

Distribution of accumulated arsenic in the seaweed Hijiki, *Sargassum fusiforme* (Harvey) Setchell (6)

Masayuki KATAYAMA^{1,2)}, Yuko YAMAMOTO²⁾, Rie SAWADA²⁾, Yohko SUGAWA-KATAYAMA^{*1,2)}

Department of Health and Nutrition, Osaka Aoyama University¹⁾

Department of Health and Nutrition, Gifu Women's University²⁾

Summary Arsenic accumulation in Hijiki, *Sargassum fusiforme* (Harvey) Setchell, a member of the *Phaeophyta* Family, was determined throughout the whole plant by thermal neutron activation analysis. We collected Hijiki plants along a sea coast of Ise Bay, where Akamoku plants have also been harvested. The arsenic distribution in the Hijiki plant body was not uniform along the stalk like in Hijiki plants harvested at other districts and Akamoku plants of this district. Arsenic distribution in the genital organs of Hijiki showed high arsenic concentrations on the wet weight basis in comparison with other tissues. (accepted. Oct. 25, 2008)

Keywords : *Sargassum fusiforme* (Harvey) Setchell, Hijiki, arsenic, arsenic distribution, thermal neutron activation analysis, genital organs, leaves, stalks, twigs

Introduction

Seaweeds of the *Phaeophyta* family, some of which have been reported to contain high amounts of arsenic¹⁻³⁾, have been traditionally eaten by Japanese ; Hijiki⁴⁾, *Sargassum fusiforme* (Harvey) Setchell, of this family has been found to contain variable levels of arsenic irrespective of their harvesting areas of the ocean⁵⁻⁹⁾. To study the arsenic accumulation behavior in the *Phaeophyta* family, the arsenic concentration in some other members of the *Phaeophyta* family has remained to be determined. In the previous studies, patterns of arsenic distribution in Umi-torano-o, *Sargassum thunbergii* (Mertens ex Roth) Kuntze⁷⁾, and Akamoku, *Sargassum horneri* (Turner) C. Agardh, were investigated^{10,11)}. In this report, we present data on arsenic distribution in Hijiki plants harvested at the same sea-coast area where Akamoku was harvested^{10,11)}.

Experimental

Plants:

Hijiki plants were harvested at Mugisaki Coast, Katada, Ise Bay. The samples were carried in an ice-cold box to our laboratory, and washed successively 3 times with artificial sea water, twice with distilled

water and once with MilliQ water. After each washing, excessive water was blotted out on filter paper. They were cut into pieces of 10 cm lengths from the downside to the upper side along the stalk, which were designated as a', b', c', etc. The leaves and the apex were separated. (Fig. 1). The separated samples were sealed in small polyethylene bags and their wet weights were measured. They were stored in a deep freezer at -40°C until lyophilization.

Arsenic determination:

The lyophilized samples were weighted and their small portions were used for determination of arsenic contents by thermal neutron activation analysis.

Thermal neutron activation analysis^{6,12)}:

The specimens sealed in small polyethylene bags were bundled in Neuma-capsules and set in the center position of a research reactor of the Research Reactor Institute of Kyoto University. The thermal neutrons were irradiated at 10^{13} neutrons $\text{cm}^{-2} \cdot \text{sec}^{-1}$ for 20 min. After 72 hr of cooling time, arsenic contents were determined by a gamma detector equipped with pure Ge.

* E-mail: katayama@osaka-aoyama.ac.jp

1) 2-11-1 Niina, Minoh City, Osaka 562-8580, Japan

2) Taromaru 80, Gifu City, Gifu 501-3592, Japan

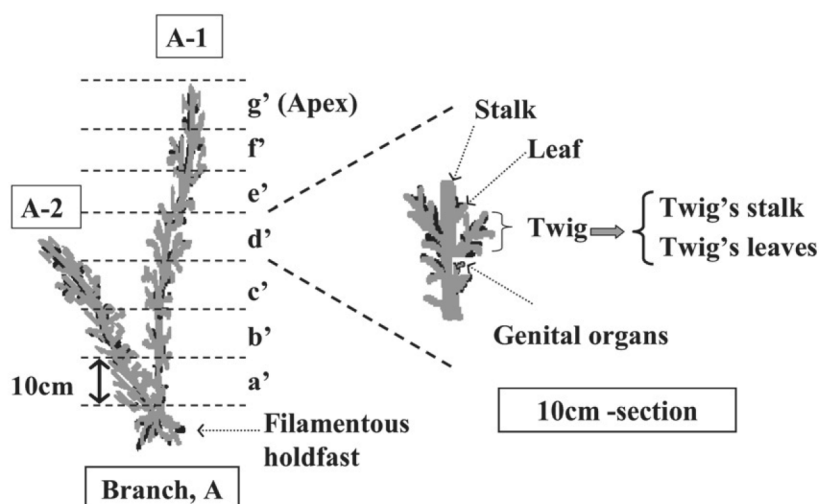


Fig.1 Fractionation of Hijiki plants.

After washing, the sample plants were cut into pieces of 10 cm length, and separated to respective tissues.

Results

Growth states of Hijiki:

A branch of Hijiki, designated as No A, had two stalks, one (A-1) 68 cm long and the other (A-2) 35 cm long, each possessing many twigs. The lengths of the twigs, mostly less than 10 cm in length, were separated from the respective 10cm-sections, and each was separated to leaves and stalks (Table 1). This branch had many genital organs, and they were also separated from the respective 10 cm-sections. (Fig. 1).

Water contents:

The ratios of the wet weight to the dry weight of the tissues suggested that the upper sections of stalks were less matured. The shorter stalk seemed to have more immature leaves than the longer stalk as shown in Table 1. A similar trend was found in the water contents of the leaves and stalks of twigs of the respective sections.

Arsenic contents on the wet weight basis:

The arsenic concentrations on the wet weight basis may reflect more physiological distribution than those on the dry weight basis, as arsenic seems to be uniformly dispersed in the wet tissues¹³⁾. The arsenic concentrations in the leaves and stalks were higher in the lower sections than in the upper sections, suggesting that the accumulated arsenic was not easily transferable in the tissues (Table 2).

It is interesting that the genital organs contained higher levels of arsenic than the other tissues in the same section (Table 2).

Arsenic contents on the dry weight basis:

The arsenic concentrations expressed on the dry weight basis (Table 3) showed generally 30 to 80 ppm in the leaves and stalks; the arsenic levels in the genital organs were sometimes more than 100 ppm. (Table 3).

Discussion

The water contents in the genital organs were mostly lower than the other tissues in the same sections (Table 1). As food for fishes or mollusks, the more rich contents with lower water contents may be preferred, and the high arsenic level might be a means to prevent the Hijiki plant from being eaten by other animals (Table 2).

The higher concentration of arsenic in the more matured tissues may indicate the more difficult of arsenic transfer between the tissues, resulting in the non-uniformity of arsenic distribution in the tissues along the stalks⁵⁻⁹⁾ (Table 2 and 3). Other elements such as calcium and iron also appeared not to be easily transferable between the tissues (our unpublished data) because of their insolubilities¹³⁾.

The arsenic levels in Hijiki were about 50% less than those in Akamoku¹¹⁾, and they perhaps depend on the duration of contact with ocean sea water¹¹⁾.

When used as food, pre-cooking treatments will be essential as shown by the water-soaking process of dried Hijiki¹⁴⁾.

Table 1. Growth state of Hijiki plants, harvested at Mugsaki-Katada district, Ise Bay

Sample*	Tissues	Sections**	Wet Weight (g)	Dry Weight (g)	Wet Weight / Dry Weight	Length of the stalks (cm)
A-1	Stalks	g'	0.807	0.099	8.126	8.0
		f'	1.383	0.193	7.179	10.0
		e'	1.553	0.236	6.582	10.0
		d'	1.645	0.268	6.136	10.0
		c'	1.625	0.257	6.329	10.0
		b'	1.298	0.225	5.767	10.0
A-2	Stalks	a'	1.027	0.199	5.156	10.0
		d'	0.473	0.038	12.327	4.5
		c'	1.170	0.168	6.982	10.0
		b'	1.246	0.182	6.827	10.0
A-1	Leaves	a'	0.751	0.127	5.927	10.0
		b'	2.705	0.253	10.674	
		g'(Apex)	1.011	0.079	12.788	
		f'	1.546	0.205	7.536	
		e'	1.056	0.120	8.765	
		d'	2.024	0.163	12.433	
A-2	Leaves	c'	1.836	0.134	13.725	
		a'	3.053	0.255	11.974	
		d'(Apex)	0.416	0.037	11.293	
		e'	0.004	0.001	7.717	
A-1	Genital Organs	f'	0.024	0.005	4.763	
		e'	0.025	0.005	4.619	
		d'	0.072	0.013	5.341	
		c'	0.042	0.008	5.413	
		b'	0.117	0.018	6.359	
		d'	0.005	0.001	4.690	
A-2	Genital Organs	c'	0.034	0.008	4.503	
		b'	0.011	0.003	4.159	
		a'	0.001	0.001	1.297	
		Filamentous holdfast	2.572	0.665	3.865	

Sample*	Tissues	Sections**	Wet Weight (g)	Dry Weight (g)	Wet Weight / Dry Weight	Length of the respective twig's stalks at the sections (cm)***
A-1	Twig's Stalks	g'(Apex)	0.132	0.020	6.50	10.5; 1; 1; 0.5; 0.5; 0.5; 0.5; 0.5;
		f'	0.478	0.061	7.90	1.5; 2; 2;
		e'	0.479	0.071	6.70	2; 2; 1.5; 2.5; 1; 2;
		d'	0.330	0.047	6.99	3; 2;
		c'	1.525	0.174	8.78	4; 3.5; 5.5; 4; 5; 4;
		b'	1.714	0.191	8.99	4; 3; 2.5; 3.5; 7; 5.5; 4;
A-2	Twig's Stalks	a'	N	N	N	N
		d'	N	N	N	N
		c'	0.200	0.025	8.12	1.5; 2; 1;
		b'	0.528	0.061	8.68	2; 2; 1.5; 2; 1.5; 2;
		a'	0.052	0.007	7.35	0.5; 1;
		g'(Apex)	1.718	0.139	12.36	
A-1	Twig's Leaves	f'	7.200	0.704	10.23	
		e'	5.200	0.666	7.81	
		d'	2.628	0.285	9.21	
		c'	12.041	1.238	9.73	
		b'	7.160	0.553	12.95	
		a'	N	N	N	
A-2	Twig's Leaves	d'	N	N	N	
		c'	1.356	0.096	14.15	
		b'	6.693	0.604	11.09	
		a'	0.834	0.051	16.48	
		Filamentous holdfast	2.572	0.665	3.87	

* The sample A-1 and A-2 constitute one branch, designated as A, connected to one common filamentous holdfast.

** The alphabetical letters, a', b', c', etc mean the sections from the bottom, filamentous holdfast, up to the top, apex, sectioned at an every 10 cm length.

*** Stalks and/or leaves of the twigs of the respective sections were gathered together and used for arsenic determination.

N : Not exist.

Table 2. Arsenic concentrations in the Hijiki plants, harvested at Mugisaki-Katada district, Ise Bay, expressed on the basis of wet weight.

Samples*	Tissues	Sections**	$\mu\text{g As/g}$ Wet weight	Percentage ratio (%***)
A-1	Stalks	g'	4.529	33.0
		f'	6.186	45.1
		e'	5.684	41.5
		d'	5.987	43.7
		c'	6.324	46.1
		b'	10.063	73.4
		a'	13.710	100.0
	Leaves	g'(Apex)	3.841	28.0
		f'	4.047	29.5
		e'	8.989	65.6
		d'	6.124	44.7
		c'	N	0
		b'	6.100	44.5
		a'	N	0
	Genital Organs	g'	71.829	100.0
		f'	9.726	13.5
		e'	13.317	18.5
		d'	9.973	13.9
		c'	13.934	19.4
		b'	12.709	17.7
		a'	N	0
Filamentous holdfast			14.961	
A-1-Twigs	g'(Apex)	Stalk	9.356	100.0
		Leaf	4.287	45.8
	f'	Stalk	7.666	81.9
		Leaf	5.574	59.6
	e'	Stalk	8.936	95.5
		Leaf	7.122	76.1
	d'	Stalk	7.966	85.1
		Leaf	6.552	70.0
	c'	Stalk	4.672	49.9
		Leaf	6.508	69.6
	b'	Stalk	6.978	74.6
		Leaf	6.719	71.8
	a'	Stalk	N	0
		Leaf	N	0

Samples*	Tissues	Sections**	$\mu\text{g As/g}$ Wet weight	Percentage ratio (%***)	
A-2	Stalks	d'	2.508	20.3	
		c'	4.584	37.1	
		b'	7.465	60.4	
		a'	12.354	100.0	
	Leaves	d'(Apex)	3.646	29.5	
		c'	5.090	41.2	
		b'	N	0	
	Genital Organs	a'	9.002	72.9	
		d'	9.056	8.6	
		c'	16.156	15.3	
		b'	b'	14.968	14.2
			a'	105.607	100.0
	Filamentous holdfast			14.961	
A-2-Twigs	c'	Stalk	5.478	72.8	
		Leaf	3.305	43.9	
	b'	Stalk	6.397	85.0	
		Leaf	5.622	74.7	
	a'	Stalk	7.523	100.0	
		Leaf	3.654	48.6	

* The sample A-1 and A-2 constitute one branch, designated as A, connected to one common filamentous holdfast.

** The alphabetical letters, a', b', c', etc means the sections from the bottom, filamentous holdfast up to the top, apex, sectioned at an every 10 cm length.

*** The highest value in the leaves and stalks, that in the genital organs, and that in the twigs (leaves and stalks) of either A-1 or A-2 were taken as 100%.

N : Not exist.

Note: The average values \pm SD ($\mu\text{g As/g}$ wet weight) of A-1 stalks, A-1 leaves and A-1-twigs were 7.5 ± 3.2 , 5.8 ± 2.1 and 6.9 ± 1.5 , respectively, and those of A-2 stalks, A-2 leaves and A-2-Twigs were 6.7 ± 4.2 , 5.9 ± 2.8 and 5.3 ± 1.6 , respectively.

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Table 3. Arsenic concentration in the Hijiki plants, harvested in Ise Bay, Mugisaki, Katada district, on the basis of dry weight.

Samples*	Tissues	Sections**	µg As/g Dry weight	Percentage ratio (%***)
A-1	Stalk	g'	36.801	48.5
		f'	44.408	58.6
		e'	37.411	49.3
		d'	36.738	48.4
		c'	40.028	52.8
		b'	58.038	76.53
		a'	70.686	93.2
	Leaf	g'(Apex)	40.999	54.1
		f'	51.750	68.2
		e'	67.741	89.3
		d'	53.677	70.8
		c'	N	0
		b'	75.840	100.0
		a'	N	0
	Genital Organs	g'	554.302	100.0
		f'	46.325	8.4
		e'	61.512	11.1
		d'	53.259	9.6
		c'	75.423	13.6
		b'	80.810	14.6
		a'	N	0
Filamentous holdfast			57.828	
A-1-Twig	g'(Apex)	Stalk	60.859	70.0
		Leaf	52.980	60.9
	f'	Stalk	60.548	69.6
		Leaf	56.996	65.5
	e'	Stalk	59.881	68.8
		Leaf	55.603	63.9
	d'	Stalk	55.693	64.0
		Leaf	60.326	69.4
	c'	Stalk	41.024	47.2
		Leaf	63.305	72.8
	b'	Stalk	62.727	72.1
		Leaf	86.980	100.0
	a'	Stalk	N	0
		Leaf	N	0

Samples*	Tissues	Sections**	µg As/g Dry weight	Percentage ratio (%***)
A-2	Stalk	d'	30.917	30.4
		c'	32.008	31.5
		b'	50.964	50.1
		a'	73.222	72.0
		d'(Apex)	50.042	49.2
	Leaf	c'	60.945	60.0
		b'	N	0
		a'	101.657	100.0
		d'	42.467	31.0
	Genital Organs	c'	72.746	53.1
		b'	62.255	45.4
		a'	137.004	100.0
		Filamentous holdfast		
A-2-Twig	c'	Stalk	44.454	71.3
		Leaf	46.777	75.1
	b'	Stalk	55.543	89.1
		Leaf	62.327	100.0
	a'	Stalk	55.306	88.7
		Leaf	60.221	96.6

* The sample A-1 and A-2 constitute one branch, designated as A, connected to one common filamentous holdfast.
 ** The alphabetical letters, a', b', c', etc means the sections from the bottom, filamentous holdfast, up to the top, apex, sectioned at an every 10 cm length.
 *** The highest value in the leaves and stalks, that in the genital organs, and that in the twigs (leaves and stalks) of either A-1 or A-2 were taken as 100%.
 N : Not exist.

Note: The average values±SD (µg As/g dry weight) of A-1 stalks, A-1 leaves and A-1-twigs were 46.3±13.1, 58.0±13.8 and 59.7±10.5, respectively, and those of A-2 stalks, A-2 leaves and A-2-Twigs were 46.8±19.9, 70.9±27.2 and 54.1±7.2, respectively.

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ヒジキ, *Sargassum fusiforme* (Harvey) Setchelの藻体各組織内における ヒ素元素の分布について (第6報)

片山 眞之^{1,2)}, 山本 裕子²⁾, 澤田 理恵²⁾, 片山 (須川) 洋子^{1,2)}

大阪青山大学健康科学部健康栄養学科¹⁾

岐阜女子大学家政学部健康栄養学科²⁾

要 旨

新鮮な伊勢湾産ヒジキの組織各部位におけるヒ素の分布を熱中性子放射化分析法によって定量した。採集された藻体は保冷して持ち帰り、人工海水、精製水で洗浄後、茎に沿って10cm毎に切断して仮盤状付着器部、茎部、葉部、生殖器部に分画後、各画分を凍結乾燥した。乾燥試料の一部分をポリエチレンバッグ中に封入し、研究用原子炉の炉心部に保持して20分間熱中性子を照射した。72時間の冷却期間をおいて取り出し、pure Geガンマ線検出器にて放射化された元素ヒ素量を定量した。

ヒ素は茎部、葉部ともに、茎に沿って不均一に分布していた。この結果はこれまで著者らが明らかにした日本の他の海域のヒジキの結果と一致するものであった。さらに今回は生殖器のついた試料について分析し、生殖器部には他の部位とくらべてかなりの濃度のヒ素が集積していることがわかった。このことの意味および、同海域に生育するアカモクのヒ素レベルとは異なることの意味について推察した。

キーワード: ヒジキ, *Sargassum fusiforme* (Harvey) Setchell, 生殖器, 葉, 茎, 小枝, ヒ素, ヒ素濃度分布, 熱中性子放射化分析