THE ⁹⁰Sr CONCENTRATION IN SURFACE SEAWATERS OF JAPAN SEA

Muslim *)

Department of Marine Science Diponegoro University, Semarang, 50275, Indonesia

Recevoid: 20 April 2006, Accepted: 30 May 2006

ABSTRACT

Distribution of ⁹⁰Sr concentrations in surface seawater of Japan Sea has been studied from 30 June 2000 to 18 July 2000. The concentrations of ⁹⁰Sr varied according to the stations position and didn't show correlation with temperature and salinity. Distribution and level of ⁹⁰Sr concentrations were influenced by any factors such as distance from the sources of radionuclides and water current. The strong system of Tsushima Warm Current System and Kuroshio branch Current System in the Japan Sea increased the leaching of ⁹⁰Sr from the bottom sediment. The results of this study were much lower than those of previous study, presumably due to ⁹⁰Sr characteristic will decrease with time and in the environment, it becomes incorporated with calcium. However, data in 1990 increased dramatically as effect of Chernobyl nuclear power plant accident.

Key words: 90 Sr, Japan Sea, current, previous study, calcium.

*Correspondence : Phone / Fax ; 024 7474698 ; e-mail : muslim_muslim@yahoo.com

INTRODUCTION

Radionuclides content in the Japan Sea (East Sea) is very interesting to be studied due to the acceptance of radionuclides in this area from various sources, both planned and accidentally from Japan, Korea, Russia and China. This sea is one of the most highly radioactive waste dumping areas in the world (Hong et al, 1999), although the total activity dumped in the NW Pacific Ocean is still lower by factor of ~ 6 in comparison to dumping sites in the Arctic Ocean. About 456 TBq of liquid radioactive waste and 252 TBq of solid waste were dumped over the last three decades in the Sea of Japan, the Sea of Okhostk and the western North Pacific Ocean, mainly by the former Soviet Union and the Russian Ferderation (White Book, 1993). The Japanese contributed radioactive as low solid waste with amount of 15.1 TBq. Republic of Korea was negligible in

comparison to the total activity. Russian Federation in 1992 and

ISSN: 1410-5217

Accredited: 23a/Dikti/ Kep/2004

1993 contributed about 1.4 TBq. This condition has consequences on the high level of radionuclides consumption on marine foods (Livingston and Povinec, 2000). Furthermore, ⁹⁰Sr is readily as soluble form in the sea and available to human beings and the transfered through food (Kasamatsu and Inatomi, 1998). Due to aforementioned problems, the study to analyze 90Sr concentration condition in the Japan Sea is urgently required. Beside to reveal the current level, the study is aimed also at comparing the concentration from the previous studies.

MATERIALS AND METHODS

The surface sea waters were collected from 30 June 2000 to 18 July 2000. About 40 ℓ of

seawater was weighed and transferred to polyethylene bucket. Oxalate acid was added according to the formula weight of seawater (kg) x 40 x 0.12 and diluted with DDW (100 gr/l) and was stirred. The pH was adjusted with NH₄OH and HCl to ~ 5.5 and then stirred thoroughly. Time was allowed for the Sr(Ca)oxalate to settle. After Sr(Ca)oxalate precipitated the solution was separated with a vacuum pump and then dried on a hot plate (50 °C) until the cracks condition were formed, and 500 ml of 70% HNO3 was added until all Sr(Ca)oxalate was diluted (adjust the total volume 400 ml) by heating and stirring. The hot plate digestion was stopped after the SrNO₃ were formed and then precipitate was sonified with ultrasonic cleaner for about 1 hour. The precipitate of SrNO₃ (white color) was separated by centrifugation. Acetone solution was added and after one hour the precipitant was centrifuged, and the solution (supernatant) was discharged. This treatment was performed twice.

The precipitate was dried in Infra Red (IR) light. After all acetone evaporated, 10 ml of HNO₃ was added and the mixture centrifuged, DDW was added to the precipitate. To complete the dilution the samples were sonified again and then centrifuged for 10 minutes. The supernatants were transferred to another bottle, and 1 ml of Fe³⁺ carrier and few drops of ammonium solution were added until a brown precipitate (FeOH) was formed. This was centrifuged for 20 minutes, and the supernatant then was separated, 7-8 ml of saturated Na₂CO₃ was added to the supernatant and then (a precipitate of centrifuged SrCO₃ was formed). Immediately, 1-2 drops of saturated Na₂CO₃ was added, if the other precipitate was formed again, centrifugation was continued, if not, the SrCO₃ was separated and then the time was recorded. To purify the SrCO₃, DDW was added and the mixture was centrifuged twice. The $SrCO_3$ was dried under an IR lamp. A few drops of 6N HNO₃ were added until all $SrCO_3$ was dissolved and made up to 25 ml. 1 ml of aliquot was added with DDW (with 10-15 drops of 70% HNO₃) and made up to 25 ml for AAS analysis. 1 ml of Y^{3+} carrier was added on 24 ml of aliquot. ⁹⁰Sr counting was carried out with low background β counters after radioequilibrium between ⁹⁰Sr and ⁹⁰Y had been established.

ISSN: 1410-5217

Accredited: 23a/Dikti/ Kep/2004

Description of the site area

A number of nuclear plants exists along the eastern coast of the Korean Peninsula, the western coast of Japanese Islands and at Peter Great Bay (Vladivostok) of the Russia. The main sources of radionuclides in Peter the Great bay are derived from the following: (1) global atmospheric fallout; (2) river input; (3) discharge from naval facilities nearby Peter the Great bay (Tkalin and Chaykovskaya, 2000). One of these facilities, the "Zvezda" shipyard, is located Bolshov Kamen; repairing decommissioning of nuclear submarines are carried out at this shipyard (Handler, 1995). Eastern Asia, especially mainland China also contribute mineral particles and pollutants to the western North Pacific Ocean through long-range atmospheric transport (Duce et al 1983; Gao et al, 1992). Previous studies concluded that 90Sr content in the surface waters of the Japan Sea were approximately uniform with some regional exceptions, along the latitudinal zones, and that their horizontal distributions corresponded nearly to the radioactive fallout (Nagava and Nakamura, 1976). However, over the past decade there has been growing concern over dumping of radioactive waste in this sea (Hong *et al*, 1999

Figure 1. Surface seawater sampling station

According to the Korean, the name of Japan Sea started to use when Japanese colonized in Korea. Korean never call Japan Sea, they always call East Sea, because the location of this sea is in the east of Korea. This case may be similar with the name of Indian Ocean and Indonesia Ocean. The station 1 and 2 are located in the Korean coastal zone. In these areas, there are some nuclear power plants operating. Station 3 and 4 are located in the Japanese coastal zone which have similar condition with station 1 and 2 as industrial zones that contributed concentration. Station 5, 6 and 7 are located in off shore as mixing area from Korea, Japan and Russia.

RESULTS AND DISCUSSIONS

The level concentration of ⁹⁰Sr in the Japan Sea varied according to the stations and did not show correlation with temperature and salinity. Generally concentration of ⁹⁰Sr in the coastal area decreased by the distance from the mainland. The concentration of ⁹⁰Sr in station 4, that is closer to the shoreland than station 3 was 2.03±0.28 mBq/L while in station 3 was 1.32±0.23 mBq/L. This was also shown between station 7 and station 1 or 2 (**Tab.1**). ⁹⁰Sr behaviour is easily diluted and dispersed and increasing distance from the mainland has helped those two processes

as have been reported by Hong *et al* (1999) and Ikeuchi *et al* (1999). They concluded that concentration of ⁹⁰Sr in surface and bottom waters at dumping areas didn't show significant difference with values observed in background area. This characteristic, however, did not occur in open sea such as at station 5 and 6. This assumption was in agreement with Hirose *et al*, (1999) who found the high level of ⁹⁰Sr in the water column of the Japan Sea, especially north central Japan Sea. The East Sea (Japan Sea) seemed to be controlled primarily by the atmospheric input (Kang *et al*, 1997).

ISSN: 1410-5217

Accredited: 23a/Dikti/ Kep/2004

Among 7 stations that were studied (Tab. 1) the ⁹⁰Sr contents in adjacent Korean coasts (station 1 and 2) were lower than those at Japanese region (station 3 and 4). It has to be the waste dumped in the Sea of Japan by the Republic of Korea was negligible in comparison to the total activity of wastes dumped in this sea (Livingston and Povinec, 2000). On the other hand that distribution concentration of 90Sr in the Japan Sea was significantly influenced by discharges from reprocessing plants. Moreover, there were slight differences between 2 stations in the Korean coastal region. The concentration at station was 1 slightly higher than at station 2. This was perhaps due to the Taiwan-Tsushima Warm Current System and Kuroshio branch Current System passing those stations (Isobe, 1999, **Fig. 2**)

Tabel 1. Salinity, Temperature and 90 Sr concentration in the surface seawaters of Japan Sea

Stations	Latitude.	Longitude	Sampling	Salinity	Temp	⁹⁰ Sr
	(°N)	(°E)	date	(psu)	(°C)	(mBq/l)
1	34°45'036	129°08'258	2000.6.30 (18.00)	32.54	22.6	1.18 <u>+</u> 0.20
2	34°50'94	129°36'49	2000.6.30 (22.00)	32.36	22.0	1.13 <u>+</u> 0.19
3	34°20'2	130°20'3	2000.7.1 (02.50)	33.81	22.7	1.32 <u>+</u> 0.23
4	34°03'475	130°45'488	2000.7.1 (05.10)	32.60	22.1	2.03 <u>+</u> 0.28
5	44o23'7	140°21'7	2000.7.16 (00.25)	33.80	17.6	1.05 <u>+</u> 0.16
6	38°02'5	131°53'7	2000.7.16 (16.10)	34.01	24.4	1.93 <u>+</u> 0.22
7	35°58'	130°18'	2000.7.18 (04.00)	33.30	21.6	1.06 <u>+</u> 0.21
					Average	1.386

Those systems are flowing from the western north Pacific to the Japan Sea transporting surface waters and influencing the sediments and leaching processes. This evidence was similar to the dynamics in the central Great Barrier Reef-Australia, in which nutrients and some trace elements increased as effect of wind strength and sediment resuspention (Muslim and Jones, 2003). In addition, radionuclides may be transported from the sediment to the water column by physical,

chemical and biological processes (Oughton *et al*, 1997). Thus, even though Japan Sea is isolated from the North Pacific Ocean and its marginal sea it still has a change in receiving the dynamic experienced by the North Pacific Ocean System. The physical connection of its channels to the North Pacific clearly support the assumption

ISSN: 1410-5217

Accredited: 23a/Dikti/ Kep/2004

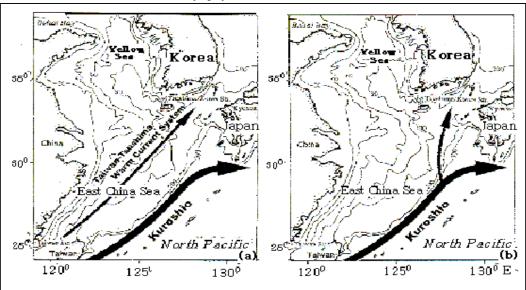


Figure 2. Schematic view of the origin of the Tsushima Warm Current as (a) a part of the Taiwan-Tsushima Warm Current System, and (b) a separation branch of the Kuroshio (Isobe, 1999).

The hazards of ⁹⁰Sr activity in the sea water to the health has attracted the concern of scientists and public. This concern is mainly with the impact of radioactive wastes on the natural environment that increased significantly. This concern has led to the increasing number of ocean sampling

expeditions. The interests in observing the ⁹⁰Sr concentration in Japan Sea in particular were started in 1964 (**Tab. 2**) and the collected data were very useful for documentation of ⁹⁰Sr concentration in the area.

Table 2. Concentration of ⁹⁰Sr in surface seawaters (mBq/l) of Japan Sea from 1964-2000

Year	Number of samples	Average conc	References
1964		19.6	Chumichev,1966
1966	4	11.1	
1966-		8.1	Chumichev,1972
1968			
1970	4	7.4	
1976-		4.1-5.1	Nagaya and Na-
1979			kamura,1981
1980	4	5.5	
1980-		2.0-3.5	Nagaya and Na-
1986			kamura,1987
1990	5	6.3	
1991	4	3.7	
1992	4	4.8	
1993	3	3.9	
1994	13	2.4	
1994	9	1.6-2.0	Joint Report, 1995
1995	2	2.0	Joint Report, 1997
2000	7	1.386	this study

 90 Sr of generally concentration decreased by the years. A number of causes was suggested for the decrease. It may be caused by radioactive decay, sedimentation (incorporated with calcium) and biological removal processes. Muslim (in press) has calculated that 90Sr concentration in the high seas and coastal regions of Korea-Japan-Russia-China decreased annually and much lower than exponential decay value. However, the concentration of 90Sr increased dramatically from 2.0-3.5 mBq/l in 1980-1986 to 6.3 mBq/l in 1990 (**Tab. 2**), presumably because on 26 April 1986 the accident of nuclear reactor of Chernobyl in which heavily contaminated in the aquatic system (Sansone et al, 1996)

CONCLUSION

The ⁹⁰Sr level story in the Japan Sea from 1964 to the present study always decreased annually, except data in 1990 after accident

of nuclear reactor of Chernobyl occurred. The current and radionuclides sources processes influenced on the distribution and concentration level of 90 Sr.

ACKNOWLEDGMENTS

I would like to specially thank to the captain and crew of Gaya training ship for their help in seawater collecting during undergraduate student training of Pukyong National University (PKNU), Busan, Korea. I also wish to thank members of Geochemistry Oceanography laboratory of PKNU for facilitating the data analysis.

REFERENCES

Chumichev., V.B. 1966. Strontium-90 content in the waters of the Pacific Ocean in 1962 and in 1964. *Trans. Inst Oceano.*, *Acadf Scie. USSR*, 82. 20-23.

ISSN: 1410-5217 Accredited: 23a/Dikti/ Kep/2004

Original paper

- Chumichev, V.B. 1972. Strontium-90 in the north-western parts of the Pacific Ocean in 1966-1968. *Trans Inst of Exp. Meth, Hydr. Ser. USSR*, 1 (32), 45-50.
- Duce, R.A., Arimoto; R., Ray; B.J., Unni, C.K and Harder, P.J. 1983. Atmospheric trace elements at Enewetak. *J. Geophy Res*, 88: 5321-5342.
- Gao, Y.,. Arimoto; R., Zhou, M.Y., Merril, J.T and Duce, R.A. 1992. Relationships between the dust concentrations over eastern Asia and the remote North Pacific. *J Geophy Res.* 97: 9867-9872.
- Handler, J. 1995. The radioactive waste crisis in the Pacific area. *Artic. Res United States*, 9, 295-306.
- Hirose, K., Amano; H., Baxter; M.S., Chaykovskaya, E., Chumichev, V.B., Hong, G.H., Isogai, K., Kim; C.K., Kim, S.H., Miyao, T., Morimoto, T., Nikitin, A., Oda, K.,. Pettersson, H.B.L., Povinec, P.P., Seto, Y., Tkalin, A., Togawa, O and Veletova, N.K. 1999. Anthropogenic radionuclides in seawater in the East Sea/Japan Sea: Result of the first-stage Japanese-Korean-Russian expedition. J. *Environ Radioac*, 43: 1-13.
- Hong G.H., Lee, S.H., Kim, S.H., Chung, C.S and Baskaran, M. 1999. Sedimentary fluxes of ⁹⁰Sr, ¹³⁷Cs, ^{239,240}Pu and ²¹⁰Pb in the East Sea (Sea of Japan). *Sci Total Environ*, 237/238, 225-240.
- Ikeuchi Y., Amono, H., Aoyama, M., Berezhnov, V.I., Chaykovskaya, E.,. Chumichev; V.B., Chung, C.S., Gastaud, J., Hirose, K., Hong, G.H., Kim, C.K., Kim, S.H., Miyao, T., Morimoto, T., Nikitin, A., Oda, K., Petterson, H.B.L., Povinec, P.P., Tkalin, A., Togowa, O and Veletova, N.K. 1999. Anthropogenic radionuclides in seawater of the Far Eastern Seas. *Sci Total Environ*. 237/238, 203-212.
- Isobe A. 1999. On the origin of the Tsushima Warm Current and its

- seasonality. *Continonl Shelf Res*, 19: 117-133.
- Joint Report. 1995. Investigation of environmental radioactivity in waste dumping areas of the far eastern sea, results from the first stage Japanese-Korean-Russian join expedition. Tokyo: Science and Technology Agency, 1-63.
- Joint Report. 1997. Investigation of environmental radioactivity in waste dumping areas of the far eastern sea, results from the second stage Japanese-Korean-Russian joint expedition. Tokyo: Sci. Tech Agency, 1-56.
- Kang, D.J., Chung, C.S., Kim, S.H., Kim, K.R and Hong, G.H. 1997. Distribution of ¹³⁷Cs and ^{239,240}Pu in the surface waters of the East Sea (Sea of Japan). *Mar. Pol Bull.* 35: 305-312.
- Kasamatsu, F and Inatomi, N. 1998. Effective environmental half lives of ⁹⁰Sr and ¹³⁷Cs in the coastal seawater of Japan. *J. Geophy Res.*. 103: 1209-1217.
- Livingstone, H.O and Povinec, P.P. 2000. Anthropogenic marine radioactivity. Ocean Coas Man 43, 689-712.
- Muslim and Jones, G. 2003. The seasonal variation of dissolved nutrients, chlorophyll *a* and suspended sediments at Nelly Bay, Magnetic Island. *Estuar Coast Shelf Sci*, 57: 445-455.
- Muslim. (in press). Distribution of ⁹⁰Sr in the high seas and coastal regions of Korea-Japan-Russia-China
- Nagaya, Y and Nakamura, K. 1976. 90 Sr and ¹³⁷Cs contents in the surface waters of the adjacent seas of Japan and the North Pacific during 1969 to 1973. *J. Oceanogr Soc Jap.* 32: 228-234.
- Nagaya, Y and Nakamura, K. 1981. Artificial radionuclides in the western north Pacific(I) ⁹⁰Sr and ¹³⁷Cs in the deep waters. *Journal of the Oceanographical Society of Japan*, 37, 135-144.
- Nagaya Y and Nakamura, K. 1987. Artificial radionuclides in the western north Pacific (II) ¹³⁷Cs and ^{239,240}Pu inventories in water and sediment column observed from 1980 to 1986. Oceanogr Soc.Jap. 43: 345-355.

Oughton, D.H., Borretzen, P., Salbu, B and Tronstad, E. 1997. Mobilisation of ¹³⁷Cs and ⁹⁰Sr from sediments: potential sources to Arctic waters. *Sci Total Environt*, 202: 155-165.

ISSN: 1410-5217

Accredited: 23a/Dikti/ Kep/2004

- Sansone, U., Belli, M., Voitsekovitch, O.V and Kanivets, V.V. 1996. ¹³⁷Cs and ⁹⁰Sr in water and suspended particulate matter of the Dnieper River-Reservoirs system (Ukraine). *Sci Totla. Environ*, 186: 257-271.
- Tkalin, A.V and Chaykovskaya, E.L. 2000. Anthropogenic radionuclides in Peter the Great bay. *Environ Radioac*, 51, 229-238.
- White Book. 1993. Facts and problems related to radioactive waste disposal in seas adjacent to the territory of the Russian Federation, Moscow: Office of the President of the Russian Federation.