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A Method to Determine Doublet Photopeak Area ¹³⁷Cs and ²¹⁴Bi, in NORM Soil by NaI-detector

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ABSTRACT

Due to the poor resolution of (76 mm×76 mm) NaI scintillation detector, ~7.5% for ¹³⁷Cs peak (661.5 keV), a mutual spectral interferences of environmental samples between ¹³⁷Cs and ²¹⁴Bi (609 keV) in the decay series of uranium peaks, is a serious problem. This overlapped problem appears in low level sample spectrum causes overestimation of the specific activity of both nuclides. The main purpose of this work is to resolve this problem and to present a methodology to correct the activity concentrations of ¹³⁷Cs by NaI(Tl) spectroscopy. In Duhok governorate of Iraq, as a real NORM case, the accumulation of ¹³⁷Cs is significant brought by west-north wind and precipitation in the years following nuclear weapons testing and Chernobyl accident in 1986. The developed correlations for any ratio of the measured areas of ²¹⁴Bi and ¹³⁷Cs overcome the overestimation and show very good results. The method is approved by comparing the relative error of the measured and corrected activity of Bi with that measured Pb. The maximum, minimum and average relative errors decreased from 40%, 7.3% and 19.4% to 6.1%, -6.5% and 0.6% respectively.

Keywords: Doublet peak area, Environmental radioactive sources, NaI scintillation spectroscopy, Cesium-137

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INTRODUCTION

Generally speaking, the enhancement in concentration of Naturally Occurring Radioactive Materials (NORM) may be insignificant or relatively small, but a large amount of NORM could be disrupted from their natural state. This is most apparent in the case of the enhanced potential for human exposure as a result of development of nuclear activities since 1960s (Froehlich, 2009). Considerable amounts of ¹³⁷Cs were deposited on Europe and all over the world by fallout from the atomic bomb tests in the 1950s and 1960s. In early April 1986, the world's worst accident occurred at the Chernobyl nuclear power plant in Ukraine and last but not least the Japanese Fukushima Daiichi reactors accident in March 2011. Most of the fission products released will have completely decayed by now but ¹³⁷Cs radionuclide which entered into the environment and, consequently, into the food chain. Over the next decades, interest will continue to focus on ¹³⁷Cs (Fairlie & Sumner, 2006). Gamma spectrometry of NORM is difficult for a number of reasons. For instance, the low activity levels, difficulties of spectrometer background and dependency upon the local environment. There may also be evidence of contamination from neutron-capture and fission-product radionuclides, ⁶⁰Co and ¹³⁷Cs being fairly common examples and interference (the overlap of peaks in a spectrum) of two or more photo peaks. All of those difficulties are then compounded by the fact that there are a large number of mutual spectral interferences between many nuclides in the decay series of uranium and thorium. The general way in which we may approach low level or the NORM sample spectrum analysis is to select a limited number of gamma-rays for each nuclide of interest and search the spectrum only for these peaks via a nuclide library to avoid true coincidence summing TCS, interference and allow deconvolution if necessary (Gilmore, 2011). (76mm×76 mm) NaI(Tl) scintillation detector is the most commonly used in gamma spectroscopy. It is cheap and readily available but poor resolution. In handling sample spectra, singlet well-defined peaks detection,

measurement and calculation of peak area is straightforward. Unfortunately, peaks interference is frequent and it is often necessary to restrict the portion of the peak measured to minimize the effect of neighbouring, possibly overlapping, peaks. The fact that not all of the peak area is taken into account should be compensated. If two photopeaks are separated by something less than their photopeak width, it can be difficult to determine if the Gaussian represents one or two photopeaks. In this study, a methodology for the determination of the activity concentrations of the overlapped peaks ¹³⁷Cs and ²¹⁴Bi (the daughter of ²³⁸U) for a shielded (76 mm×76 mm) NaI(Tl) gamma spectroscopy is presented. The method depends on the measurement of resolution of the detector and data analysis or weighting factors of the nuclides such as the separation between the centroid peaks, non significant portion and the peak area ratio. Although the zone of ²¹⁴Pb (351 keV) has a big contribution of Compton effects, which exist in both samples and background measurements and is eliminated to a good extent by subtraction of the sample spectrum from the background. The resultant activity concentrations are verified by comparison of the concentration of $^{214}\rm{Bi}$ with that of $^{214}\rm{Pb}$ (351 keV) that can be measured with good statistics and both are in secular equilibrium. The surface soil and sediment samples are taken in Duhok governorate which is located at the northernwestern part of Iraq as a third governorate of Kurdistan region. It confined with Turkey from the north, Syria from the west, Arbil governorate from the east, and Nineveh governorate from the south. It is a part of the mountain and semi-mountain. Its variant topography is a critical factor for the distribution of ¹³⁷Cs (Abdullah & Ahmed, 2012; Abdullah & Ramadhan, 2011).

DETECTION EFFICIENCY

Detector efficiency is determined using 155 Eu, 22 Na and 137 Cs gamma standard sources. All single sources have the known energies and gamma decay fraction f_{γ} (number of gammas per disintegration of the nuclide for a transition at energy E) needed in calibrated process. The energy dependent detection absolute efficiency (photo-peak efficiency) has been determined by the following equation.

$$\varepsilon = \frac{N}{Af_{\gamma}t} \times 100\% \tag{1}$$

Where N is the photo-peak counts determined by the area of the (FWHM), A is the activity of the source at the date of work calculated from the known manufacturing date and activity and t is the live time of the measurements as been fixed by the MCDWIN software. Data are fitted and plotted using EXCEL program system to get the following relationship between the efficiency and gamma energies.

$$\varepsilon = 17.478e^{-0.0016E}$$
 With $\chi^2 = 0.9991$ (2)

RESOLUTION CALIBRATION

Since photopeaks are Gaussian with a tail, the centroid represents its central value, and the full width at half max (FWHM) represents its width at half of the counts associated with the centroid value. The FWHM (ΔE) is known for its

proportional relationship to the standard deviation, which is 2.36σ . A measurement called resolution is useful to ensure that all photopeaks are identified. Fractional energy resolution is a proportionality equation between the centroid (E) and the (Δ E),

$$\frac{\Delta E}{E}$$
 (3) Theoretically and experimentally it has been found that the

Theoretically and experimentally it has been found that the square of the fractional energy resolution has a linear relationship on NaI(IT) detectors of

$$\left(\frac{\Delta E}{E}\right)^2 = \frac{m}{E} + b \tag{4}$$

Where m is the slop and b is the intersection with y-axis and can be found by obtaining centroid and FWHM values from various standard radioactive sources and performing a weighted chi-squared minimization fit (Hurtado, García-León, & García-Tenorio, 2006). The resolution equation is determined using ¹⁵⁵Eu, ²²Na and ¹³⁷Cs gamma standard sources and modelled by EXCEL, as shown in Fig.1 and Eq(5).

$$\left(\frac{\Delta E}{E}\right)^2 = \frac{1.218}{E} + 0.004\tag{5}$$

The values of ΔE of 137 Cs (661.5 keV), 214 Bi (609 keV) and 214 Pb (352 keV) calculated by Eq (5). are 50.6, 47.2 and 30.4 keV respectively. The experimental centroid and FWHM data of various gamma standard sources used to construct this resolution equation are listed in Table 1.

Table 1. The centroid and FWHM data of various gamma standard sources

Source-nergy (E)	FWHM (ΔE)	$\frac{1}{E}$	$\left(\frac{\Delta E}{E}\right)^2$
¹⁵⁵ Eu 105	13	0.009524	0.015329
²² Na 511	42	0.001957	0.006755
¹³⁷ Cs 662	50.5	0.001511	0.005819
²² Na 1274	82	0.000785	0.004143

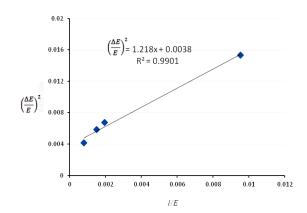


Fig 1. Resolution calibration of NaI detector

RESOLUTION OF ¹³⁷Cs and ²¹⁴Bi PEAKS

Of particular concern in the gamma spectrometry of NORM nowadays by NaI detector is its bad resolution compared to HPGE. The particular measure used in gamma

spectrometry is the Full Width of the peak at Half Maximum height (FWHM), expressed in keV. When the centers of peaks of are 3×FWHM apart, then the individual peaks are Gaussian and clearly separated, so the measurement of the peak area is straightforward. However, if the separation of two adjacent peaks is only one FWHM, as shown in Fig. 2, then simple measurement of each peak area is not possible.

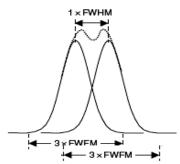


Fig 2. Centers of peaks separated by 1 FWHM (Gilmore, 2011)

Accordingly, we should be able to resolve such a double peak into its components with good accuracy, in an equivalent way of the mutual interference between ^{235}U (185.72 keV) and 226 Ra (186.21 keV). However, there is no method yet to resolve the interference and overlapping of ¹³⁷Cs peak (661.5 keV) and ²¹⁴Bi peak (609 keV) in NORM samples. For a standard (3 mm ×3 mm)NaI(Tl) detector used, the resolution of 7.5 %, is equivalent to an FWHM of 50 keV. Even, the resolution of a scintillation detector decreases as the energy increased and the peaks still become broader. These two peaks are close together by about one FWHM that deconvolution in real environmental spectra is unlikely to give results that one can have confidence in. It is, of course, possible to calculate the peak ratio by using gamma emission probabilities and the detector efficiencies for the two gammarays. However, that ratio will be in error. In principle, it would be possible to perform a peak striping operation using other peaks in the ²³⁸U spectrum to estimate its contribution to the 609 keV of ²¹⁴Bi peak. Fortunately, the emission probability of the previous intense peak at 352 keV of ²¹⁴Pb is 35.6% with no interference by any other peak and does not subject to the problems caused by True Coincidence Summing (TCS) (Gilmore, 2011).

METHODOLOGY

In principle, the methodology of correction of the measured areas under the peaks of ²¹⁴Bi and ¹³⁷Cs depends on the fact that these two peaks, after the subtraction of the sample spectrum from the background, are separated by about one FWHM. The derivation of a correlation of the net peaks areas is as following steps.

1-The spectrum analysis program MCDWIN version 2.74 is used to run the multi channel analyzer (MCD-2, 2002). The method of determination the area under the photopeaks of ^{214}Pb and ^{214}Bi is based on marking the lowest point of the spectrum to the left of the peak and fixating the ΔE to the corresponding photopeak (30.4 keV and 47.2 keV respectively). Now moving the cursor to the right side of the

peak until the fixate ΔE achieved with appropriate base line, as shown in Fig. 3 (for the sample S01as an example).

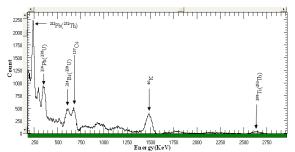


Fig 3. Gamma ray spectrum for a typical soil sample (S01) after subtracted from the background spectrum (full scale)

For 137 Cs, the area under the peak is determined by taking the lowest point to the right of the peak and fixating its ΔE to 50.6 keV. The cursor then must be moved to the left side of the peak until the same condition as before is satisfied. Fig. 3 shows gamma ray spectrum for a typical soil sample (S01) after subtracted from the background spectrum (full energy scale). While Fig.4 (energy scale zoomed) shows the way of determination of the area under each peak as described above for sample (S01) as an example.

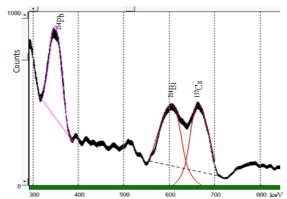


Fig 4. Determination the area under the photopeaks (energy scale zoomed)

2-After the peaks areas are measured as above, we can establish their statistical significance using the concept of the critical limits. The non-significant part of each peak, which should be subtracted from the referenced measured peak area, can be calculated as follows: From Fig.2, it is clear that there is one third of the total width of the two peaks overlapped. Since, this overlapped area is belonged to both peaks, then one half of this area is non-significant for each other peak (Gilmore, 2011). For the case of equal centroid height of the two peaks, we will have the peak area of the first peak C1 be equal to the peak area of the second C2, in other ward C1/C2 = 1. Therefore, the non significant overlapped area that must be subtracted from each peak is,

$$NS = \frac{1}{3} \frac{1}{2} = 0.167$$
 or $NS = 16.7\%$ (6)

While in the case of $C1 \neq C2$, the subtracted area is related inversely to the ratio of the two peaks areas,

$$NS1 = 0.167 \frac{c_2}{c_1}$$
 $NS2 = 0.167 \frac{c_1}{c_2}$ (7)

where NS1 and NS2 are the subtracted overlapped area from the measured area of the first C1 and second C2 peaks respectively.

3-In a practical case, it is obvious that the two peaks, 214 Bi and 137 Cs, are separated by a little bit higher than one FWHM, indeed by 661.5 -619 = 52.5 keV. This needs an extra correction of NS1 and NS2 according to the weighted Δ E's of both peaks, which can be done as follows.

$$NS1 = 0.167 \frac{c_2}{c_1} \frac{\Delta E_2}{\Delta WS}, \quad NS2 = 0.167 \frac{c_1}{c_2} \frac{\Delta E_1}{\Delta ES}$$
 (8)

Where $\Delta E1$ =47.2 keV is the FWHM of ²¹⁴Bi, $\Delta E2$ =50.6 keV is the FWHM of ¹³⁷Cs and ΔES =52.5 keV is the separation of the two peaks centroid. Accordingly, Eq (8) can be written for this case as,

$$NS1 = 0.16 \frac{c2}{c1}$$
, $NS2 = 0.15 \frac{c1}{c2}$ (9)

Depending on Eq (9). one can calculate the corrected peaks areas. The net area (corrected) of the peak N1 for 214 Bi and N2 for 137 Cs is simply the subtraction of these ratios NS1 and NS2 from one then multiply by the corresponding measured areas C1 and C2 to get,

$$N1 = (1 - 0.16 \frac{c2}{c1})C1$$
 , $N2 = (1 - 0.15 \frac{c1}{c2})C2$ (10)

Eq (10), represents best correlation for the net areas (N1 and N2) for any ratio of the measured areas (C1 and C2). Table 2 shows different values of C2/C1 and C1/C2 and the corresponding values of N1/C1 = 1-NS1 and N2/C2 = 1-NS2. Following the spectrum analysis, count rates for 214 Pb and 137 Cs detected photo-peak the activity for each of the detected nuclides are calculated. The activity A(Bq.kg $^{-1}$) is given by (Abdullah & Ramadhan, 2011).

$$A_i = \frac{N_i}{\varepsilon_i t. f_{\gamma_i.M}} (Bq. kg^{-1}) \tag{11}$$

Where ε_i is the detection photo-peak efficiency, N_i is the photo-peak counts determined (represent the measured areas of C_{214Pb} , C1 for ^{214}Bi and C2 for ^{137}Cs , and N1 for ^{214}Bi and N2 for ^{137}Cs), $f_{\gamma i}$ is a gamma decay fraction, t is the live time of the measurements and M is the mass in kg of the measured samples.

Table 2. Calculated values of N1/C1 = 1-NS1 for 214 Bi and N2/C2 = 1-NS2 for 137 Cs

$1\sqrt{2}/C2 - 1$	-1107 101	Cs			
C2/C1	NS1	1-NS1	C1/C2	NS2	1-NS2
3	0.48	0.52	3	0.45	0.55
2	0.32	0.68	2	0.30	0.70
1.5	0.24	0.76	1.5	0.23	0.78
1	0.16	0.84	1	0.15	0.85
0.75	0.12	0.88	0.75	0.11	0.89
0.5	0.08	0.92	0.5	0.08	0.93
0.25	0.04	0.96	0.25	0.04	0.96

ESULTS AND DISCUSSION

Among more than 30 samples collected and measured for different sites of Duhok government and because of similarity of some spectra, a total of 10 surface soil (S01-S10) and 3 (S11-S13) sediment samples is presented here. Depending on a standard methodology, the collected soil and sediment samples were collected and treated in the laboratory from air-dried to dry-weighed and stored for more than four weeks prior to counting to ensure equilibrium between decay products (Poschl & Nollet, 2006; Singh, Rani, & Mahajan, 2005). Gamma spectroscopic system consisted of a (3mm×3mm) NaI(TI) detector housed in a cylindrical lead shield of thickness ~6 cm. A Multi-Channel Buffer (MCB) used is a PC-based plug in PCI card consisting of more than 8k Analog Digital Convertor (ADC) with sophisticated WINDOWS based control and analysis software MCDWIN to analyze the γ-ray spectrum. Under the assumption that secular equilibrium has been reached, the areas of ²¹⁴Pb and ²¹⁴Bi decay products and that of ¹³⁷Cs are measured. The ratio C1/C2 for each sample is computed. For confirmation the credibility of this methodology, a set of six zoomed samples spectrum is represented in Fig 5.

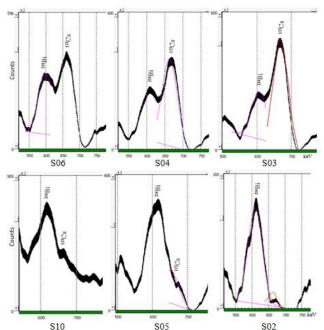


Fig 5. Spectrum of ²¹⁴Bi and ¹³⁷Cs of six energy zoomed samples

The elemental activity of the measured counts of photopeak (area counts at FWHM), for 30000 and 40000 sec spectrometric live time of soil and sediment samples respectively, are calculated by Eq (11). The resultant values of area counts and activities before correction are shown in columns 3-5 of Table 3. The measured peak areas for both radioisotopes ²¹⁴Bi and ¹³⁷Cs are used to calculate the ratios of C1 and C2 then their net areas and activities are computed by Eq(10) and Eq(11) respectively. The ratios of the measured areas of the two isotopes, the corrected areas as well as their activities are tabulated in columns 6-8 of Table3.

Table 3. The measured and corrected counts (Area) and activity A(Bq/Kg) of soil samples

	Isotopes	²¹⁴ Pb(²³⁸ U)			²¹⁴ Bi / ¹³⁷ Cs		¹³⁷ Cs
Sample	measurement	measured	measured	measured	¹³⁷ Cs / ²¹⁴ Bi	corrected	corrected
CO1	Area(count)	22900 ± 340	23900 ± 820	25500 ± 620	asured 137 Cs $^{/214}$ Bi corrected 0 ± 620 0.937 19820 ± 6 0 ± 0.40 1.067 21.63 ± 0 0 ± 740 2.232 11600 ± 7 ± 0.48 0.448 12.66 ± 0 0 ± 320 0.505 9704 ± 56 7 ± 0.21 1.979 10.59 ± 0 0 ± 330 0.591 12036 ± 4 4 ± 0.21 1.691 13.14 ± 0 $>3.226^b$ 13970 ± 18 $\le 0.310^b$ 15.25 ± 0 0 ± 390 0.890 11890 ± 5 4 ± 0.25 1.124 12.98 ± 0 >3.226 8078 ± 15 ≤ 0.310 8.82 ± 0.1 0 ± 420 0.663 13580 ± 7 5 ± 0.27 1.508 14.82 ± 0 0 ± 850 0.875 19530 ± 1 5 ± 0.55 1.142 21.32 ± 1 >3.226 14160 ± 2	19820 ± 680	21915 ± 533
301	A(Bq/Kg)	20.62±0.31	U) 214Bi(238U) measured measured measured set 23900 ± 820 25500 ± 820 16.49 ± 820 12500 ± 770 5600 ± 820 12500 ± 770 5600 ± 820 12500 ± 830 28100 ± 820 12500 ± 830 28100 ± 820 12500 ± 830 28100 ± 820 16500 ± 830 28100 ± 820 16500 ± 680 27900 ± 8.17 ± 820 16500 ± 18.01 ± 0.74 18.04 ± 820 14700 ± 190 ND a 16300 ± 820 14500 ± 700 16300 ± 8500 ± 160 ND a 16300 ± 160 ND a 17900 ± 990 17900 ± 990 17900 ± 990 17400 ± 10.54 ± 10.54 ± 10.55 17.46 ± 10.55 17.46 ± 10.55 17.46 ± 10.55 17.46 ± 10.55 17.46 ± 10.50 17.50 ± 10.5	16.49 ± 0.40	1.067	21.63 ± 0.74	14.17 ± 0.34
503	Area(count)	13300 ± 280	12500 ± 770	5600 ± 740	2.232	11600 ± 715	3725 ± 492
302	A(Bq/Kg)	11.98 ± 0.25	measured measured 137_{CS} / 21 23900 ± 820 25500 ± 620 0.937 26.08 ± 0.89 16.49 ± 0.40 1.067 12500 ± 770 5600 ± 740 2.232 13.64 ± 0.84 3.62 ± 0.48 0.448 14200 ± 830 28100 ± 320 0.505 15.50 ± 0.91 18.17 ± 0.21 1.979 16500 ± 680 27900 ± 330 0.591 18.01 ± 0.74 18.04 ± 0.21 1.691 14700 ± 190 NDa >3.226b 16.04 ± 0.21 ND ≤0.310b 14500 ± 700 16300 ± 390 0.890 15.83 ± 0,76 10.54 ± 0.25 1.124 8500 ± 160 ND >3.226 9.28 ± 0.17 ND ≤0.310 17900 ± 990 27000 ± 420 0.663 19.54 ± 1.08 17.46 ± 0.27 1.508 23900 ± 1690 27300 ± 850 0.875 26.08 ± 1.84 17.65 ± 0.55 1.142 14900 ± 310 ND >3.226 6.55 ± 0.25 ND	0.448	12.66 ± 0.78	2.41 ± 0.32	
502	Area(count)	12300 ± 270	14200 ± 830	28100 ± 320	ured 137_{CS} / 214 Bi corrected ± 620 0.937 19820 ± 680 ± 0.40 1.067 21.63 ± 0.74 ± 740 2.232 11600 ± 715 0.48 0.448 12.66 ± 0.78 ± 320 0.505 9704 ± 567 ± 0.21 1.979 10.59 ± 0.62 ± 330 0.591 12036 ± 496 ± 0.21 1.691 13.14 ± 0.54 >3.226b 13970± 180 ≤0.310b 15.25 ± 0.20 ± 390 0.890 11890 ± 574 ± 0.25 1.124 12.98 ± 0.63 >3.226 8078 ± 152 ≤0.310 8.82 ± 0.17 ± 420 0.663 13580 ± 751 ± 420 0.663 13580 ± 751 ± 850 0.875 19530 ± 138 ± 0.55 1.142 21.32 ± 1.51 >3.226 14160 ± 254 ≤0.310 13.64 ± 0.28 <0.310	9704 ± 567	25970 ± 296
Sample S01 S02 S03 S04 S05 S06 S07 S08 S09 S10 S11	A(Bq/Kg)	11.08 ± 0.24	15.50 ± 0.91	18.17 ± 0.21	1.979	10.59 ± 0.62	16.79 ± 0.19
604	Area(count)	15600 ± 230	16500 ± 680	27900 ± 330	0.591	12036 ± 496	25425 ± 301
304	A(Bq/Kg)	14.05 ± 0.21	18.01 ± 0.74	18.04 ± 0.21	1.691	13.14 ± 0.54	16.44 ± 0.19
005	Area(count)	16600 ± 230	14700 ± 190	ND^a	$>3.226^{b}$	13970± 180	0.0
202	A(Bq/Kg)	14.95 ± 0.21	16.04 ± 0.21	ND	≤0.310 ^b	15.25 ± 0.20	0.0
006	Area(count)	15000 ± 250	14500 ± 700	16300 ± 390	0.890	11890 ± 574	14125 ± 338
200	A(Bq/Kg)	13.51 ± 0.23	15.83 ± 0.76	10.54 ± 0.25	1.124	12.98 ± 0.63	9.13 ± 0.22
507	Area(count)	9900 ± 180	8500 ± 160	ND	>3.226	8078 ± 152	0.0
307	A(Bq/Kg)	8.91 ± 0.16	9.28 ± 0.17	ND	≤0.310	8.82 ± 0.17	0.0
000	Area(count)	16700 ± 290	17900 ± 990	27000 ± 420	0.663	13580 ± 751	24315 ± 378
308	A(Bq/Kg)	15.04 ± 0.26	19.54 ± 1.08	17.46 ± 0.27	1.508	14.82 ± 0.82	15.72 ± 0.24
000	Area(count)	23300 ± 310	23900 ± 1690	27300 ± 850	$\begin{array}{lll} 0.591 & 12036 \pm 496 \\ 1.691 & 13.14 \pm 0.54 \\ > 3.226^b & 13970 \pm 180 \\ \le 0.310^b & 15.25 \pm 0.20 \\ 0.890 & 11890 \pm 574 \\ 1.124 & 12.98 \pm 0.63 \\ > 3.226 & 8078 \pm 152 \\ \le 0.310 & 8.82 \pm 0.17 \\ 0.663 & 13580 \pm 751 \\ 1.508 & 14.82 \pm 0.82 \\ 0.875 & 19530 \pm 1381 \\ 1.142 & 21.32 \pm 1.51 \\ > 3.226 & 14160 \pm 254 \\ \le 0.310 & 13.64 \pm 0.28 \\ > 3.226 & 6460 \pm 247 \\ \le 0.310 & 6.22 \pm 0.22 \\ 1.405 & 4608 \pm 106 \\ \end{array}$	23715 ± 738	
309	A(Bq/Kg)	20.98 ± 0.28	26.08 ± 1.84	17.65 ± 0.55	1.142	21.32 ± 1.51	15.34 ± 0.48
C10	Area(count)	16800 ± 350	14900 ± 310	ND	>3.226	14160 ± 254	0.0
510	A(Bq/Kg)	13.35 ± 0.28	14.35 ± 0.30	ND	≤0.310	13.64 ± 0.28	0.0
C11	Area(count)	7600 ± 250	6800 ± 260	ND	>3.226	6460 ± 247	0.0
511	A(Bq/Kg)	6.04 ± 0.20	6.55 ± 0.25	ND	≤0.310	6.22 ± 0.22	0.0
010	Area(count)	5900 ± 140	5200 ± 120	3700 ± 190	1.405	4608 ± 106	2920 ± 150
S12	A(Bq/Kg)	4.69 ± 0.11	5.01 ± 0.12	2.11 ± 0.11	0.712	4.44 ± 0.10	1.67 ± 0.09
S13	Area(count)	14100 ± 250	13200 ± 170	5400 ± 520	2.444	12336 ± 159	3420 ± 329
313	A(Bq/Kg)	11.20 ± 0.20	12.71 ± 0.16	3.08 ± 0.30	0.409	11.88 ± 0.15	1.95 ± 0.19

a-Non Detactabe **b-** C1/C2>>3.226 and C2/C1=0.31 are derived to make 1-NS1=0.95 and 1-NS2=0.05, that is to say N(¹³⁷Cs)≈0.

The calculated activities (A) of the measured and corrected areas of ²¹⁴Bi and that of measured ²¹⁴Pb and the differences between the activities of both cases from the measured of ²¹⁴Pb, as well as the average values are summarized in Table 4. It is clear from Table 4 that this methodology of correction

approved the results of 214 Bi activity from the maximum, minimum and average relative errors decreased from 40%, 7.3% and 19.4% to 6.1%, -6.5% and 0.6% respectively of that of 214 Pb.

Table 4. Comparison between the measured and corrected activities of ²¹⁴Bi and ²¹⁴Pb

Comple	A(²¹⁴ Pb) Bq/kg	A(²¹⁴ Bi) Bq/kg	A(²¹⁴ Bi) Bq/kg	A(Bi)-A(Pb) Bq/kg	A(Bi)-A(Pb) Bq/kg
Sample	measured	measured	corrected	Measured	Corrected
S01	20.62	26.08	21.63	5.46	1.01
S02	11.98	13.64	12.66	1.66	0.68
S03	11.08	15.5	10.59	4.42	-0.49
S04	14.05	18.01	13.14	3.96	-0.91
S05	14.95	16.04	15.25	1.09	0.3
S06	13.51	15.83	12.98	2.32	-0.53
S07	8.91	9.28	8.82	0.37	-0.09
S08	15.04	19.54	14.82	4.5	-0.22
S09	20.98	26.08	21.32	5.1	0.34
S10	13.35	14.35	13.64	1	0.29
S11	6.04	6.55	6.22	0.51	0.18
S12	4.69	5.01	4.44	0.32	-0.25
S13	11.2	12.71	11.88	1.51	0.68
Average	12.800	15.278	12.876	2.478	0.076

The smoothness of the results of net activity obtained after correction is obvious from the relationship; A(Bi)_{Cor.}=

 $1.036A(Pb)_{Meas.}$ - 0.3852 with $R^2 = 0.9888$ as shown in Fig. 6.

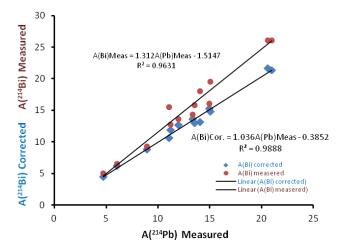


Fig 6. The relationship between the measured and corrected activities of ²¹⁴Bi and ²¹⁴Pb

CONCLUSION

The developed method is verified and approved by comparing the corrected areas optained for ²¹⁴Bi with that measured ²¹⁴Pb. As the two radioisotopes ²¹⁴Pb and ²¹⁴Bi are in secular equilibrium of the series decaying primordial radionuclide ²³⁸U, both have equal activity, A(²¹⁴Pb) = A(²¹⁴Bi). The range of activity concentration < 2.0 Bq.kg⁻¹ in samples shows no noticeable peak, then considered as below the minimum detectable count and ignored. It is noticed that the minimum detected activity measured is 2.11 Bg.kg⁻¹ (sediment S12) results corrected 1.69 Bg.kg⁻¹. The activity concentrations of ¹³⁷Cs varied from one site to another. The variation of the concentration along the landscape has been attributed to erosion and deposition in combination with surface run-of the disturbed and undisturbed soil. Also the topography affects the distribution of ¹³⁷Cs. The concentration of ¹³⁷Cs is highest at the foot of the slope and

lowers near the summit and near the creaks and lakeshore. According to the maximum areas ratio of C(Bi)/C(Cs) founds practically C1/C2 = 2.444 (sediment S13), the maximum overlapped area of 137 Cs in 214 Bi peak area is NS1 = 0.39. Accordingly the net area of Cs considered is 69%. Consequently values of C1/C2 = 3.226 and C2/C1 = 0.31 are derived for samples of non detactabe ¹³⁷Cs so that the net area (1-NS1=0.95) and (1-NS2=0.05) of ²¹⁴Bi and ¹³⁷Cs respectively are the limits of correction. The correlations derived are well compatible with the subject of confidence limit. A realistic estimate of the uncertainty of the measurement is related to the net area of the normal distribution. This method can by generalized for any other doublet peaks separated by, not only one FWHM, but much more or less. Also it can be adoubted for any other cize of NaI detectors of different resolution. In addition it can easely be programed in any spectrum analysis software.

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