Position of Sediments in Transfer of Radionuclides Released into Coastal Sea to Human Beings

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A great portion of radionuclides released into coastal sea is adsorbed into marine sediments and the adsorbed radionuclides causes the radioactive contamination of marine organisms and then transferred to human beings who consume them. In order to make a quantitative evaluation of this route, the transfer of 9 kinds of radionuclides from sediments to benthic organisms such as algae, bivalve and worm was observed. Then it was compared with the radioactivity in these organisms from radioactively contaminated sea water (Concentration factor). It was observed that the influence of sea water was larger than that of sediments as it was 10^4 times larger for ^{54}Mn , 10^3 times larger for ^{59}Fe , ^{60}Co , ^{95}Zr - ^{95}Nb , ^{106}Ru - ^{106}Rh and ^{144}Ce - ^{144}Pr , 10^2 times larger for ^{65}Zn and 10 - 10^2 times larger for $^{115\text{m}}\text{Cd}$ and ^{137}Cs .

Consequently, sea water can be considered as the main route and sediments as the secondary in the case of quantitative comparison of the effect on the accumulation of radionuclides by marine organisms and in the transfer of radionuclides to human beings.

It is well known that marine sediments accumulate radionuclides as much as toxic metals and there are several reports on the sediments-biota interaction.¹⁻⁷⁾ But it is still not clear the position of sediments in the transfer process of radionuclides from nuclear facilities to human beings via marine products.

The authors have been studied this problem by obtaining 1) transfer of radionuclides from contaminated sediments to marine organisms such as algae, bivalve and worm which are benthic and closely contact with sediments and 2) accumulation of radionuclides by marine organisms from sea water. The influences of sediments and sea water on radioactive contamination of organisms were estimated by comparing 1) and 2). Several studies on ^{60}Co , ^{95}Zr - ^{95}Nb , ^{106}Ru - ^{106}Rh , $^{115\text{m}}\text{Cd}$ and ^{137}Cs have been reported previously.⁸⁻¹²⁾ In this paper, study on ^{54}Mn , ^{59}Fe , ^{65}Zn and ^{144}Ce - ^{144}Pr is mainly reported together with summary of the former 5 radionuclides in the previous reports by us.

Materials and Methods

Radionuclides used were the following 9 kinds: $^{54}\text{Mn}(\text{MnCl}_2)$, $^{59}\text{Fe}(\text{FeCl}_3)$, $^{60}\text{Co}(\text{CoCl}_2)$, $^{65}\text{Zn}(\text{ZnCl}_2)$, ^{95}Zr - $^{95}\text{Nb}[\text{Zr}(\text{COO})_2]$, ^{106}Ru - ^{106}Rh (nitrosylruthenium nitrate complexes), $^{115\text{m}}\text{Cd}$

(CdCl_2) , $^{137}\text{Cs}(\text{CsCl})$ and ^{144}Ce - $^{144}\text{Pr}(\text{CeCl}_3)$, and with the exception of the specific activity of 93 mCi/mg Co for ^{60}Co and 0.12 mCi/mg Cd for $^{115\text{m}}\text{Cd}$, the other radionuclides were carrier-free.

All experiments were performed at room temperature.

Experiment 1. Transfer of radionuclides from sediments to organisms (Transfer ratio).

The sediments were sieved immediately after sampling from the intertidal zone of the seashore around Ajigaura in Nakaminato city and the 0.1-0.5 mm diameter portion was used. The portion accounts for 90% of the sediments. Sea water was used after filtration with a filter paper (Toyo, No. 5C).

To obtain contaminated sediments, 100 g of sediments and 300 ml of sea water were placed in 1 l polyethylene bottle, each radionuclide (0.2 mCi) added separately and shaken horizontally at a rate of 100 times per min with aeration. After exposure for one week, sediments were washed 3 times with non-radioactive sea water by decantation and the radioactivity was measured with a whole body counter (Armac Liquid Scintillation Counter, Model 446, Packard).

Eighty grams of the contaminated sediments and 800 ml of fresh sea water (quantity ratio 1:10) were placed in 5 l capped acrylic tank and or-

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ganisms were reared in the tank with aeration. The radioactivity in organisms was measured with the whole body counter. Three bivalves (*Gomphina melanaegis*, average body weight: 30 g) were dissected and the soft parts were submitted to the radioactivity measurement. In the case of marine algae (*Chondrus* sp., *Hijikia fusiforme*, *Sargassum thunbergii*, *Ahnfeltia* sp. and *Gracilaria verrucosa*), about 4 g were reared for each radionuclide. A portion of algae was taken on the day of the measurement and washed for 2 min with non-radioactive sea water. After removing sea water on the surface of algae with filter paper, the radioactivity of algae was measured with the

whole body counter.

The ratio of radioactivity in organisms to the initial radioactivity in sediments was expressed as "Transfer ratio".

Experiment 2. Accumulation of radionulides by marine organisms from sea water (Concentration factor).

Each 2 μCi of the radionuclides was separately added to 5 l of sea water in acrylic tank and aerated. The radioactivity in sea water was kept at the steady level by the addition of radionuclide. Bivalves and algae mentioned above were reared in the tank and their radioactivity was measured

Fig. 1. Transfer of radionuclides from sediments to algae.

○—○ *Sargassum thunbergii*, △—△ *Gracilaria verrucosa*, ●—● *Chondrus* sp., ▲—▲ *Ahnfeltia* sp. ×—× *Hijikia fusiforme*.

with the whole body counter. Measurement of sea water was carried out by taking 2 ml in a polyethylene test tube with the whole body counter. The ratio of the radioactivity in organisms to that in sea water was presented as concentration factor.

For the calculation of concentration factor and transfer ratio, the values of organisms on the 14th day were used in Experiment 1 and 2.

To express the comparative influence of sediments and sea water on the accumulation of radionuclide by organisms, BFS (Biological Factor of the Sediments) is used here dividing the concentration factor with the transfer ratio.

Results

Experiment 1. Transfer of radionuclides from sediments to organisms (Transfer ratio).

Fig. 1 shows the transfer of radionuclides from sediments to algae (Transfer ratio). With the exception of ^{65}Zn , the radioactivity in *Hijikia fusiforme* and *Chondrus* sp. is low, while that in *Ahnfeltia* sp. and *Sargassum thunbergii* indicates high values. The transfer of radionuclides from sediments to bivalves is shown in Fig. 2 as transfer ratio. The transfer ratios of bivalve are in the range 10^{-2} to 10^{-3} and these are much lower than those of algae.

Experiment 2. Accumulation of radionuclides by marine organisms from sea water (Concentration factor).

The accumulation of radionuclides from sea water by algae is shown in Fig. 3 as concentration factor. The accumulation by *Hijikia fusiforme* was

lowest in the algae. *Sargassum thunbergii* indicated the largest concentration factor with the exception of ^{65}Zn and ^{144}Ce - ^{144}Pr . The concentration factors for bivalve are shown in Fig. 4. The concentration factor of ^{60}Co was larger than those of ^{144}Ce - ^{144}Pr and ^{95}Zr - ^{95}Nb . The concentration factors for bivalve were lower than those for algae.

Discussion

The results from Experiment 1 and 2 are summarized in Table 1, accompanying previous data by us.¹⁰⁻¹²⁾ The transfer ratio of radionuclides is in a range of 0.857 for ^{65}Zn in the case of *Sargassum thunbergii* and 0.001 for ^{106}Ru - ^{106}Rh in the case of *Gomphina melanaegis* and the concentration factor is in a range from 2680 (*Gracilaria verrucosa* for ^{144}Ce - ^{144}Pr) to 2 (*Gomphina melanaegis* for ^{106}Ru - ^{106}Rh). The BFS values are in the order of 10^4 for ^{54}Mn , 10^3 for ^{59}Fe , ^{95}Zr - ^{95}Nb , ^{144}Ce - ^{144}Pr and ^{106}Ru - ^{106}Rh , 10^2 for ^{65}Zn and $^{115\text{m}}\text{Cd}$, 10 - 10^2 for ^{137}Cs and 10^2 - 10^4 for ^{60}Co . From these figures, it could be said that sea water gives organisms 10^4 times higher radioactivity than sediments in the accumulation of ^{54}Mn by organisms and 10 - 10^2 times higher radioactivity of ^{137}Cs when the radioactivity in sediments is the same level as in sea water. The quantity ratio of sediments and sea water in Experiment 1 is 1:10 but the transfer ratio differs when this ratio is changed. In the previous report by us,¹¹⁾ the transfer ratio decreased to about 1/3 when the quantity ratio was changed from 1:100 to 1:400. If a radionuclide is adsorbed to 2 cm depth of surface layer of the sediments and the specific gravity of the sediments is taken as 2, the ratio of 1:10, 1:100 and 1:400 indicates water depth of 0.4, 4 and 16 meter, respectively. In this experiment, the quantity ratio corresponded to the shallow sea with 0.4 m depth was used to estimate the influence of sediments as much as possible. Nevertheless, concentration factors are larger than transfer ratios. This means that sea water affects the accumulation of radionuclides by organisms more than sediments when the radioactivity in sea water is equal to that in sediments. But the authors previously reported that ^{60}Co in *Sargassum fluvellum* came from radioactive sediments,¹³⁾ and Seymour and Nelson¹⁴⁾ detected ^{65}Zn in mussel after shutdown of nuclear generator. Therefore, the possibility of contamination of organisms by sediments could not be neglected when the radioactivity in sea water is

Fig. 2. Transfer of radionuclides from sediments to bivalve *Gomphina melanaegis*.

Fig. 3. Accumulation of radionuclides from sea water by algae.

○—○ *Sargassum thunbergii*, △—△ *Gracilaria verrucosa*, ●—● *Chondrus* sp.,
▲—▲ *Ahnfeltia* sp. ×—× *Hijikia fusiforme*

Fig. 4. Accumulation of radionuclides from sea water by bivalve *Gomphina melanaegis*.

reduced. However, in general, it could be said that sea water affects the accumulation of radionuclides more than sediments.

In conclusion, the flux of radionuclides released into coastal sea can be considered as shown in Fig. 5.

I) Radionuclide along route A considerably differs depending on whether the topography of the coast is of the closed type or the open type facing the ocean.

II) Another portion remains in the coastal sea water and causes the radioactive contamination of organisms depending on concentration factors (route B).

Table 1. Accumulation of radionuclides from sediments and sea water by marine organisms

Radionuclide	(A) Initial concentration in sediments (cpm/g)		Organism	(B) Concentration in organisms (cpm/g)	(C) Transfer ratio (B/A)	(D) Concentration factor	(E) BFS (D/C)
⁵⁴ Mn	1142000	Algae	<i>Chondrus</i> sp.	30280	0.027	1320	48890
			<i>Hijikia fusiforme</i>	26260	0.023	240	10400
			<i>Sargassum thunbergii</i>	184400	0.162	2410	15700
			<i>Ahnfeltia</i> sp.	52500	0.046	720	15700
			<i>Gracilaria verrucosa</i>	130200	0.114	1970	17300
		Bivalve	<i>Gomphina melanaegis</i>	7050	0.006	150	25000
⁵⁹ Fe	284000	Algae	<i>Chondrus</i> sp.	7800	0.027	150	5560
			<i>Hijikia fusiforme</i>	2600	0.009	20	2220
			<i>Sargassum thunbergii</i>	21700	0.076	500	6580
			<i>Ahnfeltia</i> sp.	45600	0.161	340	2110
			<i>Gracilaria verrucosa</i>	17500	0.062	240	3870
		Bivalve	<i>Gomphina melanaegis</i>	560	0.002	100	5000
⁶⁰ Co	1907000	Algae	<i>Chondrus</i> sp.	61500	0.032	370	12330
			<i>Hijikia fusiforme</i>	62900	0.033	40	1210
			<i>Sargassum thunbergii</i>	25490	0.134	330	2460
			<i>Ahnfeltia</i> sp.	63800	0.033	120	3460
			<i>Gracilaria verrucosa</i>	83900	0.044	110	2500
		Bivalve	<i>Gomphina melanaegis</i>	13000	0.007	110	15710
		Worm	<i>Nereis japonica</i>	95400	0.050	6	120
		⁶⁵ Zn	152000	Algae	<i>Chondrus</i> sp.	84800	0.558
<i>Hijikia fusiforme</i>	5500				0.036	20	560
<i>Sargassum thunbergii</i>	130300				0.857	120	140
<i>Ahnfeltia</i> sp.	126200				0.830	130	160
<i>Gracilaria verrucosa</i>	64400				0.424	170	400
Bivalve	<i>Gomphina melanaegis</i>			6100	0.040	20	500
⁹⁵ Zr- ⁹⁵ Nb*	1118000	Algae	<i>Chondrus</i> sp.	73000	0.065	410	6200
			<i>Hijikia fusiforme</i>	86700	0.078	230	2950
			<i>Sargassum thunbergii</i>	521500	0.466	1390	2980
			<i>Ahnfeltia</i> sp.	419900	0.376	760	2020
			<i>Gracilaria verrucosa</i>	183400	0.164	870	5300
		Bivalve	<i>Gomphina melanaegis</i>	1900	0.002	5	2500
		Worm	<i>Nereis japonica</i>	10100	0.009	4	440
		¹⁰⁶ Ru- ¹⁰⁶ Rh*	1095000	Algae	<i>Cyrtymenia</i> sp.	59000	0.054
Bivalve	<i>Gomphina melanaegis</i>			820	0.001	2	2000
Worm	<i>Nereis japonica</i>			65700	0.060	6	1000
^{115m} Cd*	18000	Worm	<i>Nereis japonica</i>	2200	0.122	22	180
¹³⁷ Cs*	1182000	Algae	<i>Cyrtymenia</i> sp.	81000	0.069	5	70
		Bivalve	<i>Gomphina melanaegis</i>	53000	0.045	7	160
		Worm	<i>Nereis japonica</i>	211600	0.179	6	40
¹⁴⁴ Ce- ¹⁴⁴ Pr*	1617000	Algae	<i>Chondrus</i> sp.	62700	0.039	420	10800
			<i>Hijikia fusiforme</i>	34000	0.021	40	1900
			<i>Sargassum thunbergii</i>	395800	0.245	910	3700
			<i>Ahnfeltia</i> sp.	299600	0.185	1230	6650
			<i>Gracilaria verrucosa</i>	577000	0.357	2680	7500
		Bivalve	<i>Gomphina melanaegis</i>	4400	0.003	20	6670

*: cited from the previous reports.¹⁰⁻¹²⁾*Chondrus* sp.: Tsunomata, *Hijikia fusiforme*: Hijiki, *Sargassum thunbergii*: Umitoranoo, *Ahnfeltia* sp.: Harigane, *Gracilaria verrucosa*: Ogonori, *Gomphina melanaegis*: Kotanagai, *Nereis japonica*: Gokai, *Cyrtymenia* sp.: Hijirimen.

Fig. 5. Flux of radionuclides released into coastal sea.

III) The balance is adsorbed to sediments and radionuclides recycled from sediments causes the radioactive contamination of organisms (route C).

In the transfer of radionuclides to human beings via marine organisms, the primary is route B and the secondary is route C, although sediments adsorb a great portion of radionuclides released into coastal sea.^{8,9)}

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