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Measurement of naturally occurring radioactive material, ^{238}U and ^{232}Th: part 2—optimization of counting time

Nabanita Naskar[1,2] · Susanta Lahiri[2] · Punarbasu Chaudhuri[1] · Alok Srivastava[3]

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Abstract An effort has been made to optimize the counting time for low-level measurement of naturally occurring radioactive material (NORM) by considering the standard deviation between the activity values of different photopeaks and counting error. It is observed that at lower counting time, relative standard deviation (RSD) varies randomly, but attains a gradual trend with increasing time and also comes closure to the counting error. Therefore minimum counting time for low-level NORM measurement of ^{238}U and ^{232}Th would be the time required to stabilize the RSD values.

Keywords Naturally occurring radioactive materials (NORMs) · ^{238}U and ^{232}Th measurement · Gamma spectrometry · Minimum counting time

Introduction

The measurement techniques of physical quantities are highly dependent on their magnitude. Interestingly gamma spectrometry is reasonably sensitive over a wide range of magnitude, i.e., from fraction of Bq to tens of MBq, just by adjusting the source to detector distance and time of counting. In the measurement of naturally occurring radioactive materials (NORM), the count rate may come down to ~1–2 Bq in a single experimental sample. Measurement of this minuscule amount of radioactivity thus demands careful consideration of the minute details of the counting protocols as small error in the measurement would be multiplied manifolds while expressing the end result in normalized form like Bq kg^{-1}. The above statement is further supported in a paper by Durec et al. [1]. An inter comparison exercise on quantitative assessment of various natural radionuclides in river sediments were carried out by 25 laboratories of Europe. It was found that although gamma spectrometry was the most implemented technique rather than beta counting or alpha spectrometry, but all the results from gamma spectrometry were not acceptable. The activity of ^{226}Ra reported by different laboratories on same sample varied between 21 and 106 Bq kg^{-1}. This paper clearly demonstrates that for better understanding of low-level NORM measurement various parameters like proper photopeaks selection, counting time, sample geometry, detector efficiency and resolution, appropriate use of standards, etc., need to be rationalized. In a recent publication we have worked extensively on one of these parameters, selection of proper photopeaks, for measurement of ^{238}U and ^{232}Th. We could show that among more than 200 photopeaks in ^{238}U, ^{232}Th series, only few photopeaks from the daughter products of the corresponding series are reliable to get a good estimate of the level of uranium and thorium in NORM [2]. However, to the best of our knowledge till date no such attempt has been made to investigate minimum acceptable counting time for measurement of ultra-low level activity in natural samples based on the standard deviations between the activity values under different photopeaks.

In this connection, it is noteworthy to mention recent debates on the issue of selection of appropriate counting

✉ Susanta Lahiri
susanta.lahiri@saha.ac.in

1 Department of Environmental Science, University of Calcutta, 35 Ballygunge Circular Road, Kolkata 700019, India

2 Saha Institute of Nuclear Physics, 1/AF, Bidhannagar, Kolkata 700064, India

3 Chemistry Department, Panjab University, Chandigarh 160014, India

time required for measurement of NORM (^{238}U, ^{232}Th and ^{40}K). A research group assessed the activity of ^{238}U, ^{232}Th and ^{40}K radionuclides for 36000 s in sediment, soil and water samples collected from the lower basin of river Pra in Ghana [3]. In a letter to the editor a researcher objected that 36000 s was too small duration and the data should have been taken for at least 24 h [4]. The original authors in their response admitted that 24 h counting time would be better but 10 h was also not bad [5]. In all these three references, arguments on the choice of counting time were based on their experience and personal belief. Therefore it is clear that though counting time is one of the important parameters which influences the end result, never a thorough critical assessment was done on the minimum counting time required for NORM measurement.

The counting error decreases with increasing counting time. However, too long counting time also restricts the number of samples that can be analyzed by a detector. Therefore depending upon the practicality, different researchers have selected different counting time for low-level NORM measurement. A glance at literature shows that there is lot of arbitrariness in the selection of counting time irrespective of detector efficiency and type of detector [3, 6–60]. Table 1 lists the counting time taken by various researchers in different experimental set-up.

In NORM measurement it is better to take average of the activity values under few photopeaks of daughter products belonging to decay series, ^{238}U and ^{232}Th rather than to conclude the amount of U/Th/Ra in a given sample from the activity value under a single photopeak. In principle, the activity obtained under different photopeaks for same or different isotopes of a particular series should be equal (after attaining secular equilibrium). However in practice this is never observed because of several factors like intrinsic property of the detector, noise due to electronics, background, Compton edge of the other photopeaks, etc.

It is customary to express the activity in the form of $x \pm \Delta x$, where x represents activity of the sample and Δx represents the counting error (CE), which is equal to $\sqrt{x}$ according to Poisson distribution. If the average activity obtained from different photopeaks is taken as x, then Δx is represented by $\sqrt{\frac{\Delta x_1^2+\Delta x_2^2+\ldots+\Delta x_N^2}{N-1}}$, where $\Delta x_1, \ldots \Delta x_N$ denotes counting error for $x_1 \ldots x_N$. It has been shown by Naskar et al. [2] that apart from the counting error, standard deviation (SD) between the activity values obtained under different photopeaks is also a measure of error and sometime more significant than the counting error. Therefore merely representing the average value and its error in terms of counting error is not proper. Estimation of both CE and SD should be done, and the higher one should be reported as error.

The present study makes an earnest attempt to investigate the nature of SD and CE with respect to different counting time to get more meaningful result from low-level radioactivity measurements.

Experimental

High-purity Germanium (HPGe) detector with 50% relative efficiency and 3.1 keV resolution at 1332 keV was used to measure ^{238}U and ^{232}Th activities in four soil samples collected from West Bengal and Punjab state of India. The detector was well shielded with 10 cm thick lead shield. The entire detector and shielding arrangement has been described elsewhere [6]. Samples used for measurement were air-dried, grinded, weighed (50 g each) and sealed in airtight petri-plates for more than one month to maintain secular equilibrium. Energy calibration was done using point sources of ^{152}Eu, ^{137}Cs, ^{133}Ba and ^{60}Co. In addition to unknown soil samples, two ^{238}U samples of known strength, 2 Bq and 5 Bq were prepared from weighed amount of IAEA uranium ore (Pitchblende) standard (0.14 and 0.35 g correspond to 2 and 5 Bq respectively). Also two ^{232}Th samples, 2 and 5 Bq were prepared using weighed amount of thorium acetate, [$Th(CH_3COO)_4$] (0.995 and 2.49 mg correspond to 2 and 5 Bq respectively). For validation of result, in both cases of U and Th measurement, 2 Bq samples were used respectively as U and Th standards, whereas the samples having strength 5 Bq were used as samples of known activity. The samples were counted for 5000, 10000, 20000, 30000, 50000, 75000, 100000, 125000 and 150000 s. The background activity was also measured at all the above-mentioned counting times. The respective background activity was stripped from corresponding spectrum prior to analysis. The activities of ^{238}U and ^{232}Th in different samples have been taken as the average of activities obtained under different photopeaks mentioned in Table 2 as it has been observed by our group that the said peaks provide the most reliable data [2]. It is noteworthy to mention here that these photopeaks essentially represent ^{226}Ra in ^{238}U series as the equilibrium is established between ^{226}Ra and its daughter products after the samples are hermetically sealed for 30 days or more. It is further assumed that the equilibrium between ^{238}U and ^{226}Ra also exist in natural samples. However, in some cases due to geochemical cycles, this equilibrium may not exist. In the experimental samples we have observed that ^{238}U and ^{226}Ra are in equilibrium. This has been confirmed by measuring activities from 63.29 keV (4.8%) and 92.38 (2.81%), 92.80 keV (2.77%) photopeaks of ^{234}Th, which were found similar to the activities calculated from the photopeaks shown in Table 2.

Table 1 Counting time taken by different research groups for NORM measurement (HPGe detectors have been used in all the following cases)

Sl. No	Reported by [Ref]	Relative efficiency of the detector (%)	Counting time (s)	Sample weight (g)
1	Boukhenfouf W and Boucenna A [7]	10	108,000	500
2	Jaison et al. [8]	15	60,000	300
3	Kurnaz et al. [9]	16	50,000	–
4	Mahur et al. [10]	20	72,000	250
5	Tchokossa et al. [11]	20	36,000	–
6	Gupta et al. [12]	20	72,000	300–400
7	Alatise et al. [13]	20	36,000	200
8	Al-Jundi et al. [14]	20	57,600	–
9	Maxwell et al. [15]	20	21,600	~500
10	Yasmin et al. [16]	20	10,000	131 cm^3
11	Saleh H and Abu Shayeb M [17]	20	54,000	100
12	Ribeiro et al. [18]	20	60,000	300
13	Kılıç Ö and Çotuk Y [19]	22.1	259,200	–
14	Janković et al. [20]	23	70,000	–
15	Chowdhury et al. [21]	23 and 35	50,000	–
16	Adukpo et al. [3]	25	36,000	–
17	Bakim M and Uğur Görgün A [22]	25	36,000–86,400	–
18	LaBrecque et al. [23]	25	86,400	650–700
19	Alaamer AS [24]	25	54,000	200
20	Powell et al. [25]	25	18,000	225
21	Yii et al. [26]	25	86,400	–
22	Miller M and Voutchkov M [27]	28	86,400	–
23	El Samad et al. [28]	30 and 40	129,600–172,800	–
24	Stajic et al. [29]	30	21600	–
25	Aytekin et al. [30]	30	32,819–62,751	1000
26	Kobya et al. [31]	30	50,000	2000
27	Song et al. [32]	30	21,600–36,000	–
28	Agbalagba et al. [33]	30	20,000	–
29	Rahman et al. [34]	30	65,000	–
30	Ele Abiama et al. [35]	30	54,000	–
31	Ahmed NK and Mohamed El-Arabi AG [36]	30	36,000	60
32	Yang et al. [37]	30	21,600	200
33	Alabdullah et al. [38]	30 and 100	86,400	–
34	Kannan et al. [39]	33	50,000	–
35	Dragović et al. [40]	34	60,000	–
36	Chakraborty et al. [41]	35	20,000– 80,000	–
37	Latif et al. [42]	40	86,400	–
38	Murty VRK and Karunakara N [43]	41	60,000	–
39	Srilatha et al. [44]	41	60,000	–
40	Pinto P and Yerol N [45]	42	30,000	–
41	Usikalu et al. [46]	45	28,800	100
42	Mohapatra et al. [47]	50	100,000	300
43	Srivastava et al. [48]	50	80,000–170,000	50
44	Sartandel et al. [49]	50	100,000	–
45	Rajeshwari et al. [50]	50	50,000	–
46	Dusane et al. [51]	50	100,000	–
47	Canbazoğlu et al. [52]	50	72,000–86,400	1000
48	Aközcan S [53]	50	160,000	–

Table 1 continued

Sl. No	Reported by [Ref]	Relative efficiency of the detector (%)	Counting time (s)	Sample weight (g)
49	Al-Sharkawy et al. [54]	50	20,000–40,000	500
50	Chaudhuri et al. [6]	50	75,000	50
51	Wasim et al. [55]	60	57,600	200
52	Hannan et al. [56]	70	86,400	–
53	Jallad KN [57]	150	1800	350
54	Santawamaitre et al. [58]	0.50% (absolute efficiency at 662 keV)	172,800	–
55	Özmen et al. [59]	Not mentioned	500,00	–
56	Tufail et al. [60]	55.7 mm × 72.1 mm (crystal size)	72,000	–

Table 2 Sets of photopeaks considered for the present study [2]

Parent radionuclide	Daughter radionuclide	Photopeak (keV) [61]	Intensity (%) [61]
^{238}U	^{214}Pb	295.22	19.3
	^{214}Pb	351.93	37.6
	^{214}Bi	609.31	46.1
^{232}Th	^{228}Ac	338.32	11.3
	^{208}Tl	583.19	84.5
	^{228}Ac	911.20	25.8

Table 3 Observed activities and errors of sample SB1 at different counting times

Counting time, s	Observed activity of ^{238}U, Bq	RCE %	RSD %	Observed activity of ^{232}Th, Bq	RCE %	RSD %
5000	1.90	22.80	3.98	1.54	44.12	63.66
10,000	2.00	17.37	16.40	1.81	16.60	36.03
20,000	1.52	10.20	12.17	1.55	12.06	19.99
30,000	1.83	9.52	24.73	1.82	9.68	4.28
50,000	1.62	6.68	17.50	1.57	7.37	25.43
75,000	1.45	5.75	1.94	1.86	5.17	3.76
100,000	1.66	4.63	15.82	1.67	5.03	10.80
125,000	1.78	4.18	11.02	1.74	4.81	10.59
150,000	1.49	3.92	10.80	1.68	3.77	8.82

Table 4 Observed activities and errors of sample SB2 at different counting times

Counting time, s	Observed activity of ^{238}U, Bq	RCE %	RSD %	Observed activity of ^{232}Th, Bq	RCE %	RSD %
5000	1.50	25.74	34.34	1.65	37.75	35.56
10,000	1.52	18.99	14.28	1.56	17.83	44.40
20,000	1.24	11.09	19.22	1.35	11.67	1.50
30,000	1.30	10.88	28.64	1.80	9.61	18.56
50,000	1.44	6.88	4.44	1.38	7.70	13.50
75,000	1.18	6.30	3.46	1.48	5.52	10.42
100,000	1.38	4.93	9.36	1.48	5.14	9.07
125,000	1.36	4.24	8.22	1.34	4.87	8.02
150,000	1.20	3.77	5.15	1.45	3.94	9.16

Results and discussion

Tables 3, 4, 5 and 6 represents observed activity of U and Th for samples SB1, SB2, PU1, PU2 at different counting times along with their relative counting error $\left(\mathrm{RCE} = \frac{\textit{Counting error}}{\textit{mean value}} \times 100\right)$ and Relative Standard Deviation $\mathrm{RSD} = \frac{\textit{Standard deviation}}{\textit{mean value}} \times 100$. Table 7 shows the observed activity of sample of known strength of U and Th (5 Bq) along with their RCE and RSD. The two errors, namely, RCE and RSD related to measured activities of ^{238}U and ^{232}Th of the four samples have been plotted in Fig. 1a–d. Similarly the errors related to U and Th activities of the samples with known strengths have been plotted in Fig. 1e.

Table 5 Observed activities and errors of sample PU1 at different counting times

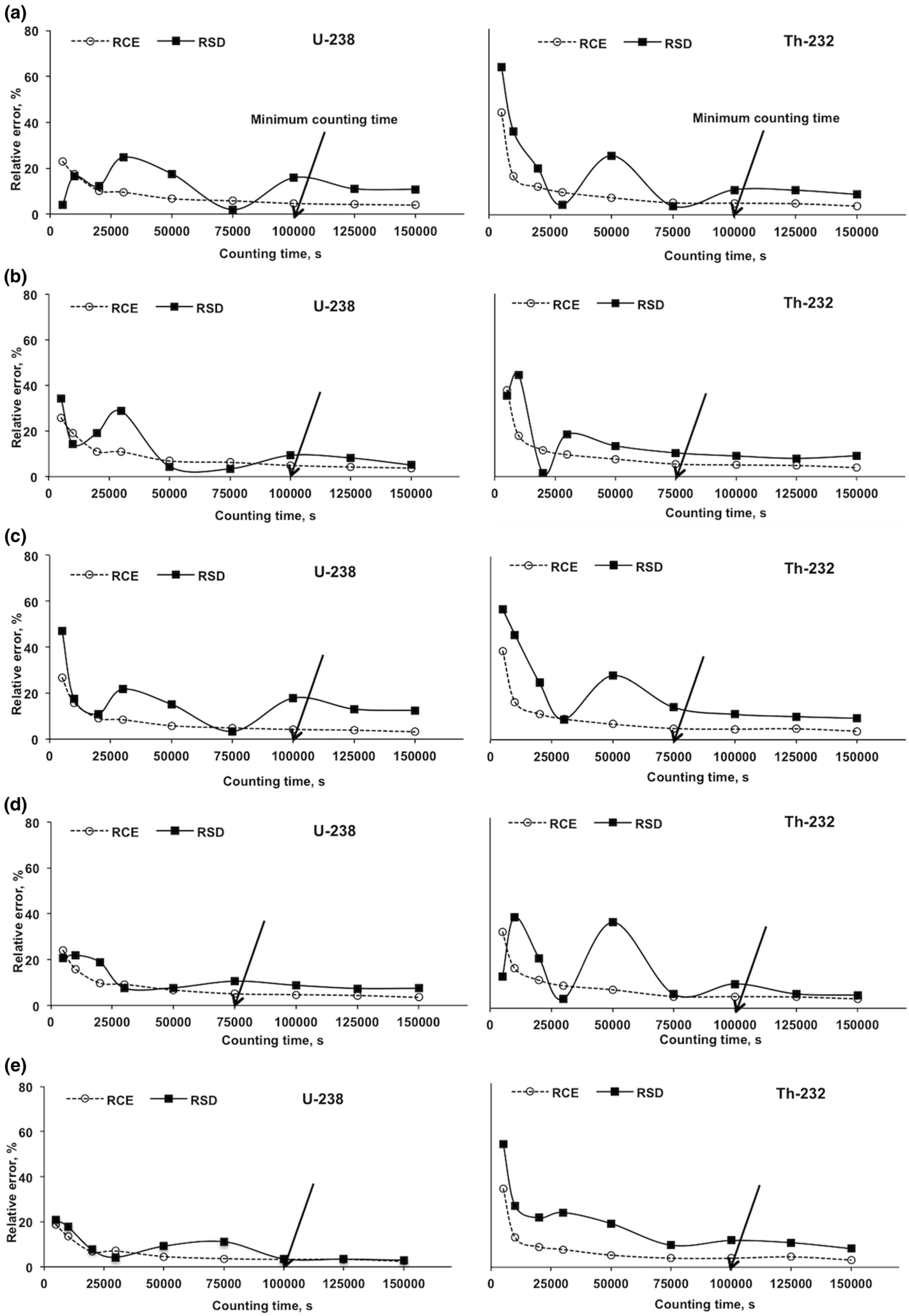
Counting time, s	Observed activity of ^{238}U, Bq	RCE %	RSD %	Observed activity of ^{232}Th, Bq	RCE %	RSD %
5000	2.11	26.72	49.92	2.44	38.51	56.78
10,000	2.62	15.90	17.51	2.12	16.42	45.78
20,000	2.23	9.10	11.03	2.05	11.24	24.80
30,000	2.71	8.49	21.77	2.36	9.04	8.86
50,000	2.54	5.86	15.21	2.00	6.77	27.90
75,000	2.16	4.91	3.41	2.22	4.87	14.09
100,000	2.33	4.25	17.92	2.01	4.63	11.04
125,000	2.42	3.93	13.13	1.97	4.74	10.06
150,000	2.21	3.21	12.52	2.02	3.71	9.35

Table 6 Observed activities and errors of sample PU2 at different counting times

Counting time, s	Observed activity of ^{238}U, Bq	RCE %	RSD %	Observed activity of ^{232}Th, Bq	RCE %	RSD %
5000	2.31	24.08	20.70	2.29	31.98	13.20
10,000	2.21	15.91	21.94	1.59	16.77	38.29
20,000	1.74	9.71	18.84	1.75	11.73	21.02
30,000	2.36	9.17	7.56	1.99	9.42	3.86
50,000	1.88	6.70	7.69	1.63	7.73	36.05
75,000	1.89	5.02	10.56	2.10	4.89	5.88
100,000	1.82	4.67	8.76	1.74	4.85	10.20
125,000	1.92	4.29	7.40	1.85	4.69	6.01
150,000	1.88	3.46	7.44	1.82	3.91	5.47

Table 7 Observed activity of 5 Bq known activity of ^{238}U and 5 Bq known activity of ^{232}Th along with their respective errors

Counting time, s	Observed activity of ^{238}U, Bq	RCE %	RSD %	Observed activity of ^{232}Th, Bq	RCE %	RSD %
5000	7.26	18.84	20.84	8.83	34.12	53.64
10,000	6.56	13.47	17.80	7.15	12.96	26.81
20,000	5.27	6.75	7.64	6.67	8.83	21.45
30,000	6.66	7.02	4.26	7.58	7.69	23.77
50,000	6.43	4.57	9.19	6.90	5.24	18.99
75,000	5.20	3.65	11.16	6.69	3.92	9.55
100,000	6.16	3.31	3.56	6.83	3.90	11.61
125,000	6.05	3.35	3.44	6.50	4.50	10.66
150,000	5.46	2.46	2.96	6.00	3.14	8.09


(a)
RCE
RSD
U-238
Th-232
Minimum counting time
Relative error, %
Counting time, s
(b)
(c)
(d)
(e)

◀**Fig. 1** **a** Variation of RCE and RSD with counting time for sample SB1. **b** Variation of RCE and RSD with counting time for sample SB2. **c** Variation of RCE and RSD with counting time for sample PU1. **d** Variation of RCE and RSD with counting time for sample PU2. **e** Variation of RCE and RSD with counting time for standards of 5 Bq known activity

In all the Fig. 1a–e, RCE decreases with increasing counting time, which is as expected. However, this is not the case for RSD, which varies rather randomly in case of lower counting time i.e. sometime RSD is very high and sometime it is too low in magnitude. The RSD is found to follow a systematic trend only beyond certain counting time. In lower counting time, relatively low RSD is explained by the fact that by chance the activity values under different peaks come closer although they may be far from the true value. This argument is further strengthened and empirically demonstrated from Fig. 2 wherein activity of ^{238}U and ^{232}Th under individual photopeaks has been plotted for known sample strength of 5 Bq. For example, in 5000 s counting time, RSD for 5 Bq ^{238}U standard is as high as 20.8%, and the individual activity values under the photopeaks of 295.2, 351.9 and 609.3 keV are 6.24, 6.53, 9 Bq respectively. The RSD decreased to only 4.2% at 30000 s with the individual activity values under the above peaks 6.58, 6.97, 6.42 Bq respectively. In both cases the average values are far from true value (5 Bq). A similar trend can be seen in the case of ^{232}Th series. RSD value for 5000 s was as high as 53.6%, which is accounted for large difference in the activity values of different daughter products of ^{232}Th.

It can be safely stated that as the counting time is increased, the data becomes more reliable. For example, in 150000 s both RCE and RSD values are smaller. However, one cannot go for very high counting time due to several restrictions. It would be a good idea for experimenters involved in low-level radioactivity measurements to empirically select a counting time for at least 1–2 samples which satisfies the conditions that RSD values come closer to RCE values and also RSD values do not fluctuate a lot during different counting time intervals, i.e. a regular trend in it is observed. In the present case, 75,000–100,000 s could be a reasonable minimum counting time, which have been indicated in the Fig. 1a–e with an arrow. However, to be in safe side one can choose 100,000 s.

All the above discussions have been made by taking average of three photopeaks from ^{238}U and ^{232}Th series as depicted in Table 2. Further we have checked the nature of RSD values with variation of counting time and by introducing more number of photopeaks. For ^{238}U we have consecutively added 1764.49 keV (15.4%) and 2204.21 keV (5.08%) photopeaks, both from ^{214}Bi. Similarly for ^{232}Th, 727.33 keV (6.58%) photopeak from ^{212}Bi and 860.56 (12.42%) from ^{208}Tl were consecutively added. It has been observed that addition of photopeaks deteriorates the situation in terms of both randomness and magnitude of RSD. To illustrate with an example, RSD values of sample SB1 for 3,4 and 5 photopeak combinations at different counting times have been tabulated in Tables 8 and 9 for ^{238}U and ^{232}Th respectively. The addition of photopeaks increased RSD values as high as 91% and 118% for ^{238}U and ^{232}Th. The five photopeaks combinations for both ^{238}U and ^{232}Th show that even 100,000 s counting time may not be sufficient to stabilize the RSD values.

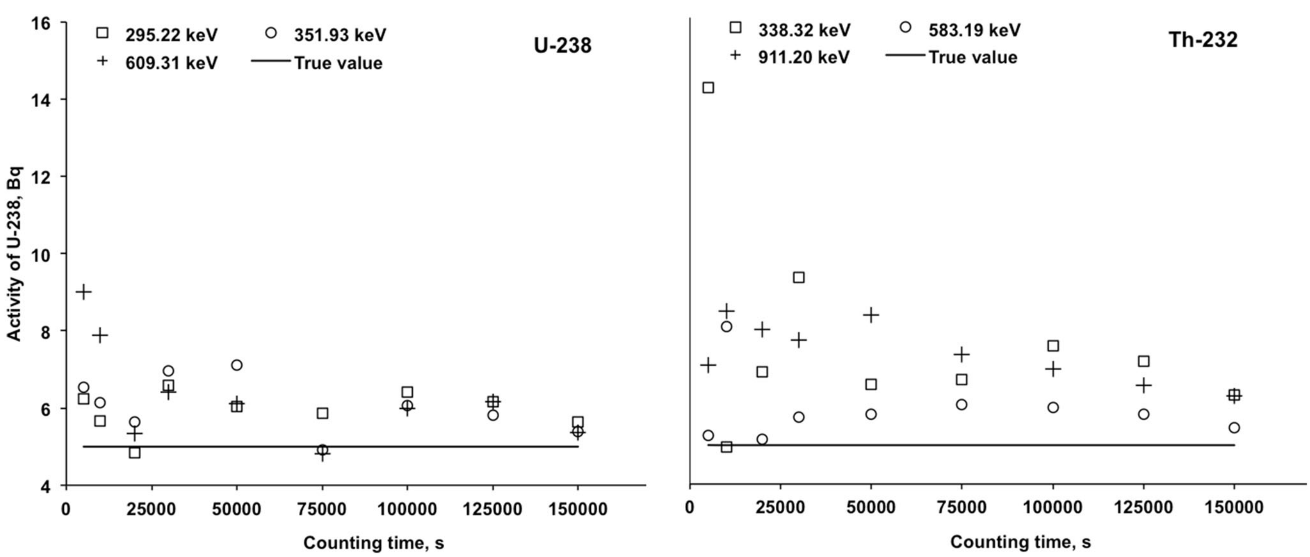


Fig. 2 Variation of activity under different photopeaks with true value for standards of 5 Bq known activity

Table 8 RSD (%) for different photopeaks combination of ^{238}U for sample SB1 at different counting times

Counting time, s	3 peaks	3 peaks + 1764.5 keV peak	3 peaks + 1764.5 keV + 2204.2 keV peaks
5000	3.99	66.81	91.41
10,000	16.40	31.72	65.61
20,000	12.17	41.04	53.61
30,000	24.73	20.55	18.33
50,000	17.50	14.28	58.00
75,000	1.94	18.25	15.97
100,000	15.82	23.93	31.28
125,000	11.02	17.15	32.62
150,000	10.80	9.46	41.36

Table 9 RSD (%) for different photopeaks combination of ^{232}Th for sample SB1 at different counting times

Counting time, s	3 peaks	3 peaks + 727.3 keV peak	3 peaks + 727.3 keV + 860.6 keV peaks
5000	63.66	96.16	118.15
10,000	36.03	30.42	64.88
20,000	19.99	31.29	65.37
30,000	4.28	27.10	47.74
50,000	25.43	21.44	33.26
75,000	3.76	3.46	16.96
100,000	10.80	11.04	14.53
125,000	10.59	18.24	15.78
150,000	8.82	20.42	22.18

Further, we have normalized the RSD values with respect to number of photopeaks, i.e., RSD values of three photopeaks combinations have been divided by 3, four photopeaks combinations have been divided by 4 and so on. The normalized values, i.e., RSD/photopeak have been plotted in Fig. 3. The figure strengthens our early data and shows that with increasing number of photopeaks RSD/photopeak also increases. Figure 3 also strongly supports the choice of three most reliable photopeaks each from ^{238}U and ^{232}Th series as described in Table 2 and our earlier paper [2].

Apart from proper selection of gamma lines, parameters like type and efficiency of detector, calibration standard and geometry, etc., may also influence the minimum counting time required for NORM measurement. Therefore it is recommended that the experimenter should consider RCE

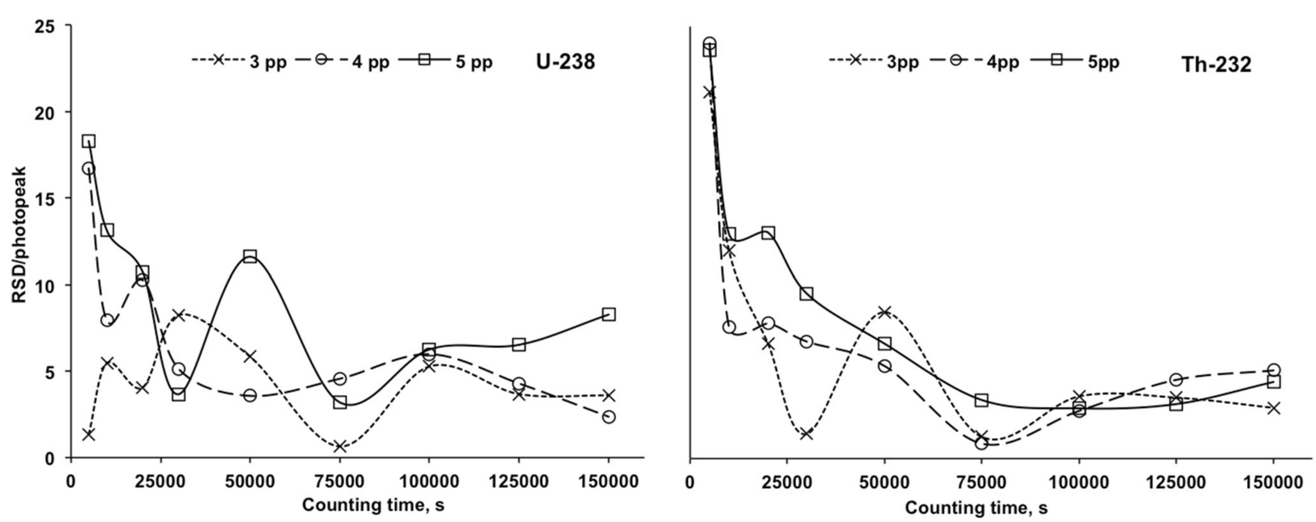


Fig. 3 Variation of RSD/photopeak with counting time and different photopeak combinations

and RSD values to select minimum reasonable counting time in their own system to get more reliable results.

Conclusion

In the study of NORM employing gamma ray spectrometric setup it is important to have prior idea of both standard deviation of activities under different photopeaks selected for NORM measurement and the counting error to determine minimum counting time required for set of similar samples to obtain meaningful result. It is generally observed that relative standard deviations (RSD) will be small in magnitude and will merge with relative counting errors (RCE) as counting time becomes longer. The recommendation of empirically determining a reasonable minimum counting time based of RSD and RCE values will also minimize the ambiguity and arbitrariness in selection of counting time in getting more consistent output by different researchers.

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References

1. Durec F, Betti M, Durecova A (2008) Intercomparison exercise on the determination of radionuclides in sediment from the Dudvah River. Appl Radiat Isot 66:1706–1710
2. Naskar N, Lahiri S, Chaudhuri P, Srivastava A (2016) Measurement of naturally occurring radioactive material, ^{238}U and ^{232}Th: anomalies in photo-peak selection. J Radioanal Nucl Chem 310:1381–1396
3. Adukpo OK, Faanu A, Lawluvi H, Tettey-Larbi L, Emi-Reynolds G, Darko EO, Kansaana C, Kpeglo DO, Awudu AR, Glover ET, Amoah PA, Efa AO, Agyemang LA, Agyeman BK, Kpordzro R, Doe AI (2015) Distribution and assessment of radionuclides in sediments, soil and water from the lower basin of river Pra in the Central and Western Regions of Ghana. J Radioanal Nucl Chem 303:1679–1685
4. Khandaker MU (2016) Letter to the editor: "Distribution and assessment of radionuclides in sediments, soil and water from the lower basin of river Pra in the central and western regions of Ghana". J Radioanal Nucl Chem 307:1–2
5. Adukpo OK, Faanu A, Lawluvi H, Tettey-Larbi L, Emi-Reynolds G, Darko EO, Kansaana C, Kpeglo DO, Awudu AR, Glover ET, Amoah PA, Efa AO, Agyemang LA, Agyeman BK, Kpordzro R, Doe AI (2016) Reply to the letter written by Mayeen Uddin Khandaker on: "Distribution and assessment of radionuclides in sediments, soil and water from the lower basin of river Pra in the central and western regions of Ghana. J Radioanal Nucl Chem 307:3
6. Chaudhuri P, Naskar N, Lahiri S (2017) Measurement of background radioactivity in surface soil of Indian Sundarban. J Radioanal Nucl Chem. doi:10.1007/s10967-016-5158-x
7. Boukhenfouf W, Boucenna A (2011) The radioactivity measurements in soils and fertilizers using gamma spectrometry technique. J Environ Radioact 102:336–339
8. Jaison TJ, Patra AK, Jha MK, Hedge AG (2010) Assessment of natural radioactivity in silt samples from Moticher lake near Kakrapar Atomic Power Station, India. J Radioanal Nucl Chem 284:583–589
9. Kurnaz A, Küçükömeroğlu B, Keser R, Okumusoglu NT, Korkmaz F, Karahan G, Çevik U (2007) Determination of radioactivity levels and hazards of soil and sediment samples in Fırtına Valley (Rize, Turkey). Appl Radiat Isot 65:1281–1289
10. Mahur AK, Gupta M, Varshney R, Sonkawade RG, Verma KD, Prasad R (2013) Radon exhalation and gamma radioactivity levels in soil and radiation hazard assessment in the surrounding area of National Thermal Power Corporation, Dadri (U.P.), India. Radiat Meas 50:130–135
11. Tchokossa P, Makon TB, Nemba RM (2012) Assessment of radioactivity contents and associated risks in some soil used for agriculture and building materials in Cameroon. J Environ Prot 3:1571–1578
12. Gupta M, Chauhan RP, Garg A, Kumar S, Sonkawade RG (2010) Estimation of radioactivity in some sand and soil samples. Indian J Pure Appl Phys 48:482–485
13. Alatise OO, Babalola IA, Olowofela JA (2008) Distribution of some natural gamma-emitting radionuclides in the soils of the coastal areas of Nigeria. J Environ Radioact 99:1746–1749
14. Al-Jundi J, Al-Bataina BA, Abu-Rukah Y, Shehadeh HM (2003) Natural radioactivity concentrations in soil samples along the Amman Aqaba Highway, Jordan. Radiat Meas 36:555–560
15. Maxwell O, Wagiran H, Ibrahim N, Lee SK, Sabri S (2013) Comparison of activity concentration of ^{238}U, ^{232}Th and ^{40}K in different layers of subsurface structures in Dei-Dei and Kubwa, Abuja, northcentral Nigeria. Radiat Phys Chem 91:70–80
16. Yasmin S, Barua BS, Kamal M, Rashid MdA (2014) An analysis for distribution of natural radionuclides in soil, sand and sediment of Potenga Sea beach area of Chittagong, Bangladesh. J Environ Prot 5:1553–1563
17. Saleh M, Abu Shayeb M (2014) Natural radioactivity distribution of Southern part of Jordan (Ma′an) soil. Ann Nucl Energ 65:184–189
18. Ribeiro FCA, da Lauria D, do Rio MA, da Cunha FG, Oliveira SW, Lima ED, Franzen M (2017) Mapping soil radioactivity in the Fernando de Noronha archipelago, Brazil. J Radioanal Nucl Chem 311:577–587
19. Kılıç Ö, Çotuk Y (2011) Radioactivity concentrations in sediment and mussel of Bosphorus and Golden Horn. J Radioanal Nucl Chem 289:627–635
20. Janković M, Todorović D, Savanović M (2008) Radioactivity measurements in soil samples collected in the Republic of Srpska. Radiat Meas 43:1448–1452
21. Chowdhury MI, Alam MN, Hazari SKS (1999) Distribution of radionuclides in the river sediments and coastal soils of Chittagong, Bangladesh and evaluation of the radiation hazard. Appl Radiat Isot 51:747–755
22. Bakım M, Uğur Görgün A (2015) Radioactivity in soils and some terrestrial foodstuffs from organic and conventional farming areas in Izmir, Turkey. J Radioanal Nucl Chem 306:237–242
23. LaBrecque JJ, Cordoves PR, Cordoves MA, Pérez K, Palacios D, Alfonso AJ (2010) Distribution of ^{137}Cs, ^{40}K, ^{232}Th and ^{238}U in coastal marine sediments of Margarita Island, Venezuela. J Radioanal Nucl Chem 283:669–674

24. Alaamer AS (2008) Assessment of human exposures to natural sources of radiation in soil of Riyadh, Saudi Arabia. Turk J Eng Environ Sci 32:229–234
25. Powell BA, Hughes LD, Soreefan AM, Falta D, Wall M, DeVol TA (2007) Elevated concentrations of primordial radionuclides in sediments from the Reedy River and surrounding creeks in Simpsonville, South Carolina. J Environ Radioact 94:121–128
26. Yii MW, Wan Mahmood Z, Ahmad Z, Jaffary NA, Ishak N (2011) NORM activity concentration in sediment cores from the Peninsular Malaysia East Coast Exclusive Economic Zone. J Radioanal Nucl Chem 289:653–661
27. Miller M, Voutchkov M (2014) Evaluation of gamma activities of naturally occurring radioactive materials in uncontaminated surface soils of Jamaica. J Radioanal Nucl Chem 300:303–313
28. El Samad O, Baydoun R, Nsouli B, Darwish T (2013) Determination of natural and artificial radioactivity in soil at North Lebanon province. J Environ Radioact 125:36–39
29. Stajic JM, Milenkovic B, Pucarevic M, Stojic N, Vasiljevic I, Nikezic D (2016) Exposure of school children to polycyclic aromatic hydrocarbons, heavy metals and radionuclides in the urban soil of Kragujevac city, Central Serbia. Chemosphere 146:68–74
30. Aytekin H, Çağatay Tufan M, Küçük C (2015) Natural radioactivity measurements and dose assessments in sand samples collected from Zonguldak beaches in Turkey. J Radioanal Nucl Chem 303:2227–2232
31. Kobya Y, Taşkin H, Yeşilkanat CM, Varinlioğlu A, Korcak S (2015) Natural and artificial radioactivity assessment of dam lakes sediments in Çoruh River, Turkey. J Radioanal Nucl Chem 303:287–295
32. Song G, Chen D, Tang Z, Zhang Z, Xie W (2012) Natural radioactivity levels in topsoil from the Pearl River Delta Zone, Guangdong, China. J Environ Radioact 103:48–53
33. Agbalagba EO, Onoja RA (2011) Evaluation of natural radioactivity in soil, sediment and water samples of Niger Delta (Biseni) flood plain lakes, Nigeria. J Environ Radioact 102:667–671
34. Rahman SU, Matiullah Malik F, Rafique M, Anwar J, Ziafat M, Jabbar A (2011) Measurement of naturally occurring/fallout radioactive elements and assessment of annual effective dose in soil samples collected from four districts of the Punjab Province, Pakistan. J Radioanal Nucl Chem 287:647–655
35. Ele Abiama P, Owono Ateba P, Ben-Bolie GH, Ekobena FHP, El Khoukhi T (2010) High background radiation investigated by gamma spectrometry of the soil in the southwestern region of Cameroon. J Environ Radioact 101:739–743
36. Ahmed NK, Mohamed El-Arabi AG (2005) Natural radioactivity in farm soil and phosphate fertilizer and its environmental implications in Qena governorate, Upper Egypt. J Environ Radioact 84:51–64
37. Yang Ya-xin Wu, Xin-min Jiang Zhong-ying, Wang Wei-xing Lu, Ji-gen Lin J, Lei-Ming Wang, Yuan-fu Hsia (2005) Radioactivity concentrations in soils of the Xiazhuang granite area, China. Appl Radiat Isot 63:255–259
38. Alabdullah J, Michel H, Barci V, Féraud G, Barci-Funel G (2013) Spatial and vertical distributions of natural and anthropogenic radionuclides and cesium fractionation in sediments of the Var river and its tributaries (southeast France). J Radioanal Nucl Chem 298:25–32
39. Kannan V, Rajan MP, Iyengar MAR, Ramesh R (2002) Distribution of natural and anthropogenic radionuclides in soil and beach sand samples of Kalpakkam (India) using hyper pure germanium (HPGe) gamma ray spectrometry. Appl Radiat Isot 57:109–119
40. Dragović S, Janković-Mandić L, Dragović R, Đorđević M, Đokić M, Kovačević J (2014) Lithogenic radionuclides in surface soils of Serbia: spatial distribution and relation to geological formations. J Geochem Explor 142:4–10
41. Chakraborty SR, Azim R, Rezaur Rahman AKM, Sarker R (2013) Radioactivity concentrations in soil and transfer factors of radionuclides from soil to grass and plants in the Chittagong city of Bangladesh. J Phys Sci 24:95–113
42. Latif SA, Kinsara AA, Molla NI, Nassef MH (2014) Natural radioactivity measurements in agricultural soil, fertilizer and crops in some specific areas of Kingdom of Saudi Arabia. Radiochim Acta 102:513–522
43. Murty VRK, Karunakara N (2008) Natural radioactivity in the soil samples of Botswana. Radiat Meas 43:1541–1545
44. Srilatha MC, Rangaswamy DR, Sannappa J (2015) Measurement of natural radioactivity and radiation hazard assessment in the soil samples of Ramanagara and Tumkur districts, Karnataka, India. J Radioanal Nucl Chem 303:993–1003
45. Pinto P, Yerol N (2014) Studies on the seasonal variation and vertical profiles of natural radionuclides in high background radiation areas of Kerala on the south west coast of India. J Radioanal Nucl Chem 302:813–817
46. Usikalu MR, Maleka PP, Malik M, Oyeyemi KD, Adewoyin OO (2015) Assessment of geogenic natural radionuclide contents of soil samples collected from Ogun State, South western, Nigeria. Int J Radiat Res 13:355–361
47. Mohapatra S, Sahoo SK, Dubey JS, Patra AC, Thakur VK, Tripathy SK, Vidyasagar D, Godbole SV, Ravi PM, Tripathi RM (2015) On the radiological assessment of natural and fallout radioactivity in a natural high background radiation area at Odisha, India. J Radioanal Nucl Chem 303:2081–2092
48. Srivastava A, Lahiri S, Maiti M, Knolle F, Hoyler F, Scherer UW, Schnug EW (2014) Study of naturally occurring radioactive material (NORM) in top soil of Punjab State from the North Western part of India. J Radioanal Nucl Chem 302:1049–1052
49. Sartandel SJ, Chinnaesakki S, Bara SV, Krishna NS, Vinod Kumar A, Tripathi RM (2014) Assessment of natural and fallout radioactivity in soil samples of Visakhapatnam. J Radioanal Nucl Chem 299:337–342
50. Rajeshwari T, Rajesh S, Kerur BR, Anilkumar S, Krishnan N, Pant AD (2014) Natural radioactivity studies of Bidar soil samples using gamma spectrometry. J Radioanal Nucl Chem 300:61–65
51. Dusane CB, Mishra S, Sahu SK, Pandit GG (2014) Distribution of ^{238}U, ^{226}Ra, ^{232}Th and ^{40}K in soil samples around Tarapur, India. J Radioanal Nucl Chem 302:1435–1440
52. Canbazoğlu C, Turhan Ş, Bakkal S, Uğur FA, Gören E (2013) Analysis of gamma emitting radionuclides (terrestrial and anthropogenic) in soil samples from Kilis province in south Anatolia, Turkey. Ann Nucl Energ 62:153–157
53. Aközcan S (2012) Distribution of natural radionuclide concentrations in sediment samples in Didim and Izmir Bay (Aegean Sea-Turkey). J Environ Radioact 112:60–63
54. Al-Sharkawy A, Hiekal MThS, Sherif MI, Badran HM (2012) Environmental assessment of gamma-radiation levels in stream sediments around Sharm El-Sheikh, South Sinai. Egypt. J Environ Radioact 112:76–82
55. Wasim M, Iqbal S, Ali M (2016) Radiological and elemental analysis of soils from Hunza in Central Karakoram using gamma-ray spectrometry and k_0-instrumental neutron activation analysis. J Radioanal Nucl Chem 307:891–898
56. Hannan M, Wahid K, Nguyen N (2015) Assessment of natural and artificial radionuclides in Mission (Texas) surface soils. J Radioanal Nucl Chem 305:573–582
57. Jallad KN (2015) Radioactive characterization of sand samples from Failaka Island in Kuwait. J Radioanal Nucl Chem 303:733–741

58. Santawamaitre T, Malain D, Al-Sulaiti HA, Bradley DA, Matthews MC, Regan PH (2014) Determination of ^{238}U, ^{232}Th and ^{40}K activity concentrations in riverbank soil along the Chao Phraya river basin in Thailand. J Environ Radioact 138:80–86
59. Özmen SF, Cesur A, Boztosun I, Yavuz M (2014) Distribution of natural and anthropogenic radionuclides in beach sand samples from Mediterranean Coast of Turkey. Radiat Phys Chem 103:37–44
60. Tufail M, Asghar M, Akram M, Javied S, Khan K, Mujahid SA (2013) Measurement of natural radioactivity in soil from Peshawar basin of Pakistan. J Radioanal Nucl Chem 298:1085–1096
61. http://www.nndc.gov/chart. Accessed 30 Dec 2016