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# The Northern Marshall Islands Radiological Survey: A Quality Control Program for Radiochemical Analyses

C. D. Jennings

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August 1983

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C. D. Jennings\*

M. E. Mount

Manuscript date: August 1983

LAWRENCE LIVERMORE NATIONAL LABORATORY  
University of California · Livermore, California · 94550



\* Oregon State University, College of Oceanography  
Corvallis, Oregon 97331

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# THE NORTHERN MARSHALL ISLANDS RADIOLOGICAL SURVEY: A QUALITY CONTROL PROGRAM FOR RADIOCHEMICAL ANALYSES

## ABSTRACT

More than 16,000 radiochemical analyses were performed on about 5,400 samples of soils, vegetation, animals, fish, invertebrates, and water to establish amounts of  $^{90}\text{Sr}$ ,  $^{137}\text{Cs}$ ,  $^{241}\text{Am}$ , and plutonium isotopes in the Northern Marshall Islands. Three laboratories were contracted by Lawrence Livermore National Laboratory to perform the radiochemical analyses: Environmental Analysis Laboratory (EAL), Richmond, California; Eberline Instrument Corporation (EIC), Albuquerque, New Mexico; and Laboratory of Radiation Ecology (LRE), University of Washington, Seattle, Washington. The analytical precision and accuracy were monitored by regularly including duplicate samples and natural matrix standards in each group of about 100 samples analyzed. Based on the duplicates and standards, over 83% of the radiochemical analyses in this survey were acceptable--97% of the analyses by EAL, 45% of the analyses by EIC, and 98% of the analyses by LRE.

## INTRODUCTION

The Northern Marshall Islands Radiological Survey (NMIRS) was a large-scale effort to collect soil, vegetation, animal, fish, invertebrate, and water samples and assess the radiation dose from the ingestion and inhalation pathway and external environments of 12 atolls and 2 islands in the Northern Marshall Islands. The Lawrence Livermore National Laboratory (LLNL) was responsible for this effort, including the collection and processing of the various samples.<sup>1</sup> Overall, about 5,400 samples were collected and over 16,000 radiochemical analyses were performed to establish the amounts of specific radionuclides in the soils, vegetation, animals, fish, invertebrates, and water. For such an extensive analytical program, no one laboratory had the capability to analyze all the samples in a reasonable time. Accordingly, three laboratories were contracted to perform the required analyses.

Any analytical program depends on the quality of the measurements being made. Most laboratories spend a certain part of their effort to establish the accuracy and reproducibility of their analytical work. Blind interlaboratory comparisons such as the Department of Energy (DOE) intercalibration exercise<sup>2</sup> and analysis of natural matrix

standards such as those available from the National Bureau of Standards and from the International Atomic Energy Agency are two methods by which the quality of a laboratory's work can be shown. In a large-scale survey such as the Northern Marshall Islands program where samples are analyzed by several laboratories, it is all the more important to assess the validity of the data by regularly having the participating laboratories analyze blind quality-control (QC) standards.

For this program we have selected three criteria for the analytical reliability of the data.

- (1) The first criterion places limits of acceptability on counting errors. Because radioactive decay is a statistical process, sufficient counts must be collected to provide a level of confidence that the number reported is a true measure of the radioactivity of the sample. Until this criterion is met it is difficult, if not impossible, to evaluate the data for the remaining two criteria. Consequently, we established a set of acceptable counting errors (Table 1). The requirements were scaled to the total radioactivity of the sample, which is the product of the amount of sample available and its specific activity (activity per unit weight of sample). Compliance could be easily checked by the individual analyst because it is based on information available to him: the measured specific activity and weight of the sample received. This criterion was developed prior to initiation of the NMIRS field-sample collection program to estimate the amount of samples required by any competent contractor to measure worldwide fallout. Samples of sufficient size with higher activity were thus well above the limits of detection of the contracting laboratories. This was done to avoid reporting machine limits that give only upper limits to the concentrations of the samples and thus will overestimate the amount of radioactivity actually present in the environment when these limits are used as real values. This is not an uncommon practice when assessing environmental data.
- (2) The second criterion required that the laboratories reproduce their results on replicate analyses. A set of blind duplicate samples was included with each group of roughly 100 samples (called DCD for the accompanying Delivery Control Document) and results of the pair of analyses were considered acceptable if they agreed within twice the measurement accuracy required in Table 1. Satisfactory performance on duplicates required acceptability on 80% of all duplicate pairs included in each DCD. Duplicate samples were prepared and distributed by LLNL.

Table 1. Allowable counting errors according to the total activity in the sample received by the contracting laboratories.

Radionuclide	pCi	1 $\sigma$ error (%)
$^{239,240}\text{Pu}$	< 0.1	50 to 100
	0.1 to 0.25	<u>&lt;40</u>
	0.25 to 1	<u>&lt;20</u>
	> 1	<u>&lt;10</u>
$^{238,241}\text{Pu}$	< 2	50 to 100
	2 to 5	<u>&lt;40</u>
	5 to 20	<u>&lt;20</u>
	> 20	<u>&lt;10</u>
$^{241}\text{Am}$	< 0.1	50 to 100
	0.1 to 0.25	<u>&lt;40</u>
	0.25 to 1	<u>&lt;20</u>
	> 1	<u>&lt;10</u>
$^{137}\text{Cs}$	< 1	50 to 100
	1 to 5	<u>&lt;30</u>
	5 to 8	<u>&lt;20</u>
	> 8	<u>&lt;10</u>
$^{90}\text{Sr}$	< 1	50 to 100
	1 to 5	<u>&lt;30</u>
	5 to 8	<u>&lt;20</u>
	> 8	<u>&lt;10</u>

- (3) The third criterion required that the laboratories accurately determine the radionuclide concentrations of blind standards. Although all three of the criteria are important, perhaps this is the most significant because it includes accuracy of measurement as well as precision (reproducibility); any systematic errors in the measurement would appear. Responsibility for preparing,

standardizing, and distributing the standard samples was assigned to Western Oregon State College (WOSC). In this way the primary responsibility for evaluating the analytical integrity of the data was vested in a disinterested party.

In some cases, small variances from these criteria were allowed to facilitate processing the data, but the deviations were never great enough to compromise the integrity of the data. Specifically, the error requirements shown in Table 1 were relaxed by 20% for some of the early DCDs to accommodate problems some of the contractors had in reducing counting errors. For example, for samples where a 10% relative standard deviation was required, a 12% relative standard deviation was allowed.

Another variance was in the number of duplicates and standards that had to be in compliance. In general, for a DCD to be considered acceptable, 80% of the duplicate pairs and 100% of the standards had to be in compliance with the QC criteria. When the number of duplicate pairs did not permit exactly 80% compliance (for example, 3 of 4 pairs would give 75% compliance), a fraction of duplicates in compliance slightly less than but near 80% was still considered to be acceptable. Less than 100% compliance on standards was allowed occasionally if the radiochemical analysis on the standard was near the accepted activity and if the laboratory had established a record of accurate radiochemical analysis on other subsamples of the standard in question.

## PARTICIPATING LABORATORIES

Laboratories participating in the radiochemical analyses of samples from the NMIRS were Environmental Analysis Laboratory (EAL), Richmond, California; Eberline Instrument Corporation (EIC), Albuquerque, New Mexico; and Laboratory of Radiation Ecology (LRE), University of Washington, Seattle, Washington.

The radionuclides measured were  $^{90}\text{Sr}$  (beta counting),  $^{137}\text{Cs}$  (beta and gamma counting),  $^{239+240}\text{Pu}$  and  $^{238}\text{Pu}$  (alpha pulse-height analysis),  $^{239}\text{Pu}$  and  $^{240}\text{Pu}$  (mass spectrometry),  $^{241}\text{Pu}$  (mass spectrometry and liquid scintillation counting), and  $^{241}\text{Am}$  (alpha pulse-height analysis). There were 16,282 analyses, including the duplicate and standard samples of the QC program, requested of the three participating laboratories. The largest fraction of the analyses was performed by EAL: 65.6% (42.3% terrestrial and 23.3% marine). Slightly over one-fourth of the analyses, 25.5%, was performed by EIC. The balance, 8.9%, was analyzed by LRE.

Table 2 summarizes the duplicate and standard analyses associated with the 15,745 analyses of soil, vegetation, terrestrial animal, marine organisms, and marine sediment

Table 2. Summary of the duplicate and standard analyses evaluated for each participating laboratory as part of the quality control program for the Northern Marshall Islands Radiological Survey.

Laboratory	Sample type	$^{90}\text{Sr}$	$^{137}\text{Cs}$	$^{239+240}\text{Pu}$	$^{238}\text{Pu}$	$^{239}\text{Pu}$	$^{240}\text{Pu}$	$^{241}\text{Pu}$	$^{241}\text{Am}$	TOTAL
EAL <sup>a</sup>	Soil									
	Duplicate	140	84	140	17	31	31	25	114	582
	Standard	22	22	22	--	--	--	--	22	88
	Vegetation									
	Duplicate	44	26	42	--	--	--	--	44	156
	Standard	13	13	13	--	--	--	--	13	52
	Terrestrial animal									
	Duplicate	6	6	6	--	--	--	--	6	24
	Standard	6	6	6	--	--	--	--	6	24
	Marine organism									
	Duplicate	71	--	70	70	--	--	--	71	282
	Standard	11	6	11	--	--	--	--	11	39
	Marine sediment									
Duplicate	7	--	7	7	--	--	--	7	28	
Standard (soil)	4	2	4	--	--	--	--	4	14	
SUBTOTAL		324	165	321	94	31	31	25	298	1289
EIC <sup>b</sup>	Soil									
	Duplicate	60	31	71	7	--	--	22	52	243
	Standard	30	30	33	--	--	--	--	32	125
	Vegetation									
	Duplicate	16	15	20	--	--	--	--	21	72
Standard	3	3	5	--	--	--	--	7	18	
SUBTOTAL		109	79	129	7	--	--	22	112	458
LRE <sup>c</sup>	Soil									
	Duplicate	63	28	63	--	--	--	18	17	189
	Standard	9	9	9	--	--	--	--	9	36
SUBTOTAL		72	37	72				18	26	225
TOTAL		505	281	522	101	31	31	65	436	1972

<sup>a</sup> Environmental Analysis Laboratory.

<sup>b</sup> Eberline Instrument Corporation.

<sup>c</sup> Laboratory of Radiation Ecology.

evaluated as part of the NMIRS QC program. The 537 water analyses were evaluated and accepted separately.<sup>3</sup> Of the 15,745 analyses evaluated herein, 12.5% were associated with the QC program. On an individual laboratory basis, QC program analyses accounted for 12.7% of EAL's, 11% of EIC's, and 15.6% of LRE's evaluated analyses.

## PREPARATION OF STANDARDS AND DUPLICATES

### PREPARATION OF STANDARDS

The samples from the survey included soil and marine sediments, plant material, marine tissue, and terrestrial animal tissue, and we prepared standards of each of the four natural matrixes. Because the purpose of the standards was to substantiate the analyses of other environmental samples, the character of the standards matched that of the samples as closely as possible. The radioactivity in the standards was from the natural environment and not merely added to the samples from a solution in which the chemical forms of radioactivity might be quite different from those in the samples. Consequently, each standard was prepared from material collected directly from the environment.

Environmental samples of vegetation and marine or terrestrial animal tissues generally did not have enough radioactivity to serve as adequate standards. These samples were spiked with environmentally labeled radioactive algae--the same algae used by Volchok and Feiner at the DOE Environmental Measurements Laboratory (EML) to prepare standards for the DOE intercalibration exercise.<sup>2</sup>

#### Soil Standard

The soil standard was collected at the Marshall Islands and a large sample was shipped to LLNL where it was dried and ball milled. It was then shipped to WOSC where it was sieved and the entire sample was blended in a large twin-cone blender. The homogenized soil was canned and labeled to conform to the style of samples prepared at LLNL. This standard was also used for the marine sediment standard.

#### Vegetation Standard

To prepare the vegetation standard, we added a known amount of algae to commercial potato flakes, blended it in a twin-cone blender, and then it was canned and labeled.

### Terrestrial Animal Standard

A large sample of beef was dried and ashed at LLNL and shipped to WOSC. The ash was spiked with a known amount of algae, blended, and reashed at 400°C. The reashed beef was blended in a small twin-cone blender, canned, and labeled.

### Marine Organism Standard

A large fish sample was collected and ashed at LLNL and then shipped to WOSC. The ashed sample was spiked with a known amount of algae, blended, and then reashed at 400°C. The reashed sample was then blended in a small twin-cone blender, canned, and labeled.

### CERTIFICATION OF STANDARDS

The first step in certifying the standards was to show that the methods used to prepare the samples produced a homogeneous sample. Because the vegetation standard was the first prepared, the greatest attention was given to it. Homogeneity was first tested by measuring  $^{137}\text{Cs}$  in a random selection of ten aliquots of the standard by nondestructive analysis on a Ge(Li) gamma-ray spectrometer. Because the samples were not in the counting geometry normally used by WOSC, only relative activities were measured. As shown in Table 3, there was excellent agreement among the ten samples. Similarly, the 2.6% relative standard deviation of 10 measurements of  $^{137}\text{Cs}$  done radiochemically and the 4.7% relative standard deviation of 16 measurements of  $^{239+240}\text{Pu}$  (Table 4) provide evidence that the procedures used to prepare the samples produced a homogeneous sample. Because the marine and terrestrial animal standards were prepared in the same manner as the vegetation standard by adding algae and blending, their homogeneity is likewise established. In the course of this survey, the homogeneity of the vegetation and animal samples was confirmed by one of the participating laboratories (EAL). Because the soil standard had no added radioactivity, it was a slightly different case, but the 5.4% standard deviation about the mean of ten  $^{239+240}\text{Pu}$  analyses shows that it was likewise homogeneous (Table 5). Moreover, many analyses by the participating laboratories have further confirmed its homogeneity.

Certified activities of the standards were established in two ways. First, the radioactivity of the algae used to spike samples of vegetation, fish, and beef had been measured at EML, and because it was used to prepare samples for the DOE

Table 3. The  $^{137}\text{Cs}$  measured by gamma-ray spectrometry in a random selection of vegetation standard samples.

Sample	$^{137}\text{Cs}$ (cpm/100 g) <sup>a</sup>	Deviation from mean (%)
1	4.996 ± 0.084	2.65
2	4.863 ± 0.091	0.08
3	4.768 ± 0.104	2.03
4	4.864 ± 0.09	0.06
5	4.675 ± 0.092	3.94
6	4.937 ± 0.096	1.44
7	4.805 ± 0.09	1.27
8	4.964 ± 0.06	1.99
9	4.918 ± 0.106	1.05
10	4.881 ± 0.098	0.29
MEAN	4.867 ± 0.097	

<sup>a</sup> Only relative counts were measured because samples were not in the counting geometry we normally used.

Table 4. Radionuclides measured in vegetation standard samples. All analyses done radiochemically except as noted.

Values or laboratory	Radionuclide (pCi/kg)			
	$^{90}\text{Sr}$	$^{137}\text{Cs}$	$^{239+240}\text{Pu}$	$^{241}\text{Am}$
Expected	--	2710 ± 20%	17.2 ± 20%	4.1 ± 20%
WOSC, OSU <sup>a</sup>	3780 ± 8% (4)	2730 ± 2.6% (10)	16.2 ± 4.7% (16)	5.4 ± 1.7% (4)
EML <sup>b</sup>	3340 ± 5.8% (3)	2390 ± 11.7% (3) <sup>c</sup>	14.9 ± 10% (3)	4.8 ± 23% (3)
LLNL <sup>d</sup>	--	2610 ± 2% (2) <sup>c</sup>	15.8 ± 2% (2)	--
Certified	3340 ± 10%	2700 ± 10%	16 ± 10%	5.4 ± 10%

NOTE: Numbers of replicates are in parentheses.

<sup>a</sup> Western Oregon State College, Oregon State University.

<sup>b</sup> Environmental Measurements Laboratory.

<sup>c</sup> Measured by gamma-ray spectrometry.

<sup>d</sup> Lawrence Livermore National Laboratory.

Table 5. Radionuclides measured in soil standard samples. All analyses done radiochemically except as noted.

Values or laboratory	Radionuclide (pCi/kg)			
	$^{90}\text{Sr}$	$^{137}\text{Cs}$	$^{239+240}\text{Pu}$	$^{241}\text{Am}$
WOSC, OSU <sup>a</sup>	--	$35.8 \pm 3.8\%$ (8)	$36.4 \pm 5.4\%$ (10)	$15.5 \pm 7.2\%$ (4)
EML <sup>b</sup>	$88.7 \pm 4.1\%$ (3)	$36.9 \pm 3.7\%$ (3) <sup>c</sup>	$34.8 \pm 9.8\%$ (3)	$14 \pm 7\%$ (3)
LLNL <sup>d</sup>	--	--	$37.4 \pm 0.6\%$ (2)	--
Certified	$89 \pm 10\%$	$36 \pm 10\%$	$36 \pm 10\%$	$16 \pm 10\%$

NOTE: Numbers of replicates are in parentheses.

<sup>a</sup> Western Oregon State College, Oregon State University.

<sup>b</sup> Environmental Measurements Laboratory.

<sup>c</sup> Measured by gamma-ray spectrometry.

<sup>d</sup> Lawrence Livermore National Laboratory.

intercalibration exercise, the measurements were well corroborated. These known additions of radioactivity were used to establish the expected radioactivity. Second, all the standards except the terrestrial animal standard were analyzed radiochemically. The results of these analyses are shown in Tables 4-6. The laboratories at WOSC and Oregon State University (OSU) have collaborated for several years and their results are combined. Corroborating analyses were performed by EML and LLNL. The radiochemical methods at WOSC and OSU have been demonstrated for the measurement of  $^{137}\text{Cs}$ ,  $^{239+240}\text{Pu}$ , and  $^{241}\text{Am}$  (see Ref. 4), and the measurements of those three radionuclides were taken as the certified values. Because the WOSC, OSU measurement of  $^{90}\text{Sr}$  had not been demonstrated, we used the analyses of EML to certify the  $^{90}\text{Sr}$ . As Tables 4-6 show, excellent agreement was found for all radionuclides. In Table 7 we present only the expected values for the terrestrial animal standard because it was prepared by the same method as the vegetation and marine organism standards by adding known amounts of the radionuclides. Consequently, no radiochemical analyses were required. We are confident that the certified activities, or expected values in the case of the terrestrial animal standard, are correct within the tolerance quoted in Tables 4-7. This has been further demonstrated by the high degree of compliance by two of the three participating laboratories.

Table 6. Radionuclides measured in marine organism standard samples. All analyses done radiochemically except as noted.

Values or laboratory	Radionuclide (pCi/kg)			
	<sup>90</sup> Sr	<sup>137</sup> Cs	<sup>239+240</sup> Pu	<sup>241</sup> Am
Expected	16.7 ± 20%	10.8 ± 20%	68.9 ± 20%	16.8 ± 20%
WOSC, OSU <sup>a</sup>	16.9 ± 20% (3)	12.1 ± 4.1% (3)	77.5 ± 10% (3)	16.7 ± 10% (2)
LLNL <sup>b</sup>	--	--	78.8 ± 5.7% (2)	17.5 ± 12% (2) <sup>c</sup>
Certified	17 ± 20%	12 ± 10%	78 ± 10%	17 ± 10%

NOTE: Number of replicates are in parentheses.

<sup>a</sup> Western Oregon State College, Oregon State University.

<sup>b</sup> Lawrence Livermore National Laboratory.

<sup>c</sup> Measured by gamma-ray spectrometry.

Table 7. Expected values of radionuclides in terrestrial animal standard samples (pCi/kg).

<sup>90</sup> Sr	<sup>137</sup> Cs	<sup>239+240</sup> Pu	<sup>241</sup> Am
67 ± 20%	48 ± 20%	0.3 ± 20%	0.074 ± 20%

#### PREPARATION OF DUPLICATES

The preparation of duplicate samples was simple and straightforward. The primary requirement was a processed sample of sufficient size (volume) to provide two aliquots for comparative radiochemistry. Once the aliquots were made, the samples were packaged in suitable containers (aluminum bean or tuna cans, plastic vials, paper cartons, etc.), labeled, and forwarded for analysis.

The procedures used at LLNL to process the various sample types are described in detail in a previous paper of this series.<sup>1</sup> For completeness, the procedures appropriate to the duplicates are briefly summarized.

### Soil Duplicates

Each soil sample (500 to 900 g) was placed in a 1-gal can. It was then dried for 48 h at 75°C, weighed, and redried for an additional 24 h and then reweighed. If constant weight was noted, it was considered dry. If not, it was returned to the oven for an additional 24 h. Once dry, eight 1-in. steel grinding balls were placed in the can with soil, the cover securely sealed, and the samples ball milled continuously for 48 h. After ball milling, the necessary number of aliquots was canned, labeled, and forwarded for analysis. Soil duplicates represented the largest fraction of duplicates prepared.

### Vegetation Duplicates

Vegetation duplicates were almost exclusively prepared from composite coconut meat samples. All vegetation was maintained frozen at LLNL until processed. To ensure against contamination, fruits and roots were washed very carefully before dissection. Once the samples were dissected into their various segments (i.e., meat, skin, and seeds), the segments were placed in plastic containers and weighed. Following weighing, the samples were freeze-dried and reweighed. The dry vegetation material was then ground to a homogeneous texture in Waring blenders, appropriate aliquots were taken and pressed into aluminum tuna or bean cans until a uniform density was achieved, the cans labeled as required, and then forwarded for analysis.

### Terrestrial Animal Duplicates

Terrestrial animal duplicates were prepared from various parts of a pig; the hindquarter being used the most. Processing procedures were the same for the animal samples as for the vegetation with one exception: formaldehyde was pipetted into the can after the sample was pressed. After sealing, the cans were appropriately labeled and forwarded for analysis.

### Marine Organism Duplicates

Marine organism duplicates were prepared from various tissues and organs of fish and clams. After dissecting, tissues and organs of a species from the same catch were pooled. Wet weights were determined and then the samples were dried to constant weight in ovens at 90°C. Following drying, the samples were dry ashed in muffle furnaces at

450°C for approximately 72 h. The ash was then homogenized and the necessary aliquots were packaged, labeled, and forwarded for analysis.

### Marine Sediment Duplicates

Marine sediment samples were processed somewhat similar to soil. After being wet weighed, they were dried in ovens at 90°C, reweighed, and then homogenized with a shaker-type ball mill. Aliquots were taken, the samples appropriately packaged and labeled, and then forwarded for analysis.

## EVALUATION OF DATA

A high degree of compliance with our QC criteria was achieved in this project. Over 83% of the samples analyzed were found acceptable for dose calculations. As shown in Table 8, 97% of the 10,685 analyses requested of EAL, 45% of the 4,152 analyses requested of EIC, and 98% of the 1,445 analyses requested of LRE were accepted. The reproducibility of the analyses is particularly apparent in Figs. A1-A45 (Appendix) that show most of the data clustered about an ideal line. The DCDs that are unacceptable do not meet the QC standards and are not certified to be used for dose assessment.

Table 8. Summary of the Northern Marshall Islands Radiological Survey radiochemical analyses.<sup>a</sup>

Laboratory	Number requested	Number accepted
Environmental Analysis Laboratory	10,685	10,379
Eberline Instrument Corporation	4,152	1,863 <sup>b</sup>
Laboratory of Radiation Ecology	1,445	1,410
TOTAL	16,282	13,652

<sup>a</sup> Includes duplicates and standards.

<sup>b</sup> Approximately 38% performed by Environmental Analysis Laboratory.

The results of the QC analyses used to evaluate the data are presented as follows. (1) Summaries of the acceptable data by laboratory, nuclide, and DCD for each environmental matrix are shown in Tables 9-16. The unacceptable data are similarly presented in Tables 17-20. (2) Shown in Figs. A1-A37 are the duplicate pairs plotted against each other for acceptable DCDs. In these figures, the broken line represents duplicates that are in perfect agreement. Solid symbols depict duplicates that overlap at  $2\sigma$ ; open symbols depict duplicates that do not overlap at  $2\sigma$ . Figures A38-A45 present duplicate pairs from unacceptable DCDs. (3) Finally, the actual data for all the duplicates and standards from acceptable DCDs are given in the attached microfiche.

When sample activity is at or near background, the resultant concentration that would be calculated may be positive or negative. Situations such as this account for the negative concentrations referred to in the figures and raw data here.

## SOIL ANALYSES

Results for acceptable soil analyses are summarized in Table 9 (EAL), Table 14 (EIC), and Table 16 (LRE). The graphic representation of soil duplicate comparisons appear in Figs. A1-A11 (EAL), Figs. A24-A29 (EIC), and Figs. A33-A37 (LRE). The range of activities measured was large:  $^{90}\text{Sr}$  ranged from 0.01 to 1000 pCi/g,  $^{137}\text{Cs}$  ranged from 0.01 to 100 pCi/g, and  $^{239+240}\text{Pu}$  and  $^{241}\text{Am}$  ranged from 0.0001 to 1000 pCi/g. It was expected that the lowest activity duplicates would show the greatest differences and this can be seen in the scatter at the lower left of Figs. A5, A11, and A27. Because these samples were low in activity, they have less stringent error requirements, as shown in Table 1. Consequently, the acceptable analyses for low-activity samples deviate further from the ideal line than for high-activity samples. When dose calculations must rely in part on soil activity,<sup>5</sup> this is advantageous because those samples that have the greatest effect on dose are those in which there is the least analytical uncertainty. Taken as a group, the acceptable soil analyses have a high degree of compliance with the QC criteria, and thus we are confident that these measurements accurately reflect the radionuclide concentrations of the localities sampled.

Results of unacceptable soil analyses are summarized in Table 17 (EAL) and Table 19 (EIC) and displayed graphically in Fig. A38 (EAL) and Figs. A40-A44 (EIC). Tables 14 and 19 show DCDs that are simultaneously acceptable and unacceptable: specifically  $^{90}\text{Sr}$  and  $^{241}\text{Am}$  for DCD-28 and  $^{239+240}\text{Pu}$  and  $^{241}\text{Am}$  for DCD-2. This apparent discrepancy arose because some initial analyses by EIC did not satisfy the QC criteria and were judged unacceptable. To rectify the problem, several reanalyses were

performed. After reanalysis, a partial approval could occur because only part of the DCD was reanalyzed either by EIC or, in some cases, by EAL. When the reanalyses were successful, approval was given only to the part reanalyzed. The part of the DCD for which insufficient samples remained for reanalysis was still unacceptable.

#### VEGETATION ANALYSES

Table 10 (EAL) and Tables 15 and 20 (EIC) summarize the results of the vegetation analyses, and the graphic comparisons of duplicates are shown in Figs. A12-A15 (EAL) and Figs. A30-A32 and A45 (EIC). More than other matrices, vegetation samples approach the limits of detection of the contracting laboratories with a resultant larger discrepancy in the results of duplicate pairs. For several vegetation samples, only the limits of detectability imposed by the radiochemical methods were reported. Although these data were not plotted on the graphs, they are included in the attached microfiche. Despite some scatter that arose from the low levels of radioactivity, the radiochemical analyses of vegetation yielded an acceptable data base especially for  $^{90}\text{Sr}$  and  $^{137}\text{Cs}$ , which contribute the most radiation dose, and also for  $^{239+240}\text{Pu}$  and  $^{241}\text{Am}$ .

#### TERRESTRIAL ANIMAL ANALYSES

Results of the analyses of terrestrial animals, all performed by EAL, are presented in Table 11 and Figs. A16 and A17. Most samples contained only small amounts of radioactivity and the analyses met the QC criteria.

#### MARINE SAMPLE ANALYSES

All samples of marine organisms and sediment were analyzed by EAL and summaries of their QC performance appear in Tables 12, 13, and 18 and in Figs. A18-A23 and A39. Departures from the ideal line in Figs. A19-A21 result both from low levels of radioactivity and, in some cases, small samples of marine organisms that yield low activity per sample. As can be seen in Table 1, counting error restrictions are less stringent in such cases. Overall, the marine samples had a high level of compliance with the QC criteria.

Table 9. Acceptable quality control results for duplicate pairs and standard samples analyzed in soil by Environmental Analysis Laboratory.

Delivery control document number	Duplicate pairs	Standard samples
<u><math>^{90}\text{Sr}</math></u>		
3	11 of 12 (92%)	--
5	10 of 11 (91%)	--
8	8 of 10 (80%)	2 of 2 (100%)
9	5 of 5 (100%)	--
11	4 of 5 (80%)	1 of 1 (100%)
12	3 of 3 (100%)	2 of 2 (100%)
14	10 of 11 (91%)	2 of 2 (100%)
15	12 of 12 (100%)	2 of 2 (100%)
18	11 of 12 (92%)	1 of 1 (100%)
19	5 of 6 (83%)	0 of 1 (0%)
20	9 of 12 (75%)	1 of 1 (100%)
21	16 of 18 (89%)	2 of 2 (100%)
36	2 of 3 (67%)	2 of 2 (100%)
42	10 of 12 (83%)	3 of 3 (100%)
43	6 of 8 (75%)	2 of 3 (67%)
TOTAL	122 of 140 (87%)	20 of 22 (91%)
<u><math>^{137}\text{Cs}</math></u>		
8	10 of 10 (100%)	2 of 2 (100%)
9	5 of 5 (100%)	--
11	4 of 4 (100%)	1 of 1 (100%)
12	3 of 3 (100%)	1 of 2 (50%)
14	10 of 11 (91%)	2 of 2 (100%)
15	10 of 11 (91%)	2 of 2 (100%)
18	11 of 12 (92%)	1 of 1 (100%)
19	5 of 6 (83%)	1 of 1 (100%)
20	8 of 12 (67%)	1 of 1 (100%)
21	1 of 2 (50%)	2 of 2 (100%)
36	3 of 3 (100%)	2 of 2 (100%)
42	4 of 4 (100%)	2 of 3 (67%)
43	1 of 1 (100%)	3 of 3 (100%)
TOTAL	75 of 84 (89%)	20 of 22 (91%)
<u><math>^{239+240}\text{Pu}</math></u>		
3	11 of 12 (92%)	--
5	11 of 11 (100%)	--
8	9 of 10 (90%)	2 of 2 (100%)
9	5 of 5 (100%)	--
11	5 of 5 (100%)	0 of 1 (0%)
12	3 of 3 (100%)	2 of 2 (100%)

Table 9: (Continued)

Delivery control document number	Duplicate pairs	Standard samples
<u>239+240Pu (continued)</u>		
14	11 of 11 (100%)	2 of 2 (100%)
15	10 of 12 (83%)	2 of 2 (100%)
18	10 of 12 (83%)	1 of 1 (100%)
19	6 of 6 (100%)	1 of 1 (100%)
21	15 of 18 (83%)	2 of 2 (100%)
36	2 of 3 (67%)	2 of 2 (100%)
42	12 of 12 (100%)	3 of 3 (100%)
43	8 of 8 (100%)	3 of 3 (100%)
TOTAL	119 of 128 (93%)	20 of 21 (95%)
<u>238Pu</u>		
3	8 of 8 (100%)	--
5	4 of 5 (80%)	--
9	4 of 4 (100%)	--
TOTAL	16 of 17 (94%)	--
<u>239Pu</u>		
8	9 of 10 (90%)	--
11	4 of 4 (100%)	--
12	3 of 3 (100%)	--
14	3 of 3 (100%)	--
15	10 of 11 (91%)	--
TOTAL	29 of 31 (94%)	--
<u>240Pu</u>		
8	9 of 10 (90%)	--
11	4 of 4 (100%)	--
12	3 of 3 (100%)	--
14	3 of 3 (100%)	--
15	9 of 11 (82%)	--
TOTAL	28 of 31 (90%)	--
<u>241Pu</u>		
8	9 of 9 (100%)	--
11	3 of 3 (100%)	--
12	3 of 3 (100%)	--
14	3 of 3 (100%)	--
15	6 of 7 (86%)	--
TOTAL	24 of 25 (96%)	--

Table 9. (Continued)

Delivery control document number	Duplicate pairs	Standard samples
	<u><math>^{241}\text{Am}</math></u>	
8	9 of 10 (90%)	2 of 2 (100%)
9	5 of 5 (100%)	--
11	4 of 4 (100%)	0 of 1 (0%)
12	3 of 3 (100%)	2 of 2 (100%)
14	10 of 11 (91%)	2 of 2 (100%)
15	11 of 12 (92%)	2 of 2 (100%)
18	11 of 12 (92%)	1 of 1 (100%)
19	3 of 6 (50%)	1 of 1 (100%)
20	11 of 12 (92%)	1 of 1 (100%)
21	16 of 18 (89%)	2 of 2 (100%)
36	1 of 2 (50%)	2 of 2 (100%)
42	12 of 12 (100%)	2 of 3 (67%)
43	7 of 7 (100%)	2 of 3 (67%)
TOTAL	103 of 114 (90%)	19 of 22 (86%)

Table 10. Acceptable quality control results for duplicate pairs and standard samples analyzed in vegetation by Environmental Analysis Laboratory.

Delivery control document number	Duplicate pairs	Standard samples
	<u><math>^{90}\text{Sr}</math></u>	
1	2 of 5 (40%)	--
7	--	1 of 1 (100%)
10	7 of 8 (88%)	1 of 1 (100%)
16	--	1 of 1 (100%)
17	5 of 6 (83%)	1 of 1 (100%)
27	8 of 8 (100%)	2 of 2 (100%)
28	--	1 of 1 (100%)
38	5 of 5 (100%)	3 of 3 (100%)
39	12 of 12 (100%)	3 of 3 (100%)
TOTAL	39 of 44 (89%)	13 of 13 (100%)

Table 10. (Continued)

Delivery control document number	Duplicate pairs	Standard samples
	<u><math>^{137}\text{Cs}</math></u>	
7	--	1 of 1 (100%)
10	8 of 8 (100%)	1 of 1 (100%)
16	--	1 of 1 (100%)
17	5 of 6 (83%)	1 of 1 (100%)
27	7 of 7 (100%)	1 of 2 (50%)
28	--	1 of 1 (100%)
38	1 of 2 (50%)	3 of 3 (100%)
39	3 of 3 (100%)	3 of 3 (100%)
TOTAL	24 of 26 (92%)	12 of 13 (92%)
	<u><math>^{239+240}\text{Pu}</math></u>	
1	5 of 5 (100%)	--
7	--	1 of 1 (100%)
10	8 of 8 (100%)	1 of 1 (100%)
16	--	1 of 1 (100%)
17	5 of 6 (83%)	1 of 1 (100%)
27	8 of 8 (100%)	2 of 2 (100%)
28	--	1 of 1 (100%)
38	5 of 5 (100%)	3 of 3 (100%)
39	9 of 10 (90%)	3 of 3 (100%)
TOTAL	40 of 42 (95%)	13 of 13 (100%)
	<u><math>^{241}\text{Am}</math></u>	
1	5 of 5 (100%)	--
7	--	1 of 1 (100%)
10	8 of 8 (100%)	1 of 1 (100%)
16	--	1 of 1 (100%)
17	6 of 6 (100%)	0 of 1 (0%)
27	8 of 8 (100%)	2 of 2 (100%)
28	--	1 of 1 (100%)
38	5 of 5 (100%)	3 of 3 (100%)
39	12 of 12 (100%)	3 of 3 (100%)
TOTAL	44 of 44 (100%)	12 of 13 (92%)

Table 11. Acceptable quality control results for duplicate pairs and standard samples analyzed in terrestrial animals by Environmental Analysis Laboratory.

Delivery control document number	Duplicate pairs	Standard samples
	<u><math>^{90}\text{Sr}</math></u>	
40	5 of 6 (83%)	3 of 3 (100%)
41	--	3 of 3 (100%)
TOTAL	5 of 6 (83%)	6 of 6 (100%)
	<u><math>^{137}\text{Cs}</math></u>	
40	4 of 6 (67%)	3 of 3 (100%)
41	--	3 of 3 (100%)
TOTAL	4 of 6 (67%)	6 of 6 (100%)
	<u><math>^{239+240}\text{Pu}</math></u>	
40	6 of 6 (100%)	3 of 3 (100%)
41	--	3 of 3 (100%)
TOTAL	6 of 6 (100%)	6 of 6 (100%)
	<u><math>^{241}\text{Am}</math></u>	
40	6 of 6 (100%)	3 of 3 (100%)
41	--	2 of 3 (90%)
TOTAL	6 of 6 (100%)	5 of 6 (83%)

Table 12. Acceptable quality control results for duplicate pairs and standard samples analyzed in marine organisms by Environmental Analysis Laboratory.

Delivery control document number	Duplicate pairs	Standard samples
	<u><math>^{90}\text{Sr}</math></u>	
4	11 of 12 (92%)	--
6	8 of 9 (89%)	--
13	9 of 9 (100%)	2 of 2 (100%)
22	12 of 12 (100%)	2 of 2 (100%)
23c	13 of 13 (100%)	2 of 2 (100%)
26c	5 of 6 (83%)	1 of 1 (100%)
30c	9 of 10 (90%)	2 of 2 (100%)
35	--	2 of 2 (100%)
TOTAL	66 of 71 (93%)	11 of 11 (100%)

Table 12. (Continued)

Delivery control document number	Duplicate pairs	Standard samples
	<u><math>^{137}\text{Cs}</math></u>	
4	--	--
6	--	--
13	--	--
22	--	2 of 2 (100%)
23c	--	1 of 1 (100%)
26c	--	1 of 1 (100%)
30c	--	1 of 2 (50%)
32	--	--
TOTAL	--	5 of 6 (91%)
	<u><math>^{239+240}\text{Pu}</math></u>	
4	12 of 12 (100%)	--
6	9 of 9 (100%)	--
13	9 of 9 (100%)	1 of 2 (50%)
22	12 of 12 (100%)	2 of 2 (100%)
23c	12 of 12 (100%)	2 of 2 (100%)
26c	2 of 6 (33%)	1 of 1 (100%)
30c	9 of 10 (90%)	2 of 2 (100%)
35	--	2 of 2 (100%)
TOTAL	65 of 70 (93%)	10 of 11 (91%)
	<u><math>^{238}\text{Pu}</math></u>	
4	12 of 12 (100%)	--
6	9 of 9 (100%)	--
13	9 of 9 (100%)	--
22	12 of 12 (100%)	--
23c	12 of 12 (100%)	--
26c	6 of 6 (100%)	--
30c	10 of 10 (100%)	--
35	--	--
TOTAL	70 of 70 (100%)	--
	<u><math>^{241}\text{Am}</math></u>	
4	10 of 12 (83%)	--
6	8 of 9 (89%)	--
13	7 of 9 (78%)	2 of 2 (100%)
22	11 of 12 (92%)	2 of 2 (100%)
23c	13 of 13 (100%)	2 of 2 (100%)
26c	6 of 6 (100%)	1 of 1 (100%)
30c	10 of 10 (100%)	2 of 2 (100%)
35	--	2 of 2 (100%)
TOTAL	65 of 71 (92%)	11 of 11 (100%)

Table 13. Acceptable quality control results for duplicate pairs and standard samples analyzed in marine sediment by Environmental Analysis Laboratory.

Delivery control document number	Duplicate pairs	Standard samples
	<u>137Cs</u>	
44	--	2 of 2 (100%)
TOTAL	--	2 of 2 (100%)
	<u>239+240Pu</u>	
32	7 of 7 (100%)	2 of 2 (100%)
44	--	2 of 2 (100%)
TOTAL	7 of 7 (100%)	4 of 4 (100%)
	<u>238Pu</u>	
32	7 of 7 (100%)	--
44	--	--
TOTAL	7 of 7 (100%)	--
	<u>241Am</u>	
32	7 of 7 (100%)	2 of 2 (100%)
44	--	2 of 2 (100%)
TOTAL	7 of 7 (100%)	4 of 4 (100%)

Table 14. Acceptable quality control results for duplicate pairs and standard samples analyzed in soil by Eberline Instrument Corporation.

Delivery control document number	Duplicate pairs	Standard samples
	<u>90Sr</u>	
2	10 of 12 (83%)	--
7	4 of 4 (100%)	2 of 3 (67%)
22	4 of 4 (100%)	2 of 2 (100%)
24	4 of 4 (100%)	2 of 2 (100%)
25	2 of 2 (100%)	3 of 3 (100%)
27	2 of 2 (100%)	2 of 2 (100%)
28	--	2 of 2 (100%)
TOTAL	26 of 28 (93%)	13 of 14 (93%)

Table 14. (Continued)

Delivery control document number	Duplicate pairs	Standard samples
<u><math>^{137}\text{Cs}</math></u>		
2	--	--
7	3 of 4 (75%)	2 of 3 (67%)
23	--	3 of 3 (100%)
24	3 of 3 (100%)	2 of 2 (100%)
25	--	3 of 3 (100%)
27	--	2 of 2 (100%)
28	--	3 of 3 (100%)
TOTAL	6 of 7 (86%)	15 of 16 (94%)
<u><math>^{239+240}\text{Pu}</math></u>		
1	--	1 of 1 (100%)
2	11 of 12 (92%)	--
7	3 of 4 (75%)	2 of 3 (67%)
11	5 of 6 (83%)	3 of 3 (100%)
18	4 of 6 (67%)	2 of 2 (100%)
22	4 of 4 (100%)	2 of 2 (100%)
24	4 of 4 (100%)	2 of 2 (100%)
25	2 of 2 (100%)	3 of 3 (100%)
27	2 of 2 (100%)	2 of 2 (100%)
28	3 of 4 (75%)	3 of 3 (100%)
TOTAL	38 of 44 (86%)	22 of 23 (96%)
<u><math>^{238}\text{Pu}</math></u>		
2	7 of 7 (100%)	--
TOTAL	7 of 7 (100%)	--
<u><math>^{241}\text{Am}</math></u>		
2	--	--
7	2 of 2 (100%)	2 of 3 (67%)
11	6 of 6 (100%)	2 of 2 (100%)
22	1 of 1 (100%)	2 of 2 (100%)
24	4 of 4 (100%)	2 of 2 (100%)
25	1 of 1 (100%)	3 of 3 (100%)
27	1 of 1 (100%)	2 of 2 (100%)
28	--	2 of 2 (100%)
TOTAL	15 of 15 (100%)	15 of 16 (94%)

Table 15. Acceptable quality control results for duplicate pairs and standard samples analyzed in vegetation by Eberline Instrument Corporation.

Delivery control document number	Duplicate pairs	Standard samples
	<u><math>^{90}\text{Sr}</math></u>	
1	1 of 5 (20%)	--
10	2 of 7 (29%)	1 of 1 (100%)
17	4 of 4 (100%)	2 of 2 (100%)
21	--	--
TOTAL	7 of 16 (44%)	3 of 3 (100%)
	<u><math>^{137}\text{Cs}</math></u>	
1	--	--
10	7 of 7 (100%)	1 of 1 (100%)
17	8 of 8 (100%)	2 of 2 (100%)
21	--	--
TOTAL	15 of 15 (100%)	3 of 3 (100%)
	<u><math>^{239+240}\text{Pu}</math></u>	
1	3 of 5 (60%)	--
17	6 of 8 (75%)	2 of 2 (100%)
21	--	2 of 2 (100%)
TOTAL	9 of 13 (69%)	4 of 4 (100%)
	<u><math>^{241}\text{Am}</math></u>	
1	1 of 5 (20%)	--
10	--	2 of 2 (100%)
17	1 of 1 (100%)	2 of 2 (100%)
21	--	--
TOTAL	2 of 6 (33%)	4 of 4 (100%)

Table 16. Acceptable quality control results for duplicate pairs and standard samples analyzed in soil by Laboratory of Radiation Ecology.

Delivery control document number	Duplicate pairs	Standard samples
	<u><math>^{90}\text{Sr}</math></u>	
1	12 of 12 (100%)	--
3	11 of 12 (92%)	--
4	13 of 18 (72%)	2 of 2 (100%)
7	9 of 9 (100%)	4 of 4 (100%)
8	12 of 12 (100%)	3 of 3 (100%)
TOTAL	57 of 63 (90%)	9 of 9 (100%)
	<u><math>^{137}\text{Cs}</math></u>	
4	5 of 8 (63%)	2 of 2 (100%)
7	7 of 9 (78%)	4 of 4 (100%)
8	11 of 11 (100%)	3 of 3 (100%)
TOTAL	23 of 28 (82%)	9 of 9 (100%)
	<u><math>^{239+240}\text{Pu}</math></u>	
1	12 of 12 (100%)	--
3	12 of 12 (100%)	--
4	15 of 18 (83%)	2 of 2 (100%)
7	9 of 9 (100%)	4 of 4 (100%)
8	11 of 12 (92%)	3 of 3 (100%)
TOTAL	59 of 63 (94%)	9 of 9 (100%)
	<u><math>^{241}\text{Pu}</math></u>	
7	8 of 9 (89%)	--
8	7 of 9 (78%)	--
TOTAL	15 of 18 (83%)	--
	<u><math>^{241}\text{Am}</math></u>	
1	2 of 2 (100%)	--
4	--	2 of 2 (100%)
7	9 of 9 (100%)	4 of 4 (100%)
8	5 of 6 (83%)	3 of 3 (100%)
TOTAL	16 of 17 (94%)	9 of 9 (100%)

Table 17. Unacceptable quality control results for duplicate pairs and standard samples analyzed in soil by Environmental Analysis Laboratory.

Delivery control document number	Duplicate pairs	Standard samples
	<u>239+240Pu</u>	
20	6 of 12 (50%)	1 of 1 (100%)
TOTAL	6 of 12 (50%)	1 of 1 (100%)

Table 18. Unacceptable quality control results for duplicate pairs and standard samples analyzed in marine sediment by Environmental Analysis Laboratory.

Delivery control document number	Duplicate pairs	Standard samples
	<u>90Sr</u>	
32	7 of 7 (100%)	0 of 2 (0%)
44	--	0 of 2 (0%)
TOTAL	7 of 7 (100%)	0 of 4 (0%)

Table 19. Unacceptable quality control results for duplicate pairs and standard samples analyzed in soil by Eberline Instrument Corporation.

Delivery control document number	Duplicate pairs	Standard samples
<u>90Sr</u>		
11	4 of 6 (67%)	0 of 2 (0%)
18	4 of 6 (67%)	0 of 2 (0%)
19	1 of 5 (20%)	0 of 3 (0%)
20	0 of 5 (0%)	0 of 3 (0%)
23	1 of 6 (17%)	0 of 3 (0%)
28	0 of 4 (0%)	0 of 3 (0%)
TOTAL	10 of 32 (31%)	0 of 16 (0%)
<u>137Cs</u>		
1	--	0 of 1 (0%)
2	--	0 of 1 (0%)
11	3 of 6 (50%)	0 of 2 (0%)
18	2 of 6 (33%)	0 of 2 (0%)
19	2 of 5 (40%)	0 of 3 (0%)
20	0 of 2 (0%)	0 of 3 (0%)
22	5 of 5 (100%)	0 of 2 (0%)
TOTAL	12 of 24 (50%)	0 of 14 (0%)
<u>239+240Pu</u>		
2	--	0 of 1 (0%)
19	0 of 6 (0%)	2 of 3 (67%)
20	8 of 9 (89%)	0 of 3 (0%)
23	11 of 12 (92%)	0 of 3 (0%)
TOTAL	19 of 27 (70%)	2 of 10 (20%)
<u>241Pu</u>		
11	1 of 4 (25%)	--
18	1 of 6 (17%)	--
19	1 of 12 (8%)	--
TOTAL	3 of 22 (14%)	--
<u>241Am</u>		
1	--	0 of 1 (0%)
2	--	0 of 1 (0%)
18	3 of 6 (50%)	0 of 2 (0%)
19	1 of 6 (17%)	2 of 3 (67%)
20	7 of 9 (78%)	1 of 3 (33%)
23	6 of 12 (50%)	1 of 3 (33%)
28	0 of 4 (0%)	1 of 3 (33%)
TOTAL	17 of 37 (46%)	5 of 16 (31%)

Table 20. Unacceptable quality control results for duplicate pairs and standard samples analyzed in vegetation by Eberline Instrument Corporation.

Delivery control document number	Duplicate pairs	Standard samples
	<u>239+240Pu</u>	
10	0 of 7 (0%)	0 of 1 (0%)
TOTAL	0 of 7 (0%)	0 of 1 (0%)
	<u>241Am</u>	
10	1 of 7 (14%)	0 of 1 (0%)
17	2 of 8 (25%)	2 of 2 (100%)
TOTAL	3 of 15 (20%)	2 of 3 (67%)

### CONCLUSIONS

The analytical reliability of the data generated from the NMIRS has been established through an extensive QC program. Blind duplicates and/or standards were included with all of the DCDs, and based on the analyses of these QC samples, the data accepted in the program accurately reflect the radioactivity in the Northern Marshall Islands. Although a high level of compliance with the QC criteria was achieved, some of the DCDs analyzed did not meet the established criteria, and these data were not approved for inclusion in the data base. As a consequence, calculations based on data approved by the QC program give accurate estimates of radiation dose to residents of the Northern Marshall Islands, while data that could give unreliable dose calculations have been rejected.

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APPENDIX. QUALITY CONTROL RESULTS FOR DUPLICATE  
PAIRS PLOTTED AGAINST EACH OTHER

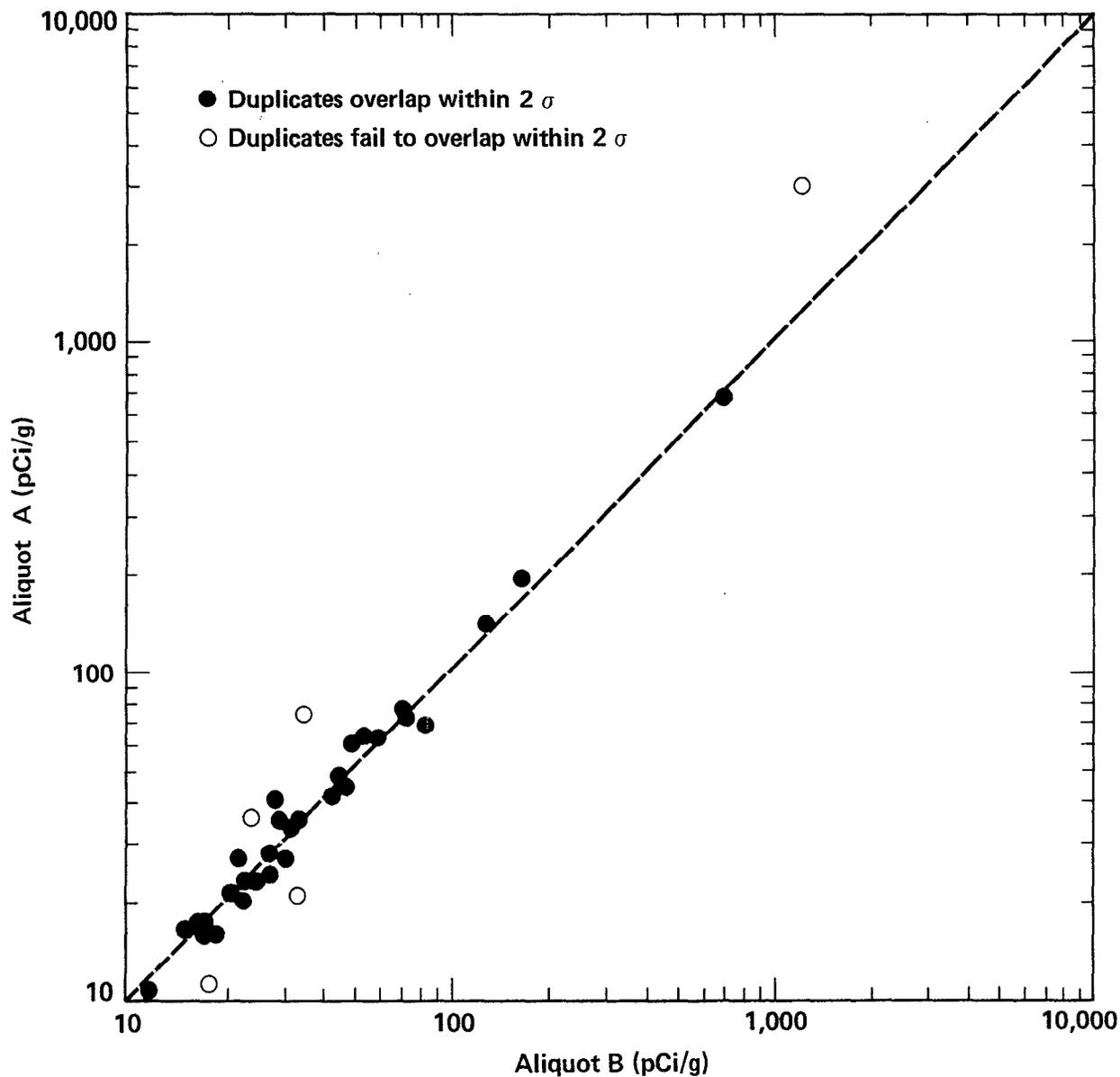


Figure A1. Acceptable quality control results for duplicate pairs analyzed for  $^{90}\text{Sr}$  in soil by Environmental Analysis Laboratory. Results depicted are for concentrations between 10 and 10,000 pCi/g. Broken line represents perfect agreement and is not a fit to the data.

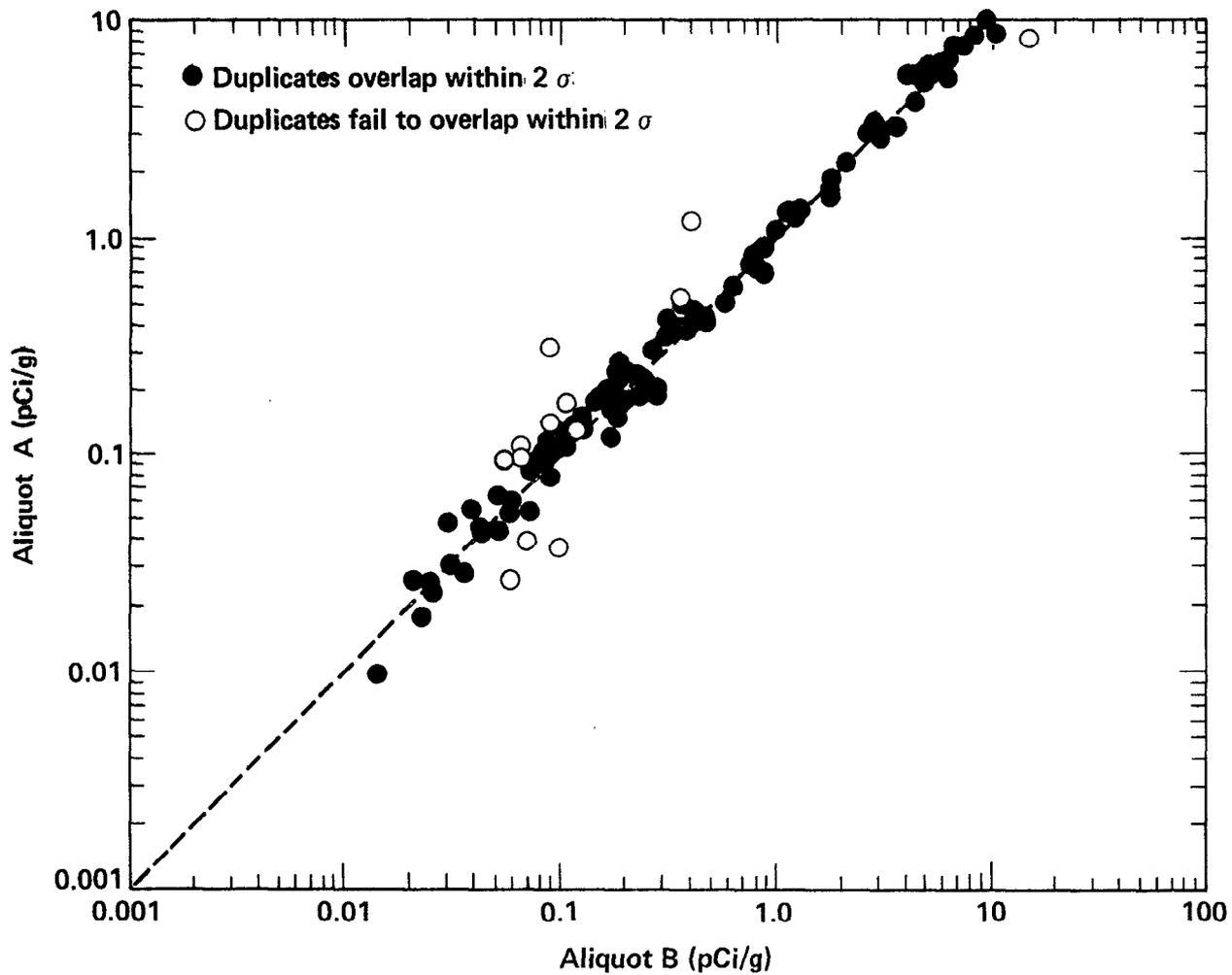


Figure A2. Acceptable quality control results for duplicate pairs analyzed for  $^{90}\text{Sr}$  in soil by Environmental Analysis Laboratory. Results depicted are for concentrations between  $1 \times 10^{-3}$  and 10 pCi/g. Broken line represents perfect agreement and is not a fit to the data.

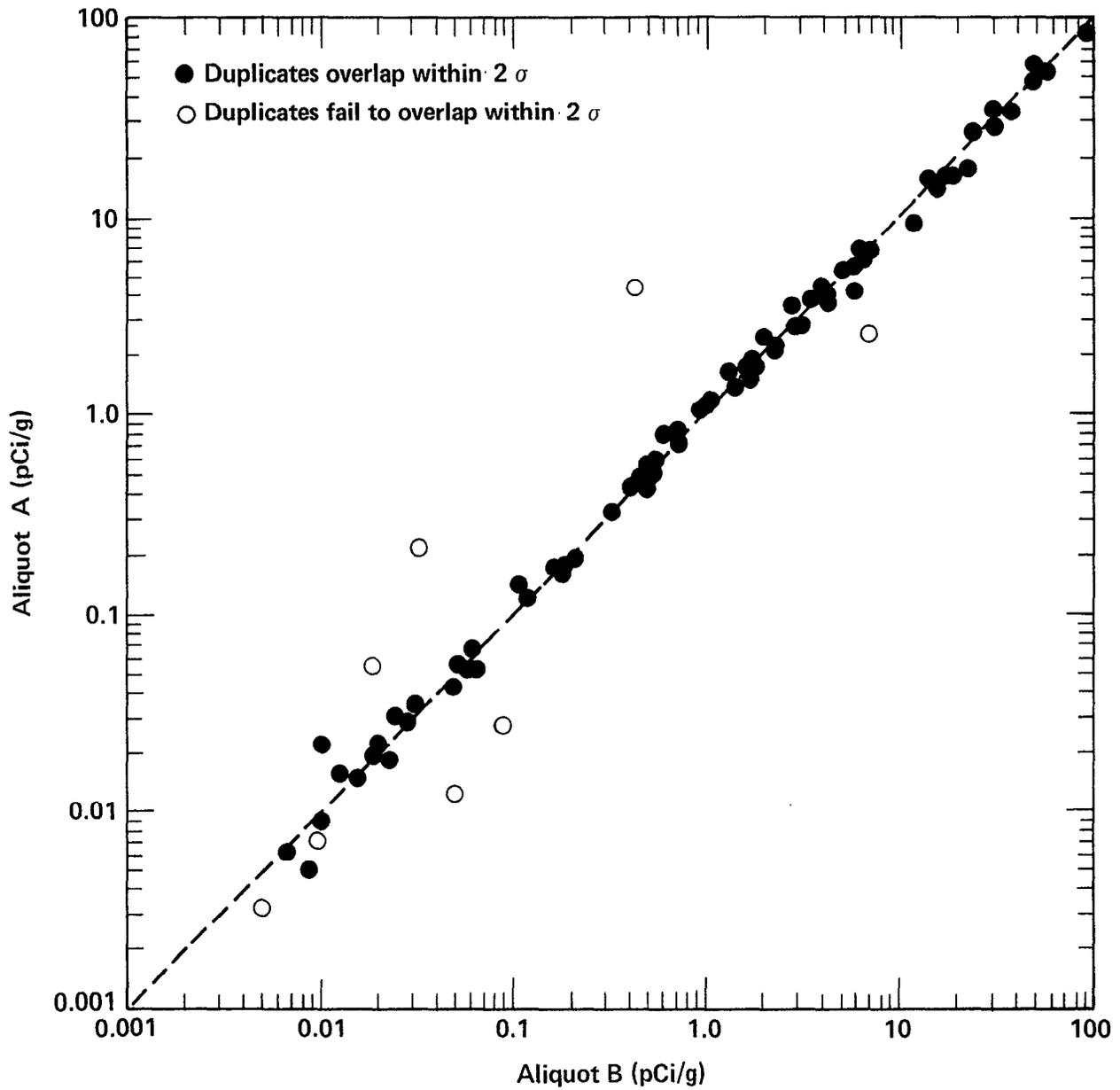


Figure A3. Acceptable quality control results for duplicate pairs analyzed for  $^{137}\text{Cs}$  in soil by Environmental Analysis Laboratory. Broken line represents perfect agreement and is not a fit to the data. Two pairs (one acceptable) involving zero concentrations are not shown.

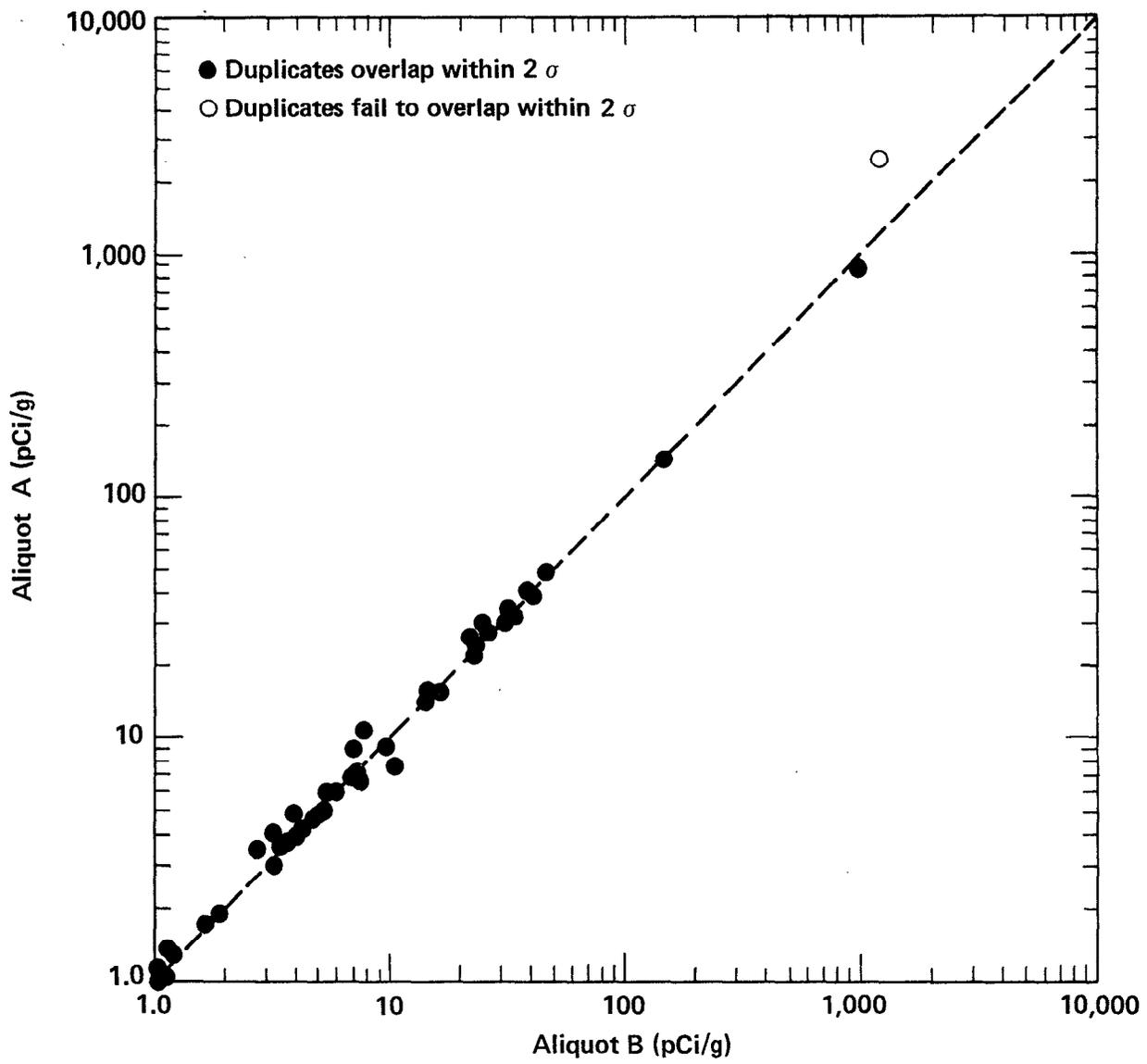


Figure A4. Acceptable quality control results for duplicate pairs analyzed for  $^{239+240}\text{Pu}$  in soil by Environmental Analysis Laboratory. Results depicted are for concentrations between 1 and 10,000 pCi/g. Broken line represents perfect agreement and is not a fit to the data. One unacceptable pair is not shown.

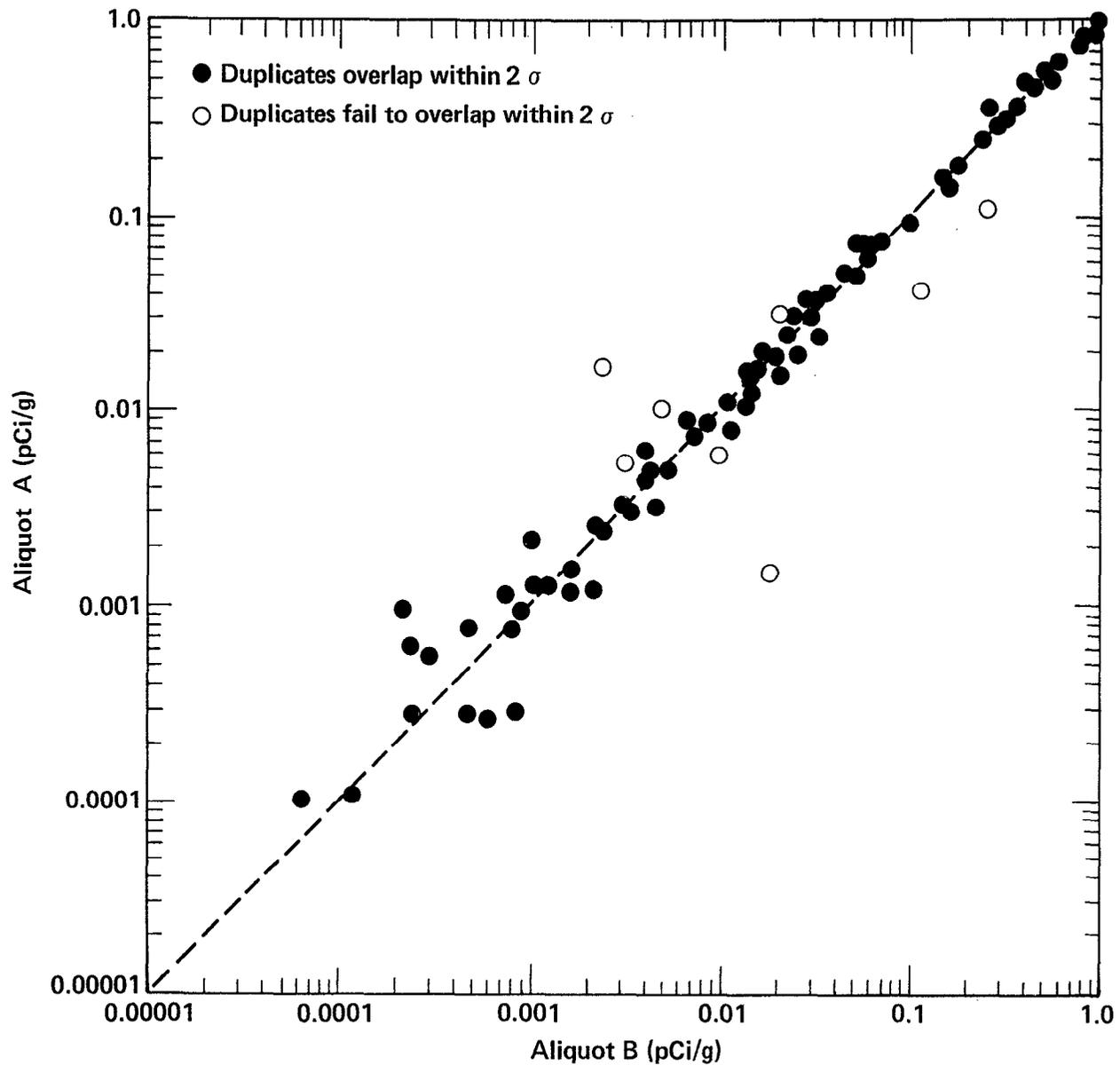


Figure A5. Acceptable quality control results for duplicate pairs analyzed for  $^{239+240}\text{Pu}$  in soil by Environmental Analysis Laboratory. Results depicted are for concentrations between  $1 \times 10^{-5}$  and 1 pCi/g. Broken line represents perfect agreement and is not a fit to the data.

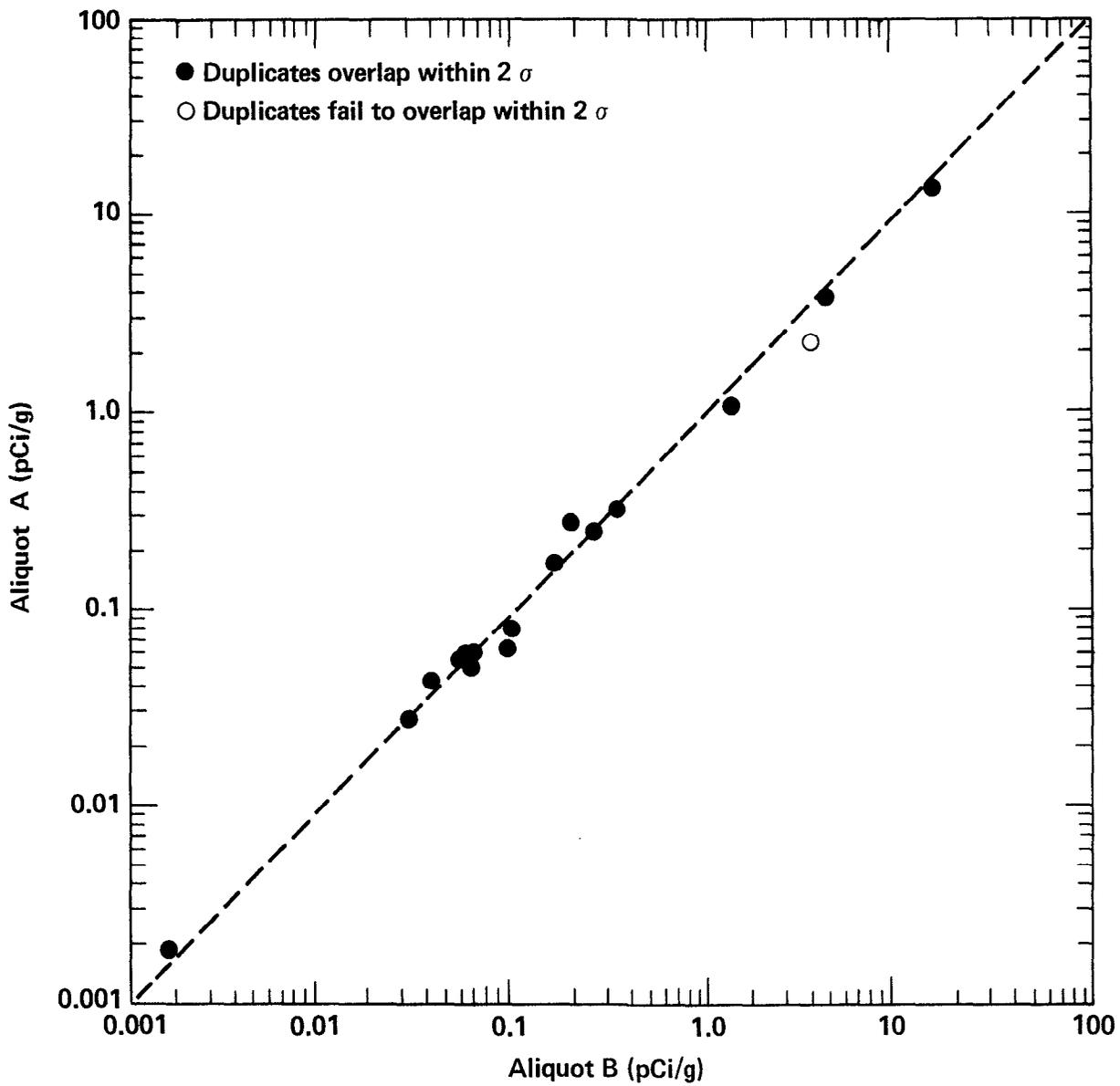


Figure A6. Acceptable quality control results for duplicate pairs analyzed for  $^{238}\text{Pu}$  in soil by Environmental Analysis Laboratory. Broken line represents perfect agreement and is not a fit to the data.

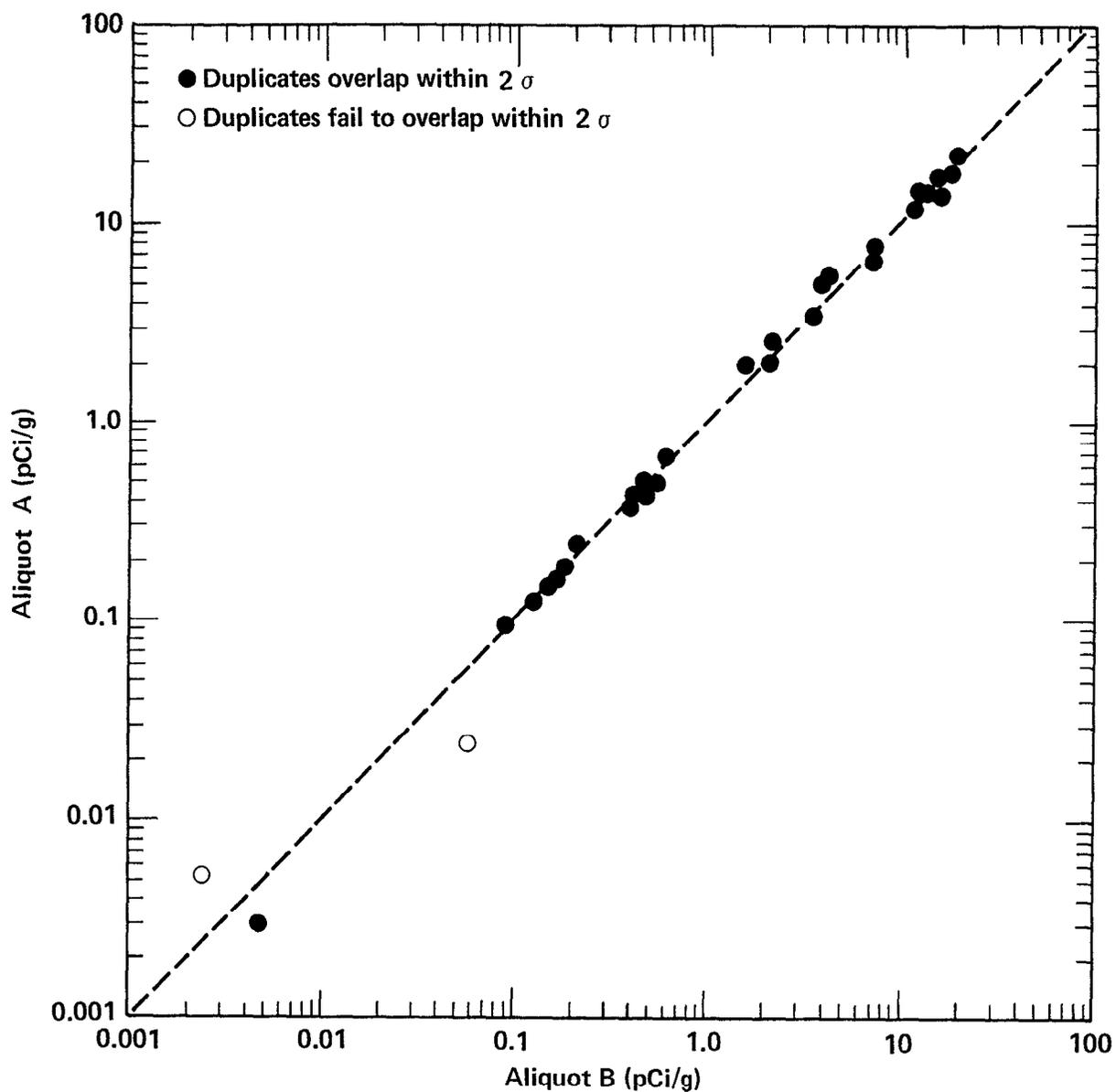


Figure A7. Acceptable quality control results for duplicate pairs analyzed for  $^{239}\text{Pu}$  in soil by Environmental Analysis Laboratory. Broken line represents perfect agreement and is not a fit to the data.

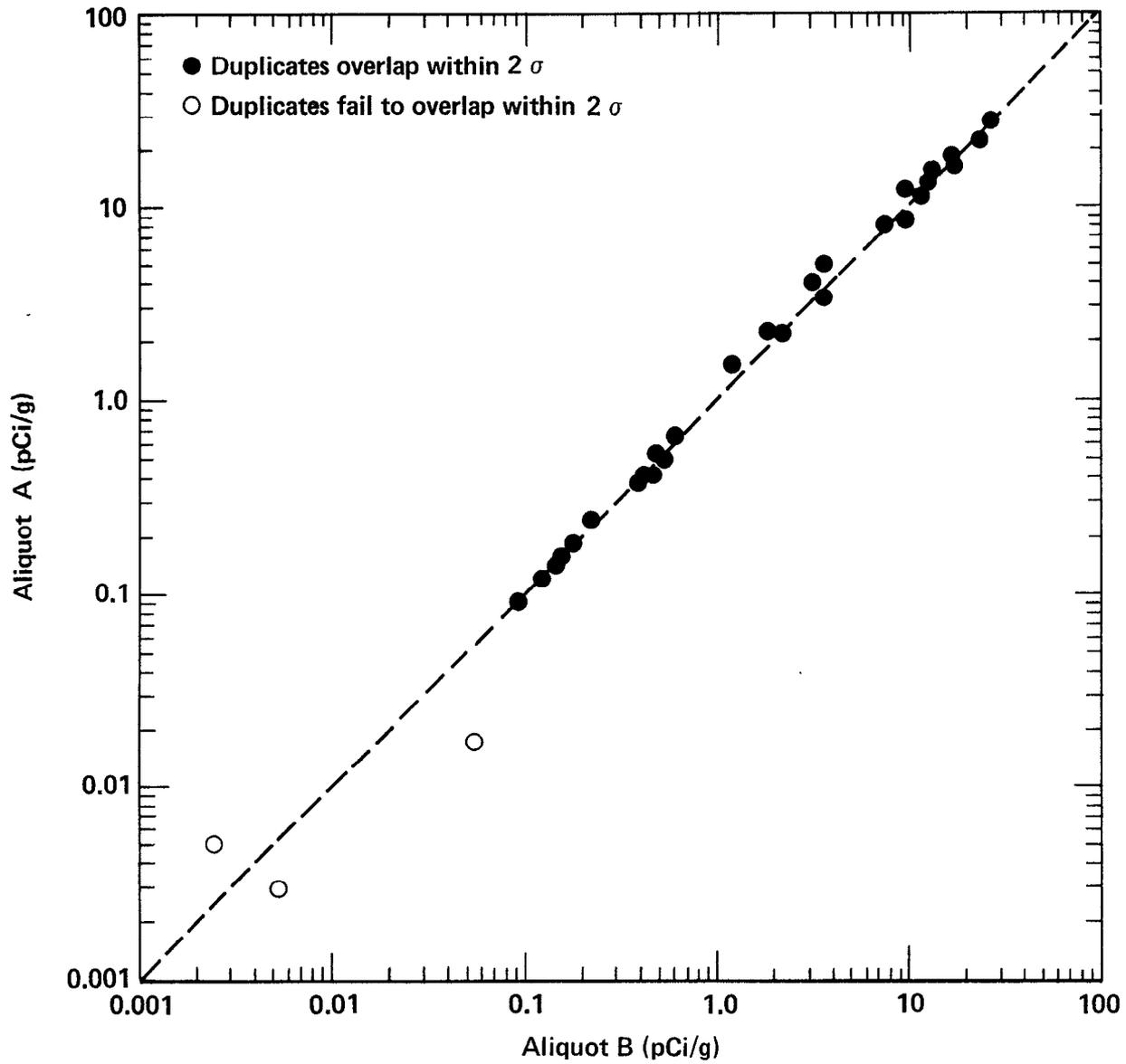


Figure A8. Acceptable quality control results for duplicate pairs analyzed for  $^{240}\text{Pu}$  in soil by Environmental Analysis Laboratory. Broken line represents perfect agreement and is not a fit to the data.

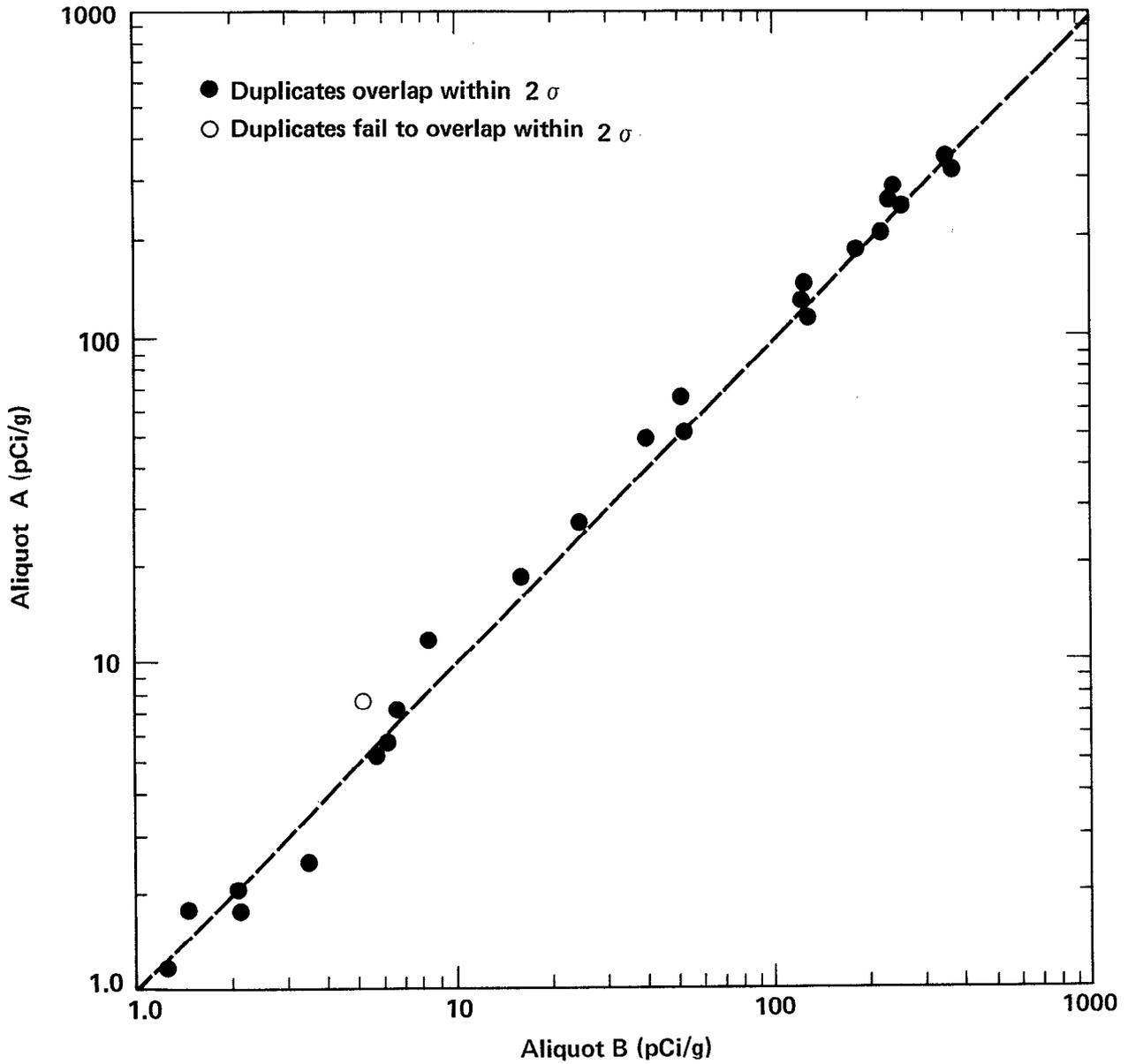


Figure A9. Acceptable quality control results for duplicate pairs analyzed for  $^{241}\text{Pu}$  in soil by Environmental Analysis Laboratory. Broken line represents perfect agreement and is not a fit to the data.

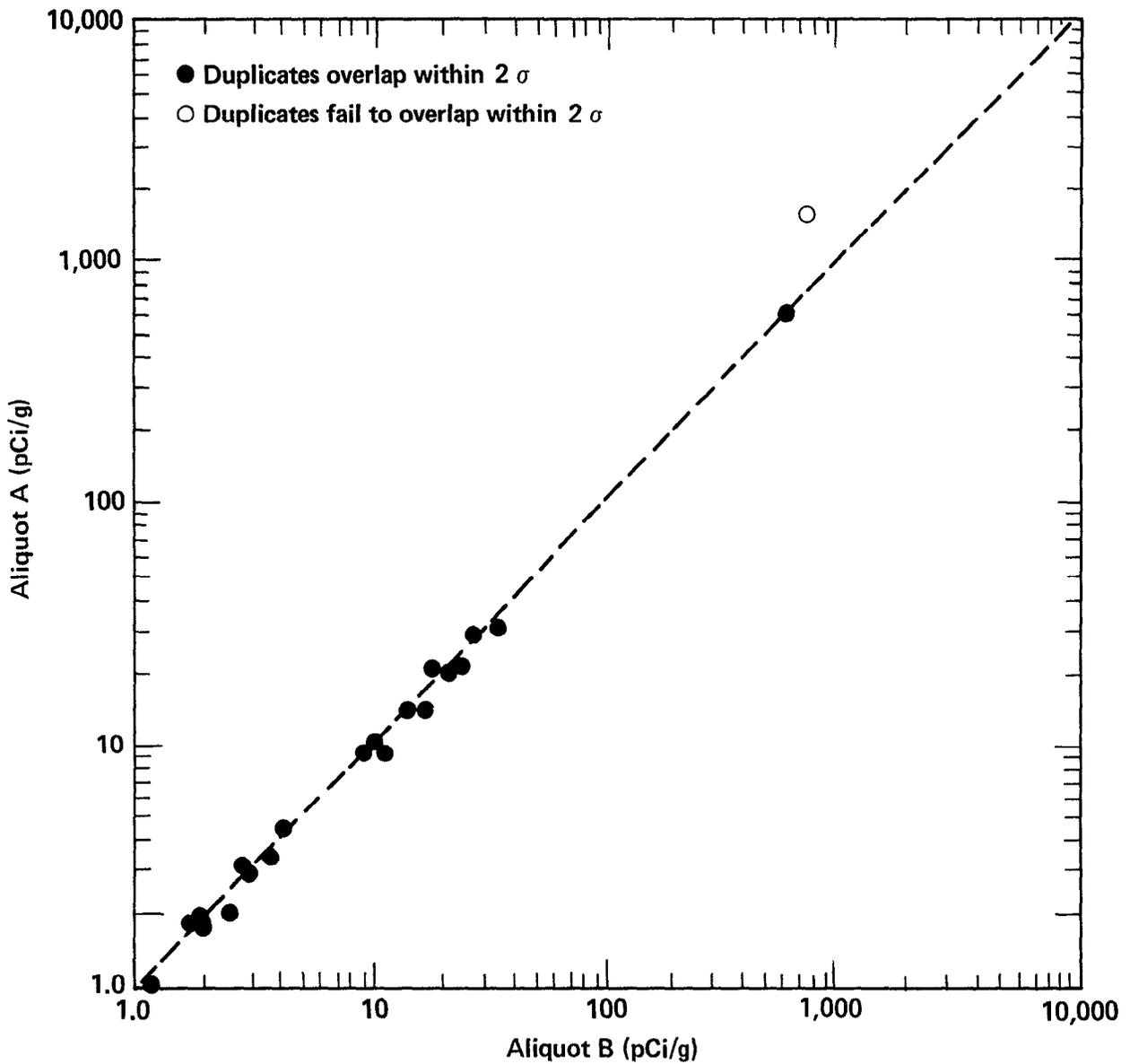


Figure A10. Acceptable quality control results for duplicate pairs analyzed for  $^{241}\text{Am}$  in soil by Environmental Analysis Laboratory. Results depicted are for concentrations between 1 and 10,000 pCi/g. Broken line represents perfect agreement and is not a fit to the data.

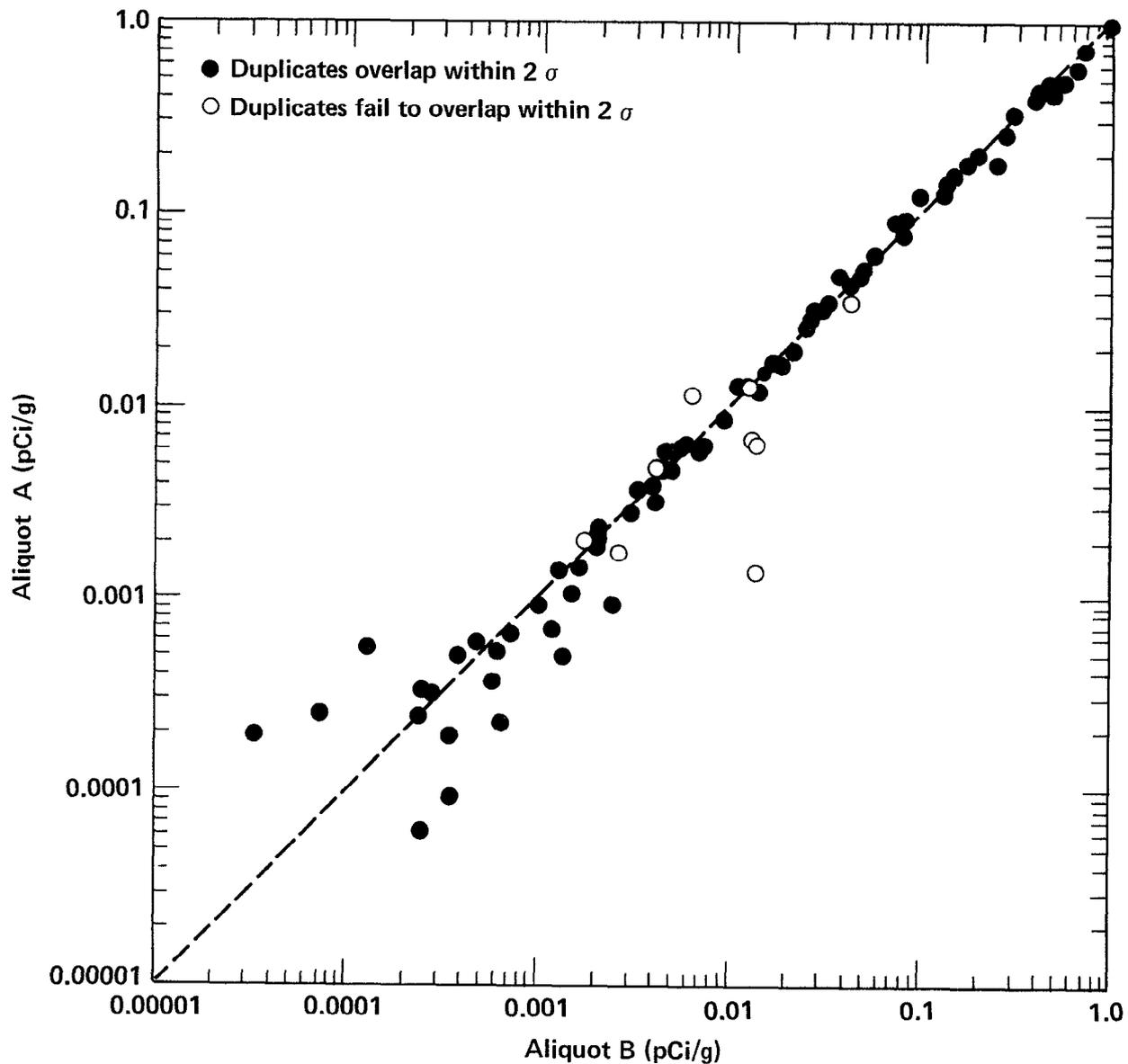


Figure A11. Acceptable quality control results for duplicate pairs analyzed for  $^{241}\text{Am}$  in soil by Environmental Analysis Laboratory. Results depicted are for concentrations between  $1 \times 10^{-5}$  and 1 pCi/g. Broken line represents perfect agreement and is not a fit to the data. Five pairs (four acceptable) involving zero or negative concentrations are not shown.







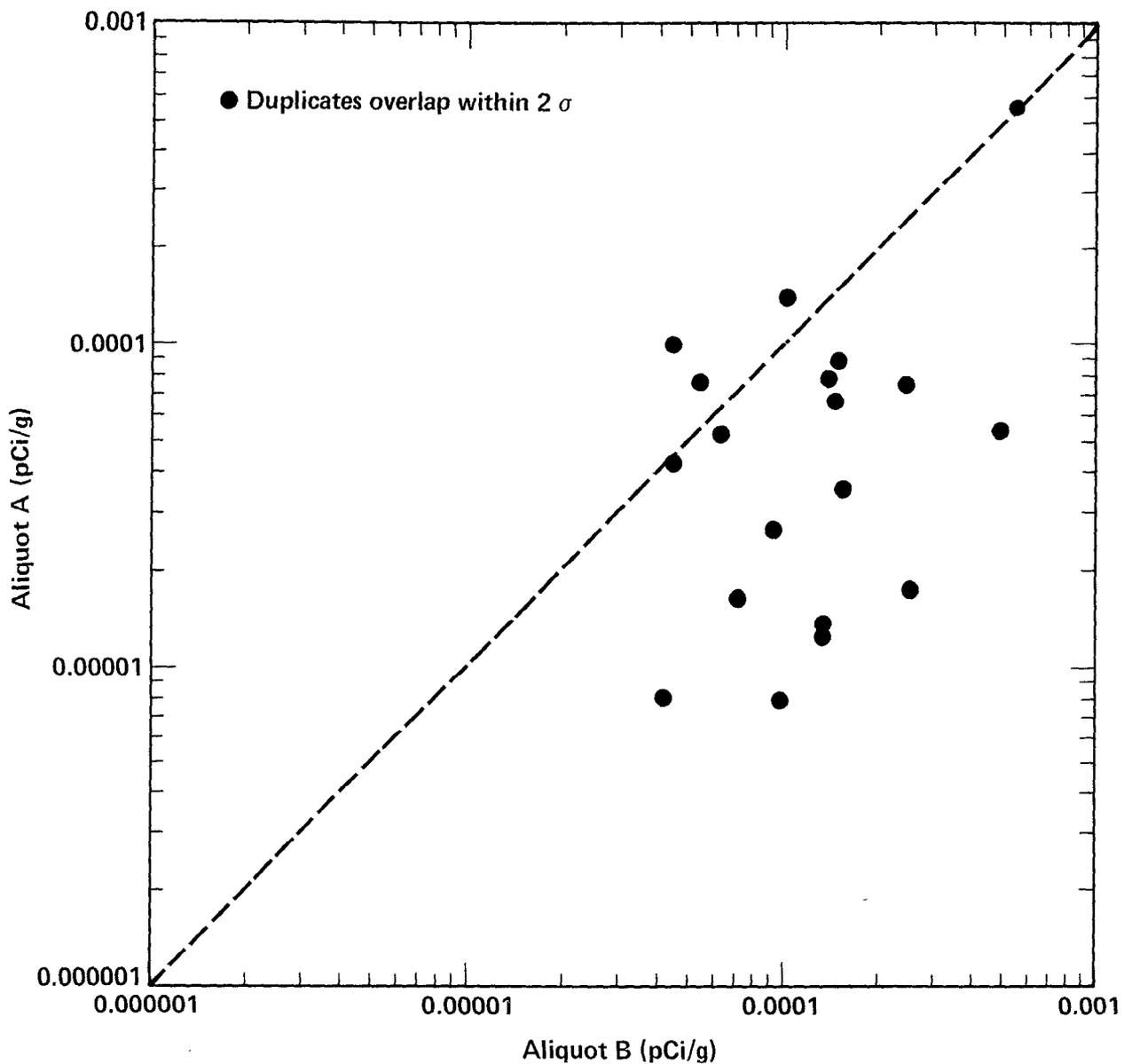


Figure A15. Acceptable quality control results for duplicate pairs analyzed for  $^{241}\text{Am}$  in vegetation by Environmental Analysis Laboratory. Broken line represents perfect agreement and is not a fit to the data. Twenty-five acceptable pairs involving zero or negative concentrations are not shown.

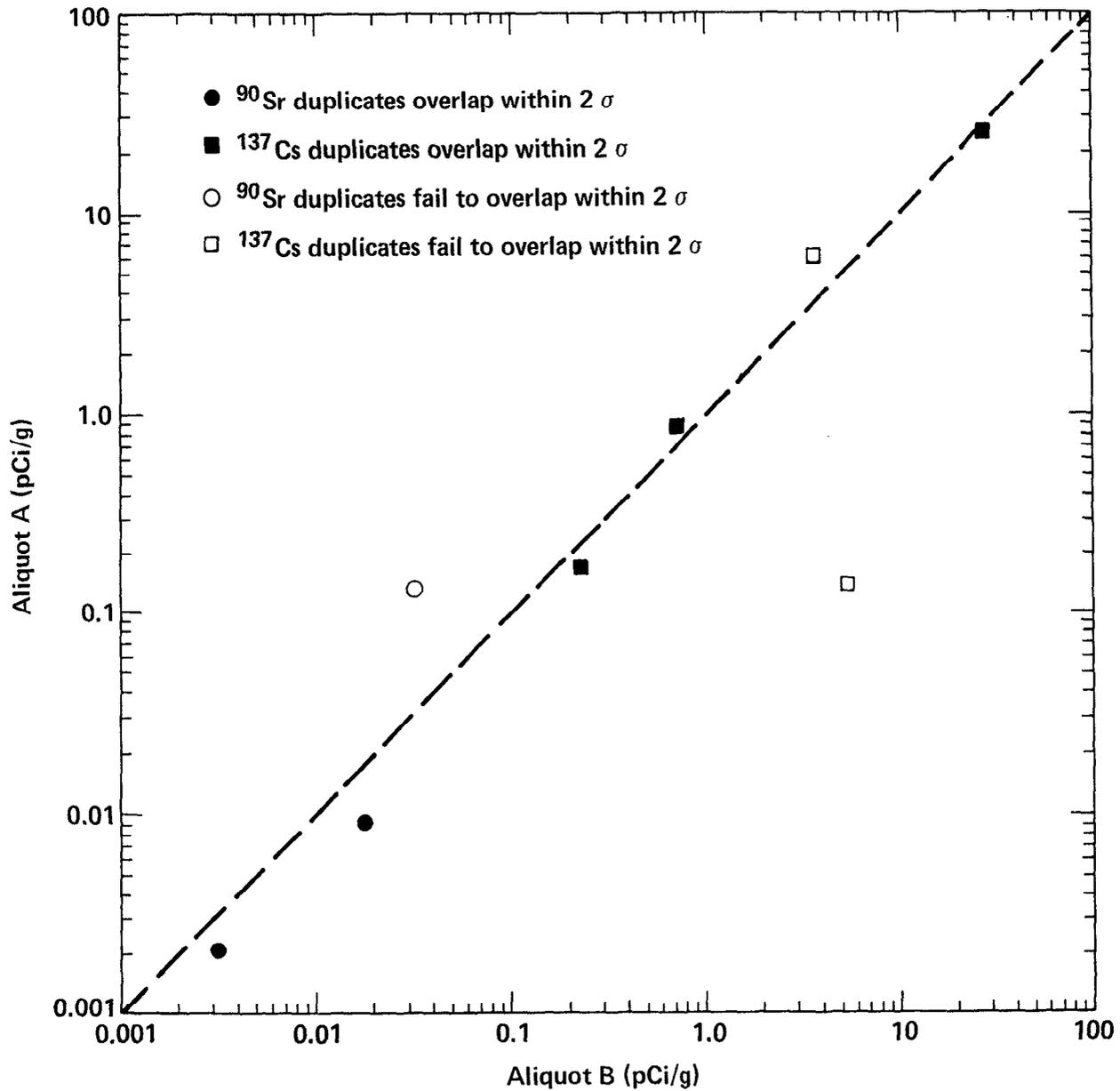


Figure A16. Acceptable quality control results for duplicate pairs analyzed for  $^{90}\text{Sr}$  and  $^{137}\text{Cs}$  in terrestrial animals by Environmental Analysis Laboratory. Broken line represents perfect agreement and is not a fit to the data. Three acceptable  $^{90}\text{Sr}$  pairs involving zero or negative concentrations are not shown.

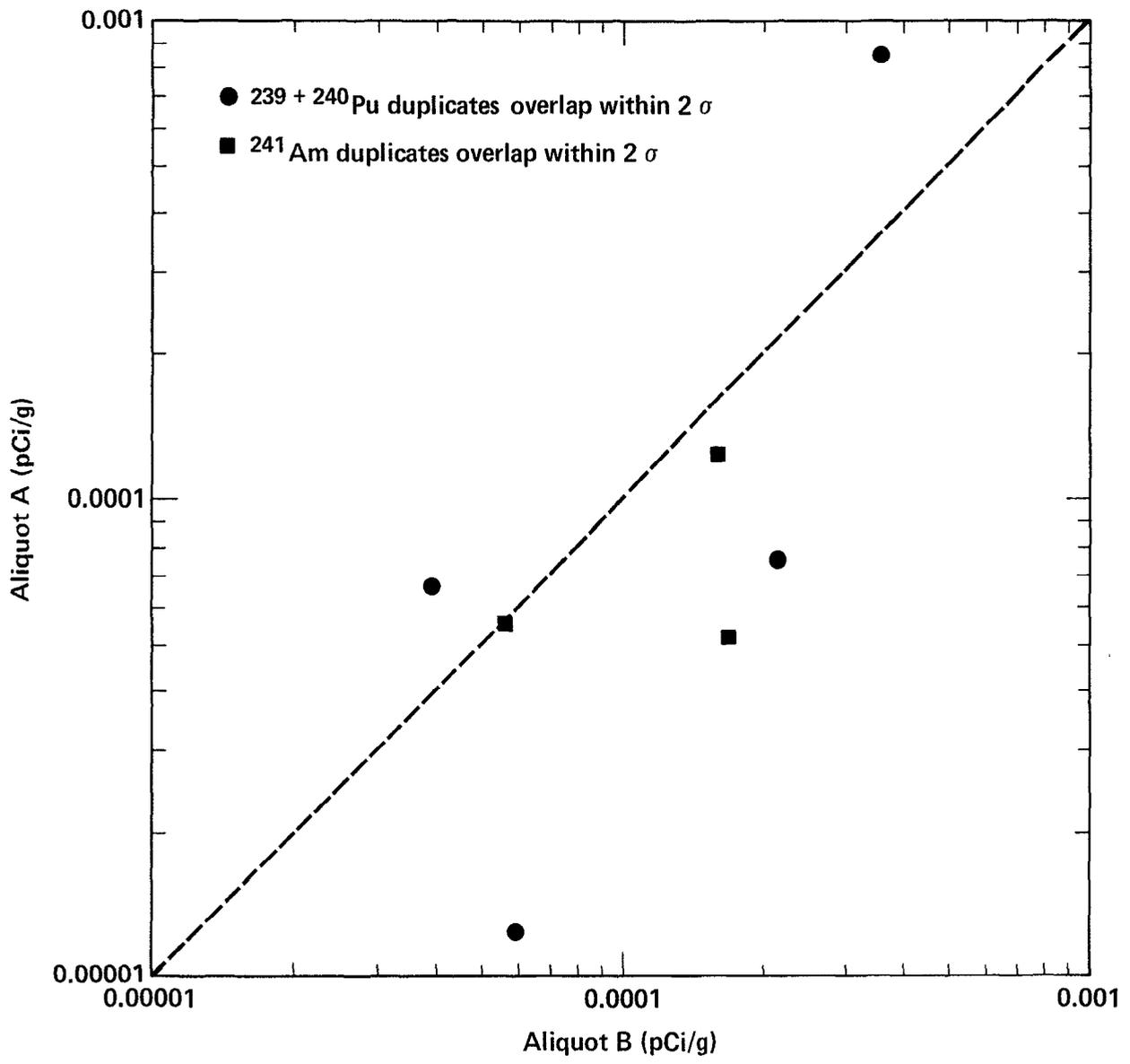


Figure A17. Acceptable quality control results for duplicate pairs analyzed for  $^{239} + ^{240}\text{Pu}$  and  $^{241}\text{Am}$  in terrestrial animals by Environmental Analysis Laboratory. Broken line represents perfect agreement and is not a fit to the data. Five acceptable pairs (three  $^{241}\text{Am}$ ) involving zero or negative concentrations are not shown.

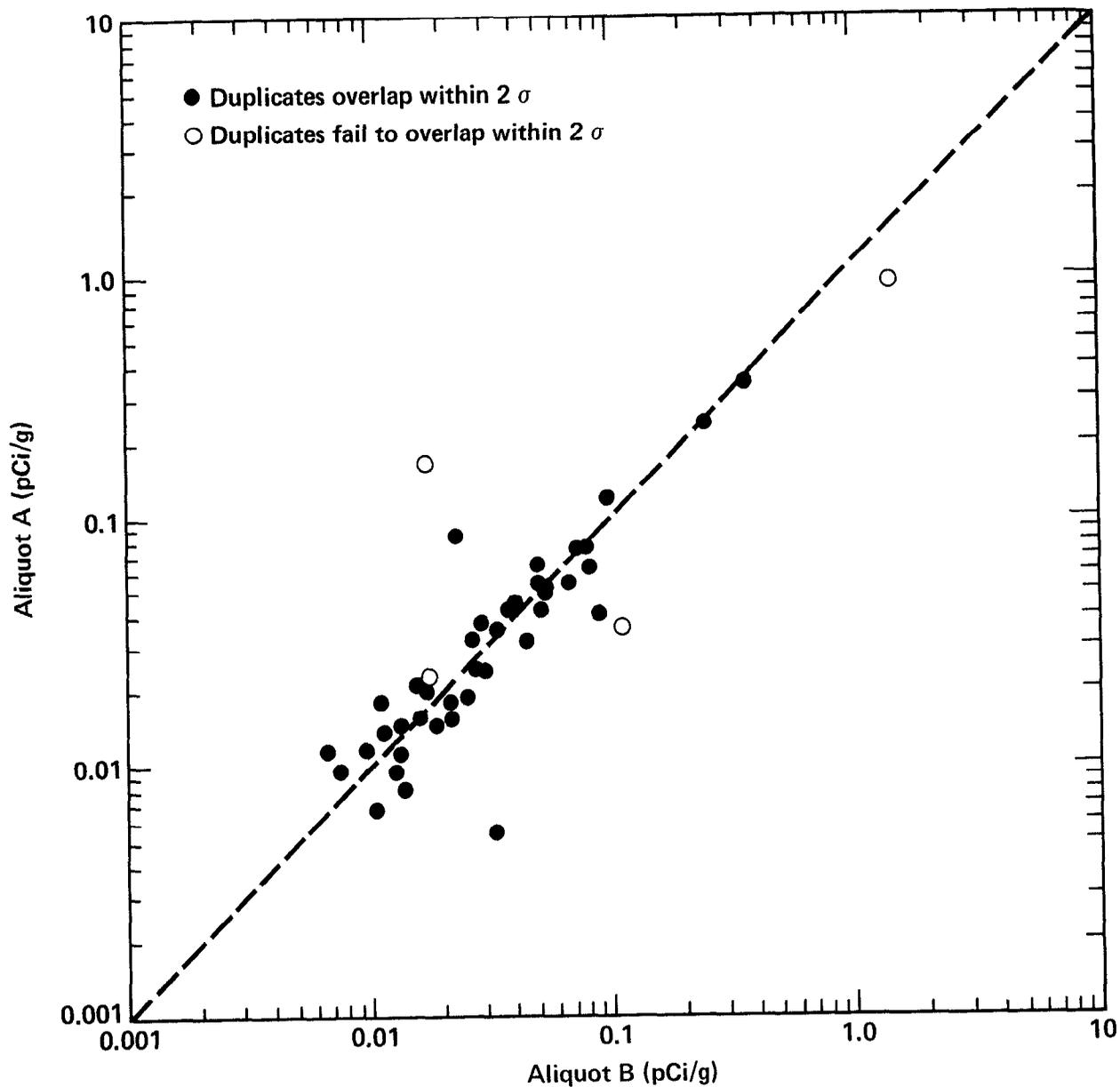


Figure A18. Acceptable quality control results for duplicate pairs analyzed for  $^{90}\text{Sr}$  in marine organisms by Environmental Analysis Laboratory. Broken line represents perfect agreement and is not a fit to the data. Twenty-five acceptable pairs involving zero concentrations are not shown.

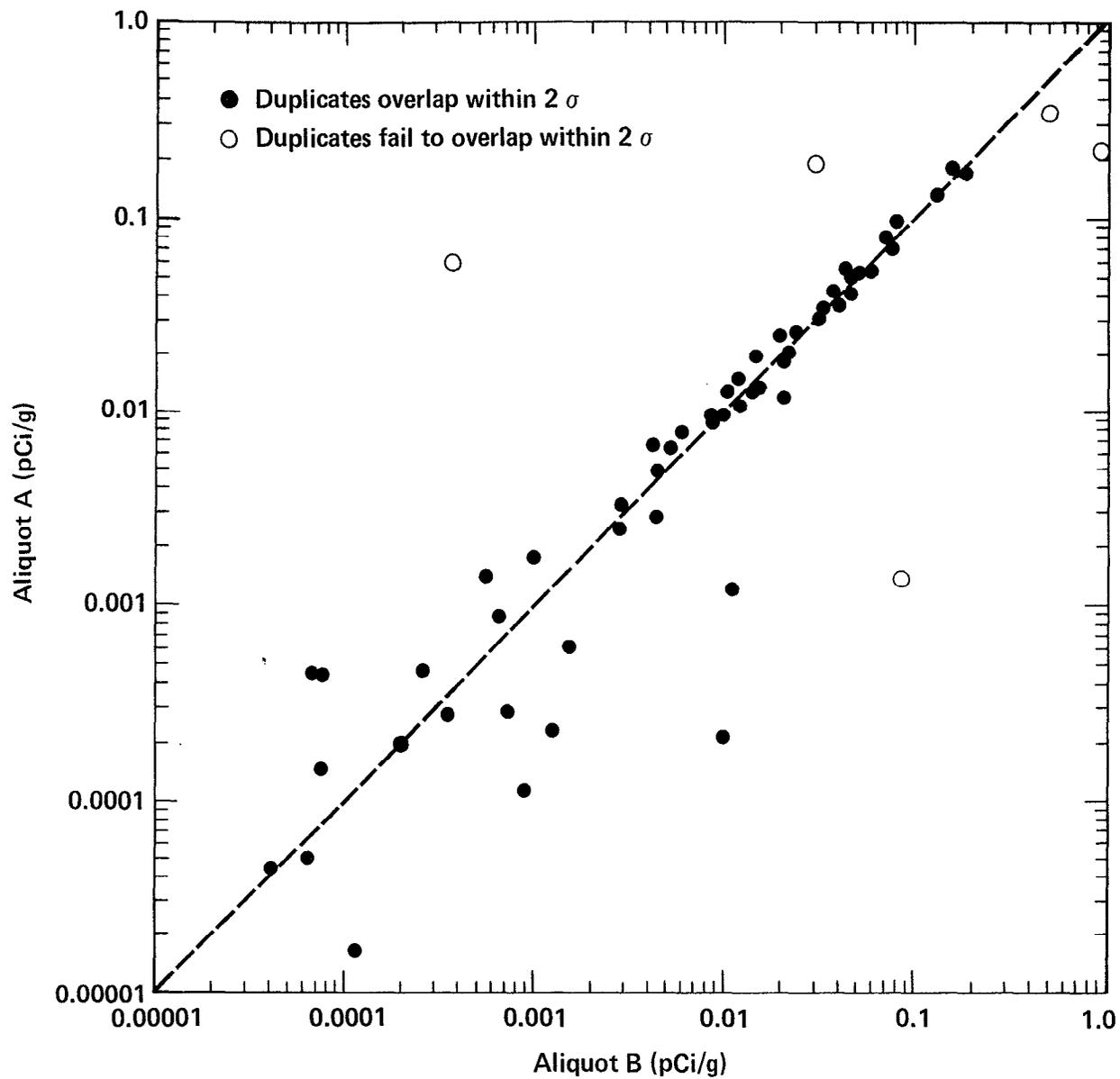


Figure A19. Acceptable quality control results for duplicate pairs analyzed for  $^{239+240}\text{Pu}$  in marine organisms by Environmental Analysis Laboratory. Broken line represents perfect agreement and is not a fit to the data. Ten acceptable pairs involving zero or negative concentrations are not shown.

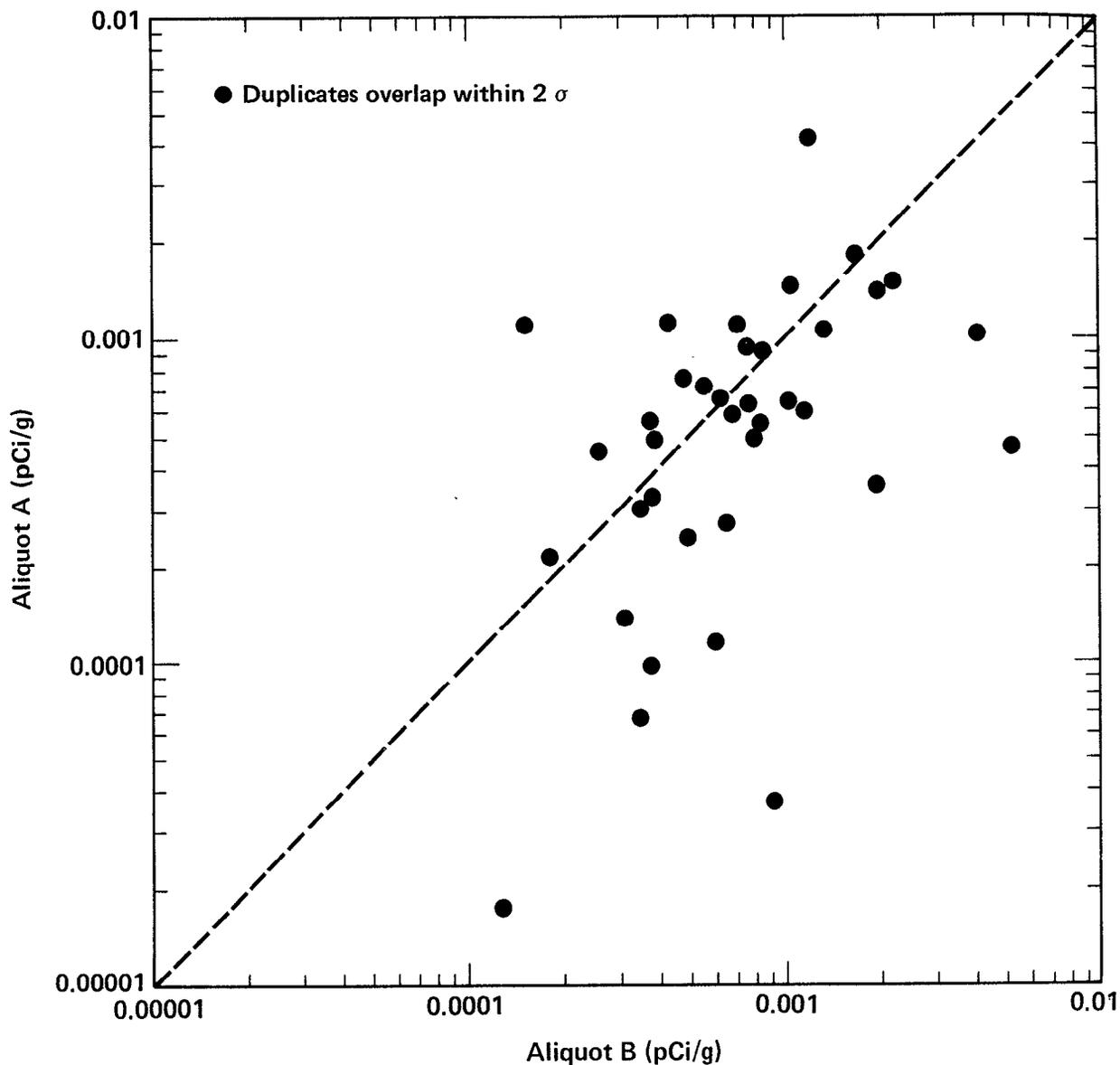


Figure A20. Acceptable quality control results for duplicate pairs analyzed for  $^{238}\text{Pu}$  in marine organisms by Environmental Analysis Laboratory. Broken line represents perfect agreement and is not a fit to the data. Thirty-two acceptable pairs involving zero or negative concentrations are not shown.

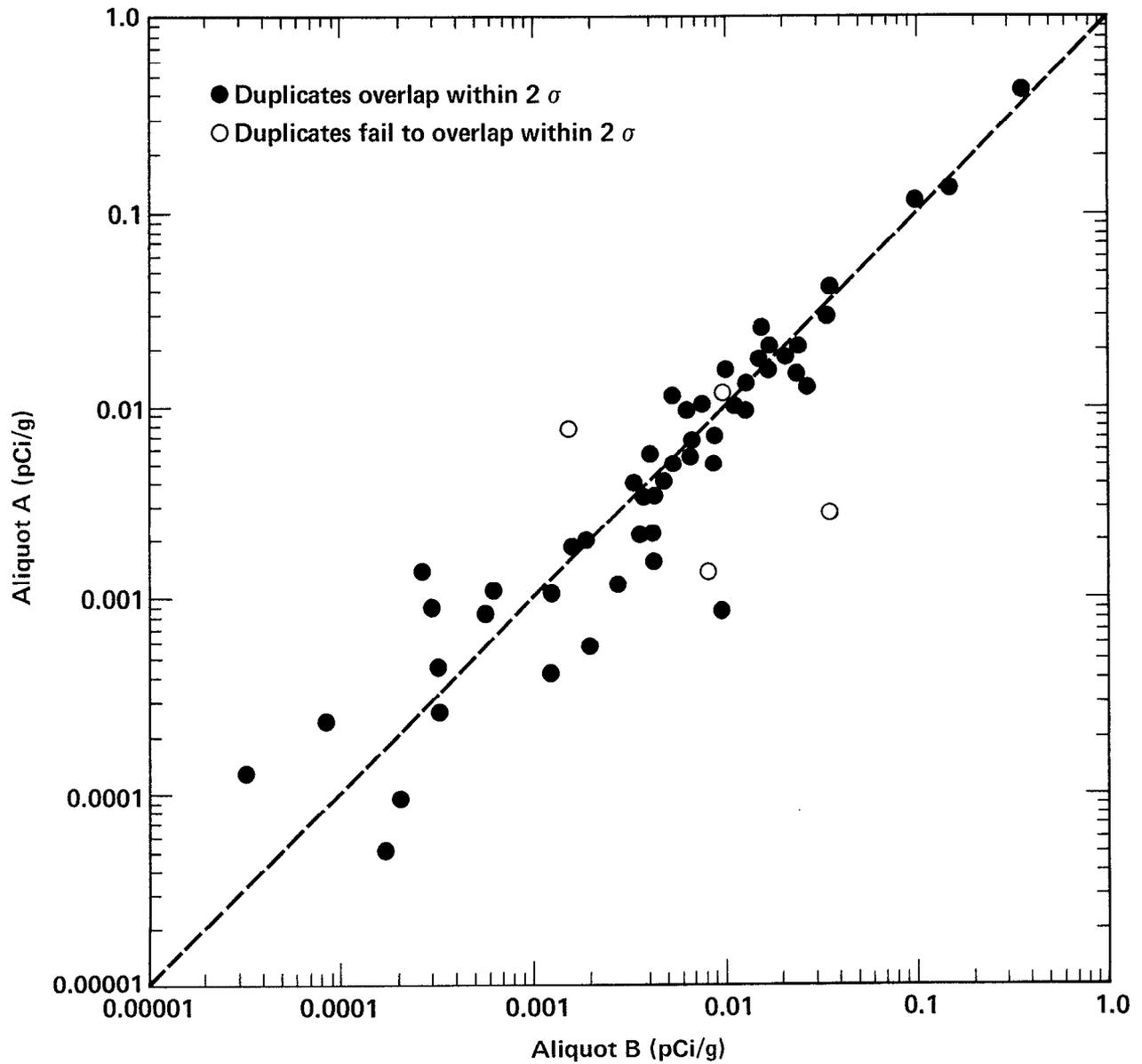


Figure A21. Acceptable quality control results for duplicate pairs analyzed for  $^{241}\text{Am}$  in marine organisms by Environmental Analysis Laboratory. Broken line represents perfect agreement and is not a fit to the data. Seventeen pairs (fifteen acceptable) involving zero or negative concentrations are not shown.

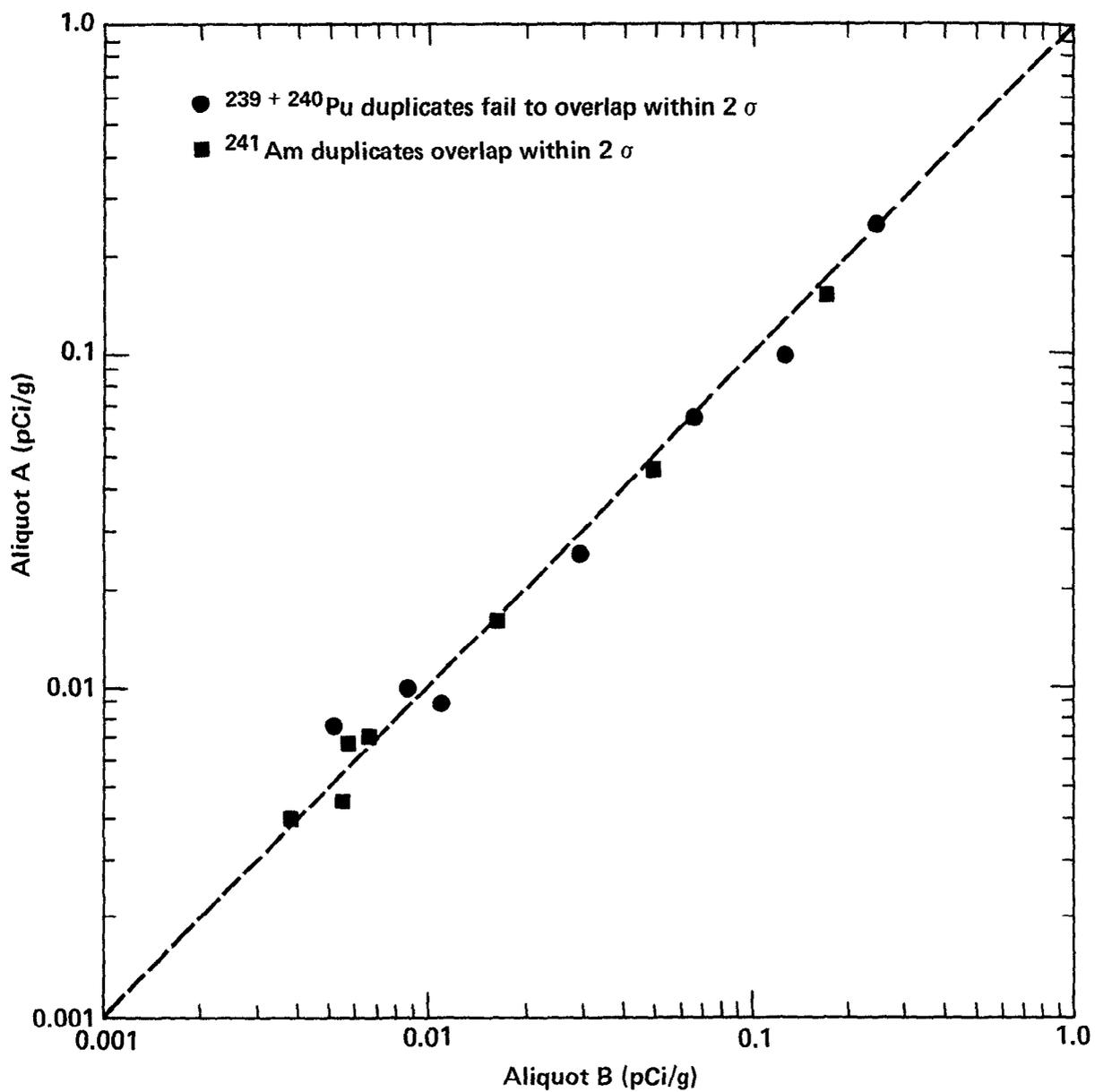


Figure A22. Acceptable quality control results for duplicate pairs analyzed for  $^{239} + ^{240}\text{Pu}$  and  $^{241}\text{Am}$  in marine sediment by Environmental Analysis Laboratory. Broken line represents perfect agreement and is not a fit to the data.

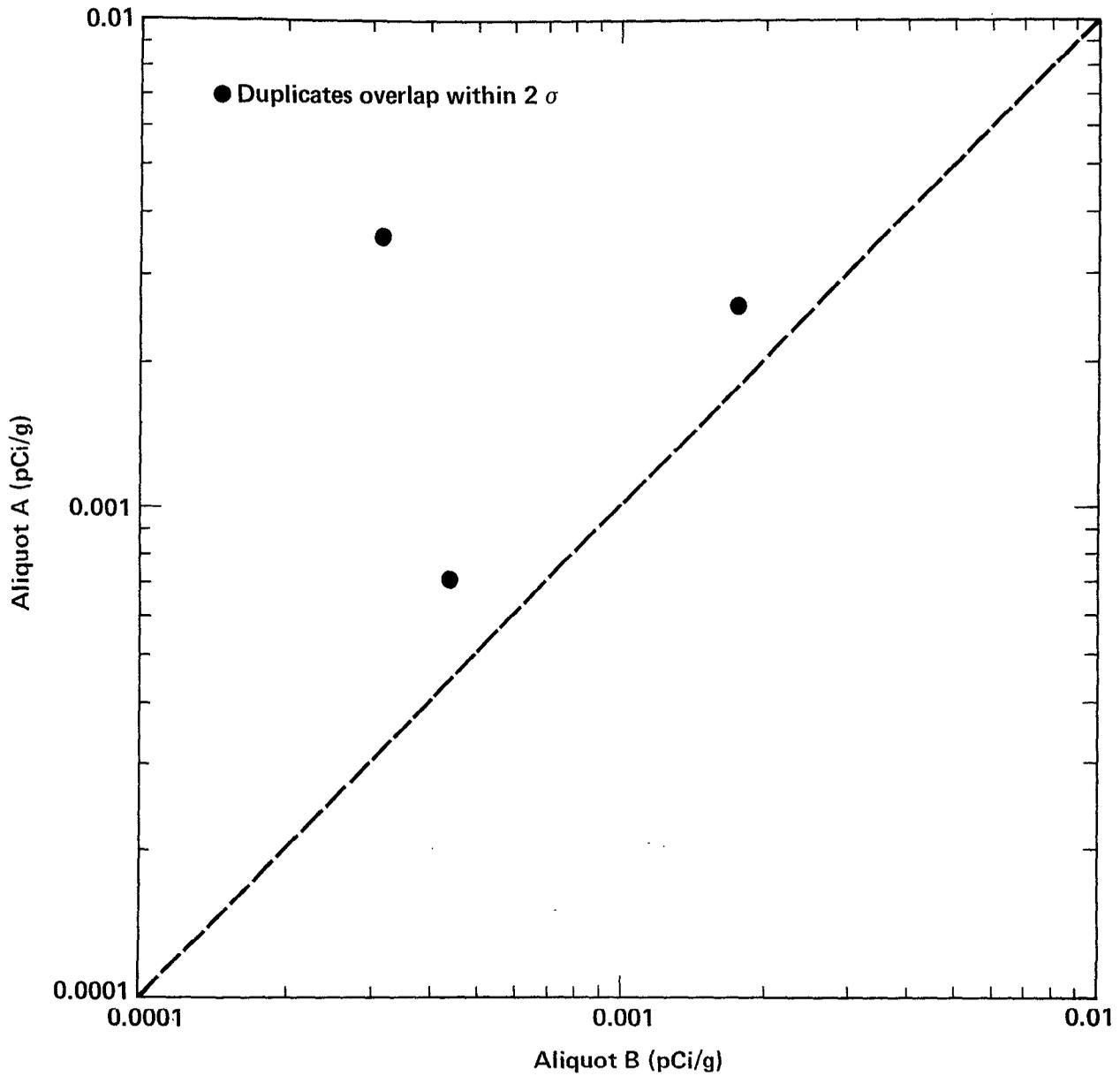


Figure A23. Acceptable quality control results for duplicate pairs analyzed for  $^{238}\text{Pu}$  in marine sediment by Environmental Analysis Laboratory. Broken line represents perfect agreement and is not a fit to the data. Four acceptable pairs involving zero or negative concentrations are not shown.

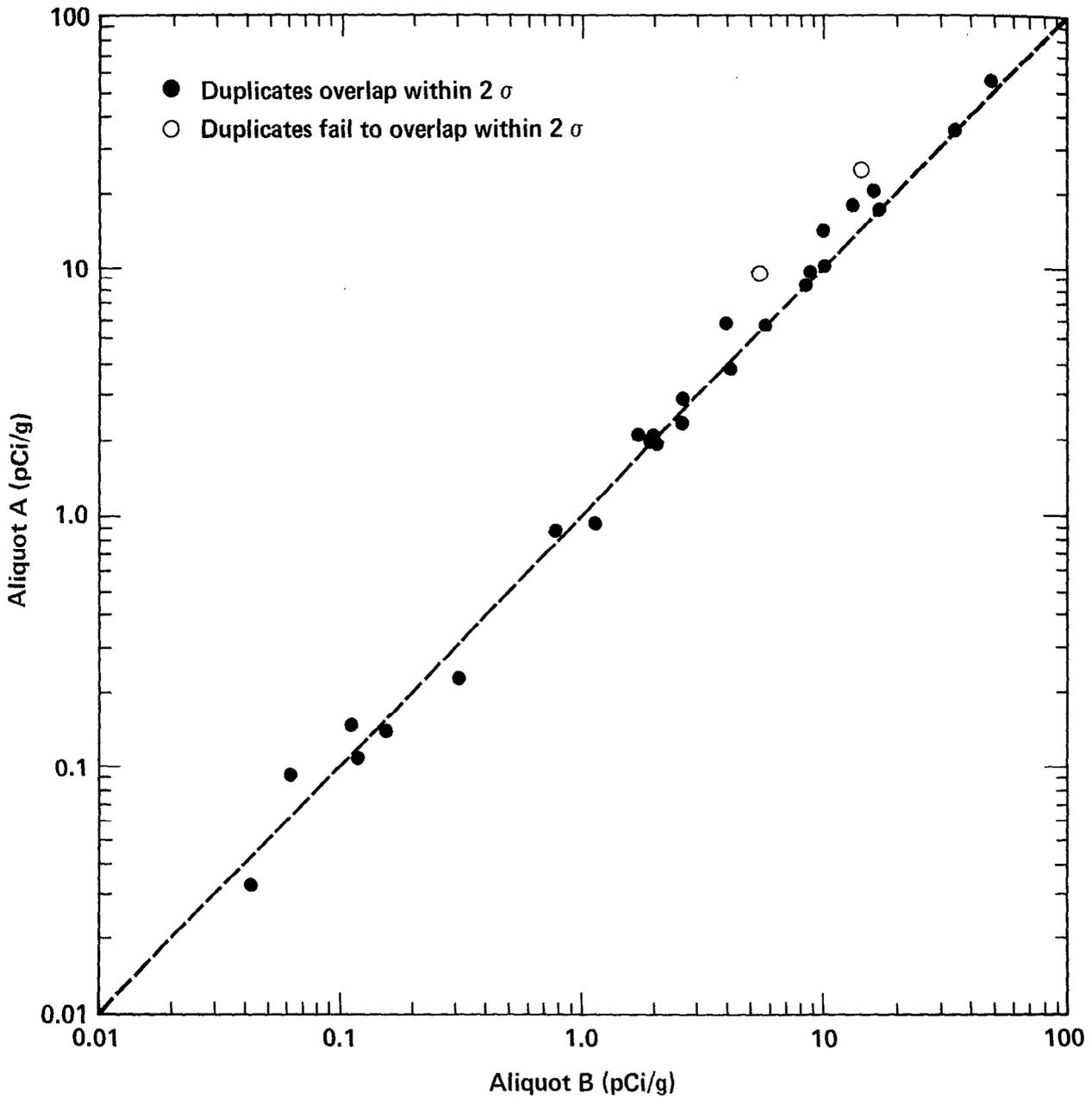


Figure A24. Acceptable quality control results for duplicate pairs analyzed for  $^{90}\text{Sr}$  in soil by Eberline Instrument Corporation. Broken line represents perfect agreement and is not a fit to the data.

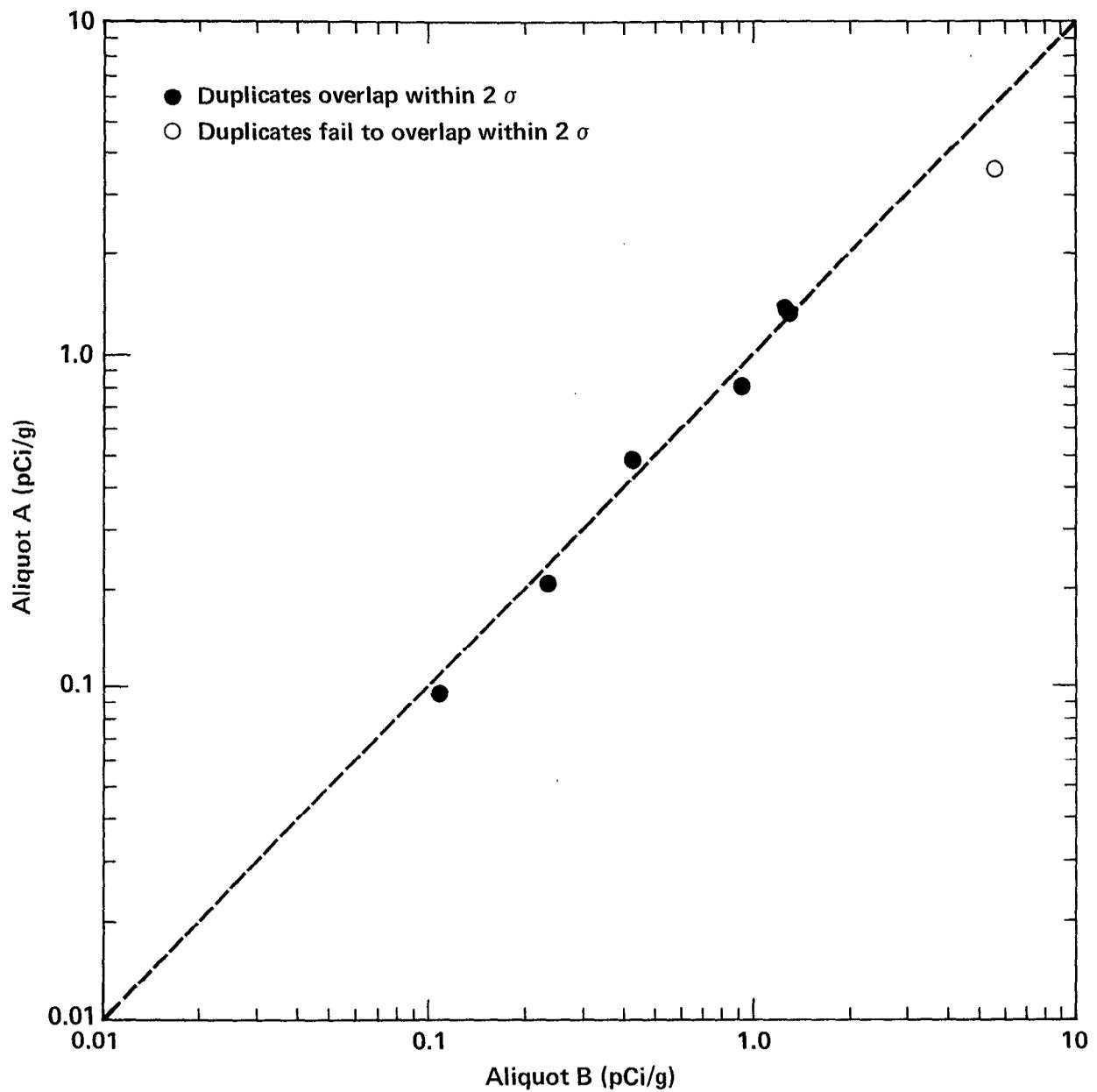


Figure A25. Acceptable quality control results for duplicate pairs analyzed for  $^{137}\text{Cs}$  in soil by Eberline Instrument Corporation. Broken line represents perfect agreement and is not a fit to the data.

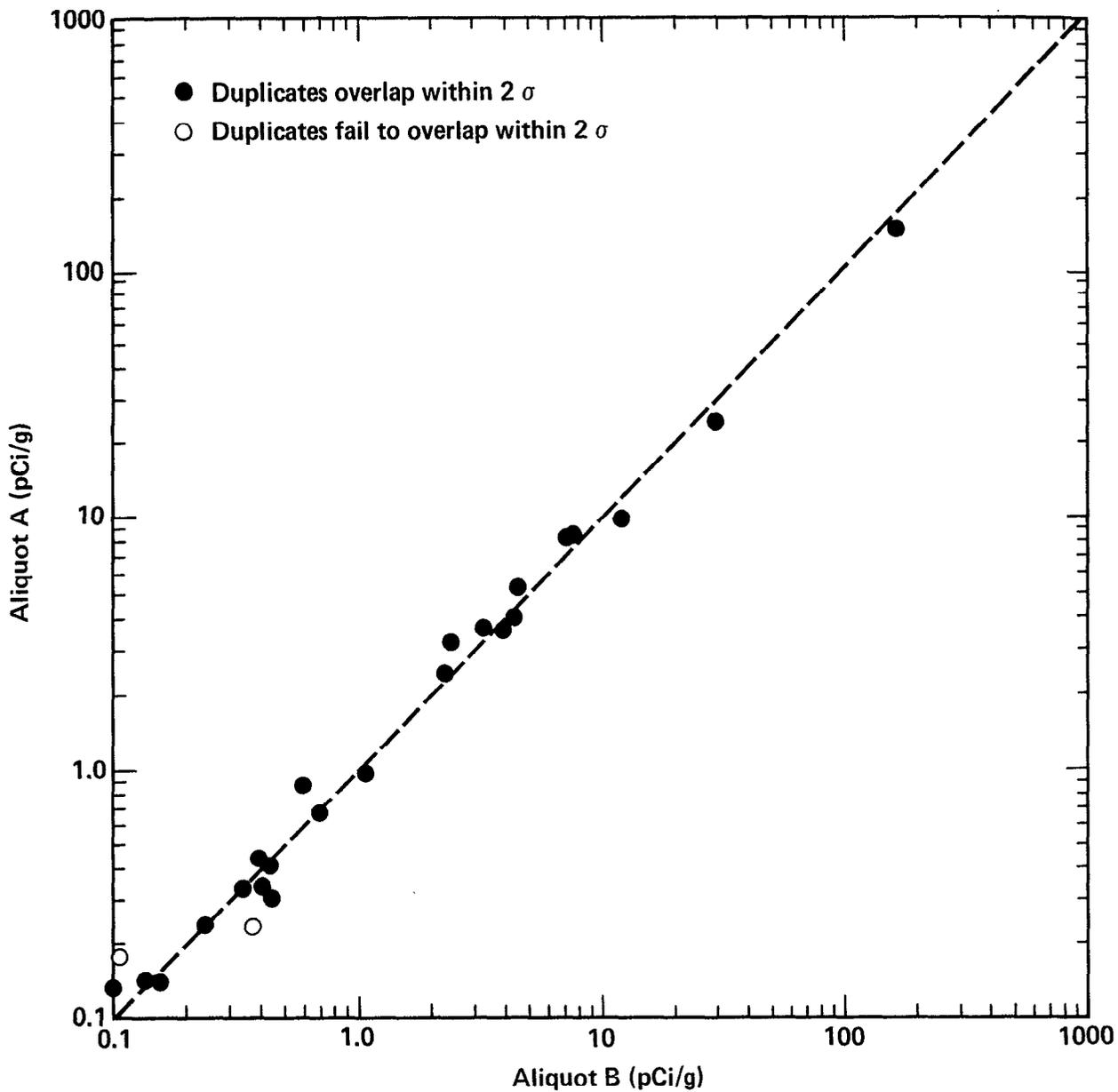


Figure A26. Acceptable quality control results for duplicate pairs analyzed for  $^{239+240}\text{Pu}$  in soil by Eberline Instrument Corporation. Results depicted are for concentrations between 0.1 and 1000 pCi/g. Broken line represents perfect agreement and is not a fit to the data.

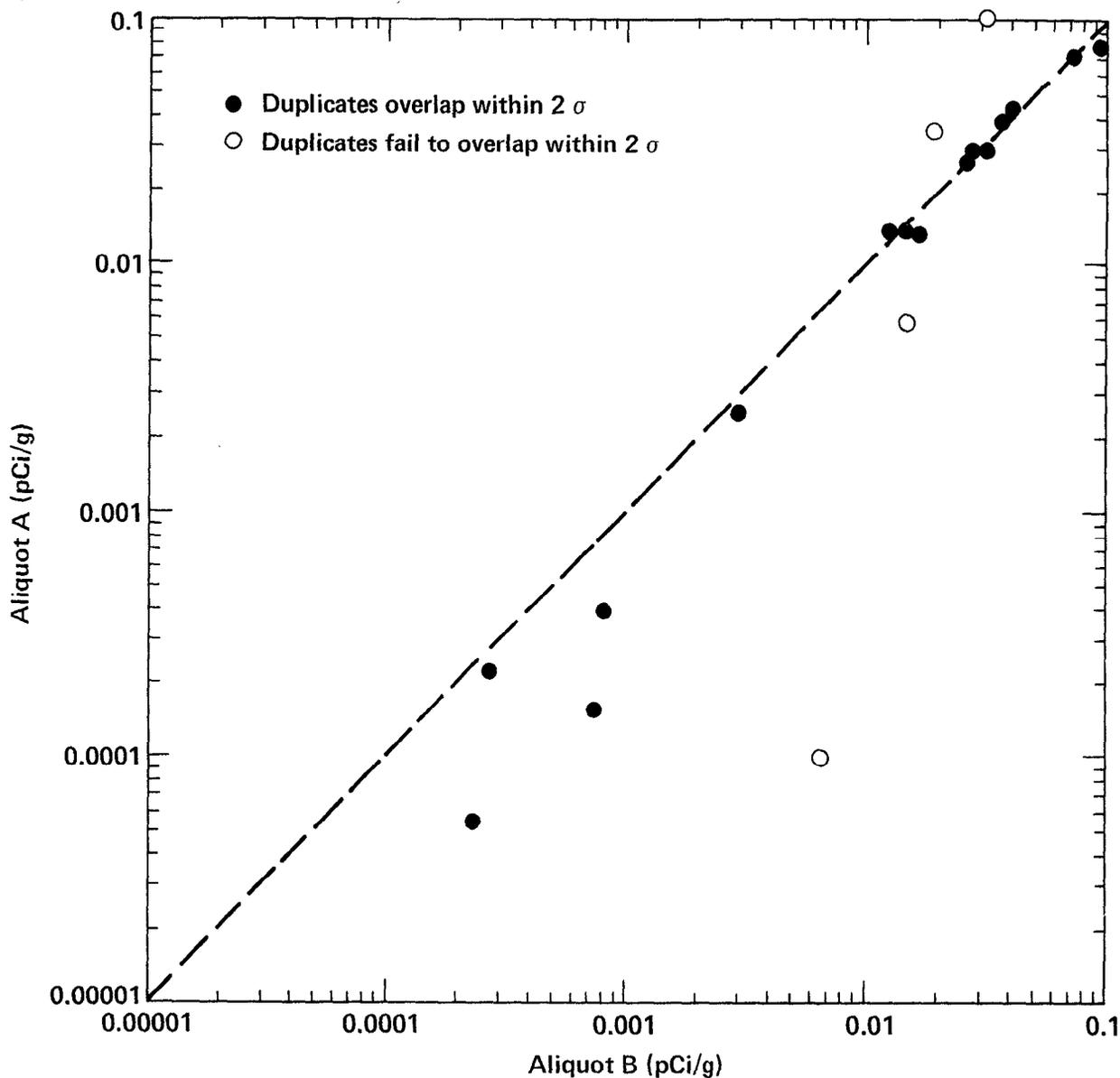


Figure A27. Acceptable quality control results for duplicate pairs analyzed for  $^{239+240}\text{Pu}$  in soil by Eberline Instrument Corporation. Results depicted are for concentrations between  $1 \times 10^{-5}$  and 0.1 pCi/g. Broken line represents perfect agreement and is not a fit to the data.

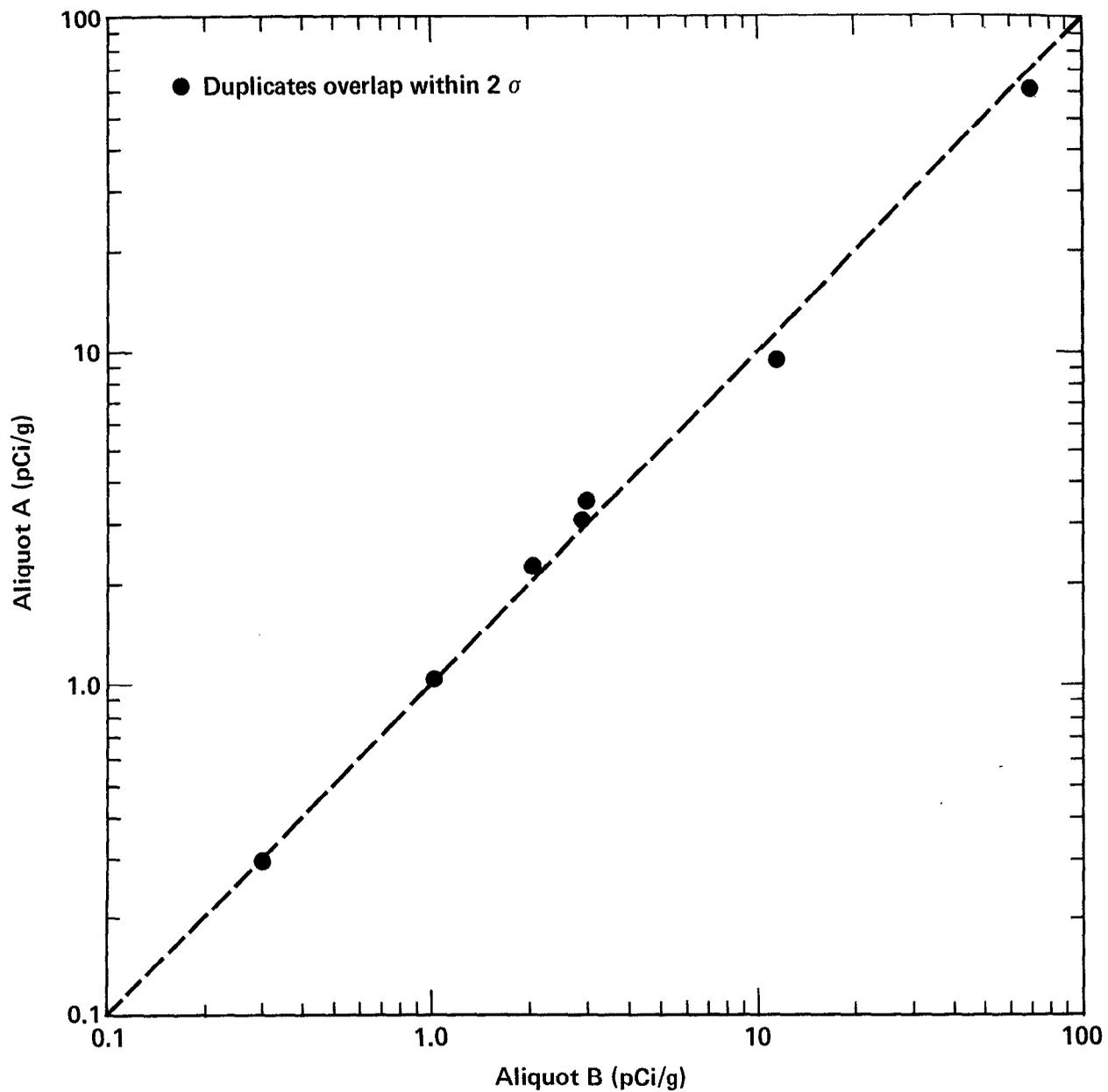


Figure A28. Acceptable quality control results for duplicate pairs analyzed for  $^{238}\text{Pu}$  in soil by Eberline Instrument Corporation. Broken line represents perfect agreement and is not a fit to the data.

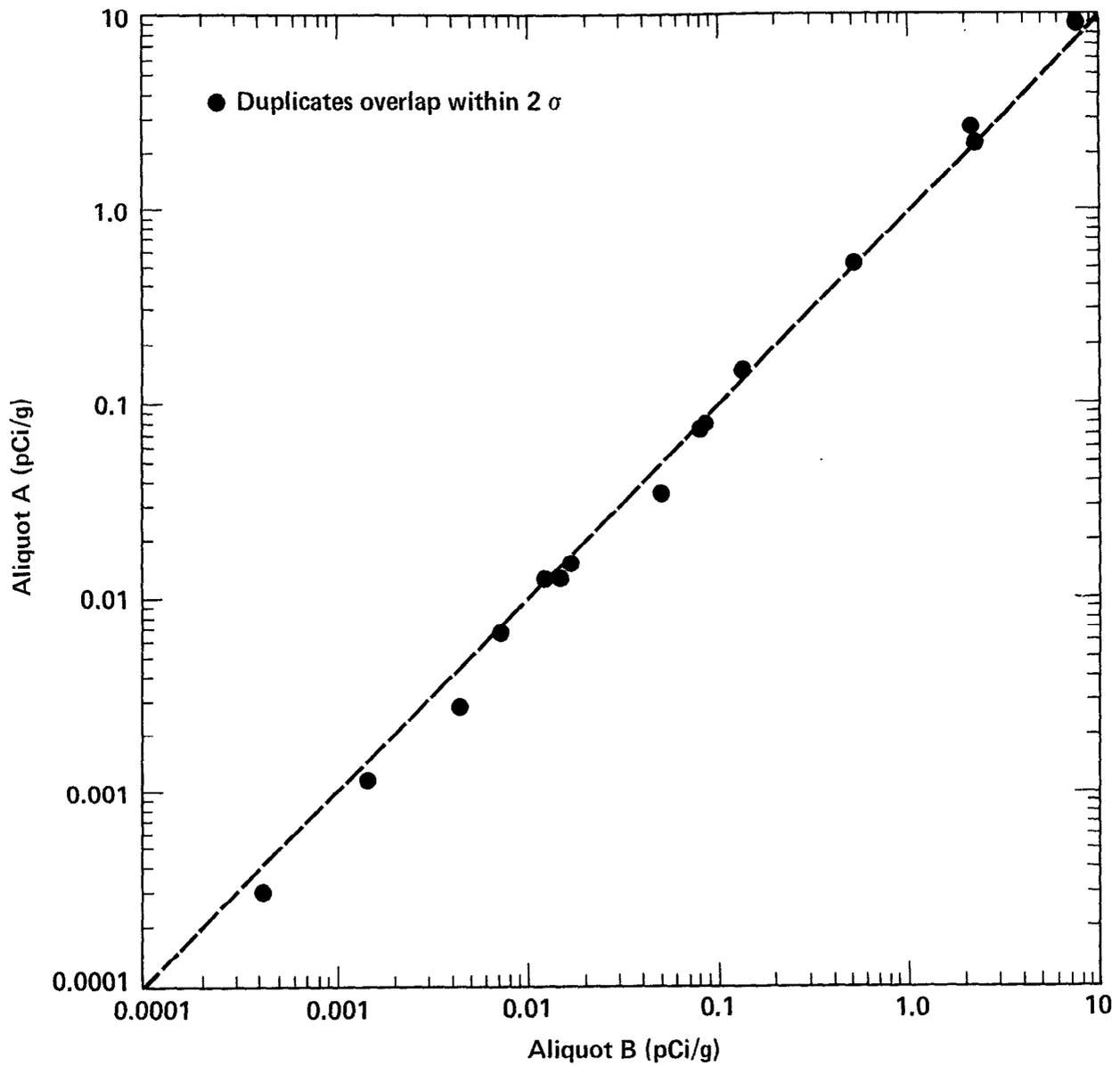


Figure A29. Acceptable quality control results for duplicate pairs analyzed for  $^{241}\text{Am}$  in soil by Eberline Instrument Corporation. Broken line represents perfect agreement and is not a fit to the data.

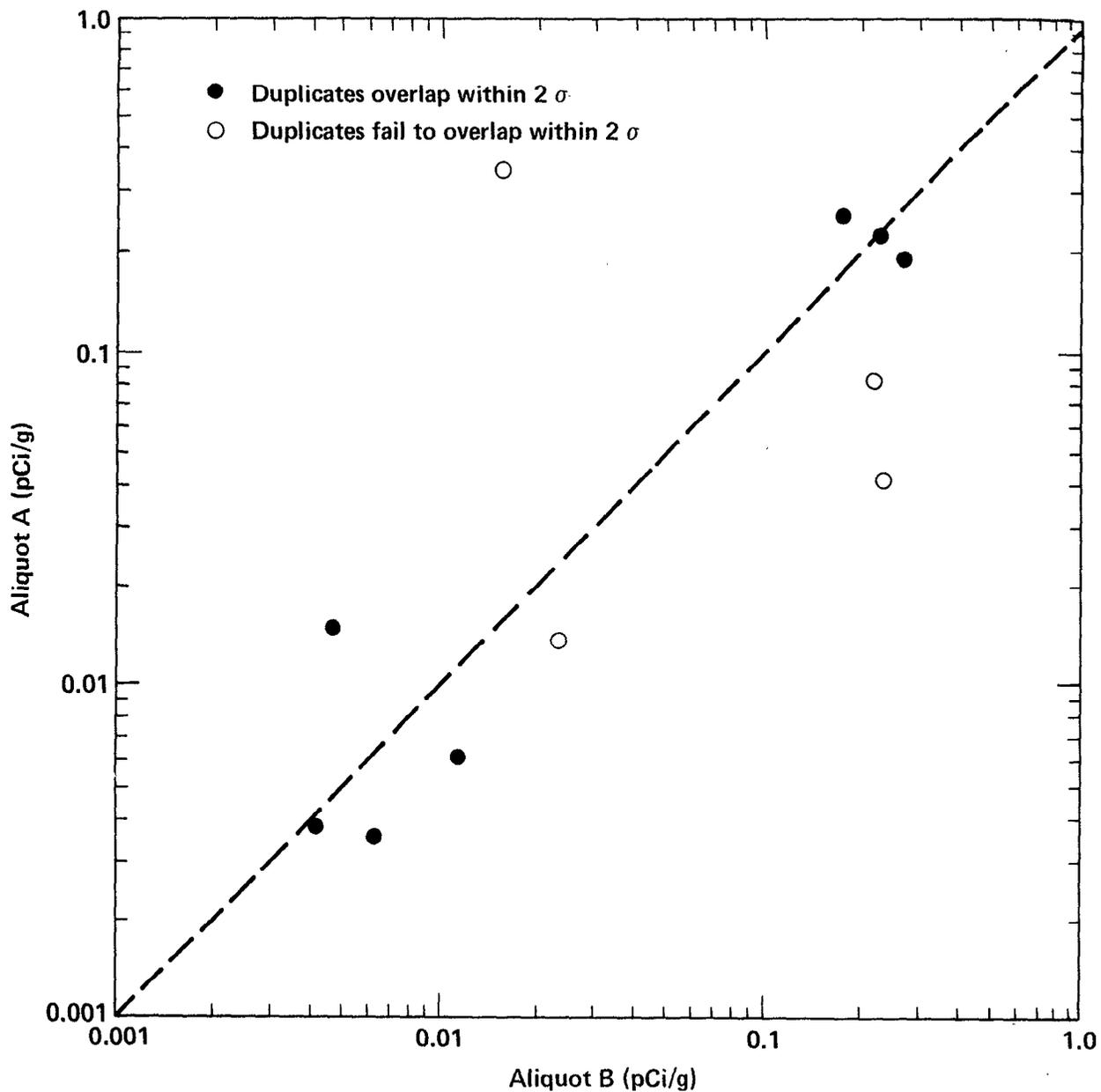


Figure A30. Acceptable quality control results for duplicate pairs analyzed for  $^{90}\text{Sr}$  in vegetation by Eberline Instrument Corporation. Broken line represents perfect agreement and is not a fit to the data. Five unacceptable pairs involving detection limits are not shown.

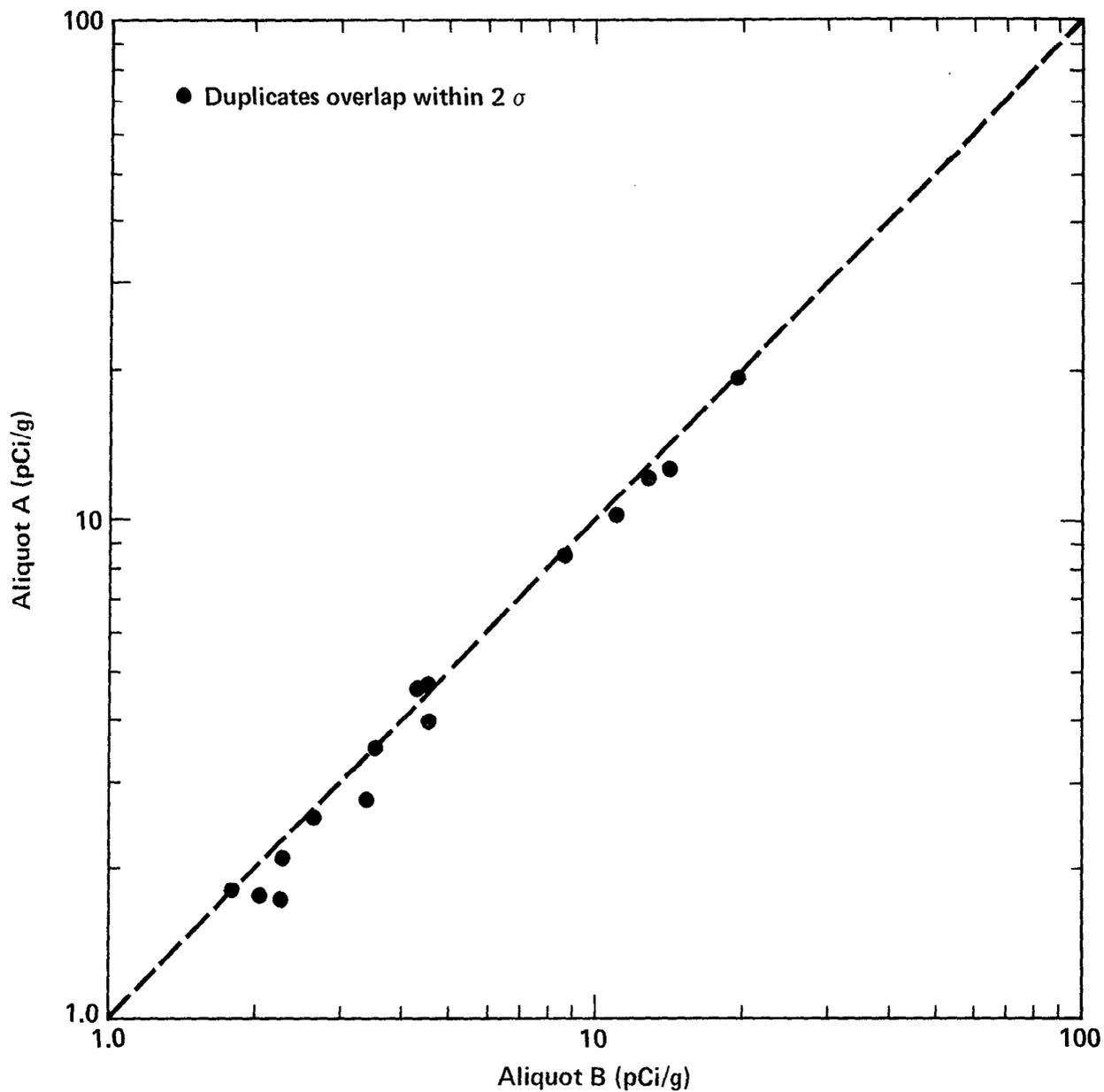


Figure A31. Acceptable quality control results for duplicate pairs analyzed for  $^{137}\text{Cs}$  in vegetation by Eberline Instrument Corporation. Broken line represents perfect agreement and is not a fit to the data.

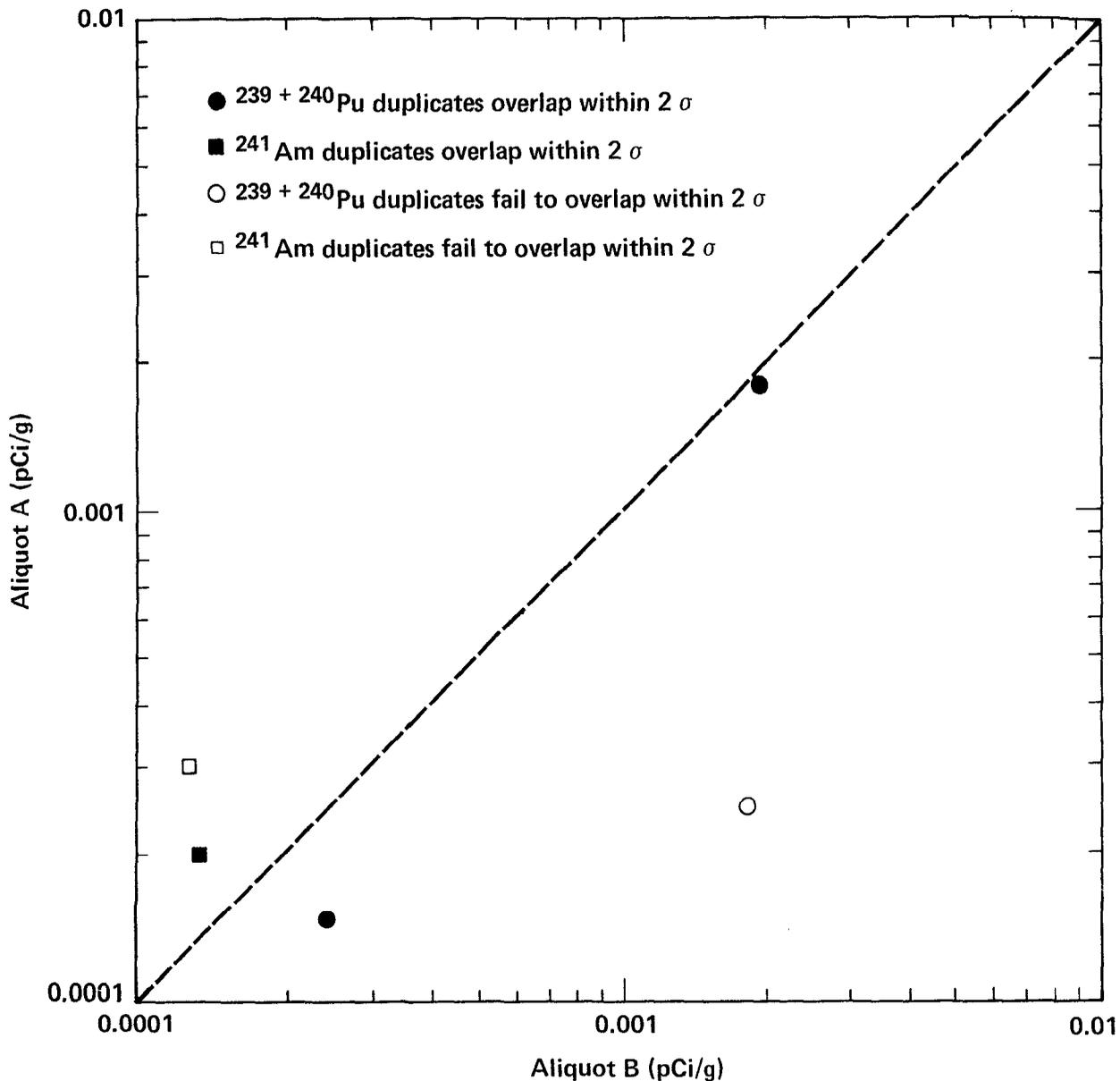


Figure A32. Acceptable quality control results for duplicate pairs analyzed for  $^{239} + ^{240}\text{Pu}$  and  $^{241}\text{Am}$  in vegetation by Eberline Instrument Corporation. Broken line represents perfect agreement and is not a fit to the data. Eight acceptable pairs (seven  $^{239} + ^{240}\text{Pu}$ ) and six unacceptable pairs (three  $^{239} + ^{240}\text{Pu}$ ) involving zero or negative concentrations or detection limits are not shown.

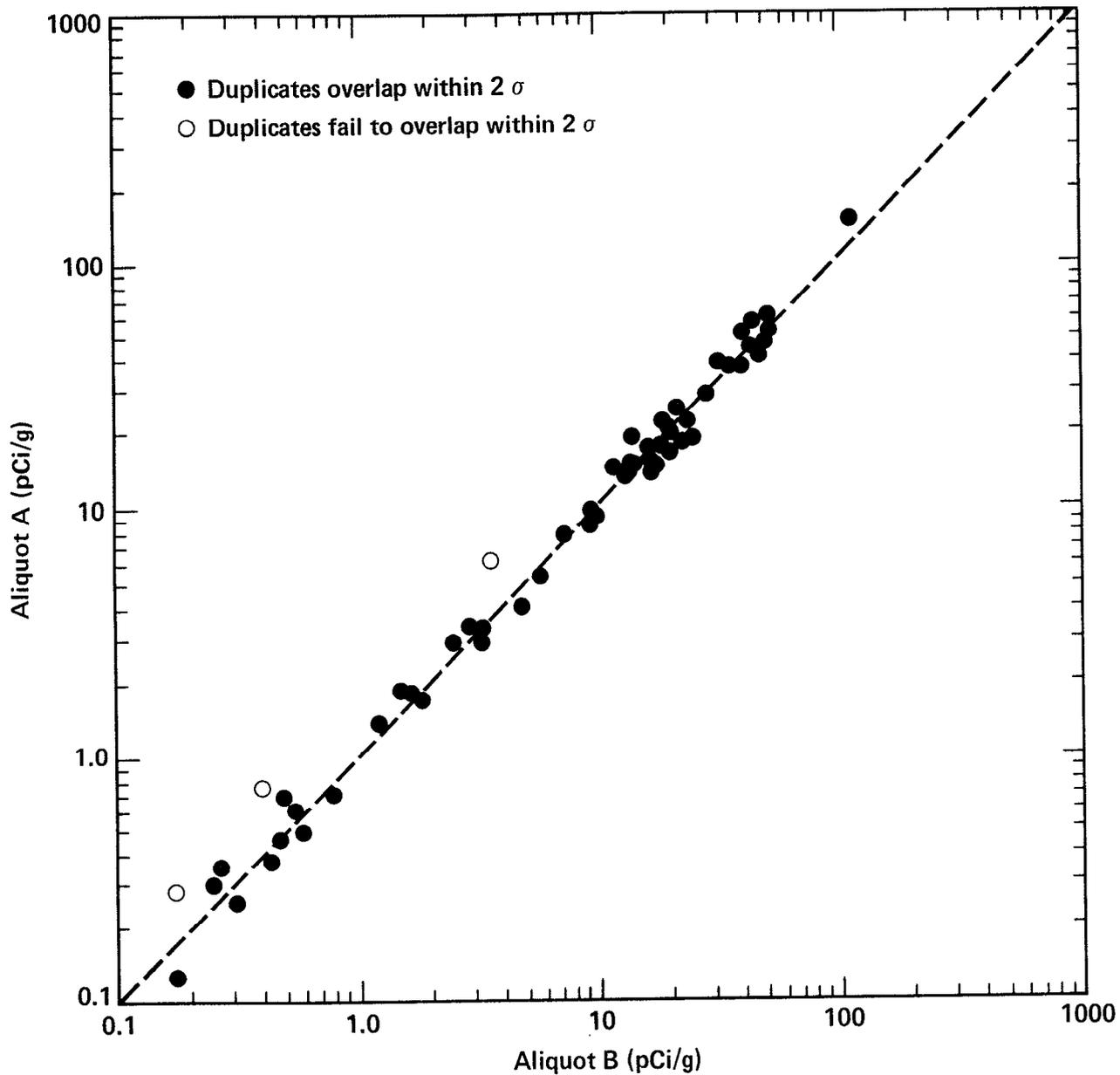


Figure A33. Acceptable quality control results for duplicate pairs analyzed for  $^{90}\text{Sr}$  in soil by Laboratory of Radiation Ecology. Broken line represents perfect agreement and is not a fit to the data. Two unacceptable pairs involving detection limits are not shown.

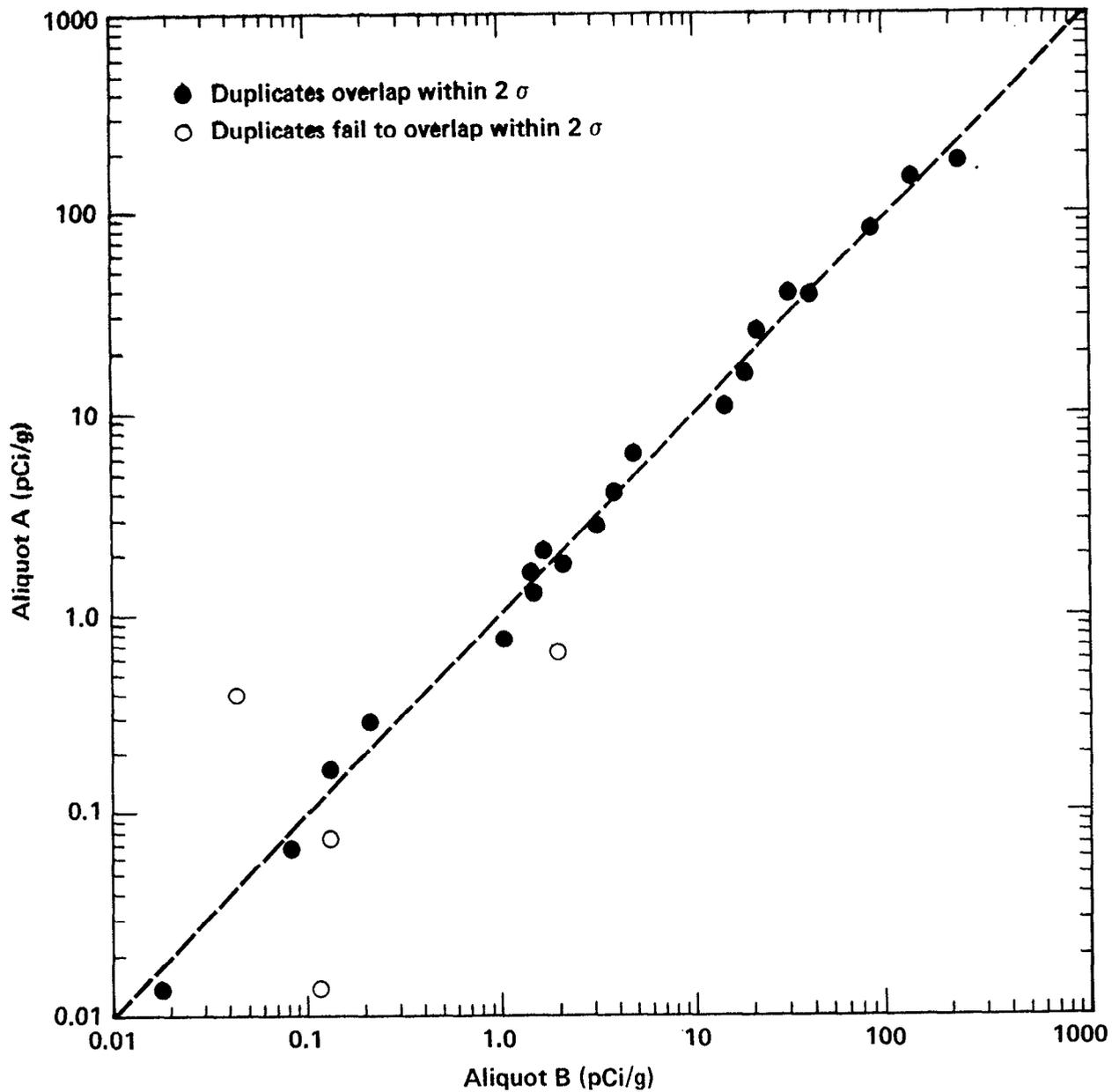


Figure A34. Acceptable quality control results for duplicate pairs analyzed for  $^{137}\text{Cs}$  in soil by Laboratory of Radiation Ecology. Broken line represents perfect agreement and is not a fit to the data. Three pairs (two acceptable) involving detection limits are not shown.

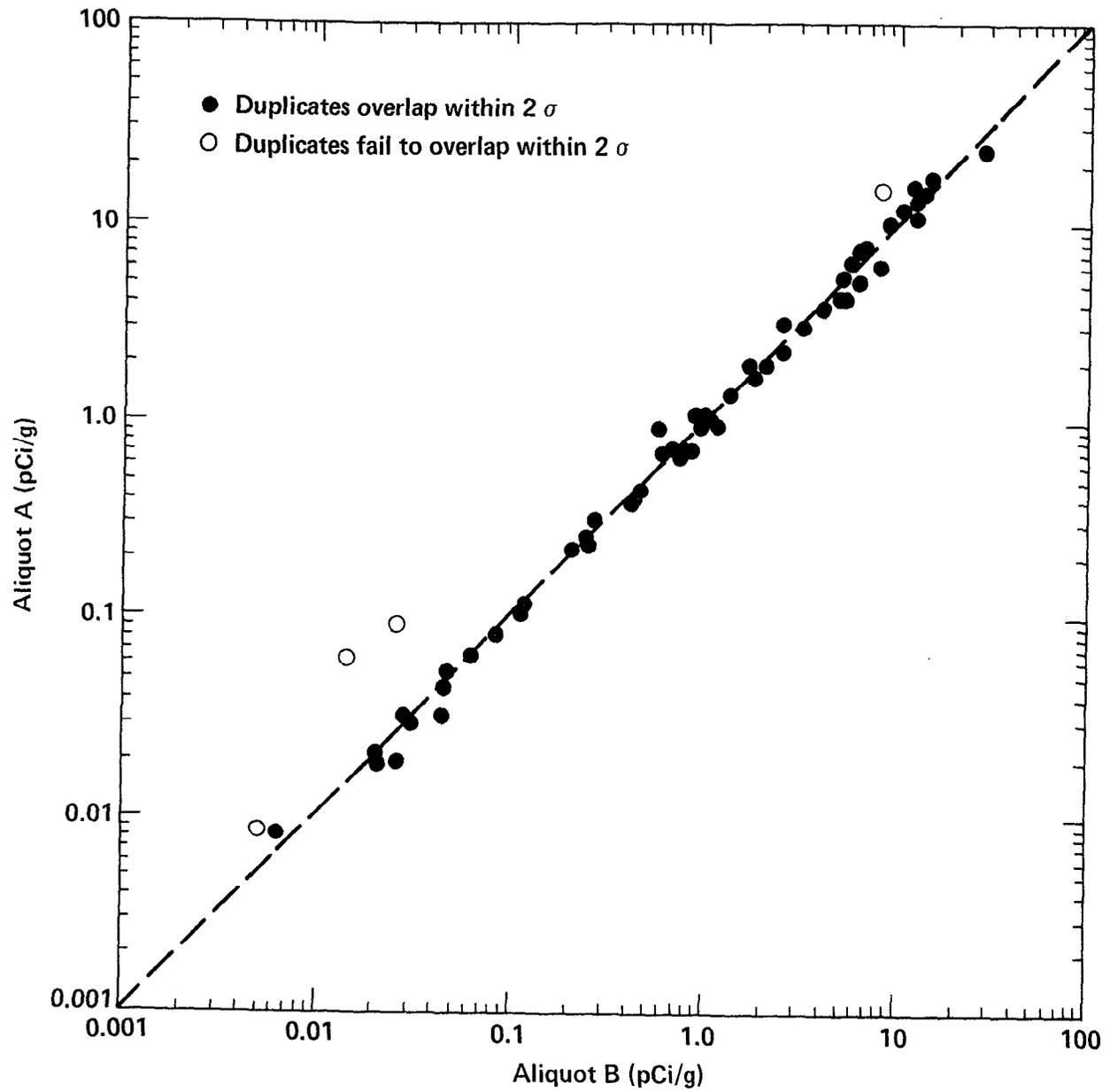


Figure A35. Acceptable quality control results for duplicate pairs analyzed for  $^{239+240}\text{Pu}$  in soil by Laboratory of Radiation Ecology. Broken line represents perfect agreement and is not a fit to the data.

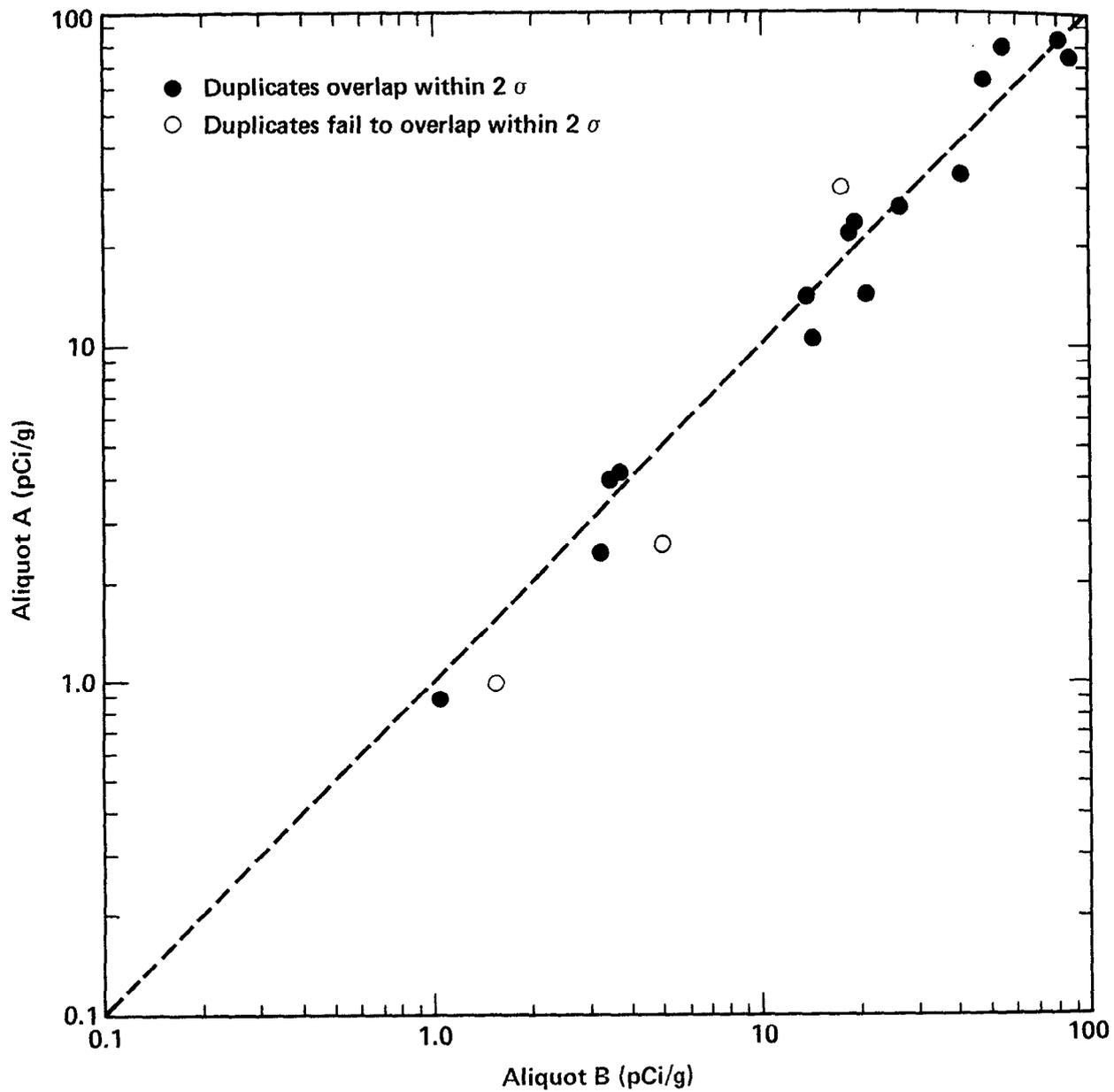


Figure A36. Acceptable quality control results for duplicate pairs analyzed for  $^{241}\text{Pu}$  in soil by Laboratory of Radiation Ecology. Broken line represents perfect agreement and is not a fit to the data.

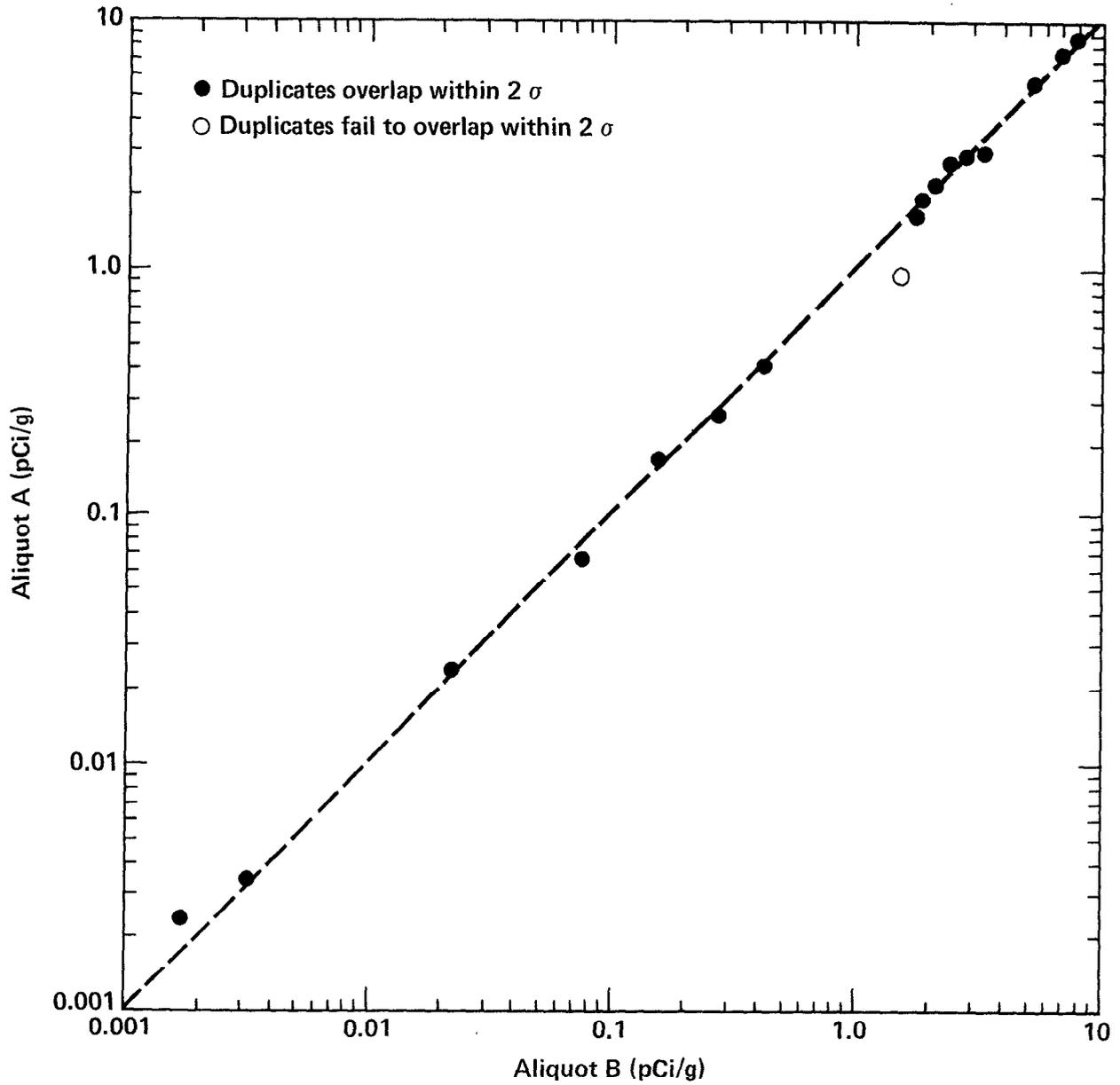


Figure A37. Acceptable quality control results for duplicate pairs analyzed for  $^{241}\text{Am}$  in soil by Laboratory of Radiation Ecology. Broken line represents perfect agreement and is not a fit to the data.

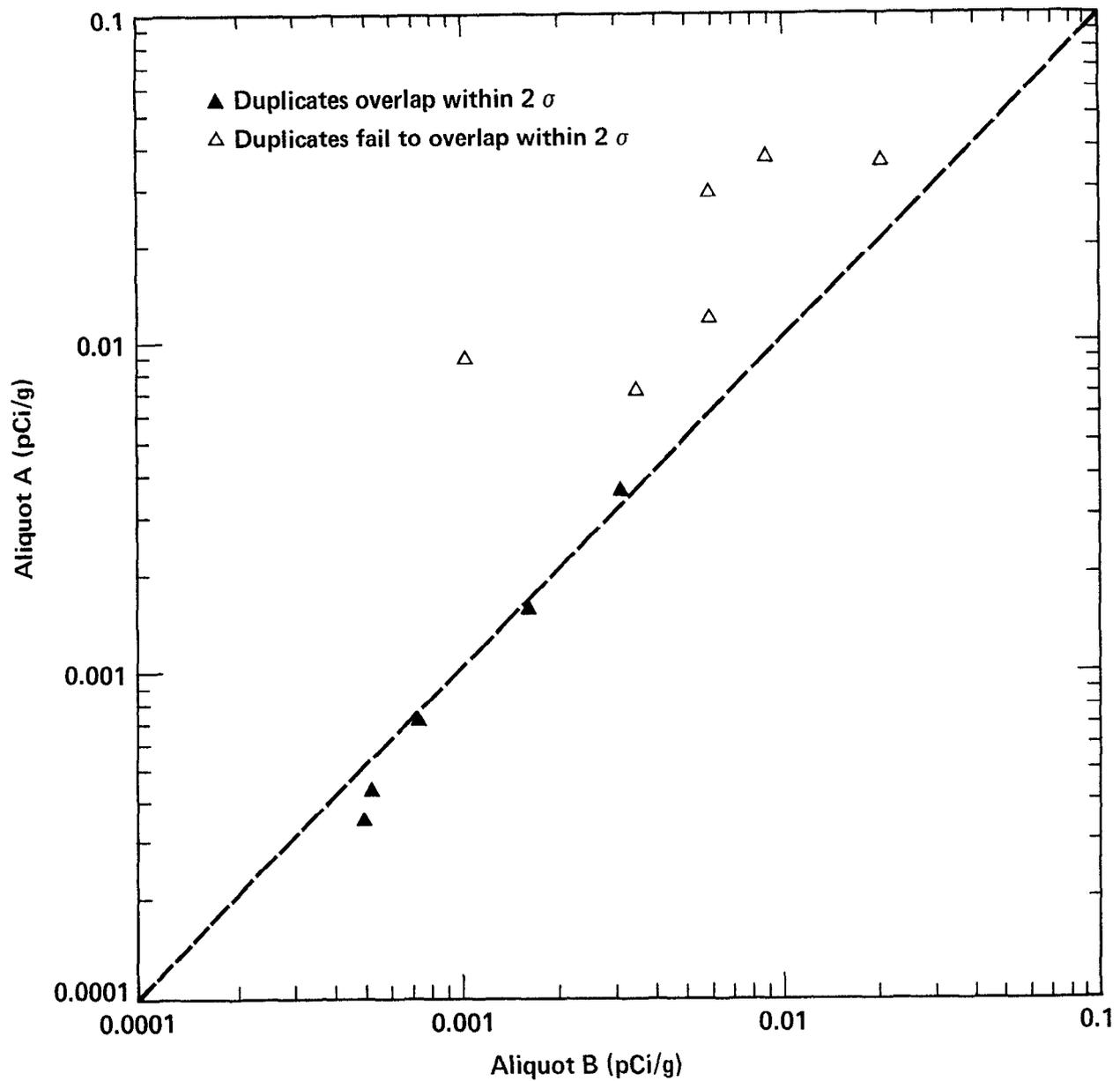


Figure A38. Unacceptable quality control results for duplicate pairs analyzed for  $^{239+240}\text{Pu}$  in soil by Environmental Analysis Laboratory. Broken line represents perfect agreement and is not a fit to the data. One acceptable pair involving zero concentrations is not shown.

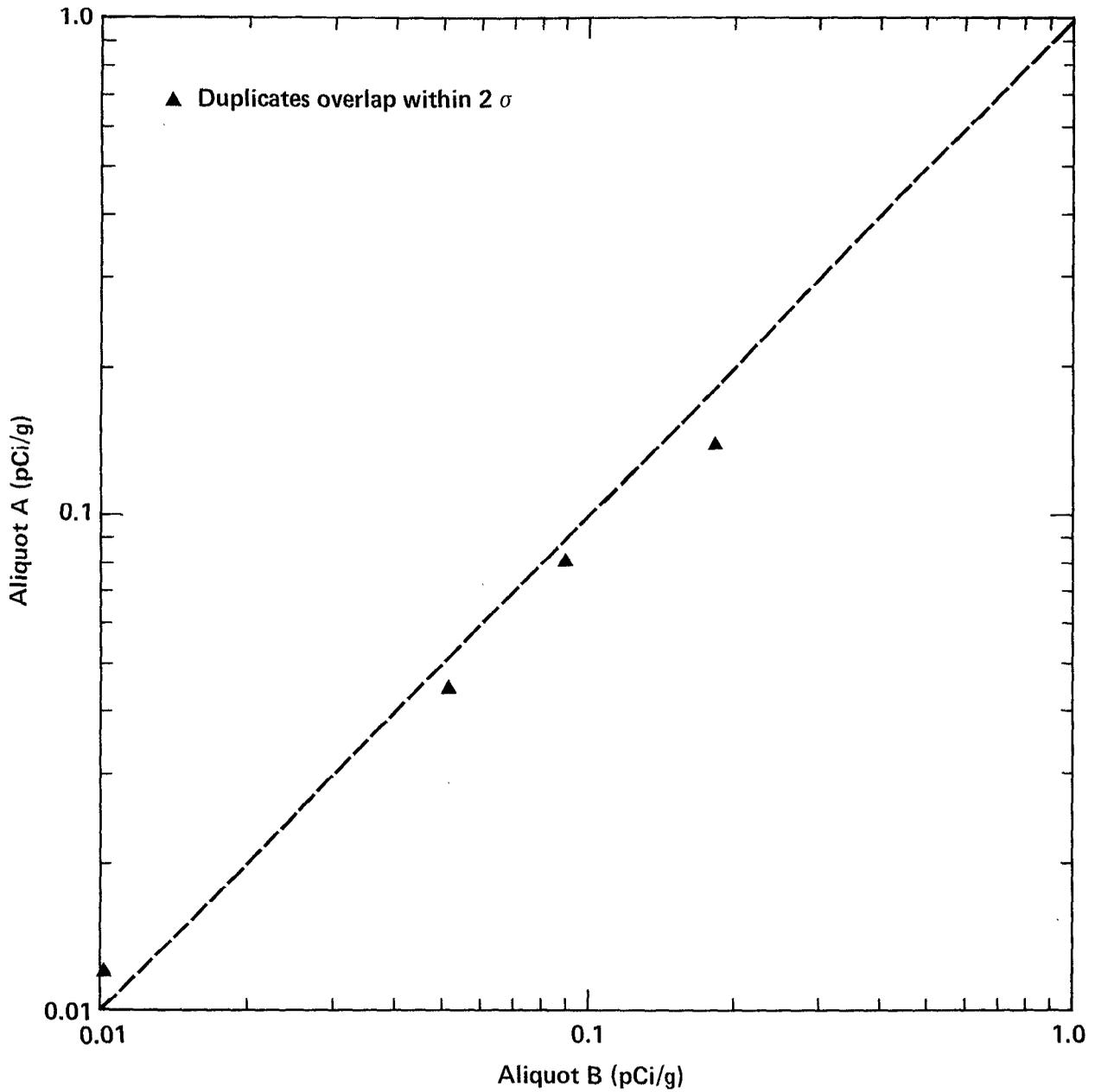


Figure A39. Unacceptable quality control results for duplicate pairs analyzed for  $^{90}\text{Sr}$  in marine sediment by Environmental Analysis Laboratory. Broken line represents perfect agreement and is not a fit to the data. Three acceptable pairs involving zero concentrations are not shown.

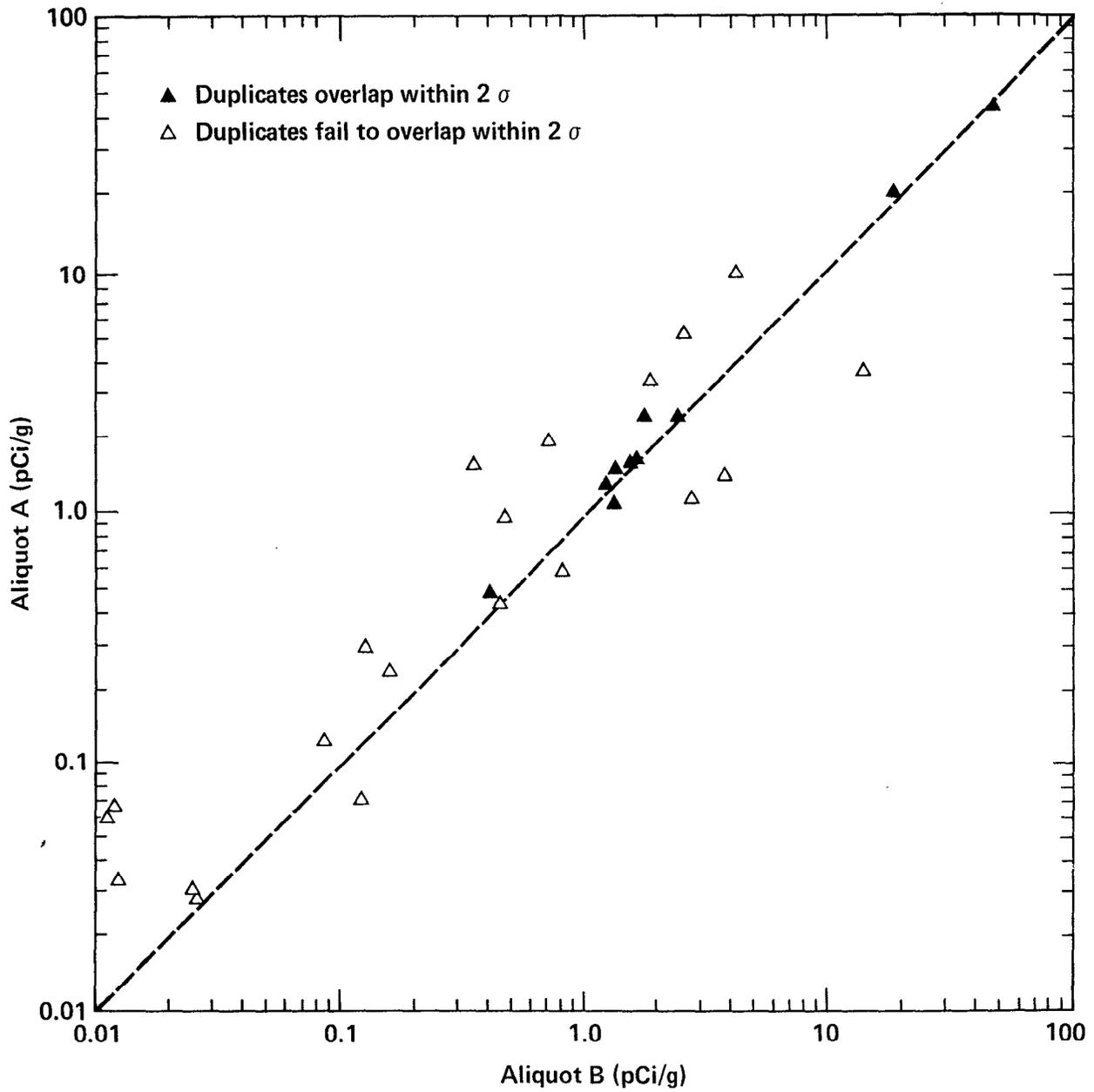


Figure A40. Unacceptable quality control results for duplicate pairs analyzed for  $^{90}\text{Sr}$  in soil by Eberline Instrument Corporation. Broken line represents perfect agreement and is not a fit to the data. Two unacceptable pairs involving negative concentrations are not shown.

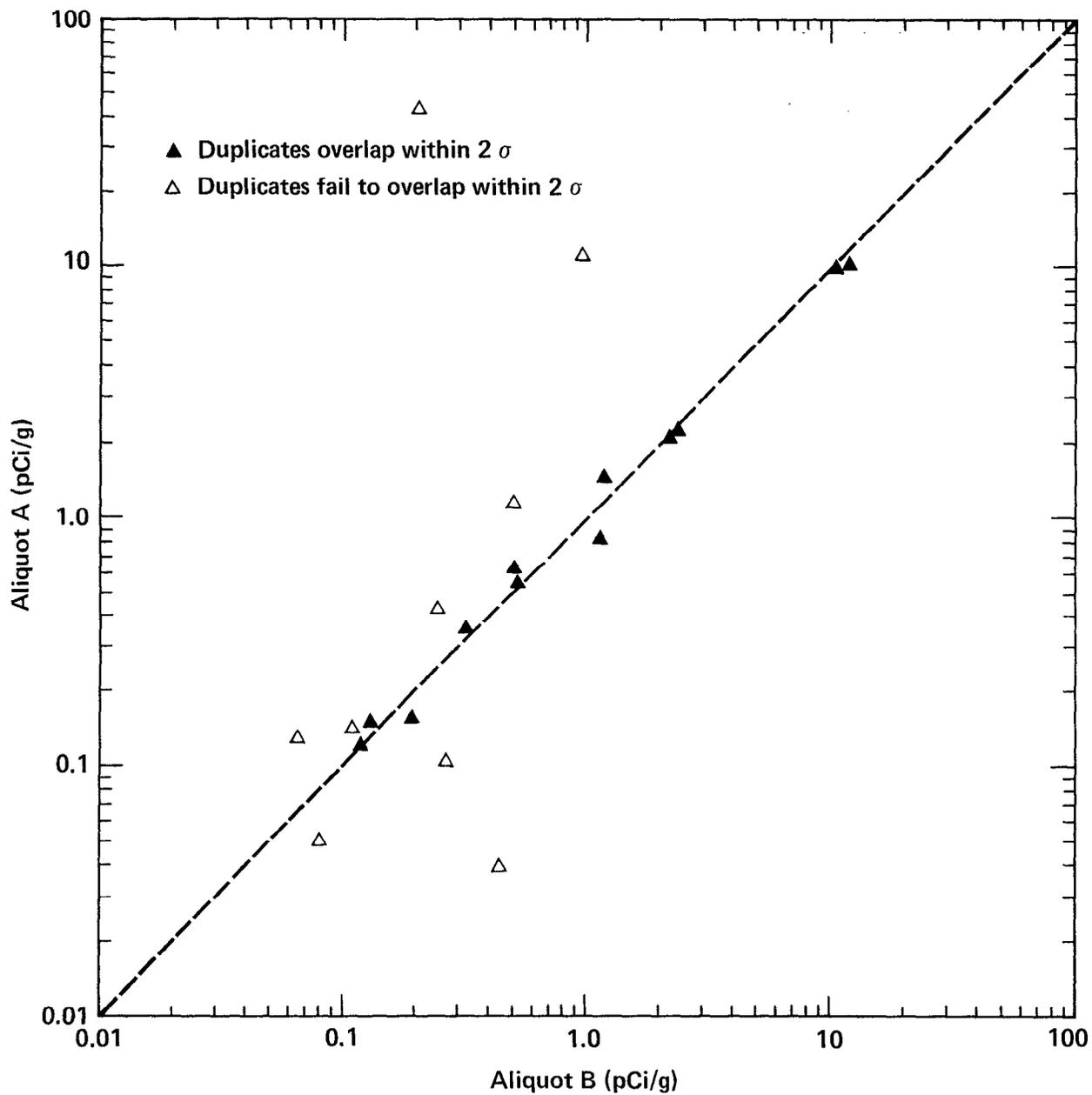


Figure A41. Unacceptable quality control results for duplicate pairs analyzed for  $^{137}\text{Cs}$  in soil by Eberline Instrument Corporation. Broken line represents perfect agreement and is not a fit to the data. Three unacceptable pairs involving detection limits are not shown.

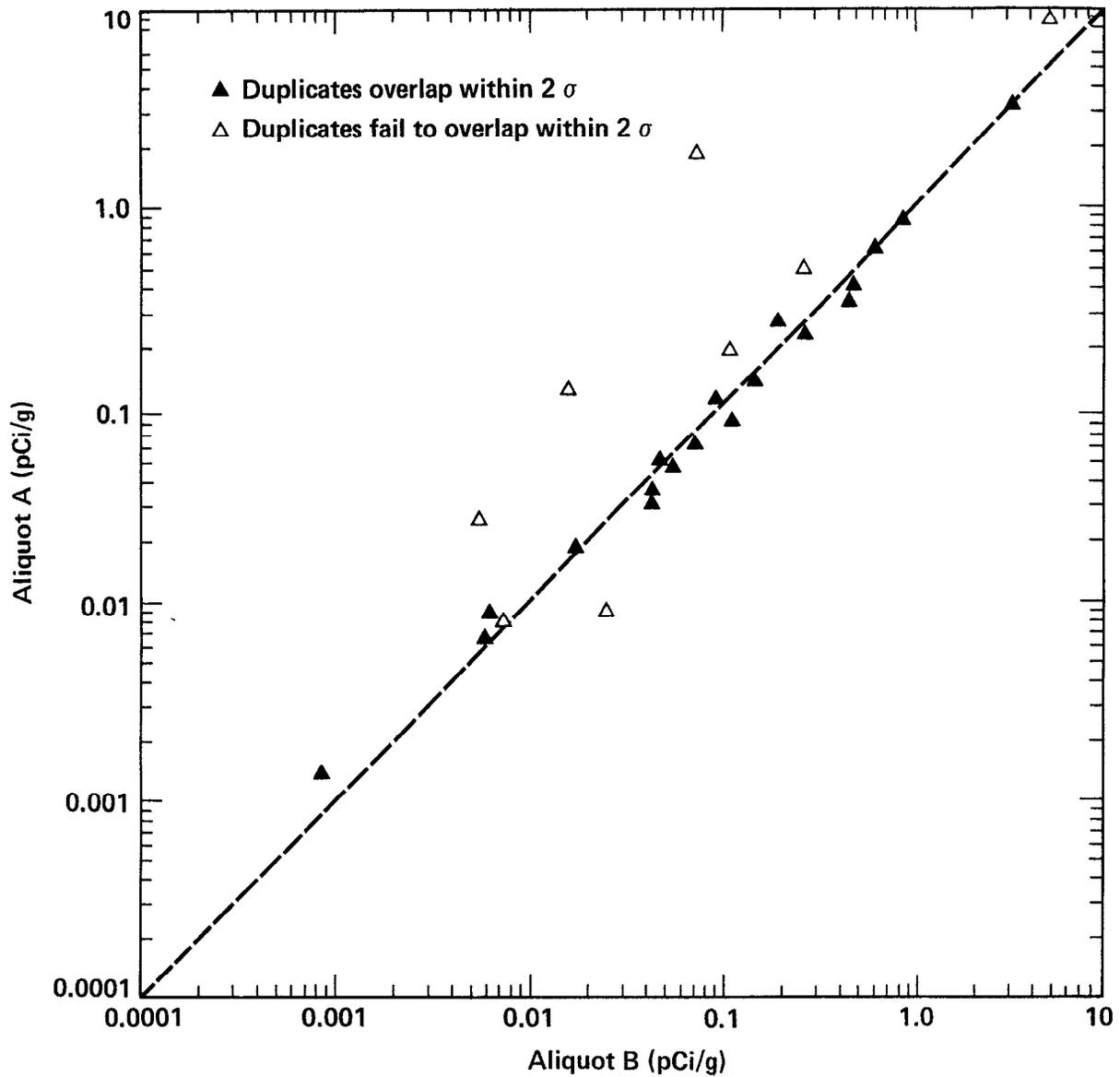


Figure A42. Unacceptable quality control results for duplicate pairs analyzed for  $^{239+240}\text{Pu}$  in soil by Eberline Instrument Corporation. Broken line represents perfect agreement and is not a fit to the data.

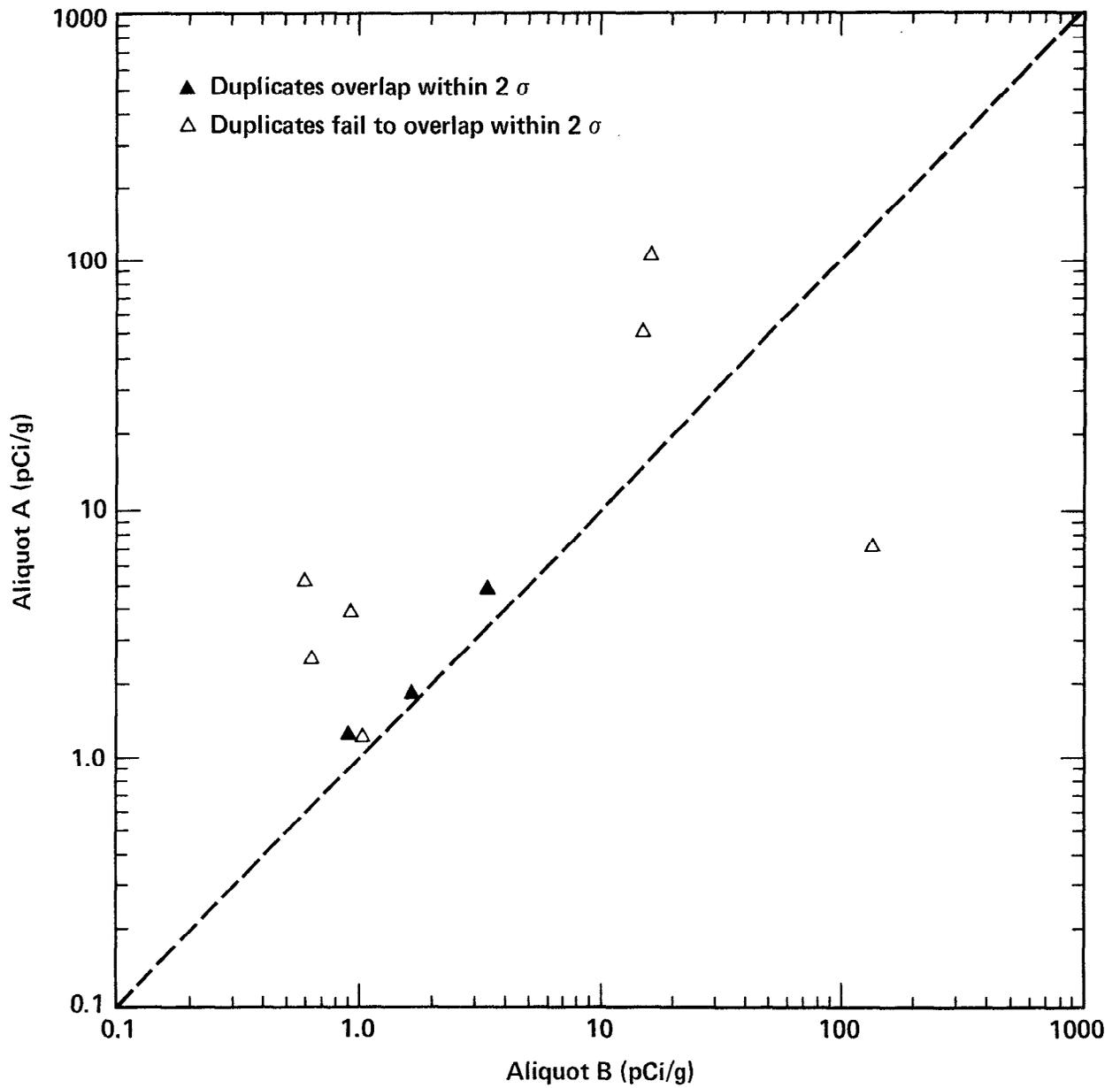


Figure A43. Unacceptable quality control results for duplicate pairs analyzed for  $^{241}\text{Pu}$  in soil by Eberline Instrument Corporation. Broken line represents perfect agreement and is not a fit to the data. Twelve unacceptable pairs involving detections limits are not shown.

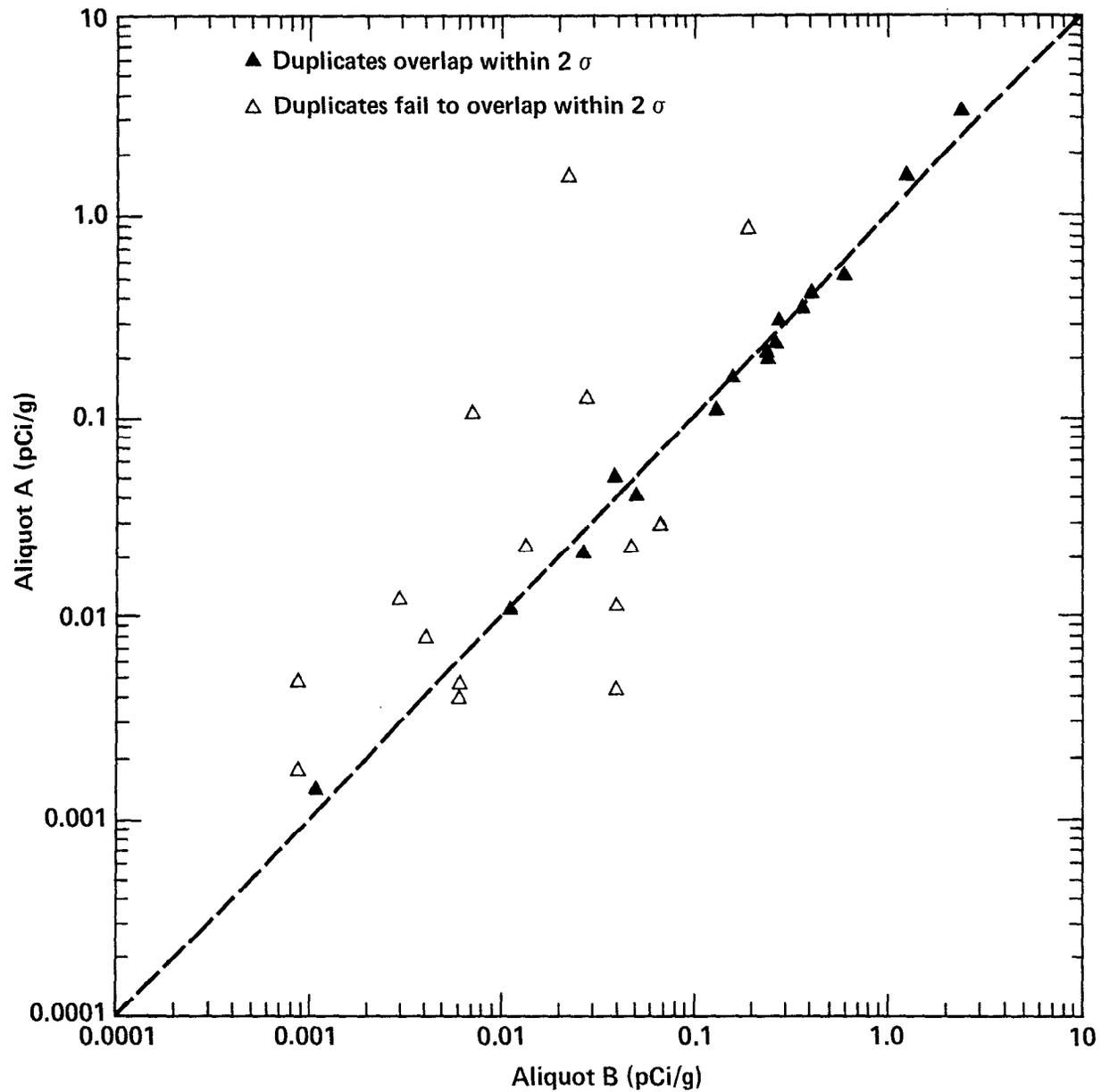


Figure A44. Unacceptable quality control results for duplicate pairs analyzed for  $^{241}\text{Am}$  in soil by Eberline Instrument Corporation. Broken line represents perfect agreement and is not a fit to the data. Five unacceptable pairs involving negative concentrations or detection limits are not shown.

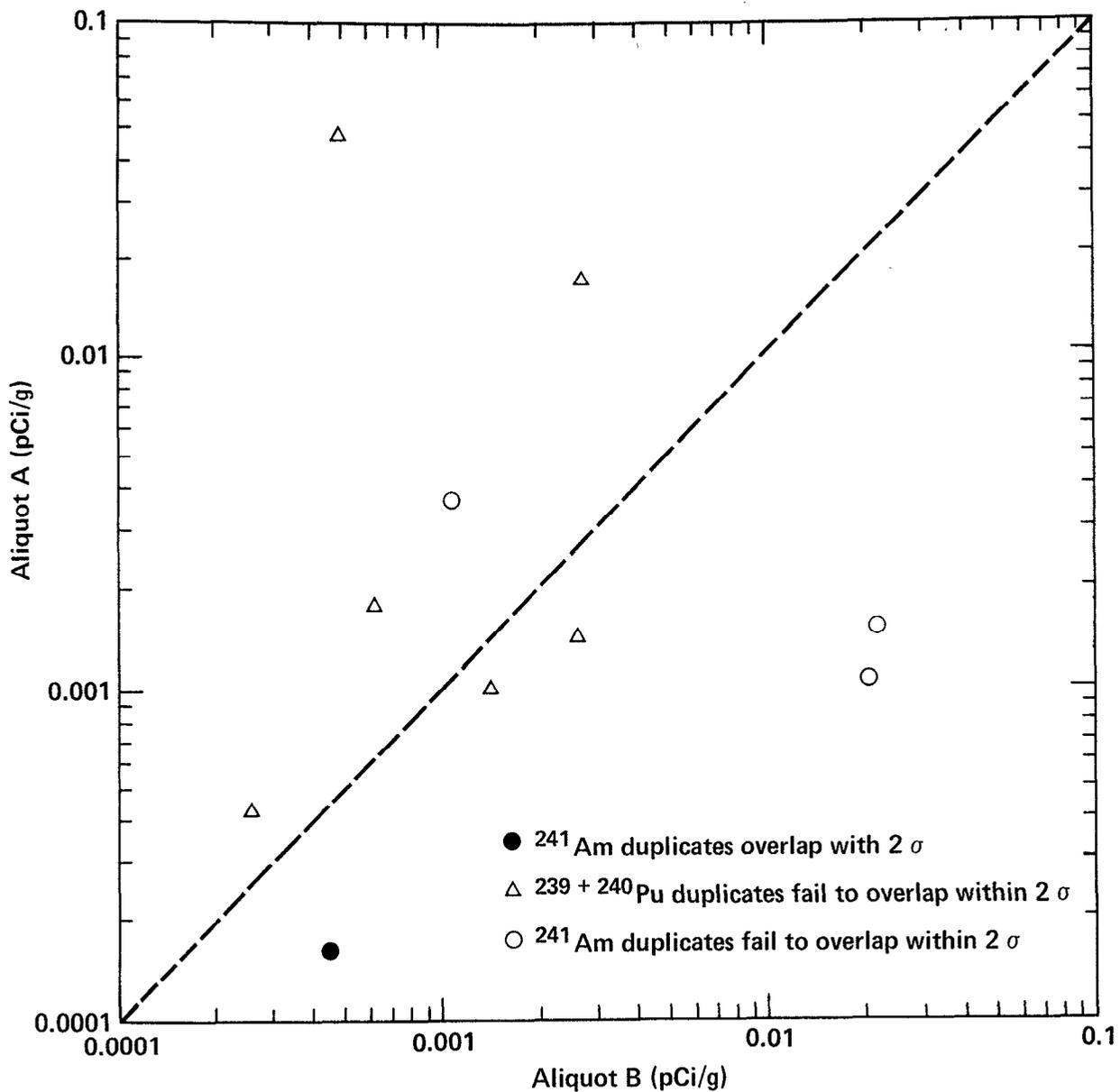


Figure A45. Unacceptable quality control results for duplicate pairs analyzed for  $^{239} + ^{240}\text{Pu}$  and  $^{241}\text{Am}$  in vegetation by Eberline Instrument Corporation. Broken line represents perfect agreement and is not a fit to the data. Ten unacceptable pairs (nine  $^{241}\text{Am}$ ) and two acceptable pairs (both  $^{241}\text{Am}$ ) involving negative concentrations or detection limits are not shown.