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DISTRIBUTION AND CHRONOLOGY OF $^{239,240}Pu$ AND ^{137}Cs IN COLORADO FRONT RANGE LAKE SEDIMENTS

by

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ABSTRACT

This study was undertaken in the summer of 1988 in order to determine the concentrations of $^{239,240}Pu$ and ^{137}Cs in Colorado Front Range Lake sediments as a result of atmospheric nuclear weapons testing. The research was initiated by Rockwell International, the current operator of the Rocky Flats plutonium processing plant north of Golden, Colorado. The research was conducted in order to compare background $^{239,240}Pu$ and ^{137}Cs concentrations in lake sediments with those of two reservoirs believed to be impacted by activities at Rocky Flats.

Triplicate cores were taken from Halligan Reservoir and Wellington Lake, control reservoirs chosen on the basis of distance from Rocky Flats and similarity of physical parameters to the impacted reservoirs. The cores were sectioned in 1 cm increments, combined, homogenized and analyzed for ^{137}Cs using gamma ray spectrometry. $^{239,240}Pu$ analysis was done using alpha spectrometry.

Results indicated $^{239,240}Pu$ maxima of 0.074 and 0.19 pCi/g for sediments in Halligan Reservoir and Wellington Lake respectively. ^{137}Cs analysis yielded maxima of 2.2 and 6.2 pCi/g respectively.

These values were compared with concentrations found in

previous studies for Standley Lake and Great Western Reservoir, which have been impacted by activities at the Rocky Flats Plant.

$^{239,240}Pu$ contamination of the sediments in Great Western Reservoir appears to be between 25 and 77x the background concentration. Plutonium contamination in Standley Lake appears to be between 9 and 43x the background concentrations. ^{137}Cs concentrations observed in Standley Lake and Great Western Reservoir are on the same order or lower than concentrations observed in both Halligan Reservoir and Wellington Lake.

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This thesis is dedicated to my mother and father

Chapter 1

Introduction

The U.S. Department of Energy Rocky Flats Plant, located northwest of Denver, CO. has been producing and reprocessing plutonium components for use in atomic weapons since 1953. Past research has suggested that drainage of the Rocky Flats Plant site by Walnut and Woman creeks into Standley Lake and Great Western Reservoir respectively, has led to offsite contamination of the lake sediments with isotopes of plutonium, cesium and other radionuclides (Figure 1.1).

In 1973, the U.S. Environmental Protection Agency (EPA) conducted a sediment survey at Standley Lake. ^{239,240}Plutonium levels in the sediments were found to be a maximum of 4.1 pCi/g dry weight.

Battelle Pacific Northwest Laboratories conducted a sediment study of Great Western Reservoir in 1974, taking a total of nine gravity cores. A single core sample was taken from Standley Lake. Determinations of the ^{239,240}Pu concentrations revealed maxima of 13.4 disintegrations per minute/gram, dpm/g (6.09 pCi/g) in Great Western Reservoir and approximately 0.85 dpm/g (0.39 pCi/g) for Standley Lake.

The Hardy, Livingston, Burke and Volchok study in 1976

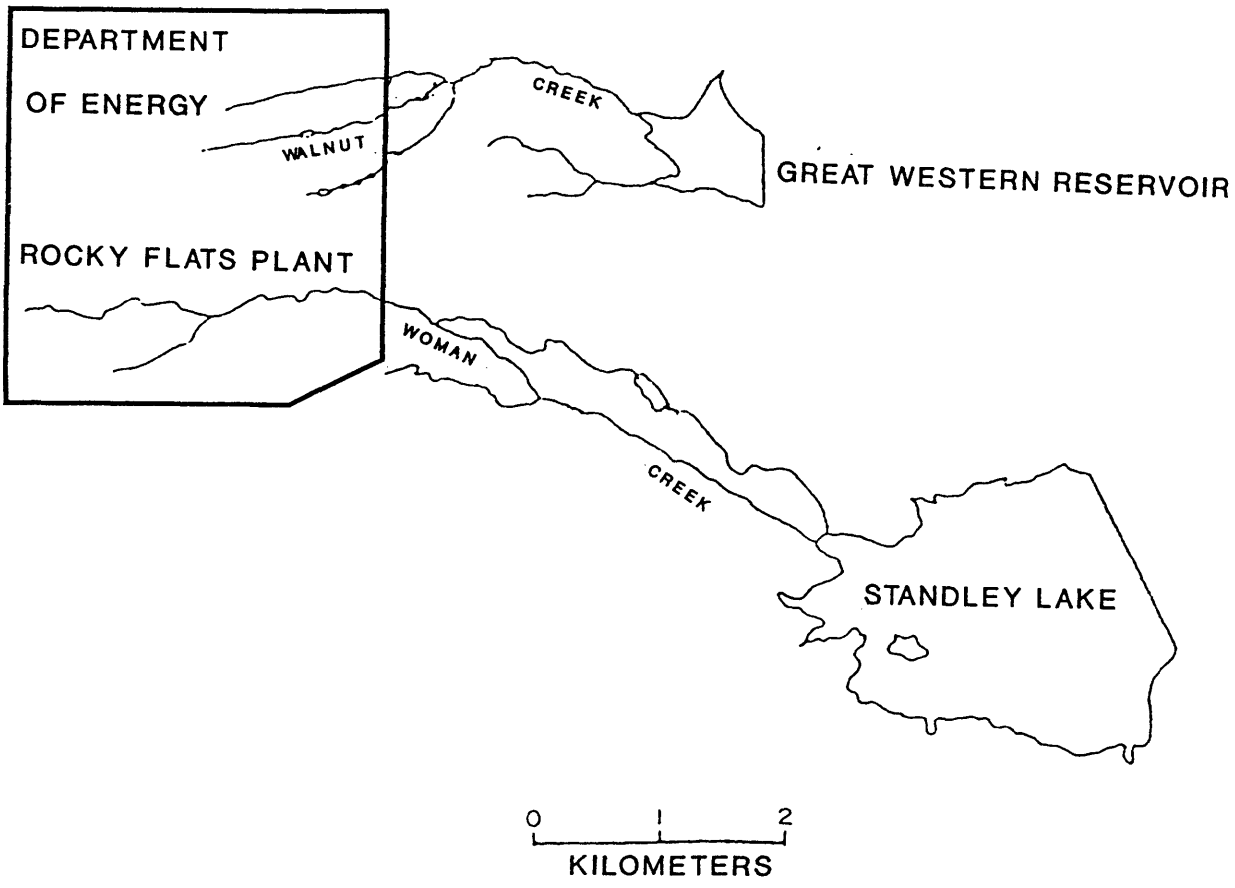

Figure 1.1 Detail of the Rocky Flats Plant and vicinity.

(Source: Rockwell International, after G. Setlock, 1983).

focused on a single, 50 cm core of Standley Lake sediment (Hardy et al., 1978). Maximum ^{137}Cesium levels were determined to be approximately 6 dpm/g. Maximum ^{239,240}Pu levels were determined to be 25.8 x 10^3 pCi/m^2. The study also suggested that approximately 70% of this quantity was a result of releases from Rocky Flats operations and occurred in the uppermost 30 cm of the sediment core.

A similar study conducted in 1983 by Rockwell International analyzed the lake sediments of Great Western Reservoir. Both core and surficial sediment samples were taken. Maximum ^{239,240}Pu levels were determined to be between 4.9 and 5.4 pCi/g. Maximum ^{137}Cs concentrations were found to be approximately 1.4 pCi/g (Setlock, 1983). Appendix 1 gives the ^{239,240}Pu and ^{137}Cs distribution with depth in the Standley Lake core analyzed in 1976. The ^{239,240}Pu distribution with depth for the Great Western Reservoir cores analyzed in 1983 is given in Appendix 2-6. All of the studies revealed levels that exceeded the Colorado Department of Health guideline of 0.9 pCi/g for plutonium in sediment. This study was undertaken to compare the levels of ^{239,240}Pu and ^{137}Cs in Standley Lake and Great Western Reservoir with the concentrations found in two reservoirs located a considerable distance from, and out of the influence of, the Rocky Flats Plant. The control reservoirs are believed to contain radioactive fallout products from

atmospheric testing only.

The peak $^{239,240}Pu$ generally attributed to weapons test fallout in the Colorado region is approximately 0.1 pCi/g. The principle objective of this study was to determine the concentrations of $^{239,240}Pu$ and ^{137}Cs residing in lake sediments as a result of atmospheric weapons testing, i.e. the background concentrations. These values were then compared with those determined by others for Standley Lake and Great Western Reservoir. The study was designed to provide a more accurate quantitative impact on the surrounding watershed by activities at the Rocky Flats Plant. The measurements were also used to yield information concerning sedimentation rates in Colorado Front Range lakes.

Chapter 2

Background

2.1 Plutonium Radiochemistry

Isotopes of plutonium were first isolated and studied by teams headed by Glenn Seaborg in 1940. The first isotope, Pu^{238},was produced by the bombardment of uranium by deuterons. Identification of isotopes 232 through 246 have since been noted (Seaborg, 1958). The main isotope of interest in nuclear power and weaponry due to its fissionability by slow neutrons (due to its high cross section) was determined to be ^{239}Pu. Production of this isotope was then focused upon in 1943.

Cyclotron produced ^{239}Pu became available in kilogram quantities by early 1944. Production of the 239 isotope involves the capture of a neutron by the ^{238}U isotope as follows;

$$^{238}U + n\ (fast) \longrightarrow {}^{239}U \longrightarrow {}^{239}Np \longrightarrow {}^{239}Pu$$

The formation of ^{239}U ($t_{1/2}$ = 23.5 min) and the subsequent beta decay to ^{239}Np ($t_{1/2}$ = 2.33 days) and ^{239}Pu occurs through another beta decay. Fission of ^{235}U produces the neutrons necessary to complete the reaction:

$^{235}U + n \rightarrow$ fission products + 2.5 n + 200 Mev

Because a fission reaction is used to generate the neutrons necessary to create plutonium, a host of highly radioactive elements accompanies the reaction. These elements (eg. ^{103}Ru, ^{95}Zr, ^{137}Cs, ^{141}Cs) are removed through the process termed "decontamination". Seaborg (1958) describes several techniques available for the decontamination process (precipitation, solvent extraction and ion exchange processes) that are beyond the scope of this review.

The end product, ^{239}Pu, emits alpha particles of energies 5.147,5.134 MeV plus others ranging in energy from 5.10 to 4.66 MeV. The daughter product of ^{239}Pu is ^{235}U, which will then follow the Actinium Series (4n + 3) decay.

The 239 isotope has a half-life of 24,360 years (see Figure 2.1). Due to this extremely long half-life, over 99.8% of the ^{239}Pu created since 1945 is still in existence as illustrated by:

$$N = N_o e^{-kt} \qquad \text{(eq.1)}$$

where:

N = the amount of the isotope remaining after time t, in percent.

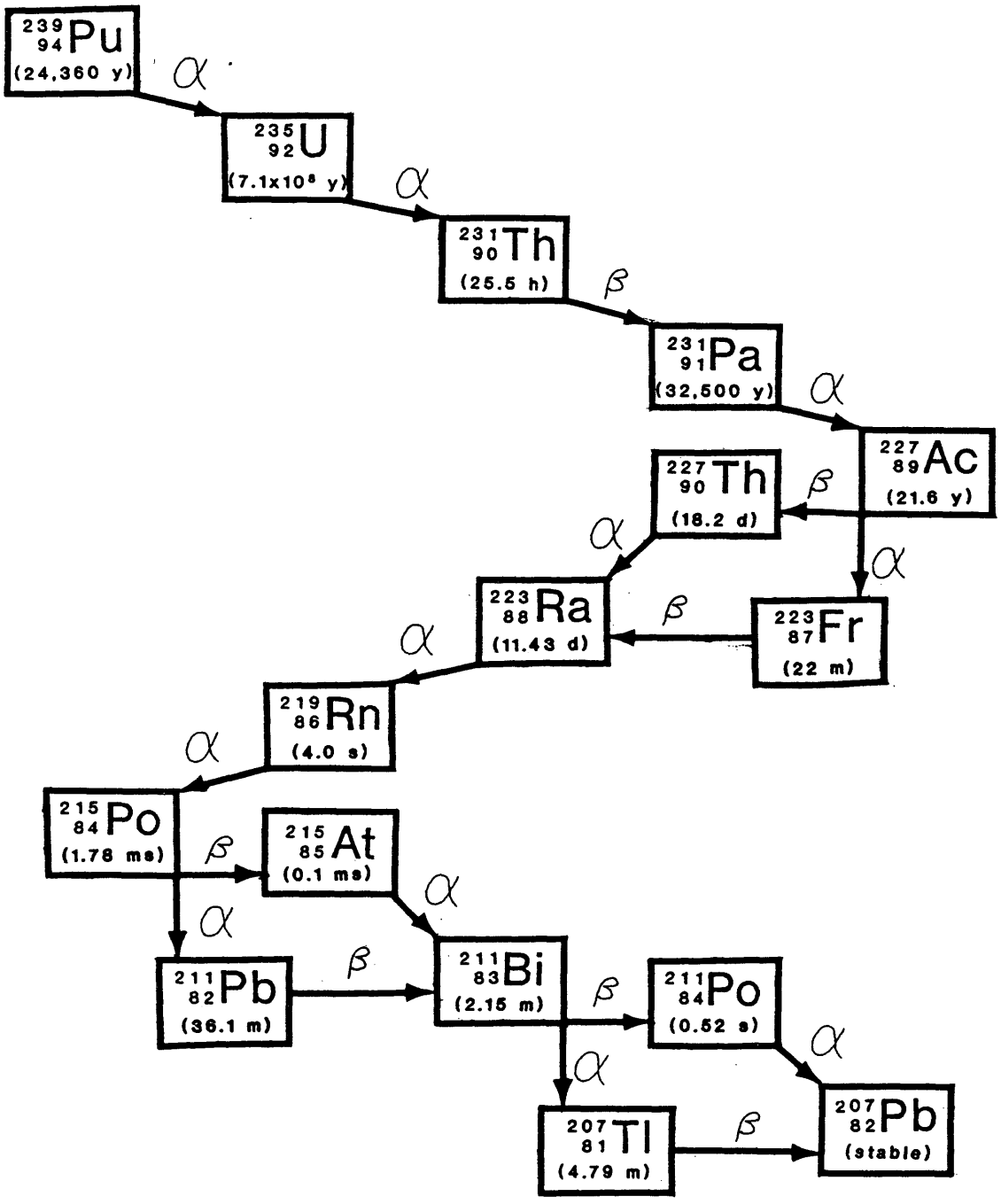

Figure 2.1 ^{239}Pu decay scheme.

(source: Dept. of Health, Education and Welfare, 1970)

N_o = the original amount of the isotope in percent,

k = the decay constant determined by $0.693/t_{1/2}$, and

t = the time since quantity N_o existed.

^{240}Pu is formed through neutron capture by a ^{239}Pu atom. This isotope also emits alpha particles at energies of 5.159, 5.115 MeV and others ranging from 5.01 to 4.85 MeV. The alpha particle energies of both the 239 and 240 isotopes are so similar that the two isotopes cannot be distinguished easily using alpha counting techniques. For this reason, the isotope activities are reported together.

^{240}Pu has a half-life of 6580 years and is considered to be a minor component of the total plutonium created for use in the defense industry or reactor fuels (see Figure 2.2). Due to the long half-life of this isotope, over 99.5% of all ^{240}Pu created since 1945 remains undecayed (calculated using eq. 1).

2.2 Natural Plutonium

Minute quantities of plutonium have been observed in uranium deposits by Seaborg and Perlman (1942) and since then by several others. Studies of Canadian pitchblende and Colorado carnotite revealed small amounts of alpha activity due to the 239 isotope of plutonium. Plutonium contents

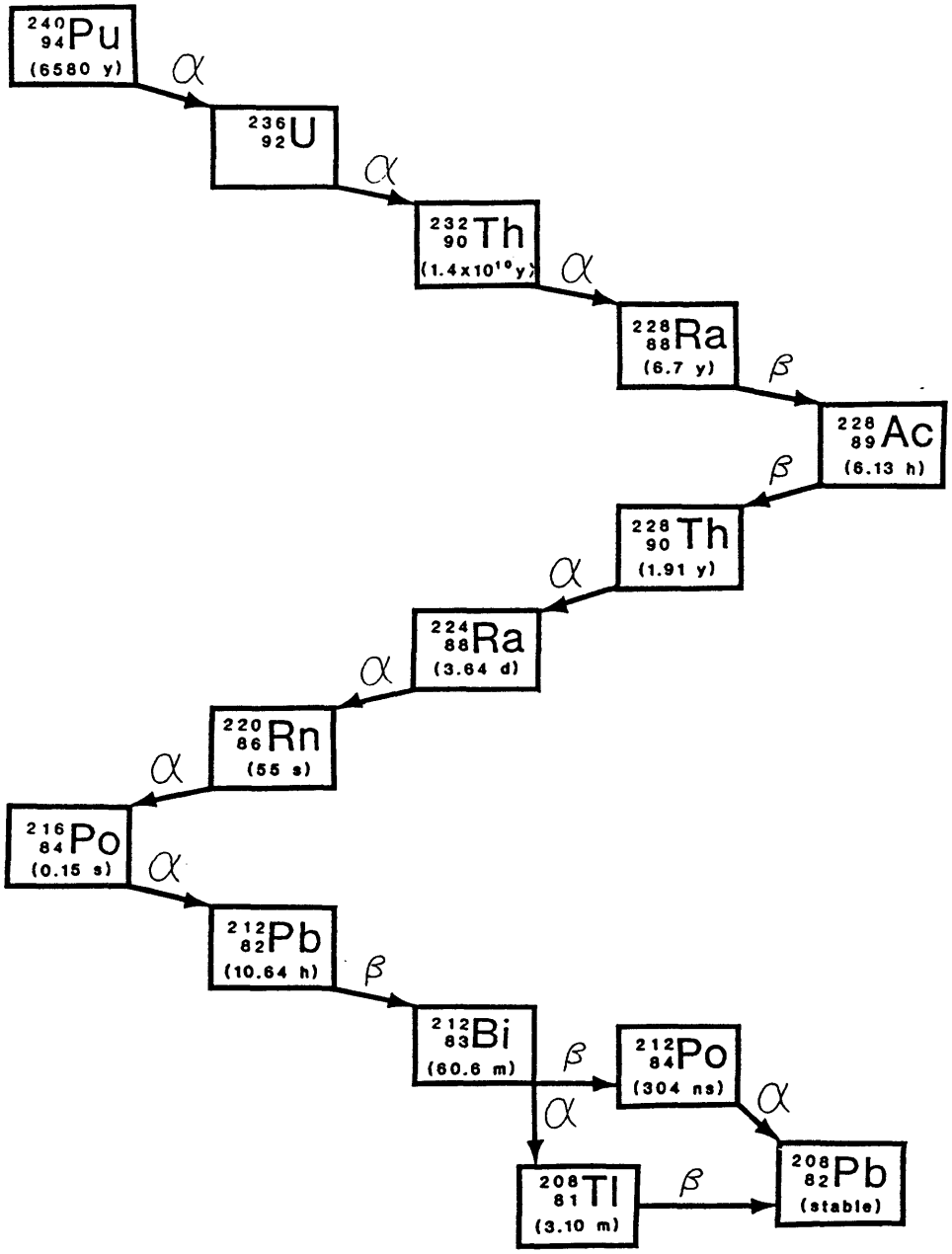

Figure 2.2. ^{240}Pu decay scheme.

(source: Dept. of Health, Education and Welfare, 1970)

have since been determined for several uranium and thorium ores that range from $< 1 \times 10^{-14}$ to 3.8×10^{-12} (ratio ^{239}Pu/ore by weight). It is believed that this plutonium is the result of the natural reaction:

$$^{238}U + n \longrightarrow {}^{239}U \longrightarrow {}^{239}Np \longrightarrow {}^{239}Pu$$

Sources for the neutrons necessary for this nuclear reaction to take place may be

a) spontaneous fission of ^{238}U,

b) neutron multiplication in ^{235}U,

c) reaction caused by alpha particles (from the decay of uranium) on lithium, boron, beryllium, fluorine, oxygen, silicon and magnesium, or

d) cosmic rays.

Of the sources listed, the most important appears to be neutrons produced in alpha particle reactions on lighter elements, however the others (with the exception of cosmic sources) do play a minor role. Regardless of the neutron source, the quantity of plutonium present in a natural ore body is dependent on the capture of the neutron by an atom of ^{238}U. Competing reactions consist mainly of neutron capture by elements of higher cross section (e.g. potassium, vanadium, tantalum). For this reason, plutonium quantities

in ores containing considerable quantities of competing elements will tend to be low. In all cases, however, since plutonium quantities, which appear to be ^{239}Pu exclusively, are of little importance outside the ore body itself (Seaborg, 1958).

2.3 Plutonium Chemistry

Seaborg (1958) indicates that the stable oxide of plutonium is the dioxide form PuO_2, and that ignition of practically any compound of plutonium in air will result in the formation of PuO_2. It can be expected then, that the plutonium released into the atmosphere during a test detonation will be of this form. Following initial injection into the atmosphere, the plutonium may oxidize to a higher oxidation state (i.e. Pu(V)). Fukai, et al. (1981) indicates that Pu(V+VI) are the primary species in rainwater.

As with other actinide elements, plutonium exhibits a wide range of oxidation states. Oxidation states range from III - VII and are able to coexist in an equilibrium state. In general, the lower oxidation states are stable at lower pH and the higher oxidation states tend to become more stable as alkalinity increases. Although states III - VI can exist in natural systems, because of the relatively

narrow band of pH values observed in the environment, the oxidation states most important to this study are Pu(IV and V). The VI and VII states are achieved in solutions through the presence of oxidizing agents such as bromate or persulfate. Pu(IV) is stable primarily at the lower end of the pH spectrum and, as the pH of the aqueous solution becomes increasingly more alkaline, the plutonium is removed from solution by adsorption to suspended particles and through plateout onto stationary objects. The oxidation state most observed in solutions of environmental interest is the Pu(V) state. Usually occurring as PuO_2^+, this oxidation state is the most stable under neutral conditions (i.e. approximately pH 7). Studies indicate that the presence of PuO_2^+ in solution is most likely if interaction between two PuO_2^+ molecules is not allowed to occur (i.e. at very low concentrations). For this reason, the concentrations of the oxide form of plutonium in natural solutions tend to be very low (10^{-13} - 10^{-17} moles per liter).

The equilibrium distribution coefficient (K_d) of plutonium in freshwater systems describes the concentration of Pu in solution versus that in the sediment and are typically on the order of 10^4. K_d can be calculated by:

$$K_d = [\text{Pu on sediment}]/[\text{Pu in water}]$$

Example K_d values are; Lake Michigan.....33 x 10^4 ml/g

Hudson River.....6.7 x 10^4 ml/g

Savannah River.....4.2 x 10^4 ml/g

These values illustrate the amount of plutonium in the sediment compared to the solution state (Watters, 1983).

Difficulties in this simple model arise when the matter of complexation arises. Experiments by Watters (1983), revealed that plutonium sorption onto sediments is an equilibrium process modified by changes in plutonium oxidation state and quantities of dissolved organic carbon. As may be expected, lakes with higher oxidation-reduction potential contain higher relative concentrations of the more oxidized plutonium states [Pu(V+VI)]. Watters' study also revealed that the oxidation state found in solution (associated with dissolved organic carbon) is primarily the Pu(IV) state. Complexing strengths of the various oxidation states associated with the same ligand are as follows:

$$Pu^{+4} > PuO_2^{+2} > Pu^{+3} > PuO_2^{+}$$

The concentration of plutonium in systems with low biological activity i.e. low oxygen consumption by respiration, can therefore be expected to have lower

quantities of plutonium in solution than those with high biological activity. Eutrophic conditions, alternatively, will tend to have higher biological activity, lower oxygen content in deeper waters and higher dissolved organic carbon. This will cause an increase in the complexation of the Pu(IV) state and a corresponding solution of plutonium into the water column. In conditions of depleted dissolved oxygen one would expect a corresponding increase in the Pu(IV) concentration (due to lower pH and eH) and a resulting selective solution of PuO_2 at the sediment water interface.

2.4 Sources of Background $^{239,240}Pu$

Although minute quantities of natural plutonium have been noted, background $^{239,240}Pu$ in the environment is considered to be entirely a result of human activity. Due to the worldwide nature of the distribution of $^{239,240}Pu$ caused by atmospheric nuclear weapons testing during the period 1945 - 1963, the associated fallout is now referred to as the background concentration.

Unfissioned $^{239,240}Pu$ used to create the critical mass necessary for the chain reaction fission process is the primary source of the 239 isotope in the environment. Creation of ^{239}U and subsequent decay to ^{239}Pu acts as a

secondary source (as shown in previous decay scheme). Although efficiencies have increased with technology, estimates of early atomic weapons reveal that as much as 90% of the plutonium mass remained unfissioned after detonation (Mahara, 1988). Modern, fission-only detonations (nonthermonuclear) have reached a fission efficiency of 35% or greater (Lapp and Andrews, 1972).

2.5 Cesium Radiochemistry

Twenty-two isotopes of cesium have been identified at present. Ranging from masses of 129 through 148, a majority of cesium isotopes have very short half-lives (on the order of days or seconds). Swan et al. (1981), identifies both the 134 and the 137 isotopes as fission products of nuclear processes. ^{134}Cs, is formed in minute quantities during an atomic detonation and is of little importance in the environment except in areas impacted by uranium or plutonium related activities. ^{137}Cs, however is a major component of uranium and plutonium fission (Swan, 1981). Kathren (1984) indicates that that approximately 6.2% of all radioactive fission products are ^{137}Cs.

With a relatively short half-life ($t_{1/2}$ = 30.23 y), ^{137}Cs is useful in sediment dating and calculation of sedimentation rates in lakes and reservoirs. ^{137}Cs decay

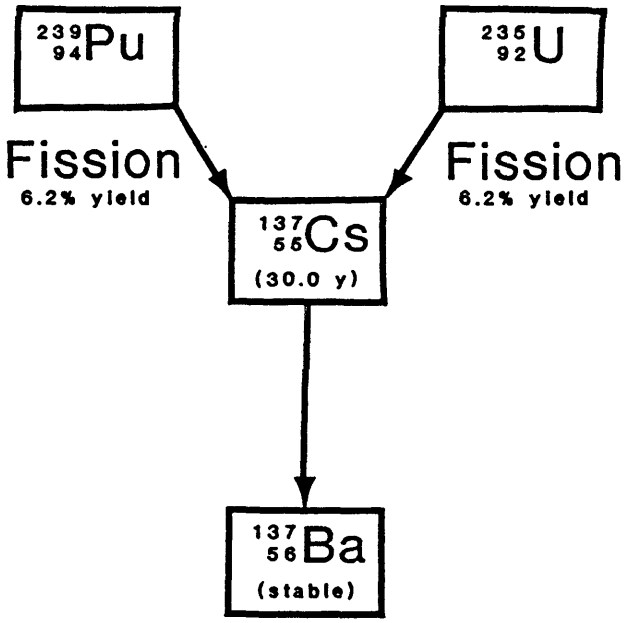

Figure 2.3 ^{137}Cs decay scheme.

(source: Dept. of Health, Education and Welfare, 1970)

scheme is given in Figure 2.3. ^{137}Cs will emit beta particles with energies averaging 0.195 MeV and a maximum of 1.167 MeV. Gamma energies average 0.662 MeV (U.S. Department of Health, Education and Welfare, 1970).

2.6 Cesium Chemistry

As a group IA element on the periodic table, cesium can be expected to be chemically similar to potassium in natural systems. Swan (1981), discusses the solvation of monovalent cesium as the primary occurrence of the metal in aqueous solutions. Due to the presence of the Cs^{+1} state, solutions of cesium tend to be relatively inactive unless the quantity of dissolved solids is high (as in most reservoir systems). Swan also identifies ion exchange to particulate matter as the primary removal mechanism of cesium from solution in natural systems. Edgington and Robbins (1975) support this claim finding that 95% of all cesium entering Lake Michigan eventually enters the sediment. Garten (1975) further supports this claim by identifying the sediments as the primary repository for ^{137}Cs. Cahill and Steele (1986) identify adsorption and ion exchange as the dominant mechanisms for cesium removal from solution. Garten (1975) continues with the observation that illite clays tend to fix the cesium into immobile forms, therefore removing it much

more effectively from the water column than kaolinitic clays which tend to bind the cesium more loosely. Kiser (1979) indicates that the equilibrium distribution coefficient (K_d) for ^{137}Cs is dependent on the pH of the solution and the concentration of ^{137}Cs in the solution. The maximum K_d observed in experiments by Kiser occurred at pH = 8 and the K_d increases with increasing ^{137}Cs concentration of the solution. Sibley (1982) found that within the pH range of environmental interest, cesium adsorption is not affected (remains at maximum). That is, in most systems the cesium is removed to the sediments via adsorption almost immediately. Distribution values residing within the sediment are typically 93 - 95% of the total ^{137}Cs within a system (Cahill and Steele, 1986).

Postdepositional migration of cesium (in the vertical direction) is limited to bioturbation, physical mixing processes (discussed later) and molecular diffusion. Diffusion of cesium in the sediments is a factor of the molecular diffusion coefficient (for ^{137}Cs = 1.78×10^{-5} cm^2s^{-1}), time, porosity and sediment properties (e.g. binding properties of the sediment constituents)(Torgerson, et al 1984).

2.7 Sources of Background ^{137}Cs

Although nearly 200 radioactive species have been observed as products of nuclear fissions (primarily fissions of ^{235}U and ^{239}Pu), only ^{137}Cs is of importance in this study. ^{137}Cs does not occur naturally and is considered to be unique to fission products. According to Kathren (1984), ^{137}Cs constitutes 6.2% of the total fission yield. For this reason, worldwide distribution of ^{137}Cs by atmospheric testing has resulted in an artificially created background similar to that of background $^{239,240}Pu$.

It is important to note that ^{137}Cs is an abundant component of discharges made during accidents at nuclear power facilities such as Chernobyl in 1986 and Three Mile Island in 1979. Minute quantities of the ^{137}Cs released from these events may cause confusion in the quantification of more recent sedimentary ^{137}Cs.

2.8 Atmospheric Testing

It has been estimated that 360,000 Ci of $^{239,240}Pu$ (Perkins et al., 1980) and 34 MCi of ^{137}Cs (Gilat, 1975) have been injected into the atmosphere by nuclear weapons testing. Although the majority of atmospheric testing ended with the 1963 test ban treaty involving the United States, Soviet Union and Great Britain, atmospheric testing by both

France and China continued until the present. Appendix 7 shows periods of nuclear testing and associated influxes of $^{239,240}Pu$ and ^{137}Cs to the environment. The testing peaks occurring in the years 1958-59 and 1960-63 can be expected to coincide with peak background plutonium and cesium levels. It can be seen from the test summary (Appendix 1; Perkins and Thomas, 1980) that the peak in testing by the United States occurred in the 1957-58 period. Nearly all of the testing carried out in the 1961-62 period was carried out by the Soviet Union. Although the exact yield of the tests carried out by the Soviets in the early sixties is unknown, it is widely believed that the 1961-62 period constituted the maximum $^{239,240}Pu$ and ^{137}Cs flux to the atmosphere. For this reason, dating techniques involving these two substances are based on the peak concentration occurrence circa 1963. It is important to note that the Soviet Union conducted tests at a much higher latitude (north of 60° N) than the U.S. (mainly at 11° N) and, although one might expect that a majority of the fallout received in the Colorado region was a result of the Soviet tests, studies by Machta, et al. (1956) revealed that detonations made by the United States (test names Mike and Bravo) at Eniwetok atoll resulted in fallout levels on the order of 100 millicuries in the continental U.S. Fallout

levels observed following the test ban treaty can be attributed to either long-term residence of the radionuclides in the stratosphere or, more importantly, continued atmospheric testing by the French and Chinese. A majority of recent fallout in the United States is probably a result of the Chinese tests that were undertaken in the northern hemisphere (Perkins and Thomas, 1980). The French tests have been carried out primarily in the southern hemisphere. The presence of convection cells within the troposphere, and particularly the zone of convergence in the equatorial region causing upward movement of air masses (and therefore movement toward the poles at high altitude), cause little mixing of contaminants between the northern and southern hemisphere (Kathren, 1984).

2.9 Transport

Plutonium compounds, generally present as oxide vapors, are injected into the atmosphere at high temperature immediately following an atomic test. These rising vapors condense as they cool adiabatically and as diffusion with cooler air takes place. Cesium compounds are expected to behave similarly to plutonium in the atmosphere.

Testing low-yield weapons at low altitudes will result in relatively short-range deposition, as the radionuclides

will be injected primarily into the troposphere. Perkins and Thomas (1980) discuss the negligible introduction of material into the stratosphere prior to 1952 when the first thermonuclear device was tested. Residence times of aerosols and particulates within the troposphere are on the order of weeks and fallout is generally limited to several hundred miles downwind of the source.

High-yield or high-altitude detonations will generally result in significant injection of materials into the stratosphere. Perkins also discussed the increasing percentage of materials injected into the stratosphere with increasing yield. Submicron aerosols have negligible settling velocities within the stratosphere, and the mixing component across the tropopause is minimal at most times of the year. Current hypotheses concerning the transfer of submicron particulates across the tropopause include:

1) decrease in temperature differential across the tropopause during local winters allow for increased mixing,

2) diffusion across the tropopause due to concentration gradient of the gas in question.

3) folding of the tropopause causing isolated "islands" of stratospheric gases within the troposphere.

Larger particles, formed by coagulation of smaller,

submicron-sized aerosols, possess settling velocities that enable direct precipitation through the tropopause.

Once within the tropopause, processes which govern fallout of radionuclides include:

1) rainout, the process by which particles enter rain water by processes within the cloud itself,

2) washout, in which particles are scavenged by droplets of falling precipitation, and

3) dry fallout, of which impaction upon solid surfaces is most important, as settling according to Stoke's Law of Settling Velocity yields extremely long residence times for a majority of these particles.

It is accepted that rainout is the most important of these processes due to the minimal particle size of the species in question.

2.10 Behavior of $^{239,240}Pu$ and ^{137}Cs in Aquatic Systems

Plutonium dissolved in rainwater is usually of the oxidation state Pu(V), while cesium is of the Cs^{+1} state. In the simplest model, plutonium and cesium in solution as precipitation are adsorbed to soil particles in the case of overland flow, or to suspended particulate matter in the case of direct precipitation onto a water body. Moving water carries the particulates until stagnation occurs

causing the radionuclides to be deposited onto the sediment bed of the receiving water body. Most of the plutonium and cesium is removed from the water column in this fashion (Allard, 1983).

Brunskill, et al. (1984), discuss the variance in removal efficiencies of sediments within the same year. Changes in oxidation potential (i.e. seasonal anoxia) will affect the adsorption ability of contaminants to the sediments. It appears that the sediments are able to adsorb higher cesium quantities during the high oxygen periods (i.e. periods of overturn).

Adsorption, a surface phenomenon, is dependent on available surface area. It follows then, that the finest sediments in suspension would possess the greatest adsorption properties. It can be expected therefore, that the highest plutonium and cesium concentrations will be associated with the finest sediments. Sly (1978), supports this observation by pointing out that in the finest materials, sorption and cation exchange are particularly important on particulate matter with high surface areas and that a majority of inorganic colloidal materials have high cation exchange capacities. Also noted is the importance of small size- fraction clays to the selective transport and accumulation of heavy metals in lake systems. Brisbin,

et.al. (1974) also supports this observation, finding that the fine sediments average 170.47 pCi/g ^{137}Cs, while the coarser sediments average only 18.10 pCi/g ^{137}Cs. Alberts (1979), disagrees with this generally accepted view, finding that a plot of the activity of an isotope per unit mass against the total mass within a particle size class generates a straight line with a slope that is not significantly different than zero. This relationship between mass (of particle size class) and particle size is not independent of grain size however. Alberts does concede that in many cases, the simple relationship between grain size and isotope activity is observed.

Spatial relationships of particle size within the reservoir bed are related to the physical characteristics of the reservoir itself. Bruk (1985) listed three factors important in the migration of sediments after deposition: wind, climate, and river flow.

River flow is probably responsible for a majority of the sediment transport to the reservoir itself. Due to the decrease in water velocity and the accompanying decrease in competence of the tributary as it enters the reservoir, the coarser bed materials can be expected to deposit in the delta region. Progressively finer materials will be carried further out into the reservoir. Aggradation in the delta

region can be expected to be composed of primarily the coarsest materials. Resuspension of the fine materials, which may happen to exist in this region, will occur due to wave action and degradation of the delta during low retention times.

Wind-caused wave erosion is important to sediment migration primarily in near-shore locations. Orbital wave energy decreases by one-half for every one-ninth of the wave length (Bruk, 1985). For this reason, only shallow regions can be expected to be affected by wave energy. In order for erosion to occur, induced shear stress from wave energy must exceed the critical shear stress of the bed material. Thomas (1972,73) therefore has found that sand-size particles exist primarily in shallow shoreline and nearshore areas. Presumably, the wave energy generated in lakes of the size of this study would be small enough to resuspend only the fines from the shallow regions. This would allow for movement of fine sediment away from the shallow regions toward deeper areas.

Climate will effect the sedimentation distribution through the presence of thermal stratifications. In temperate regions, as in Colorado, thermal stratification can be expected during the summer months (May - Sept.). Sediment entering the reservoir during these periods will be

distributed according to the density of the water (a function of temperature) and the density of the sediment load. Relatively warm water entering the reservoir may exhibit "overflow" or movement across the surface of the reservoir. Dispersion of the sediment plume will rely primarily upon wind energy causing the mixing of the plume within the epilimnion. "Interflow" occurs when the tributary water density is at or near the density of the reservoir water. This condition will result in the movement of the sediment plume above or below the thermocline depending upon the depth of the stratification. In situations where the sediment plume enters above the thermocline, the plume will become dispersed throughout the epilimnion through mixing by wind energy and diffusion. Larger sediments may, at this point, possess rapid enough settling velocities to cross the thermocline and settle to the reservoir bed, however, a large quantity of fines can be held by the thermocline without significantly changing the density of the epilimnion (Sturm and Matter, 1978). The fines will then be transported to the hypolimnion during periods of isothermal conditions or overturn. "Underflow" describes the situation which occurs when the tributary water is significantly more dense than that of the accepting reservoir. High densities can be attributed to cold water

and/or high sediment loads. This condition results in movement of the sediment plume along the sediment/water interface. Sturm and Matter (1978) point out that these turbidity currents will often travel within current channels within the sediment surface. Deposition along these channels as the turbidity current slows is dependent on the settling velocity (a function of particle size and water viscosity i.e. Stokes Law of Settling) of the individual sediment particles.

Fine sediment may posses a settling velocity too slow for deposition to occur, however flocculation or aggradation may allow these particles to be removed from the water column. Bruk (1985) points out that in most reservoirs, fragments of minerals such as quartz, feldspar, mica, and clays form a major part of the suspensions. These mineral fragments often possess negative charges on the crystal faces and positive charges on the crystal edges. Attraction of these charges allows finer material to flocculate and settle to the sediment bed. Settling velocities often remain very low however, due to the "open" nature of the floc structure allowing for high water content within the floc itself.

The result of these major processes is an overall simplification of the reservoir basin morphology over time.

Depending on the original bathymetry at the time of impoundment and the sedimentation rate, the benthic topography can be expected to become increasingly more uniform in elevation above a given datum (Bruk, 1985), with decreasing particle size as distance from shoreline and (generally) as depth increases.

2.11 Bioturbation

Following deposition, significant redistribution of the upper layers of sediment may take place as a result of bottom-feeding organisms present within the reservoir. Bennett (1987) indicates the most important of this type of organism in Lake Michigan are species of midge larvae and oligochaete worm. These organisms feed by burrowing or moving sediment and removing nutrients from water which it filters through the sediment. The result of the physical mixing of the uppermost centimeters is an increase in the time of burial of the contaminant. In the cases of plutonium and cesium, bioturbation may result in a delay in the incorporation of these substances to the sediment bed. This effect may cause errors in the calculation of sedimentation rates as the sedimentation rates are based on the relative position of peak concentrations of these two components. Study of the benthic organisms in the locations

of study was beyond the scope of this review, although species of crayfish were observed at the Halligan Reservoir location.

Chapter 3

Research Methodology

3.1 Determination of Control Sample Locations

Two control locations for sampling were chosen on the basis of:

1) distance from the Rocky Flats Plant. Halligan Reservoir is located approximately 112 km northwest and Wellington Lake approximately 72 km southwest of the Rocky Flats Plant. Prevailing westerly wind directions in this region are expected to negate any effect that emissions from the Rocky Flats Plant may have on the control locations. Table 3.1 data and figure 3.1 illustrate the primarily western component of the winds at the Rocky Flats Plant,

2) age. Sediments must date prior to the onset of atomic weapons tests (i.e. circa 1945),

3) size and depth. For reasons of comparison both Halligan Reservoir and Wellington Lake were chosen for similarity in size and depth to Standley Lake and Great Western Reservoir,

4) adequate sedimentation. The absence of water diversion projects (i.e. conduit and engineered channels) as inflow sources for the two sample locations allows for natural sedimentation conditions to exist,

TABLE 3.1

Wind Direction Frequency (in Percent) for Four Wind Speed Categories at the Rocky Flats Plant 1985.
(Source: Rockwell International, 1985; after Paricio, 1987)

Dir.	Calm (m/s)	1-3 (m/s)	3-7 (m/s)	7-15 (m/s)	>15 (m/s)	Total
-	1.33	-	-	-	-	1.33
N	-	2.98	3.56	0.40	0.00	6.95
NNE	-	3.19	2.40	0.33	0.00	5.91
NE	-	2.79	1.15	0.02	0.00	3.95
ENE	-	2.45	0.62	0.00	0.00	3.07
E	-	2.66	0.54	0.00	0.00	3.20
ESE	-	2.57	1.45	0.01	0.00	4.02
SE	-	2.92	2.68	0.03	0.00	5.68
SSE	-	2.78	3.25	0.14	0.00	6.16
S	-	2.58	2.67	0.15	0.00	5.40
SSW	-	2.72	2.62	0.19	0.00	5.53
SW	-	2.36	2.52	0.20	0.00	5.08
WSW	-	2.85	4.87	0.81	0.00	8.53
W	-	3.46	3.16	1.79	0.26	8.67
WNW	-	3.13	3.94	4.59	0.58	12.24
NW	-	2.63	3.48	1.39	0.03	7.54
NNW	-	2.70	3.43	0.53	0.00	6.67
TOTALS	1.33	44.77	42.34	10.57	0.87	100.00

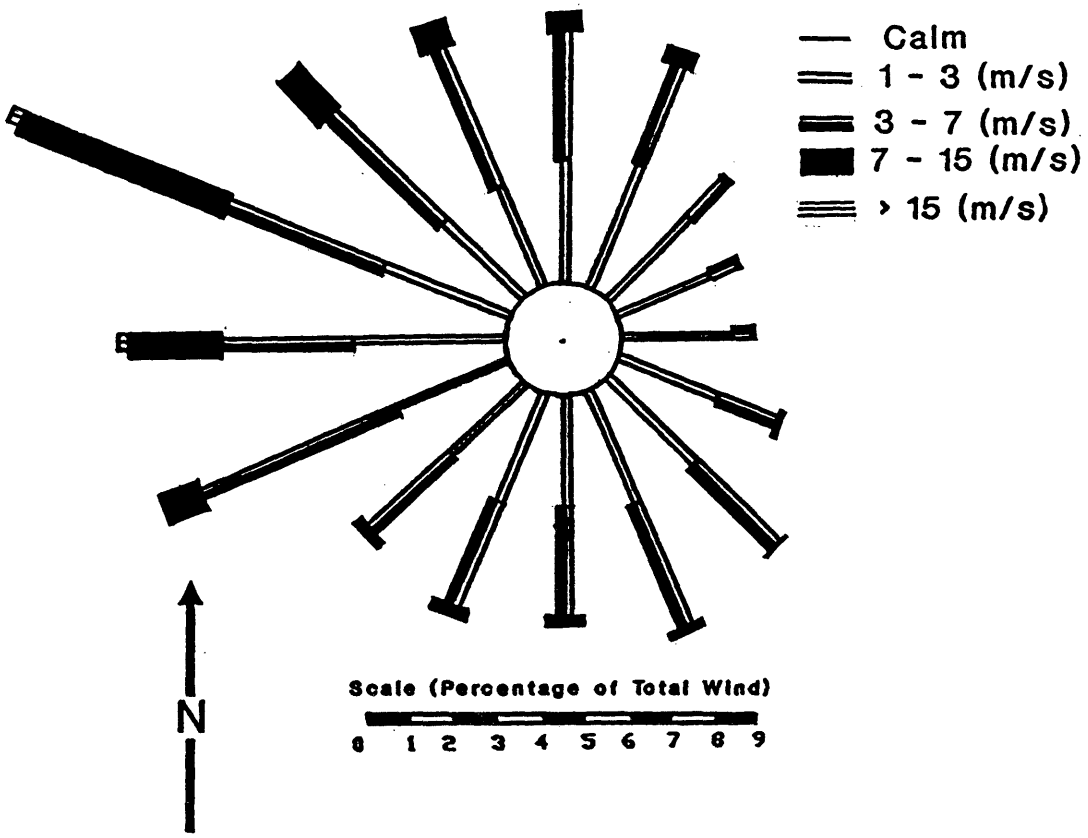

Figure 3.1 Wind Direction, Speed and Frequency.

Rocky Flats Plant (Source: Rockwell International, 1985).

5) similarity of climatic conditions. Average annual precipitation, as the most important component of fallout processes, should be similar to that of the Great Western Reservoir/Standley Lake area for purposes of comparison, and

6) minimum of human activity. Neither Halligan Reservoir or Wellington Lake have been dredged or significantly disturbed as indicated in recent records.

3.2 Site Descriptions

Standley Lake

Geographical Setting

Standley Lake is located within the city limits of Westminster, Colorado at approximately 39° 52' 30" N, 105° 07' 30" W. At maximum detention the lake surface elevation is 1678 m. (Arvada Quadrangle, Jefferson County, Colorado).

Geologic Setting

Bedrock in this region consists of the Laramie Formation which is made up of clay shale, sandy shale and sandstone (Spencer, 1961). Overlying the Laramie and probably the major influence on the sedimentation in Standley Lake are alluvial deposits of Quaternary age. This

alluvium varies in composition from sandy to clayey to sandy silt. Woman Creek, the major inflow to the reservoir, enters from the northwest and is underlain by the Piney Creek Alluvium consisting of micaceous sandy silt and a significant portion of humic silt (Van Horn, 1972).

Precipitation data in the region of both Standley Lake and Great Western Reservoir are limited to stations in Boulder, Brighton, Lakewood and Ralston Reservoir. Data from these stations are listed below.

Station	Elevation	Years of Data	Avg.Ann.Ppt
Boulder	1652 m	88	48.03 cm
Brighton	1518 m	9	33.02 cm
Lakewood	1718 m	74	37.97 cm
Ralston	1798 m	4	38.74 cm

Due to the orographically controlled precipitation patterns in this region of Colorado, the precipitation data given for these stations can be expected to approximate that of Standley Lake (i.e. circa 38 cm annually).

The Standley Lake dam was constructed circa 1902 and the initial detention took place shortly thereafter. The maximum detention is 1.248×10^8 cubic meters at a maximum

depth of 41 m (information obtained from State of Colorado Water Resources Division).

Great Western Reservoir

Geographical Setting

Located immediately south of Louisville, Colorado, Great Western Reservoir lies at an elevation of 1710 m and approximately 39° 53' N, 105° 09' W. (Louisville Quadrangle, Jefferson County, Colorado).

Geological Setting

The geology of the Great Western Reservoir region is very similar to that of Standley Lake, located approximately 5 km to the southeast. Precipitation in this region can be expected to be similar or equal to that of Standley Lake.

First retention in Great Western Reservoir occurred around 1904. The maximum retention of the reservoir is 1.122×10^7 cubic meters with a maximum depth of approximately 27 meters (Hydro-Triad Engineering, 1982).

Halligan Reservoir

Geographical setting.

Halligan Reservoir is located 25 miles northwest of Fort Collins, Colorado at approximetly 40° 52' 30" N, 105° 20' W (Figure 3.1). At maximum detention, the reservoir surface lies at an elevation of 1937 meters. (Virginia Dale Quadrangle, Larimer County, Colorado)

Geological setting.

The reservoir lies primarily within the Log Cabin batholith, forming the northern and western drainage basin. Members of the Trail Creek granite lie to the north and northeast. Localized metapegmatites exist in the area and may contribute significantly to the sediment influx to the reservoir. Major inflows to the reservoir are:

North Fork Cache La Poudre, which drains areas underlain primarily by fluvial deposits, however, the Log Cabin granite, quartz monzanite (associated with the Log Cabin event) and hornblende schists in the region also contribute to the sediment load,

Six Mile Creek, which drains regions to the north characterized by the Trail Creek granite and the sedimentary Fountain formation, and

Meadow Creek, which drains regions to the south and west characterized by the Log Cabin granite and hornblende gneisses.

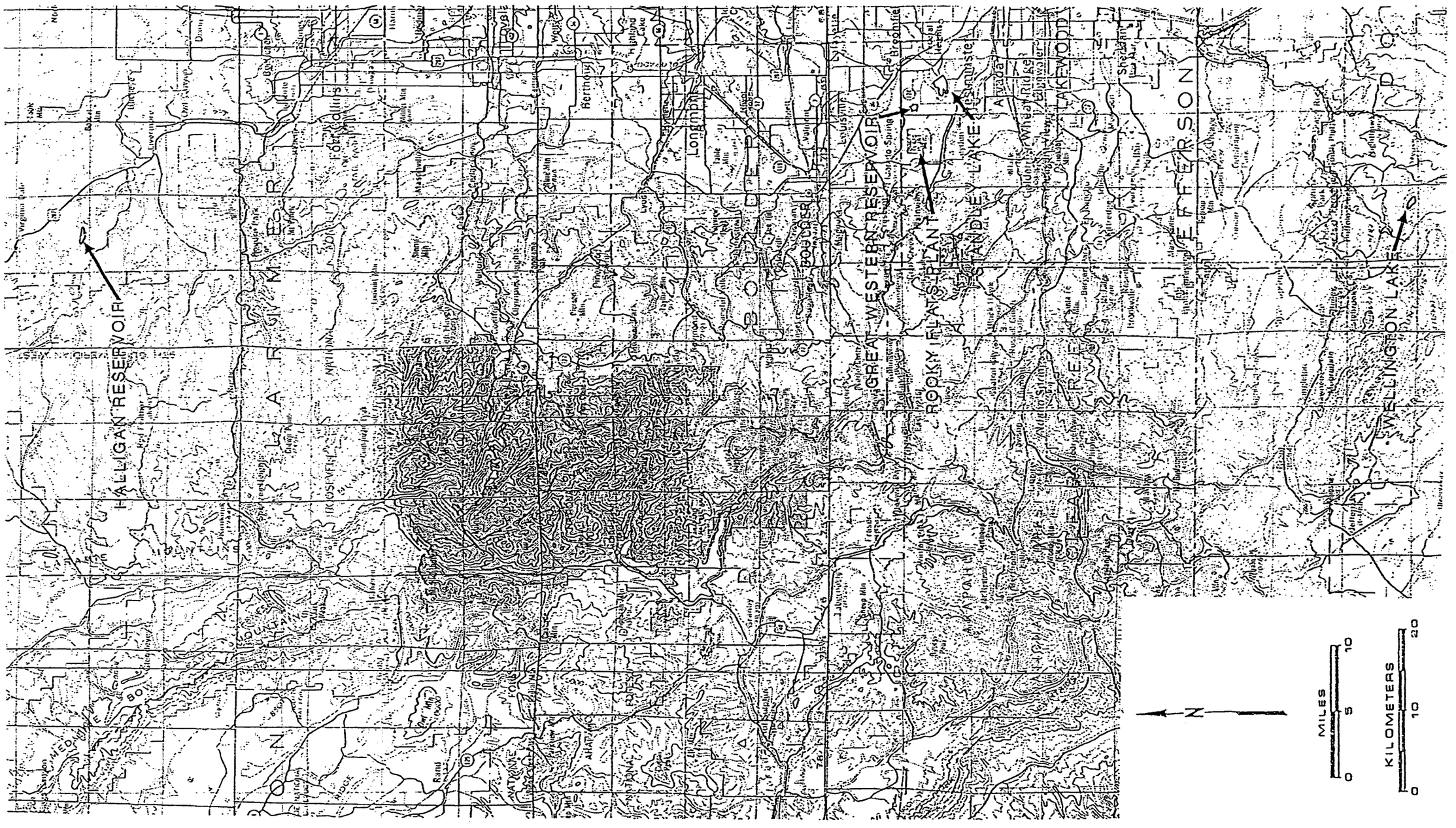

Figure 3.2 Location of Halligan Reservoir and Wellington Lake

Hydrology

Halligan Reservoir lies within the South Platte drainage basin immediately on the North Fork of the Cache La Poudre River. Meadow Creek is the only other non-intermittent tributary to the reservoir. Halligan Reservoir is owned and operated by the Phantom Canyon Ranch Water District. Water levels fluctuate significantly due to irrigation and drinking water needs throughout the year.

Precipitation data in the Halligan Reservoir region are limited to stations at Red Feather Lakes, Fort Collins and Waterdale. Data from these stations are listed below.

Station	Elevation	Years of Data	Avg.Ann.Ppt.
R.F. Lakes	2498 m	19	38.68 cm
Ft. Collins	1525 m	101	37.95 cm
Waterdale	1594 m	87	40.18 cm

The average annual precipitation at Halligan reservoir can then be expected to approximate the data given at these stations (circa 38 cm annually).

Age and dimensions.

Initial detention of water in Halligan Reservoir began in 1914. The reservoir has a maximum depth of approximately

21 m and a volume of 7.929 x 10^6 cubic meters at bankfull (information courtesy of Phantom Canyon Ranch).

Wellington Lake

Geographical setting

Wellington Lake is located 15 km southeast of Bailey, Colorado at approximately 39° 19' N, 105° 21'W (Figure 3.1). The lake surface lies at an elevation of 2448 m. at maximum retention. (Green Mountain Quadrangle, Jefferson County, Colorado)

Geological setting.

Wellington Lake lies in the region underlain by the northern member of the Pikes Peak Granite. This member is distinguished by high quartz and potassium feldspar contents. The major inflow to the lake is the North Fork Buffalo Creek which flows entirely within the region characterized by the Pikes Peak granite.

Hydrology

The tributaries to Wellington Lake consist of intermittent, seasonal streams. The North Fork of Buffalo Creek is the primary tributary. Wellington Lake is

currently owned by the City of Thornton, CO as a drinking water source, but levels are maintained relatively constant throughout the year.

Precipitation data for this region are limited to the weather collection stations at Bailey, Cheesman and Grant. Data from these stations are listed below.

Station	Elevation	Years of Data	Avg.Ann.Ppt.
Bailey	2354 m	41	42.21 cm
Cheesman	2095 m	79	41.83 cm
Grant	2642 m	19	41.12 cm

The average annual precipitation at Wellington lake can then be expected to be approximately 42 cm (All precipitation data from U.S. Department of Commerce, 1981).

Age and dimensions

Initial detention occurred circa 1900. The precise date is unknown. Maximum depth is approximately 11 m with a volume of 6.085×10^6 cubic meters at maximum detention (information courtesy of the City of Thornton and State of Colorado Water Resources Division).

Because precipitation is the major controlling factor in the deposition of airborne contaminants, Figure 3.3 is included to illustrate the similarity in average annual precipitation among the locations surveyed in this review.

3.3 Sampling Procedure

Determination of sampling locales were based upon previous studies (Setlock, 1983; Tutin, 1973; and others), which revealed that peak concentrations of radionuclides were found in the areas of greatest concentration of fines (i.e. deepest sections).

Low water levels (approximately 10 m below bankfull) at Halligan Reservoir allowed for sampling with hand-coring apparatus at a location very near the deepest section of the lake bed (Figure 3.4). Triplicate cores were taken to provide the mass necessary for gamma-ray determination of ^{137}Cs and ^{210}Pb for sediment dating purposes.

Sonar was used to determine the sampling locale at Wellington Lake (Figure 3.5). Triplicate cores were taken with a Kajak-Brinkhurst-type gravity coring device as well as a Davis-Doyle piston-type coring apparatus in what was determined to be the deepest section of the lake.

Difficulties arising from the sampling methods used in each situation included:

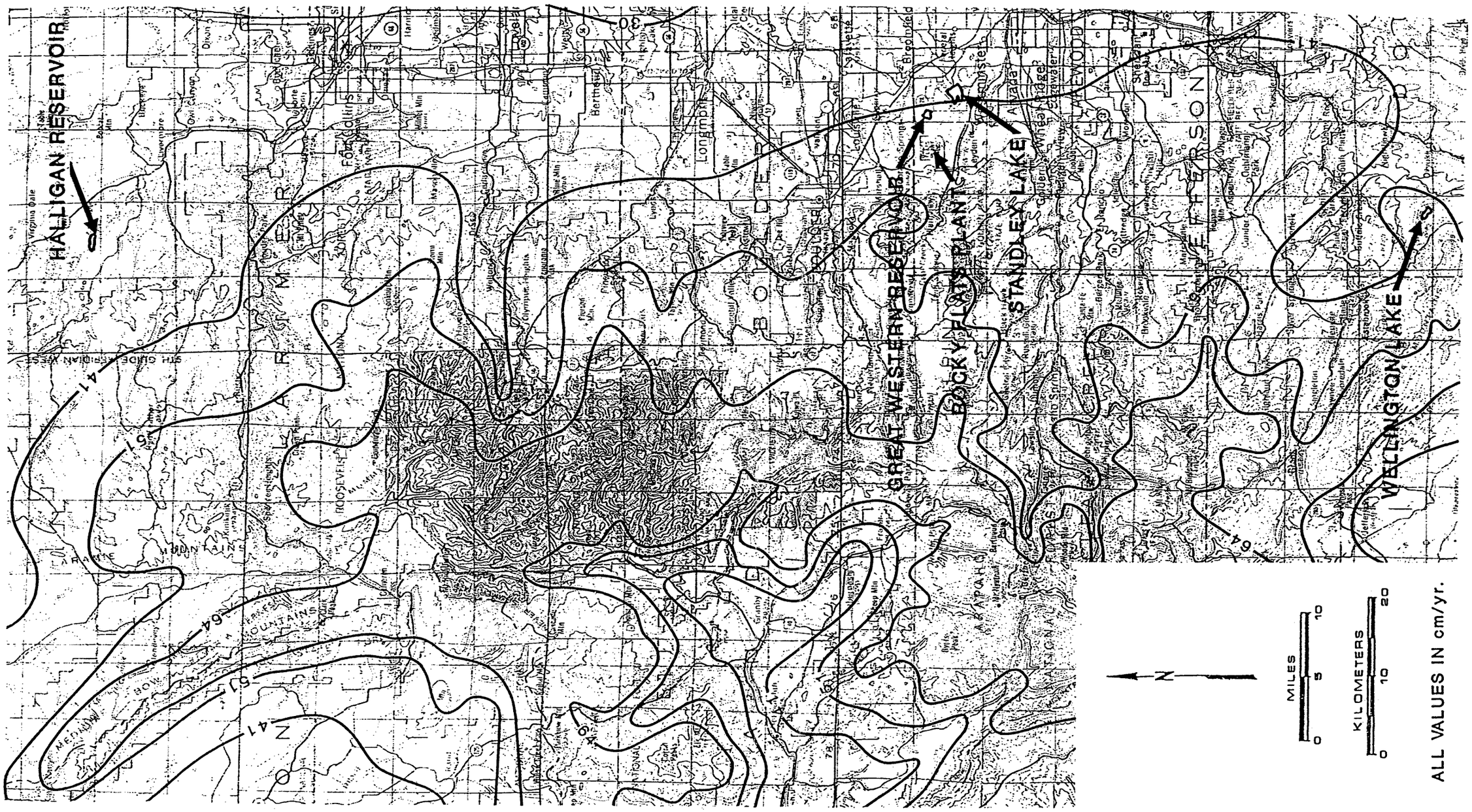

Figure 3.3 Isohyetals for the Colorado Front Range.

(source: Colorado Precipitation, 1980)

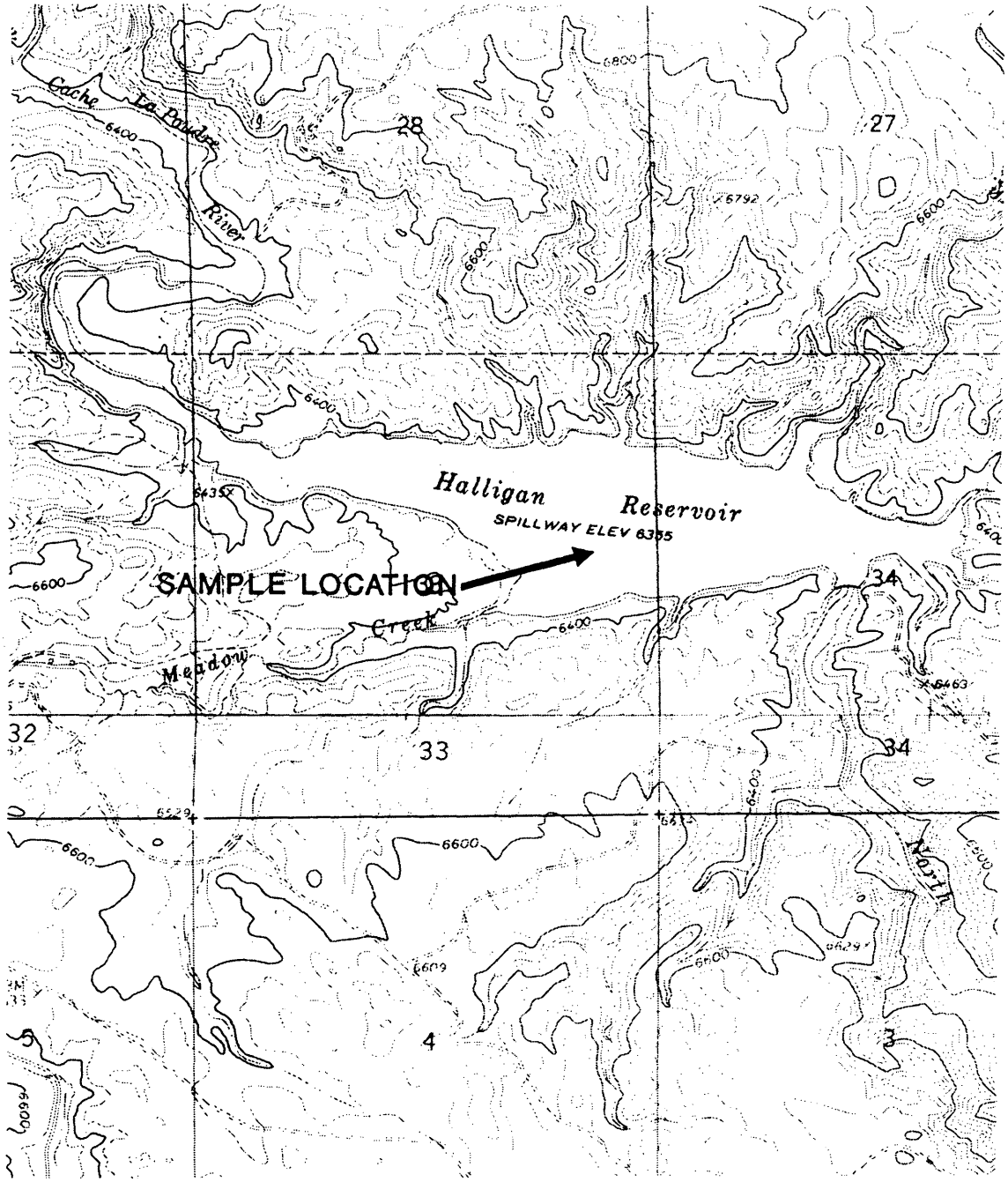

Figure 3.4. Sample location, Halligan Reservoir.

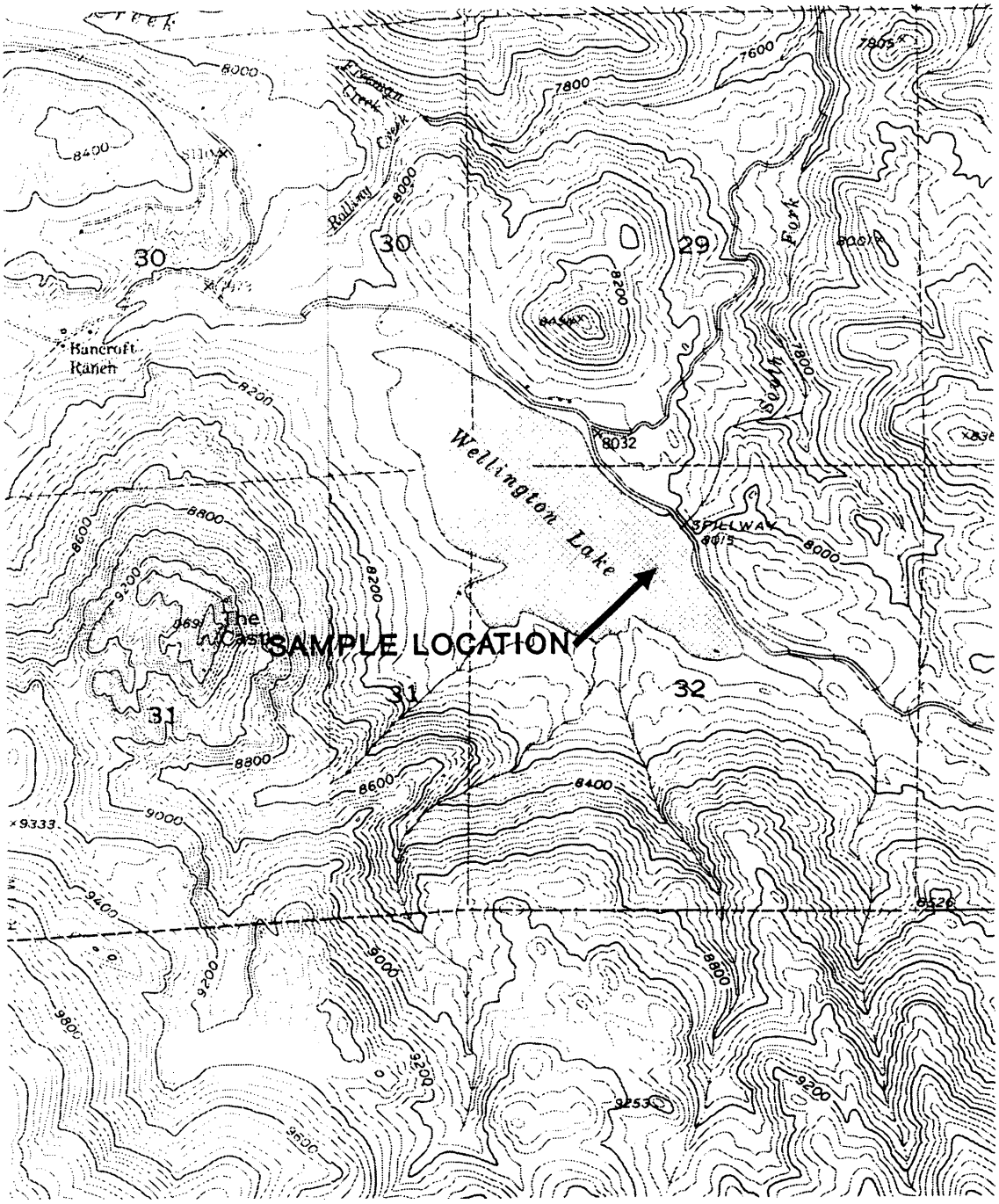

Figure 3.5 Sample location, Wellington Lake.

a) disturbance of the sediment/water interface by turbulence created by the gravity coring apparatus,

b) compaction of the sediment caused by the impact of the gravity coring device,

c) inadequate suction within the Davis-Doyle coring apparatus leading to segmentation of the sample core,

d) small sample volume of the Davis-Doyle coring device leading to greater counting error during analysis (see counting error calculation, i.e. sample volume),

e) possibility of increased bioturbation and wave energy mixing of surficial sections of the samples taken at Halligan Reservoir due to the low water levels observed at the time of sampling, and

f) possibility of unreported human activity on the sediment bed itself. Due to the high degree of fluctuation in the water level often observed in these reservoirs and the absence of reliable records concerning water levels in these reservoirs, it is impossible to determine what damage has been done to the sediment surface. It is possible that discontinuities exist within the sediment record resulting in inconsistent or erroneous conclusions drawn from the record.

3.4 Laboratory Procedures

All cores were maintained in a vertical position and all attempts were made to minimize movement following removal from the benthic environment. The Halligan Reservoir cores were sectioned within 24 hours of sampling. The Wellington Lake cores were frozen and maintained solid using CO_2 (s) and sectioned upon return to the lab facilities. Freezing was necessary in order to minimize biological activity which may have resulted due to extensive time periods between sampling and sectioning. The cores remained immobile for at least one hour prior to sectioning to allow for settling of the floc at the sediment water interface. All cores were extruded using a hand extruding device and sectioned at 1 cm increments.

The cores all appeared dark brown to gray to black in color. The sediment was generally fine to very fine in texture for all cores. There was no evident varving of the sediment. Organic matter (e.g. fine branches, twigs, etc.) was evident throughout much of all cores from both Halligan Reservoir and Wellington Lake. The sediment from Halligan Reservoir appeared to be homogeneous with respect to grain size throughout the length of the core. The sediment from Wellington Lake appeared to contain increasing amounts of organic matter with depth. The lower end of the core

appeared to be entirely organic.

Individual samples were placed on pyrex watchglasses and weighed immediately to 0.001 g using top-loading balances. Samples were then dried at 110°C. overnight. Immediately after removal from drying ovens, dry weights were determined as above for determination of porosity (Tables 3.1 and 3.2).

Samples from Halligan Reservoir were ground and incrementally homogenized (each of three corresponding replicate sections were combined prior to pulverization) using hand mortars and pestles. Samples from Wellington Lake were ground and incrementally homogenized using a rock pulverizer to approximately 200 mesh.

Preparation of samples for analysis at Acculabs Research included configuration into 11 g aliquots for gamma-ray spectrometry for ^{137}Cs determination (and sediment dating via ^{210}Pb).

Twelve blind duplicates were included for quality control and statistical analysis purposes.

Minimization of gamma ray attenuation was achieved through the use of polystyrene petri dishes as scintillation vials. The geometry achieved was that of a disk of 3 cm radius and approximately 0.5 cm in thickness.

Gamma-ray attenuation across the sample can then be

TABLE 3.2

Samples Submitted to Accu-Labs Research for ^{137}Cs, ^{210}Pb and $^{239,240}Pu$ Determination, Halligan Reservoir

Sample ID	Dry Weight (g)	Comment
HR01	11.002	
HR02	11.002	
HR03	11.004	
HR04	10.999	
HR05	10.999	
HR06	11.002	
HR07	11.002	
HR08	11.001	
HR09	10.999	
HR10	11.002	
HR11	11.000	
HR12	11.000	
HR13	11.000	
HR14	11.000	
HR15	11.000	
HR16	11.002	
HR17	11.001	
HR18	10.998	
HR19	11.001	
HR20	11.000	
HR21	11.002	
HR22	11.000	
HR23	11.000	
HR24	10.999	
HR25	11.001	
HR26	11.001	
HR27	11.001	
HR28	10.999	
HR29	11.001	
HR30	11.000	
HR31	11.001	
HR32	11.002	
HR33	10.999	

(CONTINUED)

TABLE 3.1

(CONTINUED)

Sample ID	Dry Weight (g)	Comment
HR34	11.001	
HR35	10.999	
HR36	10.998	
HR37	11.001	
HR38	10.997	DUPLICATE HR02
HR39	11.001	DUPLICATE HR06
HR40	11.000	DUPLICATE HR11
HR41	11.000	DUPLICATE HR14
HR42	11.000	DUPLICATE HR17
HR43	11.000	DUPLICATE HR20
HR44	11.000	DUPLICATE HR22
HR45	11.001	DUPLICATE HR25
HR46	11.001	DUPLICATE HR28

TABLE 3.3

Samples Submitted to Accu-Labs Research for ^{137}Cs, ^{210}Pb and $^{239,240}Pu$ Determination, Wellington Lake

Sample ID	Dry Weight (g)	Comment
WL01		
WL02		
WL03	10.900	
WL04	10.781	
WL05	11.001	
WL06	10.998	
WL07	10.999	
WL08	11.002	
WL09	10.999	
WL10	10.999	
WL11	11.002	
WL12	11.002	
WL13	11.001	
WL14	11.000	
WL15	11.000	
WL16	11.001	
WL17	11.000	
WL18	11.000	
WL19	11.000	
WL20	11.007	
WL21	11.003	
WL22	11.002	
WL23	11.003	
WL24	11.000	
WL25	11.004	
WL26	10.999	
WL27	9.307	DUPLICATE WL15
WL28	10.834	DUPLICATE WL16
WL29	11.000	DUPLICATE WL17

determined as:

$$N_1 = N_o\, e^{-kt} \qquad \text{(eq. 2)}$$

where: N_1 = number of gamma rays detected,

N_o = number of gamma rays generated by sample,

k = attenuation coefficient for ^{137}Cs (662 KeV) through soil (Dept. Health, Ed. and Welfare, 1970), and

t = thickness of sample normal to counting window surface.

Using the radius of the petri dish of approximately 1.5 cm and a sample depth of 0.5 cm, it was determined that an 11 g sample was appropriate in the geometry of the petri dish to achieve an areal density of 0.5 g/cm^2. The attenuation of gamma radiation due to absorption within the soil sample itself can then be calculated using an attenuation coefficient of 0.076 cm^2/g and $N_{(0)} = 1$, i.e. 100%:

$$0.96 = N_1 = e^{-(0.076)(0.5)}$$

The attenuation can then be assumed to be approximately 4% across the sample.

Gamma counts (of the energy 662 KeV) were determined using a reverse-electrode germanium diode detection apparatus in conjunction with a Canberra series 35

multi-channel spectrometer to define discrete gamma energies. Count times were carried out for 1000 minutes for all samples.

$^{239,240}Pu$ determinations were carried out using wet chemical separation followed by alpha spectrometry counting techniques. Following is a summary of laboratory procedures used by Accu-Labs Research.

Five g aliquots are weighed into a teflon beaker. 0.5 g ^{236}Pu is added as a tracer to each sample. Samples are digested to dryness twice with concentrated HNO_3 to remove organic material and HF to remove silicates. Samples are digested a third time with HNO_3 and boric acid to remove fluoride interference that may occur during anion exchange (Pu^{+4} + HF <-> PuF^{+3} + H^+ ; Seaborg, 1957). The samples are then redissolved in 8N HNO_3 containing 1% 1M $NaNO_2$.

100-200 mesh chloride-type anion exchange resins are prepared by washing with 250 ml HNO_3 and $NaNO_2$. The sample solutions are then passed through the anion exchange columns which are subsequently washed with additional 8N HNO_3. The columns are washed further with 9N HCl to remove thorium and the Pu is eluted with 1.2N HCl containing 2% H_2O_2. Plutonium is electroplated onto stainless steel disks for alpha-particle activity determination. The electroplating process deposits the remaining material in a geometry so

thin that attenuation of alpha particles across the sample is considered to be insignificant.

Alpha activity was determined using a Tennelec model TC256 counting apparatus in conjuction with a multi-channel spectrometer. Alpha energies of 5.16 and 5.17 MeV are counted together. No attempt to distinguish between the two energies was made. Due to the inability of the spectrometer to distinguish the alpha energies emitted by the isotopes 239 and 240, the two isotopes are quantified together. Final counts are determined by the following calculation:

$$\text{Final Result} = \frac{(\text{pCi/aliquot}) - (\text{pCi/blank})}{5\ \text{g}}$$

where:

pCi/aliquot = the total number of counts received from the sample,

pCi/blank = the total number or counts received from the reagent blank (^{236}Pu for the alpha determination, includes background alpha spectrum), and

5 g is the aliquot size (for plutonium determination).

3.5 Statistical Analysis

The inclusion of twelve blind duplicates allows for statistical determination of significance using the two-

tailed t-statistic test.

The t-statistic test is based on the initial hypothesis that the mean concentration value (for plutonium and cesium) of one population is not significantly different than the mean concentration value reported for the duplicate population. In order to test this hypothesis, it must be determined whether the variance between the two sample sets are equal. Determination of variance is based on the initial (null) hypothesis that the variance between one sample set is equal to the variance of the other ($V_1 / V_2 = 1$). The alternative hypothesis in this case would be that the absolute difference between the two sample variances is greater than expected if the population variances were equal. This hypothesis can be tested using the F test found in basic statistic literature. The F test is based on the probability of obtaining a specified F ratio, from a normal population, given a certain F ratio. Calculation of the F ratio is then compared with critical values for F specified in general statistics tables.

Not rejecting the null hypothesis indicates that there is no statistical evidence to suggest that the two duplicate samples were from different populations.

The t statistic can then be calculated using standard formulas for equal sample variance found in basic statistic

literature. Once the t statistic has been calculated, it is compared with critical values for t as a function of the degrees of freedom and levels of confidence in t statistic determination tables (Davis, 1973).

Rejection of the null hypothesis indicates that the concentration values reported in the duplicate test are statistically different than initial concentration values, however, acceptance of the null hypothesis indicates that there is not sufficient evidence to suggest that the two samples came from populations having different means in concentration. Results ofcalculations for determination of the t-statistic are given in Tables 4.6 and 4.7.

Assumptions that must be made in order to accurately use the t-test are:

1) both samples are selected at random;

2) the populations from which the the samples were drawn are normally distributed and;

3) variances of the two populations are equal.

3.6 Error Determination

All values reported for the concentration of $^{239,240}Pu$ and ^{137}Cs have an associated error (+/- value). This value is comprised only of the errors associated with counting and

analytical techniques as described below.

Total error can be defined as the analytical error plus the error in counting. Due to the random nature of radioactive disintegrations, an error exists associated with the counting of this activity. The standard deviation of the net counting rate will indicate the variability within these counts as calculated:

$$S_n = [(R_o / t_1) + (B / t_2)]^{1/2}$$

where:

R_o = gross count rate,

t_1 = counting time for the gross count,

B = background count rate, and

t_2 = counting time for the background count.

Gross count rate is defined as the net count rate plus the background count rate.

Counting error can then be determined using:

$$CE = 1.96\ S_n / [2.22\ (EVF)(e^{-kt})(P)]$$

where:

1.96 = 95% confidence factor,

2.22 = conversion factor from dpm to pCi,

E = efficiency factor, cpm/dpm,

V = volume of the aliquot analyzed in grams,

F = recovery factor,

e^{-kt} = decay constant used for tracer analysis, and

P = photon yield (used only in gamma determination)

(Source: Environmental Protection Agency, 1980)

The efficiency factor for the gamma count is determined through the analysis of the CUP-1 standard as:

Efficiency = dpm/cpm

where:

dpm = the number of disintegrations per minute calculated for the CUP-1 standard, and

cpm = the number of counts actually recorded by the gamma detector (for the CUP-1 standard).

The CUP-1 is a standardized uranium ore sample mixed with soil of similar density to that of the samples submitted. The standard was allowed to reach equilibrium (approximately 30 days) with respect to the ^{222}Rn daughters and then counted (^{214}Bi determination) for 1000 minutes.

Because the CUP-1 standard used to determine counting efficiency is of the same geometry as the samples analyzed,

the count rate observed compared with the count rate calculated for the CUP-1 will result in an integration of both the efficiency of the counting apparatus and the attenuation across the sample thickness.

Efficiency for alpha counts of plutonium activity is included with the chemical recovery factor. Recovery factor is the analytical loss of radioactive substances through the wet chemistry process. The analytical error for the ^{137}Cs determination is nonexistent as the non-destructive analysis technique was used. The recovery factor for $^{239,240}Pu$ was determined through the loss of the ^{236}Pu tracer during the wet chemistry phase of the analysis. Because the ^{236}Pu tracer is present throughout the entire process (both wet chemical and counting procedures) the expected number of counts (as calculated) can be compared with the observed counts and the result can be used as an integration of the recovery process, efficiency, and attenuation across the sample.

Radioactive decay is negligible for all samples analyzed due to the long half-lives associated with the components relative to the laboratory time. Only ^{236}Pu is of concern with respect to decay during the laboratory analysis due to its relatively short half-life (2.85 years).

Photon yield is a factor introduced to account for the

gamma activity lost to photon production. For ^{137}Cs the photon yield coefficient is 85.2%. That is, for every 1000 beta emissions there are 852 gamma emissions, the remainder are lost as photons.

Counting error can then be considered to be primarily a function of count time and efficiency of counting. Given a fixed efficiency of the counting apparatus, the variables become only aliquot geometry (to decrease attenuation) and count time. It becomes immediately clear that an increase in count time will result in a decrease in the counting error. For this reason, count times as long as economically feasible were used, i.e. 1000 minutes (information concerning calculation of error courtesy of Harry Summers, Accu-Labs Research).

Chapter 4

Results and Discussion

4.1 Data

The emphasis of this section is first, to report the background concentrations observed in both Halligan Reservoir and Wellington Lake; second, to compare these values with the previously obtained data from Great Western Reservoir and Standley Lake; and third, to compare sediment dates determined from ^{210}Pb analysis completed for the samples from Halligan Reservoir and Wellington Lake.

Results of the ^{137}Cs gamma and $^{239,240}Pu$ alpha determination for Halligan Reservoir and Wellington Lake are presented in Tables 4.1 and 4.2, respectively. Only one core from each location was analyzed by Accu-Labs Research. Lack of funding and lack of confidence (due to low mass and segmentation of core) in the Davis-Doyle core resulted in the omission of this sample from those submitted.

Immediate inspection yields respective ^{137}Cs and $^{239,240}Pu$ peaks at the same sediment interval in both cases. The Halligan Reservoir maximum occurred at the 21 - 22 cm depth interval and the Wellington Lake maximum occurred at the 10 - 11 cm depth interval. This correspondance in activity maxima between ^{137}Cs and $^{239,240}Pu$ in both cases indicated that

TABLE 4.1

Determination of $^{239,240}Pu$ and ^{137}Cs
Halligan Reservoir
(Analysis by Accu-Labs Research)

Depth Interval (cm)	$^{239,240}Pu$ (pCi/g)	^{137}Cs (pCi/g)
0 - 1	0.019 +/- 0.018	0.73 +/- 0.17
1 - 2	0.028 +/- 0.019	0.65 +/- 0.15
	0.021 +/- 0.021	0.76 +/- 0.13
2 - 3	0.024 +/- 0.019	0.63 +/- 0.15
3 - 4	0.018 +/- 0.018	0.72 +/- 0.14
4 - 5	0.028 +/- 0.019	0.70 +/- 0.17
5 - 6	0.046 +/- 0.020	0.86 +/- 0.18
	0.011 +/- 0.020	0.67 +/- 0.16
6 - 7	0.017 +/- 0.018	0.82 +/- 0.16
7 - 8	0.022 +/- 0.018	0.71 +/- 0.15
8 - 9	0.032 +/- 0.019	0.88 +/- 0.17
9 - 10	0.036 +/- 0.020	1.0 +/- 0.1
10 - 11[1]	0.033 +/- 0.019	1.1 +/- 0.2
	0.015 +/- 0.020	1.0 +/- 0.2
11 - 12	0.039 +/- 0.020	1.4 +/- 0.2
12 - 13	0.027 +/- 0.018	1.1 +/- 0.2
13 - 14[1]	0.038 +/- 0.019	1.2 +/- 0.2
	0.031 +/- 0.021	1.4 +/- 0.2
14 - 15	0.035 +/- 0.019	1.4 +/- 0.2
15 - 16	0.053 +/- 0.021	1.7 +/- 0.2
16 - 17[1]	0.040 +/- 0.019	1.9 +/- 0.2
	0.036 +/- 0.021	1.8 +/- 0.2
17 - 18	0.047 +/- 0.019	1.8 +/- 0.2
18 - 19	0.042 +/- 0.019	1.8 +/- 0.2
19 - 20[1]	0.066 +/- 0.021	2.2 +/- 0.2
	0.036 +/- 0.022	2.2 +/- 0.2
20 - 21	0.050 +/- 0.021	2.1 +/- 0.2
21 - 22[1]	0.050 +/- 0.020	2.3 +/- 0.2
	0.074 +/- 0.022	2.2 +/- 0.2
22 - 23	0.044 +/- 0.020	1.8 +/- 0.2
23 - 24	0.049 +/- 0.019	1.7 +/- 0.2
24 - 25[1]	0.049 +/- 0.021	1.5 +/- 0.2
	0.043 +/- 0.022	1.3 +/- 0.2

(CONTINUED)

TABLE 4.1

(CONTINUED)

Depth Interval (cm)	$^{238,240}Pu$	^{137}Cs
25 - 26	0.051 +/- 0.022	1.4 +/- 0.2
26 - 27	0.050 +/- 0.022	1.2 +/- 0.2
27 - 28[1]	0.026 +/- 0.021	0.69 +/- 0.16
	0.035 +/- 0.021	0.56 +/- 0.14
28 - 29	0.011 +/- 0.020	0.50 +/- 0.14
29 - 30	0.006 +/- 0.020	<0.10
30 - 31	-0.002 +/- 0.020	<0.11
31 - 32	-0.006 +/- 0.019	<0.11
32 - 33	0.001 +/- 0.020	<0.11
33 - 34	-0.002 +/- 0.020	<0.11
34 - 35	-0.005 +/- 0.019	<0.11
35 - 36	-0.003 +/- 0.020	<0.11
36 - 37	-0.002 +/- 0.020	<0.11

[1] Indicates duplicate sample analysis.

TABLE 4.2

Determination of $^{239,240}Pu$ and ^{137}Cs
Wellington Lake
(Analysis by Accu-Labs Research)

Depth Interval (cm)	$^{239,240}Pu$ (pCi/g)	^{137}Cs (pCi/g)
0 - 1	0.076 +/- 0.021	3.2 +/- 0.3
1 - 2	0.065 +/- 0.021	3.3 +/- 0.2
2 - 3	0.089 +/- 0.022	3.3 +/- 0.2
3 - 4	0.077 +/- 0.022	3.5 +/- 0.2
4 - 5	0.080 +/- 0.022	3.4 +/- 0.2
5 - 6	0.078 +/- 0.022	3.4 +/- 0.2
6 - 7	0.099 +/- 0.022	4.4 +/- 0.3
7 - 8	0.14 +/- 0.02	4.9 +/- 0.3
8 - 9	0.12 +/- 0.02	5.2 +/- 0.3
9 - 10	0.12 +/- 0.02	5.9 +/- 0.3
10 - 11	0.19 +/- 0.02	6.2 +/- 0.3
11 - 12	0.14 +/- 0.02	5.4 +/- 0.3
12 - 13	0.13 +/- 0.02	4.8 +/- 0.3
13 - 14	0.14 +/- 0.03	3.8 +/- 0.3
14 - 15[1]	0.089 +/- 0.022	2.7 +/- 0.2
	0.088 +/- 0.022	3.0 +/- 0.2
15 - 16[1]	0.048 +/- 0.020	1.0 +/- 0.2
	0.053 +/- 0.022	1.0 +/- 0.2
16 - 17[1]	0.014 +/- 0.019	0.37 +/- 0.12
	0.012 +/- 0.019	0.39 +/- 0.17
17 - 18	0.001 +/- 0.018	0.28 +/- 0.14
18 - 19	0.003 +/- 0.019	<0.11
19 - 20	-0.005 +/- 0.018	<0.11
20 - 21	-0.001 +/- 0.018	<0.11
21 - 22	0.001 +/- 0.018	<0.16
22 - 23	-0.004 +/- 0.018	<0.20
23 - 24	0.004 +/- 0.019	<0.12
24 - 25	-0.002 +/- 0.018	<0.11
25 - 26	-0.002 +/- 0.019	<0.11

[1] Indicates duplicate sample analysis.

TABLE 4.3

Recalculation of ^{137}Cs and $^{239,240}Pu$ for Halligan Reservoir.

Depth Interval (cm)	$^{239,240}Pu$ (pCi/cm^2)	^{137}Cs (pCi/cm^2)
0-1	0.029	1.15
1-2	0.044	1.02
	0.033	1.19
2-3	0.038	0.99
3-4	0.028	1.13
4-5	0.044	1.10
5-6	0.072	1.35
	0.017	1.05
6-7	0.027	1.29
7-8	0.034	1.12
8-9	0.050	1.38
9-10	0.056	1.57
10-11	0.052	1.73
	0.024	1.57
11-12	0.061	2.20
12-13	0.042	1.73
13-14	0.059	1.88
	0.049	2.20
14-15	0.055	2.20
15-16	0.083	2.67
16-17	0.063	2.98
	0.056	2.83
17-18	0.074	2.83
18-19	0.066	2.83
19-20	0.104	3.46
	0.056	3.46
20-21	0.078	3.30
21-22	0.078	3.61
	0.116	3.46
22-23	0.069	2.83
23-24	0.077	2.67
24-25	0.077	2.36
	0.068	2.04

(CONTINUED)

TABLE 4.3

(CONTINUED)

Depth Interval (cm)	$^{239,240}Pu$ (pCi/cm^2)	^{137}Cs (pCi/cm^2)
25-26	0.080	2.28
26-27	0.078	1.88
27-28	0.041	1.08
	0.055	0.88
28-29	0.017	0.78
29-30	0.0094	lim.
30-31	lim.	lim.
31-32	lim.	lim.
32-33	lim.	lim.
33-34	lim.	lim.
34-35	lim.	lim.
35-36	lim.	lim.
36-37	lim.	lim.

TABLE 4.4

Recalculation of ^{137}Cs and $^{239,240}Pu$ for Wellington Lake data.

Depth Interval (cm)	$^{239,240}Pu$ (pCi/cm^2)	^{137}Cs (pCi/cm^2)
0-1	0.119	5.03
1-2	0.102	5.18
2-3	0.139	5.18
3-4	0.121	5.50
4-5	0.126	5.34
5-6	0.122	5.34
6-7	0.156	6.91
7-8	0.22	7.70
8-9	0.19	8.17
9-10	0.19	9.27
10-11	0.29	9.74
11-12	0.22	8.78
12-13	0.20	7.54
13-14	0.22	5.97
14-15	0.14	4.24
	0.14	4.71
15-16	0.075	1.57
	0.083	1.57
16-17	0.022	0.58
	0.019	0.61
17-18	0.0016	0.44
18-19	0.0047	lim.
19-20	lim.	lim.
20-21	lim.	lim.
21-22	lim.	lim.
22-23	lim.	lim.
23-24	0.0063	lim.
24-25	lim.	lim.
25-26	lim.	lim.

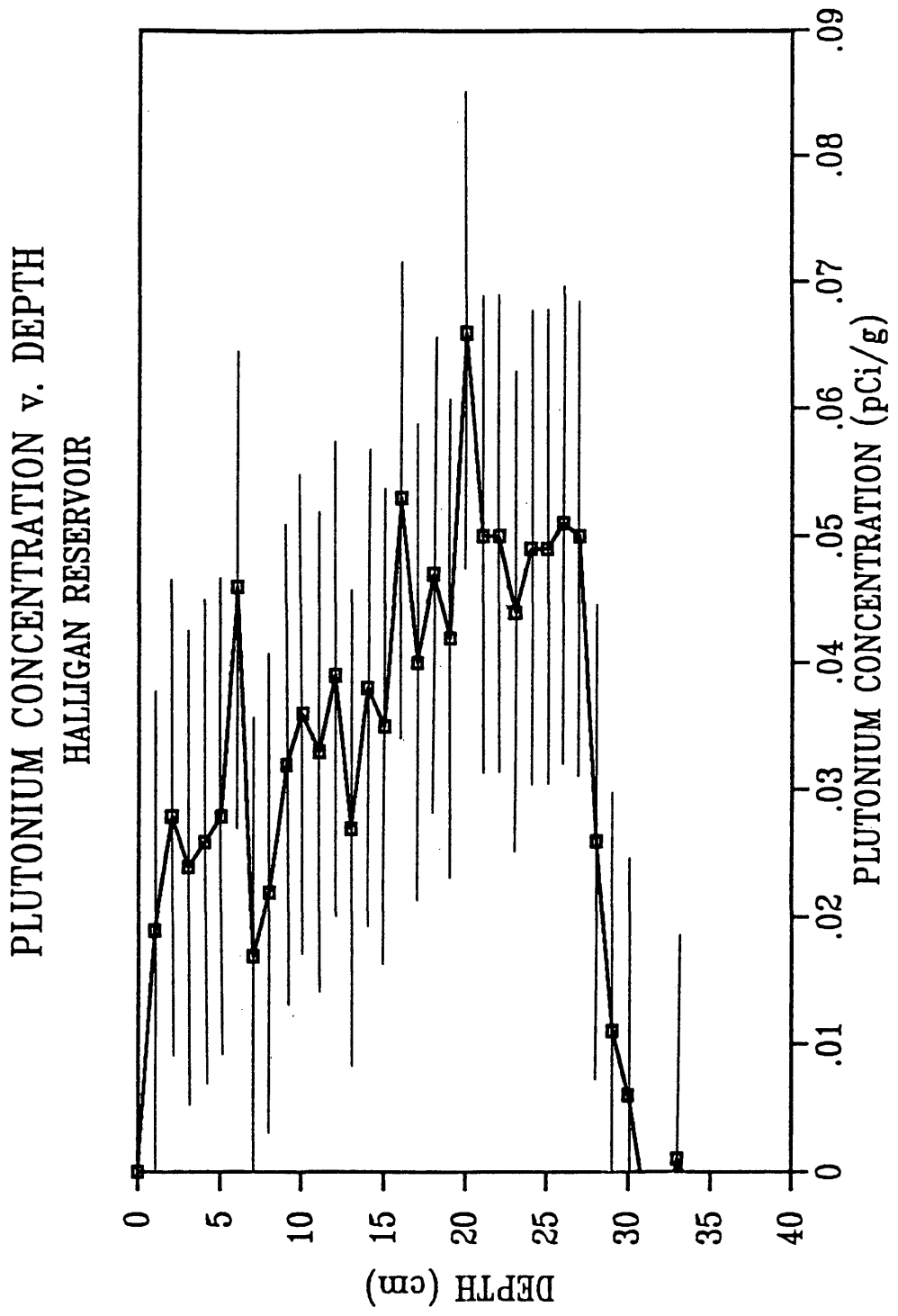

Figure 4.1. ^{239,240}Pu concentration v. depth, Halligan Reservoir

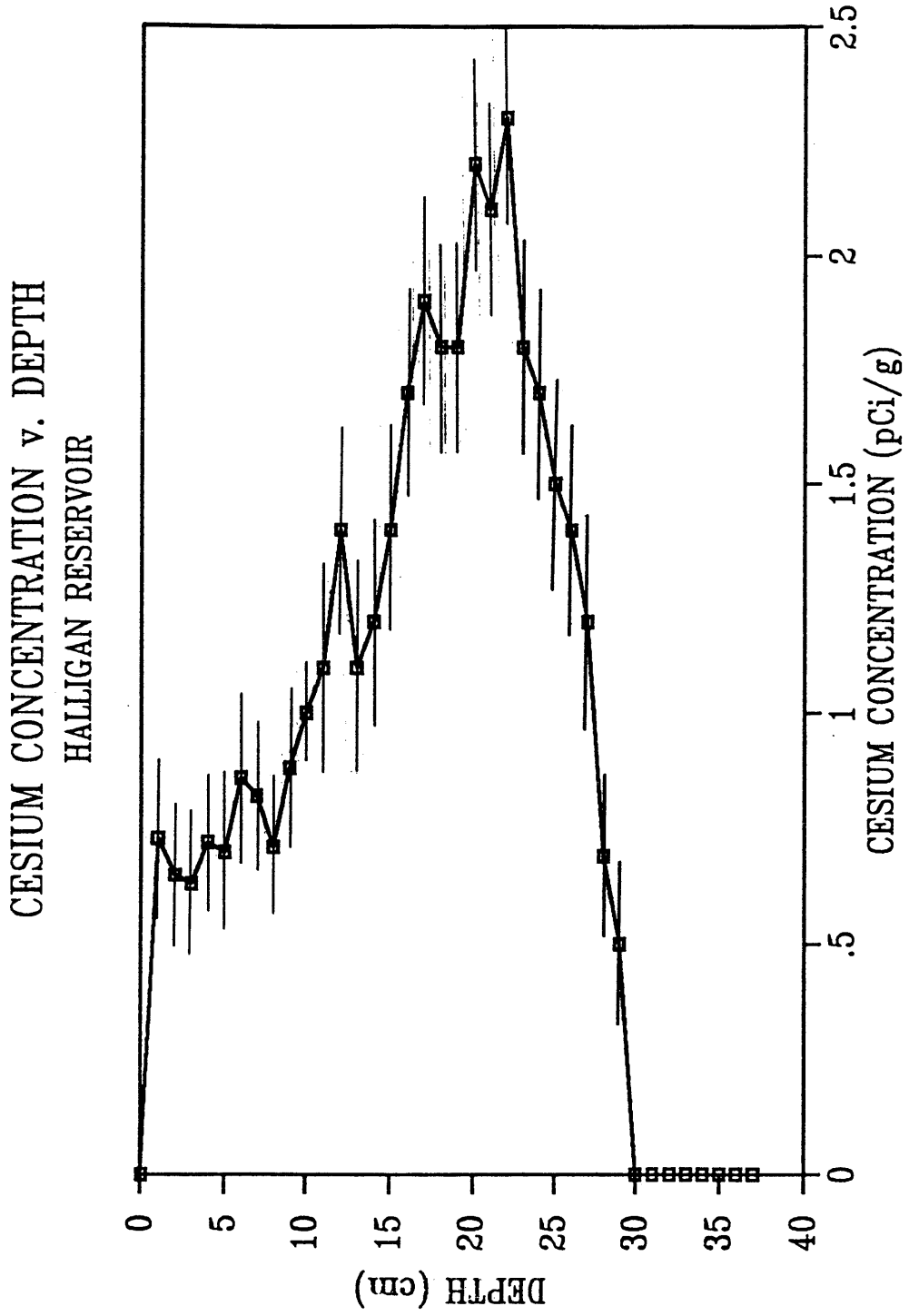

Figure 4.2. ^{137}Cs concentration v. depth, Halligan Reservoir.

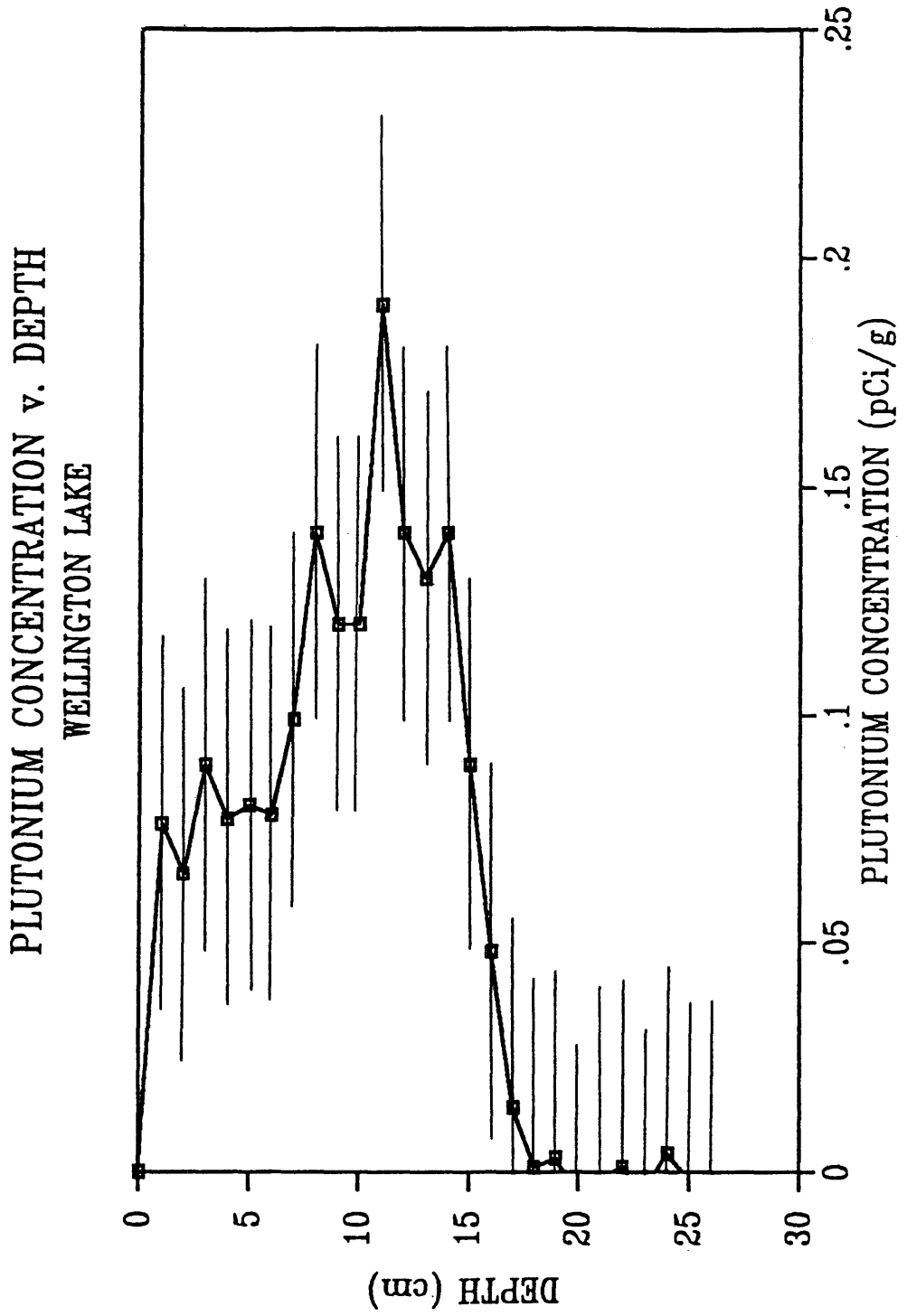

Figure 4.3. ^{239,240}Pu concentration v. depth, Wellington Lake.

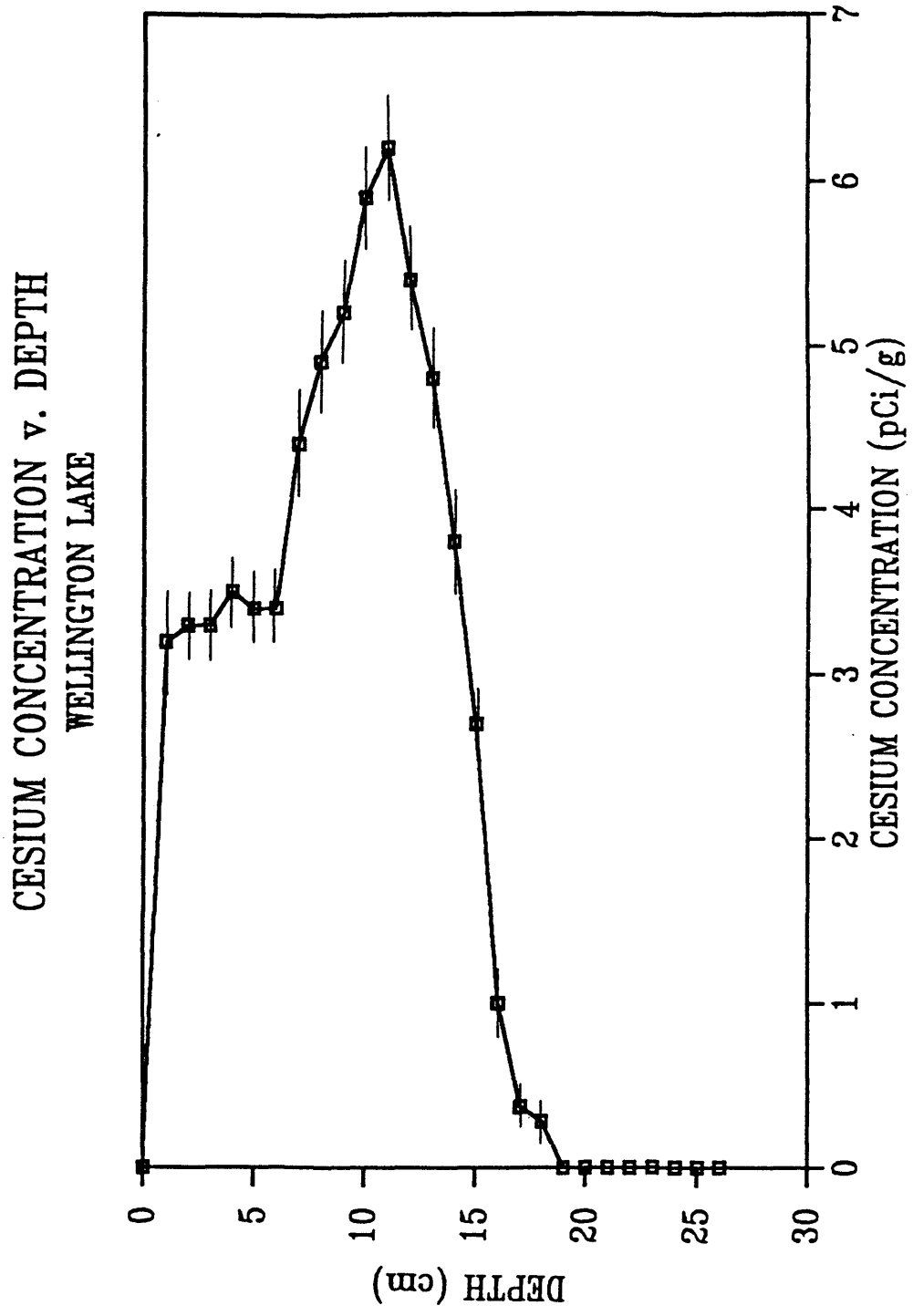

Figure 4.4. ^{137}Cs concentration v. depth, Wellington Lake.

either diffusion of the radionuclides between depth intervals is minimal or the diffusion of both the radionuclides is equal (Figures 4.1 - 4.4). Given the differences in chemical behavior between cesium and plutonium, it seems likely that if diffusion plays a major role in these systems, the peaks would be separated through the diffusion process. Because the peak concentrations occur at the same depth interval in both cases, it seems reasonable that diffusion of plutonium or cesium is not an important process in these systems.

Comparison of these data with the data from Great Western Reservoir and Standley Lake (Appendices 2 - 5) revealed that the $^{239,240}Pu$ concentrations observed in the Great Western Reservoir sediment are on the order of 2 magnitudes greater than the Halligan Reservoir maximum (i.e. 4.9 vs. 0.074 pCi/g) and 1 order of magnitude greater than the maximum (i.e. 4.9 vs. 0.19 pCi/g) observed in Wellington Lake sediments. Comparison of ^{137}Cs data from Great Western Reservoir revealed concentrations on the same order of magnitude as both Wellington Lake and Halligan Reservoir but approximately 4 times and 1.5 times lower than the reported maxima, respectively. The maximum reported concentration for ^{137}Cs in Great Western Reservoir sediments was 1.50 pCi/g vs. 2.3 and 6.2 pCi/g for Halligan Reservoir and Wellington

Lake respectively.

For comparison with the Standley Lake data from the Hardy, Livingston, Volchok and Burke study (1980), the data (reported as concentration) must be converted to activity per unit area. This can be achieved for the ^{137}Cs through the calculation,

$$A = C / D$$

where;

A = activity per unit area

C = concentration

D = areal density of the sample (cm^2/g)

Results of this conversion are given in Tables 4.3 and 4.4 corresponding to Halligan Reservoir and Wellington Lake respectively.

The peak ^{137}Cs activity from the Standley Lake core (26,300 pCi/m^2 at the 38-40 cm depth interval) can then be compared with the 97,400 pCi/m^2 peak value found for Wellington Lake and 36,100 pCi/m^2 for Halligan Reservoir.

Peak $^{239,240}Pu$ activity in the Standley Lake core was determined to be 35,700 pCi/m^2 at the 24-26 cm depth interval, while the Halligan Reservoir maximum was determined to be 1160 pCi/m^2. The Wellington Lake maximum

was identified as 2986 pCi/m^2.

The Standley Lake maximum is therefore one order of magnitude greater than the background $^{239,240}Pu$ activity, while the ^{137}Cs maximum is on the same order of magnitude as the background.

4.2 Statistical Determination

Initially, it can be seen that, with the exception of the 5-6, 19-20 and the 21-22 cm depth intervals (i.e. 0.46 +/- 0.020 vs. 0.011 +/- 0.020; 0.066 +/- 0.021 v. 0.036 +/- 0.022; 0.050 +/- 0.020 vs. 0.074 +/- 0.022 pCi/g, respectively) the reported error of the initial population was well within the error of the duplicate sample. This indicates that the values reported for each depth interval are acceptable.

Tables 4.6 and 4.7 show the results of the two-tailed t-statistic test. For both elements of concern (i.e. ^{137}Cs and $^{239,240}Pu$), the t-test implies that there is not enough evidence to suggest that the two replicate values are from differing populations. Either value for the plutonium or cesium concentration can therefore be reported as the values are not statistically different.

Figures 4.5 and 4.6 illustrate the fact that the mean

TABLE 4.5

Replicate Sample Statistical Analysis
Mean and Standard Deviation

Halligan Reservoir:

		$^{239,240}Pu$		^{137}Cs	
ID	Dup. ID	Mean	S.Dev.	Mean	S.Dev.
HR02	HR38	0.024	0.0049	0.705	0.0778
HR06	HR39	0.028	0.0247	0.765	0.1340
HR11	HR40	0.024	0.0127	1.050	0.0707
HR14	HR41	0.034	0.0049	1.300	0.1410
HR17	HR42	0.038	0.0028	1.850	0.0707
HR20	HR43	0.051	0.0212	2.200	0.0000
HR22	HR44	0.062	0.0169	2.250	0.0707
HR25	HR45	0.046	0.0042	1.400	0.1410
HR28	HR46	0.030	0.0063	0.625	0.0919

Wellington Lake:

WL15	WL27	0.088	0.0014	2.850	0.2120
WL16	WL28	0.050	0.0035	1.000	0.0000
WL17	WL29	0.013	0.0014	0.038	0.0141

TABLE 4.6

Two Sample Analysis Results
$^{239,240}Pu$ (Halligan Reservoir and Wellington Lake)

SAMPLE STATISTICS:

	SAMPLE 1	DUPLICATE	POOLED
Number of observations	12	12	24
Average	0.0439167	0.0378333	0.040875
Variance	0.00038572	0.0005669	0.000476
Standard deviation	0.0196397	0.0238092	0.021824

RATIO OF VARIANCES = 0.0068333

HYPOTHESIS TEST FOR H_0: $m_1 - m_2 = 0$

Difference in variances = 0 (95% confidence level)

Computed t statistic = 0.682775

Alpha = 0.05

Significance level = 0.501876

Conclusion: Do not reject H_0

TABLE 4.7

Two Sample Analysis Results
^{137}Cs (Halligan Reservoir and Wellington Lake)

SAMPLE STATISTICS:

	SAMPLE 1	DUPLICATE	POOLED
Number of observations	12	12	24
Average	1.3725	1.35667	1.36458
Variance	0.55455	0.63415	0.59435
Standard deviation	0.74468	0.79634	0.77094

RATIO OF VARIANCES = 0.874472

HYPOTHESIS TEST FOR H_0: $m_1 - m_2 = 0$

Difference in variances = 0 (95% confidence level)

Computed t statistic = 0.0503068

Alpha = 0.05

Significance level = 0.960332

Conclusion: Do not reject H_0

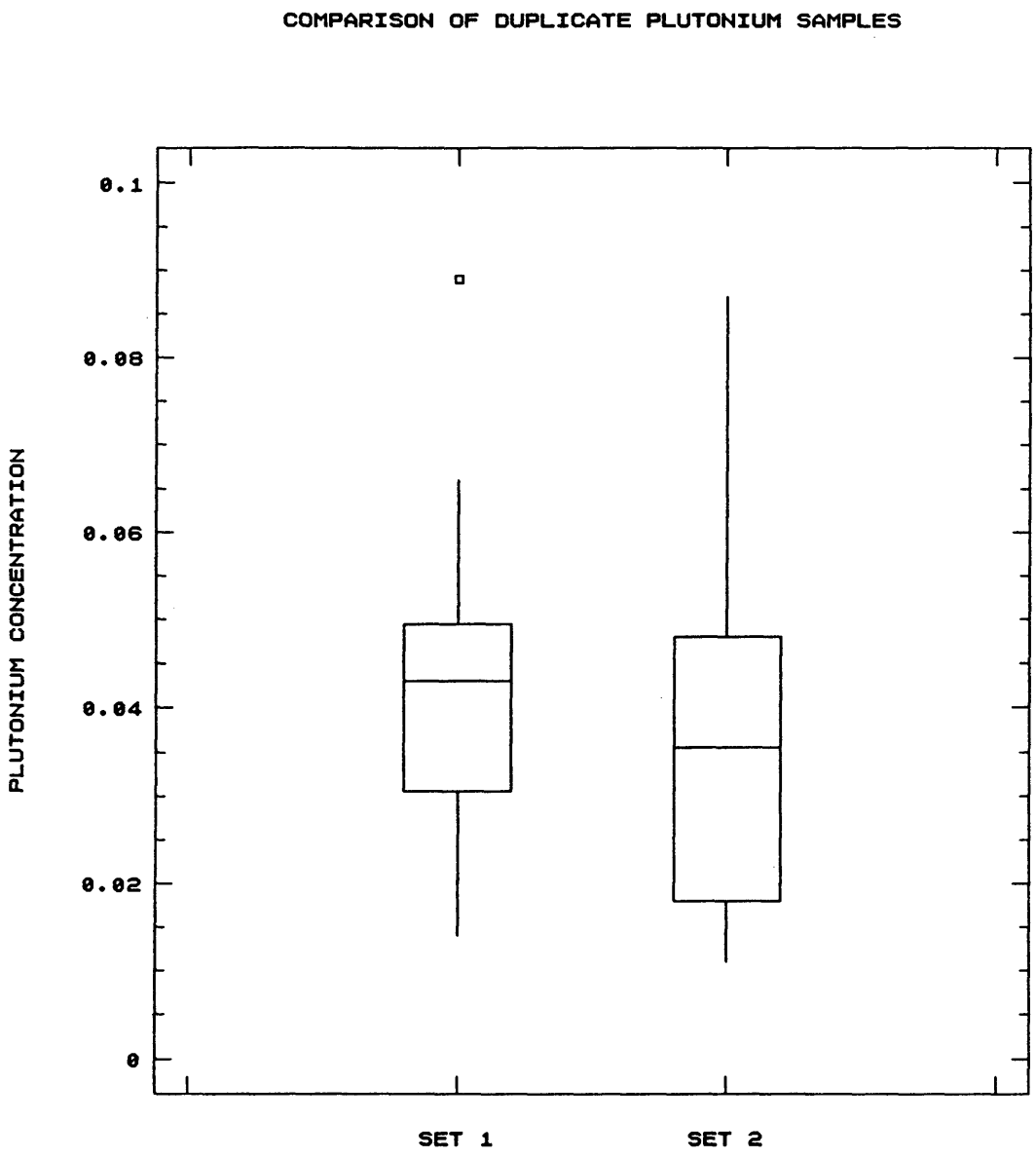

Figure 4.5. Comparison of $^{239,240}Pu$, two sample analysis.

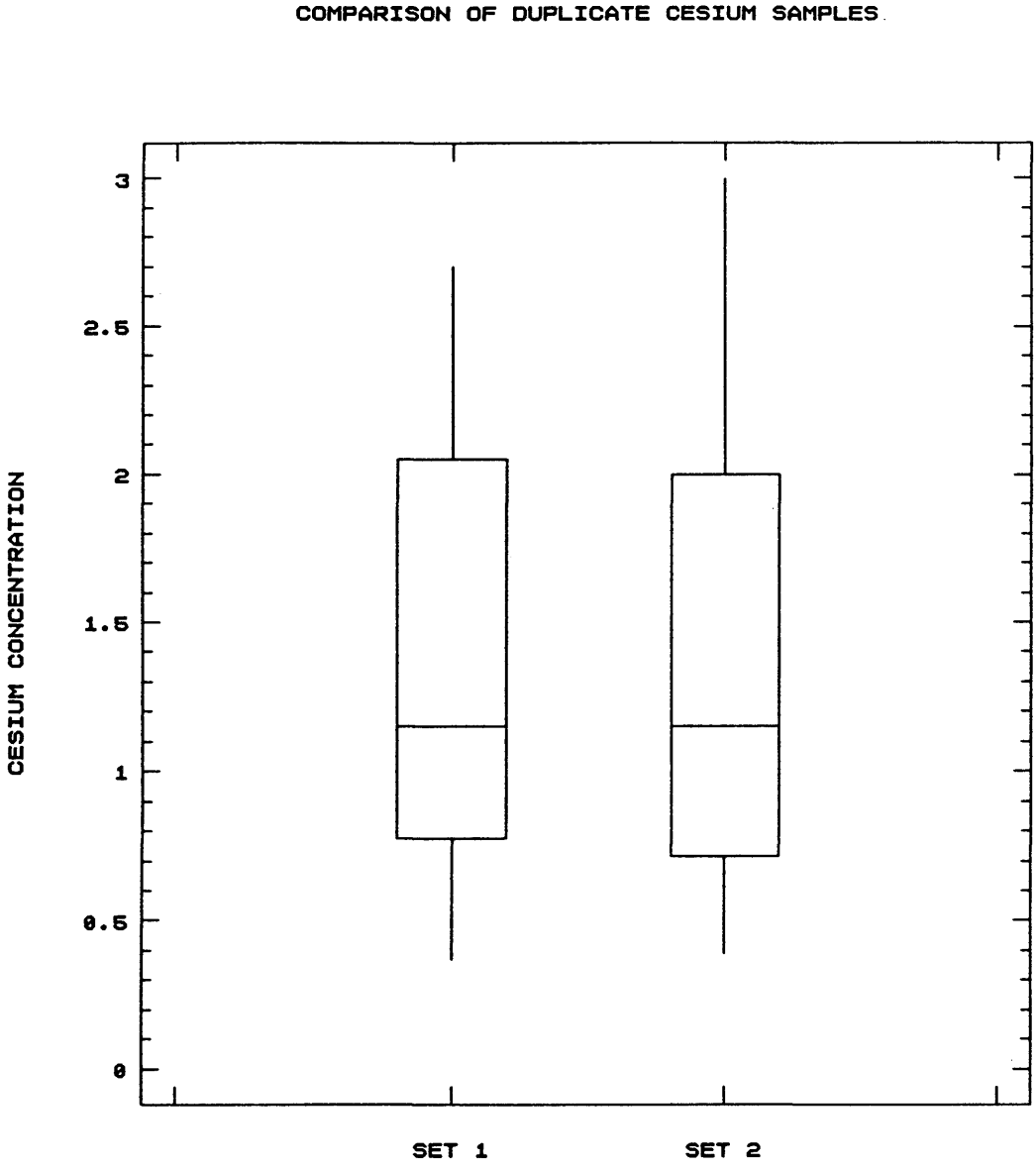

Figure 4.6. Comparison of ^{137}Cs, two sample analysis.

concentration of the duplicate population lies within 2 standard deviations of the mean concentration for plutonium.

4.3 Sedimentation Rates

^{137}Cs dating techniques have been widely established. The technique is based on the presence of a high ^{137}Cs horizon within the sediment corresponding with the peak in atmospheric testing. Assumptions governing the use of this technique are:

1) the peak ^{137}Cs activity interval within the sediments corresponds to the year 1964;

2) ^{137}Cs is absorbed to the sediments almost immediately, i.e. does not remain in the water column for long periods; and

3) migration of cesium within the sediments after deposition is insignificant (Robbins and Edgington, 1978).

The sedimentation rate is then determined as the length (in cm) of the dry sediment between the top of the core and the horizon of the maximum ^{137}Cs activity, divided by the time elapsed between 1964 and the date of core recovery (Vernet, et al. 1984).

The peak ^{137}Cs concentration in the Halligan Reservoir core occurred at the 20 cm interval, therefore:

20 cm / 25 yr = 0.80 cm/yr, and

for Wellington Lake

11 cm / 25 yr = 0.44 cm/yr.

It is important to note that the assumptions upon which this dating technique is based may not be valid. Brunskill (1984), discusses the possibility of diffusion of ^{137}Cs from intervals of high concentration to those of lower concentration. The observations have shown that ^{137}Cs may be able to move short distances through the pore water within the sediment. This may result in a significant change in the calculation of sedimentation rate. Due to the observation that both the cesium and plutonium peaks occur at the same sediment depth interval, it seems unlikely that diffusion of these elements plays a significant role in these systems.

Jaakkola, et.al. (1983) discusses the use of $^{239,240}Pu$ in the dating of sediments and in the determination of sedimentation rates. This technique is based on the same premise of the ^{137}Cs dating technique (i.e. the peak $^{239,240}Pu$ concentration corresponds to the years 1962-63) the

assumptions and possible problems are also associated with this technique. For Halligan Reservoir, the peak $^{239,240}Pu$ concentration occurred at the 20 cm depth interval. Therefore the sedimentation rate can be approximated as above:

20 cm / 25 yr = 0.80 cm/yr, and

for Wellington Lake

11 cm / 25 yr = 0.44 cm/yr.

Because the peak concentrations for $^{239,240}Pu$ and ^{137}Cs occurred at the same depth interval in both cases, the sedimentation rates determined using each technique are equal.

^{210}Pb dating techniques of the sediments are based on the constant flux of ^{210}Pb to the sediments from the decay of ^{238}U. ^{210}Pb is present in the atmosphere through the decay of ^{222}Rn (gas). The ^{210}Pb is precipitated similarly to ^{137}Cs discussed earlier. The sedimentation rates determined using this technique as established by Wolaver (in process) reveal the sedimentation rates for Halligan Reservoir to be approximately 0.24 cm/yr. This calculation is noticeably lower than the 0.80 cm/yr rate calculated using the ^{137}Cs and

$^{239,240}Pu$ techniques. Using the value reported by the ^{210}Pb determination, the maximum $^{239,240}Pu$ and ^{137}Cs concentrations should occur at the 11 cm depth interval as opposed to the 20 cm depth interval as reported earlier.

The ^{210}Pb determination for Wellington Lake revealed a sedimentation rate of approximately 0.19 cm/yr. The maximum ^{137}Cs and $^{239,240}Pu$ concentrations should occur at the 11 cm depth interval and this value correlated closely with the values calculated earlier (i.e. maximum ^{137}Cs and $^{239,240}Pu$ concentration occurrence at 11 cm).

Direct correlation of the sedimentation rates calculated (for the Wellington Lake sediments) for each of the techniques described tends to support the peak $^{239,240}Pu$ values obtained for comparison with those obtained for Great Western Reservoir and Standley Lake.

Discrepencies in the sedimentation rates calculated (for the Halligan Reservoir sediments) between the three techniques described, however, tend to call into question the reliability of the peak $^{239,240}Pu$ as well as the ^{137}Cs values reported for comparison. This discrepency may be attributed to a number of factors including, but not limited to:

a) abnormally high bioturbation of the sediments due to periods of low water retention,

b) increased wave erosion of the sediments in the region of the sample location due to periods of low water retention, or

c) exposure of the sediment bed during periods of extremely low retention causing discontinuities to occur in the sediment record.

Edgington and Robbins (1975) indicate that the $^{239,240}Pu$ and ^{137}Cs peak horizons in several Lake Michigan cores appeared deeper than predicted using ^{210}Pb sedimentation rate techniques. The authors suggest that either physical (wave energy) or biological turbation are the likely cause of this effect. Davis, et al. (1984) suggest that the occurrence of the ^{137}Cs peak at a horizon that is unexpected (lower) is due to postdepositional migration of radiocesium within the sediments. Without the benefit of geochemical studies of the clay portion of the sediments in Halligan Reservoir, it is difficult to determine the actual binding strength of ^{137}Cs (i.e. illite clays bind cesium into a highly immobile form).

Significant problems with these simple hypotheses still exist as one would expect that if bioturbation and/or wave erosion were to cause resuspension of material at the sediment/water interface, the ^{210}Pb, ^{137}Cs and $^{239,240}Pu$, all with similar adsorption properties at the pH levels seen in

this type of system, associated with that sediment should be redistributed equally. The effect of resuspension on the sedimentation rate is unclear. One would expect that resuspension would simply result in a lag (i.e. increase in residence time of the sediment in the water column) in the deposition of the ^{210}Pb, ^{137}Cs or $^{239,240}Pu$ in turn resulting in an artificially lower sedimentation rate. This simple explanation however, becomes complicated when the increased interaction of particles suspended within the water column is considered. This higher level of suspended solids may result in increased flocculation and/or coagulation of particles resulting in redeposition shortly following resuspension. This may decrease the impact of resuspension on the sedimentation rate.

Diffusion of ^{137}Cs in the Halligan Reservoir cores is probably minimal as the $^{239,240}Pu$ maximum occurs at the same depth horizon. Sources of sediment in this region also contain significant quantities of mica which will weather to form illite clays, thus preferentially fixing the cesium into an immobile form.

Further inspection of the ^{210}Pb analysis yields an inconsistant ^{226}Ra concentration with depth within the Halligan Reservoir core. ^{226}Ra is used to determine the quantity of ^{210}Pb contribution from the soil rather than the

atmosphere. The ^{226}Ra concentration is assumed to be constant throughout the depth of the core. Wolaver (in process) finds that the Halligan Reservoir core does not reveal constant concentration with depth, thus raising the suspicion that the ^{210}Pb dates are unreliable. The dates determined from ^{137}Cs and $^{239,240}Pu$ technique, therefore, may be accurate and the peak concentrations found in Halligan Reservoir are probably useful for comparison with Great Western Reservoir and Standley Lake.

4.4 Contaminant Dilution

The possibility remains that the lower concentrations of $^{239,240}Pu$ and ^{137}Cs observed in Halligan Reservoir and Wellington Lake are the result of higher sedimentation and similar fallout rate.

Comparison of the sedimentation rates in Wellington Lake (0.19-0.44 cm/yr) and Halligan Reservoir (0.24-0.80 cm/yr) indicated that the Halligan Reservoir sedimentation rate is only slightly higher than that of Wellington Lake. The degree of dilution effect is difficult to quantify, but offers a simple solution to this difference in concentration.

The sedimentation rates determined for Great Western Reservoir are within the 2.5 - 3.4 cm/yr range (Setlock,

1983) while the rate for Standley Lake averaged 3.4 cm/yr (Hardy, et al., 1976). Comparing these values to the sedimentation rates observed in Halligan Reservoir (<1.0 cm/yr) and Wellington Lake (0.19 cm/yr), it can be seen that the sedimentation rate is significantly higher in Standley Lake. If contaminant dilution plays a significant role in the comparison of these systems, it can be expected that the reported concentration values for Standley Lake and Great Western Reservoir are artificially low when compared to the concentration values for the control locations. For this reason, contaminant dilution cannot be expected to explain the difference in concentrations observed between these locations.

Chapter 5

Conclusion

The primary objective of this study was the comparison of the $^{239,240}Pu$ and ^{137}Cs concentrations in lake sediments impacted by activities at the Rocky Flats Plant vs. lake sediments considered to be unaffected by Rocky Flats. Using data received from Rockwell International (and supported by Accu-Labs Research) to quantify the impact of the Rocky Flats Plant, it was the primary goal of this project to determine the concentration of $^{239,240}Pu$ and ^{137}Cs in lake sediments attributable to global fallout from atmospheric nuclear weapons testing. Of the two lake systems surveyed, Halligan Reservoir and Wellington Lake, it was determined that the data received from the Halligan Reservoir core may be anamolous based on discrepancies in sedimentation rates calculated using three different methods. For this reason, little weight is given to these data. The sediment core from Wellington Lake however, appears to have accurate concentrations reported. The peak $^{239,240}Pu$ concentration was reported to be 0.19 +/- 0.02 pCi/g. Peak concentration values reported for Great Western Reservoir are 4.9 and 5.4 pCi/g (values from Accu-Labs Research and Rockwell International Labs, respectively). Judging from the reported values, activities at Rocky Flats Plant can be calculated to

be responsible for contamination of the sediments in Great Western Reservoir of between approximately 25x and 77x the background concentrations.

^{137}Cs concentrations appear to be no higher in the Great Western Reservoir sediments than the background levels indicate.

Maximum $^{239,240}Pu$ activity levels in Standley Lake are 26,300 pCi/m^2 compared with 2900 pCi/m^2 for Wellington Lake and 610 pCi/m^2 for Halligan Reservoir.

Peak ^{137}Cs activity levels in Standley Lake are 3570 pCi/m^2 compared with 97,400 pCi/m^2 for Wellington Lake and 22,000 pCi/m^2 for Halligan Reservoir.

The peak $^{239,240}Pu$ activity levels in Standley Lake can then be reported as between 9x and 43x background levels, while the ^{137}Cs peak activity are actually lower (between 0.04x and 0.16x) than the background levels.

It is interesting to note that the peak $^{239,240}Pu$ activity occurs at the 40 cm interval which corresponds roughly with the 1966 period (using 3.4 cm/yr, Hardy, et al.), while the ^{137}Cs peak activity occurred at the 1970 period.

Although the purpose of this study was the comparison of plutonium and cesium concentrations in Colorado lakes, the analysis revealed interesting information concerning

sedimentation rates within the lakes studied. The sedimentation rate determined for Wellington Lake was approximately 0.44 cm/yr. The sedimentation rate for Halligan Reservoir (although not supported by ^{210}Pb analysis) was found to be 0.80 cm/yr.

Recommendations

Problems encountered during this research reveal several improvements which could increase the reliability of the results obtained from this type of study.

Analysis of several cores from each lake would allow for determination of actual (as opposed to inferred) maximum contaminant concentrations.

Analysis of several lakes (more than 2) would allow for the establishment of a larger data base for comparison of maximum contaminant concentrations. This would decrease the possibility that one or two lakes may contain anomolously high contaminant concentrations due to abnormal precipitation or other climatic processes.

Establishment of a consistent sampling routine could eliminate differences that may result through the use of inconsistent sampling devices. This study utilized three different coring devices, each with its own associated and unequal error. The use of the same device for all sampling

would probably offer more consistent results.

Avoidance of lake systems which are near minimum detention would minimize the effect of sediment bed erosion and bioturbation causing resuspension of sediments, thus yielding more accurate dating of the sediments.

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APPENDIX 1

Determination of ^{137}Cs and $^{239,240}Pu$ per unit area for Standley Lake. (Source: Hardy, et.al., 1980)

Depth Interval (cm)	^{137}Cs (nCi/m^2)	$^{239,240}Pu$ (nCi/m^2)
0-2	3.8	0.31
2-4	6.8	0.81
4-6	5.8	0.49
6-8	6.4	0.56
8-10	6.3	0.58
10-12	7.6	0.74
12-14	7.7	0.86
14-16	9.4	1.18
16-18	8.1	1.22
18-20	9.3	1.68
20-22	9.1	1.93
22-24	11.5	2.96
24-26	11.7	3.57
26-28	12.8	2.87
28-30	12.6	1.46
30-32	13.1	0.66
32-34	15.3	0.27
34-36	16.4	0.22
36-38	22.0	0.37
38-40	26.3	0.59
40-42	22.8	0.59
42-44	23.4	0.60
44-46	20.0	0.57
46-48	17.0	0.38
48-50	13.6	0.29

APPENDIX 2

Determination of $^{239,240}Pu$
Great Western Reservoir
(Analysis by Rockwell International, Source, City of Broomfield, 1983)

Depth (inches)	Core 1 (pCi/g)	Core 2 (pCi/g)	Core 3 (pCi/g)	Core4 (pCi/g)	
0-2	0.20	0.19	0.16	0.25	
2-4	0.23	0.18	0.18		0.39
4-6	0.75	0.16	0.14	0.95	
6-8	1.00	0.14	0.24	1.40	
8-10	1.00	0.17	1.30	1.60	
10-12	0.11	0.67	0.75	2.10	
12-14		0.97	1.50	3.70	
14-16		1.30	1.90	3.80	
16-18				5.40	
18-20				3.30	

APPENDIX 3

Determination of $^{239,240}Pu$
Great Western Reservoir
(Analysis by Accu-Labs Research, Source; City of Broomfield, 1983)

Depth (inches)	Core 1 (pCi/g)	Core 2 (pCi/g)	Core 3 (pCi/g)
0-1	0.034	0.650	0.360
1-2	0.057	0.720	0.470
2-3	0.059	1.200	0.810
3-4	0.025	1.500	0.890
4-5	0.013	1.300	1.600
5-6	0.530	1.200	1.700
6-7			2.200
7-8			4.900
8-9			2.800
9-10			4.000
10-11			1.800

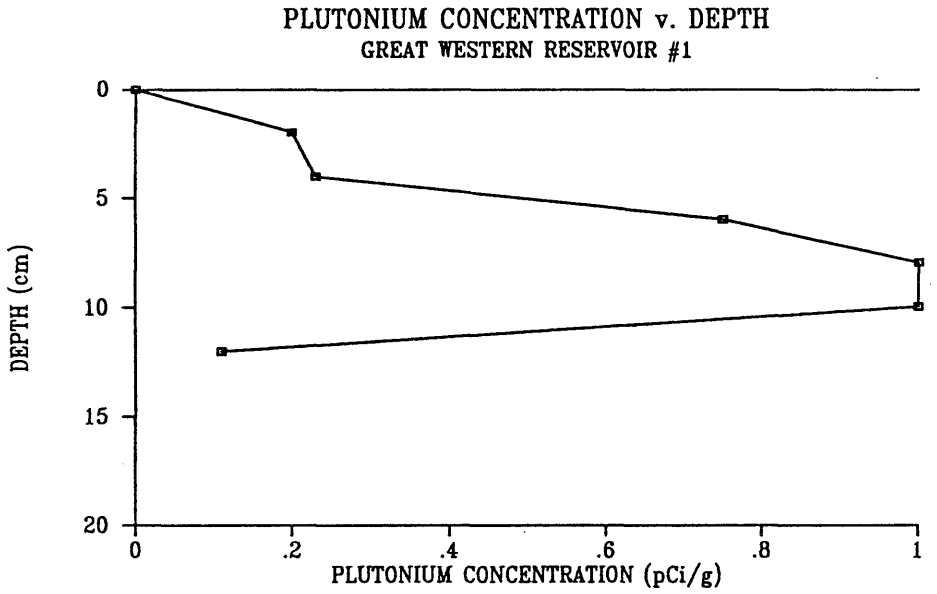
APPENDIX 4

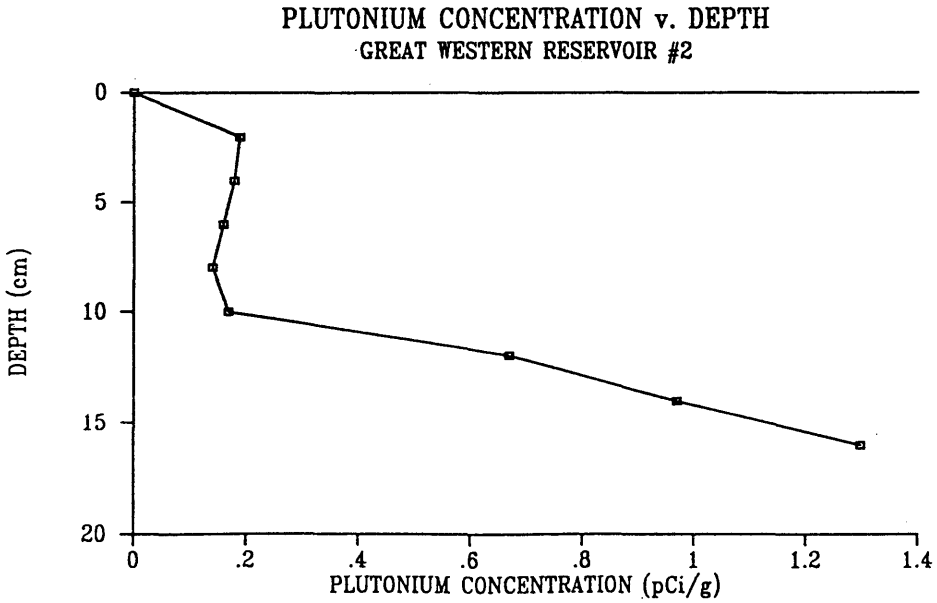
Determination of ^{137}Cs
Great Western Reservoir
Analysis by Rockwell International,
(Source: City of Broomfield, 1983)

Depth (inches)	Core 4 (pCi/g)
0-1	0.40
1-3	0.48
3-5	0.45
5-7	0.25
7-9	0.66
9-11	0.52
11-13	0.85
13-15	0.90
15-17	1.50
17-19	1.20

APPENDIX 5

^{239,240}Pu Concentration (pCi/g) v. Depth (cm)
Great Western Reservoir
(Rockwell International, 1983)





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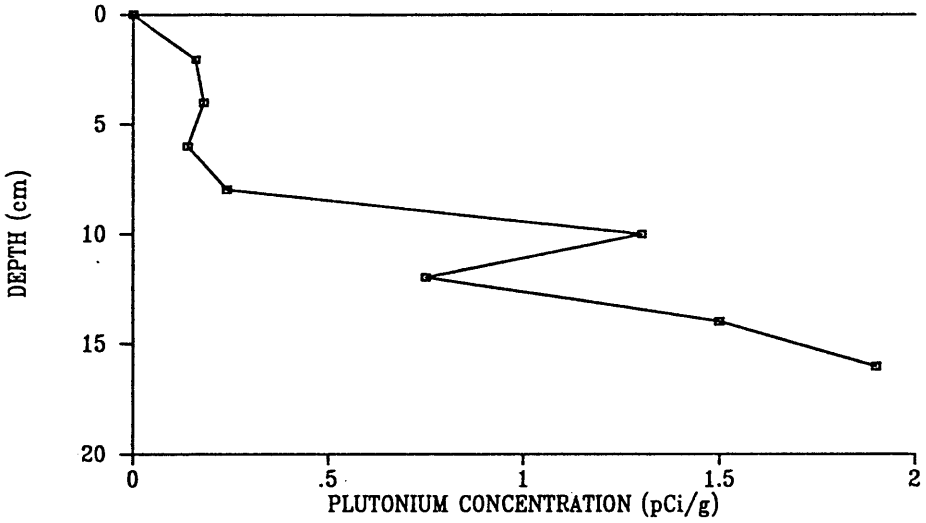
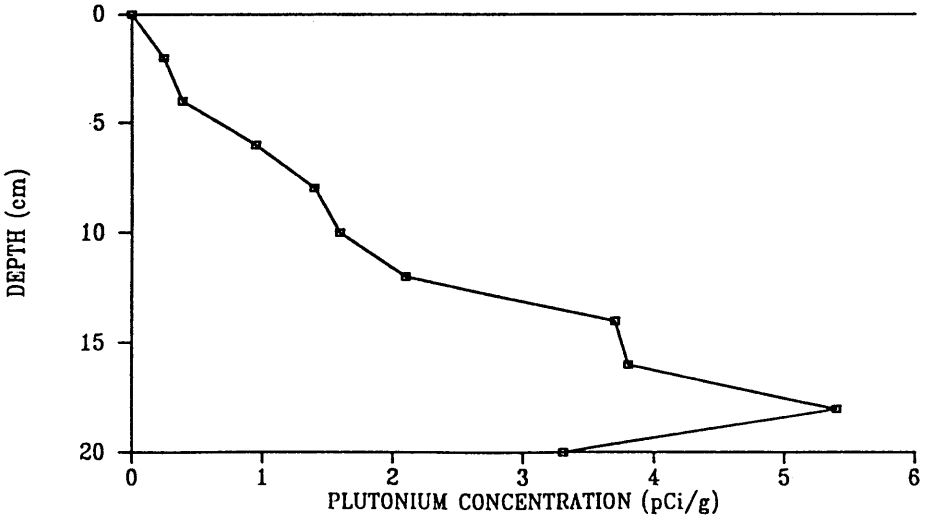
APPENDIX 5
(CONTINUED)

PLUTONIUM CONCENTRATION v. DEPTH
GREAT WESTERN RESERVOIR #3

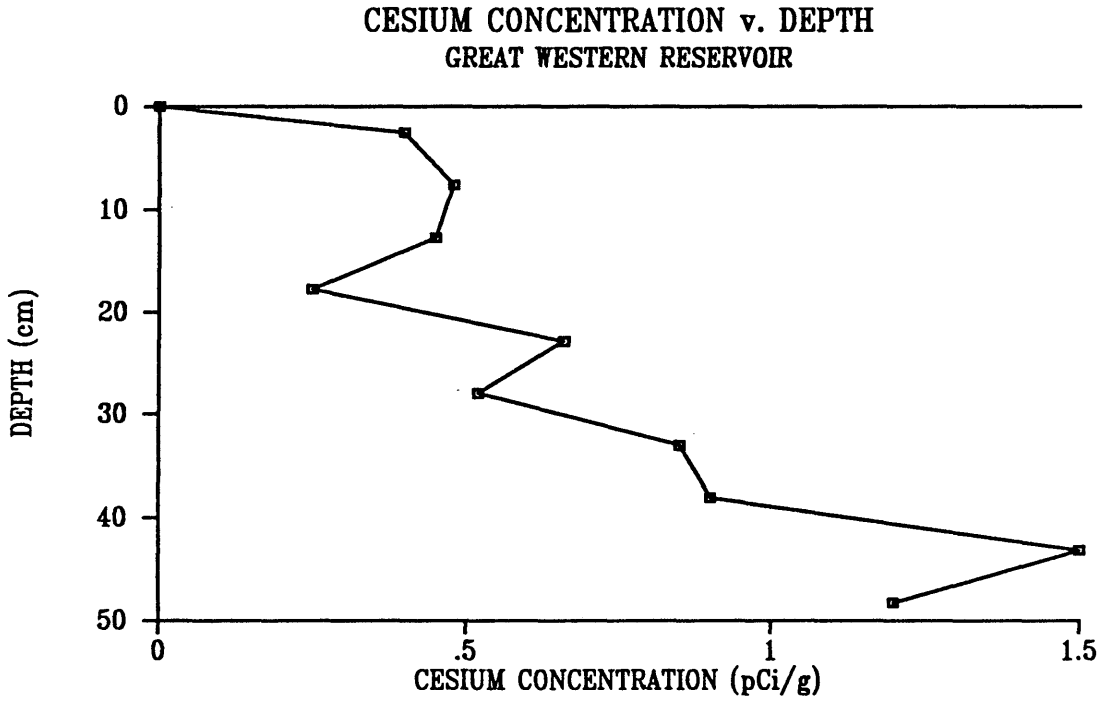


PLUTONIUM CONCENTRATION v. DEPTH
GREAT WESTERN RESERVOIR #4



APPENDIX 6

^{137}Cs Concentration (pCi/g) v. Depth (cm)
Great Western Reservoir
(Rockwell International, 1983)



APPENDIX 7

ANNOUNCED ATMOSPHERIC NUCLEAR TESTS
APPROXIMATE YIELDS

United States

Year	number of detonations	yield
1945	3	59 kt[1]
1946	1	20 kt
1948	3	104 kt
1951	15	160 kt
1952	10	14+ Mt[2]
1953	11	2.5 Mt
1954	6	15.1 Mt
1955	13	166 kt
1956	14	several Mt
1957	25	483 kt
1958	52	several Mt
1962	1	low

United Kingdom

Year	number of detonations	yield
1952	1	1 kt
1953	2	kt range
1956	6	kt range
1957	7	app. 4 Mt
1958	5	app. 3 Mt

(CONTINUED)

APPENDIX 7

(CONTINUED)

Soviet Union

Year	number of detonations	yield
1949	1	na[3]
1951	2	na
1952	2	na
1954	4	na
1955	4	Mt range
1956	7	high
1957	13	several Mt
1958	24	several Mt
1961	30	high Mt
1962	38	high Mt

France

Year	number of detonations	yield
1960	3	60-70 kt
1961	1	low
1966	5	200-300 kt
1967	3	low
1968	5	2-3 Mt
1970	8	several Mt
1971	5	Mt range
1972	3	low
1973	5	low
1974	7	na

(CONTINUED)

APPENDIX 7

(CONTINUED)

China

Year	number of detonations	yield
1964	1	20 kt
1965	1	20 kt
1966	3	500-800 kt
1967	2	3 Mt
1968	1	3 Mt
1969	1	3 Mt
1970	1	3 Mt
1971	1	20 kt
1972	2	20-200 kt
1973	1	1-3 Mt
1974	1	1 Mt
1976	3	4.2 Mt

[1] kt = kiloton equivalent TNT

[2] Mt = megaton equivalent TNT

[3] na = yield not available