Distribution of some natural and anthropogenic radionuclides in the sediments and seawater along the coastal areas of North Sulawesi

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Abstract. Studies on the radionuclide distribution of some naturally occurring radioactive materials (NORM) and anthropogenic radionuclides in the sediments of the coastal areas of North Sulawesi were conducted to provide baseline data for the effective monitoring of radioactive fallout in these areas. Sediment samples were collected by a van Veen grab samples about 100 – 300 m from the shoreline, and 60 L of seawater was pre-concentration to precipitated 137Cs. Activity concentration of the three main natural radionuclides (222Ra, 232Th, and 40K) and 137Cs were determined using a high-resolution HPGe gamma-spectroscopy system and presenting background about the radiological levels and assessing the associated hazards. The activity of 226Ra ranged from 2.1 to 9.5 Bq kg⁻¹ with an average value of 5.9 Bq kg⁻¹. The activity of 232Th varies from 2.4 to 10.4 Bq kg⁻¹ with an average value of 5.2 Bq kg⁻¹. The 40K ranges between 169.1 to 492.7 Bq kg⁻¹ with an average value of 238.8 Bq kg⁻¹. The average activity concentration of 40K (238.8 Bq kg⁻¹) in the sediment samples was lower than the worldwide average concentration (420 Bq kg⁻¹). The current level of activity 137Cs varied from 0.05 to 0.40 Bq kg⁻¹ in sediment samples and 0.98 to 1.33 Bq m⁻³ in seawater samples. 134Cs in all samples were not detected or below the detection limit. This fact indicated that radioactive cesium in Celebes Sea areas of North Sulawesi still originated from global fallout and insignificant influenced by the FDNPP accident. All radionuclide radiation values in the recent study are lower than the world average. Therefore, the potential danger of radiation generated from the surrounding environment has not yet caused a radiological health impact for the people living on the coast of North Sulawesi. Natural and artificial radionuclide activity data in this study will be used as the basis for sedimentary activity along the coast of North Sulawesi.

1. Introduction

Natural radioactive materials have existed for a long time since the earth's formation, and it was generally contained primordial radionuclides. Most of these radioisotopes in nature were form as heavy elements, and there are about 70 radionuclides exist in all environment media as soils, rocks, sediments, water (ocean, lake, and rivers), and air from the total of about 340 elements [1]. Natural occurring radionuclides were present everywhere in the earth's environment, and throughout the geological time, the concentration amount of the radioactive materials was assumed to be increased, such as 238U, 232Th, and 40K [2]. In a marine environment, natural radionuclides were formed and transported by geological processes in terrestrial rocks as weathering, erosion, and mineral recycling. Furthermore, the sedimentation processes in the aquatic environment play an important role in the accumulation and distribution of radionuclides within a geographic area [3], [4]. On the other hand, artificial radionuclides present in the environment was introduced by anthropogenic sources from
global fallout generated by atomic weapon tests, discharge from fuel reprocessing plants, and the accident of nuclear power plant facilities in Chernobyl (1986) and Fukushima Dai-ichi NPP (2011) [5], [6], [7].

Radioactive materials already present in the environment are the primary source of radiation exposure from Earth to human populations, and cosmic radiation from outer space also contributes to surface radiation with muon energies between 1 and 20 GeV [8]. Radiation from the decay of radioactive materials such as alpha, beta, and gamma becomes the primary source of background radiation. Alpha radiation sources are dominated by the decay of uranium isotopes (\(^{235}\text{U}\) and \(^{238}\text{U}\)), and the thorium series (\(^{232}\text{Th}\)) are the primary sources of beta decay followed by gamma-ray excitation. Beta radiation is mainly caused by the decay of \(^{40}\text{K}\), which significantly impacts natural radioactivity, followed by the decay of gamma-ray emission energy of 1.46 MeV. The concentration of environmental radiation activity mostly comes from prominent series decay (\(^{212}\text{Th}\) and \(^{226}\text{Ra}\) and \(^{40}\text{K}\)). Based on global data, the mean radiation activity in the sediments was 33, 33, 45, and 420 Bq kg\(^{-1}\) for \(^{238}\text{U}\), \(^{226}\text{Ra}\), \(^{232}\text{Th}\), and \(^{40}\text{K}\), respectively [8]. To estimate the total radiation exposure at the Earth's surface is necessary to add cosmic rays to the terrestrial radiation. For example, Car Nicobar Island, located in the Indian Ocean at 0 meters above sea level, has the lowest radiation dose level of 27-32 nGy h\(^{-1}\), while the highest radiation dose is at the top of Mount Everest at an altitude of 7,987 m above sea level of 502 nGy h\(^{-1}\) [5], [9]. In addition, the external average exposure rates from terrestrial gamma radiation were reported about 53 nGy h\(^{-1}\) in Indonesia [10].

The impact of radiation exposure from the decay of natural radionuclides on human activities externally and internally in the long term can cause various health problems, such as acute leukopenia, anemia, leukemia, oral necrosis, tooth fractures, and cataracts, as well as lung, liver, pancreas, and liver cancer: bone, and kidney [11], [12]. Natural radioactivity from the environment and external exposure associated with gamma rays is highly dependent mainly on the geological conditions of the soil, geological activity, and sedimentary formations in each region of the world [13]. In addition to occurring naturally, the distribution of natural radionuclides on the seabed can be used as a tracer for sediment transport mechanisms, composition, and accumulation of the seabed. The distribution of radionuclide activity in marine sediments can provide a better critical information to understand oceanographic and sedimentation processes [14], [15].

Therefore, the determination of natural radioactivity and investigation of gamma-ray exposure from the coastal areas should be monitoring. This study was reported the activity concentration of \(^{226}\text{Ra}\), \(^{232}\text{Th}\), \(^{40}\text{K}\) and \(^{137}\text{Cs}\), for coastal sediment and seawater to provide for estimation monitoring of the radiation exposure to the population around the coastal area. The main objective of this study is to evaluate the natural radioactivity and radiological hazards in sediment and seawater samples distributed along the coastal area of North Sulawesi, Indonesia. This coastal area is an important site because it was a central area for economic activity, fisheries, agriculture, and specifically, favorite recreational location in North Sulawesi. The radiological parameter hazard is determined by calculating radium equivalent activity (Ra\(_{eq}\)), external hazard (H\(_{ex}\)), internal hazard index (H\(_{in}\)), and annual dose rate (D) both from natural and artificial radionuclides to the population near the sampling site. This study would be used as baseline data on the level of natural radioactive background.

2. Materials and methods

2.1 Study area

North Sulawesi mainland is almost entirely covered by mountains, hills, and valleys landscape with a considerable climate variation. The geographical location was located at 0.3 – 4.3 North Latitude and 121 – 127 East Longitude in the northern peninsula of Sulawesi Island. The study site was located at the Celebes Sea along the coast of North Sulawesi and the Molucca Sea in the Southeast. The coastal area of Manado, Bitung, and Gorontalo capital city with a population of 451,916; 225,134; and 198,539, respectively, at the 2020 Census, was considered as a sampling area [16]. Bunaken National Park is a cluster of coral islands located near the center of the famous Coral Triangle and is a prevalent location in North Sulawesi. The number of visitors in this tourist area ranges from 32,000-39,000 [17], so it is essential to evaluate the potential hazard of environmental radiation exposure. The location of sampling and this research are shown in Figure 1.
2.2 Sample collection and preparation
A total of 6 sediments and three seawater samples were collected along the coastal area of North Sulawesi in August 2018. The distance of the sampling site was around 100-300 m from the coastline of each coastal zone. 1 kg of the sediment samples was collected using van peen sediment grab, and 60 L of seawater was collected using an 80 L plastic bucket during low tide between 10 am to 2 pm. The sediment samples were stored in plastic bags and labeled at the time of collecting. The $^{137}$Cs in seawater were prepared by preconcentration using 10 gr of K$_4$Fe(CN)$_6$ and 10 gr CuSO$_4$ to precipitated $^{137}$Cs at pH ~8.0. After settled for a night (6 – 12 h), the precipitate was filtered using 0.1 µM filter paper. At the laboratory, the sediment samples were drying in the oven at a temperature (~120 ℃). The filter paper of precipitated $^{137}$Cs was set at ~80 ℃ to dry. The dried sample was homogenized using a grinder, sieved to less than 1 mm, and filter paper was a store in a specific plastic container. 1 kg of sediment sample was transferred to the close cap plastic tube with a volume of 1.2 L and sealed to prevent air leak for about 30 days to reach an equilibrium condition between $^{226}$Ra – $^{222}$Ra, and its decay products before counted by gamma spectrometry.

2.3 Gamma spectroscopic analysis
The activity concentrations of $^{226}$Ra, $^{232}$Th, $^{40}$K, and $^{137}$Cs in the samples were determined using an HPGe planar detector with an energy resolution between 1.8 keV and 1332 keV. The gamma-ray spectrum was analyzed using MCA (multi-channel analyzer) software. The efficiency of the gamma detector was calibrated using standard multiple gamma sources for $^{137}$Cs calculated at a peak energy of 662 keV and for $^{60}$Co using two peaks at 1173 and 1332 keV energies. The detection efficiency of each radionuclide spectra peak was determined by comparing the reference material (IAEA-375) with the known weight and geometry of the sample and standard. The detector was covered, and caps with 10 cm thick of low-level background leads shield to reduce the background radiation. The activity of $^{226}$Ra was determined using gamma spectra at energy 351.9 keV (37.6%) for $^{214}$Pb and 609.3 keV (46%) for $^{214}$Bi. $^{232}$Th radioactivity was analyzed from energy 911.2 keV for of $^{228}$Ac and 238.6 keV for $^{212}$Pb. $^{40}$K activity and $^{137}$Cs were measured directly through its gamma-ray energy peak of 1460.8 keV (10.7%) and 661.6 keV (84.9%), respectively.

The minimum detectable activity (MDA) for each radionuclide was calculated using following equation:

\[
\text{MDA} = \frac{B}{\text{E}}
\]

where B is the background count rate and E is the efficiency of the detector.
The lower limit detection (LLD) was calculated using Compton (F_c) background area with a 96% confidence of selected gamma-ray spectrum in equation: LLD = 4.66 \sqrt{t \times P_g \times ef \times m}

The gamma radiation hazards

To understand the influence of gamma radiation on the population around sampling sites, some radiological parameters, such as radium equivalent activity, external and internal hazard index, and absorbed dose rate both from natural and artificial radionuclides, were calculated to evaluate the potential radiological hazards and assess the radiation risk to human. The radium equivalent activity (Raeq) was calculated to evaluate and compare gamma radiation from different mixtures of activity concentration of 226Ra, 232Th and 40K in the sediment. The radium equivalent activity was calculated using the equation below [18].

\[ Raeq (\text{Bq kg}^{-1}) = A_{Ra} + 1.43 A_{Th} + 0.077 A_K \] (1)

The potential radiological hazard caused by different radionuclides was estimated as the external hazard index (H_{ex}) and a standard safety for terrestrial radiation materials was H_{ex} \leq 1. The external hazard index was obtained from the calculation (2) [19].

\[ H_{ex} = \left( \frac{A_{Ra}}{370} \right) + \left( \frac{A_{Th}}{259} \right) + \left( \frac{A_K}{4810} \right) \leq 1 \] (2)

The internal hazard index (H_{in}) was indicate for the presence of radon and its short-lived decay products that hazardous to the respiratory organs. The H_{in} was express using the following mathematic equation (3).

\[ H_{in} = \left( \frac{A_{Ra}}{185} \right) + \left( \frac{A_{Th}}{259} \right) + \left( \frac{A_K}{4810} \right) \leq 1 \] (3)

To estimate the level of gamma radiation hazard associated with natural gamma emitters in the sediments, the representative level index of gamma radiation (I_{yr}) of the sediment can be used [20]. The gamma activity concentration index (I_{yr}) is calculated from the equation below [21].

\[ I_{yr} = \left( \frac{A_{Ra}}{300} \right) + \left( \frac{A_{Th}}{300} \right) + \left( \frac{A_K}{3000} \right) \leq 1 \] (4)

where \( A_{Ra}, A_{Th} \) and \( A_K \) (Bq kg\(^{-1}\)) are the activity concentration of 226Ra, 232Th, and 40K, respectively. The dose rate of environmental exposure associated with radiation in each coastal zone was carried out by estimating the total gamma dose rate of natural and artificial radionuclides, and the annual effective dose using equation (5) to (7), respectively [5]:

\[ D_{natural} = (0.427 \times A_{Ra}) + (0.662 \times A_{Th}) + (0.043 \times A_K) \] (5)

\[ D_{total} = (0.427 \times A_{Ra}) + (0.662 \times A_{Th}) + (0.043 \times A_K) + (0.03 \times A_{Cs}) + 53 \] (6)

\[ ED_{outdoor} = D_{total} \times T \times 0.2 \times 0.7 \times 10^{-3} \] (7)

where \( A_{Ra}, A_{Th}, A_K \) and \( A_{Cs} \) (Bq kg\(^{-1}\)) are the activity concentration of 226Ra, 232Th, 40K and 137Cs, respectively. The dose rate of gamma radiation at ground level was determined by \( D_{out} \) (nGy h\(^{-1}\)), and the dose rate of cosmic radiation exposure in the terrestrial region in Indonesia was 53 nGy h\(^{-1}\) for areas 1 m above sea level [10]. The natural dose rate of radioactive material was determined by \( D_{natural} \) (nGy h\(^{-1}\)).
and ED (µSv·y⁻¹) effective dose rate, the number 0.7 is the conversion factor for (Sv Gy⁻¹), 0.2 is the external occupancy factor, and T is the hours’ exposure time within a year (h y⁻¹). The main contribution of radiation exposure received by coastal residents was experienced mostly from people spending their time for work on the beach or just vacation activities. Therefore, the dose rate received by the population was classified into two groups: the people who work daily on the beach and tourists. The worker group was assumed to average spending their time about eight-hour per day for 18 weeks and the tourist group was assumed of 4 weeks and daily eight-hour spending at the beach. The outdoor occupancy factor (T) was estimated to be 940-1010 h y⁻¹ for workers and around 220 h y⁻¹ for tourists.

### Table 1. Sampling location, coordinates and activity concentration of ²²⁶Ra, ²³²Th, ⁴⁰K and ¹³⁷Cs in sediment with the results of radiological parameters in terms of coastal zones

| Station | Location | Activity concentration (Bq kg⁻¹) | Radiological parameters hazard |
|---------|----------|---------------------------------|-------------------------------|
|         |          | ²²⁶Ra  | ²³²Th | ⁴⁰K   | ¹³⁷Cs | Hₑ₀ | Hₑ₁ | Iₑ |
| ST-1    | 123°03′41.4″ E 0°02′45.8″ N | 7.8 ± 0.3 | 6.1 ± 0.3 | 169.1 ± 6.4 | 0.28 ± 0.09 | 121.07 ± 0.55 | 0.08 | 0.10 | 0.11 |
| ST-2    | 123°28′30.1″ E 0°09′26.8″ N | 9.5 ± 0.4 | 10.4 ± 0.4 | 492.7 ± 18.0 | 0.33 ± 0.09 | 575.03 ± 1.19 | 0.17 | 0.19 | 0.25 |
| ST-3    | 125°08′22.4″ E 0°12′15′31.8″ N | 4.3 ± 0.3 | 2.7 ± 0.3 | 236.9 ± 8.8 | 0.16 ± 0.11 | 74.31 ± 0.54 | 0.07 | 0.08 | 0.11 |
| ST-4    | 125°59′12.0″ E 0°17′36.12″ N | 4.0 ± 0.3 | 2.4 ± 0.3 | 248.3 ± 9.2 | 0.05 ± 0.11 | 70.15 ± 0.57 | 0.07 | 0.08 | 0.11 |
| ST-5    | 124°49′53.0″ E 0°13′30′2.7″ N | 7.5 ± 0.3 | 5.8 ± 0.3 | 286.6 ± 10.6 | 0.09 ± 0.08 | 190.22 ± 0.69 | 0.10 | 0.12 | 0.15 |
| ST-6    | 124°42′10.9″ E 0°13′36′52.2″ N | 2.1 ± 0.2 | 3.6 ± 0.7 | BD   | 0.40 ± 0.07 | 2.14 ± 0.22 | 0.02 | 0.03 | 0.03 |
| min-max |          | 2.1-9.5 | 2.4-10.4 | 169.1-492.7 | 0.05-0.4 Bq kg⁻¹ | 2.14-575.03 | 0.02-0.17 | 0.03-0.19 | 0.03-0.25 |
| (mean)  |          | 5.9      | 5.2      | 238.8     | 0.22      | (172.15)     | (0.09)  | (0.10) | (0.12) |
| SD      |          | 2.8      | 3.0      | 160.5     | 0.14      | 206.9       | 0.05   | 0.06  | 0.07  |

SD: Below detection limit

### 3. Results and discussion

#### 3.1 Radionuclides activity concentrations in the sediments

The activity of natural radionuclides (²²⁶Ra, ²³²Th, ⁴⁰K) and artificial radionuclides of ¹³⁷Cs, together with their average values for sediment samples, are shown in Table 1. Both natural radionuclides and artificial radionuclides (¹³⁷Cs) could be determined in sediment samples collected from different locations except ⁴⁰K in ST.6 the activity was below the detection limit. All activity value of dry weight samples was given in Bq kg⁻¹. The activity range values of ²²⁶Ra, ²³²Th, ⁴⁰K and ¹³⁷Cs are 2.1-9.5, 2.4-10.4, 169.1-492.7 and 0.05-0.4 Bq kg⁻¹, respectively. The average activity of each radionuclide are 5.9, 5.2, 238.8, and 0.22 Bq kg⁻¹ for ²²⁶Ra, ²³²Th, ⁴⁰K and ¹³⁷Cs, respectively. In all samples, the activity concentration was in the order ⁴⁰K > ²³²Th > ²²⁶Ra > ¹³⁷Cs. The ⁴⁰K dominates over the other radioisotopes due to it was the most abundant in continental rocks, and it is elevated in many light minerals [22].

The mean values of the radioactivity levels were compared with the world average, 33, 45, and 420 Bq kg⁻¹ for ²²⁶Ra, ²³²Th, and ⁴⁰K, respectively, [5]. The average activity of ²²⁶Ra and ²³²Th in the sediment samples is significantly lower than the global average as 33 Bq kg⁻¹ for ²²⁶Ra and ²³²Th was 45 Bq kg⁻¹. Taking ¹³⁷Cs activity concentration into consideration and the existing short-lived radionuclides ¹³⁷Cs were not found in the sediments, indicating any evidence of the impact from the Fukushima Dai-ichi Nuclear Power Plant accident in 2011. The existence of ¹³⁷Cs in sediment samples might be originated mainly from the impact of the global fallout of the Chernobyl disaster in 1986 or nuclear weapon test.

The activity concentration of ¹³⁷Cs in the surface seawater were varied from 0.98 to 1.33 Bq m⁻³ with an average of 1.16 ± 0.1 Bq m⁻³ in the North of Manado City Area (Table 3). Also, a similar result from the previous study by Wu et al. (2020) [23] has reported that the activity concentration of ¹³⁷Cs in the South China Sea varied from 0.47 to 1.80 Bq m⁻³, with an average of 0.92 ± 0.28 Bq m⁻³. The distribution coefficient (Kₐ) of ¹³⁷Cs in sediment samples varied from ~ 300 – 410 L kg⁻¹, one order of magnitude lower than a report from IAEA, 2004 (4000 L kg⁻¹)[24]. The lower Kₐ value from this study is probably due to the low concentration of ¹³⁷Cs activity in sediment, which is also

5
consistent with data from various places globally, which show $^{137}$Cs activity on the coast of North Sulawesi is one order lower relatively than other places (Table 3).

The comparison of the average activity concentrations of $^{226}$Ra, $^{232}$Th, $^{40}$K, and $^{137}$Cs reported from various studies in the world at different locations is shown in Table 2. This study indicates that the average activity values for natural radionuclides $^{226}$Ra, $^{232}$Th, $^{40}$K are mostly lower than the world average and some other places. For activity, the concentration of $^{40}$K is slightly higher than that reported in the Gulf of Algeciras, Spain, and the Northern Peninsula, Malaysia. Varied radionuclides activity may be due to different geographical conditions for the geochemical composition contained in each region, such as the content of phosphate, nitrate, and carbonate [25]. Differences may also influence differences in natural radionuclide concentrations in mineral sources in the coastal area for each region.

Table 2. Comparison of activity concentration of radionuclides in the sediment samples worldwide.

| Location                        | $^{226}$Ra (Bq kg$^{-1}$) | $^{232}$Th (Bq kg$^{-1}$) | $^{40}$K (Bq kg$^{-1}$) | $^{137}$Cs (Bq kg$^{-1}$) | References                  |
|---------------------------------|---------------------------|---------------------------|-------------------------|---------------------------|-----------------------------|
| Brazil, Ilha Grande Bay         | 24                        | 44                        | 678                     | 0.5                       | Martins de Carvalho et al., 2016 |
| Saudi Arabia, Arabian Gulf coast| 26.4                      | 16.3                      | 351                     | 2.16                      | Fatimh Alshahri., 2016       |
| Turkey, Marmara Sea             | 23.8                      | 23.2                      | 558.6                   | 9.14                      | Otansev et al., 2016         |
| Iran, Oman Sea                  | 14.96                     | 17.61                     | 361.6                   | 0.79                      | Darabi-Golestane, F. et al., 2017 |
| Russia, White Sea Basin         | 9.7                       | 9.2                       | 337.1                   | 1.8                       | Kiselev et al., 2018         |
| USA, Mission                    | 33.7                      | 31.9                      | 300                     | 3.7                       | Hannan et al., 2015          |
| Ghana, Greater Accra            | 14                        | 30                        | 325                     | 1.5                       | Botwe et al., 2017           |
| Spain, Bay of Algeciras         | 12.1                      | 15                        | 188                     | -                         | Gonzalez-Fernandez et al., 2012 |
| Malaysia, Northern peninsular   | 51                        | 22                        | 189                     | -                         | Muhammad et al., 2012        |
| Egypt, Red sea Marine sediment  | 27.4                      | 38.5                      | 419.4                   | -                         | El-Taher and Madykour, 2011  |
| Indonesia, Bali and Lombok      | 25.6                      | -                         | 169.4                   | 0.16                      | Putra et al., 2017           |
| Indonesia, North Sulawesi       | 5.9                       | 5.2                       | 238.8                   | 0.22                      | Present Study, 2021          |
| Worldwide                       | 33                        | 45.0                      | 420                     | -                         | UNSCEAR, 2000                |

3.2 Evaluation of radiological hazard effects

Radiological parameters in the sample sediments and the average value of the gamma radiation hazard index are shown in Table 1. The radium equivalent activity (Ra$_{eq}$) in the sediment samples in this study area ranged from 2.14-575.03 Bq kg$^{-1}$, with an average of 172.15 Bq kg$^{-1}$. These results indicate that the activity was smaller than the limit of 370 Bq kg$^{-1}$ recommended by UNSCEAR. (2000). The radium equivalent activity can be used to compare the different radioactivity content of sediments. The value of Ra$_{eq}$ activity in Table 1 shows a distribution pattern similar to $^{40}$K activity. This similarity indicates that $^{40}$K mostly dominates the radioactivity content of sediments in the coastal areas of North Sulawesi, with a maximum at Station 2.

Table 3. Concentration of $^{137}$Cs and $K_d$ in seawater samples in Celebes Sea, North Sulawesi

| Station | Location                  | $^{137}$Cs Activity (Bq m$^{-3}$) | $K_d$ (L kg$^{-1}$) |
|---------|----------------------------|-----------------------------------|---------------------|
| W-1     | 124 47° 9.2" BT 01 33° 6.5" LU | 0.98 ± 0.09                       | 4.1 x 10$^2$       |
| W-2     | 124 44° 9.3" BT 01 35° 49.7" LU | 1.33 ± 0.1                        | 3.0 x 10$^2$       |
| W-3     | 124 41° 51.9" BT 01 36° 8.5" LU | 1.16 ± 0.10                       | 3.4 x 10$^2$       |

The main purpose of calculating this radiological hazard index is to determine the potential risk caused by radioactive materials. The safety limit for this hazard index must be less than one, which is the maximum dose of exposure to individuals for one year. The external hazard concentration index,
internal hazard, and gamma activity ranged from 0.02-0.17, 0.08-0.19, and 0.03-0.25, respectively, with an average $H_{ex}$ (0.09), $H_{in}$ (0.10), and $I_{yr}$ (0.12). The index value of this study is less than 1 (< 1), and it indicates that the sediments in this area have little effect on causing harm to people living in the surrounding area near the sampling site due to the low level of gamma dose in all samples under investigation.

3.3 Annual effective dose equivalent (AEDE)

The exposure dose rate of natural and artificial radionuclides contained in sediment samples was estimated at $3.3 - 32.1$ nGy h$^{-1}$ with an average of $16.2$ nGy h$^{-1}$, while the total gamma dose rate due to terrestrial and cosmic rays were $56.3 - 85.1$ nGy h$^{-1}$ with an average value of $69.2$ nGy h$^{-1}$. The exposure level of gamma-ray was shown in Figure 2. The calculation results of gamma dose rate from the sediment samples were lower than the world average dose rate (55 nGy h$^{-1}$); however, the average dose rate of the total background radiation (terrestrial and cosmic-ray) was calculated to be slightly higher than the worldwide average. These results indicated that radiation from space has a significant effect on total background radiation. The analysis results of the effective dose derived from gamma-ray can be seen in Table 4. The annual effective dose received by residents as a worker is between 7.4 – 11.2 $\mu$Sv y$^{-1}$ with an average of 9.1 $\mu$Sv y$^{-1}$ and 1.7 – 2.6 $\mu$Sv y$^{-1}$ for tourist doses with an average of 2.1 $\mu$Sv y$^{-1}$, respectively. The highest values of gamma dose rates and annual effective dose were found in the coastal zone Station 2, and the lowest value was found near Manado Tua Island at station 6. within the shallow part, with depths of about 1-2 m.

| Station | $D_{natural}$ (nGy h$^{-1}$) | $D_{total}$ (nGy h$^{-1}$) | ED Annual effective dose ($\mu$Sv y$^{-1}$) |
|---------|--------------------------|--------------------------|---------------------------------|
| ST-1    | 14.6                     | 67.7                     | 8.9                             |
| ST-2    | 32.1                     | 85.1                     | 11.2                            |
| ST-3    | 13.8                     | 66.8                     | 8.8                             |
| ST-4    | 14.0                     | 67.0                     | 8.8                             |
| ST-5    | 19.3                     | 72.3                     | 9.5                             |
| ST-6    | 3.3                      | 56.3                     | 7.4                             |
| Average | 16.2                     | 69.2                     | 9.1                             |
| Median  | 14.3                     | 67.3                     | 8.9                             |
| SD      | 9.4                      | 9.4                      | 1.2                             |

Table 4. Gamma dose rate and annual effective dose rate in coastal sediment samples of North Sulawesi

3.4 The Pearson’s correlation

The Pearson correlation coefficient method showed the correlation between natural and artificial radionuclides ($^{137}$Cs) with radiological hazard parameters. In Table 5, it can be seen that the relationship between the activity concentrations of $^{226}$Ra and $^{232}$Th showed a significant positive correlation with the value of $R^2 = 0.875$. This result has similarity with correlation investigation by [26][27], who reported a strong relationship between the decay of $^{226}$Ra and $^{232}$Th, which occur together in nature. Furthermore, the correlation between $^{137}$Cs with $^{226}$Ra and $^{232}$Th shows a weak relationship with values of $R^2$=0.034 for $^{226}$Ra and 0.421 for $^{232}$Th. This correlation indicates differences in sources that occur in nature, as we already know that $^{137}$Cs is produced from human activities such as nuclear weapons testing, operation of nuclear facilities, and nuclear power plant accidents. All investigated radiological parameters ($R_{eq}$, $H_{ex}$, $H_{in}$, $I_{yr}$, $D_{natural}$, $ED_{total}$ and annual dose rate) showed a positive and significant correlation with the activity of $^{226}$Ra, $^{232}$Th, and $^{40}$K, as these parameters are directly linked to these radionuclides known as the primarily gamma-emitting isotopes in nature. However, the correlation between the artificial isotope of $^{137}$Cs showed negative value to the radiological parameters, indicating that the activity concentration of $^{137}$Cs in the environment did not significantly contribute to increasing radiation background.
4. Conclusions

In this study, specific activity in 6 sediment samples and three seawater samples was determined using a gamma spectrometer to determined radionuclides concentration of $^{226}$Ra, $^{232}$Th, $^{40}$K, and $^{137}$Cs. The activity of $^{226}$Ra ranged from 2.1 to 9.5 Bq kg$^{-1}$ with an average value of 5.9 Bq kg$^{-1}$. The activity of $^{232}$Th varies from 2.4 to 10.4 Bq kg$^{-1}$ with an average value of 5.2 Bq kg$^{-1}$. $^{40}$K ranges between 169.1 to 492.7 Bq kg$^{-1}$ with the average value of 238.8 Bq kg$^{-1}$. The $^{137}$Cs varied from 0.05 to 0.40 Bq kg$^{-1}$ in sediment samples and 0.98 to 1.33 Bq m$^{-3}$ in seawater samples. Measurements of all investigated samples show that the mean values of radioactivity levels are lower than the world averages for $^{226}$Ra, $^{232}$Th, $^{40}$K, and $^{137}$Cs. For radiological parameters also show results that are lower than the world average. The results of this study showed that the radium equivalent activity ranged from 2.14-575.03 Bq kg$^{-1}$, with an average of 172.15 Bq kg$^{-1}$. The hazard index of exposure to external doses was estimated at 3.3 – 32.1 nGy h$^{-1}$ with an average of 16.2 nGy h$^{-1}$. In addition, the indications of exposure rate the gamma radiation and the average annual effective dose are below the worldwide average. Thus, sediments in coastal areas will not pose a significant source of radiation hazard to residents on the beach near sampling sites.
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