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## THE $^{90}\text{Sr}$ CONCENTRATION IN SURFACE SEAWATERS OF JAPAN SEA

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### ABSTRACT

*Distribution of  $^{90}\text{Sr}$  concentrations in surface seawater of Japan Sea has been studied from 30 June 2000 to 18 July 2000. The concentrations of  $^{90}\text{Sr}$  varied according to the stations position and didn't show correlation with temperature and salinity. Distribution and level of  $^{90}\text{Sr}$  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  $^{90}\text{Sr}$  from the bottom sediment. The results of this study were much lower than those of previous study, presumably due to  $^{90}\text{Sr}$  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:**  $^{90}\text{Sr}$ , Japan Sea, current, previous study, calcium.

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### 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  $\sim 6$  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 Okhotsk and the western North Pacific Ocean, mainly by the former Soviet Union and the Russian Federation (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 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,  $^{90}\text{Sr}$  is readily as soluble form in the sea and available to human beings and transferred through the food chain (Kasamatsu and Inatomi, 1998). Due to aforementioned problems, the study to analyze  $^{90}\text{Sr}$  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

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seawater was weighed and transferred to polyethylene bucket. Oxalate acid was added according to the formula weight of seawater ( $\text{kg} \times 40 \times 0.12$ ) and diluted with DDW (100 gr/l) and was stirred. The pH was adjusted with  $\text{NH}_4\text{OH}$  and  $\text{HCl}$  to  $\sim 5.5$  and then stirred thoroughly. Time was allowed for the  $\text{Sr}(\text{Ca})\text{oxalate}$  to settle. After  $\text{Sr}(\text{Ca})\text{oxalate}$  precipitated the solution was separated with a vacuum pump and then dried on a hot plate ( $50^\circ\text{C}$ ) until the cracks condition were formed, and 500 ml of 70%  $\text{HNO}_3$  was added until all  $\text{Sr}(\text{Ca})\text{oxalate}$  was diluted (adjust the total volume 400 ml) by heating and stirring. The hot plate digestion was stopped after the  $\text{SrNO}_3$  were formed and then precipitate was sonified with ultrasonic cleaner for about 1 hour. The precipitate of  $\text{SrNO}_3$  (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  $\text{HNO}_3$  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  $\text{Fe}^{3+}$  carrier and few drops of ammonium solution were added until a brown precipitate ( $\text{FeOH}$ ) was formed. This was centrifuged for 20 minutes, and the supernatant then was separated, 7-8 ml of saturated  $\text{Na}_2\text{CO}_3$  was added to the supernatant and then centrifuged

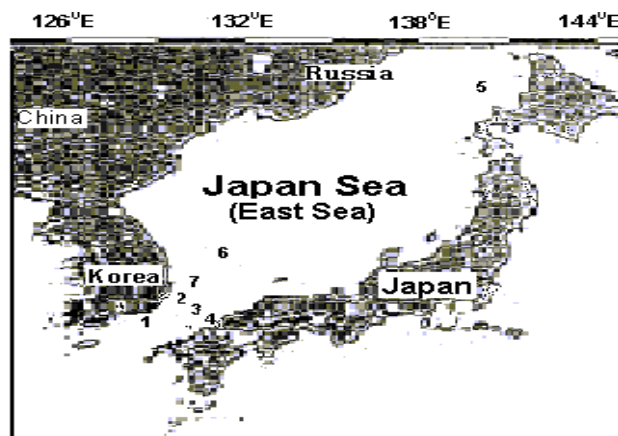
(a precipitate of  $\text{SrCO}_3$  was formed). Immediately, 1-2 drops of saturated  $\text{Na}_2\text{CO}_3$  was added, if the other precipitate was formed again, centrifugation was continued, if not, the  $\text{SrCO}_3$  was separated and then the time was recorded. To purify the  $\text{SrCO}_3$ , DDW was added and the

mixture was centrifuged twice. The  $\text{SrCO}_3$  was dried under an IR lamp. A few drops of 6N  $\text{HNO}_3$  were added until all  $\text{SrCO}_3$  was dissolved and made up to 25 ml. 1 ml of aliquot was added with DDW (with 10-15 drops of 70%  $\text{HNO}_3$ ) and made up to 25 ml for AAS analysis. 1 ml of  $\text{Y}^{3+}$  carrier was added on 24 ml of aliquot.  $^{90}\text{Sr}$  counting was carried out with low background  $\beta$  counters after radioequilibrium between  $^{90}\text{Sr}$  and  $^{90}\text{Y}$  had been established.

## 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 in Bolshoy Kamen; repairing and 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  $^{90}\text{Sr}$  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 (Nagaya 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

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**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  $^{90}\text{Sr}$  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  $^{90}\text{Sr}$  in the Japan Sea varied according to the stations and did not show correlation with temperature and salinity. Generally concentration of  $^{90}\text{Sr}$  in the coastal area decreased by the distance from the mainland. The concentration of  $^{90}\text{Sr}$  in station 4, that is closer to the shoreland than station 3 was  $2.03 \pm 0.28$  mBq/L while in station 3 was  $1.32 \pm 0.23$  mBq/L. This was also shown between station 7 and station 1 or 2 (**Tab.1**).  $^{90}\text{Sr}$  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  $^{90}\text{Sr}$  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  $^{90}\text{Sr}$  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).

Among 7 stations that were studied (**Tab. 1**) the  $^{90}\text{Sr}$  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  $^{90}\text{Sr}$  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  $^{90}\text{Sr}$  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**)

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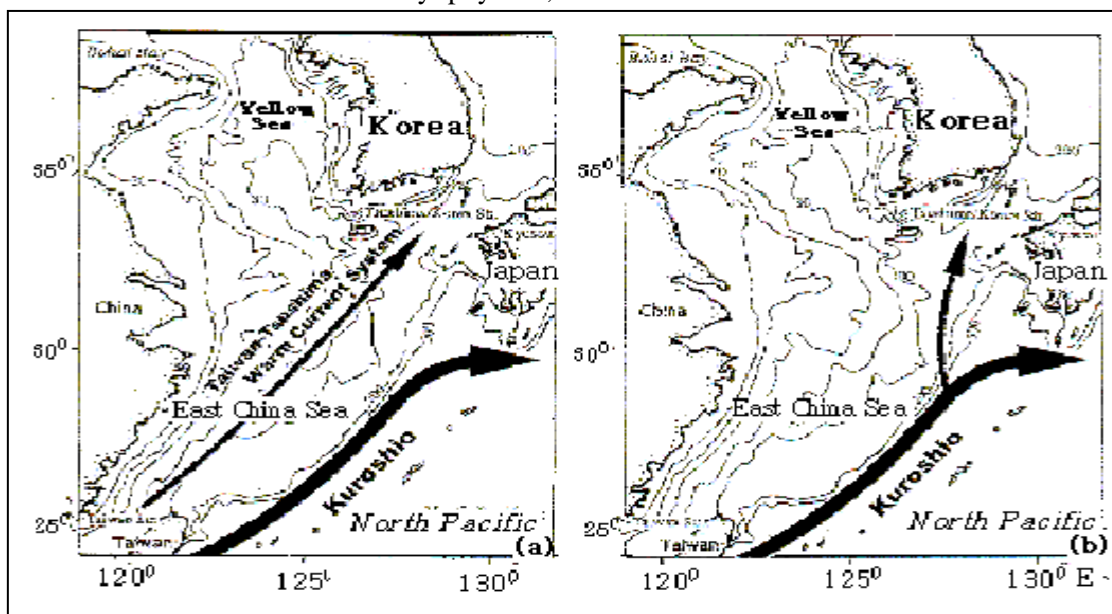
**Tabel 1.** Salinity , Temperature and <sup>90</sup>Sr concentration in the surface seawaters of Japan Sea

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



**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).

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The hazards of <sup>90</sup>Sr 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 <sup>90</sup>Sr concentration in Japan Sea in particular were started in 1964 (Tab. 2) and the collected data were very useful for documentation of <sup>90</sup>Sr concentration in the area.

**Table 2.** Concentration of <sup>90</sup>Sr 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	
1966-1968		8.1	Chumichev,1972
1970	4	7.4	
1976-1979		4.1-5.1	Nagaya and Nakamura,1981
1980	4	5.5	
1980-1986		2.0-3.5	Nagaya and Nakamura,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	<b>this study</b>

The concentration of <sup>90</sup>Sr generally 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 <sup>90</sup>Sr 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 <sup>90</sup>Sr 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 <sup>90</sup>Sr 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 <sup>90</sup>Sr.

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