^{137}Cesium Concentration of Seawater and Sediment in Medan, North Sumatra

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Abstract. *Cesium-137 is a fission product with a long half-life which can be released into natural environments from atomic bomb tests, nuclear accidents, and nuclear waste disposal. Because cesium is chemically similar to sodium, ingestion of its radioisotopes results in their deposition in the soft tissues all over the body creating an internal hazard, especially to the reproductive system. As an element in the alkali group, cesium commonly exists and migrates in the monovalent state in aquatic environments. In this research presents the results of measurements Cesium-137 from Medan seawater, North Sumatra. Sample of seawater and sediment were colected from five location on eastern sea Medan. The method has used in this experiment by precipitating Cesium-137 using Copper Hexacyanoferrates(III) to form complexes. Precipitate then dry on temperature 120° Celcius and Cesium-137 concentrations were determined by gamma spectroscopy. The result of sediment samples showed from each location was station 1 <BDL, station 2, 0.035±0.003 Bq/Kg, station 3, 0.047±0.005 Bq/Kg, station 4, <BDL, station 5, 0.19±0.02 Bq/Kg and result for seawater sample was station 1, 0.191±0.022 Bq/m^3 and station 2, 0.757±0.087.*

Keywords : *Cesium-137, Copper Hexacyanoferrates (III), Gamma spectroscopy*

Introduction

The major contribution to radioactive contamination of seawater in ocean was made by the radionuclides ^{137}Cs. These radionuclide were released to the environtment mainly through by human activity such as operation nuclear instalations, dumping of nuclear wastes into the world oceans, nuclear accidents, and atmospheric nuclear weapon tests (Eisenbud and Gesell, 1997; UNSCEAR, 2000). Among the fission products, ^{137}Cs is of special concern due to its long half life and biological hazards. Because the high solubility and chemically similiar to potassium, ingestion of its radioisotopes can be easily distributed in the soft tissues all over the body creating an internal hazard, especially to the reproductive system. As an element in the alkali group, Cs commonly exists and migrates in the monovalent state in aquatic environments (Su et al., 2000; Shakir et al. 2007).

The largest source of radioactive materials were fallout from atmospheric nuclear weapon test in the 1950s and 1960s, which had dispersed and deposited ^{137}Cs worldwide (Aarkrog, 2003). Then in recent years, the accident of the Fukushima Dai-ichi Nuclear Power Plant (FNPP) were released large amount of radionuclides ^{137}Cs and contaminated into terrestrial, aquatic and marine environment. This incident was arisen considerable interest with regard to the fate and transport of radionuclides in the aquatic environment (Kawamura et al., 2011; Koarashi et al., 2012; Thakur et al., 2013).

Therefore study the behavior of ^{137}Cs in the ecosystem were needed for future risk assessment and for management decisions concerning radioactively contaminated areas, regardless of the source of contamination. The objective of this study were measure the activities of ^{137}Cs contamination in surface seawaters and sediment of the Medan sea in North Sumatra.

Radionuclide Analysis

Environment samples were collected from 5 stations in off shore Medan, North Sumatra during June 2015. The seawater were collected approximately 100 liters at a depth 1 m below the sea surface and store in plastic container. A small quantity of sediment about 2 kg were collected in the sea floor using sediment grab. From each location 100 liters of seawater and sediment were transferred into containers and sealed. Every samples then were labeled with date and time, the Global Positioning System (GPS) locations and sample IDs noted. The samples were taken back to land-base laboratory for analysis.

For ^{137}Cs determination, the seawater samples were precipitated using chemical reaction $K_4Fe(CN)_6$ and $CuCl_2$. NaOH solutions was added for adjusted seawater sample to pH ~ 8 and stirred for 30 minutes to make a $CuFe(CN)_6^{-2}$ precipitation, then left for 24 hours settling. The clear supernate were pump out into another container and discard. The dark brown precipitated of $CuFe(CN)_6^{-2}$ were filtered using filter paper 0.45 μm and wash with distilled water. Then the precipitated were transferred into the oven at temperature 60 °C until dry and stored to bottle for counting using HPGe spectrometry.

Sediment samples were prepared by dried in oven at temperature 120°C for 2-4 days and then homogenize the dry sample until all of the material will pass through 63 μm sieve. The dried samples were transferred to polyethylene Marinelli beakers of 1200 cm^3 capacity and sealed. ^{137}Cs specific activities were measured using gamma-ray spectro-metry high purity Germanium (HPGe) detector at transition energy 661.6 keV for ^{137}Cs. The counting time of 259, 200 s was set for sediment and seawater activity and background. The background spectra were used to correct the net peak areas of gamma-rays of measured isotopes.

Result and Discussion

The dominant source of anthropogenic radionuclides such as ^{137}Cs have been release to the environment as a result of global fallout, nuclear weapon test, the Chernobyl accident and disaster of Fukushima Daiichi nuclear power plant in 11 March 2011 (Livingstone & Povinec, 2000; Buesseler et al., 2011). The environmental impacts of the nuclear accidents of Fukushima were estimate a total source term of 520 (340-800) Pbq and the majority of the radionuclides more than 80 % was transported offshore and deposit in the Pacific Ocean (Steinhauser et al., 2014). ^{137}Cs concentration in North Pasific can transport via Indonesian sea, it is well know that surface water from North Pacific through the Indonesian through flow into the Indian Ocean (Wunsch, C., 2010; Povinec et al., 2011). The distribution of ^{137}Cs after Fukushima accident in surface waters of the Pacific Ocean will reach Indian Ocean between 2014 and 2041. The North Pacific and Indian Ocean will be covered with Fukushima ^{137}Cs with concentration below 0.1 Bq/m^3 30 years after the accident (Nakano & Povinec, 2012). In this study the radionuclide ^{137}Cs analysis of seawater and sediment samples collected from various stations in offshore Medan, North Sumatra in June 2015 are shown in table 1. ^{137}Cs radioisotopes were detected in all seawater samples and 2 of 5 sediment sample shown activity below detection limit. The samples were analyzed with gamma-ray spectrometer high purity Germanium (HPGe) detector at transition energy 661.6 keV for ^{137}Cs. This instrument is up to date with its calibration frequency.

In Table 1 were shown the amount of ^{137}Cs concentrations in seawater ranges between 0.191 – 0.757 Bq/m^3 and for sediment ranged from <BDL – 0.19 Bq/kg. The result of this study when compared to previous data from observations in Indian Ocean, South China Sea, and the Pacific Ocean showed different results. Detailed data from the last few years are shown in table 2. The radionuclide ^{137}Cs were measured in the Pacific Ocean in 1996 had average concentration 2.70±0.25 Bq/m^3, the South China Sea in 1997 at 2.70±0.11 Bq/m^3, Bay of Bengal and Andaman Sea at 1.47±0.10 Bq/m^3 and Eastern Indian Ocean at 2.12± 0.19 Bq/m^3 (Yamada et al., 2006). Then the expedition BEAGLE2003 round the globe using research vessel Mirai in 2003 – 2004 by Japan Agency for Marine-Earth Science and Technology (JAMSTEC) were provided data ^{137}Cs at average concentration 1.60±0.30 Bq/m^3 in North Indian Ocean and 2.1±0.3 Bq/m^3 in South Indian Ocean (Povinec et al., 2011). Suseno and Prihatiningsih (2014) were reported concentrations ^{137}Cs in West Sumatra (Indian Ocean) for sediment sample between 0.26–0.35 Bq/kg and seawater at average concentration 0.12± 0.02 Bq/m^3 in sampling date 22 March 2012.

Table 1. The concentrations ^{137}Cs in seawater and sediment samples of marine waters in Medan, North Sumatra**.**

Sample Locations	Coordinates		Stations	^{137}Cs concentration	
	Longitude (E)	Latitude (N)		Sedimen Bq/kg	Seawater Bq/m^3
Medan	99° 05' 27.6"	03° 35' 56.4"	# 1	-	0.191±0.022
	98° 56' 31.2"	03° 40' 33.6"	# 2	-	0.757±0.087
Medan	99^0 05' 27,6"	03^0 35' 56,4"	# 1	< BDL	-
	98^0 56' 31,2"	03^0 40' 33,6"	# 2	0,035 ± 0,003	-
	98^0 57' 21,6"	03^0 40' 44,4"	# 3	0,047 ± 0,005	-
	98^0 59' 16,8"	03^0 39' 32,4"	# 4	< BDL	-
	98^0 58' 0,00"	03^0 40' 50,0"	# 5	0,19 ± 0,02	-

BDL: Below detection limit

Table 2. Average concentrations ^{137}Cs in last few year from Pasific Ocean, Indian Ocean and their adjacent seas.

Locations	Sampling Date	Average ^{137}Cs Concentration	
		Sedimen (Bq/kg)	Seawater (Bq/m^3)
Western North Pacific[a]	December 1996	-	2.70 ± 0.25
South China Sea[a]	February 1997	-	2.70 ± 0.11
Bay of Bengal and Andaman Sea[a]	January 1997	-	1.47 ± 0.10
Eastern Indian Ocean[a]	January 1997	-	2.12 ± 0.19
North Indian Ocean[b]	2004	-	1.60 ± 0.30
South Indian Ocean[b]	2004	-	2.10 ± 0.30
North West Pacific[c]	March 2011		1.30 ± 0.30
West Sumatra[d] (Indian Ocean)	March 2012	0.26 – 0.35	0.12 ± 0.02

[a] Yamada et al., 2006.
[b] Povinec et al., 2011.
[c] Ramzaev et al., 2014.
[d] Suseno & Prihatiningsih, 2014.

Results of the measurement concentration ^{137}Cs in marine waters Medan, North Sumatra were compared data ^{137}Cs from Pacific Ocean and South China Sea shown smaller concentration. Ramzaev, V et al. (2014) were measuring the concentration ^{137}Cs in Pacific Ocean with calculation before accident of Fukushima nuclear reactor at 1.3±0.3 Bq/m^3 in March 2011, then after the accident concentration ^{137}Cs were increased in Pacific Ocean ranged between 1 Bq/m^3 to 34 Bq/m^3. According to Nakano, M & Povinec, P.P (2012) contamination of radiocesium from Fukushima will be dispersed and entered into Indonesian seas at 2014 to 2016 from North Pacific to Southern Indian Ocean through Indonesian throughflow. Medan seas at North Sumatra is located in the Malacca Strait were not directly connected to current flow of water mass from the Pacific Ocean, so the small chance of ^{137}Cs can influence in the region. Therefore ^{137}Cs from Fukushima nuclear reactor accident had no impact on ^{137}Cs contamination in marine waters of Medan.

Concentration ^{137}Cs in seawater on this study were relatively lower if compared with ^{137}Cs from bay of Bengal, Andaman sea, and Indian Ocean. The climate seas of Medan that were located in Malacca strait and directly connected to Andaman sea in Eastern Indian Ocean, strongly influenced by the north-east Monsoon on equatorial current system and the circulation in Northern Indian Ocean which brings rain from December to February, and by the dry Southwest Monsoon from June to August. The weather becomes unpredictable in periods of two inter-monsoon (Chua, T.E et al. 2000; Godfrey et al., 2001; Povinec, P.P et al., 2011). In consequence contamination ^{137}Cs in marine water Medan were influence by deposition from global fallout radionuclides. The fact that region marine of Medan is free from nuclear activities most likely explains the somewhat lower levels found in this study. The observed concentration of ^{137}Cs in sediment samples on table 1, shown the radiocesium in seawater were higher than sediment. This is due to ^{137}Cs after deposition on the ocean surface is affected only by diffusions, advection, and radioctive decay (half-life of ^{137}Cs is 30.15 years) (Lee et al., 2005). Therefore deposition ^{137}Cs in sediment of Medan sea were not significant occur, so it caused concentration ^{137}Cs in sediment lower than seawater. The result from this study can be used to provide of baseline data ^{137}C radionuclides in sediment and surface waters of Medan, North Sumatra so that any further contributions from nuclear activity and possible nuclear accident can be identified.

Conclusions

1. The concentration of radionuclide ^{137}Cs in marine waters of Medan, North Sumatra from each location were station 1 <BDL, station 2, 0.035±0.003 Bq/Kg, station 3, 0.047±0.005 Bq/Kg, station 4, <BDL, station 5, 0.19±0.02 Bq/Kg and result for seawater sample was station 1, 0.191±0.022 Bq/m^3 and station 2, 0.757± 0.087.
2. The data presented here shown ^{137}Cs had lower concentration from Indian Ocean and Pacific Ocean, which is consequence of the Medan seas were located in the Malacca Strait not directly connected to current flow from Pacific Ocean. Therefore ^{137}Cs from accident Fukushima nuclear reactor had no impact on ^{137}Cs contamination in marine waters of Medan.
3. The concentration of radionuclides ^{137}Cs in Medan seas were ranged from 0.191–0.757 Bq/m^3 for seawater and sediment samples in

ranged <BDL – 0.19 Bq/kg. Because North Sumatra was free from nuclear activity so the radionuclides ^{137}Cs contaminant on this area were mostly derived from the global fallout, therefore result in this study can be used for baseline data of ^{137}Cs.

Acknowledgements

The author was grateful to Marine Radioecology Group Dr. Heny Suseno, Dr. Murdahayu Makmur and Mohamad Nur Yahya for knowledge n technical support. This work was supported by Center for Radiation Safety Technology and Metrology, National Nuclear Energy Agency (BATAN).

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