Contextual Isotope Ranking Criteria for Peak Identification in Gamma Spectroscopy Using a Large Database

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Abstract—Isotope identification is a recurrent problem in γ-spectroscopy with high purity germanium detectors. In this work, new strategies are introduced to facilitate this type of analysis. Five criteria are used to identify the parent isotopes making a query on a large database of γ-lines from a multitude of isotopes producing an output list whose entries are sorted so that the γ-lines with the highest chance of being present in a sample are placed at the top. A metric to evaluate the performance of the different criteria is introduced and used to compare them. Two of the criteria are found to be superior than the others: one based on fuzzy logic, and another that makes use of the γ relative emission probabilities. A program called histoGe implements these criteria using a SQLite database containing the γ-lines of isotopes which was parsed from WWW Table of Radioactive Isotopes. histoGe is Free Software and is provided along with the database so they can be used to analyze spectra obtained with generic γ-ray detectors.

Index Terms—Gamma-ray spectroscopy, Heuristic algorithms, Isotope identification, Ranking

I. INTRODUCTION

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γ-spectroscopy commonly uses High Purity Germanium Detectors (HPGe) to acquire the energy spectra of samples with a varied isotopic composition. Analyzing and identifying the isotopes present in the sample is a challenging problem given the multitude of gamma lines from which to choose. Thus, computational tools are needed to analyze a spectrum and extract useful information [1], [2].

Many programs have been written to make these calculations in the past [3]–[10]. However, just a few provide the capability to identify isotopes from a given spectrum. Some previous attempts have been made to perform that identification using a relational database, for example: Hyperlab [11] has a functionality to make a “graphical iteration process” followed by a procedure to solve iteratively an “identification matrix”, however, no documentation about the method nor reports on its identification accuracy are provided for this software. GammaLab [5] uses a database called “NUCDATA” which includes information “for quick calculations for 408 radioisotopes” [11], however, it is limited in extension in comparison to other well-known databases [12]–[14] and it does not present a study about its accuracy and its real capability of identifying isotopes in a sample. ASPRO-NUC [15] has a wide set of spectral analysis tools: peak search, deconvolution, background line and simulation, spectrum smoothing, among others. It also has algorithms to identify peaks using a database of 45,000 γ-lines corresponding to 2200 radionuclides. However, the authors recognized that identification in this way “is hardly possible” and they opted to develop an “actual isotope library”. Sandia National Laboratories provide two programs for assisting with analyzing spectral information from nuclear radiation. InterSpec [16] provides multipeak fitting isotope identification capabilities as well as activity calculation considering shielding from a variety of materials. The other one is called Peak Map [17] which is written in C#, it considers a set of parameters such as distance from the mean and half-life penalization (among others) to assign a score to the γ-lines and sorts with respect to that before displaying the candidate γ-lines.

There are many areas in which γ-spectroscopy is applied, e.g., high energy physics research, environmental sciences, and food contamination. An important application lies in the control of radioactive materials crossing borders around the world, where there is a need to have instruments and methods.
to identify radioactive nuclei that are potentially harmful even in small amounts. While automated analysis for this problem is desirable, a study from 2007 concluded that a "secondary analysis of spectra by a trained spectroscopist is frequently necessary" to identify isotopes through their γ-lines [18]. Since then, new techniques have been explored to make automated isotope identification more reliable independently of the field of application, to mention a few: swarm optimization [19], Fischer linear discriminant analysis [20], Bayesian statistics approach [21], [22], neural networks (NN) [23]–[29], hybrid fuzzy-genetic algorithms [30]. Some of these methods have been used to perform automated peak identification [31]. There are also developments, using the GEANT4 toolkit, for providing training data to machine learning models [32]. Herein, new methods have been proposed, implemented, and tested using the histoGe code [33], a Free Software (GNU Public License [34]) with many features that are described in its User’s Manual [35].

This work deals with the γ-line identification problem in a manner similar to what a search engine does when presenting the results of a query. It assigns a numerical value called “rank value” (RV) to each candidate γ-line that may explain the presence of the peak in a spectrum.

Five criteria to identify peaks are presented, some of them are based on simple counting while others use more complex calculations, e.g., Mamdani’s fuzzy inference system (FIS). Known γ spectra were identified using the proposed criteria and their performance was evaluated using an ad hoc metric.

This work has been developed as an effort to support the research and educational activities that will be carried out at the Laboratorio Subterrâneo de Mineral del Chico (LABChico), which will be located inside a decommissioned silver mine at the Comarca Minera, Hidalgo, México, inside the UNESCO Global Geopark [36]. LABChico will host HPGe to conduct studies of low radioactivity in water, soils, and products intended for human consumption, aiming to develop techniques to signal the presence of lead in drinking water. At the same time, it will serve as a training hub for students and researchers interested in radiation detectors, techniques for particle and astroparticle physics experiments, geology and mine engineering, among other areas. The histoGe computational software was developed as an effort to facilitate γ-ray spectroscopy and to identify isotopes from recorded spectra inside the laboratory. Technical details about its implementation and capabilities, how to use it, and its database of histoGe are described in the user’s manual [35].

This paper is organized as follows: in section II the methodology used to identify isotopes and a brief explanation of the operation of the program and some key concepts are presented; in section III five criteria to find the most suitable γ-lines that can be responsible for the peaks observed in the spectra are described; in section IV the experimental setup is described; in section V four cases are studied: one is an example of how the general method works, the second one analyzed the spectra obtained with point-like radioactive sources through the Nuclear Sciences Institute HPGe (ICN-HPGe) detector, the third example is the analysis of spectra of some samples of rocks and water and the fourth example presents the identification of isotopes using a spectrum taken from the literature. The last section shows the conclusions of this work.

II. METHODOLOGICAL APPROACH

The histoGe software is written in Python 3 [37] and can run practically in any of the mainstream operating systems available nowadays. The basic process of line sorting used in histoGe is depicted in Figure 1. Energy query ranges are determined by either processing an experimental spectrum to find peaks, or by hand. histoGe uses a Savitzki-Golay filter [38] to smooth out spectra, detect the peaks and generate these query ranges. The width of the query ranges contains information about the resolution of the detector.

An info file [35] contains the energy query ranges associated with each peak. Once the file has been read, a query to the database is performed for each peak. As a result, a set of lists, one list per interval, are obtained from those queries. The lists contain information of all γ-lines that can be located inside the specified energy ranges. Once the RVs are calculated, each γ-line list is sorted and printed on the screen or stored in a text file (figure 1). The ranking operation calculates and assigns to each γ-line the RV used for sorting in descending order depending on the criterion (section III describes them in detail). This RV, or score, can be constructed so as to take into consideration global aspects of the spectrum, such as, for example, the possibility that a given isotope or decay chain may be responsible for several peaks, bringing context into the analysis. Once a RV has been assigned to all the γ-line candidates under the peaks of interest, they are sorted with respect to the other candidates within the same peak (locally), positioning the best candidates at the top of the list. Positions span from 0 to the number of the γ-lines found in the query minus one. A position close to 0 indicates a high preference for the γ-line to explain features present in the spectrum. Although all criteria need an info file, not all criteria require the spectrum. This general procedure is schematically represented in figure 2.

The ranking methods can be classified in three broad categories according the way the RV is calculated. Those where the RV is calculated for each γ-line individually without considering the other γ-lines of the same isotope found for the other peaks (III-A), those in which the RV is calculated for each isotope using all the γ-lines found in all peaks (III-B III-C) and (III-D) and those in which the RV takes into account all isotopes in the decay chains (III-E). Depending on the category to which a criterion belongs to, the RV is assigned to a γ-line, or to all γ-lines of an isotope, or to all γ-lines of all isotopes in a chain, correspondingly.

The whole operation is based on a local database (LDB) [39] (11 MB of disk space) that was constructed in part from the one accessible in [12]. Additional data was computed and added to have enough information to be used with the ranking criteria such as decay chains and normalized emission probabilities [35]. The total number of entries in the LDB is 92453, which is 226 times larger in comparison to [11] and more than twice the used in [15]. The database entry (γ-line) with the highest energy belongs to 20Na with 11258.9
keV. Figure 3 shows the energy distribution of γ-lines in the LDB. There is an apparent under representation in the vicinity of 511 keV (≈ 50%). This might indicate a systematic over-subtraction of positron annihilation backgrounds in the reported measurements.

Using a large database introduces a problem to peak identification because peaks in a spectrum could be explained by many γ-lines, even if the peak has a narrow width. The ranking criteria presented in Section III aim to overcome this issue.

III. RANKING CRITERIA

Ten criteria were originally designed to identify γ-lines from potential isotopes that could be present in a sample using the peaks found in its γ spectrum. They were labeled arbitrarily from A to J, however, only the most significant are reported here using their original names and codes. Information about all the methods is reported in the histoGe’s User Manual [35]. The criteria can be applied individually or in combinations in arbitrary order.

These criteria differ from other previously reported methods in how they utilize the information such as the emission probability (EP), the relative emission probability (REP), the ratio between the number of peaks identified in the spectrum and the number of peaks found in the database for certain isotopes, among others. Each criterion has merit by itself, but they can be combined to obtain better results. Before the analysis begins, the spectra must be properly calibrated to obtain reliable results.

The criteria described in this work, their code and their ranking category are listed in table I.

When two gamma lines in the same peak get the same RV, other criteria are used as tiebreakers to decide in favor of one of them, e.g., for criterion F, criterion E is used and if the tie persists, then, criterion D is used. For criterion H, $\text{RMSE}_{\text{mod}}$ and criterion F were used. Next, a description of each criterion is given.

A. γ-line coincidence probability

The distance $d$ (in keV) from the peak mean to each of the γ-lines is used to calculate the probability ($P_G$) that the peak can be explained by a γ-line. This is done using the cumulative distribution function (CDF) for the Gaussian distribution:

$$P_G(d, \mu = 0, \sigma) = 2\text{CDF}(-|d|, 0, \sigma)$$  \hspace{1cm} (1)

where $\mu$ is the mean of the CDF and $\sigma$ is the peak’s standard deviation obtained from a Gaussian fit. The RV is assigned directly to each γ-line and it is given by equation (1). The γ-lines are then sorted according to their RVs.

B. Improved Peak Explanation Power

A single parent isotope could have multiple γ-lines appearing as peaks in a spectrum. Every time a γ-line of a particular isotope is matched with a peak, the chance that the isotope is present in the sample is increased, making it more suitable to explain the spectrum as a whole. The RV is calculated as the ratio of the number of γ-lines from a given isotope that fall within the peaks of the spectrum, to the number of γ-lines from that isotope expected in the whole range of the spectrum (from the first to the last peak). The RV is in the semi-closed interval (0,1] and sorting is done in descending order.

C. Relative Emission Probability (REP)

The REP was calculated and included in the LDB for each γ-line. This had to be done since many of the entries had non-numeric or missing EP values. When not available, EP were set to the minimum EP for the isotope [35]. Using the REP allows to associate a set of γ-lines with their respective parent isotopes knowing that, for a given isotope, the REP of its γ-lines should add up to 1 [35].

The RV is calculated as the sum of the REP of those γ-lines found in the queries for a particular isotope. This method also gives rank values in the (0,1] and they are sorted in descending order.

D. Ranking With a Fuzzy Inference System

Fuzzy logic [40] was used to compose a more powerful RV from the combination of three inputs. It is a method to formalize “approximate” reasoning and it is a tool to treat uncertainty and vagueness. Unlike classical logic, where propositions can only be true or false; propositions in fuzzy
logic can have a degree of truth between 0 and 1 [41]. A fuzzy inference system (FIS) performs a deductive inference through IF-THEN rules with fuzzy sets [42]. A well known and straightforward way to implement a FIS is through the Mamdani’s inference [43]. The steps to perform this type of inference are: fuzzification of the inputs; inference, which is divided in: calculation of the antecedents, implication and aggregation of rules; and finally, defuzzification via the centroid method [41]

The RMSE$_{Mod}$ is defined using some statistical methods to analyze $\gamma$-ray spectra presented by Gilmore [1] such as the net area of a peak $A = G - B$, where $G$ is the peak’s integral and $B$ is the estimated background under the peak, together with the REP. Then, RMSE$_{Mod}$ is defined as:

$$RMSE_{Mod} = \frac{P_T}{P_r} \left[ \frac{1}{N} \sum_{i=1}^{N} \left( \frac{A_i}{A_T} - \frac{I_{g_i}}{I_{g_T}} \right)^2 \right],$$

where $N$ is the total number of peaks, $A_i$ is the net area of the $i$-th peak, $A_T$ is the sum of all the net areas, $I_{g_i}$ is the REP of the $i$-th peak and $I_{g_T}$ is the sum of the REP for all lines identified for the respective isotope. The factor $\frac{P_T}{P_r}$ that modifies the RMSE in equation (2), was included to penalize those isotopes that explain fewer peaks of the spectrum in comparison to the expected number of peaks.

Each input has three fuzzy sets and the output has five fuzzy sets. Fuzzy sets were defined using the well-known sigmoid and Gaussian functions whose mathematical expressions are, respectively, given by:

$$f_g(x, \sigma, \mu) = \frac{1}{1 + e^{-k_o(x-x_o)}},$$

$$f_s(x, k_o, x_o) = e^{-\frac{(x-x_o)^2}{2\sigma^2}},$$

where $k_o$ and $x_o$ are the parameters of the sigmoid function, and $\sigma$ and $\mu$ are the parameters of the Gaussian function.

Figure 4 shows the name, curve, the mathematical function of the fuzzy sets used in the FIS. The parameters of the fuzzy sets were established considering the designer’s own knowledge about what the linguistic variables Very Low (VL), Low (L), Medium (M), High (H) or Very High (VH) could mean considering that the output hypersurface must be a monotonically increasing one.

The rules used for the inference are of the form:

IF $x_1$ is RMSE$_{n}$ and $x_2$ is PeakRatio$_n$ and $x_3$ is Ig$_{k}$

THEN $y_k$ is Affinity$_q$,

where $k$ is an index that refers to $k$-th rule as is shown in table II and $m$, $n$, $p$ and $q$ are the indices of their respective fuzzy sets which can be VL, L, M, H or VH depending on whether they are input or output fuzzy sets, as shown in figure 2. The rules used for the inference process are shown in table II. Since there are three fuzzy sets for each input, there are twenty
were observed in the results got with this method. Corrected spectra were used, but no important improvements could be used to untie isotopes with the same RV. Efficiency of the fuzzy rank method, however, other criteria such as Mamdani inference can be implemented to calculate the affinity of isotopes. All γ-lines that share the same parent get a tie. For the first query range (Co60 lines), the isotope Co, since all expected γ-lines are found in the query ranges for both isotopes. Notice that, since this criterion assigns a RV per isotope, γ-lines from the same isotope found in the two query ranges have the same RV. For the first query range (Co60: from 1160.84 keV to 1182.18 keV), the isotope 60Co is found at the top position by itself, but for the other one (Co60: 1323.41 keV to 1340.54 keV) it is tied with 53Co, since all expected gamma lines are found in the query ranges for both isotopes. A tie breaking based on criterion E is effected, which places 60Co at the top (1).

V. RESULTS

A. Example with a 60Co sealed point-like source

As a first example, criterion F (III-C) was applied over the spectrum obtained in the ICN-UNAM from the 60Co point-like source reported in Table III. For this test, an info file with two query ranges, associated with each of the two more prominent gamma lines of this isotope was used. Table V summarizes the results, showing the positions after sorting the RV of their corresponding gamma lines. The two energy ranges are shown in red in Figure 5 as well as the 10 best candidates γ-lines are shown in the inserts.

The results in sections V-A and V-B below were obtained with spectra from a set of radioactive sealed calibration sources (Table III) and they were acquired with an EG&G-ORTEC Hyper Pure Germanium detector in the Detectors Laboratory at the Institute of Nuclear Science of the National Autonomous University of Mexico (UNAM), hereafter referred to as ICN-HPGe detector, whose characterization has been reported elsewhere [44]. The data acquisition system (DAQ) was a PX5-HPGe multi-channel analyzer (MCA) and a digital pulse processor (DPP) software analyzer provided by Amptek [45].

IV. EXPERIMENTAL SETUP

Fig. 4: Parameters of the fuzzy sets used in the FIS. Sigmoid and Gaussian functions parameters are \( f_s(x, k_x, x_o) \) and \( f_g(x, \sigma, \mu) \), respectively. The names of the fuzzy sets are: VL is “Very Low”, L is “Low”, M is “Medium”, H is “High” and VH is “Very High”.

seven rules with their consequents chosen from five fuzzy sets. During the design of the FIS, it was decided that five output fuzzy sets were enough to categorize the combinations of the inputs.

Once the fuzzy sets are defined and the rules given, the Mamdani inference can be implemented to calculate the affinities of isotopes. All γ-lines that share the same parent get the same RV. They are later sorted in descending order using as a tiebreaker criterion the \( RMSE_{Mod} \). This criterion was chosen for its simplicity and because it is already an input of the fuzzy rank method, however, other criteria such as \( f_{GR} \) could be used to untie isotopes with the same RV. Efficiency corrected spectra were used, but no important improvements were observed in the results got with this method.

E. Chain using Relative Emission Probability

In some spectra, the presence of some peaks can be due to γ-lines from several isotopes that are connected to each other via a decay chain. This motivates the design of a criterion that rank chains instead of isotopes alone.

For this criterion, the RV is calculated as follows: \( REP \) of the γ-lines found in the query ranges belonging to all the isotopes in a given chain are summed and averaged.

\[ REP = \frac{1}{N} \sum_{i=1}^{N} \gamma_i \]

As described in section II the RV is assigned to all the γ-lines of all isotopes that belong to the chain. Sorting is done in descending order.

\[ \gamma_i \]

\[ a \]

\[ b \]

\[ c \]

\[ d \]

\[ e \]

\[ f \]

\[ g \]

\[ h \]

\[ i \]

\[ j \]

\[ k \]

\[ l \]

\[ m \]

\[ n \]

\[ o \]

\[ p \]

\[ q \]

\[ r \]

\[ s \]

\[ t \]

\[ u \]

\[ v \]

\[ w \]

\[ x \]

\[ y \]

\[ z \]
TABLE II: FIS RULES USED IN histoGe’s FUZZY RANK. MVL, ML, MM, MH AND MVH REFER TO THE AFFINITY OUTPUT FUZZY SETS: VERY LOW, LOW, MEDIUM, HIGH AND VERY HIGH, RESPECTIVELY.

| Source | PeakRatioLow | PeakRatioMedium | PeakRatioHigh | PeakRatioLow | PeakRatioMedium | PeakRatioHigh | PeakRatioLow | PeakRatioMedium | PeakRatioHigh |
|--------|--------------|----------------|--------------|--------------|----------------|--------------|--------------|----------------|--------------|
| REPLow | ML           | MM             | ML           | ML           | MM             | MM           | ML           | MM             | ML           |
| REPMedium | MM           | MH             | MH           | MM           | MH             | MH           | ML           | MH             | MM           |
| REPHigh | MVH          | MVH            | MVH          | MVH          | MVH            | MVH          | MVH          | MVH            | MVH          |

TABLE III: ENERGY AND HALF-LIFE OF THE POINT-LIKE SEALED SOURCES USED TO COMPARE RANKING CRITERIA.

| Source | γ-lines [keV] | half-life [yr] |
|--------|---------------|---------------|
| 241Am  | 13.81, 27.03, 33.19, 43.42, 59.76, 98.97, 102.98, 120.36, 125.33 | 432.5 |
| 133Ba  | 53.1, 79.6, 81.0, 160.6, 223.3, 276.3, 302.8, 356.0, 383.8 | 10.5 |
| 109Cd  | 88.07         | 1.27          |
| 99Co   | 122.0, 136.0  | 0.745         |
| 60Co   | 1173.2, 1332.5 | 5.27         |
| 137Cs  | 662.0         | 30.1          |
| 65Mn   | 835.0         | 0.855         |
| 22Na   | 1275.0        | 2.6           |
| 60Zn   | 1115.0        | 0.668         |

Fig. 5: 60Co spectrum obtained from the point-like source described in table III. Peak ranges for energy queries are drawn in red. Plot was made using histoGe [33]. The zoomed-in inserts show the peaks along with first 10 γ-lines (tagged by their position) in accordance to table IV.

S_criterion(\(p\gamma-line\)) = \begin{cases} 10 - p\gamma-line & p\gamma-line < 10 \\ 0 & p\gamma-line \geq 10 \end{cases}, \quad (5)

where \(p\gamma-line\) is the list position of the γ-line (starting from 0) for a given criterion. For example, in the 109Cd radioactive source (table IV), the γ-line located at 88.04 keV was sorted by criterion F at position 2 (third place) an it receives a score of 10 – 2 = 8.

Then, the following operation is done for obtaining the normalized score (NS) for a specific criterion:

\[ NS_{criterion} = \frac{\sum_{\gamma-line} S_{criterion}(p\gamma-line)}{MS} \quad (6) \]

where the maximum possible score is given by \(MS = 10 \times \text{Total}_{\gamma-lines} = 10 \times 27 = 270\). The NS range is between 0 and 1, being 0 and 1 the worst and the best possible results, respectively.

Under these expressions, criterion B performed poorly (the expected γ-line positions are mostly above 9), it was taken as the baseline criteria during comparison with other methods. This means that coincidence probability is not relevant because it depends on a good calibration and its uncertainty. The fact that the density of γ-lines is so high affects the performance, because many other γ-lines could be as near as or nearer than that of interest. Criterion E was capable to identify 133Ba and 60Co but showed a poor performance for 241Am because it has 171 γ-lines in [39] and 132 γ-lines in the range of its spectrum but only 22 γ-lines were observed. Criterion F improves the results obtained with the criterion E. In the worst case, it equals the performance of criterion E, but the fact that it uses the REP makes it able to focus on those γ-lines that
have the highest EP giving little importance to those γ-lines that could be undetectable with a certain detector. Criterion H gave the best results, in particular, when those isotopes with a relatively small REP are filtered to discard them from sorting and ranking (H+). Fuzzy ranking is able to get approximately 0.95 or 0.87 of the maximum score with and without filtering (H and H+), respectively. The overall performance of H+ makes it the best one with a score of 0.955. This result suggests that combining the criteria to make identification algorithms can give better results than using individual criteria alone. For criterion J, fair results were obtained. A simple inspection of the results reveals that this criterion correctly identifies the γ-lines 7 of the 10 radioactive sources in Table I. In particular, the γ-lines for 241Am all get ranked at positions higher than 34 and the line of 54Mn was placed at position 251. In general, 241Am was hard to be identified by histoGe and this penalizes some criteria more than others.

Nonetheless, no significant improvements were observed when efficiency corrected spectra were used as input. Therefore, identification could be achieved without knowing the detector’s efficiency [44]. Besides, a test was implemented to find the dependence of the ranking results against the peak-to-background ratio (PBR) defined as the ratio of the area enclosed by a peak to the area of the background beneath it, in an interval ±3σ around the peak, and calculated using an exponential plus second order polynomial fit. Fake peaks with variable amplitude were introduced in known spectra and it was found that, once the PDA detect the peak, the position of that γ-line is unaffected for methods E, F and J, and negligible changes in position were observed for B, H and H+. Thus, changes in position were observed for B, H and H+. Thus, that variable amplitude were introduced in known spectra and it exponential plus second order polynomial fit. Fake peaks with in an interval enclosed by a peak to the area of the background beneath it, to-background ratio (PBR) defined as the ratio of the area find the dependence of the ranking results against the peak-detector’s efficiency [44]. Besides, a test was implemented to fore, identification could be achieved without knowing the when efficiency corrected spectra were used as input. There-

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higher than 34 and the line of 54

γ

lines is unaffected for methods E, F and J, and negligible

γ

identifying the γ-lines present in the point-like sources of

Table III and an extended 210Pb source through the normalized score (NS) defined by (6). The score of InterSpec software [16] is shown in the last bar (IS).

C. Performance over generic γ spectra

To explore the capability of the program to identify isotopes in spectra that may contain an arbitrary combination of isotopes, various test were performed using previously studied samples [46], [47]: one had traces of 210Pb (sample A) and the other contained 177Lu and 131I (sample B) and a rock sample taken from the inside of LabChico’s site (sample C). It was known that sample A was spiked with lead and it was acquired with the high purity germanium detector of the Institute of Physics of the UNAM (IF-BEGe detector) [44] with a exposure time of 24 h, spectrum of sample B was acquired inside the Bouby Underground Germanium Suite (BUGS) facility [48] at Bouby Underground Laboratory [49] and a previous analysis of sample B identified 177Lu and 131I [46] and sample C is a rock taken during geotechnical studies [47] and assayed with the ICN-UNAM HPGe [44].

Table VI shows the results of applying criteria F and H to samples A, B and C. Criteria F and H were chosen for this test because they showed the best performance in the test with the point-like sealed sources. For the sample A, in addition to the 210Pb γ-line at 46.5 keV (positions 1 for rank F and 0 for rank H), two X-rays associated with 210Pb were observed, however, their assigned positions (higher than 20) are irrelevant because the database does not contain reliable information about X-rays. This motivates the addition of X-ray information to the LDB. For sample B, ranks H and H+ identified 131I, 177Lu and 40K at positions 0 for all the γ-lines which is the best possible result. The 40K seen in sample B can be safely attributed to the water given the ultra low background of the BUGS detector [50]. About sample C, 40K was successfully

Fig. 6: Comparison of the performance of the criteria in identifying the γ-lines present in the point-like sources of Table III and an extended 210Pb source through the normalized score (NS) defined by (6). The score of InterSpec software [16] is shown in the last bar (IS).
TABLE V: Position for every γ-line expected from the spectra of the radioactive sources of Table III using the criteria described in section III. The isotopes and their γ-lines properties, energy query range, the number of isotopes found in the query range, the positions of each γ-line are shown for each criterion and, besides, the IS’s individual performance. The top γ-line position is zero. 210Pb spectra was obtained from sample A from Table VI. For 241Am ranges could include more than one γ-line reported in the database, in this case all are ranked together.

| γ-line Properties | Query Range | γ-line Position |
|-------------------|-------------|----------------|
| Eg [keV] | Emin | Emax | PBR | Size | B | E | F | H | H+ | J | IS |
| 26.344 | 2.7 | 28.355 | 10.5 | 200 | 0 | 1 | 0 | 0 | 5 | 4 | 34 |
| 33.196 | 0.13 | 31.122 | 1.7 | 198 | 0 | 1 | 0 | 0 | 5 | 4 | 34 |
| 43.423 | 0.07 | 42.355 | 0.2 | 143 | 0 | 1 | 0 | 0 | 5 | 4 | 34 |
| 51.010 | 0.00 | 48.031 | 0.2 | 194 | 0 | 1 | 0 | 0 | 5 | 4 | 34 |
| 59.541 | 0.39 | 56.673 | 19.5 | 414 | 0 | 1 | 0 | 0 | 5 | 4 | 34 |
| 69.760 | 0.00 | 68.316 | 0.03 | 206 | 0 | 1 | 0 | 0 | 5 | 4 | 34 |
| 75.800 | 0.00 | 75.411 | 0.1 | 177 | 0 | 1 | 0 | 0 | 5 | 4 | 34 |
| 79.100 | 0.00 | 78.649 | 0.1 | 211 | 0 | 1 | 0 | 0 | 5 | 4 | 34 |
| 98.970 | 0.02 | 96.695 | 0.5 | 251 | 0 | 1 | 0 | 0 | 5 | 4 | 34 |
| 102.98 | 0.02 | 100.502 | 0.5 | 345 | 0 | 1 | 0 | 0 | 5 | 4 | 34 |
| 120.36 | 0.00 | 116.561 | 2.9 | 372 | 0 | 1 | 0 | 0 | 5 | 4 | 34 |
| 125.30 | 0.00 | 123.656 | 0.6 | 239 | 0 | 1 | 0 | 0 | 5 | 4 | 34 |
| 53.161 | 2.2 | 51.353 | 8.3 | 258 | 0 | 1 | 0 | 0 | 5 | 4 | 34 |
| 79.613 | 2.62 | 78.327 | 5.1 | 344 | 0 | 1 | 0 | 0 | 5 | 4 | 34 |
| 80.997 | 3.06 | 78.327 | - | 344 | 0 | 1 | 0 | 0 | 5 | 4 | 34 |
| 160.613 | 0.65 | 156 | 0.06 | 681 | 0 | 1 | 0 | 0 | 5 | 4 | 34 |
| 223.234 | 0.45 | 220 | 0.06 | 220 | 0 | 1 | 0 | 0 | 5 | 4 | 34 |
| 276.098 | 7.16 | 273.376 | 7.5 | 273 | 0 | 1 | 0 | 0 | 5 | 4 | 34 |
| 302.853 | 18.33 | 300.866 | 7.5 | 347 | 0 | 1 | 0 | 0 | 5 | 4 | 34 |
| 356.017 | 62.05 | 352.929 | 197.6 | 382 | 0 | 1 | 0 | 0 | 5 | 4 | 34 |
| 383.851 | 8.94 | 380.345 | 29.5 | 453 | 0 | 1 | 0 | 0 | 5 | 4 | 34 |
| 88.046 | 3.61 | 85.439 | 9.7 | 378 | 0 | 1 | 0 | 0 | 5 | 4 | 34 |
| 122.064 | 10.68 | 120.080 | 11.4 | 315 | 0 | 1 | 0 | 0 | 5 | 4 | 34 |
| 136.474 | 10.68 | 135.534 | 17.2 | 135 | 0 | 1 | 0 | 0 | 5 | 4 | 34 |
| 1173.237 | 99.97 | 1170.839 | 8.2 | 222 | 0 | 1 | 0 | 0 | 5 | 4 | 34 |
| 1332.501 | 99.97 | 1329.410 | 24.4 | 89 | 0 | 1 | 0 | 0 | 5 | 4 | 34 |
| 661.657 | 82.1 | 660.202 | 13.0 | 142 | 0 | 1 | 0 | 0 | 5 | 4 | 34 |
| 834.344 | 99.98 | 831.233 | 19.1 | 343 | 0 | 1 | 0 | 0 | 5 | 4 | 34 |
| 1274.53 | 99.94 | 1270.843 | 22.2 | 253 | 0 | 1 | 0 | 0 | 5 | 4 | 34 |
| 367.59 | 4.25 | 35.704 | 0.9 | 68 | 0 | 1 | 0 | 0 | 5 | 4 | 34 |
| 1115.546 | 50.6 | 1101.568 | 3.3 | 115 | 0 | 1 | 0 | 0 | 5 | 4 | 34 |

identified by ranks F, H and H+. The photopeaks observed at 2614 keV was attributed to 208Tl because it belongs to 232Th decay chain and was found at positions 3 for ranks F, H and H+. Two photopeaks were attributed to 214Bi because they belong to 238U decay chain, however, no further peaks that could have increased the confidence were identified due to the high background at energies below the potassium peak in the spectrum. 214Bi has 214 entries in the database but only two were considered resulting in a low REP which affected the behavior of H rank.

D. Ranking without raw data

It is often the case that the spectrum data is only available in image format and ranks E, F and J have the advantage of performing ranking without the raw data of the spectrum. To show this capability, a calibrated spectrum in which isotopes were identified and marked with tags (figure 2 of [51]) was analyzed using ranks F and J. The peak maxima and Full Width at Half Maximum reported (FWHM) were used to construct an info file [55] with 17 query ranges. The query ranges were defined as two times the FWHM except for 214Th in which only one FWHM was used. The positions and RV for each γ-line are shown in Table VII. For rank F, all the γ-lines known to be present were placed within the first ten positions to explain the peak where they are found. 12 out of 17 were placed among the top three and 3 were positioned fourth. Criterion J placed the lines within the first ten positions except for 234mPa and 40K which could not be associated with a specific decay chain in the LDB.

E. Comparison between histoGe and InterSpec

Table VIII shows a performance comparison between histoGe and InterSpec from Sandia Labs [16], whose output is also an ordered list. Two spectra were analyzed: sample B of Table VI and the background spectrum of the Lumpsey detector, at Boulby, (B-BKG) [50]. For sample B, rank H of histoGe outperforms InterSpec while rank F is slightly better, in particular, to identify 177Lu and the 80.158 keV photopeak of 131I. For B-BKG, InterSpec was capable of ranking correctly more γ-lines at first position in comparison to the best result of histoGe, however, there is one peak in which it fails completely (208Tl at 510.77 eV) because it is confused with e−e+ annihilation line. So, if Δ is defined as the position difference between InterSpec
TABLE VI: POSITIONS AND RANKS, ACCORDING TO CRITERIA F, H AND H+, OF PARENT ISOTOPES KNOWN TO BE PRESENT IN THE TWO WATER SAMPLES (A AND B), AND AT THE INTENDED SITE FOR LABChico (C). THE QUERY RANGES AND $\gamma$-RAY PROPERTIES ARE ALSO SHOWN. *214Bi

HAS 214 $\gamma$-LINE AND DUE TO ONLY 2 LINES WERE IDENTIFIED, REP IS SMALL WHICH AFFECTS THE BEHAVIOR OF H RANK.

| sample | isotope | $\gamma$-ray properties | Query Range | PBR | Size | Position | RV |
|--------|---------|--------------------------|-------------|-----|------|----------|----|
|        |         |                          |             |     |      | $F$  | $H$ | $H+$ |
| A      | 210Pb   | 46.53 of 4.25            | 45.70 of 47.06 | 0.950 | 69 | 1 0 0 | 1 0.873 0.873 |
| B      | 217Lu   | 112.94 of 6.4            | 111.11 of 115.14 | 1.010 | 279 | 1 0 0 | 0.965 0.904 0.999 |
|        | 217Lu   | 206.36 of 11.0           | 206.8 of 209.8 | 2.489 | 91 | 3 0 0 | 0.965 0.904 0.999 |
|        | 208Pb   | 80.18 of 2.62            | 79.3 of 82.0 | 0.169 | 158 | 9 0 0 | 0.969 0.912 0.999 |
| C      | 201Bi*  | 1460.83 of 11.0          | 1457.0 of 1463.53 | 0.436 | 423 | 0 0 0 | 0.870 0.873 0.873 |

TABLE VII: RV AND POSITIONS CALCULATED FOR 4 CRITERIA FROM THE PUBLISHED PARENT ISOTOPES ON FIGURE 2 OF [51]. NOTE FROM THE SIZE COLUMN THAT THE NUMBER OF CANDIDATES IS IN THE HUNDREDS.

and the histoGe’s best result, it can be seen that $\sum \Delta > 0$, which means that histoGe had a better overall performance when both, sample B and B-BKGD are considered.

The paradigm by which histoGe identifies the isotopes is completely different in comparison to InterSpec’s. histoGe performs better when more photopeaks of an isotope are observed in the spectrum, however, sometimes InterSpec works better when individual peaks are considered in such a way that even identification of 2 unrelated peaks could make InterSpec fail the isotopes’ identification unlike histoGe which is able to manage contextually many isotopes per run.

F. Computational cost

The computational cost of criteria F and H is due to querying the LDB, calculating the RV, and sorting all the $\gamma$-lines. A Monte Carlo analysis was performed to estimate the time used for these processes. The procedure followed to implement this test is described next: .info files were randomly constructed with a variable number of $\gamma$-lines per file, and then, ranked. The center of the energy was chosen randomly between 200 keV and 2500 keV per each $\gamma$-line and its width was calculated using a distributed normally random numbers with $\mu = 0$ and $\sigma = 5$. The minimum energy range is 1 keV to avoid small intervals that could contain few $\gamma$-lines. This procedure was repeated 100 times. The computer used to execute this analysis has an AMD Ryzen 7 processor 4800H, 16 GB of RAM and a SSD. A program was made to measure the execution time of each call to histoGe using those .info files generated randomly. For H criterion, an arbitrary spectrum was chosen considering that its energy range is larger than the query ranges. Figure 7 shows that the execution time follows a non-linear relationship in which increasing the number of peaks by 10 does not even get the execution time doubled. These results show that histoGe has reasonable (≥ 20s) execution times for real spectra in which the number of $\gamma$-lines do not exceed a hundred peaks. As expected, H is slower than F, but for a low number of peaks, there is not a considerable difference between them.

On the other hand, note that processing a spectrum form peak identification using the PDA through the histoGe’s peak finder tool to $\gamma$-line identification using some criteria could take a few minutes, however, due to the peak finding
TABLE VIII: COMPARISON OF THE RESULTS OBTAINED BETWEEN histoGe vs. InterSpec (IS). Δ MEANS THE DIFFERENCE OF POSITION OF InterSpec MINUS POSITION OF histoGe. A POSITIVE Δ FAVORS histoGe. DUE TO $^{214}$Bi HAS MORE THAN 2 HUNDREDS OF PEAKS THE INFORMATION TO IDENTIFY IT CLEARLY IS INCOMPLETE MAKING HARDER THE IDENTIFICATION FOR histoGe. RANGE HAD TO BE MADE WIDER SO IS COULD MAKE IDENTIFICATION. $^{122}$Rn, $e^- e^+$ AND $^{214}$Pb WERE IDENTIFIED AT POSITIONS 0, 1 AND 2, RESPECTIVELY AND BOTH ISO Topes WERE FOUND IN THE BACKGROUND.

| Known isotope | Range | Position | F | H | H+ | IS | Δ |
|---------------|-------|----------|---|---|----|----|---|
| spectrum | isotope | Eg [keV] | Emin | Emax | | | |
| Sample B | 177| Lu  | 112.94 | 111.131 | 115.1425 | 1 | 0 | 0 | 1 | 1 |
| | 177| Lu  | 208.36 | 206.8 | 209.8 | 3 | 0 | 0 | 2 | 2 |
| | 131| I  | 80.18 | 79.3 | 82.0 | 4 | 0 | 0 | 6 | 6 |
| | 131| I  | 284.30 | 282.5 | 285.5 | 0 | 0 | 0 | 0 | 0 |
| | 131| I  | 364.48 | 362.51 | 366.1 | 0 | 0 | 0 | 0 | 0 |
| | 133| I  | 636.98 | 634.64 | 639.0 | 0 | 0 | 0 | 0 | 0 |
| | 40| K  | 1460.83 | 1457.0 | 1463.53 | 0 | 0 | 0 | 0 | 0 |
| B-BKG | 214| Bi  | 609.31 | 607.7 | 611.3 | 2 | 1 | 2 | 0 | 1 |
| | 214| Bi  | 768.35 | 767.80 | 769.82 | 1 | 0 | 0 | 0 | 0 |
| | 214| Bi  | 1120.28 | 1119.10 | 1122.55 | 1 | 1 | 1 | 0 | 1 |
| | 214| Bi  | 1764.49 | 1763.35 | 1767.20 | 1 | 1 | 1 | 0 | 1 |
| | 214| Bi  | 2204.21 | 2203.50 | 2207.16 | 0 | 0 | 0 | 0 | 0 |
| | 40| K  | 1460.83 | 1459.5 | 1462.8 | 0 | 0 | 0 | 0 | 0 |
| | 229| Ac  | 911.20 | 909.7 | 913.16 | 1 | 1 | 2 | 0 | 1 |
| | 229| Ac  | 968.97 | 967.57 | 971.23 | 0 | 0 | 0 | 0 | 0 |
| | 229| Ac  | 1588.19 | 1587.5 | 1589.77 | 0 | 0 | 0 | 0 | 0 |
| | 212| Pb  | 47.91 | 45.1 | 47.92 | 3 | 0 | 0 | 0 | 0 |
| | 212| Pb  | 238.63 | 237.00 | 239.95 | 0 | 0 | 0 | 0 | 0 |
| | 212| Pb  | 300.087 | 298.9 | 301.3 | 0 | 0 | 0 | 0 | 0 |
| | 208| Tl  | 510.77 | 509.82 | 512.16 | 2 | 3 | 1 | 0 | 2 |
| | 208| Tl  | 583.19 | 581.8 | 584.89 | 1 | 2 | 1 | 0 | 1 |
| | 208| Tl  | 860.56 | 859.2 | 862.5 | 0 | 0 | 0 | 0 | 0 |
| | 214| Pb  | 2614.53 | 2613.65 | 2617.31 | 1 | 2 | 0 | 0 | 0 |
| | 214| Pb  | 249.99 | 240.3 | 243.2 | 1 | 0 | 0 | 4 | 4 |
| | 214| Pb  | 295.22 | 293.76 | 296.3 | 0 | 1 | 0 | 0 | 0 |
| | 214| Pb  | 351.93 | 350.52 | 353.3 | 2 | 3 | 0 | 0 | 0 |

Algorithms are not fully accurate more time could be required to adjust the query ranges in the info file by a trained spectroscopist.

VI. Conclusions

histoGe is a tool for the identification of peaks in a $\gamma$ spectrum using the information in a large database containing 92,453 gamma lines and 2,200 radioactive nuclides. The program implements different criteria to rank and sort candidate isotopes to explain the presence of peaks in a spectrum with a philosophy inspired by that of a search engine.

Five different criteria were presented and their performances were compared according to their ability to identify the $\gamma$-lines from a suite of sealed calibrated radioactive gamma sources via an ad hoc defined metric. Two of the methods stood out in performance under this test: one based on the use of Relative Emission Probabilities (F), and one using fuzzy logic (H) which combine information used in other criteria. An enhanced version of the latter (H+) where $\gamma$-lines with relatively small REPs are discarded achieved $\sim 95\%$ efficiency to identify the isotopes in a set of sealed radioactive sources. Under the same assumptions, InterSpec was also tested giving an efficiency of $70\%$, demonstrating that under this conditions histoGe performed better. $^{241}$Am was a hard isotope to identify, as only a small fraction of its $\gamma$-lines are typically visible in spectra. The score increases quite noticeably when $^{241}$Am is not considered.

histoGe was used to identify the $\gamma$-lines in three arbitrary samples: water with lead (sample A), London tap water (sample B) and the rock of LabChico’s site (sample C). For sample A, a $\gamma$-line of $^{210}$Pb was identified with accuracy but X-rays were not because the database has not complete information about X-rays. For sample B, criterion H got the best score when $^{212}$Rn, $e^- e^+$ and $^{214}$Pb were identified. For sample C, $^{214}$Bi was hardly identified by rank H due to it having 290 $\gamma$-lines but rank F performed better and H+ identified it with accuracy.

Fig. 7: Average and error bars (2$\sigma$) of the execution time of criteria F (blue) and H (red) vs. the number $\gamma$-lines per info file.
An advantage of histoGe is that it is capable of making isotope identification using only the ranges of the photopeaks without the raw data of the spectrum. Criteria E, F, and J are capable of doing this. Then, a published spectrum was ranked with criterion F, it was capable to identify all of them within the 10 first positions, in fact most of them were among the first three places. Chain rank (III-E) identified all $\gamma$-lines in the top ten places except $^{40}\text{K}$ and $^{234}\text{mPa}$ because there is not a decay chain associated with them. This result motivates further research about how to improve this rank.

histoGe's results of identification of two samples were compared to those obtained with InterSpec of Sandia Labs. In general, the best result of histoGe outperformed InterSpec. Besides, from the results presented in section VI-E and figure 6 it can be seen that histoGe is capable to make identification when multiple related or unrelated $\gamma$-lines are given at the same time, on the contrary, InterSpec cannot identify accurately unrelated peaks but is highly accurate in individual identification. These findings may change if both are tested under different conditions. Using the ideas presented in this work, a histoGe and InterSpec could be combined to get an even more accurate one.

From the Monte Carlo study, it was found that the computational cost for real spectra is affordable. The overall results have shown that histoGe is a tool capable to make reliable identification of isotopes through $\gamma$-spectroscopy with results comparable or even better than other similar software, which makes it able to be applied in teaching research and industrial applications. However, in its current state, we recommend that a trained spectroscopist analyze the results obtained with histoGe or InterSpec to get a better interpretation.

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