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Distribution of certain cations and anions in seaweeds and seawater of Saurashtra coast and their geochemical significance

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Na and Mg dominate over K and Ca respectively in seawater, but a reverse trend is observed in many seaweeds. This preferential accumulation of one element over another similar element is adjudged on the basis of differential percentage enrichment factor (DPEF). It is positive for K over Na (3986) and Ca over Mg (359). It reflects lower residence time (τ) values of K and Ca which are preferentially accumulated than Na and Mg possessing higher τ. Cl and I are more in brown seaweeds than in red and green seaweeds. Br is generally high in red seaweeds followed by brown and green seaweeds. The halogen content of investigated seaweeds is in the order Cl > Br > I > F, whereas in seawater the trend is Cl > Br > F > I [F values, *Indian J Mar. Sci*, **13** (1984) 47]. Their concentration factors, CF (median values), in seaweeds are in the order I > F > Br > Cl. DPEF of I, F, Br and Cl in seaweeds in relation to their other halogens of higher τ also show descending order:I over Cl > I over Br > I over F > F over Cl > Br over Cl > F over Cl. The trend of concentration of halogens in seaweeds compared to ambient medium (median values) suggests their relative uptake rates pattern. Br:F ratio in seaweeds indicates that the accumulation of one halide is independent of the other. In general, shorter the τ, more is the CF of cationic and anionic elements in seaweeds.

Though there are several reports[1,2] on the mineral constituents of seaweeds from Saurashtra coast, the relative amount of a group of elements in a single seaweed species with respect to seawater and their biogeochemical behaviour are not known. Distribution of F, Mn, Zn, Cu, Ni, Co and Mo in seawater and seaweeds collected from Diu, Porbandar and Okha along the Saurashtra coast (NW coast of India) has been reported[3-5]. In the present study, Na, K, Mg, Ca, Cl, Br and I contents of the same samples[3-5] are presented and Na:K, Ca:Mg, Br:F and Br:I ratios, their significance and the CF (concentration factor) of all the above cationic and anionic elements in relation to their reactivity (residence time[6], τ) in seawater are discussed.

Materials and Methods

The location of the sampling sites[3] and the topographical and hydrochemical features of these sites have already been reported[4,7]. Ambient seawater samples were directly collected in clean plastic buckets, filtered through GF/C filter paper and used for atomic absorption spectrometer (Varian Techtron model AA-6) analysis subsequent to separating Na, K, Ca and Mg by ion-exchange technique[8]. Cl, Br and I were directly analysed in filtered seawater as per the standard methods[9-11]. Coefficient of variation as a percentage of standard deviation between the triplicates for Na, K, Ca, Mg, Cl, Br and I respectively was 0.3, 1, 0.6, 0.5, 0.1, 0.6 and 1. After handpicking from their natural habitat in the intertidal region, seaweeds were cleaned with seawater and tap water (free from bleaching powder) followed by distilled water. Air dried samples were powdered, sieved and used for alkali and alkaline earth metals and halide analyses. Na, K, Ca and Mg were analysed[12] by AAS technique subsequent to oxidative decomposition of organic matter with acid digestion. Alkali salts (Na_2CO_3, KNO_3, NaOH) were used in digestion to prevent the volatilisation of halogen elements. Cl was estimated by Volhard silver nitrate titration method[13]. Br by the method of Saenger[14] and I as per the Larsen's[15] procedure. Coefficient of variation as percentage of standard deviation between the triplicates for Na, K, Ca, Mg, Cl, Br and I was 1-2.

Enrichment factor (EF) or concentration factor (CF) was calculated—the content of an element (dry wt basis) in seaweed divided by its concentration in seawater. Differential percentage enrichment factor (DPEF) between two similar elements, X relative to Y, was calculated using the equation[16],

$$\text{DPEF} = \frac{(\text{enrichment})_X - (\text{enrichment})_Y}{(\text{enrichment})_Y} \times 100$$

The conventional term EF or CF does not give any information on relative accumulation between two

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*Present address: Baddavani Peta, Urlam 532 425, India similar elements. However if the right hand side of the above equation is rewritten as

$$\frac{X/Y \text{ in seaweed}}{X/Y \text{ in seawater}} - 1 \times 100$$

then the ratio X/Y in seaweed : seawater (discriminate factor[17]) signifies the selective accumulation of element X over Y. Expression of discriminate factor in its differential percentage form is advantageous as it shows positive values as well as negative values with a greater dimension. Calculation[18] of least square fitting line between CF (log Y) and τ (log X) was also made.

Results and Discussion

The present values of Ca and Mg of seawater agree with those of the reported values for northern Arabian Sea water[19], Br with those of central west coast of India[20] and I with those of shoreline waters of Gujarat coast[11]. Thus the elemental distribution of coastal waters of Saurashtra is similar to that of rest of Indian west coast.

The concentration of Na, the major cationic ingredient in seawater, is about 27 times more than K concentration. Mg is about 3 times more than Ca concentration. However, in seaweeds Na and Mg are less accumulated than K and Ca respectively (Table 1). More Na than K is observed in *Cladophora fascicularis, Bryopsis plumosa, Caulerpa racemosa, Hypnea musciformis* and *Laurencia* sp. Similarly *Enteromorpha intestinalis, Valoniopsis pachynema, Ulva fasciata, Sarconema filiforma,* show high accumulation of Mg than Ca while no definite trend is observed in *Cheatomorpha antennina, C. fascicularis, Sargassum tenerrimum* and *H. musciformis.* The earlier reports from the Saurashtra coast[1,2,21] and elsewhere[22] also reveal low and inconsistent ratios of K:Na and Ca:Mg in certain seaweeds. However, it is interesting to note that whether K:Na ratio is more or less than one, all the seaweeds show positive DPEF values (Table 2) demonstrating the preferential accumulation of K over Na. Similarly, for Ca over Mg the DPEF values are also positive (with few exceptions - not shown in Table) manifesting the preferential accumulation of Ca over Mg. In this study certain species such as *E. intestinalis, V. pachynema* are designated as Mg rich plants in relation to Ca on the basis of their negative DPEF for Ca over Mg.

Halides not only show variation from species to species, but also in the same species collected from different places. Similarly no definite relation exists between the contents of all the halides (Table 1). Such variations have been reported earlier[22,23]. These variations may probably be due to the differences in relative uptake rates of halide ions, species specificacity, environmental conditions, and different phenological stages characterised by differential rate of metabolic activities[21,22].

In the present study (Table 1) the observed Br:I ratio in seaweeds is in general > 1. In brown seaweeds Br:I ratio is lower than in green and red seaweeds. This may be due to the high concentration of I in the former than in the latter. Similar trend of Br:I ratios has also been reported in cold water species[23]. Though Br:I ratio is > 1 in seaweeds, DPEF calculations show that seaweeds preferentially accumulate I over Br, 3 to 5 orders magnitude (Table 2). This is further supported by the depletion of I and conservative distribution of Br in productive waters[24].

The observed DPEF values (Table 2) for the individual halides in relation to other halides exhibit the sequence I over Cl > I over Br > I over F > F over Cl > Br over Cl > F over Br which follows closely their τ descendency. From the above data it seems that Cl is not accumulated by seaweeds whereas I, F and Br are being accumulated one order to several orders of magnitude.

Cl, Br and F are the major anionic elements in seawater and their ratio to salinity is constant[24]. They are interrelated as Br:Cl and F:Cl ratios are constant[24], which signifies their geochemical distribution. Similarly Br:F ratio should also be a constant. However, in seaweeds Br:F ratio is never a constant as it varies from 7.8 to 286 (Table 1). Therefore it is presumable that in seaweeds the internal concentration of Cl, Br and F are not interrelated, whatever may be the mechanism of their uptake/accumulation probably independent of other halogens present in the tissue.

Variations in the concentrations of F, Cl and Br in ambient seawater are negligible when compared with that of I (Table 1). Similar pattern is observed in median values of CF of these elements (Table 3). It is within one order of magnitude for F, Br and Cl whereas it is 3 orders of magnitude for I. Hence, according to Liebig-Blackman law of minimum[25] it can be tentatively presumed that the lowest concentration of I in ambient medium and its relatively high concentration in seaweeds probably makes I, a limiting element among the halogens for certain physiological process. On the basis of its high CF or high seaweed concentration against ambient concentration it can be said that uptake rate of I might be more than that of other halogens. The relative uptake rates of halogen ions in seaweeds are not known so far. However, on the basis of the present data, median values of individual halogen content in

Table 1—Chemical constituents of seawater and seaweeds (mg. kg^{-1}) and K:Na, Ca:Mg, Br:F and Br:I ratios in seaweeds

Name of the Sample	Place of collection	Na (×10^4)	K (×10^4)	Ca (×10^4)	Mg (×10^4)	Cl (×10^4)	Br	I	K:Na	Ca:Mg	Br:F* (×10)	Br:I
Seawater	D	1.077	0.040	0.042	0.129	1.942	67.50	0.052	—	—	—	—
	P	1.084	0.040	0.043	0.130	1.947	67.70	0.060	—	—	—	—
	O	1.100	0.041	0.043	0.132	1.980	68.82	0.056	—	—	—	—
CHLOROPHYTA												
Enteromorpha intestinalis (Linn.) Link	D	1.12	1.85	0.76	3.30	1.41	85	47	1.65	0.23	0.97	1.8
	P	0.94	1.25	0.91	2.98	1.38	78	36	1.33	0.30	0.87	2.2
	O	1.84	1.47	1.03	3.41	1.93	69	31	0.80	0.30	0.78	2.2
Enteromorpha sp.	D	1.74	1.02	1.25	3.68	2.67	58	39	0.59	0.34	1.0	1.5
	P	1.38	1.63	1.41	3.52	2.04	71	50	1.18	0.40	1.2	1.4
Ulva fasciata Delile	D	1.19	1.53	1.63	2.59	1.27	130	29	1.29	0.63	1.4	4.5
	P	0.73	2.02	1.48	2.81	1.83	147	37	2.77	0.53	1.6	4.0
	O	0.94	2.04	1.98	3.03	2.00	151	37	2.17	0.65	1.6	4.1
Chaetomorpha antennina (Bory) Kuetz	D	2.33	9.28	1.39	0.97	7.00	647	71	3.98	1.43	17.5	9.1
	P	1.36	6.84	0.83	0.87	6.25	604	60	5.03	0.95	20.0	12.1
	O	1.95	8.34	1.39	1.08	5.93	688	63	4.28	1.29	17.3	10.9
Cladophora fascicularis (Mertens) Kuetz	D	3.17	1.68	0.65	0.24	2.87	301	493	0.53	2.71	7.2	0.61
	P	3.50	1.50	0.47	0.54	2.35	270	467	0.43	0.87	6.7	0.58
	O	4.23	2.09	0.78	0.85	3.41	286	505	0.49	0.92	7.2	0.57
Bryopsis plumosa (Huds.) Ag	D	4.31	0.52	1.07	0.25	1.21	257	78	0.12	4.28	3.3	3.3
	P	4.50	0.70	1.30	0.32	0.93	230	70	0.16	4.06	3.1	3.3
	O	3.98	0.65	1.09	0.22	1.98	214	54	0.16	4.95	3.1	4.0
Caulerpa racemosa (Forssk.) Weber V. Bosse	D	6.46	0.89	1.75	0.90	2.84	326	110	0.14	1.94	6.8	3.0
	P	5.28	0.92	1.89	0.85	2.17	319	117	0.17	2.22	6.0	2.7
	O	7.37	1.33	2.12	1.03	3.01	342	135	0.18	2.06	6.9	2.5
Valoniopsis pachynema (Mertens) Boergs	D	3.72	5.50	0.41	1.88	3.53	373	564	1.48	0.22	2.3	0.66
	P	4.81	5.45	0.73	2.50	4.00	400	526	1.13	0.29	2.7	0.76
PHAEOPHYTA												
Dictyota dichotoma (Huds.). Lamour	D	1.19	3.27	1.83	1.37	2.41	236	131	2.75	1.33	1.4	1.8
	P	1.31	3.50	1.59	1.10	2.68	195	124	2.67	1.44	1.1	1.6
	O	2.00	5.10	2.65	1.30	3.17	243	157	2.55	2.04	1.5	1.5
Padina tetrastromatica Hauck	D	1.37	1.91	9.86	3.07	1.23	247	148	1.39	3.21	5.2	1.7
	P	1.58	2.30	9.13	3.37	1.65	229	161	1.46	2.71	4.6	1.4
	O	1.79	2.87	10.51	4.01	1.98	263	180	1.60	2.62	4.9	1.5
Padina sp.	P	0.89	1.73	8.93	2.83	1.35	349	218	1.94	3.15	4.6	1.6
Spatoglossum asperum J. Ag.	D	1.73	1.88	0.85	0.53	3.86	581	235	1.06	1.60	9.0	2.5
	P	1.31	1.37	0.70	0.60	3.57	654	191	1.05	1.17	10.0	3.4
	O	1.05	1.55	0.97	0.48	2.93	647	219	1.48	2.02	9.2	2.9
Cystoseira indica (Thivy *et* Doshi) Mairh	D	1.51	4.79	2.63	1.18	5.81	818	342	3.17	2.23	7.8	2.4
	P	2.20	4.58	3.68	1.47	5.74	726	319	2.08	2.50	8.5	2.3
	O	1.68	4.20	2.90	1.37	4.22	740	325	2.50	2.12	8.4	2.3
Sargassum johnstonii Setchell and Gardiner	O	1.34	1.50	3.58	1.45	1.73	190	225	1.12	3.47	1.3	0.84
S. swartzii (Turn.). C. Ag.	P	2.85	8.11	1.99	1.33	6.43	501	626	2.84	1.50	5.7	0.80
	O	1.93	7.26	1.73	1.40	5.89	486	589	3.76	1.24	5.3	0.82
S. tenerrimum J. Ag.	D	1.57	7.19	1.23	1.19	6.85	467	293	4.58	1.03	4.0	1.6
	P	1.90	8.27	1.48	1.97	6.21	516	310	4.35	0.75	4.7	1.7
	O	1.83	9.05	1.68	1.59	7.47	457	336	4.94	1.06	4.6	1.4
RHODOPHYTA												
Gelidiella acerosa (Forsk.) Feldman *et* Hamel	D	1.71	4.91	1.00	0.63	1.56	347	119	2.37	1.59	3.3	2.9
	P	2.00	5.23	0.98	0.78	2.21	371	134	2.61	1.26	3.8	2.8
	O	1.87	5.50	1.27	0.95	1.97	320	155	2.94	1.34	2.8	2.1
Amphiroa anceps (Lamk.) Decsne	D	0.90	1.27	27.78	3.09	0.63	256	83	1.41	9.00	1.9	3.1
	P	0.78	1.33	30.95	3.25	0.51	199	79	1.70	9.50	1.4	2.5
	O	0.85	1.48	29.37	3.75	0.75	228	100	1.74	7.80	1.6	2.3
Sarconema filiforma (Scond.) Kylin	P	1.92	5.85	1.11	1.51	2.96	129	70	3.05	0.73	2.4	1.8
	O	2.36	6.30	0.97	1.83	3.60	187	90	2.67	0.53	2.9	2.1

Contd

Table 1—Chemical constituents of seawater and seaweeds (mg. kg^{-1}) and K:Na, Ca:Mg, Br:F and Br:I ratios in seaweeds —*Contd*

Name of the Sample	Place of collection	Na ($\times 10^4$)	K ($\times 10^4$)	Ca ($\times 10^4$)	Mg ($\times 10^4$)	Cl ($\times 10^4$)	Br	I	K:Na	Ca:Mg	Br:F* ($\times 10$)	Br:I
RHODOPHYTA												
Hypnea musciformis	D	7.08	2.45	0.88	1.81	4.78	167	261	0.35	0.49	3.6	0.64
(Wulf.) Lamour.	P	6.21	1.98	1.63	1.15	3.39	138	198	0.32	1.42	3.2	0.70
	O	4.58	1.77	1.69	0.93	3.83	186	234	0.39	1.82	3.8	0.79
Gracilaria corticata	D	0.93	4.50	0.89	0.78	2.85	336	113	4.84	1.14	3.1	3.0
(Agadh.) J. Ag.	P	1.00	4.23	0.98	0.58	3.36	314	123	4.23	1.69	3.0	2.5
	O	1.28	5.95	1.15	0.90	3.79	359	145	4.65	1.28	3.3	2.5
Acanthophora spicifera	D	1.27	6.25	2.38	0.99	6.41	1425	571	4.92	2.40	7.1	2.5
(Vehl.) Boergs	P	1.53	7.61	2.91	1.34	5.85	1542	498	4.97	2.17	8.0	3.1
Chondria armata (Kuetz.)	P	3.76	4.93	2.23	1.22	1.93	3077	105	1.31	1.83	28.5	29.3
Okamura var. *plumaris* Boergs	O	2.85	4.25	1.97	1.00	1.37	3183	87	1.49	1.97	28.6	36.6
Laurencia sp.	D	4.67	2.89	5.27	1.29	0.90	460	151	0.62	4.08	9.2	3.0
	P	3.26	2.85	6.00	1.60	0.74	519	138	0.87	3.75	10.0	3.8
	O	3.91	2.33	5.85	1.73	0.85	443	121	0.60	3.38	7.8	3.7
Range		0.73	0.52	0.41	0.22	0.51	58	29	0.12	0.22	0.78	0.57
		to	to	to	to	to	to	to	to	to	to	to
		7.37	9.28	30.95	4.01	7.47	3183	626	5.03	9.50	28.6	36.6
Median		1.83	2.45	1.48	1.30	2.68	319	134	1.49	1.50	3.80	2.3

D = Diu; P = Porbandar; O = Okha

*Fluoride values taken from earlier data[3]

Table 2—Differential percentage enrichment factors (DPEF) of elements

	K over Na	Ca over Mg	I over			F over		Br over Cl	Cl over total halides
			Cl	Br	F	Cl	Br		
Median	3986	359	203100	52400	47300	346	14	223	−1.5
Range	226	−33	25800	3200	7700	−28	−82	−37	−19
	to	to	to	to	to	to	to	to	to
	13479	2823	642900	216700	299600	4096	567	6603	0

Table 3—Concentration factors, CF (median values) of cationic and anionic elements and oceanic residence time (τ) of elements

Element	CF	τ(y)*
Na	1.62	6.8×10^7
K	58.92	7×10^6
Ca	34.33	1×10^6
Mg	10.26	1.2×10^7
Cl	1.37	1×10^8
Br	4.84	1×10^8
F	6.77	5.2×10^5
I	2450	4×10^5

*Goldberg *et al.*[6]

seaweed to seawater concentration ratio (Table 3), it can be stated that the relative net uptake rates of halogens tentatively follow their CF patterns I > F > Br > Cl.

Distribution of elements in marine environment reveals an inverse relation between bioaccumulation (CF) of elements in seaweeds with their τ in seawater[18]. In the present investigation the observed values (Fig. 1) of a (intercept), b (slope), Y (correlation coefficient) are in agreement with those of the Japanese seaweeds[26].

The inverse relationship between CF and τ of two similar elements, verified in the present study by DPEF concept, shows preferential accumulation of lower τ element over higher τ element.

Why the seaweeds should preferentially accumulate K over Na and Ca over Mg proportionally from the ambient medium in which Na and Mg are dominant ingredients than K and Ca respectively, is not known, even though the physiological significance of these elements is well established[27]. However, it can be assumed that the selective accumulation of K over Na and Ca over Mg is due to the difference in the geochemical reactivities between 2 similar elements.

The concept of DPEF between 2 similar elements, X relative to Y, has been used to explain the

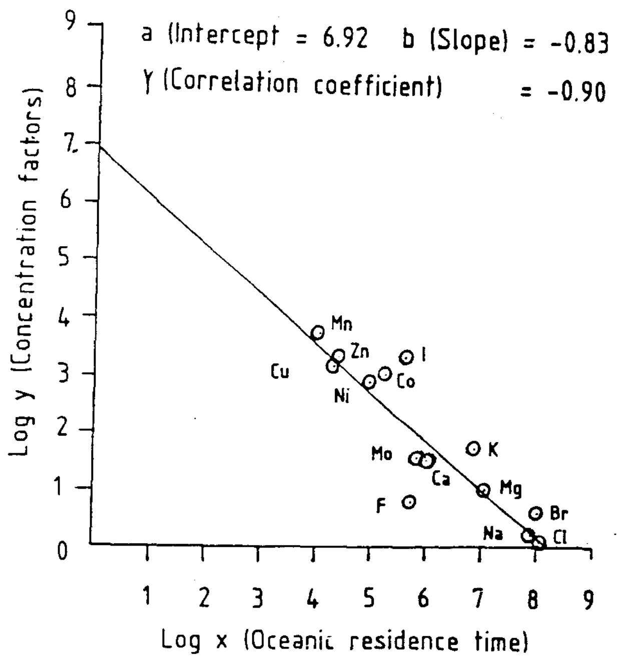

Fig. 1—Relationship between concentration factor (median values) and oceanic residence time of elements in seaweeds

lower percentage concentration of K than Na in seawater compared to river water[16]. It explains that between Na and K, the element K is geochemically more reactive (low τ) and changes more quickly than Na (higher τ) from dissolved to particulate phase, as a consequence of which it leaves the seawater system as sediment. So, it can be presumed that K and Ca, the geochemically more reactive elements, have a better chance to enter the biological system than Na and Mg, the geochemically less reactive elements.

DPEF values (Table 2) reflect the percentage abundance of K over Na (3986) and Ca over Mg (359) in seaweeds against their percentage enrichment over the seawater concentration. DPEF of K (lower τ) relative to Na (higher τ) and of Ca (lower τ) relative to Mg (higher τ) shows a wide extent of positive values (except for the latter pair of elements in few samples, not shown in Tables). Therefore, it can be inferred that K and Ca are geochemically more reactive and they change faster than Na and Mg respectively from dissolved phase in seawater to particulate/biological phase in seaweeds, thus maintaining their preferential accumulation, proportionally in seaweed tissue. Similarly I, enters biological system quicker than F, Br and Cl; F faster than Br and Cl; and Br faster than Cl according to their geochemical reactivity i.e. the accumulation of lower τ elements over the high τ elements. Thus, on the basis of DPEF calculations of individual halogens in seaweeds in relation to other halogens it is concluded that the relative uptake or bioaccumulation pattern of halogens would follow the sequence I > F > Br > Cl.

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