Radioactivity level of coastal sediments and water across the Sangihe Island as the outlying islands of Indonesia

M N Yahya1*, Untara1, M Makmur1, W R Prihatiningsih1, D I P Putra1, Y Priasetyono1

1 Marine radioecology group, Center for Technology of Nuclear Safety & Metrology-BATAN, Jl. Lebak Bulus Raya No.49 Jakarta Selatan, Indonesia

* Contribution author, E-mail: yahya_vhs@batan.go.id

Abstract. Indonesia as an archipelagic country is very concerned about maritime development, the outlying islands are the entrance for potential threats of radioactive distribution from global waters. Furthermore, the outlying islands have functions such as territorial defence, security, and sovereignty, economic and ecological. As environmental protection, level of natural and anthropogenic radioactivity was performed due to potential threats. The activity concentration of natural (226Ra, 232Th, and 40K) radionuclides inshore sediments samples are range from 3.44 ± 0.01 Bq kg⁻¹, 5.13 ± 0.03 Bq kg⁻¹ and 151.42 ± 0.04 Bq kg⁻¹ respectively and average concentration level of anthropogenic of 137Cs in sediment and seawater obtained were 0.35 Bq kg⁻¹ and 0.83 Bq m³ respectively. The concentrations value of NORM was lower compare to another region but the ratio sequence 226Ra<232Th<40K relatively have same pattern. The presence of fission product 137Cs in this region comes from global fallout since no nuclear activities in Indonesia and the level of 137Cs are almost same as the estimation the concentration 137Cs in the Pacific Ocean and comparable with other studies. Radioactivity levels in all samples are under the value of radiological hazard index (0.004-0.039, trace hold h-index=1) and annual effective dose rate are in save level (0.01 mSv/y, recommended limit AED =1mSv/year). The level of NORM radioactivity is locally and varied for each region meanwhile anthropogenic radioactivity are global and comparable since there are no nuclear activity in Indonesia.

1. Introduction
Indonesia is very concern in the development of maritime resources. As an archipelagic country the outers island potentially to be the first area to be contaminated by radioactive from global waters. Meanwhile, the outers islands have functions such as territorial defence, security and sovereignty, economic and ecological. Furthermore, maritime is civilization, but the establishment of maritime sector depending on how the sea is managed and utilized for prosperity and sustainability.

Sangihe Island is part of the North Sulawesi province located between 2° 4’ 13” – 4° 44’ 22” N and 125° 9’ 28” – 125° 56’ 57” E has boundaries as follows Republic of Phillipines and Talaud regency at northside, Pacific Ocean and maluku sea at eastside and Sulawesi sea at westside. As a consequence of being passed by global currents, Sangihe has marine biodiversity and became a tourist destination. Moreover, based on Sangihe economic growth data, one of the biggest contributors are marine
products. As a consequence of the continuous flow and mixing of seawater, contaminants that enter in one place continuously spreading throughout the world's oceans [1]. The concentration of anthropogenic radionuclides in the Ocean began in 1945 from nuclear weapons tests, Nuclear Power Plant accidents, accidents and loss of nuclear material, and the disposal of industrial waste [1].

Sangihe region was located in a subduction zone that create volcanic-magmatic activity and it has been suggested there are orogenic gold deposits and associated minerals [2]. Baru Gold Corporation (formerly East Asia Minerals) and local company recently granted operating license within area of 42,000 ha from Indonesian government (February 2021) but is awaiting final land acquisition and the project has not yet started the construction according to company announcement on official website. Mining activities in Sangihe actually have been around traditionally by native people. Nowadays Sangihe became a mining industry has potentially threatening land then transported to the coastal and marine environment.

There are 2 types of radioactivity based on the origin of their production, natural and artificial radionuclides. Artificial radionuclides are products from thermonuclear reactions of nuclear installation both normally or accident conditions and nuclear weapon testing which fall directly or indirectly into marine ecosystems from the atmosphere. Natural radionuclides in particular have been formed since the beginning of the earth's creation. Around 80% of human radiation exposure comes from naturally occurring radioactive sources, which can have a variety of negative consequences for humans, animals, and the environment. [3]. Natural materials that emit radiation are called naturally occurring radioactive materials (NORM). Sources of natural radiation that we receive come from terrestrial and extra-terrestrial (cosmic) [4]. Terrestrial background radiations are the total emitted radiations from the NORM in the earth's crust and vary with space and time.

2. Methods

2.1. Sampling area
The research was carried out in the area on the coast of Sangihe Island in August 2018. Sediment and Seawater samples were collected from the 5 Beach location in the Northern part of Sangihe. Sampling locations were chosen random method due to no nuclear activity, however, the location was chosen considered representative of the area condition were passed by global ocean currents.

Collected samples are required to be packed properly to avoid cross contamination during transfer to the laboratory and sample identity and collection information shall accompany the samples during transfer.

![Figure 1. The study region and sampling location](image)
Table 1. Details of the sampling points of Sangihe Island

| ID Sample | Latitude          | Longitude          | Depth (m) | Location         |
|-----------|-------------------|--------------------|-----------|------------------|
| ST 1      | 03° 36’ 22,4‖ N   | 125° 28’ 40,6‖ E   | 1         | Western Tahuna   |
| ST 2      | 03° 37’ 11,4‖ N   | 125° 26’ 45,8‖ E   | 1         | Western Tahuna   |
| ST 3      | 03° 38’ 42,0‖ N   | 125° 24’ 45,7‖ E   | 1         | Western Tahuna   |
| ST 4      | 03° 43’ 59,8‖ N   | 125° 25’ 23,6‖ E   | 1         | Northern Tahuna  |
| ST 5      | 03° 43’ 29,3‖ N   | 125° 30’ 14,4‖ E   | 1         | Northern Tahuna  |

2.2. Sediment sampling
Sampling was performed on the upper beach by collect the sea sediments about 1Kg each. When determining the activity of gamma radionuclide concentrations in sediment, it does not require chemical preparation. Sediment only need to be dried and make sure the water and biota content were not there. In NORM measurements samples to be sealed for certain days before measure by gamma-ray spectrometry to prevent $^{222}$Rn (noble gas) escape from container and ensure sample reach secular equilibrium between daughter and parent activity. When samples were sealed after about 30 days it will achieve $\approx 99.56\%$ of full secular equilibrium ($^{226}$Ra, $^{222}$Rn and $^{214}$Pb, $^{214}$Bi) [5]. Sediment samples were measure by using HPGe coaxial detector GC2020 type with 20% relative efficiency and GENIE 2000 as analysis software by Canberra.

2.3. Seawater sampling
Approximately 80 L of surface seawater were collected as same as sediment sampling location. Seawater samples were taken to determine the activity concentration of radionuclide anthropogenic $^{137}$Cs. Altogether seawater samples were performed pre-concentration processes due to their very low concentration. Pre-concentration procedure was conducted according to IAEA standard procedures for individual radionuclides with minor modification. This procedure uses the solution of 10 gram of $K_4(\text{Fe(CN})_6$ and 10 gram of $\text{Cu(NO}_3)_2$ for each sample until homogenous. The slurry produced from precipitation were collected using filter paper. Furthermore, filter paper was dried and packed in plastic container following the radiometric standard. Seawater samples were measure and determine the activity concentration using window analysis and full spectrum analysis as well as sediments [6].

2.4. Radioactivity analysis
Determination of radionuclides in sediment and seawater using HPGe gamma spectrometry procedure was follow in this study. The type of detector used for sample analysis in this study is an HPGe detector made in Canberra equipped with a 15cm thick Pb shielding to prevent interference from outside radiation. The GENIE2000 software was used to operate the gamma spectrometer system by Canberra. The use of the GENIE2000 software includes energy calibration, efficiency calibration, correction, as well as displaying the spectrum and interpreting the analysis results of the spectrum. Energy and Efficiency calibration of detector HPGe performed using Soil 152Eu E&Z standard and IAEA RG Th-232 reference materials. The sediment samples were counted for 3 days to achieve more than 95% confidence level and reduce counting error.

The activity concentration and uncertainty of radionuclides was calculated following formula [7]
where $a$ is the activity concentration (Bq kg), $n_g$ is net peak area samples, $n_0$ is net peak area background, $t_g$ is time of measurements (second)

$$w = \frac{1}{m_s \epsilon \xi f_s f_p D}$$

where $w$ is conversion factor, $m_s$ is mass of the sample (kg), $\epsilon$ is the efficiency of measurement, $f_s$ correction factor as the effect of geometry, element composition, density etc between samples and standard, $f_s$ is correction factor of coincidence summing, $P$ is the gamma abundance and $D$ is decay time correction. Continue with uncertainty of activity ($u(A)$) calculation following formula.

$$u(A) = \sqrt{w^2 (n_g + u^2(n_0)) + (n_g - n_0)^2 u^2(w)}$$

3. Result and discussion

Sangihe Island is composed of volcanic rocks that erupted from at least four volcanic centres, which progressively young from south to north[8]. The radioactivity level of the sediment that has been analysed for its NORM content reflects the activities on land because the sampling location is not far from the land. Sediment play important role in the recycling process as well as serve as reservoir of various pollutants.

| Location | Coordinate | Activity (Bq kg$^{-1}$) |
|----------|------------|-------------------------|
| ST 1     | 125° 28’ 40.6” E 03° 36’ 22,4” N | 3.915 ± 0.59 | 4.54 ± 0.68 | 177.05 ± 6.56 | 0.55 ± 0.06 |
| ST 2     | 125° 26’ 45.8” E 03° 37’ 11.4” N | 4.06 ± 0.61 | 7.05 ± 1.05 | 156.33 ± 6.11 | 0.37 ± 0.07 |
| ST 3     | 125° 24’ 45.7” E 03° 38’ 42” N | 3.97 ± 0.59 | 7.82 ± 1.17 | 151.85 ± 5.94 | 0.36 ± 0.07 |
| ST 4     | 125° 25’ 23.6” E 03° 43’ 59.8” N | 3.75 ± 0.56 | 4.50 ± 0.67 | 189.28 ± 7.35 | 0.30 ± 0.07 |
| ST 5     | 125° 30’ 14.4” E 03° 43’ 29.3” N | 1.50 ± 0.23 | 1.72 ± 0.25 | 82.57 ± 3.30 | 0.17 ± 0.06 |

The NORM activity concentration of $^{226}$Ra, $^{232}$Th and $^{40}$K were 1.50 ± 0.23 - 4.06 ± 0.61 Bq kg$^{-1}$, 1.72 ± 0.25 - 7.82 ± 1.17 Bq kg$^{-1}$ and 82.57 ± 3.30 - 189.28 ± 7.35 Bq kg$^{-1}$ respectively and anthropogenic activity concentration of $^{137}$Cs were 0.17 ± 0.06 - 0.55 ± 0.06 Bq/m$^3$. Base on the table 1 overall activity concentration of NORM always follow $^{226}$Ra < $^{232}$Th < $^{40}$K. That value also found in another paper [9, 10, 11, 12] while $^{40}$K activity concentration dominates is soil all over the world.

Comparing results from the collecting sites, its clearly the ST5 location shown a value that is significantly different from other sites. This shows that the dynamic character of coastal areas is able to influence the comparable radioactivity levels in both sediment and seawater. This, most probably due to the tidal effect causing continuous leaching of sediment. Furthermore, the sampling location number 5 is the side of the Sangihe island that faces directly global waters. Geologically the oldest part of the island is in the southeast and the youngest in the northwest; there are forming new volcanoes off the north western tip and western shores of Sangihe Island [13,14].
Figure 2. Correlation between $^{232}\text{Th}$ and $^{40}\text{K}$

Figure 3. Correlation between $^{226}\text{Ra}$ and $^{232}\text{Th}$

In order to identify the correlation data of distribution NORM in sediment, the data was plot in figure 2 between $^{232}\text{Th}$ and $^{40}\text{K}$ and figure 3 between $^{226}\text{Ra}$ and $^{232}\text{Th}$. The graphic shows us $^{226}\text{Ra}$ and $^{232}\text{Th}$ have a good positive correlation (0.69) compared to $^{40}\text{K}$ (0.26) because both of radionuclide are from the same parents radionuclide $^{238}\text{U}$ meanwhile $^{40}\text{K}$ is not connected and also a parent radionuclide.

Further analysis, we can determine the external dose rate in the air at 1 m above using the following formula

$$Dose = (0.462ActRa) + (0.604 ActTh) + (0.042 ActK)$$

where dose unit in nGy h$^{-1}$ and Act unit following the activity concentration were obtained. We can use the dose rate data to estimated the annual effective dose (AED) following formula [15]

$$AED uSv y^{-1} = D (nGy h^{-1}) x 24 (hours) x 365 (days) x 0.2 x 0.7 (Sv Gy^{-1}) x 10^{-3}$$

According to the table the ratio of $^{226}\text{Ra}$ and $^{232}\text{Th}$ was 1.4 and in all location, thorium is higher than radium probably this area pegmatite rocks [17]. The radiological data of NORM in soil and sediments are compared with other author in table 2. Base on the table, the dose rate and radiation hazard index in Sangihe were below all over data from other. According to the table 2, the annual effective dose rate in Sangihe were the lowest among the other and very low under ICRP recommendation dose for public 1 mSv year$^{-1}$. NORM/TENORM data collection is required due to increasing mining activity on land and plans to make Sangihe a mining industry. Marine radioecology studies include research on radionuclide content both natural and anthropogenic so that the level of radioactivity in the area can be known.

The concentration of seawater and sediment activity was plotted using ODV and then converted into contours. The contours show the distribution of anthropogenic radionuclides following the pattern of the ITF current flowing to the south. Meanwhile, the sediment counter shows sediment in the middle of the island and south of Sangihe. So that further research is needed to conduct radioecological research in the southern part of Sangihe Island to confirm the activity concentration distribution of sediment distribution. It can be concluding the level of NORM radioactivity is locally and varied for each region meanwhile anthropogenic radioactivity is global because there is no nuclear activity in Indonesia.
Table 3. Radiological NORM data compare and assessment

| Location | $^{226}$Ra Bq kg$^{-1}$ | $^{232}$Th Bq kg$^{-1}$ | $^{40}$K Bq kg$^{-1}$ | Dose nGy/hour | Annual effective dose mSv/year | Hazard index | Reff |
|----------|--------------------------|---------------------------|-----------------------|---------------|--------------------------------|--------------|------|
| Sangihe  | 1.5 – 4.06               | 1.73 – 7.82               | 82.57 – 189.28        | 11.06         | 0.01-0.0                        | 0.004 – 0.039 | Present work |
| China    | 2 - 690                  | 1 - 360                   | 9 – 1800              | 13 – 760      | 0.01 – 0.932                    | -            | 15 |
| USA      | 4 – 140                  | 4 – 130                   | 100 – 700             | 26 – 278      | 0.03 – 0.34                     | -            | 15 |
| Shore of Nakuru,Kenya | 11.2 – 92.5 | 35.4 – 74.6 | 534.8 – 837.4 | 48.86 – 106.8 | 0.08 | 0.41 | 12 |
| Red Sea, EGYPT | 4.42 – 4.90 | 4.90 – 19.12 | 198 – 547 | 13.63 – 40.34 | 0.16 – 0.5 | 0.08 – 0.271 | 16 |
| Word average | 16 – 110 | 11 – 64 | 140 – 850 | 38 – 293 | 0.04 – 0.36 | - | 15 |

**Figure 4.** Isosurface contour $^{137}$Cs in Seawater using Ocean Data View

**Figure 5.** Isosurface contour $^{137}$Cs in Sediment using Ocean Data View (Bq kg$^{-1}$)

In addition, the concentration of $^{137}$Cs is also used as a tracer to predict the transfer of global water mass [18,19] because it is easily soluble in water so that when mixed with local seawater masses make it dispersed into various water areas [20]. Indonesia has the Indonesian Through Flow (ITF) bring water mass system of water that crosses Indonesian local waters that connects the Pacific Ocean to the Indian Ocean. The ARLINDO phenomenon has become one of the characteristics of the current system in Indonesia. Circulation of seawater pass through Indonesian waters originating from the Pacific Ocean to the Indian Ocean due to the difference in density [21].
4. Conclusion

The level of radioactivity from 5 stations in Northern Sangihe have been determined by Spectrometry Gamma. The average concentration level of NORM in this study were lower compared to another region. The average activity concentration of $^{226}$Ra, $^{232}$Th and $^{40}$K results were $3.44 \pm 0.01$ Bq kg$^{-1}$, $5.13 \pm 0.03$ Bq kg$^{-1}$ and $151.42 \pm 0.04$ Bq kg$^{-1}$ respectively. The data that has been taken can be compared when the mining industry is running by seeing whether the NORM level is increasing or not. The average concentration level of anthropogenic of $^{137}$Cs in sediment and seawater obtained were $0.35$ Bq kg$^{-1}$ and $0.83$ Bq m$^{-3}$ respectively. It can be concluding the level of NORM radioactivity is locally and varied for each region meanwhile anthropogenic radioactivity is global because there is no nuclear activity in Indonesia. On other hand the anthropogenic radionuclide $^{137}$Cs level comparable and will be stable as carried from the pacific or declined very slowly until Twenty-thirty years after, levels of $^{137}$Cs in the Pacific and Indian Ocean are expected to be below 0.3 and 0.1 Bq/m$^3$ [22].

References

[1] Hong G H and Povince P 2021 Environmental Protection: The Oceans—Implications of Manmade Radiation Encyclopedia of Nuclear Energy, no. -, pp. 505 – 519.

[2] Di Leo J F, Wooley J, Hammond J O S, Kendall J M, Kaneshima S, Inoue H, Yamashina T, Harjadi P 2012 Deformation and mantle flow beneath the Sangihe subduction zone from seismic anisotropy Physics of the Earth and Planetary Interiors, vol. 194-195 pp. 38-54.

[3] Ali-Mohsen M, Zhao H, Li-Zhongyou and Maglas N N M 2019 Concentrations of TENORMs in the petroleum industry and their environmental and health effects Royal Society of Chemistry vol.-, No.9,pp.39201-39229.

[4] IAEA 2005 World Marine Radioactivity Studies (WOMARS). IAEA TECDOC-1429 pp.187

[5] Bonczyk M and Samolej K 2019 Testing of the radon tightness of beakers and different types of sealing used in gamma-ray spectrometry for $^{226}$Ra concentration determination in NORM Journal of Environmental Radioactivity vol. 205-206 pp.55-60.

[6] Yahya M N, Makmur M, Putra D I P 2016 Determination of Window Analysis and Full Spectrum Analysis Method of Gamma International Conference on the Sources, Effects and Risks of Ionizing Radiation (SERIR2) Vol 2 No.- pp.162-170.

[7] ISO 11929-1 2019 Determination of the characteristic limits (decision threshold, detection limit and limits of the coverage interval) for measurements of ionizing radiation — Fundamentals and application

[8] MORRICE M G, JEZEK P A, GILL J B, WHITFORD D J and MONOARFA M 1983 AN INTRODUCTION TO THE SANGIHE ARC: VOLCANISM ACCOMPANYING ARC--ARC COLLISION IN THE MOLUCCA SEA, INDONESIA Journal of Volcanology and Geothermal Research Vol 19 pp.135-165.

[9] Suresh G, Ramasamy V, Meenakshisundaram V, Venkatachalapathy R, Ponnusamy V 2011 A relationship between the natural radioactivity and mineralogical composition of the Ponnaiyar river sediments, India, vol 102, no.4 pp. 370–377.

[10] Srilatha M C and Rangaswamy D R 2014 Measurement of natural radioactivity and radiation hazard assessment in the soil samples of Ramanagara and Tumkur districts, Karnataka, India Journal of Radioanalytical and Nuclear Chemistry vol.302 pp.-.

[11] Yashodhara I, Karunakara N, Kumar K S, Murthy R, Tripathi R M 2011 Radiation levels and radionuclide distributions in soils of the gogi region, a proposed uranium mining region in north Karnataka Radiation Protection Environment vol.34, No. 4,pp.267-269.

[12] Langat W K, Omar N H, J ambusso W 2014 Gamma Ray Spectrometric Analysis of Sedimental Deposits at the Shores of Lake Nakuru, Kenya Journal of Natural Sciences Research vol.4, No21. 1,pp.1-66.

[13] King Julia, Jones-william A E, Hinsberg V, Glyn W J 2014 High-Sulfidation Epithermal Pyrite-Hosted Au (Ag-Cu) Ore Formation by Condensed Magmatic Vapors on Sangihe Island, Indonesia Society of Economic Geologists vol.109 pp 1705-1733.
[14] Beaulieu S 2010 InterRidge global database of active submarine hydrothermal vent fields: prepared for InterRidge World Wide Web electronic publication vol.2.0
[15] UNSCEAR 2000 Sources and effects of ionizing radiation. United Nations Scientific Committee on the Effects of Atomic Radiation Report.
[16] Arafat A A, Salama M H M, El-Sayed S A, Elfeel A A 2017 Distribution of natural radionuclides and assessment of the associated hazards in the environment of Marsa Alam-Shalateen area, Red Sea coast, Egypt Journal of Radiation Research and Applied Sciences vol.10 pp-.
[17] Megumi K, Oka T, Doi M, Tsujimoto T, Ishiyama T, Katsurayama K 1998 High-Sulfidation Epithermal Pyrite-Hosted Au (Ag-Cu) Relationship between the concentrations of natural radionuclides and mineral composition of the surface soil Radiation Protection Dpsimetry vol.24 pp 69-72.
[18] Aoyama M, Fukasawa M, Hirose K, Hamajima Y, Kawano T, Povinec P P, Sanchez-Cabeza J A 2011 Cross equator transport of 137Cs from North Pacific Ocean to South Pacific Ocean (BEAGLE2003 cruises) Progress in Oceanography vol.89 pp 7-16.
[19] Tsumune D, Aoyama M, Hirose K, Bryan F O, Lindsay K, Danabasoglu G 2011 Transport of 137Cs to the Southern Hemisphere in an ocean general circulation model Progress in Oceanography vol.89 pp 38-48.
[20] Kim Y, Kim K, Kang H D, Kim W, Doh H, Kim D-S, Kim B-K 2007 “The accumulation of Radiocesium in coarse marine sediment:effects of mineralogy and organik matter.” Marine Pollution Bulletin vol.54 pp 1341-1350.

Acknowledgments
Authors wishing to acknowledge the funding research by DIPA PTKMR – BATAN conducting by marine radioecology group. The main contributor of this research is Mohamad Nur Yahya.