Biogeochemistry **12**: 71–86, 1991

Empirical relationships between the element composition of aquatic macrophytes and their underlying sediments

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Accepted 1 October 1990

Key words: aquatic plants, element composition, QSAR

Abstract. A simple view of the role of rooted macrophytes in element cycling sees them as pumps retrieving buried elements from the sediment profile. To investigate the relationship between the elemental composition of plants and sediments, we analysed published data for 39 elements. The best general model explained 84% of the variance of the log of plant element concentration:

$$\begin{aligned} \text{LPE} = &- 0.81 + 0.90 \text{ Log Sediment Element (ug/g dry wt.)} \\ &- 0.12 \text{ Sediment Organic Content (ug/g drt wt.)} \\ &+ 0.67 \text{ Atomic radius (nm) } (r^2 = 0.84; n = 39) \end{aligned}$$

This close relationship between the concentrations of an element in plant tissues and in the underlying sediment indicates that acquatic plants do not differ markedly in element composition from the sediments in which they grow. T-tests between mean residuals indicated that these aquatic plants do not discriminate between essential and nonessential elements. Model II regression analyses showed no difference between the slopes of the functional relationships for individual elements and that of the general model. When the elements were separated into three groups (alkali, transition and related metals, and halogens), Log Sediment Element accounted for 75–96% of the variation in LPE. Element physicochemical parameters were also significant independent variables explaining an additional 3–12% variation in LPE. The relative importance of the independent variables differed for the three groups of elements.

Abbreviations: AN — Atomic number, AR — Atomic radius (nm), ELEC — Electronegativity, IPD — Ionization potential differential (eV: the difference in ionization potential between the used and next lower oxidation number), IR — Ionic radius (nm), LPE — Log plant element (ug/g dry wt.), LSE — Log sediment element (ug/g drt wt.), OP — Oxidation potential (eV), ORG — Sediment organic content (ug/g dry wt.)

Contribution 272 of the Limnology Research Centre, McGill University.

Introduction

Because rooted aquatic plants are closely associated with the sediments, the plants may accumulate substantial amounts of elements from their substrate. Root uptake may therefore be a major recycling pathway for elements buried in weed-bed sediments. If this is so, contaminant levels in the plants should reflect those in the sediment, albeit modified by a number of different biotic and abiotic factors. These factors might include species specific differences in element requirements, growth form, and habitat. Other sources of variation should include the concentration of ambient element, sediment characteristics like pH and redox potential, and the availability of complexing agents such as organic matter and amorphous iron (Luoma & Bryan 1978; Campbell et al. 1988).

Factors that determine bioavailability in sediments such as trace metal binding, partitioning and remobilization are simple functions of elemental properties like ionic radius and electronegativity (Jackson et al. 1980). Campbell et al. (1988) have suggested that measurements of total sediment metal cannot predict heavy metal accumulation in aquatic organisms because the proportion of bioavailable metal varies yet various physico-chemical properties have been related to metal toxicity for *Daphnia magna* (Kaiser 1980) and mice (Kaiser 1985) and to metal accumulation by chironomid larvae (Krantzberg 1989).

No empirical relationship between the elemental composition of aquatic plants and the sediment characteristics has been published, much less one that considers the form of the element. Previous studies demonstrate that acquatic plants can accumulate heavy metals relative to concentrations in the surrounding water (Miller et al. 1983; Heisey & Damman 1982; Kovacs 1978; Dietz 1973) and nutrients from the sediments (Barko & Smart 1981; Carignan & Kalff 1980; Bristow & Whitcombe 1971) but these studies rarely involve more than a limited number of sites in a single geographical area so the generality of these patterns is unestablished. Because the costs of primary investigation are high, the analysis of published data from many locations offers an effective first step in the development of general models. This study uses that approach to develop empirical models for element concentration of aquatic plants as a function of sediment element concentration, sediment pH, organic and amorphous iron content and elemental quantitative structure-activity relationships (QSAR's).

Materials and methods

A survey of articles in *Fisheries and Aquatic Sciences Abstracts* for the years 1978—84 identified over 200 potential abstracts which yielded 26 papers containing useful information. A computer survey performed by the Aquatic Weed Bibliography Programme of the University of Florida provided 190 titles but this resulted in a nearly identical set of useful papers.

The data set derived from the literature consists of element concentrations in plants and underlying sediment as well as estimates of sediment pH, organic and iron content from lakes (78% of samples) and running water (22% of samples). When data were obtained from seasonal studies, plant values corresponding to maximum seasonal biomass were used. When separate values were given for leaves and stems, the average value was taken to represent plant element. Roots and influorescences were not included in the analysis. Descriptive statistics are presented in Table 1. A list of the dominant aquatic plant species (Table 2) shows that the major growth forms described by Sculthorpe (1976) were represented in this survey.

Although the details of experimental procedure differed, a general, composite experimental protocol applies to all studies. Dried sediment (1—15 g) was digested at 80°—100 °C with mixtures of HNO_3, $HC1$ and $HF1$. Plants were dried at 65°—105 °C and digested in HNO_3 and $HC1O_4$. Sediment and plant extracts were cooled and brought to volume (25 to 50 mL), and then analysed for element concentration by flame atomic absorption spectroscopy. Element concentrations are expressed per gram dry weight of sediment or plant.

Multiple regression analyses (SYSTAT) fit plant element concentrations to linear functions of sediment element concentrations and, when available, sediment pH, organic and iron content using the least-squares criterion. Regression analyses assume normal distributions and homoscedastic residual variance (Sokal & Rohlf 1981). When the assumption of homoscedasticity was not met, logarithmic transformation was found to remove heteroscedasticity better than a square root transformation. Quantitative variables describing the oxidation potential (OP), ionization potential differential (IPD, the difference in ionization potential at the next lower oxidation number), electronegativity (ELEC), atomic radius (AR), ionic radius (IR), atomic number (AN) and the log of AN/IPD) were obtained from Fine (1972), Kaiser (1980) and the CRC Handbook of Chemistry and Physics (1988) and were used to explain some of the residual variation around the regressions.

Table 1. Mean, minimum and maximum concentration of elements (ug/g dry wt.) in aquatic plants and their underlying sediments.

	Sediment			Plant				
Element	$\bar{X}$	min	max	$\bar{X}$	min	max	n	Source
Na	7204	2849	11200	11405	1340	15715	6	1
K*	2204.0	10.3	8169	9638.4	0.95	26130	28	1, 10
Rb	22.2	18.8	27.9	22.8	15.2	29.4	6	1
Cs	0.37	0.28	0.52	0.72	0.36	1.77	6	1
Mg*	1505	178	3909	1604	27	3300	19	1
Ca*	17883	1063	37103	17551	0.45	65120	26	1, 10
Sr	142.9	26.2	210.9	59.5	22.7	109.1	6	1
Ba	81.1	70.3	88.9	56.6	51.1	62.8	6	1
Sc	6.6	4.8	9.5	0.6	0.3	1.2	6	1
Y	10.4	7.1	13.8	61.5	37.5	68.9	6	1
La	31.7	20.9	43.9	35.1	19.8	14.8	6	1
Ce	89.6	78.6	104.7	92.9	78.6	121.4	6	1
Nd	12.9	10.3	14.5	13.2	9.4	16.0	6	1
Ti	958.1	68	2524	7.2	2.8	20	6	1
Zr	69.5	57.9	75.7	18.5	10.7	30.0	6	1
Hf	1.01	0.85	1.10	0.03	0.02	0.04	6	1
Nb	8.8	6.4	10.5	11.8	7.6	15.2	6	1
Cr	44.0	7.7	264	7.6	0.5	57.5	58	1, 6—10, 13
Mn*	374.3	87.3	884	888.9	187	2760	29	1, 7—8, 13
Fe*	27682	9734	50772	5435	1155	59480	14	1
Co	498.7	3.9	1740	65.1	2.3	860	19	8, 9
Ni	275.7	11.7	3750	65.5	1.4	840	76	1—4, 8—10
Cu*	69.3	1.9	751	18.3	2.0	141	99	1—4, 6—13
Zn*	168.2	21	1480	124.5	12	1250	86	1—6, 8—11
Cd	15.67	0.5	119	4.57	0.01	25.5	25	1, 5—6, 9—10
Al	12112	495	33871	220	189	250	6	1

Table 1. (Continued)

Element	Sediment $\bar{X}$	min	max	Plant $\bar{X}$	min	max	n	Source
Ga	4.3	3.7	4.6	0.7	0.4	1.0	6	1
Si	55207	4633	137130	4347	595	12070	6	1
Ge	3.2	2.2	4.2	0.3	0.15	0.36	6	1
Pb	2816	3	21000	52.6	3	580	41	1, 8—10, 12
P*	1041	462.6	2125	1926	980	2580	6	1, 10
As	429.1	1.3	1695	165.9	2.6	1200	11	1, 9—10
S*	17132	8164	30471	3858	2485	5210	6	1
Se	1.75	1.5	2.0	1.93	1.4	2.5	6	1
Th	1.24	0.88	1.4	1.29	0.63	1.8	6	1
F	69.8	19.9	155.9	529.4	411.8	875.9	6	1
Cl*	549.3	434	608	15310	9650	22740	6	1
Br	23.2	12.5	29.2	23.9	11.9	35.5	6	1
I	0.42	0.27	0.52	0.46	0.25	0.56	6	1

Source:
1 Cowgill 1974
2 Baudo et al. 1981a
3 Baudo et al. 1981
4 Hutchinson et al. 1975
5 Adams et al. 1980
6 Buhl & McConville 1984
7 Guilizzoni 1975
8 Ajmal & Khan 1987
9 Mudroch & Capobianco 1979
10 Mudroch 1980
11 Campbell et al. 1985
12 Welsh & Denny 1980
13 Baudo & Varini 1976

* Essential elements (Raven et al. 1981).

Table 2. Major species sampled from the lakes and rivers included in the analysis of elemental composition of aquatic plants and their underlying sediments.

System	Dominant macrophyte(s)	Author
Wanapitei R. French R. Pickerel R. Ontario	*Equisetum polustre* *Nuphar variegatum* *Potamogeton* sp. *Elodea canadensis*	Hutchinson et al. 1975
Lake Mezzola Italy	*Potamogeton crispus* *Potamogeton perfoliatus* *Potamogeton* sp. *Callitriche stagnalis* *Fontinalis* sp. *Nasturtium officinalis*	Baudo et al. 1981a
Lake Endine Italy	*Potamogeton lucens* *Najas marina*	Guilizzoni 1975
Mississippi R. Wisconsin	*Potamogeton* sp. *Elodea* sp.	Buhl & McConville 1984
Palistine L. Indiana	*Potamogeton robbinsii* *Potamogeton pectinatus* *Potamogeton crispus* *Elodea canadensis*	Adams et al. 1980
Hindon R. India	*Eicchornia crassipes*	Ajmal & Khan 1987
Linsley Pond Connecticut	*Potamogeton praelongus* *Potamogeton crispus* *Nuphar advena* *Nymphae ordata* *Decodon verticillatus* *Pontederia cordata*	Cowgill 1974
Bend Bay, Moira Lake Hawkins Bay Ontario	*Elodea canadensis* *Pontederia cordata* *Nymphae odoratae* *Myriophyllum verticillatum*	Mudroch & Capobianco 1979
Big Creek Marsh Ontario	*Typha latifolia* *Elodea canadensis* *Nuphar advena* *Nymphae odorata* *Myriophyllum heterophyllum*	Mudroch 1980
Northwestern Quebec	*Nuphar variegatum*	Campbell et al. 1985

Regression analyses

The relationship between average plant and sediment element concentration is shown in Fig. 1. In this model LSE explained 75% of the variation in LPE, and the slope and intercept are not significantly different from 1 and 0, respectively. This finding is consistent with the view that rooted aquatic plants derive most of their elements from the sediment (Denny 1972). There is no significant difference between the relationships obtained from averaged values or from individual points (Table 3; equations 1 and 2, respectively). Thus, averaging values does not appear to have a significant effect on the observed trends. Data for many elements come from one source and location (Cowgill 1974). This could have biased our results, but a regression analysis without Cowgill's data showed the regressions not to differ (the slope dropped to 0.81, the intercept changed to −0.06 and the variation explained increased to 80%), demonstrating the relationship to be general.

Figure 1 shows that the amount of sedimentary element an aquatic

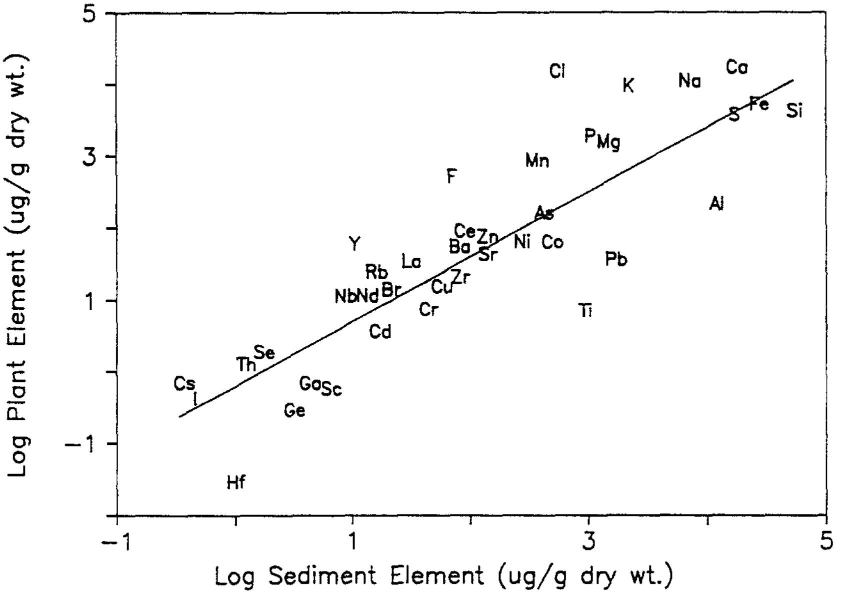

Fig. 1. The relationship between the element concentration of the above ground portion of aquatic plants and that of their underlying sediments. The symbols represent averages of all values for each of 39 elements collected in an extensive literature search. The plotted line of best fit is the equation: LPE = −0.08 (±0.21) + 0.90 (±0.09) LSE. (r^2 = 0.75; n = 39; SEest = 0.75).

Table 3. Multiple regression models for LPE as a function of all elements and subgroups based on electron configuration.

Equation	Data (n)	Model
1	All elements (39)	LPE = −0.81 (±0.21) + 0.90 (±0.09) LSE −0.12 (±0.04) ORG +0.67 (±0.31) AR $r^2 = 0.84$ SEest = 0.66
2	All elements (405)	LPE = −0.56 (±0.47) + 0.99 (±0.10) LSE −0.10 (±0.03) ORG +0.61 (±0.33) AR $r^2 = 0.74$ SEest = 0.70
3	Group 1 (21)	LPE = 1.88 (±0.35) + 0.69 (±0.09) LSE −1.71 (±0.40) Log AN/IPD $r^2 = 0.89$ SEest = 0.55
4	Group 2 (14)	LPE = −0.22 (±0.30) + 0.88 (±0.13) LSE +0.43 (±0.19) OP $r^2 = 0.86$ SEest = 0.52
5	Group 3 (4)	LPE = −0.02 (±0.38) + 1.44 (±0.21) LSE $r^2 = 0.96$ SEest = 0.47

plant absorbs depends on the concentration of the element in the sediment, but this amount may be affected by the bioavailability of the element and the selectivity of the root since some elements, e.g. Ti, K or Al, deviate from the overall trend. To examine the possibility that the scatter in Fig. 1 reflects bioavailability due to differences in sediment attributes, a series of single sediment characteristics were added in turn as a second independent variable in the regression. Both organic and iron content, which were highly correlated in our data set ($r = 0.95$), explained a small (3%) but significant ($p < 0.05$) amount of additional variation in LPE. The observed correlations between LPE and organic or iron content of the sediment were negative and therefore support the suggestion (Campbell et al. 1988) that higher levels of these sediment fractions make elements less available to biota, apparently because elements associate preferentially with the organic and iron hydroxide fraction of sediment (Forstner 1982; Tessier & Campbell 1987). Sediment pH was ineffective in explaining variation in LPE, but this likely reflects the narrow range of pH (6.17—8.32), for the effect of pH on metal speciation and bioavailability may be small in these sediment pH environments (Gambrell et al. 1976).

If aquatic plants select elements at the root epidermis, one would expect classically defined essential elements (Raven et al. 1981; Table 1) to be preferentially accumulated; Fig. 1 would then show positive residuals for essential elements and negative residuals for nonessential elements. T-tests revealed that the mean residual for essential and nonessential elements did not significantly differ from 0 (essential: $0.5 > p > 0.2$; nonessential: $p > 0.5$), nor did they differ from each other ($p > 0.5$). Since equation 1 indicates that plant burden is closely related to sediment levels and since there was no significant difference in mean residuals for essential and nonessential elements, aquatic plants appear to have no more than a limited capacity to regulate net uptake via the roots. The regulatory role of root membranes has been documented for halophytes for K^+ and Na^+ at high soil concentrations (Epstein 1969; Rozema 1976) but, in less extreme environments, ions of similar size may substitute for one another (Martin 1976). For example, organic selenium occurs primarily in proteins as sulphur analogues (Stadtman 1974) and marine algae cannot discriminate between SO_4^{3-} and Mo (Howarth & Cole 1985).

Individual element models

A sufficient data range exists to consider the relationship between macrophytes and underlying sediments for some individual elements. If the principal objective is to predict values of Y from X, Model I regression is typically employed (Snedecor & Cochran 1980); however, as the ratio of error in X relative to the range in X increases, the functional relationship (slope) between Y and X is increasingly biased in model I regressions (McArdle 1988). Thus the limited range and undiminished uncertainties associated with each element tends to bias the Model I slope calculated for each element. Model II regression provides a less biased estimate of the slope and, therefore, comparisons of the relationships of individual elements with the general model require Model II regressions. We calculated the Model II slope as b/r (Ricker 1984) where b is the Model I slope and r the Model I correlation coefficient. The variance associated with the Model II slope has been shown to be the same as that of the ordinary (Model I) regression (Teissier 1948; Kermack & Haldane 1950).

The Model II slope for the general sediment-plant relationship is 1.0 and it is this slope to which individual element slopes are compared. For the elements Cd, Co, Cr, Cu, Fe, Mn, Ni, Pb and Zn there is no significant difference between their Model II slopes and 1.0 (Fig 2). Thus, these elements have a functional relationship that is similar to that of the general model. All the Model II slopes suggest that an increase in log sediment element is accompanied by a proportional increase in log plant element. A more rigorous test would be to experimentally increase the range of sediment elements, and Pb, which includes polluted sites (Welsh & Denny

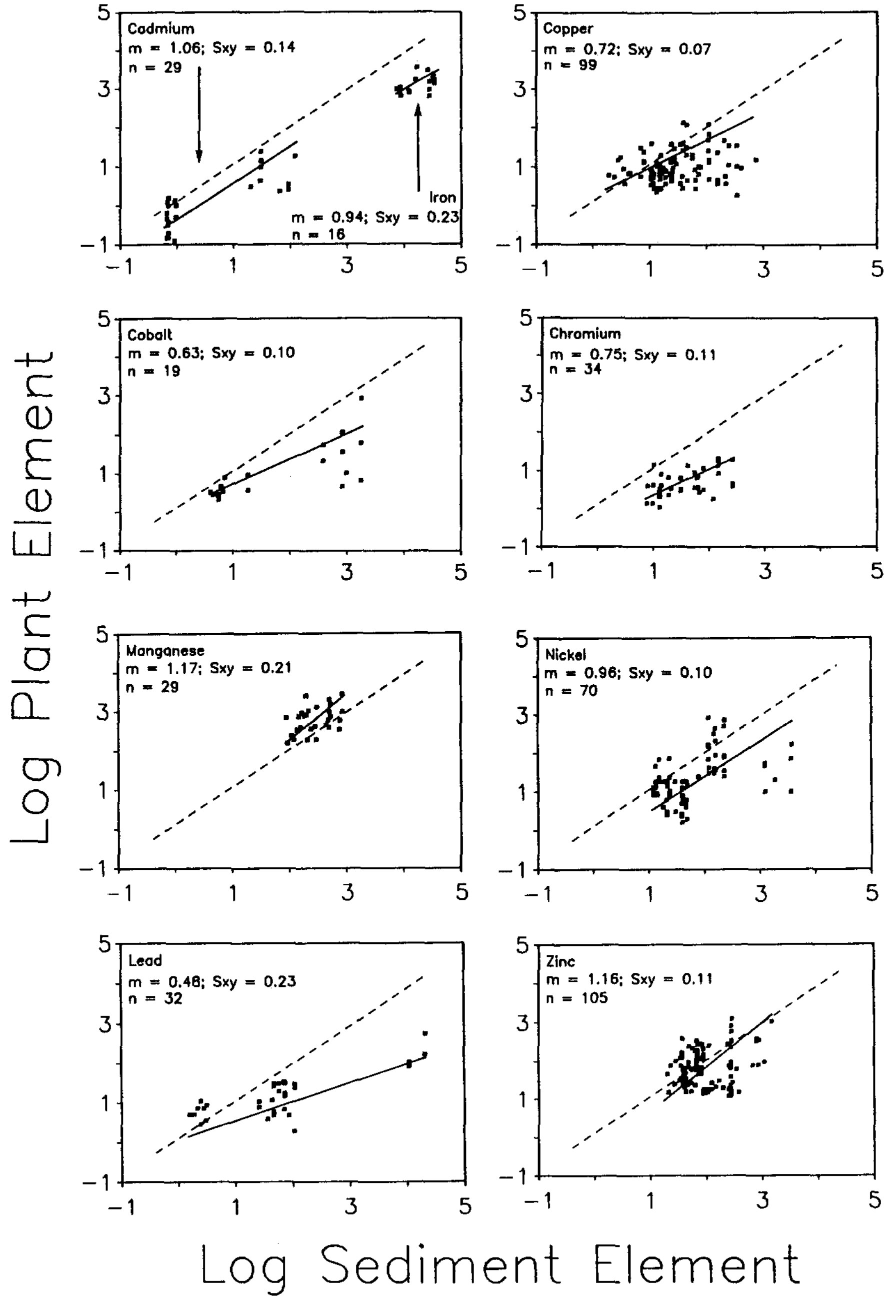

Fig 2. Model II relationships between the element concentration of the above ground portion of aquatic plants and their underlying sediments for 9 elements and the general model based on the data in Fig. 1. The functional slopes are illustrated by the dotted line for the general model (slope = 1.0) and solid line for individual elements. *Sxy* is the standard error of the slope. In some cases data points lie on top of one another.

1980) and has a sediment range of nearly 4 orders of magnitude, has the same slope as the general model.

Relationships to element physicochemical properties

Following Kaiser (1980), we separated the elements into three groups based on the number of electrons in each orbital. The first group includes ions whose oxidation states are equal to their group numbers and have completely filled *p* and *s* orbitals (Table 4). For this group, no environmental variable other than LSE was significant, but several of the QSAR variables individually explained 6—12% of the variation in LPE. Equation 3 represents the best model (Table 3).

The second group of elements consists of transition and related elements (Table 4) that have partially or completely filled *d* orbitals. They include groups in the periodic table I B through VII B, VIII and the elements from Groups IV A to VII A with oxidation numbers equal to their group number. Only LSE and oxidation potential (OP) had a significant effect on the element burden of the plants (LPE) (Table 3; equation 4).

In the third subgroup, the halogens, log sediment element explained 96% of the variation in LPE, but only represented 4 elements so additional variables are unlikely to be significant and little emphasis can be placed on the regression (Table 3; equation 5).

Jackson et al. (1980) found that non-transition metals, i.e. elements of our group 1, had a higher affinity for particulate matter than transition metals. Elements with high electronegativity should bind more closely to sedimentary material, reducing the uptake of the element and producing negative residuals. This would be consistent with our observations as negative residuals occur for highly electronegative elements (e.g., Ti and Al), and positive residuals occur for less electronegative ones, like Na and K (Fig. 1).

Jackson et al. (1980) consistently found that larger cations displayed a greater affinity for the aqueous phase in lake sediments. The positive coefficient for ionic radius likely reflects higher bioavailability due to an increased association with the aqueous phase. This trend does not apply to the subgroups based on electronic configuration; however, such categories are vertical columns for which the value of ionic radius is much less variable than that of the full periodic table.

Unexplained variation

Much of the remaining unexplained variation in our models may be due to

Table 4. Atomic properties of 39 elements. eV = electron volts. Values are from Fine (1972), Kaiser (1980) and the CRC handbook of Chemistry and Physics (1988).

Element	OP (eV)	IPD (eV)	ELEC	AR (nm)	IR (nm)	AN	Log AN/IPD
Group 1							
Ca	2.868	5.76	1.0	1.97	0.99	20	0.541
K	2.931	4.34	0.8	2.35	1.33	19	0.641
Mg	2.372	7.39	1.2	1.60	0.65	12	0.211
Na	2.71	5.14	0.9	1.90	0.95	11	0.330
Rb	2.98	4.18	0.8	2.48	1.48	37	0.947
Cs	2.92	3.89	0.7	2.67	1.69	55	1.150
Sr	2.89	5.34	1.0	2.15	1.13	38	0.852
Ba	2.912	4.79	0.9	2.22	1.35	56	1.068
Al	1.662	9.62	1.5	1.43	0.50	13	0.131
Si	1.697	11.64	1.8	1.32	0.41	14	0.080
Ti	1.63	15.77	1.5	1.47	0.68	22	0.145
Zr	1.553	11.35	1.4	1.60	0.80	40	0.547
Hf	1.505	10.00	1.3	1.67	0.81	72	0.857
Th	1.899	8.50	1.3	1.8	0.95	90	1.025
Nb	1.099	12.25	1.6	1.46	0.70	41	0.525
S	0.476	15.37	2.5	1.27	0.29	16	0.017
Sc	2.077	11.95	1.3	1.62	0.81	21	0.245
Y	2.372	8.27	1.3	1.78	0.93	39	0.674
La	2.522	7.74	1.1	1.87	1.15	57	0.867
Ce	2.483	16.52	1.1	1.81	1.14	58	0.545
Nd	2.431	16.50	1.2	1.82	1.08	60	0.561
Group 2							
Ni	0.257	10.53	1.8	1.24	0.72	28	2.659
Cu	0.342	12.56	1.9	1.28	0.69	29	2.309
Zn	0.763	8.57	1.6	1.38	0.74	30	3.501
Cd	0.403	7.91	1.7	1.54	0.97	48	6.068
Cr	0.913	17.40	1.6	1.30	0.52	24	1.379
Co	0.280	9.19	1.8	1.25	0.74	27	2.938
Mn	1.185	8.21	1.5	1.35	0.80	25	3.045
Pb	0.126	10.38	1.8	1.75	1.20	82	7.900
Fe	2.866	14.46	1.8	1.26	0.64	26	1.798
As	0.608	12.50	2.0	1.39	0.47	33	2.640
Ga	0.560	10.20	1.6	1.41	0.62	31	3.039
Ge	0.124	11.49	1.8	1.37	0.53	32	2.785
P	1.050	13.65	2.1	1.28	0.34	15	1.099
Se	0.924	13.30	2.4	1.40	0.42	34	2.537
Group 3							
F	2.866	17.422	4.0		1.36	9	0.517
Cl	1.358	12.967	3.0		1.81	17	1.311
Br	1.066	11.814	2.8		1.95	35	2.693
I	0.536	10.451	2.5		2.16	53	5.071

measurement error and/or biological selectivity. For example, the aquatic macrophyte community may influence observed patterns of element composition since plant anatomy (Franzin & McFarlane 1980; Welsh & Denny 1980), seasonal plant development (Mayes et al. 1977; Mudroch & Capobianco 1979; Larsen & Schierup 1981) and community structure (Auclair 1979; Mudroch & Capobianco 1979) have been demonstrated to affect element uptake by aquatic macrophytes. Unexplained variation could also occur if the plants take up elements from the water as Barko & Smart (1981) demonstrated for Mg^{++}.

In summary, our results demonstrate that for a wide range of environments and growth forms, aquatic plants and their underlying sediments have similar elemental composition, and that uptake of elements by the roots of these plants does not discriminate between essential and nonessential elements. Log sediment element explained 75—96% of the variation in log plant burden, and physicochemical characteristics explained up to 12% more. These models provide a first-order approximation to the relationship between element concentration of above-ground parts of aquatic plants and their associated sediments. This broad brush approach is potentially useful for determining sediment mass balances and particularly for estimating removal rates of nutrients (e.g., Carpenter & Adams 1977) or heavy metals (e.g., McIntosh et al. 1978; van der Merwe et al. 1990) by grazers or harvesting. Models that use sediment chemistry provide a background for investigating physiological processes that cause deviations from the overall trend.

Acknowledgement

This work was supported by a Natural Sciences and Engineering Research Council of Canada grant to J. Kalff.

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