IAEA-TECDOC-1104

Intercomparison of alpha particle spectrometry software packages

INTERNATIONAL ATOMIC ENERGY AGENCY

August 1999

30 - 50

The originating Section of this publication in the IAEA was:

Physics Section
International Atomic Energy Agency
Wagramer Strasse 5
P.O. Box 100
A-1400 Vienna, Austria

INTERCOMPARISON OF ALPHA PARTICLE SPECTROMETRY SOFTWARE PACKAGES
IAEA, VIENNA, 1999
IAEA-TECDOC-1104
ISSN 1011–4289

Printed by the IAEA in Austria
August 1999

FOREWORD

The Advisory Group Meeting on Software for Nuclear Spectroscopy was held in Vienna in 1994, to review the status of software for nuclear spectroscopy and to advise on future activities in the field. One of the recommendations was to conduct intercomparisons of widely available software for nuclear spectrometry.

The first intercomparison was related to gamma ray spectrum analysis software. The methods used, the characteristics of the software and the results were published in IAEA-TECDOC-1011. A companion diskette containing the complete set of test spectra and computer codes used for analysis was attached to that TECDOC.

The second intercomparison, held in Vienna in November 1997, was devoted to alpha-particle spectrometry software. This TECDOC describes the methods used in this intercomparison exercise, characterises the alpha-particle spectrometry software packages investigated and presents the results obtained. No recommendation for a particular program or method for alpha spectrum analysis is given. It is intended that the readers reach their own conclusions and make their own choices, according to their specific needs.

This TECDOC will be useful to anyone involved in alpha-particle spectrum analysis (laboratories, computer centres, quality control offices, etc.). It will be useful to a wide range of persons: university students, technical staff doing alpha spectrometry, software programmers, scientists interested in technical aspects of data analysis in alpha spectrometry, software operators and even executives or project managers who might be involved in setting up a project in this field or involved in the process of purchasing equipment and software for such projects.

This TECDOC includes a companion diskette with the complete set of test spectra and all programs used for intercomparison. The programs and test spectra on this diskette can be used to test any alpha particle analysis software package in the way described in this TECDOC.

The IAEA is grateful to M. Blaauw and E. Garcia Toraño for their help in finalising the manuscript. S. Fazinic of the Division of Physical and Chemical Sciences was the IAEA officer responsible for this publication.

EDITORIAL NOTE

In preparing this publication for press, staff of the IAEA have made up the pages from the original manuscript(s). The views expressed do not necessarily reflect those of the IAEA, the governments of the nominating Member States or the nominating organizations.

Throughout the text names of Member States are retained as they were when the text was compiled.

The use of particular designations of countries or territories does not imply any judgement by the publisher, the IAEA, as to the legal status of such countries or territories, of their authorities and institutions or of the delimitation of their boundaries.

The mention of names of specific companies or products (whether or not indicated as registered) does not imply any intention to infringe proprietary rights, nor should it be construed as an endorsement or recommendation on the part of the IAEA.

CONTENTS

1. INTRODUCTION

Nowadays, with the advent of powerful computers and structured programming languages, the software has reached an important level as the "logical controller" at different levels; from a single instrument to an entire computer-controlled experiment. This is also the case for software packages in nuclear instruments and experiments.

Today, software in nuclear techniques and experiments controls the instrument's operation and performs the data acquisition, storage, validation, and analysis.

In particular, because of the range of applications of alpha-particle spectrometry, software packages in this field are often used. There is an assortment of commercial and open-domain software packages that have been designed for the efficient and correct analysis of alpha spectra. It is the aim of this intercomparison to test and describe the abilities of four such software packages.

In the past, similar intercomparisons have been made for alpha and gamma ray spectrometry and reported in the literature. They all dealt, in one way or the other, with topics like:

- precision of the programs finding peaks (large, small one on low baseline, small ones on the Compton edge, etc.),
- precision in finding and deconvoluting multiplets,
- and many others.

They all relied on different sets of test spectra, which in many cases did not represent real measured spectra and lacked statistical correctness.

This time, the main objectives of the proposed intercomparison were:

- the ability of the programs to determine the peak areas and the peak area uncertainties,
- statistical control and stability of reported results.

The idea is: "it is not so important how successful the program is in finding (or not) a small peak (in different situations), as how is it reported, whether it is within statistical control and it reports results always with a stability". That means that results should be correctly reported in a statistical sense and that a program should operate stably and give stable results.

In this report, the task, methods and results of the intercomparison are presented in order to assist the potential users of such software and to stimulate the development of even better alpha-particle spectrum analysis software. A companion diskette with complete set of test spectra and all programs used for intercomparison is included.

2. THE TEST SPECTRA

2.1. ACQUISITION OF TEST SPECTRA

2.1.1. Introductory remarks

Alpha-particle spectra are critically dependent on the experimental set-up used in the measurements. Source preparation, detector size, and solid angle of the measurement, among other factors, have a strong influence on spectral parameters such as energy resolution and peak tailings. Therefore, selecting "typical" spectra for software testing is not an easy task. The type of programs being tested must be taken into consideration. The number of spectra

must be limited for practical reasons, but the set should at least include some "typical" spectra similar to the ones found in routine laboratory work. On the other hand, a few high resolution spectra must also be included to test the validity of the fitting models used by the different programs. Additional requirements are the existence of groups of peaks in several degrees of overlap and a wide range of peak areas.

In gamma ray spectrometry, doublets with known peak area ratios are difficult to obtain directly because the efficiency of the detector varies with photon energy. In alpha-particle spectrometry, this is not the case and such doublets can be obtained by mixing radionuclides with known activity ratios, or in many cases even by measuring a single radionuclide with known emission probabilities.

The standard equipment in alpha-particle spectrometry includes a semiconductor crystal as radiation detector. Grid ionisation chambers are also in use in many laboratories. However, the fact that ionisation chambers are essentially 2π geometry detectors implies that all particles backscattered at the source will contribute to the spectra, which then become extremely difficult to analyse. Therefore, all test spectra were acquired with Si detectors. These are manufactured in many different sizes, from about 20 mm^2 to more than 2000 mm^2. Small area detectors are used when the best energy resolution is the aim, while the large ones allow a higher counting efficiency to be obtained. From the technology viewpoint, ion-implanted detectors have the best performances, but some laboratories still have surface barrier detectors in operation. The main difference between these two types is the detector window, which for ion-implanted models can be very thin, in the order of 40 nm for the best models. Since ion-implanted detectors are widely used and produce the best spectral quality, they were selected to acquire all test spectra.

2.1.2. Experimental set-up

Spectra were measured in the Radionuclide Metrology laboratories of CIEMAT. Two types of measuring systems were used for the acquisition of the test spectra. The first type was used in most measurements, and includes equipment similar to what is found in almost any laboratory making routine alpha measurements. It features large size detectors and small source-to-detector distances. The high counting efficiencies of these set-ups are essential in the analysis of environmental samples, which usually exhibit very low activities. The second system is a temperature-stabilised chamber with small area detectors, large source-to-detector distances, beam collimators and a magnetic device to reduce the number of coincidences between alpha particles and conversion electrons. This kind of system is used to acquire spectra with the best available energy resolution.

Sixteen spectra, divided into five series, were taken. Four series were acquired with the high efficiency system, the fifth was measured with the high resolution setup. The main characteristics of these spectra, including peak areas and detectors, are summarised in Table I.

In the first system, the equipment included several uncollimated 450 mm^2 ion-implanted Si detectors from CANBERRA, measuring chambers with built-in preamplifiers from OXFORD, model 256, and a multichannel analyser card, model PCA-3 from OXFORD, plugged into a personal computer. A rotatory pump was used to maintain an air pressure of 10^{-5} Pa in the measurement chamber. Several such systems were used to acquire the spectra of the series UR1*, AM1*, RA226* and MPU*, using an analog to digital conversion range of 1k channels. Typical source-to-detector distances were 2.5 cm for a 0.04 efficiency and 0.5 cm for a 0.25 counting efficiency. The energy resolution, measured as the Full Width at Half Maximum (FWHM) of a mono-energetic peak varied between 14 and 20 keV.

TABLE I. MAIN CHARACTERISTICS OF THE TEST SPECTRA. TYPICAL PEAK AREAS REFER TO A ROUGH ESTIMATION OF THE AREAS OF THE MAIN PEAKS IN THE CORRESPONDING SPECTRA

NAME	Counting Efficiency	Typical Peak Areas	ADC conversion gain	keV/ channel	Detector Size (mm^2)	Energy range of interest (keV)	Counting time (s)	Contents
UR1A	0.25	100	1 K	7.8	450	4000–5000	1400	natural
UR1C	0.25	300	1 K	7.8	450		4000	uranium
UR1D	0.25	1500	1 K	7.8	450		20000	
UR1E	0.25	3000	1 K	7.8	450		40000	
AM1M	0.18	2000	1 K	7.8	450	5000–5600	800	americium
AM1N	0.04	13000	1 K	7.8	450		15000	mixture
AM1O	0.04	3000	1 K	7.8	450		3600	
MPU-14	0.05	490	1 K	7.8	450	4500–6000	10000	plutonium
MPU-15	0.05	380	1 K	7.8	450		7700	mixture
MPU-17	0.05	750	1 K	7.8	450		15000	
RA226-4	0.11	2700	1 K	7.8	450	4000 - 8000	5000	^{226}Ra
RA226-5	0.06	1400	1 K	7.8	450		5000	+
RA226-6	0.06	1400	1 K	7.8	450		5000	daughters
AM243-1	0.01	$4.2\ 10^6$	8 K	0.7	50	5000–5400	35722	^{243}Am
AM243-2	0.01	$3.3\ 10^6$	8 K	0.7	50		43958	
AM243-3	0.01	$4.5\ 10^6$	8 K	0.7	50		48421	

The high resolution set-up used to acquire the AM243* series has been described elsewhere [1]. It uses a collimated 50 mm^2 Si detector, a preamplifier model 2001 from CANBERRA kept at a constant temperature (±0.1°C), a spectroscopy amplifier model 244 from TENNELEC, a 8k analog to digital converter model 7421 from SILENA and a memory buffer from the same manufacturer linked by a IEEE interface to a personal computer. The vacuum system included turbomolecular and rotary pumps. The source-to-detector distance was about 8 cm and the solid angle of the measurement was 0.53% of 4π sr. The number of coincidences between conversion electrons and alpha particles was reduced by using a magnetic device. Still, residual structures can be seen in the region close to the main peak in

the spectra. The FWHM was 8.5 keV. This value approaches the limit of the technique at this time.

2.1.3. Source preparation

Two source types were used. The ^{243}Am source, with an activity of about 1 kBq, was prepared by vacuum evaporation onto a quartz disk, the diameter of the active area being 0.9 cm. The remaining sources were prepared by electrodeposition onto stainless steel disks of 2.5 cm diameter and 1 mm thickness. The diameter of the active deposit was 2.2 cm. For the mixed sources, where the relative contribution of the component isotopes had to be known, previously standardized solutions were used for the preparation, and the sources were then measured by high resolution alpha-particle spectrometry to check the nominal values. The main characteristics of the sources are shown in Table II, and the isotopic compositions of the mixed sources are given in Table III.

TABLE II. SOURCE PREPARATION DETAILS. THE COMPOSITION OF THE MIXED SOURCES OF Pu AND Am IS GIVEN IN TABLE III

Nuclide/element	Source activity(Bq)	thickness $\mu g/cm^2$	Backing material	Method of preparation
Natural U	1	6.3	stainless steel	electrodeposition
$^{241}Am + ^{243}Am$	40	10^{-4}	„	„
^{238}Pu, ^{239}Pu, ^{240}Pu, ^{242}Pu	6	10^{-3}	„	„
^{226}Ra + daughters	28	10^{-5}	„	„
^{243}Am	10^3	10^{-3}	quartz	vacuum evaporation

TABLE III. ISOTOPIC COMPOSITION OF THE MIXED SOURCES USED TO MEASURE THE AM1* AND MPU* SERIES OF SPECTRA. UNCERTAINTIES ARE GIVEN IN UNITS OF THE LEAST SIGNIFICANT FIGURE. THE SOURCES WERE PREPARED FROM STANDARD SOLUTIONS, AND THE NOMINAL VALUES CHECKED BY HIGH RESOLUTION SPECTROMETRY

Mixed Am spectra		
Nuclide	Am-241	^{243}Am
Contents (% activity) and absolute uncertainty	55.6 (0.6)	44.4 (0.6)

Mixed Pu spectra			
Nuclide	^{242}Pu	$^{239}Pu + ^{240}Pu$	^{238}Pu
Contents (% activity) and absolute uncertainty	44.1 (0.3)	23.5 (0.3)	32.4 (0.2)

2.1.4. The test spectra

As previously stated, the set includes 16 spectra divided into five series. The filenames, measuring times, typical peak areas and counting efficiencies of these spectra are given in Table I, and some selected plots are included in Appendix A.

The UR1* series is formed by a set of 4 spectra of natural uranium. These spectra present two main groups of lines with similar structures, corresponding to the isotopes 234 and 238 of this nuclide, which are estimated to be in radioactive equilibrium. A small contribution of the isotope 235 shows up in the region between the two main groups. Typical values for the FWHM are 19.5 keV. Target values for this set of spectra are the 234/238 activity ratios.

The RA226* series includes 3 spectra from ^{226}Ra and daughters. They present several isolated peaks which can be used to test the peak search features of the programs and the nuclide identification algorithms. FWHM values varied between 19 and 22 keV, depending on the spectra.

Three spectra of a mixed Am source are included in the AM1* series. ^{243}Am is often used as a tracer in ^{241}Am measurements, and the analysis of these spectra has become routine work in many laboratories. Two groups, one for each isotope, contain several overlapping peaks and can be used to test the deconvolution power of the programs, since the branching ratios of the main component peaks are well known. The main target values are the isotopic composition of the sources which are given in Table III. Typical FWHM values are around 19 keV.

The mixed Pu series MPU* consisting of 3 spectra, corresponds to a source prepared with a mixture of the isotopes 238, 239, 240 and 242. The isotopes 242 (low energy region) and 238 (high energy region) have similar doublet structures. The central group contains 5 component peaks, 3 from ^{239}Pu and 2 from ^{240}Pu. This is also a "typical" spectrum, with the isotope 242 being used as the tracer. The branching ratios of the isotopes 238 and 242 are well known, which again allows for testing of the deconvolution characteristics of the programs. As in the AM1* set of spectra, the reference values for the isotopic contents are given in Table III. For this set, the FWHM of the peaks varied around 14 keV.

The last set, the AM243* series, contains three high resolution, high statistics spectra. They can be used to test the line shape models and the deconvolution performances of all programs. Because of the large number of counts in the spectra (more than 10^6), any difference between the line shape model used by the programs and the true spectral shape would result in a significant residual structure. For the same reason, an incorrect evaluation of the uncertainty in the results by the program would become evident. Target values are the alpha-particle emission probabilities for the 5 major peaks of this nuclide [3]. A small contribution of ^{241}Am, present as an impurity in the solution, can be observed in the spectra, at higher energies than that of the ^{243}Am multiplet.

2.2. DETERMINATION OF REFERENCE DATA

For each test spectrum, a list was prepared with energies, yields and their uncertainties obtained from literature [3, 4, 5]. Mainly, the Table of Isotopes [4] was used. For ^{243}Am, the Nuclear Data Sheets [3] prevailed, and for ^{235}U the 4323.7 keV component was taken from the Table of Radioactive Isotopes [5] since it was missing in [4]. In the case of the mixed sources, the mixing ratios were taken into account. If any component was present in an unknown amount the corresponding peaks were labelled as such and are not to be used in

statistical tests (e.g. the daughters of ^{226}Ra that might have escaped through emanation of ^{222}Rn or not have been in equilibrium).

In many cases, the alpha transitions from a given nuclide feed more than one level in the daughter and the corresponding gamma de-excitations between levels can give rise to conversion electrons which are detected in coincidence with the alpha particles in a fraction which depends on the measurement geometry. This effect modifies the theoretical peak-area ratios which must be taken as targets for analysis, since the original pulse distribution of the alpha particles is deformed by several structures which show up in different parts of the spectrum, depending on the atomic shell from which the coincident conversion electrons originate. The numerical evaluation requires consideration of the measurement geometry, the gamma transitions depopulating or populating the levels, the conversion coefficients and the energies of the alpha particles and electrons involved. Such calculations were performed and the results (uncertainties resulting from uncertainties in literature data included) propagated to the expected peak areas. Second order corrections caused by X ray coincidence summing have been neglected. A detailed description of the problem, including the calculation of corrections for a simple alpha emitter are found in Appendix C.

From the results obtained with one of the tested analysis programs, the area of the most prominent peak in each spectrum was obtained. Using the literature yields of all peaks, values for the expected peak areas of the other peaks in the spectrum were computed. The uncertainty in each peak area was taken to be the combination of the Poisson uncertainty of the expected area and the propagated uncertainties from mixing ratios and yields. Expected peak areas smaller than 1 count were eliminated from the list.

Since in an intercomparison or validation test for alpha-particle spectrometry only relative peak area values need to be tested, the operation described above only served to obtain "reference" uncertainties with practical meaning in the context of a test, and to eliminate peaks from the reference lists that could not possibly be detected in the spectra.

3. THE SOFTWARE CANDIDATES — INFORMATION FROM THE MANUALS

The programs that were tested are AlphaVision 1.20 (EG&G Ortec, USA), Alps 4.21 (Westmeier GmbH, Germany), WinnerAlpha 4.0f5 (Eurisys Mesures, France) and Genie-2000 (Canberra industries inc., USA). All tested programs have been designed to work with standard personal computers and DOS or Windows operating systems. Evaluation copies were obtained from the manufacturers directly. In this section, general information on the programs is provided.

In next sections, some basic questions pertinent to all programs are answered.

3.1. ALPHAVISION

- *Is the manual clearly written and well-organised?*

The manual starts with a step-by-step example, followed with a reference section organised the same way as the Windows menus. There are 20 pages of documentation on the mathematical methods. Also, some file-formats used by the program are described.

- *Does the manual provide a quick overview for experienced users?*

  Not really, but the first chapter offers an example analysis run so the experienced as well as the inexperienced user can get started quickly.

- *What are the principles of operation?*

  The basic facts about the line shape model, including the peak parameters are covered in the manual as well as the mathematical process (Simplex method) used to fit the functions to the data. The algorithms used to smooth the data and to search for peaks are also described in detail. The only point not covered is the calculation of uncertainties. Since the Simplex method does not provide a covariance matrix, there must be some kind of approximation in the estimation of the uncertainties.

  This program can operate in three basic modes. In the first one, **ROIs** can be manually set and only basic integration is allowed.

  The second mode, **peak search and fit**, performs an automatic peak search and fits the peaks, and gives individual information on peak areas and positions. After smoothing, a peak search is performed based on normalised first derivatives. The peak with the highest energy is fitted to a Gaussian with a low-energy exponential tail. According to the manual, the regions selected for the fitting never include more than one peak. The fitting process operates on the smoothed spectrum. After having been fitted, the contribution of the peak is subtracted from the spectrum.

  The last mode, **library search and fit** performs a fit using the information from the peak search as well as the data library and gives results in terms of nuclide activities or areas. In the two modes of operation which fit the spectra, the user can not really interact with the program to modify the results of a previous fit by adding or deleting peaks, unless by editing the library in the corresponding mode.

- *What are the available energy calibration functions?*

  Linear and quadatric, depending on how many datapoints are entered.

- *What kinds of results can the software produce?.*

  Peak positions and areas, and peak energies and radionuclide "DPM"s at a specified point in time (but not Becquerels since the program does not correct for branching ratios).

- *What are the system requirements?*

  - Windows 3.10 or higher,
  - VGA or better,
  - 4 Mb memory required, 8 Mb recommended,
  - approx. 1 Mb disk space.

- *What are the supported spectrum file formats?*

  Only a particular variation of the Ortec *.CHN format.

- *Installation*

  The installation is a two-step procedure: First, an MCA-emulation program named Maestro must be installed. The only thing worth mentioning is that a reboot of the computer was unnecessary to get the program to work on our test system, and no changes in the start-up files were made even though the SETUP program claimed to have modified them. The same was true in the installation of AlphaVision itself.

The installation procedure creates a \user directory on the hard disk without informing the user or asking for an alternative filename.

- *User interface*

  The user interface is a typical Windows interface. However, not many keyboard shortcuts have been defined, and some are ambiguous. The program remembers nicely which spectrum file was treated last.

  The energy calibration can conveniently be performed by pointing out a peak in the graphical display in Maestro, preferably after a peak search has been performed by this program, and entering the energy. The back-and-forth action between Maestro and AlphaVision is confusing because some operations can be performed by either program, but Maestro's intended use is gamma ray spectrometry.

- *Other information*

  The system can be configured for several measuring chains, but can only control ORTEC hardware. It has on line help and provides support for Quality Control and a database to store and retrieve results. These two features were not tested in this evaluation.

  Only some 20 radionuclides are present initially in the libraries supplied with the program. The library can be edited and extended by the user.

  The library is not traceable. Listed are the half-lives (the real ones), peak energies and intensities, all without uncertainties.

3.2. ALPS

- *Is the manual clearly written and well-organised?*

  The manual starts with step-by-step examples, followed with an alphabetical codeword reference guide, followed by 25 pages of methods & algorithms, including an introduction to alpha-spectrometry principles. It is well-organised and clearly written, but could be improved perhaps by treating the codewords according to category rather then alphabetically. Also, the use of FORTRAN variable names (e.g. "istart" instead of "start") and expressions (e.g. "10**lim" instead of "10^{lim}") in the manual should probably be avoided to improve the clarity.

  The manual does not have an index, with the result that, every now and then, the user will have to leaf through the manual until the wanted item is found. Also, the codewords are not all intuitive, but a quick reference table is provided at the beginning of that section.

- *Does the manual provide a quick overview for experienced users?*

  No, but it starts with two example analysis runs so the experienced as well as the inexperienced user can get started quickly.

- *What are the principles of operation?*

  The spectrum is divided in regions that may contain peaks and can be treated independently. For each region, a peak search is performed based on normalised first derivatives. If peaks are found, an exponential background is applied to the region such that it's always lower than the experimental channel contents. Then, the background is subtracted. The program decides if the peaks present in the region should be treated as a multiplet or not. A Gaussian with one or two folded exponential tails is fitted to the

channel contents with a linear least squares algorithm in three passes. In the first pass, the FWHM is not varied. Insignificant peaks are then eliminated. A residual analysis is performed and if necessary, peaks are inserted. The fitting is repeated, allowing all parameters to be varied. Again, insignificant peaks are eliminated. A final fit is then performed. After the final fit, the energy calibration is used to identify the peaks.

The manual does not define the uncertainties reported by the program, but states that reported uncertainties are "greater than the usual 1-STD uncertainty". The analysis results, as compared to the other programs, show that they are indeed by a factor of 3.4.

- *What are the available energy calibration functions?*

  Linear and quadratic. Data entry may be by energy/channel pairs or be library or display driven.

- *What kinds of results can the software produce?*

  Radionuclide activities at a specified point in time.

- *What are the system requirements?*
  - DOS 2.00 or higher (a Math Coprocessor is recommended if not standard),
  - EGA,VGA or better,
  - 512 kB memory,
  - 500 kB disk space,
  - Hardware key in parallel port.
- *What are the supported spectrum file formats?*

  16 formats, including all the important ones:
  - TARGET/OXFORD,
  - CMTE (4 bytes binary without header),
  - Format free FORTRAN integers, no header,
  - EG&G (.chn and.spc),
  - CMTE (with header),
  - CANBERRA Spectran-F,
  - SILENA, VARRO and EMCAPLUS,
  - INTERTECHNIQUE,
  - NUCLEAR DATA (Accuspec and μMCA),
  - ASCII file (no header, measurement time in channel 1),
  - CANBERRA S-100,
  - CANBERRA S-80 and S-90,
  - USER1 (192 byte header, 3 bytes per channel),
  - ATMOS,
  - TMCA,
  - APTEC v4.3 or v6.3.

- *User interface*

  The program has two user interfaces: commands (called codewords) can be entered from the keyboard or selected menu-driven. A powerful and easy-to-use macro possibility is also present, that can be used to operate the program in batch mode as well.

  The menu-driven user interface is not very user-friendly: The code words are not grouped as in Windows applications, but displayed in alphabetical order, all at the same time. Codewords can then be selected with the cursor keys. In menu-driven mode, program feedback after execution of commands is displayed for a *very* short time before the screen is cleared and the codewords are displayed again. The main menu screen does not show whether a spectrum has been loaded and if so, which.

  The energy calibration can conveniently be performed by pointing out a peak in the graphical display and entering the energy.

  The user interface to control the graphical display is confusing: The X-axis of the display can be expanded or compressed, which will usually lead to display of a part of the spectrum where the cursor isn't. The "centre marker" option should compensate for that, but unfortunately it expands the X-axis to 256 channels in the process. By the time one has the cursor in view and the X-axis expanded as one wishes, one may have lost track of where one is in the spectrum.

  The results of an analysis run can be sent to file. However, the user has no control over the output filename: Characters 7 and 8 of the filename are changed to "AL" by the program. This limits the freedom of the user in selecting a spectrum naming scheme.

  Alternatively, the user can redirect all program output to a file that he *can* name as he likes. However, this output may not consist of peaks sorted on energy, depending on how the analysis run was completed. This, however, is the method used in the examples in the manual. After each analysis, the output must be redirected to some junk file to prevent all results from ending up in the same file. When using a printer, this is no problem. The interactive fitting routine refuses to send output to the screen. Either a printer must be attached or an output file created, otherwise the program will hang.

- *Installation*

  The installation procedure consists of copying files to a directory on the hard disk. No problems were encountered, except that ALPS will only run if the working or current directory is set to the directory where the program is, rather than to the directory where the spectra are.

- *Other information*

  The program is limited to spectra with a maximum of 4096 channels.

3.3. GENIE-2000

- *Is the manual clearly written and well-organised?*

  The manual comes in three volumes — Operations (containing basic spectroscopy), Options (interactive peak fitting and alpha spectrum analysis) and Customisation Tools. It is the two manuals contained in the Options volume which are of primary interest. Interactive peak fitting is common to both alpha and gamma spectrometry, whilst alpha spectrometry analysis is as the name suggests. One drawback is that both manuals assume a knowledge of the basic operation of the Genie 2000 system for gamma analysis, hence one

must also be conversant with the details contained in the basic spectroscopy manual. Invariably one finds oneself having to jump between manuals to fully clarify a point.

Once the software is loaded, full online help is provided.

- *Does the manual provide a quick overview for experienced users?*

  Such an overview is not provided. However, one could envisage an experienced user simply installing the program and using the online help as and when required.

- *What are the principles of operation?*

  Extensive mathematical details of the analysis algorithms are given in the Customisation volume. However the text relates to both alpha and gamma analysis and as such one must extract the descriptions relevant to the alpha analysis.

  In general, peak search is performed using either a library driven routine (for spectra of known contents) or by the Generalised Second Differential Method (for spectra of unknown contents). The latter was used during the current testing. Details of energy, efficiency and FWHM calibration procedures and corrections are given. Peak areas are determined either by summation (singlets) or by non-linear least squares fitting (singlets and multiplets) using a modified Marquardt algorithm for fast convergence. Residual search is available, as are MDA routines. The method of uncertainty propagation is fully documented.

- *What are the available energy calibration functions?*

  The standard energy calibration function is linear, but quadratic and cubic terms may be specified during an interactive calibration session. The energy calibration can be done in several modes, by loading previously saved files, by nuclides taken from the library or by direct entry.

- *What kinds of results can the software produce?*

  The software can perform peak searching with an adjustable significance factor. It can calculate peak areas and uncertainties for isolated and overlapped peaks and report radionuclide activities. Results can be obtained relative to a tracer. It also has a nuclide library of alpha emitters facilitating nuclide identification.

- *What are the system requirements?*

  - Windows '95 (minimum 386SX, 8MB RAM),

    or

  - Windows NT v4.0 (minimum 486, 16MB RAM),
  - VGA (640 x 480 colour) display,
  - 100MB free disk space,
  - TCP/IP networking,
  - Hardware lock on parallel port.

- *What are the supported spectrum file formats?*

  From the File menu, only CANBERRA files. A batch command, FILECNVT, is available in MS-DOS (though it was difficult to find in the manual). This will convert the following files into CAM format: S100, System AT, Gamma-AT, Series 35 Plus Toolkit, Sampo90, Ortec, ND 6* series, Nucleus, Intertechnique.

- *User interface*

  The user interface is user-friendly with easy to access pull-down menus. Comprehensive online help is available. Information as to which spectrum is loaded is clear; this is essential as more than one MCA window may be open at any one time.

  The MCA spectrum display is non optimum, it often being hard to discern individual features. This improves with expansion of the displayed area, but the split screen format in this mode limits the vertical size obtainable. The IGF display is clearer and may be expanded to full page size. The only problem with this display is that the cursors marking the fit region are often almost invisible to the eye. It is necessary to "hunt" for them using the mouse pointer, which changes format when on these cursors.

- *Installation*

  The Basic Spectrometry Package (BSP) is contained on the first four of five main diskettes provided. The fifth contains drivers for AIM units. This labelling is confusing as the "wizard" (automatic loading) quits after diskette "4 of 5", giving one the feeling that installation has not been successfully achieved. In fact, diskette 5 is independently loaded, as one optional component as are the Alpha Spectroscopy Package (ASP), and the Interactive Peak Fitting (IPF).

  Loaded software: BSP, ASP, IPF.

  Not loaded: QA, AIM drivers.

  Genie 2000 software is a client/server architecture and communications between the client and server components is through Windows sockets using the TCP/IP protocol. Thus TCP/IP must be installed as a network component even when the computer is not part of a network. Note that as ASP and IPF will not install unless this has been done.

- *Other information*

  For isolated peaks, the user can choose to integrate or fit the peaks. This is an arbitrary choice for the user. In these analyses, one user chose to integrate the peaks during automated (default) peak searches and to fit the peaks during interactive analysis; the other elected to fit the peaks for all peak area determinations. For close peaks, the user can choose to fit them as single lines or multiplets by changing a parameter expressed in terms of FWHM.

  A basic nuclide library is provided with the program, though there is no reference as to the source of the data. For each nuclide present, the main alpha emissions are listed with the most intense, the "keyline", flagged. Half lives, energies and emission probabilities are all given with uncertainties; a refreshing change. It should be noted that for short-lived nuclides in decay chains e.g. ^{226}Ra decay, the half-life is given as that of the longer lived parent nuclide. Facilities for library maintenance, library creation as so forth are provided.

3.4. WINNER ALPHA

- *Is the manual clearly written and well-organised?*

  The manual is clearly written and the basics stages of an analysis given. However, it can be very confusing. Before using the alpha analysis one must be conversant with the gamma analysis. The manual appears to be under development with sections or variable descriptions missing.

- *Does the manual provide a quick overview for experienced users?*

  Not at present.

- *What are the principles of operation?*

  There is no description of the internal workings of the program, the line shape model or the fitting methods. However it is evident that it is planned to include them at some point in time as the headings are present, just no details.

- *What are the available energy calibration functions?*

  Quadratic, obviously can also be used as linear.

- *What kinds of results can the software produce?*

  Standard output is peak position, peak area, nuclide activity and identification. The program can be used in tracer analysis mode.

- *What are the system requirements?*

  - 486 PC with 4MB RAM,
  - 4MB Hard Disk free space,
  - Windows 3.x, 95 or Windows NT,
  - A hardware key is required on the parallel port.

- *What are the supported spectrum file formats?*

  Spectra from SILENA, CANBERRA and ORTEC may be imported.

- *User interface*

  The program has a typical Windows interface. Nevertheless, some operations as the initial energy calibration can not be done by using a cursor to mark a peak position on a graphical display of the spectrum.

  One nice feature of this program is that it produces good plots, including the raw data, the fittings and the residuals. That allows a good evaluation of the quality of the analysis and makes easier the corrective actions to re-analyse the spectra.

  The program has no on line help.

  Binary and ASCII reports can be tailored by the users to suit their own requirements.

- *Installation*

  No problems were encountered in the installation. The nuclide library LARA was copied from an additional disk.

- *Other information*

  The system can be configured for several measuring chains and drivers are available for Eurisys, Silena, Oxford, Ortec and Canberra MCAs. It can support Quality Control, although this feature was not tested. The nuclide library LARA was used, after some minor modifications. The version that came with the program included 38 isotopes and 104 lines.

  Gamma analysis is the standard mode of operation. Switching to the alpha analysis mode required the use of a macro facility not covered in the basic manual, and further assistance from the manufacturer was needed to accomplish this feat.

4. INTERCOMPARISON METHOD

4.1. ANALYSIS OF SPECTRA

All test spectra were analysed with each program by two users. In total three different users, named S, E and M exploited analysis programs. Prior to the actual analysis of spectra, the manuals of the programs were studied for two hours.

TABLE IV. USER AND PROGRAM SPECIFIC REMARKS ON HOW THE ANALYSIS WERE PERFORMED

	AlphaVision	Alps	Genie 2000	Winner Alpha
user 1 "def"	no remark User E	no remark User M	integrate, continuum User S	No "automatic" mode available User S
user 1 "opt"	Realistic nuclide library employed.	Sensitivity set to maximum. Manual peak insertion employed.	Manual insertion of peaks employed.	Manual insertion of peaks employed.
user 2 "def"	Energy calibration problem compensated. User M	no remarks User S	fit, no continuum User E	No "automatic" mode available User E
user 2 "opt"	Energy calibration problem compensated. Library with artificial radionuclides employed.	Sensitivity set to maximum. Low FWHM estimate employed. Manual peak insertion employed.	Manual insertion of peaks employed.	Manual insertion of peaks employed.

Of the four programs tested, only Alps and Genie were designed to allow for easy analysis of spectra not acquired with software and hardware obtained from the same vendor.

As a result of this, in the case of AlphaVision, the menu options related to energy and efficiency calibration were not accessible. The energy calibration was performed with the related program "Maestro" and written to the spectrum file. The two programs number the channels differently, which was compensated for by offsetting the correct energies in Maestro with the slope of the calibration curve. For Winner Alpha, a special conversion of files had to be performed to label them internally as alpha-particle spectra.

For each set of spectra an internal energy calibration and, if possible, a FWHM and shape calibration was performed. Then the spectra were analyzed twice. First with all parameters, such as peak-search sensitivity and residual-search sensitivity, set to the default values or, if available, to the values suggested in the program manual. Second, with the parameters set to the user's liking, attempting to optimize for the analysis of the spectrum in each set with the best statistics. The optimization was performed using the information offered by the analysis program and its documentation, not the knowledge of the actual contents of the spectra. The optimized analysis also encompassed, if possible, manual insertion of peaks or, in the case of AlphaVision, the use of nuclide libraries tailored to the spectrum to be analyzed. This involved insertion of non-existing radionuclides in the library since only 4 peaks per radionuclide can be entered.

In addition, the libraries shipped with the programs were compared with respect to ^{226}Ra and progeny.

In Appendix D, user- and spectrum specific notes on the analysis as performed with each program are given. In Table IV program-related difficulties and remarks are given.

All results obtained, as well as the user-specific remarks in Appendix D, are labelled with the first initial of the user, i.e. "E", "M" or "S".

4.1.1. AlphaVision

In the intercomparison, spectra were to be analysed that were not acquired with an Ortec system. Even after converting to the Ortec *.chn format, some header information was still missing which made it impossible to use the program as described in the manual. For example, it was not possible to make an energy calibration in AlphaVision because the detector was not defined. In Maestro an energy calibration could be made, but apparently the two programs number the channels differently: The energies entered in Maestro had to be off by the slope of the calibration curve in order to obtain correct energies in AlphaVision.

The sensitivity of the peak search algorithm can not be modified. Therefore, if the algorithm fails, it is impossible to make a good fit, since the results of the peak search can not be edited or modified by the user.

When working in the library mode all lines of the selected nuclides are fitted, but only general information for each nuclide is provided by the program. Again, the results of the fits can not be edited or modified to start a new fit, unless the library is edited.

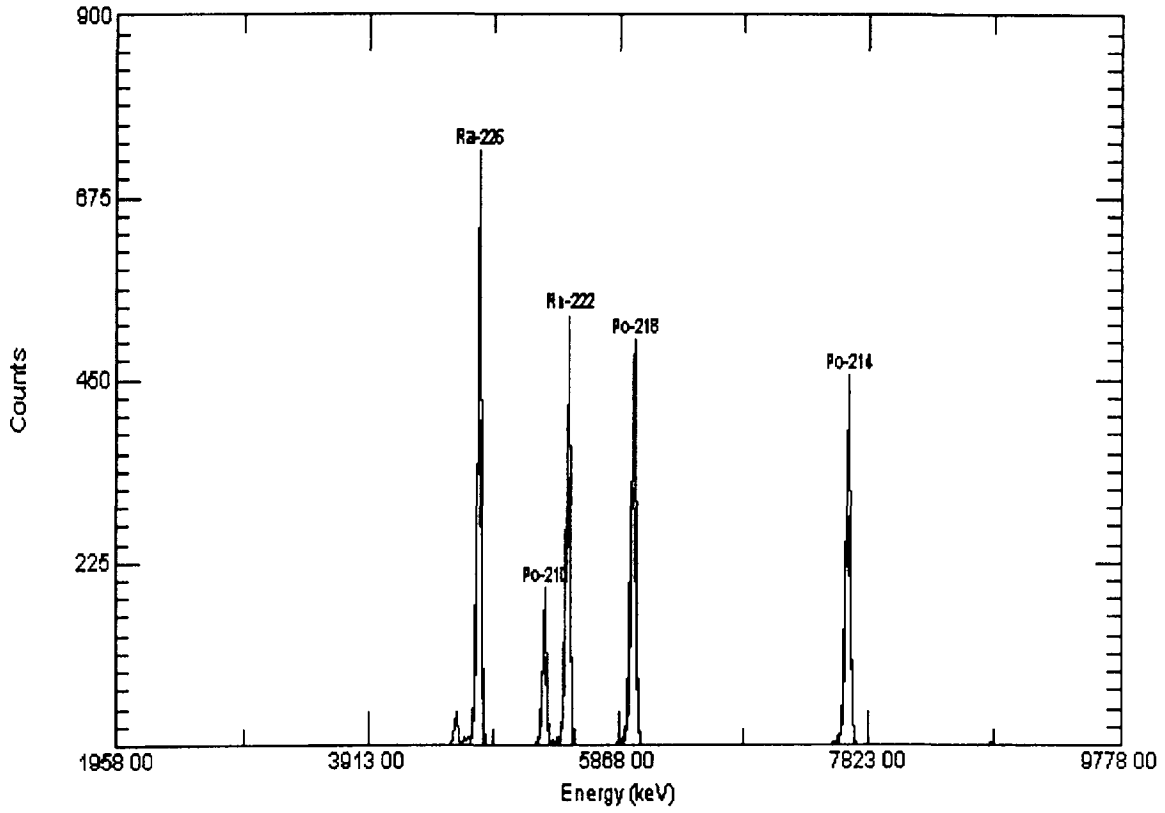

FIG 1 Alpha Vision plot of results for RA2624 spectrum

To define sample type-specific libraries, the user must point out nuclides present in a "master library" This is a rather cumbersome procedure, since the contents of the master library are presented unordered (actually, ordered with respect to the energy of the first peak of each nuclide, but that is not much help) - as the master library grows, it takes longer to spot the nuclide one wants to point out

In the test process, an artificial nuclide named "Am-241m2" was inserted as the 62nd nuclide, after which the library data structure apparently was damaged because the nuclide added could no longer be deleted from the library Similar names had been inserted before without problem

In Fig 1, a plot is shown as produced by AlphaVision

4.1.2. Alps

To perform a FWHM calibration, the user will have to analyse a spectrum first using a rough estimate of the average FWHM in the spectrum, copy the FWHM values in terms of channels obtained in the fits down on paper and then enter them in the FWHM calibration data table An energy calibration should not be performed prior to this, because, once an energy calibration is present, all FWHMs will be displayed in terms of keVs rather than

channels. Needless to say, this procedure is cumbersome and error-prone. Since FWHMs are varied in the fit in typical alpha-spectrometry problems, it was decided not to use FWHM calibration curves at all.

The program has an option to merge channels when reading a spectrum. This option was tested with the ^{226}Ra spectrum, adding channels in pairs. The sensitivity was kept at the default value of 35, but the FWHM estimate was adjusted, i.e. divided by 2. With the merged channels, the program detected the 5% ^{226}Ra peak at 4601.9 keV, with the original channel contents it did not. This means that the search algorithm is flawed: Its sensitivity decreases as amplifier gain increases.

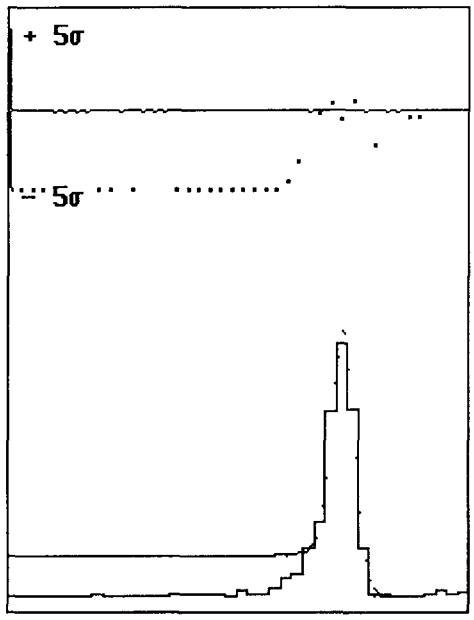

FIG. 2. Alps: Failed fit.

The fitting algorithm of the program is unstable with respect to FWHM: The failed fit in Fig. 2 was obtained with a FWHM estimate that was very close to the actual FWHM of the peak: 2.2 vs. 2.29 channels. However, with most settings fits as shown in Fig. 3 were obtained, indicating that the peak shape model is good.

If a spectrum was analysed more then once in the optimisation process, each peak was reported in the peak identification table as often as the spectrum was analysed. This means that, to clear the memory of the program, the spectrum must be read again.

The peak assignment processes are very simple; all possible assignments according to a user-specified match criterion in terms of keV are listed if the "energy oriented activity calculation" is performed. The alternative is a "nuclide oriented activity calculation" where the presence of peaks is checked for each nuclide in the library.

Both options accept a match criterion prompted for with "width of window in keV*10", but the energy-oriented routine interprets 1.5 as a window width of 15 keV, whereas the nuclide-oriented routine interprets the same number as a width of 0.15 keV.

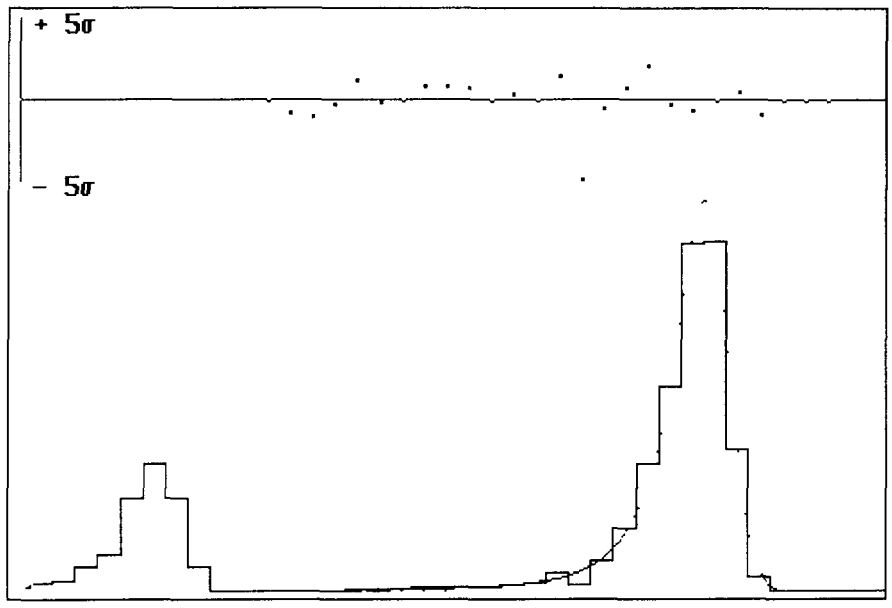

FIG.3. Alps: Good fit.

4.1.3. Genie-2000

Analysis is performed as a sequence of steps, each of which may be selected from the analysis window. A sequence may be stored and automatically run at a later date. Thus one only has to decide on a preferred methodology and load it once.

Results are not automatically written directly to disc; instead reports may be requested at each step in the analysis and are written to a file named spectrum_name.rpt. This safety feature prevents accidentally overwriting good fitting data with bad fitting data, but conversely, one may forget to save good fitting data. This is where the ability to construct an automatic sequence is useful.

For each spectrum, both users adopted the following procedure to obtain the results:

(1) Calibrate for energy,

(2) Peak locate,

(3) Peak area determination (with optional nuclide identification),

(4) Report to "default" result file,

(5) Interactive peak fit - change fit region, add/delete peaks (with optional nuclide identification),

(6) Report to "optimised" result file.

The users' preference for fitting a function to a continuum under the peaks being analysed differed slightly but converged on intent. User E fitted all spectra with no continuum,

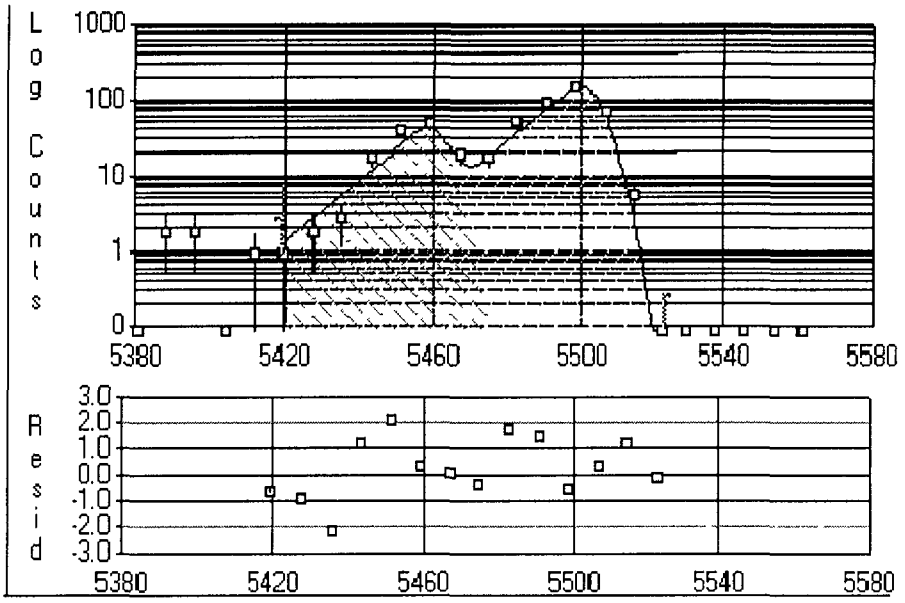

FIG.4. Genie 2000 interactive peak fit display.

as is correct. User S used it as a guide when adjusting the fit region, requiring that either a step or linear function be vanishingly small.

4.1.4. Winner Alpha

Although it was previously decided to analyse all spectra in automatic and optimised modes, the lack of default parameters meant that in all cases Eurisys needed intensive interaction with the user. This was adjudged to be equivalent to optimising the fit and hence, only the second mode was used in this intercomparison.

The methodology to analyse the spectra is not covered precisely in the manual.

After several trials (and errors) the method of analysis was established as follows:

- Import the file
- Use a MACRO program to change to the ALPHA mode.
- Provide all parameters required for the program to start. This is a difficult task since some of them are not well described in the manual. The basic parameters are:
  - (a) Peak search sensitivity.
  - (b) Tail parameters. 0.5 and 0.9 were found to be good initial values for all analysed spectra.
  - (c) Preliminary energy calibration is also required.
- One of the two possible modes of analysis, fitted areas or integration, has to be chosen.
- Perform an automatic peak search.
- If peak search is not satisfactory, manually mark the estimated regions as ROIs.

- Perform a full energy calibration. This is done by the **Edit Calibration** item in the menu: The program starts a preliminary peak fitting and presents a table of peak positions and their energies estimated according to the initial calibration provided by the user. This can be modified to improve the calibration function.
- Determine the best parameters for the peak model. Using the *edit peak parameters* function, one peak is selected and taken as a model to which fit the line shape and obtain the optimum peak shape parameters: FWHM and two tailing parameters.
- Analyse the spectra and plot results and residuals.

If necessary, ROIs were edited to facilitate the insertion or deletion of peaks before re-analysing the spectrum.

In Fig. 5, a typical display of the program is shown.

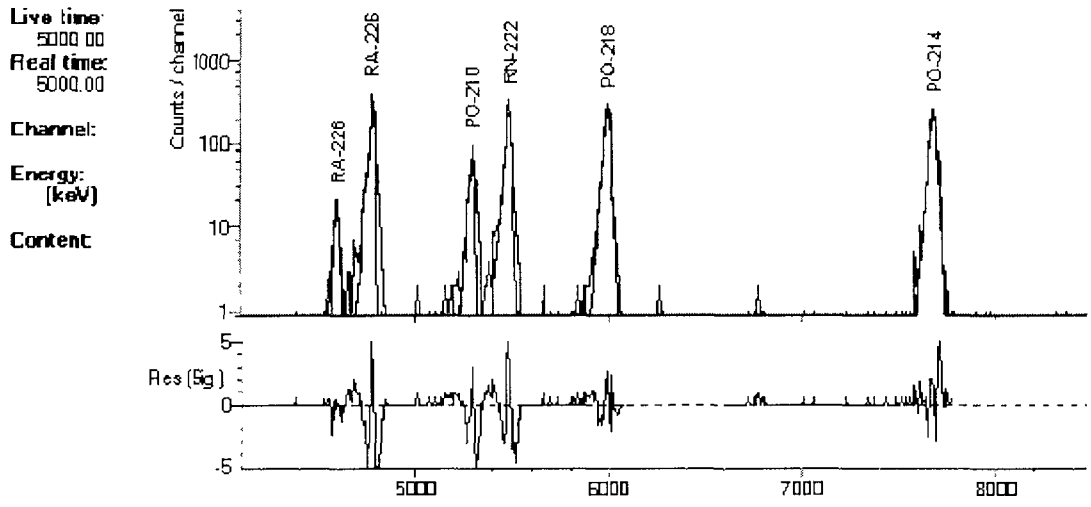

FIG. 5. Winner Alpha display.

4.2. DATA HANDLING

4.2.1. Direct doublet ratios in the natural uranium spectra

From the analysis results of the natural uranium spectra, the ratios and associated uncertainties of the peak areas, if reported for both components of the doublets of the ^{234}U and ^{238}U radionuclides, were computed and compared to the yield ratios found in the Table of Isotopes [4]. The doublet separations are approximately 2.5 FWHM in both cases, and the components are clearly visible to the eye in the spectra with the best statistics.

4.2.2. Systematic statistical testing of the analysis results

4.2.2.1. Preliminary steps

A conversion program was written that yielded output in a standard format containing peak energies and areas, both with their absolute 1 standard deviation uncertainties. This step was not entirely trivial, because only two programs did report uncertainties in the peak positions, i.e. Alps and Genie. For the other programs, which reported energies with two digits after the decimal point, implying uncertainties of less than 0.01 keV, these uncertainties were set to 0.01 keV. Also, not all programs reported 1 standard deviation uncertainties. The reported uncertainties were converted to one standard deviation absolute uncertainties according to the definitions given in the documentation of the programs. In the case of Alps, the reported uncertainties were divided by a factor of 3.4 to achieve this.

4.2.2.2. Statistical comparison

A separate program was written to perform a statistical comparison based on standardized residuals or z-scores, i.e. the differences between reported values and reference values divided by their own uncertainties.

Using all cases where both a reported area and a reference area were available ("hits"), a weighted average and its uncertainty of the ratios of reference peak areas and program output peak areas were determined. The weights used were the inversed squares of the uncertainties in the area ratios, computed from reference and reported uncertainty. The computation was performed in two passes: In the first pass, the average ratio was estimated. Peaks with reference uncertainties exceeding 11% were excluded in this pass. In the second pass, outliers at the 95% confidence level were excluded from the computation. The reported peak areas and associated uncertainties were multiplied with the average ratio before proceeding.

In the case of a "hit", two z-scores could be computed: A z-score related to the quality of area determination based on the uncertainties in the reference files

$$z_{ref} = \frac{A_{rep} - A_{ref}}{\sqrt{2\sigma_{ref}^2}}, \qquad (1)$$

and a z-score related to the statistical control of the analysis program based on both the reference uncertainty and the uncertainty reported by the analysis program

$$z_{rep} = \frac{A_{rep} - A_{ref}}{\sqrt{\sigma_{rep}^2 + \sigma_{ref}^2}}, \qquad (2)$$

where A_{ref} and A_{rep} are the reference and reported peak area, and σ_{ref} and σ_{rep} their uncertainties, respectively. Such z-scores are expected to be normally distributed with a zero mean and a unity standard deviation, i.e. z-scores higher than 2 or lower than -2 indicate that something is wrong at the 95% confidence level.

If the reference area was missing, the reported peak was considered to be a "false hit" and only the second z-score could be computed, using zero both as the reference area and as its uncertainty. If the reported area was missing, it was considered a "miss" and only the first z-score could be computed. Missing a noisy peak or reporting a false hit with a high uncertainty in the area do not result in high z-scores and are therefore "allowed" in this test.

Ratio to multiply all measured peak areas with: 1.0839 +/- 1.483 %

'TRUE' DATA AM1N.REF				MEASURED DATA AM1N.OPC				Z scores	
E		A		E		A			
val	unc	val	unc	val	unc	val	unc	rep	ref
5181.0	0.1	120.3	11.0	5190.5	4.9	128.8	18.9	0.4	0.5
5233.5	0.1	1159.7	34.1	5243.8	1.7	987.6	49.1	2.9	-3.6
5275.4	0.1	9616.5	98.1	5284.5	0.5	9433.5	180.4	0.9	-1.3
5322.0	0.1	15.2	3.9	5331.2	7.0	87.0	16.3	4.3	13.2
5350.0	0.1	17.5	4.2	5350.0	0.1	0.0	4.6		2.9
5388.0	0.1	226.1	15.0	5388.0	0.1	0.0	16.3		-10.7
5417.0	0.1	1.4	1.2	5417.0	0.1	0.0	1.3		-0.8
5442.9	0.1	1794.7	42.4	5449.3	0.8	1588.4	41.1	-3.5	-3.4
5469.0	0.1	5.5	2.3	5469.0	0.1	0.0	2.5		1.7
5485.6	0.1	11508.0	107.3	5490.4	0.4	11685.0	213.1	0.7	1.2
5512.0	0.1	31.5	5.6	5526.1	5.3	302.3	42.3	6.4	34.2
5544.3	0.1	49.3	7.0	5544.3	0.1	0.0	7.6		5.0

FIG. 6. Table of statistical results, showing a hit at 5275 4 keV and a miss at 5388 keV.

Some of the test spectra contained doublets with small separations It was decided to allow the analysis programs to determine the total area of such doublets. To this end, if two peaks in the reference list matched one peak in the analysis program output, i e. if both reference peaks were located within 1 × FWHM of the analysis result, the two reference peaks were merged before the computation of z-scores. This was also done if such two reference peaks were located within the position uncertainty reported by the analysis program.

A section from the comparison program output is shown in Fig 6.

From the z-scores, reduced sums of squares χ_r^2 were computed for different categories of peaks

- Large peaks: Hits for which the ratio of reference peak area and reference peak uncertainty is larger than 10.
- Peaks with high reference uncertainty: Peaks where the reference uncertainty is significantly due to uncertainty in literature yield data
- Small peaks Hits for which the ratio of peak area and uncertainty was less than 10
- Any hit. All peaks belonging to the previous three categories
- Misses.
- False hits
- Total: All previous categories.

For the three "hit" categories, two χ_r^2 values were computed: one based on z_{rep} values denoted X1, and one based on z_{ref} values denoted X2.

5. RESULTS

The basic statistical results are given in Appendix B. In Table V, an overview is presented of parts of the radionuclide libraries supplied with the different programs. To facilitate comparison, only data on the alpha emitters in the ^{226}Ra decay chain are shown (For AlphaVision, a more extended library is available from EG&G Ortec). In Fig. 7 and Fig. 8, the results for the natural uranium doublet ratios are presented.

TABLE V. COMPARISON OF NUCLIDE LIBRARIES AS SUPPLIED WITH THE PROGRAMS

	Alps		**Genie 2000**		**Winner Alpha**		**AlphaVision**	
^{226}RA	*1600 Y*		*1600 Y*		*1600 Y*			
	4784.5	9.445E+01	4784.2 (0.6)	94.45 (0.05)	4784.400	94.450 (0.05)		
	4601.9	5.550E+00	4601.4 (0.6)	5.55 (0.05)	4601.700	5.550 (0.90)		
	4340.0	6.500E-03						
	4191.0	1.000E-03						
	4160.0	2.700E-04						
RN-222	*3.823 D*		*1600 Y*		*1000 Y*			
	5489.5	9.992E+01	5489.7 (0.3)	99.92 (0.01)	5489.200	99.920 (0.01)		
	4987.0	7.800E-02						
	4827.0	5.000E-04						
PO-218	*3.110 M*		*1600 Y*		*1000 Y*		*0.0021 Y*	
	6002.4	9.996E+01	6002.55 (0.09)	99.98 (0.002)	6002.400	99.979 (0.00)	6000.00	100
	5181.0	1.100E-03						
PO-214	*1.643E-4 S*		*1600 Y*		*1000 Y*		*0.0133 Y*	
	7686.9	9.999E+01	7686.9 (0.06)	99.9895	7686.600	99.986 (0.00)	7690.00	100
	6905.0	1.000E-02		(0.0006)				
	6610.0	5.000E-05						
PO-210	*138.38 D*		*138.38 D*		*1000 Y*		*138.38 D*	
	5304.4	1.000E+02	5304.38 (0.12)	100.00 (0.00)	5304.400	100.00 (0.00)	5304.38	100
	4524.0	1.000E-03						
PB-210								
BI-214	*19.90 M*							
	5448.0	1.132E-02						
	5512.0	8.232E-03						
	5268.0	1.218E-03						
	5184.0	1.281E-04						
	4941.0	5.250E-05						
	5023.0	4.410E-05						
BI-210	*5.013 D*							
	4649.0	7.92E-05						
	4686.0	5.28E-05						

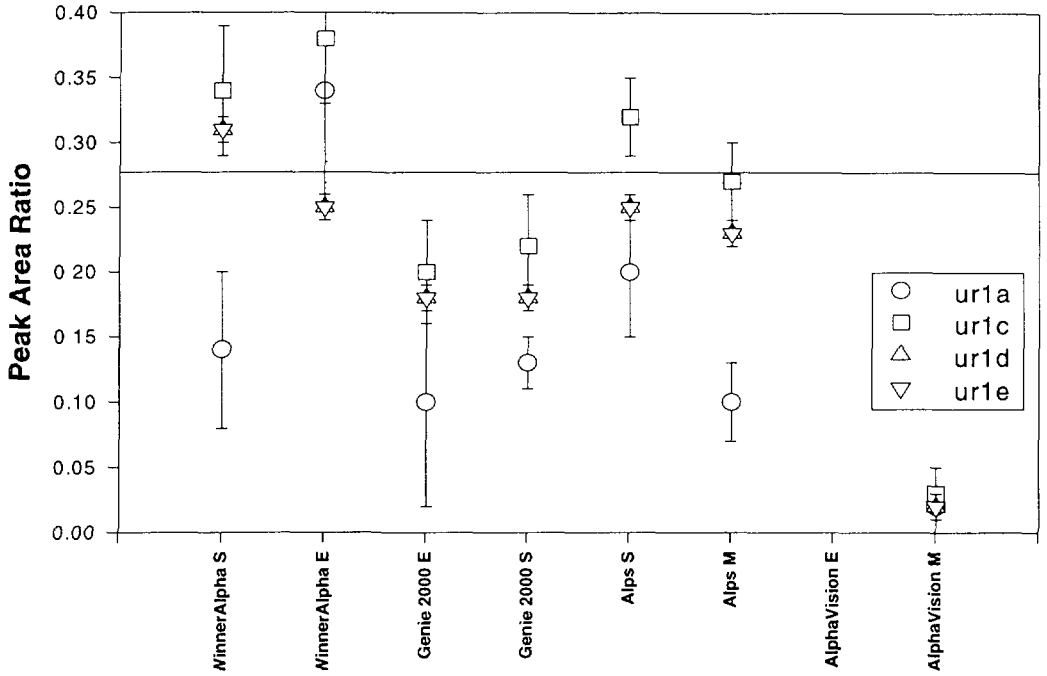

FIG. 7. Doublet ratios as found for the 4722/4775 keV doublet of ^{234}U. The horizontal line represents the target value of 0.277 ± 0.008.

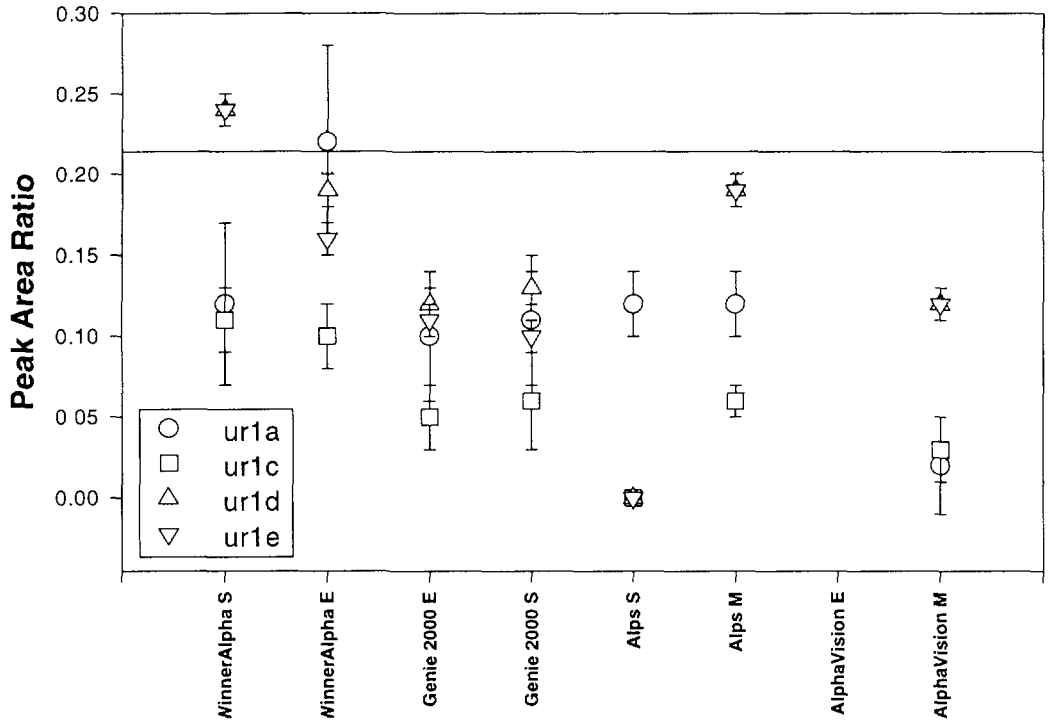

FIG. 8. Doublet ratios as found for the 4151/4198 keV doublet of ^{238}U. The horizontal line represents the target value of 0.21 ± 0.01.

6. WHAT THE NUMBERS MEAN

In this section, some examples are given of how to use the results in the tables in Appendix B to find the alpha spectrum analysis program best suited for a specific application.

6.1. LOOKING FOR A GENERAL PURPOSE ALPHA SPECTRUM ANALYSIS PROGRAM

A general purpose alpha spectrum analysis program should give X1 values in statistical control (near 1) for the "all matches" categories for all spectra. X1 and X2 values should be approximately equal for the relatively simple ^{226}Ra spectra.

6.2. LOOKING FOR A PROGRAM WITH OUTSTANDING PEAK SEARCH CAPABILITIES

Such a program should detect all relevant peaks in the ^{226}Ra spectra, in default mode. This means 2 large peaks in the ra2624 spectrum, and 1 large and 1 small peak in the ra2625 and ra2626 spectra.

The χ^2 values in the "misses" category should be near 1 for all spectra.

6.3. LOOKING FOR A PROGRAM WITH ACCURATE PEAK AREA CALCULATION IN SIMPLE SPECTRA

The X2 values in the ^{226}Ra spectra tests in the "all matches" category should be near to unity.

6.4. LOOKING FOR A PROGRAM WITH OUTSTANDING ACCURACY IN UNCERTAINTIES CALCULATION FOR SIMPLE SPECTRA

The X1 and X2 values in the "all matches" category should be approximately equal for the ^{226}Ra spectra. If X1 < X2, it indicates that the program reports uncertainties that are too large, if X1 > X2, it means that the program reports uncertainties that are too small.

6.5. LOOKING FOR A PROGRAM WITH HIGH MULTIPLET RESOLVING POWER

The more peaks in the "all matches" category for the am1, am243 and ur1 spectra, the better. However, the X value in the "false hits" and "misses" categories should also be near to one. A high value of chi-square means that some significant false peak was reported or a significant multiplet component missed. This situation usually arises from erroneous deconvolution of highly significant peaks.

7. DISCUSSION

7.1. GENERAL

The data presented in the Appendix B provide a wealth of information for intercomparison purposes. In this section, only a few of the results will be highlighted and discussed. All results in Appendix B as well as in this section are labelled with the first initial of the user who obtained the results, i.e. "E", "M", or "S".

7.1.1. Preliminary remarks

The tests produced a significant amount of information which is presented in the tables and graphs. But an accurate conclusion on the ability of the programs tested to handle a particular problem can only be drawn by the reader after defining the type of spectra and the purpose of the analysis which is to be performed. Alpha-particle spectrometry is very different from, for example, gamma ray spectrometry. Because of the very strong asymmetry inherent to alpha peaks, the low energy tails, the relatively low energy resolution of the spectrometers and the specific emission pattern of many alpha-particle emitters, it is almost impossible to find other significant peaks from the same nuclide in a different region of the spectrum. That makes the peak deconvolution features of the programs very important, especially if chemical separation has not been performed before the measurement. A program with a good deconvolution algorithm should be preferred in this case, even if it does not find all peaks automatically, since in many cases, the peaks overlap strongly and the user must introduce the peak positions manually. On the other hand, in some applications, peak deconvolution is not needed, as is the case in the analysis of the activity ratio $^{238}Pu/^{239+240}Pu$, where a careful tail extrapolation can provide accurate results. The best program here would be then the one which best estimates this low energy tail. Other situations exist in which the analyst is not interested in the individual peak areas, but only in the contributions of the different component nuclides. In these cases, a "nuclide- or library-oriented" analysis which includes complete information on the branching ratios and energies of the component peaks can represent a considerable advantage.

The statistical contents of the spectra also determine the choice of the optimal program. Using programs with complex line shape models and many peak parameters can be useless in spectra from environmental samples with a few counts while it is a very useful tool in the analysis of complex Pu mixtures with large peak areas. All these considerations must be taken into account to select, with the help of the data compiled in this paper, the program which best adapts to the user's problem.

As mentioned earlier, the peak area ratios for some nuclides have been affected by significant coincidence summing between alpha particles and conversion electrons from the gamma transition depopulating the levels fed by the alpha decay. The deformations suffered by the original pulse distributions introduce additional difficulties and sources of uncertainty in the fitting of some of the spectra. This is particularly true for nuclides with high electron conversion measured with a high efficiency. However, these are conditions as found in practical laboratory work and it was decided to keep the measuring conditions as close to reality as possible. The effects were corrected for in the computation of the reference peak areas.

7.1.2. Peak energy estimation

Only two programs reported uncertainties in the peak positions. In general, they are underestimated, and unrealistic values below 0.1 keV are reported sometimes. The problem seems to be an incomplete covariance treatment. In any case, owing to the intrinsic non-linear behaviour of the detectors, the uncertainties in the peak positions can not be easily transformed in their equivalent energy values. The reason is that the energy loss in the detector window introduces a non-linear correction and makes it very difficult, unless a small energy region is considered.

7.1.3. Peak area estimation

Because of the reasons mentioned above in this paper, none of the test spectra contains singlet peaks as one would encounter in gamma ray spectra. The separation of peaks in terms of FWHM however varies. To judge the capability of the programs to find peaks and determine the peak areas, the mixed Am and the ^{243}Am spectrum were used, with peak separations of about 2.5 and 5 FWHM, respectively. In the other groups of spectra, the peaks are less well separated.

The mixed Am spectra essentially consist of two groups of three peaks each. Smaller peaks are present but hardly detectable. The ^{243}Am spectrum contains one of the two groups also present in the mixed Am spectra with much better resolution and statistics, so the smaller components should be detectable. Also, in spectra with very good statistics, discrepancies between fitted and actual peak shape functions may show up as false hits with those programs that perform simple residual searches.

In this intercomparison, it was decided to test only relative peak areas within a single spectrum to allow for peak-shape model dependencies. This is essential in alpha-particle spectrometry, since contributions to the peak areas can be found in regions far from the peak maxima. However, for a single spectrum and a given analysis program, one would hope to find consistent peak areas independent of user or analysis mode. The actual renormalization factors to the known peak areas found for the AM1N spectrum are shown in Table VI. From the data, it seems that results obtained with Alps are biased depending on the mode of operation. The biases found are user-independent. It must be pointed out that this fact has no practical consequences since all peaks in the same spectrum are treated by the programs in an homogeneous manner.

TABLE VI. RENORMALIZATION FACTORS AND CORRESPONDING RELATIVE UNCERTAINTIES IN% FOR THE AM1N SPECTRUM

Program	default	optimized	user	default	optimized	user
AlphaVision	0.90 (2)	0.85 (2)	M	0.90 (2)	0.89 (2)	E
Alps	0.87 (2)	0.94 (1.5)	M	0.87 (2)	0.93 (1.5)	S
Genie 2000	0.97 (1.2)	0.97 (1.2)	E	1.01 (1.2)	0.98 (1.1)	S
Winner Alpha		0.97 (1.2)	E		0.96 (0.9)	S

The cause becomes apparent from Fig. 9. In this figure, the total numbers of hits for all three mixed Am spectra are shown. A score of six indicates two peaks found per spectrum, implying that the smaller components were overlooked or included in the total area of the triplets. Low numbers of hits correspond to low renormalization factors, resulting from the determination of total areas of triplets rather than the areas of a constituent peak. In the case of total area determination, renormalization factors significantly smaller than unity are to be expected.

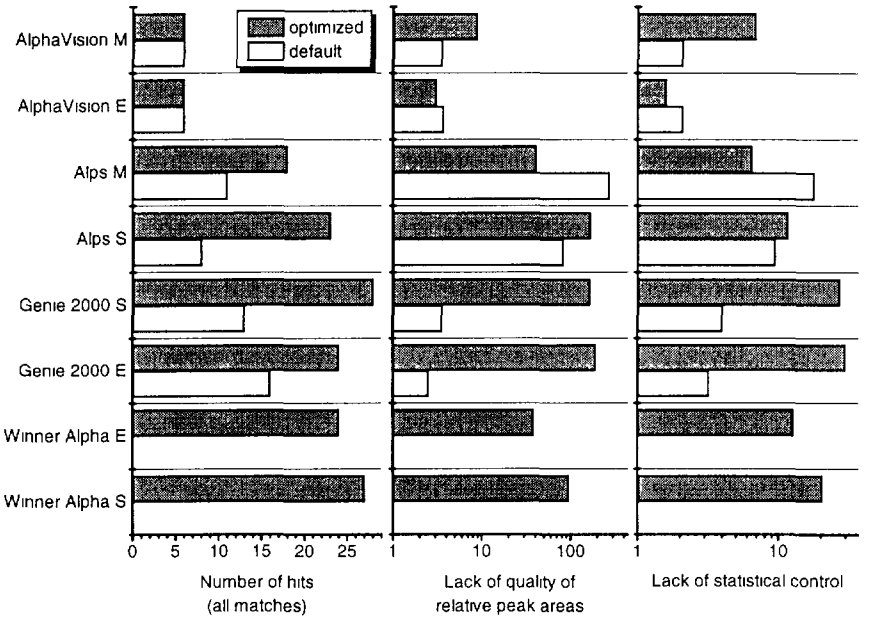

FIG. 9. All matches results for the mixed Am spectra. From left to right: number of hits, X2 and X1 values.

Also in Fig. 9, the quality of the relative peak area determination is shown in terms of X2 values for all matches. A perfect score of unity cannot be expected since the peaks were overlapping. From the figures, it can be seen that Genie 2000 in optimised mode finds more peaks than in default mode but at the expense of the quality of the relative peak areas, as opposed to Alps with user "M", where the quality of the results improves with the number of peaks found. From the underlying results it can be concluded that Genie 2000 has difficulty with the small peaks that it reports in optimised mode, not with the large ones. Also Alps and Winner Alpha score high X2 values for the small components.

The problem with the analysis of small multiplet components also becomes very clear from Figs 7 and 8. None of the programs finds the peak area ratios for ^{234}U and ^{238}U that would be in agreement with the literature values.

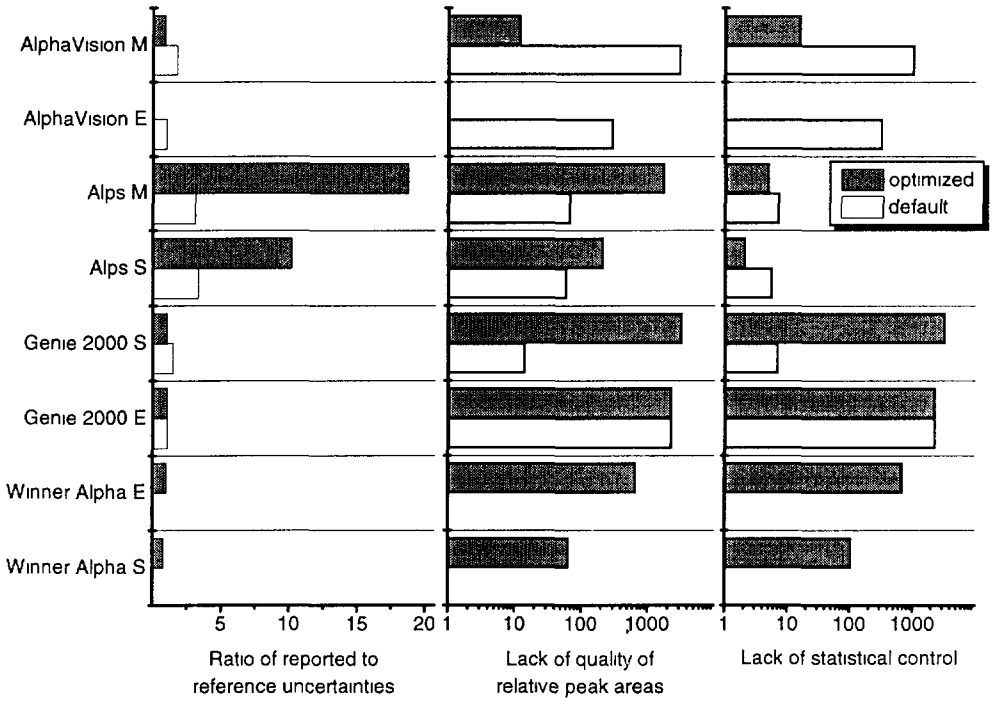

FIG. 10. All matches results for the ^{243}Am spectra. From left to right: ratio of reported to Poisson uncertainties, X2 and X1 values.

In Fig. 10, the number of hits for the ^{243}Am spectra has been omitted, since in some cases the programs failed to report any peak areas or crashed, as indicated by the "total failure" comments in the tables of result. However, the X2 values indicating the quality of the peak areas have been plotted, indicating comparable achievements by all programs. Only Alps reported false hits for the ^{243}Am spectra. AlphaVision in optimised mode with user M yields the best peak area quality.

7.1.4. Peak area uncertainty estimation (statistical control)

In Fig. 9 and Fig. 10, X1 values representing statistical control have been plotted for the "all hits" categories for the mixed Am and ^{243}Am spectra. Alps and AlphaVision (in user M optimised mode) exhibit the best control for the ^{243}Am spectra. For the mixed Am spectra, only AlphaVision is in control but at a very low number of hits. In all other cases, control is lacking, i.e. the reported uncertainties are too small (Genie 2000 as operated by user S in optimised mode sometimes even reports uncertainties below Poisson uncertainties, e.g. for 4778 keV in the UR1A spectrum it reports 83.6 ± 2.96 as peak area where the Poisson 1 s.d. uncertainty would be 9.1). In Fig. 10, the ratio of reported to reference uncertainties for the ^{243}Am spectra also has been plotted. All results seem to indicate that a full variance analysis is not performed, or at least it does not account properly for the effect of the line shape model.

7.1.5. Deconvolution capability

To discuss the deconvolution capability of the programs, the mixed Pu spectra were selected, since they contain a quadruplet with 2–2.5 FWHM separations. Results of interest are the numbers of hits, X1 values reflecting on statistical control, the numbers of misses and false hits and the associated X value. In Fig. 11 and Fig. 12, the relevant data are plotted.

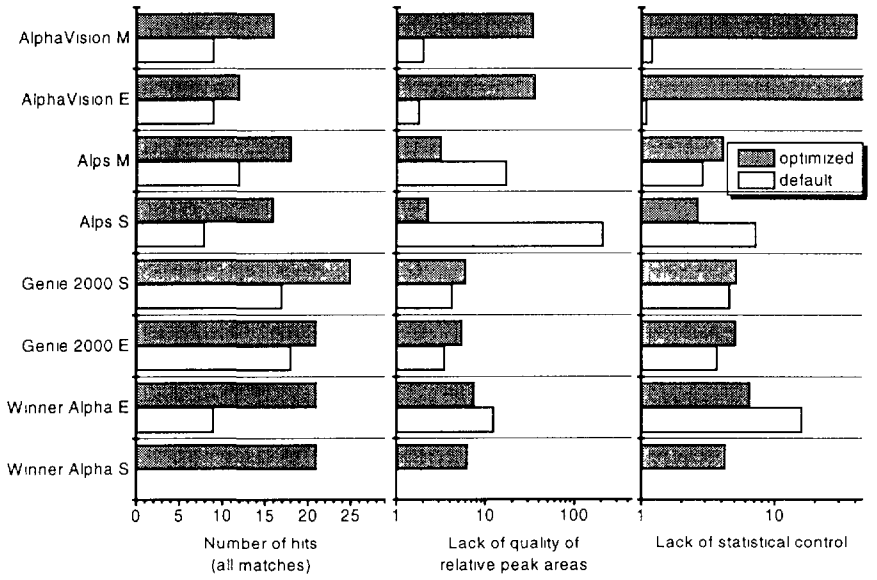

FIG. 11. Combined results for the mixed plutonium spectra: Total number of hits in the "all matches" category and associated X1- and X2 values.

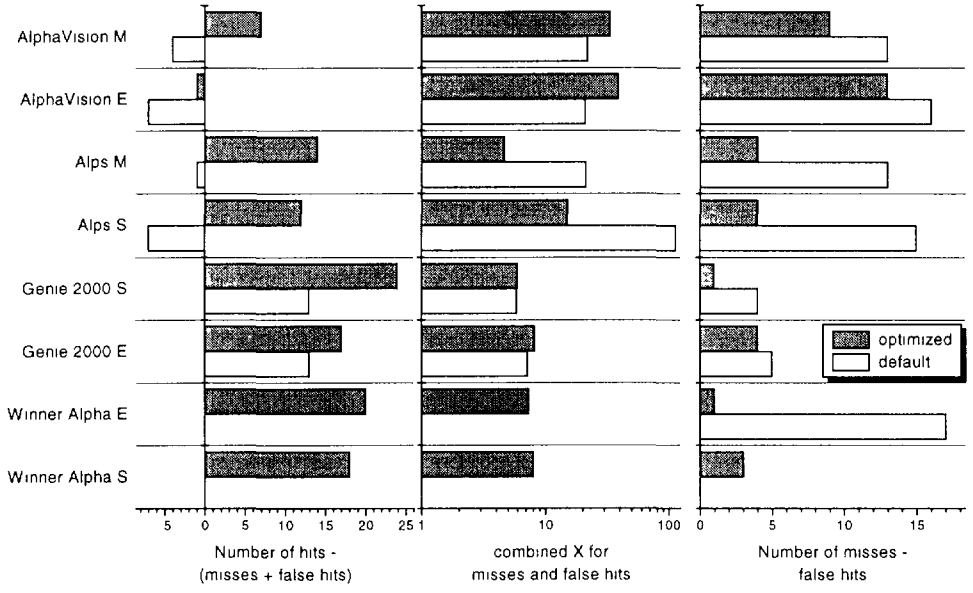

FIG. 12. Combined results for the mixed plutonium spectra: Difference between number of hits and total number of misses and false hits, X values for misses and false hits, and the difference between number of misses and number of false hits.

As was also observed in the mixed Am spectra, only Alps shows improved quality of peak areas with increased number of hits due to user optimisation. Genie 2000 and Winner Alpha exhibit the best deconvolution capabilities with roughly the same quality of peak areas and statistical control as Alps.

From the tables, it becomes clear that the programs hardly ever report false hits, but they do miss statistically significant multiplet components. This leads to the conclusion that the thresholds are set too high in the algorithms that perform residual searches looking for components that were overlooked initially.

7.1.6 User influence

As can be seen in all figures, the influence of the user operating the programs is very limited. Even though the difference between "default" and "optimised" modes of operation can be large, the differences between users are minor. Since the degree of experience with alpha analysis software of the users varied, this indicates good quality of manuals and user interfaces.

7.2. ALPHAVISION

The program came with a very poor nuclide library of alpha emitters which had to be edited to add or modify all nuclides which are present in the spectra. This is a crucial point since the program requires the user to give as much information as possible on the nuclides being analysed and this information has to be taken from the nuclide library. In fact, it is always necessary to access the library in order to select the nuclides for analysis.

There is no possibility to perform a FWHM calibration. There is no possibility to set a sensitivity for peak search or inform the program of FWHMs to expect. Since a 9-point smoothing is performed in the peak search, the only option for the user is to change the amplifier gain until he gets the sensitivity he wants. The example spectrum coming with the program has FWHMs in the order of 10 channels. 9-Point smoothing cannot be optimal for all spectra. It is unclear what the optimum FWHM for spectra to be analysed with AlphaVision would be.

Even though a plot of the fitted spectrum can be produced as part of the analysis report, the plot does not show residuals and therefore is not much of a tool when judging the quality of the results.

The program produces ASCII text report files. There is no ordering apparent in the results, and peak energies and areas are not listed on the same line which makes the reports hard to read. In the reports, peaks are identified and per peak, a "disintegrations per minute" value is given that has not been corrected for the branching ratio of the peak. The definition of this value in the manual is a little ambiguous: "net disintegrations per minute for the peak, calculated based on the CPM and the total efficiency." Obviously, however, a value called "DPM" should be corrected for branching ratio to convert counts to disintegrations.

The fact that the program does not allow a real interactive mode and only provides information either on the peaks found in the search mode or in the ones existing in the library makes it very inefficient when working out of a "repetitive" pattern. The quality of the plots is very poor since one can not estimate the quality of the fitting from the graphical output. To

work properly, the program would require an extensive library editing which probably would have to be adapted for each small change in the measuring conditions.

One concern about this program is the way in which the doublets are fitted when considered as singlet peaks. In principle one could expect to have a strong underestimation of the total area of the doublet. But this is not found in some results, which are similar for doublet and singlet peak fittings. At this moment, there is no satisfactory explanation for this effect. Automatic addition of other peaks and area renormalization are only some of the possible causes which could explain these results.

7.3. ALPS

Alps' peak shape model is very good: Singlet peaks are fitted with no apparent structure in the residuals.

The program does not have much of a memory for user input. This means that, when analysing a sequence of spectra, one has to enter the same data, like the type of spectrum to be read, or the region to be analysed, over and over again. Using macros will help in practice.

In contrast, peak analysis results are kept in memory until a new spectrum is read in. All peaks stored will show up if identification in terms of radionuclide activities is performed, as often as the spectrum was analysed. Also, after a few runs with changed settings, the results array is full and the program acts as if there are no peaks to be found in the spectrum.

7.4. GENIE-2000

This program has some nice features, i.e. the calibration menus, the good peak-search algorithm and the interactive peak fit. On the other hand, it is not easy to operate, even for simple operations, and requires a very complex manipulation of the many available menus. The colour scheme of the display is poor, which sometimes makes it difficult to correctly assign peak positions with the cursor in the calibration procedure. The interactive peak fit is quite powerful and allows an easy (although slow) modification of the number of peaks, the limits of the fitted region, etc. Using this feature, the user can have very good information on the quality of the fits, as shown in FIG.. Nevertheless, since the line shape model can not be modified, many results were identical in the automatic and interactive modes.

7.5. WINNER ALPHA

Overall this is a reasonably straightforward program to use, once the methodology has been worked out. The peak shape seems to be an extension of the gamma peak shape function, and the definition of peaks can be a bit tricky using the definition of ROIs, particularly for complex multiplets. A higher than average user interaction is required at the start of analysis with the selecting of starting values for a lot of parameters. Perhaps consideration should be given to programming in some realistic default options in order to reduce the amount of initial interaction.

ROIs had to be edited to facilitate the insertion or deletion of peaks before re-analysing the spectrum. Rather than this cumbersome method of interactive fitting, it would be very useful if some form of insert/delete at cursor could be performed such as can be done in the gamma analysis version INTERWINNER.

The determination of the peak parameters seems to be stable against different initial choices for the parameters. That makes the fitting process easy to do and allows the user to test different choices without the risk of a computer crash.

8. CONCLUSIONS

Of the four programs tested, three are more or less comparable with respect to quality of peak area determination and deconvolution power: Alps, Genie 2000, and Winner Alpha. Of these three, Genie 2000 yielded the best results by a narrow margin. Exceptional are the results for the ^{243}Am spectra, where Alps performed the best in terms of statistical control and the library-oriented AlphaVision as optimized by user M yielded the best peak areas. All programs have difficulty with small peaks close to large ones.

All programs exhibit lack of statistical control, especially where the deconvolution of multiplets or analysis of spectra with very good statistics are concerned: uncertainties in reported peak areas are underestimated in all cases. The same is true for the peak energy estimates, if reported at all.

Generally, the programs performed the same independent of the user operating them.

As a final conclusion, it is clear from the results that there is room for improvement for the developers.

REFERENCES

[1] GARCIA-TORANO, E., ACENA, M.L., BORTELS, G., MOUCHEL, D., Nucl. Instr. and Meth. **A312** (1992) 317.

[2] NYLANDSTED LARSEN, A., BORTELS, G., DENECKE, B., Nucl. Instr. and Meth. **219** (1984) 339.

[3] AKOVALI, Y.A., Nuclear Data Sheets **66** (1992) 897.

[4] FIRESTONE, R.B., SHIRLEY, V.S., Table of Isotopes, 8th edition, Wiley & Sons (1996).

[5] BROWNE, E., FIRESTONE, R.B., Table of Radioactive Isotopes, Wiley & Sons (1986).

Appendix A

THE TEST SPECTRA

In this Appendix, plots of the relevant sections of the alpha spectra used in the intercomparison are shown. Of each set of spectra, only the spectrum with the best statistics is shown. These are UR1E, AM1N, MPU-17, RA2624, and AM243-3.

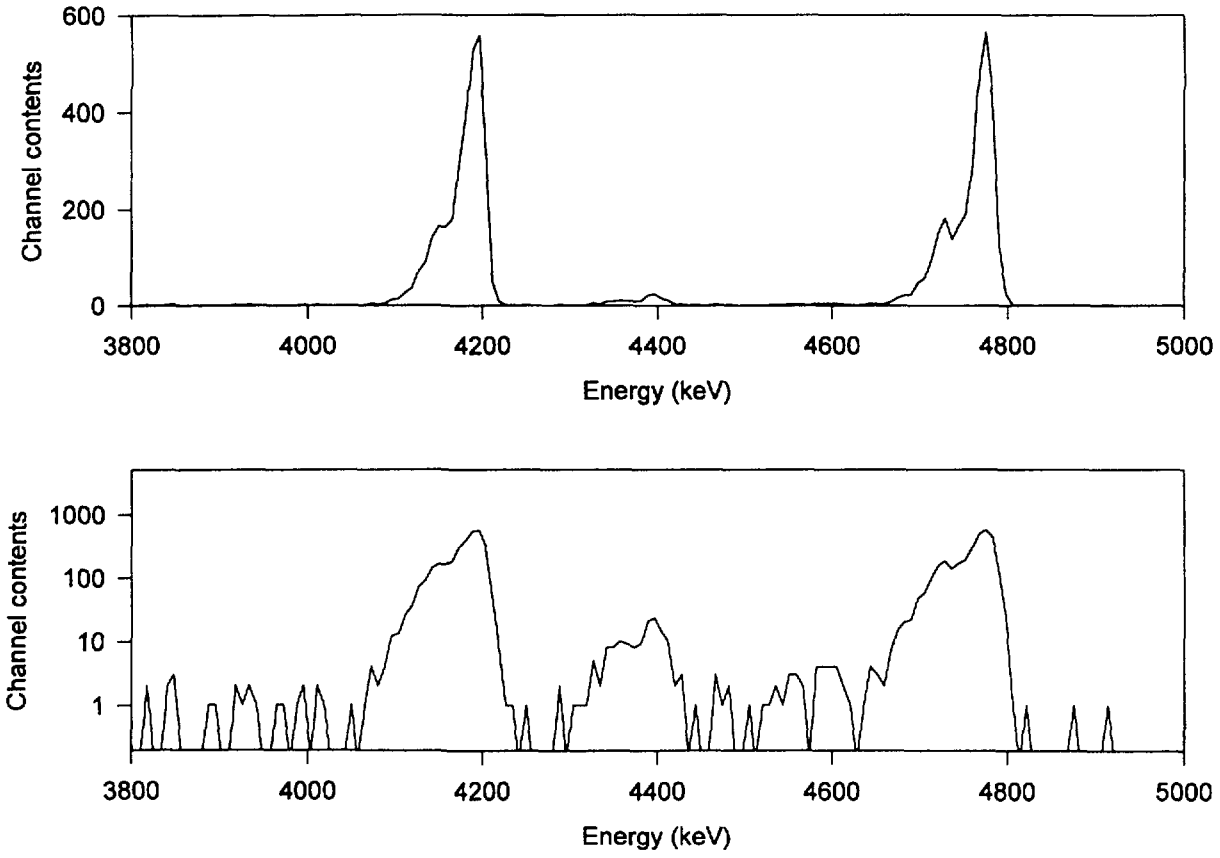

FIG.13. Spectrum UR1E.

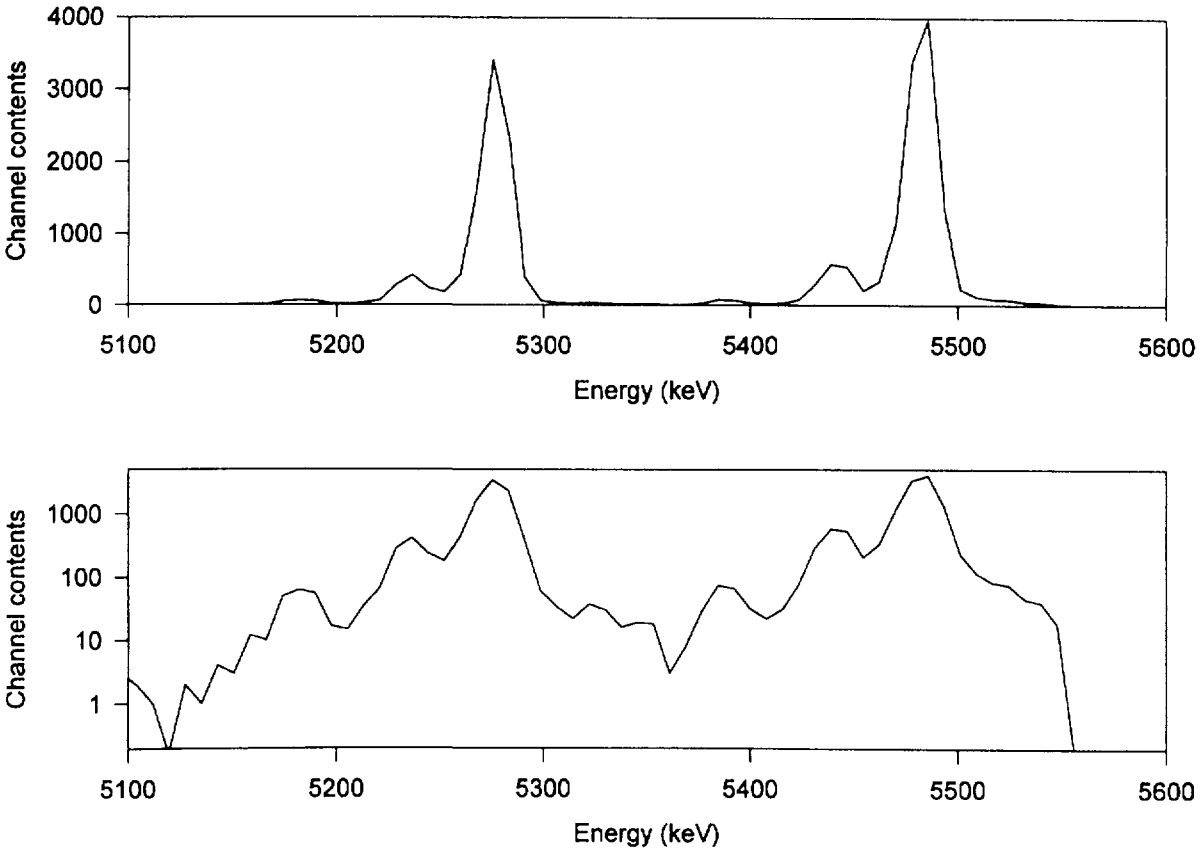

FIG.14. Spectrum AM1N.

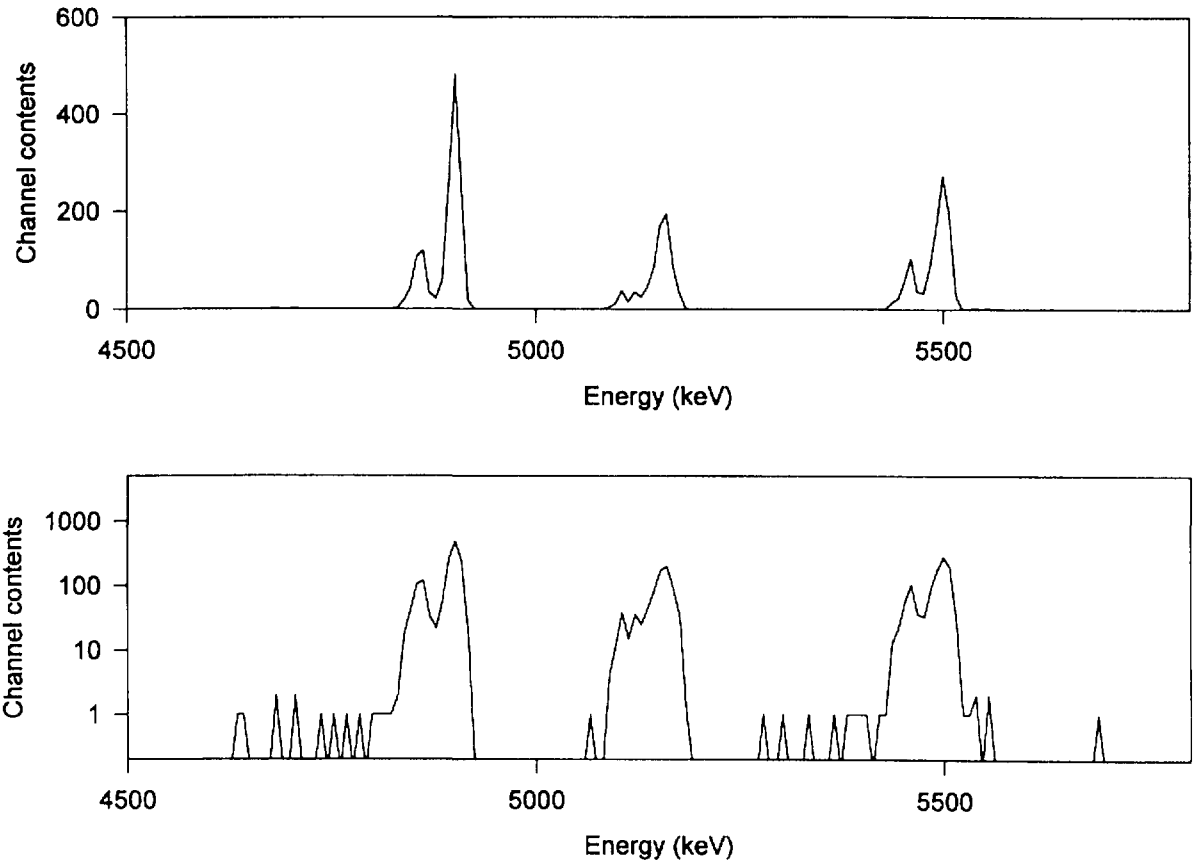

FIG.15. Spectrum MPU-17.

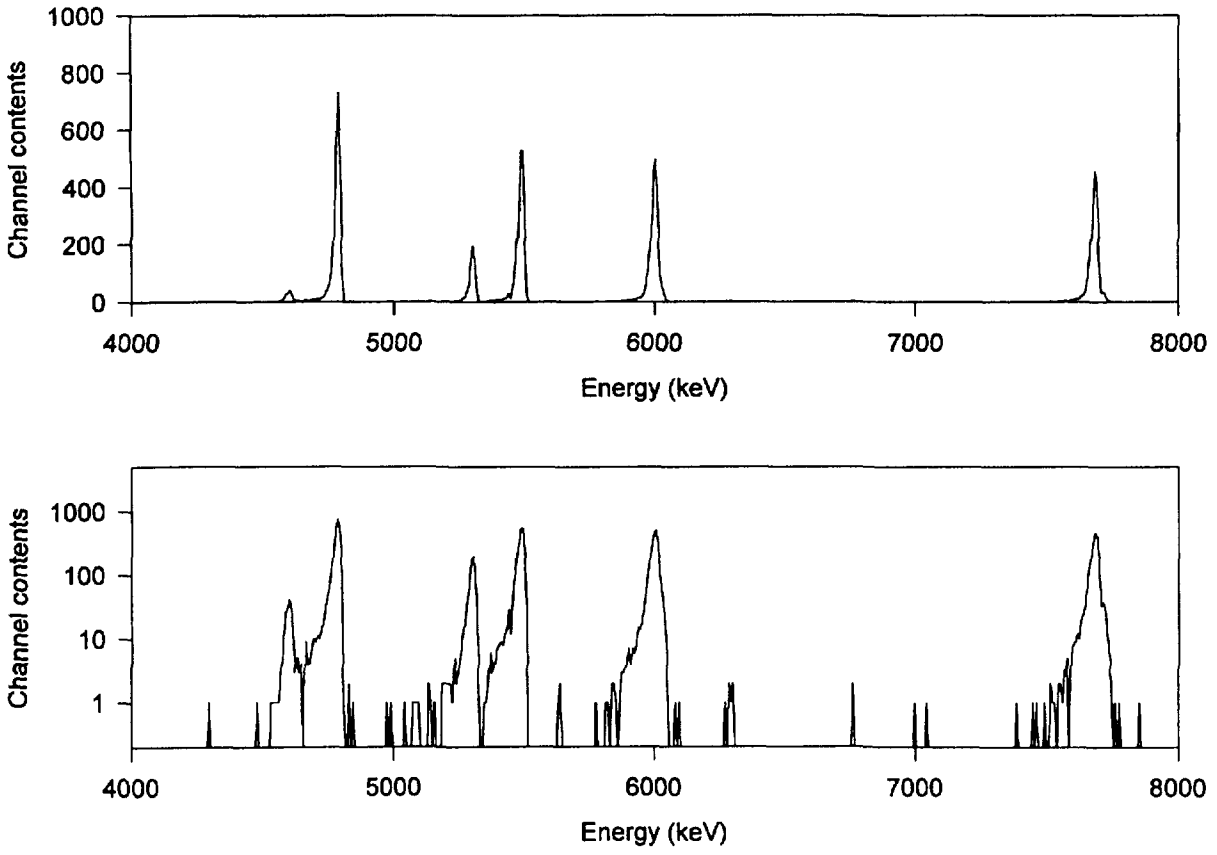

FIG.16. Spectrum RA2624.

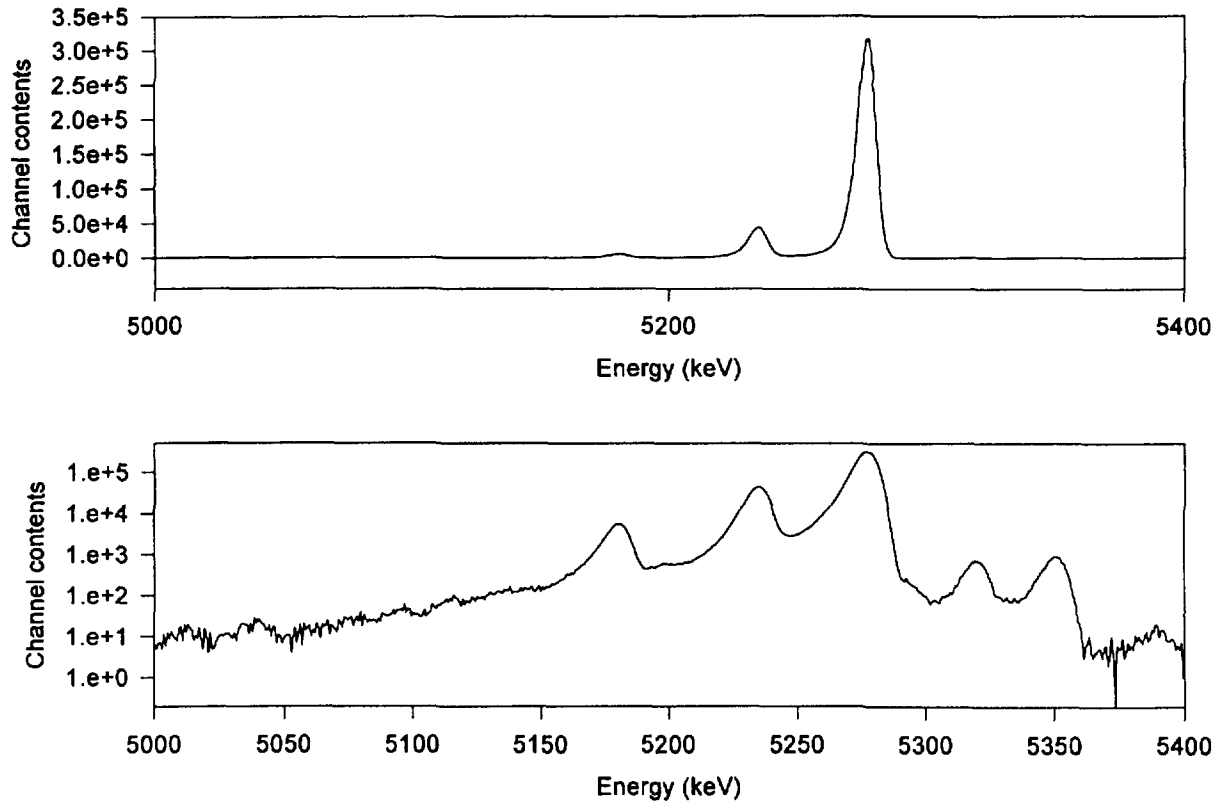

FIG.17. Spectrum AM243-3.

Appendix B

TABLES OF RESULTS

In this Appendix, the tables of statistical results are presented for all programs, program modes, spectra and users. Since renormalization was applied, no X values exist in the "large" and "all matches" categories if the number of peaks in these categories is 0 or 1. If a category does not contain any peaks at all, likewise no X values exist.

TABLE VII. ALPHAVISION [M]

	Large			High ref unc			Small			All matches			Misses		False hits		Total	
	N	X1	X2	N	X1	X2	N	X1	X2	N	X1	X2	N	X	N	X	N	X
default																		
ra226-4	2	0 5	1 4	0	-	-	0	-	-	2	0 5	1 4	0	-	0	-	2	0 5
ra226-5	1	-	-	0	-	-	1	0 1	0 2	2	0 1	0 2	0	-	0	-	2	0 1
ra226-6	1	-	-	0	-	-	1	0 3	0 9	2	0 3	0 9	0	-	0	-	2	0 3
mpu-14	3	0 7	1 3	0	-	-	0	-	-	3	0 7	1 3	4	56	0	-	7	38
mpu-15	3	1 7	3 0	0	-	-	0	-	-	3	1 7	3 0	4	44	0	-	7	30
mpu-17	3	1 0	2 0	0	-	-	0	-	-	3	1 0	2 0	7	45	2	0 0	12	29
am1m	2	0 7	1 3	0	-	-	0	-	-	2	0 7	1 3	8	33	0	-	10	29
am1n	2	3 1	4 7	0	-	-	0	-	-	2	3 1	4 7	10	1E2	0	-	12	95
am10	2	2 5	4 9	0	-	-	0	-	-	2	2 5	4 9	8	43	0	-	10	38
am243-1	3	72	59	0	-	-	0	-	-	3	72	59	7	1E2	0	-	10	1E2
am243-2	7	2E2	1E3	0	-	-	0	-	-	7	2E2	1E3	2	1E2	0	-	9	2E2
am243-3	6	3E3	8E3	0	-	-	1	0 0	0 0	7	2E3	6E3	2	2E2	0	-	9	2E3
url a	0	-	-	0	-	-	2	0 0	0 0	2	0 0	0 0	3	10	0	-	5	7 6
url c	2	0 0	0 0	0	-	-	0	-	-	2	0 0	0 0	4	20	0	-	6	16
url d	2	1 4	1 7	0	-	-	1	4 0	9 8	3	2 7	5 8	12	52	0	-	15	45
url e	2	1 3	1 7	0	-	-	1	4 0	9 9	3	2 6	5 8	12	52	0	-	15	45
optimised																		
ra226-4	2	0 6	1 6	0	-	-	0	-	-	2	0 6	1 6	0	-	0	-	2	0 6
ra226-5	1	-	-	0	-	-	1	0 1	0 1	2	0 1	0 1	0	-	1	2E2	3	78
ra226-6	1	-	-	0	-	-	1	0 4	1 0	2	0 4	1 0	0	-	0	-	2	0 4
mpu-14	5	79	63	0	-	-	1	29	18	6	69	54	2	34	0	-	8	59
mpu-15	4	35	36	0	-	-	1	17	12	5	30	30	3	47	0	-	8	37
mpu-17	4	91	80	0	-	-	1	40	23	5	78	66	6	52	1	0 0	12	57
am1m	2	0 5	1 0	0	-	-	0	-	-	2	0 5	1 0	8	33	0	-	10	29
am1n	2	16	18	0	-	-	0	-	-	2	16	18	11	10	0	-	13	93
am10	2	3 8	7 3	0	-	-	0	-	-	2	3 8	7 3	8	44	0	-	10	39
am243-1	5	8 6	4 5	0	-	-	0	-	-	5	8 6	4 5	5	1E2	0	-	10	73
am243-2	5	27	23	0	-	-	0	-	-	5	27	23	5	90	0	-	10	62
am243-3	5	13	8 7	0	-	-	0	-	-	5	13	8 7	5	1E2	0	-	10	74
url a	0	-	-	0	-	-	4	17	14	4	23	18	1	9 9	1	32	6	22
url c	2	1E2	75	0	-	-	2	33	21	4	61	39	2	11	1	95	7	50
url d	3	6E2	3E2	0	-	-	2	4E2	2E5	5	5E2	9E4	10	6 6	0	-	15	2E2
url e	3	6E2	3E2	0	-	-	2	4E2	2E5	5	5E2	1E4	10	6 4	0	-	15	1E2

TABLE VIII. ALPHAVISION [E]

	Large			High ref unc			Small			All matches			Misses		False hits		Total	
	N	X1	X2	N	X1	X2	N	X1	X2	N	X1	X2	N	X	N	X	N	X
default																		
ra226-4	2	0 5	1 4	0	-	-	0	-	-	2	0 5	1 4	0	-	0	-	2	0 5
ra226-5	1	-	-	0	-	-	1	0 1	0 2	2	0 1	0 2	0	-	0	-	2	0 1
ra226-6	1	-	-	0	-	-	1	0 3	0 9	2	0 3	0 9	0	-	0	-	2	0 3
mpu-14	3	0 9	1 7	0	-	-	0	-	-	3	0 9	1 7	5	50	0	-	8	36
mpu-15	3	1 2	2 2	0	-	-	0	-	-	3	1 2	2 2	5	39	0	-	8	28
mpu-17	3	0 9	1 7	0	-	-	0	-	-	3	0 9	1 7	8	43	2	0 0	13	29
am1m	2	0 7	1 3	0	-	-	0	-	-	2	0 7	1 3	8	33	0	-	10	29
am1n	2	3 1	4 8	0	-	-	0	-	-	2	3 1	4 8	11	94	0	-	13	87
am10	2	2 5	4 9	0	-	-	0	-	-	2	2 5	4 9	8	43	0	-	10	38
am243-1	3	2E2	2E2	0	-	-	0	-	-	3	2E2	2E2	7	1E2	0	-	10	2E2
am243-2	3	3E2	3E2	0	-	-	0	-	-	3	3E2	3E2	7	3E2	0	-	10	3E2
am243-3	2	5E2	4E2	0	-	-	0	-	-	2	5E2	4E2	8	3E2	0	-	10	4E2
ur1a	0	-	-	0	-	-	2	0 0	0 0	2	0 0	0 0	3	10	0	-	5	7 6
ur1c	2	0 0	0 0	0	-	-	0	-	-	2	0 0	0 0	4	20	0	-	6	16
ur1d	2	1 4	1 7	0	-	-	1	4 0	9 8	3	2 7	5 8	12	52	0	-	15	45
ur1e	2	1 3	1 7	0	-	-	1	4 0	9 9	3	2 6	5 8	12	52	0	-	15	45
optimised																		
ra226-4	1	-	-	0	-	-	0	-	-	1	-	-	1	67	0	-	2	67
ra226-5	1	-	-	0	-	-	0	-	-	1	-	-	1	35	0	-	2	35
ra226-6	1	-	-	0	-	-	0	-	-	1	-	-	1	35	0	-	2	35
mpu-14	4	73	54	0	-	-	0	-	-	4	73	54	4	68	0	-	8	70
mpu-15	4	42	34	0	-	-	0	-	-	4	42	34	4	48	0	-	8	45
mpu-17	4	1E2	74	0	-	-	0	-	-	4	1E2	74	7	54	2	0 0	13	57
am1m	2	1 2	2 4	0	-	-	0	-	-	2	1 2	2 4	8	33	0	-	10	30
am1n	2	0 2	0 3	0	-	-	0	-	-	2	0 2	0 3	11	95	0	-	13	87
am10	2	3 4	6 6	0	-	-	0	-	-	2	3 4	6 6	8	43	0	-	10	39
am243-1	total failure																	
am243-2	total failure																	
am243-3	total failure																	
ur1a	0	-	-	0	-	-	3	0 5	3 2	3	0 8	4 8	2	14	0	-	5	7 6
ur1c	2	0 3	0 4	0	-	-	1	0 9	2 9	3	0 6	1 6	3	27	0	-	6	16
ur1d	2	1 7	2 2	0	-	-	1	4 7	12	3	3 2	7 1	12	52	0	-	15	45
ur1e	2	1 7	2 2	0	-	-	1	4 7	12	3	3 2	7 1	12	52	0	-	15	45

TABLE IX. ALPS [M]

	Large			High ref unc			Small			All matches			Misses		False hits		Total	
	N	X1	X2	N	X1	X2	N	X1	X2	N	X1	X2	N	X	N	X	N	X
default																		
ra226-4	1	-	-	0	-	-	0	-	-	1	-	-	1	78	0	-	2	78
ra226-5	1	-	-	0	-	-	0	-	-	1	-	-	1	36	0	-	2	36
ra226-6	1	-	-	0	-	-	0	-	-	1	-	-	1	37	0	-	2	37
mpu-14	3	49	45	0	-	-	1	01	00	4	33	30	4	39	0	-	8	24
mpu-15	3	49	21	0	-	-	1	03	02	4	33	14	4	30	0	-	8	19
mpu-17	3	57	40	0	-	-	1	01	01	4	38	27	7	34	1	00	12	23
am1m	2	01	02	0	-	-	1	77	2E3	3	38	8E2	7	39	0	-	10	39
am1n	3	93	22	0	-	-	2	21	2E2	5	15	1E2	8	60	0	-	13	45
am10	2	04	13	0	-	-	1	37	20	3	21	17	7	52	0	-	10	41
am243-1	total failure																	
am243-2	total failure																	
am243-3	5	73	68	0	-	-	0	-	-	5	73	68	5	1E2	1	80	11	64
urla	0	-	-	0	-	-	1	00	00	1		-	4	20	0	-	5	20
urlc	2	16	43	0	-	-	0	-	-	2	16	43	4	19	0	-	6	15
urld	2	13	20	0	-	-	2	14	16	4	14	18	10	64	0	-	14	50
urle	1	-	-	0	-	-	0	-	-	1	-	-	15	59	0	-	16	59
optimised																		
ra226-4	2	02	03	0	-	-	0	-	-	2	02	03	0	-	0	-	2	02
ra226-5	1	-	-	0	-	-	1	03	02	2	03	02	0	-	0	-	2	03
ra226-6	1	-	-	0	-	-	1	06	04	2	06	04	0	-	0	-	2	06
mpu-14	4	63	57	0	-	-	2	30	20	6	50	42	1	17	0	-	7	70
mpu-15	4	36	25	0	-	-	2	80	52	6	53	36	1	14	0	-	7	67
mpu-17	5	73	68	0	-	-	1	03	02	6	59	55	3	88	1	00	10	62
am1m	4	08	08	0	-	-	2	52	33	6	26	14	3	44	0	-	9	32
am1n	4	63	66	0	-	-	3	18	2E2	7	12	92	5	76	0	-	12	10
am10	4	43	27	0	-	-	1	04	02	5	33	21	5	41	0	-	10	37
am243-1	total failure																	
am243-2	total failure																	
am243-3	6	50	2E3	0	-	-	0	-	-	6	50	2E3	4	76	2	45	12	38
urla	0	-	-	0	-	-	4	48	27	4	65	37	1	15	0	-	5	52
urlc	2	10	97	0	-	-	4	64	44	6	72	55	0	-	0	-	6	72
urld	3	65	43	1	46	24	2	51	78	6	29	21	7	32	0	-	13	14
urle	3	66	43	1	50	26	2	50	76	6	29	21	7	32	0		13	14

TABLE X. ALPS [S]

	Large			High ref unc			Small			All matches			Misses		False hits		Total	
	N	X1	X2	N	X1	X2	N	X1	X2	N	X1	X2	N	X	N	X	N	X
default																		
ra226-4	1	-	-	0	-	-	0	-	-	1	-	-	1	71	0	-	2	71
ra226-5	1	-	-	0	-	-	0	-	-	1	-	-	1	24	0	-	2	24
ra226-6	1	-	-	0	-	-	0	-	-	1	-	-	1	37	0	-	2	37
mpu-14	2	12	43	0	-	-	1	01	01	3	60	22	5	78	0	-	8	58
mpu-15	1	-	-	0	-	-	1	18	8E2	2	18	8E2	5	83	0	-	7	72
mpu-17	2	39	77	0	-	-	1	06	03	3	22	40	7	86	1	00	11	60
am1m	2	01	02	0	-	-	0	-	-	2	01	02	8	36	0	-	10	32
am1n	3	94	23	0	-	-	2	21	2E2	5	15	1E2	7	68	3	6E2	15	2E2
am10	1	-	-	0	-	-	0	-	-	1	-	-	9	2E2	0	-	10	2E2
am243-1	5	56	60	0	-	-	0	-	-	5	56	60	5	1E2	1	68	11	60
am243-2	total failure																	
am243-3	total failure																	
ur1a	0	-	-	0	-	-	1	00	00	1	-	-	4	20	0	-	5	20
ur1c	2	47	50	0	-	-	0	-	-	2	47	50	4	18	0	-	6	16
ur1d	2	06	21	0	-	-	0	-	-	2	06	21	14	47	0	-	16	44
ur1e	2	11	15	0	-	-	1	22	24	3	16	19	12	53	0	-	15	46
optimised																		
ra226-4	1	-	-	0	-	-	0	-	-	1	-	-	1	74	0	-	2	74
ra226-5	1	-	-	0	-	-	0	-	-	1	-	-	1	02	5	5E2	7	4E2
ra226-6	1	-	-	0	-	-	0	-	-	1	-	-	1	38	0	-	2	38
mpu-14	4	55	46	0	-	-	2	53	40	6	54	43	1	17	0	-	7	74
mpu-15	2	00	01	0	-	-	2	07	08	4	05	05	2	1E2	0	-	6	50
mpu-17	5	28	36	0	-	-	1	01	01	6	23	29	2	05	1	00	9	15
am1m	4	11	09	0	-	-	3	30	8E2	7	16	4E2	2	54	0	-	9	13
am1n	4	81	69	0	-	-	4	14	1E2	8	12	81	3	60	0	-	11	98
am10	4	17	14	0	-	-	4	13	66	8	83	39	2	32	0	-	10	72
am243-1	5	21	2E2	0	-	-	0	-	-	5	21	2E2	5	1E2	1	72	11	60
am243-2	total failure																	
am243-3	total failure																	
ur1a	0	-	-	0	-	-	4	25	16	4	34	22	1	14	0	-	5	29
ur1c	2	20	19	0	-	-	1	41	29	3	30	24	3	70	0	-	6	54
ur1d	3	34	34	0	-	-	2	09	07	5	18	17	11	45	0	-	16	80
ur1e	3	34	34	0	-	-	2	19	15	5	18	18	11	45	0	-	16	81

TABLE XI. GENIE 2000 [S]

	Large			High ref unc			Small			All matches			Misses		False hits		Total	
	N	X1	X2	N	X1	X2	N	X1	X2	N	X1	X2	N	X	N	X	N	X
default																		
ra226-4	2	0 3	0 4	0	-	-	0	-	-	2	0 3	0 4	0	-	0	-	2	0 3
ra226-5	1	-	-	0	-	-	1	2 4	2 6	2	2 4	2 6	0	-	0	-	2	2 4
ra226-6	1	-	-	0	-	-	1	0 0	0 0	2	0 0	0 0	0	-	0	-	2	0 0
mpu-14	4	6 6	7 2	0	-	-	2	9 6	7 1	6	7 8	7 2	1	17	0	-	7	9 3
mpu-15	4	3 6	3 9	0	-	-	1	0 4	0 3	5	2 8	3 0	2	16	0	-	7	7 3
mpu-17	5	15	12	0	-	-	1	0 5	0 3	6	12	9 4	2	0 5	1	0 0	9	7 7
am1m	3	4 2	4 4	0	-	-	0	-	-	3	4 2	4 4	7	20	0	-	10	16
am1n	4	2 3	1 8	0	-	-	2	3 1	2 0	6	2 6	1 9	7	3 8	0	-	13	3 3
am10	4	5 9	5 4	0	-	-	0	-	-	4	5 9	5 4	6	4 9	0	-	10	5 2
am243-1	6	8 4	17	0	-	-	1	0 1	0 5	7	7 0	14	3	1E2	0	-	10	41
am243-2	total failure																	
am243-3	total failure																	
ur1a	0	-	-	0	-	-	3	0 2	0 3	3	0 3	0 5	2	9 0	0	-	5	4 6
ur1c	2	2 8	3 8	0	-	-	0	-	-	2	2 8	3 8	4	16	0	-	6	14
ur1d	3	93	92	0	-	-	1	1 5	2 3	4	63	62	12	4 2	0	-	16	16
ur1e	3	93	91	0	-	-	1	1 5	2 4	4	62	61	12	4 2	0	-	16	16
optimized																		
ra226-4	2	1 8	1 3	0	-	-	0	-	-	2	1 8	1 3	0	-	2	68	4	46
ra226-5	1	-	-	0	-	-	1	0 9	0 7	2	0 9	0 7	0	-	5	16	7	14
ra226-6	1	-	-	0	-	-	1	0 2	0 2	2	0 2	0 2	0	-	2	27	4	18
mpu-14	5	7 0	14	0	-	-	4	3 0	3 9	9	5 0	8 7	0	-	0	-	9	5 0
mpu-15	5	13	11	0	-	-	3	1 6	1 2	8	8 0	7 0	0	-	0	-	8	8 0
mpu-17	5	20	17	0	-	-	3	3 8	2 7	8	13	11	2	0 5	1	0 0	11	9 1
am1m	4	4 8	4 0	0	-	-	4	55	7E2	8	33	4E2	2	2 3	0	-	10	26
am1n	4	2 1	1 5	0	-	-	6	56	1E2	10	38	91	2	0 5	0	-	12	31
am10	4	2 5	2 0	0	-	-	6	15	72	10	11	48	0	-	0	-	10	11
am243-1	5	3E3	3E3	0	-	-	0	-	-	5	3E3	3E3	5	1E2	0	-	10	2E3
am243-2	total failure																	
am243-3	total failure																	
ur1a	0	-	-	0	-	-	4	4 3	2 5	4	5 8	3 3	1	1 4	0	-	5	4 7
ur1c	2	1 4	1 8	0	-	-	2	14	9 9	4	9 5	7 2	2	2 8	0	-	6	6 8
ur1d	3	93	72	1	14	7 7	0	-	-	4	67	50	11	5 9	0	-	15	19
ur1e	3	93	71	1	19	10	2	0 3	0 4	6	41	31	8	2 5	0	-	14	17

TABLE XII. GENIE 2000 [E]

	Large			High ref unc.			Small			All matches			Misses		False hits		Total	
	N	X1	X2	N	X1	X2	N	X1	X2	N	X1	X2	N	X	N	X	N	X
default																		
ra226-4	2	1.0	1.0	0	-	-	0	-	-	2	1.0	1.0	0	-	0	-	2	1.0
ra226-5	1	-	-	0	-	-	1	0.9	1.0	2	0.9	1.0	0	-	0	-	2	0.9
ra226-6	1	-	-	0	-	-	1	0.0	0.0	2	0.0	0.0	0	-	0	-	2	0.0
mpu-14	4	8.0	8.3	0	-	-	2	0.6	0.7	6	5.1	5.3	2	25	0	-	8	11
mpu-15	4	5.6	4.8	0	-	-	2	1.0	0.8	6	3.8	3.2	1	25	0	-	7	7.4
mpu-17	4	11	9.2	0	-	-	2	0.6	0.5	6	6.8	5.7	3	17	1	0.0	10	9.4
am1m	4	4.2	3.7	0	-	-	2	0.3	0.2	6	2.6	2.3	4	1.9	0	-	10	2.3
am1n	4	2.2	1.9	0	-	-	2	3.2	2.0	6	2.6	1.9	7	3.9	0	-	13	3.4
am10	4	4.9	3.8	0	-	-	0	-	-	4	4.9	3.8	6	5.1	0	-	10	5.0
am243-1	6	4E3	4E3	0	-	-	0	-	-	6	4E3	4E3	4	1E2	0	-	10	2E3
am243-2	5	3E3	3E3	0	-	-	0	-	-	5	3E3	3E3	5	86	0	-	10	1E3
am243-3	5	1E2	56	0	-	-	0	-	-	5	1E2	56	5	1E2	0	-	10	1E2
ur1a	0	-	-	0	-	-	2	0.1	0.1	2	0.3	0.3	3	7.8	1	2.1	6	5.2
ur1c	2	0.9	1.0	0	-	-	1	17	13	3	8.8	6.8	3	7.5	0	-	6	8.0
ur1d	3	1E2	90	1	24	12	1	0.6	0.7	5	66	48	9	3.4	0	-	14	23
ur1e	3	1E2	89	1	24	12	1	0.6	0.7	5	66	48	9	3.4	0	-	14	22
optimized																		
ra226-4	2	0.2	0.2	0	-	-	0	-	-	2	0.2	0.2	0	-	0	-	2	0.2
ra226-5	1	-	-	0	-	-	1	1.5	1.1	2	1.5	1.1	0	-	0	-	2	1.5
ra226-6	1	-	-	0	-	-	1	0.0	0.0	2	0.0	0.0	0	-	0	-	2	0.0
mpu-14	5	3.7	4.3	0	-	-	2	2.9	3.8	7	3.4	4.1	1	33	0	-	8	7.6
mpu-15	5	14	14	0	-	-	2	0.2	0.2	7	9.2	9.6	1	27	0	-	8	12
mpu-17	5	16	20	0	-	-	2	0.7	0.6	7	11	14	3	17	1	0.0	11	12
am1m	4	4.2	3.7	0	-	-	4	54	7E2	8	33	4E2	2	2.3	0	-	10	26
am1n	4	2.2	1.9	0	-	-	4	70	2E2	8	41	1E2	4	4.4	0	-	12	28
am10	4	4.6	3.6	0	-	-	4	23	1E2	8	15	60	2	3.1	0	-	10	13
am243-1	6	4E3	4E3	0	-	-	0	-	-	6	4E3	4E3	4	1E2	0	-	10	2E3
am243-2	5	3E3	3E3	0	-	-	0	-	-	5	3E3	3E3	5	86	0	-	10	1E3
am243-3	5	82	46	0	-	-	0	-	-	5	82	46	5	1E2	0	-	10	94
ur1a	0	-	-	0	-	-	4	3.4	3.1	4	4.5	4.1	1	1.4	1	2.1	6	3.4
ur1c	2	1.5	1.7	0	-	-	2	16	11	4	11	8.2	2	2.8	0	-	6	8.0
ur1d	3	96	73	1	16	8.6	1	0.7	0.9	5	52	39	9	3.3	0	-	14	18
ur1e	3	93	70	1	17	9.0	2	0.5	0.7	6	41	30	8	2.5	0	-	14	17

TABLE XIII. WINNER ALPHA [E]. THIS PROGRAM DID NOT HAVE A "DEFAULT" MODE. THE RESULTS UNDER HEADING "OPT 1" WERE OBTAINED BY INTEGRATION OF PEAK AREAS, THE RESULTS UNDER THE HEADER "OPT 2" BY FITTING

	Large			High ref unc			Small			All matches			Misses		False hits		Total	
	N	X1	X2	N	X1	X2	N	X1	X2	N	X1	X2	N	X	N	X	N	X
opt 1																		
mpu-14	3	45	34	0	-	-	0	-	-	3	45	34	6	71	0	-	9	65
mpu-15	3	20	16	0	-	-	0	-	-	3	20	16	5	40	0	-	8	29
mpu-17	3	09	08	0	-	-	0	-	-	3	09	08	8	45	2	00	13	30
opt 2																		
ra226-4	2	07	07	0	-	-	0	-	-	2	07	07	0	-	0	-	2	07
ra226-5	1	-	-	0	-	-	1	53	40	2	53	40	0	-	0	-	2	53
ra226-6	1	-	-	0	-	-	1	20	19	2	20	19	0	-	0	-	2	20
mpu-14	4	18	16	0	-	-	3	28	35	7	15	18	0	-	0	-	7	15
mpu-15	4	16	15	0	-	-	3	30	29	7	23	22	0	-	0	-	7	23
mpu-17	5	86	79	0	-	-	2	82	82	7	85	80	2	05	1	00	10	58
am1m	4	09	08	0	-	-	4	37	1E2	8	22	82	2	17	0	-	10	17
am1n	4	09	07	0	-	-	4	15	23	8	92	14	4	29	0	-	12	69
am10	4	39	35	0	-	-	4	100	29	8	74	18	2	23	0	-	10	63
am243-1	5	1E2	63	0	-	-	0	-	-	5	1E2	63	5	10	0	-	10	1E2
am243-2	5	2E3	2E3	0	-	-	0	-	-	5	2E3	2E3	5	84	0	-	10	9E2
am243-3	5	1E2	63	0	-	-	0	-	-	5	1E2	63	5	1E2	0	-	10	1E2
urla	0	-	-	0	-	-	4	03	03	4	04	04	1	13	0	-	5	07
urlc	2	17	16	0	-	-	2	52	31	4	40	26	2	26	0	-	6	34
urld	3	37	33	1	48	26	0	-	-	4	26	23	10	59	0	-	14	11
urle	3	38	34	1	92	49	2	04	04	6	17	15	7	23	0	-	13	85

TABLE XIV. WINNER ALPHA [S]. THIS PROGRAM DID NOT HAVE A "DEFAULT" MODE

	Large			High ref unc			Small			All matches			Misses		False hits		Total	
	N	X1	X2	N	X1	X2	N	X1	X2	N	X1	X2	N	X	N	X	N	X
optimized																		
ra226-4	2	13	12	0	-	-	0	-	-	2	13	12	0	-	0	-	2	13
ra226-5	1	-	-	0	-	-	1	01	01	2	01	01	0	-	0	-	2	01
ra226-6	1	-	-	0	-	-	1	10	09	2	10	09	0	-	0	-	2	10
mpu-14	4	25	24	0	-	-	3	11	21	7	68	12	1	32	0	-	8	10
mpu-15	4	17	17	0	-	-	3	56	55	7	36	36	0	-	0	-	7	36
mpu-17	4	76	63	0	-	-	3	13	26	7	10	16	3	17	1	00	11	11
am1m	4	29	27	0	-	-	4	42	4E2	8	25	2E2	2	24	0	-	10	20
am1n	4	34	29	0	-	-	6	43	1E2	10	30	73	2	05	0	-	12	24
am10	4	20	19	0	-	-	5	83	21	9	59	14	1	22	0	-	10	55
am243-1	5	1E2	65	0		-	0	-	-	5	1E2	65	5	10	0	-	10	1E2
am243-2	total failure																	
am243-3	total failure																	
urla	0	-	-	0	-	-	4	32	25	4	43	34	1	14	0	-	5	36
urlc	2	15	14	0	-	-	2	47	32	4	36	26	2	25	0	-	6	32
urld	3	10	11	1	05	03	2	16	19	6	49	52	7	20	0	-	13	32
urle	3	10	11	1	05	03	2	17	20	6	49	52	7	20	0	-	13	32

Appendix C

ALPHA-ELECTRON COINCIDENCE SUMMING AND ITS INFLUENCE ON THE DETERMINATION OF THE REFERENCE DATA

Most alpha-particle emitting nuclides do not emit mono-energetic alpha particles: They decay by more than one alpha transition to different levels of the daughter nuclide. The excited levels fed by the transitions usually depopulate in a very short time to the ground state by gamma emission, either directly or by a series of intermediate levels. If a gamma transition is converted, atomic electrons can be emitted instead of gamma photons. The ratio between the number of conversion electrons and gamma rays for a particular transition is called the conversion coefficient α_T.

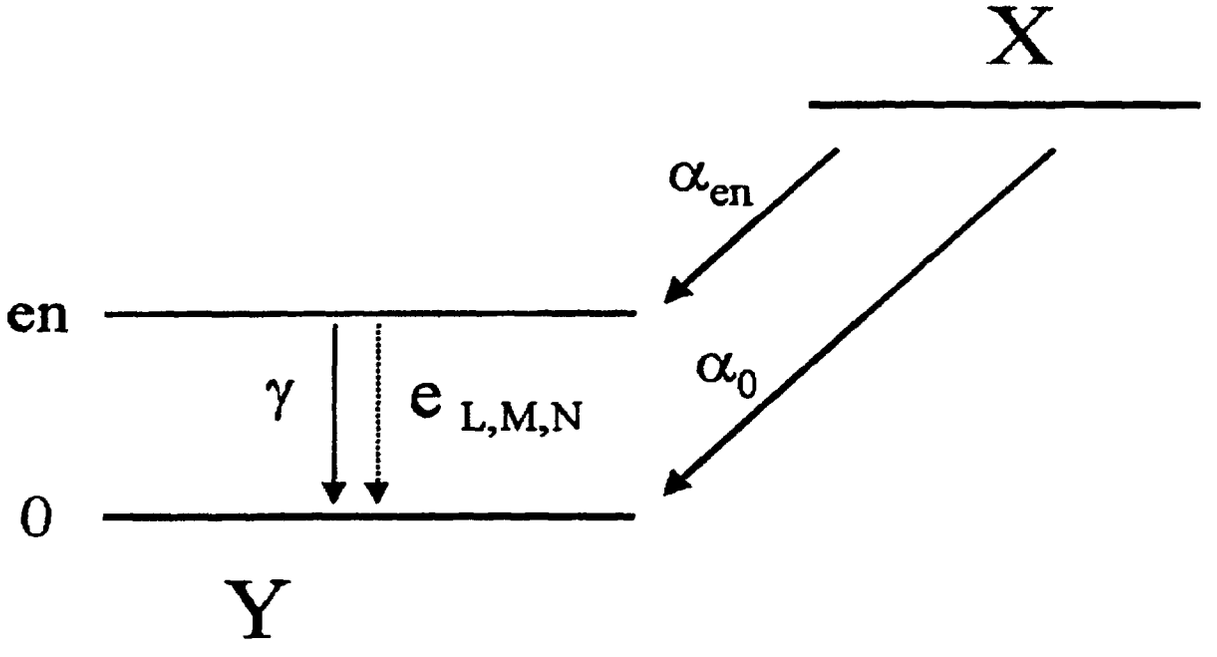

FIG 18. Example of an alpha-particle emitting nuclide with coincidence-summing.

From the point of view of the detector, the emission of alpha particles and conversion electrons depopulating the associated level can be considered as simultaneous events. Hence, the probability exists of detecting both particles in coincidence, thus obtaining a combined voltage pulse which would correspond to the sum of their energy contributions. The ratio between coincident and single pulses is proportional to the detection efficiency for both kinds of radiation, and, implicitly, to the solid angle of the measurement.

The observed effect is an apparent modification of the theoretical peak area ratios of the alpha transitions in the measured spectra, since the spectral distribution of the number of pulses corresponding to the different alpha branches are modified by the coincidence distributions of electrons and alpha particles. In the high efficiency limit, if the gamma transition depopulating the excited level was fully converted and the detector had unit efficiency for both kinds of particles, no single events would be observed. In what follows we describe the basics aspects of this effect and how to evaluate its influence in a practical case.

For the sake of simplicity let us consider the ideal case (Fig. 18) of a nuclide X decaying by emission of two alpha transitions, with energies $E_{\alpha en}$ and $E_{\alpha o}$ and probabilities $P_{\alpha en}$ and $P_{\alpha o}$

respectively The α_{en} transition feeds the excited level of energy *en* in the daughter nuclide, while α_0 feeds the ground level on Y. Let ε_α and ε_e be the detection efficiency of our system for alpha particles and electrons respectively. For the Si implanted detectors used in this work, it can be assumed that ε_α does not depend on the particle energy and is basically defined by the relative solid angle ($\Omega/4\pi$) of the experimental setup.

Let I_γ be the intensity of the gamma transition depopulating the excited level and α_T the total conversion coefficient. To simplify the calculations, let us also assume that only L,M and N+ conversion electrons are possible. For each α_{en} particle emitted, relative fractions $1/(1+\alpha_T)$ of gamma rays and $\alpha_T/(1+\alpha_T)$of conversion electrons will be emitted ($I_\gamma + \alpha_T I_\gamma = 1$).

If we measure the alpha particle spectra of this nuclide in the absence of coincidences, we will obtain two alpha peaks whose areas would correspond to the theoretical branching ratio:

$$R = (\varepsilon_\alpha P_{\alpha_{en}})/(\varepsilon_\alpha P_{\alpha_0}) = P_{\alpha_{en}}/P_{\alpha_0}$$

This expression holds when there are no coincident pulses because of a low conversion coefficient of the γ transition, and/or a detector system with a very small efficiency. In other cases, i.e., electron emission from converted transitions, the number of coincident events must be accounted for to obtain the actual peak area ratios. For the example we are considering here, that implies estimating the number of coincident pulses associated to the α_{en} transition and their contribution to the measured pulse height spectra. Alpha particles emitted in the transition α_0 are not coincident with conversion electrons, except for random coincidences, which can be neglected at low countrates and are not the issue here.

The number of conversion electrons emitted in coincidence with each α_{en} particle is:

$$N_e = \alpha_T /(1 + \alpha_T)$$

And the fraction of electrons detected in coincidence with each α_{en} particle **detected** will be:

$$\varepsilon_e N_e = \varepsilon_e \alpha_T/(1 + \alpha_T)$$

These coincident events will give rise to voltage pulses higher than those corresponding to $E_{\alpha_{en}}$. The final energy deposited into the detector depends on the atomic shell (L,M,N+) from which the converted electron has been emitted. Several sets of coincidence distributions are then produced, with different energies and probabilities. Since all events corresponding to coincident pulses are situated outside the α_{en} peak, they must be discounted from it to obtain the corrected value of single detection alpha events $P_{\alpha_{en}}$:

$$P_{\alpha_{en}} \varepsilon_\alpha (1- \varepsilon_e \alpha_T/(1 + \alpha_T)).$$

For a transition highly converted (α_T »1) and observed with a detector with $\varepsilon_e = \varepsilon_\alpha \approx 1$, no single detection of alpha particles would be observed and:

$$P_{\alpha_{en}} \varepsilon_\alpha (1- \varepsilon_e \alpha_T/(1+\alpha_T)) \approx 0.$$

by adding the energies of their components, the alpha particle and the conversion electron. A number of energies are possible:

$$E_{\alpha en} + E_L\ ,\ E_{\alpha en} + E_M\ ,\ E_{\alpha en} + E_N$$

whose relative probabilities can be calculated according to the partial conversion coefficients for each shell or subshell.

$$\alpha_L,\ \alpha_M,\ \alpha_N\ .$$

The contribution of these groups of pulses to the peak formed by the α_0 transition depends on their energy. Since the binding energy for electrons M and N+ is usually very small, their energy will be close to that of the gamma photon and these groups will mainly add to the alpha peak. In the case of distributions originated from L electrons, the coincident pulses usually show up in a part of the spectrum situated between both alpha peaks (α_0 and α_{en}) and can not always be considered to contribute to the main peak α_0. For this reason, a numerical evaluation must be done of the energies involved before making the appropriate corrections. If we assume that only M and N electron distributions will add to the α_0 peak, the corresponding probability for this transition will increase to:

$$P_{\alpha_o}\,\varepsilon_\alpha + \varepsilon_e\,\varepsilon_\alpha\,\alpha_M\,I_\gamma + \varepsilon_e\,\varepsilon_\alpha\,\alpha_N\,I_\gamma$$

and the measured branching ratio will become:

$$R^m = (\varepsilon_\alpha\,P_{\alpha en} - \varepsilon_e\,\varepsilon_\alpha\,\alpha_T\,I_\gamma)/(\varepsilon_\alpha\,P_{\alpha_o} + \varepsilon_e\,\varepsilon_\alpha\,\alpha_M\,I_\gamma + \varepsilon_e\,\varepsilon_\alpha\,\alpha_N\,I_\gamma)$$

That is:

$$R^m = (P_{\alpha en} - \varepsilon_e\,\alpha_T\,I_\gamma)/(P_{\alpha_o} + \varepsilon_e\,\alpha_M\,I_\gamma + \varepsilon_e\,\alpha_N\,I_\gamma)$$

Let us now make a numerical estimation of the magnitude of this effect for a practical case. Suppose that the atomic and nuclear parameters of our example nuclide are those presented in Table XV, with all energies expressed in keV. They must be considered as average values for each shell, since we will not detail subshell structures.

TABLE XV. VALUES GIVEN TO THE ATOMIC AND NUCLEAR PARAMETERS OF OUR IDEAL NUCLIDE

Alpha emission		**Gamma and electron emission**			
$E_{\alpha en}$	4950	E_Γ	50	α_T	9
$P_{\alpha en}$	0.22	E_L	30	α_L	6
E_{α_o}	5000	E_M	45	α_M	2
P_{α_o}	0.78	E_{N+}	49	α_{N+}	1

The theoretical branching ratio, in absence of coincidences would be:

$$R = 0.22/0.78 = 0.282$$

The total number of electrons emitted in coincidence with each α_{50} particle is:

$$N_e = \alpha_T/(1 + \alpha_T) = 0.9$$

From which 0.6 will come from the L shell, with an average energy of 30 keV, 0.2 will be from M with 45 keV in average, and the remaining 0.1 from N+, with about 49 keV.

Let us now suppose that we are using an experimental setup with a detection efficiency of 0.2 for alpha particles. This is basically defined by the relative solid angle ($\Omega/4\pi$) of the measurement, since the intrinsic efficiency of Si detectors alpha particles is close to 1. For electrons with the energies considered here and, because of the backscattering in the detector window, the efficiency would be slightly lower, let us say $0.85 \times 0.2 = 0.17$.

For every alpha particle feeding the level of 50 keV, the number and characteristics of the associated coincident events are presented in Table XVI.

TABLE XVI. PROBABILITIES AND ENERGIES OF α_{50} +CONVERSION ELECTRON COINCIDENCE-SUMMING EVENTS

Coincidence event	Relative probability	Energy (keV)
$e_L + \alpha_{50}$	$0.6 \times 0.17 = 0.102$	$4950 + 30 = 4980$
$e_M + \alpha_{50}$	$0.2 \times 0.17 = 0.034$	$4950 + 45 = 4995$
$e_{N+} + \alpha_{50}$	$0.1 \times 0.17 = 0.017$	$4950 + 49 = 4999$

The next step is to evaluate the spectral contribution of the coincidence events. The corresponding pulses can not add to the α_{50} peak, since the integration of this peak, with a maximum in the energy 4950 keV, can not include pulses corresponding to energies outside than $4950 \pm 3\sigma$, which is far from the 4980 keV of the less energetic pulses (see Fig. 2). In other words, all coincident pulses are missing from the α_{50} peak Hence, the intensity of the α_{50} peak, after correction by coincidence summing will be proportional to:

$$P_{\alpha_{50}} = 0.22\ (1\text{-}0.102\text{-}0.034\text{-}0.017) = 0.1863$$

For similar reasons, not all coincident pulses add up to the alpha peak α_0 Making a similar comparison in terms of 3σ limits, it can be seen (see Fig. 19) that only events with M and N+ electrons will be contributing to the main peak, whose intensity will then be proportional to:

$$P_{\alpha_0} = 0.78 + 0.22\ (0.034 + 0.017) = 0.7912$$

Note that now $P_{\alpha_0} + P_{\alpha_{50}} \neq 1$, since some events have been detected in coincidence.

If we renormalize the total probability to unit, we obtain:

$$P_{\alpha_{50}} = 0.1906 \text{ and } P_{\alpha_0} = 0.8094$$

And the corresponding ratio between both branches will be now:

$$R^m = 0.1906/0.8094 = 0.235$$

This is significantly different from the value R = 0.282 obtained for the situation of non-coincident detection. As an additional consequence, R will also be affected by a higher uncertainty.

The example presented and evaluated here is a simple one, but calculations for more complex decay schemes with several alpha and gamma transitions proceed in a very similar manner. All relevant gamma transitions have to be evaluated and their contributions in terms of conversion electrons must be computed and combined by adding or subtracting their respective probabilities to the distributions of alpha particles.

X rays are also emitted in the atomic rearrangements following the emission of conversion electrons. They can also give rise to coincidence-summing events. Being out of the scope of this appendix, the study of their influence has not been considered here. It must nevertheless be mentioned, that in some cases corrections of this kind cannot be neglected.

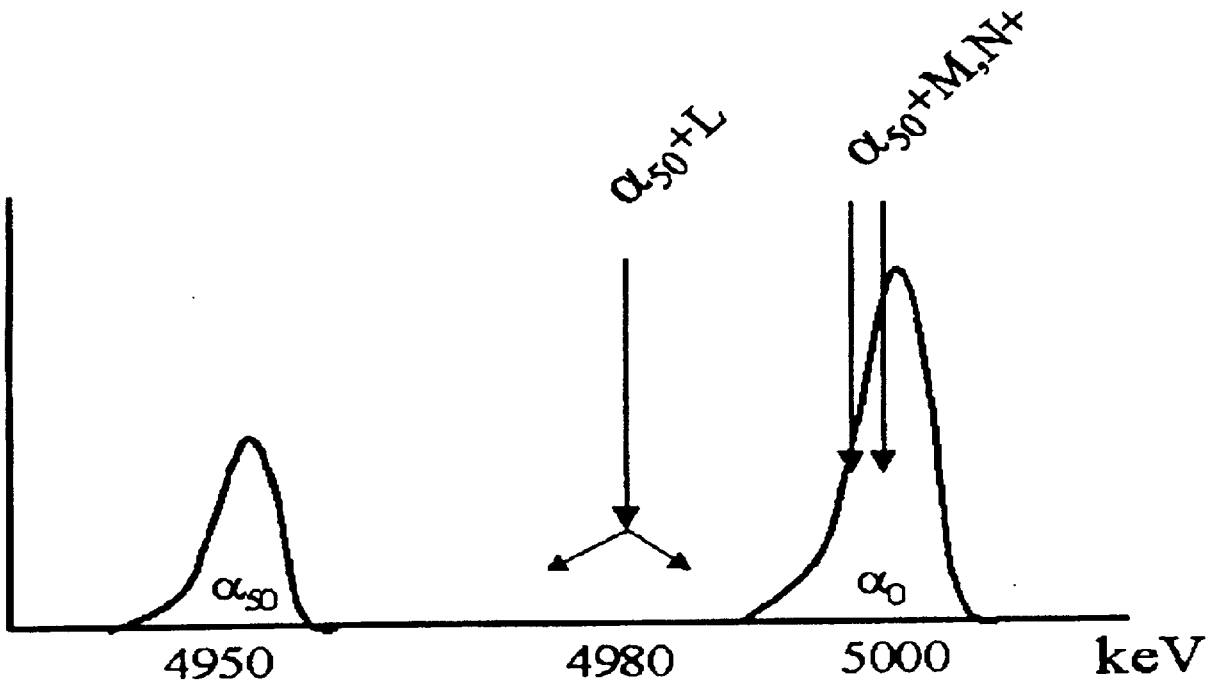

FIG. 19. Pulse distribution produced by single and coincident events.

Appendix D

USER -, PROGRAM- AND SPECTRUM-SPECIFIC NOTES

1. ALPHAVISION

1.1. User M

1.1.1. ^{226}Ra analysis

In peak search mode, the program detected all peaks in the ^{226}Ra spectrum with the best statistics. Not all peaks were identified, though: The 5% ^{226}Ra peak at 4602 keV was identified only if the 95% peak at 4785 keV was removed from the library. Apparently the identification routine is unaware of the possibility of one nuclide emitting two alpha energies. Similarly, in library-oriented mode, the program would fit and report the 4602 keV peak only after removal of the 4785 keV peak from the library. To get the program to analyse all peaks in library oriented mode, a ^{226m}Ra nuclide was inserted in the library to contain the 5% 4602 keV peak.

All this is all the more surprising because the library supplied with the program *does* contain nuclides that emit more than one alpha energy.

1.1.2. Pu-mixture analysis

The program reports many peaks with 0 areas in peak search mode. In library oriented mode, it does not find or report all peaks listed in the library.

1.1.3. Am-mixture analysis

In the spectrum with the best statistics, only the ^{243}Am was deconvoluted, but only two components were detected, one of them being an unknown detector contamination.

1.1.4. Natural uranium analysis

Remarkably, in peak search mode results were reported for all peaks in the library - most of them with 0 areas however. In library-oriented mode, all peaks reported on had non-0 areas, but not all peaks were reported on. It seems that the library is used to pad the list of peaks after analysis in peak search mode.

1.1.5. ^{243}Am analysis

The program detected too many peaks in these spectra, and in peak search mode, assigned them erroneously to the peaks in the library, e.g. the peak at 5321 keV was identified as the 5275.3 keV, with the peak match criterion set at 50 keV. With the criterion at 10 keV, however, the identification was correct.

1.2. User E

1.2.1. ^{226}Ra analysis

Peak search and fit:

The six main peaks contributing to these spectra have been found by the peak search algorithm and correctly assigned to the corresponding nuclides. The weaker peak, which corresponds to ^{226}Ra was not identified by the program, which apparently, is unable to assign more than one peak to any nuclide.

Library search mode

In this mode the result was indicated nuclide by nuclide. The areas assigned to the ^{226}Ra did not match either the area of the major peak, or the addition of both peaks.

It seems that there are some differences in the fitting process when selecting one or the other mode of operation.

1.2.2. Pu mixture analysis

Peak search and fit

Four peaks were found and assigned to the isotopes 242, 238, 239 and 240, the latter with zero area in all spectra. That implies that the three groups (242, 239 + 240 and 238) have been analyzed as if they were formed by a single peak each. This could be a serious problem in relative tracer analysis if the nuclide used as a tracer would not have a similar structure and would lead to an important bias in the results

Library search mode

In this case, results were provided separately for each nuclide, although it is almost impossible to perform a good deconvolution of the 239 + 240 multiplet.

1.2.3. Natural Uranium analysis

Peak search and fit

The small contribution of the isotope 235 was not detected in all cases. For the other two groups , the program was not able to find the doublet structure of both and were fitted as single peaks. Since the ratio of the major to the minor components in both groups is similar (2.5 for ^{234}U and 3.3 for ^{238}U) that will not produce a strong effect on the results. But the same consideration made for the mixed Pu spectra applies here.

Library search mode

The ^{235}U contribution was present in the results.

1.2.4. Am mixture analysis

Peak search and fit

The same problem as in the case of the Pu mixture arises. Two main peaks were identified, one corresponding to each nuclide. But the fitted areas are very similar in this mode and in the

library mode in which four peaks are considered. At this moment there is no a good explanation to this fact.

1.2.5. ^{243}Am analysis

Peak search and fit

The program was only able to detect the three main peaks in these high statistics spectra. The two weak lines (about 0.2% each) were ignored.

Library search mode failed completely. No report whatsoever was produced by the program..

2. ALPS

2.1. Both users

2.1.1. ^{226}Ra analysis

Good results were obtained automatically once the right sensitivity and FWHM estimate had been established. The automatic peak search works well.

2.1.2. Pu-mixture analysis

Even in the spectrum with the best statistics, the program only separated the two ^{242}Pu peaks automatically, and this with the sensitivity set to 11. The interactive fitting option, where the user specifies region and initial peak position estimates, was used for the other doublets in these spectra in the optimised runs.

2.1.3. Natural uranium analysis

As with the Pu-mixture analysis, the main peaks were automatically found, but at a sensitivity setting of 35 or less, these classic doublets of ^{234}U and ^{238}U were fitted as singlets. The exception was UR1A, with poor statistics and low count rates, where a sensitivity of 11 had to be used. Interactive fitting easily optimised the program's fits to the peaks.

2.1.4. Am-mixture analysis

Only the major components were detected in automatic mode. In interactive fitting, the small peaks to the high energy side of the main peaks were generally not fitted, even when their position had been pointed out to the program. However, perseverance and an a priori knowledge of the expected spectrum, revealed that if these peaks were specified at 1 to 2 channels higher than in reality, the program would not reject them, but would "pull" them, either as the true doublet or a synthetic singlet, into their correct fit position.

2.1.5. ^{243}Am analysis

The analyses of these spectra proved to be somewhat random in terms of success. One user, who attempted to analyse all 3 spectra, found that spectrum AM243-2 could be analysed

without any problems. However, analysis of the two other spectra consistently crashed the program.

No satisfactory explanation was found for this, particularly as the other user arbitrarily selected only one of the spectra, AM243-1, for fitting and did not experience this problem. Due to the high conversion gain, the latter user did require to compress the spectrum by combining adjacent channels in order to be able to set an appropriate ROI to obtain a reasonable continuum subtraction. In general, the fit around the centroid region of each peak was good, but such high precision spectra graphically highlight the problems (generally available) peak fitting models have with the tailing experienced in all alpha spectra; the model cannot follow the data and consequently "underfits" these regions. In any analysis of these high precision spectra, large residuals (in the order of 500) can be expected and were indeed obtained.

3. GENIE 2000

3.1. Both users

3.1.1. ^{226}Ra analysis

None of the spectra caused any undue problems with the peak fitting and optimisation. All peaks were found by the peak search algorithm by using the standard significance parameter s = 3. A poor fit can be observed in some of the most energetic peaks (5945 keV and 5448 keV) because of the small satellite peaks on the right side. All singlet peak areas were easily optimised by fitting.

3.1.2. Pu mixture analysis

Using a significance factor of 3, the ^{238}Pu and ^{242}Pu doublets tended to be identified as singlets, though this could easily be modified by adding the peaks back in during IPF. The $^{239/240}$Pu multiplet was identified as a doublet. To fit correctly, a third, and sometimes a fourth, peak was interactively inserted.

With a significance factor of 2, five or six peaks were found in automatic search mode, in the case of six peaks the Pu doublets being fitted well. Again, using IPF, a third peak was added to fit the 239+240 multiplet correctly.

For all interactive fitting, it was required to slightly modify the results by changing the region limits. The region limits as set by the automated peak fitting, invariably tended to be to narrow, restricting the ability of the program to fit the peaks.

3.1.3. Natural uranium analysis

The peak search algorithm at its default setting struggled to detect the most significant peaks in this series of spectra. Indeed in the case or spectrum UR1A, no peaks at all were found due to the low number of counts. When the significance factor was lowered, spurious peaks could be detected. Once peak regions had been defined, the fits were easily optimised interactively.

3.1.4. Am mixture analysis

The automatic mode was not able to detect all peaks in the AM1M spectrum. For the others, with a greater number of counts in the spectra, the main peaks were found. However, this showed a slight dependency on the initial parameter setup, which, as mentioned before was very slightly different between the users for the continuum. User E obtained automatic fits giving doublets in each of the Am groups for both AM1N and AM10 (no continuum), whereas user S (with continuum) obtained a triplet fit to both groups in the case of AM1N.

Additional peaks were introduced in the interactive peak fitting procedure to fully reflect the multiplet nature of these Am groups. The low intensity doublet on the high energy side of each group could normally only be successfully fitted with a singlet.

3.1.5. ^{243}Am spectra

All 5 major peaks were found and fitted in automatic mode. No modifications were included in the interactive peak fit, since the line shape can not be modified and this was the only possible improvement. Large residuals can be observed in the fits (40 standard deviations), specially in the areas between peaks. That reflects the fact that accurate peak models are needed to fit high statistics spectra.

The program crashed when analysing two of these spectra with user S, who employed a continuum function.

4. WINNER ALPHA

4.1. User E

4.1.1. ^{226}Ra analysis

The peak search function worked well after modifying the sensitivity parameter from the standard value 3 to 5. Nothing else was required and all peak ROIS were automatically marked by the program.

In the reports provided by the program, the "net" areas are sometimes larger than the "gross" areas. Although it is not indicated in the manual, It was concluded that net areas correspond to the "fitted" areas and the gross areas to the integration of the areas under the ROIS.

The uncertainties stated by the program are not realistic. For the first peak of ^{226}Ra they are even lower than the Poisson uncertainties derived from the peak areas. It seems that the fitting uncertainties are not considered or at least, they are not reasonably estimated.

4.1.2. Pu mixture analysis

These spectra were analysed in two different ways, by simple area integration and by fitting. In the second case, only three peaks were used for the multiplet 239+240 which has 5 component peaks. The peak search algorithm was not able to detect all relevant peaks and the manual ROI edition was used. The results depend on the number of channels taken for each peak, specially for the 239+240 group.

4.1.3. Natural uranium analysis

These spectra were analysed as two groups of lines (2 peaks each) corresponding to the isotopes 234 and 238, for which the activity ratio should be unity. In the last spectrum of the series two peaks of 235 were also included in the fittings. The peak search algorithm failed to find the peaks, even in the spectra with higher statistics.

4.1.4. ^{243}Am analysis

These spectra have a high statistical significance and a good energy resolution. The analysis of these peaks should assess the ability of the line shape model to closely follow the measured peaks. This is not the case for Eurisys which uses a simple (but not documented) model. The residual spectra showed very significant discrepancies, and the uncertainties assigned to the peak areas are much lower than they should be.

4.2. User S

4.2.1. ^{226}Ra analysis

Generally no problem in fitting the six main peaks in these spectra. It was found that varying the sensitivity between 1 and 10 had little effect except for RA2626 where an extra peak was located for SENS >= 3. Generally SENS = 5 was used.

4.2.2. Pu mixture analysis

For all three spectra, peak locate found 3 ROIs (i.e. 3 peaks). Those for Pu242 and Pu238 were each edited into 2 ROIs to facilitate a correct fitting of doublets and a good energy calibration. Peak shape was taken from the 4900 keV line of Pu242. Attention was then turned to the Pu239/Pu2-40 multiplet. It was found that one could easily adjust the ROI to fit a doublet, but that this caused a poor fit at the high energy side. Very judicious definition of the ROIs enabled three peaks to be fitted to this region, the residuals and visual inspection indicating that this was the best fit obtainable. As mentioned this procedure was applied to all three spectra.

4.2.3. Natural uranium analysis

A 2 point energy calibration was used with the main U234 and U238 peaks. Shape calibration was from the 4196 keV peak. Peak locate fitted both U234 and U238 as singlets. The ROIs were edited to give doublets before proceeding. For all of the spectra, the fitted peak energies often appeared to be between 5 and 10 keV adrift despite what should have been an accurate energy calibration. For all spectra the ratio of branching ratios within the nuclide (high energy/low energy) were high, but improved with improving statistics e.g. UR1D reported U238 ratio = 4.1 (expected 3.4) and U234 ratio = 3.2 (expected 2.6). For both the higher statistics spectra, UR1D and UR1E, it was noticeable that the program could not accurately fit the peak tailing (underfitting).

4.2.4. Am mixture analysis

Again a 2 point energy calibration was used fitting the 2 strongest lines. ROIs were such that the program initially fitted as doublets. Peak parameters were in each case taken from the ^{243}Am 5275 keV line.

AM1M: As with other programs, this spectrum was fitted as 2 groups with 4 components each. i.e. in each group the low intensity doublet on the high energy side was fitted as a singlet. Reasonable fit obtained.

AM1N: In this spectrum the 5321 and 5350 keV peaks were distinguishable and could be fitted separately. The 5512 and 5544 keV peaks were slightly less distinguishable, but with a very judicious selection of ROIs they could just be fitted as 2 singlets. Hence a 2 group fit each containing 5 peaks.

AM10: This spectrum fell in between the two previous. The 5321 and 5350 keV peaks from Am^{243} could be fitted as singlets, but the 5512 and 5544 keV peaks of Am241 could not be so and were fitted as a singlet covering the doublet i.e. two groups fitted, the Am^{243} group with 5 peaks and the Am241 group with 4 peaks.

In general no great difficulties were found with these spectra.

4.2.5. ^{243}Am analysis

Peak locate: SENS = 5 — many peaks!

SENS = 10 — better, but still too sensitive and giving spurious peaks.

SENS = 15 — ok, conversion electron ROI deleted before proceeding.

The 5 main Am^{243} peaks were fitted. Very high residuals were reported, due to the high precision of the data points, particularly in the tailing regions where the peak shape model could not follow the data accurately. In the report file, it was noted that very low uncertainties were reported on the peak areas (<1% in most cases). These are somewhat unrealistic and may be inductive of the uncertainties not being fully propagated. Checking the LARA library did indeed reveal that the P_{alpha} uncertainties were set to zero. Also only the three main alpha radiations were listed, explaining why only these lines were correctly labelled; the 5321 keV line was not labelled and 5350 keV was allocated to ^{228}Th.

CONTRIBUTORS TO DRAFTING AND REVIEW

Blaauw, M.	Interfaculty Reactor Institute, Delft University of Technology, Netherlands
Fazinic, S.	International Atomic Energy Agency
García-Toraño, E.	CIEMAT, Spain
Woods, S.	National Physical Laboratory, United Kingdom

The diskette may be obtained from:

Physics Section
International Atomic Energy Agency
Wagramer Strasse 5
P.O. Box 100
A-1400 Vienna, Austria