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Preparation of Papers for UCTE 2017: Natural radionuclides in Fort-Dauphin beach sands, South Madagascar

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Abstract

The Fort-Dauphin beach sand placer occurs as black sands on the East-South of Madagascar. The placer contributes 2/3 of the total heavy mineral resources of this country. The laboratorial measured activity concentration of ^{238}U, ^{226}Ra and ^{232}Th of sand samples along the beach in Fort-Dauphin beach range from 2060 Bq/kg to 4220 Bq/kg, 1500 Bq/kg to 2880 Bq/kg and 11,000 Bq/kg to 24,400 Bq/kg respectively. The absorbed dose rate ranges from 7350 to 16,000 nGy/h, the dose rates are from 120 to 270 times higher than the average terrestrial radiation background of 60 nGy/h, the effective dose rate for the human being living in this area could be up to 140 mSv/year with principal contribution from ^{232}Th activity concentration.

Introduction

The natural isotopes of ^{238}U, ^{232}Th, ^{40}K and their progeny principally contribute to terrestrial natural background radiation. The concentrations of the isotopes depend on the rock lithology, mineral compositions and underground water movement (Qu Limei et al., 2008; Strezov et al., 2009; Chau et al. 2008a; 2009; 2016). The radionuclides occurring in the Earth's crust are the sources of external terrestrial radiation which contribute of 15% of the average annual dose (2.4 mSv) (ICRP, 1987; UNSCEAR, 2008). In many regions on the Earth the high background radiation is observed, namely beach sand placers in India, Egypt, Greece, Brazil and Bangladesh (Mohanty et al, 2003a; Abd El Wahab & El Nahas, 2013; Takayuki et al, 2015; Papadopoulos et al, 2016). Deposits often contain heavy minerals, they were formed by long time weathering and erosion of different rock types, transported and deposited along the beaches. The sand beach problems were studied by several scientists especially the natural radionuclides in different regions such as Chhatrapur and Erasama beach placer deposit of Orissa in India; beach sand of Rosetta in Egypt; Placer Sands in Southeast Bangladesh; beach sands in Ilha Grande of Brazilian southeastern; beaches of Kavala, Sithonia Chalkidiki, Maronia, Samothraki, Mykonos of Greece: (Mohanty et al, 2003b; Freitas and Alencar, 2004;Mohanty et al, 2004; Sengupta et al, 2005; Alencar & Freitas, 2005; Sulekha et al, 2009; Nada et al, 2012; Abd El Wahab & El Nahas, 2013; Takayuki, et al, 2015; Papadopoulos et al, 2014a, b; 2015a, b; 2016). In this paper, we study on the natural radionuclides, effective dose rates and relations between the mentioned parameters in the Fort-Dauphin beach sand placer in the East-South of Madagascar.

Geological setting of the studied placer

The Fort-Dauphin beach sand placer is located in Flandrian Dunes surrounded by the Vohimena Mountains and the Indian Ocean in South – East of Madagascar Island (Fig. 1). In the studied region there are igneous, metamorphic and sedimentary formations. The igneous formation occupies majority part of the Fort-Dauphin region and consists of the Vohimena and the Anosyenne Mountains granitic complexes. The essential compositions of the granitic complex are charnockite and garnet - biotite granite with few dozen meter of lenses containing apatite-biotite, zircon and large crystal of monazite. The Anosyenne granite is composed of ilmenite, monazite and magnetite (Andriamanantena, 2008). The coastal sedimentary formation consists of heavy minerals transported from eroded Anosyenne granite. Due to the potassium feldspar alteration, greenish yellow color is observed along the formation (Lacroix, 1922). The metamorphic formation in this area is characterized by the banded gneiss with biotite, cordierite, spinel, sillimanite, garnet, orthoclase and plagioclase (Bazot, 1974). The placer is very rich in titanium mineral accompanied with minerals bearing natural radioactive and heavy elements. The deposit has been exploited by Rio Tinto Qit Madagascar Mineral QMM Company.

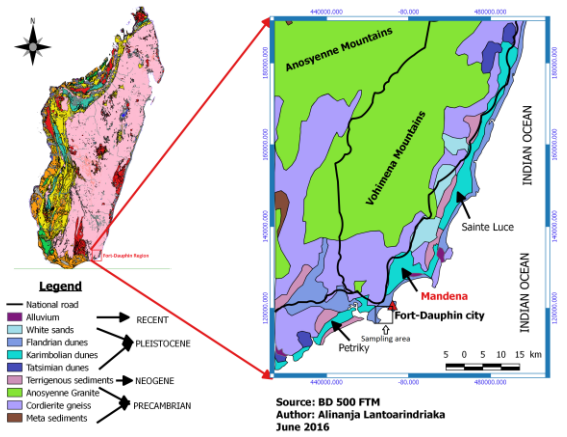

Fig.1 Geological map of the study area, modified from BD 500 FTM map (2016)

Materials and methods

Collection and pre-treatment of the samples

The sand samples were collected from the Fort-Dauphin beach sand placer. Each sample collected approximately 600 g. The sample for gamma measurement was milled until the grains became less than 2 mm, then it was dried in an oven at 120^0 C for 24 hours to ensure that moisture was completely removed, then weighted accurately and packed in an aluminum cylindrical beaker of 120 ml capacity and sealed to prevent the escape of radon. The weighed and tightly sealed samples were left for at least 22 days to reach secular equilibrium between ^{222}Rn and ^{226}Ra in the samples (Jodlowski & Kalita, 2010). After the stored period, the samples were measured using gamma spectrometer with HPGe detector.

Gamma spectroscopic analysis

The activity concentrations of ^{40}K, ^{238}U, ^{226}Ra and ^{232}Th were determined using gamma-ray spectrometer coupled with HPGe detector of the relative efficiency of 42% and resolution of 1.9 keV for 1332 keV line. The gamma spectrometer was calibrated using the IAEA reference materials RGU, RGTH, RGK as standard samples.

The gamma lines of 1000.8 keV (0.7%) from ^{234}Pa and 609.3 keV (46.1%), 1120.3 keV (15.0%) and 1764.5 keV (15.9%) from ^{214}Bi were used to determine the activity concentration of ^{238}U and ^{226}Ra, while that of ^{232}Th were determined from the gamma lines of 911.2 keV (29.0%) and 969.0 keV (2.3%) from ^{228}Ac and 583.0 keV (30.9%) and 2614.4 keV (35.8%) from ^{208}Tl (ICRP, 1983; IAEA, 1989). For ^{40}K, its activity concentration was determined from its 1461 keV gamma line. The maximum counting time for every sample was 50 hours and the obtained uncertainty was less than 3% for low active samples. The self-gamma absorption resulted from the difference in density of the studied samples and standard ones were introduced to follow the method described by Jodlowski (2006). To avoid the photo effect absorption in the sample, all of the gamma lines were used higher than 500 keV.

The activity concentrations of ^{40}K, ^{226}Ra and ^{232}Th is calculated by formula (1)

$$A_{sp} = \frac{N_{sp} M_{st} A_{st} C_i}{N_{st} M_{sp}} \quad (1)$$

Where: $\mathbf{N_{sp}}$, $\mathbf{M_{sp}}$ and $\mathbf{N_{st}}$, $\mathbf{M_{st}}$ are the net measured intensity and mass of the sample and standard sample respectively, $\mathbf{C_i}$ - the corrected factor for the differences between the densities of the sample and standard sample.

As the activity concentration of ^{232}Th was very high in the study samples, the number of counts from the gamma lines 1459,88 keV and 1000,6 keV emitted from ^{228}Ac, which is the progeny of ^{232}Th is also very high. Therefore the ^{40}K concentration in the samples of this study after subtract the amount contribute from ^{28}Ac is very low, it is not possible to determine ^{40}K concentration using gamma spectrometry in this case. The determination of the ^{40}K activity concentration was corrected to eliminate the interference of the thorium progeny and calculated as follow (2), (3):

$$A_{K-40} = A_{total}(I_{total} - I_{Ac-228}) / I_{total} \quad (2)$$

$$I_{Ac-228} = \varepsilon_{\gamma-Ac-228} A_{Th-232} M_{sp} P_e T \quad (3)$$

Where: $A_{K\text{-}40}$, A_{total}, $A_{Ac\text{-}228}$ – is the activity of potassium subtracted from the interference part of thorium progeny, active total calculated from gamma line 1461 keV, activity of ^{228}Ac from gamma line 1459,88 keV contributed in gamma line 1461 keV. $\boldsymbol{I_{total}}$, $\boldsymbol{I_{Ac-228}}$ - is the measured number of counts in the gamma line 1461 keV, number of counts in the gamma line 1459,88 keV from ^{228}Ac contributed in the gamma line 1461 keV. Due to short haft time period of ^{228}Ac (6.13h), the radioactive equilibrium between ^{232}Th and ^{228}Ac always regarded as established in the solid materials. $A_{Ac\text{-}228} = A_{Th\text{-}232}$ – is the activity of ^{232}Th. $\varepsilon_{\gamma-Ac-228}$ - is the emission efficiency of the gamma ray of 1459,88 keV from ^{228}Ac. Msp – mass of sample. P_e decay rate of gamma line 1459.88 keV of ^{228}Ac. T – time of measurement.

The gamma absorbed dose rate was calculated using the measured activity concentrations of ^{226}Ra, ^{232}Th and ^{40}K of the measured sample (UNCEAR, 2000):

$$D(nGy/h) = 0.041 \cdot K + 0.462 \cdot Ra + 0.604 \cdot Th \quad (4)$$

Where K, Ra and Th are specific activity of ^{40}K, ^{226}Ra and ^{232}Th of the sample expressed in [Bq/kg]

Results and discussions

Radioactive characteristic

The measured activity concentrations of ^{238}U, ^{226}Ra and ^{232}Th are summarized in (Tab. 1) which ranges from 2063 to 4224, 1501-2878 and 11013-24379 Bq/kg respectively. The high concentration of ^{238}U, ^{226}Ra and ^{232}Th in the study samples implies that the U and Th are the principal natural radioactive elements occurring in the beach sand, while the activity concentration of ^{232}Th is higher than ^{238}U by five times, equivalent to fifteen times in mass. It is common among some beach sand deposits in Australia (Elsner, 2010; Dean, 2011). This beach sand contribute heavy mineral and high concentration of radionuclides also, it is happening in different regions in worldwide and were being studied by many scientists such as (Mohanty et al, 2004; Sengupta et al,

2005; Alencar & Freitas, 2005; Sulekha et al, 2009; Nada et al, 2012; Abd El Wahab & El Nahas, 2013; Takayuki, et al, 2015; Papadopoulos et al, 2014a, b; 2015a, b; 2016). The average absorbed dose rate ranged from 7347 to 16055 nGy/h, the dose rates are 122 to 268 times higher than the average global terrestrial radiation of 60 nGy/h and 80 time higher than that of the world range (10-200 nGy/h). So the effective dose rate for a human living in this area could be up to 140 mSv/year.

The result show the correlations between ^{232}Th versus ^{238}U and ^{238}U versus ^{226}Ra (Fig 2, 3), this phenomenon could explain of the co-occurrence of the uranium and thorium. There are disequilibrium between ^{226}Ra and ^{238}U, the statement could be explained by the geochemistry of sea water condition on beach sand. The activity concentration of ^{238}U is higher than those of the ^{226}Ra in the samples, the activity ratio between ^{226}Ra and ^{238}U (^{226}Ra/^{238}U) varies from 0.56 to 0.73, the ratio is the characteristic for the shelf sediments (Polański 1972). Sometime the ratio ^{226}Ra/^{238}U of 1.76–2.33 in soil and sand samples are obtained from Penang, Malaysia (Almayahi et al., 2012). Thorium is the principal contributor in the radioactive anomalous in the placer (Fig. 4).

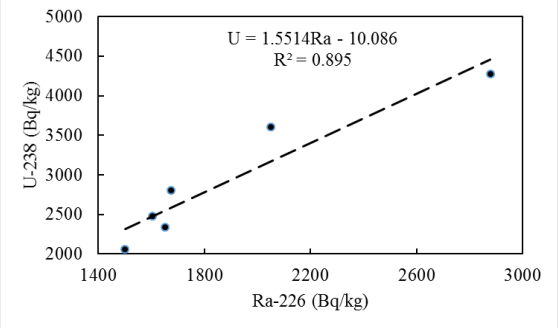

Fig. 2. The relation between Ra-226 and U-238 in sand smaples

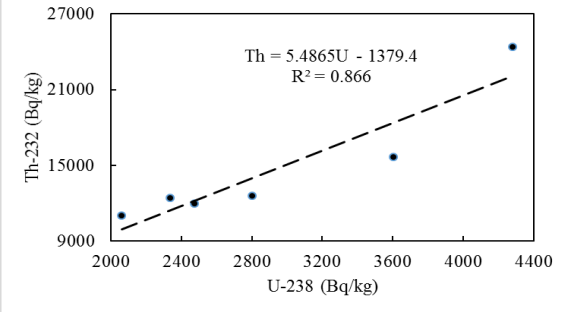

Fig. 3. The relation between U-238 and Th-232 in sand smaples

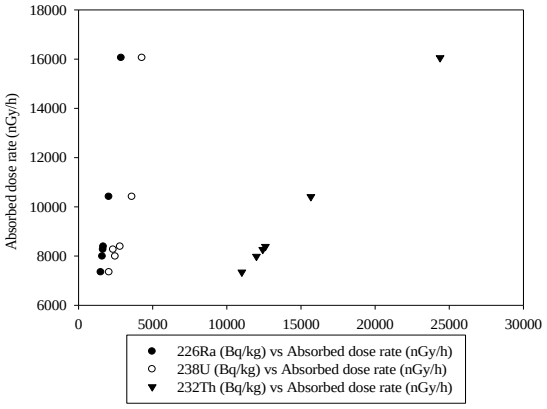

Fig. 4. Relation between estimated gamma absorbed dose rates and radium, uranium and thorium measured by gamma spectrometer coupled with HPGe detector.

Name	^{226}Ra (Bq/kg)	^{238}U (Bq/kg) After eliminate	^{232}Th (Bq/kg)	Absorbed dose rate (nGy/h)	Effective dose rate for people living in study area (mSv/year)
S 1	1501±19	2063±32	11013±165	7347	64.4
S 2	1604±20	2476±70	11994±178	7988	70.0
S 3	1675±21	2803±70	12604±189	8387	73.5
S 4	1651±21	2337±67	12421±186	8266	72.4
S 5	2050±25	3605±80	15666±235	10410	91.2
S 6	2878±36	4278±123	24379±366	16055	140.6

Tab. 1. ^{226}Ra, ^{238}U, ^{232}Th contents of the sand samples measured by gamma spectrometer coupled with HPGe detector and estimated gamma absorbed dose rates and effective dose rate for people living in the study area

Conclusions

In the Fort-Dauphin beach sand placer, there are very high natural radioactive background with the average absorbed dose rate up to 16055 nGy/h excluding cosmic radiation. The effective dose rate for the human living in this area amounts to 140 mSv/year.

There are linear correlation between ^{238}U, ^{226}Ra and ^{232}Th, and the principal anomalous radioactivity is thorium anomaly.

The ^{40}K concentration is very low in comparison with ^{232}Th concentration, undeterminable in gamma spectrometric results.

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