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CONCENTRATION OF NATURAL RADIONUCLIDE AND POTENTIAL RADIOLOGICAL DOSE OF ^{226}RA TO MARINE ORGANISM IN TANJUNG AWAR-AWAR, TUBAN COAL-FIRED POWER PLANT

Chairun Annisa Aryanti[1)], Heny Suseno[3)], Muslim[2)], Wahyu Retno Prihatiningsih[3)], & Mohamad Nur Yahya[3)]

[1)]Department of Marine Science, Faculty of Fisheries and Marine Science, Diponegoro University
Jl. Prof. H. Soedharto, SH, Tembalang, Semarang, 50275 Indonesia
[2)]Department of Oceanography, Faculty of Fisheries and Marine Science, Diponegoro University
Jl. Prof. H. Soedharto, SH, Tembalang, Semarang, 50275 Indonesia
[3)]Marine Radioecology Group, Center for Radiation Safety Technology and Metrology, National Nuclear Energy Agency
Jl. Lebak Bulus Raya No. 49, Kotak Pos 7043 JKSKL, Jakarta Selatan, 12070 Indonesia

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ABSTRACT

Fly ash and bottom ash from coal combustion can contain natural radionuclides with a certain concentration. The release of fly ash into the environment can potentially increase the concentration of natural radionuclides in the surrounding environment, including marine waters. This study aimed to determine the activity of natural radionuclides in seabed sediments and the radiological impact on marine organisms related to Tanjung Awar-Awar Coal-Fired Power Plant activities in Tuban. The sampling techniques were carried out by the purposive sampling method. The concentration measurement of natural radionuclide activity was carried out using HPGe detector gamma-ray spectrometry at marine radioecology laboratory of National Nuclear Energy Agency. The detected concentration of natural radionuclide activity was ^{40}K ranging from 159.18 to 365.88 Bq.Kg^{-1}, ^{228}Ac ranging from 14.47 to 28.05 Bq.Kg^{-1}, and ^{226}Ra ranging from 15.00 to 67.65 Bq.Kg^{-1}. By using the ERICA Assessment Tool Tier 2 program, the results of the analysis showed that there was no impact of ^{226}Ra radiation on marine organisms at the research site because the total radiation dose rate (external + internal) of ^{226}Ra on marine biota was lower than the screening level of 10 µGy / h. Thus, it will not harm marine ecosystems and the sustainability of marine organisms in the waters of Tanjung Awar-Awar Coal-Fired Power Plant, Tuban.

Keywords: Natural radionuclides, ERICA Tool, sea bed sediment, coal-fired power plant, Tuban waters.

Corresponding author:
Jl. Pasir Putih I Ancol Timur, Jakarta Utara 14430. Email: chairunannisa10@gmail.com

INTRODUCTION

Bali I Coal is one of the most important energy sources in the world (IAEA, 2010). In Indonesia, the increase in coal production is expected to continue to meet the needs of power plants, industrial activities, and overseas demand (exports). One of the factors that cause high coal production in Indonesia is coal-fired power plants (CFPP) (Sekretaris Jenderal Dewan Energi Nasional, 2019).

CFPP Tanjung Awar – Awar, Tuban, East Java, is one of the coal-fired power plants included in 10,000 MW Energy Diversification Acceleration Program (PPDE) Phase 1 with the capacity of 2 x 350 MW (Riadessy, 2015). The fuel used is low-calorie coal originating from Bontang, Kalimantan, with coal consumptions of 160 tons/hour to produce 700 MW of electrical energy (Riadessy, 2015). Coal contains several natural radionuclides or Naturally Occurring Radioactive Materials (NORM) (Ozden *et al*., 2018). In the coal combustion process in the CFPP, there will be cracking that causes NORM to come out along with other emission gases through bottom ash and fly ash which has NORM content ten times higher than the original value (Alam & Mohamed, 2011). Fly ash has the potential to release into the environment because a filtration system on CFPP chimneys such as electrostatic precipitators, baghouses, and scrubbers, if functioning properly, only reduces the emission of fly ash into the atmosphere by about 95% (Dinis *et al*., 2013). Thus, fly ash will fall into the environment around the power plant, which is usually dominated by marine waters (Alviandini *et al*., 2019) because the air has a lower density than fly ash (Amin *et al*.,2013). The coal processing process causes NORM to be concentrated and forms a radioactive concentrated called TENORM (Alam & Mohamed, 2011). TENORM is Technologically Enhanced Naturally Occurring Material defined as a radioactive element that is concentrated or exposed to the environment as a result of human activities one of which is mining (Uddin & Behbehani, 2018). Technologically Enhanced means that the properties radiological, physical, and chemical of radioactive materials have been concentrated or have been changed by a process, that increases the potential for radiation exposure to humans and the environment (Makmur *et al*., 2019). Some of the dominant elements contained in fly ash are ^{210}Pb, ^{210}Po, ^{231}Pa, ^{226}Ra, ^{232}Th, ^{228}Ac, ^{238}U, dan ^{40}K (Pandit *et al*., 2011).

NORM is classified as a dangerous element and can harm the survival of living things (Alviandini *et al*., 2019). Natural radionuclides such as ^{226}Ra, have a higher risk of long-term exposure prolonged period, resulting in harmful effects including anemia, cataracts, cancer, and death. Some of these effects may take years to develop and are mostly caused by gamma radiation emitted by radium (Peterson *et al*., 2007).

Natural radionuclides released into the marine environment will generally be spread through abiotic components (water and sediment). By these components, there is also accumulation into biota tissue, this incident can disrupt the life of biota and humans who consume these marine biotas (Suseno & Prihatiningsih, 2014). The effect of the NORM's existence of coal-fired power plant operations in the environment, especially the marine environment, can be a problem for coastal natural resources so that it needs to be observed further (Liu *et al*., 2015).

Many studies related to the presence of radionuclides in Indonesian waters have been widely conducted, but the studies only focus on the distribution of anthropogenic radionuclides ^{137}Cs. Studies on the distribution of natural radionuclides (^{232}Th, ^{226}Ra, ^{40}K) have been conducted in Tikus Island, Bengkulu (Syaher *et al*., 2015), Bengkalis Island (Makmur *et al*., 2019), and Bangka Island (Prihatiningsih & Suseno, 2012). In other countries, studies on the increase in NORM activity due to coal-fired power plant operations have been carried out in Poland (Bem *et al*., 2002), In Brazil (Flues *et al*., 2002), in Spain (Charro & Peña, 2013), in China (Liu *et al*., 2015), and India (Pandit *et al*., 2011). Similar research in Indonesia is still very limited. NORM research related to coal operations has been carried out at CFPP of Labuan (Anggarini *et al*., 2018), CFPP of Rembang (Marwoto *et al*., 2019), and CFPP of Jepara (Alviandini *et al*., 2019) but only limited to environmental monitoring. Meanwhile, NORM research at CFPP Tuban, East Java, has never been carried out. Thus, this research needs to be carried out to review NORM activities on the environment and marine resources due to the operation of CFPP Tanjung Awar - Awar, Tuban.

A further study of radionuclide monitoring in the marine environment is to examine the impact on marine waters, including marine biota in the research area (Suseno & Prihatiningsih, 2014; Prihatiningsih *et al*., 2016). The radiological effects of exposure to radioactive substances above a certain threshold can be potentially life-threatening, so they cannot be ignored (Makmur *et al*., 2019). The presence of certain amounts of radioactive elements in the marine environment will indicate whether or not a precaution is taken (Makmur *et al*., 2019). Now, several model sets have been developed to facilitate the assessment of the radiological impact of ionizing radiation on various species of biota, one of the most widely used models is the ERICA Assessment Tool (Brown *et al*., 2004; Brown *et al*., 2008; Gjelsvik *et al*., 2012; ERICA, 2021). The use of the ERICA Assessment Tool has been very widely applied to various environmental conditions

(Brown *et al*., 2004; Feroz Khan *et al*., 2014). Based on this study, comprehensive results are expected related to the distribution of natural radionuclides and their impact on marine water resources.

The study aims to provide basic information on natural radioactivity in the marine environment and assessment of the radiological impact on marine organisms from activities coal processing in the coastal area coal-fired power plant Tanjung Awar-Awar, Tuban.

The presence of radioactive elements in large quantities certain in the sediments and marine organisms of the study area will indicate whether or not precautions are taken (Makmur *et al*., 2019).

Results research is expected to be used as one of the reference data for the presence of natural radionuclide to formulate the policies related to ecosystem sustainability and environmental safety.

METHODOLOGY

Method of Determining Location

The method used in determining the location of sediment sampling stations was the purposive sampling method, which is a sampling method that represents the state of the overall sampling location (Priyono, 2016). There were ten sampling points. Station 1 represented the waters of the northern coastal power plant, station 2 represented the nearest point of the CFPP outlet, station 3 represented the coastal waters of the southeastern part of the power plant, and station 4 represented the nearest point of coal loading and unloading ships. Stations 5, 6, 7 represented mid-waters, and stations 8, 9, 10 represented off shore waters (Figure 1). Global Positioning System (GPS) was used to determine the coordinates of the research station. Meanwhile, ArcGis software 10. 6. 1. was used to make the map of the research site.

Sampling Methods

Referring to (Uddin & Behbehani, 2018), sediment sampling at each station uses the A Van Veen grab sampler. Sediment samples were placed in airtight Ziplock plastic and labeled. Sediment samples were prepared for measurement of NORM activity using gamma-ray spectrometry according to the methodology established by the IAEA (International Atomic Energy Agency, 2003).

NORM Analysis Methods

The method used in determining NORM activity in sediments is by using gamma-ray spectrometry guided by the IAEA (International Atomic Energy Agency, 2003). Sediment samples were dried in the sun for approximately seven days and then dried in an oven at 80^0C for 8 hours to remove the seawater content. The dried sediment samples were crushed using a grinder with a size of 50 microns. Samples that have been finely taken as much as 1 kg were placed into Marinelli to be allowed to stand for 28 days until it reached the Seqular Equilibrium. Furthermore, measurements were carried out using *Gama High Purity Germanium* HPGe with a relative efficiency of 20 – 25% for three days to detect gamma radiation emitted from the sample. Sample measurements were carried out for three days, calculated using the Genie 2000 *Gamma Acquisition* and analysis application to calculate the gamma energy spectrum, producing a graph of energy

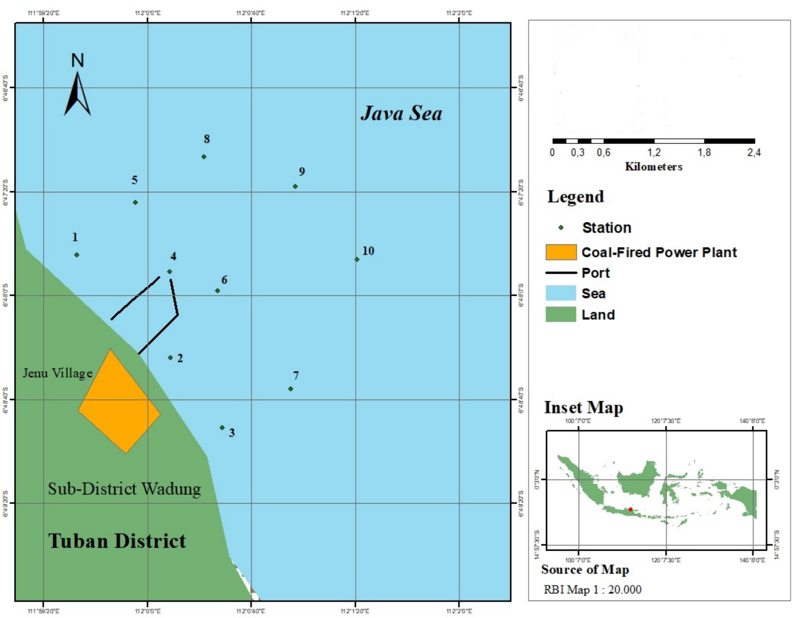

Figure 1. Research Location Map.

peaks that represented the radioactive elements contained in the sample.

Based on (Uddin & Behbehani, 2018), the activity of ^{226}Ra can be identified at low energy of 186.2 keV. In addition, the activity of ^{226}Ra can be calculated by the decay of ^{214}Pb and ^{214}Bi, at an energy of 295.2; 351.9 keV (^{214}Pb) and 609.3, 768.3, 934.1, 1120.3, 1238.1, 1764.5 keV (^{214}Bi), after 28 days, when equilibrium was reached. The activity of ^{40}K can be seen at an energy of 1460.8 keV, and activity of ^{228}Ac can be seen at an energy of 338.32 keV.

Processing of Spatial Distribution Map of Natural Radionuclides

Referring to (Sandu *et al*., 2020) to obtain a map of the spatial distribution pattern of NORM concentrations in sediments, the data analysis results in the form of radionuclide concentrations of ^{40}K, ^{228}Ac, and ^{226}Ra in sediment samples is processed by the Inverse Distance Weighted (IDW) interpolation method using ArcGIS 10.6.1. Software.

Radiological Doses Estimation in Marine Organisms

The radiological dose of ^{226}Ra in marine organisms was estimated using ERICA Assessment Tool Tier 2 software. The objective of tier 2 was to determine the possible impact of radiological hazards on non-human biota (Brown, 2008). Therefore, it was necessary to calculate the dose level of radiation exposure to marine organisms. Based on the research, the natural radionuclide ^{226}Ra was found in the Tanjung Awar-Awar waters, Tuban. The input data entered in the ERICA Assessment Tool software was the activity of ^{226}Ra in the sediment. The screening confidence level for radiation doses was set at 10 μGy/hr.

RESULTS AND DISCUSSION

Natural Radionuclide Activities (^{40}K, ^{228}Ac, and ^{226}Ra)

Based on this research, element ^{40}K had an activity value range of 113,07 to 365.88 Bq.Kg^{-1} with an average activity of 288.75 Bq.Kg^{-1}. Element 228Ac ranged from 14,47 to 28.05 Bq.Kg^{-1} with an average activity of 22.12 Bq.Kg^{-1}. Element ^{226}Ra ranged from 15.00 to 67.65 Bq.Kg^{-1}. With an average activity of 37.66 Bq.Kg^{-1} (Figure 2). The highest activity to the lowest is elements ^{40}K, ^{226}Ra, and ^{228}Ac, respectively. This is thought to be influenced by the nature of each element of the radionuclide itself. The concentration of ^{282}Ac activity in sediments tends to be low due to the influence of low geochemical mobility (Alviandini *et al*., 2019). The high value of ^{40}K element activity can occur because of ^{40}K activity in nature originating from the earth's crust that occurred since the beginning of the formation of the earth, that its content will always increase (Peterson *et al*., 2007). This is also revealed by research conducted by (Ravisankar *et al*., 2015) stated that the abundance of elements ^{40}K in nature is caused by its abundant content in rocks, moreover, ^{40}K has high solubility and geochemical mobility when compared to elements ^{226}Ra and ^{228}Ac.

Internationally, research on the concentration of radionuclides NORM and TENORM has also received high attention. Research conducted on the coast of Saudi Arabia reported that the radioactivity levels of ^{238}U, ^{232}Th, and ^{40}K which were analyzed using inductively coupled plasma - mass spectrometry (ICP-MS) showed that the activity value of element ^{40}K had a high value when compared to other natural radionuclides (Al-Trabulsy *et al*., 2013). This shows the need for great attention in managing the surrounding environment, it caused the high of 40K activity detected is related to industrial or mining activities involving raw

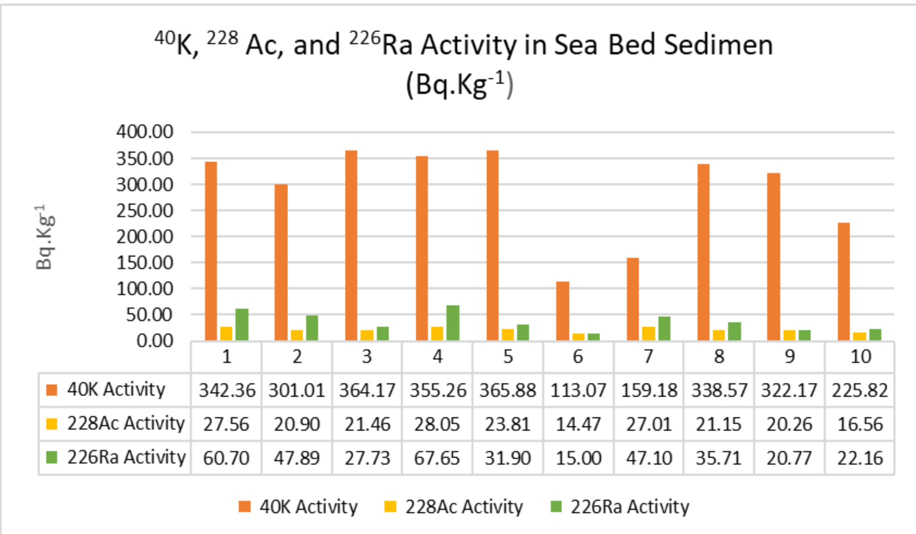

Figure 2. Graph of NORM activity in sediments in the waters of CFPP Tanjung Awar-Awar, Tuban.

materials from the earth's crust. The waste materials from industry and mining will enter water bodies and eventually form offshore sediments with weak currents (Makmur *et al*., 2019).

The natural radionuclide activity in sediments of the waters of CFPP Tanjung Awar-Awar, Tuban is still below the value of natural radionuclide activity in various worlds such as in China (Liu *et al*., 2015), in Spain (Charro *et al*., 2013), in Greece (Papaefthymiou *et al*., 2007), in India (Mishra, 2004), and in Malaysia (Amin *et al*., 2013).

In Indonesia, the activity value obtained in this study is still classified as safe when viewed from BAPETEN regulations (BAPETEN, 2013), which states that the threshold value for the derivative ^{232}Th is 228Ac of 10,000 Bq.Kg^{-1}, for the derivative ^{226}Ra is 300 Bq.Kg^{-1}, and the element ^{40}K is 3,000 Bq.Kg^{-1}.

Spatial Distribution of Natural Radionuclide (^{40}K, ^{228}Ac, and ^{226}Ra)

The spatial distribution of radionuclides in sediments can be influenced by several factors: climatological conditions (wind), type of sediment grain size, and human activities (Liu *et al*., 2015). Processing of spatial distribution maps of natural radionuclides ^{40}K, ^{228}Ac, and ^{226}Ra on seabed sediments in the waters of CFPP Tanjung Awar-Awar, Tuban, was carried out by interpolating the concentration data measured by the IDW interpolation method using ArcGIS 10.6.1 software (Figure 3, 4, and 5).

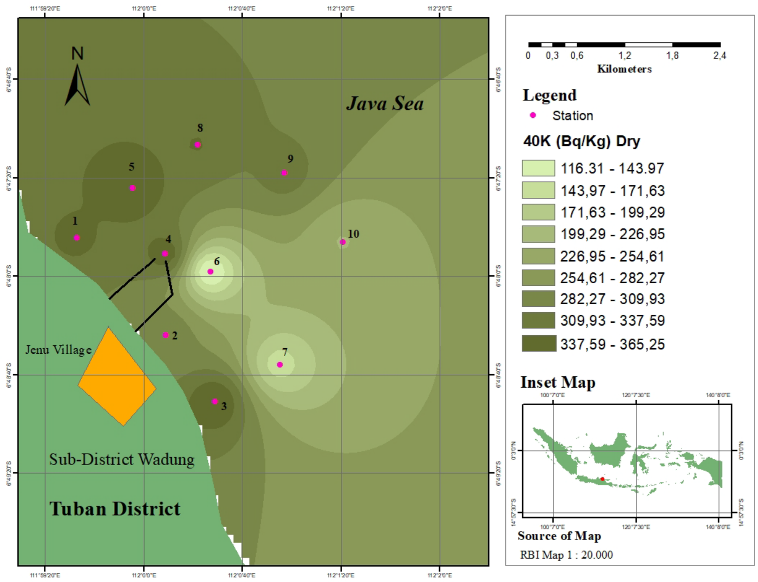

Figure 3. Spatial variation in ^{40}K concentration in sediments.

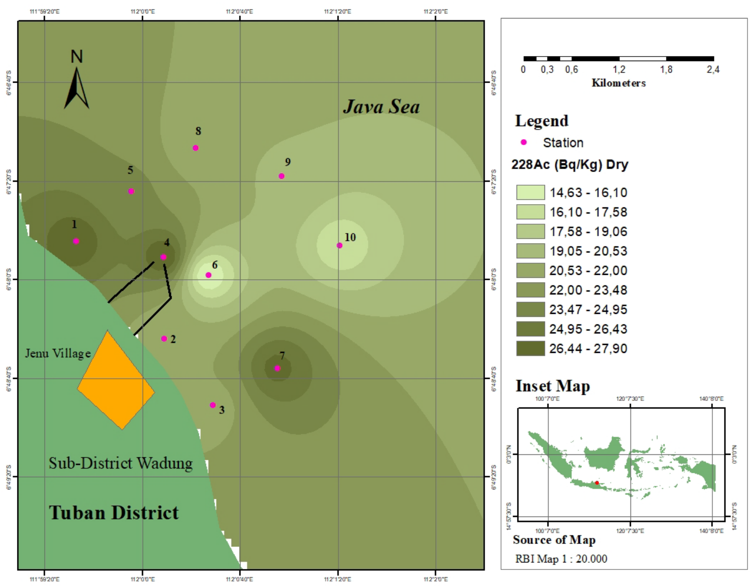

Figure 4. Spatial variation in ^{228}Ac concentration in sediments.

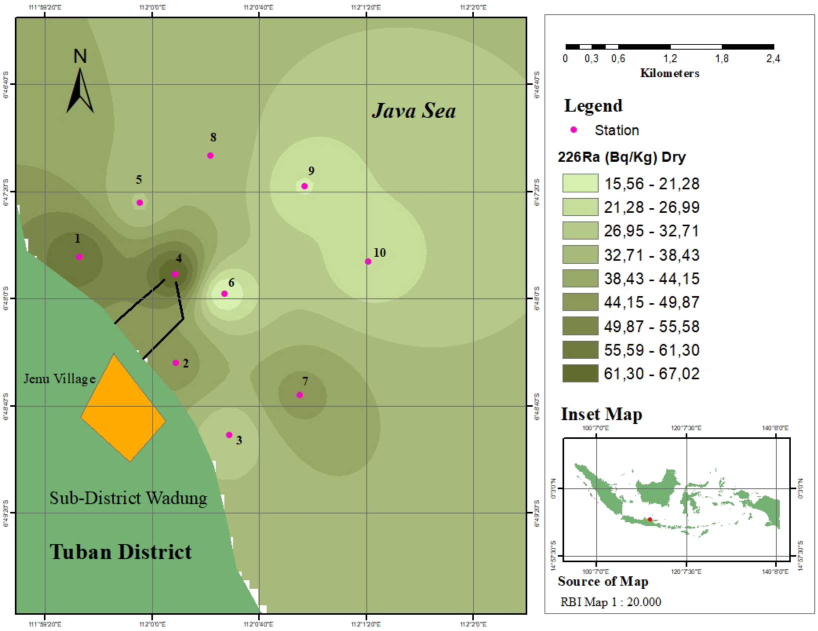

Figure 5. Spatial variation in 226Ra concentration in sediments.

The analysis of the spatial distribution map of the radionuclides ^{40}K, ^{228}Ac, and ^{226}Ra shows that station 4 had a higher activity value compared to other stations (Figures 3, 4, and 5). This can be caused in as much as station 4 is the closest point of loading and unloading coal from barges to the CFPP docks, where this activity can cause coal contamination to the marine environment. Under the statement (Ozden *et al*., 2018), coal contains several natural radionuclides or Naturally Occurring Radioactive Material (NORM), so it can increase the activity of natural radionuclides in these waters. In addition, these results follow research conducted in China by (Liu *et al*., 2015), which showed high activity of natural radionuclide in the area closest to the source of coal processing.

Station 6 is also closer to the loading and unloading site than other stations except station 4 but is consistently has the lowest value of ^{40}K, ^{228}Ac, and ^{226}Ra radionuclides. It is suspected that station 6 is influenced by ship activity (at station 6 is the path of ships sailing to loading and unloading coal) which is causing the sea bed condition of the waters to be stirred, then resulting in resuspension. Following the state of Syahrer *et al*. (2015), that resuspension causes NORM bound in sea bed sediment to be released and

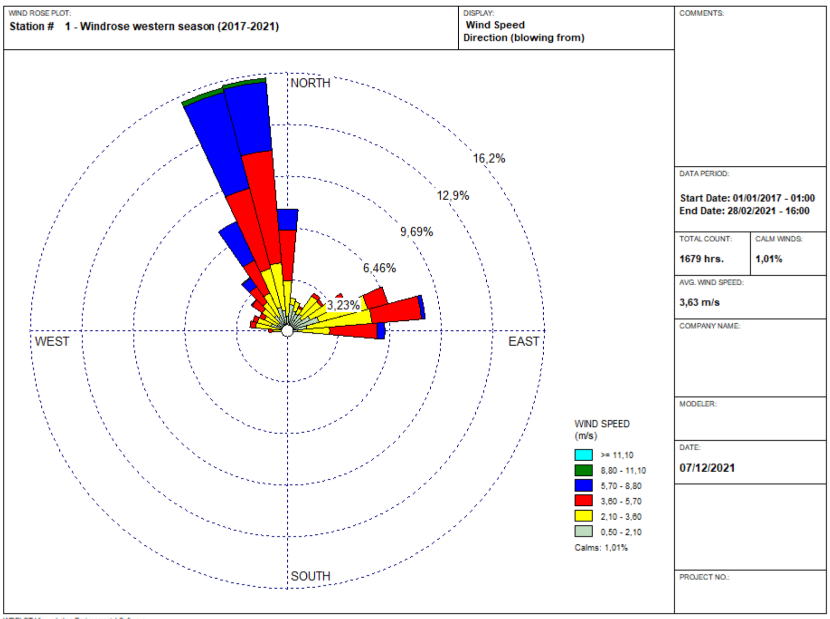

Figure 6. Windrose western season (2017-2021).

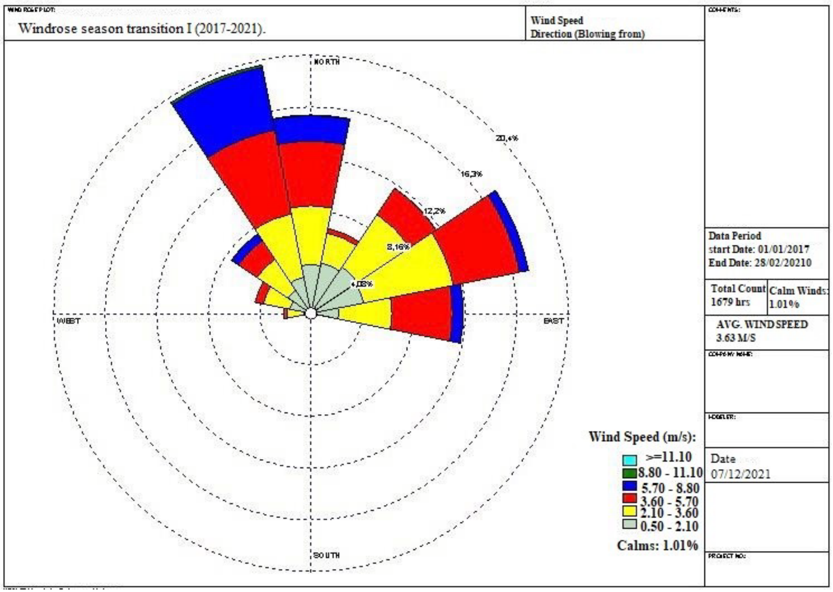

Figure 7. Windrose season transition I (2017-2021).

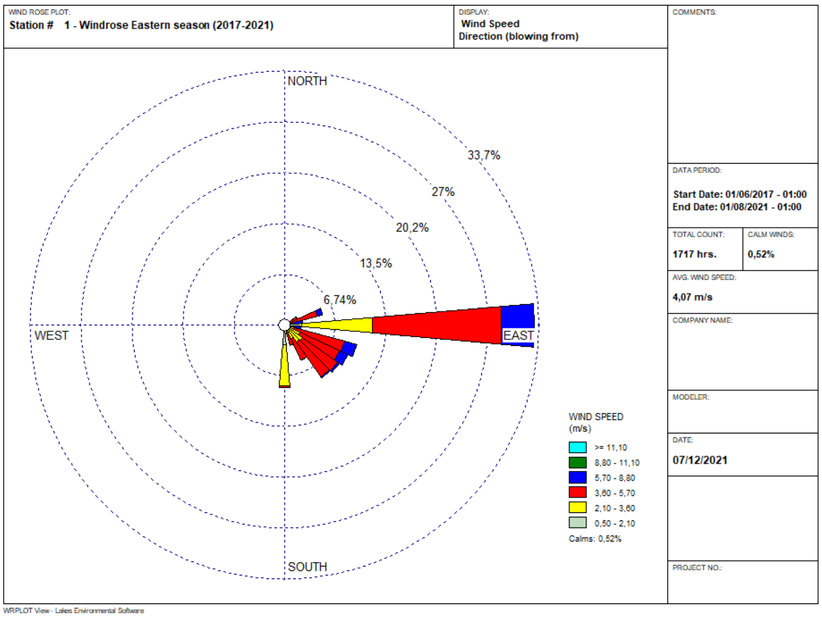

Figure 8. Windrose Eastern season (2017-2021).

rises to the water column above it then re-deposited in steady waters.

Based on the map of the spatial distribution of natural radionuclides (Figures 3, 4, and 5), the distribution of concentrations to those three elements will be more significant when heading to the northwest and west. This high value can occur due to the influence of wind movements for five years (Figures 6, 7, 8, and 9). The dominant direction is towards the northwest. Therefore, fly ash coming out of the CFPP chimney will be distributed at the location point northwest of the chimney. Then when it falls into the waters, it will be distributed by the movement of ocean currents and deposited into seabed sediments. According to (Liu *et al*., 2015), climatological conditions can affect the spatial distribution of radionuclides because fly ash in CFPP has the potential to be released into the environment through wind movement (Dinis *et al*., 2013). In addition, these results follow the research conducted by (Alviandini *et al*., 2019), which states that dominant wind movements for five years can affect the high distribution of natural radionuclide activity in waters.

Distribution of ^{228}Ac and ^{226}Ra showing higher value in the southeast (station 7) and near the coastline. It is suspected that essentially ^{226}Ra and ^{228}Ac exists naturally in soil, rocks, surface water, groundwater is generally low concentrations, in contrast with ^{40}K

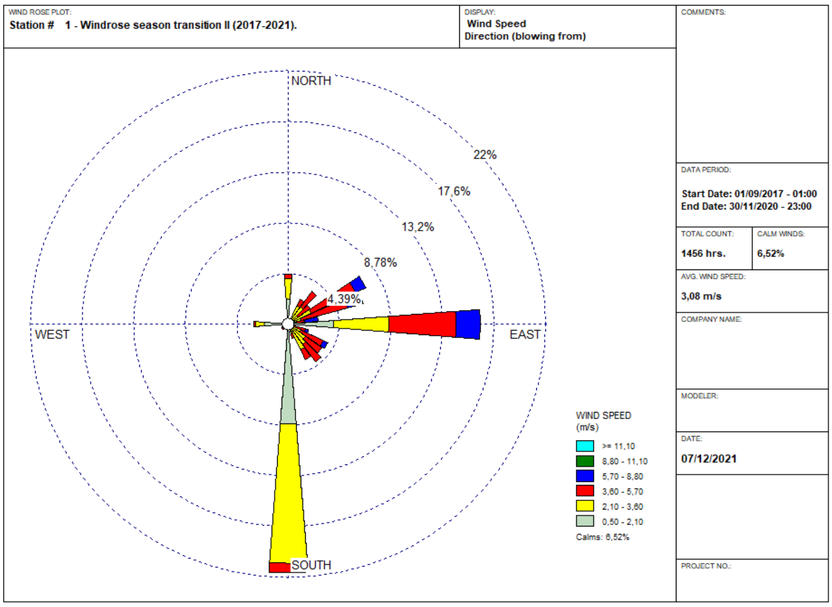

Figure 9. Windrose season transition II (2017-2021).

have been existed naturally in the earth with high concentration (Peterson *et al*., 2007). Thus, the further away from the source (coal-fired power plant), the lower concentration of the radionuclide (Liu *et al*., 2015). That causes higher concentration activity of ^{226}Ra and ^{228}Ac in the near of coastline, but the results of ^{226}Ra and ^{228}Ac activity tend to be higher in the west of the coal-fired power plant (station 1 and 4).

Station 3 has a lower activity concentration of ^{226}Ra and ^{228}Ac than station 7. It is suspected that station 3 is far from the coal-fired power plant and is located on the coastal area. Coastal areas have greater human activity than mid-waters such as at. The presence of human activity is caused the sea bed condition of the waters to be stirred, then resulting in resuspension. Following the state of Syahrer *et al*. (2015), resuspension causes NORM bound in sea bed sediment to be released and rises to the water column above it then re-deposited in steady waters such as station 7 as a mid-water. This causes station 7 to have a higher concentration than station 3.

NORM activity in sediments in the waters of CFPP Tanjung Awar-Awar, Tuban tended not to be affected by currents because of its relatively low speed. Based on the current model results processing using Delft 3D software (Figures 10 and 11), the average current

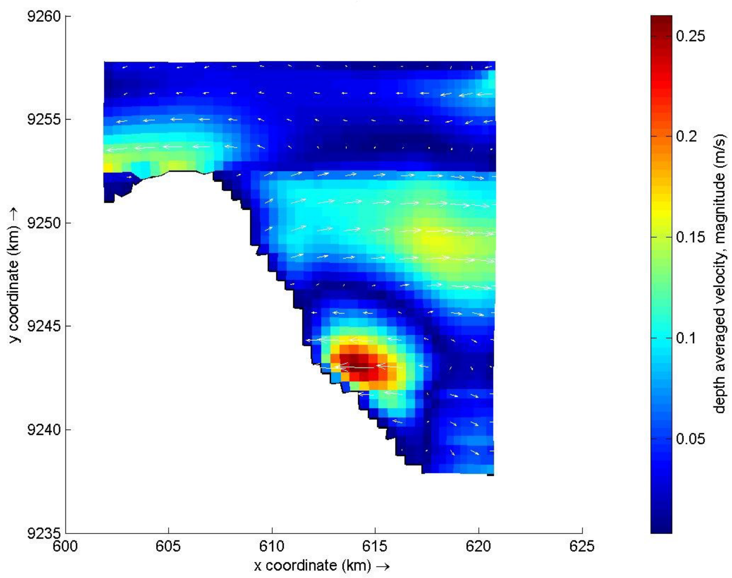

Figure 10. Current pattern at high tide in the waters of CFPP Tanjung Awar-Awar, Tuban.

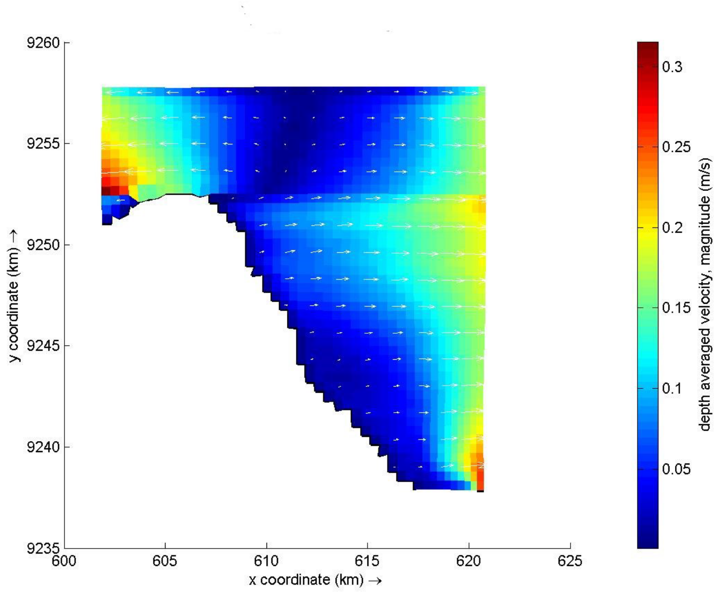

Figure 11. Current pattern at low tide in the waters of CFPP Tanjung Awar – Awar, Tuban.

velocity in the waters of CFPP Tanjung Awar-Awar, Tuban at high tide is 0.22269 m/s. This coincides with (Triatmodjo, 2007), which stated that a current speed of less than 0.5 m/s will not affect sediment transport because it cannot move seabed sediment.

Radiological Dose Estimation on Marine Organisms

The estimated dose absorbed by marine biota can be determined using the Erica Assessment Tool method, a software system for assessing the impact of radiology on biota (Brown *et al*., 2008). The system will calculate the dose rate received by biota from external and internal exposure to radionuclides, either natural or artificial (Brown *et al*., 2008).

Based on the results of laboratory tests for the activity of ^{226}Ra in sediments, the process of transferring contaminants in the food chain used transfer factor data contained in the ERICA Assessment tool database (Brown *et al*., 2008; ERICA, 2021). The database comes from experimental results in various countries and has been published in reputable international journals. The movement of contaminants in the food chain is illustrated in Figure 12. In Figure 12, if the radionuclide concentration in one component is known, the radiation dose can be predicted in the other components (ERICA, 2021).

The radiation dose level of marine biota was determined using the Erica Assessment Tool software. The study of radiation dose (external + internal) on marine organisms in CFPP Tanjung Awar-Awar Tuban waters is shown in Table 1.

Based on calculations using the Erica Tool software, the total radiation dose (external + internal) on marine organisms in CFPP Tanjung Awar-Awar waters, Tuban had a range between 0.0372 - 0.942 Gy/

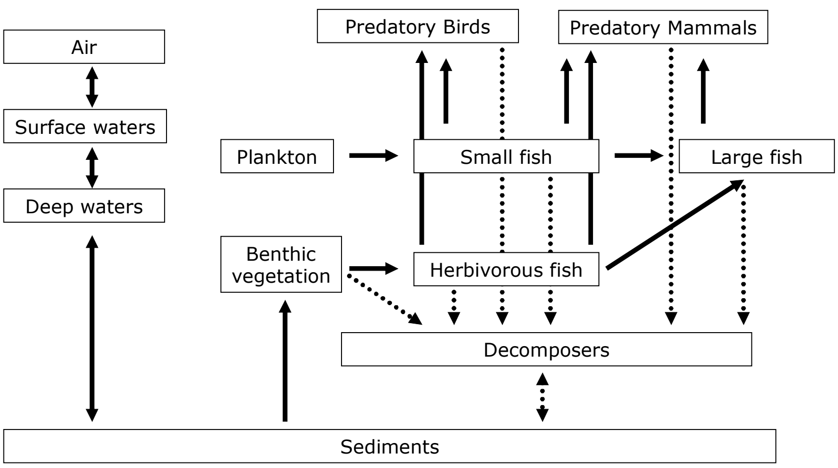

Figure 12. Current pattern at low tide in the waters of CFPP Tanjung Awar-Awar, Tuban.

Table 1. The estimated value of the total radiation dose rate of ^{226}Ra on marine biota in the waters of CFPP Tanjung Awar-Awar, Tuban

Marine Organisms	^{226}Ra total radiation dose (µGy/hr)	Screening Value (µGy/hr)
Benthic Fish	0.163	10
Bird	0.166	10
Crustacean	0.115	10
Mammal	0.167	10
Macroalgae	0.186	10
Mollusc-Bivalve	0.079	10
Pelagic Fish	0.146	10
Polychaeta worm	0.531	10
Phytoplankton	0.942	10
Reptile	0.167	10
Sea Anemon & True Coral	0.133	10
Vascular plant	0.184	10
zooplankton	0.037	10

hr. Gray (Gy) is an international unit used to express the amount of radiation dose received by a material (Brown *et al*., 2008). Using the ERICA Tool Tier 2 program, the analysis results showed that there was no impact of ^{226}Ra radiation on marine organisms at the study site. The total radiation dose rate (external + internal) of ^{226}Ra received by marine biota was lower than that of the screening level of 10 Gy/h. Thus, it will not harm marine ecosystems and the sustainability of marine organisms in CFPP Tanjung Awar-Awar waters, Tuban.

The extinction of a marine organism will result in an imbalance in the marine ecosystem due to pollution in the sea (Islam & Tanaka, 2004). One of the pollutants that can be released into sea waters is ^{210}Po natural radionuclide which can be produced from coal combustion (Alam & Mohamed, 2011). Natural radionuclides released into marine waters will generally be spread through abiotic components (water and sediment). Through these components, there is also accumulation into biota tissue so that this incident can slowly disrupt the life of biota and human that consumes marine biota (Suseno & Prihatiningsih, 2014).

Another study using the Erica Assessment Tool software was also carried out on the coast of Fukushima Beach, Japan, in June - July 2011, a few months after the nuclear reactor accident in Fukushima, Japan. The obtained dose level for marine organisms in the sampling area was lower than the screening dose level of 10 Gy/h, indicating that the radioactive release from the Fukushima nuclear accident did not significantly negatively affect marine life at the population level. However, the risk could increase through the food chain of marine biota that lives in coastal areas of Japan (Yu *et al*., 2015).

CONCLUSION

Based on this study it can be concluded that despite the region containing coal processing from the coal-fired power plant, natural radionuclide parameters in sediments on waters of CFPP Tanjung Awar -Awar, Tuban are within acceptable limits determined by BAPETEN. Moreover, by using the ERICA Assessment Tool Tier 2 program, the results of the analysis showed that there was no impact of 226Ra radiation on marine organisms at the research site. It caused the total radiation dose rate (external + internal) of 226Ra on marine biota was lower than the screening level of 10 µGy / h. Thus, it will not harm marine ecosystems and the sustainability of marine organisms in the waters of Tanjung Awar-Awar Coal-Fired Power Plant, Tuban.

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