KfK 3149
ESARDA 1/81
Juli 1981

Results from an Interlaboratory Exercise on the Determination of Plutonium Isotopic Ratios by Gamma Spectrometry

Report of the ESARDA Working Group on Techniques and Standards for Nondestructive Analysis

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RESULTS FROM AN INTERLABORATORY EXERCISE ON THE DETERMINATION OF PLUTONIUM ISOTOPIC RATIOS BY GAMMA SPECTROMETRY

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Submitted Under the Terms of the ESARDA-Agreement
No. 1423/80/12 (CG-ISP)

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Kernforschungszentrum Karlsruhe GmbH
ISSN 0303-4003

ABSTRACT

Results from interlaboratory comparison measurements on the determination of plutonium isotopic ratios by gamma spectrometry, organized by the ESARDA Working Group on Techniques and Standards for Nondestructive Analysis, are presented and discussed. Nine laboratories from nine countries or international organizations participated in the intercomparison exercise, which included both laboratories' own measurements on the plutonium isotopic reference materials NBS-SRM 946, 947, 948 and comparison analyses of gamma spectra from these materials distributed to the participating laboratories. Results from the intercomparison analyses have been used to reevaluate some gamma branching intensity ratios required for plutonium isotopic ratio measurements.

ZUSAMMENFASSUNG

ERGEBNISSE VON EINEM INTERLABORATORIUM-VERGLEICHSTEST ZUR BESTIMMUNG VON PLUTONIUMISOTOPENVERHÄLTNISSEN DURCH GAMMASPEKTROMETRIE

Unter Teilnahme von 9 Laboratorien aus 9 verschiedenen Ländern oder internationalen Organisationen führte die ESARDA-Arbeitsgruppe "Meßmethoden und Standards für Zerstörungsfreie Analyse" einen Interlaboratorium-Vergleichstest zur Bestimmung von Plutoniumisotopenverhältnissen mittels Gammaspektrometrie durch. Das Vorhaben beinhaltete sowohl Vergleichsmessungen an den Plutonium-Isotopenstandards NBS-SRM 946, 947, 948 als auch Vergleichsanalysen an Referenzspektren von diesen Standards. Die Durchführung des Vergleichstests und die gewonnenen Ergebnisse werden beschrieben. Die Resultate der Vergleichsmessungen wurden verwendet zur Neubestimmung von einigen Gammaverzweigungsverhältnissen, die für die gammaspektrometrische Plutoniumisotopenanalyse von Bedeutung sind.

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continued

PREFACE AND ACKNOWLEDGEMENT

The present report summarizes the results from interlaboratory comparison measurements of plutonium isotopic ratios by gamma spectrometry which have been initiated and organized by the ESARDA Working Group on Techniques and Standards for Non-Destructive Analysis. The decision for this experiment dates back to 1977, where the Working Group convened a meeting of specialists from ESARDA on this subject at ECN Petten, the Netherlands. It has then been concluded that an intercomparison exercise on plutonium isotopic ratio measurements by high-resolution gamma spectrometry could helpfully contribute to the process of maturing this NDA technique which in recent years has gained relevance to the accounting and safeguarding of plutonium materials.

Originally intended as a very limited intercomparison exercise among members of ESARDA, the scope of the final interlaboratory experiment has then been enlarged to some extent during the initial stage of the project by inviting other laboratories which previously had gained some experience on the matter. Finally, 9 laboratories have notified their participation.

The present report documents an intermediate stage of the Working Group's continuing efforts on the further evaluation and implementation of the non-destructive plutonium isotopic composition analysis. Those efforts are including both the specification and preparation of suitable physical standards as well as further intercomparison measurements through cooperative international programmes, of which two will be mentioned:

- On the invitation of Dr. C. Beets of the CEN/SCK, Mol, Belgium, an intercomparison was made of gamma and neutron measurements on cans of plutonium oxide containing either 0.5 or 3 kg Pu. The measurements were performed at the plant of Belgo Nucléaire in Dessel, Belgium. The results will be published around the end of this year.

- Upon the request of the Working Group new sets of plutonium isotopic intercomparison materials with an extended range of isotopic composition are currently under preparation at AERE Harwell. Those intercomparison materials will be available from Dr. A. Adamson to interested parties by the end of 1981 for further intercomparison measurements under the name of PIDIE, i.e. Plutonium Isotopic Determination Intercomparison Exercise. The samples containing about 0.5 g of Pu in a defined geometry will offer some possibilities for improvement of the measurement technique, because also it is foreseen that the material will be subjected to careful mass spectrometry analysis by the participating laboratories in parallel.

The ESARDA Working Group on Techniques and Standards for Non-Destructive Analysis gratefully acknowledges the efforts of Dr. H. Ottmar who organized the first international intercomparison study on the determination of plutonium isotopic ratios by gamma spectrometry, of which this report is the final result. In this acknowledgement we also want to include Mr. H. Eberle, staff member of KfK, who supported diligently in the compilation of the results, the analyses of the data, and the preparation of the final presentation of the results in this report. Also we are thankful to the participants in this intercomparison who contributed with their results to the completion of this first step of the ESARDA Working Group on NDA in the exploration of the possibilities of gamma spectrometry for the determination of the isotopic composition of plutonium.

Petten, April 1981.

Jörn Harry,
Convener ESARDA Working Group on
Techniques and Standards for Non-Destructive Analysis.

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I. PURPOSE AND SCOPE

A. Purpose of the Experiment

The determination of the plutonium isotopic composition by high-resolution gamma spectrometry is receiving attention for many years among the NDA techniques for nuclear materials measurements. Although now in use at several places, the technique is still in a developing stage rather than being a standardized routine method. Both feasibility studies and practical experiences with the technique have shown that there is no unique solution to plutonium isotopic composition measurements using gamma spectrometry in view of the very different plutonium materials available in the nuclear fuel cycle. For this reason the evaluation of the method and its adaptation to particular measurement problems is subject of continuing R&D work at several laboratories.

The measurement of plutonium isotopic ratios by gamma spectrometry is based on the specific gamma radiation emitted by the isotopes ^{238}Pu, ^{239}Pu, ^{240}Pu and ^{241}Pu. The isotope ^{242}Pu fails to emit a detectable gamma-ray signature. The method offers the advantage that it can determine the parameters of interest from an intrinsic calibration without the use of external standards. The isotopic ratio $N(i) / N(k)$ of isotopes i and k simply deduces from the relation

$$\frac{N(i)}{N(k)} = \frac{A(i)}{A(k)} \cdot \frac{B(k) \cdot T_{1/2}(i)}{B(i) \cdot T_{1/2}(k)} \cdot \frac{\varepsilon(k)}{\varepsilon(i)} ,$$

where the peak areas, A, and the relative detection efficiencies, ε , are determined from the measured gamma spectrum. The gamma branching intensities, B, and isotope half lives, $T_{1/2}$, are external input data required for the analysis.

The major error sources which can occur in the isotopic ratio determination arise from possible systematic and random errors associated with the adopted methods for the evaluation of peak areas and detection efficiency, and from possible systematic errors associated with the required nuclear data.

It has been the aim of the present intercomparison measurements to provide some orientation about the aforementioned various error sources, in particular:

i) to investigate the systematic error sources from the various procedures of data reduction adopted by the different laboratories, and

ii) to test the validity and to refine, if possible, the available nuclear data required for the analysis.

B. Scope of the Experiment

The present interlaboratory experiment refers to the case of intrinsically calibrated plutonium isotopic ratio measurements on samples of arbitrary physical form. In order to establish a directly comparable set of measurement results, isotopically identical plutonium materials had to be analyzed, and an agreed common set of nuclear data had to be used for the isotopic ratio evaluation from the measured spectra.

For some practical reasons the intercomparison measurements have been restricted to the analysis of the NBS plutonium isotopic standard reference materials NBS-SRM 946, 947 and 948. Those certified reference materials were available at several laboratories. As to the isotopic grade, the measured samples can be classified as aged (in terms of time elapsed since ^{241}Am separation) materials of low and medium burnup. They do not include high-burnup material.

Another restriction imposed on the intercomparison experiment refers to the gamma-ray signatures used for obtaining the isotopic ratios. The laboratories have been requested to concentrate their analysis to the limited energy region from 125 keV to 208 keV. Apart from the complex X-ray region between 94 keV and 104 keV, this energy region provides the most abundant gamma rays from the plutonium isotopes 238, 239, 240 and 241 which are detectable in gamma spectra from aged plutonium materials. Actually, most of the laboratories have employed this energy region in their previous plutonium isotopic composition measurements. Fig. 1.1 displays for illustration gamma spectra from plutonium materials of distinctly different isotopic grade in the energy region of interest.

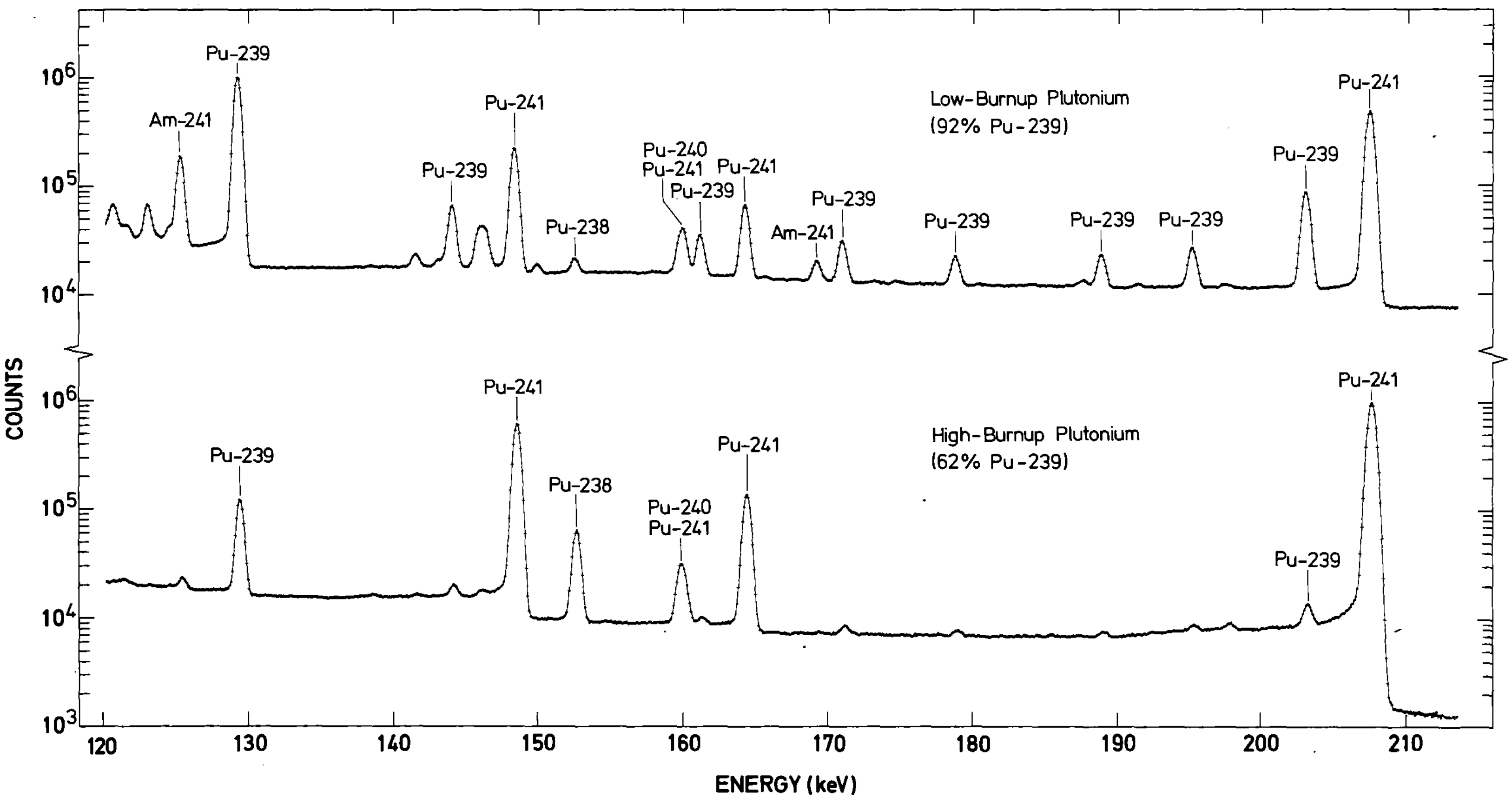

Fig. 1.1 Gamma spectra from plutonium materials of distinctly different isotopic grade ('low burnup' and 'high burnup') in the energy range between 120 and 210 keV.

The isotopic analysis in the selected energy range assumes equilibrium between ^{241}Pu and ^{237}U ($t_{1/2}$ = 6.75 d). Given this equilibrium - a condition fulfilled by the NBS reference materials - it is convenient to determine the plutonium isotopic ratios relative to ^{241}Pu. Therefore the participants have been asked to report the following data:

i) The plutonium isotopic ratios $^{238}Pu/^{241}Pu$, $^{239}Pu/^{241}Pu$ and ^{240}Pu / ^{241}Pu,

ii) the ratio $^{241}Am/^{239}Pu$ which is also easily obtained from the gamma measurement,

iii) raw data such as measured peak areas and relative detection efficiencies,

iv) experimental details.

Those data had to be obtained from the laboratories' own measurements from the 3 NBS reference materials, if available, and / or from the analysis of reference spectra distributed to the participants.

In the following section more experimental details about the experiment are given. Section III and the Appendices will present the reported data and their evaluation. Section IV will give a discussion of the obtained results for the different isotopic ratios. In Section V some gamma branching intensity ratios will be reevaluated in view of the results from the present exercise. Preliminary results have been reported previously in two conference contributions /1,2/.

II. EXPERIMENTAL DETAILS

A. The Common Set of Nuclear Data

The proposed common set of nuclear data to be used by the participants in the evaluation of the isotopic ratios comprised the absolute gamma branching intensities reported by R. Gunnink /3/, and the half lives recommended in ANSI 15.22 /4/. The source for the gamma branching intensities represents the most comprehensive and most accurate set of data which is currently available. Yet, recent experiences with gamma-spectrometric plutonium isotopic composition measurements have indicated that some of those data need some further refinements in order to improve the potential accuracy of the method.

The isotope half lives recommended in ANSI 15.22 are listed in Table 2.1. The half-life values have been used both for converting the measured peak area ratios into atom ratios and for updating the isotopic composition of the NBS reference materials.

In the meantime some of these half-life values have slightly changed due to remeasurements or reevaluations. Those changes are not relevant for the comparison of isotopic ratios reported from the present exercise, but they have been taken into account in the reevaluation of some branching intensity ratios as discussed below in Section V.

Table 2.1. Isotope Half Lives Recommended in ANSI 15.22

Isotope	Recommended Half Life (years)
^{238}Pu	87.79 ± 0.08
^{239}Pu	24082 ± 46
^{240}Pu	6537 ± 10
^{241}Pu	14.35 ± 0.02
^{241}Am	434.1 ± 0.6

B. Reference Spectra

The reference spectra distributed to the participants for intercomparison analysis have been measured previously at KfK under the experimental conditions given in Table 2.2. Two spectra have been taken from each of the 3 NBS isotopic reference materials using two different detector systems:

- A single open-ended coaxial Ge(Li)-detector, diameter 32 mm, length 25 mm, active volume 18 cm³. The spectra taken with this detector are hence referred to as 'COAX' spectra.

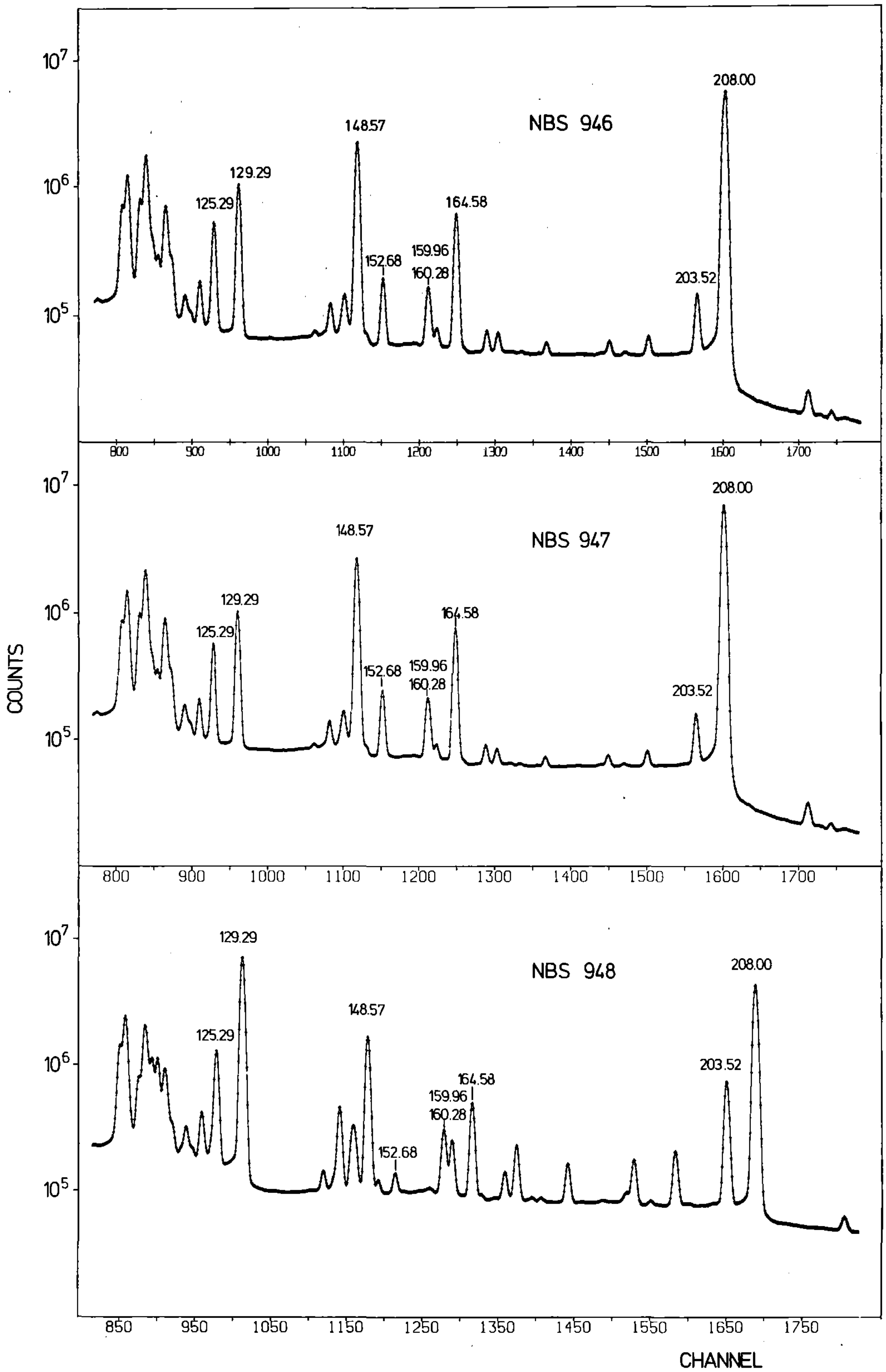

Fig. 2.1 Reference spectra from NBS-SRM 946, 947, 948 measured with the 'COAX' detector.

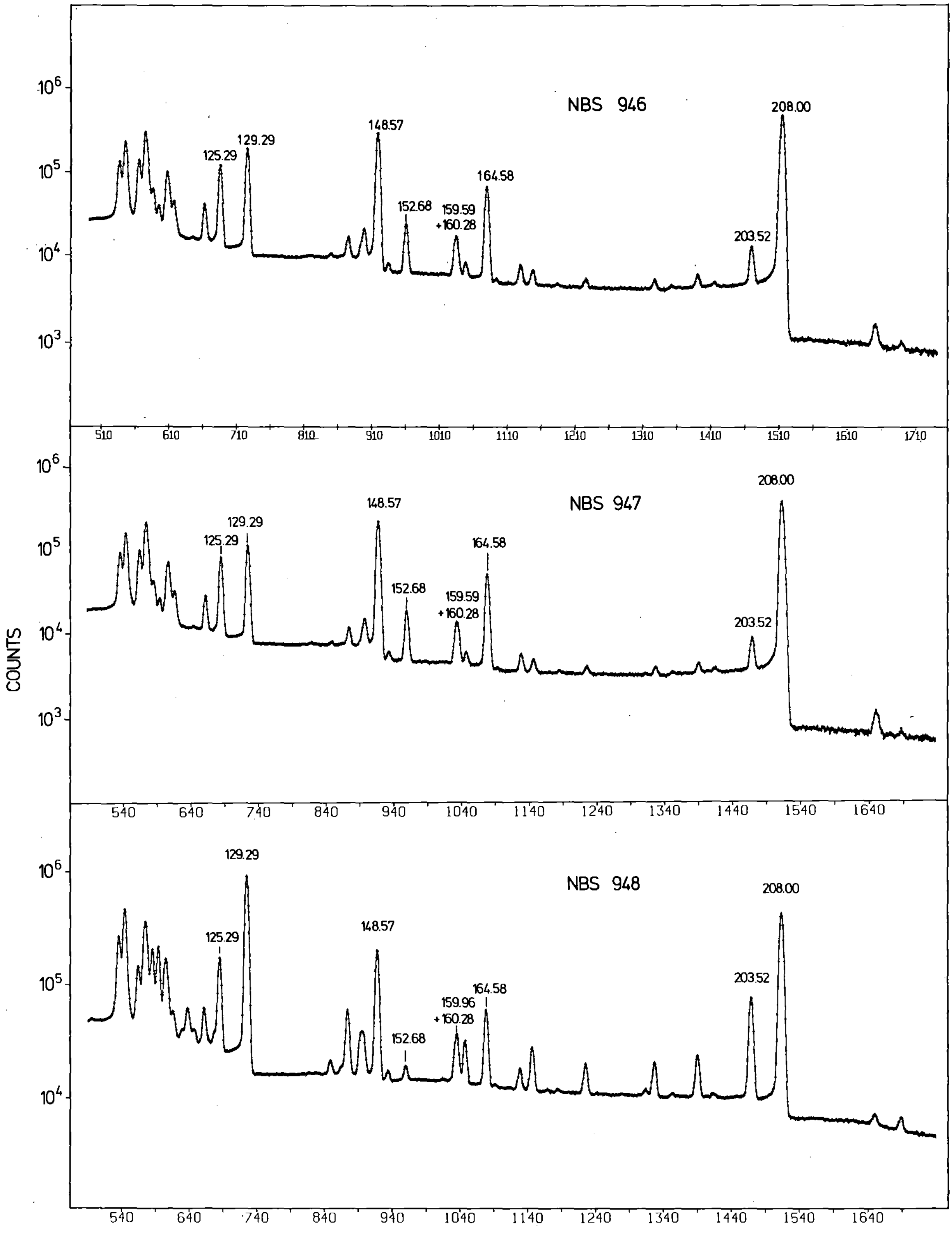

Fig. 2.2 Reference spectra from NBS-SRM 946, 947, 948 measured with the 'LEPS' detector.

- A planar intrinsic Ge detector, diameter 16 mm, depletion depth 7 mm, active volume 1.4 cm^3. The spectra taken with this detector are hence referred to as 'LEPS' (Low Energy Photon Spectrometer) spectra.

Fig. 2.1 shows the relevant portion of the reference spectra taken with the 'COAX' detector, and Fig. 2.2 the corresponding spectra taken with the 'LEPS' detector. The spectra have been distributed either on punched cards, paper tape or magnetic tape depending upon the request of the participant.

As can be seen from Table 2.2, relatively long counting times have been chosen for the accumulation of the reference spectra from the small quantities of reference materials (0.1 - 0.25 g Pu). It has been the intention to abstract in this way as far as possible from random errors due to counting statistics, thereby making the systematic errors in the analysis more evident. Table 2.3 summarizes the calculated relative standard deviation

$$\Delta A/A = \sqrt{(1 + 2B/A)/A}$$

of the net peak area, A, above the background area, B, for the relevant gamma rays in the 6 reference spectra. With the exception of the ^{238}Pu gamma line at 152.68 keV (LEPS spectrum from NBS 948), the ^{239}Pu line at 203.52 keV (LEPS spectra from NBS 946, 947) and the ^{240}Pu line at 160.28 keV, the counting errors have been kept well below 1 %, in some cases even below 0.1 %.

The isotopic composition of the 3 plutonium isotopic reference materials NBS-SRM 946, 947, 948 is given for reference dates dating back to 1971 and 1972. Remeasurements of the isotopic composition of NBS-SRM 946 and 947 by mass spectrometry and alpha spectrometry have been requested from the Central Bureau for Nuclear Measurements (CBNM), Geel (Belgium), particularly in view of a potential improvement of the accuracy for the ^{238}Pu abundance. The available isotopic compositions from the National Bureau of Standards (NBS), Washington, and from CBNM, Geel, are summarized in Table 2.4.

Table 2.2 Measurement Conditions for the Accumulation of Reference Spectra Distributed for Intercomparison Analysis

Parameter	Detector	Measurement of NBS 946	NBS 947	NBS 948
Source-to-Detector Distance (cm)	LEPS	4	4	3
	COAX	Adjusted to give 5000 cps		
Total Detector Countrate(cps)	LEPS	750	900	650
	COAX	5000	5000	5000
Slope (eV/channel)	LEPS	49.88	49.88	49.88
	COAX	123.15	123.15	116.01
Resolution (eV) Attained at 129 keV:	LEPS	549	549	549
	COAX	713	718	743
at 414 keV:	LEPS	878	868	893
	COAX	1093	1081	1044
Counting Time (Minutes)	LEPS	4300	3000	6000
	COAX	1400	1400	3400
Absorber		1 mm Cd for all measurements		
Main Amplifier Shaping Time		2 μsec for all measurements		
Digital Stabilizer		All measurements digitally stabilized using internal 59.6 keV peak for ZERO and 208.0 peak for GAIN stabilization.		
Pile-up Rejection		None used		

Table 2.3 Counting Precisions of Gamma Rays in the Reference Spectra Distributed for Intercomparison Analysis

Gamma Line (keV)	Relative Standard Deviation of Net Peak Area(%)					
	NBS 946		NBS 947		NBS 948	
	LEPS	COAX	LEPS	COAX	LEPS	COAX
125.29 + 125.21	0.18	0.09	0.23	0.09	0.18	0.05
129.29	0.12	0.05	0.13	0.05	0.05	0.02
148.57	0.08	0.03	0.09	0.03	0.11	0.04
152.68	0.53	0.39	0.60	0.18	2.38	0.72
159.96-160.28	0.66	0.21	0.68	0.18	0.51	0.14
160.28[a]	1.82	0.94	1.56	0.57	0.71	0.25
164.58	0.20	0.06	0.23	0.06	0.19	0.21
203.52	0.91	0.26	1.29	0.30	0.21	0.06
208.00	0.06	0.02	0.07	0.02	0.06	0.02

[a] After removal of ^{241}Pu interference relative to adjacent ^{241}Pu line at 164.58 keV, and of ^{239}Pu interference.

The isotopic ratios ^{238}Pu/^{241}Pu, ^{239}Pu/^{241}Pu and ^{240}Pu/^{241}Pu updated to the data of the gamma measurements from the NBS and CBNM reference analyses with the half lives from Table 2.1 are summarized in Table 2.5.

The updated ratios ^{239}Pu/^{241}Pu and ^{240}Pu/^{241}Pu for SRM 946 and 947 agree to better than 0.15 %, while the CBNM values for the ratio ^{238}Pu/^{241}Pu in SRM 946 and 947 are 0.7 % higher than the corresponding NBS values.

C. Laboratories' Own Measurements

There have been 7 participants among the 9 participating laboratories which had the isotopic reference materials SRM-946, 947, 948 available for own gamma measurements. Table 2.6 gives some experimental details on the measurement conditions for those measurements. Three laboratories

Table 2.4 Isotopic Composition of NBS-SRM 946, 947, 948

Sample	Analysis	Date	Atom Percent				
			^{238}Pu	^{239}Pu	^{240}Pu	^{241}Pu	^{242}Pu
SRM-946	NBS	09/10/71	0.247 $\pm$0.007	83.128 $\pm$0.015	12.069 $\pm$0.015	3.991 $\pm$0.005	0.565 $\pm$0.003
	CBNM	08/09/78	0.239 $\pm$0.002	84.087 $\pm$0.030	12.203 $\pm$0.020	2.899 $\pm$0.005	0.572 $\pm$0.003
SRM-947	NBS	13/10/71	0.296 $\pm$0.006	75.696 $\pm$0.022	18.288 $\pm$0.022	4.540 $\pm$0.006	1.180 $\pm$0.004
	CBNM	12/09/78	0.286 $\pm$0.002	76.703 $\pm$0.047	18.520 $\pm$0.036	3.296 $\pm$0.006	1.195 $\pm$0.003
SRM-948	NBS	01/09/72	0.011 $\pm$0.001	91.574 $\pm$0.010	7.914 $\pm$0.010	0.468 $\pm$0.001	0.0330 $\pm$0.0003

Table 2.5 Updated Isotopic Ratios at the Date of Gamma Measurement

Spectrum	Date of Measurement	Isotopic Ratios					
		$^{238}Pu/^{241}Pu$		$^{239}Pu/^{241}Pu$		$^{240}Pu/^{241}Pu$	
		NBS	CBNM	NBS	CBNM	NBS	CBNM
946 COAX	27/02/76	0.07397	0.07444	25.713	25.676	3.7317	3.7269
946 LEPS	12/01/78	0.07973	0.08029	28.145	28.103	4.0844	4.0786
947 COAX	03/03/76	0.07784	0.07834	20.605	20.597	4.9765	4.9742
947 LEPS	04/01/78	0.08370	0.08439	22.528	22.512	5.4402	5.4358
948 COAX	15/04/78	0.02969		256.804		22.185	
948 LEPS	30/04/78	0.02977		257.528		22.247	

Table 2.6 Measurement Conditions of Laboratories' Own Measurements

Laboratory	Measurement No.	Detector Type	FWHM at 129 keV (eV)	Amplifier Shaping Time (μsec)	Gain (eV/channel)	Detector Countrate (cps x 10^3)	Source Distance (cm)	Counting Time (Hours)
1	1	Ge(Li), Coaxial 39 mm Ø x 13 mm	680	4	275	2.5 - 3	1 - 3	6.7
	2	Ge(Li), Coaxial 36 mm Ø x 30 mm	780	2	210	3	1 - 3	7
2		HPGe, Planar	570	6	110	2	NA	15-23
3	1	HPGe, Planar 16 mm Ø x 7 mm	550	2	50	0.7 - 0.9	3 - 4	50 - 100
	2	Ge(Li), Coaxial 32 mm Ø x 25 mm	720	2	120	5	NA	23 - 57
4		HPGe, Planar	560	NA[a]	NA	NA	5	3 x 2.7 1 x 14
5	1	HPGe, Planar 32 mm Ø x 11 mm	800	6	120-200	NA	10 - 40	5 - 8
	2	HPGe, Planar 32 mm Ø x 11 mm	850-1080	6	200	NA	20 - 30	2 - 11
6		HPGe, Planar	507	6	100	0.7 - 1.1	0.8 - 2	70 - 94
8		HPGe, Planar	680	1	110	3	10 - 20	1.5

[a] NA = Not available

(laboratories coded 1, 3 and 5) have performed 2 measurements on each sample using different detector systems or measurement conditions. In most cases planar detectors of intrinsic germanium with an energy resolution of about 500-800 eV at 129 keV have been used. The values given for the energy resolution are those obtained in the actual measurements.

The plutonium quantity for each of the reference materials is nominally 0.25 g. In some cases only a fraction of this quantity has been available for the gamma measurements. Again, relatively long counting times have been used for spectrum accumulation, ranging from a few hours up to about 100 hours of measurement time per spectrum. Only laboratory 8 has used a relatively short counting time of 1.5 hours/ spectrum.

The selected source-to-detector distances cover a wide range from about 1 cm up to 40 cm. All participants used cadmium absorbers with a thickness ranging between 1 mm and 2.4 mm, supplemented in some cases by a copper absorber of about 0.25 to 1 mm thickness, in order to reduce the intensity of the intense ^{241}Am radiation at 59.6 keV. The total detector countrate has been kept below 5000 cps in all the measurements.

D. Methods of Spectrum Analysis

In principle, an intercomparison exercise on plutonium isotopic ratio measurements is a test of various methods for peak area determination applied to the specific case of decomposing the complex gamma spectra from plutonium materials, supplemented by a comparison of methods employed for determining relative detection efficiencies from the measured gamma spectrum. A variety of different methods for spectrum analysis has been used by the laboratories participating in the present exercise. Table 2.7 gives a survey on the various methods of spectrum analysis employed by the participants. Four laboratories (laboratories coded 1, 3, 5 and 7) have reported results from more than one approach. In these cases a second digit has been added to the laboratory code number (e.g., 1/1, 1/2) in order to distinguish between the different analyses from one participant.

Table 2.7 Methods of Spectrum Analysis

Laboratory Code ℓ	Method of Peak Area Analysis	Background Approximation	Functional Relationship for Relative Detection Efficiency $\varepsilon(E)$
1/1 1/2	SYM.GAUSSIAN ASYM.GAUSSIAN	Linear Dispersed Step	$\ln\varepsilon = \sum_{n=0}^{3} a_n (\ln E)^n$
2	CHAN.SUMMATION (3 x FWHM)	Dispersed Step	ln = a + b lnE in 140-170 keV Region
3/1 3/2 3/3	CHAN.SUMMATION ASYM.GAUSSIAN ASYM.GAUSSIAN	Dispersed Step Linear Dispersed Step	$\varepsilon = \sum_{n=0}^{3} a_n E^n$ in 125-170 keV Region
4	ASYM.GAUSSIAN	Dispersed Step	$\ln\varepsilon = \sum_{n=0}^{3} a_n (\ln E)^n + b_o/E + b_1/E^2$
5/1 5/2	SYM.GAUSSIAN ASYM.GAUSSIAN	Linear Linear	$\ln\varepsilon = \sum_{n=0}^{3 \text{ or } 4} a_n (\ln E)^n$
6	SYM.GAUSSIAN	Dispersed Step	$\ln\varepsilon = a + b \ln E$ or $\varepsilon = aE^{-b}$ in 140-170 keV Region
7/1 7/2	CHAN.SUMMATION ASYM.GAUSSIAN	Linear Linear	$\log\varepsilon = \sum_{n=0}^{3 \text{ or } 4} a_n (\log E)^n$
8	SYM.GAUSSIAN	Dispersed Step	$\varepsilon = \left[C_1 \exp(A_1/E) + C_2 \exp(A_2/E)\right]^{-1}$
9	SYM.GAUSSIAN (2.2 x FWHM)	Dispersed Step	$\ln\varepsilon = \sum_{n=0}^{3} a_n (\ln E)^n$

For the evaluation of the net peak areas of the gamma rays of interest both the channel summation method (CHANSUM) and various peak fitting routines have been employed. In the latter case either symmetrical Gaussian functions (SYM.GAUSSIAN) or Gaussian functions modified by a tailing function on the low-energy side of the gamma peaks (ASYM. GAUSSIAN) were used to describe the spectral peak shape. The background underneath the peaks has been approximated by linear or dispersed step functions.

The approaches for the determination of the relative detection efficiency can be classified as following:

i) Determination of a complete relative detection efficiency curve in the energy range from 125 keV up to about 450 keV using ^{239}Pu gamma lines alone (laboratories 5 and 7) or using gamma lines from ^{239}Pu and ^{241}Pu - ^{237}U (laboratories 1 and 9). Polynomials in a log-log representation have been the preferred functional relationship for describing the energy dependence of the relative detection efficiency ε.

ii) Determination of a complete relative detection efficiency curve in the energy range from 125 keV to 208 keV (laboratories 4 and 8) using the functional relationship given in Table 2.7.

iii) Determination of segments of the relative detection efficiency curve in energy regions of interest (laboratories 2,3 and 6). These laboratories started with the evaluation of the ratios $^{239}Pu/^{241}Pu$ and $^{241}Am/^{239}Pu$ from the 332-345 keV region, assuming a power law function for the relative detection efficiency curve in the 332-413 keV region which has been fitted to gamma-ray intensities of ^{239}Pu gamma lines. The isotopic ratios obtained from this energy region have then been used to establish segments of the relative efficiency curve in the 125-208 keV region for the determination of the remaining isotopic ratios.

III. REPORTED DATA AND THEIR EVALUATION

A. Isotopic Ratios

Most of the laboratories have reported the isotopic ratios as requested, viz., the $^{238}Pu/^{239}Pu$ ratio evaluated from the peak pair 152/148 keV, the $^{239}Pu/^{241}Pu$ ratio evaluated from the peak pairs 129/148 keV and 203/208 keV, the $^{240}Pu/^{241}Pu$ ratio evaluated from the peak pairs 160/148 keV and 160/164 keV, and the ratio $^{241}Am/^{239}Pu$ determined from the peak pair 125/129 keV. A few participants have partly reported other isotopic ratios evaluated from different gamma rays. In cases where the complete raw data such as net peak areas and relative detection efficiencies have been given, the author has evaluated the above isotopic ratios from the available raw data. Some participants have reported additional data such as $^{239}Pu/^{241}Pu$ and $^{241}Am/^{239}Pu$ ratios determined from higher energy gamma rays in the energy region between 332 and 422 keV. Those data, although included in the Tables in Appendix B and C, have not been used for comparison purposes.

For the reason of convenient comparison the measured plutonium isotopic ratios 238/241, 239/241 and 240/241 have been divided by the isotopic ratio calculated from the updated NBS reference values. The measured ratios $^{241}Am/^{239}Pu$, for which no reference values are available from the NBS certificates, have been normalized to a reference date of January 1st, 1978 in order to obtain a comparable set of data.

The normalized isotopic ratios which have been determined by the participants from their own measurements and/or from the distributed 'LEPS' and 'COAX' spectra are graphically presented in Appendix A. The corresponding numerical values are compiled in Appendix B and C.

There have been no special statistical test procedures applied to the reported isotopic ratios such as tests for the homogeneity of the data, outlier tests etc. for the following reasons:

i) For most of the reported isotopic ratios the statistical error component constitutes the minor part of the total assay error.

This holds at least for the results obtained from the analysis of the distributed spectra, which have been collected with good counting precisions (see Tables 2.3, D.1 and D.3). It is then obvious that the observed scatter of the results will be mostly governed by systematic errors associated with the different procedures for spectrum analysis. It has been anticipated that the results from the different methods of spectrum analysis will vary to some extent. Some participants have even underscored the influence of the selected method of spectrum analysis upon the evaluated isotopic ratios by providing different sets of results, which have been obtained using either different methods for peak area determination or different functional approximations for the relative detection efficiency.

ii) There has been no uniform way among the participants for estimating the overall assay error. Most of the participants only included the statistical error components from their analysis into their estimate of the assay errors. Only a few participants also accounted for some systematic errors in their error analysis. It appeared therefore difficult to weight the different results appropriately. The error bars in Figs. A.1 to A.6 and the bracket values given in the Tables in Appendix B and C correspond to the reported 1σ errors.

For the above reasons only the following evaluations have been made on the reported isotopic ratios $R_{\ell is}$,
with

ℓ = laboratory code (ℓ_{max} = 14)

i = isotopic ratio (i_{max} = 6 for 238/241 from 152/148 keV,
239/241 from 129/148 keV and 203/208 keV,
240/241 from 160/148 keV and 160/164 keV,
Am/239 from 125/129 keV)

s = sample/spectrum (s_{max} = 9 for NBS 946 LEPS, COAX, Own,
NBS 947 LEPS, COAX, Own, NBS 948 LEPS, COAX,
Own):

1. Calculation of the unweighted grand mean value

$$\bar{R}_{is} = \frac{1}{w} \sum_{\ell=1}^{n} R_{\ell is}$$

from the reported n evaluations of a given isotopic ratio i from a given spectrum s.

2. Calculation of the relative standard deviation

$$RSD(R_{\ell is}) = \frac{1}{\bar{R}_{is}} \left(\frac{\sum_{\ell=1}^{n} (R_{\ell is} - \bar{R}_{is})^2}{(n-1)} \right)^{1/2}$$

as some kind of quantitative figure for the spread of the data. The grand mean values with the associated $\pm$ 1σ limits are indicated as horizontal lines in Figs. A.1 to A.6 in Appendix A. The numerical values are given in the Tables in Appendix B and C.

3. For each evaluation ℓ of a given isotopic ratio i, the following average relative deviations

$$\overline{\Delta R_{\ell i}} = \frac{1}{m} \sum_{s=1}^{m} \left(\frac{R_{\ell is} - \bar{R}_{is}}{\bar{R}_{is}} \right) \qquad \text{and}$$

$$|\overline{\Delta R_{\ell i}}| = \frac{1}{m} \sum_{s=1}^{m} \left| \frac{R_{\ell is} - \bar{R}_{is}}{\bar{R}_{is}} \right|$$

from the grand mean values have been calculated for the number of spectra analyzed by a participant's evaluation method ℓ (typically m = 6 or 9 for NBS 946, 947, 948 Own and/or LEPS and COAX). The approximate equality of the values $\overline{\Delta R_{\ell i}}$ and $|\overline{\Delta R_{\ell i}}|$ will be an indication if there exists a bias for a given isotopic ratio from a given evaluation. Results from this test, if positive, will be discussed in Chapter IV along with the discussion of the individual isotopic ratios.

B. Peak Areas

Together with the evaluated isotopic ratios, the participants have also reported raw data such as net peak areas and relative detection efficiencies. The raw data from the analysis of the distributed 'LEPS' and 'COAX' spectra provide a good set of comparable data, which can be used to better identify the different error components contributing to the observed scatter of the reported isotopic ratios. For this reason the raw data from the evaluation of those spectra have been also included in this report.

The net peak areas of 8 single peaks or peak complexes in the energy range between 125 and 208 keV, which are of relevance for the isotopic ratio evaluation, are compiled in Appendix D for both the 'LEPS' and 'COAX' spectra. Total peak areas of the unresolved peak complexes at 125 and 160 keV are given because not all of the participants have reported the percentage contributions of the individual gamma rays to the respective peak complexes. Again, unweighted grand mean values and associated relative standard deviations have been calculated from the different sets of data as given in Tables D.1 to D.4. In addition, reduced mean values have been calculated after the exclusion of deviating results. The reduced mean values have been used for the reevaluation of some branching intensity ratios as discussed in Chapter V. For relatively uniform data sets, results deviating more than 3 % from the grand mean have been excluded from the reduced mean. For data sets with a larger scatter such as the 152 keV peak areas from NBS 948, results deviating more than 5 % from the grand mean have been omitted from the reduced mean.

It is clear that the different approaches for the peak area analysis, in particular the different approximations for the background underneath the peaks (linear, step) result in different peak areas which will vary to a larger extent than expected from counting precisions. The different biases, however, should not affect the isotopic ratio evaluation, provided the peak areas obtained from a particular peak fitting routine show the same systematic trend for all gamma lines. Consequently, the scatter of the values for peak area ratios as reported in Tables D.2 and D.4 should become narrower. This is indeed observed in most cases. However, the variation of the peak area ratio values is still considerably higher than the scatter expected from the counting precision values as given in Table 2.3.

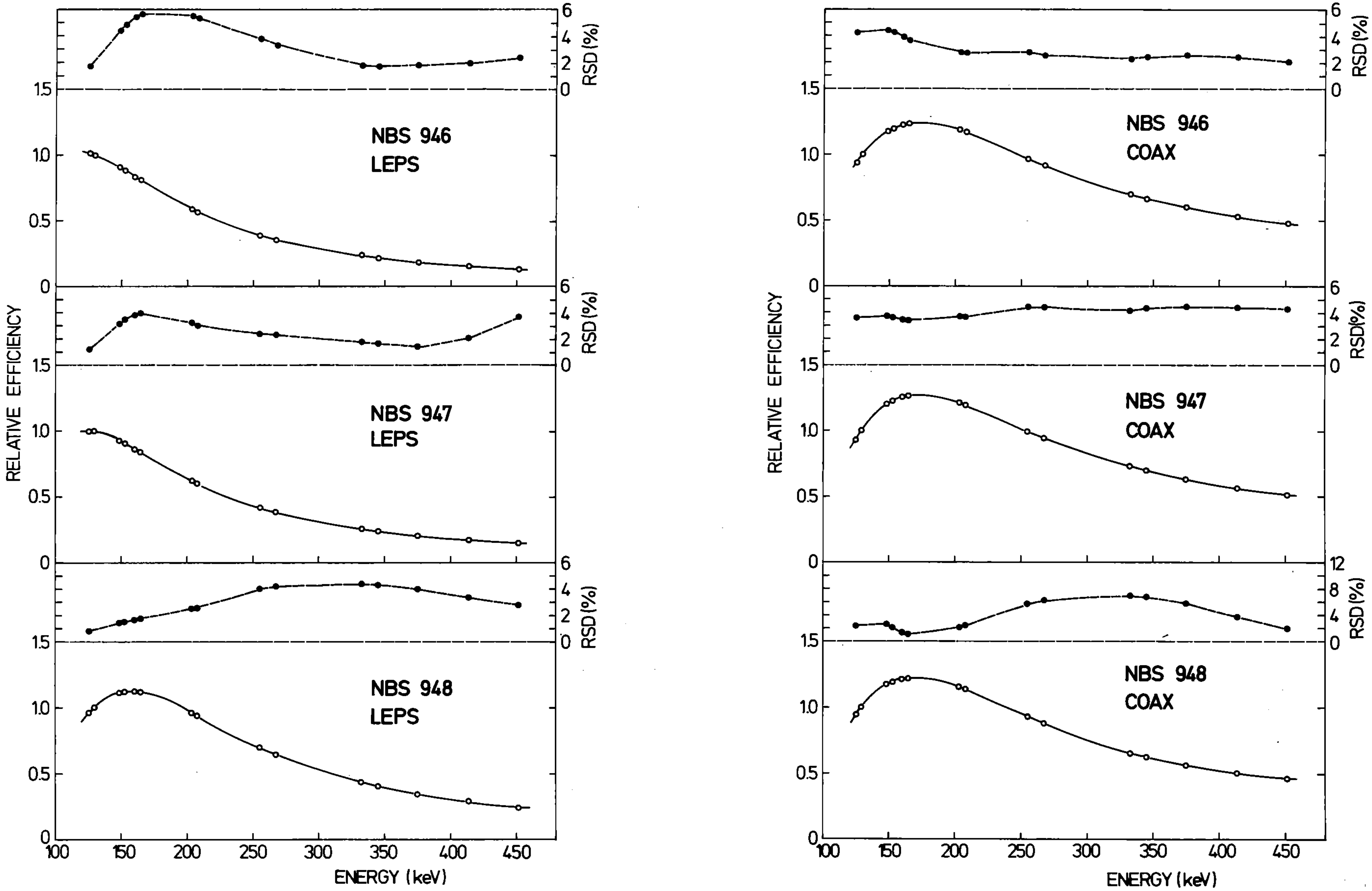

Fig. 3.1 Normalized detection efficiency and associated relative standard deviation for gamma energies between 125 and 451 keV determined from the distributed 'LEPS' and 'COAX' spectra. Values plotted are the mean values with standard deviation as given in Tables E.1 to E.6 in Appendix E.

C. Relative Detection Efficiency

Relative detection efficiencies have been reported either explicitely or as functional relationships (see Table 2.7) with numerical values for the coefficients in the functions. In the latter case the efficiency values have been calculated from the given functions. Again, only the efficiency values determined from the distributed 'LEPS' and 'COAX' spectra will provide directly comparable data. The available relative detection efficiency values from those spectra, normalized to a value of 1.00 at 129.29 keV, are compiled in Appendix E (Tables E.1 to E.6). Laboratories 2 and 6 have only reported efficiency ratios for relevant pairs of gamma energies. Those data are included in Tables E.7 and E.8, which summarize efficiency ratios for some important pairs of gamma energies together with calculated grand mean and reduced mean values. The reduced mean values for the efficiency ratios 148/152, 148/160, 164/160 and 208/203 keV have been obtained after the exclusion of results deviating more than 2 % from the grand mean, if present. For the calculation of the reduced mean of the efficiency ratios 129/125 and 148/129 keV, results deviating more than 5 % from the grand mean have been excluded.

The normalized efficiency values and associated relative standard deviations, compiled in Tables E.1 to E.6 for a number of gamma-ray energies between 125 and 451 keV, are plotted in Fig. 3.1. The gross behaviour of the relative detection efficiency curves in the given energy range is typical for small sample measurements with little gamma self attenuation. It should be noted that the given efficiency curves are probably more difficult to approximate by suitable functions than the efficiency curves from bulk sample measurements, which usually show a smooth and steady increase with energy in the given energy range.

IV. DISCUSSION OF RESULTS

A. Pu-238/Pu-241 Ratios

The evaluations of this isotopic ratio from the peak pair 152/148 keV provided relatively consistent results, with a comparatively small scatter of the ratios determined from NBS 946 and 947. There are two favourable conditions for a reliable determination of this ratio from gamma

Table 4.1 Average $^{238}Pu/^{241}Pu$ Ratios from Peak Pair 152/148 keV and Error Components from Distributed Spectra

Sample/ Spectrum	238/241(γ/NBS)		RSD(%) of A_{152}/A_{148}			RSD(%) of $\varepsilon_{148}/\varepsilon_{152}$	
	Grand Mean	RSD (%)	Grand Mean	Reduced Mean	Counting Statistics	Grand Mean	Reduced Mean
946 LEPS	0.979	0.86	1.91	0.60	0.54	0.51	-
946 COAX	0.982	2.13	2.32	1.08	0.39	0.52	-
946 Own	0.976	1.90	-	-	-	-	-
947 LEPS	0.978	0.83	1.61	0.65	0.61	0.32	-
947 COAX	0.976	1.42	1.47	1.14	0.18	0.49	-
947 Own	0.983	1.85	-	-	-	-	-
948 LEPS	1.063	2.57	3.39	2.76	2.38	0.24	-
948 COAX	1.044	3.63	5.65	2.34	0.72	0.42	-
948 Own	1.078	6.89	-	-	-	-	-

spectrometry measurements: i) the two isotopic gamma rays involved in the analysis are well resolved and free from interferences, and ii) the two gamma rays are close to each other, thereby requiring relatively small corrections for detection efficiency.

Table 4.1 summarizes the average 238/241 ratios - divided by the NBS reference values - which have been determined from the different measurements. The Table also contains informations about the variances of the peak area ratios A_{152}/A_{148} and of the detection efficiency ratios $\varepsilon_{148}/\varepsilon_{152}$ determined from the distributed 'LEPS' and 'COAX' spectra. The variances are identical to the RSD-values given in Tables D.2 and D.4 for the grand and reduced means of the peak area ratios, and in Tables E.7 and E.8 for the grand and reduced means for the efficiency ratios, respectively. In addition, the precision values for the peak area ratios A_{152}/A_{148} as expected from the counting statistics are also given in Table 4.1 for comparison purposes.

The 238/241 ratios determined from NBS 946 and 947 are, on an average, 2.1 % lower than the NBS reference values. This bias increases to -2.7 % relative to the improved 238/241 reference values determined at CBNM, Geel. We attribute this deviation to a bias in the branching intensity ratio of the 152 and 148 keV gamma rays, which will be reevaluated in Chapter V on the basis of the present results.

The standard deviations of the peak area ratios A_{152}/A_{148} determined from the 'LEPS' spectra from NBS 946, 947 (reduced data basis after exclusion of outliers) are close to the counting precision values. This indicates that the 152/148 keV peak area ratio can be reliably determined from high-resolution gamma spectra. For comparison, the standard deviations of the peak area ratios determined from the lower resolution 'COAX' spectra are about 3 to 5 times larger than the counting precision values for the ratio. The variations of the efficiency ratio $\varepsilon_{148}/\varepsilon_{152}$ are in all cases smaller than the variations of the peak area ratios.

The ratios 238/241 determined from NBS 948 are, on an average, about 6 % higher than the NBS reference value, which has an uncertainty of 9 %. The errors of the gamma measurements on NBS 948 with its very low ^{238}Pu abundance (0.011 %) are almost exclusively governed by the poor counting statistics obtained for the 152 keV line from ^{238}Pu. Calculating a weighted average from the 3 values for the 238/241 ratio given in Table 4.1, and taking into account the adjusted branching intensity ratio for the 152 and 148 keV gamma rays, the ^{238}Pu abundance in NBS 948 is determined from the present measurements to

^{238}Pu (Atom % at NBS Reference Date of 01/09/72) = 0.01196 ± 0.00030.

The corresponding NBS reference value is 0.011 ± 0.001.

An examination of the individual results for NBS 946 and 947 - the results from NBS 948 are not considered because of the relatively large counting errors - indicates an average bias from the grand means of -2.0 % for laboratory/analysis 3/1, and an average bias of -1.0 % for laboratory 4. The bias for participant 3/1 is associated with a bias for the peak area of the 148 keV line. In this analysis (channel summation method) counts have been integrated over the energy range from 143 to 150 keV and corrected for interferences. This procedure resulted in an overestimate of the 148 keV peak area. Consequently, any isotopic ratios determined from this analysis involving the 148 keV line will show some bias.

B. Pu-239/Pu-241 Ratios

1. *Ratio from peak pair 129/148 keV.* The results from this analysis are summarized in Table 4.2. The average 239/241 ratios are close to the mass-spectrometric reference values, but have relatively large uncertainties. As can be seen from the Table, these uncertainties are mostly related to the determination of the efficiency ratio $\varepsilon_{148}/\varepsilon_{129}$. As a matter of fact, the isotopic ratio determined from this peak pair does in most cases not represent a true independent measurement, because the 239/241 ratio determined from other spectral regions has been used as input data for establishing the relative detection efficiency in this energy region. Therefore, the variation of the results for the isotopic ratio 239/241 deduced from the peak pair 129/148 keV more or less reflects the goodness of the efficiency approximations in this energy region. Indeed, the observed average biases for individual results from the grand means, -4.4 % and +4.6 % for the analyses 1/1 and 1/2 from laboratory 1, and + 2.1 %, +3.1 % and +3.6 % for the analyses 3/1, 3/2 and 3/3 of laboratory 3, respectively, are mostly related to deviating efficiency ratios. The efficiency corrections improve for the low-burnup material NBS 948 where additional gamma rays from ^{239}Pu can be employed for establishing the relative detection efficiency in this energy region. We conclude that the peak pair 129/148 keV will generally not provide a reliable independent $^{239}Pu/^{241}Pu$ ratio determination from samples of arbitrary geometry with unknown gamma attenuation characteristics.

2. *Ratio from peak pair 203/208 keV.* The results from this analysis are given in Table 4.3.The measured 239/241 ratios are again close to the reference values, suggesting that the nuclear data used for the analysis are correct within 1 %. The new half life for ^{239}Pu recommended by the U.S. half-life evaluation committee (24119 $\pm$ 26 years as compared to the ANSI 15.22 value of 24082 years used in the present analyses) will increase the reported ratios by 0.15 %. For NBS 946 and 947, most of the scatter of the measured 239/241 ratios is associated with the determination of the peak area ratio A_{203}/A_{208}. For NBS 948, the error contributions from the peak area ratio and from the efficiency correction are comparable. For the range of isotopic distributions represented by the NBS reference

Table 4.2 Average $^{239}Pu/^{241}Pu$ Ratios from Peak Pair 129/148 keV and Error Components from Distributed Spectra

Sample/ Spectrum	239/241(γ/NBS)		RSD(%) of A_{129}/A_{148}			RSD(%) of $\varepsilon_{148}/\varepsilon_{129}$	
	Grand Mean	RSD (%)	Grand Mean	Reduced Mean	Counting Statistics	Grand Mean	Reduced Mean
946 LEPS	0.972	4.57	1.56	0.39	0.14	3.94	3.25
946 COAX	0.989	3.58	1.07	-	0.06	3.99	2.86
946 Own	1.028	5.71	-	-	-	-	-
947 LEPS	0.986	2.99	1.46	0.52	0.16	3.13	2.81
947 COAX	0.990	3.18	1.21	-	0.06	3.36	2.86
947 Own	1.009	5.22	-	-	-	-	-
948 LEPS	0.992	2.27	0.89	-	0.12	1.36	-
948 COAX	0.998	2.57	1.49	1.04	0.04	2.22	-
948 Own	0.999	3.07	-	-	-	-	-

Table 4.3 Average $^{239}Pu/^{241}Pu$ Ratios from Peak Pair 203/208 keV and Error Components from Distributed Spectra

Sample/ Spectrum	239/241(γ/NBS)		RSD(%) of A_{203}/A_{208}			RSD(%) of $\varepsilon_{208}/\varepsilon_{203}$	
	Grand Mean	RSD (%)	Grand Mean	Reduced Mean	Counting Statistics	Grand Mean	Reduced Mean
946 LEPS	0.986	1.59	1.48	-	0.91	0.39	-
946 COAX	0.994	1.82	1.85	-	0.26	0.51	-
946 Own	1.001	2.38	-	-	-	-	-
947 LEPS	0.980	1.60	3.32	1.22	1.29	1.82	0.68
947 COAX	0.982	2.31	2.29	1.65	0.30	0.45	-
947 Own	0.999	-	-	-	-	-	-
948 LEPS	0.998	1.03	0.46	-	0.22	0.48	-
948 COAX	0.993	2.18	0.71	0.37	0.06	0.73	-
948 Own	0.995	1.14	-	-	-	-	-

materials, the peak pair 203/208 keV provides the best choice for the $^{239}Pu/^{241}Pu$ ratio determination.

There are also a few $^{239}Pu/^{241}Pu$ ratio values reported which have been determined from the 332/345 keV region (see Tables B.3, B.4 and C.2 in the Appendices). On an average, those values are in good agreement with the reference values (average ratio Gamma/NBS = 1.002$\pm$2.42% for the 15 reported values). With respect to the $^{239}Pu/^{241}Pu$ ratio analysis from the 332/345 keV region it should be noted that for measurements with little gamma attenuation at close sample-to-detector distances possible corrections for the true coincidence summing of the cascading ^{239}Pu gamma lines 129.29 and 203.54 keV to the cross-over energy of 332.8 keV have to be considered. Taking the decay characteristics and the measurement geometries into account, the contribution of the coincidence summing events to the full energy peak of the 332.8 keV line from ^{239}Pu has been calculated to be about 1.6 % for the 'LEPS' spectra NBS 946,947,948, and about 0.5 % for the 'COAX' spectra. The effect will be more pronounced, for example, for the close source-to-detector geometries used by laboratories 1 and 6 in their own measurements. We should also mention that the coincidence summing 129+203 keV removes events from the full energy peaks at 129 and 203 keV, thereby affecting the measured relative detection efficiencies for these gamma rays.

C. <u>Pu-240/Pu-241 Ratios</u>

1. *Ratio from peak pair 160/148 keV.* The common problem encountered in the $^{240}Pu/^{241}Pu$ ratio measurements, using both the peak pair 160/148 keV and 160/164 keV, is related to the difficulty of obtaining an accurate peak area for the ^{240}Pu gamma line at 160.28 keV. This gamma line has a relatively low branching intensity, thereby providing limited counting precisions within reasonable counting times. Further, it receives strong interference from an unresolved ^{241}Pu gamma line at 159.96 keV, and to a lesser extent from an unresolved ^{239}Pu gamma line at 160.19 keV . The contribution of ^{241}Pu to the unresolved peak complex (159.96 + 160.19 + 160.28) keV amounts to about 65 %, 58 % and 22 % for NBS 946, 947 and 948, respectively. This explains the large scatter of the measured $^{240}Pu/^{241}Pu$ ratios. The ratios obtained from the peak pair 160/148 keV are summarized in Table 4.4. On an average, the gamma values for the

Table 4.4 Average $^{240}Pu/^{241}Pu$ Ratios from Peak Pair 160/148 keV and Error Components from Distributed Spectra

Sample/ Spectrum	240/241(γ/NBS)		RSD(%) of $A_{160}*/A_{148}$ [a]			RSD(%) of $\varepsilon_{148}/\varepsilon_{160}$	
	Grand Mean	RSD (%)	Grand Mean	Reduced Mean	Counting Statistics[b]	Grand Mean	Reduced Mean
946 LEPS	0.989	4.24	1.77	-	1.82	1.26	1.09
946 COAX	0.962	5.27	2.46	1.65	0.94	1.42	1.00
946 Own	1.005	8.01	-	-	-	-	-
947 LEPS	0.984	4.17	1.99	-	1.56	0.89	-
947 COAX	0.980	4.52	1.64	-	0.57	1.35	1.01
947 Own	1.002	5.76	-	-	-	-	-
948 LEPS	0.983	2.62	1.61	-	0.72	0.68	-
948 COAX	1.000	2.64	2.43	1.14	0.25	1.27	0.70
948 Own	0.999	3.73	-	-	-	-	-

[a] $A_{160}* = \sum(159.96 + 160.19 + 160.28)$ keV
[b] Counting Statistics for Ratio $A_{160.28}/A_{148}$

Table 4.5 Average $^{240}Pu/^{241}Pu$ Ratios from Peak Pair 160/164 keV and Error Components from Distributed Spectra

Sample/ Spectrum	240/241(γ/NBS)		RSD(%) of $A_{160}*/A_{164}$ [a]			RSD(%) of $\varepsilon_{164}/\varepsilon_{160}$	
	Grand Mean	RSD (%)	Grand Mean	Reduced Mean	Counting Statistics[b]	Grand Mean	Reduced Mean
946 LEPS	0.945	5.49	1.48	-	1.83	0.41	-
946 COAX	0.932	4.04	2.31	1.27	0.94	0.59	-
946 Own	0.957	8.67	-	-	-	-	-
947 LEPS	0.948	4.12	2.01	1.76	1.58	0.40	-
947 COAX	0.954	4.21	1.45	-	0.57	0.57	-
947 Own	0.952	6.48	-	-	-	-	-
948 LEPS	0.975	3.35	1.82	1.29	0.73	0.42	-
948 COAX	0.978	5.61	2.19	1.25	0.33	1.15	0.58
948 Own	0.978	3.81	-	-	-	-	-

[a] $A_{160*} = \sum(159.96 + 160.19 + 160.28)$ keV
[b] Counting Statistics for Ratio $A_{160.28}/A_{164}$

isotopic ratio are close to the reference values, showing an average bias of about -1 %. The standard deviations of the gamma values, however are relatively large for the above reason. The larger uncertainties from the laboratories' own measurements are due to the generally poorer counting precisions in the measurements. The precisions for the net peak area ratios $A_{160.28}(^{240}Pu)/A_{148}(^{241}Pu)$ for the distributed 'LEPS' and 'COAX' spectra are listed in Table 4.4 together with the standard deviations of the measured ratios $A_{160}*/A_{148}$, with $A_{160}*$ being the peak area of the total peak complex (159.96 + 160.19 + 160.28) keV. For NBS 946 and 947, the error of the peak area ratio A_{160*}/A_{148} will propagate about doubled into the measured isotopic ratio because of the large corrections for the ^{241}Pu interference in the 160* keV complex. In fact, this is observed from the relative standard deviations given in Table 4.4 . For NBS 948 with its smaller ^{241}Pu interference to the 160 keV peak, the RSD values for the ratio A_{160*}/A_{148} and for the measured isotopic ratios are closer to each other, as expected. From the Table we may also note that for the peak pair 160/148 keV the corrections for detection efficiency contribute a notable amount to the total assay error.

The following average biases from individual analyses have been deduced from the reported data: + 4.8 % for laboratory 1/1 and -4.6 % for laboratory 1/2. In these cases the isotopic ratios have been evaluated by the author from raw data supplied by the laboratory. The results from laboratory 3/1 show an average bias of -3.8 % because of an overestimation of the 148 keV peak area.

During the evaluations of the data we identified another source of error which can affect the ^{240}Pu abundance measurements involving the 160.28 keV line of ^{240}Pu. There can occur notable pulse summing effects to the 160 keV line due to chance summing of 59.54 keV gamma rays from ^{241}Am with the neptunium $K_{\alpha 1}$ X rays at 101.066 keV. For the distributed 'LEPS' and 'COAX' spectra we have calculated the chance summing contribution

$$N_{ch} = N(59.54\ \text{keV}) \cdot N(101.066\ \text{keV}) \cdot 2\tau$$

to the 160 keV line from the observed peak countrates in the 59 keV peak and in the X-ray peak. The effective time interval 2τ within the two pulses can sum up to the full summing energy has been determined from separate measurements with ^{241}Am and ^{57}Co sources to 2τ = 0.35 μsec for the given amplifier shaping time of 2 μsec. With this data, the following percentage chance summing contributions to the net 160.28 keV (^{240}Pu) peak area have been deduced for the distributed spectra:

Sample/Spectrum	Chance Summing Contribution to 160.28 keV Line (%)
NBS 946 LEPS	1.95
NBS 947 LEPS	1.36
NBS 948 LEPS	0.12
NBS 946 COAX	2.65
NBS 947 COAX	2.15
NBS 948 COAX	0.94

Taking the summing corrections into account implies a reduction of the isotopic ratios 240/241 given in Tables 4.4 and 4.5 by the above percentage values. The corrected isotopic ratios then show an enlarged bias with respect to the reference values, which we attribute to biases in the gamma branching intensities involved in the analysis as discussed in Chapter V. No corrections for chance summing could be applied to the results from the laboratories' own measurements because of the lack of the necessary raw data.

We have also investigated pulse summing effects, either random or coincident, to other gamma rays between 125 and 208 keV (for example chance summing of 59.54 + 148.57 = 208.0 keV). Those investigations, however, did not reveal any notable effects (less than 0.1 % in each case).

2. *Ratio from peak pair 160/164 keV.* The results from this evaluation are summarized in Table 4.5. The same findings as discussed above for the evaluation of the peak pair 160/148 keV also apply to the peak pair 160/164 keV. Regarding the results there are two differences to the former evaluations:

i) The isotopic ratios 240/241 from the peak pair 160/164 keV are significantly lower, showing average biases of -5.3 %, -4.9 %, and -2.3 % for NBS 946, 947 and 948, respectively. We suspect that systematic errors of the gamma branching intensities involved are the reason for the observed biases. We have reevaluated new branching intensity ratios (see Chapter V) in order to bring the results from the gamma measurements in accordance with the mass-spectrometric reference values.

ii) The errors from the efficiency corrections are relatively small, contributing only a minor part to the overall assay error.

A common observation made from the results presented in Tables 4.4 and 4.5 is the relatively large difference between the expected and measured standard deviation of the peak area ratios A_{160}/A_{164} and A_{160}/A_{148} determined from the 'COAX' spectra taken from NBS 948. Probably the problem of proper background assessment for the 160 keV peak - due to the structured background in front of the 160 keV complex (compare Fig. 2.1 on page 6) - might be the reason for this.

Individual results for the 240/241 ratio from the peak pair 160/164 keV show an average bias of -4.2 % for laboratory 1/2, and of -5.3 % for laboratory 3/1. Both biases have their origin in the measured peak area ratios A_{160}/A_{164}.

D. Am-241/Pu-239 Ratios

The $^{241}Am/^{239}Pu$ ratios have been determined from the closely spaced gamma lines 125.29 keV (^{241}Am) and 129.29 keV (^{239}Pu). The results are summarized in Table 4.6. The isotopic ratios given in the Table are normalized to a reference date of January 1st, 1978. There are no reference data from the destructive analysis available to check the validity of the measured ratios.

Table 4.6 Average $^{241}Am/^{239}Pu$ Ratios from Peak Pair 125/129 keV and Error Components from Distributed Spectra

Sample/ Spectrum	Am/239(01/01/78)		RSD(%) of $A_{125}*/A_{129}$ [a]			RSD(%) of $\varepsilon_{129}/\varepsilon_{125}$	
	Grand Mean	RSD (%)	Grand Mean	Reduced Mean	Counting Statistics [b]	Grand Mean	Reduced Mean
	x 10^{-2}						
946 LEPS	1.614	1.93	0.50	-	0.22	1.65	-
946 COAX	1.651	3.75	0.63	-	0.10	4.19	1.82
946 Own	1.640	3.19	-	-	-	-	-
947 LEPS	1.850	1.74	0.48	-	0.26	1.14	-
947 COAX	1.882	4.41	1.15	0.42	0.10	3.90	2.15
947 Own	1.854	5.24	-	-	-	-	-
948 LEPS	0.433	3.59	2.05	1.23	0.19	1.70	-
948 COAX	0.431	4.38	1.53	1.25	0.05	2.51	1.83
948 Own	0.428	3.71	-	-	-	-	-

[a] $A_{125*} = \sum(125.21 + 125.29)$ keV
[b] Counting statistics for $\sum(125.21+125.29)$ keV

Although differing by only 4 keV in energy, the efficiency correction for the two isotopic gamma rays turns out to be the most critical factor in the gamma-spectrometric $^{241}Am/^{239}Pu$ ratio measurement from the given peak pair. The data given in Table 4.6 show that the uncertainties from the efficiency ratio correction represent a significant part of the total assay errors. The necessity for extrapolating the efficiency curves down to 125 keV, and the generally large gradient of the efficiency curve versus energy in this energy region may be factors contributing to the uncertainties of the efficiency correction.

The ^{241}Am gamma line at 125.29 keV receives interference from an unresolved ^{239}Pu gamma line at 125.21 keV - with contributions to the total peak area of about 2 % for NBS 946 and 947, and of about 7 % for NBS 948 - and from a partly resolved ^{239}Pu gamma line at 124.51 keV. Defining an appropriate background level underneath the 125 keV line seems to be the most critical factor in the peak area evaluation of the ^{241}Am gamma line at 125.29 keV (compare Fig. 2.1).

We have not identified a significant average bias for the $^{241}Am/^{239}Pu$ ratio determination from any evaluation method. We observed, however, that the efficiency ratios $\varepsilon_{129}/\varepsilon_{125}$ determined from the functional relationship used by laboratory 4 (see Table 2.7) deviate significantly from the average values for the efficiency curves in the 'COAX' spectra.

A few $^{241}Am/^{239}Pu$ ratio values have been also reported from the 332/345 keV region (see Tables B.7, B.8 and C.4). On an average, those values agree within about 3 % with the values determined from the 125/129 keV peak pair, but with a tendency to give somewhat lower ratio values.

E. Figures of Merit

Regarding the overall assessment of the isotopic ratio results from the present intercomparison exercise the question raises if there are methods of spectrum evaluation for the isotopic ratio analysis which provide a significant better overall performance as compared to others. We have tried to answer this question for the results obtained from the distributed 'LEPS' and 'COAX' spectra by calculating figures of merit (FOM) defined as following:

$$FOM = \frac{1}{N} \sum_{i,s} \frac{|R_{\ell is} - \bar{R}_{is}|}{\sigma(R_{\ell is})} \quad \text{with} \quad \begin{array}{l} i = \text{isotopic ratio} \\ s = \text{sample} \\ \sum_{i,s} = N, \end{array}$$

i.e., the average absolute deviations of the results from a given evaluation method ℓ (represented by the laboratory code) from the grand mean values $\bar{R}_{i,s}$, expressed in units of the relative standard deviations $\sigma(R_{\ell is})$. We have calculated the figure of merit values separately for the results from the analysis of the distributed 'LEPS' and 'COAX' spectra, respectively, summing up the absolute deviations for the number of reported isotopic ratios (typically N = 18 for 6 ratios i from each of the 3 NBS samples). The figure of merit values are listed in Table 4.7, and plotted in Fig. 4.1. We note gradual changes of the overall performance, corresponding to average deviations from the grand mean values between about 0.6σ and 1.2σ. We also note that the performance data for a given evaluation method are not always consistent for the two types of spectra.

When interpreting the performance data we have to keep in mind that the results from the intercomparison analyses have been obtained

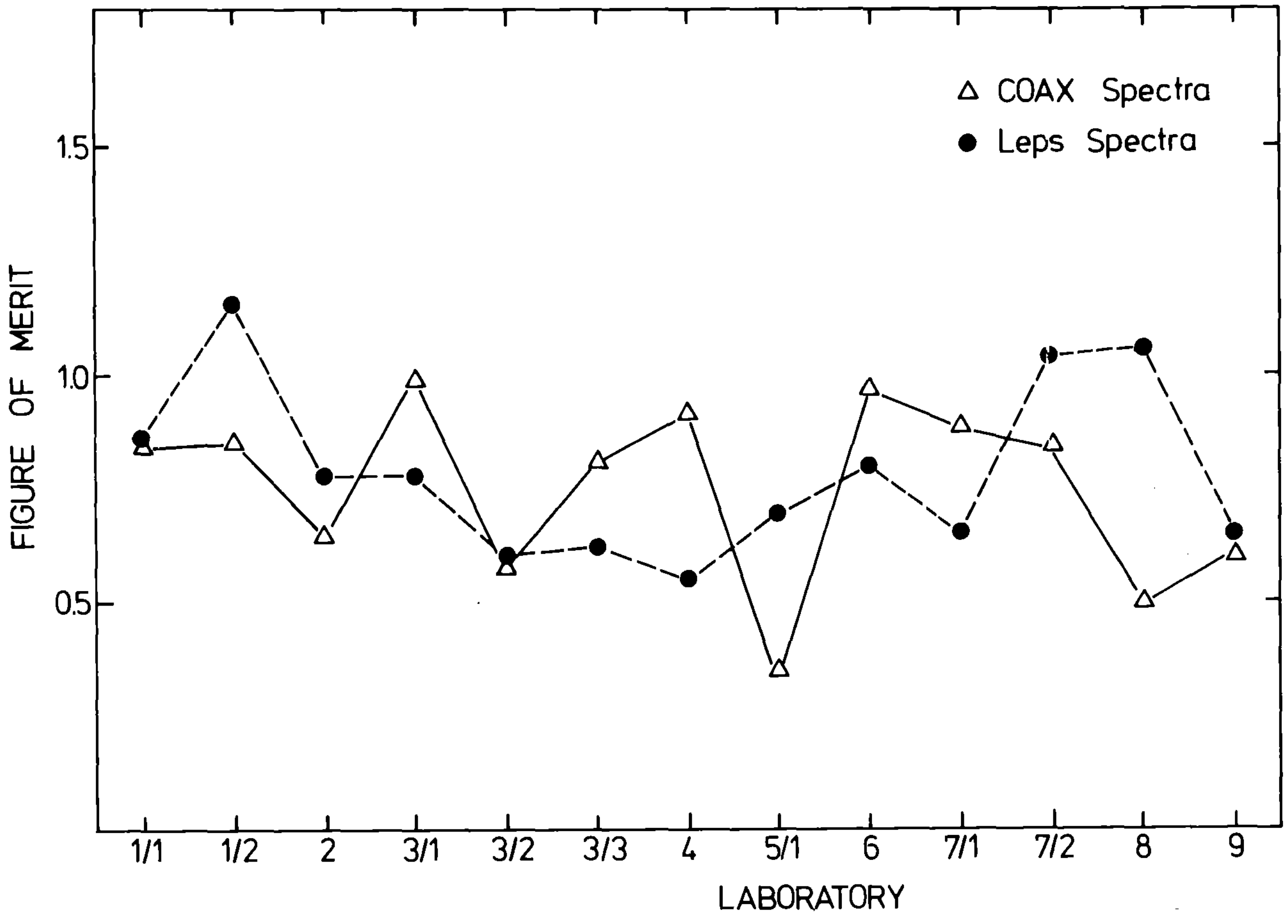

Fig. 4.1 Figures of merit for the analyses of distributed 'LEPS' and 'COAX' spectra.

i) from samples of different isotopic composition resulting in changing overall spectral features, and

ii) from gamma spectrometers exhibiting different energy resolution, peak shape parameters and energy dependence of the overall detection efficiency.

Therefore it is not surprising that the different methods of spectrum evaluation, being sensitive to some extent to any of these parameters, exhibit comparatively uniform performance data when averaging over the range of isotopic distributions and spectrometer hardware features. Another consideration refers to the fact that elaborated peak fitting programs require detailed informations about the peak shape parameters. We must admit that these data - without any a priori informations about detector characteristics - are difficult to obtain from plutonium spectra alone, because there are only a few isolated gamma peaks available from the complex plutonium spectra which can be used for an accurate determination of peak shape parameters.

Table 4.7 Figures of Merit for the Analysis of Distributed 'LEPS' and 'COAX' Spectra

Laboratory/ Analysis	LEPS Spectra		COAX Spectra	
	Number N of Ratios	Figure of Merit	Number N of Ratios	Figure of Merit
1/1	18	0.858	18	0.852
1/2	18	1.155	18	0.857
2	18	0.776	18	0.647
3/1	18	0.778	18	0.993
3/2	18	0.606	18	0.584
3/3	18	0.622	18	0.817
4	18	0.522	18	0.922
5/1	18	0.696	18	0.356
6	15	0.798	15	0.971
7/1	9	0.657	9	0.889
7/2 [a]	24	1.041	23	0.839
8	18	1.061	18	0.503
9	6	0.648	6	0.707

[a]Provided a total of 12 values for the 240/241 ratios

V. REEVALUATION OF GAMMA BRANCHING INTENSITY RATIOS

A. The Data Base

The reevaluation of some gamma branching intensity ratios starts from the assumption that the observed average deviations of the measured isotopic ratios from the mass-spectrometric reference values are due to errors of the respective gamma branching intensity ratios. This assumption implies that the number of different evaluations of peak area ratios and detection efficiency ratios involved in the present intercomparison measurements, when averaged, will provide unbiased mean values which are close to the true values. With this premise, some branching intensity ratios have been reevaluated from the available data, using the following data base:

1. $^{238}Pu/^{241}Pu$ ratios for NBS 946, 947 updated from the remeasurements performed at CBNM Geel (see Table 2.5).
2. $^{239}Pu/^{240}Pu$ and $^{239}Pu/^{241}Pu$ ratios for NBS 946, 947, 948 updated from the NBS reference data (see Table 2.5).
3. The half lives

   ^{238}Pu: 87.74 ± 0.04 y (Recommended by U.S. Half-Life

   ^{239}Pu: 24119 ± 26 y Evaluation Committee)

   ^{240}Pu: 6550 ± 20 y Ref. 5

   ^{241}Pu: 14.35 ± 0.02 y Ref. 4

   ^{241}Am: 432.6 ± 0.6 y Ref. 5
4. The grand mean values of the isotopic ratios determined from the laboratories' own measurements.
5. The reduced mean values of the peak area ratios and relative detection efficiency ratios determined from the analyses of the distributed 'LEPS' and 'COAX' spectra, with the following corrections applied to peak areas:

   correction to the 160 keV complex for chance summing and ^{239}Pu (160.19 keV) contribution, and

   correction for ^{241}Am interference to the gamma lines 164.58 keV (^{241}Pu-U), 203.54 keV (^{239}Pu) and 208.00 keV (^{241}Pu-U) using the measured $^{241}Am/^{239}Pu$ ratios and $^{239}Pu/^{241}Pu$ ratios calculated from the updated NBS data.

The applied percentage corrections for interferences are summarized in Table 5.1

Table 5.1 Percentage Corrections Applied to Reported Peak Areas from the Distributed Spectra

Sample/ Spectrum	Gamma Peak (keV)				
	164.58	203.54	208.00	160^{*a}	
	^{241}Am(%)			^{239}Pu(%)	Chance Summing(%)
NBS 946 LEPS	2.17	0.47	2.19	0.98	0.67
NBS 947 LEPS	1.99	0.53	2.01	0.74	0.57
NBS 948 LEPS	5.22	0.15	5.26	3.59	0.09
NBS 946 COAX	1.63	0.38	1.64	0.93	0.87
NBS 947 COAX	1.45	0.42	1.47	0.68	0.86
NBS 948 COAX	5.17	0.13	5.22	3.54	0.70

[a] $160^* = \sum(159.96 + 160.19 + 160.28)$ keV

B. Evaluation of Branching Intensity Ratios

Branching intensity ratios B_i/B_k have been reevaluated from the relations

$$(B_i/B_k)_{Reevaluated} = (\overline{A_i/A_k}) \times (\overline{\varepsilon_k/\varepsilon_i}) \times (T_{1/2})_i/(T_{1/2})_k \times (N_k/N_i)_{DA}$$

when using the average peak area ratios and relative detection efficiency ratios from the distributed spectra, and from the relation

$$(B_i/B_k)_{Reevaluated} = (B_i/B_k)_{Used} \times (N_i/N_k)_{Gamma}/(N_i/N_k)_{DA}$$

when using the average isotopic ratios from the laboratories' own measurements. In this way the branching intensity ratios 129 keV (^{239}Pu)/148 keV (^{241}Pu), 208 keV (^{241}Pu-U)/203 keV(^{239}Pu) and 152 keV (^{238}Pu)/148 keV(^{241}Pu) have been redetermined, using the data base as mentioned above in Section A. The results for the 3 ratios are summarized in Table 5.2.

Table 5.2 Reevaluated Branching Intensity Ratios 129/148, 152/148 and 208/203 keV

Sample/ Spectrum		Branching Intensity Ratio $B_{E_i}(keV)/B_{E_k}(keV)$				
	$B_{129.3}$ / $B_{148.6}$		$B_{152.7}$ / $B_{148.6}$		$B_{208.0}$ / $B_{203.5}$	
NBS 946	LEPS	32.76 (3.27)[a]	4.994	(0.79)	0.9678	(1.53)
	COAX	32.94 (2.95)	4.967	(1.20)	0.9581	(1.92)
	Own	34.05 (1.27)	4.973	(1.66)	0.9625	(1.62)
NBS 947	LEPS	33.21 (2.86)	4.984	(0.72)	0.9763	(1.26)
	COAX	33.14 (3.11)	4.978	(1.24)	0.9761	(1.71)
	OWN	33.71 (1.75)	4.980	(1.23)	0.9600	(1.65)
NBS 948	LEPS	33.23 (1.63)			0.9468	(0.66)
	COAX	33.51 (2.45)			0.9514	(0.82)
	Own	33.58 (2.52)			0.9578	(1.14)
Weighted Mean		33.545	4.983		0.9561	
RSD of Mean (%)		0.42	0.08		0.37	

[a]Values in parentheses are relative standard deviations in %.

For the evaluation of the branching ratio 152/148 keV only the data from NBS 946 and 947 have been used because of the lack of an accurate reference value for the ^{238}Pu abundance in NBS 948. The weighted mean values and the standard error of the mean are calculated from the expressions

$$\bar{X} = \sum g_i X_i / \sum g_i \quad \text{and}$$

$$RSD(\bar{X}) = \left(\sum g_i (X_i - \bar{X})^2 / (n-1) \sum g_i \right)^{1/2}$$

with $g_i = 1 / \sigma^2_i$.

For the reevaluation of branching intensity ratios involved in the $^{240}Pu/^{241}Pu$ ratio determination from the gamma lines 160.28 keV(^{240}Pu) and 148, 164 keV (^{241}Pu) a different procedure has been used. It is easily shown that the following relation holds:

$$\frac{N_{240}}{N_{241}} \cdot \frac{(T_{1/2})_{241}}{(T_{1/2})_{240}} \cdot \frac{\varepsilon_{160.28}}{\varepsilon_{159.96}} = -\frac{B_{159.96}}{B_{160.28}} + \frac{B_i}{B_{160.28}} \cdot \frac{A_{160*}}{A_i} \cdot \frac{\varepsilon_i}{\varepsilon_{160.28}}$$

where the subscript i stands either for 148.57 keV or 164.58 keV. With the respective input data, the branching intensity ratios $B_{159.96}(^{241}Pu)/B_{160.28}(^{240}Pu)$, $B_{164.58}(^{241}Pu\text{-}U)/B_{160.28}(^{240}Pu)$ and $B_{148.57}(^{241}Pu)/B_{160.28}(^{240}Pu)$ are obtained from linear least-squares fits to the above relation.

Table 5.3 Reevaluated Branching Intensity Ratios Involved in the $^{240}Pu/^{241}Pu$ Ratio Determination

Least-Squares Fit to Peak Pairs	Branching Intensity Ratios $B_{E_i}(keV) / B_{E_k}$ (keV)		
	$B_{159.96}/B_{160.28}$	$B_{148.6}/B_{160.28}$	$B_{164.6}/B_{160.28}$
160* / 148 keV	0.01666±0.00016	0.4723±0.0023	-
160* / 164 keV	0.01655±0.00007	-	0.11546±0.00023
Weighted Mean	0.01658±0.00010		

Table 5.4 Summary of Reevaluated Branching Intensity Ratios

Gamma Energies (Isotopes)	Branching Intensity Ratio		External Errors (%)	
	Previous /3/	Present	Half Lives	Atom Ratio
Directly Measured Ratios				
129(^{239}Pu)/148(^{241}Pu)	33.48 (0.4)	33.55 (0.4)[a]	0.25	0.18
152(^{238}Pu)/148(^{241}Pu)	5.112(0.6)	4.983(0.1)	0.15	0.90
160(^{240}Pu)/148(^{241}Pu)	2.150(0.8)	2.117(0.5)	0.45	0.22
160(^{240}Pu)/160(^{241}Pu)	59.64 (2.1)	60.31 (0.6)	0.45	0.22
160(^{240}Pu)/164(^{241}Pu-U)	8.874(0.9)	8.661(0.2)	0.45	0.22
208(^{241}Pu-U)/203(^{239}Pu)	0.950(0.5)	0.956(0.4)	0.25	0.18
Deduced Ratios[b]				
152(^{238}Pu)/160(^{240}Pu)	2.379(1.0)	2.354(0.5)	0.35	0.90
160(^{241}Pu)/148(^{241}Pu)	0.0360(2.1)	0.03510(0.8)	-	-
160(^{241}Pu)/164(^{241}Pu-U)	0.1488(2.1)	0.1436(0.6)	-	-

[a] Internal Percentage Error (1σ) from Gamma Measurement

[b] From Combinations of Directly Measured Ratios

The peak area A_{160*} refers to the sum of the 159.96 keV (^{241}Pu) and 160.28 keV(^{240}Pu) peak intensities, which is obtained from the total peak counts of the 160 keV complex after corrections for interferences from ^{239}Pu and chance summing. The peak area ratios and detection efficiency ratios determined from the distributed 'LEPS' and 'COAX' spectra for NBS 946,947,948, together with the corresponding half-life ratios and atom ratios, have been used as input data for the least-squares fitting. The branching intensity ratios thus obtained are given in Table 5.3 .

In Table 5.4 we summarize our results on some reevaluated branching intensity ratios as obtained from the evaluation of the present intercomparison measurements and analyses. Previous data taken from Ref. 3 are given for comparison. In the Table we have also included the external error components for the branching intensity ratios which are associated with the present uncertainties of the plutonium half lives, and with the isotopic ratios of the NBS reference materials. For a conservative estimate of the total uncertainty of the branching intensity ratios we have to add those external errors linearly to the internal errors from the gamma measurements. We may note that the external errors are comparable or even larger than the internal errors from the gamma comparison measurements. We hope that the outcome of the forthcoming PIDIE intercomparison exercise mentioned in the preface to this report will further extend and improve our knowledge about the nuclear data required for the plutonium isotopic analysis by gamma spectrometry.

VI. CONCLUSIONS

In summary, the following conclusions can be drawn from the results of the present intercomparison exercise on the determination of plutonium isotopic ratios from arbitrary samples of low and medium burnup, using gamma rays from the limited energy range between 125 and 208 keV:

1. Isotopic ratios have been determined with standard errors (1σ) of about 1 % to 2 % for the ratios $^{238}Pu/^{241}Pu$ and $^{239}Pu/^{241}Pu$, 3 % to 5 % for the ratio $^{240}Pu/^{241}Pu$ (from the distributed spectra), and 2 % to 4 % for the ratio $^{241}Am/^{239}Pu$.

2. The performance of the gamma-spectrometric ^{238}Pu assay is comparable with existing techniques. For reactor-grade materials, the expected accuracy obtainable from high-resolution gamma spectra is about 0.5 %. The overall accuracy of the ^{238}Pu assay by gamma spectrometry, however, is presently limited to about 1 % because of the lack of reference materials with more accurate reference data for this isotope, which would permit a more accurate determination of the gamma branching intensity ratio involved in the isotopic ratio measurement.
3. The applied methods for spectrum evaluation do not exhibit striking differences in the overall performance when averaging the results over the range of spectra from the different spectrometers and isotopic composition of the samples.
4. Highest priority should be given to satisfactory energy resolution performance, also at the expense of counting efficiency, since the isotopic ratio evaluations from high-resolution spectra tend to be less prone to systematic biases.
5. The standard errors for the detection efficiency corrections are generally small for pairs of isotopic gamma rays which differ not more than 5 keV in energy (about 0.5 % with the exception of the efficiency ratio correction $\varepsilon_{125}/\varepsilon_{129}$ for the $^{241}Am/^{239}Pu$ ratio determination). On an average, 3rd order polynomials in a log-log representation provide satisfactory efficiency correction performance. It appears that the problem of correctly defining the overall relative detection efficiency is closer related to the limited number of reference points than to the selected functional relationship for the efficiency curve. Iterative procedures, involving both the gamma rays from ^{239}Pu and ^{241}Pu-^{237}U, are recommended for establishing the overall relative detection efficiency.
6. Systematic deviations of the average gamma results from the mass-spectrometric reference values in the range of up to 5 % have been attributed to biases of gamma branching intensities. Some gamma branching intensity ratios have been reevaluated from the present results. Intercomparison measurements are considered as the most promising way to further extend and improve the data base of gamma branching intensity values required for the intrinsic calibration of plutonium isotopic ratio measurements by gamma spectrometry.

7. No attempt has been made to establish general accuracy data for isotopic ratio measurements by gamma spectrometry because of the limited range of samples, isotopic distributions and spectral informations used in the present intercomparison measurements and analyses. Some kind of standardization, both with respect to measurement conditions (type of gamma detector, counting times, pulse processing rate etc.) and spectrum evaluation, has to be achieved in order to arrive at some general accuracy data for given categories of materials.

VII. REFERENCES

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Appendix A: Graphical Presentation of Reported Isotopic Ratios from the Analysis of Distributed Spectra and from Laboratories' Own Measurements.

In the following figures A.1 to A.5 the measured plutonium isotopic ratios $^{238,239,240}Pu/^{241}Pu$, divided by the ratios calculated from the updated NBS reference values, are plotted versus the participant number. Fig. A.6 presents the ratios $^{241}Am/^{239}Pu$, normalized to a common reference date of January 1st, 1978. The solid horizontal lines represent the grand mean values of the reported data, and the dashed lines the calculated $\pm$ 1σ limits of the grand mean. Error bars, if given, correspond to reported 1σ errors.

Fig. A.1	$^{238}Pu/^{241}Pu$ Ratios from 152/148 keV
Fig. A.2	$^{239}Pu/^{241}Pu$ Ratios from 129/148 keV
Fig. A.3	$^{239}Pu/^{241}Pu$ Ratios from 203/208 keV
Fig. A.4	$^{240}Pu/^{241}Pu$ Ratios from 160/148 keV
Fig. A.5	$^{240}Pu/^{241}Pu$ Ratios from 160/164 keV
Fig. A.6	$^{241}Am/^{239}Pu$ Ratios from 125/129 keV

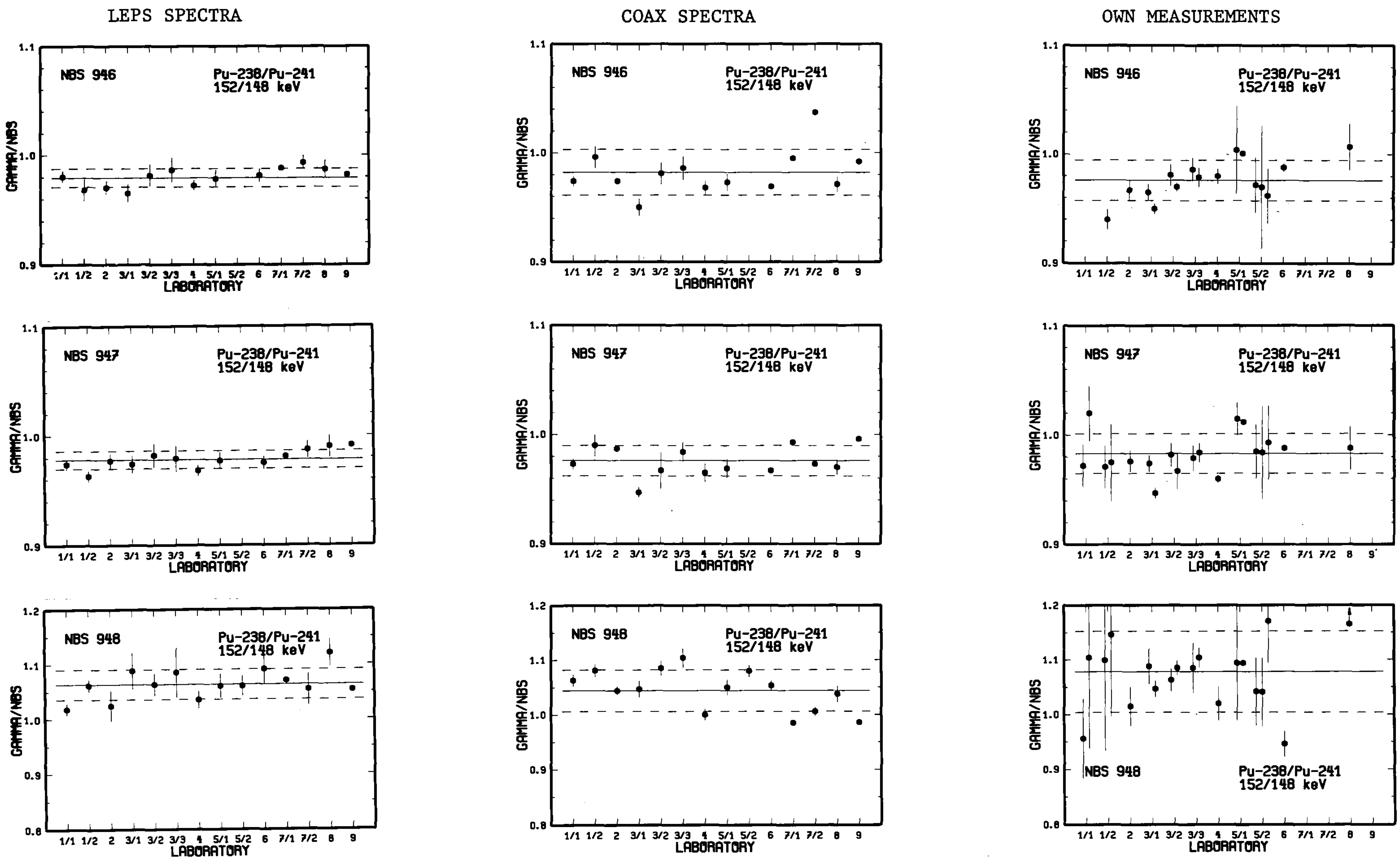


Fig. A1 ^{238}Pu/^{241}Pu ratios from peak pair 152/148 keV. From left to right: Analysis of distributed LEPS spectra, analysis of distributed COAX spectra, analysis of laboratories' own measurements.

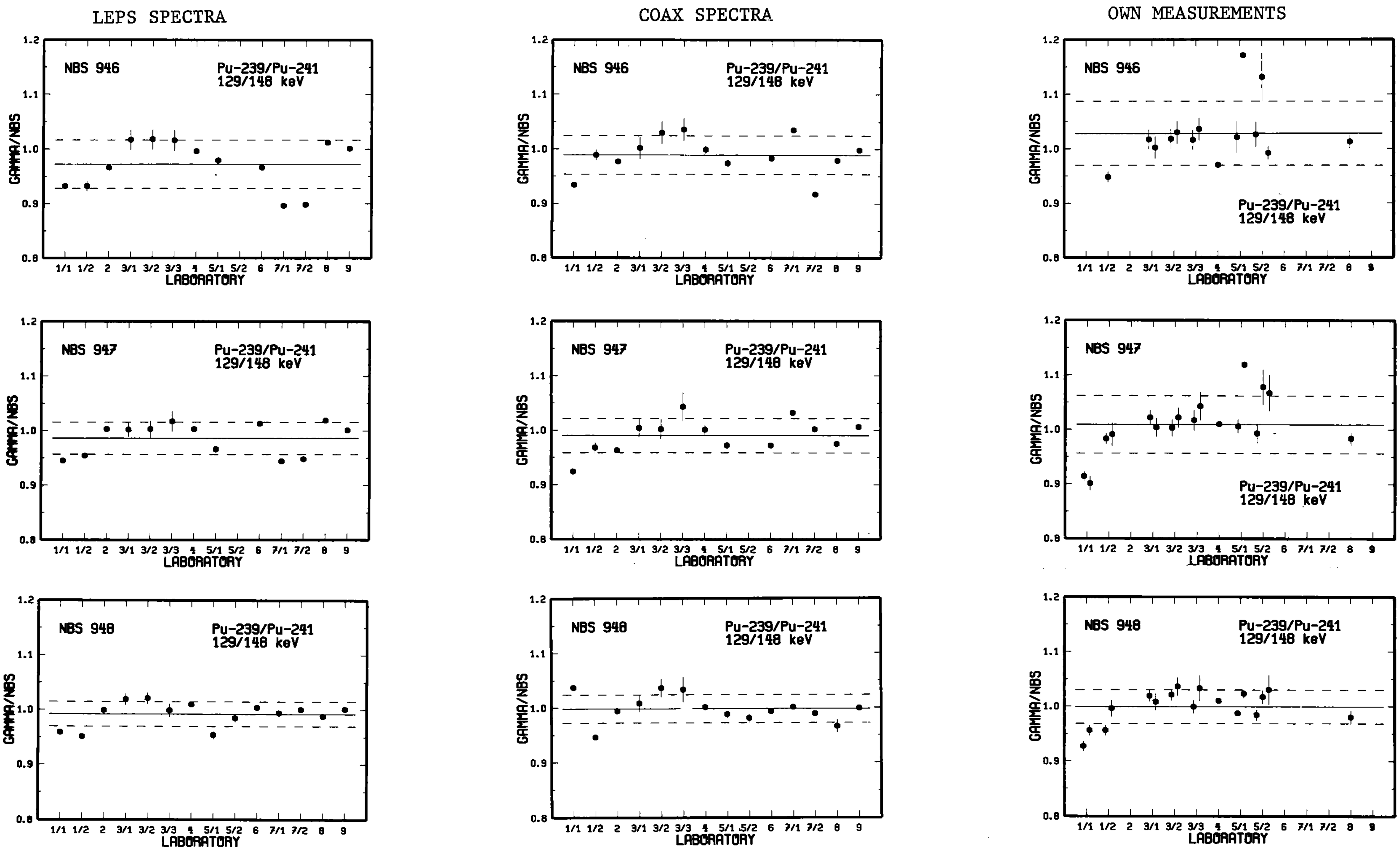


Fig. A.2 $^{239}Pu/^{241}Pu$ ratios from peak pair 129/148 keV. From left to right: Analysis of distributed LEPS spectra, analysis of distributed COAX spectra, analysis of laboratories' own measurements.

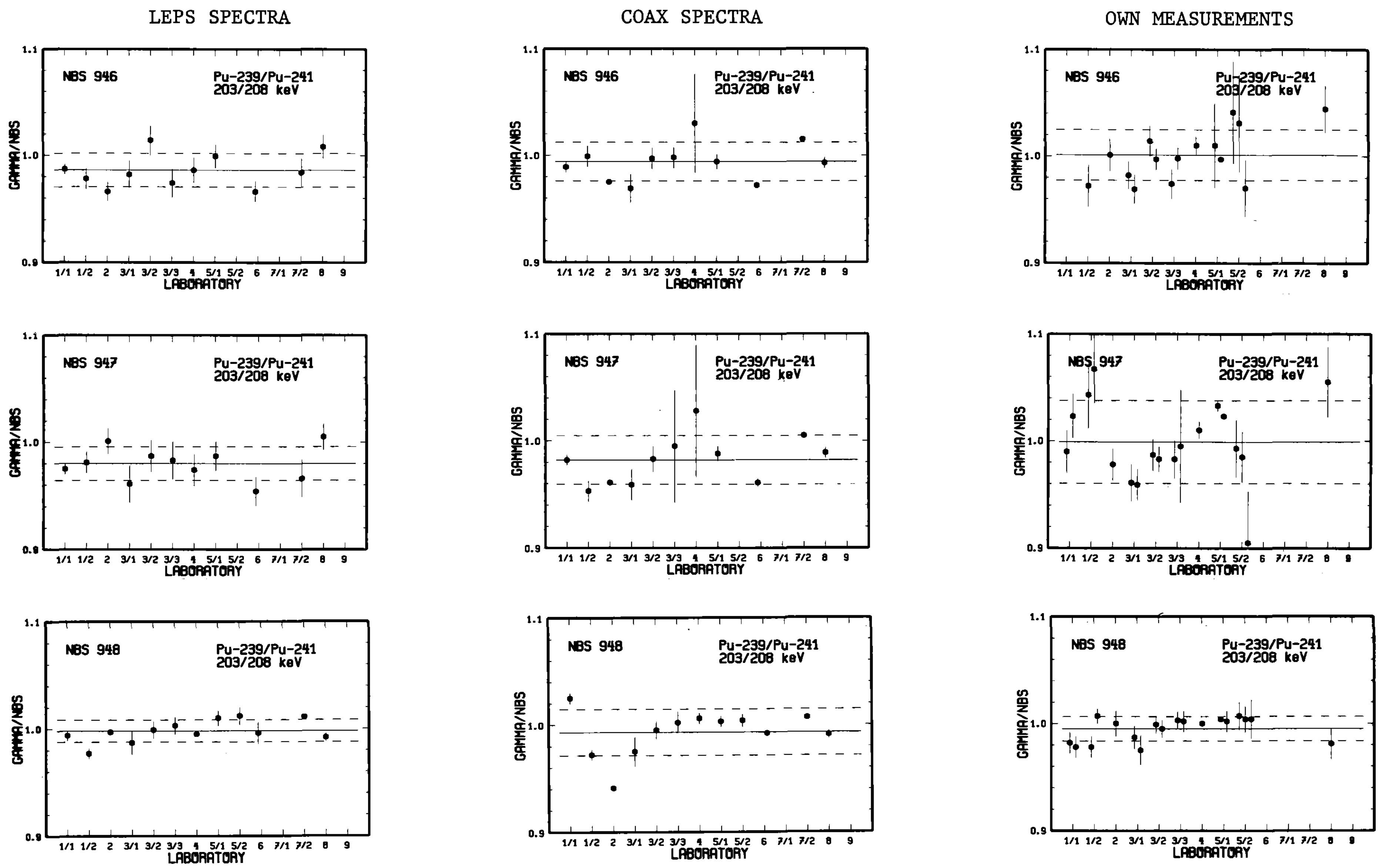


Fig. A.3 $^{239}Pu/^{241}Pu$ ratios from peak pair 203/208 keV. From left to right: Analysis of distributed LEPS spectra, analysis of distributed COAX spectra, analysis of laboratories' own measurements.

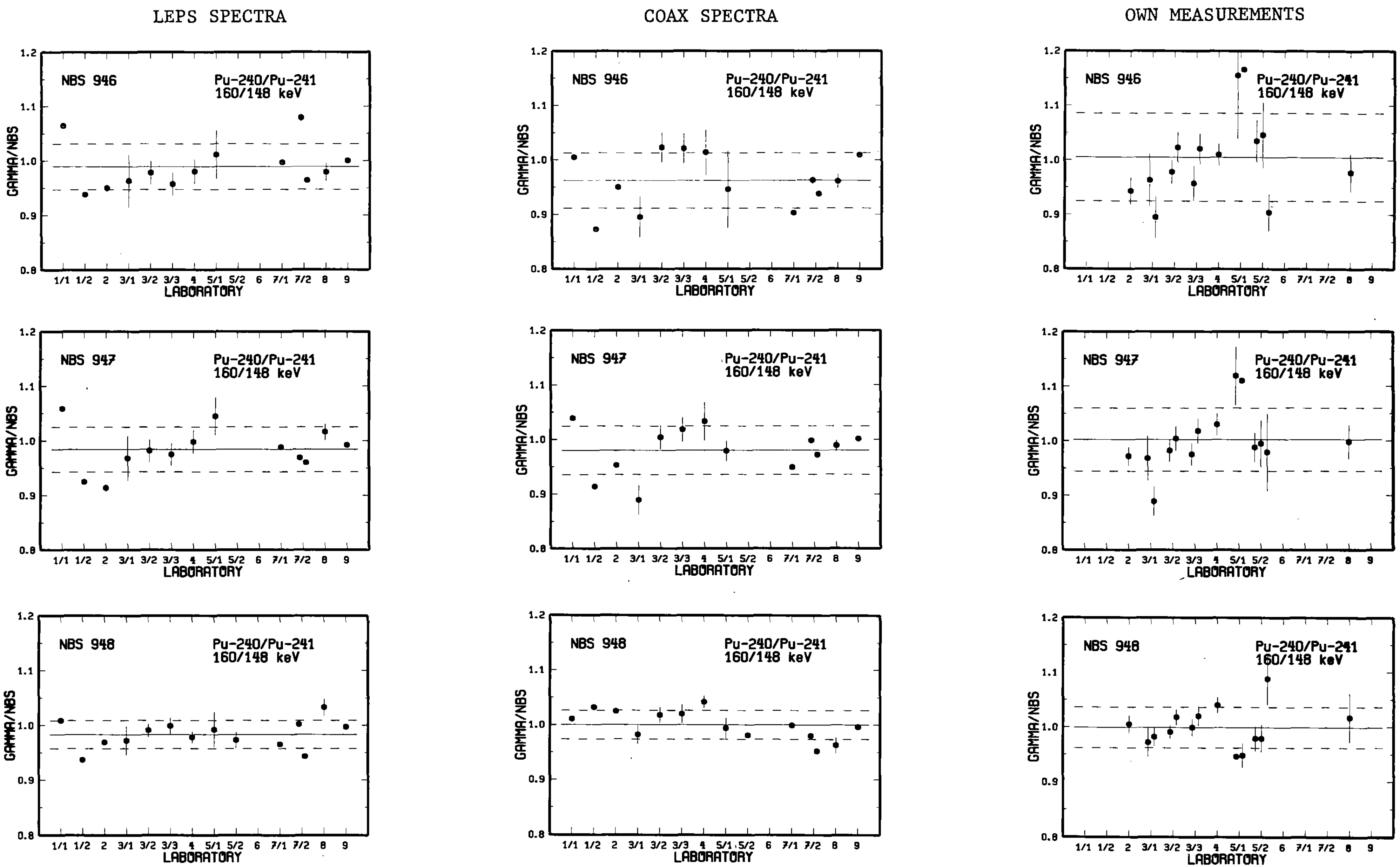


Fig. A.4 $^{240}Pu/^{241}Pu$ ratios from peak pair 160/148 keV. From left to right: Analysis of distributed LEPS spectra, analysis of distributed COAX spectra, analysis of laboratories' own measurements.

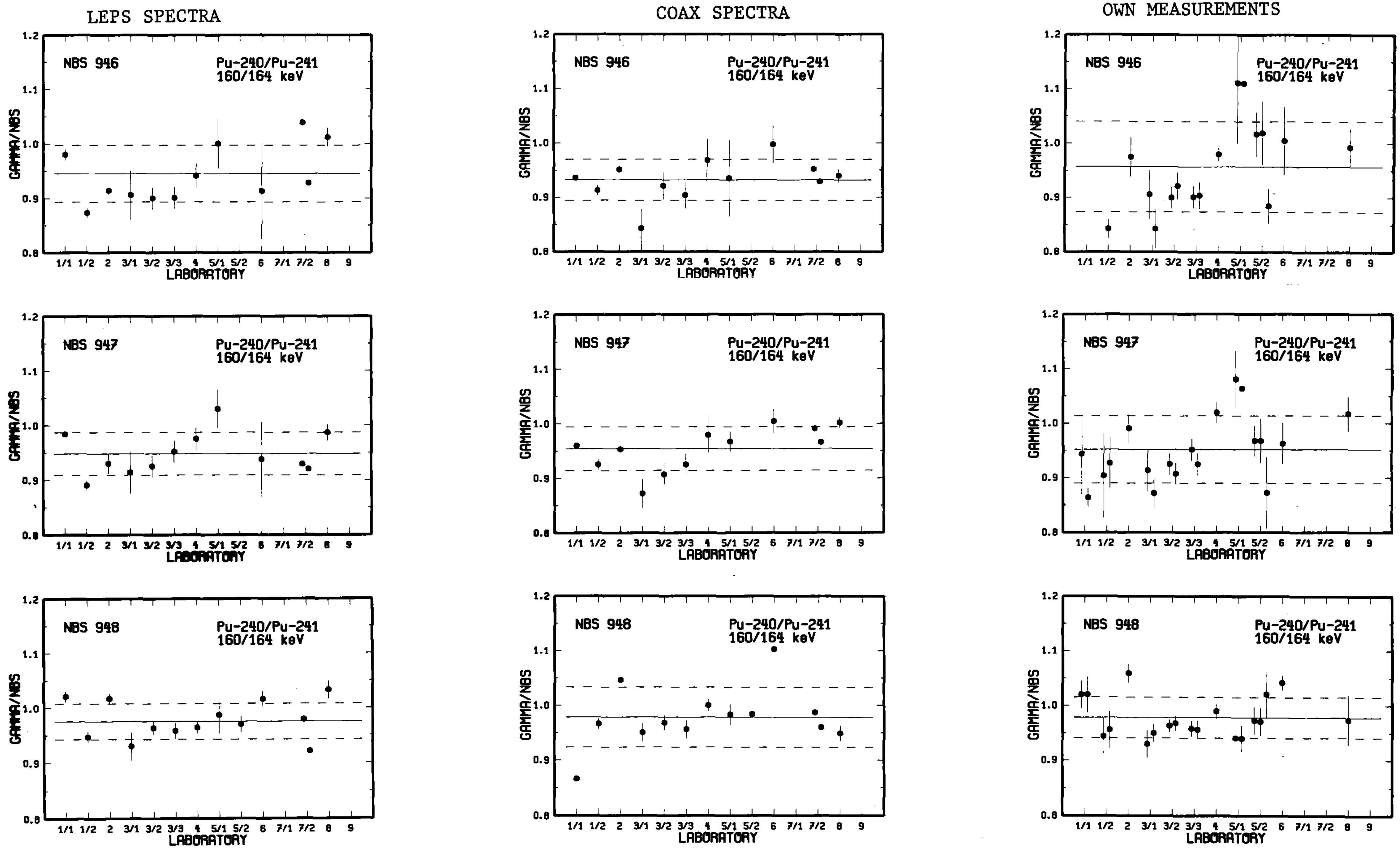
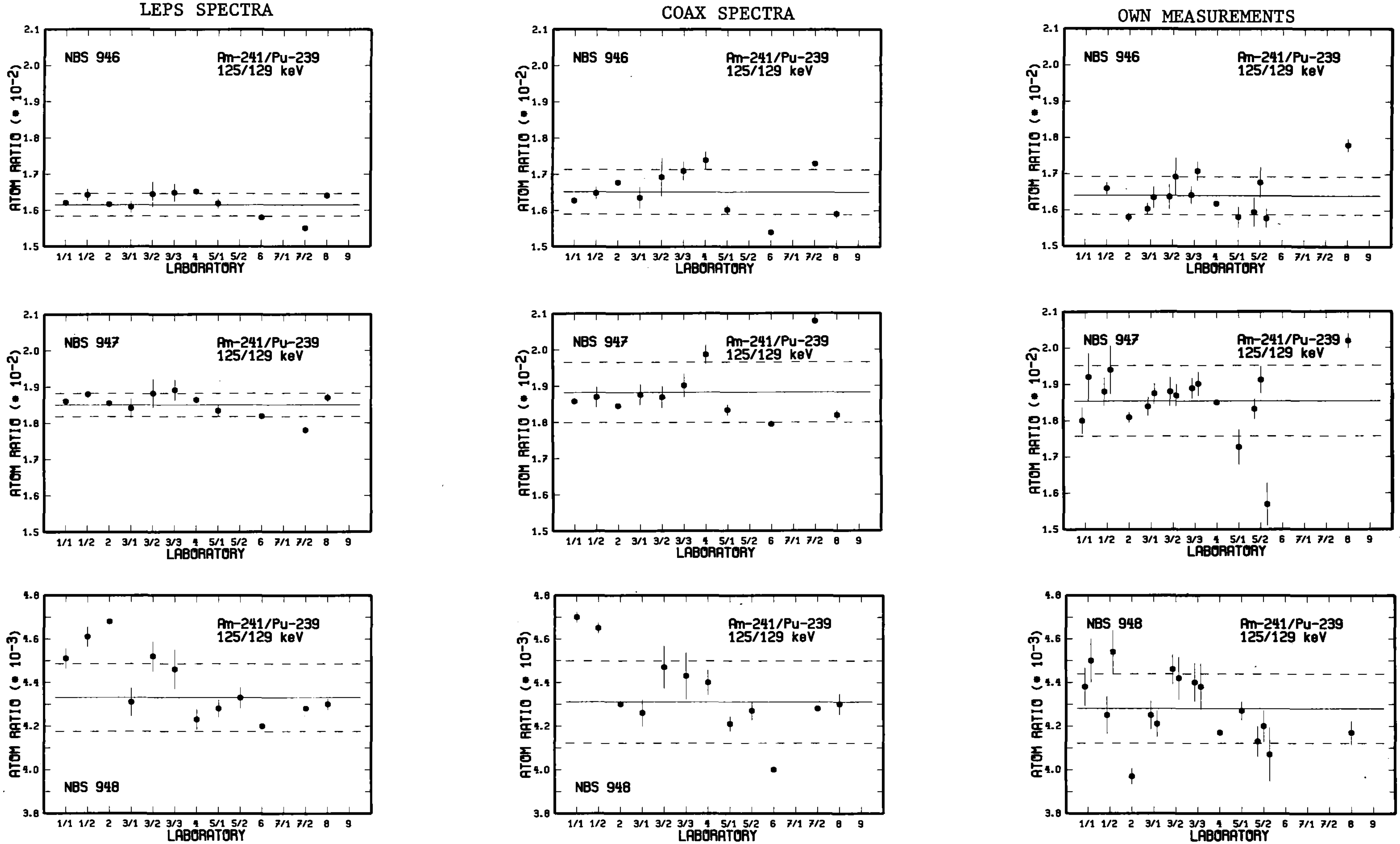


Fig. A.5. $^{240}Pu/^{241}Pu$ ratios from peak pair 160/164 keV. From left to right: Analysis of distributed LEPS spectra, analysis of distributed COAX spectra, analysis of laboratories' own measurements.



<u>Fig. A.6</u> ^{241}Am/^{239}Pu ratios from peak pair 125/129 keV. Values normalized to the reference date of January 1st, 1978. From left to right: Analysis of distributed LEPS spectra, analysis of distributed COAX spectra, analysis of laboratories' own measurements.

Appendix B: Numerical Presentation of Reported Isotopic Ratios from the Analysis of Distributed Spectra

Tables B.1 to B.8 summarize the reported isotopic ratios determined from the distributed 'LEPS' and 'COAX' spectra. The values given for the plutonium isotopic ratios are atom ratios from the gamma measurement divided by the atom ratios calculated from the updated NBS reference data. The ratios $^{241}Am/^{239}Pu$ refer to the dates of the gamma measurements (see Table 2.5 on page 11). Values normalized to a common reference date of January 1st, 1978 are also given. Values in parentheses are reported 1σ errors. The grand mean values are the unweighted mean values from all data.

Table B.1 Ratio Gamma Spectrometry / Ratio NBS for Pu-238/Pu-241 from LEPS Spectra

Lab./ Anal.		NBS 946	NBS 947	NBS 948
		152/148 keV	152/148 keV	152/148 keV
1/1		0.980 (0.5)	0.974 (0.5)	1.018 (1.0)
1/2		0.968 (1.0)	0.963 (0.5)	1.061 (1.0)
2		0.970 (0.6)	0.977 (0.7)	1.024 (2.7)
3/1		0.965 (0.8)	0.974 (0.8)	1.088 (3.0)
3/2		0.981 (1.0)	0.982 (1.1)	1.063 (1.9)
3/3		0.986 (1.1)	0.979 (1.2)	1.085 (4.2)
4		0.972 (0.45)	0.968 (0.5)	1.035 (1.51)
5/1		0.978 (0.8)	0.977 (0.8)	1.06 (2.1)
5/2				1.06 (1.7)
6		0.981 (0.6)	0.975 (0.6)	1.091 (2.5)
7/1		0.988	0.981	1.070
7/2		0.993 (0.6)	0.987 (0.8)	1.054 (2.7)
8		0.987 (0.8)	0.990 (1.0)	1.120 (2.2)
9		0.982[a]	0.991[a]	1.053[a]
	Mean	0.979	0.978	1.063
	RSD(%)	0.86	0.83	2.57

[a] Evaluated by the author from raw data given by the laboratory.

Table B.2 Ratio Gamma Spectrometry / Ratio NBS for Pu-238/Pu-241 from COAX Spectra

Lab./ Anal.		NBS 946	NBS 947	NBS 948
		152/148 keV	152/148 keV	152/148 keV
1/1		0.974 (0.5)	0.973 (0.5)	1.063 (1.0)
1/2		0.996 (1.0)	0.990 (1.0)	1.081 (1.0)
2		0.974 (0.3)	0.987 (0.3)	1.044 (0.8)
3/1		0.950 (0.8)	0.947 (0.5)	1.047 (1.4)
3/2		0.981 (1.0)	0.967 (1.7)	1.085 (1.2)
3/3		0.986 (1.1)	0.984 (0.9)	1.104 (1.6)
4		0.968 (0.65)	0.965 (0.87)	1.000 (1.03)
5/1		0.973 (0.8)	0.969 (0.9)	1.05 (1.3)
5/2				1.08 (1.0)
6		0.969 (0.2)	0.967 (0.2)	1.053 (0.8)
7/1		0.995	0.993	0.985
7/2		1.037 (0.24)	0.973 (0.2)	1.006 (0.8)
8		0.971 (0.7)	0.970 (0.7)	1.038 (1.4)
9		0.992[a]	0.996[a]	0.986[a]
	Mean	0.982	0.976	1.044
	RSD(%)	2.13	1.42	3.63

[a] Evaluated by the author from raw data given by the laboratory.

Table B.3 Ratio Gamma Spectrometry / Ratio NBS for Pu-239/Pu-241 from LEPS Spectra

Lab./ Anal.		NBS 946		NBS 947		NBS 948	
		129/148 keV	203/208 keV	129/148 keV	203/208 keV	129/148 keV	203/208 keV
1/1		0.932 (0.5)	0.987 (0.5)	0.945 (0.5)	0.975 (0.5)	0.959 (0.5)	0.994 (0.5)
1/2		0.932 (1.0)	0.978 (1.0)	0.954 (0.5)	0.981 (1.0)	0.951 (0.5)	0.977 (0.5)
2		0.966 (0.2)	0.966 (0.9)	1.003 (0.2)	1.001 (1.2)	0.999 (0.1)	0.997 (0.2)
3/1		1.017 (1.8)	0.982 (1.3)	1.002 (1.3)	0.961 (1.8)	1.019 (1.0)	0.987 (1.1)
3/2		1.018 (1.8)	1.014 (1.4)	1.003 (1.5)	0.987 (1.5)	1.021 (1.0)	0.999 (0.8)
3/3		1.016 (1.8)	0.974 (1.4)	1.017 (1.8)	0.983 (1.8)	0.999 (1.2)	1.003 (0.8)
4		0.996 (0.3)	0.986 (1.2)	1.003 (0.3)	0.974 (1.5)	1.010 (0.3)	0.995 (0.2)
5/1		0.979 (0.7)	0.999 (1.1)	0.966 (0.7)	0.987 (1.4)	0.954 (0.8)	1.010 (0.7)
5/2						0.985 (0.8)	1.012 (0.8)
6		0.966[a]	0.966[b] (1.0) 0.963[b] (1.1)	1.013[a]	0.954[b] (1.4) 1.011[b] (1.4)	1.004[a]	0.996[b] (1.0) 1.003[b] (1.2)
7/1		0.896		0.944		0.994	
7/2		0.898 (0.2)	0.984 (1.3)	0.948 (0.2)	0.966 (1.8)	1.000 (0.5)	1.011 (0.25)
8		1.012 (0.5)	1.008 (1.1)	1.019 (0.5)	1.005 (1.2)	0.988 (0.4)	0.992 (0.4)
9		1.001	0.994[c]	1.001	1.058[c]	1.001	1.034[c]
	Mean	0.972	0.986	0.986	0.980	0.992	0.998
	RSD(%)	4.57	1.59	2.99	1.60	2.27	1.03

[a] Evaluated by the author from raw data given by the laboratory.
[b] From 332-345 keV region. Values omitted from mean.
[c] From (332-345 keV)/414 keV region. Values omitted from mean.

Table B.4 Ratio Gamma Spectrometry / Ratio NBS for Pu-239/Pu-241 from COAX Spectra

Lab./ Anal.	NBS 946 129/148 keV	NBS 946 203/208 keV	NBS 947 129/148 keV	NBS 947 203/208 keV	NBS 948 129/148 keV	NBS 948 203/208 keV
1/1	0.934 (0.5)	0.989 (0.5)	0.924 (0.5)	0.982 (0.5)	1.037 (0.5)	1.025 (0.5)
1/2	0.989 (1.0)	0.999 (1.0)	0.968 (1.0)	0.953 (1.0)	0.946 (0.5)	0.972 (0.5)
2	0.977 (0.1)	0.975 (0.3)	0.963 (0.1)	0.961 (0.3)	0.994 (0.1)	0.941 (0.1)
3/1	1.002 (2.0)	0.969 (1.4)	1.004 (1.7)	0.959 (1.5)	1.008 (1.5)	0.975 (1.4)
3/2	1.030 (2.0)	0.997 (1.0)	1.002 (1.8)	0.983 (1.2)	1.036 (1.6)	0.995 (0.8)
3/3	1.036 (2.0)	0.998 (1.0)	1.043 (2.5)	0.995 (5.3)	1.033 (2.2)	1.002 (1.0)
4	0.999 (0.8)	1.030 (4.5)	1.001 (0.9)	1.028 (6.0)	1.001 (0.6)	1.006 (0.5)
5/1	0.974 (0.7)	0.994 (0.7)	0.972 (0.7)	0.988 (0.7)	0.988 (0.7)	1.003 (0.5)
5/2					0.981 (0.8)	1.004 (0.6)
6	0.983[a]	0.972[b] (0.3) 0.983[b] (0.3)	0.972[a]	0.961[b] (0.4) 0.969[b] (0.3)	0.993[a]	0.993[b] (0.1) 0.992[b] (0.3)
7/1	1.035		1.032		1.001	
7/2	0.917 (0.1)	1.015 (0.3)	1.002 (0.1)	1.005 (0.3)	0.989 (0.1)	1.007 (0.1)
8	0.979 (0.6)	0.993 (0.5)	0.975 (0.6)	0.989 (0.5)	0.966 (1.2)	0.991 (0.4)
9	0.998	1.018[c]	1.006	1.001[c]	0.999	0.997[c]
Mean	0.989	0.994	0.990	0.982	0.998	0.993
RSD(%)	3.58	1.82	3.18	2.31	2.57	2.18

[a] Evaluated by the author from raw data given by the laboratory.
[b] From 332-345 keV region. Values omitted from mean.
[c] From (332-335) / 414 keV region. Values omitted from mean.

Table B.5 Ratio Gamma Spectrometry / Ratio NBS for Pu-240/Pu-241 from LEPS Spectra

Lab./ Anal.	NBS 946		NBS 947		NBS 948	
	160/148 keV	160/164 keV	160/148 keV	160/164 keV	160/148 keV	160/164 keV
1/1	1.064[a]	0.980 (1.0)	1.059[a]	0.984 (0.5)	1.009[a]	1.021 (1.0)
1/2	0.938[a]	0.873 (1.0)	0.925[a]	0.891 (1.0)	0.937[a]	0.947 (1.0)
2	0.950 (0.7)	0.914 (0.8)	0.914 (0.8)	0.930 (2.0)	0.969 (0.6)	1.017 (0.9)
3/1	0.963 (5.0)	0.906 (5.0)	0.968 (4.2)	0.914 (4.2)	0.972 (2.7)	0.930 (2.7)
3/2	0.978 (2.2)	0.900 (2.2)	0.982 (2.1)	0.925 (2.1)	0.991 (1.2)	0.963 (1.2)
3/3	0.957 (2.2)	0.901 (2.2)	0.975 (2.1)	0.952 (2.1)	0.999 (1.5)	0.958 (1.5)
4	0.980 (2.3)	0.941 (2.3)	0.998 (2.1)	0.975 (2.1)	0.978 (1.1)	0.964 (1.1)
5/1	1.011 (4.4)	1.000 (4.5)	1.045 (3.3)	1.030 (3.4)	0.992 (3.3)	0.987 (3.4)
5/2					0.973 (1.5)	0.970 (1.5)
6		0.913 (9.7)		0.938 (7.3)		1.016 (1.4)
7/1	0.997		0.988		0.965	
7/2[b]	1.079 (0.7)	1.039 (0.7)	0.969 (0.3)	0.929 (0.4)	1.002 (0.6)	0.979 (0.7)
7/2[c]	0.964	0.928	0.960	0.920	0.943	0.921
8	0.979 (1.7)	1.012 (1.7)	1.016 (1.5)	0.987 (1.5)	1.033 (1.5)	1.033 (1.6)
9	1.000[d]		0.992[d]		0.997[d]	
Mean	0.989	0.945	0.984	0.948	0.983	0.975
RSD(%)	4.24	5.49	4.17	4.12	2.62	3.35

[a] Evaluated by the author from raw data given by the laboratory.
[b] Peak area of Pu-240 line determined from total intensity between 159.96 and 161.45 keV by subtraction of Pu-239 and Pu-241 interference.
[c] 159.96 - 160.28 keV and 161.45 keV peaks fitted separately.
[d] Evaluated by the author from the given ratios Pu-240/Pu-239 (160/129 keV) and Pu-241/Pu-239 (148/129 keV).

Table B.6 Ratio Gamma Spectrometry / Ratio NBS for Pu-240/Pu-241 from COAX Spectra

Lab./ Anal.	NBS 946		NBS 947		NBS 948	
	160/148 keV	160/164 keV	160/148 keV	160/164 keV	160/148 keV	160/164 keV
1/1	1.005[a]	0.936 (0.5)	1.039[a]	0.960 (0.5)	1.011[a]	0.866 (0.5)
1/2	0.873[a]	0.913 (1.0)	0.914[a]	0.925 (1.0)	1.032[a]	0.966 (1.0)
2	0.950 (0.3)	0.951 (0.7)	0.953 (0.5)	0.953 (0.6)	1.025 (0.2)	1.046 (0.2)
3/1	0.895 (4.2)	0.843 (4.2)	0.889 (3.0)	0.872 (3.0)	0.982 (1.7)	0.950 (1.7)
3/2	1.023 (2.7)	0.921 (2.7)	1.004 (2.2)	0.907 (2.2)	1.018 (1.4)	0.968 (1.4)
3/3	1.021 (2.7)	0.904 (2.7)	1.018 (2.2)	0.925 (2.2)	1.020 (1.7)	0.956 (1.7)
4	1.014 (4.1)	0.968 (4.1)	1.033 (3.4)	0.979 (3.4)	1.042 (1.1)	1.000 (1.1)
5/1	0.946 (7.5)	0.935 (7.5)	0.979 (1.9)	0.967 (1.9)	0.994 (1.9)	0.983 (1.9)
5/2					0.981	0.984
6		0.997 (3.5)		1.004 (2.2)		1.103 (0.4)
7/1	0.903		0.949		0.999	
7/2[b]	0.962 (0.7)	0.952 (0.7)	0.998 (0.6)	0.991 (0.6)	0.980 (0.6)	0.987 (0.6)
7/2[c]	0.938	0.929	0.972	0.966	0.952	0.960
8	0.961 (1.3)	0.940 (1.3)	0.989 (1.0)	1.002 (1.0)	0.963 (1.5)	0.949 (1.5)
9	1.009[d]		1.001[d]		0.996[d]	
Mean	0.962	0.932	0.980	0.954	1.000	0.978
RSD(%)	5.27	4.04	4.52	4.21	2.64	5.61

[a] Evaluated by the author from raw data given by the laboratory.
[b] Peak area of Pu-240 line determined from total intensity between 159.96 and 161.45 keV by subtraction of Pu-239 and Pu-241 interference.
[c] 159.96 - 160.28 keV and 161.45 keV peaks fitted separately.
[d] Evaluated by the author from the given ratios Pu-240/Pu-239 (160/129 keV) and Pu-241/Pu-239 (148/129 keV).

Table B.7 Atom Ratio Am-241/Pu-239 from LEPS Spectra

Lab./ Anal.	NBS 946		NBS 947		NBS 948	
	125/129 keV	332/345 keV	125/129 keV	332/345 keV	125/129 keV	332/345 keV
1/1	0.0162 (0.5)		0.0186 (0.5)		0.00451 (1.0)	
1/2	0.01642 (1.0)		0.0188 (0.5)		0.00461 (1.0)	
2	0.01616 (0.2)	0.01585 (1.6)	0.01855 (0.3)	0.01810 (1.9)	0.00468 (0.2)	0.00425 (1.1)
3/1	0.01609 (1.0)		0.01841 (1.4)		0.00431 (1.5)	
3/2	0.01643 (2.1)		0.01882 (2.1)		0.00452 (1.5)	
3/3	0.01648 (1.5)		0.01891 (1.5)		0.00446 (2.0)	
4	0.01651 (0.4)		0.01864 (0.5)		0.00423 (1.1)	
5/1	0.01618 (0.8)		0.01834 (0.8)		0.00428 (0.9)	
5/2					0.00433 (1.1)	
6	0.0158 (0.3)	0.0161 (1.6)	0.0182 (0.3)	0.0182 (1.9)	0.00420 (0.3)	0.00432 (1.1)
7/1						
7/2	0.01550 (0.5)		0.0178 (0.5)		0.00428 (0.2)	
8	0.0164 (0.6)		0.0187 (0.6)		0.0043 (0.6)	
9		0.01572[a]		0.01735[a]		0.00427[a]
Mean	0.01620	0.01589	0.01852	0.01788	0.00439	0.00428
Mean 01/01/78	0.01614	0.01583	0.01850	0.01786	0.00433	0.00422
RSD(%)	1.93		1.74		3.59	

[a] From (332-335) / 414 keV

Table B.8 Atom Ratio Am-241/Pu-239 from COAX Spectra

Lab./ Anal.	NBS 946		NBS 947		NBS 948	
	125/129 keV	332/345 keV	125/129 keV	332/345 keV	125/129 keV	332/345 keV
1/1	0.0130 (0.5)		0.0145 (0.5)		0.00470 (0.5)	
1/2	0.01317 (1.0)		0.0146 (1.5)		0.00465 (0.5)	
2	0.0134 (0.1)	0.01330 (0.4)	0.01440 (0.1)	0.01490 (0.4)	0.00430 (0.1)	0.00440 (0.1)
3/1	0.01306 (1.8)		0.01464 (1.5)		0.00426 (1.4)	
3/2	0.01352 (3.1)		0.01459 (1.6)		0.00447 (2.2)	
3/3	0.01365 (1.5)		0.01484 (1.7)		0.00443 (2.4)	
4	0.01389 (1.4)		0.01550 (1.3)		0.00440 (1.3)	
5/1	0.01280 (0.8)		0.01431 (0.8)		0.00421 (0.8)	
5/2					0.00427 (1.0)	
6	0.01230 (0.2)	0.01292 (0.5)	0.0140 (0.1)	0.0146 (0.4)	0.00400 (0.1)	0.00444 (0.2)
7/1 7/2	0.01382 (0.5)		0.01622 (0.1)		0.00428 (0.1)	
8	0.0127 (0.7)		0.0142 (0.7)		0.0043 (1.1)	
9		0.01292[a]		0.01467[a]		0.00442[a]
Mean	0.01319	0.01305	0.01469	0.01472	0.00436	0.00442
Mean 01/01/78	0.01651	0.01633	0.01882	0.01886	0.00431	0.00437
RSD(%)	3.75		4.41		4.38	

[a] From (332-335) /414 keV

Appendix C: Numerical Presentation of Reported Isotopic Ratios from Laboratories' Own Measurements.

The results from the laboratories' own measurements presented in this Appendix are compiled in the same manner as the results from the distributed spectra given in Appendix B.

Table C.1 Ratio Gamma Spectrometry / Ratio NBS for Pu-238/Pu-241 from Laboratories' Own Measurements

Lab./ Anal.	Meas. No.	NBS 946	NBS 947	NBS 948
		152/148 keV	152/148 keV	152/148 keV
1/1	1		0.972 (2.0)	0.956 (7.6)
	2		1.020 (2.5)	1.104 (15)
1/2	1	0.940 (1.0)	0.971 (2.0)	1.099 (15)
	2		0.975 (3.6)	1.146 (13)
2		0.967 (1.0)	0.976 (1.0)	1.015 (3.5)
3/1	1	0.965 (0.8)	0.974 (0.8)	1.088 (3.0)
	2	0.950 (0.5)	0.947 (0.5)	1.047 (1.4)
3/2	1	0.981 (1.0)	0.982 (1.1)	1.063 (1.9)
	2	0.970 (0.4)	0.967 (1.7)	1.085 (1.2)
3/3	1	0.986 (1.1)	0.979 (1.2)	1.085 (4.2)
	2	0.979 (0.9)	0.984 (0.9)	1.104 (1.6)
4		0.98 (0.7)	0.96 (0.4)	1.02 (3.0)
5/1[a]	1	1.004 (4.0)	1.015 (1.5)	1.094 (9.5)
5/1[b]	1	1.001	1.012	1.094
5/2[a]	1	0.972 (2.6)	0.985 (2.5)	1.042 (5.9)
5/2[b]	1	0.970 (5.8)	0.984 (4.3)	1.041 (6.0)
5/2[a]	2	0.962 (2.6)	0.993 (3.4)	1.171 (6.6)
6		0.988 (0.4)	0.988 (0.3)	0.946 (2.5)
8		1.007 (2.1)	0.988 (2.0)	1.278 (12)
	Mean	0.976	0.983	1.078
	RSD(%)	1.90	1.85	6.89

a 3^{rd} order polynomial log ε vs. log E for relative efficiency.
b 4^{th} order polynomial log ε vs. log E for relative efficiency.

Table C.2 Ratio Gamma Spectrometry / Ratio NBS for Pu-239/Pu-241 from Laboratories' Own Measurements

Lab./ Anal.	Meas. No.	NBS 946		NBS 947		NBS 948	
		129/148 keV	203/208 keV	129/148 keV	203/208 keV	129/148 keV	203/208 keV
1/1	1			0.914 (1.0)	0.990 (2.0)	0.927 (1.0)	0.982 (1.0)
	2			0.901 (1.4)	1.023 (2.0)	0.956 (1.0)	0.978 (1.0)
1/2	1	0.948 (1.0)	0.972 (2.0)	0.983 (1.0)	1.043 (3.0)	0.956 (1.0)	0.978 (1.0)
	2			0.991 (2.1)	1.067 (3.0)	0.996 (1.5)	1.007 (0.7)
2			1.001 (1.5)		0.978 (1.5)		1.000 (1.2)
3/1	1	1.017 (1.8)	0.982 (1.3)	1.022 (1.3)	0.961 (1.8)	1.019 (1.0)	0.987 (1.1)
	2	1.002 (2.0)	0.969 (1.4)	1.004 (1.7)	0.959 (1.5)	1.008 (1.5)	0.975 (1.4)
3/2	1	1.018 (1.8)	1.014 (1.4)	1.003 (1.5)	0.987 (1.5)	1.021 (1.0)	0.999 (0.8)
	2	1.030 (2.0)	0.997 (1.0)	1.022 (1.8)	0.983 (1.2)	1.036 (1.6)	0.995 (0.8)
3/3	1	1.016 (1.8)	0.974 (1.4)	1.017 (1.8)	0.983 (1.8)	0.999 (1.2)	1.003 (0.8)
	2	1.036 (2.0)	0.998 (1.0)	1.043 (2.5)	0.995 (5.3)	1.033 (2.2)	1.002 (1.0)
4		0.97 (0.3)	1.01 (0.8)	1.01 (0.3)	1.01 (0.8)	1.01 (0.3)	1.00 (0.3)
5/1[b]	1	1.021 (2.8)	1.010 (3.9)	1.006 (1.2)	1.033 (0.5)	0.987	1.004
5/1[c]	1	1.171	0.997	1.119	1.023	1.023 (0.8)	1.002 (1.0)
5/2[b]	1	1.026 (2.2)	1.041 (4.6)	0.993 (1.8)	0.993 (2.7)	0.984 (1.0)	1.007 (1.3)
5/2[c]	1	1.131 (3.9)	1.031 (4.4)	1.078 (3.0)	0.985 (2.4)	1.017 (1.2)	1.004 (1.2)
5/2[b]	2	0.992 (1.2)	0.970 (2.7)	1.067 (3.1)	0.905 (5.3)	1.030 (2.6)	1.004 (1.8)
6			0.986[a] (0.6)		1.001[a] (0.6)		1.019[a] (1.1)
8		1.013 (1.2)	1.044 (2.1)	0.984 (1.2)	1.055 (3.1)	0.980 (1.2)	0.981 (1.5
	Mean	1.028	1.001	1.009	0.999	0.999	0.995
	RSD(%)	5.71	2.38	5.22	3.87	3.07	1.14

[a] From 332-345 keV region. Values omitted from mean.

[b] 3rd order polynomial log ε vs. log E for relative efficiency.

[c] 4th order polynomial log ε vs. log E for relative efficiency.

Table C .3 Ratio Gamma Spectrometry / Ratio NBS for Pu-240/Pu-241 from Laboratories' Own Measurements

Lab./ Anal.	Meas. No.	NBS 946		NBS 947		NBS 948	
		160/148 keV	160/164 keV	160/148 keV	160/164 keV	160/148 keV	160/164 keV
1/1	1				0.944 (8.0)		1.020 (2.5)
	2				0.864 (2.0)		1.020 (3.2)
1/2	1		0.843 (2.0)		0.904 (8.5)		0.944 (3.5)
	2				0.927 (5.0)		0.956 (3.5)
2		0.943 (2.6)	0.975 (3.7)	0.972 (1.7)	0.991 (2.7)	1.005 (1.6)	1.059 (1.6)
3/1	1	0.963 (5.0)	0.906 (5.0)	0.968 (4.2)	0.914 (4.2)	0.972 (2.7)	0.930 (2.7)
	2	0.895 (4.2)	0.843 (4.2)	0.889 (3.0)	0.872 (3.0)	0.982 (1.7)	0.950 (1.7)
3/2	1	0.978 (2.2)	0.900 (2.2)	0.982 (2.1)	0.925 (2.1)	0.991 (1.2)	0.963 (1.2)
	2	1.023 (2.7)	0.921 (2.7)	1.004 (2.2)	0.907 (2.2)	1.018 (1.4)	0.968 (1.4)
3/3	1	0.957 (3.3)	0.901 (2.2)	0.975 (2.1)	0.952 (2.1)	0.999 (1.5)	0.958 (1.5)
	2	1.021 (2.7)	0.904 (2.7)	1.018 (2.2)	0.925 (2.2)	1.020 (1.7)	0.956 (1.7)
4		1.01 (2.0)	0.98 (1.3)	1.03 (1.9)	1.02 (1.8)	1.04 (1.4)	0.99 (1.3)
5/1[a]	1	1.155 (10)	1.111 (10)	1.119 (4.8)	1.081 (4.8)	0.946	0.941
5/1[b]	1	1.166	1.110	1.110	1.064	0.948 (2.3)	0.940 (2.5)
5/2[a]	1	1.035 (3.7)	1.017 (4.0)	0.988 (2.7)	0.968 (2.9)	0.979 (2.3)	0.973 (2.5)
5/2[b]	1	1.046 (5.7)	1.019 (5.7)	0.995 (4.3)	0.968 (4.2)	0.979 (2.5)	0.971 (2.6)
5/2[a]	2	0.904 (3.7)	0.885 (3.6)	0.979 (7.2)	0.873 (7.5)	1.088 (4.3)	1.021 (4.1)
6			1.005 (6.3)		0.963 (3.9)		1.042 (1.3)
8		0.977 (3.5)	0.993 (3.5)	0.998 (3.1)	1.017 (3.1)	1.017 (4.4)	0.974 (4.7)
	Mean	1.005	0.957	1.002	0.952	0.999	0.978
	RSD(%)	8.01	8.67	5.76	6.48	3.73	3.81

a 3rd order polynomial log ε vs. log E for relative efficiency.
b 4th order polynomial log ε vs. log E for relative efficiency.

Table C.4 Atom Ratio $^{241}Am/^{239}Pu$ on 01/01/78 from Laboratories' Own Measurements

Lab./ Anal.	Meas. No.	NBS 946	NBS 947	NBS 948
		125/129 keV	125/129 keV	125/129 keV
1/1	1		0.0180 (2.0)	0.00438 (2.0)
	2		0.0192 (3.4)	0.00450 (2.2)
1/2	1	0.0166 (1.0)	0.0188 (2.0)	0.00425 (2.0)
	2		0.0194 (3.4)	0.00454 (2.2)
2		0.01580 (0.8)	0.01809 (0.8)	0.00397 (0.9)
3/1	1	0.01603 (1.0)	0.01839 (1.4)	0.00425 (1.5)
	2	0.01635 (1.8)	0.01875 (1.5)	0.00421 (1.4)
3/2	1	0.01637 (2.1)	0.01880 (2.1)	0.00446 (1.5)
	2	0.01692 (3.1)	0.01869 (1.6)	0.00442 (2.2)
3/3	1	0.01642 (1.5)	0.01889 (1.5)	0.00440 (2.0)
	2	0.01708 (1.5)	0.01901 (1.7)	0.00438 (2.4)
4		0.01618 (0.4)	0.01850 (0.4)	0.00417 (0.4)
5/1[b]	1	0.01581 (1.8)	0.01727 (2.8)	
5/1[c]	1			0.00427 (1.0)
5/2[b]	1	0.01595 (2.5)	0.01832 (1.5)	0.00413 (1.7)
5/2[c]	1	0.01677 (2.5)	0.01912 (1.9)	0.00420 (1.7)
5/2[b]	2	0.01578 (1.6)	0.01569 (3.7)	0.00407 (3.0)
6		0.0161[a] (0.9)	0.0182[a] (0.7)	0.00425[a] (1.1)
8		0.0178 (1.0)	0.02019 (1.0)	0.00417 (1.3)
	Mean	0.01640	0.01854	0.00428
	RSD(%)	3.19	5.24	3.71

[a] From 332-345 keV region. Values omitted from mean.

[b] 3^{rd} order polynomial log ε vs. log E for relative efficiency.

[c] 4^{th} order polynomial log ε vs. log E for relative efficiency.

Appendix D: Numerical Presentation of Peak Areas and Peak Area Ratios Determined from Distributed Spectra

Peak areas and peak area ratios of important single gamma rays or complexes of gamma rays in the energy range between 125 and 208 keV as determined by the participants from the distributed 'LEPS' and 'COAX' spectra are compiled in this Appendix. Unweighted grand mean values from all data, and unweighted reduced mean values from reduced data sets after exclusion of deviating results as indicated in the Tables have been calculated from the data.

Table D.1 Net Peak Areas from LEPS Spectra

Lab. Anal.	Gamma Energy (keV)							
	125[a]	129.3	148.6	152.7	160[b]	164.6	203.5	208.0
				NBS 946 LEPS Peak Area (Counts x 10^5)				
1/1	6.094	10.226	16.997	1.088	0.8878	3.856	0.5639	31.657
1/2	6.268	10.443	17.345	1.103	0.8679	3.893	0.5697	32.124
2	6.218	10.311	17.254	1.087	0.8710	3.810	0.5635	32.326
3/1	6.233	10.377	18.275[c]	1.090		3.937	0.5675	32.166
3/2	6.209	10.330	17.224	1.101	0.8784	3.899	0.5698	32.006
3/3	6.130	10.157	16.967	1.091	0.8670	3.860	0.5582	31.372
4	6.163	10.312	17.234	1.098	0.8614	3.898	0.5664	31.908
5/1	6.148	10.213	17.063	1.085	0.8885	3.844	0.5633	31.586
6	6.265	10.325	17.207	1.097	0.8638	3.890	0.5530	31.907
7/1		10.267	17.141	1.087	0.8645	3.843	0.5529	32.270
7/2		10.253	16.983	1.088	0.8803	3.847	0.5562	31.680
8	6.197	10.297	17.072	1.099	0.8914	3.879	0.5745	31.721
9		10.263	16.984	1.086	0.8847	3.857		31.811
Grand Mean	6.191	10.290	17.211	1.092	0.8756	3.870	0.5632	31.887
RSD(%)	0.89	0.72	1.99	0.58	1.24	0.85	1.23	0.89
Reduced Mean			17.123					
RSD(%)			0.75					
				NBS 947 LEPS Peak Area (Counts x 10^5)				
1/1	4.053	5.988	12.569	0.8444	0.7379	2.872	0.3362	23.837
1/2	4.158	6.110	12.836	0.8603	0.7161	2.923	0.3442	24.193
2	4.134	6.044	12.758	0.8535	0.7202	2.906	0.3765[c]	24.318
3/1	4.136	6.080	13.449[c]	0.8557		2.926	0.3344	24.219
3/2	4.119	6.035	12.684	0.8543	0.7311	2.904	0.3408	24.138
3/3	4.073	5.940	12.587	0.8450	0.7236	2.841	0.3324	23.628
4	4.089	6.025	12.744	0.8551	0.7162	2.902	0.3379	24.034
5/1	4.081	5.980	12.617	0.8428	0.7444	2.862	0.3350	23.779
6	4.156	6.047	12.720	0.8493	0.7202	2.899	0.3295	24.034
7/1		5.991	12.676	0.8486		2.855	0.3544[c]	24.334
7/2		6.001	12.542	0.8441	0.7107	2.890	0.3305	23.851
8	4.116	6.028	12.584	0.8581	0.7409	2.894	0.3474	23.917
9		6.005	12.539	0.8522	0.7286	2.875		23.966
Grand Mean	4.112	6.021	12.716	0.851	0.7264	2.888	0.3416	24.019
RSD(%)	0.88	0.74	1.88	0.68	1.53	0.89	3.86	0.90
Reduced Mean			12.655				0.3368	
RSD(%)			0.76				1.73	
				NBS 948 LEPS Peak Area (Counts x 10^5)				
1/1	7.890[c]	50.739	11.065	0.2794	1.627	2.872	4.467	28.230
1/2	8.280	51.685	11.311	0.2941	1.587	2.930	4.532	28.650
2	8.327	50.980	11.185	0.2866	1.618	2.904	4.511	28.668
3/1	8.399	51.243	11.241	0.3080		2.938	4.464	28.589
3/2	8.147	51.109	11.186	0.2867	1.633	2.912	4.526	28.560
3/3	8.164	50.078	11.204	0.2930	1.611	2.883	4.465	28.077
4	8.076	51.134	11.173	0.2935	1.587	2.914	4.482	28.440
5/1	8.100	50.744	11.257	0.2826	1.624	2.859	4.461	28.279
5/2	8.227	50.984	11.205	0.2877	1.624	2.894	4.498	28.478
6	8.599[c]	51.063	11.185	0.3051	1.617	2.906	4.474	28.444
7/1		50.834	11.086	0.2983	1.587	2.861		
7/2	8.095	50.702	11.971	0.2901	1.554[c]	2.886	4.461	28.218
8	8.239	50.884	11.100	0.3136[d]	1.645	2.887	4.467	28.298
9		50.858	11.014	0.2905	1.622	2.866		28.047
Grand Mean	8.209	50.931	11.156	0.2935	1.610	2.894	4.484	28.383
RSD(%)	2.23	0.70	0.86	3.33	1.54	0.86	0.58	0.73
Reduced Mean	8.205			0.2920	1.615			
RSD(%)	1.31			2.80	1.17			

[a] Σ (125.21 + 125.29) keV
[b] Σ (159.96 + 160.19 + 160.28) keV
[c] Omitted from reduced mean. Deviates more than 3 % from grand mean.
[d] Omitted from reduced mean. Deviates more than 5 % from grand mean.

Table D.2 Net Peak Area Ratios from LEPS Spectra

Lab./ Anal.	$125^a/129.3$	129.3/148.6	152.7/148.6	$160^b/148.6$	$160^b/164.6$	203.5/208.0
	Net Peak Area Ratio A_{E_i}/A_{E_k} (keV/keV)					
			NBS 946 LEPS			
			x 10^{-2}	x 10^{-2}		x 10^{-2}
1/1	0.5959	0.6016	6.401	5.223	0.2302	1.781
1/2	0.6002	0.6021	6.359	5.004	0.2229	1.773
2	0.6030	0.5976	6.300	5.048	0.2286	1.743
3/1	0.6007	0.5678[c]	5.964[c]			1.764
3/2	0.6011	0.5997	6.392	5.100	0.2253	1.780
3/3	0.6035	0.5986	6.430	5.110	0.2246	1.779
4	0.5977	0.5984	6.371	4.998	0.2210	1.775
5/1	0.6020	0.5985	6.359	5.207	0.2311	1.783
6	0.6068	0.6000	6.375	5.020	0.2221	1.733
7/1		0.5990	6.342	5.043	0.2250	1.713
7/2		0.6037	6.406	5.183	0.2288	1.756
8	0.6018	0.6032	6.437	5.221	0.2298	1.811
9		0.6043	6.394	5.209	0.2294	
Grand Mean	0.6013	0.5980	6.348	5.114	0.2265	1.766
RSD(%)	0.50	1.56	1.91	1.77	1.48	1.48
Reduced Mean		0.6006	6.381			
RSD(%)		0.39	0.60			
			NBS 947 LEPS			
			x 10^{-2}	x 10^{-2}		x 10^{-2}
1/1	0.6769	0.4764	6.718	5.871	0.2569	1.410
1/2	0.6805	0.4760	6.702	5.579	0.2450	1.423
2	0.6840	0.4737	6.690	5.645	0.2478	1.548[c]
3/1	0.6825	0.4521[c]	6.364[c]			1.381
3/2	0.6857	0.4758	6.735	5.764	0.2518	1.412
3/3	0.6787	0.4719	6.713	5.749	0.2547	1.407
4	0.6787	0.4728	6.710	5.620	0.2468	1.406
5/1	0.6824	0.4740	6.680	5.900	0.2601[c]	1.409
6	0.6873	0.4754	6.677	5.662	0.2484	1.371
7/1		0.4726	6.695			1.456[c]
7/2		0.4785	6.730	5.667	0.2459	1.386
8	0.6828	0.4790	6.819	5.888	0.2560	1.453
9		0.4789	6.796	5.811	0.2534	
Grand Mean	0.6820	0.4736	6.695	5.741	0.2515	1.422
RSD(%)	0.48	1.46	1.61	1.99	2.01	3.32
Reduced Mean		0.4754	6.722		0.2507	1.401
RSD(%)		0.52	0.65		1.76	1.22
			NBS 948 LEPS			
			x 10^{-2}			
1/1	0.1555[c]	4.586	2.525	0.1470	0.5665	0.1582
1/2	0.1602	4.569	2.600	0.1403	0.5416	0.1582
2	0.1633	4.558	2.562	0.1447	0.5572	0.1574
3/1	0.1639	4.559	2.740			0.1561
3/2	0.1594	4.569	2.563	0.1460	0.5608	0.1585
3/3	0.1630	4.470	2.615	0.1438	0.5588	0.1590
4	0.1579	4.577	2.627	0.1420	0.5446	0.1576
5/1	0.1596	4.508	2.510	0.1443	0.5680	0.1577
5/2	0.1614	4.550	2.568	0.1449	0.5612	0.1579
6	0.1684[c]	4.565	2.728	0.1446	0.5564	0.1573
7/1		4.585	2.691	0.1432	0.5547	
7/2	0.1597	4.621	2.644	0.1416	0.5385[c]	0.1581
8	0.1619	4.584	2.825[d]	0.1482	0.5698	0.1579
9		4.618	2.638	0.1473	0.5659	
Grand Mean	0.1612	4.566	2.631	0.1445	0.5572	0.1578
RSD(%)	2.05	0.89	3.39	1.61	1.82	0.46
Reduced Mean	0.1610		2.616		0.5604	
RSD(%)	1.23		2.76		1.29	

[a] Σ(125.21 + 125.29) keV
[b] Σ(159.96 + 160.19 + 160.28) keV
[c] Omitted from reduced mean. Deviates more than 3 % from grand mean.
[d] Omitted from reduced mean. Deviates more than 5 % from grand mean.

Table D.3 Net Peak Areas from COAX Spectra

Lab. Anal.	Gamma Energy (keV)							
	125[a]	129.3	148.6	152.7	160[b]	164.6	203.5	208.0
				NBS 946 COAX Peak Area (Counts x 10^6)				
1/1	2.738	6.088	14.162	0.8739	0.7933	3.720	0.6519	40.634
1/2	2.822	6.288	14.705	0.9030	0.8000	3.789	0.6741	41.534
2	2.779	6.162	14.472	0.8857	0.8058	3.775	0.6522	41.379
3/1	2.773	6.170	14.889	0.8826	0.7989	3.694	0.6444	41.198
3/2	2.708	6.027	14.139	0.8603	0.7911	3.690	0.6533	40.742
3/3	2.751	6.087	14.202	0.8725	0.7941	3.724	0.6512	40.521
4	2.702	6.112	14.466	0.8792	0.7792	3.749	0.6863[c]	41.158
5/1	2.736	6.072	14.228	0.8717	0.7903	3.726	0.6496	40.822
6	2.752	6.145	14.459	0.8813	0.8107	3.780	0.6426	40.991
7/1		6.134	14.205	0.8910	0.7827	3.755	0.6533	41.181
7/2		6.098	14.193	0.9261[c]	0.7382[c]	3.731	0.6590	40.732
8	2.751	6.120	14.297	0.8798	0.7986	3.732	0.6594	40.750
9	2.752	6.109	14.144	0.8871	0.8048	3.720	0.6361	40.787
Grand Mean	2.751	6.124	14.351	0.8842	0.7914	3.737	0.6549	40.956
RSD(%)	1.20	1.02	1.64	1.84	2.31	0.83	2.01	0.75
Reduced Mean				0.8807	0.7958		0.6523	
RSD(%)				1.23	1.17		1.46	
				NBS 947 COAX Peak Area (Counts x 10^6)				
1/1	2.896	5.823	17.136	1.115	1.080	4.540	0.6426	50.373
1/2	2.992	6.015	17.833	1.152	1.078	4.613	0.6370	51.497
2	2.956	5.879	17.537	1.149	1.086	4.615	0.6374	51.395
3/1		5.897	18.058[c]	1.132		4.522	0.6341	51.179
3/2	2.871	5.772	17.318	1.109	1.075	4.583	0.6436	50.616
3/3	2.910	5.843	17.171	1.120	1.071	4.543	0.6341	50.327
4	2.819[c]	5.845	17.523	1.122	1.056	4.577	0.6826[c]	51.087
5/1	2.903	5.798	17.234	1.118	1.076	4.548	0.6413	50.574
6	2.926	5.876	17.529	1.123	1.094	4.572	0.6321	50.902
7/1		5.897	17.199	1.135	1.051	4.586	0.6371	51.146
7/2		5.826	17.189	1.117	1.082	4.544	0.6559	50.545
8	2.926	5.853	17.304	1.122	1.093	4.556	0.6523	50.566
9	2.924	5.829	17.209	1.142	1.088	4.545	0.6336	50.641
Grand Mean	2.912	5.858	17.403	1.1274	1.0775	4.565	0.6434	50.834
RSD(%)	1.60	1.02	1.62	1.20	1.23	0.63	2.15	0.77
Reduced Mean	2.923		17.349				0.6401	
RSD(%)	1.20		1.22				1.18	
				NBS 948 COAX Peak Area (Counts x 10^6)				
1/1	7.337	46.438	10.559	0.2712	1.609	2.845	4.863	31.335
1/2	7.483	47.936	11.060	0.2935	1.587	2.943	4.968	32.013
2	7.676[c]	47.145	10.895	0.2866	1.647	2.929	4.985	31.888
3/1	7.536	47.158	10.394[c]	0.2944[d]		2.916	4.808	31.719
3/2	7.223	46.589	10.732	0.2811	1.611	2.867	4.872	31.429
3/3	7.221	46.403	10.626	0.2833	1.598	2.877	4.879	31.265
4	7.243	46.956	10.901	0.2749	1.580	2.872	4.920	31.670
5/1	7.274	46.679	10.671	0.2726	1.608	2.872	4.865	31.372
5/2	7.388	46.911	10.886	0.2854	1.630	2.901	4.916	31.694
6	7.485	46.928	10.866	0.2867	1.692[c]	2.882	4.865	31.586
7/1	7.400	46.893	10.525	0.2619[d]		2.817	4.901	31.568
7/2		46.679	10.665	0.2710	1.565	2.833	4.863	31.333
8	7.510	46.877	10.844	0.2855	1.568	2.890	4.891	31.412
9	7.380	46.823	10.598	0.2648[d]	1.607	2.883	4.859	31.513
Grand Mean	7.397	46.887	10.717	0.2795	1.609	2.881	4.890	31.556
RSD(%)	1.87	0.81	1.72	3.64	2.20	1.22	0.94	0.73
Reduced Mean	7.373		10.756	0.2811	1.601			
RSD(%)	1.55		1.52	2.68	1.56			

[a] Σ (125.21 + 125.29) keV

[b] Σ (159.96 + 160.19 + 160.28) keV

[c] Omitted from reduced mean. Deviates more than 3 % from grand mean.

[d] Omitted from reduced mean. Deviates more than 5 % from grand mean.

Table D.4 Net Peak Area Ratios from COAX Spectra

Lab./ Anal.	Net Peak Area Ratio A_{E_i}/A_{E_k} (keV/keV) 125[a]/129.3	129.3/148.6	152.7/148.6	160[b]/148.6	160[b]/164.6	203.5/208.0
			NBS 946 COAX			
			x 10^{-2}	x 10^{-2}		x 10^{-2}
1/1	0.4497	0.4299	6.171	5.602	0.2133	1.604
1/2	0.4488	0.4276	6.141	5.394	0.2110	1.623
2	0.4510	0.4258	6.120	5.568	0.2135	1.576
3/1	0.4494	0.4144	5.928[c]	5.366[c]	0.2163	1.564
3/2	0.4493	0.4263	6.085	5.595	0.2144	1.604
3/3	0.4519	0.4286	6.144	5.591	0.2132	1.607
4	0.4421	0.4225	6.078	5.386	0.2078	1.667
5/1	0.4506	0.4268	6.127	5.555	0.2121	1.591
6	0.4478	0.4250	6.095	5.607	0.2145	1.568
7/1		0.4318	6.272	5.510	0.2084	1.586
7/2		0.4296	6.525[c]	5.201[c]	0.1979[c]	1.618
8	0.4495	0.4281	6.154	5.586	0.2140	1.618
9	0.4505	0.4319	6.272	5.690	0.2163	1.560
Grand Mean	0.4491	0.4268	6.153	5.512	0.2117	1.599
RSD(%)	0.63	1.07	2.32	2.46	2.31	1.85
Reduced Mean			6.151	5.553	0.2129	
RSD(%)			1.08	1.65	1.27	
			NBS 947 COAX			
			x 10^{-2}	x 10^{-2}		x 10^{-2}
1/1	0.4973	0.3398	6.507	6.303	0.2379	1.276
1/2	0.4974	0.3373	6.460	6.045	0.2337	1.237
2	0.5028	0.3352	6.552	6.193	0.2353	1.240
3/1		0.3266	6.269[c]			1.239
3/2	0.4974	0.3333	6.404	6.207	0.2346	1.272
3/3	0.4980	0.3403	6.523	6.237	0.2358	1.260
4	0.4823[c]	0.3336	6.403	6.026	0.2307	1.336[c]
5/1	0.5007	0.3364	6.487	6.243	0.2366	1.268
6	0.4980	0.3452	6.407	6.241	0.2393	1.242
7/1		0.3429	6.599	6.111	0.2292	1.246
7/2		0.3389	6.498	6.295	0.2381	1.298
8	0.4999	0.3382	6.484	6.316	0.2399	1.290
9	0.5016	0.3387	6.636	6.322	0.2394	1.251
Grand Mean	0.4975	0.3366	6.479	6.212	0.2359	1.266
RSD(%)	1.15	1.21	1.47	1.64	1.45	2.29
Reduced Mean	0.4992		6.497			1.260
RSD(%)	0.42		1.14			1.65
			NBS 948 COAX			
			x 10^{-2}			
1/1	0.1580	4.398	2.568	0.1524	0.5656	0.1552
1/2	0.1561	4.334	2.165[d]	0.1435[c]	0.5395[c]	0.1552
2	0.1628[c]	4.327	2.631	0.1512	0.5623	0.1563
3/1	0.1598	4.537[c]	2.832[d]			0.1516[c]
3/2	0.1550	4.341	2.619	0.1501	0.5619	0.1550
3/3	0.1556	4.367	2.666	0.1504	0.5554	0.1552
4	0.1543	4.307	2.522	0.1449[c]	0.5501	0.1554
5/1	0.1558	4.374	2.555	0.1507	0.5599	0.1551
5/2	0.1575	4.309	2.622	0.1497	0.5619	0.1551
6	0.1595	4.319	2.639	0.1557[c]	0.5871[c]	0.1540
7/1		4.455	2.488			0.1553
7/2	0.1585	4.377	2.541	0.1467	0.5524	0.1552
8	0.1602	4.323	2.633	0.1446[c]	0.5426	0.1557
9	0.1576	4.418	2.499	0.1516	0.5574	0.1542
Grand Mean	0.1577	4.370	2.570	0.1493	0.5580	0.1549
RSD(%)	1.53	1.49	5.65	2.43	2.19	0.71
Reduced Mean	0.1573	4.358	2.582	0.1504	0.5570	0.1551
RSD(%)	1.25	1.04	2.34	1.14	1.25	0.37

[a] Σ (125.21 + 125.29) keV
[b] Σ (159.96 + 160.19 + 160.28) keV
[c] Omitted from reduced mean. Deviates more than 3 % from grand mean.
[d] Omitted from reduced mean. Deviates more than 5 % from grand mean.

Appendix E: Numerical Presentation of Relative Detection Efficiencies and Efficiency Ratios Determined from Distributed Spectra

Tables E.1 to E.6 present relative detection efficiencies for some gamma-ray energies in the range from 125 keV up to 451 keV, determined from the distributed 'LEPS and 'COAX' Spectra. The Tables include data from those participants who have reported functional relationships for the relative detection efficiency in specified energy ranges. The efficiency values have been normalized to a values of 1.00 for 129.29 keV. Unweighted grand mean values and the associated relative standard deviations of the efficiency values for each gamma-ray energy are given in the last two columns of the Tables.

Tables E.7 and E.8 summarize relative detection efficiency ratios for some important pairs of gamma energies in the energy range between 125 and 208 keV, determined from the 'LEPS' and 'COAX' spectra. Grand mean values and reduced mean values have been calculated in the same manner as for the peak areas and peak area ratios in Appendix D.

<u>Table E.1</u> Relative Detection Efficiency for Spectrum NBS 946 LEPS

Energy (keV)	Relative Detection Efficiency										Mean Value	RSD (%)
	1/1	1/2	3	4	5/1	5/2	7/1	7/2	8	9		
125.29	1.0239	1.0221	0.9968	0.9789	1.0095		1.0372	1.0246	1.0072	1.0041	1.0116	1.7
129.29	1.0000	1.0000	1.0000	1.0000	1.0000		1.0000	1.0000	1.0000	1.0000	1.0000	Normalized
148.57	0.8759	0.8800	0.9531	0.9494	0.9181		0.8407	0.8755	0.9383	0.9320	0.9070	4.4
152.68	0.8492	0.8535	0.9324	0.9226	0.8958		0.8109	0.8491	0.9201	0.9112	0.8828	4.9
159.96	0.8027	0.8069	0.8898	0.8714	0.8544		0.7612	0.8033	0.8843	0.8708	0.8383	5.4
160.28	0.8007	0.8049	0.8879	0.8692	0.8526		0.7591	0.8013	0.8827	0.8689	0.8363	5.5
164.58	0.7739	0.7781	0.8594	0.8385	0.8276		0.7317	0.7750	0.8597	0.8440	0.8098	5.7
203.54	0.5650	0.5667		0.6053	0.6108		0.5341	0.5707	0.6249	0.6211	0.5873	5.6
208.00	0.5451	0.5464		0.5844	0.5890		0.5163	0.5513	0.5980	0.5985	0.5661	5.4
255.38	0.3795	0.3790			0.4040		0.3697	0.3884		0.4061	0.3878	3.8
267.54	0.3480	0.3473			0.3689		0.3418	0.3571		0.3697	0.3555	3.3
332.35	0.2299	0.2298			0.2390		0.2346	0.2388		0.2379	0.2350	1.8
345.01	0.2139	0.2141			0.2221		0.2195	0.2226		0.2209	0.2189	1.8
375.04	0.1824	0.1832			0.1890		0.1892	0.1904		0.1884	0.1871	1.8
413.71	0.1517	0.1536			0.1578		0.1587	0.1589		0.1584	0.1565	2.0
451.47	0.1294	0.1322			0.1361		0.1357	0.1358		0.1379	0.1345	2.3

Table E.2 Relative Detection Efficiency for Spectrum NBS 947 LEPS

Energy (keV)	Relative Detection Efficiency										Mean Value	RSD (%)
	1/1	1/2	3	4	5/1	5/2	7/1	7/2	8	9		
125.29	1.0137	1.0105	0.9909	0.9856	1.0120		1.0181	1.0211	1.0024	1.0035	1.0064	1.2
129.29	1.0000	1.0000	1.0000	1.0000	1.0000		1.0000	1.0000	1.0000	1.0000	1.0000	Normalized
148.57	0.9049	0.9133	0.9679	0.9612	0.9155		0.8978	0.8886	0.9560	0.9369	0.9269	3.2
152.68	0.8813	0.8904	0.9482	0.9395	0.8941		0.8745	0.8642	0.9403	0.9173	0.9055	3.5
159.96	0.8388	0.8480	0.9070	0.8964	0.8552		0.8333	0.8215	0.9080	0.8793	0.8653	3.8
160.28	0.8368	0.8463	0.9049	0.8944	0.8533		0.8314	0.8196	0.9064	0.8775	0.8634	3.8
164.58	0.8116	0.8206	0.8771	0.8673	0.8298		0.8072	0.7948	0.8849	0.8540	0.8386	3.9
203.54	0.6020	0.6060		0.6347	0.6287		0.6093	0.5969	0.6508	0.6402	0.6211	3.2
208.00	0.5816	0.5848		0.6122	0.6081		0.5898	0.5776	0.6232	0.6180	0.5994	3.0
255.38	0.4093	0.4061			0.4305		0.4221	0.4134		0.4278	0.4182	2.4
267.54	0.3767	0.3724			0.3956		0.3891	0.3814		0.3912	0.3844	2.4
332.35	0.2568	0.2496			0.2620		0.2625	0.2581		0.2564	0.2576	1.8
345.01	0.2412	0.2336			0.2437		0.2450	0.2410		0.2388	0.2406	1.7
375.04	0.2106	0.2028			0.2075		0.2100	0.2068		0.2046	0.2071	1.5
413.71	0.1820	0.1742			0.1723		0.1755	0.1731		0.1726	0.1749	2.1
451.47	0.1622	0.1547			0.1466		0.1502	0.1481		0.1505	0.1520	3.7

Table E.3 Relative Detection Efficiency for Spectrum NBS 948 LEPS

Energy (keV)	Relative Detection Efficiency										Mean Value	RSD (%)
	1/1	1/2	3	4	5/1	5/2	7/1	7/2	8	9		
125.29	0.9622	0.9633	0.9415	0.9572	0.9615	0.9603	0.9596	0.9610	0.9719	0.9601	0.9599	0.8
129.29	1.0000	1.0000	1.0000	1.0000	1.0000	1.0000	1.0000	1.0000	1.0000	1.0000	1.0000	Normalized
148.57	1.0976	1.0925	1.1484	1.1258	1.1083	1.1115	1.1133	1.1099	1.1073	1.1104	1.1125	1.4
152.68	1.1024	1.0969	1.1567	1.1361	1.1162	1.1202	1.1219	1.1186	1.1218	1.1185	1.1209	1.5
159.96	1.1007	1.0940	1.1560	1.1423	1.1200	1.1252	1.1263	1.1232	1.1384	1.1215	1.1248	1.6
160.28	1.1003	1.0936	1.1555	1.1422	1.1197	1.1249	1.1257	1.1232	1.1388	1.1213	1.1245	1.6
164.58	1.0932	1.0859	1.1471	1.1390	1.1163	1.1221	1.1225	1.1202	1.1424	1.1171	1.1206	1.8
203.54	0.9285	0.9197		0.9797	0.9747	0.9834	0.9784	0.9821	0.9782	0.9679	0.9659	2.5
208.00	0.9044	0.8955		0.9546	0.9521	0.9608	0.9554	0.9598	0.9422	0.9448	0.9411	2.6
255.38	0.6639	0.6547			0.7144	0.7220	0.7148	0.7217		0.7034	0.6993	4.0
267.54	0.6121	0.6030			0.6607	0.6677	0.6607	0.6677		0.6499	0.6460	4.2
332.35	0.4111	0.4013			0.4436	0.4475	0.4439	0.4480		0.4371	0.4332	4.4
345.01	0.3840	0.3740			0.4131	0.4166	0.4138	0.4171		0.4079	0.4038	4.3
375.04	0.3316	0.3209			0.3528	0.3550	0.3545	0.3555		0.3502	0.3458	4.0
413.71	0.2831	0.2714			0.2949	0.2958	0.2980	0.2959		0.2959	0.2907	3.4
451.47	0.2505	0.2377			0.2539	0.2539	0.2585	0.2537		0.2583	0.2524	2.8

Table E.4 Relative Detection Efficiency for Spectrum NBS 946 COAX

Energy (keV)	Relative Detection Efficiency										Mean Value	RSD (%)
	1/1	1/2	3	4	5/1	5/2	7/1	7/2	8	9		
125.29	0.9620	0.9510	0.9119	0.8550	0.9502		0.9274	0.9986	0.9598	0.9450	0.9401	4.3
129.29	1.0000	1.0000	1.0000	1.0000	1.0000		1.0000	1.0000	1.0000	1.0000	1.0000	Normalized
148.57	1.1257	1.1627	1.2388	1.2253	1.1705		1.2305	1.0783	1.1726	1.1893	1.1771	4.4
152.68	1.1416	1.1824	1.2573	1.2304	1.1930		1.2543	1.1000	1.2018	1.2138	1.1972	4.3
159.96	1.1615	1.2067	1.2710	1.2359	1.2215		1.2813	1.1367	1.2442	1.2452	1.2227	3.9
160.28	1.1623	1.2077	1.2710	1.2361	1.2226		1.2830	1.1381	1.2458	1.2463	1.2237	3.9
164.58	1.1695	1.2156	1.2686	1.2400	1.2331		1.2898	1.1573	1.2642	1.2578	1.2329	3.7
203.54	1.1305	1.1538		1.2334	1.1929		1.1875	1.2037	1.1987	1.2102	1.1888	2.7
208.00	1.1185	1.1375		1.2162	1.1784		1.1659	1.1945	1.1685	1.1940	1.1717	2.7
255.38	0.9569	0.9355			0.9865		0.9485	1.0057		0.9858	0.9698	2.8
267.54	0.9135	0.8851			0.9361		0.9028	0.9486		0.9328	0.9198	2.6
332.35	0.7085	0.6674			0.7112		0.7061	0.6982		0.7031	0.6991	2.3
345.01	0.6753	0.6352			0.6769		0.6774	0.6628		0.6695	0.6662	2.4
375.04	0.6053	0.5709			0.6067		0.6146	0.5938		0.6020	0.5989	2.6
413.71	0.5319	0.5086			0.5367		0.5465	0.5281		0.5375	0.5316	2.4
451.47	0.4750	0.4654			0.4863		0.4875	0.4791		0.4939	0.4812	2.1

Table E.5 Relative Detection Efficiency for Spectrum NBS 947 COAX

Energy (keV)	Relative Detection Efficiency										Mean Value	RSD (%)
	1/1	1/2	3	4	5/1	5/2	7/1	7/2	8	9		
125.29	0.9625	0.9520	0.9066	0.8565	0.9472		0.9243	0.9424	0.9572	0.9408	0.9322	3.6
129.29	1.0000	1.0000	1.0000	1.0000	1.0000		1.0000	1.0000	1.0000	1.0000	1.0000	Normalized
148.57	1.1286	1.1587	1.2597	1.2595	1.1872		1.2381	1.2019	1.1846	1.2085	1.2030	3.7
152.68	1.1459	1.1776	1.2822	1.2695	1.2127		1.2620	1.2296	1.2162	1.2365	1.2258	3.6
159.96	1.1683	1.2003	1.3017	1.2781	1.2468		1.2876	1.2653	1.2625	1.2739	1.2538	3.4
160.28	1.1691	1.2013	1.3022	1.2783	1.2481		1.2883	1.2665	1.2643	1.2748	1.2548	3.4
164.58	1.1777	1.2084	1.3032	1.2812	1.2617		1.2940	1.2800	1.2849	1.2897	1.2645	3.4
203.54	1.1556	1.1437		1.2408	1.2381		1.1840	1.2467	1.2379	1.2556	1.2128	3.7
208.00	1.1452	1.1270		1.2226	1.2243		1.1613	1.2313	1.2099	1.2400	1.1952	3.7
255.38	0.9954	0.9282			1.0348		0.9571	1.0257		1.0305	0.9953	4.4
267.54	0.9532	0.8790			0.9839		0.9135	0.9717		0.9763	0.9463	4.3
332.35	0.7467	0.6683			0.7530		0.7386	0.7408		0.7416	0.7315	4.3
345.01	0.7122	0.6374			0.7174		0.7113	0.7067		0.7072	0.6987	4.3
375.04	0.6368	0.5762			0.6447		0.6556	0.6386		0.6390	0.6321	4.5
413.71	0.5594	0.5177			0.5721		0.5889	0.5738		0.5745	0.5644	4.4
451.47	0.4966	0.4782			0.5197		0.5286	0.5304		0.5318	0.5142	4.3

Table E.6 Relative Detection Efficiency for Spectrum NBS 948 COAX

Energy (keV)	Relative Detection Efficiency										Mean Value	RSD (%)
	1/1	1/2	3	4	5/1	5/2	7/1	7/2	8	9		
125.29	0.9373	0.9510	0.9130	0.8968	0.9531	0.9506	0.9585	0.9549	0.9667	0.9502	0.9432	2.3
129.29	1.0000	1.0000	1.0000	1.0000	1.0000	1.0000	1.0000	1.0000	1.0000	1.0000	1.0000	Normalized
148.57	1.1985	1.1589	1.2222	1.2222	1.1576	1.1667	1.1528	1.1544	1.1455	1.1662	1.1745	2.5
152.68	1.2198	1.1769	1.2362	1.2310	1.1771	1.1878	1.1743	1.1745	1.1709	1.1871	1.1936	2.1
159.96	1.2414	1.1975	1.2401	1.2319	1.2014	1.2139	1.2024	1.2005	1.2088	1.2127	1.2151	1.4
160.28	1.2417	1.1983	1.2401	1.2316	1.2022	1.2147	1.2026	1.2012	1.2102	1.2134	1.2156	1.4
164.58	1.2495	1.2036	1.2331	1.2269	1.2107	1.2236	1.2149	1.2110	1.2272	1.2221	1.2223	1.1
203.54	1.1136	1.1201		1.1609	1.1567	1.1698	1.1843	1.1730	1.1648	1.1637	1.1563	2.1
208.00	1.0886	1.1023		1.1520	1.1411	1.1538	1.1711	1.1592	1.1335	1.1472	1.1388	2.4
255.38	0.8280	0.8911			0.9467	0.9534	0.9708	0.9754		0.9453	0.9301	5.7
267.54	0.7719	0.8405			0.8969	0.9024	0.9178	0.9262		0.8945	0.8786	6.2
332.35	0.5643	0.6322			0.6770	0.6791	0.6801	0.6993		0.6741	0.6580	7.0
345.01	0.5393	0.6028			0.6436	0.6456	0.6459	0.6635		0.6415	0.6260	6.8
375.04	0.4949	0.5455			0.5758	0.5780	0.5782	0.5890		0.5758	0.5625	5.8
413.71	0.4640	0.4930			0.5086	0.5115	0.5175	0.5124		0.5120	0.5027	3.7
451.47	0.4569	0.4599			0.4601	0.4646	0.4830	0.4544		0.4676	0.4638	2.1

Table E.7 Relative Efficiency Ratios for LEPS Spectra

Lab./ Anal.	Relative Efficiency Ratio $\varepsilon_{E_i}/\varepsilon_{E_k}$ (keV/keV) 129.3/125.3	148.6/129.3	148.6/152.7	148.6/160.3	164.6/160.3	208.0/203.5
			NBS 946 - LEPS			
1/1	0.9767	0.8759	1.0315	1.0940	0.9666	0.9649
1/2	0.9784	0.8800	1.0310	1.0933	0.9666	0.9642
2	0.987	0.908	1.031	1.087	0.971	0.955
3	1.0032	0.9531	1.0222	1.0735	0.9679	0.963
4	1.0215	0.9494	1.0291	1.0925	0.9747	0.9655
5/1	0.9906	0.9181	1.0250	1.0769	0.9707	0.9643
6		0.9035	1.0247	1.0686	0.9771	0.9597
7/1	0.9641	0.8407[b]	1.0368	1.1076[a]	0.9639	0.9667
7/2	0.9760	0.8755	1.0312	1.0926	0.9672	0.9660
8	0.9929	0.9383	1.0198	1.0630	0.9740	0.9570
9	0.9959	0.9320	1.0228	1.0727	0.9714	0.9636
Grand Mean	0.9886	0.9068	1.0277	1.0838	0.9692	0.9627
RSD(%)	1.65	3.94	0.51	1.26	0.41	0.39
Reduced Mean		0.9134		1.0814		
RSD(%)		3.25		1.09		
			NBS 947 - LEPS			
1/1	0.9865	0.9049	1.0268	1.0813	0.9698	0.9660
1/2	0.9896	0.9133	1.0258	1.0792	0.9696	0.9650
2	0.997	0.951	1.022	1.062	0.979	0.984
3	1.0092	0.9679	1.0209	1.0697	0.9693	0.967
4	1.0146	0.9612	1.0230	1.0746	0.9697	0.9644
5/1	0.9881	0.9155	1.0240	1.0729	0.9725	0.9674
6		0.9599	1.0221	1.0612	0.9795	1.0223[a]
7/1	0.9822	0.8978	1.0267	1.0799	0.9709	0.9680
7/2	0.9793	0.8862[b]	1.0283	1.0842	0.9697	0.9678
8	0.9976	0.9560	1.0167	1.0546	0.9762	0.9576
9	0.9965	0.9369	1.0213	1.0677	0.9732	0.9653
Grand Mean	0.9941	0.9319	1.0234	1.0716	0.9727	0.9723
RSD(%)	1.14	3.13	0.32	0.89	0.40	1.82
Reduced Mean		0.9364				0.9673
RSD(%)		2.81				0.68
			NBS 948 - LEPS			
1/1	1.0393	1.0976	0.9956	0.9975	0.9935	0.9740
1/2	1.0381	1.0925	0.9960	0.9990	0.9930	0.9737
2	1.095	1.127	0.995	0.987	1.005	0.970
3	1.0621	1.1484	0.9928	0.9939	0.9927	0.965
4	1.0447	1.1258	0.9909	0.9856	0.9972	0.9743
5/1	1.0400	1.1083	0.9929	0.9898	0.9970	0.9768
5/2	1.0413	1.1115	0.9922	0.9881	0.9975	0.9770
6		1.1292	0.9952	0.9871	1.0046	0.9738
7/1	1.0421	1.1133	0.9923	0.9890	0.9972	0.9765
7/2	1.0405	1.1099	0.9922	0.9882	0.9973	0.9772
8	1.0289	1.1073	0.9871	0.9723	1.0032	0.9632
9	1.0415	1.1104	0.9928	0.9903	0.9963	0.9761
Grand Mean	1.0467	1.1151	0.9929	0.9890	0.9979	0.9731
RSD(%)	1.70	1.36	0.24	0.68	0.42	0.48

[a] Omitted from reduced mean. Deviates more than 2 % from grand mean.
[b] Omitted from reduced mean. Deviates more than 5 % from grand mean.

Table E.8 Relative Efficiency Ratios for COAX Spectra

Lab./ Anal.	Relative Efficiency Ratio $\varepsilon_{E_i}/\varepsilon_{E_k}$ (keV/keV)					
	129.3/125.3	148.6/129.3	148.6/152.7	148.6/160.3	164.6/160.3	208.0/203.5
			NBS 946 - COAX			
1/1	1.03959	1.1257	0.9861	0.9685	1.0062	0.9894
1/2	1.0515	1.1627	0.9833	0.9627	1.0065	0.9859
2	1.052	1.163	0.983	0.957	1.015	0.985
3	1.0967	1.2388[b]	0.9853	0.9747	0.9981	0.984
4	1.1697[b]	1.2253	0.9959	0.9913[a]	1.0032	0.9861
5/1	1.0524	1.1705	0.9811	0.9574	1.0086	0.9878
6		1.1867	0.9835	0.9557	1.0160	0.9924
7/1	1.0782	1.2305	0.9810	0.9591	1.0053	0.9818
7/2	1.0014[b]	1.0783[b]	0.9803	0.9475	1.0169	0.9924
8	1.0419	1.1726	0.9757	0.9412	1.0148	0.9748
9	1.0582	1.1893	0.9798	0.9543	1.0092	0.9866
Grand Mean	1.0642	1.1767	0.9832	0.9609	1.0091	0.9860
RSD(%)	4.19	3.99	0.52	1.42	0.59	0.51
Reduced Mean	1.0588	1.1807		0.9578		
RSD(%)	1.82	2.75		1.00		
			NBS 947 - COAX			
1/1	1.0390	1.1286[b]	0.9849	0.9654	1.0074	0.9910
1/2	1.0504	1.1587	0.9840	0.9645	1.0059	0.9854
2	1.058	1.181	0.981	0.950	1.018	0.986
3	1.1030	1.2597	0.9825	0.9674	1.0008	0.987
4	1.1675[b]	1.2595	0.9921	0.9853[a]	1.0023	0.9853
5/1	1.0557	1.1872	0.9790	0.9512	1.0109	0.9889
6		1.1910	0.9836	0.9561	1.0158	0.9935
7/1	1.0189	1.2381	0.9811	0.9610	1.0044	0.9808
7/2	1.0611	1.2019	0.9775	0.9490	1.0107	0.9876
8	1.0447	1.1846	0.9740	0.9370	1.0163	0.9774
9	1.0629	1.2085	0.9774	0.9480	1.0117	0.9876
Grand Mean	1.066	1.1999	0.9816	0.9577	1.0095	0.9864
RSD(%)	3.9o	3.36	0.49	1.35	0.57	0.45
Reduced Mean	1.0549	1.2070		0.9550		
RSD(%)	2.15	2.86		1.01		
			NBS 948 - COAX			
1/1	1.0669	1.1985	0.9825	0.9652	1.0063	0.9776
1/2	1.0516	1.1589	0.9847	0.9671	1.0044	0.9841
2	1.023	1.176	0.986	0.963	0.971[a]	0.967
3	1.0953	1.2222	0.9887	0.9856[a]	0.9944	0.981
4	1.1151[b]	1.2222	0.9929	0.9924[a]	0.9962	0.9923
5/1	1.0492	1.1576	0.9834	0.9629	1.0071	0.9865
5/2	1.0519	1.1667	0.9822	0.9605	1.0073	0.9863
6		1.1782	0.9899	0.9727	1.0097	0.9808
7/1	1.0434	1.1528	0.9817	0.9586	1.0102	0.9889
7/2	1.0473	1.1544	0.9829	0.9610	1.0082	0.9882
8	1.0344	1.1455	0.9783	0.9465	1.0140	0.9731
9	1.0524	1.1662	0.9824	0.9611	1.0072	0.9858
Grand Mean	1.0573	1.1749	0.9846	0.9664	1.0030	0.9826
RSD(%)	2.51	2.22	0.42	1.27	1.15	0.73
Reduced Mean	1.0515			0.9619	1.0059	
RSD(%)	1.83			0.70	0.58	

[a] Omitted from reduced mean. Deviates more than 2 % from grand mean.
[b] Omitted from reduced mean. Deviates more than 5 % from grand mean.