Radioecology of $^{60}$Co in Urazoko Bay:
Correlation between Levels of $^{60}$Co in Sargassoes and Marine Sediments

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In order to know factors which influence $^{60}$Co contamination of sargassoes in Urazoko Bay, where a nuclear power plant is located, correlations between the contaminations of the algae and of the marine environment were examined, based on field data observed since January 1971.

It was found that: [1] correlation between the $^{60}$Co concentration in sargassoes ($Y_{algae}$ Ci/kg raw weight) and distance from discharge outlet of the nuclear power plant to the sampling points ($d$ km) was expressed by a function, $Y_{algae} = C \cdot \exp(-kd)$, where $C$ and $k$ are constants, and $k$ increased successively from 0.388 in Nov. 1971 to 1.200 in Nov. 1974, whereas $C$ decreased from 120 to 27 during the same period. [2] $^{60}$Co content in the sargassoes seemed to be contributed dominantly by the dislodged $^{60}$Co from heavily contaminated sediment rather than freshly discharged $^{60}$Co from the power plant. The amount of dislodged $^{60}$Co was estimated from both the $^{60}$Co distribution in marine sediment in Urazoko Bay and results of a tracer experiment concerning the dislodgement of $^{60}$Co from marine sediment.

INTRODUCTION

Tsuruga Nuclear Power Plant (BWR-type, 357 MWe) at Fukui Prefecture is a contributor of artificial radionuclides to Urazoko Bay, located along the west coast of Tsuruga Bay. The production of artificial radionuclides by this power plant makes it possible to use Urazoko Bay and also a portion of Tsuruga Bay as an excellent field for the radioecological study of enclosed coastal marine environment. The tidal range in Urazoko Bay is small as elsewhere on the Japan Sea coast. Fishery production in Urazoko Bay are mainly jack mackerel, flathead and sand smelt as fish, octopus and mussel as molluscus but these catches are extremely small. A sargasso (sargassum fulvellum, AGARDH) is one of the most abundant seaweeds in Urazoko Bay, grows in 15-20 cm in length about the middle of November and withers to flow out in the summer of the next year. In this article, correlation between the concentration of $^{60}$Co in this alga, a non-edible seaweed but a good indicator of radioactive contamination in Urazoko Bay, and the environment, which was examined in the view-
points of; (1) distance from discharge outlet of the power plant to sampling points and (2) amount of $^{60}$Co discharged by the power plant, based on both the field data observed and a laboratory tracer experiment.

\[
\begin{align*}
\text{Ash} & \quad 10 \text{ g} \\
+ & \quad \text{Co}^{++} \text{ carrier} 5 \text{ mg} \\
+ & \quad \text{Conc.} \text{HCl} 40 \text{ ml} \\
\text{dry up on sand bath} & \\
+2\text{HCl} & \text{dissolve} \\
\text{filtration} & \\
\text{Res.} & \\
+& \quad \text{H}_2\text{O}_2 \quad 3 \text{ ml} \\
+& \quad \text{HNO}_3 \quad 10 \text{ ml} \\
\text{dry up} & \\
+2\text{HCl} & \quad 20 \text{ ml} \\
\text{filtration} & \text{Res.} & \text{Filt.} \\
\text{Filt.} & & \\
& \quad \text{2 times repeat} \\
& \quad \text{dry up} \\
& \quad +8\text{HCl} \text{ filtration} \\
& \quad \text{Ton exchange} & \text{(Dowex 1x8 100-200 mesh,} \\
& & \text{column; 2 cm\(\phi\), 15 cm height)} \\
& & \begin{align*}
8\text{HCl} & \quad 50 \text{ ml} \\
6\text{HCl} & \quad 50 \text{ ml} \\
4\text{HCl} & \text{ Co elute} \\
\text{dry up on sand bath} & \\
+8\text{HCl} & \quad 20 \text{ ml} \\
\text{Ton exchange} & \text{(Dowex 1x8 100-200 mesh,} \\
& & \text{column; 2 cm\(\phi\), 10 cm height)} \\
& & \begin{align*}
8\text{HCl} & \quad 30 \text{ ml} \\
6\text{HCl} & \quad 30 \text{ ml} \\
4\text{HCl} & \text{ Co elute} \\
\text{dry up on sand bath} & \\
+\text{H}_2\text{O} & \quad 20 \text{ ml} \\
\text{filtration} & \text{Res.} & \text{Filt.} \\
\text{Filt.} & & \\
& & \text{neutralization with 3\% Na}_2\text{CO}_3 \\
& & \text{boil} \\
& & +3\% C_6\text{H}_4(N\text{H}_2)\text{COONa} 2 \text{ ml} \\
& & \text{stir} \\
& & \text{fix on ToyoRoshi filter paper} \\
& & \text{No. 5C (2 cm\(\phi\))} \\
& & \text{dryness, weight} \\
\text{Beta emitter counting} & \\
\text{Fig. 1. Analytical procedure for } ^{60}\text{Co.} &
\end{align*}
\end{align*}
\]
MATERIALS AND METHODS

The samples of sargasso (Sargassum fulvellum) collected for radiochemical analysis were washed with fresh water, weighed after blotting off the water, and then ashed at 450°C for 48 hours in an electric muffle furnace after drying in an electric oven at 110°C for 24 hours. Ashed samples were sifted out by a chemical sieve (0.5 mmø) to remove shells and sand, and then weighed. The ratio of ashed weight to raw weight of sargasses used for this investigation was in the range of 3.8 and 7.2%.

The method of determination of $^{60}$Co was essentially the same as that reported by Tsuruga. An outline of the chemical procedure was given in Figure 1. The beta activity of $^{60}$Co was finally measured in the form of cobalt anthranilate using a low-background gas flow counter (Tracer lab., omni/guard). The chemical yield of this procedure was in the range of 76-83%, and the counting efficiency was 15-19%.

In order to obtain dislodgement rate from sediment for $^{60}$Co, a tracer experiment was performed as follows:

Each one hundred grams of sediments collected from Area I, II and III (see Fig. 5) 500 ml of sea water (filtered with Toyo Roshi filter paper No. 5C) and $1.63 \times 10^6$ cpm of $^{60}$Co ($^{60}$CoCl$_2$) were added to three silicon-coated, capped polyethylene bottles of 1000 ml in capacity, and the bottles were horizontally shaken for ten days at the rate of 100 times per minute. Each twenty grams of the contaminated sediment prepared in the process described above (Area I; $25.16 \times 10^6 \pm 2.16 \times 10^6$ cpm, Area II; $29.73 \times 10^6 \pm 3.00 \times 10^5$ cpm and Area III; $31.24 \times 10^6 \pm 5.24 \times 10^6$ cpm) was shaken with 100 ml of fresh sea water in the same manner described above. Two ml of the sea water was measured by use of a well-type scintillation counter (Kobe Industries Ltd.), and the rest of sea water was replaced by 100 ml of fresh sea water at every measuring time.

RESULTS AND DISCUSSION

Location of the sampling points is shown in Figure 2. The points A-I are in Urazoeka Bay and the points J-N in Tsuruga Bay. Concentrations of $^{60}$Co in sargasses during the period from January 1971 to May 1975 are summarized in Table 1 expressed in unit of pCi per kilogram of the raw weight of sargasses. Detection limit for $^{60}$Co is approximately 3.0 pCi/kg raw weight. The maximum concentration observed is 178.6 pCi/kg at point F in January 1971. No seasonal variation of the concentrations could be observed. When samples were obtained at three or more points in Urazoeka Bay at the same time, it could be seen from Figure 3 calculated by the least-squares method that correlation between concentration of $^{60}$Co in sargasses ($Y_{\text{algae}}$pCi/kg raw weight) and distance from discharge outlet of the plant to sampling point (d km) was expressed by an exponential function, $Y_{\text{algae}} = C \cdot \exp(-kd)$, where C and k are constants, and k increased successively from 0.388 for November 1971 to 1.200 for November 1974, whereas C decreased from 120 to 27 during the same
Fig. 2. Location of sampling points for sargassoes.
period. From this fact, it can be estimated that the difference between the $^{60}$Co concentrations in sargassoes collected at the nearer and the farther points from the discharge outlet increases gradually, if the recent discharge pattern hold out in future. The numerical value of $C$ varies mainly with the concentration factor of marine organisms and the amount of discharged $^{60}$Co from the nuclear power plant, on the other hand, the value of $k$ is influenced by the configuration of the bay, the current of sea water, the character of marine sediment and so on. The life cycle of the sargassoes is about one year as mentioned above. As it can be generally considered that the concentration of $^{60}$Co in sargassoes is dominated by the $^{60}$Co concentration in the environmental sea water, so it would be reasonably expected that there should be a correlation between the amount of discharged $^{60}$Co and the concentration of $^{60}$Co in sargassoes. In Urazoko Bay, however no correlation was observed quantitatively within yearly life cycles of sargassoes in points B, F and I as shown in Figure 4. Therefore, it may be a main reason for this anomaly that heavily contaminated marine sediments influence the concentration of $^{60}$Co in sargassoes.

Marine sediment is not only a reservoir for $^{60}$Co, but also for most radioactive elements. It is said that adsorption rate of various metal elements to marine sediment is usually fast, and the recycle or dislodgement rate from sediment is very slow. For example, marine sediments obtained from Urazoko Bay accumulated over 90% of $^{60}$Co added in sea water for ten days. It was indicated, however, that the dislodgement of the half amount of $^{60}$Co from the contaminated sediments takes more

### Table 1
The concentrations of $^{60}$Co for sargassoes in Urazoko Bay

| St. | Dist. | '71-Jan. | '71-Jul. | '71-Nov. | '72-May | '72-Aug. | '73-Aug. | '74-Mar. | '74-Jun. | '74-Nov. | '75-May |
|-----|-------|----------|----------|----------|----------|----------|----------|----------|----------|----------|---------|
| A   | 0.5   | 27.1     |          |          |          |          |          |          |          |          |         |
| B   | 0.5   | 106.9    | 54.1     | 35.5     | 21.5     | 12.6     | 8.3      |          |          |          |         |
| C   | 0.7   |          | 29.6     | 22.0     | 15.2     |          |          |          |          |          |         |
| D   | 1.0   |          | 12.0     | 19.5     | 13.3     | 6.8      |          |          |          |          |         |
| E   | 1.3   | 13.1     | 29.8     | 8.4      | 7.7      |          |          |          |          |          |         |
| F   | 1.6   | 178.6    | 60.1     | 44.9     | 12.8     | 9.9      | 19.2     | 25.7     | 7.7      | 3.4      | 7.6     |
| G   | 1.8   |          |          |          |          |          |          |          |          |          | *       |
| H   | 1.9   |          |          |          |          |          |          |          |          |          | 8.3     | 6.5     | *
| I   | 2.0   |          |          |          |          |          |          |          |          |          | *       | 9.9     | *
| J   | 3.3   |          |          |          |          |          |          |          |          |          | *       | 4.3     | *
| K   | 3.4   |          |          |          |          |          |          |          |          |          | *       |         | *
| L   | 3.8   |          |          |          |          |          |          |          |          |          | 6.4     | 38.2    | 9.6     | 5.3     | 3.8     |
| M   | 4.6   |          |          |          |          |          |          |          |          |          |          |          | 4.8     |         |         |         |
| N   | 6.0   |          |          |          |          |          |          |          |          |          |          |          |          | 6.6     |          |         |         |

* Distance from the discharge outlet (km).

* Asterisk (*) denotes the value less than detection limit.
than one year (Fig. 6). The dislodgement of $^{60}$Co from the sediment will be described in detail later.

This species of sargasso is an annual plant, so it must be at most one year that sargassoes are influenced by the $^{60}$Co dislodged from sediments. Therefore, an equation can be described as follows;

$$B_n = \alpha A_n$$

where the time of collection of sargassoes is $t=t_n$, yearly amount of $^{60}$Co dislodged from contaminated sediment before that time is $B_n$ (the amount is considered to influence the contamination of sargassoes), the rate of dislodgement is $\alpha$, and the amount of $^{60}$Co adsorbed to sediments is $A_n$. On the other hand, when start of the power plant operation is $t=0$ and the yearly amount of $^{60}$Co discharged is expressed as $Q_1$ for the first year, $Q_2$ for 2nd year and $Q_n$ for $n$ year, and the corresponding amount of $^{60}$Co adsorbed to sediments $A_1$, $A_2$, and $A_n$ may be described as:

![Graph](image_url)
\[ A_r = r^3 \cdot f' Q_1, \quad A_2 = r^2 \cdot f \cdot Q_1 e^{-2t_1} + r^2 \cdot f \cdot Q_2 \]

and

\[ A_n = r^n \cdot f \cdot Q_1 e^{-nt_1} + r^n \cdot f \cdot Q_2 e^{-nt_2} + \ldots + r^n \cdot f \cdot Q_{n-1} e^{-nt_{n-1}} + r^n \cdot f \cdot Q_n \]

\[(n=1, 2, 3, \ldots)\]

where \(r\) = the adsorption rate, \(f\) = rate of transport of discharged \(^{60}\)Co to a point and \(\lambda\) = decay constant of \(^{60}\)Co.

Therefore, from equation (1) and (2),

\[ B_n = a \cdot \beta \cdot f (Q_1 e^{-\lambda t_{n-1}} + Q_2 e^{-\lambda t_{n-2}} + \ldots + Q_{n-1} e^{-\lambda t_{n-(n-2)}} + Q_n) \]

\[(n=1, 2, 3, \ldots)\]

In order to estimate the values of \(\beta \cdot f\), a simple triangular area model of Urazoko
Bay (including a portion of Tsuruga Bay) is set up as five areas of I, II, III, IV and V as shown in Figure 5. According to Nakamura and Nagaya,\textsuperscript{9,10} the maximum concentrations of \(^{60}\text{Co}\) observed during 1969 to 1973 for Areas I–V are 1160, 720, 200, 85 and 13 pCi/kg dry weight of sediment, respectively. Therefore, assuming that all the \(^{60}\text{Co}\) discharged accumulate on the sediments in Area I–V, and the respective maximum concentrations in sediments are in proportion to the average concentrations in sediments, the distribution rates of discharged \(^{60}\text{Co}\) are 33.4\% for Area I, 29.3\% for Area II, 9.3\% for Area III, 18.7\% for Area IV and 9.3\% for Area V, respectively. These figures are considered to correspond to \(\beta\cdot f\) in each area.

In Figure 5, it could be said that approximately 70\% of \(^{60}\text{Co}\) discharged by March 1975 seemed to be retained by the sediment in Urazoko Bay (Area I, II and III) whereas the rest went out of the Bay (Area IV and V).

Total amount of gamma-emitting radionuclides discharged into Urazoko Bay is presumed to be approximately 5.7 Ci during the period from the beginning of the plant operation to March 1975, and the amount of \(^{60}\text{Co}\) in it could be estimated to be 1731 mCi.\textsuperscript{12} The amount of \(^{60}\text{Co}\) discharged into Urazoko Bay from the power plant has been almost constant after the heavy initial discharge (Table 2). As \(^{60}\text{Co}\) has 5.27 years of physical half life, 1023 mCi of \(^{60}\text{Co}\) has been adsorbed by the sediments.
in the triangular area as of March 1975, and the distribution is 342, 300, 95, 191 and 95 mCi in Area I, II, III, IV and V, respectively. From the results of the tracer experiment concerning dislodgement of $^{60}$Co from sediments, dislodgement patterns of $^{60}$Co from marine sediments of Area I, II and III are estimated as shown in Figure 6. Therefore the amounts of $^{60}$Co dislodged from sediments of Area I, II and III ($B_{I}$, $B_{II}$ and $B_{III}$) during the period from time $t_{1}$ to $t_{2}$ can be calculated by following equations: 

| Area | Equation |
|------|----------|
| I    | $B_{I} = 14.1 \left( e^{-0.00205t_{1}} - e^{-0.00205t_{2}} \right)$ |
| II   | $B_{II} = 26.2 \left( e^{-0.0056t_{1}} - e^{-0.0056t_{2}} \right)$ |
| III  | $B_{III} = 22.3 \left( e^{-0.0013t_{1}} - e^{-0.0013t_{2}} \right)$ |

Accordingly, the dislodgement rate $\alpha_{I}$ in Area I is given as follows:
\[
\alpha_1 = \frac{14.1\left(e^{-0.0020t_1} - e^{-0.0020t_2}\right)}{14.1 e^{-0.0020t_1}} = 1 - e^{-0.0020(t_2 - t_1)}.
\]

In the same manner,

in Area II, \(\alpha_{II} = 1 - e^{-0.0005(t_2 - t_1)}\)

in Area III, \(\alpha_{III} = 1 - e^{-0.0013(t_2 - t_1)}\)

When all of measured \(^{60}\)Co in sargassoes is due to \(^{60}\)Co dislodged from sediment, \(^{60}\)Co dislodged from the sediments of three areas can be calculated by equation (3) as \(t=365, 730, 1095, 1460\) and 1825 days, and the correlation between the dislodged \(^{60}\)Co and the concentrations in sargassoes is shown in Figures 7 and 8.

It was found that correlation between the concentration of \(^{60}\)Co in sargassoes \((Y_{algae}\ pCi/kg\ raw\ weight)\) and the estimated amount of dislodged \(^{60}\)Co \((X\ mCi)\) dur-
ing the respective life span of the algae was expressed by an exponential function as \( Y_{\text{algae}} = 4.0 \times e^{0.022x} \) in Area I, and \( Y_{\text{algae}} = 4.0 \times e^{0.039x} \) in Area II. In Area III, an equation could not be introduced because there were a few data less than detection limit in the \(^{60}\text{Co}\) concentrations for sargassoes collected at point I.

It can be estimated that 50-70% of the amounts of \(^{60}\text{Co}\) in sargassoes collected in 1974 and 1975 was due to the \(^{60}\text{Co}\) dislodged from sediment in which the heavy initial discharge in 1969 and 1970 adsorbed.

Fig. 8. Correlation between concentration of \(^{60}\text{Co}\) in sargassoes and estimated amounts \(^{60}\text{Co}\) dislodged from marine sediment in Area II.

Thus, with regard to the radioactive effluent from nuclear power plants and other facilities, the role of marine sediment must be sufficiently taken into account for the radioactive contamination of marine organisms.
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