SOME RESULTS OF THE SUMMER 1999 JAERI-FERHRI EXPEDITION

Tkalin A.V.[1], Aramaki T.[2], Togawa O.[2], Volkov Yu.N.[1]

[1] Far Eastern Regional Hydrometeorological Research Institute, Vladivostok, Russia
[2] Japan Atomic Energy Research Institute, Mutsu, Japan

Introduction

The objective of this project was to study distribution of anthropogenic radionuclides in the Japan Sea. Recently, some information of radioactive waste dumping by the Soviet Union and the Russian Federation in the NW Pacific and its marginal seas has been published in the White Book (Yablokov, 1993). As a result of such radioactive waste dumping, the question on possible ecological effects has been arisen. In order to determine radionuclide activities in seawater and bottom sediments, the joint research expedition to the sites of radioactive waste dumping has been organized by the concerted government efforts of Japan, Republic of Korea and Russian Federation. The first stage of the expedition, where the researchers from Japan, Korea, Russia and IAEA participated, was carried out in March-April 1994 in the Japan Sea. The expedition results were jointly evaluated and presented in the report (Joint Expedition, 1995). The second stage of joint expedition was implemented in August-September, 1995 in the areas of radioactive waste dumping in the Japan and Okhotsk seas and in the NW Pacific. The results were also presented in the joint report (Joint Expedition, 1997).

The data obtained as a result of joint investigation allowed to estimate radioactivity levels in seawater and bottom sediments in the Russian, Korean and Japanese dumping areas and adjacent regions. However, for more complete and comprehensive understanding of radionuclide behavior in marine environment, their vertical and horizontal transport, accumulation and redistribution in bottom sediment, available data of observations are insufficient and further regular investigations need to be done.

To implement more detailed study of radionuclide distribution in the Japan Sea, FERHRI and JAERI in general agreed to conduct a few joint expeditions aboard FERHRI R/Vs in 1999-2001. Partner Agreement on Project 1389-99 has been signed at the end of March 1999 by representatives of the International Science and Technology Center (ISTC), Far Eastern Regional Hydrometeorological Research Institute (FERHRI) and Japan Atomic Energy Research Institute (JAERI). From the Russian side, specialists of Moscow State Engineering Physics Institute (MEPhI) actively participated in Project implementation.

Study Area and Program of Observations

Location of sampling stations is shown on Fig. 1, description of observations carried out at each station is given in Table 1.

The program of the first cruise included large-volume water sampling at different depths and bottom sediment sampling in the Japan Sea. Water samples from near-bottom layer (100-150 m over the bottom) and intermediate layers (200, 1000, 2000 m) were taken by special large-volume water sampler. Surface water samples were taken by submersible pumps and hoses.

Bottom sediment samples were taken by the Petersen grab. The surface layer (about 3 cm) and residual subsurface layer of bottom sediments were used to measure radioactivity separately. Water and bottom sediment samples were preserved as required (when necessary) and stored for further analysis in shore laboratories. Techniques used for radionuclide activity measurements in shore laboratories were already agreed during implementation of joint Japanese-Korean-Russian joint expedition (Joint Expedition, 1995; Joint Expedition, 1997). To investigate radionuclide transport, associated oceanographic observations were implemented: CTD casts, analysis of dissolved oxygen and nutrients, deployment of mooring with current meters, deployment of PALACE drifters. Meteorological observations were also carried out. Preliminary report was distributed immediately after the cruise (Preliminary Report, 1999).

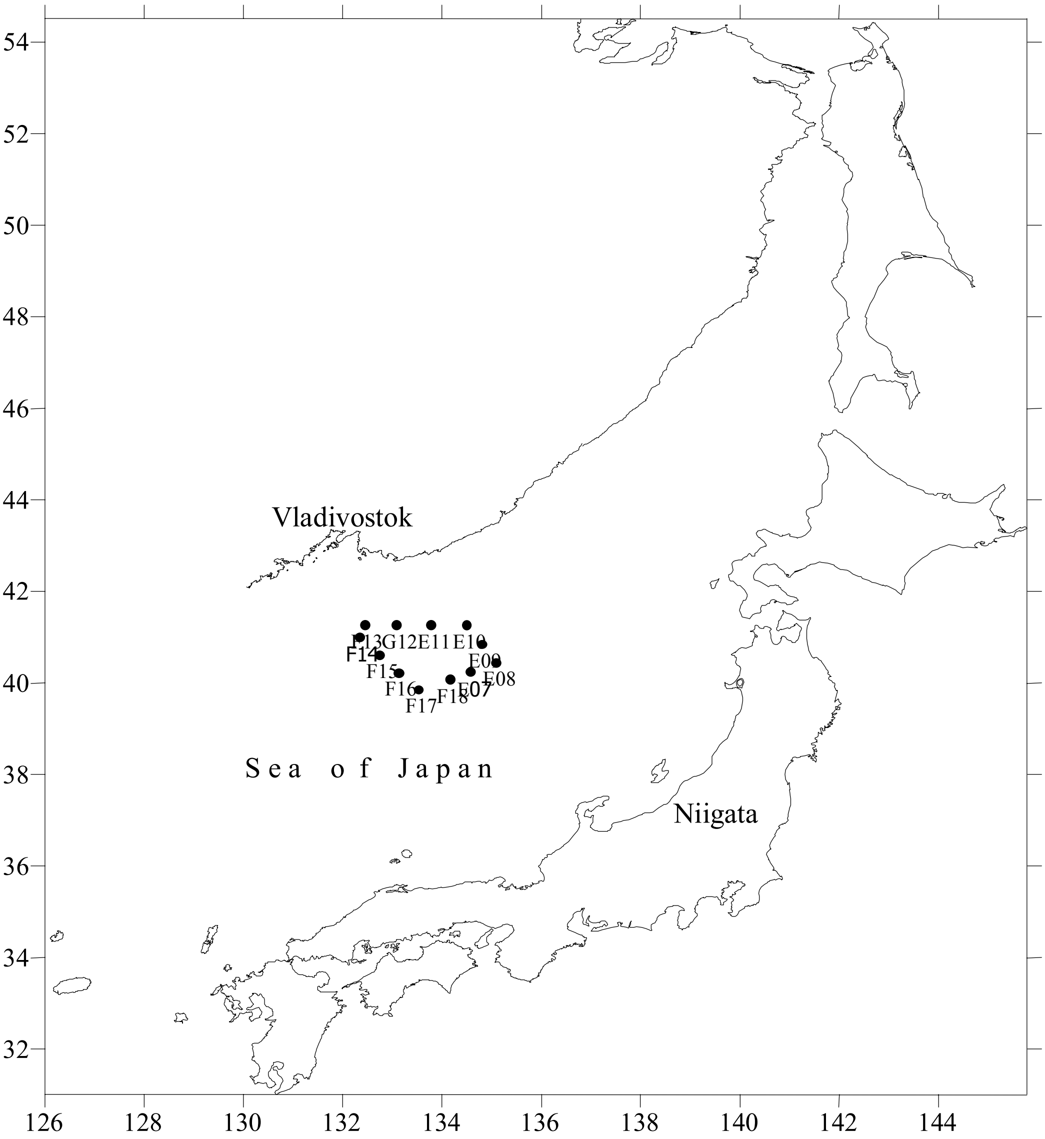

Fig. 1. Location of sampling stations, May 1999

Materials and Methods

Radionuclides in Seawater.

Co-precipitation of ^{137}Cs from seawater (100 liters) was carried out with potassium ferrocyanide and ^{90}Sr with sodium carbonate, determination of carrier yield was performed by atomic absorption spectrometry. Ferrocyanide precipitate was heated to 400 °C before gamma spectrometry using a Ge-Li detector. After 14 days, the activity of ^{90}Y produced from ^{90}Sr was measured with a low background beta-ray detector. Seawater samples (100 liters) for $^{239,240}Pu$ determination were preserved by concentrated hydrochloric acid until analysis on land. In shore laboratory, Pu was co-precipitated with iron hydroxide and purified on an ion exchange resin column. After electrodeposition of Pu on a stainless steel disk, its activity was measured on alpha spectrometer ORTEC 576-450RH using silicon semiconductor detector.

Radionuclides in Bottom Sediments

Sediment samples were dried and reduced to a powder. Then the radionuclides were measured by gamma spectrometry with a Ge-Li detector. ^{90}Sr was extracted from the powder into an acid solution. Activity of ^{90}Y produced from ^{90}Sr was measured with a low background beta-ray detector. Pu was extracted from the powder by hydrochloric acid and then purified on an ion exchange resin column. After electrodeposition of Pu on a stainless steel disk, its activity was measured on alpha spectrometer ORTEC 576-450RH using silicon semiconductor detector.

Table 1

Observations in the Japan Sea, May 1999

Station	Latitude, N	Longitude, E	Depth, m	Date	Observations
T00	40°15.0'	134°35.0'	1400	May 24	LVS/SED/SWS/CTD
F13	41°15.0'	132°21.0'	3420	May 25	Mooring deployment
F13	41°15.0'	132°27.0'	3430	May 25	LVS/SED/SWS/PD
E10	41°15.0'	134°29'.6	3560	May 27	LVS/SED/SWS/PD
E10	41°15.0'	134°29'.6	3560	May 28	CTD
E11	41°15.0'	133°47.0'	3520	May 29	CTD
G12	41°15.0'	133°05.0'	3480	May 29	CTD
F13	41°15.0'	132°27.0'	3430	May 29	CTD
F14	41°00.0'	132°21.0'	3420	May 29	CTD
F15	40°36.0'	132°45.0'	3430	May 30	CTD
F16	40°13.0'	133°08.0'	3310	May 30	CTD
F17	39°50.0'	133°32.0'	1260	May 31	CTD
F18	40°04.0'	134°10.0'	560	May 31	CTD
E07	40°14.0'	134°35.0'	840	May 31	CTD
E08	40°26.0'	135°06.0'	2540	May 31	CTD
E09	40°50.0'	134°49.0'	3340	May 31	CTD

T00 – test station
PD – PALACE deployment
LVS – large volume water sampling
SWS – surface water sampling
SED – sediment sampling
CTD – CTD cast

Gamma Spectrometry

Gamma spectrometric system consisted of Ge-Li detector (80 cm^3) with stainless steel shield (150 mm) and Russian multichannel analyzer AI-1024-95-17 with personal computer IBM PC. Gamma spectra were processed by BALTISPEKTR software. Peak resolution of gamma spectrometer at 1332 KeV (^{60}Co) was 3.8 KeV. Measurement time varied from 500 to 1200 min to allow detection limit of ^{137}Cs of about 0.7-1.1 Bq/kg (or Bq/m^3). Measurements were performed in plastic containers (150 cm^3 for bottom sediment samples and 14 cm^3 for samples of precipitate from seawater). Efficiency of gamma spectrometer has been checked with Russian standard radioactivity sources and IAEA-135 reference material.

Results and Discussion

Radionuclides in Seawater

Data on activities of radionuclides in seawater at different depths are presented in Table 2 for two stations: F-13 and E-10. Activities of ^{137}Cs and ^{90}Sr decreased with depth and became less then detection limit below 2000 m, ^{137}Cs/^{90}Sr ratio varied from 1.43 to 1.77 (global fallout value is 1.57). It's necessary to note that preliminary onboard measurements of ^{137}Cs activities in sorbents have given slightly higher values: 3.0-3.1 Bq/m^3.

Well-known feature of vertical distribution of ^{239,240}Pu activities is the subsurface maximum between 400 and 1000 m depth (*e.g.*, Nagaya & Nakamura, 1984; 1987; Hirose *et al.*, 1999). Data from Table 2 demonstrate this subsurface maximum at both stations. Elevated ^{239,240}Pu activity at the surface at station E-10 is probably a reflection of winter convection to the north of Polar front (Hirose *et al.*, 1999).

The results on ^{137}Cs activities in seawater obtained during the 1999 expedition are presented in Table 3 along with the data of Japanese-Korean-Russian joint expedition, 1994 and 1995, for the Japan Sea (median values). The results of independent measurements of Japanese (Miyao *et al.*, 1998) and Korean (Kang *et al.*, 1997) researchers are also given for comparison. The data for May 1999 are equal or below values measured in 1995, 1994 or earlier.

Similar comparative data for ^{90}Sr are given in Table 4. Again, 1999 data are equal or below the results of 1994 and 1995 cruises. It should be taken into account that geographically the study area in May 1999 is closer to the area of observations of 1994 cruise (about 41-42°N). In 1995, observations were carried out in the southern part of the Japan Sea (about 37°N).

Table 2

Activities of some radionuclides in seawater of the Japan Sea, May 1999

Depth	^{137}Cs, Bq/m^3	^{90}Sr, Bq/m^3	^{239,240}Pu, mBq/m^3
Station F-13			
Surface	2.44±0.40	1.38±0.33	22.5±2.7
200 m	2.15±0.32	1.50±0.38	22.6±4.2
1000 m	1.77±0.51	1.14±0.22	30.9±5.0
2000 m	0.31±0.10	<0.49	25.7±3.4
3350 m	<0.69	<0.47	32.6±2.9
Station E-10			
Surface	2.38±0.32	1.44±0.22	44.0±4.0
200 m	2.22±0.44	1.50±0.23	19.4±4.0
1000 m	1.57±0.27	1.04±0.23	44.0±4.0
2000 m	<0.69	<0.34	38.6±5.7
3480 m	<0.69	<0.33	21.9±3.8

Note**:** the errors given are ±1σ counting statistics.

Table 3

Activities of ^{137}Cs (Bq/m^3) in seawater of the Japan Sea

Period of observations (number of stations)	Surface waters	Bottom waters
1999 (2)	2.4	<0.7
1995 (2)	2.5-2.9	1.1-1.3
1994 (9)	2.8-3.6	0.6-2.0
1993 (19)*	2.6-3.3	–
1986-1993 (6)**	2.7-9.9	–

*Kang *et al*., 1997
**Miyao *et al*., 1998

Table 4

Activities of ^{90}Sr (Bq/m^3) in seawater of the Japan Sea

Period of observations (number of stations)	Surface waters	Bottom waters
1999 (2)	1.4	<0.5
1995 (2)	2.0	0.4-0.7
1994 (9)	1.6-2.0	0.4-1.2

Data on activities of ^{239,240}Pu in seawater measured in May 1999 expedition are presented in Table 5 along with the results of Japanese-Korean-Russian joint expedition, 1994 and 1995 (median values). The results of independent measurements of Korean (Kang *et al*., 1997) and Japanese (Miyao *et al*., 1998; Yamada *et al*., 1996) researchers are also given for comparison. The data for May 1999 are close to values measured previously in the Japan Sea. As it was already mentioned, elevated activity of ^{239,240}Pu at the surface at station E-10 is connected probably with winter convection to the north of polar front (Hirose *et al*., 1999).

Radionuclides in Bottom Sediments

The results of activity measurements of some natural and anthropogenic radionuclides in surface and subsurface bottom sediments are shown in Table 6 for two stations, F-13 and E-10. Activities of natural radionuclides are typical for marine sediments, similar values of ^{40}K (590-684 Bq/kg), ^{226}Ra (28.1-37.8 Bq/kg), ^{232}Th (43.1-51.8 Bq/kg) and ^{238}U (38.8-53.7 Bq/kg) were observed, for example, in the South-China Sea (Yu *et al*., 1994).

Table 5

Activities of $^{239,240}Pu$ (mBq/m^3) in seawater of the Japan Sea

Period of observations (number of stations)	Surface waters	Bottom waters
1999 (2)	22.5-44.0	21.9-32.6
1995 (2)	8.6-16.0	25.0-27.0
1994 (9)	8.0-25.0	15.0-29.0
1993 (3)*	6.0-10.0	–
1993 (2)**	7.4-9.5	26.1-33.0
1986-1994 (10)***	1.3-14.0	–

*Kang *et al*., 1997
**Yamada *et al*., 1996
***Miyao *et al*., 1998

Table 6

Activities of some radionuclides (Bq/kg) in bottom sediments of the Japan Sea, May 1999

Radionuclides	Station F-13		Station E-10	
	Surface sediment (0-3 cm)	Subsurface sediment (> 3 cm)	Surface sediment (0-3 cm)	Subsurface sediment (> 3 cm)
^{137}Cs	1.60±0.53	<1.1	1.27±0.92	<1.0
^{90}Sr	<0.3	<0.3	<0.3	<0.3
$^{239,240}Pu$	0.32±0.06	0.003±0.001	0.19±0.05	<0.003
^{40}K	600±20	560±30	660±30	580±30
^{226}Ra	32±5	28±7	42±8	27±6
^{232}Th	42±5	38±6	47±6	35±5
^{238}U	34±31	113±29	69±31	73±30

^{134}Cs in bottom sediments was not detected (from <0.14 to <0.32 Bq/kg) as well as ^{60}Co (from <0.17 to <0.21 Bq/kg). ^{90}Sr activities in bottom sediments in May 1999 were below detection limit, activities of ^{137}Cs were also extremely low, especially in subsurface layer of bottom sediments. As in the case with seawater, preliminary onboard measurements of ^{137}Cs in bottom sediments have given a little higher results: from <2.0 to 3.0 Bq/kg.

The results on ^{137}Cs activities in bottom sediments in May 1999 are presented in Table 7 along with the data of previous joint cruises, 1994 and 1995, for the Japan Sea (median values). Data of Korean researchers (Lee *et al*., 1998) are given for comparison. The data of 1999 expedition are well below the values measured before. Similar activities of ^{137}Cs in bottom sediments (1.52-3.11 Bq/kg) were observed in Hong Kong (Yu *et al*., 1994) as well as in the Yellow and the East China Seas (0.35-6.11 Bq/kg; Nagaya & Nakamura, 1992).

Comparison of 1999 data with the previous results on ^{90}Sr is shown in Table 8. May 1999 data are fairly similar to 1994-1995 results.

The results on $^{239,240}Pu$ activities in bottom sediments in May 1999 are presented in Table 9 along with the data of previous joint expeditions, 1994 and 1995 (median values). Data of Korean researchers (Lee *et al*., 1998) are given for comparison. The data of 1999 expedition are very similar to the values measured before. Similar activities of $^{239,240}Pu$ (0.107-0.467 Bq/kg) were observed in bottom sediments in the Yellow and the East China Seas (Nagaya & Nakamura, 1992).

Table 7

Activities of ^{137}Cs (Bq/kg) in bottom sediments of the Japan Sea

Period of observations (number of stations)	Surface sediment	Subsurface sediment
1999 (2)	1.3-1.6	<1.1
1995 (2)*	7.2	7.2
1994 (9)	1.0-2.8	<2.3
1995 (2)**	11.0-13.1	0.9-1.8

* at the station BG4 sediment sample was not divided to surface and subsurface;
** Lee *et al*., 1998, 0-2 cm and 8-10 cm layers.

Table 8

Activities of ^{90}Sr (Bq/kg) in bottom sediments of the Japan Sea

Period of observations (number of stations)	Surface sediment	Subsurface sediment
1999 (2)	<0.3	<0.3
1995 (2)*	0.2-0.8	0.2-0.8
1994 (9)	0.1-0.2	<0.3

* at the station BG4 sediment sample was not divided to surface and subsurface.

Table 9

Activities of ^{239,240}Pu (Bq/kg) in bottom sediments of the Japan Sea

Period of observations (number of stations)	Surface sediment	Subsurface sediment
1999 (2)	0.19-0.32	<0.01
1995 (2)*	0.33-1.80	0.13-1.33
1994 (9)	0.01-1.03	<0.01-0.76
1995 (2)**	2.00-3.73	0.13-0.55

* at the station BG4 sediment sample was not divided to surface and subsurface;
** Lee *et al*., 1998, 0-2 cm and 8-10 cm layers.

Conclusion

In May 1999, joint Japanese-Russian expedition to study radionuclide distribution in the Japan Sea was successfully implemented aboard FERHRI R/V "Professor Khromov". During the expedition, specialists from FERHRI, MEPhI and JAERI performed 12 CTD casts with measurements of temperature, salinity, dissolved oxygen and nutrients from the surface to the bottom, deployed two PALACE drifters and one mooring with three current meters, took samples of seawater from the surface, bottom layer and three intermediate layers (200, 1000 and 2000 m), and took samples of surface and subsurface sediments at two stations. Activities of gamma emitters, ^{90}Sr and ^{239,240}Pu were measured in samples of seawater and bottom sediments. Measured activities were low and caused by global atmospheric fallout of radionuclides. Data obtained in 1999 cruise are quite close to the results of previous expeditions. For better understanding of radionuclide inputs, distribution, transport and possible effects, further studies in the Japan Sea are necessary.

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