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Note

Dose Assessment of Radium-226 in Drinking Water from Mamuju, a High Background Radiation Area of Indonesia

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Mamuju is an area of Indonesia with high radiation exposure compared to the average across the country. It is included in the high background radiation area (HBRA) category. Radium-226 (^{226}Ra) is one of the natural radionuclides that, if contained in drinking water, can be harmful to human health. Mamuju's residents generally use well water for their daily needs. Radium-226 is easily soluble in water and emits alpha particles. Therefore, measurement of ^{226}Ra in drinking water is necessary to protect the public from radiation. A total of 13 drinking water samples were obtained from the HBRA in Mamuju. They had a concentration range of 14–238 mBq L^{-1}. These concentrations are below the World Health Organization recommendation, which is 1 Bq L^{-1}, with an annual effective dose (mSv y^{-1}) from the ingestion of ^{226}Ra in water ranging between 3–49 µSv.

Key words: ^{226}Ra , dose assessment, HBRA

1. Introduction

Natural radiation has the biggest contribution to the radiation exposure of humans. Natural radiations come from space (cosmic rays) and natural radioactive substances in the earth's crust layer (primordial radionuclides), which have existed since the earth's formation. Based on the Indonesian Radiation Level and Environmental Radioactivity Map published by Pusat Teknologi Keselamatan dan Metrologi Radiasi (PTKMR)-Badan Tenaga Nuklir Nasional (BATAN) in 2013, it was found that Mamuju city in West Sulawesi Province has an area of high natural radiation, with an average that is six times greater than levels in other regions in Indonesia, or about 40 nSv h^{-1} [1)].

Exposure to natural radiation comes from external exposure – cosmic radiation and terrestrial gamma radiation – and internal exposure – inhalation and ingestion[2)]. Radium-226 (^{226}Ra) is radioactive and emits alpha particles, and thus it is harmful to humans when inhaled or ingested. It is a product of the Uranium-228 (^{238}U) radionuclide decay series, with a half-life of 1,600 years. Among the radionuclides present in groundwater, the most radiotoxic and of greatest concern for human health is ^{226}Ra. In communities where wells are used, drinking water can be a major source of ingested radium because ^{226}Ra can be easily dissolved in water, and bone

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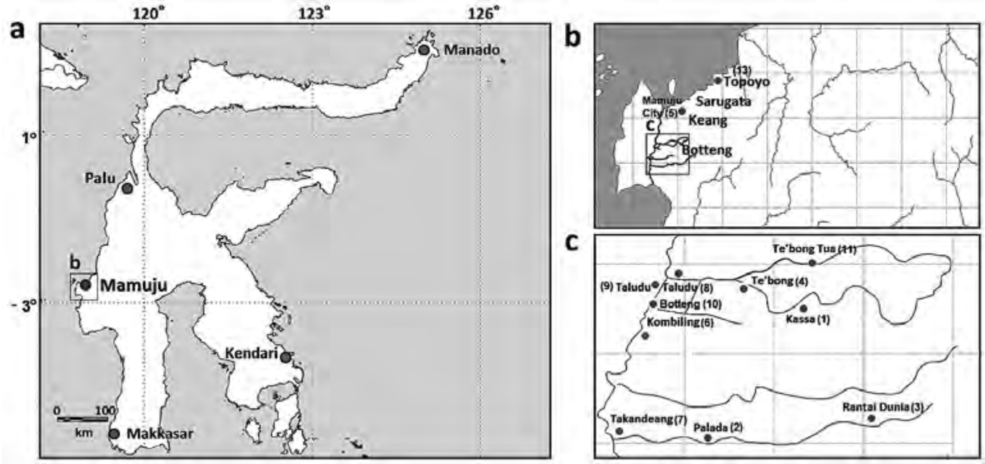

Fig. 1. The study area, showing (a) the Sulawesi island map and the location of Mamuju city, (b) the district map of Mamuju, which is the sampling point of the drinking water sample, and (c) the hamlet map for sampling points of drinking water.

becomes the principal repository for radium in the body. The fundamental reason for this is the chemical similarity between calcium and radium, and this can cause bone cancer[3, 4]. Mamuju's residents generally use well water for their daily needs. Therefore, determining the level of ^{226}Ra in water samples from Mamuju is necessary to protect the population.

Radium-226 in groundwater comes from its interaction with ^{226}Ra-bearing materials such as rocks, soil, ore bodies, and other source materials. Radium-226 in freshwater comes from groundwater inflow, sediment resuspension, resolubilisation of sediment-bound radionuclides and from the air through precipitation and particle deposition. While ^{226}Ra activity concentrations have a wide range in groundwater, depending upon the source characteristics of a region and the ionic strength of the groundwater, ^{226}Ra activities in surface water are low and lie within a relatively narrow range of concentrations[5].

2. Experimental Method

Study area

Mamuju city lies in the Province of West Sulawesi at 2° 50´110 ˝ – 2° 29´ 552 ˝ south latitude, and 118° 38´ 47 ˝ – 118° 96´ 35 ˝ east longitude (Fig. 1). Administratively, the territory consists of provinces, cities, districts, villages and hamlets. Water samples were obtained from the Mamuju Takandeang and Botteng villages, among others, and from several hamlets such as Botteng, Taludu, Te'bong Tua, Te'bong, Kassa, Palada, Takandeang, Mamuju city, Rantai Dunia, and the Kombiling hamlets. For a control area, Topoyo, around 120 km from Mamuju city, was used. Bottled mineral water, which is also often consumed by residents, was included in the water samples.

Sample collection and preparation

A total of 13 samples, measuring 1 litre each, were collected using PET bottles. Then, measures were taken of the temperature, pH, and conductivity (Laquatwin, Horiba, Japan) of the water, as well as the ambient dose rate (PDM 111, Hitachi, Japan) around the sampling area. After that, the water samples were filtered using a 0.45 µm pore membrane and adjusted for pH 2 with the addition of 65% of nitric acid. The preparation stage was then carried out, with the sample being weighed at 200 g (m_1), and evaporated at around 60 °C until the weight of the sample was approximately 20 g (m_2). The purpose of acidifying, heating, and evaporating the samples in the preparation phase is to desorb the already existing ^{222}Rn and to achieve a better detection limit[6]. Radon was extracted from the aqueous solution using an immiscible scintillation cocktail, with water inside the scintillation vial[7, 8]. A sample of approximately 10 mL was transferred into a high-performance glass vial (PerkinElmer, USA) and weighed (m_3). After that, a 10 mL scintillation cocktail (Ultima Gold uLLT, PerkinElmer, USA) was added, and the mixture was allowed to stand for 30 days to reach equilibrium with the progeny isotopes. The formula for the sample mass (m) is as follows:

$$m = \frac{m_1 m_3}{m_2} \tag{1}$$

Calibration and Measurement

Five ^{226}Ra standard solutions were prepared with various concentrations: 0.1, 0.5, 1, 3, and 5 mBq. After 30 days, the samples were measured by LSC (Quantulus 1220), applying alpha and beta discrimination: only the alpha emissions of ^{222}Rn and its short-lived progeny (^{218}Po, ^{214}Po) were considered, as this counting condition ensures a

Table 1. Drinking water properties and radiation around the sampling area

No	Area	Type of water	Temperature (°C)	pH	Conductivity (mS cm^{-1})	Dose rate (nSv h^{-1})
1	Kassa	well	25.3	7.0	0.16	585 ± 60
2	Palada	well	26.1	7.1	0.04	2,130 ± 280
3	Rantai Dunia	spring	25.1	6.2	0.08	763 ± 77
4	Tebong Tua	well	27.1	7.4	0.09	790 ± 79
5	Mamuju	tap	25.9	6.9	0.39	223 ± 23
6	Kombiling	well	25.1	7.0	0.27	600 ± 60
7	Takandeang	well	25.9	7.2	0.61	550 ± 55
8	Taludu	well	26.5	6.6	0.25	580 ± 58
9	Taludu	well	25.5	7.4	0.29	700 ± 70
10	Botteng	well	25.5	7.3	0.08	562 ± 57
11	Tebong Tua	well	25.5	7.3	0.83	770 ± 77
12	Bottled*	spring	25.2	6.8	0.16	-
13	Topoyo	tap	27.1	6.7	0.30	70 ± 7

*Bottled mineral water

better detection limit. Then, the detection efficiency was calculated using the formula below:

$$\varepsilon = \frac{r_s - r_0}{a\, m_s} \qquad (2)$$

where ε is alpha efficiency, r_s is count rate in the alpha window (s^{-1}), r_0 is blank sample count rate in the alpha window (s^{-1}), a is massic activity of the standard solution (Bq g^{-1}), and m_s is mass of the ^{226}Ra standard solution used for the preparation of the calibration sample (g).

After 30 days, the concentrated sample was then transferred into the vial with the scintillation cocktail. Then, to calculate the concentration of ^{226}Ra in the sample, the following formula was used:

$$C_A = \frac{r_g - r_0}{\varepsilon\, m}\,\rho \qquad (3)$$

where C_A is activity concentration (Bq L^{-1}), r_g and r_0 are samples and blank gross count rate in the alpha window (s^{-1}), ε is alpha efficiency, m is mass of the test sample (g), and ρ is the density of sample (g L^{-1}). The detection limit of this measurement was 1 mBq L^{-1}, with a background count rate of 0.1 s^{-1}, and the counting time was 3,600 s.

3. Result and Discussion

The results show that the water in the Mamuju area is suitable for drinking, with a pH range of 6.6–7.4, and conductivity of 0.04–0.83 mS cm^{-1}. The pH, conductivity, temperature, and radiation exposure in the sampling area were also measured, as shown in Table 1. Although pH usually has no direct impact on consumers, it is one of the most important operational water-quality parameters, with the optimum pH being in the range of 6.5–9.5. Conductivity in drinking water indicates the presence of minerals such as magnesium flour, sodium, and calcium. These minerals are needed by the body to help metabolic processes. However, conductivity over 250 mS might cause kidney damage, because metabolic waste minerals will be processed by the kidneys and may result in renal stones / urolithiasis.

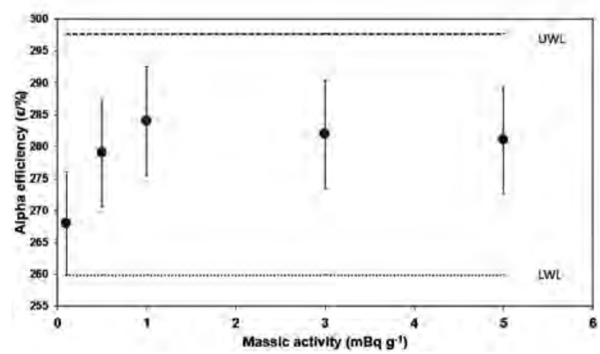

Fig. 2. The quality control chart for the calibration of ^{226}Ra standard solution. The upper line is the upper warning level (UWL) and the lower line is the lower warning level (LWL).

For quality control and calibration, five different concentration ^{226}Ra standards were prepared and measured in the vial, with the scintillation cocktail added, and this was measured after 30 days using LSC (Fig. 2). A control chart is a graph that can help a "process manager" to make better decisions regarding the actions necessary to achieve the best performance from a process. From the control chart in this study, it can be concluded that the measurement results of several standard solutions have good values. The centre line is drawn at the mean of the data. The lower and upper warning lines, labeled LWL and UWL, are placed at a distance of two standard deviations from the centre line.

The ^{226}Ra concentrations in the drinking water samples from Mamuju and the estimated internal doses for the ingestion of ^{226}Ra are shown in Table 2. This table shows that the radium concentration has a range of 8–238 mBq L^{-1}, which is below the World Health Organization recommendation. The highest concentration is from a well in the Tebong Tua area, at 238 mBq L^{-1}. For the control area and for the bottled mineral water, the observed ^{226}Ra concentrations were quite small, at around 14 mBq L^{-1}.

Pearson's correlation analysis was used to determine the relationship between ambient dose rates at sampling points and the concentration of ^{226}Ra in water (Fig. 3). The results show that radium concentration in drinking water has a weak correlation with ambient dose rates, with Pearson's coefficient at 0.3 and R^2 at 0.09. The presence of radium in water is strongly influenced by the content of radionuclides in the soil. The high ambient dose rate at the sampling points indicate that Mamuju has a HBRA, which is related to the radiation exposure from ^{238}U, ^{232}Th and ^{40}K contained in the soil. Small ^{226}Ra concentrations were observed in drinking water samples, while other studies suggest that soils in the Mamuju area have high ^{226}Ra concentrations[9].

The dose calculation from radium in drinking water is calculated by equation (4):

$$D = D_{WI} \times D_{CF} \times Y \times C_A \quad (4)$$

where D is the annual effective dose (μSv) from the digestion of ^{226}Ra in the water, C_A is the radium concentration in the water (Bq L^{-1}), D_{WI} is the daily consumption of water (estimated as 2 L d^{-1})[3], D_{CF} is the dose conversion factor for adults, (2.8×10^{-7} Sv Bq^{-1}), and Y is the ingestion period (365 d)[10]. In all calculations, the uncertainties are given within one standard deviation.

Based on calculations to determine the annual effective dose caused by ^{226}Ra through the ingestion of drinking water in the area, the results obtained for Mamuju are in the range of 3–49 μSv.

Table 2. Ra-226 concentrations in drinking water and annual effective dose due to ingestion

No	Area	^{226}Ra concentration (mBq L^{-1})	Annual effective dose (μSv)
1	Kassa	41 ± 2	8
2	Palada	133 ± 7	27
3	Rantai Dunia	117 ± 6	24
4	Tebong Tua	102 ± 5	21
5	Mamuju	75 ± 5	15
6	Kombiling	60 ± 4	12
7	Takandeang	89 ± 5	18
8	Taludu	51 ± 3	10
9	Taludu	15 ± 1	3
10	Botteng	235 ± 14	48
11	Tebong Tua	238 ± 14	49
12	Bottled*	14 ± 1	3
13	Topoyo	14 ± 1	3

*Bottled mineral water

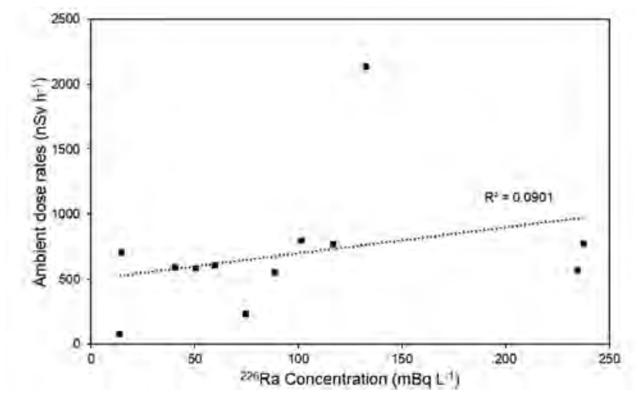

Fig. 3. Pearson's correlation analysis between ambient dose rates at sampling points and the concentration of ^{226}Ra in water.

4. Conclusion

In this study, ^{226}Ra concentrations from 14 to 238 mBq L^{-1} were observed, while other studies suggest that soils in the Mamuju area as HBRA have high ^{226}Ra concentrations. This result is below the limit of 1 Bq L^{-1} set out by the World Health Organization recommendation, with an annual effective dose range of 3–49 μSv.

Author Contributions

Conceptualization, E.D.N.,and S.T.; methodology, E.D.N., M.H. and S.T.; validation, E.D.N., N.A., and M.H.; formal analysis, E.D.N., and Y.T.; investigation, T.S., K., A.P., I.D.W., Y.T., E.D.N.; data curation, A.P., and E.D.N.; writing — original draft preparation, E.D.N.; writing — review and editing, E.D.N., Y.T., A.P., M.H., N.A., K., I.D.W., T.S and S.T.; visualization, E.D.N.; supervision, E.D.N., M.H., and S.T.; funding acquisition, S.T. All authors have read and agreed to the published version of the manuscript.

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Conflict of Interest

The authors declare no conflict of interest.

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