



## JURNAL SEGARA

<http://ejournal-balitbang.kkp.go.id/index.php/segara>

ISSN : 1907-0659

e-ISSN : 2461-1166

Accreditation Number : 158/E/KPT/2021

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### DETERMINATION OF $^{137}\text{Cs}$ IN BATAM WATER AS SOUTH CHINA SEA EXTENTION USING AMMONIUM PHOSPHOMOLIBDATE (AMP) METHOD

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Received: 23 August 2021; Revised: 1 December 2021; Accepted: 22 December 2021

#### ABSTRACT

A Study of the determination of  $^{137}\text{Cs}$  has been carried out in Batam Water. The South China Sea, through several straits, is connected to the Pacific Ocean region and the Indonesian Sea. The mass of Pacific water entering the South China Sea will reach Batam Water with radioactive contaminants. Batam Water is part of Karimata Strait, which is the Extension Line of the South China Sea, making this potentially receive  $^{137}\text{Cs}$  of radioactive contaminants from operational nuclear reactors discharged from the regional area. The purpose of the study is to obtain the current status of  $^{137}\text{Cs}$  activity concentration as a baseline data to minimize the impact of nuclear activities on the ecosystem of Batam Water, Indonesia. The  $^{137}\text{Cs}$  determination method uses the AMP with modification of base dissolution and measurement validation with Certified Reference Material (CRM). The activity concentrations of the  $^{137}\text{Cs}$  in Batam seawater ranged from  $<0.03$  to  $0.35 \pm 0.02 \text{ Bq.m}^{-3}$ , while the activity concentrations of  $^{137}\text{Cs}$  in the sediments were up to  $0.22 \pm 0.08 \text{ Bq.kg}^{-1}$  with  $^{134}\text{Cs}$  below Minimum Detectable Activity (MDA). The results suggest that the activity concentrations of  $^{137}\text{Cs}$  in seawater and sediments are comparable to previous researches related to the presence of radiocesium in the region.

**Keywords:** Batam Water, South China Sea,  $^{137}\text{Cs}$ , Ammonium Phosphomolibdate (AMP) Method.

**INTRODUCTION**

Nuclear Power Plants (NPP) in the Asia Pacific region have continued to increase in the last 10 years (Ho *et al.*, 2019). According to PRIS - IAEA 2020 data (Power Reactor Information System - International Atomic Energy Agency), China is the country with the third-largest number of nuclear reactors after the United States and France with a total capacity of 286,501 GW.h from 48 reactor operational units. In addition, Japan also operates 33 nuclear reactor units with a total capacity of 31,679 MWe and South Korea has 23,150 MWe total net capacity from operation of 24 reactors (PRIS., 2020). On the other hand, NPP in India, Pakistan and the plan to build nuclear power plants in Bangladesh and Vietnam in South Asia regional areas is thought to have potential impact on Indonesian seawater as a result of normal discharge conditions as well as in emergency conditions.

The impact of possible nuclear accidents or the normal operation of nuclear power plants in China has been predicted using Compartment Model based modeling with the Poseidon-R device (Bezhenar, 2019). The modeling results show that the waters of the Indonesian Sea as part of the South China Sea will be affected by discharge from the normal operational simulation of nuclear power plants and emergency conditions. The water of Batam Island in the Karimata Strait have a very strategic position and are traversed by the international routes. Pacific water masses that enter the South China Sea through the intrusion of the Kuroshio Flow in Japanese waters will reach Batam

Water (Li *et al.*, 2019; Nan *et al.*, 2015).

Radionuclide  $^{137}\text{Cs}$  is the result of fission (fission) from nuclear reactions in which water is able to easily distribute across countries (Suseno *et al.*, 2015). The transfer allows  $^{137}\text{Cs}$  radionuclide from nuclear power plants in China, Japan and South Korea to be distributed to Batam Water. The activity concentration of  $^{137}\text{Cs}$  in the waters of the East South China Sea was known to be 0.66 - 1.36 Bq/m<sup>3</sup> (Zhou *et al.*, 2018) while the total inventory of  $^{137}\text{Cs}$  in the South China Sea is 1,900 Bq/m<sup>3</sup> (Dong *et al.*, 2010). On the other hand, numbers of research related to the presence of  $^{137}\text{Cs}$  in the Karimata Strait has previously been done, (Suseno & Prihatiningsih, 2014) suggesting that the concentration of  $^{137}\text{Cs}$  activity in the waters of Bangka, Belitung was known to be 0.49 - 0.66 Bq/m<sup>3</sup> and in West Kalimantan seawaters was of 0.08 ± 0.01 - 0.73 ± 0.06 Bq/m<sup>3</sup> (Prihatiningsih *et al.*, 2020). The significance of this study is to provide more comprehensive baseline data of  $^{137}\text{Cs}$  in Indonesian Water, especially from Batam Water as South China Sea extention using the AMP method with base dissolution modification and validation of measurements with CRM.

**METHODOLOGY**

Seawater and sediment samples were taken during the East Monsoon transition period in December 2019 within the Batam Water (Figure 1). The coordinates of the sampling location were determined using GPS. Sediment samples from each station were taken using an Ekman Grab Corer as much as 1-2 kg and placed in

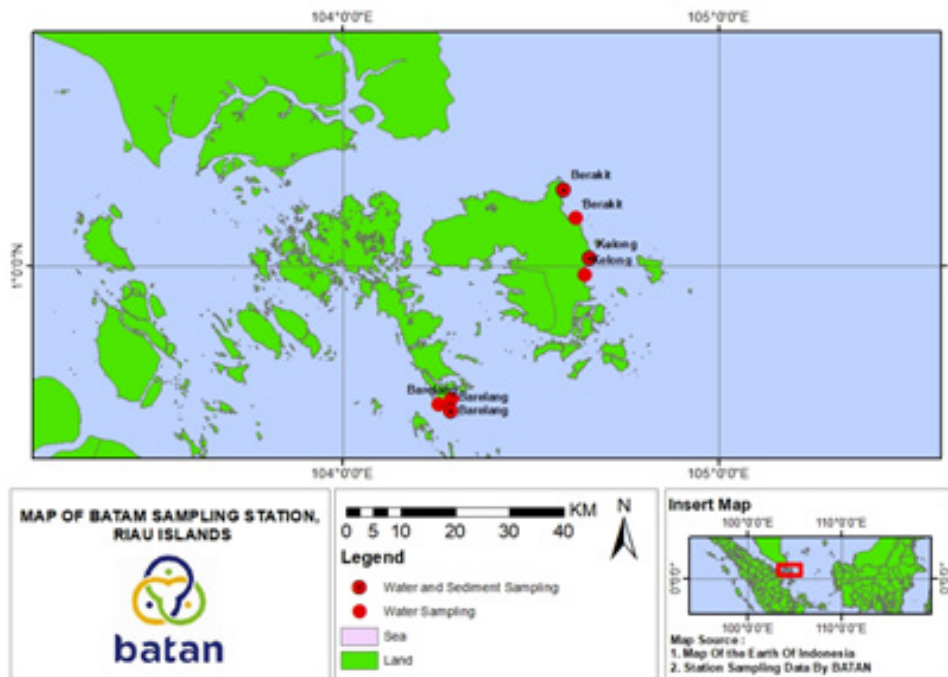


Figure 1. Map of Batam Water sampling locations.

plastic containers before being air-dried and further prepared at the PTKMR Radioecology Laboratory - BATAN. Seawater samples as much as 100-150 liters were taken using a Water Sampler and put in a plastic container and conditioned under acid conditions using 5M HCl before deposition with (AMP).

**Sample Preparation**

The seawater sample was placed in the open container, in which a carrier of Cs 40 mg and AMP were added. The method of deposition of <sup>137</sup>Cs with AMP refers to modified procedures from (Aoyama *et al.*, 2000) and (Levy *et al.*, 2011) (Figure 2). The sediment samples were then air-dried and then dried in an oven at a temperature of 105°C for 1-2 days until constant moisture content were obtained. The dried sediments were mashed with a Sediment Grinder until a uniformity of 0.5 microns were obtained, then the sediment sample were put in marinelli containers for analysis using Gamma Spectrometry.

**Gamma Spectrometric Analysis**

Analysis the concentration of <sup>137</sup>Cs activity in seawater and sediment was measured using Mirion Gamma Spectrometry equipped with a Coaxial HPGe detector integrated with the Model 2022 Amplifier Spectroscopy and High Voltage Power Supply (HVPS) Model 3106D. The Gamma Spectrometry System was calibrated with the <sup>152</sup>Eu Sediment Standard and the Cs-AMP solution standard, a cascade correction to the <sup>152</sup>Eu standard was performed with Quick Start Mode on the Genie-2000 device. Moreover, the CRM IAEA-445 and Soil Sample IAEA-315 were used for the Quality Assurance (QA) measurement results. The Standard measurements were carried out for 1-hour long, while seawater and sediment samples were measured for 72 hours. The spectrum of the measurement results was analyzed using the Genie-2000 device.

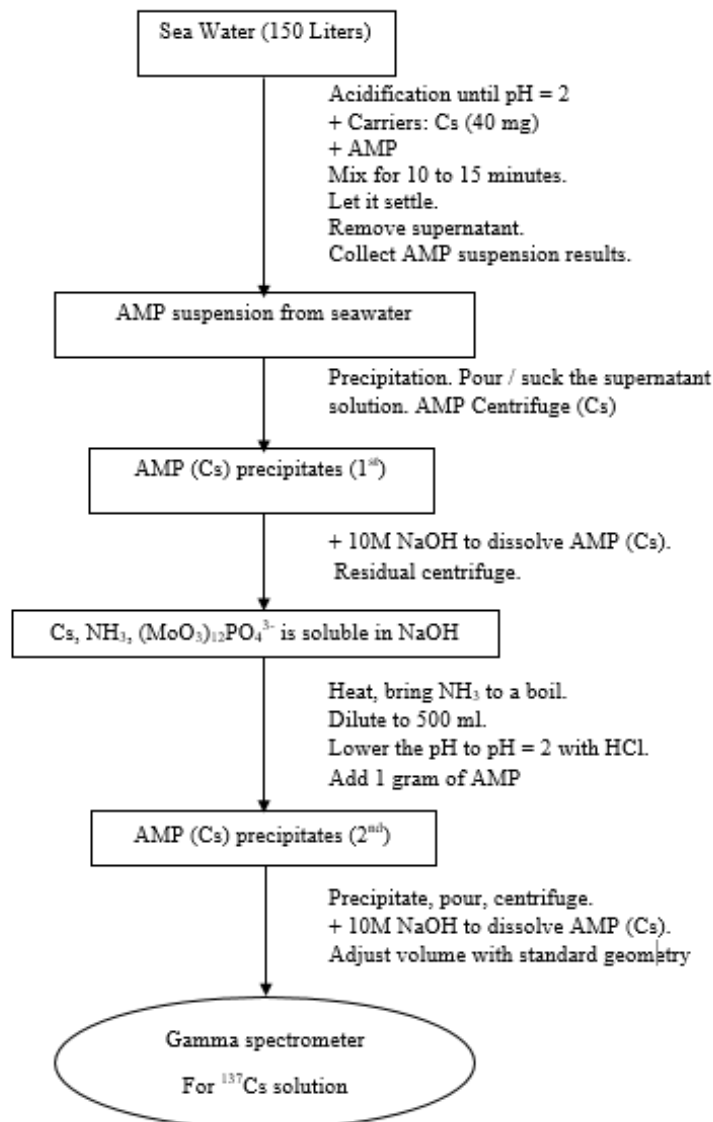


Figure 2. The cesium purification stage using the Cs-AMP dissolution method (Levy *et al.*, 2011).

## RESULTS AND DISCUSSION

China is focusing on building the "Coastal Nuclear Power Belt" in the eastern coastal area which is directly adjacent to the South China Sea (Peng *et al.*, 2019). The South China Sea, through several straits, is connected to the Pacific Ocean and the Indonesian Sea. The interconnection of the South China Sea Passage with the Indonesian Passage other than through the Mindoro and Sibutu Straits to the Makassar Strait is also known to be dominant through the Vietnam Sea to the Karimata Strait (Li *et al.*, 2019). (Susanto *et al.*, 2013) and (Wei *et al.*, 2013) predict the annual average volume of water mass transport in the Karimata strait during 2003 - 2008 to be 0.75 Sv, where in boreal winter (December - April) it is known to be 0.75 Sv. 3.6 Sv and 1.2 Sv during boreal summer (June - October). On the other hand, investigation of seawater circulation patterns in the Northwest Philippines, South China Sea and Sulu Sea by (Dong *et al.*, 2010) showed the occurrence of water mass transport from these three areas to Indonesian Water (Li *et al.*, 2019). The South China Sea Passage plays a role in carrying Pacific Water Masses from the Kuroshio Current and the North Pacific Intermediate Water Current to be forwarded to the Indonesian Cross Current through Batam Water in the Karimata Strait.

Batam Water are part of the Karimata Strait, which is the South China Sea Extension Route, so that it has the potential to receive radioactive contaminants from nuclear reactor operational releases in regional area. (Zhang *et al.*, 2019) have observed the distribution of the anthropogenic radionuclide <sup>137</sup>Cs in the Bohai Sea, Yellow Sea and Northeastern South China Waters which are indicated to come from routine local reactor operations and from Pacific waters. The concentrations of <sup>137</sup>Cs activity in the water column and sediment in the three water areas are in the respective range of 0.03 to 1.92 Bq/m<sup>3</sup> and 0.30 to 5.22 Bq/kg. The concentrations are estimated to increase if there is an emergency nuclear reactor on the South China Sea Coast and

has an impact on Batam Water (Bezhenar, 2019). In Indonesian Water, (Prihatiningsih *et al.*, 2016) have investigated <sup>137</sup>Cs may potentially be released from the nuclear facilities in Serpong to the Jakarta Bay through the Cisadane River. Although (Suseno *et al.*, 2018) have found that <sup>137</sup>Cs activity concentration in Jakarta Bay was comparable to other world regions (Table 2). In a sense, Indonesia does not have nuclear facilities that could significantly release <sup>137</sup>Cs to the environment, resulting in relatively comparable concentration of <sup>137</sup>Cs activity in the Indonesian Water including Karimata Straits.

Marine environmental monitoring of anthropogenic radioactivity is important in Batam Water. Apart from being a global water mass transit route, the potential of Batam Water as a habitat area for seagrass (Lubis *et al.*, 2017), benthic (Lubis *et al.*, 2018) and fishery products (Lubis & Anurogo, 2016) needs to be ensured to be safe from the threat of radioactive pollutants. The results of monitoring the concentration of <sup>137</sup>Cs activity in Batam seawater are in the range of < 0.03 – 0.35 ± 0.02 Bq/m<sup>3</sup>, while the concentration of <sup>134</sup>Cs activity in all stations is below the (MDA) value (0.03 Bq/m<sup>3</sup>). The concentration of <sup>137</sup>Cs activity in sediments in Batam Water is known to be a maximum of 0.22 ± 0.08 Bq/kg with <sup>134</sup>Cs below MDA. The results on seawater and sediment are comparable to previous research regarding the presence of radiocesium in the region (Table 2). (Inoue *et al.*, 2018) detected <sup>137</sup>Cs in the range of 1.6 - 1.9 mBq/l in seawater on the Korean Peninsula, while (Zhou *et al.*, 2018) in the Northeastern South China Sea and Luzon Strait detected an average of <sup>137</sup>Cs of 0.84 mBq/l. Similar research in the Pacific Northwest Waters and the South China Sea and East China Sea gave consecutive results for <sup>137</sup>Cs of 1.43 ± 0.42; 1.11 ± 0.14 and 1.10 ± 0.29 Bq/m<sup>3</sup> (Wu, 2018). The results of the <sup>134</sup>Cs research from all these locations show that <sup>134</sup>Cs were below the MDA. The waters of Bangka and West Kalimantan as the closest areas to the waters of Batam show a similar situation where the maximum concentration was found in Kalimantan

Table 1. Activity concentrations of <sup>134</sup>Cs and <sup>137</sup>Cs in seawater (Bq/m<sup>3</sup>) and sediment (Bq/kg) samples in Batam Water

Location	Sampling Coordinates	Sample	Radionuclide	
			<sup>134</sup> Cs	<sup>137</sup> Cs
Barelang	0°36'36" N 104°17'23" E	Seawater	< MDA	0.35 ± 0.02
		Sediment	< MDA	0.13 ± 0.05
	0°38'35" N 104°17'28" E	Seawater	< MDA	< 0.03
Berakit	0°37'44" N 104°15'29" E	Seawater	< MDA	0.08 ± 0.00
		1°12'17" N 104°35'20" E	Seawater	< MDA
	Sediment	< MDA	0.22 ± 0.08	
Kelong	1°07'41" N 104°37'19" E	Seawater	< MDA	< 0.03
	1°01'12" N 104°39'27" E	Seawater	< MDA	0.09 ± 0.00
		Sediment	< MDA	0.21 ± 0.08
	0°58'34" N 104°38'46" E	Seawater	< MDA	0.14 ± 0.01

Table 2. Comparison of <sup>134</sup>Cs and <sup>137</sup>Cs data in seawater (Bq/m<sup>3</sup>) and sediment (Bq/kg) samples in the region

Reference	Research Year	Location	Sample	Radionuclide	
				<sup>134</sup> Cs	<sup>137</sup> Cs
(Inoue <i>et al.</i> , 2018)	2014	Korean Peninsula	Seawater	< MDA	1.6 – 1.9
(Zhou <i>et al.</i> , 2018)	2011-2014	Northeast of South China and the Strait of Luzon	Seawater	< 0.08	0.84
(Wu, 2018)	2011-2015	Pacific Northwest	Seawater	< MDA	1.43 ± 0.42
		South China Sea		< MDA	1.11 ± 0.14
		East China Sea		< MDA	1.10 ± 0.29
(Zhang <i>et al.</i> , 2019)	2013-2014	Bohai Sea, Yellow Sea and East South China Sea	Seawater	< MDA	0.03 – 1.92
			Sediment	< MDA	0.30 – 5.22
(Suseno & Prihatiningsih, 2014)	2011-2013	West Kalimantan	Seawater	< MDA	0.27 ± 0.02
			Sediment	< MDA	0.53 ± 0.07
(Prihatiningsih <i>et al.</i> , 2020)	2017	West Kalimantan	Seawater	< MDA	0.73 ± 0.06
			Sediment	< MDA	1.26 ± 0.06
This Study	2019	Batam, Karimata Strait	Seawater	< MDA	< 0.03 – 0.35
			Sediment	<MDA	< 0.03 – 0.22

waters of 0.73 ± 0.06 Bq/m<sup>3</sup> in seawater and 1.26 ± 0.06 Bq/kg in sediment.

The determination of radiocesium in seawater is known to use several methods, including preconcentration to form Cs-AMP deposits (Aoyama *et al.*, 2000), chemical separation through CsPtCl<sub>4</sub> deposits (Park *et al.*, 2008), and filter impregnation with K<sub>4</sub>Fe(CN)<sub>6</sub>. (Yasutaka *et al.*, 2015). Preconcentration of Cs-AMP is a method that is widely used for measurement by γ-spectrometry. The high selectivity of AMP for cesium (AMP distribution coefficient, K<sub>d</sub> = ~ 5,500) was not affected by the presence of other ions in the environment (Park *et al.*, 2008). (Aoyama *et al.*, 2000) in a research on the determination of <sup>137</sup>Cs from deep seawater samples, showed the repeatability of AMP/Cs weights of 98.5 ± 2.8 and 96.5 ± 3.0 %, respectively. Furthermore, in this research, the relationship between pH and AMP/Cs repeatability of 100.5% was obtained at an optimum pH of 1.59. Another result was recently reported by (Putra *et al.*, 2021) using co-precipitation AMP/Cs in surface seawater sample and river water with efficiency removal of cesium was 96-100 %. In contrast to the research conducted by (Aoyama *et al.*, 2000) and (Putra *et al.*, 2021) the research on <sup>137</sup>Cs in Batam Water uses the Cs-AMP dissolution method with NaOH to equalize the standard geometry. On the other hand, measurement validation uses CRM-IAEA 445 (Gamma Emitting Radionuclides in Water) and CRM-IAEA 315 (Marine Sediment) to ensure measurement quality. The CRM-IAEA 445 standard is derived from the IAEA Proficiency Test for Determination of Radionuclides in Seawater. The test result certificate shows the radiocesium measurement results at the Marine Radioecology Laboratory of PTKMR-BATAN are appropriate (Accepted) with a bias of < 0.1%. Efforts to improve radionuclide detection capabilities, especially anthropogenic, need to be

continuously developed along with efforts to minimize the impact of radioactive pollutants on Indonesian marine ecosystems.

## CONCLUSION

The circulation pattern of the South China Sea Cross Current plays a role in bringing water masses containing <sup>137</sup>Cs and <sup>134</sup>Cs from the South China Sea to Indonesian Water in the Karimata Strait, Batam Water. Characteristics of <sup>137</sup>Cs and <sup>134</sup>Cs in the South China Sea in Batam Water are shown from the results of monitoring seawater and sediment, where the values obtained are comparable to previous data in the regional area. The current status shows that the activity concentration of <sup>137</sup>Cs in seawater in Batam Water is in the range of < 0.03 – 0.35 ± 0.02 Bq/m<sup>3</sup> and the maximum known sediment is 0.22 ± 0.08 Bq/kg with <sup>134</sup>Cs below the MDA for seawater and sediments. The baseline data of radioactivity <sup>137</sup>Cs in Batam Water in this research was obtained as an effort to reduce the impact of many nuclear reactor operations in China and other regional country and development plans in the region.

## ACKNOWLEDGEMENTS

The author's appreciation to the Research Team at the Marine Radioecology Laboratory for their scientific and technical support as well as the PTKMR-BATAN Institution for supporting the 2019 DIPA research funds. WRP as first author is playing role as a main contributor in this paper.

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