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INFLUENCE OF PHOSPHOGYPSUM ON FORAGE YIELD AND QUALITY AND ON THE ENVIRONMENT IN TYPICAL FLORIDA SPODOSOL SOILS

Volume II

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Steven G. Richardson	**-Reclamation**
Gordon D. Nifong	**-Environmental Services**

Florida Institute of Phosphate Research
1855 West Main Street
Bartow, Florida 33830
(863) 534-7160
Fax: (863) 534-7165

INFLUENCE OF PHOSPHOGYPSUM ON FORAGE YIELD AND QUALITY AND ON THE ENVIRONMENT IN TYPICAL FLORIDA SPODOSOL SOILS

VOLUME II.

ENVIRONMENTAL ASPECTS ASSOCIATED WITH PHOSPHOGYPSUM APPLIED AS A SOURCE OF SULFUR AND CALCIUM TO BAHIAGRASS AND ANNUAL RYEGRASS PASTURES GROWING ON FLORIDA SPODOSOL SOILS

FINAL REPORT

FIPR Project 89-01-085

J.E. Rechcigl[1], I.G. Alcordo[1], C.E. Roessler[2], R.C. Littell[3]

[1]Range Cattle Research and Education Center
Institute of Food and Agricultural Sciences, Ona, FL 33865
[2]Department of Environmental Engineering Sciences
College of Engineering, Gainesville, FL 32611
[3]Department of Statistics
Institute of Food and Agricultural Sciences
Gainesville, FL 32611
UNIVERSITY OF FLORIDA

Submitted to the

FLORIDA INSTITUTE OF PHOSPHATE RESEARCH
1855 W. Main Street
Bartow, FL 33830

CONTRACT MANAGER:
Dr. Steven G. Richardson
Director, Reclamation Research Programs

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PERSPECTIVE

When phosphate ore is treated with sulfuric acid to produce phosphoric acid, which is used primarily in the manufacture of phosphate fertilizers, a calcium sulfate byproduct is also produced, known as phosphogypsum. More than 800 million tons of phosphogypsum have accumulated in Florida, and about 30 million tons are added annually. A high priority research topic at the Florida Institute of Phosphate Research has been to investigate potential uses of this byproduct in agriculture and industry. This project is one of several funded by the Institute to examine the use of phosphogypsum as an agricultural soil amendment. The first volume of the report gives the results of the authors' research on the agronomic benefits of using phosphogypsum as a source of calcium and sulfur for enhancing bahiagrass and ryegrass forage production on native sandy soils in central peninsular Florida. The second volume contains research results addressing the potential environmental impacts of applying phosphogypsum to the land. The environmental work has been expanded in a subsequent project, "Impact of Phosphogypsum on Radon Emissions and on Radioactivity and Heavy Metals in the Soil, Groundwater and Bahiagrass Forage" (FIPR Project # 92-03-038R), which is still in progress as this report goes to press.

The reader is also referred to additional FIPR reports on the subject:

Miller, W.P. 1989. Use of Gypsum to Improve Physical Properties and Water Relations in Southeastern Soils. FIPR Publ. No. 01-20-082. 42p.

Mullins, G.L. and C.C. Mitchell, Jr. 1990. Use of Phosphogypsum to Increase Yield and Quality of Annual Forages. FIPR Publ. No. 01-048-084. 56p.

Sumner, M.E., et al. 1990. Gypsum as an Ameliorant for the Subsoil Acidity Syndrome. FIPR Publ. No. 01-024-090. 56p.

Sumner, M.E. 1995. Literature Review on Gypsum as a Calcium and Sulfur Source for Crops and Soils In the Southeastern United States. FIPR Publ. No. 01-118-118. 89p.

An additional related topic is the reclamation of phosphogypsum piles, or "stacks" as they are commonly known in the phosphate industry. The following FIPR publication addresses this topic:

Richardson, S.G. 1995. Establishing Vegetation Cover on Phosphogypsum in Florida. FIPR Publ. 01-086-116. 60p.

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EXECUTIVE SUMMARY

Phosphogypsum (PE), a by-product of phosphoric acid manufacture, is primarily gypsum ($CaSO_4.2H_2O$). Mined gypsum as well as PG have been established to be good sources of S and Ca for crops (Shainberg et al., 1989; Alcordo and Rechcigl, 1993; and Alcordo and Rechcigl, 1995). Unlike mined gypsum, PG may contain As, Ba, Cd, Cr, Pb, Hg, Se, Ag, and F, any one of which can be toxic at high concentration levels. Phosphogypsum is also highly acidic (pH>2 to <5) compared with mined gypsum (pH=7), and the high solubility of PG (2.6 g L^{-1}) relative to that of calcium carbonate (0.015 g L^{-1}) may affect the quality of surficial groundwater in terms of electrical conductivity (E,) or total dissolved solids (TDS). However, it is primarily because of the presence of small amounts of radionuclides in PG, particularly radium-226 (^{226}Ra) which is the source of the gas radon-222 (^{222}Rn), that the United States Environmental Protection Agency (USEPA) has imposed severe restrictions on PG use.

Four field experiments were conducted using forage grasses. Two experiments (one agronomic-environmental and one solely agronomic) were conducted using Pensacola bahiagrass (Paspalum notatum Flugge) on an established pasture. Two similar experiments were conducted using annual ryegrass (Lolium multiflorum Lam.) on an annually seeded field. All four experiments were conducted to determine the effect of PG, applied as a source of S and Ca, on forage yields and quality. Non-radiological environmental and radiological data were collected from the agronomic-environmental experiments to evaluate the environmental impacts associated with PG use in agriculture on soil, groundwater, plant tissue, and on the emanation of ^{222}Rn to the atmosphere. The studies were conducted by the University of Florida (UF), Institute of Food and Agricultural Sciences (IFAS) at the Range Cattle Research and Education Center (RCREC) at Ona, Florida. The soils used were Florida Spodosols (Myakka series in the case of the bahiagrass field and Pomona series in the case of the annual ryegrass field). The experimental rates of 0.0, 0.4, 2.0, and 4.0 Mg (Mg = megagram = 10^6 grams) PG ha^{-1} were broadcast by hand over the experimental plots. In the bahiagrass experiments, the PG was not mixed with the soil but was left on the surface and allowed to dissolve and leach naturally down the soil profile by rain. In the ryegrass plots, the PG was mixed with the top 15-cm of the soil using a disk prior to seeding. The 0.4 Mg PG ha^{-1} treatment was applied annually for 3 years and the 2.0 and 4.0 Mg PG ha^{-1} treatments were applied only at the beginning of the study.

The experiments ran from 1990 to 1993. The radiological analyses, except for ambient Rn and gamma radiation, were performed by commercial environmental laboratories. The non-radiological analyses were done by various UF-IFAS laboratories. Ambient Rn and gamma radiation, which were measured using

electret ion chambers (EIC) were determined at the RCREC lab. All data were statistically analyzed by the Statistics Department of UF-IFAS.

This report (Volume II) covers the agro-environmental and radiological aspects of the study. The agronomic results are reported in Volume I.

Agro-environmental Aspects. The pH in water of the PG used in the study (1:1) was 4.6. The PG dissolved at a constant rate of 2.6 g L^{-1} in water and 4.3 g L^{-1} in a mixed acid solution of 0.025 M HC1 and 0.0125 M H_2SO_4 (Mehlich 1). It contained small amounts (mg kg^{-1}) of As (5.0), Ba (46.0), Cd (0.7-1.1), Cr (2.9), Pb (4.0), Hg (<0.01), Se (<0.05-1.6), and Ag (<0.2-2.0). Fluoride content was about 0.43 %. The levels of ^{226}Ra, ^{210}Pb, and ^{210}Po activities were 18, 31, and 24 pCi g^{-1}, respectively.

The heavy metals (HMs) As, Ba, Cd, Cr, Pb, Hg, Se, and Ag are used by USEPA to determine whether a solid waste is hazardous or not under its "toxicity characteristic" (TC) category based on each metal's leaching potential (LP) from a solid waste, determined according to USEPA procedure. Using the extensive data of May and Sweeney (1983) on measured LPs or TCs of the HMs in Florida PG samples from 9 PG stacks, the LP or TC for each heavy metal in any Florida PG may be estimated by:

Estimated LP or TC (unit: mg HM L^{-1} of leachate) = [(measured mg HM kg^{-1} of PG used in the study)/(measured avg mg HM kg^{-1} in Florida PGs with measured avg LP or TC)] x [measured avg LP or TC of Florida PGs].

The estimated LPs or TCs of the various HMs in the PG used in the study were 0.12 (As), 0.09 (Ba), 0.02 (Cd), 0.02 (Cr), 0.03 (Pb), <0.001 (Hg), <0.001 (Se), and 0.30 (Ag). Compared with the USEPA's TC limits of 5.0 (As), 100 (Ba), 1.0 (Cd), 5.0 (Cr), 5.0 (Pb), 0.2 (Hg), 1.0 (Se), and 5.0 (Ag) mg L^{-1}, the estimated LP values for any HM in the PG used in the study showed that the PG did not fall under the "toxicity characteristic" category of a USEPA hazardous waste.

The projected increases in heavy metal concentrations at the 0-15 cm surface soil (assumed bulk density = 1.5 g cm^{-1}) in the ryegrass plots due to application of 4.0 Mg PG ha^{-1} were estimated to be 0.010 (As), 0.092 (Ba), 0.002 (Cd), 0.006 (Cr) 0.008 (Pb), <0.0001 (Hg), 0.003 (Se), and 0.004 (Ag) mg kg^{-1} soil. Twelve months after PG application, Cd, Cr, Pb, Hg, and Se were determined in the soil profile down to 90 cm depth at 15-cm depth intervals. The measured ranges of concentrations in plots that received 4.0 Mg PG ha^{-1} for Cd, Cr, Pb, Hg, and Se were 0.04-0.10, 0.40-0.80, 0.14-0.58, 0.00-0.16, and 0.00-0.16 mg kg^{-1} soil, respectively. The statistics showed no difference between treatments at any soil depth. These indicated that the HM contaminants in PG used in the study would not present any real

short-term or long-term environmental concern for the soil.

Assuming that the LPs of the HMs in PG now mixed with the soil were the same as in pure PG, the TCs of the top 0-15 cm soil were estimated to increase by 0.0002 (As), 0.0002 (Ba), 0.00003 (Cd), 0.00003 (Cr), 0.00005 (Pb), 0.0000003 (Hg), 0.000006 (Se), and 0.0003 (Ag) mg L^{-1}. Relative to the USEPA primary standards for drinking water which are 0.05 (As), 1.00 (Ba), 0.01 (Cd), 0.05 (Cr), 0.05 (Pb), 0.002 (Hg), 0.01 (Se), and 0.05 (Ag) mg L^{-1} the projected increments are simply insignificant. Hence, it is concluded that the estimated in&eases in the LPs or TCs due to these heavy metal contaminants in PG would not present any impact, short- or long-term, on the quality of the surficial groundwater.

The statistical analysis of the pH of surficial groundwater from the bahiagrass experiment averaged annually (1990, 1991, 1992) and over the 3-year period (1990-1992) indicated no effect of PG treatments on the pH of surficial groundwater sampled at the surface (runoff) and at 60 and 90 cm depths. The annual average pH of runoff during 3 years of sampling ranged from 4.9 to 5.4 for the control plots and from 4.3 to 6.3 for all treated plots. Groundwater sampled at 60 cm depth had pH ranging from 4.5 to 4.8 for the control plots and from 4.6 to 5.6 for all treated plots. Groundwater sampled at 120 cm depth had pH ranging from 4.9 to 5.2 for the control plots and 4.0 to 5.8 for all treated plots.

The statistical analysis of the pH of groundwater samples averaged annually (1990-91 and 1991-92) and over the 2-year period (1990-91 to 1991-92) at 60 and 120 cm depths from the ryegrass experiment also showed no effect of PG applications. The annual average pH of groundwater from the control plots ranged from 4.0 to 4.4 at 60 cm depth and from 4.3 to 4.9 at 120 cm depth. For all treated plots, groundwater pH ranged from 3.7 to 4.4 and from 4.1 to 4.9 at 60 and 120 cm depths, respectively.

In the bahiagrass experiment, the highest annual (1991) average E_c of 1190 and 3-year average of 821 umho cm^{-1} were noted in groundwater sampled at 60 cm depth from plots that received 4.0 Mg PG ha^{-1} In the ryegrass experiment, the highest annual (1990-91) and 2-year average E_c of 1421 and 1137 umho cm^{-1}, respectively, were again noted in groundwater from plots treated with 4.0 Mg ha^{-1} sampled at the 60 cm depth. The highest value of 1421 umho cm^{-1} observed in the ryegrass experiment, however, was still less than the upper E_c limit of 1500 umho cm^{-1} for potable water in the United States.

Converting the highest E_c value of 1421 umho cm^{-1} into TDS (E_c x 0.66) gave 938 mg of dissolved solids L^{-1} which, again, was well below Florida's TDS standards of <3000 mg L^{-1} for water for agricultural use and <l000 mg L^{-1} for domestic and industrial uses.

In the case of F levels in groundwater from the bahiagrass experiment, the highest annual and 3-year average F levels in surficial groundwater from all depths were 0.83 and 0.36 mg L^{-1}, respectively. Both values were observed in water samples collected at 60 cm depth from plots that received 2.0 Mg PG ha^{-1}.

In the tilled ryegrass plots, the highest annual and 2-year average F levels in groundwater from all treated plots and for all depths (60 cm and 120 cm) were 0.53 and 0.43 mg L^{-1}, respectively. The highest annual average F levels in groundwater sampled at 60 cm and at 120 cm depths from the control plots were 0.31 and 0.19 mg L^{-1}, respectively.

It is widely accepted that approximately 1.0 Mg F L^{-1} in drinking water can effectively reduce dental caries without harmful effects on health. The Florida drinking water primary standards allow for a maximum contamination level (MCL) for F from 1.4 to 2.4 mg L^{-1}. None of the individual measured F values in the bahiagrass and ryegrass experiments exceeded the MCL values. The annual and the 3-year (bahiagrass) or 2-year (ryegrass) average values were even less than 1.0 mg F L^{-1}. Therefore, it is concluded that application of PG up to 4.0 Mg ha^{-1} to an established pasture or tilled land would not lead to unacceptable levels of F in surficial groundwater.

Radiological Aspects: Established Bahiagrass Pasture. Phosphogypsum containing 18, 31, and 24 pCi g^{-1} of ^{226}Ra, ^{210}Pb, and ^{210}Po, respectively, was applied to the experimental plots which also contained measured activities of 0.55, 0.61, and 0.53 pCi g^{-1} in the top 15-cm layer of untreated soil. It is calculated that each Mg PG ha^{-1} added 1820, 3080, and 2430 pCi m^{-2} and increased the activities in the upper 15-cm layer (assumed density 1.5 g cm^{-3}) by 0.008, 0.014, and 0.011 pCi g^{-1}. For the maximum treatment of 4.0 Mg ha^{-1}, the calculated increases in the initial activities of ^{226}Ra, ^{210}Pb, and ^{210}Po, averaged over the 15-cm layer, were 0.032, 0.055, and 0.043 pCi g^{-1}, respectively. These small additions to the natural activity in the soil could not be detected. An increase in ^{210}Pb due to the 4.0 Mg PG ha^{-1} treatment was detected in the top 15 cm layer in the first year of sampling, but it could not be detected in subsequent years. Any downward transport of the three radionuclides over the three years could not be detected.

There was no consistent correlation of gamma-radiation level with PG treatment. Any influence of PG at treatment levels up to 4 Mg ha^{-1} is insignificant in comparison to temporal and spatial variations in the local gamma-radiation background.

There was limited evidence of measurable increases in ^{226}Ra uptake by bahiagrass due to PG application. However, contributions by PG at the application levels used are small relative to the variation in the background level of ^{226}Ra and were difficult to measure. In regrowth forage, increases in ^{226}Ra uptake by bahiagrass during the first three years were estimated

to range from 0.01 to 0.02 pCi g^{-1} per Mg PG ha^{-1}. In hay forage, the data indicated an uptake in the order of 0.015 to 0.017 pCi g^{-1} per Mg PG ha^{-1}. No measurable effect of PG on ^{210}Pb uptake by bahiagrass was observed. For ^{210}Po, there was only limited evidence of increased uptake at the PG treatment levels used, and any contribution was small relative to the variations in the background levels. For ^{210}Pb and ^{210}Po, the data were insufficient to support calculation of an uptake factor.

Water sampling included runoff and also groundwater from 60-cm and 120-cm depths. ^{226}Ra appeared in runoff during the first three years after PG application. The data suggested, but did not confirm, an effect at 60 cm and 120 cm. However, the maximum activity observed in water, 1.8 pCi L^{-1}, was well below the current (3 pCi L^{-1}) and proposed (20 pCi L^{-1}) drinking water standards for ^{226}Ra. The data for ^{210}Pb in water were equivocal; there was very limited evidence for an effect of PG treatment on the well water. However, comparison of observed levels (up to 2.5 pCi L^{-1}) to proposed criteria (1 pCi L^{-1}) indicated that the potential for Pb in water deserves further evaluation. No effect of PG on ^{210}Po in groundwater could be detected by this experiment for treatments up to 4 Mg ha^{-1}. The maximum values for treated and control plots, 1.5 and 1.8 pCi L^{-1}, respectively, were less than the current 15 pCi L^{-1} standards for gross alpha activity in drinking water.

Radon flux is a sensitive parameter for evaluating Rn potential of land. No significant effects of the PG treatment on Rn flux were detected. Any increase attributable to PG treatment at levels up to 4 Mg ha^{-1} is insignificant in relation to the variations in the natural levels of Rn flux. Consequently, any effect on indoor Rn in future structures built over lands treated at this level will be insignificant relative to the variations experienced in indoor Rn levels.

Radiological Aspects: Tilled Land cropped to Annual Ryegrass. As with the bahiagrass experiment, the small additions of ^{226}Ra, ^{210}Pb, and ^{210}Po to the natural radioactivity of the soil could not be consistently detected. No PG-attributable increases in background gamma radiation could be detected.

As in the case with bahiagrass, there was limited evidence of measurable ^{226}Ra uptake by ryegrass forage related to the PG treatment; the uptake factor was estimated to be on the order of 0.039 pCi g^{-1} per Mg ha^{-1} for regrowth and mature forage. There was an indication but no statistically-significant evidence, of PG-attributable ^{210}Pb and ^{210}Po uptake for treatment rates up to 4 Mg ha^{-1}.

Groundwater at a 120-cm depth was sampled five times. The data suggested that PG treatment of the soil increased the ^{226}Ra activity in groundwater; however, effects were statistically significant only during the first year after treatment, and the

treated plot results did not correlate well with treatment level. The maximum ^{226}Ra activity, 2.35 pCi L^{-1}, was below the current and proposed drinking water standards of 3 pCi L^{-1} and 20 pCi L^{-1}, respectively. The treated plots tended to have higher activities of ^{210}Pb than the controls: however, after the first year there was no consistent, meaningful correlation of activity with treatment level. Comparison of the observed levels (up to 2.5 pCi L^{-1}) to proposed criteria (1 pCi L^{-l}) indicated that the potential for ^{210}Pb in groundwater deserves further attention. For ^{210}Po in ground water, no significant effects of PG treatment up to 4 Mg ha^{-1} were observed, and all activities were well below the current and proposed 15 pCi L^{-1} standards for gross alpha activity in drinking water.

Soil surface Rn flux was determined six times at the test plots. PG treatment increased Rn flux: the estimated contribution is on the order of 0.002 pCi m^{-2} s^{-l} per Mg PG ha^{-1}.

Ambient atmospheric Rn activities 1 m above the soil surface were determined 13 times after treatment application for periods ranging from 30 to 90 days. No effects of PG treatment were observed; however, Rn activity 1 m above an individual small plot is probably not representative of the Rn emission from that plot because of atmospheric mixing.

Overall, the radiological impact on the environment from application of Central Florida PG to tilled land at levels up to 4 Mg ha^{-1} was minimal when compared to levels and variations of the natural background. Phosphogypsum-attributable additions were typically at or below the limits of detection by contemporary conventional radiation and radioactivity monitoring techniques. Thus, it is recommended that the restrictions on agricultural use be relaxed for application to tilled land.

With regard to relational data for future assessments, quantitative relationships were obscured because of the low level of radioactivity involved and the natural variability of the parameters being measured. Therefore, it was recommended that further studies incorporate higher treatment levels, additional replication, and modified procedures in order to improve the statistical power, provide measures of quantitative relationships, and describe time-dependent behavior. Such a study (Phase II) using a maximum rate of 20 Mg PG ha^{-1} is presently in progress.

In view of the minimal environmental effects of PG application to either established bahiagrass pasture or tilled agricultural land cropped to annual ryegrass up to 4 Mg ha^{-1} of PG used in the study applied once in three years, it is recommended that restrictions on the agricultural use of such PG at less than 4 Mg PG ha^{-1} be relaxed. Application of PG with (23 pCi ^{226}Ra g^{-1} at 2 Mg ha^{-1} once every three years or 0.65 Mg ha^{-1} annually would be

comparable to the USEPA allowable rate of 2,700 pounds PG $acre^{-1}$ (3.024 Mg ha^{-1}) applied biennially (1.51 Mg ha annually) for PG having $\leq$ 10 pCi Ra g^{-1}.

INTRODUCTION

Phosphogypsum (PG), a by-product of the wet-process manufacture of phosphoric acid from phosphate rock, is primarily gypsum ($CaS0_4.2H_20$). Mined gypsum and PG have been used in agriculture as (1) sources of S and Ca for crops, (2) ameliorants for Al toxicity and subsoil acidity and infertility, (3) ameliorants for sodic and non-sodic dispersive soils, (4) conditioners for hard-setting clay soils and hardpans, (5) bulk carriers for micronutrients or fillers in low analysis fertilizers, (6) additives to modify cation-to-Ca ratios such as Mg:Ca ratio of soils, and (7) absorbents for NH_3-N in urea and other volatiles in manures (Shainberg et al., 1989; Alcordo and Rechcigl, 1993; and Alcordo and Rechcigl, 1995).

Naturally-occurring uranium (U) and its radioactive decay series are associated with phosphate mineral deposits. Consequently, the U-series member radium-226 (^{226}Ra) and its progeny appear in the PG. Radionuclide levels vary depending upon the source of the phosphate rock; PG derived from Central Florida rock contains ^{226}Ra at concentrations on the order of 21-33 pCi g^{-1} each (USEPA, 1978). Because of the radionuclide content, the U. S. Environmental Protection Agency (USEPA) allows only limited uses of PG (Federal Register, 1992). First, distribution for agricultural use is permitted if the certified average ^{226}Ra concentration does not exceed 10 pCi g^{-1}. This limit is intended to prevent unacceptable risks from indoor airborne radon or Rn[1] (a radioactive decay product of ^{226}Ra) and direct gamma radiation exposure in residences constructed on land previously treated with PG. Secondly, distribution of quantities up to about 320 kg (700 pounds) for research and development (R&D) is permitted. Thirdly, the USEPA may grant approval for other uses on a case-by-case basis if the proposed use will be at least as protective of public health as disposal of the PG in a stack or mine. The application for such approval must be accompanied by a proposed control program description and a risk assessment. Each of these uses has requirements for measuring the ^{226}Ra content of the PG and certification by the distributor; R&D and specifically-permitted uses also require record keeping by the end user.

The environmental impact of certain non-radioactive elemental impurities also needs to be considered. PG may contain as many as 50 elements originally present in various forms in rock phosphate (Borris and Boody, 1980). May and Sweeney (1980, 1983) detected 30 elemental impurities in Florida PGs using emission spectrographic analysis and 44 elements using neutron activation

[1] In this report, the term "radon" and "Rn" are used to designate the isotope ^{222}Rn.

analysis. Of major concern, however, are the eight heavy metals (As, Ba, Cd, Cr, Pb, Hg, Se, and Ag) that the United States Environmental Protection Agency (USEPA) uses to determine whether a solid waste is hazardous or not under its so-called "toxicity characteristic" category (USEPA, 1992).

Although of minor concern, fluorides (F) in PG, the usually low pH of PG, and the effect of the high solubility of gypsum itself on the electrical conductivity (E_c) and on total dissolved solids (TDS) may also have some impact on surficial groundwater quality.

The general objective of the project was to evaluate the effect of PG applied at agronomic rates as a source of S and Ca to agricultural crops and to assess the environmental impacts of the treatments. The specific objectives of the field experiments are given in the appropriate sections of the report.

Two fertility and two environmental field experiments involving the application of PG as a source of S and Ca to an established bahiagrass (Paspalum notatum Flugge) pasture and to annual ryegrass (Lolium multiflorum Lam.) were conducted during 1990 to 1993 by the University of Florida, Institute of Food and Agricultural Sciences (IFAS), Range Cattle Research and Education Center at Ona, Florida.

Volume I of the report covers the agronomic aspects (forage yields and quality), and Volume II covers the radiological and the non-radiological environmental aspects.

EXPERIMENTAL PLAN OF THE PROJECT

A. General and Specific Objectives of the Project

The general objective of the project was to evaluate the effect of PG as a source of S and Ca on forage yield and quality and to assess the environmental impacts of the treatments, at agronomic rates, on an established pasture (bahiagrass) and on a tilled land cropped annually to annual ryegrass.

The specific objectives of the environmental aspect of the study were:

(a) to evaluate the potential impact of the USEPA "toxicity characteristic" heavy metal impurities in PG on the soil and on the surficial groundwater;

(b) to determine the effect of PG on surficial groundwater quality in terms of pH, E_C, and F;

(c) to evaluate the potential radiological impact of the application of PG to an established pasture (bahiagrass) and to a tilled agricultural land cropped to annual ryegrass; and

(d) to develop data to support comprehensive assessment of the potential radiological impact through three potential exposure routes: (1) airborne Rn progeny inhalation - by evaluating soil ^{226}Ra (the Rn production source), soil surface Rn flux, and ambient airborne Rn; (2) radionuclides in the water and food pathway (ingestion) - by measuring ^{226}Ra and its two long-lived decay products, ^{210}Pb and ^{210}Po, in soil, water, and forage; and (3) external gamma-radiation exposure - by direct measurement.

B. Experimental Site, Design, and Treatments.

The bahiagrass experiment was conducted on an established Pensacola bahiagrass pasture on an area that sprawled over a Myakka (sandy, siliceous, hyperthermic Aeric Haplaquods) fine sand soil. A ground penetrating radar (GPR) was run over the site by personnel of the USDA Soil Conservation Service. The test established the presence of a hardpan or spodic horizon, characteristic of Spodosols, as a continuous layer between 60 to 120 cm beneath the surface. Also established by the test was the presence a broken argillic horizon just beneath the 120 cm depth.

The annual ryegrass experiment was conducted on virgin land that sprawled over Pomona (sandy, siliceous, hyperthermic Ultic Haplaquods) fine sand soil and Myakka fine sand. Soil core sampling established the presence of a hardpan or spodic horizon, characteristic of Spodosols, as a continuous layer between 60 to 120 cm beneath the surface. Also established by soil core

sampling was the presence a broken argillic horizon just beneath the 120 cm depth. The land was tilled and cropped to annual ryegrass (Gulf variety) each year for three years.

The rates of PG used, on a dry-weight basis, were 0.0, 0.4, 2.0, and 4.0 Mg PG ha^{-1} which were applied by hand to the plots. The 0.4 Mg PG ha^{-1} treatment was applied annually for three years while the 2.0 and 4.0 Mg PG ha^{-1} treatments were applied only at the beginning of the study. The PG contained 25% Ca and 19% S on a dry-weight basis.

A randomized complete block design with two replicates and experimental plots each measuring 32 m x 32 m were used. Fertilizers N (ammonium nitrate), P (triple superphosphate), K (potassium chloride), and a commercial micronutrient mix (2.4% B, 2.4% Cu, 14.4% Fe, 6.0% Mn, 0.06% Mo, and 5.6% Zn) were applied at the rate of 180, 45, 67.5 and 28.0 kg ha^{-1}, respectively, at the start of the growing season each year.

The first year of the bahiagrass experiment ran from July (first application of PG) to December, 1990 (last harvestable regrowth). The succeeding years (1991 and 1992) ran from March or April to December. For the annual ryegrass, the growing period in the locality runs from late October-early November (seeding) to March/April (last regrowth harvest) of the following year. For the environmental data, however, for both bahiagrass and ryegrass, the by-year (crop-year) analysis consisted of data collected within a 12-month cycle, from the beginning of one growing (bahiagrass) or seeding (ryegrass) season to next growing or seeding season.

C. Sample Collection, Analysis, and Field Measurements.

Sampling and measurements for both bahiagrass and ryegrass experiments included:

Soil sampling. Soil samples were collected at the end of the growing season each year to a depth of 90 cm at 15-cm intervals, air-dried and crushed to pass a 2-mm sieve.

Forage sampling. For regrowth forage, a portion of each plot was cut every 30 to 35 days and allowed to grow for subsequent sampling. For hay forage samples, a portion of each plot was left uncut and sampled near the end of the growing period (middle of autumn for bahiagrass and late February to early March for ryegrass) each year for three years. Tissue samples were oven-dried at 60°C and ground to pass a 0.84-mm sieve.

Runoff water sampling. Runoff was collected only from the bahiagrass experiment. Connected shallow ditches, 30 cm wide with the cut sloping outward from the ground level to a depth of 15 cm, were constructed at the edge of and around each plot. Runoff samplers, each consisting of two collector plastic

containers set one after the other, were installed at the lowest spot of the ditches surrounding each plot.

Runoff from the plot first drained into the ditches, then flowed toward the lowest spot on the ditches around the plot, and was collected in the first collector unit. The first collector unit, with inflow and outflow windows, was covered to keep the rain out to avoid dilution of the runoff. When the water level in the first unit rose to the level of the outflow window, free flow to the outside of the plot begun. A certain volume of the outflow passed through a rigid l-cm diameter plastic pipe placed at the outflow level parallel to the direction of the flowing water and into the second collector unit buried right next to the first. A breather consisting of a 1.25 cm diameter PVC pipe extending down to the bottom of the container and 30 cm above the ground was built into the second collector. This breather allowed the emptying of the collector using a siphon pump. The runoff collectors were emptied after each heavy rain.

Groundwater sampling. For subsurface water samples, wells were installed at the center of the plots at two depths. The wells consisted of 5.0 cm diameter PVC pipes with the lower ends joined to 30-cm long well tips that allowed percolating and/or standing subsurface water to flow into the pipes. The upper ends of the pipes extended 30 cm above ground and were loosely capped. The "shallow" wells had their water collection zones at 30 to 60 cm below the soil surface; those of the "deep" well at 90 cm to 120 cm. Duplicate "shallow" and "deep" wells were installed 2.5 m apart.

Beginning 1991, external control wells were set up 100 to 200 m from the nearest edge of the bahiagrass and the ryegrass plots to help determine possible contamination of the internal controls.

Wells were sampled or emptied using a siphon pump. Water samples were collected two to three days after each heavy rain during the first year and at longer intervals thereafter.

All water samples were immediately analyzed for pH, electrical conductivity (E_c), and F at the Center's laboratory. One-gallon water samples were prepared for storage (Standard Methods, 1985) for radionuclide analysis.

Radionuclide analysis. Phosphogypsum, soil, plant tissue, and water samples were sent to Core Laboratories of Casper, Wyoming for ^{226}Radium (^{226}Ra), ^{210}Lead (^{210}Pb), and ^{210}Polonium (^{210}Po) analysis according to procedures in use in that laboratory.

Radon flux measurement. Large-area activated charcoal canisters (LAACC) (Hartley and Freeman, 1985) were used to measure Rn fluxes emanating from the experimental pasture plots.

The grasses were first cut down to 2 cm from the ground and the activated charcoals in the canisters were set over these spots. The guidelines set by USEPA for Rn sampling over PG stacks (Federal Register, 1989) were strictly observed. The charcoals in the canisters were exposed for 24 hr to Rn emanating from the soil, then taken out from the canisters, placed inside plastic containers, sealed, and brought within hours to Pembroke Laboratory at Fort Meade, Florida which was contracted to do Rn analysis in accordance with USEPA procedure.

Gamma radiation and airborne Rn. Both gamma radiation and Rn gas concentrations were measured at the height of 1 m from the pasture surface of each experimental plot using electret ion chamber (EIC) (Kotrappa et al., 1988; Matuszek, 1990; Hopper et al., 1990; Rechcigl, et al., 1992). Wooden posts with wood crossbars were erected at the center of the plots. Four EIC units were hung on these crossbars 1 m above the ground. Three of the four EIC units were exposed to the atmosphere for Rn gas measurements and one EIC unit was placed inside a Rn-proof plastic bag for the gamma-radiation measurement. Since the EIC is an integrating device, gamma radiation and Rn measurements were interpreted in terms of the average exposure rate (uR hr^{-1}) and average concentration (pCi L^{-1}), respectively, over the deployment period. Deployment times ranged from 30 to 90 days, more or less. Duplicate external control stations 200 m apart, denoted as external control C_{ex1} were set up west of the experimental plots for each of the two studies. The external sampling stations were located 50 to 100 m from the edge of the nearest treated bahiagrass or ryegrass plot. A single external control station located about 3 km west of the bahiagrass experimental field but east of the ryegrass location served as a common external control C_{ex2} . Pre-PG application measurements were made at the general location of the bahiagrass experimental site but not at the ryegrass site.

Non-radiological analysis. Most of the non-radiological analyses were done at the UF-IFAS laboratories, unless indicated otherwise. The heavy metals in the soil were extracted using Mehlich 3 and analyzed using inductively coupled plasma (ICP) methods (Mehlich 3, 1984; Standard Methods, 1985). Fluorides in plant tissue, PG, and water were analyzed using F-electrode method (Woltz, 1980; Standard Methods, 1985). Water pH and E_c were determined using pH and conductivity meters, respectively. Except for Al which was determined using the aluminon method (McLean, 1965), the various elements for the PG solubility determination were analyzed using a Perkin-Elmer 3030B atomic absorption spectrometer (Alcordo and Rechcigl, 1992).

D. Statistical Analysis and Data Presentation

All statistical analyses were done by the UF-IFAS Department of Statistics. The data were first analyzed by individual collection or harvest (by-sampling), then averaged over PG

application period or crop year (by-year), and finally averaged over all collections or harvests over the 3-year period of the study (by-3-year analysis).

Treatment effects were analyzed using analysis of variance (ANOVA). Pair comparison analysis was also done using Duncan's Multiple Range test (DMR test at $P \leq 0.05$) (Gomez and Gomez, 1984).

Tests for linear (P{linear}) and quadratic (P{quad.}) effects ($P<0.10$ for both) for the first year data (1990) for by-sampling and by-year analysis were done using all four treatments. Tests for the subsequent years' data (1991 and 1992) for by-sampling, by-year, as well as the by-3-year analysis were done using only the three treatments of 0.0, 2.0, and 4.0 Mg PG ha^{-1}. The P values for treatment effects are presented in the tables, whether significant or not. The absence of P(linear), P(quadratic), or DMR test entries in the table indicates non-significance (ns) at the levels of significance indicated above.

Unless noted otherwise, data presented for individual sampling represent the average of the results from the two replicate plots for the particular treatment level.

RESULTS AND DISCUSSIONS

A. AGRO-ENVIRONMENTAL ASPECTS ASSOCIATED WITH PG AND PG APPLICATION TO AGRICULTURAL LAND CROPPED TO BAHIAGRASS AND ANNUAL RYEGRASS AS A SOURCE OF S AND Ca

A.1 Chemical Analysis and Solubility of PG and Some of Its Constituents

The PG used in the study was analyzed by two independent laboratories. The analyses are shown in Table A1. For purposes of comparison, the average concentrations of some of the elements in samples obtained from several Florida PG stacks as reported by May and Sweeney (1980, 1983) are also presented.

Chemical analysis of PG. Phosphogypsum, with about 25% Ca and 19% S, can be a major source of Ca and S for crops. The small quantities of the other essential macronutrients P, K, and Mg are not likely to make any significant contribution to crop needs. Also, the small quantities of the various essential trace elements present in PG are not going to meet the needs of crops. These small amounts of trace plant nutrients as contaminants in PG, even at large PG application rates, are also not likely to present any real environmental concern.

Table Al shows the presence of F and Al in concentrations which are much higher than most of the contaminants. Although both are known to become phytotoxic in large concentrations, only F may be of some real concern as its toxicity is known to affect animal life.

The elements which are of major environmental concerns are the so-called USEPA "toxicity characteristic" metals and radionuclides. Their concentrations are listed in Table A1.

Solubility of PG and of some of its constituents. The solubilities of PG used in the study and of the Ca/Mg, P, K, F, Al, and Fe constituents were determined by dissolving 4.0 g of moisture-free PG in 200 mL of deionized water and in acid solution (0.05 N HC1 + 0.025 N H_2SO_4, Mehlich 1, 1953, 1953a). Mehlich 1 was used because this was the solution used to extract the various soil plant nutrients in the study (Table A2).

Phosphogypsum dissolved at a constant average rates of 0.26 g in 100 ml in water and at 0.43 g in 100 ml Mehlich 1 (Table A2). The purity of PG, in terms of dissolved PG and Ca, was in excess of 90% on moisture-free basis.

Table A1. Chemical analyses of phosphogypsum (PG) used in the study and of other PG samples from Florida PG stacks.

Elements	Concentration (Unit)		
	U.S. Agri. Chem. Corp.[a]	Pembroke Laboratory[a]	Average in Florida PG[a]
Major Nutrient:	---------- (Percent) ----------		
Calcium (Ca)	24.5	25.6	26.2
Sulfur (S)	19.8	-[b]	19.5
Phosphorus (P)	-[b]	0.3	0.7
Minor:	---------- (mg kg^{-1}) ----------		
Iron (Fe)	690.0	460.0	860-1,000±300-600
Sodium (Na)	260.0	260.0	520±79
Potassium (K)	38.0	110.0	200-230±83-94
Molybdenum (Mo)	-[b]	17.0	2.2-11±1.4-2.2
Magnesium (Mg)	16.0	13.0	<940±<27
Boron (B)	-[b]	<10.0	<3.0
Zinc (Zn)	6.0	6.2	<340±21
Copper (Cu)	0.6	2.1	<82±<9.6
Manganese (Mn)	-[b]	1.9	25±14
Chloride (Cl)	-[b]	-[b]	<150±<4.7
Nickel (Ni)	-[b]	-[b]	<2.0
Phytotoxic:			
Fluorides (F)	4,300	-[b]	5,000
Aluminum (Al)	-[b]	1,100	2,000±540
USEPA toxic metals for "toxicity characteristic" determination:			
Arsenic (As)	5.0	5.0	0.76-0.9±0.26-0.3
Barium (Ba)	46.0	45.0	<210±<24
Cadmium (Cd)	0.7	1.1	3.4-4.0
Chromium (Cr)	-[b]	2.9	6.0±1.4
Lead (Pb)	-[b]	4.0	2.0-13.0
Mercury (Hg)	-[b]	<0.01	0.3-0.4±0.25-0.28
Selenium (Se)	<0.05	1.6	0.7-2.1±0.44-0.72
Silver (Ag)	2.0	<0.2	<1.3±<0.64
Radionuclides:[c]	---------- pCi g^{-1} ----------		
^{226}Ra		18.1[c]	21-33[d]
^{210}Pb		30.8[c]	26.4[e]
^{210}Po		24.3[c]	26.4[e]

[a]Moisture-free basis. [b]Not determined in the analysis; [c]As determined by Core Laboratory. [d]USEPA (1978); [e]USEPA (1991);

Table A2. Solubility of PG used in the study and of some of its major mineral constituents expressed in terms of their elements.

Solubility Run	PG	F	Ca	Mg	P	K	Al	Fe
Mehlich 1:	g $100mL^{-1}$	-- g kg^{-1} --			---------- mg kg^{-1} ----------			
1	0.43	2.94	60.9	1.5	305	44.7	5.6	39.3
2	0.42	1.25	55.2	1.0	284	11.6	7.3	32.5
3	0.44	0.21	58.5	2.0	283	8.6	7.8	35.7
4	0.43	0.21	55.1	1.7	245	7.3	7.4	26.3
5	0.07	0.22	3.3	1.7	17	4.2	4.7	15.3
6	0.07	0.01	2.3	1.3	8	5.5	3.3	5.7
Total dissolved[a]	1.86	4.63	235.0	9.2	1142	81.9	36.0	155.0
Total in PG[b]	2.00	4.30	256.0	13.0	2450	110.0	1100.0	436.0
% dissolved	93.0	108.0	92.0	71.0	47	74.0	3.3	34.0
Water:								
1	0.27	0.28	32.7	0.0	125	26.0	4.0	0.0
2	0.25	0.38	32.4	0.0	122	7.5	4.0	0.0
3	0.26	0.37	32.0	0.0	136	6.0	5.1	0.0
4	0.26	0.40	33.0	0.0	144	6.2	3.8	0.0
5	0.26	0.27	31.7	0.0	154	6.3	4.3	0.0
6	0.26	0.42	31.4	0.0	172	6.0	4.9	0.0
Total dissolved[a]	1.56	2.12	193.0	0.0	853	58.0	26.0	0.0
Total in PG[b]	2.00	4.30	256.0	13.0	2450	113.0	1100.0	460.0
% dissolved	78.00	49.00	75.0	0.0	35	53.0	2.4	0.0

[a]Total PG or element dissolved after solubilizing 4.0 g PG in 200 mL solvent by shaking the mixture for 1 hr at each run using an Eberbach reciprocating shaker at low speed.
[b]Total PG or total element in PG dissolved.

The F in PG ranged from 4000 to 5000 mg kg^{-1} or 0.4 to 0.5 % (Table A2). The F appeared to come from two sources with different solubility in Mehlich 1 - a highly soluble one dissolving at about 3.0 g kg^{-1} and the other at 0.21 g kg^{-1} of PG. Both sources appeared to have similar and constant solubility in water which was about 0.35 g kg^{-1}. The F-sources were totally soluble in acid (108 %) and only partially soluble in water (49 %). In view of the high solubility of the F-sources, hence F's high availability to plants and animals, F in PG would appear to present some environmental concern.

Aluminum in PG ranged from 1000-2000 mg kg^{-1} or 0.1 to 0.2 %. The Al-source was found to be highly insoluble in both water and acid solvents. Total Al that dissolved in water and Mehlich 1 averaged only 2.4 and 3.4 % of total Al, respectively, after 6 successive dissolution runs (Table A2). The insolubility of the Al source in PG even in such an acid medium as Mehlich 1 (pH <1.5) (Alcordo and Rechcigl, 1992) rules out any adverse effect of Al in PG on crops.

A.2. USEPA "toxicity characteristic" Metals and Their Leaching Potentials (LP) in PG

The elements which are used by the USEPA as indices of toxic metal contaminations of solid wastes are As, Ba, Cd, Cr, Pb, Hg, Se, and Ag (Table A3). The USEPA may classify such contaminated waste as a "toxicity characteristic" waste (USEPA, 1992) if the concentration of any one of the metals in the leachate - the leaching and the analysis of the metals being determined according to USEPA procedures - equals or exceeds the corresponding limits listed in Table A3.

For Florida PGs, extensive data on toxic metal concentrations and leaching potentials have already been collected and evaluated by May and Sweeney (1980, 1983). Their data are presented in Table A3 for reference. May and Sweeney's data are useful in determining the toxicity characteristic of the Florida PG used in the present study without doing the actual analytical USEPA procedure. Table A3 showed that none of the samples obtained from 9 PG stacks in Florida indicated a leaching potential of a USEPA "toxicity characteristic" waste. For the samples from 9 PG stacks, As, Ba, Cd, Cr, Pb, Hg, Se, and Ag concentrations (mg L^{-1}) in the leachates were found to be well below the USEPA contamination limits (in parenthesis), namely, 0.02 (5.0), 0.2 (100), 0.01 (1.0), 0.04 (5.0), 0.01 (5.0), 0.001 (0.2), 0.003 (1.0), and 0.06 (5.0), respectively (Table A3).

Table A3. USEPA heavy metals (HM) concentrations in leachate for "toxicity characteristics" (TC) for solid wastes,[a] their concentrations in PG samples from Florida PG stacks,[b] the measured "leaching potential" (LP) of PG for these metals, and the estimated LP of PG used in the study.

Sample		EPA "toxicity characteristics" metals							
		As	Ba	Cd	Cr	Pb	Hg	Se	Ag
Limits for USEPA TC solid waste:		---------- mg L^{-1} ----------							
		5.0	100	1.0	5.0	5.0	0.2	1.0	5.0
LP of Florida PG stack samples:									
Stack A:	1	0.02	0.2	0.01	0.04	0.01	0.000	0.002	0.08
	2	0.02	0.2	0.01	0.05	0.03	0.000	0.003	0.07
	3	0.03	0.0	0.01	0.07	0.01	0.000	0.004	0.01
Stack B:	1	0.01	0.0	0.01	0.01	0.00	0.001	0.003	0.04
	2	0.01	0.2	0.01	0.01	0.00	0.001	0.005	0.04
Stack C:	1	0.02	0.2	0.02	0.02	0.00	0.001	0.005	0.04
	2	0.01	0.4	0.03	0.05	0.00	0.001	0.003	0.10
Stack D		0.01	0.0	0.01	0.11	0.01	0.001	0.003	0.09
Stack E		0.01	0.2	0.03	0.03	0.03	0.001	0.004	0.06
Stack F		0.00	0.1	0.01	0.01	0.04	0.001	0.002	0.05
Stack G		0.01	0.3	0.01	0.01	0.01	0.001	0.002	0.04
Stack H		0.02	0.3	0.01	0.05	0.00	0.001	0.003	0.04
Stack I		0.02	0.2	0.01	0.02	0.01	0.004	0.002	0.07
Avg. stack LP:		0.02	0.2	0.01	0.04	0.01	0.001	0.003	0.06
Leachability of HM in PG samples from Florida PG stacks:									
		---------- mg kg^{-1} ----------							
Total TM in PG[c]		0.85	105	0.59	6.00	1.30	0.34	1.40	0.67
Amount Leached		0.24	3.2	0.28	0.80	0.36	0.01	0.06	0.96
% leachable:		28	3	47	13	28	4	4	-[d]
Estimated LP of PG used in the study:									
		---------- mg L^{-1} ----------							
Estimates:[e]		0.12	0.09	0.02	0.02	0.03	<0.001	0.003	0.30

[a]USEPA (199233) [b]May and [d]Sweeney (1983). [c]Averages for PG stacks A1, B1, E, F, and H. [d]Indeterminate (Total in PG and amount leached were 0.67 and 0.96 mg kg^{-1}, respectively, giving a leachability >100 %). [e]({HM in PG used}/{HM in PG stacks}) x Avg. stack LP.

The leaching potential (LP) for any HM in or the "toxicity characteristic" (TC) of any Florida PG may be estimated by using the average total concentrations of these metals measured in samples from 5 PG stacks and the average LP of these metals in 9 PG stacks samples (Table A3). In the case of the PG used in the study, the LP for any heavy metal will then be:

> Estimated leaching potential (LP) of metal in PG = [(Measured concentration of metal in PG used in the study)/(Average measured metal concentration in PG stack samples)] x Average measured stack sample LP for the metal.

The estimated LP values of the metals in the PG used in the study are thus found to be 0.12 (As), 0.09 (Ba), 0.02 (Cd), 0.02 (Cr), 0.03 (Pb), <0.001 (Hg), <0.001 (Se), and 0.30 (Ag). From these values, it is concluded that the PG used in the study, as with the other Florida PG's studied and evaluated by May and Sweeney, does not fall under the "toxicity characteristic" of a hazardous waste.

A.3. Measured Impact of PG on Toxic Metal Levels in Soil

The projected increases in toxic metal concentrations at the 0-15 cm surface soil due to PG application of 4.0 Mg ha^{-1} are given in Table A4. The measured concentrations of five heavy metals in a Florida Spodosol soil profile down to 90 cm depth determined at 15-cm depth intervals as influenced by four rates of PG applications up to 4.0 Mg ha^{-1} are presented in Table A5. The various amounts of PG were broadcast at the surface and then mixed with the soil to a depth of 15 cm. The land was then prepared for seeding with annual ryegrass. Soil samples were collected about 12 months after application. The statistics show no indication of any impact of PG on heavy metal concentrations at any PG rate and soil depth. There was also no consistent trend in toxic metal concentrations with PG rates for any of the metals. The measured concentrations of heavy metals in soil treated with PG up to 4.0 Mg ha^{-1} indicated that the heavy metal contaminants in PG used in the study do not present any short-term or long-term environmental concern to the soil.

Table A4. Projected increases in concentrations of toxic metals in agricultural land due to PG if applied to crops as a source of S and Ca.

Parameter	EPA Toxicity index metals							
	As	Ba	Cd	Cr	Pb	Hg	Se	Ag
	mg kg^{-1} PG							
Concentration in PG used:	5.0	46.0	1.1	2.9	4.0	<0.001	1.6	2.0
	mg kg^{-1} soil							
Projected[a] increase in the top 0-15 cm soil due to 4.0 Mg PG ha^{-1}	0.010	0.092	0.002	0.006	0.008	0.000	0.003	0.004
	mg L^{-1}							
Projected[b] Leaching Potential (LP) due to 4.0 Mg PG ha^{-1}	0.0002	0.0002	<0.0001	<0.0001	<0.0001	<0.0001	<0.0001	<0.0001

[a]Based on a soil bulk density of 1.5 g cm^{-3}.
[b]Based on the projected increases in the 0-15 cm surface soil.

Table A5. Measured concentrations of toxic metals in a Florida Spodosol amended with PG applied as a source of S and Ca to annual ryegrass after one cropping season, 1990-91, by depth.

Depth/ Treatment	EPA Toxicity index metals							
	As	Ba	Cd	Cr	Pb	Hg	Se	Ag
Measured concentration by depth:	mg kg^{-1} soil							
0-15 cm:								
Mg PG ha^{-1}								
0.0	-	-	0.020	0.020	0.520	0.040	0.000	-
0.4	-	-	0.020	0.040	0.420	0.040	0.000	-
2.0	-	-	0.000	0.040	0.560	0.020	0.000	-
4.0	-	-	0.040	0.040	0.580	0.000	0.000	-
Statistics:								
P	-	-	0.2918	0.5000	0.4915	0.8164	-[a]	-
15-30 cm:	mg kg^{-1} soil							
Mg PG ha^{-1}								
0.0	-	-	0.000	0.020	0.220	0.000	0.000	-
0.4	-	-	0.000	0.020	0.240	0.060	0.000	-
2.0	-	-	0.000	0.040	0.200	0.000	0.000	-
4.0	-	-	0.060	0.040	0.140	0.000	0.000	-
Statistics:								
P	-	-	0.5000	0.7082	0.2101	0.5000	-[a]	-

Continued to next page:

Table A5. Measured concentrations of toxic metals in a Florida Spodosol soil amended with PG applied as a source of S and Ca (Continuation).

Depth/ Treatment	EPA Toxicity index metals							
	As	Ba	Cd	Cr	Pb	Hg	Se	Ag
30-45 cm:								
Mg PG ha^{-1}								
0.0	-	-	0.040	0.100	0.400	0.180	0.140	-
0.4	-	-	0.000	0.020	0.220	0.000	0.000	-
2.0	-	-	0.000	0.040	0.140	0.000	0.000	-
4.0	-	-	0.020	0.040	0.180	0.000	0.000	-
Statistics:								
P	-	-	0.5000	0.6927	0.6778	0.5000	0.5000	-
45-60 cm:								
Mg PG ha^{-1}								
0.0	-	-	0.080	0.300	0.730	0.440	0.400	-
0.4	-	-	0.040	0.080	0.300	0.080	0.000	-
2.0	-	-	0.020	0.060	0.220	0.000	0.000	-
4.0	-	-	0.040	0.140	0.360	0.100	0.020	-
Statistics:								
P	-	-	0.6858	0.6019	0.6105	0.5713	0.5000	-

Continued to next page:

Table A5. Measured concentrations of toxic metals in a Florida Spodosol soil amended with PG applied as a source of S and Ca (Continuation).

Depth/ Treatment	EPA Toxicity index metals							
	As	Ba	Cd	Cr	Pb	Hg	Se	Ag
	mg kg^{-1} soil							
60-75 cm:								
Mg PG ha^{-1}								
0.0	-	-	0.100	0.300	0.720	0.460	0.440	-
0.4	-	-	0.060	0.180	0.560	0.300	0.200	-
2.0	-	-	0.020	0.100	0.340	0.060	0.040	-
4.0	-	-	0.100	0.220	0.460	0.100	0.160	-
Statistics:								
P	-	-	0.3999	0.7157	0.8914	0.7392	0.7345	-
75-90 cm:								
Mg PG ha^{-1}								
0.0	-	-	0.060	0.280	0.620	0.320	0.300	-
0.4	-	-	0.060	0.200	0.460	0.140	0.120	-
2.0	-	-	0.040	0.140	0.300	0.040	0.000	-
4.0	-	-	0.060	0.200	0.320	0.160	0.140	-
Statistics:								
P	-	-	0.8429	0.6065	0.3422	0.4452	0.3656	-

A.4. Heavy Metal Concentrations, pH, E,, and F Standards in Groundwater

Projected USEPA "toxicity characteristic" metals in surficial groundwater. For reference, Florida's primary drinking water standards or maximum contamination level (MCL) for heavy metals (which are the same as USEPA's) and F are given in Table A6.

Assume that the metals in PG now mixed with the top soil behave similarly as in pure PG. It is, however, more likely that their leachabilities are going to decrease rather than remain the same as in pure PG. Considering only the increases in concentrations due to 4.0 Mg PG ha^{-1}, the LP of the metals at the top 0-15 cm soil are estimated to be on the order of 0.0002 (As), 0.0002 (Ba), 0.00003 (Cd), 0.00003 (Cr), 0.00005 (Pb), 0.0000003 (Hg), 0.000006 (Se), and 0.0003 (Ag) mg L^{-1}. Hence, it is concluded that the concentrations of these USEPA "toxicity characteristic" heavy metal contaminants in PG used in the study, as with the other Florida PGs, do not present any reasonable environmental concern to surficial groundwater now or in the foreseeable future.

Table A6. Florida primary drinking water maximum contamination levels (MCL) standards[a] for F and heavy metals and their projected "leaching potential" (LP) in pure PG and in soil upon application at 4.0 Mg ha^{-1} rate.

Elements	Florida MCL standards	LP in PG (pure PG)	LP in PG (in soil)[b]
Inorganic:	---------- mG L^{-1} ----------		
As	0.05	0.12	0.0002
Ba	1.00	0.09	0.0002
Cd	0.010	0.02	<0.0001
Cr	0.05	0.02	<0.0001
Pb	0.05	0.03	<0.0001
Hg	0.002	0.00003	<0.0001
Se	0.01	0.003	<0.0001
Ag	0.05	0.30[c]	<0.0001
F	1.4-2.4	-	-

[a]Herr and Shaw (1989). [b]From 4.0 Mg PG ha^{-1} applied to the 0-15 cm topsoil. [c]Based on the higher determined value of 2.0 in the range of <0.02 to 2.0 mg kg^{-1} PG (See Table A1).

Impact on pH of groundwater. The pH is define as the negative logarithm of the H ion concentration (activity). The standard for Florida drinking water is a minimum pH of 6.5 measured at the point of collection (Herr and Shaw, 1989).

For the bahiagrass study, the by-sampling data for pH of surficial groundwater collected during the 3-year period are given in Tables IIAB-1, IIAB-2 and IIAB-3 (Appendix) for runoff, 60-cm, and 120 cm depths, respectively. In 1991, external control (C_{ex}) wells for the 60-cm and 120-cm depths were set up to check the readings of the internal controls.

For the control, runoff pH values ranged from 4.7 to 5.5 during the 3 years of sampling. For the treated plots, runoff pH ranged from 3.9 to 6.3. All by-sampling analysis during 3 years of sampling indicated no statistically differentiable effect of treatments on pH of runoff. The by-year and 3-year average analysis also indicated no effect of PG on runoff pH (Table A7).

Groundwater at 60 cm below ground surface was sampled three times in 1990, twice in 1991 and three times in 1992 (Table IIAB-2, Appendix). The by-sampling pH values for the internal control ranged from 4.3 to 5.6 and the treated from 4.1 to 7.1 during 3 years of sampling with no statistically measurable difference. The by-year and 3-year analysis also indicated no effect of treatment on runoff pH for the internal plots (Table A7).

Four samplings of surficial groundwater at 120 cm depth were done in 1990, three in 1991, and four in 1992. For the internal controls, the pH values ranged from 4.2 to 5.5; for the treated, pH ranged from 3.5 to 6.5 (Table IIAB-3, Appendix) during 3 years of sampling, with no significant differences, and no consistent trend as function of PG rates. The by-year and 3-year average data also showed no statistically measurable effect of PG on the pH of groundwater from a bahiagrass pasture sampled at 120 cm depth (Table A7).

In the ryegrass experiment no runoff was collected from the plots. For subsurface water, samples at 60 cm depth were collected twice during 1990-91 and 1991-92, but none during 1992-93. The by-sampling pH ranged from 3.7 to 4.7 for the control and from 3.7 to 4.6 for the treated plots (Table IIAR-1, Appendix). No significant differences among treatments were noted. However, samples from plots that received 4.0 Mg PG ha^{-1} showed depressed pH relative to those from the internal control in three out of four collections. Similar results were indicated in the by-year and 3-year average analysis (Table A8).

Table A7. pH of runoff and surficial groundwater from a bahiagrass pasture amended with PG as source of S and Ca, averaged by year and over a 3-year period, 1990-1992.

Depth(cm)/ Treatment	Crop year 1990	1991	1992	Mean
A. Runoff:	---------- pH ----------			
Mg PG ha^{-1}				
0.0(C)	5.4	5.3	4.9	5.2
0.4	6.3	5.9	5.9	6.0
2.0	5.5	5.3	5.4	5.4
4.0	5.9	4.3	5.0	4.9
Statistics:				
P	0.5680	0.1961	0.3589	0.3285
DMRT	ns[1]	0.4>4.0	ns	ns
B. 60 cm:	---------- pH ----------			
Mg PG ha^{-1}				
0.0(C_{ex})	-[a]	5.3	4.5	4.9
0.0(C)	4.6	4.8	4.5	4.7
0.4	4.6	4.6	4.6	4.6
2.0	5.1	5.6	5.3	5.3
4.0	4.7	4.9	5.2	4.9
Statistics:				
P(w/C_{ex})	-[a]	0.7647	0.7249	0.7646
P(w/o C_{ex})	0.6800	0.6650	0.4612	0.6407
C. 120 cm:	---------- pH ----------			
Mg PG ha^{-1}				
0.0(C_{ex})	5.5	6.2	5.6	5.8
0.0(C)	5.2	4.9	5.1	5.1
0.4	5.8	5.3	5.5	5.6
2.0	5.2	4.5	4.9	4.9
4.0	4.9	5.3	4.0	4.8
Statistics:				
P(w/C_{ex})	0.5253	0.0493	0.2499	0.2587
P(w/o C_{ex})	0.4698	0.2404	0.2091	0.3388

[1]ns=not significant. [a]No external control wells at this time.

Table A8. pH of surficial groundwater from an annual ryegrass pasture amended with PG as source of S and Ca, averaged by crop year and over 3 crop years, 1990-91 to 1992-93, by depth.

Depth (cm)/ Treatment	Crop year			
	1990-91	1991-92	1992-93	Mean
A. 60 cm:	---------- pH ----------			
Mg PG ha^{-1}				
0.0(C)	4.4	4.0	-[a]	4.2
0.4	4.3	3.7	-[a]	4.0
2.0	4.4	3.9	-[a]	4.1
4.0	4.1	3.8	-[a]	4.0
Statistics:				
P	0.7283	0.9287	-[a]	0.8725
B. 120 cm:	---------- pH ----------			
Mg PG ha^{-1}				
0.0(C_{ex})	4.7	4.6	4.3	4.6
0.0(C)	4.9	4.4	4.3	4.6
0.4	4.9	4.3	4.1	4.5
2.0	4.6	4.3	4.2	4.4
4.0	4.7	4.2	4.1	4.4
Statistics:				
P(w/C_{ex})	0.8834	0.9622	0.9368	0.9795
P(w/o C_{ex})	0.8054	0.9625	0.9353	0.9542

[a]No water in the wells.

Groundwater samples from 120 cm depth were collected nine times from April, 1991 to February, 1993 (Table IIAR-2, Appendix). The pH values ranged from 4.1 to 5.1 for the internal control and from 4.0 to 5.6 for the treated plots. Only in one instance (April 26, 1991) was there a slight evidence of differences (P=0.0291) among treatments, with pH linearly (P{linear}<0.05) decreasing with PG rates. By-crop year and 3-crop year analysis, however, indicated no effect of PG on groundwater pH at 120 cm depth (Table A8).

Impact on E_c of groundwater. The E_c is a measure of the quantity of electricity transferred across a unit area per unit potential gradient per unit time. For water, it is related to the amount of charged particles dissolved in it. For potable waters in the U.S., the E_c ranges generally from 50 to 1500 umho cm^{-1} (Standard Methods, 1985). The E_c is converted to another quantity "TDS" or "total dissolved solids" by multiplying the measured E_c by a given factor. In this study the E_c, in micromho per cm (umho cm^{-1}), may be multiplied by 0.66 to get a

corresponding value for TDS. The TDS represents all the solid minerals in solution except suspended solids, colloids, or dissolved gases. For Florida, the recommended TDS for drinking water is < 500 mg L^{-1}. For domestic and industrial use and for agricultural use the TDS should be <1000 and <3000 mg L^{-1}, respectively (Herr and Shaw, 1989).

The by-sampling data for runoff for the bahiagrass experiment over the 3-year period indicated ranges of E_c values from 132 to 430 and from 99 to 624 umhos cm^{-1} for the control and the treated plots, respectively. Although not significant, 3 out of 5 runoff samples collected over the 3-year period indicated elevated E_c in runoff from plots treated with 4.0 Mg ha^{-1}. Two samples (October, 1990 and July, 1992 both with P{linear}<0.05) showed increasing linear trend with PG rates. The July, 1992 sample showed slight (P=0.0632) evidence that application of 4.0 Mg PG ha^{-1} would raise the E_c of the runoff over those from plots that received 2.0 or less Mg PG ha^{-1} (Table IIAB-1, Appendix). The by-year analysis of the runoff data indicated evidence of a linearly increasing E_c (P{linear}=0.0449) trend with PG rates in 1990 but none in the other years. When averaged over 3 years, the results showed no effect of PG on E_c of runoff (Table A9).

During three years of sampling, the highest E_c of groundwater sampled at 60 cm depth were 357, 444, and 1583 umho cm^{-1}, for the external control, the internal control, and the treated plots, respectively. Excluding the external controls, 7 out of 8 groundwater samples collected over 3 years from plots that received 4.0 Mg PG ha^{-1} had elevated E_c relative to those from the control wells. Samples collected in October, 1990, June, 1991, and April, 1992 showed that E_c increased linearly (P{linear}<0.05) with PG rates (Table IIAB-2, Appendix).

The 1990 and the 1991 by-year analysis with P{linear} values of 0.0623 and 0.0179, respectively, as well as the 3-year analysis (P{linear}=0.00l4) provide strong evidences of E_c increasing linearly with PG rates in groundwater at 60 cm depth (Table A9). The ANOVA of the 1991 and the 3-year data also showed that the treatments increased the E_c of the groundwater with P values of 0.0439 and P=0.0040, respectively. The DMR test of the 3-year data showed that the 4.0 Mg PG ha^{-1} application, with E_c of 821 umho cm^{-1}, effectively raised E_c over all other rates with E_c ranging from 236 to 334 umho cm^{-1} (Table A9).

Table A9. E_c of runoff and surficial groundwater from a bahiagrass pasture amended with phosphogypsum (PG) as source of S and Ca, averaged by year and over the 3-year period, 1990-1992.

Depth(cm)/ Treatment	Crop year 1990	1991	1992	Mean
A. Runoff:	---------- umho cm^{-1} ----------			
Mg PG ha^{-1}				
0.0(C)	319.0	344.5	214.5	287.4
0.4	259.0	241.3	312.0	276.7
2.0	453.0	256.8	140.5	249.5
4.0	624.5	459.0	279.5	420.3
Statistics:				
P	0.1462	0.5829	0.1057	0.4651
P(Linear)	0.0449	ns	ns	ns
P(Quad.)	ns[1]	ns	0.0802	ns
DMRT	ns	ns	0.4>2.0	ns
B. 60 cm:	---------- umho cm^{-1} ----------			
Mg PG ha^{-1}				
0.0(C_{ex})	-[a]	295.0	176.8	235.9
0.0(C)	210.6	300.3	170.5	239.9
0.4	348.0	354.9	292.0	339.1
2.0	369.6	367.8	163.7	334.4
4.0	660.1	1190.3	364.5	821.1
Statistics:				
P(w/C_{ex})	-[a]	0.0343	0.6163	0.0041
P(w/o C_{ex})	0.1891	0.0439	0.5771	0.0040
P(Linear)	0.0623	0.0179	ns	0.0014
P(Quad.)	ns	ns	ns	0.0183
DMRT	ns	4.0>all	ns	4.0>all
C. 120 cm:	---------- umho cm^{-1} ----------			
Mg PG ha^{-1}				
0.0(C_{ex})	494.5	537.5	528.8	527.4
0.0(C)	170.5	253.3	214.5	203.3
0.4	265.9	395.9	416.3	355.4
2.0	308.4	1081.3	567.8	580.8
4.0	301.1	653.3	852.6	521.5
Statistics:				
P(w/C_{ex})	0.6420	0.0913	0.2008	0.2354
P(w/o C_{ex})	0.5724	0.1178	0.0591	0.1561
P(Linear)	ns	ns	0.0172	0.0890
P(Quad.)	ns	0.0545	ns	ns
DMRT	ns	2.0>C	4.0>C,0.4	ns

[1]ns=not significant. [a]No external control wells at this time.

Eleven collections for groundwater at 120 cm depth were done during the study period. The highest E_c values for the external control, the internal control, and the treated plots were 716, 275, and 1,219 umho cm^{-1}, respectively (Table IIAB-3, Appendix). All samples from treated plots showed elevated E compared to samples from the control plots. All samples collected during 1990 indicated no significant difference between treatments. In late 1991 two groundwater samples indicated slight evidences (P{linear/quadratic}<0.10) of linear or quadratic trend. All four samples collected in 1992 indicated linear (P{linear}=0.0533, 0.0764, 0.0408, and 0.0080) increases in E_c with PG rates. The DMR test showed that application of 2.0 or 4.0 Mg PG ha^{-1} could effectively increase the E_c of groundwater at the 120 cm depth some three years after application (Table IIAB-3, Appendix).

The 1990 and 1991, by-year (annual) ANOVA showed that differences among treatments in E_c of groundwater sampled at 120 cm depth were not significant. By 1992 a slight effect of treatment (P=0.0591) was indicated, with an increasing linear (P{linear}=0.0172) trend with PG rates. In 1991, the DMR test showed that plots that received 2.0 Mg PG ha^{-1} had higher E_c than the control. In 1992, the DMR test showed that application of 4.0 Mg PG ha^{-1} had higher E_c than the control and the plots that received 0.4 Mg PG ha^{-1} annually. When averaged over 3 years, the analysis showed no significant effect of treatment, but only a slight (P{linear}=0.0890) increasing linear trends with rates (Table A9).

For the ryegrass experiment, four collections of groundwater samples at 60 cm depth were made over a 2-year period as there was no water in the wells at this depth during crop year 1992-93. The E_c for the internal control wells samples ranged from 463 to 895 and for the treated from 273 to 1469 umho cm^{-1}. During 1990-91, individual sampling data showed slight evidences of E_c increasingly linearly (August, P{linear}=0.0589; October, P{linear}=0.0482) with PG rates (Table IIAR-1, Appendix). By-year average analysis of the data also indicated a similar trend (Table A10). During 1991-92, the treated plots indicated elevated E_c relative to the control, but statistical tests showed non-significant differences among the treatments both in the individual sampling or when averaged by year. The 2-year average data showed the control with 713 and the 4.0 Mg PG ha^{-1} plots with 1137 umho cm^{-1}, but the statistical analysis showed the difference to be not significant (Table A10).

Nine collections of groundwater samples at 120 cm depth were done over the 3-year period. The E_c for the internal control wells samples ranged from 131 to 429 and for the treated from 105 to 1211 umho cm^{-1}. None of the individual collection indicated any significant effect of treatments (Table IIAR-2, Appendix). The by-year and the 3-year data also showed that the differences in E_c among the treatments were not significant (Table A10).

Table A10. E_c of surficial groundwater from an annual ryegrass pasture amended with PG as source of S and Ca, averaged by crop year and over 3 crop years, 1990-91 to 1992-93, by depth.

Depth (cm)/ Treatment	Crop year 1990-91	1991-92	1992-93	Mean
A. 60 cm:	---------- umho cm^{-1} ----------			
Mg PG ha^{-1}				
0.0(C)	678.8	747.3	-[a]	713.0
0.4	395.5	1212.3	-[a]	803.9
2.0	1045.8	1071.5	-[a]	1058.6
4.0	1420.5	854.0	-[a]	1137.3
Statistics:				
P	0.1527	0.6517	-[a]	0.6060
P(Linear)	0.0519	ns[1]	-[a]	ns
B. 120 cm:	---------- umho cm^{-1} ----------			
Mg PG ha^{-1}				
0.0(C_{ex})	114.6	166.3	165.0	136.6
0.0(C)	141.1	355.0	357.5	270.2
0.4	170.6	492.3	559.3	383.8
2.0	169.8	521.2	785.2	459.8
4.0	281.8	625.2	528.0	458.7
Statistics:				
P(w/C_{ex})	0.3667	0.3094	0.4260	0.4577
P(w/o C_{ex})	0.4807	0.2800	0.5501	0.5783

[1]ns=not significant. [a]No water in wells.

Considering all the individual values, only few values approached the upper limit of 1500 umho cm^{-1} of potable water in the U.S., and only one value (1583 umho cm^{-1} collected in June, 1991 at 60 cm depth from bahiagrass plots treated with 4.0 Mg PG ha^{-1}), exceeded this limit. The highest by-year average of 1190 (1991) and 3-year average of 821 umho cm^{-1} were also measured in samples from plots that received 4.0 Mg PG ha^{-1} and collected at 60 cm depth. In the ryegrass experiment, the highest by-year (1990-91) and 2-year average E_c of 1421 and 1137 umho cm^{-1}, respectively, were again noted in groundwater from plots treated with 4.0 Mg ha^{-1} sampled at the 60 cm depth. Converting the highest measured individual, the highest annual average, and the highest multi-year average E_c values into TDS give 1045, 938, and 750 mg L^{-1} of dissolved minerals. Using TDS values, it is concluded that runoff and subsurface waters from established bahiagrass and tilled land amended with PG at as much as 4.0 Mg ha^{-1} once every 3 years do meet water quality requirement for agricultural, industrial, domestic uses.

Impact on F in groundwater. Runoff from the bahiagrass plots was sampled only once in 1990. The lowest F level was 0.22 for the control and the highest was 0.69 mg L^{-1} for plots treated with 4.0 Mg PG ha^{-1} (Table IIAB-1, Appendix). The difference was not significant. The test for a linear trend, however, indicated a slight evidence (P{linear}=0.0566) of F levels increasing with PG rates. The F in runoff collected in June, 1991 to July, 1992 ranged from 0.07 to 0.16 mg L^{-1}. No differences among treatments were noted, but plots treated with 0.4 Mg PG ha^{-1} annually tended to have elevated values relative to all the others. In 1992, runoff from plots treated with 0.4 Mg PG ha^{-1} annually had the highest F content of 0.96 and the control the least with 0.30 mg L^{-1}. Treatment effects were in the order of 0.4>4.0>2.0>C (Table IIAB-1, Appendix).

The 1990 by-year average analysis showed slight evidence (P{linear}=0.0566) of F levels in runoff tending to increase with PG rates, while the 1992 by-year and the 3-year analysis showed that treatment differences were significant (Table A11). The 3-year analysis indicated further that F in runoff increased linearly with PG rates (P{linear}=0.0002) from 0.15 in the control to 0.35 mg L^{-1} in plots that received 4.0 Mg PG ha^{-1}.

The highest by-sampling F levels in groundwater from 60 cm depth were 0.43, 0.36, and 2.0 mg L^{-1} for the external control, the internal control, and the treated plots, respectively. Seven out of 8 collections made over the 3-year period showed elevated F levels in groundwater from treated plots relative to those from the control. However, only the October, 1990 (P=0.0902; P{linear}<0.05) and October, 1992 samplings (P<0.05; P{linear}<0.05) indicated significant treatment effects (Table IIAB-2, Appendix).

The 1990 by-year and the 3-year average analysis with P=0.0316 and P=0.0841, respectively presented further evidences that PG increased the F contents of groundwater at the 60-cm depth. Also, the 1990 and 1991 by-year and 3-year average analysis showed that the F levels increased linearly with PG rates (Table A11).

The highest by-sampling F levels in groundwater at 120 cm depth were 1.44, 0.29, and 0.60 mg L^{-1} for the external control, the internal control, and the treated plots, respectively (Table IIAB-3, Appendix). All 11 samplings over the 3-year period showed all samples from the treated plots with elevated F contents relative to those from the internal control. However, only the September, 1990 (P=0.0660) and June, 1991 (P=0.0683) samples indicated slight evidences of the effects of PG on F in groundwater at 120 cm depth. Both samplings also showed F levels increasing linearly (P{linear}=0.0254 and 0.0522) with rates (Table IIAB-3, Appendix). The by-year and 3-years statistics showed no effect of PG on F levels in groundwater at 120 cm depth under an established bahiagrass pasture (Table A11).

Table A11. F contents in runoff and surficial groundwater from a bahiagrass pasture amended with PG as source of S and Ca, averaged by year and over a 3-year period, 1990-1992.

Depth(cm)/ Treatment	Crop year 1990	1991	1992	Mean
A. Runoff:	---------- mg F L^{-1} ----------			
Mg PG ha^{-1}				
0.0(C)	0.22	0.09	0.17	0.15
0.4	0.27	0.13	0.56	0.35
2.0	0.42	0.13	0.29	0.25
4.0	0.69	0.12	0.41	0.35
Statistics:				
P	0.1914	0.8979	0.0478	0.0005
P(Linear)	0.0566	ns	ns	0.0002
P(Quad.)	ns[1]	ns	ns	0.0377
DMRT	ns	ns	0.4>C,2.0; 4.0> 2.0>C	0.4,
		4.0>C		
B. 60 cm:	---------- mg F L^{-1} ----------			
Mg PG ha^{-1}				
0.0(C_{ex}	-[a]	0.18	0.25	0.22
0.0(C)	0.11	0.14	0.23	0.15
0.4	0.14	0.15	0.43	0.20
2.0	0.17	0.33	0.83	0.36
4.0	0.22	0.30	0.71	0.36
Statistics:				
P(w/C_{ex})	-[a]	0.0883	0.2270	0.0505
P(w/o C_{ex})	0.0316	0.1269	0.3008	0.0841
P(Linear)	0.0086	0.0887	ns	0.0498
DMRT	4.0>C, 0.4;2.0>C	ns	ns	4.0>C
C. 120 cm:	---------- mg F L^{-1} ----------			
Mg PG ha^{-1}				
0.0(C_{ex})	0.24	0.30	0.85	0.50
0.0(C)	0.10	0.08	0.16	0.11
0.4	0.21	0.14	0.28	0.21
2.0	0.19	0.27	0.35	0.25
4.0	0.20	0.22	0.32	0.23
Statistics:				
P(w/C_{ex})	0.4628	0.3540	0.6085	0.5475
P(w/o C_{ex})	0.4359	0.1466	0.5053	0.3305
DMRT	ns	2.0>C	ns	ns

[1]ns=not significant. [a]No external control wells at this time.

In the ryegrass experiment, the by-sampling F concentrations in groundwater at 60 cm depth ranged from 0.14 to 0.32 for the control plots and from 0.13 to 0.70 mg L^{-1} for the treated plots. Although the treated plots had consistently elevated F levels relative to the control, the differences were not significant (Table IIAR-1), Appendix). Similar values and trends were noted in the by-year and 3-year average analysis, with no significant difference among treatments (Table 12A).

Table A12. F of surficial groundwater from an annual ryegrass pasture amended with PG as source of S and Ca, averaged by crop year and over 3 crop years, 1990-91 to 1992-93, by depth.

Depth (cm)/ Treatment	Crop year 1990-91	1991-92	1992-93	Mean
A. 60 cm:	---------- mg F L^{-1} ----------			
Mg PG ha^{-1}				
0.0(C)	0.15	0.31	-[a]	0.23
0.4	0.15	0.31	-[a]	0.23
2.0	0.17	0.38	-[a]	0.28
4.0	0.34	0.53	-[a]	0.43
Statistics:				
P	0.7290	0.7818	-[a]	0.7443
B. 120 cm:	---------- mg F L^{-1} ----------			
Mg PG ha^{-1}				
0.0(C_{ex}	0.01	0.04	0.28	0.07
0.0(C)	0.03	0.09	0.19	0.10
0.4	0.04	0.15	0.27	0.14
2.0	0.04	0.12	0.26	0.13
4.0	0.04	0.14	0.29	0.15
Statistics:				
P(w/C_{ex})	0.7697	0.3371	0.9099	0.7371
P(w/o C_{ex})	0.8088	0.4497	0.8686	0.7995

[a]No water in wells.

The by-sampling values for F in groundwater at 120 cm depth were much lower than those at 60 cm depth, ranging from 0.01 to 0.53 mg L^{-1} (Table IIAR-2, Appendix). Nine of the eight samplings done over the 3-year period showed any significant effect of treatments. The highest by-year and 3-year average F levels for the internal control samples were 0.19 and 0.10 and for the treated 0.29 and 0.15 mg L^{-1}, respectively (Table A12).

The impact of PG on F in surficial groundwater in relation to certain values and standards may now be considered.

In an established pasture of bahiagrass, the highest F levels in individual sample from the treated plots during three years of sampling were 0.96 mg L^{-1} for runoff, 2.00 mg L^{-1} for samples at the 60-cm depth (recorded from plots that received 2.0 but not from 4.0 Mg PG ha^{-1}), and 0.60 mg L^{-1} at the 120-cm depth. The highest single year and 3-year average F levels for all types of surficial groundwater samples were 0.83 and 0.36 mg L^{-1}, and both from plots that received 2.0 Mg PG ha^{-1} and at 60 cm depth.

In a tilled land where PG was mixed with the soil to a depth of 15 cm and the land planted to annual ryegrass, the highest by-sampling F levels in qroundwater for the treated plots at 60 and 120 cm depths were 0.70 and 0.53 mg L^{-1}. The highest by-year and 3-year averages for all treated plots were for both depths were 0.53 and 0.29 mg L^{-1}. It is widely accepted that approximately 1.0 Mg F L^{-1} in drinking water can effectively reduce dental caries without harmful effects on health (Standard Methods, 1985). Also, the Florida drinking water primary standards allow for a maximum contamination level (MCL) for F from 1.4 to 2.4 mg L^{-1}. None of the individual measured F values in the bahiagrass and ryegrass experiments exceeded the 2.4 MCL, and the by-year and 3-year average values were even less than 1.0 mg F L^{-1}.

Therefore, it is concluded that application of PG up to 4.0 Mg ha^{-1} to an established pasture or tilled land does not lead to unacceptable levels of F in surficial groundwater.

B. RADIOLOGICAL ASPECTS ASSOCIATED WITH PG APPLICATION AS A SOURCE OF S AND Ca TO AN ESTABLISHED BAHIAGRASS PASTURE AS A SOURCE OF S AND Ca

B.l. Radionuclides in PG

The concentrations reported for the three measured radionuclides are given in Table Bl. The ^{226}Ra value of 18.2 pCi g^{-1} is at the lower end of the range of values reported by USEPA (1992b) for Central Florida PG (5-stack average: 31, range of stack means: 25-34, range of individual samples: 16-81). The ^{210}Pb value of 30.8 pCi g^{-l}, ^{210}Po value of 24.3 pCi g^{-l}, and the $^{210}Po/^{210}Pb$ ratio of 0.79 are comparable to the EPA-reported values (36, 27 and 0.74, respectively).

Table B1. Radionuclide concentrations in PG used in the study.

Radionuclides Concentration	pCi g^{-1}
^{226}Ra	18.2
^{210}Pb	30.8
^{210}Po	24.3

B.2 Soil Radionuclides

Radionuclide concentrations by treatment and sampling depth, averaged over the 3-year study period, are summarized in Table B2. The results of analyses and statistical testing by year for 1990, 1991, and 1992 are presented in Appendix Tables IIB-1, IIB-2, and IIB-3, respectively.

Radium-226. Results are compared by treatment and depth for 1990, 1991, and 1992 in Figures IIB-1, IIB-2, and IIB-3, respectively. The most striking observation is that the year-to-year variability is greater than any treatment and depth differences. This effect was observed in the control plot (0.0 Mg ha^{-1}) which should be relatively invariant and indicates a considerable analytical variability associated with this measurement. Using the control plot 3-year averages (Table B2) as the best available estimates of the baseline radioactivity indicates that the natural radioactivity of this soil is low (<1 pCi g^{-1}) and relatively uniform with depth over the 90-cm sampling depth. For these plots, presumably unaffected by the PG treatment, the 3-year averages at each depth have standard errors on the order of 0.1 to 0.2 pCi g^{-1} and relative standard errors on the order of 20%.

Table B2. ^{226}Ra, ^{210}Pb, and ^{210}Po concentrations in a Florida Spodosol soil cropped to bahiagrass which was amended with PG as a source of S and Ca, averaged over a 3-year period, 1990-1992, by depth.

Treatment	Depth (cm)					
	0-15	15-30	30-45	45-60	60-75	75-90
Mg PG ha^{-1}	---------- pCi ^{226}Ra g^{-1} ----------					
0.0(C)	0.55	0.67	0.56	0.53	0.55	0.75
0.4	0.46	0.43	0.58	0.58	0.75	0.70
2.0	0.65	0.63	0.43	0.58	0.53	0.35
4.0	0.58	0.80	0.40	0.47	0.48	0.48
Statistics:						
P	0.6428	0.5187	0.3663	0.9637	0.1801	0.4517
P(Linear)	ns[1]	ns	ns	ns	ns	ns
Mg PG ha^{-1}	---------- pCi ^{210}Pb g^{-1} ----------					
0.0(C)	0.61	0.55	0.42	0.32	0.45	0.42
0.4	0.59	0.28	0.22	0.42	0.48	0.33
2.0	0.63	0.43	0.38	0.37	0.60	0.34
4.0	0.59	0.30	0.22	0.08	0.28	0.32
Statistics:						
P	0.9128	0.5666	0.1235	0.4896	0.3987	0.9636
DMRT	ns	ns	C>0.4,4.0	ns	ns	ns
Mg PG ha^{-1}	---------- pCi ^{210}Po g^{-1} ----------					
0.0(C)	0.53	0.28	0.23	0.24	0.21	0.23
0.4	0.58	0.68	0.75	0.64	0.40	0.32
2.0	0.90	0.32	0.44	0.49	0.31	0.44
4.0	0.79	0.45	0.23	0.64	0.79	1.07
Statistics:						
P	0.8437	0.4090	0.1547	0.2688	0.5478	0.0279
P(Linear)	ns	ns	ns	ns	ns	0.0088
DMRT	ns	ns	ns	ns	ns	4.0>all

[1]ns=not significant.

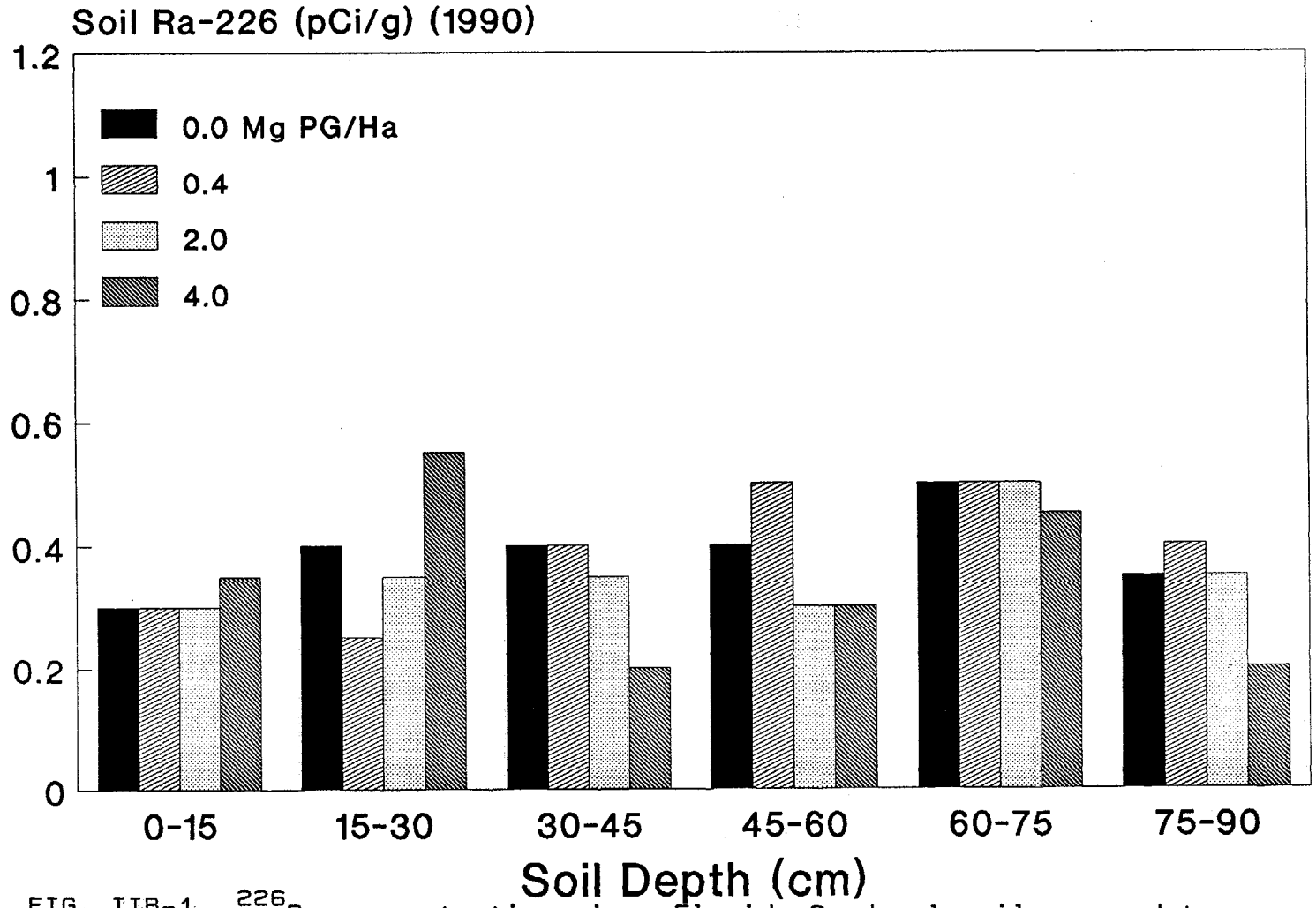

FIG. IIB-1. ^{226}Ra concentrations in a Florida Spodosol soil cropped to bahiagrass which was amended with PG as source of S and Ca, 1990, by depth.

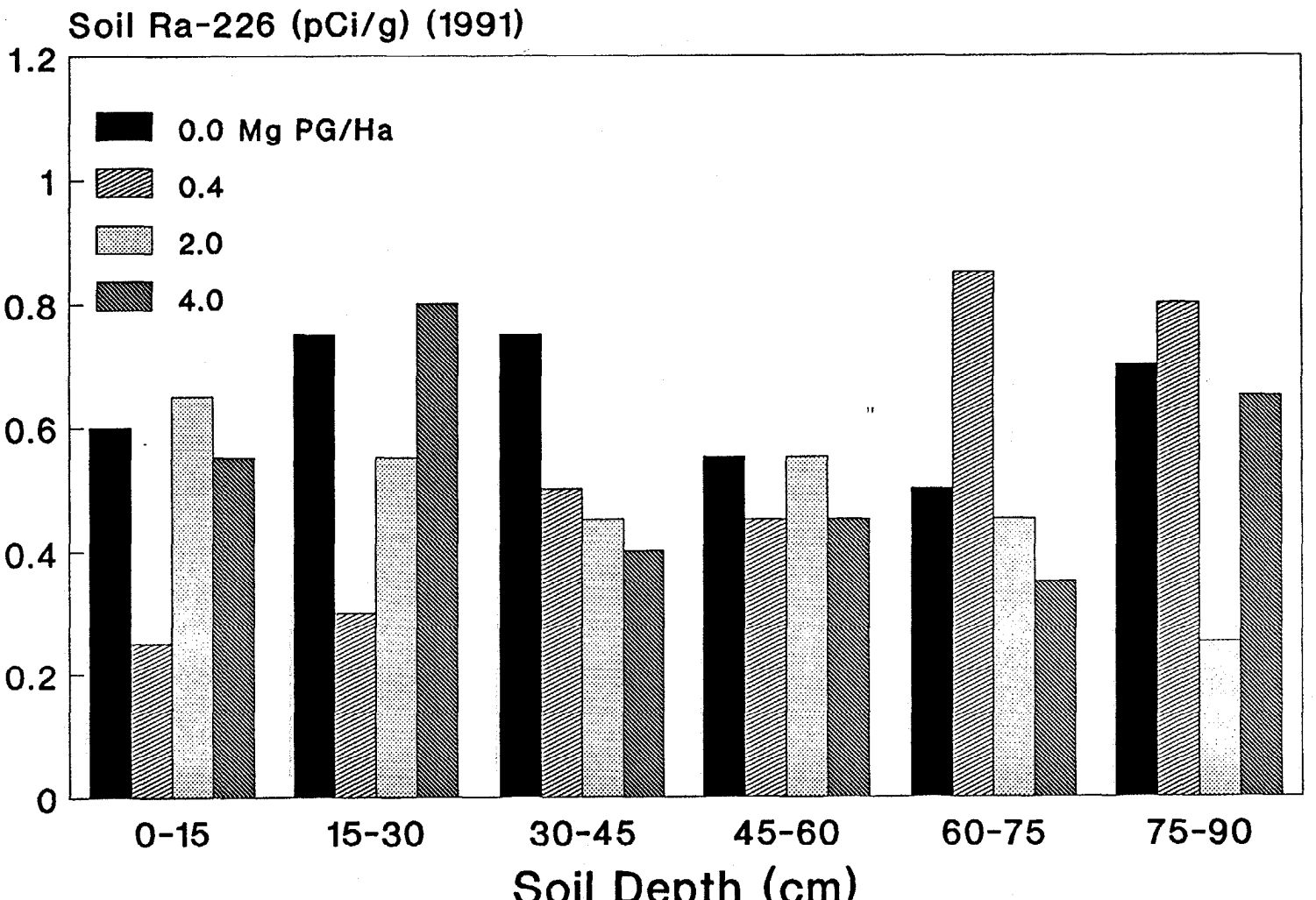

FIG. IIB-2. ^{226}Ra concentrations in a Florida Spodosol soil cropped to bahiagrass which was amended with PG as source of S and Ca, 1991, by depth.

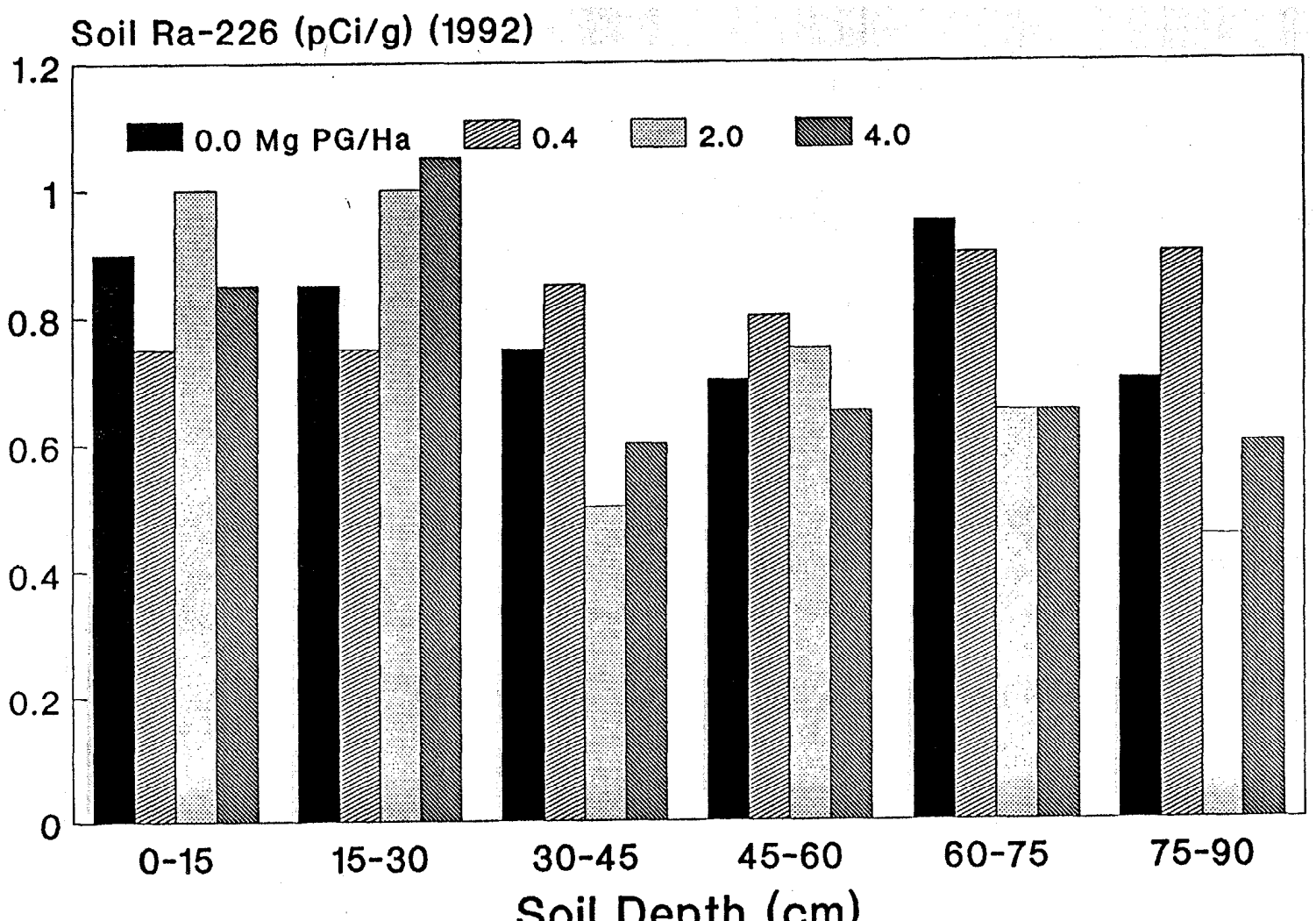

FIG. IIB-3. ^{226}Ra concentrations in a Florida Spodosol soil cropped to bahiagrass which was amended with PG as source of S and Ca, 1992, by depth.

No meaningful effects of treatment were observed. Although the treatment differences were statistically significant at the 30-45 cm depth in 1990 and the 60-75 cm depth in 1992, soil ^{226}Ra concentrations in plots treated with 4.0 Mg PG ha^{-1} were actually lower than in all the others including the control plots.

The inability to detect a treatment effect is not surprising. The maximum treatment level of 4.0 Mg PG ha^{-1}, when distributed over a 15-cm soil column, gives a calculated soil radioactivity increase of only 0.03 pCi g^{-1}. Thus, any increase in radioactivity due to the treatment would not be detected by this sampling and analytical procedure.

By comparison, the study of Mullins and Mitchell, Jr. (1990) reported soil ^{226}Ra concentrations in PG experimental plots which were sampled down to 102 cm ranging from 0.08 to 0.35 pCi g^{-1}, with no meaningful treatment differences.

Lead-210. Comparisons of ^{210}Pb concentrations are given in Figures IIB-4, IIB-5, and IIB-6 for 1990, 1991, and 1992, respectively. The values for the control plots (Table B2) indicate that the natural ^{210}Pb concentration of the soil was low ($\leq$ 0.6 pCi g^{-1}). The activity was relatively uniform with depth, but the data suggest that the activity was higher in the first 15 cm, an effect that could be attributed to deposition of ^{210}Pb produced by atmospheric Rn. The 3-year averages at each depth had standard errors on the order of 0.03 to 0.1 pCi g^{-1}.

The data in general are highly variable. The 1990 samples for the 0-15 cm depth (Table IIB-1, Appendix) showed a significant treatment difference (P=0.03, 4.0>all, P{linear} = 0.02), suggesting an initial effect on the uppermost layer; this effect was not observed in subsequent years. No significant treatment effects were observed in 1991 (Table IIB-2, Appendix). The 1992 data (Table IIB-3, Appendix) indicated a significant, linear treatment effect at the 15-30 cm depth. However, this observation is suspect since it is driven by a 1992 average control plot value at this depth that was atypically low - 0.1 pCi g^{-1} and 20% of the 0-15 cm value vs. previous measurements that were 0.4 to 0.6 pCi g^{-1} and comparable to the O-15 cm value.

Again, the very limited evidence of treatment effects is not surprising. For the maximum treatment level of 4.0 Mg PG ha^{-1} and averaging over a 15-cm depth, the calculated soil ^{210}Pb increase is only 0.05 pCi g^{-1}. The sampling procedure used would not detect such an increase in the presence of the natural variability of ^{210}Pb in the control plots.

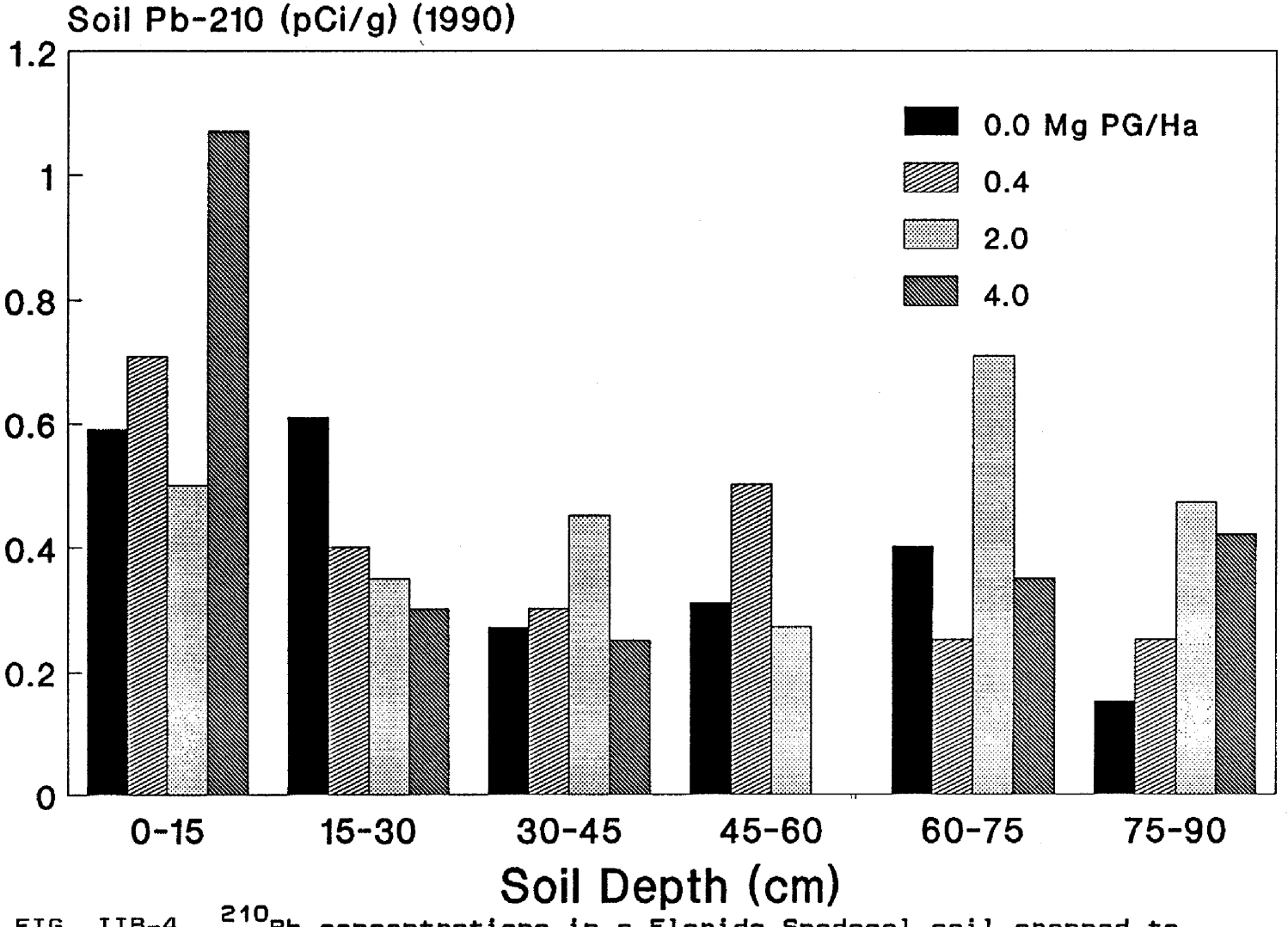

FIG. IIB-4. ^{210}Pb concentrations in a Florida Spodosol soil cropped to bahiagrass which was amended with PG as source of S and Ca, 1990, by depth.

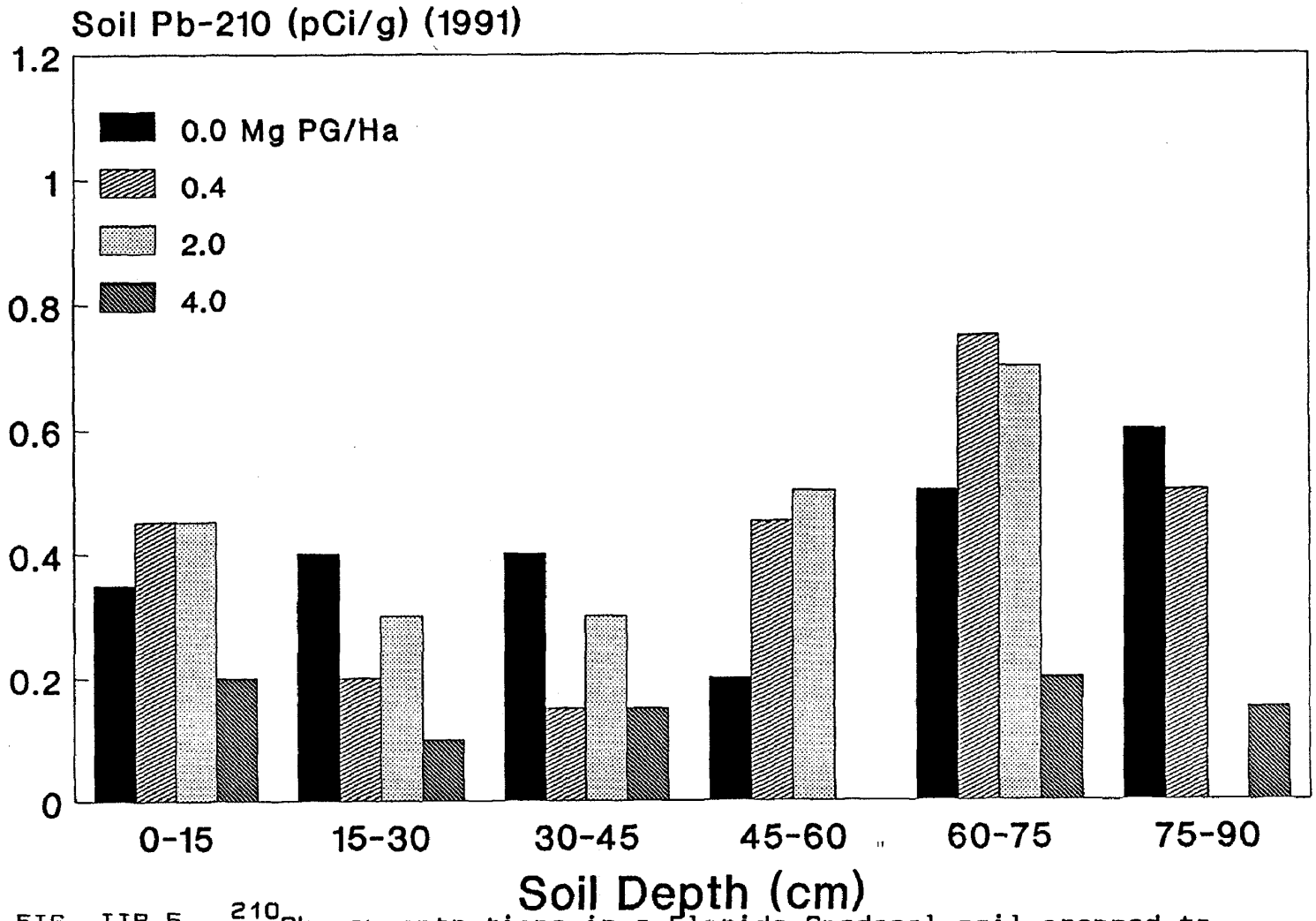

FIG. IIB-5. ^{210}Pb concentrations in a Florida Spodosol soil cropped to bahiagrass which was amended with PG as source of S and Ca, 1991, by depth.

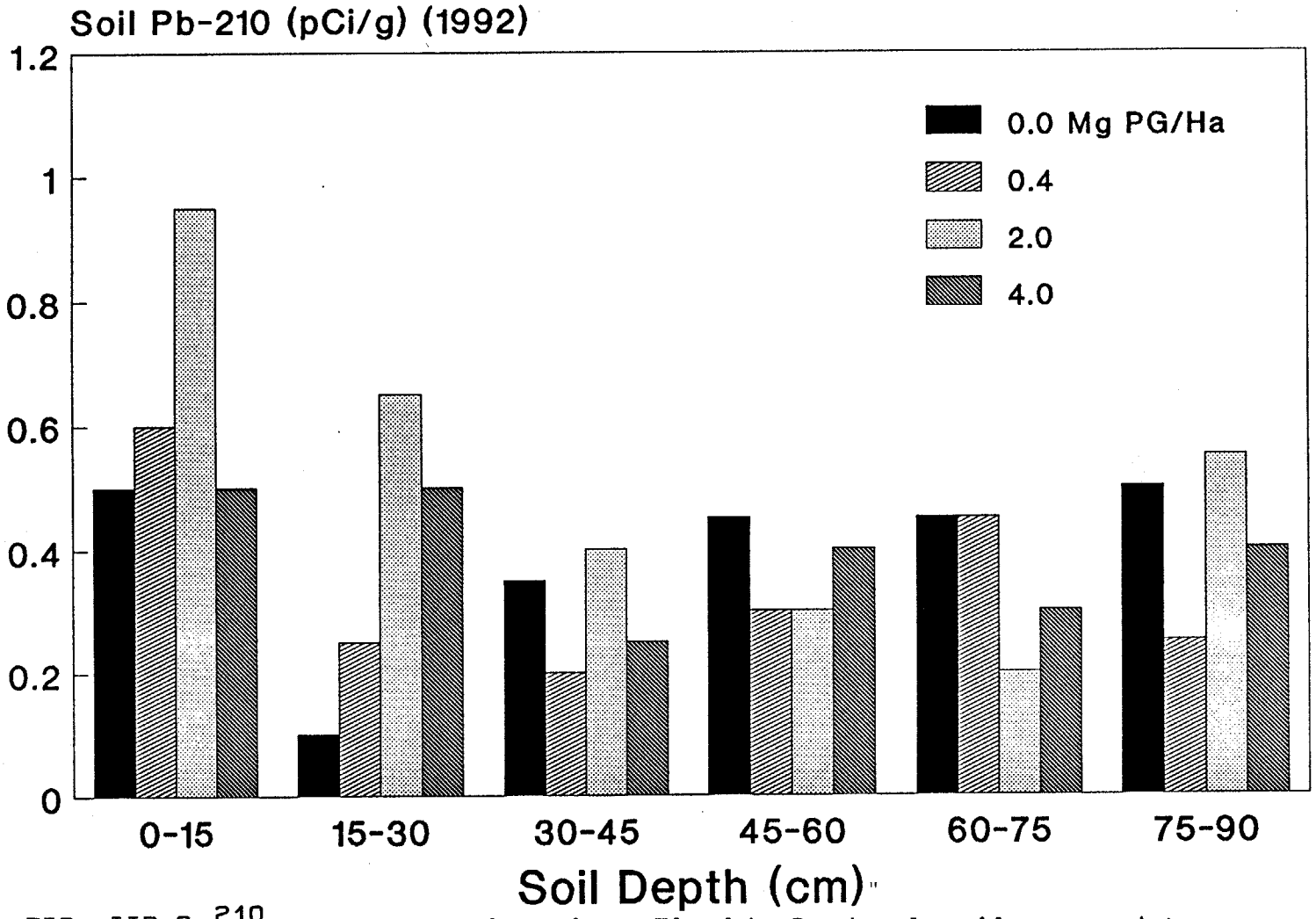

FIG. IIB-6. ^{210}Pb concentrations in a Florida Spodosol soil cropped to bahiagrass which was amended with PG as source of S and Ca, 1992, by depth.

Polonium-210. Average soil ^{210}Po concentrations in the control plots (Table B2) indicate that natural ^{210}Po concentration for the soil was low (<1.0 pCi g^{-1}) in the top 15-cm layer and relatively uniform at <0.2 pCi g^{-1} from 15- down to 90-cm depth. This depth effect was consistent with deposition of ^{210}Po produced by atmospheric Rn. The 3-year averages showed standard errors on the order of 0.2 pCi g^{-1} in the 0 to 15-cm depth and from 0.03 to 0.1 in the other layers. Mullins and Mitchell, Jr. (1990) in their study reported a soil ^{210}Po range of from 0.17 to 0.48 pCi g^{-1} from the top soil down to the 46-cm depth.

Concentrations by treatment and by depth for 1990, 1991, and 1992 are presented in Figures IIB-7, IIB-8, and IIB-9, respectively. The data are highly variable and, except for the 75-90 cm sampling in 1992 (Table IIB-3, Appendix) and in the 3-year average for that depth (Table B2), no statistically significant treatment effects were observed. The value of 2.15 pCi g^{-1} reported at 75 to 90 cm in 1992 for the 4.0 Mg ha^{-1} treatment (and the 3-year average based on this value) is highly suspect - it is considerably higher than all other reported values and represents a PG-attributable addition to baseline of about 2.0 pCi g^{-1} while the calculated increase for that treatment would be 0.04 pCi g^{-1} if the radioactivity is transported in total from the surface to the 75-90 cm layer.

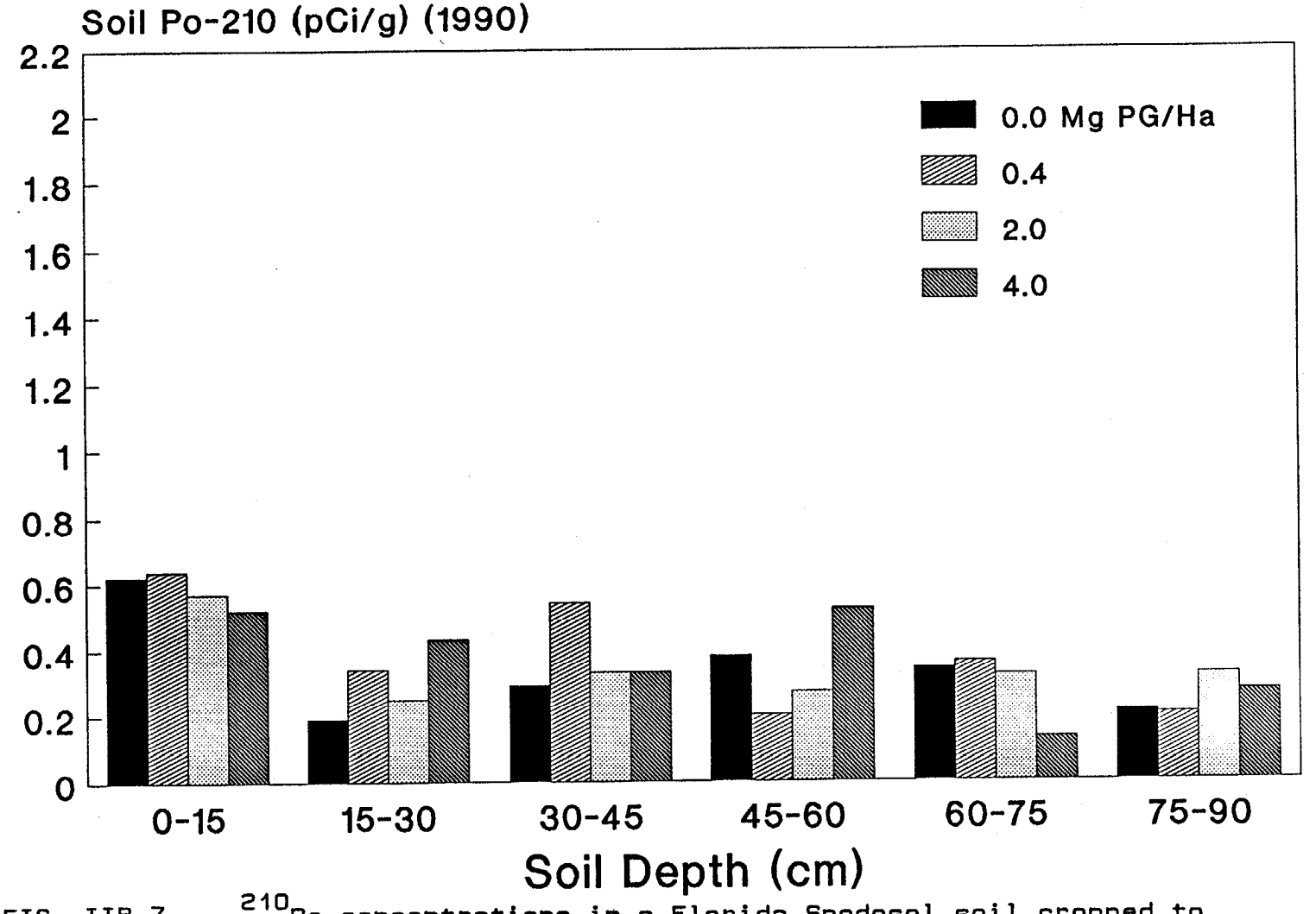

FIG. IIB-7. ^{210}Po concentrations in a Florida Spodosol soil cropped to bahiagrass which was amended with PG as source of S and Ca, 1990, by depth.

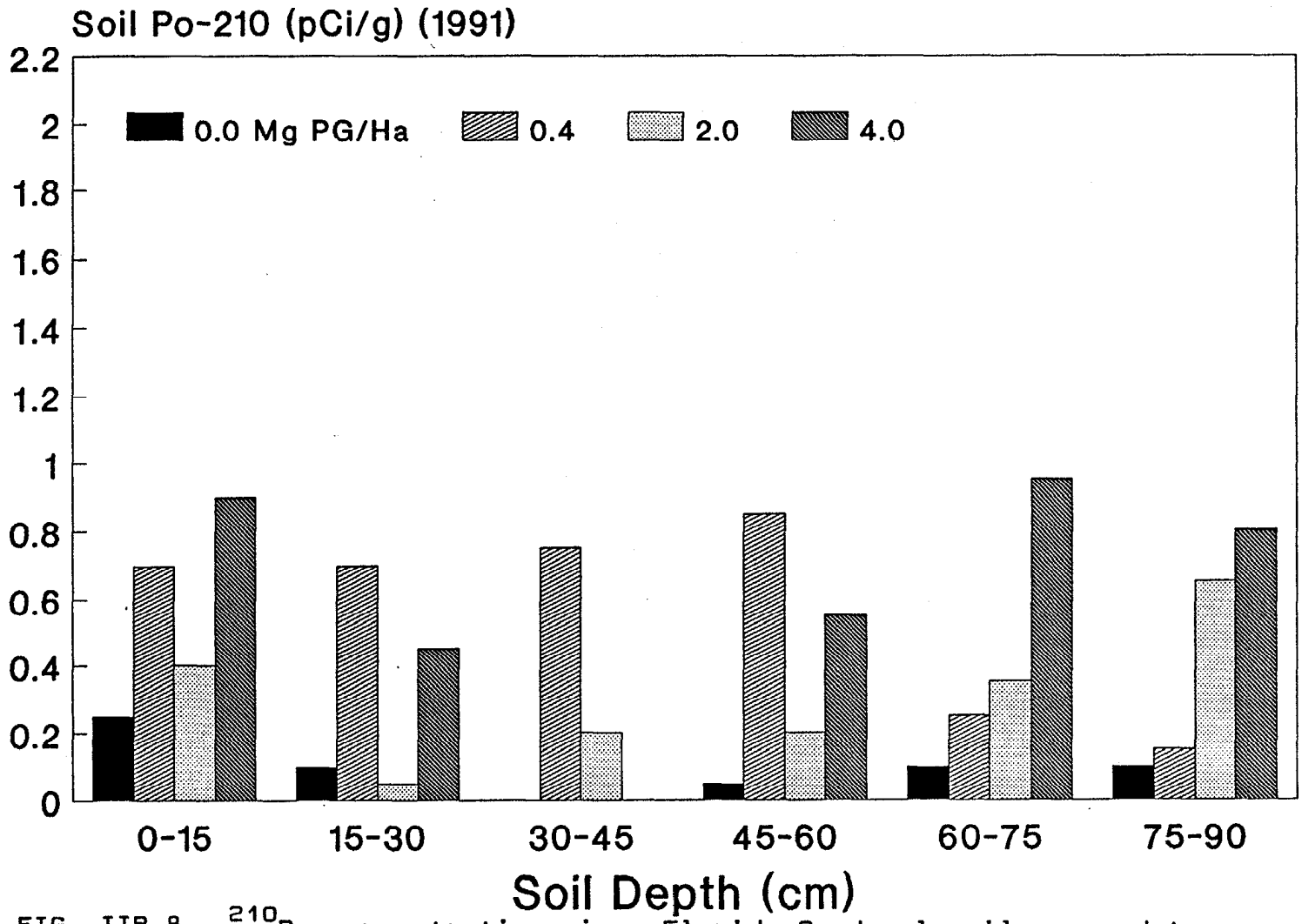

FIG. IIB-8. ^{210}Po concentrations in a Florida Spodosol soil cropped to bahiagrass which was amended with PG as source of S and Ca, 1991, by depth.

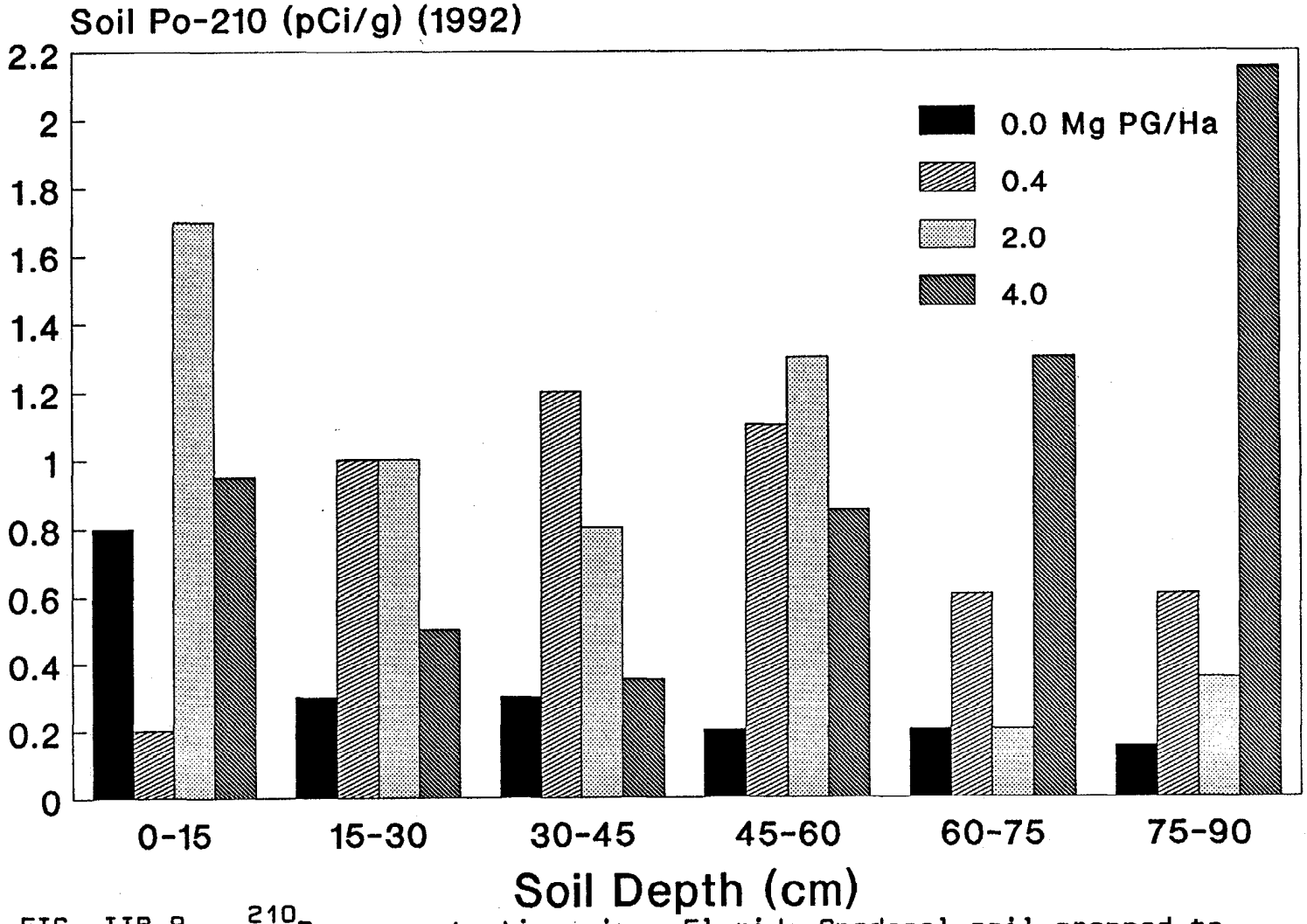

FIG. IIB-9. ^{210}Po concentrations in a Florida Spodosol soil cropped to bahiagrass which was amended with PG as source of S and Ca, 1992, by depth.

B.3. Gamma Radiation.

Gamma radiation was measured over the control plots for two consecutive time intervals during 74 days prior to application of the PG. Following PG application, measurements were made over all plots for 13 time intervals of 30 to 75 days each during the 3-year study period. Annual and 3-year average gamma radiation exposure levels are presented in Table B3. Results by individual sampling period are presented in Figure IIB-10 and in the Appendix (Table IIB-4).

Table B3. Gamma radiation over a Florida Spodosol soil cropped to bahiagrass which was amended with PG as a source of S and Ca, averaged by year and over a 3-year period, 1990-1992.

Treatment	Crop year			
	1990-91	1991-92	1992	mean
Mg PG ha^{-1}	---------- uR hr^{-1} ----------			
0.0(C_{ex1})	4.96	5.28	5.61	5.23
0.0(C_{ex2})	5.59	5.89	6.53	5.92
0.0(C)	4.29	5.28	5.48	4.81
0.4	4.54	5.06	5.45	5.09
2.0	4.55	5.37	5.35	5.04
4.0	4.71	5.08	5.23	4.99
Statistics:				
P(w/$C_{ex1,2}$)	0.0314	0.1167	0.0197	0.0130
P(w/o $C_{ex1,2}$)	0.4523	0.4908	0.6450	0.8672

Baseline levels over the study period averaged 4.8 uR h^{-1} (range of 3.85 to 6.4) for the internal control C, 5.2 (4.2 to 6.0) for C_{ex1}, and 5.9 (4.8 to 7.4) for C_{ex2}. This suggests there may be some local spatial variation in baseline gamma-radiation levels. By comparison, state averages in the U.S. range from 3.3 uR hr^{-1} in Texas to 14 in Colorado, Nevada, and Wyoming, with individual measurements ranging from <l to 34 (Myrick, et al., 1981). Previous studies have reported Florida values on the order of 5 to 10 uR hr^{-1} (Roessler, 1987).

Analysis of the data showed differences across treatments were statistically significant for about one third of the tests performed; however, there was no consistent trend with treatment. The greatest difference was typically between the external controls (highest values) and some treatment level among the contiguous plots. Thus, any influence of the PG treatment is insignificant in comparison to the temporal and spatial variations in the local background.

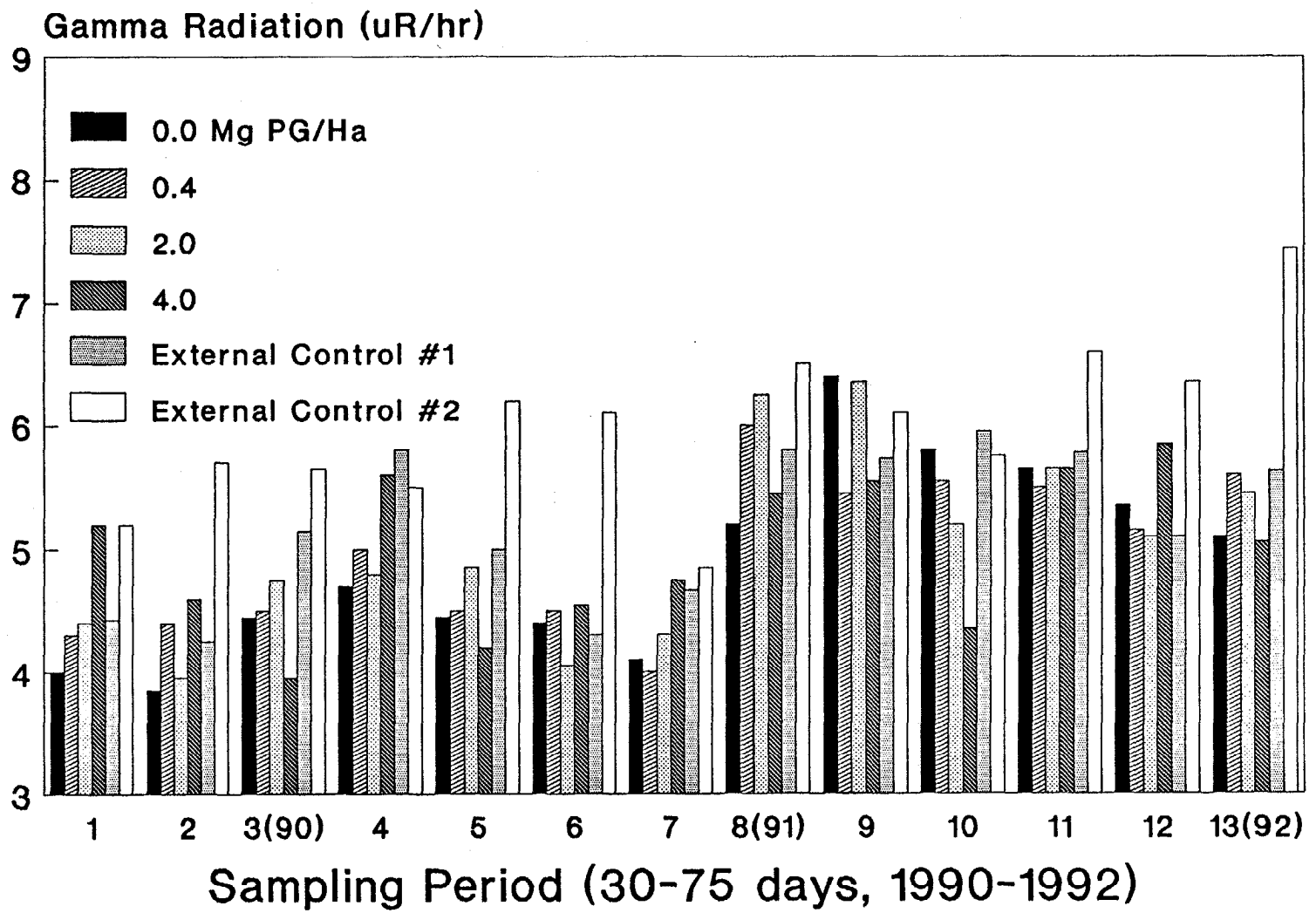

FIG. IIB-10. Gamma radiation over a Florida Spodosol soil cropped to bahiagrass which was amended with PG as source of S and Ca, 1990-1992, by sampling.

B.4. Radionuclides in Plant Tissue.

Three regrowth harvests and one hay or mature forage harvest from each of the years 1990, 1991, and 1992 were analyzed for radionuclides.

Radium-226. Annual average and 3-year average concentrations in regrowth forage and annual concentrations and 3-year average concentrations for the annual harvests of the mature forage are presented in Table B4. Results for individual regrowth harvests are presented in Figure IIB-11. Analytical and statistical testing results for individual harvests during 1990, 1991, and 1992 are presented in Tables IIB-5, IIB-6, and IIB-7 (Appendix), respectively, for all three radionuclides.

One of the most striking features of these data (see Figure IIB-11) is the variability and, in particular, the general increases from 1990 to 1991 to 1992, including in the control samples. Baseline (control plot) concentrations in regrowth forage averaged 0.05 pCi g^{-1}, with annual averages ranging from 0.01 to 0.13 and individual harvests ranging from below the limit of detection to 0.15. The concentration in control plot mature forage was 0.03 pCi g^{-1} for each of the two years sampled. By comparison, average tissue concentrations of 0.04 pCi g^{-1} were reported by Mislevy, et al. (1989) for various biomass plants growing on unmined Florida soil and concentrations ranging from 0.04 to 0.13 pCi $^{-1}$ were reported by Mullins and Mitchell (1990) for wheat forage from untreated plots in Alabama.

The regrowth data present limited evidence of a treatment effect. The 3-year mean indicated a significant treatment effect ($P = 0.06$, P{linear} = 0.02). On the other hand, treatment effects were generally not statistically significant for the individual harvests or for the annual averages. One limited exception was at the end of the 1990 season when there was evidence of a linear effect at the January 1991 harvest (Appendix, Table IIB-5, P{linear} = 0.07). Another exception was in 1991 when the August harvest showed a significant treatment effect (Appendix, Table IIB-6, $P = 0.3$, P{linear} = 0.2) and the annual average had evidence of a linear effect (Table B4, P{linear} = 0.07).

The mature forage data (Table B4, B) are limited in quantity (most of the 1992 data are missing and show considerable variability). Statistically-significant treatment effects were observed for 1990 ($P = 0.05$) and for the study average ($P = 0.03$, P{linear} = 0.02). There is no obvious explanantion of why the 0.4 Mg ha^{-1} annual treatment resulted in consistently higher tissue concentrations than single treatment at the two higher rates. Considering only the two single-application treatments, the 3-year average results indicated a concentration in mature forage that increased linearly with PG treatment rate.

Table B4. ^{226}Ra concentrations in bahiagrass regrowth and hay forage from a pasture amended with PG as a source of S and Ca and growing on a Florida Spodosol soil, averaged by year and over a 3-year period, 1990-1992.

Treatment	Crop year			
	1990[a]	1991[a]	1992[a]	Mean[a]
A. Regrowth:	pCi ^{226}Ra g^{-1}			
Mg PG ha^{-1}				
0.0(C)	0.01	0.04	0.13	0.05
0.4	0.02	0.04	0.17	0.06
2.0	0.01	0.06	0.13	0.05
4.0	0.03	0.09	0.23	0.10
Statistics:				
P	0.4583	0.1783	0.5758	0.0583
P(Linear)	ns[1]	0.0679	ns	0.0185
DMRT	ns	ns	ns	4.0>all
B. Hay:	pCi ^{226}Ra g^{-1}			
Mg PG ha^{-1}				
0.0(C)	0.03	0.03	–[b]	0.03
0.4	0.10	0.13	–[b]	0.11
2.0	0.03	0.10	–[b]	0.06
4.0	0.09	0.09	0.20	0.09
Statistics:				
P	0.0539	0.4030	–[b]	0.0327
P(Linear)	ns	ns	–[b]	0.0215
DMRT	0.4,4.0> C,2.0	ns	–[b]	0.4, 4.0>C; 0.4>2.0

[1]ns=not significant. [a]Statistics computed based solely on those harvests where each treatment had at least one detected value associated with it. [b]No analysis due to missing data.

In summary, ^{226}Ra uptake by bahiagrass regrowth forage during the first three years following PG treatment was on the order of 0.01 to 0.02 pCi g^{-1}/Mg PG ha^{-1}. Uptake in the annual accumulative (hay) growth during each of the first three years was on the order of 0.015 to 0.017 pCi g^{-1}/Mg PG ha^{-1} with the higher application rate supporting a value on the order of 0.02 pCi g^{-1}.

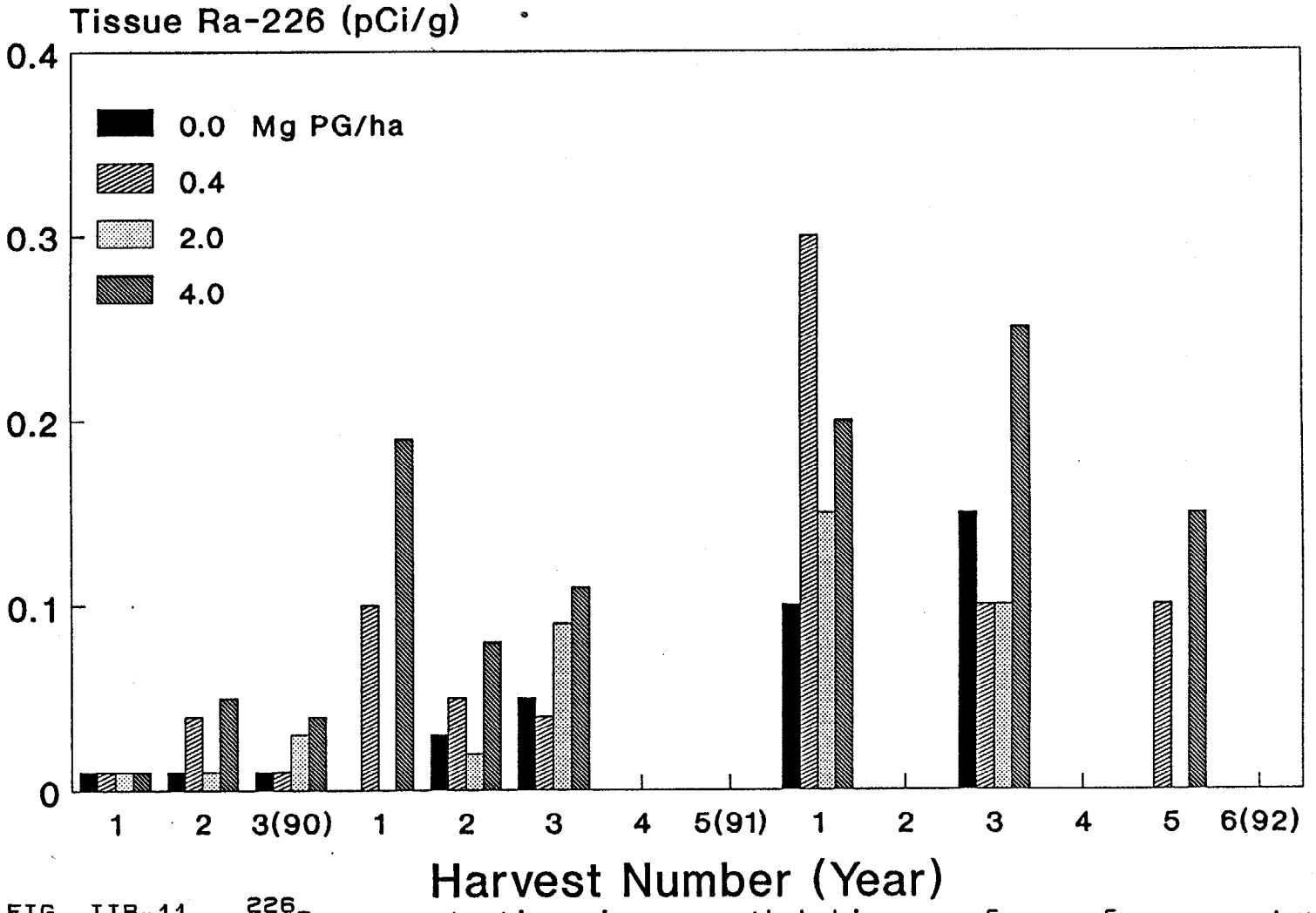

FIG. IIB-11. ^{226}Ra concentrations in regrowth bahiagrass forage from a pasture amended with PG as source of S and Ca and growing on a Florida Spodosol soil, 1990-1992, by harvest.

Lead-210. Annual average and 3-year average concentrations in regrowth forage and annual concentrations and 3-year average concentrations for the annual harvests of the mature forage are presented in Table B5. Results for individual regrowth harvests are presented in Figure IIB-12. As indicated for ^{226}Ra, analytical and statistical test results for individual harvests are presented in Appendix Tables IIB-5, IIB-6, and IIB-7.

Table B5. ^{210}Pb concentrations in bahiagrass regrowth and hay forage from a pasture amended with PG as a source of S and Ca and growing on a Florida Spodosol soil, averaged by year and over a 5-year period, 1990-1992.

Treatment	Crop year			
	1990[a]	1991[a]	1992[a]	Mean[a]
A. Regrowth:	---------- pCi ^{210}Pb g^{-1} ----------			
Mg PG ha^{-1}				
0.0(C)	0.20	0.50	2.53	0.93
0.4	0.13	0.73	2.13	0.93
2.0	0.14	0.62	2.10	0.87
4.0	0.13	0.46	2.22	0.81
Statistics:				
P	0.0719	0.0925	0.8259	0.7117
P(Linear)	0.0590	ns[1]	ns	ns
DMRT	C>all	0.4>4.0	ns	ns
B. Hay:	---------- pCi ^{210}Pb g^{-1} ----------			
Mg PG ha^{-1}				
0.0(C)	0.80	0.75	2.95	1.33
0.4	0.95	0.85	3.05	1.45
2.0	0.75	0.90	2.20	1.28
4.0	0.87	0.40	2.75	1.23
Statistics:				
P	0.8397	0.2874	0.7625	0.7513

[1]ns=not significant. [a]Statistics computed based solely on those harvests where each treatment had at least one detected value associated with it.

Again, a striking feature is the increases, including in the control data, for regrowth forage from 1990 to 1991 to 1992 (Figure IIB-12), for mature forage from 1990 to 1992 (Table B5).

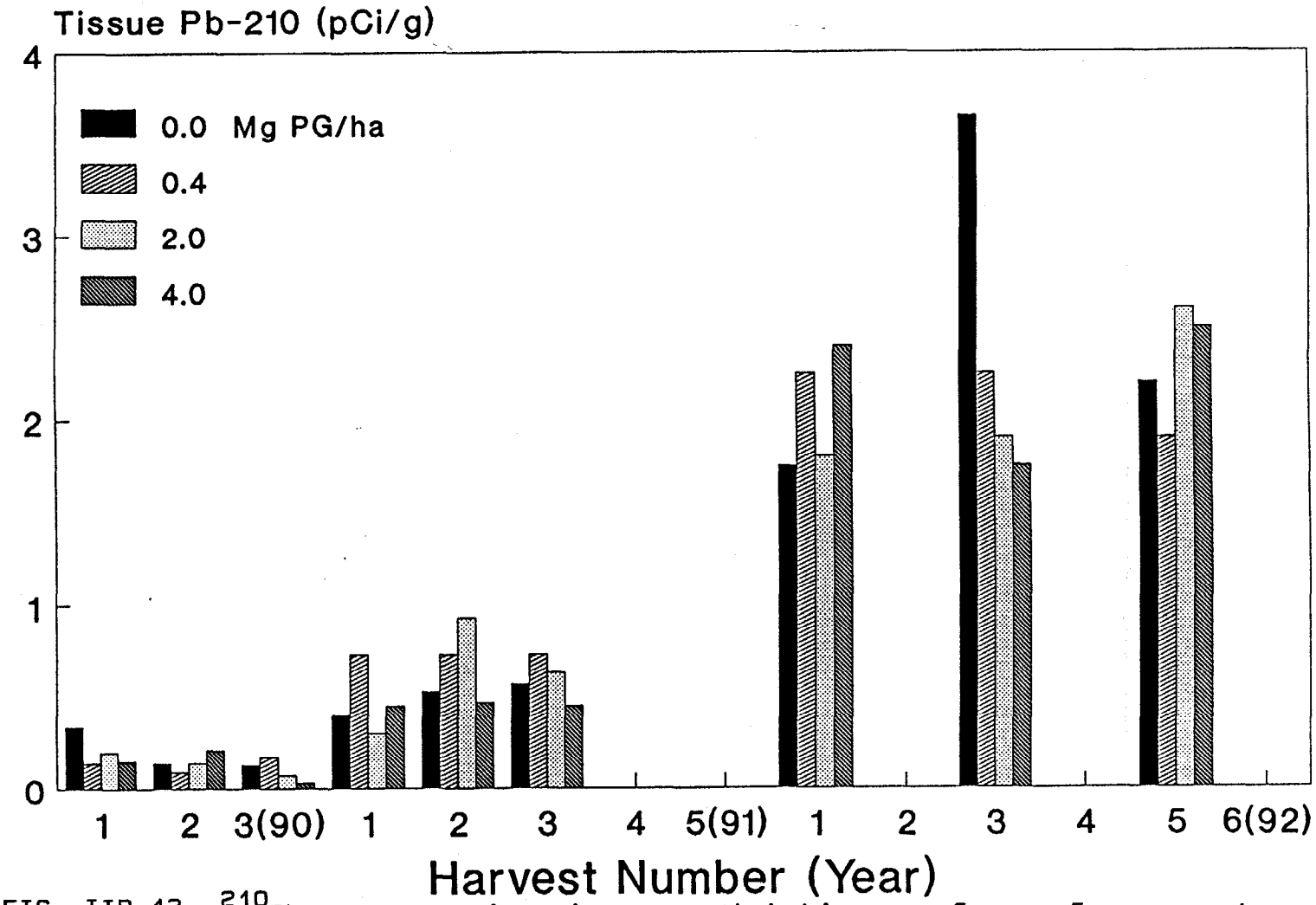

FIG. IIB-12. ^{210}Pb concentrations in regrowth bahiagrass forage from a pasture amended with PG as source of S and Ca and growing on a Florida Spodosol soil, 1990-1992, by harvest.

Control plot concentrations in regrowth forage averaged 0.93 pCi g^{-1}, with annual averages ranging from 0.2 to 2.5 and individual harvests ranging from 0.13 to 3.65. Concentrations in mature forage harvested annually averaged 1.33 pCi g^{-1} and ranged from 0.75 to 2.95. However, the order of magnitude increase in control plot concentrations from 1990 to 1992 raises questions about possible bias by year in the analyses.

Statistically-significant differences were noted in regrowth forage for three of the nine harvests and two of the three annual averages, but there was no consistency and the values appeared to have little relation to PG rates. For the mature forage (Table B5, B), there was no apparent trend with treatment level and no statistically-significant differences. No effect of ^{210}Pb uptake by bahiagrass regrowth or mature forage from PG application could be observed from these data.

Polonium-210. Annual average and 3-year average concentrations in regrowth forage and annual concentrations and 3-year average concentrations for the mature forage are presented in Table B6. Results for individual regrowth harvests are presented in Figure IIB-13. Analytical and statistical testing results for individual harvests are presented in Appendix Tables IIB-5, IIB-6, and IIB-7.

Control plot values were highly variable from year to year. Baseline concentrations in regrowth forage averaged 0.33 pCi g^{-1}, with annual averages ranging from 0.04 to 0.52 and individual harvests ranging from below the limit of detection to 0.64. Concentrations in annual mature forage harvests averaged 0.57 pCi g^{-1} and ranged from below the limit of detection to 0.73. For comparison, Mullins and Mitchell (1990) reported ^{210}Po concentrations ranging from 0.08 to 0.44 pCi g^{-1} in wheat forage from plots not treated with PG.

There is limited evidence for a treatment effect in the first year after treatment. By inspection, the concentration in regrowth tissue appears to increase with PG in the 1990 (and possibly 1992) samplings. Differences were statistically significant for two of the three 1990 crop-year harvests (Appendix Table IIB-5) and the 1990 average showed evidence of linear response (Table B6, P{linear} = 0.06). In addition, the 1992 annual average for the maximum (4.0 Mg ha^{-1}) treatment level was significantly greater than for the other treatments.

The 1991 regrowth forage results appear to be anomalous - the values observed for the control and treated plots were, for the most part, considerably higher than for the other two crop years and the concentrations increased from the control to the 0.4 Mg yr^{-1} treatment, and then decreased with treatment level for the higher treatment values!

Table B6. ^{210}Po concentrations in bahiagrass regrowth and hay forage from a pasture amended with PG as a source of S and Ca and growing on a Florida Spodosol soil, averaged by year and over a 3-year period, 1990-1992.

Treatment	Crop year			
	1990[a]	1991[a]	1992[a]	Mean[a]
A. Regrowth:	pCi ^{210}Po g^{-1}			
Mg PG ha^{-1}				
0.0(C)	0.04	0.52	0.10	0.33
0.4	0.10	0.73	0.10	0.48
2.0	0.19	0.51	0.10	0.38
4.0	0.22	0.37	0.20	0.30
Statistics:				
P	0.1713	0.1754	0.0001[b]	0.3304
P(Linear)	0.0594	ns	ns	ns
DMRT	ns[1]	ns	4.0>all[b]	ns
B. Hay:	pCi ^{210}Po g^{-1}			
Mg PG ha^{-1}				
0.0(C)	0.73	0.27	-[c]	0.57
0.4	0.62	0.59	0.20	0.61
2.0	0.50	0.78	0.10	0.61
4.0	0.87	0.85	0.10	0.86
Statistics:				
P	0.6506	0.3957	0.0001[b]	0.4750
DMRT	ns	ns	0.4>2.0, 4.0[b]	ns

[1]ns=not significant. [a]Statistics computed based solely on those harvests where each [b]treatment had at least one detected value associated with it. [b]Significance was due to MSE being very close to zero. [c]Not detected in sample.

With regard to hay forage (Table B6, B), the highest treatment level (4.0 Mg ha^{-1}) appears to produce an effect. However, differences were either not statistically significant (1990, 1991, and the 3-year average) or not meaningful in terms of PG level (1992).

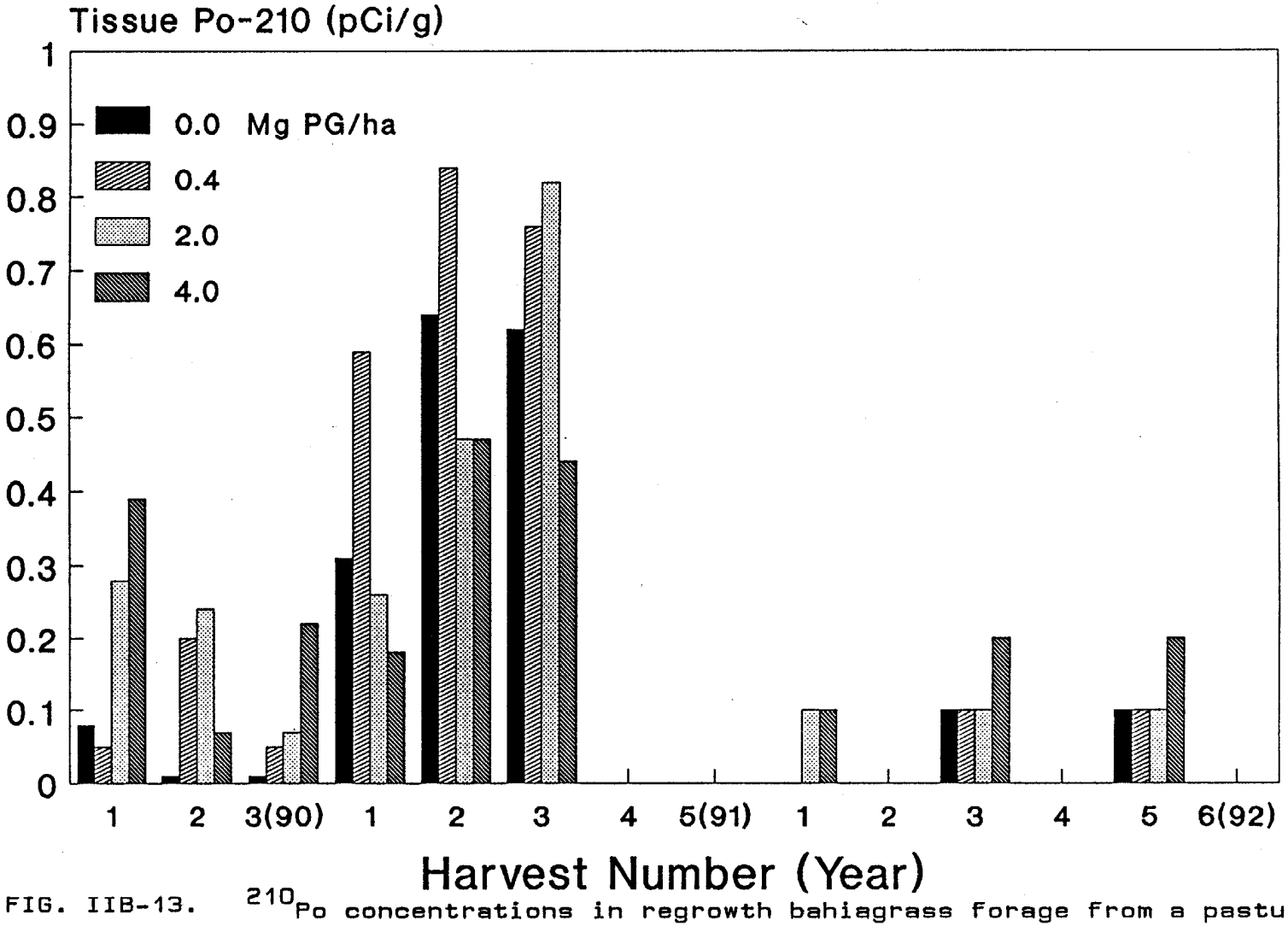

FIG. IIB-13. ^{210}Po concentrations in regrowth bahiagrass forage from a pasture amended with PG as source of S and Ca and growing on a Florida Spodosol soil, 1990-1992, by harvest.

B.5. Radionuclides in Surficial Groundwater.

Surficial groundwater from the experimental plots was collected at three depths: at the surface as runoff, at 60 cm, and 90 cm below the pasture surface. For the 60 cm depth, only one sample from each plot (July 1991) was analyzed for radionuclides. For the 3-year period, samples collected at 120 cm depth at six different times were analyzed for radionuclides.

Radium-226. Annual average and 3-year average concentrations are presented in Table B7. Concentrations are presented by individual sampling date for runoff, the 60-cm depth, and the 120-cm depth in Figures IIB-14, IIB-15, and IIB-16, respectively, and Appendix Tables IIB-8, IIB-9, and IIB-10, respectively.

For runoff, baseline (control plot) concentrations averaged 0.19 pCi L^{-1} with individual samplings ranging from 0.11 to 0.40. Concentrations from PG-treated plots ranged from 0.07 to 0.80 pCi L^{-1}. Inspection of the data suggests effects following a single application that persist into the third year and effects from annual application that cumulate into the third year. Differences were statistically significant for the first two of four sampling.

At the 60-cm depth, the control plot concentrations for the single sampling were slightly higher than for runoff - 0.30 and 0.40 pCi L^{-1} for the external and internal controls, respectively. Concentrations in samples from PG-treated plots ranged from 0.40 to 0.70 pCi L^{-1}. These data suggest a small effect with treatment (see Figure IIB-15) but the differences were not statistically significant.

At the 120-cm depth, control plot concentrations were slightly higher than at the surface and at 60 cm. Concentrations from the external control plot averaged 0.63 pCi L^{-1}, with the five individual samplings ranging from 0.35 to 0.80. Concentrations from the internal control plots were comparable, averaging 0.80 pCi L^{-1} with the six individual samplings ranging from 0.55 to 1.17. Concentrations at PG-treated plots ranged from 0.65 to 1.80 pCi L^{-1}. Concentrations for the treated plots were generally higher than for the control plots through the third year (Fig. IIB-16); however, averages did not correlate well with treatment level and the differences were not statistically significant.

The maximum ^{226}Ra concentrations observed in runoff and shallow well water from treated plots were 0.8 and 1.8 pCi L^{-1}, respectively. These values are below the current drinking water standard of 5 pCi L^{-1} (Federal Register, 1976) and well below the proposed Maximum Contaminant Level (MCL) of 20 pCi L^{-1} (Federal Register, 1991).

Table B7. ^{226}Ra concentrations in groundwater from a Florida Spodosol soil cropped to bahiagrass which was amended with PG as source of S and Ca, averaged by year and over a 3-year period, 1990-1992.

Depth (cm)/ Treatment	Crop year 1990	1991	1992	Mean
A. Runoff:	---------- pCi ^{226}Ra L^{-1} ----------			
Mg PG ha^{-1}				
0.0(C)	0.11	0.20	0.27	0.19
0.4	0.13	0.35	0.50	0.37
2.0	0.07	0.40	0.20	0.19
4.0	0.42	0.35	0.47	0.42
Statistics:				
P	0.0329	0.0001[a]	0.4547	0.5353
P(Linear)	0.0165	ns[1]	ns	ns
P(Quad.)	0.0359	ns	ns	ns
DMRT	4.0>all	0.4,4.0> C;2.0>all	ns	ns
B. 60 cm depth:	----------pCi ^{226}Ra L^{-1} ----------			
Mg PG ha^{-1}				
0.0(C_{ex})	-[b]	0.30	-[b]	0.30
0.0(C)	-[b]	0.40	-[b]	0.40
0.4	-[b]	0.60	-[b]	0.60
2.0	-[b]	0.55	-[b]	0.55
4.0	-[b]	0.70	-[b]	0.70
Statistics:				
P(w/C_{ex})	-[b]	0.4117	-[b]	0.4117
P(w/o C_{ex})	-[b]	0.4642	-[b]	0.4642
C. 120 cm depth:	---------- pCi ^{226}Ra L^{-1} ----------			
Mg PG ha^{-1}				
0.0(C_{ex})	0.70	0.65	0.57	0.63
0.0(C)	0.69	0.86	0.73	0.80
0.4	1.35	1.36	0.97	1.23
2.0	0.93	1.59	0.87	1.13
4.0	1.12	1.11	1.75	1.33
Statistics:				
P(w/C_{ex})	0.7096	0.1111	0.3518	0.4495
P(w/o C_{ex})	0.6111	0.2868	0.4024	0.5931
P(Quad.)	ns	0.0611	ns	ns

[1]ns=not significant. [a]Difference due to estimated error being too small. [b]At the 60-cm depth, only the 1991 samples were analyzed for radionuclides.

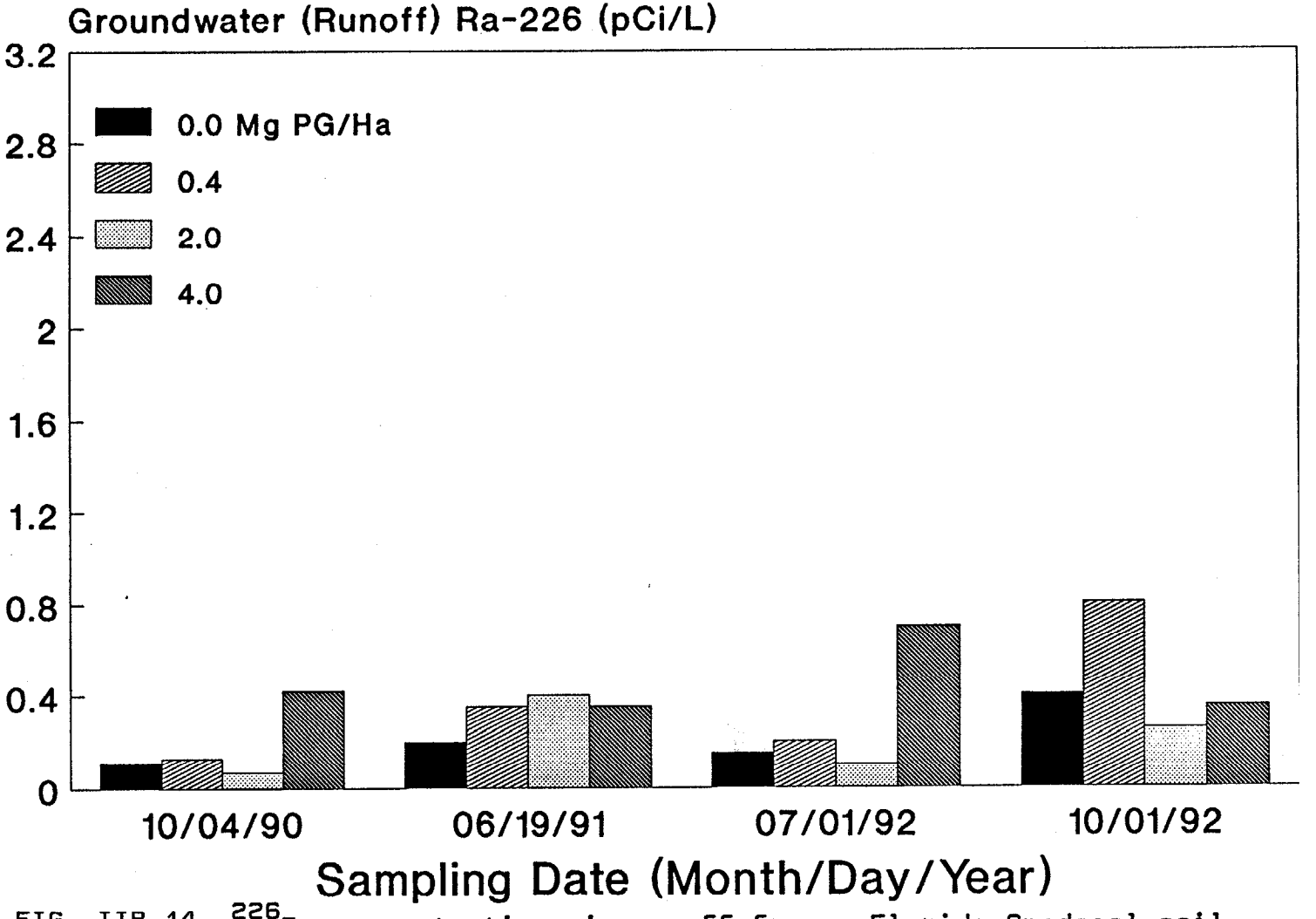

FIG. IIB-14. ^{226}Ra concentrations in runoff from a Florida Spodosol soil cropped to bahiagrass which was amended with PG as source of S and Ca, 1990-1992, by sampling.

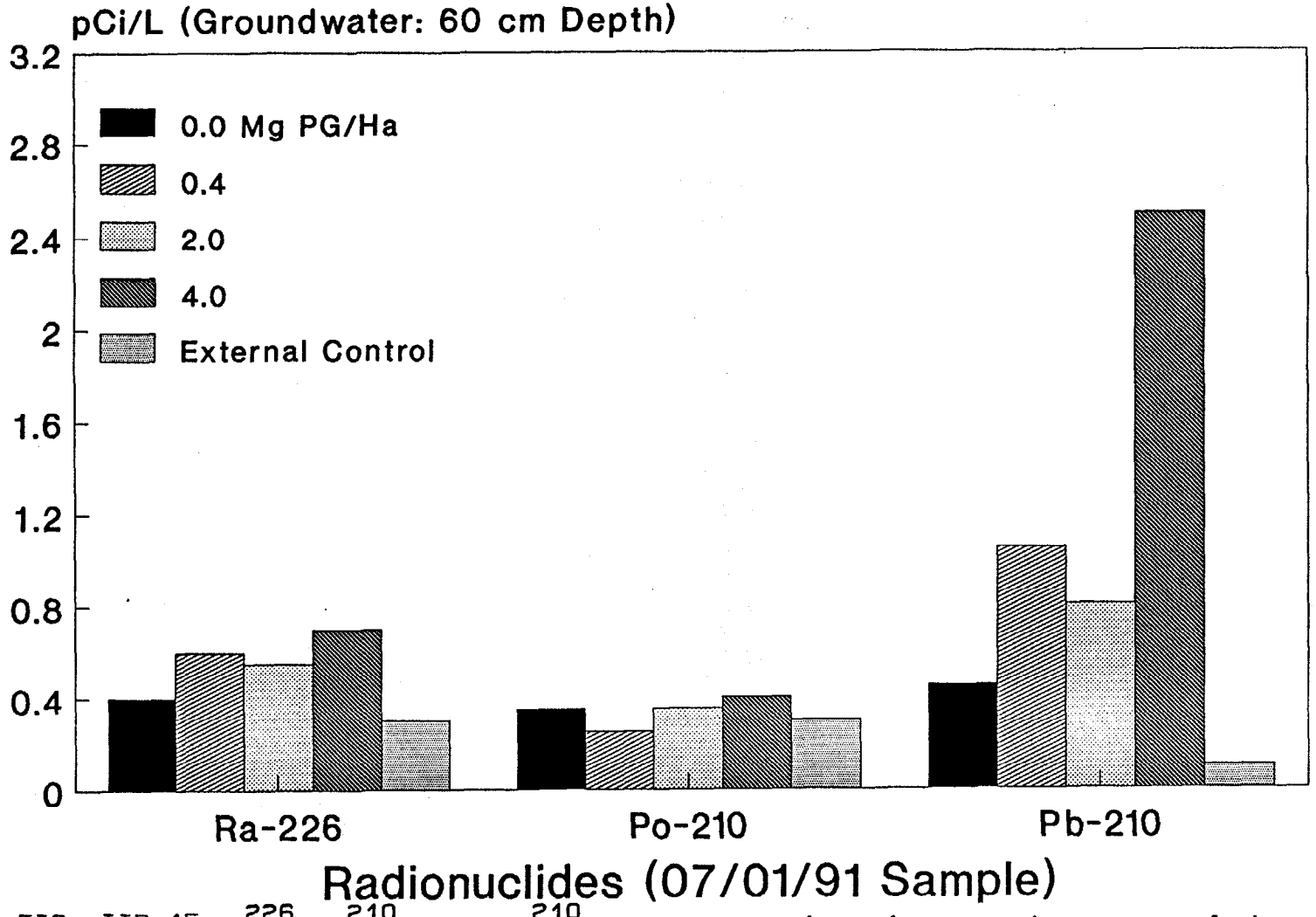

FIG. IIB-15. ^{226}Ra, ^{210}Pb, and ^{210}Po concentrations in groundwater sampled at 60 cm depth from a Florida Spodosol soil cropped to bahiagrass which was amended with PG as source of S and Ca, 1991.

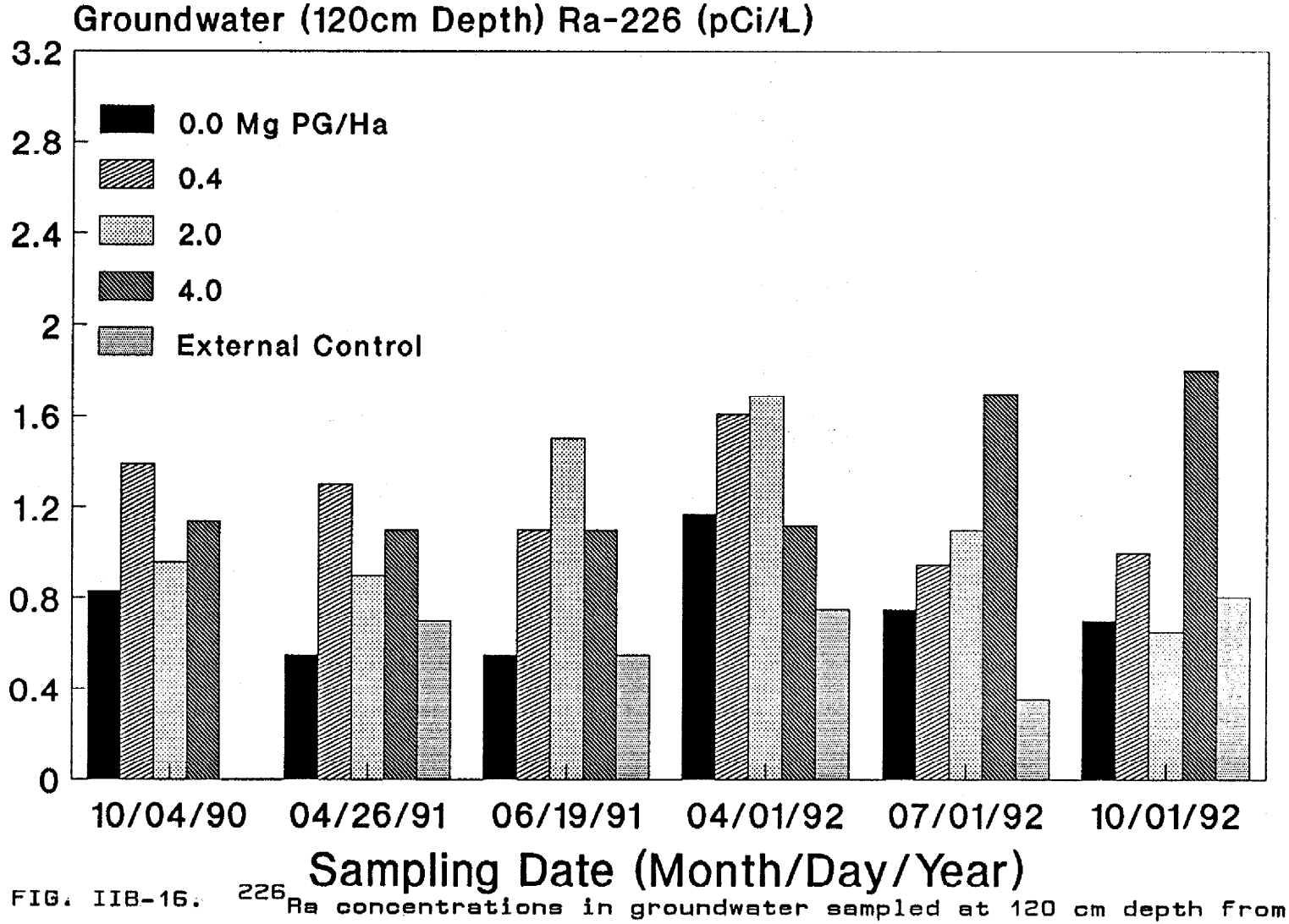

FIG. IIB-16. ^{226}Ra concentrations in groundwater sampled at 120 cm depth from a Florida Spodosol soil cropped to bahiagrass which was amended with PG as source of S and Ca, 1990-1992, by sampling.

Lead-210. Annual average and 3-year average concentrations are presented in Table B8. Concentrations are presented by individual sampling date for runoff, the 60-cm depth, and the 120-cm depth in Figures IIB-17, IIB-15, and IIB-18, respectively, and Appendix Tables IIB-8, IIB-9, and IIB-10, respectively.

For runoff, baseline (control plot) concentrations averaged 0.45 pCi L^{-1} but the four individual samplings were quite variable, ranging from 0.05 to 1.10. Concentrations from PG-treated plots ranged from 0.00 to 1.25 pCi L^{-1}. No significant differences were observed. Effects, if any, were transient. At the first sampling, the concentration reported for the maximum treatment level was eight times the very low control value but the difference was not statistically significant and was not seen in later samplings. At the 7/01/92 sampling, the value for plots treated annually at 0.4 Mg ha^{-1} was about six times the relatively low control value but the difference was not statistically significant and was not supported by any of the other samplings.

At the 60-cm depth, the control plot concentrations for the single sampling were comparable to those for runoff - 0.10 and 0.45 pCi L^{-1} for the external and internal controls, respectively. Concentrations in samples from PG-treated plots ranged from 0.45 to 2.50 pCi L^{-1}. The data indicated a statistically significant effect with linear response with PG level (P = 0.03, P{linear} = 0.008), 4.0 > all); however, this single sampling, not strongly supported by observations in the runoff, is a limited basis for any definite conclusion.

At the 120-cm depth, control plot concentrations were comparable to those at the other sampling levels. Values from the external control plot averaged 0.54 pCi L^{-1} with the five individual samplings ranging from 0.00 to 1.09 and those from the internal control plots averaged 0.42 pCi L^{-1} with the six individual samplings ranging from 0.15 to 0.80. Concentrations at PG-treated plots ranged from 0.05 to 2.40 pCi L^{-1}. The data (Figure IIB-18) suggest that the treated plots had higher levels than the control plots in the second and third years: however, these suggested effects did not correlate well with treatment level and the only statistically-significant differences were for the 2.0 Mg ha^{-1} level at the 4/26/91 sampling (Table B10) and in the 3-year average (Table B8).

Currently there is no drinking water standard for ^{210}Pb (a naturally-occurring beta emitter). The proposed MCL for beta emitters is the concentration resulting in an annual effective dose equivalent of 4 mrem yr^{-1} (Federal Register, 1991). The ^{210}Pb concentration that corresponds to this dose limit is 1 pCi L^{-1}, based on contemporary dosimetry and a 2 L day^{-1} ingestion. Of the four runoff, two 60-cm, and 11 120-cm control plot samplings, ^{210}Pb concentrations exceeded 1 pCi L^{-1} for one external control

Table B8. ^{210}Pb concentrations in groundwater from a Florida Spodosol soil cropped to bahiagrass which was amended with PG as source of S and Ca, averaged by year and over a 3-year period, 1990-1992.

Depth (cm)/ Treatment	Crop year 1990	1991	1992	Mean
A. Runoff:	---------- pCi ^{210}Pb L^{-1} ----------			
Mg PG ha				
0.0(C)	0.05	1.00	0.50	0.45
0.4	0.17	1.10	1.10	0.87
2.0	0.00	0.60	0.60	0.36
4.0	0.41	0.60	0.20	0.40
Statistics:				
P	0.2455	0.3579	0.1179	0.1981
DMRT	ns[1]	ns	0.4>4.0	ns
B. 60 cm depth:	----------pCi ^{210}Pb L^{-1} ----------			
Mg PG ha^{-1}				
0.0(C_{ex})	-[a]	0.10	-[a]	0.10
0.0(C)	-[a]	0.45	-[a]	0.45
0.4	-[a]	1.05	-[a]	1.05
2.0	-[a]	0.80	-[a]	0.80
4.0	-[a]	2.50	-[a]	2.50
Statistics:				
P(w/C_{ex})	-[a]	0.0308	-[a]	0.0308
P(w/o C_{ex})	-[a]	0.0261	-[a]	0.0261
P(Linear)	-[a]	0.0083	-[a]	0.0083
DMRT	-[a]	4.0>all	-[a]	4.0>all
C. 120 cm depth:	----------pCi ^{210}Pb L^{-1} ----------			
Mg PG ha^{-1}				
0.0(C_{ex})	0.00	0.87	0.30	0.54
0.0(C)	0.31	0.57	0.45	0.42
0.4	0.36	0.86	0.55	0.60
2.0	0.34	1.63	1.13	1.03
4.0	0.25	1.02	1.03	0.77
Statistics:				
P(w/C_{ex})	0.3109	0.4793	0.5928	0.2450
P(w/o C_{ex})	0.6734	0.2620	0.4903	0.1582
P(Quad.)	ns	ns	ns	0.0731
DMRT	ns	ns	ns	2.0>C

[1]ns=not significant. [a]At the 60-cm depth, only the 1991 samples were analyzed for radionuclides.

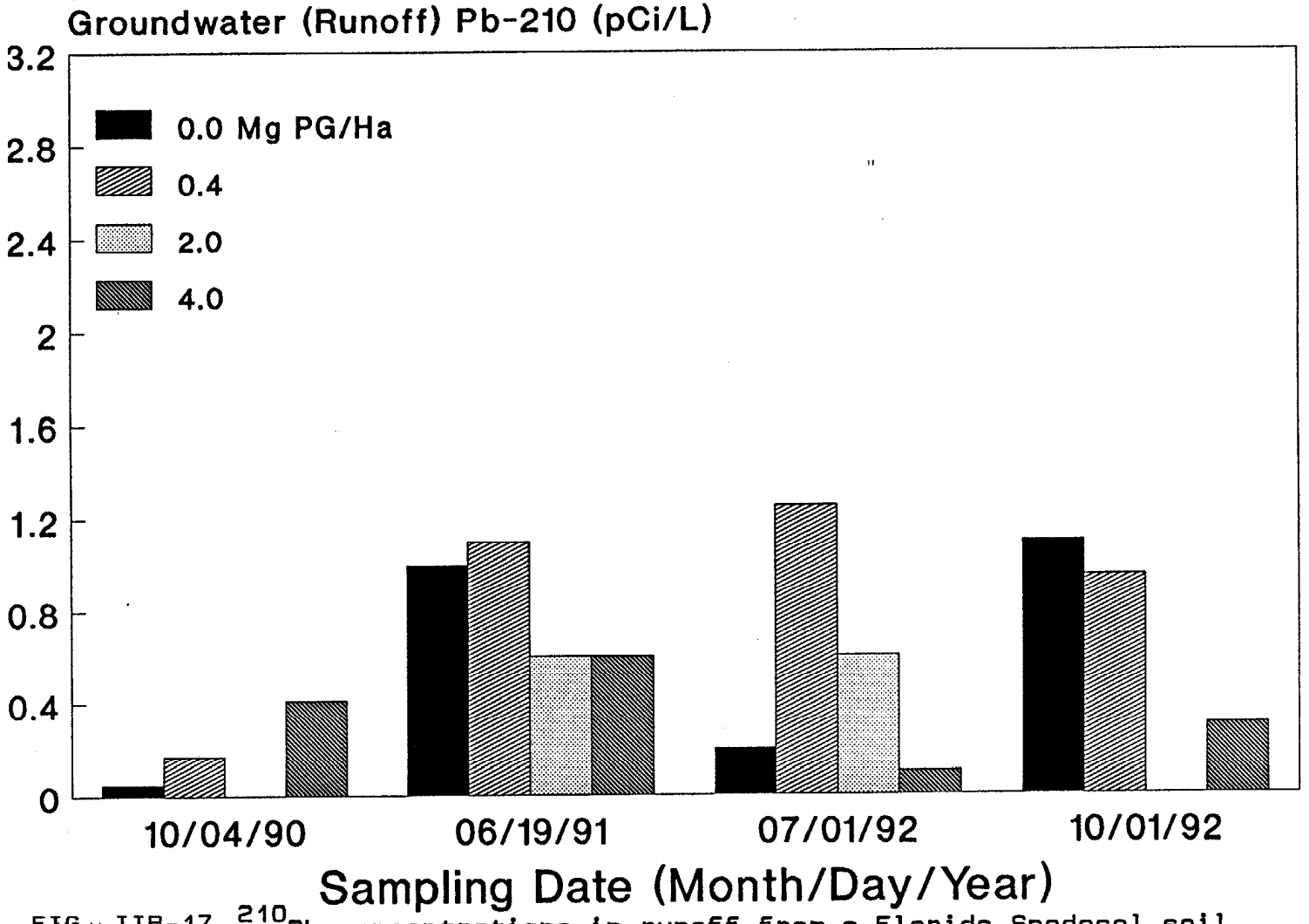

FIG. IIB-17. ^{210}Pb concentrations in runoff from a Florida Spodosol soil cropped to bahiagrass which was amended with PG as source of S and Ca, 1990-1992, by sampling.

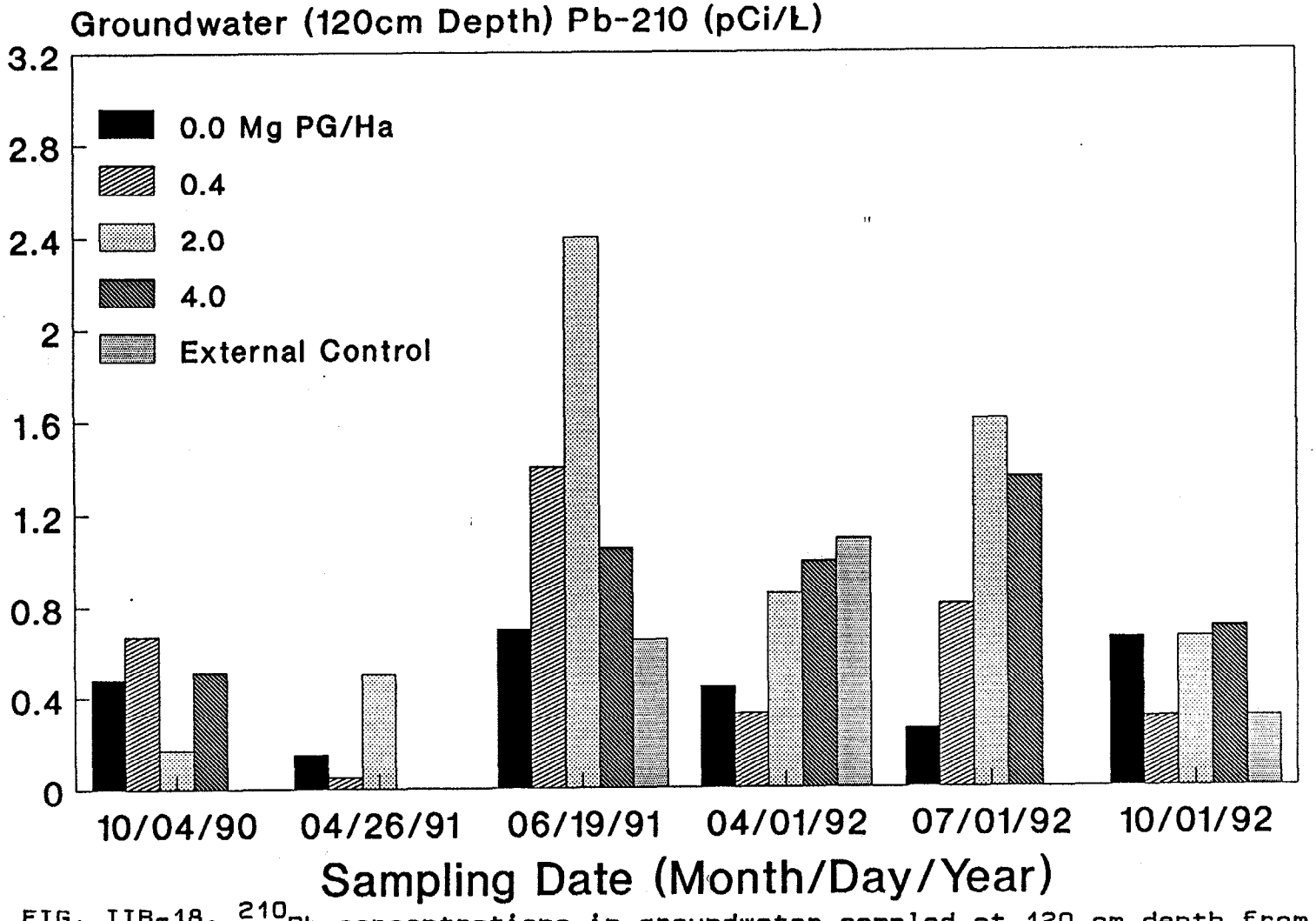

FIG. IIB-18. ^{210}Pb concentrations in groundwater sampled at 120 cm depth from a Florida spodosol soil cropped to bahiagrass which was amended with PG as source of S and Ca, 1990-1992, by sampling.

sampling (1.1 pCi L^{-1}). For PG-treated plots, ^{210}Pb concentrations exceeded 1 pCi L^{-1} for two of the three samplings at 60 cm (1.1-2.5 pCi L^{-1}) and five of the 18 samplings at 120 cm (1.1-2.4 pCi L^{-1}). While there was very little statistical evidence for a treatment effect, these data indicate that potential for ^{210}Pb in water deserves continued evaluation.

Polonium-210. Annual average and 3-year average concentrations are presented in Table B9. Concentrations are presented by individual sampling date for runoff, the 60-cm depth, and the 120-cm depth in Figures IIB-19, IIB-15, and IIB-20, respectively, and Appendix Tables IIB-8, IIB-9, and IIB-10, respectively.

For runoff, control plot concentrations averaged 0.44 pCi L^{-1} and the four individual samplings ranged from 0.37 to 0.60. Concentrations from PG-treated plots ranged from 0.10 to 1.30 pCi L^{-1}. No significant effects were observed.

At the 60-cm depth, the control plot concentrations for the single sampling were slightly lower than observed for runoff - 0.30 and 0.35 pCi L^{-1} for the external and internal controls, respectively. Concentrations in samples from PG-treated plots ranged from 0.24 to 0.40 pCi L^{-1}; no treatment effects were observed.

At the 120-cm depth, concentrations from the external control plot averaged 0.59 pCi L^{-1} with the five individual samplings ranging from below the detection limit to 0.90 and concentrations from the internal control plots averaged 1.3 pCi L^{-1} with the six individual samplings ranging from 0.10 to 2.85. The most striking feature at this depth is the variation in control plot concentrations - the values for the internal controls varied considerably from year to year, had yearly averages that were several times the corresponding values for the external control, and, for three of the samplings, exceeded the values for the treated plots. Concentrations at PG-treated plots ranged from below the limit of detection to 1.80 pCi L^{-1}. The data show no meaningful effects of PG treatment on ^{210}Po.

There are no current or proposed explicit drinking water standards for ^{210}Po (a naturally-occurring alpha emitter); rather this radionuclide is included under limits for gross alpha emitters. The current standard specifies a limit for gross alpha emitters of 15 pCi L^{-1} (Federal Register, 1976). Under proposed rules (Federal Register, 1991), the MCL for "adjusted" gross alpha activity (excluding ^{226}Ra, uranium, and ^{222}Rn) is also 15 pCi L^{-1}. All the ^{210}Po concentrations reported in this study were well below this value. The maximum observed was 2.8 pCi L^{-1} in an internal control sampling at 120 cm. The maximums for PG-treated plots were 1.3 and 1.5 pCi L^{-1} for runoff and shallow ground water, respectively.

Table B9. ^{210}Po concentrations in groundwater from a Florida Spodosol soil cropped to bahiagrass which was amended with PG as source of S and Ca, averaged by year and over a 3-year period, 1990-1992.

Depth (cm)/ Treatment	Crop year 1990	1991	1992	Mean
A. Runoff:	---------- pCi ^{210}Po L^{-1} ----------			
Mg PG ha^{-1}				
0.0(C)	0.37	0.40	0.50	0.44
0.4	0.25	1.30	0.23	0.54
2.0	0.29	0.60	0.23	0.31
4.0	0.41	0.40	0.27	0.35
Statistics:				
P	0.8963	0.4925	0.7359	0.6171
B. 60 cm depth:	---------- pCi ^{210}Po L^{-1} ----------			
Mg PG ha^{-1}				
0.0(C_{ex})	-[a]	0.30	-[a]	0.30
0.0(C)	-[a]	0.35	-[a]	0.35
0.4	-[a]	0.25	-[a]	0.25
2.0	-[a]	0.35	-[a]	0.35
4.0	-[a]	0.40	-[a]	0.40
Statistics:				
P(w/C_{ex})	-[a]	0.5201	-[a]	0.5201
P(w/o C_{ex})	-[a]	0.5000	-[a]	0.5000
C. 120 cm depth:	---------- pCi ^{210}Po L^{-1} ----------			
Mg PG ha^{-1}				
0.0(C_{ex})	0.75	0.57	0.45	0.59
0.0(C)	1.13	1.51	1.93	1.30
0.4	0.50	0.79	0.15	0.55
2.0	0.57	0.94	0.55	0.72
4.0	1.28	1.01	0.53	0.98
Statistics:				
P(w/C_{ex})	0.0459	0.7017	0.6301	0.4505
P(w/o C_{ex})	0.1103	0.7537	0.5860	0.4655
P(Quad.)	0.0376	ns[1]	ns	ns
DMRT	4.0>0.4, 2.0	ns	ns	ns

[1]ns=Not significant. [a]For the 60-cm depth, only the 1991 samples were analyzed for radionuclides.

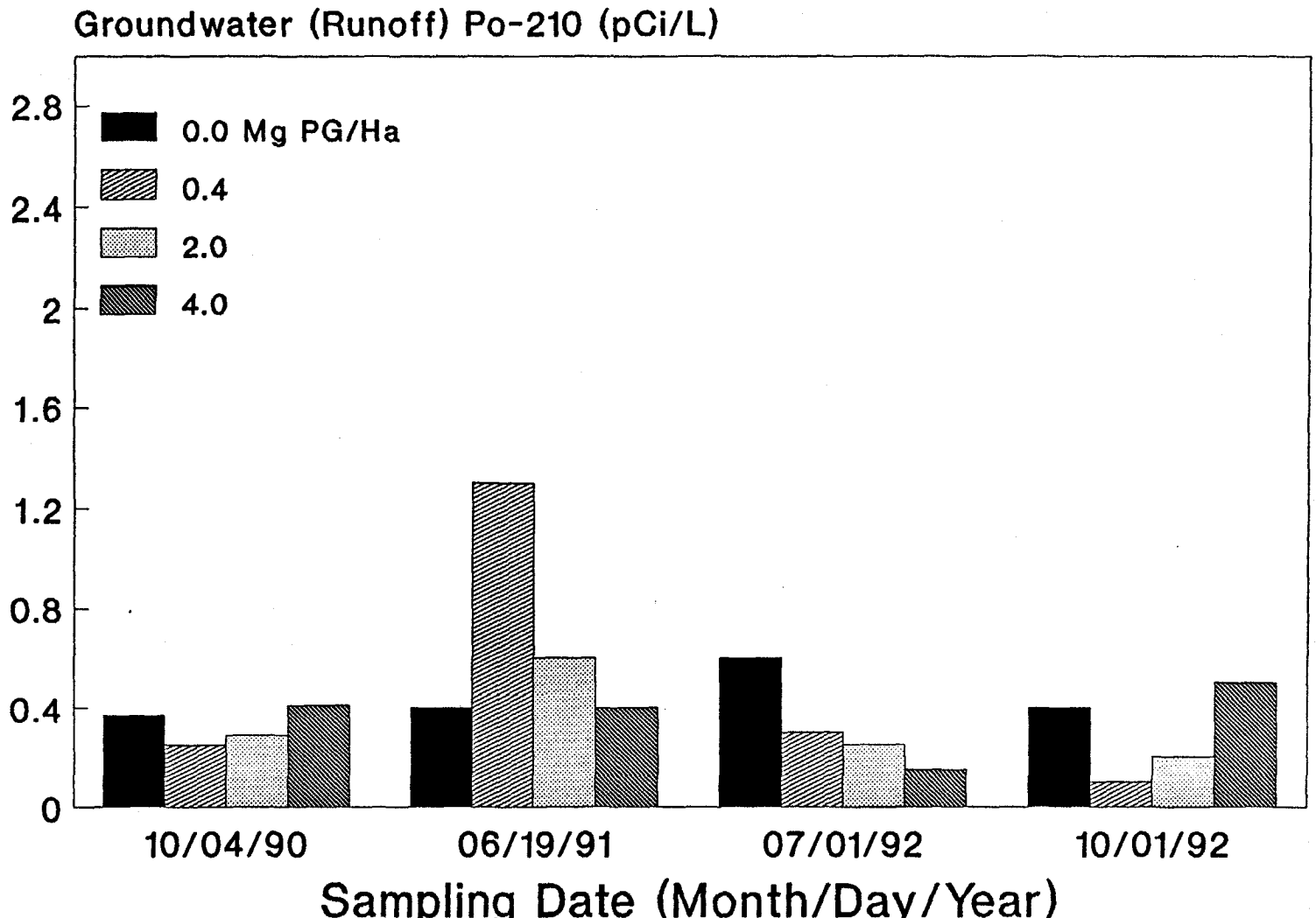

FIG. IIB-19. ^{210}Po concentrations in runoff from a Florida Spodosol soil cropped to bahiagrass which was amended with PG as source of S and Ca, 1990-1992, by sampling.

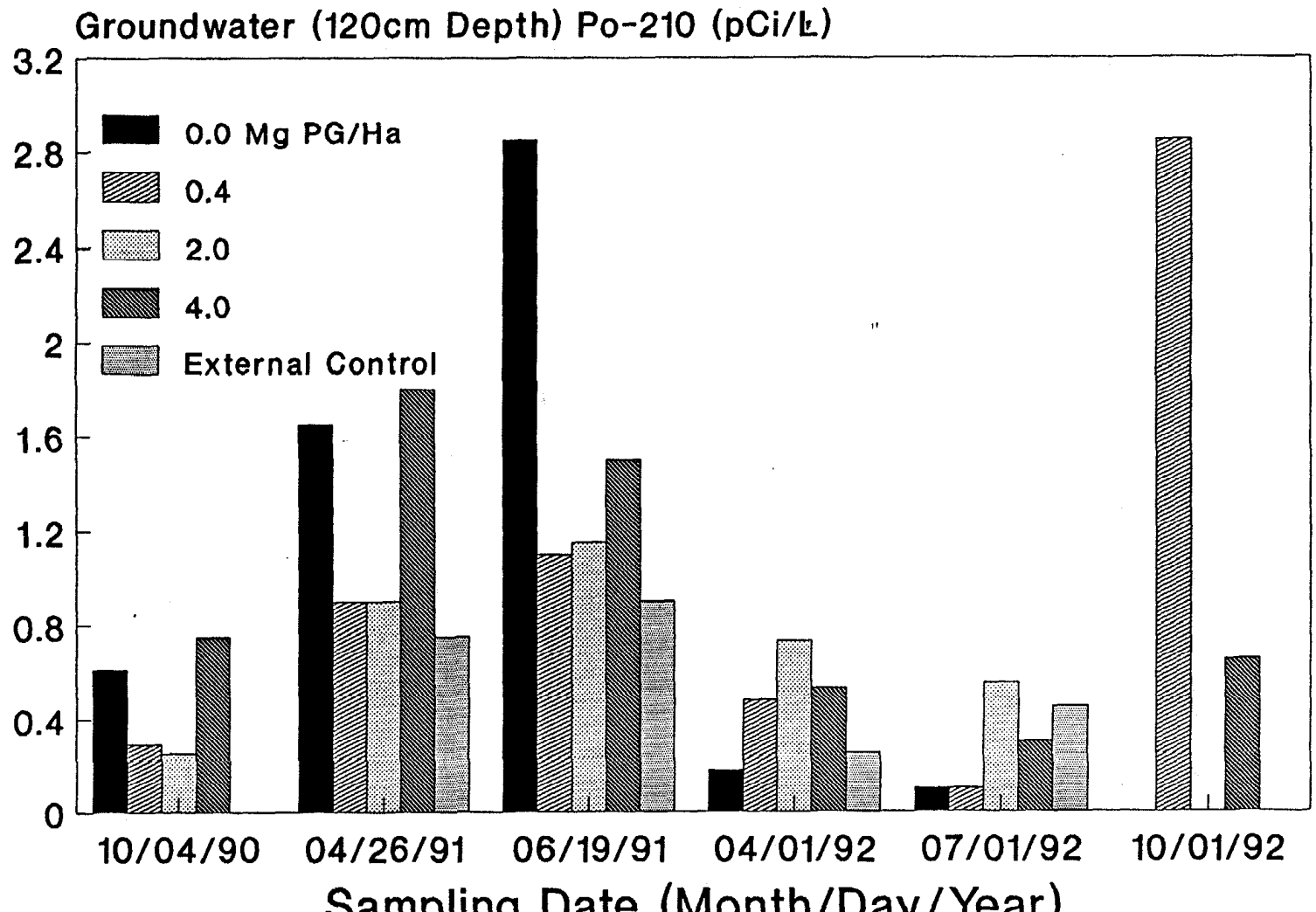

FIG. IIB-20. ^{210}Po concentrations in groundwater sampled at 120 cm depth from a Florida Spodosol soil cropped to bahiagrass which was amended with PG as source of S and Ca, 1990-1992, by sampling.

B.6. Soil Surface Rn Flux.

The Rn flux from the surface of the bahiagrass pasture was determined seven times during the 3-year period. Average values by crop year and for the 3-year period are presented in Table B10. The by-sampling statistics are given in the Appendix (Table IIB-11). The trends are graphically shown in Figure IIB-21.

Table B10. Soil surface Rn flux over a Florida Spodosol soil cropped to bahiagrass which was amended with PG as a source of S and Ca, averaged by year and over a 3-year period, 1990-1992.

Treatment	Crop year 1990-91	1991-92	1992	mean
Mg PG ha^{-1}	---------- pCi m^{-2} s^{-1} ----------			
0.0(C)	0.04	0.03	0.02	0.03
0.4	0.03	0.03	0.03	0.03
2.0	0.04	0.05	0.03	0.04
4.0	0.04	0.04	0.04	0.04
Statistics:				
P	0.2154	0.5120	0.2497	0.3211

The baseline Rn flux values for the control plots averaged 0.03 pCi m^{-2} s^{-1} over the entire observation period and ranged from 0.01 to 0.05.

For the PG-treated plots, the values ranged from 0.02 to 0.08 pCi m^{2} s^{-1}. The data for six of the seven measurements, two of the three crop-year averages, and the 3-year average suggested slightly higher values for the higher treatment level plots (2.0 and 4.0 Mg PG ha^{-1}) than for the control. However, none of the differences were statistically significant.

The values obtained for Rn flux were well within the range reported in the literature. Surface flux measurements have been reported to vary by a factor of 250, ranging from 0.005 to 1.41. The reported average for the U.S. is 0.43 pCi m^{-2} s^{-1} (Wilkening, et al., 1972; NCRPM, 1989).

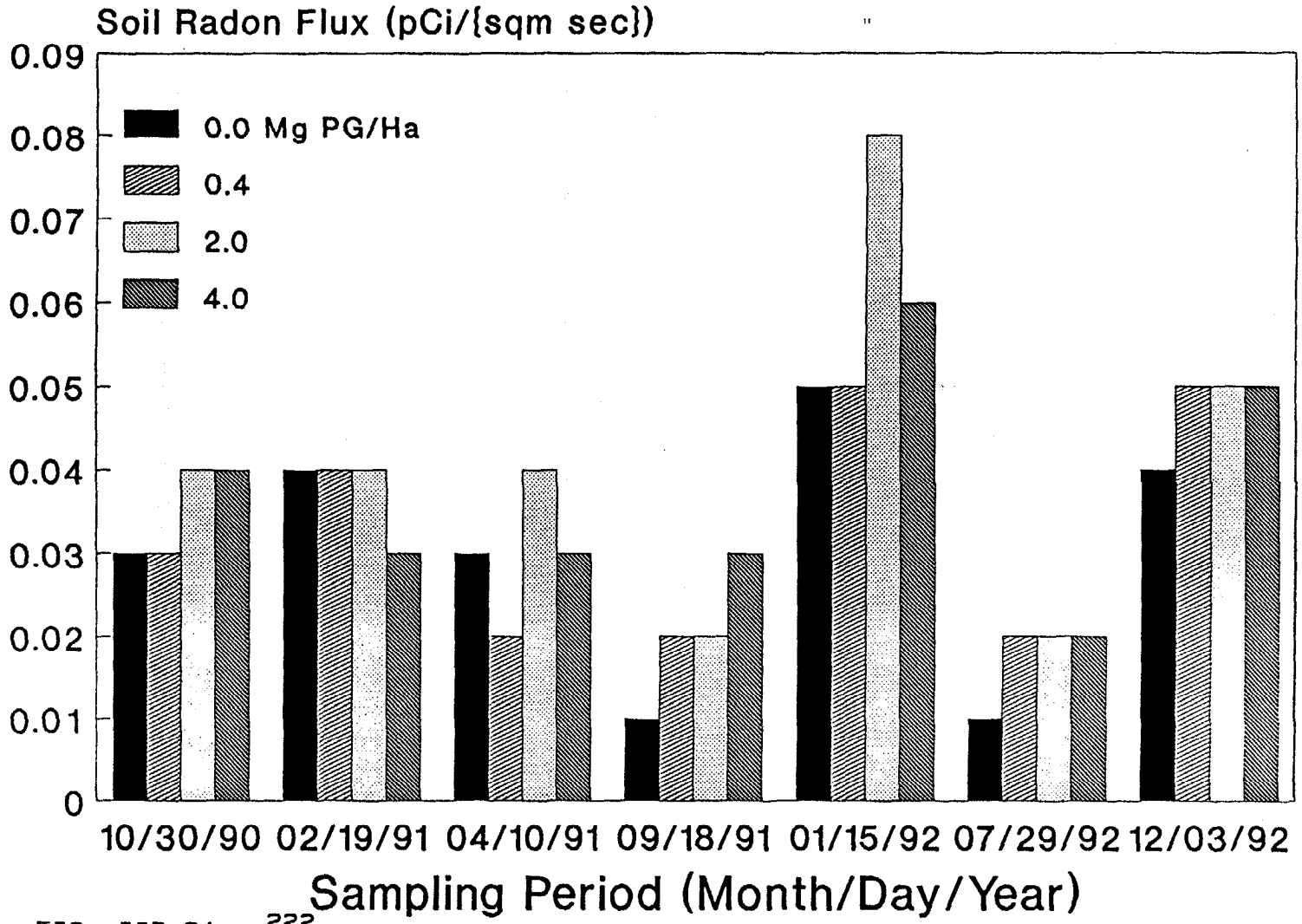

FIG. IIB-21. ^{222}Rn flux over a Florida Spodosol soil cropped to bahiagrass which was amended with PG as source of S and Ca, 1990-1992, by sampling.

B.7. Ambient Atmospheric Rn.

Airborne Rn concentrations were measured over control plots twice prior to application of the PG and over all plots determined 13 times after treatment application for periods ranging from 30 to 90 days, more or less, over the 3-year period. Table B11 summarizes the data by crop year and for the 3-year study: data for individual sampling periods are presented in Figure IIB-22 and in Appendix Table IIB-12.

Table B11. Ambient atmospheric Rn over a Florida Spodosol soil cropped to bahiagrass which was amended with PG as source of S and Ca, averaged by year and over a 3-year period, 1990-1992.

Treatment	Crop year			
	1990-91	1991-92	1992	mean
Mg PG ha^{-1}	---------- pCi L^{-1} ----------			
0.0(C_{ex1})	0.18	0.22	0.18	0.19
0.0(C_{ex2})	0.19	0.19	0.18	0.18
0.0(C)	0.22	0.22	0.20	0.20
0.4	0.27	0.26	0.19	0.24
2.0	0.19	0.24	0.20	0.21
4.0	0.20	0.21	0.21	0.20
Statistics:				
P(w/$C_{ex1,2}$)	0.0006	0.7810	0.9110	0.4557
P(w/o $C_{ex1,2}$)	0.0016	0.3910	0.8725	0.2891
P(Linear)	0.0015	ns[1]	ns	ns
P(Quadratic)	0.0758	ns	ns	ns
DMRT	0.4>all; C>2.0,4.0	ns	ns	ns

[1]ns=Not significant.

The background Rn concentration tends to vary with time, even when averaged over periods on the order of 30 to 90 days. During the two successive measurement periods in the pre-application period, concentrations were 0.22-0.23 and 0.12-0.13 pCi L^{-1}; there was no difference between the various external and internal control station measurements. During the post-application period, the averages and ranges for control station measurements were 0.19 (0.05 to 0.32) pCi L^{-1} for C_{ex1}, 0.18 (0.04 to 0.37) pCi L^{-1} for C_{ex2}, and 0.20 (0.09 to 0.43) for internal control C.

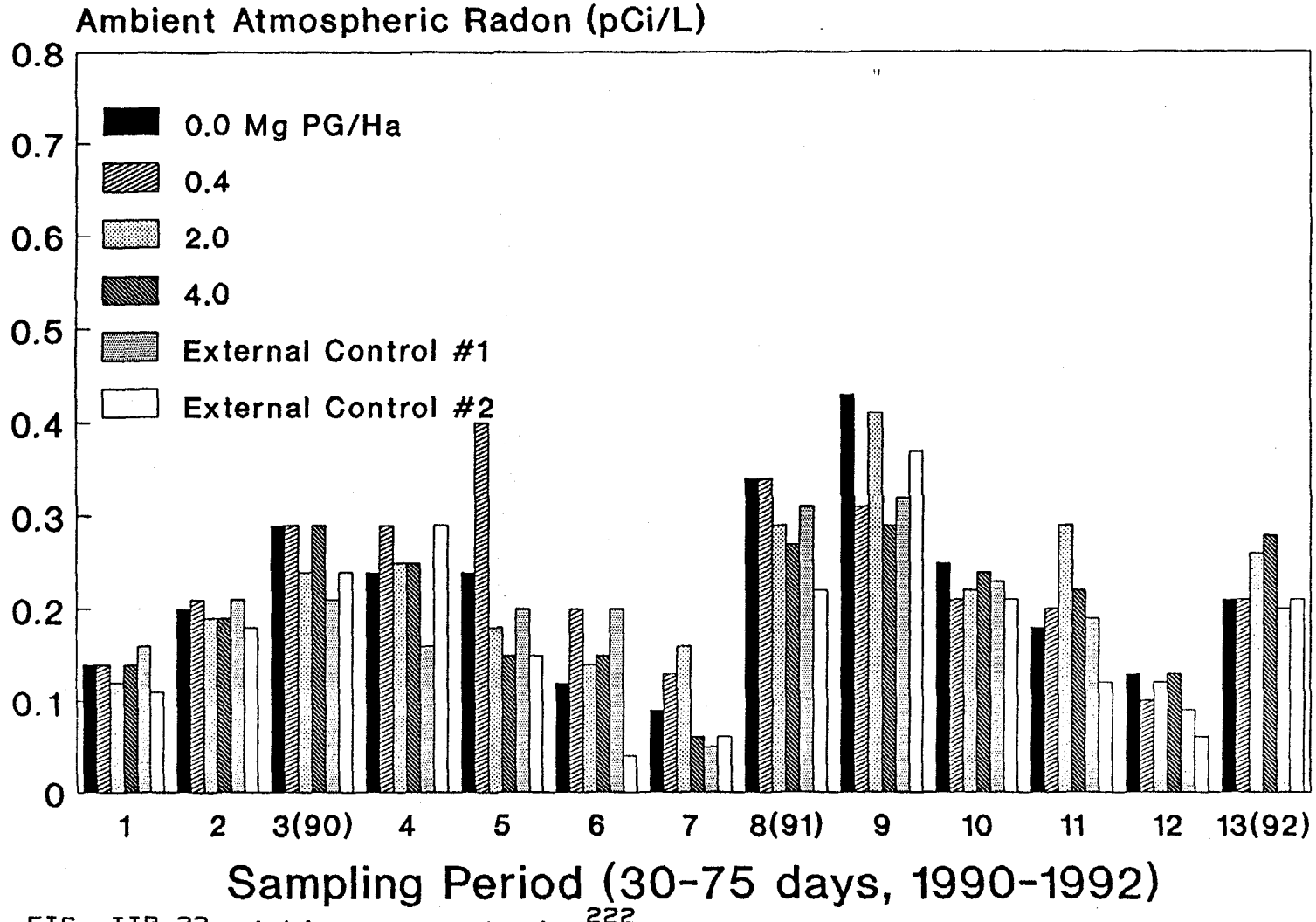

FIG. IIB-22. Ambient atmospheric ^{222}Rn over a Florida Spodosol soil cropped to bahiagrass which was amended with PG as source of S and Ca, 1990-1992, by sampling.

Concentrations over the PG-treated plots ranged from 0.06 to 0.40 pCi L^{-1}. Data were analyzed for treatment effects with and without the external controls for the 3-year average, crop-year averages, and individual sampling periods. Differences were statistically significant for about one third of the tests performed; however, there was no consistent trend with treatment level.

Inspection of the averages in Table B11 suggests that the concentrations at the external controls were slightly lower than for the internal control plots, indicates that within the contiguous plots there was no meaningful pattern with treatment level, and suggests that the concentrations over the contiguous plots in aggregate (control and treated) were slightly higher than for the external controls. Consequently, it is speculated that the treatment effect at the 1-m elevation, if any, is shared by the contiguous plots due to the relatively small area of the plots and the effect or air movement and mixing.

For comparison with another study that also used EIC's, individual outdoor ambient atmospheric Rn values for the U.S. were reported to range from 0.0 to 1.11 pCi L^{-1} with a median concentration of 0.39 pCi L^{-1}. Mean outdoor values ranged from 0.16 to 0.57 (Hopper et al., 1990).

C. RADIOLOGICAL ASPECTS ASSOCIATED WITH PG APPLICATION TO A TILLED LAND AS A SOURCE OF S AND Ca TO ANNUAL RYEGRASS

C.1. Radionuclides in PG

The same PG used in the bahiagrass experiment was used in the ryegrass study. The comments in the bahiagrass section B.1 also apply here and may be referred to.

C.2. Soil Radionuclides

Radionuclide concentrations by depth for the samples collected in 1990 prior to PG application are given in the Table C1; 3-year averages for post-treatment sampling are presented in Table C2. The results of analyses and statistical testing for post-treatment sampling for 1991, 1992, and 1993 are presented in Appendix Tables IIC-1, IIC-2, and IIC-3, respectively.

Radium-226. Results are compared by treatment assignment and depth for 1990 (pre-treatment), 1991, 1992, and 1993 in Figures IIC-1, IIC-2, IIC-3, and IIC-4, respectively.

Table C1 and Figure IIC-1 indicate that, with one possible exception, the natural ^{226}Ra concentrations prior to PG application were <1 pCi g^{-1} and comparable between the plot groups assigned to the various treatments. The value of 1.55 pCi g^{-1} at 75-90 cm for plots designated for the 4.0 Mg ha^{-1} treatment appears to be an "outlier". Statistically it was not significantly different from the other values and concentrations were <1 pCi g^{-1} in all subsequent sampling. The concentrations reported for the control plots (see Figures IIC-1 through IIC-4) show considerable variability with time - values reported for 1991 were in the range of 0.8 to 2.0 pCi g^{-1} while those for 1992 were in the range of 0.2 to 0.3 pCi g^{-1}. Since these concentrations should be relatively invariant, this suggests a considerable analytical variability associated with this measurement. Taking the average of all samples from untreated plots (pre-treatment samples from all plots and three years of post-treatment samples from control plots) as the best estimate indicates that the baseline soil ^{226}Ra concentration for the lands used in this study was low and relatively uniform over the 90-cm profile - about 0.6 pCi g^{-1} with individual 15-cm layers ranging from 0.5 to 0.7.

Table C1. ^{226}Ra, ^{210}Pb, and ^{210}Po concentrations in a Florida Spodosol soil before PG application, by depth, 1990.

Treatment	Depth (cm) 0-15	15-30	30-45	45-60	60-75	75-90
Mg PG ha^{-1}	pCi ^{226}Ra g^{-1}					
0.0(C)	0.70	0.80	0.60	0.50	0.75	0.65
0.4	0.85	0.45	0.45	0.85	0.60	0.70
2.0	0.70	0.70	0.65	0.65	0.95	0.40
4.0	0.60	0.65	0.80	0.65	0.65	1.55
Mean	0.71	0.65	0.62	0.66	0.74	0.82
Statistics:						
P	0.8215	0.9458	0.6598	0.4728	0.1941	0.3962
Mg PG ha^{-1}	pCi ^{210}Pb g^{-1}					
0.0(C)	0.90	-[a]	-[a]	0.70	0.30	1.60
0.4	1.00	1.30	0.80	1.45	1.90	1.60
2.0	0.60	-[a]	1.00	1.30	0.40	-[a]
4.0	-[a]	0.90	0.60	0.30	1.60	-[a]
Mean	0.64[b]	0.58[b]	0.61[b]	0.94	1.05	0.82[b]
Statistics:						
P	-[c]	-[c]	-[c]	0.0584	0.2721	-[c]
Mg PG ha^{-1}	pCi ^{210}Po g^{-1}					
0.0(C)	(2.25)[d]	0.80	0.45	0.75	0.25	0.55
0.4	0.80	0.65	1.40	1.30	0.36	0.70
2.0	1.05	0.80	0.80	1.10	0.35	0.20
4.0	0.30	0.50	1.25	0.45	0.75	1.20
Mean	0.72[e]	0.69	0.98	0.90	0.43	0.66
Statistics:						
P	0.6796	0.6949	0.4704	0.7446	0.3306	0.6402

[a]Not detected in samples. [b]In calculating the mean, non-detectible values were assigned ½ the lowest reported value. [c]No error term for analysis due to missing data. [d]This value is unusually high and not supported in subsequent year; extrapolating for years and depth estimates a value of 0.74 pCi g^{-1}. [e]Calculated using extrapolated value for 0.0 treatment.

Table C2. ^{226}Ra, ^{210}Pb, and ^{210}Po concentrations in a Florida Spodosol soil cropped to annual ryegrass which was amended with phosphogypsum (PG) as a source of S and Ca, averaged over a 3-year period, 1991-1993, by depth.

Treatment	Depth (cm)					
	0-15	15-30	30-45	45-60	60-75	75-90
Mg PG ha^{-1}	---------- pCi ^{226}Ra g^{-1} ----------					
0.0(C)	0.43	0.47	0.60	0.82	0.57	0.48
0.4	0.43	0.50	0.55	0.43	0.35	0.58
2.0	0.25	0.42	0.37	0.43	0.52	0.48
4.0	0.37	0.38	0.32	0.37	0.45	0.43
Statistics:						
P	0.7805	0.7781	0.7177	0.8185	0.8063	0.9571
Mg PG ha^{-1}	---------- pCi ^{210}Pb g^{-1} ----------					
0.0(C)	0.76	0.40	0.78	0.70	0.40	0.56
0.4	0.77	0.85	0.90	0.66	0.84	0.62
2.0	0.85	0.78	0.47	0.50	0.65	0.45
4.0	0.64	0.42	0.33	0.53	0.92	0.42
Statistics:						
P	0.9279	0.5073	0.2719	0.7827	0.3561	0.7586
Mg PG ha^{-1}	---------- pCi ^{210}Po g^{-1} ----------					
0.0(C)	0.38	0.30	0.30	0.15	0.28	0.30
0.4	0.37	0.30	0.23	0.42	0.34	0.26
2.0	0.68	0.22	0.23	0.15	0.33	0.15
4.0	0.45	0.27	0.18	0.17	0.35	0.28
Statistics:						
P	0.1705	0.7421	0.2964	0.6634	0.6858	0.8768
P(Quadratic)	0.0716	ns[1]	ns	ns	ns	ns
DMRT	2.0>0.4	ns	ns	ns	ns	ns

[1]ns=not significant.

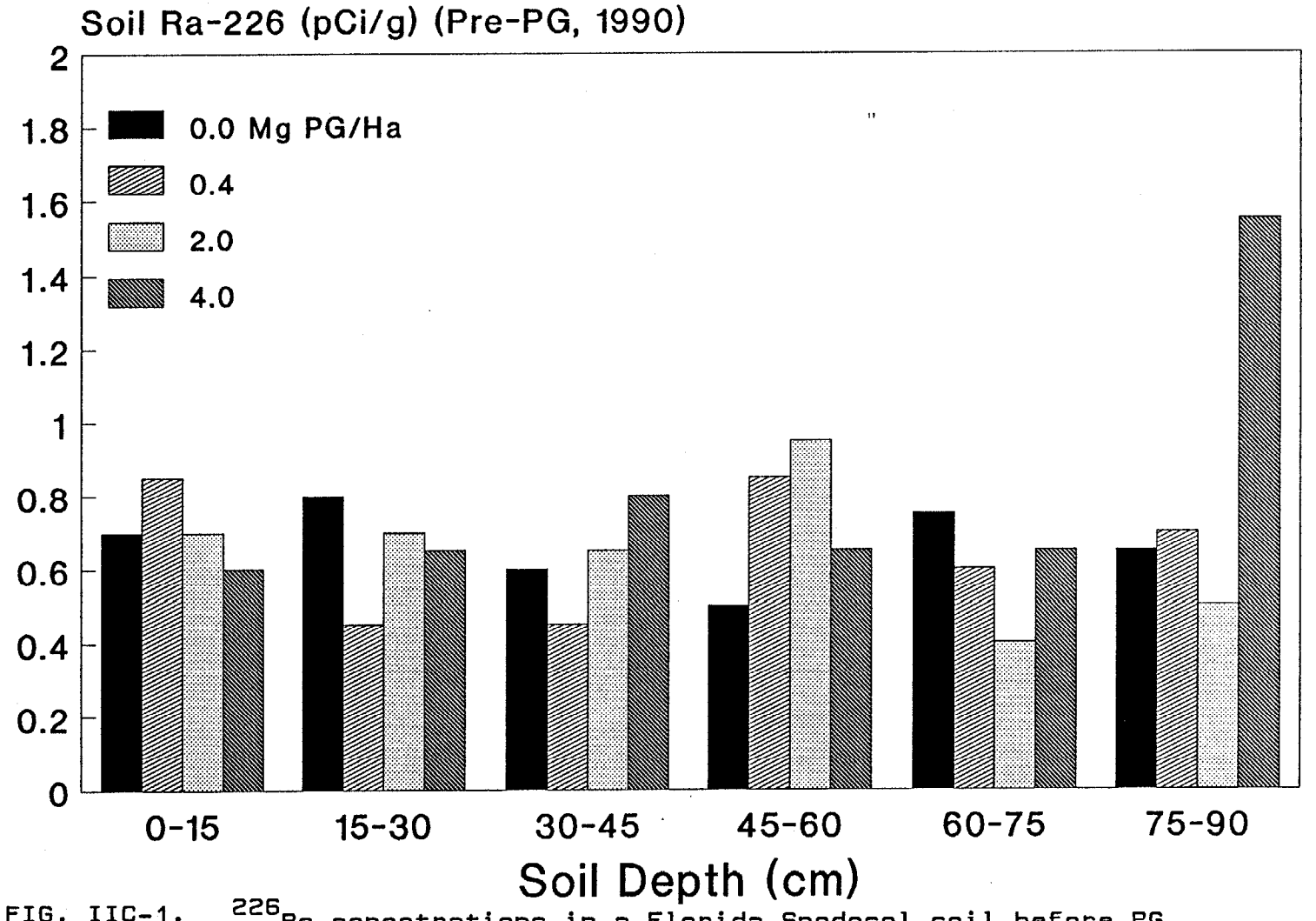

FIG. IIC-1. ^{226}Ra concetrations in a Florida Spodosol soil before PG application, by depth, 1990.

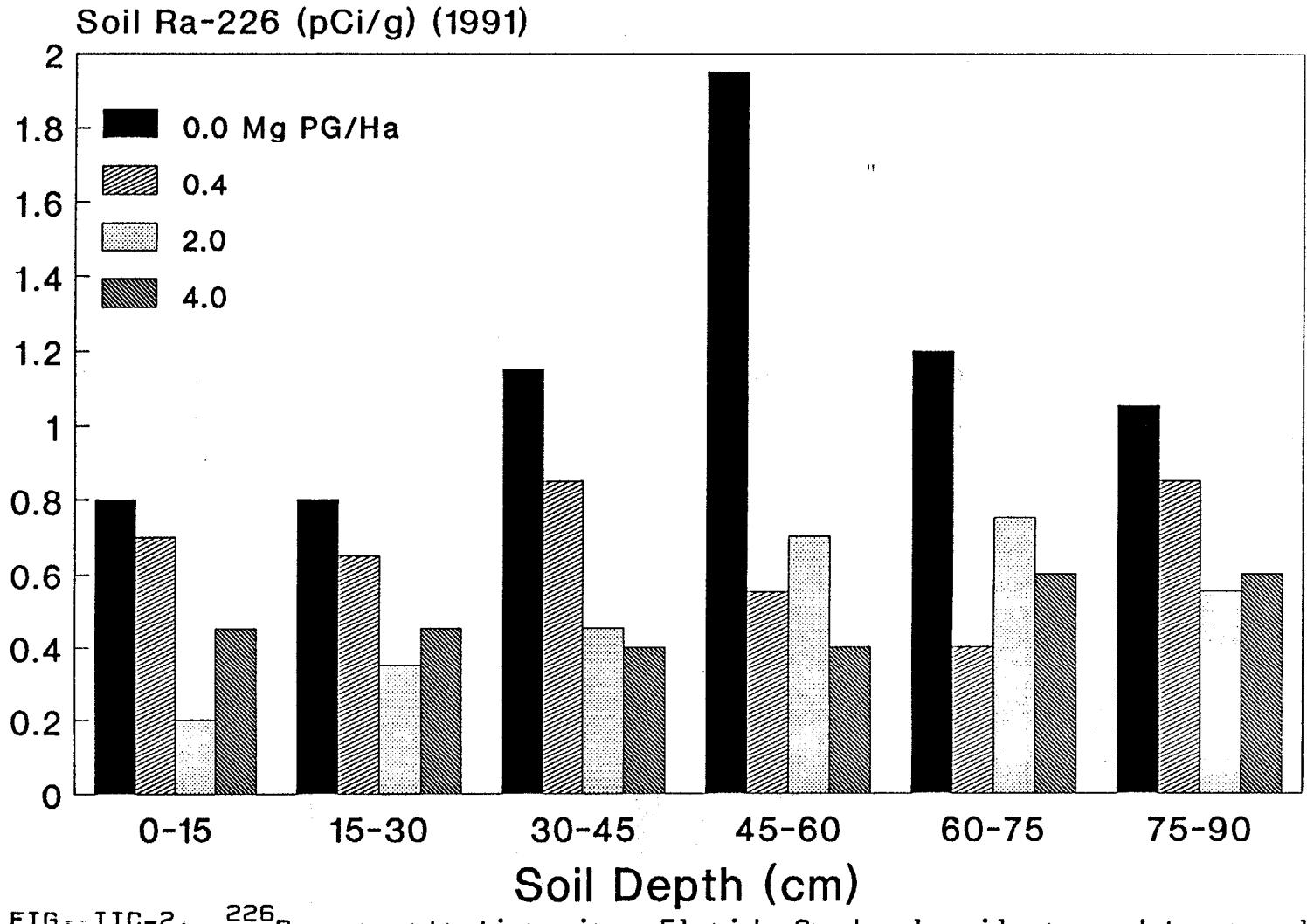

FIG. IIC-2. ^{226}Ra concentrations in a Florida Spodosol soil cropped to annual ryegrass which was amended with PG as source of S and Ca, by depth, 1991.

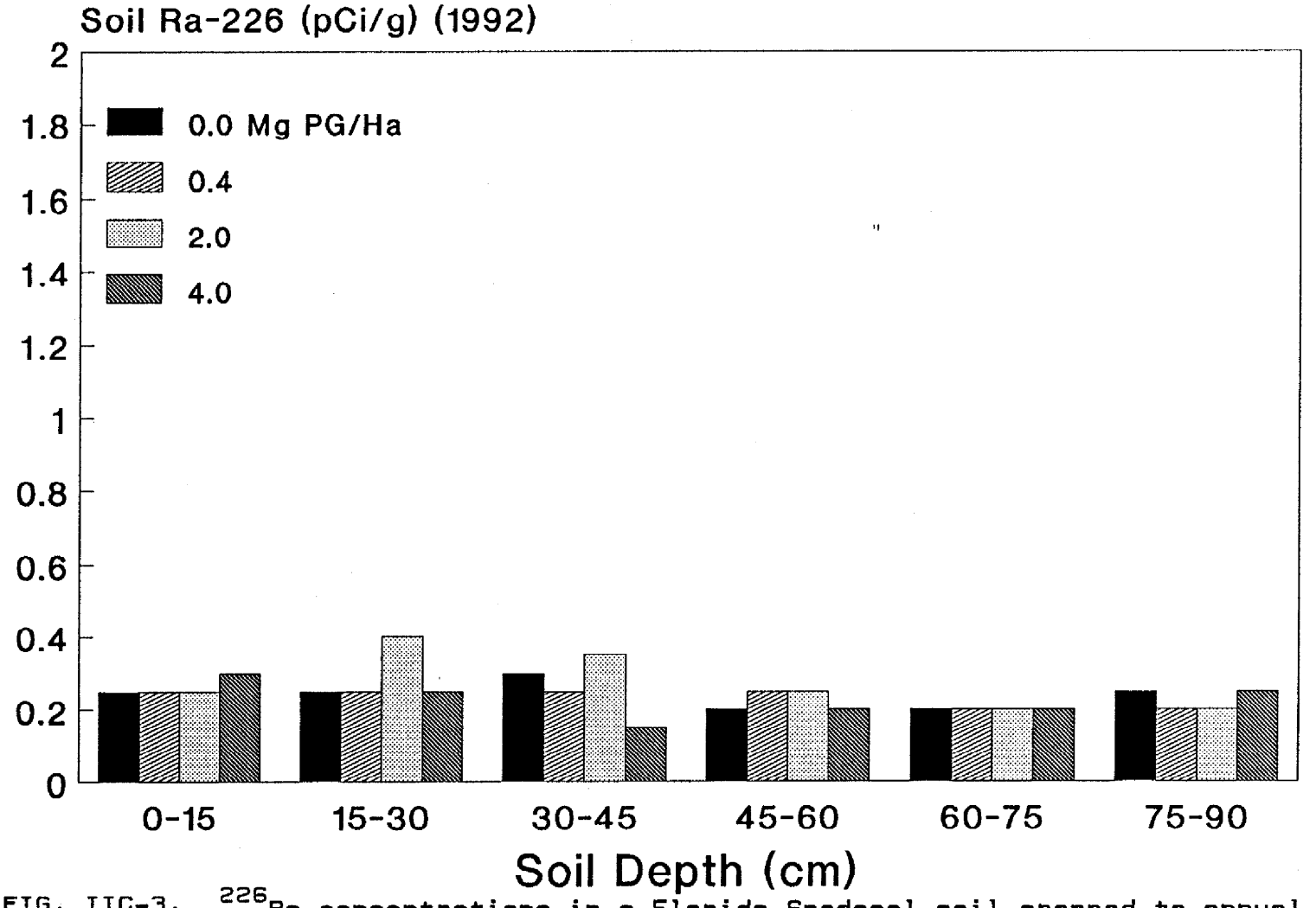

FIG. IIC-3. ^{226}Ra concentrations in a Florida Spodosol soil cropped to annual ryegrass which was amended with PG as source of S and Ca, by depth, 1992.

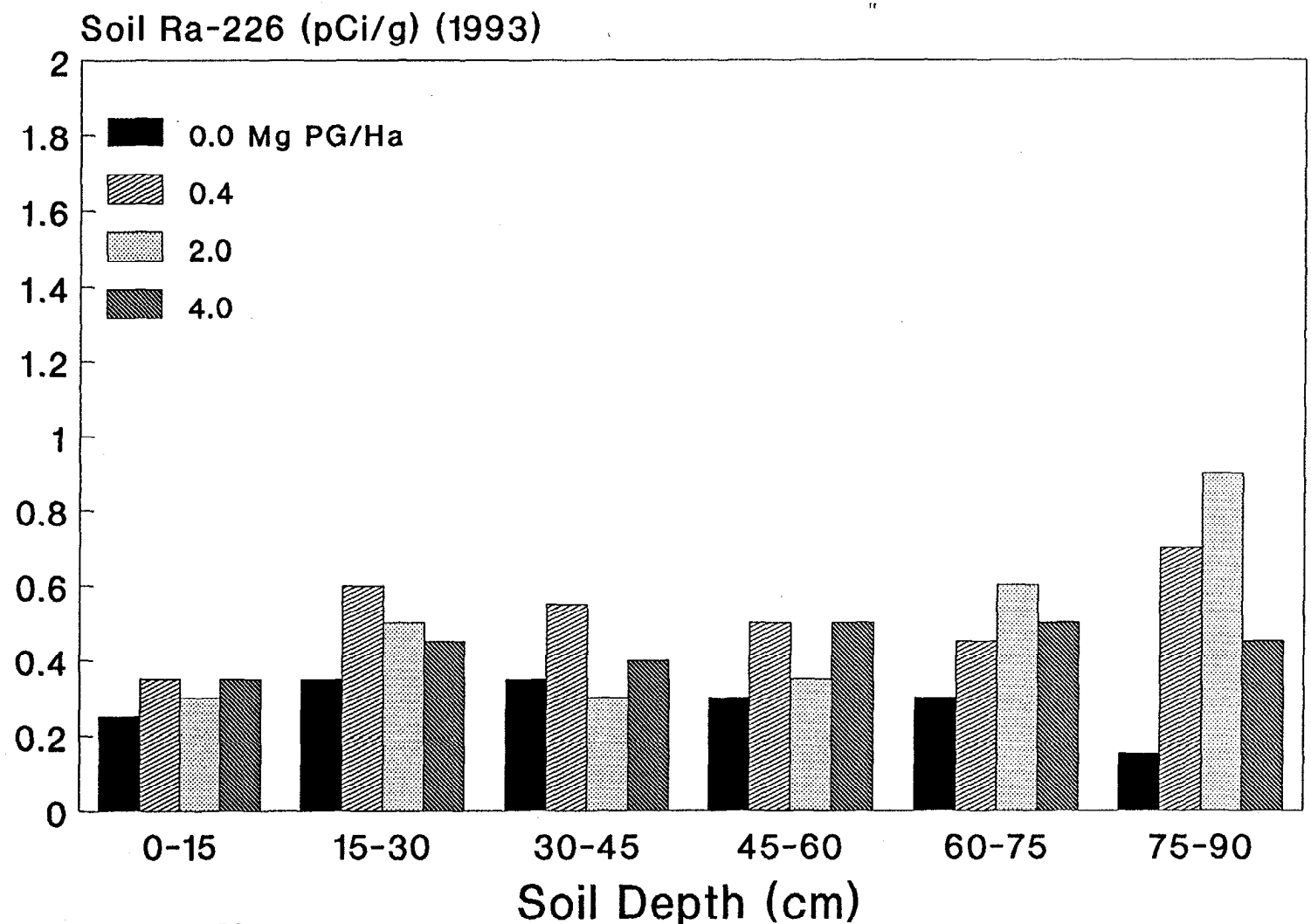

FIG. IIC-4. ^{226}Ra concentrations in a Florida Spodosol soil cropped to annual ryegrass which was amended with PG as source of S and Ca, by depth, 1993.

For the treated plots, concentrations reported for all depths of the 1991 sampling (Figure IIC-2) were lower than those reported for control plots and there was no meaningful trend with treatment level - there is no obvious explanation for this observation. For 1992 (Figure IIC-3), reported treatment and control plot values were consistently low and there was no evidence of a treatment effect. The 1993 data (Figure IIC-4) suggest a treatment effect, particularly at the deeper depths, but none of the differences were statistically significant.

The inability to detect a treatment effect is not surprising. The maximum treatment level of 4.0 Mg PG ha^{-1} when distributed over a 15-cm soil column gives a calculated soil ^{226}Ra radioactivity increase of only 0.03 pCi g^{-1}. Any increase in ^{226}Ra due to PG treatment at the levels used could not be detected with these sampling and analytical procedures.

For comparison, the study of Mullins and Mitchell (1990) reported soil ^{226}Ra concentrations in phosphogypsum experimental plots which were sampled down to 102 cm ranging from 0.08 to 0.35 pCi g^{-1} and found no meaningful treatment differences.

Lead-210. Results are compared by treatment assignment and depth for 1990 (pre-treatment), 1991, 1992, and 1993 in Figures IIC-5, IIC-6, IIC-7, and IIC-8, respectively.

The pre-treatment ^{210}Pb data for all plots (Table Cl and Figure IIC-5) exhibit considerable variability with no obvious pattern by plot group or depth. The pre- and post-treatment control plot data (Figures IIC-5 through IIC-8) also show considerable variability and no consistent pattern from year to year. Pre-treatment values ranged from non-detectible to 1.90 pCi g^{-1}, post-treatment control plot values ranged from 0.20 to 1.90. In the pooled data for untreated plots (all pre-treatment plots plus post-treatment controls), averages for individual 15-cm layers ranged from 0.50 to 0.84 pCi g^{-1}; the average for the top layer was 0.70 pCi g^{-1}.

Results for the treated plots were equivocal. Inspection of the data (Figures IIC-6 through IIC-8) suggest a limited number of cases where the concentrations for treated plots were higher than for control plots; however, typically there was no meaningful correlation of concentration with treatment level and the differences were not statistically significant. Statistically-significant differences and a trend generally consistent with treatment level were seen only for the 60-75 cm

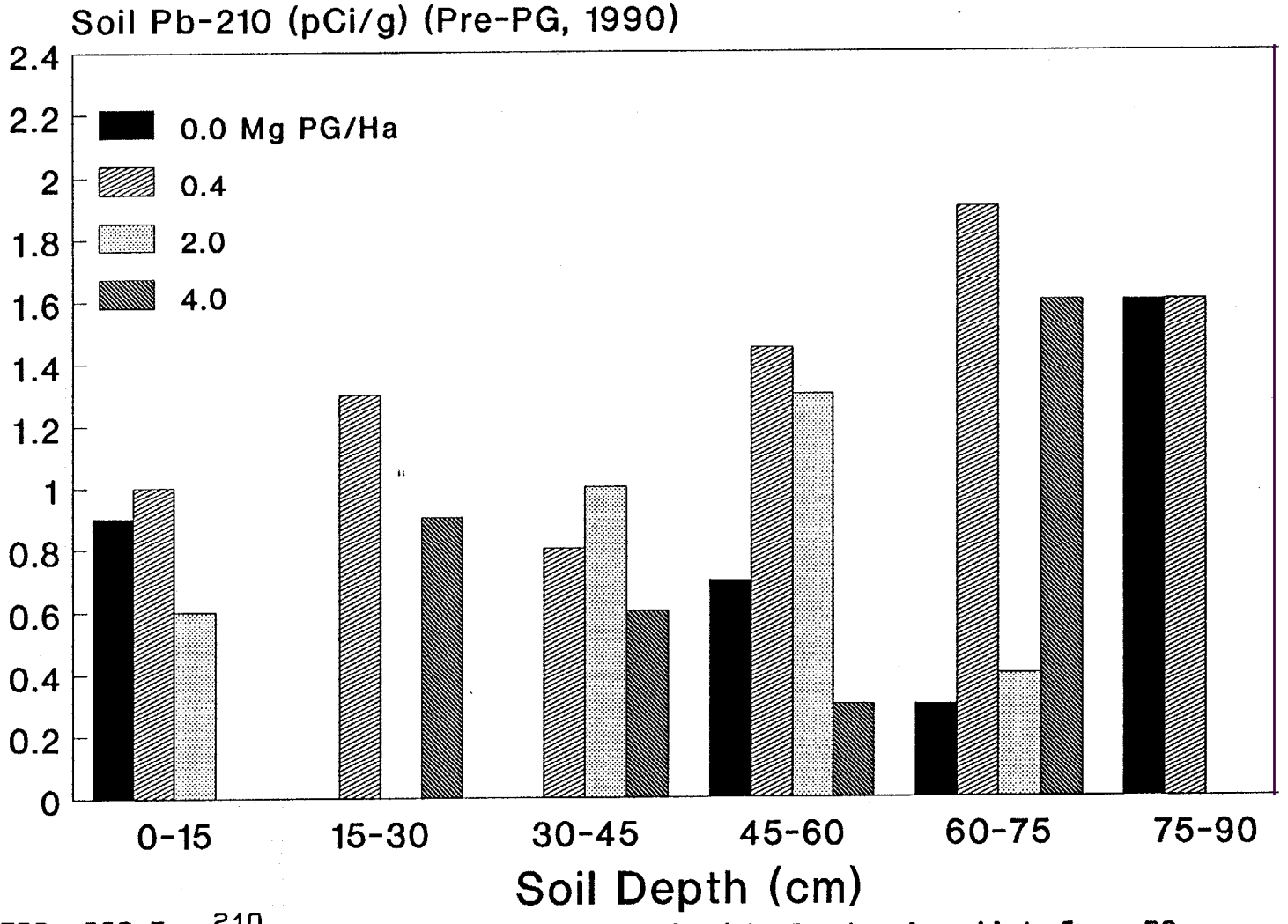

FIG. IIC-5. ^{210}Pb concentrations in a Florida Spodosol soil before PG application, by depth, 1990.

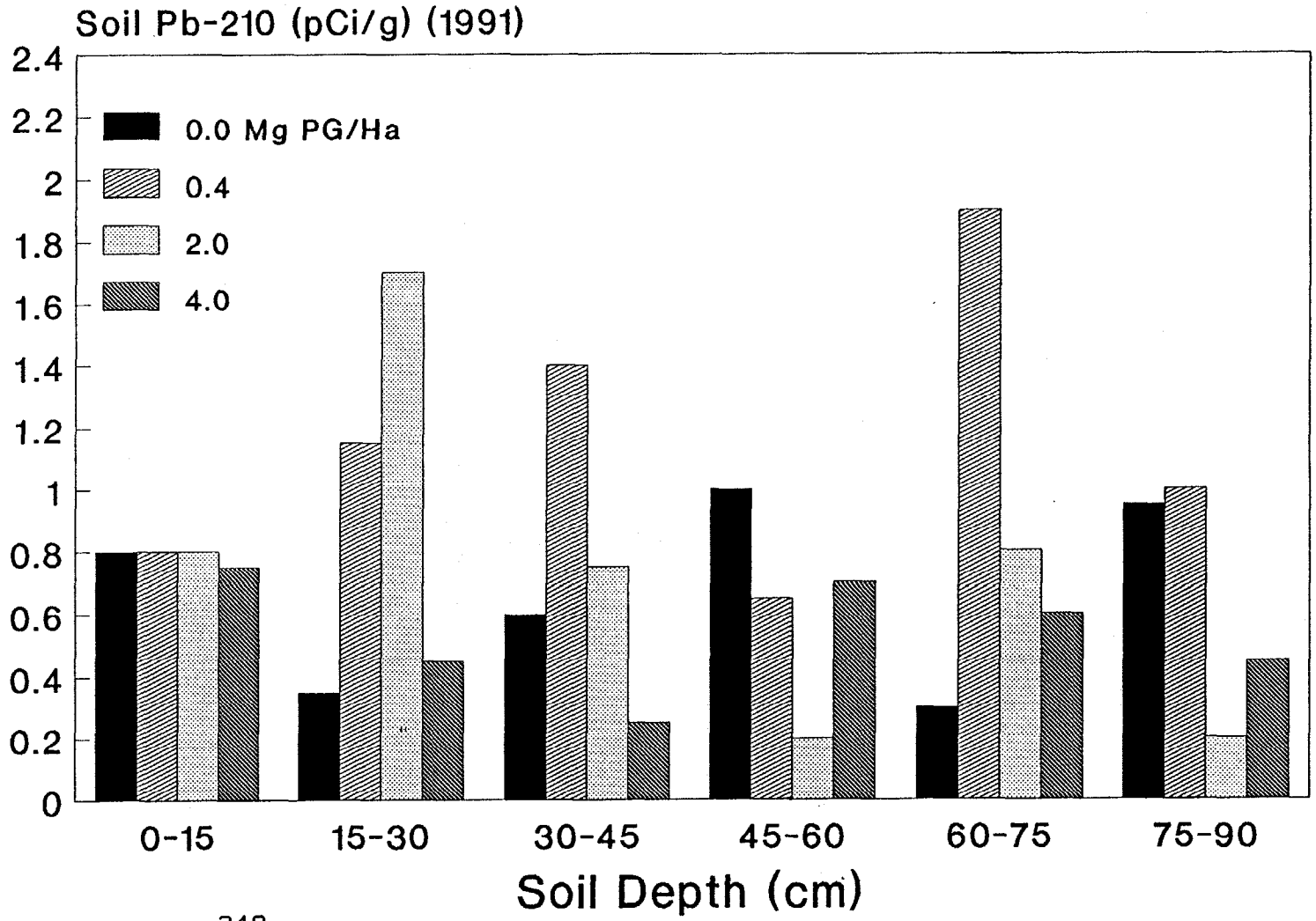

FIG. IIC-6. ^{210}Pb concentrations in a Florida Spodosol soil cropped to annual ryegrass which was amended with PG as source of S and Ca, by depth, 1991.

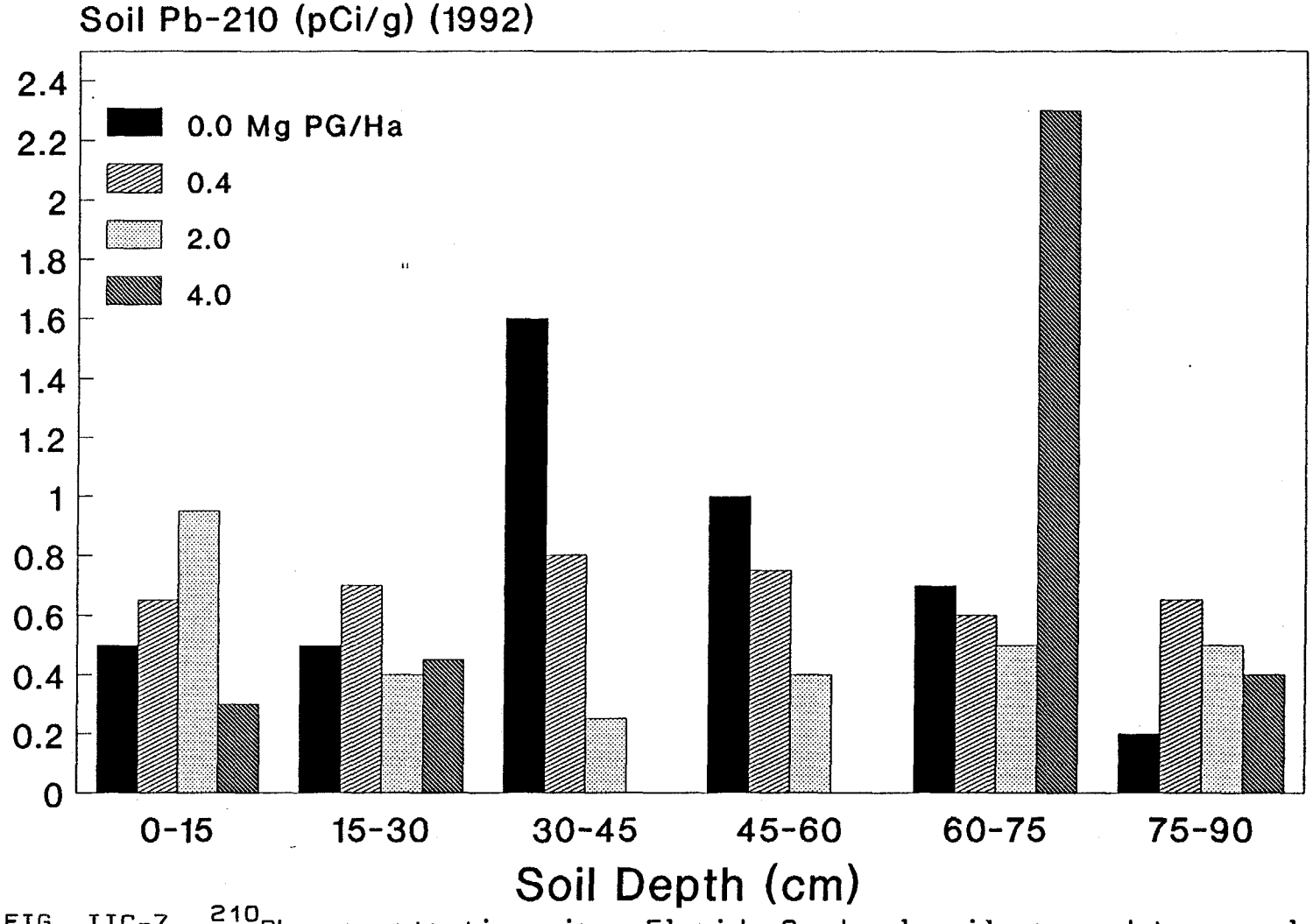

FIG. IIC-7. ^{210}Pb concentrations in a Florida Spodosol soil cropped to annual ryegrass which was amended with PG as source of S and Ca, by depth, 1992.

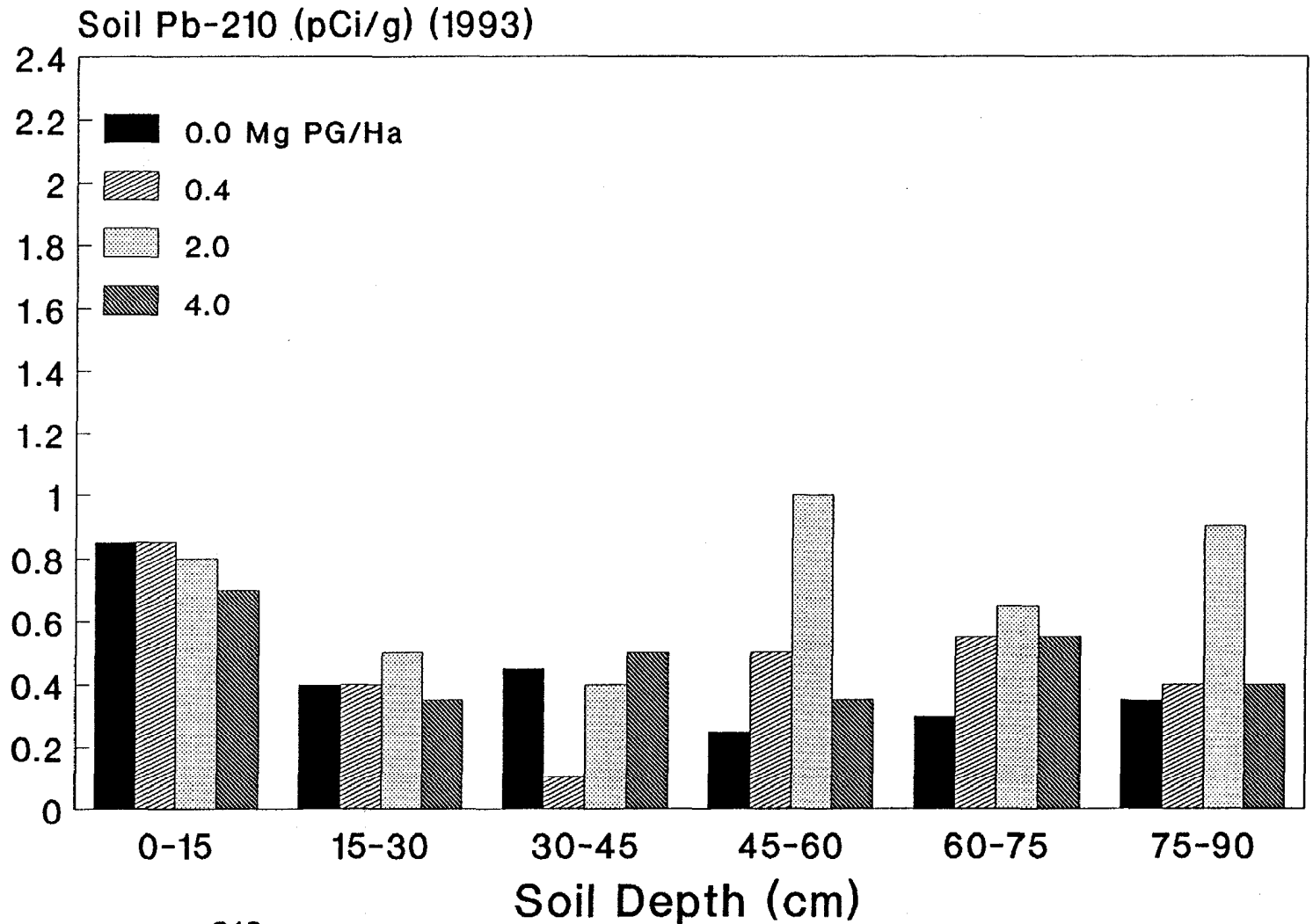

FIG. IIC-8. ^{210}Pb concentrations in a Florida Spodosol soil cropped to annual ryegrass which was amended with PG as source of S and Ca, by depth, 1993.

depth for 1992 and 1993. In two other cases, statistically-significant differences corresponded to an inverse relationship between treatment level and reported concentration in the soil! Overall, no effect of PG treatment on soil ^{210}Pb was detected.

Again, the lack of evidence of treatment effects is not surprising. For the maximum treatment level of 4.0 Mg PG ha^{-1}, the calculated soil ^{210}Pb increase over a 15-cm depth is only 0.05 pCi g^{-1}. This increase could not be detected in the presence of the variations associated with the natural background and with the measurements.

Polonium-210. Results are compared by treatment assignment and depth for 1990 (pre-treatment), 1991, 1992, and 1993 in Figures IIC-9, IIC-10, IIC-11, and IIC-12, respectively.

The 1990 pre-treatment result reported for the control plots upper layer (2.25 pCi g^{-1}) appears to be anomalous. It is 4 to 7 times the values reported in subsequent years and, unlike subsequent years, 3 to 9 times the values reported for other depths. A more consistent value for this depth layer, obtained by extrapolating across depths and years, would be 0.74 pCi g^{-1}.

Inspection of the ^{210}Po data for untreated plots (all pre-treatment plots[2] plus post-treatment controls) suggested a comparable degree of variability between years and between depths with no consistent pattern by depth. Pre-treatment values[2] ranged from 0.20 to 1.40 pCi g^{-1}, post-treatment control plot samples ranged from 0.10 to 0.55. In the pooled data for untreated plots (all pre-treatment plots[2] plus post-treatment controls), averages for individual 15-cm layers ranged from 0.37 to 0.69 pCi g^{-1}; the average for the top layer was 0.57. By comparison, Mullins and Mitchell (1990) reported a soil ^{210}Po range of 0.17 to 0.48 pCi g^{-1} from the top soil down to 46 cm.

There was very limited, if any, evidence for treatment effects. The data for 1991 suggest a treatment effect at 60-75 cm: however, the differences were not statistically significant and a very similar trend was seen at this depth before treatment (1990). The data suggest a treatment effect in the 0-15 cm layer. The 3-year average data (Table C2) indicate a trend (P{Quadratic} = 0.07) and the differences were statistically significant in 1992 (P = 0.03, 2.0>all). However, the 3-year average and 1992 values for the 4.0 Mg ha^{-1} treatment were less than for the 2.0 Mg ha^{-1} treatment. There was no indication of an effect in 1991, and the apparent differences of 1993 (Figure IIC-12) were not statistically significant. For the maximum treatment level (4.0 Mg ha^{-1}), the calculated soil ^{210}Po increase over a 15-cm depth is only 0.04 pCi g^{-1}. This increase could not be detected in the presence of the variations associated with the natural background and with the measurements.

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[2]Using the extrapolated value of the 0-15 cm layer, 1990.

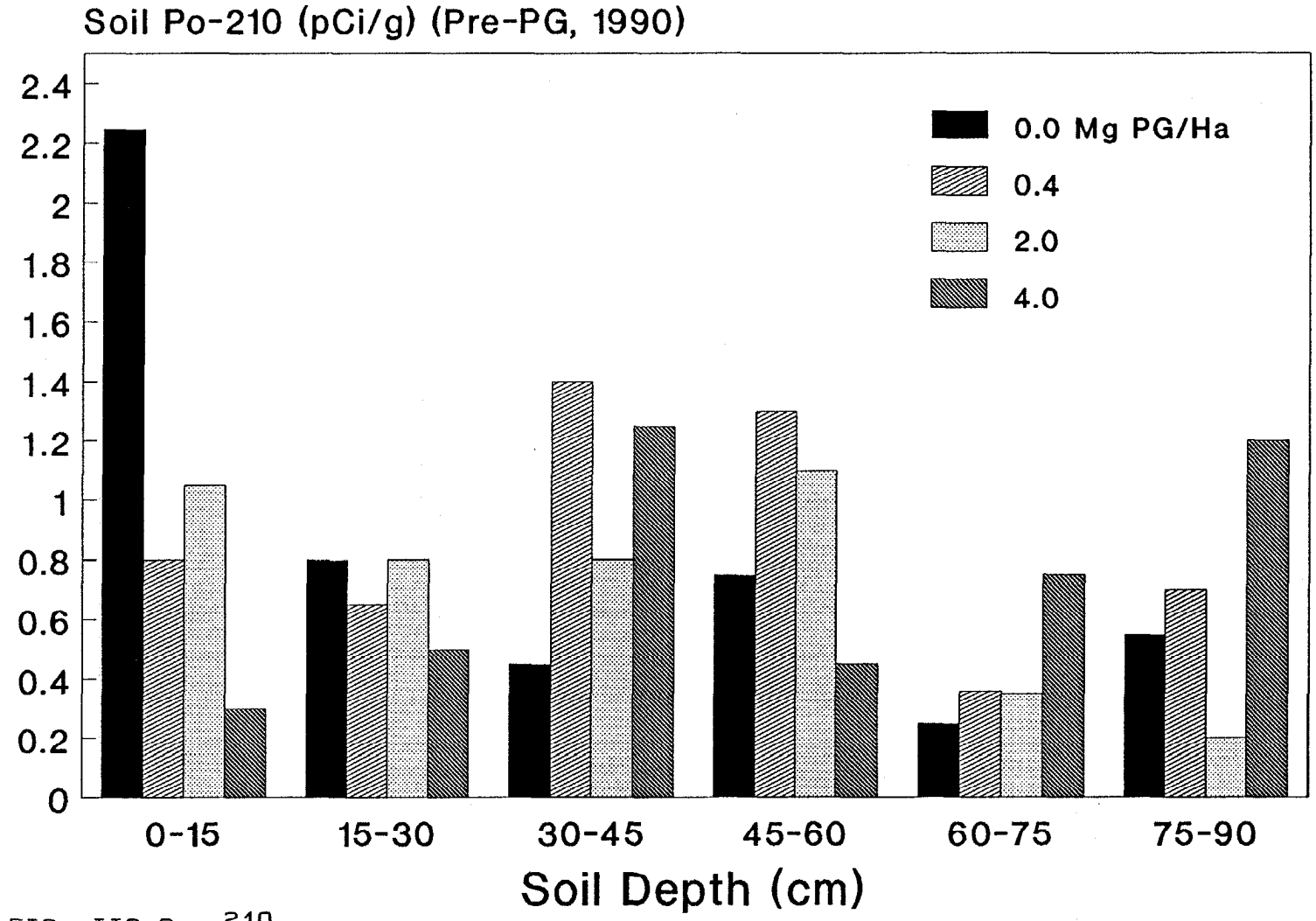

FIG. IIC-9. ^{210}Po concentrations in a Florida Spodosol soil before PG application, by depth, 1990.

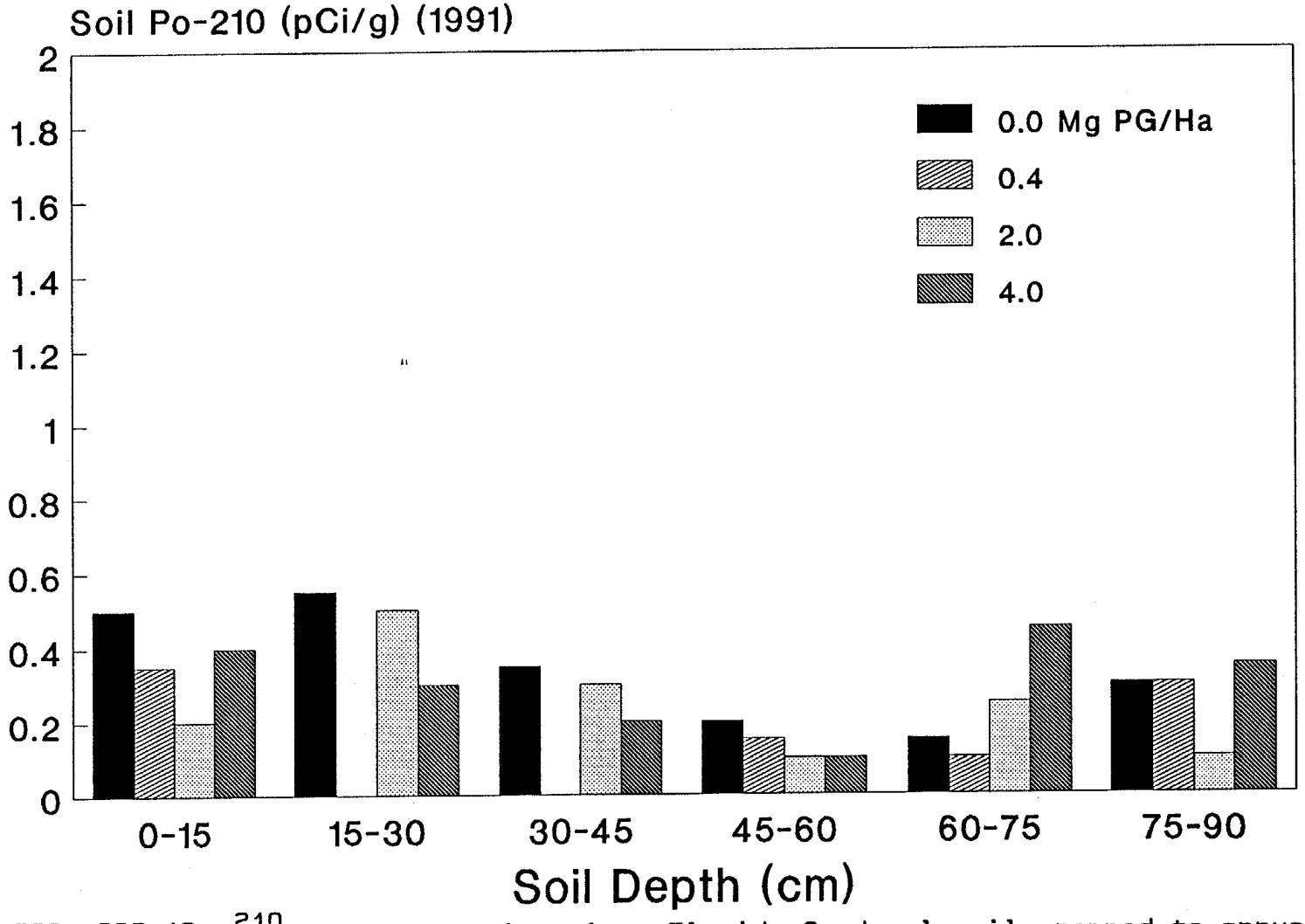

FIG. IIC-10. ^{210}Po concentrations in a Florida Spodosol soil cropped to annual ryegrass which was amended with PG as source of S and Ca, by depth, 1991.

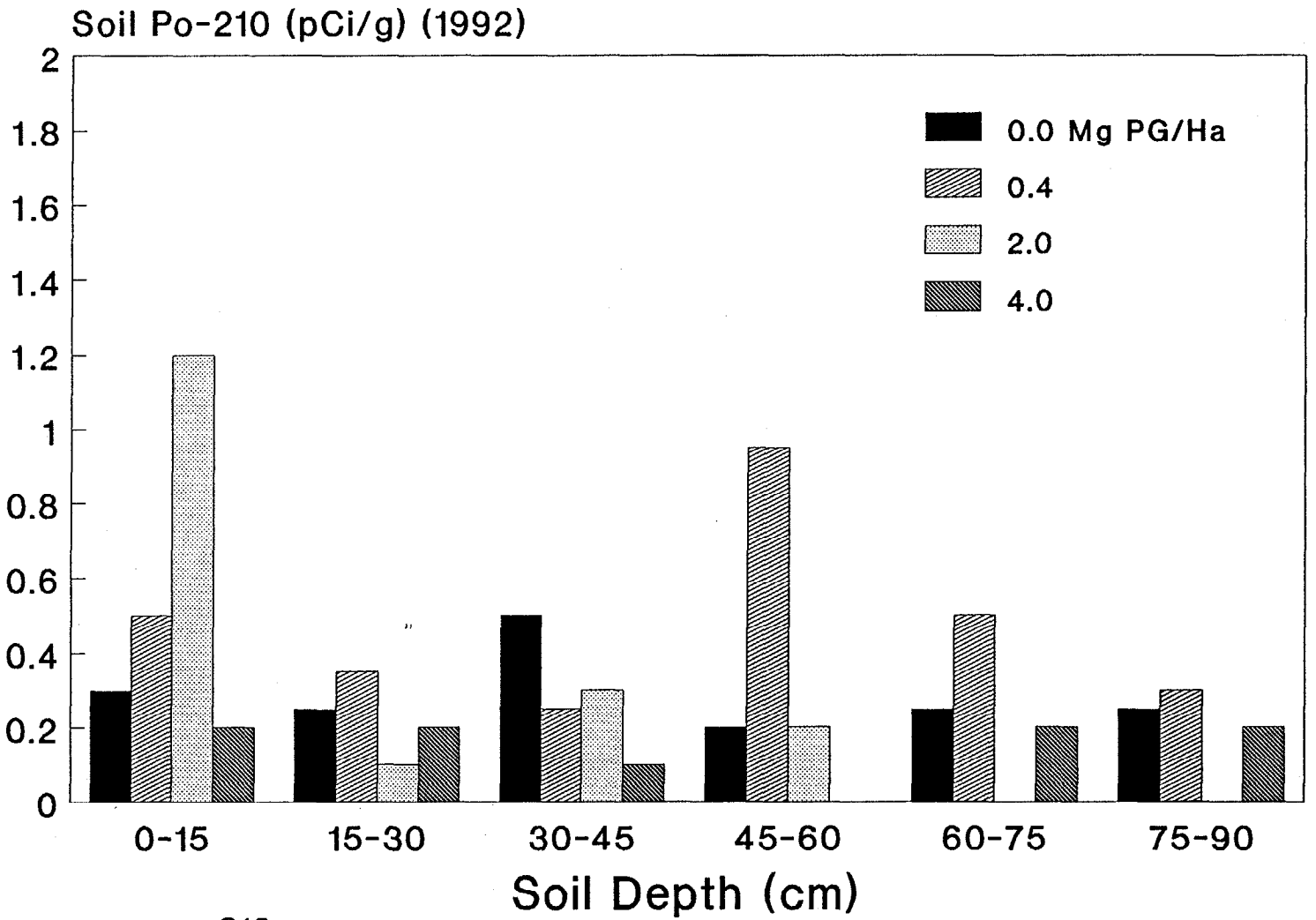

FIG. IIC-11. ^{210}Po concentrations in a Florida Spodosol soil cropped to annual ryegrass which was amended with PG as source of S and Ca, by depth, 1992.

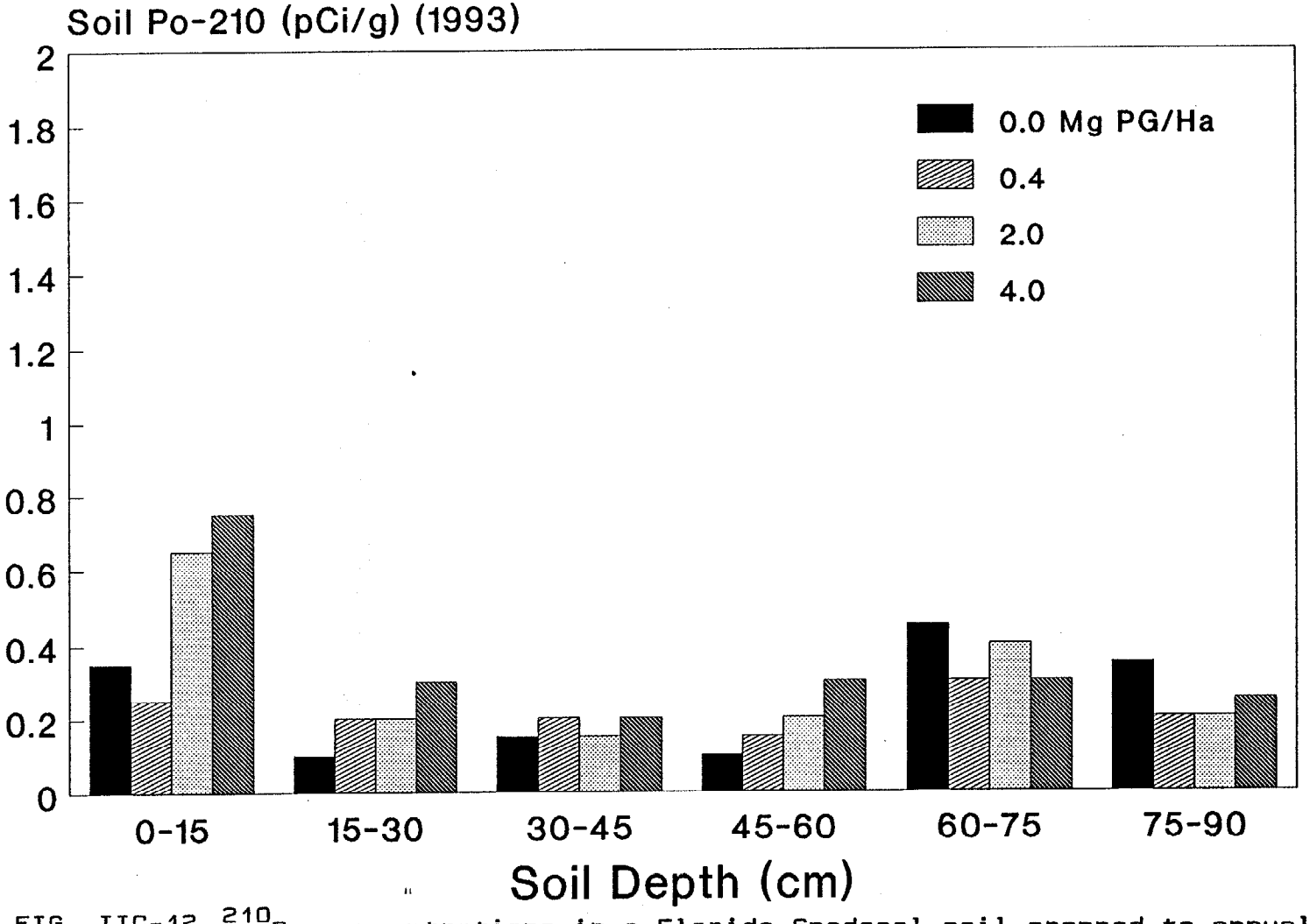

FIG. IIC-12. ^{210}Po concentrations in a Florida Spodosol soil cropped to annual ryegrass which was amended with PG as source of S and Ca, by depth, 1993.

C.3. Gamma Radiation

Gamma radiation 1 m above the experimental plots was measured during 13 time intervals of 30 to 75 days each over the 3-year period. Only partial data was collected during the first sampling period; the subsequent 12 sampling gave complete sets of data. Annual and 3-year average gamma radiation exposure levels are presented in Table C3. Results by individual sampling period are graphically presented for the 12 complete measurements in Figure IIC-13 and given for all measurements in the Appendix (Table IIC-4).

Table C3. Gamma radiation over a Florida Spodosol soil cropped to annual ryegrass which was amended with phosphogypsum (PG) as a source of S and Ca, averaged by year and over a 3-year period, 1990-91 to 1992-93.

Treatment	Crop year			
	1990-91	1991-92	1992-93	mean
Mg PG ha^{-1}		---------- uR hr^{-1} ----------		
0.0(C_{ex1})	4.69	5.55	5.83	5.39
0.0(C_{ex2})	-[a]	6.34	6.50	6.39
0.0(C)	5.02	5.71	5.73	5.45
0.4	4.77	5.45	6.23	5.38
2.0	5.10	5.50	6.38	5.54
4.0	4.86	5.23	5.35	5.13
Statistics:[1]				
P(w/$C_{ex1,2}$)	0.3349	0.1672	0.6645	0.2452
P(w/o $C_{ex1,2}$)	0.5553	0.5866	0.4803	0.4970

[a]No external control at this time.

Background gamma radiation levels over the study period averaged 5.45 /µR hr^{-1} (range of 4.45 to 6.15) for internal control C, 5.4 /µR hr^{-1} (range of 4.10 to 6.45) for external control C_{ex1}, and 6.4 (5.75 to 7.05) for external control C_{ex2}. This suggests that there is some local spatial variation in baseline gamma radiation levels. A higher background existed at C_{ex2} which was located outside the vicinity of the experimental site and right beside an asphalt-paved road, than in the immediate vicinity of the test plots. By comparison, state averages in the U.S. vary from 3.3 in Texas to 14 uR hr^{-1} in Colorado, Nevada, and Wyoming with measurements ranging from <1 to 34 uR hr^{-1} (Myrick, et al., 1981). Previous studies have reported Florida values on the order of 5 to 10 uR hr^{-1} (Roessler, 1987).

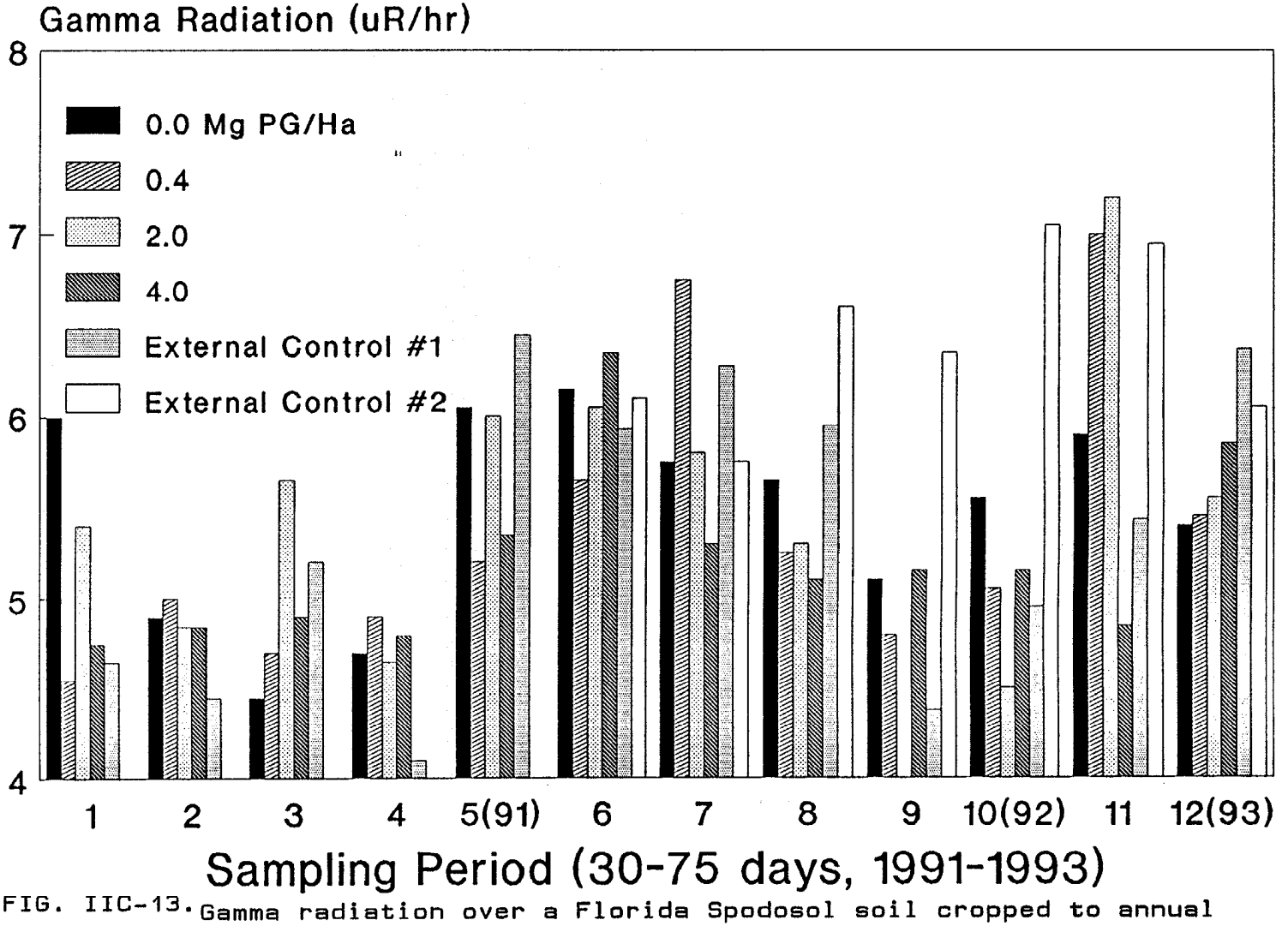

FIG. IIC-13. Gamma radiation over a Florida Spodosol soil cropped to annual ryegrass which was amended with PG as source of S and Ca, by sampling, 1991-1993.

For the treated plots, gamma radiation levels ranged from 4.50 to 7.20 uR hr^{-1} (Figure IIC-13 and Table IIC-4, Appendix). There were no statistically-significant differences when the data were analyzed without the external controls. When the data were analyzed with the external controls, differences were statistically significant for about one-third of the tests - typically because the C_{ex2} value was higher than the treated plot values. Thus, at the rates and frequency used, any influence of PG on the levels of gamma radiation over a tilled land cropped to annual ryegrass was insignificant in comparison to the temporal and spatial variations in the local background.

C.4. Radionuclides in Plant Tissue

Two regrowth harvests during crop year 1990-91 three in 1991-92, and two in 1992-93 were analyzed for ^{226}Ra, ^{210}Pb, and ^{210}Po.

Radium-226. Annual average and 3-year average concentrations in regrowth forage and annual concentrations and 3-year average concentrations for the annual harvests of the mature forage are presented in Table C4. Results for individual regrowth harvests are presented in Figure IIC-14. Analytical and statistical testing results for individual harvests are presented in Appendix Table IIC-5 for all three radionuclides.

Baseline (control plot) ^{226}Ra concentrations in regrowth forage averaged 0.13 pCi g^{-1}, with annual averages ranging from 0.10 to 0.16 and individual harvests ranging from below the limit of detection to 0.19. The average concentration in control plot mature forage was 0.23 pCi g^{-1} with individual annual harvests ranging from 0.19 to 0.30. By comparison, average tissue concentrations of 0.04 pCi g^{-l} were reported by Mislevy, et al. (1989) for various biomass plants growing on unmined Florida soil and concentrations ranging from 0.04 to 0.13 pCi^{-1} were reported by Mullins and Mitchell (1990) for wheat forage from untreated plots in Alabama.

For the treated plots, concentrations of ^{226}Ra ranged from 0.09 to 0.31 pCi g^{-1} in regrowth forage and from 0.20 to 0.45 pCi g^{-1} for annual harvest of the mature hay. The data suggest a treatment effect that persists into the third year (Figure IIC-14 and Table C4); however, statistically-significant effects were limited (Tables C4 and IIC-5). There was evidence (P=0.06) of treatment effects in the January 1992 harvest and the February 1992 data support a model of ^{226}Ra in tissue increasing linearly with phosphogypsum rates. The data suggest a PG-attributable uptake in regrowth and mature forage on the order of 0.039 pCi g^{-1} per Mg ha^{-1}.

Table C4. ^{226}Ra concentrations in annual ryegrass regrowth and hay forage from a pasture amended with phosphogypsum (PG) as a source of S and Ca and growing on a Florida Spodosol soil, averaged by year and over a 3-year period, 1990-91 to 1992-93.

Treatment	Crop year			
	1990-91[a]	1991-92[a]	1992-93[a]	Mean[a]
A. Regrowth:	---------- pCi ^{226}Ra g^{-1} ----------			
Mg PG ha^{-1}				
0.0(C)	0.10	0.16	0.10	0.13
0.4	0.10	0.16	0.13	0.14
2.0	0.15	0.23	0.20	0.20
4.0	0.15	0.23	0.40	0.25
Statistics:				
P	0.2580	0.3950	0.7180	0.4376
B. Hay:	---------- pCi ^{226}Ra g^{-1} ----------			
Mg PG ha^{-1}				
0.0(C)	0.30	0.19	0.20	0.23
0.4	0.30	0.22	0.25	0.26
2.0	0.35	0.31	0.20	0.29
4.0	0.45	0.24	0.30	0.33
Statistics:				
P	0.6265	0.7139	0.6677	0.4621

[a]Statistics computed based solely on those harvests where each treatment had at least one detected value associated with it.

Lead-210. Annual average and 3-year average concentrations in regrowth forage and annual concentrations and 3-year average concentrations for the mature forage are presented in Table C5. Results for individual regrowth harvests are presented in Figure IIC-15. As indicated for ^{226}Ra, analytical and statistical testing results for individual harvests arepresented in Appendix Table IIC-5.

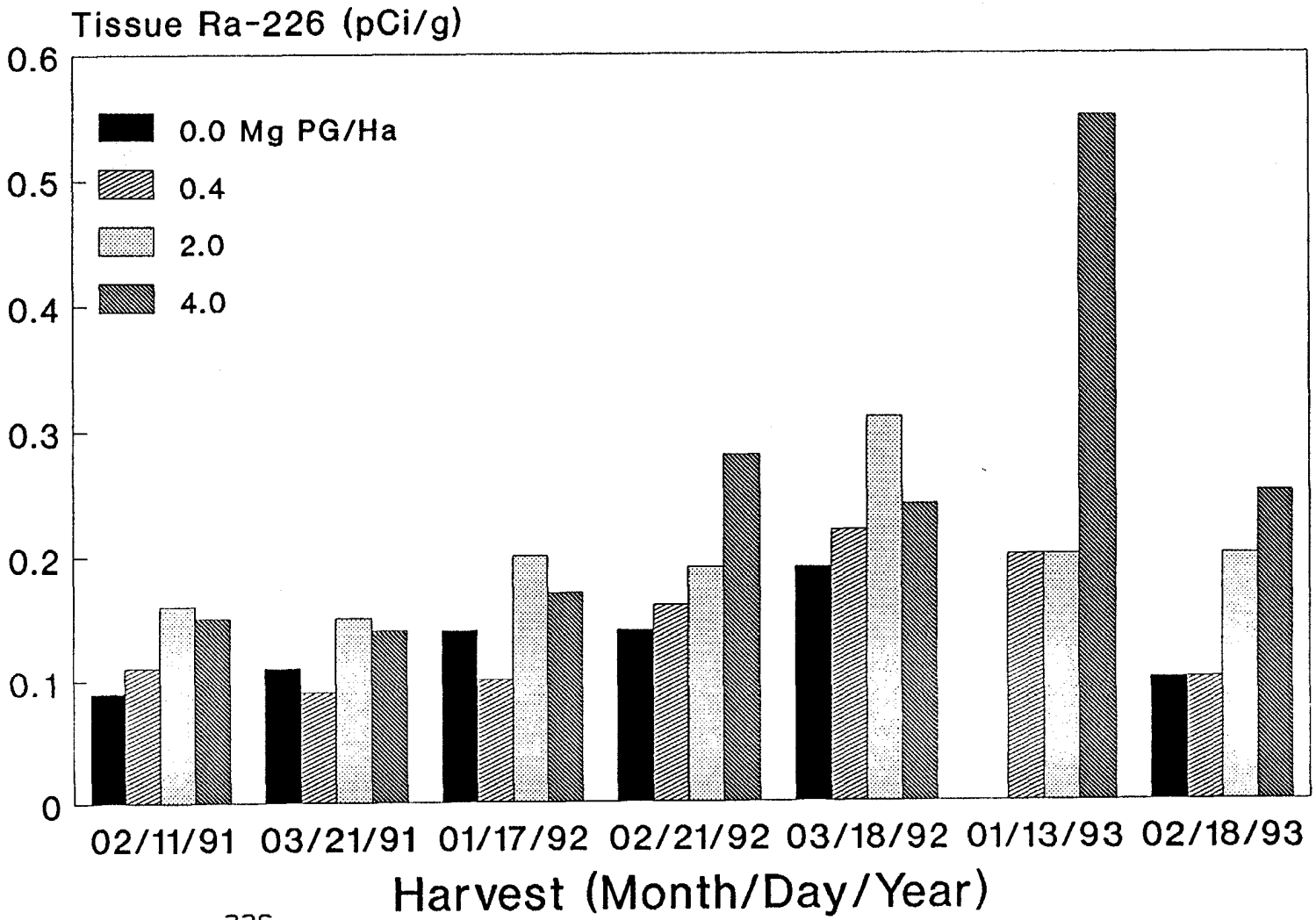

FIG. IIC-14. ^{226}Ra concentrations in regrowth annual ryegrass forage from a pasture amended with PG as source of S and Ca and growing on a Florida Spodosol soil, by harvest, 1990-91 to 1992-93.

Table C5. ^{210}Pb concentrations in annual ryegrass regrowth and hay forage from a pasture amended with phosphogypsum (PG) as a source of S and Ca and growing on a Florida Spodosol soil, averaged by year and over a 3-year period, 1990-91 to 1992-93.

Treatment	Crop year			
	1990-91[a]	1991-92[a]	1992-93[a]	Mean[a]
A. Regrowth:	---------- pCi ^{210}Pb g^{-1} ----------			
Mg PG ha^{-1}				
0.0(C)	0.07	0.43	0.27	0.27
0.4	0.05	0.34	0.27	0.23
2.0	0.23	0.31	0.25	0.26
4.0	0.17	0.26	0.43	0.28
Statistics:				
P	0.6865	0.4384	0.2055	0.9006
B. Hay:	---------- pCi ^{210}Pb g^{-1} ----------			
Mg PG ha^{-1}				
0.0(C)	0.70	0.56	0.50	0.59
0.4	0.90	0.41	0.40	0.57
2.0	0.80	0.43	0.35	0.53
4.0	0.85	0.33	0.25	0.48
Statistics:				
P	0.6608	0.7156	0.2267	0.2901
P(Linear)	ns[1]	ns	0.0714	ns

[1]ns=not significant. [a]Statistics computed based solely on those harvests where each treatment had at least one detected value associated with it.

A striking feature of these data is the wide range of ^{210}Pb concentrations observed in control plot regrowth forage (Figure IIC-15). Untreated plot concentrations averaged 0.27 pCi g^{-1}, with annual averages ranging from 0.07 to 0.43 and individual harvests ranging from 0.01 to 0.56. In control plot mature hay, the concentrations were less variable and generally higher, averaging 0.59 pCi g^{-1} with individual annual harvests ranging from 0.50 to 0.70.

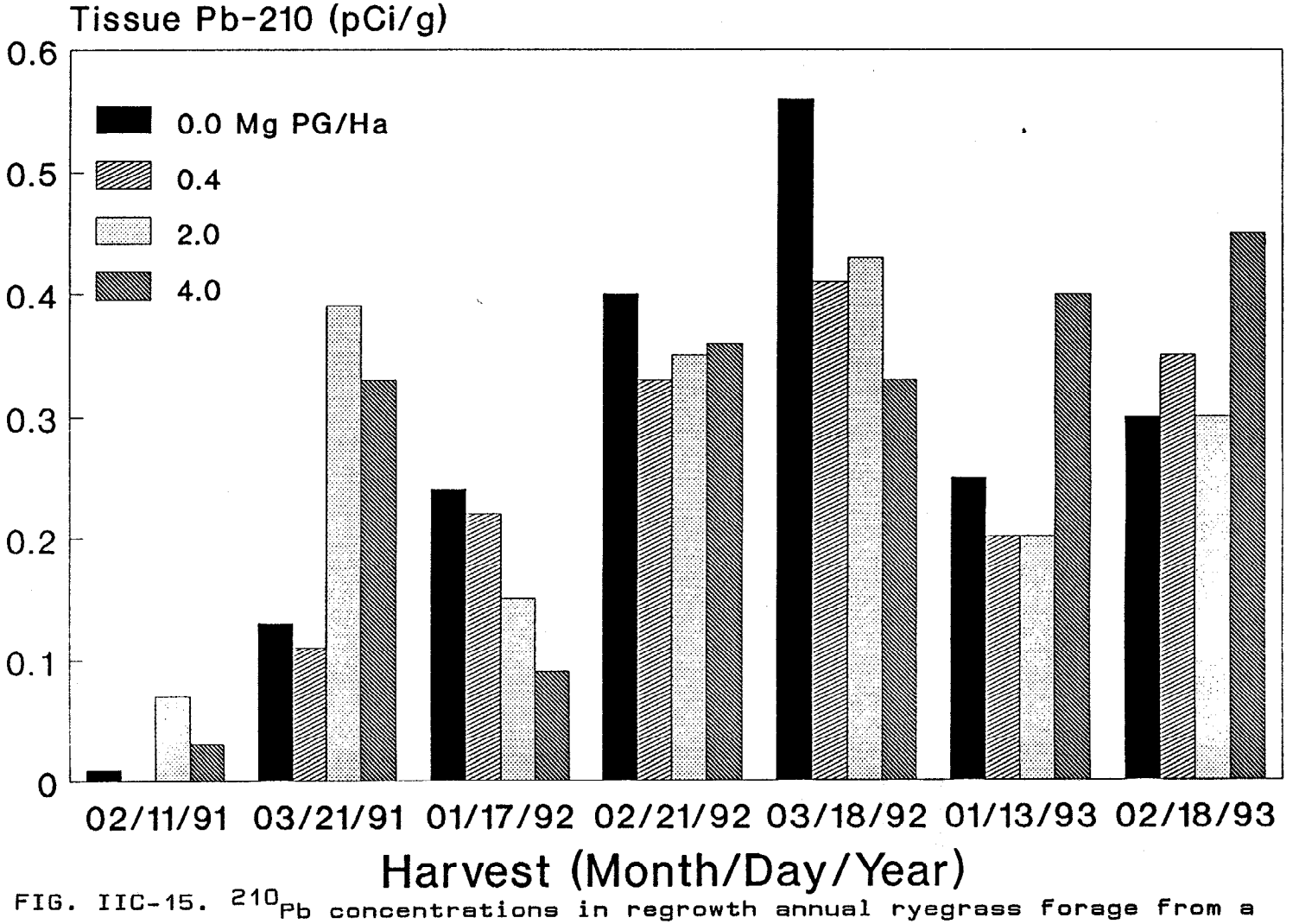

FIG. IIC-15. ^{210}Pb concentrations in regrowth annual ryegrass forage from a pasture amended with PG as source of S and Ca and growing on a Florida Spodosol soil, by harvest, 1990-91 to 1992-93.

The ^{210}Pb values reported for individual regrowth harvests from treated plots ranged from 0.0 to 0.45 pCi g^{-1} and were highly variable with no consistent trend with treatment level. For 1990-91 and 1992-93, annual averages in forages from plots treated with 4.0 Mg PG ha^{-1} appeared to be elevated relative to the control. However, there were no statistically significant differences among the individual harvests, the annual averages, or the 3-year mean. In the case of mature hay, tissue ^{210}Pb ranged from 0.25 to 0.90 pCi g^{-1} for the treated plots (Table C6); there were no statistically-significant treatment differences and the 1992-93 data suggested an inverse relationship between concentration in tissue and treatment level (P{Linear} = 0.070).

Polonium-210. Annual average and 3-year average concentrations in regrowth forage and annual concentrations and 3-year average concentrations for the mature forage are summarized in Table C6. Results for individual regrowth harvests are presented in Figure IIC-16 and Appendix Table IIC-5.

A striking, and unexplained, feature of the ^{210}Po data is progressive yearly increase in concentrations reported for control plot regrowth forage (Table C6 and Figure IIC-16). Untreated plot concentrations averaged 0.20 pCi g^{-1} over 3 years, with annual averages ranging from 0.05 to 0.50 and individual harvests ranging from undetected to 0.50. In control plot mature hay, the values were quite variable and averaged 1.40 pCi g^{-1} with individual annual harvests ranging from nondetectible to 2.35. By comparison, Mullins and Mitchell (1990) reported a range of tissue ^{210}Po from 0.08 to 0.44 pCi g^{-1} in wheat forage harvested from plots not treated with phosphogypsum.

The regrowth and mature forage ^{210}Po data for treated plots showed a high degree of variability from sampling to sampling. The regrowth forage values reported for individual harvests ranged from non-detectible to 0.50 pCi g^{-1} (Figure IIC-16) and calculated annual averages ranged from 0.04 to 2.35 (Table C6). The data suggest a possible treatment effect in the 1991 and the January 1993 sampling. However, annual averages and the 3-year average showed no evidence of a treatment effect and none of the differences were statistically significant. In the case of mature hay, there was no evidence of a treatment effect.

C.5. Radionuclides in Surficial Groundwater

Surficial groundwater at 120 cm depth was sampled five times over the 3-year period for radionuclide analysis. Results are summarized in Table C7 and data by sampling are presented in the Appendix, Table IIC-6.

Table C6. ^{210}Po concentrations in annual ryegrass regrowth and hay forage from a pasture amended with phosphogypsum (PG) as a source of S and Ca and growing on a Florida Spodosol soil, averaged by year and over a 3-year period, 1990-91 to 1992-93.

Treatment	Crop year			
	1990-91[a]	1991-92[a]	1992-93[a]	Mean[a]
A. Regrowth:	---------- pCi ^{210}Po g^{-1} ----------			
Mg PG ha^{-1}				
0.0(C)	0.05	0.19	0.50	0.20
0.4	0.14	0.11	0.30	0.19
2.0	0.06	0.09	0.23	0.13
4.0	0.09	0.08	0.25	0.13
Statistics:				
P	0.5536	0.6505	0.8501	0.8903
B. Hay:	---------- pCi ^{210}Po g^{-1} ----------			
Mg PG ha^{-1}				
0.0(C)	2.35	–[b]	0.45	1.40
0.4	2.35	0.09	0.45	1.14
2.0	1.40	0.09	0.70	0.86
4.0	1.15	0.04	0.45	0.65
Statistics:				
P	0.2978	–[c]	0.5322	0.3042

[a]Statistics computed based solely on those harvests where each treatment had at least one detected value associated with it. [b]Not detected in sample. [c]No source of error for appropriate test due to missing data.

Radium-226. The trends in the values are graphically shown in Figure IIC-17. Concentrations at the external control plots C_{ex} averaged 0.52 pCi L^{-1}, ranged from 0.20 to 0.90, and appeared to have a decreasing trend over the study period. Concentrations at the internal control plots C averaged 0.85 pCi L^{-1} and ranged from 0.15 to 1.55; concentrations were originally lower than for C_{ex} and tended to increase with time.

The data suggest a treatment effect for ^{226}Ra (Figure IIC-17 and Table C7). However, statistically-significant differences were observed only for the first collection (P=0.09, P{Linear}=0.03) and the 1991 crop-year average tested without C_{ex} (P[w/o C_{ex})=0.07, P{Linear}=0.01); and the treated plots results did not correlate well with treatment level.

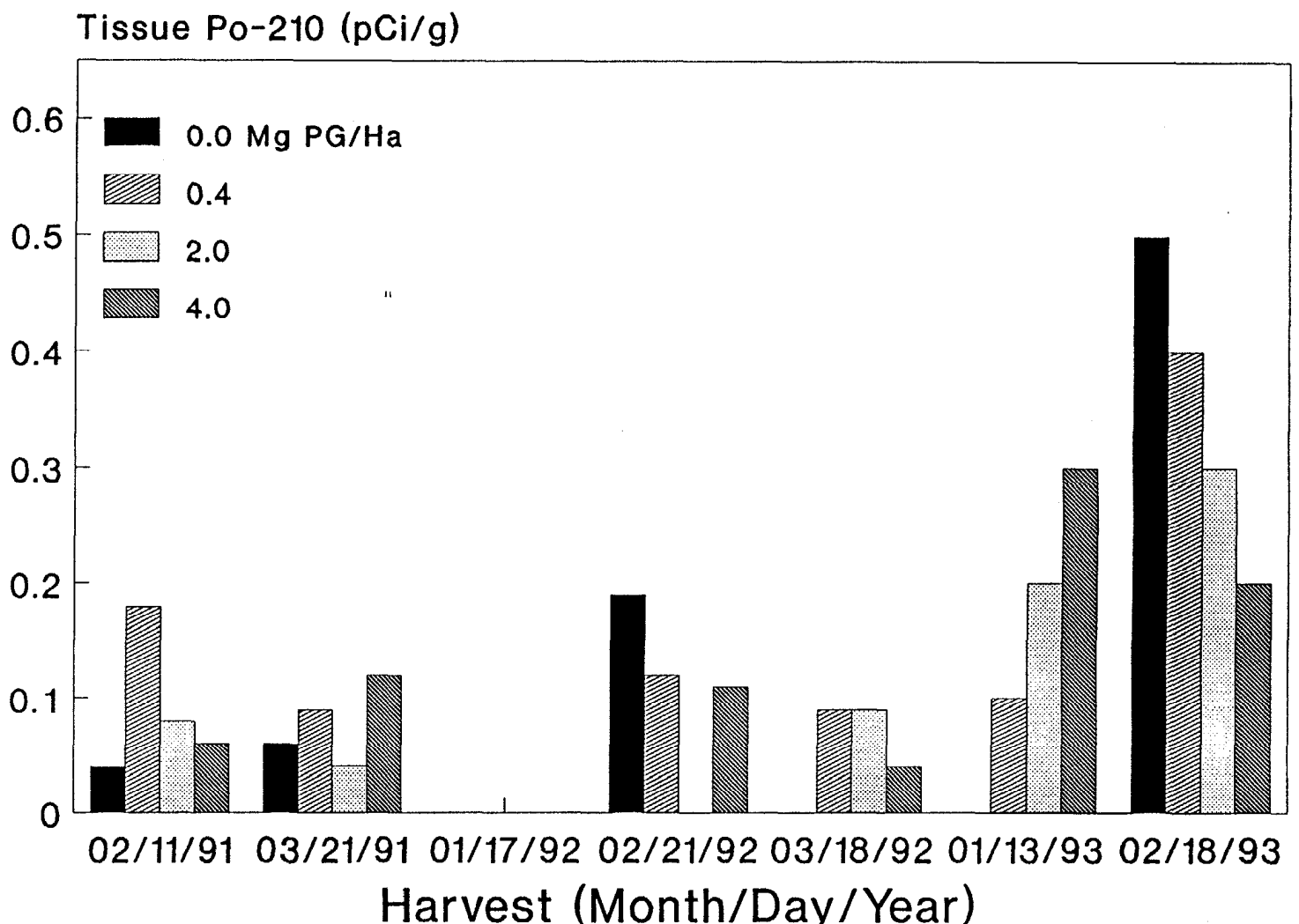

FIG. IIC-16. ^{210}Po concentrations in regrowth annual ryegrass forage from a pasture amended with PG as source of S and Ca and growing on a Florida Spodosol soil, by harvest, 1990-91 to 1992-93.

Table C7. Radionuclide concentrations in groundwater sampled at 120 cm depth from a Florida Spodosol soil cropped to annual ryegrass which was amended with phosphogypsum (PG) as a source of S and Ca, averaged by year and over a 3-year period, 1990-91 to 1992-93.

Depth (cm)/ Treatment	Crop year 1991	1992	1993	Mean
Mg PG ha^{-1}	---------- pCi ^{226}Ra L^{-1} ----------			
C_{ex}	0.90	0.47	0.20	0.52
0.0(C)	0.33	1.23	1.15	0.85
0.4	0.85	2.52	2.35	1.82
2.0	0.53	2.02	1.65	1.35
4.0	1.20	1.09	1.15	1.15
Statistics:				
P(w/C_{ex})	0.1578	0.2446	0.4432	0.2110
P(w/o C_{ex})	0.0723	0.1436	0.7075	0.3661
P(Linear)	0.0427	ns[1]	ns	ns
Mg PG ha^{-1}	---------- pCi ^{210}Pb L^{-1} ----------			
0.0(C_{ex})	0.45	0.84	0.55	0.67
0.0(C)	0.37	0.37	0.95	0.49
0.4	0.55	2.50	2.15	1.44
2.0	0.63	0.90	2.45	1.12
4.0	0.83	0.51	2.10	1.00
Statistics:				
P(w/C_{ex})	0.4082	0.0452	0.3484	0.5702
P(w/o C_{ex})	0.3831	0.0846	0.5473	0.6013
Mg PG ha^{-1}	---------- pCi ^{210}Po L^{-1} ----------			
0.0(C_{ex})	0.80	0.18	0.65	0.49
0.0(C)	0.63	0.34	0.50	0.49
0.4	0.77	0.92	0.40	0.74
2.0	1.00	0.89	0.45	0.85
4.0	0.57	0.60	0.40	0.53
Statistics:				
P(w/C_{ex})	0.8312	0.4097	0.7442	0.5351
P(w/o C_{ex})	0.7137	0.5966	0.8801	0.4225

[1]ns=not significant.

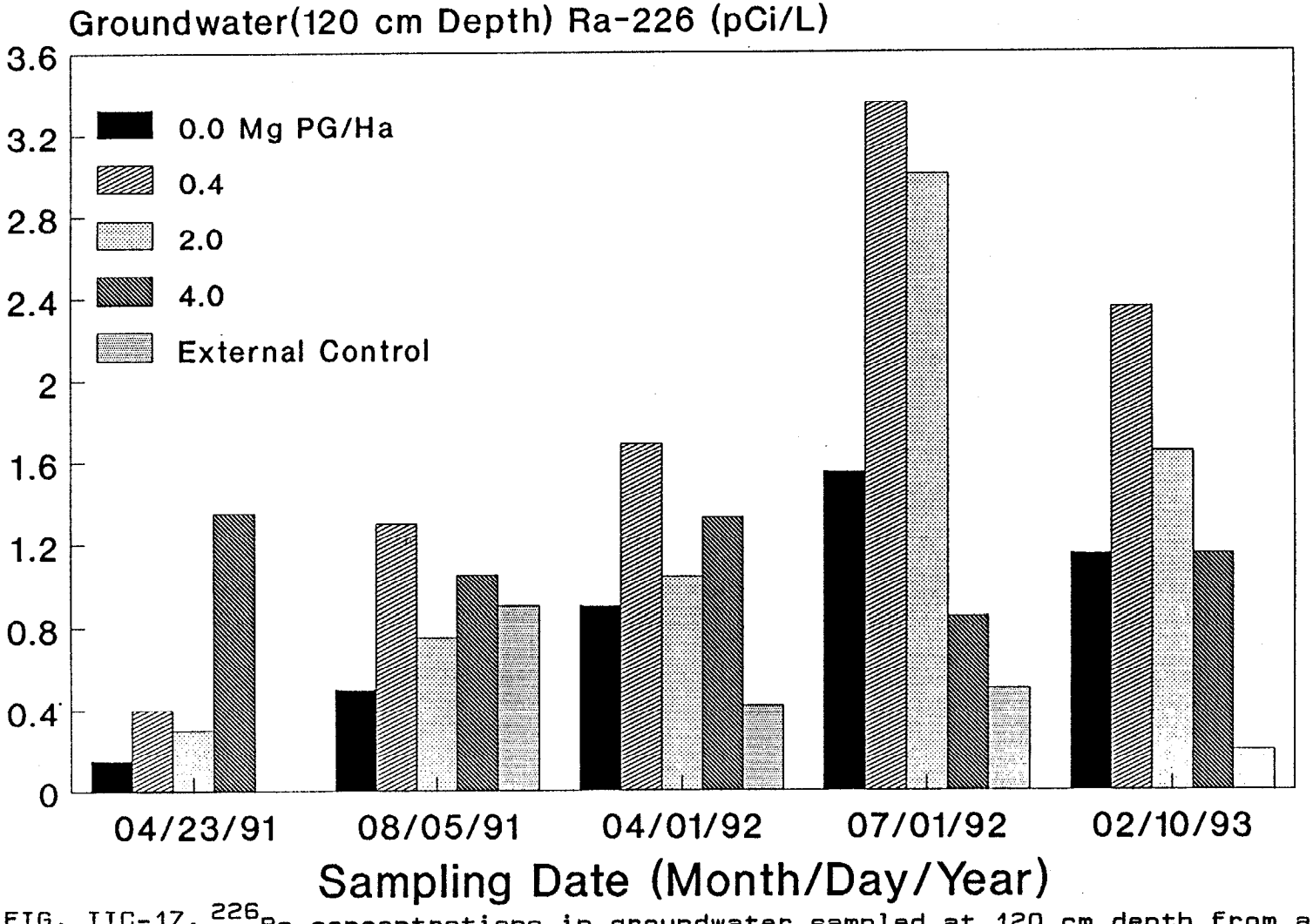

FIG. IIC-17. ^{226}Ra concentrations in groundwater sampled at 120 cm depth from a Florida Spodosol soil cropped to annual ryegrass which was amended with PG as source of S and Ca, by sampling, 1991-1993.

The maximum ^{226}Ra concentration observed in groundwater was 3.35 pCi L^{-1}. This value is below the current drinking water standard of 5 pCi L^{-1} (Federal Register, 1976) and well below the proposed Maximum Contaminant Level (MCL) of 20 pCi L^{-1} (Federal Register, 1991).

There is no obvious explanation for the apparent inverse relationship of ground water ^{226}Ra concentration with treatment level on the treated plots after the first collection. The data do raise the question as to whether there is sufficient communication in the ground water so that all the contiguous plots (including internal control) are affected by the total treatment. Observations suggesting such an effect include:

(a) the general increase with time of the ^{226}Ra concentrations for the internal controls in the absence of an increase at C_{ex},

(b) the similar behavior with time for all contiguous plots (control and treated), and

(c) the departure from a consistent relationship of concentration with treatment level.

This question of internal control contamination cannot be answered at this time.

Lead-210. The trends across treatments over time are given in Figure IIC-18. Concentrations at the external control plots C_{ex} averaged 0.67 pCi L^{-1} and ranged from 0.45 to 1.03 without any strong trend over the study period. Concentrations at the internal control plots C averaged 0.49 pCi L^{-1} and ranged from 0.05 to 0.95 without any strong trend with time.

The treated plots as a group tended to have higher ^{210}Pb concentrations than the controls and statistically-significant differences were observed for three of the four collections in 1991 and 1992 and for the 1992 annual average. However, after the first year, there was no consistent, meaningful correlation of concentration with treatment level.

Currently there is no drinking water standard for ^{210}Pb (a naturally-occurring beta emitter). The proposed MCL for beta emitters is the concentration resulting in an annual effective dose equivalent of 4 mrem yr^{-1} (Federal Register{ 1991). For ^{210}Pb, using contemporary dosimetry and a 2 L day ingestion, a concentration of 1 pCi L^{-1} corresponds to this dose limit. At four of the five collections, one or more sample sets exceeded 1 pCi L^{-1} (range of 1.03 to 2.50). These data indicate that the potential for ^{210}Pb in water deserves continued evaluation.

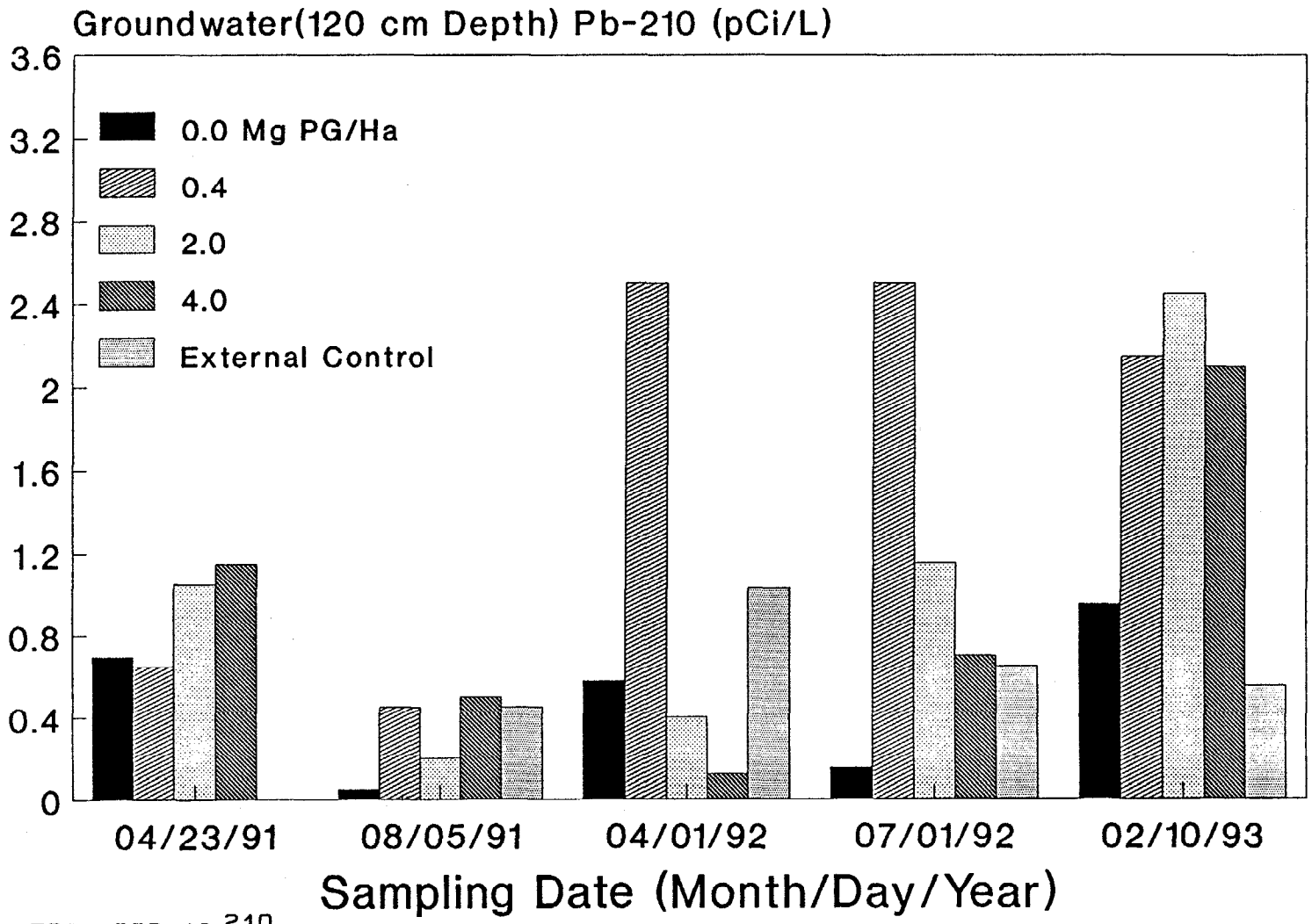

FIG. IIC-18. ^{210}Pb concentrations in groundwater sampled at 120 cm depth from a Florida Spodosol soil cropped to annual ryegrass which was amended with PG as source of S and Ca, by sampling, 1991-1993.

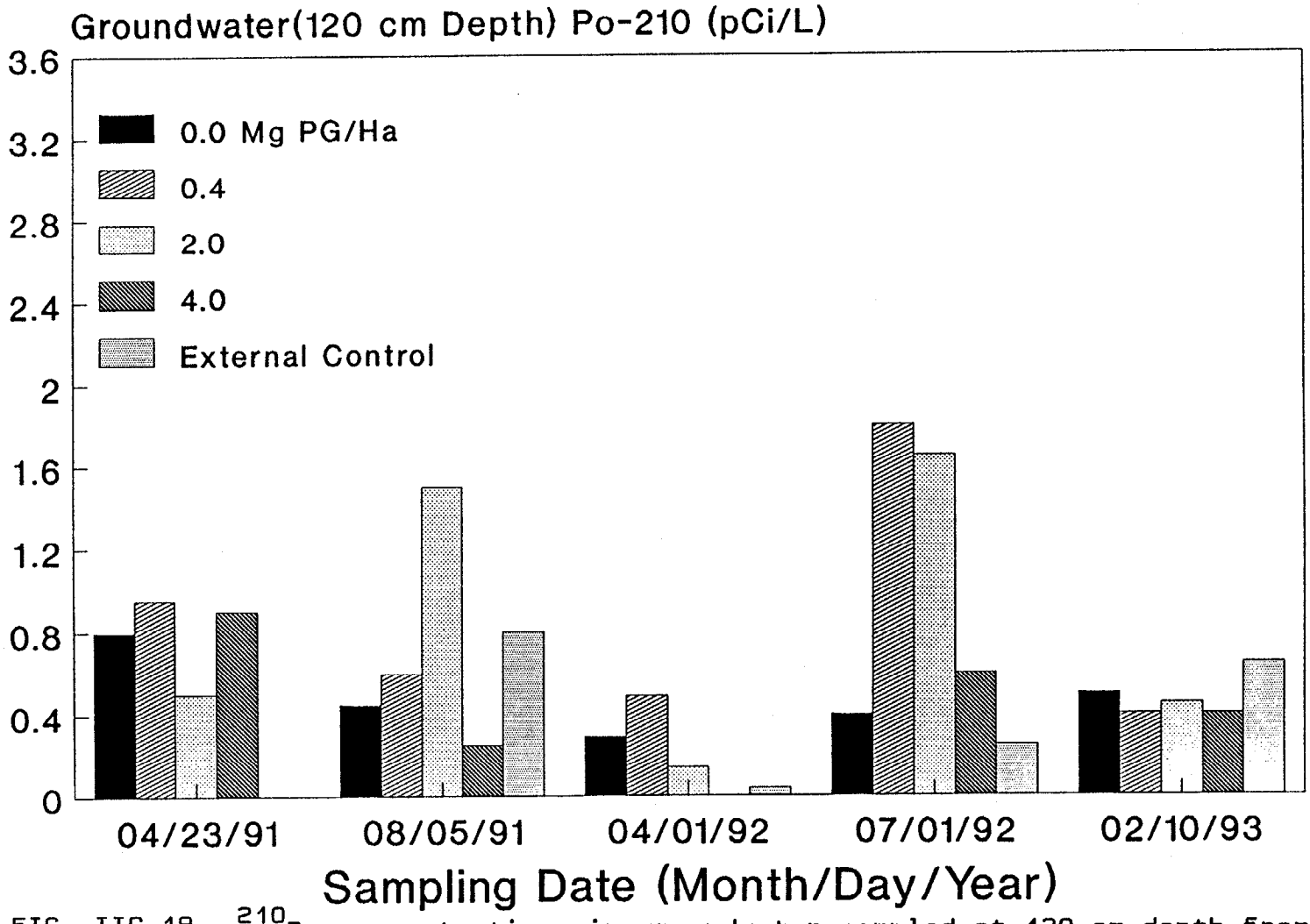

FIG. IIC-19. ^{210}Po concentrations in groundwater sampled at 120 cm depth from a Florida Spodosol soil cropped to annual ryegrass which was amended with PG as source of S and Ca, by sampling, 1991-1993.

Polonium-210. The trends with treatments and time are given in Figure IIC-19. Concentrations at the external control plots C averaged 0.49 pCi L^{-1} and ranged from 0.04 to 0.80. Concentrations at the internal control plots C were similar, averaging 0.49 pCi L^{-1} and ranging from 0.29 to 0.80. Neither external nor internal controls exhibited any strong trend with time.

While the treated plots as a group appeared to have higher ^{210}Po levels than the control plots for several collections, the treated plot concentrations did not correlate well with treatment levels. There were no statistically-significant differences observed for individual collections, annual averages, or the 3-year average.

There are no current or proposed explicit drinking water standards for ^{210}Po (a naturally-occurring alpha emitter): rather this radionuclide is included under limits for gross alpha emitters. The current standard specifies a limit for gross alpha emitters of 15 pCi L^{-1} (Federal Register, 1976). Under proposed rules (Federal Register, 1991), the MCL for "adjusted" gross alpha activity (excluding ^{226}Ra, uranium, and ^{222}Rn) is also 15 pCi L^{-1} All the ^{210}Po concentrations reported in this study were well below this value.

C.6. Soil Surface Rn Flux

The Rn flux from the surface of a tilled soil cropped to annual ryegrass was determined six times during the 3-year period. The data are summarized in Table C8; the results for individual sampling periods are shown graphically in Figure IIC-20 and presented in Appendix Table IIC-7.

The baseline (control plot) Rn flux values averaged 0.02 pCi m^{-2} s^{-1} and ranged from 0.015 to 0.030.

For the treated plots, the values ranged from 0.017 to 0.040 pCi m^{-2} s^{-1}. The data for all six readings suggested a treatment effect (Figure IIC-20). Differences were statistically significant for the 3-yr average (P=O.O05, P{linear}=0.002, 4.0>all, 2.0>0.0), the 1991-92 crop year average, and two of the individual sampling periods. There was evidence to support a linear treatment-response model for the 3-year mean, two of the annual averages, and one of the individual sampling. Over the first three years following application of PG to a tilled land cropped to annual ryegrass, the PG-attributable increase in Rn flux was on the order of 0.002 pCi m^{-2} s^{-1} per Mg ha^{-1}.

The values obtained for Rn flux were well within the range reported in the literature. Surface flux measurements have been reported to vary by a factor of 250, ranging from 0.005 to 1.41. The reported average for the U.S. is 0.43 pCi m^{-2} s^{-1} (Wilkening, et al., 1972; NCRPM, 1989).

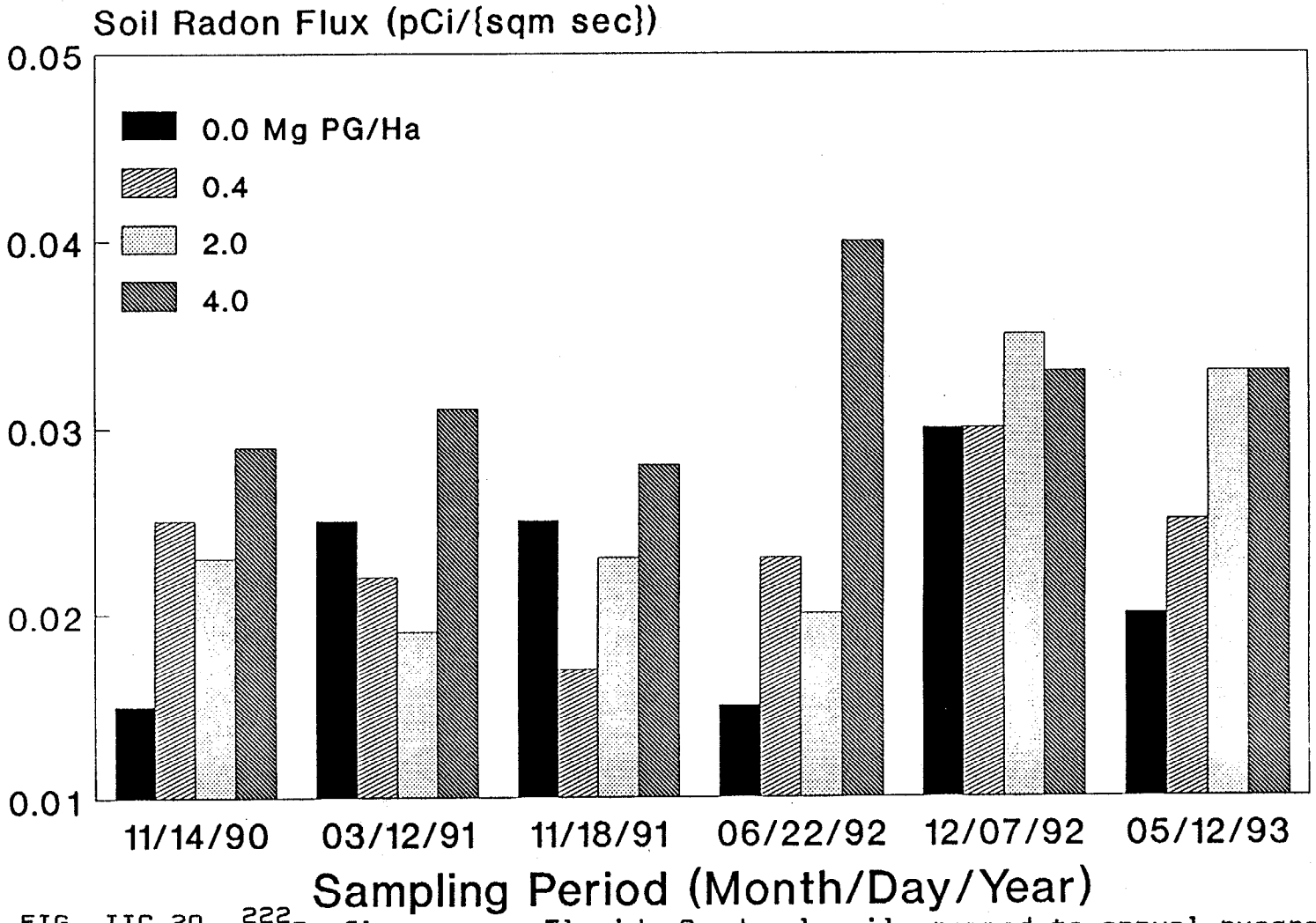

FIG. IIC-20. ^{222}Rn flux over a Florida Spodosol soil cropped to annual ryegrass which was amended with PG as source of S and Ca, by sampling, 1990-91 to 1992-93.

Table C8. Soil surface Rn flux over a Florida Spodosol soil cropped to annual ryegrass which was amended with phosphogypsum (PG) as a source of S and Ca, averaged by year and over a 3-year period, 1990-1992.

Treatment	Crop year 1990-91	1991-92	1992-93	mean
Mg PG ha^{-1}	---------- pCi $m^{-2}s^{-1}$ ----------			
0.0(C)	0.020	0.020	0.025	0.022
0.4	0.024	0.020	0.028	0.024
2.0	0.021	0.021	0.034	0.025
4.0	0.030	0.034	0.033	0.032
Statistics:				
P	0.1410	0.0380	0.2979	0.0054
P(Linear)	0.0660	0.0162	ns	0.0015
P(Quadratic)	ns[1]	ns	ns	ns
DMRT	ns	4.0>all	ns 2.0>C	4.0>all;

[1]not significant.

C.7. Ambient Atmospheric Rn

Following the initial treatment application, airborne Rn monitors were deployed continuously from 12/10/90 through 7/18/91 and from 10/07/91 through 04/30/93; during these times, average concentrations were reported for 13 time intervals ranging from 30 to 90 days, more or less. The data are summarized in Table C9; the results for individual sampling periods are shown in Fi gure IIC-21 and presented in Appendix Table IIC-8.

The averages and ranges of Rn concentrations measured at control stations were 0.20 (0.12 to 0.29) pCi L^{-1} for C_{ex1}, 0.23 (0.05 to 0.37) pCi L^{-1} for C_{ex2}, and 0.22 (0.11 to 0.32) for internal control C. The background ambient Rn concentration varied with time but there was no significant difference between the various external and internal control station measurements.

Concentrations over the PG-treated plots ranged from 0.08 to 0.37 pCi L^{-1}. There were no significant treatment effects for individual monitoring periods, annual averages, or the 3-year average. For comparison with another study that also used EIC's, individual outdoor ambient atmospheric Rn values for the U.S. were reported to range from 0.0 to 1.11 pCi L^{-1} with a median concentration of 0.39 pCi L^{-1}. Mean outdoor values ranged from 0.16 to 0.57 (Hopper et al., 1990).

Table C9. Ambient atmospheric Rn over a Florida Spodosol soil cropped to annual ryegrass which was amended with phosphogypsum (PG) as a source of S and Ca, averaged by year and over a 3-year period, 1990-91 to 1992-93.

Treatment	Crop year			
	1990-91	1991-92	1992-93	mean
Mg PG ha^{-1}	---------- pCi L^{-1} ----------			
0.0(C_{ex1})	0.19	0.20	0.20	0.20
0.0(C_{ex2})	-[a]	0.24	0.22	0.23
0.0(C)	0.20	0.25	0.15	0.22
0.4	0.18	0.23	0.21	0.21
2.0	0.17	0.23	0.17	0.20
4.0	0.14	0.18	0.24	0.17
Statistics:[1]				
P(w/$C_{ex1,2}$)	0.4083	0.6190	0.7355	0.3141
P(w/o $C_{ex1,2}$)	0.3424	0.5068	0.5529	0.2283

[a]No external control at this time.

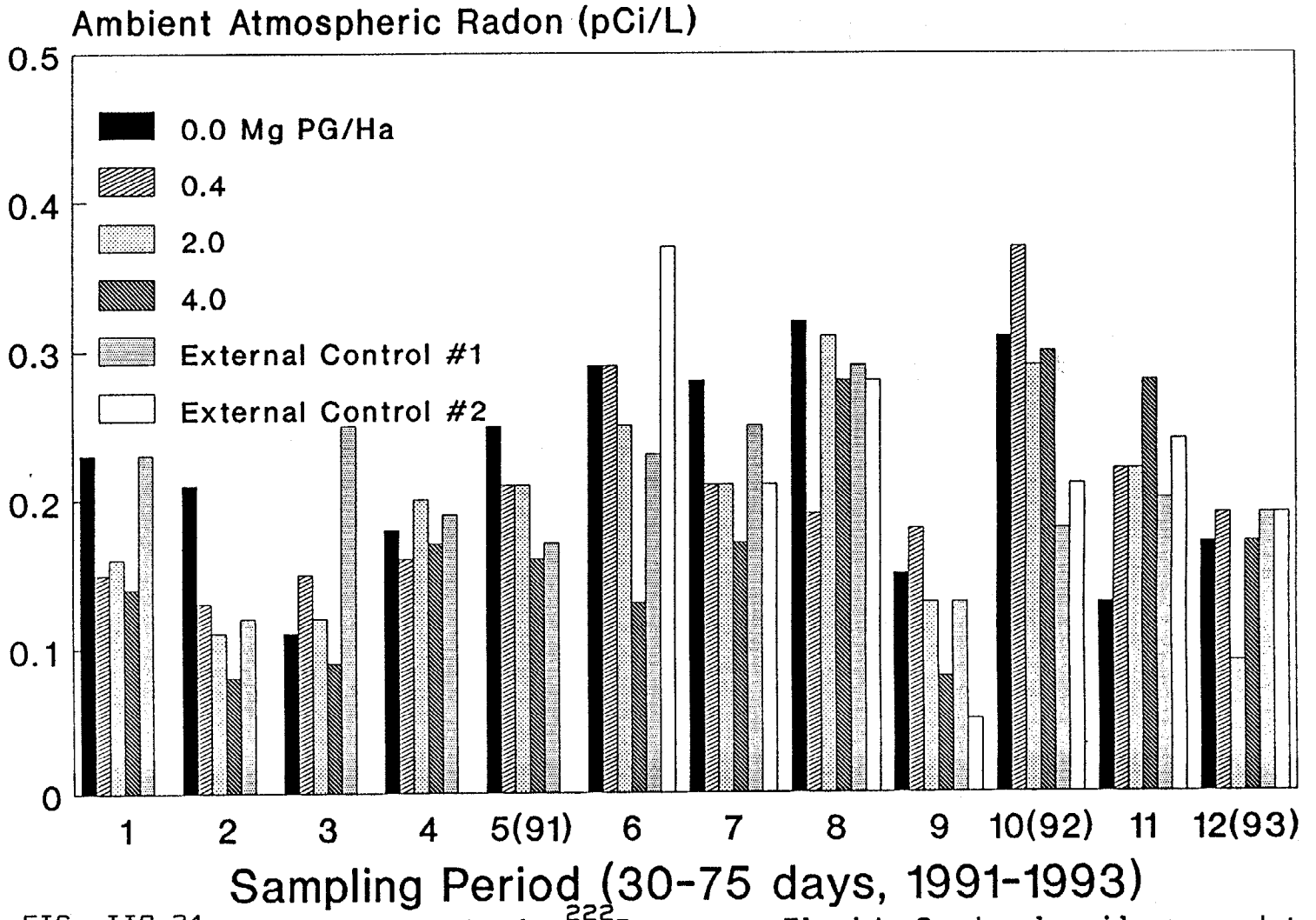

FIG. IIC-21. Ambient atmospheric ^{222}Rn over a Florida Spodosol soil cropped to annual ryegrass which was amended with PG as source of S and Ca, by sampling, 1990-91 to 1992-93.

SUMMARY, CONCLUSIONS, AND RECOMMENDATIONS

A. General Summary

Four field experiments were conducted to determine the effect of PG application to an established Pensacola bahiagrass (Paspalum notatum Flugge) pasture and to annual ryegrass (Lolium multiflorum Lam.),as a source of S and Ca, on forage yields and quality and on radiological and non-radiological environmental aspects associated with PG use. They were conducted by UF-IFAS, at RCREC, Ona, Florida. The soil was a typical Florida Spodosol soil. The experimental PG rates of 0.0, 0.4, 2.0, and 4.0 Mg PG ha^{-1} were broadcast by hand over the experimental plots. The 0.4 Mg treatment was applied annually for 3 years and the 2.0 and 4.0 Mg PG ha" treatments applied only at the beginning of the study. The PG was allowed to dissolve and leach into the soil profile by rain in the bahiagrass experiment. In ryegrass study, PG was mixed into the top 15-cm of the soil using a disk harrow before seeding.

The experiments ran from 1990 to 1993. The radiological analyses, except for ambient Rn and gamma radiation, were done by commercial environmental laboratories; the non-radiological analyses at the various UF-IFAS facilities. All data were statistically analyzed by the Department of Statistics of UF-IFAS.

1. Agro-environmental Aspects

a. Summary results:

Heavy metals and "toxicity characteristic". The pH of the PG used in the study was 4.6 in water (1:l). It dissolved at a constant rate of 2.6 and 4.3 g L^{-1} in water and in Mehlich 1 solution, respectively. Fluoride content was 0.43%.

The PG contained minute amounts (mg kg^{-1}) of As (5-0), Ba (46.0), Cd (0.7-l.l), Cr (2.9), Pb (4.0), Hg (<0.01), Se (<0.05-1.6), and Ag (<0.2-2.0). The leaching potentials (LPs) of the toxic metals in PG used in the study used by USEPA to determine the "toxicity characteristic" (TC) of solid wastes, as estimated, were 0.12 (As), 0.09 (Ba), 0.02 (Cd), 0.02 $_{-1}$(Cr), 0.03 (Pb), <0.001 (Hg), <0.001 (Se), and 0.30 (Ag) mg L^{-1} of leachate. The maximum contamination level (MCL) allowed by USEPA for solid wastes to exhibit "toxicity characteristic" for As, Ba, Cd, Cr, Pb, Hg, Se, and Ag are 5.0, 100, 1.0, 5.0, 5.0, 0.2, 1.0, and 5.0 mg L^{-1}, respectively.

"Toxicity Characteristic" increments due to PG. In ryegrass plots treated with 4.0 Mg PG ha^{-1}, the measured soil heavy metal extracted using Mehlich 3 (Mehlich, 1984) solution, in mg kg^{-1}, were 0.04-0.10 for Cd, 0.4-0.8 for Cr, 0.14-0.58 for Pb, 0.0-0.16

for Hg, and 0.0-0.16 for Se, sampled at 15 cm intervals down to 90 cm one year after application. The statistics showed no statistical difference between treatments at any soil depth.

If it is assume that the LPs of the metals in PG now mixed with the soil were the same as in pure PG, the TCs of the top 0-15 cm soil were estimated to increase by 0.0002 (As), 0.0002 (Ba), 0.00003 (Cd), 0.00003 (Cr), 0.00005 (Pb), 0.0000003 (Hg), 0.000006 (Se), and 0.0003 (Ag) mg L^{-1}. The increments in soil "toxicity characteristics" levels are well below the USEPA primary standards for drinking water of 0.05 (As), 1.00 (Ba), 0.01 (Cd) 0.05 (Cr), 0.05 (Pb), 0.002 (Hg), 0.01 (Se), and 0.05 (Ag) mg L^{-1}.

pH, E_c, (TDS), and F. In bahiagrass, the by-year average pH ranges during 3 years of sampling were 4.9 to 5.4 (control) and 4.3 to 6.3 (treated) for runoff; 4.5 to 4.8 (control) and 4.6 to 5.6 (treated) for samples at 60 cm depth: and 4.9 to 5.2 (control) and 4.0 to 5.8 (treated) for samples at 120 cm depth. In ryegrass, the by-year average pH ranges of groundwater samples from the control plots were 4.0 to 4.4 (60 cm depth) and 4.3 to 4.9 (120 cm depth), The pH ranges for samples from the treated plots were 3.7 to 4.4 (60 cm depth) and 4.1 to 4.9 (120 cm depth). In both bahiagrass and ryegrass experiments, PG had no effect on the by-year and 3-year average pH of groundwater at all depths.

The highest by-year (1990-91) and multi-year average E_c of 1421 and 1137 umho cm^{-1}, respectively, were noted in groundwater from plots treated with 4.0 Mg ha^{-1} sampled at the 60 cm depth from the ryegrass experiment. All other water samples at all depths and treatments had much lower E_cs than these values. These high values, however, are less than the upper E_c limit of 1500 umho cm^{-1} for potable water in the United States. Converting the highest measured annual (1421 umho cm^{-1}) and multi-year average (1137 umho cm^{-1}) E_c values into TDS (E_c x 0.66) gave 938 and 750 mg of dissolved solids L^{-1} which remained well below Florida's TDS standards of <3000 mg L^{-1} for agricultural use and <1000 mg L^{-1} for domestic and industrial uses.

The highest by-year and 3-year average F levels in surficial groundwater from the bahiagrass plots for all depths were 0.83 (by-year mean) and 0.36 (3-year mean) mg L^{-1}. Both were observed in samples from plots that received 2.0 Mg PG ha^{-1} and at 60 cm depth. In the tilled ryegrass plots, the highest by-year and 3-year F level averages in groundwater from all treated plots from both 60 and 120 cm depths were 0.53 (by-year) and 0.43 (2-year) mg L^{-1}, respectively.

It is widely accepted that approximately 1.0 Mg F L^{-1} in drinking water can effectively reduce dental caries without harmful effects on health. Also, the Florida drinking water primary standards allow for a maximum contamination level (MCL)

for F from 1.4 to 2.4 mg L^{-1}. None of the individual measured F values in the bahiagrass and ryegrass experiments exceeded the MCL values, and the by-year and 3-year average values were even less than 1.0 mg F L^{-1}.

2. Radiological Aspects

a. Summary Results: Bahiagrass

Effect on soil. PG containing ^{226}Ra, ^{210}Pb, and ^{210}Po at measured concentrations of 18, 31, and 24 pCi g^{-1}, respectively, was applied to land for which the top 15-cm layer of the untreated soil contained measured concentrations of 0.55, 0.61, and 0.53 pCi g^{-1}. It is calculated that each Mg PG ha^{-1} added 1820, 3080, and 2430 pCi m^{-2}, and increased the concentration in the upper 15-cm layer (at density = 1.5 g cm^{-3}) by 0.008, 0.014, and 0.011 pCi g^{-1}, respectively. For the maximum treatment of 4.0 Mg ha^{-1}, the calculated increases in the initial concentration, averaged over the 15-cm layer, were 0.032, 0.055, and 0.043 pCi g^{-1}, respectively (5.2%, 9.0%, and 8.1%).

Measurements of ^{226}Ra in the upper layer of the control plots had standard errors on the order of 0.1 to 0.2 pCi g^{-1} (20%) and the treatment effect could not be detected. Measurements of ^{210}Pb detected an effect of PG treatment in the top layer for the first year of sampling but it could not be detected in subsequent years. Measurements of ^{210}Po in the upper layer of the control plots had standard errors on the order of 0.2 pCi g^{-1} and the treatment effect could not be detected. Any downward transport of the three radionuclides over the three years could not be detected.

Gamma radiation. There was no consistent correlation of gamma-radiation level with PG treatment.

Effect on plant tissue. There was limited evidence of measurable ^{226}Ra uptake by forage. Treatment effects were statistically significant for the 3-year mean of the regrowth forage, two of the nine harvests of the regrowth forage, and the 3-year average of the mature forage. Uptake by forage during the first three years following PG treatment was on the order of 0.01 to 0.02 pCi g^{-1} per Mg ha^{-1}.

For ^{210}Pb, measured concentrations in regrowth forage had little consistent relationship to PG treatment levels; and in mature forage there were no suggested trends or statistically-significant treatment effects. Thus, no effect of ^{210}Pb uptake by bahiagrass regrowth or mature forage from PG application could be observed from these data.

For ^{210}Po, there was limited evidence of a treatment effect in regrowth forage, particularly in the first year, and a suggested, but not statistically-significant, effect on mature forage. The data were too variable to provide a basis for calculating an uptake factor.

Effect on surficial groundwater. Radium-226 was observed in the runoff during the first two samplings following PG application: this effect appeared to persist into the third year. The data suggest a small treatment effect at the 60-cm depth, but differences were not statistically significant. Results for the 120-cm depth were equivocal -- concentrations for the treated plots were generally higher than for the control plots but the results did not correlate well with treatment level and the differences were not statistically significant. The maximum concentration observed, 1.8 pCi L^{-1}, was well below the current and proposed drinking water standards of 5 and 20 pCi L^{-1}.

For ^{210}Pb, no statistically-significant differences were observed for runoff and any apparent effect was transient. The single sampling at 60 cm indicated a statistically-significant effect, linear with PG level: but this is limited data on which to base a conclusion. Data for the 120-cm depth suggested that treated plots as a group had higher concentrations than control plots, but the suggested effects did not correlate well with treatment level and statistically-significant differences were limited and equivocal. Concentrations in samples from the wells in PG-treated plots ranged up to 2.5 pCi L^{-1}; 1/3 of these samplings exceeded 1 pCi L^{-1}, the concentration corresponding to the proposed dose limit for beta emitters in drinking water.

Data for ^{210}Po exhibited considerable variability and no meaningful treatment effects were observed in runoff or from the wells. The maximum treated plot value of 1.5 pCi L^{-1} was less than the maximum control plot value of 2.8 pCi L^{-1}; and both of these values were below the current and proposed 15 pCi L^{-1} standards for gross alpha activity in drinking water.

Radon flux and atmospheric Rn. Rn flux values for control and treated plots were quite low, reported values ranged from 0.01 to 0.05 pCi m^{-2} s^{-1}. The data suggested slightly higher values for the higher treatment level plots than for the control (on the order of 0.003 pCi m^{-2} s^{-1} per Mg ha^{-1} applied PG), but differences were not statistically significant.

The background atmospheric Rn concentration, averaged over periods of 30 to 90 days, varied with time. There was no meaningful treatment effect. It was speculated that, due to atmospheric mixing, any small effect at 1 m above the surface that might have occurred was shared by the contiguous plots.

b. Summary Results: Annual Ryegrass

Effect on soil. Phosphogypsum containing ^{226}Ra, ^{210}Pb, and ^{210}Po at measured concentrations of 18, 31, and 24 pCi g^{-1}, respectively, was applied to land for which the top 15-cm layer of the untreated soil contained measured concentrations of 0.59, 0.69, and 0.57 pCi g^{-1}. It is calculated that each Mg PG ha^{-1} added 1820 ^{226}Ra, 3080 ^{210}Pb, and 2430 ^{210}Po pCi m^{-2}, and increased the concentration in the upper 15-cm layer (at density = 1.5 g cm^{-3}) by 0.008, 0.014, and 0.011 pCi g^{-1}, respectively. For the maximum treatment of 4.0 Mg ha^{-1}, the calculated increases in the initial concentration, averaged over the upper 15-cm layer, were 0.032, 0.055, and 0.043 pCi g^{-1} (5.4%, 8.0%, and 7.5%0), respectively.

No effect of PG treatment on soil ^{226}Ra was observed in 1991 and 1992. The 1993 data suggested an effect with downward migration but none of the differences by treatment were statistically significant. Results for ^{210}Pb were equivocal: overall, no meaningful relationship between concentrations in soil and treatment level could be deduced from the data. There was very limited evidence for a treatment effect for ^{210}Po in the upper soil layer. However, the effect was not seen in 1991; while the 1992 differences were statistically significant, the concentration for the 2.0 Mg ha^{-1} treatment was higher than for the 4.0 treatment; and the apparent effect in 1993 was not statistically significant. In general, the PG-attributable additions to the soil concentrations of ^{226}Ra, ^{210}Pb, and ^{210}Po could not be measured consistently in presence of the variations associated with the natural background and with the measurements.

Gamma radiation. No PG-attributable increases in background radiation were detected.

Effect on plant tissue. The data suggest a PG-attributable ^{226}Ra uptake by ryegrass regrowth and mature forage persisting into the post-application year: however, statistically-significant effects were limited. The estimated uptake factor is on the order of 0.039 pCi g^{-1} per Mg ha^{-1}.

Annual average ^{210}Pb values for regrowth forages from plots treated with 4.0 Mg PG ha^{-1} appeared to be elevated. However, there were no statistically-significant differences with respect to treatment level for regrowth or mature forage harvests or averages. There was very limited suggestive indication, and no statistical evidence, of a measurable effect of PG treatment on ^{210}Po in regrowth and mature ryegrass forage.

Effect on surficial groundwater. The data suggest that PG treatment increased the ^{226}Ra concentration in groundwater collected at 120 cm: however, statistically-significant differences were observed only for the first post-treatment collection and the 1991 crop-year average and the treated plots

results did not correlate well with treatment level. The data raised the question as to whether there might be sufficient communication in the ground water so that all the contiguous plots (including internal control) were affected by the total treatment. The maximum ^{226}Ra concentration observed in groundwater, 3.35 pCi L^{-1}, is below the current drinking water standard of 5 pCi L^{-1} and well below the proposed Maximum Contaminant Level (MCL) of 20 pCi L^{-1}.

For ^{210}Pb, the treated plots as a group tended to have higher concentrations than the controls: however, after the first year, there was no consistent, meaningful correlation of concentration with treatment level. Concentrations ranged up to 2.5 pCi L^{-1}; at four of the five collections, one or more sample sets exceeded 1 pCi L^{-1}, the concentration corresponding to the proposed dose limit for beta emitters in drinking water.

No meaningful effects of PG treatment were observed for ^{210}Po in groundwater. All concentrations were well below the current and proposed 15 pCi L^{-1} standards for gross alpha activity in drinking water.

Radon flux and atmospheric Rn. Rn was flux increased by PG treatment: the evidence was statistically significant for the 3-year average, the 1991-92 crop year average, and two of the six individual sampling. Statistically-significant evidence to support a linear treatment-response model was presented by the 3-year average and the first two annual averages.

Ambient atmospheric Rn concentrations measured at 1 m over the test plots and external control plots varied with time but no effects of PG treatment were detected.

B. Conclusions

1. Agro-environmental aspects

a. Heavy metals. Application of PG used in the study to soil at 4 Mg PG ha^{-1} had no effect on the levels of 5 of the 8 USEPA "toxicity characteristic" metals at the top O-15 cm down to 90 cm of the soil profile, determine one year from the time of application. The other three were not determined in the soil, but their concentrations in PG were also extremely low. The estimated increments in "toxicity characteristics" of the 8 metals in soils to which PG was applied are well below the MCL levels standards for drinking water. Hence, it is concluded that the heavy metals concentrations in PG, the increments in their concentrations in the soil upon PG application, and their leaching potentials in the soil are extremely low to be of any concern to the soil and to the surficial groundwater.

b. pH, E_c (TDS), and F. The pH, E_c, and F levels of runoff and groundwater sampled at 60 and 120 cm depths had not been impacted by PG application. Mean E_c converted to TDS showed that the various water samples met the Florida water quality for domestic, industrial, and agricultural uses. The mean F levels in the various water samples were less than 1.0 mg F L^{-1}, the level which is known to prevent dental caries.

2. Radiological Aspects

Established bahiagrass pasture. The following conclusions are drawn about the radiological effects of the application of PG to an established bahiagrass pasture:

(1) Effect on soil - Radioactivity contributions by PG treatments up to 4.0 Mg ha^{-1} are small relative to the natural activity of even low-background soils. (Any addition cannot be detected by conventional analytical techniques in a well-mixed sample collected from a 0-15 cm depth).

(2) Gamma radiation - Any influence of PG at treatment levels up to 4 Mg ha^{-1}, is insignificant in comparison to temporal and spatial variations in the local background radiation.

(3) Radium-226 uptake by bahiagrass forage - Contributions by PG at the application levels used are small relative to the variation in the background level of ^{226}Ra and difficult to measure. A crude estimate of uptake is 0.01 to 0.02 pCi g^{-1} per Mg PG ha^{-1}.

(4) Lead-210 uptake by bahiagrass forage - No uptake in regrowth or mature forage of Pb from PG treatment could be observed and no uptake factor can be calculated.

(5) Polonium-210 uptake by bahiagrass forage - This experiment detected only limited evidence of uptake of ^{210}Po from PG at the treatment levels used, any contribution was small relative to the variations in the background levels. The data were too variable to support calculation of an uptake factor.

(6) Radium-226 in groundwater - This radionuclide appears in runoff during the first three years after PG application. The data from this experiment suggest, but do not confirm, an effect at 60 cm and 120 cm. Concentrations in runoff and shallow well water during the first three years following PG application up to 4 Mg ha^{-1} should be well below current and proposed drinking water standards.

(7) Lead-210 in groundwater - The data were equivocal; there was very limited evidence for an effect of PG treatment up to 4 Mg ha^{-1} on the well water. However, comparison of observed levels (up to 2.5 pCi L^{-1}) to proposed criteria (1 pCi L^{-1})

indicates that the potential for ^{210}Pb in water deserves further evaluation.

(8) Polonium-210 in groundwater - No effect of PG treatment up to 4 Mg ha^{-1} could be detected by this experiment. Concentrations in runoff and shallow well water during the first three years after PG treatment up to 4 Mg ha^{-1} should be well below the 15 pCi L^{-1} existing and proposed drinking water standards.

(9) Rn flux - Any increase in Rn flux attributable to PG treatment at levels up to 4 Mg ha^{-1} is insignificant in relation to the variations in the natural levels of Rn flux. Consequently, any effect on indoor Rn in future structures built over lands treated at this level will be insignificant relative to the variations experienced in indoor Rn levels. The treatment levels used in this experiment were not sufficient to provide reliable empirical data for modeling Rn emissions from PG-application practices.

(10) Atmospheric Rn - Because of air movement and mixing, concentrations at 1 m over individual plots of the size used in this experiment are probably not uniquely representative of the Rn emission of the respective plots.

(11) Overall, the radiological impact on the environment of application of Central Florida PG to an established bahiagrass pasture at levels up to 4.0 Mg ha^{-1} was minimal when compared to the levels and variations of the natural occurrence of the radionuclides studied. Additions attributable to the PG application were typically at or below the limit of detection by contemporary conventional radiation and radioactivity monitoring techniques.

Tilled land cropped to annual ryegrass. On the basis of this work, the following conclusions are drawn about the radiological effects of the application of PG to tilled land cropped to annual ryegrass:

(1) Effect on soil - Radioactivity contributions by PG treatments up to 4.0 Mg ha^{-1} are small relative to the natural activity of even low-background soils. (Any addition cannot be detected by conventional analytical techniques in a well-mixed sample collected from a 0-15 cm depth).

(2) Gamma radiation - Any influence of PG at treatment levels up to 4 Mg ha^{-1} is insignificant in comparison to temporal and spatial variations in the local gamma-radiation background.

(3) Radium-226 uptake by ryegrass forage - Contributions by PG at the rates used are small relative to the variation in the background level of ^{226}Ra and difficult to measure. The uptake is estimated to be 0.039 pCi g^{-1} per Mg PG ha^{-1}.

(4) Lead-210 uptake by ryegrass forage - Uptake by regrowth or mature forage of ^{210}Pb from PG treatment up to 4 Mg ha^{-1} is too small to be detected by the methods employed in this study and no uptake factor can be calculated.

(5) Polonium-210 uptake by ryegrass forage - Uptake by regrowth or mature forage of ^{210}Po from PG treatment up to 4 Mg ha^{-1} is too small to be detected by the methods employed in this study and no uptake factor can be calculated.

(6) Radium-226 in groundwater - This radionuclide appeared in 120-cm groundwater during the first year after PG treatment but the data are equivocal for subsequent years. Concentrations in 120-cm depth groundwater during the first three years following PG application up to 4 Mg ha^{-1} should be well below current and proposed drinking water standards.

(7) Lead-210 in groundwater - There is limited but equivocal evidence for an effect of PG treatment up to 4 Mg ha^{-1} on 120-cm depth ground water. Comparison of observed levels (up to 2.5 pCi L^{-1}) to proposed criteria (1 pCi L^{-1}) indicate that the potential for ^{210}Pb in water deserves further evaluation.

(8) Polonium-210 in groundwater - No effect of PG treatment on 120-cm ground water could be detected by this experiment. Concentrations during the first three years after PG treatment up to 4 Mg ha^{-1} should be well below the 15 pCi L^{-1} existing and proposed drinking water standards.

(9) Rn flux - Rn flux attributable to PG applied to tilled land cropped to annual ryegrass is estimated to be on the order of 0.002 pCi m^{-2} s^{-1} per Mg PG ha^{-1} during the first several years following the PG application.

(10) Atmospheric Rn - Because of air movement and mixing, concentrations at 1 m over individual contiguous plots of the size used in this experiment are probably not uniquely representative of the Rn emission of the respective plots.

(11) Overall, the radiological impact on the environment of application of Central Florida PG to tilled land cropped to annual ryegrass at levels up to 4.0 Mg ha^{-1} was minimal when compared to the levels and variations of the natural occurrence of the radionuclides studied. Additions attributable to the PG application were typically at or below the limit of detection by contemporary conventional radiation and radioactivity monitoring techniques.

C. Recommendations

Based on the experimental data from both the established bahiagrass pasture and the tilled land cropped to annual ryegrass experiments, the following recommendations are made:

Non-radiological Aspects

At rates up to 4 Mg PG ha^{-1}, the heavy metal impurities Cd, Cr, Pb, Hg, and Se had no impact in the soil due to their very low concentrations in PG. The evaluation of the leaching potentials of the heavy metals, including As, Ba, and Ag, using the extensive experimental data of May and Sweeney (1983) and their measured concentrations in the PG used in the study, precluded any measurable impact of these metals in surficial groundwater. The $CaSO_4.2H_2O$ itself and F had little effect on the quality of surficial groundwater. To put the heavy metal issue to rest, it is recommended that their determinations in soil, groundwater, and plant tissue be included in future studies using PG at rates higher than 4 Mg ha^{-1}.

Radiological Aspects

1. The radiation and the radionuclide levels attributable to PG used in the study with 18 pCi g^{-1} of ^{226}Ra applied up to 4 Mg PG ha^{-1} were not statistically differentiable from natural background levels. Thus, current restrictions on the agricultural use of PG should be relaxed.

2. Further radiological studies to produce statistically measurable effects and define time-dependent behavior and thus provide the solid data on which to base risk assessments should include the following:

* Extend the range of treatments to higher application rates to improve the chances of observing effects and providing measurements of quantitative relationships.

* Increase the replication (number of plots per treatment) to compensate for variability, increase the sensitivity for detecting differences, and improve the statistical power of the study.

* To evaluate contributions to ambient atmospheric Rn, further studies should simulate an extended treated field and/or isolate the air columns over the plots.

* After the initial two or three years, continue observations on treated and control plots for those parameters showing measurable effects. This should be done for periods long enough to quantitatively describe the time-dependent behavior. This would provide the basis for modeling the long-term effects of single and repetitive PG application programs.

3. While the work provides suggestions of certain trends, quantitative relationships are obscured by the low level of radioactivity involved and the natural variability of the parameters being measured. To derive quantitative and kinetic relationships to use in modeling to support future assessments, further studies would require higher PG application rates, more replication, and/or more sensitive analytical techniques.

A study using PG rates up to 20 Mg ha^{-1} and a greater number of replicates to improve measurability of the radiological parameters and reliability of the measured data is now being conducted by UF-IFAS under the support of FIPR.

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APPENDIX (BY-SAMPLING STATISTICS)

Table IIAB-1. Electrical conductivity (E_c), fluoride content (F), and pH of runoff from a Florida Spodosol soil cropped to bahiagrass which was amended with PG as source of S and Ca, by sampling, 1990-1992.

Sampling date	Treatment	E_c	F	pH
10-04-1990	Mg PG ha^{-1}	umho cm^{-1}	mg L^{-1}	
	0.0(C)	319.0	0.22	5.4
	0.4	259.0	0.27	6.3
	2.0	453.0	0.42	5.5
	4.0	624.5	0.69	5.9
	Statistics:			
	P	0.1462	0.1914	0.5680
	P(Linear)	0.0449	0.0566	ns[1]
06-19-1991	Mg PG ha^{-1}	umho cm^{-1}	mg L^{-1}	
	0.0(C)	430.0	0.07	5.5
	0.4	227.5	0.11	5.7
	2.0	233.5	0.11	5.2
	4.0	347.5	0.11	4.7
	Statistics:			
	P	0.7353	0.9365	0.4841
10-04-1991	Mg PG ha^{-1}	umho cm^{-1}	mg L^{-1}	
	0.0(C)	259.0	0.10	5.3
	0.4	269.0	0.15	6.2
	2.0	280.0	0.14	5.5
	4.0	570.5	0.13	3.9
	Statistics:			
	P	0.5006	0.8826	0.2185
07-01-1992	Mg PG ha^{-1}	umho cm^{-1}	mg L^{-1}	
	0.0(C)	132.0	0.11	5.1
	0.4	171.0	0.16	5.9
	2.0	182.5	0.12	5.9
	4.0	334.0	0.12	5.3
	Statistics:			
	P	0.0632	0.5466	0.3796
	P(Linear)	0.0207	ns	ns
	DMRT	4.0>all	ns	ns

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Table IIAB-1. Electrical conductivity (E_c), fluoride content (F), and pH of runoff from a Florida Spodosol soil cropped to bahiagrass which was amended with PG (Continuation).

Sampling date	Treatment	E_c	F	pH
10-01-1992	Mg PG ha^{-1}	umho cm^{-1}	mg L^{-1}	
	0.0(C)	297.0	0.30	4.7
	0.4	453.0	0.96	6.0
	2.0	98.5	0.47	4.9
	4.0	225.0	0.71	4.7
	Statistics:			
	P	0.1829	0.0099	0.3460
	DMRT	ns	0.4>4.0 >2.0>C	ns

[1]ns=not significant.

Table IIAB-2. Electrical conductivity (E_c), fluoride content (F), and pH of groundwater sampled at 60 cm depth from a Florida Spodosol soil cropped to bahiagrass which was amended with PG as source of S and Ca, by sampling, 1990-1992.

Sampling date	Treatment	E_c	F	pH
09-12-1990	Mg PG ha^{-1}	umho cm^{-1}	mg L^{-1}	
	0.0(C)	168.0	0.13	4.6
	0.4	201.0	0.16	5.0
	2.0	89.0	0.18	5.8
	4.0	113.0	0.29	5.3
	Statistics:			
	P	0.0685	0.2588	0.0759
	DMRT	0.4>C; C>2.0, 4.0	ns[1]	ns
09-19-1990	Mg PG ha^{-1}	umho cm^{-1}	mg L^{-1}	
	0.0(C)	126.0	0.06	4.5
	0.4	192.0	0.07	4.5
	2.0	167.5	0.07	5.2
	4.0	305.5	0.07	4.7
	Statistics:			
	P	0.9208	0.6550	0.6674
10-04-1990	Mg PG ha^{-1}	umho cm^{-1}	mg L^{-1}	
	0.0(C)	253.0	0.13	4.6
	0.4	460.5	0.17	4.5
	2.0	540.8	0.23	4.9
	4.0	974.3	0.28	4.5
	Statistics:			
	P	0.0902	0.1069	0.8675
	P(Linear)	0.0271	0.0311	ns
	DMRT	4.0>C	4.0>C	ns

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Table IIAB-2. Electrical conductivity (E_c), fluoride content (F), and pH of groundwater sampled at 60 cm depth from a Florida Spodosol soil cropped to bahiagrass (Continuation).

Sampling date	Treatment	E_c	F	pH
06-19-1991	Mg PG ha^{-1}	umho cm^{-1}	mg L^{-1}	
	0.0(C)	273.0	0.15	4.7
	0.4	499.0	0.21	4.2
	2.0	450.5	0.40	4.8
	4.0	1582.5	0.33	4.1
	Statistics:			
	P	0.0214	0.1940	0.5201
	P(Linear)	0.0075	ns[1]	ns
	P(Quad.)	0.0727	ns	ns
	DMRT	4.0>all	ns	ns
10-04-1991	Mg PG ha^{-1}	umho cm^{-1}	mg L^{-1}	
	0.0(C_{ex})	357.5	0.11	5.5
	0.0(C)	444.0	0.09	5.6
	0.4	192.5	0.10	5.3
	2.0	316.0	0.15	7.1
	4.0	774.5	0.15	6.4
	Statistics:			
	P(W/C_{ex})	0.3214	0.8316	0.5960
	P(W/o C_{ex})	0.2860	0.6360	0.5883
04-01-1992	Mg PG ha^{-1}	umho cm^{-1}	mg L^{-1}	
	0.0(C_{ex})	232.5	0.25	5.1
	0.0(C)	211.0	0.18	4.4
	0.4	229.0	0.11	4.7
	2.0	254.0	0.30	5.6
	4.0	821.5	0.31	5.1
	Statistics:			
	P(W/C_{ex})	0.0720	0.4899	0.8216
	P(w/o C_{ex})	0.0998	0.4387	0.5928
	P(Linear)	0.0427	ns	ns
	DMRT	4.0>C, 0.4	ns	ns

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Table IIAB-2. Electrical conductivity (E_c), fluoride content (F), and pH of groundwater sampled at 60 cm depth from a Florida Spodosol soil cropped to bahiagrass (Continuation).

Sampling date	Treatment	E_c	F	pH
07-01-1992	Mg PG ha^{-1}	umho cm^{-1}	mg L^{-1}	
	0.0(C_{ex})	177.5	0.08	4.9
	0.0(C)	145.0	0.11	4.5
	0.4	366.5	0.16	4.1
	2.0	157.5	0.25	5.5
	4.0	277.5	0.39	5.0
	Statistics:			
	P(w/C_{ex})	0.1776	0.1095	0.6573
	P(w/o C_{ex})	0.1548	0.1922	0.3444
	P(linear)	ns	0.0663	ns
10-01-1992	Mg PG ha^{-1}	umho cm^{-1}	mg L^{-1}	
	0.0(C_{ex})	176.0	0.43	4.1
	0.0(C)	196.0	0.36	4.3
	0.4	217.5	0.69	5.1
	2.0	176.0	2.00	5.0
	4.0	451.5	1.02	5.3
	Statistics:			
	P(w/C_{ex})	0.7916	0.0329	0.5050
	P(w/o C_{ex})	0.7900	0.0304	0.5337
	P(linear)	ns	0.0418	ns
	DMRT	ns	2.0>all	ns

[1]ns=not significant.

Table IIAB-3. Electrical conductivity (E_c), fluoride content (F), and pH of groundwater sampled at 120 cm depth from a Florida Spodosol soil cropped to bahiagrass which was amended with PG as source of S and Ca, by sampling, 1990-1992.

Sampling date	Treatment	E_c	F	pH
09-05-1990	Mg PG ha^{-1}	umho cm^{-1}	mg L^{-1}	
	0.0(C)	149.0	0.12	5.1
	0.4	238.8	0.26	5.7
	2.0	250.5	0.20	5.4
	4.0	272.8	0.25	4.7
	Statistics:			
	P	0.5387	0.4818	0.4124
09-12-1990	Mg PG ha^{-1}	umho cm^{-1}	mg L^{-1}	
	0.0(C)	181.5	0.13	5.5
	0.4	284.3	0.26	6.5
	2.0	269.0	0.30	5.4
	4.0	304.5	0.26	5.5
	Statistics:			
	P	0.5711	0.4421	0.4444
09-19-1990	Mg PG ha^{-1}	umho cm^{-1}	mg L^{-1}	
	0.0(C)	163.0	0.06	5.2
	0.4	241.0	0.07	5.7
	2.0	257.5	0.07	5.2
	4.0	322.8	0.09	4.9
	Statistics:			
	P	0.1908	0.0660	0.6543
	P(Linear)	0.0693	0.0254	ns[1]
	DMRT	ns	4.0>C	ns
10-04-1990	Mg PG ha^{-1}	umho cm^{-1}	mg L^{-1}	pH
	0.0(C)	192.8	0.10	5.1
	0.4	290.3	0.25	5.5
	2.0	279.8	0.20	4.9
	4.0	335.5	0.18	4.6
	Statistics:			
	P	0.5360	0.4785	0.3438

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Table IIAB-3. Electrical conductivity (E_c), fluoride content (F), and pH of groundwater sampled at 120 cm depth from a Florida Spodosol soil cropped to bahiagrass (Continuation).

Sampling date	Treatment	E_c	F	pH
04-26-1991	Mg PG ha^{-1}	umho cm^{-1}	mg L^{-1}	
	0.0(C_{ex})	494.5	0.24	5.5
	0.0(C)	162.0	0.08	5.0
	0.4	285.0	0.17	5.6
	2.0	662.5	0.20	5.1
	4.0	238.5	0.21	5.0
	Statistics:			
	P(w/C_{ex})	0.5259	0.4580	0.8327
	P(w/o C_{ex})	0.5329	0.4094	0.6353
06-19-1991	Mg PG ha^{-1}	umho cm^{-1}	mg L^{-1}	
	0.0(C_{ex})	678.5	0.33	6.4
	0.0(C)	275.5	0.08	4.2
	0.4	504.5	0.14	5.0
	2.0	1218.5	0.39	4.3
	4.0	508.0	0.29	5.4
	Statistics:			
	P(w/C_{ex})	0.1687	0.1606	0.0377
	P(w/o C_{ex})	0.1959	0.0743	0.1431
	P(Linear)	ns	0.0683	0.0661
	P(Quad.)	0.0653	0.0522	ns
	DMRT	ns	2.0>C, 0.4	ns
10-04-1991	Mg PG ha^{-1}	umho cm^{-1}	mg L^{-1}	
	0.0(C_{ex})	558.5	0.48	6.5
	0.0(C)	252.5	0.06	5.4
	0.4	287.5	0.11	5.7
	2.0	1042.8	0.13	4.9
	4.0	749.5	0.16	5.5
	Statistics:			
	P(w/C_{ex})	0.1175	0.5527	0.1216
	P(w/o C_{ex})	0.0607	0.5055	0.3167
	P(Linear)	0.0930	ns	ns
	P(Quad.)	0.0461	ns	ns
	DMRT	2.0>C, 0.4	ns	ns

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Table IIAB-3. Electrical conductivity (E_c), fluoride content (F), and pH of groundwater sampled at 120 cm depth from a Florida Spodosol soil cropped to bahiagrass (Continuation).

Sampling date	Treatment	E_c	F	pH
04-01-1992	Mg PG ha^{-1}	umho cm^{-1}	mg L^{-1}	
	0.0(C_{ex})	374.0	0.08	5.6
	0.0(C)	210.5	0.11	5.0
	0.4	395.5	0.19	4.9
	2.0	884.0	0.28	4.0
	4.0	751.5	0.19	4.8
	Statistics:			
	P(w/C_{ex})	0.0841	0.2421	0.2221
	P(w/o C_{ex})	0.0813	0.3055	0.2876
	P(Linear)	0.0533	ns[1]	ns
	P(Quad,)	0.0760	ns	ns
	DMRT	2.0>C	ns	ns
07-01-1992	Mg PG ha^{-1}	umho cm^{-1}	mg L^{-1}	
	0.0(C_{ex})	343.5	0.28	5.4
	0.0(C)	249.0	0.08	4.9
	0.4	471.5	0.15	5.1
	2.0	824.0	0.32	4.5
	4.0	790.0	0.25	4.0
	Statistics:			
	P(w/C_{ex})	0.1311	0.5551	0.4106
	P(w/o C_{ex})	0.1593	0.3543	0.2822
	P(Linear)	0.0764	ns	0.0977
10-01-1992	Mg PG ha^{-1}	umho cm^{-1}	mg L^{-1}	
	0.0(C_{ex})	526.5	1.13	5.0
	0.0(C)	206.0	0.29	4.6
	0.4	428.5	0.45	5.3
	2.0	451.5	0.60	5.1
	4.0	773.0	0.55	4.9
	Statistics:			
	P(w/C_{ex})	0.4377	0.5896	0.6210
	P(w/o C_{ex})	0.1404	0.5373	0.1559
	P(Linear)	0.0408	ns	ns
	DMRT	4.0>C	ns	ns

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Table IIAB-3. Electrical conductivity (E_c), fluoride content (F), and pH of groundwater sampled at 120 cm depth from a Florida Spodosol soil cropped to bahiagrass (Continuation).

Sampling date	Treatment	E_c	F	pH
12-07-1992	Mg PG ha^{-1}	umho cm^{-1}	mg L^{-1}	
	0.0(C_{ex})	716.5	1.44	7.1
	0.0(C)	201.5	0.14	5.5
	0.4	382.5	0.27	5.8
	2.0	497.8	0.25	4.9
	4.0	923.8	0.23	3.5
	Statistics:			
	P(w/C_{ex})	0.1788	0.0428	0.1903
	P(w/o C_{ex})	0.0274	0.7133	0.1653
	P(Linear)	0.0080	ns	0.0820
	DMRT	4.0>all	ns	ns

[1]ns=not significant.

Table IIAR-1. Electrical conductivity (E_c), fluoride (F) content, and pH of groundwater sampled at 60-cm depth from a Florida Spodosol soil cropped to annual ryegrass which was amended with PG as source of S and Ca, by sampling, 1991-1993.

Sampling date	Treatment	E_c	F	pH
08-05-1991	Mg PG ha^{-1}	umho cm^{-1}	mg L^{-1}	
	0.0(C)	895	0.16	4.7
	0.4	519	0.17	4.6
	2.0	1045	0.22	4.5
	4.0	1469	0.37	4.3
	Statistics:			
	P	0.1512	0.7597	0.7591
	P(Linear)	0.0589	ns[1]	ns
10-04-1991	Mg PG ha^{-1}	umho cm^{-1}	mg L^{-1}	
	0.0(C)	463	0.14	4.1
	0.4	273	0.13	4.0
	2.0	1046	0.13	4.3
	4.0	1371	0.30	3.9
	Statistics:			
	P	0.1491	0.6951	0.6739
	P(Linear)	0.0483	ns	ns
07-01-1992	Mg PG ha^{-1}	umho cm^{-1}	mg L^{-1}	
	0.0(C)	744	0.30	3.7
	0.4	1218	0.27	3.8
	2.0	1247	0.47	3.9
	4.0	576	0.35	3.8
	Statistics:			
	P	0.4972	0.9051	0.9790
10-02-1992	Mg PG ha^{-1}	umho cm^{-1}	mg L^{-1}	
	0.0(C)	751	0.32	4.3
	0.4	1207	0.35	3.7
	2.0	897	0.29	3.9
	4.0	1132	0.70	3.8
	Statistics:			
	P	0.7236	0.3031	0.5778

[1]ns=not significant.

Table IIAR-2. Electrical conductivity (E_c), fluoride (F) content, and pH of groundwater sampled at 120-cm depth from a Florida Spodosol soil cropped to annual ryegrass which was amended with PG as source of S and Ca, by sampling, 1991-1993.

Sampling date	Treatment	E_c	F	pH
04-23-1991	Mg PG ha^{-1}	umho cm^{-1}	mg L^{-1}	
	0.0(C_{ex})	85	0.00	4.9
	0.0(C)	131	0.01	5.1
	0.4	105	0.01	5.6
	2.0	109	0.02	4.8
	4.0	229	0.02	4.9
	Statistics:			
	P(w/C_{ex})	0.5989	0.5000	0.3123
	P(w/o C_{ex})	0.6411	0.5000	0.2801
04-26-1991	Mg PG ha^{-1}	umho cm^{-1}	mg L^{-1}	
	0.0(C_{ex})	88	0.00	4.9
	0.0(C)	128	0.01	4.9
	0.4	110	0.01	5.1
	2.0	111	0.02	4.7
	4.0	220	0.02	4.7
	Statistics:			
	P(w/C_{ex})	0.5359	0.5000	0.0554
	P(w/o C_{ex})	0.6002	0.5000	0.0672
	P(Linear)	ns[1]	ns	0.0291
08-05-1991	Mg PG ha^{-1}	umho cm^{-1}	mg L^{-1}	
	0.0(C_{ex})	147	0.03	4.3
	0.0(C)	155	0.03	4.9
	0.4	295	0.09	4.3
	2.0	221	0.08	4.7
	4.0	293	0.05	5.1
	Statistics:			
	P(w/C_{ex})	0.5260	0.7957	0.4065
	P(w/o C_{ex})	0.6504	0.7643	0.5253

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Table IIAR-2. Electrical conductivity (E_c), fluoride (F) content, and pH of groundwater sampled at 120-cm depth from a Florida Spodosol soil cropped to annual ryegrass which was amended with PG (Continuation).

Sampling date	Treatment	E_c	F	pH
10-04-1991	Mg PG ha^{-1}	umho cm^{-1}	mg L^{-1}	
	0.0(C_{ex})	138	0.01	4.7
	0.0(C)	151	0.04	4.5
	0.4	173	0.07	4.5
	2.0	237	0.04	4.4
	4.0	385	0.06	4.1
	Statistics:			
	P(w/C_{ex})	0.3118	0.5688	0.8423
	P(w/o C_{ex})	0.3927	0.8096	0.8481
04-01-1992	Mg PG ha^{-1}	umho cm^{-1}	mg L^{-1}	
	0.0(C_{ex})	180	0.06	4.7
	0.0(C)	239	0.09	4.7
	0.4	377	0.15	4.5
	2.0	231	0.09	4.5
	4.0	557	0.18	4.3
	Statistics:			
	P(w/C_{ex})	0.4578	0.3677	0.8515
	P(w/o C_{ex})	0.4716	0.3253	0.7185
07-01-1992	Mg PG ha^{-1}	umho cm^{-1}	mg L^{-1}	
	0.0(C_{ex})	153	0.03	4.5
	0.0(C)	397	0.12	4.3
	0.4	502	0.19	4.1
	2.0	484	0.15	4.3
	4.0	459	0.13	4.3
	Statistics:			
	P(w/C_{ex})	0.3604	0.4123	0.9231
	P(w/o C_{ex})	0.8231	0.5132	0.9556

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Table IIAR-2. Electrical conductivity (E_c), fluoride (F) content, and pH of groundwater sampled at 120-cm depth from a Florida Spodosol soil cropped to annual ryegrass which was amended with PG (Continuation).

Sampling date	Treatment	E_c	F	pH
10-04-1992	Mg PG ha^{-1}	umho cm^{-1}	mg L^{-1}	
	0.0(C_{ex})	-[a]	-[a]	-[a]
	0.0(C)	429	0.08	4.3
	0.4	598	0.11	4.1
	2.0	849	0.11	4.1
	4.0	861	0.13	4.1
	Statistics:			
	P(w/C_{ex})	-[b]	-[b]	-[b]
	P(w/o C_{ex})	0.0124	0.6663	0.9767
	P(Linear)	0.0052	ns	ns
	P(Quad.)	0.0275	ns	ns
	DMRT	4.0,2.0> 0.4>C	ns	ns
12-07-1992	Mg PG ha^{-1}	umho cm^{-1}	mg L^{-1}	
	0.0(C_{ex})	-[a]	-[a]	-[a]
	0.0(C)	229	0.05	4.2
	0.4	478	0.17	4.1
	2.0	1211	0.38	4.0
	4.0	632	0.53	4.3
	Statistics:			
	P(w/C_{ex})	-[b]	-[b]	-[b]
	P(w/o C_{ex})	0.1841	0.5016	0.5182
02-10-1993	Mg PG ha^{-1}	umho cm^{-1}	mg L^{-1}	
	0.0(C_{ex})	165	0.28	4.3
	0.0(C)	420	0.27	4.1
	0.4	575	0.38	4.1
	2.0	761	0.27	4.1
	4.0	493	0.19	4.0
	Statistics:			
	P(w/C_{ex})	0.3819	0.4773	0.9259
	P(w/o C_{ex})	0.6839	0.3620	0.9946

[1]ns=not significant. [a]No water in wells. [b]No error term due to missing data.

Table IIB-1. ^{226}Ra, ^{210}Pb, and ^{210}Po concentrations in a Florida Spodosol cropped to bahiagrass which was amended with PG as a source of S and Ca, by depth, 1990.

Treatment	Depth (cm)					
	0-15	15-30	30-45	45-60	60-75	75-90
Mg PG ha^{-1}	----------	----------	pCi ^{226}Ra g^{-1}	----------	----------	----------
0.0(C)	0.30	0.40	0.40	0.40	0.50	0.35
0.4	0.30	0.25	0.40	0.50	0.50	0.40
2.0	0.30	0.35	0.35	0.30	0.50	0.35
4.0	0.35	0.55	0.20	0.30	0.45	0.20
Statistics:						
P	0.9410	0.6709	0.0275	0.7764	0.9932	0.7881
P(Linear)	ns[1]	ns	0.0078	ns	ns	ns
DMRT	ns	ns	all>4.0	ns	ns	ns
Mg PG ha^{-1}	----------	----------	pCi ^{210}Pb g^{-1}	----------	----------	----------
0.0(C)	0.59	0.61	0.27	0.31	0.40	0.15
0.4	0.71	0.40	0.30	0.50	0.25	0.25
2.0	0.50	0.35	0.45	0.27	0.71	0.47
4.0	1.07	0.30	0.25	0.00	0.35	0.42
Statistics:						
P	0.0310	0.5000	0.9219	0.8516	0.7721	0.3361
P(Linear)	0.0213	ns	ns	ns	ns	ns
P(Quad.)	0.0299	ns	ns	ns	ns	ns
DMRT	4.0>all	ns	ns	ns	ns	ns
Mg PG ha^{-1}	----------	----------	pCi ^{210}Po g^{-1}	----------	----------	----------
0.0(C)	0.62	0.19	0.29	0.38	0.34	0.21
0.4	0.64	0.34	0.54	0.20	0.36	0.20
2.0	0.57	0.25	0.33	0.27	0.32	0.32
4.0	0.52	0.43	0.33	0.52	0.13	0.27
Statistics:						
P	0.9844	0.5902	0.6930	0.6608	0.3266	0.2245

[1]ns=not significant.

Table IIB-2. ^{226}Ra, $^{2\ 10}Pb$, and ^{210}Po concentrations in a Florida Spodosol cropped to bahiagrass which was amended with PG as a source of S and Ca, by depth, 1991.

Treatment	Depth (cm)					
	0-15	15-30	30-45	45-60	60-75	75-90
Mg PG ha^{-1}	---------- pCi ^{226}Ra g^{-1} ----------					
0.0(C)	0.60	0.75	0.75	0.55	0.50	0.70
0.4	0.25	0.30	0.50	0.45	0.85	0.80
2.0	0.65	0.55	0.45	0.55	0.45	0.25
4.0	0.55	0.80	0.40	0.45	0.35	0.65
Statistics:						
P	0.6911	0.4060	0.6672	0.9873	0.1026	0.2188
DMRT	ns[1]	ns	ns	ns	0.4>4.0	ns
Mg PG ha^{-1}	---------- pCi ^{210}Pb g^{-1} ----------					
0.0(C)	0.35	0.40	0.40	0.20	0.50	0.60
0.4	0.45	0.20	0.15	0.45	0.75	0.50
2.0	0.45	0.30	0.30	0.50	0.70	0.00
4.0	0.20	0.10	0.15	0.00	0.20	0.15
Statistics:						
P	0.8705	0.5000	0.6476	0.3564	0.6532	0.4427
Mg PG ha^{-1}	---------- pCi ^{210}Po g^{-1} ----------					
0.0(C)	0.25	0.10	0.00	0.05	0.10	0.10
0.4	0.70	0.70	0.75	0.85	0.25	0.15
2.0	0.40	0.05	0.20	0.20	0.35	0.65
4.0	0.90	0.45	0.00	0.55	0.95	0.80
Statistics:						
P	0.5000	0.5000	0.5000	0.3545	0.6047	0.5492

[1]ns=not significant.

Table IIB-3. ^{226}Ra, ^{210}Pb, and ^{210}Po concentrations in a Florida Spodosol cropped to bahiagrass which was amended with PG as a source of S and Ca, by depth, 1992.

Treatment	Depth (cm)					
	0-15	15-30	30-45	45-60	60-75	75-90
Mg PG ha^{-1}	----------	----------	pCi ^{226}Ra g^{-1}	----------	----------	----------
0.0(C)	0.90	0.85	0.75	0.70	0.95	0.70
0.4	0.75	0.75	0.85	0.80	0.90	0.90
2.0	1.00	1.00	0.50	0.75	0.65	0.45
4.0	0.85	1.05	0.60	0.65	0.65	0.60
Statistics:						
P	0.9797	0.8174	0.3986	0.9592	0.0797	0.5000
P(Linear)	ns[1]	ns	ns	ns	0.0434	ns
DMRT	ns	ns	ns	ns	C>2.0, 4.0	ns
Mg PG ha^{-1}	----------	----------	pCi ^{210}Pb g^{-1}	----------	----------	----------
0.0(C)	0.50	0.10	0.35	0.45	0.45	0.50
0.4	0.60	0.25	0.20	0.30	0.45	0.25
2.0	0.95	0.65	0.40	0.30	0.20	0.55
4.0	0.50	0.50	0.25	0.40	0.30	0.40
Statistics:						
P	0.7458	0.0270	0.9374	0.3505	0.1052	0.5000
P(Linear)	ns	0.0220	ns	ns	0.0667	ns
P(Quad.)	ns	0.0215	ns	ns	ns	ns
DMRT	ns	2.0>C, 0.4;4.0>C	ns	ns	0.4,C>2.0	ns
Mg PG ha^{-1}	----------	----------	pCi ^{210}Po g^{-1}	----------	----------	----------
0.0(C)	0.80	0.30	0.30	0.20	0.20	0.15
0.4	0.20	1.00	1.20	1.10	0.60	0.60
2.0	1.70	1.00	0.80	1.30	0.20	0.35
4.0	0.95	0.50	0.35	0.85	1.30	2.15
Statistics:						
P	0.8175	-[a]	0.5921	-[a]	0.5530	0.0988
P(Linear)	ns	-[a]	ns	-[a]	ns	0.0361
DMRT	ns	-[a]	ns	-[a]	ns	4.0>C,2.0

[1]ns=not significant. [a]No analysis due to no degrees of freedom for error.

Table IIB-4. Gamma radiation (uR hr^{-1}) over a Florida Spodosol soil cropped to bahiagrass which was amended with PG as a source of S and Ca, by sampling period, (month/day), 1990-1992.

Treatment	1990 (Pre-PG)		1990-1991				
	06/11/90 07/16/90	07/16/90 08/24/90	08/24/90 09/24/90	09/24/90 10/30/90	10/30/90 11/27/90	11/27/90 02/13/91	02/20/91 03/29/91
Mg PG ha^{-1}	----------	----------	----------	uR hr^{-1} ----------	----------	----------	----------
0.0(C_{ex1})	4.00	4.76	4.43	4.25	5.15	5.80	5.00
0.0(C_{ex2})	-[a]	5.40	5.20	5.70	5.65	5.50	6.20
0.0(C)	4.30	4.58	4.00	3.85	4.45	4.70	4.45
0.4	-[b]	-[b]	4.30	4.40	-[b]	5.00	4.50
2.0	-[b]	-[b]	4.40	3.95	4.75	4.80	4.85
4.0	-[b]	-[b]	5.20	4.60	3.95	5.60	4.20
Statistics:							
P(w/$C_{ex1,2}$)	-[c]	-[c]	0.1052	0.0236	0.2421	0.0907	0.2759
P(w/o $C_{ex1,2}$)	-[c]	-[c]	0.2020	0.0896	0.5377	0.2971	0.7393
P(Linear)	-[c]	-[c]	0.0778	0.0364	ns	ns	ns
DMRT	-[c]	-[c]	ns[1]	4.0>C	ns	ns	ns

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Table IIB-4. Gamma radiation, by sampling (Continuation).

Treatment	1991-1992				1992			
	04/09/91 05/13/91	05/13/91 07/16/91	10/07/91 01/02/92	01/08/92 03/16/92	03/17/92 05/15/92	05/15/92 07/13/92	07/14/92 09/18/92	09/21/92 11/23/92
Mg PG ha^{-1}	---------- uR hr^{-1} ----------							
0.0(C_{ex1})	4.30	4.67	5.80	5.73	5.95	5.78	5.10	5.63
0.0(C_{ex2})	6.10	4.85	6.50	6.10	5.75	6.60	6.35	7.45
0.0(C)	4.40	4.10	5.20	6.40	5.80	5.65	5.35	5.10
0.4	4.50	4.00	6.00	5.45	5.55	5.50	5.15	5.60
2.0	4.05	4.30	6.25	6.35	5.20	5.65	5.10	5.45
4.0	4.55	4.75	5.45	5.55	4.35	5.65	5.85	5.05
Statistics:								
P($w/C_{ex1,2}$)	0.1422	0.5038	0.3332	0.3358	0.0054	0.6652	0.5332	0.1415
P($w/o\ C_{ex1,2}$)	0.4310	0.2364	0.0051	0.3061	0.0072	0.98818	0.8261	0.3503
P(Linear)	ns	ns	0.0877	ns	0.0022	ns	ns	ns
P(Quad.)	ns	ns	0.0018	ns	C>2.0;	ns	ns	ns
DMRT	ns	ns	0.4,2.0> 4.0,C	ns	all>4.0			

[1]ns=not significant. [a]No external control #2 at this time; [b]No electret readings. [c]No analysis due to missing data.

Table IIB-5. ^{226}Ra, ^{210}Pb, and ^{210}Po concentrations in bahiagrass regrowth forage from a pasture amended with PG as a source of S and Ca and growing on a Florida Spodosol soil, by harvest (month/day), 1990-91.

Treatment	Date of harvest		
	10/01	11/13	01/07
Mg PG ha^{-1}	---------	pCi ^{226}Ra g^{-1}	----------
0.0(C)	0.01	0.01	0.01
0.4	0.01	0.04	0.01
2.0	0.01	0.01	0.03
4.0	0.01	0.05	0.04
Statistics:			
P	0.5094	0.3971	0.2112
P(Linear)	ns[1]	ns	0.0672
Mg PG ha^{-1}	--------	pCi ^{210}Pb g^{-1}	----------
0.0(C)	0.34	0.14	0.13
0.4	0.14	0.09	0.17
2.0	0.20	0.14	0.07
4.0	0.15	0.21	0.03
Statistics:			
P	0.6890	0.7844	0.5262
Mg PG ha^{-1}	---------	pCi ^{210}Po g^{-1}	----------
0.0(C)	0.08	0.01	0.01
0.4	0.05	0.20	0.05
2.0	0.28	0.24	0.07
4.0	0.39	0.07	0.22
Statistics:			
P	0.5146	0.0418	0.0750
P(Linear)	ns	ns	0.0233
P(Quad.)	ns	0.0155	ns
DMRT	ns	2.0>4.0, C;0.4>C	4.0>0.4, C

[1]ns=not significant.

Table IIB-6. ^{226}Ra, ^{210}Pb, and ^{210}Po concentrations in bahiagrass regrowth forage from a pasture amended with PG as a source of S and Ca and growing on a Florida Spodosol soil,by harvest (month/day), 1991.

Treatment	Date of harvest		
	05/06	06/24	08/08
Mg PG ha^{-1}	----------	pCi ^{226}Ra g^{-1}	----------
0.0(C)	-[a]	0.03	0.05
0.4	0.10	0.05	0.04
2.0	-[a]	0.02	0.09
4.0	0.19	0.08	0.11
Statistics:			
P	-[b]	0.3707	0.0327
P(Linear)	-[b]	ns[1]	0.0225
DMRT	-[b]	ns	4.0>C, 0.4; 2.0>0.4
Mg PG ha^{-1}	----------	pCi ^{210}Pb g^{-1}	----------
0.0(C)	0.40	0.53	0.57
0.4	0.73	0.73	0.73
2.0	0.30	0.93	0.63
4.0	0.45	0.47	0.45
Statistics:			
P	0.0366	0.0414	0.3034
P(Quad.)	ns	0.0117	ns
DMRT	0.4>all	2.0>C,4.0	ns
Mg PG ha^{-1}	----------	pCi ^{210}Po g^{-1}	----------
0.0(C)	0.31	0.64	0.62
0.4	0.59	0.84	0.76
2.0	0.26	0.47	0.82
4.0	0.18	0.47	0.44
Statistics:			
P	0.1857	0.3689	0.4509

[1]ns=not significant. [a]Not detected in duplicate samples. [b]No analysis due to missing data.

Table IIB-7. ^{226}Ra, ^{210}Pb, and ^{210}Po concentrations in bahiagrass regrowth forage from a pasture amended with PG as a source of S and Ca and growing on a Florida Spodosol soil, by harvest (month/day), 1992.

Treatment	Date of harvest		
	04/30	07/10	09/22
Mg PG ha^{-1}	----------	pCi ^{226}Ra g^{-1}	----------
0.0(C)	0.10	0.15	-[a]
0.4	0.30	0.10	0.10
2.0	0.15	0.10	-[a]
4.0	0.20	0.25	0.15
Statistics:			
P	0.1815	0.7858	-[b]
DMRT	0.4>C	ns[1]	-[b]
Mg PG ha^{-1}	----------	pCi ^{210}Pb g^{-1}	----------
0.0(C)	1.75	3.65	2.20
0.4	2.25	2.25	1.90
2.0	1.80	1.90	2.60
4.0	2.40	1.75	2.50
Statistics:			
P	0.6771	0.1053	0.7846
P(Linear)	ns	0.0389	ns
DMRT	ns	C>2.0,4.0	ns
Mg PG ha^{-1}	----------	pCi ^{210}Po g^{-1}	----------
0.0(C)	-[a]	0.10	0.10
0.4	-[a]	0.10	0.10
2.0	0.10	0.10	0.10
4.0	0.10	0.20	0.20
Statistics:			
P	-[b]	-[c]	-[c]

[1]ns=not significant. [a]Not detected in duplicate samples. [b]No analysis due to missing data. [c]No degrees of freedom for error.

Table IIB-8. ^{226}Ra, ^{210}Pb, and ^{210}Po concentrations in runoff from a Florida Spodosol soil cropped to bahiagrass which was amended with PG as a source of S and Ca, by sampling, 1990-1992.

Sampling date	Treatment	^{226}Ra	^{210}Pb	^{210}Po
10-04-1990	Mg PG ha^{-1}	--------	pCi L^{-1}	---------
	0.0(C)	0.11	0.05	0.37
	0.4	0.13	0.17	0.25
	2.0	0.07	0.00	0.29
	4.0	0.42	0.47	0.41
	Statistics:			
	P	0.0329	0.2455	0.8963
	P(Linear)	0.0165	ns[1]	ns
	P(Quad.)	0.0359	ns	ns
	DMRT	4.0>all	ns	ns
06-19-1991	Mg PG ha^{-1}	---------	pCi L^{-1}	---------
	0.0(C)	0.20	1.00	0.40
	0.4	0.35	1.10	1.30
	2.0	0.40	0.60	0.60
	4.0	0.35	0.60	0.40
	Statistics:			
	P	0.0001[a]	0.3579	0.4925
	DMRT	2.0>all; 0.4,4.0>C	ns	ns
07-01-1992	Mg PG ha^{-1}	---------	pCi L^{-1}	---------
	0.0(C)	0.15	0.20	0.60
	0.4	0.20	1.25	0.30
	2.0	0.10	0.60	0.25
	4.0	0.70	0.10	0.15
	Statistics:			
	P	0.1395	0.1441	0.5030
10-01-1992	Mg PG ha^{-1}	---------	pCi L^{-1}	---------
	0.0(C)	0.40	1.10	0.40
	0.4	0.80	0.95	0.10
	2.0	0.25	-[b]	0.20
	4.0	0.35	0.30	0.50
	Statistics:			
	P	0.2188	-[c]	-[d]

[1]ns=not significant. [a]Difference due to estimated error term being so small. [b]Not detected in duplicate samples. [c]No analysis due to missing data. [d]No degrees of freedom for error.

Table IIB-9. ^{226}Ra, ^{210}Pb, and ^{210}Po concentrations in groundwater sampled at 60 cm depth from a Florida Spodosol soil cropped to bahiagrass which was amended with PG as a source of S and Ca, by sampling, 1991[a].

Sampling date	Treatment	^{226}Ra	^{210}Pb	^{210}Po
07-01-1991	Mg PG ha^{-1}	---------	pCi L^{-1}	---------
	0.0(C_{ex})	0.30	0.10	0.30
	0.0(C)	0.40	0.45	0.35
	0.4	0.60	1.05	0.25
	2.0	0.55	0.80	0.35
	4.0	0.70	2.50	0.40
	Statistics:			
	P(w/C_{ex})	0.4117	0.0308	0.5201
	P(w/o C_{ex})	0.4642	0.0261	0.5000
	P(Linear)	ns[1]	0.0083	ns
	DMRT	ns	4.0>all	ns

[1]ns=not significant. [a]Not originally included in the sampling program for radionuclides but a 1991 sample was run to see the range of nuclide values in water samples at this depth.

Table IIB-10. ^{226}Ra, ^{210}Pb, and ^{210}Po concentrations in groundwater sampled at 120 cm depth from a Florida Spodosol soil cropped to bahiagrass which was amended with PG as a source of S and Ca, by sampling, 1990-1992.

Sampling date	Treatment	^{226}Ra	^{210}Pb	^{210}Po
10-04-1990	Mg PG ha^{-1}	---------	pCi L^{-1}	---------
	0.0(C)	0.83	0.48	0.61
	0.4	1.39	0.67	0.29
	2.0	0.96	0.17	0.25
	4.0	1.14	0.51	0.75
	Statistics:			
	P	0.4160	0.1760	0.2740
04-26-1991	Mg PG ha^{-1}	---------	pCi L^{-1}	---------
	0.0(C_{ex})	0.70	0.00	0.75
	0.0(C)	0.55	0.15	1.65
	0.4	1.30	0.05	0.90
	2.0	0.90	0.50	0.90
	4.0	1.10	0.00	1.80
	Statistics:			
	P(w/C_{ex})	0.9207	0.0217	0.0519
	P(w/o C_{ex})	0.8810	0.0346	0.1606
	P(Quad.)	ns[1]	0.0126	0.0647
	DMRT	ns	2.0>all	ns
06-19-1991	Mg PG ha^{-1}	---------	pCi L^{-1}	---------
	0.0(C_{ex})	0.55	0.65	0.90
	0.0(C)	0.55	0.70	2.85
	0.4	1.10	1.40	1.10
	2.0	1.50	2.40	1.15
	4.0	1.10	1.05	1.50
	Statistics:			
	P(w/C_{ex})	0.1607	0.1515	0.5628
	P(w/o C_{ex})	0.2721	0.1640	0.5747
	P(Quad.)	ns	0.0497	ns

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Table IIB-10. ^{226}Ra, ^{210}Pb, and ^{210}Po in groundwater sampled at 120 cm depth (Continuation).

Sampling date	Treatment	^{226}Ra	^{210}Pb	^{210}Po
04-01-1992	Mg PG ha^{-1}	---------	pCi L^{-1}	---------
	0.0(C_{ex})	0.75	1.09	0.25
	0.0(C)	1.17	0.44	0.18
	0.4	1.61	0.32	0.48
	2.0	1.69	0.85	0.73
	4.0	1.12	0.99	0.53
	Statistics:			
	P(w/C_{ex})	0.0696	0.6249	0.3097
	P(w/o C_{ex})	0.2488	0.3665	0.3123
07-01-1992	Mg PG ha^{-1}	---------	pCi L^{-1}	---------
	0.0(C_{ex})	0.35	-[a]	0.45
	0.0(C)	0.75	0.25	0.10
	0.4	0.95	0.80	0.10
	2.0	1.10	1.60	0.55
	4.0	1.70	1.35	0.30
	Statistics:			
	P(w/C_{ex})	0.4598	0.2417	0.0001[b]
	P(w/o C_{ex})	0.6052	0.2417	-[c]
10-01-1992	Mg PG ha^{-1}	---------	pCi L^{-1}	---------
	0.0(C_{ex})	0.80	0.30	-[a]
	0.0(C)	0.70	0.65	2.85
	0.4	1.00	0.30	0.20
	2.0	0.65	0.65	-[a]
	4.0	1.80	0.70	0.65
	Statistics:			
	P(w/C_{ex})	0.1638	0.9040	0.5962
	P(w/o C_{ex})	0.1878	0.8721	0.5962
	P(Linear)	0.0815	ns	ns

[1]ns=not significant. [a]Not detected in duplicate samples. [b]Difference due to estimated error term being so small. [c]No degrees of freedom for error.

Table IIB-11. Soil surface Rn flux over a Florida Spodosol soil cropped to bahiagrass which was amended with PG as a source of S and Ca, by sampling, (month/day), 1990-1992.

Treatment	1990-91		1991-92			1992	
	10/30	02/19	04/10	09/18	01/15	07/29	12/03
Mg PG ha^{-1}	---------- pCi m^{-2} s^{-1} ----------						
0.0(C)	0.03	0.04	0.03	0.01	0.05	0.01	0.04
0.4	0.03	0.04	0.02	0.02	0.05	0.02	0.05
2.0	0.04	0.04	0.04	0.02	0.08	0.02	0.05
4.0	0.04	0.03	0.03	0.03	0.06	0.02	0.05
Statistics:							
P	0.3179	0.3984	0.4671	0.7072	0.5018	0.5772	0.3073

Table IIB-12. Ambient atmospheric Rn over a Florida Spodosol soil cropped to bahiagrass which was amended with PG as a source of S and Ca, by sampling, (month/day), 1990-1992.

Treatment	Pre-application 1990		1990-1991				
	06/11-07/16	07/16-08/24	08/24-09/24	09/24-10/30	10/30-11/27	11/27-02/13	02/20-03/29
Mg PG ha^{-1}	----------	----------	----------	pCi L^{-1}	----------	----------	----------
0.0(C_{ex1})	0.23	0.12	0.16	0.21	0.21	0.16	0.20
0.0(C_{ex2})	-[a]	0.12	0.11	0.18	0.24	0.29	0.15
0.0(C)	0.22	0.13	0.14	0.20	0.29	0.24	0.24
0.4	-[b]	-[b]	0.14	0.21	0.29	0.29	0.40
2.0	-[b]	-[b]	0.12	0.19	0.24	0.25	0.18
4.0	-[b]	-[b]	0.14	0.19	0.29	0.25	0.15
Statistics:[1]							
P(w/$C_{ex1,2}$)	0.9682	0.7802	0.4764	0.8450	0.2297	0.0112	0.0158
P(w/o $C_{ex1,2}$)	-[c]	-[c]	0.0382	0.8418	0.3603	0.0676	0.0241
P(Linear)	-[c]	-[c]	ns[1]	ns	ns	ns	0.0154
P(Quad.)	-[c]	-[c]	0.0116	ns	ns	ns	ns
DMRT	-[c]	-[c]	all>2.0	ns	ns	0.4>all	0.4>all

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Table IIB-12. Ambient atmospheric Rn (Continuation).

Treatment	1991-1992				1992			
	04/09-05/13	05/13-07/16-	10/07-01/02	01/08-03/16	03/17-05/15	05/15-07/13	07/14-09/18	09/21-11/23
Mg PG ha^{-1}	pCi L^{-1}							
0.0(C_{ex1})	0.20	0.05	0.31	0.32	0.23	0.19	0.09	0.20
0.0(C_{ex2})	0.04	0.06	0.22	0.37	0.21	0.12	0.06	0.21
0.0(C)	0.12	0.09	0.34	0.43	0.25	0.18	0.13	0.21
0.4	0.20	0.13	0.34	0.31	0.21	0.20	0.10	0.21
2.0	0.14	0.16	0.29	0.41	0.22	0.29	0.12	0.26
4.0	0.15	0.06	0.27	0.29	0.24	0.22	0.13	0.28
Statistics:								
P(w/$C_{ex1,2}$)	0.0397	0.0276	0.8062	0.8015	0.9771	0.1464	0.4329	0.9389
P(W/o $C_{ex1,2}$)	0.0051	0.0905	0.2190	0.5002	0.9160	0.2174	0.3677	0.8620
P(Linear)	0.0459	ns[1]	ns	ns	ns	ns	ns	ns
P(Quad.)	ns	0.0327	ns	ns	ns	ns	ns	ns
DMRT	0.4>4.0, 4.0>C	2.0>4.0	ns	ns	ns	ns	ns	

[1]ns=not significant. [a]No external control at this time. [b]Not detected in duplicate samples. [c]No analysis due to missing data.

Table IIC-1. ^{226}Ra, ^{210}Pb, and ^{210}Po concentrations in a Florida Spodosol soil cropped to annual ryegrass which was amended with PG as source of S and Ca, by depth, 1991.

Treatment	Depth (cm)					
	0-15	15-30	30-45	45-60	60-75	75-90
Mg PG ha^{-1}	pCi ^{226}Ra g^{-1}					
0.0(C)	0.80	0.80	1.15	1.95	1.20	1.05
0.4	0.70	0.65	0.85	0.55	0.40	0.85
2.0	0.20	0.35	0.45	0.70	0.75	0.55
4.0	0.45	0.45	0.45	0.40	0.60	0.60
Statistics:						
P	0.6369	0.5000	0.6615	0.1904	0.5046	0.8007
P(Linear)	ns[1]	ns	ns	0.0735	ns	ns
Mg PG ha^{-1}	pCi ^{210}Pb g^{-1}					
0.0(C)	0.80	0.35	0.60	1.00	0.30	0.95
0.4	0.80	1.15	1.40	0.65	1.90	1.00
2.0	0.80	1.70	0.75	0.20	0.80	0.20
4.0	0.75	0.45	0.25	0.70	0.60	0.45
Statistics:						
P	0.6805	0.4847	0.1965	0.7311	0.1738	0.5412
Mg PG ha^{-1}	pCi ^{210}Po g^{-1}					
0.0(C)	0.50	0.55	0.35	0.20	0.15	0.30
0.4	0.35	-[a]	-[a]	0.15	0.10	0.30
2.0	0.20	0.50	0.30	0.10	0.25	0.10
4.0	0.40	0.30	0.20	0.10	0.45	0.35
Statistics:						
P	0.9981	-[b]	0.6944	0.4557	0.4827	0.9591

[1]ns=not significant. [a]Not detected in samples. [b]No error terms for appropriate analysis due to missing data.

Table IIC-2. ^{226}Ra, ^{210}Pb, and ^{210}Po concentrations in a Florida Spodosol soil cropped to annual ryegrass which was amended with PG as source of S and Ca, by depth, 1992.

Treatment	Depth (cm)					
	0-15	15-30	30-45	45-60	60-75	75-90
Mg PG ha^{-1}	pCi ^{226}Ra g^{-1}					
0.0(C)	0.25	0.25	0.30	0.20	0.20	0.25
0.4	0.25	0.25	0.25	0.25	0.20	0.20
2.0	0.25	0.40	0.35	0.25	0.20	0.20
4.0	0.30	0.25	0.15	0.20	0.20	0.25
Statistics:						
P	0.9480	0.0520	0.4181	0.8904	1.0000	0.9480
Mg PG ha^{-1}	pCi ^{210}Pb g^{-1}					
0.0(C)	0.50	0.50	1.60	1.00	0.70	0.20
0.4	0.65	0.70	0.80	0.75	0.60	0.65
2.0	0.95	0.40	0.25	0.40	0.50	0.50
4.0	0.30	0.45	-[a]	-[a]	2.30	0.40
Statistics:						
P	0.8982	-[b]	0.0525	0.0875	0.0803	-[a]
P(Linear)	ns[1]	-[b]	ns	ns	0.0561	-[a]
Mg PG ha^{-1}	pCi ^{210}Po g^{-1}					
0.0(C)	0.30	0.25	0.50	0.20	0.25	0.25
0.4	0.50	0.35	0.25	0.95	0.50	0.30
2.0	1.20	0.10	0.30	0.20	-[a]	-[a]
4.0	0.20	0.20	0.10	-[a]	0.20	0.20
Statistics:						
P	0.0279	0.6221	-[b]	-[b]	0.9083	0.9186
DMRT	2.0>all	ns	-[b]	-[b]	ns	ns

[1]ns=not significant. [a]Not detected in Samples. [b]No replicated observations.

Table IIC-3. ^{226}Ra, ^{210}Pb, and ^{210}Po concentrations in a Florida Spodosol soil cropped to annual ryegrass which was amended with PG as source of S and Ca, by depth, 1993.

Treatment	Depth (cm)					
	0-15	15-30	30-45	45-60	60-75	75-90
Mg PG ha^{-1}	---------- pCi ^{226}Ra g^{-1} ----------					
0.0(C)	0.25	0.35	0.35	0.30	0.30	0.15
0.4	0.35	0.60	0.55	0.50	0.45	0.70
2.0	0.30	0.50	0.30	0.35	0.60	0.90
4.0	0.35	0.45	0.40	0.50	0.50	0.45
Statistics:						
P	0.9419	0.7365	0.5615	0.6205	0.4331	0.4105
Mg PG ha^{-1}	---------- pCi ^{210}Pb g^{-1} ----------					
0.0(C)	0.85	0.40	0.45	0.25	0.30	0.35
0.4	0.85	0.40	0.10	0.50	0.55	0.40
2.0	0.80	0.50	0.40	1.00	0.65	0.90
4.0	0.70	0.35	0.50	0.35	0.55	0.40
Statistics:						
P	0.8345	0.7562	0.3657	0.3844	0.0001[a]	0.7979
Mg PG ha^{-1}	---------- pCi ^{210}Po g^{-1} ----------					
0.0(C)	0.35	0.10	0.15	0.10	0.45	0.35
0.4	0.25	0.20	0.20	0.15	0.30	0.20
2.0	0.65	0.20	0.15	0.20	0.40	0.20
4.0	0.75	0.30	0.20	0.30	0.30	0.25
Statistics:						
P	0.3578	0.0001[a]	0.9423	0.3837	0.1224	0.6897

[a]Due to estimated mean square error (MSE) being zero.

Table IIC-4. Gamma radiation (uR/hr) over a Florida Spodosol soil cropped to annual ryegrass which was amended with PG as source of S and Ca, by sampling period (month/day), 1990-1993.

Treatment	1990-1991				
	12/10/90 02/06/91	02/06/91 03/13/91	03/13/91 04/12/91	04/12/91 05/16/91	05/16/91 07/18/91
Mg PG ha^{-1}	---------- uR hr^{-1} ----------				
0.0(C_{ex1})	5.40	4.65	4.45	5.20	4.10
0.0(C_{ex2})	-[a]	-[a]	-[a]	-[a]	-[a]
0.0(C)	5.10	6.00	4.90	4.45	4.70
0.4	-[b]	4.55	5.00	4.70	4.90
2.0	-[b]	5.40	4.85	5.65	4.65
4.0	5.10	4.75	4.85	4.90	4.80
Statistics:					
P(w/C_{ex1}	-[c]	0.0558	0.4782	0.5989	0.6940
P(w/o C_{ex1})	-[c]	0.1276	0.9602	0.6053	0.2876

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Table IIC-4. Gamma radiation, by sampling (Continuation).

Treatment	1991-92					1992-93		
	10/07/91 01/07/92	01/08/92 03/17/92	03/18/92 05/18/92	05/18/92 07/13/92	07/14/92 09/22/92	09/24/92 11/23/92	11/25/92 02/08/93	02/10/93 04/30/93
Mg PG ha^{-1}	---------- uR hr^{-1} ----------							
0.0(C_{ex1})	6.45	5.93	6.28	5.95	4.38	4.95	5.43	6.37
0.0(C_{ex2})	-[a]	6.10	5.75	6.60	6.35	7.05	6.95	6.05
0.0(C)	6.05	6.15	5.75	5.65	5.10	5.55	5.90	5.40
0.4	5.20	5.65	6.75	5.25	4.80	5.05	7.00	5.45
2.0	6.00	6.05	5.80	5.30	-[b]	4.50	7.20	5.55
4.0	5.35	6.35	5.30	5.10	5.15	5.15	4.85	5.85
Statistics:								
P(w/$C_{ex1,2}$)	0.0305	0.9572	0.7486	0.0095	0.3075	0.2105	0.3919	0.06414
P(w/o $C_{ex1,2}$)	0.1181	0.8863	0.4765	0.1891	0.3294	0.6916	0.4043	0.4924
P(Linear)	ns[1]	ns	ns	0.0605	ns	ns	ns	ns

[1]ns=not significant. [a]No external control #2 at this time. [b]No electret reading obtained. [c]No source of error.

Table IIC-5. ^{226}Ra, ^{210}Pb, and ^{210}Po concentrations in annual ryegrass regrowth forage from a pasture amended with PG as source of S and Ca and growing on a Florida Spodosol soil, by harvest (month/day), 1990-91 to 1992-93.

Treatment	1990-91		1991-92		1992-93		
	02/11	03/21	01/17	02/21	03/18	01/13	02/18
Mg PG ha^{-1}	pCi ^{226}Ra g^{-1}						
0.0(C)	0.09	0.11	0.14	0.14	0.19	-[a]	0.10
0.4	0.11	0.09	0.10	0.16	0.22	0.20	0.10
2.0	0.16	0.15	0.20	0.19	0.31	0.20	0.20
4.0	0.15	0.14	0.17	0.28	0.24	0.55	0.25
Statistics:							
P	0.3352	0.5708	0.0632	0.1717	0.7139	-[b]	0.8126
P(Linear)	ns[1]	ns	ns	0.0594	ns	ns	ns
Mg PG ha^{-1}	pCi ^{210}Pb g^{-1}						
0.0(C)	0.01	0.13	0.24	0.40	0.56	0.25	0.30
0.4	0.00	0.11	0.22	0.33	0.41	0.20	0.35
2.0	0.07	0.39	0.15	0.35	0.43	0.20	0.30
4.0	0.03	0.33	0.09	0.36	0.33	0.40	0.45
Statistics:							
P	0.4544	0.7595	0.7949	0.4572	0.7156	0.1462	0.7477
Mg PG ha^{-1}	pCi ^{210}Po g^{-1}						
0.0(C)	0.04	0.06	-[a]	0.19	-[a]	-[a]	0.50
0.4	0.18	0.09	-[a]	0.12	0.09	0.10	0.40
2.0	0.08	0.04	-[a]	-[a]	0.09	0.20	0.30
4.0	0.06	0.12	-[a]	0.11	0.04	0.30	0.20
Statistics:							
P	0.4417	0.8049	-[b]	-[b]	-[b]	-[b]	0.8412

[1]ns=not significant. [a]Not detected in duplicate samples. [b]No source of error for appropriate test due to missing data.

Table IIC-6. ^{226}Ra, ^{210}Pb and ^{210}Po concentrations in groundwater sampled at 120-cm depth from a Florida Spodosol soil cropped to annual ryegrass which was amended with PG as source of S and Ca, by sampling, 1990-91 to 1992-93.

Sampling date	Treatment	^{226}Ra	^{210}Pb	^{210}Po
04-23-1991	Mg PG ha^{-1}	---------	pCi L^{-1}	----------
	0.0(C)	0.15	0.70	0.80
	0.4	0.40	0.65	0.95
	2.0	0.30	1.05	0.50
	4.0	1.35	1.15	0.90
	Statistics:			
	P	0.0932	0.0557	0.8888
	P(Linear)	0.0346	0.0176	ns[1]
08-05-1991	Mg PG ha^{-1}	---------	pCi L^{-1}	----------
	0.0(C_{ex})	0.90	0.45	0.80
	0.0(C)	0.50	0.05	0.45
	0.4	1.30	0.45	0.60
	2.0	0.75	0.20	1.50
	4.0	1.05	0.50	0.25
	Statistics:			
	P(w/C_{ex})	0.6829	0.7103	0.4544
	P(w/o C_{ex})	0.6124	0.6817	0.3560
04-01-1992	Mg PG ha^{-1}	---------	pCi L^{-1}	----------
	0.0(C_{ex})	0.42	1.03	0.04
	0.0(C)	0.90	0.58	0.29
	0.4	1.69	2.50	0.49
	2.0	1.04	0.40	0.14
	4.0	1.33	0.12	-[b]
	Statistics:			
	P(w/C_{ex})	0.7899	0.0041	0.8140
	P(w/o C_{ex})	0.6931	ns	0.6801
	P(Quad.)	ns	ns	ns

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Table IIC-6. ^{226}Ra, ^{210}Pb and ^{210}Po concentrations in groundwater sampled at 120-cm depth (Continuation).

Sampling date	Treatment	^{226}Ra	^{210}Pb	^{210}Po
07-01-1992	Mg PG ha^{-1}	---------	pCi L^{-1}	----------
	0.0(C_{ex})	0.50	0.65	0.25
	0.0(C)	1.55	0.15	0.40
	0.4	3.35	2.50	1.80
	2.0	3.00	1.15	1.65
	4.0	0.85	0.70	0.60
	Statistics:			
	P(w/C_{ex})	0.2597	0.0167	0.1322
	P(w/o C_{ex})	0.2064	0.0482	0.3570
	P(Quad.)	ns	0.0665	ns
	DMRT	ns	0.4>all	ns
02-10-1993	Mg PG ha^{-1}	---------	pCi L^{-1}	----------
	0.0(C_{ex})	0.20	0.55	0.65
	0.0(C)	1.15	0.95	0.50
	0.4	2.35	2.15	0.40
	2.0	1.65	2.45	0.45
	4.0	1.15	2.10	0.40
	Statistics:			
	P(w/C_{ex})	0.4432	0.3484	0.7442
	P(w/o C_{ex})	0.7075	0.5473	0.8801

[1]ns=not significant.

Table IIC-7. ^{222}Rn soil surface flux over a Florida Spodosol soil cropped to annual ryegrass ryegrass which was amended with PG as source of S and Ca, by sampling period (month/day), 1990-91 to 1992-1993.

Treatment	90-91		1991-92		1992-93	
	11/14/90 11/15/90	03/12/91 03/13/91	11/18/91 11/19/91	06/22/92 06/23/92	12/07/92 12/08/92	05/12/93 05/13/93
Mg PG ha^{-1}	----------	----------	pCi $m^{-2}s^{-1}$	----------	----------	----------
0.0(C)	0.015	0.025	0.025	0.015	0.030	0.020
0.4	0.025	0.022	0.017	0.023	0.030	0.025
2.0	0.023	0.019	0.023	0.020	0.035	0.033
4.0	0.029	0.031	0.028	0.040	0.033	0.033
Statistics:						
P	0.4924	0.1059	0.5253	0.0045	0.9345	0.1955
P(Linear)	ns[1]	ns	ns	0.0013	ns	ns
P(Quad.)	ns	0.0467	ns	0.0341	ns	ns
DMRT	ns	ns	ns	4.0>all; 0.4>C	ns	ns

[1]ns=not significant.

Table IIC-8. Ambient atmospheric ^{222}Rn over a Florida Spodosol soil cropped to annual ryegrass which was amended with PG as source of S and Ca, by sampling period (month/day), 1990-91 to 1992-93.

Treatment	1990-91				
	12/10/90 02/06/91	02/06/91 03/13/91	03/13/91 04/12/91	04/12/91 05/16/91	05/16/91 07/18/91
Mg PG ha^{-1}	----------	----------	pCi L^{-1}	----------	----------
0.0(C_{ex1})	-[a]	0.23	0.12	0.25	0.19
0.0(C_{ex2})	_[b]	_[b]	_[b]	_[b]	_[b]
0.0(C)	0.26	0.23	0.21	0.11	0.18
0.4	0.31	0.15	0.13	0.15	0.16
2.0	0.31	0.16	0.11	0.12	0.20
4.0	0.19	0.14	0.08	0.09	0.17
Statistics:					
P(w/C_{ex1})	-	0.2503	0.2163	0.8195	0.8678
P(w/o C_{ex1})	0.3663	0.3855	0.2619	0.2610	0.4920

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Table IIC-8. Ambient atmospheric ^{222}Rn over a Florida Spodosol soil11 cropped to annual ryegrass (Continuation).

Treatment	1991-92					1992-93		
	10/07/91 01/07/92	01/08/92 03/17/92	03/18/92 05/18/92	05/18/92 07/13/92	07/14/92 09/22/92	09/24/92 11/23/92	11/25/92 02/08/93	02/10/93 04/30/93
Mg PG ha^{-1}	---------- pCi L^{-1} ----------							
0.0(C_{ex1})	0.17	0.23	0.25	0.29	0.13	0.18	0.20	0.19
0.0(C_{ex2})	-[b]	0.37	0.21	0.28	0.05	0.21	0.24	0.19
0.0(C)	0.25	0.29	0.28	0.32	0.15	0.31	0.13	0.17
0.4	0.21	0.29	0.21	0.19	0.18	0.37	0.22	0.19
2.0	0.21	0.25	0.21	0.31	0.13	0.29	0.22	0.09
4.0	0.16	0.13	0.17	0.28	0.08	0.30	0.28	0.17
Statistics:								
P(w/$C_{ex1,2}$)	0.3877	0.4346	0.8808	0.1725	0.4116	0.1760	0.4977	0.4134
P(w/o $C_{ex1,2}$)	0.3973	0.4497	0.8127	0.1280	0.2626	0.5549	0.4019	0.7608

[a]No electret reading obtained. [b]No external control #2 at this time.