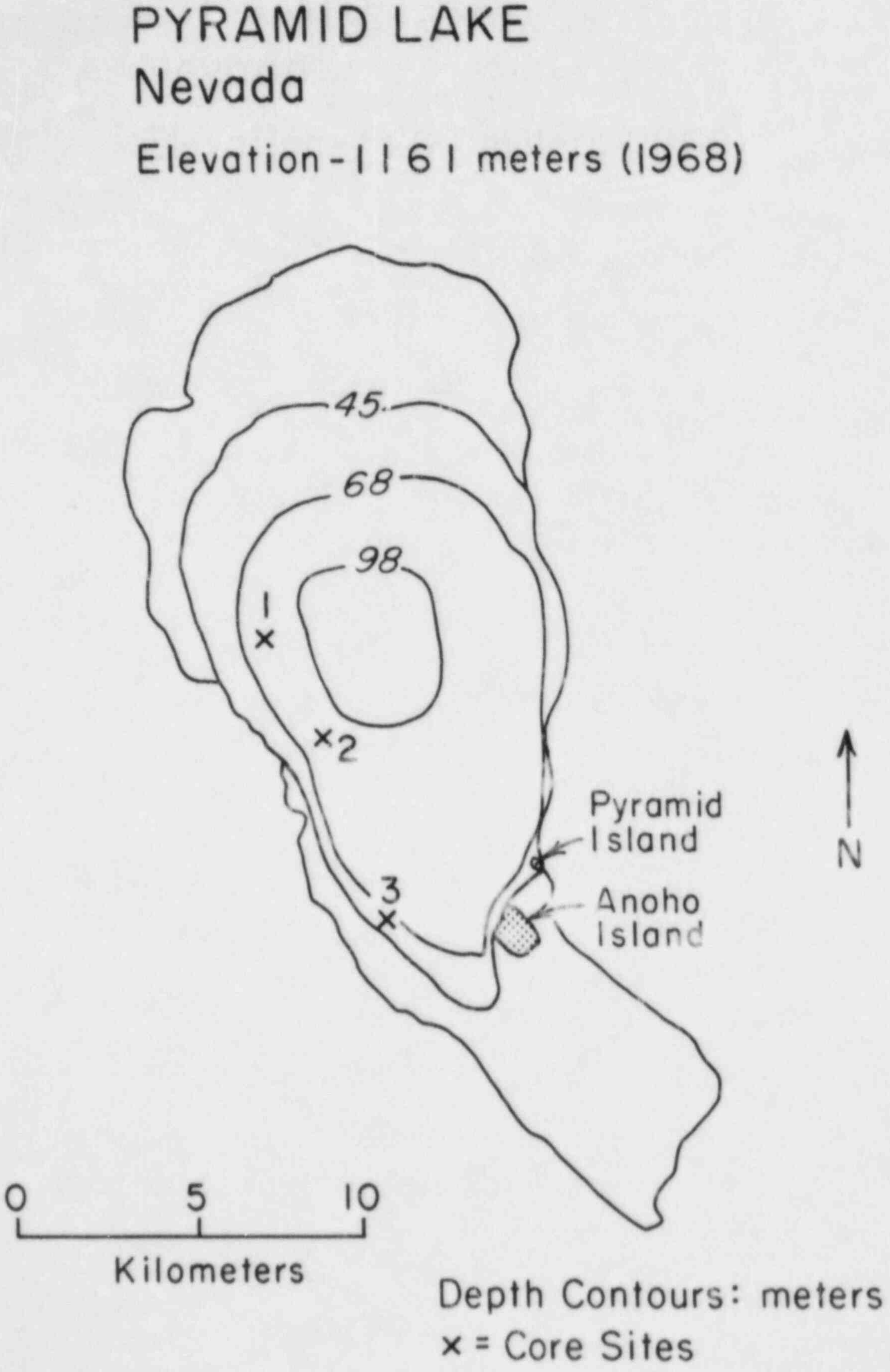

WALKER LAKE
Nevada
Elevation - 1223 meters
Walker River
18
23
N
Core Sites
Depth Contours: meters
0
5
10
Kilometers

Figure 4.



PYRAMID LAKE
Nevada
Elevation - 1161 meters (1968)

Depth Contours: meters
× = Core Sites

Figure 5.

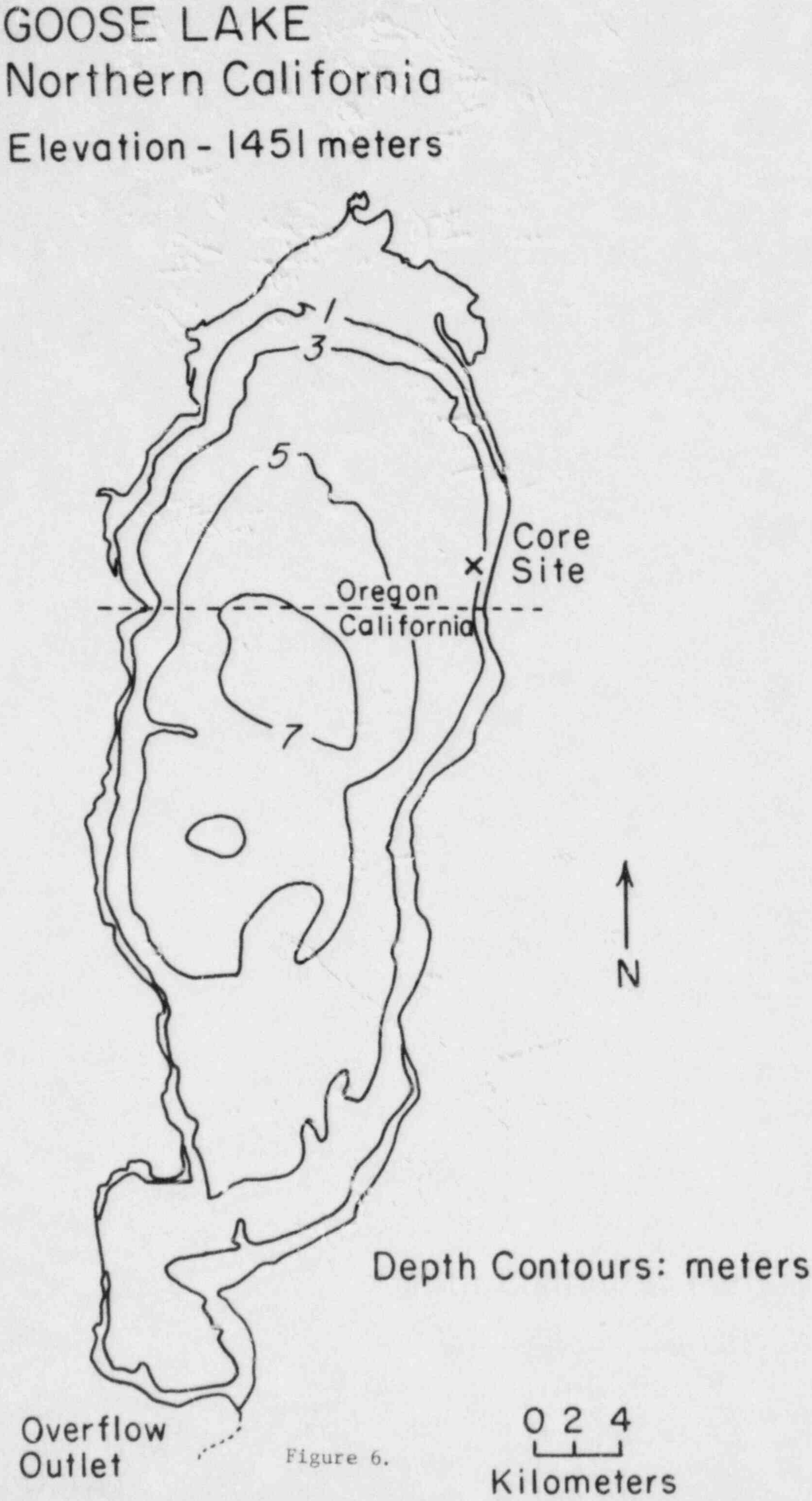
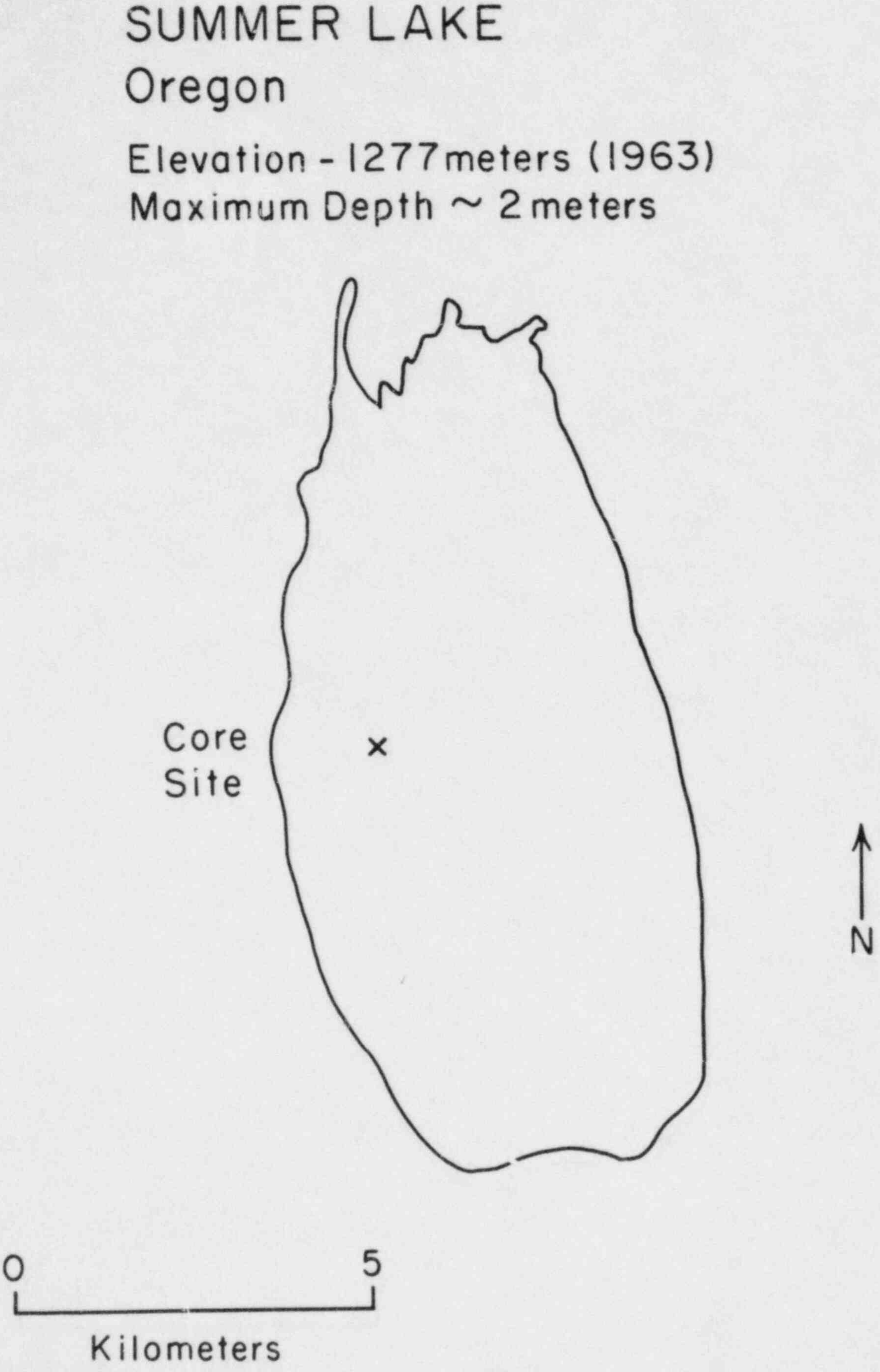

GOOSE LAKE
Northern California
Elevation - 1451 meters
1
3
5
7
Core Site
Oregon
California
N
Depth Contours: meters
Overflow Outlet
0 2 4
Kilometers

Figure 6.



SUMMER LAKE
Oregon
Elevation - 1277 meters (1963)
Maximum Depth ~ 2 meters

Figure 7.

Table 2

Dissolved Plutonium in Mono Lake

Year of Sample Collection	Laboratory Control #	Alpha Spectrum #	Sample Volume (l)	Water Column Depth (m)	^{239,240}Pu (pCi/m^3)	^{238}Pu (pCi/m^3)	^{238}Pu/^{239,240}Pu (x100)
'78	1361	357	80	1	19.5 ± 0.7	0.85 ± 0.11	4.4 ± 0.6
'78	1362	356	80	1	19.0 ± 0.7	0.65 ± 0.10	3.4 ± 0.5
'78	1363	358	240	1	22.9 ± 1.6	1.25 ± 0.25	5.5 ± 1.2
'78	1364	359	160	1	19.1 ± 0.8	0.78 ± 0.11	4.1 ± 0.6
'78	1365	360	240	1	16.4 ± 0.8	0.48 ± 0.09	2.9 ± 0.6
'78	1366	361	240	1	11.1 ± 0.7	0.54 ± 0.12	4.8 ± 1.1
'79	1406	459	75	1	12.6 ± 0.6	0.5 ± 0.1	4.0 ± 0.9
'79	1417	502	14	1	12.6 ± 1.2	N.D.	N.D.
'79	1421	484	80	1	11.2 ± 0.6	N.D.	N.D.
'79	1421	484	80	1	18.7 ± 1.2	N.D.	N.D.
'80	1541	522	80	1	8.1 ± 0.4	0.33 ± 0.07	4.1 ± 0.8
'80	1542	523	80	8	11.7 ± 0.5	0.33 ± 0.07	2.8 ± 0.6
'80	1543	524	80	12	12.1 ± 0.8	0.56 ± 0.15	4.6 ± 1.2
'80	1544	525	80	18	9.2 ± 0.4	0.27 ± 0.06	2.9 ± 0.6
'80	1545	526	80	30	12.1 ± 0.5	0.52 ± 0.09	4.3 ± 0.7
'80	1546	527	80	1(u)	12.7 ± 0.4	0.56 ± 0.08	4.4 ± 0.7
'80	1547	528	80	8(u)	11.5 ± 0.4	0.42 ± 0.07	3.7 ± 0.6
'80	1548	529	80	12(u)	11.3 ± 0.5	0.51 ± 0.10	4.5 ± 0.7
'80	1549	530	80	18(u)	3.5 ± 0.2	0.12 ± 0.03	3.4 ± 0.9
'80	1550	531	80	30(u)	12.7 ± 0.4	0.40 ± 0.06	3.1 ± 0.5
'81	1652	592	80	2	7.8 ± 0.8	N.D.	N.D.
'81	1653	593	80	7	10.4 ± 0.4	0.45 ± 0.07	4.3 ± 0.7
'81	1654	594	80	13	10.1 ± 0.3	0.35 ± 0.05	3.5 ± 0.5
'81	1655	595	80	20	11.1 ± 0.4	0.41 ± 0.06	3.7 ± 0.6
'81	1656	598	80	30	9.3 ± 0.2	0.39 ± 0.03	4.2 ± 0.4
'81	1637	681	80	2	25.1 ± 0.7	1.1 ± 0.1	4.3 ± 0.4
'81	1638	682	80	20	23.5 ± 0.5	0.90 ± 0.06	3.8 ± 0.3
'81	1639	683	80	30	23.8 ± 0.7	0.72 ± 0.08	3.0 ± 0.4
'81	1657	603	80	2	10.6 ± 0.3	0.36 ± 0.04	3.4 ± 0.4
'81	1658	596	80	2	11.9 ± 1.2	N.D.	N.D.
'81	1659	605	80	2	8.2 ± 0.2	0.34 ± 0.04	4.2 ± 0.5
'81	1660	606	80	2	9.1 ± 0.2	0.34 ± 0.04	3.8 ± 0.4
'81	1661	607	80	20	10.5 ± 0.3	0.58 ± 0.05	5.5 ± 0.5

Dissolved plutonium concentrations in L. Abert, Walker L. and Pyramid L. are listed in Table 3. Values for L. Abert were similar to those for Mono L., while those for Walker and Pyramid Lakes were more than an order of magnitude lower.

Dissolved U and Th activities in Mono Lake are listed in Table 4. Dissolved U values were quite high, averaging about 500 dpm/l for ^{234}U, which is almost two hundred times that observed in seawater. Dissolved Th values were also remarkably high for natural waters, averaging about 3 dpm/l for ^{230}Th, which is more than a factor of two thousand times that found in seawater. Dissolved U and Th concentrations in L. Abert, Walker L., Pyramid L., Goose L. and Summer L. are reported in Table 5. In all cases, the U and Th concentrations were considerably greater than in seawater, but not as high as in Mono Lake. Lake Abert had ^{238}U, ^{234}U, ^{232}Th, ^{230}Th and ^{228}Th concentrations of only 10-20% of those in Mono Lake, despite the similarity in dissolved ^{239,240}Pu concentrations in the two lakes. Several isotope ratios for U and Th are listed in Table 6 for Mono Lake and in Table 7 for the remainder of the lakes. The ratio of ^{230}Th to ^{234}U in Mono Lake was about 5x10^{-3}, indicating that for this daughter/parent pair, the parent ^{234}U is preferentially retained in solution over the daughter ^{230}Th by a factor of about 2x10^{2}. Ratios of ^{230}Th to ^{234}U are substantially lower in Walker and Pyramid Lakes (by more than an order of magnitude) than in Mono Lake, with L. Abert intermediate, as was true for thorium concentrations.

Despite the high concentrations of dissolved actinides in these alkaline lakes, there were substantial analytical difficulties for most of the nuclides which we measured. For example, there was such an enormous quantity of dissolved silica removed from L. Abert with the iron hydroxide precipitation step that considerable difficulties were encountered with several subsequent steps in the separations procedures. Fortunately, we have several independent checks on the validity of our analytical results from work at both the University of Southern California (Professor Douglas Hammond) and at Woods Hole Oceanographic Institution (Anderson et al., 1982). Dissolved U and Th concentrations from Mono, Walker, Pyramid and Goose Lakes, as measured at USC, are listed in Table 8. Data for a surface sample of Mono Lake water analyzed at Woods Hole are listed in Table 9. In addition to general confirmation of the data reported here for LDGO measurements, these independent results provide values for ^{234}Th (Table 8, USC data), ^{241}Am, ^{231}Pa and ^{227}Ac (Table 9, WHOI data) which have not been measured at LDGO.

Dissolved radium values for alkaline lakes are reported in Table 10. The first two sets of Mono Lake ^{226}Ra values were (1978, 1980) measured at USC and those for 1981 were done at LDGO. In both labs ^{226}Ra was measured by scintillation counting of the daughter product ^{222}Rn, which was allowed to grow into equilibrium with the parent isotope in a sealed container. As for Pu, U and Th, the activity of ^{226}Ra is higher in Mono Lake than for the other alkaline lakes. However, in the case of dissolved ^{226}Ra, all of the alkaline lake values were comparable to seawater concentrations. Thus, the presence of large amounts of carbonate ion does not appear to significantly enhance the solubility of radium. We have reported only one

Table 3

Dissolved Plutonium in Alkaline Lakes

Lake Sample	Laboratory Control #	Alpha Spectrum #	Sample Volume (1)	Water Column Depth (m)	$^{239,240}Pu$ (pCi/m^3)	^{238}Pu (pCi/m^3)	$^{238}Pu/^{239,240}Pu$ (x100)
L. Abert('80)	1555	538	210	1	6.1 ± 0.1	N.D.	N.D.
" ('81)	1670	608	80	1(u)	12.9 ± 0.3	0.45 ± 0.05	3.5 ± 0.4
" ('81)	1671	609	80	1	12.0 ± 0.3	0.47 ± 0.05	3.9 ± 0.4
" ('81)	1672	604	80	1	11.3 ± 0.2	0.33 ± 0.03	2.9 ± 0.3
" ('81)	1673	602	80	1	10.6 ± 0.4	0.43 ± 0.07	4.0 ± 0.7
Walker L.('79)	1428	483	85	1	0.7 ± 0.1	N.D.	-
" ('79)	1429	488	85	1	0.5 ± 0.1	N.D.	-
" ('79)	1429*		85	1	0.7 ± 0.1	N.D.	-
" ('79)	1430	489	250	1	0.5 ± 0.1	N.D.	-
" ('79)	1438	483	85	1	0.67 ± 0.13	N.D.	-
" ('80)	1553	536	200	1	0.13 ± 0.05	N.D.	-
" ('81)	1663	599	240	25	0.19 ± 0.01	(0.043 ± 0.006)	(23 ± 4)
Pyramid L.('80)	1554	537	200	1	0.30 ± 0.15	N.D.	-
" ('81)	1665	597	240	1	0.29 ± 0.03	N.D.	-
" ('81)	1666	600	80	75	N.D.	N.D.	-
" ('81)	1667	601	240	75	0.11 ± 0.02	N.D.	-

Table 4

Dissolved Uranium and Thorium in Mono Lake

Year of Sample Collection	Laboratory Control #	Alpha Spectrum #	Water Column Depth (m)	Sample Volume (1)	^{238}U (pCi/1)	^{234}U (pCi/1)	^{232}Th (pCi/m^3)	^{230}Th (pCi/m^3)	^{228}Th (pCi/m^3)
'78		-	1	1.0	-	-	611 ± 32	1334 ± 52	653 ± 40
'79	1409B	-	1	0.4	-	-	400 ± 70	1400 ± 140	-
'79	1409C	-	1	0.005	162 ± 10	251 ± 127	-	-	-
'79	1410B	-	1	0.4	-	-	750 ± 80	1080 ± 100	750 ± 130
'79	1410C	-	1	0.005	153 ± 11	232 ± 24	-	-	-
'79	1411B	-	1	0.4	-	-	710 ± 70	1390 ± 100	780 ± 113
'79	1411C	-	1	0.005	196 ± 20	270 ± 24	-	-	-
'79	1418C	-	1	0.005	116 ± 12	190 ± 15	-	-	-
'79	1419B	-	1	0.5	-	-	530 ± 90	1080 ± 120	630 ± 130
'79	1419C	-	1	0.005	130 ± 12	229 ± 17	-	-	-
'79	1420B	-	1	2.0	-	-	510 ± 70	790 ± 90	810 ± 180
'79	1420C	-	1	0.005	(47 ± 4)	(56 ± 4)	-	-	-
'80	1541	522	1	0.5	-	-	638 ± 40	1387 ± 62	999 ± 101
'80	1541	522	1	0.05	177 ± 4	220 ± 7	-	-	-
'80	1541	522	1	80	210 ± 13	252 ± 12	-	-	-
'80	1542	523	8	0.5	-	-	723 ± 33	1420 ± 49	1022 ± 101
'80	1542	523	8	0.05	185 ± 3	224 ± 4	-	-	-
'80	1543	524	12	0.5	-	-	739 ± 34	1453 ± 49	1046 ± 101
'80	1543	524	12	0.05	187 ± 3	220 ± 3	-	-	-
'80	1544	525	18	0.5	-	-	746 ± 55	1321 ± 75	952 ± 102
'80	1544	525	18	0.05	195 ± 9	233 ± 11	-	-	-
'80	1545	526	30	0.5	-	-	677 ± 57	1531 ± 91	1102 ± 109
'80	1545	526	30	0.05	182 ± 6	212 ± 7	-	-	-
'81	1652	592	2	80	-	-	777 ± 36	1340 ± 57	664 ± 30
'81	1653	593	7	80	-	-	815 ± 42	1590 ± 78	655 ± 35
'81	1654	594	13	80	-	-	871 ± 52	1461 ± 84	754 ± 45
'81	1655	595	20	80	-	-	817 ± 49	1358 ± 78	671 ± 41
'81	1656	596	30	80	-	-	-	-	-
'81	1659A	605	2	80	-	-	728 ± 30	1320 ± 48	646 ± 26
'81	1660A	606	2	80	-	-	718 ± 44	1376 ± 80	646 ± 40
'81	1661A	607	20	80	-	-	698 ± 48	1304 ± 86	668 ± 46

Table 5

Dissolved Uranium and Thorium in Alkaline Lakes

Lake Sampled	Laboratory Control #	Alpha Spectrum #	Water Column Depth (m)	^{238}U (pCi/l)	^{234}U (pCi/l)	^{232}Th (pCi/m^3)	^{230}Th (pCi/m^3)	^{228}Th (pCi/m^3)
L. Abert	1670	608	1	-	-	115 ± 5	176 ± 7	140 ± 6
"	1671	609	1	19.6 ± 1.3	55.1 ± 2.3	84 ± 3	107 ± 3	91 ± 3
"	1671	738	1	17.6 ± 1.0	38.4 ± 1.7	-	-	-
"	1672	604	1	-	-	87 ± 3	114 ± 3	100 ± 3
"	1673	602	1	-	-	90 ± 3	123 ± 3	107 ± 3
Walker L.	1663	731A	25	51.9 ± 1.7	69.6 ± 2.2	5.2 ± 0.2	8.1 ± 0.3	47 ± 1
"		731B	25	49.4 ± 2.6	75.1 ± 3.6	-	-	-
Pyramid L.	1665	597	1	-	-	3.2 ± 0.2	4.1 ± 0.2	8.3 ± 0.4
"	1665	733	1	6.1 ± 0.3	12.0 ± 0.5	-	-	-
"	1666	600	75	-	-	2.2 ± 0.1	3.0 ± 0.1	8.7 ± 0.3
"	1667	601	75	-	-	3.9 ± 0.2	5.0 ± 0.2	10.7 ± 0.3
Goose L.	1668	732	1	4.2 ± 0.2	8.8 ± 0.3	-	-	-
Summer L.	1674B	735	1	9.3 ± 0.2	15.4 ± 0.3	-	-	-

Table 6

Uranium and Thorium Isotope Ratios in Mano Lake Water

Year of Sample Collection	Laboratory Control #	Alpha Spectrum #	Water Column Depth(m)	$^{234}U/^{238}U$	$^{230}Th/^{232}Th$	$^{228}Th/^{232}Th$	$^{230}Th/^{234}U$ $(x10^{-3})$
'78				–	2.18 ± .14	1.07 ± .09	–
'79	1409	–	1	1.55 ± .79	3.5 ± .71	–	5.6 ± 2.9
'79	1410	–	1	1.51 ± .19	1.44 ± .20	1.00 ± .20	4.7 ± 0.7
'79	1411	–	1	1.38 ± .19	1.96 ± .24	1.10 ± .19	5.1 ± 0.6
'79	1418	–	1	1.64 ± .21	–	–	–
'79	1419	–	1	1.76 ± .21	2.04 ± .41	1.19 ± .32	4.7 ± 0.7
'79	1420	–	1	1.19 ± .13	1.55 ± .28	1.59 ± .42	(14.1) ± 1.9
'80	1541	522	1	1.24 ± .05	2.17 ± .17	1.57 ± .19	6.3 ± 0.4
'80	1541	522	1	1.20 ± .09	–	–	–
'80	1542	523	8	1.21 ± .03	1.96 ± .11	1.41 ± .15	6.3 ± 0.7
'80	1543	524	12	1.18 ± .02	1.97 ± .11	1.42 ± .15	6.6 ± 0.3
'80	1544	525	18	1.19 ± .08	1.77 ± .16	1.28 ± .17	5.7 ± 0.5
'80	1545	526	30	1.16 ± .05	2.26 ± .23	1.63 ± .21	7.2 ± 0.8
'81	1652	592	2	–	1.72 ± .11	0.85 ± .05	–
'81	1653	593	7	–	1.95 ± .14	0.80 ± .06	–
'81	1654	594	13	–	1.68 ± .14	0.87 ± .07	–
'81	1655	595	20	–	1.66 ± .14	0.82 ± .07	–
'81	1659	605	2	–	1.81 ± .10	0.89 ± .05	–
'81	1660	606	2	–	1.92 ± .16	0.90 ± .08	–
'81	1661	607	20	–	1.87 ± .18	0.96 ± .09	–

Table 7

Uranium and Thorium Isotope Ratios in Alkaline Lake Waters

Lake Sampled	Laboratory Control #	Alpha Spectrum #	Water Column Depth(m)	$^{234}U/^{238}U$	$^{230}Th/^{232}Th$	$^{228}Th/^{232}Th$	$^{230}Th/^{234}U$ $(x10^{-3})$
L. Abert	1670	608	1	-	1.53 ± .09	1.21 ± .05	-
"	1671	609	1	2.81 ± .22	1.27 ± .06	1.08 ± .05	1.9 ± .1
"	1671	738	1	2.18 ± .16	-	-	-
"	1672	604	1	-	1.31 ± .06	1.15 ± .05	-
"	1673	602	1	-	1.37 ± .06	1.19 ± .05	-
Walker L.	1663	731A	25	1.34 ± .06	1.56 ± .08	(9) ± .40	0.12 ± .01
"	1663	731B	25	1.52 ± .11	-	-	-
Pyramid L.	1665	597,733	1,30	1.97 ± .13	1.28 ± .10	2.59 ± .20	0.34 ± .02
"	1666	600	75	-	1.36 ± .08	2.90 ± .17	-
"	1667	601	75	-	1.28 ± .08	2.14 ± .12	-
Goose L.	1668	732	1	2.10 ± .12	-	-	-
Summer L.	1674	735	1	1.66 ± .05	-	-	-

Table 8

Dissolved Uranium and Thorium in Alkaline Lakes (U.S.C. Data)

Lake Sampled	Water Column Depth (m)	^{238}U (pCi/l)	^{234}U (pC/l)	$^{234}U/^{238}U$	^{234}Th (pCi/l)	^{232}Th (pCi/m^3)	^{230}Th (pCi/m^3)	^{228}Th (pCi/m^3)
Mono L.-'80	30	-	-	-	216 ± 18	631 ± 9	1577 ± 158	-
L. Abert-'81	1	13 ± 1	28 ± 2	2.2 ± 0.1	18 ± 3	66 ± 4	86 ± 3	-
Walker L.-'81	1	57 ± 2	74 ± 3	1.3 ± 0.06	41 ± 6	-	61 ± 5	-
"	25	61 ± 3	83 ± 4	1.4 ± 0.07	31 ± 4	-	132 ± 6	-
Pyramid L.-'81	1	-	-	-	0.59 ± 0.09	-	6.8 ± 0.5	-
"	75	0.77 ± 0.09	1.1 ± 0.14	1.4 ± 0.17	0.50 ± 0.14	-	0.18 ± 0.05	-
Goose L.-'81	1	3.8 ± 0.3	5.0 ± 0.4	1.3 ± 0.12	1.8 ± 0.3	49 ± 2	53 ± 4	-

Table 9

Mono Lake Actinide Concentrations (Woods Hole Data)*

Nuclide	Activity			
$^{239,240}Pu$	11.2	±	1.1	pCi/m^3
^{241}Am	1.22	±	0.27	pCi/m^3
^{238}U	108	±	3	pCi/l
^{234}U	123	±	4	pCi/l
^{232}Th	399	±	14	pCi/m^3
^{230}Th	754	±	20	pCi/m^3
^{228}Th	435	±	16	pCi/m^3
^{231}Pa	35.8	±	1.3	pCi/m^3
^{227}Ac	< 2.7			pCi/m^3

*Data are based on a sample collected from the western shore of the lake having a Cl^- concentration of 13.2 g/l (Anderson et al., 1982), indicating that the shoreline sample reported here was probably diluted by about one-third with fresh spring water.

Table 10

Dissolved Radium Isotopes in Alkaline Lakes

Lake Sample	Laboratory Control #	Depth (m)	^{226}Ra (pCi/m^3)		^{228}Ra (pCi/m^3)
Mono L. - ('78)	-	1		450 ± 100	-
"	-	1-11	536	506 ± 36	231 ± 41
"	-	"	536		
"	-	15-30	482		
"	-	"	468		
Mono L. - ('81)	1652	2	326	332 ± 10	-
"	"	"	339		
L. Abert - ('81)	1670	1	41	39 ± 3	-
"	"	"	37		
Walker L. - ('81)	1663	1	195	199 ± 15	-
"	"	"	216		
"	"	"	186		
Pyramid L. - ('81)	1665	30	26	26 ± 1	-
"	"	"	27		
Goose L. - ('81)	1668	1	21	20 ± 1	-
"	"	"	21		
"	"	"	19		

value of ^{228}Ra, measured by gamma spectrometry determination of ^{228}Ac using a large quantity of $CaCO_3$ precipitated from Mono Lake water. Subsequent attempts to duplicate this procedure have not been successful. At present, it appears as if the $CaCl_2$ we added to the 1981 samples to cause precipitation of $CaCO_3$ was sufficiently contaminated with radium isotopes to introduce substantial error in the final measurements. Since we used several different batches of $CaCl_2$ during the 1981 field work, we have not been able to establish consistent enough blank values to obtain reproducible ^{228}Ra values from measurement of its immediate daughter ^{228}Ac. The data available to us suggest that the ^{228}Ra to ^{226}Ra activity ratio in the alkaline lakes we sampled is of the order of 0.5.

Dissolved concentrations of ^{210}Pb and ^{210}Po, both of which are in the ^{238}U decay series, are listed in Table 11. Most of the values are of the order of 25-50% of the dissolved ^{226}Ra concentrations, with ^{210}Po/^{210}Pb ratios about 0.5. However, L. Abert had a considerably higher ^{210}Pb concentration than the other lakes, having a ^{210}Pb to ^{226}Ra ratio of greater than 10. One possible explanation could be input of ^{210}Pb by precipitation to the lake surface. Since L. Abert has the lowest mean depth of the lakes for which we have ^{210}Pb data, precipitation-supplied ^{210}Pb would be most likely to be significant in this lake relative to production in the water column by decay of dissolved ^{226}Ra through the short-lived daughter products ^{222}Rn ($t_{1/2}$ = 3.8 days), ^{218}Po ($t_{1/2}$ = 3 minutes), ^{214}Pb ($t_{1/2}$ = 27 minutes), ^{214}Bi ($t_{1/2}$ = 20 minutes), and ^{214}Po ($t_{1/2} = 2\times10^{-4}$ sec). A first-order estimate of the maximum concentration of ^{210}Pb in L. Abert which could be supported by precipitation can be made as follows, assuming no removal from solution except by radioactive decay:

$$\underset{\text{(max)}}{\text{L. Abert } {}^{210}\text{Pb}} = \frac{[\text{Precip } {}^{210}\text{Pb}] \times \dfrac{25 \text{ cm}}{\text{Yr of Precip}} \times \dfrac{22 \text{ Yrs}}{\ln 2} \quad \begin{pmatrix}\text{mean life} \\ \text{of } {}^{210}\text{Pb}\end{pmatrix}}{100 \text{ cm (mean depth)}}$$

Using a precipitation ^{210}Pb value of 4 pCi/l (average of 17 months of precipitation in Connecticut [Benninger, 1976]), the maximum L. Abert ^{210}Pb concentration would be ≈ 3×10^{4} pCi/m^3. Since the measured L. Abert ^{210}Pb concentration is only ≈ 8×10^{2} pCi/m^3, it is clear that precipitation input could easily explain the very high ^{210}Pb/^{226}Ra ratio in the lake water. Using the above precipitation input values, and assuming no stream supply of dissolved ^{210}Pb, the residence time of ^{210}Pb in the water column of L. Abert would be of the order of 0.5 year.

Also listed in Table 11 is a value of 115±10 pCi/m^3 for ^{231}Pa in Mono L., measured at USC. The only other reported ^{231}Pa concentration (35.8±1.3 pCi/m^3) was for a Mono L. surface sample, partially diluted with fresh water runoff (Table 9). Adjusting this to average Mono L. chlorinity (≈ 20 g/l), the two ^{231}Pa measured concentrations are: 115 and 54 pCi/m^3. The concentration of the parent nuclide, ^{235}U, in Mono Lake is

Table 11

Dissolved ^{210}Pb, ^{210}Po, and ^{231}Pa in Alkaline Lakes (USC Data)

Lake Sampled	Water Column Depth (m)	^{210}Pb (pCi/m^3)	^{210}Po (pCi/m^3)	^{231}Pa (pCi/m^3)
Mono L. - '79	1	261 ± 9	131 ± 5	115 ± 10
"	1	284 ± 14	126 ± 5	-
Mono L. - '80	8	157 ± 7	122 ± 14	-
"	30	94 ± 3	44 ± 6	-
L. Abert - '81	1	752 ± 45	185 ± 32	-
Walker L. - '80	1	63 ± 5	7.7 ± 2.3	-

obtained by multiplying the dissolved concentration of ^{238}U (1.7×10^{5} pCi/m^{3}) times the ^{235}U/^{238}U activity ratio (4.6×10^{-2}) = 7.8×10^{3} pCi/m^{3}. Thus, the ^{231}Pa/^{235}U ratio in Mono L. is $7\text{-}14\times10^{-3}$, or approximately 1-2 times the ^{230}Th/^{234}U ratio (Table 6). This data suggests that actinides with oxidation states of +5 (i.e. ^{231}Pa) experience complexing influence by carbonate ion similar to that for oxidation state +4 nuclides (i.e. ^{230}Th).

We have also obtained data for dissolved fission product fallout isotopes (^{137}Cs and ^{90}Sr) in several alkaline lakes (Table 12). ^{137}Cs concentrations in Mono L. averaged about 2 pCi/l for 12 samples collected in 1978, 1980 and 1981. This represents a large fraction of the total fallout delivery of ^{137}Cs to Mono L., since the total sediment inventory is relatively small. In contrast, both Walker L. and Pyramid L. have dissolved ^{137}Cs concentrations which are less than 10% of those for Mono L. Although the greater mean depth of Pyramid L. (≃3 times that for Mono L. and Walker L.) could account for part of the difference, the main cause is probably greater partitioning of ^{137}Cs to the sediments in the less saline lakes (Walker L. and Pyramid L.). The measured concentration of ^{90}Sr in Mono L. for 1978 and 1980 was about 0.16 pCi/l, or about 0.08 of ^{137}Cs. This suggests greater removal of ^{90}Sr to the sediments than for ^{137}Cs, since the fallout delivery ratio of ^{90}Sr to ^{137}Cs was about 0.67.

Tritium concentrations for four alkaline lakes are listed in Table 13. The lowest value (35 T.U.) was for L. Abert, and the highest for Walker L. (86-97 T.U.). Detailed vertical profiles for both Mono L. and Pyramid L. indicate uniform concentrations with depth in the two lakes (Mono L. ≃ 70 T.U. and Pyramid L. ≃ 45 T.U.).

5.0 SEDIMENT RADIONUCLIDE CONCENTRATIONS IN ALKALINE LAKES

Data for ^{137}Cs concentrations in twelve sediment cores collected at nine sites in Mono L. over the period 1978-1981 are listed in Table 14 (see Figure 2 for core locations). Most of the ^{137}Cs activity was confined to the top 10 cm of the sediments, and many of the cores had maximum values several centimeters below the surface, but above 5 cm depth (Figures 8 and 9). The sediment ^{137}Cs inventory per unit area was greatest in the deep central basin south of Paoha Island, and the highest ^{137}Cs activity measured was about 0.4 pCi/g. Lake Abert sediments had considerably greater ^{137}Cs activities per unit weight of sediment (up to a maximum of more than 3 pCi/g), as well as deeper penetration in the cores (Table 15, Figure 10). All four of the other alkaline lakes had greater ^{137}Cs sediment concentrations than Mono L. (Table 16, Figures 11, 12, 13 and 14), but the depth distributions for three cores from Pyramid L. (Figure 12) was similar to cores from the deepest area of Mono L.

Sediment ^{239,240}Pu concentrations in Mono L. (Table 17) had generally similar trends with depth (Figures 15 and 16) to those for ^{137}Cs, but subsurface maximum concentrations per unit weight of sediment were much higher (up to 230 pCi/kg), relative to surface values (≃ 10 pCi/kg). Thus, the ^{239,240}Pu concentration gradients in Mono L. sediments tended to be much greater than for ^{137}Cs. Lake Abert sediment ^{239,240}Pu concentrations (Table 18) were similar to the average of Mono L. values,

Table 12

Dissolved ^{137}Cs and ^{90}Sr in Alkaline Lakes

Lake Sampled	Laboratory Control #	Water Column Depth (m)	^{137}Cs (pCi/1)	^{90}Sr (pCi/1)
Mono L. - '78	1368	1	1.87 ± 0.04	0.185 ± 0.045
"	1369	1	2.01 ± 0.04	-
"	1370	1	1.57 ± 0.03	-
"	1371	1	2.00 ± 0.04	-
Mono L. - '79		1		0.023 ± 0.015
Mono L. - '80	1541	1	1.9 ± 0.2	-
"	1542	8	1.8 ± 0.2	0.162 ± 0.017
"	1545	30	1.9 ± 0.2	0.127 ± 0.013
Mono L. - '81	1629	2	2.36 ± 0.09	
"	1630	7	2.25 ± 0.11	
"	1631	13	2.31 ± 0.07	
"	1632	20	1.99 ± 0.11	
"	1676	30	2.26 ± 0.08	
Walker L. - '81	1633	25	0.13 ± 0.04	
Pyramid L. - '81	1634	1	0.11 ± 0.05	
"	1635	75	0.17 ± 0.05	
Goose L. - '81		1		0.812 ± 0.030

Table 13

Tritium in Alkaline Lakes

Lake Sampled	Sampling Time	Water Column Depth (m)	Tritium Activity (T.V.)
Mono L.	8/80	1	73.1 ± 1.1
"	"	8	72.7 ± 1.1
"	"	12	72.5 ± 1.1
"	"	18	71.8 ± 1.0
"	"	30	72.4 ± 1.1
Mono L.	7/81	1	65.5 ± 1.1
"	"	10	68.0 ± 1.2
"	"	30	67.7 ± 1.1
L. Abert	7/80	1	35.0 ± 0.9
L. Abert	7/81	1	33.8 ± 2.5
Walker L.	7/80	1	95.1 ± 1.5
Walker L.	7/81	1	86.4 ± 5.3
"	"	50	96.7 ± 3.6
Pyramid L.	7/80	1	47.2 ± 0.6
Pyramid L.	7/81	1	44.2 ± 3.0
"	"	15	41.7 ± 2.0
"	"	30	44.6 ± 1.0
"	"	45	43.8 ± 0.7
"	"	60	46.2 ± 0.9
"	"	75	44.1 ± 3.3

Table 14

Mono Lake - ^{137}Cs (pCi/kg) In Sediment Cores

Station #	1	2A	3A	4A	5A	6A	6B	7	8A	8B	9A	9B
Depth (cm)	CN 1373	CN 1433	CN 1436	CN 1439	CN 1551	CN 1677	CN 1675	CN 1678	CN 1679	CN 1680	CN 1681	CN 1682
0-1	132±50	77±30	54±47	150±56	120±39	146±72	168±26	110±50	110±60	63±56	49±51	99±37
1-2	275±40	29±24	68±47	110±41	151±42	122±61	210±17	112±51	42±77	410±85	99±80	253±44
2-3	260±50			130±70	189±34	2[illegible]±69	119±34	287±49	285±65	-104±80	333±74	31±42
3-4	16±40				152±81	-37±77	83±54	420±71	200±95	98±86	73±75	62±39
4-5	60±40				5±38	131±83	64±42	167±53	40±68	-11±76	58±49	76±48
5-6					36±16	86±81	104±81	110±64		-23±76	15±91	115±56
6-7						128±75	5±60	176±60	3±75	177±72	74±56	159±57
7-8					76±16	30±55	122±54	118±82	164±75		-18±75	75±66
8-9						60±62	121±60	158±55	97±53		-16±69	-17±77
9-11					50±20	32±48	20±45	71±31			105±50	-18±41
11-13					8±32	-34±51	25±19	107±34			105±46	37±50
13-15						-33±60	70±43	75±41			9±51	
15-20					46±33		37±23	34±22			31±25	
20-25					7±24+		24±15	17±18			11±29	
25-30					1±16*			37±17			-13±21	
					1±31†							

+ 19-23 cm

* 23-27 cm

† 27-31 cm

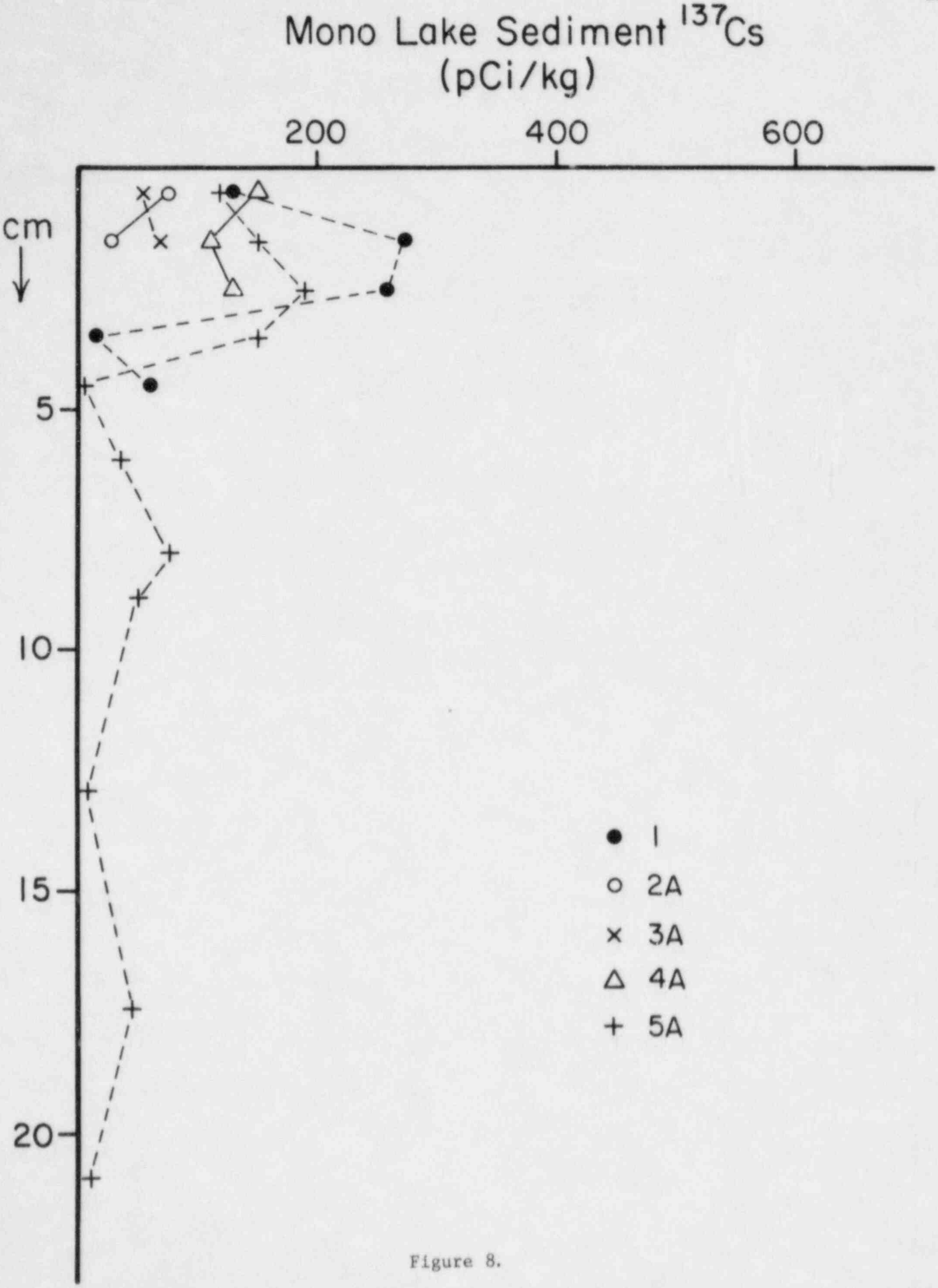

Mono Lake Sediment ^{137}Cs
(pCi/kg)
200
400
600
cm
5
10
15
20
● 1
○ 2A
× 3A
△ 4A
+ 5A

Figure 8.

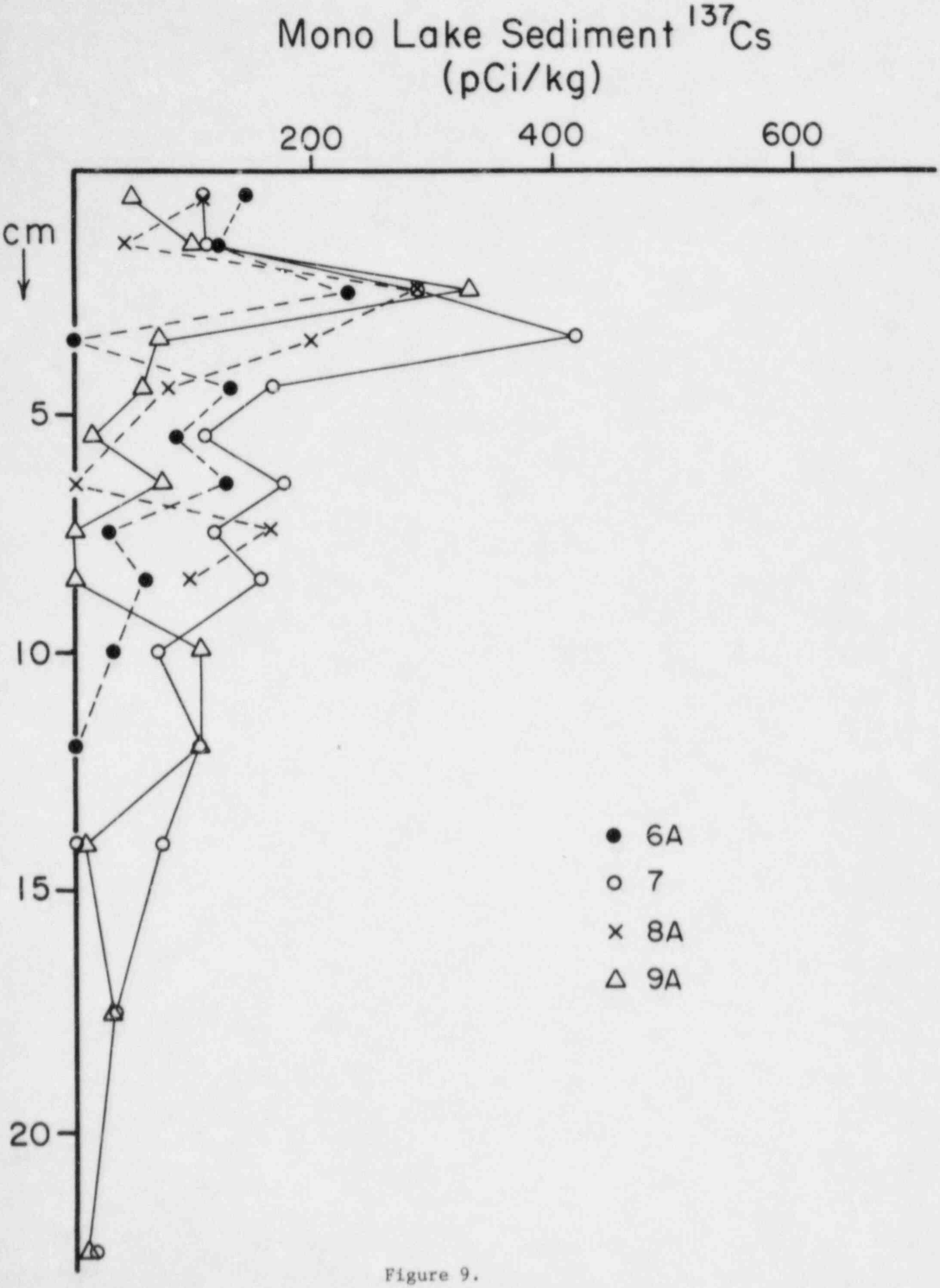

Mono Lake Sediment ^{137}Cs
(pCi/kg)
200
400
600
cm
5
10
15
20
● 6A
○ 7
× 8A
△ 9A

Figure 9.

Table 15

Lake Abert - ^{137}Cs (pCi/kg) In Sediment Cores

Station #	1	1B	2A	2B	3A	3B	4
Depth (cm)	CN 1690	CN 1691	CN 1692	CN 1650	CN 1694	CN 1695	CN 1651
0-1	2195±193	1483±260	1627±93	1456±85	854±45	826±79	1483±140
1-2	2040±82	1948±105	2110±134	1815±81	312±74	451±44	1776±123
2-3	2261±81	2222±97	2094±123	1962±131	327±44	304±44	1852±89
3-4	2847±91	2150±84	1909±127	2007±104	285±43	191±51	1368±101
4-5	2680±92	2302±117	1304±86	2185±83	243±39	79±55	1458±81
5-6	2226±91	3196±120	819±66	2105±81	125±54	34±53	1365±122
6-7	3010±96	2882±106	734±57	1987±113	123±54	-62±57	1973±98
7-8	2419±87	2849±109	885±54	1692±69	186±57	41±52	1807±75
8-9	2489±107	3255±106	531±57	941±63	72±68	-76±55	1189±79
9-11	2910±88	2722±85	324±42	821±41	15±27†	-100±61	1661±71
11-13	2781±100	2877±77	217±34	669±36		-3±21	962±41
13-15	1923±58	2678±81	44±21*	386±29			601±28
15-20	307±21	159±18		121±16			264±21
20-25	77±38+	998±35					132±15
25-30							19±13
30-35							11±14
35-40							-17±20
40-45							5±21
45-50							-5±16

+ 20-22 cm

* 13-17 cm

† 9-14 cm

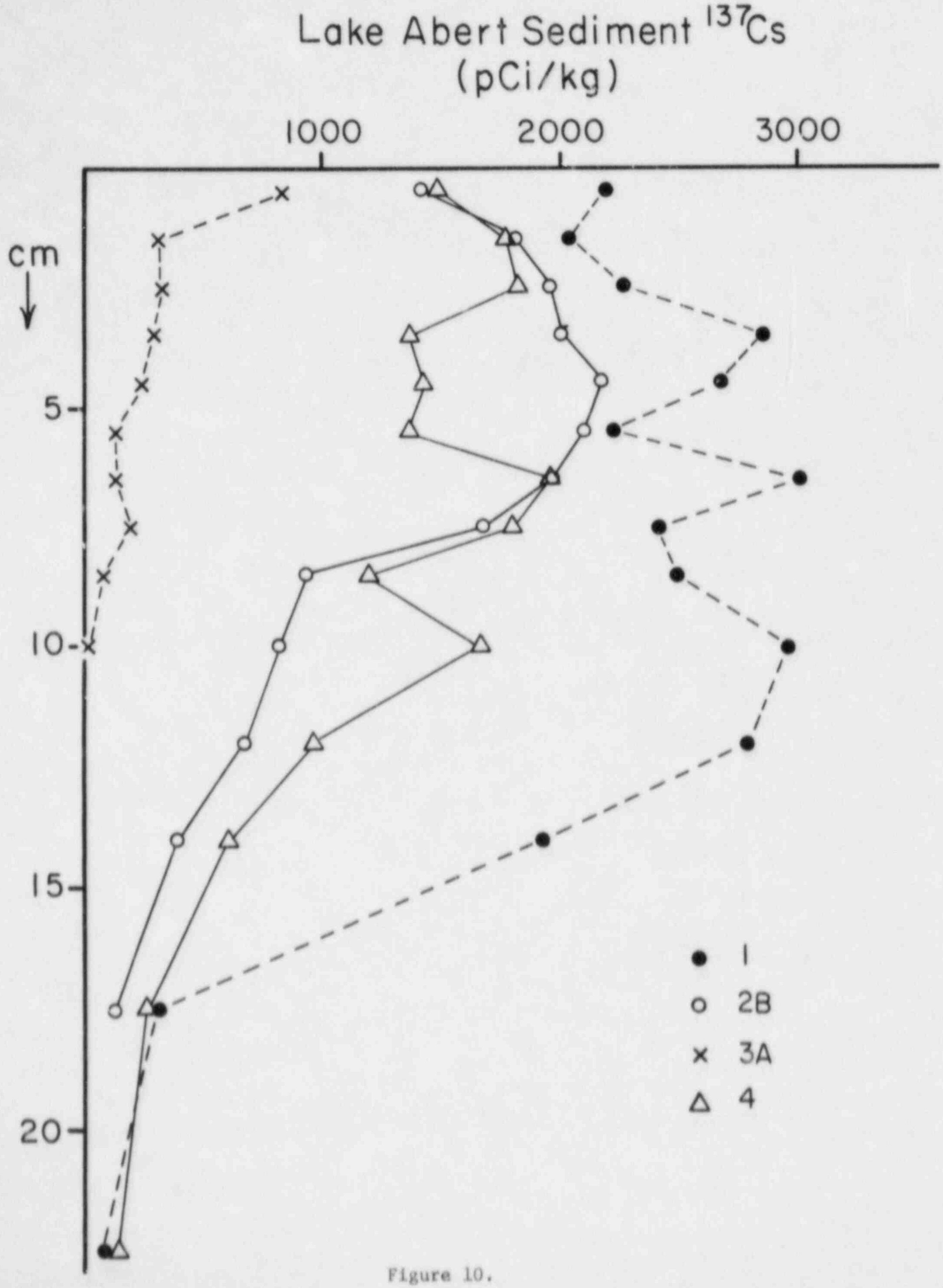

Lake Abert Sediment ^{137}Cs
(pCi/kg)
1000
2000
3000
cm
5
10
15
20
● 1
○ 2B
× 3A
△ 4

Figure 10.

Table 16

Alkaline Lake Sediment -^{137}Cs (pCi/kg)

Depth (cm)	Walker L. Station 1 CN 1442	Walker L. Station 2 CN 1697	Pyramid L. Station 1 Cn 1685	Pyramid L. Station 2 Cn 1686	Pyramid L. Station 3 CN 1687	Goose L. Station 1 CN 1683	Goose L. Station 2 N 1684	Summer L. Station 1 CN 1688	Summer L. Station 2 CN 1689
Surf.floc.		1534±161							
0-1	1915±120	1871± 79	867±179	1196±112	1270±182	662±45	829±199	660±119	702±102
1-2	2170±130	2176± 72	767±120	1235±123	1215±121	682±76	991± 86	599± 44	564± 40
2-3	420±140	{ }	1387±127	992±114	1106± 97	629±40	708± 52	249± 32	209± 21
3-4		{3020± 78}	551± 93	477±100	530±140	405±38	605± 36	169± 37	40± 24
4-5		{ }	-14± 88	138± 65	281± 83	433±34	652± 45	43± 36	47± 29
5-6		{3377± 78}	-5± 81	84± 78	197± 84	277±37	592± 40	55± 42	56± 28
6-7		{ }	-145± 93	89± 65	218± 84	217±28	571± 49	22± 31	39± 23
7-8		{3301± 73}	32± 74	-33± 73	78± 88	123±28	229± 15	10± 37	76± 28
8-9		{ }	-90± 84	186± 73	52± 81	116±23	570± 46	12± 28	21± 25
9-11		{2595± 77}	20± 45	8± 51	32± 50	82±21	384± 24	17± 23	24± 24
11-13			8± 53			33±20	215± 18	23± 22	9± 19
13-15			-13± 45			-16±19	105± 18	11± 25	2± 24
15-20			3± 33			-21±18	12± 19	-4± 16	
20-25								10± 21	

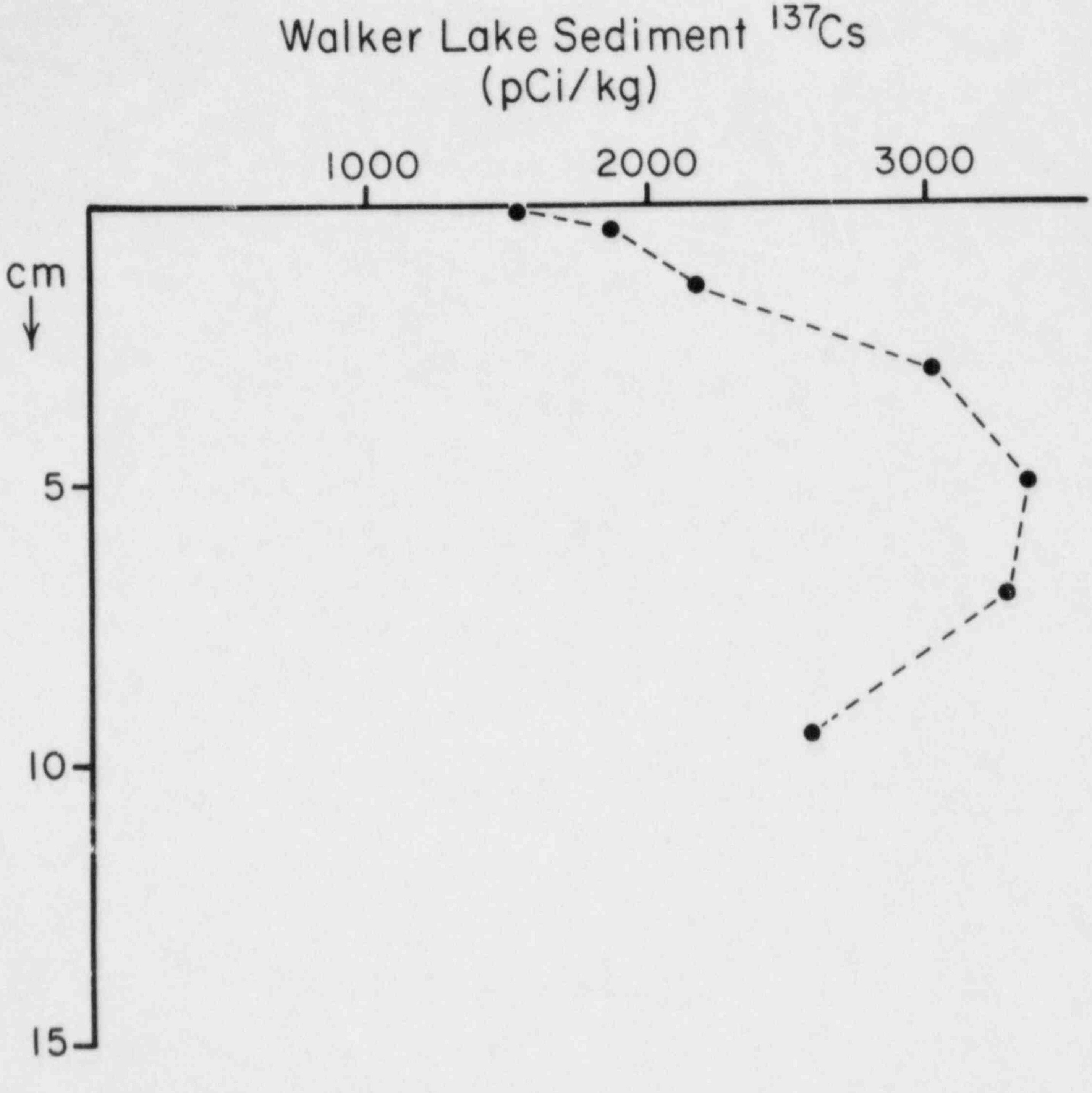


Figure 11.

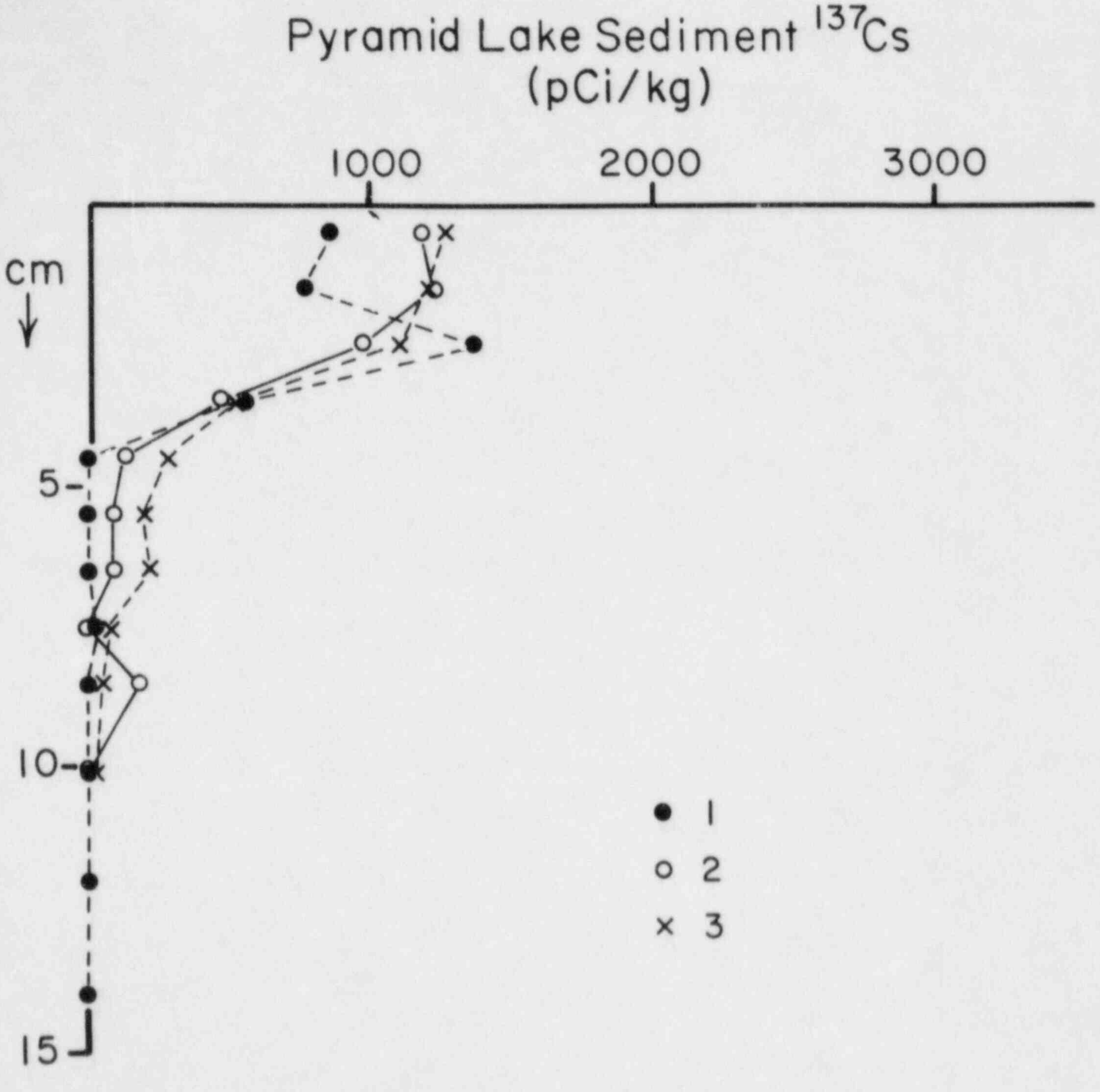

Pyramid Lake Sediment ^{137}Cs
(pCi/kg)
1000
2000
3000
cm
5-
10-
15
● 1
○ 2
× 3

Figure 12.

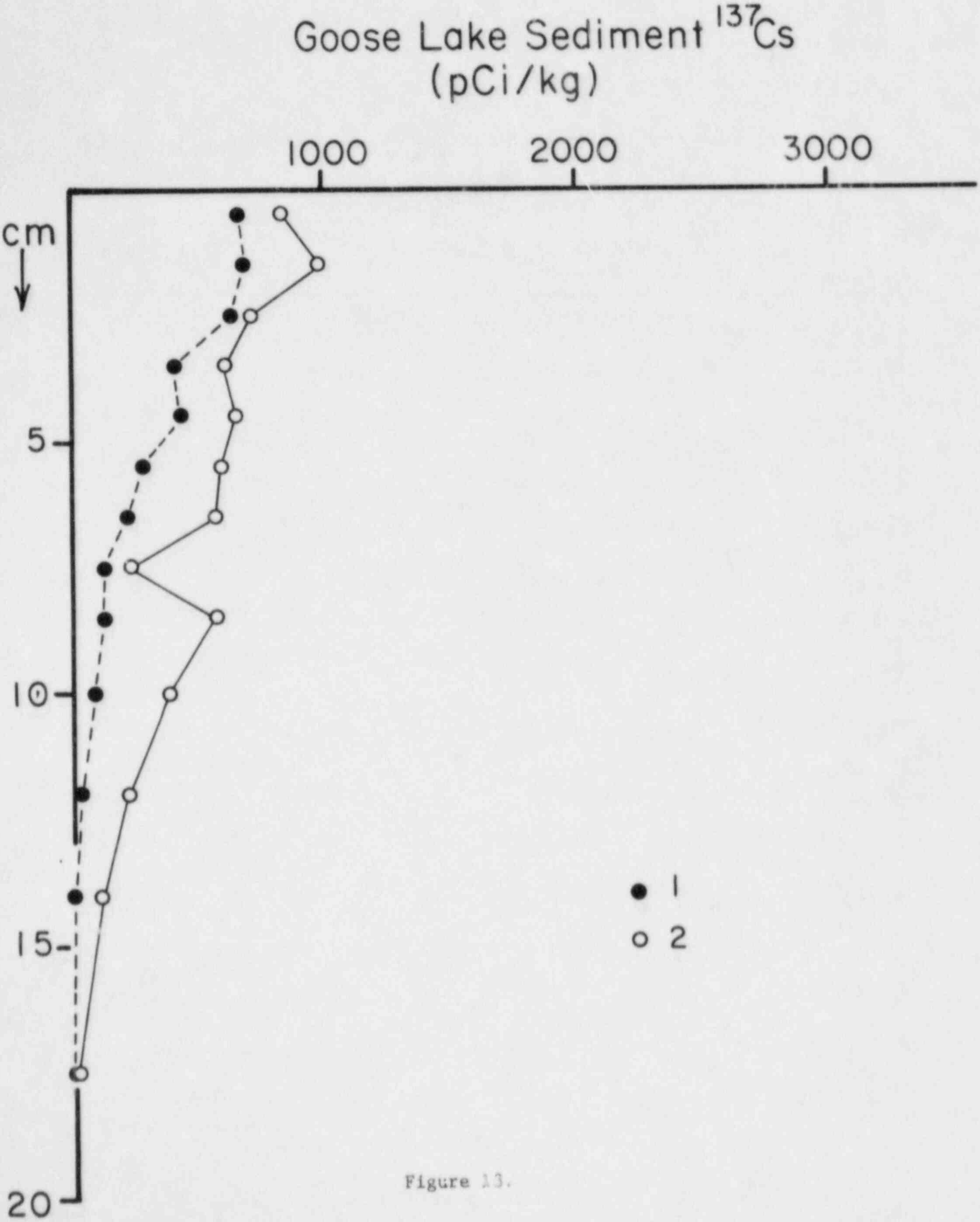

Goose Lake Sediment ^{137}Cs
(pCi/kg)
1000
2000
3000
cm
5
10
15
20
● 1
○ 2

Figure 13.

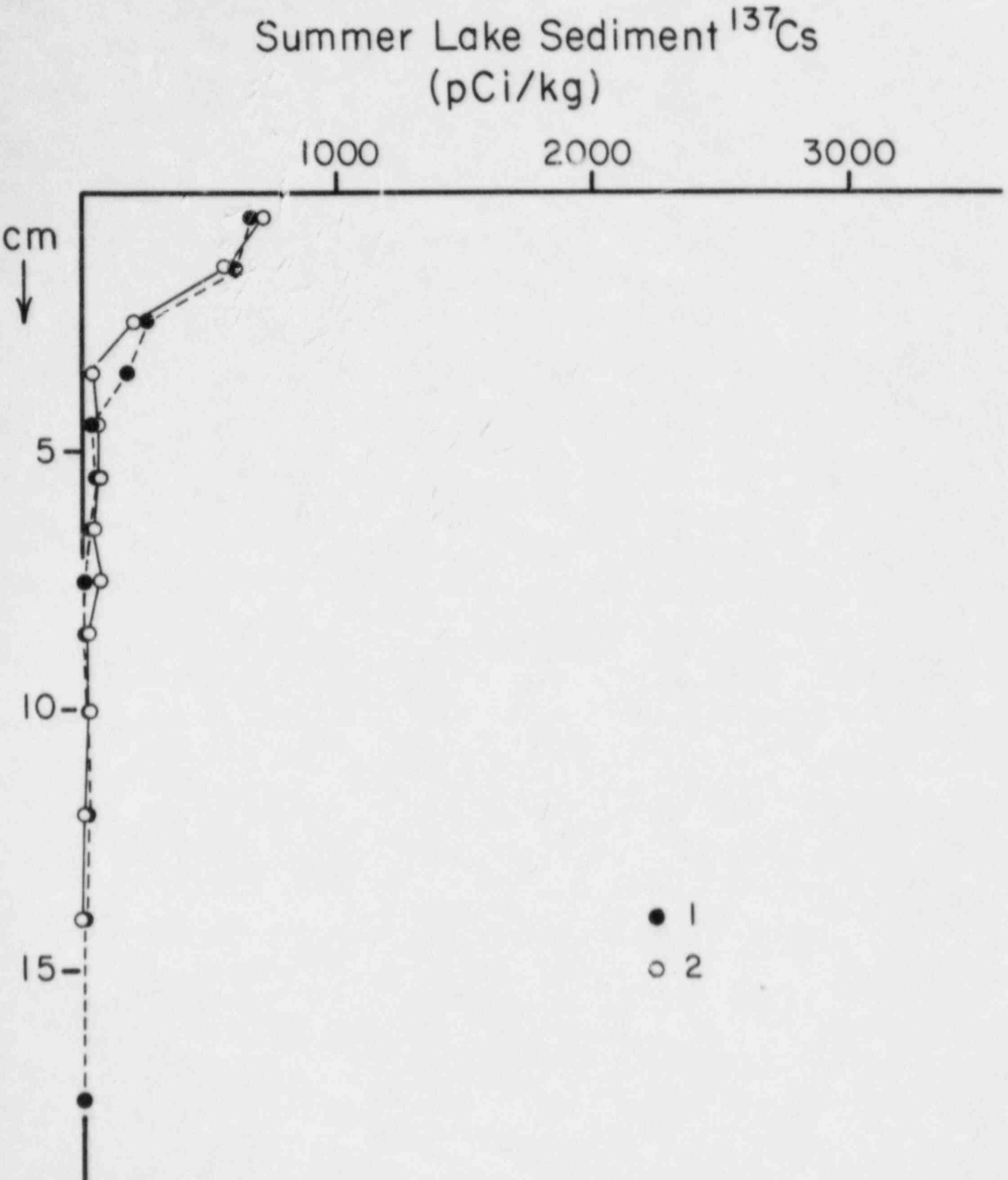


Figure 14.

Table 17

Mono Lake Sediment Plutonium

Depth (cm)	Alpha Spectrum #	$^{239\ 240}$Pu (pCi/kg)	^{238}Pu (pCi/kg)	^{238}Pu/$^{239,\ 240}$Pu (x100)	$^{239,\ 240}$Pu/^{137}Cs (x100)
Station #1-'78 (CN 1373)					
0-1	362	12.9±1.0	1.42±0.31	11.0±2.5	9.8
1-2	363	18.8±1.2	1.62±0.33	8.6±1.9	6.8
2-3	364	20.6±1.8	N.D.	N.D.	7.9
3-4	365	N.D.	N.D.	N.D.	-
4-5	366	N.D.	N.D.	N.D.	-
Station #2A-'79 (CN 1433)					
0-1	462	3.7±0.9	N.D.	N.D.	4.8
1-2	463	1.3±0.3	N.D.	N.D.	4.5
Station #3A-'79 (CN 1436)					
0-1	464	4.8±1.2	N.D.	N.D.	8.9
1-2	465	N.D.	N.D.	N.D.	-
Station #4A-'79 (CN 1439)					
0-1	466	13.5±2.7	N.D.	N.D.	9.0
1-2	467	N.D.	N.D.	N.D.	-
Station #5A-'80 (CN 1551)					
0-1	533	3.3±0.3	N.D.	N.D.	2.8
1-2	534	6.7±0.6	N.D.	N.D.	4.4
2-3	535	32.9±1.9	N.D.	N.D.	17
3-4	548	2.5±0.4	N.D.	N.D.	1.6
4-5	549	N.D.	N.D.	N.D.	-
5-7	550	N.D.	N.D.	N.D.	-
7-9	551	N.D.	N.D.	N.D.	-

Table 17 (Cont'd)

Mono Lake Sediment Plutonium

Depth (cm)	Alpha Spectrum #	$^{239\ 240}$Pu (pCi/kg)	^{238}Pu (pCi/kg)	^{238}Pu/^{239,240}Pu (x100)	^{239,240}Pu/^{137}Cs (x100)
Station #6A-'81 (CN 1677)					
0-1	698	7.0±0.6	N.D.	N.D.	4.8
1-2	699	14.4±0.8	0.60±0.15	4.2±1.1	12.
2-3	710	101.7±3.9	4.0 ±0.6	4.0±0.6	54.
3-4	711	10.9±1.9	N.D.	N.D.	7.2
Station #7-'81 (CN 1678)					
0-1	700	6.8±0.6	-	-	4.0
1-2	701	8.5±0.5	0.40±0.11	4.7±1.3	7.6
2-3	712	17.3±1.7	-	-	6.0
3-4	713	85.6±3.3	4.0 ±0.6	4.7±0.7	20.
Station #8A-'81 (CN 1679)					
0-1	702	9.7±0.6	0.49±0.14	5.0±1.4	8.8
1-2	703	18.3±1.3	0.59±0.22	3.2±1.2	(44.)
2-3	714	22.8±2.6	N.D.	N.D.	7.9
3-4	715	229.4±6.9	7.1 ±0.8	3.1±0.4	115
Station #9A-'81 (CN 1681)					
0-1	704	7.2±0.6	N.D.	N.D.	15.
1-2	705	13.7±1.0	0.6±0.2	4.4±0.4	14.
2-3	716	71.7±2.9	2.0±0.4	2.8±0.6	22.
3-4	717	14.0±1.5	N.D.	N.D.	19.

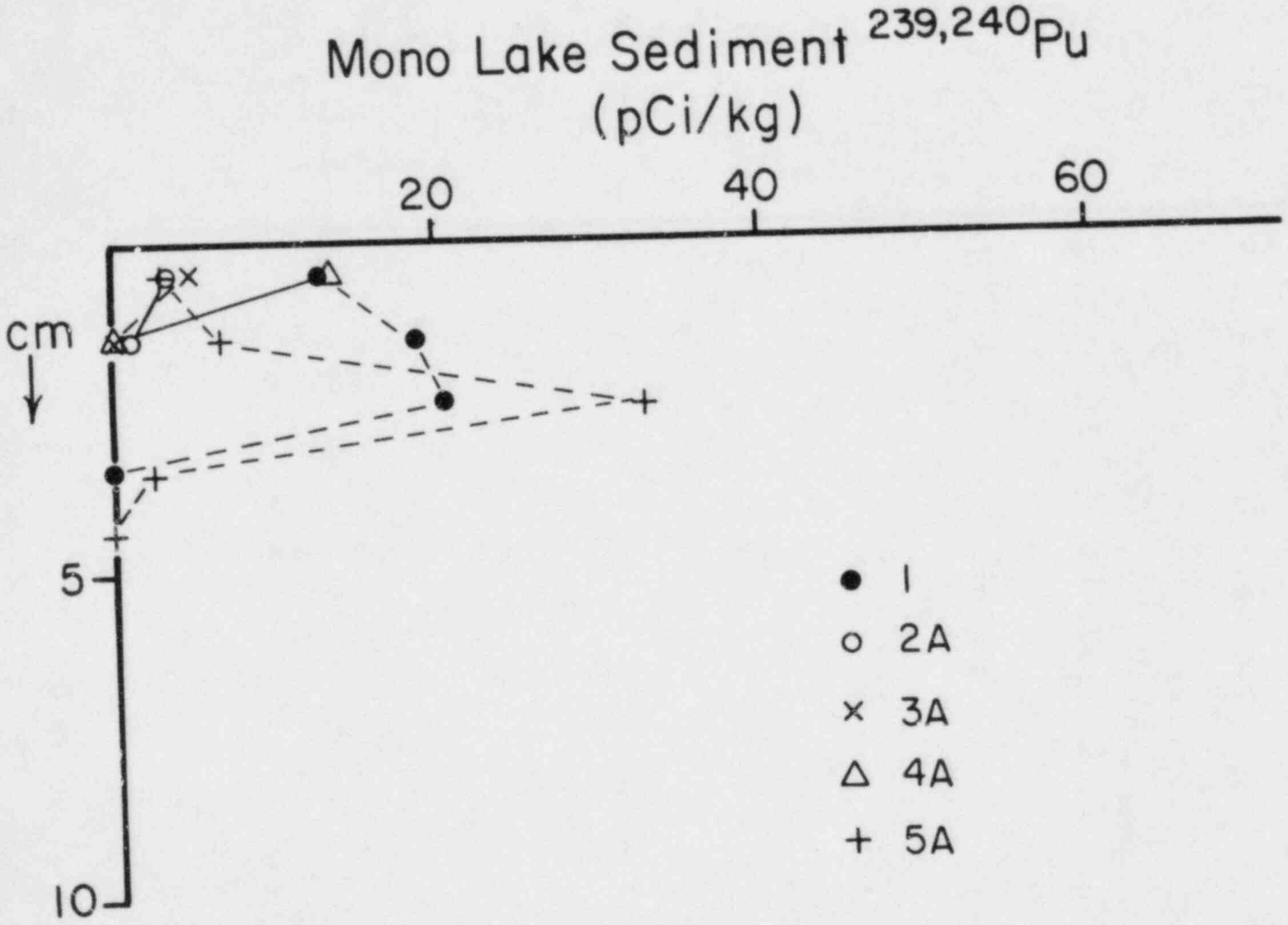


Figure 15.

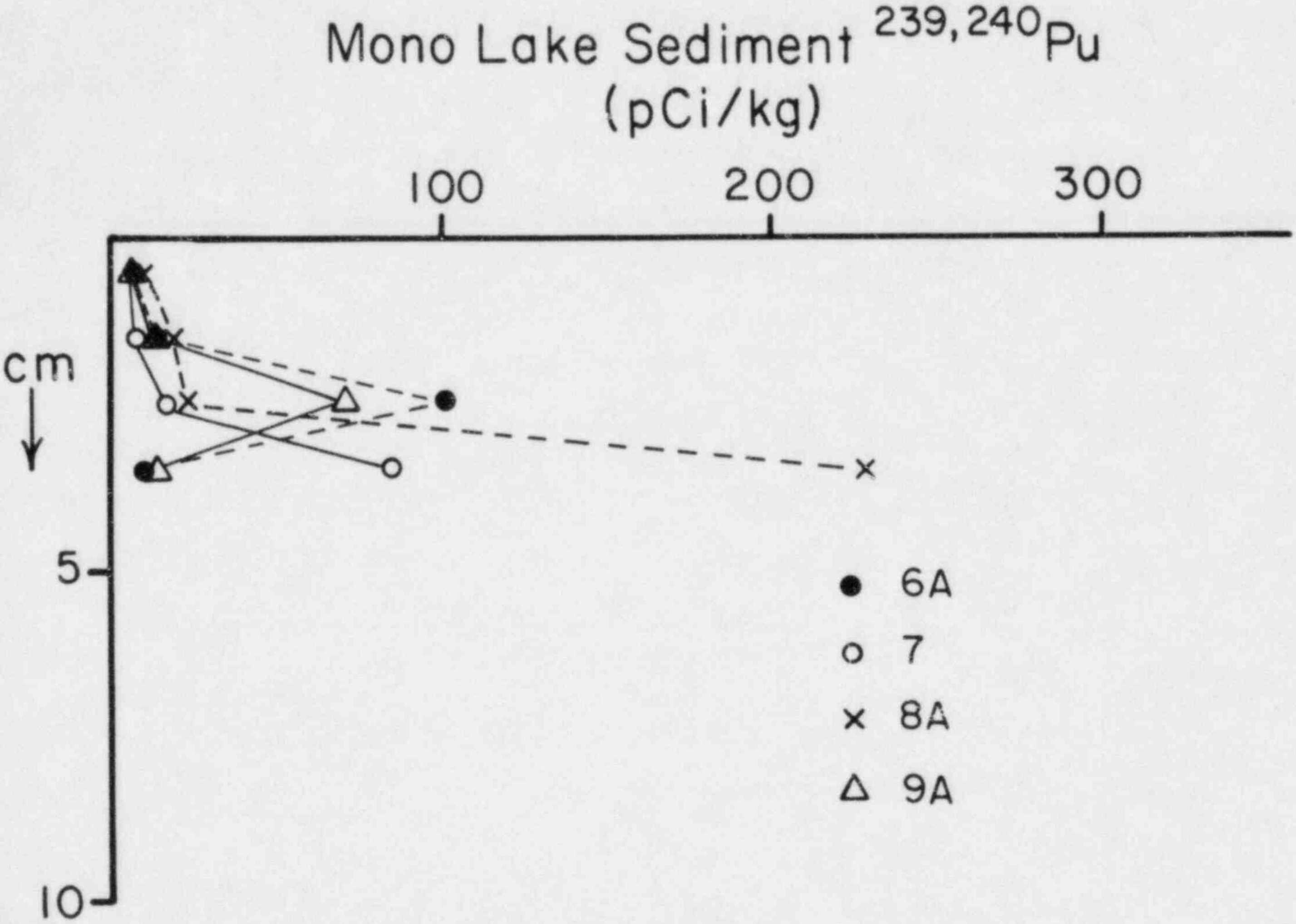

Mono Lake Sediment $^{239,240}Pu$
(pCi/kg)
100
200
300
cm
5
10
● 6A
o 7
× 8A
Δ 9A

Figure 16.

Table 18

Lake Abert Sediment Plutonium

Depth (cm)	Alpha Spectrum #	$^{239\ 240}$Pu (pCi/kg)	^{238}Pu (pCi/kg)	^{238}Pu/$^{239,\ 240}$Pu (x100)	$^{239,\ 240}$Pu/^{137}Cs (x100)
Station #2B (CN 1650)					
0-1	677	24.6 ±1.5	1.6 ±0.4	6.5±1.6	1.7
1-2	678	29.0 ±1.3	1.1 ±0.2	4.0±0.8	1.6
2-3	679	32.3 ±1.3	2.0 ±0.3	6.2±0.9	1.6
3-4	680	35.9 ±1.2	1.5 ±0.2	4.0±0.6	1.8
Station #4 (CN 1651)					
0-1	611	13.0 ±0.8	0.64±0.17	4.9±1.3	0.9
1-2	610	12.6 ±0.6	0.81±0.15	6.4±1.3	0.7
2-3	616	16.0 ±0.6	0.78±0.13	4.9±0.8	0.9
3-4	617	15.7 ±0.5	0.74±0.10	4.7±0.6	1.1
4-5	618	14.8 ±0.6	0.64±0.11	4.4±0.8	1.0
5-6	619	17.2 ±0.8	0.73±0.15	4.2±0.9	1.3
6-7	652	34.0 ±1.2	1.8 ±0.24	5.3±0.7	1.7
7-8	637	17.7 ±0.7	0.78±0.13	4.4±0.7	1.0
8-9	638	16.7 ±0.8	0.76±0.15	4.6±0.3	1.4
9-11	634	21.7 ±0.5	0.80±0.10	4.0±0.4	1.3
11-13	653	39.8 ±1.2	1.6 ±0.17	4.0±0.4	4.1
13-15	672	19.3 ±0.6	0.75±0.10	3.9±0.6	3.2
15-20	654	0.93±0.2	N.D.	N.D.	-
20-25	655	0.11±0.02	N.D.	N.D.	-

but had a much smaller range of concentrations. The shape of depth profiles of ^{239,240}Pu in L. Abert sediments (Figure 17) were quite similar to those for ^{137}Cs (Figure 10). Sediment concentrations of ^{239,240}Pu in Pyramid L., Goose L. and Summer L. (Table 19) were in the same range as those for most of the Mono L. and L. Abert sediments (10-60 pCi/kg), and depth profile shapes (Figures 18, 19 and 20) were similar to those for ^{137}Cs.

Samples of sediments from each of the six alkaline lakes were analyzed for uranium and thorium isotopes by alpha spectrometry (Table 20). Mono L. and Walker L. had the highest concentrations of ^{238}U, ^{234}U (2-3 pCi/g), ^{232}Th, ^{230}Th and ^{228}Th (0.5 to 1.5 pCi/g), while Goose L. and Summer L. had the lowest concentrations of U isotopes (0.1 to 0.4 pCi/g) and L. Abert the lowest values for Th isotopes (0.2 pCi/g). All of the alkaline lake sediments has ^{234}U/^{238}U ratios well above 1, as might be expected, but they also had ^{230}Th/^{234}U ratios of less than 1, with L. Abert being of the order of 0.1 (Table 21). It is difficult to understand why the activity ratio of ^{230}Th to ^{234}U was so low in these lake sediments.

^{214}Bi concentrations, as measured by gamma spectrometry, in eight Mono L. sediment cores were generally in the range of 0.5 to 1.5 pCi/g (Table 22), suggesting relatively uniform ^{226}Ra distributions in the fine-grained clay sediments of this lake. In L. Abert sediments, the average ^{214}Bi concentration was substantially lower (0.1 to 0.4 pCi/g), indicating much lower ^{226}Ra concentrations in its calcium carbonate-rich sediments (Table 23). ^{214}Bi data for three of the other four alkaline lakes (Table 24) were intermediate between L. Abert and Mono L., while Walker L. sediments were similar to the higher values typical of Mono L. ^{228}Ac concentrations in Mono L. were somewhat lower (0.2 to 1.0 pCi/g) than for ^{214}Bi (Table 25), indicating that these sediments had ^{228}Ra/^{226}Ra ratios of less than 1, while L. Abert sediment ^{228}Ac values (Table 26) were generally equal to or higher than ^{214}Bi concentrations. Sediments from the other alkaline lakes had ^{228}Ac concentrations (Table 27) comparable to Mono L. (0.2 to 1.0 pCi/g). Although the counting uncertainties were somewhat greater for the ^{228}Ac measurements than for ^{214}Bi, there does appear to be more variability in ^{228}Ra concentrations in the alkaline lake sediments than for ^{226}Ra.

^{40}K activities in eight Mono L. cores (Table 28) were 15-20 pCi/g, and relatively uniform with depth and between cores, suggesting large proportions of potassium-rich clay minerals in the lake sediments. L. Abert ^{40}K values were only about 6 pCi/g in two depth intervals of Core #1 (Table 29), indicating higher sand proportions near the stream inlet, but most of the depth segments in L. Abert cores were only slightly lower in ^{40}K than for Mono L. Pyramid L. had the lowest ^{40}K activities (6-12 pCi/g) for all the lakes, especially toward the bottom of cores (Table 30), suggesting a lower proportion of potassium-rich clay minerals than for the other lakes.

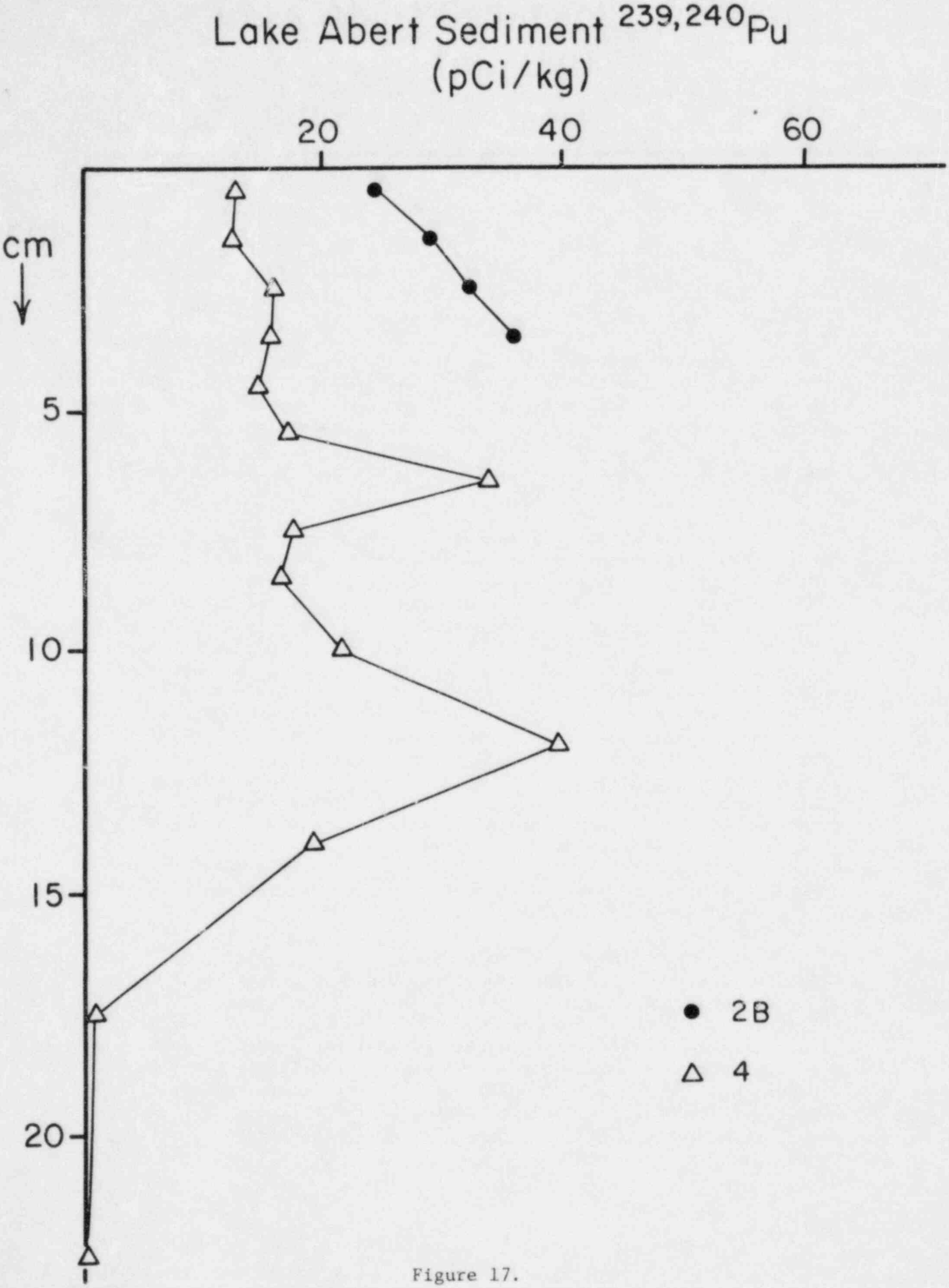


Figure 17.

Table 19

Alkaline Lake Sediment Plutonium

Depth (cm)	Alpha Spectrum #	^{239,240}Pu (pCi/kg)	^{238}Pu (pCi/kg)	^{238}Pu/^{239,240}Pu (x100)	^{239,240}Pu/^{137}Cs (x100)
Walker L. Station #1 (CN 1442)					
0-1	468	11.7 ±2.1	-	-	0.6
1-2	469	29.0 ±4.5	-	-	1.3
2-3	491	5.9 ±1.4	-	-	1.4
Pyramid L. Station #1 (CN 1685)					
0-1	673	26.7 ±2.1	2.1 ±0.6	7.9±2.2	3.1
1-2	674	33.0 ±1.8	1.8 ±0.4	5.4±1.3	4.3
2-3	675	62.8 ±1.9	4.7 ±0.5	7.6±0.8	4.5
3-4	576	40.3 ±1.6	3.6 ±0.4	8.9±1.2	7.3
4-5	693	2.4 ±0.3	N.D.	N.D.	-
5-6	694	4.3 ±0.4	N.D.	N.D.	-
Goose L. Station #2 (CN 1684)					
0-1		-	-	-	-
1-2	-	-	-	-	-
2-3	684	25.8 ±1.1	0.89±0.17	3.4±0.7	3.6
3-4	-	-	-	-	-
4-5	-	-	-	-	-
5-6	695	12.2 ±0.6	0.56±0.11	4.6±1.0	2.1
6-7	685	23.5 ±0.8	0.77±0.12	3.3±0.5	4.1
7-8	-	-	-	-	-
8-9	-	-	-	-	-
9-11	686	7.7 ±0.3	0.48±0.06	6.3±0.8	2.0
11-13	-	-	-	-	-
13-15	696	1.4 ±0.1	0.08±0.02	5.7±1.6	5.4
Summer L. Station #2 (CN 1689)					
0-1	665	12.9 ±1.2	N.D.	N.D.	1.8
1-2	661	10.4 ±0.8	0.50±0.17	4.8±1.6	1.8
2-3	669	4.0 ±0.8	N.D.	N.D.	1.9
3-4	670	0.68±0.20	N.D.	N.D.	1.7
4-5	666	N.D.	N.D.	N.D.	-
5-6	662	0.36±0.08	N.D.	N.D.	0.6
6-7	671	N.D.	N.D.	N.D.	-
7-8	667	N.D.	N.D.	N.D.	-
8-9	-	-	-	-	-
9-11	663	0.23±0.05	N.D.	N.D.	-
11-13	668	0.26±0.06	N.D.	N.D.	-
13-15	664	N.D.			

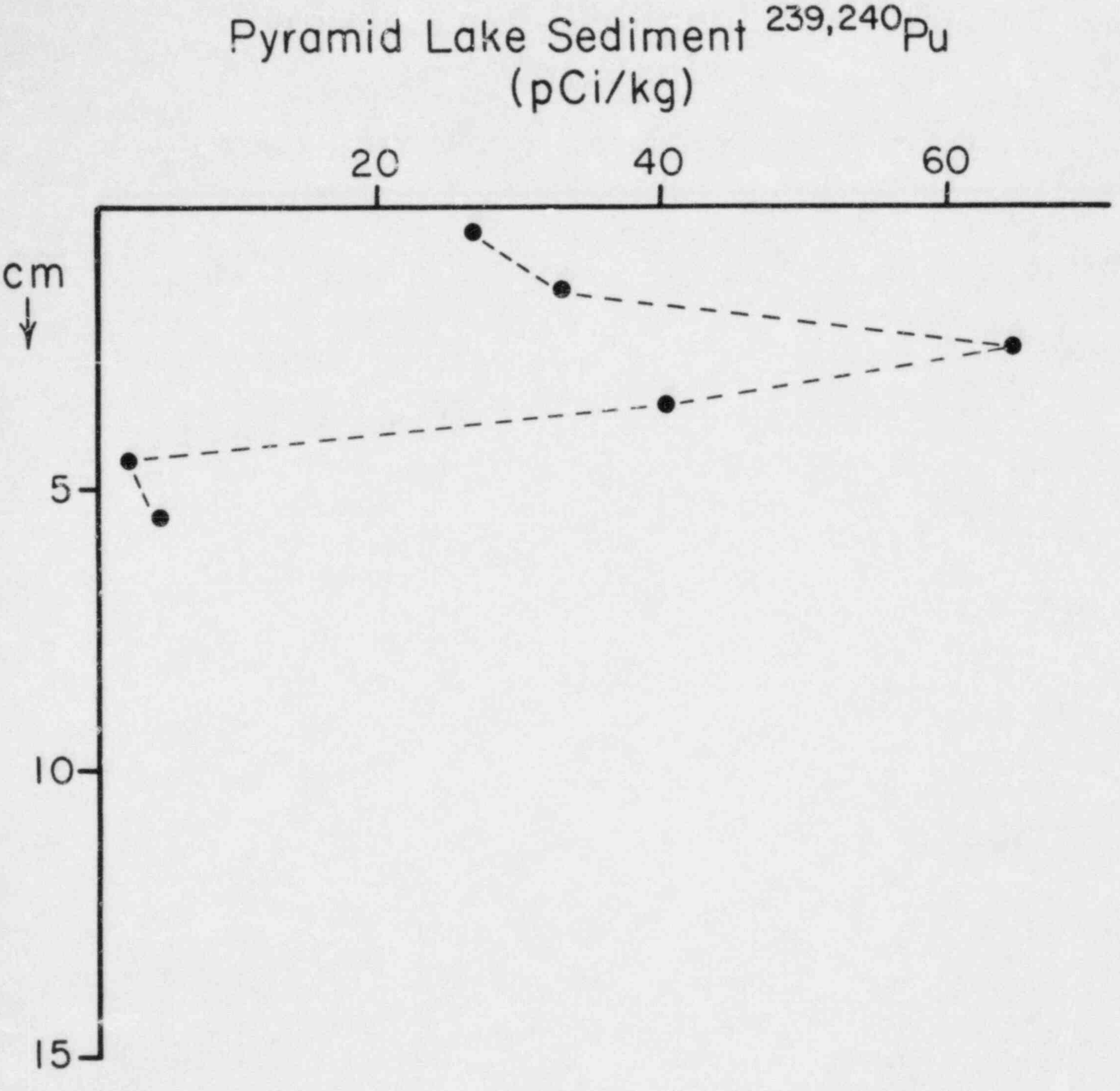


Figure 18.

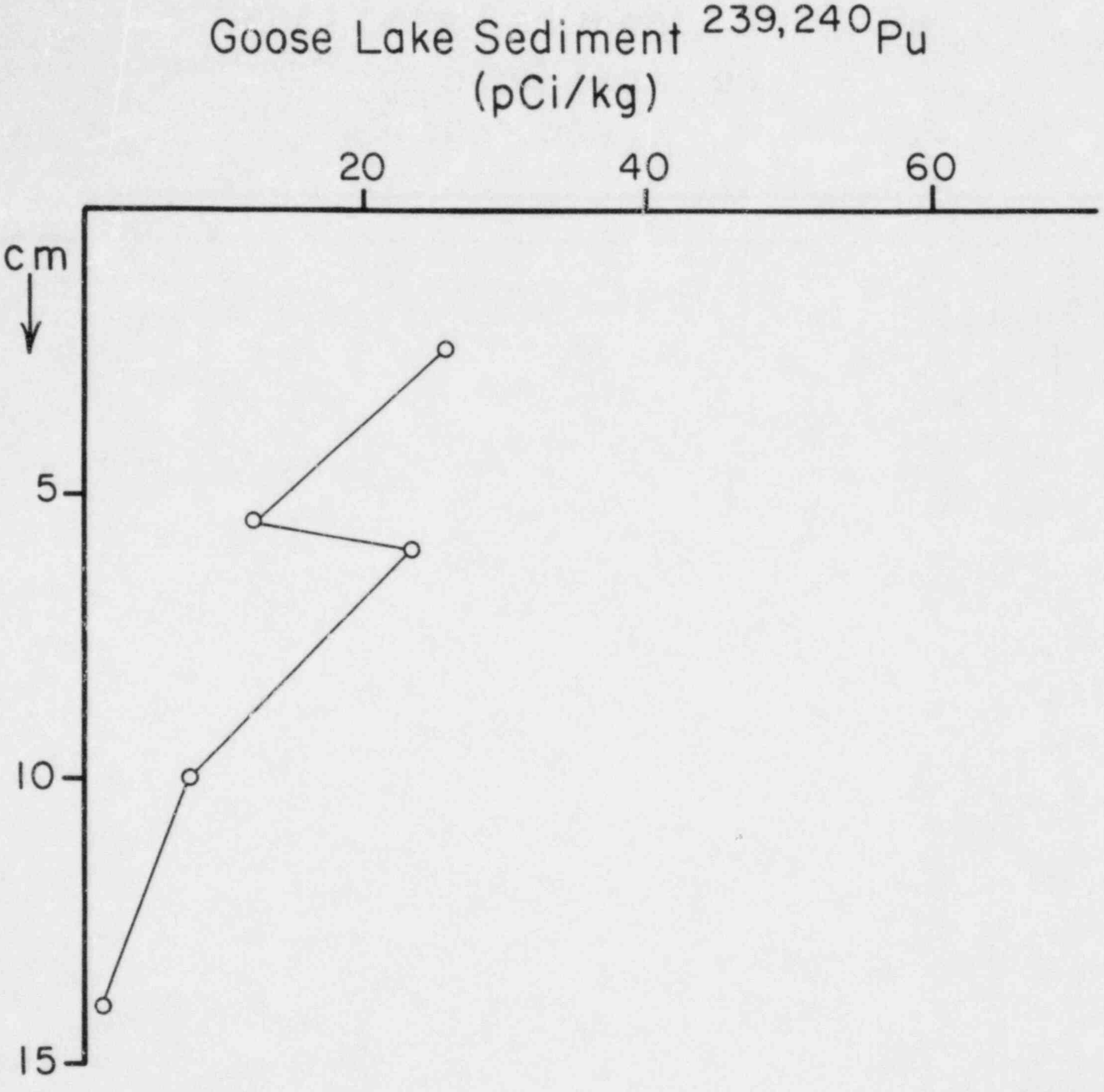


Figure 19.

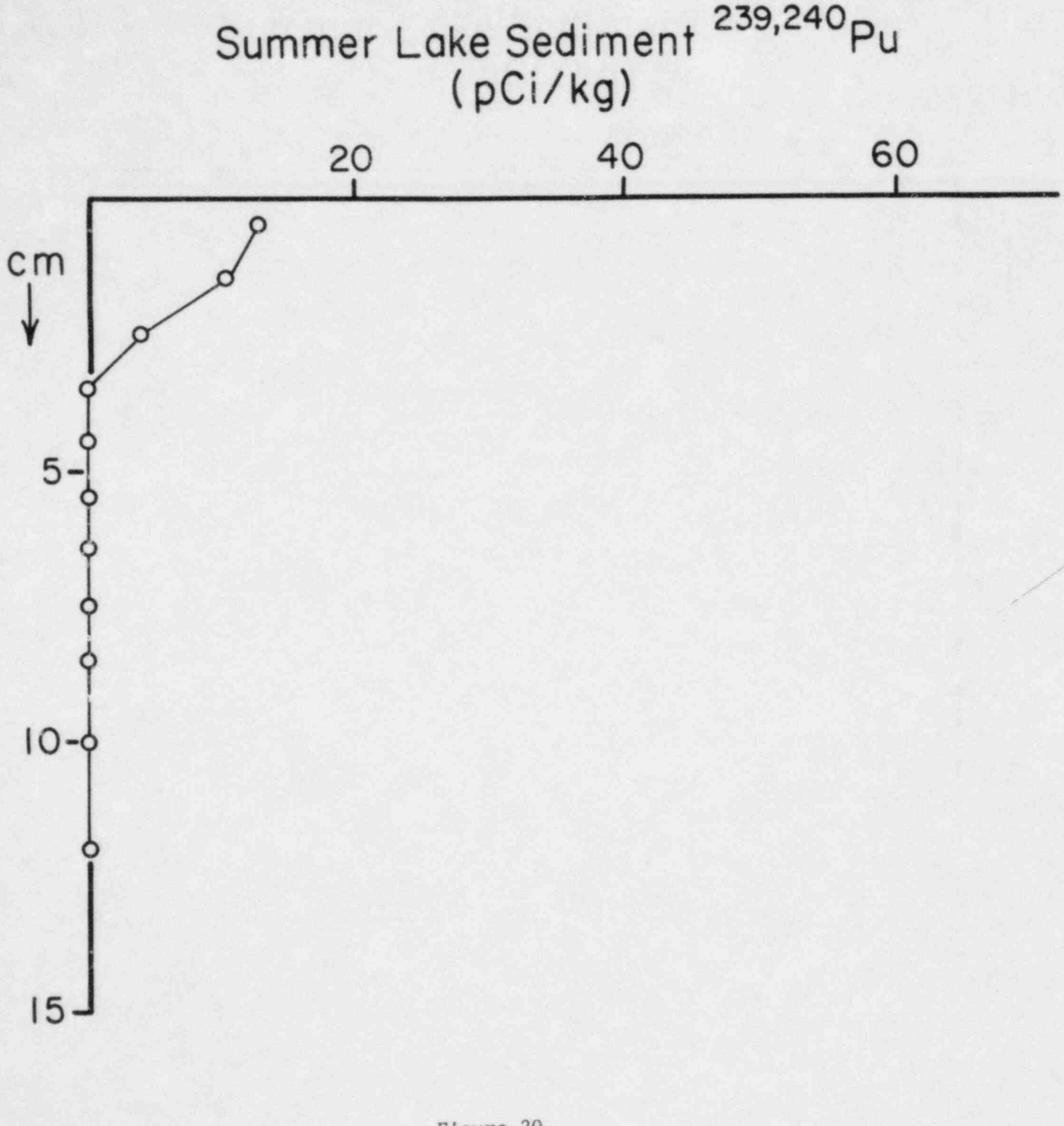


Figure 20.

Table 20

Uranium and Thorium in Alkaline Lake Sediments

Lake Sampled	Laboratory Control #	Alpha Spectrum #	Depth Interval (cm)	^{238}U (pCi/kg)	^{234}U (pCi/kg)	^{232}Th (pCi/kg)	^{230}Th (pCi/kg)	^{228}Th (pCi/kg)
Mono L. 1981-6B	1675 M	739	15-20	2350±100	2880±120	450±20	710±27	415±19
L. Abert 1981-4	1651 O	743	25-30	1090± 30	2370± 60	193± 9	205± 9	169± 8
Walker L. 1979	1442 M	747	15-20	2730±110	3500±130	1371±47	1526±51	1256±43
Pyramid L. 1981-1	1685 M	740	20-25	1620± 40	2230± 50	399±16	468±17	384±15
Goose L. 1981-2	1684 M	741	15-20	124± 9	336± 15	242±10	281±11	251±10
Summer L. 1981-2	1689 M	742	15-20	232± 12	443± 18	326± 9	284± 8	311± 9

Table 21

Uranium and Thorium Isotope Ratios in Alkaline Lake Sediments
(Alpha Spectrometry)

Lake Sampled	Depth Interval (cm)	$^{234}U/^{238}U$	$^{230}Th/^{232}Th$	$^{228}Th/^{232}Th$	$^{230}Th/^{234}U$
Mono L.	15-20	1.23±0.07	1.58±0.09	0.92±0.05	0.25 ±0.02
L. Abert	25-30	2.17±0.08	1.06±0.06	0.88±0.06	0.086±0.004
Walker L.	15-20	1.28±0.06	1.11±0.05	0.92±0.04	0.44 ±0.03
Pyramid L.	20-25	1.57±0.06	1.17±0.06	0.96±0.05	0.21 ±0.01
Goose L.	15-20	2.71±0.23	1.16±0.07	1.04±0.06	0.84 ±0.05
Summer L.	15-20	1.91±0.13	0.87±0.03	0.95±0.04	0.64 ±0.04

Table 22

Bismuth - 214 in Mono Lake Sediments (pCi/g)
(Gamma Spectrometry)

Station #	5A	6A	6B	7	8A	8B	9A	9B
Depth (cm)	CN 1551	CN 1677	CN 1675	CN 1678	CN 1679	CN 1680	CN 1681	CN 1682
0-1	0.06±0.01	0.97±0.16	0.87±0.06	1.11±0.13	1.24±0.13	1.03±0.12	0.87±0.11	0.96±0.10
1-2	0.07±0.01	0.77±0.13	0.74±0.04	1.16±0.13	0.85±0.16	1.00±0.18	0.94±0.17	0.95±0.10
2-3	0.05±0.01	0.92±0.14	0.46±0.08	1.22±0.12	0.91±0.13	0.70±0.17	0.94±0.15	1.28±0.12
3-4	0.11±0.01	0.64±0.16	0.88±0.13	0.68±0.15	1.12±0.20	1.03±0.18	0.95±0.16	0.97±0.10
4-5	0.05±0.01	0.57±0.17	0.71±0.11	0.88±0.13	0.75±0.14	0.94±0.16	0.95±0.10	1.04±0.13
5-6	0.08±0.01	0.67±0.17	0.87±0.20	1.14±0.16	0.87±0.16	0.74±0.16	1.86±0.21	1.12±0.14
6-7		0.64±0.15	2.04±0.18	0.93±0.15	0.92±0.14	0.76±0.15	1.08±0.13	1.65±0.14
7-8	0.06±0.01	0.88±0.12	1.03±0.14	2.32±0.24	1.98±0.13		0.96±0.17	0.89±0.14
8-9		1.70±0.14	0.87±0.14	1.53±0.15			1.00±0.05	0.81±0.17
9-11	0.04±0.01	1.29±0.12	0.85±0.12	1.20±0.09			1.12±0.11	1.12±0.10
11-13	0.05±0.01	1.11±0.12	0.81±0.04	0.96±1.10			1.02±0.10	1.02±0.12
13-15	0.05±0.01	1.05±0.13	0.88±0.11	0.92±0.11			1.00±0.12	
15-20	0.39±0.03		0.83±0.07	1.10±0.07			0.72±0.06	
20-25	0.38±0.02		0.66±0.05	0.88±0.06			0.84±0.07	
25-30	0.48±0.04			0.94±0.05			1.24±0.06	

Table 23

Bismuth - 214 in Lake Abert Sediments (pCi/g)
(Gamma Spectrometry)

Station #	1	1B	2A	2B	3A	3B	4
Depth (cm)	CN 1651	CN 1690	CN 1691	CN 1692	CN 1693	CN 1694	CN 1695
0-1	0.26±0.29	-1.14±0.50	0.22±0.14	0.70±0.14	0.10±0.06	0.20±0.13	0.53±0.23
1-2	0.06±0.09	0.31±0.16	0.17±0.20	0.37±0.11	0.11±0.14	0.11±0.08	0.12±0.15
2-3	0.07±0.09	0.26±0.13	0.30±0.19	0.37±0.18	0.42±0.09	0.33±0.09	0.22±0.11
3-4	0.06±0.08	0.34±0.11	0.03±0.21	0.06±0.11	0.16±0.08	0.23±0.11	0.47±0.15
4-5	0.06±0.11	0.27±0.16	0.20±0.14	0.22±0.09	0.10±0.08	0.08±0.12	0.24±0.12
5-6	0.08±0.10	0.24±0.14	0.01±0.12	-0.84±0.05	0.13±0.11	0.31±0.11	0.28±0.18
6-7	0.32±0.10	0.25±0.13	0.16±0.10	0.50±0.16	0.39±0.11	0.32±0.12	0.39±0.11
7-8	0.15±0.10	0.17±0.14	0.11±0.08	0.27±0.09	0.37±0.13	0.25±0.12	0.19±0.11
8-9	0.40±0.13	0.28±0.12	0.28±0.11	0.20±0.09	0.16±0.14	0.33±0.12	0.33±0.12
9-11	0.35±0.09	0.21±0.09	0.31±0.08	0.26±0.06	0.23±0.06	0.32±0.14	0.30±0.09
11-13	0.25±0.08	0.23±0.07	0.28±0.07	0.23±0.06		0.23±0.05	0.23±0.06
13-15	0.15±0.06	0.28±0.08	0.39±0.05	0.21±0.05			0.38±0.04
15-20	0.34±0.04	0.36±0.04		0.17±0.03			0.18±0.04
20-25	0.16±0.07	0.33±0.04					0.17±0.03
25-30							0.24±0.03
30-35							0.29±0.03
35-40							0.34±0.05
40-45							0.29±0.05
45-50							0.21±0.04

Table 24

Bismuth - 214 in Alkaline Lake Sediments (pCi/g)
(Gamma Spectrometry)

Lake	Walker L.	Pyramid L. #1	Pyramid L. #2	Pyramid L. #3	Goose L. #1	Goose L. #2	Summer L. #1	Summer L. #2
Depth (cm)	CN 1697	CN 1685	CN 1686	CN 1687	CN 1683	CN 1684	CN 1688	CN 1689
0-1	1.11±0.12	0.73±0.34	0.82±0.19	0.65±0.34	0.41±0.08	0.50±0.38	0.22±0.22	0.22±0.18
1-2	1.61±0.10	1.29±0.23	0.61±0.22	0.79±0.22	0.44±0.13	0.46±0.15	0.44±0.07	0.30±0.06
2-3	2.22±0.10	0.67±0.21	0.45±0.20	0.87±0.18	0.45±0.07	0.34±0.08	0.50±0.06	0.51±0.05
3-4		0.69±0.18	0.84±0.20	1.83±0.26	0.45±0.07	0.54±0.06	0.42±0.08	0.40±0.06
4-5	1.75±0.08	0.82±0.19	0.85±0.14	0.58±0.16	0.46±0.06	0.43±0.07	0.46±0.07	0.44±0.07
5-6		0.69±0.17	0.45±0.16	0.81±0.17	0.50±0.07	0.42±0.07	0.50±0.09	0.37±0.07
6-7	1.56±0.08	0.43±0.20	0.71±0.14	0.79±0.18	0.46±0.05	0.50±0.08	0.49±0.07	0.59±0.06
7-8		0.55±0.16	0.58±0.17	0.75±0.18	0.39±0.06	-0.08±0.03	0.42±0.08	0.47±0.07
8-9	1.19±0.09	0.48±0.17	0.78±0.17	0.56±0.17	0.36±0.05	0.45±0.08	0.48±0.06	0.39±0.06
9-11		0.64±0.10	0.64±0.12	0.34±0.10	0.37±0.05	0.44±0.04	0.59±0.06	0.46±0.06
11-13		0.70±0.12			0.36±0.04	0.42±0.04	0.48±0.05	0.49±0.05
13-15		0.72±0.10			0.34±0.04	0.38±0.04	0.49±0.06	0.44±0.06
15-20		0.61±0.08			0.27±0.04	0.36±0.04	0.40±0.04	
20-25							0.50±0.05	
*	2.00±0.30							

*Surface floc (above 0-1 cm)

Table 25

Actinium-228 in Mono Lake Sediments (pCi/g)
(Gamma Spectrometry)

Station #	5A	6A	6B	7	8A	8B	9A	9B
Depth (cm)	CN 1551	CN 1677	CN 1675	CN 1678	CN 1679	CN 1680	CN 1681	CN 1682
0-1	0.26±0.12	0.82±0.27	0.35±0.09	0.45±0.17	0.95±0.23	0.93±0.21	0.89±0.20	0.22±0.13
1-2	0.48±0.13	0.91±0.24	0.32±0.05	0.50±0.18	0.58±0.28	1.17±0.32	1.24±0.31	0.45±0.14
2-3	0.39±0.10	0.39±0.25	0.23±0.11	0.29±0.16	0.81±0.23	0.87±0.32	0.77±0.27	0.61±0.17
3-4	0.01±0.28	1.04±0.30	0.12±0.18	0.47±0.21	0.16±0.33	0.98±0.34	0.60±0.28	0.61±0.15
4-5	0.47±0.13	0.26±0.32	0.43±0.16	0.46±0.20	0.46±0.26	0.90±0.29	0.67±0.19	0.39±0.17
5-6	0.46±0.05	0.84±0.31	0.48±0.28	0.56±0.23	-	0.79±0.30	1.40±0.37	0.48±0.21
6-7		0.40±0.28	0.75±0.24	0.23±0.21	0.50±0.29	0.21±0.28	0.47±0.22	0.63±0.20
7-8	0.44±0.05	0.79±0.22	0.31±0.20	1.41±0.32	0.56±0.25		0.86±0.30	0.97±0.25
8-9		1.35±0.25	0.11±0.20	0.75±0.21	1.27±0.21		0.63±0.27	1.11±0.31
9-11	0.28±0.06	0.99±0.19	0.56±0.18	0.55±0.12			0.78±0.19	0.68±0.16
11-13	0.28±0.10	0.88±0.20	0.56±0.07	0.40±0.13			0.82±0.18	0.66±0.20
13-15	0.22±0.10	1.21±0.23	0.33±0.16	0.41±0.14			1.05±0.20	
15-20	0.30±0.09		0.34±0.09	0.60±0.09			0.51±0.10	
20-25	0.34±0.06		0.29±0.06	0.42±0.07			0.59±0.11	
25-30	0.21±0.11			0.45±0.06			0.74±0.08	

Table 26

Actinium-228 in Lake Abert Sediments (pCi/g)
(Gamma Spectrometry)

Station #	1	1B	2A	2B	3A	3B	4
Depth (cm)	CN 1651	CN 1690	CN 1691	CN 1692	CN 1693	CN 1694	CN 1695
0-1	0.13±0.41	0.19±0.98	0.53±0.26	-0.29±0.20	0.15±0.10	0.29±0.21	0.42±0.44
1-2	0.26±0.14	0.02±0.29	0.21±0.38	0.24±0.17	0.62±0.27	0.08±0.12	0.16±0.26
2-3	0.66±0.18	0.30±0.24	0.14±0.34	0.20±0.13	0.38±0.13	0.09±0.13	0.13±0.17
3-4	0.28±0.13	0.43±0.20	0.71±0.39	0.25±0.17	0.24±0.13	0.02±0.17	0.33±0.28
4-5	0.23±0.19	0.01±0.27	0.35±0.25	0.03±0.13	0.05±0.13	0.29±0.20	0.67±0.23
5-6	0.55±0.19	0.40±0.25	0.12±0.21	0.26±0.13	0.33±0.20	0.60±0.21	0.58±0.35
6-7	0.29±0.19	0.57±0.24	0.07±0.18	0.19±0.29	0.54±0.21	0.50±0.23	0.07±0.17
7-8	0.34±0.19	0.06±0.24	0.38±0.14	0.13±0.13	0.35±0.20	0.09±0.21	0.46±0.20
8-9	0.17±0.25	0.26±0.22	0.16±0.16	0.23±0.14	0.22±0.26	0.24±0.22	0.43±0.22
9-11	0.40±0.15	0.28±0.15	0.03±0.19	0.30±0.11	0.28±0.10	0.48±0.25	0.52±0.15
11-13	0.25±0.12	0.34±0.13	0.21±0.14	0.39±0.10		0.34±0.08	0.38±0.10
13-15	0.16±0.10	0.32±0.14	0.11±0.12	0.28±0.09			0.10±0.06
15-20	0.20±0.06	0.26±0.06	0.44±0.08	0.33±0.06			0.22±0.06
20-25	0.30±0.12	0.29±0.07					0.21±0.05
25-30							0.17±0.04
30-35							0.26±0.05
35-40							0.19±0.08
40-45							0.40±0.09
45-50							0.33±0.06

Table 27

Actinium-228 in Alkaline Lake Sediments (pCi/g)
(Gamma Spectrometry)

Lake	Walker L.	Pyramid L. #1	Pyramid L. #2	Pyramid L. #3	Goose L. #1	Goose L. #2	Summer L. #1	Summer L. #2
Depth (cm)	CN 1697	CN 1685	CN 1686	CN 1687	CN 1683	CN 1684	CN 1688	CN 1689
0-1	1.16±0.19	1.11±0.65	0.24±0.29	1.75±0.66	0.43±0.13	0.10±0.11	1.69±0.42	0.17±0.27
1-2	1.34±0.16	0.78±0.42	1.53±0.42	1.11±0.42	0.35±0.23	0.65±0.28	0.24±0.13	0.50±0.10
2-3	1.25±0.14	1.33±0.41	0.97±0.37	1.38±0.33	0.33±0.11	0.75±0.14	0.73±0.11	0.51±0.07
3-4		0.92±0.33	0.95±0.37	-	0.61±0.11	0.46±0.10	0.32±0.13	0.33±0.09
4-5	1.15±0.12	0.76±0.33	0.85±0.25	1.11±0.31	0.50±0.10	0.28±0.12	0.38±0.12	0.36±0.11
5-6		0.90±0.32	0.89±0.29	1.24±0.33	0.14±0.12	0.50±0.12	0.41±0.15	0.54±0.11
6-7	1.22±0.11	0.71±0.36	0.17±0.25	0.58±0.31	0.38±0.09	0.53±0.14	0.47±0.12	0.44±0.10
7-8		0.82±0.29	0.36±0.32	1.14±0.34	0.54±0.10	-	0.52±0.14	0.38±0.11
8-9	0.97±0.14	1.17±0.33	0.73±0.30	0.61±0.32	0.32±0.08	0.60±0.13	0.57±0.11	0.30±0.09
9-11		0.67±0.17	0.56±0.22	0.36±0.19	0.30±0.08	0.42±0.07	0.52±0.09	0.45±0.09
11-13		0.49±0.21			0.30±0.07	0.44±0.06	0.46±0.08	0.42±0.07
13-15		0.52±0.17			0.32±0.07	0.38±0.06	0.36±0.10	0.38±0.09
15-20		0.42±0.12			0.32±0.07	0.27±0.07	0.37±0.06	
20-25							0.58±0.08	
*	0.50±0.54							

*Surface floc (above 0-1 cm)

Table 28

Potassium-40 in Mono Lake Sediments (pCi/g)
(Gamma Spectrometry)

Station #	5A	6A	6B	7	8A	8B	9A	9B
Depth (cm)	CN 1551	CN 1677	CN 1675	CN 1678	CN 1679	CN 1680	CN 1681	CN 1682
0-1	16.1±0.9	17.7±1.6	16.6±0.6	20.0±1.1	19.4±1.3	21.6±1.2	17.5±1.1	13.7±0.8
1-2	15.6±0.9	19.4±1.3	13.4±0.4	18.0±1.2	17.9±1.7	17.8±1.7	22.7±1.8	18.8±0.9
2-3	15.6±0.8	17.6±1.5	15.0±0.8	18.5±1.1	20.6±1.4	19.0±1.8	16.6±1.5	15.2±0.9
3-4	14.5±0.9	21.0±1.7	16.8±1.2	15.6±1.3	18.9±2.0	19.6±1.9	14.6±1.6	17.4±0.9
4-5	14.9±0.9	18.6±1.8	15.1±0.9	15.7±1.2	19.7±1.5	20.7±1.7	19.1±1.1	15.5±1.1
5-6	15.4±0.6	16.4±1.8	15.3±1.7	16.3±1.3	-	21.5±1.7	17.0±1.9	17.1±1.2
6-7		18.5±1.6	15.0±1.3	17.8±1.3	18.8±1.7	17.7±1.6	19.1±1.3	14.9±1.2
7-8	15.3±0.6	19.3±1.3	17.8±1.2	17.8±1.7	17.0±1.4	-	21.0±1.7	16.8±1.4
8-9		18.5±1.3	18.6±1.3	18.5±1.2	17.0±1.1	-	21.4±1.6	19.7±1.7
9-11	14.1±0.6	19.8±1.1	18.0±1.2	18.1±0.9	-	-	19.5±1.2	18.7±1.0
11-13		18.2±1.2	17.3±0.3	18.7±0.9	-	-	18.0±1.1	14.3±1.1
13-15		17.3±1.3	17.5±1.0	15.5±0.9	-	-	20.4±1.2	-
15-20		-	16.0±0.7	18.3±0.7	-	-	16.1±0.7	-
20-25		-	11.0±0.5	14.0±0.6	-	-	18.4±0.8	-
25-30		-	-	15.8±0.5	-	-	16.9±0.6	-

Table 29

Potassium-40 in Lake Abert Sediments (pCi/g)
(Gamma Spectrometry)

Station #	1	1B	2A	2B	3A	3B	4
Depth (cm)	CN 1651	CN 1690	CN 1691	CN 1692	CN 1693	CN 1694	CN 1695
0-1	-	13.5±6.1	16.9±1.8	9.4±1.0	-	-	18.3±2.5
1-2	-	10.7±1.9	13.6±2.5	14.3±0.9	15.6±1.5	-	-
2-3	6.3±0.9	10.3±1.6	17.6±2.3	13.3±1.6	-	-	-
3-4	-	14.8±1.3	15.0±2.5	15.5±1.2	-	-	15.8±1.6
4-5	15.9±1.2	17.1±1.9	16.0±1.7	15.0±0.9	-	13.5±1.4	12.2±1.3
5-6	11.4±1.1	16.7±1.7	15.8±1.5	16.1±0.9	16.3±1.2	17.0±1.4	14.2±1.9
6-7	17.2±1.1	13.9±1.6	15.8±1.3	16.7±1.7	18.0±1.2	16.5±1.5	-
7-8	13.8±1.1	17.3±1.6	6.2±1.0	14.1±0.8	-	13.7±1.3	17.8±1.2
8-9	15.4±1.4	16.8±1.5	15.8±1.3	13.2±0.9	14.0±1.5	12.8±1.5	11.6±1.2
9-11	17.2±1.0	15.5±1.1	15.2±1.0	15.2±0.7	15.3±0.6	13.5±1.6	16.9±0.9
11-13	-	15.1±0.9	16.5±0.9	15.9±0.7		12.9±0.6	13.9±0.6
13-15	6.6±0.6	14.8±1.0	14.7±0.6	17.7±0.6			-
15-20	14.8±0.5	14.6±0.5		14.3±0.5			-
20-25	14.5±0.8	15.3±0.5					-
25-30							-
30-35							15.7±0.5
35-40							15.2±0.7
40-45							14.9±0.6
45-50							15.4±0.5

Table 30

Potassium-40 in Alkaline Lake Sediments (pCi/g)
(Gamma Spectrometry)

Lake Depth (cm)	Walker L. CN 1697	Pyramid L. #1 CN 1685	Pyramid L. #2 CN 1686	Pyramid L. #3 CN 1687	Goose L. #1 CN 1683	Goose L. #2 CN 1684	Summer L. #1 CN 1688	Summer L. #2 CN 1689
0-1	14.0±1.0	14.5±3.3	13.7±1.5	15.1±3.3	14.0±0.8	17.2±3.7	18.5±2.2	17.0±1.7
1-2	16.3±0.9	15.4±2.2	12.1±2.1	12.8±2.1	12.8±1.3	14.2±1.5	17.2±0.8	18.0±0.8
2-3	20.6±0.8	17.4±2.1	8.3±1.9	18.4±2.0	12.0±0.7	15.5±0.9	18.2±0.7	18.4±0.6
3-4		16.3±1.8	12.5±2.0	38.3±3.0	12.9±0.7	13.5±0.6	19.4±0.9	15.8±0.7
4-5	18.2±0.8	15.3±1.7	9.0±1.3	9.8±1.6	14.2±0.7	14.4±0.8	18.5±0.9	15.6±0.8
5-6		9.7±1.9	9.0±1.6	9.2±1.7	10.7±0.7	13.1±0.8	19.3±1.0	16.6±0.8
6-7	16.3±0.7	14.1±1.5	7.7±1.3	12.8±1.7	11.9±0.6	14.6±0.9	19.1±0.8	17.0±0.7
7-8		10.4±1.7	7.2±1.9	5.8±1.8	11.0±0.6	-	16.6±0.9	15.4±0.8
8-9	13.0±0.9	11.2±1.0	7.6±1.9	4.2±1.6	10.5±0.5	12.7±0.8	16.2±0.7	14.0±0.7
9-11		12.3±1.2	8.4±1.4	2.5±1.0	9.5±0.5	10.8±0.4	15.7±0.6	16.0±0.6
11-13		10.6±0.9			8.2±0.4	9.6±0.4	15.9±0.6	15.0±0.5
13-15		10.2±0.7			8.9±0.4	8.8±0.4	17.1±0.7	17.0±0.7
15-20					7.8±0.4	7.8±0.4	14.5±0.5	
20-25							17.5±0.6	
*	20.2±2.8							

*Surface floc (above 0-1 cm)

6.0 CHEMICAL DATA FOR ALKALINE LAKES

All six of the lakes discussed here were known to have high proportions of bicarbonate and carbonate ions in solution relative to saline waters more similar to seawater composition. Table 31 provides representative published data for the chemical composition of these lakes, generally for somewhat higher lake levels than existed during 1981 when our samples were collected for radionuclide measurements. Based on this published data Mono L. had the highest alkalinity, Goose L. and Pyramid L. the lowest, and all of the lakes had high pH values (8.9-9.7).

Samples from four of the lakes were equilibrated for pCO_2 measurements in the field (Table 32) at the ambient temperature of the sample. When corrected to the same temperature, Mono L. samples all gave pCO_2 values in the range of $800\text{-}900\times10^{-6}$ atmospheres, or about 2.5 to 3 times the atmospheric value for pCO_2. Thus Mono L., as well as the other lakes to a somewhat lesser extent, has a substantial net gas exchange flux of inorganic carbon to the atmosphere.

Chemical data on the samples of alkaline lake water collected in 1981 are reported in Table 33. Total alkalinity (measured by procedures described in Edmond, 1970) in Mono L. was about 0.7 eq/l, 0.5 eq/l in L. Abert, 0.3 eq/l in Summer L., 0.05 eq/l in Walker L. and Goose L. and 0.02 eq/l in Pyramid L. Carbonate alkalinity, which was measured by subtracting the measured titration alkalinity after removal of the dissolved inorganic carbon, was somewhat lower for each of the lakes, and the total range of carbonate alkalinity values was about a factor of 30 between Pyramid L. and Mono L. The lowest carbonate alkalinity value (Pyramid L.) was approximately an order of magnitude greater than values typical of seawater. Lake Abert had extremely high dissolved silicate and quite low dissolved sulfate relative to other anions in the alkaline lakes. Total boron, as measured by plasma emission spectroscopy, was about 5±3% of total inorganic carbon, with higher values for lakes with the highest alkalinity. Carbonate ion concentrations, as measured by difference between carbonate alkalinity and total inorganic carbon, range from a maximum of 200 mM for Mono L. to 1-2 mM for Pyramid L., compared to a value of ≈ 0.2 mM for seawater. Measurements of carbonate ion (0.21±0.01 M) in Mono Lake (Simpson and Takahashi, 1973) made previously by a pCO_2 titration method (Simpson and Broecker, 1973) were quite similar to those reported here (Table 33).

Representative alkaline lake anion concentrations of chloride, sulfate and carbonate alkalinity, expressed in terms of equivalents, are listed in Table 34. These data are based on measurements made on samples collected in August, 1981 (Table 33), and the percent of total anion values were computed assuming that Cl^-, $SO_4^=$, HCO_3^- and $CO_3^=$ account for all of the anion composition. These relative anion proportion values are plotted in Figure 21, indicating the range of relative anion abundances represented by these alkaline lake samples. Lake Abert had the lowest proportion of sulfate equivalents, Summer L. the highest proportion of carbonate equivalents, and Pyramid L. the lowest proportion of carbonate equivalents.

Table 31

Published Chemical Data for Alkaline Lakes
(mM)

	Mono L.[a]	L. Abert[b]	Walker L.[c]	Pyramid L.[a]	Goose L.[b]	Summer L.[b]
Na^+	935	-	101	71	11.5	106
K^+	30	14	2.4	3.1	0.7	2.9
Mg^{++}	1.4	<8	2.7	4.8	0.1	<.05
Ca^{++}	0.1	<5	0.2	0.2	0.4	<.1
Cl^-	380	403	46	56	2.3	45
$So_4^=$	77	7.6	16	2.9	0.5	3.6
ΣCO_2	261	171	29	19	9	52
Alk	433	296	36	22	10	57
pH	9.6	9.7	9.2	8.9	8.9	9.6

a) Jones, 1966
b) Phillips and Van Denburgh, 1971
c) Whitehead and Feth, 1961

Table 32

Field Equilibration Chemical Data on Alkaline Lakes - July, 1981

Lake Sampled	Depth (m)	Temp (°C)	pCO_2 insitu (µatm)	pCO_2 (22°C)	ΣAlk meq/l
Mono L.	1	20.3	854	893	710
"	"	"	–	–	706
"	7	19.8	843	894	663
"	10	"	"	"	691
"	13	15.2	750	904	683
"	15	7.9	525	792	706
"	18	6.0	523	841	698
"	30	4.8	523	874	651
L. Abert	1	24.5	686	644	504
"	"	"	672	608	508
Walker L.	1	24.0	646	612	56.6
"	75	4.5	275	487	52.4
Pyramid L.	1	22.5	487	477	24.7
"	"	"	509	502	"
"	15	10.4			25.0
"	30	9.7			23.9
"	40	7.8			–
"	45	7.5			24.3
"	75	~4.5			23.8

Table 33

Chemical Composition of Alkaline Lakes

Lake Sampled	Depth (m)	ΣAlkalinity (meg/l)	ΣCO_2 (mM)	NonCarbonate Alkalinity (meq/l)	ΣBoron (mM)	ΣSilicate (mM)	Phosphate (mM)	Chloride (mM)	Sulfate (mM)	Carbonate Alkalinity (meq/l)	ΣCarbonate Ion (mM)	pH
Mono L. - 79	1	-	-	-	-	-	-	589	-	-	-	-
Mono L. - 80	1	-	-	-	-	-	-	555	-	-	-	-
Mono L. - 81	2	-	-	-	-	-	-	597	-	-	-	10
"	10	702	453	-	-	-	-	-	134	-	-	-
"	10	703	-	48.1	48.7	0.44	0.72	-	-	655	202	-
"	10	701	-	43.1	-	-	-	-	-	658	205	-
"	18	690	-	44.2	-	-	-	-	-	646	193	-
L. Abert	1	502	330	16.6	9.8	3.48	0.72	735	12.3	485	155	10
Walker L.	1	55.1	33.7	1.3	2.5	0.014	0.018	74.2	26.7	53.8	20	9.2
Pyramid L.	30	24.1	20.8	1.9	1.3	0.008	0.0054	59.1	3.13	22.2	1.4	9.1
Goose L.	1	53.5	33.6	3.2	1.5	1.09	0.17	65.6	3.31	50.3	17	9.5
Summer L.	0.1	275	230	7.1	12	-	-	149	13.3	268	38	-
Sea Water		2.84	2.58	0.09	0.43	0.15	0.003	548	28	2.75	0.18	8.0

Table 34

Average Major Anion Composition of Alkaline Lakes

Lake	Chloride (meq/l)	Chloride (% of ΣAnions)	Sulfate (meq/l)	Sulfate (% of ΣAnions)	Carbonate Alkalinity (meq/l)	Inorganic Carbon (% of ΣAnions)	Total anions: $Cl^- + SO_4^= +$ $HCO_3 + CO_3^=$ (meq/l)	Carbonate ion (mM)
Mono L.	595	39	268	18	650	43	1513	200
Abert L.	735	59	24	2	485	39	1244	160
Walker L.	74	41	53	29	54	30	181	20
Pyramid L.	59	68	6.3	7	22	25	87.3	(10)
Gooose L.	66	54	6.6	5	50	41	122.6	20
Summer L.	150	34	27	6	270	60	447	40
Sea Water	550	95	28	5	2.5	0.4	580.5	0.2

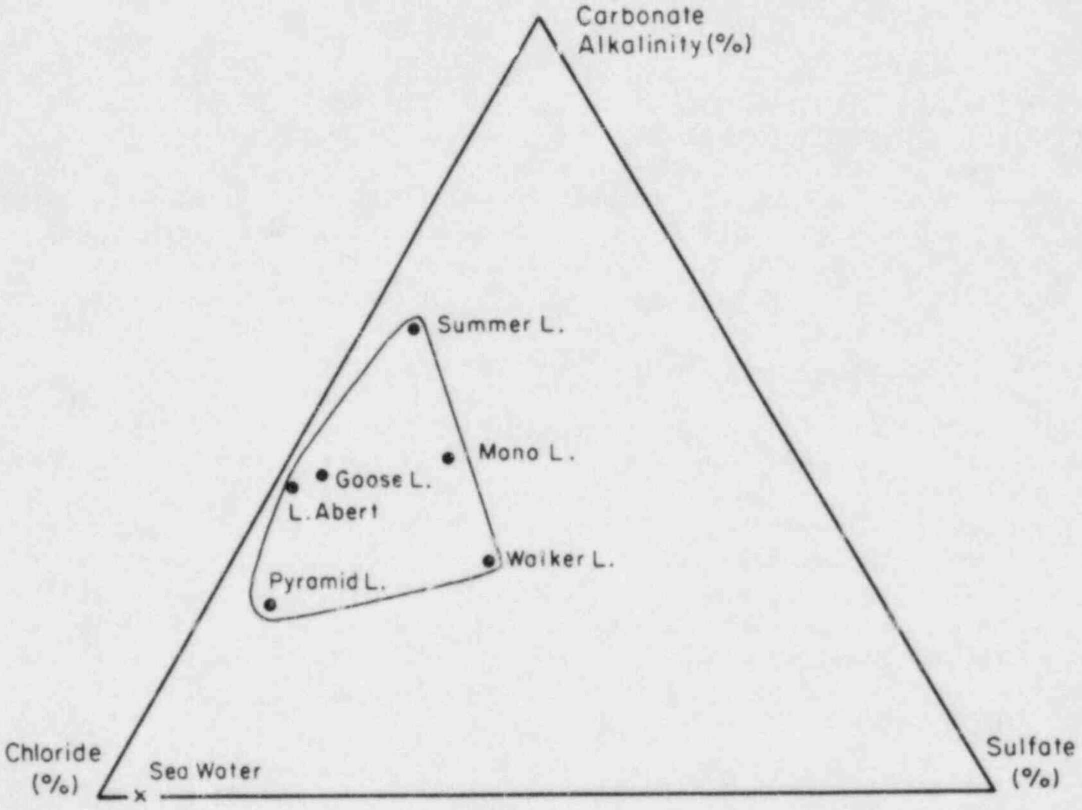

Carbonate Alkalinity (%)
Summer L.
Mono L.
Goose L.
L. Abert
Walker L.
Pyramid L.
Chloride (%)
Sea Water
Sulfate (%)

Figure 21.

7.0 SUMMARY OF RADIONUCLIDE CONCENTRATIONS IN ALKALINE LAKE WATER AND SEDIMENTS

Measurements of radionuclides in natural water systems often present quite difficult analytical problems, especially for samples with high ionic strengths. We have not provided any details of the analytical procedures, nor substantial complications encountered in making the measurements reported here. In general the procedures were those employed in our laboratory for low-level radionuclide measurements in seawater. Considerably more space would be required to document the analytical methodologies than seems justified, but suffice it to say that it would not have been possible to make the large range of measurements reported here without considerabe effort by a number of radiochemists and chemists at LDGO as well as others at USC and WHOI.

Dissolved ^{239,240}Pu concentrations in Mono L. have been measured at LDGO using two different yield tracers (^{236}Pu and ^{242}Pu), as well as several sample volumes, a range of spike equilibration times, and different oxidation states of the yield tracer. The range of analytical values obtained (Figure 22) probably does not indicate real differences in Mono L. dissolved plutonium concentrations at different times, but rather the substantial analytical difficulties encountered in measuring a few dpm of ^{239,240}Pu in a large sample of very salty water with chemical characteristics quite different from the fresh water or seawater compositions for which most of the analytical procedures have been developed. The dissolved ^{239,240}Pu concentration of 13 pCi/m^3 reported for Mono L. (Table 35) was obtained by averaging all of the LDGO data (Table 2). Although this average value was quite similar to the values most frequently obtained in our measurements (Figure 22), several samples for which we have no reason to suspect problems gave values almost twice as great as the average. In general, the dissolved radionuclide data reported in Table 35 was the average of all of the LDGO data, or a single value if that were the only result available. Values listed in parentheses were computed (Walker L. and Pyramid L.) from ratios of ^{238}Pu to ^{239,240}Pu observed in other lakes, or appear to be either too low (Pyramid L. ^{234}Th) or too high (Walker L. ^{228}Th).

The range of ^{239,240}Pu concentrations observed in Mono Lake sediments (Figure 23) was considerably larger than for sediment ^{137}Cs concentrations (Figure 24), but we believe this variation to be real. The covariance of ^{239,240}Pu with observed trends in the ^{137}Cs sediment concentrations with depth, plus the substantially less complicated analytical procedures required for measuring plutonium in a few grams of sediment compared to 80 liters of water both tend to increase confidence in the absolute values of the reported sediment ^{239,240}Pu concentrations. In this case, the difficulty of deriving one "representative" value of ^{239,240}Pu for Mono Lake sediments relates more to real heterogeneity in the system than to analytical uncertainties. For all of the individual data points we have quoted analytical uncertainties based only on counting statistics (one sigma), including counting uncertainty in background and blank values. This quoted uncertainty does not address systematic errors, for which we could not reliably assign numerical values. In general, we

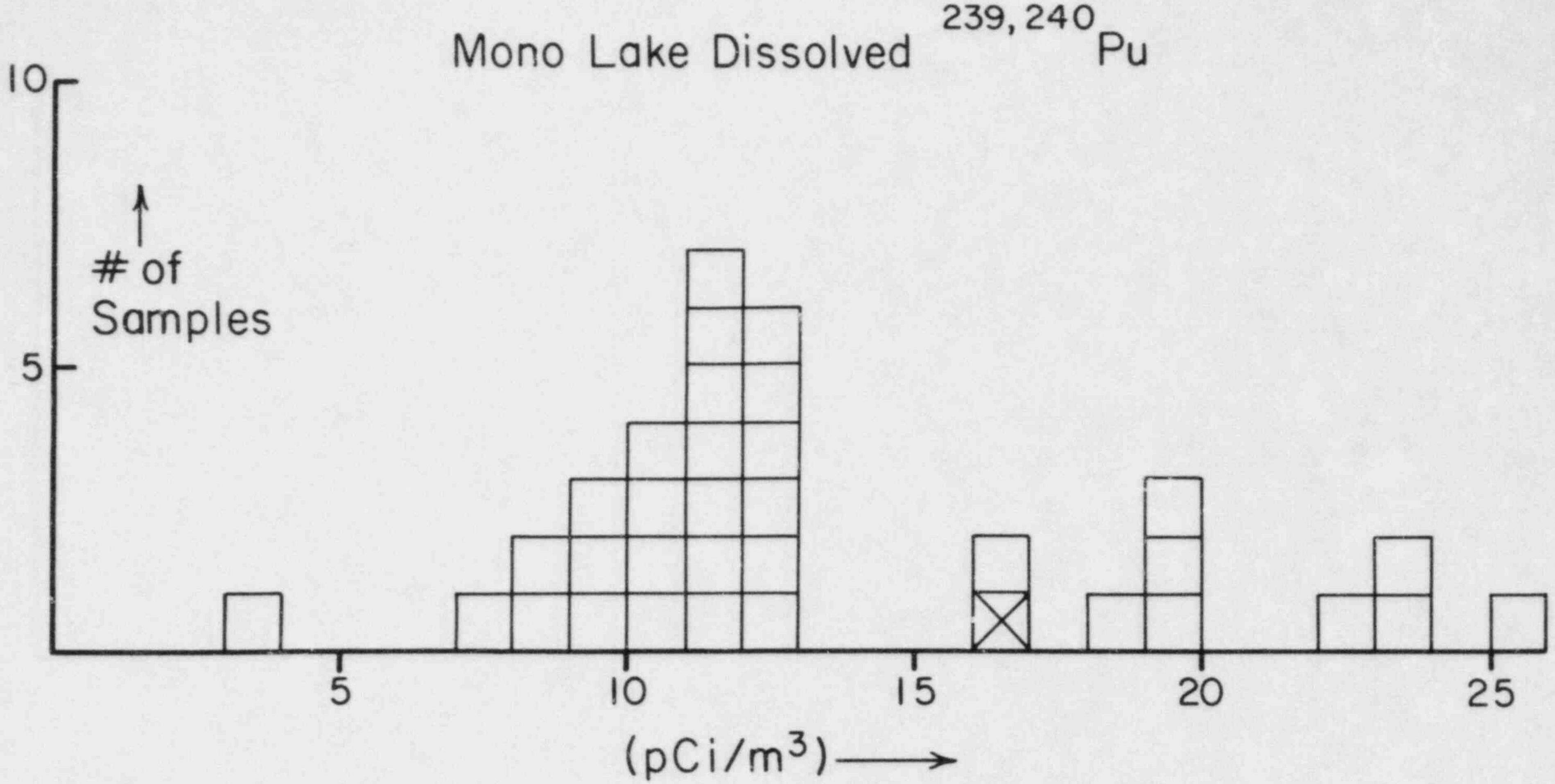

Mono Lake Dissolved ^{239,240}Pu
10
5
# of Samples
5
10
15
20
25
(pCi/m^3)

Figure 22.

Table 35

Summary of Dissolved Radionuclides in Alkaline Lakes

Nuclide	Mono L.	L. Abert	Walker L.	Pyramid L.	Goose L.	Summer L.
^{241}Am (pCi/m^3)	1.8	-	-	-	-	-
^{239,240}Pu (pCi/m^3)	13	11	0.5	0.2	-	-
^{238}Pu (pCi/m^3)	0.54	0.42	(0.02)	(0.008)	-	-
^{238}U (pCi/l)	170	19	51	6	4	9
^{234}U (pCi/l)	230	47	72	12	9	15
^{231}Pa (pCi/m^3)	115	-	-	-	-	-
^{234}Th (pCi/l)	220	18	36	(0.6)	1.8	-
^{232}Th (pCi/m^3)	690	94	5	3	-	-
^{230}Th (pCi/m^3)	1330	130	8	4	-	-
^{228}Th (pCi/m^3)	790	110	(47)	9	-	-
^{227}Ac (pCi/m^3)	<2.7	-	-	-	-	-
^{228}Ra (pCi/m^3)	230	-	-	-	-	-
^{226}Ra (pCi/m^3)	420	40	200	25	20	-
^{210}Po (pCi/m^3)	105	185	7.7	-	-	-
^{210}Pb (pCi/m^3)	200	750	63	-	-	-
^{137}Cs (pCi/l)	2.0	-	0.13	0.14	-	-
^{90}Sr (pCi/l)	0.16	-	-	-		
HTO (T.U.)	70	35	93	45	-	-

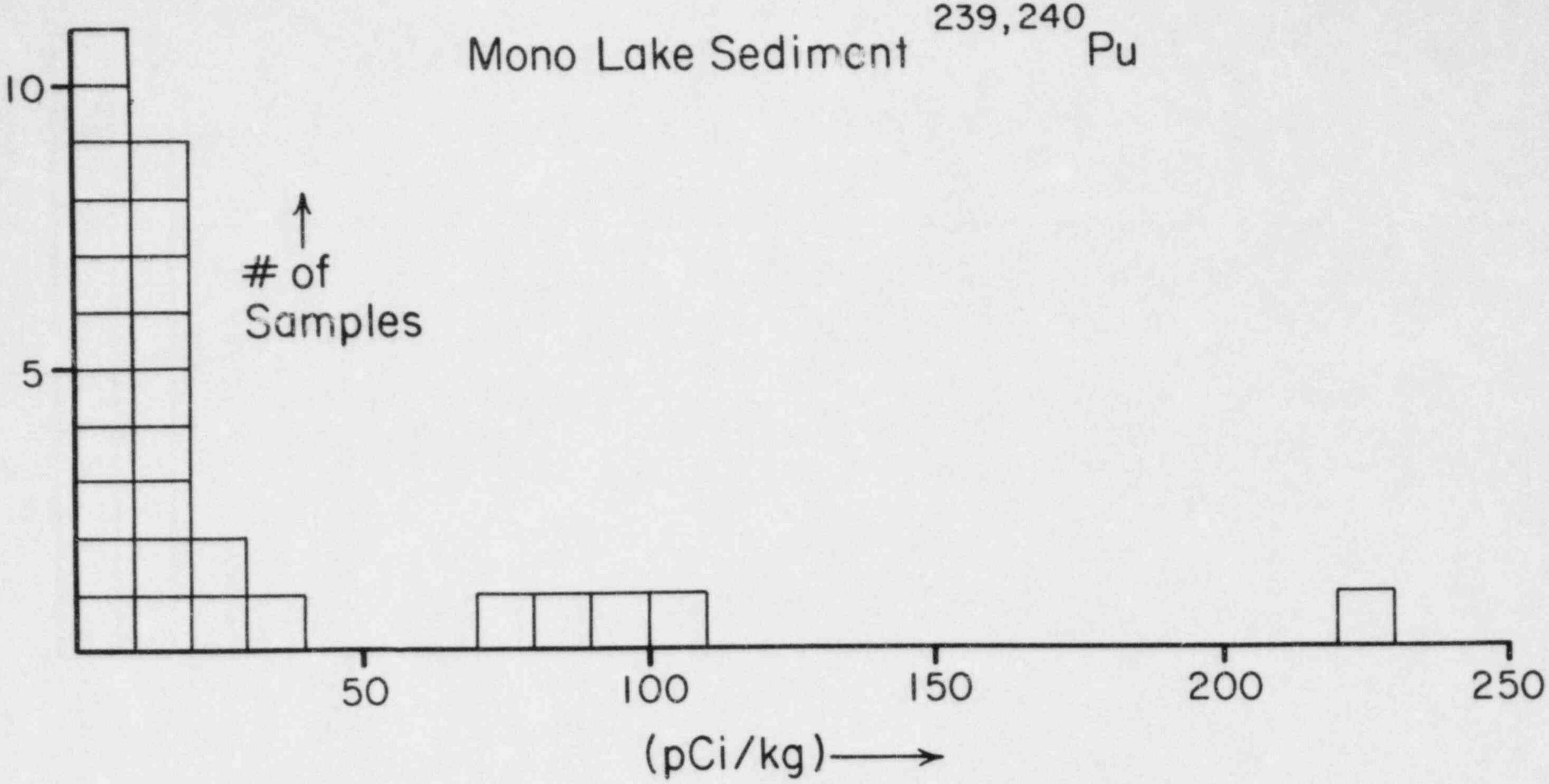

Mono Lake Sediment $^{239,240}Pu$
10
5
# of Samples
50
100
150
200
250
(pCi/kg)

Figure 23.

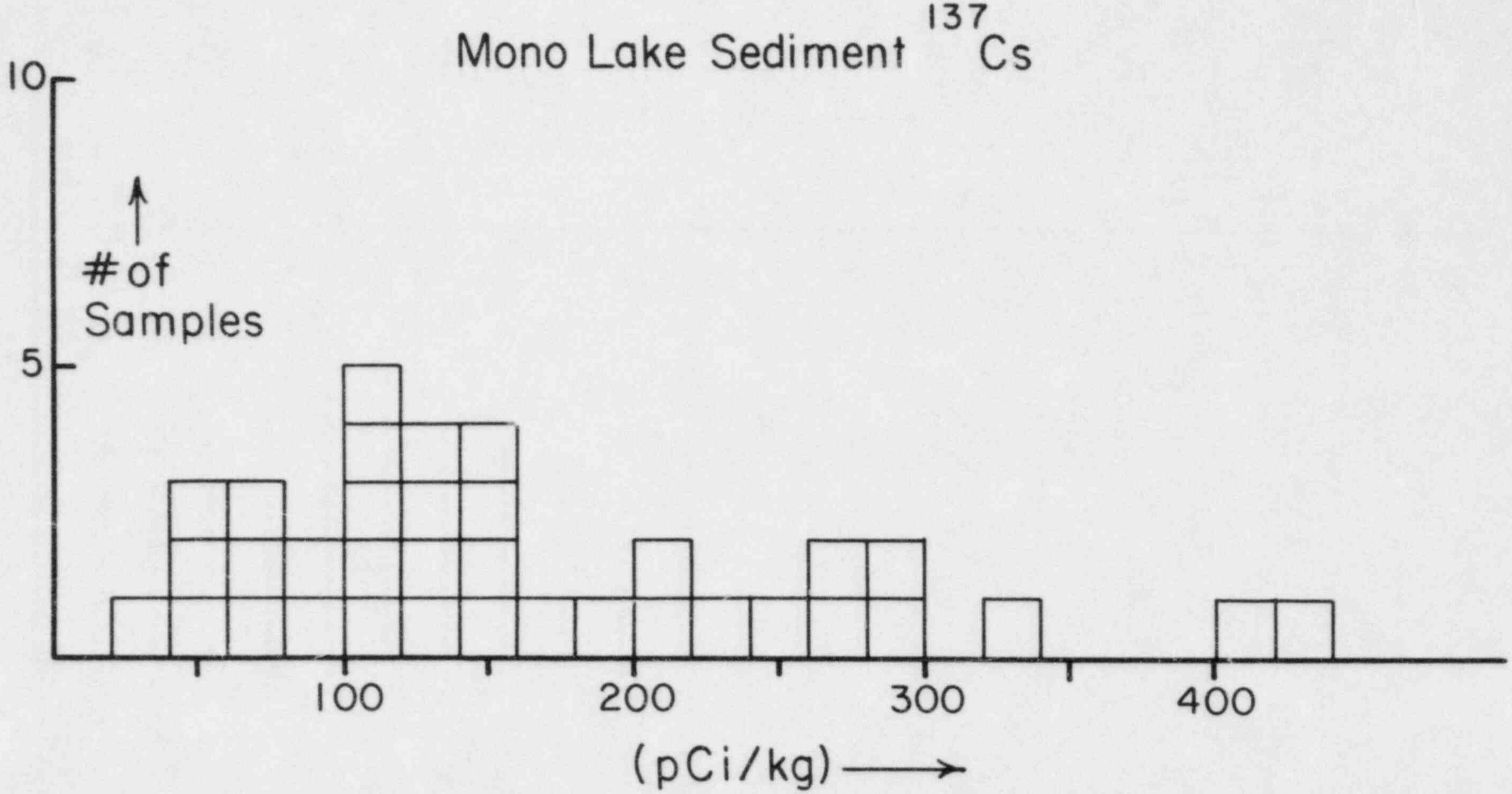

Mono Lake Sediment ^{137}Cs
10
5
# of Samples
100
200
300
400
(pCi/kg)

Figure 24.

have chosen to average all of the available sediment data (i.e. for ^{239,240}Pu) or all of that in the top 4 cm of sediment cores (i.e. for ^{137}Cs) to generate the values listed in Table 36. Sediment concentrations listed in brackets were computed assuming secular equilibrium with a measured radioactive parent nuclide or a nuclide earlier in the decay scheme whose activity could be reasonably estimated (^{231}Pa, ^{234}Th, ^{227}Ac, ^{228}Ra, ^{226}Ra, ^{210}Po, and ^{210}Pb). In the case of ^{210}Pb we know the top few centimeters of Mono L. sediment were actually about 2-3 times the activity of ^{226}Ra (Simpson et al., 1980; Hammond, personal communication), but we consider this difference to be small enough to neglect here.

If we divide the sediment radionuclide concentrations compiled in Table 36 by the dissolved radionuclide concentrations in Table 35, the results can be expressed as a distribution coefficient ($K_d = A_i$ per g sediment/A_i per ml water). These ratios, listed in Table 37, range from 1×10^1 for ^{238}U and ^{234}U in Mono L. to 4×10^5 for ^{238}Pu in Pyramid L. Values for ^{234}Th are listed in brackets because the short radioactive half-life (24 days) does not permit sufficient time for this nuclide, produced by decay of ^{238}U in solution, to "equilibrate" with the sediments. The close agreement of K_d values for ^{232}Th, ^{230}Th and ^{228}Th (except for ^{228}Th in Walker L. and Pyramid L.) argue against interpreting the computed ^{234}Th ratios as distribution coefficients. The value for ^{227}Ac in Table 37 is also listed in brackets because dissolved ^{227}Ac was only reported as a "less than" value, and thus does not have as much confidence as for nuclides actually detected at measurable activities.

Distribution coefficient values from Table 37 are plotted on a log scale as a function of the logarithm of carbonate alkalinity in Figures 25, 26, 27, 28, 29, and 30. Distribution coefficients of ^{239,240}Pu and ^{238}Pu (Figure 25) were two orders of magnitude lower in Mono L. and L. Abert ($2\text{-}3\times10^3$) than in Pyramid L. ($2\text{-}4\times10^5$). ^{238}U and ^{234}U distribution coefficients increased by only about a factor of twenty in the lower alkalinity lakes (Figure 26). The substantial difference in uranium distribution coefficients between Mono Lake and L. Abert (factor of 5-6) probably cannot be attributed only to the difference in carbonate alkalinity between these two lakes, but based on the trends observed for the other lakes, the value for L. Abert seems most anomalous. Distribution coefficients for ^{232}Th and ^{230}Th show much higher values ($1\text{-}3\times10^5$) for lower alkalinity lakes, compared to $0.5\text{-}2\times10^3$ for Mono L. and L. Abert (Figure 27). ^{234}Th is clearly anomalous among the thorium isotopes, as the result of its short half-life as discussed earlier, and even ^{228}Th ($t_{1/2} \simeq 1.9$ yrs) appears to yield a K_d value substantially lower than for ^{232}Th and ^{230}Th in Walker L. and Pyramid L. However, without data for dissolved ^{228}Ra in these two lakes, it is difficult to invoke slow ^{228}Th equilibration time following production in the water column from a soluble parent nuclide. Note that in the case of Mono L., the ratio of ^{228}Th to ^{228}Ra in the water column is actually greater than 1 (Table 35).

We have plotted distribution coefficients of ^{226}Ra (Figure 28), ^{210}Pb and ^{210}Po (Figure 29), and ^{137}Cs (Figure 30) as a function of carbonate alkalinity, but in the case of both ^{226}Ra and ^{137}Cs, the lower values of K_d for higher carbonate alkalinity probably have more to do with the

Table 36

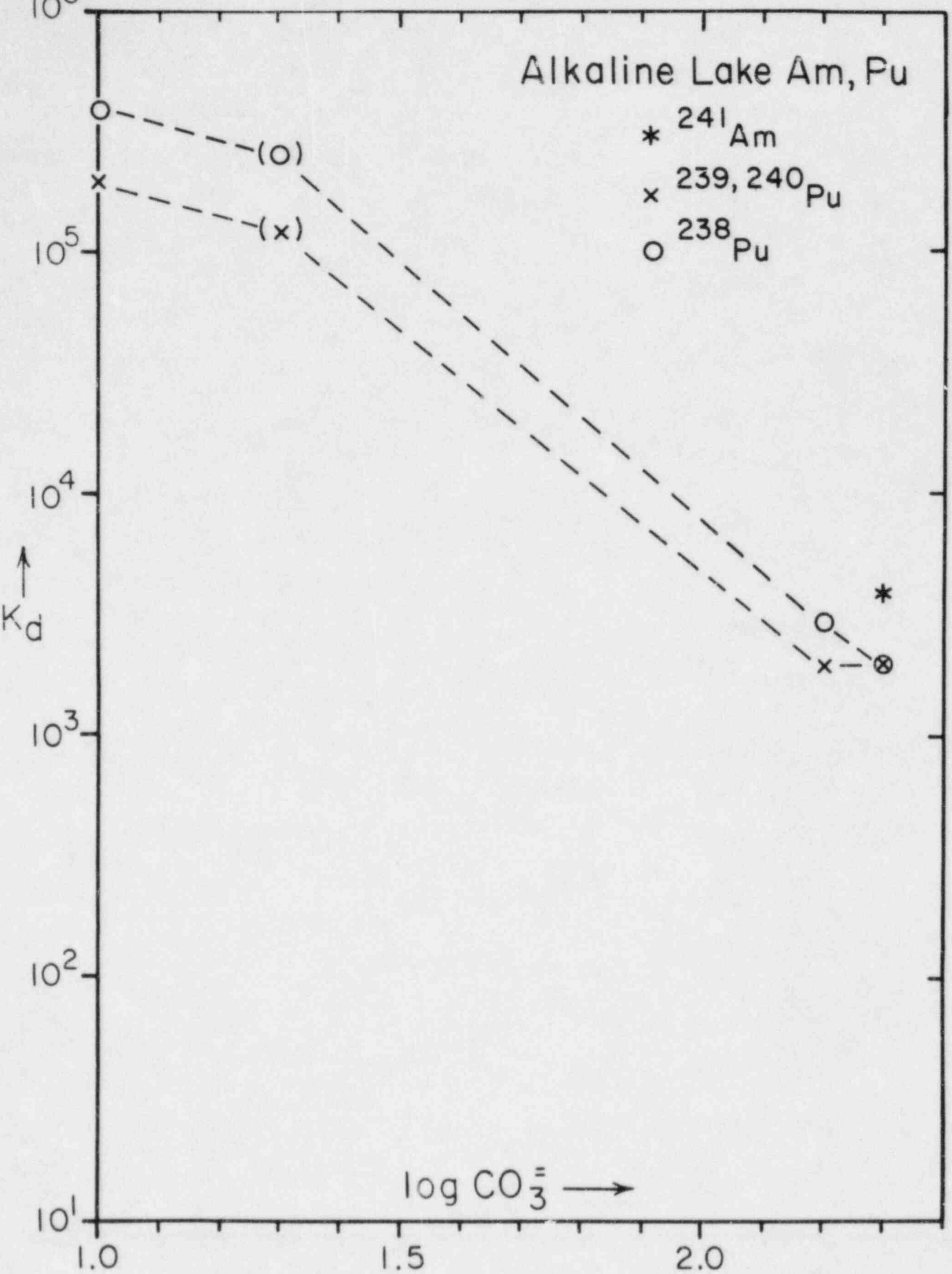
Summary of Sediment Radionuclides in Alkaline Lakes

Nuclide	Mono L.	L. Abert	Walker L.	Pyramid L.	Goose L.	Summer L.
^{241}Am (pCi/kg)	[7]	-	-	-	-	-
^{239,240}Pu (pCi/kg)	28	22	16	40	20	10
^{238}Pu (pCi/kg)	1.1	1.1	0.6	3	0.7	0.5
^{238}U (pCi/g)	2.4	1.1	2.7	1.4	0.1	0.2
^{234}U (pCi/g)	2.9	2.4	3.5	2.2	0.3	0.4
^{231}Pa (pCi/g)	[0.11]	-	-	-	-	-
^{234}Th (pCi/g)	[2.4]	[1.1]	[2.7]	[1.4]	[0.1]	[0.2]
^{232}Th (pCi/g)	0.5	0.2	1.4	0.4	0.2	0.3
^{230}Th (pCi/g)	0.7	0.2	1.5	0.5	0.3	0.3
^{228}Th (pCi/g)	0.4	0.2	1.3	0.4	0.3	0.3
^{228}Ac (pCi/g)	0.3	0.2	1.2	0.4	0.3	0.4
^{227}Ac (pCi/g)	[0.11]	-	-	-	-	-
^{228}Ra (pCi/g)	[0.5]	[0.2]	[1.4]	[0.4]	[0.2]	[0.3]
^{226}Ra (pCi/g)	[0.8]	[0.2]	[1.5]	[0.6]	[0.3]	[0.4]
^{214}Bi (pCi/g)	0.8	0.2	1.5	0.6	0.3	0.4
^{210}Po (pCi/g)	[0.8]	[0.2]	[1.5]	[0.6]	[0.3]	[0.4]
^{210}Pb (pCi/g)	[0.8]	[0.2]	[1.5]	[0.6]	[0.3]	[0.4]
^{137}Cs (pCi/kg)	150	1500	1800	1100	750	600
^{90}Sr (pCi/kg)	~1000	-	-	-	-	-
^{40}K (pCi/g)	16	15	16	12	9	17

Table 37

Ratio of Sediment to Dissolved Radionuclides
(expressed as $K_d = A_i$ per g sed./A_i per ml water)

Nuclide	Mono L.	L. Abert	Walker L.	Pyramid L.	Goose L.	Summer L.
^{241}Am	4×10^3	-	-	-	-	-
^{239,240}Pu	2×10^3	2×10^3	3×10^4	2×10^5	-	-
^{238}Pu	2×10^3	3×10^3	3×10^4	4×10^5	-	-
^{238}U	1×10^1	6×10^1	5×10^1	2×10^2	3×10^1	2×10^1
^{234}U	1×10^1	5×10^1	5×10^1	2×10^2	3×10^1	3×10^1
^{231}Pa	1×10^3	-	-	-	-	-
^{234}Th	$[1\times10^1]$	$[6\times10^1]$	$[8\times10^1]$	$[2\times10^3]$	$[6\times10^2]$	-
^{232}Th	7×10^2	2×10^3	3×10^5	1×10^5	-	-
^{230}Th	5×10^2	2×10^3	2×10^5	1×10^5	-	-
^{228}Th	5×10^2	2×10^3	3×10^4	4×10^4	-	-
^{227}Ac	$[>4\times10^4]$	-	-	-	-	-
^{228}Ra	2×10^3	-	-	-	-	-
^{226}Ra	2×10^3	5×10^3	8×10^3	2×10^4	2×10^4	-
^{210}Po	8×10^3	1×10^3	2×10^5	-	-	-
^{210}Pb	4×10^3	3×10^2	2×10^4	-	-	-
^{137}Cs	8×10^1	-	1×10^4	8×10^3	-	-
^{90}Sr	6×10^3	-	-	-	-	-



Alkaline Lake Am, Pu
* ^{241}Am
× ^{239,240}Pu
○ ^{238}Pu
10^6
10^5
10^4
10^3
10^2
10^1
Kd
log $CO_3^=$ →
1.0
1.5
2.0

Figure 25.

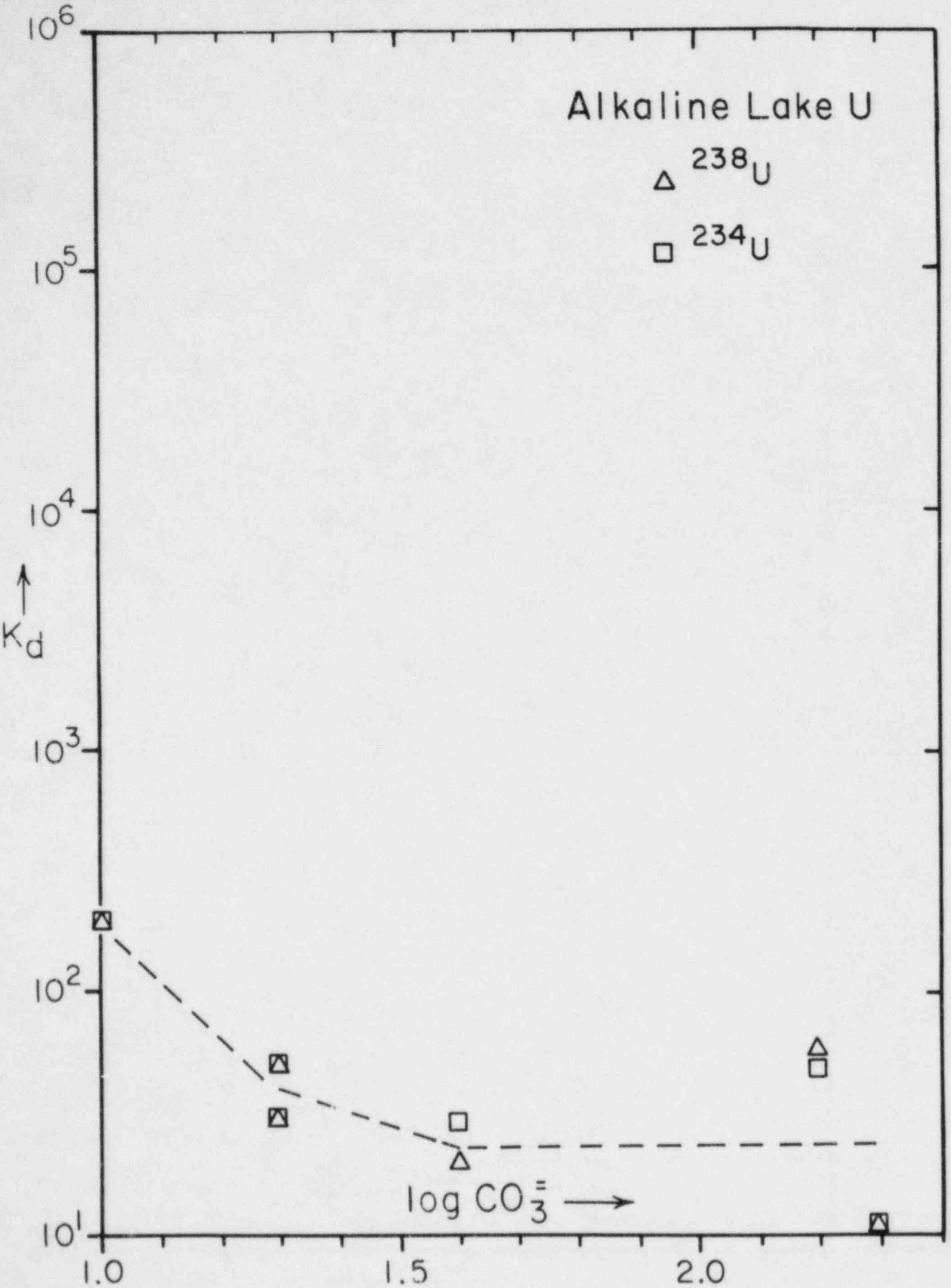

Alkaline Lake U
△ ^{238}U
□ ^{234}U
10^6
10^5
10^4
10^3
10^2
10^1
K_d
log $CO_3^{=}$
1.0
1.5
2.0

Figure 26.

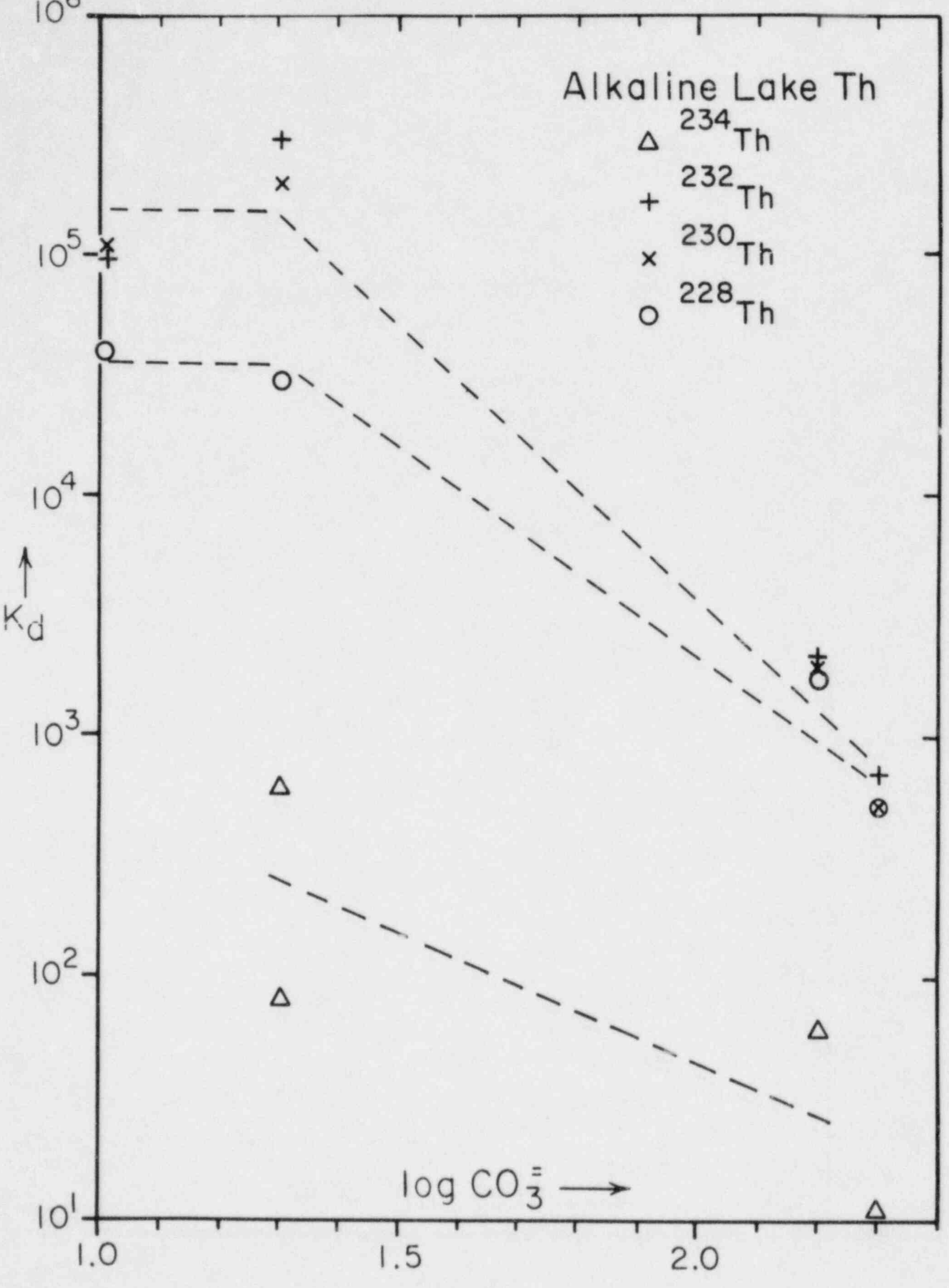

Alkaline Lake Th
△ ^{234}Th
+ ^{232}Th
× ^{230}Th
○ ^{228}Th
K_d
log $CO_3^=$
10^6
10^5
10^4
10^3
10^2
10^1
1.0
1.5
2.0

Figure 27.

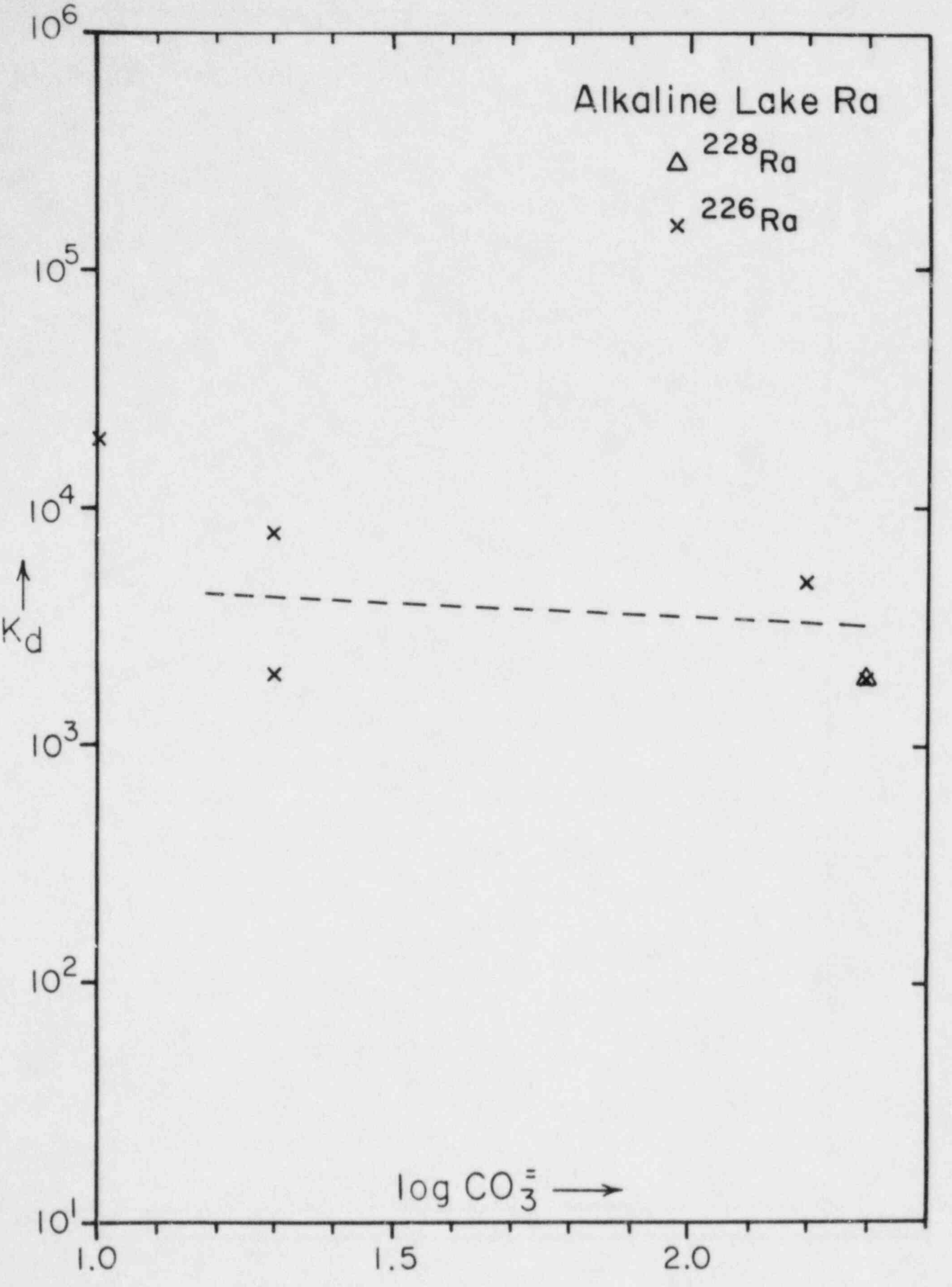

Alkaline Lake Ra
△ ^{228}Ra
× ^{226}Ra
10^6
10^5
10^4
10^3
10^2
10^1
K_d
log $CO_3^=$
1.0
1.5
2.0

Figure 28.

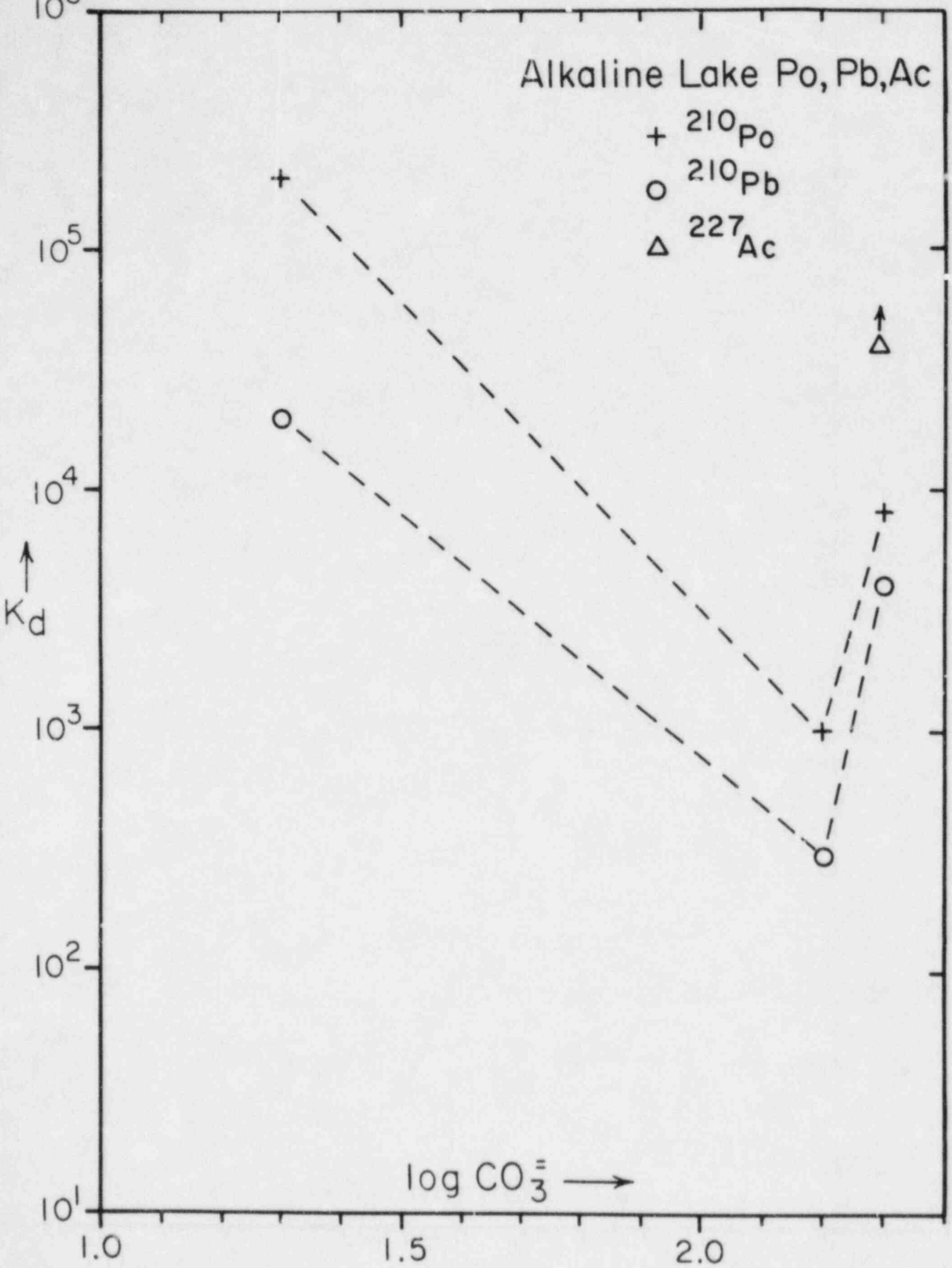

Alkaline Lake Po, Pb, Ac
+ 210Po
○ 210Pb
△ 227Ac
Kd
log CO3=
10^6
10^5
10^4
10^3
10^2
10^1
1.0
1.5
2.0

Figure 29.

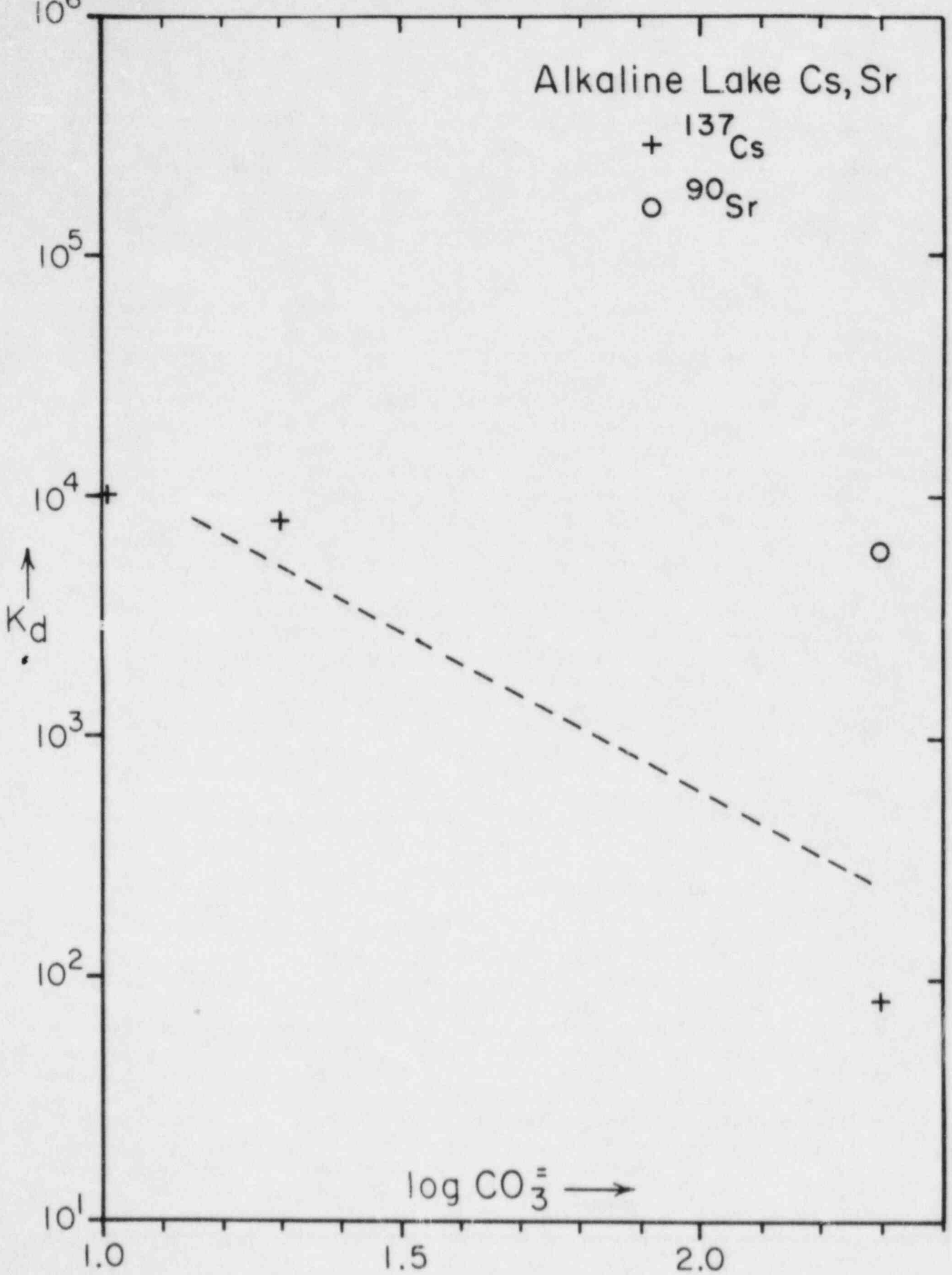

Alkaline Lake Cs, Sr
+ ^{137}Cs
○ ^{90}Sr
10^6
10^5
10^4
10^3
10^2
10^1
K_d ↑
log $CO_3^=$ →
1.0
1.5
2.0

Figure 30.

higher ionic strengths (especially higher Na^+, K^+, Cs^+) of Mono L. and L. Abert than to the effect of carbonate complexing. However, for consistency, we have reported the data for these nuclides in the same form as for Pu, U and Th isotopes, which probably all are strongly influenced by carbonate ion complexing. The distribution coefficient of ^{231}Pa (1×10^3) in Mono L. (Figure 29) is quite similar to those for ^{232}Th, ^{230}Th and ^{228}Th.

Presentation of data on log-log plots can sometimes obscure valuable information. The effects of carbonate complexing on enhancing solubility of both Pu and Th appear to be very large, and can be more readily observed on a linear plot of dissolved ^{239,240}Pu, ^{232}Th and ^{232}Th vs. carbonate alkalinity (Figure 31) for the alkaline lakes (radionuclide data from Table 35). Another illustrative comparison can be made between dissolved radionuclide concentrations in Mono L. and seawater (Table 38). In this case, concentrations typical of the upper few hundred meters of the water column in the northern hemisphere have been used for the ocean to permit reasonable comparisons for the fallout nuclides. Deep ocean ^{226}Ra values are much higher (by a factor of ≈ 5) than the value quoted in Table 38 but fallout nuclide activities in deep water are much lower. The ratio of Mono Lake ^{137}Cs to that for seawater is 15-20, primarily because of the deeper "dilution" depth for this fallout nuclide in the sea. ^{90}Sr shows a much lower ratio than for ^{137}Cs because much of this nuclide has apparently been removed from solution in Mono L., probably in association with $CaCO_3$ precipitation in localized deposits near the lake margin. The most dramatic trends revealed in Table 38 are the greatly enhanced dissolved concentrations in Mono L. of ^{238}U, ^{234}U ($\approx 10^2$), ^{230}Th, ^{228}Th, ^{231}Pa ($\approx 10^3$) and ^{232}Th ($> 10^5$), compared to seawater. Thus, these two types of saline water with very similar chloride concentrations (≈ 0.6 m) differ in dissolved concentrations of radionuclides with oxidation states of IV-VI by orders of magnitudes. The primary cause of this difference appears to be complexing by carbonate ion.

8.0 LABORATORY DISTRIBUTION COEFFICIENTS FOR MONO LAKE USING TRACER ADDITIONS

The data discussed up to now were obtained from analysis of radionuclide abundances already present in natural waters. The only tracer additions made were for yield measurements. The most common method for estimating distribution coefficients is to add tracers to small laboratory samples to simulate real systems. We have performed a few of these equilibration experiments using Mono L. water and sediments for comparison with our field sample data. For plutonium experiments, we used ^{242}Pu, an alpha emitter normally used in our laboratory as a yield tracer. Small quantities of fine-grained Mono L. sediment (10 or 50 mg, dry weight) were added to 100 ml of Mono L. water. After stirring for a few hours, approximately 100 dpm of ^{242}Pu was added in dilute acid solution to the suspension and allowed to equilibrate for 10 days. Then the particles were separated by filtration and the particle and solution phases were analyzed separately. For 10 mg of particles, about 0.3 dpm of ^{242}Pu adsorbed to the solid phase, while 108 dpm of ^{242}Pu stayed in solution (Table 39, $K_d = 3 \times 10^1$). For 50 mg of particles the K_d obtained was about 4×10^0.

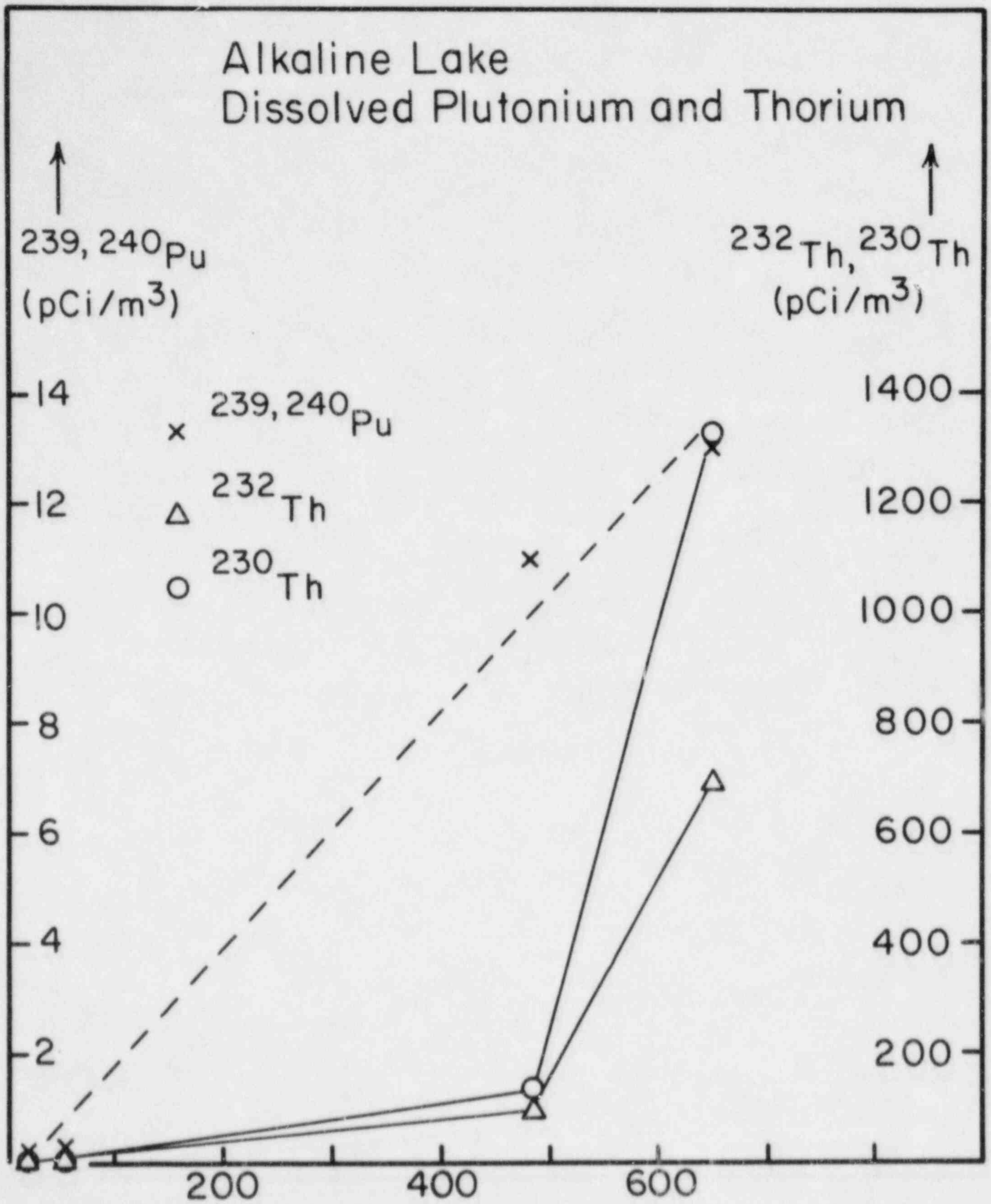

Alkaline Lake
Dissolved Plutonium and Thorium
^{239,240}Pu (pCi/m^3)
^{232}Th, ^{230}Th (pCi/m^3)
× ^{239,240}Pu
△ ^{232}Th
○ ^{230}Th
14
12
10
8
6
4
2
1400
1200
1000
800
600
400
200
200
400
600
Carbonate Alkalinity (meq/l)

Figure 31.

Table 38

Comparison of Dissolved Radionuclides in Mono Lake and Sea Water

Nuclide	Mono Lake (pCi/m^3)	Surface Sea Water (pCi/m^3)	Mono L./Sea Water
^{241}Am (III)	1.8	0.23[a]	8
^{239,240}Pu (IV-VI)	13	1.4[b]	9
^{238}Pu (IV-VI)	0.54	0.056[b]	10
^{238}U (VI)	170,000	1,100[c]	150
^{234}U (VI)	230,000	1,300[c]	180
^{231}Pa (V)	115	0.1[d]	$>1\times10^3$
^{234}Th (IV)	(220,000)	800[e]	~250
^{232}Th (IV)	690	<0.008[f]	$>8\times10^4$
^{230}Th (IV)	1,330	<0.6[d]	$>2\times10^3$
^{228}Th (IV)	790	<0.4[g]	$>2\times10^3$
^{227}Ac (III)	<2.7	-	-
^{228}Ra (II)	230	20[h]	12
^{226}Ra (II)	420	40[i]	11
^{210}Po (IV)	105	30[j]	4
^{210}Pb (II)	200	50[i]	4
^{137}Cs (I)	2,000	120[k]	17
^{90}Sr (II)	160	80[l]	2
HTO (T.U.)	70	15[m]	5

a) Livingston and Bowen, 1976
b) Bowen et al., 1980
c) Ku et al., 1977
d) Moore and Sackett, 1964
e) Bhat et al., 1969
f) Kaufman, 1969; Moore, 1981
g) Knauss et al., 1978
h) Kaufman et al., 1973
i) Bruland et al., 1974
j) Bacon et al., 1976; Nozaki et al., 1976
k) ^{137}Cs to ^{90}Sr ratio assumed to be 1.5
l) Bowen et al., 1969
m) Roether, 1967

Table 39

Laboratory Distribution Coefficient Measurements - Pu

Solution Composition	Sediment Composition	Solution Volume (ml)	Sediment Mass (mg)	Solution Activity (dpm ^{242}Pu)	Sediment Activity (dpm ^{242}Pu)	Distribution Coefficient (ml/g)
Mono L. water (A)	Mono L. Sed	100	10	107.7±1.4	0.31±0.03	3×10^{1}
Mono L. water free of inorganic carbon (B)	"	"	"	43.2±1.4	52.2 ±0.9	1.2×10^{4}
A+B (50/50)	"	"	"	101.4±1.2	0.05±0.01	5×10^{0}
Mono L. water (A)	Mono L. Sed	100	50	98.6±0.7	0.19±0.04	4×10^{0}
Mono L. water free of inorganic carbon (B)	"	"	"	31.7±0.4	70.0 ±2.0	4×10^{3}
A+B (50/50)	"	"	"	100.8±1.2	0.07±0.02	1.4×10^{0}

These values are substantially different than we obtained from our field measurements (Table 37, 2×10^3). A second set of experiments was conducted using Mono L. water which had been first acidified, stripped of CO_2, and readjusted to pH 10. In this case, a much larger fraction of the ^{242}Pu activity adsorbed to the particles (52 dpm for 10 mg and 70 dpm for 50 mg). The K_d values derived for these equilibrations were 1.2×10^4 and 4×10^3, respectively. We also used a mixture of Mono L. water plus Mono L. water free of inorganic carbon (50/50) to equilibrate with 10 and 50 mg of sediment particles. In both cases a very small fraction of the ^{242}Pu activity remained in solution, with K_d values slightly lower than for pure Mono L. water. We expected the K_d results to be slightly higher rather than lower, but do not believe the difference is very significant. These laboratory tracer experiment results clearly demonstrate the strong effect of inorganic carbon (presumably carbonate ion) complexing, but the absolute values of distribution coefficients for plutonium are quite different from those we derived by analysis of radionuclides from field samples in Mono L.

An analogous group of laboratory experiments was also conducted for ^{210}Po using Mono L. water and sediments (Table 40). Polonium became associated with the particles to a greater extent than did plutonium, but the large effect of carbonate ion was also clearly demonstrated. The distribution coefficient values for ^{210}Po in the laboratory experiments using Mono L. water (7×10^2) were about an order of magnitude lower than we derived from our field measurements in Mono L. (8×10^3).

9.0 DISTRIBUTION COEFFICIENTS FOR MONO LAKE WITHOUT TRACER ADDITIONS

In most natural waters, the concentrations of radionuclides already present are usually too low to permit distribution coefficient measurements on small samples. In the case of Mono L., however, the concentrations of ^{238}U, ^{234}U, ^{232}Th, ^{230}Th, ^{228}Th and even ^{239,240}Pu are sufficiently high to permit measurements of radionuclide concentrations in water and particle phases without tracer additions (except as yield monitors). We performed a series of large volume equilibration experiments (1980) at Mono L. (80 liters of Mono L. water and 100 grams of wet Mono L. sediment) to investigate the importance of carbonate complexing (Table 41). The sediments were from 30-50 cm below the sediment surface, and thus free of ^{239,240}Pu prior to the experiment.
After equilibration for 48 hours, the dissolved ^{239,240}Pu concentration (12.6 ± 0.6 pCi/m^3) was the same as a fresh sample of Mono L. surface water. We were not able to detect any ^{239,240}Pu activity on the particles above our background. Equlibration of sediments with Mono L. water acidified, stripped of CO_2, and restored to pH 10 resulted in a very high fraction of the total sample ^{239,240}Pu activity being adsorbed by the particles ($K_d = 3\times10^4$). An additional equilibration of water and sediment after addition of sufficient Na_2CO_3 to restore carbonate alkalinity to a value comparable to unperturbed Mono L. water (carbonate alkalinity ≈ 0.7 eq/l) resulted in approximately the same amount of ^{239,240}Pu activity associated with the two phases (K_d 1×10^3). Thus the distribution coefficient we measured by first inducing adsorption of fallout ^{239,240}Pu, followed by desorption resulting from carbonate complexing, was quite comparable to the ratio based on our field data (2×10^3).

Table 40

Laboratory Distribution Coefficient Measurements - Po

Solution Composition	Sediment Composition	Solution Volume (ml)	Sediment Mass (mg)	Solution Activity (dpm ^{210}Po)	Sediment Activity (dpm ^{210}Po)	Distribution Coefficient (ml/g)
Mono L. water (A)	Mono L. Sed	100	10	161.5±4.3	11.4±0.3	7×10^2
Mono L. water free of inorganic carbon (B)	"	"	"	18.5±0.3	137.5±1.0	7×10^4
A+B (50/50)	"	"	"	105.4±1.8	49.2±1.0	5×10^3
Mono L. water (A)	Mono L. Sed	100	50	121.1±1.6	42.7±1.0	7×10^2
Mono L. water free of inorganic carbon (B)	"	"	"	12.3±0.3	179.0±1.0	3×10^4
A+B (50/50)	"	"	"	31.1±0.7	126.6±1.0	8×10^3

Table 41

Field Distribution Coefficient Measurements - Pu

Solution Composition	Sediment Composition	Solution Volume (l)	Recovered Sediment Dry Weight(g)	Solution Activity (pCi/m^3 ^{239,240}Pu)	Sediment Activity (pCi/m^3 ^{239,240}Pu)	Distribution Coefficient (ml/g)
I Mono L. water (A)[a]	Mono L. Sed	80	23.5	12.6±0.6	N.D.	-
II Mono L. water (B)[b] free of inorganic carbon	"	80	46.6	0.5±0.1	14.2±2.7	3×10^4
III Solution B[c] plus sufficient Na_2CO_3 to restore conditions similar to A	"	80	40.8	6.9±0.4	8.9±0.4	1×10^3

a) ~100 g of wet sediment from the bottom of Mono Lake Core #1 was equilibrated for 48 hrs.

b) ~100 g of wet sediment was equilibrated for 48 hrs. with 80 liters of Mono Lake water which had been acidified, bubbled with N_2 to remove CO_2, and readjusted to pH 10 with NaOH.

c) An identical experiment to that above (b) was done, followed by reequilibration after Na_2CO_3 was added to restore conditions to those similar to unperturbed Mono Lake water.

A similar series of experiments was performed in the laboratory during 1980 using Mono L. water and sediments to establish the partitioning of ^{238}U, ^{234}U, ^{232}Th, ^{230}Th and ^{228}Th (Tables 41 and 42). The general procedures employed were the same as those described above for $^{239,240}Pu$, except that much smaller quantities of water and sediments were used. Approximately 2 liters of water and 0.5 g of sediment were sufficient to measure both uranium and thorium isotopes. In one set of experiments stirring was accomplished by bubbling a stream of air through the suspension (Table 42), and in the second set of experiments nitrogen gas was continuously flushed through the experimental vessels (Table 43). In both sets of experiments the adsorption of Th to particles was substantially increased when inorganic carbon was removed from the water ($K_d = 5\text{-}7 \times 10^3$ for ^{232}Th, ^{230}Th, and ^{228}Th), compared with mixing fresh Mono L. water and sediments ($K_d = 0.7\text{-}2 \times 10^3$ for ^{232}Th, ^{230}Th and ^{228}Th). These latter values were quite comparable to those we obtain from field sample measurements (Table 37, $K_d = 0.5$ to 0.7×10^3).

10.0 CONCLUSIONS

The data reported here for radionuclide concentrations in a group of alkaline lakes demonstrate clearly that effective solubilities of a number of nuclides with oxidation states of III to VI are substantially higher than in natural waters with lower carbonate ion concentrations. In addition, the degree of solubility enhancement is proportional to carbonate ion concentration, especially for elements such as Th and Pu (Simpson et al., 1983). Concentrations of dissolved U and Th (10^{-6} - 10^{-8} mole/l) in the highest alkalinity lakes (0.5 - 0.7 eq/l of carbonate alkalinity) demonstrate substantial mobilities for elements with oxidation states of IV and VI in natural waters with appreciable carbonate ion concentrations. Although both laboratory distribution coefficient experiments and pure-phase thermodynamic solubility calculations clearly support the importance of carbonate complexing in enhancing actinide solubility, neither of these approaches is able to accurately predict or explain distribution coefficients based on field sample radionuclide concentration measurements. There is clearly no adequate substitute for such field data in assessing radionuclide transport pathways and rates for possible releases from a high level waste repository to groundwater.

11.0 RECOMMENDATIONS

Design and performance assessment of high-level radionuclide waste repository facilities requires consideration of a large set of complicated and interactive processes and components. Some of the most important elements of choice for appropriate sites and design strategies are the geologic and hydrologic environments to be used. Solid phase and groundwater chemical conditions, as well as groundwater movement rates in the vicinity of a waste repository will be very strongly influenced by both geologic and hydrologic conditions. Behavior of radionuclides in a variety of groundwater environments can be computed on the basis of a variety of thermodynamic and laboratory simulation models to gain insights about important processes. In addition to these activities, however, it

Table 42

Laboratory Distribution Coefficient Measurements - Th, U[a]

Solution Composition	Sediment Composition	Solution Volume (l)	Recovered Sediment Dry Weight(g)	Nuclide	Solution Activity (pCi/l)	Sediment Activity (pCi/g)	Distribution Coefficient (ml/g)
I Mono L. water (A)	Mono L. Sed	2.20	0.38	^{232}Th	0.64± 0.03	0.53±0.06	8×10^2
				^{230}Th	1.28± 0.04	0.98±0.08	8×10^2
				^{228}Th	0.94± 0.11	0.66±0.09	7×10^2
				^{238}U	354 ± 9	1.6 ±0.2	5×10^0
				^{234}U	451 ±11	3.1 ±0.2	7×10^0
II Mono L. water free of inorganic carbon (B)	"	2.40	0.44	^{232}Th	0.27± 0.02	1.71±0.11	6×10^3
				^{230}Th	0.54± 0.02	3.64±0.16	7×10^3
				^{228}Th	0.44± 0.06	2.50±0.26	6×10^3
				^{238}U	352 ±24	20.4 ±0.5	6×10^1
				^{234}U	438 ±27	24.7 ±0.6	6×10^1
III Solution B plus sufficient Na_2CO_3 to restore conditions similar to A	"	2.40	0.70	^{232}Th	0.53± 0.02	0.48±0.06	9×10^2
				^{230}Th	1.10± 0.03	0.76±0.08	7×10^2
				^{228}Th	0.67± 0.05	0.58±0.10	9×10^2
				^{238}U	352 ±23	1.1 ±0.1	3×10^0
				^{234}U	486 ±19	1.9 ±0.1	4×10^0

a) Suspensions mixed for a period of 1 week with a stream of air bubbles

Table 43

Laboratory Distribution Coefficient Measurements - Th, U[a]

Solution Composition	Sediment Composition	Solution Volume (l)	Recovered Sediment Dry Weight(g)	Nuclide	Solution Activity (pCi/l)	Sediment Activity (pCi/g)	Distribution Coefficient (ml/g)
I Mono L. water (A)	Mono L. Sed	2.55	0.43	^{232}Th	0.49± 0.02	0.66±0.08	$1x10^{3}$
				^{230}Th	1.03± 0.03	1.05±0.10	$1x10^{3}$
				^{228}Th	0.58± 0.05	1.33±0.23	$2x10^{3}$
				^{238}U	357 ±11	1.5 ±0.2	$4x10^{0}$
				^{234}U	473 ± 9	2.7 ±0.2	$6x10^{0}$
II Mono L. water free of inorganic carbon (B)	"	2.45	0.51	^{232}Th	0.25± 0.01	1.80±0.10	$7x10^{3}$
				^{230}Th	0.49± 0.02	3.58±0.14	$7x10^{3}$
				^{228}Th	0.42± 0.04	2.02±0.13	$5x10^{3}$
				^{238}U	345 ±12	3.1 ±0.2	$9x10^{0}$
				^{234}U	489 ±10	3.9 ±0.2	$8x10^{0}$
III Solution B plus sufficient Na_2CO_3 to restore conditions similar to A	"	2.45	0.78	^{232}Th	0.30± 0.01	0.21±0.01	$7x10^{2}$
				^{230}Th	0.61± 0.02	0.38±0.02	$6x10^{2}$
				^{228}Th	0.54± 0.05	0.24±0.01	$4x10^{2}$
				^{238}U	353 ±12	1.5 ±0.1	$4x10^{0}$
				^{234}U	460 ±15	2.2 ±0.1	$5x10^{0}$

a) Suspensions mixed for a period of 1 week with a stream of N_2 bubbles is important to accumulate sufficient information on radionuclide behavior in natural systems to provide direct indication of likely soluble phase nuclide concentrations in the far field for a number of geologic and hydrologic conditions. We recommend that a significantly larger research effort should be devoted to the study of radionuclide behavior in natural environments than is presently occurring. This effort is not presently being accomplished under the general category of "site characterization", and represents what should be a more important component of the national program in high level nuclear waste management.

12.0 PLANNED RESEARCH

The data reported here were obtained to provide information about the influence of carbonate complexing on radionuclide mobility in natural waters. We are currently analyzing water and sediment samples collected from high sulfate and high chloride environments to establish the effect of these anions on enhancing or reducing solubilites of radionuclides in natural systems. The field environments from which these samples were collected include evaporite deposit areas (gypsum or halite) in the USA (New York and New Mexico) and saline lakes in western Canada.

Based on the information gained from the reseach presented here, and preliminary data from other field environments, some specific areas of research that need to be addressed are:
a) the apparent mobility of Pu and Th in extremely high SO_4^{2-} lake waters,
b) the kinetics of U reduction in anoxic waters,
c) redox controls on Th and Ra mobility in groundwaters, and
d) development of improved analytical procedures for determination of radionuclides in high DOC, anoxic waters.

ACKNOWLEDGEMENTS

The data reported here represent considerable field and laboratory effort by a number of people in addition to the authors, including B. Deck and G. Mathieu of Lamont-Doherty Geological Observatory, D. Hammond from the University of Southern California and C. Olsen from Oak Ridge National Laboratory. The information summarized here is based on field work initiated at Mono Lake with financial support by the Department of Energy (E-2529) and expanded to include work at a number of other alkaline lakes with financial support by the Nuclear Regulatory Commission (NRC-04-81-217). Contribution number 3680 of Lamont-Doherty Geological Observatory.

APPENDIX I

Mono Lake U and Th Concentrations

$^{238}U = 170$ pCi/l

Conversion to molar units:

$$2.22\ \frac{\text{dpm}}{\text{pCi}} \times 5.26\text{x}10^{5}\ \frac{\text{min}}{\text{Yr}} \times \frac{4.51\text{x}10^{9}\text{yr}}{\ln 2} \times \frac{1\ \text{mole}}{6.02\text{x}10^{23}\ \text{atoms}}$$

$$= 1.26\text{x}10^{-8} \times \frac{\text{pCi}}{1} = \frac{\text{mole}}{1}$$

$$^{238}U = 1.7\text{x}10^{2}\ \frac{\text{pCi}}{1} \times 1.26\text{x}10^{-8} = \underline{2.1\text{x}10^{-6}\ \text{mole/l}}$$

$^{232}Th = 690$ pCi/m^3

Conversion to molar units:

$$2.22\ \frac{\text{dpm}}{\text{pCi}} \times 5.26\text{x}10^{5}\frac{\text{min}}{\text{Yr}} \times \frac{1.39\text{x}10^{10}\text{Yr}}{\ln 2} \times \frac{1\ \text{mole}}{6.02\text{x}10^{23}\text{atoms}} \times \frac{10^{-3}\text{m}^{3}}{1}$$

$$= 3.89\text{x}10^{-11} \times \text{pCi/m}^{3} = \text{mole/l}$$

$$^{232}Th = 6.9\text{x}10^{2}\ \frac{\text{pCi}}{\text{m}^{3}} \times 3.89\text{x}10^{-11} = \underline{2.7\text{x}10^{-8}\ \text{mole/l}}$$

APPENDIX II

Uranium and Thorium Solubility Calculations for Mono Lake [a]

	U	Th
Assumed solubility limiting species	$UO_2(OH)_2(s)$	$ThO_2(s)$
Solubility product (K_s)	10^{-23}	10^{-54}
$\log[UO_2{}^{2+}]=(\log K_s+28.0-2pH)$	10^{-15}mole/l (pH=10)	--
$\log[Th^{4+}]=(\log K_s+56.0-4pH)$[b]	--	10^{-38}mole/l (pH=10)
$UO_2{}^{2+}+3CO_3{}^{2-}\rightleftharpoons UO_2(CO_3)_3{}^{4-}$ (β_3)[c]	10^{24}	--
$Th^{4+}+5CO_3{}^{2-}\rightleftharpoons Th(CO_3)_5{}^{6-}$ (β_5)[c]	--	10^{37}
U(VI) complex/$UO_2{}^{2+}$-Mono Lake[d]	10^{22}	--
Th(IV) complex/Th^{4+}-Mono Lake[d]	--	10^{33}

---

[a] Most of the calculations summarized in this table are based on results from Allard, 1982.

[b] Values of $[Th^{4+}]$ tabulated were computed from the expression shown in this table (taken from Allard, 1982). Another calculation of ThO_2 solubility suggests $\Sigma Th(IV)$ concentrations of $\approx 10^{-14}$ mole/l above pH=8, including hydrolysis effects but not carbonate ion (Langmuir and Herman, 1980).

[c] These complexes were chosen to illustrate representative actinide carbonate complex constants for Mono Lake, based on personal communications with Prof. I. Grenthe, Royal Institute of Technology, Stockholm, Sweden.

[d] Mono Lake water contains total carbonate ion concentrations of $2x10^{-1}$ mole/l (Simpson and Takahashi, 1973).

APPENDIX III

Fallout Nuclide Inventories in Mono Lake and Lake Abert

Our primary attention has been directed to obtaining dissolved and sediment fallout and natural radionuclide concentrations in alkaline lakes to establish effective distribution coefficients for these nuclides. In the case of fallout nuclides, because information is available on delivery rates per unit area, we can also compare our measured inventories with those expected from regional fallout data. Our water and sediment data is most comprehensive for Mono L. and L. Abert, so we confine inventory estimates to those two lakes.

Dissolved ^{239,240}Pu was measured a number of times in both Mono L. and L. Abert (Tables 2 and 3), and our best estimates of average water column concentrations for the two lakes were 13 pCi/m^3 and 11 pCi/m^3, respectively (Table 35). Total dissolved ^{239,240}Pu can then be estimated by multiplying by lake volumes of 2.7 km^3 and 0.13 km^3 for Mono L. and L. Abert (Table 1). Mono L. has declined in volume substantially since diversion of a major fraction of its stream inputs by the Los Angeles Department of Water and Power began in 1940 and expanded in later years, especially since 1970. Our estimate of volume for the period of our sampling probably has an uncertainty on the order of ± 10%, but should not be much greater because Mono L. is relatively deep (mean depth = 17 meters). The total quantity of ^{239,240}Pu in the water column of L. Abert is somewhat more diffcult to estimate, primarily because of the large changes in volume experienced by this very shallow lake (mean depth = 1.1 meters in August, 1981) over fairly short periods of time. At the level existing in 1981, a change in surface elevation of 0.5 meter can result in a total volume change approaching a factor of two. We have estimated the volume of L. Abert during our sample collection (August, 1981) by comparison of the chloride concentration in the lake at the time of sampling (26.1 ‰) with published data (Table A-1) on chloride and lake surface elevation between 1939 and 1963 (Figure A-1) to estimate the lake level. This computed lake level was then used to estimate the lake volume, based on published (Table 2 in Van Denburgh, 1975) bathymetry information (Figure A-2). Two of the data points (obtained in the 1940's) shown in Figure A-1 were ignored in estimating the relationship between chloride and surface elevation because they fall well outside the range of the other points, and because the sampling locations at the lake were not reported. Significant errors in estimating the surface elevation of a very shallow lake can be made in the absence of careful surveying to fixed bench mark positions during low wind conditions. From the information plotted in Figures A-1 and A-2, we estimate the volume of L. Abert in August, 1981 to have been ≈ 0.13 km^3, which is less than 5% of the volume of Mono L. The total quantities of dissolved ^{239,240}Pu in Mono L. and L. Abert were estimated to be 35 mCi and 1.4 mCi, respectively (Table A-2). The total quantities of other dissolved fallout nuclides were estimated (Table A-2) by multiplying the concentrations listed in Table 35 by the volume of Mono L. as discussed above: ^{241}Am (4.9 mCi), ^{137}Cs (5400 mCi), ^{90}Sr (430 mCi) and HTO (6.1x10^5 mCi).

Estimation of the total sediment burden of fallout radionuclides in the two lakes is considerably more difficult than for the water column, primarily because sediment deposition patterns tend to be extremely heterogeneous. Fallout radionuclide concentrations in sediments are listed for Mono L. in Table A-3 and for L. Abert in Table A-4. Integrals of ^{137}Cs and ^{239,240}Pu accumulation are listed for each core in Table A-5, expressed in terms of

Table A-1

Lake Abert Surface Elevation and Chloride Concentrations*
(1939-1963)

Year of Sampling	Lake Level (ft)	Chloride (ppm)
'39	4,248.2	28,500
'44	4,249.1	16,600
'45	4,247.8	22,900
'55	4,252.8	14,000
'56	4,257.6	7,760
'57	4,256.7	7,440
'58	4,259.1	6,780
'58	4,260.5	5,140
'59	4,259.2	7,040
'59	4,258.9	7,570
'61	4,253.86	13,900
'61	4,253.86	14,100
'61	4,253.3	15,500
'61	4,252.04	19,200
'61	4,251.5	20,800
'62	4,251.81	19,300
'62	4,252.14	18,100
'62	4,252.3	17,900
'62	4,252.36	16,700
'62	4,251.19	22,000
'62	4,250.7	25,000
'63	4,253.08	15,500

*Phillips and VanDenburgh, 1971

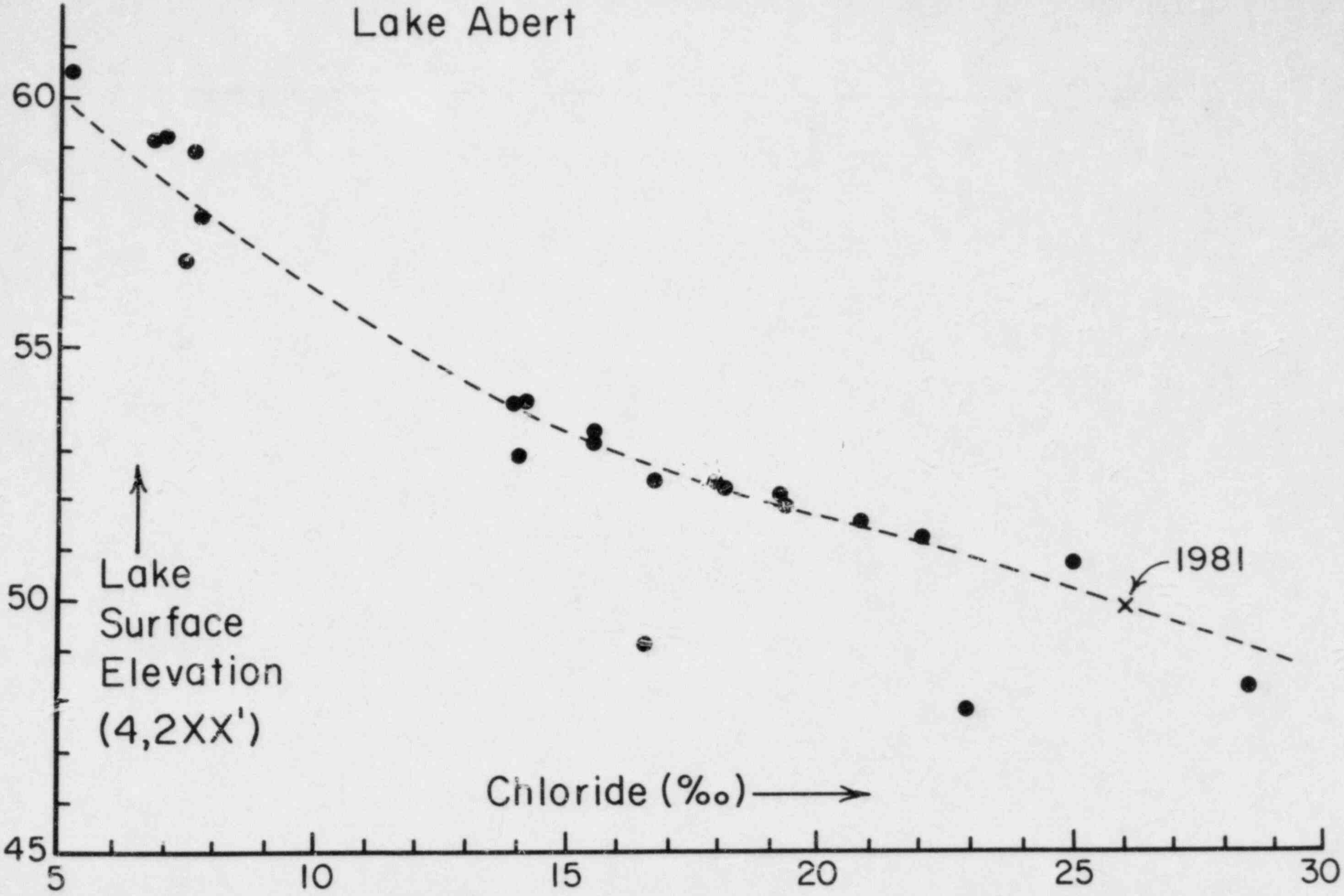

Lake Abert
60
55
50
45
Lake Surface Elevation (4,2XX')
Chloride (‰)
5
10
15
20
25
30
1981

Figure A-1.

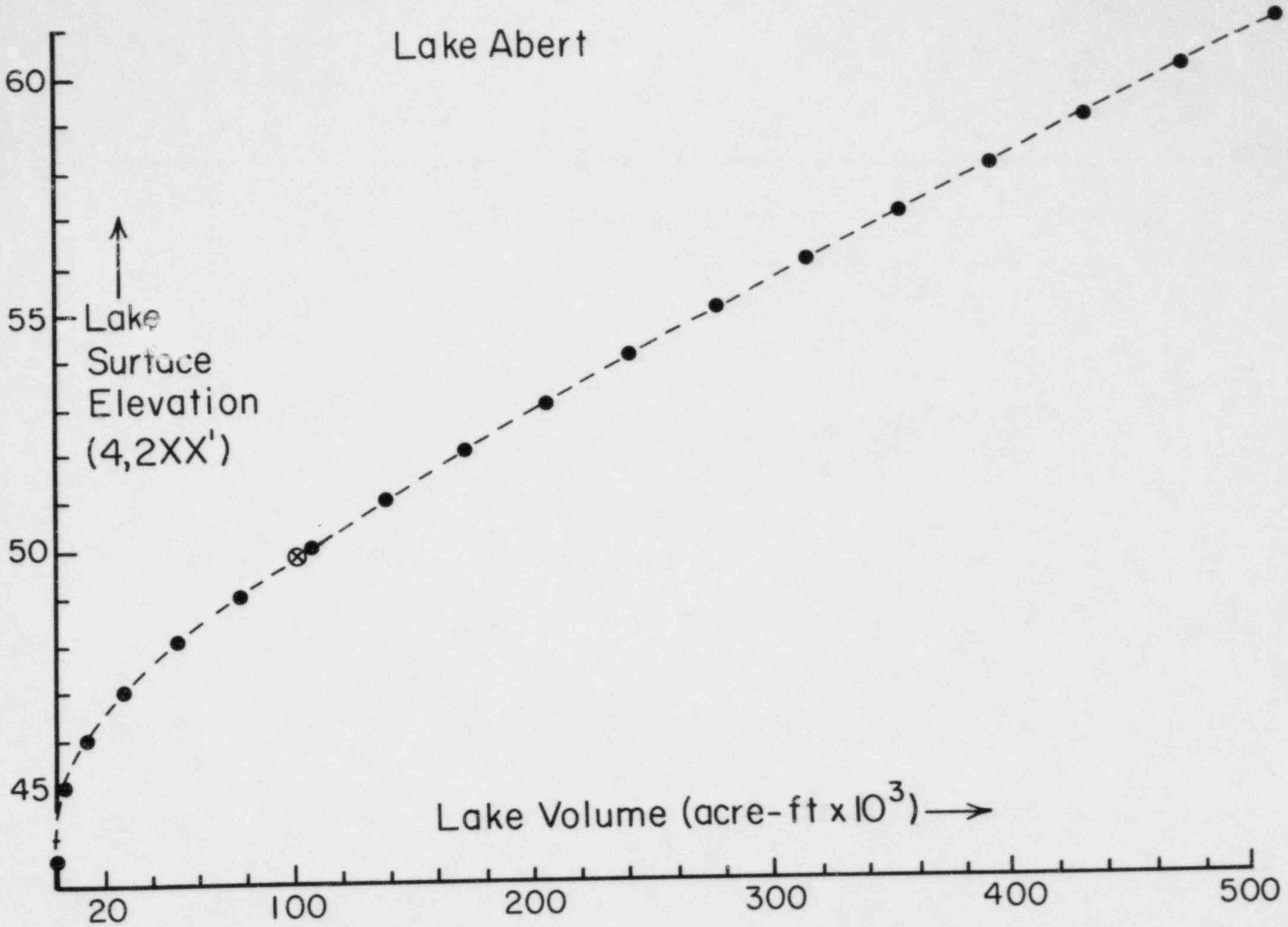

Lake Abert
60
55
50
45
Lake Surface Elevation (4,2XX')
Lake Volume (acre-ft x 10^3)
20
100
200
300
400
500

Figure A-2.

Table A-2

Alkaline Lake Radionuclide Inventories

	Mono Lake	Abert Lake
Dissolved ^{239,240}Pu (mCi)	3.5x10^{1}	1.4x10^{0}
Dissolved ^{241}Am (mCi)	4.9x10^{0}	-
Dissolved ^{137}Cs (mCi)	5.4x10^{3}	(3x10^{2})
Dissolved ^{90}Sr (mCi)	4.3x10^{2}	-
Tritium (mCi)*	6.1x10^{5}	1.5x10^{4}
Sediment ^{239,240}Pu (mCi)	1.4x10^{1}	6.5x10^{1}
Sediment ^{137}Cs (mCi)	1.6x10^{2}	4.3x10^{3}
Sediment ^{90}Sr (mCi)	(3.3x10^{3})	-
Total lake ^{239,240}Pu (mCi)	4.9x10^{1}	6.6x10^{1}
Total lake ^{137}Cs (mCi)	5.6x10^{3}	(4.6x10^{3})
Total lake ^{90}Sr (mCi)	(3.7x10^{3})	-
Total lake ^{239,240}Pu/^{137}Cs	9x10^{-3}	1.4x10^{-2}

*To obtain pCi/m^{3} from T.U., multiply by 3.24x10^{3}

Table A-3

Fallout Radionuclide Inventory in Mono Lake Sediments

Laboratory Control #	Core #	Depth (cm)	Sample wt. (g)	Dry Weight Density (g/cm^3)	^{137}Cs (pCi/kg)	^{137}Cs (mCi/km^2)	^{239,240}Pu (pCi/kg)	^{239,240}Pu (mCi/km^2)
1373A	1	0-1	6.4	0.32	132	0.42	12.9	0.041
B	"	1-2	9.0	0.45	275	1.24	18.8	0.085
C	"	2-3	6.7	0.34	260	0.87	20.6	0.069
D	"	3-4	7.1	0.36	16	0.06	N.D.	-
E	"	4-5	6.5	0.33	60	0.20	N.D.	-
						2.79		0.195
1433A	2A	0-1	5.9	0.30	77	0.23	3.7	0.011
B	"	1-2	7.8	0.39	29	0.11	1.3	0.005
						0.34		0.016
1436A	3A	0-1	4.4	0.22	54	0.12	4.8	0.011
B	"	1-2	3.7	0.19	68	0.13	N.D.	-
						0.25		0.011
1439A	4A	0-1	3.5	0.18	150	0.26	13.5	0.024
B	"	1-2	4.1	0.21	110	0.23	N.D	-
C	"	2-3	4.1	0.21	130	0.27	0.6	0.001
						0.76		0.025
1551A	5A	0-1	10.6	0.53	120	0.64	3.3	0.017
B	"	1-2	11.4	0.57	145	0.83	6.7	0.038
C	"	2-3	9.1	0.46	195	0.89	32.9	0.150
D	"	3-4	4.4	0.22	80	0.18	2.5	0.006
E	"	4-5	8.8	0.44	5	0.02	N.D.	-
F	"	5-7	13.7	0.34	32	0.21	N.D.	-
G	"	7-9	13.6	0.34	69	0.47	N.D.	-
H	"	9-11	13.4	0.34	45	0.31		-
						3.55		0.211

Table A-3 (Cont'd)

Fallout Radionuclide Inventory in Mono Lake Sediments

Laboratory Control #	Core #	Depth (cm)	Sample wt. (g)	Dry Weight Density (g/cm^3)	^{137}Cs (pCi/kg)	^{137}Cs (mCi/km^2)	^{239,240}Pu (pCi/kg)	^{239,240}Pu (mCi/km^2)
1677A	6A	0-1	15.4	0.77	146	1.12	7.0	0.05
B	"	1-2	14.2	0.71	122	0.87	14.4	0.10
C	"	2-3	15.0	0.75	231	1.73	101.7	0.76
D	"	3-4	11.1	0.56	-	-	10.9	0.06
E	"	4-5	10.7	0.54	131	0.70	-	
F	"	5-6	10.3	0.52	86	0.44	-	
G	"	6-7	11.9	0.60	128	0.76	-	
H	"	7-8	13.1	0.66	30	0.20	-	
I	"	8-9	11.3	0.57	60	0.34	-	
J	"	9-11	20.7	0.52	32	0.33	-	
						6.49		0.87
1675A	6B	0-1	14.4	0.72	168	1.21	-	
B	"	1-2	17.9	0.90	210	1.88	-	
C	"	2-3	14.4	0.72	119	0.86	-	
D	"	3-4	9.3	0.47	83	0.39	-	
E	"	4-5	10.4	0.52	64	0.33	-	
F	"	5-6	10.8	0.54	104	0.56	-	
G	"	6-7	10.5	0.53	5	0.03	-	
H	"	7-8	11.6	0.58	122	0.71	-	
I	"	8-9	10.8	0.54	121	0.65	-	
J	"	9-11	19.6	0.49	20	0.20	-	
K	"	11-13	18.7	0.47	25	0.24	-	
L	"	13-15	17.3	0.43	70	0.60	-	
M	"	15-20	36.3	0.36	37	0.67	-	
N	"	20-25	53.6	0.54	24	0.65	-	
						8.98		

Table A-3 (Cont'd)

Fallout Radionuclide Inventory in Mono Lake Sediments

Laboratory Control #	Core #	Depth (cm)	Sample wt. (g)	Dry Weight Density (g/cm^3)	^{137}Cs (pCi/kg)	^{137}Cs (mCi/km^2)	^{239,240}Pu (pCi/kg)	^{239,240}Pu (mCi/km^2)
1678A	7	0-1	12.7	0.64	110	0.70	6.8	0.04
B	"	1-2	15.0	0.75	112	0.84	8.5	0.06
C	"	2-3	14.5	0.73	287	2.08	17.3	0.16
D	"	3-4	11.3	0.57	420	2.37	85.6	0.49
E	"	4-5	12.1	0.61	167	1.01	-	
F	"	5-6	11.1	0.56	110	0.61	-	
G	"	6-7	9.8	0.49	176	0.86	-	
H	"	7-8	10.5	0.53	118	0.62	-	
I	"	8-9	11.4	0.57	158	0.90	-	
J	"	9-11	23.9	0.60	71	0.85	-	
K	"	11-13	20.3	0.51	107	1.09	-	
L	"	13-15	18.1	0.45	75	0.68	-	
M	"	15-20	42.6	0.43	34	0.72	-	
N	"	20-25	52.4	0.52	17	0.45	-	
O	"	25-30	44.7	0.45	37	0.83	-	
						14.61		0.75
1679A	8A	0-1	15.7	0.79	110	0.86	9.7	0.08
B	"	1-2	11.9	0.60	42	0.25	18.3	0.11
C	"	2-3	14.0	0.70	285	2.00	22.8	0.16
D	"	3-4	10.2	0.51	200	1.02	229.4	1.17
E	"	4-5	11.9	0.60	40	0.24	-	
F	"	5-6	-	-	-	-	-	
G	"	6-7	13.5	0.68	3	0.02	-	
H	"	7-8	11.1	0.56	164	0.91	-	
I	"	8-9	12.4	0.62	97	0.60	-	
						5.90		1.52

Table A-3 (Cont'd)

Fallout Radionuclide Inventory in Mono Lake Sediments

Laboratory Control #	Core #	Depth (cm)	Sample wt. (g)	Dry Weight Density (g/cm^3)	^{137}Cs (pCi/kg)	^{137}Cs (mCi/km^2)	^{239,240}Pu (pCi/kg)	^{239,240}Pu (mCi/km^2)
1680A	8B	0-1	17.0	0.85	63	0.54	-	
B	"	1-2	11.2	0.56	410	2.30	-	
C	"	2-3	9.8	0.49	-	-	-	
D	"	3-4	11.2	0.56	98	0.55	-	
E	"	4-5	10.7	0.54	-	-	-	
F	"	5-6	13.0	0.65	-	-	-	
G	"	6-7	11.6	0.58	177	1.03	-	
						4.42		
CN1681A	9A	0-1	15.5	0.78	49	0.38	7.2	0.06
B	"	1-2	11.7	0.59	99	0.58	13.7	0.08
C	"	2-3	11.9	0.60	333	1.98	71.7	0.43
D	"	3-4	12.3	0.62	73	0.45	14.0	0.09
E	"	4-5	19.2	0.96	58	0.56	-	
F	"	5-6	9.9	0.50	15	0.07	-	
G	"	6-7	15.7	0.79	74	0.58	-	
H	"	7-8	11.6	0.58	-	-	-	
I	"	8-9	11.8	0.59	-		-	
J	"	9-11	21.5	0.53	105		-	
K	"	11-13	19.5	0.49	105		-	
						4.60		0.66
1682A	9B	0-1	17.9	0.90	99	0.89	-	
B	"	1-2	13.2	0.66	253	1.67	-	
C	"	2-3	16.8	0.84	31	0.26	-	
D	"	3-4	17.2	0.86	62	0.53	-	
E	"	4-5	14.9	0.75	76	0.57	-	
F	"	5-6	11.4	0.57	115	0.66	-	
G	"	6-7	11.8	0.59	159	0.94	-	
H	"	7-8	13.9	0.70	75	0.52	-	
I	"	8-9	11.8	0.59	-	-	-	
J	"	9-11	23.7	0.59	-	-	-	
K	"	11-13	20.0	0.50	37	-	-	
						6.04		

Table A-4

Fallout Radionuclide Inventory in Lake Abert Sediments

Laboratory Control #	Core #	Depth (cm)	Sample wt. (g)	Dry Weight Density (g/cm^3)	^{137}Cs (pCi/kg)	^{137}Cs (mCi/km^2)	^{239,240}Pu (pCi/kg)	^{239,240}Pu (mCi/km^2)
1690A	1	0-1	6.7	0.34	2195	7.4	-	
B	"	1-2	14.3	0.72	2040	14.7	-	
C	"	2-3	18.2	0.91	2261	20.6	-	
D	"	3-4	15.9	0.80	2847	22.8	-	
E	"	4-5	16.5	0.83	2680	22.2	-	
F	"	5-6	19.6	0.98	2226	21.8	-	
G	"	6-7	17.2	0.86	3010	25.9	-	
H	"	7-8	15.3	0.77	2419	18.6	-	
I	"	8-9	14.2	0.71	2489	17.7	-	
J	"	9-11	26.4	0.66	2910	38.4	-	
K	"	11-13	25.2	0.63	2781	35.0	-	
L	"	13-15	30.1	0.75	1923	28.9	-	
M	"	15-20	105.0	1.05	307	16.1	-	
N	"	20-22	41.5	1.03	77	1.6	-	
						291.7		
1691A	1B	0-1	3.1	0.16	1483	2.2	-	
B	"	1-2	11.9	0.60	1948	11.5	-	
C	"	2-3	15.1	0.76	2222	16.9	-	
D	"	3-4	16.8	0.84	2150	18.1	-	
E	"	4-5	15.8	0.79	2302	18.2	-	
F	"	5-6	15.3	0.77	3196	24.6	-	
G	"	6-7	14.1	0.71	2882	20.2	-	
H	"	7-8	15.6	0.78	2849	22.2	-	
I	"	8-9	14.5	0.73	3255	23.4	-	
J	"	9-11	29.1	0.73	2722	39.6	-	
K	"	11-13	28.5	0.71	2877	41.0	-	
L	"	13-15	30.5	0.76	2678	40.8	-	
M	"	15-20	83.5	0.84	159	6.6	-	
N	"	20-25	91.9	0.92	998	45.9	-	
						331.2		

Table A-4 (Cont'd)

Fallout Radionuclide Inventory in Lake Abert Sediments

Laboratory Control #	Core #	Depth (cm)	Sample wt. (g)	Dry Weight Density (g/cm^3)	^{137}Cs (pCi/kg)	^{137}Cs (mCi/km^2)	^{239,240}Pu (pCi/kg)	^{239,240}Pu (mCi/km^2)
1692A	2A	0-1	12.5	0.63	1627	10.2	-	
B	"	1-2	10.7	0.54	2110	11.3	-	
C	"	2-3	10.7	0.54	2094	11.2	-	
D	"	3-4	7.9	0.40	1909	7.5	-	
E	"	4-5	13.8	0.69	1304	9.2	-	
F	"	5-6	15.7	0.79	819	6.4	-	
G	"	6-7	18.8	0.94	734	6.9	-	
H	"	7-8	21.5	1.08	885	9.5	-	
I	"	8-9	21.8	1.09	531	5.8	-	
J	"	9-11	32.9	0.82	324	5.3	-	
K	"	11-13	31.1	0.78	217	3.4	-	
L	"	13-17	77.0	0.96	44	1.7	-	
						88.4		
1650A	2B	0-1	7.2	0.36	1456	5.2	24.6	0.09
B	"	1-2	10.7	0.54	1815	9.7	29.0	0.16
C	"	2-3	8.9	0.44	1962	8.7	32.3	0.14
D	"	3-4	16.1	0.81	2007	16.1	35.9	0.29
E	"	4-5	14.0	0.70	2185	15.3	-	
F	"	5-6	15.0	0.75	2105	15.8	-	
G	"	6-7	13.6	0.68	1987	13.5	-	
H	"	7-8	12.3	0.62	1692	10.4	-	
I	"	8-9	16.9	0.85	941	8.0	-	
J	"	9-11	29.7	0.74	821	12.2	-	
K	"	11-13	30.1	0.75	669	10.1	-	
L	"	13-15	31.7	0.79	386	6.1	-	
M	"	15-20	101.5	1.02	121	6.1	-	
						137.2		0.68

Table A-4 (Cont'd)

Fallout Radionuclide Inventory in Lake Abert Sediments

Laboratory Control #	Core #	Depth (cm)	Sample wt. (g)	Dry Weight Density (g/cm^3)	^{137}Cs (pCi/kg)	^{137}Cs (mCi/km^2)	^{239,240}Pu (pCi/kg)	^{239,240}Pu (mCi/km^2)
1694A	3A	0-1	18.2	0.91	854	7.8	-	
B	"	1-2	17.5	0.88	312	2.7	-	
C	"	2-3	15.3	0.77	327	2.5	-	
D	"	3-4	14.1	0.71	285	2.0	-	
E	"	4-5	15.6	0.78	243	1.9	-	
F	"	5-6	14.1	0.71	125	0.9	-	
G	"	6-7	14.4	0.72	123	0.9	-	
H	"	7-8	13.9	0.70	186	1.3	-	
I	"	8-9	16.1	0.81	72	0.6	-	
J	"	9-14	36.3	0.36	15	0.3	-	
						20.9		
1695A	3B	0-1	11.8	0.59	826	4.9	-	
B	"	1-2	16.9	0.85	451	3.8	-	
C	"	2-3	16.5	0.83	304	2.5	-	
D	"	3-4	14.3	0.72	191	1.4	-	
E	"	4-5	18.5	0.93	79	0.7	-	
F	"	5-6	16.1	0.81	34	0.3	-	
G	"	6-7	17.3	0.87	-	-	-	
H	"	7-8	17.2	0.86	41	0.4	-	
						14.0		

Table A-4 (Cont'd)

Fallout Radionuclide Inventory in Lake Abert Sediments

Laboratory Control #	Core #	Depth (cm)	Sample wt. (g)	Dry Weight Density (g/cm^3)	^{137}Cs (pCi/kg)	^{137}Cs (mCi/km^2)	^{239,240}Pu (pCi/kg)	^{239,240}Pu (mCi/km^2)
1651A	4	0-1	8.2	0.41	1483	5.9	13.0	0.053
B	"	1-2	9.6	0.48	1776	8.5	12.6	0.060
C	"	2-3	10.8	0.54	1852	10.0	16.0	0.086
D	"	3-4	13.8	0.69	1368	9.4	15.7	0.108
E	"	4-5	13.1	0.66	1458	9.6	14.8	0.098
F	"	5-6	11.4	0.57	1365	7.8	17.2	0.098
G	"	6-7	15.7	0.79	1973	15.6	34.0	0.269
H	"	7-8	12.2	0.61	1807	11.0	17.7	0.108
I	"	8-9	13.4	0.67	1189	8.0	16.7	0.112
J	"	9-11	31.5	0.78	1661	26.2	21.7	0.339
K	"	11-13	28.3	0.71	962	13.6	39.8	0.563
L	"	13-15	34.4	0.86	601	10.3	19.3	0.332
M	"	15-20	75.0	0.75	264	9.9	0.93	0.035
N	"	20-25	78.8	0.79	132	5.2	0.11	0.004
O	"	25-30	79.5	0.80	19	-	-	-
P	"	30-35	91.9	0.92	11	-	-	-
Q	"	35-40	88.3	0.88	-	-	-	-
R	"	40-45	94.3	0.94	5	-	-	-
						151.0		2.27

Table A-5

Alkaline Lake Radionuclide Inventories in Sediments

	^{137}Cs(mCi/km^2)	^{239,240}Pu(mCi/km^2)	^{239,240}Pu/^{137}Cs
Mono Lake Core #			
1	2.8	0.20	0.071
2A	0.34	0.016	0.047
3A	0.25	0.011	0.044
4A	0.76	0.025	0.033
5A	3.55	0.211	0.059
6A	6.5	≥0.87	≥0.134
6B	9.0	-	-
7	14.6	≥0.75	≥0.051
8A	5.9	≥1.52	≥0.26
8B	4.4	-	-
9A	≥4.6	≥0.66	≥0.143
9B	6.0	-	-
Lake Abert Core #			
1	292	-	-
1B	≥331	-	-
2A	88	-	-
2B	137	≥0.68	≥0.0050
3A	21	-	-
3B	14	-	-
4	151	2.27	0.0150

mCi/km^2. The range of ^{137}Cs accumulation rates in Mono L. was from 0.25 mCi/km^2 to ≈ 15 mCi/km^2, while in L. Abert the sediment ^{137}Cs inventories were much greater (14-330 mCi/km^2). For ^{239,240}Pu, the sediment accumulation rates varied from 0.01-1.5 mCi/km^2 for Mono L. and 0.7-2.3 mCi/km^2 for L. Abert.

Our coring sites were not chosen to obtain samples of the lake sediments which were equally representative of the different depositional environments. Instead we generally selected sites which were expected to have high accumulation rates to obtain sediment samples indicative of the fine-particle activities during the years of maximum fallout. Thus the total sediment inventory for the lakes should not be derived simply by averaging accumulation rates for all the cores and multiplying by lake surface area. In the case of Mono L., the cores collected in the deep basin south of Paoha Island have much higher fallout nuclide accumulation rates than for the rest of the lake. Lake Abert has its highest fallout nuclide accumulation rates near the stream input at the south end of the lake, with relatively high values also in the deepest central area. Considering these uncertainties, we have estimated total fallout nuclides in the sediments as follows:

1) Mono Lake

A) ^{137}Cs - average accumulation rate for 12 cores = 4.9 mCi/km^3.
^{137}Cs - lake average estimate, assuming high deposition areas represent ≈ 10% of total area (cores 1, 5A, 6A, 6B, 7, 8A, 8B, 9A, 9B - average 6.4 mCi/km^2) and low deposition areas represent ≈ 90% of total area (cores 2A, 3A, 4A - average 0.45 mCi/km^2) = 1 mCi/km^2.
1 mCi/km^2 x 160 km^2 = 160 mCi.

B) ^{239,240}Pu - average accumulation rate for 9 cores = 0.48 mCi/km^2.

^{239,240}Pu - lake average estimate, assuming high deposition areas represent ≈ 10% of total area (cores 1, 5A, 6A, 7, 8A, 9A - average 0.70 mCi/km^2) and low deposition areas represent ≈ 90% of total area (cores 2A, 3A, 4A - average 0.017 mCi/km^2) = 0.09 mCi/km^2.
0.09 x 160 = 14 mCi.

2) Lake Abert

A) ^{137}Cs - average accumulation rate for 7 cores = 148 mCi/km^2.
^{137}Cs - lake average estimate, assuming high deposition areas represent ≈ 10% of total area (cores 1, 1B, 2A, 2B, 4 - average 200 mCi/km^2) and low deposition areas represent ≈ 90% of total area (cores 3A, 3B - average 18 mCi/km^2 = 36 mCi/km^2
36 mCi/km^2 x 120 km^2 = 4300 mCi.

B) ^{239,240}Pu - accumulation rate for 1 core = 2.3 mCi/km^2.
^{239,240}Pu - lake average estimate, assuming value of 36 mCi/km^2 for ^{137}Cs and a ^{239,240}Pu/^{137}Cs ratio of 0.0150 (core # 4) = 0.54 mCi/km^2.
0.54 mCi/km^2 x 120 km^2 = 65 mCi.

We have included several numerical values in Table A-2 in addition to those discussed above. We included estimates for both total lake (sediments plus dissolved) ^{90}Sr and sediment ^{90}Sr, by assuming a ratio of ^{137}Cs/^{90}Sr = 1.5 for the input of these fallout nuclides to Mono L. We also derived a value for total lake ^{137}Cs in L. Abert, by assuming that dissolved ^{137}Cs concentrations in L. Abert were equal to those in Mono L.

The total quantities (including the water column and sediments) of ^{239,240}Pu in Mono L. and L. Abert were thus estimated to be 49 mCi and 66 mCi, respectively. For ^{137}Cs the total inventories were estimated to be 5.6×10^3 mCi and 4.6×10^3 mCi. These "measured" inventories can be compared with inputs obtained by multiplying fallout delivery rates to lake surface area during the years of maximum fallout. A recent attempt to make fallout delivery estimates in Utah (Beck and Krey, 1983) reported a simple linear relationship between total fallout delivery and mean annual precipitation rate. Using the relationship derived for Utah, we computed total fallout delivery to Mono L. and L. Abert of about 4-5 times the amounts we measured for ^{239,240}Pu and about three times as much for ^{137}Cs (Table A-6). Estimates of total fallout delivery based on ^{90}Sr deposition in Washington, Oregon and California during the mid-1960's (Table A-6) were consideraly lower, yielding amounts quite comparable to the inventories we derived from measurements in the water column and sediments of Mono L. and L. Abert. Thus the total quantities of fallout ^{239,240}Pu and ^{137}Cs observed in these alkaline lakes are generally compatible with regional fallout delivery rates for the states of Washington, Oregon and California. We conclude that the unusually high ^{239,240}Pu concentrations observed in the water column of Mono L. and L. Abert result from complexing by carbonate ion, and not from the delivery of any large "extra" component of fallout.

Table A-6

Estimated Fallout Delivery of Radionuclides

Nuclide	mCi/km^2 (P=20 cm/yr)	mCi (P=20 cm/yr) (A=200 km^2)	mCi/km^2 (P=25 cm/yr)	mCi P=25 cm/yr (A=150 km^2)
$^{137}Cs^a$	70	$1.4x10^4$	82	$1.2x10^4$
$^{239,240}Pu^b$	1.3	$2.6x10^2$	1.5	$2.3x10^2$
$^{137}Cs^c$	31	$6.2x10^3$	34	$5.1x10^3$
$^{239,240}Pu^c$	0.59	$1.2x10^2$	0.64	$9.6x10^1$
$^{137}Cs^d$	20	$4.0x10^3$	22	$3.3x10^3$
$^{239,240}Pu^d$	0.38	$7.6x10^1$	0.42	$6.3x10^1$
$^{137}Cs^e$	18	$3.6x10^3$	19	$2.9x10^3$
$^{239,240}Pu^e$	0.34	$6.8x10^1$	0.36	$5.4x10^1$

Table A-6 (Cont'd)

Estimated Fallout Delivery of Radionuclides

a) Beck and Krey (1983) reported the following relationship for fallout ^{137}Cs delivery (decay corrected to 1979) and mean annual precipitation rate (P in cm/yr) for Utah (15-50 cm/yr of annual precipitation):

$$^{137}\text{Cs (mCi/km}^2\text{)} = 2.22\text{xP}+26 \text{ mCi/km}^2$$

b) Beck and Krey (1983) reported a ratio of ^{137}Cs to $^{239,240}Pu$ in integrated global fallout for samples collected in 1979 of 53±0.5. Expressed as the reciprocal, this value gives a $^{239,240}Pu$ to ^{137}Cs ratio of $1.89\text{x}10^{-2}$.

c) Simpson (1970) found the following relationship for ^{90}Sr deposition vs. annual precipitation in cm for 5 sites in the state of Washington during the mid 1960's.

$$^{90}\text{Sr (mCi/km}^2\text{)} = 0.46\text{xP}+20 \text{ mCi/km}^2$$

^{137}Cs in 1980 can be estimated from the above expression for ^{90}Sr by multiplying by 1.5 and dividing by 1.41 to correct for radioactive decay to 1980. $^{239,240}Pu$ to ^{137}Cs was assured to = $1.89\text{x}10^{-2}$

d) Simpson (1970) found the following relationship for ^{90}Sr deposition vs. annual precipitation in cm for 2 sites in the state of Oregon during the mid 1960's.

$$^{90}\text{Sr (mCi/km}^2\text{)} = 0.36\text{xP}+12 \text{ mCi/km}^2$$

^{137}Cs and $^{239,240}Pu$ deposition in 1980 can be estimated as described in footnote c.

e) Simpson (1970) found the following relationship for ^{90}Sr deposition vs. annual precipitation in cm for 3 sites in the state of California during the mid 1960's.

$$^{90}\text{Sr (mCi/km}^2\text{)} = 0.28\text{xP}+11 \text{ mCi/km}^2$$

^{137}Cs and $^{239,240}Pu$ deposition in 1980 can be estimated as described in footnote C.

REFERENCES

Allard, B., 1982. Solubilities of actinides in neutral or basic solutions, N. Edelstein (Ed.) *Actinides in Perspective*, Pergamon Press, Oxford, pp. 553-580.

Anderson, R. F., 1980. The marine geochemistry of thorium and protactinium. Ph.D. Thesis, Massachusetts Institute of Technology-Woods Hole Oceanographic Institution Joint Program in Oceanography, Woods Hole, Massachusetts. 287 pp.

Anderson, R. F., M. P. Bacon and P. G. Brewer, 1982. Elevated concentrations of actinides in Mono Lake. Science, 216: 514-516.

Bacon, M. P., D. W. Spencer and P. G. Brewer, 1976. ^{210}Pb/^{226}Ra and ^{210}Po/^{210}Pb disequilibria in seawater and suspended particulate matter. Earth Planet. Sci. Lett., 32: 277.

Beck, H. L., and P. W. Krey, 1983. Radiation exposures in Utah from Nevada nuclear tests. Science, 220: 18-24.

Benninger, L., 1976. The uranium series radionuclides as tracers of geochemical processes in Long Island Sound. Ph.D. Thesis, Yale University, New Haven, Connecticut. 151pp.

Bhat, S. G., S. Krishnaswami, D. Lal, Rama and W. S. Moore, 1969. ^{234}Th/^{238}U ratios in the ocean. Earth Planet. Sci. Lett., 5: 483.

Bowen, V. T., V. E. Noshkin, H. L. Volchok and T. T. Sugihara, 1969. Strontium-90: concentrations in surface waters of the Atlantic Ocean. Science, 164: 825.

Bowen, V. T., V. E. Noshkin, H. D. Livingston and H. L. Volchok, 1980. Fallout radionuclides in the Pacific Ocean: vertical and horizontal distributions, largely from GEOSECS stations. Earth Planet. Sci. Lett., 49: 411.

Broecker, W.S., 1957. Application of radiocarbon to oceanography and climate chronology. Ph.D. Thesis, Columbia University, New York, NY

Broecker, W. S. and A. F. Walton, 1959. Re-evaluation of the salt chronology of several Great Basin lakes. Geol. Soc. Am. Bull., 31:411-419.

Bruland, K. W., M. Koide and E. D. Goldberg, 1974. The comparative marine geochemistries of lead-210 and radium-226. J. Geophys. Res., 79:3083.

Christensen, M.N., C.M. Gilbert, K.R. Lajoie, and Al.-R. Yehya, 1969. Geological-geophysical interpretation of Mono Basin, California-Nevada. J. Geophys. Res., 74: 5221-5239.

Duursma, E. K. and C. J. Bosch, 1970. Theoretical, experimental and field studies concerning diffusion of radioisotopes in sediments and suspended particles of the sea. Part B: methods and experiments. Netherlands J. of Sea Res., 4: 395-469.

Edmond, J.M., 1970. High precision determination of titration alkalinity and total carbon dioxide content of seawater by potentiometric titration. Deep Sea Res., 17: 737-750.

Garrels, R.M. and Mackenzie, F.T., 1967. Origin of the chemical composition of some springs and lakes. In: Equilibrium Concepts in Natural Water Systems (R. F. Gould, ed.). American Chemical Society, Advances in Chemistry, Series No. 67, Washington, D. C. pp. 222-242.

Hutchinson, G. E., 1957. A Treatise on Limnology, Vol. I. Geography, Physics and Chemistry. John Wiley and Sons, New York. 1015 pp.

Jones, B. F., 1966. Geochemical evolution of closed basin water in the western Great Basin. In: Second Symposium on Salt (J. L. Rau, ed.). Cleveland, Ohio, Northern Ohio Geological Society, Vol. 1, pp. 181-199.

Kaufman, A., 1969. The ^{232}Th concentration of surface ocean water. Geochim. Cosmochim. Acta, 33: 717-724.

Kaufman, A., R. M. Trier, W. S. Broecker and H. W. Feely, 1973. Distribution of ^{228}Ra in the world ocean. J. Geophys. Res., 78: 8827.

Knauss, K. G., T. L. Ku and W. S. Moore, 1978. Radium and thorium isotopes in the surface waters of the East Pacific and coastal southern California. Earth Planet. Sci. Lett., 39: 235.

Ku, T.L., 1968. Protactinium 231 method of dating coral from Barbados Islands, J. Geophys. Res., 73: 2271-2276.

Ku, T. L., K. G. Knauss and G. G. Mathieu, 1977. Uranium in the open ocean concentration and isotopic composition. Deep Sea Res., 24:1005.

Langmuir, D. and J.S. Herman, 1980. The mobility of thorium in natural waters at low temperatures, Geochim. Cosmochim. Acta 44: 1753-1766.

Livingston, H. D. and V. T. Bowen, 1976. Americium in the marine environment-relationships to plutonium, in Environmental Toxicity of Aquatic Radionuclides: Models and Mechanisms. Ann Arbor Science, Ann Arbor, Michigan. pp. 107-130.

Mason, D.T., 1967. Limnology of Mono Lake, California. University of California Publications in Zoology, Vol. 83, University of California Press, Berkeley and Los Angeles. 110p.

Moore, W. S. and W. M. Sackett, 1964. Uranium and thorium series in equilibrium in seawater. J. Geophys. Res., 69: 5401.

Moore, W.S., 1981. The thorium isotope content of ocean water. Earth Planet. Sci. Lett., 53: 419.

Nozaki, Y., J. Thomson and K. K. Turekian, 1976. The distribution of ^{210}Pb and ^{210}Po in the surface waters of the Pacific Ocean. Earth Planet. Sci. Lett., 32: 304.

Olsen, C.R., H. J. Simpson, and R.M. Trier 1981. Plutonium, radiocesium, and radiocobalt in sediments of the Hudson River estuary. Earth Planet. Sci. Lett., 55: 377-392.

Phillips, K.N. and A.S. Van Denburgh, 1971. Hydrology and geochemistry of Abert, Summer and Goose Lakes, and other closed-basin lakes in south-central Oregon. U.S. Geological Survey Professional Paper 502-B. 86p.

Roether, W., 1967. Tritium in Wasser Kreislauf. Ph.D. Thesis, University of Heidelberg, West Germany.

Rush, F.E., 1974. Hydrologic regimen of Walker Lake, Mineral County, Nevada. Water Resources Information Series Report 21. Nevada Division of Water Resources.

Russell, I.C., 1885. Geological History of Lake Lahontan. U.S. Geological Survey Monograph, 11: 287pp.

Russell, I.C., 1889. Quaternary history of Mono Valley, California. U.S. Geological Survey Eighth Annual Report: 261-394.

Schell, W. R. and T. H. Sibley, 1982. Distribution coefficients for radionuclides in aquatic environments, final summary of report. NUREG/CR-1869, Office of Nuclear Regulatory Research, U.S. Nuclear Regulatory Commission, Washington, D.C. 21 pp.

Simpson, H. J., 1970. Closed basin lakes as a tool in geochemistry. Ph.D. Thesis, Columbia University, New York. 325 pp.

Simpson, H. J. and T. Takahashi, 1973. Interstitital water studies, Leg 15 - chemical model of seawater and saline water. In: Initial Reports of the Deep Sea Drilling Project, 20, 877 (B. C. Heezen, I. D. MacGregor, eds.). U.S. Govt. Printing Office, Washington, D.C.

Simpson, H. J., R. M. Trier and C. R. Olsen, 1980. Transport of plutonium by rivers. In: Transuranic Elements in the Environment (W. C. Hansen, ed.). Technical Information Center/U.S. Department of Energy (DOE/TIC-22800). pp. 684-690.

Simpson, H. J., R. M. Trier, C. R. Olsen, D. E. Hammond, A. Ege, L. Miller and J. M. Melack, 1980. Fallout plutonium in an alkaline, saline lake. Science, 207: 1071-1073.

Simpson, H. J., R. M. Trier, J. R. Toggweiler, G. Mathieu, B. L. Deck, C. R. Olsen, D. E. Hammond, C. Fuller and T. L. Ku, 1982. Radionuclides in Mono Lake, California. Science, 216: 512-514.

Simpson, H.J., R.M. Trier and Y.-H. Li, 1983, Field experiment determinations of distribution coefficients of actinide elements in alkaline lake environments, Report to Nuclear Regulatory Commission, March 1983, 129 p.

Thurber, D., 1965. The concentration of some natural radioelements in the waters of the Great Basin. Bull. Volcanologique, 28: 1. Napoli, Italy, 1-7.

Van Denburgh. A.S., R.D. Lamke, and J.L. Hughes, 1973. A brief water resources appraisal of the Truckee River Basin, western Nevada. State of Nevada Water Resources - Reconnaissance Series Rept. 57.

Van Denburgh, A.S., 1975. Solute balance at Abert and Summer Lakes, South-Central Oregon. Closed-Basin Investigations, U.S. Geological Survey Professional Paper 502-C. 29pp.

Wahlgren, M.A. and J.S. Marshall, 1975. The behaviour of plutonium and other long-lived radionuclides in Lake Michigan. I. Biological transport, seasonal cycling and residence times in the water column. In: Impacts of Nuclear Releases into the Aquatic Environment. IAEA SM 198/139: 227-243.

Wahlgren, M.A. and D.M. Nelson, 1975. Plutonium in the Laurentian Great Lakes: comparison of surface waters. Verh. Internat. Verein. Limnol., 19: 317-322.

Wahlgren, M.A., J.J. Alberts, D.M. Nelson, and K.A. Orlandini, 1976. Study of the behaviour of transuranics and possible chemical homologues in Lake Michigan water and biota. In: Tranuranium Nuclides in the Environment. IAEA STI/PUB, 410: 9.

Whitehead, H.C. and J.H. Feth, 1961. Recent chemical analyses of waters from several closed basin lakes and their tributaries in the western United States. Geol. Soc. Amer. Bull., 72: 1421-1426.

Wong, K.M., 1971. Radiochemical determination of plutonium in seawater, sediments and marine organisms. Anal. Chim. Acta, 56: 355.

NRC FORM 335
(6-83)

U.S. NUCLEAR REGULATORY COMMISSION

BIBLIOGRAPHIC DATA SHEET

1 REPORT NUMBER *(Assigned by TIDC, add Vol. No., if any)*
NUREG/CR-3940

2 Leave blank

3 TITLE AND SUBTITLE
Field Experiment Determinations of Distribution Coefficients
of Actinide Elements in Alkaline Lake Environments

4 RECIPIENT'S ACCESSION NUMBER

5 DATE REPORT COMPLETED

MONTH	YEAR
March	1983

6 AUTHOR(S)
H. J. Simpson, R. M. Trier, Y.-H. Li, R. F. Anderson, and
A. L. Herczeg

7 DATE REPORT ISSUED

MONTH	YEAR
August	1984

8 PERFORMING ORGANIZATION NAME AND MAILING ADDRESS *(Include Zip Code)*
Lamont-Doherty Geological Observatory
Columbia University
Palisades, NY 10964

9 PROJECT/TASK/WORK UNIT NUMBER

10 FIN NUMBER
B7057

11 SPONSORING ORGANIZATION NAME AND MAILING ADDRESS *(Include Zip Code)*
Division of Radiation Programs and Earth Sciences
Office of Nuclear Regulatory Research
U.S. Nuclear Regulatory Commission
Washington, DC 20555

12a TYPE OF REPORT
Topical

12b PERIOD COVERED *(Inclusive dates)*
N/A

13 SUPPLEMENTARY NOTES
N/A

14 ABSTRACT *(200 words or less)*

Measurements of the radioisotope concentrations of a number of elements (Am, Pu, U, Pa, Th, Ac, Ra, Po, Pb, Cs, and Sr) in the water and sediments of a group of alkaline (pH = 9-10), saline lakes demonstrate greatly enhanced soluble-phase concentrations of elements with oxidation states of (III)-(VI) as the result of complexing by carbonate ion. Ratios of soluble radionuclide concentrations in Mono Lake to those in seawater ($[CO_3^{2-}]$ in Mono Lake = 200 times that of seawater) were: Pu(≈10), ^{238}U(≈150), ^{231}Pa, ^{228}Th, ^{230}Th(≈10^3), and ^{232}Th(≈10^5). Effective distribution coefficients of these radionuclides in high CO_3^{2-} environments are several orders of magnitude lower (i.e., less particle reactive) than in most other natural waters. The importance of CO_3^{2-} ion on effective K_d values was also strongly suggested by laboratory experiments in which most of the dissolved actinide elements became adsorbed to particles after a water sample normally at a pH of 10 was acidified, stripped of all CO_2, and then returned to pH 10 by adding NH_4OH. Furthermore, the effect of complexation by organic ligands is of secondary importance in the presence of appreciable carbonate ion concentration.

Neither pure phase solubility calculations nor laboratory scale K_d determinations accurately predicted the measured natural system concentrations. Therefore, measurements of the distribution of radionuclides in natural systems are essential for assessment of the likely fate of potential releases from high level waste repositories to groundwater.

15a KEY WORDS AND DOCUMENT ANALYSIS

15b DESCRIPTORS

actinide, carbonate complexation, effective distribution coefficient

16 AVAILABILITY STATEMENT
Unlimited

17 SECURITY CLASSIFICATION
(This report)
unclassified

18 NUMBER OF PAGES

19 SECURITY CLASSIFICATION
(This page)

20 PRICE
$

UNITED STATES
NUCLEAR REGULATORY COMMISSION
WASHINGTON, D.C. 20555

OFFICIAL BUSINESS
PENALTY FOR PRIVATE USE, $300

FOURTH-CLASS MAIL
POSTAGE & FEES PAID
USNRC
WASH. D. C.
PERMIT No. G 67

120555078877 1 1AN1RW
US NRC
ADM-DIV OF TIDC
POLICY & PUB MGT BR-PDR NUREG
W-501
WASHINGTON DC 20555

NUREG/CR-3940

FIELD EXPERIMENT DETERMINATIONS OF DISTRIBUTION COEFFICIENTS OF ACTINIDE ELEMENTS IN ALKALINE LAKE ENVIRONMENTS

AUGUST 1984