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Distribution of ^{137}Cs Radionuclide in Industrial Wastes Effluents of Gresik, East Java, Indonesia

Muslim[1*], H. Suseno[2] and F. Rafsani[1]

[1]*Department of Marine Science, Diponegoro University,*
Jl. Prof. H. Soedharto, SH, Tembalang, Semarang 50275, Indonesia
[2]*Center for Radiation Safety Technology and Metrology, National Nuclear Energy Agency,*
Jl. Lebak Bulus Raya No.49, Jakarta 12070, Indonesia

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ABSTRACT

The distribution of anthropogenic radionuclides ^{137}Cs was measured from industrial waste effluent of Gresik to Gresik Sea in east Java, Indonesia. The activity of ^{37}Cs detected at all stations was much lower than in northeast Japan both before and after NPP Fukushima accident. This indicated that in Gresik industrials waste did not consist of ^{137}Cs. The lowest activity ^{137}Cs occurred at the station nearest to the industrial waste effluent that contained some particle ions that were able to scavenge ^{137}Cs and then precipate this radionuclide. Furthermore, the greatest ^{137}Cs occured at the station that has high current speeds that stirred up sediment to release ^{137}Cs in seawater as a secondary source. The lowest salinity did not effect on the activity of ^{137}Cs even though the lowest salinity and activity ^{137}Cs occured at the same station.

INTRODUCTION

Indonesia is one of the countries in the world which do not use nuclear energy, but several artificial radionuclides have been detected in several locations in the sea water, sediment, and marine organisms in Indonesian waters [1]. Transport of artificial radionuclides to Indonesia from the sources of the radionuclides can cause surface infusion of these isotopes into the sea, including atmospheric transport and subsequent transport to off-shore and deeper regions [2] by current and horizontal mixing [3-5], or by wind-driven circulation of the surface waters [6] and global currents such as the Mindanao Current, the Great Ocean Conveyor Belt and the Indonesian Throughflow, which are the important currents which transport huge amounts of seawater from the Pacific Ocean to the Indian Ocean [7].

^{137}Cs is soluble in seawater and is anthropogenic [8], and has been recognized as a part of global nuclear fallout since the earliest nuclear weapons were detonated in New Mexico and Japan in 1945. This radionuclide is transported globally with water masses in the oceans [9,10] which is then advected and diffused to the worlds ocean [11]. The Chernobyl and Fukushima accidents on 26 April 1986 and 11 March 2011, respectively, have also produced some artificial radionuclides globally. Therefore, serious attention must be given to this worldwide pollution problem as these radionuclides can be concentrated in surface soils and sediments [12]; thus, many countries need to seriously investigate the activity concentration and distribution of ^{137}Cs in the sea as this radionuclide has a long half life (30.2 y) and can accumulate in the soil, plants, and marine organisms [13-15].

Gresik is a city in the East Java province of Indonesia which is located in the northern part of the Java Island and connected to the Java Sea by the Madura Strait in the north part. Its industries are diverse, among others including palm oil, industrial chemicals (e.g. oxygen, nitrogen, argon, acetylene), phosphate fertilizers, detergents, ceramics, tiles, paper, wood processing, rubber, electrics,

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[*] Corresponding author.
E-mail address: muslim.ir@undip.ac.id
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steel, cement, asbestos, fish processing, glass, shoes, paints, pesticides, textiles, and wood furnitures. All of the produced industrial wastes flow through the river which flows to the Java Sea. Janssen *et al.* [16] and Morita *et al.* [17] found that ^{137}Cs will accumulate in soil, plants and marine organisms and are affected by competing ions such as potassium, calcium, ammonium, and other inorganic minerals such as other monovalent cations [18]. In order to determine the sources and present levels of ^{137}Cs in the coastal waters of Gresik, hydrological factors which affect the nuclide's distribution process in the water column, such as currents, salinity, and pH, were determined.

The objective of this study is to determine the distribution of ^{137}Cs in the waters around Gresik, Indonesia (part of the Java Sea), and whether this distribution is affected by the pollution in the area which can be correlated with hydrological factors. A previous study about the monitoring ^{137}Cs and ^{134}Cs at marine coasts in Indonesia between 2011 and 2013 had been conducted. ^{137}Cs was detected in all study areas but ^{134}Cs was not detected and it was concluded that the ^{137}Cs in marine coasts in Indonesia originated from global fallout. The concentration of ^{137}Cs in Gresik waters were compared with distribution of industrial waste effluents.

EXPERIMENTAL METHODS

Sampling

In September 21, 2013, the collection of seawater samples was carried out. The surface seawater samples were taken using a 10 L plastic bucket and then collected into big polyethylene containers (80 L) aboard the ship. The containers were rinsed at least twice with seawater collected from the location before the final samples were taken. The total volume of seawater samples taken at each station was 60 L. Sampling site stations were located in the northern region of the Gresik area, as shown in Fig. 1, and the coordinates and depths of the sampling stations are presented in Table 1. The cruises were a part of the monitoring program of the impact of the Fukushima radioactive releases in the Asia-Pacific Region in Indonesia, itself a part of IAEA program. Seawater samples were taken from surface seawater and along the water profile with simultaneous current, salinity, pH, and temperature profiling.

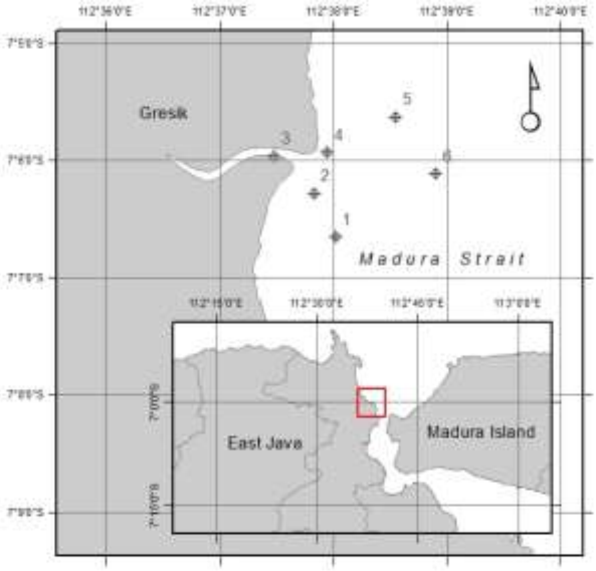


Fig. 1. Map of the sampling sites.

Table 1. Station site information

Station	Coordinate		Depth
	Longitude	Latitude	(Cm)
1	112° 37.792'	07° 06.473'	80
2	112° 37.725'	07° 06.190'	68
3	112° 37.534'	07° 05.930'	341
4	112° 37.874'	07° 05.868'	127
5	112° 38.038'	07° 05.76'	143
6	112° 38.289'	07° 05.984'	123

Analysis

The activity concentration of ^{137}Cs in sea water was determined by using the ammonium molybdophosphate (AMP) method according to IAEA procedure. This method has been successfully applied to the analysis of ^{137}Cs in small volumes of seawater [19,20]. Cesium was precipitated as a chloroplatinate. The overall chemical yield was determined gravimetrically and the samples were counted on a low-background beta counter.

Before precipitating the contained cesium by AMP, the 60 L of seawater was acidified with 4 M HCl (to a pH of about 1). As much as 20 mg of Cs carrier was added and stirred for 1-2 hours by bubbling air or N_2 gas. Then, the samples were mixed with 13 g AMP to make a slurry, and the solution was stirred for at least 30 minutes to absorb Cs into the AMP. The AMP settled in the container overnight. The supernatant was siphoned off and discarded. The AMP was collected in a smaller beaker and allowed to settle. This solution was centrifuged for 10 minutes and the supernatant was discarded. The AMP was then dissolved in 10 N NaOH and centrifuged to remove undissolved solids. Afterward, the solution was boiled to eliminate ammonia; the vapor was checked with wet pH paper until NH_3 emission ceased. The sample was subsequently diluted with 500 mL of distilled water and the pH was brought to 1.5-2 when the solution turned yellow. The ^{137}Cs activity was determined by its photon/gamma emission at 661 keV while the ^{134}Cs activity was also determined from emissions at 605 and 796 keV. The detector was calibrated by counting a standard solution of ^{137}Cs, using the same counting geometry.

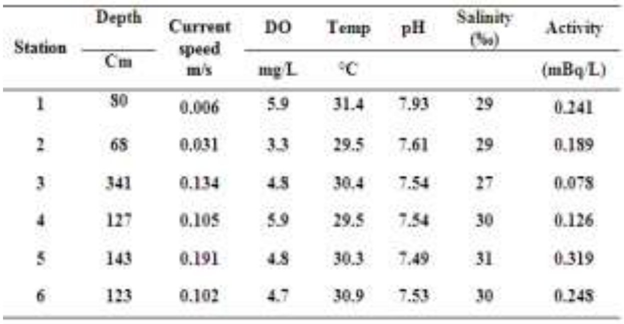
RESULTS AND DISCUSSION

^{137}Cs activity

The ^{137}Cs activity concentration in Gresik seawaters was detected and found to be persistent even though the Gresik site is very far from the source of the Fukushima incident. This radionuclide activity could also reflect fallouts from past nuclear bomb tests and the nuclear power plant accident of Chernobyl because ^{137}Cs is a long-lived radionuclide ($T_{1/2}$ = 30.2 y).

The activity concentration of ^{137}Cs fluctuated at each station sampled with a range of activity concentration from 0.078 to 0.319 mBq/L (Table 2) which is similar to the results of the previous study in marine coasts in Indonesia between 2011 and 2013 (0.11 to 0.66 mBq/L) but much lower than the activity concentration in northeast Japan both before and after the Fukushima nuclear power plant (NPP) accident, as measured in April-December 2009 and May 2012, respectively. Concentrations reported have been 2-3 mBq/L and 2.5-4 mBq/L, respectively [2]. These results indicated that ^{137}Cs was not present in the industrial waste effluent of Gresik. The lowest activity concentration occurred at station 3 which had the greatest depth. The low activity concentration is probably due to the soil particles (suspended sediments) at station 3 which contained some ions such as potassium, calcium, ammonium and other inorganic ions which scavenged ^{137}Cs and precipitated the radionuclide to the bottom sediments; thus, the concentration of ^{137}Cs in sediment which have received deposition of radionuclides will increase [21]. However, Aoyama *et al*. [19], Evangeliou *et al*. [22], and Nakanishi *et al*. [23] found that the activity concentration of ^{137}Cs in deep water and coastal water in Japan decreased with depth or was concentrated in surface sediments. The activity concentration of ^{137}Cs in the surface seawater was much higher than in the sub-surface layers.

Table 2. Water quality, current speed and ^{137}Cs activity at Gresik.



Station	Depth	Current speed	DO	Temp	pH	Salinity (‰)	Activity
	Cm	m/s	mg/L	°C			(mBq/L)
1	80	0.006	5.9	31.4	7.93	29	0.241
2	68	0.031	3.3	29.5	7.61	29	0.189
3	341	0.134	4.8	30.4	7.54	27	0.078
4	127	0.105	5.9	29.5	7.54	30	0.126
5	143	0.191	4.8	30.3	7.49	31	0.319
6	123	0.102	4.7	30.9	7.53	30	0.248

The highest activity concentration of the^{137}Cs radionuclide occurred at station 5 (0.319 mBq/L) caused by the resuspension of bottom sediments caused by high current speeds (0.191 m/s) which produced elevated ^{137}Cs activity concentration from those sediments as a secondary source.

Dissolved oxygen, temperature, pH, and salinity did not show any effects on the distribution of ^{137}Cs even though both the lowest salinity (27‰) and the lowest ^{137}Cs activity concentration (0.078 mBq/L) occurred at station 3. In this case, the lowest ^{137}Cs activity concentration was not caused by salinity, but it was most likely caused by the station's proximity to the estuary and its being the nearest station to the industrial waste effluent of Gresik that contained ions capable of scavenging^{137}Cs. Zalewska and Lipska [24] found that reduction of the activity concentration of ^{137}Cs occurs at stations closest to river-estuaries and in low-salinity water, because there ^{137}Cs is diluted by freshwater, and because the behavior of the radionuclide in the water column is affected by physical processes such as advection, mixing, and diffusion [25,26].

CONCLUSION

The activity concentration of ^{137}Cs in Gresik seawater was detected even though the concentration was very low compared with activities in the Sea of Japan, both before and after the Fukushima NPP accident. The results indicate that ^{137}Cs in Gresik did not come from industrial wastes effluent, but it probably originated from global fallout. Industrial waste particles could accumulate ^{137}Cs from the seawater and precipitate in bottom sediment, thus explaining why the activity concentration of dissolved ^{137}Cs at this location was low. The sediment could release its ^{137}Cs to the seawater and become a secondary source when current speeds are high.

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