J Radioanal Nucl Chem (2016) 310:1381–1396
DOI 10.1007/s10967-016-4988-x

CrossMark

Measurement of naturally occurring radioactive materials, ^{238}U and ^{232}Th: anomalies in photopeak selection

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Received: 19 June 2016 / Published online: 17 August 2016

Abstract There are more than 200 photopeaks of various daughter radionuclides of ^{238}U and ^{232}Th series, some of which have been randomly used for quantitative measurement of U/Th in natural samples. It has been observed that arbitrariness in photopeak selection may fail to stipulate statistically consistent data. This paper judiciously selects set of three photopeaks from each series whose respective averages could present statistically reliable measurement of ^{238}U and ^{232}Th based on minimum relative standard deviation (RSD) under the selected photopeaks. RSD is also proposed as an important parameter in NORM measurement.

Keywords Naturally occurring radionuclide materials (NORMs) · ^{238}U and ^{232}Th measurement · Gamma-ray spectrometry · Photopeak selection

Introduction

The ubiquitous natural background radiation felt on the Earth is mainly due to terrestrial and cosmic radiation [1]. Long-lived, primordial naturally occurring radionuclides or NORMs like ^{238}U ($T_{1/2} = 4.468 \times 10^9$a), ^{235}U ($T_{1/2} = 7.04 \times 10^8$a), ^{232}Th ($T_{1/2} = 1.40 \times 10^{10}$a) and ^{40}K ($T_{1/2} = 1.248 \times 10^9$a) have geological presence since formation of the Earth [2]. They along with their daughter products (^{226}Ra, ^{212}Pb, ^{212}Bi, ^{228}Ac, ^{210}Pb, ^{208}Tl, etc.) are prime contributors of background radiation. The global mean of ^{238}U, ^{232}Th and ^{40}K in terrestrial system reported are 35, 45 and 420 Bq/kg respectively [3]. The enhanced concentration of natural and anthropogenic radionuclides resulting due to human activities like mining, refining, nuclear experiments, etc., is termed as technologically enhanced naturally occurring radioactive materials or TeNORMs [4]. It could be further stated that nuclear weapon testing (1960–1970), Chernobyl accident (1986) and recent Fukushima-Daichii accident (2011) have made significant contribution to the global inventory of anthropogenic radionuclides.

There are several reports on measurement of NORMs (^{238}U, ^{232}Th and ^{40}K) all over the globe. These measurements have come out from laboratories with moderate experimental facilities as well as from renowned laboratories equipped with state-of-art detectors. The sample size for NORM measurement generally varied in the reported works from 20 to 50 g, which was further normalized to Bq/kg. The estimated radioactivity level of ^{238}U and ^{232}Th in such sample could be around only 1–2 Bq. Therefore slight discrepancy in measurement would reflect in terms of high uncertainty in the final normalized value. Low-level radiation measurement requires selection of high efficiency detector, accurate energy and efficiency calibration, optimum counting time, proper selection of photopeaks, etc. The literature review reveals that researchers in many cases have arbitrarily fixed the above-mentioned experimental parameters. In the present work we have discussed about proper selection of photopeaks from the daughter radionuclides of ^{238}U and ^{232}Th series to get

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Table 1 List of photopeaks taken by different groups of researchers for measurement of ^{238}U and ^{232}Th activity

Sl No.	Reported by	Parent radionuclide ^{238}U		Parent radionuclide ^{232}Th	
		Photopeak	Energy (keV)	Photopeak	Energy (keV)
1	Mohapatra et al. [1]	^{234}Th	63.29	^{208}Tl	2614.53
		^{214}Pb	351.93		
		^{214}Bi	609.31, 1764.49		
2	Srivastava et al. [3]	^{214}Pb	351.93	^{228}Ac	911.20, 968.97
		^{214}Bi	609.31		
3	Sartandel et al. [5]	^{234}Th	63.29	^{228}Ac	Not mentioned
		^{214}Pb	Not mentioned	^{208}Tl	Not mentioned
		^{214}Bi	Not mentioned		
4	Wang et al. [6]	^{234}Th	63.29, 92.6	^{228}Ac	338.32, 911.20, 968.97
		^{214}Pb	351.93		
		^{214}Bi	609.31		
5	Al-Sharkawy et al. [7]	^{234}Th	63.29, 92.38, 92.8	^{228}Ac	911.20, 964.76, 968.97, 1588.19
		^{214}Pb	295.2, 351.93		
		^{214}Bi	609.31, 1120.29, 1764.49, 2204.21, 2447.86	^{208}Tl	583.19, 860.56, 2614.53
				^{212}Bi	727.33, 1620.7
6	Chowdhury et al. [8]	^{234}Th	63.29	^{212}Pb	238.63
		^{234m}Pa	1001.03	^{212}Bi	727.33
		^{214}Pb	295.22, 351.93	^{228}Ac	338.32, 911.20, 968.97
		^{214}Bi	609.31, 1120.29, 1764.49	^{208}Tl	583.19
7	Janković et al. [9]	^{234}Th	63.29	^{228}Ac	911.20
		^{234}Pa	1001.03		
		^{214}Pb	351.93		
		^{214}Bi	609.31		
8	Song et al. [10]	^{234}Th	63.29, 92.6	^{228}Ac	911.20
		^{214}Pb	295.22, 351.93	^{212}Pb	238.63
		^{214}Bi	609.31	^{208}Tl	583.19
9	Mahur et al. [11]	^{214}Pb	295.22, 351.93	^{228}Ac	338.32, 463.00, 911.20, 968.97
		^{214}Bi	609.31, 1120.29, 1764.49	^{212}Bi	727.33
		^{234}Pa	1001.03	^{212}Pb	238.63
10	Santawamaitre et al. [12]	^{226}Ra	186.21	^{228}Ac	338.32, 911.20,
		^{214}Pb	295.22, 351.93		968.97
		^{214}Bi	609.31, 1120.29, 1238.11, 1764.49, 2204.21	^{212}Pb	238.63, 300.09
				^{212}Bi	727.33, 1620.5
				^{208}Tl	583.19, 2614.53
11	Gupta et al. [13]	^{226}Ra	186.21	^{228}Ac	338.32, 463.00, 911.20, 968.97
		^{214}Pb	295.22, 351.93	^{212}Bi	727.33
		^{214}Bi	609.31, 1120.29, 1764.49	^{212}Pb	238.63
12	Boukhenfouf and Boucenna [14]	^{226}Ra	186.21	^{228}Ac	338.32, 911.20, 964.76 968.97
		^{214}Pb	295.22, 351.93	^{212}Pb	238.63
		^{214}Bi	609.31, 1120.29, 1764.49	^{208}Tl	583.19, 860.56
13	Aközcan [15]	^{226}Ra	186.21	^{228}Ac	911.20
		^{214}Pb	351.93	^{208}Tl	583.19
		^{214}Bi	609.31		

Table 1 continued

Sl No.	Reported by	Parent radionuclide ^{238}U		Parent radionuclide ^{232}Th	
		Photopeak	Energy (keV)	Photopeak	Energy (keV)
14	Yang et al. [16]	^{226}Ra	186.21	^{212}Pb	238.63
		^{214}Pb	351.93	^{228}Ac	338.32, 911.20, 968.97, 974.2
		^{214}Bi	609.31, 768.35, 1120.29, 1238.11, 1764.49		
				^{208}Tl	583.19
15	Alaamer [17]	^{226}Ra	186.21	^{228}Ac	911.20
		^{214}Pb	351.93	^{208}Tl	583.19
		^{214}Bi	609.31		
16	Kurnaz et al. [18]	^{226}Ra	186.21	^{228}Ac	911.20
		^{214}Pb	351.93	^{208}Tl	583.19
		^{214}Bi	609.31		
17	Ele Abiama et al. [20]	^{214}Bi	609.31, 768.35, 1120.29, 1238.11, 1764.49	^{212}Pb	238.63
				^{228}Ac	338.32, 911.20, 968.97, 974.2
				^{208}Tl	583.19
18	Aytekin et al. [21]	^{214}Pb	295.22, 351.93	^{208}Tl	583.19
		^{214}Bi	609.31	^{228}Ac	338.32, 911.20
19	Alfonso et al. [22]	^{214}Pb	295.22, 351.93	^{228}Ac	911.20
		^{214}Bi	609.31	^{212}Pb	238.63
20	Hannan et al. [23]	^{214}Bi	609.31	^{228}Ac	911.20
21	Adukpo et al. [24]	^{214}Bi	609.31	^{228}Ac	911.20
22	Ravisankar et al. [25]	^{214}Bi	1764.49	^{208}Tl	2614.53
23	Potoki et al. [26]	^{214}Bi	Not mentioned	^{228}Ac	Not mentioned
				^{208}Tl	
24	Bakim and Ugur Görgün [27]	^{214}Pb	295.22, 351.93	^{208}Tl	2614.53
25	Kobya et al. [28]	^{214}Pb	Not mentioned	^{208}Tl	Not mentioned
		^{214}Bi		^{212}Pb	
26	Isinkaye and Emelue [29]	^{214}Bi	1764.49	^{208}Tl	2615.53
27	Chakraborty [30]	^{214}Pb	351.93	^{208}Tl	583.19
		^{214}Bi	609.31, 1120.29	^{228}Ac	911.20
28	Manigandan and Chandar Shekar [31]	^{214}Bi	1764.49	^{208}Tl	2614.53
29	Yadav et al. [32]	^{214}Bi	1764.49	^{208}Tl	2614.53
30	Bala et al. [33]	^{214}Bi	1764.49	^{208}Tl	2614.53
31	Canbazoğlu et al. [34]	^{214}Pb	351.93	^{228}Ac	911.20
		^{214}Bi	609.31	^{208}Tl	583.19
32	Tchokossa et al. [35]	^{214}Pb	351.93	^{228}Ac	911.20
		^{214}Bi	609.31, 1120.29	^{208}Tl	583.19
33	Singh et al. [36]	^{214}Bi	1764.49	^{208}Tl	2614.53
34	Kannan et al. [37]	^{214}Bi	609.31	^{228}Ac	911.20
35	Alatise et al. [38]	^{214}Bi	1764.49	^{208}Tl	2614.53
36	Agbalagba and Onoja [39]	^{214}Pb	295.22	^{212}Pb	238.63
37	Ahmed et al. [40]	^{214}Pb	351.93	^{228}Ac	911.20
		^{214}Bi	609.31	^{208}Tl	583.19
38	Rajeshwari et al. [41]	^{214}Pb	351.93	^{228}Ac	911.20
		^{214}Bi	609.31	^{212}Pb	238.63
				^{208}Tl	583.19, 2614.53

Table 1 continued

Sl No.	Reported by	Parent radionuclide ^{238}U		Parent radionuclide ^{232}Th	
		Photopeak	Energy (keV)	Photopeak	Energy (keV)
39	Matiullah and Malik [42]	^{214}Pb	295.22, 351.93	^{228}Ac	338.32, 911.20, 968.97
		^{214}Bi	609.31, 1120.29		
40	Pinto and Yerol [43]	^{214}Bi	609.31, 1120.29, 1764.49	^{208}Tl	583.19, 2614.53
41	Jeevarenuka et al. [44]	^{214}Bi	1764.49	^{208}Tl	2614.53
42	El-Taher and Madkour [45]	^{214}Pb	351.93	^{212}Pb	238.63
		^{214}Bi	609.31, 1764.49	^{228}Ac	911.20
43	Powell et al. [46]	^{214}Pb	351.93	^{228}Ac	911.20
		^{214}Bi	609.31		
44	Özmen et al. [47]	^{214}Pb	351.93	^{228}Ac	911.20
		^{214}Bi	609.31		
45	Rani and Singh [48]	^{214}Bi	1764.49	^{208}Tl	2614.53
46	Al-Jundi et al. [49]	^{214}Pb	351.93	^{212}Pb	238.63
		^{214}Bi	609.31	^{228}Ac	911.20, 968.97
				^{208}Tl	583.19
47	Murty and Karunakara [50]	^{214}Pb	351.93	^{228}Ac	911.20
		^{214}Bi	609.31, 1120.29, 1764.49	^{208}Tl	583.19, 2614.53

reliable estimate of uranium and thorium present at ultra-low level concentration in natural matrices.

Different investigators have measured activity of ^{238}U and ^{232}Th by selecting different photopeaks; most of them selected multiple gamma-peaks from different daughter radionuclides of the corresponding series, and presented the average value of the activity of ^{238}U and ^{232}Th. Even when multiple photopeaks were used, different groups selected different sets of photopeaks (not necessarily the most intense peaks). In Table 1, we list the sets of photopeaks taken by various research groups to measure ^{238}U and ^{232}Th activity [1, 3, 5–18, 20–50]. A careful look to this table shows some interesting and apparently illogical choice of photopeaks. Few of them are illustrated here. Mohapatra et al. [1], Sartandel et al. [5], Wang et al. [6], Al-Sharkawy et al. [7], Chowdhury et al. [8], Janković et al. [9] and Song et al. [10], have considered low intensity (4.8 % only) 63.29 keV (^{234}Th) photopeak for ^{238}U activity measurement. Chowdhury et al. [8], Janković et al. [9], Mahur et al. [11] have included very low intensity (0.65 % only) 1001.03 keV photopeak of ^{234}Pa along with other peaks to measure ^{238}U. Many authors [12–18] have considered 186.21 keV photopeak of ^{226}Ra, member of ^{238}U decay series, to measure ^{238}U. However, this photopeak may have significant interference from ^{235}U, which could be as high as 11.4 % [19], therefore should be avoided otherwise correction for ^{235}U should be made. Al-Sharkawy et al. [7], have selected both 92.38 and 92.8 keV photopeaks for ^{238}U measurement. Both of these photopeaks have low intensities (2.81 and 2.77 % respectively). They also reported that they have measured using 50 % *p*-type HPGe detector, which normally will be unable to resolve these photopeaks. Similarly for ^{232}Th measurement many authors [7, 14, 16, 20] have measured 964.76 keV (4.99 %) and 974.2 keV (0.05 %) photopeaks, both from ^{228}Ac. These peaks are situated on the shoulder and on the trail of 968.97 (15.8 %) keV photopeak respectively and therefore difficult to have statistically reliable area count.

The pertinent question therefore boils down to which photopeaks are preferable for low-level measurement? In this paper we made an attempt towards optimization of NORM measurement (^{238}U and ^{232}Th) with respect to selection of photopeaks from different daughter radionuclides of ^{238}U and ^{232}Th decay series. To the best of our knowledge, despite large number of measurements on NORM reported in literature, this type of detailed analysis has been attempted for the first time.

Initial screening of photopeaks

In Table 2, we list the gamma energies of different daughter radionuclides of ^{238}U, ^{232}Th and major photopeaks of ^{235}U. As ultra-low level activities are measured in NORM measurement, we excluded the photopeaks having

Table 2 List of different photopeaks of the radionuclides belonging to natural decay series and their suitability for measurement

Parent	Daughter	Energy (keV)	Intensity (%)	Score
^{238}U	^{234}Th	63.29	4.8	1
^{232}Th	^{208}Tl	72.805 (X-ray)	2.09	0[a,b]
^{238}U	^{214}Pb	74.815 (X-ray)	4.8	0[b]
^{232}Th	^{212}Pb	74.815 (X-ray)	10.41	0[b]
^{232}Th	^{208}Tl	74.969 (X-ray)	3.51	0[b]
^{238}U	^{214}Pb	77.107 (X-ray)	8	0[b]
^{232}Th	^{212}Pb	77.107 (X-ray)	17.5	0[b]
^{235}U	^{223}Ra	81.069 (X-ray)	15.2	0
^{235}U	^{223}Ra	83.787 (X-ray)	25.2	0
^{232}Th	^{212}Pb	86.83 (X-ray)	2.09	0[a,b]
^{232}Th	^{212}Pb	87.349 (X-ray)	4.01	0[b]
^{238}U	^{234}Th	92.38	2.81	0[a,b]
^{238}U	^{234}Th	92.8	2.77	0[a,b]
^{232}Th	^{228}Ac	93.35 (X-ray)	3.19	0[a,b]
^{235}U	^{235}U	93.35 (X-ray)	11	0[b]
^{238}U	^{234}Pa	94.654 (X-ray)	14.4	0[b]
^{238}U	^{234}Pa	98.434 (X-ray)	23.3	0[b]
^{238}U	^{234}Pa	99.853	3.2	0[a,b]
^{238}U	^{234}Pa	110.421 (X-ray)	2.87	0[a,b]
^{238}U	^{234}Pa	111.298 (X-ray)	5.44	0[b]
^{238}U	^{234}Pa	114.445 (X-ray)	2.1	0[a,b]
^{232}Th	^{228}Ac	129.065	2.42	0[a]
^{238}U	^{234}Pa	131.29	18	1
^{235}U	^{235}U	143.764	10.96	0
^{238}U	^{234}Pa	152.72	6	1
^{235}U		185.712	57.2	0[b]
^{238}U	^{226}Ra	186.2111	3.59	0[a,b]
^{232}Th	^{228}Ac	209.253	3.89	0[a]
^{238}U	^{234}Pa	226.5	4.2	0[b]
^{238}U	^{234}Pa	227.25	5.8	0[b]
^{235}U	^{227}Th	235.971	12.3	0[b]
^{232}Th	^{212}Pb	238.632	43.3	0[b]
^{232}Th	^{224}Ra	240.986	4.1	0[b]
^{238}U	^{214}Pb	241.997	7.43	0[b]
^{238}U	^{234}Pa	249.22	2.5	0[a]
^{235}U	^{223}Ra	269.46	13.7	0[b]
^{232}Th	^{228}Ac	270.245	3.46	0[b]
^{235}U	^{219}Rn	271.23	10.8	0[b]
^{232}Th	^{208}Tl	277.351	6.31	1
^{238}U	^{234}Pa	293.79	2.99	0[a]
^{238}U	^{214}Pb	295.224	19.3	1
^{232}Th	^{212}Pb	300.087	3.28	0[a]
^{232}Th	^{228}Ac	328	2.95	0[a]
^{232}Th	^{228}Ac	338.32	11.27	1
^{235}U	^{211}Bi	351.059	12.91	0[b]
^{238}U	^{214}Pb	351.9332	37.6	1

Table 2 continued

Parent	Daughter	Energy (keV)	Intensity (%)	Score
^{238}U	^{234}Pa	369.5	2.47	0[a]
^{232}Th	^{228}Ac	463.004	4.4	1
^{232}Th	^{208}Tl	510.77	22.6	0[c]
^{238}U	^{234}Pa	568.9	3.6	0[b]
^{238}U	^{234}Pa	569.5	8.2	0[b]
^{232}Th	^{208}Tl	583.19	84.5	1
^{238}U	^{214}Bi	609.31	46.1	1
^{238}U	^{234}Pa	699.03	3.6	0[a]
^{238}U	^{234}Pa	705.9	2.27	0[a]
^{232}Th	^{212}Bi	727.33	6.58	1
^{238}U	^{234}Pa	733.39	6.9	1
^{238}U	^{234}Pa	742.81	2.06	0[a]
^{238}U	^{214}Bi	768.356	4.94	1
^{232}Th	^{228}Ac	794.947	4.25	0[b]
^{238}U	^{234}Pa	796.1	2.58	0[a,b]
^{238}U	^{234}Pa	805.8	2.52	0[a]
^{238}U	^{234}Pa	831.5	4.12	1
^{232}Th	^{208}Tl	860.564	12.42	1
^{238}U	^{234}Pa	876	2.524	0[a]
^{238}U	^{234}Pa	880.5	4.2	0[b]
^{238}U	^{234}Pa	880.5	6	0[b]
^{238}U	^{234}Pa	883.24	9.6	0[b]
^{238}U	^{234}Pa	898.67	3.24	0[a]
^{232}Th	^{228}Ac	911.204	25.8	1
^{238}U	^{234}Pa	925	7.8	0[b]
^{238}U	^{234}Pa	926.72	7.2	0[b]
^{238}U	^{214}Bi	934.061	3.03	0[a]
^{238}U	^{234}Pa	946	13.4	1
^{232}Th	^{228}Ac	964.766	4.99	0[b]
^{232}Th	^{228}Ac	968.97	15.8	1
^{238}U	^{234}Pa	980.3	2.7	0[a]
^{238}U	^{234m}Pa	1001.03	0.837	0[a]
^{238}U	^{214}Bi	1120.287	15.1	1
^{238}U	^{214}Bi	1238.11	5.79	1
^{238}U	^{214}Bi	1377.669	4	0[a]
^{238}U	^{214}Bi	1407.98	2.15	0[a]
^{238}U	^{214}Bi	1509.228	2.11	0[a]
^{232}Th	^{228}Ac	1588.19	3.22	0[a]
^{238}U	^{214}Bi	1729.595	2.92	0[a]
^{238}U	^{214}Bi	1764.49	15.4	1
^{238}U	^{214}Bi	1847.42	2.11	0[a]
^{238}U	^{214}Bi	2204.21	5.08	1
^{232}Th	^{208}Tl	2614.53	99	1

[a] Low intensity peaks

[b] Closely spaced, detector cannot resolute

[c] Coincides with 511 keV annihilation peak

Table 3 Photopeaks of ^{238}U series suitable for measurement of ^{238}U after preliminary screening

Sl. no.	Daughter	Energy (keV)	Intensity (%)
1	^{234}Th	63.29	4.8
2	^{234}Pa	131.29	18
3	^{234}Pa	152.72	6
4	^{214}Pb	295.22	19.3
5	^{214}Pb	351.93	37.6
6	^{214}Bi	609.31	46.1
7	^{234}Pa	733.39	6.9
8	^{214}Bi	768.35	4.94
9	^{234}Pa	831.5	4.12
10	^{234}Pa	946	13.4
11	^{214}Bi	1120.29	15.1
12	^{214}Bi	1238.11	5.79
13	^{214}Bi	1764.49	15.4
14	^{214}Bi	2204.21	5.08

Table 4 Photopeaks of ^{232}Th series suitable for measurement of ^{232}Th after preliminary screening

Sl. no.	Daughter	Energy (keV)	Intensity (%)
1	^{208}Tl	277.35	6.31
2	^{228}Ac	338.32	11.27
3	^{228}Ac	463.00	4.4
4	^{208}Tl	583.19	84.5
5	^{212}Bi	727.33	6.58
6	^{208}Tl	860.56	12.42
7	^{228}Ac	911.20	25.8
8	^{228}Ac	968.97	15.8
9	^{208}Tl	2614.53	99

intensities less than 2 % in ^{238}U and ^{232}Th series. However, some of them are even listed in the table, if frequently taken by different research groups (e.g., 1001.00 keV photopeak of ^{234}Pa having intensity 0.65 %). Also we have excluded all the photopeaks of ^{210}Tl and ^{206}Tl, which belong to ^{238}U series. The reason of exclusion is extremely low population from their parent radionuclides, e.g. ^{214}Bi decays to ^{210}Tl with branching ratio 0.02 % only. Similarly, ^{210}Bi decays to ^{206}Tl by emitting α-particle with only 1.3×10^{-4} % probability. We have assigned a score to each photopeak listed in Table 2, 0 or 1 where 0 denotes unsuitability of the gamma line for quantification of the parent radionuclide of the series; whereas the score 1 denotes the suitability of the gamma line based on the preliminary observation. The reason for assigning 0 is based on either very low intensity in the specific energy region or possibility of overlapping with the neighboring photopeaks either from the same series or from inter-series interference. While overlapping with another photopeak is considered, it is assumed HPGe detectors are used for NORM measurement that have generally 2–3 keV resolution in the higher energy region and ~1–2 keV in the lower energy region. All the photopeaks from ^{235}U series have been assigned score zero because of its very low natural abundance, 0.7204 %. However, they have been included in the table to show possible interference to the radionuclides, like 185.71 keV interferring with 186.21 keV ^{226}Ra photopeak and 351.06 keV interferring with 351.93 keV ^{214}Pb photopeak. From the preliminary screening it is revealed that only 14 photopeaks from ^{238}U series, and 9 photopeaks from ^{232}Th series qualify for quantitative measurements of low-level NORMs. Tables 3 and 4 represent these useful photopeaks as deduced from Table 2 for measurement of the activity of uranium and thorium respectively. Rest of the investigation has been carried out using only the useful photopeaks.

Experimental

Four soil samples were collected from different parts of India, e.g., from Sundarban region (SB1, SB2) and from Punjab state (PU1 and PU2). It is noteworthy to mention Sundarban is world's largest mangrove ecosystem known for its luxuriant floral-faunal diversity. The samples were air-dried until moisture was driven out and then further pulverized in grinder to obtain homogenized form. Each of the pulverized samples were weighed to 50 g, hermetically sealed in leak-proof petri-plates and kept aside for 40 days to ensure the state of secular equilibrium. The dimension of the petri-plates as well as that of the soil samples was 7.5 cm diameter and 1.1 cm height. In addition to four test samples, four standards (two each of ^{238}U and ^{232}Th) were also prepared. For preparation of two ^{238}U standards (2 and 5 dps), weighed amount of IAEA Uranium Ore (Pitchblende); S-8 standard (0.35 and 0.14 g correspond to 5 and 2 dps respectively) was taken in leak-proof petri-plate. For ^{232}Th standards (2 and 5 dps), weighed amount of thorium acetate, $[Th(CH_3COO)_4]$ (0.995 and 2.49 mg correspond to 2 and 5 dps respectively) was taken in leak-proof petri-plate. To maintain the geometry at par with the test samples, all the four standard samples were mixed thoroughly with silica gel to attain the total weight of 50 g, equivalent to the sample size. The petri-plates were also hermetically sealed for 40 days to establish the secular equilibrium between the parent and daughter isotopes. One of the two standards (2 dps) was used as standard for all

Table 5 Calculated activity of different daughter radionuclides of ^{238}U under different photopeaks using 2 dps as U standard (SU)

Sl. no.	Radionuclide	Photopeak (keV)	Intensity (%)	SB1	SB2	PU1	PU2	U-5 dps
1	^{234}Th	63.29	4.8	4.20 ± 0.28	3.69 ± 0.26	5.53 ± 0.36	5.17 ± 0.34	5.47 ± 0.34
2	^{234}Pa	131.29	18	0	0	0	0	0
3	^{234}Pa	152.69	6	0	0	0	0	0
4	^{214}Pb	295.22	19.3	1.48 ± 0.08	1.16 ± 0.08	2.09 ± 0.11	1.69 ± 0.09	5.86 ± 0.21
5	^{214}Pb	351.93	37.6	1.39 ± 0.04	1.04 ± 0.04	2.20 ± 0.06	1.81 ± 0.05	4.77 ± 0.10
6	^{214}Bi	609.31	46.1	1.56 ± 0.06	1.24 ± 0.05	2.14 ± 0.07	2.07 ± 0.07	4.95 ± 0.13
7	^{234}Pa	733.39	6.9	0	0	0	0	0
8	^{214}Bi	768.4	4.94	0	0	0	0	0
9	^{234}Pa	831.5	4.12	0	0	0	0	0
10	^{234}Pa	946	13.4	0	0	0	0	0
11	^{214}Bi	1120.29	15.1	1.86 ± 0.26	1.56 ± 0.24	3.95 ± 0.44	2.55 ± 0.32	8.23 ± 0.79
12	^{214}Bi	1238.11	5.79	1.27 ± 0.33	0	1.79 ± 0.39	0.76 ± 0.31	3.7 ± 0.48
13	^{214}Bi	1764.49	15.4	2.20 ± 0.22	1.6 ± 0.17	2.69 ± 0.26	2.32 ± 0.23	6.88 ± 0.55
14	^{214}Bi	2204.21	5.08	1.86 ± 0.50	0.72 ± 0.26	3.34 ± 0.75	2.52 ± 0.59	6.13 ± 1.19
Mean values of activities obtained from photopeaks having serial nos. 1, 4, 5, 6, 11, 12, 13 and 14 (%RSD)				1.98 ± 0.76 (47.98)	1.38 ± 0.48 (76.81)	2.97 ± 1.06 (42.42)	2.36 ± 0.86 (53.81)	5.75 ± 1.66 (24)
Mean values of activities obtained from photopeaks having serial nos. 4,5,6 and 13 (%RSD)				1.66 ± 0.25 (22.29)	1.26 ± 0.2 (19.05)	2.28 ± 0.29 (11.84)	1.97 ± 0.26 (14.21)	5.62 ± 0.61 (17.25)
Mean values of activities obtained from photopeaks having serial nos. 4, 5 and 6 (%RSD)				1.48 ± 0.11 (5.4)	1.15 ± 0.1 (8.69)	2.14 ± 0.14 (2.8)	1.86 ± 0.13 (10.21)	5.19 ± 0.27 (11.37)
Mean values of activities obtained from photopeaks having serial nos. 4, 5, 6, 12, 13 and 14 (%RSD)				1.63 ± 0.65 (20.85)	0.96 ± 0.33 (57.3)	2.38 ± 0.9 (23.10)	1.86 ± 0.72 (33.33)	5.38 ± 1.42 (21.19)
Mean values of activities obtained from photopeaks having serial nos. 4,5,6,13 and 14 (%RSD)				1.70 ± 0.56 (19.41)	1.15 ± 0.33 (27.83)	2.49 ± 0.81 (21.28)	2.08 ± 0.65 (16.35)	5.72 ± 1.33 (15.21)
Mean values of activities obtained from photopeaks having serial nos. 4, 5, 6 and 14 (%RSD)				1.57 ± 0.51 (12.74)	1.04 ± 0.28 (22.11)	2.44 ± 0.77 (24.59)	2.02 ± 0.61 (17.82)	5.43 ± 1.22 (12.33)

$RSD = \frac{\text{standard deviation}}{\text{mean value}} \times 100$; RSD values have been given in parenthesis

Table 6 Photopeaks of ^{238}U series suitable for quantitative analysis of ^{238}U

Sl. no.	Daughter	Energy (keV)	Intensity (%)
1	^{234}Th	63.29	4.8
2	^{214}Pb	295.22	19.3
3	^{214}Pb	351.93	37.6
4	^{214}Bi	609.31	46.1
5	^{214}Bi	1120.29	15.1
6	^{214}Bi	1238.11	5.79
7	^{214}Bi	1764.49	15.4
8	^{214}Bi	2204.21	5.08

measurements in both cases of ^{238}U and ^{232}Th. The other one (5 dps) was used as sample of known activity (SU for ^{238}U and STh for ^{232}Th) to validate the result.

All samples and standards were measured for 75000 s using reverse electrode coaxial high-purity Germanium (HPGe) detector with 50 % relative efficiency and FWHM (full width at half maxima) of 3.3 and 0.96 keV respectively at 1.33 MeV and 122 keV. Shielding of this detector had CANBERRA model 747 lead shield with 9.5 mm thick low carbon outer jacket, 10 cm thick low background lead as bulk shield, also graded lining of 1 mm tin and 1.6 mm copper preventing the interference by lead X-rays [3]. Samples were kept at 1 cm distance from top of central HPGe detector. Energy calibration was performed using single elemental standards or point sources of ^{133}Ba, ^{60}Co, ^{137}Cs and ^{152}Eu. Count of 50 g silica gel was taken also for 75,000 s in a similar petri-plate. This was considered as background spectrum. This background spectrum was stripped from all sample and

Table 7 Activity of ^{238}U obtained for four test samples with different sets of photopeaks as taken by different researchers (see Table 1)

Sl. no.	Research group	Photopeaks (keV)	Intensity (%)	Average ^{238}U activity (Bq), relative standard deviation (RSD in %)				
				SB1	SB2	PU1	PU2	U-5dps
1	Mohapatra et al. [1]	63.29	4.8	2.34 ± 0.37	1.89 ± 0.32	3.14 ± 0.45	2.84 ± 0.42	5.51 ± 0.67
		351.93	37.6	55.12	64.55	51.27	55.28	17.42
		609.31	46.1					
		1764.49	15.4					
2	Wang et al. [6]	63.29	4.8	2.38 ± 0.29	1.99 ± 0.27	3.29 ± 0.37	3.02 ± 0.35	5.06 ± 0.38
		92.38[a]	2.77	65.97	73.87	58.97	61.92	7.11
		351.93	37.6					
		609.31	46.1					
3	Al-Sharkawy et al. [7]	63.29	4.8	2.08 ± 0.68	1.57 ± 0.48	3.13 ± 0.99	2.59 ± 0.8	6.04 ± 1.59
		92.38[a]	2.81	46.63	62.42	40.25	45.56	19.87
		92.8[b]	2.77					
		295.22	19.3					
		351.93	37.6					
		609.31	46.1					
		1120.29	15.1					
		1764.49	15.4					
		2204.21	5.08					
		2447.86	1.57					
4	Chowdhury et al. [8]	63.29	4.8	1.81 ± 0.46	1.47 ± 0.41	2.66 ± 0.64	2.23 ± 0.54	5.17 ± 1.05
		295.22	19.3	69.61	75.51	64.66	69.06	49.71
		351.93	37.6					
		609.31	46.1					
		1001.03	0.837					
		1120.29	15.1					
		1764.49	15.4					
5	Jankovic et al. [9]	63.29	4.8	1.79 ± 0.29	1.49 ± 0.27	2.47 ± 0.37	2.26 ± 0.35	3.79 ± 0.38
		351.93	37.6	97.76	104.7	92.31	95.13	67.28
		609.31	46.1					
		1001.03	0.837					
6	Song et al. [10]	63.29	4.8	2.16 ± 0.31	1.78 ± 0.28	2.99 ± 0.39	2.68 ± 0.37	5.26 ± 0.43
		92.38[a]	2.77	62.96	71.35	56.52	61.94	9.31
		295.22	19.3					
		351.93	37.6					
		609.31	46.1					

Table 7 continued

Sl. no.	Research group	Photopeaks (keV)	Intensity (%)	Average ^{238}U activity (Bq), relative standard deviation (RSD in %)				
				SB1	SB2	PU1	PU2	U-5dps
7	Mahur et al. [11]	295.22	19.3	1.42 ± 0.36	1.10 ± 0.31	2.18 ± 0.52	1.74 ± 0.41	5.12 ± 0.99
		351.93	37.6	52.82	52.73	58.71	52.29	55.08
		609.31	46.1					
		1001.03	0.837					
		1120.29	15.1					
		1764.49	15.4					
8	Santawamaitre et al. [12]	186.21	3.59	1.60 ± 0.71	1.10 ± 0.42	2.48 ± 1.01	1.92 ± 0.79	5.62 ± 1.63
		295.22	19.3	21.25	48.18	32.26	30.73	25.98
		351.93	37.6					
		609.31	46.1					
		1120.29	15.1					
		1238.11	5.79					
		1764.49	15.4					
		2204.21	5.08					
9	Gupta et al. [13], Boukhenfouf and Boucenna [14]	186.21	3.59	1.62 ± 0.38	1.34 ± 0.34	2.45 ± 0.55	2.01 ± 0.44	5.85 ± 1.02
		295.22	19.3	22.22	17.16	33.06	18.40	24.96
		351.93	37.6					
		609.31	46.1					
		1120.29	15.1					
		1764.49	15.4					
10	Aközcan [15], Alaamer [17], Kurnaz et al. [18]	186.21	3.59	1.38 ± 0.14	1.25 ± 0.13	1.99 ± 0.17	1.82 ± 0.17	4.71 ± 0.26
		351.93	37.6	13.76	16.8	15.58	12.64	5.52
		609.31	46.1					
11	Yang et al. [16]	186.21	3.59	1.35 ± 0.49	0.99 ± 0.33	2.06 ± 0.66	1.57 ± 0.53	4.71 ± 1.10
		351.93	37.6	51.11	71.72	57.77	57.96	54.99
		609.31	46.1					
		768.4	4.94					
		1120.29	15.1					
		1238.11	5.79					
		1764.49	15.4					

Table 7 continued

Sl. no.	Research group	Photopeaks (keV)	Intensity (%)	Average ^{238}U activity (Bq), relative standard deviation (RSD in %)				
				SB1	SB2	PU1	PU2	U-5dps
12	Ele Abiama et al. [20]	609.31	46.1	1.38 ± 0.48	0.88 ± 0.3	2.12 ± 0.64	1.54 ± 0.5	4.75 ± 1.08
		768.4	4.94	60.87	93.18	67.92	71.43	66.95
		1120.29	15.1					
		1238.11	5.79					
		1764.49	15.4					
13	Aytekin et al. [21], Alfonso et al. [22]	295.22	19.3	1.48 ± 0.11	1.15 ± 0.10	2.14 ± 0.14	1.86 ± 0.13	5.19 ± 0.27
		351.93	37.6	5.41	8.69	2.80	10.21	11.37
		609.31	46.1					
14	Chakraborty [30], Tchokossa et al. [35]	351.93	37.6	1.6 ± 0.27	1.28 ± 0.25	2.77 ± 0.45	2.14 ± 0.33	5.98 ± 0.81
		609.31	46.1	14.37	20.31	37.18	17.76	32.44
		1120.29	15.1					
15	Matiullah et al. [42]	295.22	19.3	1.57 ± 0.28	1.25 ± 0.26	2.59 ± 0.46	2.03 ± 0.34	5.95 ± 0.83
		351.93	37.6	12.74	17.6	35.13	18.72	26.72
		609.31	46.1					
		1120.29	15.1					
16	Pinto et al. [43]	609.31	46.1	1.87 ± 0.34	1.47 ± 0.3	2.93 ± 0.51	2.31 ± 0.4	6.68 ± 0.97
		1120.29	15.1	17.11	12.92	31.74	10.39	24.7
		1764.49	15.4					
17	El-Taher and Madkour [45]	351.93	37.6	1.72 ± 0.23	1.29 ± 0.18	2.34 ± 0.27	2.06 ± 0.25	5.53 ± 0.57
		609.31	46.1	25	21.7	12.39	12.62	21.16
		1764.49	15.4					
18	Murty and Karunakara [50]	351.93	37.6	1.75 ± 0.35	1.36 ± 0.30	2.75 ± 0.51	2.18 ± 0.4	6.21 ± 0.98
		609.31	46.1	20	19.11	30.54	14.68	26.57
		1120.29	15.1					
		1764.49	15.4					

[a] 92.38 keV photopeak was not observed, hence excluded from the calculation

[b] 92.38 and 92.8 keV photopeaks were not resolvable and not observed, hence excluded from the calculation

Table 8 Calculated activity of different daughter radionuclides of ^{232}Th under different photopeaks using 2 dps Th standard (STh)

Sl. no.	Radionuclide	Photopeak (keV)	Intensity (%)	SB1	SB2	PU1	PU2	Th-5 dps
1	^{208}Tl	277.35	6.31	0.36 ± 0.14	0.26 ± 0.17	1.05 ± 0.19	0.82 ± 0.2	3.91 ± 0.41
2	^{228}Ac	338.32	11.27	2.16 ± 0.10	1.57 ± 0.08	2.14 ± 0.1	2.26 ± 0.1	7.7 ± 0.28
3	^{228}Ac	463.00	4.4	1.25 ± 0.21	2.14 ± 0.27	1.99 ± 0.29	2.51 ± 0.32	9 ± 0.9
4	^{208}Tl	583.19	84.5	1.78 ± 0.07	1.65 ± 0.06	2.43 ± 0.08	2.14 ± 0.07	6.05 ± 0.17
5	^{212}Bi	727.33	6.58	2.02 ± 0.2	0.85 ± 0.13	1.92 ± 0.19	2.42 ± 0.22	5.43 ± 0.43
6	^{208}Tl	860.56	12.42	0.16 ± 0.16	1.56 ± 0.23	2.36 ± 0.29	2.32 ± 0.27	5.84 ± 0.54
7	^{228}Ac	911.20	25.8	1.82 ± 0.07	1.34 ± 0.06	2.25 ± 0.09	2.09 ± 0.08	6.97 ± 0.21
8	^{228}Ac	968.97	15.8	2.61 ± 0.2	2.81 ± 0.2	4.19 ± 0.28	2.32 ± 0.18	12.54 ± 0.77
9	^{208}Tl	2614.53	99	1.75 ± 0.09	1.09 ± 0.07	1.92 ± 0.11	2.19 ± 0.11	5.33 ± 0.24
Mean values of activities obtained from photopeaks having serial nos. 1, 2, 3, 4, 5, 6, 7, 8 and 9 (%RSD)				1.55 ± 0.45 (52.26)	1.47 ± 0.48 (49.66)	2.25 ± 0.6 (36.89)	2.12 ± 0.58 (23.58)	6.96 ± 1.5 (36.78)
Mean values of activities obtained from photopeaks having serial nos. 2, 3, 4, 5, 6, 7 and 9 (%RSD)				1.56 ± 0.37 (43.59)	1.46 ± 0.40 (28.77)	2.14 ± 0.49 (9.81)	2.28 ± 0.51 (6.58)	6.60 ± 1.22 (20.76)
Mean values of activities obtained from photopeaks having serial nos. 2, 4, 5, 6 and 9 (%RSD)				1.57 ± 0.29 (51.59)	1.34 ± 0.29 (26.12)	2.15 ± 0.39 (11.16)	2.27 ± 0.39 (4.84)	6.05 ± 0.81 (16.19)
Mean values of activities obtained from photopeaks having serial nos. 2,4,5,6,7 and 9 (%RSD)				1.62 ± 0.31 (45.06)	1.34 ± 0.29 (23.13)	2.17 ± 0.39 (10.13)	2.24 ± 0.39 (5.35)	6.20 ± 0.83 (15.32)
Mean values of activities obtained from photopeaks having serial nos. 4,7 and 9 (%RSD)				1.78 ± 0.14 (1.68)	1.36 ± 0.11 (20.59)	2.20 ± 0.16 (11.82)	2.14 ± 0.16 (2.34)	6.09 ± 0.36 (14.28)
Mean values of activities obtained from photopeaks having serial nos. 2, 4 and 7 (%RSD)				1.92 ± 0.14 (10.94)	1.52 ± 0.12 (10.53)	2.27 ± 0.16 (6.17)	2.17 ± 0.15 (4.15)	6.91 ± 0.39 (11.87)

%RSD value in parenthesis

Table 9 Photopeaks of ^{232}Th series suitable for quantitative analysis of ^{232}Th

Sl. no.	Daughter	Energy (keV)	Intensity (%)
1	^{228}Ac	338.32	11.27
2	^{228}Ac	463.00	4.4
3	^{208}Tl	583.19	84.5
4	^{212}Bi	727.33	6.58
5	^{208}Tl	860.56	12.42
6	^{228}Ac	911.20	25.8
7	^{228}Ac	968.97	15.8
8	^{208}Tl	2614.53	99

standard spectra. Analysis of the obtained gamma-spectra was done using GENIE 2K software, also procured from CANBERRA.

Result and discussion

In principle, the activities of ^{226}Ra, ^{214}Pb and ^{214}Bi, (all of them are member of ^{238}U series) should be same as they are in secular equilibrium. But in practice slight difference is always observed between the measured activities of different isotopes or even in between the different peaks of same isotope. We have measured activities of ^{238}U for all four samples SB1, SB2, PU1, PU2 and 5 dps test sample (SU) using 2 dps standard for all the photopeaks listed in Table 3 and tabulated the activity values in Table 5. It is clear from Table 5 that still some of the photopeaks do not qualify for quantitative measurement of ^{238}U. These photopeaks are 131.3 keV (^{234}Pa), 152.7 keV (^{234}Pa), 733.4 keV (^{234}Pa), 768.4 keV (^{214}Bi), 831.5 keV (^{234}Pa) and 946 keV (^{234}Pa). These peaks give either too low or too high value, as compared to other photopeaks. There may be multiple reasons for the disqualification of these photopeaks, such as low intensity and overlapping with other low abundance nearby photopeaks, location of the peak at the Compton edge of other photopeak, etc. Therefore we have not considered these photopeaks suitable for quantitative analysis of U content from natural samples and deleted in the next stage of selection of photopeaks. In Table 6 we have listed photopeaks of ^{238}U series still suitable for analysis of uranium content. Now the pertinent question is whether all the photopeaks listed in Table 6 have same merit? More elaborately, whether one can take average of all the photopeaks listed in Table 6 to report uranium content of the sample or one can take arbitrarily average of activities obtained from few of these photopeaks? To answer these questions, we go back to bottom part of Table 5, wherein we have calculated the activity of U in samples SB1, SB2, PU1, PU2 by taking average of activities under various combinations of photopeaks and also calculated relative standard deviation ($\text{RSD} = \frac{\text{standard deviation}}{\text{mean value}} \times 100$) of the activities obtained in different photopeaks. The RSD values varied from 2.8 % to as high as 76.8 %. The RSD value need to be as low as possible to get the best estimate using set of good photopeak combinations. Table 5 suggests that average of activity calculated from 295.22, 351.93 and 609.3 keV gives minimum RSD value and therefore can be used to report uranium content of natural samples in a statistically reliable manner.

To further validate our result, we have calculated the activity of our four test samples with different combinations of photopeaks as taken by different researchers in Table 7. Only those results have been taken into account where the researchers selected three or more photopeaks. In some cases the RSD was even close to 100 %. For example, the RSD was $\sim$100 % for all the samples, when photopeaks were selected as per Jankovic et al. [9] (entry no 5 in Table 7). This is because along with two good peaks they also selected two very low intensity peaks, 63.29 and 1001.03 keV. Only two groups of researchers, Aytekin et al. [21], and Alfonoso et al. [22] selected the photopeaks as proposed by us (295.22, 351.93 and 609.3 keV). However, these authors never mentioned the reason for choosing such photopeaks and therefore their selection can be considered "accidentally right selection." The RSD value was found to be minimal for these photopeaks compared to any other entry in the table, which corroborates and strongly validates our recommended approach.

The same approach has been resorted to for the quantification of ^{232}Th in all four samples by measuring activity under different photopeaks listed in Table 4. In all measurements 2 dps ^{232}Th standard was used. Also a 5 dps ^{232}Th (STh) sample was taken as known strength. All such results have been tabulated in Table 8. The RSD values between various sets of photopeaks are closer in ^{232}Th series when compared to that of ^{238}U series. The 277.35 keV photopeak from ^{208}Tl gave very low activity for all four samples. However, all other photopeaks provided more or less acceptable results. Therefore, using the same analogy as that of uranium, we have listed acceptable photopeaks of ^{232}Th series in Table 9, which indicates 8 numbers of photopeaks might be suitable for ^{232}Th analysis. However, the same questions arise again. Whether all of these photopeaks have same merit? Whether one can choose any number of photopeaks from Table 9, and report the mean as ^{232}Th content in the sample? To answer this question, we have shown few combinations at the bottom of Table 8, with RSD for each combination.

Table 10 Activity of ^{232}Th obtained for four test samples with different sets of photopeaks as taken by different researchers (see Table 1)

Sl. no.	Research group	Photopeaks (keV)	Intensity (%)	Average ^{232}Th activity (Bq), relative standard deviation (RSD in %)				
				SB1	SB2	PU1	PU2	Th-5dps
1	Wang et al. [6]	338.32	11.27	2.19 ± 0.24	1.91 ± 0.23	2.86 ± 0.31	2.22 ± 0.23	9.07 ± 0.84
	Matiullah and Malik [42]	911.20	25.8	17.81	41.36	40.56	5.41	33.41
		968.97	15.8					
2	Al-Sharkawy et al. [7]	583.19	6.58	2.08 ± 3.55	2.14 ± 5.05	2.76 ± 3.89	2.92 ± 4.53	7.22 ± 7.35
		727.33	84.5	86.06	127.10	59.42	64.38	56.23
		860.56	12.42					
		911.20	25.8					
		964.76[c]	4.99					
		968.97	15.8					
		1588.19	3.22					
		1620.7	1.49					
		2614.53	99					
3	Chowdhury et al. [8]	238.63	43.3	2.08 ± 0.32	1.66 ± 0.27	2.57 ± 0.38	2.25 ± 0.32	7.55 ± 0.97
		338.32	11.27	13.94	39.16	31.91	5.33	34.04
		583.19	6.58					
		727.33	84.5					
		911.20	25.8					
		968.97	15.8					
4	Song et al. [10]	238.63	43.3	1.89 ± 0.11	1.58 ± 0.09	2.38 ± 0.13	2.17 ± 0.12	6.54 ± 0.29
		583.19	6.58	7.94	13.92	5.04	4.15	7.03
		911.20	25.8					
5	Mahur et al. [11], Gupta et al. [13]	238.63	43.3	1.99 ± 0.37	1.75 ± 0.38	2.49 ± 0.47	2.31 ± 0.45	8.04 ± 1.31
		338.32	11.27	22.61	38.28	34.54	6.06	31.09
		463.00	4.4					
		727.33	6.58					
		911.20	25.8					
		968.97	15.8					
6	Santawamaitre et al. [12]	238.63	43.3	2.27 ± 3.51	2.24 ± 5.04	2.64 ± 3.68	2.65 ± 4.32	7.43 ± 7.22
		300.09	3.28	72.69	119.64	67.42	78.49	38.89
		338.32	11.27					
		583.19	84.5					
		727.33	6.58					
		911.20	25.8					
		968.97	15.8					
		1620.5	1.49					
		2614.53						
7	Boukhenfouf and Boucenna [14]	238.63	43.3	1.51 ± 0.29	1.53 ± 0.33	2.38 ± 1.21	2.21 ± 1.28	6.53 ± 1.02
		338.32	11.27	67.55	53.59	41.59	4.98	56.35
		583.19	84.5					
		860.56	12.42					
		911.20	25.8					
		964.76[c]	4.99					
		968.97	15.8					

Table 10 continued

Sl. no.	Research group	Photopeaks (keV)	Intensity (%)	Average ^{232}Th activity (Bq), relative standard deviation (RSD in %)				
				SB1	SB2	PU1	PU2	Th-5dps
8	Yang et al. [16], Ele Abiama et al. [20]	238.63	43.3	1.74 ± 0.25	1.52 ± 0.24	2.25 ± 0.32	1.85 ± 0.24	6.64 ± 0.86
		338.32	11.27	51.72	59.21	59.55	49.19	60.39
		583.19	84.5					
		911.20	25.8					
		968.97	15.8					
		974.2[d]	0.050					
9	Aytekin et al. [41]	338.32	11.27	1.92 ± 0.14	1.52 ± 0.12	2.27 ± 0.16	2.17 ± 0.15	6.91 ± 0.39
		583.19	84.5	10.94	10.53	6.17	4.15	11.87
		911.20	25.8					
10	Rajeshwari et al. [41]	238.63	43.3	1.85 ± 0.14	1.46 ± 0.12	2.27 ± 0.17	2.18 ± 0.16	6.22 ± 0.37
		583.19	84.5	7.57	20.55	11.01	3.67	12.06
		911.20	25.8					
		2614.53	99					
11	Al-Jundi et al. [49]	238.63	43.3	2.07 ± 0.23	1.89 ± 0.22	.84 ± 0.31	2.21 ± 0.22	8.04 ± 0.82
		583.19	6.58	18.36	33.86	232.04	4.98	37.56
		911.20	25.8					
		968.97	15.8					
12	Murty and Karunakara [50]	583.19	6.58	1.78 ± 0.14	1.36 ± 0.11	2.19 ± 0.16	2.14 ± 0.16	6.09 ± 0.36
		911.20	25.8	1.68	20.59	11.87	2.34	14.28
		2614.53	99					

[c] 964.76 keV photopeak is present on the shoulder of 968.97 keV photopeak, therefore couldnot be resolved and not included in calculation

[d] 974.2 keV photopeak is present on the trail of 968.97 photopeak, therefore counldnot be resolved and not included in calculation

Table 11 Final recommended photopeaks of ^{238}U and ^{232}Th series suitable for low-level radioactivity measurement

Sl. no.	Daughter	Energy (keV)	Intensity (%)
Photopeaks of ^{238}U			
1	^{214}Pb	295.22	19.3
2	^{214}Pb	351.93	37.6
3	^{214}Bi	609.31	46.1
Photopeaks of ^{232}Th			
1	^{228}Ac	338.32	11.27
2	^{208}Tl	583.19	84.5
3	^{228}Ac	911.20	25.8

The minimum and maximum RSD amongst different combination was 1.7 and 52.6 % respectively. However, average of activity obtained from 338.32, 583.19 and 911.20 keV yielded minimum RSD value, and hence recommended as the best combination of photo-peaks to measure ^{232}Th.

Again to validate our approach for ^{232}Th series, we have tabulated the activity of four test samples SB1, SB2, PU1, PU2 with different combinations of photopeaks of ^{232}Th series as taken by different researchers in Table 10. The results are more consistent vis-a-vis the U series, but as high as 127 % RSD was observed in particular combination of photopeaks. Again, brilliantly Aytekin et al. [21] reported natural radioactivity in Black sea region of Turkey using the photopeaks as proposed by us (338.32, 583.19, and 911.20 keV), and have the lowest RSD compared to any other entry in the table, further corroborating and validating our proposed approach for measurement of low level environmental radioactivity.

In Table 11, we list our final proposed recommendations related to the appropriate selection of photopeaks from ^{238}U series and ^{232}Th series for carrying out statistically reliable quantitative measurement of NORMs like ^{238}U and ^{232}Th. It should however be kept in mind that this recommendation should not be treated as the ultimate one as the role of detector used, sample size, counting time etc. still needs to be further investigated. However, the above discussion advocates to take at least three photopeaks for quantitative measurement of low-level ^{238}U/^{232}Th, especially in natural samples and the best combination of photopeaks is that one where minimal RSD value is obtained.

Conclusion

Measurement of naturally occurring radionuclide materials (NORMs) is becoming increasingly important in the present world scenario. In conclusion it can be stated that the present work is the first attempt to systematically investigate the contribution of photopeaks and has come out with a prescription to get a better and statistically reliable estimate of activity/concentration of radionuclides while carrying out low-level radioactivity measurements. It would be interesting to extend the work further to understand the role of parameters like nature of detector, sample size, counting time etc. in the study of environmental radioactivity. This paper also states that RSD between different photopeaks is one of the important criteria to impose restrictions on the arbitrariness on choice of photopeaks for quantitative measurement of low-level ^{238}U or ^{232}Th in natural samples.

Acknowledgments This work is part of DAE-SINP 12 five-year plan project TULIP (Trace, ULtratrace Analysis and Isotope Production). One of the authors, NN would like to thank University Grants Commission (UGC) for providing the necessary fellowship. AS would like to place in record the financial support given to him by UGC-DAE and Alexander von Humboldt Foundation, Bonn, Germany to enable him to participate in NORM related studies in India and Germany. The financial support used for collection of sample given under DST PURSE program to Panjab University, Chandigarh is also thankfully acknowledged. PC would like to acknowledge Scientific and Engineering Research Board (SERB), Department of Science and Technology, Govt. of India for financial support.

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