

TRACE ELEMENT ENRICHMENTS IN DECOMPOSING LITTER OF *SPARTINA ALTERNIFLORA**

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ABSTRACT

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Concentrations of mercury, copper, zinc, chromium, iron and manganese were measured in *Spartina alterniflora* Loisel. during different stages of decomposition. Samples were collected from a salt marsh treated with a metal-containing sewage sludge and from a control marsh area. Overall, substantial increases were found in the concentrations of Hg, Cu, Fe, and Zn in all the *Spartina* litter samples collected. The trace-metal enrichment in the decaying *Spartina* grass was best explained by the adsorption of metal-enriched organic substances during tidal flooding of the marsh surface. No metal adsorption was observed from the sludge-treated marsh soil in spite of an elevation in several of the metals studied.

INTRODUCTION

Spartina alterniflora Loisel., the smooth marsh cord grass, is commonly found in salt marshes along the east coast of North America. This grass grows in a tall form (0.5-3 m in height) in low intertidal areas of the marsh, predominantly along tidal creeks, while a dwarf growth form (5-50 cm in height) occupies higher intertidal regions. Morphological differences, as well as some of the factors controlling the growth forms of this grass, have been described previously (Squiers and Good, 1974; Valiela et al., 1978).

Following the death and subsequent decomposition of *Spartina alterniflora*, marked increases in several heavy metals have been reported (Williams and Murdoch, 1969; Pellenbarg, 1978; Giblin et al., 1980). Pellenbarg (1978)

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found that increased Cu, Zn, and Fe in the litter of *S. alterniflora* was higher than could be explained by decay weight losses alone, and showed that an adsorption of trace metals from the aqueous surface microlayer had taken place. These processes are of interest, since it has been well-established that detritus from *S. alterniflora* forms a major link between primary and secondary productivity in coastal salt marshes (Teal, 1962; Odum and de la Cruz, 1967). Three concurring and interacting processes may be involved in metal concentration changes of dead plants during decomposition. These processes are:

- (1) relative metal increases due to decay losses of plant components with below average metal content;
- (2) absolute concentration increases caused by an adsorption of metal-enriched components by the plant material;
- (3) loss of trace metals by desorption processes, or by decay losses of metal-enriched plant parts. This study deals with the relative changes in the concentrations of Hg, Cu, Fe, Zn, Cr and Mn during the decomposition of litter of short and tall *S. alterniflora*.

The hypothesis was tested that metal concentration increases during the decomposition of *Spartina* litter were caused by adsorption of metal-rich organic components accumulated in the aqueous surface microfilm; and that the ultimate trace metal content of the litter is controlled by both sorption and desorption of metal chelates to the plant surface. To ascertain if metal concentrations in the *Spartina* litter were related to possible changes in the chemical concentration of the grasses during decay, the content of carbon, nitrogen and silver were monitored throughout the experimental period.

METHODS

Litter was collected from Great Sippewissett Marsh, Cape Cod, Massachusetts. In this marsh, experimental plots have been treated since 1974 with a metal-containing fertilizer based on sewage sludge. This material was spread over the marsh surface during low tides, twice every month, in dosages of 151.2 g m^{-2} . The metal additions took place throughout the growing seasons (April–October) of 1974–1976. Further details of experimental procedures and treatment effects on *S. alterniflora* have been presented elsewhere (Valiela et al., 1974, 1975, 1976).

During the fall of 1977, whole plants of *S. alterniflora* (ca. 400 g dry wt.) were collected from the sludge-treated marsh and from a control marsh, in the entire vegetated intertidal zone. The plants were divided into two groups, each of which were put inside a nylon bag (8 mm mesh). These bags were returned to their original location and attached to the marsh surface with aluminum pins. In subsequent sampling periods, two to ten plants, depending on their size, were retrieved from each bag; these samples were collected after 0, 32, 70, and 168 days, respectively. In addition to the dead grasses, plants surrounding the bags were also collected. This provided a

check of the validity of the experimental procedure with respect to the metal composition changes during plant decomposition. The most prominent difference between undetached and bagged plants was that standing plants remained in their upright position for several weeks during the initial period of the experiment, and were therefore often not submerged entirely during high tides.

Dead plant litter was rinsed carefully with deionized water, freeze-dried, ground in a Virtis stainless steel micro-homogenizer and analyzed for Hg, Cu, Fe, Zn, Cr and Mn. Details of the analytical procedure used for the determination of mercury have been described elsewhere (Breteler et al., 1981a). Other heavy metals were analyzed by atomic absorption spectrometry (Perkin-Elmer 403) after hot (60°C) digestion of the homogenized sample in nitric acid and oxidation with hydrogen peroxide. From each duplicate set of samples, one sample was also analyzed for carbon, nitrogen, and silver contents, using a Perkin-Elmer 240 elemental analyzer.

RESULTS AND DISCUSSION

The standing plants served as a control to check for possible effects of the bagging procedure on the experimental outcome, since the sturdy dwarf *Spartina* grasses remained standing for the majority of the experimental period. No significant differences in metal contents were found between the two types of litter collected, i.e. from litter bags and undetached plants

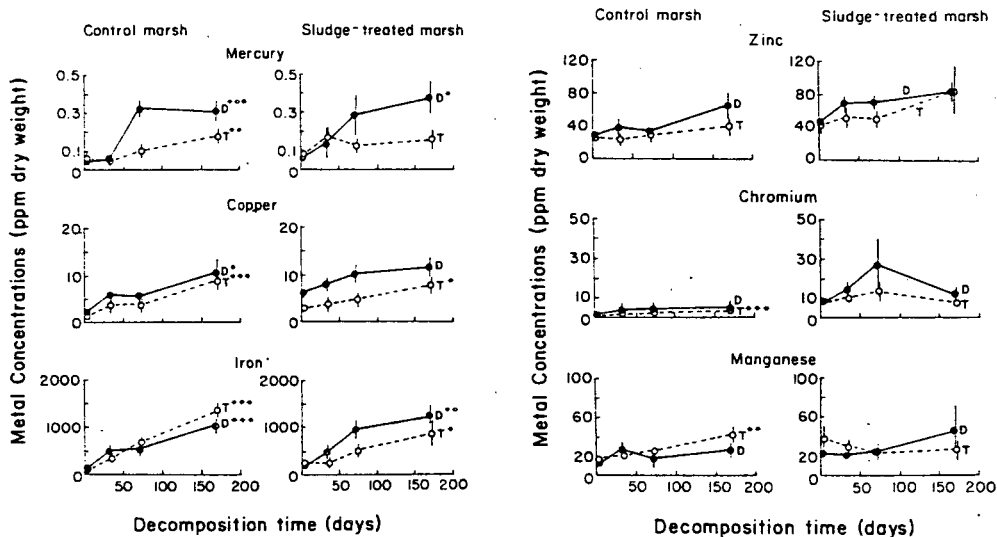


Fig.1. Relative metal concentrations (in mg kg^{-1} dry litter) of ageing litter of short form (D) and of tall form (T) *Spartina alterniflora* from control and sludge-treated salt marsh plots. Asterisks indicate the level of significance of differences between the mean concentrations within each curve (one-way analyses of variance): * = 95%; ** = 99%; and *** = 99.9%.

(analysis of variance; $P < 0.05$). To obtain more precise estimates of the mean metal concentrations, data obtained from samples collected by the two different procedures were pooled.

Changes in the metal concentrations during the decay of short and tall *S. alterniflora* are shown in Fig. 1 for litter from both control and fertilized marsh. The overall statistical significance of the metal concentration increases with time were tested (three-way analysis of variance) and found to be highly significant for Hg, Cu, Fe, ($P < 0.001$) and Zn ($P < 0.01$). There were no significant statistical interactions ($P > 0.05$) among decomposition times, growth forms of *S. alterniflora*, and fertilized vs. control grasses for any of the metals.

While concentrations of Cr in the fertilized litter appeared to rise during the first seven weeks of litter decay, levels dropped markedly towards the end of the experimental period. It is not clear whether this trend was real; but it appears to be, since it was observed in six out of eight cases. The gradual concentration increases of zinc were not steep enough to be significant at the 95% confidence level. However, the pattern of concentration increases was consistent under all experimental conditions and significant when the data were treated simultaneously (three-way analysis of variance). Mercury enrichments were considerably more pronounced in the short form than in tall litter. No differences in the mercury contents were found between control and fertilized plants, in agreement with earlier findings that this ele-

TABLE I

Heavy metal concentrations (mean \pm 3.182 S.E.) in short and tall *Spartina alterniflora* during decomposition. Litter kept on untreated and sludge-fertilized marsh plots. Calculated concentrations, assuming no loss of metal during plant decay, are in parentheses

<i>S. alterniflora</i> litter	Sampling date	% wt. loss	Metal concentrations in <i>S. alterniflora</i> (mg metal/kg litter dry wt. ⁻¹)		
			Mercury	Copper	Iron
<i>Control marsh</i>					
Short form	Oct. 11	0	0.047 \pm 0.015 (0.047)	1.60 \pm 2.23 (1.60)	126 \pm 32 (126)
	Nov. 22	22	0.052 \pm 0.033 (0.060)	5.84 \pm 0.37 (2.05)*	497 \pm 266 (162)*
	Dec. 20	26	0.326 \pm 0.125 (0.064)*	5.47 \pm 2.98 (2.16)*	573 \pm 490 (170)
	Mar. 28	36	0.304 \pm 0.170 (0.073)*	10.7 \pm 9.27 (2.50)	1143 \pm 461 (197)*
Tall form	Oct. 11	0	0.058 \pm 0.053 (0.058)	1.51 \pm 0.72 (1.51)	112 \pm 71 (112)
	Nov. 12	36	0.048 \pm 0.070 (0.091)	3.73 \pm 3.27 (2.36)	354 \pm 117 (177)*
	Dec. 20	40	0.098 \pm 0.061 (0.097)	3.96 \pm 3.20 (2.52)	701 \pm 197 (188)*
	Mar. 28	54	0.174 \pm 0.087 (0.126)	9.25 \pm 3.32 (3.28)*	1357 \pm 432 (246)*
<i>Fertilized marsh</i>					
Short form	Oct. 11	0	0.057 \pm 0.014 (0.057)	6.10 \pm 1.41 (6.10)	195 \pm 68 (195)
	Nov. 12	27	0.135 \pm 0.253 (0.078)	8.02 \pm 3.35 (8.36)	484 \pm 326 (267)
	Dec. 20	36	0.286 \pm 0.348 (0.089)	10.3 \pm 5.40 (9.52)	953 \pm 648 (304)*
	Mar. 28	50	0.378 \pm 0.266 (0.114)*	11.8 \pm 6.15 (12.2)	1240 \pm 734 (390)*
Tall form	Oct. 11	0	0.079 \pm 0.041 (0.079)	2.85 \pm 1.47 (2.85)	248 \pm 182 (248)
	Nov. 12	42	0.172 \pm 0.156 (0.136)	3.70 \pm 1.93 (4.91)	272 \pm 219 (428)
	Dec. 20	48	0.125 \pm 0.061 (0.152)	4.78 \pm 2.20 (5.48)	529 \pm 312 (477)
	Mar. 28	59	0.157 \pm 0.154 (0.193)	8.21 \pm 4.84 (6.95)	870 \pm 861 (605)

*Metal increase explained by adsorption processes.

confidence range of the actual metal values. Conversely, metal sorption probably was important in those cases where the measured means were significantly higher than the theoretical concentration increases. The results in Table I indicate that Cu, Fe, Hg, Cr, and perhaps Mn enrichments appear to have been caused by adsorption, especially in litter growing on untreated marsh.

Metal-concentration increases in the litter may be related to the stability of the metal-sediment associations. Metals which are strongly bound to insoluble soil colloids are less available for uptake by the live grasses or by the decomposing litter. Some metals, such as Cr, Zn, and Cd, may be available for uptake, but are also leached easily from the grasses (Giblin et al., 1980). With the exception of Cr, the heavy metals investigated appeared not to be sorbed from the marsh peat, in spite of the considerable metal enrichments found in response to the sewage sludge treatments.

The *Spartina* litter, therefore, may have taken up metals by sorption of

TABLE II

Rate and magnitude of trace metal enrichment in litter of *Spartina alterniflora*

Metal	Litter type	Marsh treatment	Regression slope (S.E.)	Correlation coefficient (r^2)	$f_e = \frac{[M]_{t=168}}{[M]_{t=0}}$
Hg	Short	Control	1.65 (0.92)	0.62	6.5
		Fertilized	0.76 (0.14)	0.94	6.6
	Tall	Control	1.89 (0.44)	0.90	3.0
		Fertilized	—*	0.27	2.0
Cu	Short	Control	0.049 (0.011)	0.90	6.7
		Fertilized	0.032 (0.008)	0.88	1.9
	Tall	Control	0.044 (0.006)	0.97	6.1
		Fertilized	0.032 (0.001)	1.0	2.9
Fe	Short	Control	5.7 (0.78)	0.96	9.1
		Fertilized	7.4 (0.33)	1.0	6.4
	Tall	Control	6.1 (1.47)	0.89	12.1
		Fertilized	3.9 (0.48)	0.97	3.5
Zn	Short	Control	0.23 (0.060)	0.88	2.7
		Fertilized	0.20 (0.075)	0.77	1.9
	Tall	Control	0.10 (0.020)	0.93	1.6
		Fertilized	0.23 (0.050)	0.78	2.0
Cr	Short	Control	0.014 (0.006)	0.73	2.8
		Fertilized	0.017 (0.002)	0.96	1.8
	Tall	Control	—	0.02	4.8
		Fertilized	—	0.03	1.0
Mn	Short	Control	—	0.39	2.5
		Fertilized	0.17 (0.012)	0.99	2.2
	Tall	Control	0.17 (0.040)	0.90	2.7
		Fertilized	-0.10 (0.032)	0.82	0.5

*Value not given because of poor correlation.

ment was not accumulated by marsh grasses under the experimental conditions (Breteler et al., 1981b). Although copper was accumulated by the live grasses, and concentrations of fertilized plants were higher than those of control plants during senescence (Giblin et al., 1980), no differences in copper contents were found at the end of the experimental period. Manganese exhibited the least consistent pattern of change during the litter decay; although the mean values of Mn were higher after 6 months of litter exposure than at the end of the growing season.

To ascertain whether the observed metal increases could be explained by the decay loss of plant matter, the maximum possible metal increase through this mechanism was calculated (Table I). Decay weight losses of the grasses were obtained from a decomposition study carried out parallel with and at the location of the experiment described in this report (Valiela et al., unpublished data). It was assumed that adsorption had taken place whenever the calculated metal increase was less than the lower 95% confidence limit of the measured values. It is important to remember that the calculated concentrations presume no loss of the metal during the plant decay, so that the relative importance of the metal sorption process is likely to be underestimated by these calculations. This may be especially true in cases when the inherent variability of metal concentrations of the litter resulted in large confidence intervals. Therefore, it is not certain that metal sorption had not taken place in those cases where the calculated concentrations fell within the

Zinc	Chromium	Manganese
23.9± 1.6 (23.9)	1.50± 1.31 (1.50)	10.5± 7.7 (10.5)
37.9±32.6 (30.6)	3.11± 2.49 (1.92)	26.7±12.4 (13.5)*
31.9± 4.2 (32.3)	3.79± 2.33 (2.03)	18.3±22.8 (14.2)
64.8±52.0 (37.3)	4.25± 2.32 (2.34)	26.6±17.4 (16.4)
25.0± 9.0 (25.0)	0.76± 0.28 (0.76)	16.6± 4.6 (16.6)
23.4±13.8 (39.1)	1.37± 0.89 (1.19)	21.3± 7.3 (25.9)
28.8± 9.1 (41.7)	2.45± 0.54 (1.27)*	25.7± 5.2 (27.7)
39.8±23.1 (54.4)	3.62± 1.01 (1.65)*	44.8±23.1 (36.1)
45.4±11.0 (45.4)	6.93± 3.81 (6.93)	22.2± 6.4 (22.3)
69.4±37.7 (62.2)	14.3 ±12.4 (9.49)	20.4± 6.0 (30.0)
71.7±19.9 (70.9)	34.9 ±50.0 (10.8)	25.4±24.5 (34.8)
84.3±45.2 (90.8)	12.6 ± 9.73 (13.9)	48.4±75.7 (44.6)
42.6±28.5 (42.6)	8.10± 2.96 (8.10)	38.3±43.6 (38.3)
52.3±33.7 (73.5)	10.9 ± 4.83 (14.0)	30.5±24.3 (66.0)
51.2±26.0 (81.9)	13.6 ±13.1 (15.6)	24.2±10.2 (73.6)
86.5±95.5 (103.9)	8.00± 5.75 (19.8)	20.6±21.8 (93.4)

confidence range of the actual metal values. Conversely, metal sorption probably was important in those cases where the measured means were significantly higher than the theoretical concentration increases. The results in Table I indicate that Cu, Fe, Hg, Cr, and perhaps Mn enrichments appear to have been caused by adsorption, especially in litter growing on untreated marsh.

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* Value not given because of poor correlation.

metal-enriched organics contained in the aqueous surface film (Pellenbarg, 1978). To test this hypothesis, the magnitude of metal enrichments were compared between grasses from control and fertilized plots (Table II). If metals were taken up from the marsh soil, metal concentrations would be expected to increase more rapidly in the sludge-treated plots. To measure the total metal increase during the experimental period, two parameters were used: the regression slope of the plot of the metal concentration of the litter vs. the decomposition time, provided that a high correlation coefficient was found; and an enrichment factor (fe), defined as the ratio between the final and initial metal concentration in the litter. The latter parameter provided useful information in cases where the initial metal concentration of fertilized grasses approximately equalled that of untreated plants. It was assumed that the same enrichment mechanism accounted for the metal increases of both fertilized and untreated litter. Evaluation of the results of Table II shows that metals were generally not taken up preferentially from the fertilized marsh. However, zinc uptake in tall *S. alterniflora* litter appeared to be higher in the fertilized marsh, although the total increase of zinc could be accounted for by weight loss of plant components with low zinc content during decay (Table I). Evaluation of the results of Table II further indicated that the Cr and Cu concentrations of control plants increased parallel with those of the fertilized plants. This was noteworthy, since the initial concentrations of these metals were enriched by four-fold in the sludge-treated marsh plots. This trend therefore typifies a mechanism of metal sorption from aquatic sources, by which similar amounts of the heavy metals are added to the litter, regardless of the initial concentrations.

Comparison of the enrichment factors and regression slopes of the metal increases between litter employed at high marsh sites (short form) and low marsh (tall form) made it possible to evaluate effects of tidal floodings, such as reduction—oxidation potentials, temperature, and salinity on the metal increases in the *Spartina* litter. Tall form grasses decompose more rapidly than the short form (Table I). These grasses are also more frequently in contact with metal chelates accumulated in the aqueous surface film (Pellenbarg and Church, 1979). Therefore a steeper rise might be expected in the metal concentrations of the tall form grasses. The results of Table II, however, show the inverse trend; and differences in rate and magnitude of the metal enrichments between the growth forms, therefore, seem to be related to some other factor.

Table III presents the chemical component analyses of the dead grasses from all treatment combinations. The results show that the percentages of C, H and S in the decaying grasses fluctuated with time. The nitrogen contents of the fertilized grasses dropped considerably during the first weeks of plant decay and remained unchanged thereafter. No single pattern could be discerned for the sulfur content as a function of the decomposition time. Significantly higher contents of sulfur ($P < 0.05$) and nitrogen ($P < 0.001$) were found in the short form *S. alterniflora* in comparison with the

TABLE III

Decompositional changes in C, N, and S in tall and short *Spartina alterniflora* from untreated and from fertilized marsh plots. Mean percentages ($n = 2$) \pm S.E.

Element	Marsh treatment	Growth form	Decomposition time			
			0 Days	32 Days	70 Days	168 Days
Carbon (%)	Control	Short	43.1 \pm 0.7	40.8 \pm 0.9	42.0 \pm 0.0	41.8 \pm 1.7
		Tall	41.9 \pm 0.05	40.1 \pm 1.3	41.4 \pm 0.2	42.4 \pm 1.1
	Fertilized	Short	41.9 \pm 0.8	37.7 \pm 1.6	39.7 \pm 1.5	40.6 \pm 1.0
		Tall	41.7 \pm 0.4	38.7 \pm 0.1	39.9 \pm 0.1	42.1 \pm 1.0
Nitrogen (%)	Control	Short	0.77 \pm 0.10	1.00 \pm 0.10	0.73 \pm 0.09	0.90 \pm 0.11
		Tall	0.53 \pm 0.09	0.54 \pm 0.03	0.48 \pm 0.06	0.72 \pm 0.04
	Fertilized	Short	2.54 \pm 0.01	1.48 \pm 0.0	1.60 \pm 0.56	1.63 \pm 0.38
		Tall	1.20 \pm 0.17	0.82 \pm 0.05	0.92 \pm 0.06	0.83 \pm 0.18
Sulfur (%)	Control	Short	1.22 \pm 0.07	1.16 \pm 0.02	1.60 \pm 0.48	1.25 \pm 0.12
		Tall	1.10 \pm 0.07	1.11 \pm 0.03	1.21 \pm 0.08	1.08 \pm 0.02
	Fertilized	Short	1.29 \pm 0.06	1.33 \pm 0.17	1.71 \pm 0.60	1.24 \pm 0.25
		Tall	1.23 \pm 0.15	1.10 \pm 0.04	1.17 \pm 0.13	1.30 \pm 0.01

tall form. This trend was of interest because sulfhydryl and amino groups are known to be active in the binding of heavy metals. Nitrogen contents were also significantly higher in the fertilized grasses ($P < 0.001$) due to increased uptake of nitrogen from the sludge-treated marsh (Valiela et al., 1978). However, heavy metal increases of the litter did not appear to be enhanced due to the fertilization of the marsh. The results, therefore, do not support the hypothesis that quantitative differences in nitrogenous compounds affect the heavy metal uptake of dead *S. alterniflora*. Since a similar comparison for sulfur could not be made, the role of this element in the metal enrichment process remains uncertain.

Squiers and Good (1974) measured a larger crude fiber (cellulose and hemicellulose) content in litter from the tall form of *S. alterniflora* than in litter of the dwarf form. These structural carbohydrates, low in nitrogenous compounds, form an effective sorbent for organo-metallic complexes (Pellenbarg, 1978). Thus, dead tall *S. alterniflora* may be a better sorbent for surfactant, metal-rich compounds than the short form. Since metal concentrations were generally lower in the tall form grasses, it would appear that grasses in the low elevations of the marsh were subject to a more rapid loss of loosely adhered metal-rich organic materials than litter at higher marsh elevations. Such a desorption process may, perhaps, result from a shearing force caused by tidal actions. If this is so, then the net accumulation of heavy metals during the detritus formation of *S. alterniflora* is controlled by an equilibrium between adsorption and desorption processes. The selective weight loss of plant matter with low heavy metal content during decay may further add to the metal enrichments, depending both on the characteristics of the element and the decomposition rate of the plants.

CONCLUSIONS

The chemical analyses of trace metals and other components of *S. alterniflora* during the litter decay on untreated and fertilized salt marsh plots yielded the following results:

(1) Concentrations of Hg, Zn, Cu and Fe in litter of *S. alterniflora* increased proportionately with the length of time of the litter exposure to the marsh surface. No overall enrichments were found in Cr and Mn.

(2) Metal increases were generally highest in the short growth form, which is found higher in the intertidal zone than the tall grasses. This phenomenon could not be explained by differences in the chemical composition of the growth forms of this grass.

(3) In general, no uptake of trace metals was found in litter of grasses grown within experimental plots treated with sewage sludge. Metals in the enriched plant litter therefore appear to be adsorbed as metallo-organic complexes accumulated in the aqueous surface film of the tidal waters.

(4) Metal enrichments were caused by adsorption processes, but in some incidences could be explained by decay losses of plant materials low in metal content.

(5) The ultimate trace metal enrichment in ageing *S. alterniflora* appeared to depend on the extent to which the sorbed metal complexes were held by the litter.

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