

## ON THE ACCUMULATION OF RADIOACTIVE MATERIALS IN MARINE ORGANISMS ALONG THE COAST OF KOREA\*

### 2. Strontium-90 Activities in Several Edible Marine Algae

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#### ABSTRACT

Continued to the previous paper, the present investigation deals with strontium-90 activities and calcium contents among 54 samples of edible marine algae collected along the coast of Korea during September, 1973 and April, 1974.

The calcium contents are variable 2.0-17.8%, and 6.38% on an average. Most of the members investigated contain 2-4% in green and red algae, and 8-10% in brown algae. Strontium-90 activities are 0.32-0.37 pCi/l in sea-water, and 0.80-28.66 pCi<sup>90</sup>Sr/g-Ca in edible seaweeds. Among the algal phyla, they are 12.49 in green, 3.34 in brown, and 9.39 pCi<sup>90</sup>Sr/g-Ca in red algae, while they are 6.25 pCi<sup>90</sup>Sr/g-Ca on an average.

In a single species collected at the same season, the highest activities appear mostly from the eastern coast, and from the western and southern coasts, in turn. The green algae, *Capsosiphon-Enteromorpha* complex show about 3 times higher activities, 28.66 pCi<sup>90</sup>Sr/g-Ca compared with the other members showing rather higher activities, and would be an indicator plant of strontium-90 activities of the marine algae along the coast of Korea.

#### INTRODUCTION

Continued to the previous paper (Yang *et al.*, 1975), the present investigation is carried out to detect the accumulation of strontium-90 activities among the edible seaweeds along the coast of Korea. Strontium-90 activities in sea-water are also analysed in order to clarify

the concentration factors of them in the seaweeds.

Considering the problems of radiation protection, the levels of radioactivity in the edible marine algae would be also estimated from the maximum permissible concentrations recommended by International Commission on Radiological Protection.

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## MATERIALS AND METHODS

### Materials

In the present investigation, about 54 seaweed samples (24 species from 8 sampling sites) were used for the analysis of calcium contents and strontium-90 activities. The seaweed samples were collected during September, 1973 and April, 1974 from Kangnung, Samchuck and Kizang (Pusan), Zindo, Yeosu, Cheju-island, Kunsan, and Anmyundo along the coast of Korea. The sampling dates, sites, and the materials collected are summarized in the previous paper (Yang *et al.*, 1975).

On the other hand, sea-water sampling is carried out from 4 sites (Kangnung, Kori (Pusan), Yeosu, and Kunsan) along the coast of Korea in May, 1974 for the analysis of strontium-90 activities.

### Methods

The materials are washed in water for several times in order to purify, and are ashed under the temperature regulated at 400°C. After ashing, grinding and blending, a suitable aliquot of the ash is taken for analysis. The seaweed ash is treated with dilute nitric acid in presence of strontium carrier, and dissolved as completely as possible by heating on the hot plate. The solution should be complete except for traces of carbon and silica by means of filtering through Toyo No 5B paper to remove traces of insoluble materials. The filtrate is transferred to a 250ml centrifuge tube and then a few ml of phosphoric acid was added. Strontium was precipitated with carbon free ammonia water by centrifugation.

Strontium which was precipitated as phosphate is dissolved in fuming nitric acid and repeatedly precipitated as nitrate. Other radioelements are removed by scavenging with ferric hydroxide and barium chromate. After storage for 14 days in the presence of yttrium carrier, yttrium is

milked and counted with low beta counting system and then precipitated as carbonate for gravimetric strontium yield measurement (Suschny, 1967, Harley 1972, Yang 1973).

On the other hand, calcium oxalate is precipitate for calcium determination by excess ammonium oxalate from an ammonium acetate solution buffered to pH 4.5 to 5.0. The precipitate is dissolved in dilute sulfuric acid and the liberated oxalic acid is determined by n/10 permanganate solution (Pak and Yang, 1971).

The counting system used is a low background beta counter, LOW BETA II, Beckman. The background count rates of the counting system are less than 1 count per minute at 1 inch planchet for strontium-90. The counting efficiency of strontium-90 is about 34%. The estimation of the radioactive purity of strontium-90 and calculations of half-life are made on seaweeds which have high activities of yttrium-90.

The abbreviations used in this paper are as follows:

Kangnung: Ka	Sept. 1973 : 9
Samchuck: S	Dec. 1973 :12
Kizang: Ki	Jan. 1974 : 1
Yeosu: Y	Mar. 1974 : 3
Anmyundo: A	Apr. 1974 : 4
Cheju-isl.: C	
Zindo: Z	
Kunsan: Ku	

## RESULTS AND DISCUSSIONS

### Calcium contents

The calcium contents obtained from the present investigation are summarized in Table 1.

As shown in the table, the calcium contents fluctuate from 2% of *Porphyra yezoensis* to 17.8% of *Myelophycus caespitosus*. They are divided into two groups, 2—4% and 8—10%. Most of the members of green and red algae contain 2—4%, and those of brown algae 8—

Table 1. Calcium contents of edible seaweeds in Korea

Materials	Mean	Calcium percentage(%)
Green Algae		
<i>Caposiphon fulvelescens-Enteromorpha clathrata</i>	4.16	5.1(Ku-12), 3.1(Ku-3), 5.5(A-1), 3.2(Ki-3), 3.9(Y-3)
<i>Enteromorpha</i> -complex	3.80	3.4(Ku-12), 4.2(Ku-3)
<i>Enteromorpha linza</i>	3.35	3.1(Ki-3), 3.6(Y-3)
<i>Ulva pertusa</i>	4.10	5.2(Ki-3), 3.1(Y-3), 4.0(C-4)
<i>Codium fragile</i>	2.15	2.3(Ku-12), 2.0(A-3)
Brown Algae		
<i>Scytosiphon lomentaria</i>	9.82	9.2(S-3), 11.9(Ka-3), 8.4(Y-3), 9.8(C-4)
<i>Myelophycus caespitosus</i>	17.80	17.80(C-4)
<i>Ishige okamurai</i>	12.70	12.7(C-4)
<i>Undaria pinnatifida</i>	9.22	9.2(Ku-12), 11.1(Ki-3), 7.5(Ka-12), 9.5(Ka-3), 8.8(C-4)
<i>Ecklonia cava</i>	4.00	4.0(C-4)
<i>Costaria costata</i>	7.20	3.7(S-3), 10.7(Ka-3)
<i>Laminaria japonica</i>	9.50	10.3(Y-3), 8.7(Z-3)
<i>Kjellmaniella crassifolia</i>	5.90	5.9(Ka-3)
<i>Hizikia fusiforme</i>	8.35	9.1(Y-3), 7.61(C-4)
<i>Pelvetia wrightii</i>	9.90	11.0(Ku-12), 8.8(Ku-3)
<i>Sargassum thunbergii</i>	8.18	7.9(Ku-3), 5.2(Ki-3), 8.6(Y-3), 9.2(Ka-3), 10.0(Y-3)
<i>Sargassum fulvellum</i>	10.20	10.2(Ku-12), 10.4(Ku-3), 10.0(Y-3)
<i>Sargassum confusum</i>	12.40	12.4(C-4)
<i>Sargassum horneri</i>	13.85	13.9(A-3), 13.8(Y-3)
Red Algae		
<i>Porphyra tenera</i>	3.00	2.4(Ku-12), 2.7(Ku-3), 3.7(A-1), 3.2(Y-3)
<i>Porphyra yezoensis</i>	2.00	1.9(Ku-12), 2.1(Ka-3)
<i>Gelidium amansii</i>	5.50	5.5(C-4)
<i>Chondrus pinnulatus</i>	3.50	3.5(Ka-3)
<i>Enteromorpha-Porphyra</i> complex	6.50	6.5(A-1)

10%, whereas the mean value of calcium contents is 3.6% in green algae, 3.5% in red algae and 9.9% in brown algae. As a result, the brown algae contain about 3 times higher in contents compared with green and red algae.

The results are comparable with 5.27% of green algae, 11.87% of brown algae and 3.58% of red algae, reported by Yamamoto(1960) from the Japanese seaweeds, while they are higher than 1.7—2.5% of brown algae, reported by Lee *et al.* (1970). from edible brown algae in Korea. The tendency of calcium contents in the present investigation, which is generally higher in brown algae compared with green and red

algae, accords well with the one obtained by Yamamoto (1960).

#### Strontium-90 activities

Strontium-90 activities of the edible seaweeds investigated are summarized in Table 2 and Figure 1, and those of sea-water are summarized in Table 3. Before the analysis of strontium-90 activities, the purity test for the activities by detecting the half-life of yttrium-90 is carried out with green alga, *Enteromorpha linza* and brown alga, *Sargassum thunbergii*, as shown in Table 4 and Figure 2. By the test, the purity of strontium-90 is demonstrated

Table 2. Strontium-90 activities of edible seaweeds in Korea

Materials	Mean	Activities (pCi <sup>90</sup> Sr/g-Ca)
<b>Green Algae</b>		
<i>Capsosiphon fulvelescens-Enteromorpha clathrata</i>	28.66	23.85(Y-3), 37.26(Ku-12), 39.35(Ku-3), 14.18(A-1)
<i>Enteromorpha</i> -complex	10.68	4.29(Ku-3), 17.06(Ku-12)
<i>Enteromorpha linza</i>	7.22	7.22(Y-3)
<i>Ulva pertusa</i>	4.56	7.12(Ki-3), 2.0(C-4)
<i>Codium fragile</i>	11.30	9.13(Ku-12), 13.46(A-3)
<b>Brown Algae</b>		
<i>Scytosiphon lomentaria</i>	7.26	2.35(C-4), 4.13(S-3), 13.62(Ka-3), 8.93(Y-3)
<i>Myelophycus caespitosus</i>	0.84	0.84(C-4)
<i>Ishige okamurai</i>	1.97	1.97(C-4)
<i>Undaria pinnatifida</i>	3.40	1.74(Ku-12), 1.62(Ki-3), 6.27(Ka-12), 3.30(Ka-3), 4.09(C-4)
<i>Ecklonia cava</i>	0.80	0.80(C-4)
<i>Costaria costata</i>	2.60	1.73(S-3), 3.46(Ka-3)
<i>Laminaria japonica</i>	3.10	3.10(Z-3)
<i>Kjellmaniella crassifolia</i>	3.56	3.56(Ka-9)
<i>Hizikia fusiforme</i>	2.39	2.42(Y-3), 2.37(C-4)
<i>Pelvetia wrightii</i>	5.65	3.91(Ku-12), 7.39(Ku-3)
<i>Sargassum thunbergii</i>	7.00	5.35(Y-3), 7.59(Ku-3), 12.6(Ka-3), 12.46(C-4)
<i>Sargassum fulvellum</i>	2.62	2.74(Ku-12), 2.71(Ku-3), 2.40(Y-3)
<i>Sargassum confusum</i>	1.37	1.37(C-4)
<i>Sargassum horneri</i>	4.19	4.96(A-3), 3.41(Y-3)
<b>Red Algae</b>		
<i>Porphyra tenera</i>	10.78	10.0(Ku-12), 13.33(Ku-3), 13.24(A-1), 6.56(Y-3)
<i>Porphyra yezoensis</i>	13.87	11.53(Ku-3), 16.20(Ka-3)
<i>Gelidium amansii</i>	5.46	5.46(C-4)
<i>Chondrus pinnulatus</i>	7.43	7.43(Ka-3)
<i>Enteromorpha-Porphyra</i> complex	3.23	3.23(A-1)

clearly.

The strontium-90 activities of edible marine algae investigated are variable from 0.80 pCi/<sup>90</sup>Sr/g-Ca of *Ecklonia cava* to 28.66 pCi <sup>90</sup>Sr/g-Ca of *Capsosiphon fulvelescens-Enteromorpha clathrata* complex, and 6.25 pCi <sup>90</sup>Sr/g-Ca on an average.

The average value of the strontium-90 activities is 12.49 pCi <sup>90</sup>Sr/g-Ca in green algae, 3.34 pCi <sup>90</sup>Sr/g-Ca in brown algae and 9.39 pCi<sup>90</sup> Sr/g-Ca in red algae. Therefore, the activities decrease from green to red and brown algae, in turn. The tendency is quite contrary to those of gross alpha and beta activities

among the phyla of algae (Yang *et al.*, 1975).

Except for *Porphyras* (10.78 pCi<sup>90</sup> Sr/g-Ca of *P. tenera* and 13.87 pCi <sup>90</sup>Sr/g-Ca of *P. yezoensis*), all the members of red and brown algae show less than 8 pCi <sup>90</sup>Sr/g-Ca, whereas except for *Enteromorpha linza* (7.22 pCi <sup>90</sup>Sr/g-Ca) and *Ulva pertusa* (4.56 pCi<sup>90</sup> Sr/g-Ca) the members of green algae show more than 10 pCi <sup>90</sup>Sr/g-Ca.

Especially it is a noticeable fact that the strontium-90 activities of *Capsosiphon-Enteromorpha* complex are about 3 times higher than the other members that indicate comparatively high activities. As seen in the gross beta activi-

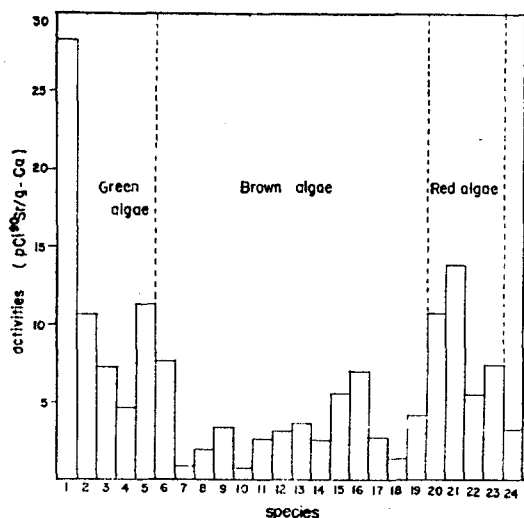


Fig. 1. Strontium-90 activities of edible seaweeds in Korea.

1. *Capsosiphon fulvelescens* - *Enteromorpha clathrata*
2. *Enteromorpha*-complex
3. *Enteromorpha linza*
4. *Ulva pertusa*
5. *Codium fragile*
6. *Scytosiphon lomentaria*
7. *Myelophycus caespitosus*
8. *Ishige okamurai*
9. *Undaria pinnatifida*
10. *Ecklonia cava*
11. *Costaria costata*
12. *Laminaria japonica*
13. *Kjellmaniella crassifolia*
14. *Hizikia fusiforme*
15. *Pelvetia wrightii*
16. *Sargassum thunbergii*
17. *Sargassum fulvellum*
18. *Sargassum confusum*
19. *Sargassum horneri*
20. *Porphyra tenera*
21. *Porphyra yezoensis*
22. *Gelidium amansii*
23. *Chondrus pinnulatus*
24. *Enteromorpha*-*Porphyra* complex

Table 3. Contents of Sr-90 in sea-water along the coast of Korea

Site	Sr-90 pCi/l
Eastern coast	0.32
Southern coast	0.35
Western coast	0.37
Average	0.35

ties of *Sargassum thunbergii* in the previous paper (Yang *et al.*, 1975), the plants inhabit also rather commonly, and survive among seaweeds more strongly in variously contaminated environment. Considering the concentration of strontium-90 from sea-water, it is presumed that these plants would be also an indicator to

Table 4. Yttrium-90 counting data

Material	Date	Time	Activity
<i>Enteromorpha linza</i>	Jun. 17, '74	11 : 56	430.14CPM
	Jun. 18, '74	12 : 04	334.40CPM
	Jun. 19, '74	10 : 00	262.36CPM
	Jun. 20, '74	09 : 33	208.24CPM
	Jun. 21, '74	14 : 57	150.42CPM
<i>Sargassum thunbergii</i>	Jun. 17, '74	12 : 47	32.36CPM
	Jun. 19, '74	10 : 51	19.50CPM
	Jun. 20, '74	10 : 37	16.30CPM
	Jun. 21, '74	15 : 48	12.40CPM

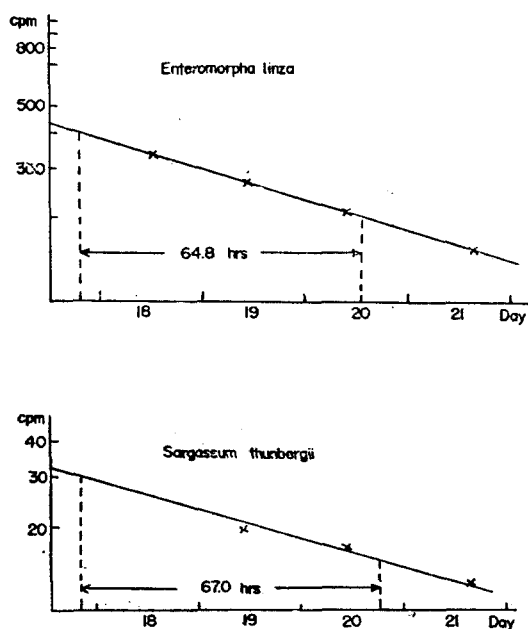


Fig. 2. Yttrium-90 decay curves of *Enteromorpha linza* (above) and *Sargassum thunbergii* (below).

detect the strontium-90 activities of marine algae along the coast of Korea.

Comparing the strontium-90 activities in a single species with the materials collected at the same season, the highest one is generally from eastern coast, and then from western coast, while the less one is from southern coast; for example, in *Capsosiphon-Enteromorpha* complex they show 39.35 (Ku-3) v.s. 23.85(Y-3), in *Ulva pertusa* 2.0 (C-4) v.s. 7.12 (Ki-3), in

*Undaria pinnatifida* 1.74 (Ku-12), v.s. 6.27 (Ka-12), in *Sargassum thunbergii* 7.59 (Ku-3) v.s. 5.35 (Y-3) v.s. 12.6 (Ka-3) in *Porphyra tenera* 13.33 (Ku-3) v.s. 6.56 (Y-3), and in *Porphyra yezoensis* 11.53 (Ku-3) v.s. 16.20 (Ka-3), etc.

To compare the data of green laver 8.49 pCi<sup>90</sup> Sr/g-Ca and dulce 3.21 pCi <sup>90</sup>Sr/g-Ca reported by Yang and Pak (1973) with the present investigation of green laver (= *Enteromorpha linza* and *Enteromorpha*-complex) 8.95 pCi <sup>90</sup>Sr/g-Ca and dulce (= *Undaria pinnatifida*) 3.40 pCi<sup>90</sup> Sr/g-Ca, the both results accord quite well with each other.

The contamination of strontium-90 in sea-water, as shown in Table 3, is 0.35 pCi/l on an average, while 0.32 pCi/l in eastern coast, 0.35 pCi/l in southern coast, and 0.37 pCi/l in western coast, respectively. As a result, the strontium-90 activities are highest in western coast among the three parts of the of Korea. Considering the fact that western coast of Korea is facing to China, the result is very interesting and noticeable. Moreover, the contents of strontium-90 along the coast of Korea are slightly higher than 0.28-0.31 pCi/l reported by Ervet *et al.* (1966) from the coast of France.

Wilber (1969) proposed the maximum allowance of the radiation for seaweeds as below:

	Max. allowance in sea-water (pCi/ml)	Max. allowance in seaweeds(pCi/g)
gross beta	$2 \times 10^{-4}$	676
strontium-90	$8 \times 10^{-7}$	207

Considering the proposal, these species investigated contain the gross beta activities of about 1/110 times in green algae, 1/60 in brown algae and 1/80 of the maximum allowance in red algae, while *Sargassum thunbergii*, showing the highest activities among the species investigated, contains about 1/30 times of the maximum allowance (referred from the previous paper, Yang *et al.*, 1975).

The strontium-90 activities of the species investigated, on the other hand, contain about 1/16 times in green algae, about 1/62 times in brown algae and about 1/22 times of maximum allowance in red algae, while *Capsosiphon-Enteromorpha* complex, showing the highest concentration of strontium-90 among the species investigated, contains about 1/7 times of maximum allowance.

According to Bouvoyiannis and Mimikos (1966), and I.C.R.P. data (1962) the strontium-90 activities, ones of the most dangerous radiations for human being, are permitted 1 nCi in maximum for a person, which is calculated to 1,100 pCi <sup>90</sup>Sr/g-Ca for a 60 kg weight person. Considering the maximum permission, these plants investigated contain about 1/90 times in green algae, 1/330 times in brown algae and 1/120 times of the maximum permission in red algae, while *Capsosiphon-Enteromorpha* complex contains about 1/38 times of the maximum permission.

## CONCLUSION

The calcium contents fluctuate from 2% to 17.8%, and 6.38% on an average among the edible seaweeds investigated. They are divided into two groups, 2-4% and 8-10%. Most of the members of green and red algae contain 2-4% of calcium, and those of brown algae 8-10%. The mean value of calcium contents among the phyla of algae is 3.6% in green, 3.5% in red and 9.9% in brown algae. Therefore, the brown algae contain about 3 times higher than the green and red algae investigated. Such a tendency accords well with the result obtained by Yamamoto (1960) from the Japanese seaweeds.

On the other hand, strontium-90 content in sea-water is 0.35 pCi/l on an average, while it is 0.32 pCi/l in eastern coast, 0.35 pCi/l in

southern coast and 0.37 pCi/l in western coast of Korea. The result is slightly higher than the one reported by Ervet *et al.* (1968) from the coast of France. However, the strontium-90 activities of the samples investigated are variable from 0.80 pCi  $^{90}\text{Sr/g-Ca}$  to 28.66 pCi $^{90}\text{Sr/g-Ca}$ , and 6.25 pCi  $^{90}\text{Sr/g-Ca}$  on an average. Among the algal phyla, they are 12.49 pCi $^{90}\text{Sr/g-Ca}$  in green algae, 3.34 pCi  $^{90}\text{Sr/g-Ca}$  in brown algae and 9.39 pCi  $^{90}\text{Sr/g-Ca}$  in red algae. Therefore, the activities decrease from green to red and brown algae, in turn. Especially it is a noticeable fact that the strontium-90 activities of *Capsosiphon-Enteromorpha* complex are about 3 times higher than the other members that show comparatively high activities. Considering the concentration factor of strontium-90 from sea-water, it is presumed that the plants would be an indicator to detect the activities of marine algae along the coast of Korea.

Comparing the strontium-90 activities in a single species with the materials collected at the same season, the highest one is generally from the eastern coast, and then from western coast, while the less one is from southern coast.

Considering the maximum permission of strontium-90 activities for human being proposed by Bouvoyiannis and Mimikos (1966) and I.C.R.P. data (1962), such as 1,100 pCi  $^{90}\text{Sr/g-Ca}$  for a 60 kg weight person, the samples investigated contain about 1/90 times of maximum permission in green algae, 1/330 in brown algae and 1/120 times in red algae. The member *Capsosiphon-Enteromorpha* complex, showing the highest concentration of strontium-90 among the species investigated, contains about 1/38 times of the maximum permission.

On the other hand, considering the proposal of Wilber (1969) these species investigated contain the strontium activities of about 1/16 times in green algae, 1/62 times in brown algae, and

1/22 times of maximum allowance in red algae, while *Capsosiphon-Enteromorpha* complex contains about 1/7 times of the maximum allowance in seaweeds.

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