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Calculating radiation dose rate from online real-time environmental γ-spectrum using NaI(Tl) detector

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Abstract: Calculating gamma radiation dose rate from online real-time environmental gamma spectrum using NaI(Tl) detector has been developed into a software named RADAPROC V.1 in the Center for Operating the National Network of Environmental Radiation Monitoring And Warning (CONNERMAW). Currently, hundreds of online gamma spectra per day from online monitoring stations are processed to calculate the total ambient dose equivalent rate and the ambient dose equivalent rate of typical natural radioactive isotopes such as K-40, Bi-214, Tl-208 according to the method of using the function *G(E)* and the photo-peak area method. The calculated results have been compared with the results of calculating the dose rate from the specific activity of radioactive isotopes in soil samples collected at the same monitoring location and analyzed in the laboratory. The difference between the methods is less than 25%. The ambient dose equivalent rates of typical natural radioactive isotopes are a bit higher than those calculated with SARA-NMC software. The software will be improved shortly for better results.

Keywords: *Gamma radiation dose rate; online NaI(Tl) spectrum; G(E) function; CONNERMAW; RADAPROC V.1; Ka; H*(10); K-40; Bi-214 and Tl-208.*

I. INTRODUCTION

Gamma spectrometer using NaI(Tl) detector is used quite popular in the world to determine the environmental gamma dose rate in many different cases such as routine monitoring or emergency response. Because it is highly sensitive to gamma rays, useful for measuring low-level dose rates, and quite stable under environmental conditions. The photo-peak area method has been developed first by Back H.L. and et al since 1972 [1], then simulation method by Monte Carlo technique, method of using *G(E)* function, method of conversion coefficients have been derived [3 - 8, 10 and 12].

Online radioactive monitoring networks of many countries around the world such as Korea, Japan, Germany, Hongkong, Bulgaria, Russia, Ukraine ... all use NaI(Tl) detectors. Each online monitoring and alert network has central management software, operating the entire network and integrating online data automatically, systematically, data processing and results display as the requirement, and can automatically alert all abnormal radioactive situations 24/24 hours.

In our country, according to the decision on the planning of the national environmental radioactive monitoring and

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warning network No. 1636/QD-TTg, signed on August 31, 2010, the National Network will include a National Control Center, 4 Regional Stations, and 16 Local Stations. Center for Operating the National Network of Environmental Radiation Monitoring And Warning (CONNERMAW) is located in the Institute of Nuclear Science and Technology (INST). This Center has been building and put into operation continuously, online since the end of 2015. However, the online monitoring systems in use today are designed and manufactured by various measuring technologies. The online transmitting and storing of monitoring data are also conducted by different technologies (technologies of Korea, Japan, Germany, and Vietnam). Especially, the database type is very different, heterogeneous, even the original data is encrypted. At each system, measured data is transmitted to a separate Server, so it is not systematically manageable, cannot integrate simultaneously, and display the environmental radiation status at any desired time.

Every day, the Operations Center receives hundreds of gamma radiation spectra from the stations but cannot process them automatically. To overcome this difficulty, software for calculating gamma radiation dose rate from online real-time environmental gamma spectrum using NaI(Tl) detector has been developed named RADAPROC V.1 in the CONNERMAW.

II. METHOD OF CALCULATING GAMMA RADIATION DOSE RATE

INST is operating three different types of online NaI(Tl) gamma spectrometer. These are SARA (made in Germany), RADMON (Vietnam), and NAH (Japan) spectrometers. Gamma spectral data obtained from these spectrometer systems have very different structures and formats. The processing of these gamma spectra is therefore very complicated. Calculating the gamma radiation dose rate from online real-time environmental gamma spectra acquired with NaI(Tl) detector has been researched and put into application.

A. Energy calibration

Energy calibration for gamma spectrometers is extremely important and necessary. That is to establish the relationship between the peak positions in the gamma spectrum and the corresponding gamma radiation energy. This dependency usually takes the following polynomial form.

$$E(ch) = a.ch^2 + b.ch + c \quad (1)$$

Where:

ch: Channel position in gamma radiation spectrum.

E(ch): Corresponding gamma radiation energy in the channel ch (keV).

a, b and c are the constant coefficients that will be determined using the least-squares method.

For the NaI(Tl) gamma spectrum, photo-peaks 609.3 keV (Bi-214), 1460.8 keV (K-40), 1764.5 keV (Bi-214) and 2614.4 keV (Tl-208) are commonly used. Based on the real spectral form, the corresponding channel position will be the channel with the maximum count in the energy regions of interest (Fig.1 and Tab.I).

For a RADMON spectrometer, the photo-peak locations and the corresponding gamma radiation energies are shown in Table 1. The found constant coefficients of the energy calibration curves are: a = 9E-05; b = 2.4404; c = 45.497 and the found energy calibration curve (Fig.2) is:

$$E\ (keV) = 9E\text{-}05.ch^2 + 2.4404.ch + 45.497 \quad (2)$$

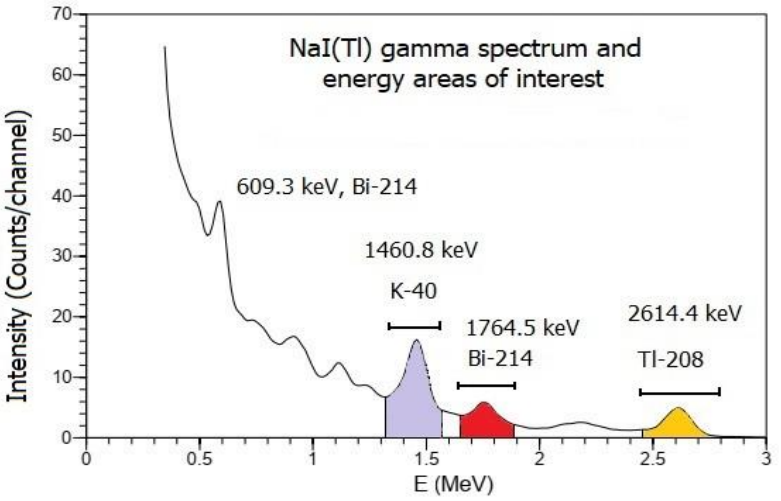

Fig. 1. Illustration of a NaI(Tl) gamma spectrum and energy regions of interest

Table I. Photo-peak locations and energy regions of interest in a NaI(Tl) gamma spectrum

Radiation isotopes	Photo-peak locations	Corresponding γ radiation energies, keV	Energy regions of interest, keV [4]
Bi-214	229	609.3	(520 – 690)*
K-40	568	1460.8	E1: 1370 - 1570
Bi-214	687	1764.5	E2: 1660 - 1860
Tl-208	1015	2614.4	E3: 2410 - 2810

* *The author's choice.*

The energy calibration curves for gamma spectra obtained from the SARA and NAH spectrometer systems were also performed similarly, but the position of the photo-peak in spectra would be different due to the different amplification factor of each spectrometer system.

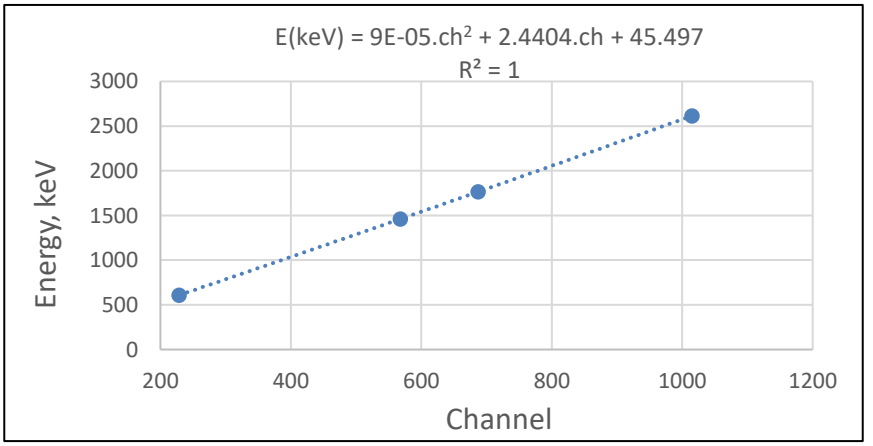

Fig. 2. Energy calibration curve for RADMON spectrometer

B. Calculating air kerma rate in free air (Ka) using G(E) function method

The method of calculating the air kerma rate in free air obtained from NaI(Tl) gamma spectrometer systems using the *G(E)* function has been studied by Moriuchi and Miyanaga [8] and it has been applying quite popular in the world. Accordingly, the function *G(E)* for cylindrical-shaped NaI(Tl) scintillation detector with size 3x3 inches has the following polynomial form [8]:

$$G(E) = \sum_{K=1}^{Kmax} A(K).(log_{10}\, E)^{K-M-1} \qquad (3)$$

Where:

Kmax = 16 (degree of polynomial);

M = 3 (integer determined by the size of the detector);

A(K): is the coefficients of polynomial identified and shown in Table II [14].

Then, the air kerma rate in free air is calculated by the following formula:

$$D(E) = \sum_{E=50keV}^{3000keV} N(E).G(E) \qquad (4)$$

Where:

D(E): Air kerma rate is unit nGy/h;

N(E): Counts per minute (cpm);

G(E): Value of function *G* at energy *E* (nGy/h/cpm).

Table II. The coefficients *A(K)* for converting to air kerma rate

A(1)	4.64477815E+01	*A(9)*	3.71066321E+00
A(2)	-7.53203815E+01	*A(10)*	-1.76560076E+00
A(3)	-2.36700398E+01	*A(11)*	6.24042285E-01
A(4)	1.33979261E+02	*A(12)*	-1.13338185E-01
A(5)	-1.16280743E+02	*A(13)*	-1.20970045E-03
A(6)	3.84485329E+01	*A(14)*	4.45227288E-03
A(7)	2.12501936E+00	*A(15)*	-7.38983490E-04
A(8)	-6.65937345E+00	*A(16)*	4.04368069E-05

To ensure higher accuracy, the above sum function was determined by the approximation fractional integration (trapezoidal method). The above formula can be rewritten as follows:

$$D(E) = \int_{50keV}^{3000keV} N(E).G(E).dE \qquad (5)$$

For terrestrial radiation fields, the following equation was used to convert the air kerma rate *D(E)* into ambient dose rate equivalent rate *H*(10)* in nSv/h [15].

$$H^*(10) = 1.21\, D(E) + 1.26 \qquad (6)$$

C. Calculating γ-ray ambient dose equivalent rate of interesting radionuclides with photo-peak area method

NaI(Tl) gamma-ray spectroscopy system provides a practical way to characterize dispersed radionuclides in or on the soil to

ascertain possible changes in the environmental radioactivity. Therefore, online or in-situ gamma spectroscopy is often used for monitoring and assessment of radioactivity and radiation dose rates in the environment due to both natural and anthropogenic sources. The full-energy peak (photo-peak) count rate of the measured radionuclide is one of the factors when calculating its gamma radiation dose rate. So, sometimes it is named the photo-peak area method.

The theoretical principles of this method were developed by Back H.L. et al. (1972) [1]. The principal three factors formula as follows:

$$\frac{N_f}{I} = \frac{N_f}{N_0} \cdot \frac{N_0}{\phi} \cdot \frac{\phi}{I} \quad (7)$$

Where: N_f is the photo-peak count rate of the measured radionuclide (in counts per second);

N_o is the photo-peak count rate of that radionuclide for a parallel beam of gamma-rays that is incident on the detector parallel to its symmetry axis, is the gamma-ray un-scattered flux on the detector ($cm^{-2}.s^{-1}$);

I is the exposure rate (R/h);

ϕ/I is the ratio of the flux due to gamma-rays of energy E to the corresponding exposure rate for that nuclide; this value was taken from Beck's tabulated data [1] and it is expressed in ($\gamma.s^{-1}.cm^{-2}/R.h^{-1}$);

N_f/N_o is angular correction factor, which for the detector used was assumed to be one, in the energy range of interest (50-3000 keV);

N_o/Φ is the count rate under a peak area due to unit flux of energy E incident of the detector parallel to its axis of symmetry. It is determined experimentally and it is express in $cps/\gamma.s^{-1}.cm^{-2}$ [9].

For a cylindrical 3×3 inches NaI(Tl) detector, Rahman has constructed the dependence of N_o/Φ on E as follows:

$$\ln(N_o/\Phi) = 4.48\text{-}1.03\ln E \quad (8)$$

Where, E is in MeV.

For scintillation NaI(Tl) gamma-ray spectrum, it is quite difficult to calculate the gamma dose rate due to radionuclides with energies lower than hundreds of keV. Therefore, it is usually to calculate only the gamma radiation dose rate through some typical radioisotopes such as K-40 (1460.8 keV), Bi-214 (1764.5 keV) represents for U-238 decay series and Tl-208 (2614.4 keV) represents for Th-232 decay series.

From Beck's tabulated data and equation (7), the values of No/Φ and Φ/I have been derived and shown in Table III.

The gamma exposure dose rate in term of ambient dose equivalent rate $H^*(10)$ due to each or all radionuclides presented in the spectrum can be calculated by the formula:

$$D = k1.k2 \sum_i \frac{(N_f)_i}{(N_f/I)_i} \quad (9)$$

Where, the sum is extended over all the peaks registered by the detector;

$(N_f)_i$ are the counts per second of the peaks experimentally measured;

$k1$ is the conversion factor from Roentgen to Gray ($k1$ = 8760);

$k2$ is the conversion factor from Gray to Sievert ($k2$ = 1.07 for natural background) [2].

Table. III. Values of No/Φ and Φ/I for a cylindrical 3×3 inches NaI(Tl) detector

Radionuclide	E, keV	No/Φ (cps/γ.s^{-1}.cm^{-2})	Φ/I (γ.s^{-1}.cm^{-2}/R.h^{-1})
K-40	1460.8	0.04854	0.2030
Bi-214	1764.5	0.03996	0.0296
Tl-208	2614.4	0.02665	0.0592

III. RESULTS AND DISCUSSION

A. Determination of N_f for ambient dose equivalent rate of K-40, Bi-214 and Tl-208

N_f is the photo-peak count rate of the measured radionuclide (in counts per second). Each on-line gamma spectroscopy measurement is usually performed for 10 minutes, so the count/channel in the energy regions corresponds to the 3 radioisotopes of interest (Table I). To determine N_f more accurately, the obtained spectral data are often smoothed before treatment. The following 5-point smoothing formula of Savitzky-Golay (1964) [11] is commonly used:

$$N'_{-2} = \frac{1}{35}[31.N_{-2} + 9.N_{-1} - 3.N_0 - 5.N_1 + 3.N_2] \quad (10)$$

Where: N and N' are the count before and after the smoothing respectively.

Then, the photo-peak area in the energy region *Ei* is calculated by the following formula:

$$N_f(Ei) = \frac{1}{T}\sum_{j=Li}^{Ri} N'(j) - \frac{N'^{(Li)} + N'(Ri)}{2}.(Ri - Li + 1) \quad (11)$$

Where:

$N_f(Ei)$ is in unit cps (count in 1 sec);

$i = 1 \div 3$ (3 energy regions *E1*, *E2* and *E3* shown in Table I)

T = 600 (for spectrum acquired in 10 min)

T = 3600 (for spectrum acquired in 1 h)

T = 86400 (for spectrum acquired in 1 d)

Li is the left channel of the i-th interesting photo-peak

Ri is the right channel of the i-th interesting photo-peak

Substituting the calculated N_f into Equation (7), the gamma radiation dose rate (I) of each interesting radioisotope is determined.

The calculating the total ambient dose equivalent rate and the ambient dose equivalent rate of interesting radionuclides from online real-time environmental gamma spectrum using NaI(Tl) detector has been developed into a software named RADAPROC V.1. The software is using continuous at the CONNERMAW.

The calculated results using the RADAPROC.V.1 have been compared with those obtained with the SARA-NMC software (Table IV and Table VII). The average ambient dose equivalent rate of K-40, Bi-214 and Tl-208 in 2020 at 3 stations Cao Bang, Da Nang and Vinh provinces are presented in Table IV.

Table IV shows that the results calculated by RADAPROC V.1 are a bit higher than those calculated with SARA-NMC software. It looks quite good for K-40, but not so good for Bi-214 and Tl-208. This could be explained by a poor counting statistic at the photo peaks of 1764.5 keV (Bi-214), 2614.4 keV (Tl-208) and the technique of smoothing applied in the software. So, the software will be improved in the near future for better results.

Table IV. Average ambient dose equivalent rate of interesting radionuclides at some provinces in 2020

Station	2020 average ambient dose equivalent rate of K-40, Bi-214 and Tl-208 calculated with different softwares, (nSv/h)								
	RADAPROC V.1 (a)			SARA-NMC (b)			Ratio (a/b)		
	K-40	Bi-214	Tl-208	K-40	Bi-214	Tl-208	K-40	Bi-214	Tl-208
Cao Bang	8.538	6.558	8.279	5.577	4.772	5.672	1.531	1.374	1.460
Da Nang	20.262	8.399	23.394	21.970	7.374	13.666	0.922	1.139	1.712
Vinh	4.935	10.347	11.990	4.877	6.292	9.064	1.012	1.644	1.323
Average values	11.245	8.435	14.554	10.808	6.146	9.467	1.155	1.386	1.498

B. Evaluating results of ambient dose equivalent rate calculated with RADAPROC V.1

To evaluate the results of calculating the ambient dose equivalent rate (*H*(10)*) of gamma radiation using RADAPROC V.1 software, some soil samples around the position of the NaI(Tl) spectrometer were collected and analyzed. The specific activity of the interesting radioisotopes (K-40, Bi-214 and Tl-208) in those soil samples have been determined with a low-background high purity semiconductor gamma spectrometer with CANBERRA's HPGe GC5019 detector. The gamma spectrometer has an energy resolution at the peak of 1332.5 keV of Co-60 of 1.8 keV and relative efficiency of 50%. From the specific activity of the radioisotopes of interest, their respective gamma dose rates were calculated using the conversion factors for the natural radioactive series in secular equilibrium and having a uniform distribution in the ground and for K-40 (Table V).

Table V. Specific activity to gamma dose rates conversion coefficients (*CF*)

Isotope/ Radioisotopes	Conversion coefficient (*CF*)		
	CF for *Ka* [10] ($nGy.h^{-1}/Bq.kg^{-1}$)	*CF* for *Ka* [6] ($nGy.h^{-1}/Bq.kg^{-1}$)	*CF* for *H*(10)* [6] ($nSv.h^{-1}/Bq.kg^{-1}$)
Th-232 series	0.604	0.599	0.749
Tl-208	0.326	0.319 (53.2% total)	0.387 (51.7% total)
U-238 series	0.463	0.450	0.564
Ra-226	0.00125	0.00148	0.00228
Bi-214	0.401	0.390 (86.7% total)	0.480 (85.1% total)
K-40	0.0417	0.0427	0.0512

According to Lemercier et al. [6], *H*(10)* for area monitoring was calculated by the following formula:

$$H^*(10) = 0.0512 \times C(K\text{-}40) + 0.48 \times C(Bi\text{-}214)/0.851 + 0.387 \times C(Tl\text{-}208)/0.517 \quad (12)$$

Where *H*(10)* is ambient dose equivalent rate in unit of nSv/h and C(K-40), C(Bi-214) and C(Tl-208) are the activity concentrations of K-40, Bi-214 (U-238 series) and Tl-208 (Th-232 series) in unit of Bq/kg, respectively.

Specific radioactive activity concentrations of the interesting radioisotopes in the soil samples were presented in the following Table VI.

The results of comparing the total online gamma radiation dose rate, the dose rate calculated by RADAPROC V.1 and the analytical results of soil samples in the laboratory are presented in Table VII.

Table VI. Specific radioactive activity concentrations of the interesting radioisotopes in the soil samples collected quarterly around each station in 2020

Station	Number of samples	Specific radioactive activity, Bq/kg					
		K-40	Error	Bi-214	Error	Tl-208	Error
Cao Bang	4	832.14	11.62	45.62	0.55	30.24	0.35
Da Nang	4	861.21	29.05	46.61	1.57	72.64	2.44
Vinh	4	268.08	9.07	59.00	1.99	59.74	2.00

The comparing results in Table VII shown that gamma dose rates obtained from different methods of determination are quite similar to each other. The difference of results between the methods is less than 25%. Shuichi Tsuda (2016) [13] had also found that one could overestimate the air dose rates in the environment at the most by 20-30% using NaI(Tl) detectors. The software RADAPROC V.1 is useful and believable.

Table VII. The average radiation dose rate in 2020 obtained from the different methods of determination

Station	The total dose rate calculated from specific activities, nSv/h		SARA online total dose rate (a), nSv/h		Dose rate calculaed by RASAPROC (b), nSv/h		Ratio (b)/(a)
	Total dose rate	Error	Total dose rate	Error	Total dose rate	Error	
Cao Bang	90.50	1.97	90.29	6.19	109.90	7.80	1.22
Da Nang	123.50	7.21	132.36	8.08	162.90	10.20	1.23
Vinh	90.69	5.29	86.54	10.88	107.07	14.25	1.24

IV. CONCLUSIONS

The first version software of RADAPROC V.1 has been developed in the CONNERMAW. This software allows to calculating the total ambient dose equivalent rate and the ambient dose equivalent rate of interesting radionuclides such as K-40, Bi-214, Tl-208 from online real-time environmental gamma spectrum using NaI(Tl) detector.

The software RADAPROC V.1 has been using very effective running in the background at the CONNERMAW. Hundreds of gamma spectra are processing per day with this software. It helps greatly reducing the time as well as manpower in processing online monitoring data.

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