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# **The AS-76 Interlaboratory Experiment on the Alpha Spectrometric Determination of Pu-238**

**Part I:  
Performance and Results**

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Projekt Spaltstoffflußkontrolle

**Kernforschungszentrum Karlsruhe**



KERNFORSCHUNGSZENTRUM KARLSRUHE

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## Abstract

In cooperation with 26 laboratories of 11 countries or international organizations, the Safeguards Project of the Karlsruhe Nuclear Research Center carried out the interlaboratory program AS-76. It focused on the alpha-spectrometric determination of the Pu-238 isotope. The performance of the program as well as the results obtained are described.

## Zusammenfassung

Das Interlaborexperiment AS-76 zur alphaspektrometrischen Bestimmung von Pu-238

Teil I: Durchführung und Ergebnisse

In Zusammenarbeit mit 26 Laboratorien aus 11 Ländern oder internationalen Organisationen führte das Projekt Spaltstoffflußkontrolle des Kernforschungszentrums Karlsruhe das Interlaboratoriumsprogramm AS-76 durch, das sich mit der alphaspektrometrischen Bestimmung des Isotops Pu-238 befaßte. Die Programmdurchführung sowie die gewonnenen Ergebnisse werden beschrieben.

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## Preface

Since the mass spectrometric determination of the Pu-238 isotope might be complicated by interference with the abundant U-238 isotope, alpha-spectrometry is preferred by many laboratories for this application. Therefore, for practical safeguards it is of interest to determine the error sources essentially contributing to this analytical technique as well as to estimate the uncertainty involved in this type of measurement.

For this reason, the Safeguards Project of the Karlsruhe Nuclear Research Center initiated in 1976 the AS-76 analytical intercomparison program on the alpha-spectrometric measurement of the Pu-238 isotope. The purpose of this program was to study the capability of the complete procedure, starting with 'unknown' sample material and reaching a final result for the alpha-activity ratio  $\text{Pu-238}/(\text{Pu-239} + \text{Pu-240})$ .

The sample material used originated from the GWK reprocessing plant, Karlsruhe; the samples were prepared by the JRC-CBNM, Geel, Belgium, which also characterized the sample composition in cooperation with the AERE Harwell, U.K., and the NBS, Washington, U.S.A. . Inclusive of the institutions above, a total of 26 laboratories of 11 countries or international organizations participated.

The data were evaluated by statistical methods and compiled in three 'preliminary reports'. On February 13 to 15, 1979 the results were discussed in detail at a meeting of the participants at the Karlsruhe Research Center; the conclusions arrived at provided the basis of this final report and have been compiled in Section 8.

Dipak Gupta

Head of the Nuclear Safeguards Project

Acknowledgement

In this report the results have been compiled which were obtained by the laboratories named below. The authors like to acknowledge the fruitful cooperation with these laboratories; they also like to thank Dr. Dipak Gupta, Head of the Nuclear Safeguards Project, for his interest in this study. The valuable discussions with Priv.-Doz. Dr. Rudolf Avenhaus are appreciated.

In data evaluation and in the preparation of this report the authors were assisted by Anton Bajer, Axel Berghoff, Veronika Glienecke, Jürgen Henkenhaf, Rudolf Hulata, Ruth Klausmann-Stern, and Edith Wortmann, to whom they wish to express their gratitude.

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## Introduction

Already with the JEX 70 /1/ and the IDA-72 /2/ interlaboratory comparison programs some alpha-spectrometric results for Pu-238 determinations were obtained, and in 1974 an alpha-spectrum evaluation test (ASET-74) /3/ was carried out. The objective of this exercise was to determine the scattering of results, caused by application of different evaluation methods to identical spectra, and to investigate the influence of spectrum resolution on the Pu-238/(Pu-239 + Pu-240) alpha-activity ratio obtained. This influence was found to be of major importance. But, besides resolution, this test indicated further possible error components affecting the alpha-activity ratio. The AS-76 program was initiated to study these possible error components.

The participating laboratories were requested to use their routine procedures for sample preparation and measurement. In addition, they were asked to keep for later their records as well as their original alpha-spectra recordings in order to allow a more detailed investigation of this experiment, possibly induced by the results obtained in the first steps of evaluation.

For description of this experiment and its results the report had to be divided into three parts. Part I is mainly the compilation and evaluation of the reported data including the study of the overall capability of the analytical method investigated. Part II contains graphs of representative alpha-spectra measured by the participating laboratories and the results obtained through application of one common method of evaluation. Part III provides information on the preparation of the samples and on their characterization.

## 1. Experimental

To study the error components affecting the alpha-activity ratio of Pu-238/(Pu-239 + Pu-240) - consequently affecting the interesting Pu-238 determination - the main steps of the analytical procedure as well as their error contributions have to be identified.

With respect to the nuclear fuel cycle, the Pu-238 isotope has to be determined in materials comprising either relatively pure plutonium or quite a number of additional elements. For example, the plutonium product of a reprocessing plant might be relatively pure but Am-241 will be generated through beta-decay of Pu-241 ( $T_{1/2} = 14.89$  a); Am-241 is an alpha-emitter making some contribution in the range of energy where also Pu-238 must be determined. This Am-241 has to be handled by a suitable method, depending on the time elapsed since the 'production' of the plutonium as well as on the procedures preferred by the laboratories. This correction might introduce some error sources.

On the other hand, the input solution of a reprocessing plant will contain fission products comprising numerous alpha-emitters; therefore, a straightforward alpha-spectrometry of Pu-238 is not possible; chemical separation must precede in any case. This chemical step might contribute some error component, for instance, if purification was incomplete.

Figure 1.1 presents schematically a typical alpha-spectrum of plutonium in the energy range between 4.26 and 5.79 MeV. Obviously, the (Pu-239 + Pu-240) alpha-peak is influenced by the low-energy tail of the Pu-238 alpha-peak. Consequently, the separation of these two alpha-peaks might contribute some error too.

Furthermore, for alpha-counting usually several sources are prepared and each source is counted several times. Therefore, also estimates of the reproducibilities associated with these two steps of the procedure are of interest.

Based on these considerations it was decided to use sample solutions of different fission-product contents and of varying isotopic compositions and to propose a structure of the analytical procedure to be followed in the way presented in Fig. 1.2:

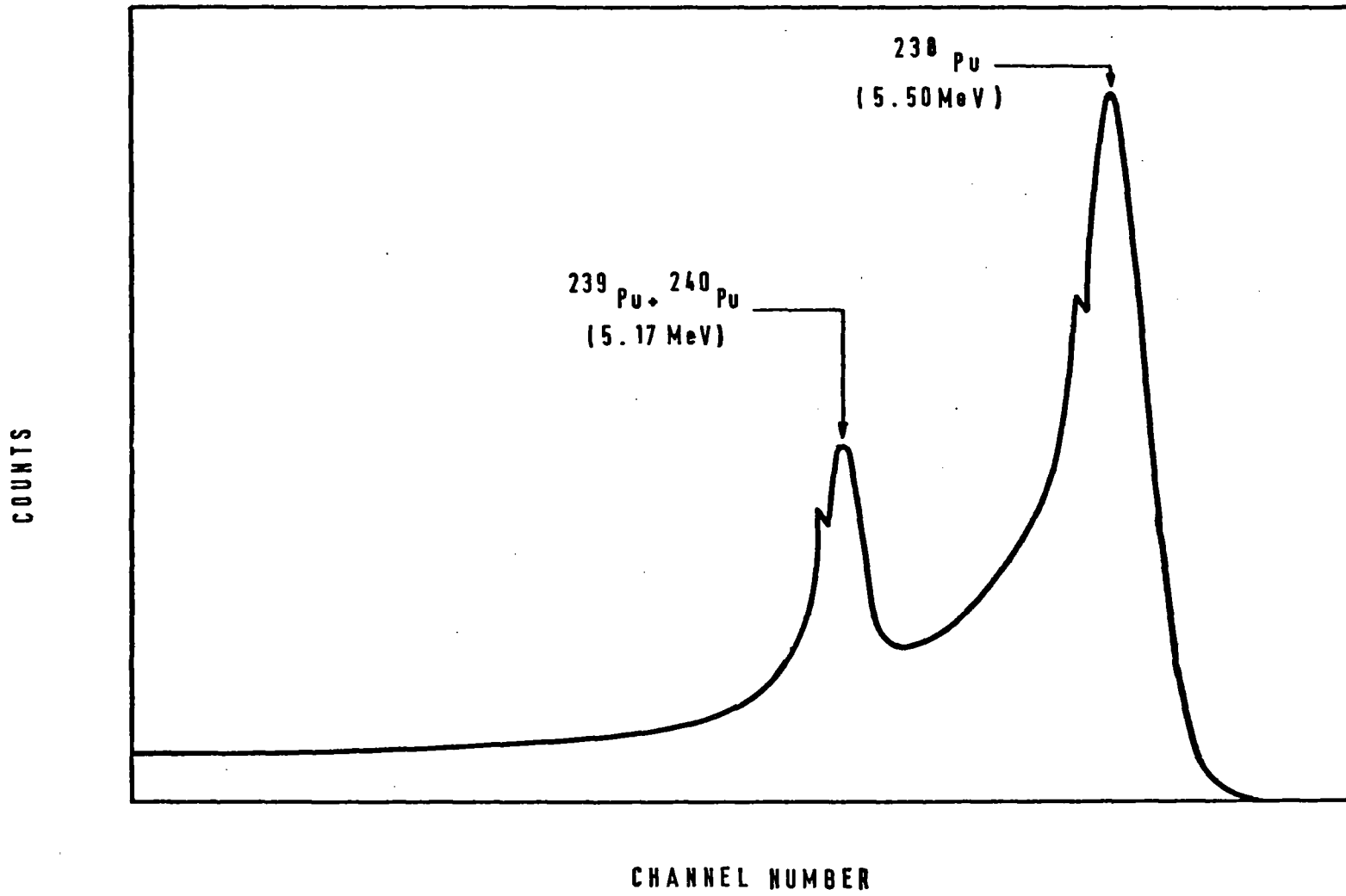
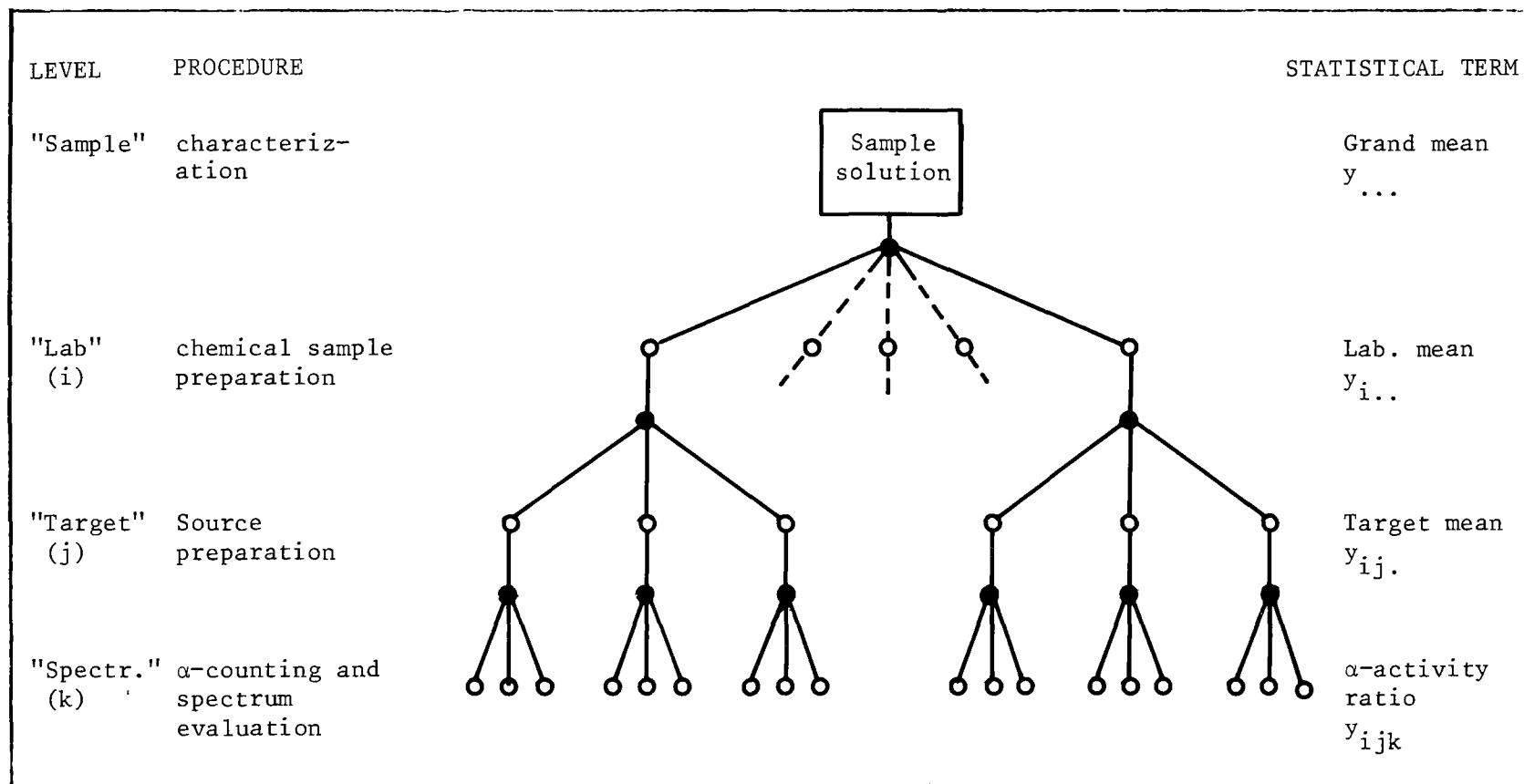


Fig. 1.1: Plutonium alpha-particle spectrum (schematic).



**Fig. 1.2:** Layout of the experimental design and structure of the statistical evaluation procedure applied (scheme per sample).

Each laboratory was asked to purify the total amount of each sample solution obtained in one single chemical procedure (which may consist of several repetitions), to prepare then three alpha sources, and to count each of them three times, which resulted in nine spectra per sample and laboratory.

Applying three-stage variance analysis this experimental design allows to study the interlaboratory deviation as well as the error contributions from the preparation of the alpha-source (called 'target error') and the random errors from alpha counting and spectrum evaluation (called 'spectrum error').

The interlaboratory deviation will be affected by errors resulting from the chemical purification step as well as by any systematic error contribution from spectrum evaluation. However, it seemed possible to distinguish between the contributions of these two error sources by applying one common procedure of evaluation to the spectra of all laboratories <sup>1</sup>).

Fig. 1.2 also shows the structure of the statistical procedure of evaluation applied with the symbols explained below:

For each sample solution investigated

$y_{ijk}$  describes the measurement of the alpha-activity ratio Pu-238/(Pu-239 + Pu-240) reported by laboratory  $i$  on target  $j$  for spectrum  $k$

with

$i = 1 \dots 23$  (laboratories)  
 $j = 1 \dots 3$  (targets)  
 $k = 1 \dots 3$  (spectra)

---

<sup>1</sup>) Separation of these two error components by appropriate experimental design requires an additional branching point. For the laboratories this would have been implied a considerable increase of the workload.

$$y_{ij.} = \frac{1}{3} \sum_{k=1}^3 y_{ijk}$$

mean value related to target j  
of laboratory i ('target mean') (1-1)

$$y_{i..} = \frac{1}{3} \sum_{j=1}^3 y_{ij.}$$

mean value related to  
laboratory i ('laboratory mean') (1-2)

$$y_{...} = \frac{1}{23} \sum_{i=1}^{23} y_{i..}$$

'grand mean' of all measurement  
results obtained in this experiment. (1-3)

2. Sample Materials Used

The detailed information on sample preparation as well as on its characterization is the subject of Part III of this report.

To the participating laboratories three sample materials (called A, B and C) containing fission products and differing with respect to the Pu-238 content, and one fission-product free sample material (called D) were distributed.

The original material used for samples A to C was drawn from the accountability tank of the reprocessing plant operated by the Gesellschaft für Wiederaufarbeitung von Kernbrennstoffen mbH (GWK); for sample D, product material obtained in this installation was used.

These original sample materials were handled further by the Central Bureau for Nuclear Measurements (JRC-CBNM), Geel. For the sample material: A and C the desired isotopic compositions were obtained by adding suitable amounts of the isotopes Pu-239 and Pu-240 (sample A) and Pu-238 (sample C). The approximate Pu-238 abundances and further related data of the AS-76 samples are given in Tab. 2.1.

1	2	3	4	5
SAMPLE	NOMINAL Pu-238 CONTENT (%)	NOMINAL Pu-238/ (Pu-239 + Pu-240) $\alpha$ -ACTIVITY RATIO	FISSION PRODUCTS CONTAINED	ISOTOPEES ADDED
A	0.2	0.4	YES	Pu-239, Pu-240
B	0.8	1.4	YES	NONE
C	1.6	3.0	YES	Pu-238
D	0.9	1.6	NO	NONE

Tab. 2.1: AS-76 sample materials.

The four sample materials were characterized by the alpha-spectrometry laboratories at JRC-CBNM, Geel and AERE, Harwell and, in addition, by the mass spectrometry laboratories at JRC-CBNM, Geel, and NBS, Washington.

Tab. 2.2 presents the certified values CV <sup>1)</sup> of the alpha-activity ratio of Pu-238/(Pu-239 + Pu-240) as well as the isotopic composition <sup>2)</sup> for the sample materials B, C and D <sup>3)</sup>. Nov. 1, 1977 was chosen as the date of reference.

Sample	$\alpha$ -Activity Ratio	Isotopic Abundances [atom %]				
	(Pu-238)	Pu-238	Pu-239	Pu-240	Pu-241	Pu-242
	(Pu-239 + Pu-240)					
( $\alpha$ -Spectrometry)	(Mass Spectrometry)					
B	1.446	0.80	65.37	23.55	7.42	2.86
C	2.985	1.65	63.59	24.04	7.60	3.12
D	1.637	0.91	63.46	24.47	7.83	3.33

**Tab. 2.2:** Certified values CV of the alpha-activity ratio of Pu-238/(Pu-239 + Pu-240) and isotopic composition of the AS-76 sample materials B, C and D (reference date Nov. 1, 1977).

For the certified values of alpha-activity ratios the accuracy ( $3\sigma$ ) stated is  $\pm 0.3\%$ .

For safeguards purposes, the interesting quantity is the atom ratio of Pu-238/Pu-239 which can be derived from the alpha-spectrometric ratio Pu-238/(Pu-239 + Pu-240) using the equation

<sup>1)</sup> Mean values derived from the  $\alpha$ -spectrometric results obtained at JRC-CBNM, Geel, and AERE, Harwell.

<sup>2)</sup> Mean values from NBS, Washington, and JRC-CBNM, Geel, mass spectrometric measurements.

<sup>3)</sup> Due to inhomogeneous Pu-isotopic composition for the sample material A no characterization values were communicated.



$$R(238/239) = \alpha(238/(239 + 240)) \left[ \frac{T_{1/2}(238)}{T_{1/2}(239)} + R(240/239) \frac{T_{1/2}(238)}{T_{1/2}(240)} \right]$$

where e.g.  $R(238/239)$  stands for the atomic ratio of Pu-238/Pu-239. Using  $T_{1/2}(\text{Pu-238}) = 87.74$  a,  $T_{1/2}(\text{Pu-239}) = 24\ 110$  a, and  $T_{1/2}(\text{Pu-240}) = 6\ 553$  a /4/, for the sample materials B, C and D, Pu-238/Pu-239 atomic ratios were derived which, within the accuracies stated, were in agreement with the isotopic ratios measured by mass spectrometry.

### 3. Analytical Procedures Applied

Besides the alpha-activity ratio of Pu-238/(Pu-239 + Pu-240), the participating laboratories also reported information on the chemical purification step, on the spectrum evaluation methods, and on records related to alpha counting. It follows from this information that a wide variety of procedures have been used. For illustration as well as to facilitate the understanding of the reported results, a brief survey is given in Tab. 3.1. The information compiled is based on the descriptions given by the laboratories; a classification, especially of spectra evaluation methods, has been performed together with JRC-CBNM, Geel. In the specification of this survey care is taken that the codes are not disclosed.

The following explanations are related to the information compiled:

Column 2: IE and SE stand for ion exchange and solvent extraction, respectively.

Column 3: Materials for substrates were stainless steel (Fe) or other materials (O.M.) such as Au, Au-plated copper, glass, Ni-plated brass, Pt and Ta.

Column 4: ED and DE stand for electro-deposition and direct evaporation, respectively.

Column 5: Approximate figures are given for the solid angle in percent of  $4\pi$ ; they are calculated from the detector area and the target to detector distance, assuming a point source.

A dash stands in case the information necessary for this estimation was not available or could not be evaluated for any other reason.

Column 6: The figures given were obtained through averaging all values reported by a laboratory (total number of counts and counting times for all four samples). Together with the information in columns 4 and 5, this conveys an idea about the source strength.

1	2	3	4	5	6	7	8
Lab. Code	Separation Procedure	Substrate	Target Preparation	Counting Geometry (%)	Counting Rate (cpm)	Classification of Spectra Evaluation Method	Remarks about column 7
1	IE	O.M.	ED	-	1 700	STC	Similar to ASET-74, lab. 10.
2	IE	O.M.	DE	8.0	1 100	STC	Tail at the low energy end of the spectrum is decomposed according to $\alpha$ -ratio, and Pu-238 tail is interpolated.
3	SE	Fe	DE	2.8	1 200	STC	Similar to ASET-74, lab. 10.
4	IE	Fe	DE	8.3	1 600	NTC	
5	IE	O.M.	ED	1.3	7 000	IF	Iterative method; sloping tail is fitted to double exponential; constant tail of Pu-238 is obtained as by lab. 2.
6	IE	Fe	DE	-	1 200	EA	Similar to ASET-74, lab. 5.
7	SE	O.M.	ED	0.5-2.0	2 500	NTC	
8	IE	Fe	DE	3.1	24 000	ETF	Similar to ASET-74, lab. 3.
9	SE	Fe	ED	1.6	30	(NTC)	
10	IE	O.M.	ED	-	13 500	NTC	
11	SE	Fe	DE	24	300	NTC	
12	SE	O.M.	DE	3.0	70	STC	Similar to ASET-74, lab. 10.
13	SE	O.M.	DE	11	26 000	(ETF)	Graphically estimated tail correction; similar to ASET-74, lab. 9.
14	IE	Fe	DE	1.6	2 900	EA	Similar to ASET-74, lab. 8.
15	IE	Fe	ED	-	40 000	EA/IF	Iterative procedure according to empirical formula.
16	SE	Fe	DE	0.3	690	NTC	
17	IE	O.M.	ED	-	840	NTC	
18	SE	Fe	ED	16	31 200	EA	Similar to ASET-74, lab. 8.
19	SE	O.M.	ED	0.2	2 100	ETF/IF	Two methods used: a) similar to ASET-74, lab. 3, but with linear extrapolation towards lower energies; b) empirical algorithm to construct Pu-238 tail by iteration.
20	SE	Fe	DE	1.3	235	NTC	
21	IE	Fe	DE	-	11 200	ETF	Similar to ASET-74, lab. 3.
22	SE	O.M.	ED	-	11 200	(NTC)	
23	IE	Fe	DE	0.9	10 400	IF	Iterative fitting to construct Pu-238 tail; similar to ASET-74, lab. 7.

Tab. 3.1: Survey of methods used.

Column 7: This is an approach to classify the evaluation methods reported, where

NTC 'No tail correction';

STC 'Simple tail correction'.

Linear background (in a linear plot) under each peak such that the corrected channel content is zero at the beginning and end of the peak intervals;

ETF 'Extrapolated tail fit'.

Part of Pu-238 tail above (Pu-239 + Pu-240) peak is fitted with exponential. This exponential is extrapolated to lower energies;

EA 'Empirical algorithm'

IF 'Iterating fit'.

Brackets indicate that the classification is uncertain.

Tabs. 3.2 to 3.5 give some information on the data reported in connection with alpha counting.

Column 2: The average value of the counts per spectrum calculated for each laboratory by

$$N_i = \frac{1}{9} \sum_{\ell=1}^9 n_{i\ell} \quad (3-1)$$

where  $n_{i\ell}$  means the number of counts reported by laboratory  $i$  for the 9 spectra obtained for a given sample.

Column 3: The average counting times (minutes) per spectrum calculated for each laboratory by

$$CT_i = \frac{1}{9} \sum_{\ell=1}^9 ct_{i\ell} \quad (3-2)$$

where  $ct_{i\ell}$  means the counting time reported by laboratory  $i$  for the 9 spectra obtained for a given sample.

Column 4: The average counting rates  $N_i/CT_i$  (counts per minute) calculated with the values given in columns 2 and 3.

Column 5: The time lags (days) between chemical separation and the counting of the 9 spectra; this survey provides only the minimum ( $TL_i^{MIN}$ ) and maximum ( $TL_i^{MAX}$ ) time lags as reported by the laboratories for each sample.

*****						
1	2	3	4	5		
*****						
LABORATORY CODE	AVERAGE NUMBER OF COUNTS PER SPECTRUM	AVERAGE COUNTING TIME PER SPECTRUM	AVERAGE COUNTING RATE	TIME LAG BETWEEN CHEMICAL SEPARATION AND COUNTING		
i	$N_i$	$CT_i$	$N_i/CT_i$	$TL_i^{MIN}$	$TL_i^{MAX}$	
		(MIN)	(CPM)	(DAYS)		
*****						
1	162200	85	1917	6	8	
2	64765	60	1076	2	9	
3	27009	33	810	1	28	
4	200000	244	821	6	25	
5	364222	39	9367	1	2	
6	90877	83	971	2	6	
7	115149	54	2144	1	2	
8	357601	15	23840	1	1	
9	2514	100	25	0	2	
10	346106	30	11537	2	7	
11	16741	61	273	8	24	
12	18477	283	65	4	7	
13	123870	5	24774	0	0	
14	53535	30	1784	42	42	
15	237000	8	28440	11	11	
16	12548	31	411	10	10	
17	17966	15	1198	8	8	
18	71516	14	5108	12	12	
19	2000000	2010	995	2	23	
20	1727	10	173	1	1	
21	70765	5	14153	0	1	
22	24512	3	4085	2	2	
23	1177444	333	3532	0	3	
*****						

Tab. 3.2: Data reported by the laboratories in connection with the alpha-spectrometric measurement of sample A.

*****					
1	2	3	4	5	
*****					
LABORATORY CODE	AVERAGE NUMBER OF COUNTS PER SPECTRUM	AVERAGE COUNTING TIME PER SPECTRUM	AVERAGE COUNTING RATE	TIME LAG BETWEEN CHEMICAL SEPARATION AND COUNTING	
i	$N_i$	$CT_i$ (MIN)	$N_i/CT_i$ (CPM)	$TL_i^{MIN}$	$TL_i^{MAX}$
*****					
1	15444C	69	2240	3	14
2	71794	6C	1197	3	9
3	4251C	33	1275	27	28
4	196667	163	1203	5	33
5	371061	50	7421	2	2
6	95914	83	1151	9	13
7	93466	52	1803	1	2
8	381227	15	25415	1	1
9	3422	10C	34	1	6
10	397687	30	13256	4	7
11	16135	6C	269	8	25
12	15560	263	59	1	7
13	118033	5	23607	1	1
14	4960C	30	1653	44	44
15	454556	8	54547	10	10
16	21231	3C	708	8	8
17	9536	15	636	4	4
18	22704C	3	68112	10	10
19	25000C	72C	3472	14	45
20	1771	10	177	0	7
21	58421	5	11684	0	0
22	28524	3	4754	3	3
23	98880C	5C	17776	1	3
*****					

Tab. 3.3: Data reported by the laboratories in connection with the alpha-spectrometric measurement of sample B.

*****					
1	2	3	4	5	
*****					
LABORATORY CODE	AVERAGE NUMBER OF COUNTS PER SPECTRUM	AVERAGE COUNTING TIME PER SPECTRUM	AVERAGE COUNTING RATE	TIME LAG BETWEEN CHEMICAL SEPARATION AND COUNTING	
i	$N_i$	$CT_i$ (MIN)	$N_i/CT_i$ (CPM)	$TL_i^{MIN}$	$TL_i^{MAX}$
*****					
				(DAYS)	
1	157778	112	1407	3	7
2	63077	58	1081	7	11
3	56579	33	1697	29	30
4	200000	117	1713	6	14
5	310324	44	6981	3	3
6	74675	83	896	7	9
7	146028	33	4381	2	4
8	262492	15	17499	2	2
9	3700	100	37	5	7
10	338404	30	11280	4	14
11	15875	65	246	7	30
12	37228	395	94	4	11
13	147776	5	29555	1	1
14	146215	30	4874	43	43
15	273778	8	32853	10	10
16	23488	33	712	10	10
17	5942	15	663	1	1
18	309104	13	23183	12	12
19	2000000	1050	1905	3	6
20	2260	10	226	0	5
21	83904	5	16781	0	1
22	31296	3	5216	2	2
23	923989	50	18480	0	4
*****					

Tab. 3.4: Data reported by the laboratories in connection with the alpha-spectrometric measurement of sample C.



*****					
1	2	3	4	5	
*****					
LABORATORY CODE	AVERAGE NUMBER OF COUNTS PER SPECTRUM	AVERAGE COUNTING TIME PER SPECTRUM	AVERAGE COUNTING RATE	TIME LAG BETWEEN CHEMICAL SEPARATION AND COUNTING	
i	$N_i$	$CT_i$	$N_i/CT_i$	$TL_i^{MIN}$	$TL_i^{MAX}$
*****					
		(MIN)	(CPM)	(DAYS)	
*****					
1	54444	48	1145	6	8
2	55058	60	918	8	48
3	40236	33	1207	30	33
4	200000	74	2700	7	14
5	350325	83	4204	3	4
6	149570	83	1800	3	5
7	67336	44	1515	1	2
8	457127	15	30680	2	2
9	2683	100	27	1	2
10	541809	30	18060	4	14
11	24083	60	401	7	30
12	15740	274	57	5	10
13	130812	5	26162	1	1
14	103578	30	3453	41	41
15	370111	8	44413	10	10
16	27835	30	917	9	11
17	13335	15	889	3	3
18	147445	5	28264	13	13
19	2000000	900	2222	1	75
20	3643	10	364	1	2
21	52691	27	2303	2	2
22	52400	3	8733	1	1
23	555111	333	1695	1	7
*****					

Tab. 3.5: Data reported by the laboratories in connection with the alpha-spectrometric measurement of sample D.

#### 4. Reported Data and their Evaluation

The alpha-activity ratios of Pu-238/(Pu-239 + Pu-240) reported by the participating laboratories were corrected for Pu-238 decay, taking Nov. 1, 1977 as the reference date. The detailed information is compiled in Appendix A, while the corrected data are presented in Figs. 4.1 to 4.4. In these figures the values belonging to the three repetition spectra taken for each target are plotted vertically and are connected by a line.

This presentation gives a first survey of the spectra reproducibility per target, the deviation between targets, and the distribution of the laboratory related data sets.

Further evaluation of laboratory related data was based on the corrected alpha-activity ratios, using statistical procedures as already published /5/.<sup>1)</sup>

Table 4.1 comprises the target mean values calculated from the corrected alpha-activity ratios by use of Eq. (1-1).

In Tabs. 4.2 to 4.5 the following information is compiled:

Column 2: The mean value  $y_{i..}$  of the laboratory  $i$  as defined by (1-2).

Column 3: An estimate of the standard deviation of the error component 'spectrum'  $S_i^s$  defined by

$$S_i^s = \sqrt{\frac{1}{3(3-1)} \sum_{j=1}^3 \sum_{k=1}^3 (y_{ijk} - y_{ij.})^2} . \quad (4-1)$$

This value characterizes the reproducibility of spectra recording, including possible random errors from the evaluation, obtained on an average by laboratory  $i$ .

---

<sup>1)</sup> Although the inhomogeneity of sample A ruled out an exact characterization, this inhomogeneity was small enough to allow meaningful evaluation of the routine measurements discussed here.

ALPHA-ACTIVITY RATIO PU238/(PU239+PU240)

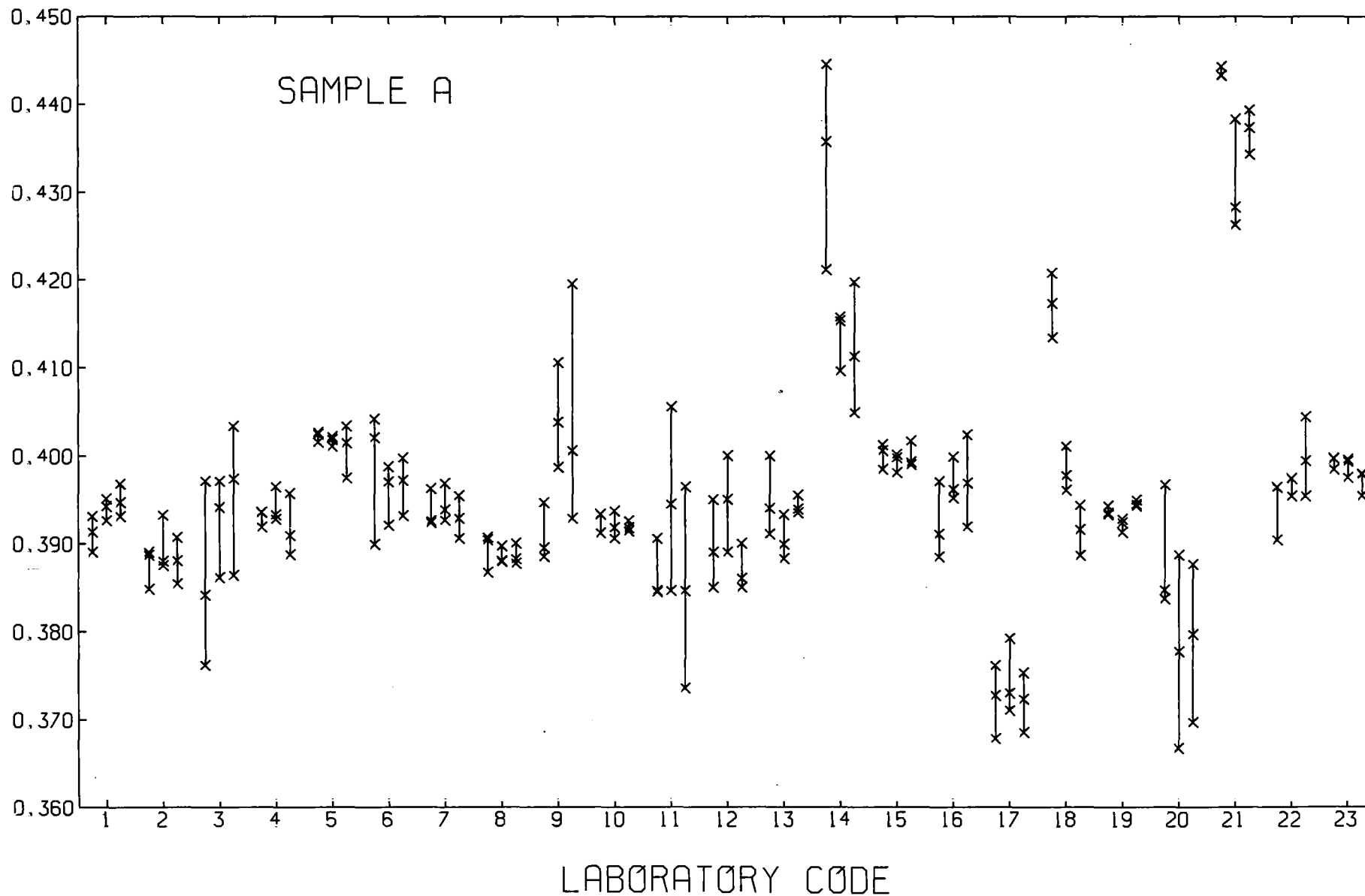


Fig. 4.1: Reported alpha-activity ratios of Pu-238/(Pu-239 + Pu-240) after correction with respect to reference date November 1, 1977 for sample A; Pu-238 abundance about 0.2 % (each division marked on the ordinate indicates an average 2.5 % change of the alpha-activity ratio).

ALPHA-ACTIVITY RATIO PU238/(PU239+PU240)

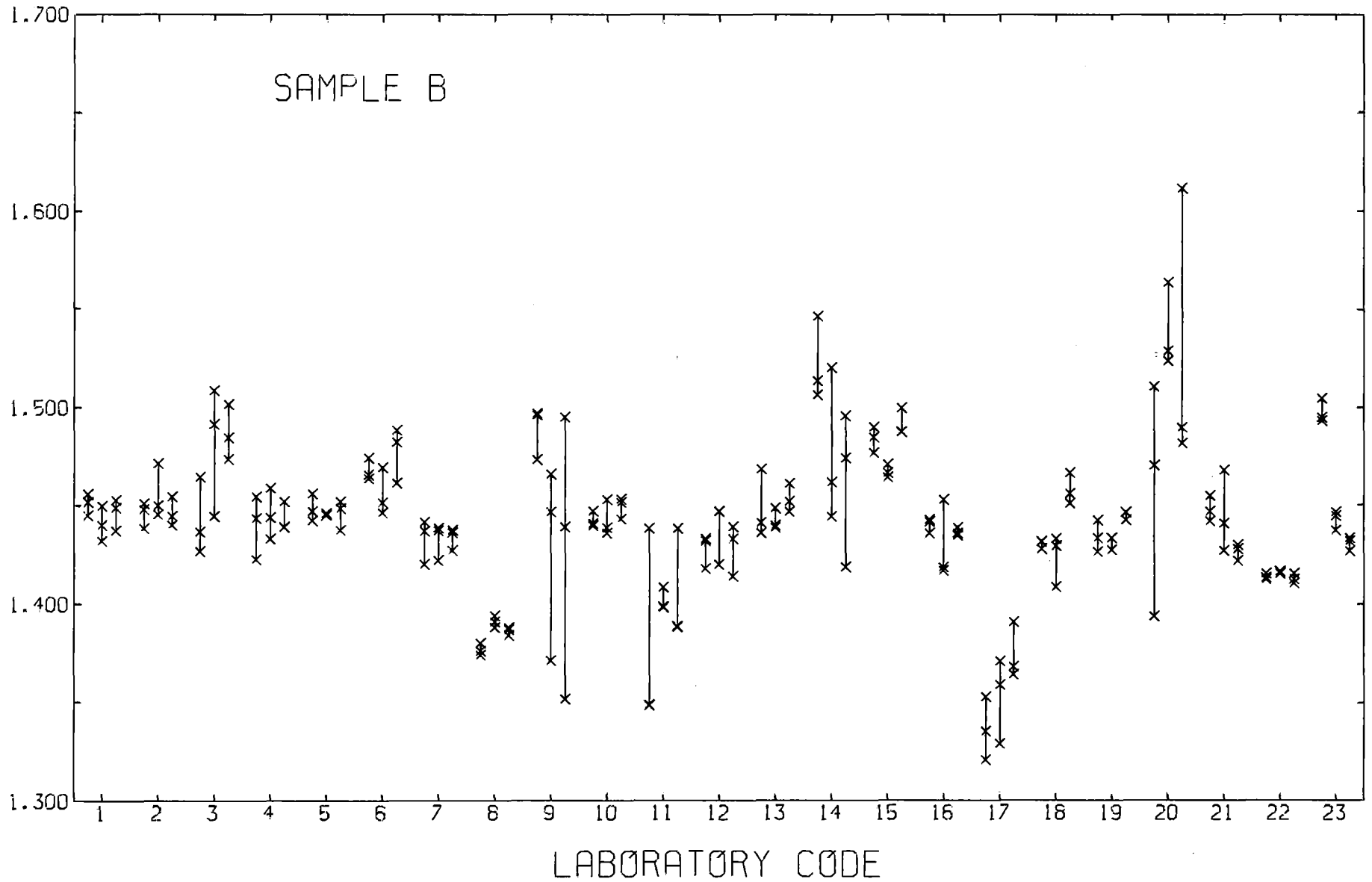


Fig. 4.2: Reported alpha-activity ratios of Pu-238/(Pu-239 + Pu-240) after correction with respect to reference date November 1, 1977 for sample B; Pu-238 abundance about 0.8 % (each division marked on the ordinate indicates an average 3.5 % change of the alpha-activity ratio).

ALPHA-ACTIVITY RATIO PU238/(PU239+PU240)

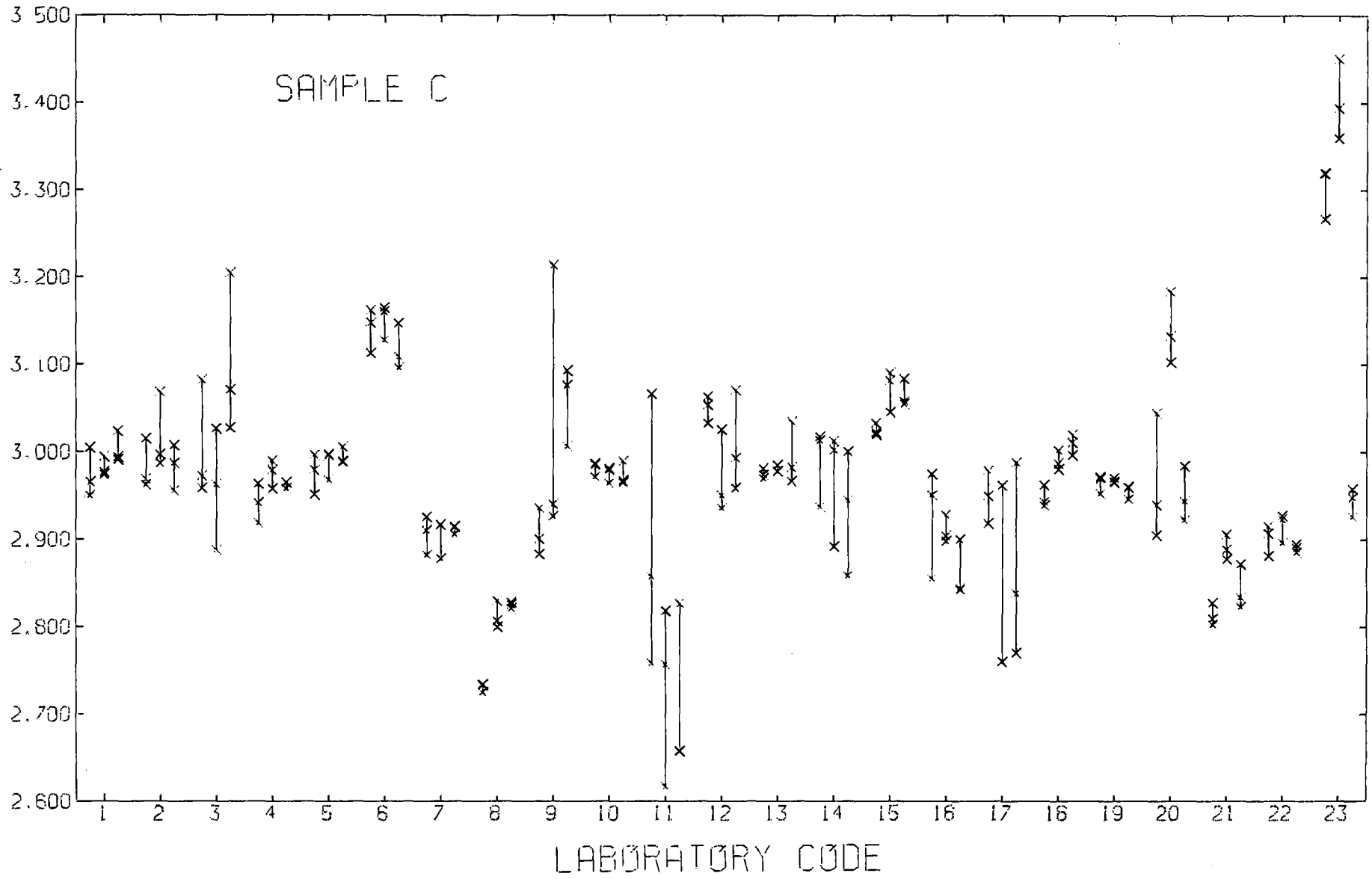


Fig. 4.3: Reported alpha-activity ratios of Pu-238/(Pu-239 + Pu-240) after correction with respect to reference date November 1, 1977 for sample C; Pu-238 abundance about 1.6 % (each division marked on the ordinate indicates an average 3.3 % change of the alpha-activity ratio).

ALPHA-ACTIVITY RATIO PU238/(PU239+PU240)

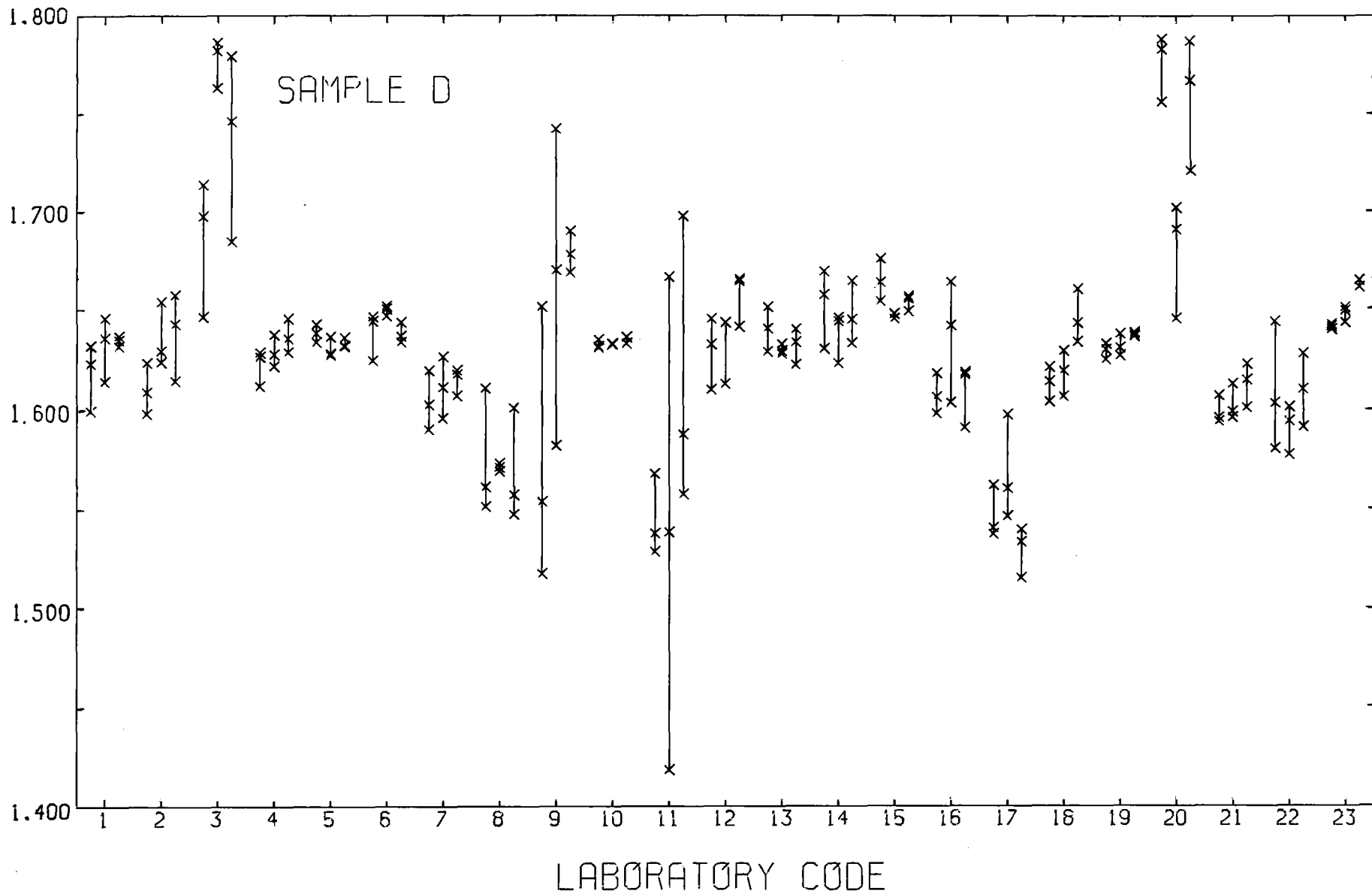


Fig. 4.4: Reported alpha-activity ratios of Pu-238/(Pu-239 + Pu-240) after correction with respect to reference date November 1, 1977 for sample D; Pu-238 abundance about 0.9 % (each division marked on the ordinate indicates an average 3.1 % change of the alpha-activity ratio).

1	2	3	4	5	6	7	8	9	10	11	12	13
LABORATORY	TARGET 1	TARGET 2	TARGET 3	TARGET 1	TARGET 2	TARGET 3	TARGET 1	TARGET 2	TARGET 3	TARGET 1	TARGET 2	TARGET 3
CODE	SAMPLE A			SAMPLE B			SAMPLE C			SAMPLE D		
I	YI(I,1,*)	YI(I,2,*)	YI(I,3,*)	YI(I,1,*)	YI(I,2,*)	YI(I,3,*)	YI(I,1,*)	YI(I,2,*)	YI(I,3,*)	YI(I,1,*)	YI(I,2,*)	YI(I,3,*)
1	0.39118	0.39401	0.39450	1.45109	1.44077	1.44650	2.97358	2.98192	3.00288	1.61882	1.63281	1.63545
2	0.38754	0.38962	0.38809	1.44592	1.45579	1.44658	2.98230	3.01751	2.98347	1.61077	1.63668	1.63916
3	0.38586	0.39251	0.39573	1.44281	1.48175	1.48678	3.00451	2.95897	3.10143	1.68688	1.77742	1.73726
4	0.39305	0.39425	0.39184	1.44049	1.44545	1.44355	2.94127	2.97567	2.96275	1.62327	1.63007	1.63783
5	0.40226	0.40173	0.40081	1.44863	1.44580	1.44620	2.97555	2.98696	2.95452	1.63935	1.63163	1.63436
6	0.39877	0.39601	0.39675	1.46820	1.45604	1.47756	3.14120	3.15114	3.11728	1.63950	1.65100	1.63528
7	0.39379	0.39452	0.39303	1.43303	1.43273	1.43377	2.90580	2.90365	2.91117	1.60478	1.61201	1.61560
8	0.38934	0.38861	0.38871	1.37680	1.39110	1.38645	2.73034	2.81185	2.82416	1.57506	1.57173	1.56507
9	0.39094	0.40440	0.40437	1.48899	1.42810	1.42870	2.90626	3.02672	3.05880	1.57492	1.66563	1.68017
10	0.39264	0.39208	0.39198	1.44275	1.44275	1.44957	2.98104	2.97445	2.97397	1.63358	1.63391	1.63622
11	0.38660	0.39492	0.38493	1.37859	1.40190	1.40524	2.89364	2.73060	2.71393	1.54514	1.54178	1.61507
12	0.38973	0.39473	0.38706	1.42788	1.43822	1.42888	3.04984	2.97075	3.00735	1.63039	1.63435	1.65828
13	0.39508	0.39055	0.39437	1.44881	1.44269	1.45364	2.97504	2.98031	2.99450	1.64145	1.63111	1.63316
14	0.43386	0.41360	0.41203	1.52220	1.47570	1.46307	2.98947	2.96504	2.93451	1.65359	1.63889	1.64878
15	0.40014	0.39938	0.40001	1.48416	1.46753	1.49179	3.02519	3.07267	3.06564	1.66577	1.64815	1.65498
16	0.39224	0.39711	0.39707	1.44030	1.42967	1.43663	2.92706	2.91006	2.86206	1.60830	1.63740	1.60983
17	0.37222	0.37442	0.37206	1.33627	1.35304	1.37434	2.94904	2.82735	2.86556	1.54697	1.56856	1.52970
18	0.41719	0.39831	0.39155	1.43058	1.42389	1.45613	2.94793	2.98599	3.00930	1.61391	1.61923	1.64681
19	0.39372	0.39216	0.39459	1.43429	1.43159	1.44532	2.96442	2.96742	2.95542	1.63022	1.63277	1.63850
20	0.38838	0.37772	0.37900	1.45837	1.53863	1.52797	2.96315	3.13982	2.95015	1.77605	1.68014	1.75677
21	0.44369	0.43101	0.43703	1.44819	1.44552	1.42684	2.81316	2.89061	2.84258	1.59952	1.60319	1.61354
22	0.39444	0.39678	0.39978	1.41392	1.41626	1.41292	2.90126	2.91561	2.88992	1.60981	1.59146	1.61048
23	0.39930	0.39887	0.39715	1.49764	1.44318	1.43096	3.30228	3.40154	2.94417	1.64228	1.64912	1.66469

Tab. 4.1: Target mean values calculated from the corrected alpha-activity ratios by use of Eq. (1-1).

LAB CODE	LAB MEAN	ESTIMATE FOR SD OF SPECTRUM	ESTIMATE FOR RSD OF SPECTRUM	ESTIMATE FOR SD OF TARGET	ESTIMATE FOR RSD OF TARGET	ESTIMATE FOR RSD OF SPECTRUM PLUS TARGET	ESTIMATE FOR RSD OF LABORATORY MEAN	DEVIATION FROM GRAND MEAN	DEVIATION FROM CHARACTERIZATION VALUE
i	$y_{i..}$	$s_i^s$	$RS_i^s$ (%)	$s_i^t$	$RS_i^t$ (%)	$RS_i^{s,t}$ (%)	$RSM_i^{s,t}$ (%)	$\Delta GM_i$ (%)	$\Delta CV_i$ (%)
1	0.3933630	0.0017555	0.45	0.0016596	0.42	0.61	0.29	-0.17 (-0.70)	
2	0.3884178	0.0027410	0.71	0.0000000	0.30	0.71	0.24	-1.43 (-1.94)	
3	0.3913651	0.0085278	2.18	0.0010636	0.27	2.20	0.74	-0.68 (-1.20)	
4	0.3930467	0.0024344	0.62	0.0000000	0.00	0.62	0.21	-0.25 (-0.78)	
5	0.4016021	0.0017989	0.45	0.0000000	0.00	0.45	0.15	1.92 (1.38)	
6	0.3971751	0.0052328	1.32	0.0000000	0.00	1.32	0.44	0.80 (0.27)	
7	0.3937788	0.0022671	0.59	0.0000000	0.00	0.59	0.19	-0.07 (-0.59)	
8	0.3888845	0.0015725	0.40	0.0000000	0.00	0.40	0.13	-1.31 (-1.88)	
9	0.3999017	0.0088498	2.21	0.0088404	1.46	2.65	1.12	1.49 (0.96)	
10	0.3922358	0.0012071	0.31	0.0000000	0.00	0.31	0.10	-0.46 (-0.58)	
11	0.3888197	0.0091779	2.36	0.0007531	0.19	2.37	0.79	-1.32 (-1.84)	
12	0.3905058	0.0045645	1.17	0.0028674	0.73	1.38	0.56	-0.90 (-1.42)	
13	0.3933321	0.0030669	0.78	0.0016751	0.43	0.89	0.36	-0.18 (-0.70)	
14	0.4198297	0.0083052	1.98	0.0111938	2.67	3.32	1.67	6.55 (5.99)	
15	0.3998423	0.0013603	0.34	0.0000000	0.00	0.34	0.11	1.47 (0.94)	
16	0.3954747	0.0042099	1.06	0.0013907	0.35	1.12	0.41	0.37 (-0.16)	
17	0.3729016	0.0039711	1.06	0.0000000	0.00	1.06	0.35	-5.36 (-5.85)	
18	0.4023504	0.0030650	0.76	0.0131707	3.27	3.36	1.91	2.11 (1.57)	
19	0.3934882	0.0005802	0.15	0.0011875	0.30	0.34	0.18	-0.14 (-0.66)	
20	0.3816992	0.0092063	2.41	0.0023729	0.62	2.49	0.83	-3.13 (-3.64)	
21	0.4372410	0.0040032	0.92	0.0059050	1.35	1.63	0.84	10.96 (10.33)	
22	0.3970016	0.0033537	0.84	0.0018479	0.47	0.96	0.29	0.75 (0.22)	
23	0.3984416	0.0011360	0.29	0.0009317	0.23	0.37	0.17	1.12 (0.59)	

Evaluation not possible due to missing characterization value

Tab. 4.2: Compilation of laboratory related results of evaluation for sample A



1	2	3	4	5	6	7	8	9	10
LAB	LAB	ESTIMATE FOR	ESTIMATE FOR	ESTIMATE FOR	ESTIMATE FOR	ESTIMATE FOR	ESTIMATE FOR	DEVIATION	DEVIATION FROM
CODE	MEAN	SD OF	RSD CF	SD OF	RSD CF	RSD OF	RSD CF	FROM	CHARACTERIZATION
		SPECTRUM	SPECTRUM	TARGET	TARGET	SPECTRUM PLUS	LABORATORY	GRAND MEAN	VALUE
						TARGET	MEAN		
i	$Y_{i..}$	$S_i^s$	$RS_i^s$	$S_i^t$	$RS_i^t$	$RS_i^{s,t}$	$RSM_i^{s,t}$	$\Delta GM_i$	$\Delta CV_i$
		(%)	(%)		(%)	(%)	(%)	(%)	(%)
1	1.4461174	0.0076779	0.53	0.0026559	0.18	0.56	0.21	0.29	0.01
2	1.4494276	0.0098726	0.68	0.0000000	0.00	0.68	0.23	0.52	0.24
3	1.4704447	0.0236704	1.61	0.0199080	1.35	2.10	0.54	1.98	1.69
4	1.4431629	0.0127682	0.88	0.0000000	0.00	0.88	0.29	0.09	-0.20
5	1.4468765	0.0060034	0.41	0.0000000	0.00	0.41	0.14	0.35	0.06
6	1.4672642	0.0111837	0.76	0.0086453	0.59	0.96	0.42	1.76	1.47
7	1.4331779	0.0090059	0.63	0.0000000	0.00	0.63	0.21	-0.60	-0.89
8	1.3847828	0.0027429	0.20	0.0071213	0.51	0.55	0.30	-3.96	-4.23
9	1.4485960	0.0514423	3.55	0.0184808	1.28	3.77	1.39	0.47	0.18
10	1.4450188	0.0065955	0.46	0.0010067	0.07	0.46	0.16	0.22	-0.07
11	1.3952408	0.0344704	2.47	0.0000000	0.00	2.47	0.82	-3.23	-3.51
12	1.4316597	0.0127389	0.89	0.0000000	0.00	0.89	0.30	-0.71	-0.93
13	1.4483786	0.0113543	0.79	0.0000000	0.00	0.79	0.26	0.45	0.16
14	1.4869881	0.0346596	2.33	0.0238589	1.60	2.83	1.21	3.13	2.43
15	1.4811583	0.0058683	0.40	0.0119306	0.81	0.90	0.48	2.72	2.13
16	1.4355326	0.0120218	0.84	0.0000000	0.00	0.84	0.28	-0.44	-0.72
17	1.3545465	0.0175911	1.30	0.0161538	1.19	1.76	0.81	-6.06	-6.32
18	1.4375334	0.0089934	0.63	0.0173912	1.21	1.36	0.73	-0.30	-0.59
19	1.4370661	0.0052167	0.36	0.0066224	0.46	0.59	0.29	-0.33	-0.62
20	1.5082912	0.0556908	3.69	0.0293984	1.95	4.18	1.67	4.61	1.31
21	1.4401817	0.0128514	0.89	0.0099598	0.62	1.09	0.47	-0.12	-0.10
22	1.4143667	0.0017342	0.12	0.0013898	0.10	0.16	0.07	-1.61	-0.19
23	1.4572582	0.0049634	0.34	0.0353820	2.43	2.45	1.41	1.07	0.78

Tab. 4.3: Compilation of laboratory related results of evaluation for sample B

*****									
1	2	3	4	5	6	7	8	9	10
*****									
LAB	LAB	ESTIMATE FOR	ESTIMATE FOR	ESTIMATE FOR	ESTIMATE FOR	ESTIMATE FOR	ESTIMATE FOR	DEVIATION	DEVIATION FROM
CODE	MEAN	SD OF	RSD OF	SD OF	RSD OF	RSD OF	RSD CF	FROM	CHARACTERIZATION
		SPECTRUM	SPECTRUM	TARGET	TARGET	SPECTRUM PLUS	LABORATORY	GRAND MEAN	VALUE
						TARGET	MEAN		
i	$Y_{i..}$	$S_i^S$	$RS_i^S$	$S_i^t$	$RS_i^t$	$RS_i^{S,t}$	$RSM_i^{S,t}$	$\Delta GM_i$	$\Delta CV_i$
			(%)		(%)	(%)	(%)	(%)	(%)
*****									
1	2.9861221	0.0204276	0.68	0.0094265	0.32	0.75	0.29	0.62	0.04
2	2.9944258	0.0340711	1.14	0.0036077	0.12	1.14	0.39	0.90	0.32
3	3.0216351	0.0774492	2.56	0.0573942	1.90	3.19	1.39	1.82	1.23
4	2.9598942	0.0161735	0.55	0.0146552	0.50	0.74	0.34	-0.26	-0.84
5	2.9856739	0.0171473	0.57	0.0000000	0.00	0.57	0.19	0.61	0.02
6	3.1365385	0.0242261	0.77	0.0103576	0.33	0.84	0.32	5.69	5.08
7	2.9068727	0.0184393	0.63	0.0000000	0.00	0.63	0.21	-2.05	-2.62
8	2.7887802	0.0096933	0.35	0.0506763	1.82	1.85	1.06	-6.03	-6.57
9	2.9972582	0.0586364	3.29	0.0567862	1.89	3.80	1.55	1.00	0.41
10	2.9764881	0.0106329	0.36	0.0000000	0.00	0.36	0.12	0.30	-0.29
11	2.7753865	0.1223142	4.40	0.0698009	2.51	5.07	2.06	-6.34	-6.89
12	3.0093231	0.0441870	1.47	0.0302654	1.01	1.78	0.76	1.41	0.81
13	2.9832830	0.0210647	0.71	0.0000000	0.00	0.71	0.24	0.53	-0.06
14	2.9643412	0.0624503	2.11	0.0000000	0.00	2.11	0.70	-0.11	-0.69
15	3.0544977	0.0169944	0.56	0.0236713	0.77	0.95	0.48	2.93	2.33
16	2.8957269	0.0423014	1.46	0.0232361	0.80	1.66	0.67	-2.29	-2.86
17	2.8806477	0.0947872	3.29	0.0256331	1.03	3.45	1.25	-2.93	-3.50
18	2.9824018	0.0118970	0.40	0.0306208	1.03	1.10	0.61	0.50	-0.09
19	2.9624205	0.0077757	0.26	0.0043391	0.15	0.30	0.12	-0.17	-0.76
20	3.0177050	0.0515170	1.71	0.1016958	3.37	3.78	2.03	1.69	1.10
21	2.8487816	0.0185202	0.65	0.0376023	1.32	1.47	0.79	-4.00	-4.56
22	2.9022636	0.0143998	0.50	0.0098338	0.34	0.60	0.26	-2.20	-2.77
23	3.2159958	0.0331881	1.03	0.2398158	7.46	7.53	4.32	8.37	7.74
*****									

Tab. 4.4: Compilation of laboratory related results of evaluation for sample C

LAB CODE	LAB MEAN	ESTIMATE FOR SD OF SPECTRUM	ESTIMATE FOR RSD OF SPECTRUM	ESTIMATE FOR SD OF TARGET	ESTIMATE FOR RSD OF TARGET	ESTIMATE FOR RSD OF SPECTRUM PLUS TARGET	ESTIMATE FOR RSD OF LABORATORY MEAN	DEVIATION FROM GRAND MEAN	DEVIATION FROM CHARACTERIZATION VALUE
i	$y_{i..}$	$S_i^s$	$RS_i^s$ (%)	$S_i^t$	$RS_i^t$ (%)	$RS_i^{s,t}$ (%)	$RSM_i^{s,t}$ (%)	$\Delta GM_i$ (%)	$\Delta CV_i$ (%)
1	1.6290245	0.0137020	0.84	0.0041530	0.25	0.88	0.32	-0.17	-0.49
2	1.6288719	0.0174957	1.07	0.0120537	0.74	1.30	0.56	-0.18	-0.50
3	1.7338514	0.0348296	2.01	0.0406630	2.35	3.09	1.51	6.25	5.92
4	1.6303921	0.0086414	0.53	0.0053087	0.33	0.62	0.26	-0.09	-0.40
5	1.6351137	0.0042043	0.26	0.0030714	0.19	0.32	0.14	0.20	-0.12
6	1.6432590	0.0077959	0.47	0.0049660	0.30	0.56	0.24	0.70	0.38
7	1.6107950	0.0131046	0.81	0.0000000	0.00	0.81	0.27	-1.29	-1.60
8	1.5719490	0.0248658	1.58	0.0000000	0.00	1.58	0.53	-3.67	-3.97
9	1.6402369	0.0617489	3.76	0.0445195	2.71	4.64	2.01	0.52	0.20
10	1.6345682	0.0016910	0.10	0.0010587	0.06	0.12	0.05	0.17	-0.15
11	1.5673256	0.0845894	5.40	0.0000000	0.00	5.40	1.80	-3.95	-4.26
12	1.6410389	0.0166857	1.02	0.0116866	0.71	1.24	0.53	0.56	0.25
13	1.6352406	0.0084971	0.52	0.0024311	0.15	0.54	0.19	0.21	-0.11
14	1.6470871	0.0165002	1.00	0.0000000	0.00	1.00	0.33	0.94	0.62
15	1.6562967	0.0066982	0.40	0.0080008	0.48	0.63	0.31	1.50	1.18
16	1.6185112	0.0209266	1.29	0.0110565	0.68	1.46	0.58	-0.82	-1.13
17	1.5484076	0.0186785	1.21	0.0162134	1.05	1.60	0.73	-5.11	-5.41
18	1.6266165	0.0114202	0.70	0.0164238	1.01	1.23	0.63	-0.32	-0.63
19	1.6338310	0.0040179	0.25	0.0035492	0.22	0.33	0.15	0.12	-0.19
20	1.7383175	0.0277958	1.60	0.0485384	2.79	3.22	1.70	6.53	6.19
21	1.6054153	0.0092345	0.58	0.0049375	0.31	0.65	0.26	-1.62	-1.53
22	1.6039133	0.0227240	1.42	0.0000000	0.00	1.42	0.47	-1.71	-2.02
23	1.6520281	0.0026737	0.16	0.0113835	0.69	0.71	0.40	1.24	0.92

Tab. 4.5: Compilation of laboratory related results of evaluation for sample D

Column 4: The relative value of the standard deviation as given in column 3 defined by

$$RS_i^s = \frac{S_i^s}{y_{i..}} 100 (\%) \quad . \quad (4-2)$$

Column 5: An estimate of the standard deviation of the error component 'target'  $S_i^t$  defined by

$$S_i^t = \sqrt{\frac{1}{3} \left[ \frac{\sum_{j=1}^3 (y_{ij.} - y_{i..})^2}{(3-1)} - (S_i^s)^2 \right]} \quad . \quad (4-3)$$

This value characterizes the reproducibility of target preparation for laboratory  $i$ <sup>1)</sup>.

Column 6: The relative value of the standard deviation as given in column 5 defined by

$$RS_i^t = \frac{S_i^t}{y_{i..}} 100 (\%) \quad . \quad (4-4)$$

Column 7: An estimate of the relative standard deviation of the error component 'spectrum plus target'  $RS_i^{s,t}$  calculated by

$$RS_i^{s,t} = \sqrt{(RS_i^s)^2 + (RS_i^t)^2} (\%) \quad . \quad (4-5)$$

This value characterizes the relative standard deviation of a measurement result obtained by laboratory  $i$  in counting one target once.

Column 8: An estimate of the relative standard deviation  $RSM_i^{s,t}$  of the laboratory mean  $y_{i..}$  calculated by

$$RSM_i^{s,t} = \sqrt{\frac{1}{9} (RS_i^s)^2 + \frac{1}{3} (RS_i^t)^2} (\%) \quad . \quad (4-6)$$

---

<sup>1)</sup> If zero is printed out, the value calculated by (4-3) is zero or imaginary; it is customary to consider the standard deviation 'target' in such cases as not significant.

The values calculated using this expression depend on the special measurement procedure applied for AS-76, counting each of three targets three times.

Column 9: The deviation of the laboratory mean  $y_{i..}$  from the grand mean  $y_{...}^*$  after elimination of outliers as described below

$$\Delta GM_i = \frac{y_{i..} - y_{...}^*}{y_{...}^*} 100 \quad (\%) \quad . \quad (4-7)$$

The value given in brackets represents the deviation of the laboratory mean  $y_{i..}$  from the grand mean  $y_{...}$  as defined by (1-3).

Column 10: The deviation of the laboratory mean  $y_{i..}$  from the characterization value CV stated in Section 2 (Tab. 2.2)

$$\Delta CV_i = \frac{y_{i..} - CV}{CV} 100 \quad (\%) \quad . \quad (4-8)$$

The values calculated for the grand means  $y_{...}$  as defined by (1-3) are compiled in Tab. 4.6, column 2. In column 3 of the same table, the relative standard deviations of these grand means

$$RSM = \frac{100}{y_{...}} \sqrt{\frac{1}{23(23-1)} \sum_{i=1}^{23} (y_{i..} - y_{...})^2} \quad (\%) \quad (4-9)$$

are given <sup>1)</sup>.

To check whether or not the values of the grand means and their RSD's were influenced significantly by extraordinary results of individual laboratories, an outlier test according to Dixon /6/ was carried out for each sample on the basis of the laboratory means  $y_{i..}$ . For samples B, C and D no outliers were detected, if error probabilities of 5 % and more were accepted. However, in the case of sample A, three laboratory means were identified to

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<sup>1)</sup> Eq. (4-9) is only valid in the orthogonal case which holds for this experiment.

be outliers according to this test with a probability of error of less than 1 % <sup>1</sup>). For this reason, the grand mean  $\bar{y}_{...}^*$  and its associated relative standard deviation  $RSM^*$  were calculated on the basis of the 20 laboratory means remaining after the rejection of the 3 outlier values. These figures are given in columns 5 and 6 of Tab. 4.6. Possible error sources responsible for these outliers will be discussed in Section 7.

In Fig. 4.5, the relative deviations of the laboratory means from these grand means are shown. For sample A, this is the grand mean  $\bar{y}_{...}^*$  calculated without the three values considered as outliers. For sample B, C and D the grand mean  $\bar{y}_{...}^*$  equals the grand mean  $\bar{y}_{...}$  since no data had to be rejected.

The error bars of the laboratory means indicated correspond to the values given in column 8 of Tabs. 4.2 to 4.5.

The dashed line represents the characterization value CV given in Section 2. Although this value is slightly higher in all cases than the grand means, no significant difference can be stated if one takes into consideration the uncertainty ranges indicated. This means that there is no evidence for the existence of any bias common to all laboratories and specific for this analytical method.

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<sup>1</sup>) This concerns the laboratories 14, 17 and 21; as shown in Part II of this report, the values obtained by laboratories 14 and 17 came into line when a common spectrum evaluation procedure was applied.

1	2	3	4	5	6
Sample	Grand mean $y_{\dots}$	Rel. stand. dev. of grand mean $y_{\dots}$ RSM (%)	Number of outliers <sup>1)</sup>	Grand mean after elimination of outliers $y_{\dots}^*$	Rel. stand. dev. of grand mean $y_{\dots}^*$ RSM* (%)
A	0.39612	0.65	3	0.39404	0.27
B	1.44187	0.48	0	1.44187	0.48
C	2.96758	0.69	0	2.96758	0.69
D	1.63183	0.55	0	1.63183	0.55

Tab. 4.6: Grand means of the alpha-activity ratios Pu-238/(Pu-239 + Pu-240) and their relative standard deviations.

<sup>1)</sup> Dixon criterion applied to laboratory means with probability of error below 1 %

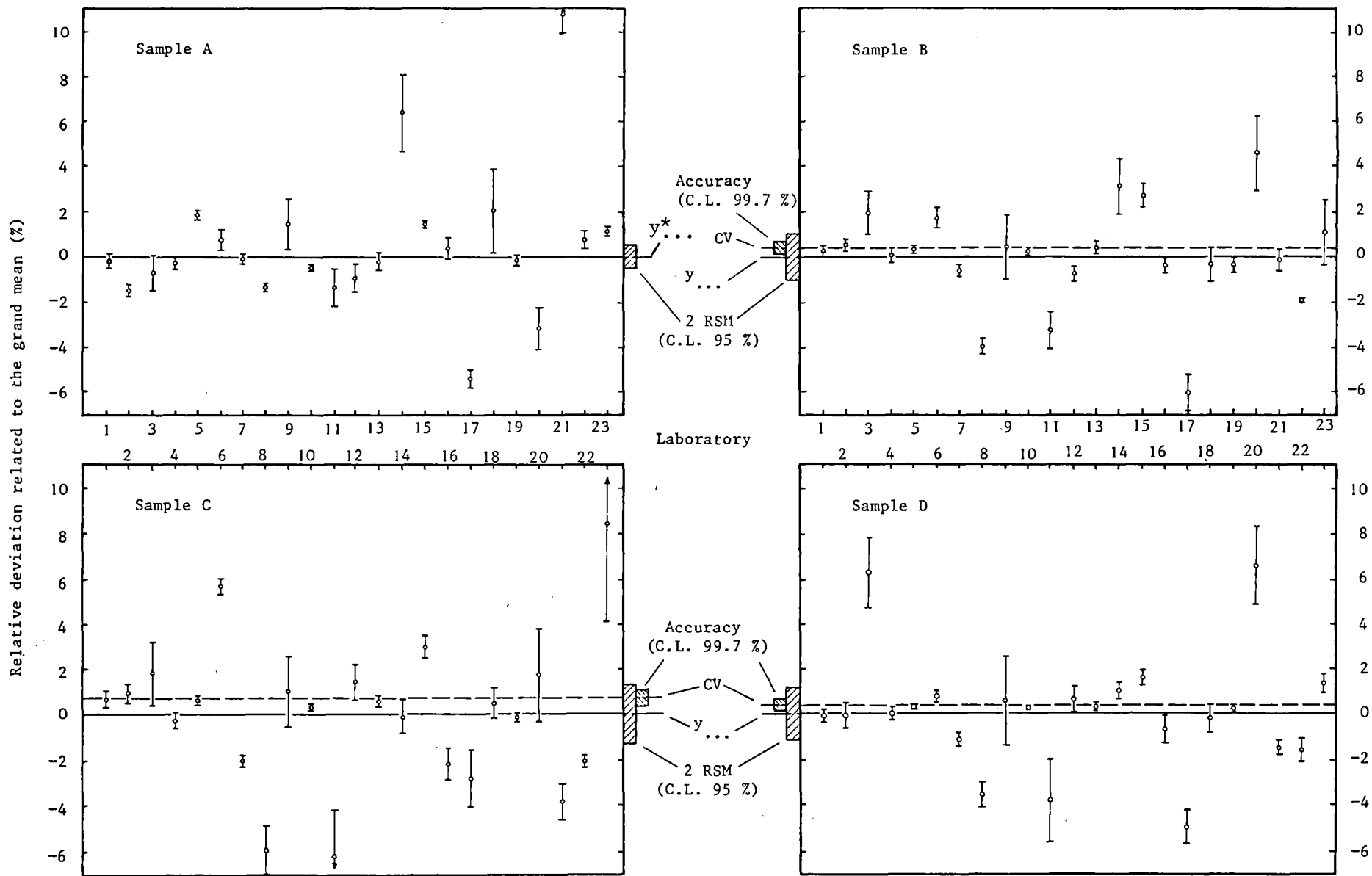


Fig. 4.5: Deviations of the laboratory means  $y_{i..}$  from the grand means  $y_{...}$  (for sample A calculated without laboratories 14, 17, and 21); error bars indicate RSDs of the laboratory means.



## 5. Application of Statistical Tests

A series of statistical tests have been performed in order to

- verify the statistical homogeneity of the data material used for the calculations presented in Section 4;
- examine the significance of possible effects related to source preparation;
- investigate whether the calculated deviations of the laboratory means from the reference values might be explained as laboratory related systematic errors.

The results obtained are compiled in Tabs. 5.1 to 5.4. In these tables, for example in column 2 the symbol '-' indicates that 'no deviation' from homogeneity was found, the values '5', '0.5', and '0.1' indicate that a deviation was found from homogeneity with a probability of error less than 5 %, 0.5 %, and 0.1 %, respectively. In detail, the test procedures applied and their results are:

Column 2: Results of the Bartlett test /7/ ('Test 1') applied to the 3 spectrum variances associated with the three target means per sample and laboratory.

Only if the homogeneity of these data is confirmed (test result '-'), calculation of the spectrum reproducibility on the basis of all 9 spectrum measurements by analysis of variances was permitted from the statistical point of view <sup>1)</sup> (see (4-1) and Tabs. 4.2 to 4.5, column 3).

In almost all the cases to be checked, application of the Bartlett test confirmed the homogeneity of the data. As to the exemption, a deviation from homogeneity is indicated if a prob-

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<sup>1)</sup> Furthermore, the 3 results of spectrum measurement of each source must be homogeneous. Checking this, however, requires the application of an outlier criterion to a data set of 3 values only which does not seem meaningful.

```

*****
      1          2          3          4          5          6
*****
LABORATORY  TEST 1    TEST 2    TEST 3    TEST 4    TEST 5
*****
      1          -          -          -
      2          -          -          0.1
      3          -          -          -
      4          -          -          -
      5          -          -          0.1
      6          -          -          -
      7          -          -          -
      8          -          -          0.1
      9          -          -          -
     10          -          -          0.1
     11          -          -          -
     12          -          -          -
     13          -          -          -
     14          -          5          0.1
     15          -          -          0.1
     16          -          -          -
     17          -          -          0.1
     18          -          0.1        -
     19          -          0.5        -
     20          -          -          0.5
     21          5          5          0.1
     22          -          -          5
     23          -          -          0.1
*****

```

Evaluation not possible due to missing characterization value

Evaluation not possible due to missing characterization value

Tab. 5.1: Results obtained with statistical tests applied to measurement data of sample A.

```

*****
  1           2           3           4           5           6
*****
LABORATORY  TEST 1     TEST 2     TEST 3     TEST 4     TEST 5
*****
  1           -           -           -           -           -
  2           -           -           5           -           -
  3           -           -           5           5           -
  4           -           -           -           -           -
  5           5           -           5           -           -
  6           -           -           0.1         0.5         0.5
  7           -           -           0.5         0.5         5
  8           -           0.5         0.1         0.1         0.1
  9           -           -           -           -           -
 10          -           -           -           -           -
 11          -           -           0.5         0.5         0.5
 12          -           -           5           0.5         5
 13          -           -           -           -           -
 14          -           -           0.5         5           5
 15          -           0.5         0.1         0.1         0.1
 16          5           -           -           5           -
 17          -           -           0.1         0.1         0.1
 18          -           0.5         -           -           -
 19          -           5           -           0.5         -
 20          -           -           5           5           5
 21          -           -           -           -           -
 22          -           -           0.1         0.1         0.1
 23          -           0.1         -           -           -
*****

```

Tab. 5.2: Results obtained with statistical tests applied to measurement data of sample B.

```
*****
  1          2          3          4          5          6
*****
LABORATORY  TEST 1    TEST 2    TEST 3    TEST 4    TEST 5
*****
```

LABORATORY	TEST 1	TEST 2	TEST 3	TEST 4	TEST 5
1	-	-	5	-	-
2	-	-	5	-	-
3	-	-	-	-	-
4	-	-	-	0.5	5
5	-	-	5	-	-
6	-	-	0.1	0.1	0.1
7	-	-	0.1	0.1	0.1
8	-	0.1	0.1	0.1	0.1
9	-	-	-	-	-
10	-	-	5	5	-
11	-	-	0.5	0.5	0.5
12	-	-	5	-	-
13	5	-	5	-	-
14	-	-	-	-	-
15	-	5	0.1	0.1	0.1
16	-	-	0.5	0.1	0.5
17	-	-	5	5	5
18	-	0.5	-	-	-
19	-	-	-	0.1	0.5
20	-	0.5	-	-	-
21	-	0.5	0.1	0.1	0.1
22	-	-	0.1	0.1	0.1
23	-	0.1	0.5	0.5	5

```
*****
```

Tab. 5.3: Results obtained with statistical tests applied to measurement data of sample C.

```
*****
  1           2           3           4           5           6
*****
LABORATORY  TEST 1     TEST 2     TEST 3     TEST 4     TEST 5
*****
  1           -           -           -           -           -
  2           -           -           -           -           -
  3           -           -           0.1       0.1       0.1
  4           -           -           -           -           -
  5           -           -           -           -           -
  6           -           -           0.5       -           -
  7           -           -           0.1       0.1       0.1
  8           5           -           0.1       0.1       0.1
  9           -           -           -           -           -
 10           -           -           0.5       0.5       -
 11           -           -           5         5         5
 12           -           -           -           -           -
 13           -           -           -           -           -
 14           -           -           5         -           -
 15           -           5           0.1       0.1       0.5
 16           -           -           -           5         -
 17           -           -           0.1       0.1       0.1
 18           -           5           -           -           -
 19           -           -           -           -           -
 20           -           5           0.1       0.1       0.1
 21           -           -           0.1       0.1       0.1
 22           -           -           0.5       0.5       0.5
 23           -           0.1       0.1       0.5       5
*****
```

Tab. 5.4: Results obtained with statistical tests applied to measurement data of sample D.

ability of error up to 5 % is allowed. Considering the overall picture (Figs. 4.1 to 4.4) it seemed justified to accept also these data as being homogeneous.

Column 3: Results on the significance of differences between the three target means per sample obtained by each laboratory.

Such differences might indicate the introduction of errors in connection with source preparation. In the statistical test applied the ratio of the standard deviations related to 'target' and 'spectrum' is checked essentially (Tabs. 4.1 to 4.4, columns 5 and 3, respectively), applying the F-distribution /8/, /9/ ('Test 2'). It should be noted that in case of relatively high 'spectrum' standard deviations, the significance of a test result might be camouflaged. For example, for sample A this effect is displayed by the data reported by laboratories 9 and 11 (see Fig. 4.1).

Columns 4 to 6: Several t-test procedures /7/ were performed in order to obtain information on the significance of the difference between the laboratory means  $y_{i..}$  and the grand mean  $y_{...}^*$  ('Test 3') as well as on the characterization value CV ('Test 4')<sup>1)</sup>.

Furthermore, for such laboratory means which lie outside the range given by  $CV \pm 0.3 \%$  the significance of their deviations from these range limits was tested ('Test 5'). For such laboratory means lying within this range, '-' is printed out in column 6 of Tabs. 5.1 to 5.4 as if the test result would indicate 'no deviation'. (For the values of these differences see Tabs. 4.1 to 4.4, columns 9 and 10).

Significant deviations might indicate an error source, related to the laboratory internal preparation of the sample or, in

---

<sup>1)</sup> In these tests the standard deviation of the grand mean  $y_{...}^*$  and the accuracy of the characterization value CV was not taken care of.

particular if they exist for the results of several samples, a systematic error of the laboratory, e.g. introduced by the method of spectrum evaluation. As with 'Test 2', a relatively high standard deviation associated with the laboratory mean might veil the significance of a test result.

## 6. Overall Capability of the Analytical Method

Besides the results presented in Section 4 which are in particular interesting for the individual laboratory, further information can be derived from this experiment, which concerns the overall capability of the analytical method investigated.

As already mentioned in Section 1, the structure of the experiment (see Fig. 1.2) allows to break down into three single error components the total error obtained in the determination of the alpha-activity ratio, and to calculate estimated values of their variances (using the method of variance analysis) /8/. Such values are means based on the measurements of all laboratories. They describe

- the error contribution associated with alpha-counting (error component 'spectrum');
- the error contribution associated with the preparation of the alpha-source (error component 'target');
- the error contribution associated with all laboratory specific procedures, such as chemical sample preparation and method of spectrum evaluation (error component 'interlaboratory deviation').

The variance analysis for the four samples A, B, C and D based on the complete sets of measurement data available resulted in the values compiled in Tab. 6.1 and plotted in Fig. 6.1 versus the alpha-activity ratios. Instead of the calculated estimates of the variances themselves, the more customary relative standard deviations of the error components 'spectrum' ( $RS^s$ ), 'target' ( $RS^t$ )<sup>1)</sup> and 'interlaboratory deviation' ( $RS^{id}$ ) are given.

The most striking observations are the relatively high values for sample A, in particular as far as the interlab deviation is concerned, and for the error component 'target' of sample C. As already mentioned, for sample A the means obtained by three laboratories (14, 17 and 21) were identified as outliers according to the Dixon criterion. Furthermore, review of the labora-

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<sup>1)</sup> These values can be considered as average values for the expressions  $RS_i^s$  and  $RS_i^t$  calculated in Section 4 for each laboratory i.



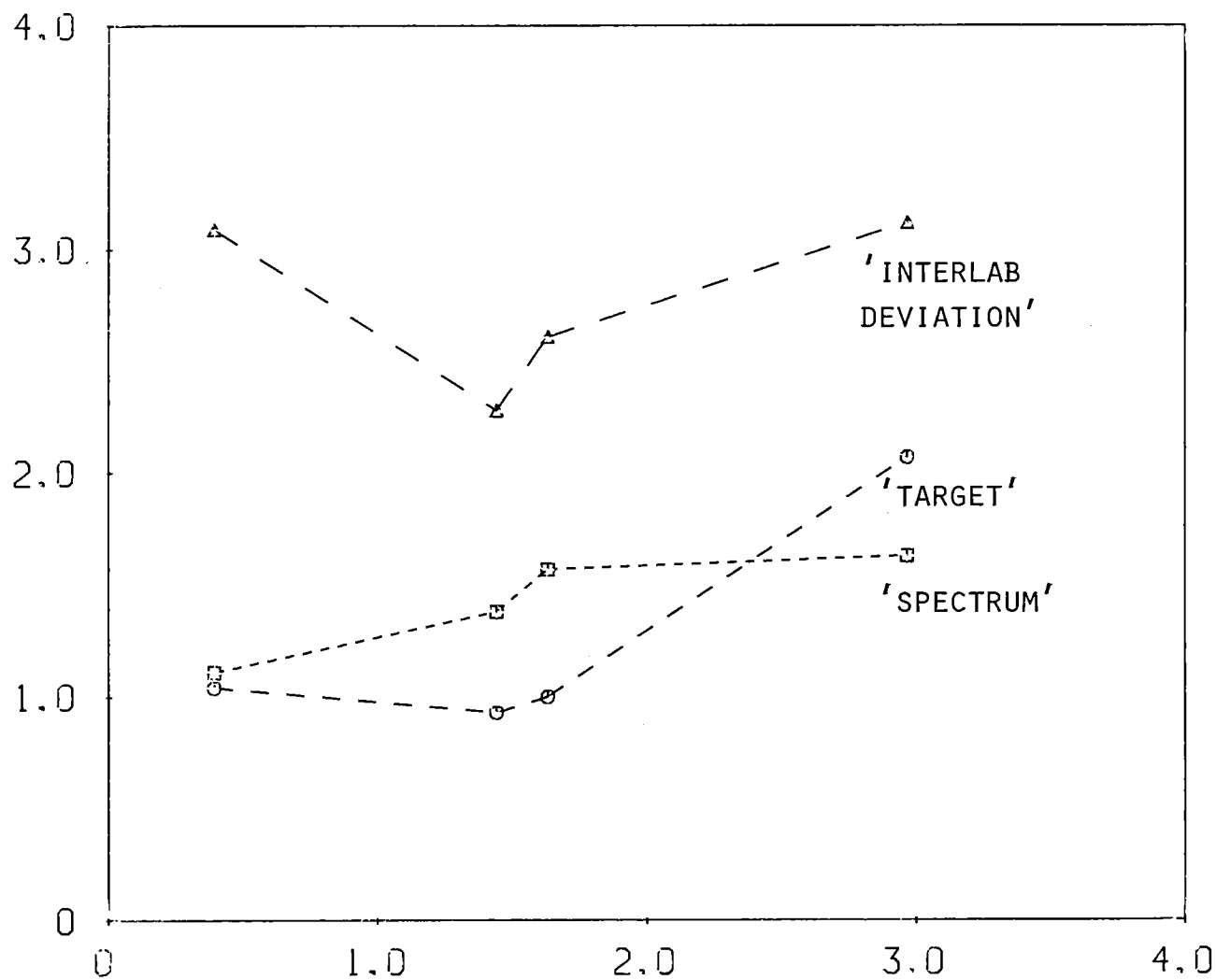
Sample	RS <sup>s</sup> (%)	RS <sup>t</sup> (%)	RS <sup>id</sup> (%)
A	1.11	1.04	3.09
	1.10 <sup>1)</sup>	0.85 <sup>1)</sup>	1.26 <sup>1)</sup>
B	1.38	0.93	2.28
C	1.63	2.07	3.12
	1.65 <sup>2)</sup>	1.24 <sup>2)</sup>	2.78 <sup>2)</sup>
D	1.57	1.00	2.61

<sup>1)</sup> Calculated without the data of laboratories 14, 17 and 21

<sup>2)</sup> Calculated without the data of laboratory 23

Tab. 6.1: Calculated estimates for the RSD's of sample error components.

ESTIMATES FOR RSD'S OF  
ERROR COMPONENTS (%)



$\alpha$ -RATIO PU-238/(PU-239 + PU-240)

Fig. 6.1: Estimates of the relative standard deviations of the single error components versus the alpha-activity ratio; calculated using all data available.

tory related data <sup>1)</sup> reveals the relative standard deviation for the error component 'target' to be excessively high for sample C in the case of laboratory 23. Therefore, the analysis of variances was repeated for samples A and C without the data of the respective laboratories. The results now obtained are also included in Tab. 6.1 and are presented in Fig. 6.2.

There is no doubt that the set of data remaining after these exclusions is still 'inhomogeneous' in the strict statistical sense. Complete 'homogenization', however, would require the exclusion of so many values that the rest could no longer be considered as representative of the actual situation of measurement. However, since the exclusion of the four data sets (values of 3 labs for sample A and 1 lab for sample C) caused the significant changes evident from comparison of Fig. 6.1 and 6.2 whereas rejection of further data has no marked influence, it seems justified to consider the data in Fig. 6.2 as the best estimate obtainable of the actual situation of measurement. It can be considered to be representative since it is based on more than 95 % of the data available.

The total uncertainty of any mean value calculated from the results obtained by  $\ell$  laboratories each preparing  $m$  targets and performing  $n$  spectrum measurements per target is expressed as RSD by (see also /8/ <sup>2)</sup>).

$$\text{RSM} = \sqrt{\frac{(\text{RS}^{\text{id}})^2}{\ell} + \frac{(\text{RS}^{\text{t}})^2}{\ell \cdot m} + \frac{(\text{RS}^{\text{s}})^2}{\ell \cdot m \cdot n}} \quad (6-1)$$

$$= \frac{100}{y_{\dots}} \cdot \sqrt{\frac{1}{\ell \cdot (\ell - 1)} \sum_{i=1}^{\ell} (y_{i\dots} - y_{\dots})^2} \quad (6-2)$$

Formula (6-1) allows to estimate the total uncertainty associated with a laboratory mean value in general by choosing  $\ell=1$ . To demonstrate this, the

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<sup>1)</sup> Tabs. 4.2 to 4.5.

<sup>2)</sup> Due to the identity of (6-1) with (6-2) the RSM values for  $\ell=23$  (after exclusion of outliers  $\ell=20$  for sample A and  $\ell=22$  for sample C) and  $m=n=3$  are identical with the values calculated for the RSD of the grand mean using (4-9).

ESTIMATES FOR RSD'S OF  
ERROR COMPONENTS (%)

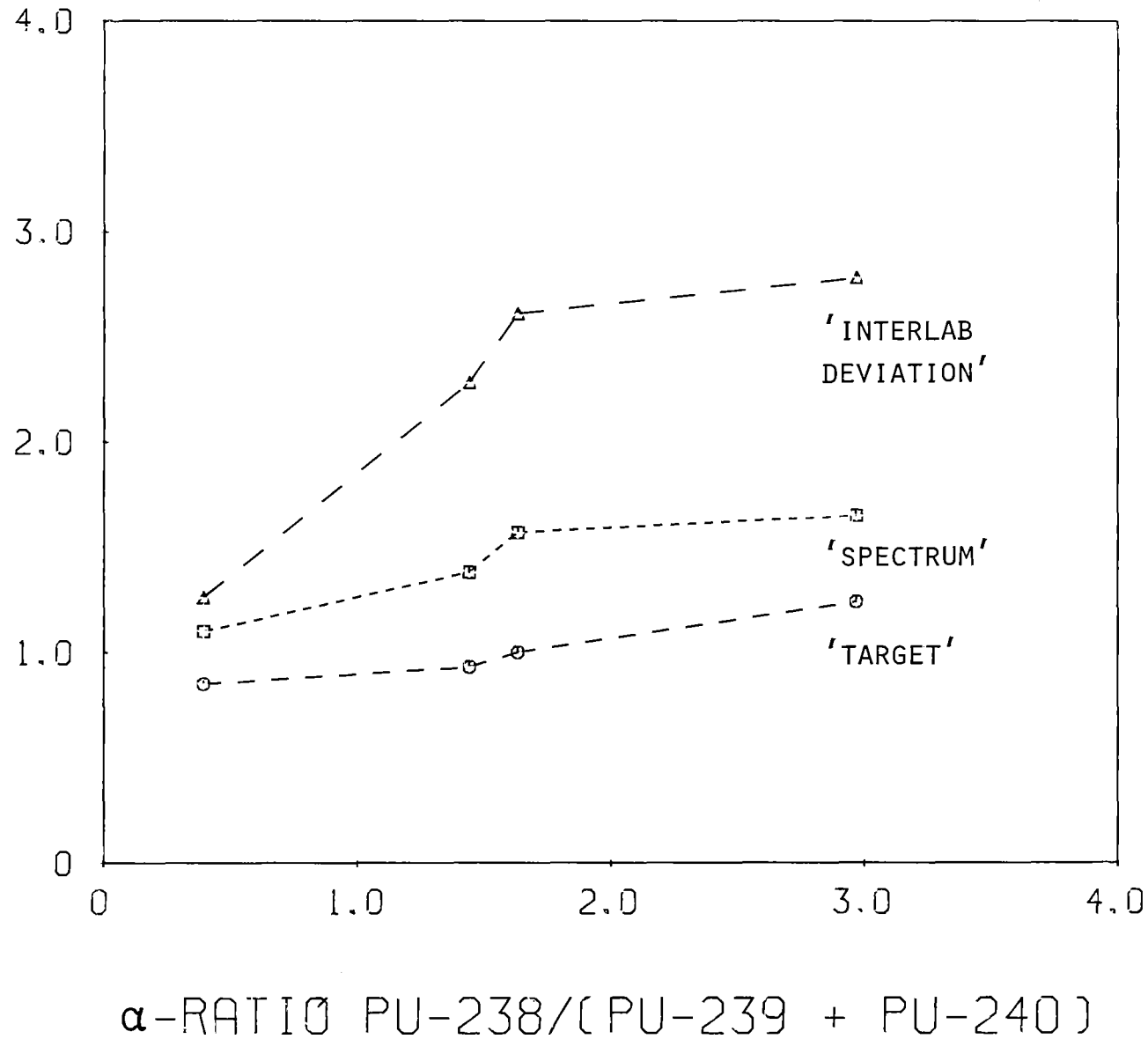
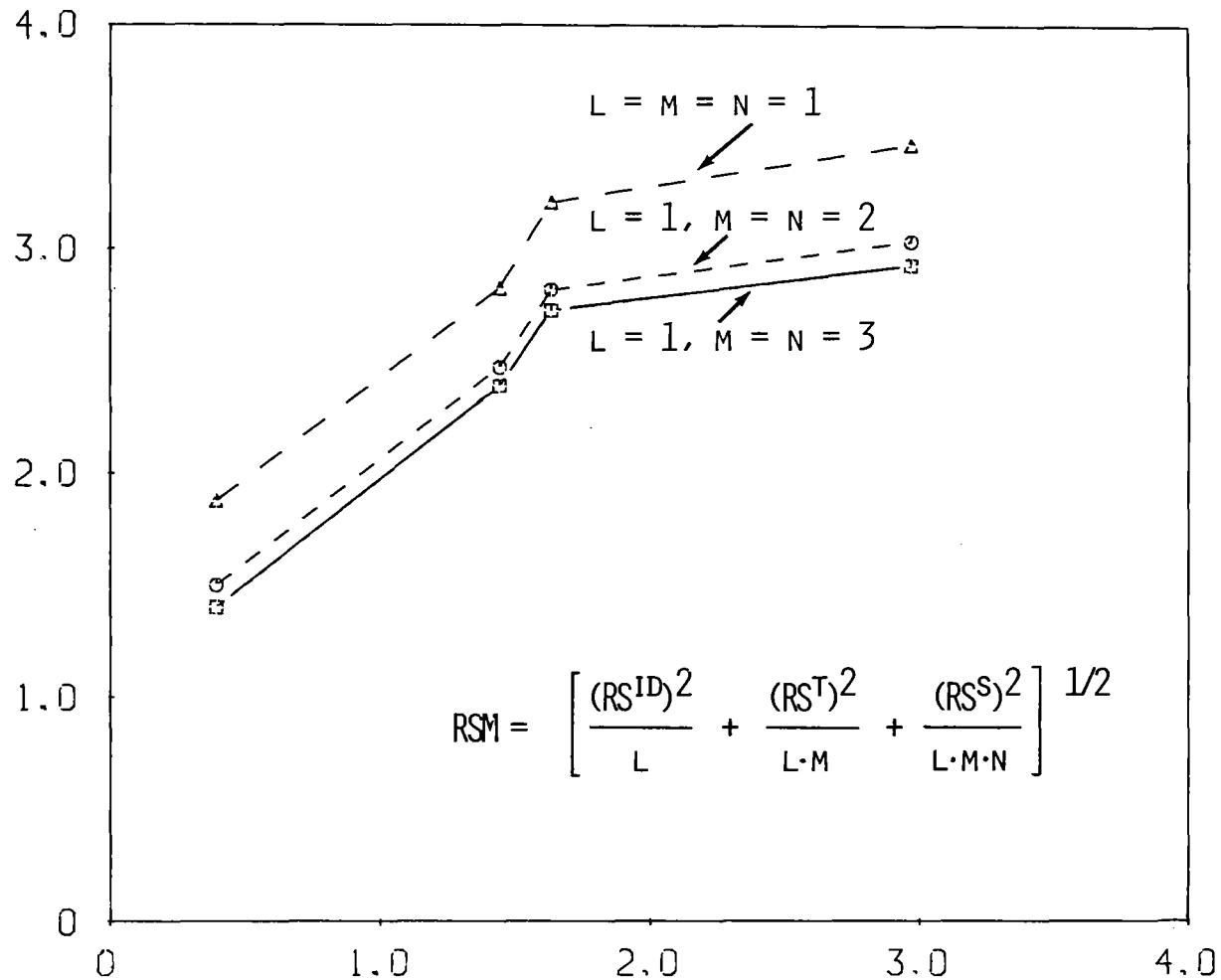


Fig. 6.2: Estimates of the relative standard deviations of the single error components versus the alpha-activity ratio; calculated without data of laboratories 14, 17 and 21 for sample A, without data of laboratory 23 for sample C.

solid curve in Fig. 6.3 was calculated using  $\ell=1$  and  $m=n=3$ . Furthermore, it seemed interesting to present the special cases  $\ell=m=n=1$ , and  $\ell=1$ ,  $m=n=2$ , describing the situations to be expected if a laboratory prepares only one source to be counted once, or two sources, each counted twice. A comparison of the curves obtained for these cases illustrates the influence of repetition measurements on the total uncertainty and shows that for routine analysis an overall uncertainty of about 2 to 3 % can be estimated for the determination of the alpha-activity ratio of Pu-238/(Pu-239 + Pu-240) within the range investigated.

ESTIMATES FOR RSD'S OF  
TOTAL ERROR RSM (%)



$\alpha$ -RATIO PU-238/(PU-239 + PU-240)

**Fig. 6.3:** Estimates of the relative standard deviations of the total error versus alpha-activity ratio; calculated without data of laboratories 14, 17 and 21 for sample A, without data of laboratory 23 for sample C.

## 7. Discussion of Error Components

It follows from Fig. 6.2 that the main error contribution is caused by the interlaboratory deviation  $RS^{id}$ . This finding correlates very well with the results obtained by previous intercomparison programs. This deviation increases with increasing alpha-activity ratios, varying between approximately 1 and 3 %. It comprises all systematic error components, especially those which might be induced by the evaluation procedure applied and, as discussed in Section 1, due to the setup of this experiment, the errors from the chemical sample preparation step performed but once (per sample) by each participant in this program.

As discussed more thoroughly in Part II of this report, application of one common evaluation procedure to the alpha-spectra revealed that the results communicated by the laboratories 14 and 17 might include some error contributed by the spectrum evaluation procedure applied by these laboratories.

Furthermore, it should be mentioned that for sample A (after elimination of outliers),  $RS^{id}$  turned out to be smaller than  $RS^{id}$  of the samples B, C and D. This finding suggests that the error component induced by inhomogeneity in sample A (see Part III of this Report) is exceeded in the samples B, C and D by some error component increasing with higher Pu-238 abundance.

From Fig. 6.2 follows also that the error components 'spectrum' and 'target' display a nearly constant behavior over the alpha-activity range investigated. The error component 'spectrum' depends almost completely on the total number of counts and has been found in the range between 1 and 1.5 %.

Since the number of counts taken per spectrum essentially determines the spectrum reproducibility, it seemed of interest to check to which degree this presumption holds in this experiment. For this purpose, in Figs. 7.1 to 7.4 the RSD estimates of the error component 'spectrum' (reproducibility of spectrum) calculated for each laboratory (see Tabs. 4.2 to 4.5, column 4) were plotted versus the average number of counts reported (see Tabs. 3.2 to 3.5, column 2). To compare for each laboratory this reproducibility with the value to be expected from counting statistics, the curve derived from

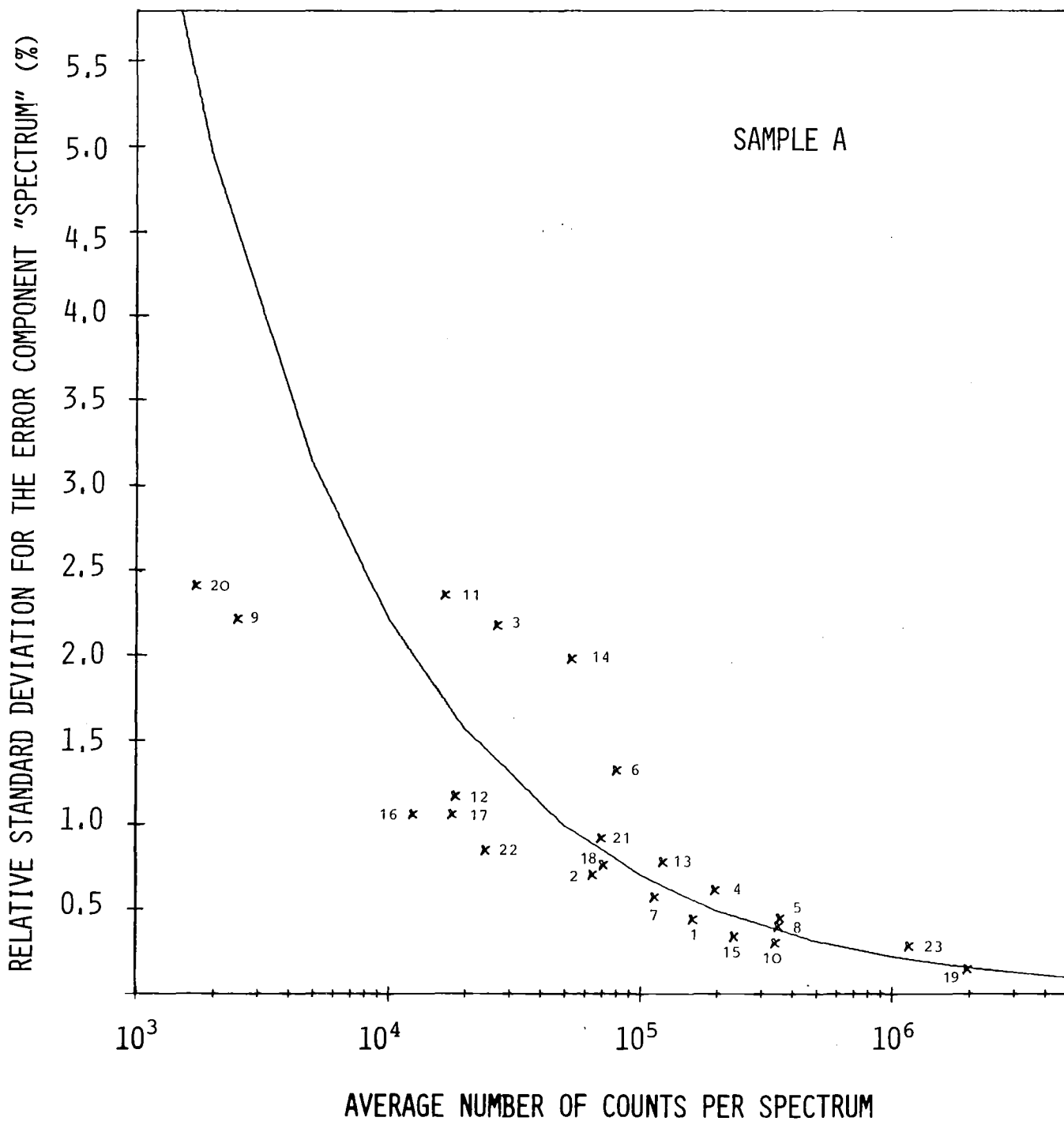


Fig. 7.1: Spectrum reproducibility versus average number of counts taken per spectrum; the curve represents values to be expected only on the basis of the number of counts.



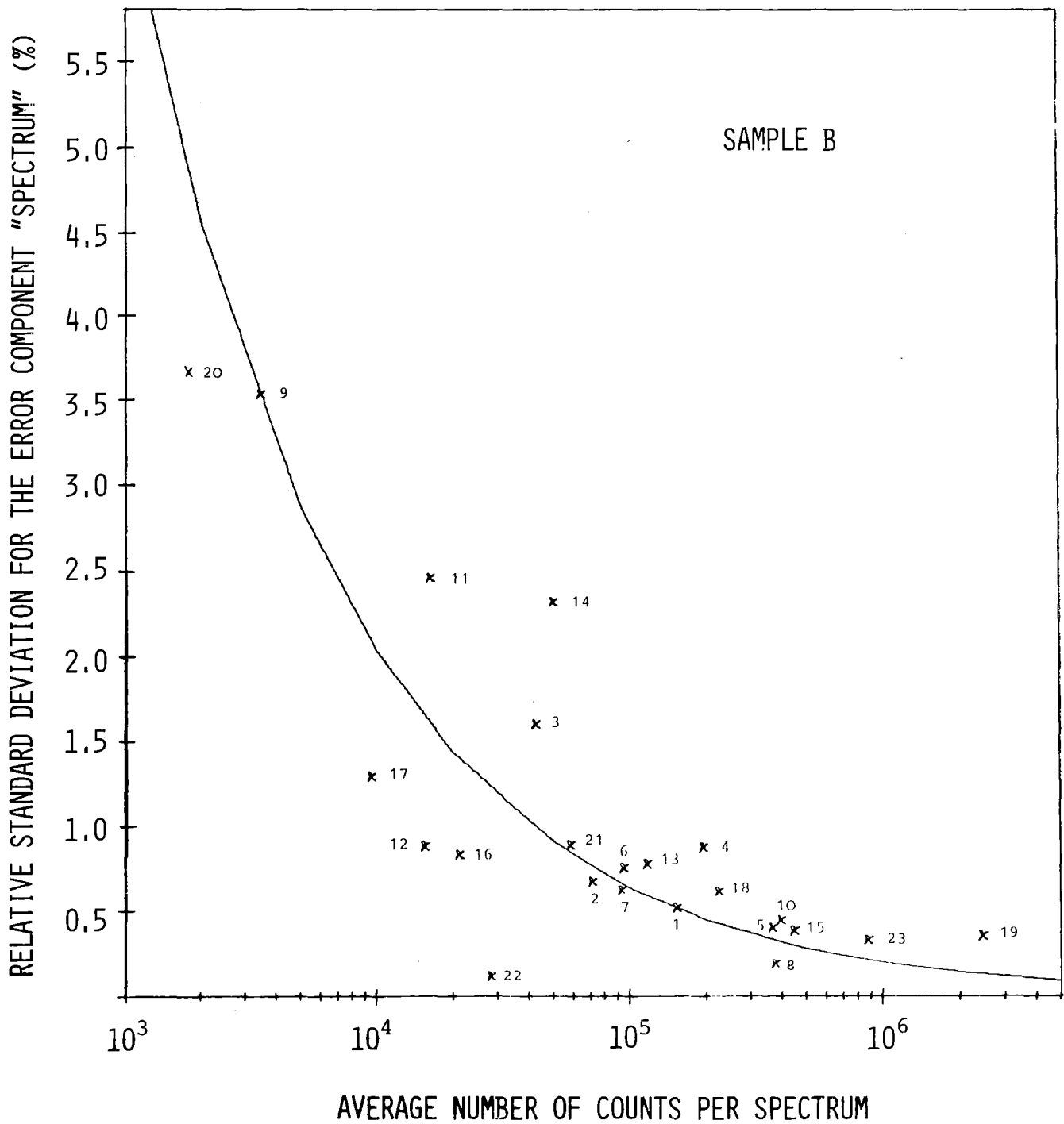


Fig. 7.2: Spectrum reproducibility versus average number of counts taken per spectrum; the curve represents values to be expected only on the basis of the number of counts.

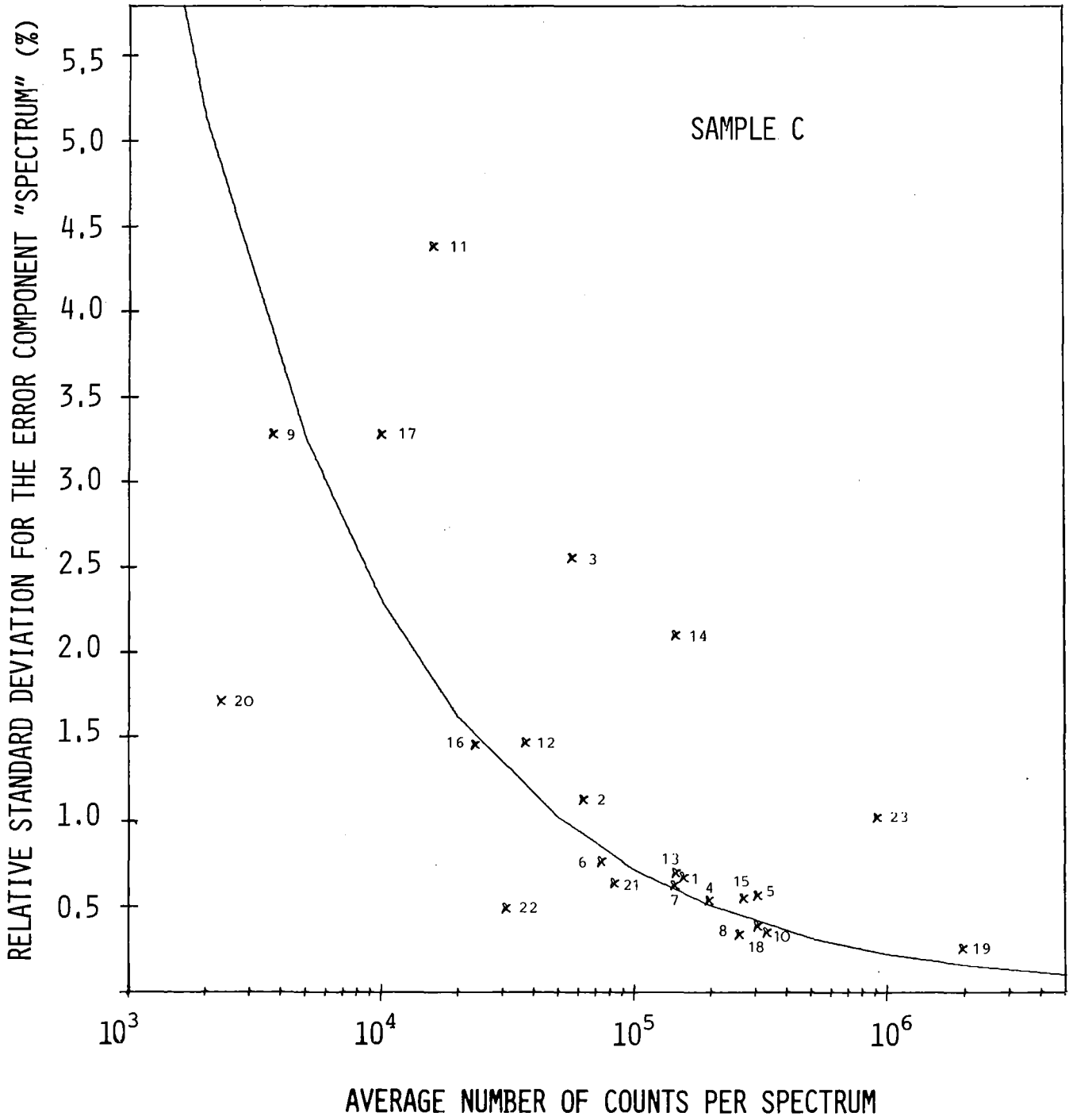


Fig. 7.3: Spectrum reproducibility versus average number of counts taken per spectrum; the curve represents values to be expected only on the basis of the number of counts.

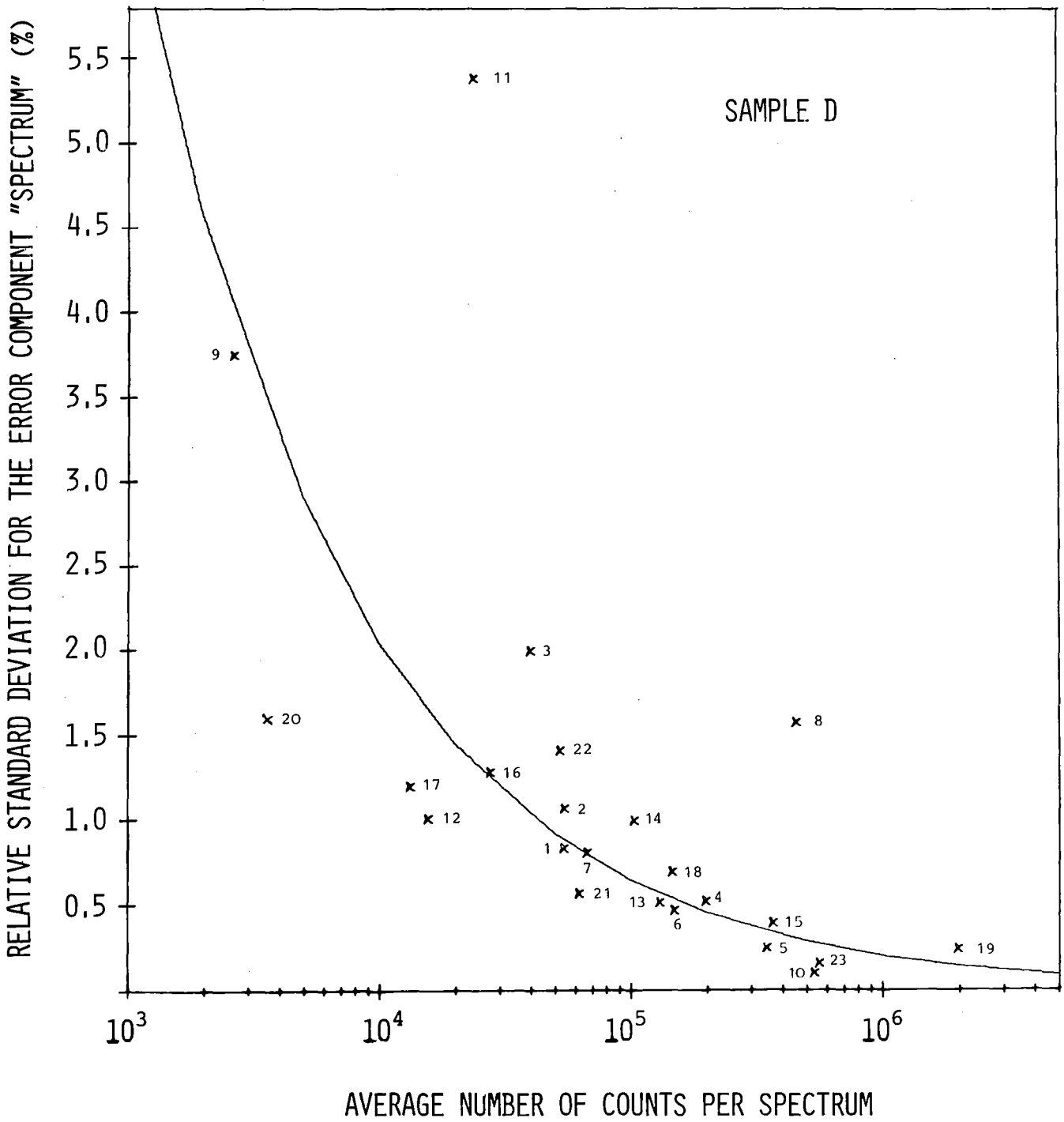


Fig. 7.4: Spectrum reproducibility versus average number of counts taken per spectrum; the curve represents values to be expected only on the basis of the number of counts.

$$F(N) = \frac{1+\alpha}{\sqrt{N \cdot \alpha}} 100 \quad (\%) \quad (7-1)$$

has been plotted also <sup>1)</sup>, where

$\alpha$ : alpha-activity ratio Pu-238/(Pu-239 + Pu-240)

N: total number of counts per spectrum

With the specific experimental layout of AS-76, in which 9 spectra per sample were counted from each laboratory, it should be noted that only one third of the relative standard deviation for the error component 'spectrum' shown in Figs. 7.1 to 7.4 contributes to the relative standard deviation of the laboratory mean <sup>2)</sup>.

According to the layout of the experiment the error component 'target' represents the random variations related to the actual alpha-source preparation (e.g. errors through source thickness effects). This error component turned out to be the smallest one (about 1 %).

Tab. 7.1 presents a survey of the main error contributions observed for the individual laboratories. The '+'-symbol appearing in columns 2 to 5 of this table stands in all cases where  $RS_i^S$  was found to be greater than 2 % <sup>3)</sup> (see Figs. 7.1 to 7.4); the '++'-symbol indicates that (in addition to  $RS_i^S > 2 \%$ ) the average number of counts taken per spectrum was below  $10^4$  counts.

In columns 6 to 9 error contributions related to the target preparation are indicated. The values 0.1, 0.5, and 5 correspond to the probabilities of error already listed in column 3 of Tabs. 5.1 to 5.4. Finally, in columns 10

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<sup>1)</sup> Eq. (7-1) might be derived under the basic assumption that the standard deviation of the statistical counting error is given by  $\sqrt{N}$ .

<sup>2)</sup> This was treated in (4-6) by  $RS_i^S/\sqrt{9}$ .

<sup>3)</sup> The 2 %-value was chosen as the highest acceptable value for this error component; it contributed about 0.7 % to the total uncertainty of measurement.

1	2	3	4	5	6	7	8	9	10	11	12	
Lab. Code	Observed Error Contributions from											
i	'Spectrum'				'Target'				'Interlab Deviation'			
	RS <sub>i</sub> <sup>s</sup> 1)				RS <sub>i</sub> <sup>t</sup> 2)				ΔCV <sub>i</sub> 3)			
	A	B	C	D	A	B	C	D	B	C	D	
1												
2												
3	+		+	+					5	§	0.1 §	
4										0.5	*	
5												
6									0.5	0.1	*	
7									0.5	0.1	*	
8						0.5	0.1		0.1	*	0.1 *	
9	++	++	++	++								
10										5	*	
11	+	+	+	+					0.5	*	0.5 *	
12									0.5		*	
13												
14		+	+		5				5	*		
15						0.5	5	5	0.1	*	0.1 *	
16									5	0.1	*	
17			++						0.1	*	0.1 *	
18					0.1	0.5	0.5	5	*	*		
19					0.5	5			0.5	*	0.1	
20	++	++					0.5	5	5	§	0.1 §	
21					5		0.5				0.1 *	
22									0.1	§	0.1 §	
23						0.1	0.1	0.1			0.5 §	

1) '+'-symbol :  $RS_i^s > 2\%$  (Section 4, Tabs. 4.2 to 4.5)  
 '++'-symbol:  $RS_i^s > 2\%$  and  $N < 10^4$  counts (Section 3, Tabs. 3.2 to 3.5).  
 2) Test 2-results from Section 5, Tabs. 5.1 to 5.4.  
 3) Test 4-results from Section 5, Tabs. 5.1 to 5.4;  
 n.b.:  $\Delta CV_i = y_{i..} - CV$  represents a quantity related to the interlaboratory deviation (see also Section 4, Figs. 4.5).  
 '\*'-symbol : the application of one common evaluation procedure induced a reduction of  $\Delta CV_i$ .  
 '§'-symbol : the application of one common evaluation procedure was not possible.

Tab. 7.1: Observed Error Contributions.

to 12 the results from test 4 (Tabs. 5.1 to 5.4, column 5) <sup>1)</sup> are presented as a measure of estimating the interlaboratory deviation; again the numbers indicate the probability of error.

In addition, in columns 10 to 12 the '\*'-symbol stands in all cases where the application of one common spectrum evaluation procedure resulted in a reduction of the difference  $\Delta CV_i$  between the reported laboratory means  $y_{i..}$  and the characterization value CV (see also Part II of this report). The '\$'-symbol indicates that application of one common procedure was not possible.

Obviously, application of one common evaluation procedure to the alpha-spectra reduced the difference  $\Delta CV_i$ , especially in those cases where this difference was indicated with a probability of error below 0.1 %. This reduction corresponds to a reduction of the interlaboratory deviation, which, as mentioned before, contributes essentially to the overall uncertainty.

It should be pointed out that the values of the error components of samples B and D are approximately equal although sample B contained fission products while sample D did not.

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<sup>1)</sup> T-test on the deviation of the laboratory mean  $y_{i..}$  from the characterization value CV.

## 8. Conclusions and Recommendations

On February 13/15, 1979, the results presented above as well as those described in Parts II and III of this report were discussed at Karlsruhe by the representatives of the participating laboratories listed in Appendix B. The conclusions and recommendations elaborated at this meeting will be given below.

### A. Conclusions

1. For routine analyses, an overall uncertainty <sup>1)</sup> of about 2 to 3 % can be estimated for the alpha-activity ratio of Pu-238/(Pu-239 + Pu-240) within the range investigated. This uncertainty is caused mainly by the 'inter-laboratory deviation'. It can be tolerated for the Pu element assay in the nuclear fuel cycle for most purposes, excluding calorimetry.
2. From the analysis of variances it was found that
  - the error component 'spectrum' (depending mainly on the total number of counts) was in the range between 1 and 1.5 % <sup>1)</sup>,
  - the error component 'target' was about 1 % (for the specific layout of this experiment which concerns only actual alpha source preparation, without any error contributions from chemical separation steps),
  - the error component 'interlaboratory deviation' varied between about 1 and 3 % with increasing alpha-activity ratio (0.4 up to 3.0).

A common evaluation procedure applied in this test showed that this variation might be caused by difficulties in evaluating the low-energy tail of the Pu-238 peak.
3. From the samples distributed in AS-76 there was no indication that the presence of fission products had a detrimental effect on the accuracy achieved.

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<sup>1)</sup> If not stated otherwise,  $1\sigma$  values are given.

4. Ion exchange and solvent extraction were used for the chemical separation steps by about the same number of participating laboratories. The results do not show that either method has any great advantage over the other.
5. Many techniques were used for alpha source preparation including electro-deposition, direct evaporation, vacuum evaporation, use of spreading agents (e.g. tetraethylene glycol), etc. None of the methods was preferred by the majority of laboratories.
6. The parameter values of the pulse height analysis (e.g. solid angle, counting rates, measuring time) varied considerably from laboratory to laboratory.
7. As already observed with ASET-74, a great variety of spectra evaluation methods are used. They are difficult to classify in a meaningful way.
8. In some spectra peaks have been observed which cannot be identified readily.
9. For the AS-76 sample materials the characterization laboratories determined the isotopic ratios Pu-238/Pu-239 by mass spectrometry and the alpha-activity ratios Pu-238/(Pu-239 + Pu-240) by alpha-spectrometry; using the latter ratios and the values of half-lives given in the report, isotopic ratios Pu-238/Pu-239 were calculated which, within the stated accuracies, were in agreement with the isotopic ratios measured by mass spectrometry.
10. In samples such as those used in AS-76 a ratio of alpha-activities of Pu-238/(Pu-239 + Pu-240) in the range of 0.4 to 3.0 can be certified by selected laboratories with an accuracy of 0.3 %. This accuracy includes a random component at the 99.7 % confidence level ( $3\sigma$ ) as well as systematic uncertainties.
11. There was no significant difference between the characterization values and the grand means calculated from the results of all laboratories.



B. Recommendations concerning the Alpha-spectrometric Determination of Pu-238 in Routine Operation

1. In every case, it must be considered carefully whether valency adjustment is required prior to separation.
2. As a rule of thumb, the alpha source should be counted less than 10 days after chemical separation, if no correction for americium ingrowth is made. (In a sample with an isotopic ratio  $\text{Pu-241/Pu-238} = R_i$  the activity ratio  $\text{Am-241/Pu-238}$  builds up at a rate of  $R_i \cdot 2.6 \cdot 10^{-5}$  per day (Am-241 ingrowth is almost linear with time over at least 20 days)).
3. To achieve optimum results, a thin clean source is required. Electro-deposition, vacuum evaporation and TEG mounted sources have been successfully applied and are recommended.
4. The substrate material chosen should be appropriate to the chemical reagents and source preparation technique used. Its surface should be uniform, scratch free and preferably highly polished. For instance, substrates prepared from highly polished stainless steel, type BS 316 S 16<sup>1</sup>), have been found to be very satisfactory.
5. In a given geometrical arrangement of source and detector, only a fraction of the alpha-particles emitted (in  $4\pi$ ) by the source enter the detector.

The following table gives guidance figures for the measured fraction from a source whose area of active deposit is equal to the effective detector area.

---

<sup>1</sup>) Available 0.4 ± 0.025 mm thick, diameters 25 and 27 mm, from Nuclear Supplies, 123 Knowle Road, Mirfield, West Yorkshire, WF14 9RL.

<u>Effective detector area (mm<sup>2</sup>)</u>	<u>Source to detector distance (mm)</u>	<u>Measured fraction of 4π (in percent)</u>
100	10	5.6
	15	3.0
	20	1.8
	25	1.2
	30	0.8
150	15	4.1
	20	2.6
	25	1.7
	30	1.2
	35	0.9

(For an axial point source, the geometry is about 10 % larger at the shortest distance; the difference between a point source and an extended source diminishes with increasing source to detector distance).

6. In order to get optimum results, relatively low counting rates are recommended to eliminate pulse pileup. As a guideline it can be stated that with most equipment available commercially, a counting rate of 100 to 200 cps is suitable.
7. For Pu-238/(Pu-239 + Pu-240) ratio measurements (in the range 0.4 to 3.0) on a separated plutonium fraction, the integrated counts in the spectrum should be at least 50 000 in order to keep the uncertainty due to counting statistics around 1 % <sup>1)</sup>.
8. With increasing Pu-238/(Pu-239 + Pu-240) ratios, particular care should be taken in evaluation since tailing corrections get more important.
9. If a laboratory suspects that deviations are systematic, it is recommended that the evaluation procedure used be checked.

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<sup>1)</sup> If not stated otherwise, 1σ values are given.

C. General Recommendations

1. Further interlaboratory tests on this subject are not required now.
2. Plutonium isotopic standards of compositions similar to those used in this program are needed.
3. With regard to the inhomogeneity observed in one of the prepared sample solutions, it is recommended that the problems encountered in chemical sample treatment should be investigated by a limited group of laboratories, before starting any new intercomparison program involving input solutions of reprocessing plants.

9. References

- /1/ R. Kraemer, W. Beyrich, 'Joint Integral Safeguards Experiment (JEX 70) at the EUROCHEMIC Reprocessing Plant, Mol, Belgium', KfK 1100/EUR 4576e (1971)
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- /6/ M.G. Natrella, 'Experimental Statistics', National Bureau of Standards Handbook 91
- /7/ G. Gottschalk und R.E. Kaiser, 'Elementare Tests zur Beurteilung von Meßdaten', B.I. Hochschultaschenbücher, Bd. 774 (1972), und 'Einführung in die Varianzanalyse und Ringversuche', B.I. Hochschultaschenbücher, Bd. 775 (1976)
- /8/ R. Avenhaus, 'Analysis of Variances', see p. 7-130 in: R. Kraemer and W. Beyrich (Editors) 'Joint Integral Safeguards Experiment (JEX 70) at the EUROCHEMIC Reprocessing Plant, Mol, Belgium', KfK 1100/EUR 4576e (1971)
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Appendix A

Reported Alpha-activity Ratios and their Correction  
with Respect to the Pu-238 Decay

The alpha-activity ratios of Pu-238/(Pu-239 + Pu-240) reported by the participating laboratories (here called  $y_{ijk}^r$ ) are compiled in Tabs. A.1 to A.4. These data were corrected for Pu-238 decay, taking November 1, 1977 as the date of reference. Tabs. A.5 to A.8 show the time differences  $\Delta T$  (days) between the dates of spectra recording and the date of reference for the four samples, respectively. A negative sign indicates that the date of measurement was before the reference date and vice versa.

The corrected data (here called  $y_{ijk}$ ) are given in Tabs. A.9 to A.12.

*****									
1	2	3	4	5	6	7	8	9	10
*****									
LABORATORY	TARGET 1			TARGET 2			TARGET 3		
CODE	SPECTRUM 1	SPECTRUM 2	SPECTRUM 3	SPECTRUM 1	SPECTRUM 2	SPECTRUM 3	SPECTRUM 1	SPECTRUM 2	SPECTRUM 3
i	$y_{i11}^r$	$y_{i12}^r$	$y_{i13}^r$	$y_{i21}^r$	$y_{i22}^r$	$y_{i23}^r$	$y_{i31}^r$	$y_{i32}^r$	$y_{i33}^r$
*****									
1	0.39390	0.39210	0.38980	0.39340	0.39590	0.39500	0.39390	0.39550	0.39760
2	0.38953	0.38987	0.38563	0.39407	0.38831	0.38881	0.38621	0.39150	0.38883
3	0.39800	0.38500	0.37700	0.39800	0.38700	0.39500	0.40400	0.38700	0.39800
4	0.39420	0.39250	0.39420	0.39330	0.39700	0.39380	0.38920	0.39140	0.39620
5	0.40220	0.40310	0.40330	0.40250	0.40170	0.40280	0.40210	0.40400	0.39810
6	0.39048	0.40475	0.40263	0.39761	0.39265	0.39930	0.40031	0.39373	0.39774
7	0.39330	0.39350	0.39720	0.39780	0.39480	0.39360	0.39150	0.39640	0.39380
8	0.39160	0.38760	0.39130	0.38880	0.38890	0.39060	0.39090	0.38910	0.38860
9	0.38961	0.38861	0.39475	0.39876	0.41068	0.40389	0.40066	0.39294	0.41961
10	0.39190	0.39400	0.39400	0.39130	0.39440	0.39250	0.39250	0.39330	0.39210
11	0.38500	0.39100	0.38500	0.39500	0.40600	0.38500	0.39700	0.37400	0.38500
12	0.39500	0.38500	0.38900	0.40000	0.38900	0.39500	0.39000	0.38500	0.38600
13	0.39996	0.39400	0.39107	0.38990	0.39328	0.38826	0.39551	0.39350	0.39390
14	0.42113	0.44455	0.43571	0.41572	0.40960	0.41529	0.41129	0.41972	0.40489
15	0.39870	0.40080	0.40150	0.40000	0.39830	0.40040	0.40190	0.39950	0.39920
16	0.39710	0.39110	0.38850	0.39520	0.39620	0.39990	0.39190	0.40240	0.39690
17	0.37270	0.37610	0.36780	0.37920	0.37300	0.37100	0.36850	0.37230	0.37530
18	0.42072	0.41338	0.41726	0.39601	0.39768	0.40102	0.38860	0.39432	0.39154
19	0.39380	0.39360	0.39460	0.39250	0.39140	0.39290	0.39460	0.39510	0.39440
20	0.38500	0.39700	0.38400	0.37800	0.36700	0.33900	0.38000	0.33800	0.37000
21	0.44300	0.44300	0.44400	0.42600	0.42800	0.43800	0.43700	0.43400	0.43900
22	0.39000	0.39600	0.39600	0.39700	0.39500	0.39700	0.40400	0.39900	0.39500
23	0.39977	0.39979	0.39852	0.39945	0.39762	0.39970	0.39806	0.39798	0.39553
*****									

Tab. A.1: Alpha activity ratios Pu-238/(Pu-239 + Pu-240) reported by laboratories for sample A.

*****									
1	2	3	4	5	6	7	8	9	10
*****									
LABORATORY	TARGET 1			TARGET 2			TARGET 3		
CODE	SPECTRUM 1	SPECTRUM 2	SPECTRUM 3	SPECTRUM 1	SPECTRUM 2	SPECTRUM 3	SPECTRUM 1	SPECTRUM 2	SPECTRUM 3
i	$y_{i11}^r$	$y_{i12}^r$	$y_{i13}^r$	$y_{i21}^r$	$y_{i22}^r$	$y_{i23}^r$	$y_{i31}^r$	$y_{i32}^r$	$y_{i33}^r$
*****									
1	1.45500	1.45900	1.44800	1.45300	1.43500	1.44300	1.45600	1.45200	1.44000
2	1.44140	1.45410	1.45100	1.47450	1.45300	1.44840	1.45750	1.44760	1.44300
3	1.42900	1.43900	1.46700	1.44700	1.51100	1.49400	1.48700	1.50400	1.47600
4	1.44600	1.45700	1.42500	1.46100	1.43500	1.44600	1.44100	1.45400	1.44100
5	1.44930	1.45840	1.44460	1.44840	1.44740	1.44800	1.45440	1.45090	1.43970
6	1.46582	1.47621	1.46765	1.47140	1.44829	1.45348	1.48423	1.46336	1.49012
7	1.42330	1.44400	1.44010	1.44180	1.42520	1.44030	1.43940	1.44070	1.43030
8	1.38300	1.37900	1.37700	1.35400	1.39700	1.39100	1.38700	1.39000	1.39100
9	1.49644	1.49711	1.47361	1.46624	1.37121	1.44695	1.45516	1.43527	1.35159
10	1.44980	1.44340	1.44220	1.43850	1.44130	1.45560	1.45430	1.45590	1.44570
11	1.35000	1.44000	1.35000	1.40000	1.41000	1.40000	1.35000	1.44000	1.39000
12	1.41900	1.43300	1.43400	1.44800	1.42100	1.44800	1.43400	1.41500	1.44000
13	1.46854	1.44110	1.43595	1.43985	1.43870	1.44867	1.44688	1.45200	1.46120
14	1.51331	1.50615	1.54625	1.44434	1.51998	1.46193	1.41860	1.47419	1.49556
15	1.49090	1.47790	1.48500	1.46530	1.47190	1.46750	1.48840	1.48850	1.50060
16	1.44300	1.43600	1.44200	1.45320	1.41880	1.41710	1.43500	1.43600	1.43500
17	1.32060	1.35250	1.33510	1.37080	1.35880	1.32890	1.36790	1.39060	1.36390
18	1.43156	1.42777	1.43158	1.43298	1.42935	1.40851	1.45557	1.45100	1.46658
19	1.43400	1.44300	1.42700	1.43300	1.43300	1.42700	1.44300	1.44700	1.44700
20	1.47200	1.51200	1.39500	1.53000	1.56500	1.52500	1.61300	1.48300	1.49100
21	1.45400	1.44100	1.44600	1.46700	1.44000	1.42600	1.42900	1.42100	1.42700
22	1.41400	1.41100	1.41200	1.41500	1.41400	1.41500	1.41100	1.40900	1.41400
23	1.50480	1.49350	1.49520	1.43770	1.44520	1.44710	1.42720	1.43380	1.43230
*****									

Tab. A.2: Alpha activity ratios Pu-238/(Pu-239 + Pu-240) reported by laboratories for sample B.

*****									
1	2	3	4	5	6	7	8	9	10
*****									
LABORATORY	TARGET 1			TARGET 2			TARGET 3		
CODE	SPECTRUM 1	SPECTRUM 2	SPECTRUM 3	SPECTRUM 1	SPECTRUM 2	SPECTRUM 3	SPECTRUM 1	SPECTRUM 2	SPECTRUM 3
i	$y_{i11}^r$	$y_{i12}^r$	$y_{i13}^r$	$y_{i21}^r$	$y_{i22}^r$	$y_{i23}^r$	$y_{i31}^r$	$y_{i32}^r$	$y_{i33}^r$
*****									
1	2.97100	3.01000	2.95500	3.00000	2.98200	2.97900	3.02900	2.99600	2.99900
2	2.97490	2.96820	3.02100	3.07440	2.99270	3.00270	3.01300	2.96120	2.99270
3	2.96300	3.08700	2.97700	2.89200	2.96700	3.03100	3.21000	3.07600	3.03200
4	2.96800	2.92300	2.94600	2.98300	2.96200	2.99400	2.96900	2.96200	2.96900
5	3.00060	2.95540	2.98360	2.97240	3.00000	3.00150	2.99380	3.01010	2.99270
6	3.15206	3.11676	3.16607	3.16435	3.16875	3.13159	3.05964	3.11250	3.15078
7	2.93150	2.88780	2.91600	2.88360	2.92240	2.92270	2.92050	2.91950	2.91130
8	2.73900	2.73000	2.73800	2.80500	2.81200	2.83500	2.82600	2.83300	2.83000
9	2.93568	2.88266	2.90020	2.92593	2.93996	3.21387	3.09300	3.00658	3.07623
10	2.97630	2.99010	2.99140	2.98570	2.96870	2.98320	2.99460	2.96980	2.97170
11	3.07000	2.86000	2.76000	2.76000	2.62000	2.82000	2.83000	2.66000	2.66000
12	3.05600	3.03500	3.06500	2.93800	2.95300	3.02800	2.96100	3.07300	2.99500
13	2.98056	2.96921	2.97362	2.97753	2.98442	2.97724	3.03421	2.96578	2.98177
14	3.01700	3.01292	2.93676	3.00218	3.01207	2.89114	3.00000	2.85765	2.94417
15	3.03460	3.02130	3.02400	3.04710	3.09230	3.08300	3.08520	3.05960	3.05650
16	2.85500	2.97500	2.95100	2.90400	2.92800	2.89800	2.84200	2.84400	2.90000
17	2.97820	2.94940	2.91780	2.96140	2.75940	2.75960	2.83750	2.98760	2.76950
18	2.96129	2.94217	2.93783	2.97975	2.98626	3.00143	2.99561	3.01028	3.01947
19	2.95300	2.97000	2.97200	2.96600	2.96700	2.97100	2.94700	2.96100	2.96000
20	2.90700	2.94200	3.04700	3.10500	3.13500	3.18600	2.98600	2.94600	2.92400
21	2.80700	2.82500	2.80000	2.87500	2.90300	2.88600	2.86900	2.82000	2.83100
22	2.87800	2.91200	2.90400	2.89300	2.92000	2.92400	2.88700	2.89100	2.88200
23	3.26740	3.32060	3.31970	3.35990	3.39470	3.45090	2.95830	2.92630	2.94850
*****									

Tab. A.3: Alpha activity ratios Pu-238/(Pu-239 + Pu-240) reported by laboratories for sample C.



*****									
1	2	3	4	5	6	7	8	9	10
*****									
LABORATORY	TARGET 1			TARGET 2			TARGET 3		
CODE	SPECTRUM 1	SPECTRUM 2	SPECTRUM 3	SPECTRUM 1	SPECTRUM 2	SPECTRUM 3	SPECTRUM 1	SPECTRUM 2	SPECTRUM 3
i	$y_{i11}^r$	$y_{i12}^r$	$y_{i13}^r$	$y_{i21}^r$	$y_{i22}^r$	$y_{i23}^r$	$y_{i31}^r$	$y_{i32}^r$	$y_{i33}^r$
*****									
1	1.63700	1.62800	1.60400	1.61900	1.64100	1.65100	1.63700	1.64000	1.64200
2	1.61255	1.62742	1.60156	1.62768	1.65818	1.63324	1.64658	1.61777	1.66024
3	1.65000	1.71700	1.70100	1.78500	1.78900	1.76600	1.68800	1.74900	1.78200
4	1.63200	1.61500	1.63000	1.62500	1.64100	1.63100	1.63900	1.64900	1.63200
5	1.63720	1.64180	1.64620	1.63140	1.63060	1.63990	1.63560	1.63500	1.63550
6	1.62772	1.64761	1.64588	1.65039	1.65398	1.65539	1.63718	1.64022	1.64717
7	1.59380	1.60640	1.62360	1.63080	1.59950	1.61520	1.62380	1.62170	1.61080
8	1.55500	1.56500	1.61500	1.57700	1.57500	1.57300	1.55100	1.60500	1.56100
9	1.51794	1.55451	1.65302	1.58285	1.74317	1.67163	1.67950	1.67035	1.69131
10	1.63890	1.63460	1.63510	1.63670	1.63630	1.63680	1.64000	1.63670	1.63970
11	1.54000	1.57000	1.53000	1.67000	1.42000	1.54000	1.56000	1.59000	1.70000
12	1.64700	1.63400	1.61100	1.64500	1.61400	1.64500	1.64300	1.66600	1.66700
13	1.62970	1.64141	1.65229	1.63359	1.62869	1.63010	1.63447	1.64101	1.62306
14	1.63123	1.65844	1.67025	1.64510	1.64689	1.62384	1.66546	1.64600	1.63403
15	1.66580	1.65620	1.67770	1.64990	1.64730	1.64960	1.65870	1.65750	1.65110
16	1.61900	1.59870	1.60700	1.60400	1.66500	1.64300	1.61960	1.59140	1.61850
17	1.56250	1.53770	1.54100	1.54680	1.59820	1.56100	1.53380	1.51560	1.54000
18	1.60431	1.62181	1.61457	1.62000	1.62991	1.60706	1.66116	1.63450	1.64403
19	1.63700	1.63400	1.62900	1.63300	1.62900	1.64000	1.63900	1.64000	1.63900
20	1.79000	1.78500	1.75800	1.64800	1.69300	1.70400	1.76900	1.78900	1.72300
21	1.59300	1.60600	1.59500	1.59800	1.59500	1.61200	1.61400	1.62200	1.60000
22	1.60200	1.57900	1.64300	1.60000	1.59300	1.57600	1.60900	1.62700	1.59000
23	1.64350	1.64230	1.64090	1.65050	1.65210	1.64450	1.66220	1.66550	1.66570
*****									

Tab. A.4: Alpha activity ratios Pu-238/(Pu-239 + Pu-240) reported by laboratories for sample D.

```

*****
1          2          3          4          5          6          7          8          9          10
*****
LABORATORY          TARGET 1          TARGET 2          TARGET 3
CODE          SPECTRUM 1 SPECTRUM 2 SPECTRUM 3          SPECTRUM 1 SPECTRUM 2 SPECTRUM 3          SPECTRUM 1 SPECTRUM 2 SPECTRUM 3
i           $\Delta T_{i11}$            $\Delta T_{i12}$            $\Delta T_{i13}$            $\Delta T_{i21}$            $\Delta T_{i22}$            $\Delta T_{i23}$            $\Delta T_{i31}$            $\Delta T_{i32}$            $\Delta T_{i33}$ 
*****
1          -90          -89          -88          -90          -89          -88          -90          -89          -88
2          -97          -96          -92          -92          -92          -92          -90          -90          -90
3          -97          -97          -97          -97          -97          -97          -70          -70          -70
4          -69          -69          -69          -53          -52          -52          -50          -50          -50
5          -69          -69          -65          -69          -69          -69          -68          -68          -68
6          -61          -60          -60          -61          -60          -57          -61          -60          -57
7          -103          -102          -103          -103          -103          -102          -103          -103          -102
8          -98          -98          -98          -98          -98          -98          -98          -98          -98
9          -6          -6          -5          -5          -5          -5          -5          -4          -4
10         -80          -76          -75          -80          -76          -75          -78          -76          -75
11         -56          -47          -40          -56          -47          -40          -56          -47          -40
12          6          7          8          6          8          9          6          7          9
13          8          8          8          8          8          8          8          8          8
14          7          7          7          7          7          7          7          7          7
15         -22          -22          -22          -22          -22          -22          -22          -22          -22
16          1          1          1          1          1          1          1          1          1
17          3          3          3          3          3          3          3          3          3
18          8          8          8          8          8          8          8          8          8
19         -33          -33          -33          -13          -13          -13          -13          -13          -12
20         -34          -34          -34          -34          -34          -34          -41          -41          -41
21          37          37          37          37          37          37          38          38          38
22          52          52          52          52          52          52          52          52          52
23          -7          -7          -6          -6          -6          -5          -5          -5          -4
*****

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Tab. A.5: Time differences between dates of spectra recording and Nov. 1, 1977 for sample A.

*****									
1	2	3	4	5	6	7	8	9	10
*****									
LABORATORY	TARGET 1			TARGET 2			TARGET 3		
CODE	SPECTRUM 1	SPECTRUM 2	SPECTRUM 3	SPECTRUM 1	SPECTRUM 2	SPECTRUM 3	SPECTRUM 1	SPECTRUM 2	SPECTRUM 3
i	$\Delta T_{i11}$	$\Delta T_{i12}$	$\Delta T_{i13}$	$\Delta T_{i21}$	$\Delta T_{i22}$	$\Delta T_{i23}$	$\Delta T_{i31}$	$\Delta T_{i32}$	$\Delta T_{i33}$
*****									
1	-95	-92	-91	-95	-92	-91	-95	-92	-84
2	-95	-92	-92	-90	-90	-90	-89	-89	-89
3	-70	-70	-70	-70	-70	-70	-69	-69	-69
4	-70	-70	-69	-60	-60	-60	-57	-57	-57
5	-68	-68	-68	-68	-68	-68	-68	-68	-68
6	-54	-53	-53	-54	-53	-53	-54	-53	-50
7	-98	-98	-97	-98	-98	-97	-98	-97	-97
8	-96	-96	-96	-96	-96	-96	-96	-96	-96
9	-4	-1	-1	-1	-1	-1	1	1	1
10	-78	-76	-75	-78	-76	-75	-78	-76	-75
11	-56	-46	-40	-56	-46	-40	-55	-46	-39
12	-28	-26	-22	-27	-26	-22	-27	-27	-22
13	9	9	9	9	9	9	9	9	9
14	9	9	9	9	9	9	9	9	9
15	-22	-22	-22	-22	-22	-22	-22	-22	-22
16	-1	-1	-1	-1	-1	-1	-1	-1	-1
17	7	7	7	7	7	7	7	7	7
18	9	9	9	9	9	9	9	9	9
19	-12	-12	-12	19	19	19	-11	-11	-11
20	-41	-41	-41	-41	-41	-41	-34	-34	-34
21	38	38	38	38	38	38	38	38	38
22	52	52	52	52	52	52	52	52	52
23	-6	-6	-6	-5	-5	-5	-5	-4	-4
*****									

Tab. A.6: Time differences between dates of spectra recording and Nov. 1, 1977 for sample B.

*****									
1	2	3	4	5	6	7	8	9	10
*****									
LABORATORY	TARGET 1			TARGET 2			TARGET 3		
CODE	SPECTRUM 1	SPECTRUM 2	SPECTRUM 3	SPECTRUM 1	SPECTRUM 2	SPECTRUM 3	SPECTRUM 1	SPECTRUM 2	SPECTRUM 3
i	$\Delta T_{i11}$	$\Delta T_{i12}$	$\Delta T_{i13}$	$\Delta T_{i21}$	$\Delta T_{i22}$	$\Delta T_{i23}$	$\Delta T_{i31}$	$\Delta T_{i32}$	$\Delta T_{i33}$
*****									
1	-81	-78	-78	-81	-78	-77	-81	-78	-77
2	-89	-89	-88	-88	-88	-88	-85	-85	-85
3	-69	-69	-69	-68	-68	-68	-68	-68	-68
4	-69	-69	-69	-62	-62	-62	-61	-61	-61
5	-67	-67	-67	-67	-67	-67	-67	-67	-67
6	-56	-55	-55	-56	-55	-54	-55	-55	-54
7	-95	-95	-94	-95	-94	-93	-95	-94	-93
8	-90	-90	-90	-90	-90	-90	-90	-90	-90
9	1	1	2	2	2	2	3	3	3
10	-78	-75	-74	-76	-74	-71	-78	-74	-68
11	-57	-48	-39	-57	-48	-34	-57	-48	-34
12	-35	-34	-29	-36	-35	-34	-36	-36	-33
13	9	9	9	9	9	9	9	9	9
14	9	9	9	9	9	9	9	9	9
15	-22	-22	-22	-22	-22	-22	-22	-22	-22
16	1	1	1	1	1	1	1	1	1
17	9	9	9	9	9	9	9	9	9
18	13	13	13	13	13	13	13	13	13
19	-11	-8	-8	-11	-8	-8	-11	-8	-8
20	-34	-34	-34	-32	-32	-32	-29	-29	-29
21	41	41	41	41	42	42	42	42	42
22	52	52	52	52	52	52	52	52	52
23	-4	-4	-4	-4	-4	-4	-3	-3	-3
*****									

Tab. A.7: Time differences between dates of spectra recording and Nov. 1, 1977 for sample C.

*****									
1	2	3	4	5	6	7	8	9	10
*****									
LABORATORY	TARGET 1			TARGET 2			TARGET 3		
CODE	SPECTRUM 1	SPECTRUM 2	SPECTRUM 3	SPECTRUM 1	SPECTRUM 2	SPECTRUM 3	SPECTRUM 1	SPECTRUM 2	SPECTRUM 3
i	$\Delta T_{i11}$	$\Delta T_{i12}$	$\Delta T_{i13}$	$\Delta T_{i21}$	$\Delta T_{i22}$	$\Delta T_{i23}$	$\Delta T_{i31}$	$\Delta T_{i32}$	$\Delta T_{i33}$
*****									
1	-119	-119	-119	-119	-118	-118	-120	-119	-118
2	-88	-88	-88	-85	-85	-85	-76	-76	-48
3	-67	-67	-67	-67	-67	-67	-64	-64	-64
4	-68	-68	-68	-64	-64	-64	-61	-61	-61
5	-67	-67	-67	-66	-66	-66	-66	-66	-66
6	-64	-63	-62	-64	-63	-62	-64	-63	-62
7	-91	-91	-90	-91	-90	-90	-91	-90	-90
8	-96	-96	-96	-96	-96	-96	-96	-96	-96
9	-7	-7	-7	-7	-7	-7	-6	-6	-6
10	-78	-76	-68	-78	-76	-74	-76	-74	-68
11	-55	-48	-34	-57	-48	-34	-57	-47	-34
12	-10	-9	-5	-10	-9	-8	-10	-9	-5
13	9	9	9	9	9	9	9	9	9
14	8	8	8	8	8	8	8	8	8
15	-22	-22	-22	-22	-22	-22	-22	-22	-22
16	2	2	2	2	2	2	0	0	0
17	-3	-3	-3	-3	-3	-3	-3	-3	-3
18	7	7	7	7	7	7	7	7	7
19	-88	-88	-88	-35	-35	-34	-14	-14	-14
20	-42	-42	-42	-42	-42	-42	-41	-41	-41
21	44	44	44	44	44	44	44	44	44
22	52	52	52	52	52	52	52	52	52
23	1	1	2	2	2	3	6	6	7
*****									

Tab. A.8: Time differences between dates of spectra recording and Nov. 1, 1977 for sample D.

*****									
1	2	3	4	5	6	7	8	9	10
*****									
LABORATORY	TARGET 1			TARGET 2			TARGET 3		
CODE	SPECTRUM 1	SPECTRUM 2	SPECTRUM 3	SPECTRUM 1	SPECTRUM 2	SPECTRUM 3	SPECTRUM 1	SPECTRUM 2	SPECTRUM 3
i	Y <sub>i11</sub>	Y <sub>i12</sub>	Y <sub>i13</sub>	Y <sub>i21</sub>	Y <sub>i22</sub>	Y <sub>i23</sub>	Y <sub>i31</sub>	Y <sub>i32</sub>	Y <sub>i33</sub>
*****									
1	0.39313	0.39135	0.38906	0.39263	0.39514	0.39425	0.39313	0.39474	0.39684
2	0.38871	0.38906	0.38486	0.39329	0.38754	0.38804	0.38546	0.39074	0.38807
3	0.39716	0.38419	0.37621	0.39716	0.38619	0.39417	0.40339	0.38641	0.39740
4	0.39361	0.39191	0.39361	0.39285	0.39655	0.39336	0.38878	0.39098	0.39577
5	0.40160	0.40250	0.40270	0.40190	0.40110	0.40220	0.40151	0.40341	0.39751
6	0.38996	0.40422	0.40211	0.39709	0.39214	0.39881	0.39978	0.39322	0.39725
7	0.39242	0.39263	0.39032	0.39691	0.39392	0.39273	0.39063	0.39552	0.39293
8	0.39077	0.38678	0.39047	0.38798	0.38808	0.38977	0.39007	0.38628	0.38778
9	0.38956	0.38856	0.39471	0.39872	0.41064	0.40385	0.40062	0.39291	0.41957
10	0.39122	0.39335	0.39336	0.39062	0.39375	0.39186	0.39184	0.39265	0.39146
11	0.38453	0.39060	0.38467	0.39452	0.40559	0.38467	0.39052	0.37362	0.38467
12	0.39505	0.38506	0.38907	0.40005	0.38907	0.39508	0.39005	0.38506	0.38608
13	0.40003	0.39407	0.39114	0.38997	0.39335	0.39833	0.39558	0.39357	0.39347
14	0.42119	0.44462	0.43578	0.41578	0.40966	0.41535	0.41135	0.41978	0.40495
15	0.39951	0.40061	0.40131	0.39981	0.39811	0.40021	0.40171	0.39931	0.39901
16	0.39711	0.39111	0.38851	0.39521	0.39621	0.39991	0.39191	0.40241	0.39691
17	0.37272	0.37612	0.36782	0.37922	0.37302	0.37102	0.36852	0.37232	0.37532
18	0.42079	0.41345	0.41733	0.39608	0.39775	0.40109	0.38867	0.39439	0.39161
19	0.39352	0.39332	0.39432	0.39235	0.39129	0.39279	0.39449	0.39499	0.39430
20	0.38472	0.39671	0.38372	0.37772	0.36673	0.38871	0.37966	0.38766	0.36967
21	0.44335	0.44335	0.44436	0.42634	0.42834	0.43835	0.43736	0.43436	0.43936
22	0.39044	0.39645	0.39645	0.39745	0.39544	0.39745	0.40446	0.39945	0.39544
23	0.39971	0.39973	0.39847	0.39940	0.39757	0.39966	0.39802	0.39794	0.39550
*****									

Tab. A.9: Alpha activity ratios Pu-238/(Pu-239 + Pu-240) corrected to Nov. 1, 1977 for sample A.

*****									
1	2	3	4	5	6	7	8	9	10
*****									
LABORATORY	TARGET 1			TARGET 2			TARGET 3		
CODE	SPECTRUM 1	SPECTRUM 2	SPECTRUM 3	SPECTRUM 1	SPECTRUM 2	SPECTRUM 3	SPECTRUM 1	SPECTRUM 2	SPECTRUM 3
i	Y <sub>i11</sub>	Y <sub>i12</sub>	Y <sub>i13</sub>	Y <sub>i21</sub>	Y <sub>i22</sub>	Y <sub>i23</sub>	Y <sub>i31</sub>	Y <sub>i32</sub>	Y <sub>i33</sub>
*****									
1	1.45201	1.45610	1.44515	1.45001	1.43214	1.44016	1.45301	1.44911	1.43738
2	1.43844	1.45121	1.44811	1.47163	1.45017	1.44558	1.45469	1.44481	1.44022
3	1.42684	1.43682	1.46478	1.44481	1.50871	1.49174	1.48478	1.50175	1.47380
4	1.44381	1.45479	1.42287	1.45910	1.43314	1.44412	1.43922	1.45221	1.43922
5	1.44717	1.45625	1.44247	1.44627	1.44527	1.44587	1.45226	1.44876	1.43758
6	1.46411	1.47452	1.46597	1.46968	1.44663	1.45181	1.48249	1.46168	1.48851
7	1.42028	1.44174	1.43708	1.43874	1.42218	1.43728	1.43635	1.43768	1.42730
8	1.38013	1.37614	1.37414	1.39110	1.39410	1.38811	1.38412	1.38711	1.38811
9	1.49631	1.49708	1.47358	1.46621	1.37118	1.44692	1.49519	1.43930	1.35162
10	1.44735	1.44103	1.43986	1.43607	1.43893	1.45324	1.45185	1.45351	1.44335
11	1.34836	1.43857	1.34883	1.39830	1.40860	1.39879	1.38834	1.43657	1.38883
12	1.41814	1.43219	1.43332	1.44715	1.42020	1.44731	1.43316	1.41417	1.43931
13	1.46883	1.44138	1.43623	1.44013	1.43898	1.44895	1.44716	1.45228	1.46148
14	1.51360	1.50644	1.54655	1.44462	1.52028	1.46221	1.41887	1.47448	1.49585
15	1.49019	1.47720	1.48509	1.46460	1.47120	1.46680	1.46769	1.48779	1.49988
16	1.44297	1.43597	1.44157	1.45317	1.41877	1.41707	1.43897	1.43597	1.43497
17	1.32080	1.35270	1.33530	1.37101	1.35900	1.32910	1.36811	1.39081	1.36411
18	1.43184	1.42805	1.43186	1.43326	1.42963	1.40878	1.45625	1.45128	1.46686
19	1.43363	1.44262	1.42663	1.43359	1.43359	1.42759	1.44266	1.44665	1.44665
20	1.47069	1.51066	1.39376	1.52864	1.56361	1.52365	1.61181	1.48191	1.48990
21	1.45520	1.44219	1.44719	1.46821	1.44118	1.42717	1.43018	1.42217	1.42817
22	1.41559	1.41259	1.41359	1.41659	1.41559	1.41659	1.41259	1.41059	1.41559
23	1.50460	1.49331	1.49500	1.43754	1.44504	1.44694	1.42704	1.43367	1.43217
*****									

Tab. A.10: Alpha activity ratios Pu-238/(Pu-239 + Pu-240) corrected to Nov. 1, 1977 for sample B.

*****									
1	2	3	4	5	6	7	8	9	10
*****									
LABORATORY	TARGET 1			TARGET 2			TARGET 3		
CODE	SPECTRUM 1	SPECTRUM 2	SPECTRUM 3	SPECTRUM 1	SPECTRUM 2	SPECTRUM 3	SPECTRUM 1	SPECTRUM 2	SPECTRUM 3
i	Y <sub>i11</sub>	Y <sub>i12</sub>	Y <sub>i13</sub>	Y <sub>i21</sub>	Y <sub>i22</sub>	Y <sub>i23</sub>	Y <sub>i31</sub>	Y <sub>i32</sub>	Y <sub>i33</sub>
*****									
1	2.96579	3.00492	2.95001	2.99474	2.97697	2.97404	3.02369	2.99094	2.99400
2	2.96917	2.96249	3.01525	3.06855	2.98700	2.99698	3.00746	2.95575	2.98720
3	2.95858	3.08239	2.97256	2.88774	2.96263	3.02654	3.20529	3.07147	3.02754
4	2.96357	2.91864	2.94160	2.97900	2.95803	2.98998	2.96508	2.95809	2.96508
5	2.99625	2.95111	2.97927	2.96809	2.99565	2.99715	2.98546	3.00574	2.98836
6	3.14824	3.11305	3.16230	3.16051	3.16498	3.12793	3.09595	3.10879	3.14710
7	2.92548	2.88187	2.91007	2.87767	2.91646	2.91682	2.91450	2.91356	2.90544
8	2.73367	2.72468	2.73267	2.79954	2.80652	2.82948	2.82050	2.82748	2.82449
9	2.93574	2.88272	2.90032	2.92605	2.94009	3.21401	3.09320	3.00677	3.07643
10	2.97128	2.98525	2.98661	2.98079	2.96395	2.97862	2.98955	2.96504	2.96733
11	3.06621	2.85703	2.75767	2.75659	2.61728	2.81792	2.82651	2.65724	2.65804
12	3.05368	3.03277	3.06308	2.93571	2.95076	3.02577	2.95869	3.07061	2.99286
13	2.98114	2.96979	2.97420	2.97811	2.98500	2.97782	3.03480	2.96636	2.98235
14	3.01759	3.01351	2.93733	3.00276	3.01266	2.89170	3.00058	2.85821	2.94474
15	3.03315	3.01986	3.02256	3.04565	3.09083	3.08153	3.06373	3.05814	3.05504
16	2.85506	2.97506	2.95106	2.90406	2.92806	2.89806	2.84206	2.84406	2.90006
17	2.97878	2.94997	2.91837	2.96198	2.75994	2.76014	2.83845	2.98818	2.77004
18	2.96212	2.94300	2.93866	2.98059	2.98710	3.00227	2.99645	3.01113	3.02032
19	2.95230	2.96548	2.97148	2.96529	2.96649	2.97048	2.94630	2.96049	2.95949
20	2.90486	2.93983	3.04476	3.10265	3.13283	3.18379	2.98412	2.94415	2.92216
21	2.80949	2.92751	2.80249	2.87755	2.90564	2.89863	2.87161	2.82257	2.83357
22	2.88124	2.91528	2.90727	2.89626	2.92329	2.92729	2.89025	2.89426	2.88525
23	3.26712	3.32031	3.31941	3.35961	3.39440	3.45060	2.95811	2.92611	2.94831
*****									

Tab. A.11: Alpha activity ratios Pu-238/(Pu-239 + Pu-240) corrected to Nov. 1, 1977 for sample C.



*****									
1	2	3	4	5	6	7	8	9	10
*****									
LABORATORY	TARGET 1			TARGET 2			TARGET 3		
CODE	SPECTRUM 1	SPECTRUM 2	SPECTRUM 3	SPECTRUM 1	SPECTRUM 2	SPECTRUM 3	SPECTRUM 1	SPECTRUM 2	SPECTRUM 3
i	$Y_{i11}$	$Y_{i12}$	$Y_{i13}$	$Y_{i21}$	$Y_{i22}$	$Y_{i23}$	$Y_{i31}$	$Y_{i32}$	$Y_{i33}$
*****									
1	1.63279	1.62381	1.59587	1.61483	1.63681	1.64679	1.63275	1.63578	1.63781
2	1.60948	1.62432	1.59351	1.62469	1.65513	1.63024	1.64367	1.61511	1.65851
3	1.64761	1.71451	1.65853	1.78241	1.78641	1.76344	1.68566	1.74658	1.77953
4	1.62960	1.61262	1.62760	1.62275	1.63873	1.62874	1.63684	1.64682	1.62985
5	1.63483	1.63942	1.64381	1.62907	1.62827	1.63756	1.63726	1.63266	1.63316
6	1.62547	1.64536	1.64767	1.64810	1.65172	1.65317	1.63491	1.63798	1.64496
7	1.59066	1.60324	1.62044	1.62759	1.59639	1.61205	1.62060	1.61854	1.60766
8	1.55177	1.56175	1.61165	1.57372	1.57173	1.56973	1.54778	1.60167	1.55776
9	1.51771	1.55427	1.65277	1.58261	1.74290	1.67136	1.67528	1.67013	1.65109
10	1.63613	1.63191	1.63265	1.63394	1.63361	1.63418	1.63730	1.63408	1.63729
11	1.53817	1.56837	1.52887	1.66794	1.41852	1.53887	1.55807	1.58838	1.69875
12	1.64664	1.63372	1.61082	1.64464	1.61368	1.64471	1.64264	1.66567	1.66692
13	1.63002	1.64173	1.65261	1.63391	1.62901	1.63042	1.63479	1.64133	1.62337
14	1.63151	1.65873	1.67054	1.64538	1.64717	1.62412	1.66575	1.64628	1.63431
15	1.66501	1.65541	1.67650	1.64911	1.64651	1.64881	1.65791	1.65671	1.65031
16	1.61907	1.59877	1.60707	1.60407	1.66507	1.64307	1.61960	1.59140	1.61850
17	1.56240	1.53760	1.54090	1.54670	1.59810	1.56090	1.53370	1.51550	1.53990
18	1.60455	1.62206	1.61481	1.62024	1.63016	1.60730	1.66141	1.63475	1.64428
19	1.63388	1.63089	1.62590	1.63176	1.62777	1.63879	1.63750	1.63950	1.63350
20	1.78837	1.78338	1.75640	1.64650	1.69146	1.70245	1.76743	1.78741	1.72147
21	1.59452	1.60753	1.59652	1.59952	1.59652	1.61354	1.61554	1.62355	1.60152
22	1.60300	1.58078	1.64485	1.60180	1.59479	1.57777	1.61081	1.62883	1.59179
23	1.64353	1.64233	1.64057	1.65057	1.65217	1.64461	1.66242	1.66572	1.66595
*****									

Tab. A.12: Alpha activity ratios Pu-238/(Pu-239 + Pu-240) corrected to Nov. 1, 1977 for sample D.

Appendix B:

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