Distribution of Accumulated Arsenic in the Seaweed Hijiki, *Sargassum fusiforme* (Harvey) Setchell* (5)

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Summary

In the preceding papers, we reported the distribution of arsenic accumulated by Hijiki plants harvested along the seashores of Tsushima Archipelago. To investigate them harvested in some other areas, we collected Hijiki plants along the seashores of Goto Archipelago, Japan, in May 2001. Hijiki plants were separated into several parts such as leaves, stalks and filamentous holdfasts. After washing them with distilled water, the samples were lyophilized. Arsenic was determined by neutron activation analysis in the nuclear reactor of Research Reactor Institute, Kyoto University. The non-uniformity of arsenic localization in individual plants along their stems was observed as shown previously when expressed in μg arsenic per g of wet or dry weight (ppm).

Key words: arsenic; *Hizikia fusiforme* Okam.; *Sargassum fusiforme* (Harvey) Setchell; seaweeds; Goto Archipelago; neutron activation analysis

We have investigated the distribution of arsenic accumulated by Hijiki plants harvested along the seashores of Tsushima Archipelago[2, 3]. To investigate Hijiki harvested in some other areas, we collected Hijiki plants along the seashores of six districts in Goto Archipelago[4, 5], Nagasaki, Japan in May 2001. In both Archipelagoes, Hijiki plants grew to more than 100 cm in length in some districts. There were large differences in arsenic concentration along the stalk, and these differences may depend on the environmental sea water conditions[6]. This non-uniform localization of arsenic along the Hijki stalk seems to be a typical phenomenon also occurring in some other seaweeds[4].

In the present study, we intended to present distribution data of arsenic accumulated by Hijiki plants harvested along the seashores of other three districts in Goto Archipelago, Nagasaki, Japan.

Experimental

Hijiki, Sargassum fusiforme (Harvey) Setchell

We collected Hijiki plants, *Sargassum fusiforme* (Harvey) Setchell, in May 2001 along the seashores of three districts, Tomie, Taihou and Tamanoura of Goto Archipelago, Japan. Samples of the Hijiki plants were stored in an ice-box until we brought them back to our laboratory.

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*Newly proposed taxonomic name[1] of ***Hizikia fusiforme* Okam.**

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Preparation of samples[2)]

Hijiki plants were washed with artificial seawater three times and washed more than twice with distilled water. Excessive water on the samples was removed by blotting them on filter paper. Hijiki plants were cut into pieces of 10 cm in length along their main branches from the top (shoot apex) down to the filamentous holdfast, and separated into leaves and stalks. The shoot apexes of the respective main branches and filamentous holdfast (radicle-like) were separated. After washing them with distilled water, the samples were separately lyophilized in small polyethylene bags.

Arsenic determination[7)]

The amount of arsenic in each sample was determined in duplicate in comparison with the arsenic standard. The dried samples were mixed well and separately packaged in small polyethylene bags. To determine the arsenic concentration in the samples, 40 of those bags were put together in a polyethylene Neuma-Capsule, along with 10 bags of various amounts of a standard arsenic compound; two standard specimens were arranged for every 8 specimens of Hijiki.

Thermal neutron activation analysis[7)]

The samples in the Neuma-capsules were irradiated in a flux of 10^{13} neutrons $\cdot$ cm^{-2} $\cdot$ sec^{-1} for 20 min in the center position of the nuclear reactor of the Research Reactor Institute, Kyoto University. After a cooling time of 72 hr, the arsenic content in the samples was determined by gamma radiation from ^{76}As using a pure Ge gamma-detector at 559.1 keV. The energy levels of ^{60}Co and ^{137}Cs were used for calibration.

Results

Growing state of Hijiki

The Hijki plants from the three districts showed different growth rates (Table 1).

Tomie District: The Tomie district produced Hijiki plants with two to four branches from one filamentous holdfast and they were about 10 to 20 cm in length.

Taiho District: The Taihou district produced plants with three to four branches from one filamentous holdfast and they were about 10 cm or less in length. The ratios of the dry weight to the wet weight suggest that they are more mature than those of the other districts in spite of their shortness.

Tamanoura District: The Tamanoura district produced plants with three branches from one filamentous holdfast and the three branches were different in length, about 10 cm, 40 cm and 60 cm, respectively.

Arsenic contents in individual tissues

The arsenic contents in the respective sections are shown in Table 2 and Table 3.

The arsenic contents along the main branches in the respective sections (10 cm in length) were 0.02 μg to 0.6 μg (Tomie district) and 0.03 μg to 1.6 μg (Taihou district) in the leaves and 0.004 μg to 0.6 μg (Tomie district) and 0.007 μg to 0.4 μg (Taihou district) in the stalks. The samples from Tamanoura showed larger amounts of arsenic in their leaves than in their stalks. Moreover, the total amounts of arsenic in the respective sections of the samples of Tamanoura were ten times higher than those of the other districts, although their wet weights or dry weights were more or less similar to those of the other samples.

Arsenic concentrations in the tissues on the wet weight basis (Table 2)

The distribution patterns of the arsenic concentration expressed on the wet weight (g) basis were not uniform along their main branches (stalks) (Table 2), and the values were generally greater in the samples from Tamanoura than those from the other districts.

Table 1 Growth states of Hijiki plants, harvested at the Tomie, Taihou and Tamanoura districts.

Tomie

Samples*	Tissues	Sections**	Wet weight (g)	Dry weight (g)
A-1	Apex		0.67	0.07396
	Stalk	a	0.58	0.10603
		b	0.23	0.04868
	Leaf	a	7.77	0.8611
	Filamentous holdfast		0.22	0.048
A-2	Apex		0.92	0.08424
	Stalk	a	0.51	0.08246
	Leaf	a	3.81	0.39069
	Filamentous holdfast		0.22	0.048
A-3	Apex		0.94	0.0886
	Stalk	a	0.29	0.05112
	Leaf	a	2.69	0.30123
	Filamentous holdfast		0.22	0.048
A-4	Apex		0.31	0.03404
	Stalk	a	0.14	0.02568
	Leaf	a	1.02	0.12376
	Filamentous holdfast		0.22	0.048
B-1	Apex		0.39	0.0383
	Stalk	a	0.59	0.0963
		b	0.24	0.04527
	Leaf	a	6.43	0.66131
		b	0.32	0.03218
	Filamentous holdfast		0.24	0.04321
B-2	Apex		0.21	0.01908
	Stalk	a	0.06	0.00837
	Leaf	a	0.55	0.0515
	Filamentous holdfast		0.24	0.04321
B-3	Apex		0.35	0.03244
	Stalk	a	0.23	0.03681
	Leaf	a	1.01	0.09945
	Filamentous holdfast		0.24	0.04321
C-1	Apex		0.69	0.07495
	Stalk	a	0.55	0.09626
		b	0.24	0.04837
	Leaf	a	8.75	0.9534
		b	0.21	0.0237
	Filamentous holdfast		0.36	0.07672
C-2	Apex		0.26	0.02579
	Stalk	a	0.14	0.0225
	Leaf	a	1.3	0.14034
	Filamentous holdfast		0.36	0.07672

For the samples of the respective districts:

* The samples, A-1, A-2, A-3 etc mean the main branches, connected to one common filamentous holdfast (the same as expressed in A-1 column). B-1, B-2, etc and C-1, C-2, etc mean the same as A-1, A-2 etc.

** The alphabets, a, b, c, etc mean the 10 cm sections from the top down to the filamentous holdfast.

Taihou

Samples*	Tissues	Sections**	Wet weight (g)	Dry weight (g)
A-1	Apex		0.23	0.03207
	Stalk	a	0.36	0.07559
	Leaf	a	1.77	0.27842
	Filamentous holdfast		0.91	0.17623
A-2	Apex		0.08	0.0096
	Stalk	a	0.09	0.01684
	Leaf	a	0.87	0.11316
	Filamentous holdfast		0.91	0.17623
A-3	Apex		0.04	0.00515
	Stalk	a	0.25	0.05168
	Leaf	a	1.68	0.24918
	Filamentous holdfast		0.91	0.17623
B-1	Apex		0.25	0.02927
	Stalk	a	0.12	0.02188
	Leaf	a	0.93	0.11301
	Filamentous holdfast		0.56	0.09274
B-2	Apex		0.37	0.04541
	Stalk	a	0.12	0.02408
	Leaf	a	1.68	0.20359
	Filamentous holdfast		0.56	0.09274
B-3	Apex		0.44	0.05265
	Stalk	a	0.18	0.03684
	Leaf	a	2.07	0.26048
	Filamentous holdfast		0.56	0.09274
B-4	Apex		0.15	0.0192
	Stalk	a	0.21	0.03755
	Leaf	a	0.81	0.10768
	Filamentous holdfast		0.56	0.09274

Tamanoura

Samples*	Tissues	Sections**	Wet weight (g)	Dry weight (g)
A-1	Apex		0.17	0.01664
	Stalk	a	0.47	0.06342
		b	0.61	0.08822
		c	0.7	0.1147
		d	0.63	0.10985
		e	0.63	0.12042
		f	0.38	0.08912
	Leaf	a	4.33	0.41658
		b	6.03	0.64008
		c	6.9	0.77875
		d	7.41	0.82216
		e	8.21	0.85512
		f	3.51	0.3851
	Filamentous holdfast		0.08	0.02095
A-2	Apex		0.14	0.01675
	Stalk	a	0.44	0.06073
		b	0.55	0.08645
		c	0.67	0.11812
		d	0.47	0.10065
	Leaf	a	6.37	0.36202
		b	4.06	0.45431
		c	6.3	0.69734
		d	2.76	0.3261
	Filamentous holdfast		0.08	0.02095
A-3	Stalk	a	0.29	0.04891
	Leaf	a	1.19	0.12385
	Filamentous holdfast		0.08	0.02095

Table 2 Arsenic concentrations in Hijiki plants, harvested at the Tomie, Taihou and Tamanoura districts, expressed on the wet weight basis.

Tomie

Samples*	Tissues	Sections**	μg As/g Wet weight	Percentage ratio (%***)
A-1	Apex		0.0386	1.82
	Stalk	a	0.0564	2.66
		b	2.1210	100.00
	Leaf	a	0.0382	1.80
	Filamentous holdfast		0.0649	3.06
A-2	Apex		0.0297	45.79
	Stalk	a	0.0567	87.36
	Leaf	a	0.0177	27.20
	Filamentous holdfast		0.0649	100.00
A-3	Apex		0.0191	29.46
	Stalk	a	0.0395	60.78
	Leaf	a	0.0230	35.47
	Filamentous holdfast		0.0649	100.00
A-4	Apex		0.0321	49.50
	Stalk	a	0.0500	76.98
	Leaf	a	0.0410	63.10
	Filamentous holdfast		0.0649	100.00
B-1	Apex		0.0361	3.40
	Stalk	a	0.5634	53.05
		b	1.0620	100.00
	Leaf	a	0.0409	3.85
		b	0.0646	6.08
	Filamentous holdfast		0.0674	6.35
B-2	Apex		0.0375	55.61
	Stalk	a	0.0580	85.98
	Leaf	a	0.0391	57.91
	Filamentous holdfast		0.0674	100.00
B-3	Apex		0.0288	42.70
	Stalk	a	0.0624	92.51
	Leaf	a	0.0460	68.16
	Filamentous holdfast		0.0674	100.00
C-1	Apex		0.8516	34.13
	Stalk	a	0.0573	2.30
		b	0.1135	4.55
	Leaf	a	0.0711	2.85
		b	2.4948	100.00
	Filamentous holdfast		0.0632	2.53
C-2	Apex		0.0450	59.01
	Stalk	a	0.0763	100.00
	Leaf	a	0.0478	62.57
	Filamentous holdfast		0.0632	82.77

Taihou

Samples*	Tissues	Sections**	μg As/g Wet weight	Percentage ratio (%***)
A-1	Apex		0.0278	5.49
	Stalk	a	0.5071	100.00
	Leaf	a	0.0328	6.47
	Filamentous holdfast		0.0721	14.21
A-2	Apex		0.0337	1.83
	Stalk	a	1.5237	82.68
	Leaf	a	1.8429	100.00
	Filamentous holdfast		0.0721	3.91
A-3	Apex		0.0438	2.85
	Stalk	a	1.5361	100.00
	Leaf	a	0.0728	4.74
	Filamentous holdfast		0.0721	4.69
B-1	Apex		0.2038	31.04
	Stalk	a	0.6566	100.00
	Leaf	a	0.5118	77.95
	Filamentous holdfast		0.0052	0.79
B-2	Apex		0.0027	0.30
	Stalk	a	0.9160	100.00
	Leaf	a	0.0351	3.83
	Filamentous holdfast		0.0052	0.57
B-3	Apex		0.0333	2.60
	Stalk	a	1.2799	100.00
	Leaf	a	0.0249	1.94
	Filamentous holdfast		0.0052	0.40
B-4	Apex		0.0203	58.87
	Stalk	a	0.0322	93.70
	Leaf	a	0.0344	100.00
	Filamentous holdfast		0.0052	15.05

Tamanoura

Samples*	Tissues	Sections**	μg As/g Wet weight	Percentage ratio (%***)
A-1	Apex		0.9212	52.86
	Stalk	a	0.5731	32.88
		b	0.5512	31.63
		c	0.6557	37.62
		d	0.8104	46.50
		e	1.3386	76.81
		f	1.6757	96.14
	Leaf	a	0.4111	23.59
		b	0.4047	23.22
		c	0.3763	21.59
		d	0.7491	42.98
		e	1.1110	63.74
		f	1.7429	100.00
	Filamentous holdfast		0.9708	55.70
A-2	Apex		0.7660	66.17
	Stalk	a	0.5792	50.03
		b	0.0263	2.28
		c	0.2894	24.99
		d	0.8010	69.18
	Leaf	a	0.4034	34.85
		b	1.1578	100.00
		c	0.6988	60.35
		d	0.8905	76.92
	Filamentous holdfast		0.9708	83.85
A-3	Stalk	a	0.5969	45.99
	Leaf	a	1.2977	100.00
	Filamentous holdfast		0.9708	74.81

For the samples of the respective districts:
*, ** : The same as described in Table 1.
*** : The highest value was taken as 100%.

Table 3 Arsenic concentrations in Hijiki plants, hrvested in the Tomie, Taihou and Tamanoura districts, on the dry weight basis.

Tomie

Samples*	Tissues	Sections**	μg As/g Dry weight	Percentage ratio (%***)
A-1	Apex		0.3495	3.49
	Stalk	a	0.3087	3.08
		b	10.0211	100.00
	Leaf	a	0.3444	3.44
	Filamentous holdfast		0.2975	2.97
A-2	Apex		0.3246	92.56
	Stalk	a	0.3507	100.00
	Leaf	a	0.1722	49.09
	Filamentous holdfast		0.2975	84.83
A-3	Apex		0.2029	68.19
	Stalk	a	0.2238	75.23
	Leaf	a	0.2056	69.10
	Filamentous holdfast		0.2975	100.00
A-4	Apex		0.2926	86.69
	Stalk	a	0.2724	80.70
	Leaf	a	0.3376	100.00
	Filamentous holdfast		0.2975	88.13
B-1	Apex		0.3678	6.53
	Stalk	a	3.4520	61.31
		b	5.6305	100.00
	Leaf	a	0.3976	7.06
		b	0.6424	11.41
	Filamentous holdfast		0.3746	6.65
B-2	Apex		0.4128	98.97
	Stalk	a	0.4157	99.66
	Leaf	a	0.4171	100.00
	Filamentous holdfast		0.3746	89.81
B-3	Apex		0.3107	66.55
	Stalk	a	0.3898	83.50
	Leaf	a	0.4669	100.00
	Filamentous holdfast		0.3746	80.23
C-1	Apex		7.8396	35.46
	Stalk	a	0.3275	1.48
		b	0.5631	2.55
	Leaf	a	0.6523	2.95
		b	22.1060	100.00
	Filamentous holdfast		0.2965	1.34
C-2	Apex		0.4541	95.61
	Stalk	a	0.4750	100.00
	Leaf	a	0.4425	93.16
	Filamentous holdfast		0.2965	62.42

Taihou

Samples*	Tissues	Sections**	μg As/g Dry weight	Percentage ratio (%***)
A-1	Apex		0.1996	8.27
	Stalk	a	2.4152	100.00
	Leaf	a	0.2087	8.64
	Filamentous holdfast		0.3721	15.41
A-2	Apex		0.2808	1.98
	Stalk	a	8.1432	57.47
	Leaf	a	14.1684	100.00
	Filamentous holdfast		0.3721	2.63
A-3	Apex		0.3399	4.57
	Stalk	a	7.4310	100.00
	Leaf	a	0.4911	6.61
	Filamentous holdfast		0.3721	5.01
B-1	Apex		1.7410	41.34
	Stalk	a	3.6012	85.50
	Leaf	a	4.2118	100.00
	Filamentous holdfast		0.0313	0.74
B-2	Apex		0.0224	0.49
	Stalk	a	4.5647	100.00
	Leaf	a	0.2893	6.34
	Filamentous holdfast		0.0313	0.68
B-3	Apex		0.2780	4.44
	Stalk	a	6.2535	100.00
	Leaf	a	0.1976	3.16
	Filamentous holdfast		0.0313	0.50
B-4	Apex		0.1582	61.14
	Stalk	a	0.1803	69.66
	Leaf	a	0.2588	100.00
	Filamentous holdfast		0.0313	12.08

Tamanoura

Samples*	Tissues	Sections**	μg As/g Dry weight	Percentage ratio (%***)
A-1	Apex		9.4116	59.25
	Stalk	a	4.2473	26.74
		b	3.8114	23.99
		c	4.0019	25.19
		d	4.6479	29.26
		e	7.0034	44.09
		f	7.1451	44.98
	Leaf	a	4.2735	26.90
		b	3.8121	24.00
		c	3.3340	20.99
		d	6.7511	42.50
		e	10.6668	67.15
		f	15.8859	100.00
	Filamentous holdfast		3.7072	23.34
A-2	Apex		6.4027	61.88
	Stalk	a	4.1964	40.56
		b	0.1676	1.62
		c	1.6414	15.86
		d	3.7403	36.15
	Leaf	a	7.0990	68.61
		b	10.3465	100.00
		c	6.3128	61.01
		d	7.5369	72.85
	Filamentous holdfast		3.7072	35.83
A-3	Stalk	a	3.5389	28.38
	Leaf	a	12.4687	100.00
	Filamentous holdfast		3.7072	29.73

For the samples of the respective districts:
*, ** : The same as described in Table 1.
*** : The highest value was taken as 100%.

Tomie District: The concentration of arsenic expressed on the wet weight basis was smaller in the leaves than those in the stalks, reflecting the relatively smaller weight of stalk tissues. The values did not differ greatly between the respective leaves.

It is noticeable that a section or two of one branch out of several branches connecting to the filamentous holdfast (A－1 or B－1 in the Table 2) showed a higher concentration of arsenic. A similar finding was obtained in the sections of the apex and leaves of another branch (C－1 in Table 2).

Taiho District: The concentrations of arsenic were mostly higher in the stalks than those in the leaves, although the differences were not large between the sections of the leaves.

Tamanoura District: The plants from Tamanoura showed good growth rates and even the smallest values of arsenic concentrations showed higher values in comparison with the samples from the other two districts. The higher values of arsenic concentrations were observed generally in sections near the filamentous holdfast.

Arsenic concentrations in the tissues on the dry weight basis (Table 3)

The distribution patterns of the arsenic concentration expressed on the dry weight (g) basis were different along their main branches, although the patterns were not different from those on the wet weight basis.

Tomie District: Arsenic concentrations of the whole plant tissues of Hijiki plants from this district were about 0.5 to 1.3ppm in average, although the stalk of Section b of the sample A－1 and the leaf of Section b of the sample C－1 had more than ten times as high arsenic concentrations as the average.

Taiho District: The arsenic concentrations in the leaves of some sections were higher than those of the stalks, but most sections showed levels lower than one-tenth the concentrations in the leaves. The average arsenic concentration was about 1 to 2.5ppm in the whole plant tissues.

Tamanoura District: In most sections, the arsenic concentration was higher in the leaves than in their stalks; even in the stalks, the arsenic level was higher than those of the plants from the other districts, indicating about 7ppm in the average of the whole plant tissues.

Discussion

In the samples harvested in Goto Archipelago, the concentration of total arsenic was lower than those of other specimens hitherto investigated in our laboratory[7, 8]. As has been discussed in the previous report[2, 4], the arsenic level in Hijiki seems to change depending on the environmental conditions of ocean seawater. This seems to coincide with the great discrepancy of arsenic concentrations between the analyzed samples of some 'Giant clams' (*Tridacna maxima and T. derasa*) of Lizard Island and Davies Reef[6].

The arsenic content of dried Hijiki decreases during the pre-cooking processes, such as soaking it in water and thereafter discarding the water. The total amount of arsenic in daily diets is mostly derived from other marine products. Moreover, a beneficial role of Hijiki's components accelerating arsenic metabolism for arsenic detoxication is observed in the animal body[10, 11].

Seaweeds including Hijiki are rich sources of minerals[9] as well as beneficial dietary fibers[12, 13]. This usefulness will contribute to healthy life in the aging society of Japan, through traditional Japanese diets, as aimed in the policy of ***Healthy Japan 21***[14].

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