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Presence of Radionuclide Concentrations in Himalayan Region of Uttarakhand, India- A Review

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Abstract: Earth is a major source of several elements which are occurring with different physical and chemical properties in nature. Some of the elements are categorized as Naturally Occurring Radioactive Materials and termed as NORMs. These natural sources and anthropogenic sources combined make the earth's environment radioactive and human beings continuously receive the amount of the total radiation dose more than 50% of the radiation that comes from radon and thoron. It has been evident that soil is the furthermost important feature that affects the radon/thoron level in the human living surroundings which increases human exposure to radioactivity. The concentration of natural radionuclides elements present in soil or rocks is the main feature of levels of natural background radiation dose. This review paper represents the activity level of Ra-226, Th-232, and K-40 in soil samples of the Himalayas region of Uttarakhand. The data so obtained from previous studies of radionuclides shows the high abundance of radioactive mineralization in the soil of the Himalayan region in the Uttarakhand state of India.
Keywords: Soil, Radionuclides, Ra-226, Th-232 and K-40, Gamma Ray Spectroscope

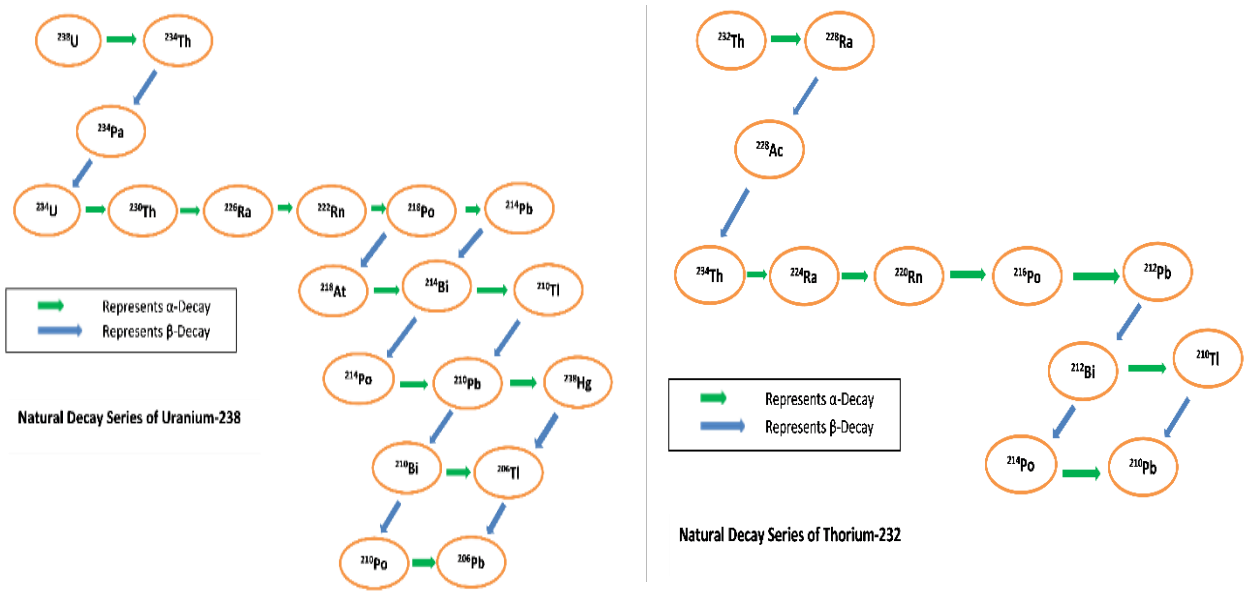
Introduction

Earth is made up of a variety of elements, each with its own set of physical and chemical properties, and some of them are radioactive elements (Krebs, 2006; Yadav et al., 2015). The earth's atmosphere is radioactive due to natural and human-made causes (Al-Khawlany et al., 2018). The toxicity due to radioactivity is a very natural occurrence all over the world for the Human. Important causes of radiation exposure include the presence of radon (Rn-222) and thoron (Rn-220) in the human atmosphere (Kaur et al., 2019). Radiation from radon and thoron accounts for more than half (50%) of the overall exposure received from humans and up to 70% of the global contamination caused by natural sources of radiation that contributes to substantial radiological research in order to estimate the effect on living being (UNSCEAR, 2000).

Rn-222 and Rn-220 are noble radioactive gases and are found as a daughter radioactive element of the U-238 and Th-232 decay sequence, which disintegrates into the stable product like Pb-206 and Pb-208, respectively, and are abundantly present in rocks/soil (Alnour et al., 2012) Fig.1 (a) and (b). The energy of alpha particles emitted during the disintegration of the U-238 and Th-232 is harmful to the human tissue and caused health

issues (Ahamad et al., 2021). The primary source of radon and thoron exhalation into the atmosphere also radio-nuclides found in soil or rocks (Semwal et al., 2018).

The main causes of human exposure to radioactive compounds are radon, thoron, and their progeny. Ra-226, Th-232, and K-40 are naturally occurring radionuclides found in soil and rocks (Ramola et al., 2008). It has been observed that a high exhalation rate is obtained in those soil samples which have a high amount of Ra-226 activity (Yadav et al., 2016). Indoor radon, thoron concentration has a significant correlation with the activity concentration of radionuclides (Kumar et al., 2021).



(a) Decay series of Uranium **(b)** Decay series of Thorium

Fig. 1: *Decay series and daughter product of (a) Uranium and (b) Thorium*

The World Health Organization has recommended the reference limit which is get reduced from 200 Bq/m3 to 100 Bq/m3 and has identified radon as the second most carcinogenic gas for humans after smoking (WHO, 2009). The majority of the radiation doses are determined by the concentration of these radionuclides in the earth's crust. Radionuclides are helpful for the estimation of Radiological Health Hazard Index (H_{ex} = external and H_{in} = internal health effects), Radium Equivalent Activity (Ra_{eq}), Absorbed dose (D), Annual effective dose (for Indoor and Outdoor), and Gamma Activity Index (I_{γ}) (Ramola et al., 2008).

The present study represents a review of the presence of Radionuclide concentrations in the Himalayan Region of Uttarakhand, India based on the past studies done in various parts

of the Uttarakhand state, India which gives strong evidence for the presence of radioactive mineralization across the state.

Environmental Radioactivity in ambient Air

Environmental radioactivity is termed as natural radioactivity in the ambient air which increases by the natural background radiation by radon (Rn-222 and Rn-220) and gamma exposure. Terrestrial background radiation, or gamma radiation released from NORMs (Naturally-Occurring Radioactive Materials), is a major indirect cause of human body radiation exposure. Natural ambient radioactivity and the resulting external gamma radiation emission are largely determined by geological and geographical factors. Soil contains radioactive elements such as plutonium, thorium, and potassium along with organic and inorganic compounds (Ramola et al., 2008). U-234, U-235, U-238, Ra-226, Th-232 etc., natural radionuclide distribution is not constant, and external radiation exposure varies by a factor. Natural radiation is the most significant contributor to the global population's total exposure (UNSCEAR, 2000). Radon and its isotopes are transferred to indoor air via. dwellings include windows and doors, as well as construction materials used in the walls, floors, and ceilings, which increase the natural background radiation exposure. It is being researched in a number of countries and it was also named as the second leading cause of lung cancer, after smoking (Kreienbrock et al., 2001; Letoureau et al., 1994; Jonsson et al., 1988; Kandari et al., 2016).

More than half (50%) of the ionizing radiation received by humans comes from radon, thoron, and their daughter products, when inhaled (UNSCEAR, 2000). The doses received as a result of radon and thoron were primarily contributed by their daughter products, according to a subsequent study (UNSCEAR, 2008). Therefore, it is essential to measure their daughter products (Ramola et al., 2016; Ramola et al., 2014; Prasad et al., 2016; Bangotra et al., 2015).

Geology of Uttarakhand

The present study is based on previous studies conducted in Uttarakhand state of Himalaya region, which is located at the northern part of India on the foothills of the Himalayan mountain range, located on 29-30 º N latitude and 78-79 º E longitude with Himalayan peaks (second highest peak Nanda Devi peak with 7681 m altitude) and glaciers and it has a total covered area of the state is 53,483 km^2 with 86% mountain. The significant value of the activity of radionuclides was recently reported in Uttarakhand and hence, the present study is based on the data of radionuclides distribution over the state. Generally, Uttarakhand is covered by hilly forest (65% total forest) with a population of 10.08 million as per 2011 senses. Garhwal and Kumaun divisions are the two main divisions of the state (wikipedia/Geology of Uttarakhand). Minerals

and ores abound in the province. Mylonitized porphyritic, lime, granite, quartz porphyry, phyllites, quartzite, schist, slates, and a wide variety of sedimentary rocks are found in the state. The final and stable state of radioactive elements is lead (Pb) (Valdiya et al., 1980; Ramola et al., 2006). The state of Uttarakhand has a high concentration of Pb in its various districts like Dehradun, Chamoli, Pauri, etc. Uttarakhand's lithotectonic units like the main central thrust (MCT) area are prone to a significant number of tectonic events per year with lesser Himalayan zone, the main boundary thrust (MBT) separates tertiary and pre-tertiary rocks and involves sediment portion (Bilham, 2004; Ansari, 2018). A significant activity of radionuclides has been estimated in MCT and MBT regions of Uttarakhand Himalayan region, India earlier (Ramola et al., 2014; Kandari et al., 2018).

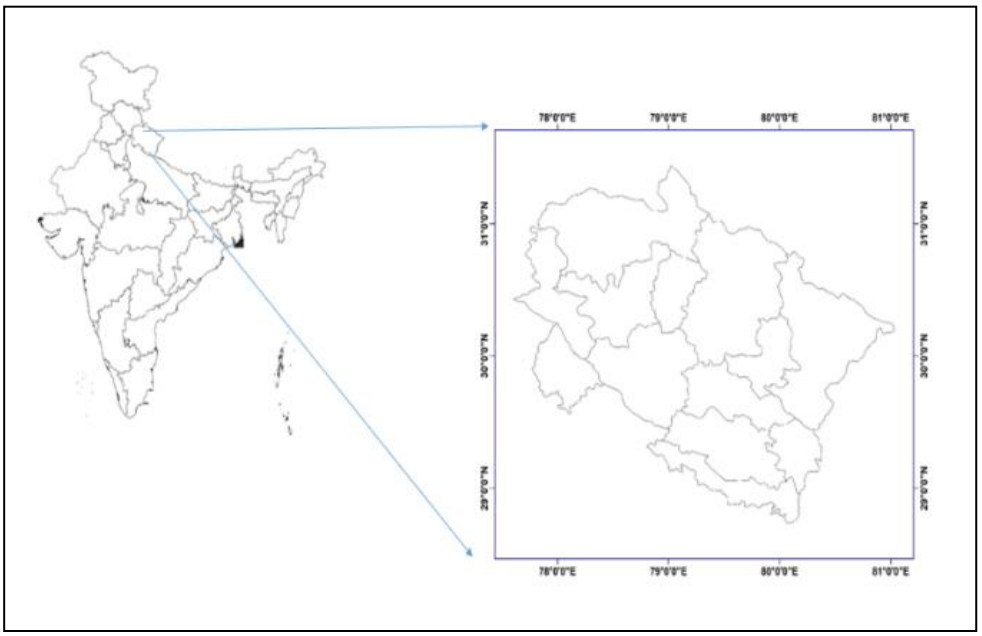


Fig. 2: *Map of Uttarakhand*

Materials and Methodology

Collection and preparation of Soil samples for analysis

During the soil sample's collection across the different parts of the study area**, o**ther entities such as pebbles, small rocks, roots, and leaves are removed from the soil, which is then packed in airtight packaging before analysis starts. Radon is transported into the air from the soil profile mainly by the emanation and exhalation process.

After soil sampling, a mesh with a mesh size of 150 microns was used to dry and prepare the samples. The soil samples were then sealed by using the airtight container for avoiding moisture and placed for a month to attain secular equilibrium between Ra-226, Rn-222, and their daughter products present in the soil. Scintillation NaI (Tl) gamma radiation detector (gamma-ray spectrometry) of scale 63 mm x 63 mm with several channel analyzers are used to measure radionuclides in soil

samples for 3 hours of counting time to have the values of radionuclide activity in soil (Rautela et al., 2012).

Radionuclide Measurement (Mehra et al., 2007)

Gamma-ray spectrometry is a combined two-detector spectrometric and radiometric instrument to measure mixed gamma beta radiation. The device was designed to identify and quantify gamma-emitting isotopes in a number of soil matrices. It can detect multiple Gamma emitting radionuclides in a single sample with minimal sample preparation. The calculation yields a continuum of lines, whose amplitude is proportional to the radionuclide's operation and whose location on the horizontal axis indicates its emitted energy. The spectrometer detects gamma radiation in the energy range from 50 to 3000 keV and beta radiation in the range of boundary from 150 to 3500 keV. It can measure gamma and beta radiation simultaneously. The spectrometer provides stabilization of the energy range on the energy line of 661.6 keV from the reference source Cs-137, 9 kBq, and also its operability and calibration safety controls with this reference source. The unit of Gamma detection is made up of a scintillation detector NaI (TI) with a scale of 63 x 63 mm, as well as electronic components such as a photomultiplier, amplification unit, selector, and light-emitting diode (Reguigui, 2006).

For sample preparation, the soil was dried in an oven at a temperature of 100°C to 120°C, and the sample size should be 150 microns. 250g of dried soil was packed in an airtight Marinelli vessel and for four weeks, the radionuclide and its daughter products were held in a state of secular equilibrium with each other. The acquire peak of Gamma spectra was normalized by comparing it to the 661 keV peak of Cs-137. The central component of gamma spectroscopy is the detector. In the case of annihilation, the gamma photons interfere with the detector substance and pass their energies to electrons or positrons. Ionized atoms and ion pairs are formed as these created particles lose their energy inside the detector. The detector signal is made up of these alternative entities (Reguigui, 2006).

A photomultiplier tube converts a light pulse generated by a gamma-ray communicating with a scintillator into an electric pulse. A photomultiplier is made up of a photocathode, electrode used for focusing, and dynodes (10 or more), each of which multiplies the number of electrons striking it several times. Transparency, large size availability, and large light intensity relative to gamma-ray energy are all properties of scintillation material that are needed for good detectors and very few materials have strong detector properties. NaI and CsI crystals that have been activated with thorium are widely used. The high Z of iodine in NaI provides good gamma-ray detection ability. To unlock the crystal, a slight amount of Tl is added, so the crystal is normally

referred to as NaI (Tl). For the 662 keV gamma-ray from Cs-137, the highest resolution achieved varies from 7.5 percent to 8.5 percent for a 3 in. diameter by 3 in. long crystal, which is significantly weaker for smaller and larger sizes. To massive crystals with several phototubes for X-ray measurements with a comparatively thin detector (to maximize resolution at the cost of performance at higher energies). A common configuration is crystals used with a well to allow virtually spherical 4P geometry for counting filler samples. Because of its good gamma-ray resolution and low cost, NaI is still the most used material for gamma detection. Commercially affordable NaI detector configurations are available. The detailed work can be found elsewhere (Reguigui, 2006).

Fig. 3: *Gamma Spectroscopy*

Radium equivalent activity

The following formula (eq. 1) is used for the calculation of radium equivalent activity which gives radionuclide concentration used to define radiation hazard associated with Ra-226, Th-232, and K-40 (UNSCEAR, 2000; Ramola et al., 2008).

$$Ra_{eq.} = A_{Ra} + 1.43 \cdot A_{Th} + 0.77 \cdot A_K \quad (1)$$

Where A_{Ra} = Activity concentration of Ra-226, A_{Th} = Activity concentration of Th-232, A_K = Activity concentration of K-40.

Absorbed Gamma Dose rate

As per the UNSCEAR, 2000 the absorbed dose rate due to natural radio-nuclide is calculated using the by using conversion factor (eq. 2)

$$D = 0.604A_{Th} + 0.463A_{Ra} + 0.0417A_K \quad (2)$$

Here A_{Ra} = Avg. Activity concentration of Ra-226, A_{Th} = Avg. Activity concentration of Th-232, A_K = Avg. Activity concentration of K-40.

Results and Discussion

The measured activity concentration of Radium, Thorium, and Potassium in different areas of Himalayan regions are found from 50 to 285 Bq/Kg, 38 to 384 Bq/Kg, and 310 to 2126 Bq/Kg with an average value of 112.78, 126.46, and 1210.51 Bq/Kg respectively. The details of radionuclide activity of a variable region of Himalaya Uttarakhand are shown in table 1. The activity concentration of radionuclides radium, thorium, and potassium is much higher in comparison to the world average value of 35, 30, and 400 Bq/kg according to UNSCEAR 2000 report. The highest Radium concentration was found in the Uttarkashi area while the lowest value was found in the Ukhimath area highest value may be due to the presence of a thrust plane and Uttarkashi is a highly uranium mineralized area (Bist et al., 1999; Ramola et al., 1988).

Table 1. Ra, Th, K-40, Absorbed Gamma Dose Rate, and Average $Ra_{eq.}$

Study Area	Ra-226 (Bq/kg)	Th-232 (Bq/kg)	K-40 (Bq/kg)	External Absorbed Gamma Dose Raten (nGy/h)	Average Radium Equivalent (Bq/kg)	Reference
S1	131±18	384±53	1406 ± 175	306	300	(Ramola et al., 2008)
S2	166.6±12.9	94.7±9.7	1378±37	179.6	384.2	(Gusain et al., 2009)
S3	141.7+11.9	155.9+12.4	672.8+25.9	169.1	276.9	(Rautela et al., 2012)
S4	50 ± 10	88 ± 16	885 ± 132	113	86	(Yadav et al., 2014)
S5	79.6 ± 18.4	99 ± 8.8	1201.3 ± 418.3	144	266.8	(Kandari et al., 2018)
S6	285 ± 28	136 ± 15	1588 ± 185	188	176.6	(Ramola et al., 2014)
S7	55± 10	101± 13	1310± 154	93	221	(Yadav et al., 2015)
S8	73±12	99±14	2126±176	148	228	(Kumar et al., 2021)
S9	69.85 ± 9.77	38 ± 7.37	1991 ± 256	-	-	(Semwal et al., 2018)
S10	76±11	69±10	549±67	96	174	(Anamika et al., 2020)

The maximum Thorium activity concentration was found in Budhakedar and the minimum concentration was found in Chai Khan, Almora. The value of thorium in soil samples

represents highly thorium mineralization. The maximum Potassium activity was found in Bageshwer and the minimum value was found in Shestradhara, Dehradun the high value was due to the usage of potassium-rich fertilizers. The overall data shows that the concentration of naturally occurring radionuclides directly is subject to the geological and geophysical features of the area (Bist et al., 1999). The distribution of radionuclide is shown in graph Fig. 4.

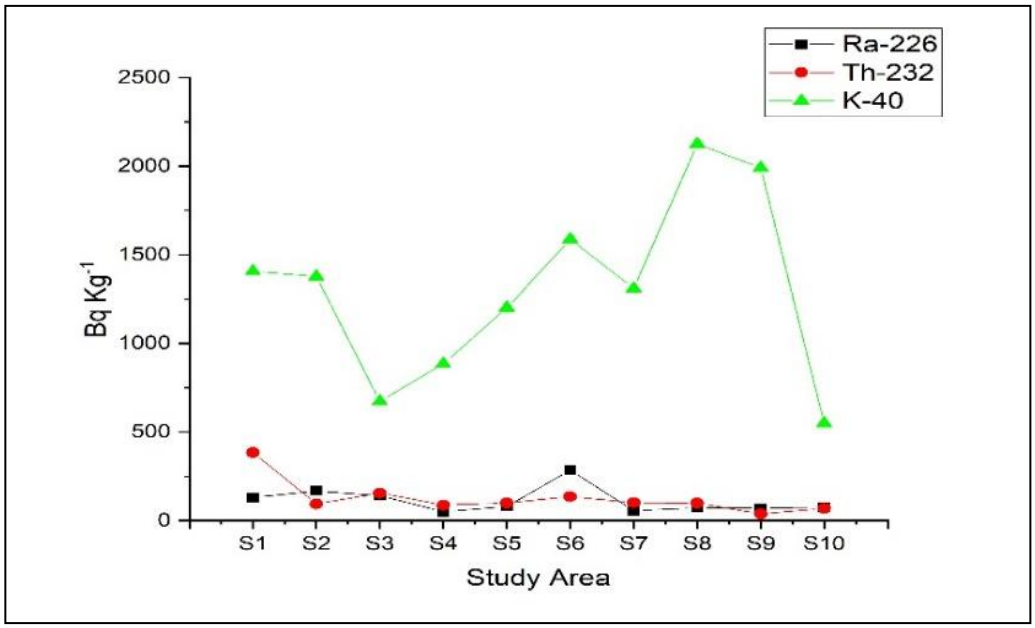


Fig. 4: Distribution graphs Ra, Th, K-40 radionuclides

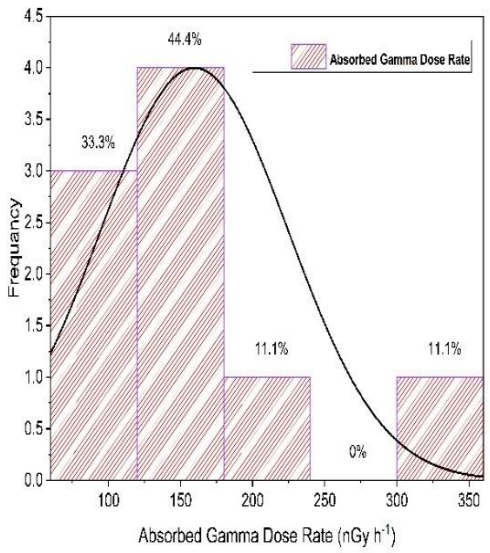
The distribution of radionuclide is not even hence exposure due to radionuclides is measured in terms of average radium equivalent activity. The average Radium equivalent in the area is shown in table 1 and range varies from 86 to 384.20 Bq/Kg with an average value of 234.84
Bq/Kg. The average value of radium equivalent was lower than recommendation level 370 Bq/Kg and acceptable for use (OECD, 1979). Approx 44.4% of radium equivalent found near 250 Bq/Kg Fig. 6(b).

External absorbed Gamma Dose rate was also calculated from soil samples and gamma dose rate varies from 93 to 306 nGy/h [with an average value of 159.63 nGy/h which is greater than the global average value of 57 nGy/h (Ramola et al., 2016; UNSCEAR, 2008). The frequency distribution of external absorbed Gamma Dose rate shown in fig. 5 (a). Box plots of data are shown in Fig. 6. Table 2 represents the statistical data representation of study locations mentioning Max., Min., Average and Std. Deviation.

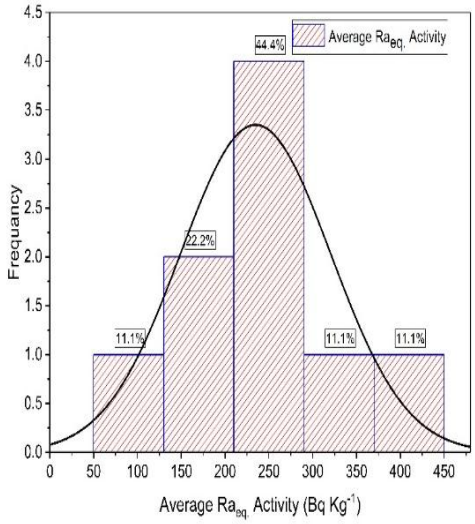
Conclusion

The study displays that the total activity concentration of Ra-226, Th-232, and K-40 in different areas of the Himalayas is higher than the reference level given by the UNSCEAR 2000. The average radium equivalent activity was found below the recommended level while the external absorbed gamma dose rate

was found above the reference. The Uttarakhand has associated with highly seismic areas and comes under IV and V seismic zone and most of the locations are situated along with the MCT, MBT, and MFT and holds various types of geology. The state consists most tectonically active zone from a geophysical point of view. Because of tectonic activity in the Himalayan region, there may be a huge change in the radionuclide concentration. The higher value of radionuclide is may be due to the geochemical composition of the soil and geophysical conditions. As all these radionuclide elements are not uniformly distributed in soil and rocks will play a significant role in the measurement of Radon transportation and help to find health Hazard due to indoor radon-thoron concentration.



(a) Frequency Distribution plot of absorbed dose rate



(b) Frequency distribution of average radium activity

Fig. 5: Frequency Distribution plot of (a)absorbed dose rate and (b) average radium activity

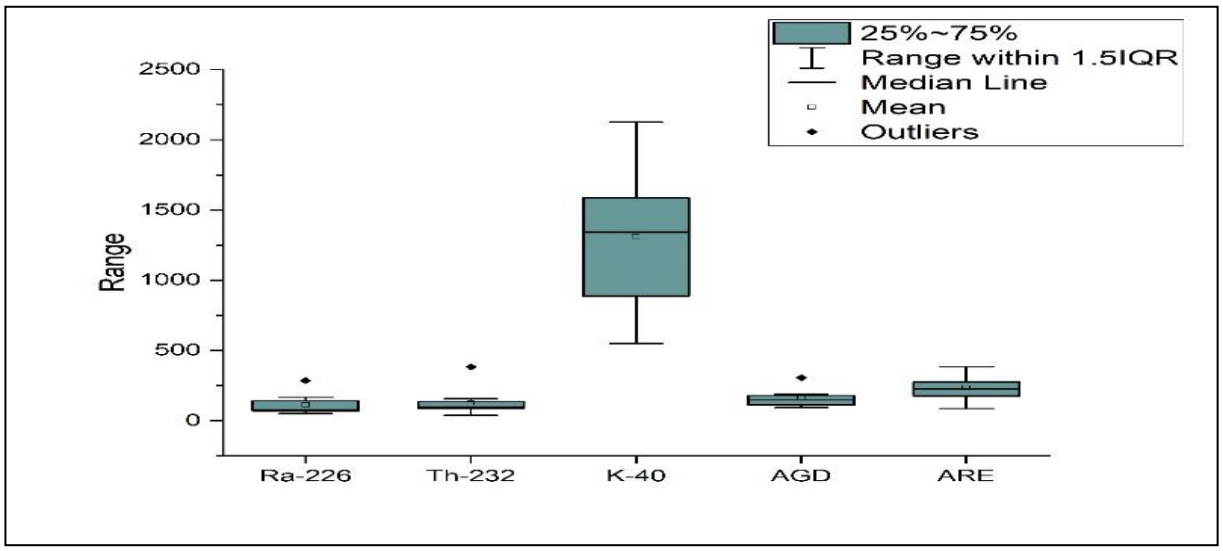


Fig. 6: Box plots of statistical data

Table 2. Statistical values of data

Study Area	Ra-226 (Bq/kg)	Th-232 (Bq/kg)	K-40 (Bq/kg)	External absorbed Gamma Dose Raten (nGy/h)	Average Radium Equivalent (Bq/kg)
Max.	285±28	384±53	2126±176	306.00	384.20
Min.	50±10	38±7.37	549±154	93.00	86.00
Averge	112.78	126.46	1310.71	159.63	234.83
S.D.	72.18	96.06	675.08	64.96	85.75

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