Omni-Akuatika, 13 (2): 180 – 189, 2017
ISSN: 1858-3873 print / 2476-9347 online

Research Article

Marine Radioecology in Bali and Lombok Waters: Present Status of ^{137}Cs and Natural Radionuclides in Lombok Strait

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Received 4 May 2017; Accepted 10 October 2017; Available online 28 November 2017

ABSTRACT

The accident of Fukushima Dai-Ichi nuclear power plant in 2011, were released a large amount of ^{137}Cs into terrestrial and marine environment in Pasific Ocean. ^{137}Cs has a relatively long half-life and easily soluble in sea water can spread out by ocean currents. ^{137}Cs can entered to Indonesian waters from Pacific Ocean by Indonesian Through flow (ITF). This study aims to obtain baseline data of artificial radionuclide ^{137}Cs in Bali and Lombok Straits. The concentration of natural radionuclides was also measured to determine the dose rate and its impact to the ecosystem using Erica Tools. The sampling location were covered territorial waters of Bali and Lombok. The radionuclide samples were analyzed by co-precipitation process using $K_4Fe(CN)_6$ and $CuCl_2$ at pH 8-9. Their concentration activities of natural radionuclides and ^{137}Cs were measured using Gamma HPGe spectrometer with 20 – 25 % efficiency. This study indicate the average concentration of ^{40}K ^{226}Ra ^{212}Pb ^{214}Bi ^{228}Ac ^{214}Pb and ^{137}Cs in the sediments of Strait Bali waters 165.16 Bq kg^{-1}, 25.11 Bq kg^{-1}, 8.36 Bq kg^{-1}, 7.51 Bq kg^{-1}, 7.05 Bq kg^{-1}, 6.68 Bq kg^{-1} and the lowest of 0.15 Bq kg^{-1} respectively. In the waters of Lombok concentration activity of radionuclides ^{40}K ^{226}Ra ^{212}Pb ^{214}Bi ^{228}Ac ^{214}Pb and ^{137}Cs in the sediments was 172.00 Bq kg^{-1}, 25.88 Bq kg^{-1}, 10.10 Bq kg^{-1}, 6.50 Bq kg^{-1}, 6.39 Bq kg^{-1}, 5.70 Bq kg^{-1} and 0,162 Bq kg^{-1}. The concentration activity of ^{137}Cs in seawater of Bali and Lombok was 0.113 Bq m^{-3} and 0.644 Bq m^{-3} respectively.

Keywords : *Natural Radionuclide, ^{137}Cs, Lombok Strait*

1. Introduction

Lombok Strait located in coordinates of 115 ° 37 'E - 116 ° 02' E and 8 ° 20 'S - 8 ° 50' S represents the marine waters between the islands of Bali and Lombok and connects western seas of Flores and the Indian Ocean. The Lombok Strait was route of water mass from Pacific Ocean into Indian Ocean via Indonesian Throughflow (ITF), which is influenced by differences of sea level, temperature and salinity (Brink and Robinson, 2005). The water mass from the Pacific Ocean enters to Indonesian Seas through two entrances i.e west channel and east channel. The western channel which is the main inflow water from the Pacific Ocean transports 11.8 Sv through the Makassar Strait and then exits the Lombok Strait and partially into the Banda Sea through the Flores Sea and Timor Sea into the Indian Ocean. The eastern channel of water mass enters through Halmahera and Maluku Seas and pass through the strait of Ombai and Timor Passage to Indian Ocean (Gordon et al, 2003, Gordon et al., 2010). In addition the large quantities of this water mass from ITF also contains ^{137}Cs radionuclide caused by nuclear activity in the Pacific Ocean.

The main source of contamination ^{137}Cs radionuclide in marine waters comes from nuclear weapons tests proceeding within the period of 1945 - 1980 with a total release of 189 megatons from 543 nuclear weapons and mostly testing in northern hemisphere. The accident of Fukushima Nuclear Power Plant in March 2011 also give a contribution impact on radionuclide contamination in Pacific Ocean with an estimated released of ^{137}Cs about 12 to 41 PBq into the atmosphere (UNSCEAR, 2000; Hu et al. ; Baily du Bois et al., 2012).

Until now data of radionuclides in Lombok Strait is limited, so it is difficult to estimate the radiological impacts in these marine waters. Previous research in monitoring radionuclide concentration has been conducted on some coastal areas of Indonesian Seas, the southern of Java Sea (Indian Ocean) and the eastern sea of Indonesia (Suseno and Prihatiningsih, 2014; Suseno et al., 2017). Furthermore monitoring of ^{137}Cs on previous research was only conducted on surface seawater. On the other hand, ITF from Pacific

Ocean is the upper thermocline sea flow in the depth of 50 - 200 m. The upwelling in Lombok Strait annualy was caused by the southeast monsoon cycle, with the surface temperature being colder than that during the northwest monsoon. This condition takes place to compensate for the divergence of the horizontal flow and produces upward flow, carrying cold and nutrient-rich deep waters to the surface. This upwelling phenomenon brings many nutrients from the seafloor including radioactive soluble metals such as ^{137}Cs, ^{226}Ra, $^{212/214}$Pb, ^{228}Ac, ^{214}Bi and ^{40}K (Krishnaswami and Cochran, 2016). This paper reports the concentrations of the Lombok strait natural and artificial radionuclides ^{137}Cs that was carried by ocean currents from the Pacific Oceans resulting from the global fallout and the Fukushima Dai-ichi nuclear accident in 2011.

2. Material and methods

Sampling sites were conducted surrounding the islands of Bali and Lombok in August 2016. The sampling sites were shown in Figure 1 and Figure 2.

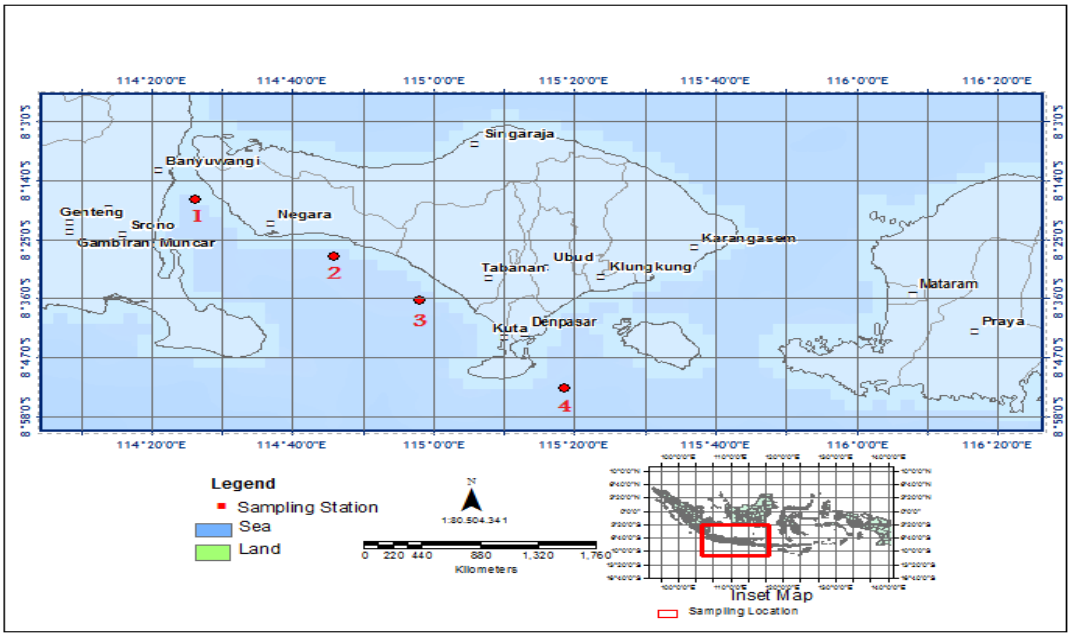

Figure 1. Sampling location in Bali sea waters

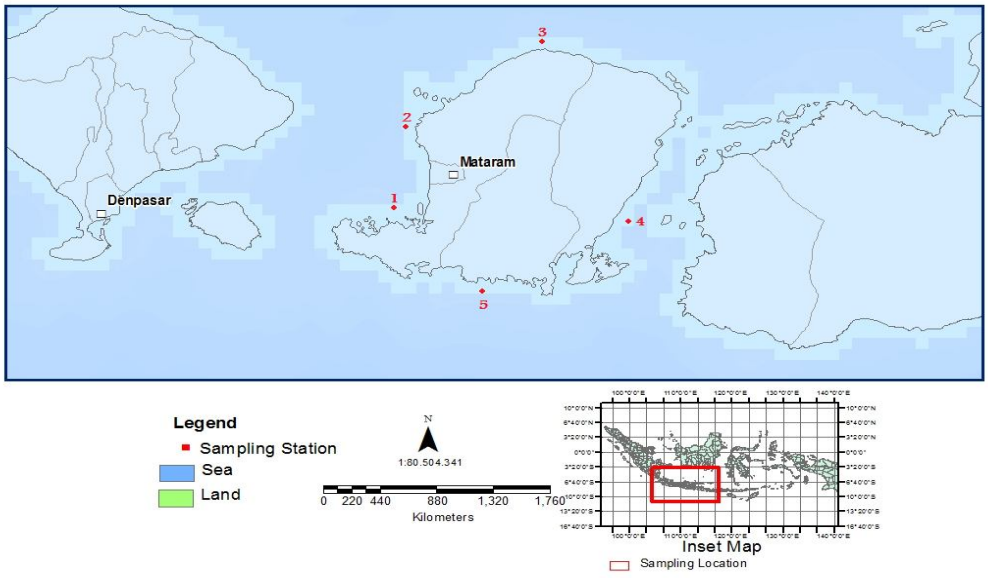

Figure 2. Sampling location in Lombok sea waters

Each location 150 L of sea water sample was collected at a depth of 0 - 5 m using water pump and sediments from the seabed using sediment grab of approximately 2 kg. Pre-concentration of all sea waters was conducted in the field near shore using PVC containers. To each sea water sample was adding by 10 grams of potassium ferrocyanic(II) ($K_4Fe(CN)_6$) and 10 gram copper chloride ($CuCl_2$) subsequently, then stirred until homogeneous and allowed to stand up to 24 hours for the precipitation process. The precipitate on bottom container was separated from the supernatant by a 0.5 micron filter paper (Wo and Ahmad, 2004). The precipitate was then analyzed to the radiochemistry laboratory for further processing and analysis.

Sediment samples and precipitate from previous sampling in the field were dried in oven at temperature 120 °C for 2 - 4 days to dry. The sediment samples were then smoothed by grinder and sieved using a 63 µm sieve. Dry precipitate and sediments were fed to 1 kg marinelli tube for measurement using a gamma spectrometer. Each sample was measured during 259,200 - 345,600 seconds with an HPGe detector which has a 20 - 25% efficiency.

The radioactivity concentration of ^{137}Cs and ^{40}K was calculated as follows (IAEA, International Atomic Energy Agency, 1989):

$$C_r\ (Bq\ kg^{-1}) = A/(\varepsilon \times T \times \gamma \times W)$$

Where C_r is the activity concentration of the radionuclide (^{40}K ^{226}Ra ^{212}Pb ^{214}Bi ^{228}Ac ^{214}Pb ^{137}Cs) in seawater and sediment (Bq kg^{-1}), A is the count of the radionuclide (counts), ε is the detector efficiency of the specific gamma-ray, γ is the absolute transition probability of the specific gamma-ray, T is the time (second; s) and W is dry weight of sediment (kilogram; kg). Minimum Detectable Activity (MDA) is commonly used to evaluate the measurement sensitivity and limit detection concentration for radiocesium is 0.03 Bq m^{-3}. The MDA calculation is derived from the following formula

$$\mathrm{MDA} = \frac{2.71 + 4.65\sqrt{B}}{tmEY}$$

Where *B* is background count at the peak energy of interest, *t* is the counting time (s), m is the sample mass, *E* is the detector efficiency at the energy and geometry of interest and Y is the effective yield at the energy.

Radiological risk assessment in the marine environment of the monitoring area was evaluate using Erica Assessment Tool (version 1.2 April 2017). Isotope ^{137}Cs and a dose screening value of 10 µGyH^{-1} were selected in the parameter Tier 1 and Tier 2 of assessment. The highest concentration of ^{137}Cs in the seawater monitoring results were input in the assessment as the environment concentration and the parameters of distribution coefficient (Kd), concentration ratio (CR), dose conversion coefficient of radiation, occupancy factors, uncertainty factor were set as the default value in ERICA Tool. The weighing factors of internal low beta, internal beta/gamma and internal alpha were set as 3.1 and 20 (Yu et al., 2015).

3. Results and Discussion

^{137}Cs and ^{134}Cs were conservative radionuclides that are very soluble in sea water, so their distribution in the ocean is strongly influenced by physical process of mixing and diffusion. Radionuclides ^{137}Cs and ^{134}Cs can be involved in sea water and transported to various areas within a considerable distance. The ^{137}Cs radioactive elements in the northern Pacific Ocean were mostly derived from nuclear activities occurring in the 1950s and 1960s such as nuclear weapons testing, radioactive waste and operations of nuclear reactor (Livingstone and Povinec, 2000). In March, 2011 the Fukushima Dai-ichi nuclear power plant were crashed and released radioactive ^{137}Cs into the atmosphere with an estimated total concentration activity of 12 - 41 PBq. Contaminants of ^{137}Cs were dispersed into the Pacific Ocean and contaminated most of Japan area including Tohoku and northern areas of Kanto (Chino et al., 2011; Steinhauser et al., 2014).

Radionuclides were released from the Fukushima nuclear accident and entered to the waters of north Pacific Ocean and through the Kuroshio stream and were transported to South Pacific Ocean (Kaeriyama, 2016). Contaminant of ^{137}Cs radionuclide in Pacific Ocean can spread out and enter to Indonesian seas by through Kuroshio current from Japan. This current flows from the northern Philippines along the southeastern sea of Japan to the north of American coast, then leading to southward and enters to northern equatorial currents to south of Philippines. This Kuroshio stream then seperated into 2 direction, one direction leading to Indonesia seas through south Mindanao and the other towards to Japanese stream of Kuroshio in north Philippines (Sprintall., 2009). ^{137}Cs with a half-life over 30.07 years has a potential to spread out in ocean currents from Japanese waters into Indonesian waters through Indonesian Throughflow (ITF).

Table 1. Concentration of ^{137}Cs in the water and sediment of the Bali and Lombok waters

Location	Station	Concentration ^{137}Cs	
		Sea water (Bq m^{-3})	Sediment (Bq kg^{-1})
Bali	1	0.11 ± 0.02	0.18 ± 0.02
	2	0.09 ± 0.01	-
	3	-	0.16 ± 0.03
	4	0.14 ± 0.03	0.12 ± 0.02
Lombok	1	0.41 ± 0.05	0.27 ± 0.007
	2	1.16 ± 0.10	0.187 ± 0.02
	3	1.13 ± 0.11	0.16 ± 0.02
	4	0.05 ± 0.007	0.082 ± 0.01
	5	0.47 ± 0.05	0.11 ± 0.02

Tabel 2. Concentration of ^{137}Cs dan ^{134}Cs from previous research in Indonesia and Pacific Ocean.

Sampling year	Location	Concentration ^{137}Cs		Concentration ^{134}Cs	Reff.
		Sediment (Bq kg^{-1})	Sea water (Bq m^{-3})	Sea water (Bq m^{-3})	
September 2012	Manado, North Sulawesi	0.10 – 0.23	0.12 – 0.31	-	Suseno and Prihatiningsih (2014)
March 2012	Makassar, South Sulawesi	0.58 – 0.59	0.18 – 0.27	-	
June 2013	Jogjakarta, Indian Ocean	0.63 – 1.05	0.12 – 0.14	-	
September 2012	South Java Sea	-	<MDA – 0.30	<MDA	Suseno et al. (2015)
November 2011	West Sumatra	-	<MDA – 0.13	<MDA	
September 2012	Flores Sea	-	0.29 – 0.30	<MDA	Suseno et al. (2017)
	Halmahera Sea	-	0.20 – 0.38	<MDA	
	Lifamatola Passage	-	0.20 – 0.30	<MDA	
August 2016	Bali	0.12 – 0.18	0.09 – 0.14	<MDA	This research
	Lombok	0.082 – 0.27	0.05 – 1.16	<MDA	
2011 - 2012	Western North Pacific and Japan Sea	-	1.00 – 34.0	0.2 – 29.0	Ramzaev et al. (2014)

MDA ^{137}Cs = 0,01 Bq m^{-3}, MDA ^{134}Cs = 0,02 Bq m^{-3}

The current that pass through Mindanao enters to Indonesia waters via western Indonesian Throughflow to the west of Sulawesi leading to Makassar Strait with about 15 million m^3 (15 Sv) volume of sea water. This stream then exit to the Indian Ocean via Lombok Strait current with a volume of 20-25% of the total water mass Indonesian Throughflow. The remaining 75 to 80 % turns towards to Banda Sea and exit through the Ombai and Timor passage (Susanto and Gordon, 2005). The concentration of ^{137}Cs activities in the waters of Bali and Lombok can be seen in Table 1.

The ^{137}Cs concentration for sediment samples in Bali waters ranged from 0.12 to 0.18 Bq kg^{-1} with an average of 0.153 Bq kg^{-1}. The concentration of ^{137}Cs in seawater were ranged from 0.09 - 0.14 Bq m^{-3} with an average of 0.113 Bq m^{-3} in Lombok water. The concentration of ^{137}Cs ranged from 0.05 to 1.16 Bq m^{-3} with average of 0.64 Bq m^{-3} in seawater and ranged from 0.082 - 0.27 Bq kg^{-1} with an average of 0.16 Bq kg^{-1} in sediment. Detection limit on this measurement were 0.03 Bq m^{-3} for radioisotopes ^{137}Cs and ^{134}Cs. Cesium radionuclide from the Fukushima Dai-Ichi nuclear reactor accident possibly entering to Indonesian waters can be determined by measuring of ^{134}Cs isotope. One of the ^{235}U fission products with a half-life of 2,065 years (Povinec et al., 2013).

^{137}Cs were predicted entering to Indonesian waters in 2016 based on modeling results by Povinec et. al. (2013). In this study the ^{134}Cs in seawater and sediment samples were undetectable or below the detection limit indicating the ^{137}Cs and ^{134}Cs of radionuclides originating from the Fukushima nuclear reactor accident did not impact on Indonesian waters.

The ^{137}Cs concentrations in water and sediments samples in Lombok strait were possibly caused by the global fallout from nuclear activity in 1950s until 1960s in northern hemisphere. ^{137}Cs with a long half life time were still left in the environment with less than 50 % from that of original sources. Table. 2 showed data of ^{137}Cs from previous research in Indonesia and some places in the world. Comparing with those previous research it can be seen that concentration of ^{137}Cs in the waters of Bali and Lombok has not much different value from those of other regions in Indonesia. This indicates that ^{137}Cs contamination has insignificantly changed and only possibly influenced by sea water flows.

The concentrations radionuclides ^{226}Ra, ^{212}Pb, ^{214}Pb, ^{214}Bi, ^{228}Ac, ^{40}K and ^{137}Cs in sediments of Bali and Lombok straits were shown in Table 3. and Figure 3. Concentration for ^{40}K ^{226}Ra ^{212}Pb ^{214}Pb ^{214}Bi ^{228}Ac and ^{137}Cs in Bali waters respectively ranged 145.89 - 183.45 Bq kg^{-1}; 21.06 - 31.13 Bq kg^{-1}; 6.12 - 10.46 Bq kg^{-1}; 5.95 - 7.10 Bq kg^{-1}; 6.34 - 9.45 Bq kg^{-1}; 6.19 - 7.86 Bq kg^{-1} and 0.12 - 0.18 Bq kg^{-1}. The average concentration of those radionuclides was 165.16 Bq kg^{-1}, 25.11 Bq kg^{-1}, 8.36 Bq kg^{-1}, 7.51 Bq kg^{-1}, 7.05 Bq kg^{-1}, 6.68 Bq kg^{-1} and 0.15 Bq kg^{-1}. Concentration ^{40}K ^{226}Ra ^{212}Pb ^{214}Pb ^{214}Bi ^{228}Ac and ^{137}Cs in sediment Lombok respectively ranged from 150.31 - 189.94 Bq kg^{-1}; 18.17 - 32.52 Bq kg^{-1}; 7.42 - 12.81 Bq kg^{-1}; 4.01 - 7.53 Bq kg^{-1}; 4.74 - 8.92 Bq kg^{-1}; 4.23 - 9.22 Bq kg^{-1} and 0.082 - 0.27 Bq kg^{-1}. The average of those radionuclide was 172.00 Bq kg^{-1}, 25.88 Bq kg^{-1}, 10,10 Bq kg^{-1}, 6.50 Bq kg^{-1}, 6.39 Bq kg^{-1}, 5.70 Bq kg^{-1} and 0.162 Bq kg^{-1}.

Table 3. Concentration of Natural radionuclides and ^{137}Cs in sediment sample of Bali and Lombok waters

Location	Station	Analysis results (Bq kg^{-1})						
		^{226}Ra	^{212}Pb	^{214}Pb	^{214}Bi	^{137}Cs	^{228}Ac	^{40}K
Bali	1	23.13 ± 3.87	8.51 ± 0.91	7.10 ± 0.80	6.73 ± 0.83	0.18 ± 0.02	7.86 ± 0.09	145.89 ± 16.98
	2	21.06 ± 2.31	10.46 ± 1.89	6.99 ± 0.79	9.45 ± 1.03	0.16 ± 0.03	7.11 ± 0.08	166.13 ± 17.32
	4	31.13 ± 4.01	6.12 ± 0.73	5.95 ± 0.68	6.34 ± 0.77	0.12 ± 0.02	6.19 ± 0.77	183.45 ± 19.42
Lombok	1	30.88 ± 1.35	7.42 ± 0.33	5.17 ± 0.20	5.06 ± 0.20	0.27 ± 0.007	5.35 ± 0.36	189.94 ± 3.28
	2	32.52 ± 4.12	10.62 ± 0.23	4.86 ± 0.05	6.87 ± 0.71	0.187 ± 0.02	5.92 ± 0.61	150.31 ± 10.13
	3	25.32 ± 3.51	12.81 ± 0.27	4.01 ± 0.05	6.91 ± 0.72	0.16 ± 0.02	7.23 ± 0.76	182.32 ± 11.81
	4	18.17 ± 2.49	10.24 ± 0.20	6.91 ± 0.07	4.74 ± 0.53	0.082 ± 0.01	4.23 ± 0.50	166.29 ± 14.07
	5	22.52 ± 2.98	9.41 ± 0.19	7.53 ± 0.08	8.92 ± 0.91	0.11 ± 0.02	9.22 ± 0.91	171.12 ± 15.34

Natural radionuclides in an environment come from two main sources: i.e primordial radionuclides existed since the beginning of the earth with long half life and those formed naturally by the interaction of cosmic rays with materials in atmosphere and the earth's crust. The primordial radionuclides are derivatives from ^{238}U, ^{235}U, ^{232}Th, ^{40}K, ^{87}Rb and ^{187}Re, while those cosmogenic radioisotopes were i.e ^{3}H, ^{14}C and ^{10}Be (UNSCEAR, 1982). Distribution of all these radionuclides in ocean waters was determined by their origin, half-life, water circulation, and reactivity of biogeochemistry in seawater. If the biogeochemical processing in sea water was passive, water circulation becomes the most dominant factor to spread out of radioisotopes in ocean. Distribution of natural radionuclides and ^{137}Cs found in sediments of Bali and Lombok can be seen in Figure 3. and Figure 4.

The total average concentration of natural radionuclides and ^{137}Cs in sediments of Bali and Lombok islands can be seen in Figure 5. Content of ^{40}K in sediments about 75.57% was highest level when compared with other elements. Highest value of ^{40}K was influenced by mineral content of potassium salts dissolved in seawater and the sediments. The average activity concentration of radionuclide ^{40}K ^{226}Ra ^{212}Pb ^{214}Bi ^{228}Ac ^{214}Pb and ^{137}Cs from high to low was 169.43 Bq kg^{-1}, 25.59 Bq kg^{-1}, 9.45 Bq kg^{-1}, 6.88 Bq kg^{-1}, 6.64 Bq kg^{-1}, 6.07 Bq kg^{-1}, and lowest 0.16 Bq kg^{-1}.

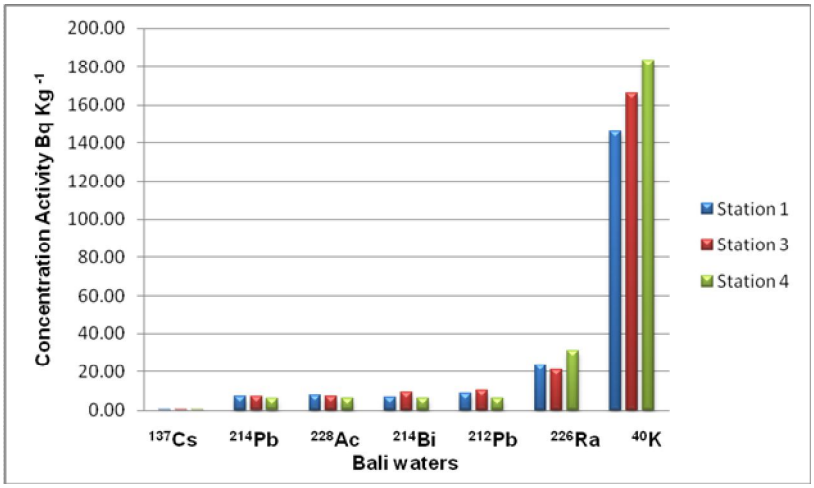

Figure 3. Concentration activity radionuclides in the sediments of Bali waters

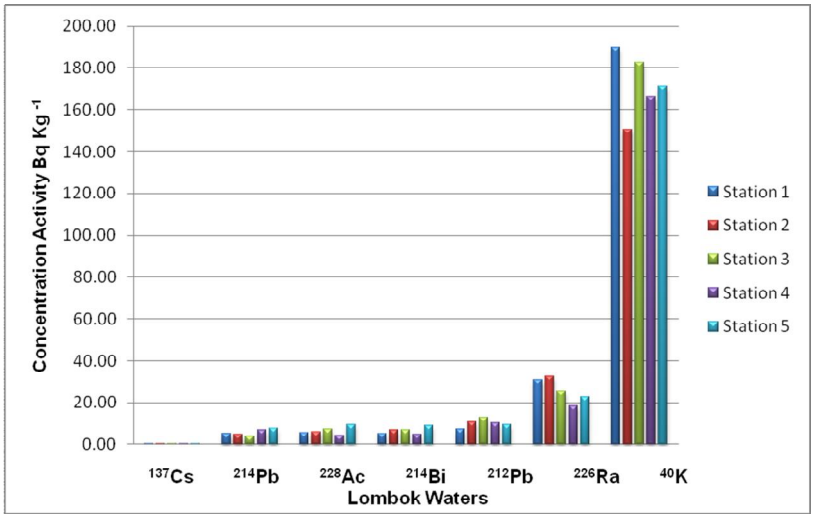

Figure 4**.** Concentration activity radionuclides in the sediments of Lombok waters

Comparison results of this study with other regions in the world can be seen in Table 4. ^{40}K from this study were still below the average of other regions. Based on data from UNSCEAR 2000, the upper limit of radionuclide content in environment was 400 Bq kg^{-1} for ^{40}K and 35 Bq kg^{-1} for ^{226}Ra. This indicates that concentrations of radionuclides in this study were still significantly below the threshold.

Impact of radionuclides ^{137}Cs on marine ecosystems in this study were estimated and analyzed using Erica Tool software version 1.2.1 update 12 February 2016. The highest concentrations of ^{137}Cs in seawater and sediment Bali and Lombok were used as radionuclide parameters with dose rate limits of 10 µGy h^{-1}. In Table 5. were shown results and analysis from Erica Tool that indicate of internal dose rate, external dose rate, and total dose rate in some types of marine water organisms. These results indicating that ^{137}Cs contents do not significantly affect on marine biota because the dose rate were still below from the established dose rate of 10 µGy h^{-1}. Nevertheless, potential of danger from contamination ^{137}Cs in marine biota still occur through the chain food and continue to accumulate in biota which is will eventually to be consumed by humans.

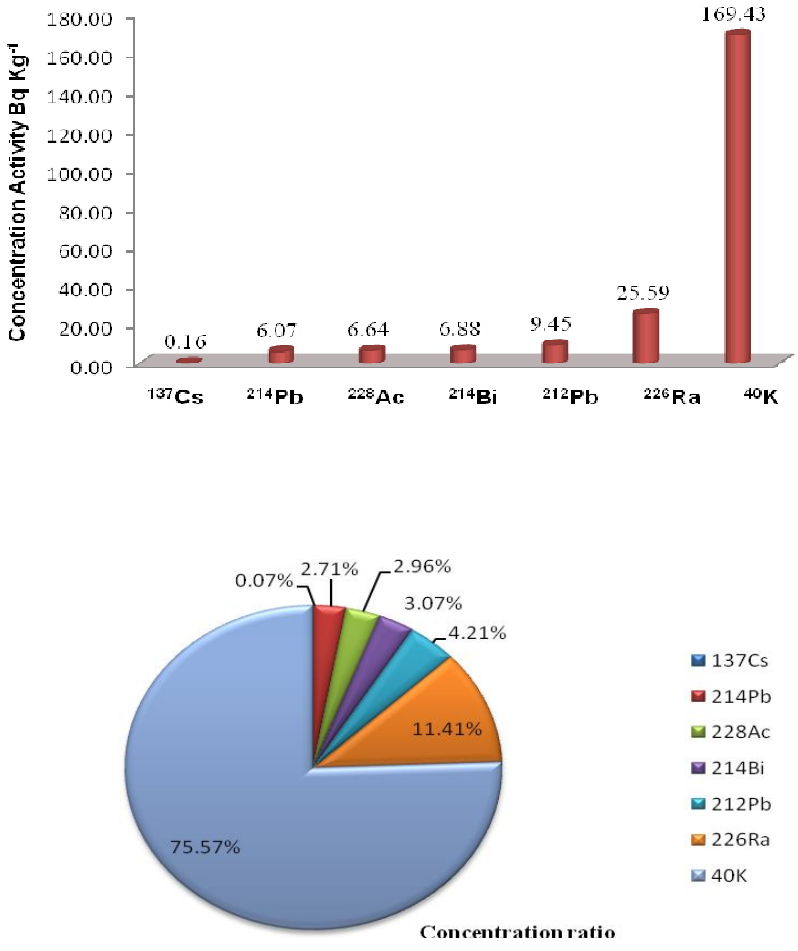

Figure 5. Average and concentration ratio of natural radionuclides and ^{137}Cs in sediment samples of Bali and Lombok waters.

Table 4. Data of natural radionuclide concentration and ^{137}Cs in sediment from other regions in the world.

Location	Concentration (Bq kg^{-1})			Reff.
	^{40}K	^{226}Ra	^{137}Cs	
Yangtze Estuary, China	628	24,3	-	Wang, J. et al., 2017
Guanghai Bay, China	571	36,6	-	Zhao et al., 2015
Pakistan	532	50,7	-	Qureshi et al., 2014
India	782	34	-	Tripathi et al., 2013
Thailand	1145	22,3		Kessaratikoon et al., 2013
Malaysia	189	51	-	Muhammad et al., 2012
Bali and Lombok	169.43	25.59	0.16	This research
World recomended	400	35	-	UNSCEAR, 2000

Table 5. Results of total dose rate ^{137}Cs using Erica Tool tier 2

Biota	**External dose rate ^{137}Cs [µGy h^{-1}]**	**Internal dose rate ^{137}Cs [µGy h^{-1}]**	**Total dose rate ^{137}Cs [µGy h^{-1}] per organism**
Crustacean	3.93E–5	1.11E–5	5.04E–5
Macroalgae	4.47E–5	1.56E–6	6.03E–5
Mollusc - bivalve	4.34E–5	8.70E–6	5.21E–5
Pelagic fish	3.36E–7	1.75E–5	1.79E–5
Phytoplankton	4.67E–7	6.61E–7	1.13E–6
Zooplankton	4.06E–7	1.81E–5	1.85E–5
Sea anemones & True Coral	4.47E–5	3.74E–5	8.21E–5
Polychaete worm	8.91E–5	2.92E–5	1.18E–4
Benthic fish	4.07E–5	1.66E–5	5.72E–5
Bird	3.25E–7	1.06E–4	1.06E–4
Mammals	1.62E–7	8.42E–5	8.44E–5
Vascular plant	4.34E–5	1.74E–6	4.51E–5

4. Conclusion

The monitoring results from the analysis showed that activities of ^{137}Cs were found in the surface seawater of the Lombok strait not much different from previous research with average concentration in seawater 0.38 Bq m^{-3} and 0.16 Bq kg^{-1} for sediment.^{134}Cs concentration in seawater and sediment were not detected or below from the detection limit. This result indicate that waters of strait Lombok were not contaminated from accident Fukushima Dai-ichi nuclear power plant. According to the radiological assessment result of ERICA Tool, the dose rates for marine organisms in the sampling area were lower than the screening dose rate of 10 µGy H^{-1}, indicating that the radioisotope ^{137}Cs would not have a significant adverse effect on marine biota at the population level.

Acknowledgment

The authors would like to thank Marine Radioecology team for technical support. This research were supported and funded by the Center for Technology Radiation Safety and Metrology, National Nuclear Energy Agency (BATAN).

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