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Enclosed for your information is a recent Colorado State University progress report on the Radieoecological Investigations at the Rocky Flats Plant.

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bmb

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EG&G-RF/ASC 83749AM/CSU-3

PROGRESS REPORT

ON

RADIOECOLOGICAL INVESTIGATIONS

AT ROCKY FLATS

TO

EG&G ROCKY FLATS, INC.
ROCKY FLATS PLANT
P.O. BOX 464
GOLDEN, COLORADO 80402-0464

AGREEMENT ASC 83749AM

BY

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JUNE 24, 1991

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I. INTRODUCTION AND OVERVIEW

This constitutes a report on progress to date on Contract ASC 83749AM since the earlier report of January 31, 1991. The main objectives of this project are: (1) to evaluate the current distribution of plutonium, americium and several natural radionuclides in the terrestrial environment near the 903 Pad at Rocky Flats, and (2) to provide an updated, state-of-the-art assessment of an appropriate cleanup standard for plutonium and americium in soil east of the 903 Pad. These studies are deemed important for documentation of environmental conditions as required under the National Environmental Policy Act, and for future decisions concerning environmental remediation as required under the Resource Conservation and Recovery Act and the Superfund Amendments and Reauthorization Act.

This work builds upon research conducted and experiences gained at Rocky Flats in the early 1970s by Colorado State University. This new work is adding several radionuclides of interest and an updated assessment of dose by virtue of new analysis capabilities and a state-of-the-art modeling capability developed over the last ten years. Drs. F. W. Whicker, S. A. Ibrahim and L. Fraley, Jr., along with several graduate students, are conducting the work at Rocky Flats. Students involved in the project include Kathy Higley, Scott Webb, Ernest Antonio and James Jarvis. Effective February 1, 1991, Sharren Reuss was appointed as a research manager for the project (50%).

The laboratory at CSU is now generating data on radionuclide concentrations in environmental samples at a good pace. The data quality appear to be excellent, by virtue of good facilities, equipment, experience, and a rigorous quality assurance program. Plant uptake studies are continuing at the CSU greenhouse. Good progress has been made on the development of a

computer simulation model for a risk assessment of radionuclides in soil at Rocky Flats.

We wish to acknowledge the NEPA personnel at Rocky Flats for their excellent cooperation and assistance.

II. TASK A. EVALUATION OF STANDARDS FOR PLUTONIUM AND AMERICIUM IN SOIL AT ROCKY FLATS.

A. Objective

To provide the best estimate of potential dose to the public from exposure to plutonium and americium contaminated soil in the buffer zone at the Rocky Flats Plant. This estimate can be used to develop a risk-based approach to setting reasonable cleanup standards.

B. Synopsis of the Project

Plutonium and americium have been detected in the soil around the Rocky Flats Plant. The presence of these radionuclides in the plant environment is primarily attributed to the leakage of plutonium contaminated cutting oil from 55-gallon drums previously stored on the 903 Pad at the southeast corner of the site. Because of the potential hazards of plutonium, the question has arisen as to the need for remediation (i.e., removal) of the contaminated soil. The purpose of this project is to provide the best possible estimate of the potential dose to the public from the presence of plutonium and other radionuclides in the environment at Rocky Flats. This objective will be accomplished by using a mathematical model (a computerized version of the PATHWAY model by Whicker and Kirchner, 1987) to predict the range of potential exposures to the public. In order to provide the best possible estimates of potential exposure, site-specific data will be developed and used as input for the model.

The information required to run the model includes current and past data on the distribution of plutonium in the environment. The data to be collected and used in the model will include distribution of the radioisotopes as a function of soil depth, particle size distributions of the contamination, radionuclide concentration on and in vegetation, and resuspension factors for various soil and meteorological conditions.

Data will be collected from transects extending from the center of the 903 Pad outward and will include a reference location as well as one down gradient of the contaminated area. The transect through the contaminated area will extend to Indiana Street. Sampling plots are to be located along each transect, and the spacing between plots will increase as one moves toward the east.

The exact locations of the sampling sites are to be determined after the distribution of plutonium extending from the 903 Pad has been ascertained from field surveys and soil assays.

Site-specific data on plant uptake of plutonium and americium from Rocky Flats soil are also needed for the model, since this pathway becomes relatively more important as the potential for resuspension declines. A variety of greenhouse experiments with Rocky Flats soil are underway to measure this parameter

C. Progress on Soil Measurements and Modeling (K. Higley, T. Kirchner and W. Whicker)

The soil collected in conjunction with the FIDLER surveys was analyzed for ^{241}Am. Based on these results and other data collected at the RFP, soil parameter values for the computer model were selected.

The mathematical model describing movement of ^{241}Am and ^{239,240}Pu in the RFP environment was constructed and tested. Site specific input parameters, such as isotopic ratios for plutonium, were used for the initial application of the model. An on site hypothetical exposure scenario was selected and uncertainty/sensitivity analyses performed. The results of the analyses identified those parameters driving the dose assessment, and indicated where additional information is needed to improve model performance.

This work is included in the Master's thesis of K. Higley, which was successfully defended in late April 1991. This document will be finalized and made available in the near future. The modeling results were also presented at a technical seminar to EG&G personnel on May 3, 1991.

D. Plant Uptake Studies (J. Jarvis and L. Fraley, Jr.)

Blue grama (*Bouteloua gracilis*) was harvested from the mixed soil containers in December 1990. Similarly, native species from the uncovered and covered intact soil cores were harvested in November and December 1990, respectively. Refer to Whicker et al. 1990 for the experimental design.

Radiochemical analysis has been completed for the Blue Grama and uncovered and covered core vegetation samples. Vegetation weight analyzed for Pu varied from 1 to 5 g (dry). The majority of $^{239,240}Pu$ values were below detection limits with the exception of the uncovered cores which were, on average, above the detection limits (Table 1). There is a significant difference ($p < 0.05$) in Pu concentrations between vegetation data for covered and uncovered soil cores. This difference is most likely due to Pu contamination of the external surfaces of the plants during greenhouse watering procedures. Covering the contaminated soil with sand and gravel, and harvesting vegetation grown under these conditions resulted in a 75 fold reduction in mean Pu concentration.

Soil cores were also measured for ^{241}Am by gamma spectroscopy using a 7.65 cm diameter LEPS (low energy photon spectrometer) HPGe detector. The ^{241}Am data were then used to obtain a weighted estimate of the $^{239,240}Pu$ concentration in soil for each container assuming a 20% ^{241}Am ingrowth (Table

2). Each container was measured at three different levels, each level being measured at four equally spaced points surrounding the container. This indirect method provided an initial estimate of the Pu concentration in soil. Radiochemical analysis was undesirable at this time because it would require destruction of some or all of the cores.

The mixed soil containers had a higher soil Pu concentration than the intact cores. This was to be expected as the mixed soil was obtained from the top 9 cm of soil while the cores contain soil from a maximum depth of about 30 cm. Sampling to this depth has the effect of diluting the overall container concentration. The estimated core $^{239,240}Pu$ concentration measurements agree well with the field depth distribution found on the Macroplot 1 site (Whicker et al. 1990). The negative slope of the regression line indicates a decreasing concentration as the measurements approach the bottom layers of the container (Figure 1).

Mean vegetation uptake ratio (UR) and concentration ratio (CR) values were generally low and ranged from 10^{-3} to 10^{-5} (Table 3). In comparison, CR values from the 1976 study (Little 1976) ranged from 1.5×10^{-2} to 5.4×10^{-2} with a mean CR of 3.4×10^{-2} (CV = 0.29). These values are higher than those of the present greenhouse study by a factor of 10^1 to 10^3. To some extent the higher values presented earlier may be explained by experimental differences such as analyzing the vegetation unwashed, use of liquid scintillation methods for counting, and, more importantly, the differences between greenhouse and field conditions.

Repeated harvesting of the core samples will continue for the next 4 to 8 months to obtain large sample size for Pu measurements. Furthermore, specimens obtained from the cores will be separated into grass and forbs (provided that a sufficient sample size is available).

Wheat and cabbage seeds are now being prepared for germination and subsequent planting. The wheat seeds require an eight week period of vernalization or "cold hardening" in order to produce grain at maturity. The number of wheat container replicates will be increased to eight pots at planting time in order to increase the sample size.

III. TASK B. DISTRIBUTION AND TRANSPORT OF RADIONUCLIDES (S. Webb and S. Ibrahim)

A. Objectives

This task constitutes a resurvey of plutonium concentrations in soil, litter, vegetation, and small mammals in two study plots established in 1972. The survey will determine whether work conducted by Colorado State University during the period 1972-1976 is still relevant and applicable. Of specific interest is whether and to what extent the distribution patterns of plutonium have changed. Soil depth profile data for Pu will indicate the rate of downward transport of radionuclides and determine if changes have occurred. Plant/soil and animal/plant concentration ratios will indicate whether the Pu mobility has changed over the past 15 years. The plutonium data will also verify whether or not measurable quantities of new plutonium have been deposited in the study plots since the earlier work.

Other aspects of this task not previously studied are to measure levels and transport rates of ^{241}Am, an ingrowth product of ^{241}Pu decay. Data from washed and unwashed vegetation should provide valuable information regarding the relative contribution of dust resuspension on plant surfaces versus true plant uptake for both Pu and Am isotopes. Comparative information on ^{232}Th, ^{226}Ra and ^{238}U will indicate how the biological mobility of natural radioactivity compares to Pu and Am. Another man-made radionuclide of interest in the present study is ^{137}Cs. Soil profile data for ^{137}Cs at Rocky Flats will be compared to worldwide fallout data from offsite locations. Finally, we will correlate gross alpha and gamma survey measurements with the radiochemical analysis data to develop a sensitive but more rapid survey technique for Pu contamination.

B. Materials and Methods

1. Collection, Preparation, and Analyses of Soil and Vegetation Samples

Two study plots (Macroplot 1 and Macroplot 2) initially established during the period 1972-1976 were relocated and sampled for soil and vegetation (Figure 2). Five background sampling locations were also selected and sampled: Woman Creek drainage west boundary (Plot WC); southwestern most corner of the buffer zone (Plot SW); northwest part of the buffer zone (this plot was previously established by CSU in 1974 (Plot NW); Flatiron Vista Trailhead near the intersection of Colorado Highway 93 & 128 (Plot FV); and Trailhead Greenbelt Plateau (Plot GP) near the same intersection. Vegetation, litter and soil samples were collected from each sampling location using methods comparable to those used by Little et al. 1980. Collection and preparation of soil and vegetation samples as well as the analysis of plutonium have been described in detail in our progress report of September 15, 1990.

2. Analytical Quality Control

An internal quality control program and participation in interlaboratory comparisons were instituted to document the validity of the analytical results. Detector backgrounds are documented on a routine basis to evaluate laboratory contamination. Standards traceable to the National Institute of Standards and Technology (NIST, formerly NBS) are used routinely to determine detector efficiencies, to set energy regions, and to track instrument drift. Reagent blanks are processed to detect any external contamination. One blank is analyzed with every 6-10 samples. Replicate analyses are being performed on a number of large samples.

The internal quality control program including replicate analyses provides an estimate of the laboratory precision. The accuracy of the analytical technique will be determined by participating in interlaboratory comparisons. Considerable effort was devoted to analyze and report Pu concentrations in soil samples for such comparisons. The intralaboratory comparisons program is sponsored by the International Atomic Energy Agency (IAEA). Preliminary results reported by members participating in this program were published early in 1991 (Table 4).

C. **Results and Discussion**

Some elements of this task have been completed, while others are still in progress. The results to date were presented at a technical seminar to EG&G personnel on May 3, 1991 by Scott B. Webb. With respect to estimates of standing crop (vegetation and litter) biomass, we presented the complete data in the September 15, 1990 report. Average dry mass in g/m^2 of vegetation and litter observed from this study is generally within the range reported earlier for data collected and averaged over several years. These data will be used to calculate radionuclide inventories in the biological compartment of the Rocky Flats ecosystem.

The analysis of $^{239,240}Pu$ and ^{238}Pu in washed and unwashed vegetation and litter for both macroplots have been completed and data discussed in the previous progress report. Plutonium concentrations in vegetation and litter from Macroplot 2 were lower than from Macroplot 1 by over two orders of magnitude. The mean vegetation concentration in Macroplot 1 was an order of magnitude lower than for litter (Figures 3 and 4). There is probably a significant fraction of Pu activity associated with surface soil that was included in both washed and unwashed litter samples.

Total plutonium concentrations ($^{239,240}Pu$ + ^{238}Pu) in vegetation from the earlier study (Little 1976) were much higher than those observed in the current work. With regard to litter, values from the earlier study were 9 to 14 times greater than reported herein. These data provide strong evidence that a decrease in the biological availability of Pu for plant uptake and accumulation may be taking place over time. Continued investigation to obtain soil values and plant/soil concentration factor estimates is underway to elucidate any changes of Pu bioavailability with time.

Generally, ultrasonically washed vegetation and litter from both macroplots had lower plutonium concentrations than for unwashed samples (Figure 5). The removal efficiency for the ultrasonic washing procedure used was estimated from Ti tracer experiments to be about 75% for a similar environment, but for a different plant type. Therefore, residual surficial activity probably contributed somewhat to the observed values from washed vegetation and litter.

The isotopic ratio of $^{239,240}Pu/^{238}Pu$ in vegetation and litter from Macroplot 1 ranged from 48 to 53 and 67 to 89, respectively (Figure 6). Most of ^{238}Pu concentrations from Macroplot 2 were below detection limits, thus the above ratio could not be calculated. Most of the plutonium analyses in the earlier study resulted from liquid scintillation counting in which no distinction between $^{239,240}Pu$ and ^{238}Pu can be made. Three vegetation and four litter samples were, however, processed for isotopic plutonium by a commercial laboratory. Based on the limited data from the earlier study, the Pu isotopic ratio appeared to be within the range observed presently.

With respect to Pu measurements in soil, all soil profiles have been completed from both macroplots for the < 2 mm particle size fraction. Preliminary observations based on the available data are:

1) Concentrations of $^{239,240}Pu$ in the top 3 cm of soil ranged from 200 to 575 dpm/g at the various sampling locations on Macroplot 1 (Figure 7).

2) The concentrations of plutonium in soil decreased exponentially with depth and were about two orders of magnitude lower at 21 cm depth relative to the soil surface.

3) Plutonium-238 concentrations were much lower than for $^{239,240}Pu$ in the same soil samples and the isotopic ratio decreased with depth (Figure 8).

4) There was a positive correlation (r^2 = 0.88) between Pu concentrations and previously reported gamma measurements for ^{241}Am from Macroplot 1 (Figure 9). These data will be refined when completed to develop a field survey technique for Pu in soil.

5) Data from soil samples analyzed from Macroplot 2 indicate Pu concentrations of about two orders of magnitude lower than in Macroplot 1 for the soil particle size (Figure 10).

6) Our Pu soil values are generally lower than those reported in the earlier study by Little et al. 1980.

Soil samples collected from ten locations in Macroplot 1 and five locations in Macroplot 2 were also analyzed for ^{241}Am (Figure 11) and gross alpha (Figure 12). Levels of ^{241}Am in soil from Macroplot 2 were below the detection limits by gamma counting, and thus will be radiochemically analyzed. A more complete gamma measurement to include ^{241}Am, ^{137}Cs, ^{226}Ra, and ^{232}Th are planned for the same samples after sieving to various particle size fractions in sealed metal containers.

The average concentration of ^{241}Am varied from soil depth between 100 dpm/g on the surface and 0.8 dpm/g at the 21 cm depth. The data indicate that most of the ^{241}Am is still near the surface some 25 years after the initial contamination. Surface soil concentrations for ^{241}Am ranged from 20 to 100 dpm/g, which is comparable to ground measurements reported recently at two sampling locations near Macroplot 1 (EG&G 1990). The detectability of ^{241}Am at the 21 cm soil depth indicates a slow, but definite downward movement. In one sampling location, there appears to be two ^{241}Am peaks at the 6 to 9 and 9 to 12 cm depths. This sampling location was particularly rocky and possible mechanisms are that surface contamination migrated downward by gravity acting on particulates and/or with water percolation. Observed sample variability may be attributed to the initial heterogeneous pattern of contamination. The concentration of ^{241}Am was strongly correlated with depth; as soil depth increased the Am concentration decreased (Figure 11).

Gross alpha measurements conducted on the same samples also indicate a strong correlation with soil depth. There was a positive correlation between gamma measurements for ^{241}Am and gross alpha measurements. These values will also be correlated with plutonium concentrations obtained via radiochemical analysis to develop and refine a field survey technique for Pu concentration.

D. Small Mammal Trapping (E. Antonio and S. Ibrahim

The purpose of this task is to determine the concentration of Pu and Am in small mammal tissues sampled from Macroplots 1 and 2. These data will be used to compare with the results obtained by Little (1976), to determine if the concentrations in the small mammal tissues have changed significantly in the last 15 years and to determine concentration ratios between small mammal tissues and soil.

Work on this project began in late March 1990 when traps in Macroplot 2 were located. The plot measured 63 x 63 m and 36 trap locations were marked with flags. Macroplot 1 in the Am zone was not established until May 1990. Trapping in Macroplot 2 commenced with setting out and baiting 36 traps. Five trapping dates were set and a total of 26 rodents were obtained from Macroplot 2. In June 1990 the 36 traps were removed from Macroplot 2 and placed on Macroplot 1. A total of 31 specimens have been collected from Macroplot 1.

The small mammal specimens have been identified to species with *Peromyscus maniculatus* being the predominant species trapped (Table 5). All specimens have been dissected and five tissue samples (pelt, muscle, liver, lungs, and bone) have been obtained from each specimen. The remainder of the dissected carcasses were archived for future use.

Radiochemical analysis for ^{239,240}Pu is in progress with 46 samples completed. Only 13 samples have shown any levels of Pu above detection limits. They were large samples of hide and muscle from a 13-lined ground squirrel (*Spermophilus tridecmlineatus*), the hide samples of voles (*Microtus ochrogaster*), Hispid pocket mice (*Perognathus hispidus*), and deer mice (*Peromyscus maniculatus*). To date only pelts have shown any detectable levels of Pu other than the one muscle sample. Our findings are consistent with the previous work (Little 1976) which concluded that a very small portion of the total Pu in the Rocky Flats ecosystem was associated with the small mammal tissues.

We are pooling like tissues to increase sample size prior to Pu analysis. Analysis of the remaining samples is continuing.

IV. REFERENCES

EG&G, Inc. 1990. An aerial radiological survey of the United States Department of Energy's Rocky Flats Plant. EG&G, 10617-1044, May 1990.

Little, C. A. 1976. Plutonium in a grassland ecosystem. Colorado State University, Fort Collins, Colorado. Dissertation 1976.

Little, C. A., F. W. Whicker, and T. F. Winsor. 1980. Plutonium in a grassland ecosystem at Rocky Flats. J. Environ. Qual. 9(3):350.

Whicker, F. W., S. A. Ibrahim, A. W. Alldredge and L. Fraley, Jr. 1990. Progress report on radioecological investigations at Rocky Flats. Colorado State University, Fort Collins, Colorado, September 15, 1990.

Table 1. Summary $^{239,240}Pu$ concentrations for vegetation specimens. The minimum detectable concentration was approximately 850 mBq kg^{-1}

SAMPLE	MEAN $^{239,240}Pu$ CONCENTRATION mBq kg^{-1} (± 1 SD)	CV
Blue grama	306 ± 542	1.77
Uncovered cores	7750 ± 10400	1.34
Covered cores	104 ± 271	2.61

Table 2. Estimated mean $^{239,240}Pu$ soil concentration for core specimens and mixed soil containers based upon ^{241}Am measurements.

SAMPLE	MEAN $^{239,240}Pu$ CONCENTRATION kBq kg^{-1} (± 1 SD)
CORE	4.2 ± 1.2
MIXED (B)	8.1 ± 1.3
MIXED (S)	8.6 ± 0.8

B = Blue grama
S = Standard

Table 3. Mean $^{239,240}Pu$ UR and CR values for Blue grama, uncovered core, and covered core specimens.

SAMPLE	Mean $^{239,240}Pu$ UR or CR (± 1 SD)	CV
Blue grama UR	$3.8 \times 10^{-5} \pm 7.2 \times 10^{-5}$	1.91
Uncovered core CR	$1.9 \times 10^{-3} \pm 2.6 \times 10^{-3}$	1.39
Covered core UR	$2.5 \times 10^{-5} \pm 6.1 \times 10^{-5}$	2.4

$$CR = \frac{\text{Radionuclide concentration in vegetation (Bq/kg)}}{\text{Radionuclide concentration in soil (Bq/kg)}}$$

$$UR = \frac{\text{Radionuclide concentration in vegetation via roots (Bq/kg)}}{\text{Radionuclide concentration in soil (Bq/kg)}}$$

Table 4. Preliminary results of the intercomparison of radionuclide measurements in marine sediments. IAEA samples IAEA-367 and IAEA-368.

	$^{239,240}Pu$ Activity (Bq kg^{-1})	
	IAEA-367	IAEA-368
Number of reports	25	26
Maximum value	54.7	51.1
Minimum value	24	18.5
Average	38.8	32.1
Relative Standard Dev. (%)	7.5	19.5
Median	38.2	27.4
CSU Average	39.8	27.4
CSU Relative Standard Dev. (%)	8.2	3.8

Table 5. Small mammal trapping success from Macroplot 1 and Macroplot 2.

Species	Macroplot 1	Macroplot 2
Peromyscus maniculatus (Deer Mouse)	23	25
Perognathus hispidus (*Hispid Pocket Mouse*)	4	0
Mus musculus (House Mouse)	1	1
Microtus ochrogaster (Vole)	2	0
Spermophilus tridecemlineatus (13-lined Ground Squirrel)	1	0
Total	31	26

Figure 1. Estimated $^{239,240}Pu$ soil concentration as a function of the layer measured for the eight soil-vegetation core specimens.

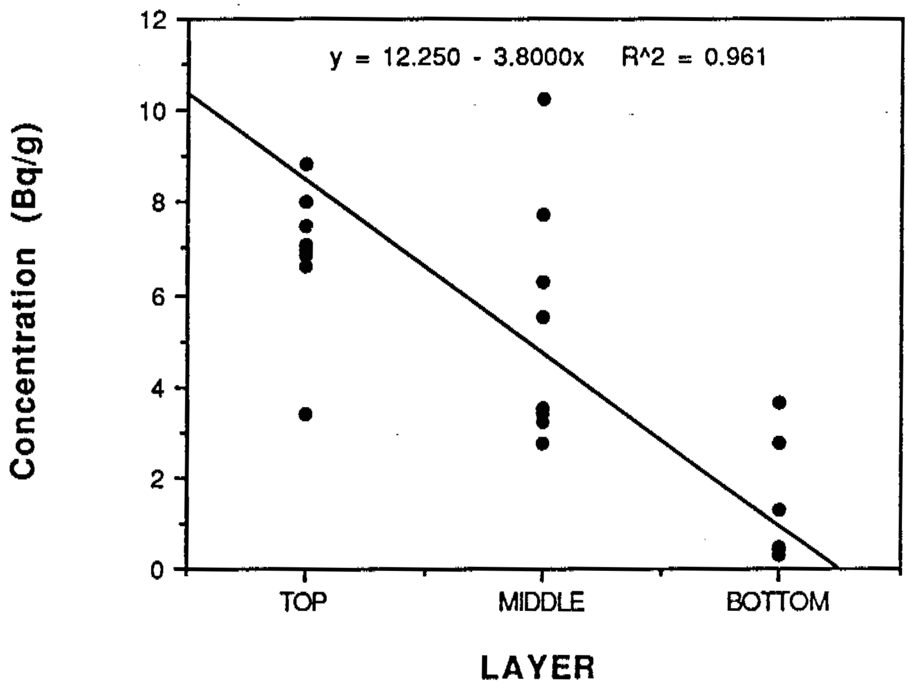

Figure 2. Map of the Rocky Flats Plant site showing CSU study plots.

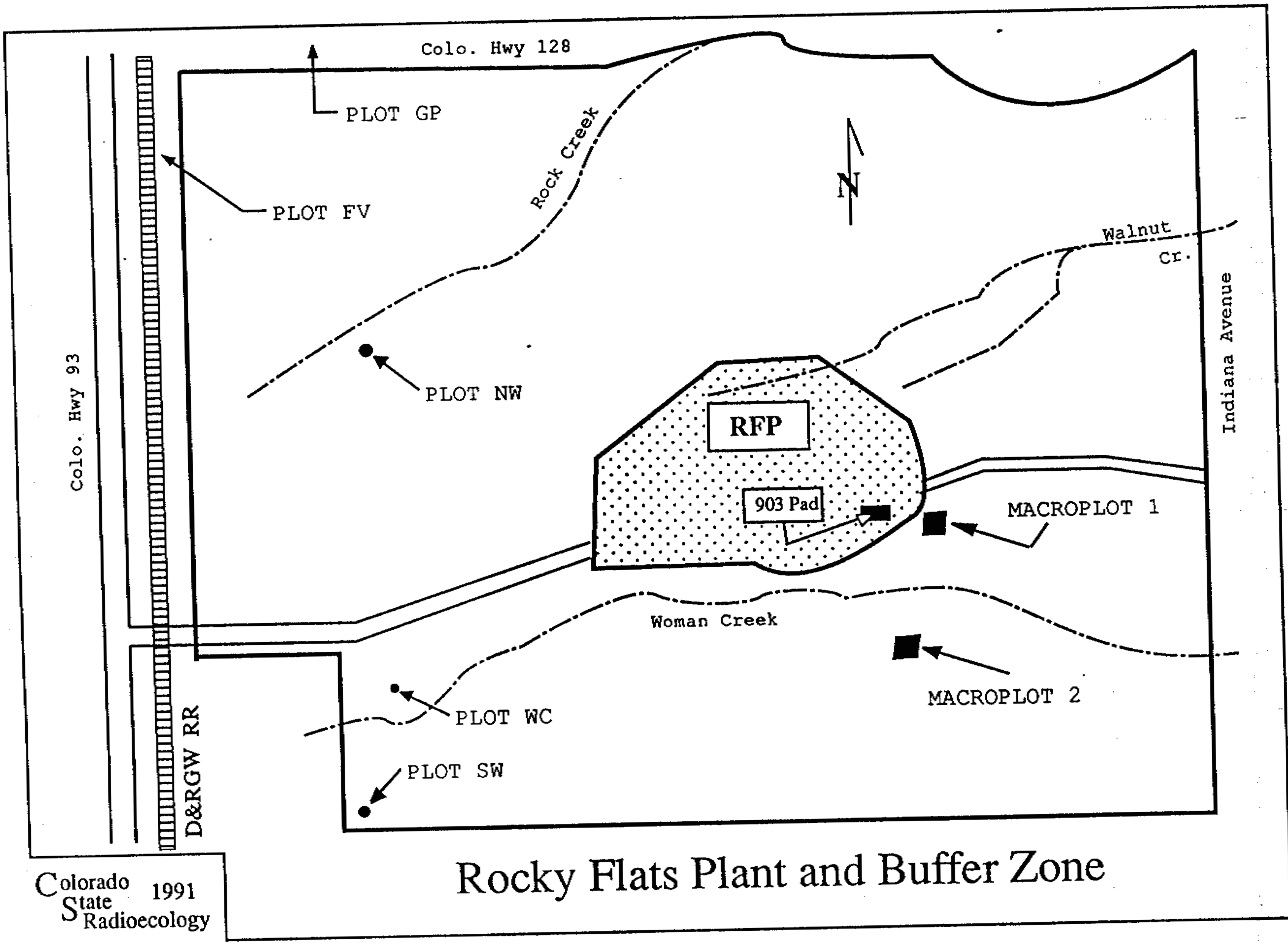

Figure 3. Plutonium concentration in vegetation and litter from Macroplot 1 for the ongoing and previous work.

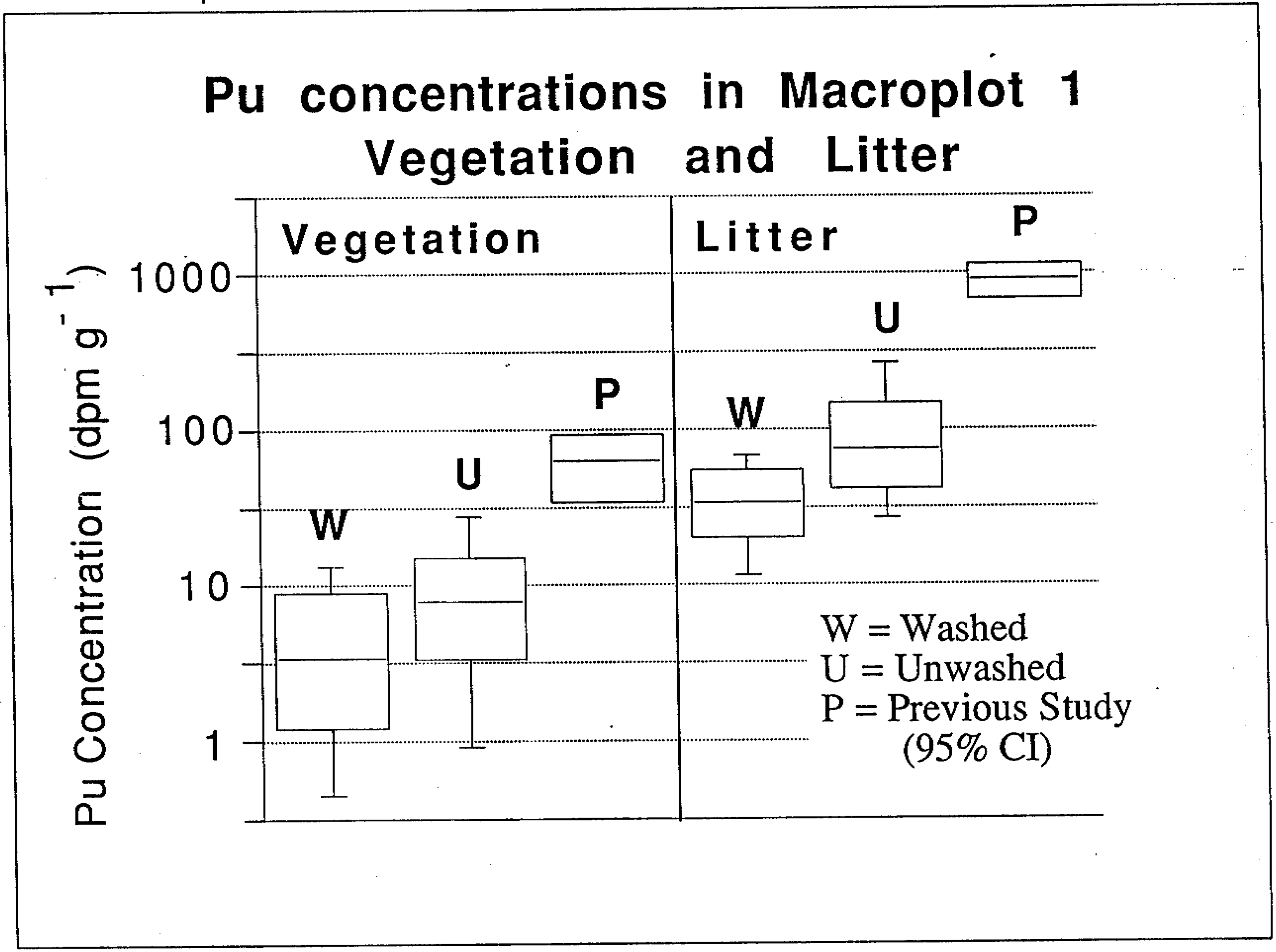

Figure 4. Plutonium concentration in vegetation and litter from Macroplot 2.

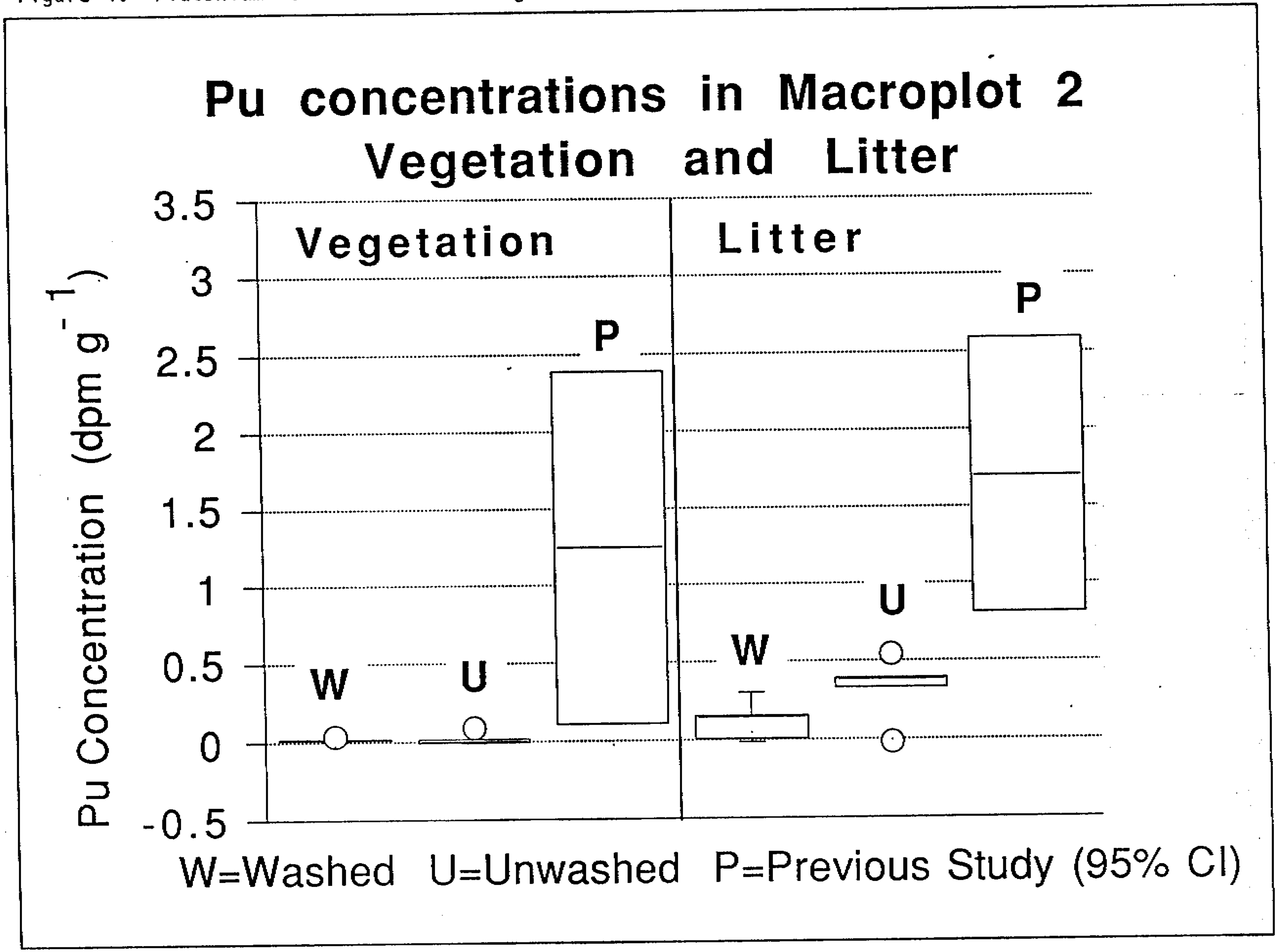

Figure 5. Plutonium ratios of unwashed to washed vegetation and litter for Macroplot 1.

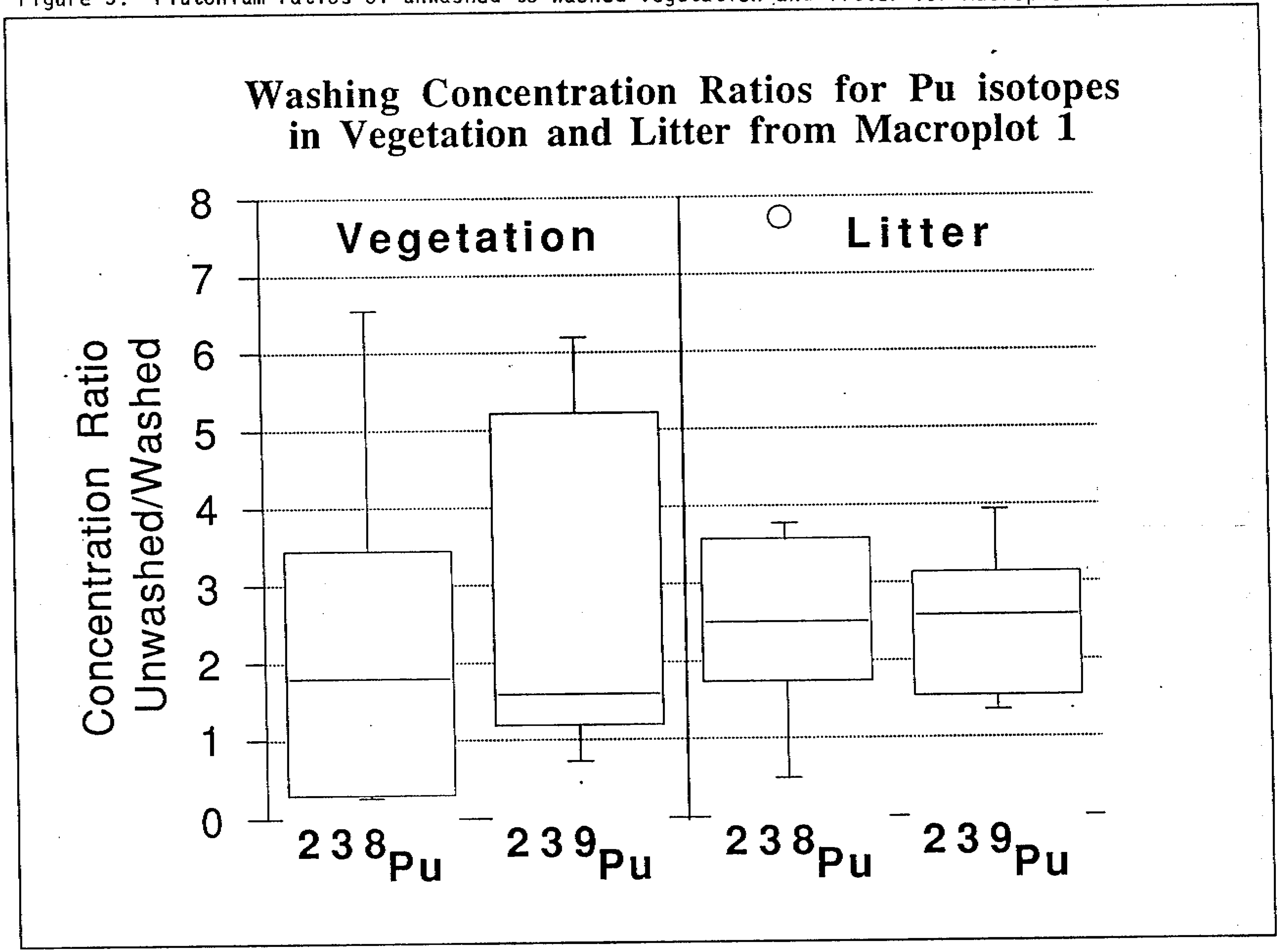

Figure 6. Ratios of $^{239,240}Pu$ to ^{238}Pu for Macroplot 1 vegetation and litter. The ongoing and previous work are compared.

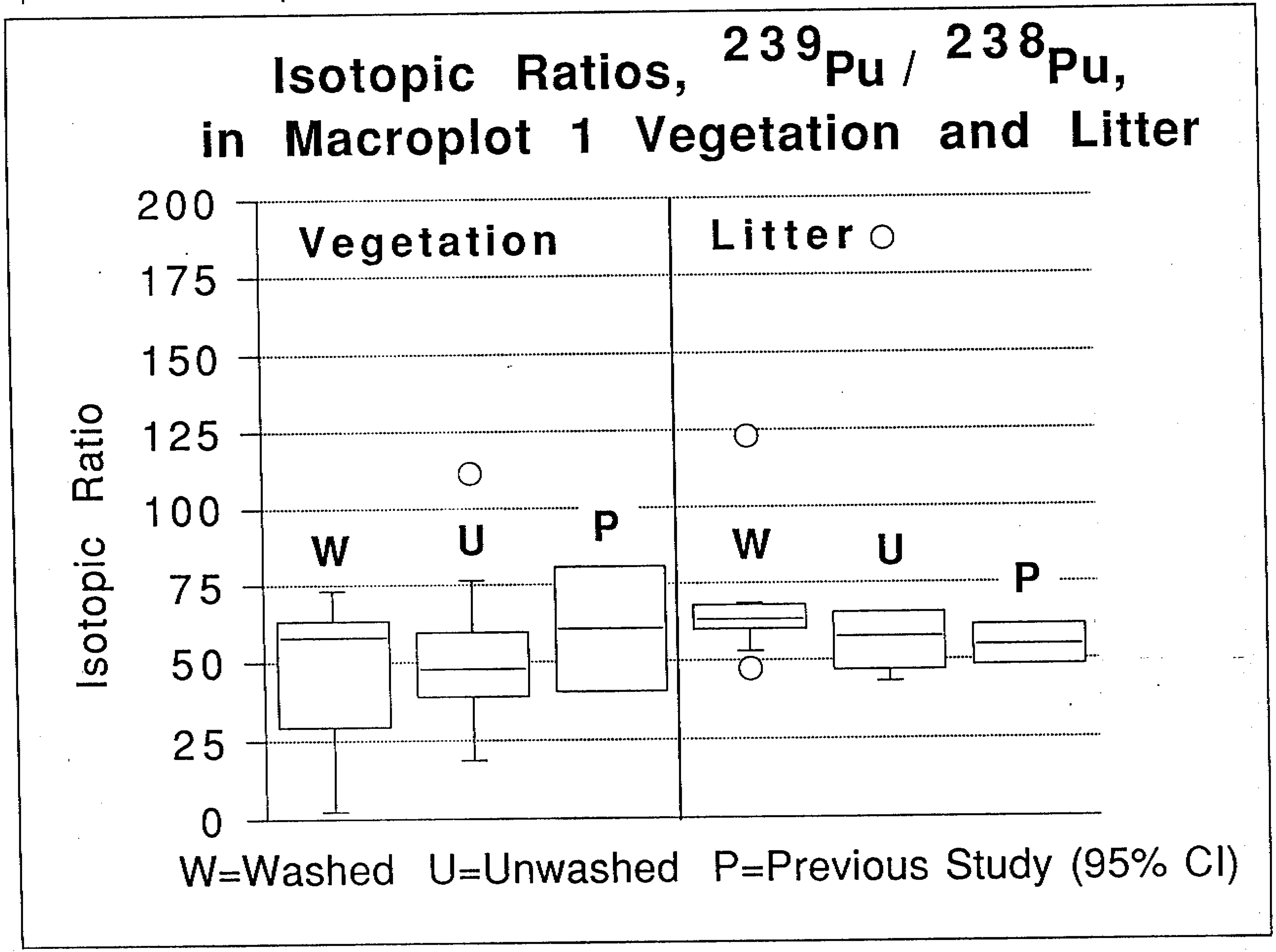

Figure 7. Plutonium concentrations in Macroplot 1 soil (< 2 mm size) by depth. Data from the previous study are compared to the current values.

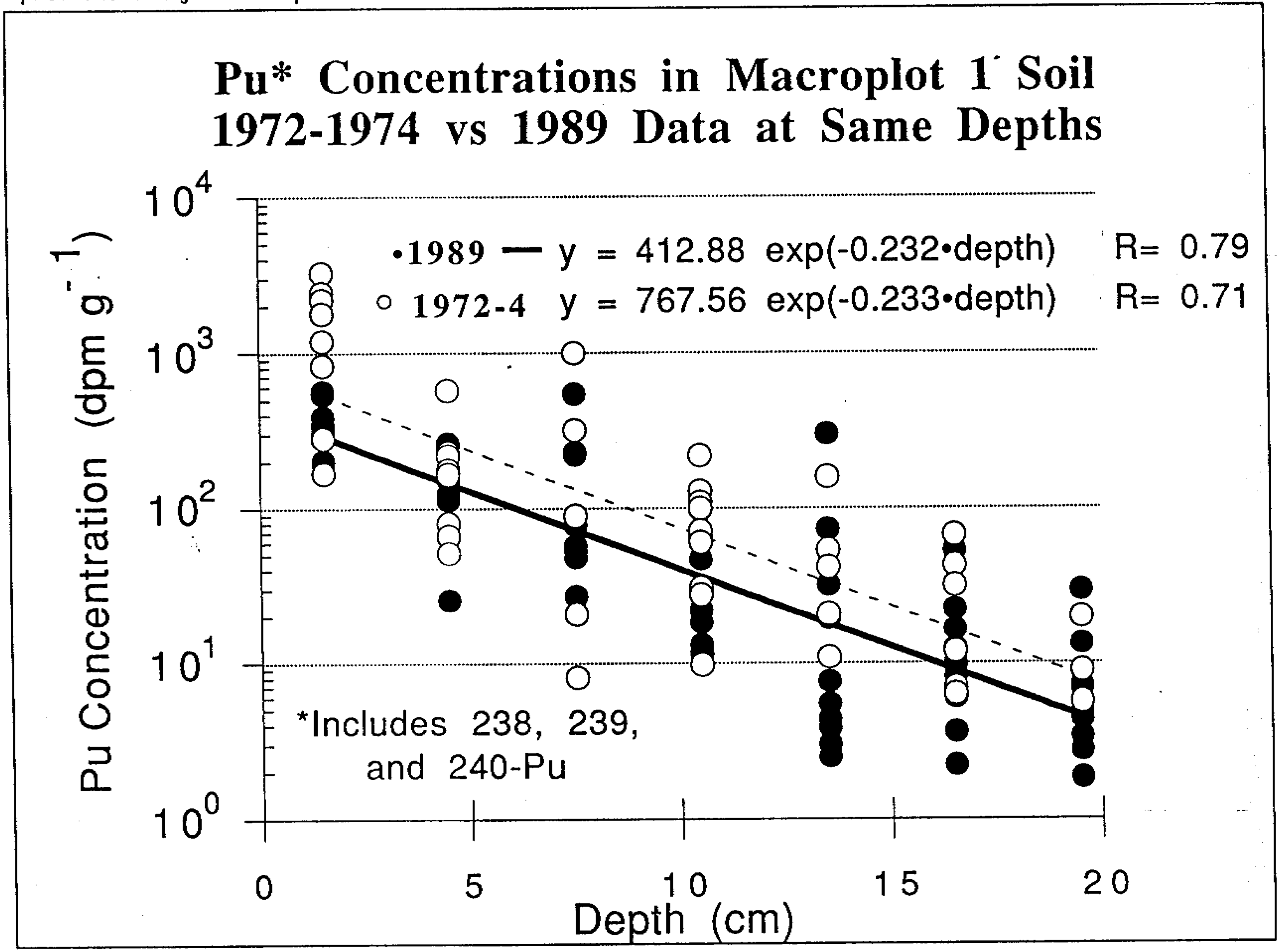

Figure 8. Isotopic ratios for Macroplot 1 soil (< 2 mm size).

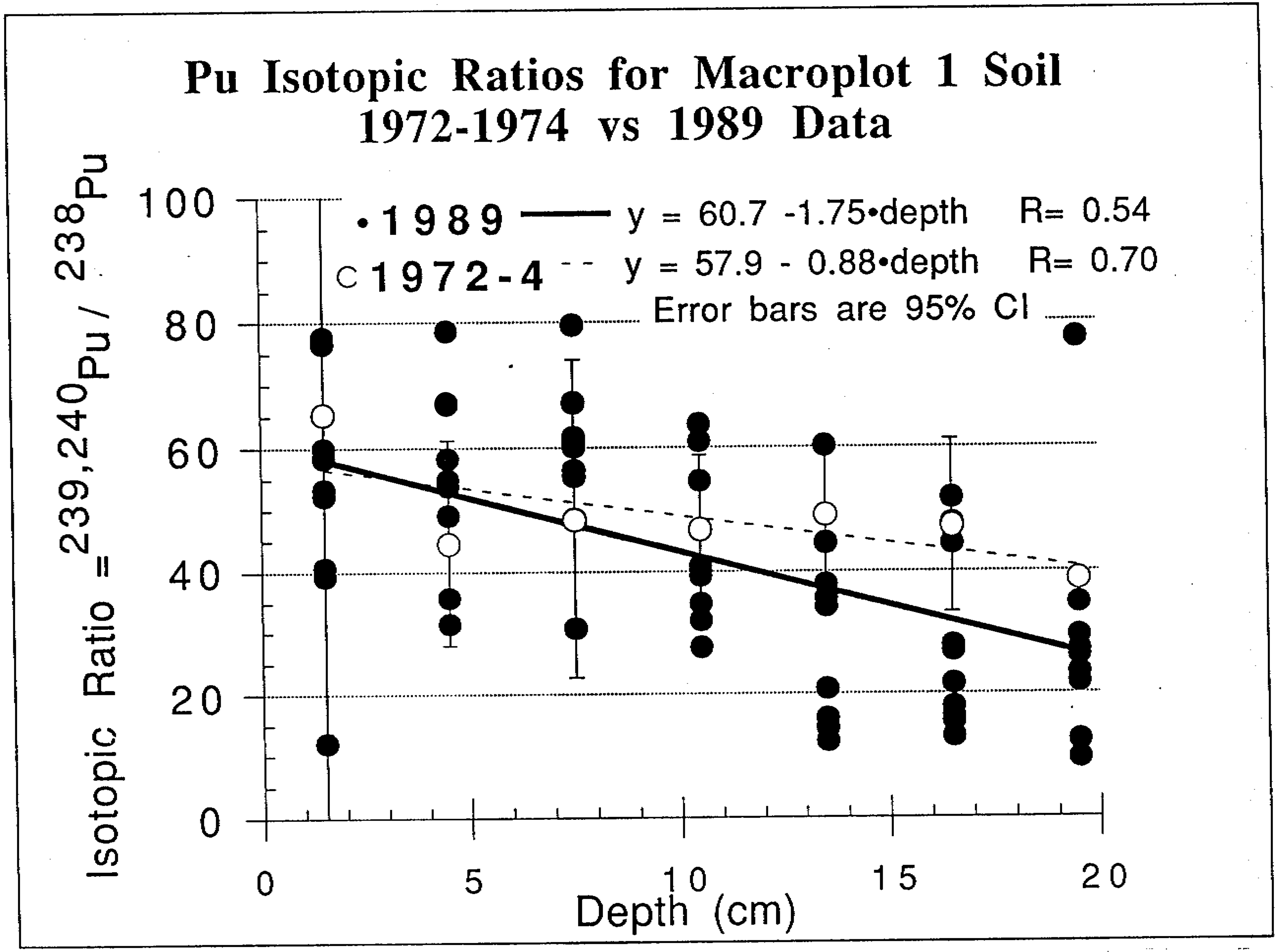

Figure 9. Regression of $^{239,240}Pu$ measurements versus ^{241}Am concentration in soil from Macroplot 1.

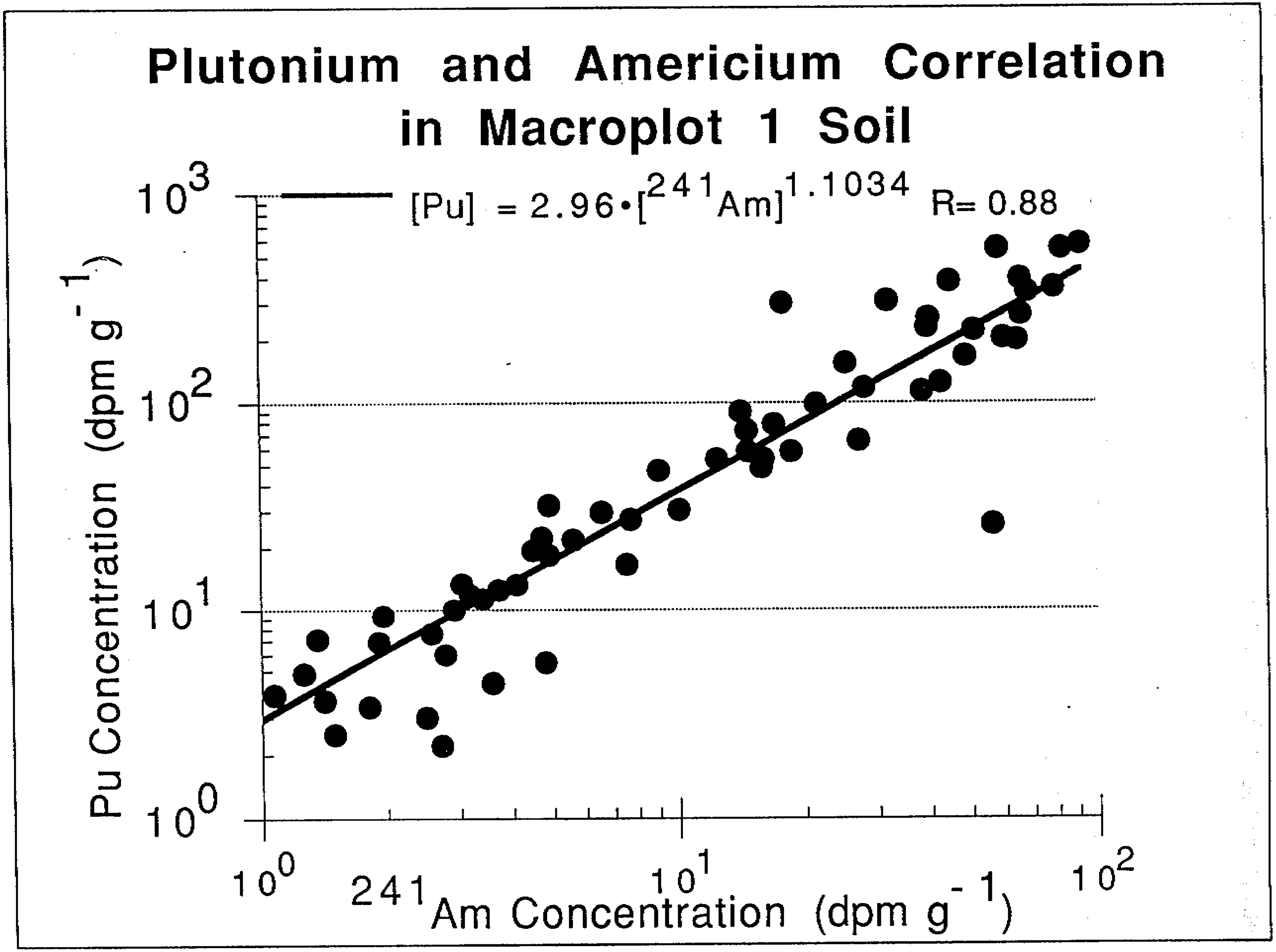

Figure 10. Plutonium concentrations in Macroplot 2 soil for the < 2 mm size. Data from the ongoing and the previous studies are compared.

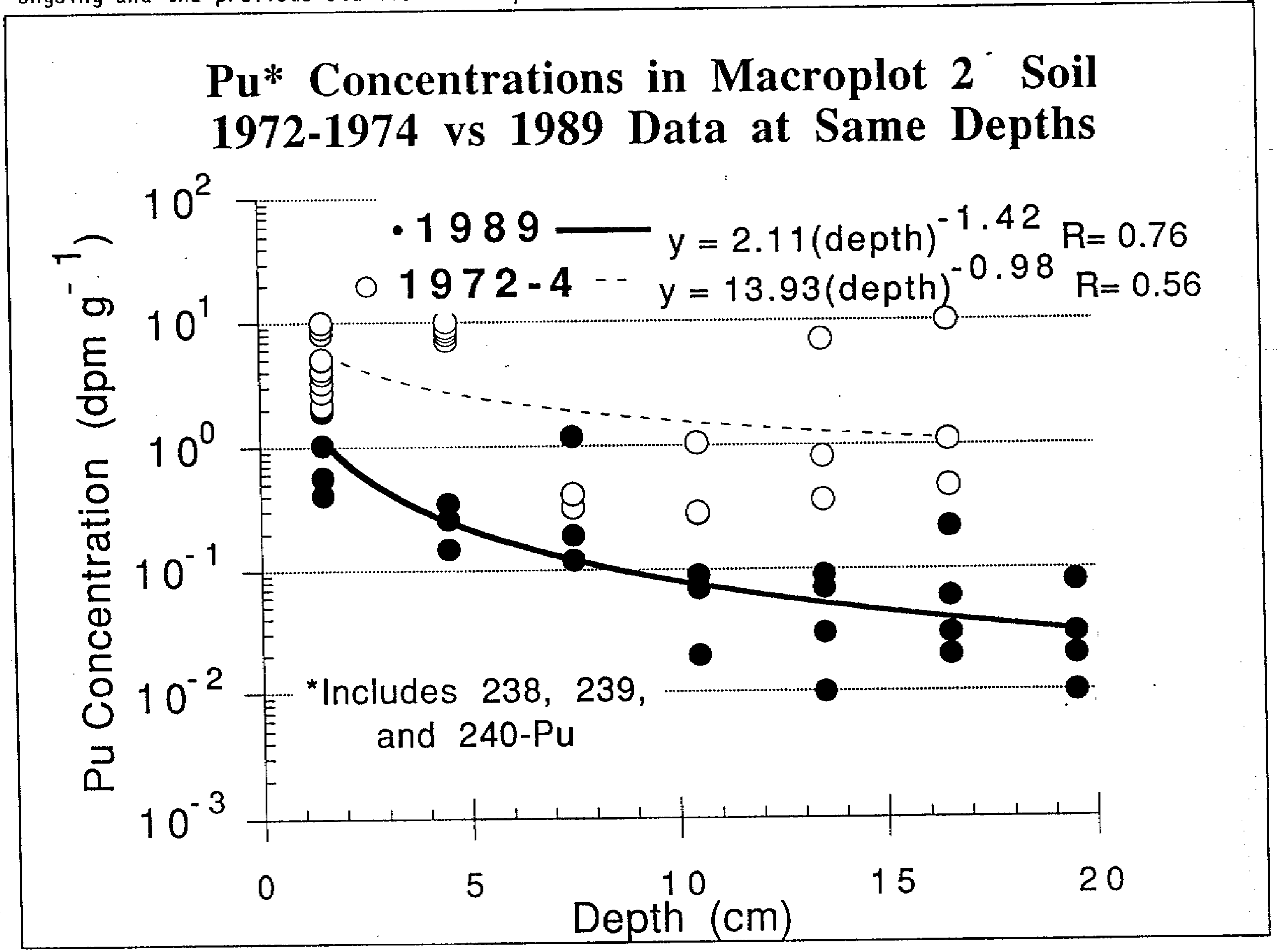

Figure 11. ^{241}Am concentration in soil from Macroplot 1 measured by gamma spectroscopy.

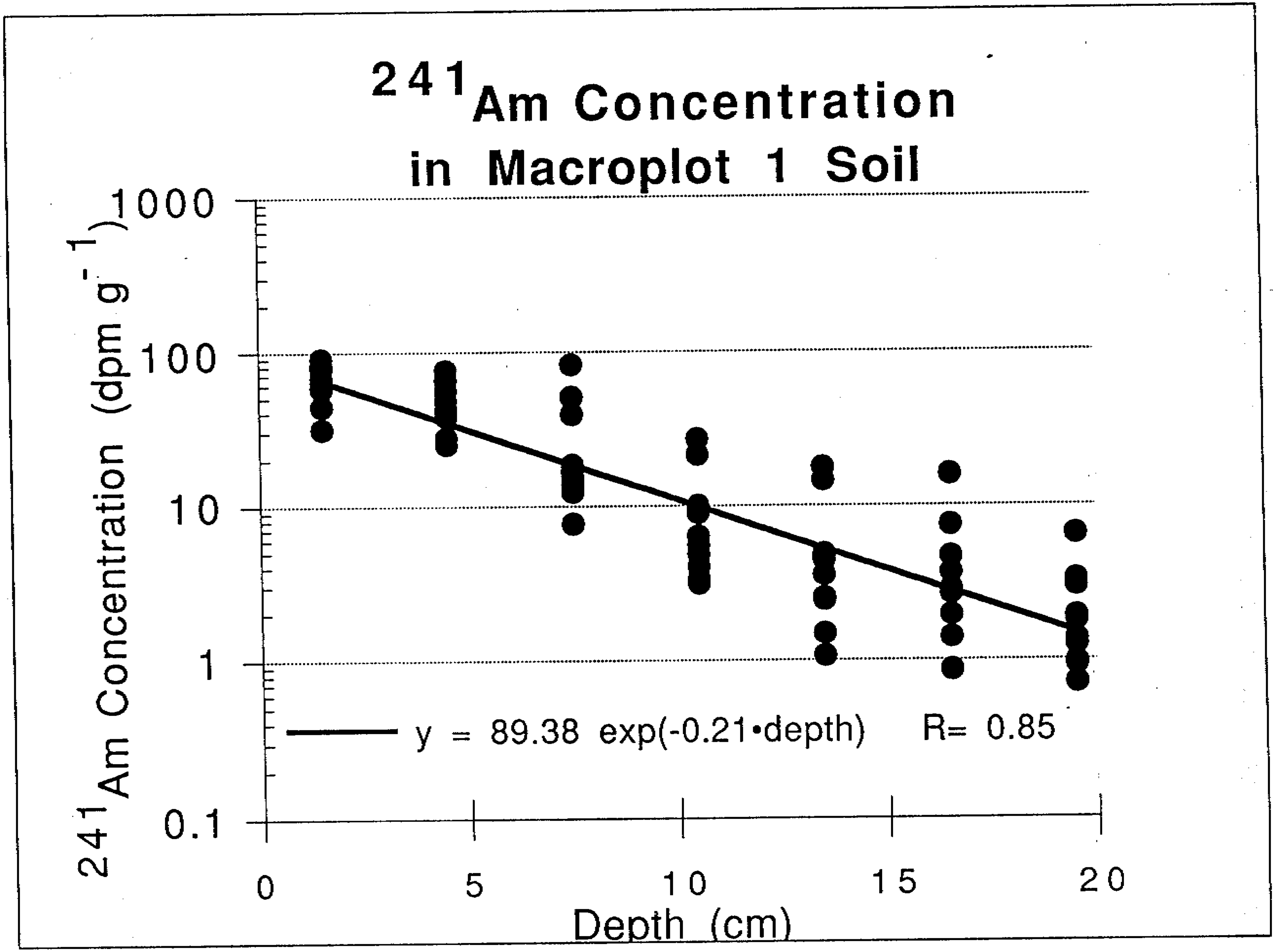

Figure 12. Gross alpha survey results for Rocky Flats soil.