Nucl. Sci. and Tech., Vol.13, No. 3 (2023), pp. 27-37

Nuclear Science and Technology

Journal homepage: http://jnst.vn/index.php/nst

Simulation of gamma spectra from soil samples by using MCNP: A case study

An Trung Nguyen[1], Hao Quang Nguyen[2], Thi Thu Ha Nguyen[2], Duc Thang Duong[2]

[1] Non-Destructive Evaluation Center, 140 Nguyen Tuan, Thanh Xuan Dist, Hanoi
E-mail: natrung@most.gov.vn

[2] Institute of Nuclear Science and Technology, 179 Hoang Quoc Viet, Cau Giay Dist, Hanoi;
E-mail: nhquang@most.gov.vn

Abstract: A comparison between the MCNP simulated gamma spectrums based on the nuclear data from NuDat with the current version 3.0 and Nucléide-Lara against measured spectra based on the IAEA reference samples has been performed to assess the influence of nuclear data of photon decay processes and cross-section of photon interactions on the quality of the simulated spectra. As the results, the appearance of some abnormal energy peaks in the MCNP simulated spectrums; namely 215 keV, 571 keV, 675 keV, 1227 keV of the NuDat-based simulated spectra and 90 keV, 94 keV, 106 keV, 416 keV of the Nucléide-Lara-based simulated spectra, which were present in neither the measured spectrum nor remaining simulated spectra, indicating issues with accuracy and completeness of these dataset. In addition, the good correlation between the combined dataset-based simulated spectra and reference samples-based measured spectra within the range of 50 keV to 2620 keV suggests that this MCNP simulation configuration can be used to generate a large simulated dataset for Machine Learning (ML) models that automatically identify and qualify radioactive isotope from gamma-ray spectra, overcoming the practical limitation of number of reference samples to sufficiently generate data for training and testing ML algorithms in the field of environmental radiation [1].

Keywords: *Gamma spectra; soil samples; MCNP; simulation; NuDat; Nucléide-Lara.*

I. INTRODUCTION

Nuclear data plays a crucial role in modeling codes, such as MCNP or GADRAS, used to simulate nuclear physics processes to bridge gaps where experimental data is lacking or impractical to obtain. However, modeling applications often rely on nuclear data that may have limited or no experimental validation, resulting in outcomes based on low-quality data that may contain gaps and inconsistencies. Recognizing the significance of this issue, the nuclear physics communities have given high priority to nuclear data measurements [2].

Gamma spectrometry simulation is widely used in Calibration of gamma-ray spectrometers by comparing the simulated spectra with experimental data, in Design and Optimization of gamma-ray detectors by studying the response of different detector materials and geometries to gamma-ray interactions, in Nuclear physics Research to understand the behavior of gamma rays emitted during nuclear reactions, in Identification and Quantification of

https://doi.org/10.53747/nst.v13i3.416
Received 23 August 2023, accepted 12 January 2024

radioactive isotopes in environment, in Optimize imaging systems and Improve the accuracy of diagnostic techniques in nuclear medicine, in Radiation Dosimetry by studying the dose distribution of gamma radiation in various materials, in Security and Nuclear Safeguards, etc..

Gamma spectrometry simulation requires knowledge of the atomic and nuclear data that involve in radioactive decay, e.g., half-life, decay modes and branching ratios, the energies and intensities of the various gamma emissions, etc. To provide this information, there are two dominant web-based databases in the world, namely, NuDat and Nucléide-Lara which were developed by the Brookhaven National Laboratory (USA) and Laboratoire National Henri Becquerel (France), respectively. There is a difference in radionuclides data (presence of radionuclides, half-life, decay mode, emission energy and intensity, decay scheme...) between these two databases. In this study, simulated spectra by using MCNP based on NuDat and Nucléide-Lara databases and measured spectra from IAEA reference samples are compared to investigate the impact of nuclear data of photon decay processes on the quality of the simulated spectra. The role of the interaction cross-section for photons is also investigated.

II. METHODOLOGY

The IAEA reference soil samples used in this study were RGU-1 (m=167.644 gam, 224 Bq/kg of U-235, 4941 Bq/kg of U-238) and RGTh-1 (m=157.861 gam, 3250 Bq/kg of Th-232, 3.6 Bq/kg of U-235, 78 Bq/kg of U-238, 6.3 Bq/kg of K-40). In the gamma spectroscopy-related study, only 40 gamma and X-ray emitters in the decay chains were paid much attention and selected. 1691 and 1670 energies of photon and their intensities together with chain coefficients of 40 nuclei from the NuDat 3.0 [3] and Nucléide-Lara [4], respectively, were collected as inputs for MCNP simulation.

Geometry of MNCP simulation was complied with configuration of gamma spectroscopy using GMX series n-type HPGe coaxial detector system and the Model 747 Mirion Lead Shield which are manufactured by Canberra. The soil samples were contained in the 2piL-type plastic box with 10.3 cm inner diameter, 0.2 cm wall thickness and 2.0 cm sample height. The detector’s crystal dimensions given by the manufacturer are: 6.17 cm diameter, 6.1 cm height and 0.4 cm from the top of the detector’s crystal to the entrance window.

The simulation process was conducted by following the procedure [5] [6], taking into account the increase of inactive germanium layer of detector after 13 years of operation [7] [8] [9][10], geometry of measuring system and resolution of detector. Selecting 100 millions of source particles for simulation was complied with the emitted particles in real samples. The graphic output of the MCNP6 computer code for vertical cross-section of the geometry is demonstrated in the Fig. 1.

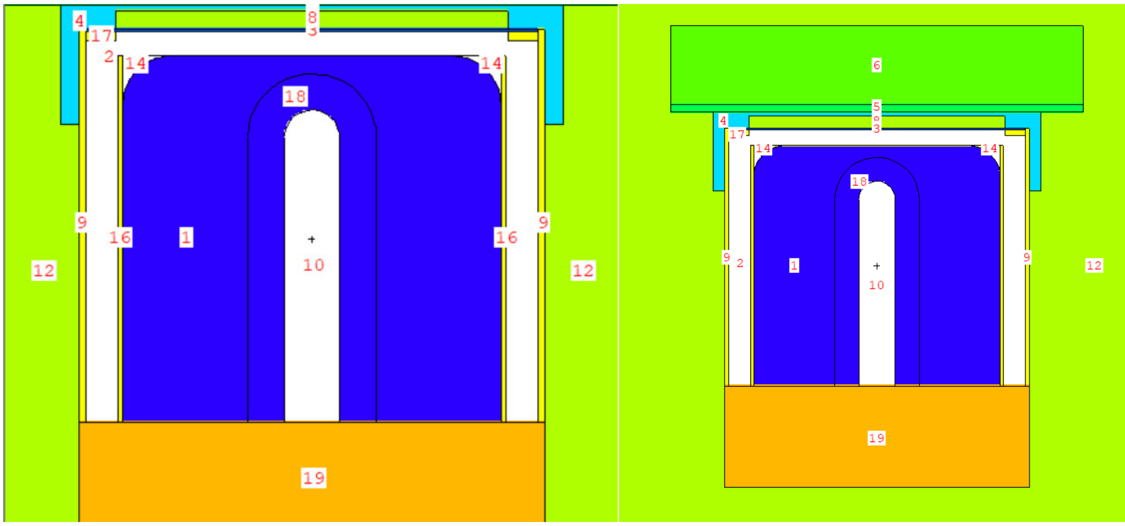

Fig. 1. The graphic output of the MCNP6 computer code: Left - Geometry of Detector, Right - Geometry of Detector and Sample Box

Note for Fig. 1:

1: Active volume of Detector's Crystal
2: Vacuum outside the Detector's Crystal
3: Beryllium window
4: Polystyrene Base
5: Bottom of box containing reference sample
6: Reference sample
8: Air buffer
9: Protective aluminum layer
10: Vacuum inside the Detector's Crystal
12: Air outside the detector
14: Upper layer of the Detector's Crystal
16: Dead layer outside the Detector's Crystal
17: Aluminum layer for supporting the Beryllium window
18: Dead layer (inside the Detector's Crystal and outside Vacuum column)
19: Copper base for supporting Detector's Crystal

In this study, we aim to compare the gamma spectra obtained from MCNP simulation, which include the measured background spectrum, with the measured spectra of the IAEA reference samples. This comparison was performed by using the InterSpect software [12], with the objective of identifying any abnormal peaks present in the Nucléide-Lara and NuDat based simulated spectra. These abnormal peaks are defined as those with a significantly different peak area compared to both the measured spectrum and simulated spectrum from the remaining data source among the Nucléide-Lara and NuDat.

A new dataset, after that, was developed on the combination of Nudat and Nucléide-Lara databases by adjustment of the intensity of energies (intensity of gamma emission) which contributed to abnormal peaks according to the corresponding peaks in the measured spectrum. This is expected to remove identified abnormal peaks in simulated spectrum in comparison with measured spectrum.

This combined dataset was then re-checked by MCNP simulation and compared with the measured spectrum to check the agreement between the simulation results and the measured results and evaluate the quality of the simulation performed in the study. In addition to using the RGTh-1 and RGU-1 reference samples, the new dataset is further used to simulate the gamma spectrum of the IAEA-447 (m=144.4 gam, 425 Bq/kg of Cs-137, 420 Bq/kg of Pb-210, 423 Bq/kg of Po-210, 37 Bq/kg of Pb-212, 25.1 Bq/kg of Ra-226, 37 Bq/kg of Ac-228, 21.8 Bq/kg of U-234, 22.2 Bq/kg of U-238, 0.15 Bq/kg of Pu-238, 5.3 Bq/kg of Pu-239+240, 550 Bq/kg of K-40, 37.3 Bq/kg of Th-232, 2.2 Bq/kg of Am-241, 8 Bq/kg of Pu-241).

The combined dataset was also used in MCNP simulation to discover the difference between MCNP's library versions of interaction cross-section for photons, including 01p, 02p, 03p, 04p, 12p, 63p and 84p.

III. RESULT AND DISCUSSION

A. Study on the influence of nuclear data of photon decay processes on the quality of the simulated spectra

The MCNP simulation results based on the individual datasets of NuDat and Nucléide-Lara which were implemented on the RGTh-1 and RGU-1 samples are shown in Fig. 2 to 4 *(in these figures, the spectra are represented by three different components: the Foreground (depicted in black) corresponds to the Nucléide-Lara-based spectrum, the Background (shown in violet) represents the measured spectrum, and the Second Foreground (illustrated in green) corresponds to the NuDat-based spectrum)*.

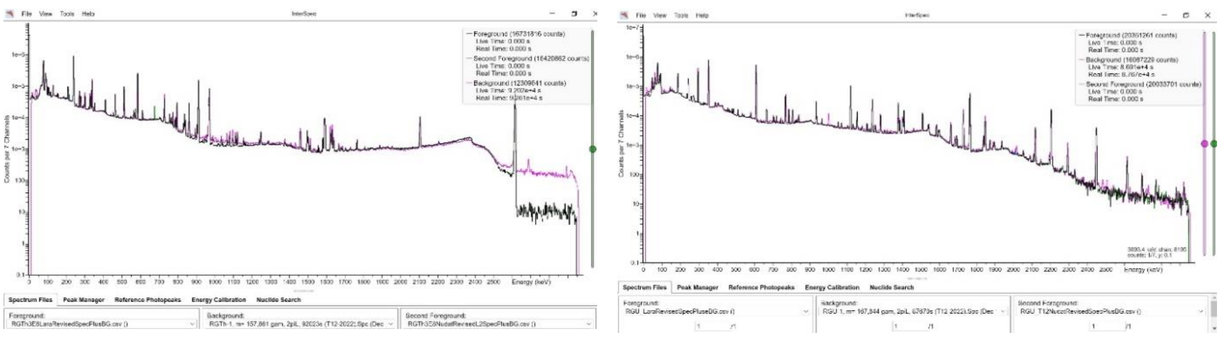

Fig. 2. Comparison of simulated gamma spectrum based on NuDat and Nucléide-Lara against measured spectrum for over full energy range: Left - RGTh-1 sample, Right - RGU-1 sample

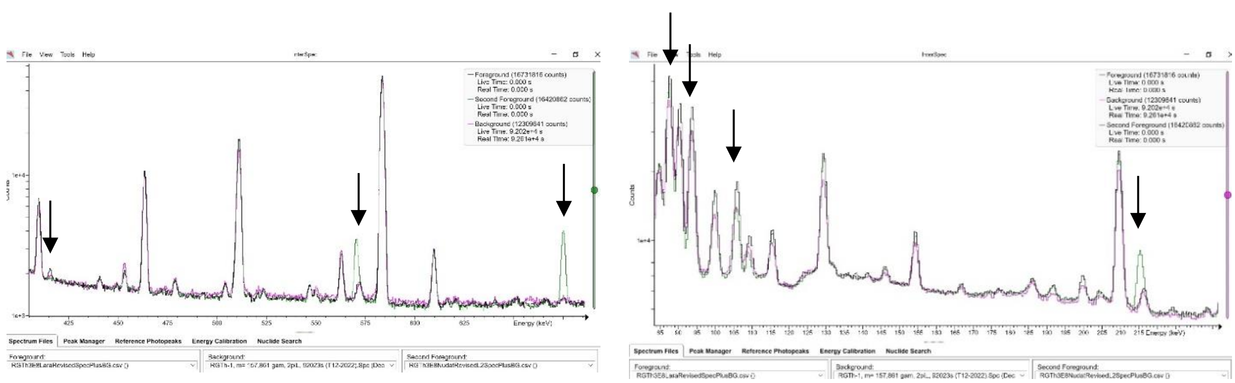

Fig. 3. Comparison of simulated gamma spectrum based on NuDat and Nucléide-Lara against measured spectrum of RGTh-1 sample at abnormal peaks - from NuDat: 215 keV, 571 keV and 675 keV; from Nucléide-Lara: 90 keV, 94 keV, 106 keV, 416 keV

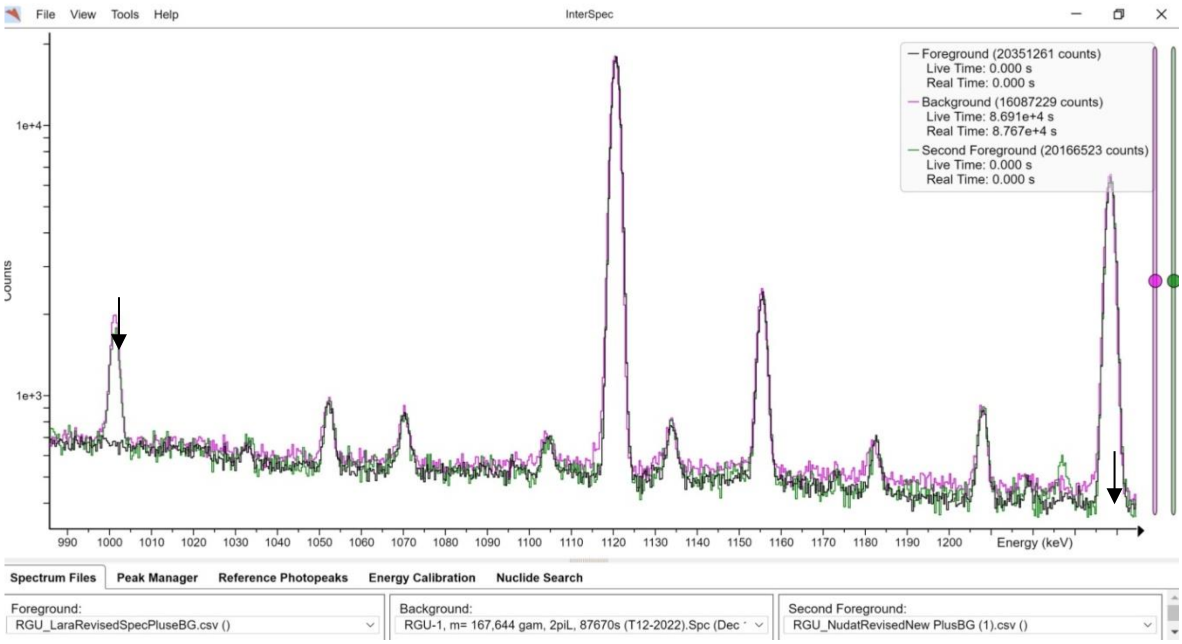

Fig. 4. Comparison of simulated gamma spectrum based on NuDat and Nucléide-Lara against measured spectrum of RGU-1 sample at abnormal peak: 1227 keV (from NuDat), 1001 keV (no peak from Nucléide-Lara)

Comparison of simulated gamma spectra based on NuDat and Nucléide-Lara against measured spectra of RGTh-1 and RGU-1 samples over full energy range in the Fig. 2, in general, shows the good correlation between the simulated and measured spectra within the range of 50 keV to 2620 keV. There was an abnormality in the simulation result at the energy higher than 2620 keV. Since the energy in this range is outside the peaks of interest, it is not necessary to concern this region.

As shown in the Table I, the results of the quantitative comparison of correlation between the simulated and measured spectra were done through mean deviation (MD) by using the following equation:

$$MD = \frac{1}{k-n+1}\sum_{i=n}^{k}|N_i - M_i|$$

Where n and k are the index of the smallest and the biggest channel within defined energy range; N_i and M_i are the counts of the channel i in the spectrum N and spectrum M, respectively.

It should also be noted that this comparison was made for the part of the spectrum in the energy range of 50 keV to 2620 keV to avoid significant disturbances outside this range. This result confirms that, overall, the difference between the Nucléide-Lara and NuDat data is not significant. In addition, for the RGTh-1 sample, the simulation based on the NuDat dataset gave results that were closer to the measured spectrum than the simulation based on the Nucléide-Lara dataset. However, for the RGU-1 sample, the result is the opposite. Therefore, it is not confirmed whether Nucléide-Lara or NuDat data give better simulation results.

Table I. Quantitative comparison of simulated and measured spectra

Sample	**Comparison of 2 pair of spectra**	**Mean deviation** (within the range of 50 keV to 2620 keV)
RGTh-1	Nucléide-Lara-based simulated spectra against measured spectrum	163
	NuDat-based simulated spectra against measured spectrum	156
	Nucléide-Lara-based simulated spectra against NuDat-based simulated spectra	6.78
RGU-1	Nucléide-Lara-based simulated spectra against measured spectrum	293
	NuDat-based simulated spectra against measured spectrum	301
	Nucléide-Lara-based simulated spectra against NuDat-based simulated spectra	6.88

The Fig. 2 also shows the very good agreement between 02 simulated spectra based on NuDat and Nucléide-Lara in the full range, except for several abnormal peaks which will be further discussed below. This demonstrates that except for some anomalies, the nuclear data of NuDat and Nucléide-Lara are quite similar.

Specifically, the Fig. 3 and 4 show the existing of several abnormal peaks, including 215 keV, 571 keV, 675 keV and 1227 keV of the NuDat-based simulated spectra, which were present neither in the measured spectrum nor Nucléide-Lara-based simulated spectra (two latter spectra are good agreement at these peaks). Similarly, the Fig. 3 also show the existing of several abnormal peaks, including 90 keV, 94 keV, 106 keV and 416 keV of the Nucléide-Lara -based simulated spectra, which were present neither in the measured spectrum nor NuDat-based simulated spectra. The Fig. 4 present the peak of 1001 keV in both NuDat-based simulated and measured spectrum, but not existing in Nucléide-Lara -based simulated spectra.

The contribution of main energies, with corresponding intensities, to abnormal peaks is named in Table II. For example, the peak of 215 keV was corresponding to the energy of 214.85 keV with the intensity of 0.76% from the decay of Ac-228 and the energy of 214.983 keV with the intensity of 0.247 % from the decay of Po-212 (both in the Th-232 chain). Both energies were from the NuDat nuclear dataset and they did not exist in the Nucléide-Lara. These abnormal peaks, therefore, indicate issues with accuracy and completeness of the existing nuclear datasets from both NuDat and Nucléide-Lara, potentially at energies and their intensities identified in the column (4) of Table II.

Table II. Identified energies from both NuDat and Nucléide-Lara nuclear datasets contributing to abnormal peaks

No.	Abnormal peaks (keV)	Corresponding peak area	Energies contributing to abnormal peaks			
			Energy (*) (keV) - Intensity (%)		Nuclide (Decay chain)	Nuclear Data Library
(1)	(2)	(3)	(4)		(5)	(6)
1	215	Nucléide-Lara-based simulated/measured spectra: 6383 NuDat-based simulated spectra: 23119	214.85	0.76	Ac-228 (Th-232)	NuDat
			215.983	0.247	Th-228 (Th-232)	
2	571	Nucléide-Lara-based simulated/measured spectra: 2838 NuDat-based simulated spectra: 14837	570	2	Po-212 (Th-232)	
			570.91	0.182	Ac-228 (Th-232)	
3	675	Nucléide-Lara-based simulated/measured spectra: 1051 NuDat-based simulated spectra: 17513	674.75	2.1	Ac-228 (Th-232)	
4	1227	Nucléide-Lara-based simulated/measured spectra: 271 NuDat-based simulated spectra: 575	1226.7	0.13	Bi-214 (U-238)	
5	90	NuDat-based simulated/measured spectra: 60095 Nucléide-Lara-based simulated spectra: 81222	89.8087	0.244	Bi-214 (U-238)	Nucléide-Lara
			89.95	1.01	Th-231 (U-235)	
			89.954	2.5	Ac-228 (Th-232)	
			89.954	3.56	U-235 (U-235)	
			90.0757	1.77	Pb-212 (Th-232)	
			90.0757	1.1	Pb-214 (U-238)	
			90.885	1.16	Pa-231 (U-235)	
6	94	NuDat-based simulated/measured spectra: 99549 Nucléide-Lara-based simulated spectra: 124370	94.666	2.18	Pa-234 (U-238)	
			94.8547	8.5	Ra-223 (U-235)	
7	106	NuDat-based simulated/measured spectra: 33919 Nucléide-Lara-based simulated spectra: 49715	105.554	1.5	Ac-228 (Th-232)	
			105.554	2.05	U-235 (U-235)	
			106.1133	0.136	Pa-231 (U-235)	

8	416	NuDat-based simulated/measured spectra: 862 Nucléide-Lara-based simulated spectra: 1752	415.272	0.144	Pb-212 (Th-232)	
9	1001	NuDat-based simulated/measured spectra: 1979 Nucléide-Lara-based simulated spectra: 0	Not existing			
Note (*): The energies with small intensity are neglected.						

Based on input from the new dataset of 1693 energies of photons and their intensities from 40 nuclei by combining Nudat and Nucléide-Lara databases, MCNP simulations were performed again for RGTh-1, RGU-1 and the IAEA-447 samples. Spectra were compared as shown in Fig. 5 to 7. Accordingly, there were no longer abnormal peaks and therefore it can be confirmed that this dataset gave better simulation results than with those using single Nudat and Nucléide-Lara databases, which was closer to the measured spectrum.

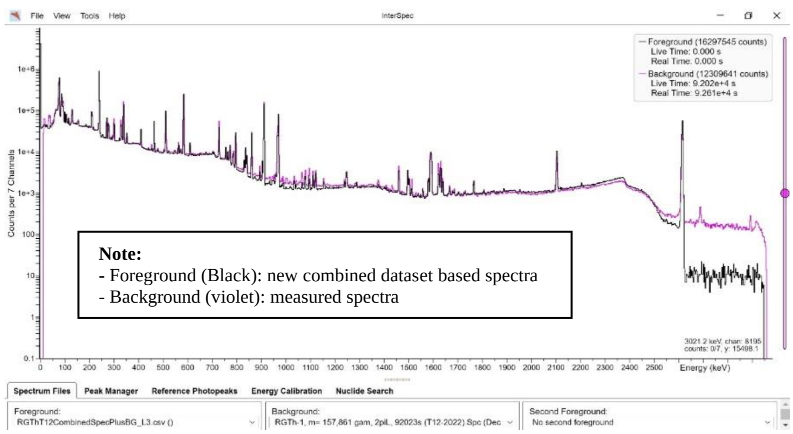

Fig. 5. Comparison of simulated gamma spectrum based on the new combined dataset against measured spectrum for over full energy range (for RGTh-1 sample)

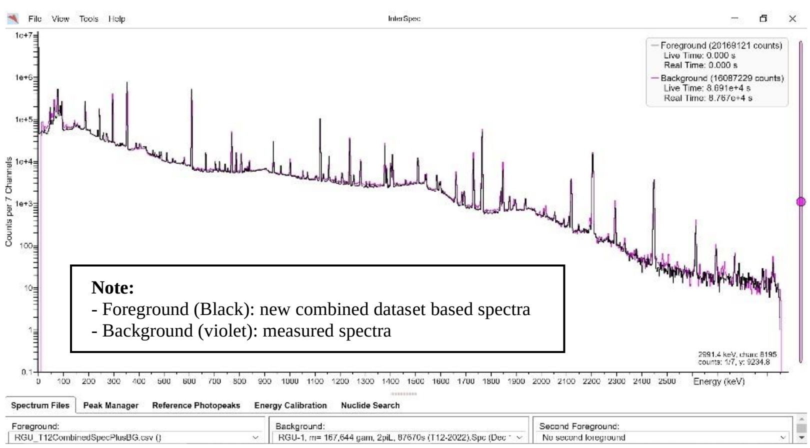

Fig. 6. Comparison of simulated gamma spectrum based on the new combined dataset against measured spectrum for over full energy range (for RGU-1 sample)

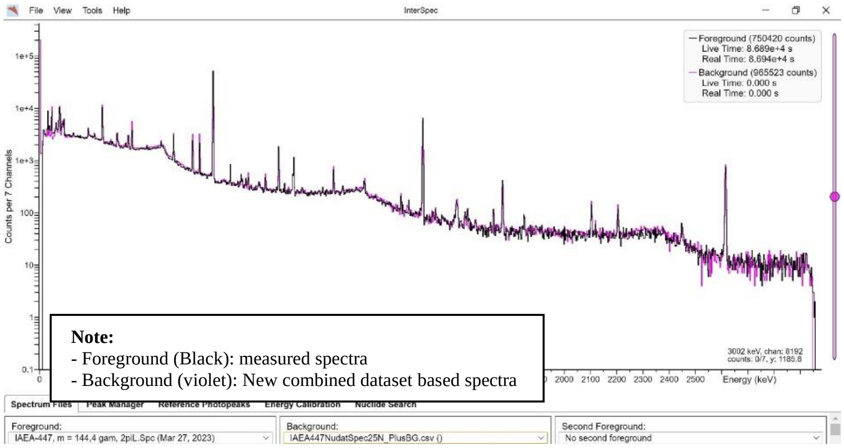

Fig. 7. Comparison of simulated gamma spectrum based on the new combined dataset against measured spectrum for over full energy range (for IAEA-447 sample)

3.2. Study on the influence of cross-section of photon interactions on the quality of the simulated spectra

An investigation was conducted to examine the influence of the cross-section of photon interactions on the quality of simulated spectra. The study utilized selected versions of MCNP's library for photon interaction cross-sections, namely 01p, 02p, 03p, 04p, 12p, 63p, and 84p. The simulation results revealed no significant variation between the 01p, 02p, and 03p libraries, as well as among the 04p, 12p, 63p, and 84p libraries. However, a slight difference was observed within the energy range of 410 keV to 440 keV, as depicted in Fig. 8, between these two groups. Notably, the spectra from the latter group exhibited a stronger correlation with the measured spectra. These findings indicate that the impact of photon interaction cross-sections on the quality of simulated spectra is minimal. Nevertheless, it is recommended to employ the more updated versions, namely 04p, 12p, 63p, and 84p libraries, for improved simulation results.

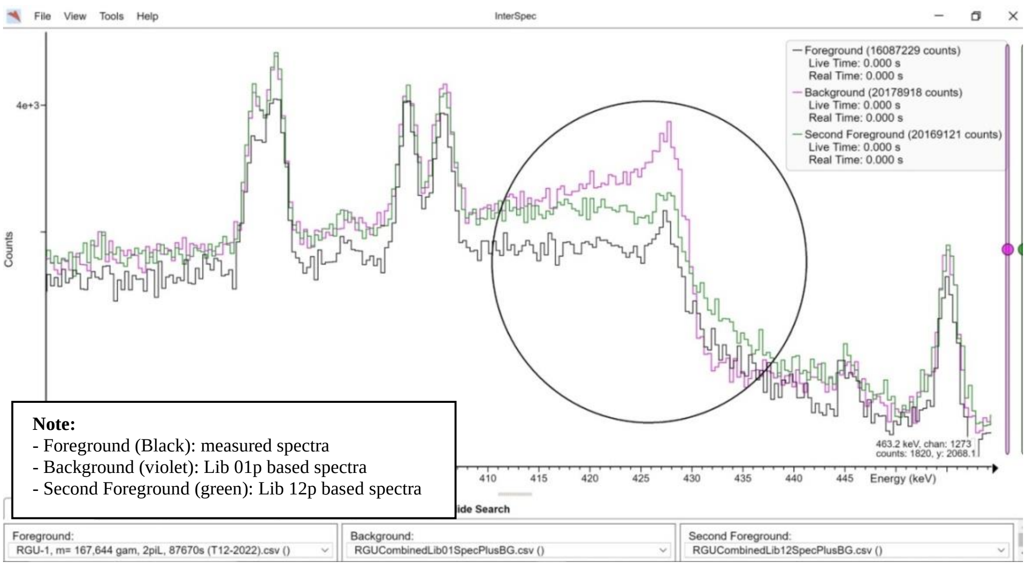

Fig. 8. The difference between gamma spectra using various MCNP's library versions of interaction cross-section for photons (in the circle mark)

IV. CONCLUSIONS

The simulation of gamma spectra from soil samples by using MCNP performed in this study, based on combined dataset, is adequate to be used for the future problem of generating a large simulated dataset for training and testing Machine Learning (ML) models that automatically identify and quantify radioactive isotope from gamma-ray spectra of soil samples in the analysis of environmental radioactivity for configuration of type-n HPGe detector and 2piL-type plastic sample container. It is noted that only the energy range between 50 keV and 2620 keV was considered because this range contains the energy peaks of interest and removes noise outside this range. To be able to apply to other configurations, it will be necessary to adjust some geometry related parameters in the input of MCNP simulation.

The very good agreement between 02 simulated spectra based on NuDat and Nucléide-Lara in the full energy range demonstrates the similarity of the nuclear data of NuDat and Nucléide-Lara. The existence of some abnormal energy peaks from the NuDat and Nucléide-Lara -based simulated spectra, however, raises a need of reviewing the accuracy and completeness of these dataset, especially at energies identified in the Table II.

The impact of photon interaction cross-sections data on the quality of simulated spectra is negligible. However, it is still suggested to employ the more updated library versions for improved simulation results.

V. ACKNOWLEDGEMENTS

The case study was conducted as a part of the Ministerial Project ĐTCB.03/23/VKHKTHN titled "Study on the Application of Artificial Intelligence (AI) for isotope identification and qualification from gamma-ray spectra getting from HPGe-type Detectors in environmental radiation research" as assigned by Ministry of Science and Technology to Institute of Nuclear Science and Technology from 2023 to 2024.

VI. REFERENCES

[1]. Turner, A. *et al.* Convolutional Neural Networks for Challenges in Automated Nuclide Identification. *Sensors* **21**, 5238 (2021).

[2]. Gauld, I. C. *et al.* Quantifying Nuclear Data Uncertainties in National Security Applications.

[3]. National Nuclear Data Center. NuDat 3.0. *NuDat 3.0*.

[4]. Laboratoire National Henri Becquerel. Nucléide-Lara. *Nucléide-Lara*.

[5]. 5. Conti, C. C., Salinas, I. C. P. & Zylberberg, H. A detailed procedure to simulate an HPGe detector with MCNP5. *Progress in Nuclear Energy* **66**, 35–40 (2013).

[6]. Hendricks, J. S., Swinhoe, M. T. & Favalli, A. *Monte Carlo N-Particle Simulations for Nuclear Detection and Safeguards: An Examples-Based Guide for Students and Practitioners*. (Springer International Publishing, 2022). doi:10.1007/978-3-031-04129-7.

[7]. Elanique, A. *et al.* Dead layer thickness characterization of an HPGe detector by measurements and Monte Carlo simulations. *Applied Radiation and Isotopes* **70**, 538–542 (2012).

[8]. Chuong, H. D., Thanh, T. T., Ngoc Trang, L. T., Nguyen, V. H. & Tao, C. V. Estimating thickness of the inner dead-layer of n-type HPGe detector. *Applied Radiation and Isotopes* **116**, 174–177 (2016).

[9]. Huy, N. Q., Binh, D. Q. & An, V. X. Study on the increase of inactive germanium layer in a high-purity germanium detector after a long time operation applying MCNP code. *Nuclear Instruments and Methods in Physics Research Section A: Accelerators, Spectrometers, Detectors and Associated Equipment* **573**, 384–388 (2007).

[10]. Loan, T. T. H., Ba, V. N., Thy, T. H. N., Hong, H. T. Y. & Huy, N. Q. Determination of the dead-layer thickness for both p- and n-type HPGe detectors using the two-line method. *J Radioanal Nucl Chem* **315**, 95–101 (2018).

[11]. Mezerreg, N., Azbouche, A. & Haddad, M. Study of coincidence summing effect using Monte Carlo simulation to improve large samples measurement for environmental applications. *Journal of Environmental Radioactivity* **232**, 106573 (2021).

[12]. William Johnson. InterSpec gamma radiation analysis software. (2018).