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Review

In vitro and *in vivo* approaches for the measurement of oral bioavailability of lead (Pb) in contaminated soils: A review

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ABSTRACT

We reviewed the published evidence of lead (Pb) contamination of urban soils, soil Pb risk to children through hand-to-mouth activity, reduction of soil Pb bioavailability due to soil amendments, and methods to assess bioaccessibility which correlate with bioavailability of soil Pb. Feeding tests have shown that urban soils may have much lower Pb bioavailability than previously assumed. Hence bioavailability of soil Pb is the important measure for protection of public health, not total soil Pb. Chemical extraction tests (Pb bioaccessibility) have been developed which are well correlated with the results of bioavailability tests; application of these tests can save money and time compared with feeding tests. Recent findings have revealed that fractional bioaccessibility (bioaccessible compared to total) of Pb in urban soils is only 5–10% of total soil Pb, far lower than the 60% as bioavailable as food-Pb presumed by U.S.-EPA (30% absolute bioavailability used in IEUBK model).

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1. Introduction

Concern about potential risks from Pb in urban and garden soils have been widely expressed because of the history of soil contamination from domestic use of Pb paints, deposition of automotive Pb emissions, and from mine waste and smelter contaminated areas developed for housing (Hunt et al., 1992; Levin et al., 2008; Mielke et al., 1983). Among direct exposure pathways for Pb in urban environments, inadvertent ingestion of soil is considered the major concern compared with dermal and respiratory pathways (Chaney et al., 1989; Davies et al., 1990; Johnson and Bretsch, 2002; Paustenbach, 2000; Thornton et al., 1990; Thornton et al., 1995). In modeling risks from diet, water and soil Pb, U.S.-EPA presumes that soil-Pb is 60% as bioavailable as other dietary Pb (U.S.-EPA, 1994, 1999). The default value for soil Pb bioavailability is 30% (60% as bioavailable as Pb from water and food) in the Integrated Exposure Uptake Biokinetic (IEUBK) model (U.S.-EPA, 1994). Implications of soil Pb risk and several recommendations have been based on this assumed soil Pb fractional bioavailability figure (U.S.-EPA, 2001).

Inner city soils are considerably more contaminated than suburban soils, although exterior Pb paint scrapped to soil can cause high soil Pb contamination wherever it occurs, easily causing soil to exceed 10,000 mg Pb kg^{-1} (Murray and Hendershot, 2000). High soil Pb has become a worrisome source of risk to children because Pb has become widely dispersed in urban soils (Mielke et al., 1983, 1984, 2007; Demetriades et al., 2010; Laidlaw and Taylor, 2010). Soil Pb is a greater risk thru soil ingestion than thru uptake by garden food crops (Chaney et al., 1984; Chaney and Ryan, 1994). Thornton et al. (1985) concluded that “the concentration of lead in house dusts is significantly related to that in garden soil, and is highest at older homes.” [In the UK and some other nations, “garden” refers to all land surrounding housing, while in the US, “garden” usually refers to soils used in food and flower production.] A meta-analysis of the contribution of soil vs. housedust to blood-Pb of urban children has shown that housedust is considerably more important and interior paint Pb comprised far greater risk than soil Pb (Lanphear et al., 1998). Analysis of housedust suggested that paint, road dust, and garden soil may all be important lead sources (Hunt et al., 1992). These contributions are a function of particle size, and the importance of the contributions is dependent on whether the apportionment is based on particle population or estimated particle volume or mass (Hunt et al., 1992).

Nriagu (1972, 1974) provided the first indication that the extremely insoluble Pb mineral pyromorphite [$Pb_5(PO_4)_3$ X, where X is OH, Cl, or F] should be formed in Pb contaminated soils having some amounts of phosphorus compounds. Discovery of pyromorphite in

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urban soil was first reported when researchers in the UK found no correlation between high Pb levels in garden soil and housedust with elevated blood levels of children who lived with the soils, and scanning electron microscopy coupled with X-ray analysis of the soil grains revealed that many Pb-rich particles were composed of pyromorphite, a stable Pb mineral with a characteristic of low bioavailable Pb (Cotter-Howells and Thornton, 1991). Following this Ma et al. (1993) examined formation of chloro-pyromorphite from Pb compounds and soil Pb; their work suggested that this could be an effective soil Pb remediation technology. Later, pyromorphite was identified in some mine-waste and industrially contaminated soils (Cotter-Howells et al., 1994). Then Thornton et al. (1995) noted that "although lead of mine-waste origin may be present at elevated levels in dusts and soils, it does not necessarily contribute to elevated blood lead levels when the lead is present in relatively insoluble forms." Analysis of housedusts from hand-wipe samples collected from children in Winster, UK showed that in total, for the five soils, 28.4% of soil particles on hands fell into the "pyromorphite" class (Watt et al., 1993). Within the next few years Cotter-Howells (1996) and Cotter-Howells et al. (1999) discovered formation of this highly insoluble mineral within the rhizosphere of *Agrostis capillaris* roots, indicating that root growth could promote formation of pyromorphite, perhaps by the phosphate solubilizing activity of roots. Thornton et al. (2008) in another study, employing sequential chemical extraction, discovered that "significant proportions of Pb in brownfield contaminated soils were relatively insoluble and suggested that the low solubility of Pb did not necessarily present a risk to exposed population groups." However, bioavailability or bioaccessibility of soil/dust lead directly ingested by young children was not determined by these authors. Further, the common sequential extraction methods applied to contaminated soils actually promote transformation of the Pb species present (Scheckel and Ryan, 2004); only use of an *in situ* spectroscopic method such as X-ray absorption spectroscopy can measure the Pb species present in a contaminated soil material.

At present, for total Pb concentration, U.S. EPA and U.S. Department of Housing and Urban Development (HUD) have established a 400 ppm standard for children's play areas and an average of 1200 ppm standard in the remainder of the yard (U.S.-EPA, 2001). The Agency believes that more than 12 million homes exceed a 400 ppm yard-wide standard while at 1200 ppm Pb threshold, only 4.7 million homes exceed the standard.

Lead bioavailability in soils is largely controlled by phosphate, iron oxides, organic matter and pH. Iron (oxy)hydroxides [such as goethite and amorphous $Fe(OH)_3$ (ferrihydrite)] and organic matter create surface sorption or chelation sites for binding Pb^{2+}, and dissolved phosphate causes its precipitation (Appel and Ma, 2002; Brown and Chaney, 2003; Ryan et al., 2004; Traina and Laperche, 1999). In a similar way, an increase in pH also decreases Pb mobility and bioavailability as fewer H^+ ions are available to compete with Pb^{2+} ions for binding sites (Cao et al., 2008; Hettiarachchi and Pierzynski, 2004) in addition to the formation of Pb carbonate minerals.

For over a decade, researchers have explored *in situ* soil treatments such as phosphate (e.g., single/triple superphosphate, phosphoric acid, rock phosphate, monoammonium phosphate, bone meal), and amorphous iron oxides to reduce the phyto- and bioavailability of soil Pb (Basta et al., 2001; Porter et al., 2004; Brown et al., 2004; Brown et al., 1999; Cao et al., 2008; Cotter-Howells and Caporn, 1996; Hettiarachchi et al., 2001; Hettiarachchi and Pierzynski, 2002; Hodson et al., 2001, 2000; Laperche et al., 1996, 1997; McGowen et al., 2001; Moseley et al., 2008; Ryan et al., 2004; Sneddon et al., 2006, 2008; Sterrett et al., 1996; Yang et al., 2001). As noted above, while urban soils often have evidence of pyromorphite formation (Cotter-Howells, 1996), Pb mine wastes usually contain very low phosphate levels inhibiting natural formation of pyromorphite (Harwood et al., 1987).

Use of conventional soil extraction methods [5 mM diethylenetriaminepentaacetic acid (DTPA), 0.01 M $Ca(NO_3)_2$] to assess phytoavailable Pb was unable to identify the significant reduction in soil Pb bioavailability induced by application of phosphate or Fe-rich biosolids which were demonstrated through *in vivo* and *in vitro* trials (Brown et al., 2003; Ryan et al., 2004). Because conventional soil tests yield results unrelated to bioavailability of soil Pb risk to children, alternative tests are needed. In a recent study on soils contaminated with Pb and As, bioaccessibility of As and Pb measured in artificial gastric and small intestinal solutions decreased with phosphate and iron application except for the bioaccessibility of As in the gastric phase with single super phosphate addition (Cui et al., 2010). Combined application of phosphates and iron could be an effective approach to lower bioaccessibility of As and Pb, but had opposing effects on mobility of As and Pb in contaminated soils, the authors concluded.

Trials using animals for measurements of soil Pb *bioavailability* are expensive (about $30,000 per test soil with swine) and time consuming (Casteel et al., 2006). Thus, several chemical test methods have been developed to measure soil Pb *bioaccessibility* to animals (*in vitro* approach) starting with Ruby et al. (1993). These kind of *in vitro* approaches need not necessarily represent all the physiological processes provided bioaccessible Pb is well correlated with bioavailable Pb (Ruby et al., 1993; Drexler and Brattin, 2007; Scheckel et al., 2009). In order to obtain improved bioaccessibility tests, numerous authors have conducted tests of variation of the methods by adding different digestion factors from the human digestive system assuming that the more lifelike the method, the better the reliability would be (e.g., Oomen, 2000; Oomen et al., 2002, 2003a,b, 2004, 2006). Others (e.g., Thums et al., 2008) have simply used 0.12 M HCl ignoring the pH buffering aspect of stomach secretions and ingested soil and foods. In our view, the relevant issue is the relationship of the bioaccessibility test and an acceptable bioavailability measure, where bioavailability to humans is the ultimate best estimate.

Ruby et al. (1993) introduced a chemical extraction method to estimate the bioavailability of soil Pb which was well correlated with bioavailability measured by rabbits, and this extraction result was labeled "*bioaccessibility*" to make clear it was not a biological measurement of bioavailability. Within the next few years, Ruby et al. (1996, 1999) extended the development of their earlier bioaccessibility extraction method and called it the Physiologically-Based Extraction Test (PBET). The modified test was more complicated, but seemed well correlated with results of available feeding studies. After this development, there were many studies along similar lines by several researchers (Oomen, 2000; Oomen et al., 2002, 2003a,b, 2004, 2006) who tried to see if some chemicals similar to gastric juice/GI secretions could simulate living animal digestion behavior for measurement of Pb (and other metals as well) entry to the systemic circulation (Intawongse and Dean, 2006; Hooda, 2010). Presently, a variety of *in vitro* GI methods have been developed and proposed, with multi-phase tests involving a wide range of pH (1.07–7.5), different size fractions of soil (<2.00 mm to <125 μm), residence time (5 s to 16 h), temperature 20–37 °C, and soil:solution ratio (1:2 to 1:5000) (Wragg and Cave, 2002; Intawongse and Dean, 2006). The pH values used in such tests don't necessarily represent a close approximation of the human GI system (Hooda, 2010). Mean stomach fasting pH ranges from 1 to 4 in children (Anderson et al., 1999), and from 1.5 to 2 in adults (Charman et al., 1997), but when food is consumed stomach pH commonly rises to 6 (Malagelada et al., 1979).

Recently Drexler and Brattin (2007) introduced a modified *in vitro* procedure for estimation of lead bioaccessibility that was well correlated with the results of bioavailability tests using pigs fed mine waste and smelter contaminated soils, and this method

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