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Heavy metal concentrations in sediments of streams affected by a sanitary landfill: A comparison of metal enrichment in two size sediment fractions

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Abstract The purpose of this study was to determine if metal concentrations are enriched in two size sediment fractions of streams that receive landfill effluent and, if so, whether there is a greater extent of metal enrichment in one of the fractions. Sediment samples were collected from three streams adjacent to a sanitary landfill. Sediments representing control for the study were also collected from a stream not influenced by the landfill. All samples were sieved and the <0.0625-mm and <0.25-mm to >0.149-mm size fractions from each sample were used in this study. The concentrations of acid-extractable Cu, Zn, Pb, and Cr for all samples were determined by atomic absorption techniques. Mean concentrations, coefficient of variation values, a *t* test, and the variation of metal concentrations along the stream were used to analyze the data. Results indicated that Cu, Zn, Pb, and Cr concentrations were enriched in both size sediment fractions from the stream whose channel originated at the base of the landfill. Copper, Zn, and Pb concentrations were enriched in the <0.0625-mm size sediments of the stream whose channel did not intersect the landfill. Copper, Zn, Pb, and Cr concentrations appear enriched in both size sediment fractions of the third stream, which formed from the confluence of the other two streams. The extent of metal enrichment was greater in the <0.0625-mm size sediments. A decreasing trend of metal concentrations in a downstream direction was not present in the enriched sediments. This was true for each metal in both size sediment fractions.

Key words Metal enrichment · Acid-extractable metals · Landfill emission plume

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Introduction

Various studies have reported elevated concentrations of metals in stream sediments caused by contamination sources. There have been studies dealing with enrichment of heavy metals in river sediments influenced by industrial wastes (Ramamoorthy and Rust 1978; Castaing and others 1986; Rule 1986). These include Cu, Pb, Zn, Ni, Cd, Hg, Co, Cr, Fe and Mn. Some authors have reported Cu, Pb, Zn, Cd, Sn, As, Cr, Fe, and Mn concentrations in sediments of rivers affected by mining activities (Reece and others 1978; Wolfenden and Lewin 1978; Yim 1981; Chapman and others 1983; Mann and Lintern 1983; Moore 1985; Leenaers and others 1988; Axtmann and Luoma 1991). Studies of the enrichment of Cu, Pb, Zn, Cd, Ag, and Ba concentrations in stream sediments affected by landfills and a water treatment facility have been reported (Mantei and Coonrod 1989; Mantei and Foster 1991).

Traditionally, fine-grained sediment fractions have been used to study metal contamination in sediments. Eliminating coarse grained portions reduces biases resulting from differences among samples (Solomons and Forstner 1984; Bradley and Cox 1987). Higher concentrations of metals generally accumulate in smaller sediment grain fractions because of the higher surface area-to-grain size ratio (Whitney 1975; Ramamoorthy and Rust 1978; Harding and Brown 1978; Sinex and Helz 1981; Solomons and Forstner 1984; Horowitz and Elrick 1987; Moore and others 1989). Some authors have used restricted fine-grain size sediments in studies of heavy metal emissions from landfills to reduce the grain size bias in metal concentrations. Mantei and Coonrod (1989) used the <0.25-mm to >0.149-mm (fine sand) size. The <0.088-mm to >0.074-mm (very fine sand) size fraction was used by Mantei and Foster (1991). Some studies have used less restricted grain size fractions. Compest (1991) found that trace metal trends in sediments downstream from an emission source may not be recognized in the <2-mm sediment size fraction. Rule (1986) concludes the <0.0625-mm size sediment fraction contains the greatest concentration of

heavy metals. Axtmann and Luoma (1991) and Luoma and others (1989) used the <0.060-mm size sediment fraction to study metal trends in sediments affected by mining activities.

In this paper we report the affect of landfill leachate on the sediments of three streams located adjacent to the landfill. We determine if the <0.0625-mm and <0.25-mm to >0.149-mm grain size sediments have elevated concentrations of Cu, Zn, Pb, and Cr, and if so, whether there is evidence of a greater extent of enrichment in one size compared to the other. There is an absence in the literature of studies dealing with enrichment of heavy metals in the <0.0625-mm grain size sediments of streams influenced by landfill effluent. Landfill records indicate the aforementioned heavy metals have been periodically added to the landfill primarily in the form of industrial wastes. These metals could be emitted from the landfill into the streamwaters and enrich in the sediments through adsorption and/or precipitation. Sediments from a nearby stream unaffected by the landfill are used as a control.

Location and setting

The Wright County sanitary landfill is situated in the Ozarks physiographic province in south central Missouri, USA. The study area is located 6.5 km southeast of Hartville, Missouri. The three study streams are located west-northwest of the landfill and the control stream lies 1.5 km east of the landfill (Fig. 1). The channel of stream 1 originates at the base of the landfill. There are several leachate seeps that flow from the base of the landfill to the head of stream 1. Stream 2 originates from a small spring and is located downslope from the landfill. Consequently, this stream may receive leachate materials from the landfill through runoff and infiltration. Stream 3 forms from the confluence of streams 1 and 2. The landfill is underlain by dolomite bedrock. The channels of the three study streams and control stream are also located in this geologic rock formation. The physical nature of all four streams is similar. All streams are similar in size, associated with similar vegetation cover, and have similar peak and average water flow.

Other than landfill activity, there are no known land use practices that would affect the study streams or control stream. The landfill operated with a permit from the Missouri Department of Natural Resources (MDNR) from 1975 to 1986.

Experimental procedure

Sample collection

Forty-five, 24, and 10 sediment samples were collected from streams 1, 2, and 3, respectively. Each sample was collected at approximately 10-m intervals (Fig. 2). In addition, 30 sediment samples were collected from the control stream at the same interval. All samples were taken as near to the center of the stream as possible and from the top 6–15 cm of the sediment deposit. The samples were placed in plastic bags and returned to the laboratory for physical and chemical treatment and analysis. A similar collection procedure is reported elsewhere (Mantei and Coonrod 1989). The pH of the streamwaters was measured at every

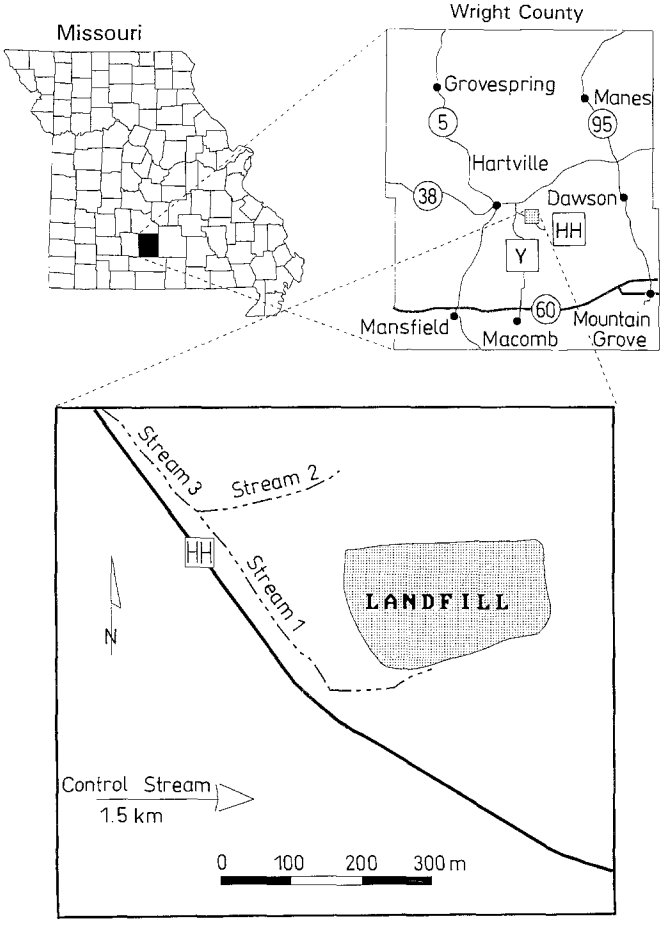


Fig. 1 Location of study area

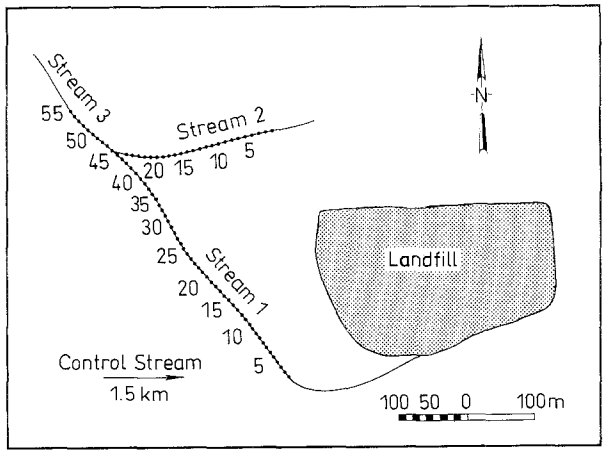


Fig. 2 Sediment sample locations

fourth sediment collection site along streams 1, 2, 3, and the control stream to determine if and to what extent it may have influenced variation of heavy metal concentrations in the sediments.

Sample preparation, chemical and data methods

All sediment samples were dried at room temperature, disaggregated, and slightly mixed with a glass stirring rod. No aggregates of particles were apparent after this activity. Each sample was passed through stainless steel sieves and the <0.0625-mm (silt and finer) and <0.25-mm to >0.149-mm (fine sand) size portions saved for chemical analyses. To establish if there was contamination from the sieving procedure, five fine-grained pure silica portions were sieved in the same manner as the sediment samples. A 1-g portion of each sample was subjected to a hot nitric acid extraction procedure (Mantei and Coonrod 1989). This procedure does not represent a total extraction and separates some chemical phases and speciated metals from the sediments.

The Cu, Zn, Pb, and Cr concentrations in each individual stream sediment and pure silica sample were determined using a Varian 1475 atomic absorption spectrophotometer. The mean and standard deviation concentrations and the coefficient of variation value for each heavy metal in each sediment sample from streams 1, 2, and 3 and the control stream were determined. The two-sample t test was used to compare the concentration of each metal in the sediments of streams 1 and 2 with the same in the control stream. The values representing mean concentration, standard deviation, coefficient of variation, and t tests were computed with the aid of a computer program called Minitab. A trend chart representing the variation of the concentrations of each metal in the sediment samples along each stream was generated using Quattro Pro.

Results and discussion

Table 1 summarizes the mean concentration, standard deviation, and coefficient of variation values for Cu, Zn, Pb, and Cr in the two size sediment fractions. The results of the t test also are included.

Comparison of mean concentrations

The mean concentration for each metal is higher in the silt and finer-size sediments of streams 1, 2, and 3 than in the control sediments. The same is true for the metals in the fine sand size sediments of streams 1 and 3. Although the concentration of Cr is higher in the silt and finer-size sediments of stream 2 than in the control sediments, the two values are similar. It is well known that pH of natural waters may affect concentrations of metals adsorbed or precipitated on stream sediments (Moore and Ramamoorthy 1984). The mean pH and range values for waters in each stream are 7.31 and 7.14–7.38 for stream l; 7.43 and 7.20–7.63 for stream 2; 7.22 and 7.11–7.36 for stream 3; 7.41 and 7.19–7.61 for the control stream. These pH values and variance of pH are similar to that of other waters in this area (Mantei and Coonrod 1989; Mantei and Foster 1991). Since the pH mean and range values of the streams are similar, the difference of metal concentrations in the stream sediments cannot be attributed to pH differences. Analyses of the fine-grained silica control samples also indicated a lack of detectable amounts of heavy metals present after sieving. Hence, the sediment samples were not contaminated by the sieving procedure.

In both size fractions, there is a higher mean concentration of each metal in the sediments of stream 1 than in the sediments of stream 2. This might be expected since stream 1 appears to be situated more in the emission plume of the landfill than does stream 2. In a similar study of two streams affected by leachate from another landfill, the fine sand size sediments of the stream whose channel originated at the base of the landfill contained higher concentrations of Cu, Zn, and Ag (Mantei and Coonrod 1989). The drainage of stream 1 into stream 2 appears to dilute the concentration of heavy metals in the sediments of stream 1 below the confluence. In both size fractions, the mean value for each heavy metal in the sediments of stream 3 falls between those values found in the sediments of streams 1 and 2. The mean concentration for each metal in the fine sand size sediments of stream 2 is similar to the same in the control sediments. The mean content of each metal in the finer-grained sediment fraction is higher than the same in the coarser-grained fraction except for Cr in the control sediments. These values are the same.

Comparison of coefficient of variation values of heavy metal populations

A difference in the variability of metal concentrations in the sediments of the study streams compared to that in the control sediments might indicate the sediments of the study streams are affected by landfill leachate. The coefficient of variation (CV) value may be used as a measure for the above comparisons. Axtmann and Luoma (1991) used CV values to show high variability of metal concentrations in bed sediments affected by metal contamination from mining activity. Table 1 shows the CV for each heavy metal population in the two size sediment fractions of each stream. The value for each metal in both size sediment fractions of stream 1 is higher than the same in the control stream. The CV values for Cu, Zn, and Pb are higher in the silt and finer size sediments of stream 2 compared with that in the control stream, while that for Cr is similar.

Statistical t test comparison

The concentration of each metal in both size sediment fractions of streams 1, 2, and the control stream represent

Table 1 Content and *t* test results of metals in stream sediments

Metal	Silt and finer				Fine sand size			
	Stream 1	Stream 2	Stream 3	Control stream	Stream 1	Stream 2	Stream 3	Control stream
Cu	139[a,b]	64.4[e]	125	52.2	11.2[b]	5.9	8.2	5.7
	144[c]	23.7	96.2	13.2	9.8	4.8	4.1	1.6
	104[d]	37	77	25	88	81	50	28
Zn	59.9[b]	34.5[f]	55.9	28.0	41.6[b]	16.7	30.8	17.9
	24.6	9.94	15.3	4.5	22.1	2.1	5.9	3.7
	41	29	24	16	53	13	19	21
Pb	66.9[b]	57.3[e]	58.1	46.0	50.1[b]	41.1	48.8	43.1
	16.1	9.1	9.9	5.2	6	6.1	2.9	3.6
	24	16	17	11	12	15	6	8
Cr	14[b]	8.6	11.4	8.5	12[b]	7.6	11	8.5
	4.6	2	1.8	1.9	3.9	2.6	2.5	1
	33	23	16	22	32	34	23	12

[a] Mean concentrations (μg/g)
[b] *t* test results showing the mean significant at 0.001 alpha level.
[c] Standard deviation (μg/g)
[d] Coefficient of variation (CV) in percentage
[e] *t* test results showing the mean significant at 0.05 alpha level
[f] *t* test results showing the mean significant at 0.01 alpha level

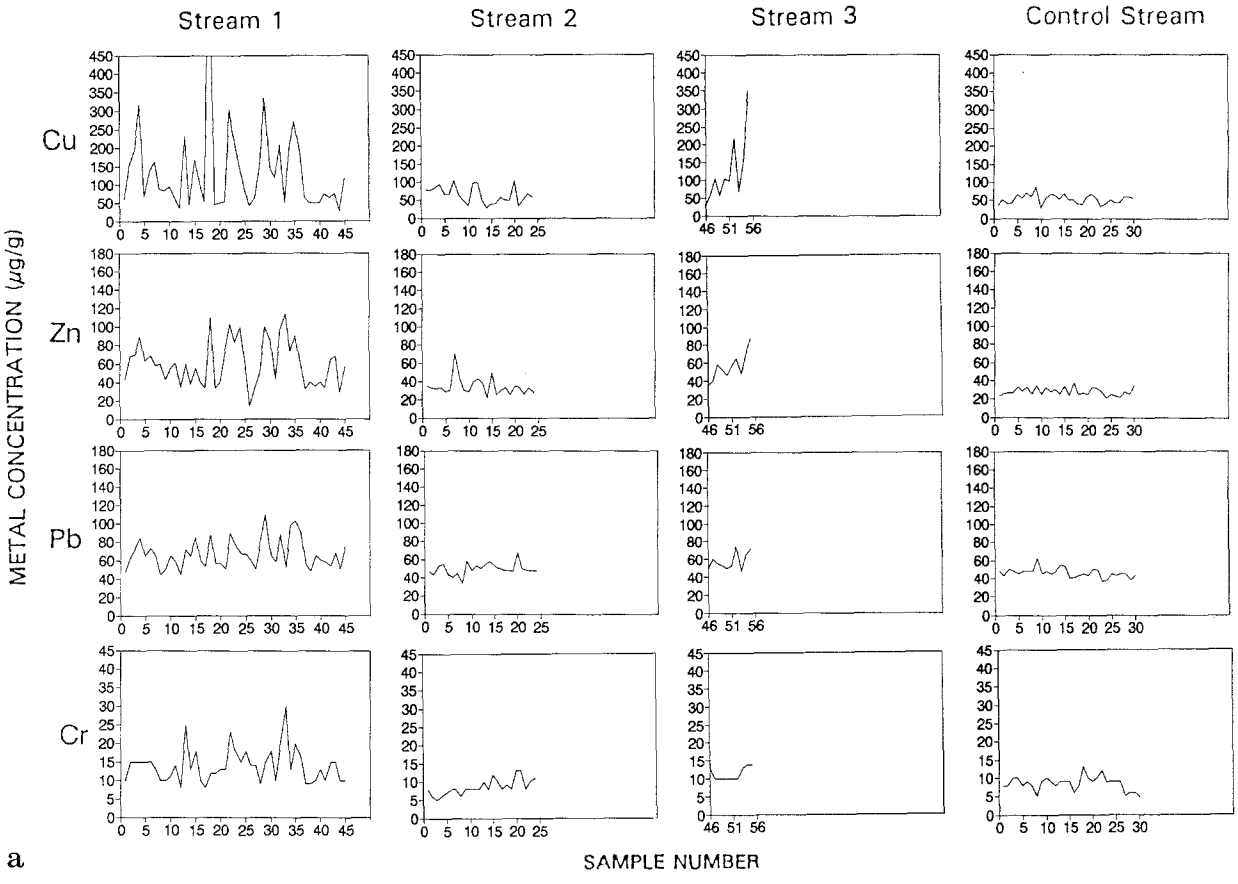
single symmetrically distributed populations. The two-sample *t* test was used to compare the concentration of each heavy metal in both size sediment fractions of streams 1 and 2 with the same in the control sediments. The specific purpose was to determine if the mean of each heavy metal representing its population in the sediments of streams 1 and 2 was significantly different from the mean of the same metal representing its population in the control stream samples. The null hypothesis used was that there was no difference between the concentration of each metal in the sediments of the control stream and that in streams 1 and 2. A failure to reject the research hypothesis for a given metal in a size fraction of the sediments of streams 1 and 2 would indicate that the metal concentrations are not different than the background, assuming the control sediments represent the background. A rejection of the research hypothesis would indicate that the metal concentrations are different than the background. The *t* test was performed at the 0.05, 0.01, and 0.001 alpha levels for all metals. Table 1 shows the *t* test results. There was a rejection of the hypothesis for Cu, Zn, Pb, and Cr in both size sediment fractions of stream 1 and for Cu, Zn, and Pb in the silt and finer size sediments of stream 2 at the 0.05 and greater confidence levels. There was a failure to reject the research hypothesis for Cr in the silt and finer-size sediments of stream 2 and for all metals in the fine sand size sediments of stream 2 based on the same confidence levels.

The results from the *t* test indicate that concentrations of Cu, Zn, Pb, and Cr in both size sediment fractions of stream 1 and Cu, Zn, and Pb in the silt and finer-size sediments of stream 2 are enriched with respect to the control sediments. The same result is observed from the analysis of the mean concentrations and CV values. This enrichment is probably caused by leachate from the landfill. Chromium concentrations from the landfill leachate drainage may have diminished through adsorption or precipitation on soils and rocks before reaching stream 2, resulting in the lack of enrichment in the silt and finer-size sediments. A definitive reason for the lack of enrichment of all metals in the fine sand size sediments of stream 2 cannot be given. Possibly the diminished metal concentrations in the leachate were too small to register an enrichment in this sediment size. However, Mantei and Coonrod (1989) report an enrichment of Zn and Ag in the fine sand size sediments of a stream that also originated from a spring and received leachate material from a landfill through runoff and infiltration.

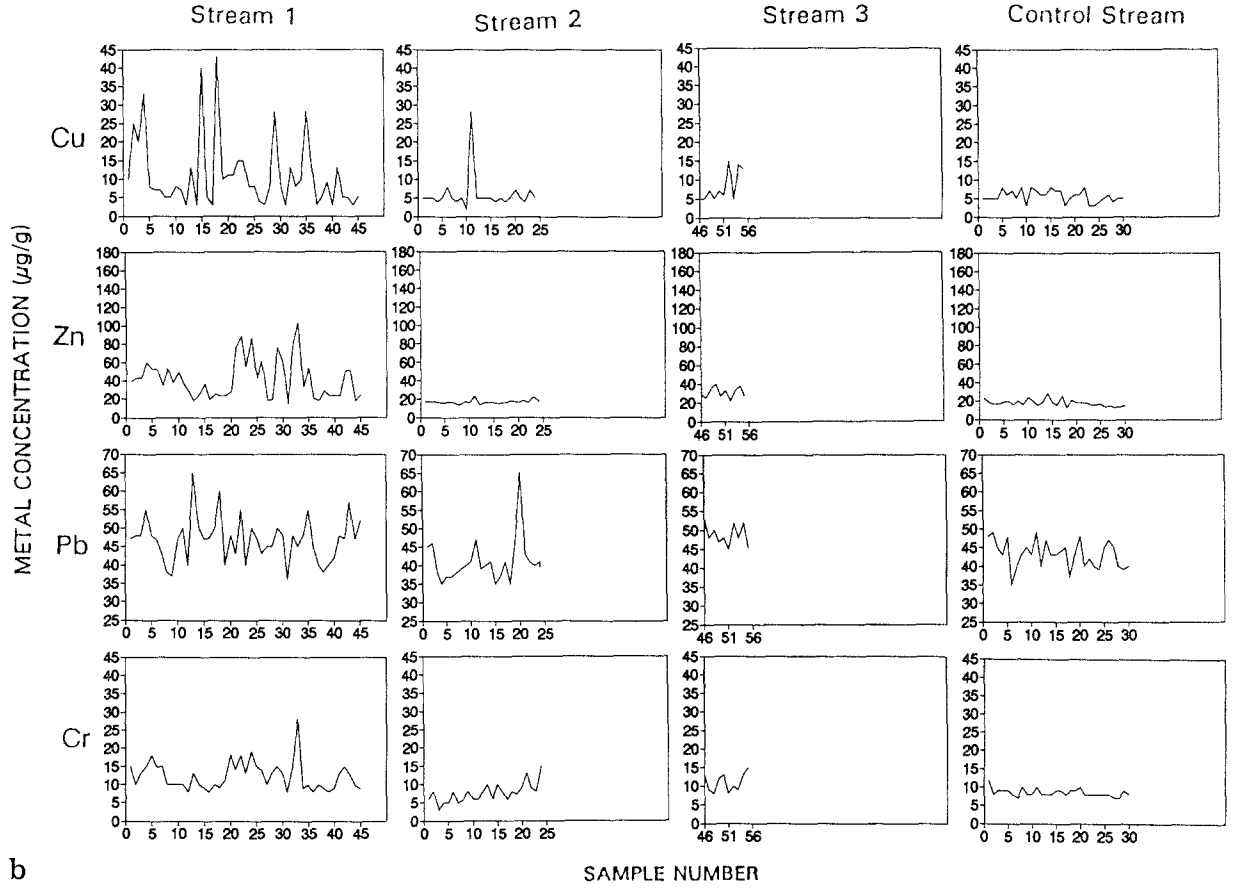
Comparison of heavy metal concentration trends

Various studies report decreasing metal concentrations in sediments in a downstream direction from a contamination source. Axtmann and Luoma (1991) report this trend for Cu, Zn, Pb, Cd, and Ag concentrations in the <0.060-mm size sediments affected by mining activity over a distance of 380 km. There are other studies that show this trend for metals over smaller distances (Wolfenden and Lewin 1978; Chapman and others 1983; Mann and Lintern 1983; Rybicka and Kyzoil 1987; Leenaers and others 1988). Figure 3A and B show the trends of metal concentrations in both size sediment fractions along each stream. There appears to be a lack of decreasing concentration for each metal downstream in the enriched sediments. This is true for both size sediment fractions. Such a trend may be undetectable over small distances, as used in this study. Mantei and Coonrod (1989) also observed a lack of such a trend for elevated concentrations of Cu, Zn, and Ag in fine sand size sediments from a stream affected by landfill emission. Their study included samples collected over a distance of 1.5 km beginning at the junction of the landfill and stream. Land ownership restrictions did not permit access for collection of sediments from stream 1 immediately below the landfill. The first sample was collected in excess of 200 m from the junction of the landfill and stream

Fig. 3a The trends of the heavy metal concentrations in the silt and finer size sediments along the streams. **b** The trends of the heavy metal concentrations in the fine sand size sediments along the streams


Stream 1
Stream 2
Stream 3
Control Stream
Cu
Zn
Pb
Cr
METAL CONCENTRATION (μg/g)
SAMPLE NUMBER

a


Stream 1
Stream 2
Stream 3
Control Stream
Cu
Zn
Pb
Cr
METAL CONCENTRATION (μg/g)
SAMPLE NUMBER

b

(Fig. 2). Information concerning metal concentrations in the sediments in this area may have been important to this study.

Summary and conclusions

The analyses of the data indicates that concentrations of Cu, Zn, Pb, and Cr were emitted by the Wright County Sanitary Landfill. The enrichment of these metals in both size sediment fractions of stream 1 clearly indicates this. The lack of metal enrichment in the fine sand size fraction of stream 2 indicates that the extent of metal enrichment caused by the landfill leachate was greater in the silt and finer-size sediment fraction. There was a lack of decreasing metal concentrations present in the metal-enriched sediments in a downstream direction from the landfill. Such a trend for a metal may be undetectable over small distances, such as that used in this study.

Currently there is a lack of definite regulatory standards established for heavy metal toxicity in stream sediments. Metal concentrations in stream sediments thought to be affected by a landfill may be compared with the same in stream sediments known to be affected by a landfill on the Superfund cleanup list. Studies such as this, which generate information on metal concentration in stream sediments affected by landfills, can aid in this comparison. Furthermore, studies such as this, which define elevated heavy metal concentrations in sediments, can aid biomonitoring studies in a natural setting. Toxicity studies dealing with assimilation of heavy metals in life forms such as algae, snails, and ostracods have been directed to areas of elevated metal concentrations in stream sediments affected by landfill leachate (Koontz 1992; Havel and Talbott 1994).

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