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## Review

# Estimation of leaded (Pb) gasoline's continuing material and health impacts on 90 US urbanized areas

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## ABSTRACT

The subject of this paper is lead (Pb) additives in gasoline and their material and health impact from Pb dust inputs into 90 US urbanized areas (UAs). The mass of Pb additives for 90 UAs as a total of the US Pb additives in 1982 were estimated from vehicle travel, vehicle fuel economy (miles/gallon), ratio of leaded to unleaded fuel, and Pb/gallon. About 500 billion ( $10^9$ ) miles of travel in 90 UAs during 1982 account for ~18,000 metric tons (MT), or nearly 30% of the US Pb additives in 1982. Applying the 1982 proportions to the 90 UAs for 1950 through 1982 fuel sales by state accounts for ~1.4 million MT Pb of the US national total of 4.6 million MT during the same years. Fates of Pb additives in engine systems were used to calculate Pb aerosol inputs into the 90 UAs. The inputs range from 100's to more than 100,000 MT of Pb depending on a given UA's traffic flow patterns. Soils are the reservoir of urban Pb dust. The median background soil Pb for the US is 16.5 mg/kg (range 10.3 to 30.1 mg/kg), and less by an order of magnitude or more than soil Pb within larger UAs. Recognizing the US input of massive gasoline Pb additives into UAs assists with comprehending soil Pb differences between large and small UAs, inner and outer areas of UAs, health disparities, and school achievement issues within UAs. The findings underscore the need for controlling accumulated exterior urban Pb dust from gasoline additives along with paint sources that have accumulated in soil to meet the goal of primary childhood Pb exposure prevention.

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Abbreviations: ft<sup>2</sup>, 0.09290304 m<sup>2</sup> or 929 cm<sup>2</sup>; 40 µg/ft<sup>2</sup>, 431 µg/m<sup>2</sup>; DVMT, Daily vehicle miles of travel; MT, Metric tons; NHANES, National Health and Nutrition Examination Survey; US gallon, 3.79 L; US miles, 1.61 km; VMT, Vehicle miles traveled per year.

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## 1. Introduction

The subject of this paper is lead (Pb) additives to gasoline and their material and health impact from Pb dust inputs into 90 US urbanized areas (UAs). The history of the discovery, early industrial accidents, opposition to, commercial use, and the January 1, 1986 rapid phase out of ~90% of the Pb additives to gasoline are well described in the literature (Bridbord and Hanson, 2009; Kovarik, 2005; LaBelle et al., 1987; Needleman, 1998; Nriagu, 1990; Rosner and Markowitz, 1985). About the same mass (5–6 million MT) of anthropogenic Pb was used in the commercial production of nonpaint Pb additives to gasoline as in paint production (Mielke and Reagan, 1998). The World Health Organization emphasizes accumulated Pb dust from paint and nonpaint sources in soil as a major concern for children's health (Louis et al., 2006). US policy emphasizes lead-based paint as the major concern for children's health and indicates that nonpaint sources are "insufficiently characterized... [and] ... often underestimated" (Levin et al., 2008). In 1978, paint sources of Pb were restricted to 600 mg/kg, and in 1995 the ban was completed on Pb additives to gasoline for US highway travel (Levin et al., 2008; US EPA, 1996a). The use of Pb-based paint on older buildings is the subject of intense scrutiny. Estimation of the magnitude of additives in gasoline as a nonpaint source of Pb inputs into urban environments must also be scrutinized to comprehend the impact of essentially equal masses of Pb in paint and gasoline on the environment and health.

The purposes of this paper are: 1) to estimate the mass of gasoline Pb aerosols dispersed into US urbanized areas from 1950 through 1982, 2) to review studies concerning US background soil Pb and soil Pb in US urbanized areas, and 3) to characterize the environmental health impact that the input of Pb dust may have on various sized UAs.

## 2. Methods

First, the mass of gasoline Pb additive from 1950 to 1982 were calculated from state fuel sales and compared with data from the Ethyl Corporation (US Senate, 1984). Second, the proportions of Pb additives used in 90 US urbanized areas were calculated for 1982 from vehicle travel data and public records. Third, the proportion of the Pb additives in each UA in 1982 were applied to state by state fuel consumption records from 1950 to 1982 to derive the quantities of Pb used in the 90 UAs. Finally, a review of soil Pb and health studies was conducted to assist with characterizing the environment and health effects of gasoline Pb additives on the 90 UAs.

### 2.1. Mass of gasoline lead additives in the US 1950–1982

Fuel sales by state for all grades of gasoline for 1950–1975 were obtained from Highway Statistics Summary (US DOT, 1987). The fuel

volume by gasoline grade (regular leaded, regular unleaded, premium leaded, and premium unleaded) sold in each state from 1975 to 1982 were obtained from the Ethyl Corporation (1982) yearly report. The gasoline quantities were converted into the amounts of Pb from data on the historical trends of grams of Pb per gallon for the various grades of gasoline (Shelton et al., 1982).

### 2.2. 1982 Traffic lead emissions for 90 urbanized areas

Vehicle traffic data is collected by every state using guidelines from the US Department of Transportation, Federal Highway Administration (US DOT 1999). These data are an essential requirement for setting priorities for taxpayer expenditures of highway construction and maintenance in the US. Vehicle traffic data originating from the Federal Highway Administration are compiled by the Texas Transportation Institute (TTI, 2009). The TTI vehicle traffic data were sorted and tabulated for 90 urbanized areas. To estimate the quantity of Pb attributable to each of the urbanized areas, the following data were assembled for 1982: vehicle traffic mileage for 1982 for 90 UAs (TTI, 2009), fuel efficiency in miles per gallon or mpg (US DOT), percentage of various grades of gasoline (Ethyl, 1982), and grams of Pb per gallon of leaded fuel for 1982 (Lewis, 1985). All of these data components for 90 UAs were available only for 1982.

Lead dust for each urbanized area were estimated as follows: daily vehicle miles traveled (DVMT) were summed for freeway and arterial street travel and then multiplied by 365 to obtain the annual vehicle miles traveled (VMT) for 1982. The 1982 gallons of gasoline were estimated by dividing the annual VMT by the 1982 average miles per gallon (14.1 mpg). The volume of gasoline was multiplied by state proportions of leaded gasoline as reported by the Ethyl Corporation (1982). In the cases where UAs are linked with multiple states, the averages of the combined state proportions of leaded gasoline were used. Then mass of Pb for each UA were estimated by multiplying the number of gallons of leaded gasoline times 1.1 g per gallon (Lewis, 1985). Proportions were assigned by dividing the mass of Pb additives from vehicle traffic of each UA by the total mass of Pb additives for the nation in 1982 (Mielke et al., 2010; US Senate Hearings, 1984).

### 2.3. Estimated Pb additives and Pb aerosols into 90 UAs 1950–1982

Assuming approximately the same proportion of Pb for each of the 90 UAs in 1950–1982 as for 1982, estimates of total Pb were made for each UA. The fates of Pb additives in gasoline were based on information from the US EPA (1986). According to the EPA, 75% of the Pb additives were emitted as exhaust directly into the atmosphere. Particle sizes of the Pb aerosols were allocated into two categories, >10 micrometers (µm) (40%) and <0.25 µm (35%), also 25% of the Pb in gasoline remained in the engine and adhered to

engine or exhaust surfaces (15%), and the remaining Pb (10%) entered the lubricating oil (US EPA, 1986).

### 3. Results

#### 3.1. Annual mass of US Pb gasoline additives from 1950–1982

Fig. 1 is a graph of the annual masses of Pb in metric tons (MT) calculated from fuel sales in the US between 1950 and 1982 along with the quantities of Pb additives reported by the Ethyl Corporation for the same time period at the Senate Hearings on the Airborne Lead Reduction Act of 1984, S.2609 (US Senate, 1984). The total amount of US Pb additive calculated from state fuel consumption records between 1950 and 1982 is 4.64 million MT. This compares with 4.61 million MT reported for 1950 through 1982 by the Ethyl Corporation (US Senate, 1984); the correlation coefficient is 0.981 (P-value  $0.74 \times 10^{-11}$ ) between the two data sets. The close match between the quantities of Pb additives calculated from fuel sales and the amounts reported at the US Senate Hearings by the Ethyl Corporation provide confidence in the calculation methods. The 1950 through 1982 period accounts for about 86% of the total mass of 5.37 million MT Pb used as a gasoline additive during the years from 1927 to 1994 and brackets the most intensive years of commercial sales of gasoline with Pb additives.

#### 3.2. 1982 lead estimates for 90 urbanized areas

The 1982 proportions of Pb for the 90 urbanized areas (18,222 MT) were nearly 30% of the total amount of Pb (61,526 MT) sold for the US consumption in the same year. The US EPA (1986) fate of Pb in the engine including the particle sizes of the Pb aerosols along with the quantity of Pb remaining in the engine system and the lubricating oil were also calculated for the 90 UAs.

#### 3.3. 1950–1982 estimates for ninety urbanized areas

Using the same proportions for each UA as calculated for 1982, the amount of Pb additive for the 90 urbanized areas for 1950 through 1982 is 1.37 million MT or about 30% of the 4.64 million MT Pb additive total in the US calculations of the particle sizes of Pb aerosols and Pb remaining in the engine or crankcase lubricant are given in parentheses.

Table 1 is a summary of the total quantity of Pb in MT (and in parenthesis the estimated mass of Pb aerosol emitted) for 1950 through 1982.

Table 2 lists the rank by metric tons of Pb additive estimated for each UA from highest (1) to lowest (90) for 1950 through 1982, and categorizes each of the UAs into quartiles according to the metric tons of Pb additives and aerosol emissions given in Table 1. The approximate location, rank of each of the UAs and symbols for each quartile are illustrated in Fig. 2.

## 4. Discussion

### 4.1. Limitations

Estimates for the Pb additives in 1982 were calculated from vehicle traffic data characteristics for each UA as a proportion of the total

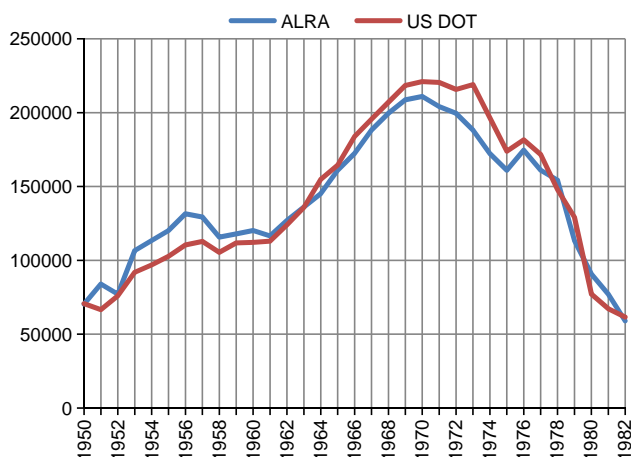


Fig. 1. Comparisons between Pb additives in gasoline by methods using state by state fuel consumption data from 1950 to 1982 (US DOT) and Ethyl Corporation data presented at the Airborne Lead Reduction Act (ALRA) hearings conducted by the US Senate in June, 1984 (US Senate 1984). All numbers on the left axis are in metric tons per year.

Table 1

Estimated mass of lead additives (and aerosol emissions) by a group of urban areas and percentiles for 90 urbanized areas from 1950 to 1982. See also Fig. 2. (In metric tons).

Percentiles	1st group	2nd group	3rd group	4th group
	Additives (aerosols)	Additives (aerosols)	Additives (aerosols)	Additives (aerosols)
Min	17,686 (13,265)	8403 (6302)	4662 (3497)	608 (456)
25%	24,859 (18,644)	9506 (7130)	5374 (4031)	1907 (1430)
50%	31,601 (23,701)	11,737 (8803)	5709 (4282)	3080 (2310)
75%	44,821 (33,616)	14,310 (10,733)	6802 (5102)	4055 (3041)
Max	149,938 (112,454)	16,623 (12,467)	8108 (6081)	4570 (3428)
N =	23	22	22	23

amount of Pb additives in 1982. The proportions were then applied to total Pb additives calculated from state by state gasoline sales from 1950 to 1982. The main limitation is a consequence of the assumption that the 1982 proportions of Pb for each UA are appropriate to apply to the 1950 through 1982 period for each of the 90 UAs. For example, Los Angeles UA grew more rapidly than most other UAs from 1950 to 1982 (Mielke et al., 2010). As a result, by applying the 1982 proportions to the entire 1950 through 1982 period, the estimates of Pb for the 1950s, 1960s and 1970s would be overestimated for the Los Angeles UA. An overestimate of Pb additives for the Los Angeles UA would result in an underestimate of Pb for the other UAs. However, the 1950 through 1982 Pb additive estimates shown in Tables 1 and 2 represent ranges of Pb additives and Pb aerosols inputs within various sized urbanized areas. These results are a reasonable estimate of the mass of Pb from gasoline associated with each UA and provide a beginning point for characterizing the effects of the inputs of gasoline Pb additives on the various UAs.

### 4.2. Characterization of the impact of gasoline Pb additives

General characterization of Pb additives must begin by recognizing the massive quantity of Pb mined and smelted for commercial use in gasoline. Lead is an element and when emitted as an aerosol it persists in the environment. Lead accumulates mainly in the loose surface materials (i.e. pedosphere) such as soils and/or stream and marine sediments. The Pb accumulated in soil will remain for hundreds of years (LaBelle et al., 1987). Han et al. (2002) estimated that by the year 2000, the cumulative global industrial production of Pb was about 235 million MT. Thus, during the short span of about 100 years, US paint and gasoline additives accounted for a combined total of 10–12 million MT of Pb, or about 5% of the several thousand year global history of anthropogenic Pb production.

#### 4.2.1. Pb aerosol particle sizes

Characterization of gasoline Pb additives requires a description of the physical characteristics of Pb aerosol particles. EPA (1986) estimated that 75% of Pb additives were emitted as exhaust. The tonnages of the relatively large  $>10 \mu\text{m}$  particles of Pb probably settled locally, especially in the urbanized areas where Pb in soils are elevated adjacent to roadways and decrease with distance away from the roadway (Laidlaw and Filippelli, 2008). Thirty-five percent of the particles were  $<0.25 \mu\text{m}$  size. The bulk of the smallest aerosol particles were emitted in the ultra-fine particle size, or less than  $0.1 \mu\text{m}$  diameter (Blom et al., 2000). The tonnages of the ultra-fine particles of lead were dispersed throughout the planet and deposited in oceans and on glaciers such as Greenland and the Antarctic where ice cores disclose a geochemical record of the atmospheric lead input from many centuries of industrial production (Han et al., 2002; Murozami et al., 1969).

However, a portion of the ultra-fine particles of Pb were also retained in urbanized areas. Because of their density, ultra-fine Pb particles tend to pierce through the boundary layer of air surrounding

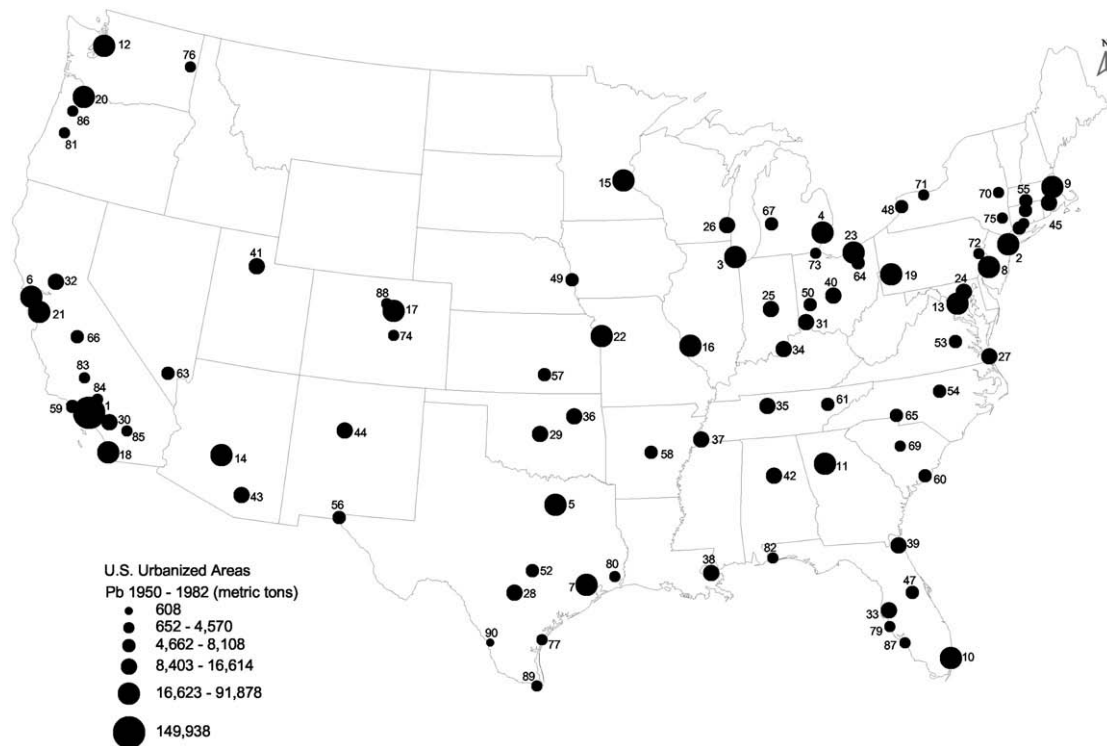
**Table 2**  
Urbanized areas listed and grouped by rank of lead additives during 1950–1982. See Fig. 2 for locations numbered by rank for the contiguous states, not including Alaska and Hawaii.

Group 1	Group 2	Group 3	Group 4
1 LA-Long Bch-Santa Ana CA	24 Baltimore MD	46 Bridgeport-Stamford CT-NY	68 New Haven CT
2 New York-Newark NY-NJ-CT	25 Indianapolis IN	47 Orlando FL	69 Columbia SC
3 Chicago IL-IN	26 Milwaukee WI	48 Buffalo NY	70 Albany-Schenectady NY
4 Detroit MI	27 Virginia Beach VA	49 Omaha NE-IA	71 Rochester NY
5 Dallas-Ft Worth-Arlington TX	28 San Antonio TX	50 Dayton OH	72 Allentown-Bethlehem PA-NJ
6 San Francisco-Oakland CA	29 Oklahoma City OK	51 Hartford CT	73 Toledo OH-MI
7 Houston TX	30 Riverside-San Bernardino CA	52 Austin TX	74 Colorado Springs CO
8 Philadelphia PA-NJ-DE-MD	31 Cincinnati OH-KY-IN	53 Richmond VA	75 Poughkeepsie-Newburgh NY
9 Boston MA-NH-RI	32 Sacramento CA	54 Raleigh-Durham NC	76 Spokane WA
10 Miami FL	33 Tampa-St. Petersburg FL	55 Springfield MA-CT	77 Corpus Christi TX
11 Atlanta GA	34 Louisville KY-IN	56 El Paso TX-NM	78 Anchorage AK
12 Seattle WA	35 Nashville-Davidson TN	57 Wichita KS	79 Sarasota-Bradenton FL
13 Washington DC-VA-MD	36 Tulsa OK	58 Little Rock AR	80 Beaumont TX
14 Phoenix AZ	37 Memphis TN-MS-AR	59 Oxnard-Ventura CA	81 Eugene OR
15 Minneapolis-St. Paul MN	38 New Orleans LA	60 Charlotte NC-SC	82 Pensacola FL-AL
16 St. Louis MO-IL	39 Jacksonville FL	61 Knoxville TN	83 Bakersfield CA
17 Denver-Aurora CO	40 Columbus OH	62 Honolulu HI	84 Lancaster-Palmdale CA
18 San Diego CA	41 Salt Lake City UT	63 Las Vegas NV	85 Indian-Cathdrl Cty-Plm Sprng CA
19 Pittsburgh PA	42 Birmingham AL	64 Akron OH	86 Salem OR
20 Portland OR-WA	43 Tucson AZ	65 Charleston-No. Chrliston SC	87 Cape Coral FL
21 San Jose CA	44 Albuquerque NM figure	66 Fresno CA	88 Boulder CO
22 Kansas City MO-KS	45 Providence RI-MA	67 Grand Rapids MI	89 Brownsville TX
23 Cleveland OH			90 Laredo TX

surfaces, collect on buildings, painted and unpainted, (along with trees and other surfaces), and wash into the soil below (Mielke et al., 1984; Mielke, 1999; Rolfe and Haney 1975; US EPA, 1986). Lead in soils within non-smelter urbanized areas is derived from a mixture of Pb from paint and gasoline with a ratio that is spatially variable dependent on the proximity of roadways and/or age, condition, and maintenance of homes with exterior Pb paint (Wu et al., 2010). The characteristics of fine and ultra-fine particles of Pb collecting on building surfaces assists with understanding why, regardless of the building materials such as brick vs. painted siding, the soils next to

buildings are generally more contaminated than soils away from buildings (Button, 2008; Clark et al., 2006; Linton et al., 1980; Mielke et al., 1984; Mielke, 1999; Schmitt et al., 1988).

Another comparison of Pb from paint and vehicle source was from a study that weighed the paint scraped from an old house and found that if the same house had been power-sanded, 7.4 kg of Pb would have been released as fine particles (Mielke et al., 2001). A substantial amount, considering that the total tolerable daily intake (TTDI) for a child under 6 years of age is 6 µg Pb from all sources (Ross et al., 2000). Converting 7.4 kg Pb to dust means an equivalent of more than



**Fig. 2.** Estimated lead aerosol inputs from gasoline into 90 US urbanized areas (UA) from 1950 to 1982. The numbers on the map are rankings of each UA. The size of each dot refers to the quartile for each group of UAs reported in Table 2, with the extremes, Los Angeles UA ranked 1 and Laredo, Texas ranked 90. Some of the UAs have been sites of soil Pb studies as indicated in Table 3.

1 billion ( $10^9$ ) times the TTDI. For perspective with vehicle traffic related emission, the one-time release of  $7.4 \times 10^9 \mu\text{g}$  of Pb dust from power sanding compares with the annual emission of  $50 \times 10^9 \mu\text{g}$  of Pb dust per 0.1 mile (0.16 km) during the peak use of leaded gasoline from the nearby arterial street (Mielke et al., 2001).

#### 4.2.2. Bioavailability of Pb particles

An important characteristic of anthropogenic Pb deposited in soil is that it is speciated in easily absorbed carbonate, iron, and manganese hydroxide soil fractions, whereas natural Pb in soils consists of the non-bioavailable portion and is speciated in the residual fractions (Chlopecka et al., 1996; Lee et al., 1997). This means that soils contaminated by anthropogenic Pb aerosols from gasoline and paint are more bioavailable than the small portion of Pb associated with natural soils. This observation is supported by the ease of extraction of Pb from soils with a relatively mild 1 molar nitric acid at room temperature (Mielke et al., 1983, 2005a). This extraction gives similar results for Pb as boiling concentrated nitric acid extraction methods (US EPA, 1996b). In Los Angeles California, analysis of 550 soil samples indicated that total Pb (extracted using boiling concentrated nitric acid) and bioavailable Pb were highly correlated ( $r = 0.96$ ) (Wu et al., 2010).

The bioavailability and toxicology characteristics of Pb are related to particle size. The toxicity of Pb additives was indicated by the phenomenally rapid decline of blood Pb, especially for children, when the Pb additives were removed from gasoline (Annest et al., 1982). During the rapid phase-down beginning in January, 1986, the US childhood prevalence of blood Pb decreased by 77% (Pirkle et al., 1994), and similarly rapid declines of human blood Pb were experienced by every nation that eliminated Pb additives from gasoline (Thomas et al., 1999). Thus, gasoline Pb aerosols consisted of fine to ultra-fine particles which, after deposition, are easily extracted from soils, highly bioavailable and very toxic to humans, especially children.

#### 4.3. US background and urban soil lead concentrations

Soils are the most accessible of the loose materials, or pedosphere, of the Earth. The United States Geological Survey undertook a survey of soil ( $n = 1323$ ) to establish the background metals in the US and found that the background soil Pb median is 16.5 mg/kg and ranges from 10.3 to 30.1 mg/kg (Gustavsson et al., 2001 p. 22). Even smaller quantities of Pb were identified in fresh alluvium from the Mississippi River, the parent materials of the urbanized area of New Orleans (Mielke et al., 2000). Thus, natural soils contain relatively small quantities of Pb. The practical meaning of this finding is that all UAs have an available source of low Pb soils nearby.

In contrast to background soil lead, significantly larger quantities of soil Pb are reported in urbanized areas. The remnants of Pb additives in gasoline are observed by studying the variations of Pb content of the pedosphere. Updating a review of urban soil Pb studies by Burgoon et al. (1995) assists with characterizing the material impact that Pb additives to gasoline may have had on environments and the health of inhabitants of urbanized areas. Table 3 is a list of soil Pb studies sorted by rank and group of urbanized areas according to the estimated quantity of Pb additives and Pb emissions, given in Tables 1 and 2, and illustrated in Fig. 2. Whenever possible, the number of samples, the median (or geometric mean), and the ranges are given in Table 3.

#### 4.4. Characteristics of soil lead in urbanized areas

In Table 3, the highest density collections for urbanized areas to date have been conducted in Baltimore MD, cities of Minnesota, Milwaukee WI, cities of Louisiana (especially New Orleans), Illinois parks, Syracuse NY, and Detroit MI. One protocol developed in Minnesota for high density sampling involved stratifying samples by

enumeration districts) and collecting samples along residential streets, near foundations of residences, and from open spaces (Mielke, 1991, 1994; Schmitt et al., 1988). The protocol was revised to include busy streets in the second survey in metropolitan New Orleans, LA, where the median N of soil collection was 19 samples per  $\text{km}^2$  (Mielke et al., 2005a). The following discussion lists six characteristics of UAs that were derived empirically from the high density sampling studies of urbanized areas.

#### 4.4.1. Soil Pb, city size, inner-city vs. outer-city location

Larger UAs are locations where vehicle traffic input larger quantities of Pb aerosols than vehicle traffic input in smaller UAs. As a result, the storage of Pb in soils in larger cities is higher than storage of Pb in soils of smaller towns, a characteristic observed in Table 3 for Minnesota, Louisiana and Michigan. It is important to note that median refers to the middle Pb result of a given set of soil Pb results. The number (N) of samples collected for each of the cities ranged from 38 for Pontiac to 5467 for Survey 2 of New Orleans (see Table 3). In Table 3 the progression of median soil Pb (from larger to smaller cities) of Minneapolis, Saint Paul, Duluth, Saint Cloud and Rochester is a median of 230, 170, 144, 41 and 25 mg/kg Pb, respectively (Mielke et al., 1984/85, 1989, 1997; Mielke, 1993; Schmitt et al., 1988). Table 3 also shows that the median soil Pb for New Orleans, the largest Louisiana city, is in the  $>200$ – $399$  mg/kg range while the median soil Pb in the small community of Lafourche is  $<25$  mg/kg (Mielke et al., 1997). The median soil Pb concentration of Metro New Orleans soil: Survey 1 = 167 mg/kg ( $N = 4026$ ), Survey 2 = 112 mg/kg ( $N = 5467$ ) (Mielke et al., 2005a). In Michigan, the median soil Pb for Detroit is 189 mg/kg while in the smaller city of Pontiac the median soil Pb is 86 mg/kg (Mielke et al., 2003).

Furthermore, soils of inner-city communities contain larger quantities of Pb than outlying communities of the same city. This was noted in a soil survey of urban gardens of Baltimore and then in Minneapolis-St. Paul (Mielke et al., 1983, 1984). LaBelle (1986) studied soil Pb in parks where buildings with Pb-based paint do not generally exist and noted that parks of inner-city Chicago, suburbs, and rural areas, have median soil Pb of 262, 87, and 37 mg/kg respectively. The soil in inner city of Milwaukee has a median soil Pb of 240 mg/kg while the suburban communities have a median Pb of 50 mg/kg (Brinkmann 1994b). The footprint of soil Pb in UAs requires taking into account both lead-based paints and nonpaint sources in order to comprehend the input of Pb in various sized cities and different locations within the same region (Mielke et al., 2001, 2008). LaBelle et al. (1987) studied soils that were not likely to be impacted by buildings or industrial sites and noted that Pb in Illinois soils vary widely depending on traffic density, distance from the busiest nearby roadway, and traffic volume. Laidlaw and Filippelli (2008) reviewed the quantity and distribution of soil Pb in numerous cities of North America, and to date, all urbanized areas exhibit the same city size and community location characteristics that have been described above.

#### 4.4.2. Soil is an enormous Pb dust reservoir

Children characteristically expose themselves to Pb dust via hand-to-mouth activity (Sayre et al., 1974; Ko et al., 2007). Children's hands tested with wipes at childcare centers before and after outdoor play in New Orleans indicated that children had more Pb after playing outside than after playing inside (Viverette et al., 1996). If the children engaged in hand-to-mouth behavior at private inner-city childcare centers the amount of Pb per hand exceeded the  $6 \mu\text{g}$  Total Tolerable Daily Intake (TTDI) of Pb by a factor of 5 or more; public ("Head Start") childcare centers, where play areas are covered with rubberized playground surfacing, children's hands did not exhibit Pb differences before and after outdoor play (Viverette et al., 1996). Another study by Nielsen and Kristiansen (2005) compared exposure to Pb on

**Table 3**  
Soil lead studies in the US. Median Pb results in mg/kg. See Tables 1, 2 and Fig. 2.

	N	Min	Med	Max	Reference	
Background soil Pb, US	1319		16.5		Gustavsson et al. (2001)	
<i>Rank</i>	<i>City-State</i>					
1-1st	Los Angeles, California	550	9	216,174	Wu et al. (2010)	
1-1st	Los Angeles, California	343			Sutton et al. (1995)	
1-1st	Chicago Illinois	57			Cannon and Horton (2008)	
3-1st	Chicago Illinois	667			LaBelle et al. (1987)	
3-1st	Chicago-Urban Parks	255	12	262	1312	LaBelle (1986)
3-1st	Chicago-Suburban Parks	245	12	87	1637	LaBelle (1986)
	Illinois, Rural Parks	177	12	37	937	LaBelle (1986)
4-1st	Detroit, Michigan	59	13	189	1345	Mielke et al., 2003, Supplement
4-1st	Detroit-Suburbs, Michigan	76	4	16	810	Mielke et al., 2003, Supplement
	Pontiac, Michigan	38	15	86	495	Mielke et al., 2003, Supplement
6-1st	Oakland, California	358	7		347,900	Sutton et al. (1995)
6-1st	Alameda, California	138	22		3187	Teichman et al. (1993)
9-1st	Boston, Massachusetts	195	7		13,240	Rabinowitz and Bellinger (1988)
10-1st	Miami, Florida	240	2		1060	Chirenje et al. (2004)
12-1st	Seattle, Washington	51	150		74,000	Roberts et al. (1991)
13-1st	Washington, D.C.	240	12		6015	Elhelu et al. (1995)
15-1st	Mpls/St Paul, Minnesota	90	5		7650	Mielke et al. (1984)
15-1st	Minneapolis, Minnesota	898	1	230	20,136	Schmitt et al. (1988)
15-1st	St. Paul, Minnesota	832	1	170	7994	Schmitt et al. (1988)
	Duluth, Minnesota	229	1	144	11,110	Schmitt et al. (1988)
	St. Cloud, Minnesota	124	1	41	1952	Schmitt et al. (1988)
	Rochester, Minnesota	165	1	25	1930	Schmitt et al. (1988)
	Outstate farms Minnesota	781	1	31	7111	Schmitt et al. (1988)
23-2nd	Cleveland, Ohio	50	19		811	Petersen et al. (2006)
24-2nd	Baltimore, Maryland	122	0.01		5620	Yesilonis et al. (2008)
24-2nd	Baltimore, Maryland	422	1	100	10,900	Mielke et al. (1983)
25-2nd	Indianapolis, Indiana	116	46		565	Filippelli et al. (2005)
26-2nd	Milwaukee, WI summary data	372	1	160	880	Brinkmann (1994b)
26-2nd	Milwaukee cent city + No and So Side	256	1	240	7220	Brinkmann (1994b)
26-2nd	Milwaukee, WI suburbs	122	1	50	1780	Brinkmann (1994b)
32-2nd	Cincinnati, Ohio	60	2		3166	Leeuwen et al. (1990)
31-2nd	Cincinnati, Ohio-Childcare centers	69	17		4636	Button (2008)
31-2nd	Cincinnati, Ohio	60				Tong (1990)
32-2nd	Sacramento, California	232	57	229	320,834	Sutton et al. (1995)
33-2nd	Tampa, Florida	146	<1	100	9160	Brinkmann (1994a)
38-2nd	New Orleans, LA Survey 1	4026	18	134	183,588	Mielke et al. (2005a)
38-2nd	New Orleans, LA Survey 2	5467	3	100	52,798	Mielke et al. (2005a)
38-2nd	LA Orleans Parish	~1540	<25	>200	<1000	Mielke et al. (1997)
	LA Lafourche Parish	~190		<25		Mielke et al. (1997)
44-2nd	Albuquerque, New Mexico	43	3		5280	Franz and Hadley (1981)
49-3 rd	Omaha, Nebraska	176				Angle and McIntire (1979)
50-3 rd	Dayton, Ohio	22	22		461	Ritter and Rinefierd (1983)
56-3 rd	El Paso, Texas	94	<20		8700	Pingitore et al. (2009)
62-3 rd	Honolulu, HI	18				Fu et al. (1988)
65-3 rd	Charleston So. Carolina	164	9		7890	Galke et al. (1975)
68-4th	New Haven, Connecticut	487	30		7000	Stark et al. (1982)
77-4th	Corpus Christi, Texas	485	21		2969	Harrison (1987)
	Pueblo, Colorado	33				Diawara et al. (2006)
	Connecticut	174	<10		2200	Stilwell et al. (2008)
	Gainesville, Florida	202	2.13		1091	Chirenje et al. (2004)
	Champaign Illinois	116	20		1061	Solomon and Hartford (1976)
	Louisiana and Minnesota	6,342	Urban–rural comparisons.			Mielke (1993)
	S.E. Michigan	171	3		7400	Murray et al. (2004)
	Mt. Pleasant, Michigan	189	100		16,839	Francek (1992)
	Syracuse, New York	2998	45			Griffith et al. (2009)
	Syracuse, New York	162				Griffith (2002)
	Syracuse, New York	194		80 (GM)		Johnson and Bretsch (2002)
	Lubbock, Texas	52		35		Brown et al. (2008)
	Maine urban soils	100				Kruger and Duguay (1989)

children's hands at three kindergartens before and after remediation. One kindergarten had very small amounts of Pb in soil (control) and two kindergartens had soil Pb concentrations of 100–200 mg/kg. Following remediation of the 100–200 soil Pb to below 10 mg/kg the study found a good agreement (with some overlap) between the average concentration of Pb in soil and the amount of Pb on the hands of the children (Nielsen and Kristiansen, 2005).

One critical characteristic is Pb loading of the soil surface. For example, in a study in New Orleans measuring the Pb loading of the surface of bare soil it was noted that a soil containing 400 mg/kg (the current EPA standard for soil Pb) exceeds the interior floor guideline of Pb loading (currently 40  $\mu\text{g}/\text{ft}^2$ ) by a factor of about 35 (Mielke et al., 2007a). Children are also exposed to soil Pb by track-in of contaminated soil into homes on shoes (Hunt et al., 2006) family pets,

and resuspension and deposition of Pb contaminated urban soil dust which penetrates interiors of homes and settles onto contact surfaces (Laidlaw and Filippelli, 2008; Layton and Beamer, 2009; Paustenbach et al., 1997; Polidori et al., 2009). Considering the quantity of Pb loading on the soil surface, it is easy to understand how Pb contaminated exterior soil results in children picking up Pb on their hands as well as the large potential for track-in of exterior Pb into building interiors.

#### 4.4.3. Association of soil Pb and blood Pb

Soil Pb concentrations have been associated with children's blood Pb concentrations using multiple study designs—cross-sectional, ecological spatial, ecological temporal, prospective soil removal, and isotopic studies (Laidlaw and Filippelli, 2008). Comparing various communities, both soil Pb and blood Pb of children living in smaller towns' exhibit lower blood Pb than children living in larger cities (Mielke et al., 1989, 1997; Mielke and Reagan 1998). To observe this characteristic requires full spectrum blood Pb surveillance rather than truncated blood Pb at an arbitrary level such as 10 or 15  $\mu\text{g}/\text{dL}$ . In New Orleans the blood Pb response of children to soil Pb is curvilinear (Mielke et al., 1997, 1999, 2007b). Johnson and Bretsch (2002) observed a similar curvilinear relationship between soil Pb and children's blood Pb in Syracuse, NY. The most recent New Orleans urban soil Pb and blood Pb study shows the following results: below 100 mg/kg soil Pb children's blood Pb response is steep at 1.4  $\mu\text{g}/\text{dL}$  per 100 mg/kg, between 100 mg/kg and 300 mg/kg the curve flexes, and above 300 mg/kg soil Pb children's blood Pb response becomes gradual 0.32  $\mu\text{g}/\text{dL}$  per 100 mg/kg (Mielke et al., 2007b). Soil Pb levels are also associated with a risk of blood Pb concentrations greater than 10  $\mu\text{g}/\text{dL}$  at soil Pb concentrations lower than indicated by soil Pb–blood Pb curves (Mielke et al., 2007b). For example, Malcoe et al. (2002) found that logistic regression of yard soil Pb > 165.3 mg/kg was associated independently with blood Pb greater than or equal to 10  $\mu\text{g}/\text{dL}$  (OR, 4.1; CI, 1.3–12.4). Similarly, the Texas Department of Health (2004) used a large database from El Paso, Texas Area and found an odds ratio 4.5 (1.4, 14.2) for the relationship between a 500 mg/kg increase in soil Pb above the background and blood Pb level > 10  $\mu\text{g}/\text{dL}$ . Similar soil Pb and blood Pb responses of children are expected as a general characteristic in other UAs of the US. The close connection between children and their environment is one of the critical qualities shared as part of the developmental processes of children (Louis et al., 2006).

#### 4.4.4. Blood Pb seasonality and Pb dust resuspension

Lead dust is so ubiquitous, especially within the 1st and 2nd group of urbanized areas (Tables 1 and 2), that seasonal wet–dry weather patterns characteristically influence the blood Pb of children. This characteristic was described by Laidlaw et al. (2005) for three cities with climates as different as Syracuse, NY, Indianapolis, IN, and New Orleans, LA. In all three cities blood Pb increased during droughty periods when soil is dusty and decreased blood Pb during rainy periods when soil dust is settled (Laidlaw et al., 2005). Similarly, in Mexico City, Rosas et al. (1995) observed that during rainy seasons of the year,  $\text{PM}_{10}$  dust was settled and atmospheric Pb concentrations were lower; during seasons with low rainfall  $\text{PM}_{10}$  and atmospheric Pb concentrations increased. In Milwaukee, Wisconsin, blood Pb levels also follow a seasonal pattern with peaks in the summer and autumn (Havlena et al. 2009). They observed that particulate matter less than  $\text{PM}_{2.5}$  correlated with the seasonal variation in 10 month old children's blood Pb, and suggested that the Pb in the  $\text{PM}_{2.5}$  was causally related to seasonal variations in children's blood Pb levels.

The characteristic of Pb contaminated soil being resuspended is supported by Pingitore et al. (2009) who used synchrotron-based XAFS (X-ray absorption fine structure) to quantify the Pb species in the air of El Paso. Pingitore et al. (2009) concluded that the soil is the dominant source of Pb, and that it was being resuspended as aerosols

into the atmosphere. Harris and Davidson (2005) calculated that annually at least 54 MT of Pb aerosol particles in the Southern California Air Basin were being resuspended from soils contaminated by previous Pb additives in gasoline. Climates differ markedly between the UAs shown in Fig. 2, and children's blood Pb responses to soil Pb are expected to be related to local weather factors that affect resuspension in each UA.

Pb dust in urban environments can result in elevated lead loading of both interior and exterior contact surfaces (Caravanos, et al., 2006a, b). Lead loading (mass/unit area) is well known to correlate with urban children's blood lead levels. However, exterior lead loading is not routinely measured in US cities, and is likely a better measure of risk to children blood lead levels than air lead concentrations (mass/volume). Caravanos et al. (2006b) demonstrated that exterior lead loading in the five boroughs of New York City was highly elevated when compared to the HUD/EPA indoor Pb in dust standard of 40  $\mu\text{g}/\text{ft}^2$ . Caravanos et al. (2006b) measured the following median dust loadings in New York: Brooklyn (730  $\mu\text{g}/\text{ft}^2$ ), Staten Island (452  $\mu\text{g}/\text{ft}^2$ ), the Bronx (382  $\mu\text{g}/\text{ft}^2$ ), Queens (198  $\mu\text{g}/\text{ft}^2$ ) and Manhattan (175  $\mu\text{g}/\text{ft}^2$ ). In a related study, Caravanos et al. (2006a) demonstrated how exterior particulate Pb can accumulate rapidly on interior surfaces. They observed that interior settled dust in a Pb-free room with a window slightly open exceeded the HUD/EPA indoor Pb in dust standard of 40  $\mu\text{g}/\text{ft}^2$  (43  $\mu\text{g}/\text{m}^2$ ) within a 6 week period.

#### 4.4.5. Association between blood lead <10 $\mu\text{g}/\text{dL}$ and children's clinical health outcomes

Low blood lead levels are also associated with a reduction in children's intelligence. Canfield et al. (2003) observed that when lifetime average blood lead concentrations in children increased from 1 to 10  $\mu\text{g}/\text{dL}$ , the intelligence quotient (IQ) declined by 7.4 points. Jusko et al. (2008) observed that compared with children who had lifetime average blood lead concentrations <5  $\mu\text{g}/\text{dL}$ , children with lifetime average concentrations between 5 and 9.9  $\mu\text{g}/\text{dL}$  scored 4.9 points lower on Full-Scale IQ (91.3 vs. 86.4,  $p=0.03$ ). Similarly, Surkan et al. (2007) observed that children with 5–10  $\mu\text{g}/\text{dL}$  had 5.0 (S.D. 2.3) points lower IQ scores compared to children with blood lead levels of 1–2  $\mu\text{g}/\text{dL}$  ( $p=0.03$ ). Interestingly, multiple studies have shown that the strongest lead effects on IQ occurred within the first few micrograms of blood Pb (Schnaas et al., 2006; Canfield et al., 2003; Lanphear et al., 2005). Low blood lead levels have also been associated with various physiological outcomes such as kidney damage (Fadrowski et al., 2010), puberty delay in boys (Williams et al., 2010) and girls (Selevan et al., 2003) and cardiovascular outcomes in adults (Navas-Acien et al., 2007).

#### 4.4.6. Soil Pb, blood Pb and school achievement

Lower blood Pb levels (<10  $\mu\text{g}/\text{dL}$ ) typically associated with urban soil Pb exposure are now being associated with health outcomes related to subtle nervous disorders. Needleman et al. (1979) identified the impact of children's early Pb exposure to classroom performance. Low blood Pb <10  $\mu\text{g}/\text{dL}$  is associated with ADHD (Nigg et al., 2010) and a reduction in children's test scores for reading (OR 0.51,  $p=0.006$ ) and writing (OR 0.49,  $p=0.003$ ) and mathematics (Chandramouli et al., 2009; Miranda et al., 2007). Reduced student performance has been demonstrated as one of the characteristics of the accumulation of Pb dust in communities of UAs. The literature of the effects of Pb on student performance is large (Martin, 2008).

Support for the idea that soil Pb in a community is associated with student performance was indicated in Minnesota by the observation that dropout rates of high school students followed city size and soil Pb (Mielke et al., 1989). In New Orleans, soil Pb on the play areas of elementary public schools follow the same trend as the urban soil Pb map of the entire city; however, soils on school grounds are significantly less Pb contaminated than soils of neighboring residential properties where the most vulnerable preschool children reside

and play before they attend school (Higgs et al., 1999). New Orleans school achievement test scores were found to be associated with soil metals including Pb (Mielke et al., 2005b; Mielke and Berry, 2007, pp 116–123). Finally, standardized test performance of children of local schools is significantly linked with soil Pb and blood Pb of the same school district and this phenomenon indicates the serious neurotoxicity effects of Pb (Zahran et al., 2009). These studies characterize the more insidious disparities and costly influences that environmental Pb imposes on urban society (Campanella and Mielke, 2008; Chandramouli et al., 2009; Gould, 2009; Muennig, 2009; Nigg et al., 2008; Zahran et al., 2009).

#### 4.5. Precautionary approaches for primary exposure prevention

As described in the sections above, the current consensus is that for children a blood Pb of 5 µg/dL or even 2 µg/dL will cause clinical signs. The critical problem for children Pb is to successfully curb the blood Pb in the 2–10 µg/dL range which, as reported by Gould (2009), currently prevails among 24.5%, or 9.6 million US children. To reduce the Pb exposure to meet lower guidelines, large US urbanized areas will probably require extensive environmental treatment, and soil Pb is one factor that can be changed to reduce children's Pb burdens (Filippelli and Laidlaw, 2010). Fortunately, as described previously clean soil with a median Pb content of 16.5 mg/kg is available nearby all US urbanized areas (Gustavsson et al., 2001).

In Minneapolis a pilot project was conducted that successfully reduced children's exposure and prevented the expected seasonal summertime blood Pb increases (Mielke et al., 1992). The method was refined in New Orleans where abundant and remarkably clean (median ~5 mgPb/kg) Mississippi River sediments are available as a natural resource for covering Pb contaminated urban soils (Mielke, 2005; Mielke et al., 2006a,b). Other pilot projects focused on soil Pb were conducted in Boston and Chicago with varying success (US EPA, 2001; Binns et al., 2004).

A full-scale national program is underway in Norway to clean up contaminated soils at all childcare centers, elementary schools and parks in the ten largest cities (Ottesen et al., 2008). The differences between the US approach and Norway's program are striking: in the US secondary prevention is conducted after a child is identified with elevated blood Pb (CDC blood Pb guideline is  $\geq 10$  µg/dL, or even higher depending on local health department jurisdictions). In Norway, the WHO principles of the precautionary approach are followed (Louis et al., 2006). Therefore, the emphasis is on primary prevention whereby environmental contaminants are directly addressed and treated; children's blood Pb samples are not included as part of the protocol. In the US the current residential soil guideline is 400 mg/kg Pb for the bare soil of a play area, and 1200 mg/kg for the remaining areas of the property. In Norway the soil Pb guideline for play areas is 100 mg/kg, and there is discussion about reducing the guideline to 60 mg/kg.

Research in New Orleans indicates that communities with a median soil Pb  $\leq 80$  mg/kg would generally prevent children from reaching a blood Pb  $\geq 10$  µg/dL (Mielke et al., 1999). If a stricter blood Pb guideline is observed that reflects the current clinical health effects of Pb, then the soil Pb guideline must also be significantly lower. As shown in Table 3, many US urbanized areas have soil Pb medians above 100 mg/kg. Norway's precedence for implementing a national clean play area program serves as a model for commitment to primary prevention that benefits children and ultimately all community members of Pb impacted urbanized areas (Ottesen et al., 2008).

## 5. Conclusions

The principal message from the Pb emission estimates for 90 urbanized areas is that precaution must accompany the selection of

fuel additives used in urban-industrial society. Lead additives in fuel created the situation whereby vehicles traveling on roadways functioned as a Pb dust delivery system into the most populated communities of urbanized areas. As a result, the commercial use of Pb additives in gasoline had the consequence of materially contaminating urbanized areas, with 100's to over 100,000 MT of Pb dust, depending directly on vehicle traffic patterns in urbanized areas. Although Pb additives were removed from vehicle fuels for highway use, the mass of Pb dust inputs continue to materially impact the environment. Lead dust is associated with an array of chronic health problems of the people living in urbanized areas.

Lead-based paint coating is visible and presents a hazard when it deteriorates or is treated in a manner that creates Pb dust. Past use of Pb additives in gasoline resulted in ~75% emitted as tiny Pb aerosols particles that were essentially invisible. Empirical means for visualizing the material impact of both paint and nonpaint sources of Pb on urbanized areas involves soil Pb mapping. High density soil Pb mapping (i.e., ~19 samples per km<sup>2</sup>) has been conducted in a few cities, but as indicated in Table 3, most urban soil Pb surveys have insufficient sample density to guide intervention efforts. All US cities are in close proximity to soils containing 10.3–30.1 mg/kg Pb thereby making intervention with clean soils possible for every city.

Young children have no possibility of creating safe environments for themselves—this is an adult responsibility. Characterization of Pb additives in gasoline and its legacy of effects on urbanized areas should result in better understanding of actions needed for hazard control to protect communities from the health effects of both paint and nonpaint sources of Pb.

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