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Radioactivity concentrations and dose assessments of therapeutic peloids from some Turkish spas

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ABSTRACT: The activity concentrations of natural radionuclides in peloids were studied to assess the radiologic hazard from 18 Turkish spas. The peloids are mainly used for therapeutic treatments, rheumatic diseases and aesthetic purposes. The concentrations of the natural radionuclides ^{226}Ra, ^{232}Th, ^{40}K and ^{137}Cs were determined with a gamma ray spectrometer using a HPGe detector. The average activity concentrations of ^{226}Ra, ^{232}Th, ^{40}K, and ^{137}Cs in the peloids studied were 110.69, 71.52, 576.48 and 0.447 Bq/kg, respectively. The radium equivalent activities in the peloid samples ranged from 63.3 to 766.77 Bq/kg. The absorbed dose rate (D_{out}) varied between 37.52 and 330.67 nGy/h and most of the observed spa doses are greater than the worldwide recommended values. The annual effective dose values range from 0.26 to 2.78 μSv/y. The annual gonadal dose equivalents of the samples vary from 224.07 to 2283.55 with a mean of 821.99 μSv/y.

KEYWORDS: dose, peloids, radionuclides, spa, Turkey.

In situ thermal muds are being used for therapeutic, aesthetic and pharmaceutical purposes, outdoors and/or indoors in 30 spa centers in Turkey (Karakaya *et al*., 2010). Activity concentrations of certain radionuclides, together with the mineralogical, chemical and some physical properties of these peloids, were investigated in this study. The peloids studied, which were sourced from areas near the spas, were generally alluvium or marsh soils and, consequently, they are complex mixtures of different compounds from the source areas and from matured thermal waters. Radionuclides were the primary source of radiation in the samples; with the amount of radiation, or dose, being proportional to the concentration. In a typical preparation of the peloids, a mud bath tub is filled with 50 cm of thick clay and thermal water is added to make a mud bath. The patients lie in a bathtub and a ~20 cm thick peloid cover is placed on their bodies while they use the mud bath for ~10–15 min each day for 15 days. After a period of time, the tub is drained completely. In addition, the patients use the thermal waters in a bath or pool, kept at ~40–45ºC. The mud baths, masks and cataplasms are used to treat nearly all types of rheumatism as well as to cleanse and beautify the skin. Peloids are also used to treat neuralgia, neuritis, polyneuritis, fractures, dislocations and polio disease in women (Legido *et al.*, 2007; Viseras *et al.*, 2007; Williams & Haydel, 2010; Quintela *et al.*, 2012 and references therein). Some patients cover themselves in mud and wait

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for it to dry in the sun before they are cleaned in the pool water.

Exposure to radioactivity is commonly considered objectionable at all exposure levels, even though no harmful effects have been reported at very low levels of radioactivity (UNSCEAR, 1993). Natural radioactivity in soils is caused mainly by the $^{238}U/^{232}Th$ decay series and natural ^{40}K (UNSCEAR, 2000).

Pelotherapy is famous as an extremely effective therapy worldwide and is a commonly used method for the cure of numerous syndromes, e.g. skin, rheumatic and joint diseases (Kikouama & Balde, 2010 and references therein). The patients are irradiated by mud, water and air in spas because they contain natural radioisotopes. Radon gas (^{222}Rn) in mud, water and air constitutes a major part of natural radioactivity. The radon gas undergoes alpha decay with energy 5.49 MeV with a half-life of 3.82 days. Therefore, the decay products, which emit α particles themselves, have relatively short lifetimes; for example, ^{218}Po and ^{222}Rn, can be a threat to human life at high concentrations. Radon gas in particular, can be inhaled from mud, air or water during ventilation (Appleton, 2005). During the spa treatments, natural radioactivity can be beneficial to patients, but negative side effects may occur at greater doses. Hence, measurement of gamma radiation levels provides valuable information about dose levels. The radioactivity levels in soils and water in Turkey have been studied previously (Karahan & Bayülken, 2000; Karakelle *et al.*, 2002; Erees *et al.*, 2006; Merdanoğlu & Altınsoy, 2006; Kam & Bozkurt, 2007; Kılıç *et al.*, 2008: Kurnaz *et al.*, 2007; Değerlier *et al.*, 2008; Bozkurt *et al.*, 2007; Taşkın *et al.*, 2009; Yüce *et al.*, 2009; Yüce and Gasparon, 2013) and some of the mineralogical, chemical and physical properties of the thermal muds have been investigated (Karakaya *et al.* 2010). Nevertheless, there are very few studies evaluating the radioactivity of thermal muds or clay minerals (Gold *et al.*, 1990; Doretti *et al.*, 1992; Manic *et al.*, 2006; Silva *et al.*, 2011) and the radioactivity of the thermal muds in Turkey have not been assessed at all.

The main aims of this study are to determine the radioactivity levels of peloid samples, for estimating doses for patients seeking treatment for musculoskeletal disorders, or aesthetic purposes, in spas and to compare spa, soil and rock results from different parts of Turkey and internationally.

MATERIALS AND METHODS

Eighteen matured peloid samples were taken from 16 of the 20 main spa centres (Fig. 1). After sieving, the peloids were treated with thermal spa water for 24 h. Subsequently, they were homogenized, dried and pulverized for 5 min in a porcelain ball mill for mineralogical and chemical analyses. The mineralogical analyses of the samples were conducted on random, oriented samples (total fraction) (<2 μm clay-sized fractions) using X-ray diffraction (XRD) with CuKα radiation from 2−70º2θ at a scanning speed of 1º2θ/min. The mineral proportions were determined from chemical analysis combined with the powder XRD patterns, according to the external standard method developed by Gündoğdu (1982), (Table 1). The samples

FIG. 1. Locations of the main peloid using spas.

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TABLE 1. Mineralogical composition, K_2O (wt.%) and Cs, Th and U contents (ppm) of the 18 peloid samples examined. The values in parentheses indicate the proportions (%) of individual minerals in the peloids.

	Mineralogy and mineral contents (wt.%)	K_2O	Th	U	Cs
P-1	Sme(60)+Cal(12)+Ms/Bio(10)+Fsp(8)+Qz(5)+Kln(3)+Dol (2)	3.01	29.1	7.20	20.8
P-2	Sme(65)+Cal(13)+Dol(8)+Ms/Bio(6)+Qz(4)+Kln(4)	1.08	5.60	1.60	7.70
P-5	Sme(38)+Ms/Bio(30)+Cal(13)+Fsp(9)+Qz(5)+Kln(3)+Dol(1)+Gp(1)	1.43	7.10	2.90	42.50
P-5/1	Cal(34)+Ms/Bio(32)+Sme(18)+Fsp(5)+Qz(4)+Kln(4)+Dol(2)+Gp(1)	2.34	7.60	3.40	152.10
P-6	Sme(36)+Ms/Bio(27)+Cal(22)+Qz(5)+Dol(4)+Fsp(3)+Kln(3)	0.83	4.20	2.10	40.50
P-7	Sme(26)+Dol(18)+Cal(15)+Py(12)+Srp(10)+Kln(8)+Qz(7)+Gp(4)	0.95	5.00	1.50	15.90
P-8	Sme(42)+Srp(18)+Cal(9)+Ms/Bio(8)+Dol(6)+Kln(6)+Qz(5)+Fsp(4)+Hl(2)	1.05	4.40	1.10	3.50
P-9	Sme(66)+Hl(11)+Cal(8)+Fsp(7)+Qz(5)	2.34	11.40	6.70	12.40
P-11	Sme(52)+Ms/Bio(21)+Fsp(9)+Qz(8)+Dol(6)+Kln(4)	4.79	19.60	4.90	43.80
P-12	Sme(57)+Ms/Bio(15)+Cal(11)+Fsp(8)+Qz(4)+Kln(3)+Gp(2)	2.07	11.70	1.90	121.7
P-14	Sme(32)+Ms/Bio(22)+Cal(17)+Fsp(11)+Qz(7)+Kln(4)+Py(4)+Hl(2)	2.50	10.90	2.50	21.10
P-15	Sme(36)+Ms/Bio(26)+Cal(12)+Kln(10)+Dol (7)+Qz(4)+Fsp(3)+Gp(2)	1.69	11.10	1.80	139.50
P16	Sme(73)+Fsp(6)+Qz(6)+Kln(4)+Gp(4)+Py(4)+Cal(3)	2.24	12.60	1.80	289.10
P-17	Sme(60)+Cal(15)+Fsp(12)+Kln(4)+Qz(4)+Py(4)	1.74	6.60	1.60	3.70
P-18	Sme(52)+Cal(40)+Fsp(3)+Qz(3)+Do(2)	1.37	5.80	1.30	10.20
P19	Man(90)+Spe(10)	1.41	0.70	4.80	11.70
P-20	Ms/Bio(37)+Cal(18)+Sme(7)+Fsp(26)+Qz(12)	2.66	25.40	3.50	48.60
P-20/1	Ms/Bio(38)+Cal(17)+Sme(11)+Fsp(21)+Qz(13)	3.18	27.10	3.90	52.70
MDL		0.01	0.20	0.10	0.50

MDL: Method Detection Limit, Bio: Biotite, Cal: Calcite, Dol: Dolomite, Fsp: feldspar, Gp: Gypsum, Hl: Halite, Kln: Kaolinite, Man: magnesite; Ms: Muscovite, Qz: Quartz, Sme: Smectite, Spe: sepiolite, Srp: serpentine, Py: pyrite (abbreviations from Whitney & Evans, 2010).

were mounted according to Temel and Gündoğdu (1996) and Gündoğdu (1982) and the characteristic peak intensities (I) of the minerals were normalized to the (104) reflection of dolomite. The sensitivity ratios of the minerals relative to dolomite (K) were determined for each mineral (including clay minerals with peaks between 19 and 20º2θ) by weight in a 1:1 dolomite/mineral mixture, as follows: $K = I_{\text{dolomite}}/I_{\text{mineral}}$. The mineral percentages were calculated using equation 1:

$$\% \text{ mineral a} = (100 \times K_a \times I_a)/(K_a \times I_a + K_b \times I_b + ...K_n \times I_n) \quad (1)$$

The accuracy of the method is better than ±15%.

The total abundances of the major oxides and the minor elements of the peloids were determined at ACME Laboratories (Vancouver, British Columbia, Canada) by inductively coupled plasma optical emission spectrometry and mass spectrometry (Spectro ICP-OES and Perkin Elmer ELAN 9000 ICP-MS, USA, respectively). Samples (0.1 g) were fused with Li metaborate/tetraborate (1 g) and digested with nitric acid.

The peloid samples were sieved, the >1 mm fractions were removed and the <1 mm particles were suspended in water, the <2 μm fractions separated by sedimentation, centrifuged, dried and then the dried powders were kept in polyethylene bottles. The bottles were closed to prevent the escape of gas and stored for 1 month to allow the radioactive elements to reach equilibrium with their decay products. The activity concentrations of the radionuclides (^{226}Ra, ^{232}Th, ^{40}K and ^{137}Cs) were determined using an n-type HPGe (high purity germanium) detector with energy resolution 2.1 keV at 1.33 MeV, energy rate range from 40 keV to 10 MeV and relative efficiency 50%. Detector calibrations were performed with multi-nuclide standard sources, which had an activity of 1.365 μCi (^{57}Co, ^{60}Co, ^{88}Y, ^{109}Cd, ^{139}Ce, ^{137}Cs, ^{203}Hg, ^{210}Pb and ^{241}Am). Sample spectra were taken at 54,000 s (15 h) intervals. Spectra analyses were performed using *GammaVision* (ORTEC) software and the activity concentrations of 5 radionuclides were produced. To decrease the background effect, a 10 cm thick lead covering, lined with 2 mm thick copper and cadmium foils, protected the detector. The energy and efficiency calibrations were made with a certified standard gel source with a similar density to the measured samples. The energy and efficiency calibration curves are shown in Fig. 2.

The sample activity (Bq/kg) for a given radionuclide can be calculated using equation 2:

$$A = \frac{N_S - N_B}{\varepsilon \times P_\gamma \times t} \times \frac{1}{m} \quad (2)$$

where A, N_S, N_B, ε, Pγ, t and m are the activity, the counts of the sample (Bq/kg), the background count, the absolute efficiency, the branching ratio, the counting live time (s) and the mass of the sample (kg or L (density ~1 kg L^{-1})), respectively, for certain radionuclides in a gamma ray with energy E. For the determination of specific activities, the daughter radionuclide gamma ray lines of 351.9 keV (^{214}Pb) and 609.3 (^{214}Bi) for ^{226}Ra, 911.2 keV (^{228}Ac) for ^{232}Th and the 662 keV

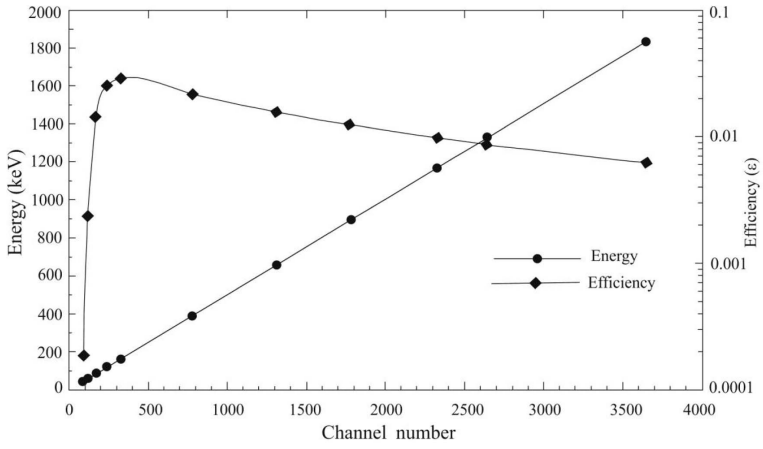

FIG. 2. Energy and efficiency calibrations of the HPGe detector.

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line for ^{137}Cs, were used. Because ^{40}K does not belong to a decay series as it has a stable daughter product, its characteristic gamma peak at 1460.8 keV was used, with a branching ratio of 11% of the energy. The concentration of ^{40}K is related directly to the total potassium content and the amounts of ^{226}Ra and ^{232}Th were determined using the peaks of the decay products in equilibrium with their parent nuclides. The minimum detectable activity (MDA) was calculated using equation 3:

$$MDA = (2.71 + 4.65\sqrt{N_B})/(\varepsilon \times P\gamma \times t) \quad (3)$$

RESULTS AND DISCUSSION

Most peloid compositions are generally homogenous and composed mainly of Ca-montmorillonite, illite and illite-smectite, with smaller proportions of quartz and feldspar, some calcite, dolomite and amorphous silica, with rare traces of kaolinite, halite, serpentine and gypsum (Table 1). The exception is sample P-19, which contains 90% magnesite and 10% sepiolite. The proportion of the clay minerals was generally between 50% and 60%, and the most abundant clay mineral was Ca-montmorillonite and the concentrations of K_2O and some of the trace elements in the peloids were mostly similar (Karakaya *et al.*, 2013). A moderate positive correlation was observed between K_2O, Th and U concentrations. The activity concentrations of the radionuclides (^{226}Ra, ^{232}Th, ^{40}K, and ^{137}Cs) were determined and were compared with the world soil average (UNSCEAR, 2008; Fig. 3, Table 2). Sample P-14 displayed the maximum activity concentration and the sample P-15 the lowest activity/concentration for ^{226}Ra (392.59 Bq/kg and 9.42 Bq/kg respectively, Table 2). The average ^{226}Ra activity concentration of the samples is 110.69 Bq/kg, with most of the concentrations being greater than the UNSCEAR average (37 Bq/kg). The greatest activity concentration of ^{232}Th was measured for sample P-2 (445.36 Bq/kg) and the smallest was found in sample P-19 (2.61 Bq/kg). The average ^{232}Th activity concentration of the samples was 71.52 Bq/kg, with several samples having greater concentrations than the UNSCEAR average (33 Bq/kg).

Potassium is an important constituent of rock-forming minerals including some clay minerals such as illite. It may also be present as an exchangeable cation in smectites and vermiculites. The abundance of potassium in the Earth's crust is 2.6%. ^{40}K is a naturally occurring radioactive isotope of potassium with half-life ~1.26×10^9 y and an abundance of 0.00117%. Activity concentrations of ^{40}K were

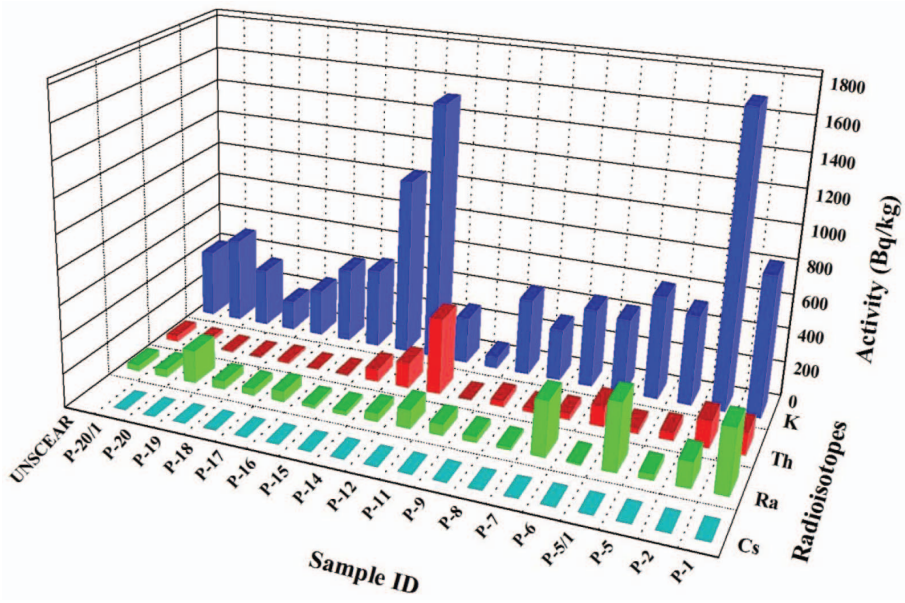

FIG. 3. Activity concentrations of ^{226}Ra, ^{232}Th, ^{40}K and ^{137}Cs in the peloid samples.

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TABLE 2. Activity concentrations (Bq/kg) of the radionuclides in the peloids examined.

Sample number	^{226}Ra		^{232}Th	^{40}K	^{137}Cs
	351.9 keV	609.3 keV	911.2 keV	1461.8 keV	662 keV
P-1	374.44±4.42	373.07±4.66	128.16±6.03	818.32±24.92	MDA
P-2	95.56±2.50	122.64±2.91	445.36±11.25	270.20±14.32	0.559±0.307
P-5	25.67±1.54	17.92±1.26	3.96±1.06	465.42±18.79	0.419±0.140
P-5/1	51.57±1.97	73.98±2.35	2.79±0.89	438.38±18.24	MDA
P-6	42.24±1.51	39.73±1.62	14.78±2.05	277.12±14.50	0.425±0.159
P-7	51.09±1.69	49.31±1.74	4.68±1.15	174.76±11.51	0.459±0.162
P-8	181.36±3.30	212.39±3.75	8.13±1.52	344.68±16.17	0.259±0.172
P-9	54.40±1.76	37.71±1.71	22.55±2.53	506.63±19.61	1.265±0.309
P-11	171.62±3.03	122.07±3.12	153.65±6.61	1698.60±35.90	MDA
P-12	46.81±1.70	27.23±1.48	46.66±3.64	522.07±19.90	0.199±0.227
P-14	383.54±4.59	401.64±4.84	27.81±2.81	599.96±21.33	0.473±0.184
P-15	9.94±1.00	8.91±1.30	119.02±5.82	430.73±18.08	MDA
P-16	322.40±4.54	317.92±4.73	34.99±3.15	456.16±18.60	MDA
P-17	17.05±0.95	15.58±0.99	18.88±2.32	301.71±15.13	MDA
P-18	32.07±1.45	33.47±1.58	31.85±3.01	448.77±18.45	0.486±0.213
P-19	56.86±1.75	74.06±2.08	2.61±0.86	64.82±7.01	MDA
P-20	56.23±2.29	39.90±2.18	147.95±6.48	1516.37±33.92	0.242±0.202
P-20/1	20.12±1.14	24.36±1.33	73.48±4.57	1041.93±28.12	0.136±0.109
MDA	0.034	0.032	0.025	0.057	0.005
Minimum	9. 94	8.91	2.61	64.82	MDA
Maximum	383.54	401.64	445.36	1698.60	1.265
Average	110.72	110.66	71.52	576.48	0.447
UNSCEAR (2008)	37	37	33	400	

reported in some studies as 670 Bq/kg (clay) (Somlai *et. al.*, 2008), 112 Bq/kg (red mud) (Akinci & Artir, 2008), 110.3 Bq/kg (mud) (Saad & Al-Azmi, 2002), 618−548 Bq/kg (sand) (El-Arabi, 2005) and 643 Bq/kg (soil) (Kurnaz *et. al.*, 2007). The activity concentrations measured in those studies are due to the presence of potassium-rich minerals in the samples such as orthoclase and micas. Potassium activity values for soil samples are given as 400 Bq/kg (UNSCEAR, 2008). In the present study, the highest activity concentration of ^{40}K was determined for sample P-11 (1698.60 Bq/kg), while the lowest activity concentration was observed in sample P-19 (64.82 Bq/kg). The mean ^{40}K activity concentration of the samples is 576.48 Bq/kg, with most samples having concentrations greater than the UNSCEAR average (400 Bq/kg) (Table 2).

Being a product of radioactive fallout, ^{137}Cs is not present naturally in soils and sediments (Kurnaz 2007). The ^{137}Cs activity varies from above the MDA value to 1.265 Bq/kg in this study with a mean of 0.447 Bq/kg (for detectable activities). The highest ^{137}Cs activity concentration was measured in the P-9 sample. A threshold value for Cs activity is not available in the literature for soils and peloids. However, there is background work on the activity concentrations of the remaining radionuclides in soils both in Turkey and internationally. The activity concentrations obtained for the radionuclides excluding Cs are greater than other Turkish soils and global soil averages (UNSCEAR, 2008) (Table 3).

The activity concentrations of radionuclides in the peloids are generally different from the concentrations in naturally occurring soils from Turkey and some Mediterranean countries. Most concentrations in the peloids are greater than those of the soils (Table 3). Natural radionuclides may cause many health problems after long-term exposure, e.g. chronic lung diseases, leukemia and bone, kidney and pancreas cancers (Taşkın *et al.*, 2009). The activity concentrations of ^{226}Ra, ^{232}Th and ^{40}K in the present study are comparable to those of the pharmaceutical and cosmetic clays (Silva *et al.*, 2011; Papadopoulos *et al.*, 2014). The concentrations of the radionuclides vary widely and the maximum values are greater than those

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TABLE 3. Comparison of the activity concentrations of the radionuclides in peloids with those in soils from Turkey and elsewhere.

Location	^{226}Ra	^{232}Th	^{40}K	^{137}Cs	Reference
İstanbul, Turkey		35±7	322±87		Karahan & Bayülken (2000)
Eskişehir,Turkey		306	1265		Örgün *et al*. (2005)
Çanakkale, Turkey		192	1207		Merdanoğlu & Altınsoy (2006)
Rize, Turkey	–	42.0	653.0	85.0	Kurnaz *et al*. (2007)
Thrace, Turkey	32.0	23.0	1319.0	21.0	Aközcan *et al*. (2014)
Amman, Jordan		28.8	501.3		Ahmad *et al*. (1997)
Karak, Jordan		27.2	410.2		
Egypt		1.23–32.15	17–99		Yousef *et al*. (2007)
	16.7±2.7	19.4±5.0	262±82		Saleh *et al*. (2007)
Malaysia		14.9–301	–		Ramli *et al*. (2005)
Kohistan, Pakistan	42.11±1.11	43.27±1.72	418.27±15.69	9.4>±0.64	Khan *et al*. (2011)
Punjab, Pakistan		50–103	374–1082		Khan *et al*. (2012)
Cyprus	7.1±8.6	5.0±7.1	105±95		Tzortzis *et al*. (2004)
India		104±77	217±145		Selvasekarapandian *et al*. (2000)
Vojvodina, Serbia		53±8	554±92		Bikit *et al*. (2005)
Stromboli		68±1	754±9		Brai *et al*. (2002)
This study	55–425	3–445	65–1699	0.02–65.06	
World average	37	33	400		UNSCEAR (2008)

TABLE 4. Comparison of the activity concentrations (Bq/kg) of the radionuclides in the peloids of the present study with those from some spa and cosmetic clays.

Activity Bq/kg	Gold *et al.* (1990)	Doretti *et al.* (1992)	Manic *et al.* (2006)	Silva *et al.* (2011)*	Papadopoulos *et al.* (2014)*	This study
^{226}Ra	–	–	259	32–54	58–168	9.9–384
^{232}Th	10	33	12.9	40–111	36–80	2.6–445
^{40}K	260	443	219	76–1146	211–996	65–1698
^{137}Cs	–	–	0.5	–	–	MDA–1.25

* cosmetic clays

observed in spas internationally (Tables 3, 4). Therefore, according to their radioactivity many of the peloids examined may be classified as radioactive muds (Manic *et al.*, 2006; Silva *et al.*, 2011). However, this is not confirmed when the radiological hazard point of view is considered (see below).

Nevertheless, the peloids collected were formed *in situ* near each spa and the geological characteristics of each area differ. Therefore, different mineralogical and chemical compositions of the source rocks and soils, as well as the chemical and physical properties of the thermal waters, might cause the radionuclide concentrations to vary markedly.

The radium equivalent activity (Ra_{eq}), absorbed dose rate (D_{out}), the external annual effective dose (AED), the annual gonadal dose equivalent (AGDE) and the lifetime cancer risk (LCR) for the peloids were calculated (Table 5). Radium equivalent activity (Ra_{eq}) is a commonly used risk index and was calculated using the equation of Krieger (1981) and Beretka & Matthew (1985):

$$Ra_{eq} = A_{Ra} + 1.43A_{Th} + 0.077A_K \quad (4)$$

where A_{Ra}, A_{Th} and A_K are the activity concentrations of ^{226}Ra, ^{232}Th and ^{40}K, respectively, in the peloids. The radium equivalent activity (Ra_{eq}) weights the activities of ^{226}Ra, ^{232}Th and ^{40}K and, in this study, ranges from 63.3 to 766.77 Bq/kg, with a mean of 257.35 Bq/kg (Table 5). Most Ra_{eq} values are below the guideline value of 370 Bq/kg (Alaamer, 2008 and references therein).

The effects of gamma ray exposure, which may be sourced from the host material, are usually expressed as an absorbed dose rate in air. There are direct links between terrestrial gamma radiation and radionuclide concentration. The total absorbed dose rate D_{out} (nGy/h) in air at 1 m above surface level, due to the presence of ^{226}Ra, ^{232}Th and ^{40}K in the peloids studied, was calculated using the UNSCEAR (2008) equation:

$$D_{out}\ (\text{nGy/h}) = 0.462A_{Ra} + 0.604A_{Th} + 0.0417A_K \quad (5)$$

The calculated absorbed dose rates owing to the radionuclides vary between 31.52 and 330.675 nGy/h, with a mean of 118.37 nGy/h (Table 5). Most of the calculated absorbed dose rates are in the range of 28–120 nGy/h recommended by UNSCEAR (2008).

Khan *et al.* (2010) has provided a means to estimate effective doses, in order to check the influence of the effective dose (μSv/y) due to soil radioactivity on the residents of a surveyed area. The annual effective dose equivalent (AEDE) values were calculated using the recommended conversion factors (UNSCEAR, 2008; Kurnaz *et al.*, 2007; Khan *et al.*, 2010):

$$\text{AEDE} = D_{out}(\text{nGy/h}) \times \text{CF}(0.7\ \text{Sv/Gy}) \times \text{ET}_{out}(0.00137) \times 8760\ (\text{hy}^{-1}) \times 10^{-3} \quad (6)$$

In the present study, the value 0.7 $SvGy^{-1}$ was used as the conversion factor (CF) for the absorbed dose rate in air, the value of 0.00137 was used as the outdoor occupancy factor fraction (ET_{out}) and 8760 h was used as the time factor, i.e. 365×24 h for annual exposure, as recommended by UNSCEAR (2008).

The treatment with the peloids is applied ~18 times at 40 min. intervals. Hence:

$$\text{ET}_{out} = (18 \times 40/60)/(365.25 \times 24) = 0.00137 \quad (7)$$

The annual gonadal dose equivalent (AGDE) was calculated using equation 8:

$$\text{AGDE}\ (\mu\text{Sv/y}) = 3.09A_{Ra} + 4.18A_{Th} + 0.314A_K \quad (8)$$

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The excess lifetime cancer risk (ELCR) was calculated using equation 9:

$$\text{ELCR}\ (\mu\text{Sv/y}) = \text{AEDE} \times \text{DL} \times \text{RF} \quad (9)$$

where DL is the duration of life (70 y) and RF is the risk factor (Sv^{-1}), i.e. fatal cancer risk per Sievert. For stochastic effects, RFs of ICRP 103, BEIR(6) VII and ICRP 60 for the public, 0.057, 0.064 and 0.072, respectively, were used.

The absorbed dose rates vary across a wide range (Table 5). The calculated annual effective radiation dose from the 18 peloids is less than the reported safe limit (70 μSv/y). The dividing cells of bone marrow, gonads, epithelial cells of the stomach, intestines and skin are more sensitive to radioactivity than the cells of the liver, kidney, muscle, cartilage and bone. Therefore, the effective dose, i.e. the sum of the dose of each organ, should be computed. The total worldwide average effective dose from natural radiation is approximately 2.4 mSv/y (UNSCEAR, 2000) and consecutive 5-year averages vary between <1 and 5 mSv/y (whole body) in special cases (ICRP 1993). Correlations between the radioactivity concentrations and the subsequent dose calculations, i.e. Ra_{eq} and D_{out}, were not found. It is suggested that activity concentrations alone are not sufficient to classify the peloids as radioactive muds.

CONCLUSIONS

Most of the peloids examined have been used for pelotherapy in Turkey since ancient times. The radionuclide activity concentrations of the peloids are not uniform and although the concentration of ^{232}Th, ^{226}Ra and ^{40}K in some of them are greater than the world average, they cannot be considered as radioactive muds because, from the review, the Ra_{eq} values were >370 Bq/kg for only six of the peloids and the AEDE values were less than those suggested from the guideline values. During peloid treatment, a team of experts should consider the patient's age, gender, health problems, tissue/organ type, duration of the therapy and other similar factors, to evaluate the positive and negative health effects and thus ensure the health benefits of peloid

TABLE 5. Radium equivalent activity (Ra_{eq}), absorbed dose rates (D_{out}), annual effective dose equivalent (AEDE), annual gonadal dose equivalent (AGDE) and excess lifetime cancer risk (ELCR) of the peloid samples.

Sample number	Ra_{eq} (Bq/kg)	D_{out} (nGy/h)	AEDE (uSv/y)	AGDE (uSv/y)	ELCR (%)		
					ICRP 103	BEIR VII	ICRP 60
P-1	620.03	284.20	2.39	1947.54	0.010	0.011	0.012
P-2	766.77	330.67	2.78	2283.55	0.011	0.012	0.014
P-5	63.30	31.87	0.27	230.05	0.001	0.001	0.001
P-5/1	100.52	48.97	0.41	343.28	0.002	0.002	0.002
P-6	83.46	39.42	0.33	275.44	0.001	0.001	0.002
P-7	70.35	33.30	0.28	229.54	0.001	0.001	0.001
P-8	235.04	110.24	0.93	750.56	0.004	0.004	0.005
P-9	117.31	56.02	0.47	395.65	0.002	0.002	0.002
P-11	497.35	231.48	1.95	1629.36	0.008	0.009	0.010
P-12	143.94	67.06	0.56	473.35	0.002	0.003	0.003
P-14	478.55	223.19	1.88	1517.72	0.007	0.008	0.009
P-15	212.79	94.20	0.79	661.87	0.003	0.004	0.004
P-16	405.32	188.07	1.58	1278.79	0.006	0.007	0.008
P-17	66.54	31.52	0.26	224.07	0.001	0.001	0.001
P-18	112.87	53.09	0.45	375.29	0.002	0.002	0.002
P-19	74.18	34.52	0.29	233.52	0.001	0.001	0.001
P-20	376.39	174.80	1.47	1243.08	0.006	0.007	0.007
P-20/1	207.55	98.11	0.82	703.05	0.003	0.004	0.004
Minimum	63.30	31.52	0.26	224.07	0.001	0.001	0.001
Maximum	766.77	330.67	2.78	2283.55	0.011	0.012	0.014
Mean	257.35	118.37	1.00	821.99	0.004	0.004	0.005

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treatments while minimizing the negative effects of radiation exposure.

In terms of radioactivity, some of the measured, and all of the calculated values, are lower than the limits, so they mostly do not pose a health risk. This arrangement may contribute to the spas' healing effects for certain maladies. As it was not possible to determine the health effects of the radionuclides or describe safe therapeutic programs, future studies should make use of these new data to determine the effects, therapeutic or otherwise, of radionuclides on pelotherapy. The results of this study may provide a suitable baseline and may assist in establishing safe dosage levels of peloid radionuclides, in order to prevent unintended adverse health effects during spa therapy.

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