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Litter Accumulation in Woodlands Contaminated by Pb, Zn, Cd and Cu

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Summary. Close to a primary lead-zinc-cadmium smelter the standing crop of litter in woodlands was found to be elevated relative to more distant sites. The total litter accumulation is similar to that from contaminated sites reported by other authors but in this case the concentrations of heavy metals are considerably lower than those reported for other sites. Evidence is provided to support the hypothesis that within the woodlands studied, litter accumulation is not closely pH dependent, but is clearly related to both cadmium and zinc concentrations in litter. Litter accumulation occurs in certain particle size ranges and fractionation shows that the weight of accumulated litter in these size ranges is highly correlated to cadmium concentrations. These results are discussed in relation to the reported possible long term effects of metal contamination on decomposition processes and the possibility of adaptation to these adverse effects.

Introduction

Several recent publications have concerned the effect of heavy metals on woodland litter decomposition (ie. Strojan, 1978; Watson, 1975). The present paper concerns a survey of metal concentrations and litter standing crops in woodlands surrounding a primary lead-zinc-cadmium smelter at Avonmouth, England, and provides information concerning the relationships between lead, zinc, cadmium and copper concentrations and litter accumulation in these woods.

Sampling Sites, Materials and Methods

Seven woodlands were studied at varying distances from a smelting complex at Avonmouth, England. The woodlands studied were as follows:

Blaise Wood	Beech, Sycamore, Yew, Oak and Ash	ST554782[1]
Moorgrove Wood	Beech Plantation	ST552791
Hallen Wood	Oak-Hazel	ST553803
Haw Wood	Oak-Hazel	ST559800
Midger Wood	Oak-Hazel	ST795893
Leigh Wood	Oak, Beech, Sycamore, Yew, Ash and Hazel	ST552732
Wetmoor Wood	Oak-Hazel	ST745875

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[1] British Ordnance Survey Map Reference

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Woodland sampling sites were chosen for similar aspect, topography and drainage. Blaise, Moorgrove, Hallen and Haw Woods are within an area affected by prevailing winds from the smelter and receive maximal contamination (eg. Little and Martin, 1972, 1974) while the other woodlands are comparatively uncontaminated although they may well receive small amounts of contamination, especially cadmium, by long distance transport (Gill et al., 1975).

At each woodland at least 5 quadrats (15×30 cm) were placed randomly and all litter layers were removed within each quadrat. Litter was transported to the laboratory for subsequent analysis and all woodlands were sampled within a period of two weeks in the summer of 1977.

After collection, litter from each quadrat was separated into two subsamples, one was immediately weighed and then reweighed after overnight drying at 80° C. These weights were used to obtain estimates of standing crops of litter at each sampling site. The dried subsamples were then milled, further divided into subsamples, weighed into pyrex flasks, digested in boiling concentrated nitric acid, and after dilution, analysed for lead, zinc, cadmium and copper concentrations. Flame atomic absorption spectroscopy with automatic background correction was used for all analyses.

The other initial subsample of undried litter was sieved through a range of meshes, 8,4,2,1,0.5 and 0.25 mm to obtain data for the size range distribution of litter particles at the woodland sites. Samples of > 8,8-4,4-2,2-1,1-0.5 mm size range were then analysed for lead, zinc and cadmium concentrations as above.

pH was determined on unground, undried litter samples. All statistical treatments of data were carried out using GENSTAT (Lawes Agricultural Trust, 1977).

Results

Mean standing crops of litter at the woodlands studied, along with mean cadmium, lead, zinc and copper concentrations and pH are summarised in Table 1. It is apparent from these data that at woodlands close to the smelter, total standing crop of litter is greater than at more distant woodlands. Data for individual quadrats were subjected to partial regression and correlation analysis. Table 2 contains the correlation matrix between metal concentration, litter accumulation and H^+ concentration after logarithmic transformations of all data. Litter weights are closely correlated with all metal concentrations. However the correlation coefficient with H^+ is much lower although still significant ($p < 0.05$) and similar results are obtained whatever transformations and combinations of transformations are applied to the data. Partial regression shows that 76.6% of the variation in litter weights can be accounted for by the following relationship:

$$W = 0.4321\,(Zn) + 0.1784\,(Cd) + 0.0512\,(H^+) + 0.6182$$

$$SE_{rc} = 0.1703 \qquad 0.1712 \qquad 0.0329 \qquad 0.3469$$

where W = weight of litter, g/quadrat (dry weight); Zn, Cd, H^+ = zinc, cadmium and hydrogen ion concentrations; SE_{rc} = Standard Error of regression coefficients.

This confirms that the relationship of metal concentrations and pH with standing crop of litter is complex. The significance of zinc and cadmium in this relationship is confirmed by partial correlation analysis (Table 3) in which correlations between metal concentrations (including H^+) and litter accumulation have been calculated eliminating the effect of each of the other variables in turn. It is apparent that Cd and Zn concentrations are closely interrelated since partial correlation coefficients of litter with cadmium and zinc are both

Table 1. Standing crop of litter, metal concentrations (μg/g) and pH at seven woodlands surrounding the smelting complex at Avonmouth

Site	Standing crop of litter (g/m²)	Distance from smelter (km)	Aspect °N	Cadmium	Lead	Zinc	Copper	pH
Blaise ($n=5$)	13,160 ± 1,411	2.9	108	32 ± 2.5	721 ± 12	1,844 ± 116	47 ± 1.7	6.30 ± 0.14
Moorgrove ($n=5$)	8,345 ± 2,131	2.5	91	23 ± 2.3	1,052 ± 123	764 ± 187	73 ± 4.7	3.95 ± 0.09
Hallen ($n=12$)	8,343 ± 598	2.9	65	62 ± 6.1	2,179 ± 272	2,469 ± 221	135 ± 12.4	3.88 ± 0.07
Haw ($n=5$)	7,910 ± 640	3.1	75	98 ± 8.2	1,545 ± 141	2,814 ± 730	100 ± 5.9	5.03 ± 0.12
Midger ($n=10$)	3,104 ± 268	28.5	72	5.7 ± 0.7	103 ± 13	202 ± 15	15 ± 0.9	5.66 ± 0.27
Leigh ($n=5$)	1,784 ± 157	6.8	153	7.2 ± 1.0	191 ± 17	169 ± 8.2	16 ± 0.9	5.76 ± 0.10
Wetmoor ($n=10$)	913 ± 100	23.0	70	1.5 ± 0.1	44 ± 3.1	80 ± 9.1	20 ± 7.9	5.45 ± 0.24

(n = number of quadrats studied at each site)

Table 2. Correlation coefficients between Log_{10} standing crop of litter (g), metal concentrations (μg/g), and H^+ concentrations for woodlands surrounding the Avonmouth smelting complex

Variable					
Cd	0.8595***				
Pb	0.8149***	0.9308***			
Zn	0.8748***	0.9644***	0.9247***		
Cu	0.7115***	0.8766***	0.8902***	0.8416***	
H^+	0.2918*	0.4484**	0.5564***	0.4363**	0.6180***
	Litter	Cd	Pb	Zn	Cu

*$p<0.05$; **$p<0.01$; ***$p<0.001$

low when zinc and cadmium concentrations are taken into account. With both zinc and cadmium, partial correlation coefficients are highly significant when Pb, Cu or H^+ effects are accounted for.

From sieving the litter at each site the percentage weight distribution of litter within certain size ranges was obtained (Table 4). It is apparent from these data that the distribution of litter in these different size ranges changes between contaminated and relatively uncontaminated woodlands.

Metal analysis of different particle sizes show that there is an increase in concentration with decreasing particle sizes (Table 5) which is most marked in the case of cadmium at the contaminated woodlands, ie. Hallen, Moorgrove, Blaise and Haw Woods.

Table 3. Partial correlation coefficients ($r_{ml.v}$) between metal and hydrogen ion concentrations (m) and litter standing crops (l) calculated from the data of Table 4 using the method of Bailey (1959)

Variable accounted for (v)	(m)				
	Cd	Pb	Zn	Cu	H^+
Cd	–	0.0796	0.3396*	−0.1747	−0.2049
Pb	0.4767***	–	0.5496***	−0.0527	−0.3356*
Zn	0.1237	0.0325	–	−0.0945	−0.2062
Cu	0.6979***	0.5670***	0.7272***	–	−0.2677
H^+	0.8523***	0.8211***	0.8685***	0.7064***	–

*$p<0.05$; **$p<0.01$; ***$p<0.001$

Table 4. Percentage distribution of total woodland litter in different particle size ranges for seven woodland sites

Site	Size range (mm)					
	>8 mm	8–4 mm	4–2 mm	2–1 mm	1–0.5 mm	<0.5 mm
Haw	10.40	13.87	21.13	20.73	28.29	5.19
Blaise	12.76	16.19	15.35	23.76	29.70	2.03
Hallen	29.55	31.31	23.70	11.20	3.84	0.40
Moorgrove	29.98	22.28	17.64	13.60	12.42	3.30
Midger	31.37	17.90	17.89	15.84	12.90	3.35
Leigh	28.87	32.20	22.53	10.65	2.98	1.17
Wetmoor	66.42	23.49	6.33	2.38	0.82	0.38

(—— delineates contaminated and uncomtaminated woods, above and below)

Correlation and regression analysis has also been applied to these data with litter weights (dry) in each particle size range (apart from those < 0.5 mm). Correlation coefficients are shown in Table 6 along with regression coefficients obtained for litter weights on metal concentrations. Inter-metal correlations are not given but were highly significant in all size ranges ($r>0.868$, $p \ll 0.001$).

Discussion

It is apparent (Table 1) that standing crops of litter are greater in woodlands close to the Avonmouth smelting complex than in more distant, and less contaminated woodlands. This increase is closely related to metal concentrations in litter and to zinc and cadmium concentrations in particular. The results of statistical analyses (Tables 2 and 3) show that, in the sites studied, increase in litter crop is not closely related to H^+ concentration; a result which is consistent with that of Tyler (1975). The highly significant correlation for copper and lead concentrations with H^+ concentrations (Table 2) confirm our previous results showing the affinity of lead, and especially copper, for the organic frac-

Tabelle 5. Metal concentrations (μg/g)[a] in litter of different particle sizes at seven woodlands

Site	Particle size (mm)	Cd	Pb	Zn
Hallen	0.5–1.0	60 ± 7.9	2,908 ± 274	3,320 ± 306
	1.0–2.0	60 ± 7.7	3.004 ± 252	3,094 ± 336
	2.0–4.0	53 ± 7.7	3,052 ± 407	2,555 ± 270
	4.0–8.0	50 ± 9.4	2,748 ± 248	2,392 ± 261
	> 8.0	39 ± 6.2	2,334 ± 210	2,048 ± 191
Moorgrove	0.5–1.0	30 ± 3.3	1,241 ± 143	1,188 ± 308
	1.0–2.0	23 ± 2.4	1,151 ± 103	1,153 ± 100
	2.0–4.0	20 ± 2.5	1,145 ± 164	1,011 ± 107
	4.0–8.0	15 ± 3.5	946 ± 124	949 ± 79
	> 8.0	18 ± 1.9	785 ± 215	993 ± 119
Blaise	0.5–1.0	54 ± 5.4	988 ± 47	2,902 ± 194
	1.0–2.0	49 ± 4.1	880 ± 51	2,705 ± 183
	2.0–4.0	36 ± 4.5	621 ± 40	1,998 ± 152
	4.0–8.0	23 ± 3.8	389 ± 83	1,262 ± 212
	> 8.0	12 ± 2.7	219 ± 57	649 ± 166
Haw	0.5–1.0	112 ± 15	1,547 ± 149	3,511 ± 740
	1.0–2.0	104 ± 7.5	1,636 ± 246	3,490 ± 385
	2.0–4.0	86 ± 6.5	1,615 ± 77	3,384 ± 358
	4.0–8.0	59 ± 5.3	1,201 ± 174	2,565 ± 249
	> 8.0	50 ± 7.3	1,016 ± 142	1,884 ± 194
Midger	0.5–1.0	5.4 ± 0.7	157 ± 6.4	221 ± 24
	1.0–2.0	3.9 ± 0.7	157 ± 6.7	175 ± 15
	2.0–4.0	4.2 ± 0.6	133 ± 8.5	147 ± 18
	4.0–8.0	2.9 ± 0.5	97 ± 9.8	140 ± 10
	> 8.0	1.1 ± 0.2	63 ± 7.1	115 ± 10
Leigh	0.5–1.0	5.3 ± 0.4	201 ± 18	292 ± 24
	1.0–2.0	4.9 ± 0.2	170 ± 15	252 ± 16
	2.0–4.0	4.3 ± 0.2	157 ± 5.3	226 ± 15
	4.0–8.0	4.7 ± 0.7	158 ± 18	209 ± 4.4
	> 8.0	2.3 ± 0.3	122 ± 9.5	140 ± 12
Wetmoor	0.5–1.0	0.93 ± 0.22	107 ± 9.1	204 ± 5.9
	1.0–2.0	1.00 ± 0.10	84 ± 6.6	196 ± 37
	2.0–4.0	0.96 ± 0.11	64 ± 5.0	118 ± 9.4
	4.0–8.0	0.89 ± 0.06	52 ± 3.5	99 ± 5.8
	> 8.0	0.77 ± 0.11	45 ± 5.5	86 ± 6.3

[a] Mean ± S.E.

tions of these contaminated litters (Martin et al., 1976). Litter accumulation close to metal smelting activities has been reported previously by Tyler (1972, 1976) and by Strojan (1978). Strojan suggested than an increased litter accumulation could be related to decreased decomposition rates, as measured by litter-bag experiments. Similar results have been obtained for a mine-waste site by Williams et al. (1977).

The value of 8.1 kg/m^2 reported by Strojan (1978) for litter standing crops close to a zinc smelter is in close agreement with our figures of 7.9–13.2 kg/m^2 for contaminated sites. In 1976 and 1977 annual litter fall at Hallen Wood

Table 6. Correlation coefficients(r) and regression coefficients (b, a) between Log_{10} metal concentrations and litter weights within several particle size ranges. %F = percentage of variation in litter weights accounted for by regression. For discussion see text

Particle size:	0.5–1.0 mm			
Metal	r	b	a	%F
Cd	0.7502	0.8876	−0.4459	55.4
Pb	0.5096	0.8183	−1.7021	24.4
Zn	0.5068	0.8033	−1.8042	24.1
1.0–2.0 mm				
Cd	0.8141	0.8124	−0.2343	65.6
Pb	0.6527	0.8243	−3.4008	41.4
Zn	0.6726	0.8563	−3.9221	44.1
2.0–4.0 mm				
Cd	0.8501	0.7137	0.5338	71.7
Pb	0.7981	0.7424	−2.1311	62.9
Zn	0.7701	0.7584	−2.6323	58.5
4.0–8.0 mm				
Cd	0.7710	0.4733	1.5059	58.6
Pb	0.8147	0.5374	−0.5970	65.7
Zn	0.8144	0.6013	−1.2136	65.6
>8 mm				
Cd	0.5180	0.1992	2.4028	25.3
Pb	0.5893	0.2455	1.3943	33.4
Zn	0.5580	0.2811	1.0624	29.7

Table 7. Metal concentrations in litter from sites where litter accumulation has previously been reported (μg/g dry weight)

Smelter type	Date smelter started operations	Current metal conc.	Reference
Primary Pb	1968	Pb = 60,000 Zn = 2,000 Cu = 1,000 Cd = 100	Watson (1975)
Primary Zn	1911	Pb = 2,333 Zn = 25,750 Cu = 340 Cd = 885	Strojan (1978)
Brass Mill	1661	Pb = 870 Zn = 18,800 Cu = 14,900 Cd = 53	Tyler (1975, 1976)

was recorded (Coughtrey, unpublished) and was 250 and 300 g/m^2/annum respectively. These data are close to the 320 g/m^2/annum quoted by Bray and Gorham (1964) for deciduous angiosperm woodlands in general, to 210–380 g/m^2/annum quoted by Ovington (1962), and to 212–325 g/m^2/annum for leaf fall of two English oakwoods (Brown, 1976). Apart from Ovington's figure of 370–600 g/m^2 there are few data for the normal standing crop of litter within deciduous woodlands. However, if 900 g/m^2 (that of Wetmoor) is chosen as a background figure for standing crop, then the excess litter at Hallen Wood represents 25–30 years of litter input and as such may represent a large proportion of the total capital of nutrients involved in cycling within this woodland. Smelting activities began at the Avonmouth site in 1929, 48 years before this study was completed. Compared to previous studies in which litter accumulation has been reported, metal concentrations in our contaminated woodlands are rather low. Table 7 summarises data for metal concentrations in litter from sites where litter accumulation has previously been reported. Cadmium, lead and zinc concentrations were an order of magnitude, and copper two orders of magnitude higher than those recorded in this study (Table 1).

Tyler (1972) reported higher metal concentrations in more humified components of litter and our data for metal concentrations in different particle sizes (Table 5) support this observation. It is also apparent from Table 6 that the most significant correlations between metal concentrations and litter weights are for particle sizes of 1–8 mm. Litter cadmium concentrations and total standing crop are highly correlated (Tables 2 and 3, Fig. 1), however when litter is fractionated the correlation is weakest for particles > 8 mm. Figure 2 demonstrates the cadmium regression line for different particle size ranges (data from Table 6), and shows the slope of this line tending to decrease with increasing particle size; strongly supporting the view that the influence of metal contamination is not at the initial stage of litter fragmentation but at a later stage involving more fragmented material. This is further supported by the data for percentage distribution of litter in different particle size ranges (Table 4) which are summarised for contaminated and relatively uncontaminated woodlands in Fig. 3.

The data presented augment those of other authors and support the hypothesis that litter decomposition in woodlands is, in some way, affected by heavy metal contamination. This occurs at relatively low metal concentrations and in certain size ranges of litter (ie. 0.5–4 mm) and in these size ranges, litter accumulation is most closely related to cadmium concentrations. Other authors do not appear to agree as to the causes of litter accumulation in metal contaminated areas; Strojan (1975, 1978) supports the view that litter accumulation is related to the absence of certain groups of invertebrates, while Jordan (1971) and Jordan and Lechavalier (1975) take the view that litter accumulation is the result of reduced microbiological activity in contaminated sites. There is evidence that numbers and activity of soil micro-organisms are decreased by high concentrations of heavy metals (eg. Cole, 1977; Imai et al., 1975; Liang and Tabatabai, 1977; Foully, 1976) and there are also reports that micro-organisms can become tolerant to metals deposited via aerial contamination (ie. Tatsuyama et al., 1975a, b; Uchida et al., 1973; Coughtrey et al., in press).

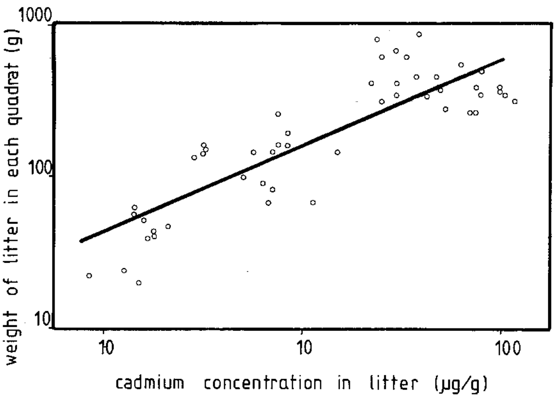

Fig. 1. Plot of litter standing crops against litter cadmium concentrations and regression line for woodland sites studied

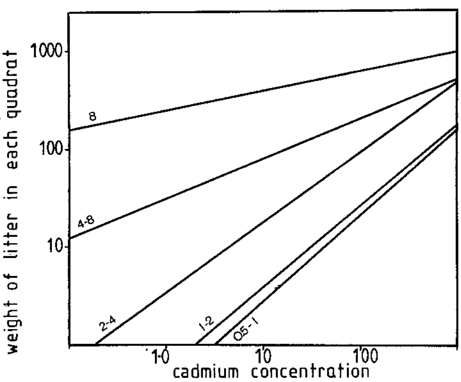

Fig. 2. Regression lines for litter weights in different size ranges against respective cadmium concentrations

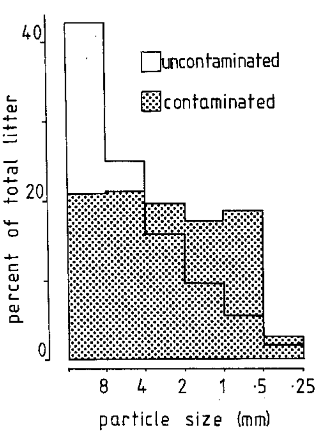

Fig. 3. Mean percentage distribution of litter in different size ranges in contaminated and uncontaminated woodlands

Furthermore, at Avonmouth sites certain groups of invertebrates do take up considerable quantities of metals (Martin and Coughtrey, 1976; Coughtrey and Martin, 1976) although at present there are no published data describing adverse effects of these metals on decomposer invertebrates, nor are there any published data concerning the ability of these organisms to produce tolerant races. In comparison, in some lead and copper contaminated rivers tolerant races of the isopod *Asellus meridianus* occur (Brown, 1976, 1977). Contamination at Avonmouth has, however, allowed the development of lead-zinc-cadmium tolerant races of higher plants (Coughtrey and Martin, 1977).

The relationships of time scale and magnitude of contamination with litter decomposition processes and tolerance in metal contaminated sites have received little attention. The long term result of metal contamination may be a reduction in ecosystem productivity as suggested by Tyler (1972) and further discussed by Strojan (1978), but there is little evidence to support this hypothesis. The soil-litter component of the ecosystem is an important sink for metal contaminants and, in the case of cadmium at Hallen Wood, contains in excess of 90% of the total ecosystem load (Coughtrey, unpublished). An important factor in the long term may be the ability of soil fauna and microflora to develop tolerance to metals. Such tolerance could result in increased litter comminution and decomposition leading to a release of previously unavailable metal reservoirs. This release would complicate any predictions of the effects of heavy metal contamination on woodland ecosystems.

Apart from the recent study of Van Hook et al. (1977) there are no detailed studies of heavy metal cycling in deciduous woodlands. Until such studies are available, along with details of the long term action of certain metals on soil biological and chemical activity, it appears to be unwise to speculate about the effects of heavy metals on ecosystem functioning.

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