MASTER

CONF-790903--5

ANALYSIS OF SOIL SAMPLES FROM OMRE DECOMMISSIONING PROJECT

O. D. Simpson, J. A. Chapin, R. E. Hine,
J. W. Mandler, M. P. Orme and G. A. Soli

EG&G Idaho, Inc.
Idaho National Engineering Laboratory
Idaho Falls, Idaho 83401

NOTICE
This report was prepared as an account of work sponsored by the United States Government. Neither the United States nor the United States Department of Energy, nor any of their employees, nor any of their contractors, subcontractors, or their employees, makes any warranty, express or implied, or assumes any legal liability or responsibility for the accuracy, completeness or usefulness of any information, apparatus, product or process disclosed, or represents that its use would not infringe privately owned rights.

INTRODUCTION

The decontamination and decommissioning (D&D) of retired nuclear reactor plants and the restoration of the landsite to its original state is of primary importance to the nation. As of December 1977, there have been fifty-two reactors constructed at the Idaho National Engineering Laboratory (INEL). Seventeen of these are currently operable; the others are in various stages of retirement ranging from defueled and placed under protective confinement to completely decontaminated and open to the public. Several reactors at the INEL have undergone D&D in the past, but these projects were done individually without an integrated plan. An integrated, site-wide, long-range D&D plan[1] has been developed at the INEL, and the Organic Moderated Reactor Experiment (OMRE) facility is the first reactor to be addressed by this plan.

Criteria are being developed at the INEL to be used as guidelines for D&D of land, structures, and equipment at the INEL in order that such land, structures, and equipment may safely be returned to unrestricted use whenever or wherever practicable. In order to accomplish this for the land, the radioactive contamination in the soil must be reduced below a maximum permissible concentration. Criteria suggested in the INEL D&D long-range plan for maximum permissible concentration are 100 pCi/g of soil for transuranic alpha-emitting radionuclides and 1 nCi/g of soil for beta-gamma-emitting radionuclides. The above values assume natural backgrounds have been subtracted. These criteria are still under review, and as data are developed, they may be revised.

The D&D long-range plan also requires a cost/benefit analysis to be performed on each project to evaluate the feasibility of decommissioning to the unrestricted levels listed above. The planned use of the INEL, in the near term, may preclude the expenditure of large sums of money and manpower for D&D when the site will be reused for nuclear applications. A different set of criteria would be considered if the site is to be released for activities such as farming or housing. Each end use may require a separate, specific set of criteria.

Until a critical pathway analysis can be performed for all possible alternatives, specific criteria cannot be established for each facility. Current criteria will be influenced most heavily by safe access considerations, near-term site utilization, radioactive storage space, and cost/benefits.

In order to establish that the present OMRE site does not exceed the above criteria for radioactive contamination, samples obtained from the remainder of the facility that was not removed such as soil, concrete pads, various structural materials, and the leach pond area were analyzed to determine their radioactive content. This paper presents the results of the analyses performed on soil samples.

REACTOR HISTORY AND DESCRIPTION OF REACTOR AREA

The general purpose of the OMRE facility was to study feasibility, economics, and behavior of organic moderator and coolant under power conditions. The nominal reactor power was from 2 to 12 MW thermal. The reactor was designed by Atomics International, constructed by Wadsworth and Arrington and Fluor Maintenance Co., and owned by the United States Atomic Energy Commission. Construction was started in June 1956 and completed in May 1957. The reactor went critical in September 1957, attained full power in February 1958, and was operated until 1963. The facility remained in a deactivated condition from 1963 until October 1977, at which time D&D of the facility was begun. The reactor vessel, buildings, and other structures were removed from the site during 1978.

The reactor core was of rectangular shape having dimensions of 57 cm x 69 cm x 91 cm high. It had a critical mass of 16.4 kg ^{235}U. Fuel elements were constructed from alloyed plates containing 25 wt.% fully enriched UO_2 and 75 wt.% stainless steel. Cladding made of 0.013-cm-thick 304 stainless steel was metallurgically bonded to the fuel plate. The moderator consisted of 380 liters of Santowax OM (diphenyl 16.0%, ortho-terphenyl 46.1%, meta-terphenyl 31.8%, para-terphenyl 6.1%). The mixture is partially solid at 21°C. Melting begins at approximately 38°C

and is completely liquified slightly below 93°C, depending on irradiation history. Total volume of moderator and coolant was 19,000 liters.

A picture of the OMRE facility before D&D was started is shown in Figure I. The reactor was located in the corrugated silo shown on the left side of the picture. Figure II shows how the reactor pit area appears today. All structural materials have been removed including the silo building. The bottom of the pit is where the base of the reactor vessel rested.

SOIL SAMPLING TECHNIQUES

Four sampling techniques (vertical augering, vertical punching, vertical coring, and trenching with horizontal coring) were tested in order to determine a method which will allow radionuclide

Figure I. The OMRE facility before decontamination and decommissioning was started. The reactor is housed in the corrugated silo shown at the left. The reactor core is ~5.5 m below ground level.

Figure II. The OMRE reactor pit area as it appeared in August 1979. Since this time an additional 1 m of basalt and soil has been removed from the bottom of the pit in order to meet soil contamination criteria at INEL. The basalt at the bottom of the pit is where the concrete pad rested that supported the base of the reactor vessel. The Ge(Li) detector used for an *in situ* measurement is shown on the right side of the reactor pit near the center of the picture. The motor home that houses the electronic equipment can be seen in the background.

characterization of the soil with a minimum amount of cross contamination from level to level yet minimize cost. The amount of time required for sampling and analysis, personnel exposure, and the complexity of equipment necessary to perform the sampling process were considered.

Vertical augering was done using a 15.2-cm-diameter, two-man, motor-driven post-hole digger. Samples were collected in 15-cm-thick increments as they were expelled by the digger. The auger was brushed clean between samplings to minimize cross contamination. These samples were sent to the laboratory for radionuclide analysis.

Vertical punching was accomplished by using an 8-cm-diameter by 91-cm-long steel punch driven by a sledge hammer. A beta-gamma radiation detector was lowered into the hole and total radiation levels recorded as a function of depth.

Vertical cores were taken by driving a core-sampling probe into the soil. The probe was retracted through the center of the coring tool, the coring tool then driven to the depth of the sample required. Samples were removed and sent to the laboratory for radionuclide analysis.

Trenching and horizontal coring were done as follows. A trench was dug down to the basalt level. The wall face was gently scraped to remove surface contamination that may have been deposited during the digging process. Horizontal coring was then accomplished by driving a 5-cm-diameter, 30-cm-long hollow pipe into the wall. Core samplings were normally spaced 15 cm apart starting at the base of the trench.

Core sampling of the basalt regions was done as follows. After cleaning of the surface area of the basalt, an air hammer was used to first spall the top 2-3 cm of basalt and then to drill a hole in 15-cm increments to a depth of up to 76 cm. Samples were taken from each 15-cm-thick segment of crusted rock and sent to the laboratory for radionuclide analysis.

In all of the above sampling procedures great care was used to minimize cross contamination. Most soil samples ranged from 50-500 g in size.

ANALYSIS METHODS

Laboratory and *in situ* analyses were performed to establish the concentrations of the alpha-, beta-, and gamma-emitting isotopes in the soil samples.

Alpha Analysis

Three different techniques were used: (1) A general survey of the area was done using a hand-held proportional counter. (2) Samples were prepared for gross counting by sieving and leaching 10 g of soil. The sample was then gross counted using a gas-flow proportional counter. (3) Ten grams of soil were fused so that plutonium and americium isotopes could be extracted from the sample. Isotopic identification was performed using electrodeposition techniques and surface barrier spectrometry.

Beta Analysis

General mapping of the OMRE area was accomplished with hand-held end-window G.M. detectors. Laboratory analysis of the samples was carried out as follows: (1) The samples that were prepared for gross alpha analysis were also counted for beta activity by changing the operating voltage of the gas-flow proportional counter. (2) The ^{90}Sr activity was determined by fusing a 10-g soil sample, separating the strontium with wet chemistry, letting the sample decay for seven days, separating the ^{90}Y that grew into the sample from the decay of ^{90}Sr, and then counting the ^{90}Y in the gas-flow proportional counter. Yields were established by using a ^{85}Sr gamma-emitting tracer and yttrium as a carrier.

Gamma-ray Analysis

A shielded beta-gamma survey G.M. instrument was used to measure the gross gamma activity at the site area. A mobile Ge(Li) spectrometer was used to collect gamma-ray spectra at the site area. This spectrometer was calibrated to give reliable relative answers only. Quantitative measurements with this spectrometer were not attempted. Three types of laboratory analyses were performed on the samples obtained from the OMRE site: (1) Gross count using methane flow chambers, (2) Na(I) spectrometry, and (3) Ge(Li) spectrometry. Ge(Li) spectrometry was used to establish the concentration of the various gamma-ray emitting isotopes. The spectrometers were calibrated using National Bureau of Standards samples which were distributed uniformly in various soil samples. From these calibrations absolute numbers could be obtained. The samples were counted as received with no sieving or drying procedures being used. Uncertainties in sampling procedures, uniformity, soil dryness, etc., can cause activity uncertainties to be as large as 200-300%. Variations of this amount are considered to be acceptable.

Natural Backgrounds

The thorium (^{232}Th) and uranium (^{238}U) atoms' natural radioactive decay set off a series of nuclear transformations which are

called the Th-U daughter series. Emission of an alpha particle from ^{238}U transforms the atom into ^{234}Th which is the beginning of a long chain of naturally occurring radioactivities. Likewise, the natural emission of an alpha particle from ^{232}Th produces ^{228}Ra which also starts the production of a long chain of other radioactive isotopes. Potassium (^{40}K) is also a naturally occurring radioactive isotope. The ^{40}K produces about 85% and the Th-U daughters 15% of the natural gamma-ray background in the vicinity of the OMRE site. There are also naturally occurring beta emitters such as tritium (^{3}H), ^{14}C, and ^{87}Rb that add to the natural background. At the INEL, the natural background radioactivity in the soil is approximately 63 pCi/g (24 pCi/g due to alpha-emitting and 16 pCi/g due to beta-gamma-emitting isotopes in the Th-U chain, 19 pCi/g due to ^{40}K, and about 4 pCi/g due to pure beta-emitting isotopes). There is also a man-made background, mostly ^{137}Cs and ^{90}Sr, that is produced from weapons tests which amounts to about 2-3% of the naturally occurring background at the INEL.

These naturally occurring backgrounds sometimes make measurement for the man-made isotopes difficult to perform.

RESULTS

Reactor Area

Since the OMRE reactor was located below ground with little shielding between the core and the surrounding soil, this soil became radioactive by neutron-gamma reactions. The majority of this soil was removed to the waste storage area. Approximately 150 samples taken from the remaining soil and from the basalt base were analyzed for radionuclide concentrations. Table I shows a summary of the concentrations observed. The primary radionuclides observed in the soil samples taken from the reactor area were ^{60}Co, ^{152}Eu and ^{154}Eu. Fission product concentrations were established to be < 3 pCi/g of soil around the reactor pit perimeter and in the basalt that supported the base of the reactor. Small quantities of ^{137}Cs (< 3 pCi/g) and ^{90}Sr (< 7 pCi/g) were observed in some of the soil that was collected above the basalt base. Off-site backgrounds for these two isotopes are approximately 1.0 and approximately 0.5 pCi/g, respectively.

Additional soil and basalt is being removed from the reactor pit area so that soil activities will meet the criterion of < 1 nCi/g. When the < 1 nCi/g criterion has been met, the pit will be backfilled with clean soil. The area will then be restored to its original state by grading and seeding with native grass.

Table I. A Summary of the Radionuclide Concentrations Observed in Soil Sources Taken at the OMRE Reactor Area Site

Location	pCi/g					Beta-Gamma Natural Background
	^{60}Co	^{90}Sr	^{137}Cs	^{152}Eu	^{154}Eu	
Perimeter	0.6-2	0.4	0.4-3	0-5	<0.5	~39
Soil Above Basalt (0-61 cm)	20-370	6.4	0-3	9-960	6-70	~39
Basalt (0-61 cm)	10-210	---	0-0.2	26-650	2-50	~39
Off Site	<0.1	~0.5	~1	<0.3	<0.03	~39

An *in situ* survey of the reactor pit area and the fill dirt was carried out to verify that the gamma-ray-emitting radionuclides measured in the soil samples were the only ones present in the area (excluding natural radioactivity). This was done utilizing a motorized laboratory containing a high-resolution Ge(Li) gamma-ray detector. Figure II shows the mobile laboratory with the Ge(Li) detector positioned on the edge of the pit and viewing the general pit area.

Results of the *in situ* measurements (see Table II) indicate that 99% of the gamma-ray activity in the fill dirt is due to natural backgrounds of ^{40}K and Th-U daughters and 1% is due to ^{137}Cs. Approximately 75% of the gamma-ray activity in the reactor pit was found to be from ^{152}Eu and ^{154}Eu, 18% from ^{60}Co, 1% from ^{134}Cs (the ^{134}Cs activity came from the neutron-gamma reaction on ^{133}Cs), and 6% from ^{40}K. The Ge(Li) mobile spectrometer was also used to measure the activities in other areas: 3 m back from the edge of the reactor pit, the cement dirt pile, and an area approximately 2 km from the OMRE site (at the OMRE-PBF road junction). These results are also shown in Table II.

Leach Pond Area

The OMRE leach pond is a trench situated on natural basalt bedrock. The pond size at the berm (brim of the trench) is approximately 8 m wide by 22 m long. The sides slope to a base approximately 5 m wide by 15 m long. The depth of the soil above

Table II. Results of the *in situ* Ge(Li) Gamma-ray Spectral Measurements Taken at the OMRE Area

	Percentage of Activity					
	Full View Reactor Pit Base	3 m Back Reactor Pit Edge	Cement-Dirt Pile	Edge Leach Pond	Fill Dirt Pile	OMRE-PBF Road Junction
^{40}K	6	11	71	24	83	86
Th-U Dau.	--	--	27	3	16	14
^{60}Co	18	20	--	21	--	--
^{134}Cs	1	--	--	--	--	--
^{137}Cs	--	--	1	52	1	--
^{152}Eu	69	63	1	--	--	--
^{154}Eu	6	6	--	--	--	--

the basalt base varies from 30 to 46 cm. Figure III shows the leach pond with the mobile Ge(Li) detector being set into position to measure the gamma-ray field.

Approximately 200 samples were obtained from the leach pond area and analyzed for their radionuclide content. Only four radionuclides (^{60}Co, ^{90}Sr, ^{137}Cs, and traces of ^{152}Eu) were detected in samples from the pond. The small traces of ^{152}Eu were observed only in surface samples. A compilation of the radionuclide concentrations measured in the soil samples is given in Table III. The ^{60}Co concentrations ranged from approximately 1 pCi/g at the pond perimeter to 1500 pCi/g at the basalt surface. The ^{137}Cs concentrations ranged from approximately 1 pCi/g at the perimeter to 6700 pCi/g at the basalt surface. The average concentrations of the four radionuclides and an estimate of their total activities in the leaching pond are given in Table IV.

Selected samples were analyzed for alpha-emitting radionuclides. The results indicated that the gross alpha-emitting radionuclide concentration (including the natural Th-U chain) is

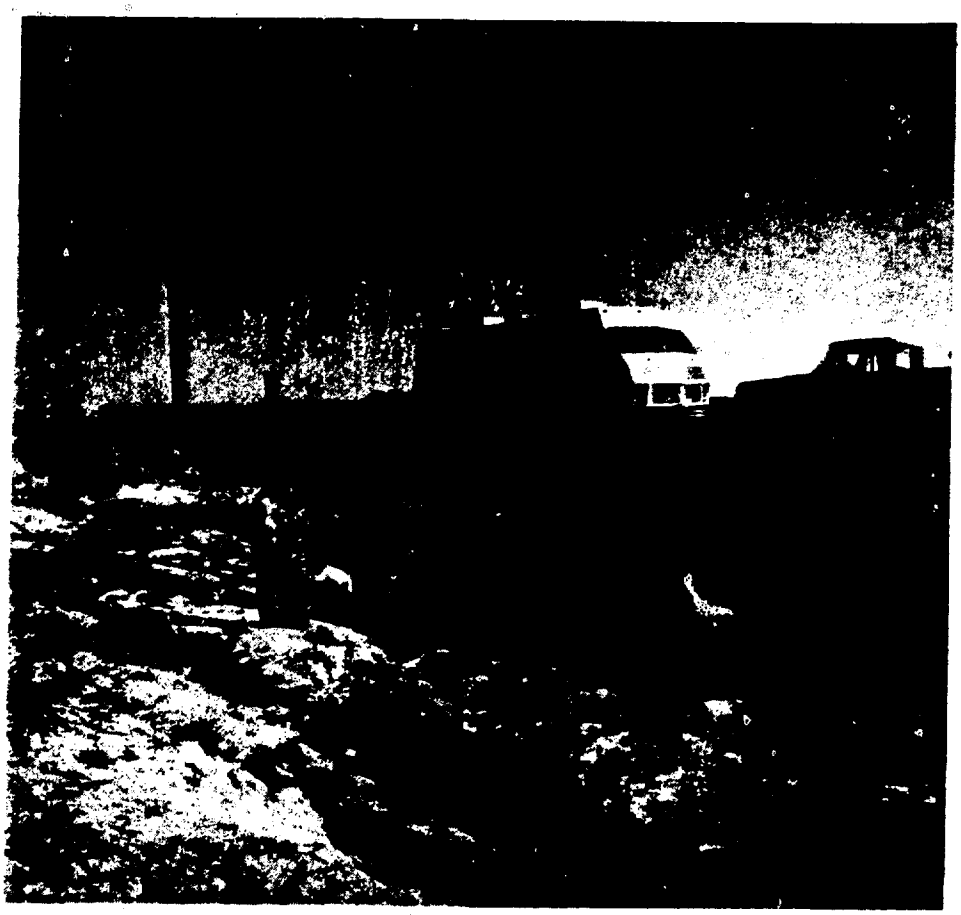

Figure III. The OMRE leach pond. The pond is approximately 8 m wide and 22 m long. The Ge(Li) gamma detector is shown in the background at the doorway of the motor home where the electronic equipment is housed.

Table III. A Summary of the Radionuclide Concentrations in Soil Samples Taken from the OMRE Leach Pond

Location	pCi/g				Beta-Gamma Natural Background
	^{60}Co	^{90}Sr	^{137}Cs	^{152}Eu	
Perimeter	0.3-3.0	--	0.2-4.0	0-0.5	~39
Berm	2-220	2-43	1-425	0-3	~39
Base Soil	3-1500	6-650	3-6700	0-19	~39
Basalt (0-15 cm)	0.4-9.0	--	0.3-26.0	<0.3	~39
Off-Site	<0.1	~0.5	~1.0	<0.3	~39

Table IV. The Average Concentration of the Four Radionuclides and an Estimate of their Total Activity for the OMRE Leach Pond

Isotope	Half Life	Activity			
		Total (mCi)	Berm Area (mCi)	Pond Base (mCi)	Average (pCi/g)
^{60}Co	5.27 yrs	22	9	13	151
^{90}Sr	29.1 yrs	~18	~7	~11	~123
^{137}Cs	30.0 yrs	36	14	22	246
^{152}Eu	13.33 yrs	0.3	0.2	0.1	1.4

Note: 220,000 kg soil in berm
62,000 kg soil in base

< 20 pCi/g. Concentrations for specific radionuclides are < 0.04 pCi/g for ^{238}Pu, < 0.04 pCi/g for $^{239,240}Pu$, and < 0.01 pCi/g for ^{241}Am.

Results of the in situ measurements obtained using the mobile Ge(Li) spectrometer (see Table II) indicate that about 50% of the gamma-ray activity in the leach pond is due to ^{137}Cs, 20% is due to ^{60}Co, and almost 30% is due to Th-U daughters.

Figure IV shows how the average concentrations of the detected radionuclides will drop off as a function of time. These concentrations are compared to the INEL D&D criterion (< 1 nCi/g), INEL natural background (mainly ^{40}K and daughters of Th-U), ^{137}Cs and ^{90}Sr background (from weapons tests), and ^{60}Co and ^{152}Eu INEL backgrounds.

CONCLUSIONS

Results of this study indicate that the activity at the OMRE decommissioned area is confined to localized areas (i.e., the leach pond area and reactor area). Comparisons of radionuclide concentrations measured in soil taken from the lip of the leach pond with concentrations in soil obtained outside the INEL site boundaries indicate that the concentration in the soil at the edge of the leach pond is at background levels.

The vertical augering technique was determined to be the best approach for obtaining shallow soil samples at the INEL. Selection of this technique was based on ease of operation and analytical results. Less area is disturbed per sample than with the horizontal trenching and coring techniques.

The radionuclide analysis of the samples presented in this report shows the existence of a few regions in the reactor and leach pond areas that were still above INEL release criteria. These regions have been or are being further decontaminated.

The following specific conclusions are worthy of note.

Reactor Area

1. Major radionuclides came from neutron activation of the soil (^{60}Co, ^{152}Eu and ^{154}Eu). Highest concentrations were in the soil samples taken above the basalt base.

2. Concentrations of the above isotopes are less than INEL D&D criterion (< 1 nCi/g).

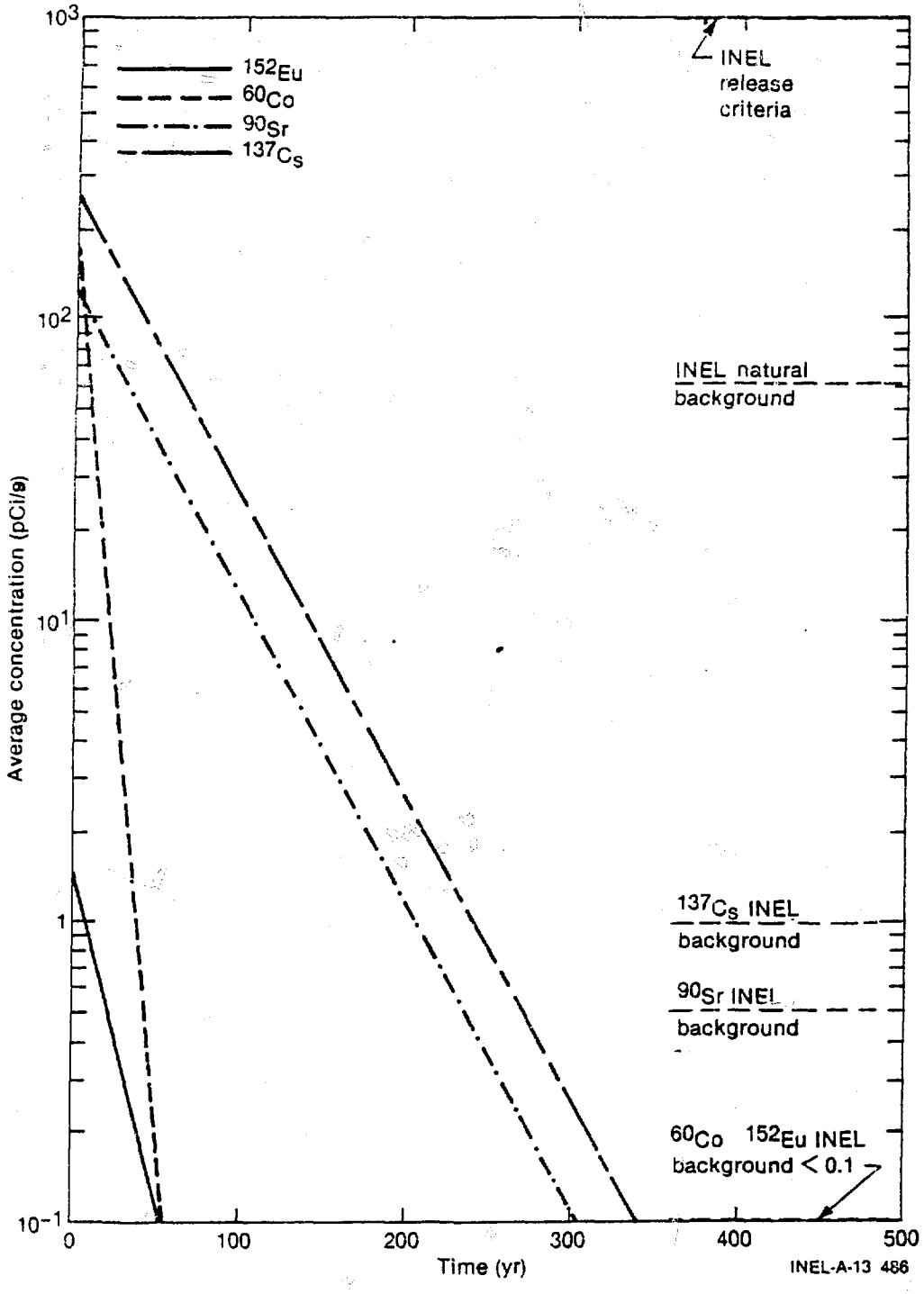

Figure IV. The OMRE leach pond average radionuclide concentrations versus decay time. These values are compared to the INEL D&D criteria.

3. No significant amounts of fission products were observed in the reactor area (< 3 pCi/g).

4. The reactor perimeter area including fill dirt had radionuclide concentrations at essentially background levels.

5. Basalt core samples taken from a depth of 30-60 cm showed ^{60}Co concentrations < 10 pCi/g, ^{137}Cs < 0.2 pCi/g, ^{152}Eu < 28 pCi/g, and ^{154}Eu < 3 pCi/g.

Leach Pond Area

1. The major radionuclides observed were ^{137}Cs, ^{60}Co, and ^{90}Sr. Only trace quantities of ^{152}Eu were measured.

2. The highest contamination was located in the soil next to the basalt surface.

3. Radiation readings taken on a few basalt core samples indicate extremely low level contamination in the upper 15 cm of the basalt (< 26 pCi/g of ^{137}Cs and < 9 pCi/g of ^{60}Co).

4. Analysis of samples taken at the perimeter of the pond show activities at essentially background levels.

5. The concentration of transuranic radionuclides was measured to be < 0.1 pCi/g. This is three orders of magnitude below the INEL D&D criterion.

ACKNOWLEDGEMENTS

The authors would like to express their appreciation to E. D. Cadwell and C. L. Rowsell for their assistance and direction of the Radiation Measurements Laboratory personnel who did the Ge(Li) gamma-ray spectral analysis of the many soil samples. Thanks are also given to R. A. Stevenson, J. L. Alvarez and J. F. Sommers for their support and recommendations. Appreciation is also expressed to C. W. Filby who directed the beta and alpha analyses that were performed by the Radioanalysis Group of Exxon Nuclear Idaho Corp.

REFERENCES

1. J. A. Chapin and R. E. Hine, Decontamination and Decommissioning Long Range Plan, Idaho National Engineering Laboratory, EG&G Idaho, Inc., TREE-1250 Vol. 1, June 1978.