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James R. Morrissey for the Master of Science

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Title: Metal Pollution Associated with a Landfill: Concentrations in Water, Sediment, Crayfish and Fish

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Water, sediment, crayfish (Orconectes nais) and orangespotted sunfish (Lepomis humilis) were analyzed for Pb, Cd, Al, Zn, and Cu in samples taken from four sites in the vicinity of a Lyon County, Kansas landfill. Site one was immediately above the landfill on a tributary of the Cottonwood River and site two was immediately below the landfill on the same tributary. Sites three and four were above and below, respectively, the confluence of the tributary and river.

There is no evidence that the landfill is a source of metal contamination for water, crayfish or fish. Concentrations of Pb, Cd, Al, Zn and Cu in sediment were, however, significantly higher below the landfill than above. Thus the landfill appears to be a major source of these metals for the tributary below. Al levels, however, may be due to naturally occuring Al minerals in the clay fraction of the sediment. The general patterns for

concentrations of metals in the various components were Pb: water, fish, crayfish < sediment; Cd, Al, Zn: water < fish < crayfish < sediment; Cu: water < fish < sediment < crayfish.

Al was the only metal for which the concentration in water was significantly different among sites; it was higher at site two. Pb, Cd and Al were significantly higher in sediment at site two. Zn was higher in sediment at site two than at site one, but was not significantly different from sites three and four. Cu in sediment was different among all sites, as follows: site three > site two > site four > site one. There were no significant differences among sites for any metal levels in crayfish. Al was significantly lower in fish at site two, whereas Cu was significnatly higher in fish at site one.

Comparing crayfish to fish showed crayfish had significantly higher mean concentrations of Cd, Al, Zn and Cu. These differences could possibly be explained by the close association of crayfish with the sediment. Cu, however, is a necessary component of hemocyanin in crayfish, and this could account for the higher levels.

Although there were no significant differences among sites for Pb, Cd, Zn and Cu in water, some sites had levels higher than Kansas Department of Health and Environment recommended maximum safe levels for water within the state.

METAL POLLUTION ASSOCIATED WITH A LANDFILL: CONCENTRATIONS IN WATER, SEDIMENT, CRAYFISH AND FISH

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James R. Morrissey

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Rodney J Sobieski

Approved for Major Department

David R. Eddy

Committee Chairman

[illegible]

Committee Member

David C Schroeder

Committee Member

Faye N Vowell

Approved for Graduate Council

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CONTENTS

LIST OF TABLES

LIST OF FIGURES

INTRODUCTION

Anthropogenic activities produce many, if not most, of the elements of environmental pollution. Metals are an important segment in the growing assemblage of chemicals which comprise this contamination. Lead, cadmium, aluminum, zinc and copper, metals used extensively by industrial nations, contribute to this widespread problem. Pb is used for positive plates in storage batteries, in making glass, and was widely used in the past as a gasoline additive and for pigments in paints. Cd is used mostly for electroplating onto steel, iron, copper, brass and other alloys to protect them from corrosion. Some Cd is converted to pigments for paints, used in storage batteries and combined with other metals to form alloys for making solder. Al has many uses, including construction of beverage containers, lawn furniture, storm windows and has numerous other applications requiring a non-corroding light-weight metal. Zn is used mainly for galvanizing iron and steel and for making brass. Cu also has several uses such as copper plumbing and wiring, and is combined with other metals to form alloys.

Landfills are repositories for a large proportion of the waste materials discarded by humanity. Trace metals undoubtedly find their way into landfills through the refuse of homes, factories and cities. These metals may manage to escape the confines of landfills through erosion, dissolution and their subsequent transportation by surface water.

Metals in water and sediment

Sprenger and McIntosh (1989) studied six northwestern New Jersey lakes whose water had detectable concentrations of Al, Cd, Pb, and Zn. These lakes had pH's ranging from 3.6 to 5.8, and researchers found lower concentrations of these metals in the least acidic lake and generally higher concentrations in the two most acidic lakes. This approximately inverse relationship between pH and concentrations of heavy metals in water has also been found by other researchers (Burrows and Whitton, 1983; Hakannson et al., 1989). Phillips (1977) stated that using water analysis to compare pollution at various sites has the disadvantage of large variations in metals concentrations due to factors such as time of day, season, extent of freshwater run-off and sampling depth. Thus, the study of heavy metal pollution in aquatic environments requires more than the sampling of water alone.

At pH's typically found in Kansas streams, trace metals may not be detectable in the water column. A study done in 1990 by Ron Falwell, an Emporia State University chemistry student, in the same area as the present study, showed no detectable levels of Pb or Cd in the water (D. Schroeder, pers. comm.). The pH of the water was 7.0 or above at the time of sampling.

Many times very low concentrations of trace metals in water belie higher concentrations in the sediment. At pH's of 7.0 and above, concentrations of trace metals in water are generally low, because at these pH levels heavy metals tend to form insoluble and sparingly soluble salts, which precipitate and coprecipitate out of the water column. Metals also can be absorbed in or adsorbed on detritus and sediment (Anderson et al., 1978; Manahan, 1984; Douben and Koeman, 1989; Timmermans et al., 1989). It is therefore necessary to analyze sediments along with water to ascertain if a pollution problem exists.

Sediments act as a sink and repository for metals and can release these metals into the rest of the environment (Douben and Koeman, 1989). Some metals return to the overlying water column by remobilization and upward diffusion. Even though the original source of contamination may have disappeared, contaminated sediments may continue releasing metals (Mohapatra, 1988). Natural processes and human-induced environmental changes, such as complexing and pH changes due to acid rain, may cause mobilization of accumulated pollutants. Activities such as dredging can cause remobilization when this causes sediment to be brought from anoxic to oxic conditions (Salomons et al., 1987). In northwestern New Jersey, Sprenger and McIntosh (1989) found that in the most acidic lakes studied the concentrations of metals in the water were the highest while concentrations in the upper 2.5 cm of sediment in the two most acidic lakes were considerably lower. This may indicate a flow of metals from the sediment into the water column.

Metals can be associated with both organic and inorganic fractions of the sediment. Pugsley et al. (1988) found a positive correlation between Pb and Cd concentrations and the amount of organic carbon present in the sediments of Lake St. Clair and the Detroit and St. Clair rivers. Anderson et al. (1978) stated that in the Fox River in Illinois, detritus usually had the highest metal concentration of all the abiotic components. These researchers pointed out, however, that much of this may have been contained in the microfauna and flora associated with the detritus, although this fraction was not analyzed.

Metals in organisms

In laboratory experiments Cd, Pb and Al have been shown to be lethal to bivalves, gastropods, amphipods, fish, chironomids and odonates (Pascoe and Cram, 1977; Wright and Frain, 1981; Pascoe and Shazili, 1986; Pascoe et al., 1986; McCahon et al., 1988; McCahon and Pascoe, 1988a; McCahon and Pascoe, 1988b; Pascoe et al., 1989; Mackie, 1989). Trace metals, while not reaching lethal levels, can have other health consequences at non-lethal levels. Enzymes can be immobilized because heavy metals attack sulfur bonds (Hodson, 1988). Cd, Cu, Pb and Hg ions bind to cell membranes, interfering with transport functions. Cd replaces Zn biochemically, which leads to high blood pressure and kidney damage, destruction of testicular tissue and red blood cells in humans and is toxic to aquatic life (Manahan, 1984). Lead causes anemia and dysfunction of the kidneys, reproductive system, liver, brain and central nervous system (Manahan, 1984).

Some studies have shown that shifts in sediment composition may affect contaminant concentrations, and thus sediment analysis alone may not produce an accurate portrayal of sites of contamination (Brook and Moore, 1988; Hakansson et al., 1989; Moore et al., 1989). Pugsley et al. (1988) showed that mollusks were better suited as an indicator of metal pollution than were sediments.

It might be expected to see a strong correlation between sediment concentration and body burden if consumption of tainted sediment was the principal source of heavy metals in clams. Just the opposite, however, was found by Pugsley et al. (1988). Such correlation studies fail to prove that clams obtain heavy metals from the sediment, and suggest they obtain most of their contamination directly from the water column. However, Pugsley et al. (1988), in their study of mollusks, did not determine concentrations of Pb and Cd in the overlying water column. Roesijadi and Klerks (1989) determined the oyster, *Crassostrea virginica*, accumulated Cd from water, primarily in the gills. Timmermans et al. (1989) found Pb and Cd levels in gastropods, bivalves, leeches, copepods, amphipods, isopods, water mites and insects lower or equal to those in the sediment. Anderson (1977) found body burdens of Cd, Cu, Pb and Zn in freshwater clams generally reflected concentrations of the sediment. Conversely, Douben and Koeman (1989) showed that stone loach (*Noemacheilus barbatulus*) in aquaria with sediments enriched with Pb and Cd had elevated body burdens while those kept with acid-washed sand usually had lower metal levels than control fish.

Selection of organisms for investigation of heavy metal pollution requires careful planning as not all aquatic organisms have the same affinity for accumulation of trace metals. Several investigators have shown correlations of body burdens with feeding habits and trophic levels (Smock, 1983a; Timmermans et al., 1989; Pugsley et al., 1988; Barak and Mason, 1989). Whether or not trace metals are subject to biomagnification or biotransference is a subject of debate. Some researchers have shown that these do occur while others have found no evidence to support their occurrence.

Enk and Mathis (1977) found aquatic insects had over five times the concentrations of Pb and Cd found in fish. In general, these researchers found both metals increased successively from water to fish to sediments to aquatic invertebrates. Burrows and Whitton (1983) compared metal concentrations in two carnivorous stoneflies and a carnivorous caddisfly with other invertebrates, many of which probably represented potential prey, and found that these concentrations did not increase up the food chain. In a study of Cu, Pb, Zn and Cd in invertebrates, biomagnification appeared to take place only for Zn in some predators and their prey (Timmermans et al., 1989). Patrick and Loutit (1978) fed tubificid worms that had high concentrations of Cr, Cu, Mn, Pb and Zn to tropical fish (Hyphessobrycon serpae) but found only Mn to be higher in predators than their prey. In their study of herbivorous, omnivorous and carnivorous vertebrates and invertebrates, Anderson et al. (1978) showed that while Cu and Zn appeared to demonstrate biomagnification in crayfish and snails, respectively, Pb and Cd did not. The reason for this, they felt, could be due partly to the ability of organisms to regulate Cu and Zn.

Several factors appear to affect body burdens of metals. Food is a prime suspect, but questions arise concerning how and where the it is obtained. Feeding habits in aquatic insects were implicated by Smock (1983a) as increasing whole-body metal concentrations by ingesting metal-enriched sediments and weight of the organism is increased by weight of ingested sediments, decreasing metal concentrations reported on a weight basis. Although both operate simultaneously, Smock (1983a) indicated the former appears to be of greater importance.

Thus, many factors may influence the concentration of metals in an ecosystem. Investigation and determination of metal concentrations using only one parameter may result in a distorted view of the pollutants in question. As past studies indicate, water, sediment and more than one organism at different trophic levels, need to be included in any study in order to fairly assess environmental contamination.

The present study was designed to investigate the possible impact of surface run-off from a landfill located on a tributary of the Cottonwood River. The Cottonwood River is a back-up source of drinking water for Emporia, Kansas and a primary source for communities downstream. Also, the river is fished heavily downstream from the landfill. The biota of the river and the health of the people who drink its water and eat its fish may be in jeopardy if heavy metals enter the river from the landfill. To investigate possible trace metal pollution, I sampled metal levels in the water, sediments, fish and crayfish in the area.

MATERIALS AND METHODS

Study sites

My study area was in central Lyon County, Kansas, southwest of the city of Emporia, and consisted of four sites; two on an unnamed intermittent stream which flows through a Lyon County landfill and empties into the Cottonwood River, and two sites in the Cottonwood River

(Figure 1). The old channel was cut off from the stream a few months prior to the commencement of this study. Site one was in the stream above the landfill; site two was in the same stream below the landfill; sites three and four were above and below, respectively, the confluence of the stream with the Cottonwood River. Site one served as a "control" for site two and site three served as a "control" for site four.

Field sampling

Four samples each of water, sediment, fish and crayfish were taken at each of the four sites. Fish and crayfish were collected by seine on 25 August 1990. Sediment was collected on 11 September 1990 with a home-made sampler consisting of 43 mm diameter PVC pipe with a rubber stopper. All samples were stored in polyethylene freezer bags. Water samples were also collected on September 11, 1990 and were taken from the surface with 30 ml acid-washed polyethylene sample bottles, taking care not to incorporate sediment into the samples. All samples were frozen and stored at -18°C (0°F) immediately upon returning from the field.

Orangespotted sunfish, *Lepomis humilis*, and crayfish, *Orconectes nais*, were selected for analysis because they

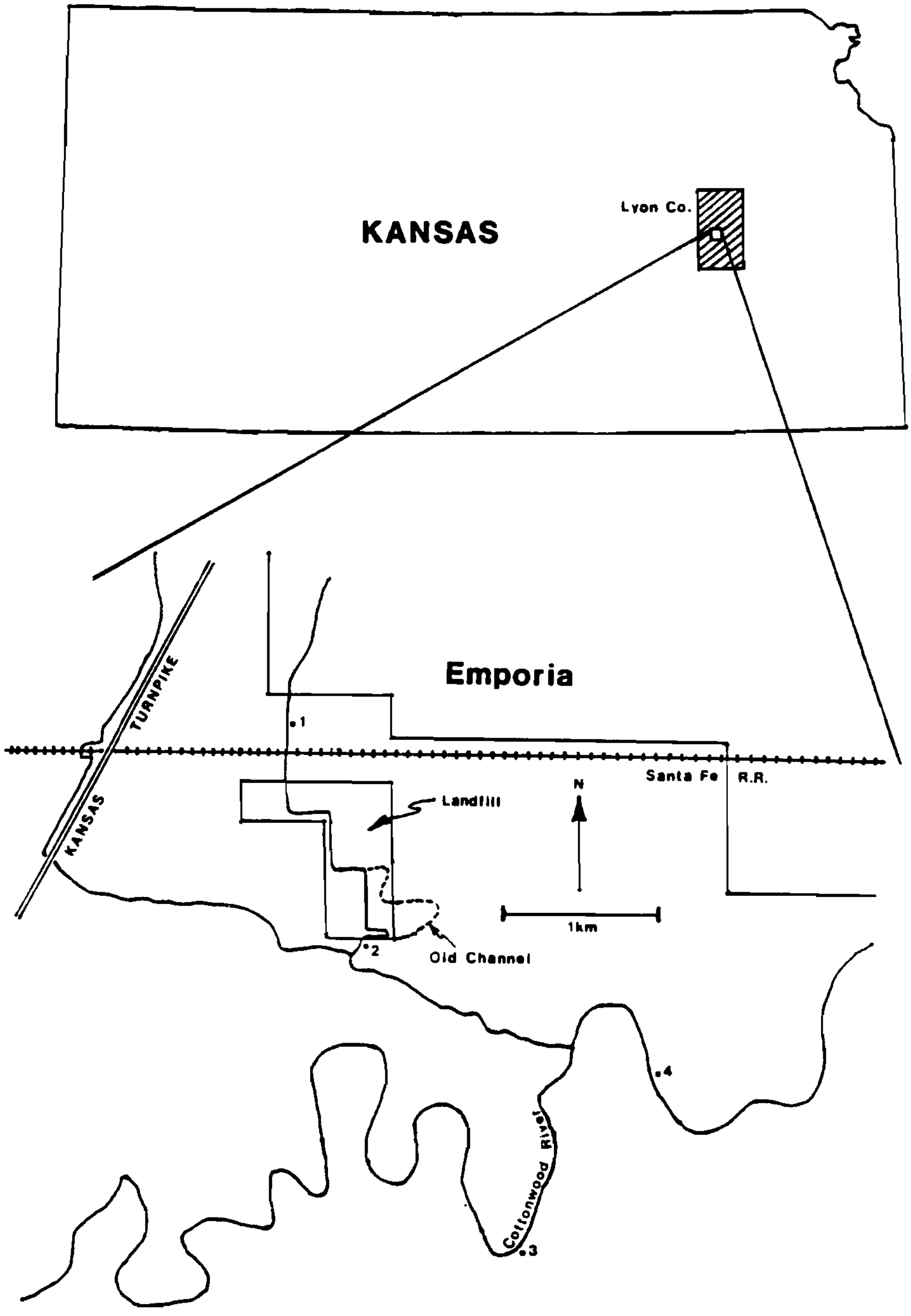

Figure 1. Map of study area in Lyon Co., Kansas.

were present at all four sampling sites. These two organisms represent different trophic levels as well as different feeding habits. Crayfish are considered omnivores and scavengers and frequently feed on detritus at the water-sediment interface (Pennak, 1981). Orangespotted sunfish are predators that consume small crustaceans, aquatic insects and small fishes (Pflieger, 1975).

Laboratory analysis

Analysis for metals in water, sediment and tissues was done in a variety of similar procedures (Beeby and Eaves, 1983; Hopkin et al., 1986; Pugsley et al., 1988; Barak and Mason, 1989; Case et al., 1989; Mackie, 1989; Sprenger and McIntosh, 1989). The procedure for this study was a modification of these.

Water samples (20 ml) were acidified with 5 ml of 1:1 nitric acid-deionized water solution. All other samples were dried at 105°C and ashed in a muffle furnace. Sediment samples were pulverized with a mortar and pestle. Residues were digested with 20 ml nitric acid and heated to just below the boiling point for one hour. Digested samples were transferred to volumetric flasks and diluted to 25 ml with deionized water. All samples were filtered to remove particulate matter. All glassware were acid washed with 1:1

nitric acid-deionized water and rinsed with deionized water.

There was potential for some loss of Pb and Cd because of the high ashing temperature and the volatility of these metals at this temperature. To determine extraction efficiency of the procedure, quality control samples were prepared by spiking two sediment and two fish samples with 5.0 mg Pb and 5.62 mg Cd. Sediment from site three and two wild orangespotted sunfish kept in an aquarium for several months were used for the quality control samples. These four samples were then prepared in the same manner as all other samples.

Concentrations of metals were determined by inductively coupled argon plasma atomic emission spectroscopy (ICAP) at the Kansas Department of Health and Environment laboratory in Topeka, Kansas. The ICAP detection limits were: Pb, 0.020 ppm; Cd, 0.002 ppm; Al, 0.026 ppm; Zn, 0.001 ppm; and Cu, 0.003 ppm. Two determinations for each metal were done on each sample and the mean used for the sample values. Concentrations of metals in sediment, crayfish and fish were converted to mg/kg dry weight.

Statistical analysis

Differences in concentrations of metals among sites were analyzed for water, sediment, crayfish and fish by one-way ANOVA (Sokal and Rohlf, 1981) with BIOSTAT II computer software (Pimentel and Smith, 1986). Dixon's test (Sokal and Rohlf, 1981) was used to determine if the lead levels found in crayfish, fish and sample two, site three sediment were outliers. Bartlett's test for homogeneity of variance was employed for all samples with mean greater than zero. Five of the 12 sample sets analyzed by ANOVA failed Bartlett's test, therefore, all data were log-10 transformed to improve normality. To determine which sites had significant differences, the Student-Newman-Keuls (S-N-K) multiple range test was used. In the case of Pb in water, in which only one site had detectable levels, ANOVA was not appropriate because there was no variance. In this situation a one-tailed Student's _t_-test was used to determine if the mean for this site two significantly greater than zero (Zar, 1974). Values for crayfish and fish from all four sites were also pooled into their respective groups and these two groups were compared by one-way ANOVA to determine if there were significant differences in their metal concentrations. All samples below ICAP detection limits were considered as zero in the statistical analysis.

RESULTS

Extraction efficiency for spiked quality control samples were as follows: Pb in sediment, 74.4%; Cd in sediment, 88.6%; Pb in fish, 73.5%; and Cd in fish, 67.0%.

Lead

Except for two samples of water (0.02 mg/l) and one sample each of crayfish (30.87 mg/kg) and fish (0.29 mg/kg), all samples of these three groups were below the ICAP Pb detection limit of 0.02 ppm (Table 1). The one-tailed Student's *t*-test for Pb in water was not significant (t = 1.74, d.f. = 3, $P > 0.05$).

Pb was present in sediment in all samples at all sites (Table 1). One of the samples from site three, however, was such a gross anomaly that it was suspect. Dixon's test for this value proved highly significant ($P < 0.01$), thus it was excluded from the ANOVA. Pb levels for the crayfish and fish samples were also anomalous. Dixon's test for these values was also highly significant ($P < 0.01$) and they were also excluded from statistical analysis. Possible

Table 1: Concentrations of lead, with S-N-K multiple range test results for samples of water, sediment, crayfish and orangespotted sunfish, Lyon Co., Kansas, 1990. Within columns, values followed by the same letter are not significantly different. All zero values were below detection limit.

	SAMPLE	WATER mg/l	SEDIMENT mg/kg*	CRAYFISH mg/kg*	FISH mg/kg*
SITE ONE	1	0.00	37.33	0.00	0.00
	2	0.00	21.52	0.00	(0.29)**
	3	0.00	19.41	(30.87)**	0.00
	4	0.00	23.69	0.00	0.00
	MEAN	0.00 a	25.49 a	7.72 a	0.07 a
	(±SE)	****	(±4.04)	(±7.72)	(±0.07)
SITE TWO	1	0.00	42.13	0.00	0.00
	2	0.02	28.14	0.00	0.00
	3	0.00	36.54	0.00	0.00
	4	0.02	32.38	0.00	0.00
	MEAN	0.01 a	34.80 b	0.00 a	0.00 a
	(±SE)	(±0.006)	(±2.99)	****	****
SITE THREE	1	0.00	24.01	0.00	0.00
	2	0.00	(26333.36)**	0.00	0.00
	3	0.00	21.67	0.00	0.00
	4	0.00	21.61	0.00	0.00
	MEAN	0.00 a	22.43 a	0.00 a	0.00 a
	(±SE)	****	(±0.79)	****	****
SITE FOUR	1	0.00	17.70	0.00	0.00
	2	0.00	17.51	0.00	0.00
	3	0.00	18.65	0.00	0.00
	4	0.00	18.43	0.00	0.00
	MEAN	0.00 a	18.07 a	0.00 a	0.00 a
	(±SE)	****	(±0.28)	****	****

* DRY WEIGHT

** OUTLIER EXCLUDED FROM STATISTICAL ANALYSIS explanations for these high values might be that lead shot, lead fishing weight or perhaps a piece of lead metal from old automobiles in the river at this site was incorporated into these samples.

Pb levels in sediment were significantly different among sites ($F = 8.94$, d.f. $= 3,11$, $P < 0.003$). The S-N-K multiple range test showed site two, below the landfill, was significantly higher than the other three sites (Table 1).

Cadmium

Six of the 16 water samples had Cd levels at or above the ICAP detection limit of 0.002 ppm (Table 2). Cd was detected in all samples of sediment, crayfish and all but one sample of fish (Table 2). There were no significant differences among sites for Cd levels in water, crayfish or fish. Levels in sediment did, however, exhibit a significant difference among sites ($F = 8.22$, d.f. $= 3,12$, $P < 0.004$). The S-N-K multiple range test showed site two, below the landfill, had significantly higher levels of Cd than the other three sites (Table 2).

Table 2: Concentrations of cadmium, with S-N-K multiple range test results for samples of water, sediment, crayfish and orangespotted sunfish, Lyon Co., Kansas, 1990. Within columns, values followed by the same letter are not significantly different. All zero values were below detection limit.

	SAMPLE	WATER mg/l	SEDIMENT mg/kg*	CRAYFISH mg/kg*	FISH mg/kg*
SITE ONE	1	0.003	0.761	0.243	0.060
	2	0.002	0.358	0.095	0.100
	3	0.002	0.413	0.076	0.073
	4	0.000	0.454	0.131	0.000
	MEAN	0.002 a	0.497 a	0.136 a	0.058 a
	(±SE)	(±0.001)	(±0.090)	(±0.037)	(±0.021)
SITE TWO	1	0.000	0.987	0.193	0.047
	2	0.010	0.805	0.294	0.037
	3	0.000	0.932	0.083	0.021
	4	0.008	0.647	0.098	0.052
	MEAN	0.005 a	0.843 b	0.167 a	0.039 a
	(±SE)	(±0.003)	(±0.076)	(±0.049)	(±0.007)
SITE THREE	1	0.000	0.678	0.243	0.048
	2	0.000	0.462	0.463	0.046
	3	0.002	0.790	0.121	0.052
	4	0.000	0.451	0.248	0.084
	MEAN	0.001 a	0.595 a	0.269 a	0.058 a
	(±SE)	(±0.001)	(±0.083)	(±0.071)	(±0.008)
SITE FOUR	1	0.000	0.345	0.161	0.025
	2	0.000	0.364	0.275	0.116
	3	0.000	0.365	0.251	0.039
	4	0.000	0.371	0.236	0.074
	MEAN	0.000 a	0.361 a	0.231 a	0.064 a
	(±SE)	*****	(±0.006)	(±0.025)	(±0.035)

* DRY WEIGHT

Aluminum

Al was detected in all samples of water, sediment, crayfish and fish (Table 3). There was no significant difference among sites for Al in crayfish samples (Table 3). There were significant differences among sites for water ($F = 7.29$, d.f. = 3,12, $P < 0.005$), sediment ($F = 35.10$, d.f. = 3,12, $P = 0.00001$) and fish ($F = 7.21$, d.f. = 3,12, $P < 0.006$). Al levels in water were significantly higher at site two (Table 3). Levels of Al in sediment at site two were significantly higher than the other three sites and site three was significantly lower than site two but significantly higher than the remaining two sites (Table 3). For fish the test showed significantly lower levels at site two, below the landfill (Table 3).

Zinc

Concentrations of Zn were found in all samples of water, sediment, crayfish and fish (Table 4). There were no significant differences for Zn levels among sites for water, fish or crayfish (Table 4). There was a significant difference among sites for sediment ($F = 3.79$, d.f. = 3,12, $P < 0.05$). The S-N-K multiple range test showed site two, below the landfill, was significantly higher than site one, above the landfill, but neither was significantly different

Table 3: Concentrations of aluminum, with S-N-K multiple range test results for samples of water, sediment, crayfish and orangespotted sunfish, Lyon Co., Kansas, 1990. Within columns, values followed by the same letter are not significantly different.

	SAMPLE	WATER mg/l	SEDIMENT mg/kg*	CRAYFISH mg/kg*	FISH mg/kg*
SITE ONE	1	3.071	23201	644.03	248.36
	2	0.906	23440	1026.62	536.75
	3	0.418	21462	1537.05	136.98
	4	0.699	21941	1243.11	110.53
	MEAN (±SE)	1.274 a (±0.607)	22511 a (±480)	1112.70 a (±188.00)	258.16 a (±97.55)
SITE TWO	1	4.168	36325	1510.00	55.99
	2	3.213	28625	2062.80	54.88
	3	3.203	36046	1923.71	23.84
	4	9.608	33721	2195.63	17.21
	MEAN (±SE)	5.048 b (±1.537)	33679 b (±1783)	1923.03 a (±148.46)	37.98 b (±10.17)
SITE THREE	1	2.424	28753	1304.53	125.64
	2	1.137	27837	2199.41	79.89
	3	1.278	31237	1416.12	104.57
	4	1.129	30083	1012.50	79.94
	MEAN (±SE)	1.492 a (±0.313)	29478 c (±746)	1483.14 a (±253.47)	97.51 a (±11.03)
SITE FOUR	1	0.773	23246	2848.33	46.70
	2	0.893	23649	889.62	198.60
	3	0.984	22741	664.49	111.06
	4	1.708	22341	1781.54	144.52
	MEAN (±SE)	1.090 a (±0.211)	22994 a (±286)	1546.00 a (±496.61)	125.22 a (±31.78)

* DRY WEIGHT

Table 4: Concentrations of zinc, with S-N-K multiple range test results for samples of water, sediment, crayfish and orangespotted sunfish, Lyon Co., Kansas, 1990. Within columns, values followed by the same letter are not significantly different.

	SAMPLE	WATER mg/l	SEDIMENT mg/kg*	CRAYFISH mg/kg*	FISH mg/kg*
SITE ONE	1	0.058	200.32	100.693	40.607
	2	0.035	71.94	87.470	54.146
	3	0.040	50.17	102.429	60.524
	4	0.032	70.65	57.917	59.785
	MEAN	0.041 a	98.27 a	87.127 a	53.766 a
	(±SE)	(±0.006)	(±34.38)	(±10.294)	(±4.612)
SITE TWO	1	0.034	199.95	119.350	47.086
	2	0.029	161.10	167.055	45.872
	3	0.029	192.01	87.426	48.125
	4	0.328	178.10	75.603	36.911
	MEAN	0.105 a	182.79 b	112.359 a	44.499 a
	(±SE)	(±0.074)	(±8.52)	(±20.439)	(±2.571)
SITE THREE	1	0.026	126.94	84.114	24.324
	2	0.021	120.32	129.167	76.088
	3	0.011	93.93	53.667	50.943
	4	0.014	100.77	124.264	30.409
	MEAN	0.018 a	110.49 ab	97.803 a	45.441 a
	(±SE)	(±0.003)	(±40.32)	(±17.840)	(±11.695)
SITE FOUR	1	0.014	125.12	132.362	58.117
	2	0.011	121.86	85.227	156.334
	3	0.014	78.41	77.942	67.611
	4	0.012	120.33	91.404	43.251
	MEAN	0.013 a	111.43 ab	96.774 a	81.328 a
	(±SE)	(±0.001)	(±11.05)	(±12.191)	(±25.499)

* DRY WEIGHT

from sites three and four (Table 4).

Copper

Cu was present in all samples of water, sediment, crayfish and fish (Table 5). There were no significant differences in Cu levels among sites for water or crayfish (Table 5). There were significant differences among sites for Cu levels in sediment ($F = 64.34$, d.f. = 3,12, $P < 0.00001$) and fish ($F = 5.24$, d.f. = 3,12, $P < 0.016$). The S-N-K multiple range test showed there were significant differences among all sites for sediment samples (Table 5). The rankings of the means for Cu in sediment were: site 3 > site 2 > site 4 > site 1. For fish, the test showed site one, above the landfill, was significantly higher than the other three sites (Table 5).

Crayfish vs. Fish

Values of metal levels for Cd, Al, Zn and Cu at all four sites were pooled for crayfish and fish then compared. Levels of all four metals were significantly higher in crayfish than fish (Cd: $F = 31.43$, d.f. = 1,30, $P < 0.00001$; Al: $F = 130.19$, d.f. = 1,30, $P < 0.00001$; Zn: $F = 22.57$, d.f. = 1,30, $P < 0.00001$; Cu: $F = 368.38$, d.f. = 1,30, $P < 0.00001$).

Table 5: Concentrations of copper, with S-N-K multiple range test results for samples of water, sediment, crayfish and orangespotted sunfish, Lyon Co., Kansas, 1990. Within columns, values followed by the same letter are not significantly different.

	SAMPLE	WATER mg/l	SEDIMENT mg/kg*	CRAYFISH mg/kg*	FISH mg/kg*
SITE ONE	1	0.025	9.963	35.955	3.608
	2	0.018	7.633	92.608	5.479
	3	0.016	7.746	65.301	2.495
	4	0.014	7.513	47.537	17.650
	MEAN	0.018 a	8.214 a	60.350 a	7.308 a
	(±SE)	(±0.002)	(±0.585)	(±12.330)	(±3.502)
SITE TWO	1	0.013	15.403	56.217	2.105
	2	0.014	13.093	52.003	1.438
	3	0.012	15.573	55.108	1.034
	4	0.038	14.377	61.521	1.174
	MEAN	0.019 a	14.612 b	56.212 a	1.438 b
	(±SE)	(±0.006)	(±0.571)	(±1.982)	(±0.238)
SITE THREE	1	0.013	17.922	57.098	0.687
	2	0.012	18.424	56.702	1.375
	3	0.007	18.660	61.968	1.798
	4	0.009	16.458	69.582	2.308
	MEAN	0.010 a	17.866 c	61.338 a	1.542 b
	(±SE)	(±0.001)	(±0.494)	(±2.998)	(±0.343)
SITE FOUR	1	0.012	12.369	75.557	2.535
	2	0.006	12.784	77.781	1.896
	3	0.005	12.557	69.751	2.300
	4	0.004	12.181	87.913	3.349
	MEAN	0.007 a	12.473 d	77.751 a	2.520 b
	(±SE)	(±0.002)	(±0.129)	(±3.787)	(±0.306)

* DRY WEIGHT

DISCUSSION

Water

As expected, because of typical alkaline pH levels in local streams, Pb, Cd, Zn and Cu in water were below or slightly above the detection limits of the ICAP. Although pH levels were not measured, water in streams and rivers in eastern Kansas generally is neutral or slightly alkaline (Geiger et al. 1991), due to underlying limestone. As noted earlier, metals generally precipitate out of the water at these pH's. Another factor which can affect metal concentrations in water is dilution. Run-off from areas free of the source of contamination can reduce metal concentrations.

Al, unlike the other metals, was well above the ICAP detection limit. Sprenger and McIntosh (1989) found Al to be in relatively higher concentrations in water than Pb, Cd and Zn, as did this study. Al levels in water were significantly higher at site two, below the landfill, than at other sites. This suggests that the landfill is a source of Al in water at this site.

Al is a naturally occuring mineral in many clays (Donahue et al., 1983), and soils in the vicinity of the landfill are high in clays (USDA, 1981). This is a likely reason for the relatively high concentrations of Al seen in all samples. Differences among sites may be due to the relative amount of clay at each site. The relative proportions of clay were not determined for the sediment samples. A general observation, however, was that sediment samples taken from below the landfill were much finer in texture than those of the other sites. Since clays are the finer particles found in sediment it can be postulated that the sediment samples from below the landfill would have higher levels of naturally occuring aluminum. Furthermore, these higher levels would also be reflected in the water and aquatic organisms.

Sediment

Sediment generally had higher levels of metals than water, crayfish and fish. Several other researchers have also found very low levels of metals in the overlying water column although there were relatively high levels in sediments (Anderson, 1977; Enk and Mathis, 1977; Anderson et al., 1978; Burrows and Whitton, 1983; Barak and Mason, 1989). This evidence supports the hypothesis that sediments are a sink for trace metal pollution.

The landfill appears to be a major source of metal contamination in sediment. Pb, Cd and Al were all significantly higher at site two, below the landfill. As explained earlier elevated Al levels could be due to possibly higher proportions of clay in the samples at this site. Zn was significantly higher at site two than site one though neither was significantly different from sites three and four, which are also polluted by metal debris. Automobiles, major household appliances and common household trash can be seen littering the banks of the river. This could have been the reason for the higher levels of metals encountered in sediment at sites three and four. The pattern for Cu in sediment was quite dissimilar. Cu was significantly different at all sites, with the trend being 3 > 2 > 4 > 1. Site three had the highest mean concentration of Cu. Why site three had the highest mean concentration of Cu is unclear. The landfill is, however, a source of Cu contamination as the site below the landfill did have the second highest mean concentration in the sediment.

Crayfish

Pb concentrations, or lack of, in crayfish are in contrast with sediment levels. The lone crayfish sample containing Pb is not consistent with all other samples, although the two runs were similar (1.475, 1.549 ppm). This sample's Pb level could be due to contamination from an outside source. It is also possible that small particles of Pb metal could have been ingested by the crayfish along with its food. Keeping crayfish in aquaria just long enough until they cleared their gut would eliminate this possibility.

Cd was present in all crayfish samples. However, since there were no significant differences among sites, there is no evidence that the landfill is a source of Cd body burdens in crayfish. Al, Zn and Cu were also present in crayfish, but again, no significant differences among sites implies sources other than the landfill.

Fish

The only fish sample containing Pb may be erroneous. The two ICAP runs on the extraction solution were relatively far apart (-0.0001, 0.0405 ppm), and therefore questionable. Lack of any significant differences among sites for Cd suggests there are sources other than the landfill that are contributing to fish body burdens. However, there was a significant difference among sites for Al in fish. Site two, below the landfill, was significantly lower than the other sites. If clay is the source of Al in sediment, the levels found in fish could be a function of the amount of the clay fraction of the sediment ingested along with food. Since fish are mobile, the fish sampled at each site may have been feeding at areas with lower or higher relative amounts of clay in the sediment. Perhaps, the fish should also be kept alive in an aquarium just long enough to clear their gut to eliminate this possibility. Once again, due to the relatively high amounts of debris at all sites, Al levels could be due to ingestion of minute particles of the metal. Although Zn was present at all sites there were no significant differences. Cu in fish was significant at site one, and was almost three times as high as the next highest site. This was due mainly to one relatively high sample at site one. As with all relatively high samples, there is the possibility that the organism ingested a particle of metal debris.

Comparison of Crayfish and Fish

Significantly lower body burdens of Cd, Al, Zn and Cu in fish compared to crayfish could be due to the higher mobility of fish, as fish have the ability to move in and out of areas with higher contamination levels. This difference in body burdens between fish and crayfish is more likely due, however, to either food sources or the close association of crayfish with sediment. Smock (1983a) found that sediment dependent organisms, those that generally live within the sediment and indiscriminately ingest it along with detritus, had the highest concentrations of most metals. Filter feeders usually had the next highest concentrations, followed by sediment associated organisms, which live on and in the sediment but, to some extent, selectively ingest detritus, some animals and plant materials. Comparing trace metal concentrations in deposit feeders and filter feeders, Timmermans et al. (1989) found substantial differences, and postulated those differences showed that factors other than trophic position also played a role as deposit feeders live in closer association with the sediment than filter feeding organisms, though they both occupy the same trophic level. The authors hypothesized that lead, zinc and copper concentrations, which were all higher (although not always significantly) in deposit feeders, indicated the influence of proximity to the sediment. Anderson et al. (1978) found that detritus usually had the highest metal concentrations of the abiotic components of the aquatic ecosystems of the rivers studied, and as crayfish frequently feed on detritus, this could account for the higher concentrations of metals observed in crayfish in my study.

Ney and Van Hassel (1983) studied two centrarchids, rock bass (*Ambloplites rupestris*) and redbreast sunfish (*Lepomis auritus*), which suspend themselves in the water column, and four bottom-dwelling species, northern hog sucker (*Hypentelium nigricans*), white sucker (*Catostomus commersoni*), blacknose dace (*Rhinicthys atratulus*) and fantail darter (*Etheostoma flabellare*). Of these six species, they found that the bottom-dwelling species generally had higher levels of metals. The orangespotted sunfish, a centrarchid examined in this study, is also a species that suspends itself in the water column. This lack of close association with the sediment appears to be a limiting factor for body burdens of metals in contaminated streams.

Absolute Levels of Metals

The Kansas Department of Health and Environment is currently proposing new maximum acute and chronic levels for pollutants in surface water in Kansas (Robert Angelo, pers. comm.) (Appendix). No levels have been proposed for Al, nor have maximum levels been proposed for sediment or aquatic organisms.

In a laboratory study of tolerances of benthic invertebrates to levels of Cd, Pb and Al, Mackie (1989) found Cd generally equal to or more toxic than Pb, depending on the species. But even at the lowest pH, 1000 µg/l of Al was not acutely toxic (96-hr LC50) to any of the test species. Ten of the sixteen water samples in my study exceeded this level. It is unknown if these higher levels pose a health risk to organisms.

In my study, Pb was present in water only at site two. This was not significantly higher than the other sites, but the mean concentration was above the KDHE proposed maximum safe levels for hardness < 251 mg/l $CaCO_3$ (Appendix). The mean concentration of Cd at both sites one and two was above recommended acute levels for hardness < 150 mg/l and above chronic levels for hardness < 251mg/l. Cd at site three was above the chronic level for hardness < 150 mg/l hardness. The mean concentration of Zn at site one was above both acute and chronic safe levels below 150 mg/l. For Cu, all four sites exhibited mean levels above both acute and chronic safe levels for hardness < 150 mg/l hardness. Sites one and two for Cu were also above safe chronic levels for hardness < 251 mg/l. Thus, although metal concentrations at some sites were not significantly different from others, they exceeded recommended maximum safe levels.

Metals - General Patterns

The general patterns for concentrations of metals in the various components of my study were Pb: water, fish, crayfish < sediment; Cd, Al, Zn: water < fish < crayfish < sediment; and Cu: water < fish < sediment < crayfish.

Copper is a requirement for hemocyanin, an oxygen-carrying pigment, in the blood of crayfish and several other invertebrates (Villee et al., 1968). This is the most probable explanation for its higher observed concentration in crayfish compared to fish, although close association with the sediment may also be a factor.

Other researchers' results vary from study to study regarding general trends of metal levels. Enks and Mathis (1977) found the trend for Pb and Cd to be water < fish < sediment < invertebrates. Burrows and Whitton (1983) found the pattern for Pb, Cd and Zn to be water < invertebrates < sediment. Timmermann et al.'s (1989) results for Pb showed water < invertebrates < sediment and for Cd, Zn and Cu to be water < sediment < invertebrates. One pattern common to all of these and my study was that water had the lowest concentrations of metals of all components studied.

Factors Affecting Body Burdens

Concentration of pollutants along food chains involves not only ecology, but is also a function of physiology and biochemistry. Moriarty and Walker (1987) postulated that for aquatic species, there is relatively less of a chance for accumulation along food chains than for terrestrial species, because the water could provide a certain amount of metals to all organisms regardless of trophic position.

The origin of body burdens of metals in fish and crayfish in my study area is unclear at this time. Some or all of the metal concentrations could have come from ingestion of sediments and some of the levels could be due to absorption directly from the water.

Organism size is another factor which appears to influence body burdens, and it may do so in two ways (Timmermans et al., 1989; Smock, 1983b). The first is due to the surface to volume ratios of the organisms. The larger the surface area in relation to volume, the more metals can be absorbed on a weight basis. The second is the influence of body size on the size and type of material which can be ingested. Smaller particles, due to higher surface-to-volume ratios, may have higher concentrations of metals, although metals may differ in their afffinity for soil particle size (Moore et al., 1989). Body size could thus influence metal concentrations by restricting ingestion to smaller particles.

Anderson et al. (1978) found that even though two sites studied had different inputs of trace metals from two cities, one with run-off from an industrial complex and one without, Cu and Zn showed no significant differences in the biota between sites. They believed this to be due to physiologic control of these metals.

Handy and Eddy (1990) showed that body mucus of rainbow trout, *Oncorhynchus mykiss*, can absorb aluminum and zinc from the water. They believed this mucus may have a protective role by reducing passive ion loss and toxic metal accumulation. Perhaps metal levels in fish with large surface-to-body ratios should not be determined using whole-body methods, as this could cloud the picture of true internal body burdens. A large surface-to-body ratio increases the relative size of the mucous layer and thus increases the amount of metals which could be absorbed. In cases where metals in the water are high, specific organs need to be analyzed instead of whole fish, or else the mucous layer should be removed prior to analysis.

Ney and Van Hassel (1983), in their study of heavy metals in fish, concluded that long-lived benthic species should be selected, and tissues with demonstrated affinities for specific metals be analyzed. This conclusion was based on evidence which showed that fish species which had a close association with the sediment had higher whole-body metal concentrations than those that didn't have a close association. As metal concentrations may increase with exposure time, they also suggested that older fish should be included.

Thus, there are many parameters which can and should be investigated in a study of heavy metal pollution in aquatic environments. Their inclusion at the outset of experimental design is of prime importance. The present study investigated the three most studied and obvious of these elements: water, sediment and organisms. Other factors which need to be considered during study design include physicochemical parameters, trophic levels, feeding habits, seasonal changes and toxicity.

Many factors affect concentrations of metals in water, sediment and organisms. There is no evidence the landfill is a major source of contaminants for water or organisms. However my data show the landfill is a significant contributor of trace metals to the sediment of the stream that flows through it.

SUMMARY

Concentrations of five metals, Pb, Cd, Al, Zn and Cu, were determined in samples of water, sediment, crayfish (Orconectes nais) and orangespotted sunfish (Lepomis humilis). Samples were taken from four sites in the vicinity of a Lyon County, Kansas landfill. Site one was on a tributary stream immediately above the landfill. Site two was immediately below the landfill on the same stream. Site three was above the confluence of the stream and the Cottonwood River and site four was below this confluence.

The general patterns for concentrations of metals in the various components were Pb: water, fish, crayfish < sediment; Cd, Al, Zn: water < fish < crayfish < sediment; Cu: water < fish < sediment < crayfish.

The landfill appears to be a source of Pb, Cd, Al, Zn and Cu in sediment. The clay content of the samples was not determined, however, and since clays in the sediment may be a major source for Al there is still some doubt about its origin. There is no clear indication the landfill is a source of metals for water, crayfish or orangespotted sunfish. Al was significantly higher in water at site two, but concentrations in water could be influenced by the clay

in the sediment.

Al was the only metal with significant differences in concentration among sites for water; it was higher at site two. Pb, Cd and Al were significantly higher in sediment at site two. Zn was higher in sediment at site two than at site one, but was not significantly different from sites three and four. Cu in sediment was different among all sites, as follows: site three > site two > site four > site one. There were no significant differences among sites for any metal levels in crayfish. Al was significantly lower in fish at site two, whereas Cu was significicnatly higher in fish at site one.

Comparing crayfish to fish showed crayfish had significantly higher mean concentrations of Cd, Al, Zn and Cu. These differences could possibly be explained by the close association of crayfish with the sediment. Cu, however is a necessary component of hemocyanin in crayfish and this could account for the higher levels.

Although there were no significant differences among sites for Pb, Cd, Zn and Cu in water, some sites had levels higher than Kansas Department of Health and Environment recommended maximum safe levels for water within the state.

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APPENDIX

Maximum safe levels in water for aquatic life and public health proposed by the Kansas Department of Health and Environment (µg/l). Levels are hardness dependent, with hardness expressed as mg/l $CaCO_3$

METAL	HARDNESS < 150	150 - 250	251 - 400	> 400
LEAD				
Acute	34.0	138.0	265.0	478.0
Chronic	1.3	5.4	10.3	18.6
CADMIUM				
Acute	1.8	6.2	11.1	18.7
Chronic	0.66	1.56	2.34	3.37
ALUMINUM				
Acute	None Proposed			
Chronic	None Proposed			
ZINC				
Acute	65.0	165.0	255.0	458.0
Chronic	59.0	149.0	231.0	414.0
COPPER				
Acute	9.2	26.0	42.0	65.4
Chronic	6.5	16.7	26.0	38.7

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Jackie Tolbert
Signature of Graduate Office Staff Member

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