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THE ORIGIN OF THE  
CHEMICAL COMPOSITION OF  
SANTA FE LAKE, NEW MEXICO

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by

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of the Requirements for the Degree of  
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### Abstract

A mass balance study of precipitation and lake water chemistry from samples collected monthly over a period of one year at Santa Fe Lake, New Mexico, show that about 40% of the  $\text{Ca}^{++}$ ,  $\text{Mg}^{++}$ , and  $\text{Na}^+$  found in the lake water and about 50% of the  $\text{SO}_4^{--}$  is of atmospheric origin. The importance of the atmospheric inputs of  $\text{K}^+$  is unclear due to  $\text{K}^+$  contamination of precipitation by vegetation.

X-ray diffraction analysis of the clay sized fraction of the lake sediment shows that it is mostly amorphous with trace quantities of kaolinite. Analysis of the lake water chemistry using the computer program WATEQ and mineral stability diagrams indicates that kaolinite is thermodynamically stable. Apparently an amorphous geochemical alteration product of kaolinitic composition is being formed.

Concentrations of dissolved constituents found in the lake water beyond that found in precipitation is assumed to come from weathering. Back reacting the dissolved chemical constituents attributable to weathering with kaolinite to produce primary minerals found in the basin shows that chemical breakdown of plagioclase and biotite are the two most important weathering processes in the basin. Approximately 70% of the dissolved solids found in Santa Fe Lake water which originate from weathering can be attributed to the

breakdown of plagioclase.

Because of mobility problems and delayed entry into lake sediments, dating of sediment from Santa Fe Lake as well as four other lakes from northern New Mexico using Cs-137 was unsuccessful. Excessive time delay in transportation of Pb-210 from lake basin soils to lake sediment rendered Pb-210 dating invalid for two of the five lakes (Lagunitas #3 and Sugarloaf Lakes). Pb-210 dating was successful for the remaining three lakes with sedimentation rates of 0.07, 0.08, and 0.13 cm/year for Lost, Santa Fe, and Truchas Lakes respectively.

An attempt at using lithium distribution in Santa Fe Lake sediment to determine trends in weathering rate over time was inconclusive. However, an increasing trend in iron concentration beginning about 160 years ago suggests that weathering rates have been increasing over that time.

## INTRODUCTION

### Purpose:

This study examines the relative importance of atmospheric deposition and geochemical weathering as sources of the dissolved constituents found in the waters of Santa Fe Lake. Determination of the contribution of atmospheric inputs to the overall chemical composition of the lake water will provide insight into the sensitivity of this lake to changes in the composition of precipitation entering the basin. Exactly which weathering reactions were responsible for the production of basin derived inputs were determined to provide a more complete understanding of the origin of the dissolved solutes found in the lake water.

Dating of lake sediments from Santa Fe Lake as well as four other subalpine lakes of the Northern New Mexico Region was performed. Dates obtained for these sediments allow correlation of trace metal concentrations within the lake sediments determined by Lynch et al. (1988) and Svec (in progress) with absolute ages. Comparison of the years in which changes in trace metal deposition trends occur with historical events, may allow future workers to identify the causes of these changes.

### Nature of the Study Area:

Sediment cores from five lakes were dated in this

study. All of the lakes occur at high elevations in areas with high relief and have low lake basin to lake surface area ratios (table 1). For these reasons precipitation has little opportunity to react with dust in the atmosphere or rocks in the lake basins before entering the lakes themselves. Since contact between precipitation and particulate matter is minimal, solutes present in the precipitation are likely to have a significant effect on the chemical composition of these lake waters.

The lakes are located in three distinct areas (figures 1-4). Only Sugarloaf Lake in the Brazos Uplift area is accessible by motor vehicle. However, even this lake receives no motor boat traffic and being on private land receives limited use. Geologic descriptions of the five lake basins are given in Appendix A.

#### Mass Balance Approach to Origin of Chemical Composition

A modified mass balance approach for determining the source of the chemical composition of Santa Fe Lake was used. Since the absolute mass of solutes entering and leaving the lake are not calculated this is not a true mass balance study. This approach has been used by a number of investigators with varying degrees of success (e.g. Garrels and Mackenzie (1967), Bricker et al. (1968), Cleaves et al. (1970), and Reynolds and Johnson

Table 1. Lake elevations, basin areas, surface areas and basin area to surface area ratios.

<u>Lake Name</u>	<u>Elevation (meters)</u>	<u>Basin Area (hectares)</u>	<u>Surface Area (hectares)</u>	<u>Basin area: Surface Area</u>
Santa Fe	3530	15.8	1.9	8.3
Lost	3504	69.9	3.4	20.6
Truchas	3618	32.5	1.0	32.5
Lagunitas #3	3222	24.8	0.3	82.7
Sugarloaf	3005	197.2	2.4	82.2

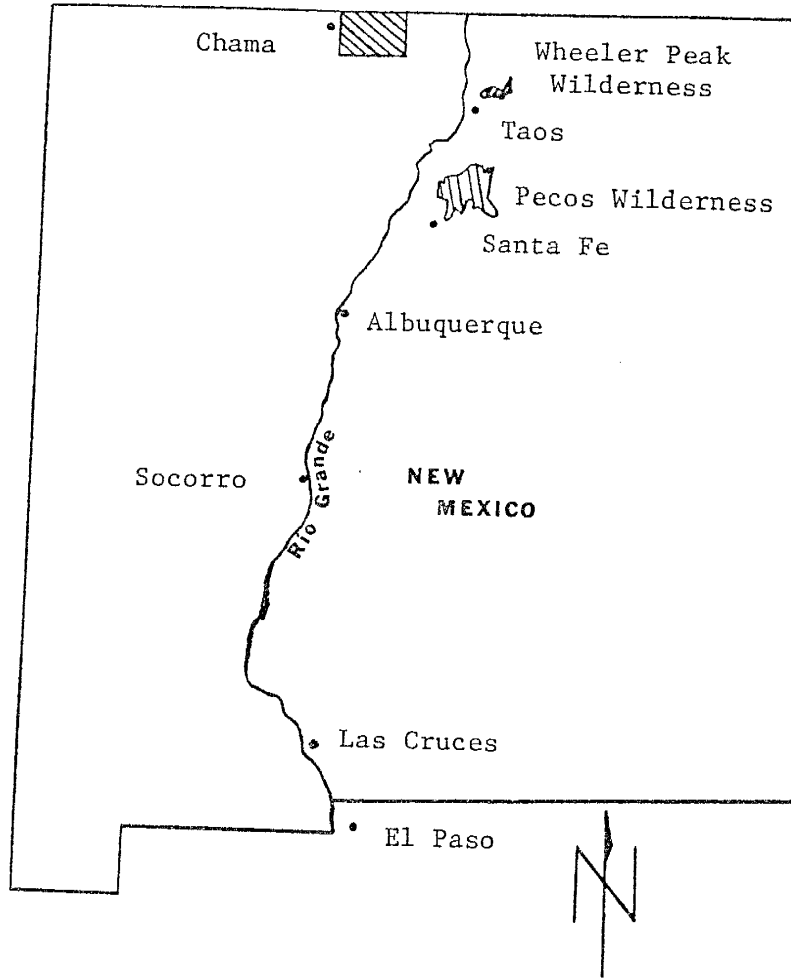


Figure 1. Location map of the three study areas.



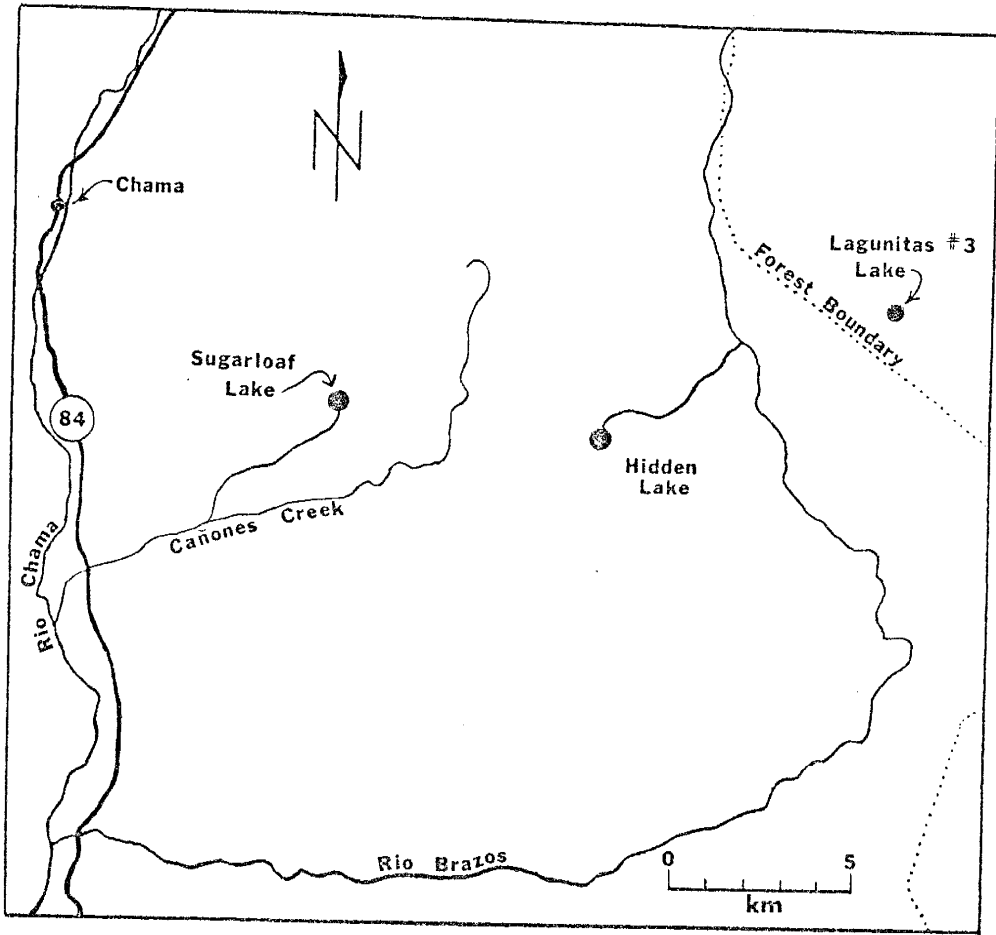


Figure 2. Detailed location map of the Chama area.

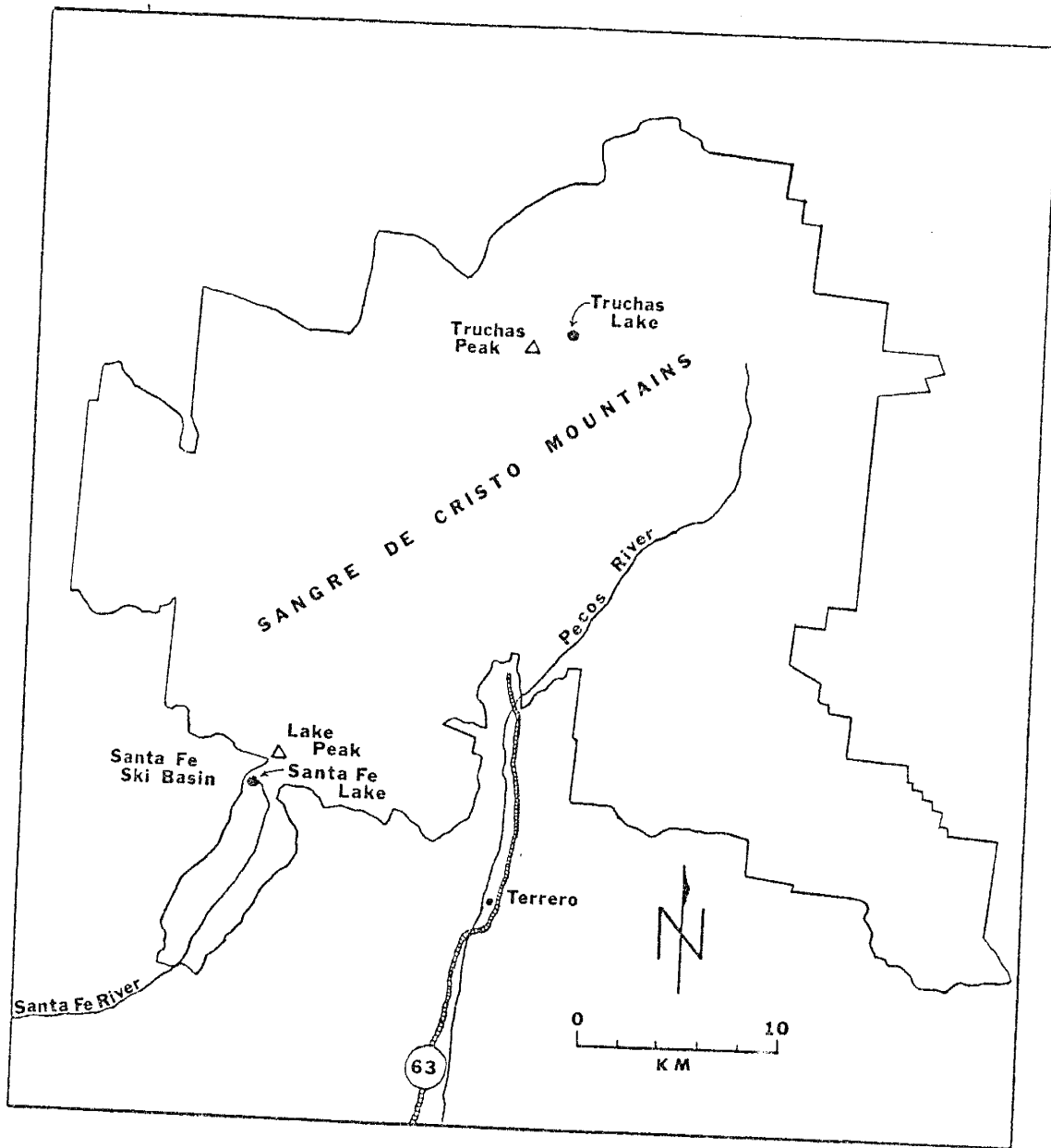


Figure 3. Detailed location map of the Pecos Wilderness study area showing the location of Truchas and Santa Fe Lakes.

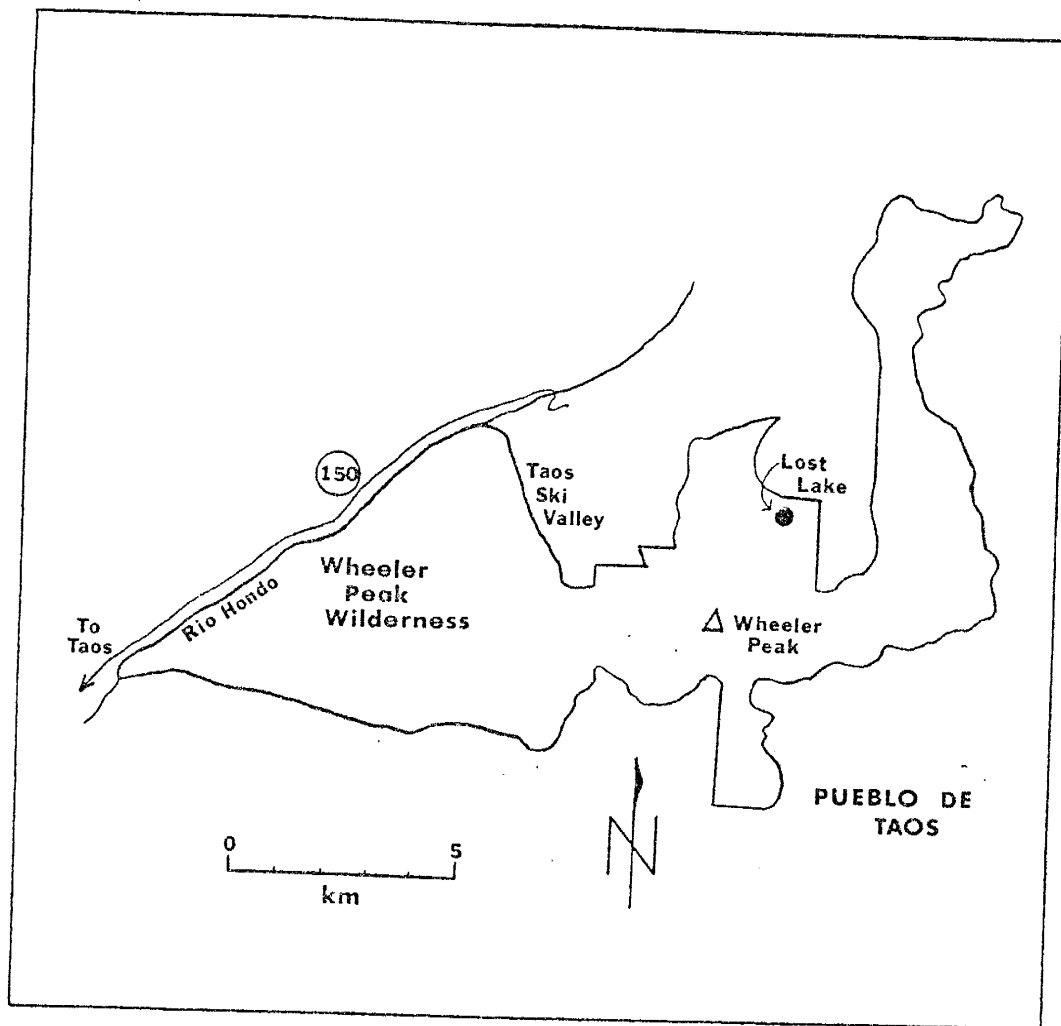


Figure 4. Detailed location map of the Wheeler Peak study area showing the location of Lost Lake.

(1972)). This method involves subtracting the mean concentrations of solutes in precipitation from the mean concentrations in lake water, and assuming differences are a result of inputs due to geochemical weathering reactions. Exactly which reactions are occurring may be determined by identifying the weathering products and then back reacting the products with the solutes to form the minerals present in the basin.

#### Lake Sediment Chronology:

##### Stratigraphic Markers:

Stratigraphic markers are distinctive layers occurring in the sedimentary record. If they can be correlated with an event of known age then they are of use in dating. Dating based on this method must assume constant deposition rate. For example, if a stratigraphic marker associated with a ten year old event occurs ten cm from the surface in a sediment core then the sedimentation rate is assumed to be one cm per year. This technique results in only an average sedimentation rate, and changes in the sedimentation rate with time cannot be detected.

##### Radiometric Dating:

###### Cs-137:

Cs-137 is an artificial nuclide formed by nuclear

fission. It has been used extensively to date recent lake sediments (e.g. Pennington et al. (1973), Robbins and Eddington (1975), Engstrom et al. (1985), Davis et al. (1983), Popp et al. (1988), and Anderson et al. (1987)). It first appeared in the environment in 1945 as a result of above ground nuclear weapons testing. Since then nuclear weapons testing has resulted in a sporadic release of Cs-137 into the atmosphere where it has a mean residence time of about one year after which time it is deposited on to the surface of the earth (Bhandari et al., 1966).

Pennington et al. (1973) determined that the rate of Cs-137 fall-out peaked in 1959 and again in 1963 (figure 5). This sporadic fall-out pattern is recorded in the stratigraphic record. For this reason the first appearance of Cs-137 as well as the 1959 and 1963 peaks can be used as stratigraphic markers. This dating method is only applicable to sediments where Cs-137 has remained immobile since its deposition.

#### Pb-210:

Pb-210 is an unstable intermediate in the U-238 decay series (figure 6) with a half-life of 22.3 years (Krishnaswami and Lal, 1978). Because of this, Pb-210 is very useful for dating lake sediments deposited within the last 100 years. The reliability of Pb-210 dating has been confirmed by many workers (e.g. Koide et al. (1973),

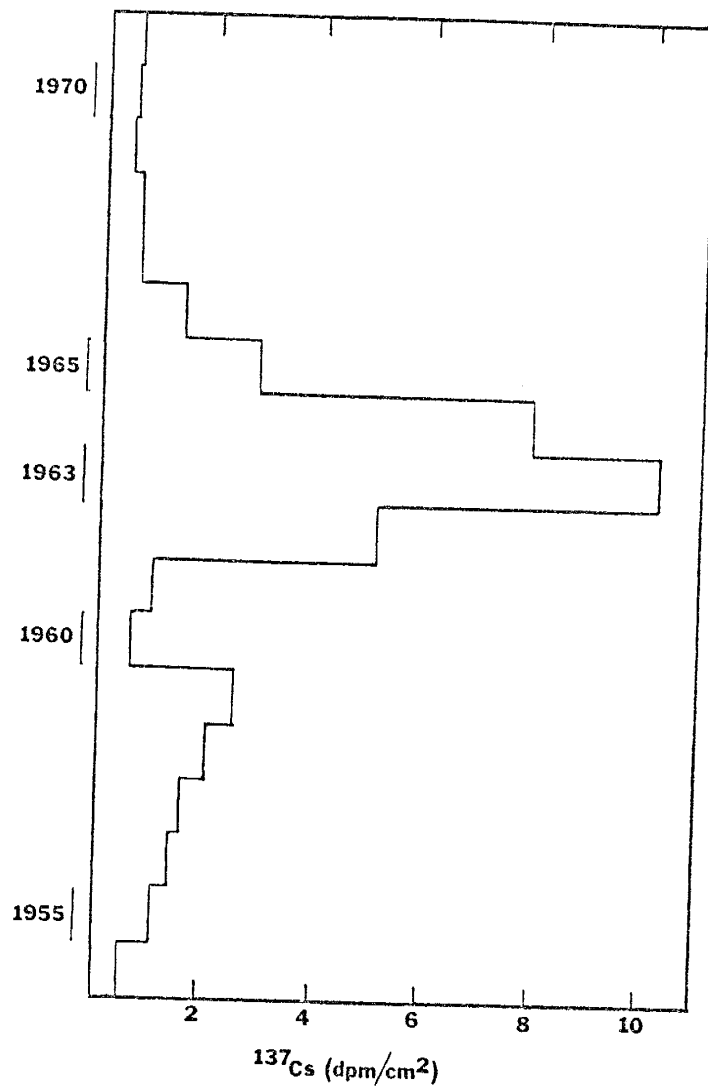


Figure 5. The atmospheric fall-out pattern for Cs-137 at Windermere Lake (Pennington et al., 1973)

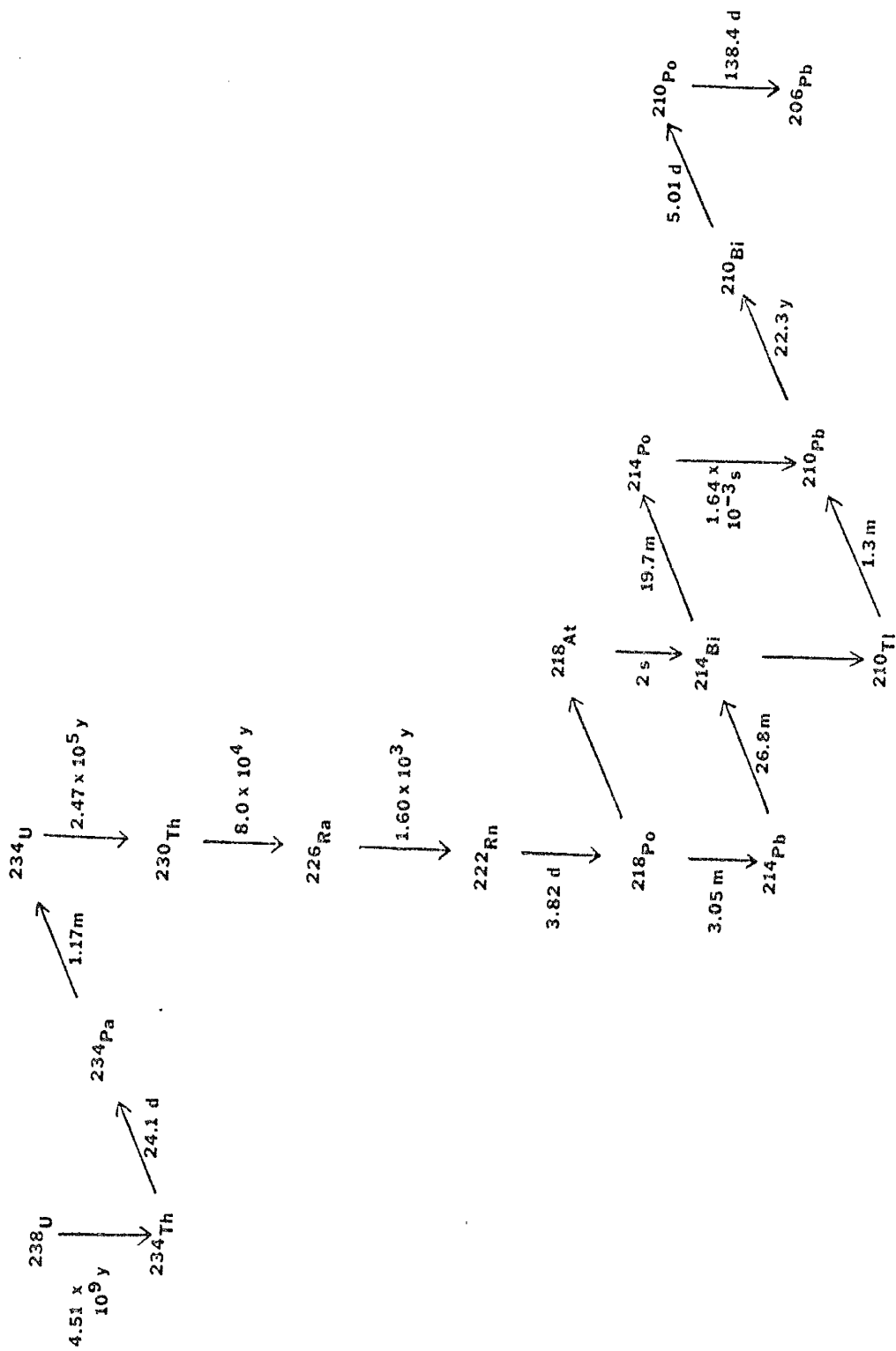


Figure 6. The U-238 decay series (Dehn, 1982).

Matsumoto (1975), Robbins and Eddington (1975), Engstrom et al. (1985), Davis et al. (1983), and Anderson et al. (1987)).

U-238 within the earth's crust decays via several unstable intermediates to Rn-222. Some of this Rn-222, which is a gas, diffuses out of the earth's crust into the atmosphere where it quickly decays to Pb-210. Pb-210 is then rapidly removed from the atmosphere to the earth by both wet and dry deposition.

Pb-210 that falls into a lake is quickly adsorbed onto suspended particles and incorporated into the lake's sediments (Krishnaswami and Lal, 1978). Once a layer of sediment is buried, no new atmospheric Pb-210 can be added to it, and the amount of Pb-210 in that layer will decay over time.

In addition to the atmospheric Pb-210 (hereafter referred to as "unsupported Pb-210"), there is also some Pb-210 which forms within the lake sediments from the decay of nuclides in the U-238 series. This Pb-210 is referred to as "supported Pb-210" because its presence is supported by the presence of other nuclides in the U-238 series.

The activity of supported Pb-210 can be determined by measuring the activity of another nuclide in the U-238 series which is in secular equilibrium with Pb-210. At equilibrium the disintegration rate of a long-lived parent will determine the disintegration rates of all of



its relatively short-lived daughters (Faure, 1986).

Therefore the activities of other gamma emitters in the U-238 series such as Ra-226 or Pb-214 which have half-lives significantly shorter than U-238, should be equal to the activity of supported Pb-210. Since Ra-226 is the parent of Rn-222, Pb-214 was used to determine supported Pb-210 in order to avoid error caused by degassing of Rn-222.

## Methods and Procedures

### Sampling:

Sediment cores for the purpose of dating were obtained using a Brinkhurst core sampler (Brinkhurst et al., 1969). This device consists essentially of a small metal frame which holds a 50 cm long brass coring tube which is 5 cm in diameter. A removable plastic tube is contained within the brass tube and it is therefore within the plastic tube where sediment cores are collected. Originally the sampler contained a plunger with a triggering mechanism which acted to create a suction in the coring tube which would retain the sample. Since this proved to be cumbersome and inefficient the sampler was modified by removing the plunger and triggering mechanism and by threading a lead doughnut onto the line used to lower the sampler to the lake bottom. The sampler, which was suspended from an inflatable raft, was lowered to the bottom partially penetrating the lake sediments. A second line tied to the lead doughnut allowed the doughnut to be raised and lowered thereby pounding the corer further into the lake bottom.

After retrieving the core sampler to the surface the coring tube containing the sediment was removed from the sampler and capped. The cores were extruded in the field

by pushing on the core inside the core tube with a plunger from the bottom of the core up. As the core was extruded it was sliced into 1 cm thick slices and stored in individual plastic sealable Whirl-Pak bags.

Each core slice was labelled with an abbreviation of the lake of origin and its numerical position in the core. For example, the top 1 cm slice from Lost, Sugarloaf, Lagunitas #3, Santa Fe, and Truchas Lakes were labelled L-1, SGL-1, Lag3-1, SF-1, and T-1 respectively. Successive slices were labelled according to depth in the core. For example slices taken 2, 3, and 4 cm from the top of the Lost Lake core were labelled L-2, L-3, and L-4.

Upon returning to the laboratory the slices were frozen until they could be freeze-dried at  $-50^{\circ}\text{C}$  and 25 microtorr in a Labconco Model 5 lypholyzer. The samples were prepared for freeze-drying in a Labconco ethanol bath shell freezer.

Monthly sampling of precipitation was performed for Santa Fe Lake from July 1987 to June 1988. Snow samples were collected by directly scooping the snow into polyethylene bottles. Remnant snow drifts within the lake basin were also sampled during the warmer months.

Rain water was collected using a 28 cm diameter polyethylene funnel connected to a 20 L polyethylene jug with a Tygon hose. The funnel was set up in an open area so that rain water dripping off vegetation would not be

collected.

Santa Fe Lake water was also collected monthly during this same time period. However, the January sample could not be collected due to avalanche danger. Lake water was collected in nitric acid/distilled water rinsed polyethylene bottles.

After collection, lake water was filtered through 0.45  $\mu$ m micromembrane filters and the pH and alkalinity were measured in the field. pH was measured with a portable pH meter calibrated with buffers of pH 4 and 7. Alkalinity was determined by the Gran method (Stumm and Morgan, 1981) with the aid of a microliter buret (Gilmont Instruments). The water was titrated with dilute sulfuric acid standardized by titration with sodium hydroxide which in turn had been standardized against a carefully prepared potassium hydrogen phthalate solution. EPA alkalinity standards were titrated along with each lake sample to check for accuracy.

Precipitation samples were returned to the laboratory for filtering and pH determination. The study area has been found to receive acid precipitation (Popp et al., 1982; Popp et al. 1984). For this reason the accuracy of the pH determinations was checked against artificial rain samples made of dilute nitric acid solutions which had been previously titrated with standardized sodium hydroxide to determine pH. This was necessary because it was found that the glass electrodes

used to measure pH failed to provide accurate results in the low ionic strength solutions tested unless they were fairly new and operating at peak efficiency. The artificial acid rain of known pH was therefore used as a check to ensure the reliability of the electrode. The pH of the artificial acid rain ranged from pH 3 to 4.

#### Cation and Anion Analysis:

All water samples were divided into two acid-rinsed polyethylene bottles for cation and anion analysis. The water samples destined for cation analysis were acidified with a few drops of redistilled concentrated nitric acid to prevent any adsorption or precipitation of dissolved species.

Chloride, nitrate, and sulfate were analyzed with a Dionex 2000i/SP ion chromatograph using either a p/n 039590 Fast Sep Anion-1 exchange column or a p/n 037041 HPIC AS4A anion exchange column.

Calcium, magnesium, sodium, potassium, lithium, and iron were analyzed by atomic absorption (AA) spectrophotometry using an Instrumentation Laboratories, Inc. Model 857 AA or a Varian Techtron Model 1250 AA.

Silica was determined colorimetrically using the molybdosilicate method (American Public Health Association, 1976). This method involves acidifying the water sample with HCl to about pH 1.2. Ammonium molybdate is then added, reacting with the silica in

the water to form yellow molybdosilicic acid which has an absorption maximum at 410 nm. Oxalic acid was also added to avoid interferences from molybdophosphoric acid. The percent transmittance for standards and samples was determined using a Bausch and Lomb Spectronic 20 spectrophotometer.

Aluminum was determined colorimetrically using the eriochrome cyanine method (American Public Health Association, 1976). This method involves buffering each water sample to a pH of 6.0 with acetic acid and sodium acetate and then adding eriochrome cyanine dye to produce a pink aluminum complex. Ascorbic acid was added to eliminate interferences from manganese and iron. A lake water sample containing EDTA was used as a blank to correct for any errors caused by the normal color of the water. Percent transmittance was read at a wavelength of 535 nm on a Bausch and Lomb Spectronic 20 spectrophotometer and converted to absorbance.

#### Determination of Radionuclide Activities:

The activities of Pb-210, Pb-214, and Cs-137 were determined using an Ortec high purity N-type germanium detector. All of the samples were counted for approximately 24 hours.

The freeze-dried core slices were prepared for analysis by placing them in glass petri dishes sealed with tape to prevent escape of Rn-222 gas. The reasoning

behind this was that if Rn-222 escaped, Pb-210 would not attain true secular equilibrium. This is theoretically correct, however activities of Pb-210 and Pb-214 measured from samples counted in unsealed petri dishes did not differ significantly from activities measured after the same samples were sealed for 2 weeks and then recounted (table 2).

In order to be as consistent as possible, 5 grams of sample from each core slice was the usual amount added to each petri dish. However, some core slices did not contain 5 grams and therefore sediment from 2 slices was combined to obtain the necessary quantity. A few slices which weighed slightly less than 5 grams were counted "as is" without any additional material added from an adjacent slice. A sample such as T-8 which did not contain 5 grams and required some additional material from T-9 was relabelled T-8,9.

The efficiency of the detector over the energy range from 0 to 1500 KeV was determined from reference standards using a computer program prepared by Nuclear Data, Inc. (1980). The standard used in preparing the efficiency curve was 5 grams of medium quartz sand in a petri dish spiked with 464 pCi of Cs-137, 232 pCi of Co-60, and 200 pCi of Pb-210. The source of the Cs-137 and the Co-60 was Amersham Corporation (Amersham, England) standard QCY.44 and the source of the Pb-210 was Isotope Products Laboratories (Burbank, California) standard

Table 2. Comparison of nuclide activities in sediments recently sealed in petri dishes and nuclide activities in the same samples about 2 weeks later. Both samples were sealed on July 9. Sample T-8,9 was first counted on July 11 and then recounted on July 23. Sample T-9,10 was first counted on July 12 and then recounted on July 25. Nuclide activities are given in picocuries per gram.

<u>Sample</u>	Pb-210 activity		Pb-214 activity		unsupported Pb-210	
	<u>count 1</u>	<u>count 2</u>	<u>count 1</u>	<u>count 2</u>	<u>count 1</u>	<u>count 2</u>
T-8,9	6.499	7.495	4.043	4.604	2.456	2.891
T-9,10	6.516	6.469	4.456	4.368	2.060	2.101



218-1. The efficiency program calculates the efficiency of the detector at the energies of the gamma rays emitted by the nuclides in the standard. An efficiency curve is then calculated for the entire spectrum by extrapolating between the known points.

The gamma counts per second associated with the peak centroid energies corresponding to the nuclides of interest were converted to picocuries per gram of sample using equation 1.

$$\text{pCi/g} = \frac{\text{counts/second}}{(3.7 \times 10^{-12}) (\text{g sample}) (\text{abundance}) (\text{efficiency})}$$

Equation 1. Equation used to convert counts per second to picocuries per gram.

#### Sediment Composition:

#### Lithium Analysis:

Several slices from the Santa Fe Lake sediment core were digested and analyzed for lithium. The purpose of this was to see if inputs from geochemical weathering have changed over time. Such would be the case if acidification of precipitation has had a significant effect in accelerating weathering rates. Lithium was chosen because the amounts coming in from the atmosphere are insignificant. However, rocks from the lake basin that were digested and analyzed for lithium were found to

contain as much as 27 ppm (table 3). Therefore changes in lithium concentrations occurring in a sediment core would be a good indicator of changes in weathering rates through time.

Digestion Procedure:

One gram portions of freeze-dried core slices were placed in Teflon beakers. Approximately 3 mL of deionized water were added to each sample followed by 6 mL of distilled concentrated HF and 6 mL distilled concentrated aqua regia (3:1 HCl:HNO<sub>3</sub>). The beakers were heated on a hot plate until just before dryness was achieved. At this time they were removed from the hot plate and 5 mL of concentrated distilled HNO<sub>3</sub> and 5 mL of 30% H<sub>2</sub>O<sub>2</sub> were added. The samples were heated until just short of dryness. Complete dryness was avoided to inhibit loss of analyte due to volatilization. At this point the samples were transferred through Whatman #4 qualitative filters into 100 mL volumetric flasks, diluted up to volume with 10% HNO<sub>3</sub>, and stored in polyethylene bottles until they could be analyzed.

Rock samples were pulverized in a Bico pulverizer using ceramic pulverizing plates and then digested. The same procedure used to digest the core slices was also used to digest the rocks except that no H<sub>2</sub>O<sub>2</sub> was used. The procedure had to be repeated twice to dissolve all of the rock.

Table 3. Lithium concentrations in rocks from the Santa Fe Lake basin (ppm dry weight).

<u>Rock Type</u>	<u>Lithium (ppm)</u>
quartz diorite	14
pegmatite	14
biotite gneiss	27
migmatite	14
granite	6

Textural Analysis:

Particle size analysis was performed on several slices from each core. This was important because radionuclide activities in sediments have been shown to be partly dependent upon texture (Smith and Walton, 1980). Therefore if significant changes in sediment texture was found to exist within the cores, steps would be required to correct for this when calculating sediment ages.

Organic matter first had to be removed from the core slices. This was usually accomplished by removing the large pieces (sticks, pine needles, etc.) with a pair of tweezers. Smaller pieces were removed by wetting the sediment with water and skimming off the organic matter as it floated to the surface.

Several samples (Lag3-1, Sgl-8, and Sgl-2) contained so much organic matter that it was necessary to oxidize the smallest pieces with 30%  $H_2O_2$  using the method of Janitzky (1986). This method involved placing the samples in beakers and adding three, 4 to 5 mL aliquots of 30%  $H_2O_2$  at 5 minute intervals. The beakers were then covered with watchglasses and set aside for about 1/2 hour to let the initial foaming subside. The beakers were then heated for 1 hour at 70 C on a hot plate. While the sediment was heated, 4-5 mL aliquots of 30%  $H_2O_2$  were added every 10 minutes. The samples were then removed

from the heat to cool and the sediment was separated from the solution by alternately centrifuging, decanting the supernatant, rinsing with distilled water, and recentrifuging.

After removal of organic matter, all samples were dried and weighed. Sand sized and larger sediment was removed with a 230 mesh seive (110  $\mu\text{m}$ ) and weighed. The remaining silt and clay sized portion was then diluted to 1000 mL with distilled water and the silt particles ( $> 2 \mu\text{m}$ ) were allowed to settle for 1/2 hour. After this time, 20 mL of solution from the the very top of the beaker were pipetted off and put into a previously weighed evaporation dish for drying. The resulting clay fraction (which represents 20/1000 or 1/50 of the total clay in the sample) was then weighed and the figure multiplied by 50 to determine the total clay fraction in the sample. The results of the particle size analysis are listed in Appendix B.

#### Clay Mineral Analysis

In order to thoroughly characterize the sediment samples, x-ray diffraction analysis of the less than 2 micron fraction of the lake sediments was also performed. A semi-quantitative procedure developed by Austin (pers. comm.) was used. This involved mixing some of the silt and clay sized portion of the sediment left over from the textural analyses in distilled water to form a slurry.

The sediment was then allowed to settle for 1/2 hour. After this time some of the clay sized fraction remaining at the surface was removed with a pipette and placed on a petrographic slide and allowed to dry. This resulted in an oriented mount. Each sample was then run in a Rigaku Dmax IIA x-ray diffractometer using Cu K $\alpha$  radiation and a scan speed of 2° 2 $\theta$  per minute 3 ways: 1. From 2° to 35° 2 $\theta$  with no treatment; 2. From 2° to 15° 2 $\theta$  after 24 hours in an ethylene glycol atmosphere; 3. From 2° to 15° 2 $\theta$  after heating the slide at 375°C for 1/2 hour. If both chlorite and kaolinite were found to be present, a slow, no treatment run from 24° to 26° 2 $\theta$  at 0.5° 2 $\theta$  per minute was also necessary.

The purpose of these various treatments was to differentiate between mixed layer clay minerals and clay minerals with expanding and non-expanding lattices. The proportions of the various clay minerals present were then calculated from the relative peak intensities using the following empirically derived equations:

$$\text{Illite} = \frac{I(1G)}{T} \times 10$$

$$\text{Montmorillonite} = \frac{\frac{M(1)}{4}}{T} \times 10$$

$$\text{Chlorite} = \frac{C(3)}{I(2)} \times \frac{I(1G)}{T} \times 10$$

$$\text{Mixed Layer Illite/Smectite} = I(1H) - \frac{\left[ \frac{I(1G)}{4} + \frac{M(1)}{4} \right]}{T} \times 10$$

$$\text{Kaolinite} = \frac{K(1)}{T} \times 10$$

or if chlorite is present

$$\text{Kaolinite} = \frac{K(2)}{2C(4)} \times \frac{C(3)}{I(2)} \times \frac{I(1G)}{T} \times 10$$

where T = total counts:

$$T = I(1H) = K(1)$$

or, if chlorite is present:

$$T = I(1H) + \frac{C(3) \times I(1G)}{I(2)} + \frac{K(2) \times C(3) \times I(1G)}{2C(4) \times I(2)}$$

I(1H) = intensity of peak at 8.8° 2θ on the heated run

I(1G) = intensity of peak at 8.8° 2θ on the glycolated run

M(1) = intensity of peak at 5.2° 2θ on the glycolated run

K(1) = intensity of peak at 12.4° 2θ (first order kaolinite peak)

K(2) = intensity of peak at 25.1° 2θ (second order kaolinite peak) on the slow no treatment run

I(2) = intensity of peak at 17.8° 2θ (second order illite peak) on the initial no treatment run

C(3) = intensity of peak at 18.4° to 18.9° 2θ (third order chlorite peak) on the initial no treatment run

C(4) = intensity of peak at 25.1° 2θ (fourth order chlorite peak) on the slow no treatment run

where intensity is taken to be the peak height above background. The results of the clay mineral analyses are shown in Appendix C.

## Results and Discussion

### Atmospheric Contributions:

Appendices D and E show the chemical compositions of the monthly lake water and precipitation samples. Rain and snow samples were not analyzed for aluminum or iron since these elements would very likely occur at levels below the detection limits of the methods used. Only one silica determination was performed on a precipitation sample. The concentration of silica was very low compared to that found in the lake water. The silica concentration was assumed to be low in all samples and approximately equal to that in the single snow sample analyzed (snow sample collected 7/1/87).

Bicarbonate concentration was calculated assuming that precipitation was in equilibrium with CO<sub>2</sub> in the atmosphere. Other assumptions necessary for this calculation are that the atmospheric pressure at 3530 m equals 0.6492 atm (CRC Handbook, 1983), CO<sub>2</sub> content in the atmosphere equals 0.0314%, the vapor pressure of water at 5°C equals 0.0086 atm (Manahan, 1984), and the Henry's Law constant for CO<sub>2</sub> is 6.324 x 10<sup>-2</sup> mol/L·atm at 5°C (Cole, 1983). On the basis of these assumptions:

$$[\text{HCO}_3^-] = \frac{[4.45 \times 10^{-7}][6.56 \times 10^{-6} \text{ M}]}{[\text{H}^+]}$$

It should be noted that for the precipitation analyses the anion-cation balances are off considerably. This is believed to arise from not analyzing for all of the possible ionic species present. Previous work in



this region produced acceptable cation-anion balances when all significant species were accounted for (Popp et al., 1982).

Concentrations of solutes in lake water above that found in precipitation were assumed to originate from geochemical weathering. This assumption actually underestimates the importance of atmospheric inputs because atmospherically derived solutes can enter the lake basin via dry deposition as well as wet deposition. No studies have been done in this area to determine the importance of dry deposition relative to wet deposition.

Barrie et al. (1984) found that the importance of dry deposition of sulfur compounds approached that of wet deposition at sampling sites near sources of anthropogenic inputs in eastern Canada but became less important at sampling sites farther away from these sources. Barrie and Sirois (1986) determined that dry deposition accounted for about 22% of the total annual atmospheric  $\text{SO}_4^{--}$  inputs and about 30% of the total annual atmospheric  $\text{NO}_3^-$  inputs at six rural sampling sites in eastern Canada.

Dry deposition almost certainly accounts for a significant portion of the atmospheric inputs into Santa Fe Lake. However the precipitation samples collected at Santa Fe Lake were either collected as snow which had lain on the ground for long periods of time or as rain

collected from an open bucket set out for about 1 month. Because of the time lapse between precipitation events and sample collection these samples actually represent accumulations of both wet and dry deposition.

Evaporation of lake water also presents a problem because its effect would be to increase the concentration of lake solutes and therefore make weathering inputs appear larger than their true value. The significance of evaporation effects is very difficult to estimate because water is constantly flowing both into and out of the lake. However, the assumption that evaporation effects are very small is probably valid because evaporation is kept to a minimum by the cold climate and the ice which covers the lake about 7 months of the year. Also since the lake is small, the residence time of water in the lake must be low thereby allowing little time for evaporation to take place.

In order to determine the relative importance of atmospheric inputs versus inputs from weathering, species concentrations were averaged over all samples collected (table 4). In computing these averages some analyses had to be discarded. Nitrate was particularly troublesome because unless analyzed soon after collection it was found that concentrations rapidly dropped, presumably due to reduction by microorganisms. Samples not included in the calculation of average  $\text{NO}_3^-$

Table 4. Chemical species concentrations for Santa Fe Lake water and precipitation collected at Santa Fe Lake averaged from data obtained from monthly sampling. The average concentrations were used to estimate the relative amounts of atmospherically derived and weathering derived dissolved solids.

	Ca <sup>++</sup> (ppm)	Mg <sup>++</sup> (ppm)	Na <sup>+</sup> (ppm)	K <sup>+</sup> (ppm)	SiO <sub>2</sub> (ppm)	Cl <sup>-</sup> (ppm)	NO <sub>3</sub> <sup>-</sup> (ppm)	SO <sub>4</sub> <sup>--</sup> (ppm)	HCO <sub>3</sub> <sup>-</sup> (ppm)
average concentration in lake water	2.36	0.66	1.53	0.40	1.99	0.39	0.85	2.11	9.57
(std. dev. in parentheses)	(0.41)	(0.15)	(0.50)	(0.14)	(0.22)	(0.22)	(0.55)	(0.52)	(1.82)
average concentration in precipitation	0.91	0.27	0.64	0.49	0.18	0.47	0.79	1.09	0.27
(std. dev. in parentheses)	(0.66)	(0.56)	(0.73)	(0.71)	(0.00)	(0.32)	(0.54)	(0.95)	(0.62)
concentration attributable to weathering (= difference (molarity x E5 in parentheses)	1.45	0.39	0.89	-0.09	1.81	-0.08	0.06	1.02	9.30
	(3.62)	(1.60)	(3.87)	(-0.23)	(3.01)				(15.24)
percent of total concentration in lake water which is atmospherically derived	38%	41%	42%	122%	9%	120%	93%	52%	3%

concentrations because of the time delay involved between collection and analysis which resulted in significant biodegradation were the snow sample collected on 10/14/87 and the lake water samples collected on 11/12/87, 7/1/87, 6/14/87, 10/16/87, and 8/11/87.

The  $\text{SiO}_2$  analysis for the lake water sample collected on 5/12/88 was also discarded because insufficient sample was available for analysis. The measuring out of reagents required for this analysis was not highly accurate because of the irregular volumes required to complete the analysis for the small amount of sample on hand. Apparently the errors involved in measuring out volumes of reagents were significant because this result is markedly different from the other samples analyzed.

Table 4 shows that atmospheric contributions to the chemical constituents found in Santa Fe Lake vary considerably depending on the species. Nevertheless atmospheric inputs are quite large in comparison to weathering inputs for all species except  $\text{HCO}_3$  and  $\text{SiO}_2$ .

This data shows that approximately half of the sulfate found in the lake water originates from the atmosphere. This conflicts with the sulfur isotope data of Lynch et al. (1988) which suggests that essentially all of the sulfate in Santa Fe Lake is atmospherically derived. One problem with the weathering scenario is

that there is no readily apparent source of sulfate in the basin. If sulfide minerals (the most likely source of sulfate in the area) are present in the lake basin, they either occur as microscopic crystals, are not readily visible on surface outcrops, or both.

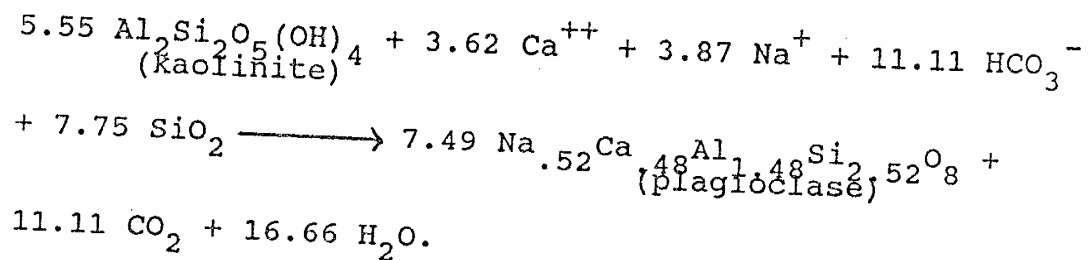
Evidence for the reliability of this data is shown by chloride. There are no chloride minerals present in the basin. Some chloride probably exists as an impurity in non-chloride minerals, but this amount would be insignificant compared to the amount coming in from the atmosphere. Therefore the chloride concentrations in both precipitation and lake water should be equal. Table 4 shows that this is very nearly so, the difference between the two being the amount of experimental error.

Nitrate is another species that is not derived from weathering. Once again table 4 shows that the concentration of  $\text{NO}_3^-$  is very nearly equal in both precipitation and lake water. However, unlike chloride,  $\text{NO}_3^-$  can also be derived from the oxidation of  $\text{NH}_4^+$  produced by nitrogen fixing bacteria and it is also rapidly cycled by many organisms. The significance of nitrogen fixing bacteria in contributing  $\text{NO}_3^-$  to Santa Fe Lake and the effects of nitrogen cycling on the concentration of  $\text{NO}_3^-$  in the lake water is unknown. Therefore the significance of  $\text{NO}_3^-$  as an indicator of the reliability of the data presented is not clear.

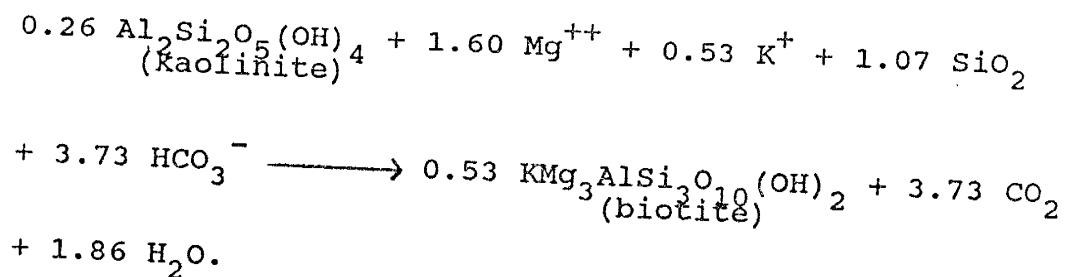
Mass Balance Determination of Weathering Reactions for Santa Fe Lake:

XRD analysis of the lake sediment showed that the clay sized fraction was mostly amorphous. Therefore there are no clearly defined alteration products. The XRD analysis did reveal a trace amount of kaolinite. Apparently some kaolinite formation is occurring.

On the basis of this, an attempt was made to back react kaolinite with the chemical species in the lake water which were produced by geochemical weathering to produce minerals present in the basin. Sodium and  $\text{Ca}^{++}$  are the two most abundant cations which originate from weathering. Since no limestone exists in the basin the only reasonable source for  $\text{Ca}^{++}$  and  $\text{Na}^+$  is plagioclase. Back reacting all of the  $\text{Ca}^{++}$  and  $\text{Na}^+$  with kaolinite gives:



Biotite is the only reasonable source of  $\text{Mg}^{++}$  so assuming an ideal biotite composition, all of the  $\text{Mg}^{++}$  is back reacted with kaolinite to make biotite:



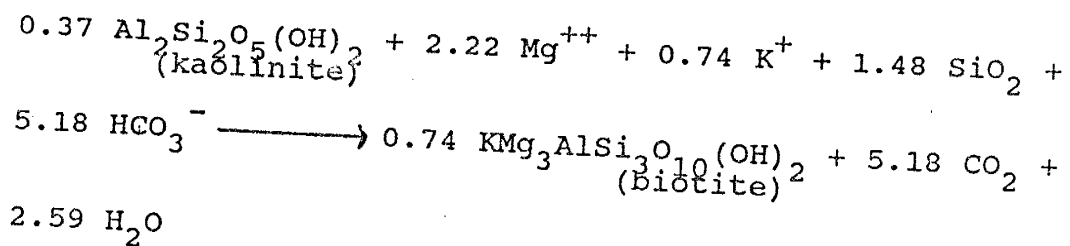
This creates some problems because although the balance works out correctly for  $\text{HCO}_3^-$ , these reactions require more  $\text{K}^+$  and  $\text{SiO}_2$  than is available. Appendix E shows that the rain sample collected from 9/3 to 10/13/87 has an anomalously high value for  $\text{Mg}^{++}$ . It also shows that the  $\text{K}^+$  concentration in the snow sample collected 5/25/86 is also abnormally high. Assuming that these values are erroneous, average  $\text{K}^+$  and  $\text{Mg}^{++}$  levels in precipitation were recalculated without including these values. The new  $\text{K}^+$  and  $\text{Mg}^{++}$  averages were subtracted from the average concentrations found in lake water and the differences, which are the amount contributed from weathering, were converted to molarity  $\times 10^5$  (table 5).

Using these revised values for  $\text{Mg}^{++}$  and  $\text{K}^+$ , kaolinite was again back reacted with the dissolved chemical components originating from weathering to form primary minerals. Since only  $\text{Mg}^{++}$  and  $\text{K}^+$  values have been changed the reaction involving plagioclase is the same, only biotite is affected.

Table 5. Average  $Mg^{++}$  and  $K^+$  concentrations in both Santa Fe Lake and precipitation collected at Santa Fe Lake as well as contributions from geochemical weathering and the atmosphere. The  $K^+$  value from the snow sample collected 5/25/86 and the  $Mg^{++}$  value from the rain collected from 9/3 to 10/13/87 are not averaged into these figures because of their anomalously high values.

	$Mg^{++}$ <u>(ppm)</u>	$K^+$ <u>(ppm)</u>
average concentration in lake water (std. dev. in parentheses)	0.66 (0.15)	0.40 (0.14)
average concentration in precipitation (std. dev. in parentheses)	0.12 (0.09)	0.31 (0.28)
concentration attributable to weathering (= difference) (molarity $\times 10^5$ in parentheses)	0.54 (2.22)	0.09 (0.23)
percent of total which is atmospherically derived	18%	78%





The mass balance calculated which includes the anomalous  $\text{Mg}^{++}$  and  $\text{K}^+$  values balances well for all species except  $\text{SiO}_2$  and  $\text{K}^+$ . All of the  $\text{Ca}^{++}$ ,  $\text{Mg}^{++}$ , and  $\text{Na}^+$  are accounted for and only about 7% of the  $\text{HCO}_3^-$  is unaccounted for (well within experimental error). The revised mass balance which ignores the anomalous  $\text{Mg}^{++}$  and  $\text{K}^+$  values works out slightly better with the shortage of  $\text{K}^+$  and  $\text{SiO}_2$  needed to obtain a satisfactory balance only slightly less. In either case however, the results are essentially the same with some  $\text{K}^+$  and  $\text{SiO}_2$  apparently lacking. These deficits can be explained by non-uniform dissolution of mineral grains and contribution of  $\text{K}^+$  to precipitation by plants.

Some of the snow samples were collected beneath trees. Precipitation falling through a forest canopy picks up solutes from the plants and potassium is enriched significantly. Likens et al. (1977) found that precipitation was enriched as much as 91 times in potassium after falling through a northern hardwood forest canopy in New Hampshire. Freiesleben et al. (1986) found a 34% increase in potassium content of precipitation after falling through the canopy of a spruce forest in Denmark. The enrichment of potassium

in both studies far exceeded the enrichment of any of the other solutes measured. Although rain samples were collected in an open area away from dripping vegetation, most of the precipitation samples collected were snow. Usually the snow had lain on the ground for at least several days and in some cases several weeks and was noted to contain significant amounts of plant debris. It therefore seems reasonable to assume that the elevated  $K^+$  levels in precipitation were due to leaching from plant materials.

Experiments by Luce et al. (1972) suggest that weathering of freshly exposed Mg-silicate minerals can result in silica rich alteration rinds on exposed surfaces. Evidence for similar effects in feldspars has been provided by Paces (1973) and Busenburg and Clemency (1976). If the outer layers of minerals undergoing weathering are silica enriched then it follows that the solutions participating in the weathering would be silica depleted.

There was some question as to how realistic it is to use kaolinite as the alteration product since only trace amounts showed up in x-ray diffractograms. In order to help answer this question, chemical data from the 12 Santa Fe Lake water samples shown in Appendix D were analyzed using the chemical equilibrium computer program WATEQ (Truesdell and Jones, 1974). This program converts analytical concentrations to activities of

aqueous species and determines the thermodynamic stability of various mineral phases with respect to the solution.

Kaolinite was found to be supersaturated with respect to all 12 of the water samples examined. Furthermore, all of the primary minerals occurring in Santa Fe Lake basin that are included in the WATEQ program were found to be undersaturated (table 6).

Activities of chemical species calculated by the WATEQ program were plotted on mineral stability diagrams (figures 7,8, and 9). All samples were found to plot within the kaolinite stability fields.

All of the data examined suggests that kaolinite is the mineral alteration product being produced in the Santa Fe Lake basin yet only trace quantities were detected by x-ray diffraction analysis. This can be explained by the fact that even if a mineral phase is thermodynamically stable it may not be as kinetically favored as some other phase (Drever, 1982). This seems to fit in well with the results because amorphous phases are always thermodynamically less stable than their corresponding crystalline phases but amorphous phases also tend to be kinetically more favorable. What seems to be happening in Santa Fe Lake is the production of a kaolinitic amorphous phase (i.e. an amorphous phase of kaolinitic composition) such as allophane.

Table 6. Log IAP/KT values for Santa Fe Lake water samples computed by the WATEQ program for mineral phases found in Santa Fe Lake basin. Positive values indicate supersaturation with respect to the lake water whereas negative values indicate undersaturation. Since only the sodium and calcium plagioclase endmembers are considered by WATEQ both of these minerals are included. Similarly phlogopite (the magnesium biotite endmember) is listed whereas actual biotite found in the lake basin almost certainly contains some iron.

<u>Sample Date</u>	<u>Albite</u>	<u>Anorthite</u>	<u>Quartz</u>	<u>Phlogopite</u>	<u>Kaolinite</u>
7/1/87	-5.017	-5.573	-0.247	-19.943	3.339
12/16/87	-3.625	-4.514	-0.022	-21.757	5.371
5/27/88	-6.902	-9.510	-0.233	-29.365	1.967
11/12/87	-4.246	-5.383	-0.111	-21.505	4.205
5/12/88	-7.761	-11.217	-0.268	-34.080	1.254
4/17/88	-7.521	-11.898	-0.056	-33.441	0.697
2/11/88	-4.802	-6.307	-0.096	-23.926	3.873
3/17/88	-6.433	-9.638	-0.094	-30.175	2.217
9/3/87	-6.116	-8.866	-0.181	-27.786	2.340
6/14/88	-6.271	-8.277	-0.294	-26.667	2.452
10/16/87	-6.905	-9.319	-0.297	-27.510	1.573
8/11/87	-5.529	-6.672	-0.302	-21.464	2.717

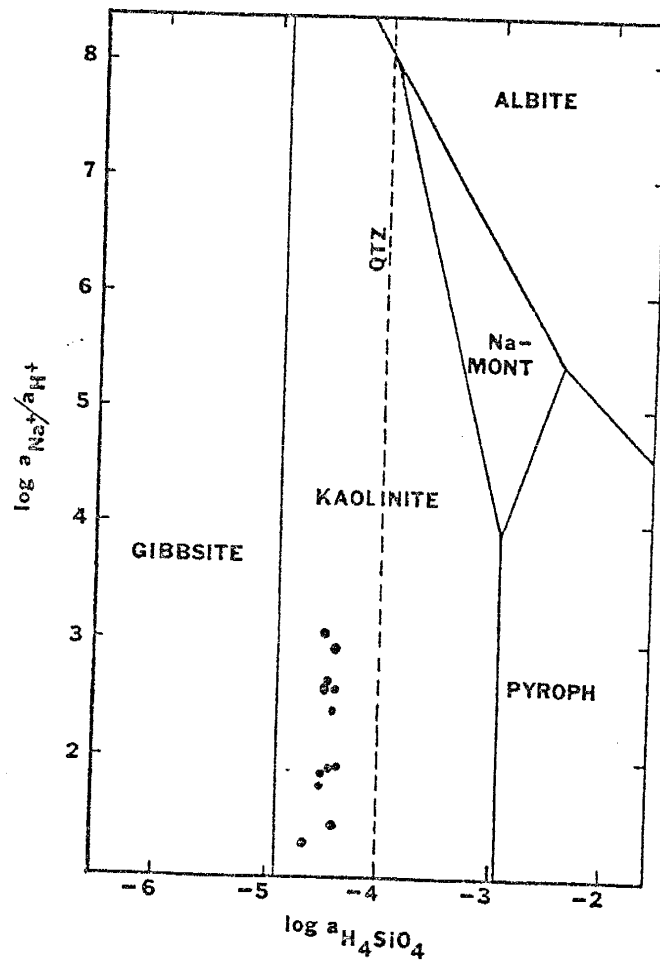


Figure 7. Water compositions from Santa Fe Lake plotted on a  $\log a_{\text{Na}^+}/a_{\text{H}^+}$  versus  $\log a_{\text{H}_4\text{SiO}_4}$  stability diagram (Riese and Popp, 1980).

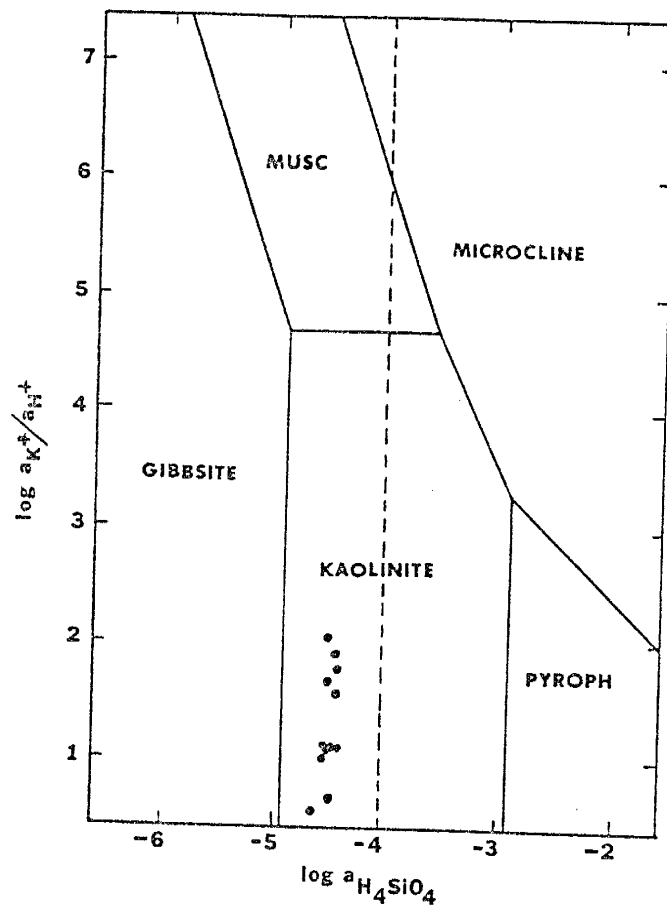


Figure 8. Water compositions from Santa Fe Lake plotted on a  $\log a_{K^+}/a_{H^+}$  versus  $\log a_{H_4SiO_4}$  stability diagram (Riese and Popp, 1980).

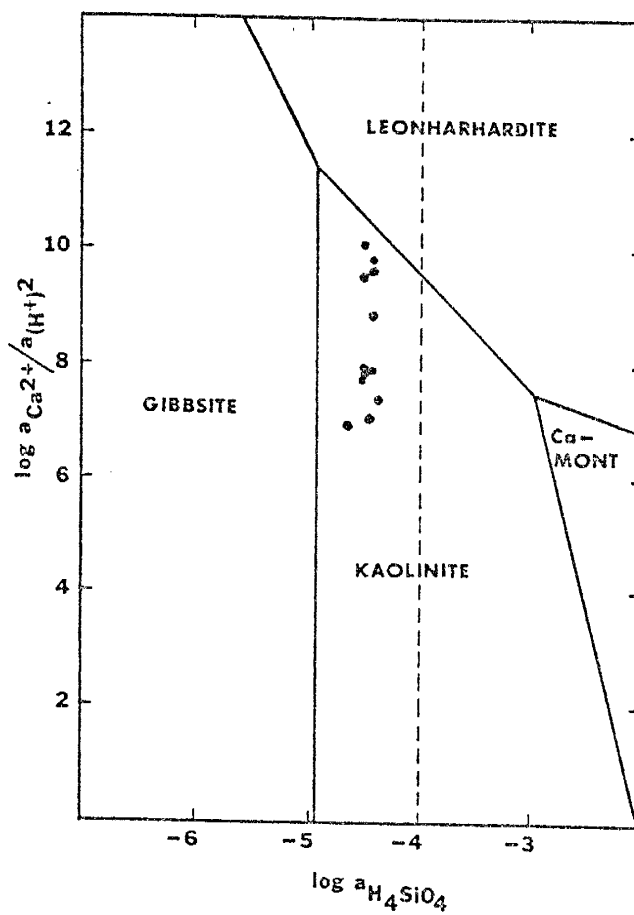


Figure 9. Water compositions from Santa Fe Lake plotted on a  $\log a_{Ca^{2+}}/a_{(H^+)^2}$  versus  $\log a_{H_4SiO_4}$  stability diagram (Riese and Popp, 1980).

### Lithium Distribution in Santa Fe Lake Sediment

A plot of lithium concentration in Santa Fe Lake sediment versus depth is shown in figure 10. A linear regression analysis of the data produces a line with a slightly positive slope (0.176). However, the correlation coefficient for this line is only 0.34. Given the low correlation coefficient, and the small number of data points, the slope of the line does not vary significantly enough from zero to arrive at any conclusions about trends in weathering rates through time.

### Radionuclide Activities in Lake Sediments

#### Pb-210 Dating

Tables 7-11 show radionuclide activities in sediment from the 5 lakes studied. The activity of unsupported Pb-210 was determined by subtracting Pb-214 activity from total Pb-210 activity.

Since each core slice is thick enough to contain several years worth of deposition, Pb-210 activity measured in a core slice is actually an average of the Pb-210 activity for the whole slice. For this reason the Pb-210 activity measured for a core slice was assumed to represent the activity in the middle of the slice. For example, the Pb-210 activity measured in the top 1.0 cm slice of a sediment core was assumed to be representative of the activity exactly 0.5 cm from the

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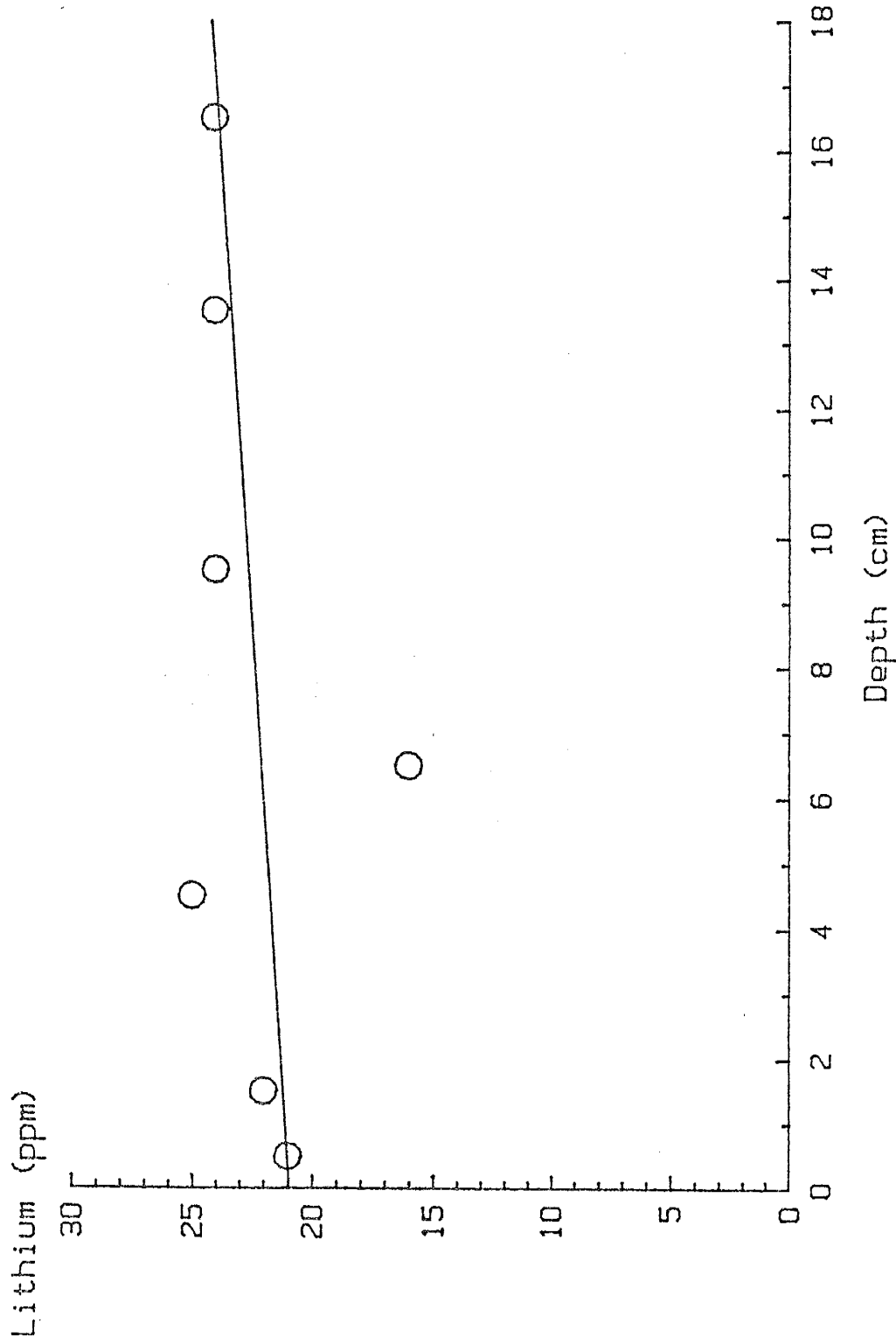


Figure 10. Lithium concentration plotted versus depth for Santa Fe Lake sediments. Linear regression line has a slope of 0.176 and a correlation coefficient of  $r = 0.34$ .

Table 7. Radionuclide activities in the sediment core from Lost Lake.

<u>Sample</u>	Total Pb-210 (pCi/g)	Pb-214 (pCi/g)	Unsupported Pb-210 (pCi/g)	Cs-137 (pCi/g)
L-1	13.988	3.984	10.004	5.143
L-2	12.939	3.630	9.309	6.606
L-4	7.285	3.600	3.685	3.194
L-5	6.906	4.337	1.759	1.354
L-6,7	4.698	4.131	0.567	1.068
L-8	4.911	4.405	0.506	0.976

Table 8. Radionuclide activities in the sediment core from Sugarloaf Lake.

<u>Sample</u>	Total Pb-210 (pCi/g)	Pb-214 (pCi/g)	Unsupported Pb-210 (pCi/g)	Cs-137 (pCi/g)
SGL-1,2	3.227	1.570	1.657	0.833
SGL-3	2.317	1.229	1.088	0.401
SGL-4	2.617	1.564	1.053	0.410
SGL-5	1.707	1.574	0.133	0.309
SGL-6	2.112	1.584	0.528	0.290

M.M.T.

Table 9. Radionuclide activities in the sediment core from Lagunitas #3 Lake.

<u>Sample</u>	Total Pb-210 <u>(pCi/g)</u>	Pb-214 <u>(pCi/g)</u>	Unsupported Pb-210 <u>(pCi/g)</u>	Cs-137 <u>(pCi/g)</u>
Lag3-1	6.078	3.069	3.009	0.878
Lag3-8	5.521	3.175	2.346	4.339
Lag3-18	2.294	3.039	-0.745	0.662
Lag3-24	3.805	3.275	0.530	0.602
Lag3-30	4.376	4.253	0.123	1.158

Table 10. Radionuclide activities in the sediment core from Santa Fe Lake.

<u>Sample</u>	Total Pb-210 (pCi/g)	Pb-214 (pCi/g)	Unsupported Pb-210 (pCi/g)	Cs-137 (pCi/g)
SF-1,2	27.349	3.145	24.204	5.919
SF-3,5	13.277	3.276	10.001	2.074
SF-6,7	6.650	2.833	3.817	1.204
SF-8,9	4.522	3.188	1.334	1.000
SF-10,11,12	2.746	2.700	0.046	0.771

Table 11. Radionuclide activities in the sediment core from Truchas Lake.

<u>Sample</u>	Total Pb-210 (pCi/g)	Pb-214 (pCi/g)	Unsupported Pb-210 (pCi/g)	Cs-137 (pCi/g)
T-1	30.883	6.883	24.000	9.642
T-2	25.994	4.043	21.951	10.285
T-3,4	20.847	4.191	16.656	5.768
T-4,5	10.202	4.368	5.834	3.127
T-5,7	7.588	4.073	3.515	2.006
T-8,9	6.997	4.323	2.674	1.164
T-9,10	6.492	4.412	2.080	1.094
T-11	5.530	3.925	1.605	1.198
T-13	5.234	4.161	1.073	0.751
T-15	6.556	5.614	0.942	1.114

R.M.M.T.

top.

The difference in age between slices in a sediment core was determined using equation 2. By measuring the distance between the mid-slice depths of slices for which unsupported Pb-210 was used to calculate age and dividing this distance by the age difference calculated using equation 2, a sedimentation rate for each core was determined. By using these sedimentation rates, the absolute ages of each slice in the five sediment cores were calculated (tables 12-21). Since the half-life of Pb-210 is 22.3 years, dating using this method can only be used for sediments less than five half-lives old (about 100 years). However, if sedimentation rate is assumed to not have changed significantly in sediments older than 100 years, then the sedimentation rate calculated for the top 100 years of sediment can be used to estimate the age of older sediments.

$$t = \frac{-\ln N/N_0}{\lambda}$$

Equation 2. The equation used to determine the difference in age between 2 successive core slices based on unsupported Pb-210 activity.  $N_0$  = unsupported Pb-210 activity in a core slice,  $N$  = unsupported Pb-210 activity in the next slice down in a sediment core,  $\lambda$  is the decay constant which is equal to  $3.108 \times 10^{-2}$  1/yrs, and  $t$  is the difference in age between the two slices.

Table 12. The sedimentation rate for Lost Lake determined using Pb-210.

<u>Sample</u>	<u>Mid-slice Depth (cm)</u>	<u>Age Difference (years)</u>	<u>Sedimentation Rate (cm/yr)</u>
L-1	0.5	} 96	0.07
L-8	7.5		

MMMT



Table 13. The mid-slice age of core slices from Lost Lake determined from sedimentation rate.

<u>Slice</u>	<u>Mid-slice Depth (cm)</u>	<u>Mid-slice Age (years)</u>
L-1	0.5	7
L-2	1.5	21
L-3	2.5	36
L-4	3.5	50
L-5	4.5	64
L-6	5.5	78
L-7	6.5	93
L-8	7.5	107
L-9	8.5	121
L-10	9.5	136
L-11	10.5	150
L-12	11.5	164
L-13	12.5	178
L-14	13.5	193
L-15	14.5	207

Table 14. The sedimentation rate for Sugarloaf Lake determined using Pb-210.

<u>Sample</u>	<u>Mid-sample Depth (cm)</u>	<u>Age Difference (years)</u>	<u>Sedimentation Rate (cm/yr)</u>
SGL-1,2	1.0	) 81	0.04
SGL-5	4.5		

Table 15. The mid-slice age of core slices from Sugarloaf Lake determined from sedimentation rate.

<u>slice</u>	<u>Mid-slice Depth (cm)</u>	<u>Mid-slice Age (years)</u>
SGL-1	0.5	12
SGL-2	1.5	38
SGL-3	2.5	62
SGL-4	3.5	88
SGL-5	4.5	112
SGL-6	5.5	138
SGL-7	6.5	162
SGL-8	7.5	188
SGL-9	8.5	212
SGL-10	9.5	238
SGL-11	10.5	262

2000

Table 16. The sedimentation rate for Lagunitas #3 Lake determined using Pb-210.

<u>Sample</u>	<u>Mid-sample Depth (cm)</u>	<u>Age Difference (years)</u>	<u>Sedimentation Rate (cm/yr)</u>
Lag3-1	0.5	} 8.01	0.87
Lag3-8	7.5		

Table 17. The mid-slice age of core slices from Lagunitas #3 Lake determined from sedimentation rate.

<u>Slice</u>	<u>Mid-slice Depth (cm)</u>	<u>Mid-slice Age (years)</u>
Lag3-1	0.5	0.6
Lag3-2	1.5	1.7
Lag3-3	2.5	2.9
Lag3-4	3.5	4.0
Lag3-5	4.5	5.2
Lag3-6	5.5	6.3
Lag3-7	6.5	7.5
Lag3-8	7.5	8.6
Lag3-9	8.5	10
Lag3-10	9.5	11
Lag3-11	10.5	12
Lag3-12	11.5	13
Lag3-13	12.5	14
Lag3-14	13.5	16
Lag3-15	14.5	17
Lag3-16	15.5	18
Lag3-17	16.5	19
Lag3-18	17.5	20
Lag3-19	18.5	21
Lag3-20	19.5	22
Lag3-21	20.5	24
Lag3-22	21.5	25
Lag3-23	22.5	26
Lag3-24	23.5	27
Lag3-25	24.5	28
Lag3-26	25.5	29
Lag3-27	26.5	30
Lag3-28	27.5	32
Lag3-29	28.5	33
Lag3-30	29.5	34
Lag3-31	30.5	35
Lag3-32	31.5	36

Table 18. The sedimentation rate for Santa Fe Lake determined using Pb-210.

<u>Sample</u>	<u>Mid-sample Depth (cm)</u>	<u>Age Difference (years)</u>	<u>Sedimentation Rate (cm/yr)</u>
SF-1,2	1.0	} 93	0.08
SF-8,9	8.0		

Table 19. The mid-slice age of core slices from Santa Fe Lake determined from sedimentation rates.

<u>Slice</u>	<u>Mid-slice Depth (cm)</u>	<u>Mid-slice Age (years)</u>
SF-1	0.5	6
SF-2	1.5	19
SF-3	2.5	31
SF-4	3.5	44
SF-5	4.5	56
SF-6	5.5	69
SF-7	6.5	81
SF-8	7.5	94
SF-9	8.5	106
SF-10	9.5	119
SF-11	10.5	131
SF-12	11.5	144
SF-13	12.5	156
SF-14	13.5	169
SF-15	14.5	181
SF-16	15.5	194
SF-17	16.5	206
SF-18	17.5	219

Table 20. The sedimentation rate for Truchas Lake determined using Pb-210.

<u>Sample</u>	<u>Mid-sample Depth (cm)</u>	<u>Age Difference (years)</u>	<u>Sedimentation Rate (cm/yr)</u>
T-1	0.5	} 104	0.13
T-15	14.5		



Table 21. The mid-slice age of core slices from Truchas Lake determined from sedimentation rates.

<u>Slice</u>	<u>Mid-slice Depth (cm)</u>	<u>Mid-slice Age (years)</u>
T-1	0.5	4
T-2	1.5	12
T-3	2.5	19
T-4	3.5	27
T-5	4.5	35
T-6	5.5	42
T-7	6.5	50
T-8	7.5	58
T-9	8.5	65
T-10	9.5	73
T-11	10.5	81
T-12	11.5	88
T-13	12.5	96
T-14	13.5	104
T-15	14.5	112
T-16	15.5	119
T-17	16.5	127
T-18	17.5	135
T-19	18.5	142
T-20	19.5	150
T-21	20.5	158

### Cs-137 Dating

Cs-137 is an artificial nuclide that did not exist in the environment prior to above ground nuclear weapons testing in 1945 (Krishnaswami and Lal, 1979). However, Cs-137 was detected throughout all of the cores analyzed in sediments found to be decades older than 1945 by Pb-210 dating. Also, maximum Cs-137 activities existed at or near the top of all of the cores (figures 11-15). Clearly the Cs-137 activity profiles for the sediment cores are not representative of the fallout history. This combined with the evidence indicating that Cs-137 has been mobilized renders Cs-137 useless for dating. Davis et. al (1984) found Cs-137 behaved similarly in soft water lakes in Scandinavia and New England. They attributed the presence of Cs-137 at pre-fallout depths to molecular diffusion. The surface Cs-137 maxima were attributed to net upward diffusion and adsorption, recycling of sedimentary Cs-137 into the water column, and delayed input into the watershed.

### Reliability of Pb-210 Dates

Delayed entry of Pb-210 into a lake would not affect the reliability of Pb-210 dates as long as the delay was steady state (Davis et al., 1984). This is because once incorporated into lake sediments unsupported Pb-210 would still decay according to equation 2.

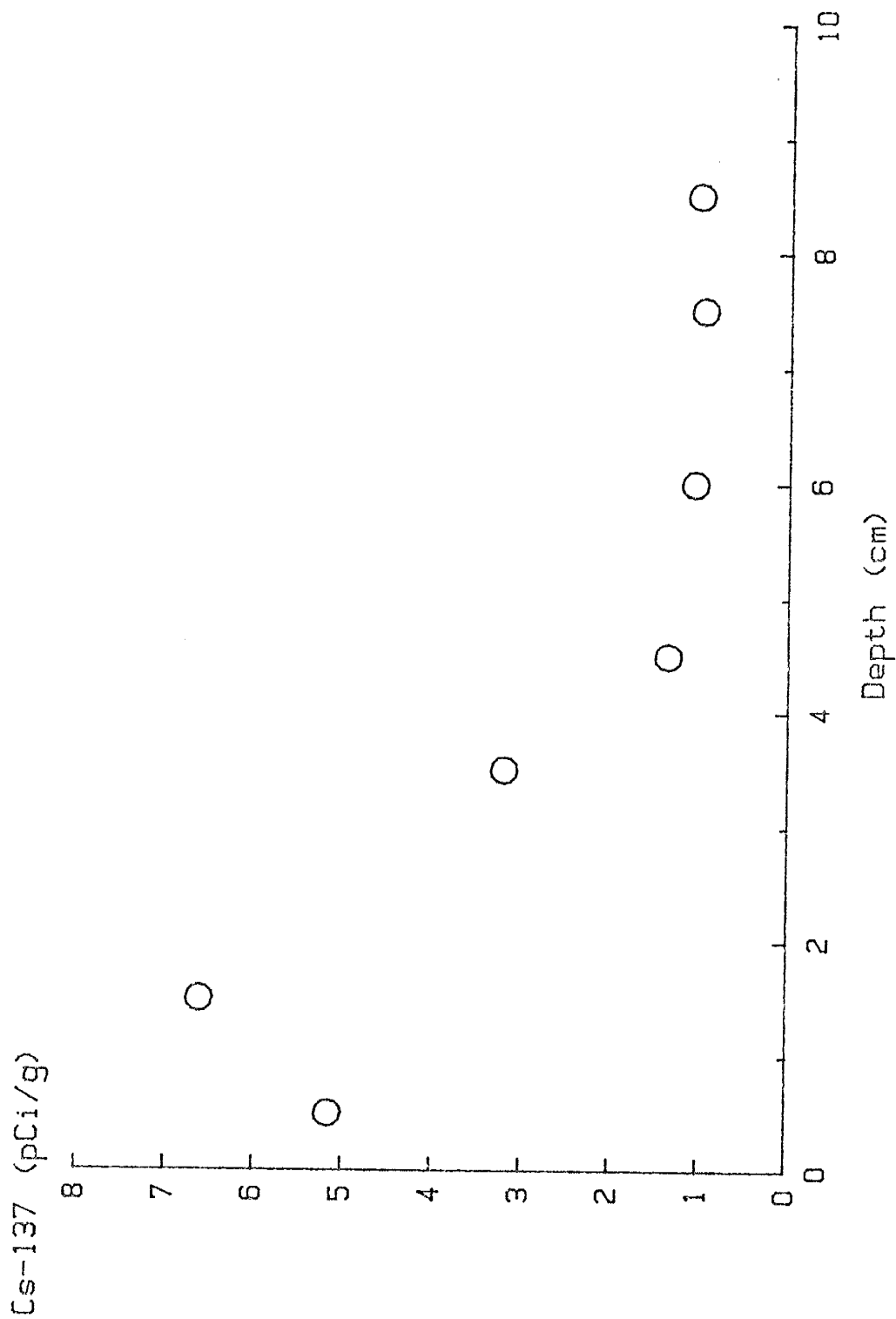


Figure 11. Cs-137 activity profile for the sediment core from Lost Lake.

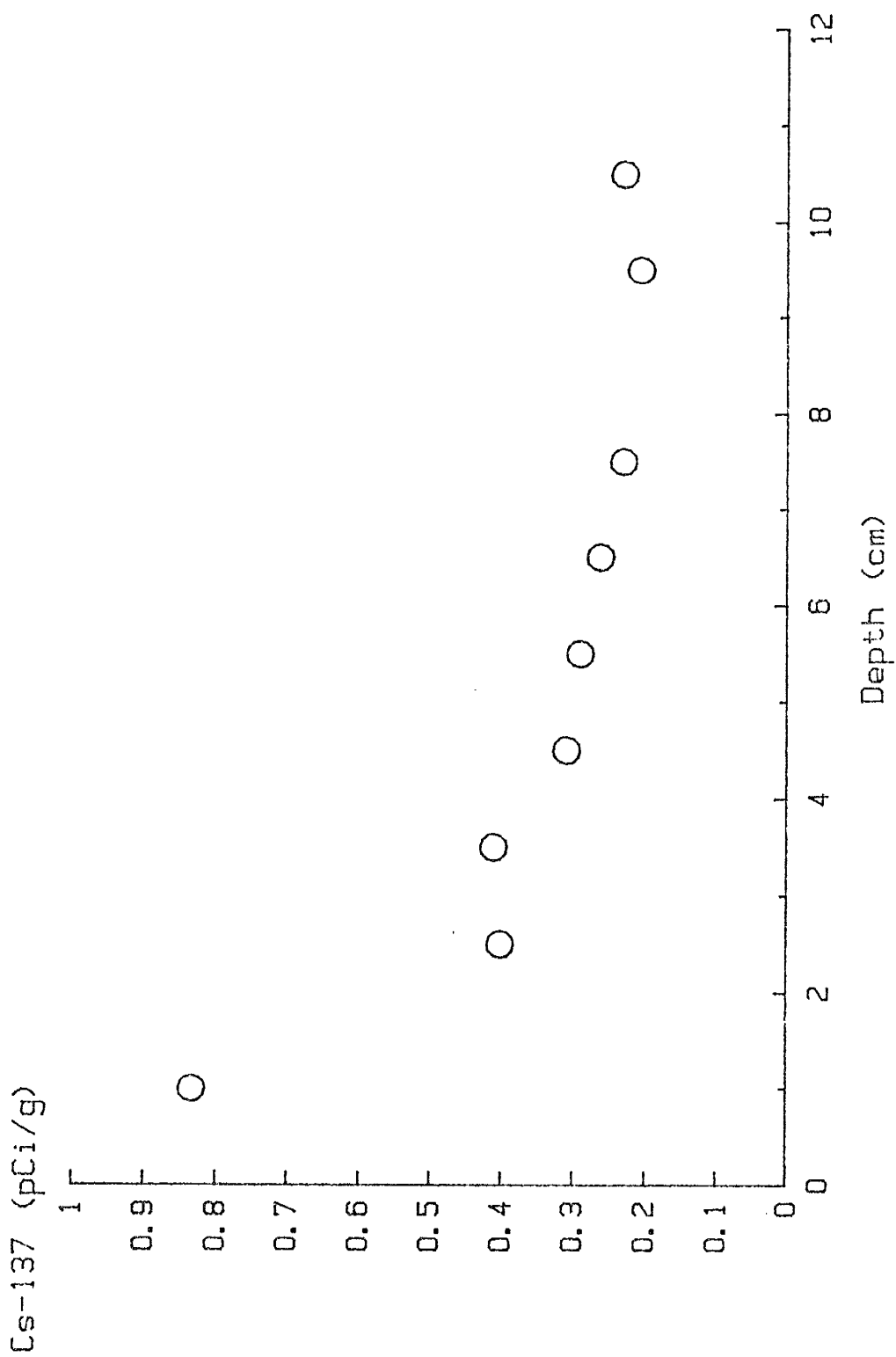


Figure 12. Cs-137 activity profile for the sediment core from Sugarloaf Lake.

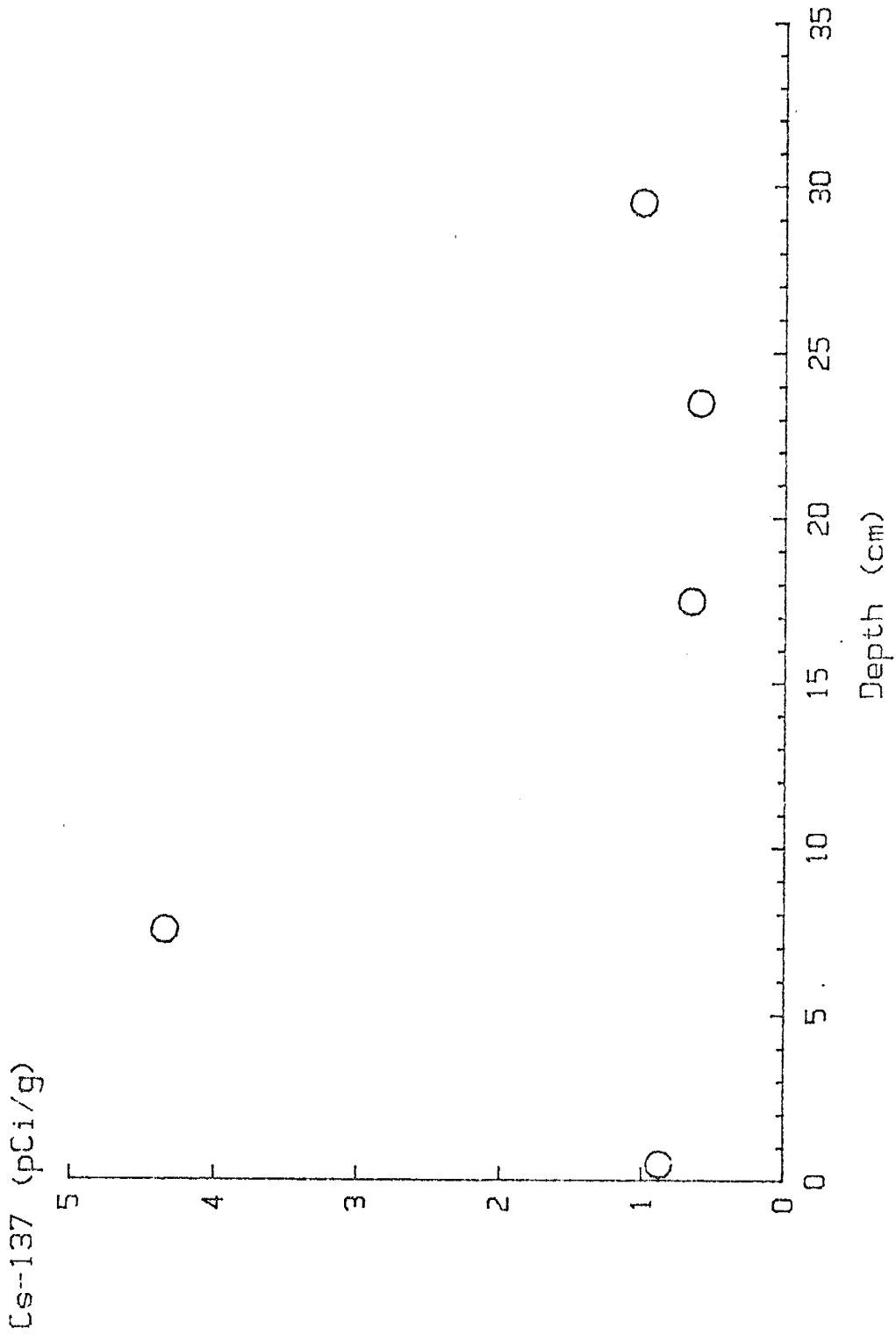


Figure 13. Cs-137 activity profile for the sediment core from Lagunitas #3 Lake.

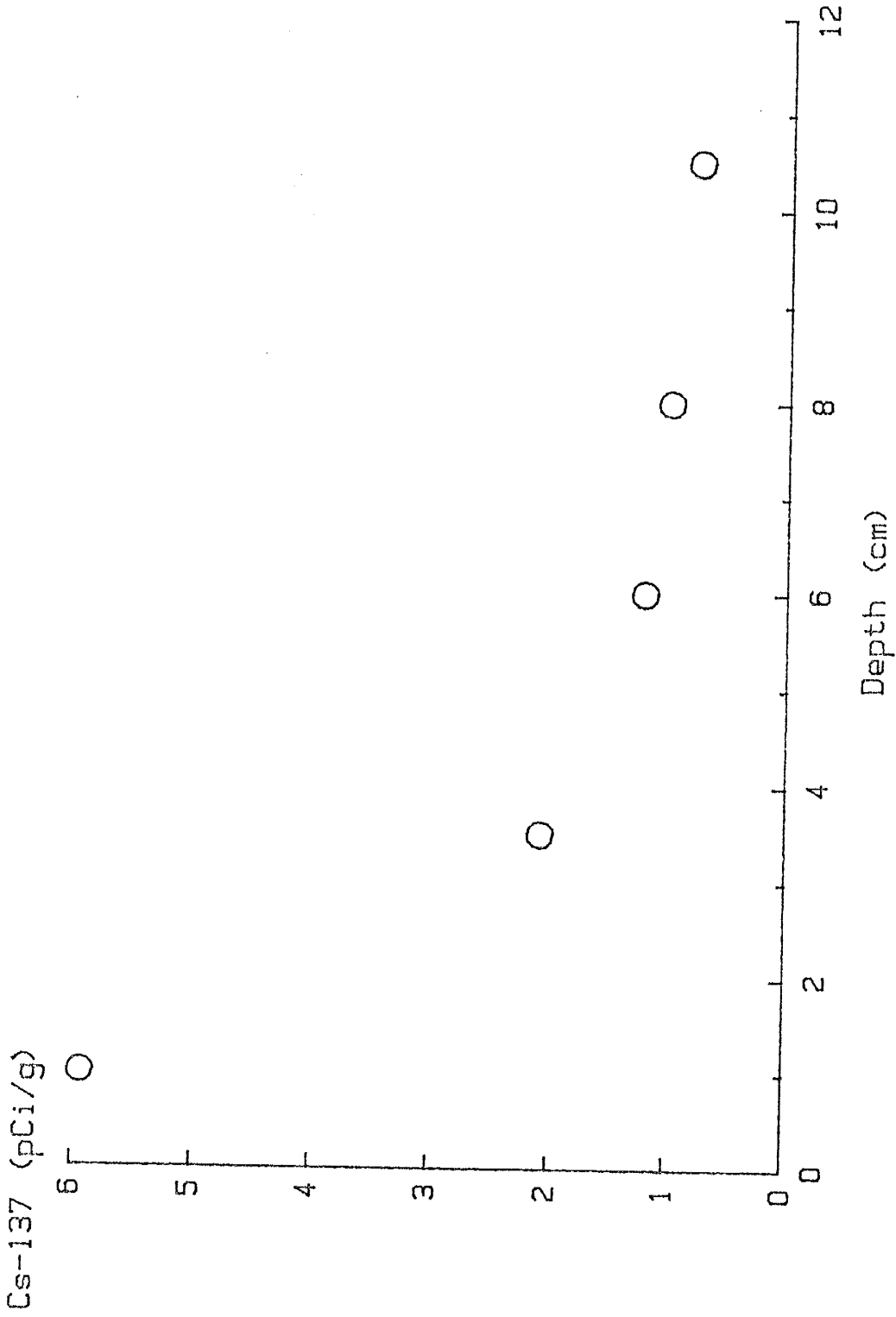


Figure 14. Cs-137 activity profile for the sediment core from Santa Fe Lake.

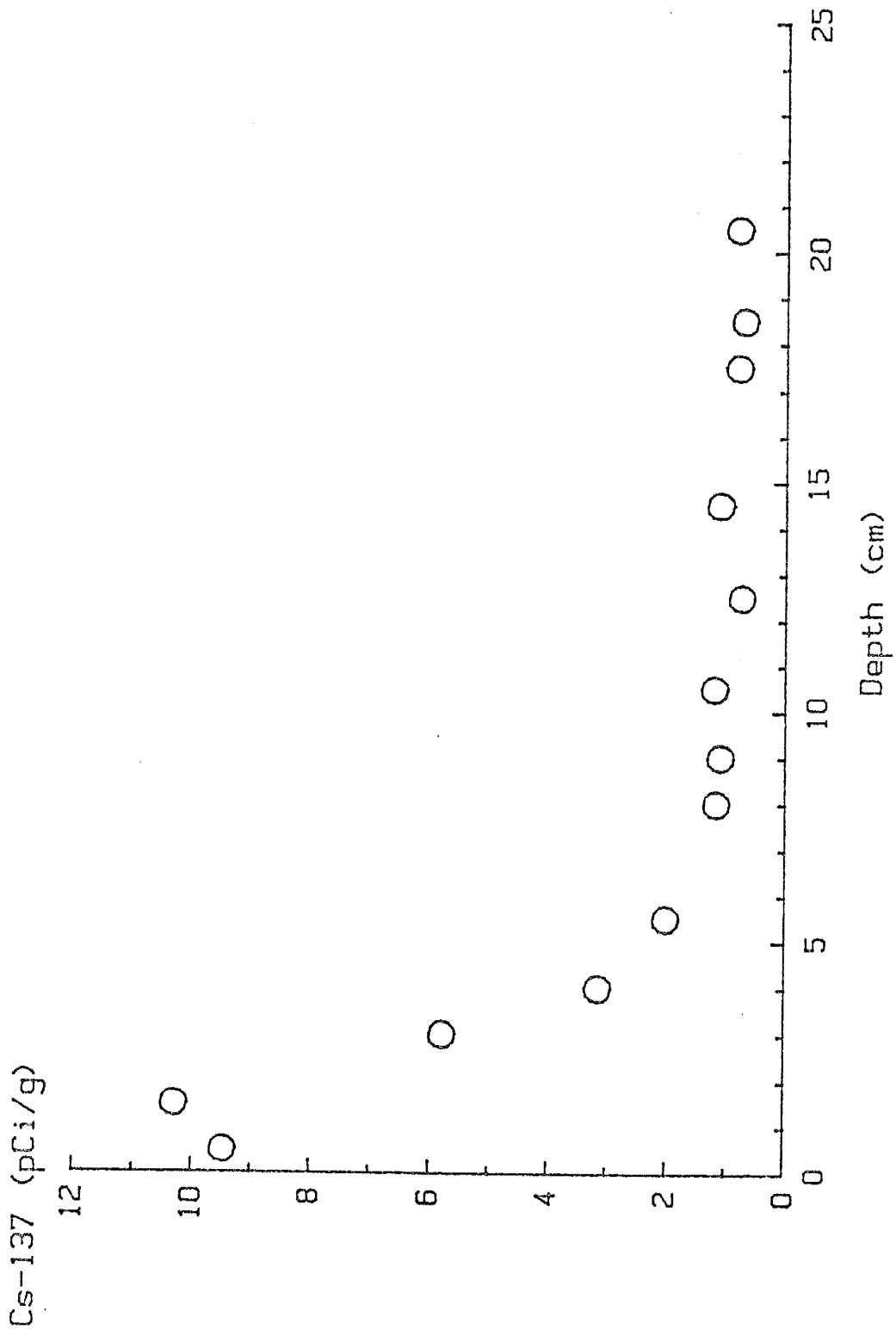


Figure 15. Cs-137 activity profile for the sediment core from Truchas Lake.

Pb-210 may not be affected by mobility problems such as those which plague Cs-137. Pb-210 behaves similarly to transition metals which have been found to be very strongly adsorbed. The probable reason for this is that the structure of these atoms allow some covalent bonding to occur between the lead and the clay surface (Bowman, pers. comm.).

The Pb-210 in sediments from Santa Fe, Lost, and Truchas lakes all exhibit exponential decay profiles (figures 16, 17, and 18). The profiles from Lost and Truchas lakes both deviate somewhat from this exponential profile at the top of the cores. This may very well be caused by increased sedimentation rates (which was the assumption used when calculating the ages shown in tables 13 and 21). However, there is no evidence to show that physical mixing of the upper-most sediments due to something such as bioturbation or sample collection and processing occurred.

The Pb-210 in sediments from Lagunitas #3 and Sugarloaf lakes does not exhibit exponential decay profiles (figures 19 and 20). This may be explained by increasing sedimentation rates through time but the increases are rather extreme (tables 14 and 16).

Sugarloaf Lake is a man made lake dammed in 1953 (Leo Smith, pers. com.). The soil surrounding the lake is rocky. The sediment core from Sugarloaf Lake was not collected using the modified Brinkhurst corer but rather



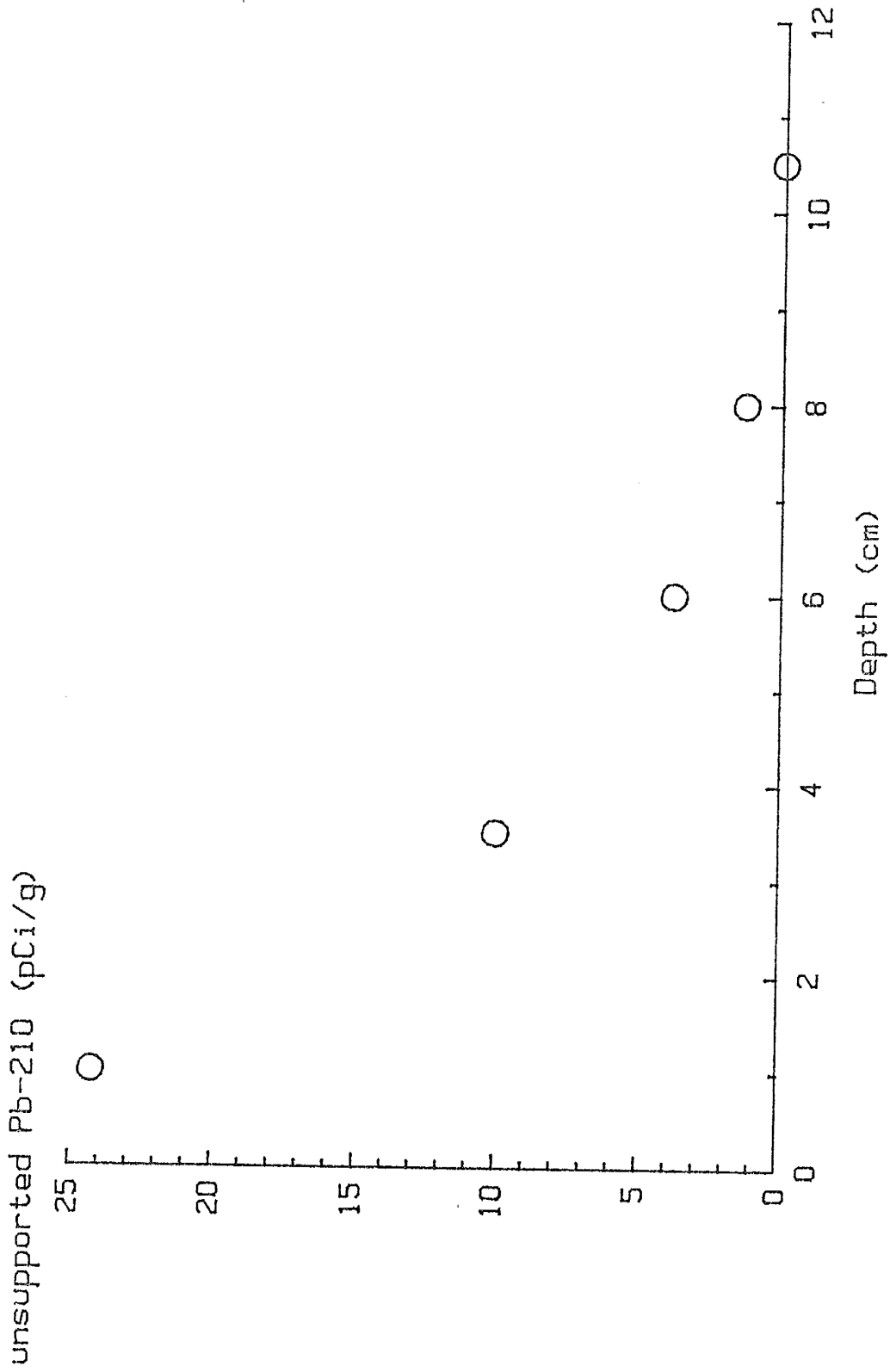


Figure 16. Unsupported Pb-210 activity profile for the sediment core from Santa Fe Lake.

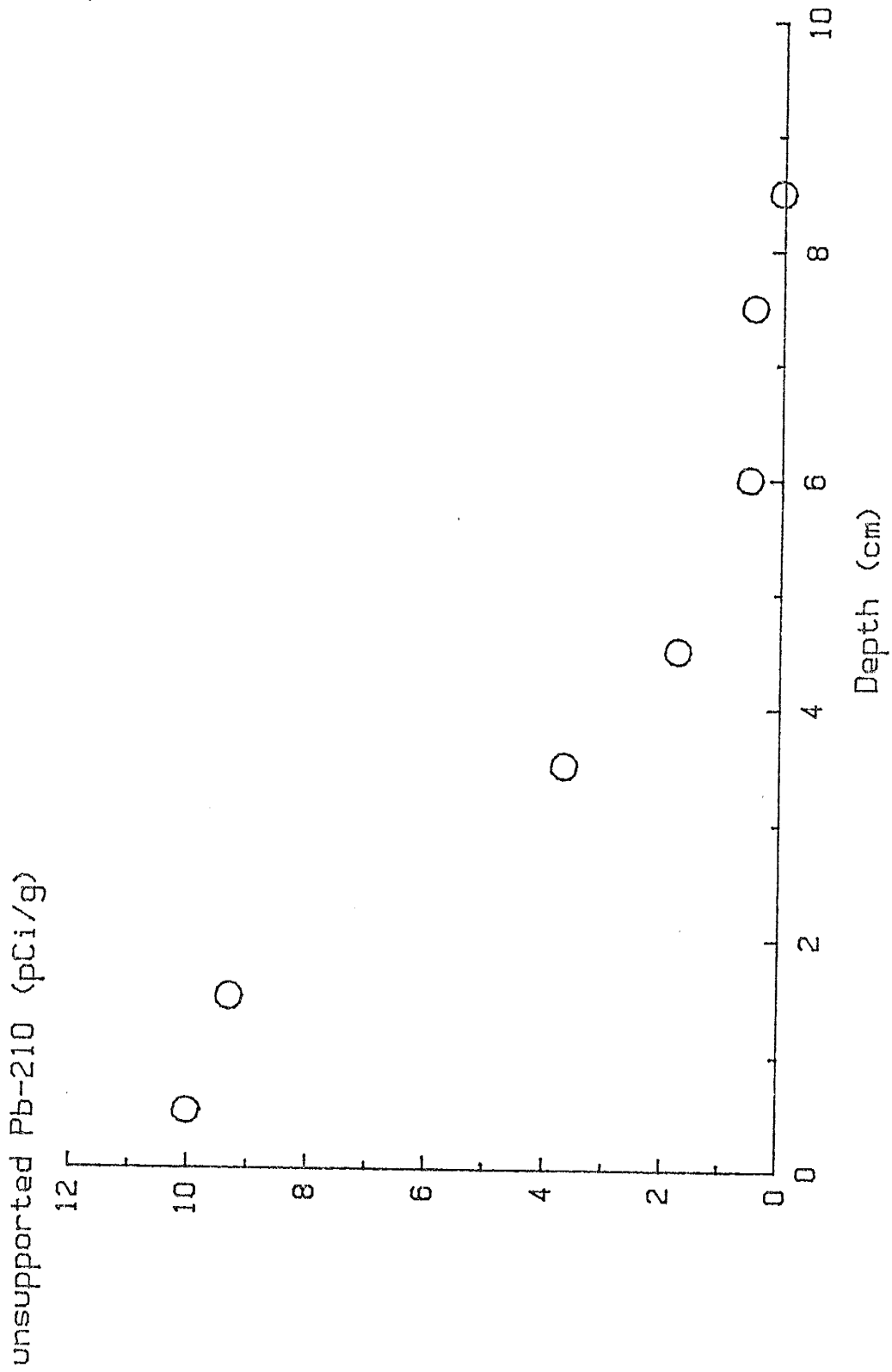


Figure 17. Unsupported Pb-210 activity profile for the sediment core from Lost Lake.

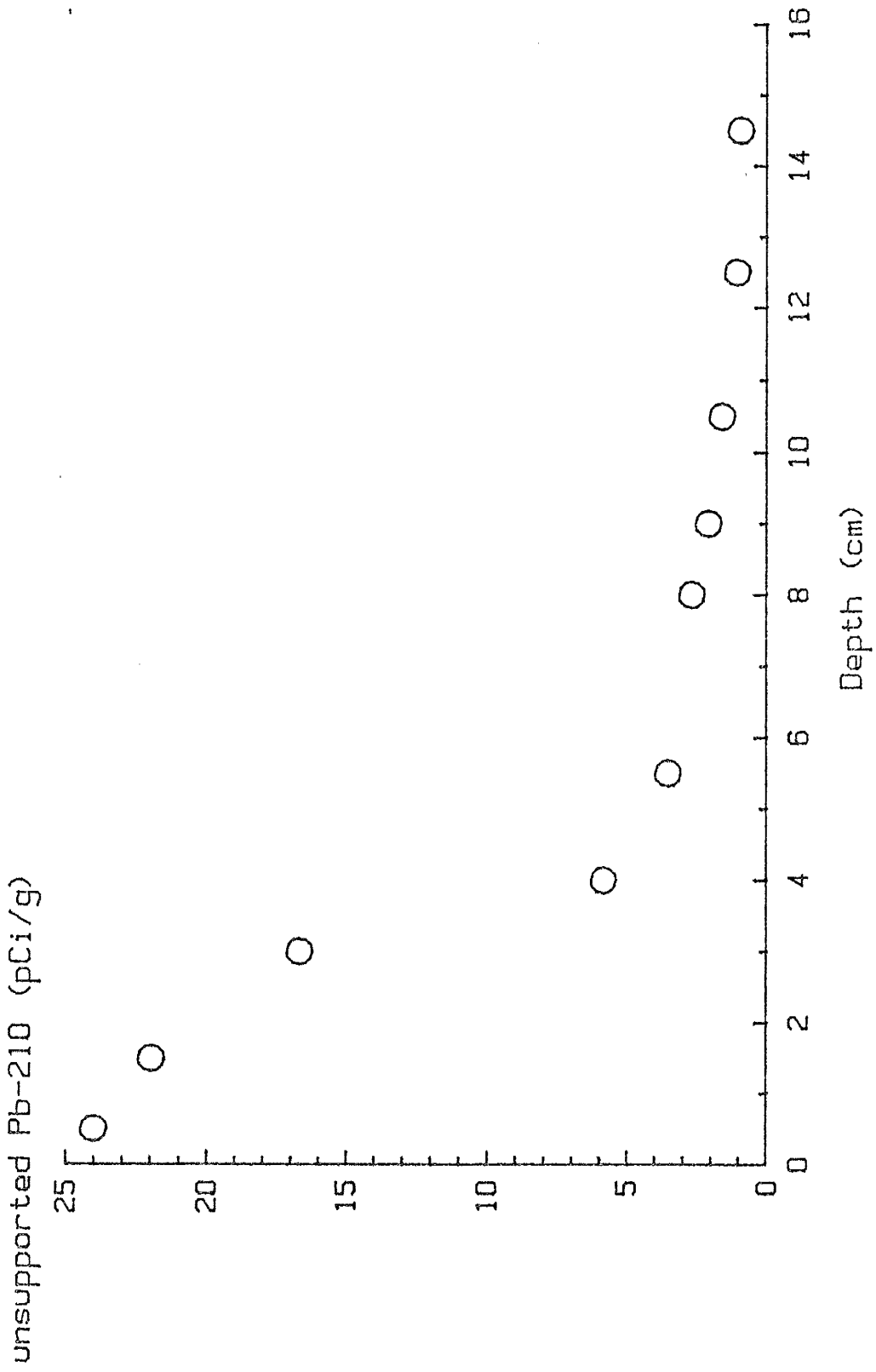


Figure 18. Unsupported Pb-210 activity profile for the sediment core from Truchas Lake.

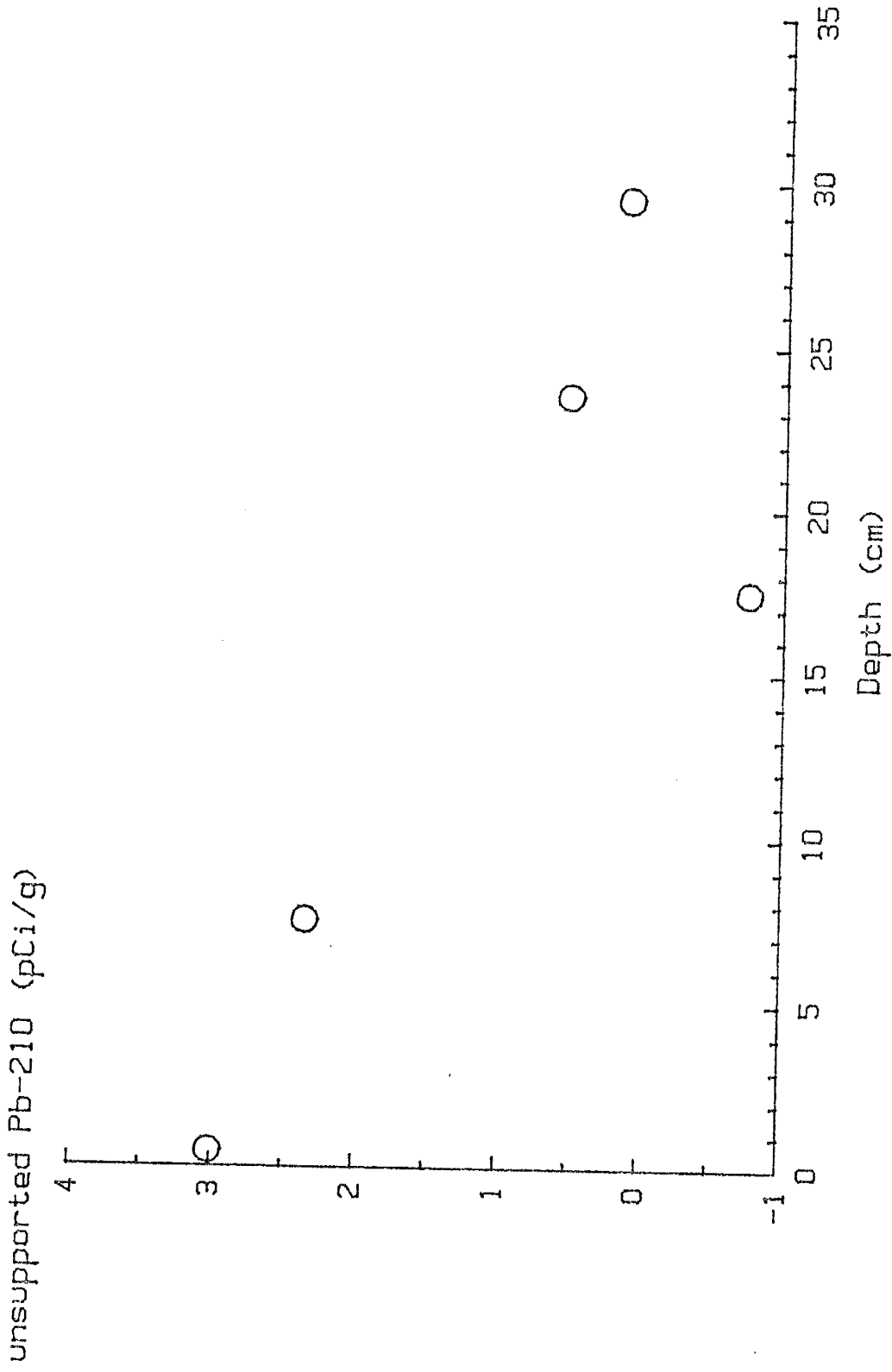


Figure 19. Unsupported Pb-210 activity profile for the sediment core from Lagunitas #3 Lake.

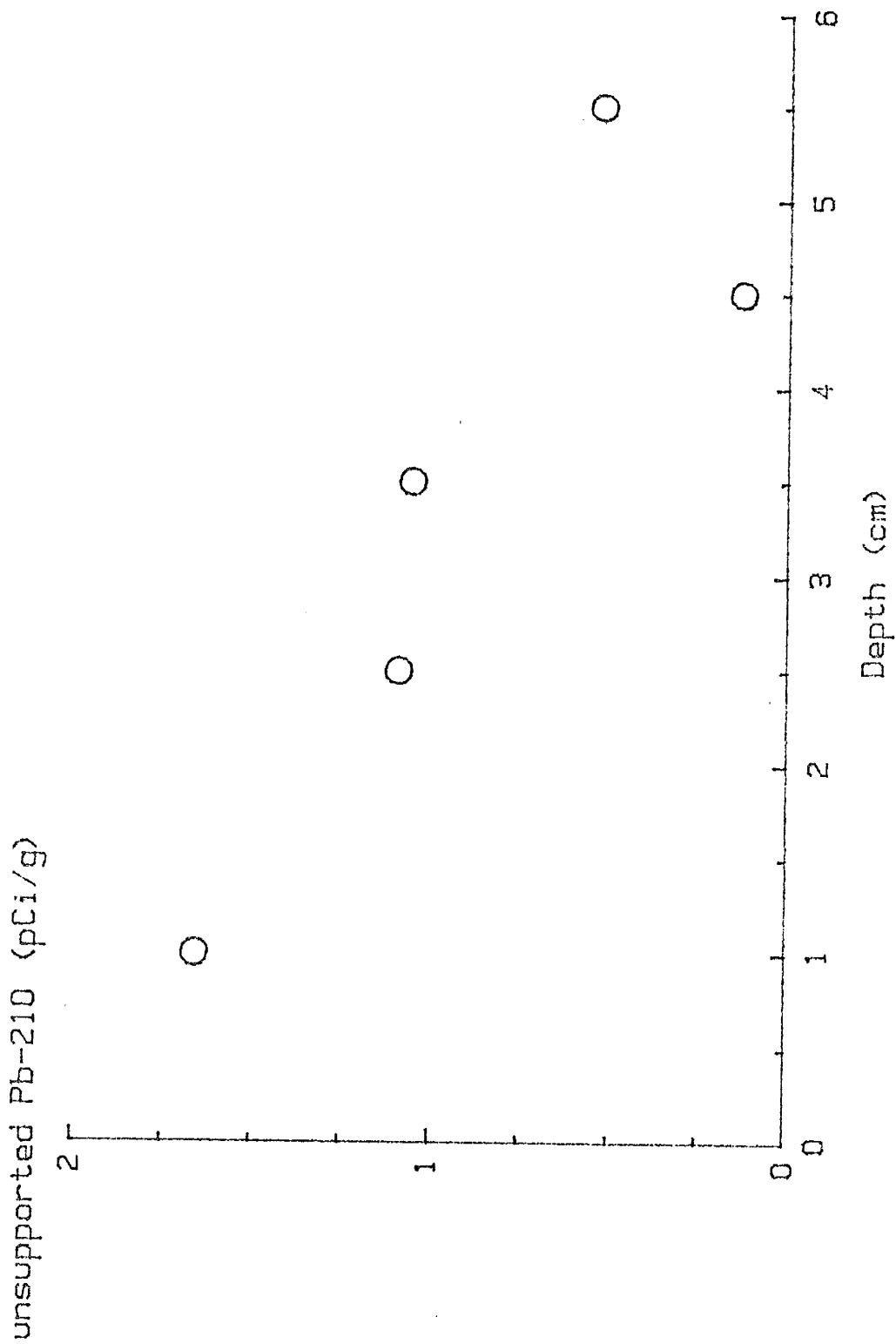


Figure 20. Unsupported Pb-210 activity profile for the sediment core from Sugarloaf Lake.

the core tube was fitted to the end of a long pole and pushed into the lake sediment by hand until bedrock was contacted. Considering the rockiness of the soil surrounding the lake it is probable that the sediments penetrated by the corer (which contained no large rocks) were deposited since 1953. The dates obtained using Pb-210 are much older than this (table 15). Although it is not certain that all of the sediments contained in the core were deposited since 1953, the doubt which this problem casts upon the Pb-210 dates combined with the non-exponential Pb-210 profile suggests that the Pb-210 dates probably are not accurate.

Lagunitas #3 Lake is a naturally occurring beaver pond of unknown age. The Pb-210 profile for this lake exhibits the same kinds of problems as the Sugarloaf Lake profile. The profile is non-exponential and the unsupported Pb-210 activities are very low in even the top core slice. Because of this the Pb-210 dates for this core are probably not accurate.

The unsupported Pb-210 activities in the top core slices from both Sugarloaf and Lagunitas #3 are very low with respect to the activities measured in top core slices from Lost, Santa Fe, and Truchas lakes. The basin area to lake surface area ratios are much higher for Sugarloaf and Lagunitas #3 lakes than for the other three lakes examined (table 1). Soil development is more extensive in the heavily forested Sugarloaf and

Lagunitas #3 basins than in the basins containing Lost, Santa Fe, and Truchas lakes. Clearly the opportunities for delayed entry of unsupported Pb-210 are highest for Lagunitas #3 and Sugarloaf lakes.

Apparently Pb-210 is held up in the lake basin soils so long that most of the unsupported Pb-210 decays before it can reach the lakes to be incorporated into their sediments. As a result unsupported Pb-210 activities in these sediments are just too low to allow accurate Pb-210 dating.

To test the significance of the basin area to lake surface area ratio on the amount of Pb-210 delivered to lake sediments, unsupported Pb-210 activity was measured in the top core slice from Hidden Lake. Hidden Lake lies about 7.5 km east of Sugarloaf Lake. The rock types found in the Hidden Lake basin are identical to those found in Sugarloaf basin. However, the basin area to lake surface area ratio is only 6.2 as compared to 82.2 for Sugarloaf Lake. Unsupported Pb-210 activity measured in the top core slice from Hidden Lake was 16.729 pCi/g as opposed to 1.657 pCi/g in the top two slices from Sugarloaf Lake.

#### Correlation of Trace Metal Concentrations in Lake Sediment with Age

Trace metal concentrations within Santa Fe Lake and Truchas sediments were determined by Svec (in progress)

and Lynch et al. (1988). The Pb-210 dating performed for these two lakes now allows these concentrations to be correlated with absolute ages (figures 21 and 22).

The iron profile in Santa Fe Lake sediment is another indicator of the weathering history in the Santa Fe Lake basin. Like lithium, iron inputs from weathering should greatly exceed atmospheric inputs. Iron concentration begins to increase in sediments deposited about 150 years ago. This suggests an increase in the weathering rate in the Santa Fe Lake basin over the last 150 years. This increasing trend continues all of the way to the top of the core with the greatest increase taking place in the time span beginning about 110 years ago and ending about 50 years ago. Such a change in the weathering pattern is probably caused by increased precipitation.



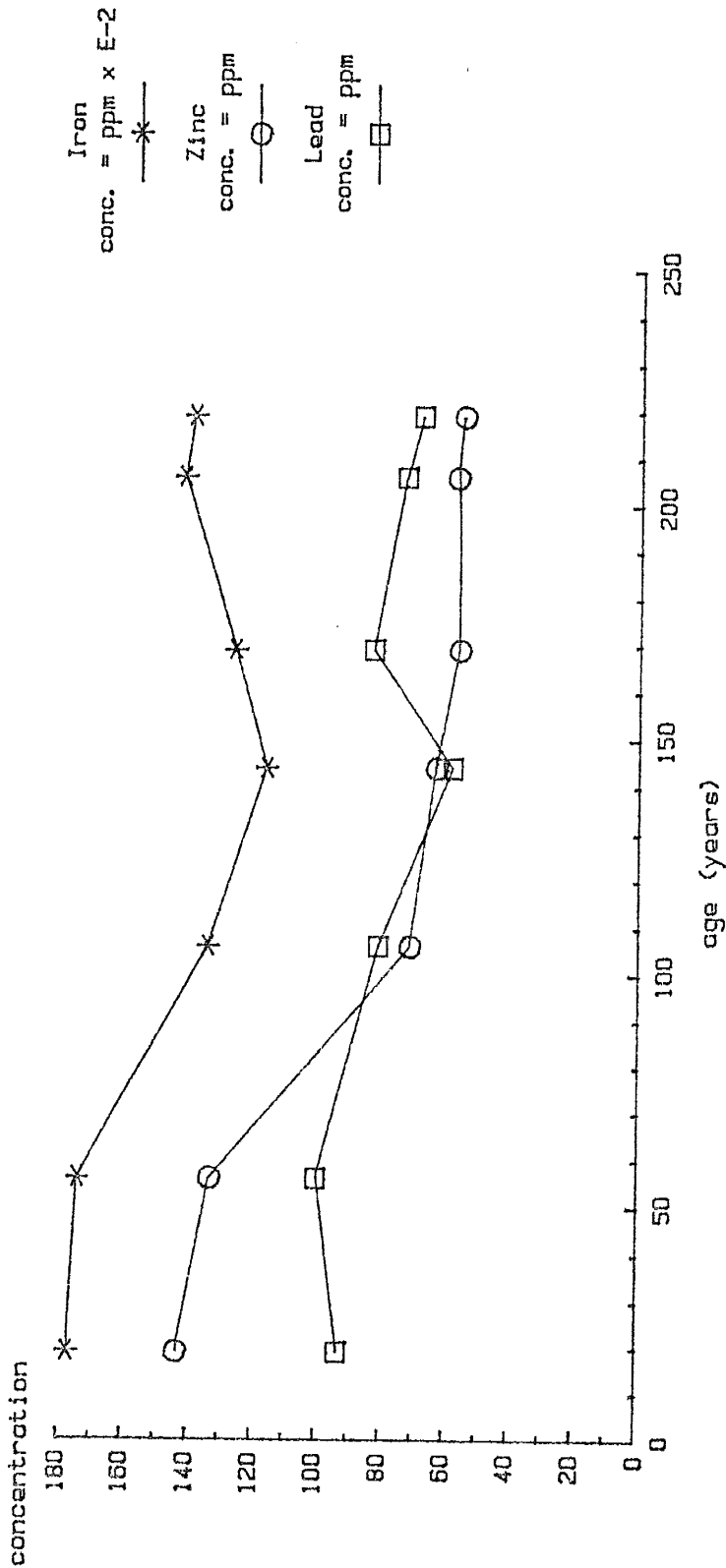


Figure 21. Iron, zinc, and lead concentrations in Santa Fe Lake sediment plotted versus age.

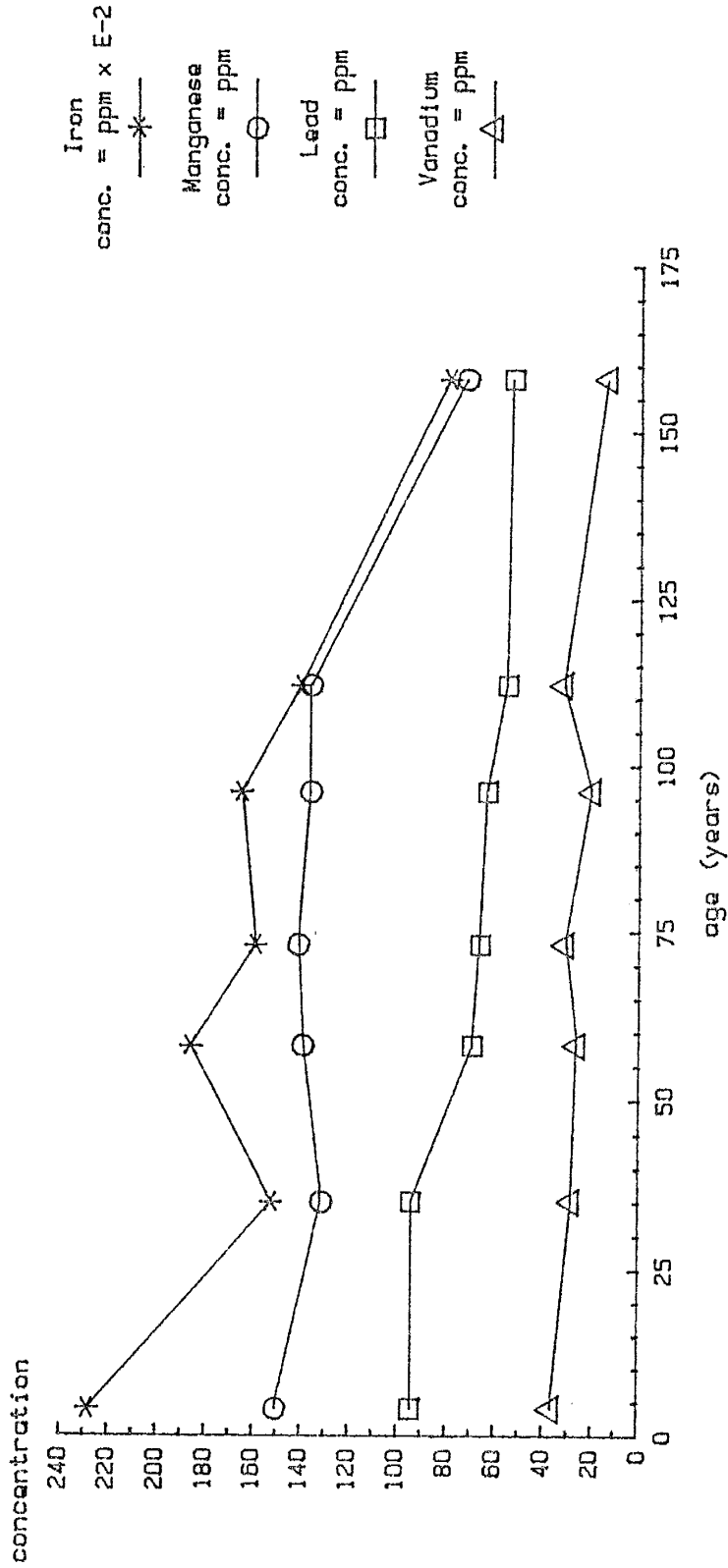


Figure 22. Iron, manganese, lead, and vanadium concentrations in Truchas Lake sediment plotted versus age.

### Conclusions

Atmospheric inputs account for about 40% of the  $\text{Ca}^{++}$ ,  $\text{Mg}^{++}$ , and  $\text{Na}^+$  found in Santa Fe Lake water as well as about 50% of the  $\text{SO}_4^{--}$  and virtually all of the  $\text{Cl}^-$ . The concentrations of  $\text{K}^+$  in precipitation samples analyzed are also large relative to concentrations measured in lake water samples but this is probably caused by contact with vegetation and therefore represents  $\text{K}^+$  originally derived from both weathering and the atmosphere.

The weathering of biotite and plagioclase to an amorphous phase of kaolinitic composition accounts for virtually all of the basin derived inputs. However, an unidentified source of sulfate is also present in the basin. Although never observed, the most likely source of sulfate is a sulfide mineral such as pyrite. Some weathering of microcline almost certainly exists as well but its significance is masked by  $\text{K}^+$  inputs from vegetation.

Over 70% of the dissolved inputs (on a molar basis) are derived from the weathering of plagioclase. This follows the classical weathering scheme of Goldich (1938) which shows that andesine is more easily weathered than biotite which in turn is more easily weathered than microcline or muscovite.

Changes in lithium concentrations with depth in the

Santa Fe Lake core are rather erratic. The linear regression line for this data suggests a slight increase in lithium concentration with depth but with a correlation coefficient of 0.34 this data is rather inconclusive.

The iron profile in the Santa Fe Lake sediment core provides a clearer record of the history of weathering trends in the Santa Fe Lake Basin than does the lithium profile. The pronounced increase in the concentration

of iron which first occurs in sediments deposited about 150 years ago suggests that weathering rates have been increasing since that time.

Because of mobility problems and time delays between fall-out and incorporation into lake sediments, Cs-137 profiles in all of the lake sediments examined were found to not be representative of documented fall-out histories and therefore were not of use in dating.

Entry delays of Pb-210 into lake sediments were found to be so long for Sugarloaf and Lagunitas #3 Lakes that radioactive decay eliminated so much of the unsupported Pb-210 before it could be incorporated into lake sediments that dating was not possible.

The exponential unsupported Pb-210 profiles for sediment from Lost, Santa Fe, and Truchas lakes indicate that Pb-210 dating for these lake sediments is probably accurate. The non-exponential behavior exhibited in the tops of the Lost Lake and Truchas Lake cores may be

explained by accelerated sedimentation rates in recent years. However, no evidence is available at this time to discount the possibility of physical sediment mixing as the cause of this non-exponential behavior.

### Further Studies

Since Cs-137 dating was found to be unreliable in all of the lakes studied, no backup dating method was available to serve as a cross-check for the Pb-210 dates. An alternative dating method such as pollen chronology would be desirable to see if the dates measured using both methods are in agreement.

The importance of solute sources not considered in this study are assumed to be insignificant but their actual significance is not clear. Data presented by Lynch et al. (1988) suggests that at least for a short time each year respiration is a significant contributor to alkalinity in Santa Fe Lake. The magnitude of  $\text{HCO}_3^-$  inputs from this source with respect to total  $\text{HCO}_3^-$  inputs on an annual basis is unknown. The importance of dry deposition versus wet deposition is also unknown.

It would be possible to check if the proposed weathering reactions are indeed representative of what is actually occurring at Santa Fe Lake if the weathering mechanisms could be duplicated in the more controlled environment of the laboratory. This could be approached by performing an artificial weathering experiment using water and crushed rock from the Santa Fe Lake basin. Are ions really produced in the proportions calculated in this study? Does acid rain significantly affect weathering rates and if so could this be used to

reconstruct acid deposition histories in other lakes?

Appendix A. Geologic descriptions of the lake basins.

Lost Lake

Lost Lake basin contains mostly Precambrian tonalite, granite, and amphibole gneiss. Some paleozoic sedimentary outcrops occur in this basin as well. These consist mostly of quartz arenites and quartz pebble conglomerates with minor amounts of calcilutite.

Sugarloaf Lake

This basin contains Tertiary volcanoclastic rocks of intermediate composition belonging to the Conejos Formation. These rocks are mostly andesite and quartz latite breccias but small amounts of tuffaceous sandstone are also present. A basaltic member of the Conejos Formation caps the ridge to the north of the lake.

Lagunitas #3 Lake

Lagunitas #3 Lake basin contains quartz latite and volcanoclastic sandstone and conglomerate belonging to the Tertiary Treasure Mountain Tuff. Capping the ridge to the north of the lake is a rhyolitic welded tuff belonging to the Tertiary Masonic Park Tuff.



Santa Fe Lake

Santa Fe Lake basin contains Precambrian granodiorite, quartz diorite, biotite gneiss, and numerous pegmatite dikes. Common minerals are quartz, plagioclase, microcline, muscovite, and biotite.

Truchas Lake

Truchas Lake basin lies entirely within the Precambrian Ortega Quartzite. Although this rock is almost entirely quartz, it also contains a wide variety of accessory minerals. These include muscovite, kyanite, sillimanite, specular hematite, piedmontite, chlorite, and garnet.

Appendix B. Sediment texture of selected core slices from Lost, Sugarloaf, Lagunitas #3, Santa Fe, and Truchas Lakes.

<u>Sample</u>	<u>%Sand</u>	<u>%Silt</u>	<u>%Clay</u>
Lost-3	7.2	88.3	4.5
Lost-11	44.8	51.1	4.0
Lost-14	75.5	23.0	1.5
SGL-1	33.1	50.7	16.1
SGL-7	38.1	51.6	10.3
Lag3-7	1.2	82.0	16.8
Lag3-23	0.3	78.1	21.6
Lag3-32	0.2	66.8	33.0
SF-4	1.5	80.7	17.9
SF-13	1.6	93.7	4.8
SF-18	0.9	87.3	11.7
T-5	5.4	86.2	8.4
T-15	3.6	88.7	7.7
T-19	5.2	88.6	6.2

Appendix C. Clay mineralogy of selected core slices from Lost, Sugarloaf, Lagunitas #3, Santa Fe, and Truchas Lakes. Clay mineral abundances are given in terms of parts in ten. Because of errors introduced by rounding off, the sum of all of the clay mineral components does not always add up to ten.

Lost Lake

<u>Clay Mineral</u>	<u>Parts In Ten</u>		
	<u>Lost-3</u>	<u>Lost-11</u>	<u>Lost-14</u>
illite	4	4	4
chlorite	3	2	3
kaolinite	1	1	1
mixed layer illite/smectite	2	2	2

Sugarloaf Lake

<u>Clay Mineral</u>	<u>Parts In Ten</u>	
	<u>SGL-1</u>	<u>SGL-7</u>
illite	2	2
smectite	1	0
kaolinite	4	4
mixed layer illite/smectite	4	3

Lagunitas #3 Lake

<u>Clay Mineral</u>	<u>Parts In Ten</u>		
	<u>Lag3-7</u>	<u>Lag3-23</u>	<u>Lag3-32</u>
smectite	9	8	4
kaolinite	0	0	1
mixed layer illite/smectite	2	1	5

Santa Fe LakeClay Mineralogy

trace of kaolinite; mostly x-ray amorphous

Truchas Lake

Clay Mineral	<u>Parts In Ten</u>		
	<u>T-5</u>	<u>T-15</u>	<u>T-19</u>
illite	2	2	2
smectite	1	<1	<1
kaolinite	6	6	6
mixed layer illite/smectite	<1	1	1

Appendix D. Santa Fe Lake water chemistry

date	pH	Ca++ (ppm)	Mg++ (ppm)	Na+ (ppm)	K+ (ppm)	Fe (ppm)	Al+++ (ppm)	SiO2 (ppm)	Cl- (ppm)	NO3-- (ppm)	SO4-- (ppm)	HCO3-- (ppm)	D.O. (ppm)	Temp. (deg. C)
7/1/87	6.92	2.50	0.55	1.12	0.30	0.10	0.06	2.21	0.16	0.02	2.49	9.38	7.8	13.5
8/11/87	6.56	2.45	0.69	1.70	0.44	0.10	0.06	2.18	0.16	0.01	2.30	10.18	7.5	16.5
9/3/87	5.86	2.50	0.65	2.75	0.74	0.22	0.14	2.38	0.60	0.23	2.38	12.40	7.1	11.5
10/16/8	5.95	3.40	0.86	1.50	0.58	0.16	0.06	1.82	0.24	0.01	2.32	8.12	7.6	11.5
11/12/8	7.15	2.40	0.78	1.90	0.33	0.12	0.06	1.82	0.27	0.02	2.60	7.44	9.2	0.8
12/16/8	7.03	2.40	0.84	1.90	0.30	0.25	0.22	2.18	0.37	0.18	2.52	11.40	6.15	0.2
2/11/88	6.89	2.40	0.75	1.40	0.25	0.10	0.06	1.82	0.57	1.50	0.80	10.12	4.8	0.0
3/17/88	6.10	1.73	0.60	1.29	0.40	0.10	0.06	1.87	0.48	1.15	2.25	8.36	3.1	0.5
4/17/88	5.69	2.09	0.76	1.31	0.39	0.10	0.04	2.04	0.37	1.16	2.36	10.56	4.2	0.5
5/12/88	5.68	1.90	0.34	0.93	0.30	0.16	0.11	1.36	0.90	0.44	1.46	12.02	4.9	2.5
5/27/88	5.97	2.40	0.65	1.00	0.45	0.25	0.08	1.70	0.34	1.29	1.97	7.26	6.4	6.0
6/14/88	6.11	2.12	0.49	1.54	0.39	0.16	0.11	1.87	0.20	0.01	1.87	7.56	6.8	12.0

## Appendix E. Santa Fe Lake precipitation chemistry

date	sample type	pH	Ca++ (ppm)	Mg++ (ppm)	Na+ (ppm)	K+ (ppm)	SiO2 (ppm)	Cl- (ppm)	NO3- (ppm)	SO4-- (ppm)	HCO3- (ppm)
5/25/86	snow	5.41	1.74	0.16	2.14	2.75	NA	BDL	1.85	2.40	0.09
7/1/87	snow	4.95	1.18	0.30	0.73	0.52	0.18	0.57	0.23	0.38	0.03
9/3-10/13/87	rain	5.35	0.25	2.20	1.70	0.66	NA	1.29	1.22	3.72	0.08
10/14/87	snow	4.20	0.40	0.05	0.40	0.19	NA	0.36	0.01	0.55	0.08
10/15-11/11/87	rain	5.39	0.49	0.15	0.58	0.17	NA	0.28	0.49	1.14	0.08
11/11/87	snow	5.73	1.25	0.24	1.95	0.10	NA	0.39	1.22	1.52	0.18
12/16/87	snow	4.76	0.58	0.06	0.21	0.16	NA	0.66	0.53	0.39	0.02
1/7/88	snow	5.37	0.50	0.03	0.10	0.10	NA	0.50	0.83	0.98	0.08
2/11/88	snow	5.85	2.60	0.11	0.13	0.97	NA	0.15	0.86	1.01	0.24
3/17/88	snow	5.33	0.86	0.05	0.30	0.12	NA	0.78	1.64	1.11	0.07
4/17/88	snow	5.12	0.50	0.03	0.20	0.07	NA	0.62	0.29	0.38	0.04
5/12/88	snow	6.11	1.10	0.19	0.15	0.67	NA	0.38	0.30	0.56	0.44
5/27/88	snow	6.84	1.10	0.14	0.21	0.20	NA	0.24	0.63	0.89	2.38
6/14/88	snow	4.50	0.19	0.03	0.15	0.16	NA	0.31	0.23	0.25	0.01

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