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A Nonlinear Preconditioning Transformation-Based Method for Gamma Spectrum Inversion

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Abstract

Building upon the Gold algorithm, this work proposes an iterative inversion method based on nonlinear preconditioning for quantitative analysis of overlapping full-energy peaks in low-resolution γ-ray spectra. The approach replaces the transpose of the response matrix with that of its element-wise cubed form, enhancing spectral separation in densely overlapped regions. Evaluated on synthetic and experimental spectra with varying peak separations, the method achieves lower mean absolute relative deviation (MARD) and standard deviation (SD) than both Gold and ML-EM when peak spacing is $\geq$ ¾ FWHM or randomly distributed. Even under extreme overlap (½ FWHM), it maintains superior accuracy in medium-and highenergy regions. While its computational cost per fixed iteration lies between Gold (fastest) and ML-EM (slowest), faster convergence reduces practical runtime. The algorithm improves inversion accuracy and stability while preserving efficiency, offering a practical solution for low energy resolution γ-ray spectrometry.

Full Text

Preamble

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Inversion

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Abstract

Building upon the Gold algorithm, this work proposes an iterative inversion

method based on nonlinear preconditioning for quantitative analysis of overlapping

full-energy peaks in low-resolution γ-ray spectra. The approach replaces the transpose of the

response matrix with that of its element-wise cubed form, enhancing spectral separation in

densely overlapped regions. Evaluated on synthetic and experimental spectra with varying

peak separations, the method achieves lower mean absolute relative deviation (MARD) and

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accuracy in medium- and high-energy regions. While its computational cost per fixed

iteration lies between Gold (fastest) and ML-EM (slowest), faster convergence reduces

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efficiency, offering a practical solution for low energy resolution γ-ray spectrometry.

Keywords

Gamma spectrum; Inversion; Energy resolution; Nonlinear preconditioning

transformation

1 Introduction

Gamma-ray spectrometry is a well-established technique that enables the performance of

qualitative and quantitative analyses of gamma-ray emitting radionuclides [1], and is widely

applied in fields such as environmental radioactivity surveys and assessments [2-4], radioactive waste regulation [5], and geological and mineral exploration [6]. Accurate

analysis hinges on the precise extraction of characteristic peak information —specifically,

peak position and count —which correspond to the energy and intensity of gamma rays.

Reliable peak deconvolution and integration typically require that peaks be separated by

several times the full width at half maximum (FWHM). In practice, a minimum separation of

FWHM), it maintains superior

approximately 2.5 FWHM is commonly adopted as a guideline to ensure robust and accurate

spectral analysis. To mitigate the effects of spectral overlap, various spectrum deconvolution

and inversion techniques have been developed over the past decades, such as Maximum

Entropy

(ML-EM) [9], the Gold algorithm [10], Maximum A Posteriori (MAP) estimation [11], and

direct deconvolution approaches [12]. Of these, ML-EM and the Gold algorithm have

emerged as the most widely adopted in nuclear spectroscopy owing to their stability,

convergence

conditions [13–20].

During routine spectral inversion tests with a NaI(Tl) system, we empirically identified that

introducing a nonlinear preconditioning transformation —specifically, replacing the

conventional transpose of the detector response matrix with the transpose of its element-wise

cubed form —consistently improved the separation of closely spaced peaks. Although this

modification originates from empirical observation rather than a priori theoretical derivation, validation using both simulated and experimental data demonstrates its superior performance

compared to ML-EM and Gold.

In this work, Simulated spectra were generated for multiple gamma lines with controlled

=1/2, 3/4, and 1 times FWHM, as well as randomly

Method

(MEM) [7,8],

behavior,

Maximum

practical

Likelihood-Expectation

performance

under

realistic

Maximization

measurement

energy separations corresponding to

detector and a mixed point source containing 241Am, 137Cs, 60Co, and 133Ba. The proposed

method is compared with the Gold and ML-EM algorithms in terms of convergence behavior,

computational efficiency, and inversion accuracy. The results show that the proposed method

generally outperforms both the Gold and ML-EM algorithms in terms of quantitative accuracy,

while maintaining computational efficiency comparable to that of the Gold algorithm. This

practical enhancement offers a viable route to higher-precision radionuclide analysis with

cost-effective, low-resolution detection systems.

2.1 Problem description

Gamma-ray spectrum unfolding is fundamentally a linear inverse problem. The measured

distributed combinations. Experimental validation was conducted using a 4″$×4’ ‘$$×16’’ $ NaI(Tl)

spectrum

is related to the incident gamma-ray spectrum

through the detector response matrix

where

denotes the number of spectral channels, and

incident energy bins. Each column of

represents the number of

corresponds to the detector response to a

monoenergetic gamma ray at a specific energy.

In practice, R is typically ill-conditioned, and the measurement Y is contaminated by

is numerically unstable

statistical noise. Consequently, direct inversion

and often produces non-physical solutions (e.g., negative intensities). To address this, the

iteration methods are used preferably [21]. Among iterative methods, the

Gold and ML−EM algorithms have been used more than other methods due to providing positive answers and better accuracy [22].

2.2.1 Gold algorithm

The Gold algorithm is based on the Van Cittert analytical algorithm [23,24]. By multiplying

RT on both sides of the Eq. (1), one obtains the Toeplitz linear system, as shown in Equation

(2). Equation (2) is then solved using Equation (3) [25].

where iteration.

denotes the estimated intensity in the

-th energy bin after the

2.2.2 ML-EM algorithm

The maximum-likelihood expectation-maximization (ML-EM) algorithm is derived under the

assumption that the detected photon counts follow a Poisson distribution [9]. Starting from

the likelihood function of the observed data, the iterative update rule for estimating the

unknown intensity vector is formulated as [26]:

2.2.3 The proposed method

We propose a nonlinear preconditioning strategy to enhance convergence and stability.

Specifically, we replace the standard transpose operator   transformation. Let

this modified operator:

Substituting

with an element-wise cubic

denote the matrix obtained by raising each element of

the third power. We then define

and pre-multiply Eq. (1) by

into the Gold update formula (Eq. 3) leads to the proposed

iteration:

This approach amplifies contributions from larger response elements while suppressing

noise-dominated components, thereby acting as an implicit regularizer.

2.3 Geant4 simulation and response matrix generation

A detailed Monte Carlo model of a 4″$×4’ ‘$$×16’ ’ $ NaI(Tl) scintillation detector was developed

using Geant4 to simulate the detector response to monoenergetic gamma-ray point sources.

The source was positioned on-axis at a fixed distance from the detector face, consistent with

the experimental geometry (Fig. 1 [Figure 1: see original paper]).

43.43cm 42.16cm 40.64cm

MgO radiative layer

57.96cm

right view 5.71cm

stainless steel housing

10.72cm 10.62cm 10.16cm

3.12cm

10.72cm 10.62cm 10.16cm

NaI(Tl) crystal

4.86cm

central point

point source

radioactive point source

For each incident gamma-ray energy—from 30 keV to 3 MeV in steps of

energy points)—1$×$109primary particles were simulate. The energy deposited in the NaI(Tl)

crystal during each event was recorded as the ideal (noise-free) pulse height. To account for

the finite energy resolution of the real detector, a Gaussian energy broadening was applied in

post-processing according to the empirical resolution function:

2.93 keV (1014

Where E is the gamma-ray energy, MeV. Parameters a=-0.00556, b=0.0668, c=0.000228 were

determined by least-squares fitting to measured FWHM values (table 1) obtained from

calibration with standard point sources (241Am : 59.5 keV; 137Cs : 661.7 keV; 60Co : 1173.2

and 1332.5 keV).

The resulting broadened spectra were normalized to unit source intensity and assembled into

a 1014 × 1014 response matrix R , where each column corresponds to the detector response at a specific incident energy.

Energy (MeV)

FWHM (MeV)

3.1.1 Simulated spectrum construction

To evaluate the performance of the inversion algorithms under varying degrees of spectral

overlap, two types of synthetic γ -ray spectra were constructed. Each spectrum consists

of three energy regions, initiated at

recurrence relation

, 661.67 , and 1173.2 keV,

respectively. Within each region, 10 full-energy peaks are generated sequentially using the

0 = 59.54

-1 +

1,2, , 9)

resulting in a total of 30 peaks per spectrum.

Type I (fixed separation): A single fixed value of

nine inter-peak intervals in the 59.54 keV region, the nine in the 661.67 keV region,

. This yields three

and the nine in the 1173.2 keV region all use the same distinct spectra, one for each choice of

, each containing 30 peaks with globally

consistent peak spacing.

applied uniformly across all three energy regions. That is, for a given spectrum, the

{1/2,3/4,1}

Type II (random separation): Within each energy region, the nine values

are independently sampled from {1/2,3/4,1} . The specific sequences

used for the three regions are listed in Table 2 .

The intensity

(in Bq) of the incident γ-ray corresponding to the i-th peak is shown

in Table 3 : the first column provides intensities for the 10 peaks in the 59.54 keV region, the second column for the 661.67 keV region, and the third column for the 1173.2 keV region. All

intensities are randomly generated integers below 100 Bq.

The measured spectrum

where

channel

is then modeled as a linear superposition:

= 1,2, , 30)

denotes the probability that the

-th gamma ray contributes to

E0=59.54 keV

E0=661.67 keV

E0=1173.2 keV

E0=59.54 keV

Intensity of incident gamma rays E0=661.67 keV

E0=1173.2 keV

3.1.2 Peak position estimation

Inversions were conducted using the proposed method, Gold, and ML-EM. Except for slight

shifts observed in several peaks within the low-energy group (starting at 59.54 keV) of the

n=1/2 spectrum, no peak center channel offsets were found in any other energy groups or

spectra (Fig. 2 [Figure 2: see original paper]).

Inversion results for the synthetic spectrum with n = 1/2 (peak separation = 1/2FWHM)

3.1.3 Gamma-ray intensity estimation

The net counts rate under each full-energy peak in the inverted spectrum was taken as the

estimate of the corresponding gamma-ray intensity deviation from the reference intensity

. The relative

relative deviations were evaluated over all 30 peaks (

(Table 3) was defined as:

The mean absolute value of the relative deviation (MARD) and standard deviation (SD) of

where

is the mean of

= 30) as:

Results are summarized in Fig. 3 [Figure 3: see original paper] and lead to the following observations:

For all methods, both MARD and SD decreased monotonically as peak separation

increased.

separation, indicating greater sensitivity to inter-peak spacing.

At separations of 3/4 FWHM, 1 FWHM, and the random case, the proposed method achieved the lowest MARD, outperforming both Gold and ML-EM.

The proposed method exhibited a steeper decline in MARD and SD with increasing

For the 1/2 FWHM spacing, the proposed method had larger MARD and SD than the others.

1/2 FWHM 61.36%

MARD=0.057 SD=0.15

MARD=0.052 SD=0.085

MARD=0.031 SD=0.049

-39.40%

-18.60%

3/4 FWHM

Mean line

Gamma-ray intensity deviation

Gamma-ray intensity deviation

Mean line

MARD=0.013 SD=0.044

-23.41%

MARD=0.023 SD=0.042

MARD=0.038 SD=0.073

-19.18%

-34.91%

The proposed method

ML-EM

1 FWHM

The proposed method

Mean line

ML-EM

Random FWHM

Mean line

2.94%

MARD=0.0014 SD=0.0054

MARD=0.0092 SD=0.014

MARD=0.0089 SD=0.012

-6.40%

-4.70%

Gamma-ray intensity deviation

Gamma-ray intensity deviation

MARD=0.013 SD=0.045

MARD=0.026 SD=0.052

MARD=0.042 SD=0.073

-24.08%

-25.18%

-35.33%

The proposed method

ML-EM

The proposed method

ML-EM

inversion methods

Segmentation into low-energy (first 10 peaks) and medium/high-energy (last 20 peaks)

regions (Figs. 4–5) reveals that the proposed method excels in medium/high-energy ranges,

where detector resolution is better. For the full spectrum, it outperforms competitors when

peak separation $\geq$ 3/4 FWHM.

1/2 FWHM Gamma-ray intensity deviation

Gamma-ray intensity deviation

MARD=0.17

MARD=0.069 SD=0.091

-40% SD=0.28

MARD=0.048 SD=0.062

The proposed method

Gamma-ray intensity deviation

MARD=0.036 SD=0.073

MARD=0.052 SD=0.063

MARD=0.085 SD=0.11

MARD=0.0033 SD=0.0093

MARD=0.0067 SD=0.011

MARD=0.0089 SD=0.014

ML-EM

Random FWHM

Mean line

MARD=0.036 SD=0.075

MARD=0.059 SD=0.083

MARD=0.087 SD=0.11

The proposed method

ML-EM

The proposed method

ML-EM

inversion methods (low-energy segment)

1/2 FWHM

Mean line

MARD=0.0020

SD=0.0042 MARD=0.043 SD=0.085

MARD=0.023 SD=0.041

Mean line

MARD=0.0011 SD=0.0017

MARD=0.0084 SD=0.011

MARD=0.015 SD=0.018

ML-EM

1 FWHM

Mean line

MARD=0.00047 SD=0.00053

MARD=0.0094 SD=0.015

MARD=0.010 SD=0.013

The proposed method

ML-EM

Random FWHM

Gamma-ray intensity deviation

The proposed method

3/4 FWHM

Gamma-ray intensity deviation

Gamma-ray intensity deviation

Gamma-ray intensity deviation

The proposed method

Mean line

ML-EM

1 FWHM

Mean line

Gamma-ray intensity deviation

3/4 FWHM

Mean line

Mean line

MARD=0.0010 SD=0.00079

MARD=0.0091 SD=0.010

MARD=0.019 SD=0.025

The proposed method

ML-EM

The proposed method

ML-EM

inversion methods (medium/high-energy segment)

3.1.4 Convergence and computational speed

Convergence evaluated using the root-mean-square error (RMSE) between spectra at different

iterations:

where

iteration

1 , and

denotes the counts rate in channel

is the total number of channels.

As shown in Fig. 6 [Figure 6: see original paper], the proposed method stabilizes in significantly fewer iterations than Gold or ML-EM.

The proposed method ML-EM

number of iterations

number of iterations

The proposed method ML-EM

number of iterations Random FWHM

The proposed method ML-EM

1 FWHM

The proposed method ML-EM

3/4 FWHM

1/2 FWHM

number of iterations

All algorithms were executed for a fixed number of iterations (100,000) under identical

computational conditions (MATLAB R2023a, Windows 7, Intel® Core™ i7-4790K CPU @

4.00 GHz, 16 GB RAM). As shown in Fig. 7 [Figure 7: see original paper], the total runtime required to complete these

iterations depends on both the algorithm and the degree of spectral overlap:

For the 1/2 FWHM, 3/4 FWHM, and random FWHM cases, the ordering is ML-EM > proposed method > Gold;

For the 1 FWHM case, the ordering reverses to ML-EM > Gold > proposed method.

Despite exhibiting a longer total runtime than Gold in most simulated scenarios when run for

a fixed number of iterations, the proposed method converges significantly faster (Fig. 6).

Consequently, when the stopping criterion is based on convergence rather than a preset

iteration count, the proposed method typically achieves a stable solution in less total time than

both Gold and ML-EM, as demonstrated in the experimental validation (Fig. 9 [Figure 9: see original paper]).

1/2 FWHM

time(s-1)

time(s-1)

number of iterations

1 FWHM

time(s-1)

number of iterations

number of iterations

The proposed method ML-EM

Random FWHM

The proposed method ML-EM

The proposed method ML-EM

time(s-1)

3/4 FWHM

The proposed method ML-EM

number of iterations

3.2 Measured spectrum inversion

The measured spectrum from a mixed point source containing 241Am , 137Cs, 60Co, and 133Ba、

was inverted using the proposed method, Gold, and ML-EM. (Fig. 8 [Figure 8: see original paper]). All methods

successfully recovered the full-energy peaks without any observable channel offset.

Am 59.54keV

measured spectrum

Cs 661.67keV

unfolded spectrum by the proposed method

unfolded spectrum by Gold

unfolded spectrum by ML-EM

Ba 356.01keV

Co 1173.20keV

Ba 80.99keV

Unfolded spectrum(counts/s $×$103)

Measured spectrum(counts/s)

Co 1332.50keV

Ba 302.84keV

Ba 276.40keV

383.85keV

Channel

spectrum using the three methods

Source activities were calculated using:

where

is the net full-energy peak counts rate (cps), and

emission probability obtained from the IAEA nuclear data database.

is the gamma-ray

Quantitative results are summarized in Table 4 . For both closely spaced peaks (separation < 1

FWHM) and well-separated peaks, the proposed method yields the lowest mean absolute

relative deviation (MARD=0.085) and smallest standard deviation (SD=0.10), demonstrating

superior accuracy and stability across all energy regions. ML-EM performs second best, while

Gold exhibits the largest deviations—consistent with the trends observed in Fig. 3 for the

simulated n = 3/4 and random-FWHM cases.

Nuclide

Energy (keV)

Reference Activity (kBq)

Overall

ML-EM

71.24(-11.09%) 47.61(0.87%) 10.45(-6.33%) 10.30(-7.68%) 33.58(3.68%) 46.66(44.06%) 40.84(26.10%) 35.38(9.24%) 30.24(-6.63%) MARD: 0.13 SD:0.18

69.32(-13.49%) 46.62(-1.23%) 10.35(-7.23%) 10.03(-10.10%) 32.81(1.30%) 28.72(-11.33%) 39.83(22.98%) 33.88(4.61%) 28.40(12.31%) MARD: 0.094 SD:0.12

*Percentage deviations from reference values are shown in parentheses

For the measured spectrum, the proposed method achieves RMSE stabilization at

approximately 60,000 iterations, compared to ~80,000 for Gold and ~100,000 for ML-EM

(Fig. 9). Although all methods were executed for the same total number of iterations, the

cumulative computation time at their respective convergence points is lowest for the proposed

method, followed by Gold, and highest for ML-EM. This suggests that the faster convergence

of the proposed method may also translate into a shorter total runtime required to obtain a

stable solution, even when the algorithms are allowed to run for longer.

The proposed method ML-EM

The proposed method ML-EM

time(s-1)

The proposed

method

72.00(-10.14%) 48.35(2.44%) 10.67(-4.36%) 10.52(-5.71%) 34.66(7.01%) 37.50(15.78%) 39.11(20.75%) 34.01(5.01%) 30.74(-5.09%) MARD: 0.085 SD:0.10

number of iterations

number of iterations

4 Conclusion

This study systematically evaluated the proposed nonlinear preconditioning iterative

algorithm against the Gold and ML-EM algorithms. Using simulated spectra with varying

peak spacings (n = 1/2, 3/4, 1, and random combinations) and measured mixed-source spectra,

comparisons were made in computational efficiency, inversion accuracy (MARD), and

stability (SD).

Under a fixed iteration count, the total runtime follows the ordering: Gold < proposed method

< ML-EM for ½, ¾, and random-FWHM cases, while Gold > proposed method in the 1

FWHM case. Despite this, the proposed method exhibits significantly faster convergence (Fig.

6), enabling it to reach a stable solution in fewer iterations. Consequently, when termination is

based on convergence rather than a preset iteration limit, it often achieves a lower practical

runtime than Gold, as demonstrated in the experimental validation (Fig. 9).

In terms of accuracy and stability, the proposed algorithm demonstrates superior performance.

In simulated spectra, it achieves the lowest mean absolute relative deviation (MARD) and

standard deviation (SD) across all tested configurations—including fixed spacings of ¾

FWHM and 1 FWHM, as well as randomly spaced peaks—consistently outperforming both

Gold and ML-EM. In the measured mixed-source spectrum, it also yields the smallest MARD

(0.085) and competitive SD (0.10), surpassing Gold (MARD = 0.13, SD = 0.18) and slightly

improving upon ML-EM (MARD = 0.094, SD = 0.12).

The proposed method performed best when peak spacing was $\geq 3/4$ FWHM, yielding the

smallest MARD and SD in both simulated and measured spectra. This advantage was

confirmed for realistically complex "random spacing" simulated spectra and measured mixed

spectra, demonstrating higher accuracy.

Notably, under extreme overlap (½ FWHM), the overall MARD of the proposed method is

slightly higher than that of Gold and ML-EM. However, segmental analysis reveals that this

degradation is confined to the low-energy region near 59.54 keV, where detector resolution is

poorest. In contrast, it maintains superior performance in the medium- and high-energy regions (661.7 keV and 1173.2 keV). This indicates that algorithmic efficacy may not solely

determined by mathematical peak overlap.

In summary, the proposed inversion algorithm provides more accurate and stable quantitative

results under most practical conditions while maintaining good computational efficiency,

making it a highly practical method for gamma spectrum analysis.

Author Declarations:

We declare that this manuscript is original, has not been published before, and is not currently

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During the preparation of this work, the authors used Qwen-3.5 for the purpose of language

editing and grammar checking only. The tool was used to improve the readability and clarity

of the text. After using this service/tool, the authors reviewed and edited the content as needed

and take full responsibility for the content of the published article. No AI tool was used to

generate scientific data, perform analysis, or formulate conclusions.

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Note: Figure translations are in progress. See original paper for figures.

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