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Anthropogenic ¹³⁷Cs distribution and Radiological risk assessment in the Coastal Environments of Sri Lanka

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Abstract

The radionuclide concentration from one location to another is varied due to spreading, transferring and mixing in the huge amount of seawater. Further, the 2011 nuclear accident in Japan and the massive expansion of nuclear power plants in neighbouring countries has highlighted the importance of continuously monitoring the radioactivity of the coastal marine environment of Sri Lanka. Anthropogenic 137Cs activity concentration, in surface seawater along the coast of Sri Lanka has been estimated using the co-precipitation technique. The classical ammonium molybdophosphate (AMP) pre-concentration method has been used routinely to determine 137Cs (Cesium-137) in seawater. All the samples were analyzed by High Purity Germanium (HPGe) detector with relative efficiency of 32.6% for 72,000 seconds in order to improve the counting statistics. The radioisotope, 137Cs, which has a half-life of 30.17 years ranged from 0.33 Bqm-3 to 0.94 Bqm-3, with an average of 0.73 Bqm-3 in 2019. 134Cs (Cesium-134) was not detected. The effective half-life of 137Cs in surface water was calculated to be 15.1 years on average in the range from 13.1 to 17.8 years in surface waters in all the monitoring areas. The results prevailed that the spatial distribution confers no fresh input of 137Cs in Sri Lanka coastal region, thus showing pre-existing contamination due to past fallout.

Keywords: ¹³⁷Cs activity Sri Lanka coast, Marine radioactivity, Radiological risk assessment, Effective half-life

Introduction

The radionuclide concentration from one location to another is depended on the variation of source inputs and subsequent spreading, mixing and transferring throughout the world's oceans and seas (Jha, et al., 2012). During the last few decades, the world's seas and oceans have been contaminated in varying degrees with anthropogenic radionuclides due to the nuclear power plant accident, weapon tests and waste dumping. The northern hemisphere is more contaminated than southern hemisphere due to the large quantities of radioactive isotopes (especially ¹⁴C, ¹³⁷Cs, and ⁹⁰Sr) released into the atmosphere during nuclear weapons tests. However, the most abundant anthropogenic radio-nuclides in the marine environment is ¹³⁷Cs (Povinec, Hirose, & Aoyama, 2013).

The Fukushima nuclear power plant (FNPP) accident in 2011 caused the release of radioactive material into the environment and the atmosphere, is drawing attention important of monitoring the marine environment in this region (Men, et al., 2014) (Jha, et al., 2012). The accident contaminated the ocean with a very large amount of radioactive material that was released into the ocean, and within a week or so, it traveled around the globe by ocean currents. (Buesseler, 2016), (Tinker & Carpenter, 2012). Thus, the radionuclides dissolved in seawater travels very far through ocean currents (IRSN, 2011). Most of the attention has been on three radioactive isotopes released in large amounts: iodine-131, cesium-137, and cesium-134 (Buesseler, 2016). Due to their short half-life of Iodine-131 were present in the first few days and weeks after the accident, they can no longer be detected. Among all the radionuclides release into the environments, the mainly caused by Cesium-137 and Cesium-134 because Cesium-137 has a half-life of 30 years and remains in the environment for decades but it is also present in the ocean as a result of nuclear weapons testing in 1950s and 1960s. Earlier studies of ¹³⁷ Cs concentrations in the Indian ocean, Miyake et al. in 1975-1978, observed that ¹³⁷Cs in surface water ranged from 1.85 to 5.96 Bq/m3 lower than the south and north pacific oceans (Miyake, Saruhashi, Sugimura, Kanazawa, & Hirose, 1988). Cesium-134, with a half-life of only two years, is an unequivocal marker of Fukushima accident contamination which means that any detected in seawater samples most likely would have come from Fukushima (Becky Oskin, 2014) (Buesseler, 2016).

Radionuclides with a long half-life can remain in the environment for long periods of time and transfer to food web and have a significant impact on the environment as well as humans. (Men, et al., 2014). It has been necessary to establish the baseline levels and conduct regular monitoring of marine radioactivity in sea water and sediments of Sri Lanka to enable comparisons of any possible contamination events in the future. In addition, regular monitoring will be useful to identify any potential impact to Sri Lanka shores due to 22 nuclear reactors in operation in 7 nuclear power plants as of March 2018, the nearest Nuclear Power Plant (NPP) is situated in Kudankulam, India and the aerial distance to closest place in Sri Lanka, Mannar is 258 km, and Bangladesh is constructing an NPP with 2 nuclear reactors.

The risk of release of radioactive material into the environment can be determined by software by using available standardized software, such as the ERICA-Tool (Environmental Risk from Ionizing Contaminants: Assessment and Management), which has undergone substantial capability upgrades in recent years (Suseno, Budiawan, Muslim, Makmur, & Yahya, 2018). The radionuclide activity concentrations in soil, water and air are basically the inputs that should be provided to this software to assess radioactive risk. Using the ERICA Tool, whole body activity concentrations of radionuclides in biota are predicted from media activity concentrations, by using equilibrium concentration ratios.

However, relatively few studies have been conducted in the Indian ocean on the behavior and spatial variation of anthropogenic radionuclides (Provinec, et al., 2005; Provinec, et al., 2003). Recently Asia Pacific Marine Radioactivity Data-base (ASPAMARD) was developed jointly by the IAEA Regional Co-operative Agreement (RCA) and United Nation Development Program with the objectives to characterize the fate and behavior of key radionuclide contaminants in the regional seas. Input for most of the data contributed in ASPAMARD was through the RCA projects IAEARAS/7021 and 7028 which have focused on generating bench-mark data on ¹³⁷Cs activity concentration in coastal area.

After the Fukushima Dai-Ichi Nuclear Power Plant (FDNPP) accident in March 2011, a comprehensive marine environmental monitoring program has been carried out around the coastline of Sri Lanka. In this paper, radioactivity levels anthropogenic radionuclides is presented based on the data obtained during the monitoring program with an emphasis placed on the potential radiological risk associated with these marine samples. In this study, the activity concentration of ¹³⁷Cs and ¹³⁴Cs was measured selected locations around the coastline of Sri Lanka.



Figure 1 Sampling Location around the Sri Lanka coast

Methods

2.1 Sampling locations and sample preparation:

Sri Lanka is an island lying to the south east of India between the latitudes 5^0 55'- 9^0 51' north and longitudes 79° 41'- 81° 54' east and the country has a 1700 km long coastline (Koralagama, 2008). Marine water samples were collected along the Sri Lanka coastline and sampling locations were selected to cover the coastal area as uniformly as possible. Each sampling point was located approximately 1 km away from the shore. The sample locations were recorded in terms of degree - minute -second (Latitudinal and Longitudinal position) using hand-held Global Positioning System (GPS) (Model: GARMIN GPS-12) unit. Surface sea water samples were collected (100 L per sample, at an average depth of 500 m). The sampling locations are shown in fig 01. All samples were brought to the laboratory at the Sri Lanka

Atomic Energy Board and samples were filtered, after measuring the pH, and stored at room temperature $(25 \, {}^{0}\text{C})$ in a dark place.

1.2 Sample preparation and analysis for AMP pre-concentration method:

Reagents and reference solution: AMP, CsCl, HCl and NaOH (analytical grade).

The classical ammonium molybdophosphate (AMP) pre-concentration method has been used routinely to determine ¹³⁷Cs in seawater (Suseno, Budiawan, Muslim, Makmur, & Yahya, 2018), (Men, et al., 2014), (Visetpotjanakit & Tumnoi, 2016). The filtered seawater sample was transferred to a plastic water bath and pH was adjusted to below 2 to using HCl prevent AMP dissolution, and 100 g of AMP was added. The samples were stirred for 30 minutes to ensure that the sample was mixed well, and left overnight to allow the precipitate to settle. The supernatant was removed by siphoning, and the residue was centrifuged. After rinsing three times using deionized water, the residue was dried in the oven for two hours at 60 $^{\circ}$ C. The resultant precipitate was dried and packed in the specific container called GI geometry (8.4 x2.9 cm radon impermeable plastic container) chosen for the study and placed in the gamma detector with the relative efficiency of 32.6%. The weight of the sample was measured, and the activity was counted to determine the activities of ¹³⁴Cs and ¹³⁷Cs.

for 72,000 seconds counting time.

A proficiency testing (IAEA-TEL 2018) solution which contained 2010 Bq m⁻³ of 137 Cs activity, was diluted into 15 ml of sea water in the calibrated container. The samples were acidified with HCl to adjust the pH below 2. These spiked seawater samples were analyzed for 137 Cs using two different methods.

Method A: Fifteen g of AMP was added to the one of 15 l of sea water sample and allowed to settle overnight. The weight of the sample was measured, and the activity was counted for 72,000 seconds counting time.

Method B: Three g of AMP were added to the 15 l of spiked sea water sample. The samples were stirred for one hour and left overnight to allow the precipitate to settle. The precipitate samples were centrifuged and washed with 1 M HCl twice. The precipitate samples were dissolved with 10 M NaOH. The solutions were heated to remove ammonia. The precipitation step was repeated. The second precipitate samples were dissolved with 10 M NaOH. The solutions were transferred to calibrated containers (the same shape and type as prepared calibration source) and made up to the same volume as the calibration source.

3. Instrument

3.1 Gamma-ray spectrometry

All the samples were analyzed by High Purity Germanium (HPGe) detector with relative efficiency of 32.6% for 72,000 seconds in order to improve the counting statistics. The gamma spectrometry system was equipped with a coaxial p-type High Purity Germanium (HPGe) detector connected through spectroscopy amplifiers and multi-channel analyzer driven by a computer-based operating system. Genie 2000 software package (Canberra) was used for data acquisition and analysis. The detector had coaxial closed facing geometry with energy resolution of 1.84 keV at 1.3 MeV gamma line of ⁶⁰Co. Detector mode GX3020(Canberra) with the detector was shielded by a cylindrical lead shield, which had average thickness of 11 cm to reduce the background radiation. Soil 6, standard reference material was used for detector calibration. Genie 2000 LABSOCS software was used to check the detector performance. DSA1000 (Digital spectrum analyser) was used to analyse the spectra.

Results and discussion

4.1 Method verification

The quality control/assurance of analysis carried out at the laboratory has been confirmed by participating in the proficiency tests organized by the IAEA ALMERA network and other international organizations. Table 1 shows the analytical performance evaluation of the spiked samples. The laboratory results are in good agreement with the target value. The performance evaluation in the IAEA proficiency tests confirms the reliability and traceability of the analytical measurement result of the laboratory.

Sample	Spiked	Activity concentration of ¹³⁷ Cs /Bq m ⁻³		Recovery,
name	volume/g		%	
		Target value/ Bq m ⁻³	Laboratory value/Bq m ⁻³	
А	9.61	1.24	1.22	98.4%
В	9.56	1.23	1.16	94.0%

Table 1: Results of analysis of spike samples for method verification

3.2 Variation of radioactivity level of surface sea water

The spatial distribution of 137 Cs is shown in table 02.

Table 2 Activity concentration of ¹³⁷Cs of sea water samples collected in 2019

Location Name	Location GPS		¹³⁷ Cs Activity/Bq m ⁻³	Unc/ Bq m ⁻³
	Latitude	Longitudinal		(k=2)
Kalutara	6.5696	79.9545	0.65	0.11
Tangalle	5.9931	80.7525	0.75	0.12
Trincomalee	8.5225	81.2339	0.73	0.10
Kankesanthurai	9.8319	80.0267	0.72	0.12
Kalpitiya off	8.4128	79.7758	0.85	0.13
Negombo	7.2186	79.8072	0.87	0.16
Batticaloa	7.7850	81.7867	0.94	0.17
Paanama	6.7708	81.8453	0.33	0.13

The radioisotope, ¹³⁷Cs, which has a half-life of 30.17 years, ranged in concentration from 0.33 Bqm⁻³ to 0.94 Bqm⁻³, with an average of 0.73 Bqm⁻³ in 2019. In 2015, ¹³⁷Cs activity concentration was observed to range from 0.68 Bqm⁻³ to 1.22 Bqm⁻³, with an average of 1.01 Bqm⁻³. The data shows a non-uniform distribution of ¹³⁷Cs concentrations at the different sampling locations covering the Sri Lanka coastal region. A previous study in 2013 reported that ¹³⁷Cs in seawater from the eastern and western coastal area of Sri Lanka ranged from 0.72-1.72 Bq m⁻³ with an average of 1.20 Bq m⁻³ (Wickramasooriya, Waduge, Attanayake, & Udugala-Ganehenege, 2015). The ¹³⁷Cs activity concentration in 2019 was with region plotted to have a better representation of spatial variation. ¹³⁷Cs activity concentration in surface sea water in region depicts non uniform distribution (Fig. 2). Variation in the ¹³⁷Cs activity concentration in sea water may be due to factors such as scavenging to sediment, biogeochemical processes or primary productivity prevailing in the region.



Figure 1: Average ¹³⁷*Cs activity according to the location*

The activities of ¹³⁴Cs were not detected in any of the surface sea water samples because ¹³⁴Cs, with a half-life of only two years (2.06 yrs.) and ¹³⁴Cs has been released the FNPP accident has decayed to an extremely low level, which is below the detection limit. There are no nuclear power plants in the vicinity of the sampling location and Sri Lanka has not conducted any nuclear weapons testing. The anthropogenic radioactivity present is most likely due to atmospheric fallout from nuclear weapons tests or accidents such as Chernobyl which occurred in the North hemisphere.

To understand the temporal variation of 137 Cs in surface sea water, published data for the Indian ocean at locations adjoining Sri Lanka coast were assessed. Table 3 gives the temporal variation of 137 Cs activity concentration which includes results published by various authors for region adjoining of Sri Lanka coast. The 137 Cs activity concentrations in surface sea water were plotted against time to have a better understanding of decrease covering the entire region along the coast of Sri Lanka. This data also suggests a decreasing trend in 137 Cs with time. This implies that the physical radioactive decay is a major factor in the changes of 137 Cs concentrations over time and there are also no new, significant inputs of 137 Cs in the general region.

Year	Activity	Uncertainty/ Bqm	Reference	
	Concentration/Bqm ⁻³	3		
1962-1963	8.00	-	Average activity concentration of 137Cs in	
			surface seawater of Arabian Sea and Bay of	
			Bengal (Sreekumaran, Gogate, Doghi,	
			Sastry, & Viswanathan, 1968)	
1977	4.3	1.3	Average activity of Bay of Bengal and	
			Andaman Sea (Yamada, Zheng, & Wang,	
			2005)	
1996	1.47	0.10	Overall average activity of Bay of Bengal	
			and Andaman Sea (Yamada, Zheng, &	
			Wang, 2005)	
6				

Table 3 Comparison of ¹³⁷Cs activity concentration in Asia Pacific Regional Sea water.

2013	1.20	0.25	(Wickramasooriya, Waduge, Attanayake Udugala-Ganehenege, 2015)	
2015	1.01	0.20	Data base of Sri Lanka Atomic Energy Board	
2019	0.73	0.02 Present study		

2957



Figure 2: Time series analysis of ²²⁷ Cs activity concentration in surface water.

Fig. 2 shows the temporal change in the surface radionuclide concentration expressed by an exponential function in time. In general, Jha et al and Kusakabe et al studies that the concentration of 137Cs in surface water appears to decrease exponentially with time (Jha, et al., 2012) and this study data also shows good agreement with recent publications suggestions. Surface seawaters is shown the declining trend of ¹³⁷Cs concentration based on the assumption that the ¹³⁷Cs concentrations decrease exponentially with time,

$$Cs = C_{so} e^{-(k+\gamma)t}$$

 C_s , C_{so} , γ , and t are measured ¹³⁷Cs concentration (Bq L⁻¹ or Bq kg⁻¹), initial ¹³⁷Cs concentration (Bq L⁻¹ or Bq kg⁻¹), decrease rate (year⁻¹) and time (year), respectively (Kusakabe & Takata, 2020).

An index for the declining trend can be expressed as an effective half-life, which was calculated as follows.

effective half-life(year) = $\ln(2)/\gamma$

Effective half-life of ¹³⁷Cs in surface water was calculated to be 15.1 year on average in the range from 13.1 to 17.8 year in surface waters in all the monitoring areas.

3.3 Radiological risk assessment:

In this study, the impact of radionuclides ¹³⁷Cs on marine ecosystems was estimated and analyzed using the Erica Tool software version 1.2.1 update 12 February 2016. The highest concentrations of ¹³⁷Cs in seawater and sediment were used as radionuclide parameters with dose rate limits of 10 μ Gy h⁻¹. Table 2 shows the results and analysis from Erica Tool of

internal dose rate, external dose rate, and total dose rate in some types of marine aquatic organisms. These results indicate that the ¹³⁷Cs concentration does not significantly effect on marine biota because the dose rates are still below the established dose rate of 10 μ Gy h⁻¹. Nevertheless, potential danger from contamination of ¹³⁷Cs in marine biota may still occur through the food chain and continue to accumulate in biota which is eventually consumed by humans.

	External Dose	Internal Dose	Total Dose	Total Dose
	Rate (μ Gy/h)	Rate(μ Gy/h)	Rate(μ Gy/h)	Rate(μ Gy/h)
	¹⁵⁷ Cs	¹⁵ /Cs	¹⁵⁷ Cs	¹⁵⁷ Cs
Macroalge	1.52E-04	1.35E-02	1.36E-02	1.36E-02
Pelgic Fish	2.64E-04	3.96E-02	3.99E-02	3.99E-02
Vascular Plant	1.47E-04	1.50E-03	1.65E-03	1.65E-03
Benthic Fish	1.38E-04	1.70E-06	1.40E-04	1.40E-04
Phytoplankton	3.67E-04	5.70E04	9.36E-04	9.36E-04
Zooplankton	3.19E-04	1.56E-02	1.59E-02	1.59E-02
Mammals	1.27E-04	7.26E-02	7.27E-02	7.27E-02
Sea anemones or	1.52E-04	3.22E-02	3.24E-02	3.24E-02
True Corals				
Polychate worm	3.30E-06	2.52E-02	2.52E-02	2.52E-02

Conclusion

The marine environment is a dynamic system. The data on key anthropogenic radionuclides such as ¹³⁷Cs show that physical radioactive decay contributes to changes in radionuclide concentrations over time at a given sampling station. The first monitoring was conducted in February 2012. Comparing the monitoring ¹³⁷Cs activity concentration from 2012 to 2019 implies that downward trend during this period. Apart from fallout, there has been no substantial input from any source, including the operation of nuclear power plants in Sri Lanka coast. The results confirm that the dominant source of anthropogenic radionuclides in the marine environment is global fallout. These results indicating that ¹³⁷Cs contents do not significantly affect marine biota because the dose rates were still below from the established dose rate of 10 μ Gy h⁻¹. The result will be useful as an international reference source on the average level of ¹³⁷Cs radionuclides in the surface waters of the Indian coastal environment so that any further contribution.

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