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IODINE-129 IN THE ENVIRONMENT OF A NUCLEAR FUEL REPROCESSING PLANT BY NEUTRON ACTIVATION ANALYSIS

by

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IODINE-129 IN THE ENVIRONMENT OF A NUCLEAR FUEL REPROCESSING PLANT BY NEUTRON ACTIVATION ANALYSIS*

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ABSTRACT

^{129}I has been analyzed in both aqueous and soil samples to characterize the environmental impact of the Savannah River Plant (SRP) reprocessing operations. ^{129}I was quantified by a neutron activation procedure with γ-ray spectrometric detection [Ge(Li)]. For many samples, natural iodine (^{127}I) was also quantified. A wide range of samples can be accommodated by a combustion-based preirradiation isolation technique. ^{129}I amounts as low as 3.8 fCi can be determined with counting times of less than 30 minutes (s_{rel} = 10%).

Deposition of ^{129}I in the environment via aqueous and airborne emissions has been studied. Data from analysis of seepage basin contents, spring water, on-plant streams, and the Savannah River

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show that ^{129}I from the seepage basins migrates easily through the soil with the groundwater, eventually reaching the Savannah River. Annual aqueous release rates were estimated to be 16-27 mCi/year. Data from analysis of soils from distances up to 159 km from SRP show above background levels of ^{129}I in both the minimum and maximum airborne transport directions. The soil results are compared with a wind dispersion model. The vertical distribution of ^{129}I in the soil was determined to a depth of 61 cm.

INTRODUCTION

Measurable quantities of anthropogenic ^{129}I now exist in the world due to fission product releases associated with nuclear weapons testing, nuclear reactor operation, and fuel reprocessing. The buildup of ^{129}I is due to a fission yield of 0.9% from ^{235}U and a very long half-life (1.7×10^7 years).

The short-term biological behavior of radioiodine (as ^{131}I) and its pathways to man have been thoroughly studied.[1] Major pathways for thyroid dose by airborne radioiodine are consumption of milk by infants and milk and leafy vegetables by adults. Infants are the most critical element of the population. Aqueous radioiodine can reach man through drinking water and seafood.[2,3] Some soils retain radioiodine, possibly as a result of organic and/or microbial soil fractions.[4,5] However, radioiodine can be highly mobile in water-saturated soil systems.[6,7,8]

This study presents the results of the ^{129}I analysis of a number of environmental samples from the environs of the Savannah River Plant. This nuclear facility encompasses fuel fabrication, nuclear materials production reactors, fuel reprocessing, and waste disposal operations, the latter two being the principal sources of ^{129}I emissions. Both aqueous and airborne routes were studied.

The aqueous samples were primarily related to two seepage basin systems which receive low-level radioactive aqueous discharges from two fuel reprocessing areas. Included were seepage basins, monitoring wells, a spring receiving seepage from the basins, on-plant streams, and the Savannah River.

The impact of airborne ^{129}I released from the fuel reprocessing areas on the surrounding environment was observed relative to an existing dispersion model by collecting vegetation-litter and soil samples out to a distance of 159 km from SRP. Vertical distribution in the soil was also observed to a depth of about 60 cm. With two exceptions, the sampling sites were well-forested pine or oak stands.

All analyses were conducted with a neutron activation procedure derived from the work of Brauer et al.[9] This procedure has been used at the Savannah River Laboratory for over 3 years for a wide variety of sample types. The procedure simultaneously determines natural iodine, permitting calculation of the dose-significant ^{129}I/^{127}I atom ratio.

ANALYTICAL METHODOLOGY

Sampling

All aqueous samples except those from the seepage basins were grab samples taken in new polyethylene containers. Seepage basin samples were 0.5-mL aliquots removed from the monthly grab samples collected by SRP's Health Physics organization.

Vegetation-litter and soil samples were collected and processed with particular attention to the following: Whenever possible, the number of each type of sample collected at a site was large enough to form a good representative composite. The sampling techniques defined the area of each sample allowing easy comparison with dispersion models. Finally, soil core processing techniques were developed which eliminated the possible problem of vertical cross-contamination resulting from driving the core tubes into the ground.

Ten vegetation-litter samples were combined in a large plastic bag at each sampling site to prepare a good representation of the site. The ten individual portions were collected by removing, down to the soil surface, all vegetation and litter contained within a 30.5 × 30.5 cm *Lucite*® (Du Pont) frame placed on the ground.

A surface soil core (2.54 cm thick by 5.08 cm diameter) was taken within each square of ground from which the vegetation and litter were removed, and all such cores at a given site were combined in a plastic bag. Also within each square of ground, a 7.30 cm ID by 91.4 cm long thin-wall steel coring tube (Acker #120037-5) was driven into the ground to a depth of about 76 cm. The protruding length of core tube was measured for later calculation of the soil compaction. In some instances, the distance from the top of the tube to the top of the soil column was measured at the site; and in others, it was measured just before the tube was processed. After extraction from the ground, the tube ends were sealed with plastic caps. The core tubes were transported in a vertical position to the processing location where they were frozen. Whenever possible, 10 core tubes were collected at each site. In some cases, weather or soil conditions limited that number to as few as 5.

Pretreatment

The volumes of aqueous samples ranged from 0.5 to 2000 mL. Each 0.5-mL sample (i.e., seepage basin and spring) was placed in a 1-mL disposable polyethylene pipet tip whose narrow end was heat-sealed shut. The other end was capped with a small square of *Parafilm*® (American Can Company) after addition of the small volume of ^{125}I spike solution. The resultant capsule was ready for combustion. For larger samples (i.e., river and streams), a known amount of ^{125}I was added to the sample as soon after sampling as practical. The samples were then passed through a 15 × 120 mm, 20-50 mesh, *Dowex*® 1-X8 (Dow Chemical Company) anion exchange column in the Cl^- form. The resin containing the strongly retained iodine was reserved for the combustion procedure described below.

The combined vegetation and litter samples from each site were frozen until processed. After the total weight was determined, the samples were chopped into smaller pieces for easier blending and sampling. Weighed, manageable portions of the blended samples were dried overnight in an 100°C oven to determine moisture content. After spiking with a known amount of ^{125}I, weighed portions of the dried samples were combusted.

The soil core tubes from each site were frozen until processed. Based on measurements of soil core length and core tube penetration, the core tubes were sawed at levels corresponding to depths of 15.2, 30.5, and 61.0 cm with a handheld portable band saw. Secondary cores (2.54 cm thick by 5.08 cm diameter) were taken downward from each of the cuts after careful surface cleaning designed to eliminate potential contamination by the cutting. The secondary cores at each level were composited for each site. After weighing, the composites were dried overnight at 100°C and reweighed to determine moisture content. Any lumps in the dried composites were crushed, and the samples were sieved with a 35-mesh screen. The <35-mesh fraction was blended for 30 minutes in a 1/2-quart acrylic V-blending shell in a Patterson-Kelly model YB-B blender. Portions of the blended samples were spiked with ^{125}I and subjected to the combustion procedure. The volume of the >35-mesh samples allowed each to be burned in its entirety.

The 10 surface soil cores combined at each site were also dried, blended, and sieved as above. Combustion was similarly performed after spiking.

Combustion, Irradiation, and Counting

The resin, capsule, vegetation-litter, and soil samples were burned in a quartz combustion apparatus according to a procedure designed to isolate and concentrate the sample and spike iodine in a 2.4 × 35 mm evacuated sealed quartz ampoule. Six such ampoules, four samples and two standards containing known amounts of ^{127}I and ^{129}I, were sealed in a 15 × 74 mm evacuated aluminum capsule by electron beam or TIG welding. The capsule was irradiated for 2 hours in a reactor at a neutron flux of about $4 \times 10^{14}/cm^2$-sec. After irradiation, the contents of the ampoules were subjected to carrier-aided, redox-controlled, distillation and solvent-extraction steps. Finally, the iodine from each ampoule was isolated as a AgI precipitate on a 25-mm-diameter membrane filter for counting.

^{128}I, the activation product of ^{127}I, was quantified immediately after the isolation by counting at 443 keV with a 44 × 71 mm Ge(Li) detector and multichannel analyzer. After decay of the 25-minute ^{128}I, ^{130}I (the activation product of ^{129}I) was determined at 536 keV with the same counting equipment. Finally, after decay of the 12.5-hour ^{130}I, ^{125}I was counted with a low energy photon detector and multichannel analyzer to determine the iodine recovery of the entire procedure. Typically, recoveries of 50% were obtained. The ^{127}I and ^{129}I concentrations in the samples were then calculated from the decay-corrected ^{128}I and ^{130}I count rates, the measured iodine recoveries, the standard concentrations, and the sample weights. Samples which used the anion exchange resin technique were corrected for the natural iodine blank of the resin. By Currie's definition of the quantification limit,[10] the procedure can determine as little as 4 fCi of ^{129}I (s_{rel} = 10%) with a 30-minute counting period. More detailed descriptions of the reactions, equipment, and procedures are available.[9,11]

RESULTS

Seepage Basins and Spring Samples

Table 1 summarizes 9 months of ^{129}I and ^{127}I data from the last and largest basins of the two seepage basin systems receiving low-level radioactive waste from the fuel reprocessing areas. Basin F-3 exceeds Basin H-4 in ^{129}I concentration and the ^{129}I/^{127}I atom ratio. The differences between the two basin systems reflect chemical differences in the reprocessing schemes which cause most of H-Area's aqueous iodine emissions to be routed to high-level waste storage tanks rather than to the seepage basins.

Also in Table 1 are data from analysis of a spring which receives seepage from the F-Area basins and empties into nearby Four Mile Creek. Analysis of water from nine monitoring wells placed between the seepage basins and Four Mile Creek resulted in ^{129}I values ranging from 0.083 to 297 pCi/L. The highest values correlated well with known major groundwater movement patterns determined earlier from ^{3}H migration studies. These results indicate that ^{129}I moves easily with the groundwater through the sandy clay, clayey sand, silt, and clay soils at the site.

Data from analysis of Four Mile Creek, other on-plant streams, and the Savannah River are presented in Table 2. As expected from its relationship with the seepage basins and spring, Four Mile Creek conveys most of the aqueous ^{129}I from SRP to the Savannah River. Based on flows at the time the samples were taken, the output rate is estimated to be 16-27 mCi/yr. Periodic Savannah River and stream sampling is underway to generate a larger data base for a more confident estimate of aqueous ^{129}I emission and to detect any time-dependent factors influencing ^{129}I concentrations. In all cases, the concentrations found in the Savannah River have been several orders of magnitude below the Federal concentration guides for releases to uncontrolled areas.[12]

Vegetation-Litter and Soil Samples

Data from the analysis of vegetation-litter and soil samples are shown in Tables 3 and 4. Table 3 records the 103° azimuth, the direction in which the airborne transport of ^{129}I is predicted to be maximum by the probability distribution meteorological model described below. Table 4 records the minimum transport direction.

Since the area of soil sampled was set by the dimensions of the coring tool and the number of cores taken, it was possible to calculate the analyte content of a unit area layer of the coring thickness. Similarly, the area of ground from which the vegetation and litter were removed was also set allowing normalization of the analyte content to unit area. The data in Tables 3 and 4 are expressed in terms of such "area concentrations."

The area concentrations of ^{129}I for soil samples can be empirically expressed as a function of depth by the following equation:

$$\ln [^{129}I] = mx + b \tag{1}$$

where

$[^{129}I]$ = area concentration of ^{129}I
x = midpoint depth of layer sampled

m and b are the slope and intercept of a least-squares fit of the soil data to equation (1). Figure 1 illustrates this relationship for one site.

Integrating equation (1) to an infinite soil depth gives a finite integral whose value represents the total amount of ^{129}I deposited on a unit area of soil. Adding the vegetation-litter area concentrations to the integrated soil concentrations results in an estimate of the total unit area deposition of ^{129}I. Since weapons fallout and natural ^{129}I concentrations are insignificant relative to the measured concentrations,[2] the total ^{129}I deposited per unit area is attributed to SRP.

R. E. Cooper's CHIDIS probability distribution code[13] for determining airborne pollutant concentrations from a continuous point source is based on the following equation:

$$\chi = \frac{Q}{\alpha \pi \bar{u} \sigma_y \sigma_z} \left\{ \exp - \left[\left(\frac{H - xV_g/\bar{u}}{\sqrt{2}\, \sigma_z} \right)^2 + \frac{\lambda x}{\bar{u}} \right] \right\} \qquad (2)$$

where χ = downwind concentration (m^{-3})

Q = source term. If Q is the release per unit time for a continuous release, the χ will be a steady state concentration.

$\bar{u}$ = effective downwind transport velocity, generally taken to be the measured windspeed at a height H (m/sec)

H = effective release height for Q (m)

σ_y, σ_z = distance equal to one standard deviation of the material distribution in the vertical and crosswind axes relative to the center of the plume (m)

V_g = gravitational settling velocity (m/sec)

x = downwind distance at which χ is being computed (m)

$\alpha = 1.0$ if $V_g = 0$, $\alpha = 2.0$ if $V_g > 0$

λ = radioactive decay constant (sec^{-1})

Solving equation (2) for 15-minute average meteorological records of a 2-year data base, and assuming an average release rate of 6×10^{-9} Ci/sec of ^{129}I for 21 years and a nominal gravitational settling velocity of 0.01 m/sec[14,15] result in the maximum and minimum transport predictions shown as solid lines in Figure 2. Also indicated are the total ^{129}I area concentrations found for those two directions.

Both the maximum and minimum transport data describe curves of the same general shape as the model predicts. The maximum transport data describe a smooth curve with little scatter. On the other hand, considerable scatter is seen in the minimum transport data. The 19- and 79-km sites of the minimum transport azimuth were unlike the remaining sites in that they lacked forestation: The 19-km site was a forest clearing and the 79-km site was a pasture. All other sites were well-forested. The low ^{129}I area concentrations of the two unique sites may in large part be due to the lower efficiencies of non-forested sites for collection of airborne pollutants.[16] The remaining six sites could be described reasonably well by a single curve. The agreement with the model is quite acceptable when the uncertainties in assumed deposition velocities and historical emission rates are considered.

CONCLUSIONS

The aqueous sample results demonstrate that ^{129}I from the seepage basins moves easily through the sandy clay, clayey sand, silt, and clay soils at the Savannah River Plant to the external aqueous environment.

The impact of SRP airborne ^{129}I emissions is clearly visible at 160 km from the source with the high sensitivity neutron activation analysis used. An existing probability distribution model based on 2 years of meteorological data provides a reasonable estimate of ^{129}I deposition. However, more data are needed in other transport directions for a definitive comparison.

The vertical distribution of ^{129}I in sandy clay soils resulting from deposition of airborne emissions was inversely related to depth by a logarithmic function. Since half-depths on the order of 8 cm were found, very slow downward migration is indicated. This, in turn, implies a long residence time for ^{129}I in regions of the soil which are subject to agricultural activity and wind-driven resuspension. However, ^{129}I/^{127}I atom ratios in all soil samples in this study were orders of magnitude below the limiting specific activity calculated for the maximum permissible thyroid burden.[3] Furthermore, little is known about soil ^{129}I uptake by typical crops under such environmental conditions. As clearly seen in the comparison of the migration of aqueous and airborne ^{129}I through soil, large differences in behavior can exist. These are probably related to differences both in matrices and the chemical/physical forms of ^{129}I.

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TABLE 1

^{129}I and ^{127}I in Seepage Basin and Spring Samples

Sample	*Date*	*^{129}I (pCi/L)*	*^{127}I (μg/L)*	*^{129}I/^{127}I Atom Ratio*
Basin F-3	8-12-77	140 ±4	83 ±1	(9.9 ±0.3) × 10^{-3}
	12-30-77	152 ±6	NA[a]	-
	1-27-78	54 ±3	44 ±2	(7.2 ±0.5) × 10^{-3}
	3-23-78	198 ±5	33 ±5	(35 ±5) × 10^{-3}
	4-21-78	143 ±4	48 ±6	(18 ±2) × 10^{-3}
Basin H-4	8-12-77	74 ±3	61 ±11	(7.2 ±1.3) × 10^{-3}
	12-30-77	71 ±4	NA	-
	1-27-78	50 ±4	55 ±4	(5.4 ±0.6) × 10^{-3}
	3-23-78	44 ±2	57 ±4	(4.6 ±0.6) × 10^{-3}
	4-21-78	18 ±2	78 ±7	(1.4 ±0.2) × 10^{-3}
Spring	8-29-77	224 ±6	25 ±7	(53 ±14) × 10^{-3}

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[a] NA = not analyzed.

TABLE 2

^{129}I in Stream and River Samples

All samples taken 8/25/77

Sample Location	*^{129}I (fCi/L)*	*$^{129}I/^{127}I$ Atom Ratio*
Four Mile Creek[a]	43 ±2	17×10^{-6}
Upper Three Runs[b]	1.1 ±0.3	1.2×10^{-6}
Lower Three Runs	0.2 ±0.2	0.1×10^{-6}
Steel Creek	1.3 ±0.2	0.5×10^{-6}
Savannah River (Upstream)	1.6 ±0.5	0.5×10^{-6}
Savannah River (Downstream)	5.2 ±0.4 5.4 ±0.3 (duplicate)	1.6×10^{-6}

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[a] Major stream receiving seepage from basin system.

[b] Originates off-plant. Other creeks originate on-plant.

TABLE 3

^{129}I and ^{127}I in Vegetation-Litter and Soil Samples

103° Azimuth — Direction of Maximum Predicted Transport

Distance From Source (km)	*Layer Depth (cm)*	^{129}*I (pCi/m^2)*	^{127}*I (mg/m^2)*	^{129}*I/*^{127}*I Atom Ratio*
19	Veg-Litter	3.8 ±0.2	2.0 ±0.1	(11 ±0.8) × 10^{-6}
19	0 - 2.7	20.4 ±0.6	18 ±1	(6.7 ±0.4) × 10^{-6}
19	15.2 - 17.9	4.4 ±0.2	32 ±1	(0.81 ±0.04) × 10^{-6}
19	30.5 - 33.2	1.5 ±0.2	47 ±2	(0.19 ±0.03) × 10^{-6}
19	61.0 - 63.7	-0.06 ±0.3	55 ±4	-
48	Veg-Litter	0.80 ±0.06	1.4 ±0.1	(3.4 ±0.3) × 10^{-6}
48	0 - 2.7	6.8 ±0.4	18 ±1	(2.2 ±0.2) × 10^{-6}
48	15.2 - 17.9	1.7 ±0.2	39 ±2	(0.26 ±0.03) × 10^{-6}
48	30.5 - 33.2	0.8 ±0.1	37 ±2	(0.12 ±0.02) × 10^{-6}
48	61.0 - 63.7	0.3 ±0.3	51 ±3	-
84	Veg-Litter	0.21 ±0.02	0.76 ±0.03	(1.6 ±0.2) × 10^{-6}
84	0 - 2.7	5.7 ±0.2	35 ±2	(0.96 ±0.06) × 10^{-6}
84	15.2 - 17.9	0.68 ±0.07	57 ±3	(0.070 ±0.008) × 10^{-6}
84	30.5 - 33.2	0.19 ±0.07	63 ±3	(0.018 ±0.007) × 10^{-6}
84	61.0 - 63.7	0.2 ±0.3	52 ±3	-
159	Veg-Litter	0.11 ±0.04	1.0 ±0.1	(0.6 ±0.2) × 10^{-6}
159	0 - 2.7	1.2 ±0.2	40 ±2	(0.18 ±0.03) × 10^{-6}
159	15.2 - 17.9	0.17 ±0.06	56 ±3	(0.018 ±0.003) × 10^{-6}
159	30.5 - 33.2	0.2 ±0.2	80 ±3	-
159	61.0 - 63.7	0.7 ±0.4	49 ±3	(0.008 ±0.005) × 10^{-6}

TABLE 4

^{129}I and ^{127}I in Vegetation-Litter and Soil Samples

170° Azimuth — Direction of Minimum Predicted Transport

Distance From Source (km)	*Layer Depth (cm)*	^{129}I *(pCi/m^2)*	^{127}I *(mg/m^2)*	$^{129}I/^{127}I$ *Atom Ratio*
19	Veg-Litter	0.29 ±0.02	0.52 ±0.03	(3.3 ±0.3) × 10^{-6}
19	0 - 2.5	6.6 ±0.3	17 ±0.5	(2.3 ±0.04) × 10^{-6}
19	16.8 - 19.6	1.2 ±0.2	41 ±1	(0.17 ±0.03) × 10^{-6}
19	33.3 - 36.1	0.24 ±0.08	47 ±1	(0.03 ±0.01) × 10^{-6}
19	66.5 - 69.3	0.1 ±0.1	29 ±0.5	-
40	Veg-Litter	0.56 ±0.05	1.6 ±0.02	(2.1 ±0.2) × 10^{-6}
40	0 - 2.6	9.1 ±0.4	14 ±0.2	(3.8 ±0.2) × 10^{-6}
40	15.7 - 18.4	1.8 ±0.1	40 ±1	(0.27 ±0.02) × 10^{-6}
40	31.5 - 34.1	0.38 ±0.05	40 ±1	(0.056 ±0.008) × 10^{-6}
40	63.0 - 65.6	0.40 ±0.09	173 ±2	(0.014 ±0.003) × 10^{-6}
79	Veg-Litter	0.006 ±0.004	0.1 ±0.001	-
79	0 - 2.9	0.66 ±0.08	24 ±0.3	(0.16 ±0.02) × 10^{-6}
79	17.1 - 20.0	0.13 ±0.08	46 ±1	(0.016 ±0.010) × 10^{-6}
79	34.3 - 37.1	0.15 ±0.08	64 ±1	(0.014 ±0.007) × 10^{-6}
79	68.6 - 71.4	0.16 ±0.08	68 ±2	(0.014 ±0.007) × 10^{-6}
150	Veg-Litter	0.04 ±0.02	1.1 ±0.01	(0.21 ±0.11) × 10^{-6}
150	0 - 2.8	0.7 ±0.1	NA[a]	-
150	16.9 - 19.7	0.21 ±0.06	50 ±1	(0.025 ±0.007) × 10^{-6}
150	33.8 - 36.6	0.05 ±0.04	40 ±1	-
150	67.5 - 70.3	0.05 ±0.03	27 ±1	(0.011 ±0.007) × 10^{-6}

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a. NA = not analyzed

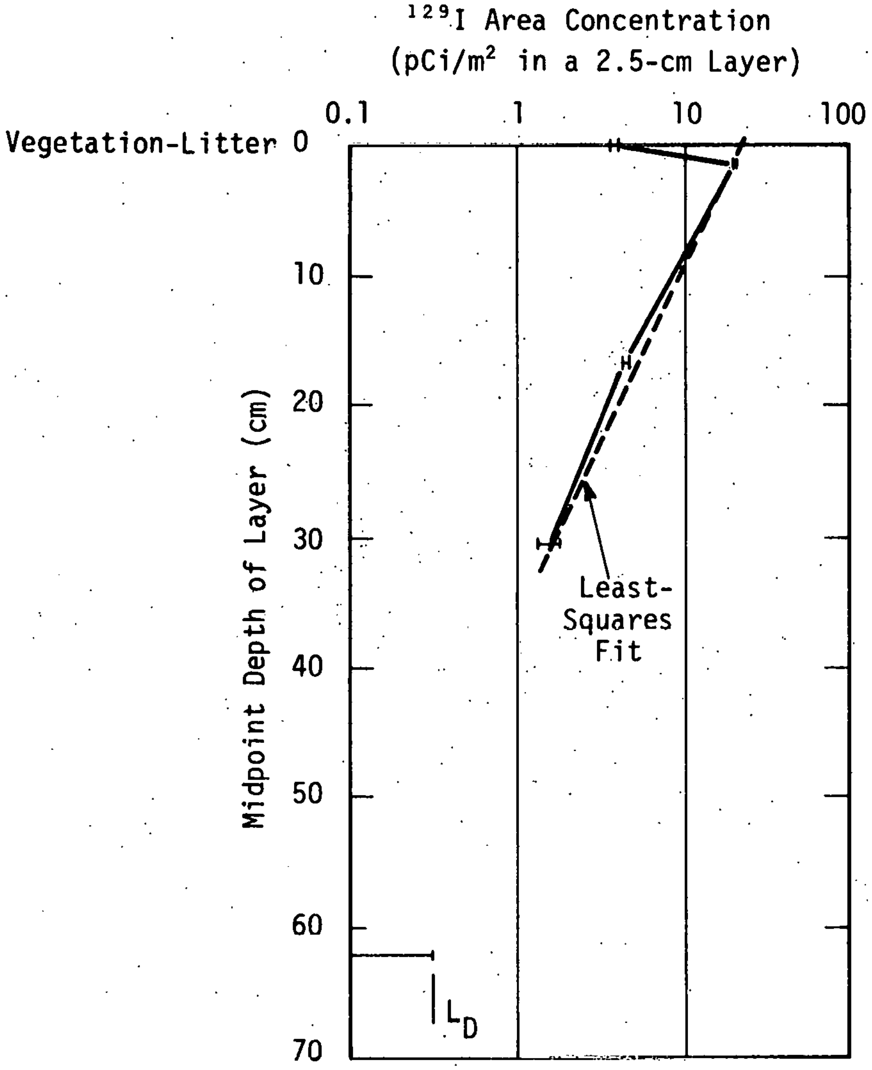

FIGURE 1. ^{129}I Area Concentration Versus Depth for 19-km Site — 103° Azimuth

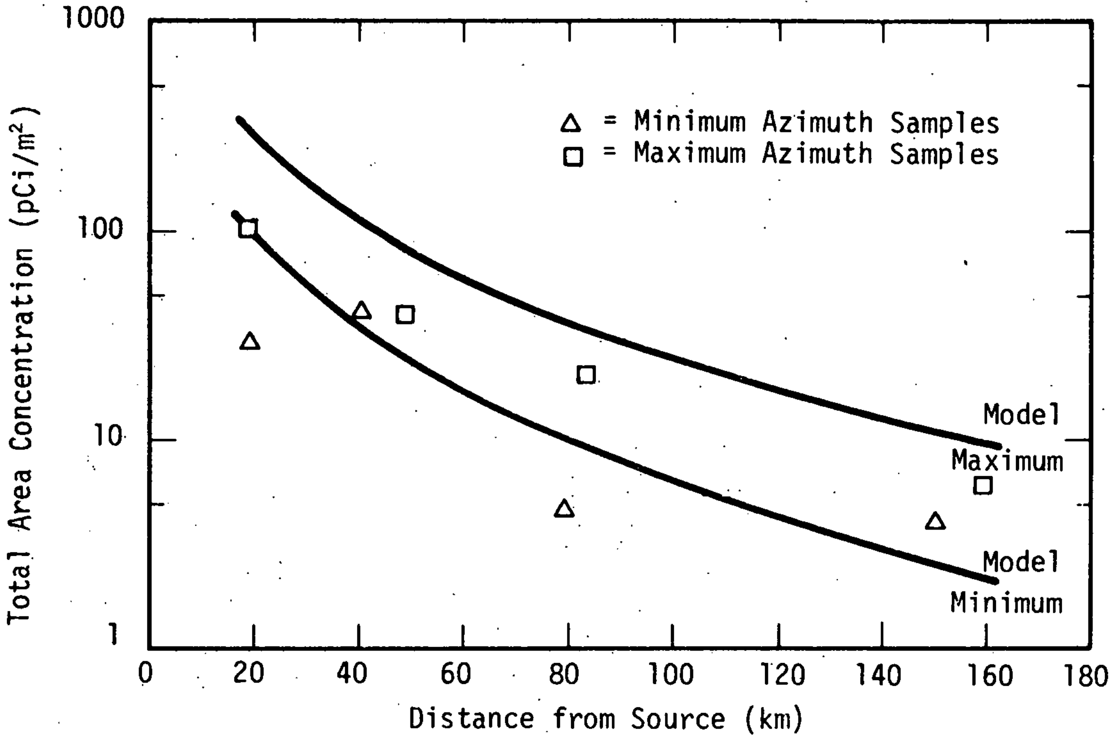

FIGURE 2. ^{129}I Area Concentration Versus Distance from SRP Reprocessing Areas