

Accumulation of Lead in Soil from Waste-derived Commercial Fertilizer

Eden Mann

Abstract Non-nutritive metals from industrial waste are recycled into fertilizer as a cheap method of disposal. Soil naturally contains traces of heavy metals; however, the repetitive application of fertilizers containing these heavy metals may result in accumulation of lead to levels that may have toxic consequences for all life forms. This study estimates and compares accumulation of lead in agricultural soils from waste-derived commercial fertilizers under new California regulations by calculating the time it will take to reach USEPA standards for hazardous concentrations of lead under different worst-case scenarios and also for specific real-life crop examples. Grain, root, and vegetable crops were compared in the worst-case scenarios. Fertilizer usage was based on high-end phosphate nutrient requirements for each of the categories. Tillage depth and soil densities were specific to California soils. It was assumed the maximum allowable concentration of lead under the new California regulations was present in the fertilizer. The specific crop examples in various California counties were compared: bell pepper in Kern, onions in Riverside, and grapes in Napa. Variables such as fertilizer use and tillage depth were specific to local growing practices; soil density was specific to the location of the crop. The results show that lead poses a negligible risk of accumulating to toxic concentrations within the next 100 years under the worst-case scenarios and the specific crop scenarios. Therefore, new California standards for lead in fertilizers may be adequate; however, states without regulations may want to set standards for lead in fertilizer to prevent rapid accumulation to hazardous levels.

Introduction

A commercial fertilizer is “a substance containing one or more recognized plant nutrients that is used for its plant nutrient content or is designated for use or claimed to have value in promoting plant growth” (WSDA 2003). Primary nutrients are defined as nitrogen, phosphorus, and potassium. Secondary nutrients include calcium, magnesium, and sulfur (WSDA 2003). Micronutrients are defined as boron, manganese, chlorine, molybdenum, cobalt, sodium, copper, zinc, and iron (WSDA 2003). Recently, however, non-nutritive substances including lead, mercury, arsenic, cadmium, chromium, and dioxin have been recycled into some fertilizers as a cheaper alternative to proper disposal (USEPA 1999, Environmental Working Group 1998, The Fertilizer Institute 2000, WSDA 2001). This practice is occurring all over the world. Toxic waste from cement kiln dust, pulp mills, mining, incinerated medical supplies, spent battery acid, contaminated industrial phosphoric and sulfuric acid, and the aluminum industry is sent to fertilizer companies where it is then rolled into fertilizer pellets if it contains primary or secondary nutrients such as zinc, nitrogen, or phosphorus (USEPA 1999, Environmental Working Group 1998, The Fertilizer Institute 2000, WSDA 2001). A total of 454 companies identified as fertilizer manufacturers and farms in the Toxic Release Inventory received 271 million pounds of toxic waste containing 69 types of toxics and 6.2 million pounds of lead compounds in a five-year period (Environmental Working Group 1998). There are economic incentives for both the waste providing industries and fertilizer manufactures—it is cheap for industries to dispose of their hazardous waste in this fashion and benefits fertilizer manufacturers because they receive free or discounted nutrients (Environmental Working Group 1998).

Although the United States Environmental Protection Agency (USEPA) admits that “no specific regulations exist requiring fertilizer producers to list non-nutritive constituents on fertilizer labels, so it is difficult to quickly ascertain the levels of heavy metals (and other chemicals) in fertilizers” and “chemicals such as radionuclides and persistent organics (e.g., chlorinated dibenzodioxins/furans) are in this category,” they claim that there is no specific evidence showing that the heavy metals recycled into fertilizers pose a threat to human health (USEPA 1999).

In 2002, the California State Department of Agriculture implemented new regulations which establish maximum concentration limits of arsenic, cadmium, and lead in fertilizer that vary depending upon the concentrations of specified nutritive constituents (i.e. iron, zinc, and

manganese for mineral products and phosphate for commercial fertilizers) in the fertilizer (CDFA 2001). The higher the concentration of “nutrients,” the more “non-nutritive” metals the California State Department of Agriculture allows (CDFA 2001). This means root crops, which require more phosphate than grain crops will receive a larger quantity of arsenic, cadmium and lead.

Many studies conclude that ingestion of crops contaminated with cadmium from waste-derived fertilizers, as opposed to other methods of exposure, is the biggest risk posed to humans (CDFA 1998, The Fertilizer Institute 2000, Kuo et. al. 1999). These studies, however, neglect the ecological impacts and long-term impacts of heavy metal accumulation in soil due to fertilizer application. While cadmium may be taken up into plants, arsenic and lead remain in the soil where they may accumulate rapidly with persistent fertilizer application (CDFA 1998, USEPA 1999a). Lead exposure from dermal absorption, inhalation, or ingestion of crops and soil, can target the nervous system of humans and animals and may result in toxicity or death (ASTDR 1997). Children are more likely to directly ingest contaminated soil than adults; even low levels of lead may cause developmental problems, poor academic performance, lowered IQ, juvenile delinquency, and neurological damage (Hamel et. al. 2003). According to USEPA Administrator Carol Browner, “lead poisoning continues to be one of the most serious environmental threats to children in this country” (USEPA 2000).

New California standards for non-nutrient metals in fertilizer do not anticipate long-term health risks from exposure and environmental impacts that may become more likely as pollutant concentrations increase. The standards for non-nutrient metals in fertilizer fail to consider the health risks or environmental impacts of long-term heavy metal accumulation in the soil. The longest study of heavy metal accumulation in soil was conducted by Brobst et. al. (2003) which lasted only six years. Lead has a tendency to bind to soil particles and may be found to reside near the surface of the soil even many years after the original deposition occurred (Hamel 2003). Therefore, it is necessary to investigate possible scenarios of long-term heavy metal accumulation since adequate long-term field data is not available.

This study estimates accumulation of lead in soil to levels deemed hazardous to human health by the United States Environmental Protection Agency from repetitive fertilizer applications under different worst-case scenarios that depend on different combinations of tillage depth, soil

density, fertilizer concentration, and application rate. In addition this study also modeled three specific crop examples and recommendations for them were made based on the outcomes.

Methods

My study attempts to model a worst-case scenario of accumulation of lead in soil from repetitive use of waste-derived fertilizers using different combinations of tillage depths, soil densities, fertilizer concentrations, and application rates. The worst-case scenarios assume the maximum allowable concentration of lead in fertilizer is present in the fertilizer. The scenarios were compared using the time it will take for lead to accumulate to hazardous concentrations in agricultural soils from continuous application of waste-derived commercial fertilizers. The United States Environmental Protection Agency determined levels of lead hazardous to the health of humans to be 400 parts per million (ppm) in residential play areas, 1200ppm in residential non-play areas, and 2000ppm for non-residential areas (USEPA 2001). At these concentrations it is recommended that remedial action be taken either by removing contaminated soil or by covering the contaminated soil with cement (USEPA 2000). Preliminary data showed the time for lead accumulation to reach concentrations of 1200ppm or 2000ppm was on the order of hundreds to thousands of years. Although farms are not residential play areas this study's final data focused on the time it will take for lead accumulation to reach the hazardous concentration of 400ppm (USEPA 2001). Nevertheless, lead concentrations of 400ppm in soil pose a risk to certain members of the human population and therefore the concentration is still significant whether it is found in a residential play area or on an agricultural farm. The Rutgers Cooperative Extension of the State University of New Jersey recommends vegetables should not be grown in soils containing lead concentrations above 400ppm (Hamel et. al. 2003).

In order to calculate the time it will take for lead to accumulate to the lowest designated hazardous level in soil of 400ppm under the worst case scenarios, I found the ranges for fertilizer use, soil density, and tillage depth in California using data from documents made available to the public such as various government agency reports.

Percent phosphate in agricultural fertilizers can vary from 2-70% (USEPA 1999a). The amount of lead added to the soil is the same if a higher percentage of phosphate is used rather than a lower percentage because less fertilizer is needed. Since the amount of lead in fertilizer is a function of the amount of phosphate in fertilizer, the amount of lead applied to the soil depends on the phosphate requirements of the fertilized crop. Therefore, the total amount of fertilizer

used in the scenarios was determined by nutrient need. The yearly, high-end phosphate need for a root crop, for example, is 25 grams (g) of phosphate per m^2 (USEPA 1999a). This high-end number represents the 95% upper confidence level of the mean for root crop nutrient needs assuming a normal distribution (USEPA 1999a). If the nutrient need is 25g of phosphate per m^2 and a 70% phosphate fertilizer is used then 35.7g of fertilizer is needed per m^2 , the maximum allowable lead would be 1400ppm because 20ppm of lead are allowed for each percent available phosphate (70% x 100 x 20ppm). This means approximately 50mg of lead would be applied to the soil per m^2 (0.0357kg x 1400ppm). The nutrient requirements used in this study were 16.5g of phosphate per m^2 for grain crops, 22g of phosphate per m^2 for vegetable crops, and 25g of phosphate per m^2 for root crops (USEPA 1999a).

In California, approximately 96% of bulk soil densities ranged from $1.1\text{g}/\text{cm}^3$ in clay-like soils to $1.6\text{g}/\text{cm}^3$ in sandy soils (Brady and Weil 2003, CDFA 1998). Tillage depths in agricultural practices range from 0cm (no-till) to about 30cm under deep-till conditions (Brady and Weil 2003).

This study goes further to calculate and compare accumulation times for real life California scenarios using actual data for grapes in Napa County, onions in Riverside County and bell pepper in Kern County. These particular crops were chosen because of their high economic value for California and also because data was readily available. California is the number one producer of wine, table, and raisin grapes and is responsible for 90% of all grape production in the United States (California Agricultural Statistics Service. 1999). California is also the number one producer of onions, growing approximately 26% of onions in the United States (USDA 1999). The state is also the number two producer of bell peppers after Florida and in 1998 the crop was valued at \$133 million (USDA 2000). Farm location coordinates were identified and compared to the specific county's soil survey maps to reveal the dominant soil type nearest to the location the crops are grown. The soil density for the specific soil type was then noted. The soil densities used were $1.45\text{g}/\text{cm}^3$ for Imperial Clay soil in Riverside County, $1.21\text{g}/\text{cm}^3$ for Aiken Loam soil in Napa County, and $1.30\text{g}/\text{cm}^3$ for Kettleman Clay soil in Kern County (WSDA 2001a, WSDA 2001b, WSDA 2001c). Common application rates used in this study for the specific crops in California are as follows: bell pepper 10g of phosphate per m^2 , grapes 2.2g of phosphate per m^2 , onions (dry bulb) 28g of phosphate per m^2 (Lorenz & Maynard 1990, UCCE 1990, UCDAAS 1973). The tillage depths used are as follows: 10cm for onions because they are shallow rooted,

20cm for grapes, and 35cm for bell peppers because they have deep, extensive root systems (Kliwer et. al. 1975, UCDAAS 1996, UCDAAS 1999). The actual background concentration of lead for these particular soils was found in a publication by the Kearney Foundation of Soil Science: 37ppm in Imperial Clay, 34.3ppm in Aiken Loam, and 14.6ppm in Kettleman Clay (Bradford et. al. 1996).

With the data for the worst-case scenarios I first calculated the maximum allowable concentration of lead under the new California regulations that would be permitted in the amount of fertilizer per unit area applied. The maximum allowable concentration of lead can be found by multiplying the percent phosphate by 20ppm of lead (CDFA 2001). With this concentration of lead per unit area and the tillage depth, I calculated the lead added to the volume of soil. Using the bulk density of the soil I found the concentration of lead applied per mass unit of soil. This gave me the accumulation rate of lead into soil in units of milligrams of lead per kilogram of soil per year. Finally, I found the time it will take for lead to accumulate to the chosen hazardous level of 400ppm by dividing the difference between 400ppm and the average background concentration of lead in the soil, 17ppm, by the yearly application rate (Davies 1995).

Equation 1: Application rate of lead (Pb) into soil (mg/kg*y), where F represents waste-derived fertilizer=

$$\frac{[\text{conc. of phosphate in F (\%)}] \times [20\text{ppm(mg/kg) of Pb}] \times [\text{mass of F applied per year/unit area (kg/m}^2\text{y)}]}{100 \times [\text{tillage depth (m)}] \times [\text{soil density (kg/m}^3\text{)}]}$$

Equation 2: Time to reach critical level (y)=

$$\frac{[\text{critical concentration of Pb(mg/kg)} - \text{background level of Pb (mg/kg)}]}{[\text{application rate (mg/kg*y)}]}$$

The calculation for the real life scenarios was similar except the actual values for phosphate input, soil density, background level of lead in soil, and tillage depth were used. The amount of lead in fertilizer used in this portion of the calculations was the maximum allowable because preliminary data showed even with the maximum allowable lead there was little risk of accumulation to toxic levels

The median background level of lead in soil used in my worst-case scenario calculations was 17mg/kg (Davis 1995). I assumed the same amount of fertilizer with the same concentration of phosphate and lead is applied only once each year. I assumed that there will be no leaching of

lead. The California Department of Food and Agriculture explains there is minimal leaching in California agriculture soils because of the arid climate (CDFA 1998). Degradation, volatilization, and erosion were also determined to be negligible as loss pathways for lead (CDFA 1998). The CDFA states lead exists in a relatively immobile form because of the pH of soil in California therefore it is believed that accumulation will occur (CDFA 1998). Lead remains in the surface soil layers after application in an insoluble or stable form. (Davies 1995). Lead has a long residence time in soil compared to most other pollutants—it accumulates in soil because of its low solubility and relative freedom from microbial degradation (Davies 1995). Also, I assumed there is no plant uptake of lead and that the only input of lead is from the waste-derived fertilizer (WSDA 2001). I neglected other inputs of lead because lead was removed from gasoline in 1986, so there is currently minimal input from the atmosphere.

Results

Tables 1, 2, and 3, represent worst-case scenarios which show the time it will take for soils growing grain, vegetable, and root crops to reach the lowest established hazardous concentration of lead (400 ppm) under various tillage depths and soil densities.

Tillage Depth (m)	0.3	3830	4178	4526	4875	5223	5571
	0.25	3192	3482	3772	4062	4352	4642
	0.2	2553	2785	3018	3250	3482	3714
	0.15	1915	2089	2263	2437	2611	2785
	0.1	1277	1393	1509	1625	1741	1857
	0.05	638	696	754	812	870	928
	0.01	128	139	151	162	174	186
		1.1	1.2	1.3	1.4	1.5	1.6
		Soil Density (g/cm3)					

Table 1: Time for the lead in soil with grain crops ($P_2O_5=16.5g/m^2y$) to reach the 400ppm under various soil

densities and tillage depths

0-500yrs. >500yrs.

Tillage Depth (m)	0.3	2873	3134	3395	3656	3917	4178
	0.25	2394	2611	2829	3047	3264	3482
	0.2	1915	2089	2263	2437	2611	2785
	0.15	1436	1567	1697	1828	1959	2089
	0.1	958	1045	1132	1219	1306	1393
	0.05	479	522	566	609	653	696
	0.01	96	104	113	122	131	139
		1.1	1.2	1.3	1.4	1.5	1.6

Soil Density (g/cm³)

Table 2: Time for the lead in soil with vegetable crops ($P_2O_5=22g/m^2y$) to reach the 400ppm under various soil densities and tillage depths

0-500yrs. >500yrs.

Tillage Depth (m)	0.3	2553	2785	3018	3250	3482	3714
	0.25	2128	2321	2515	2708	2902	3095
	0.2	1702	1857	2012	2166	2321	2476
	0.15	1277	1393	1509	1625	1741	1857
	0.1	851	928	1006	1083	1161	1238
	0.05	426	464	503	542	580	619
	0.01	85	93	101	108	116	124
		1.1	1.2	1.3	1.4	1.5	1.6
		Soil Density (g/cm ³)					

Table 3: Time for the lead in soil with root crops ($P_2O_5=25g/m^2y$) to reach the 400ppm under various soil densities and tillage depths

0-500yrs. >500yrs.

	Yearly P_2O_5 application (g/m ² *y)	Soil Type	Soil Density (g/cm ³)	Tillage Depth (cm)	Current Level of Pb in Soil (ppm)	Time for Pb in soil to reach 400ppm in years (y)
Onions (dry bulb) in Riverside County	28	Imperial Clay	1.21	10	37	784
Grapes in Napa County	2.3	Aiken Loam	1.45	20	34.3	23609
Bell Peppers in Kern County	10	Kettleman Clay	1.30	35	14.6	8768

Table 4: Summary of variables and findings for the specific crop examples.

Discussion

The results for the worst case scenarios (Tables 1-3) show that possible accumulation could occur in as little as 85 years under a shallow tillage scenario. The results from the specific crop scenarios (Table 4) show that the time it will take for lead to reach the hazardous level of 400ppm is long and therefore the risk to humans is negligible (i.e. the results reveal it will take approximately 784 years to reach 400ppm). Fertilizer applications, tillage depths, and soil densities used in the specific crop scenarios are much more representative of California conditions and agricultural practices than the worst-case scenarios. These figures were also more

representative because the background concentration of lead in soil for the particular region was identified and used in the calculations.

Tillage depth seems to affect lead accumulation more than soil density. Soils growing crops requiring a high level of nutrients, such as root crops, under low till conditions will have more rapid accumulation than if they are grown under high till conditions. Tillage depth affects erosion by loosening soil, which may cause it to blow away or be washed away by rainwater (Botkin and Keller 2000). If the top layers of nutrient rich soil are blown or washed away, more fertilizers may be necessary to compensate for the loss of nutrients (Botkin and Keller 2000). Erosion, although it is a legitimate threat caused directly by tillage, was neglected in this calculation; however, if it were included as an output, the time to reach the lowest established hazardous level would be less than calculated here. Also, it was assumed the soil was tilled every year. In crops such as grapes and citrus fruits that are not planted annually, soils tillage may not occur every year which may also mean the results may have underestimated the risk of accumulation. Overall the model shows low tilled soils will accumulate lead more rapidly than deep tilled soils.

The average background level of lead in soil used in the worst case scenarios was 17ppm (Davies 1995). The actual concentration of lead found in California agricultural soils presently may be much higher or lower than the average used. If there were no regulations of lead in fertilizer prior to the year 2002 it can be assumed that some accumulation of lead from commercial fertilizer use may already exist in agricultural soil and thus exact present conditions were neglected in the calculations which may have resulted in an underestimation of the risks of lead from fertilizer input. The Kearney Foundation of Soil Science in a publication regarding background concentrations of heavy metals in California soils reveal lead concentrations in California soils range from 12ppm to 97ppm (Bradford et. al. 1996). This means the time for lead to accumulate to 400ppm for a root crop grown in soil with a density of 1.5g/cm^3 under no till conditions could range from 93 years to 117 years, instead of 116 years as calculated with the averaged background level of lead of 17ppm.

There are many limitations to this study. This model provides an easy way to estimate the risk of lead accumulation in soil. It may not be the best way to estimate the risk, but it is one of the safest ways as opposed to something like a long-term field study. Future studies should model the accumulation of other hazardous substances such as arsenic, cadmium, chromium,

nickel, mercury, and selenium. While this study focused on estimating the risk of lead accumulation in soil for humans, future studies should also investigate the risk to nematodes, insects, and wildlife. Future models may want to incorporate other variables into this model like fertilizer/contaminant uptake into plants, pH of the soil, runoff, soil erosion, topography, dust emission, and metal mobility. Designing a computer program to model accumulation would be the best way to incorporate these variables because having outputs as well as inputs requires differential calculus equations.

While this study reveals the new California standards for lead in fertilizers are adequate, the study does not address the other non-nutritive substances currently found in waste-derived fertilizer. It is also important to note that only California, Washington, and Texas have set regulations for metal concentrations in commercial fertilizers. In 1998, a study by the California State Department of Agriculture of random unidentified fertilizer samples revealed high concentrations of lead (CDFA). A sample containing 14.4-23.4-0, or 23.4% phosphate, for example, showed a concentration of lead of 4650ppm. As the current regulations are 20ppm for each percent of available phosphate, the previously referenced pre-regulation datum would be equivalent to 200ppm for each percent phosphate. If this particular sample was used to fertilize a root crop in soil with a density of 1.45g/cm^3 under no till conditions, it would only take 11 years to reach the hazardous concentration of 400ppm. This means without proper regulation, soil in other states may be at a serious risk for accumulating hazardous levels of lead in a short amount of time.

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