



Forschungszentrum Karlsruhe
in der Helmholtz-Gemeinschaft

Wissenschaftliche Berichte

FZKA 7377

**Automation of a Gamma
Spectrometric Analysis
Method for Naturally Occuring
Radionuclides in Different
Materials (NORM)**

O. Marzocchi

Institut für Strahlenforschung

Juni 2009

Forschungszentrum Karlsruhe

in der Helmholtz-Gemeinschaft

Wissenschaftliche Berichte

FZKA 7377

**Automation of a Gamma
Spectrometric Analysis Method for
Naturally Occuring Radionuclides in
Different Materials (NORM)**

Olaf Marzocchi

Institut für Strahlenforschung

Forschungszentrum Karlsruhe GmbH, Karlsruhe

2009

Für diesen Bericht behalten wir uns alle Rechte vor

Forschungszentrum Karlsruhe GmbH
Postfach 3640, 76021 Karlsruhe

Mitglied der Hermann von Helmholtz-Gemeinschaft
Deutscher Forschungszentren (HGF)

ISSN 0947-8620

urn:nbn:de:0005-073774

Zusammenfassung

Automatisierung einer Gamma-spektrometrischen Analyse-Methode für natürlich vorkommende Radionuklide in verschiedenen Materialien (“NORM”)

Dieser Bericht beschreibt eine Technik, welche die Standardanalyseroutine verbessert, die in einem physikalischen Messlabor genutzt wird. Die neue Routine kann Peaks in Spektren von Norm-Proben (“Naturally occurring radioactive materials”, natürlich vorkommende radioaktive Stoffe) besser identifizieren. Der Einsatz einer neuen spezifischen Bibliothek mit Gamma Linien hilft der Software mehr Peaks zu finden. Das Ergebnis ist eine schnellere Analyseroutine, die weniger Benutzerzeit braucht und die weniger Fehler erzeugt.

Ein neues Programm mit einer einfacheren und optimierten Schnittstelle wurde entwickelt und validiert.

Abstract

This work presents an improvement over the standard analysis routine used in the Physikalisches Messlabor to detect gamma peaks in spectra from naturally occurring radioactive materials (NORM). The new routine introduces the use of custom libraries of known gamma peaks, in order to ease the work of the software than can therefore detect more peaks. As final result, the user performing the analysis has less chances of making errors and can also analyse more spectra in the same amount of time.

A new software, with an optimised interface able to further enhance the productivity of the user, is developed and validated.

Contents

Contents	III
List of Tables	V
List of Figures	VI
Foreword	IX
Introduction	XI
1 Principles of Gamma Spectrometry	1
1.1 Radioactive Decays	1
1.2 Radiation Detectors	2
1.3 Interactions of Photons with Matter	3
1.4 Gamma Detector Components	6
1.5 Analysis of Gamma-Emitting Samples	10
1.5.1 Peak Locate Calculations	11
1.5.2 Peak Area Calculations	13
1.5.3 Background Subtraction and Efficiency Correction	14
1.5.4 Peak Identification and Nuclide Identification	15
1.5.5 Decay Chain Equilibrium Calculation	15
1.5.6 Reporting	15
1.6 Example of a Complete Analysis Sequence	16
2 Problems of the Traditional Routine	21
2.1 Missing Peaks	21
2.1.1 Peaks Closely Spaced	21
2.1.2 Peaks Between Peaks of Greater Magnitude	23
2.1.3 Peaks Lost in the Background Noise	25
2.1.4 Solution to the Problem	27
2.2 Non-Existing Peaks	27
2.3 Peak Identification Step	28
2.3.1 Solution to the Problem	30
2.4 Activity and Decay Chain Calculations	30

3	The Program	32
3.1	Main Guidelines and Program Structure	32
3.2	Main Window and Sample Properties	33
3.3	Peak Locate Step and Correction Step	35
3.4	Interactive Peak Fit	35
3.5	Peak Identification Step	37
3.6	Nuclide identification Step	42
3.7	Reporting Step	43
4	Validation of the Results	45
4.1	Low Activity Sample	45
4.1.1	Peak Locate Results	45
4.1.2	Peak Identification	49
4.1.3	Nuclide Identification	49
4.2	High Activity Sample	52
4.2.1	Peak Locate and Peak Identification	52
4.2.2	Nuclide Identification	53
5	Conclusions	55
A	Application code	57
B	Gamma Analysis User Guide	62
B.1	Overview	62
B.2	Installation	62
B.3	User Interface Elements Description	62
B.3.1	Main Window	62
B.3.2	Information Window	64
B.3.3	Peak Locate	64
B.3.4	Interactive Peak Fit Window	65
B.3.5	Peak Identification Window	65
B.3.6	Nuclide Identification Window	66
B.3.7	Report Window	66
B.3.8	Preferences Window	68
B.4	Usage Guide	71
	Bibliography	75

List of Tables

1.1	Example of an Unidentified Peaks results, with the sample information omitted.	19
1.2	Example of a Nuclide Activities results, with the sample information omitted.	20
2.1	Results of a complete analysis, grouped by decay chain, with the peaks in the 241 keV region not correctly identified.	23
2.2	Results of a complete analysis, grouped by decay chain, with the peaks in the 241 keV region correctly identified.	25
4.1	List of peaks (1-25) found in the first sample spectrum.	46
4.2	List of peaks (26-43) found in the first sample spectrum.	47
4.3	Peaks not identified by the old routine.	49
4.4	Peaks not identified by the new routine.	49
4.5	Unidentified peaks: details of peak nr. 5.	50
4.6	Unidentified peaks: details of peak nr. 6.	51
4.7	Comparison of the results of the nuclide identification and activity calculation step, executed with the old and the new routines on a low-activity sample.	52
4.8	Summary of the peak locate step on a high-activity sample.	52
4.9	Comparison of the results of the nuclide identification and activity calculation step, executed with the old and the new routines on a high-activity sample.	54

List of Figures

1.1	Polar plot of the Compton-scattering cross section for several energies.	4
1.2	Energy distribution of electrons from Compton scattering.	5
1.3	Linear attenuation coefficients in Germanium.	6
1.4	The detector, as seen from outside. Inside the lead shield is visible the copper shield.	7
1.5	Close-up of the detector.	7
1.6	Germanium detector assembly.	8
1.7	Efficiency calculated for a coaxial germanium detector.	9
1.8	A typical sample, ready for the measurement.	11
1.9	Two examples of a spectrum along with its second derivatives.	12
1.10	Peak locate parameters.	16
1.11	Area calculation parameters.	17
1.12	The spectrum as appears after peak locate and area calculations.	17
1.13	Two peaks not found by the automatic algorithm.	18
1.14	Example of a poor peak fitting.	19
1.15	The settings window for the <i>Nuclide Identification with Interference Correction</i> step.	19
2.1	Two peaks missed by the peak locate algorithm and their residuals.	22
2.2	The same two peaks of figure 2.1, now correctly marked.	22
2.3	A “non-existing” peak, useful only to improve the fitting.	23
2.4	The 233-245 keV region of the spectrum after <i>Unidentified 2nd Difference</i> peak locate and area calculation.	24
2.5	The 233-245 keV region of the spectrum, with the 241.14 keV peak added.	24
2.6	The peak centred on 84 keV, not recognised.	25
2.7	The same peak of figure 2.6, now correctly marked.	26
2.8	A missed peak in the high-energy region of the spectrum.	26
2.9	The same peak of figure 2.6, now correctly marked.	26
2.10	A peak left by the area calculation algorithm.	28
2.11	The region of figure 2.10, after the manual correction.	29
2.12	<i>Library (Simple)</i> parameters.	29
3.1	Software structure flow chart.	33
3.2	Efficiency correction step.	33
3.3	The main program window.	34

3.4	Sample properties window.	35
3.5	Background file mask in the global preferences.	36
3.6	Peak identification window.	37
3.7	Peak identification flow chart.	38
3.8	K_c value as function of nuclide half-life.	40
3.9	Preferences of the new peak identification step.	42
3.10	“Nuclide Identification” window with some test results.	43
3.11	Reports window.	44
4.1	Peak 42, as marked by the old routine.	47
4.2	Peak 41, marked correctly by the new routine.	48
4.3	Peak 41 and 43, marked correctly by the user.	48
B.1	Main software window.	63
B.2	Sample information window.	64
B.3	Interactive Peak Fit window.	65
B.4	Peak Identification window.	66
B.5	Nuclide Identification window.	67
B.6	Reports window.	67
B.7	Preferences window: Profiles.	68
B.8	Preferences window: Peak Locate.	69
B.9	Preferences window: Area Calculation/Peak Rejection.	69
B.10	Preferences window: Interactive Peak Fit.	69
B.11	Preferences window: Area Correction.	70
B.12	Preferences window: Peak Identification.	70
B.13	Preferences window: Nuclide Identification.	71
B.14	Preferences window: Reports.	71
B.15	Preferences window: Common Options.	72

Foreword

The field of NORM is gaining wider and wider recognition as a source of radiological risk to the general public and to the workers of numerous types of industry: the fertilizer, ceramic tile and oil industries to name but a few. Radon in particular is attracting growing attention as a potential health hazard both on the workplace and at home: it has been estimated that Italians are exposed yearly on the average to doses in the few millisievert from radon inhalation. Nor can the many other NORM and TENORM sources of radiological risk be disposed of lightheartedly.

In recent times, a collaboration has developed between our laboratory of Montecucolino, University of Bologna, and the Radiation Protection body in Forschungszentrum Karlsruhe that, within his activities, runs a laboratory very active in the measurement and qualification of NORM. In the framework of this burgeoning collaboration my Colleague Professor Manfred Urban, head of radiation protection at the Forschungszentrum, offered to host a student from our University for several months, to do his Master Thesis work in the wake of the ongoing project of automation of their gamma spectrometry laboratory. My student Olaf Marzocchi then spent the second half of 2006 working actively on this project, with excellent result as I could see and as was confirmed to me by Professor Urban.

The whole experience proved to be a success story, and I would like to take this opportunity to thank Professor Urban and all his team for being so hospitable to my student and for guiding him through this challenging learning experience.

And of course, I wish to congratulate Olaf Marzocchi on completing his demanding task with most excellent results, holding high the name of his Alma Mater.

PROF. ING. DOMIZIANO MOSTACCI

The radiation protection community has been aware of the contribution from natural occurring radionuclides to the radiation exposure of man at least since the European Council Directive 96/29 Euratom, laying down the basic safety standards for the health protection of workers and general public against the dangers arising from ionizing radiation. Since then, also, materials sometimes containing naturally high concentrations of radionuclides of the uranium and thorium decay chains, used for example in industrial processes, have to be considered as sources of exposure to the workers and to the public. In the treatment and storage of the waste originated by these industrial processes, the level of contamination with natural radionuclides must also be taken into account. From this duties arises the need of measurements of the radioactivity content of these materials.

The Forschungszentrum Karlsruhe is running laboratories in its Central Safety Department, where these measurements of natural radionuclides concentration in different sample matrices can be performed by gamma spectrometry and also, after radiochemical sample preparation, by alpha spectrometry.

The Central Safety Department (HS) is in general responsible for licensing, supervising, monitoring and, to some extent, executing measures of radiation protection, industrial health and safety as well as physical protection and security at and for the institutes and departments of the Forschungszentrum Karlsruhe GmbH (Karlsruhe Research Centre). It is also responsible for monitoring liquid effluents and the environment of all facilities and nuclear installations on the premises of the Centre.

Preserving at least the current knowledge in Radiation Protection is considered as essential for the future. Therefore junior scientists, young engineers, PhD- and Masters students do research in the Central Safety Department in the field of biokinetic modelling, internal and external dosimetry, radiochemistry, alpha- and gamma-spectrometry and measurement instrumentation in general. This happens in close cooperation with the Karlsruhe Scientific University, with the Karlsruhe University of Cooperative Education and, internationally, with several European universities such as the University of Bologna, Italy.

The *HS – Physical Measurements Laboratory* determines the activity concentrations in the waste water at the installations, and decides whether these liquid effluents have to be decontaminated or can be passed directly to the sewage treatment plant. It also establishes balances of the activity discharges. Additionally, the Group is responsible for carrying out all the spectrometric nuclide assays, including the very time consuming gamma spectrometry of NORM materials. In order to improve and to speed up this type of gamma spectrometric analyses, we decided to invite a student to do his master thesis in this field.

In the discussion of this project with my Italian Colleague Professor Domiziano Mostacci from the University of Bologna, we agreed to offer the project “Automation of a gamma spectrometric analysis method for natural occurring radionuclides in different materials (NORM)” as Master Thesis to Mr. Olaf Marzocchi, master student at the University of Bologna. As can be seen in the following paper, he managed his task in an excellent manner.

I would like to express my congratulation to Olaf Marzocchi for his work, to thank my Colleague Professor Domiziano Mostacci for his very fruitful cooperation and Dipl.-Ing. Christoph Wilhelm and Dipl.-Ing. Susanne Kaminski for accompanying and supervising the thesis.

PROF. DR.-ING. HABIL MANFRED URBAN
Karlsruhe Institute of Technology KIT
(Forschungszentrum Karlsruhe and Scientific University of Karlsruhe)

Introduction

There are many techniques useful to analyze a sample and know its composition, some of them are chemical analyses, neutron activation and gamma analysis, where each one has advantages and disadvantages. Chemical analyses involve chemical reactions, so they have the complete destruction of the sample as side effect, but can, theoretically, find each atomic species contained in the sample; neutron activation is able to find many atomic species too, but samples that undergo this type of analysis require a long and expensive preparation step: the sample must be first irradiated with nucleons that cause nuclear reactions and create radioactive nuclides, then these new radioactive nuclei are detected by an appropriate instrumentation. Gamma analysis has a big disadvantage too, as general purpose analysis method: it is able to detect only gamma rays emitted by radioactive nuclides. It has, however, a big advantage: it is completely non-destructive and does not require any sample modification. The measurement in gamma analysis is done by counting and then analyzing gamma rays emitted during the natural decays of the nuclides; from a more practical point of view, the user simply has to put the sample inside the device and start the counting. This process takes from some minutes to many hours, depending on the sample activity level and on the required accuracy of the measurement, and produces a file where informations about the collected gamma rays are stored. The analysis of these data, done with different softwares, is the main topic of this work.

In the HS-ÜM measurement laboratory of the *Forschungszentrum Karlsruhe* samples from both the research center itself and external customers are analyzed routinely. Examples are air filters from the research center or soil, wastes and other environmental samples from external customers. Many of the materials analyzed fall in the *NORM* category, “naturally occurring radioactive materials”: they are usually expected to contain only or almost only natural radioactive nuclides, artificial nuclides are unusual.

The specific goal of this project is to speed up the analysis of the results of the gamma counting process for *NORM* samples. The traditional routine takes up to two hours to complete and requires a great amount of user interaction, while the new one must take less than 30 minutes, while achieving the same accuracy of the old one. The need for a new, more efficient, routine comes from the expected increase in the number of samples the HS lab will be asked to analyze, task too long to be completed with the current staff.

This work is divided in three parts:

Principles of Gamma Spectrometry describes the principle of gamma analysis, the elements of a gamma detector and the routines used during the analysis;

Problems of the Traditional Routine describes the limitations found in the traditional

procedure and the steps that take most of the time during the analysis;

The Program describes how the problems listed in the previous chapter have been solved, with a description of the algorithms chosen and images of the new graphical user interface;

Validation of the Results compares the results of the traditional routine with the results obtained with the new software, highlighting the improvements achieved and the issues left.

Chapter 1

Principles of Gamma Spectrometry

1.1 Radioactive Decays

Atomic nuclei are governed by three forces: the *strong nuclear force*, the *electrostatic repulsion* and the *weak nuclear force*. The result of the interaction of these forces is a complex potential field, where every constituent of the nucleus (*nucleon*) has a specific energy, depending on its relative position.

This force field is quantised, so nucleons cannot change position and energy state freely. Instead, they require, just like the electrons in the orbitals, a specific quantum of energy. This activation energy can either come from an external energy source or from the statistical fluctuations of the void, that can supply finite amounts of energy for a very small time, according to the Heisenberg *Uncertainty principle* (or *Indeterminacy principle* as called by Niels Bohr). This principle states that

$$\Delta E \Delta t \geq \frac{\hbar}{2} \quad (1.1)$$

The nucleus undergoes nuclear decay when two specific conditions are satisfied: the proper quantum energy is supplied to a nucleon or the nucleon is in an unstable state, so that every change in its state lead to a lower energy rearrangement of the nucleus. The final result of the decay is the emission of at least one particle and an energy release.

Depending on the rearrangement and on the particle released, the decay takes different names. The most common radioactive decays in natural radionuclides are alpha decay and beta decay. Alpha decays are characterised by the release of a helium nucleus (${}^4_2\text{He}$), while beta decays release an electron. Regardless of the decay type, the new nucleus is often left in an excited state, so that it then emits gamma rays, carrying away the excess energy.

Alpha particles, beta particles and gamma rays interact very differently with matter. Alpha particles, as a result of the strong electrostatic charge and of the heavy mass, stop in just a few milligrams per square centimetre of matter, so that even a paper sheet is enough to stop them and their detection is therefore difficult. Beta particles, weighting eight thousand time less and with half the charge of alpha particles (absolute value), stop in about one gram per square centimetre of matter, so that an aluminium plate is required to shield them effectively.

Gamma rays, being photons of high energy (mostly more than about 50 keV) and

without electrostatic charge, are the most difficult particle to stop. A good shielding therefore requires a wide thickness of heavy material: depending on the gamma rays energy and on the initial beam intensity, the shield can be made of centimetres of lead or decimetres of concrete.

Another difference between both alpha and beta particles and gamma rays is the way they lose energy. Gamma rays, being energy quanta, can either travel in space or be completely absorbed, thus transferring all the energy it carried to the interacting particle, while alpha and beta particles collide with atoms and lose energy progressively.

1.2 Radiation Detectors

The different properties of the particles emitted in nuclear decays greatly affect the design of detectors, but all of them share the same principle: alpha, beta and gamma ionise the matter they interact with, the instrument detects this ionisation and produces a signal the operator is able to understand. With this principle in mind, I will describe the three most common detector principles: gas-filled detectors, scintillation detectors, semiconductor detectors.

The basic principle of *gas-filled* detectors is very simple: a sealed chamber filled with gas has a potential applied to it, so when the gas is ionised by the alpha-, beta- or gamma-particle, the ionised atoms travel toward one of the two electrodes, thus producing an electric pulse. If the instrument operates under a voltage low enough to avoid a cascade effect, the result is a proportional counter (used mostly with alpha particles), if the potential applied is able to generate further ionisation after the first event, it operates as simple detector (*Geiger-Mueller detector*).

The sample can be outside or inside the chamber. In the former case, the chamber window must be thin enough to let the radiation enter, in the latter case, common with proportional counters, the instrument is able to collect more particles and to better measure their energies.

Liquid scintillation works by converting the particle energy into photons, then detected by optical sensors.

This type of instruments is made of several components: the first one is the sample itself, which releases energy to a first medium, chosen for its absorption properties. The energy is then transferred to a second medium, able to efficiently convert it into photons. These photons usually have energies outside the photo-detectors range, so they must be frequency shifted with a third medium. Photons, finally, are converted into photo-electrons and multiplied by photo-multipliers, then detected by sensors.

When alpha- or beta-particles are involved, the three media used and the sample must be all liquid and miscible, so the sample must be completely destroyed and a solvent is required too. When gamma-rays are involved, the particle energy conversion into ionisation can be done by crystal (i.e. a Sodium Iodide crystal), so the sample is not liquified and destroyed. Sodium Iodide detectors are often used as portable gamma rays detectors.

Semiconductor detectors work by using a semiconductor as diode, with an high electric potential applied to the surrounding electrodes. The most common materials used are silicon, germanium or highly pure germanium crystals (*HPGe*, used for

gamma rays detection). The ionising particles release their energy and create electron-hole pairs, then detected as an electric current pulse. The advantage of this technique is the low energy required to create such electron-hole pairs, so higher resolution spectrometry is possible. the downside is the low detection efficiency.

Semiconductor detectors can be built in different shapes, depending on the particle to detect. Coaxial detectors, with one electrode near the axis and the other on the surface, are used to capture gamma rays, because this geometry allows for bigger detectors and higher collecting efficiencies. Another possibility is to put the active region on the surface, to collect better particles with low free-mean-paths, such as alpha- and beta-particles.

1.3 Interactions of Photons with Matter

Gamma rays can interact with the matter in three different ways: *photoelectric absorption*, *Compton scattering* and *pair production* [1].

In the *photoelectric effect*, most common at low energies, a photon directly releases the energy to an electron. Gamma rays usually interact with the two innermost shells: the K-shell or the L-shell. After the interaction, the electron is free from atomic bounds and will usually interact with the surrounding matter, ionising other atoms and releasing all its energy. The energy of the electron after the interaction with the gamma ray is

$$E_e = \hbar\nu - E_b \quad (1.2)$$

Where:

E_e is the final electron energy;

\hbar is the Planck's constant;

ν is the frequency;

E_b is the binding energy of the electron in its original atomic shell.

The K-shell of Germanium has a binding energy of 11.103 keV [3], its L-shell has, depending on the electronic spin, 1.4146 keV, 1.248 keV, 1.217 keV [4].

The released electron will then interact with the surrounding nuclei and ionise them, thus creating a cascade effect until each electron no longer has enough energy to ionise other atoms. The final result is a current pulse, with amplitude related to the energy of the incident gamma ray.

The cross section of this interaction is given by

$$\tau = \text{const} \cdot Z^{4.5} \cdot E_\gamma^{-3} \quad (1.3)$$

Where Z is the atomic number and E is the energy of the incident photon. This effect is important for high Z atomic species but not for high energy rays.

The second possible interaction, the *Compton scattering*, takes places at medium energies and transfers to a loosely bound atom electron only part of the energy of the incident photon. The equation describing the phenomenon is

$$E_\gamma = E_e + E'_\gamma \quad (1.4)$$

that can be rewritten as

$$E_e = E_\gamma [1 - P(E_\gamma, \theta)] \quad (1.5)$$

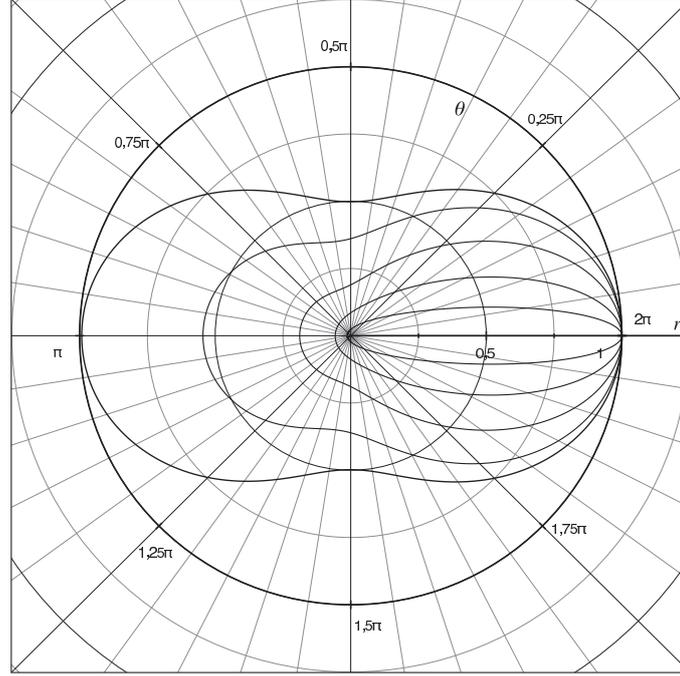


Figure 1.1: Polar plot of the Compton-scattering cross section for several energies.

Where:

E_γ is the incident gamma energy;

E'_γ is the scattered gamma ray energy;

θ is the angle between the original and the final photon travel direction;

m_0 is the rest mass of the electron;

c is the speed of light

$P(E_\gamma, \theta)$ is the ratio of photon energy after and before the collision:

$$P(E_\gamma, \theta) = \frac{1}{1 + \left(\frac{E_\gamma}{m_0 c} \cdot \frac{1 + \cos(\theta)}{511} \right)} \quad (1.6)$$

The energy released to the electron is therefore maximum when the photon is scattered backward, but this latter always retains at least

$$E'_\gamma = \frac{E_\gamma}{1 + 2 \frac{E_\gamma}{m_0 c}} \quad (1.7)$$

The angular probability distribution is given by the Klein-Nishima formula:

$$\frac{d\sigma}{d\Omega} = 0.5 r_e^2 \left[P(E_\gamma, \theta) - P(E_\gamma, \theta)^2 \cdot \sin^2(\theta) + P(E_\gamma, \theta)^3 \right] \quad (1.8)$$

where r_e is the classic electron radius. The function is plotted in figure 1.1.

As result, the incident gamma ray energy can be spread in a significant volume of the detector. The resulting spectrum is shown in figure 1.2. The cross section of this interaction is given by the formula

$$\sigma = \text{const} \cdot Z \cdot E^{-1} \quad (1.9)$$

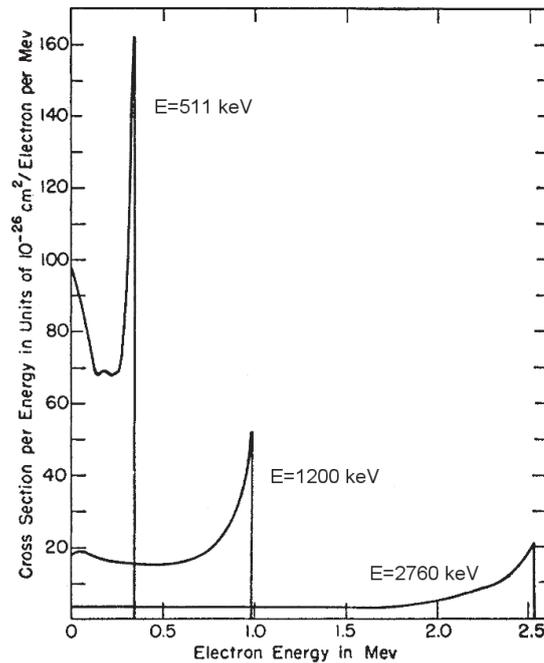


Figure 1.2: Energy distribution of electrons from Compton scattering for primary photons of 511, 1200 and 2760 keV (Modification of figure from Davisson and Evans 1952 [7]).

The third possible interaction with the matter is *pair production*. This effect consists in the creation of an electron-positron pair and can be observed only for gamma rays with at least the sum of the energies of the two particles at rest: 1.022 MeV. The excess of energy is shared as kinetic energy between the two particles, which will interact with the surrounding matter to create more ionisation and free electrons. The positron, however, will annihilate immediately with an electron, to create two gamma rays of about 511 keV each. The energy of each photon will be lower than the rest mass of the electron because some energy and momentum are required to free the interacting electron from the originating atom. With pair production, as in the Compton effect, the original gamma energy is not released in the location of the first interaction, but is carried away by the gamma generated during the positron annihilation.

The pair production effect is the most important at high gamma energies and in high-Z materials, since its cross section is proportional to Z^2 . The graph of the total cross section is shown in figure 1.3.

A phenomenon associated with the pair production is “*escape peak*” detection. “Escape peaks” are groups of counts not belonging to any known nuclide, with an energy shift of 511 keV or 1022 keV compared to a higher-energy known peak. The cause of this phenomenon is the loss of one or both 511 keV gamma rays generated during a positron-electron annihilation inside a crystal of finite size. In the case of the loss of only one of the quanta, the counting system will detect a peak with 511 keV less energy than it should, while if both are lost, a peak shifted of 1022 keV towards lower energies will be detected. The ratio of single-to-double peaks counts depends on many factors, among them detector type, detector shape, detector size. Such peaks

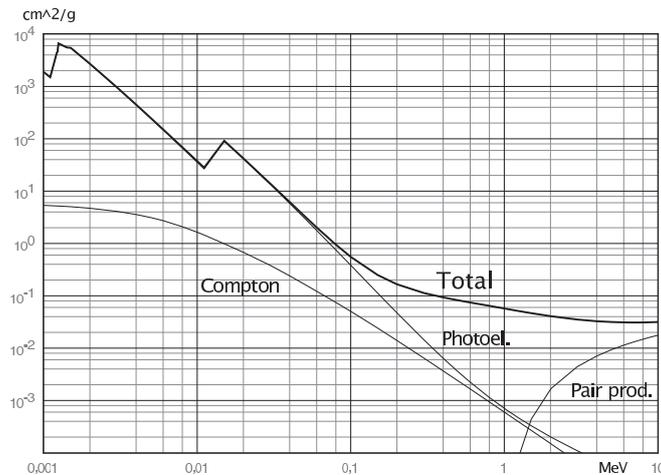


Figure 1.3: Linear attenuation coefficients in Germanium for photoelectric effect, Compton scattering and pair production (from [5]).

are not usually taken into account during activity calculations, but must be recognised as such during sample analyses.

The range of possible interactions with the surroundings of the escaping gamma is quite broad, but the probability of them is low so they are not actually taken into account.

1.4 Gamma Detector Components

A gamma detector is made of several components:

active element is made of a Germanium crystal, with about 10^{10} impurities per cubic centimetre in HPGe versions. Silicon crystals used in microelectronics have three orders of magnitude or more impurities per cubic centimetre. An external view of a detector is shown in figure 1.4.

lead shield shields the active element of the detector from external sources of radiation to lower the background counts as much as possible (figure 1.4 and 1.5).

electronics is used to amplify the first current pulse originating from the crystal after each gamma interaction. Shown in figure 1.6.

acquisition system converts the analog amplified pulses into digital signals, in order to send the information to a computer and to count pulses of different energies separately.

The germanium crystal is characterised by a conduction band only 0.7 eV higher than the unexcited state. This leads to thermal electric noise, so the crystal must be cooled down at very low temperatures in order to reduce this background unwanted signal. The most common way to reach this goal is to use liquid nitrogen at constant 77 K temperature (boiling temperature), even if germanium crystals does not always need such low temperature. The preamplifier is cooled down as well, to reduce the



Figure 1.4: The detector as seen from outside, the lead shield is painted blue. Inside the lead shield is visible the orange copper shield, used to absorb the low energy x-rays originated in the sample.

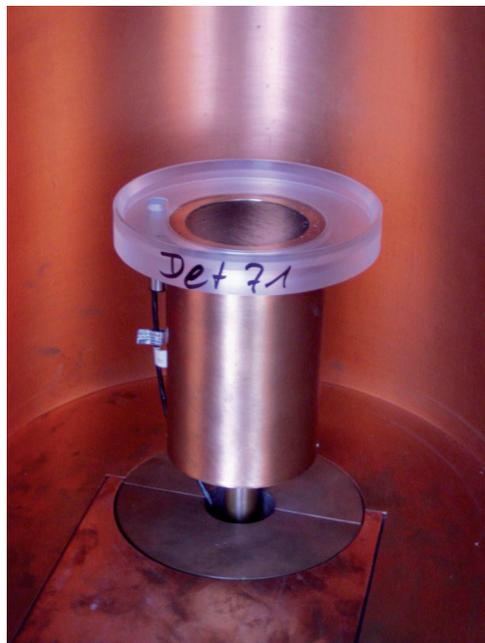


Figure 1.5: Close-up of the detector.

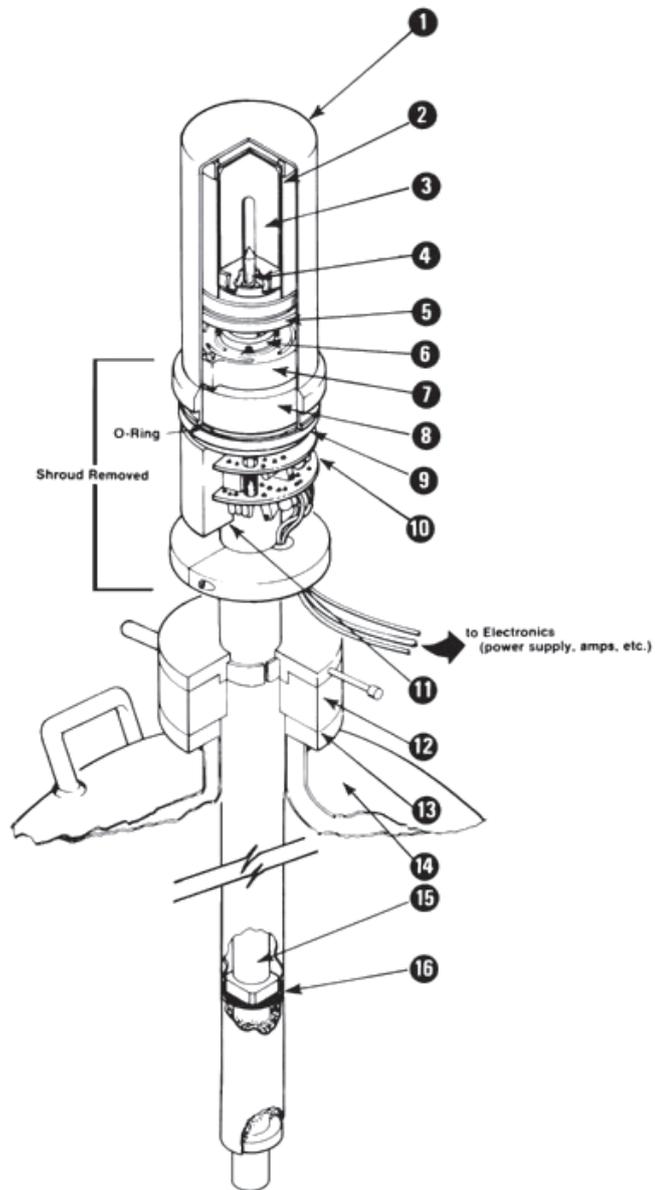


Figure 1.6: Germanium detector assembly [6]. Legend: 1 - End-cap; 2 - Detector mount cup; 3 - Germanium detector element; 4 - Pedestal nut; 5 - Pedestal; 6 - Cooling rod clamp; 7 - Thermal insulator; 8 - Shield; 9 - Cryostat flange; 10 - Preamplifier; 11 - HV filter; 12 - Fill collar; 13 - Dewar flange; 14 - Dewar; 15 - Cooling rod; 16 - Cryosorption material.

noise.

The liquid nitrogen is kept in a dewar under the detector, with a copper “cold finger” extending from the crystal to the bottom of the dewar (figure 1.6).

The results of the measurements depend on all the components used, but the response of analog electronic components cannot be known in advance, so the whole instrumentation must be calibrated periodically. Many calibrations are possible, but two of them are routinely checked: efficiency calibration and energy calibration.

Energy calibration is required to match the current pulses with the original gamma energy. The electronics components can output different values depending on many factors such as temperature and other environmental conditions, but the problem in most situations is only an energy shift, so the correction is straightforward. The recalibration is done by putting a source with peaks spread over the whole spectrum and then counting the gamma. Once the measurement is completed, the best linear interpolation is chosen as new energy calibration and the two variables used to define the interpolating line are saved in a calibration file. A calibration source with only two peaks can be enough for rough approximations. Newer instruments with less analog and more digital components suffer less from this energy shift problem, so less recalibrations are required.

The other calibration step required prior to any sample counting is the efficiency calibration. This step associates the number of counts in each “channel” (energy bin in the final spectrum) with the original number of gamma rays emitted within the corresponding energy bin and, consequently, with the sample activity. This correction is required because the efficiency of a gamma detector depends on the photon energy: a higher-energy photon has a lower interaction probability, so the collecting efficiency is lower. An example efficiency plot is given in figure 1.7.

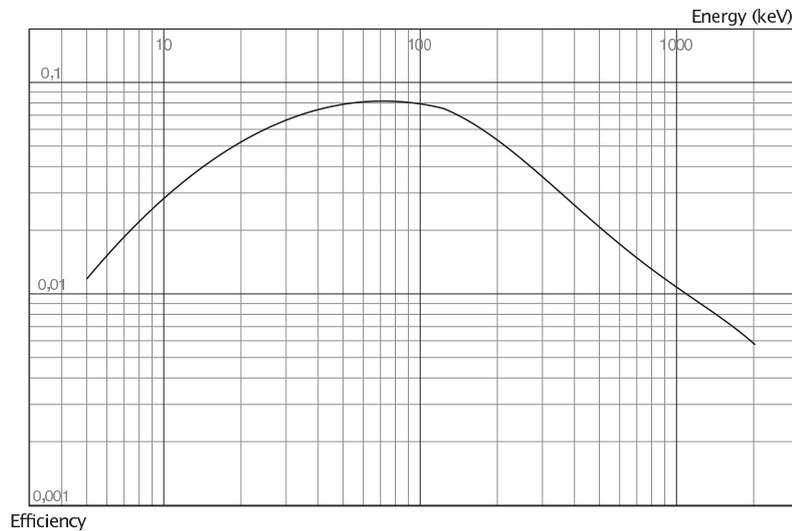


Figure 1.7: Efficiency calculated for a coaxial germanium detector.

Recently, thanks to the increase in computing power, a mathematical calculation of the detector efficiency is possible with Monte Carlo simulations. They simulate the possible interactions of gamma rays emitted from the whole sample and output

the photon-detector interaction probability for each energy bin. The advantage of this new method is the accuracy with generic shapes and the speed: each experimental efficiency calibration requires a significant amount of time and is correct only with the specific test sample, while a mathematical calibration can be done for the real sample geometry, density and composition. Experimental calibrations are then needed only to check the coherency of the mathematical calibration with experimental results.

1.5 Analysis of Gamma-Emitting Samples: Steps and Algorithms Used

The complete process of a sample measurement requires the following steps:

sample preparation the sample must be put in a standard container with a standard shape, able to fit comfortably in the detector (figure 1.8);

mathematical calibration required when the sample has a density too much different from water, used for the traditional efficiency calibration;

counting the counting time is set in the software, the counting procedure is automatic;

peak locate the spectrum only lists counts per each bin, peaks must be marked for the subsequent analysis;

area calculation and non-existing peak removal all the peaks marked during the previous step are analysed and their area calculated;

background subtraction to isolate the contribution of the environment from the contribution of the sample itself;

efficiency correction the calculated area must be corrected for the detector efficiency to know the original peak activity;

peak identification each peak must be assigned to a nuclide, to know if there are peaks from artificial nuclides;

nuclide identification peaks correctly assigned to nuclides must be taken into account and their area weighted to calculate the nuclide activities;

decay chain equilibrium calculation activities of nuclides of the same decay chain could be in equilibrium if the sample was not subject to manipulations;

reporting to summarise the result obtained.

This work affected the steps from the peak locate to the last step, so these steps will be analysed in more detail in the following sections.



Figure 1.8: A typical sample, ready for the measurement.

1.5.1 Peak Locate Calculations

A peak is defined as an increase in the spectrum of the number of counts, compared to the local background. The locate algorithm must be able to correctly identify peaks even when they are masked by stochastic noise, that appears as peaks with small amplitude and width.

The software *Gamma analysis* of the *GENIE-2K* software suite offers four different automatic peak locate algorithms [8]: *Library (Simple)*, *Library (Gamma-M)*, *Unidentified 2nd Difference*, *VMS Standard Peak Search*.

Library (Simple) is the simplest algorithm: given an external nuclide library¹ where energy lines² and nuclides of interest are listed, the algorithm marks a peak in the spectrum for each energy line found in the library. The next area calculation algorithm will then discard non-existing peaks (peaks with the value of the area equal or less than zero).

This algorithm has two big limitations, so it is useful only in specific situations. The first issue is the energy calibration, that must be accurate and similar from sample to sample: if it changes, the energy lines will not match the peaks of the spectrum anymore. The second limitation is the nuclide library: the algorithm does not ‘look’ at the spectrum, but uses only the library. A peak not listed will never be found.

The *Library (Gamma-M)* algorithm works in four different steps: peak erosion,

¹A nuclide library is a database where gamma ray energies are associated to radioactive nuclides. Nuclides are characterised by their decay time and, optionally, mother nuclide, while gamma ray energies (also called energy lines) are characterised by per-decay emission probability, energy of the gamma ray and other data useful to the *GENIE-2K* software.

²From now on, the term *energy line* will be used when speaking of theoretical data contained in a nuclide library, while *energy peak* will refer to the actual increase of counts found in the spectrum. As consequence, we could have missing peaks, when the nuclide library lists gamma ray energies not matched by any increase in counts in the spectrum, and also missing energy lines, when we have peaks not matched by any known gamma energy.

peak fitting, unknown peak locate, peak rejection and area calculation. *Peak erosion* is the first step and is used to calculate the background level. It works by ‘eroding’ the spectrum, until all the peaks have been smoothed out and fall below the continuum background. The background is then subtracted from the spectrum and the resulting peaks are marked with boundary limits.

The *Peak fitting* step tries to find the best Gaussian fitting of each peak listed in an external library by using a least squares algorithm. This algorithm does not use the low-tail calibration, that models the shape of the lower-energy peak tail, but is able, if requested, to try to detect shifts of the peak from the energy line listed in the library.

The *unknown peak locate* is an optional step, used to detect peaks not known in advance. This algorithm does not try to fit them with a Gaussian shape, but works as a digital filter: it analyses a group of channels, recalculates the background to improve the final area estimation and then averages the number of counts. If the final result is greater than a defined threshold, the peak is marked.

The *peak rejection* algorithm works in two steps. The first one depends on the peak type: library peaks are rejected when the FWHM distance is greater than a predefined value and merged when it is smaller, unknown peaks are rejected if they are located within the energy tolerance of a library peak. The presence of the library peak in the list of peaks found is not checked. The second step checks the peaks for MDA³: if the peak falls below the area or height threshold, the peak is rejected.

The *Unidentified 2nd difference* algorithm works by ‘looking’ directly at the spectrum: each peak is characterised by a second derivative with a negative and two positive peaks. This behaviour is shown in figure 1.9, where an example with one peak is shown on the left and an example with two overlapping peaks (a “multiplet”) is shown on the right.

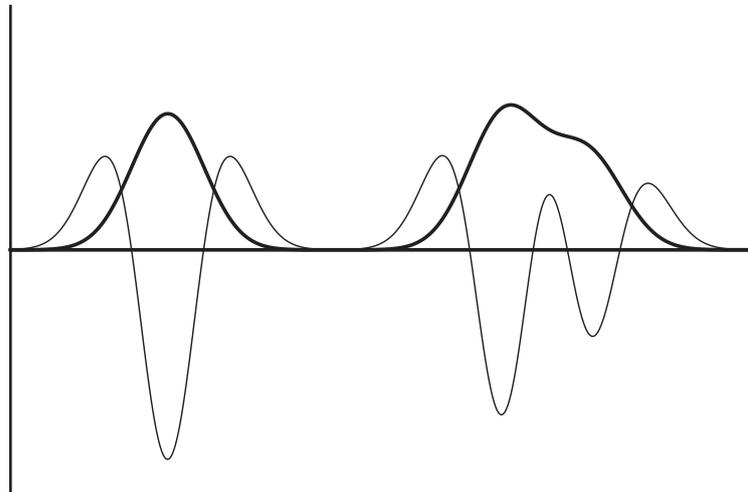


Figure 1.9: Two examples of a spectrum (thick line) along with its second derivatives (thin lines). The example on the right is made by placing two overlapping Gaussians like the first one. The derivatives are not re-scaled.

This algorithm first calculates the value of the second difference and of the standard

³The acronym stands for *Minimum Detectable Activity* and it is an estimate of the smallest peak that can be distinguished from the background.

deviation with following two formulas, respectively:

$$dd_i = \sum_{j=-k}^{j=+k} c_j y_{i+j} \quad (1.10)$$

$$sd_i = \sqrt{\sum_{j=-k}^{j=+k} c_j^2 (y_{i+j})} \quad (1.11)$$

where y are the counts of each channel, i is the channel we are analysing and c_j is a weight coefficient calculated as following:

$$c_j = \frac{100 [j^2 - (c_w)^2]}{(c_w)^2} \cdot \exp\left(-\frac{j^2}{2(c_w)^2}\right) \quad (1.12)$$

In this equation, c_w is the calibrated expected Gaussian width in units of channels: $c_w = \text{FWHM}/2.355$. The FWHM value is taken from the detector peak shape calibrations.

The value of k , used in equations 1.10 and 1.11, is not known in advance: the algorithm starts with $c_0 = -100$ and the set of coefficients is terminated at k , when the next coefficient would be less than 1. The second k coefficient is then adjusted so that the sum of the coefficients is zero. The set of coefficients are recalculated every 100 channels.

Each peak centroid is then defined by as

$$\text{centroid} = \frac{\sum_i i \cdot ss_i}{\sum_i ss_i} \quad (1.13)$$

Where i is the channel number and

$$ss_i = \frac{dd_i}{sd_i} \quad (1.14)$$

is the significance value. The centroid is calculated only for channels where the significance value is negative, so it is the weighted average of these channels. Peaks with a significance value below in absolute value than a predefined parameter are discarded.

This algorithm saves, along with the peak energy and peak centroid, a peak energy uncertainty, to account for the standard deviation calculated during the fitting.

The *VMS Standard Peak Search* is an algorithm that locates the peaks and calculates the area in a single step and is an alternative to the *Unidentified 2nd difference* peak locate algorithm followed by a *Sum Non-Linear Least Squares Fit*. More details can be found in the documentation [8].

1.5.2 Peak Area Calculations

The *GENIE-2K* software suite has two area calculation algorithms: *Library (Gamma-M)* and *Sum/Non-Linear Least Square Fit*.

The first one uses the peak erosion technique, described in the previous section, to calculate the background and then calculates the area with a least square fit algorithm.

The *Sum/Non-Linear Least Square Fit* algorithm, in addition, calculates the peak region limits and merges single peaks (“singlets”) into group of peaks (“multiplets”) if they are close together, a required step after the *Library (Simple)* and *Unidentified 2nd difference* algorithms, since they do not mark peak boundaries.

The algorithm checks the peaks centroids from the smallest to the largest. Each consecutive peak is compared to its neighbour to the right and checked for the following condition:

$$C_{i+1} - C_i < n_w \cdot W \quad (1.15)$$

Where:

C_{i+1} is the centroid of peak $i + 1$ (in units of channels);

C_i is the centroid of peak i (in units of channels);

w is the expected FWHM at peak i (in units of channels);

n_w is the user selectable separation parameter.

If it is satisfied, the peaks are merged into a multiplet and the last peak of it will be compared to the next peak.

To define the left boundary, the algorithm first calculates the average of 5 channels (or the expected peak width, rounded to the nearest odd integer), going backwards and starting from 0.8 FWHM before the peak centroid. The left boundary is then set as the channel where the calculated average reaches a minimum. The same algorithm is applied on the right side of the peak or multiplet. The number of channels used to determine the background can be either fixed and set by the user or variable, depending on the expected peak width at the centre of the peak region.

The area calculation algorithm depends on the peak type. If it is a singlet, the area is calculated as sum of the channels counts minus the background, if it is a peak within a multiplet or a singlet with forced Gaussian fitting, the area is the result of the peak fitting with a Gaussian curve or a series of Gaussian curves. The background estimation can be either *linear* or *step*, more useful when there are steep variations of the background level. More details and the complete algorithm description are found in [8].

1.5.3 Background Subtraction and Efficiency Correction

The peak areas calculated in the previous step can be corrected by environmental background subtraction or by peak reference. The environmental background subtraction algorithm uses background peaks activity data, stored in an external file, to correct the peaks areas found in the spectrum, while the peak reference correction uses one peak to normalise all the other peak areas. In the HS-ÜM laboratory only background subtraction is done.

The efficiency correction works by calculating the local efficiency based on data previously saved. The models available are Dual, Linear, Empirical, Average and Tracer. For details on the algorithms, refer to [8], only the Dual model will be described here, since it is the most used for the samples analysed in the HS-ÜM laboratory.

The efficiency using the Dual curve is calculated as

$$\varepsilon = \exp \sum_{i=0}^n b_i \cdot (\ln E)^i \quad (1.16)$$

where b_i are the parameters of the calibration curve, as saved during the calibration step.

The variance of the efficiency σ_ε^2 is calculated as

$$\sigma_\varepsilon^2 = \varepsilon^2 \sum_n \sum_m (\ln E)^{n-1} (\ln E)^{m-1} (M^{-1})_{n,m} \quad (1.17)$$

where $(M^{-1})_{n,m}$ is the inverse of the matrix resulting from the least squares determination of the efficiency calibration coefficients b_n .

1.5.4 Peak Identification and Nuclide Identification

The nuclide identification algorithm takes two inputs (the list of peaks found and a nuclide library) and outputs a list of identified nuclides, each one with its activity and activity uncertainty. Peaks used for the identification are marked with the name of the nuclide, the other peaks are marked with a flag. For a complete description of the algorithm refer to [8].

This algorithm is executed two times in a gamma analysis routine. In the first time, the goal is to match as many peaks as possible and leave as unidentified only the peaks from artificial sources, so the library used contains all the known energy lines of natural radionuclides. Once the user knows which peaks do not belong to natural sources, he executes the nuclide identification algorithm again, with a smaller library. Such library, with only the most important energy lines per each nuclide, is able to calculate the activities better, since energy lines with a low emission probability would only increase the uncertainty of the final calculation because the algorithm uses all the energy lines and their branching ratios while estimating the activities.

The user can also execute an MDA calculation to know the minimum detectable activity and the limit of detection of each nuclide, as specified in the ISO standards and in the German regulations.

1.5.5 Decay Chain Equilibrium Calculation

In this step the user checks whether the activities of all the nuclides of a specific decay chain are in equilibrium or not. The step is done by comparing the nuclides activities, normalised by their abundance. An example could be Thallium-208 in the Thorium-232 decay chain: each Thorium Becquerel leads to 0.362 Becquerels of Thallium-208, so its activity must be divided by this value.

1.5.6 Reporting

Each gamma analysis requires four different reports:

peak search used to list all the peaks found in the spectrum, it lists peak energy, width, original area, background area subtracted, final area, uncertainty in area calculation;

unidentified peaks used to list peaks not known to the NORM library (artificial radionuclides, unknown peaks, escape peaks), with energy, area and uncertainty, peak width;

nuclide activities NORM nuclides found in the spectrum are listed here, with activity and uncertainty, minimum detectable activity and limit of detection of the instrument;

customer report this report contains the global sample activity and details about the activity of each nuclide found.

The first three reports are executed from inside the *Gamma Acquisition & Analysis* software from Canberra, by selecting manually the report to print. This process requires three steps and each time also creates a text file with the report just printed. The file is overwritten each time.

The last report, addressed to the customer, requires the text file generated by the third printed report, from which a Microsoft Excel macro reads the values. The values are automatically checked for completeness (all the natural radionuclides of interest must be listed) and the final report is then created.

1.6 Example of a Complete Analysis Sequence

In this section I will describe a complete analysis sequence, with the software output and the parameters used.

The sample, put in the container, is shown in figure 1.8 and the detector used for the measurement is shown in figure 1.4 and figure 1.5.

The sample data must be first saved inside a centralised database: name, source, weight, rough composition. After this step, if the density of the sample is significantly different from the standard water used for the routine calibrations, the user must execute a mathematical calibration with the Canberra software to produce a calibration file, specific for the sample density and composition.

After the counting is completed, the first step of the manual analysis is the peak locate step, shown in figure 1.10, followed by a peak area calculation (figure 1.11). After these two steps, the spectrum shows some single peaks and some multiplets (peaks closely spaced, shown in figure 1.12 in the zoom box on the top with a cyan colour).

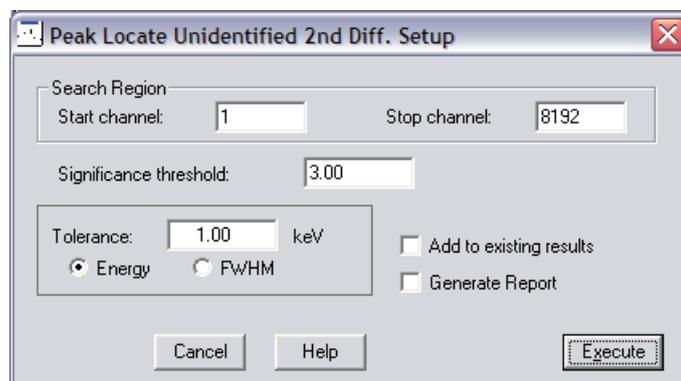


Figure 1.10: Peak locate parameters.

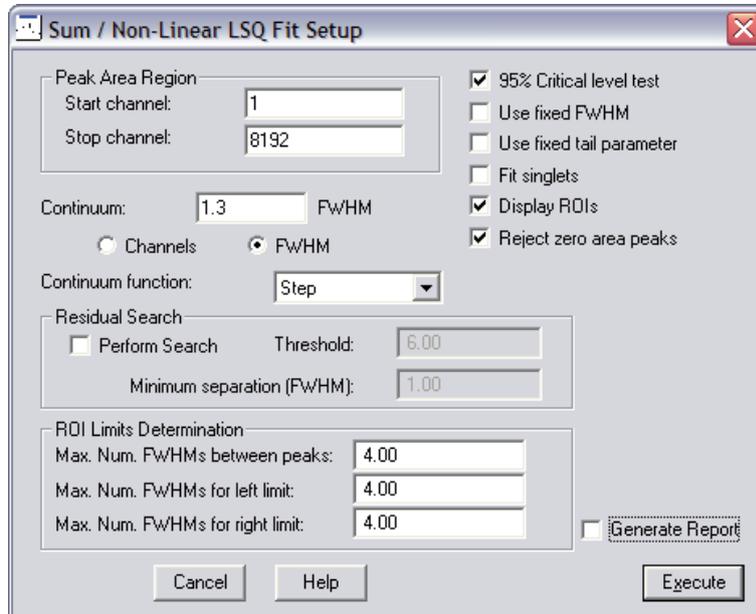


Figure 1.11: Area calculation parameters.

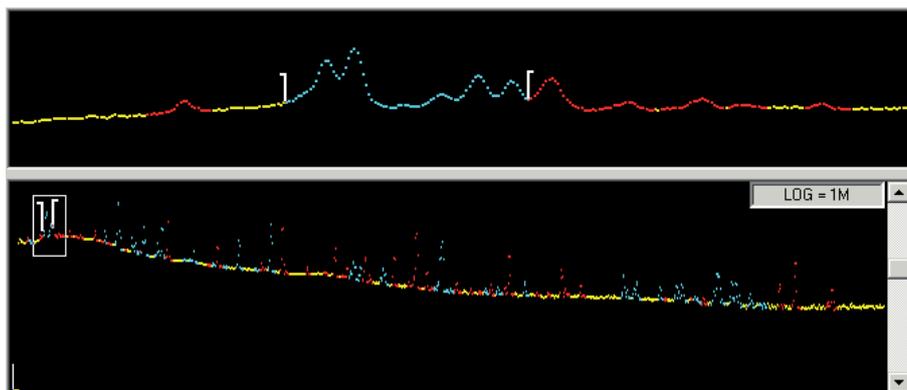


Figure 1.12: The spectrum as appears after peak locate and area calculations. Single peaks are marked with red colour, multiplets with cyan colour.

The user has now to check the spectrum manually, peak by peak, with the *Interactive Peak Fit* software. From within this program, the user can add and delete peaks, change the regions limits, merge regions to create multiplets and split regions to create singlets. Adding peaks is the most time consuming step, since it requires the user to:

1. look at the spectrum in the main application window, searching for a localised increase in the number of counts (figure 1.13);
2. start the Interactive Peak Fit with the energy value of the new (supposed) peak as parameter;
3. add the peak;
4. check the correct fitting, background level and region limits (figure 1.14).

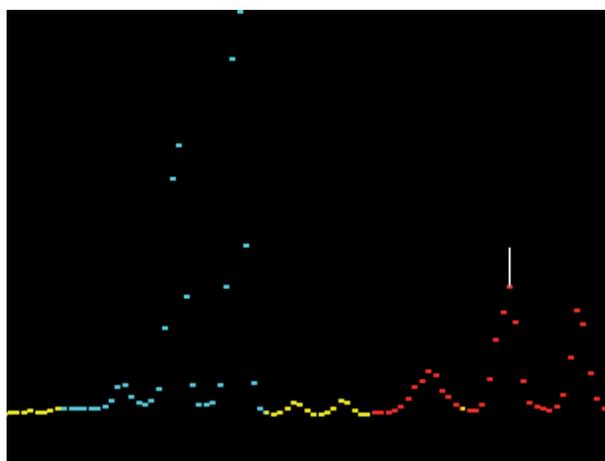


Figure 1.13: Two peaks not found by the automatic algorithm are visible between the multiplet, marked in cyan colour, and the series of single peaks, marked in red colour.

The next step is the background area subtraction and efficiency correction. These two steps are almost automatic and require only few clicks, since the user only has to check for the correct detector-background file matching and the correct efficiency calibration type.

Once the peak data are complete and correct, the user executes a *Nuclide Identification with Interference Correction*, with a library containing all the energy lines of natural radionuclides (figure 1.15).

The result of this step is a report where only peaks not matched by any energy line are listed, along with some sample information (table 1.1). The user now checks manually their fitting and their sources, with the aid of an external library of artificial radionuclides energy lines.

Once unknown peaks are all identified, the user can execute the *Nuclide Identification with Interference Correction* step again, with a library containing only the most important energy lines from natural radionuclides, a reduced version of the aforementioned library. The result of this analysis are the nuclides activities and uncertainties.

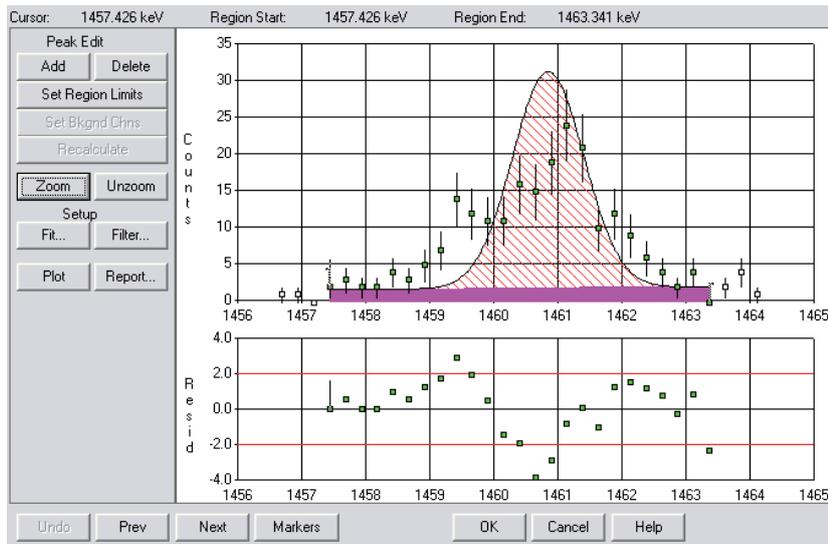


Figure 1.14: Example of a poor peak fitting.

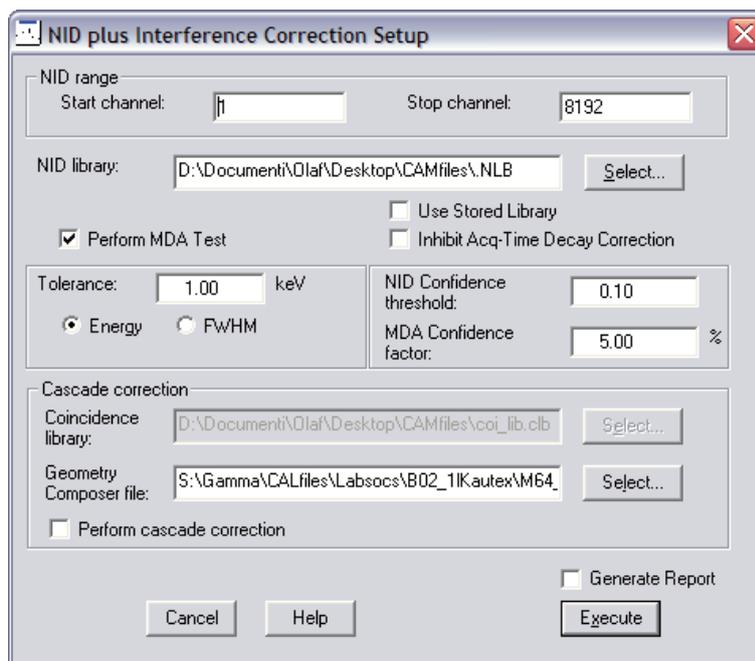


Figure 1.15: The settings window for the *Nuclide Identification with Interference Correction* step.

Unidentified peaks				
Peak nr.	Energy (keV)	Peak area	Peak FWHM	Area Uncertainty
6	105.83	190.83	0.62	43.57
31	1001.38	46.07	0.25	36.33
37	1621.49	23.83	0.80	41.94

Table 1.1: Example of an Unidentified Peaks results, with the sample information omitted.

Nuclides activities				
Nuclide name	Activity (Bq/g)	Uncertainty (%)	LD (Bq/g)	MDA (Bq/g)
K-40			1.84E-02	9.19E-03
Tl-208	3.24E-01	5.5	4.03E-03	2.02E-03
Bi-212	8.93E-01	7.3	4.25E-02	9.19E-03
Pb-212	8.33E-01	8.5	3.86E-03	9.19E-03
Ra-224	1.25E+00	21.2	3.83E-02	9.19E-03
Ac-228	9.09E-01	4.4	7.28E-03	9.19E-03

Table 1.2: Example of a Nuclide Activities results, with the sample information omitted.

The level of detection is obtained with an additional calculation. An example is shown in table 1.2.

The result obtained must now be checked for equilibrium of the activities. This step is done by hand and the comparison nuclide is the one with the lowest uncertainty. In the example shown in the table 1.2, the calculations to normalise the activities are:

$$A'_{Ac-228} = 9.09 \exp(-01)/1.000 = 0.909 \quad (1.18)$$

$$A'_{Ra-224} = 1.25 \exp(+00)/1.000 = 1.25 \quad (1.19)$$

$$A'_{Pb-212} = 8.33 \exp(-01)/1.000 = 0.833 \quad (1.20)$$

$$A'_{Bi-212} = 8.93 \exp(-01)/1.000 = 0.893 \quad (1.21)$$

$$A'_{Tl-208} = 3.24 \exp(-01)/0.362 = 0.891 \quad (1.22)$$

The activities are clearly in equilibrium, given the uncertainties of the activity measurements.

If the activities are not in equilibrium, the user has to check how the peaks have been matched with energy line, check again the peak fitting and execute again all the steps.

Chapter 2

Problems of the Traditional Routine

The analysis routine described in section 1.6 produces good results, but requires a great amount of time to be completed. In this chapter I will discuss in detail the main time consuming steps and how to improve them, in order to reduce the time required to complete an analysis.

2.1 Missing Peaks

The peak locate step, executed with the *Unidentified 2nd Difference*, is not able to find all the peaks, as shown in figure 1.13, and requires the user to fill the gap by inspecting the spectrum and manually adding additional peaks.

There are mainly three situations where the locate algorithm used can miss some peaks:

- two peaks are very closely spaced, so that the distance between the centroids is of the order of magnitude of the FWHM;
- a peak amplitude is very low, compared to the adjacent peaks;
- a peak amplitude is very low, compared to the background noise.

2.1.1 Peaks Closely Spaced

The first problem is shown in figure 2.1: two peaks are so closely spaced that the residuals of the simple fitting, calculated with only one peak, are very low and, thus, accepted by the locate algorithm. Peaks of this type, however, are easy to spot and to correct when the user manually scans all the spectrum with the *Interactive Peak Fit* software. Correcting this problem is a matter of a couple of clicks, but the user must first check if the new peak is found in a nuclide library: sometimes the spectrum can be fitted much better with an additional peak, even if it does not exist in nature (more on this topic in section 2.2). An example of a “non-existing” peak is shown in figure 2.3.

The problem of missing peaks is often found with Ra-224 and Pb-214 at respectively 241 keV and 241.9 keV. The two peaks are critical for the detection and activity

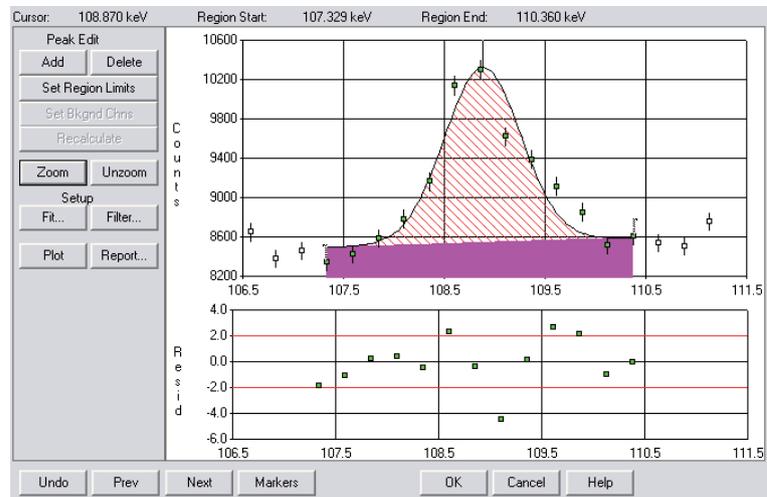


Figure 2.1: Two peaks so closely spaced that the peak locate algorithm is unable to discern them. The residuals are shown below the spectrum.

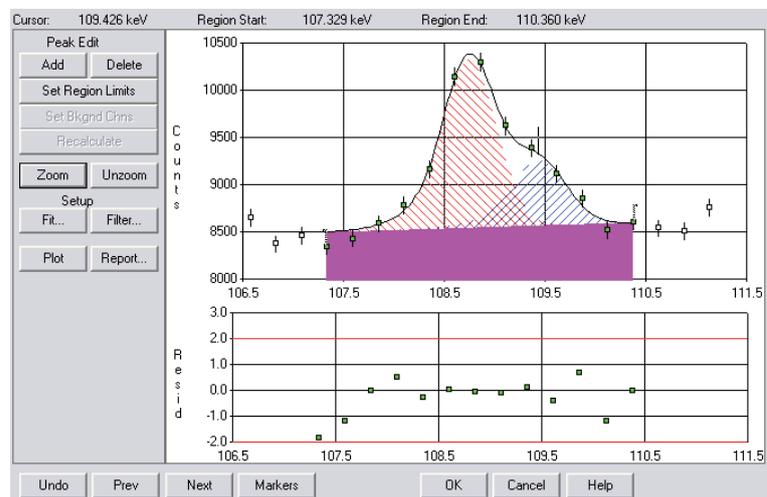


Figure 2.2: The same two peaks of figure 2.1, now correctly marked. The residuals are much lower and the interpolating curve fits perfectly the input data. The energies of the peaks are 108.6 keV and 109.1 keV and belong to Uranium-235.

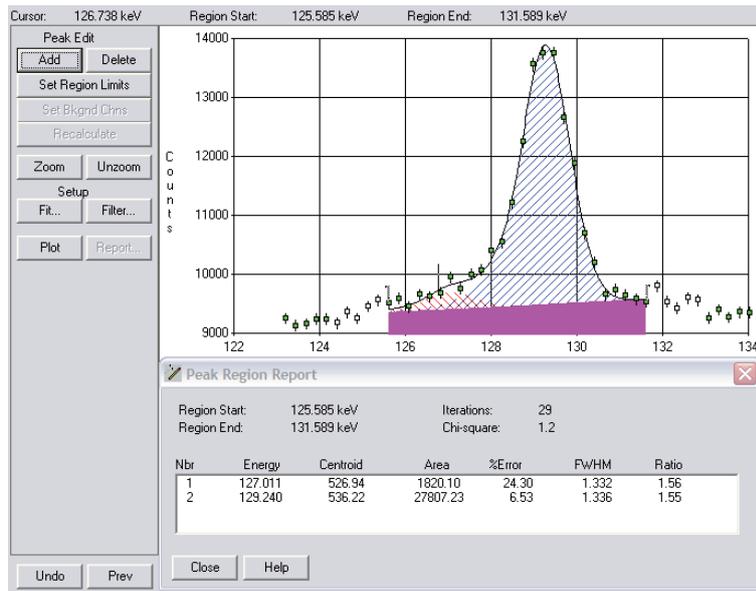


Figure 2.3: The red peak, on the left of the main one, is not listed in the library of natural radionuclides, it only improves the fitting of the low-energy peak tail.

Nuclides activities			
Nuclide name	Activity (Bq/g)	Uncertainty (%)	LD (Bq/g)
Tl-208	2.75E-00	5.26	1.28E-02
Ra-224	6.24E+00	26.06	1.27E-01
Ac-228	7.79E-00	4.18	2.81E-02
Bi-214	4.05E-00	3.69	1.91E-02
Pb-214	4.05E-00	8.28	2.71E-02
Pa-234m	5.64E-00	23.15	5.38E-01

Table 2.1: Results of a complete analysis, grouped by decay chain, with the peaks in the 241 keV region not correctly identified.

calculation of the two nuclides: merging them or missing the first peak prevents Radium from being identified and causes an higher calculated activity of Lead-214. The results of the analysis with the peak of Radium missing (figure 2.4) are shown in table 2.1, the results of the analysis with the peaks correctly marked (figure 2.5) are shown in table 2.2. The 241 keV region is, however, an extreme case: the residuals are very high and the 241 keV peak should be easily found, but the software is still not able to mark it. Tests with different tolerance parameters did not produce any good results.

2.1.2 Peaks Between Peaks of Greater Magnitude

The second type of missed peaks (peaks between much bigger ones) requires a greater amount of time to be corrected, since the user has to add the missing peaks with the complete procedure described in section 1.6. The advantage is that peaks are located

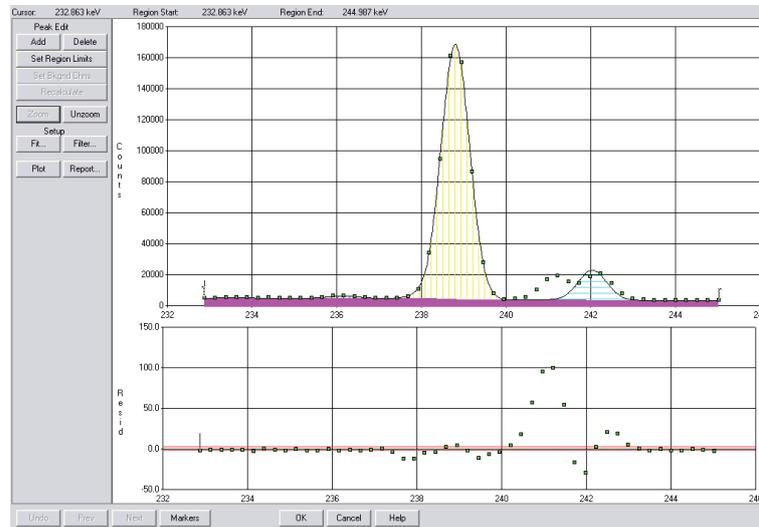


Figure 2.4: The 233-245 keV zone of the spectrum, immediately after *Unidentified 2nd Difference* peak locate and area calculation.

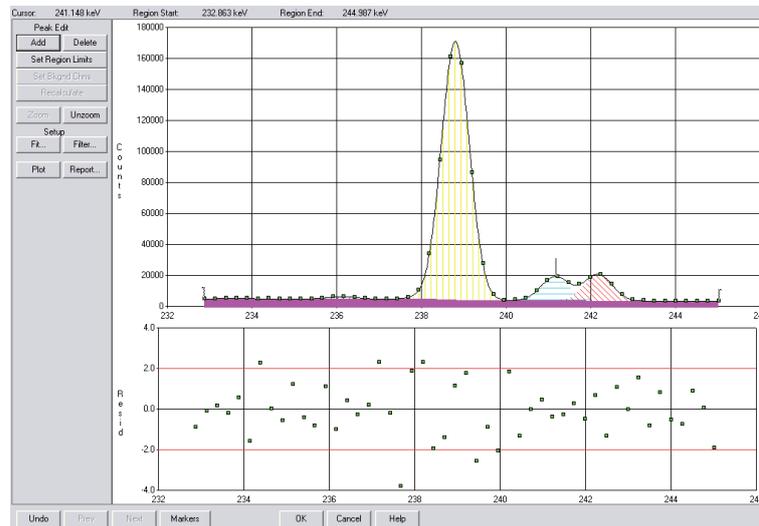


Figure 2.5: The 233-245 keV zone of the spectrum, with the 241.14 keV peak added (light blue).

Nuclides activities			
Nuclide name	Activity (Bq/g)	Uncertainty (%)	LD (Bq/g)
Tl-208	2.75E-00	5.26	1.28E-02
Ra-224	8.05E+00	11.54	1.27E-01
Ac-228	7.79E-00	4.18	2.81E-02
Bi-214	4.05E-00	3.69	1.91E-02
Pb-214	4.20E-00	6.83	2.71E-02
Pa-234m	5.64E-00	23.15	5.38E-01

Table 2.2: Results of a complete analysis, grouped by decay chain, with the peaks in the 241 keV region correctly identified. The value of Radium-224 activity is 30% larger and the uncertainty is lower.

near other peaks, so they are easy to spot. An example of this problem is shown in figure 2.6: the locate algorithm was unable to mark the two peaks, probably because the peaks on both sides are much bigger and the background continuum is very high.

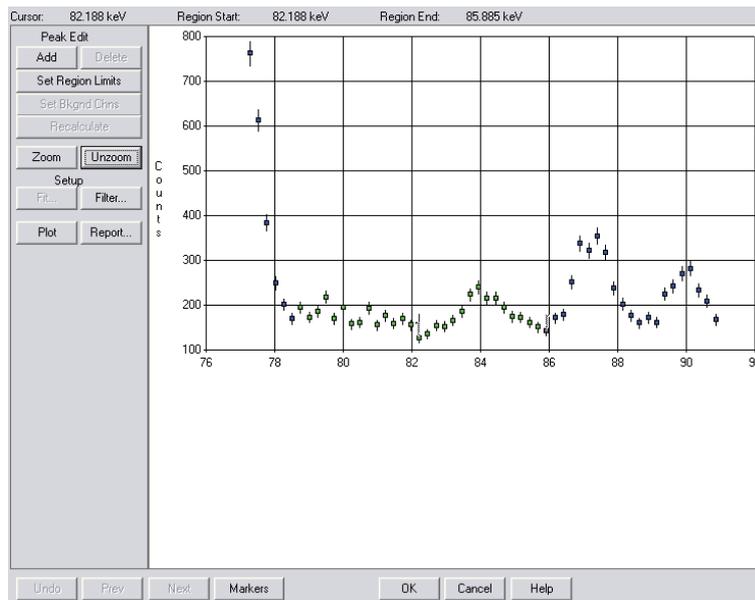


Figure 2.6: The peak centred on 84 keV was not recognised because it is located between much bigger peaks.

2.1.3 Peaks Lost in the Background Noise

The third type of missed peaks is found in the high-energy region of the spectrum, due to the low detector efficiency and the resulting low signal-to-noise ratio. An example of such peaks is shown in figure 2.8 and figure 2.9.

This type of peaks is the most difficult to recognise, because the user has to check manually the spectrum with the “zoom” mode, as shown in figure 1.12, go to the *Interactive Peak Fit*, try to add the peak, check the fitting and sometimes discard the

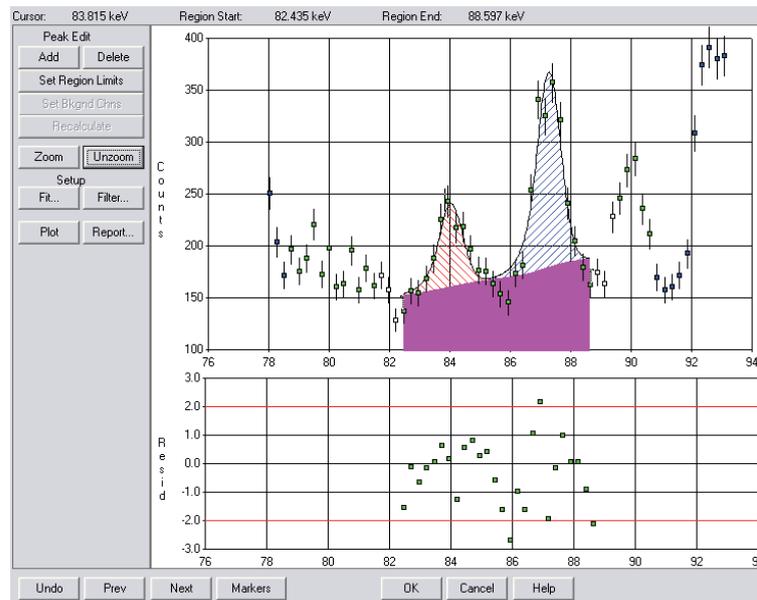


Figure 2.7: The same peak of figure 2.6, now correctly marked.

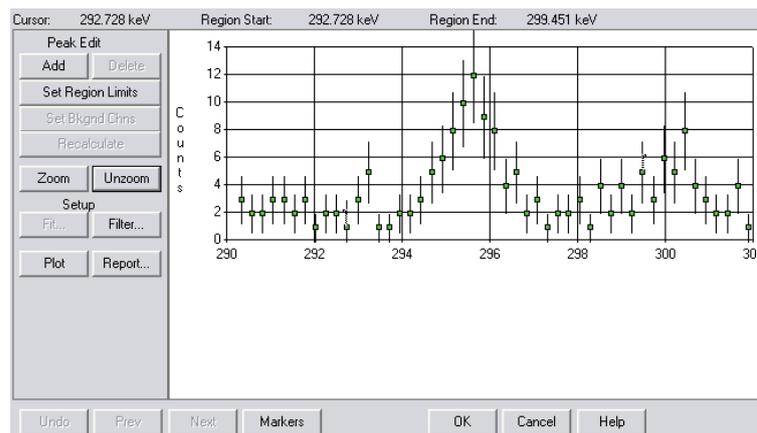


Figure 2.8: A missed peak in the high-energy region of the spectrum.

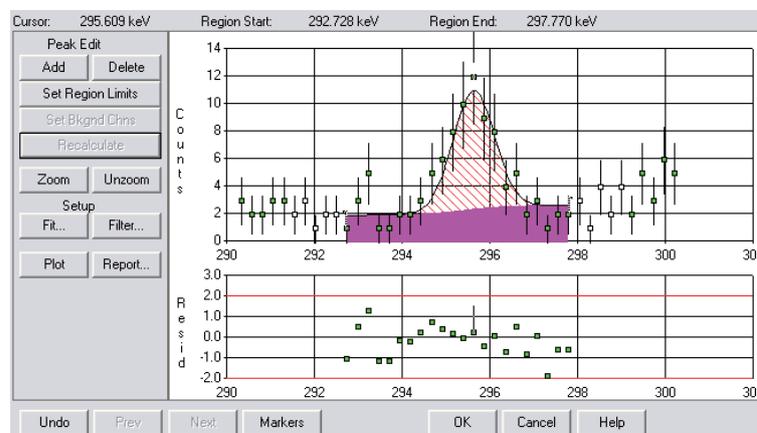


Figure 2.9: The same peak of figure 2.6, now correctly marked.

peak as originated by noise.

There are few small important peaks in the high-energy region, but, if the goal is to mark everything contained in the sample, the process is very time consuming.

The first attempt to solve this issue was focused on the “significance” value of the *Unidentified 2nd Difference*, able to increase or decrease the sensitivity of the algorithm. Changing this parameter, however, proved to have more drawbacks than positive results: when the value was decreased, the background noise was sometimes considered as peak, while if the value was increased, more peaks were not recognised as such.

2.1.4 Solution to the Problem

With the goal of improving the *Unidentified 2nd Difference* algorithm behaviour, several different significance values were experimented, but the result were not positive. Values between 3 and 5 lead to almost no differences, while values outside this interval brought up the problems previously described.

Other algorithms, among those offered by the Canberra software, were then evaluated. *Library (Gamma-M)* required the change of the area calculation algorithm from the previously used, well known *Sum/Non-Linear Least Square Fit* to *Library (Gamma-M)*, but still was not able to find all the peaks, so it was discarded. *VMS Standard Peak Search* found many peaks, but multiplets were not correctly marked, so I tried *Library (Simple)*. This algorithm is not suited to be used alone, since it is not able to detect unknown peaks, but worked perfectly as additional peak locate step, to improve the results of the standard algorithm.

The *Library (Simple)* algorithm, however, requires a nuclide library with the energy lines to mark in the spectrum. An optimisation of the library was then required: the goal of the additional step was to mark peaks not already found automatically by the standard algorithm, so only small peaks and overlapping peaks were kept, while peaks already marked by the *Unidentified 2nd Difference* algorithm were discarded.

The results of the addition of *Library (Simple)* to the standard routine were positive: more peaks in the high-energy region and more overlapping peaks were now correctly marked. The downside of this change was its increased sensitivity in the specific regions where the nuclide library listed an energy line: as already explained, the library locate algorithm always marks a peak for each energy line, but, sometimes, the area calculation algorithm is not able to reject it, resulting in a “non-existing” peak listed among the other peaks.

2.2 Non-Existing Peaks

“Non-existing” peaks are false peaks, marked by the software but not really found in the spectrum, and they can be seen in two different regions of the spectrum. The first one is the high-energy, low signal-to-noise ratio region, where the noise sometimes is marked as peak if the local shape is similar to a Gaussian. This type of peaks, easily recognised by the user, must be deleted manually. The traditional routine sometimes marked peaks of this type, the addition of the *Library (Simple)* peak locate algorithm increased the number of such peaks.

The second type of non-existing peak is found either in the low-energy tail of real Compton peaks or in the tails of peaks located in the high-noise regions. In both cases, an additional peak improves or does not worsen the spectrum fitting and the residuals magnitude, so the area calculation step cannot reject it. An example of this problem is shown in figure 2.10 and figure 2.11: The additional peak, marked with the colour blue, fits the spectrum nicely, only the user can recognise it.

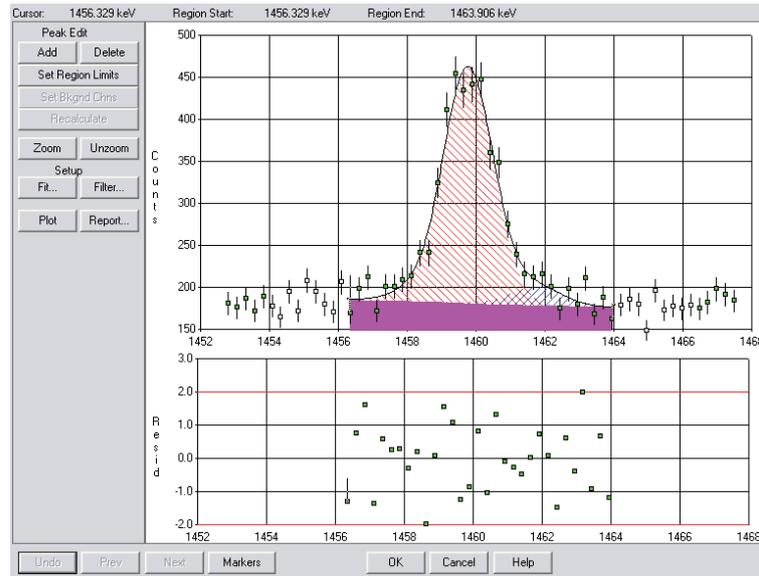


Figure 2.10: A peak left by the area calculation algorithm.

This type of peaks is originated mostly by the peak locate step: the library locate algorithm does not mark peaks within the energy tolerance of existing ones, but wide peaks can be fitted by two peaks, if there are enough energy lines listed in the nuclide library used by the *Library (Simple)* algorithm. Once marked, the peak cannot be rejected any more by the area calculation step because the fitting is good.

This type of non-existing peak must be corrected by the user manually, during the scan of the spectrum with the *Interactive Peak Fit* software. The process of identifying such false peaks is quite easy for the experienced user: they always appear as very small peaks located in the lowest part of the tail of the main peak. Moreover, the software reports their uncertainty as extremely high: typical values range from 50% to 200%.

2.3 Peak Identification Step

The peak identification step is used, as previously explained, to isolate peaks that cannot be matched to any energy line, both from natural or artificial radionuclides.

The traditional routine, used to identify peaks marked during the locate step, starts with the *Nuclide Identification with Interference Correction* software algorithm, used with a nuclide library containing all the natural radionuclides energy lines. The software would then identify the nuclides and mark as known all the peaks belonging to

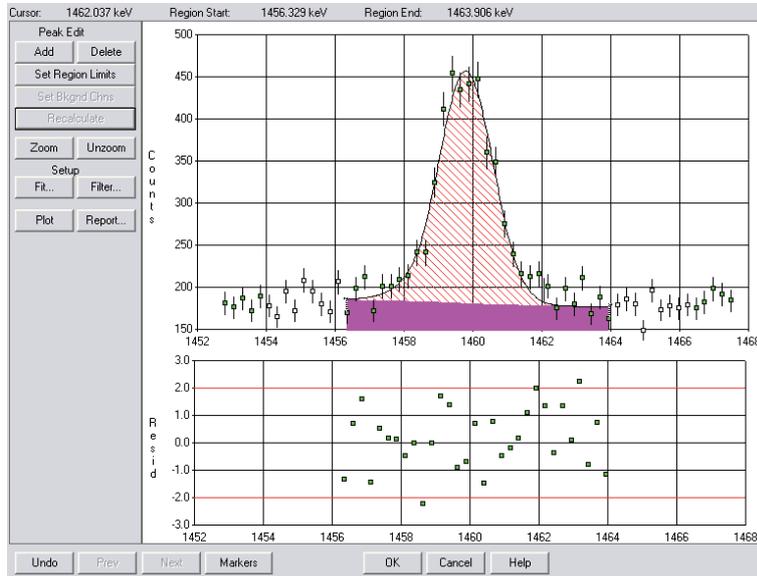


Figure 2.11: The region of figure 2.10, after the manual correction.

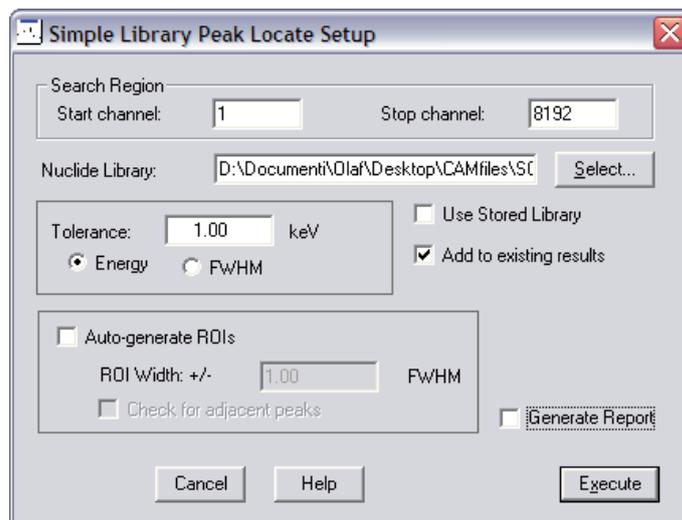


Figure 2.12: Library (Simple) parameters.

them. With the assumption that all the radionuclides found in the sample are always identified, the method is correct.

The downside of the method is the identification algorithm: the Canberra algorithm always uses, during the identification of a nuclide, all the energy lines associated to it. When many energy lines are listed, but only a fraction of them is found due to the low sample activity, the nuclide sometimes gets completely discarded and all its peaks found in the spectrum are left as unidentified. To solve this problem, the user can only execute the identification step again, with a smaller library, to exactly know which nuclides are found. Once this information is known, he then checks manually, against the big library, the peaks left as unidentified by the previous step.

The whole process is very time consuming: to summarise, the user first executes a nuclide identification with a small library containing only the main energy lines, then prints the results, executes again the identification with the complete library and, finally, checks manually the peaks still left as unidentified. The manual check is done first against the complete natural radionuclide library, then against a library containing only and all the artificial radionuclides energy lines. A peak not matched by any of them will be marked as non-existing peak, a peak matched against the artificial radionuclides nuclide library must be highlighted in the final customer report.

2.3.1 Solution to the Problem

To solve both the problem of the non-existing peaks and the problem of the long time required for each peak identification, the software I developed has an enhanced identification routine.

The main concept is the following: first, nuclides are identified with the small nuclide library in order to find all all of them, then the software checks automatically the unidentified peaks against the complete nuclide library. Peaks correctly identified are automatically removed from the final list, while peaks not identified by the new software are shown to the user, with detailed information about energy, area and calculated activity. The user has always available the list with all the energy lines of the nuclide libraries, to avoid the use of an external software and the manual search of the energy lines. More details on the topic can be found in section 3.5.

2.4 Activity and Decay Chain Calculations

During the nuclide activity calculation step, the user needs to check which peaks the software used for the activity calculation and the calculated activity of each peak. The traditional way to accomplish this task was to execute a report and to look manually at it. If a peak had an activity much different from the average nuclide activity, the user had to manually go to the *Interactive Peak Fit* and check the peak fitting. After the nuclide identification and activity calculation, the user then needed to check the decay chain equilibrium, as previously explained. This step required the user to do some calculation manually.

The new software has a revamped nuclide identification interface, where all the needed information are shown at the same time and manual computation are avoided. The software shows graphically the activities of the identified nuclides, grouped by

decay chain, with the addition of the activity uncertainties and the level of detection of each nuclide, useful when a nuclide is missing. Every value is scaled by the relative nuclide abundance in the decay chain, in order to avoid the manual calculation previously mandatory.

Each peak used for the activity calculation is also shown, along with its specific activity and level of detection, along with peaks listed in the nuclide library but not found in the spectrum: if needed, the user can go directly to the *Interactive Peak Fit* to check the energy region where the peaks should have been found.

Chapter 3

The Program

3.1 Main Guidelines and Program Structure

The software I developed has been created to solve the problems described in the previous chapter. It relies on the standard Canberra libraries for the core routines, while the user interface and the new routines I developed were implemented in *Visual Basic .Net*.

The standard Canberra libraries ensure coherency of the new results with the past while keeping the development time short.

The new interface has been designed to be easy and straightforward, where all the functions and every data the user in each step needs are immediately visible and available. To better accomplish this goal, the software has only one window per analysis step.

All the parameters left unchanged while executing similar analyses have been grouped and moved into a separate preferences window, to clean up the other windows and to make the introduction of “profiles” possible. A “profile” is a set of values saved to disk and loaded as a whole upon user request; the ability to use them is extremely useful during the testing of new parameters and when analysing different samples types: instead of making the specific changes manually, the user simply loads the correct profile.

The introduction of profiles was significantly simplified thanks to the use of standard Canberra analysis sequence files to execute the different steps: each step is a separate ASF file with the algorithm names and all the needed parameters stored inside: tracking profiles is, as consequence, only a matter of keeping different files and executing the correct one. Moreover, no custom file format with the stored settings is required, is used only a standard Canberra file, editable with the Canberra tools as well. As instance, the *Nuclide Identification* step is constituted of a single file, where both the *Nuclide Identification with Interference Correction* and the *Detection Limits/KTA MDA* steps are saved: my code simply calls the Canberra’s generic analysis routine with the specific file name as parameter.

To speed up the processing, I decided to fully automate the steps that do not require user interaction. Two examples are the two area calculation and efficiency correction steps that must be executed after each *Interactive Peak Fit* execution and the consequent change of the peak fitting. As shown in figure 3.1, the user could have to go

back to the peak fitting from almost every step of the analysis, and after each change at least the two steps mentioned must be executed again, mechanically (the original efficiency correction window is shown in figure 3.2).

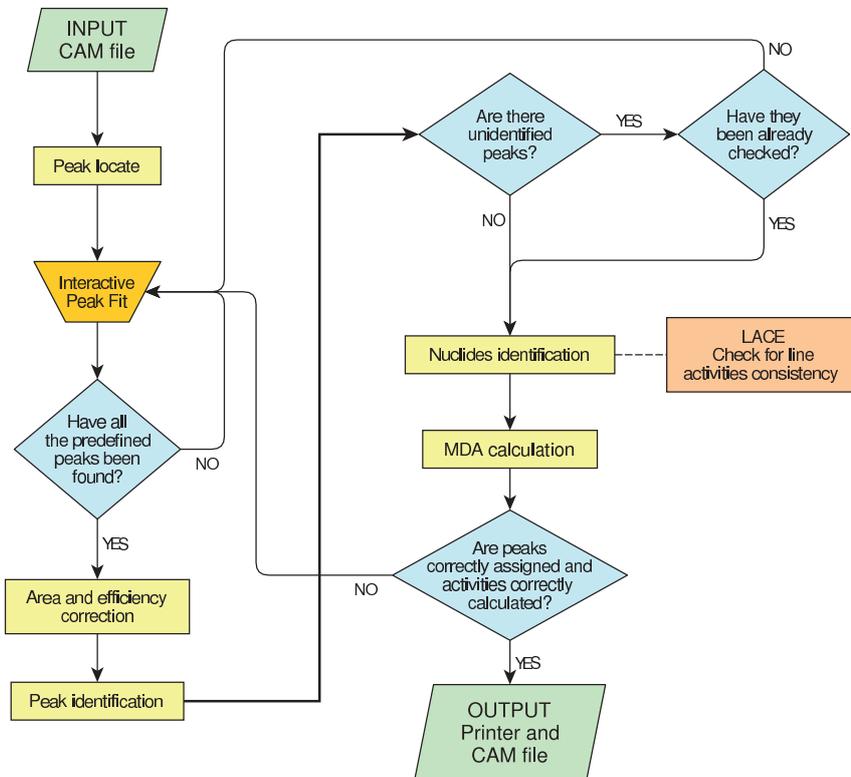


Figure 3.1: Software structure flow chart.

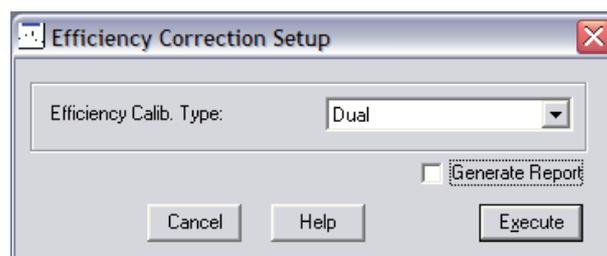


Figure 3.2: Efficiency correction step.

3.2 Main Window and Sample Properties

The main window (figure 3.3) is used to access to all the program's functions and to show both the spectrum and the reports.

The buttons on the left are grouped by function: visualisation, reporter related, data file related, processing related.

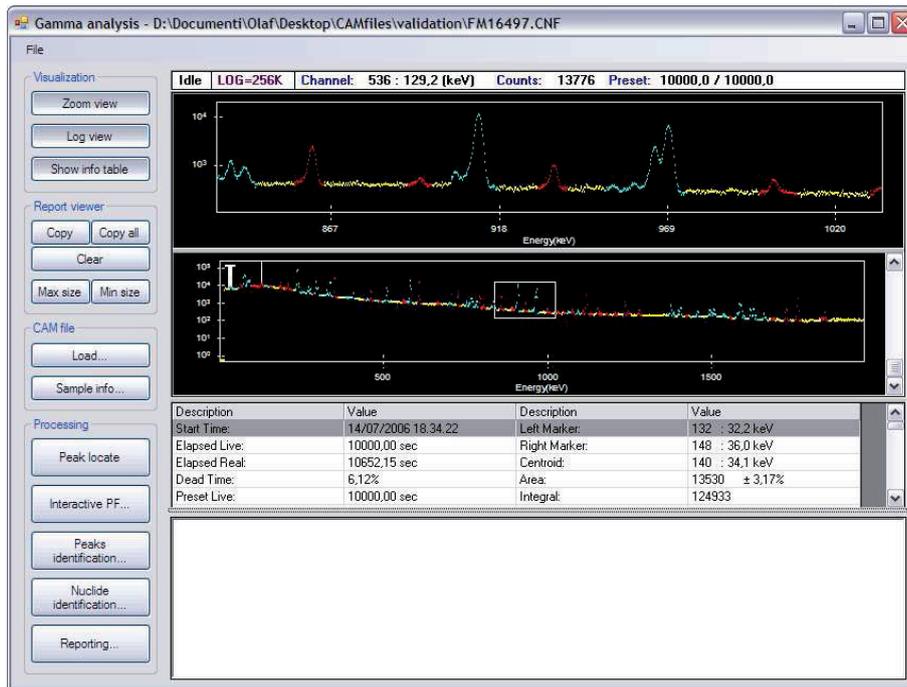


Figure 3.3: The main program window, with enabled Zoom view, Logarithmic scale and the additional data table. The report pane has been resized manually, to make more space for the spectrum view.

In the first group there are three buttons: “Zoom view” changes how the spectrum is displayed a detailed view in the upper half of the spectrum area; “Log view” changes the vertical scale, from linear to logarithmic, better suited to high-activity samples; “Show info table” shows, in the lower part of the spectrum area, a table with detailed data about the sample, such as the acquisition time.

The second group affects the contents and the appearance of the reporter pane, the white space where reports are displayed: they increase and reduce its size, clear the contents or copy the contents to the system clipboard.

The next two buttons load the CAM file, where the initial counts and the results of the calculation are stored, and show a window with the sample properties. This window, shown in figure 3.4, let the user check and change the most important sample properties. Some of them are editable, some of them read-only. The “Geometry/efficiency”, “GIS file” and “Energy calibration” cannot be edited manually, because they represent a set of parameters that can only be loaded as a whole from existing data files (calibration files).

Back in the main application window, the last group of buttons has an item per analysis step. Steps requiring additional user input or producing some output will open a child window, while the others will only show a confirmation of the successful completion of the work.

Figure 3.4: Sample properties window.

3.3 Peak Locate Step and Correction Step

This step does not require any user input, it only requires a confirmation because existing peaks are overwritten.

This step executes the *Unidentified 2nd Difference* routine, to first mark unknown peaks and peaks easily detected, then the *Library (Simple)* routine to try to find peaks missed by the previous routine. This way, if there is a small energy shift, a peak that could be found by both algorithm will be marked in the correct position, while the library locate algorithm would have positioned it in the correspondence of the energy stored in the nuclide library, disregarding the actual position.

After the peak locate step, the area calculation step is always required, so it is executed immediately.

The areas of the peaks must be corrected for both background counts and detector efficiency, so a correction step is needed after the peak locate and after each *Interactive Peak Fit* execution.

The CAM file with background counts data has a name that depends on the detector name, so the user usually checked the correct matching. To avoid any user interaction, I put in the preferences window a field where a file name “mask” can be defined (figure 3.5). The software will substitute the detector number in the user-defined mask, to create on-the-fly the correct background file name.

The efficiency calibration required by the specific sample can be changed in the “Sample properties” window (figure 3.4).

3.4 Interactive Peak Fit

The check for correct peak fitting is a step always executed by user after each peak locate, so a window has been dedicated to it. Not only, I used this window also to

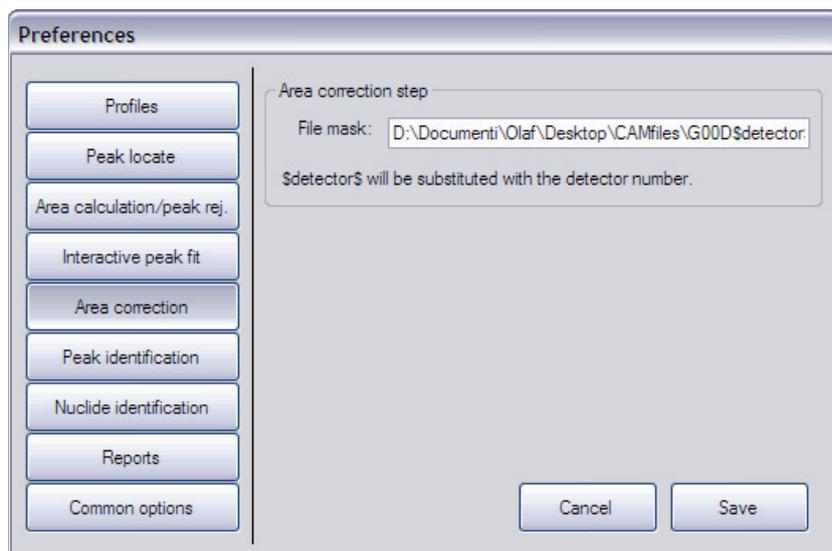


Figure 3.5: Background file mask in the global preferences.

help the user check if some peaks were missing, an important step because I found a problem in the Canberra routines: some peaks are discarded after being correctly marked by the peak locate routines. This is, sometimes, the cause of the loss of the Radium-224 241 keV peak: such peak, if missed by the *Unidentified 2nd Difference* algorithm, is correctly marked by the *Library (Simple)* peak locate, but then it is also discarded by the *Sum/Non-Linear Least Square Fit* algorithm. Changing the area calculation step parameters (shown in figure 1.11) did not help, because the two peaks are extremely close and a change in the algorithm parameters would affect many other peaks, correctly marked with the current values.

The solution, offered in this window, is a custom list where the user adds “usually missed by the software” peaks. The software will then show one list with peaks listed in it and one with peaks found in the spectrum. The original idea was to have the software make the matching and hide from the custom list all the peaks correctly found in the spectrum, but it was impossible, so the user has to complete this task manually. The problem I encountered was the peaks spacing: when the distance between two peaks is of the same order-of-magnitude of the user-defined uncertainty, a wrong matching can occur.

This window name, *Interactive Peak Fit*, has been chosen to remember the user he can directly jump to the external peak fitting software with each one of the buttons positioned in the bottom of the window and also with a double click on a peak in either list. One of the buttons also allow the user to jump to a specified energy in the spectrum, ability useful while inspecting the spectrum to check for additional peaks not caught by any peak locate routine.

3.5 Peak Identification Step

The identification step must correctly identify peaks belonging to nuclide found in the sample, while showing enough details of the other peak to help the user to understand better and faster the nature of the peaks.

With this goals in mind, I designed a window where both a list of unidentified peaks and a list of energy lines taken from nuclide libraries are shown. As result (figure 3.6), there are four different sections: “Unidentified peaks” (top-left), details of the selected peak (centre-left), “Nuclides” (right), nuclide libraries selection and options (bottom).

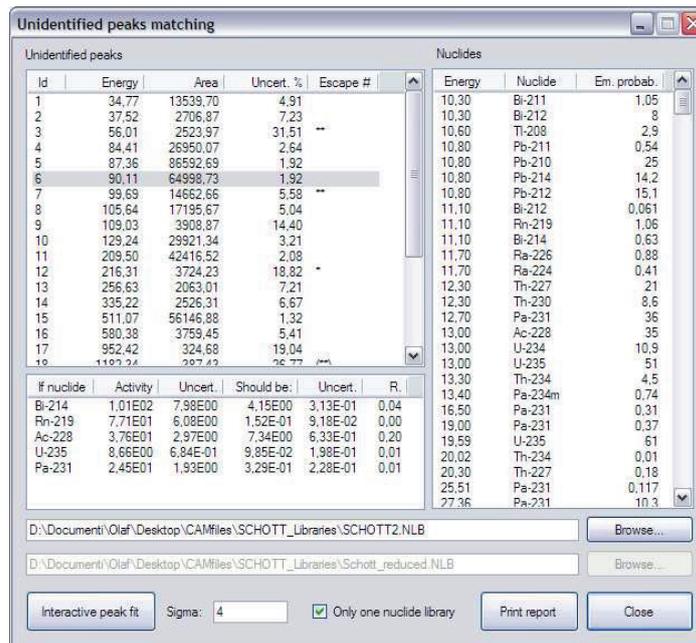


Figure 3.6: Peak identification window. The option *Only one nuclide library* is selected, so only the first library is displayed in the list on the right.

The “Nuclides” section shows all the energy lines listed in, depending on the use preference, the first or both the nuclide libraries displayed in the lower part of the window. The list contains nuclide name and energy of each entry and is sorted by energy.

The “Unidentified peaks” and the list with details of the peaks show the results of the peak identification algorithm displayed in the flow chart of figure 3.7. The algorithm is divided in two parts: in the first one the goal is to try to match the unidentified peak with an energy line of an identified nuclide, while, in the second part, the peak is matched against other peaks in order to try to identify it as escape peak.

Associating a peak with an identified nuclide requires two consecutive steps: in the first, the nuclide must match an energy line found in the complete NORM-nuclides library; if this first step succeed, the peak activity is calculated and compared to the value of the identified nuclide. The list of identified nuclides is generated by the Canberra libraries during the execution of the *Nuclide Identification with Interference Correction* algorithm, called every time the “Peak Identification” window is opened. The library

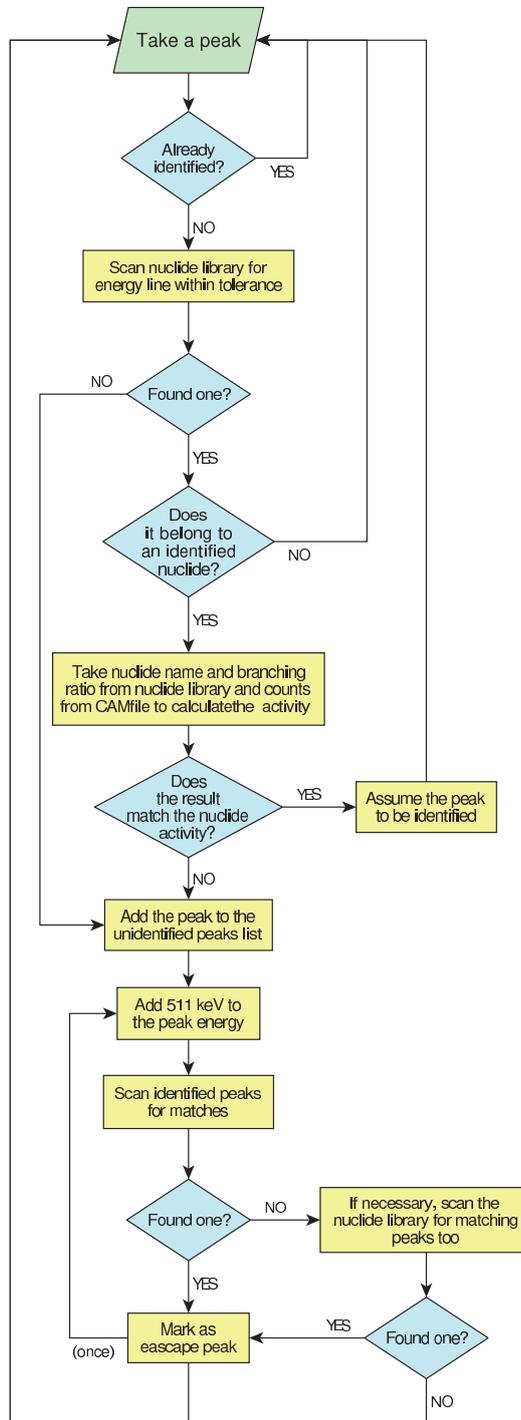


Figure 3.7: Peak identification flow chart.

used during this nuclide identification is the small nuclide library that ensures all the nuclides found in the sample are correctly identified. The unidentified peak matching I will describe is, as already stated, executed against the complete NORM-nuclides library, to match more peaks.

The matching against energy lines found in the nuclide library is satisfied when the following relation is satisfied:

$$E_{\text{unid}} - \delta E_{\text{unid}} \leq E_{\text{lib}} \leq E_{\text{unid}} + \delta E_{\text{unid}} \quad (3.1)$$

Where:

E_{unid} is the unidentified peak energy;

δE_{unid} is its uncertainty;

E_{lib} is the energy line value.

These three values are stored, respectively, in the CAM_F_PSENERGY, CAM_F_TOLERANCE, CAM_F_NLENERGY parameters of the sample CAM file. The uncertainty value is set by the user in the preferences and fixed for all the sample peaks.

When a match is found, the list of nuclides identified in the sample is checked for the presence of the nuclide the energy line belongs to. If a match is found, the activity calculation step is executed.

Activity calculation requires the knowledge of many different data about the peak and about the sample as a whole. Short-lived radionuclides can decay very fast, so the time elapsed during the counting and the time elapsed since the sample was taken must be taken into account. When the activity calculation is done using the raw number of counts, the detector efficiency must also be included in the computation. To summarise, the formula used by the software for each peak activity calculation is the following:

$$C = \frac{S}{M \cdot \epsilon' \cdot y \cdot T_1 \cdot U_f \cdot K_c \cdot K_w} \quad (3.2)$$

Where:

C is the activity per volume unit;

S is the number of counts (parameter CAM_F_PSAREA);

M is the sample mass (parameter CAM_F_SQUANT);

ϵ' is the attenuation corrected efficiency (parameter CAM_F_PSEFF);

y is the peak branching ratio (parameter CAM_F_NLABUN);

T_1 is the live time of the collect in seconds (parameter CAM_X_ELIVE);

U_f is the conversion factor required to have the activity in μCi , the fixed unit of measurement used by all the Canberra algorithms;

K_c is the correction factor for the nuclide decay during counting (equation 3.4);

K_w is the correction factor for the nuclide decay from the time the sample was obtained to the start of the counting (equation 3.5).

The attenuation corrected efficiency is defined as:

$$\epsilon' = \epsilon \exp[-\mu(E) \rho t] \quad (3.3)$$

Where:

ϵ is the non-attenuation corrected detection efficiency at the peak energy;

$\mu(E)$ is the mass attenuation (in units of cm^2/g) at gamma energy E ;
 ρt is the average sample mass per unit area.

The correction factor for the nuclide decay during counting, K_c , is defined as:

$$K_c = \frac{T_{1/2}}{\ln_2 t_c} \left[1 - \exp\left(-\frac{\ln_2 t_c}{T_{1/2}}\right) \right] \quad (3.4)$$

Where:

$T_{1/2}$ is the half-life of the nuclide in question;

t_c is the elapsed real clock time during the measurement in the same time unit as $T_{1/2}$.

The K_w correction factor for the nuclide decay from the time the sample was obtained to the start of the collect, K_w , is defined as:

$$K_w = \exp\left(-\frac{\ln_2 t_w}{T_{1/2}}\right) \quad (3.5)$$

where t_w is the elapsed clock time from the time the sample was taken to the beginning of the measurement (in the same time units as $T_{1/2}$).

The theoretical formula was, however, simplified by ignoring the two correction coefficients K_c and K_w . The latter is not known because the time from when the sample is taken to when the counting starts is not recorded, while the former is always close to 1, as the graph in figure 3.8 shows. Only nuclides with a very short half-life would need the correction coefficient, but not when they are in equilibrium with mother- and daughter-nuclide, so it can be safely removed.

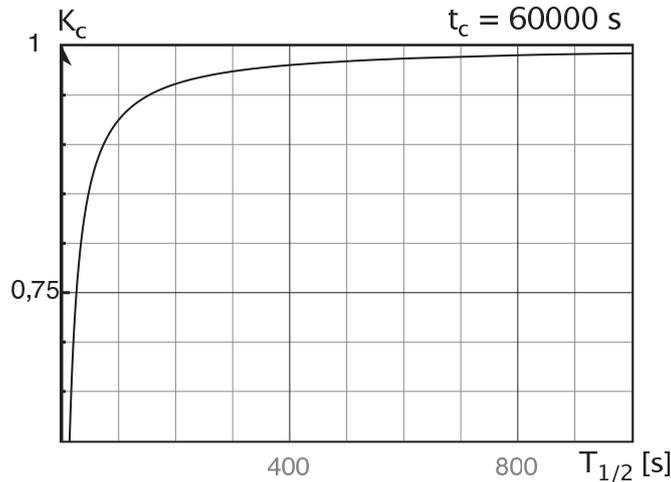


Figure 3.8: K_c value as function of nuclide half-life.

The formula used in the calculation, with the approximations described, is:

$$C = \frac{S}{M \cdot \epsilon' \cdot \gamma \cdot T_1 \cdot U_f} \quad (3.6)$$

The activity uncertainty is calculated as:

$$\delta C = C \cdot k \cdot \left[\frac{\sigma_{\text{syst}}}{100} + \sqrt{\left(\frac{\sigma_{\text{rand}}}{100}\right)^2 + \left(\frac{\sigma_{\epsilon}}{\epsilon}\right)^2 + \left(\frac{\sigma_S}{S}\right)^2} \right] \quad (3.7)$$

Where:

- k is the user-defined global uncertainty multiplier;
- σ_{syst} is the user-defined systematic uncertainty (parameter CAM_F_SSYSTERR);
- σ_{rand} is the user-defined random uncertainty (parameter CAM_F_SSYSERR);
- σ_{ϵ} is the detector efficiency uncertainty for the peak in question (parameter CAM_F_PSEFERR); σ_S is the net peak area uncertainty (parameter CAM_F_PSDAREA).

The final activity comparison is satisfied when

$$C - \delta C \leq C_{\text{nuc}} + \delta C_{\text{nuc}} \wedge C + \delta C \geq C_{\text{nuc}} - \delta C_{\text{nuc}} \quad (3.8)$$

Where:

- C is the calculated peak activity;
- δC is the calculated peak activity uncertainty;
- C_{nuc} is the nuclide activity (parameter CAM_G_NCLWTMEAN);
- δC_{nuc} is the nuclide activity uncertainty (parameter CAM_G_NCLWTMERR).

The activity calculation described is executed for all the possible energy matches of each unidentified nuclide, as long as the possible originating nuclide has already been identified. This way, if the same unidentified peak energy can be matched against more energy lines in the nuclide library, the software will be able to show the user the different alternatives in a table (middle-left of the window of figure 3.6). The details shown in each entry are the matching nuclide name, as taken from the library energy line, the calculated activity and uncertainty of the unidentified peak when the peak is assumed to belong to the nuclide listed in the first column, and the nuclide activity and uncertainty, as written in the CAM file.

The check for escape peaks (discussed in section 1.3) is done in a similar manner, after a 511 keV or 1022 keV energy shift is applied to the unidentified peak:

$$E_{\text{unid}} - \delta E_{\text{unid}} \leq E_{\text{peak}} + \delta E_{\text{peak}} \wedge E_{\text{unid}} + \delta E_{\text{unid}} \geq E_{\text{peak}} - \delta E_{\text{peak}} \quad (3.9)$$

where E_{peak} is the energy of the already identified peak and δE_{unid} is its energy uncertainty. The latter is the same as δE_{unid} , since they are both equal to the user-defined energy tolerance.

Once this step has been completed, another comparison is executed with energy lines found in the nuclide library and characterised by an higher energy than the most energetic identified peak. This check is done because the CAM file lists only peaks within the detector energy range, while an escape peak could be originated by a gamma ray of higher energy. Energy lines found in the library have no uncertainty, so the check is the same of equation 3.1.

The preferences of this analysis step are shown in figure 3.9.

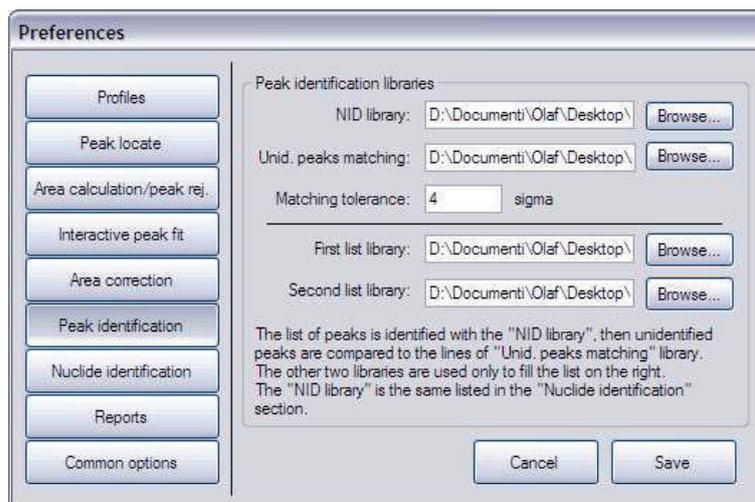


Figure 3.9: Preferences of the new peak identification step.

A result of this new, improved identification algorithm, not based on the double nuclide identification algorithm execution, is the ability to leave as unidentified more peaks and, thus, to be able to better analyse the spectrum. The cause of the increased number of unidentified peaks is the following: when, in the traditional routine, the nuclide identification with the complete library was executed, the Canberra algorithm used all the energy lines associated to each nuclide and calculated the mean activity weighting them all, with the branching ratios as weights.

This led to a bigger number of identified peaks, but allowed for the identification of peaks with extremely different activity values too: the activities were, sometimes, spread over more than three orders of magnitude but the peaks were marked as identified anyway. The final mean activity was not always influenced significantly because this problem arose only with small peaks, but such peaks were then hidden and the user never noticed them. With the new routine, instead, the software will list them and the user will act consequently. Maybe a better fitting is possible, maybe the peak is only made of noise and must be removed.

3.6 Nuclide identification Step

The “Nuclide Identification” window (figure 3.10) has been designed to show nuclides activities numerically and graphically, both with uncertainties and levels of detection. This helps the user to compare the activities of the nuclides of the same decay chain and to associate a missing nuclide to an high level of detection. To help the user doing the decay chain activity comparison, I introduced an uncertainty factor; the effects of every change of this value are applied to the graphs in real-time.

All the peaks associated to each nuclide are shown, upon request, in a list on the right side of the window, each one with the specific activity, uncertainty, branching ratio and minimum level of detection. The last two data are specially useful with peaks not found, displayed along with peaks found and identified: a missing peaks can be associated to an high level of detection in the specific spectrum region and,

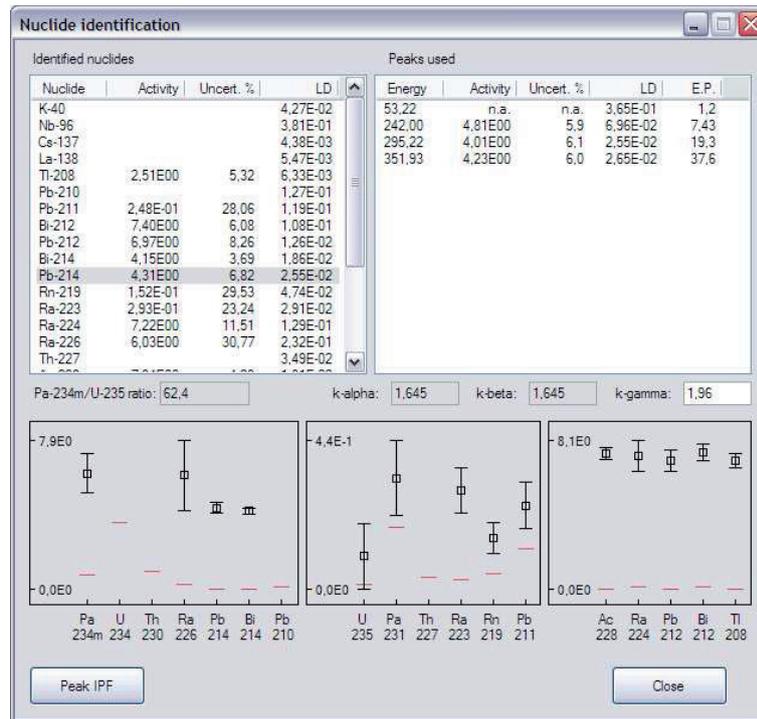


Figure 3.10: “Nuclide Identification” window with some test results.

thus, justified. The user can, however, jump directly to the external *Interactive Peak Fit* application to check the fitting of existing and to look manually for missing peaks, with a simple double click on a peak.

Another feature I implemented in the “Nuclide Identification” window is the Uranium-238-to-Uranium-235 activity ratio calculation. The traditional routine required the user to do the calculation manually, I automated the procedure. Not only, I made the calculation more flexible, so that if the mother nuclide or another nuclide of the two decay chains is missing, the algorithm find the new best choice and uses it. The actual nuclide used for the calculation is displayed next to the ratio value.

A feature requested but impossible to implement was the identification of peaks used for interference correction by the Canberra identification algorithm. Theoretically, the algorithm keeps track of them, but, when I tried to read the CAM file variables associated to it, they were empty. A bug report was sent to Canberra for this problem, but no feedback was received, so the feature has not been implemented.

3.7 Reporting Step

The reports window has been designed to ease the execution of four different reports, required for each sample printed documentation: nuclide activities report, unidentified peak list report, found peaks list, Excel report for the customer. The new peak identification routine has its own report print function, so three reports can be printed. To speed up the process, I designed a window able to offer the user all the reporting steps at the same time, as shown in figure 3.11.

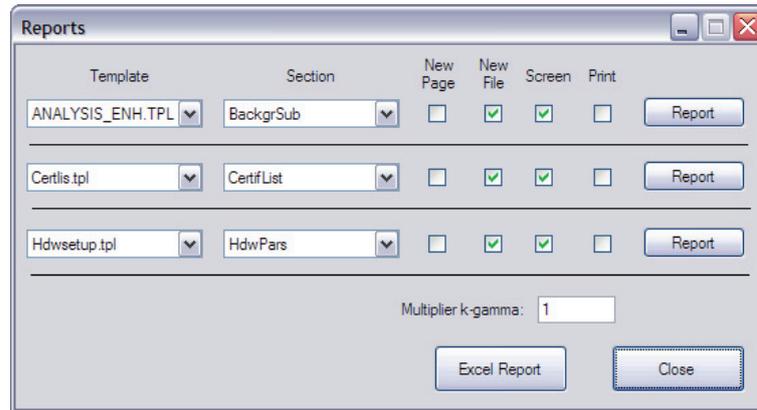


Figure 3.11: Reports window.

In the window three different, customisable reports ready for printing. The “Excel report” button writes all the nuclides activities, uncertainties and levels of detection in an external Excel file, that acts as mask, containing some macros. After all the required fields have been filled, the Excel file is opened and the user can see and print the results.

Compared to the previous routine, where the values were copied to the Excel file almost manually and with only two meaningful decimals, this procedure lead to an improvement in the final report precision and to a speedup of the procedure.

Chapter 4

Validation of the Results

The validation of the new routine has been done with many samples of both high and low activity. The conclusions are always similar, so I will show here only the results of the analysis of two samples. The first one has a low number of counts and shows the behaviour of the software in presence of noise, the second sample has high activity levels and shows the behaviour with well defined peaks.

4.1 Low Activity Sample

4.1.1 Peak Locate Results

The comparison of the results of the peak locate step has been done first by executing the automatic search and then by checking manually the spectrum for missed peaks. The results are listed in tables 4.1 and 4.2.

The results show that the new algorithm is able to mark all the peaks the old algorithm would, plus almost all the remaining peaks the user would have to add manually.

Peaks 1 and 2 are the same peak in the spectrum, the different energy depends on the algorithm used to mark them: the new routine first marks a peak with the *Unidentified 2nd Difference* algorithm and then another one, with *Library (Simple)*, in the energy specified in the nuclide library. The peak area calculation algorithm keeps only the second one because it fits the spectrum the best. The old routine has only one peak (the first one), so it is kept.

Peak 7 was missed by both routines. The *Library (Simple)* algorithm, used by the the new routine, cannot mark it because the peak has been considered as important during the nuclide library optimisation, so it was not included: either it is marked by the *Unidentified 2nd Difference* algorithm or it is lost. However, when I tried to add it to the nuclide library, I found the peak to be correctly found, so a re-introduction of the peak in the library can be evaluated in the future.

Peak 18 was found only by the new routine, but it was not marked correctly: during the manual spectrum check, it was removed and peaks 16 and 17 have been marked in the same region.

Peak 40, 41 and 42 belong to the same multiplet, but they were marked differently depending on the routine used (figures 4.1, 4.2 and 4.3). The new routine correctly marks peak 41 (source: Actinium-228), but misses the much bigger peak from Potassium-40.

Peak Nr.	Energy (keV)	Found by the old routine	Found by the new routine	Manually removed
1	39.88		•	
2	40.03			
3	46.51		•	
4	63.38	•	•	
5	74.85	•	•	
6	77.14	•	•	
7	84.00			
8	87.30	•	•	
9	89.81	•	•	
10	92.71	•	•	
11	94.81			
12	185.96	•	•	
13	238.70	•	•	
14	240.73		•	
15	242.00	•	•	
16	270.01			
17	270.99			
18	271.23		•	•
19	295.23	•	•	
20	300.31		•	
21	328.07		•	
22	338.36	•	•	
23	351.90	•	•	
24	462.75	•	•	
25	510.96	•	•	

Table 4.1: List of peaks (1-25) found in the first sample spectrum. Peaks not found by any algorithm have been added manually.

Peak Nr.	Energy (keV)	Found by the old routine	Found by the new routine	Manually removed
26	583.14	●	●	
27	609.30	●	●	
28	665.37		●	
29	727.42	●	●	
30	768.34	●	●	
31	806.22	●	●	
32	860.67		●	
33	911.11	●	●	
34	934.19	●	●	
35	968.89	●	●	
36	1120.12	●	●	
37	1238.09	●	●	
38	1377.03	●	●	
39	1408.14	●	●	
40	1459.68		●	
41	1460.83	●		●
42	1461.17			
43	1764.53	●	●	

Table 4.2: List of peaks (26-43) found in the first sample spectrum. Peaks not found by any algorithm have been added manually.

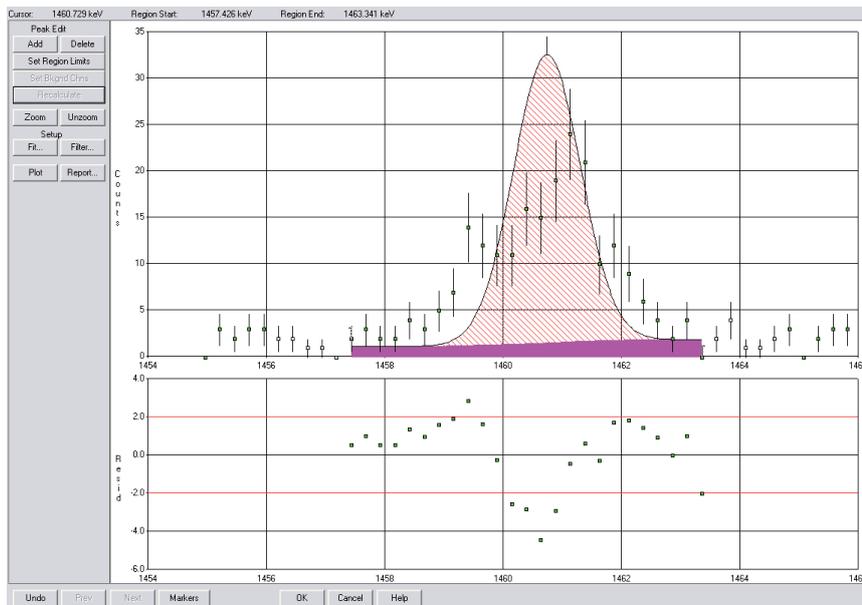


Figure 4.1: Peak 42, as marked by the old routine.



Figure 4.2: Peak 41, marked correctly by the new routine.

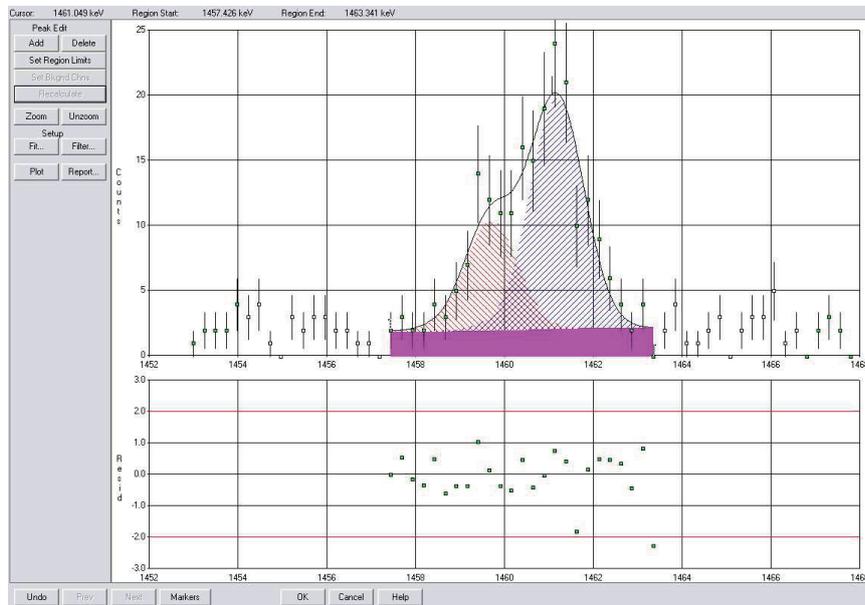


Figure 4.3: Peak 41 and 43, marked correctly by the user.

This peak is not listed in the nuclide library used during the identification because it is a big peaks that should be found automatically. The old routine, too, is not able to distinguish the two peaks and marks only one wrong entry, near the weighted centre of the multiplet. The user always has to add a peak.

4.1.2 Peak Identification

The results of the Peak Identification step executed with the two routines are perfectly comparable, as shown in tables 4.3, 4.4, 4.5, 4.6.

Peak Nr.	Energy (keV)	Uncertainty (%)
23	510.96	33.18

Table 4.3: Peaks not identified by the old routine.

Peak Nr.	Energy (keV)	Uncertainty (%)
5	74.85	4.90
6	77.14	2.74
23	510.96	33.18

Table 4.4: Peaks not identified by the new routine.

The new routine is however more sensitive and also shows details about the peaks not identified. The user can use the values in the “Ratio” column to check if the peak failed identification is caused by interference of more nuclides. This is the case of this sample, where both peaks 5 and 6 are the sum of two smaller peaks: the sum of the “Ratio” values is about the unity.

4.1.3 Nuclide Identification

The output of this step is shown in table 4.7. The activities of most nuclides, calculated with the new analysis routine, are exactly the same or within 5% of the reference value, obtained with the old routine. The values are always within the uncertainties, while the uncertainty values slightly change when switching from the old to the new routine. This behaviour is related to the differences of the peaks energies, resulting from the peak locate step. The final effect on the values is an increase of the uncertainty associated to some nuclides and a decrease of the uncertainty associated to other nuclides. Tests with several samples rarely show a global significant increase, while a slight global decrease is more often achieved. If needed, the user can go directly to the *Interactive Peak Fit* to change the positions of the peaks in the spectrum.

Matching Nuclide	Calculated Peak Activity (Bq/g)	Calculated Peak Activity Uncertainty (Bq/g)	Nuclide Activity (Bq/g)	Nuclide Activity Uncertainty (Bq/g)	Ratio of the Activities
Th-234	5.09E-01	5.70E-02	1.15E-03	1.29E-03	0.00
Pb-212	1.35E-02	2.66E-03	4.74E-03	1.18E-03	0.35
Pb-214	2.18E-02	4.29E-03	1.43E-02	2.29E-03	0.66
Tl-208	4.11E-02	8.10E-03	1.82E-03	6.95E-04	0.04
U-235	2.33E00	4.61E-01	1.99E-03	8.16E-04	0.00

Table 4.5: Unidentified peaks: details of peak nr. 5.

Matching Nuclide	Calculated Peak Activity (Bq/g)	Calculated Peak Activity Uncertainty (Bq/g)	Nuclide Activity (Bq/g)	Nuclide Activity Uncertainty (Bq/g)	Ratio of the Activities
Rn-219	5.09E-01	5.70E-02	1.15E-03	1.29E-03	0.00
Bi-214	6.65E-01	7.44E-02	1.36E-02	2.06E-03	0.02
Pb-212	1.54E-02	1.73E-03	4.74E-03	1.18E-03	0.31
Pb-214	2.50E-02	2.80E-03	1.43E-02	2.29E-03	0.57

Table 4.6: Unidentified peaks: details of peak nr. 6.

Nuclide Name	Old algorithm		New algorithm	
	Activity (Bq/g)	Uncertainty (%)	Activity (Bq/g)	Uncertainty (%)
Tl-208	1.52E-03	10.0	1.82E-03	9.53
Pb-210	3.30E-02	17.2	3.01E-02	20.3
Bi-212	5.88E-03	21.2	5.88E-03	21.3
Pb-212	4.44E-03	6.27	4.74E-03	6.22
Bi-214	1.37E-02	3.78	1.36E-02	3.78
Pb-214	1.41E-02	4.06	1.43E-02	4.00
Rn-219	1.00E-03	31.6	1.15E-03	28.2
Ra-224	3.77E-03	31.7	3.85E-03	27.6
Ra-226	3.24E-02	10.1	3.24E-02	10.2
Ac-228	4.44E-03	7.75	4.42E-03	6.70
Th-232	3.13E-01	17.6	3.13E-01	17.5
Th-234	1.71E-02	22.2	1.71E-02	22.2
U-235	1.99E-03	10.3	1.99E-03	10.3

Table 4.7: Comparison of the results of the nuclide identification and activity calculation step, executed with the old and the new routines on a low-activity sample.

4.2 High Activity Sample

4.2.1 Peak Locate and Peak Identification

Both routines were able to detect a significantly higher number of peaks in the high-activity sample, thanks to the reduced statistical uncertainty: the first sample, analysed in the previous section, has peaks with less than 1,000 counts, while this high-activity sample has the biggest peaks of the order of magnitude of 128,000 counts.

The total number of peaks found by the new routine was 138, while the old routine detected 117 peaks, so I report in table 4.8 only a summary of the changes I made while checking both spectra.

Peaks	Old routine	New routine
Detected	117	138
Added	15	9
Deleted	0	10
Not added	2	0
Not recognised	3	0
Total	132	137

Table 4.8: Summary of the peak locate step on a high-activity sample.

The results of the peak locate step depend on the routine used. The new routine was able to mark automatically more peaks, but some of them were then deleted during the manual spectrum check because considered as false positives.

The old routine, having identified less peaks automatically, required a great amount of user interaction to correct the spectrum fitting, but it was still impossible to obtain the same results. During the manual cross-check with the results of the other routine, two types of peaks were skipped and not added at all: peaks impossible to recognise as such by only looking at the spectrum and peaks impossible to place in the correct position. The latter problem is related to the algorithm the *Interactive Peak Fit* software uses to position peaks in multiplets. The algorithm does not allow a fine control of the position of the new peak, so a peak could get placed in a position completely different from the specified one (quite a rare situation anyway: it happened only once). During the test of this sample I skipped peaks placed more than 1 keV apart of the specified position.

Just like in the previous sample, the new peak identification step reports an higher number of unidentified peaks compared to the standard routine: 26 versus 14 peaks. Among the additional 12 peaks, 2 are marked as escape peaks and 5 can be considered as interference peaks. The remaining 5 peaks cannot be immediately associated to existing energy lines or identified nuclides: the user has to better study the fitting and/or keep track of them in the final report.

4.2.2 Nuclide Identification

This step produces different results depending on the routine used, because the list of peaks found in the spectrum is different. Most of the additional peaks found by the new peak locate routine are, however, not used for the nuclide identification: given the low emission probability and the high uncertainty, they were not included in the nuclide library used during the nuclide identification.

The comparison of the results is shown in table 4.9. They fall always within the uncertainties, as in the previous sample, especially if we consider that the table reports the raw uncertainty values: during the normal analyses, a multiplier of 1.96 is applied to them, so the tolerances are almost doubled.

Thanks to the improved peak localisation and to the increased number of peaks recognised in the spectrum, the new software was also able to identify two more nuclides, albeit with high uncertainties (14-18%).

Nuclide Name	Old algorithm		New algorithm	
	Activity (Bq/g)	Uncertainty (%)	Activity (Bq/g)	Uncertainty (%)
Tl-208	2.50E+00	2.72	2.51E+00	2.71
Pb-211			2.48E-01	14.3
Bi-212	7.09E+00	2.70	7.40E+00	3.10
Pb-212	6.52E+00	4.17	6.97E+00	4.21
Bi-214	4.19E+00	1.98	4.15E+00	1.88
Pb-214	4.03E+00	3.48	4.31E+00	3.48
Rn-219	1.62E-01	12.0	1.52E-01	15.1
Ra-223	2.61E-01	11.5	2.93E-01	11.9
Ra-224	7.08E+00	5.88	7.22E+00	5.87
Ra-226	5.96E+00	16.6	6.03E+00	15.7
Ac-228	7.18E+00	2.14	7.34E+00	2.16
Pa-231			3.29E-01	17.3
Th-232	4.98E+01	9.59	4.98E+01	9.58
Pa-234m	6.14E+00	8.64	6.14E+00	8.64
Th-234	2.72E+00	16.7	2.72E+00	16.7
U-235	1.03E-01	51.0	9.85E-02	50.2

Table 4.9: Comparison of the results of the nuclide identification and activity calculation step, executed with the old and the new routines on a high-activity sample.

Chapter 5

Conclusions

The results obtained with the software I developed have been checked against the previous routine and the match is good. Slight differences are inevitable (for example, the peak identification routine is more sensitive and leaves more peaks as unidentified), but the nuclides activities are well within uncertainties.

The software is, therefore, already in use in the laboratory and its effectiveness in reducing the time required per analysis has been verified: a complete analysis can now be completed in far less time and the quality of the results equals or surpasses the old ones.

After a first test period, the users of this new software were asked to list the most important and useful improvements they experienced. They resulted to be:

- the automatic peak identification: the user does not have to execute manually the *Nuclide Identification with Interference Correction* algorithm two times and to change manually the library used, but everything is done by the software. Furthermore, the user has more knowledge about the unidentified peaks, thanks to the different peak activity calculations, one per possible match;
- the availability of details about the peaks used by the nuclide identification algorithm and their respective data (activity, uncertainty, level of detection);
- the complete knowledge of which peaks have been found and used for the nuclide identification and which peaks have not been found in the sample, even if there is a matching energy line in the nuclide library;
- the automatic Uranium-238-to-Uranium-235 ratio calculation, able to adapt to the possibility of a missing nuclide too;
- the graphical display of the decay chains activities and the ability to change on-the-fly the uncertainty factor, to account for greater tolerances in the measurement;
- the ability to execute the Excel customer report with only one click, instead of having to copy and paste manually the results found in an external text file

and, of course, the generalised reduction in the time required per analysis.

Possible Future Improvements

Concerning the second point of the previous list (the availability of detailed data about each peak used for the nuclide identification), an additional note must be made. As shown in the flow chart of figure 3.1, the nuclide identification step should be connected to a “LACE analysis” step, but nothing has been described under that name all throughout this thesis. *LACE* is the acronym for “Line Activity Consistency Evaluation” and its purpose is to check if peaks belonging to the same nuclide and used for the activity calculations have comparable activities even when they are spread through all the spectrum.

This analysis, initially planned for inclusion, has been removed from the software because of two different causes. The first one is technical: the “LACE” analysis module from the Canberra libraries did not work under Visual Basic .Net. The second reason for the removal of this feature is functional: the “Nuclide identification” window, with the ability to show each peak activity, is a good replacement for the “LACE” analysis. As a future improvement, “LACE” analysis can be introduced again, to offer the user a graphical view of the relative peak area, scaled by the relative abundances.

Another possible future improvement is an automatic energy calibration of the detector. Energy shifts are, with analog electronics, very common and are connected to many external factors. The idea that could be developed and implemented in the software is the automatic re-calibration of the spectrum with the results of the nuclide detection: the software detects the nuclides contained in the sample and uses them as reference sources for the new energy calibration. This process could be executed with good precision and results because the energy calibration does not depend on the activities of the sources, only on the positions of the peaks. Once the instrument has been automatically re-calibrated, a new, more accurate peak locate can be executed again and the user-controlled analysis can start. This improvement is, however, much less useful with new digital electronic components, much less subject to energy shifts.

The last possible improvement I have in mind is the use of the new “Library Correlation NID” algorithm for the nuclide identification step. This new algorithm has been made available only in the latest software revision, obtained too late to be tested actively. The new algorithm should be able to identify better nuclides, with less false positives, but needs good energy, shape and efficiency calibrations, so maybe the automatic energy re-calibration should be implemented in advance.

Appendix A

Application code

Follows the code of the peak identification routine.

```

Private Sub identifyPeaks()
  On Error GoTo errHdlr
  Dim tempCursor As Windows.Forms.Cursor = MyBase.Cursor
  Windows.Forms.Cursor.Current = Cursors.WaitCursor

  Dim nuclides As New CanberraDataAccessLib.DataAccess
  ' the nuclide file seems to be always sorted by nuclide energy, the function can be faster
  If nuclides.IsOpen Then nuclides.Close()
  nuclides.Open(autoId, CanberraDataAccessLib.OpenMode.dReadOnly)

  seqAnalyzer.Analyze(CAMFile, 1, xmlLocation & "\profile" & activeProfNumber & "\
    nid.ASF", False, False, False, True, "", Nothing)
  changed = True
  loadedCamfile_lines = False
  loadCamfileArrays()

  camfile_unidPeaks = 0

  Dim i As Integer ' position in the camfile peaks list
  Dim j As Integer ' position in the library
  Dim k As Integer ' position in the list of found nuclides
  Dim m As Integer ' position in the array with identification details
  Dim peakEnergy As Single
  Dim peakUncert As Single
  Dim nuclides_lineEnergy As Single
  Dim nuclides_lines As Integer = nuclides.NumberOfRecords(CAM_CLS_NLINES)
  Dim nuclides_lineEnergyA(nuclides_lines) As Single
  Dim nuclides_NIndexA(nuclides_lines) As Integer
  Dim nuclides_name As String
  Dim nuclides_names As Integer = nuclides.NumberOfRecords(CAM_CLS_NUCL)
  ReDim nuclides_namesA(nuclides_names)
  Dim camfile_peaks As Integer = CAMFile.NumberOfRecords(CAM_CLS_PEAK)
  Dim camfile_nuclideActivity As Single
  Dim camfile_nuclideActivityUncert As Single
  Dim key_line_factor As Single

```

```

Dim camfile_unidActivity As Single
Dim camfile_unidActivityUncert As Single
Dim sigma_s_s, sigma_eff_eff As Single

Dim key_line As Integer

Dim k_max As Integer = 50

' shadows the library nuclide energies (big list)
For i = 1 To nuclides_lines
    nuclides_lineEnergyA(i) = CSng(nuclides.Param(CAM_F_NLENERGY, i))
Next
' shadows the library nuclide names (smaller list)
For i = 1 To nuclides_names
    nuclides_namesA(i) = CStr(nuclides.Param(CAM_T_NCLNAME, i))
Next
' shadows the library nuclide indexes that every energy line has (big as the first, linking
    each line with a nuclide name)
For i = 1 To nuclides_lines
    nuclides_NIndexA(i) = CInt(nuclides.Param(CAM_L_NLNUCL, i))
Next

' first check: if a peak is listed in the full nuclides library and belongs to one
' of the identified nuclides, it's considered identified and it's not shown
ReDim camfile_unidPeaksEnergyA(k_max)
ReDim camfile_unidPeaksEUncertA(k_max)
ReDim camfile_unidPeaksAreaA(k_max)
ReDim camfile_unidPeaksAUncertA(k_max)
ReDim camfile_unidPeaksEscapeA(k_max)

ReDim camfile_tentativePeakIdent(5, k_max * 5)
Dim lastm As Integer = 1

For i = 1 To camfile_peaks
    If camfile_peakIsIdentified(i) = 1 Then Continue For
    ' lastm is used to go back in the list of tentativePeakIdent
    lastm = m

    j = 1
    peakEnergy = camfile_peaksEnergyA(i)
    peakUncert = tolerance + CSng(CAMFile.Param(CAM_F_PSDENERGY, i))

    ' if the energy of the peak is higher than the energy of the line listed in the library, go to
    next line
    While (peakEnergy - peakUncert) > nuclides_lineEnergyA(j) And j < nuclides_lines
        j += 1
    End While
    ' if the peak energy and the line energy (with uncertainties) are overlapping, check for
    nuclide matching
    While (peakEnergy - peakUncert) <= nuclides_lineEnergyA(j) And (peakEnergy +
        peakUncert) >= nuclides_lineEnergyA(j)
        nuclides_lineEnergy = nuclides_lineEnergyA(j)
        nuclides_name = nuclides_namesA(nuclides_NIndexA(j))

```

```

' for each of the identified nuclides
For k = 1 To camfile_names
  ' these two lines check for identification: is the line nuclide in the list of nuclides
    found in the sample?
  ' is also marked as identified? (not all nuclides listed in the sample are identified)
  If StrComp(camfile_namesA(k), nuclides_name, CompareMethod.Text) <> 0
    Then Continue For
  If camfile_nuclideIdentifiedA(k) = 0 Then Continue For

  ' check for correct activity
  ' reads activity of the nuclide
  camfile_nuclideActivity = camfile_nuclideActivityA(k)
  camfile_nuclideActivityUncert = sigmaErr * camfile_nuclideActivityUncertA(k)

  ' calculates activity of the unid. peak as it was the specified nuclide
  camfile_unidActivity = camfile_peaksAreaA(i) / CSng(nuclides.Param(
    CAM_F_NLABUN, j)) * 100 / _
    camfile_peaksEfficiencyA(i) / CSng(CAMFile.Param(CAM_F_SQUANT, i)) / _
    37000 / CSng(CAMFile.Param(CAM_X_ELIVE))

  ' calculates uncertainty
  sigma_s_s = camfile_peaksAreaUncertA(i) / camfile_peaksAreaA(i)
  sigma_eff_eff = CSng((CSng(CAMFile.Param(CAM_F_PSEFERR, i)) /
    camfile_peaksEfficiencyA(i)) ^ 2)
  camfile_unidActivityUncert = sigmaErr * (CSng(Math.Sqrt((CSng(CAMFile.Param
    (CAM_F_SSYSERR)) / 100) ^ 2 + _
    sigma_eff_eff ^ 2 + sigma_s_s ^ 2)) + CSng(CAMFile.Param(CAM_F_SSYSTEMERR)
    ) * camfile_unidActivity / 100)

  ' compares activities with uncertainties
  If (camfile_nuclideActivity + camfile_nuclideActivityUncert) > (
    camfile_unidActivity * (1 - camfile_unidActivityUncert)) And _
    (camfile_nuclideActivity - camfile_nuclideActivityUncert) < (
    camfile_unidActivity * (1 + camfile_unidActivityUncert)) Then
    ' the nuclide is identified
    m = lastm
    GoTo Next_iteration
  End If

  ' if not identified, add the peak activity and the nuclide activity to the list of possible
    matches
  ' They have matching energies but not matching activities
  ' (list on the bottom of the window)
  camfile_tentativePeakIdent(0, m) = camfile_unidPeaks + 1
  camfile_tentativePeakIdent(1, m) = nuclides_NIndexA(j)
  camfile_tentativePeakIdent(2, m) = camfile_unidActivity * 37000
  camfile_tentativePeakIdent(3, m) = camfile_unidActivityUncert *
    camfile_unidActivity * 37000
  camfile_tentativePeakIdent(4, m) = camfile_nuclideActivity * 37000
  camfile_tentativePeakIdent(5, m) = camfile_nuclideActivityUncert * 37000
  m += 1
  If m = UBound(camfile_tentativePeakIdent, 2) Then ReDim Preserve
    camfile_tentativePeakIdent(5, 2 * m)

```

```

        ' now go back and check for escape peaks
        Exit For
    Next k

    j += 1
End While

' add the peak to the list of unidentified peaks
camfile_unidPeaks += 1
camfile_unidPeaksEnergyA(camfile_unidPeaks) = peakEnergy
camfile_unidPeaksEUncertA(camfile_unidPeaks) = peakUncert
camfile_unidPeaksAreaA(camfile_unidPeaks) = camfile_peaksAreaA(i)
camfile_unidPeaksAUncertA(camfile_unidPeaks) = camfile_peaksAreaUncertA(i) /
    camfile_unidPeaksAreaA(camfile_unidPeaks) * 100
If camfile_unidPeaks = k_max Then
    k_max = k_max * 2
    ReDim Preserve camfile_unidPeaksEnergyA(k_max)
    ReDim Preserve camfile_unidPeaksEUncertA(k_max)
    ReDim Preserve camfile_unidPeaksAreaA(k_max)
    ReDim Preserve camfile_unidPeaksAUncertA(k_max)
    ReDim Preserve camfile_unidPeaksEscapeA(k_max)
    ReDim Preserve camfile_tentativePeakIdent(5, k_max)
End If

' second check: escape peaks
peakEnergy = peakEnergy + 511
For j = (i + 1) To camfile_peaks
    If camfile_peakIsIdentified(j) = 0 Then Continue For

    If (camfile_peaksEnergyA(j) + tolerance) > (peakEnergy - tolerance) And _
    (camfile_peaksEnergyA(j) - tolerance) < (peakEnergy + tolerance) Then
        ' it's an escape peak (first type: first escape peak, found in the sample)
        camfile_unidPeaksEscapeA(camfile_unidPeaks) = 1
        Exit For
    End If
    If (camfile_peaksEnergyA(j) - tolerance) > (peakEnergy + tolerance) Then Exit For
Next j
If j >= camfile_peaks Then ' since we reached the last peak without finding anything, I
    will look into the library, from the top
    j = nuclides_lines
    While nuclides_lineEnergyA(j) > camfile_peaksEnergyA(camfile_peaks)
        If nuclides_lineEnergyA(j) > (peakEnergy - tolerance) And nuclides_lineEnergyA(j)
        < (peakEnergy + tolerance) Then
            ' it's an escape peak (second type: first escape peak, found in the library)
            camfile_unidPeaksEscapeA(camfile_unidPeaks) += 2
        Exit While
    End If
    j -= 1
End While
End If

' second escape peak

```

```

peakEnergy = peakEnergy + 511
For j = (i + 1) To camfile_peaks
  If camfile_peakIsIdentified(j) = 0 Then Continue For

  If (camfile_peaksEnergyA(j) + tolerance) > (peakEnergy - tolerance) And _
  (camfile_peaksEnergyA(j) - tolerance) < (peakEnergy + tolerance) Then
    ' it's an escape peak (third type: second escape peak, found in the sample)
    camfile_unidPeaksEscapeA(camfile_unidPeaks) += 4
  Exit For
End If
If (camfile_peaksEnergyA(j) - tolerance) > (peakEnergy + tolerance) Then Exit For
Next j
If j >= camfile_peaks Then ' we reached the last peak without finding anything, now I
  will look into the library, from the top
  j = nuclides_lines
  While nuclides_lineEnergyA(j) > camfile_peaksEnergyA(camfile_peaks)
    If nuclides_lineEnergyA(j) > (peakEnergy - tolerance) And nuclides_lineEnergyA(j)
    < (peakEnergy + tolerance) Then
      ' it's an escape peak (fourth type: second escape peak, found in the library)
      camfile_unidPeaksEscapeA(camfile_unidPeaks) += 8
    Exit While
  End If
  j -= 1
End While
End If

Next_iteration:
Next i
ReDim Preserve camfile_tentativePeakIdent(5, m)

nuclides.Close()

Windows.Forms.Cursor.Current = tempCursor
Exit Sub

errHdlr:
  Windows.Forms.Cursor.Current = tempCursor
  ProcessError()
End Sub

```

Appendix B

Gamma Analysis User Guide

B.1 Overview

The *Gamma Analysis* software is designed to speed up the analysis of NORM samples. The result is obtained by showing at the same time all the informations the user needs, while in the previous software the user had to switch back and forth between many applications and also do calculations by hand. Not only, a new peak location sequence and peak matching algorithm have been developed. The user interface is designed to be straightforward and to guide the user in the steps needed to obtain the result.

The main program input is the CAM file produced by the measurement software, no pre-processing is required, the final output can be a printed report, a screen report, or an Excel file ready for the print.

B.2 Installation

The application does not require an installation procedure but only to be copied in a local drive. Network drives are not currently supported. Program settings are stored in an XML file located in the “settings” subfolder. Each saved profile is made of ASF files located in subdirectorues of the “settings” folder.

B.3 User Interface Elements Description

B.3.1 Main Window

While no files are open, only the “Load...” button and the Report viewer buttons are enabled. The user can load a CAM file with the “Load...” button or from the File menu.

The “File” menu contains the following items:

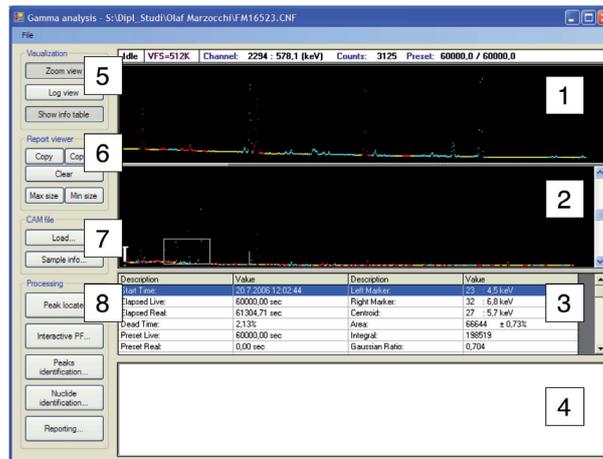


Figure B.1: Main software window.

Load... open a windows to select a file to load. The default folder depends on the local Canberra installation.

Save saves the open file overwriting the old one.

Save as... open a window to select where to save the file.

Close closes the open file.

Print... prints the contents of the Report viewer after a confirmation window.

Preferences opens the Preferences window.

Exit exit from the application, asking to save changes if needed.

The main window is made of eight sections:

Section 1 zoomed view of the section 2, activated with the first button of the section 5;

Section 2 main view of the spectrum, always visible, empty when no files are loaded;

Section 3 detailed information on the sample, activated with the third button of section 5;

Section 4 report viewer;

Section 5 buttons affecting the visualization of the spectrum. The first button activates the zoom view (section 1), the second button affects sections 1 and 2 and changes the vertical scale, from linear to logarithmic, to better show peaks in the high-counts regions. The third button activated the section 3;

Section 6 buttons affecting the report viewer. The first two buttons copy the selected text and all the content of the report viewer to the clipboard, the third button clears the report viewer, the last two buttons make the report viewer as big as possible or as small as possible. These last two behaviours can be obtained by dragging the horizontal bar between sections 3 and 4;

Section 7 buttons to load a CAM file and to show the Information window of image B.2;

Section 8 the buttons represent the steps needed to complete an analysis. The steps should be executed from the first button to the last button. The steps are described below.

B.3.2 Information Window

Figure B.2: Sample information window.

The Informations window show the main attributes of the sample and let the user change them. Fields with dark background can either be read only or be automatically updated after the corresponding “Browse...” button is pressed. The “Browse...” button of each field is used to load data from external files, such as another energy calibration, another geometry and another GIS file.

B.3.3 Peak Locate

The peak locate step executes the *Unidentified 2nd Difference* and the *Library (Simple)*. The settings for these two algorithms can be seen and changed in the Preferences window (image B.8). After the peak location, area subtraction and efficiency correction are performed on the results. The user is asked for confirmation before the peak locate is executed, because the peak locate step overwrites the previous peaks saved in the

CAM file. A window with a notice of the successful execution is shown after the peak locate step.

B.3.4 Interactive Peak Fit Window

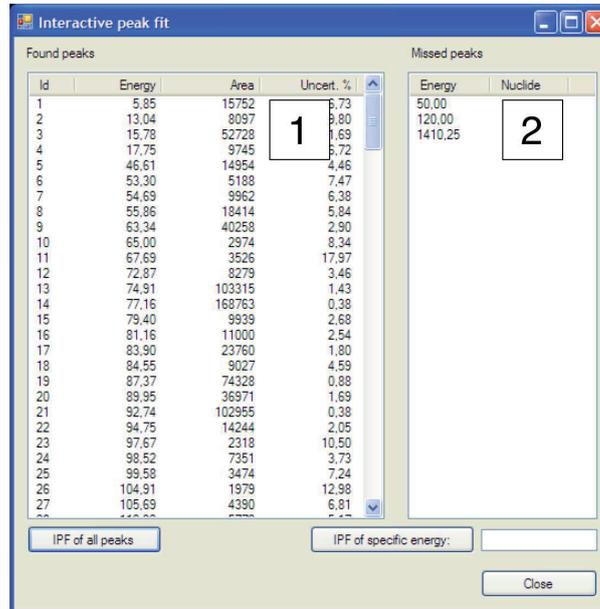


Figure B.3: Interactive Peak Fit window.

The list 1 on the left shows energy, area and area uncertainty, the list 2 on the right is a user-defined list. This list is useful to list peaks that the software is not usually able to find with its algorithms. The “IPF of all peaks” opens the external Interactive Peak Fit program, where the user can check the correct fitting, add and delete peaks. The “IPF of specific energy” button is used to execute the external program and to go directly to the energy specified in the text box. To analyze a specific peak the user can click on it, so that its energy appears in the text box, or double click on the peak in the list. Once the external program is closed, area correction and efficiency correction are calculated again and the list is updated.

B.3.5 Peak Identification Window

The peak matching is done while the window is loading and the results are shown in the list 1. The list 2 shows possible choices for matching the unidentified peaks selected in the list 1. In the list 2 only nuclides identified in the sample are listed, a peak that could belong to a nuclide not found in the sample won't show up in the list. The user can double click on one of the unidentified peaks or click on the *Interactive Peak Fit* button to go directly to the peak selected in the first list and check its fitting

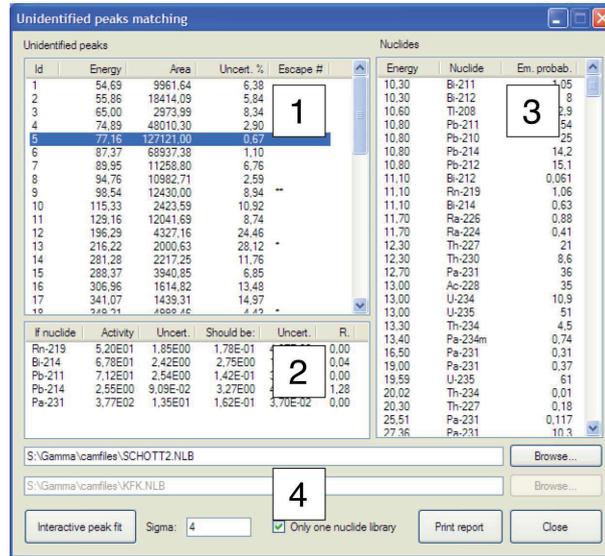


Figure B.4: Peak Identification window.

and boundaries. The list 3 is made of all the energy lines contained in the libraries shown in the section 4 or only of the first library if the appropriate option is checked. Duplicate lines found in the libraries are shown only once. The user can double click on an energy line to go to the external Interactive peak fit of the line energy. The error multiplication factor is set with the textbox of section 4, the list 1 will be immediately updated after a change.

B.3.6 Nuclide Identification Window

The list 1 shows the results of the *Nuclide Identification with Interference Correction*. Activities, activities uncertainties and the level of detection are shown. The list 2 contains the details for the selected nuclide. Energy, calculated activity, uncertainty and emission probability are shown for each line. Lines not found in the spectrum have “n.a.” as activity and uncertainty. The three graphs 3 show graphically the equilibrium of the decay chains. Each graph represents a decay chain, where activities and uncertainties are shown in black and the levels of detection are marked in red. Missing nuclides leave an empty space, but their level of detection is shown. The user can go to the interactive peak fitting by double clicking on an energy line or by using the “Peak IPF” button.

B.3.7 Report Window

The window let the user easily do three different reports without the need to change settings. For each report, the user can choose the template, the section of the template

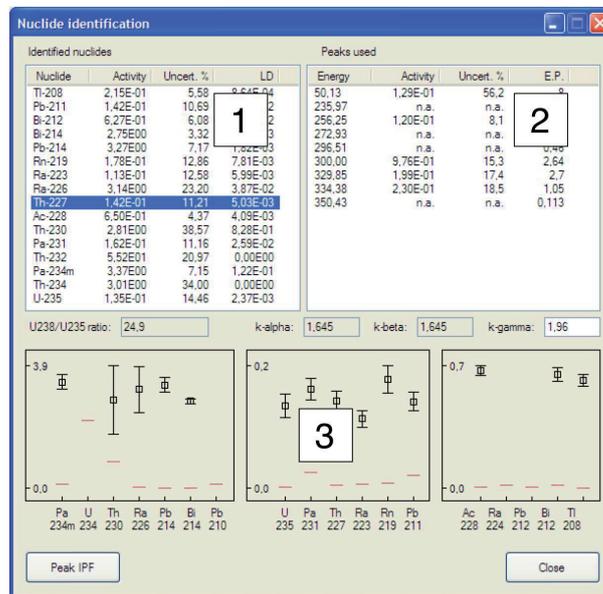


Figure B.5: Nuclide Identification window.

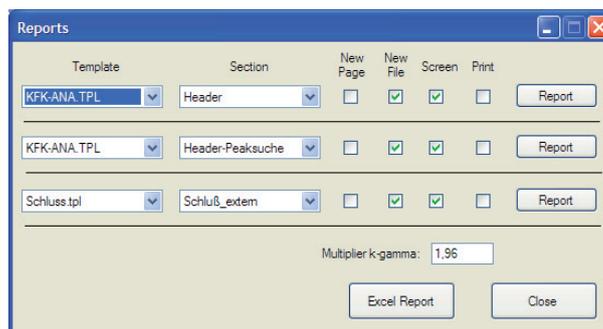


Figure B.6: Reports window.

and the destination of the report. The report is executed upon pressing of the “Report” button. The error multiplier is set in the text field on the lower part of the window and the default settings are set in the preferences.

B.3.8 Preferences Window

The Preferences window is divided in two parts. On the left there are buttons to select the different sections of the preferences, while the selected group of preferences is shown on the right.

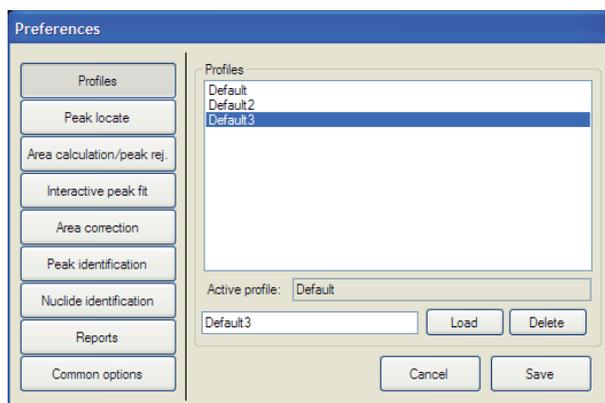


Figure B.7: Preferences window: Profiles.

Profiles: in this section the user can define multiple profiles, so that sets of preferences can be saved for later use. The active profile is shown in the “Active profile” text box, to load another one the user has to select a profile from the list and then to press the “Load” button. The “Delete” button, just like “Load”, operates on the profile shown in the white text box on its left. The first profile cannot be deleted. The “Save” button saves the current set of preferences into the profile written in the white text box. The name of the profile can contain spaces. If the white text box is empty, the preferences are saved over the active profile. This is useful if the user wants to change only one setting: load a profile, change the setting, set the name for the new profile and then press the save button. The “Cancel” button is used to undo all the changes.

Peak locate: in this section, preferences affecting the peak locate algorithms are shown. The “Library locate” refers to the nuclide library used for the *Library (Simple)* step. When only the *Unidentified 2nd Difference* algorithm is preferred, simply choose here an empty nuclide library.

Area calculation/peak rejection: this section shows the preferences affecting the area calculation step.

Interactive peak fit: in this section the user can set the text file containing the list of “usually missed peaks”. The file has a simple syntax and can be customized by hand:

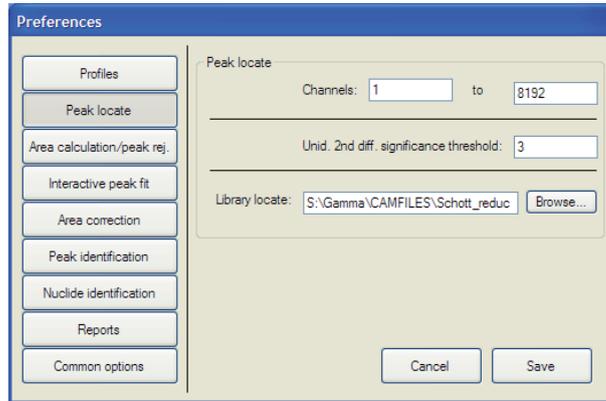


Figure B.8: Preferences window: Peak Locate.

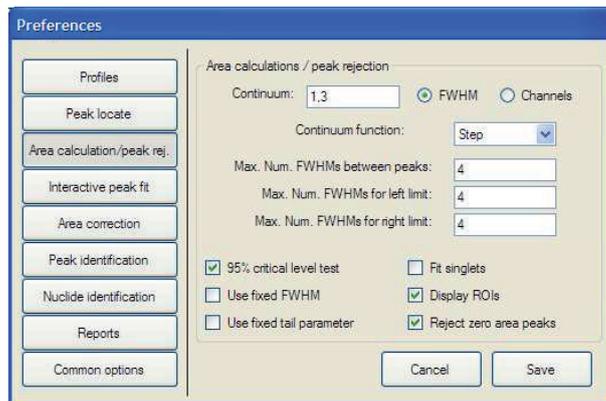


Figure B.9: Preferences window: Area Calculation/Peak Rejection.

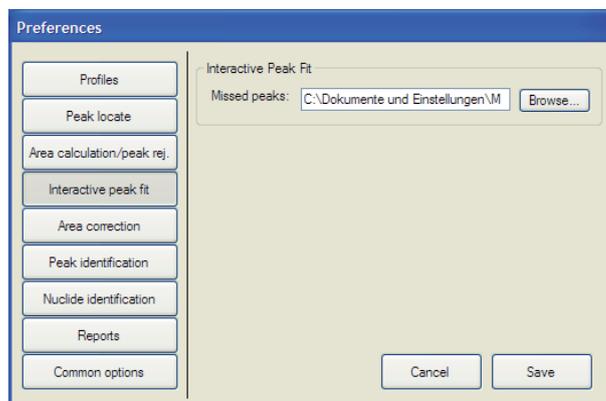


Figure B.10: Preferences window: Interactive Peak Fit.

each line must start with either an “N-” or a “E-”. In the first case, the text following that prefix will be considered as a nuclide name, in the second case the text will be considered an energy (the user must use the comma or the dot as decimal separator depending on the language of the operating system). Each missed peak is made of two lines in this file: first a line starting with “N-” and containing the nuclide name, then a line starting with “E-” containing the energy. If there are more peaks of the same nuclide, only one “N-” line for them is enough: each energy value will be associated with the last nuclide defined.

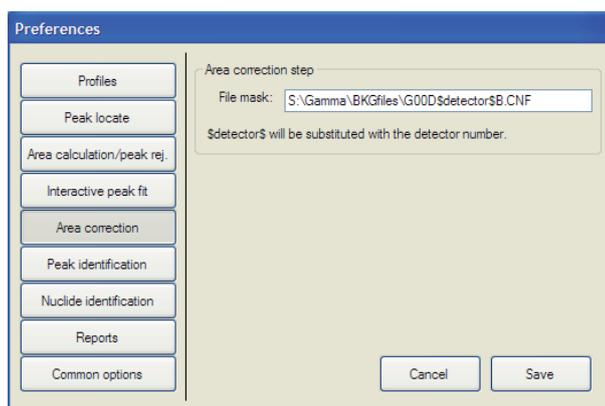


Figure B.11: Preferences window: Area Correction.

Area correction: this section is used to define the file mask for the correction step (background subtraction). The user can set the file name and location in the text box, then has to replace the detector number with “\$detector\$”. The software will substitute the string “\$detector\$” with the actual detector number, depending on the sample.

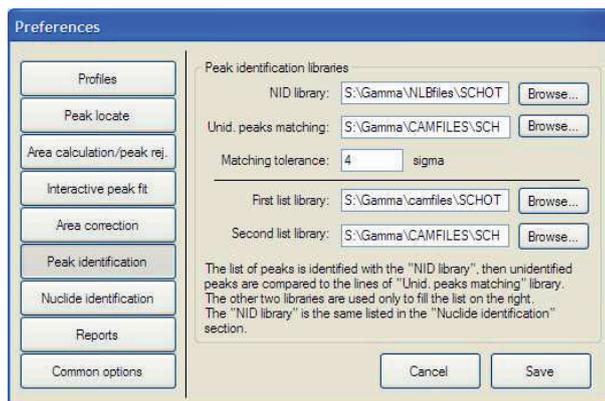


Figure B.12: Preferences window: Peak Identification.

Peak identification: in this section are listed the libraries used in the peak identification step. The first library is used for the nuclide identification (useful to know all the nuclides found in the spectrum), the second library is used by the identification al-

gorithm, to match peaks left unidentified by the first algorithm. The other two libraries are NOT used for the identification, but only to compile the list of nuclides shown on the right in image B.4.

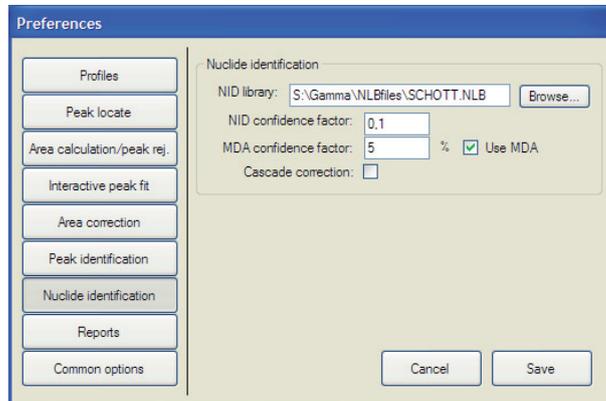


Figure B.13: Preferences window: Nuclide Identification.

Nuclide identification: this section lists the parameters for the Nuclide identification step. The library listed is the same of the first field of the previous section, here the user can set other parameters affecting the identification.

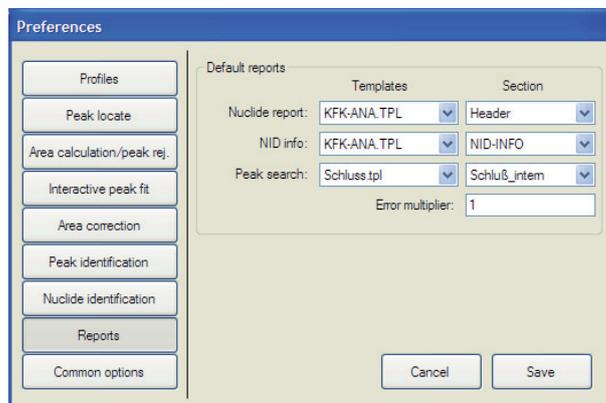


Figure B.14: Preferences window: Reports.

Reports: in this section the user can set the default reports.

Common options: this section is used to set the value of the energy tolerance, used for every step of the program except for the interactive peak fit, where no matching is done.

B.4 Usage Guide

The first step is to set all the preferences and save them as profile, with a descriptive name useful to remember the sample type and other informations. The user should

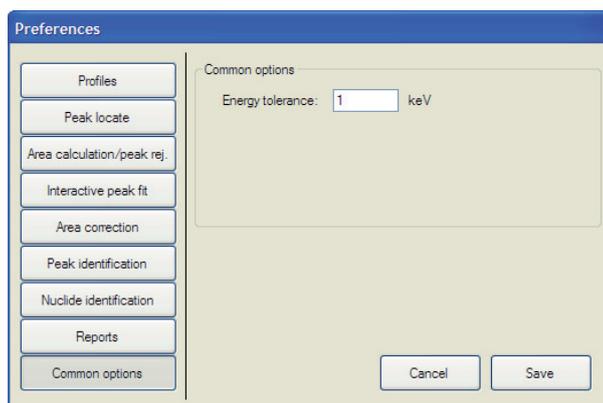


Figure B.15: Preferences window: Common Options.

then customize the file with the list of missed peaks (each profile can have a different file). Once the profile has been saved, the next time the user will only have to load it without changing any parameter. The next step is the peak locate. The user is asked for confirmation before the step is executed, after the step has been completed the new peaks are shown graphically in the MCA display (sections 1, 2, 3 of image B.1). The user can now check for the correct peak fitting by selecting *Interactive Peak Fit* and then “IPF of all peaks”. The external *Interactive Peak Fit* application from Canberra will be executed. The execution of the external application always requires the CAM file to be saved to disk (it happens in every window of the program when the external application is executed), so all the changes applied (peak location, nuclide identification, sample properties) are saved to disk, overwriting the previous CAM file. The check for the correct fitting of the peaks can be usually skipped, since the user will be able in each step of the analysis to go back to the peak fitting. This check is useful mostly to remove unneeded peaks, peaks that the software marks only to better fit the spectrum. This behaviour comes from the *Library (Simple)* step, so it can be tuned by changing the number of peaks in the *Library (Simple)* library (image B.8). The next step is the peak identification (image B.4). In this step the software first identifies the nuclides in the sample, then reads the lists of peaks not assigned to any nuclide. This list is then checked, in order to try to match each peak with a energy lines from a nuclide library. Peaks not matched are also checked as escape peaks and marked accordingly. This procedure is useful because the first nuclide identification uses a nuclide library with few lines, so that no nuclides are missed, then the second matching can compare unidentified peaks with a much bigger library. The user can click on a peak of the section 1 of this window to view, in the section 2, a list of possible matching nuclides. None of them has an activity equal (within uncertainty) to the activity of the peak (because the peak would have been marked as identified and wouldnt be in the list), but all the choices are shown so that the user can check directly.

More specifically, every line of the section 2 shows in the second column the activity of the peak when it is assumed to belong to the nuclide listed in the first column and, in the fourth column, the activity of that nuclide. The ratio of these two activities is shown in the last column, so that the user can check the sum of the ratios to identify peaks with interference. On the right side of the window there is a list of the energy lines listed in the specified libraries. This list can be used to know which nuclides have an energy similar to the energy of a peak or to identify peaks from non-natural occurring radionuclides. After peak identification, the user can close the window and then go to the nuclide identification step. The new window shows the nuclides activities, both numerically and graphically. The user can change the tolerance value k -gamma, the same error multiplier of the report window, and all the uncertainties are immediately updated.. When the user clicks on a nuclide, a list of the lines used for its identification is shown on the right (section 2 of image B.5). In this list, lines found in the library but not in the sample have “n.a.” as activity and uncertainty. The emission probability of a line is always shown in the last column. A double click on a energy line of this list opens the external Interactive peak fit application. Once all the analysis steps have been completed, the user can print to screen or to paper the reports from the “Report window” (image B.6). The user can also print only the content of the main reporter (section 4 of image B.1) by selecting the menu item “Print...”.

Bibliography

- [1] K. Debertin and R. G. Helmer. *Gamma- and X-Ray spectrometry with semiconductor detectors*, Elsevier Science Publishers, 1988.
- [2] Peter J. Mohr and Barry N. Taylor. CODATA recommended values of the fundamental physical constants: 2002. *Reviews of Modern Physics*, 77(1):1, 2005. An in-depth discussion of how the CODATA constants were selected and determined. Available on <http://link.aps.org/abstract/RMP/v77/p1>.
- [3] J. A. Bearden and A. F. Burr. Reevaluation of X-Ray Atomic Energy Levels. *Rev. Mod. Phys.*, 39(125), 1967.
- [4] M. Cardona and L. Ley, Eds. *Photoemission in Solids I: General Principles*, Springer-Verlag, Berlin, 1978. With additional corrections.
- [5] NIST XCOM library. <http://physics.nist.gov/PhysRefData/Xcom/Text/XCOM.html>.
- [6] Ortec website. <http://www.ortec-online.com>.
- [7] Davisson, Charlotte Meaker and Evans, Robley D. Gamma-Ray Absorption Coefficients. *Rev. Mod. Phys.*, 24(2):79, Apr 1952.
- [8] Canberra Industries. *GenieTM2000 3.0 Customization Tools Manual*, chapter Genie Algorithms, pages 229361. 2004. Description of the algorithms used in the Genie-2K software.

Acknowledgements

This paper is the result of an effort lasted more than six months, made possible by the aid and support of many people. Among them, I wish to thank Prof. Mostacci and Prof. Urban, for the opportunity to study abroad they gave me, and Ing. Wilhelm and Ing. Kaminski, for welcoming and helping me throughout all my work in the HS-ÜM laboratory of the Forschungszentrum Karlsruhe.

A special thanks goes to my family, for the continuous support during my years of study in Bologna and abroad, to “Lo’p” and “Masa”, the other two members of the “Unità di produziù” (the group of people I studied with all the last five years), and to my good high-school friends I am still in touch with, Camillo and Giacomo.