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Evaluation of excess life time cancer risk due to natural radioactivity of the Lignite samples of the Nichahoma, lignite belt, North Kashmir, India

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Abstract: Naturally occurring radionuclides of the ^{226}Ra, ^{232}Th, and ^{40}K present in the lignite samples was measured by using a low-background Pb-shielded gamma spectroscopic counting assembly utilizing NaI(Tl) detector for the measurement and to evaluation the radiation hazard indices and excess life time cancer risk. The average values of specific activity concentrations in the investigated lignite samples was found to be 45.36 Bq kg^{-1} for ^{226}Ra, 21.42 Bq kg^{-1} for ^{232}Th, 40.51 Bq kg^{-1}for ^{40}K and 79.11 Bq kg^{-1}for Ra_{eq} respectively. The average value excess life time cancer risk was found to be relatively higher than the world average. Moreover, the correlation analysis shows the strong dependence of excess lifetime cancer risk on measured dose and the radium equivalent activity.

Keywords: Excess life time cancer risk, dose rate, correlation analysis and activity concentration.

1 Introduction

Human beings are always exposed to background radiation that arises both from natural and man-made sources. Natural radioactivity is widespread in the earth's environment and they exist in various geological formations such as rocks, earth crust, plants, water and air [1]. When an ionizing radiation passes through a living tissue, it deposits energy in the tissue randomly and rapidly via excitation and ionization, in turn produces moving electrons. These electrons interact with atoms and molecules leading to chemical and molecular changes thereby altering the structure of the cells. These cells may be damaged directly by the radiation or indirectly by the free radicals (OH and H) produced in the adjacent cells. Many forms of damage could occur from radiation but the most important is that done to the deoxyribonucleic acid (DNA). A damage to the DNA results in gene mutation, chromosomal aberration, breakages or cell death, oncogenic transformation and acute radiation sickness. More frequently, repairs can take place. This however depends on the condition that the damage is not a lethal damage. If repair is not perfect, it may result in a genetically modified cell. When human cells in an organ or tissue are killed or prevented from reproducing and functioning normally, there will be loss of organ function. A modified germ cell for instance in the gonads of an individual may transmit incorrect hereditary information, which may cause severe hereditary effects. Exposure to ionizing radiation over extended period is known to result in non-leather mutation, which could increase the risk of cancer [2]. There is a linear, no-threshold (LNT) relationship between radiation dose and the occurrence of cancer. This dose-response hypothesis suggests that any increase in radiation dose, no matter how small, could results in an increase in cancer risk [3]. Diseases caused by radioactivity exposure include lung cancer, pancreas, hepatic, skin, kidney cancers, cataracts, sterility, atrophy of the kidney and Leukemia [4].

A radiation-induced cancer can develop from a single damaged cell independently of other damaged cells in the tissue of interest. The period between radiation exposure and the detection of cancer is known as the latent period and could be many years. Therefore, excess lifetime cancer risk is the probability that an individual will develop cancer over his/her lifetime of exposure. Our initial study by Mudasir Ashraf et al. [5] looked at the radiological health assessment due to gamma radiation levels of nat-

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ural radioactivity of soil in the vicinity of Nichahoma lignite belt, Kashmir Valley. This particular study is focused mainly on the health hazard indices and the absorbed dose to the radionuclide concentration in the surface soil of the lignite belt and in the soil of the villages surrounding the lignite belt.

Lignite, also known as brown coal, is a low grade form of coal containing relatively high moisture and low energy. Toxic metals such as cadmium, lead, chromium, selenium, nickel, vanadium, copper, sulphur and fluorine as well as radioactive elements such as uranium, thorium and radium and their progenies, have been identified as potential risks in mining operations. All these agents, singly or in combination, could cause major detrimental health effects in the miner and to the human population living in the vicinity of the mines. The major lignite deposits in the valley of Kashmir occur in the vicinity of Nichahoma, Chowkibal, Budhasheng, Lanylab, Shaliganga, Raithan and Tangmarg. The lignite samples under investigation were collected from Nichahoma locality, Handwara. This lignite seam exists in the Karewa formations, right from Nichahoma to Lolab. These deposits are associated with clays and loams. The report of Geological Survey of India envisages that there are lignite coal deposits of about 5.6 crore tons in the valley. Drilling operation was started first in the Nichahoma and Chowkibal areas and reserves are estimated at 4.5 million tons to depth of 40 m [6].

Since the dawn of the universe natural environmental radioactivity of terrestrial nature are present in the earth. The levels of these environmental radiations of terrestrial nature, depends on the geological, soil types and geographical conditions. The natural gamma radiation levels may vary considerably from one type of soil to another. The higher concentrations of radionuclides in the earth' s crust such as ^{238}U, ^{232}Th and ^{40}K, which occur in minerals, such as monazites, zirons and in marble baring area. The distribution of these radionuclides in the earth's crust depend on the distribution of rocks and soil from which they originate and the process which solidifies them [7, 8]. There are few regions in the world, which are known for high background radiation areas (HBRAs), are due to local geology, geochemical processes which cause enhanced levels of background radiations [9].

The aim of the present study is to measure the natural environmental radiation levels, know the distribution of minerals which serve as the source of these radionuclides. The importance of these studies are understood when considering the radiation exposure to human being only, the external radiation exposure on the earth's surface is mainly caused by ^{238}U, ^{232}Th and ^{40}K in soil [10, 11]. The natural radiation levels under certain conditions can reach radiological hazard levels. Therefore, it is felt necessary to study the natural radioactivity in soil and to assess the exposure levels to the population in order to know the health risks with special emphasis to the excess life time cancer risk and to have a baseline data to carry extensive research in the field of radiological investigations in future. The areas that are covered in the present study are the samples of lignite belt of Nichahoma, Kupwara, North Kashmir, India.

2 Geological outline of the study area

The study region has quartz veins carrying sulphides of copper and iron with some oxide, carbonates and arsenides. The presence of gold and silver in traces is indicated in the quartz veins carrying sulphides of copper and iron in association with some oxides, carbonates and arsenides in Lolab area of Jammu and Kashmir region. The upper Cambrian region of Kupwara in the northwest Kashmir contains several thick bands of hard and siliceous recrystallised limestone, which is currently being mined and marked as "Kupwara Marble". The estimated lignite reserves at Nichahoma are found to be 80 million tons. This lignite occurs in a track which is around 80 km long and 16 km wide that has lignite showing rapid variation in thickness and quality. The geological survey of India proved 4.5 million tons reserves of lignite up to a depth of 36.5 m in Nichahoma area. The Indian Bureau of Mines indicated proven reserves of 7.26 million tons. The quarriable reserves in Nichahoma area were estimated to be 5.26 million tons [6].

3 Material and methods

3.1 Sample preparation and radiometric analysis

The radiometric analysis for the radioactivity concentrations of ^{226}Ra, ^{232}Th, and ^{40}K in the lignite samples collected from the Nichahoma, lignite belt was performed by using the gamma-ray spectrometer consisting of a NaI(Tl) detector (crystal size 40.0 mm × 60.0 mm) connected to 1024 channel multichannel analyser (MCA). Before measurement, the samples were pulverized, heated and dried in an oven at a temperature of 125 °C for 24 h so as to make them moisture and sieved through a 2 mm mesh. 1000 g of samples was filled and sealed in leak-proof, air tight PVC

merinelli beakers, weighed and stored for a period of four weeks to enable the samples to attain a state of secular equilibrium, where the rate of progeny becomes equal to that of the parent (^{226}Ra and ^{232}Th) [12] and the system is calibrated using ^{137}Cs and ^{60}Co radioactive sources produce γ-ray energies of 662 KeV, 1173 KeV and 1332 KeV, respectively.

The spectrum was analysed by Leybold Cassy Lab Multi-Channel Analyser model Pocket- CASSY 559901 (Germany made). The activity of ^{40}K was estimated directly with 1460.7 (10.7%) keV peak of the gamma ray spectrum. To determine the activity concentration of ^{226}Ra, the average value of gamma ray energies 295.1 (19.2%) and 351.9 (37.1%) keV from ^{214}Pb to 609.3 (46.1%) and 1764.5 (15.9%) keV gamma ray from ^{214}Bi are used. The activity concentration of ^{232}Th was determined using the average value of gamma rays peaks 238.6 (43.6%) keV from ^{212}Pb, 338.4 (12%), 911.1 (29%) and 968.9 (17.4%) keV from ^{228}Ac, 583.1 (86%) and 2614 KeV from ^{208}Tl [13]. Each sample was examined for 18 000 s. The activity concentrations in the lignite samples were calculated according to the following relation [14, 15]:

$$A = \frac{C}{\varepsilon \times P_{\gamma} \times M_{s} \times T} \quad (1)$$

Where C is the count rate of gamma rays, ε is the detectors efficiency of the specific γ-rays, P_{γ} is the absolute transition probability of the γ-decay, M_{s} is the mass of the sample in kg, and T is the counting time in seconds obtained for the measured radionuclides and are expressed in Bq kg^{-1} per dry weight.

4 Radium equivalent activity

The radium equivalent activity, Ra_{eq}, was introduced to identify the uniformity to radiation exposure. The calculated values of Ra_{eq} were generally used to compare the specific activity of materials containing different amounts of activity concentration ^{238}U, ^{232}Th, and ^{40}K. Besides, Ra_{eq} data can be used to assess the health hazard effects produced from the activity concentrations of ^{238}U, ^{232}Th, and ^{40}K radionuclides in the lignite samples. The measured values of Ra_{eq} were obtained by making use of the following equation [9, 11].

$$Ra_{eq}(\text{Bq kg}^{-1}) = A_{Ra} + 1.43A_{Th} + 0.077A_{K} \quad (2)$$

Where A_{Ra}, A_{Th}, and A_{K} are the specific activities of ^{226}Ra, ^{232}Th, and ^{40}K respectively in Bq kg^{-1}. It is calculated based on the assumption that 370 Bq kg^{-1} of ^{226}Ra, 259 Bq kg^{-1} of ^{232}Th and 4810 Bq kg^{-1} of ^{40}K produce an equal gamma ray dose rate. Radium equivalent activity is directly related to the external and internal gamma dose due to radon and its progenies [12].

4.1 Absorbed dose rate in air

In order to assess any radiological hazard, the exposure to radiation arising from radionucleides present soil can be estimated in terms of many parameters. A direct connection between radioactivity concentrations of natural radionucleides and their exposure rate is known as the absorbed dose in the air at 1 meter above the ground surface. The mean activity concentrations of ^{226}Ra (of the ^{238}U series), ^{232}Th, and ^{40}K (Bq kg^{-1}) in soil samples are used to calculate the absorbed dose rate given using the following formula provided by United Nations Scientific Committee on Effects of Atomic Radiations [4] and European Commission [13]. UNSCEAR and the European Commissions have provided the dose conversion coefficients for the standard room centers.

$$D(\text{nGy h}^{-1}) = 0.462A_{Ra} + 0.604A_{Th} + 0.0417A_{K} \quad (3)$$

Where D is the absorbed dose rate in nGy h^{-1}, A_{Ra}, A_{Th}, and A_{K} are the activity concentration of ^{226}Ra (^{238}U), ^{232}Th and ^{40}K, respectively.

4.2 Annual effective dose equivalent

The annual effective dose equivalent (AEDE) received by individuals was estimated from the calculated values of absorbed dose rate by applying the dose rate conversion factor of 0.7 Sv Gy^{-1} and the occupancy factors of 0.2 (5/24) and 0.8 (19/24) for outdoors and indoors, respectively [14]. The annual effective outdoor doses, D_{out}; the annual effective indoor doses, D_{in}; and total annual effective doses, D_{tot}, were calculated according to the following equation [15].

$$D_{out}(\text{mSv y}^{-1}) = D_{r}(\text{mGy h}^{-1}) \times 24\,\text{h} \times 365.25\,\text{d} \times 0.2 \times 0.7\,\text{Sv Gy}^{-1} \times 10^{-6} \quad (4)$$

$$D_{in}(\text{mSv y}^{-1}) = D_{r}(\text{mGy h}^{-1}) \times 24\,\text{h} \times 365.25\,\text{d} \times 0.8 \times 0.7\,\text{Sv Gy}^{-1} \times 10^{-6} \quad (5)$$

$$D_{tot}(\text{mSv y}^{-1}) = D_{out} + D_{in} \quad (6)$$

4.3 External and internal hazard indices

To limit the radiation exposure attributable to natural radionuclides in the samples to permissible dose equivalent limit of 1 mSv y^{-1}, the H_{ex} index based on a criteria have been introduced using a model Krieger which is given by [4, 16]:

$$H_{ex} = \frac{A_{Ra}}{370} + \frac{A_{Th}}{259} + \frac{A_K}{4810} \leq 1 \quad (7)$$

In order to keep the radiation hazard insignificant, the value of H_{ex} 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 Bq kg^{-1} [16, 17] measured dimensions and calculated densities.

In addition to H_{ex}, radon and its short lived products are also hazardous to the respiratory organs. The H_{in} due to internal exposure to radon and its daughter products is quantified by H_{in}, which is given by the following equation as [13]:

$$H_{in} = \frac{A_{Ra}}{185} + \frac{A_{Th}}{259} + \frac{A_K}{4810} \leq 1 \quad (8)$$

where A_{Ra}, A_{Th} and A_K are the concentration in Bq kg^{-1} of ^{226}Ra, ^{232}Th and ^{40}K respectively.

4.4 Excess lifetime cancer risk

The excess lifetime cancer risk (ELCR) values are calculated using the equation [14, 18].

$$ELCR = D_{in} \times D_L \times R_F \quad (9)$$

Where D_L is the duration of life (approximately 70 years), and R_F is the risk factor (Sv^{-1}), which reflects the fatal cancer risk per Sievert. For stochastic effects, ICRP 60 uses values of 0.05 for the public [19].

5 Results and discussions

Table 1 presents the estimated values of the specific activity concentration of the lignite samples of the study area and the comparison of their mean values of the ^{226}Ra, ^{232}Th, and ^{40}K activity of the lignite samples with different scientific groups across the world is presented in the Table 2. The mean values of specific activity concentrations in the investigated lignite samples are found to be 45.36 Bq kg^{-1} for ^{226}Ra, 21.42 Bq kg^{-1} for ^{232}Th, 40.51 Bq kg^{-1} for ^{40}K and 79.11 Bq kg^{-1} for Ra_{eq}. The observed average values of ^{226}Ra, ^{232}Th are relatively higher than the data presented by the German, UK, USA, and Indian coal. Also, the ^{40}K activity concentration of the samples under investigation was found to be higher than the activity concentration of the Drage Mine, Bosinia. The radiation hazard indices H_{ex}, H_{in}, dose rate (nGy h^{-1}), annual effective dose equivalent (mSv y^{-1}) and excess life time cancer risk are estimated for the investigated samples are presented in the Table 3.

Frequency histograms and the associated distribution curves for ^{226}Ra, ^{232}Th and ^{40}K are presented in the Fig-

Table 1: The activity concentration of the ^{226}Ra and ^{232}Th series and ^{40}K (Bq kg^{-1}) of the studied lignite samples.

Sample Code	Specific activity concentration (Bq kg^{-1})			Ra_{eq} (Bq kg^{-1})
	^{222}Ra	^{232}Th	^{40}K	
L-1	40.4	17.4	37.4	68.16
L-2	43.4	19.8	36.6	74.53
L-3	47.1	21.3	38.2	80.50
L-4	42.9	18.6	39.6	72.55
L-5	49.9	19.5	40.7	80.92
L-6	40.3	23.4	31.8	76.21
L-7	50.6	22.6	42	86.15
L-8	41.6	24.1	43.7	79.43
L-9	53.7	25.9	46.2	94.29
L-10	43.7	21.6	48.9	78.35
Mean	45.36	21.42	40.51	79.11
Standard Deviation	4.69	2.65	4.97	7.29
Minimum	40.3	17.4	31.8	68.17
Maximum	53.7	25.9	48.9	92.29

Table 2: The comparison of average value of concentration of the natural radionucleides in lignite samples (Bq kg^{-1}) reported for different parts of world.

Mine and country	Specific activity concentration (Bq kg^{-1})			Reference
	^{226}Ra	^{232}Th	^{40}K	
Drage Mine, Bosinia	1191.34±4.83	26.67±1.86	32.92±7.20	[26]
Table Mine, Herzegovina	263.33±2.43	11.54±1.04	137.64±6.06	[26]
Indian Coal Mines	16–27	8–27	50–100	[27]
Fly-Ash India	200–150	50–150	250–700	[9]
Australia	19–24	11–69	23–140	[27]
Brazil	72	62	–	[27]
Germany Lignite	32	21	225	[27]
Greece Lignite	44–206	–	–	[27]
Romania	126	62	–	[27]
UK	8–22	7–19	55–314	[27]
USA	9–59	4–21	–	[27]
Nichahoma Kupwara, Kashmir, India	45.36	21.42	40.51	Current study

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

D (nGy h^{-1})	E_T (mSv y^{-1})	H_{ex}	H_{in}	ELCR × 10^{-3}
30.74	0.04	0.18	0.29	0.53
33.55	0.04	0.20	0.32	0.58
36.22	0.05	0.22	0.34	0.62
32.71	0.04	0.20	0.31	0.56
36.53	0.04	0.22	0.35	0.63
34.08	0.04	0.21	0.31	0.59
38.78	0.05	0.23	0.37	0.67
35.60	0.04	0.21	0.33	0.61
42.40	0.05	0.25	0.40	0.73
35.28	0.04	0.21	0.33	0.61
Mean = 35.58	Mean = 0.044	Mean = 0.21	Mean = 0.34	Mean = 0.61
Standard Deviation = 3.3	Standard Deviation = 0.004	Standard Deviation = 0.019	Standard Deviation = 0.031	Standard Deviation = 0.05
Maximum = 42.34	Maximum = 0.5	Maximum = 0.25	Maximum = 0.4	Maximum = 0.73
Minimum = 30.73	Minimum = 0.5	Minimum = 0.18	Minimum = 0.29	Minimum = 0.53

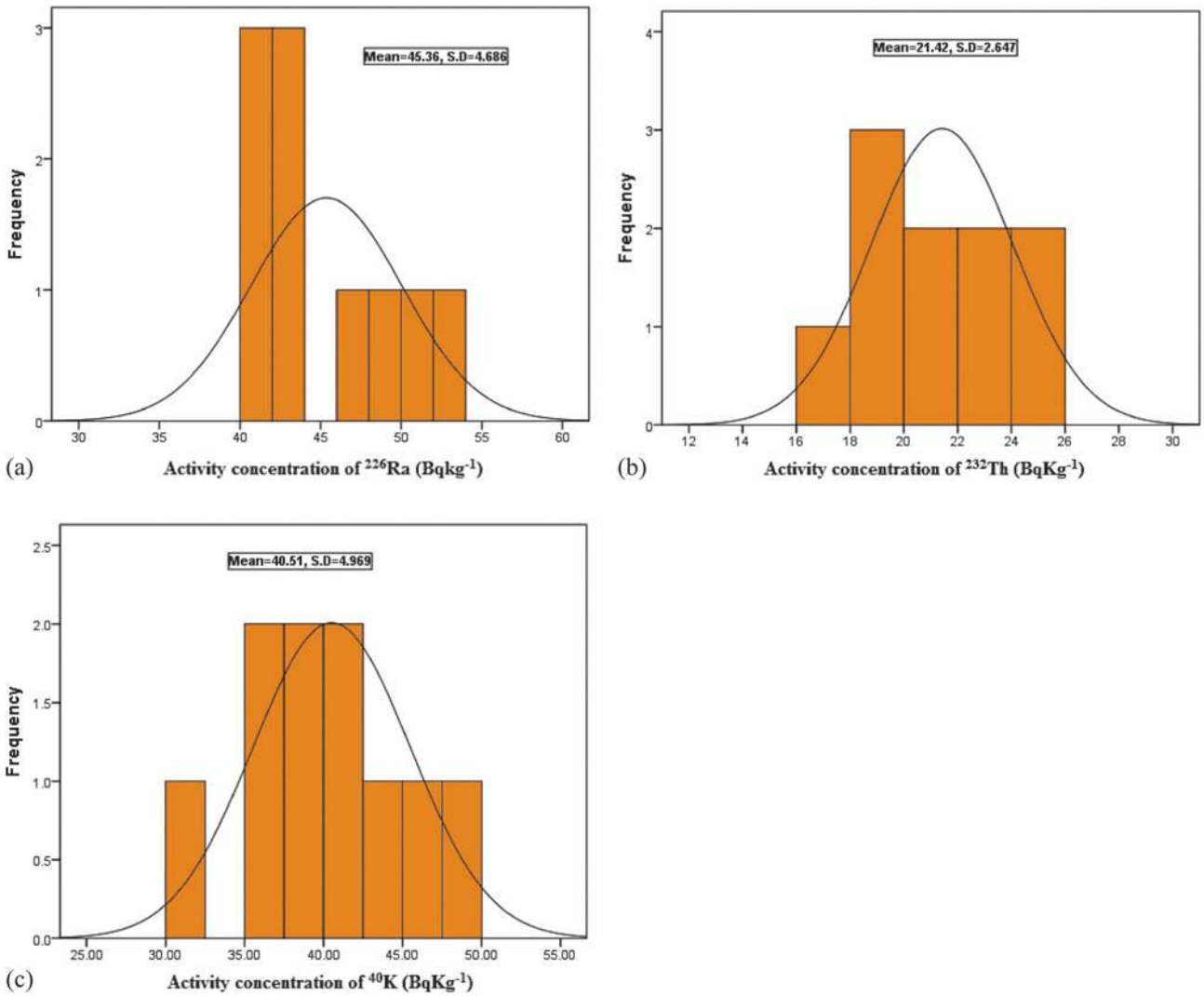

Figure 1: Frequency distribution of the ^{226}Ra, ^{232}Th and ^{40}K concentrations of the lignite samples.

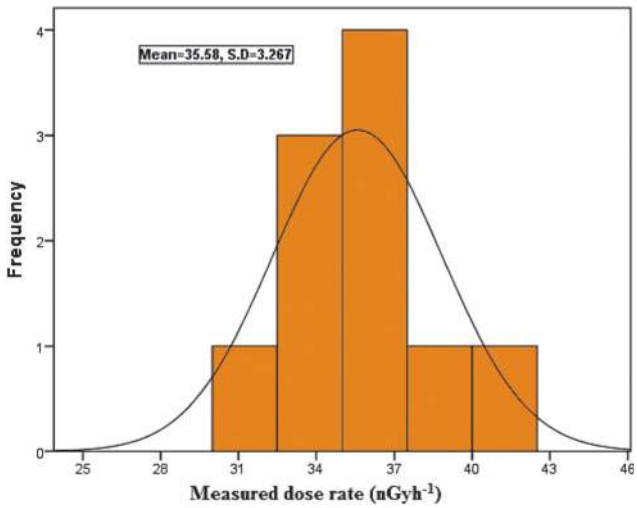

Figure 2: Frequency distribution of the absorbed dose.

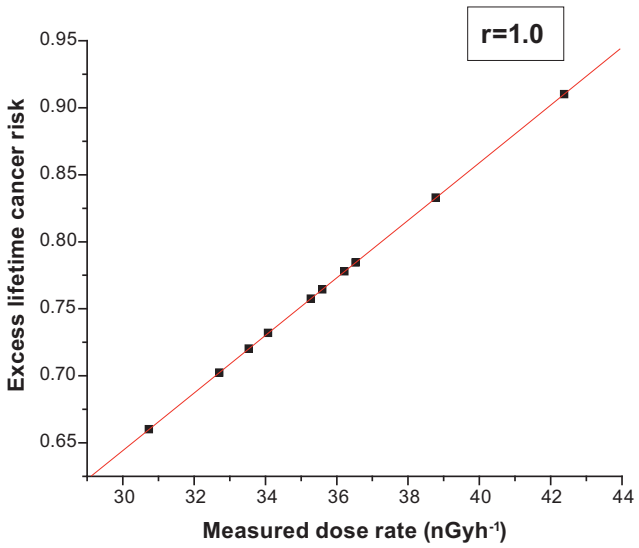

Figure 3: Correlation between excess life time cancer risk and the measured dose rate.

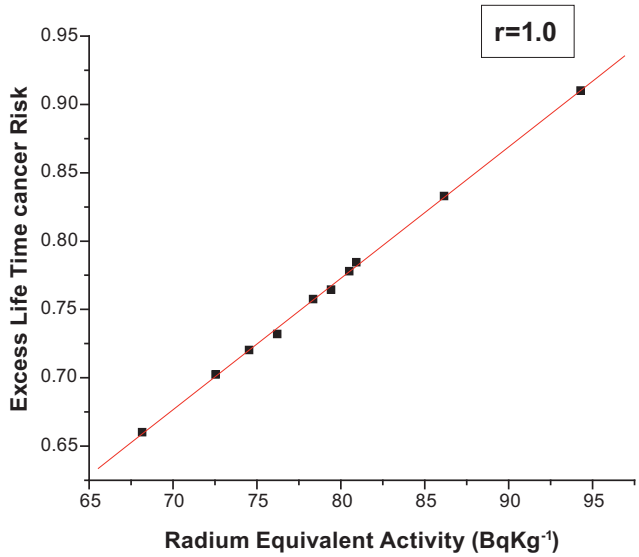

Figure 4: Correlation between excess life time cancer risk and radium equivalent activity.

ure 1. The frequency distribution for the absorbed dose rate for the samples under investigation is provided in the Figure 2. The activity and absorbed dose rate fitted to normal distribution with an asymmetric curve indicating its dominance in particular samples of the lignite belt.

Radiation induced cancer being a stochastic effect may appear only some years after the irradiation and the probability of occurrence increases with increasing absorbed dose and there is no threshold. Radium being a solid radioactive element is chemically analogous to calcium, and is absorbed from soil by plants and passed up the food chain to humans. Microscopic quantities of radium in the environment can lead to some accumulation of radium in bone tissue whereby it degrades bone marrow and can mutate bone cells. Ingestion or body exposure to radium causes serious health effects which included sores, anemia, bone cancer and other disorders. ^{226}Ra is a product of ^{238}U decay series. Emitted energy from the decay of radium causes vexed on the skin and produces many other detrimental effects. Radium is a naturally occurring radioactive metal moreover it is present in soil, sand, rock, water, plants and animals. Higher values of radium in sand contribute significantly in the enhancement of indoor radon in dwellings [20]. Radium is one million times more radioactive than the same mass of uranium. Its decay occurs in at least seven stages, the following main products were called radium emanation recognized as radon. Radon is a heavy gas and the later products are solids. These products are themselves radioactive in nature. Radon is the first leading cause of lungs cancer among non smokers and second leading cause in smokers.

The excess life time cancer risk is determined and presented in Table 3. The values of excess lifetime cancer risk range from 0.7×10^{-3} to 0.5×10^{-3} with an average of about 0.61×10^{-3}. In order to find the dependence of excess life time cancer risk on the activity concentration and the dependence of excess life time cancer risk with other radiological hazard indices, correlation analysis was performed between the Ra_{eq} and excess life time cancer risk and measured dose rate and excess life time cancer risk Figures 3 and 4. A very strong correlation was observed between the Ra_{eq} and excess lifetime cancer risk ($r = 1.0$) and measured dose rate and excess life time risk of cancer ($r = 1.0$). The average value of excess life time risk of can-

cer in the present study is higher than the world average of 0.29 [28]. There is no safe limit and all doses of radiation carry some form of risk. Hence radiation induced cancer and life time cancer cannot be prevented, only can be reduced by minimizing the radiation dose.

6 Conclusion

Radiation carcinogenesis is a complex phenomenon involving the process of initiation fallowed by a long latent period before the onset of cancer. Furthermore, sensitivity to induction varies enormously with the type of the tissue or organ involved. The mechanism of induction of carcinogenic lesions is not well understand and is still a challenge among the scientific community.

The radiometric analysis of lignite samples of the Nichahoma lignite belt was performed by using a NaI(Tl) gamma ray spectrometer with an intention that the present study will serve as the baseline data for carrying out the extensive research in the area surrounding the lignite belt and its impact on the residents of this rural area. The average value of absorbed dose rate in air due to lignite was found to be 35.58 ($nGy\,h^{-1}$). The specific activity concentration of ^{226}Ra and ^{232}Th was found to be relatively higher than the Indian coal mines, the data presented by Germany, USA, and UK. Further, the ^{40}K is higher than the activity concentration of Drage Mine, Bosinia. The excess life time cancer risk estimated for the investigated lignite samples was found to be relatively higher than the world average of 0.29. Moreover, the correlation analysis performed shows the strong linear dependence of excess life time cancer risk on the Ra_{eq} ($Bq\,kg^{-1}$) and the estimated dose rate ($nGy\,h^{-1}$) from the investigated lignite samples.

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