Conference Paper

Assessment of Some Trace Chemical Elements in Cajío Beach Peloid Using Nuclear Analytical Techniques

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Abstract

A peloid is a maturated mud or muddy dispersion with healing and/or cosmetic properties, composed of a complex mixture of fine-grained natural materials of geologic and/or biologic origin, mineral water or seawater, and common organic compounds from biological metabolic activity. Cajio beach peloid is a traditional peloid of the south coast of Güira de Melena municipality located in Artemisa province, Cuba. The peloid is a sea mud, used *in situ*, by the population of the locality. The present study has tree aims: ones is to determine trace chemical elements content (Sc, Cs, La, Ce, Nd, Sm, Eu, Gd, Tb, Tm, Yb, Hf, Ta, Th and U). The second aim is to determine natural (^{226}Ra, ^{238}U, ^{232}Th, ^{210}Pb, ^{40}K) and the anthropogenic (^{137}Cs) radionuclides in the peloid. Finally, to assess the level of contamination and radiological risk for the users of the peloid therapeutic practices.

Keywords: peloid, Cajío beach, trace elements, radionuclides

1. INTRODUCTION

According to the International Society of Medical Hydrology (ISMH) the term peloid comprises the "natural products composed of a mixture of mineral water (including sea water and salt lake water), with organic or inorganic matter, resulting from geologic or biologic process, or from both geologic and biologic process, which are utilized for therapeutic purposes under the form of packs or bath". [1]

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When used for therapeutic purposes peloid may not be exempted, *a priori*, of possibly causing adverse health effects. [2] Some recent investigations demonstrated the necessity of studding the geochemical abundance of potential hazardous elements in peloids, including radioactive ones. [3]

Cajio beach peloid is a traditional peloid of the south coast of Güira de Melena municipality located in Artemisa province, Cuba. The peloid is a sea mud, used *in situ*, by the population of the locality. The aim of the present study is to determine trace chemical elements content (Sc, Cs, La, Ce, Nd, Sm, Eu, Gd, Tb, Tm, Yb, Hf, Ta, Th and U) as well as natural (^{226}Ra, ^{238}U, ^{232}Th, ^{210}Pb, ^{40}K) and the anthropogenic (^{137}Cs) radionuclides in the peloid, to assess the level of contamination and radiological risk for the users of the peloid therapeutic practices.

2. MATERIALS AND METHODS

2.1. Field methodology and pretreatment of samples

Samples of Cajío beach stockade (SCB) and Cajío beach bridge (BCB) were collected directly from the deposits in Cajío beach. This beach is located at 22° 40′ 54.0″ N and 82° 27′ 42.1″ W of the Güira de Melena municipality, Artemisa, Cuba. The selected sampling areas are located in the deposit where the population extract the peloid for medical uses in Cajio beach. In both cases, a composite sample was prepared from the different collected fractions. After sampling, peloids samples were sealed in clean polyethylene containers, placed in a cooler at 4°C, and transported to the Analytical Laboratory of the Higher Institute of Applied Technologies and Sciences (InSTEC), Havana, Cuba, for further analysis.

The samples were air-dried after collection at the Analytical Laboratory of InSTEC and then sieved, in order to obtain a peloid fraction of size smaller than 63 μm, since metal concentrations in peloids are highly depend on size.

2.2. Total content determination of trace chemical elements

The total content of the trace chemical elements (Sc, Cs, La, Ce, Nd, Sm, Eu, Gd, Tb, Tm, Yb, Hf, Ta, Th and U) were determined by Instrumental Neutron Activation Analysis (INAA) performed at the pulsed fast reactor IBR-2 of the Joint Institute for Nuclear Research (JINR), Dubna, Russia. To analyze as many as possible elements, both short term (60 s) and long term (63 h) irradiations were used, the channel while, in the case of long term irradiation, the Cd-screened irradiation channel was used.

To determine the elemental concentrations a comparison method was used employing IAEA standard reference materials: 1633b (coal fly ash), 2709 (Trace elements in soil), 2710 (Montana Soil), 2711 (Montana Soil) and 433 (Marine sediment).

After short time irradiation, each sample was measured twice, after 2 and 10 minutes, for about 5 and 12 minutes, respectively. In the case of long time irradiation, gamma ray spectra were recorded after 4 and 14 days cooling time. All gamma ray spectra were recorded and processed using both Genie 2000 software and proprietary software developed at the Frank laboratory for Neutron Physics of JINR. In this way, the final uncertainties were calculated by taking into account statistic counting, sample preparation and detector calibration obtaining values between 3% for Na, Al, Zn and As and 40% for I.

The radionuclide activities in peloid samples were determinates by gamma ray spectrometry using the certified reference material (CRM) IAEA-375 (IAEA 1994b) and the standards UC-1 and UC-2 prepared in the University of Cantabria (Spain). [4] IAEA-375 and UC-2 were used for calibration, while UC-1 was used for quality control. CRM, standards and samples preparation was standardized at 50 g (dry weight) and put in the hermetic closed plastic container during 30 days so secular equilibrium between ^{226}Ra, ^{222}Rn and shorter half lives daughters of ^{222}Rn was assured. Samples, CRM and standard were measured during 24 h in the low-background gamma spectrometer (LBGS) at the Nuclear Analytical Laboratory at InSTEC. [5] LBGS is composed of a low-background chamber (LBC), using an n-type closed and coaxial high-purity germanium detector (DSG, NGC-3018, 130 cm^3, FHWM = 2.04 keV for 1332 keV ^{60}Co gamma line) equipped with an 8192 channel multichannel analyzer (webMASTER TARGET coupled to PC).

The gamma spectra were processed using the Gamma-W version 18.03 code (Dr. Westmeier Gesellschaft für Kernspektometrie GmbH). The gamma ray transitions of energies 186.3 keV (^{226}Ra), 351.9 keV (^{214}Pb), 661.9 keV (^{137}Cs) and 1460 keV (^{40}K) were used to determine the concentration of the radionuclides of interest. The minimum detectable activity (MDA) of the system for 24 h count acquisition were 1.0 Bq·kg^{-1} for ^{226}Ra, 0.6 Bq·kg^{-1} for ^{137}Cs, 1.9 Bq·kg^{-1} for ^{232}Th and 7.1 Bq·kg^{-1} for ^{40}K. The Determination Limit were calculated according to Currie criteria. [6] The activities of the radionuclides present in UC-1 standard measured in the LBGS (Table 1) shows "excellent" results (SR < 25%) for all determined activities. The deviation between the obtained results and the reference values was always less than 5 %, an excellent precision for environmental radioactivity measurements. [7]

Table 1: Activities (Bq·kg^{-1}) determined in UC-1 standard by relative method.

Nuclide	^{226}Ra	^{137}Cs	^{232}Th	^{40}K
Reference activity (Gomez and Soto, 1998)	24	45	30	480
Standard deviation (%)	13	4	10	4
Measured activity	25	44	31	460
Standard deviation (%)	8	9	10	7
Deviation from the reference value (%)	4.2	-2.2	3.3	-4.2
SR (%)	21	20	23	18

2.3. Peloid quality assessment

The references do not describe specific regulation of metal content for mud application with medicinal purposes. [2] Consequently, in this study to evaluate background concentrations and compare it with the obtained data, to determine if the element content in the analyzed sediment represents a natural concentration or if it has being affected by anthropogenic contamination. These values establish the minimum content above which elements can be considered as a risk of anthropogenic contamination compared to background levels (this criteria is the one commonly applied to metals). This approach is more realistic since it is based in real data from the studied place, and allows the calculation of the contamination factor (C_f) and the contamination degree (C_d). [7] The contamination factor for each i element is calculated as:

$$C_f^i = \frac{\overline{C_{0-1}^i}}{C_n^i}$$

$\overline{C_{0-1}^i}$ and C_n^i are the mean content and the background value for each metal, respectively; and n is the number of elements. The background value is defined as the original elemental concentration in the sediment sample, before possible anthropogenic or environmental contamination. However, as not data of the original elemental concentration of the chemical elements in Cajío beach peloide have been reported; background values of chemical elements will be assumed as the mean values of the earth crust (geochemical background), reported by Turekian, 1961 (**appendix 1**). [8] The contamination degree is calculated as:

$$C_d = \sum_{i=1}^{n-1} C_f^i = \sum_{i=1}^{n-1} \frac{\overline{C_{0-1}^i}}{C_n^i}$$

In this equation, the sum of all contamination factors are taken into account to evaluate the combined risk. [7, 9]

On the other hand, to evaluate the radiological quality of the peloid, the determined radioactive concentrations were compared with the average radioactive concentrations, reported by UNSCEAR, 2000 (table 2). [10]

Table 2: Average and range of radioactive concentrations UNSCEAR, 2000 [10].

Nuclide	^{226}**Ra**	^{238}**U**	^{232}**Th**	^{40}**K**
Mean (Bq·kg^{-1})	35	35	30	400
Range (Bq·kg^{-1})	17-60	16-110	11-64	140-850

3. RESULTS AND DISCUSSION

3.1. Total content of trace chemical elements

The trace chemical elements determinates in Cajío beach peloid are Sc, Cs, La, Ce, Nd, Sm, Eu, Gd, Tb, Tm, Yb, Hf, Ta, Th and U (**Table 3**). Trace elements play an important role in the functioning of life on our planet. Some of these elements can be highly toxic to various life forms; others are considered essential, but can become toxic at higher doses. [11]

Table 3 shows also the values of the contamination factor (Cf). These values are less than unity ($Cf < 1$) for the main part of trace chemical elements determined (Sc, Cs La, Ce, Nd, Sm, Eu, Gd, Tb, Tm, Yb, Hf, Ta and Th). These results means that the main part of chemical elements Sc, Cs La, Ce, Nd, Sm, Eu, Gd, Tb, Tm, Yb, Hf, Ta and Th are in concentrations less than the geochemical background, that have low contamination factor ($Cf < 1$). While, regarding U, the samples are considered as moderately contaminated ($1 \leq Cf < 3$).

On other hand, the table 4 shows the values of radioactive concentrations of the natural radionuclides ^{226}Ra, ^{238}U, ^{232}Th, ^{210}Pb, ^{40}K and the anthropogenic radionuclide ^{137}Cs, present in the Cajío beach peloid. Likewise, in the Figure 1 is showed the gamma spectrum obtained whit the data acquisition system. In the initial part of the spectrum (< 250 keV) are the radionuclides ^{235}U (185.7 keV 57%) and ^{226}Ra (186.2 keV 3%) which overlap in the spectrum. For that reason, the concentration of ^{226}Ra was calculated using the peak of ^{210}Pb with energy of 6.54 keV, considering that during the storage period the secular radioactive equilibrium between ^{226}Ra, ^{222}Rn and their daughters was reached.

Table 3: Total content of trace chemical elements determinates in Cajío beach peloid.

No.	Element	Total Content Determined (mg·kg^{-1})	Uncertainty (mg·kg^{-1})	Geochemical Background (mg·kg^{-1})	Contamination factor (Cf)
1	Sc	2.0850	0.1043	19	0.11
2	Cs	0.4475	0.0224	6	0.07
3	La	3.2150	0.4823	115	0.03
4	Ce	6.4500	0.9675	345	0.02
5	Nd	25.3992	3.8099	140	0.18
6	Sm	0.453	0.068	38	0.01
7	Eu	1.0995	0.1649	6	0.18
8	Gd	2.0150	0.6045	38	0.05
9	Tb	0.0886	0.0044	6	0.01
10	Tm	0.2575	0.0773	1.2	0.21
11	Yb	3.064375	0.3064	15	0.20
12	Hf	0.4255	0.0426	4.1	0.10
13	Ta	0.0792	0.0040	0.1	0.79
14	Th	1.0410	0.0521	7	0.15
15	U	3.660	0.183	1.3	2.82

Table 4: Radioactive concentration of nuclides in Cajío beach peloid.

Nuclide	Radioactive concentration (Bq·kg^{-1})	Uncertainty (Bq·kg^{-1})
^{226}Ra	8	1
^{238}U	11	3
^{232}Th	6	3
^{210}Pb	29	4
^{40}K	31	8
^{137}Cs	< 1.35	

The radioactive concentration of ^{40}K (31 ± 8 Bq·kg^{-1}) is significantly lower than that of other Cuban peloids: 236 ± 61 Bq·kg^{-1} (San Diego de los Baños), 115 ± 16 Bq·kg^{-1}(Elguea), 365 ± 40 Bq·kg^{-1} (Santa Lucía) (table 5). [12] In the same way, the radioactive concentration of ^{40}K determined in the Cajío beach peloid is less than the values reported in the literature for similar matrices (327-675 Bq·kg^{-1}) and less than the limit reported by UNSCEAR, 2000 for this radionuclide (140-850 Bq·kg^{-1}). [10, 13–15]

From the table 5, the results obtained in the present study it is possible to compare whit the results obtained for the same peloid in a previous study developed by Díaz

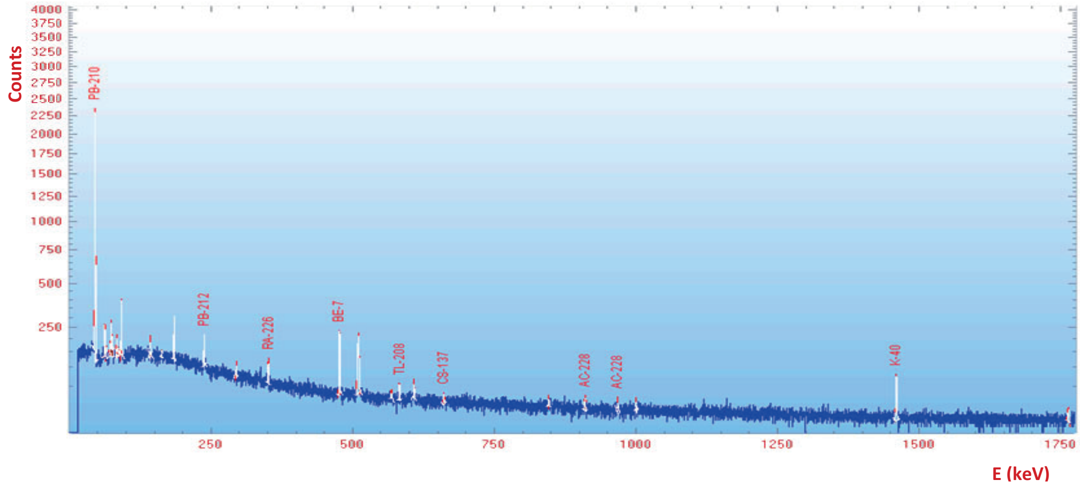

Figure 1: Gamma spectrum for the Cajío beach peloid.

Rizo, 2015. Also, it possible to compare whit some other Cuban peloids (San Diego de los Baños, Elguea and Santa Lucía) and other peloids around the world (Salsomaggio, Eugenian Hill, Safaga, Hugada and Abano). The results from the present study and the results obtained by Díaz Rizo, 2015 do not differ significantly. However, the determined values of the main part of radionuclides in the Cajío beach peloid are lower than the values reported by the rest Cuban and the world peloids. On the other hand, the radioactive concentration of ^{137}Cs in Cajío beach is similar or lower too than the radioactive concentrations reported for Cuban and world peloids. The determined value is within the range of concentrations reported for surface sediments in the northern hemisphere only affected by global radioactive precipitation (1-17 Bq·kg^{-1}). [13, 14, 16–22]

Table 5: Radioactive concentrations (Bq·kg^{-1}) of different peloids around the world.

Location	^{226}**Ra**	^{137}**Cs**	^{232}**Th**	^{40}**K**	**Reference**
Cajío, Cuba	8 ± 1	< 1,35	6 ± 3	31 ± 8	Present study
Cajío, Cuba	6 ± 1	< 1,6	6 ± 3	47 ± 7	Díaz Rizo et. al., 2015 [12]
San Diego de los Baños, Cuba	37 ± 3	5 ± 1	27 ± 5	236 ± 61	Díaz Rizo et. al., 2015 [12]
Elguea, Cuba	1 800 ± 298	< 1,6	38 ± 16	115 ± 16	Díaz Rizo et. al., 2015 [12]
Santa Lucía, Cuba	405 ± 65	< 1,6	21 ± 6	365 ± 40	Díaz Rizo et. al., 2015 [12]
Salsomaggio, Italia	30		48	659	Tateo et al., 2009 [12]
Eugenian Hill, Italia	286	2	35	368	Cantaluppi et. al., 2014 [12]
Safaga, Egipto	25		21	618	El-Arabi 2005 [12]
Hugada, Egipto	21		22	548	El-Arabi 2005 [12]
Abano, Italia	1 208	5	580	460	Doretti et. al., 1992 [12]

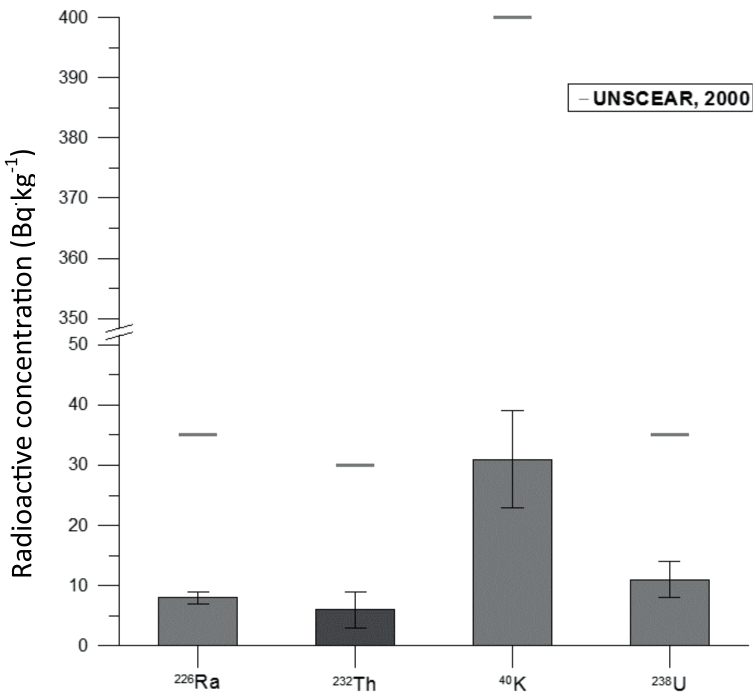

Figure 2: Comparison whit average radioactive concentrations UNSEAR, 2000.

^{226}Ra have been detected between 16 and 37 Bq·kg^{-1} in sediments of estuaries, bays or coast of different zones of normal radioactive background. [12] However, the radioactive concentration determined in Cajío beach peloid (8±1 Bq·kg^{-1}) is approximately half of the lowest of the reported values (16 Bq·kg^{-1}). In a similar way, the radioactive concentration of ^{238}U is lower than the lowest reported value (15 Bq·kg^{-1}). Nevertheless, other studies of Cuban coastal sediments report radioactive concentrations of 226Ra, similar to those of the peloid object of study: Havana Bay (10 Bq·kg^{-1}) and Cienfuegos Bay (9 Bq·kg^{-1}) [23, 24]

Due to the low radioactive concentrations of ^{226}Ra, radioactive concentration values of ^{210}Pb (29±4 Bq·kg^{-1}) are also lower than those reported in the literature for normal background areas. In addition, the value determinated is lower than the values reported for the San Diego de los Baños peloid (61±6 Bq·kg^{-1}) and Elguea peloid (352±14 Bq·kg^{-1}). The same situation happen when this value is compared whit other results reported, such as Jiaojiang, China (40-62 Bq·kg^{-1}) and the Turkish coast of the Aegean Sea (40-62 Bq·kg^{-1}). However, the radioactive concentration obtained in the present study for 210Pb is in the range reported for the Havana Bay sediment (25-184 Bq·kg^{-1}). [24]

Table 6

			Deep-Sea Sediments
			Clay
21	Scandium	Sc	19
55	Cesium	Cs	6
57	Lanthanum	La	115
58	Cerium	Ce	345
60	Neodymium	Nd	140
62	Samarium	Sm	38
63	Europium	Eu	6
64	Gadolinium	Gd	38
65	Terbium	Tb	6
69	Thulium	Tm	1.2
70	Ytterbium	Yb	15
72	Hafnium	Hf	4.1
73	Tantalum	Ta	0.1
90	Thorium	Th	7
92	Uranium	U	1.3

3.2. Quality evaluation

The contamination factors calculated were less than one for all chemical elements assessment except U (table 3). Therefore, according to the contamination degree the Cajío beach (Cd = 4.93) have low degree of contamination (Cd < n = 15). On the other hand, the results of the radioactive concentrations obtained for the natural radionuclides present in the peloid of Cajío beach (Figure 2) are lower than the average radioactive concentrations of sediments of the normal radioactive background (^{40}K = 400 Bq·kg^{-1}, ^{238}U, ^{232}Th and ^{226}Ra = 35 Bq·kg^{-1}). [10] In the correspondence with the results obtained, it can be assumed that the doses in the skin by superficial contact for patients should not be significant.

4. CONCLUSIONS

The total content of the trace chemical elements (Sc, Cs, La, Ce, Nd, Sm, Eu, Gd, Tb, Tm, Yb, Hf, Ta, Th and U) was determined and taking into consideration that the contamination degree, can be conclude that this content of trace chemical elements determined is not an impediment for its use with therapeutic purposes. On the other hand, the radioactive concentrations of natural (^{226}Ra, ^{238}U, ^{232}Th, ^{210}Pb, ^{40}K) and the anthropogenic (^{137}Cs) radionuclides determined do not represent any radiological risk for patient users. The comparison with some peloids and worldwide used for different medical proposes show that radionuclides content in Cajío beach peloid is suitable for its medical purposes.

APPENDIX

Appendix 1. Distribution of the Elements in the Earth´s Crust (Expressed in part per million).

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