



Behavior of ^{137}Cs Activity in the Sayung Waters, Demak, Indonesia

Muslim^{1*}, H. Suseno² and M.J. Pratiwi¹

¹Department of Oceanography, Diponegoro University, Tembalang, Semarang 50275, Indonesia

²Marine 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

The behavior of anthropogenic radionuclide ^{137}Cs in the water and sediment has been examined with regard to particle size, organic sediment, and other physical conditions in the Sayung estuarine waters of Demak. Previously, this estuary was used as shrimp culture brackish water ponds that were affected by serious erosion and intrusion. Furthermore, this regency rapidly industrialized over the last three decades. The activities of ^{137}Cs in the six stations in water and sediment were in the 0.0116-0.4509 mBq/L and 0.3747-1.2442 Bq/kg ranges with mean activities of 0.2278 mBq/L and 0.8594 Bq/kg, respectively, depending on the prevailing physicochemical regime at the sampling station. The highest activities of aqueous ^{137}Cs occurred in station 2 but the highest in sediment occurred in station 6 located closed to the main land of Sayung. Fast current effectively diluted ^{137}Cs as a conservative radionuclide in seawater, thus the activity of ^{137}Cs in seawater decreased when the current speed increased. This effect was caused in part by industrial pollutants that adsorbed ^{137}Cs and precipitated onto the surface sediment. Organic matters and clay materials in sediment were not significantly effective at binding ^{137}Cs in Sayung waters in the industrial coastal area, but the correlation increased in open areas separated by a breakwater (second group stations) and the location further from the beach or in open sea. This indicates that industrial pollutants are more effective at binding ^{137}Cs than organic matters and clay materials. Fine grain size sediment is also more ineffective at binding ^{137}Cs ; it may be caused by the sediment's high total organic matter content.

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INTRODUCTION

The Fukushima Daiichi Nuclear Power Plant (NPP) accident on 11 March 2011 has caused panic throughout all countries in Asia Pacific, including Indonesia, the largest island country in the world. The long-lived artificial radioisotope ^{137}Cs (half life 30.2 years) is the main hazardous radionuclide of several produced in nuclear fission processes [1] and has become a focus of serious attention in addition to ^{90}Sr and $^{239,240}\text{Pu}$, since they are the most abundant anthropogenic radionuclides in the marine environment and can lead to the highest radiation doses to humans and marine biota, such as fish and

algae [2-4]. Several herbivores are capable of accumulating radiocaesium and can serve as reliable indicators of the degree of radiocaesium pollution. ^{137}Cs also represents a major source of radiation exposure to humans [5]. This radionuclide can be trapped by adsorption on clay mineral soil particles [6], remobilization, bioturbation, and migration due to sea currents [7] or ion exchange and precipitation as hydroxides or sulfides [8]. However, the association of organic matter with clay material may decrease their affinity for Cs and thus enhance its bioavailability [9]. Thus, the ^{137}Cs activity in seawater and bottom marine sediment can be influenced by chemical composition of seawater and sediment [10,11], water mixing and turbulence [12], and the sampling period and the physical and chemical characteristics of the sampling locations [13,14].

* Corresponding author.

E-mail address: muslim.ir@undip.ac.id

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Surveys in Indonesia between 2011 and 2013 have detected ^{137}Cs in sea water, sediment, and some marine organisms in most coastal areas and two offshore locations [10,14,15]. However, ^{134}Cs (half life 2.1 years) was never detected in any samples, indicating that ^{137}Cs was not produced in Indonesia, but resulted from global fallout [10,14,15]. Furthermore, radiocesium data in other Indonesian marine environments is still needed to obtain an accurate picture of radioecological condition in Indonesian marine waters. Anthropogenic ^{137}Cs is introduced to Indonesian sea surface waters by currents and other processes. In the water column, processes such as advection, dispersion, and precipitation are effective in removing this radionuclide from the water column by transporting it both horizontally and vertically to the bottom waters and sediments [16,17]. This study is part of the IAEA program that coordinated Asia-Pacific countries to form Regional Cooperation Agreements (RCA) to monitor the effects of the Fukushima Daiichi NPP accident in the Asia-Pacific region.

Demak is a regency in the Central Java province of Indonesia that is located east of Semarang, the capital city of Central Java. Annually, in the rainy season, flooding occurs in most of the coastal area, but the dry season is very arid, thus leading to the high sedimentation rate in rivers of the coastal area. This condition is the reason for the name of this regency, Demak, meaning swamp. Previously, the coastal area of Demak was used as brackish water ponds for semi intensive shrimp culture that had a serious negative effect on erosion. To restore the region, mangrove plants are being planted along the coastal area, and a breakwater was built to reduce erosion. However, industrial activities have developed quickly, increasing the rate of pollution. It is likely that sedimentation and pollution concentrate in the area between the coastline and the breakwater. Burial of marine-sourced organic carbon (OC) into the sediment, as commonly occurring in continental margins, also occurred in this area as a contribution from rivers [18,19] that most commonly linked to oceanographic regulation of bottom-water oxygenation (anoxia) and/or biological productivity [20].

Due to the aforementioned problems, this study attempts to determine the behavior of ^{137}Cs in the seawater and sediment in the waters of the Sayung district of Demak. It also assesses the relation between sediment carbon concentration, sediment composition, and particle size on the one side, and the ^{137}Cs activity on the other side.

EXPERIMENTAL METHODS

Sampling site

Water and sediment samples were collected onboard fishery ships at six sampling stations located in the Sayung waters of Demak, as shown in Fig. 1. The coordinates and depths of the sampling stations are presented in Table 1. The cruises are part of the marine science student water quality monitoring program in mid-October 2014. Seawater samples were taken from the surface, and sediments were collected from the sea floor. In addition, salinity, temperature, pH, and current profiles for water quality monitoring data were also collected simultaneously with the seawater and sediment samples. The sampling stations were separated into two groups. The first group consisted of stations 1 and 6, located to the southeast of the break water, in the closed coastal area. The second group, located to the northwest of the break water, in the open area, consisted of stations 2, 3, 4, and 5. The stations in the first group are more polluted and turbid compare with the stations in the second group, because the terrestrial waste or pollution concentrates in this area both in water column and bottom sediment.

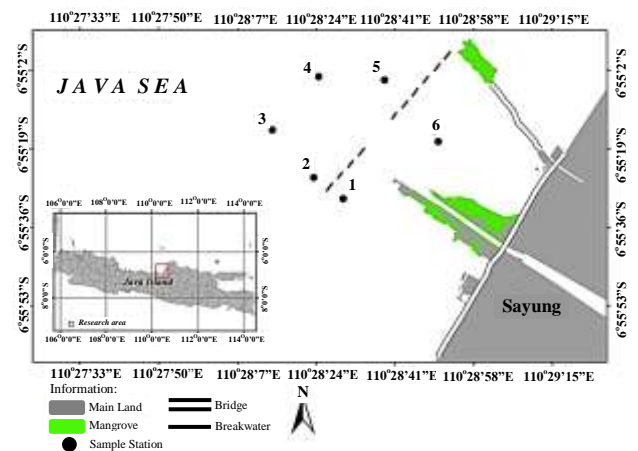


Fig. 1. Sampling station sites.

Table 1. Station site information

Station	Coordinate		Depth (m)
	Longitude	Latitude	
1	06 ° 55' 29.76"	110 ° 28' 29.82"	1.22
2	06 ° 55' 25.17"	110 ° 28' 23.45"	2.35
3	06 ° 55' 14.88"	110 ° 28' 14.52"	2.25
4	06 ° 55' 3.34"	110 ° 28' 24.51"	2.50
5	06 ° 55' 4.07"	110 ° 28' 38.75"	1.38
6	06 ° 55' 17.39"	110 ° 28' 50.34"	0.33

Field work

The predetermined stations were located by shipboard GPS (Global Position System). Sixty

liters of surface seawater samples for ^{137}Cs analysis were collected in ninety-liter acid-cleaned polyethylene buckets that were rinsed at least twice with surface seawater, before filling with the water sample. ^{137}Cs in seawater samples was precipitated with ammonium molybdophosphate (AMP), stirred for 30 minutes, and then allowed to settle in the form of cesium chloroplatinate sediment. The settled solid samples were then collected by siphon using plastic tubing into polyethelene bottles and stored in a plastic box and transported to the laboratory.

Sediment samples for analyzing ^{137}Cs , sediment texture, and organic content were obtained using sediment grab. The sediments were placed in black zippered plastic bags and transported to the laboratory.

Laboratory treatment and measurements seawater for ^{137}Cs analysis

The settleable solid samples were filtered using filter paper (Selectron BA 85, 0.45 μm) and the particles were dried in the oven at 80 $^{\circ}\text{C}$ for 2-3 days. The activity of ^{137}Cs in dried samples was counted using a gamma ray spectrometer. The gamma spectrometer system consisted of an HPGe detector (Canberra type GX2018) and a multichannel analyzer connected to a PC.

^{137}Cs and carbon analysis in sediment

Sediment samples were dried in an oven at 80 $^{\circ}\text{C}$ for 2-3 days and powdered with an agate mortar prior to analysis. To homogenize, the sediment samples were treated using a grinder. One kg of sediment was put on a Marinelli beaker. Then, the ^{137}Cs activity was measured using a gamma ray spectrometer.

To determine the total organic carbon (TOC) content, the powdered sediments were treated with 0.1 N HCl to remove the carbonate minerals. After drying, the sediments were heated at 550 $^{\circ}\text{C}$ for six hours. Particles sizes were analyzed according to Eleftheriou and McIntyre [21].

RESULTS AND DISCUSSION

^{137}Cs activity in seawater samples

The results of ^{137}Cs measurements for the surface seawater obtained by gamma-ray spectrometry are shown in Table 2. The activity of

^{137}Cs in the seawater of the second group (stations 2, 3, 4, and 5) were relatively more uniform compared to in the first group (stations 1 and 6), and appeared to decrease with increasing current speed. This means that the ^{137}Cs in the surface seawater is conservative and easy to dilute. The ^{137}Cs activity in station 3 was the lowest compared with other stations in the second group, due to the current being the strongest at that station. Furthermore, the ^{137}Cs was diluted by mixing, diffusion, and advection processes and/or small contributions of radioactive deposition [22]. The mean ^{137}Cs activity in seawater in the first group of stations is much lower than in the second group. This result is similar to a previous study within industrial waste effluent [10] which showed that ^{137}Cs activity decreased in the seawater with high pollutant concentration. The lowest ^{137}Cs activity occurred at station 6, which was the nearest to industrial waste effluent. The temperature, salinity, and pH of surface seawater in all stations were relatively consistent, indicating that these parameters did not affect the behavior of ^{137}Cs , especially since pH was unaffected [23].

Table 2. ^{137}Cs activity and the temperature, salinity, current speed, and depth of seawater at the sampling stations

Station	^{137}Cs Activity (mBq/L)	Temp ($^{\circ}\text{C}$)	Salinity (‰)	pH	Speed Current (m/s)	Depth (m)
1	0.1966	30.3	30.0	7.78	0.0129	1.22
2	0.4509	30.4	30.0	7.93	0.0109	2.35
3	0.0910	30.9	30.6	7.77	0.0178	2.25
4	0.3778	30.4	30.0	7.67	0.0054	2.50
5	0.2388	29.8	30.0	8.02	0.0089	1.38
6	0.0116	31.2	31.0	7.93	0.0075	0.33
Mean	0.2278					

^{137}Cs , carbon concentration, and particle size in sediment samples

It is important to note that the physical and chemical process that deposit ^{137}Cs in sediment do not occur in a short time frame. Thus, the current speed data in the surface seawater in this study can not be used to as a rationale for the behavior of ^{137}Cs in sediment. However, there is increasing evidence that organic matter plays a role in soil caesium affinity, causing bioavailability to increase as the organic matter content increases [24]. The activity of ^{137}Cs in sediment in this study ranged from 0.3747 to 1.2442 Bq/kg. The sediment's ^{137}Cs activity in the second group stations was relatively more uniform than first group stations. The concentrations of TOC in the second group stations

are also relatively uniform and the mean concentration was lower than in the first group stations. The observed difference may be due to TOC from terrestrial activities being prevented by the breakwater from getting advected to second group stations. The highest and the lowest ¹³⁷Cs activity in sediment occurred in the first group stations, namely stations 6 and 1, respectively (Table 3). The lowest ¹³⁷Cs activity in sediment occurred in station 1, but TOC and clay material concentrations were the highest at this station. The highest ¹³⁷Cs activity occurred in station 6 when the TOC concentration was lowest and the clay material content was relatively low. This suggests that ¹³⁷Cs activity will decrease when both TOC and clay material contents in sediment are high. Decreasing TOC and clay material contents in sediment will have the opposite effect, increasing ¹³⁷Cs activity. ¹³⁷Cs activity in sediment did not have a positive correlation with TOC and clay in the first group stations. Whereas, in the second group stations, when the TOC and clay contents were very low (station 5) the ¹³⁷Cs activity became the highest. It is thought that the increased ¹³⁷Cs activity in sediment was not caused by TOC and clay, but rather by industrial waste pollution in sediment that contains some minerals such as some trace metals that were able to bind ¹³⁷Cs. Other authors found that although clay contains illite mineral that has a strong capability for adsorbing ¹³⁷Cs, other pollutants such as heavy metals decreased this adsorption capability [25]

Table 3. ¹³⁷Cs activity, particle size, and carbon concentration in sediment

Sta- tion	¹³⁷ Cs activity in sedi- ment (Bq/kg)	Particle size (µm) (texture)				Carbon in sediment		
		Sand (62.5- 250) (%)	Silt (3.9- 62.5) (%)	Clay (<3.9) (%)	Silt+Clay (<63) (%)	TC (%)	TIC (%)	TOC (%)
1	0.3747	9.33	73.33	17.34	90.67	13.0	1.0	12.0
2	0.6466	47.61	42.59	9.81	52.40	9.6	1.4	8.2
3	0.8773	55.57	36.08	8.36	44.44	10.8	1.4	9.4
4	0.8654	34.02	49.37	16.61	65.98	6.6	0.8	5.8
5	1.1479	67.60	26.20	6.20	32.40	5.8	1.0	4.8
6	1.2442	64.80	28.32	6.88	35.20	12.8	3.4	9.4
Me an	0.8594							8.27

This phenomenon was different from what was observed in a previous study where ¹³⁷Cs was adsorbed by clay and organic matter; that is, ¹³⁷Cs activity increased when the clay and organic matter increased. In this study, the stations 1 and 6 as the first-group stations were located in industrial waste pollution area, where the chemical composition in

sediment is more complex, causing weaker ¹³⁷Cs adsorption to clay and organic matter. The increase of ¹³⁷Cs sediment in the industrial waste pollution area was possibly caused by some ions contained in waste pollution, which scavenged ¹³⁷Cs into sediment or an ion exchange and precipitation reaction as hydroxide or sulfide. Usually in coastal areas, there are higher levels of pollutants that compete with ¹³⁷Cs than in open seas [10].

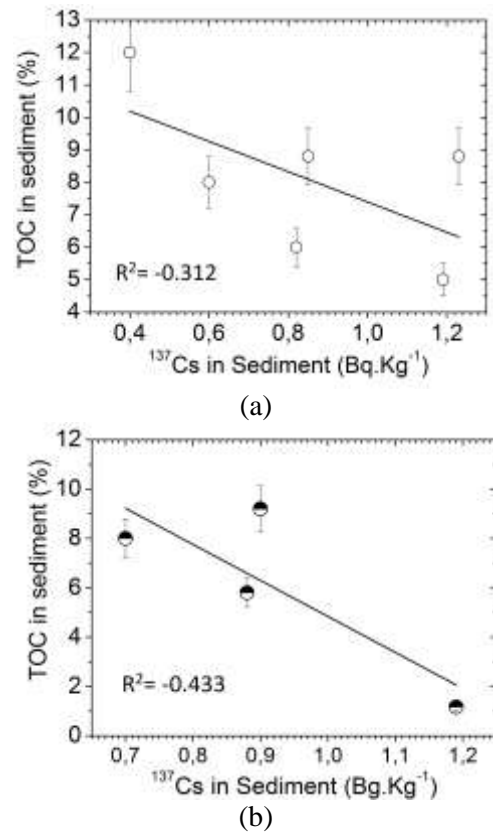


Fig. 2. Regression correlation between TOC (total organic carbon) and ¹³⁷Cs in sediment (a) all stations, (b) the four stations northwest of breakwater (second-group stations).

Figure 2 shows that, based on data from all stations, the TOC did not show a strong correlation with ¹³⁷Cs in sediment ($R^2 = -0.312$); however, when only the stations in the second group (station 2,3,4, and 5) were used, the correlation between TOC and ¹³⁷Cs in sediment was stronger ($R^2 = -0.433$). This may be caused by waste pollution, where the first group was more polluted than the second group and the two groups were separated by a breakwater. Furthermore, the correlation between % fine grain size (< 63 µm) and ¹³⁷Cs in sediment increased (Fig. 3; $R^2 = -0.781$). It has been recognized that ¹³⁷Cs does not have the ability to attach to fine-grained particles, and is able to serve as a tracer for water mass movement in the aquatic environment [26]. Thus, fine grain size sediment is more effective to protect ¹³⁷Cs binding than organic content.

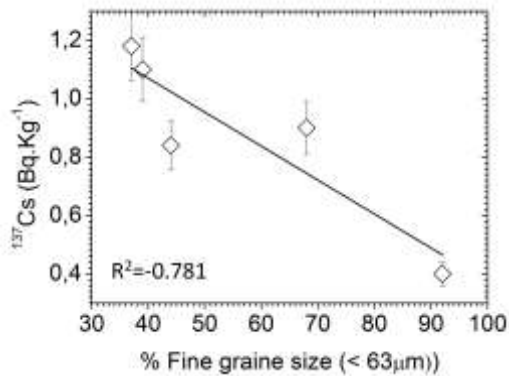


Fig. 3. Correlation between ¹³⁷Cs in sediment (Bq/kg) and % fine grain size (< 63 μm)

Table 3 shows that particles with size >250 μm (coarse) were not found, which means that the composition of sediments in Sayung estuary, Demak, did not contain any animal shells shale, such as sea shells and bivalves rich in CaCO₃ and/or SiO₂. Pyrtle and Scott [27] had found through their investigation that the Lena River estuary (Russia) exhibited strong inverse correlations between ¹³⁷Cs and % SiO₂ ($R^2 = -0.91$) and between ¹³⁷Cs and quartz ($R^2 = -0.91$), but strong positive correlation between ¹³⁷Cs and % Al₂O₃ ($R^2 = 0.97$). The results indicate that ¹³⁷Cs was not adsorbed by particles with a high % SiO₂ and % quartz; however, they show a stronger adsorption by particles containing high % Al₂O₃, which are generally present in clay materials (< 3.9 μm). In this study, the negative correlation between % fine grain size (< 63 μm) and ¹³⁷Cs in sediment increased (Fig. 3; $R^2 = -0.781$), despite the lack of coarse particles in sediment. This may be caused by the relatively high TOC concentration in the sediment in the Sayung, Demak estuary (mean: 8.27 %, Table 3). This TOC value is much higher compared with other sites, such as in Changjiang (Yangtze) River estuary of China, with a mean TOC concentration of only 0.57 % [28]. Thus, % fine grain particle size in Sayung-Demak did not bind ¹³⁷Cs, perhaps due to the high TOC in the sediment, produced from terrestrial erosion, despite the sediment's not containing coarse-size grains that are usually rich in SiO₂ and Al₂O₃ [27]. Thus, intensive human activities and increasing fertilizer use in the coastal area of Sayung-Demak contributed to the increasing organic carbon coastal environment, which will affect radiocesium activity in the environment.

CONCLUSION

The behavior of ¹³⁷Cs in Sayung-Demak waters is influenced by several factors as follows. First, the increasing current speed correlated with decreased ¹³⁷Cs activity in the seawater column, but not in the sediment. Second, the activity of ¹³⁷Cs in the open sea (second group) is more uniform than in closed area (first group) that contains much more pollutant from industrial wastes. Third, organic matters and clay sediments did not more strongly adsorb ¹³⁷Cs that comes from global fallout than pollutants did. Finally, the fine grain-size (< 63 μm) particles were not effective in adsorbing ¹³⁷Cs even though coarse particles were absent.

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