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The measurement of gamma-emitting radionuclides in beach sand cores of coastal regions of Ramsar, Iran using HPGe detectors

Marziyeh Tari [a,*], Sayyed Ali Moussavi Zarandi [b,*], Kheirollah Mohammadi [b], Mohammad Reza Zare [c]

[a] *Department of Physics, Faculty of Sciences, Central Tehran Azad University, Tehran, Islamic Republic of Iran*
[b] *Department of Physics and Energy Engineering, University of Amir Kabir, Tehran, Islamic Republic of Iran*
[c] *Department of Physics, Faculty of Sciences, University of Isfahan, Isfahan 81746-73441, Islamic Republic of Iran*

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ABSTRACT

Radionuclides which present in different beach sands are sources of external exposure that contribute to the total radiation exposure of human. ^{226}Ra, ^{235}U, ^{232}Th, ^{40}K and ^{137}Cs analysis has been carried out in sand samples collected at six depth levels, from eight locations of the northern coast of Iran, Ramsar, using high-resolution gamma-ray spectroscopy. The average Specific activities of natural radionuclides viz., ^{226}Ra, ^{235}U, ^{232}Th, ^{40}K and ^{137}Cs, in the 0–36 cm depth sand were found as: 19.2 ± 0.04, 2.67 ± 0.17, 17.9 ± 0.06, 337.5 ± 0.61 and 3.35 ± 0.12 Bq kg^{-1}, respectively. The effects of organic matter content and pH value of sand samples on the natural radionuclide levels were also investigated. Finally, the measured radionuclide concentrations in the Ramsar beach were compared with the world average values, as reported by UNSCEAR (2000). None of the studied beaches were considered as a radiological risk.

1. Introduction

In general, approximately 85% of the annual total radiation dose of any person comes from natural radionuclides of both terrestrial and cosmogenic origins (Wild, 1993; NCRP, 1994; UNSCEAR, 2000; Belivermis et al., 2010). In addition to naturally occurring radionuclides, a large number of anthropogenic radionuclides have been produced and released into the environment by human nuclear activity, including nuclear weapons testing, the operation of nuclear power plants, research reactors, and nuclear fuel reprocessing. Nuclear accidents, such as Fukushima (on March 11, 2011) and Chernobyl accident (on April 26, 1986), have also released large amounts of radionuclides such as ^{137}Cs and ^{90}Sr into the environment (Abdi et al., 2009; Zare et al., 2012).

For several decades, extensive studies had been conducted worldwide to determine the natural radionuclides. Many papers reported the natural radioactivity level of sands and soils from different coastal zones (Malathi et al., 2005; Lakshmi et al., 2005; Alencar and Freitas, 2005; Lu and Zhang, 2008). In most places on the earth, the natural radioactivity varies only within narrow limit, whereas there are few regions in the world which are known for high background radiation areas (HBRAs). These are due to the local geological controls and geochemical effects and cause increased levels of terrestrial radiation (UNSCEAR, 1993, 2000; Mohanty et al., 2004). Very high background radiation areas are found at Ramsar and Mahallat in Iran (Sohrabi, 1993; Ghiassi-nejad et al., 2002); Brazil (Bennett, 1997; Paschoa, 2000; Mohanty et al., 2004); China (Wei et al., 1993); southwest coast of India (Sunta et al., 1982; Sunta, 1993; Paul et al., 1998), and in some other coun-

* Corresponding authors. Tel.: +98 2164543746; fax: +98 2188385777.
E-mail addresses: marziyeh.t_nph@yahoo.com (M. Tari), moussavi@aut.ac.ir (S.A. Moussavi Zarandi).

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ties (UNSCEAR, 2000). The radium in soil/water and the radon in air are responsible for the radiation at Ramsar in Iran, whilst the sources of high background radiations are the monazite sand deposits in the other cases. Some of these areas have been studied for many years in order to determine the risks and effects of long-term, low-level and natural radiation exposure (Sohrabi, 1998).

The objective of this work was focused **1.** to determine activity concentrations of ^{226}Ra, ^{235}U, ^{232}Th, ^{40}K and ^{137}Cs in the beach sand profile for studied beaches of Ramsar, by high – resolution Gamma ray spectrometry; **2.** to study the distribution of natural radioactivity in the beach sand profile and **3.** to investigate the relationship between some sand parameters and natural radionuclide levels.

The area of interest was located in southwest part of Caspian Sea. Ramsar is a northern coastal county on the west of Mazandaran province in Iran. The regional sampling was performed using the systematic random sampling method in Visual Sample Plan (VSP) software, According to the international standard (IAEA-TEC-DOC-1360, 2003). In January 2012, 48 beach sand samples from 8 different coastal zones were collected, ranging from 55°44′51.5″N, 36°52′58.4″E to 50°35′56.2″N, 36°57′46.6″E, along southwest coast of Caspian Sea. The sampling stations are presented in Table 1 and the map of sampling spots is indicated in Fig. 1. In order to analyze the activity concentration variations of ^{226}Ra, ^{235}U, ^{232}Th, ^{40}K and ^{137}Cs, 6 samples were taken at a step of 6 cm depth covering 36 cm depth. The process of sampling was carried out by a metal corer (radius = 7 cm). Each sample profile was obtained from five subsamples collected from four corners and the center of an area corresponding to 1 m^2. The five subsamples at each depth interval were homogenized in situ, and weighing approximately 1.5 kg, as a representative profile, were properly stored in a thick plastic bag and marked for processing before analysis. During sampling a hand-held EXPLORANIUM detector was used to monitor the environment.

In the laboratory, the sand samples were first dried at room temperature for several days. Then stones, and other foreign particles were removed and all the sand samples were placed in a drying oven. The drying temperature was set to (100–110 °C ± 4 °C) for 24 h. The dried samples were pulverized into fine quality powder, homogenized and sifted through an ASTM sieve of 300 μ mesh size (Lee et al., 2001). All of the samples were then weighed and finally, Samples of approximate mass 950 ± 1 g each transferred into marinelli beakers of 1000 ml capacity (Degerlier et al., 2008). The marinelli beakers were hermetically and externally sealed with an adhesive tape over the caps to prevent the escape of gaseous ^{222}Rn and ^{220}Rn from the samples and stored for at least 5 weeks to allow secular equilibrium to be attained between long-lived radioisotopes ^{226}Ra, ^{232}Th and their associated radioactive progenies (Mollah et al., 1987; Abdi et al., 2009; Zare et al., 2012). After this period, the samples were analyzed using gamma ray spectrometry.

Table 1
Collected samples along the Ramsar coastline of Iran.

Ser. no.	Sampling location code	Geographical position		
		Longitude (Deg, Min, S)	Latitude (Deg, Min, S)	Tourist resort
1	RAETTBP	50, 44, 51.5	36, 52, 58.4	Ettehad
2	RAMIYBP	50, 44, 30.2	36, 53, 04.6	Miyanhaleh
3	RASSHB1	50, 42, 18.1	36, 53, 56.5	Sadat Mahalleh
4	RASSHB2	50, 42, 13.1	36, 53, 55.4	Sadat Mahalleh
5	RATOSBP	50, 40, 33.8	36, 55, 02.4	Toskasara
6	RATALBP1	50, 38, 27.6	36, 56, 49.5	Safa Rood
7	RATALBP2	50, 39, 01.1	36, 56, 33.5	Safa Rood
8	RAGOLBP	50, 35, 56.2	36, 57, 46.6	Golestan

Measurements of the activity concentrations of primordial radionuclides (^{226}Ra, ^{235}U, ^{232}Th and ^{40}K) and anthropogenic radionuclide ^{137}Cs in Bq kg^{-1} dry weight of the samples were performed using low-background gamma-ray spectrometry based on two coaxial P-type HPGe detectors of 38.5% and 55% relative efficiencies. The resolutions (FWHM) of the spectrometers were found to be 1.98 keV and 1.8 keV for the 1332 keV gamma-ray line of ^{60}Co, respectively. In order to achieve the lowest unwanted ambient radioactivity from room background sources, each of detectors was housed in a vertical position in a passive lead cylindrical shield of about 10 cm thickness. The lead shield contained two internal concentric cylinders of cadmium and copper sheets by 2 mm and 3 mm thickness respectively, which significantly reduced the effects from photo fluorescence of the lead X-rays (Bikit et al., 2006;Kurnaz et al., 2007). The output of each detector was coupled to a standard spectroscopy amplifier. The data was taken directly to a multi channel analyzer (4096-channels). Spectrum acquisition was done by using the computer software (MAESTRO – 32) and also Spectral analysis and peak fitting were performed using the OMNIGAM software. Each spectrum was accumulated for period of 86,400 S in order to have statistically relevant results.

Three types of calibrations were carried out for gamma spectrometer. These are energy, resolution and efficiency calibrations (El-Gamal et al., 2007). The energy calibrations of detectors were done using two standard point sources of ^{241}Am and ^{226}Ra. The resolution calibration (called FWHM calibration) that establishes a function to describe the peak width versus the spectral energy was done using three standard point sources of ^{57}Co, ^{137}Cs and ^{60}Co. The absolute photopeak efficiency calibrations of systems were performed using photopeak from a standard mixed source containing ^{241}Am, ^{109}Cd, ^{57}Co, ^{133}Ba, ^{137}Cs and ^{60}Co of known activities, that were manufactured by Institute of Atomic Energy POLATOM, Radioisotope Center. The counting geometry of the sand samples and the standard mixed source used for efficiency calibration were kept constant (Al-Trabulsy et al., 2011). A wide range of different gamma-ray energy lines ranging from about 59 keV up to 1765 keV, were covered using these standard sources.

The absolute photo peak efficiencies were determined using two following polynomial fits:

$$y = a + b\ln x + c(\ln x)^2 + d(\ln x)^3 + e(\ln x)^4 + f(\ln x)^5$$
: For detector with 38.5% relative efficiency (1)

$$y = a + bx + cx^{2.5} + d/\ln x + e/x^{0.5}$$
: For detector with 55% relative efficiency (2)

These expressions are adequate for determining efficiency of gamma energies from 59.5 keV to 2000 keV for both detectors. The parameter values corresponding to measuring conditions are shown in Figs. 2 and 3. As a fit becomes more ideal, the r^2 values approach 1.0 (0 represent a complete lack of fit), the standard error decreases toward zero, and the F-statistic goes toward infinity.

Following the spectrum analysis, count rates for each detected photopeak and activity per mass unit (radiological concentration) for each of the detected nuclides is calculated. On the basis of the measured photopeaks of relevance for this work that mainly are emitted by specific radionuclides in the ^{232}Th and ^{238}U decay series and single occurring ^{40}K, their radiological concentrations in collected samples were determined. While ^{40}K can be measured directly by its own gamma-rays, ^{232}Th and ^{226}Ra can be specified with gamma-rays of their decay products. Calculations were combined under the assumption of secular equilibrium, due to the much smaller life time of daughter radionuclides in the decay series of ^{232}Th and ^{226}Ra. The initial activity concentration of ^{232}Th was inferred by several gamma-ray transitions at 911.0 keV,