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TRACE METALS IN SEVERAL EDIBLE MARINE ALGAE OF KOREA

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ABSTRACT

The present paper deals with nine kinds of trace metal contents among 79 samples, more than 20 species of edible marine algae collected during 1973~1975 along the coasts of Korea. Among the metals detected by the atomic absorption spectrometer, Mg content is variable in 2.09~13.84mg /g-dry on an average and 5.31mg/g-dry in a mean of green, brown and red algae, whereas Fe is 0.148~2.343 and 0.747mg/g-dry; Mn 0.015~0.191 and 0.063mg/g-dry; Zn 0.007~0.082 and 0.030 mg/g-dry; Cu 0.005~0.054 and 0.017mg/g-dry; Pb 0.0017~0.082 and 0.0043mg/g-dry; Co 0.0009~ 0.0024 and 0.0016mg/g-dry; Cd 0.0002~0.0034 and 0.0011mg/g-dry, and Cr is 0.0007~0.0074 and 0.0021mg/g-dry, both on an average and in a mean respectively.

The trace metal contents in the seaweeds investigated are variable according to species, collecting seasons, places and kinds of metal. It is interesting that in *Sargassum thunbergii* some metal contents such as Fe, Mn, and Zn are specially high in September 1974 through the year investigated.

INTRODUCTION

Reports on the trace metal contents in marine algae from Korea are lack of data than inorganic components in the terrestrial plants or organic matters in the seaweeds.

Yang(1964), Lee *et al.* (1961, 1962 a, b), Lee & Lee (1963), and Hong (1964) reported qualitatively several organic substances including alginic acids, crude protein, amino acids, or free sugars, etc., whereas Yang(1964) and Lee *et al.* (1971) investigated some inorganic metals in edible marine algae. On the other hand, Yang *el al.* (1975 a, b, c) reported on the accumulation of radioactive materials in the edible marine algae of Korea.

This paper deals with the contents of trace heavy metals such as magnesium, iron, manganese, zinc, lead, copper, cadium, chromium, and cobalt in the edible marine algae collected along the coasts of Korea during September, 1973 and September, 1975.

MATERIALS AND METHODS

Materials

For the analysis of trace heavy metals 79 samples of more than 20 edible seaweed species collected along the coasts of Korea during 1973 ~1975 were used in this experiment. Among them the samples collected during September, 1973~April, 1974 were referred to the previous paper (Yang *et al.* 1975 a) and the further collections during May, 1974~September, 1975 were summarized in Table 1.

Methods

The materials experimented were rinsed with tap water to remove adhering debris and then

Table 1. Sampling sites and dates, and the kinds of seaweeds (2nd year: May, 1974~Sept. 1975)

Name	Sites	Dates
⟨Green Algae⟩		
Enteromorpha complex	Kunsan	Apr. 1974
Enteromorpha linza	Yeosu(Namhaedo)	Mar. 1974
Ulva pertusa	Yeosu(Nanchodo)	May 1974
Ulva pertusa	Yeosu(Odongdo)	May 1974
Ulva pertusa	Yeosu(Myodo)	Sep. 1974
Ulva pertusa	Yeosu(Nanchodo)	Mar. 1975
Ulva pertusa	Cheju Isl.	Sep. 1975
Codium fragile	Cheju. Isl.	Sep. 1975
⟨Brown Algae⟩		
Undaria pinnatifida	Kunsan	Apr. 1974
Undaria pinnatifida	Mokpo	Apr. 1974
Undaria pinnatifida	Kizang	Mar. 1975
Undaria pinnatifida	Cheju Isl.	Sep. 1985
Sargassum thunbergii	Yeosu(Myodo)	May 1974
Sargassum thunbergii	Yeosu(Odongdo)	May 1974
Sargassum thunbergii	Yeosu(Nakpori)	May 1974
Sargassum thunbergii	Yeosu(Nanchodo)	May 1974
Sargassum thunbergii	Yesou(Myodo)	Sep. 1974
Sargassum thunbergii	Yeosu(Sudo)	Nov. 1974
Sargassum thunbergii	Yeosu(Namhaedo)	Nov. 1974
Sargassum thunbergii	Yeosu(Myodo)	Jan. 1975
Sargassum thunbergii	Yeosu(Nanchodo)	Mar. 1975
Sargassum thunbergii	Yeosu(Namhaedo)	Mar. 1975
Sargassum thunbergii	Cheju Isl.	Sep. 1975
Hizikia fusiforme	Cheju Isl.	Sep. 1975
⟨Red Algae⟩		
Gelidium amansii	Cheju Isl.	Sep. 1975

dried at 100°C in the electric oven. The dried samples were ashed under the temperature regulated at 400°C. After ashing, grinding and blending, 0.5~1g of ash sample were placed in conical pyrex flask. 15ml of concentrated nitric acid and 3ml of 70% perchloric acid were added and dissolved as completely as possible by heating on the hot plate. After dissolving, the solution was filtered through Toyo No. 5B filter paper into 100ml volumetric flask and made to volume with deionized distilled water. Appropriate blank solutions were similarly prepared for each analysis at the same time. The standard were prepared by diluting aqueous solution of sample metal salts to the concentration required. All measurements were performed by an atomic absorption flame photometer, Model AA-610S, SHIMAZU, JAPAN, equipped with a 5cm/10cm convertible burner-head. All analytical procedures adopted for the trace metals were referred to the following reports; manganese (Allan 1959, David, 1962), zinc (David 1958, Allan, 1961 a), copper (Allan 1961 b, Morgan 1964), iron (David 1962), magnesium (David 1962), lead (Chakrabarti *et al.* 1966) cadmium (Dagnall *et al.* 1966, Pulido *et al.* 1966), Cobalt (Salvin *et al.* 1964), and chromium (Krumpaek 1967).

The abbreviations used in this paper are as follows:

Sampling sites Kangnung: Ka, Samchuck: S, Kizang: Ki, Yeosu: Y, Anmyondo: A, Cheju Isl.: C, Zindo: Z, Kunsan: Ku, Mokpo: M.

Sampling dates Sep. 1973: 9-73, Dec. 1973: 12-73, Jan. 1974: 1-74, Mar. 1974: 3-74, Apr. 1974: 4-74 (1st year). May 1974: 5-74, Apr. 1974: 4-74, Sep. 1974: 9-74, Nov. 1974: 11-74, Jan. 1975: 1-75, Mar. 1975: 3-75, Sep. 1975: 9-75 (2nd year).

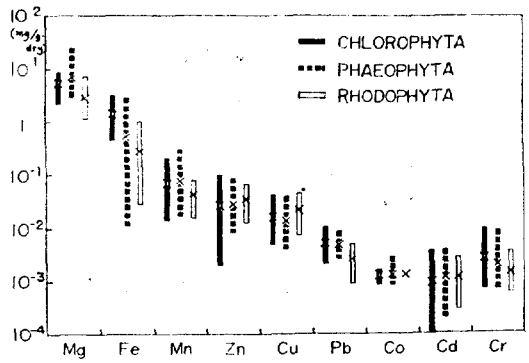

Fig. 1. The variation of trace metal contents in several edible marine green, brown and red algae of Korea.

RESULTS AND DISCUSSIONS

The experimental results of heavy trace metals in marine algae were summarized in Table 2 and Fig. 1. Among the metals investigated Mg contents are as high as 5.31mg/g-dry in a mean and 2.09~13.84mg/g-dry on an average in fluctuation among green, brown and red algae, while they are ranged in 2.25~8.85mg/g-dry and 5.53mg/g-dry on an average in green algae investigated, 2.89~25.19mg/g-dry and 7.35mg /g-dry in brown algae, and 1.13~7.48mg/g-dry and 3.06mg/g-dry in red algae, respectively. Among the samples experimented Mg content is the highest in *Sargassum fulvellum*(Ku-12-73) as 25.19mg/g-dry, and the lowest in *Porphyra tenera* (A-1-74) as 1.13mg/g-dry. These results are comparable with the mean value reported by Yamamoto (1961) of Japanese seaweeds. (green algae, 21.3, brown algae 11.9 and red algae 5.0mg/g-dry) in that our samples show rather lower Mg contents. Such a result was also mentioned in edible brown algae of Korea by Lee *et al.*(1971).

Fe contents are variable among 0.148~2.343 mg/g-dry on an average and 0.747mg/g-dry of a mean. They show 0.404~3.160mg/g-dry in fluctuation and 1.448mg/g-dry on an average in green algae, 0.011~2.817mg/g-dry and 0.518 mg/g-dry in brown algae, and 0.028~1.051mg /g-dry and 0.275mg/g-dry in red algae, respectively. *Codium fragile* (A-3-74) shows the highest Fe content as 3.160mg/g-dry among the samples investigated, and *Kjellmaniella crassifolia* (Ka-9-73) the lowest as 0.011mg/g-dry. The results are also comparable with the reports on Japanese algae of Morii(1962) and Yamamoto (1960), and on Korean edible brown algae of Lee *et. al.*(1971) in that they show generally a same tendency as the present results.

Mn contents are variable from 0.015 to 0.191 mg/g-dry on an average and 0.063mg/g-dry in a mean. They are ranged in 0.014~0.202mg/g-dry and 0.068mg/g-dry on an average in green algae, 0.016~0.290mg/g-dry and 0.077mg/g-dry in brown algae, and 0.015~0.080mg/g-dry and 0.043mg/g-dry in red algae, respectively. *Sargassum thunbergii* (Y-9-74) shows the highest Mn content as 0.290mg/g-dry and *Enteromorpha linza* (Ki-3-74) the lowest as 0.014mg/g-dry among the samples examined. The results accord quite well with the Mn contents of Japanese algae metioned by Ishibashi & Yamamoto(1958) as 0.30 of green, 0.07 of brown and 0.05mg/g-dry of red algae, while they are somewhat lower on an average compared with several brown algae of England reported by Black & Mitchell(1952) as 0.009~0.800mg/g-dry.

Zn contents are ranged in 0.007~0.082mg/g-dry on an average and 0.030mg/g-dry in a mean. They show 0.002~0.095mg/g-dry in fluctuation and 0.026mg/g-dry on an average in green algae, 0.008~0.084 mg/g-dry and 0.028 mg/g-dry in brown algae, and 0.012~0.068mg/g-dry and 0.035mg/g-dry in red algae, respectively. *Ulva pertusa* (Ki-3-74) shows the highest Zn content as 0.095mg/g-dry and *Enteromorpha linga* (Ki-3-74) the lowest as 0.002mg/g-dry. Lee *et al.* (1971) mentioned that Zn contents in edible brown algae were fluctuated much among the species investigated such as 0.0282~0.0984 % of dry weight. However, in the present result, they do not show specially broad fluctuation among the species experimented.

Cu contents are varable from 0.005 to 0.054 mg/g-dry on an average and 0.017mg/g-dry in a mean. They are ranged in 0.005~0.040mg/g-dry and 0.016mg/g-dry on an average in green algae, 0.004~0.038mg/g-dry and 0.014mg/g-dry in brown algae, and 0.007~0.045mg/g-dry and 0.022mg/g-dry on an average in red algae, res-

Table 2. Heavy Metals in several edible marine algae of Korea.

(Unit : mg/g-dry)

Materials	Sampling Site & Date	Mg	Fe	Mn	Zn	Cu	Pb	Co	Cd	Cr
Green Algae										
Capsosiphon fulvelescens-Enteromorpha clathraea	A-1-74	5.79	0.770	0.030	0.010	0.013	0.0026		0.0009	
	Ku-3-74	6.49	0.486	0.063	0.021	0.021	0.0061		0.0034	0.0029
	Y-3-74	7.68	0.441	0.072	0.040	0.014	0.0107		0.0006	0.0014
	Ki-3-74	2.72	0.404	0.048	0.014	0.014	0.0021		0.0007	0.0007
Enteromorpha complex	Ku-12-73	5.31	2.009	0.109	0.067	0.040	0.0074		0.0008	0.0018
	Ku-3-74	5.02	1.343	0.123	0.037	0.023	0.0027		0.0014	0.0014
	Ku-4-74	5.21	2.025	0.032	0.014	0.012		0.0017	0.0010	0.0058
Enteromorpha linza	Y-3-74	6.12	2.763	0.101	0.018	0.021	0.0085	0.0015	0.0006	0.0097
	Ki-3-74	3.50	2.397	0.014	0.002	0.008	0.0034		0.0038	0.0013
Ulva pertusa	Y-3-74	2.72	1.848	0.068	0.034	0.027	0.0058		0.0008	0.0012
	Y-5-74	7.42	1.945	0.063	0.014	0.010		0.0015	0.0007	0.0047
	Y-9-74	8.85	1.787	0.101	0.021	0.018		0.0017	0.0010	0.0036
	Y-3-75	7.12	1.457	0.050	0.014	0.015		0.0010	0.0007	0.0041
	C-4-74	7.78	0.596	0.049	0.017	0.009	0.0056		0.0009	0.0012
	C-9-75	5.94	1.357	0.034	0.024	0.013		0.0013	0.0004	0.0034
	Ki-3-74	8.80	0.518	0.041	0.095	0.015	0.0031		0.0001	0.0008
Codium fragile	A-3-74	2.25	3.160	0.024	0.023	0.015	0.0053		0.0008	0.0012
	Ku-12-73	2.85	1.452	0.202	0.020	0.009	0.0062		0.0003	0.0010
	C-9-75	3.48	0.750	0.063	0.006	0.005		0.0009	0.0004	0.0041
Brown Alage										
Myelophycus caespitosus	C-4-74	6.26	0.593	0.039	0.039	0.007	0.0053		0.0005	0.0009
Ishige okamurai	C-4-74	4.20	0.032	0.028	0.045	0.007			0.0004	0.0006
Undaria pinnatifida	Ku-12-73	5.04	0.162	0.044	0.018	0.010	0.0031		0.0012	0.0027
	Ku-4-74	5.56		0.016	0.030	0.004		0.0009	0.0010	0.0088
	M-4-74	5.75	0.331	0.018	0.036	0.034		0.0012	0.0010	0.0060
	C-4-74	4.68	0.087	0.039	0.029	0.038	0.0058		0.0007	0.0018
	C-9-75	5.91	0.490	0.018	0.028	0.010		0.0009	0.0009	0.0023
	Ki-3-74	6.34	0.387	0.053	0.028	0.008	0.0037		0.0003	0.0009
	Ki-3-75	5.47	0.373	0.019	0.030	0.029		0.0010	0.0010	0.0029
	Ka-12-73	6.92	0.134	0.025	0.023	0.013	0.0036		0.0003	0.0011
	Ka-3-74	8.70	0.309	0.072	0.049	0.016	0.0084		0.0024	0.0013
Ecklonica cava	S-3-74	4.16	0.335	0.025	0.023	0.007	0.0043		0.0009	0.0028
Costaria costata	S-3-74	7.35	0.108	0.050	0.084	0.022	0.0028		0.0002	0.0009
	Ka-3-74	9.18	0.373	0.067	0.034	0.012	0.0057		0.0004	0.0028
*Laminaria japonica**	Y-3-74	6.26	0.048	0.032	0.014	0.015	0.0033		0.0010	0.0007
	Z-3-74	5.43	0.231	0.042	0.016	0.016	0.0055		0.0005	0.0058
Kjellmaniella crassifolia	Ka-9-73	6.42	0.011	0.037	0.033	0.011	0.0038		0.0003	0.0010
Hizikia fusiforme	Y-3-74	6.27	0.029	0.048	0.020	0.016	0.0061		0.0010	0.0009
	C-4-74	2.89	0.022	0.031	0.014	0.009	0.0046		0.0006	0.0014
	C-9-75	7.82	0.176	0.022	0.008	0.007		0.0030	0.0013	
Pelvetia wrightii	Ku-12-73	9.91	0.523	0.107	0.027	0.024	0.0026		0.0037	0.0014
	Ku-3-74	7.54	0.510	0.137	0.024	0.009	0.0037		0.0012	0.0012
Scytosiphon lomentaria	Y-3-74	5.44	0.325	0.185	0.056	0.013			0.0021	0.0008

Materials	Sampling Site & Date	Mg	Fe	Mn	Zn	Cu	Pb	Co	Cd	Cr
	C-4-74	5.25	0.526	0.058	0.031	0.013	0.0052		0.0009	0.0008
	S-3-74	4.04	0.651	0.091	0.053	0.010	0.0048		0.0004	0.0009
	Ka-3-74	5.03	0.350	0.071	0.037	0.007	0.0044		0.0019	0.0017
Sargassum thunbergii	Ku-4-74	4.05	0.303	0.189	0.054	0.014	0.0064		0.0016	0.0019
	Y-3-74	8.78	0.367	0.109	0.013	0.013	0.0064		0.0014	0.0020
	Y-5-74	5.87	1.739	0.176	0.009	0.010		0.0016	0.0010	0.0043
	Y-9-74	8.61	2.817	0.290	0.010	0.013		0.0021	0.0017	
	Y-11-74	8.25	0.719	0.104	0.012	0.006		0.0019	0.0015	
	Y-1-75	10.02	0.989	0.162	0.013	0.006		0.0019	0.0017	
	Y-3-75	8.21	1.488	0.196	0.030	0.007		0.0023	0.0017	0.0018
	C-4-74	5.08	0.429	0.123	0.027	0.013	0.0040		0.0005	0.0009
	C-9-75	3.91	0.443	0.104	0.008	0.011		0.0015	0.0007	0.0085
	Ki-3-74	3.52	0.535	0.107	0.036	0.022	0.0040		0.0011	0.0012
	Ka-3-74	4.87	0.339	0.116	0.026	0.022	0.0037		0.0004	0.0011
Sargasssum fulvellum	Ku-12-73	25.19	0.825	0.064	0.023	0.019			0.0030	0.0011
	Ku-3-74	22.81	1.113	0.102	0.014	0.012	0.0042		0.0020	0.0013
	Y-3-74	14.73	0.470	0.046	0.011	0.008	0.0068		0.0024	0.0011
Sargassum confusum	C-4-74	4.73	0.078	0.029	0.014	0.007	0.0058		0.0016	0.0010
Sargassum horneri	A-3-74	9.16	0.997	0.061	0.028	0.009	0.0089		0.0027	0.0008
	Y-3-74	10.38	0.998	0.049	0.061	0.025	0.0067		0.0017	
Red Algae										
Porphya tenera	A-1-74	1.13	0.164	0.028	0.030	0.016	0.0013		0.0018	0.0036
	Ku-12-73	1.19	0.350	0.044	0.031	0.022	0.0017		0.0011	0.0012
	Ku-3-74	1.59	0.204	0.021	0.058	0.045	0.0009		0.0012	0.0012
	Y-3-74	1.19	0.254	0.075	0.012	0.016	0.0051		0.0004	0.0012
Porphyra yezoensis	Ku-3-74	1.28	0.239	0.068	0.027	0.020	0.0023		0.0018	0.0011
	Ka-3-74	2.27	0.128	0.022	0.037	0.013	0.0034		0.0003	0.0011
Gelidium amansii	C-4-74	2.69	0.061	0.036	0.014	0.007	0.0041		0.0007	0.0008
	C-9-75	7.48	1.051	0.080	0.034	0.012		0.0017	0.0007	0.0029
Chondrus pinnulatus	Ka-3-74	2.48	0.028	0.015	0.068	0.043	0.0018		0.0028	0.0006
Enteromorpha-Porphyra complex	A-1-74	3.09	0.237	0.056	0.019	0.016	0.0022		0.0030	0.0012

* This plant does not occur in the coast of South Korea, but is cultivated in several places.

pectively. *Porphyra tenera* (Ku-3-74) is the highest as 0.045mg/g-dry and *Codium fragile* (C-9-75) the lowest as 0.005mg/g-dry among the samples experimented.

On the other hand Pb contents fluctuate between 0.0017~0.0082mg/g-dry on an average and 0.0043mg/g-dry in a mean of green, brown and red algae. They are ranged in 0.0021~0.0107 mg/g-dry and 0.0053mg/g-dry on an average in green algae, 0.0026~0.0089mg/g-dry and 0.0050 mg/g-dry in brown algae, and 0.0009~0.0051 mg/g-dry and 0.0026mg/g-dry in red algae, respectively. *Capsosiphon fulvelescens-Enteromorpha clathrata* (Y-3-74) show the highest Pb content as 0.0107mg/g-dry and *Porphyra tenera* (Ku-3-74) the lowest as 0.0009mg/g-dry among the samples experimented.

Co contents in edible seaweeds of Korea are ranged in 0.0009~0.0024mg/g-dry on an average and 0.0016mg/g-dry in a mean. Since they are detected only in 19 samples among 72, the fluctuation range may be changed in further

investigaton. However, Co contents among the experimented samples show rather a narrow fluctuation of the variation. They are 0.009~0.0017mg/g-dry and 0.0014mg/g-dry on an average of 7 samples in green algae, 0.0009~0.0030mg/g-dry and 0.0017mg/g-dry on an average of 11 samples in brown algae, and 0.0017mg/g-dry in *Gelidium amansii* (C-9-75) of red algae. Among the samples experimented, *Hizikia fusiforme* (C-9-75) shows the highest Co content as 0.0030mg/g-dry, and *Codium fragile* (C-9-75) and *Undaria pinnatifida* (Ku-4-74, C-9-75) the lowest as 0.0009mg/g-dry.

Cd contents are 0.0002~0.0034mg/g-dry on an average and 0.0011mg/g-dry in a mean of green, brown and red algae. They show 0.0001~0.0038mg/g-dry in fluctuation and 0.0010mg/g-dry on an average in green algae, 0.0002~0.0037mg/g-dry and 0.0012mg/g-dry in brown algae, and 0.0003~0.0028mg/g-dry and 0.0012 mg/g-dry in red algae, respectively. *Enteromorpha lina* (Ki-3-74) shows the highest in Cd content as 0.0038mg/g-dry and *Ulva pertusa* (Ki-3-74) the lowest as 0.0001mg/g-dry.

Cr contents are 0.0007~0.0074mg/g-dry on an average and 0.0021mg/g-dry in a mean of green, brown and red algae. They show 0.0007~0.0097mg/g-dry in variation and 0.0028mg/g-dry on an average in green algae, 0.0007~0.0088mg/g-dry and 0.0021mg/g-dry in brown algae, and 0.0006~0.0036mg/g-dry and 0.0015 mg/g-dry in red algae, respectively. *Enteromorpha linza* (Y-3-74) contains the highest as 0.0097mg/g-dry and *Chondrus pinnulatus* (Ka-3-74) the lowest as 0.0006 mg/g-dry. These results indicate somewhat higher Cr content compared with those observed by Yamamoto (1960) in Japanese algae as 0.00047~0.00253 mg/g-dry and by Black (1952) in British seaweeds as 0.0004~0.0037 mg/g-dry.

Considering the investigations mentioned above, the concentrations of heavy metals are variable according to species, localities and seasons. However, in a single species they show characteristic range according to the metals (Fig. 1). It is interesting that in *Sargassum thunbergii* collected bimonthly at Yeosu, the southern coast of Korea, Zn, Mn and Fe contents are especially high in September through the year.

The high concentrations of heavy metals such as Cu, Cd, Co, Pb, and Cr in edible seaweeds may arise a damage to human health. Yang *et al.*(1975 a-c) mentioned the indicator plants of some radioactive substances in the edible seaweeds of Korea. So far as the present results are concerned, the concentration factors of these heavy metals are not so remarkable among the species investigated.

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