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## Review

## Resuspension of urban soils as a persistent source of lead poisoning in children: A review and new directions

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## ABSTRACT

Urban soils act as the repository for a number of environmental burdens, including Pb. Significant attention has been devoted to reducing Pb burdens to children with outstanding success, but the fact that blood Pb levels above 10  $\mu\text{g}/\text{dL}$  are disproportionately found in children living in many USA cities (15–20% in some cities compared to a national average of less than 2%) indicates that not all of the sources have been eliminated. Although the health risk of fine particulates has begun to raise concerns in cities, little attention has been paid to Pb associated with these particulates and the potential role of this pathway for continued Pb burdens of urban youth. This review summarizes recent work on particulate resuspension and the role of resuspension of Pb-enriched urban soils as a continued source of bio-available Pb both outside and inside homes, then presents recent efforts to model Pb burdens to children based on the atmospheric parameters that drive particulate resuspension. A strong seasonal relationship is found between atmospheric particulate loading and blood Pb levels in children, and new particulate loading models are presented for a range of US cities involved in the Interagency Monitoring of Protected Visual Environments (IMPROVE) program. These seasonal particulate loading models have implications for a number of respiratory health impacts, but can also be used to calculate seasonal patterns in bio-available Pb redistribution onto contact surfaces (the primary pathway for ingestion-related uptake in toddlers) and assist clinicians in interpreting time-specific blood Pb tests.

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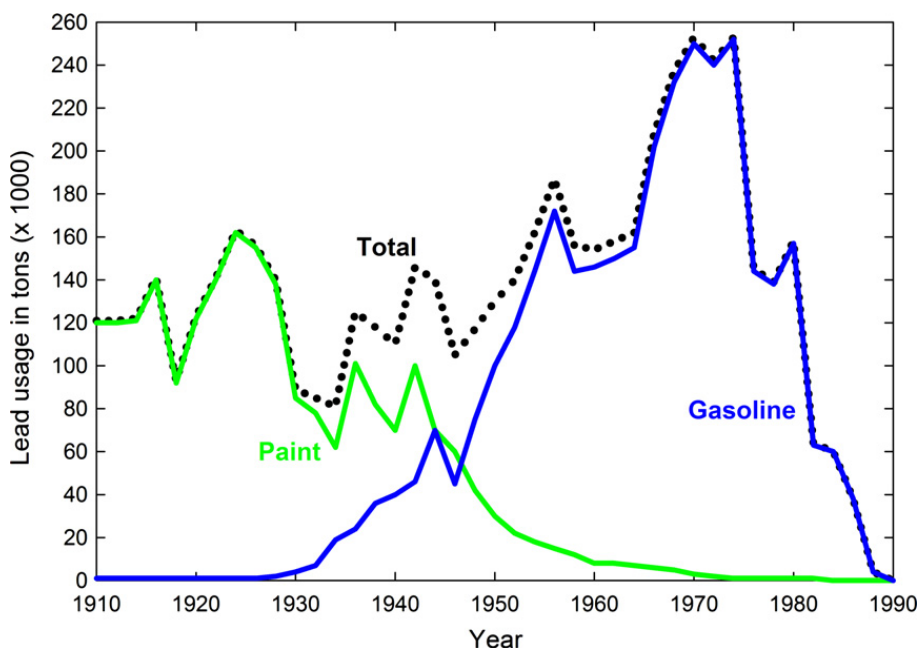
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**1. Introduction**

Compared to other chemicals of environmental concern, the uptake mechanisms and toxicological effects of Pb are relatively well understood. Lead exhibits a clear industrial legacy, with the millions of tons of anthropogenically sourced Pb used in Pb-based paints and leaded gasoline (Fig. 1) still largely present in the surface environment. The major pathway of Pb uptake in humans is via ingestion, where Pb is absorbed in the intestine and incorporated in the body (e.g., Maddaloni et al., 1998; Manton et al., 2001). Age is a strong variable in Pb absorption and metabolism, with the portion of ingested Pb that is taken up in the body typically being less than 5% for adults but as high as 50% for children due to their less-developed gastrointestinal pathway (Ziegler et al., 1978; Roberts et al.,

2001). Once absorbed, Pb moves through a number of biological systems in the body. First, due to similar charges and ionic radii, Pb is utilized in biological processes much like Ca, including as a critical component of converting the electrical neural signal into a chemical signal and as a component of the key mineral component of bone formation (Rothenberg et al., 2001). If the Pb is deposited in a neuron, it does not function as a neurotransmitter like Ca does, effectively creating permanent neural differentiation defects resulting in mental retardation, learning disorders, and attention-deficit/hyperactivity disorder (ADHD; Nigg et al., 2008). Because of children’s high ingestion efficiency and the rapid neural differentiation during early brain and nervous system development, they are especially vulnerable to permanent effects of Pb poisoning. When Pb is incorporated in bone, the bone becomes a longer-term source of



**Fig. 1.** History of Pb usage in paints and in gasoline during most of the 20th century, showing the early dominance of Pb-based paints followed by the boom in transportation resulting in a high use of leaded gasoline (after Mielke, 1999). The decline after the mid-1970s was due controls put into place to eliminate leaded gasoline.

Pb to the biological system – bone is regenerated on time-scales of months to years, resulting in a continued internal source of Pb to blood. For this reason, children treated by medical interventions like blood chelation may continue exhibiting toxic levels of Pb in their blood. Furthermore, as neither the placenta nor mammary glands are a perfect barrier to Pb, pregnant and lactating mothers with elevated blood Pb levels may themselves pose a health risk to babies and fetuses.

The health standards for Pb levels in blood have been steadily revised downward over the years as medical research has determined toxicological effects of Pb in even low quantities. The US Centers for Disease Control and Prevention (CDC) in 1991 chose 10  $\mu\text{g}/\text{dL}$  as an initial screening level for Pb in children's blood, although subsequent studies are still unable to find a "safe" lower level of Pb, with levels below 10  $\mu\text{g}/\text{dL}$  still causing some toxicological effects (e.g., Nigg et al., 2008). Recent epidemiology studies have found that blood Pb levels below 10  $\mu\text{g}/\text{dL}$ , a blood Pb level previously thought safe, can result in significant cognitive impairment in children (Canfield et al., 2007; Chiodo et al., 2007; Jusko et al., 2007; Schnaas et al., 2006; Surkan et al., 2007; Miranda et al., 2007). The National Health and Nutrition Examination Survey (NHANES) III 1999–2002 database indicates that approximately 2.4 million children have blood Pb levels between 5 and 9.9  $\mu\text{g}/\text{dL}$  (Iqbal et al., 2008) and that within that population of 1–5-year olds with blood Pb levels of 5  $\mu\text{g}/\text{dL}$  or higher, the prevalence was 47% for non-Hispanic Black children, 28% for Mexican American children, and 19% for non-Hispanic White children (Bernard, 2003). The fact that children of color are nearly 4 times more likely than white children to have blood Pb levels between 5 and 10  $\mu\text{g}/\text{dL}$  (and 13 times more likely to have blood Pb levels above 20  $\mu\text{g}/\text{dL}$ ; Bernard and McGeehin, 2003) raises concerns about social justice and the long-term health of these children. The full spectrum of toxicological effects of Pb in the human system is still not known, and deserves further study. But the persistent presence of Pb in children is a public health issue of a first order. Sources of Pb that could contribute to acute Pb poisoning have been highly publicized in the media, with the focus on consumer product safety (e.g., toys with Pb-based paints from China) and seriously degraded Pb-based paints in dilapidated homes. The continued source of chronic low levels of Pb to children, however, is not always easy to constrain, and is most easily assessed by examining environmental loading of Pb from multiple sources.

Lead loading is defined as the mass of Pb per unit area, usually expressed as  $\mu\text{g}/\text{ft}^2$  (0.093  $\text{m}^2$ ) or as a Pb loading rate as  $\mu\text{g}/\text{ft}^2$  (0.093  $\text{m}^2$ )/time. Lead concentration is expressed as mg/kg or ppm. Lead loading inside homes is correlated with blood Pb concentrations, while Pb concentration of dust is not correlated with blood Pb concentration (Lanphear et al., 1995; Sterling et al., 1999). It is possible to have a high Pb concentration in dusts inside the home, but a low Pb exposure (dose) to inhabitants of the home if the amount of dust is low. Alternatively, in a home with a large amount of dust with a low to moderate concentration, children's hand to mouth exposure (dose) may be high. While this concept is well understood, there

is debate about the source of the Pb inside homes. A percentage of the public health establishment hypothesizes that Pb loading inside homes originates primarily from the deterioration of Pb based paints – "the paint-only hypothesis". However, there is a large body of literature that argues that an appreciable percentage of the source of elevated Pb loadings inside homes originates from an enormous reservoir of Pb contaminated dusts and soils that exists outside the home and is transported inside the home via soil resuspension (Laidlaw et al., 2005) and tracked in via Pb contaminated dust and soil attached to shoes (Hunt et al., 2006) and on the feet of family pets. This outdoor reservoir is considered a combination of Pb in soils from past use of leaded gasoline and Pb-based paints that have deteriorated from exterior paints. The debate about the source of Pb in interior dust is critical because Pb poisoning in children cannot be eliminated until the primary source has been identified and eliminated (Elwood, 1984; Kurkjian and Flegal, 2003). But as with many controversies, both "sides" may be right; the authors suggest that neither the "paint-only" nor the "soil only" hypotheses are supported, but instead that interior paint AND Pb-enriched soils are both harmful sources of Pb to children, with paint the likely culprit in cases of acute Pb poisoning but soil an important source of Pb in the myriad examples of chronic Pb poisoning of urban children.

This paper presents a literature review that suggests that Pb contaminated soils and dusts from the outdoor environment are entering the indoor environment and contributing to high Pb exposure rates to children, introduces evidence of seasonal atmospheric soil resuspension in a number of US cities, and presents a climate and soil moisture model that predicts atmospheric soil resuspension. To test the hypothesis that atmospheric soil concentrations display a seasonal pattern, time series of atmospheric soil concentrations were obtained at 8 Inter-agency Monitoring of Protected Visual Environments (IMPROVE) locations across the United States. Results indicate that continental atmospheric soil concentrations exhibit strong seasonality and increase up to an order of magnitude between winter (minimum) and summer (maximum). Additionally, to test the hypothesis that atmospheric soil seasonality is related to weather and soil moisture variables, atmospheric soil concentrations at the Bondville, Illinois IMPROVE site were regressed against the independent variables minimum relative humidity, field measured soil moisture, precipitation, temperature, wind speed and atmospheric pressure obtained from the Illinois Climate Network (ICN, 2007) while adjusting for the month of the year. The model time period was 37 months. Results indicate that 83% of the temporal variation of atmospheric soil concentrations ( $r^2 = 0.83$ ,  $p < 0.001$ ,  $DW = 2.06$ ), could be explained by these variables. The empirical model indicates that when temperatures are high, relative humidity is low, and evapotranspiration maximized, soil moisture decreases, and soil dust is resuspended into the atmosphere. Correlation of local atmospheric soil concentrations with local soil moisture and atmospheric data suggests that a significant proportion of the atmospheric soil is derived from local sources. The findings that weather and soil moisture variables predict

atmospheric soil seasonality and children's blood Pb levels (Laidlaw et al., 2005) support the hypothesis that seasonal resuspension of local Pb contaminated soils in urban environments is driving seasonal Pb poisoning. Furthermore, this finding can provide the basis for an additional tool for clinicians to use when faced with time-specific blood Pb values from patients, and planning effective recommendations for the best course of continued care, based on predicted seasonal trends in blood Pb levels.

## 2. Review of soil Pb, particulate resuspension and blood lead

### 2.1. Urban soil Pb

In the USA, motor vehicles used gasoline containing tetramethyl and tetraethyl Pb additives from the 1920s to 1986. By the 1950s, Pb additives were contained in virtually all grades of gasoline. By 1986, when leaded gasoline was banned, 5–6 million metric tons of Pb had been used as a gasoline additive, and about 75% of this Pb was released into the atmosphere (Chaney and Mielke, 1986; Mielke and Reagan, 1998). Thus, an estimated 4–5 million tons of Pb has been deposited into the US environment by way of gasoline-fueled motor vehicles (Mielke, 1994). Accumulation of soil Pb created by leaded gasoline is proportional to highway traffic flow (Mielke et al., 1997).

In the 1970s, the presumed dominant source of soil Pb contamination was Pb-based house paint (Ter Haar and Aronow, 1974). A subsequent study of garden soils conducted in metropolitan Baltimore, Maryland, began to raise questions about that assumption. Soil around Baltimore's inner city buildings, predominantly unpainted brick, exhibited the highest amounts of Pb, and soils outside of the inner city, where buildings were commonly constructed with Pb-based paint on wood siding, contained comparatively low amounts of Pb, suggesting that Pb-based house paint could not account for the observed pattern of soil Pb (Mielke et al., 1983). The same pattern was also found in Ottawa, Canada (Ericson and Mishra, 1990). Recent exceptions also exist; work (Clark et al., 2008) finds evidence for a paint Pb source for urban garden soils in Boston, but notes that this source is not proximal but rather due to regional seasonal resuspension of Pb-enriched fine particulates. The quantity and distribution of soil Pb have been studied in numerous places: cities in Minnesota (Mielke et al., 1984/85); New Orleans, Louisiana (Mielke, 1994); Milwaukee County, Wisconsin (Brinkmann, 1994); Washington, DC (Elhelu et al., 1995; Roux and Marra, 2007); Indianapolis, Indiana (Laidlaw, 2001; Filippelli et al., 2005); Syracuse, New York (Johnson and Bretsch, 2002; Griffith, 2002); Cleveland, Ohio (Peterson et al., 2006); El Paso, Texas (SCERP, 2007); Pueblo, Colorado (Diawara et al., 2006); Oakland, California (CDC, 2007); Gainesville and Miami, Florida (Chirenje et al., 2004); Oslo, Norway (Tijhuis et al., 2002). All these cities exhibited the same distance decay characteristic of high soil Pb contamination in the inner city and decreasing contamination toward the outer parts of the city as initially identified in garden soils of Baltimore (Mielke et al., 1983). Further,

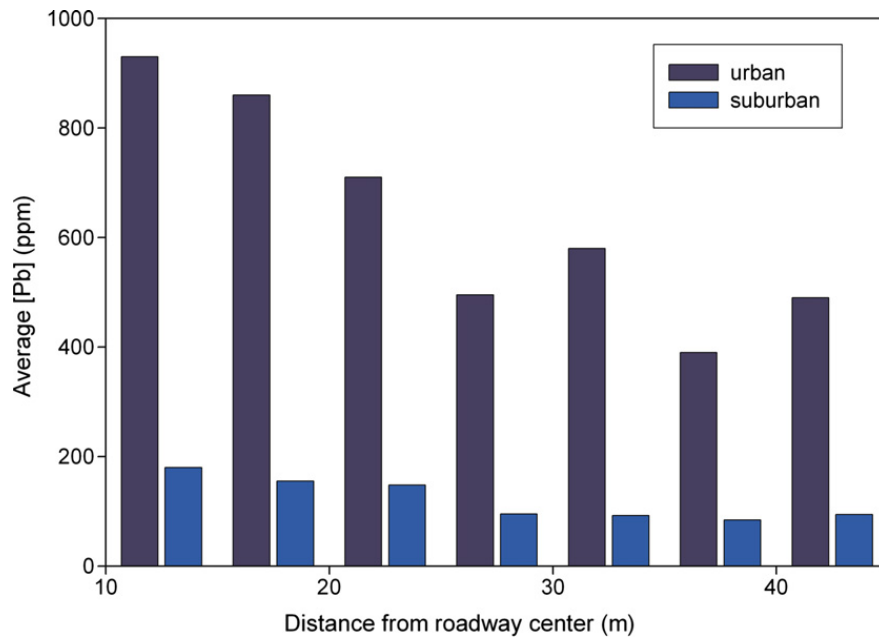
similarities in this distance decay pattern of soil Pb supports the idea that Pb-based house paint was not the sole source contributing to these observed differences.

Lead deposited by human activity onto and retained by surface soils has been added to the relatively small quantities of Pb naturally occurring in the soil. This anthropogenic Pb is generally speciated in the highly bio-available carbonate, and Fe and Mn hydroxide soil fractions, whereas the Pb in natural soils is speciated in the residual, or non-bio-available fractions (Chlopecka et al., 1996; Lee et al., 1997). Therefore, the anthropogenic Pb in dust originating from urban soils is more toxic than that containing naturally occurring Pb. Lead is associated with the smallest particles, the clay grain size fraction in urban soils (Dong et al., 1984); therefore, Pb in dust originating from urban soils is more potent and concentrated than would be expected from simple measurements of the Pb content of the soil (Young et al., 2002). Based on Pb isotope ratios in soils of Dorchester county, Massachusetts, Clark et al. (2006) observed that the source of the Pb in the fine fraction of soil was leaded gasoline.

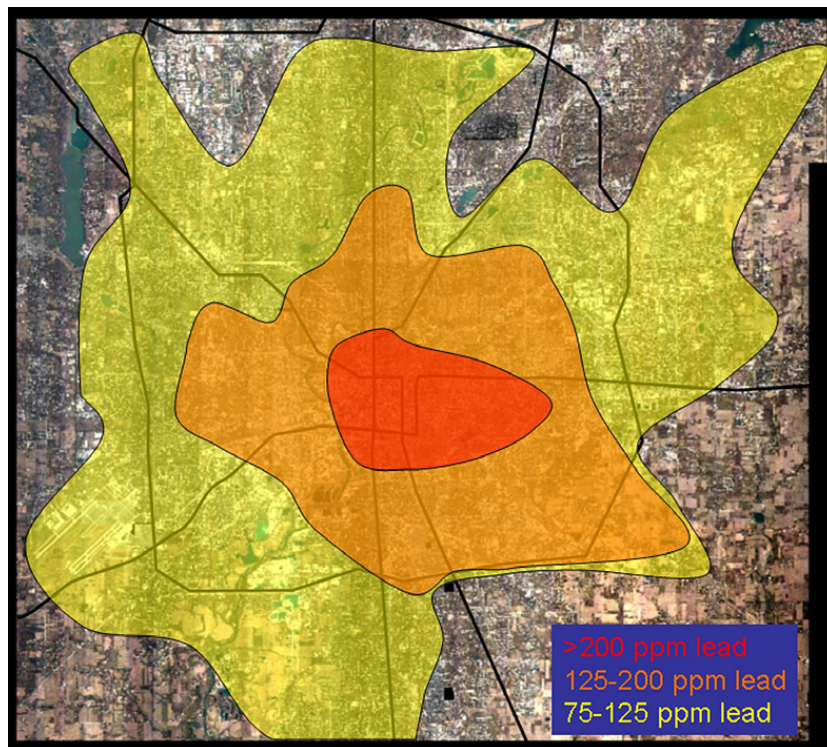
### 2.2. Urban soil Pb topology

Urban soil Pb topology is well understood. Soil Pb decays exponentially away from the roadside, with the concentration proportional to historical traffic volume (Laidlaw, 2001; Filippelli et al., 2005; Lejano and Ericsson, 2005). A large percentage of the Pb emitted from automobiles was deposited within approximately 50 m of the roadside (Fig. 2). However, Filippelli et al. (2005) suggest that due to resuspension of the roadside soil, Pb originally deposited near the roadside has been transported longer distances beyond the roadside fringe (Fig. 2), and can display a city-wide pattern best described as a "Pb Bulls-Eye" (Fig. 3). In addition, Lejano and Ericsson (2005) found that the bioavailability of Pb in roadway soils is highest adjacent to the roadway and also decays exponentially as soil blends with the less bio-available natural Pb found in the parent soil.

Soil Pb also decays exponentially with distance away from the house-side towards the roadside. In homes that used exterior Pb-based paint, the Pb in the house-side soil is a mixture of vehicular Pb and paint Pb (Linton et al., 1980), with automotive Pb concentrated in the finer grain size which is susceptible to resuspension (Clark et al., 2006). The house-side vehicular Pb was deposited when emission particles came into contact with the house-side and deposited to the soils below. This is evidenced by elevated Pb concentrations being found adjacent to brick homes facing the roadway (Mielke et al., 1983). Mielke et al. (2007) measured soil Pb concentrations in two same aged New Orleans housing authority properties, a property in the urban core of New Orleans and a property in the outlying communities, and observed that the soils of the inner city housing complex were much more contaminated with Pb. Mielke et al. (2007) concluded that the inner city housing authority property had many more years of traffic congestion than the outlying housing authority community; therefore, the Pb additives to gasoline, and not Pb-based paint, best explain the differences of soil Pb concentrations.



**Fig. 2.** Average Pb concentrations in surface soil as a function of distance from the roadway using the urban and suburban transects from Indianapolis, IN (after Filippelli et al., 2005). The decrease away from the roadway source is apparent, but more importantly, the significantly higher values in the urban transect, even at distances up to 42.5 m from the road center, beyond the range of direct deposition of Pb particulates from the combustion of leaded gasoline. Additionally, the significant near-roadway loading of surface soils in the urban transect is reflective of higher daily traffic volumes and much greater duration of the urban roadway as an important traffic artery.



**Fig. 3.** The concentration of diffuse soil Pb in surface soils of Indianapolis (in colored regions) displays a characteristic pattern of urban enrichment trending toward background values in suburban and agricultural regions. The overprint of high diffuse soil Pb presented here corresponds roughly with the distribution of elevated blood Pb levels in children (after Filippelli et al., 2005) (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.).

### 2.3. Roadside soil resuspension

Roadside soils are subjected to turbulence as vehicles pass by, and roadside dust emissions are related to vehicle

speed (exponentially), traffic volumes and fractions of heavy trucks (Kuhns et al., 2001; Lough et al., 2005; Nicholson et al., 1989). Resuspension rates have also been found to be dependent on weather conditions (Lough et al., 2005). In

Kuopio Finland, Hosiokangas et al. (2004) found that the mass of Pb in the particulate matter less than 2.5  $\mu\text{m}$  in diameter (PM<sub>2.5</sub>) and particulate matter less than 10  $\mu\text{m}$  in diameter (PM<sub>10</sub>) increased during dust episodes, indicating that factors other than wind speed, such as turbulence induced by traffic, affect the emergence of resuspended soil dust. Hopke et al. (1980) found that street dust was composed of approximately 76% soil materials, and Hunt et al. (1993) estimated that soil contributes between 57% and 90% of road dust, demonstrating the relationship between roadside soil and street dust composition.

#### 2.4. Soil resuspension as a contributor to PM<sub>10</sub>

Analysis of the composition of PM<sub>10</sub> indicates that a portion is composed of soil that has been resuspended. In Bakersfield, California, Young et al. (2002) found that 74% of PM<sub>10</sub> from July through September 1988 was composed of soil. In 6 northern China cities (Tianjin, Jinan, Shijiazhuang, Taiyuan, Urumqi and Yinchuan) during 2000 and 2002, resuspended dust was the greatest source type of ambient PM<sub>10</sub> in 6 cities (Zhao et al., 2006). In Berlin, Germany, Lenschow et al. (2001) observed that at curbsides on main streets, the PM<sub>10</sub> concentration is up to 40% higher than the urban background with half of this additional pollution due to motor vehicle exhaust emission and tire abrasion and the other half due to resuspended soil particles. Soil resuspension has also been found to display a seasonal pattern (Lee et al., 1994; Wells et al., 2007). In two locations in Central Chile, Hedberg et al. (2005) observed that PM<sub>10</sub> was dominated by soil from soil resuspension during the summer months. Arimoto et al. (2002) observed that soil resuspension in 1998 and 1999 was pronounced between May and July in Carlsbad, New Mexico.

Soil resuspension has the capability of entraining significant volumes of Pb into the air of urban areas. Harris and Davidson (2005) calculated that resuspension of soil is responsible for generating 54,000 kg of airborne Pb each year in the South Coast Air Basin of California (SOCAB) and will remain a major source well into the future. Similarly, Lankey et al. (1998) concluded that 43% of Pb emissions in the South Coast Air Basin in California resulted from the resuspension of soil and road dust. A lead isotopic study conducted in 1998 in Yerevan Armenia suggested that 75% of the atmospheric lead was derived from re-suspended soil (Kurkjian and Flegal, 2003). Simons et al. (2007) observed significant differences in particulate loading between urban and suburban areas in Baltimore, Maryland, with urban PM<sub>10</sub> loadings of 47  $\mu\text{g}/\text{m}^3$  versus 8.7  $\mu\text{g}/\text{m}^3$  in suburban areas, and urban PM<sub>2.5</sub> loadings of 34  $\mu\text{g}/\text{m}^3$  versus 18  $\mu\text{g}/\text{m}^3$  in suburban areas. Since PM<sub>10</sub> often consists of large portions of soil particles, this data suggests that urban atmospheric soil loading rates are significantly greater than suburban soil loading rates, irrespective of seasonal differences.

#### 2.5. Reduction in household Pb loading due to soil abatement

Several studies have demonstrated that landscape covering can reduce dust Pb loadings in homes. Binns et al. (2004) evaluated landscape coverings to reduce the poten-

tial for exposure to Pb contaminated soil in an urban neighborhood and found that after 1 year there was a 50% decrease in Pb tracked onto the floor mats. In Boston, Massachusetts, Dixon et al. (2006) applied low-cost soil interventions to reduce exposure to soil Pb hazards by applying ground coverings and ground barriers, with following up after 1 year. The treatments were effective in reducing entryway dust Pb in the rear of the building if the residents reported they had maintained the yard treatments. In addition, each additional 100 ft.<sup>2</sup> (9.3 m<sup>2</sup>) of yard treated was predicted to lower 1-a floor dust loading at the rear dwelling unit entry by 19%. Furthermore, Mielke et al. (2006) placed 680 metric tons (750 tons) of clean soil cover on a 6424 m<sup>2</sup> (69,153 ft.<sup>2</sup>) soil Pb contaminated lot and reduced the median surface soil Pb to 6 mg/kg (range 3–18). Interior entrance wipe samples were collected at 10 homes before and after soil treatment and showed a decreasing trend of Pb ( $p$ -value = 0.048) from a median of 52 g/ft.<sup>2</sup> (560 g/m<sup>2</sup>) to a median of 36 g/ft.<sup>2</sup> (366 g/m<sup>2</sup>) (25th and 75th percentiles are 22 and 142 g/ft.<sup>2</sup> (237 and 1528 g/m<sup>2</sup>) and 12 and 61 g/ft.<sup>2</sup> (129 and 657 g/m<sup>2</sup>), respectively).

#### 2.6. Transport of Pb from outside to inside

##### 2.6.1. Particulate penetration and air exchange rates

Penetration of Pb into homes from the exterior is based on an understanding of particulate penetration, which has both point sources (e.g., particulates on clothes, shoes, pets, etc.) and diffuse sources related to the “leakiness” of a home or apartment. The point sources depend strongly on occupant-specific behavior, which is difficult to generalize and best accessed on a case-by-case basis. But the diffuse sources can be generalized, with the generalizations sometimes helpful for understanding the intrusion of particulates, and associated Pb, into homes (e.g., Mosley et al., 2001).

The rate at which outdoor air replaces indoor air is described as the air exchange rate (AER). The air exchange rate is generally expressed in terms of air changes per hour (ACH, with units of h<sup>-1</sup>), the ratio of the airflow (m h<sup>-1</sup>) to the volume (m). “Air exchange is the balanced flow into and out of a building, and is composed of three processes: (1) infiltration – air leakage through random cracks, interstices, and other unintentional openings in the building envelope; (2) natural ventilation – airflows through open windows, doors, and other designed openings in the building envelope; and (3) forced or mechanical ventilation – controlled air movement driven by fans” (USEPA, 2008).

A study in Boston, Massachusetts (Abt et al., 2000) observed that low air exchange rates (<1 exchange/h) resulted in longer air residence times and more time for particle concentrations from indoor sources to increase and at higher exchange rates (>1 exchange/h), the impact of indoor sources was less pronounced, as indoor particle concentrations tracked outdoor levels more closely. A study of ambient and indoor elements in New York City observed that personal, indoor and outdoor median concentrations of most particle-associated elements were similar, suggesting that ambient sources drive indoor and personal exposures for most elements and concluded that there was little evidence for major indoor sources of particle-associated elements (Kinney et al., 2002). This study

also found that seasonal differences in home AERs (lower in winter than in summer) appeared to influence I/O ratios of elements. The age of the home also affects AERs. For example, Sherman and Chan (2004) noted that houses built prior to 1980 showed a clear increase in leakage with increasing age and were leakier, on average, than newer houses. They further observed that newer homes (post-1980) did not show any trend in leakiness with age. Finally, window opening/closing patterns have an effect on air exchange rates. In a study of an occupied home, Wallace et al. (2002) observed that the strongest influence on air change rates was opening windows, which could increase the rate to as much as  $2 \text{ h}^{-1}$  for extended periods, and up to  $3 \text{ h}^{-1}$  for short periods of a few hours. The use of the attic fan also increased air change rates by amounts up to  $1 \text{ h}^{-1}$ .

### 2.6.2. Exterior Pb loading and indoor Pb penetration

In New York City, Caravanos et al. (2006a) 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 time period of 6 weeks. In a related study, Caravanos et al. (2006b) measured the following median dust loadings in New York: Brooklyn ( $730 \mu\text{g}/\text{ft}^2$ ;  $7858 \mu\text{g}/\text{m}^2$ ), Staten Island ( $452 \mu\text{g}/\text{ft}^2$ ;  $4865 \mu\text{g}/\text{m}^2$ ), the Bronx ( $382 \mu\text{g}/\text{ft}^2$ ;  $4112 \mu\text{g}/\text{m}^2$ ), Queens ( $198 \mu\text{g}/\text{ft}^2$ ;  $2131 \mu\text{g}/\text{m}^2$ ) and Manhattan ( $175 \mu\text{g}/\text{ft}^2$ ;  $1884 \mu\text{g}/\text{m}^2$ ). When compared to the HUD/EPA indoor Pb in dust standard of  $40 \mu\text{g}/\text{ft}^2$  ( $431 \mu\text{g}/\text{m}^2$ ), their data show that this value is exceeded in 86% of the samples taken. Fergusson (1986) reviewing work by Butler et al. (1975), Diemel et al. (1981), Gloag (1980), Facchetti et al. (1982) and Elwood (1984) concluded that aerosol Pb penetrates houses, with detected levels varying between 45% and 95% of outside levels. Fergusson (1986) concluded that houses do not offer substantial protection from aerosol Pb.

The particle size in which the mass of Pb resides in resuspended soil dust is important as the penetration efficiency is maximized at the particle size which fractionates strongly for Pb. In Thessaloniki, northern Greece, the mean mass median aerodynamic diameter (MMAD) of Pb was observed to be  $0.96 \pm 0.71 \mu\text{m}$  (Samara and Voutsas, 2005). Similarly, in July 1994 in Chicago, Illinois, Paode et al. (1998) observed that the size distribution of atmospheric Pb peaked in the 0.5 micron grain size fraction. Abt et al. (2000) observed that outdoor particles ( $0.02$ – $0.5$  and  $0.7$ – $10 \mu\text{m}$ ) were found to contribute significantly to indoor particle levels and effective penetration efficiencies ranged from 0.38 to 0.94 for  $0.02$ – $0.5 \mu\text{m}$  particles and from 0.12 to 0.53 for  $0.7$ – $10 \mu\text{m}$  particles. Furthermore Abt et al. (2000) explained that the penetration efficiency for  $0.7$ – $10 \mu\text{m}$  particles decreased with increasing particle size, reflecting the influence of deposition losses from gravitational settling.

Doors and windows are primary entry points of Pb bearing dust into homes (Lepow et al., 1974, 1975; Archer and Barratt, 1976; Sayre and Katzel, 1979; Bornschein et al., 1986; Fergusson, 1986; Davies et al., 1987; Farfel and Chisolm, 1990, 1991; Al-Radday et al., 1993). Demonstrating the effect of indoor dust penetration, Sayre and Katzel (1979) analyzed the Pb content on windowsills (containing no Pb paint), floors, and other surfaces in 24 vacant houses with all windows closed in urban Rochester

and Buffalo, New York. Results indicated that high yields of Pb were obtained from windowsills and floor areas adjacent to windows. Sayre and Katzel (1979) suggest that the Pb deposited on the windowsills and floors originated from dust entering the house from outside the home around the loose-fitting windows of older homes. In Hong Kong, Tong and Lam (2000) observed that homes that do not have their windows opened often had a lower level of contaminants in their house dust.

Attic dust (ceiling dust) accumulations in urban areas can be substantial and are composed primarily of soil particles – a strong indicator that soil is being resuspended and deposited within homes in urban environments (Davis and Gulson, 2005). In a study of 38 houses in Sydney, Australia, (Davis and Gulson, 2005) observed that approximately 75% of the ceiling dust in attics originated from soil or plant matter, with 25% composed of anthropogenic matter. The ceiling dust was as thick as 10 cm and has been observed to weigh in excess of 800 kg. The volume of the dust ranged from 7 to  $233 \text{ g}/\text{m}^2$ . This volume of accumulated exterior dust is possible because wind-borne particles can be drawn through gaps in the roof structure due to pressure differences, where they are rapidly deposited due to a slackening of air flow (Davis and Gulson, 2005).

### 2.7. Source apportionment of Pb in house dust

Chemical mass balance was used to apportion the major proximate contributors of Pb mass to house dust (HDPb) in 64 urban Jersey City, New Jersey, homes with Pb-based paints (Adgate et al., 1998b). Coarse (up to  $\sim 60 \mu\text{m}$ ) and PM10 ( $<10 \mu\text{m}$ ) particle size fractions of vacuum dust samples were collected. Adgate et al. (1998b) observed that crustal materials and deposited airborne particulates were responsible for approximately? of the HDPb mass and interior Pb-based paint sources contributed the remaining? Furthermore, Using Pb isotope ratio analysis, Adgate et al. (1998a) found that about half of the Pb in house dust of 10 homes in Jersey City, originated from sources outside the home, such as soil.

In Port Pirie, Australia, in an area with Pb contaminated soils near a large Pb smelter, Kutlaca (1998) measured dust loading and dust concentrations inside 16 homes which were subsequently thoroughly cleaned. In each of the homes doors and windows remained closed for 47 days. At the completion of the test, the Pb loadings exceeded the pre-cleaning Pb loading. Kutlaca (1998) concluded that dust accumulation (as detected at floor level) within homes is an ongoing process, and that dust detected after the cleaning program originates as newly entering (room/house) dust. Similarly, Campbell et al. (2003) conducted a study to determine the effects of a follow-up professional Pb dust cleaning of homes in Philadelphia at 3 month intervals for 18 months after an initial cleaning. This study found that dust Pb re-accumulated to pre-cleaning levels within 3–6 months.

### 2.8. Association between soil Pb and blood Pb study designs

A large number of studies have tested the hypothesis of an association between soil Pb and blood Pb. Generally, 5

study designs have been used: ecological-temporal, ecological-spatial, cross-sectional, prospective soil removal and descriptive. Each of the study designs has methodological limitations.

#### 2.8.1. Association between soil Pb and blood Pb study designs: cross-sectional

The cross-sectional study design is the most frequently applied but is severely limited due to exposure misclassification and outcome misclassification. Exposure misclassification is best characterized as a limitation in field-related measurements. For example, it is well known that in urban areas, soil Pb concentrations decay exponentially away from the roadside (e.g., Filippelli et al., 2005). It is also known that soil Pb concentrations are elevated adjacent to homes due to the effect of past aerosols coming into contact with the side of buildings and deposited to the soil below. In homes with exterior Pb-based paint, the soils adjacent to homes can become contaminated due to deterioration of leaded paint. In addition, there is variability in exposure within and between homes due to variable hand to mouth behaviors. For example, Ko et al. (2007) found significant correlation between children's hand to mouth frequency and blood Pb levels in the children in a smaller video assessment. Cross-sectional studies that are based on single sample exposure will result in exposure misclassification because point samples can be highly variable for the above-mentioned reasons. Outcome misclassification may arise due to the blood Pb seasonality effect. Blood Pb seasonality of groups of children has shown that blood Pb concentrations can increase over 150% between winter and summer (USEPA, 1995). The exposure and outcome misclassification makes it very difficult to correlate due to the high variability of exposure (soil Pb) and outcome (blood Pb). In addition, these studies often have low sample sizes. This likely explains why cross-sectional studies sometimes have historically low correlations, due to the inadequacy of the study design. An example of a cross-sectional study was a study of the association between soil Pb and blood Pb at 4 superfund sites which used the Agency for Toxic Substances and Disease Registry's data (Lewin et al., 1999). This study found that the predicted blood Pb level corresponding to a soil Pb level of 500 mg/kg was 5.99 µg/dL with a 95% prediction interval of 2.08–17.29. Additional cross-sectional studies or literature summaries detailing positive associations between soil Pb and blood Pb include Rabinowitz et al. (1985), Reagan and Silbergeld (1989), Fett et al. (1992), Bates et al. (1995), Jacobsen (1996) and Jin et al. (1997).

#### 2.8.2. Association between soil Pb and blood Pb study designs: ecological-spatial

The ecological-spatial study design takes the average blood Pb of a population of a small area for a given period of time, and correlates that blood Pb with the average soil Pb concentration for the same small unit, across all the small area units in a city. This study design has been successfully applied ( $r^2 > 0.65$ ) in Syracuse, NY (Johnson and Bretsch, 2002; in this case, a regression of geographic mean blood Pb on median soil Pb values), New Orleans (Mielke et al., 1997) and Shenyang, China (Wang et al., 2006). This

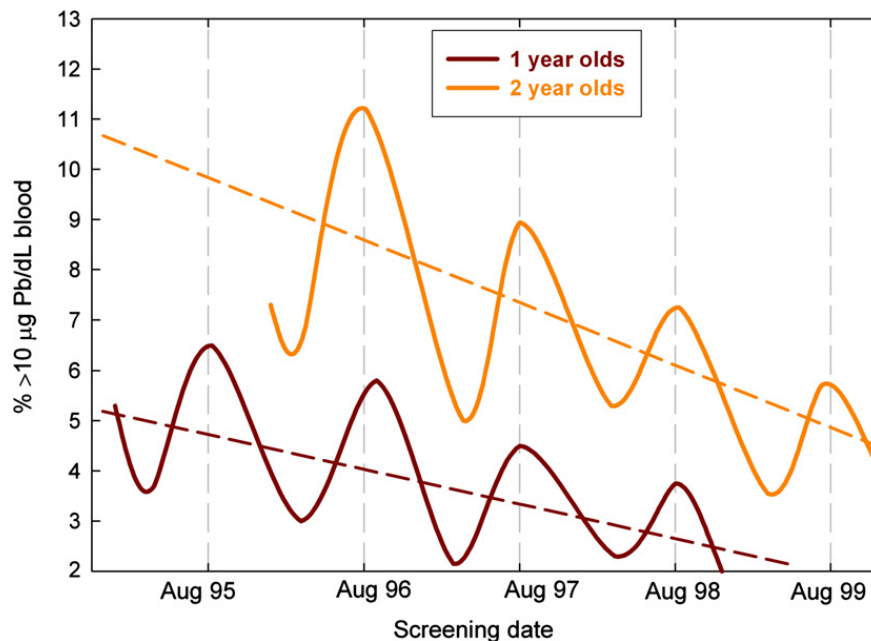
study design appears to be successful because the exposure (soil Pb) is generally an average of a large number of soil samples in a small area, thus dampening out the exposure misclassification that occurs in the cross-sectional study design. Furthermore, since it is time invariant, the misclassification from seasonality effect that is found in the cross-sectional study design is not a factor. The limitation of this study design is that the confounding variable, homes with paint Pb, is not considered, although it can be. In fact, Mielke et al. (1997) found that the p value for spatial association between soil Pb and blood Pb was many orders of magnitude lower than the association between the percentages of homes with paint Pb with blood Pb. Another limitation is that in the ecological study design, the association is between the average blood Pb of the group and not the individuals – thus the inference of a blood Pb-soil Pb association at the individual level should be made cautiously.

#### 2.8.3. Association between soil Pb and blood Pb study designs: ecological-temporal

The ecological-temporal study design is conducted by selecting a city, then taking the average monthly blood Pb concentration of all the children, for a specific time period, then regressing against soil moisture and climate variables (e.g., Laidlaw et al., 2005). This approach is believed to be successful because soil moisture and climate variables are proxy indicators of urban Pb contaminated soil resuspension (Laidlaw and Filippelli, 2006). This approach eliminates spatial concerns, such as confounding by paint Pb, and examines temporal fluctuations only. The limitation of the study design is that it does not account for variation in Pb particles released from the opening and closing of windows. However, the opening and shutting of windows is not related to temporal changes in variables such as PM10 and soil moisture, and thus Pb particles released from the opening and closing of windows may not be related to a significant proportion of the variation in children's blood Pb seasonality. In addition, ecological studies are based upon the group, rather than the individual, and inferences at the individual level must be made with caution.

#### 2.8.4. Association between soil Pb and blood Pb study designs: prospective soil removal

Prospective soil removal studies involve removing Pb contaminated soil from yards of selected homes while leaving Pb contaminated soil in yards of other homes. Blood Pb concentrations of children in the homes are then compared between treatment and controls. These study designs have limitations. First, the sample sizes can be very low. Second, the blood Pb samples must be collected at the same point in time to prevent outcome misclassification due to blood Pb seasonality which could result in lower correlations and p values. Some of these studies have failed to account for differences in how the homes are sealed as well. These studies can also fail because they do not take into account the fact that dust that travels to a home does not originate from the soil from the home but the surrounding community (von Lindern et al., 2003). This was illustrated by Sheldrake and Stifelman (2003) near the



**Fig. 4.** Seasonal patterns in children's blood Pb levels from New York State, showing summer peaks and a general decline from 1995 to 1999 (after Haley and Talbot, 2004). Note the generally higher levels of Pb poisoning in 2-year olds, who are generally more mobile with consequently greater hand exposure to Pb contaminated surfaces and more access to the outdoors.

Bunker Hill Superfund site where they found that cleanup of residences has a 3-fold greater reduction of children's blood Pb levels compared with cleaning only those homes where children currently reside by reducing exposures attributable to neighboring properties. This approach has been successfully applied by Maisonet et al. (1997) who conducted a pair-matched, case-control study of yard soil remediation and found that yard soil removal was a strong protective factor for elevated blood Pb levels in children (odds ratio, 0.28; confidence interval, 0.08–0.92). Additional studies that demonstrated reduction in blood Pb due to soil abatement include Aschengrau et al. (1994), Gagne (1979), Langlois et al. (1996), Lanphear et al. (2003), and de Freitas et al. (2007).

#### 2.8.5. Association between soil Pb and blood Pb study designs: Descriptive

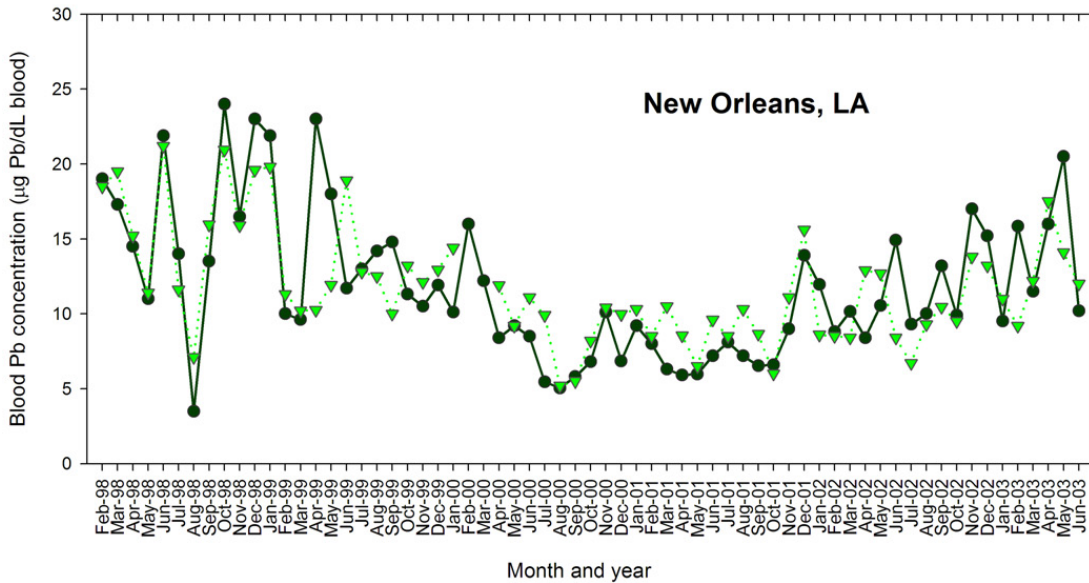
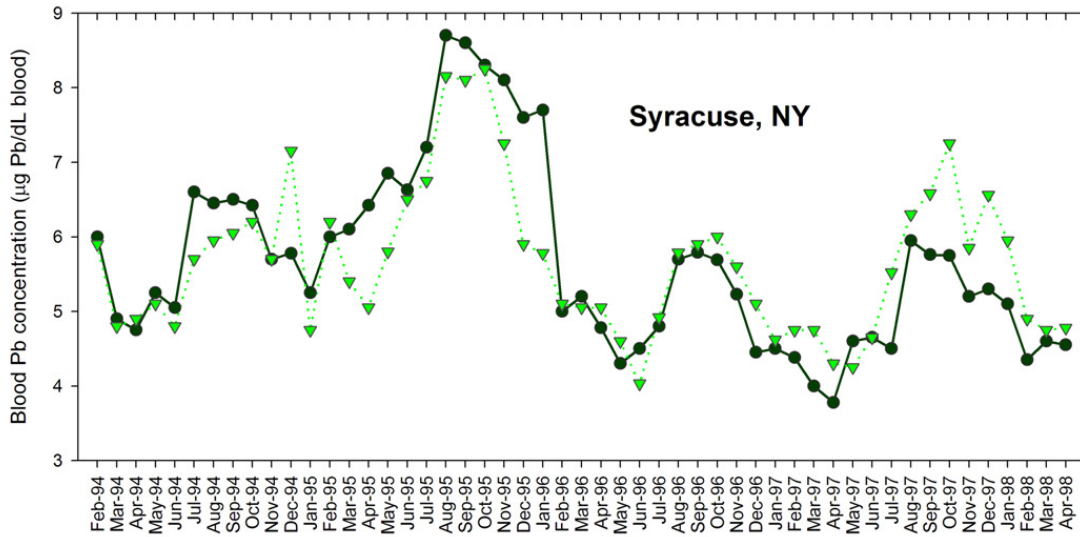
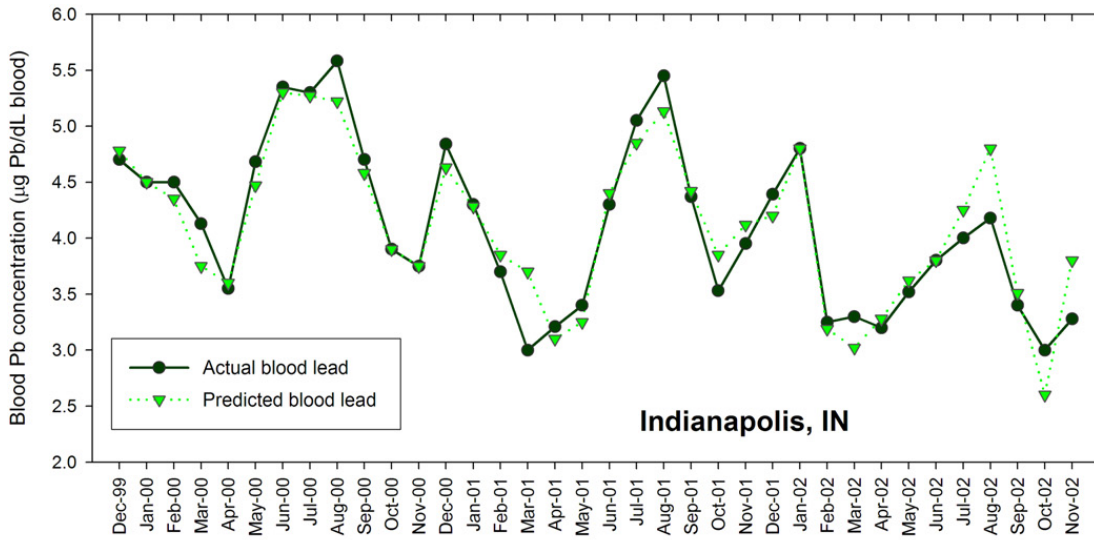
Isotopic studies can assist in determining the source of lead in children's blood, however their application has limitations (Duzgoren-Aydn and Weiss, 2008). The use of isotopic ratios have shown promise in Armenia where the isotopic ratio of lead in roadside soil has been found to match lead isotope ratios in blood (Kurkjian et al., 2002).

#### 2.8.6. Blood Pb seasonality

Average monthly blood Pb (BPb) values of children from urban areas tends to increase significantly in summer months (Fig. 4; Billick et al., 1979; Blanksma et al., 1969; Blatt and Weinberger, 1993; Haley and Talbot, 2004; Hayes et al., 1994; Hunter, 1977; Hwang and Wang, 1990; Johnson and Bretsch, 2002; Johnson et al., 1996; Kimbrough et al., 1984; Laidlaw et al., 2005; Marrero et al., 1983; Mielke and Reagan, 1998; Rabinowitz and Needleman, 1982; Rothenberg et al., 1996; Stark et al., 1980; USEPA,

1995, 1996; Yiin et al., 2000). Early work by Mielke et al. (1992), Johnson and Bretsch (2002) and Johnson et al. (1996) suggested that blood Pb seasonality may be related to the interaction between climate and Pb contaminated soils. Yiin et al. (2000) and USEPA (1995) actually measured seasonal changes in dust Pb levels and correlated blood Pb levels with seasonal dust Pb concentrations. Yiin et al. (2000) conducted a study to examine seasonal changes in residential dust Pb content and its relationship to blood Pb in preschool children. The study found that windowsill wipe samples were most correlated with blood Pb concentration and the variation of dust Pb levels for floor Pb loading, windowsill Pb loading, and carpet Pb concentration were consistent with the variation of blood Pb levels, showing the highest levels in the hottest months of the year (June, July and August). This study demonstrated that children appear to receive the highest dust Pb exposure indoors and outdoors during the summer, when they have the highest blood Pb levels. Laidlaw et al. (2005) recently found that that weather and soil moisture variables robustly predicted seasonal changes in children's blood Pb levels in a number of US cities (Fig. 5), concluding that weather and soil moisture variables were a proxy of seasonal soil resuspension and exposure of children to Pb contaminated urban soils, with the potential to use site-specific weather and soil moisture determinations to better assess clinical blood Pb data.

USEPA (1995) examined the temporal variation in blood- and environmental-Pb levels in data observed for a sample of 249 children in Boston between 1979 and 1983 at the Brigham and Women's Hospital. For each child in the study, blood Pb and environmental-Pb measurements were collected longitudinally over a period of 2 years. Blood Pb levels were found to have highly significant seasonal variations ( $p < 0.0001$ ), with the maximum



modeled to occur in late June, and the minimum in March. The estimated maximum- to-minimum ratio was 2.5. Air-, floor dust-, furniture dust-, and window sill dust Pb levels all exhibited highly significant seasonal variation. The estimated maximum-to-minimum ratios were 2.3 for air Pb, 1.5 for floor dust Pb, 1.4 for furniture dust Pb and 1.6 for window sill dust Pb. The extent to which levels of Pb in blood were correlated with levels of Pb in the environment was also evaluated. There was abundant evidence supporting the existence and parallelism of the seasonal variations among blood-, air-, floor dust-, and furniture dust Pb levels. The 3 environmental-Pb measures peak in July, which is very near the blood Pb peak month of June. In addition, the maximum-to-minimum ratios in the environmental-Pb measures, ranging from 1.4 to 2.3, are of the same order of magnitude as the blood Pb ratio of 2.5. The USEPA concluded that seasonal variations in environmental-Pb levels contribute to the blood Pb rhythms in Boston children.

Further evidence of seasonal variation of household dust loading was found in New Jersey, where Edwards et al. (1998) found that the mean summertime household dust loadings were 68% higher than mean winter household dust loadings. In addition, Paode et al. (1998) observed that Pb loading was highest during the summertime in Chicago, while Blanksma et al. (1969) observed that the incidence of high blood Pb values was lowest in November through January and highest in June.

#### 2.8.7. Blood Pb seasonality and climate forcing

To explore the potential link between climate, soil Pb, dust Pb, and blood Pb, Laidlaw et al. (2005) examined the temporal relationship between pediatric BPb, weather, soil moisture, and dust in Indianapolis, Indiana; Syracuse, New York; and New Orleans, Louisiana. This study modeled average monthly soil moisture, PM10, wind speed and temperature. Of temporal variation in urban children's BPb, 87% in Indianapolis ( $r^2 = 0.87$ ,  $p = 0.000$ ), 61% in Syracuse ( $r^2 = 0.61$ ,  $p = 0.001$ ) and 59% in New Orleans ( $r^2 = 0.59$ ,  $p = 0.000$ ) are explained by these variables. Laidlaw et al. (2005) suggested a conceptual model of urban Pb poisoning: when temperature is high and evapotranspiration maximized, soil moisture decreases and soil dust is resuspended and deposited. Under these combined weather conditions, Pb-enriched PM10 dust disperses in the urban environment and causes elevated Pb dust loading. Laidlaw et al. (2005) concluded that seasonal variation of children's Pb exposure is probably caused by inhalation and ingestion of Pb brought about by the effect of weather on soils and the resulting fluctuation in Pb loading. Direct

measurements of seasonal variations in air Pb levels have previously been shown to be highly correlated with children's blood Pb levels. In New York City between 1970 and 1976 it was demonstrated that seasonal variations in blood Pb levels of large populations of children (>300,000) were associated with seasonal variations in exterior Pb ( $r^2 = 0.60$ ,  $p < 0.0001$ ; Billick et al., 1979). Edwards et al. (1998) measured dust loading inside 4 homes in New Jersey in the summer and winter and observed that the dust mass deposition rate in summer ( $0.37 \pm 0.13 \mu\text{g}/\text{cm}^2/\text{day}$ ) was almost twice as great as winter ( $0.22 \pm 0.13 \mu\text{g}/\text{cm}^2/\text{day}$ ), arguing for summer ventilation as a primary driver of interior dust loading.

#### 2.8.8. Alternative theories of blood Pb seasonality

Alternative theories about the causal mechanism for child blood Pb seasonality have been posited. One theory is that changes in Vitamin D induced by diet, sunlight exposure, age, skin pigmentation and other factors, may modify gastrointestinal Pb absorption or release of Pb stored in bones into the bloodstream. A second competing theory is that the opening and closing of windows covered with Pb-based paint during the warmer months results in higher Pb loading rates, causing blood Pb seasonality.

Recent research (Kemp et al., 2007) refutes the theory that changes in vitamin D are related to seasonal changes in blood Pb levels. Kemp et al. (2007) collected paired blood samples from 91 urban African-American children in the winter and summer to study the seasonal increase in blood Pb and its relationship to vitamin D nutrition, age, and race. They observed that elevated blood Pb concentrations were frequent in the 91 African-American children, especially those 1–3 years of age and the percentage with elevated blood Pb levels increased by 32.4% in summer. Kemp et al. (2007) concluded that: "Because the seasonal increase of 32.4% in blood Pb in the 1- to 3-year-old children was not accompanied by a significant increase in serum 25-OH-D concentrations, there must be other as yet unknown causes for this phenomenon in the children studied besides sunlight-induced vitamin D synthesis. These causes may include increased summertime exposure of children to Pb in dust and soil".

The second alternative hypothesis that the opening and closing of windows during the summertime releases Pb-enriched particles that cause blood Pb seasonality also finds little support. Although there is indication that friction surfaces of windows with non-intact Pb paint can cause an increase in dust Pb on the window sill (e.g., Dixon et al., 2007), Hunt et al. (1993) measured seasonal changes

**Fig. 5.** Best-fit model results to predict Pb lead levels in children from Indianapolis, Syracuse (NY), and New Orleans compared to actual monthly average blood Pb levels (after Laidlaw et al., 2005). This type of effort can be used to better treat Pb poisoning from a public health perspective by providing clinicians with predicted trends of blood Pb levels (functionally calculated as a percent deviation from mean) at a given blood sampling event, allowing them to calculate the potential increase or decrease with time given normal exposure. The independent variables for each model consisted of soil moisture, wind speed, PM10, temperature, atmospheric Pb, interaction variables and monthly dummy variables. The dependent variables for the models included monthly child blood Pb data from (1) a variety of subregions, (2) a variety of blood Pb levels, and (3) a variety of ages (i.e., 0–1.0, 1.01–2.0, 2.01–3.0, 3.01–4, 4.01–5, and 5.01–7.0 years). The independent variables temperatures, PM10, minimum relative humidity, soil moisture, atmospheric Pb concentration, were computed as the arithmetic mean, while the wind speed was computed as the median. Actual monthly average BPb versus predicted monthly average BPb in Indianapolis, Indiana for a 36-month time period between December 1999 and November 2002 ( $n = 15,969$ ,  $r^2 = 0.87$ ,  $p = 0.0004$ ,  $DW = 1.71$ ,  $LM = 0.85$ ). Actual monthly average BPb versus predicted monthly average BPb in Syracuse, New York for a 51-month time period between January 1994 and March 1998 ( $n = 14,467$ ,  $DW$   $r^2 = 0.61$ ,  $p = 0.0012$ ,  $= 2.05$ ,  $LM = 0.049$ ). Actual monthly average BPb versus predicted monthly average BPb in New Orleans, Louisiana for a 65-month time period between January 1996 and May 2003 ( $n = 2,295$ ,  $r^2 = 0.59$ ,  $p = 0.0000078$ ,  $DW = 1.71$ ,  $LM = 0.85$ ).

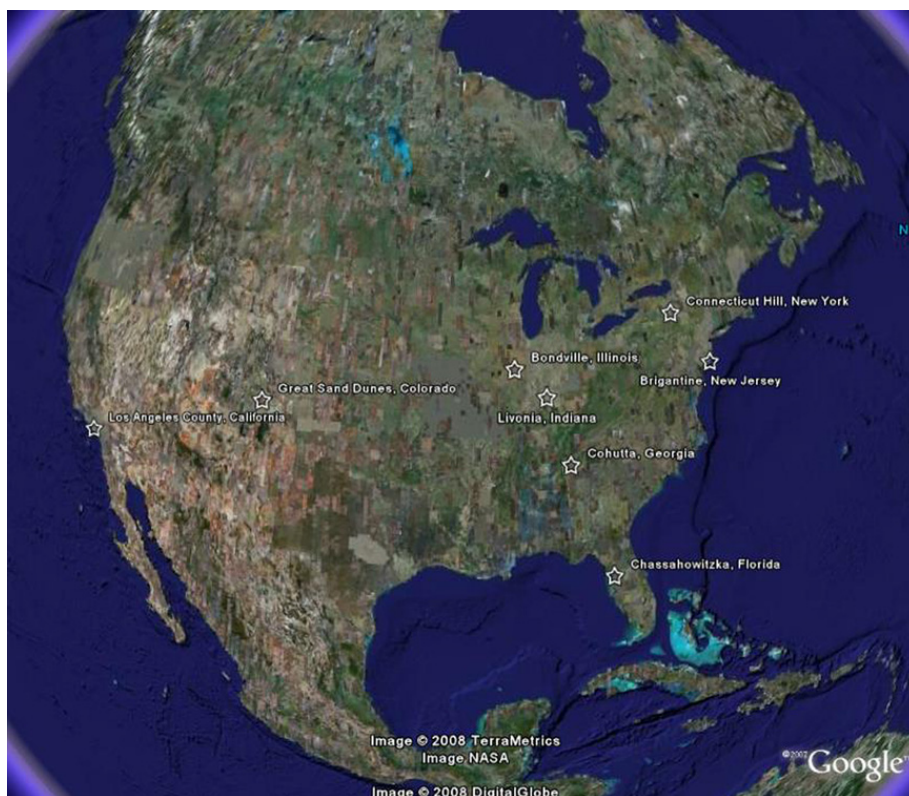


Fig. 6. Map of the 8 IMPROVE sites used for atmospheric soil concentration modeling.

in Pb-based paint particles adjacent to Pb paint covered windows. They observed no statistical difference of Pb-based paint derived particle loading between spring and late summer and early autumn. Clearly, frictional processes can cause fine particulate paint debris which may contain very high Pb and might be very accessible to children using a lower window sill as a lifting point, but it is unclear whether this Pb source is a seasonally significant one. Although the seasonality link does not appear to derive from windows, degraded Pb paint near poorly maintained windows can be an important general source of Pb to children, and window replacement with Pb safe windows, along with Pb stabilization efforts, may have long-term health, energy, and economic benefits (e.g., Nevin et al., 2008).

### 3. New directions in soil resuspension and Pb loading

Based on the review above, the authors argue that it is plausible to consider soil resuspension as both a contributor to Pb loading in urban areas and one that can be modeled in terms of potential health impacts. In the following, a soil resuspension model derived from soil moisture and meteorological variables is applied to atmospheric soil data from a number of US cities involved in the IMPROVE program to test the following two hypotheses:

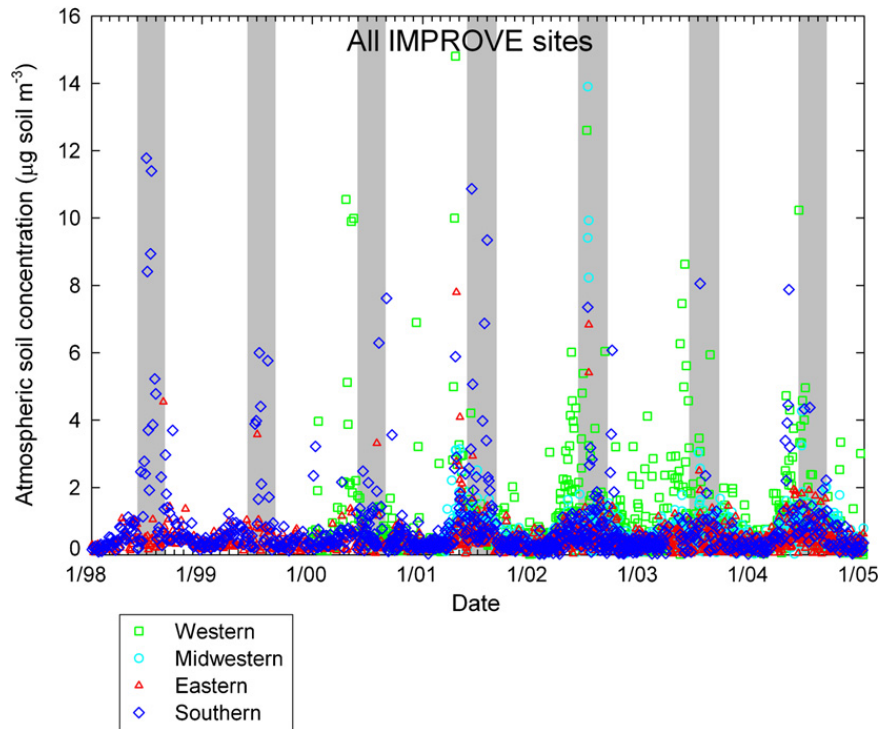
1. Atmospheric soil loading in the United States displays a seasonal pattern with higher soil loadings during periods of higher evapotranspiration.
2. Seasonal changes in atmospheric soil loading can be predicted by applying a multiple linear regression model using soil moisture and meteorological variables.

The predictive capability of this model is illustrated by a 3 city blood Pb model (Laidlaw et al., 2005) which was applied to municipalities with extensive blood Pb epidemiological data. The areas examined in the current study do not have these blood Pb databases, but the results provide some constraints on human Pb exposure in different climatic zones based on seasonal records of higher soil resuspension.

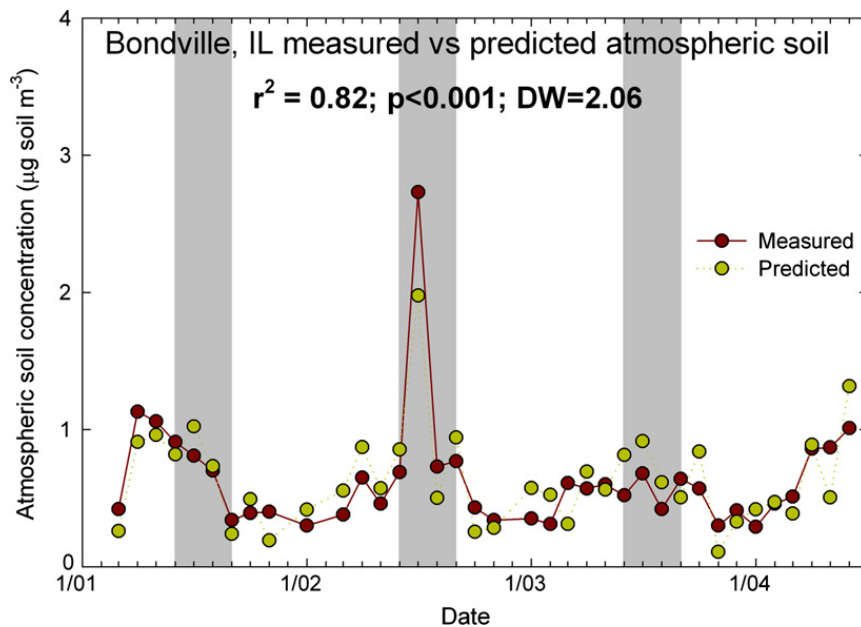
#### 3.1. Methods

##### 3.1.1. Data

The dependent variable for the model was atmospheric soil, which was obtained from the Interagency Monitoring of Protected Visual Environments (IMPROVE) station in Bondville, Illinois (IMPROVE, 2007). The independent variable, soil moisture, was field measured from the top 6 in. (15.2 cm) of soil at the Illinois Water Survey station 81 near Champaign, Illinois (Hollinger and Isard, 1994). The meteorological variables maximum temperature, wind speed, precipitation, and relative humidity were obtained from the Illinois Climate Network (ICN, 2007) and atmospheric pressure was obtained from NOAA (NOAA, 2007). The atmospheric soil data was obtained from the IMPROVE monitoring networks in Los Angeles County, California; Great Sand Dunes, Colorado; Bondville, Illinois;



**Fig. 7.** Composite of individual atmospheric soil records from IMPROVE sites, with “Western” sites including Los Angeles County and Great Sand Dunes (green squares), “Midwestern” sites including Bondville and Livonia (blue circles), “Eastern” sites including Connecticut Hill and Brigantine (red triangles), and “Southern” sites including Cohutta and Chassahowitzka (blue diamonds). Gray vertical bars represent summer months June, July and August (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.).



**Fig. 8.** Relationship between measured atmospheric soil loading (red curve) and atmospheric soil loading as predicted by meteorological and soil moisture parameters for the Bondville, Illinois site. This model used roughly the same parameters as that of Laidlaw et al. (2005) for predicting children’s blood Pb levels, indicating that atmospheric soil loading can be successfully modeled in the absence of direct measurement records and could also be used for predicting seasonal patterns in children’s blood Pb levels (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.).

Livonia, Indiana; Cohutta, Georgia; Chassahowitzka, Florida; Brigantine, New Jersey; and Connecticut Hill, New York (Fig. 6; IMPROVE, 2007).

### 3.1.2. Statistical model

The monthly atmospheric soil concentrations were used as the dependent variable in a backward elimination mul-

multiple regression model with soil moisture and meteorological parameters maximum temperature, atmospheric pressure, wind speed, precipitation and relative humidity as independent variables. Interaction variables were also modeled. Monthly dummy variables were used as independent variables to control for the month of the year. Statistical analysis was performed using SPSS (version 11.5; SPSS Inc., Chicago, IL).

## 4. Results

### 4.1. Hypothesis #1 – USA atmospheric soil seasonality

Atmospheric soil concentrations at all 8 locations demonstrate strong seasonal loading rates with fluctuations varying by an order of magnitude between peaks and troughs (Fig. 7). The fact that the variable is soil, and it was sampled from the atmosphere, implies that the atmospheric soil was resuspended from surface soils. The summer months of June, July and August (JJA), representing a period of high evapotranspiration, are highlighted in Fig. 7. While the summertime soil loading rate is periodic, the wave height can be highly irregular from year to year (Fig. 7). The results demonstrate that atmospheric soil loadings display a strong seasonality across the United States, but that seasonal patterns vary from one climatic regime to another. For example, Eastern and Southern sites exhibit the strongest JJA signal in terms of soil resuspension, while the more arid Western sites have more erratic signals. Los Angeles in particular displays strong seasonality, but with highest atmospheric resuspension occurring in the spring and fall, coincident with the monsoonal climate and the peak intervals of Santa Ana wind.

### 4.2. Hypothesis #2 – atmospheric soil prediction model

The Bondville area atmospheric soil seasonality model (Fig. 8) indicates that 83% ( $r^2 = 0.83$ ,  $p < 0.001$ ,  $DW = 2.06$ ) of the variation in atmospheric soil was explained by meteorological and soil moisture parameters, roughly the same parameters used to predict children's blood Pb levels by Laidlaw et al. (2005). Thus, the hypothesis that meteorological and soil moisture variables could predict atmospheric soil loading could not be rejected.

## 5. Discussion

Numerous studies have demonstrated that soil moisture concentration is a significant control of dust (PM10) suspension and loading (Edlefsen and Anderson, 1943; Chen et al., 1996; Clausnitzer and Singer, 1996, 2000; Nickovic et al., 2001; Cornelis and Gabriels, 2003; Harris and Davidson, 2005). Soil moisture is a predictor of wind erosion because soil moisture contributes to bind particles together (McKenna-Neuman and Nickling, 1989). Soil particles will become deflated when destabilizing forces such as drag, lift, and aerodynamic forces become greater than stabilizing forces such as particle weight and interparticle binding forces (Iverson et al., 1976).

The threshold shear velocity of a particle is the wind velocity required to deflate (suspend) a particle in the atmosphere (Cornelis and Gabriels, 2003). Most models that predict wet threshold shear velocity ( $u_{tw}$ ) of a particle take the form  $u_{tw} = u_t f(\text{moisture})$ , where  $u_t$  is the threshold shear velocity under dry conditions. The function  $f(\text{moisture})$  is a function of the surface moisture expressed in terms of moisture content  $w$  (kg/kg) or capillary potential (Pascal) (Cornelis and Gabriels, 2003).

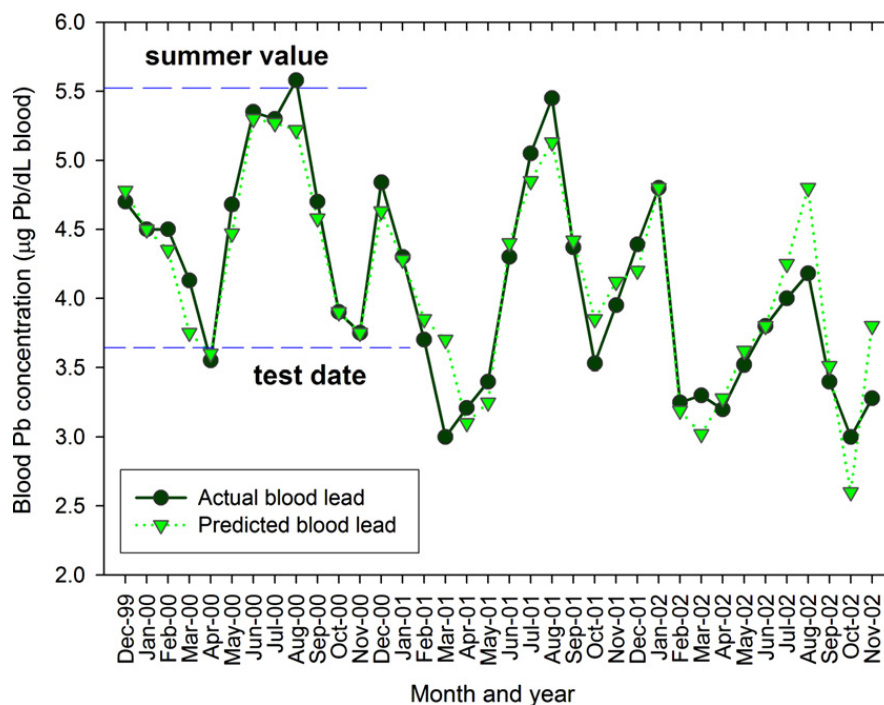
Most models of the threshold shear velocity predict a rise in deflation threshold with increasing moisture content (Cornelis and Gabriels, 2003). With decreasing soil matrix potential from a dry soil, the  $u_{tw}$  will increase exponentially until a soil matrix potential of  $-1.5$  MPa occurs, at which no soil deflation takes place. The matrix potential ( $\psi$ ) has been found to be a function of temperature ( $T$ ), air humidity ( $e/e_s$ ), molar volume of water ( $V_w$ ;  $0.0224 \text{ m}^3/\text{mol}$ ), and the universal gas constant ( $R$ ;  $8.3145 \text{ J/mol K}$ ) (Edlefsen and Anderson, 1943):

$$\psi = [(RT)/V_w][\ln(e/e_s)].$$

These equations suggest that when temperature is high, soil moisture is low and relative humidity is low, soils are susceptible to deflation. The modeling approach used in this study may have successfully explained the temporal variation in atmospheric soil concentrations because the matrix potential variables soil moisture (volumetric water content –  $V_w$ ), relative humidity ( $e/e_s$ ), and temperature ( $T$ ) were incorporated, which permits prediction of when soils are susceptible to dust emission. Since meteorological and soil moisture variables predict seasonal (temporal) atmospheric soil concentrations (Fig. 8) and children's blood Pb levels (Fig. 5), it is suggested that seasonal atmospheric soil loading (Fig. 7) may provide a proxy predictor of seasonal Pb loading.

The authors suggest that in urban areas with Pb contaminated soils (and seasonal fluctuations in climate), spatio-temporal variations in Pb loading in urban areas arises due to a complex interaction between seasonal susceptibility of soils to be resuspended, urban soil Pb contamination distributions, variations in lawn maintenance (and subsequent soil moisture) and spatial patterns of vehicular (Kuhns et al., 2001; Lough et al., 2005; Nicholson et al., 1989; Hosiokangas et al., 2004) and lawn mowing induced turbulence. Furthermore, the seasonally resuspended fine soil dust particles ( $<10 \mu\text{m}$ ) may then enter urban homes via open and looser fitting windows and doors (Lepow et al., 1974, 1975; Archer and Barratt, 1976; Sayre and Katzel, 1979; Bornschein et al., 1986; Fergusson, 1986; Davies et al., 1987; Farfel and Chisolm, 1990, 1991; Al-Radday et al., 1993; Kutlaca, 1998) which have been demonstrated to have effective penetration efficiencies ranged from 0.38 to 0.94 for  $0.02\text{--}0.5 \mu\text{m}$  particles and from 0.12 to 0.53 for  $0.7\text{--}10 \mu\text{m}$  particles (Abt et al., 2000). In addition soil Pb is also tracked into homes via soil attached to shoes (Hunt et al., 2006) and on the feet of family pets.

The source of the exterior Pb loading entering homes is likely a combination of Pb from past use of leaded gasoline with lesser amounts from Pb in paint. Clark et al. (2006) analyzed Pb isotopes in various grain size fractions in the urban soil of Massachusetts and observed that the source



**Fig. 9.** Potential clinical application of seasonality modeling. Because of the random nature of blood Pb testing dates for individuals and the seasonal pattern in children's blood Pb levels presented here, an individual test result may not provide adequate information for a clinician to assess, and potentially treat, a child patient. With awareness of the atmospheric influence on seasonality of blood Pb levels and the capacity to model this seasonality in any region with meteorologic and soil moisture data, graphs such as this could guide better assessment of a patient's longer-term Pb history and allow the clinician who is analyzing blood Pb test results from, for example, late winter-early spring to infer that the patients summertime blood Pb levels from the previous summer were likely about 50% higher. If this higher predicted level is above the level of concern for the clinician, a follow-up test in the succeeding summer might be recommended.

of the Pb in the fine fraction of soil originated from leaded gasoline, and the larger particles that do not penetrate cracks in homes (above) were composed of Pb paint particles.

The interaction between turbulence and resuspension of highly contaminated and bio-available roadside soils during periods of low evapotranspiration may be an important driver of seasonal Pb loading rates in urban areas; however, the relative contribution of the various resuspension processes (wind versus human turbulence) to urban exterior Pb loading is not known. However, it is strongly suspected that turbulence induced resuspension may be a dominant process of exterior Pb loading in urban environments. Lenschow et al. (2001) observed that at curbsides on main streets in Berlin Germany, the PM10 concentration is up to 40% higher than the urban background with half of this additional pollution due to motor vehicle exhaust emission and tire abrasion and the other half due to resuspended soil particles. In addition, in Indianapolis, Laidlaw (2001) observed that historical spatial patterns of PM10 displayed a bulls-eye pattern with PM10 concentrations highest in the inner city and decreasing with distance away from the city center. Based upon previous estimates of the contribution of resuspended soil to PM10 (above), it is suggested that elevated PM10 in areas of Pb contaminated soil may also signal elevated exterior Pb loading. The potential role of particulates of greater size than PM10 in Pb loading and as a pathway for Pb poisoning is not discounted, but little measurement

work has been done to examine the distribution of these larger particulates nor their potential role in human health.

Blood Pb seasonality is the dominant feature in urban Pb poisoning. In New York City between 1970 and 1976 it was demonstrated that seasonal variations in blood Pb levels of large populations of children (>300,000) were associated with seasonal variations in exterior Pb ( $r^2 = 0.60$ ,  $p < 0.0001$ ; Billick et al., 1979). Since blood Pb in New York was previously correlated with exterior Pb, this corroborates the observations of Laidlaw et al. (2005) and Filippelli et al. (2005) that blood Pb seasonality results from an exterior source, i.e., summertime soil and dust Pb resuspension in areas where soils have been contaminated by past use of leaded gasoline and exterior paints. Lead is not the only toxicant that children are exposed to from urban soil resuspension. The most highly enriched contaminant metals in soils of urban environments are Cu, Pb and Zn. This has been found in inner city areas of Baltimore (along with Ni and Cd; Mielke et al., 1983), Indianapolis, Indiana (Laidlaw, 2001), New Orleans, Louisiana (Mielke et al., 2001), Seville, Spain (Madrid et al., 2002), Naples, Italy (Imperato et al., 2003), Nanjing and Shenyang, China (Wu et al., 2003; Lu et al., 2004; Wang et al., 2006); and Istanbul, Turkey (Sezgin et al., 2004). Geochemical mapping studies conducted worldwide indicate that urban soil metal contamination is universal and that metal concentrations are highly correlated. In New Orleans, Louisiana, Mielke et al. (2001) found that the contaminant metals Pb, Zn, Cr and Cu were the most enriched and were

positively inter-correlated with correlation coefficients ranging from 0.83 to 0.98, while the contaminant metals Cd, Mn, Ni and V were less enriched with positively inter-correlation coefficients between 0.40 and 0.68. Mercury (Manta et al., 2002) has also been found to be inter-correlated with Pb, Zn and Cu.

One of the limitations of current blood Pb screening programs in the United States is that the assessment of blood Pb exposure is based upon analysis of blood Pb levels collected throughout the year. However, due to blood Pb seasonality, blood Pb levels in the United States generally peak in the summer and early autumn. The authors argue that children's blood Pb levels should be assessed based on the seasonal peaks in atmospheric soil loading, which for most locations would be during late summer or early Autumn to fully measure exposure levels. In the event that blood Pb sampling during this time period is not possible, we suggest that in cities with large historical child blood Pb databases, blood Pb seasonality models could be generated and validated for use in predicting peak blood Pb levels in late summer and early autumn. For example, using a blood Pb measurement in March, it would be possible to use meteorological and soil moisture data with a validated city specific model to predict a blood Pb level in August or September (Fig. 9). This would allow clinicians to better assess the blood Pb level exposure of a patient throughout the year.

## 6. Conclusions

Seasonal changes of Pb in urban children's blood, a major feature of urban Pb poisoning in the United States, may be eliminated or reduced if urban soil Pb contamination is isolated or removed. It is recommended that the spatio-temporal variation in exterior Pb loading rates be measured in urban areas where the incidence of children's blood Pb levels are elevated – although air Pb concentrations are important, particularly as they are part of the Pb loading calculation, these measurements should be expanded to the Pb flux rate to more accurately affect Pb exposure. In addition, further research should be conducted on the relationship between exterior Pb loading rates and large scale isolation of urban soil Pb using either landscaping or irrigation during periods of high evapotranspiration. One imperative is that efforts need to progress beyond the “Pb paint-only” or “soil only” arguments for Pb exposure pathways, and recognize that both of these sources act to imperil the long-term health and socio-economic futures of urban children.

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