



Use of simulated epithelial lung fluid in assessing the human health risk of Pb in urban street dust



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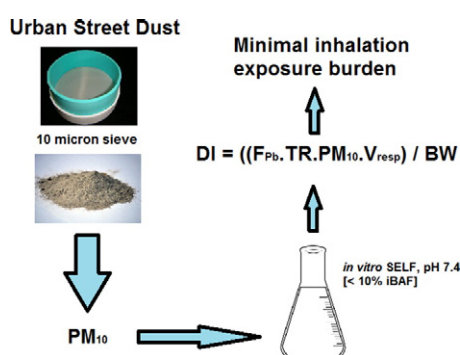
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HIGHLIGHTS

- Exposure assessment of inhalation pathway
- High pseudo-total Pb in PM₁₀ fraction of urban dusts
- Extraction using simulated epithelial lung fluid
- Inhalation bioaccessibility of Pb in PM₁₀ < 10%
- Exposure burden from the inhalation pathway observed to be minimal

GRAPHICAL ABSTRACT



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ABSTRACT

In many urban contexts, non-dietary Pb exposure from street dusts may add to the overall exposure burden, and the presence of high total Pb content is well documented in urban street dust from across the globe. Given the increasing recognition of the potential adverse health effects from both the quantity and the chemical and physical composition of the inhaled fraction, and the recognition that it is the soluble fraction rather than the total element content that has more direct links to health effects, attention has focused in this study on the human health risks via this exposure pathway. In order to investigate the environmental exposure to Pb from the inhalation of urban street dusts, a newly developed *in vitro* simulated epithelium lung fluid (SELF) has been applied to the < 10 μm fraction of urban street dusts. In this context, 21 urban street dust samples, across five UK cities, were selected based on their high pseudo-total Pb content. The work revealed that inhalation bioaccessibility, and hence inhalation dose, varied across the cities but was generally found to be low (< 10%). Indeed, the lung bioaccessibility was far lower (% lung bioaccessibility ranged from 1.2 to 8.8) than is currently applied in two of the most commonly employed risk assessment models i.e. the Integrated Exposure Uptake Biokinetic model (IEUBK, USA) and the Contaminated Land Exposure Assessment model (CLEA, UK). The estimated inhalation dose (for adults) calculated from the PM₁₀ bioaccessibility ranged from 7 ng kg⁻¹ BW day⁻¹ (Edinburgh) to 1.3 ng kg⁻¹ BW day⁻¹ (Liverpool). The results indicate a low potential inhalation bioaccessibility for Pb in these urban street dust samples when modelled using the neutral pH conditions of the SELF.

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1. Introduction

Particulate matter (PM) is ubiquitous in the atmosphere and is comprised of a wide range of materials sourced from both natural processes and human activity: sea-salt, combustion-derived carbonaceous particles, sulfates, silicates, oxides, carbonates, alloys, glass and biogenic material (Kelly and Fussell, 2012). Lead is one of the most enriched metals in urban particulate matter (De Silva et al., 2016; Goix et al., 2016; Laidlaw et al., 2012; Lei et al., 2016; Sharma and Pervez, 2003; Xu et al., 2012; Zereini et al., 2005) and despite the phasing out of Pb in fuel in many countries around the world it continues to be a potentially harmful element (PHE) of concern in the urban environment. Lead is a well-known neurotoxin, with exposure leading to neurobehavioural effects in children (e.g. Lanphear et al., 2005), and cardiovascular (hypertension) and renal toxicity effects in adults (EFSA, 2010). Indeed, a growing number of studies suggest that the threshold for clinical concern should be reduced to a blood Pb level below $5 \mu\text{g dL}^{-1}$ (Budtz-Jorgensen et al., 2013; Navas-Acien et al., 2009; Zahran et al., 2013). Whilst there are strict regulations in many developed countries on the use and release of lead into the environment, particularly, on the former use of lead in petrol as an anti-knock additive (Kaysi et al., 2000; Kummer et al., 2009), this is not the case with most developing countries where leaded petrol is still in use (Gwilliam, 2003). Over the years the emphasis has been on leaded fuel (Oudijk, 2010; Nriagu et al., 1996; Romieu et al., 1992), but there are many other activities that release Pb into the environment particularly in the urban/industrial setting: activities such as metal mining, smelting and processing, the use of Pb in lead-acid batteries, pigments, alloys, lead wool, chemical manufacturing, cables, solders, plumbing components, food cans, coal combustion, lead based paint (including that in road markings), and industrial waste (Ajmore-Marsan and Biasioli, 2010; Brown and Longoria, 2010; De Silva et al., 2016; Laidlaw and Taylor, 2011; Mielke et al., 2010; Shen et al., 2002). Studies have shown that the Pb retained in soil/dust because of anthropogenic activity typically occurs in highly bioavailable, exchangeable and carbonate forms, whereas, Pb retained because of natural occurrence is often found in residual or less-bioavailable forms (Chlopecka et al., 1997; Cox et al., 2013; Palumbo-Roe et al., 2013; Pelfrène et al., 2012; Laidlaw and Filippelli, 2008; Reis et al., 2014; Ruby et al., 1994). Direct exposure to urban dust through inhalation is expected thus the respiratory tract is a potentially significant pathway through which urban dusts can enter the human body.

Urban dust represents a significant health risk to humans due to its small size and ubiquitous nature. Though the human respiratory system is naturally equipped with coordinated mechanisms to provide protection against inhaled particulates not all inhaled dusts are expelled; in addition, there is a significant possibility that the soluble fraction would be dissolved by interstitial lung fluids. In assessing risk to humans, particle size is a very important parameter. The particle size (and shape) of urban dusts and associated chemical composition determine their behaviour in the human respiratory system and ultimately their pathogenic mechanism. The particle size fraction considered potentially significant in this study is the $<10 \mu\text{m}$ as it represents the easily inhalable fraction. This particle size is of growing concern because it can easily be carried and re-mobilised by air flows generated by wind, traffic or human movement. Moreover, it has been shown that toxic element concentrations in urban dusts increase with decreasing particle size fraction (Duong and Lee, 2009; Duong and Lee, 2011). Smaller particle size fractions have an increased surface to volume ratio and as result this provides an increased surface area for the deposition of potentially harmful elements (PHEs), relative to a similar mass of larger particles. Therefore, the $<10 \mu\text{m}$ potentially represents a higher risk to human health.

Lead remains of concern in the urban environment, particularly those associated with legacy Pb from fossil-fuel and paint-derived sources, such as in cities with a rich industrial heritage and in smelting

and mining communities (e.g. Mielke et al., 2001; Mielke et al., 2010; Mielke et al., 2011; Taylor et al., 2013; Taylor et al., 2014). A survey (Thornton et al., 1990) of metals in UK dusts has shown that most of the dusts collected from urban environments had widespread Pb contamination ranging from 172 to 9600 mg/kg; other studies have reported (Wang et al., 2006) that for example in China, 80% of children exposed to urban dust had an excessive blood lead level. Similarly, the current authors had previously investigated the human health risk from Pb in urban street dust in five UK cities (Elom et al., 2014) using an oral bioaccessibility protocol. The study (Elom et al., 2014), which covered 75 dust samples from five cities, revealed that both pseudo-total Pb content and the oral bioaccessible Pb fractions were significantly elevated in nearly a third of the dust samples analysed. In vitro experiments use simulated fluids that are used to represent the natural physiological fluids in assessing the human health risks from contaminants. Exposure from environmental contaminants are normally considered against three exposure pathways, specifically, ingestion, inhalation and dermal contact. The oral (ingestion) pathway, considered the critical exposure route when modelling public open space, residential, and commercial settings (DEFRA, 2014a), has been the focus of much research to assess the human risk assessment from exposure to environmental contaminants predominantly via hand-to-mouth (deliberate) or unintentional ingestion of soils and related materials (Boisa et al., 2013; Cai et al., 2016; Intawongse and Dean, 2006; Li et al., 2014; Lorenzi et al., 2012; Okorie et al., 2012; Oomen et al., 2002; Smith et al., 2011; Wragg et al., 2009; Wragg et al., 2011). However, given the increasing acknowledgement of the association of PM concentrations (the chemical composition, as well as the physical presence) with both short-term and long-term health consequences (Kelly and Fussell, 2015), and the recognition that it is the soluble fraction rather than the total element content that has more direct links to health effects (Adamson et al., 2000; Ghio and Devlin, 2001; Heal et al., 2005), the inhalation exposure pathway was the focus of this current study. Indeed, in many urban and high dust generating contexts it is now timely, given increasing evidence of the link between PM10s and a range of human disease pathologies (Kelly and Fussell, 2012; Kelly and Fussell, 2015; Uzu et al., 2011), to consider the potential inhalation burden and bioaccessibility of PHEs in airborne PM and other environmental samples with a particle size fraction $<10 \mu\text{m}$. The probability of inhalation depends on the particle size fraction, air movement within the exposure routes, and breathing rate. Inhalable particulate matter could be inhaled through the nose and thus the fate of inhaled particulates depends on the nature of the physiological fluids and physiochemical properties of the particulates. The inhalable particle size fraction (aerodynamic diameters $<10 \mu\text{m}$) penetrates, deposits and is retained in different compartments of the human respiratory tract with the larger components commonly found in the nasopharynx and tracheobronchial region whilst the finer ($<1-2 \mu\text{m}$) particles are deposited in the deepest region (alveolar) (Lippmann and Albert, 1969; Gokhale and Patil, 2004; Plumlee et al., 2006). An understanding of the respiratory compartments and their functions, as well as chemical composition is fundamental in formulating fluids that truly represent the respiratory system. Simplistically, the respiratory system is comprised of three compartments; the nasopharynx, tracheobronchial, and the pulmonary (Task Group on Lung Dynamics, 1966; U.S. EPA, 2008). Classification of the respiratory system into different compartments gives a clearer understanding of the processes involved in particle inhalation, deposition and removal. Neutral synthetic lung fluids, largely based on modifications of the original 'Gamble's solution', are widely used for the exposure assessment of humans to inhalable pollutants (Wragg and Klinck, 2007; Caboche et al., 2011; Lima et al., 2013). More recently, the crucial (though often unclear) role of high molecular mass proteins, antioxidants and surfactant lipids in determining bioaccessibility has been recognised, along with the development and application of a new generation of in vitro lung fluid formulations (Boisa et al., 2014; Gray et al., 2010; Li et al., 2014; Stebounova et al., 2011). The development

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