RESUSPENSION OF CONTAMINATED SOIL AS A SOURCE OF AIRBORNE LEAD

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Abstract

Although airborne Pb levels have declined significantly in California over the past two decades, levels above 65 μ g/m³ are still occasionally observed. Suspension of contaminated soils by wind or mechanical abrasion is suspected as a possible source. To examine the potential significance of this mechanism, surface soil samples with a range of bulk soil Pb concentrations were obtained near 8 industrial facilities and along roadsides and were resuspended in a specially designed laboratory chamber. The concentration of Pb was measured in the bulk soil, soil size fractions and in particles less than 10 μ m in aerodynamic diameter (PM₁₀) generated during resuspension of soils and fractions. Average yields of PM₁₀ from dry soils ranged from 0.169 to 0.869 mg PM₁₀/g soil. The resulting PM₁₀ had average Pb concentrations as high as 2283 mg/kg for samples from a secondary Pb smelter. Concentrations of Pb in the PM₁₀ were as much as 8 times higher than concentrations in the bulk soil. Total potential emissions of PM₁₀ bound Pb from soil were between 0.012 and 1.2 mg/kg. The results suggest that contaminated soil may be a locally important source of airborne Pb under a narrowly defined set of scenarios.

Executive Summary

BACKGROUND

Although impressive progress has been made in reducing airborne lead levels, certain Air Resources Board monitoring sites occasionally exhibit lead concentrations above 65 ng/m^3 in the total suspended particulate fraction. These elevated levels are infrequent but troublesome, since lead has been declared a toxic air contaminant. One possible source of the airborne particulate-bound lead is from resuspended contaminated soils. Lead concentrations near high traffic density highways, lead processing facilities, and older painted homes may exceed 1000 mg/kg. For comparison, one Air Resources Board monitoring site with relatively high airborne lead concentrations between 1990-1996 had average solid-phase concentration of lead in the fraction of particulate matter less than 10 μ m aerodynamic diameter (PM₁₀) of 800 mg/kg. This study investigates the linkage between airborne PM_{10} lead and lead concentrations in shallow soils near roadways and lead-using commercial and industrial facilities. Specific project objectives included (i) identify facilities and locations that are likely to have elevated lead concentrations in soils, (ii) determine some representative concentration profiles of lead in bulk surface soils near the identified facilities, (iii) estimate the potential for leadbearing soils to be resuspended as PM_{10} and measure the lead content of the resuspended material, (iv) examine relationships between lead and various co-contaminants at particular classes of facilities as a method of source identification, (v) predict the exposure to airborne lead for a variety of scenarios including differences in soil type, lead source, and site conditions, and (vi) evaluate the efficiency of possible control methodologies.

METHODS

A major goal of the project was to sample a wide range of facility types to determine the concentration of lead and some co-contaminants in soils on or near the site. A list of facilities that might have elevated lead levels in soil was generated by combining an Air Resources Board list of 54 current lead emitting facilities with a Department of Toxic Substances Control list of 382 facilities that were known to have lead contamination problems. Letters requesting site access so that the project team could collect soil samples were sent to all of the 436 facilities on the combined list for which addresses could be obtained. Responses permitting site access were obtained from only 5 facilities. To get a more complete sample of facility types, it was decided to collect samples at the property boundaries of some additional facilities. A list of 19 facilities, including the five that had granted site access, was generated from the original list. Samples were successfully collected from eight facilities and from three highway sites.

Soil samples were collected from the top 10 cm using a coring device. At least two samples were taken at each facility and samples were collected both upwind and downwind of the presumed emission source based on the prevailing wind direction at most facilities. Samples were taken at random distances from the emission source subject to accessibility constraints (e.g., pavement and buildings). Soil samples were oven-dried and homogenized prior to analysis. Sub-samples of soils collected downwind of the source at each site were dry sieved into six particle size ranges. Lead, and potential cocontaminants Nickel, Copper, Zinc and Arsenic were analyzed in bulk samples and their size fractions using x-ray fluorescence (XRF). Soil samples and their fractions were resuspended using a specially designed system and the resulting PM_{10} was collected on Teflon filters. The concentration of lead and other elements on the PM_{10} was determined using XRF.

RESULTS

For soil resuspension to be a significant contributor to locally elevated airborne lead levels, the soil must contain elevated levels of lead, the soil must yield appreciable amounts of fine particulate matter, and lead concentrations in the particulate matter must be high. Of the facilities tested only the lead smelter had average soil lead concentrations significantly above the 12.4-97.1 mg/kg range reported previously for uncontaminated California soils. Yields of fine particulate matter from the soils collected ranged from 0.20 to 0.87 mg PM_{10} /g soil. The concentration of lead on this particulate matter was higher than the corresponding bulk soil concentration in every case tested, with the most significant enrichment, by a factor of 8.6, observed for the glass manufacturing facility. Combining the PM_{10} yield and its lead concentration resulted in production of between 0.06 and 1.23 µg of particle bound lead per gram of bulk soil. As an example of a worstcase scenario, if soil at the lead smelter site were tilled to a depth of 1 cm and this operation efficiently stripped away the fine particulate matter, the operation would produce approximately 80 g of PM_{10} bound lead per acre. Under at least some circumstances, therefore, it appears that PM₁₀ derived from lead contaminated soils could be a significant transient local source of atmospheric lead.

CONCLUSIONS

Lead concentrations in PM_{10} derived from soil samples were on average 3.6 times higher than the corresponding bulk soil concentrations. The most contaminated sample from the lead smelter site produced PM_{10} lead concentrations in excess of 5600 µg Pb/g PM_{10} . The median concentration of lead in PM_{10} from all sites was only 152 µg Pb/g PM_{10} , suggesting that elevated lead levels are a site specific, rather than generic to all lead using or emitting industrial sites.

Although lead was correlated with different co-contaminants at different facilities, these correlations do not appear to be immediately useful for distinguishing among different industrial sources of PM_{10} .

Contaminated soils may be a significant local source of airborne lead contamination under "worst-case" conditions that include very dry fully exposed soils subject to mechanical agitation (e.g., by tilling). Under these conditions, the top 1 cm of the average lead smelter soil has the potential to release 80 g Pb/acre in the form of PM_{10} . If the area were moist, well-vegetated, or paved emissions would be significantly lower.

This report demonstrates the potential, under a narrow set of conditions, for contaminated soil resuspension to produce locally elevated airborne lead levels. Proof of this link is not offered and would need to be provided by future research monitoring resuspension and airborne lead concentrations simultaneously at a facility with exposed contaminated soil.

1. BACKGROUND

1.1. Statement of the Problem

The elimination of lead from gasoline, and the control of point sources of lead emissions from commercial and industrial facilities have lowered ambient lead concentrations in California by an order of magnitude over the last two decades. Despite this impressive progress, certain California Air Resources Board monitoring sites occasionally exhibit lead concentrations above 65 ng/m³ in the total suspended particulate fraction. These elevated levels are infrequent but troublesome, since lead has been declared a toxic air contaminant. Additional special monitoring sites have been established near sites that have historical lead contamination problems; these sites occasionally record even higher concentrations.

One possible source of the airborne particulate-bound lead is from resuspended contaminated soils. Lead can be retained in soils for many decades without significant migration (Haneberg *et al.* 1993) and bulk soil lead concentrations near high traffic density highways, lead processing facilities, and older painted homes may exceed 1000 mg/kg (Haneberg *et al.* 1993; Teichman *et al.* 1993; Gulson *et al.* 1995). Lead concentrations in fine soil fractions (<100 μ m diameter), which are most likely to become airborne dust, may be 3-8 times higher than in the bulk soil (Gulson *et al.* 1995). Review of data from one CARB monitoring site with relatively high airborne lead concentrations between 1990-1996 indicated that the solid-phase concentration of lead in the fraction of particulate matter less than 10 μ m aerodynamic diameter (PM₁₀) averaged 800 mg/kg with a standard deviation of 440 mg/kg, values that could easily be produced by resuspension of contaminated soils. Designing effective control strategies for airborne lead and assessing the potential for population exposures obviously requires an understanding of the sources of the lead, which is currently unknown.

1.2. Project Objectives

The study investigates the linkage between airborne PM_{10} lead and lead concentrations in shallow soils near roadways and lead-using commercial and industrial facilities. Specific project objectives include:

- Identify facilities and locations that are likely to have elevated lead concentrations in soils,
- Determine some representative concentration profiles of lead in bulk surface soils near the identified facilities,
- Estimate the potential for lead-bearing soils to be resuspended as PM_{10} and measure the lead content of the resuspended material,
- Examine relationships between lead and various co-contaminants at particular classes of facilities as a method of source identification,
- Predict the exposure to airborne lead for a variety of scenarios including differences in soil type, lead source, and site conditions,
- Evaluate the efficiency of possible control methodologies.

1.3. Sources, Distribution and Chemical Reactions of Lead in Soils

1.3.1. Natural lead content in soils

Lead content in the average crustal rock is approximately 16 mg/kg. There is a tendency for Pb concentration to rise with increasing silica content i.e, from ultrabasic to acid igneous rocks. For example, mean values of Pb content were 1.9 mg Pb/kg for gabbro, 8.3 mg Pb/kg for andesite and 22.7 mg Pb/kg for granite (Nriagu 1978). Estimates of Pb for uncontaminated soils ranged from a mean of 17 mg Pb/kg (Nriagu 1978) to 29 mg/kg (Ure and Berrow 1982). A higher mean was reported for organic samples (30 mg Pb/kg) than mineral samples (13 mg Pb/kg). A comprehensive survey of 50 benchmark soils throughout the state of California reported normal background concentrations for Pb in the range of 12.4-97.1 mg/kg with a mean of 21.7 mg/kg (Bradford *et al.* 1996). In these soils, Pb content was significantly correlated with the content of Zr (0.42), Rb (0.57), Cs (0.57), Sb (0.38), La (0.41), Zn (0.50), Tl (0.40), Ga (0.36), As (0.37), Th (0.44), Na (0.63) and Ce (0.42).

1.3.2. Anthropogenic sources of lead in soils

Urban soil lead levels are often significantly higher than natural background levels. For example, one report found up to 540 mg Pb/kg soil although most values were in the range of 70-150 mg/kg (Fleming and Parle 1977). A list of major lead emitting industrial activities and the amount of lead they emit per unit of production is provided in Table 1.

Facility	Typical Lead Emission
Primary Lead Smelting	5-150 g/ton
Secondary Lead Smelting	6-5200 g/ton
Portland Cement Manufacturing	0.03-0.05 g/ton
Lead-Acid Battery Production	0.2-171 g/1000 battery
Lead Oxides Manufacturing	65-7000 g/ton
Ceramic Ware Manufacturing	1500 g/ton
Lead Zirconate Titanate (PZT) Man.	23 g/ton
Municipal Waste Combustion	0.5-100 g/ton
Drum and Barrel Reclamation	1.6 g/barrel
Open Burning Scrap Tires	0.1-0.3 g/ton
Crematories	0.03 g/body
Glass Manufacturing	0.2 g/ton
Batch-mix Asphalt-mix Plants	$9*10^{-4}$ g/ton

Table 1. Lead Emission Factors for Industrial Facilities

1.3.2.1. Vehicle exhaust

Early reports documented that soil and vegetation samples collected near roads contain unusually high Pb contents (Warren and Delavault 1960). Subsequently, Pb contaminated grass was found within 152 m downwind of roads in Denver, Colorado and concentrations declined moving away from the road edge in an exponential manner (Cannon and Bowles 1962). Later reports confirmed this important source of Pb for soil

and the exponential decay in concentration with distance from the roadside (Singer and Hanson 1969; Lagerwerff and Specht 1970; Davies and Holmes 1972). The literature suggests that there is a zone of about 15 m wide on either side of most roads in which the concentration of Pb exceeds local background levels and that contamination of the roadside environment is a worldwide consequence of the use of leaded gasoline.

1.3.2.2. Contamination from smelting

Metallurgical industries can contribute to soil pollution in several ways: (i) by the emission of aerosols and dusts which are transported in air and eventually deposited on soils or vegetation; (ii) by liquid effluents which may pollute soils at times of flooding; (iii) by the creation of waste dumps in which metals become corroded and leached into the underlying soils. In surveys around smelters, maximum Pb concentrations occurred close to the stack. There is a rapid, usually exponential, decline in concentrations with distance from the stack (Ragaini *et al.* 1977). It is difficult to be categorical about the soil Pb levels likely to be encountered around metal smelters. However, using the data reported above it is concluded that within 3 km of a well-established smelter, soils may contain lead levels between 1500 to 2200 mg/kg but that 95% of the samples contain <355 mg Pb/kg. Isopleth plots of the soil Pb content showed a strong association between concentrations >355 mg Pb/kg soil and old mine workings, dressing floors and railway ore loading places i.e., enhancement of 15 times the background level. Accumulations of Pb may also be expected in the soils around secondary smelters and other Pb using industries.

1.3.2.3. Atmospheric deposition

Lead aerosols emitted to the atmosphere from vehicle exhausts or industrial discharges can be carried over long distances (Nriagu and Pacyna 1988; Kral *et al.* 1992). For example, Pb contents of surface soils in Norway decreased from an average of <120 mg Pb/kg in the south of the country to <10 mg Pb/kg in the far north (Steinnes 1984). Deposition of Pb aerosols in a remote sub-alpine ecosystem in Yosemite National Park ranged from 92-270 pg Pb/cm²/day (mean 158) over a two year study period (Elias and Davidson 1980). These rates were around two orders of magnitude below those found in urban environments. Atmospheric deposition of Pb in a forest in eastern Tennessee within 20 km of three coal-fired power plants ranged from 3 to 15 μ g/m²/day (Lindberg and Harris 1981). Lindberg and Harris (1981) also listed information on total annual atmospheric deposition of Pb measured by other workers at remote, rural and industrial locations in different parts of the world. Total atmospheric deposition of Pb ranged from 3.1 to 31 mg/m²/yr in remote and rural locations and from 27 to 140 mg/m²/yr in suburban and industrial areas.

1.3.2.4. Other sources

Other significant sources of lead pollution of soils can be the manufacture, use and/or disposal of batteries, tires, pigments and paints, rubber, plastics, printing and graphics, insecticides, sewage sludge, fuel additives and lubricants.

1.3.3. Chemical behavior of lead in soil

Lead appears to accumulate naturally in surface horizons (7-10 cm) of soils (Smith 1976). Pb showed the greatest proportional accumulation in surface horizons of soils (podzolics and grey-brown podzols) in eastern Canada (Wright *et al.* 1955). Other

authors have also reported Pb accumulations in surface horizons of uncontaminated soils (Archer 1963; Merry *et al.* 1983). A topsoil "enhancement factor" has been calculated as the ratio of Pb concentration in topsoil (0-15 cm) to that in subsoil (Colbourn and Thornton 1978). In normal agricultural areas of the UK, the enhancement factor ranged from 1.2 to 2.0, whereas in locations affected by mining and smelting operations, values between 4 and 20 were common. The main compartments for Pb in soil are in the soil solution, the adsorption surfaces of the clay-humus exchange complex, precipitated forms, secondary Fe and Mn oxides and alkaline earth carbonates, the soil humus and silicate lattices.

The solubility of Pb in non-calcareous soils was found to be regulated by $Pb(OH)_2$, $Pb_3(PO_2)_2$, $Pb_5(PO_4)_2OH$ and in calcareous soils by $PbCO_3$ (Santillan-Medrano and Jurinak 1975). Several studies have examined the chemical forms of Pb in roadside soils. More than 75% of the Pb in samples from two areas was associated with soil density >3.32 g/ml, and PbSO₄ was the primary compound present (Olson and Skogerboe 1975). However, other authors found that PbSO₄ was not of any significance in roadside soils; instead, Pb was associated with Fe-Mn oxides and organic phases and, to a lesser extent, with carbonates.

Fractionation procedures have also been used to infer the chemical forms of soil Pb. It took 24-48 hours for Pb added to soils as $PbNO_3$ to become distributed among the soil compartments, and in the range 20-40 °C temperature did not affect the equilibration (Zimdahl and Skogerboe 1977). From a statistical study analysis of this data pH and CEC were concluded to be the main soil properties involved in the immobilization of Pb and that soil organic matter was more important in this process than precipitation as the carbonate or sorption by hydrous oxides. In soils from near roads < 0.7 % of the Pb was in readily exchangeable forms and most of the Pb was associated with the organic and residual fractions or specifically bonded to inorganic sites while the amount occluded in iron oxides was low (Garcia-Miragaya 1984). Pb concentrations tended to increase with a decrease in particle size for profile samples in Scotland (Berrow and Mitchell 1991). Lead released by weathering appeared to be largely adsorbed by silt and clay fractions. Lead concentrations were 3-8 times higher in the finer fractions of the bulk soil than in the average bulk soil (Gulson et al. 1995). Soil humus, especially high-molecular weight humic acids, was found to contribute most to the immobilization of Pb via coordinated binding by free electron pairs.

2. METHODS

2.1. Site selection

A major goal of the project was to sample a wide range of facility types to determine the concentration of lead and some co-contaminants in soils on or near the site. Such materials were viewed as possible precursors to lead in airborne particulates. A list of facilities that might have elevated lead levels in soil was generated by combining an Air Resources Board list of 54 current lead emitting facilities with a Department of Toxic Substances Control list of 382 facilities that were known to have lead contamination problems. These facilities were classified by industry and the prevalence of each type of industry on the combined list was calculated (Table 2). It is immediately clear from the table that "obvious" sources of lead such as primary metal production represent only a small fraction, albeit a potentially serious one, of the possible sources of airborne lead.

facilities ultimately sampled							
Facility Type	Frequency on	Number	Number				
	the list, %	(frequency)	(frequency)				
		among selected	among				
		facilities	sampled				
			facilities				
Electric, gas, sanitary services	14.1	2 (11.1%)	1 (12.5%)				
Primary metal products	11.3	1 (5.6%)	1 (12.5%)				
Fabricated metal products	9.2	1 (5.6%)	0				
Chemicals and allied products	9.2	0	0				
Electronic and other electric equipment	5.8	2 (11.1%)	0				
Nonclassifiable establishments	5.0	0	0				
Auto repair, services and parking	4.5	0	0				
Petroleum and coal products	4.2	0	0				
Wholesale trade-durable goods	3.7	0	0				
Railroad transportation	3.4	0	0				
Trucking and warehousing	2.4	0	0				
Transportation equipment	2.4	1 (5.6%)	0				
National security/international affairs	2.4	3 (16.7%)	1 (12.5%)				
Lumber and wood products	2.1	1 (5.6%)	1 (12.5%)				
Auto dealer and service stations	1.8	0	0				
Stone, clay and glass products	1.6	4 (22.2%)	2 (25.0%)				
Agricultural production-crops	1.3	0	0				
Food and kindred products	1.0	0	0				
Metal mining	0.8	2 (11.1%)	2 (25.0%)				
Miscellaneous	14.1	1 (5.6%)	0				

Table 2. Types of lead emitting facilities and their frequency in the combined AirResources Board and Department of Toxic Substances Control lists and among
facilities ultimately sampled

Letters requesting site access so that the project team could collect soil samples were sent to all of the 436 facilities on the combined list for which addresses could be obtained. Many of these letters were returned because of insufficient or incorrect addresses particularly for the DTSC list, which included many closed businesses and industries. Responses permitting site access were obtained from only 5 facilities. To get a more complete sample of facility types, it was decided to collect samples at the property boundaries of some additional facilities that had not granted permission to sample on-site. A list of 19 facilities, including the five that had granted site access, was generated by considering both the prevalence of a facility type (Table 2), the potential severity of the contamination around the facility as judged by emission factors (Table 1), and the desire to include both northern and southern California facilities. All but one of the selected facilities were visited by the project team during the period July 15-22, 1999. The last facility was not visited because of time limitations. The distribution of the selected facilities within each industrial category is provided in Table 2. Samples were successfully collected from eight of the facilities. Samples were not collected from the other facilities because no suitable and accessible sampling location was identified. The facilities ultimately sampled do not include many of the industries listed in Table 2, but do include representative facilities from 6 of the 20 classifications, and several of these are expected to be major lead emitters based on the data in Table 1.

Soils along highway rights-of-way are also known to be lead impacted, particularly for highways that were heavily traveled during the period when leaded gasoline was common (Lagerwerff and Specht 1970). Splits of three soil samples collected by the California Department of Transportation for a separate study were obtained and included in the study.

2.2. Field sampling program and sample processing

A summary of the soil samples collected during the field sampling program is provided in Table 3. At each site the source of contamination, typically a stack, contributing to possible lead contamination was identified. Soil samples were taken randomly at varying distances from the source of the emission. Distances from the emission source were measured with a range finder. One goal of the sampling was to collect samples both upwind and downwind of the source based on prevailing wind directions and in most cases this was accomplished. When it was not, it was because of problems that prevented taking a sample such as failure to gain access to a portion of a site or the presence of payement. A minimum of two samples was collected at each industrial site to a depth of 10-cm using a 5-cm diameter intact soil corer containing plastic sleeves. The ends of each sleeve were closed with plastic caps just after sampling. Sleeves placed in plastic bags and kept in coolers on ice until being returned to the laboratory. At one site (Borax Processing) soil samples from the plastic sleeve were emptied into a labeled plastic bag because the soil sample was not cohesive. All samples were returned to the laboratory and the top 5 cm of each intact sample was removed from the sleeve and dried overnight at 105°C. Samples from the Borax facility were homogenized and approximately 120 g of each sample were dried. Dried samples were then homogenized and split into two sub-samples for analyses of lead and co-contaminant concentrations in the bulk soil and selected size fractions, and PM₁₀ formation potential in bulk samples and size fractions.

Site #	Type of Site	Number of	Wind	Emission
		Samples	Direction	Source
			Sampled	Distance (m)
S-1	Waste Treatment	5	Upwind	7-182
			Downwind	
S-2	Glass Manufacturing	4	Upwind	64-71
			Downwind	
S-3	Saw Mill	2	Upwind	44-63
S-4	Perlite Mining	4	Upwind	98-105
			Downwind	
S-5	Borax	6	Upwind	60-73
	Processing		Downwind	
S-6	Cement	8	Upwind	58-69
	Manufacturing		Downwind	
S-7	Lead Smelter	9	Upwind	123-256
S-8	Sand Blasting	12	Upwind	1-35
			Downwind	
RS-27	Roadside	1	Unknown	Not
				Measured
RS-50	Roadside	1	Unknown	Not
				Measured
RS-103	Roadside	1	Unknown	Not
				Measured

Table 3. Site type and sample information

Approximately 5 grams of each bulk soil sample were powdered to be analyzed for lead and co-contaminants using x-ray fluorescence (XRF). Soil samples taken in the downwind of the source at each site were dry sieved into six particle size ranges (A>300 μ m, 300>B>150 μ m, 150>C>75 μ m, 75>D>45 μ m, 45>E>38 μ m and 38 μ m>F). The initial soil weight and the weight of each fraction were measured and the particle size distribution was determined. Approximately 4 grams of fraction A of each sample was powdered for XRF analysis; similar preparation of fractions B, C, D, E, and F was not necessary because of their small particle sizes. An unpowdered sample of each fraction A was reserved for PM₁₀ formation potential measurement. All prepared samples and fractions were placed in sealed plastic bags and stored in a cool, dry place until analysis.

2.3. Analysis of lead and co-contaminants in bulk soils and fractions

Lead (Pb), and potential co-contaminants Nickel (Ni), Copper (Cu), Zinc (Zn) and Arsenic (As) were analyzed in bulk samples and their size fractions using XRF (Kevex, 0700 XES Control with 5230 Energy/Digital Converter and 4460 Pulse Converter). The elements other than lead selected for analysis were chosen because they are environmentally important compounds, are likely to be found in some industrial soils and they could be analyzed by XRF conveniently and accurately in conjunction with lead. The analysis conditions used for each element are summarized in Table 4.

XRF Conditions	Elements					
	Pb	Ni	Cu	Zn	As	
Target Element	Zr	Ge	Ge	Ge	Zr	
Counts	350	350	350	350	350	
KV	30	20	20	20	30	
mA	0.2	0.5	0.5	0.5	0.2	

Table 4: XRF conditions used to analyze bulk soils and fractions

Up to 16 sample cells can be loaded into the instrument tray for each batch of analyses. For each batch either Zr targeted elements (Pb or As) or Ge targeted elements (Ni, Cu, and Zn) were analyzed along with the corresponding standard samples. Standards were obtained from the National Institute of Standards and Technology (NIST) and the United States Geological Survey (USGS). The concentrations of the elements in all standards used for determination were certified by the supplying agency. Information about the standards used is given in Table 5. After analysis of a sample batch the concentrations of Pb and co-contaminants were calculated by an automated calibration procedure.

Standards	Elements	Conc.
		mg/kg
NIST 2709	Pb	18.9
San Joaquin Soil	Ni	88
San Joaquin Son	Cu	34.6
	Zn	106
	As	17.7
NIST 2711	Pb	1162
Montana Soil	Ni	20.6
Moderately Elevated Traces	Cu	114
	Zn	350.4
	As	105
NIST 2710	As	626
Montana Soil		
Highly Elevated Traces		
USGS-AEG GXR4	Pb	52
Copper Mill-Head		
USGS-AEG GXR5	Ni	75
Soil	Cu	354
	Zn	49

Table 5. Standards used in XRF analysis of bulk soils and fractions

2.4. Measurement of PM₁₀ Formation Potential

Resuspension of soil to create dust for the collection of PM₁₀ and PM_{2.5} under laboratory conditions permits the evaluation of both the PM₁₀ potential of the soils under controlled conditions and the analysis of the soil independent of crop and other field parameters. The four basic components of the UC Davis CNL dust resuspension chamber include (i) a fluidizing bed dust resuspension chamber that separates PM₁₀ from the soil sample, (ii) a collection chamber that collects suspended airborne dust, (iii) a Sierra Andersen PM₁₀ inlet that separates PM₁₀ in the resuspended dust from larger resuspended material and, (iv) an IMPROVE (Interagency Monitoring of Protected Visual Environments) sampler which collects PM₁₀ onto teflon filters (Figure 1).

The fluidizing bed of the resuspension chamber uses upward flowing air to churn the sample and suspend particles less than approximately 50 μ m aerodynamic diameter into the air stream. These particles are then carried into the dust collection chamber. The dust collection chamber is a 90 liter painted wood box that contains a Sierra-Anderson PM₁₀ inlet attached to an IMPROVE sampler. With the IMPROVE sampler, four filter samples can be collected in sequence without pausing to change filters. Prior to measurement, the soil is oven-dried overnight at 105 °C.

Approximately 1 g of sieved soil material is placed in the dust resuspension chamber, which is then sealed. A 1.0 cm diameter aluminum tube connects the top end of the dust resuspension chamber to the inside of the dust collection chamber. A measured volume of air (3.5 lpm for 15 seconds) is forced through the soil sample at the base of the fluidizing bed. The upward air velocity is sufficient to suspend dust particles up to ~50 μ m aerodynamic diameter, which are then carried out of the resuspension chamber and into the collection chamber. The particles are collected on 47 mm Teflon filters at 16.7 lpm after passing through a Sierra Anderson PM₁₀ inlet.

For this study, the PM_{10} index measurement procedure was modified from its original total sampling time of 150 minutes (Carvacho 2001) to a total of 60 minutes of sampling time. The sample was puffed every 5 minutes using a timer delay relay and the resulting dust was collected onto a single 47 mm Teflon filter. Preliminary work with soils of different textures indicated a linear relationship between the PM_{10} mass obtained at 150 minutes and the modified procedure using 60 minutes of sampling time (Figure 2). All PM_{10} mass obtained during the 60 minutes sampling interval was converted to a PM_{10} mass at 150 minutes by using the regression equation described in Figure 2. The PM_{10} formation potential is calculated as the weight of PM_{10} in the sample per gram of soil suspended.



Figure 1. Diagram of Soil Resuspension Apparatus (Carvacho 2001)



Figure 2. Relationship between PM₁₀ mass at 150 minutes and 60 minutes of sampling time for three different soils.

2.5. Measurement of Lead Concentrations on Filters

Lead concentrations on the filters from the resuspension experiments and on bulk soils were determined by energy-dispersive x-ray fluorescence (XRF) using the Compton peak ratio method and a suite of secondary standards. This system was custom designed to provide a high flux of x-rays over a relatively small beam spot. The system uses a Mo anode, which maximizes sensitivity in the region of Se. Blank filters were initially checked for trace levels of lead contamination. Figure 3 shows the minimum detectable limit for all elements routinely measured in the IMPROVE program. The minimum detectable limits illustrated in Figure 3 have been converted to mass/volume of air sampled in a typical IMPROVE sample; the shape of the curve would remain the same for the samples analyzed in this study.



Figure 3. Minimum detectable limits for elements H, Mg to Zr and Pb. H and Pb have the proper minimum detectable limits, but are drawn at arbitrary atomic numbers for display.

The x-ray tube in the CNL system (Figure 4) is a General Electric grounded anode diffraction type, with a molybdenum anode. The high-resolution SiLi detector has pulsed optical feedback to provide high count rate capabilities. The data from the detector is processed by standard pulse processing electronics and is accumulated in a dual ported analog to digital converter connected to an Ethernet local area network.

The XRF system was involved as a NIST collaborative laboratory for the development of standard reference material SRM 1833 multi-element x-ray standard.



Figure 4. Setup of XRF analysis system for filters

The baseline calibration of the system consists of analyzing a series of 30 commercial and NIST elemental standards. At the beginning of every analytical session, the calibration is checked with a series of 15 standards and by reanalyzing several samples from the previous analysis session.

The spectra are analyzed by an automated code developed at CNL especially for particulate samples. The program removes the background, searches for all peaks, then fits each peak to a Gaussian. The peaks are identified and the areal density of each element in ng/cm² is calculated. Overlapping emission line interferences are eliminated by using a series of algorithms, appropriate matrix corrections are applied, and (for air samples) the volume and area are incorporated to give concentrations in ng/m³.

The analysis is performed only after the precision requirements are met. Scatter plots of the major elements are prepared and checked for consistency. If the calibration and reanalysis are within the accepted 4%, the regular analysis is allowed to proceed. At the end of the session, the standard and reanalysis trays are reanalyzed.

2.6. Calculations

```
\begin{array}{l} PM_{10} \mbox{ Formation Potential (mg PM_{10}/g soil) is calculated as:} \\ PM_{10} \mbox{ Formation Potential = mg PM_{10} (t = 150 mins)/g soil} \\ PM_{10} \mbox{ Pb Concentration (} \mu g \mbox{ PM}_{10} \mbox{ Pb}/g soil) is calculated as:} \\ PM_{10} \mbox{ Pb Concentration = A x B} \\ where, \\ A = (mg \mbox{ PM}_{10}/g soil)/1000 \\ B = \mu g \mbox{ Pb}/g \mbox{ PM}_{10} = (ng \mbox{ Pb}/cm^2) \ x \ (Masked area of filter/mg \mbox{ PM}_{10}) \\ Masked area of filter = 2.2 \ cm^2 \end{array}
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3. RESULTS AND DISCUSSION

3.1. Overview of Results

The major objective of this study was to determine whether resuspension of lead containing soils around industrial facilities might be a significant contributor to locally elevated airborne lead levels. Before proceeding with a thorough examination of the data from the study, it is instructive to make an initial estimate of this potential contribution. For soil resuspension to be a problem in this context, the soil must contain elevated level of lead, the soil must yield appreciable amounts of fine particulate matter, and lead concentrations in the particulate matter must be high. Table 6 summarizes average values across each site for the amount of lead in the bulk soil, the yield of fine particulate matter in resuspension experiments, the concentration of lead on the resulting particulate matter and an overall amount of particle bound lead produced per gram of soil. Of the facilities tested only the lead smelter had average soil lead concentrations significantly above the 12.4-97.1 µg/g range reported for California soils (Bradford et al. 1996). Yields of fine particulate matter from the soils collected ranged from 0.20 to 0.87 mg PM_{10}/g soil. The concentration of lead on this particulate matter was higher than the corresponding bulk soil concentration in every case tested, with the most significant enrichment, by a factor of 8.6, observed for the glass manufacturing facility. Combining the PM₁₀ yield and its lead concentration resulted in production of between 0.06 and 1.23 µg of particle bound lead per gram of bulk soil. As an example of a worst-case scenario, if soil at the lead smelter site were tilled to a depth of 1 cm and this operation efficiently stripped away the fine particulate matter, the operation would produce approximately 80 g of particle bound lead per acre. To put this number into perspective, the median daily lead emission from the 58 California counties reporting lead emissions in 1996 was 527 g/d (California Toxics Inventory, 1996). Under at least some circumstances, therefore, it appears that PM₁₀ derived from lead contaminated soils could be a significant transient local source of atmospheric lead. The remainder of this section explores the measurement and interpretation of each of the parameters in Table 6 in more detail.

Tuble of overview of fiverage results by Facility Type						
Facility	Average	Average PM ₁₀	Average PM ₁₀	Average PM ₁₀		
	Bulk	Yield	Pb Conc.	Pb Yield		
	Conc.	$(mg PM_{10})$	(µg Pb/g PM ₁₀)	(µg PM ₁₀ Pb		
	(µg Pb/g soil)	/g soil)		/g soil)		
Waste Treatment	134	NM	NM	NM		
Glass Manufacturing	65.2	0.20	563.5	0.10		
Saw Mill	53.5	NM	NM	NM		
Perlite Mining	99.4	0.70	110.3	0.07		
Borax Processing	66.3	0.56	118.1	0.06		
Cement Manufacturing	65.2	NM	NM	NM		
Lead Smelter	622	0.58	2283	1.23		
Sand Blasting	132	0.87	183.8	0.15		
Roadside	135	NM	NM	NM		

Table 6. Overview of Average Results by Facility Type

NM=not measured

3.2. Soil Lead Concentrations

The average, maximum and minimum bulk soil lead concentrations observed at each facility are shown in Figure 5. Only the waste treatment plant (site 1), the lead smelter (site 7), the sandblasting facility (site 8) and the roadside samples display any significant inter-sample variability in bulk lead concentrations. Since samples were taken at varying distances from the emission source at each facility, similar lead concentrations across the site suggest that the emission source was not controlling soil lead levels. The sites with minimal variation in lead levels (sites 2-6) also have the lowest average soil lead concentrations, with none exceeding 105 ug/g, suggesting that these sites probably feature typical industrial background levels of lead in their soils. For the potentially contaminated sites, maximum lead concentrations range from 190 to 1540 ug/g.



Figure 5. Bulk soil lead concentration by facility type

Additional detail about the bulk soil lead concentrations in samples from each of the facilities is provided in tables 7 through 14. For each sample collected at the facility the table lists the distance of the sample from the emission source, the direction of the sample from the source, and the concentration of lead, nickel, copper, zinc and arsenic in the sample. Samples collected downwind from the emission source along the prevailing wind direction are indicated with an asterisk in each table.

Elemental concentrations at the wastewater treatment facility are provided in Table 7. The source of contamination at this site was a soil pile contaminated with leaded gasoline. Similar types of contamination might not exist at a typical wastewater treatment facility or on other portions of the property at the sampled facility. Lead levels at this site were higher downwind of the source than they were upwind of it. Only two samples were taken downwind of the source making it impossible to determine the functional form of the concentration decay with distance. There is a statistically significant (p<0.05) linear correlation between the concentration of lead and copper (0.92), zinc (0.94) and arsenic (0.99) suggesting that each of these elements is a contaminant being introduced from common source.

	wastewater treatment facility (Site 1)						
Sample	Distance	Direction	Pb	Ni	Cu	Zn	As
Number	(m)		(ppm)	(ppm)	(ppm)	(ppm)	(ppm)
1-1	182	W	65.5	20.4	21.8	92.9	10.4
1-2	22.1	W	70.9	33.0	60.3	141	10.8
1-3	6.60	W	76.9	34.3	41.1	129	11.9
1-4*	32.0	E	153	40.3	95.0	176	22.9
1-5*	42.4	E	304	45.9	130	228	44.7

 Table 7. Lead and co-contaminant concentrations in bulk soil samples from a wastewater treatment facility (Site 1)

Samples collected near the glass manufacturing facility had consistent and relatively low lead concentrations (Table 8) and there is no statistical difference between the lead concentration in samples collected upwind and downwind of the source. It is possible that these results were found solely because of the long distance between the source and the sampling locations (>242 m). Samples were collected from off-site as close as possible to the source. Other elements measured in the soil also displayed consistent values with little difference between upwind and downwind samples. A significant linear correlation (0.99) is observed between lead and nickel in these samples, but because both levels are within a normal range for California soils this may be a natural occurrence rather than evidence of contamination.

 Table 8. Lead and co-contaminant concentrations in bulk soil samples from a glass manufacturing facility (Site 2)

Sample	Distance	Direction	Pb	Ni	Си	Zn	As
Number	(m)		(ppm)	(ppm)	(ppm)	(ppm)	(ppm)
2-1	259.7	NE	70.8	52.6	26.3	73.5	11.8
2-2	242.0	NE	61.6	33.9	24.6	85.8	10.1
2-3*	382.0	NW*	64.2	41.6	24.5	88.1	9.6
2-4*	382.1	NW*	64.1	40.9	27.3	99.8	10.8

Samples were collected from the saw mill with the expectation that lead levels would be low and might represent an "industrial background" for lead in soils (Table 9). Only two samples were collected from the site, both from downwind of the mill. Both of the samples contained lead concentrations within the normal range for California soils, indicating that the site was probably not contaminated. Concentrations of co-contaminants were also within normal ranges. The sample collection point closest to the source was more than 139 m away; higher concentrations might have been observed closer to the source. With only two samples collected from the site, nothing can be said about correlations between the elements measured.

	linin (Site 5)						
Sample	Distance	Direction	Pb	Ni	Си	Zn	As
Number	(m)		(ppm)	(ppm)	(ppm)	(ppm)	(ppm)
3-1*	139.4	E*	62.7	59.1	62.7	85.6	10.9
3-2*	195.6	E*	44.4	29.8	75.0	92.2	8.92

 Table 9. Lead and co-contaminant concentrations in bulk soil samples from a saw mill (Site 3)

The perlite mining facility had lead concentrations slightly above the "normal range" determined by Bradford et al. (1996), but the levels did not vary significantly across the site (Table 10). All samples were collected downwind of the source so it is possible that upwind levels might have been lower than the 91-105 range observed. No trend of decreasing lead concentrations was observed for samples collected at increasing distances from the emission source. Lead concentrations were significantly correlated with copper (0.93) in these samples, but lead was not strongly correlated with any other element measured.

 Table 10. Lead and co-contaminant concentrations in bulk soil samples from a perlite mining facility (Site 4)

			0		/		
Sample	Distance	Direction	Pb	Ni	Си	Zn	As
Number	(m)		(ppm)	(ppm)	(ppm)	(ppm)	(ppm)
4-1*	23.7	W*	105	2.19	18.1	58.9	18.7
4-2*	122	W*	97.7	3.04	8.90	56.6	15.9
4-3*	156	W*	91.3	3.57	7.60	56.2	16.0
4-4*	189	W*	103	3.73	14.5	80.0	17.7

The Borax processing plant had soil lead levels between 59.6 and 73.2 mg/kg (Table 11). The small difference between samples and the low level relative to benchmark California soils suggests little or no lead contamination emanating from the processing plant. Contamination may exist closer to the emission source inside the facility boundary. The closest sample collected for this study was 174 m from the emission source. Statistically there is no difference between the upwind and downwind lead concentrations. None of the other element's soil concentration is correlated with measured lead concentrations at the 0.05 confidence level.

Table 11. Lead and co-contaminant concentrations in bulk soil samples from aborax processing plant (Site 5)

			8	(13-11)	/		
Sample	Distance	Direction	Pb	Ni	Си	Zn	As
Number	(m)		(ppm)	(ppm)	(ppm)	(ppm)	(ppm)
5-1	203	SE	67.2	12.9	16.2	70.6	11.9
5-2	214	SE	65.7	5.24	20.1	49.9	10.2
5-3	287	SE	73.2	6.18	12.8	52.7	13.1
5-4*	174	SW*	64.0	4.09	11.0	43.8	11.2
5-5*	232	SW*	68.3	4.97	6.37	44.7	11.2
5-6*	238	SW*	59.6	3.19	6.00	35.4	10.0

Since access to several cement manufacturing facilities was denied to the project team, all samples were collected off-site at significant distances (>400 m) from the emission source (Table 12). None of these distant samples displayed obviously elevated lead levels and there was no statistical difference between samples collected upwind and downwind from the emission source. Lead concentrations in these samples were not significantly correlated with any of the other elemental concentrations measured. All of these findings may arise because of the large distance between emission source and sampling locations, or they may reflect the absence of significant emissions of these chemicals at the sampled facility.

)		
Sample	Distance	Direction	Pb	Ni	Си	Zn	As
Number	(m)		(ppm)	(ppm)	(ppm)	(ppm)	(ppm)
6-1*	479	N*	65.6	10.8	24.8	130	10.6
6-2*	490	N*	65.8	12.4	29.3	125	9.88
6-3*	506	N*	64.4	11.2	27.4	121	11.9
6-4	474	NE	64.6	17.1	49.1	173	11.0
6-5	500	NE	68.8	16.3	35.7	143	11.1
6-6	518	NE	69.0	15.4	33.8	129	11.1
6-7	419	Е	58.5	5.00	16.5	78.9	8.79
6-8	419	SE	64.8	3.78	8.54	93.8	9.14

 Table 12: Lead and co-contaminant concentrations in bulk soil samples from a cement manufacturing plant (Site 6)

The lead smelter exhibited the highest soil lead levels of any of the facilities sampled in this study, ranging from 91 to 1540 mg/kg (Table 13). This elevation in lead concentrations above background levels is comparable to an enhancement of 15 observed for most lead smelters (Ragaini *et al.* 1977). The decrease in lead concentrations with distance from the emission source is approximately exponential (R^2 =0.77) as shown in Figure 6. At the sampling locations furthest from the emission source (<200 m), soil lead concentrations begin to approach industrial background levels observed at other sites in the study. The high level of Pb contamination closest to the smelter and the rapid decline with distance is comparable to similar observations reported by Ragaini *et al.* (1977). Lead levels in the soils are significantly correlated with both zinc (0.93) and arsenic (0.99) concentrations suggesting that these elements may be emitted along with lead from the smelter.

		511		.)			
Sample	Distance	Direction	Pb	Ni	Си	Zn	As
Number	(m)		(ppm)	(ppm)	(ppm)	(ppm)	(ppm)
7-1	123	E	1540	31.5	69.5	126	165
7-2	129	E	551	21.6	41.5	98.7	73.2
7-3	195	E	577	21.2	39.3	108	77.6
7-4	234	Е	116	24.9	27.6	80.9	17.5
7-5	248	E	91.0	19.4	29.2	78.1	13.8
7-6	256	Е	124	20.0	27.8	89.7	19.6
7-7*	161	NE*	1350	26.7	39.9	114	147
7-8*	176	NE*	628	26.2	37.3	91.1	82.0
7-9*	193	NE*	751	22.2	38.6	166	93.9

Table 13: Lead and co-contaminant concentrations in bulk soil samples from a lead smelter (Site 7)



Figure 6. Decreasing lead levels in soil with increasing distance from the emission source at a secondary lead smelter

Lead levels at the sand blasting facility were above industrial background concentrations at locations near the source but appear to fall to background by about 30 m away (Table 14). With one exception (sample 8-7), concentrations were significantly higher downwind of the source than in the other directions. Lead concentrations generally decline with increasing distance from the source in each of the directions. Arsenic is significantly correlated (0.99) with lead in these samples, as is copper to a lesser extent (0.89). Although zinc is not strongly correlated with lead levels, the concentrations are quite high compared to other soil samples analyzed in this study suggesting an alternative source of this element.

biasting facinty (Site 0)								
Sample	Distance	Direction	Pb	Ni	Си	Zn	As	
Number	(m)		(ppm)	(ppm)	(ppm)	(ppm)	(ppm)	
8-1	0.87	SW	157	7.97	23.9	1430	23.6	
8-2	1.64	\mathbf{SW}	130	6.59	17.8	1218	21.1	
8-3	3.82	\mathbf{SW}	113	5.52	23.3	1794	18.0	
8-4	17.1	\mathbf{SW}	120	42.8	22.3	271	19.0	
8-5	33.0	\mathbf{SW}	54.9	6.69	8.36	24.9	8.53	
8-6*	10.9	SE*	322	13.3	30.3	1140	48.2	
8-7*	12.2	SE*	80.8	10.1	13.7	312	12.1	
8-8*	14.5	SE*	203	7.35	26.8	442	33.5	
8-9*	25.1	SE*	208	10.3	27.3	592	31.5	
8-10	23.8	S	63.6	16.0	11.6	63.2	11.1	
8-11	34.5	W	61.0	13.5	9.41	39.7	9.70	
8-12	13.1	NW	68.6	4.72	11.4	125	11.0	

 Table 14. Lead and co-contaminant concentrations in bulk soil samples from a sand blasting facility (Site 8)

The concentration of lead and co-contaminants in bulk roadside soils are summarized in Table 15. Roadside soil samples analyzed in this project were initially collected for a California Department of Transportation study and therefore the procedures differed in some respects from those used at the other facilities. The three roadside samples were collected from three highways in different parts of the state rather than at three different distances from a single emission source. All samples were collected 5 feet (1.52 m) from the edge of the pavement. These samples were the first analyzed under this project and arsenic levels were not determined. Although many studies have documented the relationship between roadside soil lead levels and traffic volume, for these three soils there is little relationship between lead concentrations and the most recent average daily traffic volumes (ADT). This is probably because some of the highways were widened, rerouted or improved since the phase-out of leaded gasoline when most of the lead was deposited. Lead concentrations in two of the samples, RS-27 and RS-50 were below 50 mg/kg and probably represent industrial background levels. The third sample, RS-103, had a lead level of 322 mg/kg, comparable to some of the more contaminated industrial locations sampled. No information was collected when the samples were taken regarding the prevailing wind direction or the direction of the sampling location from the roadway so no conclusions can be drawn about directional effects. There is a statistically significant correlation between lead concentrations and the concentration of nickel (0.99), copper (0.99) and zinc (0.91). Each of these elements is known to be a product of automotive traffic so this finding is not surprising.

Sample	Distance	1996	Pb	Ni	Си	Zn			
Number	(m)	ADT	(ppm)	(ppm)	(ppm)	(ppm)			
RS-27	1.52	216,000	46.0	19.0	26.0	93.0			
RS-50	1.52	107,000	38.0	4.10	22.0	50.0			
RS-103	1.52	202,000	322	306	42.0	149			

 Table 15. Lead and co-contaminant concentrations in bulk soil samples collected along California highways

In addition to measuring lead levels in bulk soil samples, a critical element of this project was to determine which, if any, soil size fractions produced the largest amount of lead bearing particulate matter. This information was considered essential for future efforts to extrapolate the current results to additional sites without performing the full analysis. Particle size distributions were measured only for the downwind samples from each industrial facility. A graphical summary of the average mass percentage of soil particles in each size range is provided in Figure 7. Samples from sites 1, 2, 3 and the roadside are dominated by coarse materials, with particles larger than 300 μ m constituting over 75% of the sample mass. Finer materials dominate the samples from sites 4, through 8. Site 6, for example, averages nearly 50% of the soil passing a 150 μ m sieve.



Figure 7. Summary of average soil particle size distributions by site

A second important reason for the fractionation procedure was to identify any significant enrichment of lead in particular size fractions in comparison to the bulk concentrations. As an example of how such an enrichment might be facility specific, some investigators have observed Pb enrichment in large soil size fractions at smelter sites from large Pb-containing particles emitted by these operations (Ma *et al.* 1997).

However, the results revealed no systematic enrichment of lead in any one fraction at any of the sampling sites, with concentrations in most fractions generally within a factor of 2 of the average bulk value (data not shown). A summary of all the lead concentration data within various size fractions is provided in Figure 8. The figure reveals a slight tendency for the finer soil fractions to contain higher concentrations of lead, but these differences are probably insignificant in the context of the present investigation.

Accumulation of lead in the finer size fraction is consistent with previous research findings. For example, lead in soil sampled from mining areas showed a clear concentration in the finest (2-20 μ m) fractions (Queralt and Plana 1992). Lead in several mining wastes was similarly concentrated in the finest (23 μ m) particle size (Clevenger 1990). The large surface areas in the silt and clay soil size fractions may play a role in lead accumulation (Queralt and Plana 1992).



Figure 8. Lead concentration distribution in soil size fractions from all locations sampled. The box defines the 25th to 75th percentiles and the whiskers delineate the 10th to 90th percentiles.

As a quality control check on the lead concentration measurements, the lead concentration measured directly on whole soil samples was compared to the concentration calculated by weighting the concentration of each size fraction by its mass fraction in the soil sample. Figure 9 shows that there is good agreement ($R^2=0.99$) between these measured and calculated values but that the calculated values are consistently about 20% larger than those measured for the whole soils. Reasons for this discrepancy are not obvious, but may relate to the crushing of the whole soil samples in a ball mill prior to XRF analysis, which might have caused some loss of highly concentrated fine lead bearing materials, although similar losses are likely to have affected the sieving process. A discrepancy of this magnitude does not compromise the

ability of this study to determine whether lead contaminated soils may pose a threat to air quality.



Figure 9. Comparison of lead content measured and calculated for bulk soils

3.3. PM₁₀ Generation

An effort was made to resuspend each bulk soil sample and each soil fraction. This was not always possible, either because an insufficient quantity of a particular size fraction was available or because time constraints prevented the experiment from being conducted. No resuspension experiments were performed for sites 1, 3 and 6 for these reasons. Most resuspension experiments were conducted in duplicate, although in some cases more replicates were performed to determine the variability of the procedure and occasionally only a single replicate was performed because of sample limitations. More than 300 resuspension experiments were conducted in the course of this study.

For the samples that were resuspended, yields of PM_{10} averaged 0.60 mg/g soil with a range from 0.03 to 2.01 mg PM_{10} /g soil. To examine whether any particular soil size fraction was driving these results, resuspension of the individual soil size fractions was conducted. As expected, the coarsest soil fractions produced the smallest amount of PM_{10} per gram of material. For most of the sites, the PM_{10} yield was the greatest in either the 45-75 µm fraction or the 38-45 µm fraction (data not shown). Because the relationship between PM_{10} yield and size fraction was similar across the sites, a distribution of yields for all sites is displayed in Figure 10. The figure confirms that particles passing a 75 µm sieve produce the most significant amount of PM_{10} , with sizes from 38-75 mm exhibiting especially high yields. Efforts to develop a simple correlation that would allow prediction of a soil's PM_{10} yield from individual soil texture

characteristics were not successful, however. It is likely that soil aggregate structure and composition, in addition to particle size, are important determinants of the PM_{10} formation potential of a soil.



Figure 10. PM₁₀ yield for each soil size fraction from all facilities. The box defines the 25th to 75th percentile of the data and the whiskers define the 10th to 90th percentiles.

The yields for each fraction shown in Figure 10 use a basis of 1 gram of the particular soil fraction rather than 1 gram of whole soil. To understand which size fraction contributes most significantly to PM_{10} formation, it is necessary to multiply these results by the mass percentage of each fraction in a particular soil. Figure 11 shows the calculated contribution of each size fraction to the overall PM_{10} yield for each site. Although the larger soil particle sizes produce relatively little PM_{10} per gram, they are still significant contributors to a soil's overall PM_{10} formation potential because they comprise the bulk of most of the soils. After accounting for the relative abundance of each fraction, the 45-75 µm fraction appears to be the most significant for PM_{10} formation.



Figure 11. Contribution of each size fraction to the PM₁₀ yield at various sites.

As a quality control check on the resuspension results, PM_{10} yields measured when bulk soil samples were resuspended were compared to the yield calculated as described above and shown in Figure 11. If PM_{10} yields were strictly additive and no errors were made during the experiments, the PM_{10} values obtained by direct measurement and by summation should be identical. Figure 12 plots the yield calculated from the fractions against that measured directly. When the two yields match exactly, the data will fall on the 1:1 line in the figure. Most of the data points fall below the line indicating that the yield calculated from the fractions underestimates the true yield for the whole soil. One possible explanation is that during the dry sieving process that some PM_{10} precursor material was lost. A loss of 1% of the total soil mass charged to the sieve nest is typical during soil fractionation, equivalent to a 10 mg/g loss of material. Since the largest deviations between the data and the 1:1 line in Figure 12 are on the order of 0.5 mg/g, this explanation is clearly plausible.



Figure 12. Comparison of PM₁₀ yield measured directly on bulk samples to yield calculated as a weighted sum of soil fraction contributions.

Very little is known about how the PM_{10} yields measured here relate to actual PM_{10} formation under field conditions of wind erosion or mechanical disturbance. It is thought that these numbers represent the maximum potential for a soil to produce PM_{10} since the soils are thoroughly dried and vigorously resuspended. Consequently, these results should be viewed as "worst case" rather than "typical" amounts of PM_{10} formed from soil.

3.4. PM₁₀ Lead Content

For contaminated soil to be a significant contributor to elevated levels of airborne lead, the soils must produce appreciable quantities of PM_{10} and the resulting PM_{10} must have high lead concentrations relative to those observed during past periods of high ambient lead. For example, concentrations of lead in PM_{10} at the Air Resources Board's Los Angeles-North Main monitoring station during the period of 1990-1996 averaged 800 µg/g with a 10th percentile of 425 µg/g and a 90th percentile of 1200 µg/g. High, low and average values of lead concentrations in PM_{10} from bulk soil at various facilities are shown in Figure 13. Lead concentrations in PM_{10} formed during resuspension of bulk soil samples ranged from 11.5 to 5630 ug/g. The highest of these levels, recorded for samples from the lead smelter, are clearly significant in comparison to levels in particulates observed during peaks in airborne lead levels in recent years, while the lowest levels appear to be of little concern during periods of normal particulate loading. Lead concentrations in PM_{10} from sites 2, 7 and 8 exceeded the 10th percentile from the North Main monitoring station while those from sites 4 and 5 did not.



Figure 13. Maximum, minimum and average lead concentrations in PM₁₀ generated from bulk soils from several industrial facilities

Resuspension experiments are not routine laboratory measurements so it would be useful if bulk soil lead concentrations, which are routinely measured, could be used to identify soils that might pose a risk of producing lead contaminated airborne particulate matter. The concentration of lead in PM_{10} derived from whole soils is plotted against the corresponding soil lead concentration in Figure 14. The higher concentration data is well described (R^2 =0.95) by a line passing through the origin with a slope of 3.60 indicating that, on average, the lead concentration in PM_{10} is enriched by a factor of 3.6 compared with the bulk soils from which it was derived. This equation can not be used generally because it is driven by the data points above 1000 µg Pb/g PM₁₀, all of which are from the lead smelter site. The figure also includes a line of slope one to show that the lower concentration data is also enriched in lead to varying degree with only a few data points falling below the 1:1 line.



Figure 14. Lead enrichment in PM₁₀ derived from bulk soils from the industrial facilities. (A) Full concentration range (B) expanded view of the low concentration range

To determine whether specific soil fractions produced especially high lead concentrations in PM₁₀, the resuspended soil fractions described above were analyzed for lead and a suite of co-contaminants. The maximum, minimum and average production of lead in the PM₁₀ fraction are shown for five of the facilities and the roadside samples in Figures 15-20. In each case the potential of the soil fraction to produce lead-bearing PM_{10} is expressed as the mass of lead found in PM₁₀ per gram of the soil fraction charged to the resuspension apparatus ($\mu g PM_{10} Pb/g$ soil fraction). These results were obtained by multiplying the PM_{10} yield of the soil fraction (mg PM_{10} /g soil fraction) by the lead concentration of the PM_{10} from that fraction (µg PM_{10} Pb/g PM_{10}). Since the PM_{10} yields do not differ across sites and soil fractions as much as the PM_{10} lead concentrations do, the PM₁₀ lead generation results are therefore primarily determined by the PM₁₀ lead concentrations. The highest PM₁₀ lead generation potentials are found in fine soil fractions (38-75 µm) with lower generation potentials for both larger and smaller size fractions in most cases. The soils from the glass manufacturing facility (Facility 2) is an exception, with the highest average PM_{10} lead generating potential found in the soil material passing a 38 µm sieve. For most of the sites the maximum PM₁₀ lead generation potential does not exceed 1 μ g PM₁₀ Pb/g soil fraction. The roadside soil and material from the sandblasting facility were slightly higher, while fine material from the lead smelter exhibited values approaching $12 \ \mu g \ PM_{10} \ Pb/g$ soil fraction



Figure 15. Glass manufacturing (Facility 2) PM₁₀ lead generation by soil fraction



Figure 16. Perlite mining (Facility 4) PM10 lead generation by soil fraction



Figure 17. Borax processing (Facility 5) PM_{10} lead generation by soil fraction



Figure 18. Lead smelter (Facility 7) PM_{10} lead generation by soil fraction



Figure 19. Sand blasting (Facility 8) PM₁₀ lead generation by soil fraction



Figure 20. Roadside soil PM₁₀ lead generation by soil fraction

Since soil particles between 38 and 45 μ m comprise only a small fraction of most soils tested, production from the bulk soil will not approach the maximum values observed in Figures 15-20. To determine the soil fraction having the biggest impact on the formation of lead contaminated PM₁₀ it is important to consider both the productivity of that fraction and its abundance in the whole soil. Multiplying the PM₁₀ lead generation potential of each fraction by its mass fraction in the whole soil allows the identification of

while the 45-75 mm fraction dominates PM_{10} lead production for most of the soils, in the soil from each site is shown in Figure 21. The most striking feature of this graph is that filtration effect of the large gravel along highway rights-of-way. originally deposited as vapor or very fine particulate matter and was trapped by the roadside soils the coarsest size fraction dominates. percentage contribution of each fraction to the average PM_{10} lead generation potential of soil size fractions most responsible for producing lead contaminated PM₁₀. The This may be because the lead was



Figure 21. Percentage contribution of each size fraction to total particulate lead emissions

previously mentioned loss of a small amount (<1 % by mass) of fine soil material during The lower production of lead contaminated PM₁₀ by the fractions is consistent with the agreement ($R^2=0.99$) between these two values but that the slope is less than unity (0.84). whole soil samples was compared to the summation of the fractionated values as was source of airborne lead. affect the robustness of conclusions about the potential for contaminated soil to be a the sieving process. done previously for the PM_{10} formation potential. Figure 22 shows that there is a close For quality control purposes, the PM₁₀ lead generation potential measured in As noted previously, a discrepancy of this magnitude should not



Figure 22. Comparison of PM₁₀ Pb content measured by resuspending bulk soil and soil fractions

3.5. Exposure to Airborne Lead from Soils

The present lack of experimentation relating the amount of PM_{10} produced during laboratory soil resuspension to that produced during various types of realistic resuspension events in the field prevents any detailed exposure calculations from being performed. What has been demonstrated is that for worst-case conditions of very dry soils combined with mechanical agitation, at least some industrial sites have the potential to emit sufficient amounts of lead contaminated PM_{10} to be a significant, transient source of lead to the local airshed. On a more positive note, most of the facilities sampled in this project did not have soil lead levels high enough to pose a danger to air resources even under worst-case conditions. None of the whole soil samples with less than 100 µg/g of lead produced PM_{10} with lead concentrations at even the 10th percentile of that observed at an urban Los Angeles monitoring station, suggesting that such materials could not exacerbate the airborne lead problem at that particular monitoring location.

Relatively simple and reasonably inexpensive source control measures are likely to be effective in eliminating this route of exposure to lead contamination. Increased soil moisture, vegetative cover or pavement would all be effective in reducing the PM_{10} formation potential of the soils to levels that probably would negate the lead threat to air resources. The exact efficacy of these measures in combating this particular problem is unknown; however, they have been proven effective in controlling fugitive dust at industrial sites in previous research.

4. CONCLUSIONS

- 1. Soil lead levels were within the range expected for California soils (12.4-97.1 μ g/g) at most of the industrial sites sampled, but were above the mean for uncontaminated soils (Bradford *et al.* 1996). These sites were considered to represent "industrial background" lead levels of <150 μ g/g. Soil lead concentrations above 150 μ g/g were observed at only three of the eight industrial facilities sampled, all of which were selected because they use or emit lead, and in only one of the three highway samples. The maximum soil lead concentration was 1540 μ g/g, measured at a secondary lead smelter.
- 2. PM_{10} yields from soils averaged 0.60 mg/g and 80% of the values were between 0.18 and 0.97 mg/g. Yields were generally greatest from finer soil fractions (38-75µm) and lower for both coarse (>75 µm) and very fine (<38 µm) fractions.
- 3. Lead concentrations in PM₁₀ derived from soil samples were on average 3.6 times higher than the corresponding bulk soil concentrations. The most contaminated sample from the lead smelter site produced PM₁₀ lead concentrations in excess of 5600 μg Pb/g PM₁₀. The median concentration of lead in PM₁₀ from all sites was only 152 μg Pb/g PM₁₀, suggesting that elevated lead levels are a site specific, rather than generic to all lead using or emitting industrial sites.
- 4. The total yield of particulate bound lead from soil had a median value of 0.10 μ g PM₁₀ Pb/g soil and a maximum value of 4.41 μ g PM₁₀ Pb/g soil. Differences in these values between samples and facilities were primarily determined by differences in the PM₁₀ lead concentration since PM₁₀ vields were more uniform.
- 5. Although lead was correlated with different co-contaminants at different facilities, these correlations do not appear to be immediately useful for distinguishing among different industrial sources of PM_{10} .
- 6. Contaminated soils may be a significant local source of airborne lead contamination under "worst-case" conditions that include very dry fully exposed soils subject to mechanical agitation (e.g., by tilling). Under these conditions, the top 1 cm of the average lead smelter soil has the potential to release 80 g Pb/acre in the form of PM₁₀. If the area were moist, well-vegetated, or paved emissions would be significantly lower.
- 7. This report demonstrates the potential, under a narrow set of conditions, for contaminated soil resuspension to produce locally elevated airborne lead levels. Proof of this link is not offered and would need to be provided by future research monitoring resuspension and airborne lead concentrations simultaneously at a facility with exposed contaminated soil.

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Glossary

- ADT Average daily traffic
- CARB California Air Resources Board
- CNL Crocker Nuclear Laboratory
- DTSC Department of Toxic Substances Control
- IMPROVE Interagency Monitoring of Protected Visual Environments
- NIST National Institute of Standards and Technology
- PM_{10} Particulate matter less than 10 μ m aerodynamic diameter
- SRM Standard reference material
- USGS United States Geological Society
- XRF X-ray fluorescence

Elements

- As Arsenic
- Ce Cerium
- Cs Cesium
- Ga Gallium
- La Lanthanum
- Li Lithium
- Mo Molybdenum
- Na Sodium
- Ni Nickel
- Pb Lead
- Rb Rubidium
- Sb Antimony
- Si Silicon
- Th Thorium
- Tl Tellurium
- Zn Zinc
- Zr Zirconium