Review on studies of radioactivity in soil

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Abstract: Natural radioactivity is a source of continuous exposure to human beings. It originates from both extraterrestrial sources and radioactive elements in the earth's crust. The amount of radioactivity in soil varies widely. The main radionuclides contaminant in soil are mainly from ^{238}U and ^{232}Th decay series and ^{40}K. Activity concentrations of soil samples are normally measured by means of high-resolution gamma-ray spectrometry. Several studies on natural radioactivity in soil as well as building materials has been done and various observations were reported. In this article, a review studies on natural studies in soil was carried out.

Keywords: Natural radiation, soil analysis, radioactivity, radiation dose rate

INTRODUCTION

External gamma radiation exposure to public arises from terrestrial radionuclides present at trace levels in all soil types. The specific levels of a particular radionuclide are related to the types of rock from which the soils originate. Igneous rocks, such as granite are usually showing higher radiation levels as compared with sedimentary rocks. However, some shale's and phosphate rocks have relatively high radionuclide content as some exceptions [1]. These natural radionuclides in soil could pose potential health hazard [2-4]. The assessment of gamma radiation dose from natural sources is of particular importance as natural radiation is the largest contributor of the external dose to the world population [2,3-4]. There have been many radiological surveys to determine the background levels of radionuclides in soils [5-9]. As per UNSCEAR, 2000 report [10], there exist about 340 naturally occurring nuclides, of which about 70 are radionuclides and, are found mainly among the heavy elements, with the mass number (A) >200. Extensive studies of the background exposure levels in different countries indicates that the three main radionuclides namely ^{238}U and ^{232}Th series and ^{40}K make approximately equal contributions to the externally incident gamma radiation dose to individual in both outdoors and indoors [4]. 98.5% of the radiological effects in the ^{238}U series are produced by radium and its daughter radionuclide products, so radium and its decay

radionuclides are radiologically very important [11]. Radon, thoron, and their progenies contribute the maximum natural radiation dose to occupational workers and general public [12]. Radon and Thoron concentration are expected to be more in indoors than outdoors. Radon and Thoron exhalation is associated with the presence of ^{226}Ra and ^{232}Th in soil as well as in building materials. This naturally occurring Radon and Thoron gas is considered the second commonest cause of lung cancer [13,14]. Studies of natural radioactivity are necessary not only because of their radiological impact to public, but they also act as excellent biochemical and geochemical tracers in the environment. U-series radionuclides present in nature have been of particular interest due to their relatively high biological mobility [1]. Although natural radioactivity is available through the earth, the accession in specific areas varies relatively within narrow limit [15]. Hence, monitoring natural radioactivity in natural soil becomes important to evaluate any possible risk to human health and develop any precautionary measures. This review work aims to survey literatures on radiometric analysis of soil.

Sample preparation

Soil samples of most studies on radioactivity are collected and crushed into small sizes [16-19]. The soil samples are then treated in a hot air oven at 110°C for 24 hr or a suitable temperature and duration. After that, the soil samples are ground into a fine powder, homogenized, and sieved through appropriate mesh sizes. Then, samples are packed in suitable air tide plastic containers to restrict the escape of radon gas from the packed. The processed samples are then stored carefully for about one month to achieve the equilibrium condition of ^{226}Ra and ^{232}Th along with their respective daughter nuclides [17-19].

Radiological analysis of soil

The prepared Soil samples from the study areas are then sieved through a mesh size of about 0.45 mm [16-19], approximately 250 g of this homogeneous fine mesh of each soil sample is then packed inside a plastic container with predefined geometry, weighed, and properly sealed to restrict the escape of radon gas from the packed. The processed soil samples are then stored carefully for about one month to achieve the equilibrium condition of ^{226}Ra and ^{232}Th along with their respective daughter nuclides [17-19]. A suitable scintillation detector (e.g. 3" x 3" NaI(Tl)) or Germanium-based gamma spectrometer is employed with adequate shielding [19-21]. Measurements of the radiation emitted from soil samples are normally performed using calibrated standard source samples, which contain a known activity of gamma-ray emitters radionuclides, namely ^{133}Ba (356.1 keV), ^{137}Cs (661.6 keV), ^{60}Co (1173 KeV and 1332 KeV), and ^{226}Ra (1764.5 keV) [13-14, 16, 22-24]. All soil samples are then subjected to gamma spectral analysis with a suitable counting time of e.g. 36,000 s [25]. The radioactivity concentration of ^{226}Ra is determined from the average activity concentration obtained from the prominent gamma lines of ^{214}Bi (1.76 MeV) and ^{214}Pb (0.35 MeV) and that of ^{232}Th is estimated from the average concentration obtained from the gamma lines of ^{212}Bi (0.73 MeV), ^{228}Ac (0.91 MeV), and ^{208}Tl (2.61 MeV) respectively. However, ^{40}K is evaluated from its gamma photo-peak (1.46 MeV) [16,19-20]. The activity concentrations of ^{226}Ra, ^{232}Th series, and ^{40}K are calculated using the following equation [22]

$$A \left(\frac{Bq}{Kg} \right) = \frac{N}{\epsilon \beta M} \qquad (1)$$

where, N = the net gamma counting rate (counts per second), ε = the detector efficiency of the specific gamma-ray, β = the absolute transition probability of gamma decay, and M = the mass of the sample (kg).

The relative concentration and distribution of ^{226}Ra, ^{232}Th, and ^{40}K are usually not uniform in the soil materials. Hence, Radium equivalent (Ra$_{eq}$) is the most commonly used single quantity to represent natural radioactivity associated with those soil materials containing ^{226}Ra, ^{232}Th, and ^{40}K. Estimation of the radioactivity concentration is made on the assumption that 370.0 Bq/kg of ^{226}Ra, 259.0 Bq/kg of ^{232}Th, and 4810.0 Bq/kg of ^{40}K produce similar gamma dose rates as mentioned by the following equation [26].

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

where, A_{Ra}, $A_{Th,}$ and A_K are the specific activities of ^{226}Ra, ^{232}Th, and ^{40}K in Bq/kg, respectively.

External exposure index (EEI), formerly used term "Hazard index," which indicates the level of exposure from the natural gamma radiation. The value of EEI, when exceeds unity, it does not cause any significant exposure which might cause adverse health effects. To cause any adverse health effects, the external exposure index value should approximately exceed about a million. Thus, the use of the word "Hazard" in exposure indices may be considered inappropriate for ambient background exposures as suggested by Rao [27]. It is evaluated by using the following equation given as:

$$\text{EEI} = {}^{226}\text{Ra}/370 + {}^{232}\text{Th}/259 + {}^{40}\text{K}/4810 \qquad (3)$$

The maximum value of EEI should be lower than unity [7].

The external Absorbed dose rate (nGy/h) due to terrestrial gamma radiation at 1 m above the ground surface was evaluated from the activity concentration of ^{226}Ra, ^{232}Th, and ^{40}K present in the soil using the Monte Carlo method given by the UNSCEAR, 2008[28] as:

$$\text{Dose (nGy/h)} = 0.462C_{Ra} + 0.604C_{Th} + 0.0417\ C_K \qquad (4)$$

Where C_{Ra}, C_{Th}, and C_K are the activity concentrations in Bq/kg for ^{226}Ra, ^{232}Th, and ^{40}K, respectively.

The annual effective dose (D$_{eff}$) is calculated by using the dose conversion factor of 0.7 Sv/Gy from the absorbed dose in air received by an adult and a value of 0.8 as an indoor occupancy factor [10]. The D$_{eff}$ due to gamma radiation from soil materials was evaluated as

$$D_{eff}(mSv/y) = D(nGy/h) \times 8760(h/y) \times 0.8 \times 0.7(Sv/Gy) \times 10^{-6} \qquad (5)$$

Discussions

The radioactive contaminants in the soil are the main sources of gamma radiation. These radionuclides include radioactive decay chains headed by ^{238}U,^{235}U, and ^{232}Th, as well as ^{40}K,^{87}Rb, and other radioactive isotopes. The percentage of ^{235}U present in soil is relatively small amounts (0.7%) compared to ^{238}U (99.3%) and therefore is not considered further. Most natural radioactivity researchers in the world worked in the field of analysing soil and rock samples to show the natural radioactivity levels and associated radiological hazards in the environment by using gamma-rays' spectroscopy system since most soil and rock samples contain naturally occurring radioactive elements. The two abundant natural radioactive series, which can be represented by the isotopes ^{238}U, ^{232}Th, and primordial radionuclide ^{40}K, are the most important radioisotopes. The presence of these radioisotopes contaminant in the rocks and soils causes external and internal exposure to the people. The Radon and Thoron from ^{238}U

and ^{232}Th series can also enhance the concentration of these radioactive gaseous and its radionuclide daughters in the house.

Radioactive contaminants of ^{226}Ra, ^{232}Th, and ^{40}K in soil of some countries and their respective annual absorbed dose rate, effective dose rate, radium equivalent and external exposure index are shown in Table 1. Countries such as Egypt (southern part) [29], Istanbul, Turkey [30], Algeria [33], coast of Bushehr, Iran [34], Al- Rakkah, Saudi Arabia [35] are showing lesser annual absorbed dose rate, effective dose rate, Radium equivalent as well as EEI as compared with world average report (UNSCEAR, 1993, 2000) [3,10].

The study of natural radioactivity levels in environmental rock samples from Taiz, Yemen, [32] showing mean activity concentrations of ^{226}Ra, ^{232}Th, and ^{40}K to be 65.58 ± 1.38, 82.93 ± 0.93, and 976.40 ± 6.11 Bq/kg, respectively. These observed values exceed the maximum international reference values (UNSCEAR, 2000) of 35, 30, and 400 Bq/kg, respectively [10].

Similarly, building materials of Pakistan [36] and soil samples in China [37] were studied and average radioactivity concentrations of ^{226}Ra, ^{232}Th, and ^{40}K were found higher than world reference values (UNSCEAR, 2000) of 35, 30, and 400 Bq/kg, respectively [10].

Natural radioactive contaminants of ^{226}Ra, ^{232}Th, and ^{40}K in soil of some parts of India and their respective annual absorbed dose rate, effective dose rate, radium equivalent and external exposure index are shown in Table 2. Radium equivalent, Annual absorbed dose rate, effective dose rate and external exposure index are all higher than world respective references [3,10].

The natural radioactivity in soil samples collected from some areas of Himachal Pradesh, India, were measured using gamma-ray spectrometry. The results of the radioactivity concentrations of ^{226}Ra and ^{232}Th in soil samples collected from these regions are higher and ^{40}K is lower than the world average value. The annual effective exposure dose rate has been determined from the content of these radionuclides in soil and found in the range of 0.07–0.13 mSv [46].

The natural radioactivity levels in beach sediments collected from the northeast coast of Tamil Nadu, India, were determined using gamma-ray spectrometry [47]. The mean activity concentrations of ^{238}U, ^{232}Th, and ^{40}K and radiation hazard indices were evaluated, and it was found that all the values were lesser than the worldwide average values. On the basis of this lower levels of natural radioactivity, beaches of the northeast coast of Tamil Nadu in India may be considered as a low natural background radiation area.

The radionuclide concentration in different high-background radiation areas of coastal Kerala was measured [48]. The radioactivity study of soil samples collected from sea waterline at different depths and at different distances using gamma-ray spectrometry and no definite correlation was found between the variation of ^{232}Th and ^{226}Ra concentrations with depth. A study carried out using NaI (Tl) detector along the coastal belt of Kerala [49]. The natural radioactivity assessment showed that the concentration of ^{226}Ra and ^{40}K is well within the

permissible limit, but the concentration of ^{232}Th was found higher compared to the world and Indian average values.

Radiological investigations recently carried out in the Eastern coast of Odisha, India, to measure the radiation dose rates have revealed that there is an enhanced level of natural radiation in the area [50]. The activity concentration of ^{232}Th was found to be 2825 Bq/kg, whereas that of ^{238}U was 350 Bq/kg.

Natural background gamma radiation levels in and around Loktak Lake of Manipur, India, and radioactive contamination level due to terrestrial gamma radiation were measured using NaI (Tl) scintillator-based Micro-R-survey meter and high-purity germanium detector.[18] The averaged values of radioactivity concentrations of ^{226}Ra, ^{232}Th, and ^{40}K were reported as 74.6, 112.1, and 792.9 Bq/kg, respectively, and the average value of the annual effective dose in this study was 0.7 ± 0.1 mSv/y, which is higher than the world average value of about 0.4 mS/y reported by UNSCEAR, 2000 [10].

The distribution of radionuclides contaminant in Indian soils had been studied, and it has been reported that the terrestrial dose rates range from 18 to 144 nGy/h, and the contribution of ^{137}Cs toward the external dose is very small [51]. Whereas places in peninsular India like Hyderabad and Visakhapatnam and Chingleput located in the East coast exhibit higher concentrations of ^{232}Th compared to that of other normal areas of India. The natural radioactivity level of ^{238}U and ^{232}Th in soil samples from monazite regions in India varies up to 3400 and 15,400 Bq/kg, respectively.

Some developed countries such as Germany, China, Canada, Finland, Sweden, the USA, and the UK, have already analyzed the relationship between concentrations of radon in houses and the incidence of lung cancer. All these studies indicate that higher lung cancer rates occur in people exposed to higher levels of radon gas.

Conclusion:

Humans are always exposed to natural radionuclides present in soil. The radioactivity concentration in soils is an indicator of natural radioactive accumulation in the environment, which affects humans, animals, and plants. These natural radionuclides have a long life half-lives, often about hundreds of millions of years. Natural gamma radiation exposures depend on the location, elevated levels of NORMs in specific localized regions, and human activities and practices. Building materials of various types of houses and the design and ventilation systems strongly influence the indoor levels of radon and its decay products, which contribute to the doses through inhalation. The total contribution of radiation absorbed dose from natural radiation sources to the general population is about 2.4 mSv/y. Most of the exposures are produced by the natural radiation of radionuclides present in the soil and building materials.

Table 1: Natural Radioactivity concentration of in soil of some countries

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Country	Radioactivity concentration (Bq/Kg)			Dose$_{ad}$ (nGy/h)	D$_{eff}$ (mSv/y)	Ra$_{eq}$ (Bq/Kg)	EEI
	^{226}Ra	^{232}Th	^{40}K				
Egypt [29]	13.0	6.0	433.0	27.69*	0.17	54.92	0.15
Istanbul, Turkey [30]	21.0	37.0	342.0	46.31*	0.28	100.24	0.27
N. Sana'aYemen [31]	48.2	41.7	939.1	86.62*	0.53	180.14	0.49
Taiz,Yemen[32]	65.6	82.9	976.4	121.09	0.74	259.33	0.70
Algeria [33]	47.0	33.0	329.0	55.37*	0.34	119.52	0.32
Iran [34]	35.0	17.0	285.0	38.32*	0.23	81.26	0.22
Saudi Arabia [35]	23.0	20.0	233.0	32.42*	0.20	69.54	0.19
Pakistan [36]	46.7	60.8	698.6	87.43*	0.54	187.44	0.51
China [37]	44.0	47.0	593.1	73.45*	0.45	156.88	0.42
Bangladesh [38]	14.0	25.0	158.0	28.16*	0.17	61.92	0.17
World average [3]	32.00	45.0	420.0	59.48*	0.36	128.69	0.35

evaluated using equation (4).

Table 2: Natural radioactivity concentration in soil of some parts of India

India	Activity concentration (Bq/Kg)			Dose$_{ad}$ (nGy/h)	D$_{eff}$ (mSv/y)	Ra$_{eq}$ (Bq/Kg)	EEI
	^{226}Ra	^{232}Th	^{40}K				
India [39]	44.0	64.0	456.0	78.00*	0.48	170.63	0.46
Garhwal Himalayas [40]	131.0	384.0	1406.0	351.09*	2.15	788.38	2.13
Punjab Malwa region [41]	35.0	80.0	317.0	77.71*	0.48	173.81	0.47
North Rajasthan [42]	52.0	19.0	1627.0	103.35*	0.63	204.45	0.55
Hyderabad [43]	32.0	134.0	890.0	132.83*	0.81	292.15	0.79
Bengaluru [44]	26.2	53.1	635.1	70.66*	0.43	151.04	0.41
Imphal, Manipur [20]	94.2	146.5	1222.9	183.00	1.12	397.86	1.07
Bishnupur Manipur [45]	73.6	80.4	984.4	123.61	0.76	264.37	0.71

*Re-evaluated using equation (4).

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