

Concentration of Selected Radionuclides in Sediment and Surface Seawater in Belitung Island, Indonesia

Murdahayu Makmur, Mohamad Nur Yahya and Deddy Irawan Permana Putra

Center for Technology of Radiation Safety and Metrology, National Nuclear Energy Agency of Indonesia
Jl. Lebak Bulus Raya No. 49 Jakarta Selatan, 12440 Indonesia, E-mail: mdhayu@batan.go.id

Abstract. Global fallout radionuclides were injected into the oceans mainly in the late 1950s and early 1960s, after large-scale atmospheric nuclear weapons tests were carried out. The highest deposition occurred in the mid-latitudes of the western North Pacific Ocean and Western North Atlantic Ocean. The global sea water exchange between the different oceans plays an important role in the movement mass, heat and other substances include anthropogenic matters. As a part of the Indonesian maritime continent, Karimata Strait where the Belitung Islands lied, plays an important role as a transfer matters of the oceanic conjunction between Pacific and Indian Ocean. No baseline existed for the radionuclides in Belitung Island, where the baseline study is imperative to create a reference and to record the influence functioning of upcoming power plants. The present baseline concentration of the considered radionuclide's show low concentration of cesium and plutonium in sediment and surface seawater and their concentration is comparable to most oceanic waters.

Keyword: Plutonium, cesium, baseline data, Belitung Island, Karimata Strait

Introduction

Plutonium and cesium in the Pacific Ocean has originated mostly from global fallout from large atmospheric nuclear weapons tests, from which the major oceanic input occurred in the early 1960s. There are two principal sources of ^{137}Cs , ^{239}Pu and ^{240}Pu , that is global stratospheric fallout and close-in fall out from the North Pacific Proving Grounds (NPPG) in the Marshall Islands. (Hirose et al., 2006), (Yamada et al., 2006). Besides that, plutonium isotopes normally produced in reactor fuel as a mixture of isotopes. The predominant isotope, ^{239}Pu is produced by neutron capture in ^{238}U . If a fuel element containing ^{239}Pu is left in a reactor for any length of time further neutron capture can occur to yield higher isotopes like ^{240}Pu , ^{241}Pu and ^{242}Pu . In addition, small quantities of two other isotopes, ^{236}Pu and ^{238}Pu , are produced during the irradiation (Zheng & Aono, 2012).

Plutonium in the ocean is transported by physical and bio geochemical processes. The residence time of plutonium in surface waters of the open ocean ranges from 6 to 21 years, and is generally shorter than that of ^{137}Cs . In contrast to ^{137}Cs , plutonium is a typical particle-reactive radionuclide; plutonium in particulate matter of surface waters occupies from 1% to 10%, whereas particulate ^{137}Cs is less than 0.1% of the total. Plutonium vertically moves with sinking biogenic particles and regenerates into soluble forms in deep waters as a result of microbial decomposition of particles (Hirose et al., 2006).

Average ^{137}Cs activities, $^{239,240}\text{Pu}$ activities in the surface waters of the western North Pacific are $2.70 \pm 0.25 \text{ Bq/m}^3$ and $1.65 \pm 0.12 \text{ Bq/m}^3$ (Yamada et al., 2006). ^{137}Cs levels in surface seawater have been declining to its present median value of about 3 Bq/m^3 . $^{239,240}\text{Pu}$ levels in surface seawater are much lower, with a median of about 6 mBq/m^3 . $^{239,240}\text{Pu}$

appears to be partly scavenged by particles and is therefore more readily transported down the water column. As with seawater, $^{239,240}\text{Pu}$ concentrations are lower than ^{137}Cs in surface sediment. The median ^{137}Cs concentration in surface sediment is 1.4 Bq/kg dry , while that of $^{239,240}\text{Pu}$ is only 0.2 Bq/kg dry (Duran et al., 2004).

The Indonesian Through Flow (ITF) is the connection between two oceanic basins in the tropical region, Pacific Ocean and Indian Ocean. There are three Indonesian Through Flow (ITF) branches: the flow through the Makassar Strait; through the South China Sea; and through the eastern Indonesian basins (Du & Qu, 2010). In the South China Sea, the water exchanged between the Pacific Ocean and South China Sea within the depth range of the thermocline, later observations revealed that there is a Kuroshio branch toward the South China Sea both winter and summer and the intrusion of the North Pacific Tropical Water seems to occur during most. The South China Sea connects in the south with the Sulu and Java Seas through the Shallow Mindoro ($\sim 200 \text{ m}$) and Karimata (50 m) straits and in the north with East China Sea through the shallow Taiwan Strait ($< 100 \text{ m}$) and with the Pacific Ocean through the deep Luzon Strait ($> 2000 \text{ m}$) (Qu et al., 2009).

The ITF also distributed the radionuclides from Pacific Ocean to the Indian Ocean. The result study of The Pisces Austrinus Expedition of R/V Hakuho-Maru was carried out in the western North Pacific Ocean, eastern Indian Ocean and their adjacent seas in 1996-1997 summarize that average ^{137}Cs activities and $^{239,240}\text{Pu}$ activities in the surface waters of the western North Pacific are $2.70 \pm 0.25 \text{ Bq/m}^3$ and $1.65 \pm 0.12 \text{ Bq/m}^3$, in the Sulu and Indonesian seas are eastern Indian Ocean are $2.42 \pm 0.12 \text{ Bq/m}^3$ and $3.16 \pm 0.32 \text{ Bq/m}^3$. Otherwise, in the South China Sea are $2.70 \pm 0.11 \text{ Bq/m}^3$ and $2.52 \pm 0.19 \text{ Bq/m}^3$. (Yamada et al., 2006).

The work reported here will present the ^{239,240}Pu and ¹³⁷Cs activity at Belitung Sea in Sediment and surface seawater that lied in the Karimata Strait in connection with South China Sea, as a baseline data before the first Indonesia NPP built in Bangka Belitung Province.

Material and Methods

Sediment and Sea Water Sample Collection *Sampling Location*

The locations of the sampling sites are shown in Fig. 1. Belitung Island, located in Bangka Belitung Province, is an island lied in the Karimata Strait. The Belitung sea is well suited to study and characterize the Cesium and Plutonium, can be representative the anthropogenic input from South China sea through the Secondary ITF (Indonesian Through Flow).

Sediment Sample Collection

Sediment sample was collected around Belitung Island in February 2015, using the sediment corer. Surface water sample (80 l) were collected around the Belitung Sea and pre-concentration processes are required for analysis of anthropogenic radionuclides in seawater due to extremely low concentration.

Preconcentration of cesium from seawater and sediment

All sediment sample were oven dried, then 1 kg placed in marinally to direct count by gamma spectrometer. Preconcentration for cesium, using copper nitrate salt and potassium hexacyanoferrates precipitation, before separated from seawater, and dried before placing in the PVC container. The cesium preconcentration was conducted according procedure described by Wo and Ahmad study. (Wo & Ahmad, 2004).



Fig. 1. Map showing the sediment and surface sea water sampling location in the Belitung Sea, Bangka Belitung Province, Indonesia. The coordinate of the sampling point could be seen in the Table 1.

Tabel 1. Coordinate of sampling point in the coast of Belitung Island

Latitude	Longitude	¹³⁷ Cs Point Sampling		^{239,240} Pu Point Sampling	
		Seawater	Sea Sediment	Seawater	Sea Sediment
02 42 35.7	107 35 38.3	Water Cs 1	Sediment Cs 1	Water Pu 1	Sediment Pu 1
02 33 37.4	107 39 27.5	Water Cs 2	Sediment Cs 2	Water Pu 2	Sediment Pu 2
02 33 39.1	107 41 58.1	Water Cs 3	Sediment Cs 3	Water Pu 3	Sediment Pu 3
03 09 13.5	107 37 33.8		Sediment Cs 4		Sediment Pu 4
02 51 38.1	108 17 47.8		Sediment Cs 5		Sediment Pu 5
02 53 04.0	108 16 54.9		Sediment Cs 6		Sediment Pu 6

Preparation plutonium from sea seawater and sediment

For plutonium, a few gram were heated in a furnace at 550°C for 6 hours to decompose the organic matter sediment samples were digested using micro-wave digestion (Milestone Start D). After digestion, the liquid sample would go to Plutonium separation by anion exchange and column chromatography section. The preconcentration from 80 l seawater using permanganate and manganese chlorida described by levy (Levy et al., 2011).

Measurement cesium with gamma spectrometer and plutonium with alpha spectrometer

Cesium (¹³⁷Cs) was measured using gamma spectrometer Canberra type GX 2018, Canberra type GC 2020 and Ortec type GMX 25P4-76, completed with HPGe Detector with counting efficiencies of 20–25% (relative to NaI(Tl) detector). The counting time for each sample was 172.800-259.200s. Plutonium was counted by alpha analyst consisted of 8 vacuum chambers with solid-state detectors, a pulse height analyzer and a computer system with counting efficiency of 18-23%). The contents of ^{239,240}Pu were deduced from the countrates of plutonium isotopes, including a yield tracer of ²⁴²Pu, as measured by alpha spectrometry. Because the energies of alpha rays from ²³⁹Pu and ²⁴⁰Pu are too close to be distinguished from one another, we determined the sum of activities ^{239,240}Pu. (Kinoshita et al., 2011).

Result and Discussion

Surface Sea Water

The activity concentrations of cesium (¹³⁷Cs) and plutonium (^{239,240}Pu) radionuclides, in surface water Belitung Sea are presented in Figure 2. Activity ¹³⁷Cs and ^{239,240}Pu in the surface seawater in Belitung Sea were 9.5E-02±7.16E-3 Bq/m³ and 3.01E-3±8.1E-4Bq/m³ respectively. In the figure 2 could be seen, that the activity ¹³⁷Cs higher than ^{239,240}Pu. Generally, the activity ¹³⁷Cs in seawater higher than ^{239,240}Pu because plutonium vertically moves with sinking biogenic particles and regenerates into soluble forms in deep waters as a result of microbial decomposition of particles (Hirose et al., 2006).

In monitoring ¹³⁷Cs in sediment and sea water in the Indonesia Sea in 2011-2013 reported that on the western coasts of West Sumatra, Bangka Island, North Java, South Java and Madura Island, the ¹³⁷Cs concentrations in the seawater were 0.12±0.66 Bq/m³. In the along coast of the Bangka Island, the activity of ¹³⁷Cs in the sea water were 0.31±0.03 Bq/m³ in 2011 and 0,26±0.02 Bq/m³ in 2013 (Suseno & Prihatiningsih, 2014). No significant different value between these data from Bangka Sea (10⁵–10⁶) and Belitung Sea (10⁷), because no radioactive input from these island.

In the other paper, (Suseno & Budi, 2015) report that the activity concentration of ¹³⁷Cs in the surface seawater of the South Java Sea ranged from below MDA to 0.13 Bq/m³. On the other hand, the activity concentration of ¹³⁷Cs in the surface seawater of the West Sumatra Sea ranged from below MDA to 0.28 Bq/m³. The concentration of this study was comparable with the concentration data from South Java and West Sumatera Sea.

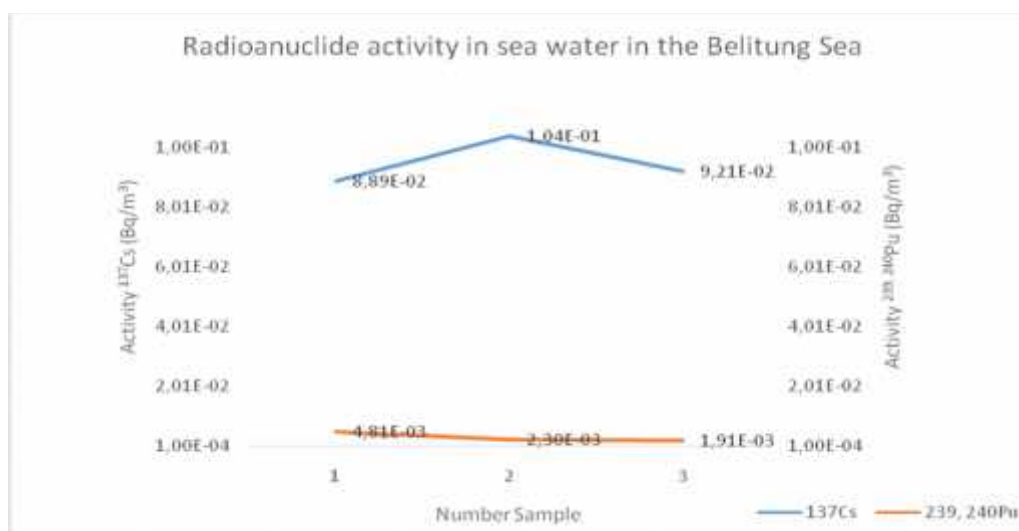


Figure 2. Activity ¹³⁷Cs and ^{239,240}Pu in the Surface Sea Water in Belitung Sea

The study that performance by the other (Wo & Ahmad, 2004), found that the activities of ¹³⁷Cs in the range of 2.33 to 5.00 Bq/m³ and 1.76 to 4.76 Bq/m³ for the South China Sea and the Straits of Malacca, respectively (Wo & Ahmad, 2004). If we compare with other study in the Western North Pacific, activity of ¹³⁷Cs in the surface sea water are 2.70±0.25 Bq/m³, in the Sulu and Indonesian seas are eastern Indian Ocean are 2.42±0.12 Bq/m³. Otherwise, in the South China Sea are 2.70±0.11 Bq/m³. (Yamada et al., 2006).

Data concentration of ¹³⁷Cs in the surface water of the northwest Pacific after the Fukushima nuclear accident at 2011 around 826 mBq/l (Yu et al., 2015). The Fukushima accident also add the value of the anthropogenic matter to the environment and according TEPCO, estimated published in May 2012, the release ¹³⁷Cs into atmosphere about 10 pBq. Radionuclides released into atmosphere are subjected to a variety of physical processes that determine their eventual fate, ¹³⁷Cs can rapidly bond to aerosol id is thus highly subject to washout removal (scavenging) by rain from contaminated air masses. In the Asia, the air mass from Fukushima was detected in the Philippines (0.48 mBq/m³), Vietnam (0.03 mBq/m³), South Korea (1,25 mBq/m³), Taiwan (1,44 mBq/m³) and China (1,55 mBq/m³), and no data from Indonesia (Thakur et al., 2013).

The Activity of ^{239,240}Pu in the surface waters of the western North Pacific is 1.65±0.12 Bq/m³, in the Sulu and Indonesian seas are eastern Indian Ocean are 2.42±0.12 Bq/m³ and 3.16±0.32 Bq/m³. Otherwise, in the South China Sea is 2.52±0.19 Bq/m³. (Yamada et al., 2006). Duran (Activities ^{239,240}Pu from this research about 3.01E-3±8.1E-4 Bq/m³, if we compare, it is found that the ^{239,240}Pu in the Belitung sea lower than western North Pacific, the Sulu and Indonesia Sea and also from South China Sea. In ASPAMARD data compilation for Pacific Ocean, activity ^{239,240}Pu in surface seawater ranged from 0.8 to 84.3 mBq/m³ with a median of

5.9 mBq/m³ (n=261). However, about 92% of the data were between the range of 0.8–14 mBq/m³ only (Duran et al., 2004).

Sea Sediment

The Activity of ¹³⁷Cs and ^{239,240}Pu in the sea sediment around the Belitung Island range from 3.11E-01±3.36E-02 Bq/kg to 6.43E-01±6.57E-02 Bq/kg and 6.66E-04±2.52E-04Bq/kg to 3.65E-02±1,62E-02Bq/kg respectively, are show in the Figure 3.

In the ASPAMARD, ¹³⁷Cs concentrations was observed in surface sediment (0-2 cm) collected from areas in the Northern Hemisphere ranged from 0.08 to 23.4 Bq/kg dry, with a median concentration of 1.4 Bq/kg dry (n=86). The highest ¹³⁷Cs levels in surface sediment were observed in the East Sea/Sea of Japan within the 35° to 40° N band. The ^{239,240}Pu concentrations in surface sediment (0-2 cm) ranged from 0.02 to 3.7 Bq/kg dry with a median of 0.2 Bq/kg dry (n = 30). (Duran et al., 2004).

Activity Ratio ^{239,240}Pu/¹³⁷Cs in Seawater and Sedimen at Belitung Sea

The activity ratio ^{239,240}Pu/¹³⁷Cs indicate the source of the anthropogenic radionuclide measured in some place. The activity ratio of ^{239,240}Pu/¹³⁷Cs in sea water at Belitung Sea are shown at Table 2.

The activity ratio ^{239,240}Pu/¹³⁷Cs from this study were 0.021–0.054 are significantly near by the ratio predicted from the atmospheric fallout of 0.025 however lower than the mean global fallout ratio of 0.176±0.014. (Yamada et al., 2006). Its mean that, the anthropogenic sources in Belitung sea come from global fallout. The ratio from this study also lower than the atom ratios in the surface waters of the western North Pacific Ocean and its adjacent seas range from 0.199±0.026 to 0.248±0.027. (Yamada et al., 2006).

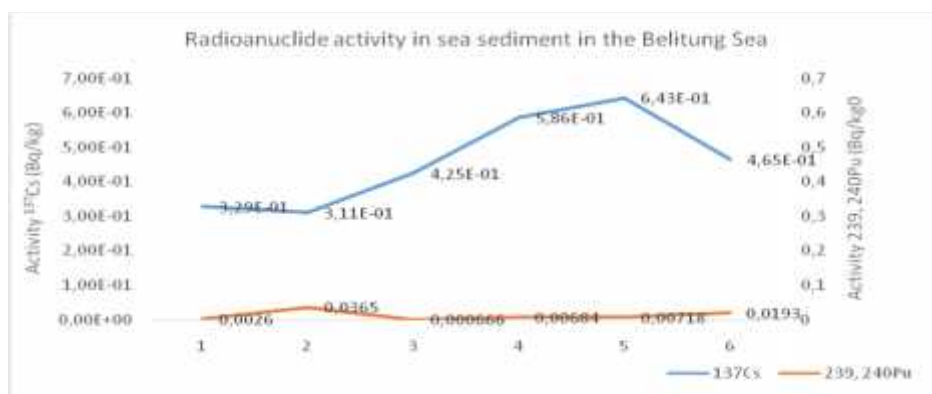


Figure 3. Activity ¹³⁷Cs and ^{239,240}Pu in Sea Sediment in Belitung Sea

Tabel 2. Activity Ratio of ^{239,240}Pu/¹³⁷Cs in the Surface Seawater in Belitung Sea

Latitude	Longitude	^{239,240} Pu(Bq/m ³)	¹³⁷ Cs (Bq/m ³)	Activity Ratio Pu/Cs
02 44 07.5	107 35 49.3	8.89E-02±1.02E-02	4.81E-03±1.51E-03	0.054
02 42 35.7	107 35 38.3	1.04E-01±1.19E-02	2.30E-03±4.50E-04	0.022
02 33 37.4	107 39 27.5	9.21E-02±1.058E-03	1.91E-03±4.71E-04	0.021

Summary

The present baseline concentration of the considered radionuclide's show low concentration of cesium and plutonium in sediment and surface seawater and their concentration is comparable to most oceanic waters. The activity ¹³⁷Cs and ^{239,240}Pu in the surface seawater in Belitung Sea were 9.5E-02±7.16E-3 Bq/m³ and 3.01E-3±8.1E-4 Bq/m³ respectively and in the sea sediment were s and ^{239,240}Pu in the sea sediment around the Belitung Island range from 3.11E-01±3.36E-02 Bq/kg to 6.43E-01±6.57E-02 Bq/ kg and 6.66E-04±2.52E-04 Bq/kg to 3.65E-02± 1,62E-02 Bq/kg respectively. This value is lower than the ASPAMARD data from Pacific Ocean. The activity ratio ^{239,240}Pu/¹³⁷Cs from this study were 0.021-0.054 are significantly near by the ratio predicted from the atmospheric fallout of 0.025 however lower than the mean global fallout ratio, its mean that, the anthropogenic sources in Belitung sea come from global allout.

Acknowledge

This work was supported by the DIPA PTKMR BATAN, and many thanks for marine radioecology group have been supported this research in sampling, did the laboratory work and measurement.

References

- Du, Y., & Qu, T. (2010). Three in flow pathways of the Indonesian through flow as seen from the simple ocean data assimilation. *Dynamics of Atmospheres and Oceans*, 50(2), 233–256. <http://doi.org/10.1016/j.dynatmoce.2010.04.001>.
- Duran, E. B., Povinec, P. P., Fowler, S. W., & Airey, P. L. (2004). Pu levels in the Asia-Pacific regional seas. *Journal of Environmental Radioactivity*, 76, 139–160. <http://doi.org/10.1016/j.jenvrad.2004.03.023>.
- Hirose, K., Aoyama, M., Kim, C. S., Kim, C. K., & Povinec, P. P. (2006). *Plutonium isotopes in seawater of the North Pacific: Effects of close-in fallout. Radionuclides in the Environment: International Conference on Isotopes in Environmental Studies* (Vol. 8). Elsevier Masson SAS. [http://doi.org/10.1016/S1569-4860\(05\)08005-8](http://doi.org/10.1016/S1569-4860(05)08005-8).
- J. Zheng, T. Aono, S. U. et al. (2012). Distribution of Pu Isotopes in Marine Sediments in the Pacific 30 km off Fukushima after the Fukushima Daiichi Nuclear Power Plant Accident. *Geochemical*, 46, 361–369.
- Kinoshita, N., Sumi, T., Takimoto, K., Nagaoka, M., Yokoyama, A., & Nakanishi, T. (2011). Science of the Total Environment Anthropogenic Pu distribution in Tropical East Pacific. *Science of the Total Environment*, The, 409, 1889–1899. <http://doi.org/10.1016/j.scitotenv.2011.01.047>
- Levy, I., Povinec, P. P., Aoyama, M., Hirose, K., Sanchez-cabeza, J. A., Comanducci, J., ... Yim, S. A. (2011). Marine anthropogenic radiotracers in the Southern Hemisphere: New sampling and analytical strategies. *Progress in Oceanography*, 89, 120–133. <http://doi.org/10.1016/j.pocean.2010.12.012>.
- Qu, T., Song, Y. T., & Yamagata, T. (2009). An introduction to the South China Sea through-flow: Its dynamics, variability, and application for climate. *Dynamics of Atmospheres and Oceans*, 47, 3–14. <http://doi.org/10.1016/j.dynatmoce.2008.05.001>
- Suseno, H., & Budi, I. (2015). Radiocesium monitoring in Indonesian waters of the Indian Ocean after the Fukushima nuclear accident. *Marine Pollution Bulletin*, 97(1-2), 539–543. <http://doi.org/10.1016/j.marpolbul.2015.05.015>.
- Suseno, H., & Prihatiningsih, W. R. (2014). Monitoring and 2013 Cs at marine coasts in Indonesia between 2011. *Marine Pollution Bulletin*, 88, 319–324.
- Thakur, P., Ballard, S., & Nelson, R. (2013). Science of the Total Environment An over-view of Fukushima radionuclides measured in the northern hemisphere. *Science of the Total Environment*, The, 458-460, 577–613. <http://doi.org/10.1016/j.scitotenv.2013.03.105>.
- Wo, Y. M., & Ahmad, Z. (2004). Determination of ¹³⁷Cs in Seawater Surrounding Peninsular Malaysia Case Study. *Journal of Nuclear and Related Technologies*, 1(2), 19–28.
- Yamada, M., Zheng, J., & Wang, Z. (2006). ¹³⁷Cs, ^{239,240}Pu and atom ratios in the surface waters of the western North Pacific Ocean, eastern Indian Ocean and their adjacent seas. *Science of the Total Environment*, The, 366, 242–252. <http://doi.org/10.1016/j.scitotenv.2005.08.014>.

Yu, W., He, J., Lin, W., Li, Y., Men, W., Wang, F., & Huang, J. (2015). Distribution and risk assessment of radionuclides released by Fukushima nuclear accident at the northwest Pacific. *Journal of Environmental Radioactivity*, 142, 54–61. <http://doi.org/10.1016/j.jenvrad.2015.01.005>.

Discussion

Q : Andreas Belhofer

How far is Belitung Island from the location of planned Power Plant ?

A : Murdahayu

It is not so far (it is about < 100 km)

Q : Stephen Solomon

Regarding the result of activity, I found that error looks very high, it is a mistake in typing the numerical result ?

A : Murdahayu

It is a miss type, I will make a correct typing in the revision paper, Thank for the correction.

Q : Gede S.Wijaya

Did you use a tracer to check the recovery process of Pu measurement? And how much is that recovery.

A : Murdahayu

We used the ²⁴²Pu tracer to check the recovery. The recovery of ²⁴²Pu is 25-30% from initial concentration.