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Multivariate statistical analysis of radiological data of building materials used in Tiruvannamalai, Tamilnadu, India

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HIGHLIGHTS

- To develop reference data of radiological parameters in building materials.
- The radiological data were processed using multivariate statistical methods.
- The values obtained in the study are within the recommended safety limits.

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ABSTRACT

Using γ spectrometry, the concentration of the naturally occurring radionuclides ^{226}Ra, ^{232}Th and ^{40}K has been measured in soil, sand, cement, clay and bricks, which are used as building materials in Tiruvannamalai, Tamilnadu, India. The radium equivalent activity (Ra_{eq}), the criterion formula (*CF*), indoor gamma absorbed dose rate (D_R), annual effective dose (H_R), activity utilization index (AUI), alpha index (I_α), gamma index (I_γ), external radiation hazard index (H_{ex}), internal radiation hazard index (H_{in}), representative level index (RLI), excess lifetime cancer risk (ELCR) and annual gonadal dose equivalent (AGDE) associated with the natural radionuclides are calculated to assess the radiation hazard of the natural radioactivity in the building materials. From the analysis, it is found that these materials used for the construction of dwellings are safe for the inhabitants.

The radiological data were processed using multivariate statistical methods to determine the similarities and correlation among the various samples. The frequency distributions for all radionuclides were analyzed. The data set consisted of 15 measured variables. The Pearson correlation coefficient reveals that the ^{226}Ra distribution in building materials is controlled by the variation of the ^{40}K concentration. Principal component analysis (PCA) yields a two-component representation of the acquired data from the building materials in Tiruvannamalai, wherein 94.9% of the total variance is explained. The resulting dendrogram of hierarchical cluster analysis (HCA) classified the 30 building materials into four major groups using 15 variables.

1. Introduction

Construction is one of the main beneficiaries of private and public investments in the world. The building industry requires large quantities of low-cost materials and novel products that may substitute for more widely used conventional building materials. However, such novel building materials may contain significant quantities of naturally or technologically enhanced naturally occurring radioactive materials. Generally, the specific activities of ^{238}U, ^{232}Th and ^{40}K in raw building materials and their products depend on geological and geographical conditions as well as the geochemical features of those materials (Rizzo et al., 2001).

The assessment of the population's exposure to indoor radiation is very important; therefore, knowledge regarding the

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concentration of natural radionuclides in construction materials is required. Construction materials are derived from both natural sources (e.g., rock and soil) and waste products (e.g., phosphogypsum, alum shale, coal, fly ash, oil-shale ash, some rare minerals and certain slugs) as well as from industry products (e.g., power plants, phosphate fertilizer and the oil industry) (O'Brien, 1997). Although building materials act as sources of radiation to the inhabitants in dwellings, they also shield against outdoor radiation (Akkurt et al., 2007). Knowing the level of the natural radioactivity in building materials is important to assess the associated radiological hazards to human health and to develop standards and guidelines for the use and management of these materials.

The natural radioactivity of building materials in many countries has been reported (Ahmad and Matiullah Hussain, 1988; Amrani and Tahtat, 2001; Beretka and Mathew, 1985; Chong and Ahmad, 1982; Mollah et al., 1986; Viresh Kumar et al., 1999; Zaidi et al., 1999; El-Arabi, 2005; El-Tahaway and Higgy, 1995; Khalid Khan and HasanKhan, 2001; McAulay and Moran, 1988; Safdar Ali et al., 1996; Turhan, 2008; Stoulos et al., 2003; Tufail et al., 1992). Natural radioactivity in some Indian building materials has also been reported by other authors (Viresh Kumar et al., 1999; Ravisankar et al., 2012; Ajay Kumar et al., 2003; Nageswara Rao et al., 1996). However, detailed information for each state is scant. Data regarding the concentration of ^{226}Ra, ^{232}Th and ^{40}K in the building materials of Tiruvannamalai in the state of Tamilnadu in India is not available in the literature. Tiruvannamalai is a world-renowned temple town in Tamilnadu, which is a synonym for deepam (fire). Here, Shiva (Arunachaleswar) is worshiped in the form of fire. This temple city is located approximately 180 km from Chennai. It is believed that this temple on the foot hills of Annamalai Hill was built in approximately 750 AD, based on the details available from archeological sculptures. It is the largest temple in India dedicated to Lord Shiva.

In the present work, the concentration of natural radionuclides was measured in 30 samples of building materials that were commonly used in Tiruvannamalai city in Tamilnadu, India, using gamma spectrometry. The potential radiological hazards associated with the studied materials were assessed by calculating the radium equivalent activity (Ra_{eq}), criteria formula (CF), indoor gamma absorbed dose rate (D_R), annual effective dose (H_R), activity utilization index (AUI), alpha index (I_α), gamma index (I_γ), external radiation hazard index (H_{ex}), internal radiation hazard index (H_{in}), representative level index (RLI), excess lifetime cancer risk (ELCR) and annual gonadal dose equivalent (AGDE). The obtained results were compared to the recommended values to assess the radiation hazards to humans resulting from the building materials, and these results were also compared to the corresponding values of building materials from different countries. The radiological data were processed using multivariate statistical methods such as the Pearson correlation coefficient as well as cluster and principal component analyses to determine the similarities and correlations among the various samples.

2. Materials and methods

2.1. Sample collection and preparation

Commonly used structural building materials (clay, soil, brick, sand and cement) were collected randomly from sites where housing and other buildings were under construction and from building-material suppliers in Tiruvannamalai for measurement of the specific radioactivity of ^{226}Ra, ^{232}Th and ^{40}K. These materials were used in bulk amounts. The materials were studied in their natural form. Each sample was properly cataloged, marked and coded according to its origin and the location of the sampling site. After crushing, powdering, coning and quartering, representative samples with a maximum grain size of 1 mm were dried in an oven at approximately 110 °C until the sample weight became constant. These samples were sealed in radon-impermeable plastic containers. The samples were then stored for more than 30 days to bring ^{222}Rn and its short-lived daughter products into equilibrium with ^{226}Ra (Ravisankar et al., 2012).

2.2. Gamma-ray spectroscopic technique

All selected samples were subjected to gamma spectral analysis using a 7.62 cm × 7.62 cm NaI (Tl) detector. The energy resolution of the NaI (Tl) detector measured in terms of the full width at half maximum (FWHM) is 50 keV at the energy of 662 keV gamma of ^{137}Cs at 25 cm from the top of the detector. The detector is shielded with 15 cm thick lead on all sides including the top, to reduce the background contribution from the surroundings. The inner sides of the lead shielding are lined with 2 mm thick cadmium and 1 mm thick copper to attenuate lead X-rays and cadmium X-rays, respectively. The certified IAEA reference materials RGU, RGTh and RGK were used for the energy and efficiency calibration of the system in the energy range from 186.21 to 2614.53 keV. The activity contents of the IAEA reference materials, which are housed in 250 ml bottles, are known with ± 3% accuracy. The efficiency percentages for ^{40}K (1.461 MeV), ^{214}Bi (1.764 MeV) and ^{208}Tl (2.615 MeV) were found to be 0.154, 0.357 and 0.301 cps Bq^{-1}, respectively. The samples were sealed in radon-impermeable plastic containers. The samples were then stored for more than 30 days to bring ^{222}Rn and its short-lived daughter products into equilibrium with ^{226}Ra. The samples were then counted in the same source-to-detector geometry used for the establishment of the efficiency calibration. The spectra were acquired for 20,000 s and the photo peaks were evaluated by the MCA software. The gamma-ray photo peaks corresponding to 1.461 MeV (^{40}K), 1.764 MeV (^{214}Bi) and 2.615 MeV (^{208}Tl) were considered to determine the activities of ^{40}K, ^{226}Ra and ^{232}Th in the samples. The detection limits of the NaI (Tl) detector system at the 95% confidence level for ^{40}K, ^{226}Ra and ^{232}Th are 8.50, 2.21 and 2.11 Bq kg^{-1}, respectively, for a counting time of 20,000 s. The results for the activity concentrations of the samples are reported with 2σ errors.

3. Results and discussion

3.1. Specific radioactivity

The activity concentrations of the detected radionuclides ^{226}Ra ^{232}Th and ^{40}K in these building materials are presented in Table 1. As shown in Table 1, the highest values observed for the specific activities of ^{226}Ra, ^{232}Th and ^{40}K are 54 (Cement-1), 198 (Sand-5) and 724 Bq kg^{-1} (Soil-4), respectively, while the lowest observed values of the specific activities of the same radionuclides are BDL, 11 (Clay-3) and 149 (Cement-1) Bq kg^{-1} respectively. As shown in Table 1, the activity of ^{226}Ra varies from BDL to 54 Bq kg^{-1} and the arithmetic mean is 9 Bq kg^{-1}. The activity concentration of ^{232}Th varies from BDL to 11 (Clay-3) Bq kg^{-1}, and the arithmetic mean is 49 Bq kg^{-1}. The activity concentration of ^{40}K varies from 149 to 724 Bq kg^{-1}, and the arithmetic mean is 356 Bq kg^{-1}.

The mean values are lower than the corresponding worldwide average values, which are 35, 30 and 400 Bq kg^{-1} for ^{226}Ra, ^{232}Th and ^{40}K, respectively (UNSCEAR, 2000). If the radionuclide activities of the present study are compared to the world average values, the ^{226}Ra activity is lower by a factor of 0.29, while the activity of ^{232}Th is found to be higher by a factor of 1.63 and the ^{40}K activity is lower by a factor of 0.89. Fig. 1 shows the activity concentrations of the natural radionuclides for the different building materials.