DP-1585

DEPOSITION OF AIR-BORNE ^{238}Pu NEAR A CHEMICAL SEPARATION FACILITY

J. H. HORTON AND D. D. GAY

E. I. du Pont de Nemours & Co.
Savannah River Laboratory
Aiken, SC 29808

PREPARED FOR THE U. S. DEPARTMENT OF ENERGY UNDER CONTRACT DE-AC09-76SR00001

Printed in the United States of America

Available from

National Technical Information Service
U. S. Department of Commerce
5285 Port Royal Road
Springfield, Virginia 22161

Price: Printed Copy A02; Microfiche A01

726635

DP-1585
UC-11

DEPOSITION OF AIR-BORNE ^{238}Pu NEAR A CHEMICAL SEPARATION FACILITY

J. H. HORTON* AND D. D. GAY

* Deceased

Approved by

T. V. Crawford, Research Manager
Environmental Transport Division

January 1982

E. I. du Pont de Nemours & Co.
Savannah River Laboratory
Aiken, SC 29808

PREPARED FOR THE U. S. DEPARTMENT OF ENERGY UNDER CONTRACT DE-AC09-76SR00001

ABSTRACT

Three methods were compared to measure deposition of ^{238}Pu released from a chemical separation facility at the Savannah River Plant, Aiken, SC. The methods were (1) adhesive paper, (2) a collector of rain and dryfall, and (3) soil samples. Excellent agreement among the three methods was found. The measured deposition for the particular source term and meteorological conditions at the Savannah River Plant is described by $y \propto x^{-1.36}$ where y is the pCi of ^{238}Pu deposited per square meter per mCi ^{238}Pu released, and x is distance in meters from the source.

CONTENTS

DEPOSITION OF AIR-BORNE ^{238}Pu NEAR A CHEMICAL SEPARATION FACILITY

INTRODUCTION

Much attention is being devoted to dispersion and deposition of pollutants released to the atmosphere from stacks. If the released materials are gases, they can be removed from the air and deposited by contact of the stack plume with the earth surface or by rain. If the released materials are particles, they are deposited by the effects of earth contact, rain, and gravitational forces.

Two nuclear fuel separation facilities at the Savannah River Plant (SRP) release minute quantities of $^{238,239,240}Pu$ to the atmosphere. The Pu is nearly always attached to nonradioactive particles, usually crustal materials (Sanders[1,2]). The measured diameter of 71 Pu-bearing particles in stack air ranged from 0.2 to 29 μm. The geometric mean diameter was 5.43 μm. Most of the particles contained <1 fCi of $^{239,240}Pu$, as measured by fission track counting.

The purpose of this report is to compare the deposition patterns of stack-released Pu-238 as measured by three methods:

(1) Collection of air-borne particles on adhesive paper

(2) Collection of rain and dryfall in a pan

(3) Collection of soil samples

METHODS

Stack releases from the chemical separation facilities, F Area and H Area (Figure 1), are routinely measured about 2 m from the top of 62 m stacks. Particles are removed from the sample air stream by Gelman Type AE filter paper. The filters are 63.5 mm in diameter and are changed daily.

Rain is routinely collected at 31 air monitoring stations (Figure 1). The collectors are large, open stainless steel pans with sloping bottoms. No provision is made for enclosure, so these collectors contain deposited material from dry fall as well as rainfall. The samples are composited into the following groups for

analysis: F Area, H Area, on-plant (Dunbarton, Par Pond, and Williston gate), plant perimeter, and 25 mile (40 km) radius. However, the 25 mile radius results were not used since they did not differ significantly from the plant perimeter results. Both are dominated by weapon fallout in the atmosphere. Ion exchange chromatography was used to remove Pu from the water.

An extensive study of Pu concentrations in soil (0-15 cm depth) was made during 1973. Samples were collected at 36 of the 46 locations shown in Figure 2. Locations 1, 2, and 9 through 16 were not used because of their proximity to F Area. Some of these results and the methods employed have been previously reported (McLendon et al.[3]). Results from two intensively sampled locations, 230 and 420 m from the H Area stack, (Pinder et al.[4]) were also used.

Adhesive papers were used to collect Pu-bearing particles from July 1976 through June 1977 at the four locations shown in Figure 3. The papers, 15.2 x 15.2 cm, were suspended horizontally 7.6 m above the ground and were changed weekly.

All Pu analyses were made using standard radiochemical separations and alpha spectrometry (Pinder et al.[4]).

The influence of wind direction and velocity upon Pu deposition at the various sampling locations was minimized by using only data representing one year or more of deposition. A generally prevailing wind direction does not exist at the Savannah River Plant so, the longer the sampling period, the more uniformly distributed the wind direction becomes. This does not completely eliminate the influence of wind direction and velocity since 85 percent of the ^{238}Pu release from H Area occurred during several days in 1969 following the failure of a sand filter. However, the concentrations in soil do not show a major directional effect.

Curvilinear regression analysis was used to obtain equations relating deposition to distance from the stack. In these equations, x and y stand for the following:

x = distance from stack, in meters (m)

y = pCi ^{238}Pu deposited per m^2 at distance x per mCi ^{238}Pu released from the stack.

RESULTS

From startup in 1954 through 1977, F Area reported releases to the atmosphere of 2.44 Ci of $^{239,240}Pu$ and 0.0062 Ci of ^{238}Pu. From startup in 1955 through 1977, H Area reported releases of 0.58 Ci of $^{239,240}Pu$ and 0.66 Ci of ^{238}Pu. Because the H-Area ^{238}Pu release is more than 100 times that of F-Area, only ^{238}Pu was considered in this study, and H Area was considered the only source. This permitted the evaluation of deposition per quantity released without the necessity of resolving the influence of two sources of release at the different sampling locations. Except for soil, no effort was made to remove the effect of nuclear weapon fallout. Using the results reported by Hardy et al.[5] for weapon fallout, 42 pCi/m^2 was subtracted from the depositions measured in soil.

Deposition of $^{238}Pu/m^2$ per mCi released from H-Area during the period July 1976 through June 1977, as collected by adhesive papers, is shown in Figure 4. During this period, H Area released 13.56 mCi of ^{238}Pu, and F Area released 0.93 mCi. At the location nearest the H-Area stack (230 m), the deposition was 50.4 pCi/m^2 per mCi released and this decreased to 0.36 pCi/m^2 per mCi released at the most distant sampling location (9000 m). Using curvilinear regression analysis, the relation between deposition and distance from the stack was determined to be:

$$\ln y = 10.89 - 1.34 \ln x \qquad (1)$$

Deposition of $^{238}Pu/m^2$ per mCi released during the years 1975, 1976, and 1977, as measured in pan collectors, is shown in Figure 5. During this three-year period, H Area released 21.4 mCi of ^{238}Pu and F Area released 1.8 mCi. At the location nearest the H Area stack (435 m), the deposition was 9.96 pCi/m^2 per mCi released and this decreased to 0.053 at the plant perimeter (average distance, 16,400 m). The relation between deposition and distance from the stack was determined to be:

$$\ln y = 11.0 - 1.36 \ln x \qquad (2)$$

Deposition of $^{238}Pu/m^2$ per mCi released from H Area from startup in 1955 through 1973, as measured in the top 15 cm of soil, is shown in Figure 6. During that period, H Area released 660 mCi of ^{238}Pu and F Area released 6.2 mCi. Previous studies have shown that nearly all the deposited ^{238}Pu remains in the surface 0-15 cm of soil. At a distance of 230 meters from the H-Area stack, the deposition was 86.4 pCi/m^2 per mCi released, and this decreased to about 0.2 pCi/m^2 per mCi released (from the best fitting line) at 8,500 m. The relation between deposition and distance from the stack was determined to be:

$$\ln y = 10.92 - 1.38 \ln x \qquad (3)$$

CORRELATION

The equations of the curves for the adhesive paper collector data, the rain and dryfall collector data, and the soil sampling data (listed below) show

Adhesive Papers	$\ln y = 10.89 - 1.34 \ln x$	(Equation 1)
Rain and Dryfall Collectors	$\ln y = 11.00 - 1.36 \ln x$	(Equation 2)
Soil Samples	$\ln y = 10.92 - 1.38 \ln x$	(Equation 3)

a remarkable similarity. Plotting the best fitting lines for the three measurements (Figure 7) bears out this similarity. Both collection systems, adhesive paper and pan collectors, compare favorably with the soil data.

Examination of the data on a monthly basis resulted in considerable variation with no well defined trend. By pooling the data at yearly intervals or longer, the relationship of activity vs. distance is extremely well differentiated by the two different collection methods. The most plausible explanation to account for this is that there is no generally prevailing wind direction on the Savannah River Plant site.

DISCUSSION

Several general points can be deduced from the data. First, whatever the accuracy is in determining the amount of ^{238}Pu released from the H-Area stack, determinations of the amounts released are extremely consistent over 22 years. Second, the general composite slope for activity vs. distance from all 3 equations depicting the data from 3 different sample types must be indicative of the true slope of activity vs. distance. Third, the steep slope of the composite curve clearly demonstrates the benefit of locating nuclear fuel separation facilities at a considerable distance from the public zone. The deposition rate at 10,000 m from the stack is only 0.4% of the deposition rate at 200 m from the stack. At 10,000 m, the total deposit from SRP operations is small compared to that from global fallout. Fourth, the consistency of the adhesive paper data with the data obtained by the other methods indicates dry deposition is the principal mechanism influencing plutonium deposition. During rain, particles in suspension are not retained on the paper and hence plutonium-bearing particles removed from the atmosphere during rain are lost from the adhesive paper.

Equations 1, 2, and 3 are unique to SRP because they are dependent upon the method to measure the source term, the particle

size of the source term, and the meteorological conditions prevailing in the area. Work is currently underway to characterize the particle size distribution in order to permit evaluation of meteorological models describing deposition of particulate material. These models can then be used to estimate deposition if the particle size distribution and meteorological conditions are different.

REFERENCES

1. Sanders, S. M., Jr., 1977. **Characterization of Airborne Plutonium-Bearing Particles from a Nuclear Reprocessing Plant.** U.S. Department of Energy Report DP-1470. Savannah River Laboratory, Aiken, S.C.

2. Sanders, S. M., Jr., and Boni, A. L., 1980. **The Detection and Study of Plutonium Particles Following the Reprocessing of Nuclear Fuel,** in Transuranic Elements in the Environment, W. C. Hanson, Editor, DOE/TIC-22800, pp 107-144. NTIS.

3. McLendon, H. R., Stewart, O. M., Boni, A. L., Corey, J. C., McLeod, K. W., Pinder, J. E., III, 1976. **Relationships Among Plutonium Contents of Soil, Vegetation, and Animals Collected On and Adjacent To an Integrated Nuclear Complex in the Humid Southeastern United States of America,** in Transuranic Nuclides in the Environment, pp 347-363.

4. Pinder, J. E. III, Smith, M. H., Boni, A. L., Corey, J. C., and Horton, J. H., 1979. **Plutonium Inventories in Two Old Field Ecosystems in the Vicinity of a Nuclear-Fuel Reprocessing Facility,** Ecology 60 (6), 1141-1150.

5. Hardy, E. P., Krey, P. W., and Volchok, H. L., 1973. **Global Inventory and Distribution of Fallout Plutonium,** Nature (London) 241, 444.

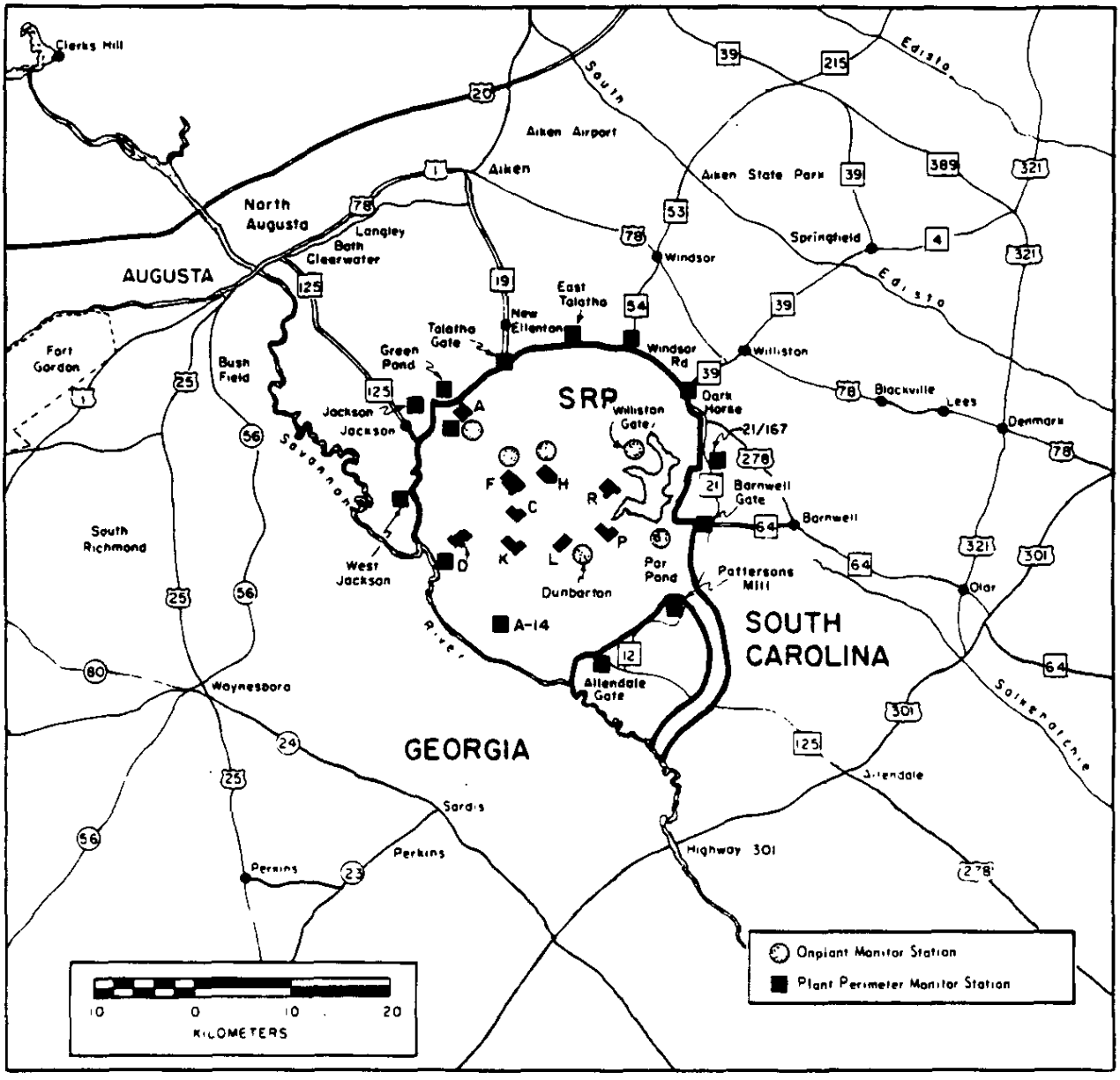

FIGURE 1. Location of Rain and Dryfall Collectors

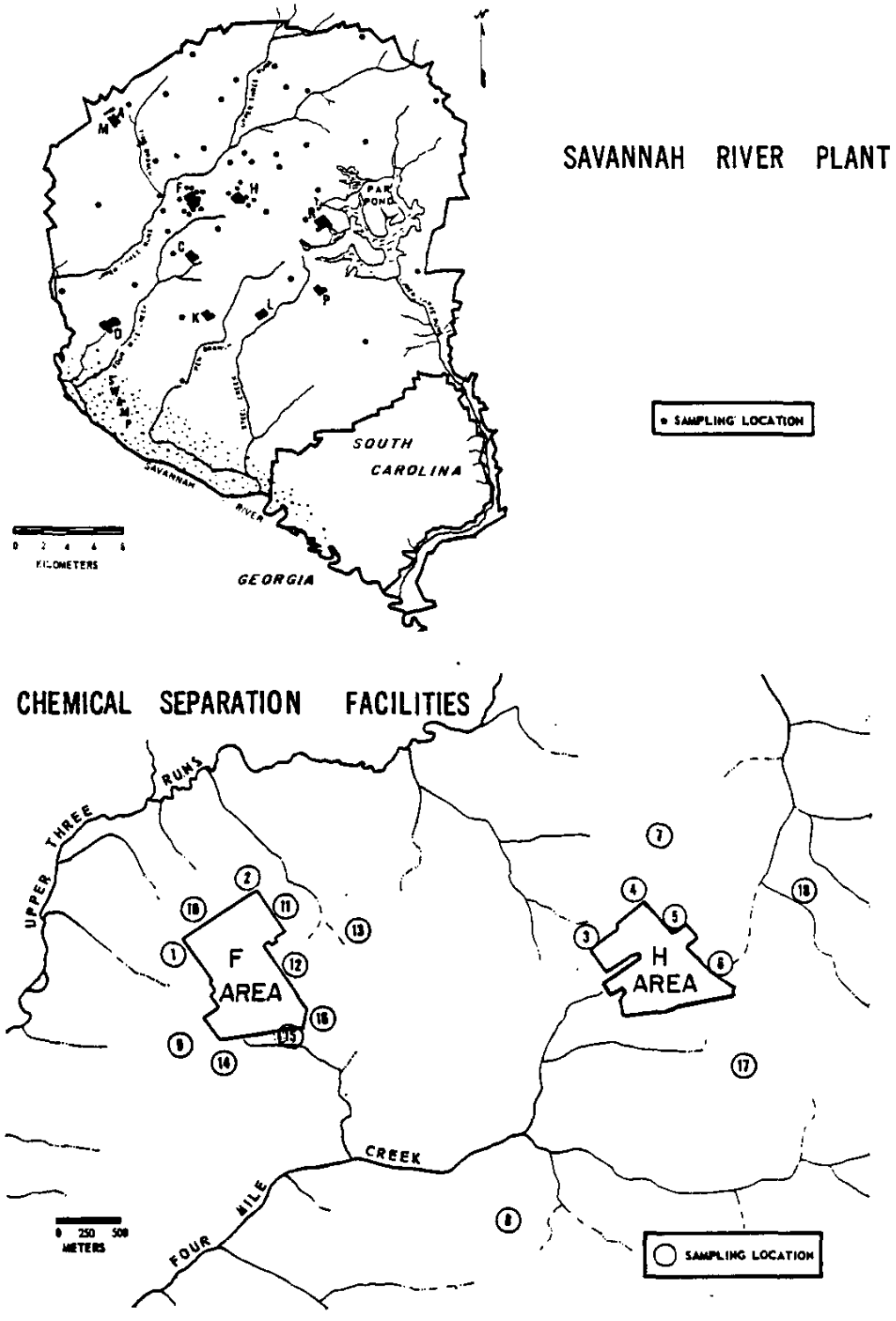

FIGURE 2. Location of Soil Sample Points

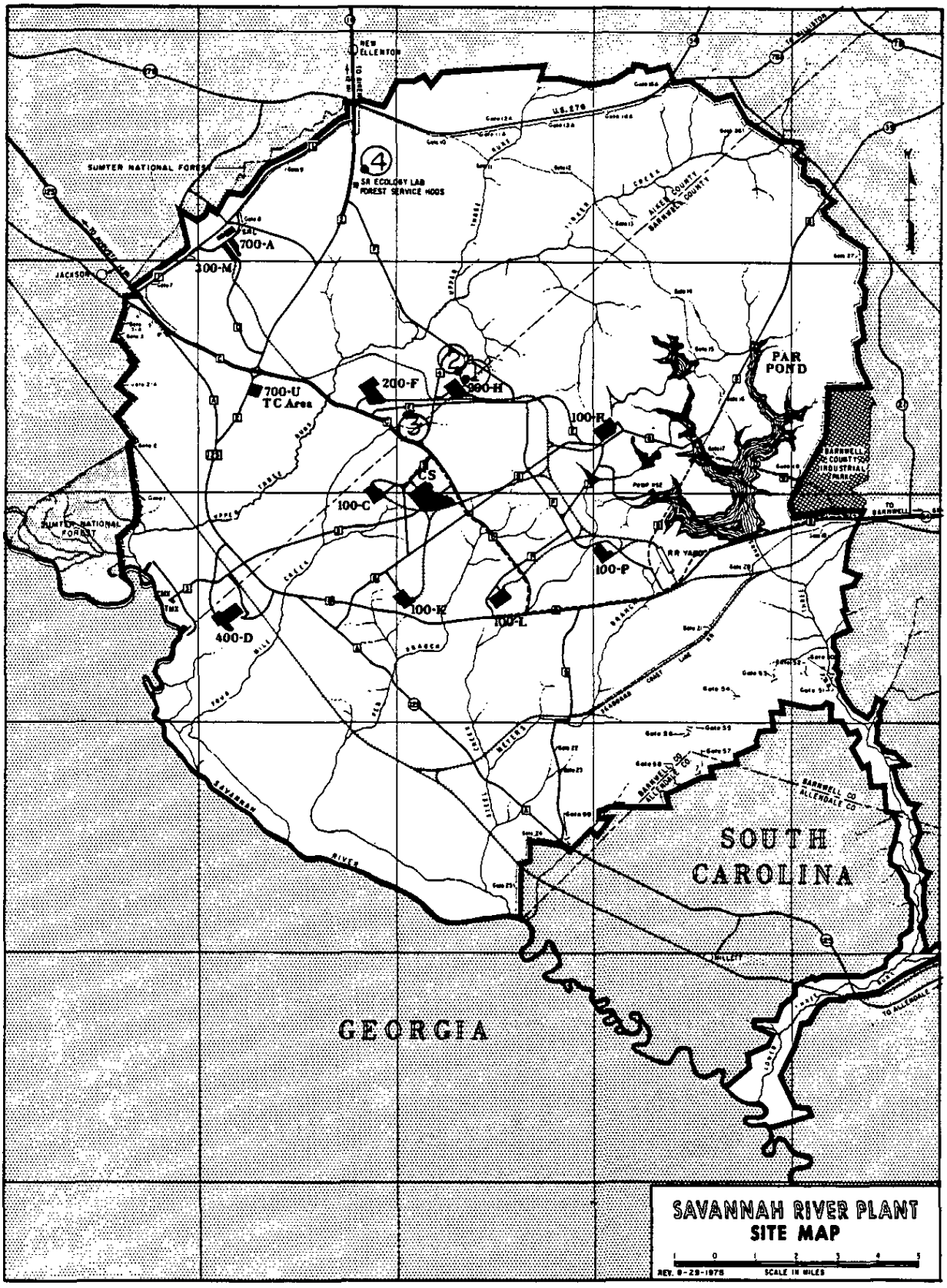

FIGURE 3. Location of Adhesive Papers Collectors

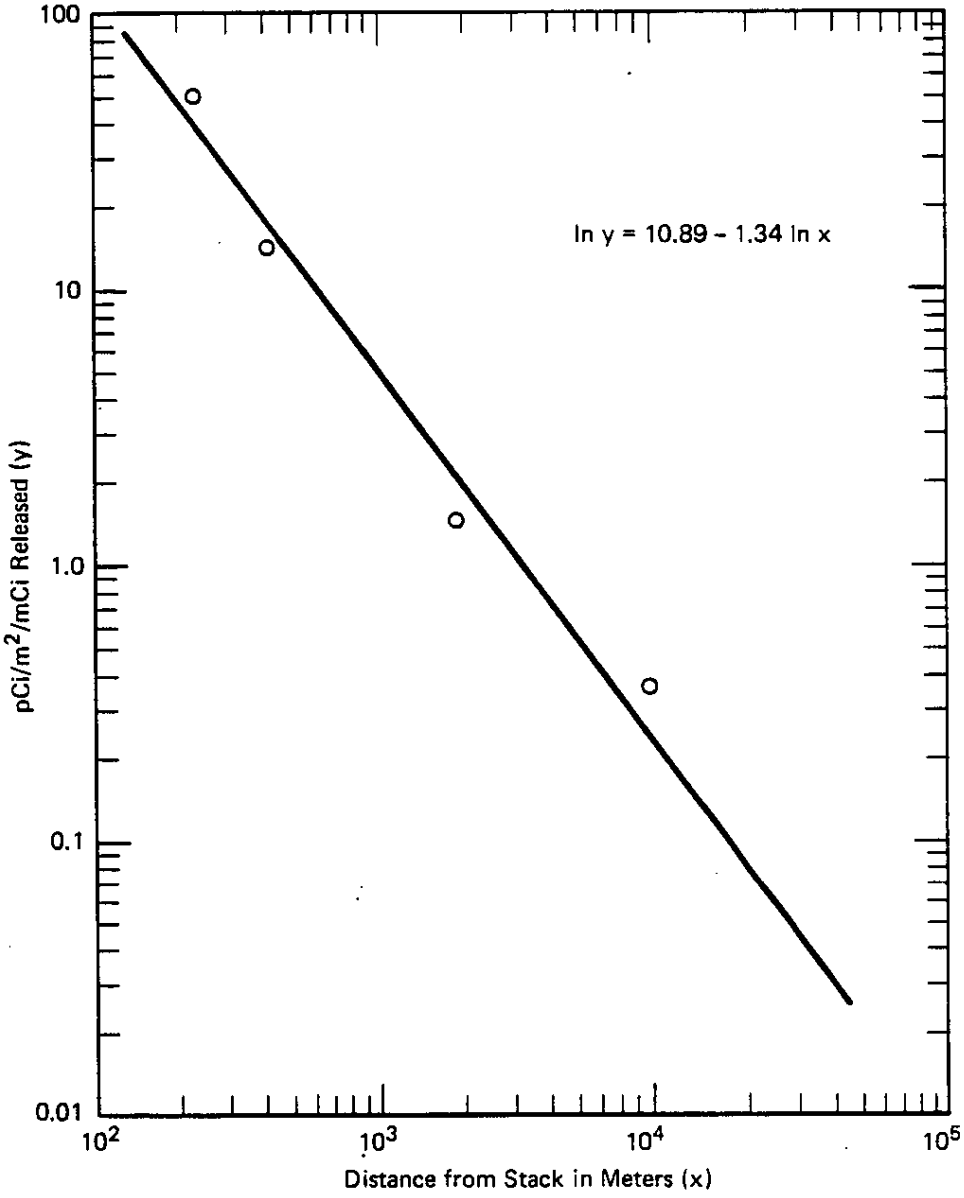

FIGURE 4. Deposition of Pu-238 from H-Area Stack from July 1976 through June 1977 Determined by Adhesive Paper Collectors

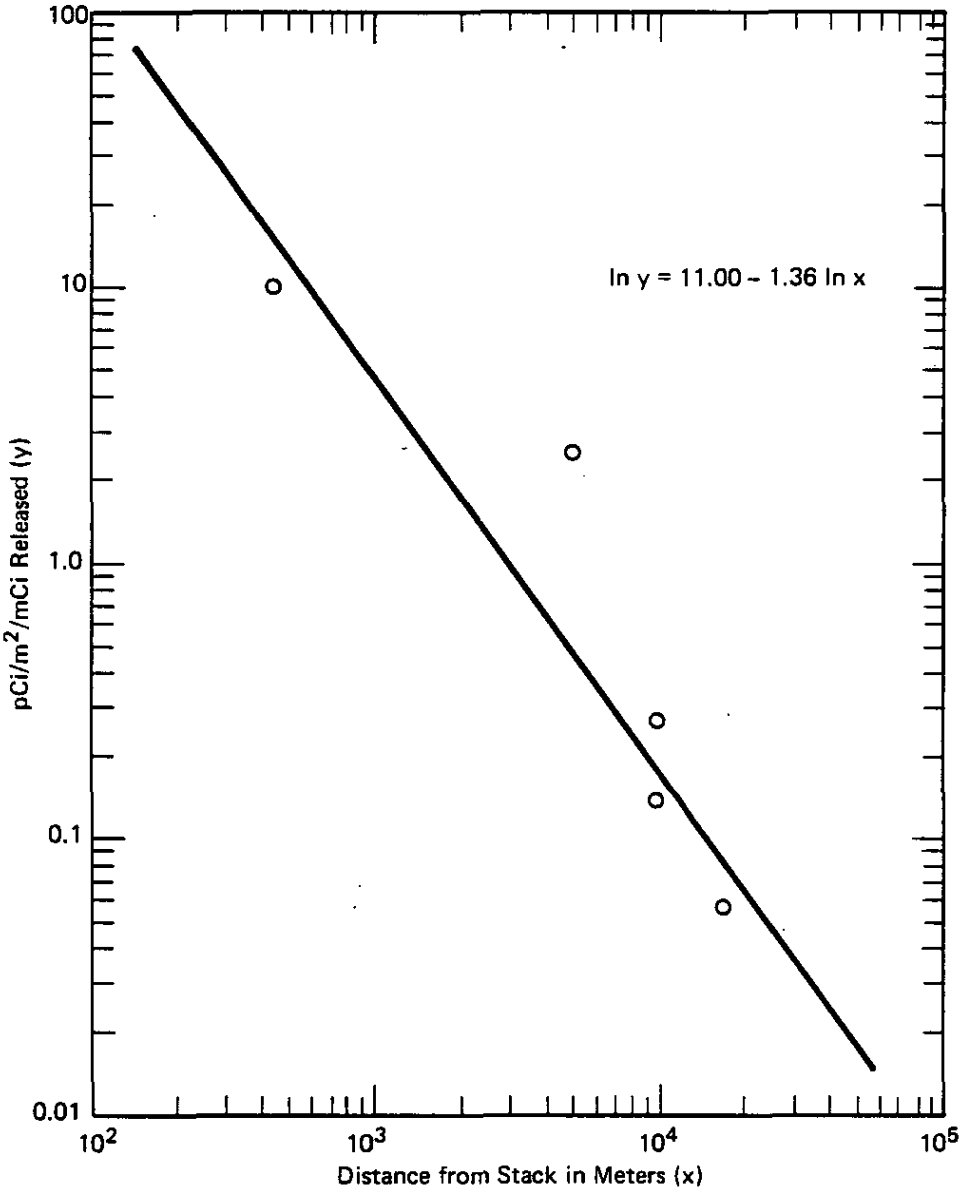

FIGURE 5. Deposition of Pu-238 from H-Area Stack from 1975 to 1977 Determined by Rain and Dryfall Collectors

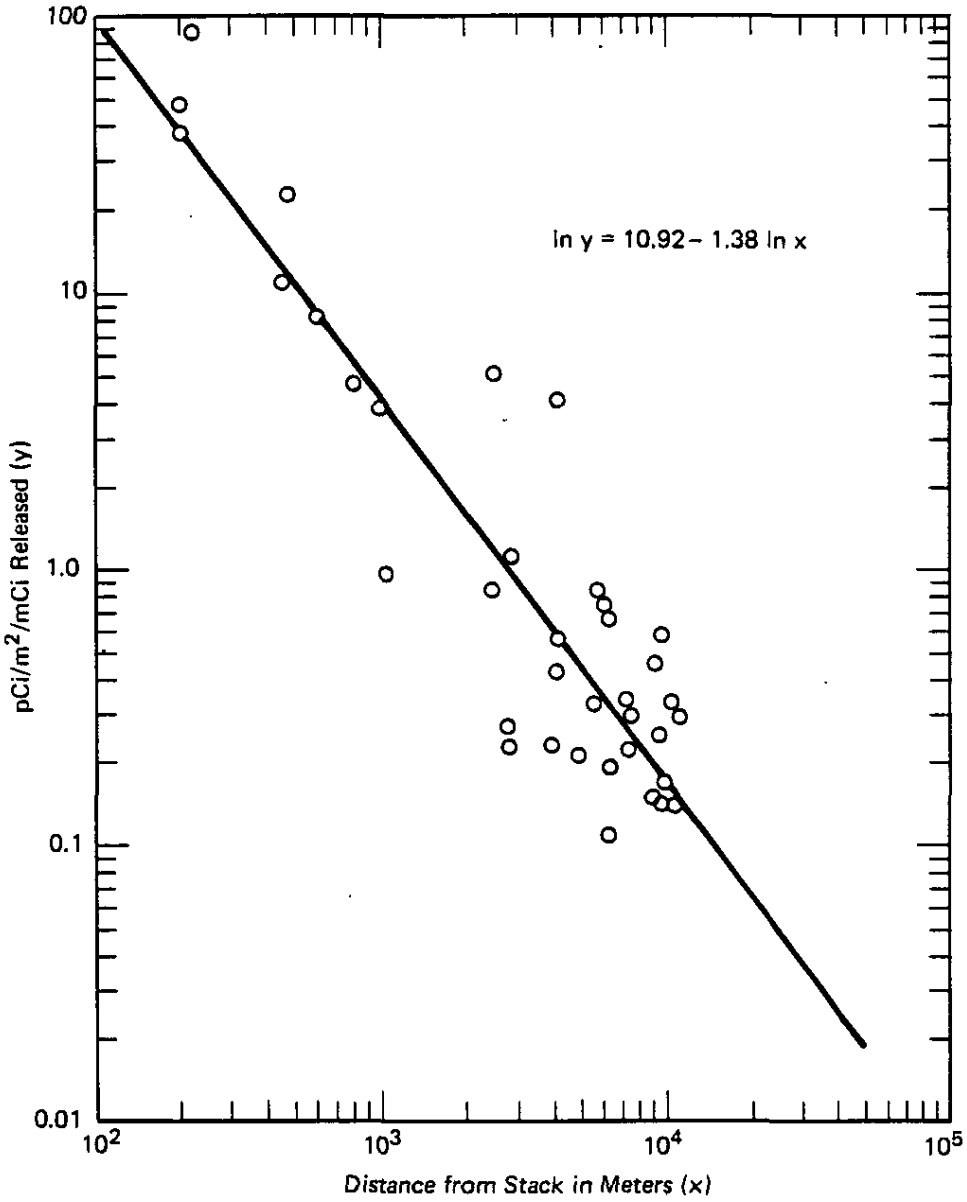

FIGURE 6. Deposition of Pu-238 from H-Area Stack from 1955 to 1973 Determined by Soil Samples

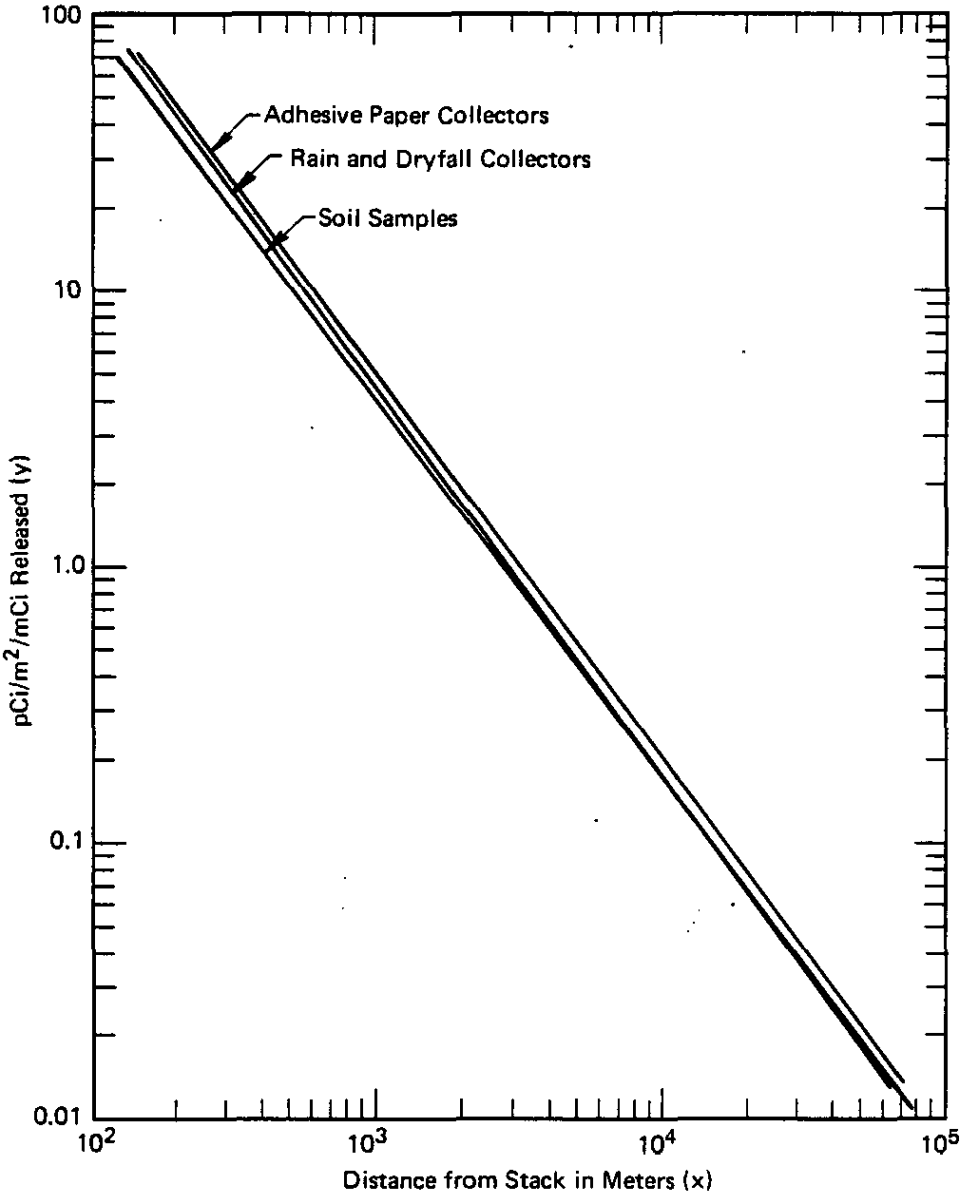

FIGURE 7. Comparison of Three Methods for Measurement of Pu-238 Deposition for Releases from H-Area Stack