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Contamination of the O2 Soil Horizon by Zinc Smelting and its Effect on Woodlouse Survival[1]

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ABSTRACT

Samples of litter from the O2 horizon of Dekalb soil (loamy-skeletal, mixed, mesic Typic Dystrochrept) were collected from 18 ridgetop sites on a transect that ran by two Zn smelters in Palmerton, Pa. Metal concentrations increased by regular gradations from a minimum at a site 105 km west of the smelters (67 mg/kg Zn, 0.85 mg/kg Cd, 150 mg/kg Pb, 11 mg/kg Cu) to a maximum 1.2 km east of the smelters (35 000 mg/kg Zn, 1300 mg/kg Cd, 3200 mg/kg Pb, 280 mg/kg Cu), and then decreased until they reached an eastern minimum at the easternmost site, 19 km from the smelters. An increase in the P concentrations near the smelters showed that the emissions were disrupting nutrient flow through the ecosystem. An increase in the pH near the smelters was attributed to the high concentrations of Zn. The log of the distance of the sites from the smelters was significantly correlated ($r = -0.80$, $p < 0.05$) with the mortality of woodlice (*Porcellio scaber* Latreille) fed samples of the litter during an 8-week test. There was substantial mortality of woodlice observed even in the O2 litter collected 19 km east of the smelters. Zinc, cadmium, lead, copper, and sulfur were experimentally added, alone or in combination, to O2 litter collected far from any known source of metal emissions. The highest concentration of Zn added (20 000 mg/kg) was toxic enough to account for the mortality observed in the earlier test. A lower concentration of Zn (5000 mg/kg) as well as the concentration of Cd (500 mg/kg) tested also significantly ($p < 0.05$) increased the mortality of woodlice.

Additional Index Words: Porcellio scaber, lead, cadmium, copper, phosphorus, pH.

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Strojan (1978a) showed a reduction in the densities of a wide variety of arthropods inhabiting the litter on Blue Mountain near two Zn smelters in Palmerton, Pa. Emissions from the smelters have done much environmental damage, including stunting and killing of forest trees (Jordan, 1975), reducing the density and species diversity of lichens (Nash, 1975) and mosses, decreasing the numbers of bacteria, actinomycetes, and fungi in the soil (Jordan & Lechevalier, 1975), and poisoning of farm animals (Gunson et al., 1982) and wildlife (Beyer, 1983). The organic horizons near the smelters are abnormally thick and have a reduced rate of respiration compared with organic horizons farther from the smelters (Strojan, 1978b). Knowledge of the metals in the litter is critical to understanding their movement through the ecosystem. The O2 litter is where much of the aerially deposited emissions from the smelters becomes concentrated, and it is an important source of both nutrients and contaminants to plants, to organisms

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feeding in the litter, and to animals higher in the food chain. Tyler (1972) suggested that the organisms in an ecosystem that are most sensitive to heavy metal contamination may be the decomposers of plant material.

Previous studies of woodlice (*Oniscus asellus*) have shown that they may accumulate high concentrations of metals (Martin et al., 1976), that the concentration of Cu in their bodies is a good indicator of Cu concentrations in litter (Wieser et al., 1976), and that the uptake of Ca is influenced by the uptake of Pb (Beeby, 1978). The present study shows that woodlice may also be used to evaluate the toxicity of litter contaminated with metals.

The O2 litter at Palmerton was sampled previously by Buchauer (1973), who concluded that the smelter emissions greatly increased the concentrations of metals near the smelters, and that the theoretical limits of the contamination of Zn, Cd, Pb, and Cu were 19 to 39 km to the east and 12 to 16 km to the west. The present study examines the contamination of the O2 litter in detail; it shows the extent of the contamination by analyzing litter collected as far away as 100 km from the smelters, shows how the survival of woodlice (*Porcellio scaber* Latreille) in litter is related to the distance from the smelters, and shows that the concentrations of Zn in some of the samples of litter are toxic.

METHODS

Collection of O2 Soil Litter

Eighteen sites were selected along a transect at various distances from two Zn smelters in Palmerton, Pa. Distances were measured to the main smokestack of the east plant. Fifteen of the sites were along a line on Blue Mountain, extending from a site 19 km east of the smelters to a site 72 km west of the smelters. At its closest point the transect passed about 1 km south of the smelters. Three western sites were located on Peters Mountain (85 km) and Broad Mountain (98 km and 105 km). The soils at all sites belonged to the Dekalb series (loamy-skeletal, mixed, mesic Typic Dystrochrept), and were either a very stony loam or a very stony sandy loam (Ackerman, 1970; Fisher et al., 1962; Kunkle et al., 1972; Staley, 1974). No sites farther than 19 km east of the smelters were sampled because it was thought that industrial emissions from New Jersey might obscure the pattern of deposition of metals from the smelters. All sites were ridgetops, had an elevation of between 425 and 475 m, and were located at least 0.5 km from all roads. The prevailing winds in the region were from the west (Kunkle et al., 1972; Staley, 1974).

At each site 15 samples of the O2 litter horizon were collected with trowels and were combined into one composite sample. The O2 horizon is defined as an "organic horizon in which the original form of most plant and animal matter cannot be recognized with the naked eye" (SCS, 1975). Additional, relatively uncontaminated O2 litter was collected from a site on Broad Mountain, 98 km to the west of the smelters, and far from any other known significant sources of metal emissions. Roots and other coarse fragments were removed from the samples, and the litter was subsampled for chemical analysis.

Handling of Woodlice and Litter

Porcellio scaber Latreille, a woodlouse, was selected as a litter arthropod to test the toxicities of the litters collected at different sites. It is common in the northern United States and is frequently associated with human habitations (Van Name, 1936). Its selection was not meant to imply that it is normally an important part of the fauna of Dekalb soil or that it normally feeds solely on O2 litter. The woodlice were collected in July from the grounds of a private residence in Bowie, Md. and were kept in the laboratory for a few weeks to allow them to acclimate. All animals used were at least 6 mm long. Gravid females were identified by their brood pouches. Remaining females were separated from the males after they were narcotized with CO_2 and examined under a microscope.

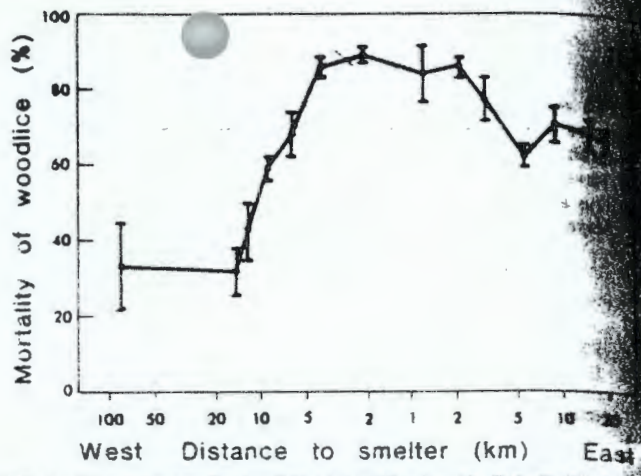

Fig. 1—Percent mortality (± SE) of woodlice kept in Dekalb O2 litter collected at various distances from two Zn smelters in Palmerton, Pa. The most western point is an average from five sites (one replicate of each) and the other points are from individual sites (four replicates of each).

The woodlice were kept at about 20°C in open plastic "freezer" boxes (15 by 15 by 20 cm high) that contained about 500 mL of moistened O2 litter, a shallow clay dish to provide shelter, and a dish of $CaCO_3$ to ensure that they had adequate Ca for their exoskeletons. Four replicate boxes were prepared from the litters of the easternmost sites. The five westernmost collection sites were thought to be far enough away from the smelters (63–105 km to the west) their litters to be considered as "controls" and, consequently, only one replicate was prepared from the litter from each of these sites. Seven male and seven female woodlice were put into each container. Once a week about 20 mL of distilled water was added to the litter. Surviving woodlice were counted after 2, 4, 6, and 8 weeks.

After this first experiment showed that some of the samples of litter were toxic to the woodlice, a second experiment was designed to identify the toxic agent. Zinc, cadmium, lead, copper, and arsenic (either alone or in combination) were added to the relatively uncontaminated O2 litter collected 98 km to the west of the smelters (Table 2). The concentrations used were those that would be present at about 4 or 5 km east of the smelters. A lower concentration of Zn (50 mg/kg) was also tested. Metals were added as oxides (ACS or Fisher certified), since they are the main emissions from the smokestack (Buchauer, 1973). The samples were shaken in an Eberbach mechanical shaker to disperse the metal powders evenly. The procedure was tested with a dye (methylene blue) to be sure that the samples were adequately mixed. Each treatment was replicated six times, with six male and six female woodlice to a replicate. In other respects, the experimental procedures were the same as those used in the first experiment. Survival of woodlice in the various treatment groups was compared with survival of woodlice in the control group by means of analysis of variance (ANOVA) and Dunnett's (1955) procedure. Dunnett's procedure allows the simultaneous comparison of several treatments to a control at a significance level that applies to all comparisons jointly.

Chemical Analyses

Samples of the O2 litter were analyzed at the Soil and Plant Analysis Laboratory of the University of Georgia; Ca, Mg, K, and Na were determined by plasma spectrograph, soil pH with a Corning Model 7 pH meter, soil-buffer pH with Adam's lime buffer solution, and S by a Leco sulfur analyzer. Cation exchange capacities were calculated by adding together the milliequivalents of Ca, Mg, K, and Na. Organic matter was determined by dry combustion at the Soil and Forage Testing Laboratory at Pennsylvania State University.

Metal concentrations were determined at the Patuxent Wildlife Research Center. Five-gram portions of the homogenized litter were dried in Vycor crucibles for 2 h at 110°C, then ashed overnight at 550°C in a muffle furnace. The cooled ash was dissolved in 3 mL HNO_3 and 1 mL HCl over a hot plate, transferred to a 12-mL polypropylene tube, and diluted to 10 mL with distilled, deionized water.

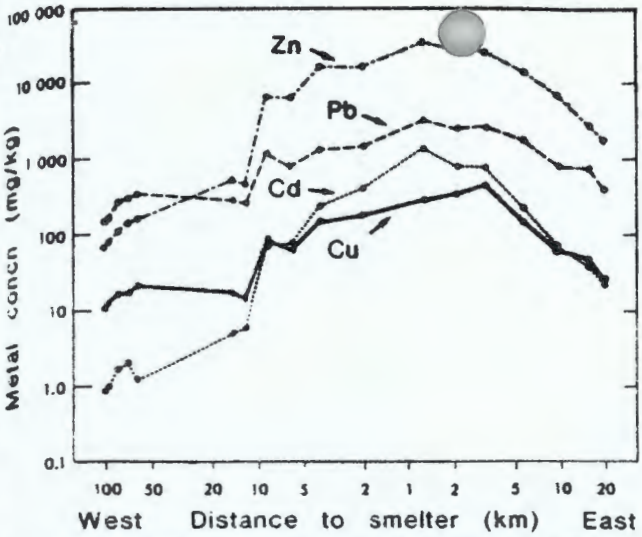

Fig. 2—Concentrations (dry wt.) of Zn, Cd, Pb, and Cu in O2 litter from Dekalb ridgetop soils collected at various distances east or west of two Zn smelters in Palmerton, Pa.

Determinations were made on a Perkin-Elmer model 703 atomic absorption spectrophotometer, by comparison to aqueous standards at 213.9 nm (Zn), 228.8 nm (Cd), 217.0 nm (Pb), and 324.7 nm (Cu) wavelengths. Background correction was used for Pb and Cd. Recoveries from spiked samples ranged from 70 to 101%, and the lower limit of quantification was 0.1 mg/kg. Concentrations were reported as mg/kg, dry weight.

RESULTS

The mortality of woodlice after 8 weeks was highest (84–89%) in the O2 litters collected nearest the smelters and decreased gradually to about 32% in the O2 litters collected from sites 15 km or more west of the smelters (Fig. 1). Substantial mortality was observed even in the O2 litter collected 19 km east of the smelters. The mortality was significantly correlated ($r = -0.80$, $p < 0.05$) with the log of the distance from the smelters (Table 1). Because the mortalities during each of the 2-week intervals were similar, only data collected after 8 weeks were shown.

Concentrations of metals in the O2 litter also were related to the distance of the collecting sites from the

Table 1—Correlation coefficients (r) relating the log of the distance to the smelters and the mortality of woodlice to 12 litter variables.

Litter variable	Log of distance to smelter	Mortality of woodlice
Mortality of woodlice	−0.80*	-
Log of conc of metals in litter		
Zn	−0.96*	−0.85*
Cd	−0.96*	−0.87*
Pb	−0.91*	−0.82*
Cu	−0.90*	−0.80*
Other litter variables		
S	−0.64*	−0.56*
pH	−0.78*	−0.71*
Ca	−0.39	−0.50*
Mg	0.45	0.28
K	0.78*	0.80*
P	−0.68*	−0.66*
Organic matter	0.43	0.33
Cation exchange capacity	0.43	0.18

* Significant at the 0.05 level.

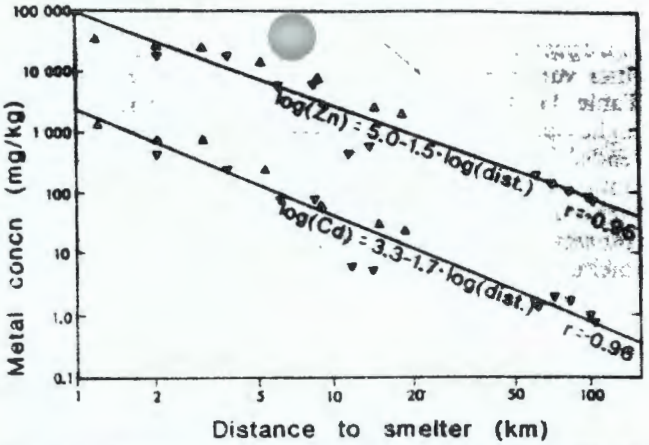

Fig. 3—Concentrations (dry wt.) of Zn (open triangles) and Cd (closed triangles) in O2 litter collected at various distances from two Zn smelters in Palmerton, Pa. Sites to the west of the smelters are represented by triangles pointing down and sites to the east by triangles pointing up.

smelter (Fig. 2, Table 1), ranging from a minimum at a site 105 km west of the smelter (67 mg/kg Zn, 0.85 mg/kg Cd, 150 mg/kg Pb, 11 mg/kg Cu) to a maximum at the site 1.2 km east of the smelters (35 000 mg/kg Zn, 1300 mg/kg Cd, 3200 mg/kg Pb, 280 mg/kg Cu). The concentration of Zn was higher at the site nearest the smelter by a factor of 500, and that of Cd by 1500. The actual degree of contamination was greater than these factors imply, since there was an accumulation of organic matter in the O2 horizon near the smelters (Strojan, 1978b). The relation between the distance to the smelters and the concentrations of Zn and Cd was evident even 100 km away (Fig. 3).

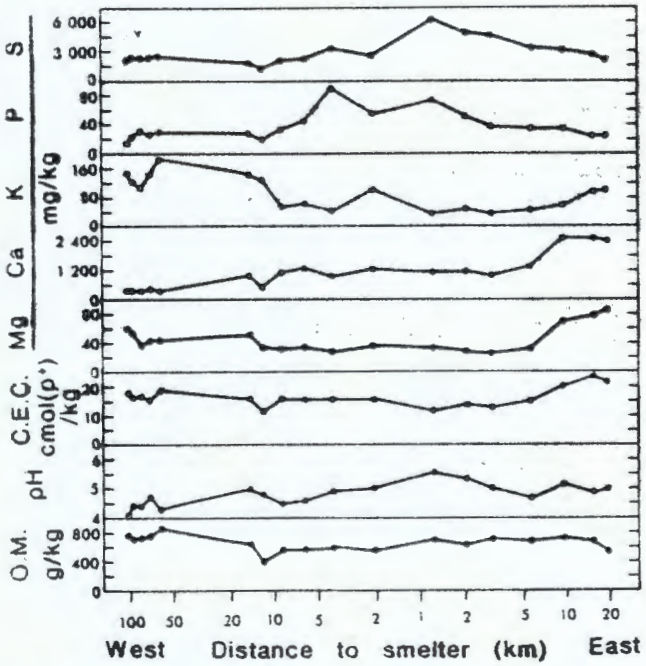

Fig. 4—Values of eight litter variables in the O2 horizons of Dekalb soils collected at various distances east or west of two Zn smelters in Palmerton, Pa. O.M. = organic matter; CEC = cation exchange capacity.

Concentrations of several other elements in the O2 litter varied with the distance to the smelters (Fig. 4, Table 1). Concentrations of S and P tended to be highest at sites nearest the smelters. Concentrations of K tended to be lowest at the sites nearest the smelters. Both the pH and the exchangeable Ca content were higher at the eastern end than at the western end of the transect; however, an increase in the pH near the smelters was not associated with an increase in the exchangeable Ca content.

Most of the litter variables examined were correlated with both the log of the distance from the smelters and the observed mortality of woodlice (Table 1), and most of the litter variables were also correlated with each other. For example, the correlation coefficients (r) for the log of the concentration of Zn vs. the log of the concentrations of the other metals were as follows: Cd, 0.99; Pb, 0.96; and Cu, 0.94 (all significant at $p < 0.05$).

The O2 litter used in the second experiment had a nutrient content similar to that of the samples collected 63 to 105 km to the west of the smelters: 16 mg/kg P, 150 mg/kg K, 300 mg/kg Ca, and 44 mg/kg Mg. The samples treated with 20 000 mg/kg Zn had the highest pH (5.3) and the samples not treated with Zn had pH values in the range of 3.7 to 4.0. The control litter contained 98 mg/kg Zn, 1.4 mg/kg Cd, 106 mg/kg Pb, and 9.4 mg/kg Cu. The metal concentrations in the treated samples, measured by atomic absorption spectrophotometry (data not shown), were similar to the concentrations calculated from the amounts of metals added experimentally (Table 2), differing by an average of < 15%.

An ANOVA showed statistically significant (F = 39.4; 7 and 40 df; $p < 0.05$) differences in the rates of survival of woodlice after 8 weeks in the experimentally amended litters. The rate of survival in the control litter was 86% (Table 2). The rate of survival of woodlice exposed to 20 000 mg/kg Zn was only 29%. There was no significant difference ($p < 0.05$) in the survival of woodlice exposed either to 20 000 mg/kg Zn or to Zn and the four other elements combined. Survival of woodlice in O2 litter treated with 5000 mg/kg Zn was intermediate between that in control O2 litter and that in O2 litter treated with 20 000 mg/kg Zn (both differences significant at $p < 0.05$). Fewer woodlice survived in O2 litter treated with Cd, Pb, or S than in the control O2 litter, but the difference was significant (p [illegible]) only in the O2 litter treated with Cd. Copper at [illegible] mg/kg did not affect mortality. The large number of woodlice used per treatment and the generally [illegible] agreement in mortality among replicates resulted in a relatively small standard error (± 3.5% of the percent survival for each treatment).

Table 2—Survival of woodlice after 8 weeks in Dekalb O2 litter amended with Zn, Pb, Cd, Cu, or S, either alone or in combination.

Treatment†	Mean % woodlice survival	% Survival compared with controls
1) Control O2 litter	86	–
2) 20 000 mg/kg Zn	29	34*
3) 5 000 mg/kg Zn	64	74*
4) 2 000 mg/kg Pb	78	91
5) 500 mg/kg Cd	69	80*
6) 200 mg/kg Cu	88	102
7) 4 000 mg/kg S	79	92
8) 20 000 mg/kg Zn + 2 000 mg/kg Pb + 500 mg/kg Cd + 200 mg/kg Cu + 4 000 mg/kg S	36	42*

* Treatment is significantly different from the control (by Dunnett's Method. $P < 0.05$).

† Each treatment consisted of six replicates, each containing 12 woodlice.

DISCUSSION

The abnormally low density of arthropods inhabiting the soil litter on Blue Mountain near the smelters (Strojan, 1978a) can be related to the high concentrations of Zn in the soil. Whereas as much as 35 000 mg/kg Zn is present in the O2 litter collected near the smelter, concentrations of both 5000 mg/kg Zn and 20 000 mg/kg Zn increased mortality of woodlice in an 8-week test. Even lower concentrations of Zn probably could be shown to be toxic to woodlice if the test period were substantially lengthened and the woodlice allowed to complete their life cycle. Concentrations of other contaminants (Cd, Pb, S) on Blue Mountain may also be toxic to woodlice, but not nearly as toxic as the concentration of Zn.

Previous estimates of the metal content of the O2 horizon near the smelters (Buchauer, 1973; Strojan, 1978b) are similar to our estimates. Nevertheless, we believe that Buchauer underestimated the extent of the contamination from the smelters (12–16 km to the west, 19–39 km to the east) because she did not sample far enough away from the smelters to accurately estimate background concentrations, which she suggests to be about 330 mg/kg Zn, 6 mg/kg Cd, 300 mg/kg Pb, and 30 mg/kg Cu. Figure 3 shows that the emissions of Zn and Cd can be detected even 100 km from the smelters. This finding is consistent with the work of Freedman and Hutchinson (1980), who showed that about 60% of the emitted Ni and Cu and about 97% of the emitted S were carried at least 60 km away from a very tall smelter stack (380 m) at a smelter in Sudbury, Canada. We attribute the excellent fit of the data points to the regression lines to the lack of other important sources of Zn or Cd emissions in the region and to the relative uniformity of the soils and of the topography of the sampling sites.

Metal concentrations detected in the litter near Palmerton are much higher than those reported in relatively uncontaminated litter. In a survey of O1 and O2 horizons from 26 oak-dominated forests in eastern Pennsylvania and in neighboring states, Andresen et al. (1980) found a mean of 139 mg/kg Zn, 174 mg/kg Pb, and 22 mg/kg Cu. These concentrations are about the same as those detected in the O2 litters from the five sites located 63 to 105 km west of the smelters. The highest Zn (1800 mg/kg) and Pb (520 mg/kg) concentrations detected by Andresen et al. were from Wind Gap, Pa., which is on Blue Mountain about 25 km east of the smelters. They attributed these high concentrations to the smelters in Palmerton. Wind Gap is just 5 km east of the easternmost site sampled in the present study, which had similar concentrations of Zn (1800 mg/kg) and Pb (400 mg/kg).

Emissions from the smelters have changed the structure and function of the soil. The O2 horizon is ab-

normally thick, is decomposing at an abnormally slow rate (Strojan, 1978b), and the soil contains reduced numbers of bacteria, actinomycetes, and fungi (Jordan & Lechevalier, 1975). Tyler (1972) predicted that heavy metal contamination could reduce the rate of decomposition of soil litter and lead to a decrease in the turnover of P, since it would be bound in the undecomposed litter. Figure 4 supports Tyler's prediction, showing an increase in the P concentrations in the O2 litter near the smelters. The decreased concentration of K may be due to its rapid turnover in litter (Tyler, 1972).

Although S emissions from smelters commonly decrease soil pH in their vicinity, the pH of the soil at Palmerton is higher than normal (Buchauer, 1973; Fig. 4). Generally Ca, Mg, and K are the main cations that determine the percent base saturation and the pH of the soil. Near the smelters, however, the increase in the pH is not associated with an increase in the concentrations of these cations. Instead, the increase is probably due to the high concentration of Zn, which may function as a basic cation (Buchauer, 1973). Most of the S from the roasters is removed by a S plant, and most of the S that is emitted from the smokestack is probably carried far away, as shown by Freedman and Hutchinson (1980).

The findings from this and previous studies (Jordan, 1975; Jordan & Lechevalier, 1975; Strojan, 1978a, 1978b) demonstrate how the Palmerton smelters are changing the surrounding environment. The populations of soil organisms, the soil profile, and the nutrient flow through the ecosystem have been altered at sites near the smelters. This study has demonstrated the value of using laboratory populations of woodlice as bioassays to help interpret the observations from a polluted environment. The results from the analyses of the O2 litter show the extent of the contamination. If the emissions have been carried as far in other directions as they have been carried to the west, then the smelter emissions have increased the metal concentrations in soil on millions of hectares of land in eastern Pennsylvania, much of which is farmland.

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