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RESEARCH ARTICLE

ACTIVITY CONCENTRATION OF RA²²⁶, TH²³², AND K⁴⁰ IN THE LIGNITE SAMPLES OF NICHAHOMA LIGNITE BELT OF KASHMIR VALLEY

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ABSTRACT

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Specific activity, Dose rate, Annual effective Dose Equivalent, External hazard index. Since the creation of the universe, natural radioactivity has been an integral part of our environment and all living things are living and have to live in a sea of radiations. The natural radioactivity is caused by γ -radiation originating from the uranium, thorium series and K⁴⁰. In the present study the activities of Ra²²⁶, Th²³², and K⁴⁰ have been measured in the lignite samples of the of Nichahoma Lignite belt of the Kashmir valley, India using NaI(Tl) Gamma ray spectrometer. The measured specific activity concentration of Ra²²⁶ and Th²³² was found to be higher than the Indian coal mines, the data presented by Germany, USA, and UK. The radiation Hazard indices are also calculated and reported.

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INTRODUCTION

Lignite is formed from the remains of trees and ferns which grew in swamps some 500 million years ago. Initially bacterial and chemical action on such debris products produced peat as an intermediate product. The deposition of minerals caused the peat to sink, which is composed of dead leaves, sterns and roots of plants, mainly cellulose $(C_6H_{10}O_5)_n$. As a result of high pressure and temperature during the geological changes under earth's surface peat was transformed into Lignite. Since the dawn of the universe, natural radiation has been an integral part of our environment. Humans have always been living in a sea of radiations and have to live. The amount of back ground radiations is different in the terms of height, the amount of nuclei present in the soil, geology, geography and rainfall of a place. Radioactivity is common in rocks, soil, beach sand, in rivers and oceans, and even in building materials and homes (Sannapa et al., 2010).

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Some radioactive nuclides are detectable in soil. They belong to natural radionuclides such as the members of the Uranium and Thorium series. More specifically, natural environmental radioactivity and associated external exposure due to gamma radiation levels depends on the geological and geographical conditions and appear at different levels in soil of each region in the world. The specific levels of terrestrial radiations are related to the geological composition of each lithologically separated area and the content of the rock from which the soils originate in each area in the radioactive element of Thorium (Th²³²), Uranium (U²³⁸) and Potassium (K⁴⁰) (Cunha Primal *et al.*, 2012; Ramachandran, 2011). The objective of the present study was to determine the activity concentration due to Ra²²⁶, Th²³², and K⁴⁰ in the lignite samples of lignite belt of Nichahoma, Kupwara, Kashmir Valley.

Experimental Details

Geological outline

The Kupwara district of Kashmir Valley has Latitude of 34.52 degrees North and Longitude of 74.25 degrees East.

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The area has Quartz veins carrying sulphides of copper and iron with some oxides, arsenides, gold, silver traces, and marbles brands. Further, sizeable deposits of lignite associated with Karewa group occur in Nichahoma and Shaliganga – Lonyalob sectors in Handwara tehsil of Kupwara district (Geology and mineral Resources, 2004).

Sampling and Sample Preparation

In this study 10 different samples of lignite were randomly collected. The collected samples are placed in labeled polyethane bags then transferred to our lab for preparation and analysis. The samples are crushed, homogenized and sieved through a 200 mesh, which is the optimum size enriched in heavy minerals and were dried in an oven at a temperature of 110^{0} C for 24 hours to ensure that the moisture is completely removed. The samples are weighted, packed in cylindrical geometry, labeled and hermetically sealed in air tight plastic containers. The sealed samples are then stored for four weeks in a safe place to enable them to attain a state of secular equilibrium, where the rate of progeny becomes equal to that of the parent (Ra²²⁶ and Th²³²) (Akkurt *et al.*, 2010).

Experimental Setup

The radioactivity of Ra²²⁶, Th²³², and K⁴⁰ in the samples was determined using the gamma-ray spectrometer consisting of a NaI(Tl) detector (crystal size 40.0 mm x 60.0 mm) connected to 1024 channel multichannel analyser (MCA). Before measurement, the system is calibrated using Cs¹³⁷ and Co⁶⁰ radioactive sources produce γ -ray energies of 662 KeV, 1173 KeV and 1332 KeV, respectively. The related fit has been displaced in Figure 1.

The spectrum is analysed by Leybold Cassy Lab Multi-Channel Analyser model Pocket- CASSY 559901 (Germany made). The activity of K⁴⁰ was measured directly with 1460.7 (10.7%) KeV peak of the gamma ray spectrum. To determine the activity concentration of Ra²²⁶, the average value of gamma ray lines 295.1 (19.2%) and 351.9 (37.1%) KeV from Pb²¹⁴ to 609.3 (46.1%) and 1764.5 (15.9%) KeV gamma ray from Bi²¹⁴ is used. The activity concentration of Th²³² was determined using the average value of gamma rays peaks 238.6 (43.6%) KeV from Pb²¹², 338.4 (12%), 911.1 (29%) and 968.9 (17.4) KeV from Ac²²⁸, 583.1 (86%) and 2614 KeV from Tl²⁰⁸. Each sample was examined for 7200 seconds. The analysis of results is performed using Microsoft Excel software.

The activities for the natural radionucleides were calculated using the following relation (Beretka and Mathew, 2003):

$$A(Bqkg^{-1}) = \frac{N}{\gamma \times \varepsilon \times t \times m}$$
(1)

Where A is the activity of the radionuclide in $Bqkg^{-1}$, N represents the counts under the most prominent photo peak, calculated from subtracting the respective count rate from the back ground spectrum obtained for the same counting time, ϵ is the detector efficiency of the specific gamma-ray, γ is the absolute transition probability of gamma decay, t is the counting time(s) which is 7200 seconds and m is the mass of the sample.

Calculation of Radiological effects

The commonly used radiological hazard index Ra_{eq} is called the radium equivalent activity.



Figure 1. The related fit obtained for Calibration of the instrument

It is defined as the weighted sum of activities of the Ra²²⁶, Th²³², and K⁴⁰ radionuclides based on the assumption that 370 $Bqkg^{-1}$ of Ra²²⁶, 259 $Bqkg^{-1}$ of Th²³² and 4810 $Bqkg^{-1}$ of K⁴⁰ produce the same gamma ray dose constant. The index is calculated from the following relation suggested by Beretka and Mathew: (Beretka and Mathew, 2003)

Where A_{Ra} , A_{Th} , and A_K are the mean activity concentrations of Ra²²⁶, Th²³² and K⁴⁰, respectively, in $Bqkg^{-1}$. The above equation has been derived for calculating the absorbed dose rate in air at a height of 1.0m above the ground from the measured radionuclide concentrations in environmental materials.

Table 1. The activity concentration of the Ra²²⁶ and Th²³² series and K⁴⁰ in (Bq/Kg) of the studied Lignite Samples

Sample Code	Specific Activity Concentration (Bq/Kg)						
	Ra ²²²	Th	²³² K ⁴⁰	Raeq(Bq/Kg)			
L-1	40.4±1.9	17.4±1.3	37.4±1.7	68.0±3.5			
L-2	43.4±0.6	19.8±0.5	36.6±2.2	74.4±1.4			
L-3	47.1±0.5	21.3±0.5	38.2±1.3	80.3±0.4			
L-4	42.9±0.8	18.6±0.9	39.6±0.5	72.4±2.1			
L-5	49.9±1.4	19.5±0.6	40.7±0.1	80.8±0.6			
L-6	40.3±1.6	23.4±0.5	31.8±4.9	76.1±0.9			
L-7	50.6±1.6	22.6±0.4	42.0±0.8	86.0±2.2			
L-8	41.6±1.3	24.1±0.9	43.7±1.8	79.3±0.1			
L-9	53.7±2.6	25.9±1.4	46.2±3.2	94.1±4.8			
L-10	43.7±0.5	21.6±0.5	48.9±4.7	78.2±0.3			
Mean Value	45.4±1.2	21.4±0.9	40.5±2.1	78.95±1.63			

Table 2. The average value of concentration of the natural radionucleides in lignite samples (BqKg⁻¹) reported for different parts of world

Mine and country	Specific Ac	Reference		
	Ra ²²⁶	Th ²³²	K^{40}	
Drage Mine, Bosinia	1191.34±4.83	26.67±1.86	32.92±7.20	[13]
Table Mine, Herzegovina	263.33±2.43	11.54±1.04	137.64±6.06	[13]
Indian Coal Mines	16-27	8-27	50-100	[3]
Fly-Ash India	200-150	50-150	250-700	[3]
Australia	19-24	11-69	23-140	[12]
Brazil	72	62	-	[12]
Germany Lignite	32	21	225	[12]
Greece Lignite	44-206	-	-	[12]
Romania	126	62	-	[12]
UK	8-22	7-19	55-314	[12]
USA	9-59	4-21	-	[12]
Nichahoma Kupwara, Kashmir, India	45.4±1.2	21.4±0.9	40.5±2.1	Current Study

Table 3. The average values of radiation hazard indices of the investigated samples

Sample	D (nGy/h)	$E_T (mSv/y)$	H _{ex}	$I_{\gamma}(Bq/Kg)$
Lignite	44.7	54.9	0.21	0.55

 $Ra_{eq} = (A_{Th} \times 1.43) + A_{Ra} + (A_K \times 0.077)$ (2)

Where A_{Ra} , A_{Th} , and A_K are the specific activities of Ra²²⁶, Th²³², and K⁴⁰ respectively.

Another radiation hazard index used to estimate the level of γ -radiation hazard associated with natural radionucleides is called the representative index I_{γ} , is defined by the following relation: (Miah *et al.*, 1998)

$$I_{\gamma} = \frac{1}{150} A_{Ra} + \frac{1}{100} A_{Th} + \frac{1}{1500} A_{K}$$
(3)

Where $A_{Ra}A_{Th}$, and A_K have the same meaning as in the equation (2).

The total air absorbed dose rate $(nGyh^{-1})$ due to the mean activity of Ra²²⁶, Th²³², and K⁴⁰ $Bqkg^{-1}$ can be calculated using the formula [Sannapa *et al.*, 2010; Alam *et al.*, 1999]:

$$D = 0.462A_{Ra} + 0.604A_{Th} + 0.042A_K \tag{4}$$

The absorbed dose rate in air at 1 meter above the ground surface does not directly provide the radiological risk to which an individual is exposed (Krieger, 1987). The absorbed dose can considered in terms of the Annual Effective Dose Equivalent (E_T) from the outdoor terrestrial gamma radiation which is converted from absorbed dose by taking into account two factors, namely the conversion coefficient from the absorbed dose in air to effective dose and the occupancy factor. The E_T can be estimated using the following formula (UNSCEAR, 2000):

$$E_T(mSv. y^{-1}) = D(nhh^{-1}) \times 24h \times 365.25 days \times 0.2 \\ \times 0.7(SvGy^{-1})$$
(5)

To limit the radiation exposure attributable to natural radionucleides in the samples to permissible dose equivalent limit of $1 mSv. y^{-1}$. The External hazard index (H_{ex}) due emitted gamma rays of the samples is calculated and examined according to the following relation: (UNSCEAR, 2000)

$$H_{ex} = \frac{A_{Ra}}{3\ 7\ 0} + \frac{A_{Th}}{2\ 59} + \frac{A_k}{4\ 8\ 10} \le 1 \tag{6}$$

In order to keep the radiation hazard insignificant, the value of H_{ex} index must not exceed the limit of unity.

The maximum value of H_{ex} equal to unity corresponds to the upper limit of Ra_{eq} 370 $Bqkg^{-1}$ measured dimensions and calculated densities.

RESULTS AND DISCUSSION

The activity concentration of Ra^{226} , Th^{232} , K^{40} and Ra_{eq} in lignite samples have been calculated and presented in the tables 1 and the comparison of the mean values of the Ra²²⁶ Th^{232} , and K^{40} activity of the lignite samples with the data presented by International Atomic Energy Report 419 (IAEA, 2014) are reported in the Table 2. The specific activities of the natural radionucleides of Ra^{226} , Th^{232} , and K^{40} are determined by using the gamma- ray spectroscopy in different lignite samples are reported with the associated uncertainties and are presented in the Table 1. The mean specific activity concentrations in the investigated lignite samples are found to be $45.5\pm2.1 \ Bqkg^{-1}$ for Ra²²⁶, $21.4\pm0.9 \ Bqkg^{-1}$ for Th²³², $40.5\pm2.1 \ Bqkg^{-1}$ for K⁴⁰ and $78.95\pm1.63 \ Bqkg^{-1}$ for Ra_{eq}. The observed average values of Ra²²⁶, Th²³² are higher than the data presented by the German, UK, USA and Indian coal. The radiation Hazard indices Hex, Dose in air (nGy/h), Annual effective dose equivalent (mSv/y), and Gamma Radiation Hazard Index (I_{γ}) determined for different lignite samples is presented in the Table 3.

Conclusion

The measurement of Activity Concentration of Ra^{226} , Th^{232} , and K^{40} in the lignite samples lignite belt of Nichahoma, Kupwara District of Kashmir Valley India was made using gamma ray spectroscopy. The average value of absorbed dose rate in air due to lignite was found to be 44.7 nGy/h. The specific activity concentration of Ra^{226} and Th^{232} was found to be higher than the Indian coal mines, the data presented by Germany, USA, and UK. The radiation hazard indices determined are well below the limits as recommended by the UNSCEAR.

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