

Analysis of ^{137}Cs Radionuclide Content in Sediment in Musi Watershed Using Gamma Spectrometer and its Affecting Factors

T. A. Jaya¹, A. Mara¹, G. F. Amri²

¹Department of Chemistry, Faculty of Mathematics and Natural Sciences, Sriwijaya University,
Jl. Raya Palembang - Prabumulih Km. 32 Indralaya, OI, Sumatra Selatan 30662, Indonesia

²Center for Safety Technology and Radiation Metrology, National Nuclear Energy Agency (BATAN),
Jl. Lebak Bulus Raya No. 49, Jakarta 12440, Indonesia

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ABSTRACT

The concentration of the radionuclide ^{137}Cs on sediment in watershed in Palembang has been analyzed. This study aims to determine the influence of sampling location and the water quality indicators of water pH, sediment pH, conductivity, turbidity, and sediment type on the concentration of ^{137}Cs and to determine the distribution pattern of ^{137}Cs in sediments. Sampling was conducted at seven stations spaced approximately 5 km apart, placed from the western end to the eastern end of the Musi river segment located within Palembang City. Sediment samples were prepared and their ^{137}Cs contents were measured with gamma spectrometry. The results showed that their ^{137}Cs concentrations ranged from below MDC (minimum detectable concentration) to 1.51 Bq/kg. This was within the 1×10^3 Bq/kg limit set by the quality standard. The varied and very low concentrations of ^{137}Cs are thought to have originated from global fallout. The location point of sampling affects the concentration of radionuclide ^{137}Cs while the characteristics of water quality are do not. The ^{137}Cs concentration spread pattern in Musi sediment is influenced by tidal currents and river morphology.

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INTRODUCTION

Nuclear science and technology is progressing rapidly, as is the application of radioisotopes in various countries in the world including Indonesia. Radioactive substances can occur naturally or be created by humans [1]. Artificial radionuclides are widely used to improve human welfare in various fields such as industry, agriculture, hydrology, and medicine [2]

One of the artificial radionuclides is a product from nuclear reactor fission, namely ^{137}Cs which has a long half-life of 30.17 years [3]. The radionuclide ^{137}Cs is widely used for equipment calibration, radiation detection, and medicine. It is also used in petroleum, construction, paper, and plastic industries [4].

The artificial radionuclide ^{137}Cs have negative effects. It can be spread by industrial or medical

wastes from facilities using the nuclide and from radioactive substance released due to by nuclear experiments in the air or also known as fallout [5]. These pollutants will settle into the riverbed and affect such organisms as macrozoobenthos [6]. Meanwhile, the Musi River supports the needs of the people of Palembang City for such uses as washing, bathing, drinking water, irrigation for rice fields, and industrial water supply.

River water that continuously carries sediment particles from the land can cause sedimentation. The distribution of sedimentation has an impact on the transport of the ^{137}Cs radionuclide. The nuclide's transport is also influenced by currents and river morphology. The nuclide can affect human health if entering the body, either directly through water consumption or indirectly through the food chain. If the dose imparted by the nuclide that enters the body is excessive, it can cause serious effects such as cancer and even death, since it affects organ function after accumulating in the human body. The ^{137}Cs radionuclide has a high radiotoxicity and

*Corresponding author.

E-mail address: tataabdianjaya6@gmail.com

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can accumulate in the body with the critical organs being the liver, muscle, and spleen [7].

Based on a previous determination of the ^{137}Cs content in the soil in several areas in South Sumatra Province [8], the concentration of ^{137}Cs in the soil in South Sumatra Province at depths of 5-20 cm ranged from 0.17 Bq/kg to 1.50 Bq/kg, while at depths of 0-5 cm it is below the minimum detectable concentration limit (MDC) of 0.17 Bq/kg. Please note that in this work, the concentration of ^{137}Cs refers to ^{137}Cs activity per unit mass of sediment. Based on the description above, this research project was carried out on the content of ^{137}Cs in the sediments in the Musi River watershed passing through Palembang City because the sediment there contains most of the chemical elements in the waters in the form of deposited particles [9].

This research aims to analyze the effect of sampling locations and the effect of water quality characteristics on concentrations of ^{137}Cs in sediments in the Musi River Basin passing through Palembang City, South Sumatra, and determine the distribution pattern which will later be compared with the threshold values set by the IAEA.

EXPERIMENTAL METHODS

Sampling location

The sampling locations were only on the segment of Musi River within Palembang City. As shown in Fig. 1 (attached), sampling was performed from the western end of the aforementioned river segment to the eastern end that is in direct contact with the Palembang City boundary. The positions of the sediment sampling points were determined using GPS (Global Position System) with a purposive sampling method. Sampling was carried out at seven stations spaced approximately 5 km. [10,11].

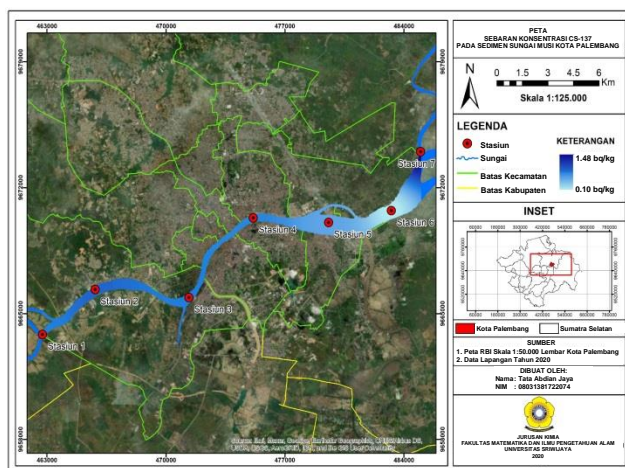


Fig. 1. Map of the radionuclide distribution pattern that showing locations of sampling of study.

Sediment sample preparation

The sediment samples that have been taken are cleaned of various impurities and dried at 105 °C in an oven. Then the samples were pounded with a mortar and sieved with a 200-mesh sieve. The results of the sieve were put into a plastic clip and the samples were taken to the PTKMR BATAN laboratory. Each sample was put in a 1000-mL Marinelli beaker and labeled. The samples were then weighted, the containers were closed, and their weights were recorded. The samples were then ready to be measured with a gamma spectrometer [12].

Measurement of water quality characteristics

Water quality characteristics were measured by determining the pH using a pH meter. Determination of sediment pH was carried out using the procedure proposed in [13]. Turbidity measurements were done using a turbidimeter. Conductivity measurements were carried out using a conductometer. The determination of the type of sediment was carried out in reference to the research [14]. Calculation of the percentage by weight of the sediment fraction is done using Eq. (1).

$$\text{Percent by weight} = \frac{\text{fraction weight } i}{\text{total sample weight}} \times 100 \quad (1)$$

Determination of the ^{137}Cs radionuclide concentration using a gamma spectrometer

Before measurements, the gamma spectrometer was calibrated for energy and efficiency. The energy calibration of the gamma spectrometer was carried out to determine the relation between radionuclide energy and the channel. The energy-channel relation was determined by counting ^{137}Cs point sources. Those sources were placed simultaneously at a distance of 15 cm from the detector [15].

Quantitative analysis using a gamma spectrometer requires efficiency calibration. A radioactive source always emits radioactive rays in all directions. Since radioactive samples are measured at a certain distance from the detector, only a part of the gamma rays emitted by the samples are detected. Therefore, it is necessary to calibrate the absolute efficiency of the total absorption peak. The efficiency can be written mathematically as in Eq. (2) [16].

$$\mathcal{E} = \frac{N_s \cdot N_{BG}}{t_s \cdot t_{BG}} \cdot \frac{A_t \cdot P_\gamma}{A_t \cdot P_\gamma} \quad (2)$$

In (2), \mathcal{E} is counting efficiency (%), N_s is standard count (count), N_{BG} is background count (count), t_s is duration of standard counting (seconds), t_{BG} is duration of background counting (seconds), A_t is standard concentration at the time of measurement (Bq), and P_γ is abundance of gamma energy (%).

Sample measurement

Before counting the sample, the background count rate measurement was carried out by counting the empty Marinelli tube. Samples were counted using a gamma spectrometer with a high-purity germanium (HPGe) detector. The radionuclide ^{137}Cs was counted directly using a gamma spectrometer for 3600 s. The concentration of ^{137}Cs was determined at a peak energy of 662 keV with P_γ of 85 %.

Data analysis

Both qualitative and quantitative descriptive analyses were performed to estimate the ^{137}Cs concentration. The results of the analysis of the concentration of ^{137}Cs were compared with the highest levels permitted by the IAEA regarding the determination of the quality standard of ^{137}Cs radionuclide in sediments, which is 1.108×10^3 Bq/kg.

Qualitative analysis

Qualitative analysis is needed to determine the elements contained in the sample. It can be performed manually by looking at the energy of the gamma rays emitted. Each radionuclide has a certain, distinct, and specific energy. The ^{137}Cs radionuclide emits gamma rays with an energy of 662 keV.

Quantitative analysis

The concentration of ^{137}Cs in the sample can be obtained after determining the time of counting using Eq. (3) [17].

$$A_t = A_0 e^{-\frac{0.693t}{T}} \quad (3)$$

In (3), A_t is concentration at the time of counting, A_0 is initial concentration, t is time delay between the stipulated initial moment and the counting time (days), and $T_{1/2}$ is half-life (days). Furthermore, A_t is used in the calculation of the efficiency calibration. The efficiency of counting can be calculated by (2).

The radionuclide concentration can be calculated using Eq. (4).

$$C = \frac{N_s - N_{BG}}{\mathcal{E} \cdot P_\gamma \cdot W} \times 100 \quad (4)$$

In Eq. (4), C is the concentration of radionuclides (Bq/kg), \mathcal{E} is efficiency of count (%), and W is sample weight (kg).

Distribution pattern of ^{137}Cs radionuclide on the Musi River, Palembang City

This distribution pattern was generated using data that includes field data (primary data) and supporting data (secondary data). The field data in the form of ^{137}Cs radionuclide concentration data was obtained from samples, while secondary data or supporting data was obtained from the map data of the Indonesian map of Palembang City with the help of SMS (Surface Water Modeling System) 8.01 software.

RESULTS AND DISCUSSION

Characteristics of water quality in the Musi River, Palembang City

The results of this study on several environmental parameters, namely water pH, sediment pH, conductivity, turbidity, and type of sediment, can be seen in Table 1.

Table 1. Water quality characteristics of samples collected in the study area in South Sumatra.

Environmental Parameters	Station						
	1	2	3	4	5	6	7
Water pH	6.6	6.8	7.0	6.7	6.8	6.4	6.0
Sediment pH	6.5	6.8	6.9	6.8	6.7	6.4	6.0
Conductivity	39.7	61.9	46.2	85.9	89.0	94.0	63.3
Turbidity	4.4	4.6	4.8	4.9	6.3	7.1	4.1
Type of Sediment	silt	silt	silt	silt	silt	silt	silt

The pH value of the water in this study ranged between 6.0 and 7.0. Meanwhile, the pH value of the sediment ranged from 6.0 to 6.9 with an average of 6.6. Based on the quality standard Regional Regulation of Palembang City Number 02 of 2003 [18], water is accepted as good if it has a pH value in the range of 6.0 - 9.0. Therefore, it can be concluded that the pH of the Musi River in the study area is still within the permitted limits. The compound that greatly affects the pH value is the carbonate contained in water. Carbonate content is influenced by CO_2 input from the air and from the water.

The pH value can affect the toxicity of a chemical compound; the higher the pH value, the lower the carbon dioxide level. The ions Cl^- , $\text{CO}_3^{2-}/\text{HCO}_3^-$, and SO_4^{2-} are anions that are generally found in water. These anions can accumulate in aquatic sediments and under certain conditions can be released back into the water body [19].

The conductivity values obtained in this study ranged from 39.7 $\mu\text{S}/\text{cm}$ to 94.0 $\mu\text{S}/\text{cm}$ with an average 72.8 $\mu\text{S}/\text{cm}$. The electrical conductivity of the surface water sample did not exceed the limit determined based on Government Regulation No. 20 of 1990 [20] which is 125 $\mu\text{S}/\text{cm}$. Thus, the conductivity in the study area is still relatively good. Conductivity is greatly influenced by the content of ions entering the water body. Those include the ions of such elements as lead (Pb), arsenic (As), cadmium (Cd), mercury (Hg), chromium (Cr), nickel (Ni), calcium (Ca), and magnesium (Mg). Their presence results in a polluted environment [21].

The turbidities found in this study ranged from 4.1 NTU to 7.1 NTU with an average of 5.2 NTU (Nephelometric Turbidity Unit). Based on the Regulation of the Minister of Health of the Republic of Indonesia Number 70 of 2016 regarding the standards and health requirements of the industrial work environment, the maximum acceptable turbidity is 25 NTU. The turbidities obtained in this research area satisfy the specified quality standard. Turbidity is basically caused by the presence of colloids, organic substances, mud, clay, and floating objects that do not settle.

Table 2. Classification of sediment grain size of samples collected in study area in South Sumatra.

Station	Sediment Weight Percentage (%)			
	Coarse sands	Medium sand	Fine sand	silt
	(0.5-2 mm)	(0.25 - 0.5 mm)	(62.5 - 250 μm)	(3.9 - 62.5 μm)
1	0.53	2.67	20.15	76.65
2	0.27	0.89	26.40	72.44
3	0.42	2.32	24.80	72.46
4	1.48	3.66	24.36	70.51
5	1.22	3.02	20.56	75.20
6	1.53	3.12	20.55	74.80
7	2.40	3.35	20.84	73.41

The analysis of sediment types can be seen in Table 2 which is determined in terms of the percentage weight of the sediment fraction calculated using Eq. (1). The results of the classification analysis of sediment types show that the types of sediment found in the research location include coarse sand, medium sand, fine sand and silt with sediment weight percentages in the ranges of

0.27-2.40; 0.89-3.66; 20.15-26.40, and 70.51-76.65, respectively. From these results, it can be seen that the type of sediment in the water of the Musi River in the study location is dominated by silt (3.9-62.5 μm).

Determination of ^{137}Cs radionuclide concentration in sediment samples using a gamma spectrometer

Energy calibration

Energy calibration was carried out by counting the standard mixed gamma radioactive source coded EW670, containing ^{137}Cs and ^{60}Co radionuclides with known concentrations. The distribution of channel number data and energy on the standard mixed-radionuclide gamma source EW679 can be seen in Fig. 2.

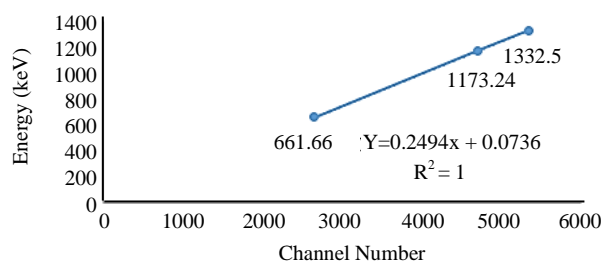


Fig. 2. Energy calibration graph showing a relationship between energy and channel number.

The result of the energy calibration in Fig. 2 shows a linear relationship between the energy and the channel number for the standard radionuclide. When expressed in graphical form, a straight line will be formed which can be seen in Fig. 2 with the regression value $R^2 = 1$ which indicates that the gamma spectrometer is suitable for use.

Efficiency calibration

Efficiency calibration is done by counting the standard mixed gamma source EW679 whose energies are known. The census was done using the HPGe detector model GEM F5930 with MAESTRO software and was carried out for 3600 seconds.

Table 3. Concentration of standard sources at the time of counting.

Radionuclide	Initial concentration (Bq/kg)	Delay Time (day)	Half Time (day)	Concentration during enumeration (Bq/kg)
Cs-137	1806.60	146	10.958	1789.99
Co-60	142.9	146	1925.5	135.58
Co-60	142.9	146	8021	135.58

It can be seen in Table 3 that the standard source concentration at the time of counting is calculated using Eq. (2). The value of the standard source concentration at the time of counting is used in calculating the efficiency calibration. This calibration was performed to determine the relationship between energy and efficiency. The efficiency is calculated using Eq. (3). Radioactive substances always emit radioactive rays in all directions. The measurement of samples of radioactive substances is carried out at a certain distance from the detector. Therefore, only a part of the gamma rays are detected by the detector.

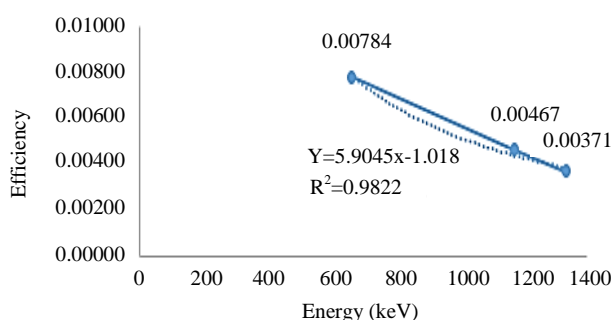


Fig. 3. Efficiency graph showing a relationship between efficiency and energy.

The results of the efficiency calibration used to calculate the efficiency of the count can be seen in Fig. 3. In this study, the efficiencies were obtained as 0.00784 cps/dps, 0.00467 cps/dps, and 0.00371 cps/dps for energies of 661.66, 1173.24, and 1332.50 keV, respectively. This result is reasonable when compared with another study [15] which gives an efficiency of 0.00104 % at 661.66 keV.

Data on spectrum and concentration of radionuclide ¹³⁷Cs

The results of examining the spectrum showed that the Cs-137 radionuclide was present at all stations. This can be seen by examining the peaks in the spectrum from the sample counting at an energy of 661.66 keV. Each radionuclide exhibits distinct and specific radiation energies. The gamma energy spectrum of ¹³⁷Cs can be seen in Fig. 4.

Quantitative calculation of the ¹³⁷Cs radionuclide concentration is carried out by looking at the data from the gamma spectrometer calculation contained in the samples at each station. The calculation uses Eq. (4) in order to obtain the results presented in Table 4.

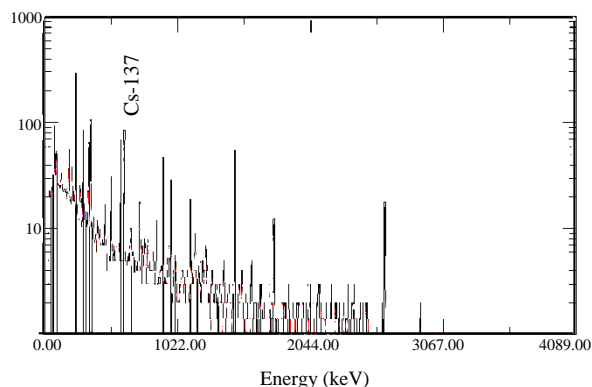


Fig. 4 Gamma energy spectrum of Cs-137.

Table 4. ¹³⁷Cs radionuclide concentration at each station.

Station No.	Concentration(Bq/kg)
1	0.78 ± 0.03
2	0.94 ± 0.04
3	0.79 ± 0.03
4	0.75 ± 0.02
5	0.53 ± 0.02
6	<MDC
7	1.51 ± 0.10

The data from the quantitative analysis is shown in Table 4. It shows that the concentration of ¹³⁷Cs in the sediments ranges from undetected, or below the MDC, to 1.51 Bq/kg. It can be seen that the concentration of ¹³⁷Cs is the lowest, below the MDC, for station 6. This result shows that station 6 has a lower concentration than the detector's ability to detect radionuclides. Meanwhile, the highest radionuclide content was found at station 7, namely 1.51 Bq/kg. Overall, the results of this study indicate that the content of ¹³⁷Cs in the sediment of the Musi River within Palembang City is still far from reaching the threshold of 1×10³ Bq/kg [22], or failing the quality standard set. The aforesaid standard is based on the Decree of the Head of the Nuclear Energy Supervisory Agency on the Standard of Radioactivity Level in the Environment [22].

Table 4 shows that the ¹³⁷Cs concentration is low and greatly varied. This may be due to a radionuclide source from a global fallout or radioactive fallout. Radionuclides released as a result of nuclear experiments or from nuclear facilities will be scattered into the air until they reach the stratosphere, causing ¹³⁷Cs to spread over a wider area due to wind gusts that can descend to the other hemisphere.

Even though Indonesia is far from nuclear activities, the global fallout has an impact on the presence of ¹³⁷Cs in Indonesia. The reactive nature of ¹³⁷Cs makes it easy to bind to particulates and get carried by the transfer of global water masses that pass through Indonesia to Palembang City.

In addition, radioactive fallout into the sea will undergo an evaporation process, and radioactive particles will participate in cloud formation and fall as rain in other areas, entering river waters [23].

A previous research in 2003 on the content of ^{137}Cs in soil in several areas in South Sumatra Province by Emlinarti and Buchari [8] showed that ^{137}Cs concentrations in soil in the South Sumatra Province ranged from 1.5 to 1.99 Bq/kg, while in this study, the highest content of ^{137}Cs in sediment in the Musi River was 1.51 Bq/kg.

Relation of water quality characteristics to radionuclide concentration of ^{137}Cs

Environmental parameters that tend to be dynamic do not have a strong relationship with the concentration of ^{137}Cs . We can see this by comparing the graph of the relationship between water characteristics and the concentration of ^{137}Cs .

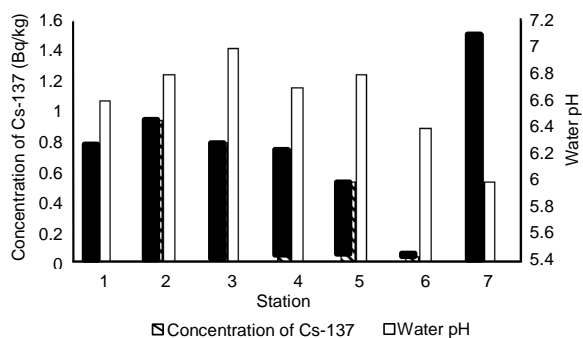


Fig. 5. Graph of relationship between water pH and sampling location on one hand and the concentration of ^{137}Cs on the other.

It can be seen in the graph in Fig. 5 that the water pH is quite varied while the concentration of ^{137}Cs varies. For example, at station 7, the concentration of ^{137}Cs is higher than at the previous station, while the pH is lower. In contrast, at station 6, the ^{137}Cs concentration is below the minimum detection limit while the water pH is high. It is assumed that water pH is not related to the concentration of ^{137}Cs .

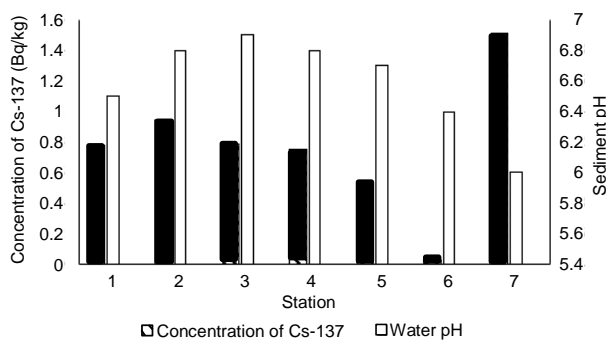


Fig. 6. Graph of the relationship between pH sediment and sampling location against the concentration of ^{137}Cs .

The graph of the relationship between sediment pH and sampling location to the concentration of ^{137}Cs can be seen in Fig. 6 which is almost the same as the graph of the relationship between the pH of water, where the sediment pH is quite varied while the ^{137}Cs concentration varies. It is assumed that the measured sediment pH has no relationship to ^{137}Cs concentration.

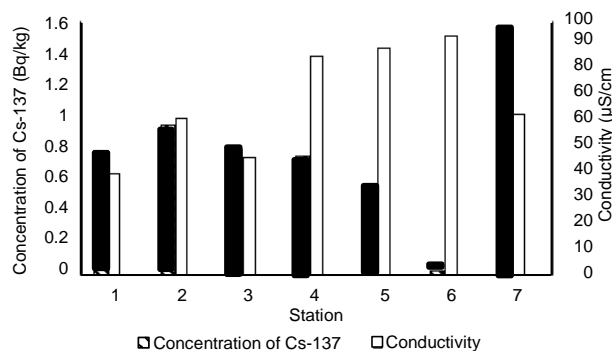


Fig. 7. Relationship between conductivity and sampling location against the concentration of ^{137}Cs .

It can be seen in Fig. 7 that station 6 has a conductivity of 94.0 $\mu\text{S}/\text{cm}$ which is the highest of all stations. However, the concentration of ^{137}Cs at station 6 was below the MDC. In contrast, as water flows from station 4 to 5 and 6, the conductivity stays quite high despite the ^{137}Cs concentrations decreasing to below the MDC. This shows that water conductivity and concentration of ^{137}Cs have no relationship.

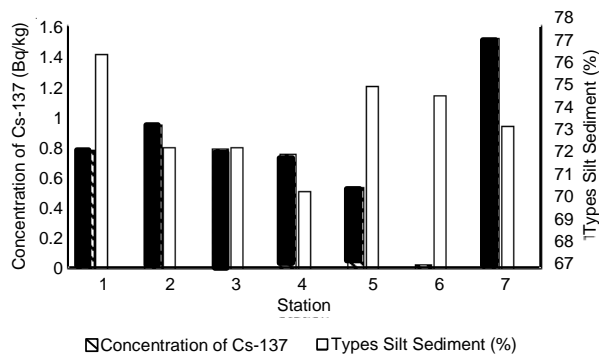


Fig. 8. Relationship graph percentage of species silt in sediment and sampling location and concentration of ^{137}Cs .

Figure 8 presents a graph of the relationship between the percentage of silt in the sediment and the location of sampling on one hand and the concentration of ^{137}Cs on the other. It is apparent that the percentage of silt sediment types has no relationship with the concentration of ^{137}Cs . This can be seen as at station 1 which has a fairly high percentage of silt sediment and a low concentration of ^{137}Cs . In comparison, station 6 has a concentration of ^{137}Cs below the MDC despite a fairly high percentage of silt in the sediment.

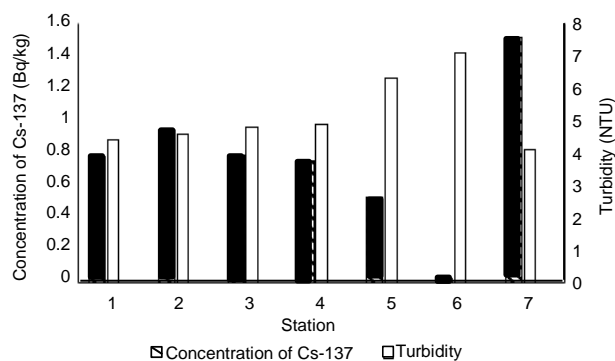


Fig. 9. Relationship between turbidity and location sampling against concentration of ^{137}Cs .

As shown in Fig. 9, station 7 has the lowest turbidity of 4.1 NTU, despite the highest ^{137}Cs concentration among all stations. This shows that there is no significant relationship between turbidity and the concentration of ^{137}Cs .

The distribution pattern of ^{137}Cs radionuclide on the Musi River, Palembang City

The preparation of this distribution pattern used data set that includes field data (primary data) and supporting data (secondary data). Field data in the form of ^{137}Cs radionuclide concentration data was obtained from samples, whereas secondary data or supporting data was obtained from the Indonesian map of Palembang City.

The results of the model approach with SMS 8.01 software package presented in Fig. 1 shows the distribution pattern of ^{137}Cs radionuclides in the sediments in the Musi River, Palembang City. The concentration is very low and varied. This is thought to have been influenced by the input of radionuclide sources from the fallout [24] and also due to the influence of the current pattern at high tide and low tide and the river morphology.

Station 7 has a fairly high concentration of ^{137}Cs . It is located in the downstream or eastern part. However, stations in the western or upstream areas such as stations 1 and 2 also have quite high concentrations of ^{137}Cs . This is influenced by the backflow of the Musi River immediately following a high tide; at that time, water from the sea flows to the river in an east-to-west direction. At low tide, however, the direction of the flow will be out of the river toward the sea/strait or from west to east.

The Musi exhibits a single tidal type where in one day there are one high tide and one low tide. This allows the ^{137}Cs radionuclide to be carried to the western station area when the river is in high tide, with the tide causing water to flow from the sea/strait to the river. The area under research has a river topography in the form of a basin with a

relatively greater river depth than other areas, making the current calmer and slower, thus allowing radionuclides to be transported in this area. At low tide, the current moves from the river to the sea or from west to east. Therefore, the radionuclides will be carried to the eastern region where, for instance, station 7 is situated. This is also confirmed by the results of research by Surbakti [25] which states that at high tide the water mass tends to move toward the southwest (toward the river), while at low tide the water mass moves to the northeast.

CONCLUSION

The point of the sampling location affected the distribution of ^{137}Cs radionuclide concentration. Water quality characteristics had no significant effect on the concentration of ^{137}Cs radionuclides. The distribution pattern of ^{137}Cs radionuclide concentrations in the sediments in the Musi watershed is influenced by tidal currents and river morphology

AUTHOR CONTRIBUTION

T. A. Jaya conducted the research and wrote the original manuscript. A. Mara and G.F. Amri performed the revision and improved the manuscript.

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