Assessment of natural radioactivity levels in soil sample from Botteng Utara Village, Mamuju Regency Indonesia

Nurokhim, Kusdiana, Eko Pudjadi
Center for Technology of Radiation Safety and Metrology, BATAN.
Jl. Lebak Bulus Raya No.49, Jakarta Selatan, 12440 Indonesia
E-mail : nurokhim@batan.go.id

Abstract. High background radiation areas usually are correlated to the high concentration of primordial radionuclides activity from Uranium/Thorium series and -K. This paper reported the natural radioactivity level in soil sample was taken from Botteng Utara village area. The activity of primordial radionuclide such as -Ra, -Th and -K were investigated from the soil by gamma spectrometry. In order to evaluate the radioactivity levels of the area, Radium equivalent activity (Ra_e), the absorb dose rate (D), the annual effective dose rate (E), the external hazard index (H_e), and internal hazard index (H_i) were calculated and compared with the internationally approved value. The concentration of -Ra, -Th and -K measured by gamma spectrometry are lies in the range 268.90 to 2921.17, 993.07 to 3153.81, and 115.72 to 438.26 Bq.kg respectively. The average and maximum annual effective dose received by Botteng Utara resident from terrestrial gamma rays are 10.40 and 18.62 mSv y⁻¹, which is the maximum received by resident of Tande-Tande hamlets. This present work clarify that Botteng Utara is an area with high background radiation exposure from primordial radionuclides activity.

1. Introduction
Natural radioactivity is spread in the environment in various formations such as soil, rock, sediment, water, plants and air. The main radioactive substances contained in the environment are long-life radionuclides known as NORMs (Naturally Occurring Radionuclide Materials), especially -U (-Ra) series, -Th series and -K. Humans are continuously exposed to ionizing rays emitted by a radioactive substance that can come from external and internal sources. External sources are in the form of terrestrial radiation and cosmic rays radiation, while internal sources come from radioactive substances that enter the body along with the entry of food, drinks and breathing [1- 3]. The estimated average radiation dose received by the world population 85% comes from natural radioactivity while the remaining ~15% comes from artificial sources. The total dose received by the population in a particular area depends on the content of radioactive NORMs present in the area. Land is a source of sustained exposure to radiation in humans and also as a medium for the transfer of radioactive
substances to other environmental media, therefore an analysis of the concentration of radioactive substances in the soil is particularly importance to estimate the total and effective doses received by the community in certain areas [4], [5], [6].

The objectives of the present study are to assess the natural radioactivity levels of Botteng Utara Village, Mamuju Regency, a district which according to previous studies was an area with high radiation exposure in Indonesia. The assessment is done by measurement of the natural radioactivity level of $^{238}$U ($^{226}$Ra), $^{232}$Th and $^{40}$K in the surface soil (0-5 cm) and shallow soil (5-20 cm) from Botteng Utara. In laboratory, presented of $^{238}$U is represented by concentration of $^{226}$Ra. Assessment of radiation levels in Botteng Utara village are investigated by calculation of Radium equivalent activity, air absorbed external gamma radiation exposure, annual effective radiation dose, and external and internal radiation hazard index.

2. Materials and methods

2.1 Area study

Botteng Utara is one of several villages in Simboro sub-district, Mamuju Regency, West Sulawesi Province. Botteng Utara is divided into 10 hamlets namely: Pasada, Popanga, Sendana, Adi-Adi, Oniang, Punaga, Pakkaroang, Salu Kalo, Tande-Tande and Salu Rumbia as shown in Figure 1. Botteng Utara topography is hilly with village access roads, most of which are quite good to accessed, except the access roads to Tande-Tande and Pakkaroang, that are difficult to reach although by motor cycle. According of Central Bureau of Statistics (2017 ) the population of Botteng Utara is 1850 people and gathered in 487 families. The population of that regions are being continues to grow, especially those area around the Trans Sulawesi Highway, Pasada, Sendana, Popanga and Adi-Adi.

![Figure 1. The map of Botteng Utara (North Botteng), Mamuju Regency, West Sulawesi.](image)

2.2 Sample collection and preparation

Two kinds of soil samples 0-5 cm and 5-20 cm depth were taken from ten hamlets of Botteng Utara. Around 2 kg soil is collected for each sample. After collection, sample was wrapped in black plastic bag and labelled according to the name of the location. The geographical coordinates of the sampling point were recorded by Global Positioning System (GPS).

In Laboratory, the soil sample was dried in an oven at temperature of 105°C to remove moisture and make it constant weight. After drying the sample were crushed and sieved through 60 mesh sieve diameter to obtain a sample with a homogeneous grain of 250 µm. Afterward the homogenized samples were packed in 1 litter marinelli beakers. These filled marinelli are stored for a period four weeks to allow radioactive equilibrium among the decay products of radon ($^{222}$Rn) and thoron ($^{220}$Rn) prior to gamma spectrometric analysis.
2.3 Gamma Spectrometry

All samples were subjected to gamma spectral analysis using High Pure Germanium gamma spectrometer. The HPG(c) gamma spectrometer GEM-F590 with 20% relative efficiency and controlled by ORTEC Gamma Vision Software was used to determine the activity concentration of \(^2\)Ra, \(^\alpha\)Th and \(^\alpha\)K. The Certified Reference Material (CRM) of soil mixed gamma source (MGS) in 1 litre of marinelli was used for energy and efficiency calibration. The samples were placed in a shielded gamma ray spectrometry unit for a counting time of 61,200 s and analyzed. The activity of \(^\alpha\)Ra obtained from 609.3 (46.1%), 1120.3 (15.1%) and 1764.5 (15.4%) keV, photopeak of \(^\alpha\)Bi, \(^\alpha\)Th obtained from 338.3 (11.3%), 911.2 (25.8%), 968.9 (15.8%) keV photo peak of \(^\alpha\)Ac and \(^\alpha\)K obtained from 1460.8 (11%) keV. [7]

The specific activity of a nuclide is measured by gamma spectrometer and calculated using equation 1 [6,8]:

\[
C_n = \frac{N_n}{(\gamma_n, \eta_n, T_m)}
\]  

(1)

Where \(C_n\) is specific activity of nuclide \(n\) for a peak of energy \(E\), \(N_n\) is the net counts of a nuclide \(n\) in a peak at energy \(E\), \(T\) is the time counting of a sample, \(\gamma_n\) is the yield of gamma ray abundance per disintegration of the specific nuclide \(n\) at energy \(E\), \(m\) is the mass of sample and \(\eta_n\) is the detection efficiency of HPG(c) detector at energy \(E\).

2.4 Determination of radium equivalent activity

Radium equivalent activity is a single index, which expresses the gamma yield from various mixture of \(^\alpha\)Ra, \(^\alpha\)Th and \(^\alpha\)K in the sample. The radium equivalent activity index was given as in equation 2 [1, 9-11]:

\[
\text{Raeq(Bq kg}^{-1}) = C_{\text{U}} + 1.43 C_{\text{Th}} + 0.077 C_{\text{K}}
\]

(2)

Where \(C_U\), \(C_{\text{Th}}\), and \(C_{\text{K}}\) were the radioactivity concentration in Bq kg\(^{-1}\) of \(^\alpha\)U, \(^\alpha\)Th, and \(^\alpha\)K. It may be noted the decay product of \(^\alpha\)Ra replaced \(^\alpha\)U, although there may be disequilibrium between \(^\alpha\)U and \(^\alpha\)Ra [12]. It is accepted that 370 Bq kg\(^{-1}\) of \(^\alpha\)U or 259 Bq kg\(^{-1}\) of \(^\alpha\)Th or 4810 Bq kg\(^{-1}\) of \(^\alpha\)K produces the same gamma dose rate [13]. The maximum value of Ra, must be less than 370 Bq kg\(^{-1}\).

2.5 Absorbed gamma dose rate (D)

According to guideline by UNSCEAR [2], the absorbed gamma dose rate in air 1 m above the ground surface for uniform distribution of natural radionuclide’s has been calculated by equation 3 [1, 9-11]:

\[
D \text { (nGy h}^{-1}) = 0.462 C_U \text{ (Bq kg}^{-1}) + 0.604 C_{\text{Th}} \text{ (Bq kg}^{-1}) + 0.0417 C_{\text{K}} \text{ (Bq kg}^{-1})
\]

(3)

Where 0.462, 0.604, and 0.0417 nGy/Bq kg\(^{-1}\) are dose conversion factors for \(^\alpha\)U (\(^\alpha\)Ra), \(^\alpha\)Th, and \(^\alpha\)K respectively and \(C_U\), \(C_{\text{Th}}\) and \(C_{\text{K}}\) are the radioactivity concentration of \(^\alpha\)U (\(^\alpha\)Ra), \(^\alpha\)Th, an \(^\alpha\)K in Bq kg\(^{-1}\) respectively.

2.6 Annual Effective dose (E)

The annual effective dose received by the population from the radioactivity in soil is estimated with employing a conversion factor of 0.7 Sv/Gy. The adults spend about 80% of their time indoors, while the remaining 20% time is spent outdoors. Therefore, the indoor and outdoor occupancy factors were given as 0.8 and 0.2, respectively [1,9]. The indoor and outdoor annual effective doses are calculated by equations 4 and 5:

\[
E_i \text{ (m Sv y}^{-1}) = D_i \text{ (nGy h}^{-1}) \times 8670 \text{ h} \times 0.8 \times 0.7 \text{ (Sv Gy}^{-1}) \times 10^6
\]

(4)

\[
E_o \text{ (m Sv y}^{-1}) = D_o \text{ (nGy h}^{-1}) \times 8670 \text{ h} \times 0.2 \times 0.7 \text{ (Sv Gy}^{-1}) \times 10^6
\]

(5)
Where $E_i$ and $E_e$ are annual effective doses for indoor and outdoor environments and $D_i$ and $D_e$ are absorbed gamma dose rate ($D$) calculated by equation (3) with radioactivity concentration of natural radionuclides in shallow and surface soil.

2.7 External Hazard Index
The external exposure is caused by direct gamma radiation whereas internal exposure is caused by the inhalation of radon ($^{222}$Rn), thoron ($^{220}$Rn) and their short-lived decay products [14]. For the estimation of gamma radiation dose measurement anticipated that is conveyed externally from building materials, the external hazard index ($H_e$) is measured by equation 6 [15, 16]:

$$H_e = C_i/370 + C_e/259 + C_d/4810$$

The external hazard index should be less than unity to stay the radiation risk to be minimal, which is equivalent to a maximum value of radium equivalent 370 Bq kg$^{-1}$.

2.8 Internal Hazard Index
Radon and thoron are natural radionuclide decay products in gaseous form, which can be released to the air. The existences of radon and thoron in soil are the reason of lungs cancer when inhaled by inhabitants. Henceforth, to determine of radon exposure, Beretka and Mathew [14] reported the equation 7 for the internal hazard index calculation:

$$H_i = C_i/185 + C_e/259 + C_d/4810$$

The values of $H_e$ and $H_i$ index must be less than 1 mSv y$^{-1}$ in order to cause any harmful effect to population [17].

3. Results and Discussion
Radioactivity content of $^{222}$Ra, $^{232}$Th and $^{40}$K in surface (0-5 cm) and shallow (5-20 cm) soil sample from Botteng Utara has been measured by gamma spectrometer. The content of $^{222}$Ra, $^{232}$Th, $^{40}$K, calculated radium equivalent and air absorbed dose rate are shown in Tables 1-2. The content of U, Th and K in soil sample are varies from 268.90 ± 25.52 Bq kg$^{-1}$ (Adi-Adi) to 2921.17±274.51 Bq kg$^{-1}$ (Tande-Tande), 993.07±93.29 Bq kg$^{-1}$ (Salu Kalo) to 3153.81±296.26 Bq kg$^{-1}$ (Popanga) and 115.72±11.75 Bq kg$^{-1}$ (Adi-Adi) to 438.26±42.02 Bq kg$^{-1}$ (Punaga) respectively.

Average activity concentration of $^{222}$Ra and $^{232}$Th in surface soil are 1042.29±98.16 and 1756.12±165.21 Bq kg$^{-1}$ that higher 29.78 and 58.54 times then world’s average concentration reported by UNSCEAR which is 35 Bq kg$^{-1}$. Meanwhile in shallow soil the concentration are 1283.60±120.02 and 1869.89 Bq kg$^{-1}$, that are 36.67 and 60.33 times higher than world’s average concentration. In the other hand average concentration of $^{40}$K is 232.63±22.66 Bq kg$^{-1}$ lower then the world’s average concentration (400 Bq kg$^{-1}$)[1, 2].

As shown in Table 2, the maximum concentration of $^{222}$Ra found in Tande-Tande was 2921.17 Bq kg$^{-1}$ its mean 83.46 times higher than world’s average, and maximum of $^{232}$Th found in Popanga with concentration 3153.81 Bq kg$^{-1}$ mean 105.13 times higher than world’s average.

Table 1. Measurement result activity of $^{222}$Ra, $^{232}$Th and $^{40}$K together with calculated radium equivalent and absorbed dose rate in surface soil (0-5 cm) sample of Botteng Utara.

| Sample Location | Radioactivity concentration (Bq kg$^{-1}$) | $^{222}$Ra | $^{232}$Th | $^{40}$K | $^{222}$Ra | $^{232}$Th | $^{40}$K | $^{222}$Ra | $^{232}$Th | $^{40}$K |
|-----------------|------------------------------------------|----------|--------|--------|----------|--------|--------|----------|--------|--------|
| Pasada          | 697.09±65.75                             | 1868.04±176.64 | 115.72±11.75 | 3377.30 | 1455.18 |
| Sendana         | 1133.64±106.70                           | 1534.57±144.32 | 205.73±20.16  | 3343.92 | 1459.20 |
Table 2. Measurement result activity of $^{226}$Ra, $^{232}$Th and $^{40}$K together with calculated radium equivalent and absorbed dose rate in shallow soil (5-20 cm) sample of Botteng Utara.

| Sample Location | Radioactivity concentration (Bq kg$^{-1}$) | Ra$_{eq}$ (nGy h$^{-1}$) | D (nGy h$^{-1}$) |
|-----------------|------------------------------------------|--------------------------|-----------------|
|                 | $^{226}$Ra | $^{232}$Th | $^{40}$K |                 |                                        |
| Pasadena        | 870.61 ± 82.01 | 2046.57 ± 192.37 | 151.18 ± 15.02 | 3808.85 | 1644.65 |
| Sendana         | 882.81 ± 75.66 | 1633.17 ± 153.59 | 142.58 ± 14.23 | 3229.22 | 1400.24 |
| Adi-Adi         | 268.90 ± 25.52 | 1726.77 ± 162.34 | 116.73 ± 11.73 | 2915.00 | 1277.31 |
| Oniang          | 1100.14 ± 103.54 | 1254.55 ± 118.02 | 270.83 ± 26.20 | 2915.00 | 1277.31 |
| Punaga          | 950.37 ± 89.49 | 1104.82 ± 103.97 | 361.39 ± 34.72 | 2558.09 | 1121.45 |
| Popanga         | 1783.52 ± 167.67 | 3153.81 ± 296.26 | 226.87 ± 22.06 | 6310.94 | 2738.35 |
| Pakaroang       | 1783.61 ± 167.72 | 1574.86 ± 148.12 | 196.16 ± 19.29 | 4050.76 | 1783.42 |
| Salu Kalo       | 915.25 ± 86.19 | 1292.66 ± 121.60 | 344.54 ± 33.14 | 2790.28 | 1217.98 |
| Tande-Tande     | 2921.17 ± 274.51 | 2943.40 ± 276.59 | 200.46 ± 19.77 | 7145.67 | 3135.75 |
| Salu Rumbia     | 1359.61 ± 127.90 | 1968.29 ± 185.02 | 315.58 ± 30.43 | 4198.56 | 1830.15 |
| Average         | 1283.60 ± 120.02 | 1869.89 ± 175.79 | 232.63 ± 22.66 | 3975.45 | 1732.14 |

**Figure 2.** Calculated radium equivalent of soil samples

Radium equivalent (Ra$_{eq}$) in soil samples studied are ranges from 2040.81 (Punaga) to 7145.67 Bq kg$^{-1}$ (Tande-Tande) with average value 3773.42 Bq kg$^{-1}$. Averages Ra$_{eq}$ of surface and shallow soil are 3571.39 Bq kg$^{-1}$ and 3975.45 Bq kg$^{-1}$ that mean 9.65 and 10.74 times higher than global average. As shown in Figure 3, radium equivalent in shallow soil is higher than in surface soil except for the three hamlets Popanga, Adi-Adi and Salu Rumbia that are slightly higher in surface compare to shallow soil.
The observed value of air absorbed dose rate in surface soil samples of study are ranges from 888.84 to 2642.18 nGy h⁻¹ with an average value of 1551.90, meanwhile in shallow soil on the ranges from 1121.45 to 3135.75 nGy h⁻¹ with average value 1732.14 nGy h⁻¹. It is observed that air absorbed dose rate much greater than the population-weighted average value of global primordial radiation of 60 nGy h⁻¹ [1, 2].

Table 3. Annual Effective dose, internal and external hazard index in soil sample of hamlets in Botteng Utara

| Sample Location | Annual Effective dose ( mSv y⁻¹) | Hazard index |
|-----------------|----------------------------------|--------------|
|                 | Outdoor | Indoor | Total | Hex | H_in |
| Pasada          | 1.78    | 8.07   | 9.85  | 9.12 | 12.64 |
| Sendana         | 1.79    | 6.87   | 8.66  | 9.03 | 11.11 |
| Adi - Adi       | 1.63    | 5.75   | 7.38  | 8.41 | 8.14  |
| Oniang          | 1.49    | 6.27   | 7.76  | 7.51 | 10.85 |
| Punaga          | 1.21    | 5.50   | 6.72  | 6.14 | 9.48  |
| Popanga         | 2.70    | 13.43  | 16.13 | 13.72 | 21.86 |
| Pakaroang       | 1.79    | 8.75   | 10.54 | 9.00 | 15.76 |
| Salu Kalo       | 1.09    | 5.97   | 7.06  | 5.51 | 10.01 |
| Tande-Tande     | 3.24    | 15.38  | 18.62 | 16.33 | 27.20 |
| Salu Rumbia     | 2.31    | 8.98   | 11.29 | 11.67 | 15.01 |
| Average :       | 1.90    | 8.50   | 10.40 | 9.65 | 14.21 |

The outdoor and indoor annual effective dose, external and internal hazard index are represented in Table 3. The outdoor and indoor annual effective dose in soil samples of Botteng Utara ranges from 1.09 to 3.24 mSv y⁻¹ and 5.50 to 15.38 mSv y⁻¹ with average values of 1.90 mSv y⁻¹ and 8.50 mSv y⁻¹. And the total annual effective dose ranges from 7.06 to 18.62 mSv y⁻¹ with average 10.40 mSv y⁻¹. All calculated outdoor and indoor annual effective dose from the samples are higher than the worldwide average value of 0.07 and 0.41 mSv y⁻¹ [1, 2]. The average annual Effective dose receive by Botteng Utara residents 10.40 mSv y⁻¹ (21.67 times greater than global average), and the greatest annual effective dose received by residents of Tande-tande hamlet with effective dose 18.62 mSv y⁻¹ (38.80 times higher than global average).

External and internal hazard index in this study are varies from 5.51 to 16.33 and 8.14 to 27.20 with average values 9.65 and 14.21. As shown in Table 3 and Figure 3 hazard index for all soil has been studied above unity, all area has been study have a high cancer risk. The external hazard index is used to assess radiological hazard from external exposure, the purpose of these value is to limit the external gamma radiation dose to the admissible dose limit 1 mSv y⁻¹ for a safe radiation hazard.
Figure 3. Calculated annual effective dose and hazard index of soil samples

4. Conclusion

- Average activity concentration of radionuclides $^{238}$U ($^{226}$Ra) and $^{232}$Th and $^{40}$K are above the world average values. The concentration of $^{238}$U ($^{226}$Ra) and $^{232}$Th in soil of Botteng Utara are 29.78 and 58.54 times higher than the world’s average concentration of Uranium and Thorium.
- The average and maximum annual effective dose received by Botteng Utara resident from terrestrial gamma rays are 10.40 and 18.62 mSv y$^{-1}$, which are 21.67 and 38.80 times higher than the world’s global effective dose.
- External hazard index and internal hazard index for all samples investigated are greater than recommended safety limits and hence need to future attention in relation to radiation hazards to the inhabitants.
- It is confirmed that Botteng Utara is high background radiation area influenced by primordial radioactivity.

Acknowledgments
The authors are thankful to the residents of the area studied, the Government of Botteng Utara Village and Mamuju regency for their cooperation during fieldwork. We are also very grateful to the Center for Technology of Radiation Safety and Metrology, National Nuclear Energy Agency, the providing fund of the research during strategic plan 2015-2019.

References
[1] UNSCEAR 2000 Radiation Sources and Effects of Ionizing Radiation Report of the United Nations Scientific Committee on the Effect of Atomic Radiation to General Assembly, New York USA
[2] UNSCEAR 1994, Sources and effects of ionizing radiation Report to General Assembly, New York USA
[3] Singh J, Singh H, Singh S, Bajwa B S and Sonkawade R G 2009. Comparative study of natural radioactivity in soil samples from the Upper Siwaliks and Punjab India using gamma ray spectrometry J. Env. Radioactivity 100 p.94-98
[4] Duggal V, Rani A, Mehra R and Ramola C 2013, Assesment of Natural Radioactivity Levels and Associated Dose Rate in Soil Samples from Northern Rajasthan, India, J. Rad. Prot. Dosimetri p.1-6.
[5] Mubarak F, Fayez-Hassan M, Mansour NA, Ahmed TA and Ali A 2017 Radiological
Investigation of High Background Radiation Areas J. Nat. Lib. Of Medicine Published online

[6] Amanjeet, Kumar A, Kumar S, Singh J, Singh P and Bajwa BS 2017 Assessment of natural radioactivity levels and associated dose rates in soil samples from historical city Panipat India J. Rad. Research and App. Sci. 10 p.283-88

[7] IAEA 2007, Update of X rays and Gamma rays decay data standards for detector calibration and other applications, vol 2, Vienna, Austria.

[8] Abdel-Rahman M A E and El-Mongy S A 2017 Analysis of radioactivity levels and hazard assessment of black sand samples from Rashid area Egypt J Nuc. Eng. & Tech. 49 p.1752-57 Korean Nuclear Society

[9] Amrani D and Tahtat M 2001 Natural radioactivity in algerian building materials J. App. Rad. & Isotopes 54(4), p.687-89

[10] Derin M T, Vijayagopal P, Venkatraman B, Chaubey R C and Gopinathan A 2012 Radionuclides and Radiation Indices of High Background Radiation Area in Chavara-Neendakara Placer Deposits (Kerala, India) J Suminori Akiba Kagoshima University Japan

[11] Raghu Y, Ravisankar R, Chandrasekaran A, Vijayogopal P and Venkatraman B 2017 Assessment of natural radioactivity and radiological hazards in building materials used in the Tiruvannamalai District, Tamilnadu, India, using a statistical approach J. Taibah Univ. for Sci. 11, p.523-33.

[12] Iqbal M, Tufail M and Mirza S M 2000 Measurement of natural radioactivity in marble found in Pakistan using NaI(Tl) gamma-ray spectrometer J. Env. Radioactivity 51 255-65

[13] Yu K N, Guan Z J, Stoks M J and Young E C 1992 The assessment of natural radiation dose committed to the Hong Kong People J. Env. Radioactivity 17 p.31-48

[14] Turham S, Baykan U N and Sen K 2008 Measurement of the natural radioactivity in building materials used in Ankara and assessment of external doses J. Radiological Prot. 28 p.83-91

[15] Beretka J and Mathew P J 1985 Natural radioactivity of Australian building materials, waste and by-products J. Health Physics 48 p.87-95

[16] Tholkappian M, Ganesh D, Devanesan E, Harikrishnan N, Jebakumar J P P and Ravisankar R 2018 Data on natural radioactivity and associated radiation hazards in coastal sediment of Chennai Coast Tamilnadu India using gamma ray spectrometry J Data in Brief 17 p.551-58 ScienceDirect

[17] Quindos L S, Fernandez P L and Soto J 1987 Building material as source of exposure in houses J. Indoor Air 87(2) p.365