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Particle size effects on bioaccessible amounts of ingestible soil-borne toxic elements

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HIGHLIGHTS

- Ingestion of contaminated soils and soil-derived dusts poses a risk to human health.
- Bioaccessible amounts of 9 elements in multi-contaminated soils were determined.
- Soil particle size significantly affected bioaccessible amounts of potentially toxic elements.
- It also had significant effects on bioaccessibility of As and Al in intestinal environment.
- Further division of the <0.25 mm soil fraction is necessary for better health risk assessment.

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ABSTRACT

The unified BARGE method was used to examine the effects of soil particle size on the bioaccessible amounts of potentially toxic elements in multi-contaminated soils from a closed landfill site. The results show that bioaccessible As, Al, Cd, Cr, Cu, Mn, Ni, Pb and Zn increased with decreasing soil particle size and the <0.002 mm soil fraction contained much greater amounts of the bioaccessible elements, as compared to other soil fractions (0.002–0.063 mm, 0.063–0.125 mm, and 0.125–0.250 mm). As, Al and Cr had much lower bioaccessibility, as compared to the six cationic heavy metals. In contrast with other elements, As bioaccessibility tended to be higher in the gastrointestinal phase than in the gastric phase. There was a significant soil particle size effect on bioaccessibility of As and Al in the gastrointestinal phase: As bioaccessibility decreased with decreasing particle size, and the finer soil fractions tended to have a higher Al bioaccessibility, as compared to the coarser soil fractions. The research findings prompt the need for further division of soil particle size fractions in order to more accurately assess the bioaccessible amounts of soil-borne potentially toxic elements in contaminated lands.

1. Introduction

In vitro methods to simulate dissolution of soil-borne potentially toxic elements (SBPTEs) in human gastrointestinal tract have gained increasing acceptance as a routine protocol for assessing the human health risk from ingesting SBPTEs (Oomen et al., 2002; Li et al., 2015). The unified bioaccessibility method (UBM) developed by the Bioaccessibility Research Group of Europe (BARGE) recommended the use of <250 μm soil fraction for assessment of bioaccessibility of SBPTEs (Wragg et al., 2011; Denys et al., 2012; Collins et al., 2015). This assumes that all soil particles with a diameter less than 250 μm are proportionally ingested by human during a given period of time. However, it is highly likely that finer soil particles tend to be more ingestible, as compared to coarser soil particles. For example, Ikegami et al. (2014) found that over 90% of soil particles adhered to children's hands were <100 μm in diameter. Ingestible dust particles derived from soils could be much finer than dermally adhered soil particles (Mahowald et al., 2014). Since heavy metal concentrations tend to be higher in the finer fraction than in the coarser fraction (Li et al., 2014; Zong et al., 2016), the standard method using <250 μm soil fraction has its limitations when being used to assess human health risk associated with soil ingestion by children or ingestion of soil-derived dusts in the areas adjacent to contaminated lands (Martin et al., 2015). In addition, most of the work reported so far focused on a limited number of SBPTEs, especially lead and arsenic (Basta et al., 2007; Denys et al.,

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2007; Palumbo-Roe and Klinck, 2007; Bosso and Enzweiler, 2008; Denys et al., 2009; Broadway et al., 2010; Roussel et al., 2010; Lu et al., 2011; Mingot et al., 2011; Appleton et al., 2012). For soils contaminated with multiple SBPTEs, the damaging effects of ingesting such soil materials on human health could be much more severe due to combined toxicity of various SBPTEs. It is therefore important to understand the simultaneous responses of different SBPTEs to chemical attack from gastrointestinal fluids in order to better assess the combined toxic effects of multiple SBPTEs on human beings exposed to different sizes of ingestible soil/soil-derived dust particles.

In this study, four further-divided fractions of <250 μm soil fraction were used to examine the effects of soil particle size on extracting a range of potentially toxic elements from contaminated soils using the UBM in vitro gastrointestinal tract method. The objective was to determine whether further particle size division is necessary in order to more accurately assessing the human health risk from ingestion of multi-contaminated soil/soil dust particles originated from contaminated lands.

2. Materials and methods

2.1. The soil materials

Two multi-contaminated soil materials were selected for this study following a soil screening test to identify appropriate soil samples from 27 locations within a closed landfill site adjacent to a residential area in the Greater Manchester region, England. This site received substantial amounts of industrial wastes during the period of Industrial Revolution. The soil samples were collected from the surface soil layer (0–10 cm), which is a readily available source of contaminated soil materials for human ingestion. The soil samples chosen contained a wide range of multiple SBPTEs at elevated concentrations with Sample FG 1 having a slightly alkaline pH and Sample FG2 having a slightly acidic pH (Table 1).

After collection, the soil materials were oven-dried at 40 °C and then crushed to pass a series of sieves (2.00, 0.25, 0.125 and 0.063 mm) to obtain the following soil particle fractions: 0.25–2.00 mm, 0.125–0.250 mm, 0.063–0.125 mm, and <0.063 mm. The <0.063 mm fraction was then suspended in deionized water and stirred by a magnetic stirrer for 1 h. The dispersed soil suspension was placed in a 1 L cylinder and stood for 24 h to allow the >0.002 mm soil particles to settle (McCarty et al., 2016). The remaining suspended materials were then separated from the settled soil materials to obtain two further soil particle fractions: <0.002 mm and 0.002–0.063 mm. The 0.25–2.00 mm fraction was not used in this experiment. The four soil fractions tested in this study therefore include: <0.002 mm, 0.002–0.063 mm, 0.063–0.125 mm and 0.125–0.250 mm. The percentage share of these different soil fractions in the <0.250 mm soil mass was 1% (<0.002 mm fraction), 13% (0.002–0.063 mm fraction), 32% (0.063–0.125 mm fraction) and 54% (0.125–0.250 mm fraction) for Sample FG1, and 1% (<0.002 mm fraction), 20% (0.002–0.063 mm fraction), 32% (0.063–0.125 mm fraction) and 47% (0.125–0.250 mm fraction) for Sample FG2 (Table 1).

Table 1
Some major physical and chemical characteristics of the soil materials used in this study.

Soil parameter	FG1	FG2
pH	7.12	6.62
EC (dS/m)	0.039	0.035
Organic carbon content (%)	1.11	1.27
0.125–0.25 mm fraction (%)	54	47
0.063–0.125 mm fraction (%)	32	32
0.002–0.125 mm fraction (%)	13	20
<0.002 mm fraction (%)	1	1
Total As (mg/kg)	29.6	27.2
Total Al (mg/kg)	21,013	19,247
Total Ca (mg/kg)	32,166	32,173
Total Cd (mg/kg)	6.19	9.28
Total Cr (mg/kg)	68.8	68.8
Total Cu (mg/kg)	2768	3377
Total Fe (mg/kg)	28,131	21,833
Total K (mg/kg)	1859	2423
Total Mn (mg/kg)	3865	3511
Total Na (mg/kg)	959	799
Total Ni (mg/kg)	811	925
Total Pb (mg/kg)	1498	1597
Total Zn (mg/kg)	1276	1031

2.2. Element extraction procedure

The unified BARGE method (Denys et al., 2012) was slightly modified and used for extraction of a range of elements in the soil samples. Briefly, for the gastric phase extraction, 9.0 mL of synthetic saliva was added to 0.6 g of each soil sample in a 50 mL centrifuge tube. After shaking by hand for 10 s, 13.5 mL of synthetic gastric fluid was added to the tube. The pH of the suspension was adjusted to 1.20 ± 0.05 with HCl or NaOH. The capped tube was placed in a water bath and the content was incubated at 37 °C for 1 h. After incubation, the gastric phase sample was centrifuged for 15 min at 4500 rpm. The supernatant was then filtered, acidified with 0.5 mL concentrated HNO_3, and stored at 4 °C prior to analysis. For the gastrointestinal phase extraction, a subsample after gastric phase extraction described above was further treated with 9 mL of synthetic bile and 27 mL of synthetic duodenal fluids. The pH of the extracting solution was adjusted to 6.3 ± 0.5. The sample was then placed in a water bath and the content was incubated at 37 °C for a further 4 h. After incubation, the gastrointestinal phase sample was centrifuged for 15 min at 4500 rpm. The supernatant was then filtered, acidified with 1.0 mL concentrated HNO_3 and stored at 4 °C prior to analysis.

2.3. Analytical methods

The total concentration of various elements in different soil particle fractions was determined by ICP-OES (Agilent Varian 720-ES) after HNO_3-HCl-H_2O_2 digestion in a microwave digester (CEM Corporation MARS 5 Digestion Microwave System). Element concentration in the gastric and gastrointestinal phase extracts was also measured by ICP-OES. pH in various soil-fluid mixtures was measured using a calibrated pH meter (JENWAY-3510).

2.4. Statistical analysis methods

Significant difference analysis was conducted using SPSS (version 17.0). The experimental data were analyzed by one-way analysis of variance (ANOVA) and the means compared using significant difference (Duncan) method at 5% level. The relationships between the total concentration of various elements and that extracted by the simulated gastric or gastrointestinal fluids for different soil particle fractions were determined by linear regression.

2.5. Quality control and quality assurance

All the chemicals used in the experiment were of analytical grade. Ultrapure water was used throughout the entire experiment. The experiment was performed in triplicate and all the samples were run in a single batch to allow reasonable comparison.