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The ASET-74 Intercomparison Experiment on the Evaluation of Alpha Spectra of Plutonium

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The ASET-74 Intercomparison Experiment on the Evaluation of Alpha Spectra of Plutonium

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The ASET-74 Intercomparison Experiment on the Evaluation of Alpha Spectra of Plutonium

Abstract

Alpha spectra of plutonium with different degrees of peak overlapping were prepared and evaluated by 10 international laboratories. The methods applied were grouped according to the effort involved and the results were compared.

It is shown that significant errors may be introduced by the use of unsatisfactory procedures for peak tail correction and recommendations on suitable methods are made.

The study was performed in the framework of the Safeguards Project of the Federal Republic of Germany, the spectra were prepared by the European Institute for Transuranium Elements (EURATOM).

Das Vergleichsexperiment ASET-74 über die Auswertung von Plutonium Alpha-Spektren

Zusammenfassung

Alpha-Spektren von Plutonium mit verschieden stark überlappenden Peaks wurden hergestellt und zur Auswertung an 10 internationale Laboratorien gegeben. Die dabei verwendeten Methoden wurden entsprechend ihrem Aufwand in Gruppen zusammengefaßt und die Ergebnisse verglichen.

Es wird gezeigt, daß durch Verwendung unzulänglicher Verfahren für die Korrektur der Peaküberlagerungen signifikante Fehler hervorgerufen werden können. Geeignete Auswahlmethoden werden empfohlen.

Die Studie wurde im Rahmen des Projekts Spaltstofflußkontrolle der Bundesrepublik Deutschland durchgeführt, die Spektren wurden im Europäischen Institut für Transurane (EURATOM) hergestellt.

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Acknowledgement and List of Participants

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1. Introduction

For safeguards as well as for commercial reasons exact knowledge is required of the isotopic composition of nuclear materials. As far as destructive analytical methods are applicable, mass spectrometry is, in general, used successfully for this purpose. However, if for any reason uranium and plutonium have to be analyzed in the same mass spectrometer - in particular if there is only one instrument available - alpha spectrometry is preferred by the laboratories for the determination of the Pu 238 isotope because of its interference with U 238.

The IDA-72 interlaboratory comparison experiment [1)] /1/ carried out a few years ago permitted to compare the alpha spectrometric results of 11 laboratories, which were obtained on samples containing plutonium with about 0.005 % and 1.0 % Pu 238. In some cases the agreement was bad and it was assumed that for poorly resolved spectra this may have been caused by the use of unsatisfactory peak tail corrections in spectra evaluation.

For this reason it was proposed to produce a set of spectra with different degrees of peak overlapping and to distribute copies of it for evaluation amongst the laboratories willing to participate in such a test.

According to this suggestion, in summer 1974 the intercomparison experiment "ASET-74" (Alpha-Spectra-Evaluation-Test) was initiated. The necessary spectra were prepared in the laboratories of the European Transuranium Institute of EURATOM in Karlsruhe, Germany, and distributed to 10 laboratories interested in this study. Besides the evaluation of the set of spectra, each laboratory was asked to give a short description of the method applied.

The data reported by the laboratories were evaluated statistically and compiled in a preliminary report. This paper is based on that report and the critical remarks and comments of the participants are taken into account.

[1)] This experiment was mainly concerned with the mass spectrometric isotope dilution technique.

As mentioned before, the poorly resolved spectra used in this experiment were produced on purpose in order to obtain strong overlapping of peaks, which was suitable to prove the performance of the different correction methods under study. There is no doubt that much better conditions can be obtained by the use of targets having thinner layers. However, in the case of Pu 238 abundances of about one percent or more, peak tail correction may become important even for the spectra of targets prepared very carefully.

Two reports on methods of spectra evaluation are given in references /2, 3/ which proved to be very satisfactory in this test.

The complete set of spectra on which this test was based is given in Appendix I. It may be used by laboratories which did not participate in this experiment to check and compare the performance of their own methods of evaluation.

2. Preparation of Samples and their Theoretical Composition

Three plutonium samples A, B and C, containing about 1.5, 0.8 and 0.2 % of Pu 238 respectively, were prepared by mixing precalculated quantities of two Pu solutions (in 8 N - HNO_3) which were calibrated by mass spectrometry combined with isotopic dilution analysis technique [1]. Their isotopic composition together with the concentration of each nuclide (atoms per gram of solution) are reported in Table 2-1.

Tab. 2-1: ASET-74: Concentrations and Isotopic Composition of the Standard Solutions.

Nuclide	Solution 1		Solution 2	
	Concentration (atoms/g sol.)	Isotopic compos. (atom %)	Concentration (atoms/g sol.)	Isotopic compos. (atom %)
Pu 238	4.292 E 16	80.08	0.0	0.0
Pu 239	8.753 E 15	16.33	2.439 E 17	97.13
Pu 240	1.608 E 15	3.00	3.038 E 15	1.21
Pu 241	2.465 E 14	0.46	2.486 E 15	0.99
Pu 242	6.968 E 13	0.13	1.682 E 15	0.67

It can be calculated from the concentration of the different nuclides in each of the two standard solutions that 1 aliquot part of the first solution has to be mixed with 11.182, 21.153 and 85.821 aliquot parts of the second solution in order to get the Pu 238 content mentioned above in samples A, B and C, respectively.

[1] Separation of Am-241 was performed before the isotope dilution analysis. About one month elapsed between this procedure and alpha counting of the targets.

For practical reasons, about 0.1 ml of solution 1 was taken, mixed with about 1.1 ml, 2.1 ml, and 8.5 ml of solution 2 and diluted into about 8.8 ml, 7.8 ml and 11.4 ml of 8 N nitric acid to prepare the sample solutions A, B and C, respectively. The aliquots mentioned above were weighed on an analytical balance. The three samples were then carefully homogenised by shaking; no redox cycle was made.

From the weighing data it was calculated back that one aliquot part of solution 1 was added to 11.0347, 21.0103 and 84.829 aliquot parts of solution 2. The Pu 238 concentrations in the three samples were about 0.17 µg/g of solution for sample A and B and 0.085 for sample C. The calculated isotopic composition of the samples is reported in Table 2-2 below.

Tab. 2-2: ASET-74: Calculated Isotopic Composition of Samples A,B, and C

Nuclide	Isotopic composition (atom %)		
	Sample A	Sample B	Sample C
Pu 238	1.52	0.80_5	0.20_1
Pu 239	95.60	96.32	96.93
Pu 240	1.24	1.23	1.21
Pu 241	0.98	0.98	0.99
Pu 242	0.66	0.66	0.67

3. Preparation of Targets and Spectra Recording

In order to study the influence of tails overlapping on the evaluation of the peak areas, two types of targets (target '1' and target '2') were prepared with source layers of different thicknesses.

In the case of targets '1', some drops of the three sample solutions A, B and C were dropped on stainless steel discs and evaporated. By this way of target preparation, a relative thick source layer is obtained and consequently important tail effects may be expected.

In order to get thin source layers, the electrodeposition method was used to prepare the targets '2'. Again stainless steel discs were employed as supporting material. Spectra of higher quality (lower FWHM) can be obtained with this method and, moreover, the energy degradation of the α-particles, which causes the peak tails, is strongly reduced.

For each of the six targets prepared by the two different methods, three repetition spectra 'I', 'II' and 'III' were taken.

The electronic equipment used to record the spectra, consisted of:

- a silicon semiconductor detector with a resolution of about 20 KeV at 5.5 MeV and a surface of about 50 mm^2,

- an Ortec power supply, pre-amplifier and amplifier,

- an Intertechnique 400 channels pulse-height analyzer.

The spectra were stored during 3600 or 7200 seconds (real time) under vacuum (better than 10^{-2} Torr).

The 18 digital and analogous spectra are given in Appendix I. The different qualities of the two sets of spectra (targets '1' and targets '2'), appear clearly from the analogous spectra.

4. Reduction and Summary of the Data Reported

The individual values for the peak areas of the Pu 238 and (Pu 239+Pu 240) alpha activities reported by the laboratories are compiled in Appendix II. From these figures, the alpha activity ratios of Pu 238/(Pu 239+Pu 240) were calculated as the basic data for all subsequent statistical considerations. The spectra A2 I and B2 III were disregarded as they were obviously disturbed by external noise (electronics or vacuum system).

From the activity ratios of the three[1] repetition spectra taken from each target, the mean values per laboratory were calculated with their standard deviation (SD) and relative standard deviation (RSD) as well as the SD and RSD of the single spectra evaluation[2]. These data are compiled in the upper parts of Tables 4-1 to 4-3. The definitions of the statistical terms are given in Appendix IV.

Comparison of these laboratory mean values shows that meaningful mean values of the activity ratio per target (and the associated SD's and RSD's) can only be obtained after rejection of some outlier values. They were identified by application of the Dixon Criterion[3] /4/ to each group of laboratory mean values belonging to the same target. The same procedure was used to identify excessively high values obtained for the SD's and RSD's [4].

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[1] Two only in case of targets A2 and B2 because of the rejection of the spectra A2 I and B2 III.

[2] For a more detailed interpretation of this term, which can be considered as the "precision", see Sections 5.3 and 5.4.

[3] Here and in all other applications of the Dixon Criterion, a probability of 5 % was allowed for the rejection of a value which belongs to the group. In some cases this probability was considerably lower.

[4] Although the Dixon Criterion was delineated for groups of independent single observations and not for groups of mean values or even standard deviations it seems to be the most suitable procedure also in such cases to decide on the exclusion of outlier values in an objective manner.

Table 4-1: ASET-74: Laboratory and Target Means of Sample A and Associated Data

Column	1	2	3	4	5	6	7	8	9	10	11	12
	Sample A; Target 1						Sample A; Target 2					
Lab. Code	Lab.mean value of activity ratio Pu 238/(Pu 239 + Pu 240)	SD of single spectrum evaluation 1)	RSD of single spectrum evaluation 1) [%]	Number of repetition spectra evaluated	SD of lab.mean value (column 1)	RSD of lab.mean value (column 1) [%]	Lab.mean value of activity ratio Pu 238/(Pu 239 + Pu 240)	SD of single spectrum evaluation 1)	RSD of single spectrum evaluation 1) [%]	Number of repetition spectra evaluated	SD of lab.mean value (column 7)	RSD of lab.mean value (column 7) [%]
	$A_{11k\cdot}$	σ_{11k}	s_{11k}	n	$\sigma(M)_{11k}$	$s(M)_{11k}$	$A_{12k\cdot}$	σ_{12k}	s_{12k}	n	$\sigma(M)_{12k}$	$s(M)_{12k}$
1	3.662	±.301	± 8.21	3	±.174	± 4.74	(4.391)	(±.423)	(±9.63)	2	(±.299)	(±6.81)
2	4.000	±.070	± 1.76	3	±.040	± 1.02	3.960	±.057	±1.44	2	±.040	±1.02
3	3.913	±.050	± 1.27	3	±.029	± .73	3.910	±.086	±2.20	2	±.061	±1.56
4	3.289	±.044	± 1.34	3	±.025	± .77	3.885	±.096	±2.46	2	±.068	±1.74
5	3.571	±.206	± 5.76	3	±.119	± 3.33	3.912	±.087	±2.21	2	±.061	±1.56
6	(5.631)	±.178	± 3.16	3	±.103	± 1.82	4.011	±.098	±2.44	2	±.069	±1.73
7	3.983	±.102	± 2.55	3	±.059	± 1.47	3.918	±.094	±2.39	2	±.066	±1.69
8	3.706	±.063	± 1.70	3	±.036	± .98	3.935	±.106	±2.69	2	±.075	±1.90
9	4.004	±.043	± 1.06	3	±.025	± .61	3.971	±.079	±2.00	2	±.056	±1.41
10	4.268	±.066	± 1.53	3	±.038	± .88	3.956	±.078	±1.96	2	±.055	±1.39
Target mean $\bar{A}_{11\cdot\cdot}$; $\bar{\sigma}_{11(G)}$; $\bar{s}_{11(G)}$	3.821_8	$\pm.139_5$ 2)	$\pm 3.65_0$ 2)			$\bar{A}_{12\cdot\cdot}$; $\bar{\sigma}_{12(G)}$; $\bar{s}_{12(G)}$	3.939_8	$\pm.087_8$ 2)	$\pm 2.22_5$ 2)			
SD of single lab.mean σ_{11}	±.292					σ_{12}	±.039					
RSD of single lab.mean s_{11}	± 7.65					s_{12}	±.98					
SD of target mean $\sigma(M)_{11}$	±.098					$\sigma(M)_{12}$	±.013					
RSD of target mean $s(M)_{11}$	± 2.55					$s(M)_{12}$	±.33					

Note: Values in brackets were not used for calculating target means and associated data.

1) This term can be considered as "precision" /Par. 5.3 and 5.4/.

2) For calculation see App. IV, Par.2 (4) and (5).

Table 4-2: ASET-74: Laboratory and Target Means of Sample B and Associated Data

Column	1	2	3	4	5	6	7	8	9	10	11	12
	Sample B; Target 1						Sample B; Target 2					
Lab. Code	Lab.mean value of activity ratio Pu 238/Pu 239+Pu 240)	SD of single spectrum evaluation	RSD of single spectrum 1) evaluation [%]	Number of repetition spectra evaluated	SD of lab.mean value (column 1)	RSD of lab.mean value (column 1) [%]	Lab.mean value of activity ratio Pu 238/Pu 239+Pu 240)	SD of single spectrum 1) evaluation	RSD of single spectrum 1) evaluation [%]	Number of repetition spectra evaluated	SD of lab.mean value (column 7)	RSD of lab.mean value (column 7) [%]
	$A_{21k.}$	σ_{21k}	s_{21k}	n	$\sigma(M)_{21k}$	$s(M)_{21k}$	$A_{22k.}$	σ_{22k}	s_{22k}	n	$\sigma(M)_{22k}$	$s(M)_{22k}$
1	2.321	(±.436)	(±18.80)	3	(±.252)	(±10.85)	(2.169)	±.018	±.82	2	±.013	±.58
2	2.283	±.024	±1.04	3	±.014	±.60	2.109	±.006	±.27	2	±.004	±.19
3	2.162	±.018	± .82	3	±.010	±.47	2.090	±.005	±.22	2	±.004	±.16
4	2.087	±.024	±1.17	3	±.014	±.68	2.086	±.008	±.38	2	±.006	±.27
5	2.123	±.030	±1.43	3	±.017	±.83	2.087	±.014	±.66	2	±.010	±.47
6	(2.607)	±.042	±1.61	3	±.024	±.93	2.121	±.004	±.19	2	±.003	±.13
7	2.111	±.018	±.87	3	±.010	±.50	2.097	±.009	±.43	2	±.006	±.30
8	2.011	±.041	±2.02	3	±.024	±1.17	2.104	±.006	±.29	2	±.004	±.21
9	No data reported						2.092	±.025	±1.18	2	±.018	±.83
10	2.129	±.023	±1.07	3	±.013	±.62	2.104	±.007	± .31	2	±.005	±.22
Target mean $A_{21..}$; $\bar{\sigma}_{21(G)}$; $\bar{s}_{21(G)}$	2.153_4	$\pm.028_9$ 2)	$\pm1.34_2$ 2)			$A_{22..}$; $\bar{\sigma}_{22(G)}$; $\bar{s}_{22(G)}$	2.098_9	$\pm.012_0$ 2)	$\pm .57_4$ 2)			
SD of single lab.mean σ_{21}	±.102					σ_{22}	±.012					
RSD of single lab.mean s_{21}	±4.74					s_{22}	±.55					
SD of target mean $\sigma(M)_{21}$	±.036					$\sigma(M)_{22}$	±.004					
RSD of target mean $s(M)_{21}$	±1.68					$s(M)_{22}$	±.18					

Note: Values in brackets were not used for calculating target means and associated data.

1) This term can be considered as "precision" /Par. 5.3 and 5.4/.

2) For calculation see App. IV, Par. 2 (4) and (5).

Table 4-3: ASET-74: Laboratory and Target Means of Sample C and Associated Data

Column	1	2	3	4	5	6	7	8	9	10	11	12
		Sample C; Target 1						Sample C, Target 2				
Lab. Code	Lab.mean value of activity ratio Pu 238/(Pu 239 + Pu 240)	SD of single spectrum evaluation 1)	RSD of single spectrum evaluation 1) [%]	Number of repetition spectra evaluated	SD of lab.mean value (column 1)	RSD of lab.mean value (column 1) [%]	Lab.mean value of activity ratio Pu 238/(Pu 239 + Pu 240)	SD of single spectrum evaluation 1)	RSD of single spectrum evaluation 1) [%]	Number of repetition spectra evaluated	SD of lab.mean value (column 7)	RSD of lab.mean value (column 7) [%]
	$A_{31k\cdot}$	σ_{31k}	s_{31k}	n	$\sigma(M)_{31k}$	$s(M)_{31k}$	$A_{32k\cdot}$	σ_{32k}	s_{32k}	n	$\sigma(M)_{32k}$	$s(M)_{32k}$
1	(.4832)	(±.0762)	(±15.77)	3	(±.0440)	(±9.10)	(.5278)	(±.0412)	(±7.81)	3	(±.0238)	(±4.51)
2	.5794	±.0015	± .26	3	±.0009	± .16	.5654	±.0042	± .74	3	±.0024	± .43
3	.5480	±.0058	± 1.06	3	±.0033	± .61	.5580	±.0019	± .34	3	±.0011	± .20
4	.5441	±.0026	± .48	3	±.0015	± .28	.5604	±.0022	± .39	3	±.0013	± .23
5	.5594	±.0034	± .61	3	±.0020	± .35	.5661	(±.0076)	(±1.34)	3	(±.0044)	(± .77)
6	.6131	±.0023	± .38	3	±.0013	± .21	.5671	±.0025	± .44	3	±.0014	± .25
7	.5559	±.0027	± .49	3	±.0016	± .28	.5609	±.0022	± .39	3	±.0013	± .23
8	.5384	±.0066	± 1.23	3	±.0038	± .70	.5621	±.0034	± .60	3	±.0020	± .35
9	.5589	±.0027	± .48	3	±.0016	± .28	.5596	±.0040	± .71	3	±.0023	± .41
10	.5589	±.0020	± .36	3	±.0012	± .21	.5614	±.0002	± .04	3	±.0001	± .02
Target mean $A_{31\cdot\cdot}$; $\bar{\sigma}_{31(G)}$; $\bar{s}_{31(G)}$	$.5617_8$	$\pm.0036_8$ 2)	$\pm .65_4$ 2)			$A_{32\cdot\cdot}$; $\bar{\sigma}_{32(G)}$; $\bar{s}_{32(G)}$	$.5623_3$	$\pm.0028_4$ 2)	$\pm.50_6$ 2)			
SD of single lab.mean σ_{31}	±.0225					σ_{32}	±.003					
RSD of single lab mean s_{31}	± 4.00					s_{32}	± .55					
SD of target mean $\sigma(M)_{31}$	±.0075					$\sigma(M)_{32}$	±.001					
RSD of target mean $s(M)_{31}$	± 1.33					$s(M)_{32}$	± .18					

Note: Values in brackets were not used for calculating target means and associated data.

1) This term can be considered as "precision" /Par. 5.3 and 5.4/.

2) For calculation see App. IV, Par.2 (4) and (5).

The target mean values and the associated SD's and RSD's, - calculated after rejection of the outlier values put in brackets - are compiled in the lower part of the tables.

A graphical survey of these data is given in Figs. 4-1 to 4-6 which present separately for each target and sample the relative deviations of the laboratory means from the target mean values.

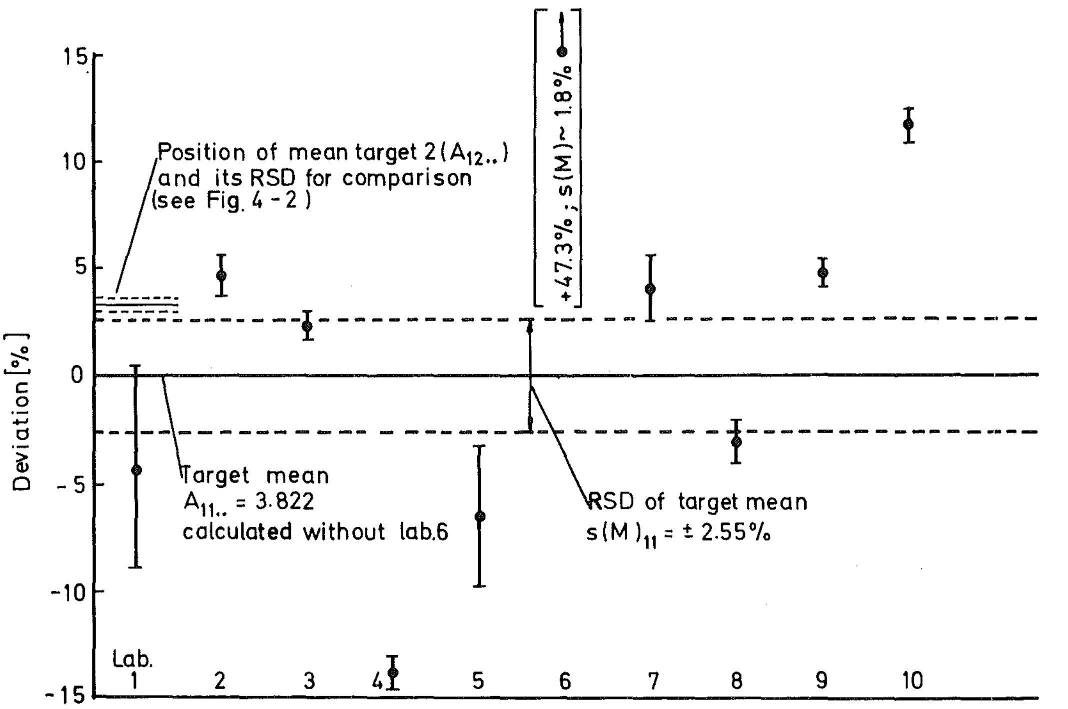
From the results shown in these tables and figures, the following observations and statements can be made:

- The supposed effect of peak overlapping on the accuracy of evaluation is clearly confirmed: the spread of the laboratory means around the target mean decreases with decreasing Pu 238 content from sample A (1.5 %) to sample C (0.2 %) and is distinctly smaller for spectra obtained from targets prepared by electrodeposition ("2") than by evaporation ("1").

- Even typical "outlier" values may be caused by unsatisfactory evaluation procedures.

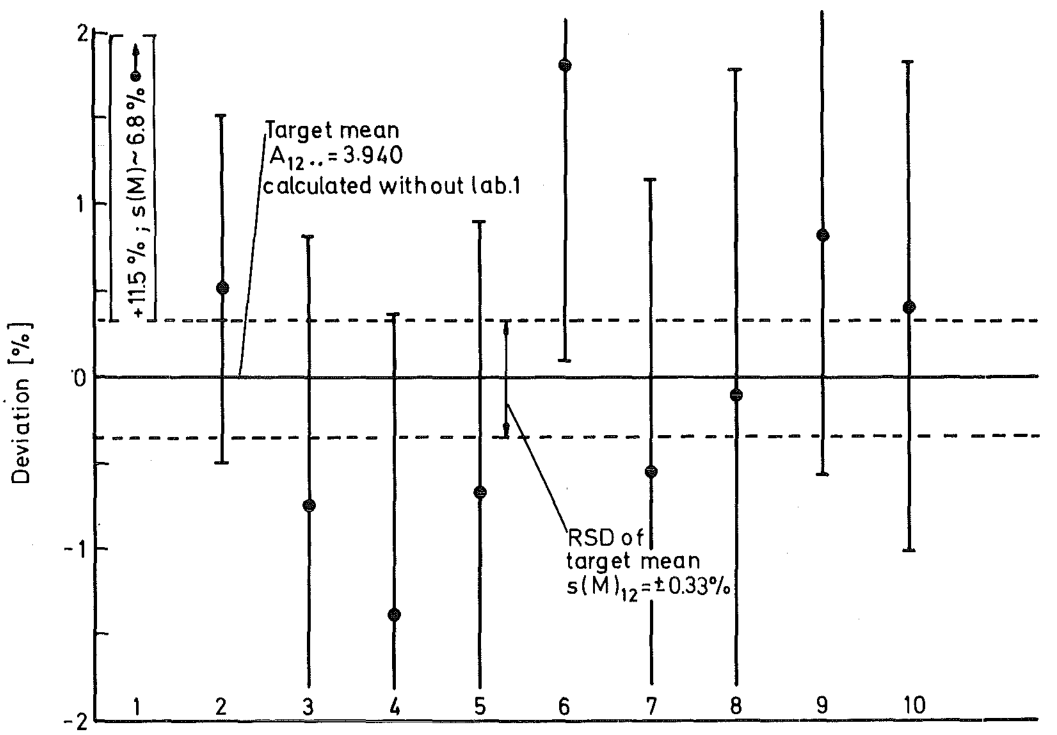
- For samples A and B with the higher Pu 238 content significant differences in the target mean values of the two preparation methods seem to exist. However, the opposite signs of these deviations for samples A and B indicate that they do not reflect any realistic effect. Furthermore, as it will be shown later /Section 5.2/, the significance of the discrepancies is considerably reduced if those laboratory mean values are excluded which were obtained by unsuitable methods of evaluation.

  In case of sample C there is an excellent agreement of both target means /Fig. 4-5/.



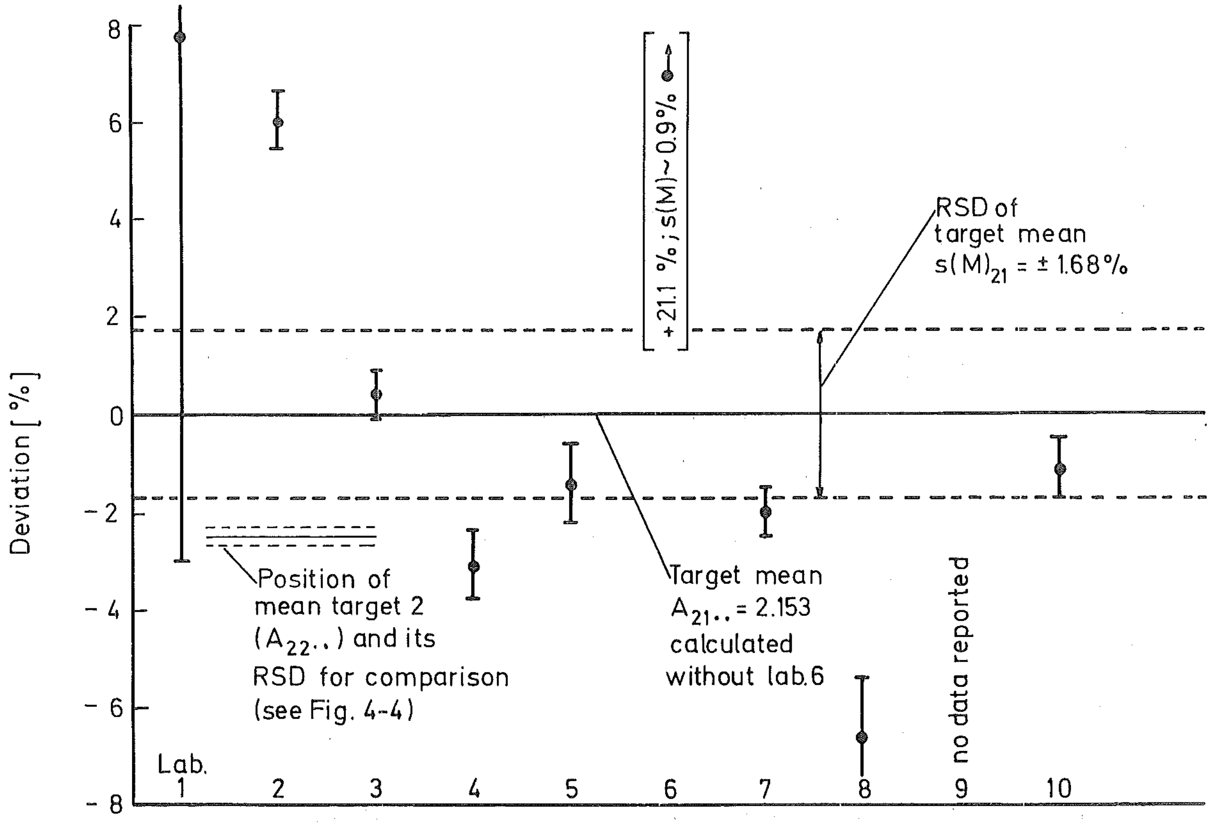
(Laboratory mean values $A_{11k.}$; error bars indicate RSD of these means $s(M)_{11k}$)

Fig. 4 - 1 ASET - 74 α-Activity Ratios Pu238/(Pu239 + 240) of Sample A, Target 1



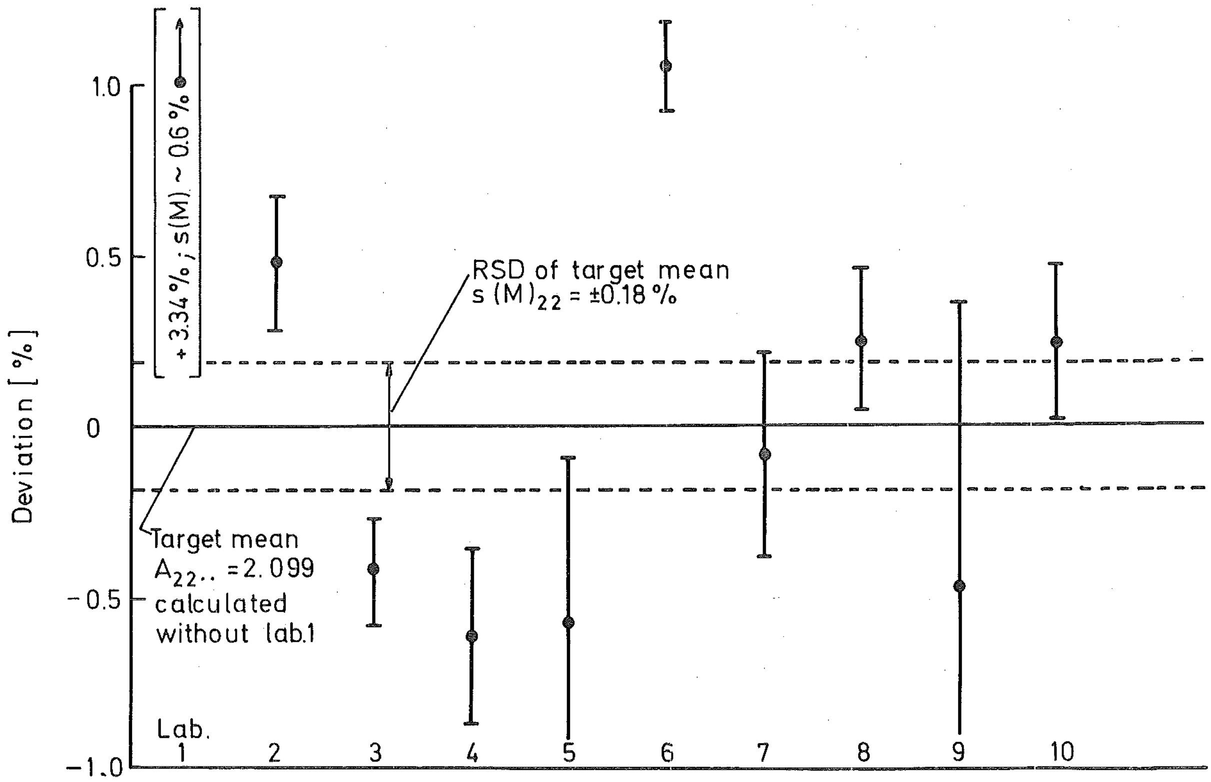
(Laboratory mean values $A_{12k.}$; error bars indicate RSD of these means $s(M)_{12k}$)

Fig. 4 - 2 ASET - 74 α-Activity Ratios Pu238/(Pu239 + Pu240) of Sample A, Target 2



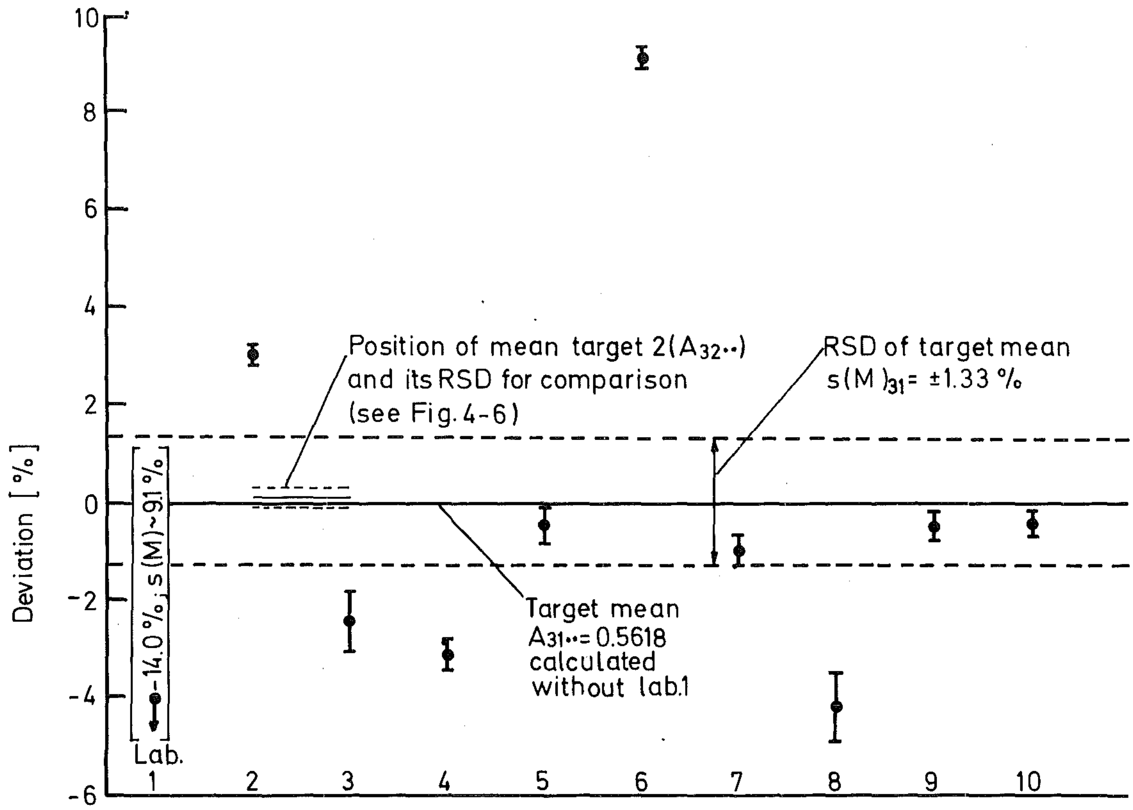
(Laboratory mean values $A_{21k.\,i}$ error bars indicate RSD of these means $s(M)_{21k}$)

Fig. 4 - 3 ASET - 74: α-Activity Ratios Pu238/(Pu239 + Pu240) of Sample B, Target 1



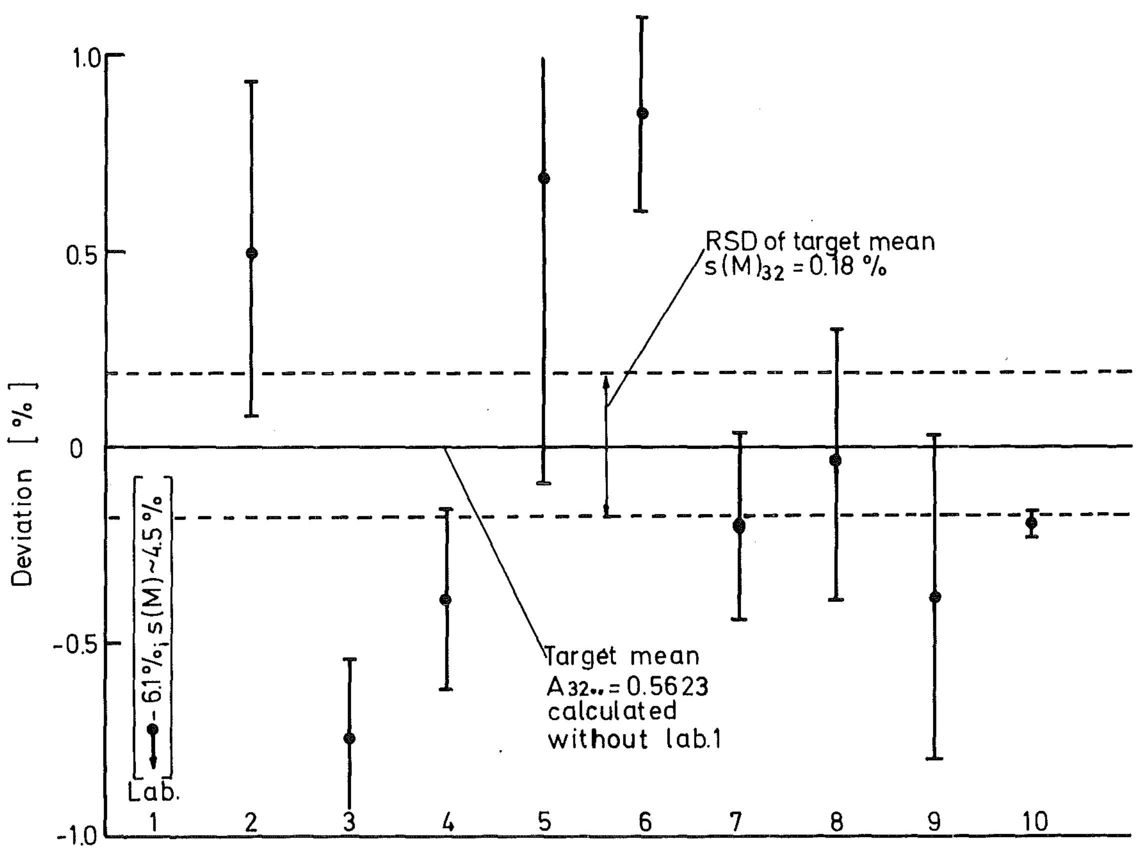
(Laboratory mean values $A_{22k.\,i}$ error bars indicate RSD of these means $s(M)_{22k}$)

Fig. 4-4 ASET - 74: α-Activity Ratios Pu238/(Pu239 + Pu240) of Sample B, Target 2



(Laboratory mean values $A_{31k\cdot i}$ error bars indicate RSD of these means $s(M)_{31k}$)

Fig. 4-5 ASET-74: α-Activity Ratios Pu 238/(Pu 239+Pu 240) of Sample C, Target 1



(Laboratory mean values $A_{32k\cdot\cdot i}$ error bars indicate RSD of these means $s(M)_{32k}$)

Fig. 4-6 ASET - 74: α- Activity Ratios Pu 238/(Pu 239 + Pu 240) of Sample C, Target 2

5. Discussion of the Results

5.1 Methods of Spectra Evaluation Used and Tentative Appreciation

Each laboratory participating in the experiment was asked to give a short description of the method used for the evaluation of the alpha spectra. The descriptions of the methods are presented in Appendix III as reported by the laboratories.

It is evident from the survey of the methods reported that each laboratory according to its experience and to the quality of the spectra uses its own method which differs to some extent from the methods used by other laboratories. This confirms that practically no standard methods are available to evaluate alpha spectra, especially for the spectra obtained from evaporated sources.

The methods used in this experiment were divided into three groups, which exclusively served the purpose of contributing to a selection of some methods susceptible of giving in every case, i.e. independent of the quality of the spectrum, more reliable results:

- i) Simple methods,
- ii) normal methods,
- iii) elaborated methods.

The main features of each group are:

Simple methods: - The peak area is obtained by summing up the channel content between predetermined channels. The correction for the tail effect of the Pu 238 peak is very poor or is not performed at all.

Normal methods: - The peak area is evaluated more or less as in case I. The contribution of the tail to the preceding peak is taken into account and corrected in a more efficient way.

Elabor. methods:- To evaluate the peak area and to make corrections for the overlapping of tails, non-linear least squares fitting procedures are used.

Regardless of the results obtained by each laboratory and according to the general features defined above the methods were grouped as in Table 5-1:

Tab. 5 - 1: ASET-74: Grouping of the Methods Used for Evaluating the Alpha Spectra

Method	Laboratory Code
SIMPLE	1, 4, 6
NORMAL	2, 8, 9, 10
ELABORATED	3, 5, 7

The analytical results obtained by the laboratories make evident that the goodness of each group of methods and their relative appreciation should appear also from the reported single values of the Pu 238/(Pu 239+ Pu 240) activity ratio and from some statistical evaluations on the values obtained by pooling the results of each group of methods. The following considerations confirm that this grouping of the methods is meaningful:

- The 6 laboratory mean values of the Pu 238/(Pu 239 + Pu 240) activity ratio considered as outliers belong to the group defined as "simple" method /Tabs. 4-1 to 4-3/.

- Considering the means of the relative standard deviations of single spectra evaluation[1] /Tab. 5-2/ for the three groups of methods results for 5 of the 6 targets in higher RSD's obtained by the group of "simple" methods than by the others.

- Influence of the tails and the worth of the correction applied are indicated for each laboratory by the differences of the Pu 238/(Pu 239 + Pu 240) activity ratios obtained for the two targets of the same sample. For each sample and each group of methods the mean of these relative differences is given in Table 5-3. It is evident from the analysis of these data that for a given sample the difference of the activity ratios between the two targets varies according to the appreciation of the group of method. These data give, essentially, an idea of the quality of the corrections applied to the overlapping of tails. Moreover, looking at the results for a given group of methods, it can be concluded that, in general, the influence of tails decreases with decreasing Pu 238 content from sample A (1.5 %) to sample C (0.2 %), which was expected.

The same conclusions on the appreciation of the group of methods can be drawn from the data reported in Table 5-4. They show for each group and for each target the mean of the relative deviations of the laboratory means from the target mean, the latter calculated after rejection of the 6 outliers as in Tables 4-1 to 4-3.

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[1] This term can be considreed as "precision" /Sections 5.3 and 5.4/.

Tab. 5-2: ASET-74: Means of the RSD's of Single Spectra Evaluation ("Precision") for the Groups of Evaluation Methods

Sample	A(i=1)		B(i=2)		C(i=2)	
Target j	1	2	1	2	1	2
Group of Methods	$\bar{s}_{11}(G)$ [%]	$\bar{s}_{12}(G)$ [%]	$\bar{s}_{21}(G)$ [%]	$\bar{s}_{22}(G)$ [%]	$\bar{s}_{31}(G)$ [%]	$\bar{s}_{32}(G)$ [%]
SIMPLE (G:Lab.1,4,6)	5.32	6.51	11.77	.55	7.84	4.25
NORMAL (G:Lab.2,8,9,10)	1.60	2.08	1.40	.64	.67	.59
ELABORATED (G:Lab.3,5,7)	3.55	2.25	1.06	.47	.75	.83

Tab. 5-3: ASET-74: Relative Difference of the Activity Ratios Obtained for Targets 1 and 2; Mean Values for the Groups of Evaluation Methods [%]

Sample	A(i=1)	B(i=2)	C(i=3)
Group of Methods	$\frac{100}{t}\sum_{G}\left\lvert\frac{A_{11k\cdot}-A_{12k\cdot}}{A_{12k\cdot}}\right\rvert$	$\frac{100}{t}\sum_{G}\left\lvert\frac{A_{21k\cdot}-A_{22k\cdot}}{A_{22k\cdot}}\right\rvert$	$\frac{100}{t}\sum_{G}\left\lvert\frac{A_{31k\cdot}-A_{32k\cdot}}{A_{32k\cdot}}\right\rvert$
SIMPLE (G:Lab.1,4,6)	24.11	9.99	6.49
NORMAL (G:Lab.2,8,9,10)	3.89	4.62	1.82
ELABORATED (G:Lab.3,5,7)	3.48	1.95	1.29

Tab. 5-4: ASET-74: Relative Difference between Laboratory and Target Mean; Mean Values for the Groups of Evaluation Methods [%]

Sample	A(i=1)		B(i=2)		C(i=3)	
Target j =	1	2	1	2	1	2
Group of Methods	$\frac{100}{t \cdot A_{1j..}} \sum_G \lvert A_{1jk.} - A_{1j..} \rvert$		$\frac{100}{t \cdot A_{2j..}} \sum_G \lvert A_{2jk.} - A_{2j..} \rvert$		$\frac{100}{t \cdot A_{3j..}} \sum_G \lvert A_{3jk.} - A_{3j..} \rvert$	
SIMPLE (G:Lab.1,4,6)	21.82	4.88	10.64	1.67	8.76	2.44
NORMAL (G:Lab.2,8,9,10)	6.03	.46	4.59	.32	2.08	.31
ELABORATED (G:Lab.3,5,7)	4.39	.67	1.26	.36	1.31	.56

It can be concluded form these considerations that by the methods of the first group (simple methods) unsatisfactory results are obtained quite easily. As indicated by the data given in Table 5-4, this is also true for targets prepared by electrodeposition, in particular at high Pu 238 concentration.

By contrast, no important difference exists, in general, between the normal and elaborated groups of methods, especially for high-resolution spectra. Only some of the data given in Tabs. 5-3 and 5-4 indicate a superiority of the elaborated methods compared to the normal ones.

5.2 Comparison between Experimental and Theoretical Values

Due to the overlapping of the alpha particle energies of the Pu 239 and Pu 240 nuclides, only the activity ratio between Pu 238 and (Pu 239+Pu 240) can be obtained by the alpha spectrometric technique. Consequently, in order to compare theoretical and experimental values, it is necessary to calculate the theoretical activity ratio. Moreover, for safeguards applications it is important to compare also the Pu 238/Pu 239 isotopic ratio, which can be obtained from the experimental data of the activity ratio. The formulae for these calculations are compiled in Appendix IV, Section 5.

The values of the specific activities and those of the half-lives used are reported in Table 5-5[1].

Tab. 5-5: ASET-74: Nuclear Data Used for Calculations

Nuclide	Specific activity [d.p.m/atom]	Half-life [years]
Pu 238	1.5027 E-8	87.7
Pu 239	5.4013 E-11	2.44 E 4
Pu 240	2.0029 E-10	6.58 E 3

[1] These values were recommended by the participants of the IDA-72 experiment and used for its evaluation /Ref. 1, Vol. I, Section 8.3.5 and Vol. II, Sect. 1.3/. They differ only slightly from those recommended more recently /5/.

In Table 5-6 the theoretical and experimental values of the Pu 238/(Pu 239+Pu 240) activity and of the Pu 238/Pu 239 isotopic ratios are compiled together with their relative deviations.

With respect to the considerations above /Section 5-1/ the experimental data were calculated excluding the results of the laboratories 1, 4 and 6 which belong to the group of "simple" evaluation methods. After exclusion of these values there is no more indication that the target mean values belonging to the same sample have to be considered as significantly different. As shown in the table, their error ranges defined by their relative standard deviations overlap or at least touch each other. It is for this reason that for comparison with the theoretical data the mean value per sample has been calculated.

As can be seen from the figures given on the last line of the table, some discrepancies exist between the experimental and the theoretical values. These deviations decrease with decreasing Pu 238 content of the sample. However, no dependence is indicated on the type of target, i.e. on the degree of peak overlapping, because of the good agreement of the two target mean values calculated for each sample. Therefore, it seems reasonable to assume that the deviations are due to uncertainties of the theoretical values which are affected by at least four sources of error :

- the inaccuracy associated with the isotopic concentrations of the two standard solutions A and B determined by isotopic dilution analysis;
- the errors involved in the weighing data of the different aliquots of the two solutions;
- the accuracy of the half-life values;and
- the degree of homogeneity of the sample solutions.

Tab. 5-6: ASET-74: Theoretical and Experimental Values of the Activity and Atom Ratios

Sample	A (i = 1)		B (i = 2)		C (i = 3)	
Target j =	1	2	1	2	1	2
Exp. Pu 238/(Pu 239+ Pu 240) activity ratio; Target mean $A_{ij..} \pm \sigma(M)_{ij}$	3.921 ± .085	3.937 ± .010	2.137 ± .036	2.098 ± .003	0.5570 .0047	0.5619 ± .0011
Exp. Pu 238/(Pu 239+ Pu 240) activity ratio; Sample mean $A_{i...}$	3.927		2.119		0.5595	
Theor. Pu 238/(Pu 239+Pu 240) activity ratio	4.220		2.220		0.5514	
Exp. Pu 238/ Pu 239 atom ratio	0.01479		0.007977		0.002104	
Theor. Pu 238/Pu 239 atom ratio	0.01590		0.008357		0.002074	
Relative deviation exp.- theor. value [%]	- 6.94		- 4.55		+ 1.47	

Note: Values of Labs.1, 4 and 6 were not used for the calculation of the experimental data.

It is very complicated to estimate the combined influence of the single errors on the overall accuracy of the theoretical values. For example, the error associated with the Pu 238 half-life value contributes by different amounts to the activity ratios of the three samples. Moreover, for the fourth source of error it is not possible to give any estimation of error.

Taking into account these different sources of error it seems to be impossible to explain on the basis of this experiment the differences observed between the theoretical and experimental data.

5.3 Estimation of Error Components

In the following paragraphs an attempt is made to estimate quantitatively the error contributions of spectra recording and their evaluation to the total error of alpha spectrometric determinations.

For these considerations three error components are distinguished:

- First, the "counting component" which represents the spread in the Pu 238/(Pu 239 + Pu 240) ratio of counts if several repetition spectra are taken from the same target.

- Secondly, the "evaluation component" describing the spread of the results if the same spectrum is evaluated several times by the same method. Assuming graphical evaluation of an analogous spectrum, the meaning of this component is obvious. In the case of non-computerized evaluation of a digital spectrum it is caused by different assigning of counts to the Pu 238 or (Pu 239 + Pu 240) peak from one evaluation to the next (e.g. evaluation of the same spectrum by different persons). However, when the spectrum evaluation is completely computerized, this error component is found to be zero. The corresponding error now adds to the bias of the method and contributes to the third error component considered.

- This third error component corresponds to the interlaboratory deviation when measurement results are compared. Since in this test the differences in the results of the laboratories are caused by the application of different evaluation methods, the expression "between methods deviation" seems to be more suitable for this study.

These error components will be denoted s(c), s(e) and s(b), respectively, and are expressed in terms of RSD.

The "counting component" and the "evaluation component" together can be considered as the "precision" of an alpha activity ratio determination performed within an individual laboratory, since it describes the reproducibility observed if several repetition spectra are taken from the same target and each repetition spectrum is evaluated once.

The RSD of its estimate is given by $(s(c)^2 + s(e)^2)^{1/2}$ and corresponds to the value s_{ijk} in Tabs. 4-1 to 4-3.

The structure of the experiment does not allow to calculate by one single analysis of variances the estimates for all three types of errors. However, by analysis of variances for two types of error, the estimate s(b) for the RSD of the "between methods deviation" and the estimate for the RSD of the sum $(s(c)^2 + s(e)^2)^{1/2}$ of the two other error components, i.e. for the "precision" /App. IV, Section 6/, can be obtained.

The values calculated for the various samples and targets are given in Tab. 5-7, columns 1 and 2[1)].

[1)] These and the following calculations are based on the values obtained by the laboratories using a "normal" or "elaborated" evaluation method /Section 5.1/.

Splitting of the precision into its two components "counting" and "evaluation" becomes possible if data on the spread of the alpha activity ratios from one repetition to the next can be obtained which are not influenced by any evaluation error and therefore represent the "counting component" alone. This can be done by calculation of the channel content of that fraction of the Pu 238 peak which does not overlap the (Pu 239 + Pu 240) peak related to the total channel content of all peaks. The channel of the Pu 238 peak selected for these calculations and the corresponding ratios of counts Pu 238/(Pu 239 + Pu 240) are compiled in Table 5-8. Then the RSD for a single spectrum was calculated from the three repetition spectra taken on each target[2)] and can be considered as a good estimate of the counting component s(c). These values are given in column 3 of Tab. 5-8.

Tab. 5-7: ASET-74: Calculated Estimates for the RSD's of Error Components

Column		1	2	3	4
Sample i	Target j	RSD of "between methods deviation" $s_{ij}(b)$ [%]	RSD of "precision" $(s_{ij}(c)^2+s_{ij}(e)^2)^{1/2}$ [%]	RSD of "counting component" $s_{ij}(c)$ [%]	RSD of "evaluation component" $s_{ij}(e)$ [%]
A(i=1)	1	5.57	2.56	.64	2.48
A(i=1)	2	not significant	2.15	.35	2.12
B(i=2)	1	4.07	1.12	.27	1.09
B(i=2)	2	not significant	.58	.14	.56
C(i=3)	1	2.21	.71	.27	.66
C(i=3)	2	0.31	.71	.25	.66

Note: Calculations are based on all data except those obtained by Labs.1,4 and 6

2) Two only in case of targets A2 and B2 /Section 4/.

Finally, in column 4 the data for the "evaluation component" are given, which are calculated by appropriate subtraction of the RSD of the counting component from that of the precision.

By comparison of the data compiled in Tab. 5-7 the following observations can be made:

- The deviations between methods are the main error contribution in case of targets prepared by evaporation ("1") but nearly insignificant for electrodeposition targets ("2").
- The two components contributing to the precision (counting and evaluation) are of the same order of magnitude for samples B and C. Only for high Pu 238 content (sample A), the counting component becomes negligible.
- For the counting component, higher values are obtained for the targets prepared by evaporation ("1") than for those made by electrodeposition ("2").

Tab. 5-8: ASET-74: Channels of Pu 238 Peak Selected for Estimation of Counting Error and Ratio of their Content to Total Counts

Sample	Target	Spectrum	Channels selected for Pu 238 counts	Ratio of Pu 238 counts in selected channels to total counts
A	1	I	205 - 330	.705
		II	210 - 330	.697
		III	209 - 330	.704
A	2	II	212 - 330	.7913
		III	212 - 330	.7952
B	1	I	210 - 350	.6330
		II	212 - 350	.6364
		III	208 - 350	.6346
B	2	I	210 - 330	.6739
		II	210 - 330	.6726
C	1	I	210 - 330	.3420
		II	210 - 330	.3432
		III	212 - 330	.3437
C	2	I	210 - 330	.3559
		II	212 - 330	.3576
		III	210 - 330	.3573

5.4 Precision of the Alpha Activity Ratio Determinations as a Function of their Absolute Value

According to the previous consideration /Section 5.2/ the precision of an alpha activity ratio determination performed in one individual laboratory is understood as the reproducibility observed if several repetition spectra are taken from the same target and each repetition spectrum is evaluated once. The average values for each target were calculated in Section 5.3 and compiled in Tab. 5-1, column 2. The theoretical values of the alpha activity ratios themselves were calculated in Section 5.2 and compiled in Tab. 5-6.

In Fig. 5-1 these data are plotted and connected by straight lines for each type of target preparation. This presentation clearly confirms the loss in precision with increasing overlapping of peaks.

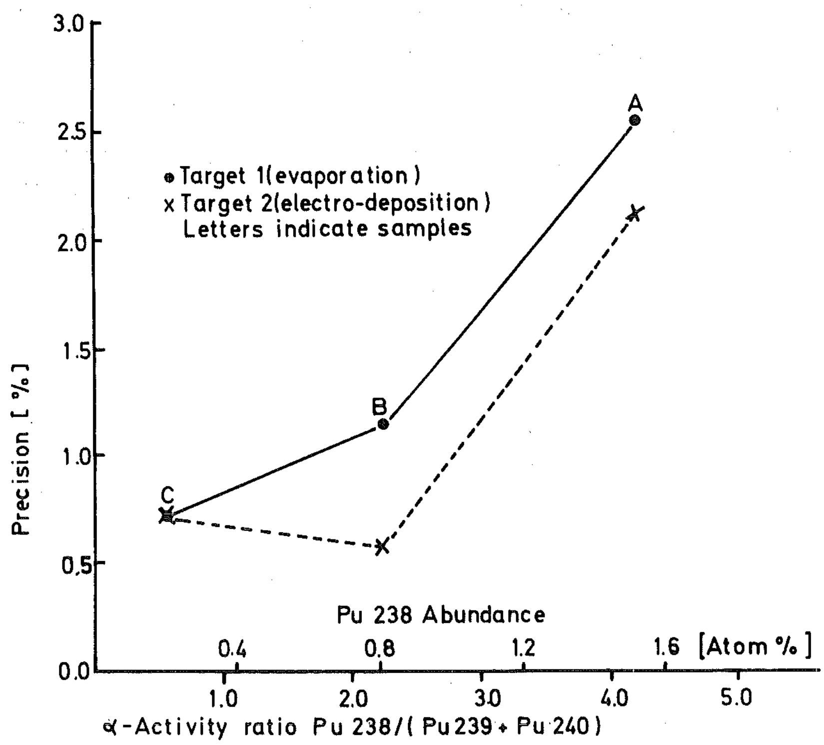

Fig. 5-1 ASET-74: Precision of α-Activity Ratio Determination Pu238/(Pu239 + Pu240)
(Errors of target preparation not included)

6. Summary and Conclusion

The results of this experience confirm clearly the strong influence of peak tail overlapping on the Pu 238/(Pu 239+ Pu 240) alpha activity ratio obtained. Considerable errors can be introduced if unsatisfactory procedures are applied for its correction.

Depending on their performance, three groups of evaluation methods could be distinguished:

Methods belonging to the group of the simplest ones should not be used. At least for Pu 238 abundances of about 1 % and more they may fail even if peak tail overlapping is minimized by the use of very carefully prepared targets.

With the methods of medium complexity satisfactory results can be obtained in the whole concentration range of Pu 238 investigated (up to about 1.5 %) if optimum peak resolution is ensured by the use of suitable methods for the preparation of the targets.

The most elaborated methods which include non-linear least squares fitting procedures are of the highest performance and recommended. However, if applied, they should be computerized to keep the effort within acceptable limits.

Significant differences are observed between the theoretical values of the alpha activity ratios and those obtained in this test. These deviations decrease with decreasing Pu 238 content. Their explanation is not possible from the data of this experiment but would require more extensive studies.

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/3/ S.K. Aggarwal, H.C. Jain, C.K. Mathews and M.V. Ramaniah
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/4/ W.J. Dixon
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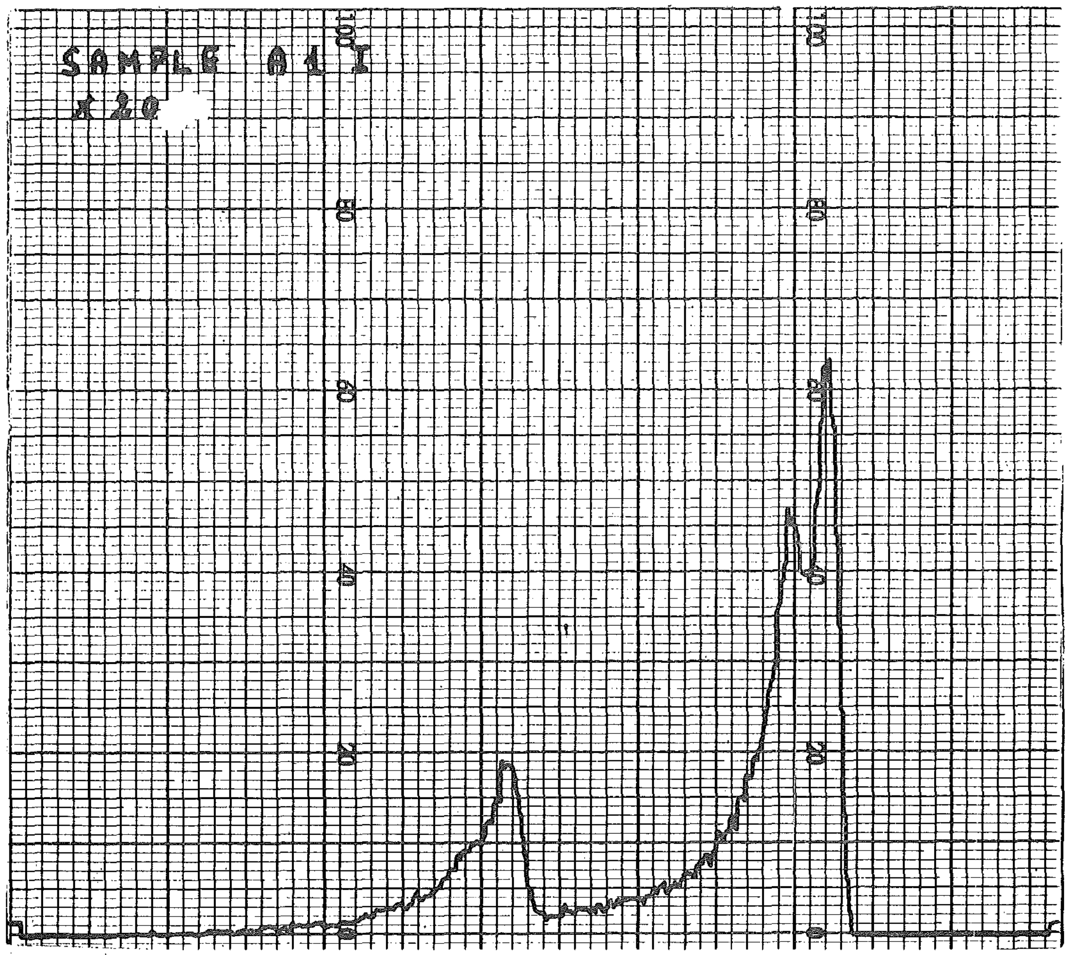
"The Half-Life of some Long-Lived Actinides: A Compilation"
EUR 5194e (1974)

APPENDIX I:

Distributed Alpha Spectra
(Analoguous and Digital)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80
60
40
20
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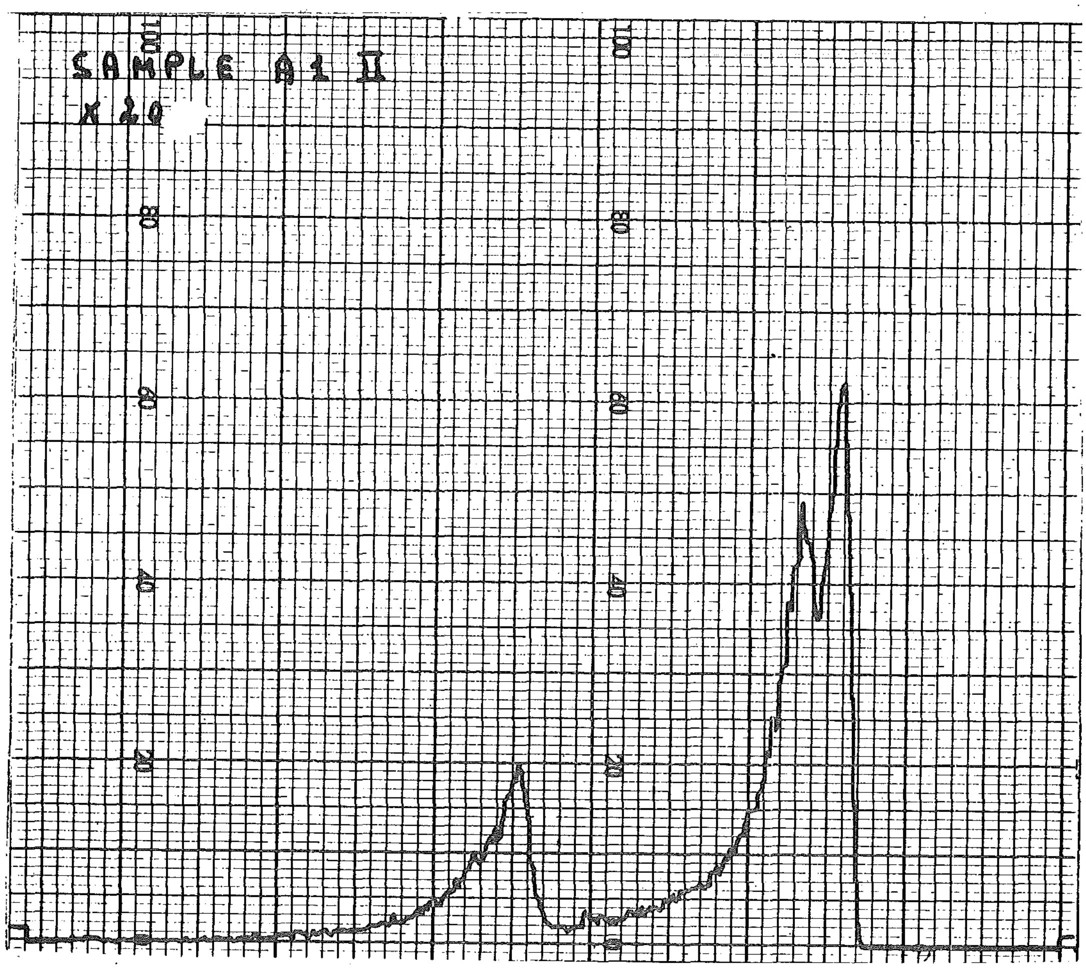
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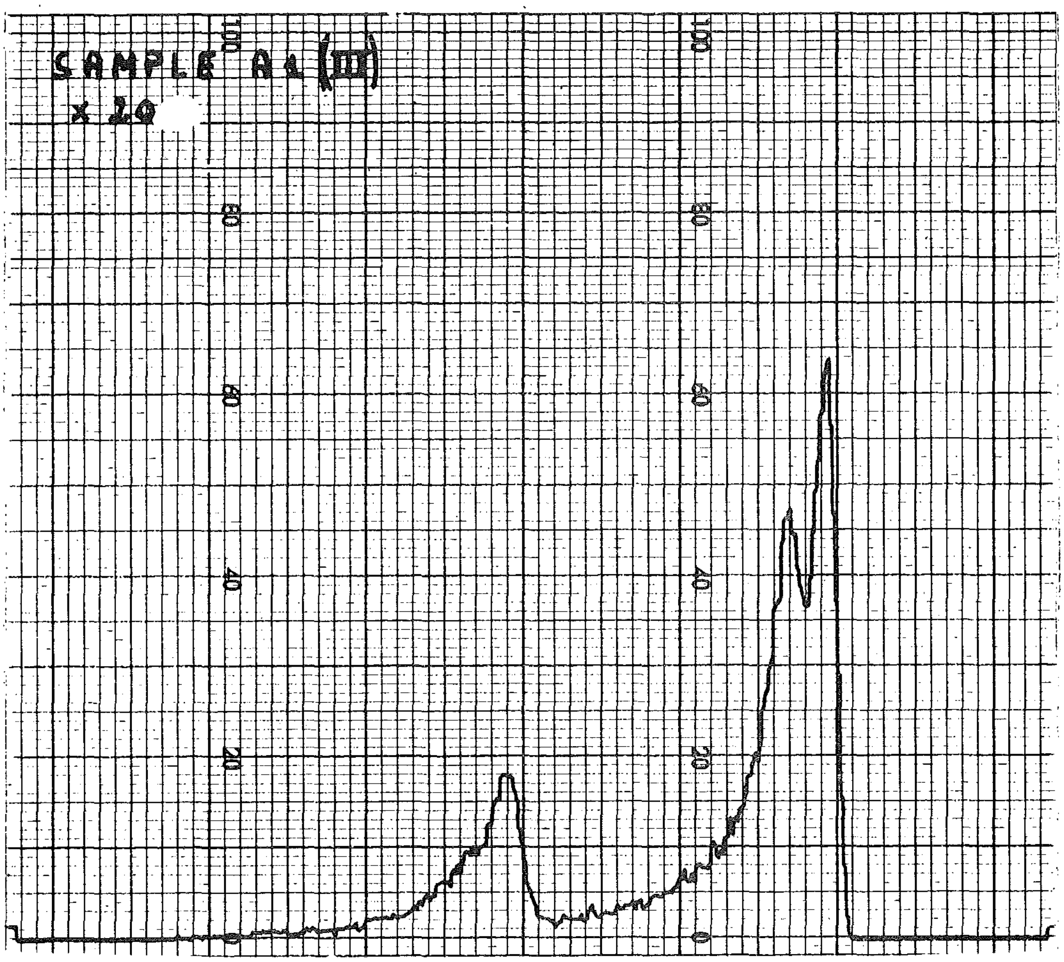
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20
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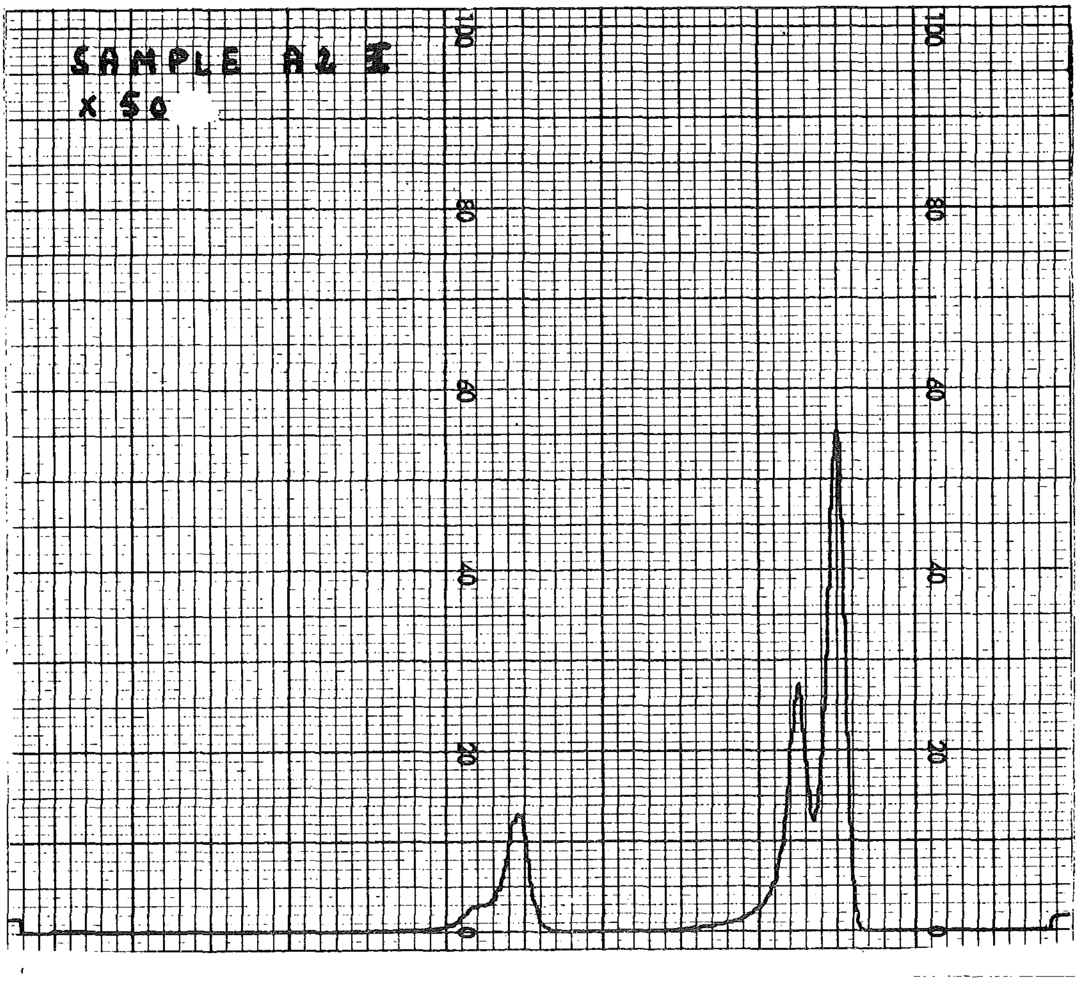
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20
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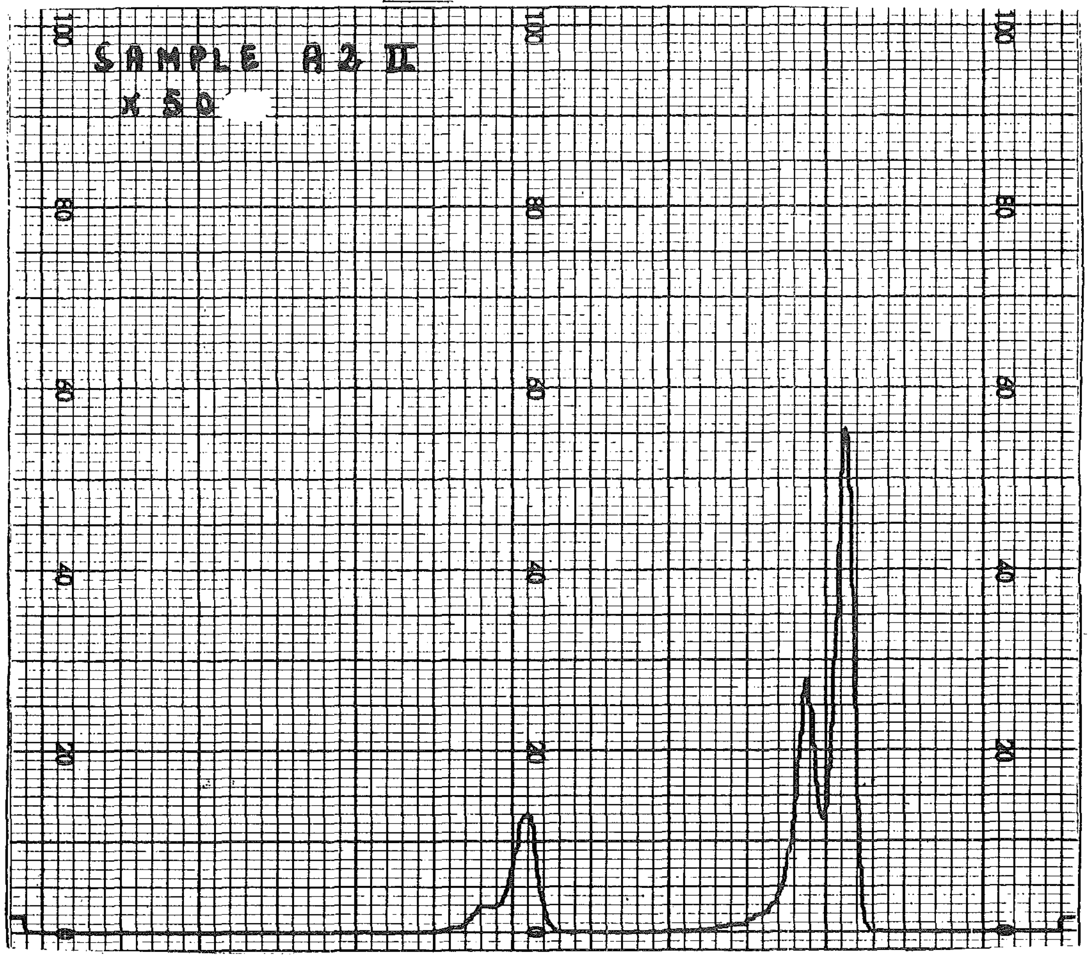
60
40
20
0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SAMPLE A2 III
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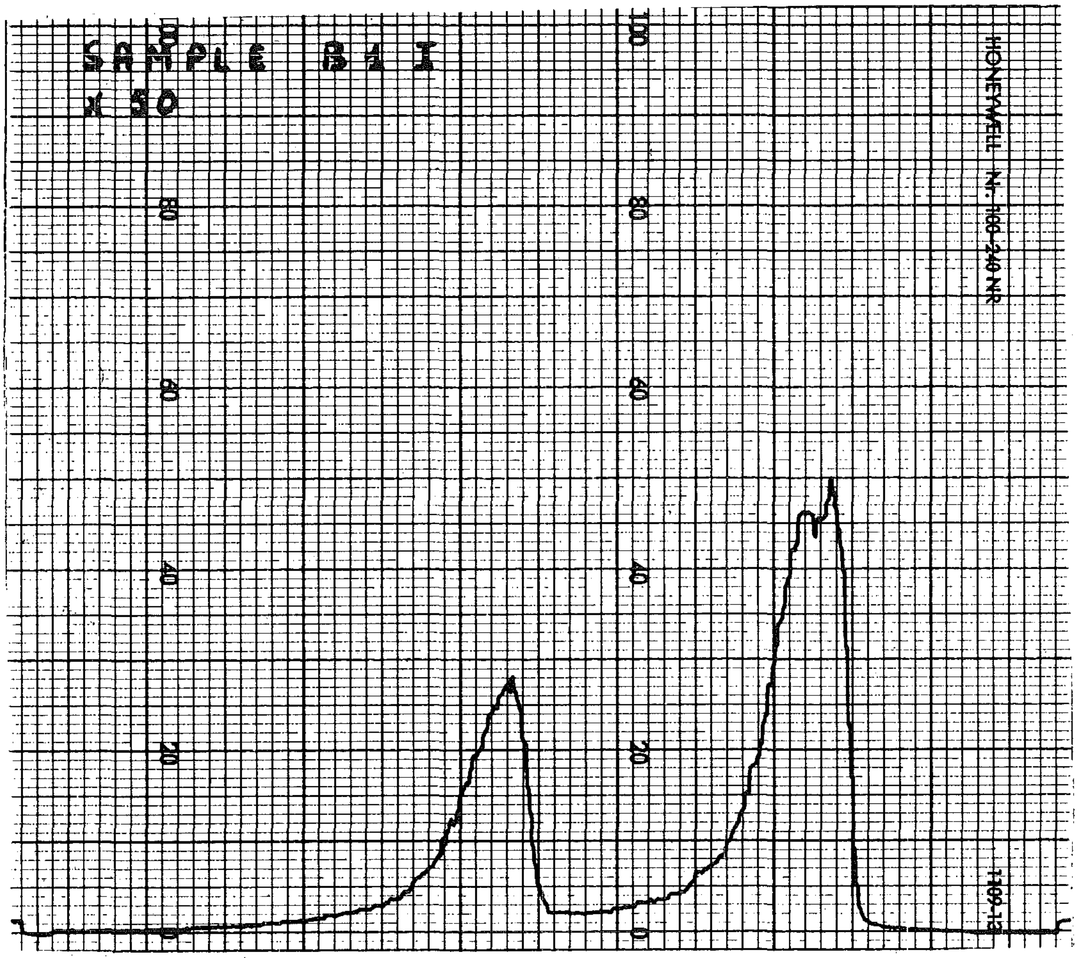
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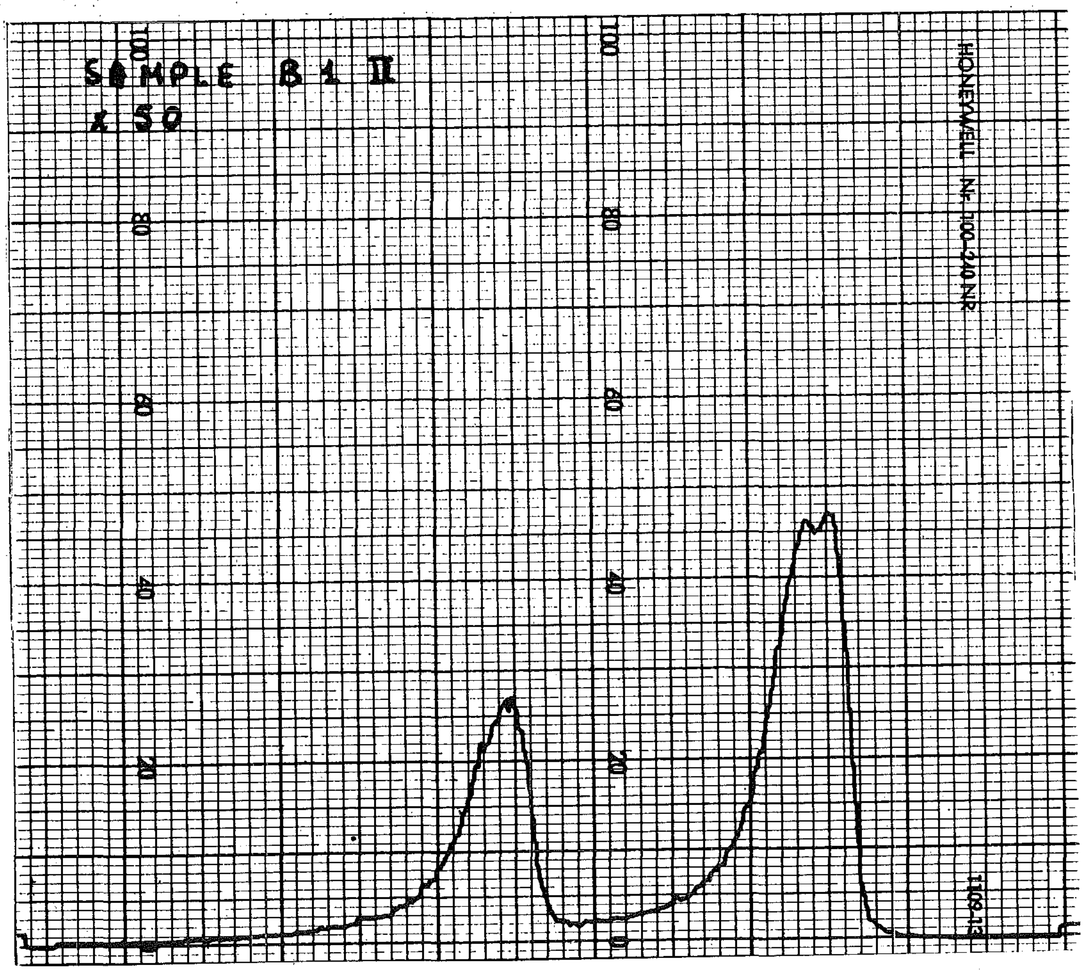
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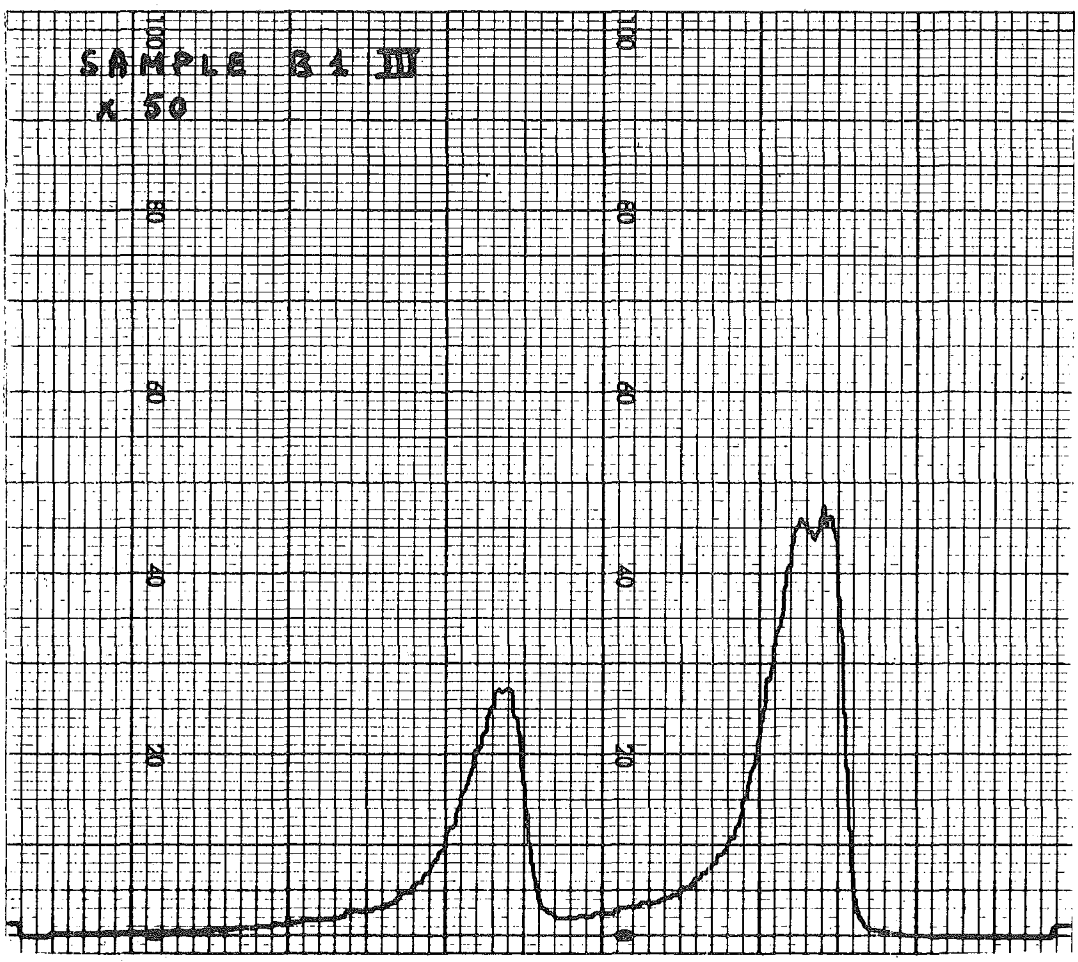
HONEYWELL NF 100-240 NR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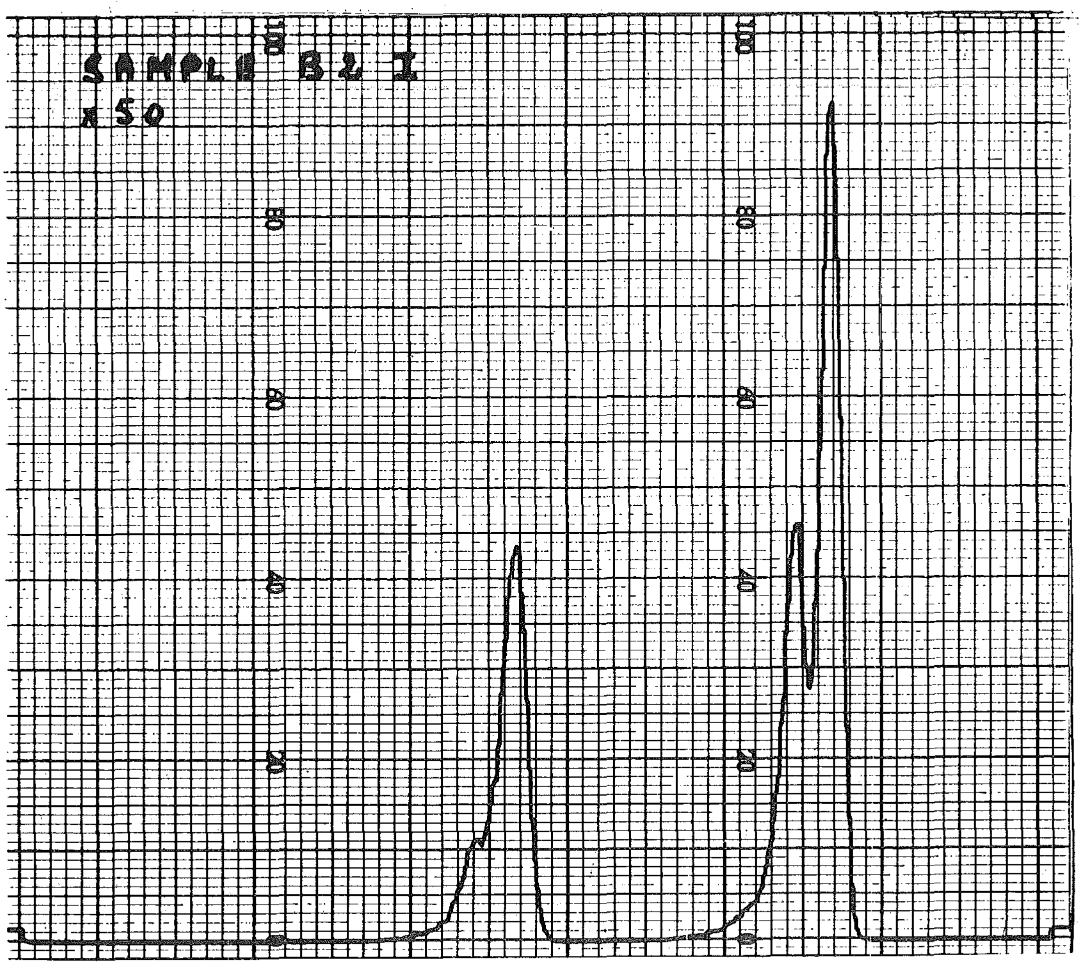
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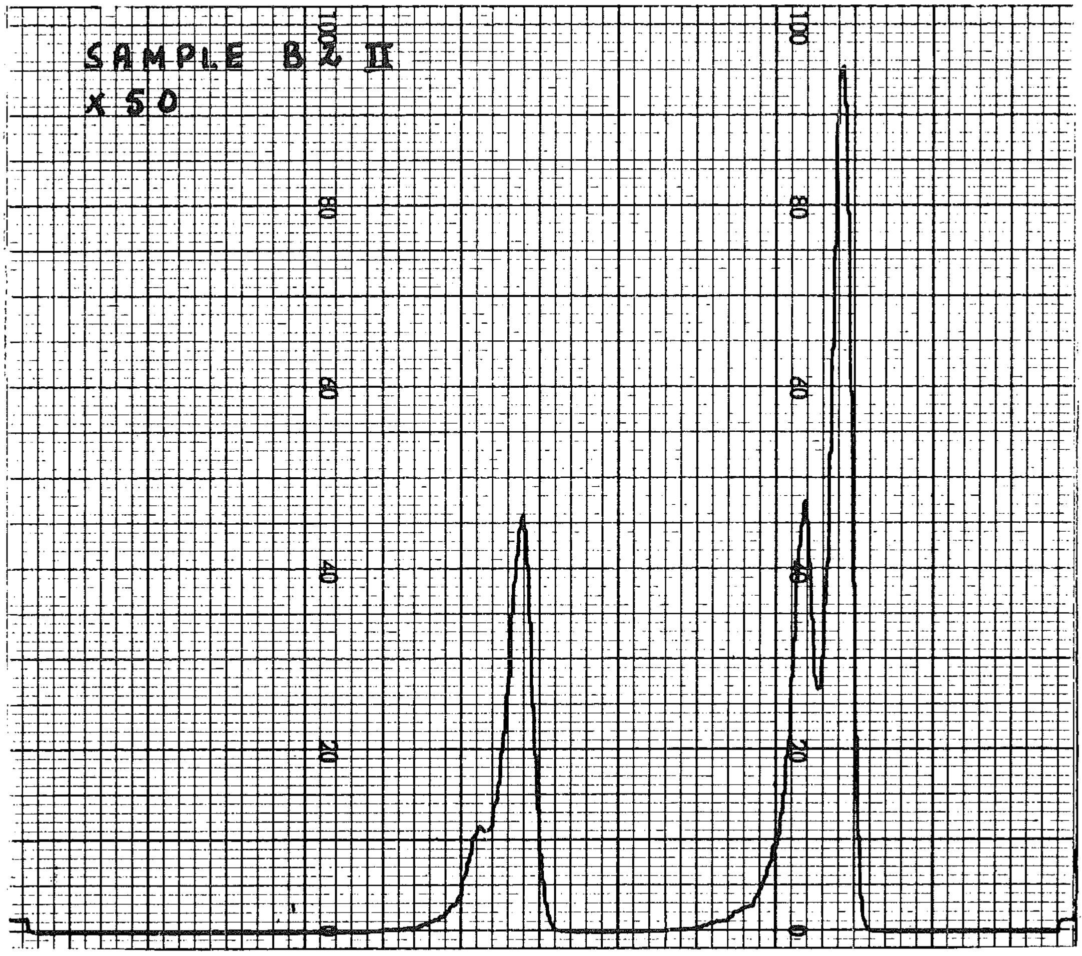
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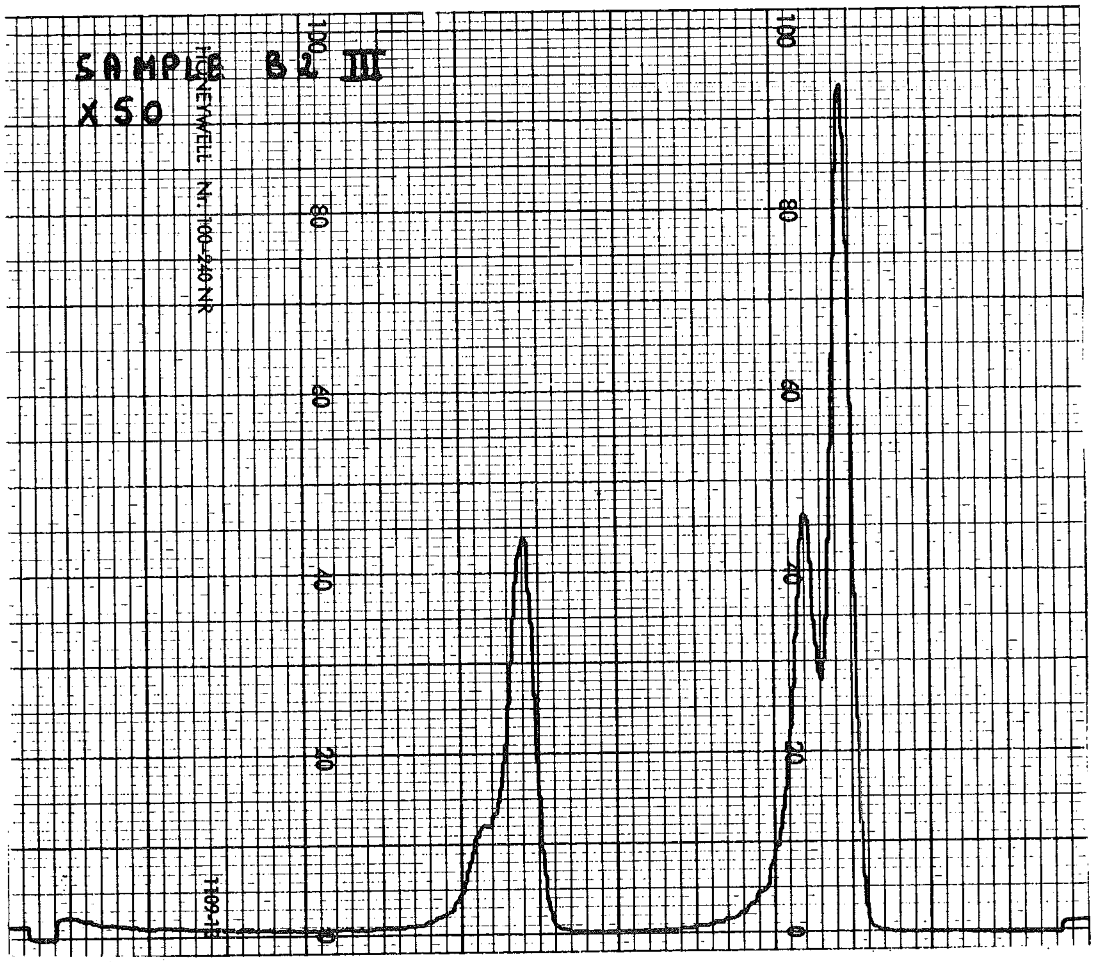
40
20
0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HONEYWELL Nr 100-240 NR
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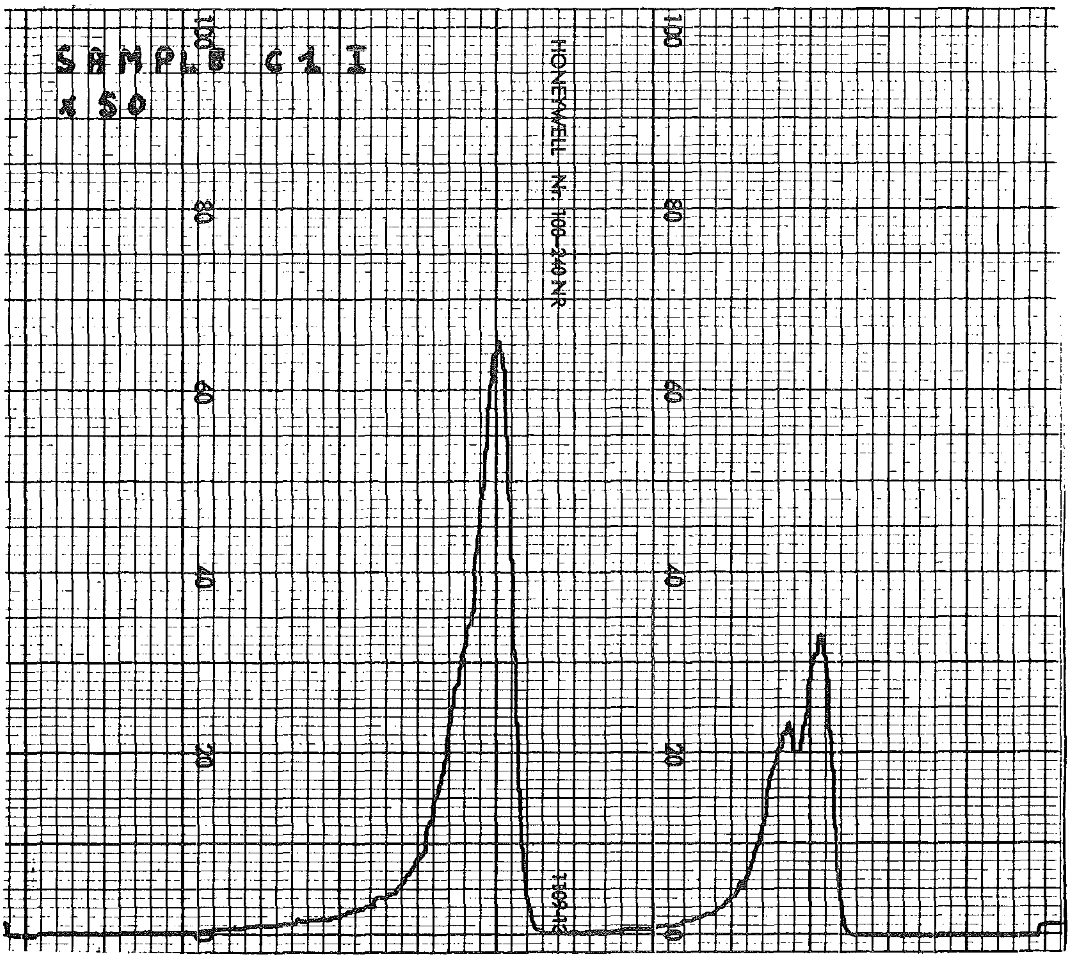
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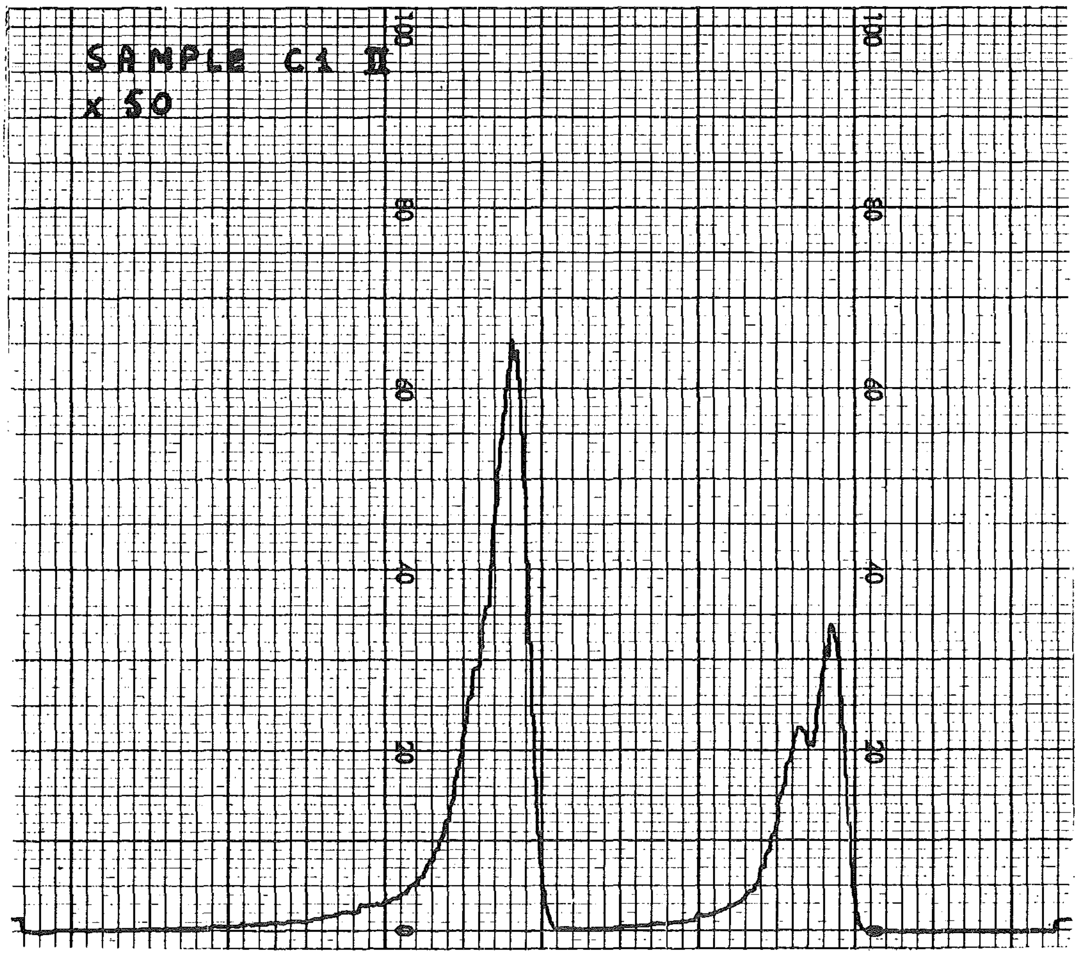
HONEYWELL Nr. 100-240 NR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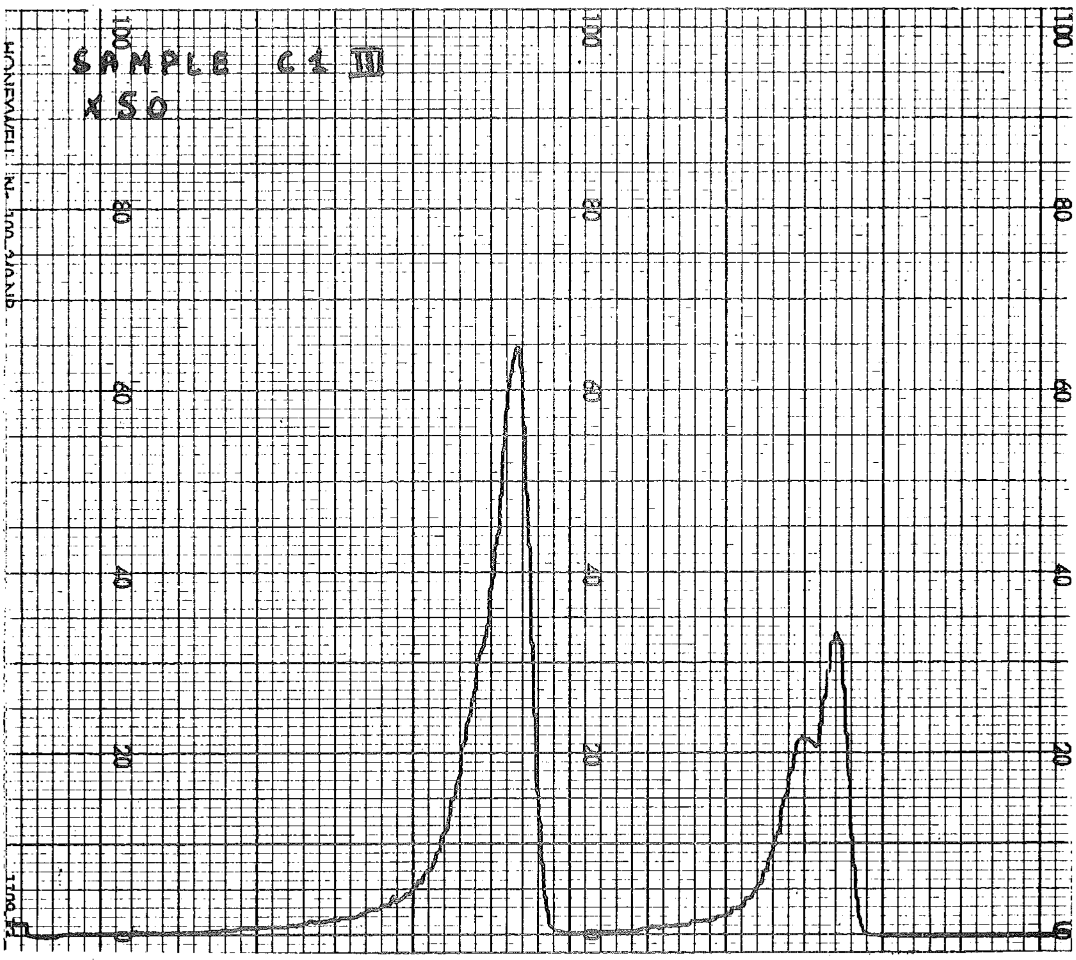
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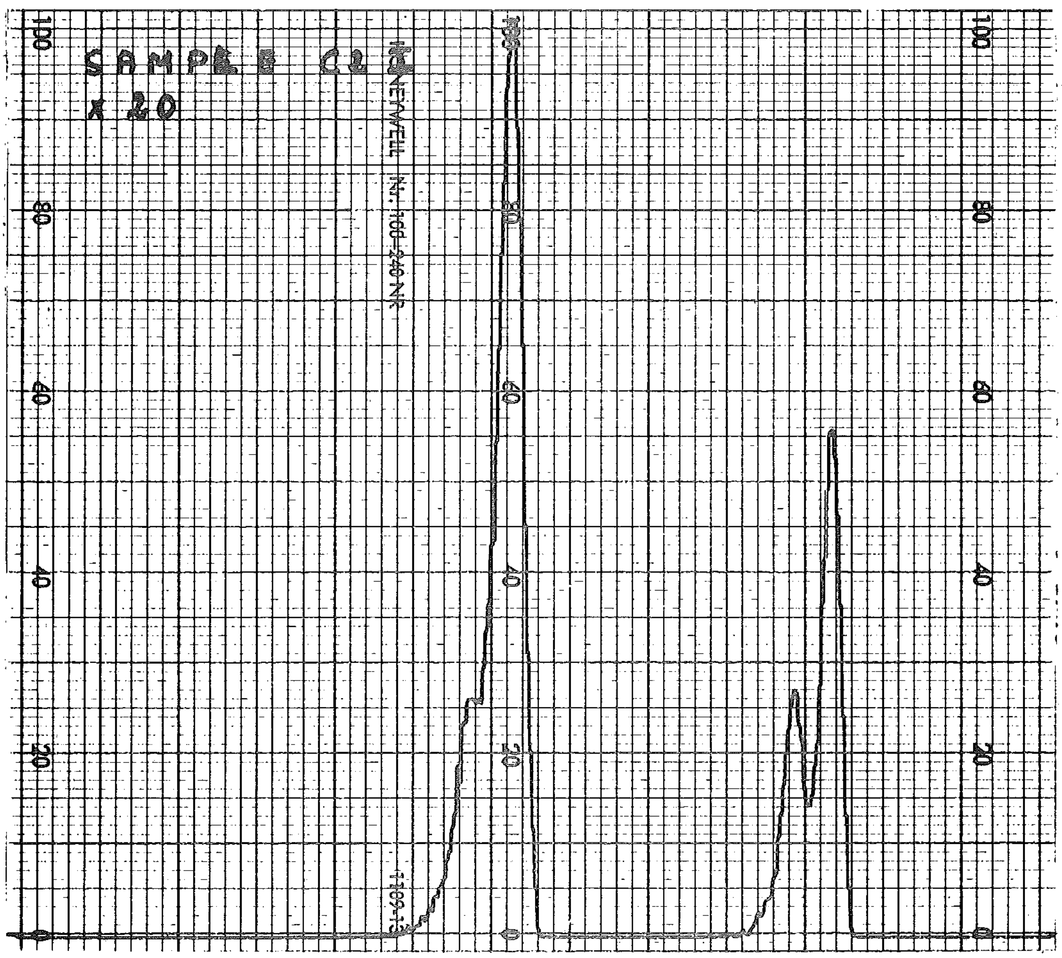
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X 20
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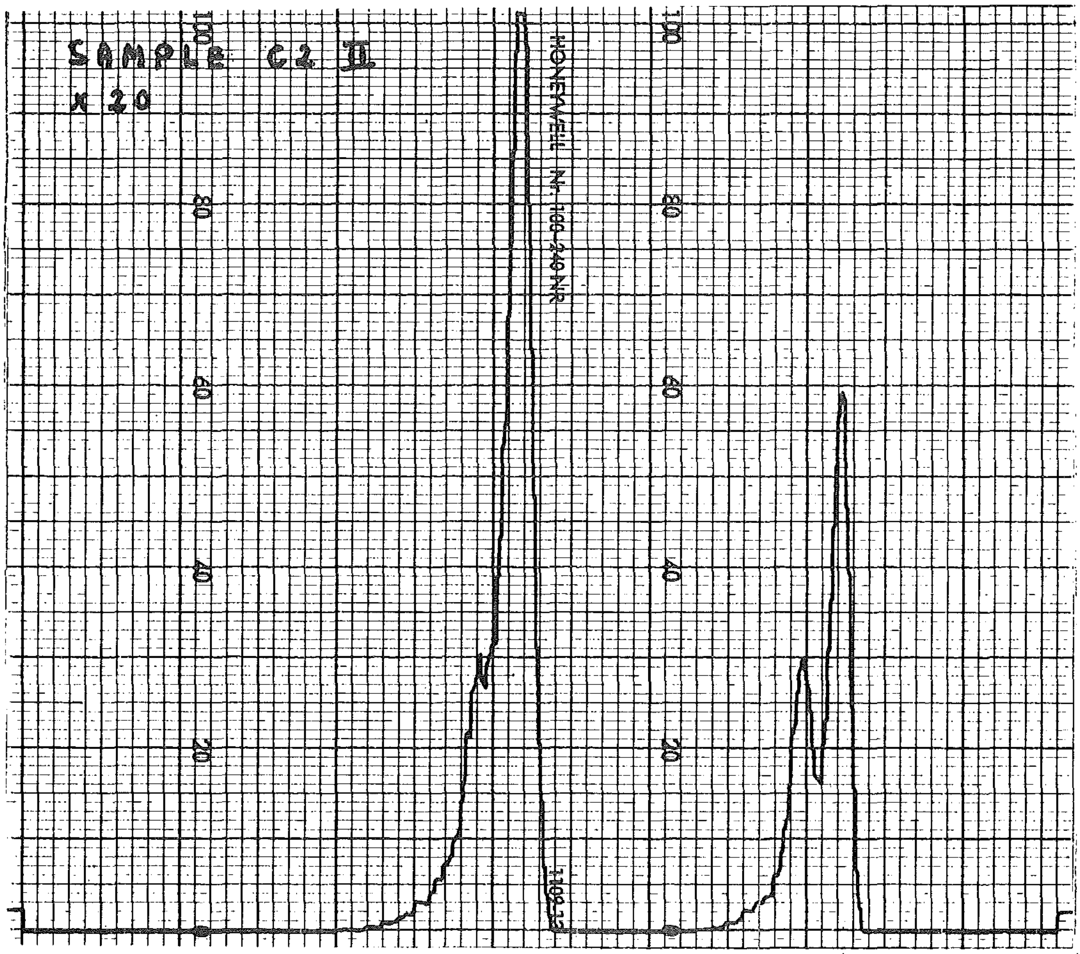

SAMPLE C2 II
x 20
HONEYWELL Nr. 100-240 NR
100
80
60
40
20


SAMPLE C2 (III)
x 20
HONEYWELL Nr. 100-240 NR
100
80
60
40
20

CHANNEL CONTENT: SAMPLE A 1 I

1	003600	000000	000000	000000	000000	000000	000000	000000	000000	000000
11	000008	000031	000024	000022	000037	000027	000028	000036	000027	000022
21	000026	000028	000018	000026	000027	000029	000037	000028	000018	000023
31	000035	000027	000028	000018	000028	000032	000024	000035	000021	000020
41	000023	000033	000031	000026	000034	000030	000028	000031	000023	000029
51	000040	000033	000024	000024	000028	000032	000026	000033	000035	000032
61	000037	000030	000036	000030	000032	000031	000035	000025	000038	000026
71	000034	000032	000049	000041	000033	000036	000040	000036	000039	000046
81	000039	000032	000027	000031	000043	000048	000040	000034	000046	000047
91	000033	000047	000035	000051	000043	000054	000053	000044	000050	000038
101	000040	000047	000054	000039	000032	000067	000049	000050	000055	000043
111	000055	000055	000055	000052	000068	000056	000061	000046	000056	000047
121	000052	000056	000048	000054	000067	000067	000053	000057	000058	000061
131	000063	000068	000072	000066	000086	000069	000075	000068	000090	000080
141	000081	000086	000090	000087	000091	000087	000103	000106	000103	000089
151	000084	000103	000104	000125	000130	000103	000100	000127	000115	000120
161	000130	000135	000142	000143	000161	000151	000165	000184	000178	000179
171	000212	000192	000224	000216	000219	000230	000231	000246	000226	000239
181	000254	000287	000281	000279	000322	000331	000315	000417	000381	000405
191	000396	000389	000365	000303	000287	000238	000199	000127	000102	000098
201	000075	000077	000078	000071	000058	000079	000079	000067	000072	000071
211	000077	000103	000073	000089	000075	000072	000095	000081	000082	000082
221	000087	000073	000092	000079	000103	000093	000075	000092	000096	000114
231	000084	000100	000110	000099	000110	000100	000111	000117	000117	000103
241	000108	000108	000099	000123	000111	000126	000144	000118	000140	000143
251	000152	000110	000136	000147	000154	000134	000140	000160	000175	000162
261	000163	000150	000151	000198	000192	000180	000210	000209	000176	000213
271	000224	000263	000241	000226	000267	000264	000291	000298	000250	000335
281	000333	000319	000382	000369	000387	000423	000403	000449	000461	000495
291	000526	000565	000580	000616	000732	000759	000805	000849	000960	000912
301	000949	000909	000885	000807	000804	000802	000806	000815	000859	000978
311	001007	001148	001261	001228	001287	001143	001119	000896	000690	000506
321	000281	000146	000078	000026	000007	000003	000001	000001	000000	000000
331	000000	000000	000000	000001	000000	000000	000000	000000	000000	000001
341	000000	000000	000000	000000	000000	000001	000000	000000	000000	000000
351	000000	000000	000001	000000	000000	000001	000000	000000	000000	000000
361	000000	000000	000001	000000	000000	000000	000001	000000	000000	000000
371	000000	000000	000000	000000	000000	000001	000000	000000	000000	000000
381	000000	000000	000001	000001	000000	000000	000001	000000	000000	000000
391	000002	000000	000000	000000	000000	000000	000000	000000	000000	000000

CHANNEL CONTENT: SAMPLE A 1 II

1	003600	000000	000000	000000	000000	000000	000000	000000	000000	000000
11	000002	000027	000032	000034	000029	000032	000026	000045	000028	000030
21	000030	000028	000044	000033	000031	000030	000030	000030	000029	000021
31	000033	000031	000028	000026	000018	000032	000034	000030	000023	000033
41	000030	000026	000033	000031	000034	000033	000026	000036	000035	000030
51	000035	000027	000030	000031	000034	000034	000031	000032	000035	000027
61	000031	000031	000048	000028	000035	000039	000032	000035	000032	000043
71	000029	000035	000038	000042	000040	000035	000032	000033	000037	000042
81	000028	000033	000041	000048	000042	000047	000042	000037	000052	000049
91	000033	000045	000037	000041	000049	000045	000046	000050	000040	000040
101	000052	000044	000049	000036	000067	000051	000050	000048	000053	000037
111	000052	000069	000047	000055	000069	000062	000062	000049	000047	000061
121	000061	000065	000062	000066	000059	000061	000063	000068	000063	000064
131	000064	000069	000072	000074	000071	000064	000074	000080	000075	000084
141	000069	000082	000103	000073	000073	000087	000088	000096	000095	000097
151	000105	000111	000113	000098	000123	000107	000132	000117	000130	000118
161	000117	000146	000153	000159	000145	000153	000187	000155	000186	000172
171	000201	000207	000221	000243	000214	000203	000225	000234	000266	000276
181	000259	000291	000259	000274	000317	000353	000369	000362	000390	000395
191	000436	000408	000368	000322	000299	000205	000198	000139	000122	000106
201	000096	000072	000079	000069	000062	000070	000070	000062	000078	000056
211	000069	000076	000078	000073	000074	000075	000111	000096	000081	000088
221	000088	000074	000102	000098	000088	000081	000077	000091	000071	000099
231	000086	000099	000109	000116	000106	000094	000117	000095	000129	000091
241	000116	000124	000107	000128	000116	000108	000121	000146	000124	000138
251	000159	000123	000147	000161	000144	000161	000151	000160	000163	000164
261	000173	000166	000157	000179	000169	000201	000190	000219	000184	000212
271	000230	000227	000258	000246	000233	000277	000281	000281	000312	000334
281	000328	000337	000335	000371	000381	000407	000461	000441	000538	000495
291	000515	000610	000647	000644	000788	000754	000864	000862	000886	001000
301	000939	000902	000902	000864	000763	000735	000751	000831	000863	000904
311	000991	001024	001121	001219	001253	001261	001114	000967	000798	000563
321	000335	000178	000071	000036	000010	000002	000000	000000	000000	000000
331	000000	000000	000000	000000	000000	000000	000000	000001	000001	000000
341	000000	000000	000000	000000	000001	000000	000000	000000	000000	000000
351	000000	000000	000000	000000	000000	000000	000000	000000	000000	000000
361	000000	000000	000000	000000	000000	000000	000000	000000	000000	000000
371	000001	000000	000000	000000	000000	000000	000000	000000	000001	000000
381	000000	000000	000001	000000	000000	000002	000000	000000	000000	000000
391	000000	000000	000001	000000	000000	000000	000000	000000	000000	000000

CHANNEL CONTENT:

SAMPLE A 1 III

1	003600	000000	000000	000000	000000	000000	000000	000000	000000	000000
11	000002	000031	000029	000036	000036	000034	000028	000034	000025	000030
21	000034	000027	000022	000024	000020	000027	000026	000029	000012	000030
31	000023	000030	000024	000025	000029	000036	000018	000027	000021	000024
41	000025	000027	000022	000024	000030	000032	000035	000028	000026	000029
51	000024	000029	000026	000033	000027	000032	000034	000028	000034	000029
61	000032	000030	000041	000029	000036	000030	000029	000024	000045	000037
71	000031	000024	000045	000038	000034	000036	000040	000037	000046	000038
81	000031	000035	000039	000045	000034	000042	000046	000035	000036	000032
91	000043	000055	000045	000044	000046	000052	000044	000051	000046	000056
101	000042	000049	000041	000051	000053	000038	000047	000044	000043	000048
111	000053	000048	000065	000048	000063	000058	000049	000046	000058	000052
121	000050	000055	000054	000039	000064	000059	000056	000063	000065	000066
131	000065	000071	000044	000066	000063	000082	000078	000083	000088	000077
141	000083	000083	000087	000076	000077	000094	000072	000086	000089	000085
151	000099	000097	000102	000113	000094	000125	000104	000136	000134	000113
161	000134	000131	000166	000160	000161	000151	000166	000144	000194	000179
171	000207	000179	000218	000234	000224	000216	000246	000217	000229	000242
181	000248	000248	000294	000284	000336	000346	000332	000394	000392	000382
191	000381	000375	000346	000325	000262	000234	000198	000149	000130	000113
201	000108	000072	000081	000072	000078	000071	000068	000078	000059	000074
211	000070	000097	000072	000074	000073	000075	000073	000076	000098	000069
221	000082	000109	000112	000083	000078	000082	000095	000091	000095	000088
231	000087	000092	000114	000090	000107	000110	000106	000113	000121	000094
241	000129	000121	000096	000117	000112	000132	000133	000138	000120	000121
251	000148	000132	000140	000138	000157	000152	000173	000169	000192	000157
261	000167	000187	000156	000208	000207	000187	000193	000185	000200	000256
271	000235	000203	000234	000218	000265	000261	000304	000255	000308	000281
281	000321	000325	000358	000392	000391	000429	000449	000419	000461	000539
291	000563	000577	000629	000642	000759	000761	000774	000905	000950	000945
301	000970	000905	000917	000825	000780	000767	000746	000758	000823	000921
311	001016	001076	001174	001186	001279	001293	001136	000959	000750	000591
321	000339	000187	000079	000039	000011	000003	000003	000001	000000	000000
331	000001	000000	000000	000000	000000	000000	000000	000000	000000	000000
341	000000	000000	000000	000001	000000	000000	000000	000000	000000	000000
351	000000	000000	000000	000000	000000	000000	000000	000000	000000	000000
361	000000	000000	000000	000000	000000	000000	000000	000000	000000	000000
371	000000	000000	000000	000000	000000	000000	000000	000000	000000	000000
381	000000	000000	000000	000000	000000	000000	000000	000000	000000	000000
391	000002	000000	000000	000000	000000	000000	000000	000001	000000	000001

CHANNEL CONTENT:

SAMPLE A 2 I

1	003600	000000	000000	000000	000000	000000	000000	000000	000000	000000
11	000003	000029	000038	000031	000022	000025	000025	000028	000028	000029
21	000029	000024	000025	000023	000027	000026	000017	000020	000017	000014
31	000022	000014	000018	000014	000014	000017	000018	000015	000018	000014
41	000015	000016	000009	000007	000013	000016	000017	000012	000010	000017
51	000020	000011	000014	000008	000007	000015	000012	000013	000019	000012
61	000008	000019	000010	000008	000012	000012	000013	000014	000013	000012
71	000014	000012	000009	000012	000016	000012	000016	000014	000007	000010
81	000010	000012	000012	000011	000009	000011	000010	000012	000010	000011
91	000012	000008	000010	000014	000007	000011	000009	000010	000015	000014
101	000014	000007	000011	000010	000014	000013	000010	000008	000008	000010
111	000008	000010	000014	000014	000010	000006	000007	000007	000011	000010
121	000007	000007	000018	000006	000010	000010	000013	000007	000011	000012
131	000015	000014	000008	000012	000008	000021	000008	000020	000011	000010
141	000014	000017	000011	000011	000017	000023	000017	000023	000021	000012
151	000026	000024	000016	000018	000027	000021	000027	000034	000033	000031
161	000023	000033	000043	000037	000041	000047	000052	000057	000056	000085
171	000087	000110	000100	000137	000158	000160	000167	000155	000184	000170
181	000167	000185	000189	000216	000205	000272	000263	000330	000399	000457
191	000539	000640	000612	000684	000648	000659	000524	000394	000263	000173
201	000148	000085	000042	000026	000021	000012	000007	000007	000006	000007
211	000008	000009	000001	000004	000008	000004	000010	000006	000005	000001
221	000004	000005	000011	000006	000008	000008	000006	000012	000008	000011
231	000008	000009	000010	000009	000013	000011	000008	000009	000009	000016
241	000014	000010	000014	000008	000012	000014	000021	000012	000013	000014
251	000016	000016	000019	000012	000020	000025	000023	000023	000034	000031
261	000029	000040	000033	000027	000048	000048	000043	000043	000049	000040
271	000060	000051	000057	000061	000071	000072	000067	000083	000083	000074
281	000109	000087	000114	000121	000135	000157	000157	000157	000183	000208
291	000238	000255	000304	000304	000407	000470	000510	000662	000829	001073
301	001141	001291	001389	001273	001092	000927	000765	000639	000616	000702
311	000835	001000	001242	001546	001912	002367	002621	002777	002667	002261
321	001713	001084	000588	000273	000105	000037	000007	000005	000003	000001
331	000003	000005	000003	000002	000000	000000	000000	000000	000001	000001
341	000001	000001	000001	000000	000002	000000	000000	000000	000001	000000
351	000002	000002	000005	000000	000002	000000	000000	000000	000002	000000
361	000000	000001	000002	000000	000001	000001	000003	000001	000000	000002
371	000001	000002	000001	000000	000001	000002	000001	000002	000000	000000
381	000002	000003	000000	000000	000001	000001	000000	000000	000000	000000
391	000000	000001	000001	000000	000000	000001	000000	000000	000000	000000

CHANNEL CONTENT: SAMPLE A 2 II

1	003600	000000	000000	000000	000000	000000	000000	000000	000000	000000
11	000000	000000	000002	000003	000000	000003	000002	000003	000001	000002
21	000001	000002	000001	000001	000002	000004	000001	000000	000001	000003
31	000001	000004	000000	000004	000000	000000	000002	000004	000001	000000
41	000002	000003	000001	000002	000001	000004	000001	000002	000001	000001
51	000000	000000	000002	000000	000001	000002	000002	000000	000001	000001
61	000000	000001	000003	000000	000000	000000	000000	000000	000002	000001
71	000003	000002	000001	000001	000003	000003	000002	000001	000003	000001
81	000003	000002	000001	000002	000000	000005	000000	000002	000003	000002
91	000002	000003	000002	000001	000003	000001	000002	000001	000000	000001
101	000001	000001	000001	000001	000005	000005	000003	000002	000003	000007
111	000003	000001	000002	000004	000002	000005	000002	000002	000003	000001
121	000003	000007	000005	000004	000005	000003	000004	000008	000003	000010
131	000004	000010	000006	000007	000004	000007	000008	000009	000007	000006
141	000009	000008	000014	000016	000010	000009	000010	000007	000007	000018
151	000014	000016	000018	000014	000014	000026	000019	000021	000026	000024
161	000032	000026	000035	000043	000043	000050	000047	000047	000050	000063
171	000081	000100	000097	000134	000144	000171	000178	000163	000164	000159
181	000141	000169	000185	000184	000201	000241	000279	000334	000394	000484
191	000514	000640	000662	000668	000689	000645	000583	000423	000309	000211
201	000169	000109	000054	000038	000014	000010	000006	000001	000004	000001
211	000001	000004	000001	000004	000002	000007	000008	000002	000005	000002
221	000004	000004	000003	000002	000004	000004	000005	000003	000004	000002
231	000004	000004	000007	000006	000009	000010	000003	000009	000006	000009
241	000008	000012	000012	000010	000011	000008	000015	000018	000016	000015
251	000019	000014	000021	000010	000011	000024	000017	000021	000021	000023
261	000024	000032	000025	000028	000030	000031	000037	000058	000043	000047
271	000054	000059	000062	000054	000056	000069	000067	000065	000062	000097
281	000099	000094	000123	000105	000127	000126	000140	000134	000182	000190
291	000192	000255	000271	000327	000395	000481	000528	000650	000777	000994
301	001122	001336	001416	001243	001207	001017	000808	000674	000620	000669
311	000739	000932	001176	001530	001751	002162	002552	002783	002707	002417
321	001885	001226	000711	000351	000125	000040	000010	000002	000003	000001
331	000000	000000	000000	000000	000000	000001	000000	000000	000000	000000
341	000000	000001	000000	000000	000001	000000	000000	000000	000000	000000
351	000000	000000	000000	000000	000000	000000	000000	000000	000000	000000
361	000000	000002	000000	000000	000000	000000	000001	000000	000000	000002
371	000001	000000	000001	000001	000001	000001	000001	000000	000000	000000
381	000000	000000	000000	000000	000001	000000	000000	000000	000000	000000
391	000001	000000	000000	000000	000000	000000	000000	000000	000000	000001

CHANNEL CONTENT: SAMPLE A 2 III

1	003600	000000	000000	000000	000000	000000	000000	000000	000000	000000
11	000000	000000	000001	000001	000002	000000	000001	000002	000000	000002
21	000001	000000	000001	000000	000001	000002	000001	000001	000002	000003
31	000001	000001	000001	000003	000001	000003	000000	000002	000004	000002
41	000004	000000	000002	000000	000000	000002	000004	000000	000000	000002
51	000000	000003	000001	000003	000000	000002	000002	000001	000002	000001
61	000002	000002	000000	000002	000002	000002	000006	000000	000001	000000
71	000002	000001	000001	000002	000002	000000	000002	000005	000001	000003
81	000000	000004	000002	000002	000003	000004	000004	000001	000004	000005
91	000003	000001	000003	000003	000002	000004	000003	000002	000001	000001
101	000000	000001	000004	000002	000004	000001	000005	000003	000003	000002
111	000004	000004	000004	000003	000005	000005	000002	000004	000001	000006
121	000004	000000	000000	000005	000008	000005	000003	000003	000004	000007
131	000005	000005	000007	000000	000012	000003	000006	000004	000007	000006
141	000005	000005	000011	000005	000015	000012	000011	000015	000018	000012
151	000013	000010	000015	000015	000021	000016	000019	000016	000016	000027
161	000034	000021	000030	000032	000038	000049	000055	000056	000057	000064
171	000083	000097	000105	000118	000154	000186	000195	000164	000175	000159
181	000150	000178	000157	000211	000230	000254	000276	000312	000374	000436
191	000521	000557	000633	000674	000711	000653	000541	000408	000306	000234
201	000146	000089	000047	000037	000012	000009	000005	000003	000001	000002
211	000005	000002	000003	000003	000001	000003	000005	000001	000002	000002
221	000005	000005	000002	000003	000005	000006	000004	000004	000005	000007
231	000005	000004	000008	000005	000006	000010	000010	000011	000010	000013
241	000014	000009	000007	000011	000015	000012	000013	000010	000017	000021
251	000010	000017	000014	000019	000014	000022	000026	000027	000028	000021
261	000022	000029	000023	000036	000039	000039	000024	000030	000038	000044
271	000046	000052	000055	000063	000055	000071	000066	000078	000073	000097
281	000081	000108	000097	000106	000119	000136	000163	000165	000183	000214
291	000198	000231	000307	000298	000382	000448	000527	000644	000849	000934
301	001160	001353	001342	001308	001196	000981	000769	000659	000592	000615
311	000809	000951	001167	001487	001976	002304	002705	002910	002769	002485
321	001800	001218	000663	000330	000117	000026	000011	000003	000000	000000
331	000000	000001	000000	000000	000000	000000	000000	000000	000000	000000
341	000000	000000	000000	000001	000000	000001	000000	000000	000000	000000
351	000000	000000	000000	000001	000000	000001	000000	000001	000000	000000
361	000000	000000	000000	000000	000000	000000	000000	000000	000001	000000
371	000000	000000	000001	000000	000000	000000	000000	000000	000000	000000
381	000000	000000	000001	000000	000000	000000	000000	000000	000000	000000
391	000001	000001	000000	000000	000000	000000	000000	000000	000000	000001

CHANNEL CONTENT: SAMPLE B 1 I

1	003600	000000	000000	000000	000000	000000	000000	000000	000000	000000
11	000002	000031	000045	000043	000037	000032	000037	000039	000040	000044
21	000027	000033	000031	000039	000027	000034	000042	000035	000035	000035
31	000038	000044	000042	000043	000039	000033	000033	000039	000041	000037
41	000039	000036	000047	000048	000045	000038	000048	000049	000050	000040
51	000047	000045	000045	000053	000046	000054	000041	000042	000056	000037
61	000053	000041	000045	000063	000037	000060	000049	000061	000045	000069
71	000048	000061	000051	000064	000049	000070	000046	000064	000061	000056
81	000065	000074	000058	000063	000050	000060	000072	000075	000059	000083
91	000083	000054	000076	000091	000079	000089	000085	000085	000079	000076
101	000108	000095	000093	000080	000097	000081	000096	000104	000082	000088
111	000102	000101	000113	000119	000105	000117	000103	000132	000126	000128
121	000104	000127	000134	000142	000147	000149	000142	000146	000156	000155
131	000173	000156	000161	000150	000160	000187	000190	000181	000170	000183
141	000215	000207	000233	000206	000220	000229	000247	000272	000260	000266
151	000302	000331	000356	000330	000370	000366	000395	000404	000407	000447
161	000467	000489	000578	000508	000618	000673	000604	000667	000725	000807
171	000837	000867	000851	000943	001019	001015	001042	001063	001140	001160
181	001234	001238	001273	001237	001332	001345	001371	001395	001310	001433
191	001317	001265	001213	001066	001035	000789	000655	000481	000376	000275
201	000232	000186	000160	000117	000121	000127	000130	000145	000122	000122
211	000141	000118	000124	000131	000136	000121	000119	000122	000135	000127
221	000147	000136	000139	000159	000125	000141	000127	000139	000172	000169
231	000159	000152	000170	000168	000177	000170	000156	000214	000192	000203
241	000181	000203	000221	000184	000195	000220	000215	000212	000221	000225
251	000261	000258	000247	000256	000267	000257	000273	000304	000321	000322
261	000385	000363	000357	000398	000380	000416	000415	000420	000434	000474
271	000468	000461	000557	000554	000594	000630	000659	000702	000729	000816
281	000788	000955	000965	000968	001012	001080	001193	001231	001391	001369
291	001507	001616	001737	001754	001821	001954	001986	002141	002114	002178
301	002332	002352	002313	002340	002314	002291	002173	002318	002285	002300
311	002336	002428	002539	002416	002338	002219	002107	001972	001556	001142
321	000851	000477	000269	000136	000097	000076	000072	000054	000059	000053
331	000054	000047	000038	000046	000052	000034	000040	000044	000054	000037
341	000032	000022	000038	000044	000035	000037	000020	000036	000028	000021
351	000024	000020	000025	000015	000018	000025	000022	000024	000010	000021
361	000014	000015	000010	000007	000010	000008	000005	000005	000010	000006
371	000009	000009	000008	000001	000003	000002	000004	000004	000002	000005
381	000001	000001	000007	000004	000005	000001	000001	000005	000004	000001
391	000004	000002	000002	000001	000000	000001	000001	000000	000000	000001

CHANNEL CONTENT: SAMPLE B 1 II

1	003605	000000	000000	000000	000000	000000	000000	000000	000000	000000
11	000001	000038	000050	000042	000036	000040	000039	000039	000048	000047
21	000038	000033	000035	000036	000034	000034	000035	000037	000039	000039
31	000036	000044	000027	000044	000031	000041	000044	000062	000029	000041
41	000034	000050	000046	000043	000039	000035	000033	000048	000037	000040
51	000037	000036	000046	000042	000047	000059	000042	000042	000056	000050
61	000047	000060	000059	000055	000056	000060	000065	000050	000060	000065
71	000038	000067	000061	000072	000069	000049	000064	000061	000059	000060
81	000060	000053	000066	000076	000068	000072	000068	000084	000069	000067
91	000081	000077	000076	000073	000085	000068	000080	000078	000089	000080
101	000087	000075	000085	000098	000103	000102	000081	000097	000097	000101
111	000092	000100	000109	000121	000109	000102	000101	000121	000129	000124
121	000119	000101	000114	000123	000133	000140	000143	000135	000183	000163
131	000165	000153	000165	000162	000171	000189	000175	000185	000195	000199
141	000169	000214	000233	000232	000253	000228	000247	000266	000282	000249
151	000282	000297	000308	000375	000352	000348	000399	000398	000388	000460
161	000465	000497	000521	000550	000594	000604	000649	000630	000701	000764
171	000783	000833	000868	000946	001021	000992	001145	001071	001134	001139
181	001185	001247	001252	001247	001325	001343	001364	001278	001382	001301
191	001275	001164	001171	001053	000991	000835	000685	000557	000424	000347
201	000299	000243	000200	000174	000148	000127	000141	000123	000135	000135
211	000099	000130	000129	000094	000115	000125	000152	000129	000121	000132
221	000151	000142	000126	000136	000120	000141	000160	000148	000161	000144
231	000156	000148	000150	000173	000163	000174	000180	000192	000182	000192
241	000191	000201	000197	000203	000202	000224	000214	000227	000232	000258
251	000235	000297	000252	000238	000279	000255	000289	000300	000306	000344
261	000330	000353	000376	000379	000358	000428	000431	000419	000478	000455
271	000455	000542	000550	000570	000602	000629	000667	000704	000774	000779
281	000804	000852	000971	000991	001084	001072	001180	001261	001326	001410
291	001490	001648	001642	001729	001842	001910	002005	002049	002110	002192
301	002229	002291	002355	002329	002285	002304	002240	002298	002289	002336
311	002372	002396	002355	002366	002271	002128	001992	001746	001570	001260
321	000978	000744	000536	000319	000247	000170	000106	000110	000094	000073
331	000070	000057	000049	000045	000043	000040	000034	000027	000028	000024
341	000025	000025	000022	000020	000016	000018	000014	000010	000017	000010
351	000025	000014	000011	000012	000015	000013	000004	000010	000007	000011
361	000006	000003	000001	000002	000001	000005	000005	000004	000004	000005
371	000006	000000	000002	000002	000002	000002	000002	000001	000003	000001
381	000001	000001	000000	000000	000001	000001	000000	000001	000000	000001
391	000000	000000	000000	000003	000000	000000	000000	000000	000000	000001

CHANNEL CONTENT: SAMPLE B 1 III

1	003600	000000	000000	000000	000001	000000	000002	000000	000000	000000
11	000003	000043	000048	000044	000045	000039	000035	000028	000054	000039
21	000035	000037	000047	000036	000050	000031	000034	000034	000032	000031
31	000043	000033	000037	000055	000050	000035	000043	000047	000047	000041
41	000042	000049	000041	000042	000053	000048	000048	000043	000031	000052
51	000059	000047	000036	000047	000046	000059	000059	000048	000055	000062
61	000053	000045	000033	000057	000039	000061	000058	000045	000046	000059
71	000061	000064	000067	000063	000064	000070	000076	000049	000051	000071
81	000068	000078	000065	000079	000080	000079	000077	000084	000065	000062
91	000061	000080	000091	000080	000086	000080	000088	000076	000113	000092
101	000094	000081	000087	000115	000104	000103	000102	000112	000111	000127
111	000108	000124	000112	000122	000106	000121	000001	000114	000130	000109
121	000111	000126	000137	000129	000152	000123	000188	000164	000154	000173
131	000174	000148	000171	000168	000175	000178	000170	000207	000191	000192
141	000175	000227	000204	000227	000229	000255	000250	000281	000283	000286
151	000273	000299	000334	000346	000340	000334	000392	000394	000395	000449
161	000478	000471	000528	000526	000516	000628	000640	000632	000672	000745
171	000757	000834	000842	000884	000933	000974	001070	001052	001089	001165
181	001161	001252	001290	001294	001383	001389	001341	001371	001385	001377
191	001346	001207	001203	001111	000915	000834	000689	000518	000406	000329
201	000259	000194	000168	000150	000137	000154	000142	000108	000111	000124
211	000114	000119	000132	000132	000103	000125	000150	000134	000127	000113
221	000149	000129	000163	000162	000148	000158	000156	000142	000150	000138
231	000181	000154	000208	000179	000184	000202	000198	000189	000197	000203
241	000231	000218	000218	000203	000206	000225	000227	000236	000253	000246
251	000257	000256	000272	000245	000278	000279	000323	000283	000313	000305
261	000349	000373	000343	000375	000403	000377	000423	000432	000459	000442
271	000506	000480	000518	000530	000570	000599	000582	000659	000679	000722
281	000803	000801	000890	000960	000982	001057	001133	001222	001294	001459
291	001426	001527	001643	001710	001747	001807	001961	002078	002074	002210
301	002281	002309	002341	002289	002312	002226	002265	002185	002257	002273
311	002354	002418	002264	002353	002323	002234	002184	001858	001672	001220
321	000921	000595	000361	000214	000149	000109	000084	000051	000059	000050
331	000046	000057	000046	000048	000038	000042	000032	000032	000025	000031
341	000022	000028	000018	000016	000021	000014	000018	000016	000021	000016
351	000019	000010	000013	000009	000014	000006	000013	000008	000011	000014
361	000014	000008	000012	000010	000010	000006	000005	000007	000013	000005
371	000008	000007	000006	000004	000006	000005	000006	000005	000008	000003
381	000008	000005	000006	000006	000005	000005	000009	000007	000003	000007
391	000003	000006	000004	000008	000008	000005	000003	000005	000011	000003

CHANNEL CONTENT: SAMPLE B 2 I

1	007200	000000	000000	000000	000000	000000	000000	000000	000000	000000
11	000000	000003	000003	000005	000005	000005	000000	000004	000005	000002
21	000004	000006	000005	000001	000004	000003	000006	000005	000005	000004
31	000004	000002	000003	000005	000002	000003	000002	000005	000003	000003
41	000004	000004	000004	000002	000005	000002	000004	000005	000002	000003
51	000004	000003	000002	000003	000004	000004	000003	000006	000001	000002
61	000008	000004	000005	000004	000004	000006	000003	000005	000003	000007
71	000003	000006	000004	000005	000004	000003	000006	000008	000008	000008
81	000005	000001	000006	000005	000006	000005	000003	000005	000005	000007
91	000001	000004	000002	000004	000004	000004	000004	000002	000004	000006
101	000004	000006	000011	000005	000006	000009	000007	000012	000009	000008
111	000007	000011	000007	000010	000011	000012	000004	000007	000012	000003
121	000010	000017	000014	000012	000017	000014	000012	000011	000015	000011
131	000017	000013	000017	000014	000019	000027	000020	000014	000024	000032
141	000022	000030	000026	000035	000039	000045	000040	000039	000052	000051
151	000058	000046	000072	000065	000079	000068	000084	000083	000102	000096
161	000104	000116	000141	000135	000149	000168	000217	000260	000260	000339
171	000351	000405	000468	000554	000566	000592	000611	000592	000549	000627
181	000665	000712	000817	000929	000937	001131	001400	001542	001709	001934
191	002069	002177	002224	002130	001907	001705	001324	000958	000682	000441
201	000292	000184	000093	000059	000036	000007	000007	000008	000006	000005
211	000005	000006	000006	000006	000009	000007	000011	000006	000008	000003
221	000006	000011	000006	000007	000008	000005	000009	000012	000014	000014
231	000011	000009	000011	000009	000009	000018	000009	000014	000019	000010
241	000017	000016	000018	000014	000025	000023	000018	000015	000028	000019
251	000017	000025	000033	000024	000034	000039	000047	000037	000046	000053
261	000054	000051	000058	000053	000052	000058	000082	000068	000088	000081
271	000078	000096	000090	000125	000121	000137	000139	000171	000150	000173
281	000211	000208	000234	000240	000252	000265	000329	000343	000414	000476
291	000553	000607	000724	000861	000951	001236	001378	001721	001900	002161
301	002296	002339	002309	001970	001717	001506	001383	001459	001556	001891
311	002194	002732	003184	003752	004354	004595	004643	004354	003759	002871
321	001990	001183	000680	000309	000129	000050	000018	000014	000005	000002
331	000001	000001	000004	000002	000001	000002	000001	000000	000000	000001
341	000000	000001	000000	000000	000001	000001	000000	000001	000000	000001
351	000000	000000	000000	000000	000000	000000	000000	000000	000000	000000
361	000001	000000	000000	000002	000001	000000	000000	000000	000000	000001
371	000001	000000	000000	000000	000000	000002	000000	000000	000000	000002
381	000001	000001	000000	000000	000000	000001	000001	000000	000001	000001
391	000000	000001	000000	000001	000000	000001	000000	000001	000001	000002

SAMPLE B 2 II

CHANNEL CONTENT:

1	007200	000000	000000	000000	000000	000000	000000	000000	000000	000000
11	000000	000004	000010	000005	000005	000002	000005	000003	000006	000003
21	000003	000002	000002	000006	000007	000008	000003	000004	000006	000005
31	000003	000002	000003	000002	000006	000002	000001	000004	000002	000003
41	000002	000006	000005	000004	000009	000006	000003	000002	000002	000005
51	000005	000002	000003	000003	000003	000004	000004	000004	000001	000003
61	000006	000005	000007	000002	000002	000005	000003	000005	000005	000004
71	000004	000002	000003	000007	000002	000005	000004	000007	000004	000006
81	000006	000004	000008	000002	000006	000005	000010	000007	000002	000009
91	000005	000002	000005	000005	000005	000007	000003	000009	000007	000010
101	000008	000005	000008	000009	000006	000010	000006	000007	000011	000012
111	000007	000009	000008	000012	000011	000012	000004	000012	000009	000008
121	000011	000008	000011	000010	000013	000009	000012	000009	000015	000011
131	000018	000013	000012	000026	000019	000016	000015	000032	000030	000022
141	000028	000026	000031	000029	000030	000032	000049	000044	000048	000041
151	000036	000050	000068	000059	000071	000055	000095	000085	000105	000087
161	000123	000099	000136	000150	000183	000176	000197	000231	000248	000348
171	000374	000436	000489	000575	000588	000621	000586	000563	000594	000602
181	000626	000741	000763	000861	001036	001144	001345	001449	001786	001963
191	002118	002267	002332	002202	002002	001696	001236	000971	000682	000422
201	000272	000153	000088	000044	000014	000008	000004	000005	000010	000003
211	000004	000007	000007	000005	000008	000004	000006	000005	000007	000010
221	000009	000012	000008	000008	000008	000002	000015	000009	000012	000010
231	000011	000011	000007	000014	000010	000018	000010	000015	000013	000020
241	000017	000016	000011	000019	000014	000021	000019	000021	000024	000033
251	000021	000025	000024	000029	000037	000036	000035	000034	000043	000042
261	000054	000058	000063	000045	000077	000076	000075	000090	000075	000086
271	000092	000082	000112	000118	000149	000139	000154	000161	000167	000161
281	000177	000167	000238	000245	000280	000324	000355	000409	000434	000527
291	000511	000585	000697	000808	001006	001093	001431	001647	001971	002168
301	002286	002410	002251	002011	001700	001398	001322	001362	001540	001846
311	002070	002661	003246	003808	004470	004754	004798	004533	003954	002969
321	001918	001082	000535	000191	000069	000023	000007	000004	000000	000000
331	000002	000001	000000	000000	000003	000000	000000	000000	000001	000000
341	000000	000000	000000	000000	000000	000001	000001	000000	000000	000000
351	000002	000001	000000	000000	000000	000001	000000	000000	000000	000000
361	000001	000000	000001	000000	000000	000000	000001	000001	000002	000000
371	000000	000000	000000	000000	000000	000002	000000	000000	000000	000000
381	000000	000000	000000	000001	000001	000000	000002	000000	000000	000002
391	000001	000000	000000	000001	000002	000000	000000	000001	000000	000001

SAMPLE B 2 III

CHANNEL CONTENT:

1	007200	000000	000000	000000	000001	000004	000000	000000	000000	000001
11	000039	000155	000153	000153	000152	000159	000136	000131	000147	000113
21	000128	000108	000114	000134	000096	000103	000093	000084	000093	000094
31	000084	000104	000094	000094	000082	000091	000081	000091	000088	000078
41	000095	000099	000068	000084	000074	000066	000069	000086	000069	000082
51	000082	000078	000088	000082	000082	000069	000080	000067	000086	000074
61	000062	000069	000053	000073	000069	000062	000078	000068	000064	000061
71	000066	000060	000058	000073	000066	000047	000074	000061	000065	000058
81	000045	000064	000071	000060	000057	000056	000067	000058	000063	000054
91	000051	000069	000052	000055	000042	000056	000051	000058	000049	000066
101	000059	000042	000062	000046	000045	000045	000051	000058	000054	000058
111	000054	000058	000056	000064	000046	000072	000049	000059	000053	000055
121	000060	000065	000058	000064	000063	000048	000056	000048	000072	000046
131	000059	000060	000051	000061	000078	000055	000064	000077	000077	000056
141	000072	000057	000073	000061	000071	000078	000085	000087	000078	000078
151	000098	000085	000087	000106	000116	000094	000115	000120	000142	000138
161	000157	000152	000174	000167	000186	000232	000230	000272	000297	000353
171	000411	000461	000513	000594	000590	000643	000657	000637	000628	000638
181	000715	000716	000782	000860	001000	001102	001319	001547	001780	001988
191	002137	002181	002238	002165	001888	001716	001326	000975	000764	000477
201	000333	000194	000132	000079	000055	000040	000048	000041	000039	000035
211	000027	000049	000034	000036	000035	000037	000032	000029	000042	000038
221	000030	000029	000037	000039	000046	000034	000039	000040	000042	000035
231	000039	000047	000044	000043	000033	000034	000032	000040	000051	000035
241	000038	000040	000047	000042	000049	000044	000055	000053	000037	000049
251	000055	000055	000061	000062	000048	000066	000062	000069	000080	000071
261	000068	000068	000064	000095	000082	000108	000107	000092	000092	000125
271	000107	000105	000115	000129	000135	000164	000156	000159	000194	000210
281	000209	000223	000289	000250	000285	000294	000320	000430	000447	000537
291	000528	000637	000734	000847	001029	001186	001396	001670	001890	002211
301	002357	002314	002272	002036	001719	001572	001421	001397	001549	001775
311	002206	002702	003230	003774	004235	004668	004708	004382	003806	002917
321	001867	001189	000622	000310	000158	000078	000048	000044	000034	000029
331	000031	000023	000030	000018	000019	000026	000023	000018	000021	000021
341	000018	000017	000019	000017	000017	000023	000019	000024	000022	000015
351	000017	000017	000021	000012	000018	000016	000020	000012	000021	000016
361	000022	000014	000016	000016	000009	000012	000019	000015	000013	000009
371	000015	000008	000009	000013	000014	000009	000015	000009	000013	000017
381	000016	000015	000012	000014	000016	000014	000014	000012	000012	000018
391	000011	000011	000011	000019	000019	000007	000011	000015	000014	000008

CHANNEL CONTENT: SAMPLE C 1 I

1	003600	000000	000000	000000	000000	000000	000000	000000	000000	000000
11	000000	000022	000031	000035	000032	000035	000037	000026	000032	000046
21	000026	000038	000027	000026	000031	000035	000037	000024	000033	000022
31	000032	000029	000025	000041	000030	000032	000029	000041	000036	000039
41	000044	000041	000032	000034	000035	000039	000042	000029	000046	000039
51	000029	000030	000038	000036	000032	000031	000048	000039	000031	000035
61	000044	000046	000045	000049	000036	000049	000044	000046	000043	000050
71	000043	000057	000059	000035	000053	000062	000054	000041	000049	000056
81	000056	000070	000062	000074	000058	000069	000064	000054	000065	000075
91	000072	000071	000068	000082	000061	000068	000064	000069	000071	000060
101	000070	000091	000080	000087	000069	000091	000088	000104	000093	000111
111	000091	000081	000102	000085	000107	000109	000105	000111	000101	000113
121	000103	000124	000128	000124	000106	000135	000129	000136	000146	000159
131	000151	000155	000175	000179	000180	000160	000169	000190	000215	000187
141	000206	000218	000262	000191	000233	000240	000251	000275	000241	000287
151	000312	000319	000358	000306	000387	000402	000399	000414	000450	000501
161	000514	000601	000614	000627	000705	000794	000808	000888	000924	001085
171	001115	001213	001256	001332	001388	001549	001584	001611	001679	001747
181	001943	002011	002193	002256	002467	002705	002920	003059	003138	003208
191	003261	003239	003108	002780	002439	002133	001609	001214	000792	000610
201	000392	000211	000132	000056	000047	000032	000038	000033	000025	000022
211	000037	000026	000038	000033	000028	000034	000030	000038	000041	000023
221	000033	000029	000026	000046	000042	000036	000034	000044	000039	000055
231	000046	000042	000043	000041	000031	000049	000050	000052	000066	000063
241	000052	000060	000081	000066	000070	000047	000075	000071	000090	000068
251	000068	000083	000089	000083	000080	000094	000075	000090	000111	000104
261	000100	000103	000107	000118	000096	000110	000116	000124	000144	000130
271	000145	000141	000168	000147	000192	000178	000178	000201	000209	000251
281	000234	000256	000285	000318	000331	000333	000408	000382	000474	000508
291	000552	000617	000608	000751	000807	000827	000936	000964	001049	001110
301	001115	001134	001081	001113	001079	001075	001046	001087	001202	001336
311	001359	001491	001644	001622	001696	001612	001590	001352	001111	000873
321	000549	000386	000202	000095	000044	000012	000004	000001	000001	000001
331	000000	000000	000001	000000	000000	000000	000000	000000	000000	000000
341	000000	000000	000000	000000	000003	000001	000000	000001	000001	000000
351	000000	000000	000000	000000	000000	000001	000000	000000	000000	000000
361	000000	000000	000000	000001	000000	000000	000000	000001	000000	000000
371	000000	000000	000000	000000	000000	000001	000000	000000	000000	000001
381	000001	000000	000001	000000	000000	000000	000000	000000	000000	000000
391	000000	000000	000001	000000	000000	000000	000001	000000	000002	000001

CHANNEL CONTENT: SAMPLE C 1 II

1	003600	000000	000000	000000	000000	000000	000000	000000	000000	000000
11	000003	000031	000036	000034	000031	000032	000047	000027	000023	000032
21	000031	000039	000041	000027	000032	000041	000035	000023	000023	000032
31	000037	000025	000032	000027	000031	000018	000031	000046	000029	000041
41	000033	000028	000032	000035	000045	000037	000032	000031	000033	000043
51	000047	000032	000048	000050	000038	000045	000029	000039	000033	000033
61	000037	000053	000040	000030	000040	000034	000032	000038	000043	000047
71	000046	000044	000036	000045	000050	000052	000050	000046	000050	000063
81	000054	000049	000060	000057	000065	000068	000050	000076	000046	000061
91	000055	000070	000070	000086	000071	000076	000082	000082	000083	000072
101	000067	000072	000070	000088	000084	000097	000080	000091	000085	000090
111	000084	000097	000120	000088	000109	000097	000107	000112	000103	000101
121	000110	000115	000125	000099	000111	000126	000132	000130	000136	000152
131	000148	000175	000148	000171	000153	000171	000188	000177	000207	000192
141	000195	000189	000254	000243	000246	000232	000239	000277	000257	000267
151	000286	000325	000320	000340	000384	000372	000416	000430	000470	000467
161	000490	000517	000641	000675	000677	000797	000825	000879	000938	001003
171	001137	001207	001290	001396	001407	001515	001582	001623	001738	001737
181	001815	002131	002231	002444	002495	002696	002921	003067	003225	003220
191	003290	003191	003093	002726	002393	002069	001573	001167	000797	000489
201	000359	000216	000146	000077	000034	000040	000033	000022	000026	000029
211	000030	000029	000023	000035	000034	000027	000037	000035	000044	000035
221	000042	000022	000039	000037	000041	000040	000041	000049	000057	000030
231	000041	000039	000045	000039	000043	000044	000053	000044	000063	000052
241	000060	000055	000056	000066	000054	000053	000055	000063	000055	000064
251	000074	000085	000076	000081	000081	000072	000078	000082	000097	000080
261	000101	000117	000097	000102	000102	000121	000129	000111	000142	000142
271	000157	000139	000143	000171	000186	000190	000193	000227	000223	000251
281	000252	000259	000299	000341	000282	000354	000389	000425	000461	000521
291	000554	000616	000665	000706	000851	000921	000960	000995	001074	001055
301	001153	001123	001209	001124	001011	001022	001030	001110	001198	001254
311	001430	001521	001575	001562	001680	001648	001512	001397	001119	000902
321	000595	000354	000193	000091	000036	000016	000004	000001	000000	000000
331	000000	000000	000003	000000	000000	000000	000000	000001	000001	000000
341	000000	000000	000000	000000	000000	000000	000001	000000	000000	000000
351	000000	000000	000000	000000	000000	000000	000000	000000	000000	000001
361	000000	000000	000000	000000	000000	000000	000000	000000	000000	000000
371	000000	000000	000001	000000	000000	000000	000000	000000	000000	000000
381	000000	000001	000000	000000	000000	000000	000000	000000	000000	000000
391	000000	000001	000000	000001	000000	000000	000000	000000	000001	000000

CHANNEL CONTENT: SAMPLE C 1 III

1	003600	000000	000000	000000	000000	000000	000000	000000	000000	000000
11	000000	000021	000036	000034	000029	000030	000026	000030	000039	000038
21	000030	000038	000018	000036	000026	000019	000031	000031	000030	000034
31	000033	000037	000024	000028	000024	000029	000040	000031	000029	000028
41	000026	000029	000039	000027	000024	000044	000033	000045	000030	000044
51	000034	000036	000031	000032	000039	000043	000031	000043	000034	000049
61	000056	000035	000045	000045	000041	000036	000043	000051	000037	000052
71	000051	000036	000043	000061	000054	000051	000055	000051	000057	000057
81	000055	000059	000057	000058	000071	000058	000061	000066	000056	000070
91	000065	000074	000059	000063	000070	000085	000064	000062	000080	000066
101	000075	000069	000093	000075	000088	000099	000086	000078	000074	000083
111	000091	000100	000106	000104	000098	000104	000121	000100	000111	000119
121	000126	000120	000140	000142	000142	000139	000125	000118	000140	000147
131	000193	000155	000170	000169	000174	000176	000192	000169	000177	000219
141	000200	000205	000225	000229	000218	000248	000258	000257	000277	000302
151	000308	000314	000317	000350	000344	000385	000421	000444	000423	000509
161	000502	000569	000581	000662	000653	000743	000774	000895	000952	001021
171	001139	001193	001314	001338	001489	001461	001497	001605	001769	001821
181	001823	002065	002292	002405	002599	002740	002898	003022	003134	003293
191	003151	003239	003036	002720	002456	002039	001581	001196	000849	000542
201	000379	000233	000131	000087	000048	000027	000039	000035	000030	000036
211	000031	000024	000033	000037	000029	000033	000027	000036	000030	000033
221	000036	000035	000032	000048	000031	000030	000046	000039	000038	000059
231	000046	000051	000055	000043	000056	000056	000059	000055	000059	000054
241	000057	000053	000066	000073	000059	000071	000064	000065	000064	000063
251	000081	000068	000087	000063	000075	000091	000074	000096	000092	000092
261	000125	000120	000102	000091	000113	000116	000124	000127	000148	000147
271	000154	000157	000166	000184	000161	000189	000199	000205	000206	000238
281	000251	000271	000245	000276	000311	000330	000401	000399	000480	000494
291	000582	000573	000640	000773	000813	000858	000978	000986	001043	001125
301	001164	001142	001115	001119	001030	001070	001035	001153	001279	001366
311	001473	001601	001517	001729	001669	001603	001501	001283	001106	000857
321	000591	000373	000192	000095	000037	000013	000001	000002	000004	000000
331	000000	000002	000000	000000	000001	000000	000001	000000	000001	000000
341	000000	000000	000000	000001	000000	000000	000000	000001	000000	000000
351	000000	000000	000000	000000	000000	000000	000000	000000	000000	000000
361	000000	000000	000000	000001	000000	000000	000000	000000	000000	000000
371	000000	000000	000000	000001	000001	000001	000000	000000	000001	000000
381	000000	000000	000000	000000	000000	000000	000000	000000	000000	000000
391	000000	000000	000001	000000	000000	000000	000000	000002	000000	000000

CHANNEL CONTENT: SAMPLE C 2 I

1	003600	000000	000000	000000	000000	000000	000000	000000	000000	000000
11	000000	000004	000008	000003	000002	000001	000004	000004	000003	000005
21	000002	000002	000004	000001	000004	000001	000000	000002	000001	000000
31	000003	000002	000005	000001	000003	000002	000001	000006	000002	000004
41	000004	000002	000004	000001	000002	000001	000000	000002	000000	000001
51	000002	000004	000001	000001	000003	000004	000002	000001	000000	000003
61	000006	000002	000003	000005	000000	000005	000004	000002	000004	000006
71	000002	000002	000004	000001	000001	000007	000006	000006	000005	000003
81	000003	000003	000002	000008	000004	000005	000002	000006	000004	000007
91	000007	000006	000006	000004	000009	000002	000011	000008	000004	000008
101	000013	000009	000004	000004	000007	000008	000009	000012	000005	000007
111	000005	000012	000013	000013	000006	000011	000015	000013	000009	000017
121	000013	000026	000022	000012	000018	000013	000024	000018	000022	000020
131	000017	000034	000033	000031	000025	000032	000028	000032	000040	000036
141	000048	000039	000046	000048	000060	000055	000047	000068	000057	000068
151	000081	000071	000085	000075	000074	000082	000105	000091	000111	000123
161	000119	000131	000141	000160	000162	000238	000187	000239	000275	000293
171	000349	000414	000492	000514	000595	000563	000575	000567	000564	000581
181	000605	000647	000672	000728	000870	000999	001132	001337	001580	001821
191	001964	002127	002084	002141	001936	001588	001304	000923	000617	000430
201	000272	000167	000081	000033	000014	000008	000001	000000	000000	000003
211	000000	000002	000000	000004	000003	000002	000005	000002	000002	000000
221	000004	000002	000003	000004	000005	000004	000002	000002	000007	000003
231	000005	000008	000004	000007	000004	000005	000001	000005	000009	000010
241	000002	000007	000008	000008	000007	000009	000008	000012	000011	000010
251	000013	000005	000017	000020	000013	000025	000014	000017	000023	000027
261	000019	000030	000025	000019	000023	000024	000035	000021	000026	000024
271	000039	000025	000039	000051	000053	000038	000054	000059	000055	000050
281	000057	000065	000071	000078	000065	000085	000084	000131	000125	000121
291	000141	000162	000190	000218	000252	000305	000338	000428	000488	000524
301	000619	000587	000571	000547	000457	000389	000356	000330	000399	000494
311	000557	000621	000769	000902	001023	001091	001126	001094	001013	000757
321	000501	000291	000138	000053	000017	000007	000001	000000	000000	000000
331	000000	000001	000000	000000	000000	000001	000000	000000	000000	000000
341	000000	000000	000000	000000	000000	000000	000001	000000	000000	000000
351	000000	000000	000000	000000	000000	000000	000000	000000	000000	000000
361	000001	000000	000000	000000	000000	000000	000000	000000	000000	000000
371	000000	000000	000000	000001	000000	000000	000000	000000	000001	000002
381	000000	000001	000000	000000	000000	000000	000000	000001	000000	000001
391	000000	000000	000000	000000	000000	000000	000000	000001	000000	000000

CHANNEL CONTENT: SAMPLE C 2 II

1	003600	000000	000000	000000	000000	000000	000000	000000	000000	000000
11	000000	000001	000002	000003	000000	000000	000004	000002	000003	000003
21	000002	000003	000000	000002	000002	000004	000002	000003	000001	000001
31	000002	000002	000001	000002	000003	000002	000003	000000	000002	000001
41	000003	000003	000003	000001	000001	000001	000000	000000	000003	000002
51	000002	000003	000002	000004	000001	000008	000003	000000	000002	000002
61	000004	000004	000002	000000	000003	000005	000008	000001	000000	000001
71	000005	000002	000005	000003	000005	000007	000003	000001	000002	000003
81	000004	000003	000002	000004	000002	000006	000004	000003	000009	000010
91	000003	000006	000006	000003	000001	000008	000002	000008	000005	000005
101	000011	000007	000006	000010	000009	000010	000014	000009	000011	000007
111	000010	000013	000012	000008	000014	000012	000006	000011	000011	000007
121	000013	000020	000014	000020	000025	000011	000023	000017	000019	000019
131	000029	000024	000034	000021	000033	000021	000027	000027	000040	000035
141	000039	000031	000044	000048	000054	000055	000054	000061	000059	000059
151	000060	000091	000086	000082	000085	000077	000097	000111	000110	000147
161	000136	000136	000172	000169	000182	000196	000226	000232	000241	000311
171	000350	000450	000430	000540	000556	000578	000625	000540	000530	000604
181	000625	000646	000644	000792	000883	001077	001126	001351	001517	001713
191	001930	002090	002086	002053	001925	001580	001275	000952	000623	000422
201	000293	000157	000096	000040	000015	000012	000001	000004	000002	000000
211	000003	000000	000003	000001	000005	000003	000000	000005	000006	000004
221	000005	000004	000005	000004	000006	000003	000004	000001	000007	000005
231	000002	000004	000002	000006	000005	000008	000006	000005	000003	000008
241	000005	000007	000007	000013	000016	000015	000011	000010	000013	000008
251	000008	000018	000012	000017	000016	000018	000016	000022	000019	000024
261	000022	000024	000025	000022	000027	000024	000030	000028	000025	000036
271	000032	000043	000033	000049	000036	000048	000056	000052	000070	000064
281	000063	000068	000088	000092	000081	000082	000103	000100	000101	000102
291	000150	000148	000165	000209	000256	000277	000346	000457	000475	000545
301	000599	000616	000572	000531	000482	000401	000340	000342	000331	000439
311	000489	000630	000744	000908	001031	001141	001198	001166	000931	000789
321	000498	000320	000145	000062	000019	000005	000001	000001	000000	000000
331	000001	000001	000000	000001	000001	000000	000000	000000	000000	000000
341	000000	000000	000000	000000	000000	000000	000000	000001	000000	000000
351	000000	000000	000000	000000	000000	000000	000000	000001	000000	000000
361	000001	000000	000000	000000	000000	000000	000000	000001	000000	000000
371	000000	000001	000000	000000	000000	000000	000000	000000	000003	000000
381	000000	000000	000000	000000	000000	000000	000001	000001	000000	000001
391	000001	000000	000000	000000	000001	000000	000001	000000	000001	000000

CHANNEL CONTENT: SAMPLE C 2 III

1	003600	000000	000000	000000	000000	000000	000000	000000	000000	000000
11	000000	000001	000005	000000	000001	000003	000001	000002	000001	000000
21	000004	000005	000000	000004	000002	000003	000001	000001	000002	000002
31	000001	000002	000002	000001	000002	000000	000001	000006	000001	000002
41	000000	000003	000001	000006	000002	000002	000003	000001	000002	000002
51	000004	000001	000002	000000	000004	000002	000001	000003	000001	000001
61	000004	000003	000003	000003	000000	000000	000005	000004	000004	000001
71	000007	000008	000002	000003	000005	000004	000002	000003	000005	000004
81	000003	000009	000002	000010	000002	000001	000002	000003	000003	000004
91	000004	000010	000005	000006	000005	000007	000008	000007	000006	000007
101	000007	000011	000009	000007	000006	000009	000010	000008	000010	000012
111	000017	000013	000008	000014	000014	000007	000012	000015	000011	000011
121	000018	000012	000019	000010	000028	000014	000022	000031	000031	000019
131	000022	000021	000024	000029	000036	000029	000031	000030	000036	000045
141	000045	000038	000048	000050	000052	000061	000060	000063	000074	000068
151	000079	000081	000099	000079	000086	000096	000093	000117	000099	000115
161	000137	000115	000161	000150	000183	000197	000206	000245	000261	000317
171	000349	000441	000396	000531	000535	000580	000585	000577	000564	000589
181	000569	000655	000735	000794	000924	000933	001147	001344	001558	001702
191	001911	001981	002106	002083	001809	001619	001340	000955	000674	000466
201	000293	000165	000074	000039	000017	000008	000002	000002	000003	000000
211	000003	000001	000004	000000	000001	000003	000003	000003	000003	000006
221	000004	000004	000002	000002	000005	000002	000005	000001	000007	000004
231	000007	000005	000003	000005	000005	000005	000006	000004	000004	000012
241	000006	000006	000008	000013	000008	000008	000005	000009	000013	000011
251	000016	000013	000012	000017	000019	000009	000012	000019	000024	000025
261	000020	000025	000017	000027	000027	000029	000036	000037	000041	000025
271	000032	000031	000041	000040	000049	000040	000071	000062	000047	000057
281	000061	000080	000068	000060	000086	000093	000107	000123	000109	000122
291	000151	000157	000169	000204	000212	000298	000342	000408	000475	000528
301	000578	000608	000589	000514	000484	000374	000349	000350	000387	000442
311	000496	000611	000791	000897	001042	001163	001167	001092	000970	000771
321	000536	000329	000160	000072	000035	000009	000000	000002	000001	000001
331	000002	000000	000002	000003	000001	000001	000000	000000	000000	000000
341	000000	000000	000000	000001	000000	000000	000000	000000	000000	000000
351	000000	000000	000000	000000	000000	000000	000000	000000	000001	000000
361	000000	000000	000000	000000	000000	000001	000000	000000	000000	000000
371	000000	000000	000001	000000	000001	000000	000001	000000	000000	000000
381	000000	000000	000000	000001	000000	000002	000000	000000	000000	000001
391	000001	000000	000000	000000	000000	000000	000000	000000	000000	000000

APPENDIX II:

Data Reported by the Laboratories

Remark: The spectra A2 I and B2 III were not used for evalution. Therefore data reported on these spectra are put in brackets.

LABORATORY CODE: 1

RESULTS OF THE ALPHA SPECTRA EVALUATION.

PEAK AREA / SAMPLE		Pu-238 [counts]	Pu-239 + Pu-240 [counts]
A 1	I	30121	8676
	II	32277	8053
	III	31960	9118
A 2	I	(36403)	(8485)
	II	38308	8168
	III	38770	9475
B 1	I	83081	29729
	II	70309	36317
	III	69060	30956
B 2	I	73618	33739
	II	71284	33052
	III	(66824)	(31552)
C 1	I	35937	66229
	II	36583	92099
	III	35848	70333
C 2	I	17128	34478
	II	16564	28829
	III	15749	30762

LABORATORY CODE: 2

RESULTS OF THE ALPHA SPECTRA EVALUATION.

SAMPLE	PEAK AREA	Pu-238 [counts]	Pu-239 + Pu-240 [counts]
A 1	I	44242	11177
	II	44683	11285
	III	44731	1o961
A 2	I	(41296)	(1o526)
	II	41o2o	1o464
	III	41617	1o4o3
B 1	I	1o1767	44171
	II	1o1695	44435
	III	1oo279	44421
B 2	I	77354	36617
	II	77446	368o2
	III	(81167)	(39o59)
C 1	I	46o1o	79664
	II	46116	79461
	III	46281	79779
C 2	I	19767	35265
	II	19933	351o4
	III	19934	351o7

LABORATORY CODE: 3

RESULTS OF THE ALPHA SPECTRA EVALUATION.

SAMPLE \ PEAK AREA		Pu-238 [counts]	Pu-239 + Pu-240 [counts]
A 1	I	45411	11514
	II	45483	11796
	III	45347	115o9
A 2	I	-	-
	II	4o813	1o6o4
	III	41331	1o41o
B 1	I	1o1533	46642
	II	1o1o2o	46648
	III	99715	46548
B 2	I	76956	36772
	II	77o68	36943
	III	-	-
C 1	I	44988	82924
	II	45374	819o2
	III	45398	82856
C 2	I	19697	35438
	II	197o9	35254
	III	19736	353o9

LABORATORY CODE: 4

RESULTS OF THE ALPHA SPECTRA EVALUATION.

SAMPLE \ PEAK AREA		Pu-238 [counts]	Pu-239 + Pu-240 [counts]
A 1	I	40885	12419
	II	41217	12708
	III	41272	12389
A 2	I	~~(40665)~~	~~(10649)~~
	II	40586	10632
	III	41020	10378
B 1	I	95864	45944
	II	96342	45612
	III	94867	45977
B 2	I	76585	36617
	II	76654	36845
	III	(77839)	(39219)
C 1	I	44005	81292
	II	44177	81098
	III	44436	81324
C 2	I	19522	34987
	II	19557	34878
	III	19590	34831

LABORATORY CODE: 5

RESULTS OF THE ALPHA SPECTRA EVALUATION.

SAMPLE	PEAK AREA	Pu-238 [counts]	Pu-239 + Pu-240 [counts]
A 1	I	45153	12898
	II	45419	13319
	III	46184	12144
A 2	I	(41o26)	(1o733)
	II	4o849	1o6o9
	III	41358	1o41o
B 1	I	1oo584	47585
	II	1o3o42	47768
	III	1o1236	48242
B 2	I	77o59	36762
	II	77131	37142
	III	(77642)	(39723)
C 1	I	46448	82886
	II	46272	83265
	III	46635	82952
C 2	I	19667	35173
	II	19785	34452
	III	19789	35o31

LABORATORY CODE: 6

RESULTS OF THE ALPHA SPECTRA EVALUATION.

SAMPLE \ PEAK AREA		Pu-238 [counts]	Pu-239 + Pu-240 [counts]
A 1	I	41467	7388
	II	41797	7651
	III	41893	7202
A 2	I	(41049)	(10023)
	II	40857	10365
	III	41369	10139
B 1	I	96743	37713
	II	97287	36726
	III	95832	36751
B 2	I	77078	36293
	II	77153	36427
	III	(79517)	(35771)
C 1	I	45351	74269
	II	45448	74081
	III	45629	74184
C 2	I	19721	34951
	II	19790	34825
	III	19804	34824

LABORATORY CODE: 7

RESULTS OF THE ALPHA SPECTRA EVALUATION.

SAMPLE	PEAK AREA	Pu-238 [counts]	Pu-239 + Pu-240 [counts]
A 1	I	49662	12528
	II	5o155	12889
	III	5o337	123o1
A 2	I	(48156)	(11591)
	II	4o886	1o616
	III	41428	1o399
B 1	I	1o438o	49417
	II	1o494o	493o5
	III	1o299o	49239
B 2	I	77369	36786
	II	77331	36994
	III	(1o4o7o)	(47242)
C 1	I	46555	84222
	II	47754	85651
	III	472o1	8469o
C 2	I	19772	354o7
	II	19849	35341
	III	19842	35265

LABORATORY CODE: 8

RESULTS OF THE ALPHA SPECTRA EVALUATION.

SAMPLE	PEAK AREA	Pu-238 [counts]	Pu-239 + Pu-240 [counts]
A 1	I	46112	12246
	II	46514	12534
	III	45994	12635
A 2	I	(40972)	(12242)
	II	41037	10630
	III	41620	10379
B 1	I	100899	50715
	II	102003	49558
	III	100056	50375
B 2	I	77574	36804
	II	77677	37002
	III	(79064)	(49578)
C 1	I	45044	84602
	II	45256	84216
	III	45854	84069
C 2	I	19854	35541
	II	19907	35407
	III	19985	35347

LABORATORY CODE: 9

RESULTS OF THE ALPHA SPECTRA EVALUATION.

SAMPLE \ PEAK AREA		Pu-238 [counts]	Pu-239 + Pu-240 [counts]
A 1	I	4o984	1o245
	II	41366	1o437
	III	41255	1o191
A 2	I	(41586)	(1o436)
	II	4o849	1o435
	III	41311	1o259
B 1	I	-	-
	II	-	-
	III	-	-
B 2	I	77o52	36524
	II	77144	37181
	III	-	-
C 1	I	44436	79859
	II	44547	79752
	III	44849	79844
C 2	I	1972o	35492
	II	19846	35455
	III	19833	35195

LABORATORY CODE: 1o

RESULTS OF THE ALPHA SPECTRA EVALUATION.

SAMPLE \ PEAK AREA		Pu-238 [counts]	Pu-239 + Pu-240 [counts]
A 1	I	33338	7822
	II	338o6	8o38
	III	33577	7743
A 2	I	(4o359)	(1o66o)
	II	4o297	1o329
	III	4o685	1o143
B 1	I	8o816	375o3
	II	78o75	36858
	III	77511	36682
B 2	I	75933	36oo5
	II	761oo	36242
	III	(75216)	(35343)
C 1	I	4o84o	73319
	II	4o976	73341
	III	41oo2	73o93
C 2	I	19335	34454
	II	19298	34378
	III	19294	34355

APPENDIX III:

Descriptions of the Evaluation Methods
Used by the Laboratories

LABORATORY CODE: 1

TYPE OF SPECTRA: Digital and Paper Tape

Short description of evaluation method used:

A) Determination of Peak Boundaries

1. For Pu-238

a) Higher energy side:
The first channel after the top of the peak having a content of 0 or 1 was defined as boundary.

b) Lower energy side:

I. For well resolved spectra:
Within the range of channels having a content between 5 and 15 % of the content at the peak top, that channel was used as peak boundary which had a content significantly smaller than the arithmetic mean of the contents of the two adjacent channels.

II. For bad resolved spectra:
As described in I., however the range was shifted to 15 to 25 % of the content at the peak top in order to obtain a clear delimitation from the Pu-239/240 peak.

2. For Pu-239/240

a) Higher energy side:

I. For well resolved spectra:
The first channel after the peak top having a content of 0 or 1 was defined as boundary.

II. For bad resolved spectra:
The first channel after the peak top with a content smaller than the arithmetic mean of the adjacent channels was used as boundary.

b) Lower energy side:

I. For well resolved spectra:
As described for Pu-238, section I.

II. For bad resolved spectra:
As described for Pu-238, section II.

B) Calculation of Peak Area

a) The contents of all channels in between the boundaries defined in section A) were summed up. The contents of the limiting channels were taken into consideration.

b) In order to obtain the peak area, from this sum the product of the arithmetic mean of the contents of the peak boundary channels with the number of channels was subtracted.

LABORATORY CODE: 2

TYPE OF SPECTRA: Digital

Short description of evaluation method used:

1. We prepare our targets principally by electrodeposition on platinum. The spectra of the evaporated samples and of sample B 2 III are so bad, that we would not evaluate them in routine operation.

2. For spectra evaluation we used the following formulae:

   Pu-238 = Σ channels 210 to 329 + U
   Pu-(239+240) = Σ channels 100 to 209 - U
   U = balanced value of channel 210 times 50

3. The evaluation is only directed to the determination of the ratio Pu-238/Pu-(239+240). It is assumed that the counts of the channels 1 to 100 have to be attributed in the ratio of the two groups.

LABORATORY CODE: 3

TYPE OF SPECTRA: Digital

Short description of evaluation method used:

1. Direct Evaporated Sources

Least squares fitting was done assuming the degraded portion to be exponential in nature. The contribution of Pu-238 peak under (Pu-239 + Pu-240) peak was determined by extrapolating the exponential curve obtained by least squares fitting of the degraded portion of the Pu-238 peak.

2. Electrodeposited Sources

a) For the electrodeposited sources, the peak areas were calculated by summing up the counts under the peaks.

b) In the case of sample A 2, the counting statistics is not good. The sample should have been counted for a longer time.

c) Samples A 2(I) and B 2(III) had more degradation though the alpha spectra were taken on the same electrodeposited sources. As the reason for this is not known, the values from these two spectra are not reported.

LABORATORY CODE: 4

TYPE OF SPECTRA: Digital

Short description of evaluation method used:

Sample No.:	A_2)	peakarea	Pu-238	: Σ 82 channels-from 251 to 332
	B_2)	peakarea	Pu-(239+240)	: Σ 82 channels-from 131 to 212
	C_2)			

Sample No.:	C_1	peakarea	Pu-238	: Σ 111 channels-from 220 to 330
		peakarea	Pu-(239+240)	: Σ 111 channels-from 102 to 212

Sample No.:	A_1-)	peakarea	Pu-238	: Σ 124 channels-from 209 to 332
	B_1:)	peakarea	Pu-(239+240)	: Σ 124 channels-from 85 to 208

Pu-238 background of channels 120 to 208 subtracted

Data-Sheet - Plutonium - Isotopic Analysis Code 4

Date:	No.:	KT:	Operator

Pu 238 :	$T_{1/2}$	= 87,7 a	spec. act.	=	17,1 Ci/g
Pu 239 :	$T_{1/2}$	= 24400 a	"	=	0,062 Ci/g
Pu 240 :	$T_{1/2}$	= 6600 a	"	=	0,226 Ci/g
Pu 241 :	$T_{1/2}$	= 14,8 a	" (α)	=	0,0025 Ci/g

Pu - Isotopes

Alpha Spectrometry

Impuls Rate:

Imp. Pu 238: = P 1

Imp. Pu 239: = P 2
+ 240
+ 241

Mass Spectrometry

Mass Ratios

Pu 240/239: =CM

Pu 241/239: =EM

Pu 242/239 =FM

KM = 1 + CM + EM + FM

=

$$\frac{P\,1}{P\,2 \times 17{,}1} = A$$

$$\frac{1}{KM} \times 0{,}062 + \frac{CM}{KM} \times 0{,}226 + \frac{EM}{KM} \times 0{,}0025 = B$$

	atom %	mass:	wt. %
$\frac{A \times B \times 100}{1 + (A \times B)}$ = % Pu 238 =		238.05 =	
$\frac{100\,\% - \%\,Pu\,238}{KM}$ = % Pu 239 =		239.05 =	
% Pu 239 x CM = % Pu 240 =		240.05 =	
% Pu 239 x EM = % Pu 241 =		241.06 =	
% Pu 239 x FM = % Pu 242 =		242.06 =	
sum =			
Pu 239 + 241 = Pu fiss =			

LABORATORY CODE: 5

TYPE OF SPECTRA: Digital

Short description of evaluation method used:

1. The evaluation method should be separated to two cases as follows; case 1; two peak areas required to separate are clearly separated, case 2; the peak area of the higher energy side tails to the other peak area of the lower energy side.

2. In case of the case 1, each total counts should be calculated in each peak area.,

   (A-2, B-2, and C-2 samples were determined with this method.)

3. In case of the case 2, the determinations should be carried out by the following prodecure:

3.1 General base of the determinations

The calculation is carried out on the assumption that the tailing of any single peak to the lower energy region is caused only by the sample thickness. The differential count N(E)dE is able to described on as the following equations:

$$E_0 > E > E_0 - (dE/dx)_{E_0} \cdot t \qquad N(E)dE = A \qquad (1)$$

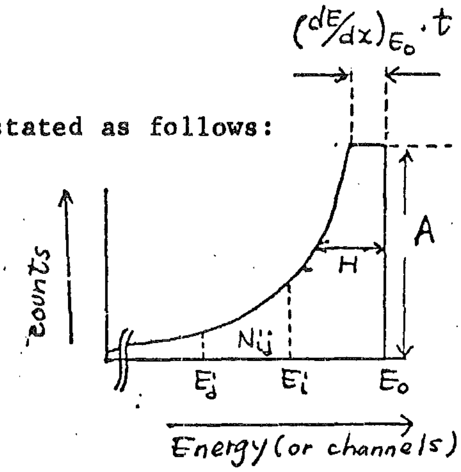
$$E_0 - (dE/dx)_{E_0} \cdot t > E \qquad N(E)dE = A((dE/dx)_{E_0} \cdot t)^2/(E_0-E)^2 \qquad (2)$$

$H = \sqrt{2} \cdot (dE/dx)_{E_0} \cdot t$, where H is HWFM.

The total counts, N_{ij}, between the energy (or the channel) E_i and E_j are as follows

$$N_{ij} = \int_{E_j}^{E_i} N(E)\,dE$$

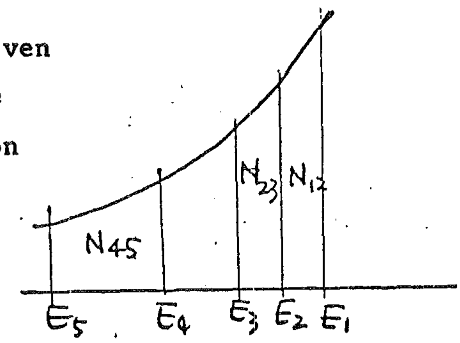
Accordingly, N_{ij} on the tail region is stated as follows:



$$N_{ij} = K(E_i-E_j)/(E_i-E_0)(E_j-E_0) \qquad (3)$$

where $K \equiv AH^2/2$.

3.2 When N_{12}, N_{23}, E_1, E_2, and E_3 are given by the experimental work, N_{45} can be calculated with the following equation derived from the equation (3):



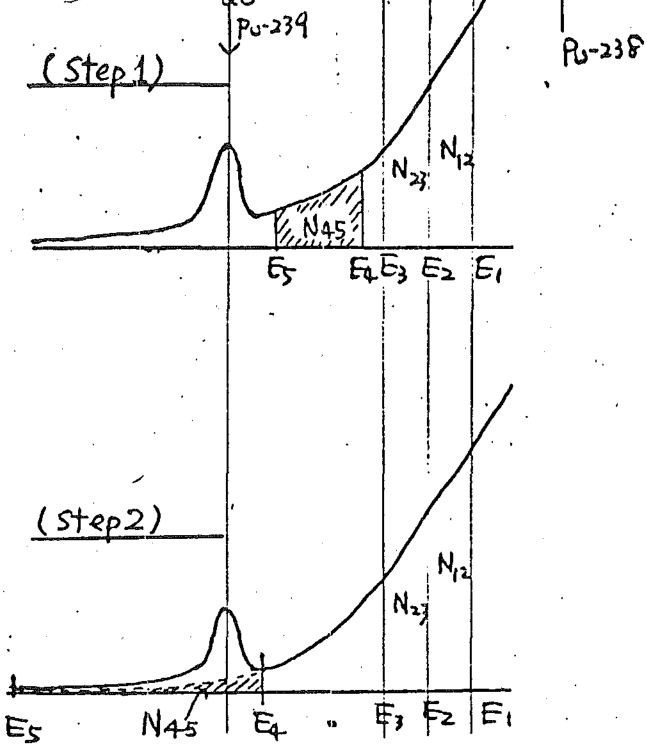
$$N_{45} = N_{12}N_{23}(N_{12}+N_{23})(E_1-E_3+1)(E_4-E_5+1)/(N_{12}(E_1-E_4+1)-N_{23}(E_3-E_4+1))(N_{12}(E_1-E_5+1)-N_{23}(E_3-E_5+1)) \qquad (4)$$

however the following condition should be satisfied;

$E_1-E_2 = E_2-E_3$.

3.3 The method of evaluation of the activity ratio of Pu-238 to Pu-239(+Pu-240 +Pu-241 +Pu-242)

Step 1. E_1 and E_3 should be chosen as the channels which give the best value N_{45} compared with the actual value by the use of the equation (4).



(A-1-I------E_1=290, E_3=270, and the calculated value N_{45} was 7359 counts between the channels 270 and 210. The actual value was 7387.)

Step 2. Then, the total counts N_{45} by the tail of the Pu-238 between E_4 and E_5 of the energy region of the Pu-239 should be calculated by the same manner of the step 1. (A-1-I------N_{45} was 4443 counts between the channels 210 and 21.)

Step 3. Finally, the acivity ratio of Pu-238 (total counts P_8) to Pu-239 (total counts P_9) can be determined as follows:

$P_9 = M_{45} - N_{45}$

$P_8 = T - P_9$, where M_{45} is the total counts between E_4 and E_5 at the step 2, and T is the total counts of the energy regions of Pu-238 and Pu-239. (A-1-I------17341 for M_{45} and 58051 for T were counted. Accordingly, P_9 and P_8 were determined 12898 and 45153, respectively.)

LABORATORY CODE: 6

TYPE OF SPECTRA: Paper Tape

Short description of evaluation method used:

Using a Digital Equipment Corp. PDP-15 computer paper tape reader, the low energy (plutonium 239,40) and high energy plutonium 238) peak areas, between predetermined channels, were read in and integrated. At the same time areas of 10 channels above and below the low energy peak were read and used to calculate the "toe" correction resulting from low energy scatter from the high energy peak.

Normally, when analyzing plutonium for isotope 238, our sample plates do not contain nearly as much solid material and we do not contain nearly as much solid material and we do not require a "toe" correction as was done above. Thus the above procedure was devised just to analyze these spectra. In addition, we were not able to read the paper tape reader and thus were required to use the reader on our computer.

LABORATORY CODE: 7

TYPE OF SPECTRA: Digital and Paper Tape

Short description of evaluation method used:

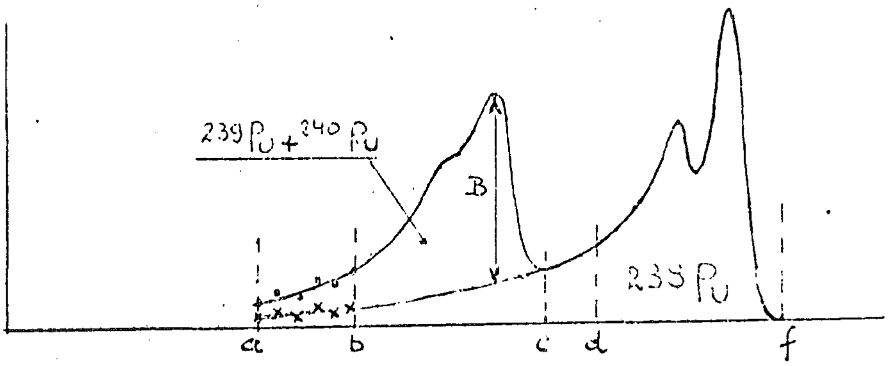
1. Fitting Model

According to M.P. Trivedi (Progress Report 1969-1970, NYO-844-81) the assumption is made that to each α-ray decay energy there corresponds a single peak spectrum (SPS) which can be described by the function

(1)
$$y_i(E) = A1_i \exp\left(\frac{-(E-A3_i)^2}{A2\left[1+A5_i(E-A3_i)^{A4}\right]} \right)$$

This model has the peak height $A1_i$, resolution parameter A2, peak energy $A3_i$, skewing parameter A4 and tailing parameter $A5_i$. For the ^{238}Pu group i is 1,2 whereas for the ^{239}Pu+^{240}Pu group i is 3,4,5,6,7. All SPS are presumed to have equal shape parameters A2, A4, $A5_i$. The high energy part of the single peaks is presumed to be of Gaussian shape hence $A5_i$=0 for $E \geq A3_i$. The set of parameters A4, A5 which applies to the tail (that is below d for the ^{238}Pu group and below b for the ^{239}Pu+^{240}Pu group) is different from that for the peak regions (d,f) and (b,c).

2. Method of evaluation



The parameters of the ^{238}Pu group are obtained from an iterative procedure of nonlinear least squares fits of a sum of two functions Eq (1) to the data in the region (d,f). From this the energy offset is also found.

The contribution of the ^{238}Pu group to the experimental data in the region (a,b) is found by iteration as follows: With an assumed fractional contribution of ^{238}Pu in (a,b), the tail (a,d) of the ^{238}Pu group is fitted and gives the parameters A4 A5 in that region. Using these parameters and assuming that the heights of the ^{239}Pu and ^{240}Pu subgroups are both B/2, the fractional contribution of ^{238}Pu in (a,b) is calculated.

The area under the ^{238}Pu group is obtained by integrating the tail in the region (0,c) and by summing up all data in (c,f). For the $^{239}Pu+^{240}Pu$ group the same procedure is carried out for the regions (0,a) and (a,c) respectively.

3. Remark

This evaluation method has been applied to all 18 spectra. The shape of the spectra A2.1 and B2.3 in the region (a,b) was found to cause a high value for the area under the tail. Here it would have been possible to make some corrections to the areas but we prefered to apply no such corrections.

LABORATORY CODE: 8

TYPE OF SPECTRA: Digital

Short description of evaluation method used:

The evaluation of the alpha spectra data is performed on the basis of an empirical algorithm which allows to calculate the surface of peaks taking into account the overlapping of the tails of adjacent peaks. This is achieved considering the hight of the minimum C_a, C_b, C_c (see fig.) between two peaks and his projections at the beginning (C_{a1}, C_{b1}, C_{c1}) and at the end (C_{a2}, C_{b2}, C_{c3}) of the spectrum.

More in detail, the surface S of the four main peaks of the spectrum represented in the figure, is given by the following relations:

$$S_A = \Sigma C_a + (\Sigma C_e - \Sigma C_{a2}) - \Sigma C_{a1}$$

$$S_B = (\Sigma C_b - \Sigma C_a) + (\Sigma C_e - \Sigma C_{b2}) - (\Sigma C_e - \Sigma C_{a2}) - \Sigma C_{b1} + \Sigma C_{a1}$$

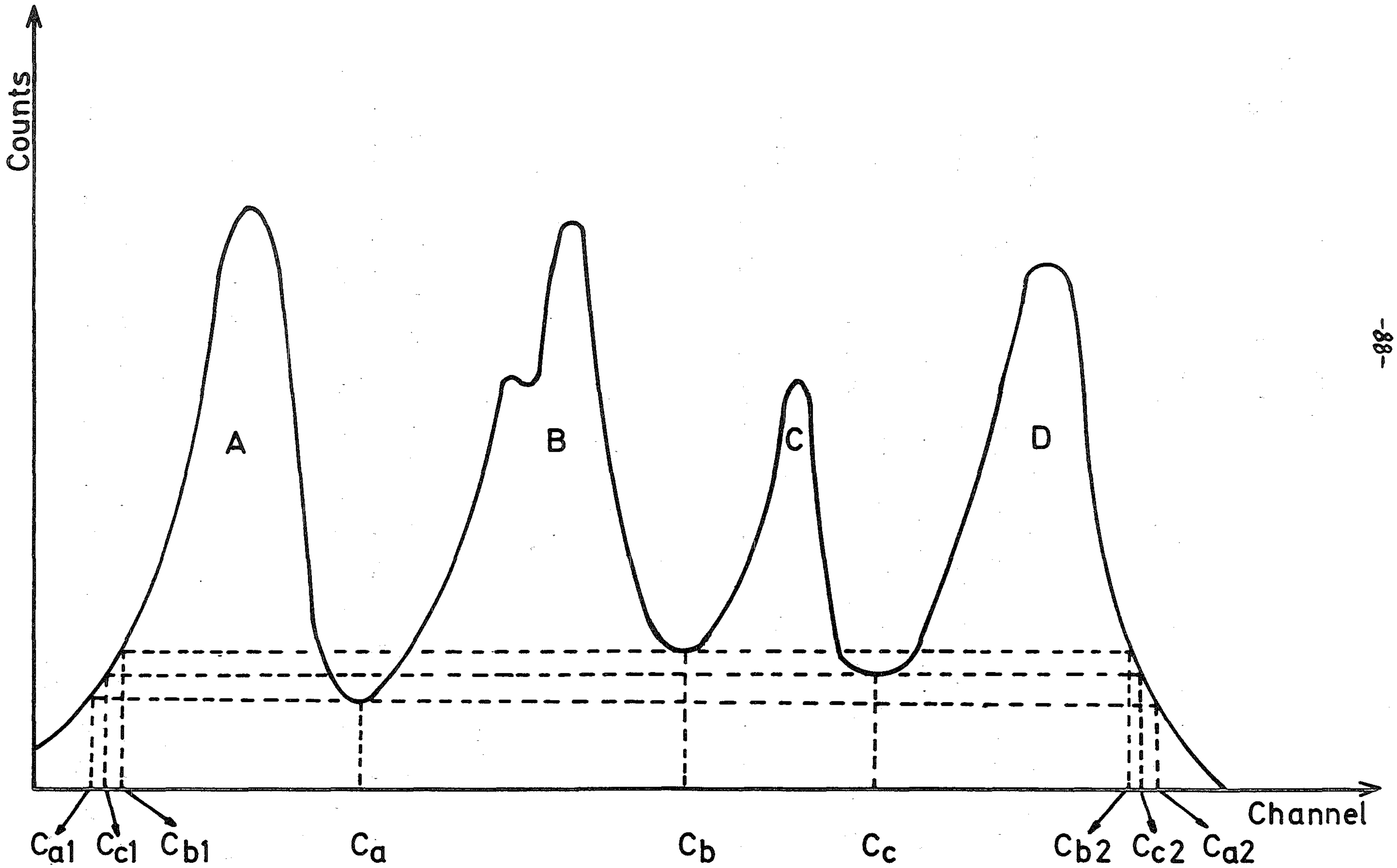
$$S_C = (\Sigma C_c - \Sigma C_b) + (\Sigma C_e - \Sigma C_{c2}) - (\Sigma C_e - \Sigma C_{b2}) - \Sigma C_{c1} + \Sigma C_{b1}$$

$$S_D = (\Sigma C_e - \Sigma C_c) + \Sigma C_{c1} - \quad (\Sigma C_e - \Sigma C_{c2})$$

in which $\Sigma C_a, \Sigma C_{a1}, \Sigma C_{a2}, \ldots, \Sigma C_{c2}$ represent the sum of the content of the channels up to the channel C_a, C_{a1}, C_{a2},....., C_{c2} respectively and C_e is the analogous sum up to the last channel used to analyse the spectrum.

For spectra containing two or three peaks, more simple relations are used.

The precedent relations, after reduction and generalization, are used in a FORTRAN computer program which automaticaly evaluate the surface of any number of peaks.


Counts
A
B
C
D
Channel
C_{a1} C_{c1} C_{b1}
C_a
C_b
C_c
C_{b2} C_{c2} C_{a2}

LABORATORY CODE: 9

TYPE OF SPECTRA: Digital and Paper Tape

Short description of evaluation method used:

120 channels were summed for each peak. Background under the ^{238}Pu peak was estimated graphically.

LABORATORY CODE: 10

TYPE OF SPECTRA: Digital

Short description of evaluation method used:

To determine the alpha peak areas, the sum of the channel contents was performed taking as limits the channels corresponding to the following energies:

Pu 239 + Pu 240	4.960 - 5,210 MeV	(250 KeV energy range)
Pu 238	5,290 - 5.540 MeV	(" " " ")

The background to be subtracted from the considered peak area has been calculated as follows:

- In order to reduce the statistical errors, the activity at the selected limits has been calculated taking in account together with the single channel chosen as limit, also the channels immediately before and after this limit (usually 4 channels before and 4 after). An average of the counts accumulated in each of the nine considered channels has been taken as the value of the activity at the selected limit.

- The variation of the background between the limits was assumed to be linear.

The above mentioned procedure is usually applied in our laboratory to calculate the alpha activity ratios, being the alpha spectra obtained from good quality sources (electrodeposed).

In fact the ratios between the alpha peak areas and not the single peak areas, are the values mainly required by the practical applications.

The agreement between the ratios of the alpha peak areas calculated for B 1 and B 2 sources as well as for C 1 and C 2 sources (see table), indicates our procedure can be used with good accuracy also for sources prepared by evaporation.

However, the disagreement between the alpha peak ratios of the A 1 and A 2 sources (see table) indicates the procedure fails when the background to be subtracted is an important fraction of the total area as in the case of sources with a high content of Pu 238 and/or prepared by evaporation.

In fact the background to be subtracted in the case of the source A 1 is about 50 % of the total area.

Table: Calculated Pu 238/(Pu 239 + Pu 240) Activity Ratios

Counting / Samples	I	II	III	Mean value	Activity at selected limits
A 1	4.262	4.206	4.336	4.268	Calculated from the average of 9 points
A 2	4.012	3.901	4.011	3.974	
B 1	2.155	2.118	2.113	2.129	Calculated from the average of 5 points for B 1 and 9 points for B 2
B 2	2.109	2.100	2.128	2.112	
C 1	0.5570	0.5587	0.5610	0.5589	Calculated from the average of 9 points
C 2	0.5612	0.5613	0.5616	0.5612	

APPENDIX IV:

Definitions of the Statistical Terms
and Other Formulae

1. Alpha Activity Ratios

Basic elements for the statistical considerations are the alpha activity ratios Pu 238/(Pu 239+Pu 240) calculated from the data reported for the Pu 238 and (Pu 239+Pu 240) peak areas / App. II /.

The ratio A_{ijkl} is the result obtained on the l - th repetition spectrum evaluated by laboratory k and taken from target j with sample i.

It is

$i = 1 \ldots r$	with r = 3 corresponding to the three samples "A", "B" and "C"
$j = 1 \ldots s$	with s = 2 corresponding to the two types of targets prepared by evaporation ("1") or electrodeposition ("2")
$k = 1 \ldots t$	with $t \leq 10$ corresponding to the number of laboratories in consideration
$l = 1 \ldots n$	with n = 3 corresponding to the three repetition spectra "I", "II" and "III". As the evaluation of the spectra "A2I" (these are the ratios A_{12k1}) and B2III (ratios A_{22k3}) were not taken into consideration, there is n = 2 in these cases.

2. Laboratory Mean and Associated Terms

The mean value of the activity ratio obtained by laboratory k for sample i, target j, is defined by

$$A_{ijk.} = \frac{1}{n} \sum_{l=1}^{n} A_{ijkl} \qquad (1)$$

Therefore, the standard deviation (SD) of a single spectrum evaluation is given for this laboratory by

$$\sigma_{ijk} = \sqrt{\frac{1}{n-1} \sum_{l=1}^{n} (A_{ijkl} - A_{ijk.})^2} \qquad (2)$$

and the corresponding relative standard deviation (RSD) by

$$s_{ijk} = \frac{\sigma_{ijk}}{A_{ijk.}} \cdot 100 \qquad (3)$$

For some considerations, mean values of these SD's and RSD's were calculated for specific groups G of laboratories. If t is the number of laboratories belonging to the group G, these means are given by

$$\bar{\sigma}_{ij}\ (G) = \sqrt{\frac{1}{nt-t} \sum_{k=1}^{t} \sum_{l=1}^{n} (A_{ijkl} - A_{ijk.})^2} \qquad (4)$$

or

$$\bar{s}_{ij}\ (G) = \frac{\bar{\sigma}_{ij}\ (G)}{A_{ij..}} \cdot 100 \qquad (5)$$

(for $A_{ij..}$ see Par. 3(8).)

The SD and RSD of the laboratory mean value itself are expressed by

$$\sigma(M)_{ijk} = \frac{\sigma_{ijk}}{\sqrt{n}} \qquad (6)$$

and

$$s(M)_{ijk} = \frac{s_{ijk}}{\sqrt{n}} \qquad (7)$$

3. Target Mean and Associated Terms

The mean value of the activity ratio for target j (sample i) is given by

$$A_{ij..} = \frac{1}{t} \sum_{k=1}^{t} A_{ijk.} \qquad (8)$$

as - for each target - the number n of evaluated repetition spectra is always the same for all laboratories in this experiment (orthogonal case[1]), SD and RSD of a single laboratory mean value are given by

$$\sigma_{ij} = \sqrt{\frac{1}{t-1} \sum_{k=1}^{t} (A_{ijk.} - A_{ij..})^2} \qquad (9)$$

and

$$s_{ij} = \frac{\sigma_{ij}}{A_{ij..}} \cdot 100 \qquad (10)$$

The SD and RSD of the target mean value itself are expressed by

$$\sigma(M)_{ij} = \frac{\sigma_{ij}}{\sqrt{t}} \qquad (11)$$

and

$$s(M)_{ij} = \frac{s_{ij}}{\sqrt{t}} \qquad (12)$$

---

1) The more general expression is

$$A_{ij..} = \frac{1}{\sum_{k=1}^{t} n_k} \sum_{k=1}^{t} \sum_{l=1}^{n_k} A_{ijkl}$$

with n_k = number of repetition spectra evaluated by laboratory k.

4. Sample Mean

Calculation of this term is complicated by the fact that in the case of samples A and B the number n of evaluated repetition spectra is different for target 1 and target 2 ($n_1 = 3$; $n_2 = 2$)[1] and, furthermore, that in the case of sample B also the number t of laboratory mean values per target is different[2]. The most simple expression for the mean of sample i in this particular non orthogonal case is given by

$$A_{i...} = \frac{1}{n_1 t_1 + n_2 t_2} \left(n_1 \sum_{k=1}^{t_1} A_{i1k.} + n_2 \sum_{k=1}^{t_2} A_{i2k.} \right) \qquad (13)$$

5. Correlation between Alpha Activity Ratio and Isotopic Composition

If the isotopic composition of the plutonium is known, the alpha activity ratio $\alpha(238/(239 + 240))$ can be calculated by

$$\alpha(238/(239+240)) = \frac{At(238) \times Sp(238)}{At(239) \times Sp(239) + At(240) \times Sp(240)} \qquad (14)$$

At(...)	is the relative abundance and
Sp(...)	the specific activity of the indicated isotope.

---

1) This is caused by the rejection of spectra A2I and B2III, see Section 4.

2) This is caused since laboratory 9 reported for sample B no data on the evaluation of spectra from target 1.

On the other side, the isotopic ratio R(238/239) can be obtained from the alpha activity ratio $\alpha(238/(239+240))$ by

$$R(238/239) = \tau(238) \times \alpha(238/(239+240)) \times (1/\tau(239) + \frac{R(240/239)}{\tau(240)}) \quad (15)$$

$\tau(\ldots)$ is the half life of the indicated isotope,

R(240/239) the isotopic ratio Pu 240/Pu 239.

6. Analysis of Variances

A scheme of the variance analysis is given in Fig. IV-1. It was performed for each group of single evaluation results A_{ijkl} of the alpha activity ratio belonging to the same target and sample; i.e., for all data used in one analysis, the indices i and j remain unchanged.

It is assumed that two error components b_{ijk} and p_{ijkl} contribute to each value A_{ijkl} so that it can be written as

$$A_{ijkl} = \mu_{ij} + b_{ijk} + p_{ijkl} \quad (16)$$

In this equation, μ_{ij} is the true value of the alpha activity ratio of the target j loaded with sample i.

As indicated in the figure, in this specific layout the error component p_{ijkl} consists of two parts c_{ijkl} and e_{ijkl}. As all laboratories obtained identical spectra, the error components c_{ijkl} are independent from k.

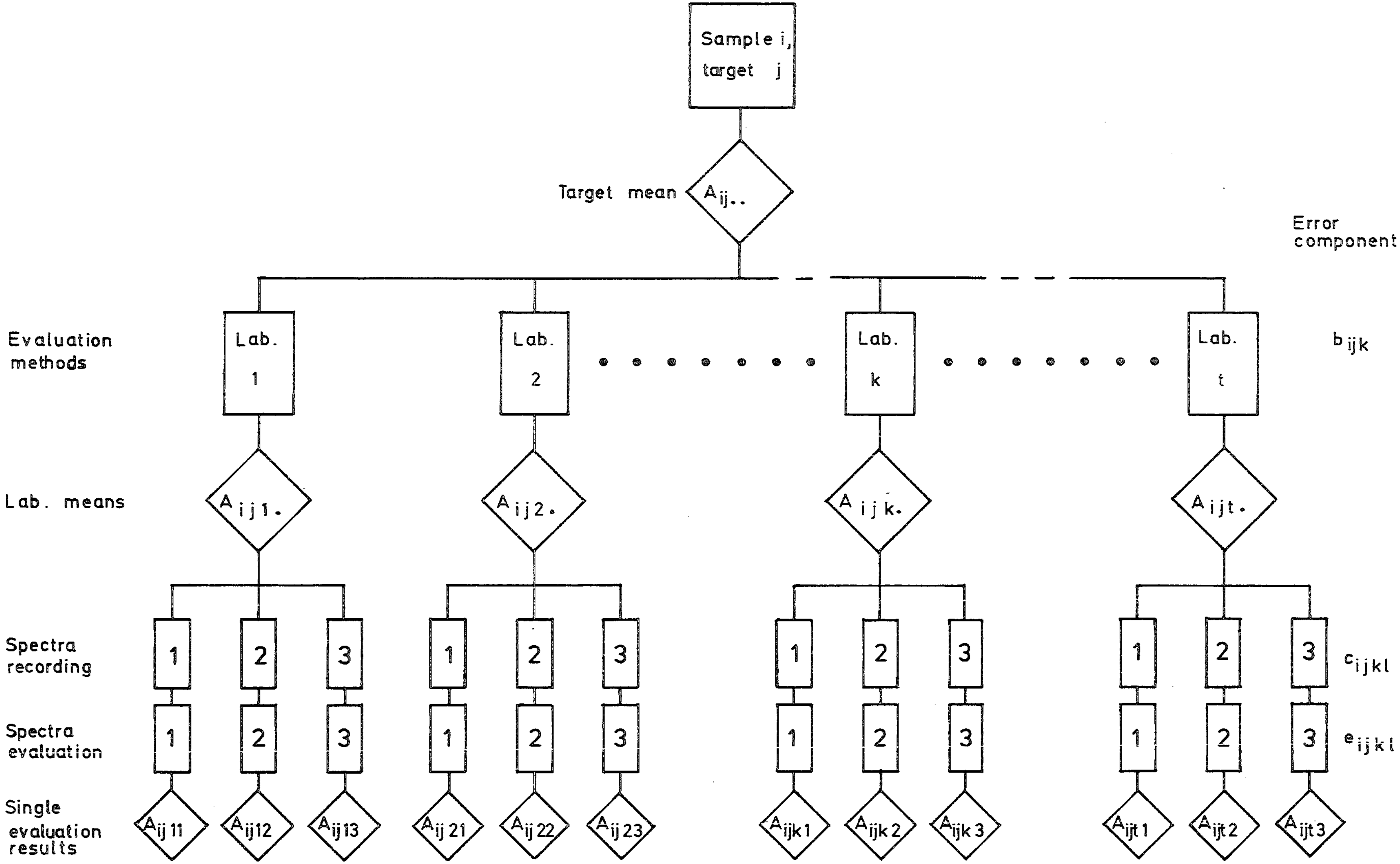


Fig. IV-1: ASET-74: Scheme of the Analysis of Variances

The estimates for the variances $\sigma_{ij}(p)^2 = \sigma_{ij}(c)^2 + \sigma_{ij}(e)^2$ and $\sigma_{ij}(b)^2$ are given by

$$\sigma_{ij}(p)^2 = \sigma_{ij}(c)^2 + \sigma_{ij}(e)^2 = \frac{1}{nt-t} \sum_{k=1}^{t} \sum_{l=1}^{n} (A_{ijkl} - A_{ijk.})^2 \quad (17)$$

and

$$\sigma_{ij}(b)^2 = \frac{1}{t-1} \sum_{k=1}^{t} (A_{ijk.} - A_{ij..})^2 - n\sigma_{ij}(p)^2 \quad (18)$$

From these variances, the RSD's are obtained as

$$s_{ij}(p) = (s_{ij}(c)^2 + s_{ij}(e)^2)^{1/2} = \frac{\sigma_{ij}(p)}{A_{ij..}} \cdot 100 \quad (19)$$

and

$$s_{ij}(b) = \frac{\sigma_{ij}(b)}{A_{ij..}} \cdot 100 \quad (20)$$

Please note that the formulae given are only correct for the orthogonal case as in this specific application. For the more general expressions, reference is made to R. Avenhaus: "Analysis of Variances", pp. 7-130 of KFK 1100, EUR 4576e (1971).