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T. W. Struttmann
University of Kentucky

Lyle V. A. Sendlein
University of Kentucky

Dave Fletcher
University of Kentucky

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ABSTRACT

This study reviewed the literature on the occurrence of lead in soils and its relationship to waste oil tank leaks. Many studies have been conducted on the natural occurrence of lead in rocks, soils and water. Very low levels (0.001mg/l to 0.01 mg/l) are found in surface and groundwater, variable levels are found in soils (from less than 10 ppm to as high as 700 ppm but more normally a high of 70 ppm), and the lead levels found in rocks range from 7 to 80 mg/kg.

Risk assessment calculations have been made by several individuals, also with variable results. One value was computed at 20 mg/kg lead in soil and another was calculated as high as 200 mg/kg. A standard of 25 mg/kg is proposed for Kentucky based on the natural background level found in soils studied in the state. For sample values taken in tank pits that do not meet this standard a background value must be determined by taking five soil samples at one-meter depth upgradient from the tank pit and averaging them.

Even though the recommended cleanup level is not based on risk analysis methods, when referring to various risk documents, the value chosen here is in accord with the value derived in some of these papers. The value selected here is slightly lower than the mean value (36 mg/kg) derived from an analysis of background samples at various remediation sites across the state. Standards in place in other states range from less than 0.1 mg/kg to as high as 2000 mg/kg. Texas uses 20 times the MCL, and Pennsylvania uses 200 ppm for non-industrial sites and 600 ppm for industrial sites.

BACKGROUND

The occurrence of heavy metals in soils is of concern because they have been reported in soils found to be contaminated by leaking waste oil tanks and pits. The heavy metal that has drawn the most attention is lead (Pb), because it has been found in the environment at elevated concentrations that can be toxic to humans (USHHS, 1992). Human use of lead has caused the element to be widely dispersed in the environment in places it historically has not been found. Specifically, beginning in the 1920s the use of tetra-ethyl lead in gasolines for automobile engines has resulted in increased concentrations of lead in soils covering large segments of the nation. This lead was expelled as an exhaust product into the air. With the proliferation of automobiles, the amount of lead released into the environment has increased substantially. Recently, lead has been banned as a gasoline additive.

Lead is a heavy, ductile, bluish-white metal that is found in Group IVB of the periodic table. It is fairly common in the earth's crust, found in concentrations of 12 $\mu g/g$. Daily oral intake of lead in this country averages about 100 μg; some European countries average less than 30 μg/day (Grandjean, 1991). Toxicological effects of lead include anemia, encephalopathy and other central and peripheral nervous system effects, impaired kidney function, muscle weakness, anorexia, constipation and memory deficits. Children are much more susceptible to the effects of lead than adults. It is estimated that absorption of lead in the gastrointestinal tracts of children may be as high adults. It is estimated that absorption of lead in the gastrointestinal tracts of children may be as high as 50%.

The purpose of this document is to establish an acceptable cleanup level for lead during underground storage tank (UST) remediations. The cleanup level that is determined will be compared to background levels found in other studies. This cleanup level will be generic, conservative, and will apply to all UST remediations in the state. This type of generalization has been done before in other states and has been shown to be acceptable and defendable (Logan and Miller, 1983).

Natural Occurrence

The natural occurrence of lead has been disturbed by the emissions from automobiles and other industrial sources and thus it is very difficult to determine what a natural background level for any given area is. Lead does occur in rocks in various concentrations and has been measured in surface water and groundwater. A brief discussion of these occurrences in general and in Kentucky is presented below.

Land Surface Occurrence

Struttmann (1993) presented information collected in Kentucky for the surface occurrence of lead in soils. His data showed that there is a wide range in soil concentrations with an average concentration of 25 mg/kg soil. The data that follow in the results section of this document list average lead levels found approximately 3 feet below the surface at UST sites in Kentucky.

Rocks and Soils

The occurrence of heavy metals in natural rocks has been compiled by several individuals (Pettijohn, 1957; Drever, 1982). The values for lead range from 20 to 80 mg/kg for shale; 9 to 16 mg/kg for limestone; and 7 to 20 mg/kg for sandstone. The resulting soils that develop over these rocks as weathering products will generally have smaller values than these, but under certain conditions the values may be higher. The U. S. Geological Survey (Shacklette and Boerngen, 1984) sampled soils or other regolith at a depth of 20 centimeters from locations approximately 80 kilometers apart throughout the conterminous United States. Twenty-seven sample sites were located in Kentucky with lead values ranging from less than 10 ppm to greater than 30 ppm.

Studies conducted in Missouri (Tidball, 1984) and Kentucky (Karathanasis and Seta, 1993; Wells, Henson and Kelly, 1993) for agricultural soils provide lead values that range from lows of 5 mg/kg to highs of 65 mg/kg. The Missouri study analyzed 1140 samples and found lead to have a wide range of values, with a geometric mean of 20 mg/kg and a geometric deviation of 1.55 mg/kg. Wells et al. (1993) studied soil samples in the top six inches. The mean values for 30 sites studied gave a range of 32 - 50 mg/kg of lead. These values are closer to those found by Struttmann (1993) and those in this study. Karathanasis and Seta (1993) looked at soils spread across Kentucky from 15 counties with 20 soils sampled. Their results showed that lead in surface soil horizons had a mean of 26.2 mg/kg and a standard deviation of 13.9 mg/kg, and that surface and subsoils had a mean of 24.4 mg/kg and a standard deviation of 13.1 mg/kg.

Water

The natural mobility of lead is very low due to its low solubility. The solid forms of lead are commonly found as hydroxy carbonates and phosphates in the soils and rocks (Hem and Durum, 1973; Hem, 1976b; Bilinski and Schindler, 1982; Nriagu, 1974). The adsorption of lead on organic and inorganic sediments and the co-precipitation of lead with manganese oxide also tend to maintain low concentrations in surface waters and groundwaters (Hem, 1976a; Hem, 1980).

The principal dissolved inorganic forms of lead are the free ion Pb^{2+}, hydroxide complexes, and probably the carbonate and sulfate ion pairs. The importance of organic complexes is uncertain, but they may constitute a significant part of the dissolved lead in some waters. Typical concentration in surface water is 1 microgram per liter, with some readings as high as 10 micrograms per liter. Some rain waters have been measured to have as much as 200 micrograms per liter in highly populated areas, but more common concentrations are near 1 microgram per liter.

Fate and Transport of Lead

Various lead compounds have solubility products that indicate that under the right Eh-pH conditions, lead solubility would be limited in natural waters. For example $PbCl_2$ has a K_{sp} = 10E-4.8 and other lead compounds' solubility products range upwards to 10E-27.5. Lead and other metals are cations that can be expected to undergo cation exchange with clays. Hence, the mobility of lead in groundwater is limited. A study (Hem, 1985) at a battery manufacturing facility at Medley, Florida, showed the soil to be contaminated to lead amounts of 98,600 mg/kg; however, the shallow groundwater immediately beneath the contaminated soil averaged less than 10 micrograms/liter of lead with the maximum value measured being 31 micrograms/liter of lead. The soils at the site were high in carbonate and clay. This exemplifies the low mobility of the lead in the soil.

A K_{oc} value for lead has been listed by Pavlou (1987) for marine sediments as 38×10^4 with a standard deviation of 40×10^4. This is another indicator that the lead in solution will have a high affinity for the soil materials and will not move far from the point of release.

RISK ASSESSMENT ANALYSIS

Lead has been reported in waste oils at levels of 200 to 300 ppm (USHHS, 1992). The USEPA has been unable to document toxicological research that allows for standard risk assessment analysis.

Status Of Current Knowledge

Birge et al. (1993) pointed out that attempts were made to perform a risk-based evaluation of lead from used oil USTs but several problems were encountered. Because slope factors for lead as an IARC Class B2 carcinogen are problematic the USEPA recommended that a numerical estimate of cancer potency or risk based on animal dose response data not be used due to uncertainties involved in the extrapolation process (USHHS, 1992). When using the oral slope factor of 1×10^{-7} for tetraethyl lead the values obtained were far below the background levels for this metal.

Westerman, KNREPC (1991, 1993), reviewed the literature and made a risk assessment analysis for lead, arriving at a level of 20 ppm to leave in the soil. He pointed out that not only animal test data are available but that information on humans has also been obtained. He also pointed out that the cancer potency factor for humans has not been determined for lead and that even though data are available on the effect to humans, a reference dose has not been developed.

The bottom line is that although a risk-based level for lead may not have been scientifically determined, the analyses to date seem to point to 20 mg/kg for soils and somewhere between 0.3 and 9 micrograms/liter for groundwater.

Standards In Other States and Nations

Soil quality and cleanup levels used by other nations (Chrostowski, 1994) range from 0.1 to 2000 mg/kg. For example, West Germany identifies normal levels to be between 0.1 and 20 mg/kg and tolerable as 100 mg/kg, and the Netherlands identifies good soil quality as 85 mg/kg, with moderate contamination as 150 mg/kg and severe contamination as 600 mg/kg. England has two levels, domestic gardens, with allotments at 500 mg/kg, and parks, playing fields, open space, with allotments at 2000 mg/kg. Ontario, Canada, identifies residential/agricultural land uses to have acceptable levels of 60 mg/kg, commercial/parkland uses of 500 mg/kg, and industrial land uses of 1000 mg/kg.

In Texas, the cleanup level of lead is determined by taking 20 times the MCL for lead regardless of the background level. A higher level can be used if a risk assessment is conducted for the site. In Tennessee the UST Branch does not regulate the amount of lead in the soil but refers it to the Solid Waste Branch where they use 250 ppm for residential and 500 ppm for industrial sites. In Pennsylvania, levels are set at 200 ppm for non-industrial sites and 600 ppm for industrial sites. These standards were developed to protect human health, ecological receptors and groundwater and are based exclusively on protecting human health to a blood lead level of 10 micrograms/deciliter because this is considered the most sensitive receptor (Pennsylvania Bureau of Waste Management, 1994).

METHODS

Data were gathered from two separate databases containing background lead levels. One database consisted of the closure files for waste oil UST removals at the Kentucky Department for Environmental Protection (Appendix Table 1). Background level, analytic method used to determine that level, type of sample collected (composite or grab), and date of analysis were recorded. To create a representative sample of closure files, 105 sites were selected randomly from a list of more than 5000 sites, but only 101 were used in the analysis. Some files did not have background data given. For these files without background data, alternate files were selected in the same county. No other restrictions were placed on the sites selected, even though some of the sites selected had very high lead concentrations. Of the 120 counties in Kentucky, 49% are represented in this subset of the available data.

The other database (Appendix Table 2) consisted of background files from Kentucky counties. These samples were collected by the Kentucky Department for Environmental Protection and analyzed by the Division of Environmental Services. This second database consists of samples from all Kentucky counties. Some of the files did not have results for soil lead. Counties for which results were unavailable were excluded from the data set. Thus, 52% of the counties in the state are represented. When combined, the closed UST files and the background files represent 69% of the Kentucky counties.

RESULTS

Data from the waste oil tank closure files (101 sites in 59 counties) gave a background mean of 35.98 mg/kg lead with standard deviation of 48.75. The upper 95% confidence interval was 42.90 mg/kg. The background files (62 sites in 62 counties) showed a background mean of 30.23 mg/kg lead, with standard deviation of 18.0; the upper 95% confidence interval was 34.7. Where the results were less than the detection limit, one-half of the detection limit was used in the calculation of the mean. The results are shown in Table 1 and the data included in the Appendix.

The range of values was from 0.4 to 292.2 mg/kg for the closed USTs and 3.3 to 351 mg/kg for the background files. This represents a wide range of values. It is interesting to note that in the undisturbed soils that were analyzed by Karathanasis and Seta (1993), the values ranged from <5 to 62 mg/kg.

Table 1.

	Closed UST files	Background files
number of data points	101	62
mean (mg/kg)	35.98	30.23
standard deviation	48.74	18.0
upper 95% confidence interval (mg/kg)	42.9	34.7
range	0.4 to 292.2	3.3 to 351

DISCUSSION

The lead levels found in soils from the two databases listed above, while not significantly different, may be slightly higher than those found in previous studies of contaminated sites. There are a few possible reasons for these higher values. For the data obtained from the tank closure files an upward bias may be introduced by the persons taking the samples. The levels found in the waste oil tank files were used as background, and the basis for cleanup purposes. A higher background level meant easier and less costly cleanup. It is also possible that the parent rock material for the tank site location consisted of higher levels of lead. And finally, sample sites selected to determine the background may have been chosen at locations that were not representative of the surrounding area.

Although all of the counties in the state are not included in the study, we believe that our data can be generalized to represent statewide lead values.

CONCLUSIONS

While risk assessment calculations can be made for lead, the parameters used are not well documented and acceptable risk values cannot be determined. The soil studies conducted in Kentucky indicate that for Kentucky the natural background levels of lead are near 25 mg/kg with a variance of <5 to 65 mg/kg. The data collected in this study from sites that contained waste oil tanks provided results with a wide variance (from 0.4 to 351 mg/k/) and a mean soil lead concentration of 30 to 36 mg/kg. A wide variance is also found in the standards set by other states and governments, with a range of <1 mg/kg to as high as 2000 mg/kg.

It has also been found that lead is immobile in almost all soils except under acid conditions. Under the closure procedures adopted by the UST Branch in Kentucky, up to two meters of clean soil will be placed on top of the contaminated soil so the exposure will be eliminated or greatly reduced. Computations of risk made by Westerman (1993) produced a level of 20 mg/kg for a 1 X 10^{-6} risk, which is very close to the levels of lead found in the rural soil samples (probably least contaminated) tested by Karathanasis and Seta (1993).

Because it will be difficult to determine how much lead contamination has been contributed to the soil from sources other than the waste oil tanks at most business sites (non rural), and because it is not possible to compute a reliable risk assessment value, it is proposed that the standard be set at a level of 25 mg/kg, which has been found to be the mean level found in Kentucky soils. If the measurements made in the tank pit, according to accepted sampling procedures, do not fall below this level, the background level of lead in the soil should be determined in the upgradient direction of the tank from samples collected one meter below the surface. The sampling and testing procedure described by the UST Branch should be followed. The background level can then be used as the level required for closure.

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Appendix Table 1

Removed Tanks Files

ID No.	County	Pb Conc.(mg/kgm)		Type	Method	Notes
5404001	Adair	TRM	19.6		3010/7420	
176607	Bell	TRM	11.0		6010	
9259008	Boone	TRM	9.0	Composite	6010	
5955009	Bourbon	TRM	63.34	Composite	7420	1992
2744010	Boyd	TRM	26.5		7420	1990
2921010	Boyd	TRM	33.0		7420	1991
2926010	Boyd	TRM	109.0		7420	1991
4874011	Boyle	TRM	0.4		6010	1989
7296011	Boyle	TRM	10.3		7421	1992
7510012	Bracken	TRM	27.0			1991
7271014	Breckenridge	TRM	15.6		9071	1990
0034017	Caldwell	TRM	4.9	Composite		1990
3566018	Calloway	TRM	6.8	Composite	6010	1990
3564019	Campbell	TRM	288.0		7421	1992
0313022	Carter	TRM	35.5			1991
0070024	Christian	TRM	8.6		7420	1992
8592024	Chrisitan	TRM	1.7		7421	1993
3085025	Clark	TRM	33.0		7420	1989
3085025	Clark	TRM	35.1		7420	1994
3245025	Clinton	TRM	14.0		7421	1993
6496028	Crittenden	TRM	21.9		6010	1993
5370029	Cumberland	TRM	71.8		7420	1990
4792030	Daviess	TRM	28.1		3050	1989
8923030	Daviess	TRM	32.0		7420	1991
0445034	Fayette	TRM	10.62		6010	1992
1911034	Fayette	TRM	21.1		7421	1991
0484034	Fayette	TRM	12.8		7421	1992
9056034	Fayette	TRM	830.0		6010	1989
2336034	Fayette	TRM	54.0		7421	1992
0432034*	Fayette	TRM	<10.0		6010	1991
7435034	Fayette	TRM	11.8		7421	1992
3653034	Fayette	TRM	75.0		7421	1992
7209035	Fleming	TRM	54.0		7421	1991
3564037	Franklin	TRM	17.4		7420	1990
0175037	Franklin	TRM	24.0		6010	1993
9161037	Franklin	TRM	7.8			1993
7348037*	Franklin	TRM	>9.0			1992
3035042	Graves	TRM	17.0	Composite	7420	1992
3174043	Grayson	TRM	29.5	Composite		1991

3318046	Hancock	TRM	55.0		FAA	1992
4876047	Hardin	TRM	16.2			1994
3423047	Hardin	TRM	30.0		6010	1992
499049	Harrison	TRM	8.33		7421	1990
3280050	Hart	TRM	28.4		7420	1993
1791051*	Henderson	TRM	<5.0			1991
1519054	Hopkins	TRM	36.0	Composite	7420	1991
5558055	Jackson	TRM	26.3		6010	1992
1513056	Jefferson	TRM	15.57		6010	1992
6480056	Jefferson	TRM	11.3		7420	1990
5000056	Jefferson	TRM	24.0	Composite	6010	1991
5006056	Jefferson	TRM	86.0	Composite	7421	1992
9700056	Jefferson	TRM	44.0	Composite	7421	1992
0404056	Jefferson	TRM	28.29		7420	
2472056	Jefferson	TRM	26.8		3050	1991
0697056	Jefferson	TRM	17.0			1992
9059057	Jessamine	TRM	20.2			1989
3140058	Johnson	TRM	14.5		7420	1991
9020059	Kenton	TRM	21.0		7420	1989
3565061	Knox	TRM	7.0		6010	1991
4128064	Lawrence	TRM	5.5			1993
2918067	Letcher	TRM	134.0		7421	1991
1601067	Letcher	TRM	14.3		7420	1992
7608068	Lewis	TRM	13.0			1991
7698076	Madison	TRM	21.0		7420	1992
3967076	Madison	TRM	79.6			1990
3558076	Madison	TRM	26.0			1990
0276078	Marion	TRM	15.5			1991
0134078	Marion	TRM	28.0		200.7	1992
2939079	Marshall	TRM	14.7		7420	1990
0044081	Mason	TRM	20.2		7420	1992
8820081	Mason	TRM	12.6			1991
2591073	McCracken	TRM	180.0		6010	1992
9149073	McCracken	TRM	15.8		7000	1993
4896082	Mead	TRM	16.5		7421	1993
2498082	Mead	TRM	98.5	Composite		1990
1092084	Mercer	TRM	18.9		6010	1990
6009087	Montgomery	TRM	24.3		7421	1993
3575089	Muhlenberg	TRM	5.2		7421	1993
1840090	Nelson	TRM	30.0		6010	1990
1675092	Ohio	TRM	26.0	Composite		1991
1044092	Ohio	TRM	9.2		7420	1992
2316093	Oldham	TRM	90.0			1991
6484093	Oldham	TRM	24.0		6010	1990
4294096	Pendleton	TRM	182.0		7420	1993
0112098	Pike	TRM	9.59		7421	1992
2793098	Pike	TRM	22.1		7420	1991
5516100	Pulaski	TRM	19.0		6010	1991
4956105	Scott	TRM	37.0			1992
1833105	Scott	TRM	19.0			1993
2564105	Scott	TRM	116.0		7420	1992
1233106	Shelby	TRM	86.0		6010	1992
1249106	Shelby	TRM	292.2	Composite	9071	1992

9799107	Simpson	TRM	14.6			1990
3291109	Taylor	TRM	9.23		7420	1992
4502110	Todd	TRM	25.2			1991
1880113	Uniion	TRM	10.6	Composite		1992
1622114	Warren	TRM	33.2			1991
6938114	Warren	TRM	41.7		3050	1990
9594114	Warren	TRM	24.0	Composite	7421	1991
3545114	Warren	TRM	50.1		7420	1990
4279120	Woodford	TRM	80.0		7421	1993

Summary

Average	35.98
Standard Deviation	48.75
c.i.	9.60
Number	101

The items marked with * above have been used in the calculations at half their value. The standard practice is to take 50% of the detection limit and use it in the calculation.

Appendix Table 2
Background Files

County	mg/kg lead
Adair	19.2
Anderson	47.9
Allen	17.7
Ballard	21.9
Barren	22.1
Bell	12.5
Boone	25.0
Boyd	56.8
Boyle	26.9
Bullitt	19.4
Calloway	38.2
Caldwell	9.3
Carlisle	30.6
Carroll	13.3
Carter	63.5
Casey	39.1
Christian	40.0
Clark	34.3
Crittenden	17.9
Daviess	42.8
Fayette	90.9
Floyd	24.7
Franklin	25.2
Graves	24.2
Grayson	45.5
Greenup	32.2
Hancock	15.1
Hardin	5.13
Harlan	16.0
Hart	16.6
Hopkins	17.9
Jefferson	19.0
Jessimine	29.2
Johnson	13.8
Larue	24.8
Laurel	10.7
Lawrence	15.8
Letcher	43.5
Logan	41.7
Madison	46.4
Marshall	23.6
McCracken	20.2

County	mg/kg lead
Meade	98.5
Montgomery	28.3
Morgan	27.7
Muhlenburg	29.6
Nicholas	20.2
Ohio	40.3
Oldham	33.8
Owen	42.5
Perry	67.8
Powell	21.9
Pulaski	23.9
Rowen	15.5
Simpson	28.5
Union	21.7
Warren	15.2
Washington	59.0
Wayne	26.5
Webster	16.3
Whitley	45.1
Woodford	11.9

Sum	1874.75
Count	62
Mean	30.2379
Min.	5.13
Max.	98.5
SD	18.0081
CI	4.8