Heavy Metal Content in the Stream Sediments Adjacent to a Sanitary Landfill

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ABSTRACT / Samples of stream sediments were collected along two streams adjacent to a sanitary landfill. One of the streams drained the landfill directly. In addition, control of background samples were also analyzed from a stream not affected by the landfill. All samples were analyzed for Ag, Zn, Cu, Cd, and Pb content using atomic adsorption techniques. The pH of the three streams were monitored since pH differences in the streams may affect the quantities of metals adsorbed or precipitated on the sediments. The comparison between the content of Ag, Zn, and Cu in the sediments of the two study streams and the same metals in the control sediments indicated the landfill emitted these metals into the two adjacent streams. However, since the Cd and Pb contents in the sediments of both streams were similar to that of the control stream sediments, these metals may not be emitted into the two study streams from the landfill and they represent only background quantities. The comparisons of each metal in the sediments of each stream were made by the use of a metal trend chart, the individual calculated mean metal content values, and by the statistical two sample t-test. No decreasing trends of the quantities of Ag, Zn, or Cu as a function of increasing distance from the landfill was present in the sediments along the stream that drained the landfill directly. These sediments might have been affected by stream action and became mixed with other sediments downstream.

Further related studies comparing the metal content in the stream sediments of an older landfill to a younger or a more active landfill to a less active one might show interesting results.

Introduction

There have been various studies reported in the literature concerning heavy metals that have been adsorbed or precipitated on stream sediments. Castaing and others (1986), Ramamoorthy and Rust (1978), and Rule (1986) studied the enrichment of heavy metals in river sediments influenced by industrial wastes. Reece and others (1978), and Yim (1981) have reported the quantities of heavy metals in the sediments of rivers affected by mining activities.

However, there appears to be an absence in the literature of studies on the enrichment of heavy metals in the sediments of streams directly influenced by a sanitary landfill. The authors have made a study of the content of some heavy metals in the sediments of two intermittent streams adjacent to a sanitary landfill. This landfill was constructed 13 years ago and has recently closed operations.

Location and Geology

The Webster County sanitary landfill is located in southwest Missouri approximately 4 km south of the city of Marshfield, Missouri (Fig. 1). The physiographic setting of the study area is the Ozark Plateau, situated near the boundary between the Springfield and the Salem Plateaus.

The climate in the region is temperate and humid. The mean annual temperature is 13° C and the mean annual precipitation is 86.1 cm.

The geology of the area includes Mississippian age formations (Fig. 2). The Burlington, Elsey, and Pierson formations are all limestones in the Osagean series. The Compton limestone and the Northview shale-silt formations belong to the Kinderhookian series. All the formations dip slightly to the west (approximately 30 ft/mi).

The sediments in this study were taken from two streams located to the west of the landfill. Both streams drain the aforementioned Mississippian rocks. One stream drains the landfill directly and junctions the second stream. The waters continue at this confluence and discharge into the James River. This location is approximately 35 km upstream from a water purification facility that provides the waters of this river as a portion of the municipal water for the city of Springfield, Missouri.

Methods

Records indicated that various materials were added to the landfill including large quantities of pho-

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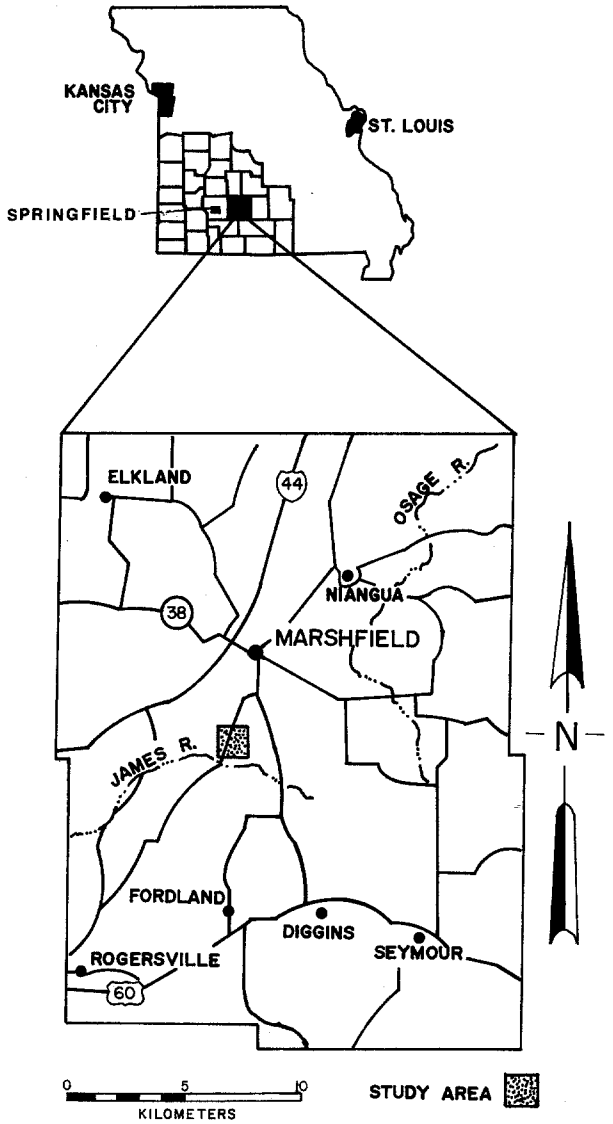

Figure 1. The location of the Webster County, Missouri sanitary landfill.

tographic, battery and electronic wastes. The metals Ag, Zn, Cu, Cd, and Pb would be found in such items and could be emitted from the landfill into the stream waters and concentrate on the sediments through adsorption or precipitation.

Sample Collection

One hundred and thirteen sediment samples were collected from the two study streams. Stream 1 was divided into an upstream and downstream portion because the drainage of stream 2 into stream 1 could affect the results of the quantities of metals in the sediments in stream 1 below this junction. The sampling sites are shown in Figure 3. An additional 30 sediment samples were collected from the control or background stream that is not affected by the landfill. This control stream is located 1.4 km east (up dip) of the landfill. It flows southward, and like the two study streams, drains the same Mississippian rocks.

The 143 samples were collected at 16 m intervals along each stream and at a depth of 10 cm in the sediments. It was hoped this depth would represent a zone relatively undisturbed by stream action where metals could concentrate on the sediments. From the analysis this could indicate a decreasing trend of the content of one or more of the metals in the sediments of stream 1 as a function of increasing distance from the landfill. If such a trend exists, it would appear that the sediments in stream 1 would show this because it drains the landfill directly.

All the sediment samples collected were placed in plastic bags for laboratory processing.

Choice of Sediment Grain Size

The samples were dried at room temperature, disaggregated, and passed through stainless steel sieves. The sediment fraction, <0.25 mm to >0.149 mm (fine sand size) was saved in plastic bags for further analysis.

Much of the research on heavy metals adsorbed or precipitated on sediments has been performed using a fine grained fraction of the sediment (Gibbs 1973; Griffen and others 1976; Harding and Brown 1978; Ramamoorthy and Rust 1978; Rule 1986; Sinex and Helz 1981). It is well known that higher quantities of metals accumulate on the smaller grain fractions because of the higher surface area to grain size ratio. It was believed the use of a coarser grain size may better document a trend of increasing quantities of one or more of the metals in the sediments of stream 1 with respect to their distance from the landfill because of a limited transport and longer residence at a site.

Chemical Extraction of Metals

Various chemical extraction methods for heavy metals accumulated on sediments using HNO_3 or HNO_3/HCl have been reported (Bloom and Ayling 1977; Rule 1986; Trefry and Presley 1976). Different extraction times were used with HNO_3 and HNO_3/HCl on 10 sediment samples to find the best combination of extraction times and acid(s) to use in this study. This included boiling samples for 5 min, 20 min, or gently heating them or letting them stand at room temperature overnight. None of these methods showed a more complete extraction of any of the metals used in this study.

A 2-g portion of each sediment sample was boiled for 5 min in 5 mL of 6*M* HNO_3, filtered, and diluted with double deionized water to a final acid concentra-

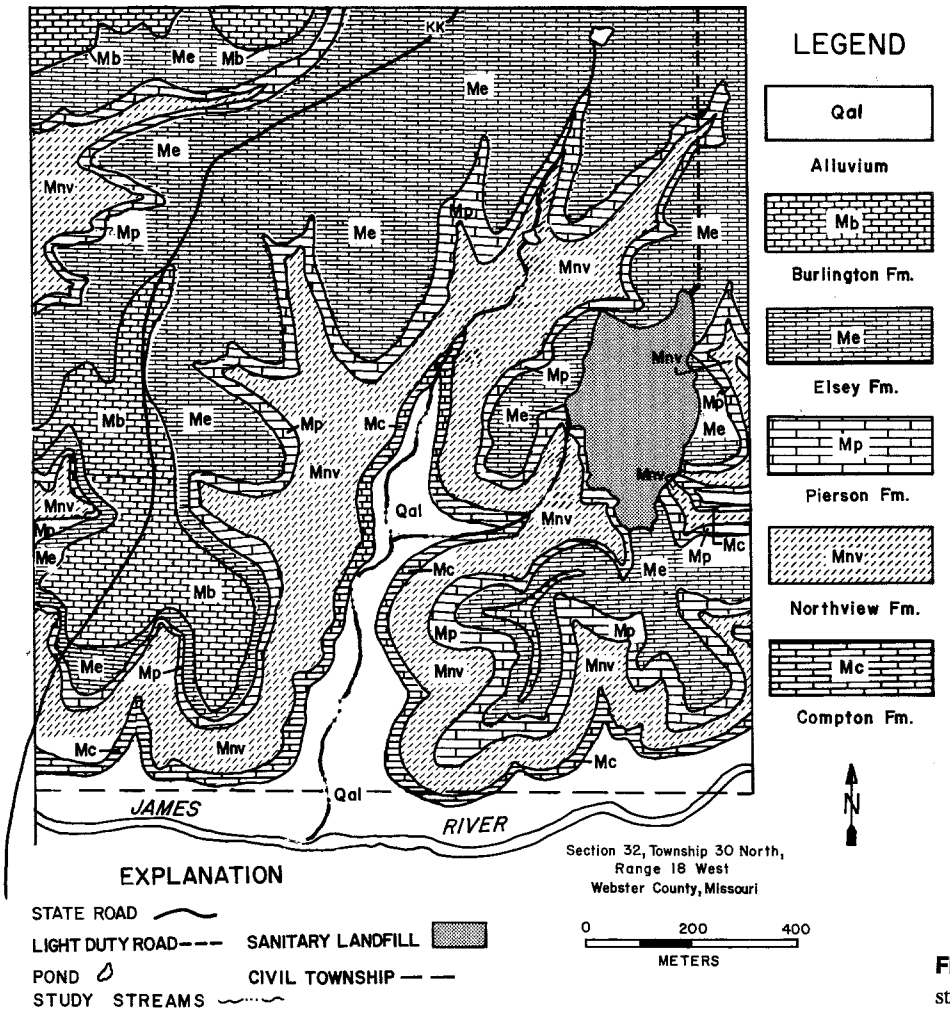

Figure 2. The geology of the study area.

tion of 10 percent. The content of the metals Ag, Zn, Cu, Cd, and Pb were determined for each sample using the Varian 1475 atomic absorption spectrophotometer. A duplicate analysis for each metal on 10 samples was performed to obtain a precision for the analysis.

Results and Discussion

Metal Content and General Trends

The content of the metals at each collection site in the stream sediments of streams 1 and 2 are given in Table 1, and those for the control stream sediments are shown in Table 2. Figure 4 shows the variation and the trends of the Ag, Zn, Cu, Cd, and Pb content in the sediments of streams 1 and 2, and the control stream. The mean content of Ag, Zn, and Cu in the samples appear to vary from stream to stream, while that of the Cd and Pb appear to be about the same in the sediments of each stream. Table 3 summarizes the mean content of each metal in the sediments of the streams. A ±value is shown next to each metal value and represents the analytical precision based on duplicate sample analyses using the relative error (relative range) method.

Specific Metal Content, Trends, and Comparisons

The content of Ag and Zn is higher in the sediments of the upper and the lower portions of stream 1, and in stream 2, than in the control sediments. Copper has a higher content in the sediments of stream 1 than in the control sediments.

If the control stream sediments act as a background in this study, it would appear that the drainage from the landfill is a source for the enrichment of the Ag, Zn, and Cu quantities in the sediments of stream 1, and the Ag and Zn quantities in the sediments of stream 2. Stream 1 drains the landfill directly and has a higher content of Ag, Zn, and Cu metals in its sediments than stream 2. This might be expected.

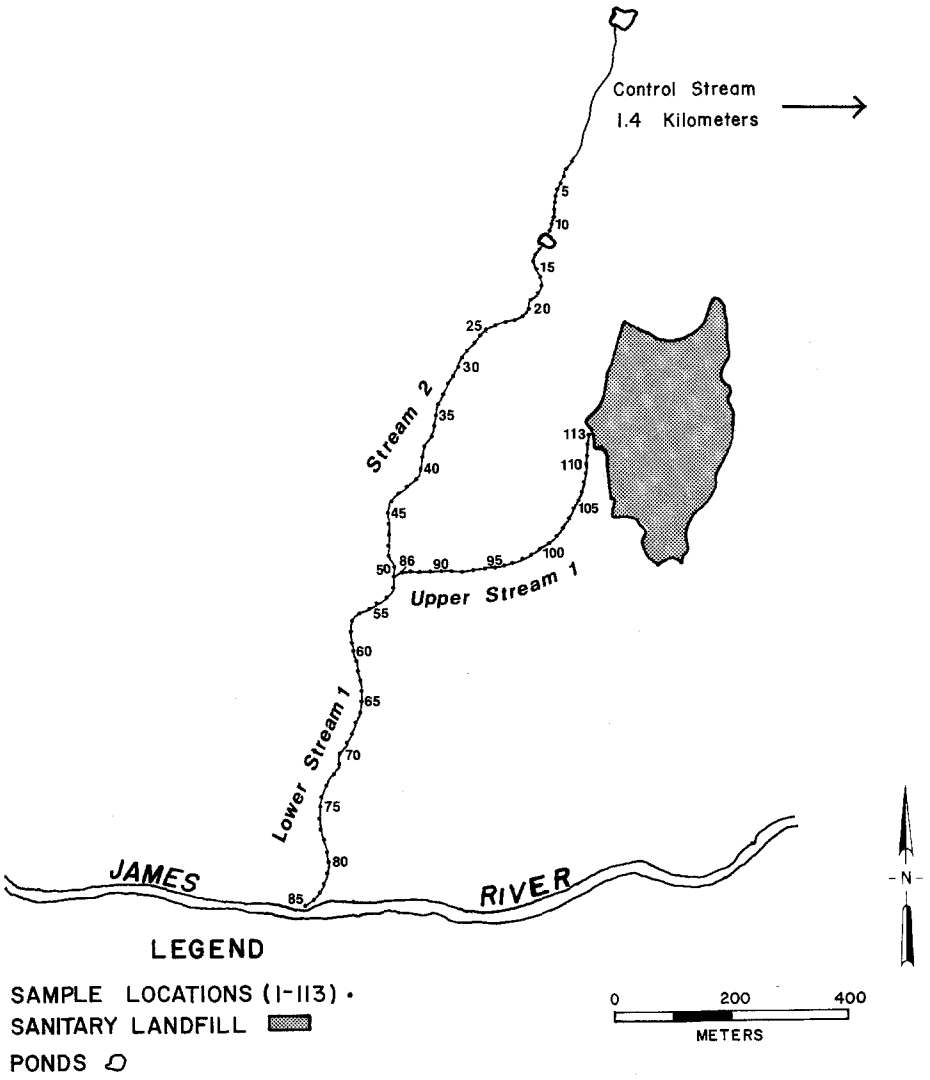

Figure 3. The sample location sites in the stream sediments of the streams adjacent to the sanitary landfill.

The geology of the area may explain why the sediments in stream 2 have been enriched in the metals emitted from the landfill. It was observed that landfill leachate material penetrates the Elsey and Pierson formations to the impermeable Northview silt-shale. Since the formations dip slightly to the west, this leachate containing the metals then could enter stream 2 by way of leachate springs (Fig. 2). It is interesting to note that copper, a relatively mobile metal, is not enriched in the sediments of stream 2 above the background quantity for Cu (Table 3 or Fig. 4).

The drainage of stream 2 into stream 1 does not appear to dilute the contents of the Ag, Zn, and Cu in the sediments of stream 1 below the junction. The content of these metals in the sediments in the upstream and downstream portions of stream 1 are essentially the same. The higher content of these metals in stream 1 as compared to stream 2 becomes apparent at or near the junction of these two streams where a jump in the metal content exists (Fig. 4).

A trend of increasing quantities of Ag, Zn, or Cu in the sediments along stream 1 towards the landfill does not exist. The coarser grain size sediment used in this study at the depth of collection might be transported by stream action and, therefore, does not have sufficient residence time at a given site to establish such a trend.

The Cd and Pb content in the stream sediments of streams 1 and 2 do not vary much from the content in the control stream sediments. It would appear that the landfill may not be emitting these metals into the streams and the content of these metals in the sediments of streams 1 and 2 represent only background quantities. Moore and Ramamoorthy (1984) mention that virtually all of the quantities of Cd and Pb metals would be adsorbed or precipitated on river sediments

Table 1. Heavy metal content (μg/g) in the stream sediment locations adjacent to the landfill.

Sample	Ag	Zn	Cu	Cd	Pb	Sample	Ag	Zn	Cu	Cd	Pb
1	0.8	20.0	7.8	0.40	36.0	57	2.0	34.0	15.6	0.40	32.0
2	0.7	10.4	7.4	0.32	42.0	58	1.4	20.0	8.4	0.38	26.0
3	0.6	27.0	7.8	0.44	30.0	59	2.2	37.4	18.0	0.40	26.0
4	1.4	22.0	5.8	0.34	24.0	60	0.6	33.4	5.6	0.40	32.0
5	0.8	19.0	6.0	0.38	26.0	61	1.6	34.6	10.4	0.40	44.0
6	0.6	6.0	3.6	0.42	34.0	62	2.0	37.2	10.6	0.41	34.0
7	0.6	22.0	8.2	0.40	34.0	63	1.6	34.6	7.8	0.44	38.0
8	0.6	8.6	4.0	0.24	26.0	64	1.8	35.2	7.6	0.56	44.0
9	0.6	10.8	5.6	0.25	40.0	65	1.6	37.8	9.6	0.50	48.0
10	0.6	8.0	5.6	0.30	30.0	66	1.4	28.2	6.4	0.40	40.0
11	0.8	15.0	4.4	0.38	26.0	67	1.8	32.0	10.4	0.42	34.0
12	0.6	12.2	3.6	0.32	34.0	68	1.4	28.6	6.6	0.28	40.0
13	0.4	9.6	2.6	0.36	40.0	69	1.6	28.2	6.4	0.36	34.0
14	0.8	10.0	4.0	0.36	40.0	70	1.4	28.2	7.6	0.42	34.0
15	0.7	10.0	4.4	0.37	30.0	71	1.8	37.4	12.0	0.44	33.0
16	0.8	12.0	6.4	0.34	30.0	72	1.5	28.0	8.6	0.38	34.0
17	0.7	12.0	4.4	0.40	34.0	73	1.4	32.0	7.6	0.40	40.0
18	1.4	17.6	5.4	0.32	36.0	74	1.7	32.2	7.8	0.48	42.0
19	0.8	16.0	4.2	0.32	38.0	75	1.8	32.6	9.6	0.42	42.0
20	0.4	18.2	4.0	0.30	36.0	76	1.7	33.6	9.2	0.36	40.0
21	0.6	18.0	3.8	0.34	32.0	77	1.7	33.0	9.2	0.44	46.0
22	0.6	22.2	4.6	0.44	34.0	78	1.7	37.0	14.0	0.40	36.0
23	0.7	22.2	4.6	0.24	32.0	79	0.6	22.2	8.2	0.38	32.0
24	0.9	19.6	4.6	0.26	30.0	80	2.0	38.6	20.0	0.60	46.0
25	0.9	19.2	4.4	0.48	44.0	81	1.8	37.6	16.0	0.38	34.0
26	0.6	15.6	4.8	0.36	26.0	82	0.7	30.0	7.6	0.20	34.0
27	0.59	20.4	4.4	0.36	28.0	83	0.6	51.0	12.0	0.48	38.0
28	0.6	21.0	5.8	0.34	28.0	84	1.7	35.6	12.0	0.40	34.0
29	0.58	17.4	5.6	0.36	34.0	85	0.6	35.6	11.6	0.38	32.0
30	0.65	11.4	4.0	0.24	26.0	86	1.8	37.4	8.2	0.40	34.0
31	0.6	9.6	4.0	0.36	30.0	87	1.6	28.6	6.0	0.42	26.0
32	0.62	16.4	4.0	0.34	32.0	88	1.8	36.0	7.8	0.32	26.0
33	0.6	20.0	7.6	0.36	36.0	89	1.4	31.8	7.8	0.30	34.0
34	0.8	22.2	4.8	0.38	30.0	90	2.0	33.6	13.0	0.30	26.0
35	0.55	18.4	4.6	0.44	42.0	91	1.9	33.0	7.0	0.42	30.0
36	0.6	20.4	6.6	0.43	26.0	92	1.7	30.6	7.6	0.44	32.0
37	0.57	22.2	7.4	0.32	32.0	93	1.8	37.8	14.6	0.42	36.0
38	0.6	11.6	4.2	0.40	36.0	94	2.0	37.6	15.4	0.40	32.0
39	0.8	20.0	4.6	0.34	38.0	95	2.4	37.2	20.0	0.40	34.0
40	0.5	14.2	4.0	0.24	28.0	96	3.0	37.4	15.0	0.38	36.0
41	0.6	28.2	4.0	0.38	44.0	97	2.0	37.2	8.2	0.38	32.0
42	0.7	33.0	4.6	0.40	24.0	98	1.7	35.6	9.8	0.34	36.0
43	0.6	22.0	2.2	0.44	40.0	99	1.4	30.6	13.0	0.38	40.0
44	0.6	13.0	1.8	0.40	20.0	100	0.6	23.6	9.2	0.24	24.0
45	0.6	22.2	3.6	0.48	34.0	101	0.6	11.0	7.6	0.28	26.0
46	0.9	27.6	2.2	0.44	36.0	102	2.4	37.4	13.8	0.44	42.0
47	0.6	20.4	2.2	0.40	40.0	103	1.6	37.2	16.2	0.40	40.0
48	1.4	37.0	4.2	0.56	40.0	104	2.0	37.2	14.6	0.40	30.0
49	1.5	30.6	3.6	0.40	36.0	105	2.4	33.8	9.8	0.48	38.0
50	1.8	37.6	6.4	0.42	42.0	106	0.8	36.4	10.0	0.38	10.0
51	1.4	23.6	16.0	0.40	26.0	107	1.4	20.4	7.8	0.30	34.0
52	2.0	38.0	8.6	0.46	34.0	108	0.8	15.6	9.8	0.36	52.0
53	2.4	38.2	12.8	0.44	38.0	109	3.0	22.8	8.2	0.30	42.0
54	1.6	23.6	8.6	0.42	34.0	110	0.9	22.2	6.8	0.32	34.0
55	1.4	31.0	17.6	0.46	34.0	111	0.8	35.8	8.6	0.38	30.0
56	1.8	35.0	17.4	0.44	56.0	112	2.2	30.0	11.6	0.38	32.0
						113	2.0	23.8	8.6	0.36	32.0

Table 2. Heavy metal content (μg/g) in the sediments of the control stream.

Sample	Ag	Zn	Cu	Cd	Pb
C1	0.43	13.4	5.6	0.29	48
C2	0.21	12.8	5.4	0.26	35
C3	0.40	10.3	6.0	0.39	39
C4	0.28	12.2	6.4	0.40	23
C5	0.26	10.7	4.1	0.33	40
C6	0.48	11.4	7.1	0.47	51
C7	0.40	13.5	5.2	0.30	31
C8	0.28	15.2	5.2	0.42	42
C9	0.43	11.8	5.1	0.50	41
C10	0.42	10.6	6.3	0.48	32
C11	0.37	11.0	6.5	0.30	39
C12	0.43	14.2	5.3	0.35	30
C13	0.43	14.5	5.8	0.39	41
C14	0.28	12.8	6.0	0.43	40
C15	0.36	10.6	5.6	0.39	29
C16	0.41	13.3	5.5	9.34	33
C17	0.29	12.9	7.3	0.38	31
C18	0.49	13.2	5.7	0.43	53
C19	0.33	12.5	5.8	0.47	42
C20	0.26	10.8	5.9	0.39	28
C21	0.48	11.3	7.1	0.45	44
C22	0.40	13.6	7.0	0.51	32
C23	0.28	15.4	6.5	0.48	38
C24	0.43	12.1	6.0	0.40	39
C25	0.43	9.8	7.5	0.38	31
C26	0.37	10.9	5.2	0.46	43
C27	0.43	14.2	5.8	0.41	50
C28	0.42	16.1	6.1	0.28	28
C29	0.28	11.0	7.8	0.31	35
C30	0.36	10.1	5.6	0.40	39

at a pH $\geq$ 7 (pH > 6 for Cd and pH $\geq$ 7 for Pb). The authors monitored the pH along streams 1 and 2 during different wet and dry periods. The lowest pH was 6.5 at the junction of stream 1 and the landfill, but increased to a pH > 7 within 16 m along stream 1. Cd and Pb may be leached within the landfill. However, these metals could be largely adsorbed or precipitated before leaving the landfill.

The mean content of Ag, Zn, and Cu in the sediments of streams 1, 2, and the control stream could be explained largely as a function of the differences in the mean pH of the individual streams. It is well known that the pH of a stream may effect the quantities of metals adsorbed or precipitated on the stream sediments. However, no large mean pH differences between the streams were observed. Each of the streams were monitored four times and each tested on the same day. These times represented different weather conditions. The mean pH of each stream was 7.2 for the upper portion of stream 1, 7.4 for the lower portion of stream 1, 7.4 for stream 2 and 7.3 for the control stream. These are typical pH values for streams in this area.

The content of Zn and Cu found in the sediments of streams 1 and 2 are within the range reported by Moore and Ramamoorthy (1984) for river sediments polluted by industrial or municipal sources (Cu, 1–82 μg/g; Zn, 6–339 μg/g). However, the size fraction of sediment analyzed was not cited. No such data for Ag was found in the literature to compare with the Ag content in this study.

Statistical T-Test Comparisons

The 2-sample t-test was used to compare the content of each metal in the sediments of streams 1 and 2 with the same in the control sediments. The specific purpose was to determine if the mean of each metal representing its population in the sediments of each of the streams 1 and 2, was substantially different from the mean of the same metal representing its population in the control stream samples. The research hypothesis used was that there was no difference between the content of each metal in the sediments of the control stream and the sample streams. A failure to reject the research hypothesis for a given metal in the samples of stream 1 or 2 would indicate that the metal quantity represents the background. A rejection of the research hypothesis would indicate that drainage from the landfill affected the content of that metal in the sediments of those two streams. A statistical computer program, minitab, was used to analyze the data from Tables 1 and 2. The t-test was performed at the 0.05 alpha level.

There was a rejection of the research hypothesis for the Ag, Zn, and Cu in the sediments of stream 1, and also for Ag and Zn in the sediments of stream 2. There was a failure to reject the research hypothesis for the Cu in the samples of stream 2, and for the Cd and Pb in the samples of streams 1 and 2.

The results from the t-test would then indicate the drainage from the landfill enriched the content of Ag, Zn, and Cu in the sediments of stream 1, and the content of Ag and Zn in the sediments of stream 2. The Cu content in the sediments of stream 2 and the Cd and Pb content in the sediments of both streams 1 and 2 represent the background. This agrees with those results discussed earlier.

Conclusion

The purpose of this research was to study the content and variation of some heavy metals in the sediments of streams affected by a sanitary landfill. It appears that certain metals are enriched in these sediments. The sediments in the stream draining the landfill do not show a decreasing trend of metal quan-

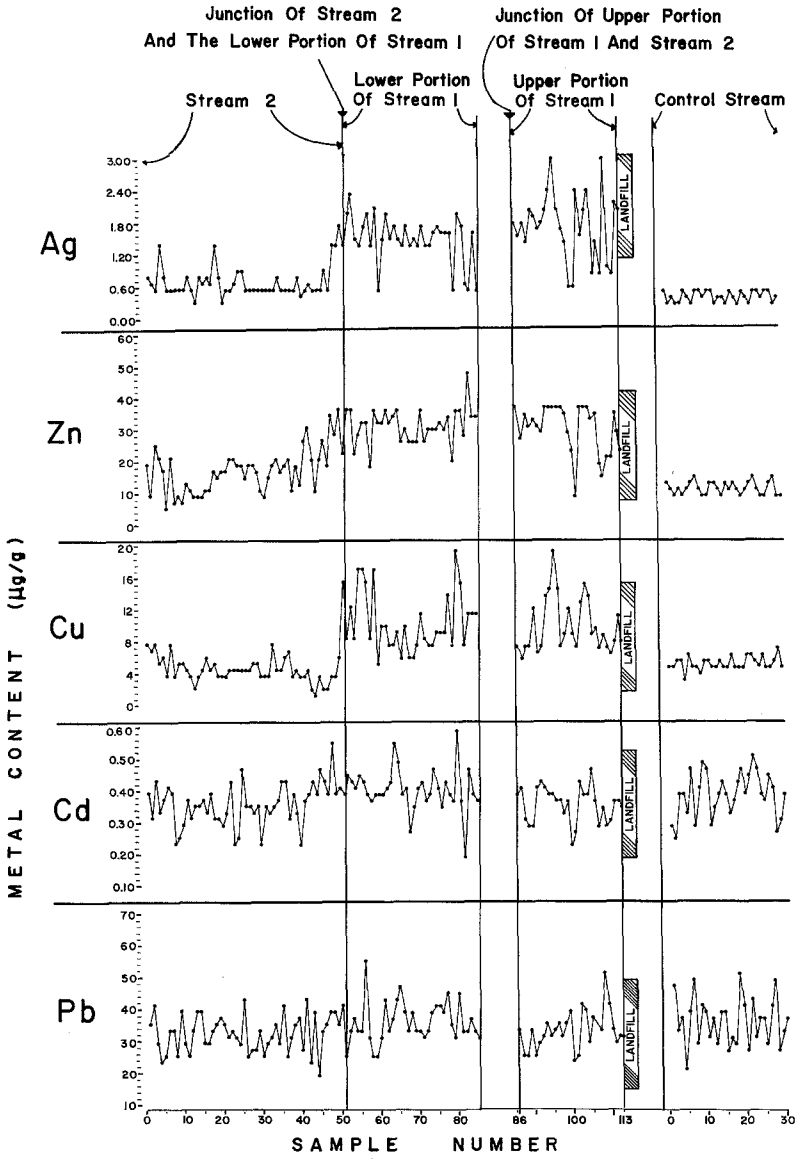

Figure 4. The trends of the Ag, Zn, Cu, Cd, and Pb content in the stream sediments of streams 2, 1, and the control stream.

Table 3. Mean content (µg/g) of the metals in the stream sediments.

Metal	Content in stream 2	Content in lower portion of stream 1	Content in upper portion of stream 1	Content in control stream
Ag	0.75 ± 0.18[a]	1.57 ± 0.38	1.71 ± 0.41	0.37 ± 0.09
Zn	18.40 ± 1.20	33.01 ± 2.14	31.13 ± 2.02	12.41 ± 0.81
Cu	4.77 ± 0.10	10.80 ± 0.24	10.61 ± 0.23	5.83 ± 0.13
Cd	0.37 ± 0.03	0.41 ± 0.04	0.37 ± 0.03	0.39 ± 0.04
Pb	33.32 ± 1.93	36.90 ± 2.14	32.88 ± 1.91	37.60 ± 2.18

[a]Relative error value referring to the precision of the analytical method

tities with increasing distance from the landfill. A possible answer for this is that the sediments chosen for analysis might have been affected by stream action and became mixed with other sediments downstream. Sediments located at a greater depth in the stream bed may not be affected by stream transport. Further studies of the heavy metal content in the sediments of streams draining an older landfill compared to that of a younger landfill, or comparing the same heavy metal content in the sediments of a more active landfill to

the same in a less active landfill might prove interesting.

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