Soil particle size fraction and potentially toxic elements bioaccessibility: A review

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ABSTRACT

In the last decade, extensive studies have been conducted to quantify the influence of different factors on potentially toxic elements (PTE) bioaccessibility in soil; one of the most important is soil size fraction. However, there is no agreement about the size fraction and the methods to investigate bioaccessibility, as very few review articles are available on soil PTE bioaccessibility and none addressed the influence of particle size on PTE bioaccessibility. This study provides a review of the relations between PTE bioaccessibility and soil particle size fractions. The available research indicates that PTE bioaccessibility distribution across different size fractions varies widely in soil, but a general trend of higher bioaccessibility in finer size fraction was found. The different elements may exhibit different relationships between bioaccessibility and soil size fraction and, in some cases, their bioaccessibility seems to be more related to the source and to the chemico-physical form of PTE in soil. Often, soil pollution and related health risk are assessed based on PTE total concentration rather than their bioaccessible fraction, but from the available studies it appears that consensus must be pursued on the methods to determine PTE bioaccessibility in the fine soil size fractions to achieve a more accurate human health risk assessment.

1. Introduction

Soils are a source and a sink of various inorganic and organic compounds, natural or anthropically-derived, depending on the soil type, soil use, parent material and anthropic pressure, e.g due to industrial and mining activities, traffic, waste disposal and others causing contamination and related environmental and health impacts (Alloway, 2013).

Soil contamination from potentially toxic elements (PTE) has become of great concern worldwide in the past few decades as PTE, contrarily to most organic pollutants, cannot be degraded and are difficult to remove, persisting for a long time in the soil environments (Ma et al., 2009; Frohne et al., 2014; Padoan et al., 2020a).

PTE may occur naturally in soils; however additional contributions come from anthropogenic activities such as mining, smelting, fertilization, and urbanization, leading to their build up and gradual release into the environment. In these cases, soils could become a sink of PTE, causing various degrees of pollution in urban, agricultural, and industrial contexts (Ajmone-Marsan et al., 2019; Cai et al., 2016; Li et al., 2014; Mehta et al., 2019; Mokhtarzadeh et al., 2020).

Most studied PTE include the xenobiotic and highly toxic lead (Pb), arsenic (As), chromium (Cr), cadmium (Cd) and mercury (Hg), as well as some essential elements, such as copper (Cu), zinc (Zn), cobalt (Co), manganese (Mn) and nickel (Ni) that can become toxic when accumulated at high concentrations in soils (Alloway et al., 2013; Li et al., 2018). These studies rise from the fact that human exposure to PTE in soils is a major public concern and is associated with serious risks to human health. PTE in soil can exert their toxicity to human and plants when they encounter the receptor or are absorbed. Human exposure scenarios to PTE in soil are, therefore, inhalation, inadvertent oral ingestion, and dermal contact (Shi et al., 2011; Ruby and Lowney, 2012). Even though exposure routes are manifold, inadvertent oral ingestion has been considered, and calculated, to be the main exposure path in most health risk assessment studies, especially through outdoor hand-to-mouth activities by children (Oomen et al., 2002; USEPA (United States Environmental Protection Agency), 2011; Pelfrêne et al., 2012).

Risk estimation due to oral ingestion of contaminated soil particles should be defined considering potential exposure, thus quantity and quality of ingested particles (USEPA (United States Environmental

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Protection Agency), 2011), and potential adsorption of PTE after ingestion (Paustenbach, 2000). Since not all the ingested PTE are easily released from the soil matrix, and thus adsorbed from the body, methods for the assessment of bioaccessible concentrations have been developed. The *bioaccessibility* of an element represents the fraction of the element/compound that is soluble in the gastrointestinal tract and, therefore, available for absorption (Ruby et al., 1999). Soil PTE absorption can be assessed using in vivo methods, to evaluate the bioavailability of soil PTE alone or in comparison with a PTE-salt reference dose, or by in vitro methods, using digestion models based on human physiology that have been developed to reduce the difficulty and the costs of in vivo studies (Oomen et al., 2002; Juhasz et al., 2009).

Although in the last years many studies investigated PTE bioaccessibility in different soils from different areas with different contamination histories (Cui et al., 2018; Valido et al., 2018; Li et al., 2020a; Mehta et al., 2020), the factors controlling bioaccessibility, such as elemental speciation and soil physical properties, have seldom been investigated.

In this regard, an important soil property which influences PTE bioaccessibility appears to be the soil particle-size distribution since PTE partitioning in various soil particle sizes can affect both exposure and PTE extractability. For example, as the ingestion of contaminated soil is largely the result of hand-to-mouth contact, we have to consider the size fraction adhering to human hands, especially of children, as for adults soil ingestion is considered to be lower (Yamamoto et al., 2006; USEPA (United States Environmental Protection Agency), 2011). Various studies assessed the soil particle size of relevance, although a definitive upper size cutoff has not yet been defined (Yamamoto et al., 2006; Siciliano et al., 2009; Ruby and Lowney, 2012). Since soil texture and organic matter directly influence dust adherence to hands, different studies achieved different cutoff sizes, ranging from 50 to 250 μm in diameter (Driver et al., 1989; Edwards and Lioy, 1999; Yamamoto et al., 2006; Ruby and Lowney, 2012). Moreover, fine soil particles are normally more concentrated in clay minerals and organic matter, influencing the total concentration and the extractability of PTE in the finer soil fraction (Ajmone-Marsan et al., 2008; Madrid et al., 2008).

Many studies in recent years started investigating the behavior of PTE in different soil size fractions but methods, soil fractions investigated and purposes of the studies have been quite dissimilar, making it hard to draw some general conclusions on PTE bioaccessibility and a possible unified method. Up to now, research groups have tested in vitro PTE bioaccessibility in different soil size fractions, however they may not be comparable as no in vivo data was obtained in different soil fractions.

This review is aimed to i) compare the most commonly used methods to estimate PTE bioaccessibility in soil particles to identify the optimal size for soil bioaccessibility and risk assessment; ii) discuss the variation of PTE bioaccessibility within particle sizes; iii) compare the efficacy of the methods to estimate metals and metalloids bioaccessibility.

2. PTE bioaccessibility methods

The in vitro bioaccessibility is defined as the fraction of an element that is soluble in simulated biological conditions such as, for oral bioaccessibility, the gastric and intestinal juices (Ruby et al., 1999; Juhasz et al., 2009). The determination relies on the extraction of the elements with solutions mimicking the chemical conditions encountered in the human stomach or intestines (Turner, 2011). The bioaccessible fraction measured using in vitro methods is usually greater than the actual bioavailable fraction (Paustenbach, 2000) and its use provides a conservative measure of bioavailability. These tests have been mostly developed using soil contaminated with one or few elements, but they have been extended to various PTE for which they have not yet been demonstrated to be good analogs of in vivo conditions.

The most used methods include the Solubility/Bioavailability Research Consortium method (SBRC), the Simple Bioaccessibility

Extraction Test (SBET), the In Vitro Gastrointestinal method (IVG), the Physiologically Based Extraction Test (PBET), the Deutsches Institutfür Normunge V method (DIN), and the Unified BARGE Method (UBM) which is based partly on the previously developed Dutch National Institute for Public Health and the Environment method (RIVM) (Ruby et al., 1996; Rodriguez et al., 1999; Oomen et al., 2002; Drexler and Brattin, 2007; Juhasz et al., 2009; Wragg et al., 2011). The SBET method consists, however, only in the gastric phase extraction of the SBRC method and, thus, in this manuscript, will be called as SBRC gastric to diminish the acronyms.

In brief, soil aliquots are mixed with solutions simulating the gastric and intestinal environments, incubated for a particular time with a specific soil/solution ratio and then extracted PTE concentrations are quantified (Fig. 1).

In vitro PTE relative bioaccessibility (RBA) can be calculated as follows:

$$PTE\ RBA(\%) = \frac{in\ vitro\ concentration}{total\ concentration} \times 100$$

All the solutions simulating the gastrointestinal fluids have a physiologically-based composition, except the SBRC gastric phase, and they are mainly composed of salts to control pH and competing ions, complexants as organic acids, enzymes, and proteins. The differences between these physiologically driven methods are mostly variations in the gastrointestinal fluid composition and extraction parameters as the incubation time in the gastric and small intestinal phases or the solution pH. For example, in the *gastric phase*, the soil residence time is 1 h for PBET, SBRC, UBM, and IVG but 2 h for DIN. In the *small intestinal phase*, the extraction time is 1 h for IVG, 2 h for UBM, 4 h for PBET and, 6 h for DIN (Li et al., 2020a).

A scheme of the main parameters and of the differences involving SBRC, PBET, IVG, DIN, and UBM methods is reported in Table 1.

These methods have been established starting from one or a limited number of key soil contaminants, chosen in view of their toxicity to humans and of their presence in most of contaminated sites. All the five procedures have been tested for As and Pb using a large number of contaminated soils in different studies, while for Cd, compared to As and Pb, only 4 studies provided paired bioaccessibility and bioavailability data (Li et al., 2020a). In vitro assays were tested to assess the correlation between in vivo and in vitro results, the most important result for the validation of the methodology, along with its repeatability and reproducibility (Wragg et al., 2011).

After their first publication the methods have been stretched and utilized for a wide variety of elements for whom the methods had not been yet validated, counting on the fact that the reliability of the results for Pb, Cd and As could be extended to elements having different chemical reactivity.

In recent years, some studies assessed the comparability of the different in vitro methods using a range of contaminated or reference soil samples, and results varied significantly depending on target element, and contamination source (Juhasz et al., 2015; Li et al., 2020a). Generally, gastric phases were better correlated with in vivo data, with SBRC and UBM methods, the simplest one (SBRC) and the most complex one (UBM) of the assays, producing the highest bioaccessibility values, providing more conservative results. Both methods were also been reported as the best assays for As, Pb and Cd bioaccessibility measurement in contaminated soils (Li et al., 2015; Li et al., 2020a), with the gastric phase of SBRC also accepted by USEPA to estimate Pb bioaccessibility (USEPA (United States Environmental Protection Agency), 2013). For this review, the above-depicted in vitro methods will be considered as comparable, to focus on the soil-dependent factors affecting the results.

2.1. Factors affecting PTE bioaccessibility

Beside the chemical nature of the analytes, and of the solubility and the extractability of elements in the specific conditions adopted by the
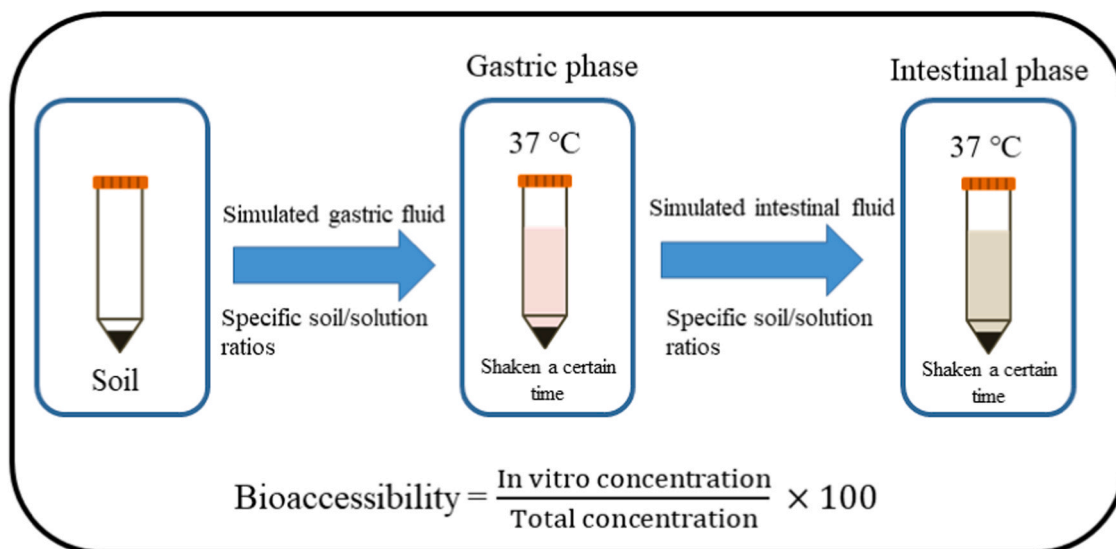
Fig. 1. Schematic diagram of in vitro soil PTE bioaccessibility test procedure.

Table 1

Composition of gastrointestinal fluids and operational parameters of SBRC, PBET, IVG, DIN, and UBM methods.

Method	Phase	Fluid composition (L^{-1})	pH	Soil:Solution Ratio	Duration (h)	Reference
SBRC	Gastric	30.03 g glycine	1.5	1:100	1	Drexler and Brattin (2007)
	Intestinal	1.75 g bile, 0.5 g pancreatin	7.0	1:100	4	
PBET	Gastric	1.25 g pepsin, 0.5 g malate, 0.5 g citrate, 0.42 mL lactic acid, 0.5 mL acetic acid, 8.77 g NaCl	2.5	1:100	1	Ruby et al. (1996)
	Intestinal	1.75 g bile, 0.5 g pancreatin	7.0	1:100	4	
IVG	Gastric	10 pepsin, 8.77 g NaCl	1.8	1:150	1	Rodriguez et al. (1999)
	Intestinal	3.5 g bile, 0.35 g pancreatin	5.5	1:150	1	
DIN	Gastric	1 g pepsin, 3 g mucin, 2.9 g NaCl, 0.7 g KCl, 0.27 g KH$_2$PO$_4$	2.0	1:100	2	Juhasz et al. (2009)
	Intestinal	9.0 bile, 9.0 pancreatin, 0.3 g trypsin, 0.3 g urea, 0.3 g KCl, 0.5 g CaCl$_2$, 0.2 g MgCl$_2$	7.5	1:100	6	
UBM	Saliva	0.45 g KCl, 0.44 g NaH$_2$PO$_4$, 0.1 g KSCN, 0.28 g Na$_2$SO$_4$, 0.15 g NaCl, 0.1 g urea, 0.15 g amylase, 50 mg mucin, 15 mg uric acid, NaOH	6.5	1:15	0.25	Wragg et al. (2011)
	Gastric	1.85 g NaCl, 0.163 g NaH$_2$PO$_4$, 0.41 g KCl, 0.2 g CaCl$_2$, 0.15 g NH$_4$Cl, 0.32 g glucose, 20 mg glucuronic acid, 40 mg urea, 165 mg glucosamine hydrochloride, 1.0 g BSA, 3.0 g mucin, 1.0 g pepsin, HCl	1.2–1.7	1:37.5	1	
	Intestinal	Duodenal: 3.5 g NaCl, 2.8 g NaH$_2$PO$_4$, 40 mg KH$_2$PO$_4$, 0.28 g KCl, 25 mg MgCl$_2$, 50 mg urea, 0.2 g CaCl$_2$, 1.0 g BSA, 3 g pancreatin, 0.5 g lipase Bile: 2.6 g NaCl, 2.9 g NaHCO$_3$, 0.18 g KCl, 0.12 g urea, 0.2 g CaCl$_2$, 1.8 g BSA, 6.0 g Bile, HCl	6.3	1:97.5	4	

different tests, it is well known that the bioaccessibility of PTE is also strongly dependent from soil characteristics, such as pH, organic matter and texture (Petruzzelli et al., 2020). For instance, previous studies have shown that the bioaccessibility of several PTE (Cu, Cd, Cr, Co, Pb, Zn, and Ni) in soil was significantly correlated with the total elemental content and soil texture (Mendoza et al., 2017; Cai et al., 2016). Other studies have attempted to identify the effect of specific soil properties on PTE bioaccessibility, revealing, not surprisingly, that pH is one of the most important parameters governing it (Li et al., 2003). Particularly, most PTE increase their bioaccessibility when the pH decreases (Du et al., 2020), which may be due to its influence on the chemical speciation and to the competition of H$^+$ ions for the same adsorption sites on the soil surfaces (Basta et al., 1993; Petruzzelli et al., 2015). Moreover, proton-promoted dissolution of Fe/Al/Mn oxyhydroxides, which could be a sink of PTE, also contribute to the increased bioaccessibility when pH decrease (Fu et al., 2010). Other important soil components influencing PTE bioaccessibility are soil organic matter (SOM), as well as Fe, Al and Mn oxides. Mostly, oxides can increase the stability of PTE in soils via inner and outer sphere complexation reactions, leading to a decrease in bioaccessibility, while SOM could also enhance the dissolution of PTE due to complexation from acid functional groups of low molecular weight SOM components (Palmer et al., 2014; Cai et al., 2017; Yin et al.,

2015).

The effect of SOM on PTE bioaccessibility has been recently investigated directly in the simulated gastrointestinal (GI) fluids from Liu et al. (2019). They depicted the Pb speciation and the role of organic complexes on Pb bioavailability. The authors stressed that increasing SOM content could increase Pb bioaccessibility due to the presence of Pb-humic complexes in the GI fluid.

2.2. Soil size fractionation

Soil texture and, more specifically, the size of particles investigated, may also have a considerable impact on bioaccessibility results.

All the extraction methods have been firstly developed using bulk soils, sieved at < 2 mm, and have been then validated using the < 250 μm soil fraction, historically chosen because of its likely representation of the soil particles that would be expected to adhere to children's hands (Ruby and Lowney, 2012).

However, the choice of a specific particle size fraction is of crucial importance for the evaluation of PTE bioaccessibility. Different soil size fractions may exhibit different properties and compositions, affecting the behavior of pollutants in soil microenvironments (Acosta et al., 2011). Previous studies have shown that fine soil fractions accumulated

Table 2
Summary of PTE bioaccessibility studies addressing different soil size fractions.

Method (in vitro)	Soil provenance	Particle size (μm)	Element (s)	Reference
IVG	8 soil samples, chromated copper arsenate contaminated area	< 250 < 90	As	Girouard and Zagury (2009)
SBRC gastric	29 soil samples: 6 railway corridor soils, 6 mining soils, 10 cattle-dip soils, and 7 geogenic soils	< 2.5 2.5–10 10–100 100–250	As	Smith et al. (2009)
UBM	72 sidewalk dust samples, urban area	0.3–5 5–50 50–150 150–250 250–1000 > 1000	Cd, Pb	Pelfrêne and Douay (2018)
UBM	21 dust samples, urban area	< 38 38–63 63–250	Cu, Fe	Patinha et al. (2015)
SBRC gastric	10 soil samples, urban area	< 2 2–10 10–22 22–50 > 50	Cr, Cu, Ni, Pb, Zn	Madrid et al. (2008)
SBRC gastric	18 soil samples and 29 road dust, peri-urban area	< 2.5 2.5–10 10–200 200–2000	Fe, Mn, Cu, Cd, Cr, Ni, Pb, Sb, Zn	Padoan et al. (2017)
SBRC gastric	33 soil samples, different mining areas	< 10 10–200	Fe, Mn, Cd, Cr, Cu, Co, Ni, Pb, Zn	Padoan et al. (2020b)
PBET	8 surface soils, highly urbanized area	< 2 2–10 10–50 50–100 100–280 280–2000	Cu, Zn, Pb, Ni, Co, Cr, Mn	Luo et al. (2011)
PBET	27 soil samples, gold mining area	< 45 < 150 < 250	As	Meunier et al. (2011)
SBRC gastric	Soil samples in urban school and kindergarten from 3 cities	< 63 63–150 150–250	Al, Fe, Mn, Cd, Cr, Cu, Ni, Pb, Zn	Ma et al. (2019)
SBRC	16 soil samples, including 2 shooting ranges sites, 3 incinerator sites, 2 urban sites, 8 mining sites and 1 gasworks site	< 50 < 100 < 250 < 2000	Pb	Juhasz et al. (2011)
UBM, PBET, SBET, SBRC, IVG	20 soil samples, urban area	< 1 1–5 5–50 50–250	As, Ba, Co, Cr, Mn, Ni, Pb, Zn	Li et al. (2020b)
SBRC gastric	4 reference soils, mining and smelting area	< 150 < 250	Pb, As	Karna et al. (2017)
UBM	2 composite soil dust samples, smelting and mining zone	< 2 2–20 20–50 50–200 200–2000	As, Cd, Cu, Pb, Sb, Sn, Zn	Goix et al. (2016)
UBM	2 soil samples, residential area	< 2 2–63 63–125 125–250	Al, As, Cd, Cr, Cu, Fe, Mn, Ni, Pb, Zn	Qin et al. (2016)
SBRC	5 street dusts and 5 soil samples, urban area	< 10 10–70 70–105 105–250	Cu, Pb, Zn	Dehghani et al. (2018)
IVG	10 soil samples, chromated copper arsenate contaminated area	< 20 20–90 90–250	As, Cu, Cr, Pb, Zn	Van der Kallen et al. (2020)
SBRC gastric	30 playground soil samples, urban area	< 50; < 150	As, Bi, Cd, Co, Cr, Cu, Fe, Mn, Ni, Pb, Sb, Sn, V, Zn	Hiller et al. (2020)
SBRC gastric	38 soil samples, gardens	< 10 < 63	As, Cd, Pb	Manjón et al. (2020)

higher PTE concentrations than coarser ones (Ajmone-Marsan et al., 2008; Luo et al., 2011). The fact has been attributed to the higher specific surface area, content of clay minerals, organic matter, and Fe, Al, and Mn oxides of the fine fraction compared to the coarser ones (Hardy and Cornu, 2006; Ajmone-Marsan et al., 2008; Luo et al., 2011; Liu et al., 2018). All these components present a variety of adsorption sites absent in coarser particles, enriched in harder minerals such as quartz, affecting thus PTE stability and bioaccessibility.

Up to now, research groups tested PTE bioaccessibility in different soil fractions, depending on the scope of the work, with or without a direct comparison between different particle sizes. Thus, the investigated particles commonly vary from clays (<2 μm particles) to particles of environmental relevance, such as the < 10 μm fraction, which could represent a possible source of atmospheric particulate matter (Thorpe and Harrison, 2008), up to the < 250 μm fraction.

In the studies where a single particle size fraction is analyzed, and in most of the studies considering a range of sizes, usually the < 250 μm size fraction is considered (Wang et al., 2018; Yin et al., 2017). This size has been used since the first investigations, and most of the methods have been developed against in vivo models using this size fraction. However, in recent years, findings demonstrated that fine fractions (e.g. < 150 μm or < 63 μm) adhere more easily to children's hands and, in addition, are more likely to be dissolved, increasing their possibilities to traverse the gastric mucosa (Ruby and Lowney, 2012; Yutong et al., 2016).

In 2016, the US Environmental Protection Agency (USEPA) reviewed recent studies on soil adherence to hands, summarizing the relationship between particle size and adherence, and endorsing the use of the < 150 μm size fraction for human health risk assessment (USEPA (United States Environmental Protection Agency), 2016). From a former study (Ikegami et al., 2014), emerged that using this fraction would allow to consider an average of 98% of particles adhering to children's hands. However, other researchers pointed out that finer particles (e.g. < 63 μm or < 100 μm) are more likely transferred into the human body, thus the use of a coarser fraction may underestimate the risk of direct soil contaminant intakes (Yamamoto et al., 2006; Siciliano et al., 2009).

3. Elemental bioaccessibility in size fractions

In most studies, different PTE exhibited different relationships between soil particle size and bioaccessibility, indicating that the influence of particle size fraction on oral bioaccessibility is metal(loid)-dependent (Ma et al., 2019; Van der Kallen et al., 2020).

The summary of the different size fractions investigated and in vitro methods used in recent studies dealing with PTE bioaccessibility is reported in Table 2.

The reviewed studies used in an equal manner the different methods for bioaccessibility estimation (Fig. 2), with a slight preference for the more simple and rapid extraction: the gastric phase SBRC (also called SBET), which is also the one endorsed from U.S. E.P.A. (USEPA (United States Environmental Protection Agency), 2013).

3.1. Arsenic

Arsenic is a widely distributed metalloid, occurring in rocks, soil, water, and air (Jarup, 2003). However, its concentration in soils may rise due to anthropogenic activities (Morin and Calas, 2006). In these cases, the exposure to As is a major public concern due to the serious risks posed to human health (Yin et al., 2015).

Previous studies have shown that soil total As concentrations increase with decreasing size fraction (Lombi et al., 2000; Smith et al., 2006), while some researchers also reported that the bioaccessible As concentrations increased with decreasing size fraction, due to a higher total As content, while the RBA (expressed as bioaccessible % of the total content of the element in the fraction) generally increased (Ruby, 2004; Girouard and Zagury, 2009). Smith et al. (2009) investigated the bioaccessibility of As in the < 2.5, 2.5–10, 10–100, and 100–250 μm soil size fractions in 50 contaminated soils. The results indicated that As was evenly distributed across fractions apart the < 2.5 μm fraction, presenting elevated As concentrations associated with a marked increase in the iron content. Arsenic RBA increased with decreasing particle size and varied depending on As origin; anthropogenic sources were more bioaccessible than geogenic ones.

Conversely, Meunier et al. (2010), working on a small number of soil samples (n = 9) reported no systematic variation of As bioaccessibility within particle size fractions (< 250 μm, < 150 μm, < 45 μm). In a successive study, Meunier et al. (2011), found the highest bioaccessible As concentrations in soil particles < 45 μm in size due to higher total As concentrations, while no systematic variation of RBA was observed between < 250 μm, < 150 μm and < 45 μm particle size fractions. Results arising from these studies may be due to the different As speciation in the different soils, as a lower As bioaccessibility was associated with encapsulated As, whereas the highest As bioaccessibility resulted, according to the authors, from the presence in soils of pentavalent As in amorphous minerals on the surface of the particles or coating the grains (Meunier et al., 2011).
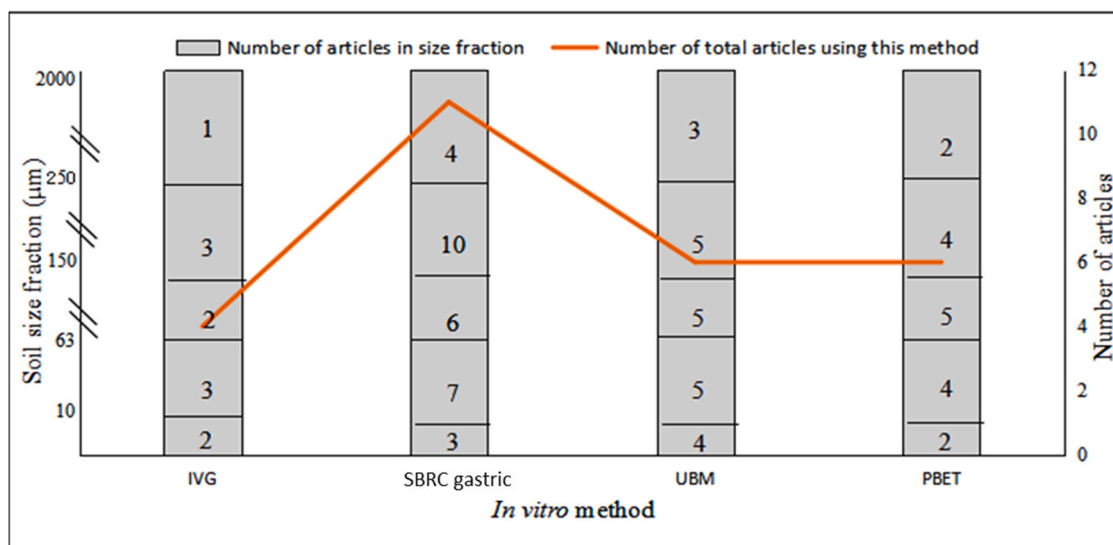

Fig. 2. Summary of studies published in the 2000–2020 period addressing PTE bioaccessibility in different soil size fractions divided according to the used method.

3.2. Lead

Lead is one of the most common environmental pollutants (Weber, 2014). Its exposure via ingestion and inhalation is a global public health hazard, especially for children (< 6 year of age), for whom Pb poisoning via hand-to-mouth exposure is of critical concern (Gao et al., 2020). Several studies looked at Pb total concentrations in soil size fractions and found that Pb concentration increased with decreasing particle size (Tawinteung et al., 2005; Weiss et al., 2006; Momani, 2006). Conversely, one long-term study, analyzing soils weathered for 70 years since the end of the contamination, noticed a decreasing concentration trend with decreasing particle size (Bright et al., 2006).

Among the studies, a lower number focused on the bioaccessibility in different particle size fractions. Juhasz et al. (2011) evaluated the influence of soil particle size fractions (Table S1) on Pb bioaccessibility and reported that, in 6 out of 16 tested soils, RBA significantly increased with decreasing particle size. Pelfrêne and Douay (2018) also found the highest RBA in the finest particles (0.3–5 μm) and a decrease when the particle size increased (5–50, 50–150, 150–250 μm), suggesting a stronger bound of Pb to coarse particles, in agreement with previous studies (Luo et al., 2011; Karna et al., 2017; Qin et al., 2016). Other studies found contrasting results; Dehghani et al. (2018) found the highest Pb bioaccessibility in 10–70 μm soil size fraction rather than the finest size fraction (< 10 μm) and Padoan et al. (2017) found an almost constant Pb RBA in all size fractions. Lead bioaccessibility vary differently among the size fractions in the different gastrointestinal phases. Yu et al. (2006), assessed the relationship among Pb bioaccessibility in three particle size fractions (< 75, 75–150, 150–250 μm). Changes in Pb RBA as a function of particle size fractions were not significant in the gastric phase while they were significant in the intestinal one. The intestinal bioaccessibility of the < 75 μm size fraction was significantly lower than 75–150 μm and 150–250 μm (same for these two size fractions), indicating that Pb was dissolved uniformly in the gastric phase but it can be quickly reabsorbed onto the surface of smaller particles after the addition of the artificial intestinal juice.

3.3. Cadmium and copper

Cadmium, one of the most toxic PTE, is an element naturally occurring in soils, with high background values in many regions. However, most of the anthropogenic emissions are due to mining and smelting, together with irrigation with sewage waters, phosphate fertilizers and other agricultural amendments, which essentially make Cd as a worldwide contaminant of soils (Ajmone-Marsan and Biasioli, 2010; Alloway, 2013).

Due to its low concentrations, Cd bioaccessibility has been mostly studied on bulk soil, while when it was assessed in different particle size fractions, its concentration decreased below the detection limit, for example in Padoan et al. (2017).

Most of the selected studies found that the RBA of Cd-containing particles was high in coarse sized particles (Goix et al., 2016; Qin et al., 2016; Ma et al., 2019), but, in some samples, Goix and Pelfrêne and Douay et al. (2016, 2018) found the highest RBA in the finest studied fractions. These differences between Cd RBA in the studies were most probably due to the different chemical speciation of Cd in the diverse soils and sampling areas (such as its adsorption onto clay minerals, oxides, its presence as a constituent of minerals or as a coprecipitate onto the surface of particles), thus to the different sources of soil contamination, as industrial sources such as batteries production, coating and plating, or agricultural ones.

Copper is also a widespread contaminant, mostly released by industrial and manufacturing activities together with traffic (Hu et al., 2020). Some researchers have found that total Cu concentration tends to be higher in finer fractions than in coarser ones (Ajmone-Marsan et al., 2008; Acosta et al., 2009; Li et al., 2014), and smaller particles are likely more ingestible than coarser ones. Recently, several researchers

determined Cu bioaccessibility in different particle sizes. Ma et al. (2019) observed a lower RBA in the fine particles (< 63 and 150 μm) than in coarser ones (150–250 and 250–2000 μm), which has been associated with high Cu affinity to organic matter, more present in fine soil fractions such as silt and clay (Kadhum et al., 2017). Differently, Dehghani et al. (2018) tested Cu bioaccessibility in different size fractions (Table 2), finding higher RBA in the finest one (< 10 μm).

3.4. Other PTE

Ma et al. (2019) found that Pb and Zn generally exhibited the highest RBA in the coarsest particle sizes (250–2000 μm); contrarily, the highest RBA of Ni occurred in the finest sizes (<63 μm). The other studied elements (Cd, Cr and Cu) did not exhibit any obvious relationships with size fraction. Li et al. (2020b) evaluated the bioaccessibility of As, Ba, Co, Cr, Cu, Mn, Ni, Pb, and Zn in different size fractions (<1, 1–5, 5–50, 50–250 μm). The results showed that elemental RBA in finer particles was higher than in larger ones and that RBA in bulk soil was mostly correlated to soil properties whereas in fine fractions was possibly associated to the diverse pollution sources and their chemical forms. Goix et al. (2016) considered soils contaminated from smelting and mining activities. In the smelting area, all elements exhibited the same pattern, with a diminution of RBA when particles diameter decreased from 200 to 2 μm and an increase in the finest fraction (2–20 μm). Arsenic, Cd, Pb, Sb and Sn showed the maximum RBA in the 50–200 μm fraction, whereas Cu and Zn were more bioaccessible in the finest fraction. In the mining area, the minimum RBA of all the metals was found for 20–50 μm particles while the highest in 2–20 μm and < 2 μm fractions. Padoan et al. (2017), examining urban soils, pointed out that the RBA of Fe, Mn, Cr, Ni and Zn increased in the fine fractions (< 2.5 and 2.5–10 μm), with significantly higher values at traffic than at background sites, consistently with the idea of a lithogenic origin of the larger particles and a more anthropic, and bioaccessible, input of fine particles for these metals. Conversely, Cu, Pb, Sb, and Zn seem to have almost constant RBA in all size fractions.

Also, Qin et al. (2016) found that size had significant effects on the RBA of landfill contaminated soils, with a clear trend showing that As, Al, Cd, Cr, Cu, Mn, Ni, Pb, and Zn RBA increased with decreasing soil particle size, and the < 2 μm soil fraction presented also a higher concentration of the elements, as compared to coarser soil fractions.

Hiller et al. (2020), observed significantly higher RBA in < 50 μm particles than in < 150 μm for six metals, namely Co, Cu, Pb, Sn, Mn and Zn. However, no differences between fractions RBA were found for As, Cr, Ni and Cd. This could have been due to multiple factors; the primary explanation of the authors was the greater contact area of fine particles available for extracting solutions. Nevertheless, some other factors, as different physicochemical, geochemical and mineralogical properties among the grain sizes, also contribute. The bioaccessibility of Cd, Cr, Cu, Pb, Sn, Zn, and Ni showed a positive correlation with the total concentration, while As, Sb and Mn bioaccessibility was negatively correlated with Fe oxides. Correlation results also indicated a negative correlation between As, Co, Cr, Mn, Pb and Ni bioaccessibility and clay and silt contents and a weak positive correlation of As, Cd, Cu, Pb and Zn with TOC.

4. Discussion and health risk assessment

Generally, PTE contaminated soils can directly harm humans via oral ingestion, inhalation and dermal contact and people exposure to polluted soils may lead to serious health problems. In most cases, health risk assessment has been based on total concentration of PTE. However, not all PTE in soils are available to absorption (Huang et al., 2018; Han et al., 2020) and the use of total contents would somewhat overestimate the risk comparing with the use of bioaccessible contents.

The presented results highlighted differences in PTE distribution among different soil particle sizes, in bioaccessible concentrations and in

RBA between the various PTE and sources of contamination.

Fine soil particles generally contained higher amounts of PTE, as previously reported, thus they contained, giving the same RBA between fine and coarser particles, higher bioaccessible amounts of elements, increasing the health risk associated to fine fraction.

Until now, we found only one article reporting the PTE bioaccessibility of nano-sized particles in soils. Dang et al. (2018) showed that the bioaccessibility of silver nanoparticles (0.5–10.9%), was significantly lower than that of $AgNO_3$ particles (4.7–14.4%), as a result of the lower adsorption of nanoparticles to soil residues during the digestion process. Conversely, many studies investigated the bioaccessibility of PTE in different nanoparticles, such as Niu et al. (2010) on nanoparticles in atmospheric particulate matter. The results emphasized a general trend of bioaccessibility increase from coarse (1000–10000 nm) to fine (100–1000 nm) and nano (57–100 nm) fractions. Bohmert et al. (2014), described that nanoparticles can reach the intestinal epithelial cells after ingestion with only a slight reduction in their cytotoxic potential. Therefore, further studies should start to also investigate soil nanoparticles and their related health risks.

Although the number or studies is still limited, PTE relative bioaccessibility seemed to be more related to the source, and to the chemico-physical form of the PTE emitted from the source, than to specific soil parameters.

For diffuse sources, such as Pb in urban soils or PTE from smelting activities and in landfill soils, clay fraction ($< 2\,\mu m$) had a higher RBA for most of the elements, due to the higher amount of adsorption sites, organic matter or oxides, accumulating PTE in a more reactive form, thus in a more bioaccessible form. In the case of point or natural contamination, the size of the particles concentrated in PTE depended strongly on the source.

Very few investigations compiled a risk assessment study using data from different particle sizes, allowing a comparison of the calculated risks. This is very important, as the choice of an incorrect size would underestimate or overestimate the risk. Li et al. (2020b) recommended to qualify the exposure risk from PTE using the fraction $< 1\,\mu m$, the most toxic one in the case of the article. With the same idea, Goix et al. (2016) recommended to sieve dust/soil samples at $< 50\,\mu m$ before analysis to limit risk underestimation. Ma et al. (2019) used $< 150\,\mu m$ and $< 250\,\mu m$ size fractions for bioaccessibility estimation, concluding that their use could result in an underestimation of the carcinogenic risk, while the use of the $< 63\,\mu m$ fraction yielded carcinogenic risks close to the results obtained including detailed calculations of the proportions of particle sizes that adhere to hands. However, the use of such a small size would alter the results in case of contamination due to the addition of coarser particles, as, for example, in the case of soils in mining areas were very coarse debris could be added to nearby soils (Mehta et al., 2020).

The choice of the optimal size of particles for the bioavailability estimation and risk assessment is thus greatly dependent on the contamination source, but the $< 150\,\mu m$ size is generally a good compromise between easiness of sample preparation, as only a dry sieving is needed, and trustworthy and conservative bioaccessibility results.

5. Conclusions

The quest for a more accurate definition of PTE bioaccessibility has led to a plethora of methods and procedures for different elements and particle sizes. In fact, there is growing evidence that there is a strong relationship between PTE bioaccessibility and soil particle size. A general trend of higher PTE bioaccessibility in finer fraction was found, which is related to total concentration, element chemical form, and anthropogenic influence. However, it does not mean that the highest PTE bioaccessibility is always found in the finest size fraction due to the considerable variation in the distribution of pollutants across soils; also, different elements may exhibit different relationships between

bioaccessibility and soil particles. Nevertheless, remediation of contaminated soils requires an assessment of the risk to human health and the environment and that should be based on more and more accurate measurement of the actual threat posed by PTE. Consensus should be sought towards a measure of bioaccessibility that is rapid and convenient.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Appendix A. Supporting information

Supplementary data associated with this article can be found in the online version at doi:10.1016/j.ecoenv.2020.111806.

References

Acosta, J.A., Cano, A.F., Arocena, J., Debela, F., Martínez-Martínez, S., 2009. Distribution of metals in soil particle size fractions and its implication to risk assessment of playgrounds in Murcia City (Spain). Geoderma 149, 101–109.

Acosta, J.A., Martínez-Martínez, S., Faz, A., Arocena, J., 2011. Accumulations of major and trace elements in particle size fractions of soils on eight different parent materials. Geoderma 161, 30–42.

Ajmone-Marsan, F., Biasioli, M., 2010. Trace elements in soils of urban areas. Water Air Soil Pollut. 213, 121–143.

Ajmone-Marsan, F., Biasioli, M., Kralj, T., Grčman, H., Davidson, C.M., Hursthouse, A.S., Madrid, L., Rodrigues, S., 2008. Metals in particle-size fractions of the soils of five European cities. Environ. Pollut. 152, 73–81.

Ajmone-Marsan, F., Padoan, E., Madrid, F., Vrščaj, B., Biasioli, M., Davidson, C., 2019. Metal release under anaerobic conditions of urban soils of four European cities. Water Air Soil Pollut. 230, 1–16.

Alloway, B.J., 2013. Sources of heavy metals and metalloids in soils. Heavy Metals in Soils. Springer, pp. 11–50.

Basta, N.T., Pantone, D., Tabatabai, M., 1993. Path analysis of heavy metal adsorption by soil. Agron. J. 85, 1054–1057.

Bohmert, L., Girod, M., Hansen, U., Maul, R., Knappe, P., Niemann, B., Weidner, S.M., Thunemann, A.F., Lampen, A., 2014. Analytically monitored digestion of silver nanoparticles and their toxicity on human intestinal cells. Nanotoxicology 8, 631–642.

Bright, D.A., Richardson, G.M., Dodd, M., 2006. Do current standards of practice in Canada measure what is relevant to human exposure at contaminated sites? I: a discussion of soil particle size and contaminant partitioning in soil. Hum. Ecol. Risk Assess. 12, 591–605.

Cai, M., McBride, M.B., Li, K., 2016. Bioaccessibility of Ba, Cu, Pb, and Zn in urban garden and orchard soils. Environ. Pollut. 208, 145–152.

Cai, M., McBride, M.B., Li, K., Li, Z., 2017. Bioaccessibility of As and Pb in orchard and urban soils amended with phosphate, Fe oxide and organic matter. Chemosphere 173, 153–159.

Cui, J.L., Zhao, Y.P., Li, J.S., Beiyuan, J.Z., Tsang, D.C., Poon, C.S., Chan, T.S., Wang, W. X., Li, X.D., 2018. Speciation, mobilization, and bioaccessibility of arsenic in geogenic soil profile from Hong Kong. Environ. Pollut. 232, 375–384.

Dang, F., Jiang, Y.Y., Li, M., Zhong, H., Peijnenburg, W., Shi, W.L., Zhou, D.M., 2018. Oral bioaccessibility of silver nanoparticles and ions in natural soils: Importance of soil properties. Environ. Pollut. 243, 364–373.

Dehghani, S., Moore, F., Vasiluk, L., Hale, B.A., 2018. The influence of physicochemical parameters on bioaccessibility-adjusted hazard quotients for copper, lead and zinc in different grain size fractions of urban street dusts and soils. Environ. Geochem. Health 40, 1155–1174.

Drexler, J.W., Brattin, W.J., 2007. An in vitro procedure for estimation of lead relative bioavailability: with validation. Hum. Ecol. Risk Assess. Int. J. 13 (2), 383–401.

Driver, J.H., Konz, J.J., Whitmyre, G.K., 1989. Soil adherence to human skin. Bull. Environ. Contam. Toxicol. 43, 814–820.

Du, H., Yin, N., Cai, X., Wang, P., Li, Y., Fu, Y., Sultana, M.S., Sun, G., Cui, Y., 2020. Lead bioaccessibility in farming and mining soils: the influence of soil properties, types and human gut microbiota. Sci. Total Environ. 708, 135227.

Edwards, R.D., Lioy, P.J., 1999. The EL sampler: a press sampler for the quantitative estimation of dermal exposure to pesticides in housedust. J. Expo. Anal. Environ. Epidemiol. 9, 521–529.

Frohne, T., Rinklebe, J., Diaz-Bone, R.A., 2014. Contamination of floodplain soils along the Wupper River, Germany, with As, Co, Cu, Ni, Sb, and Zn and the impact of pre-

definite redox variations on the mobility of these elements. Soil Sediment Contam. 23, 779–799.

Fu, H., Cwiertny, D.M., Carmichael, G.R., Scherer, M.M., Grassian, V.H., 2010. Photoreductive dissolution of Fe-containing mineral dust particles in acidic media. J. Geophys. Res. 115 (D11), D11304.

Gao, Y., Li, X., Dong, J., Cao, Y., Li, T., Mielke, H.W., 2020. Snack foods and lead ingestion risks for school aged children: a comparative evaluation of potentially toxic metals and children's exposure response of blood lead, copper and zinc levels. Chemosphere 261, 127547.

Girouard, E., Zagury, G.J., 2009. Arsenic bioaccessibility in CCA-contaminated soils: Influence of soil properties, arsenic fractionation, and particle-size fraction. Sci. Total Environ. 407, 2576–2585.

Goix, S., Uzu, G., Oliva, P., Barraza, F., Calas, A., Castet, S., Point, D., Masbou, J., Duprey, J.-L., Huayta, C., Chincheros, J., Gardon, J., 2016. Metal concentration and bioaccessibility in different particle sizes of dust and aerosols to refine metal exposure assessment. J. Hazard. Mater. 317, 552–562.

Han, Q., Wang, M., Cao, J., Gui, C., Liu, Y., He, X., He, Y., Liu, Y., 2020. Health risk assessment and bioaccessibilities of heavy metals for children in soil and dust from urban parks and schools of Jiaozuo, China. Ecotoxicol. Environ. Saf. 191, 110157.

Hardy, M., Cornu, S., 2006. Location of natural trace elements in silty soils using particle-size fractionation. Geoderma 133, 295–308.

Hiller, E., Filová, L., Jurkovič, Ľ., Mihaljevič, M., Lachká, L., Rapant, S., 2020. Trace elements in two particle size fractions of urban soils collected from playgrounds in Bratislava (Slovakia). Environ. Geochem. Health 42, 3925–3947. https://doi.org/10.1007/s10653-020-00656-6.

Huang, H., Jiang, Y., Xu, X., Cao, X., 2018. In vitro bioaccessibility and health risk assessment of heavy metals in atmospheric particulate matters from three different functional areas of Shanghai, China. Sci. Total Environ. 610–611, 546–554.

Hu, B., Shao, S., Ni, H., Fu, Z., Hu, L., Zhou, Y., Min, X., She, S., Chen, S., Huang, M., Zhou, L., Li, Y., Shi, Z., 2020. Current status, spatial features, health risks, and potential driving factors of soil heavy metal pollution in China at province level. Environ. Pollut. 266, 114961.

Ikegami, M., Yoneda, M., Tsuji, T., Bannai, O., Morisawa, S., 2014. Effect of particle size on risk assessment of direct soil ingestion and metals adhered to children's hands at playgrounds. Risk Anal. 34, 1677–1687.

Jarup, L., 2003. Hazards of heavy metal contamination. Br. Med. Bull. 68, 167–182.

Juhasz, A.L., Herde, P., Herde, C., Boland, J., Smith, E., 2015. Predicting arsenic relative bioavailability using multiple in vitro assays: validation of in vivo-in vitro correlations. Environ. Sci. Technol. 49 (18), 11167–11175.

Juhasz, A.L., Weber, J., Smith, E., Naidu, R., Rees, M., Rofe, A., Kuchel, T., Sansom, L., 2009. Assessment of four commonly employed in vitro arsenic bioaccessibility assays for predicting in vivo relative arsenic bioavailability in contaminated soils. Environ. Sci. Technol. 43, 9487–9494.

Juhasz, A.L., Weber, J., Smith, E., 2011. Impact of soil particle size and bioaccessibility on children and adult lead exposure in peri-urban contaminated soils. J. Hazard. Mater. 186, 1870–1879.

Kadhum, S.A., Ishak, M.Y., Zulkifli, S.Z., 2017. Estimation and influence of physicochemical properties and chemical fractions of surface sediment on the bioaccessibility of Cd and Hg contaminant in Langat River, Malaysia. Environ. Geochem. Health 39, 1145–1158.

Karna, R.R., Noerpel, M., Betts, A.R., Scheckel, K.G., 2017. Lead and arsenic bioaccessibility and speciation as a function of soil particle size. J. Environ. Qual. 46, 1225–1235.

Liu, Z.D., Li, H.B., Fang, X., Zhang, H., Ma, L.Q., Luo, J., 2019. Investigating lead species and bioavailability in contaminated soils: coupling DGT technique with artificial gastrointestinal extraction and in vivo bioassay. Environ. Sci. Technol. 53 (10), 5717–5724.

Liu, G., Wang, J., Liu, X., Liu, X., Li, X., Ren, Y., Wang, J., Dong, L., 2018. Partitioning and geochemical fractions of heavy metals from geogenic and anthropogenic sources in various soil particle size fractions. Geoderma 312, 104–113.

Li, Y.M., Chaney, R., Brewer, E., Roseberg, R., Angle, J.S., Baker, A., Reeves, R., Nelkin, J., 2003. Development of a technology for commercial phytoextraction of nickel: economic and technical considerations. Plant Soil 249, 107–115.

Li, X., Gao, Y., Zhang, M., Zhang, Y., Zhou, M., Peng, L., He, A., Zhang, X., Yan, X., Wang, Y., 2020b. In vitro lung and gastrointestinal bioaccessibility of potentially toxic metals in Pb-contaminated alkaline urban soil: the role of particle size fractions. Ecotoxicol. Environ. Saf. 190, 110151.

Li, J., Li, K., Cave, M., Li, H.B., Ma, L.Q., 2015. Lead bioaccessibility in 12 contaminated soils from China: correlation to lead relative bioavailability and lead in different fractions. J. Hazard. Mater. 295, 55–62.

Li, H.B., Li, M.Y., Zhao, D., Li, J., Li, S.W., Xiang, P., Juhasz, A.L., Ma, L.Q., 2020a. Arsenic, lead, and cadmium bioaccessibility in contaminated soils: measurements and validations. Crit. Rev. Environ. Sci. Technol. 50 (13), 1303–1338.

Li, Z., Ma, Z., Van der Kuijp, T.J., Yuan, Z., Huang, L., 2014. A review of soil heavy metal pollution from mines in China: pollution and health risk assessment. Sci. Total Environ. 468, 843–853.

Li, G., Sun, G.X., Ren, Y., Luo, X.S., Zhu, Y.G., 2018. Urban soil and human health: a review. Eur. J. Soil Sci. 69, 196–215.

Lombi, E., Sletten, R., Wenzel, W., 2000. Sequentially extracted arsenic from different size fractions of contaminated soils. Water Air Soil Pollut. 124, 319–332.

Luo, X.S., Yu, S., Li, X.D., 2011. Distribution, availability, and sources of trace metals in different particle size fractions of urban soils in Hong Kong: implications for assessing the risk to human health. Environ. Pollut. 159, 1317–1326.

Madrid, F., Biasioli, M., Ajmone-Marsan, F., 2008. Availability and bioaccessibility of metals in fine particles of some urban soils. Arch. Environ. Contam. Toxicol. 55, 21–32.

Manjón, I., Ramírez-Andreotta, D., Eduardo Sáez, A., Root, R., Hild, J., Janes, K., Alexander-Ozinskas, A., 2020. Ingestion and inhalation of metal(loid)s through preschool gardening: an exposure and risk assessment in legacy mining communities. Sci. Total Environ. 718, 134639.

Ma, J., Li, Y., Liu, Y., Lin, C., Cheng, H., 2019. Effects of soil particle size on metal bioaccessibility and health risk assessment. Ecotoxicol. Environ. Saf. 186, 109748.

Ma, Y., Rajkumar, M., Freitas, H., 2009. Inoculation of plant growth promoting bacterium Achromobacter xylosoxidans strain Ax10 for the improvement of copper phytoextraction by Brassica juncea. J. Environ. Manag. 90, 831–837.

Mehta, N., Cipullo, S., Cocerva, T., Coulon, F., Dino, G.A., Ajmone-Marsan, F., Padoan, E., Cox, S.F., Cave, M.R., De Luca, D.A., 2020. Incorporating oral bioaccessibility into human health risk assessment due to potentially toxic elements in extractive waste and contaminated soils from an abandoned mine site. Chemosphere 255, 126927.

Mehta, N., Cocerva, T., Cipullo, S., Padoan, E., Dino, G.A., Ajmone-Marsan, F., Cox, S.F., Coulon, F., De Luca, D.A., 2019. Linking oral bioaccessibility and solid phase distribution of potentially toxic elements in extractive waste and soil from an abandoned mine site: case study in Campello Monti, NW Italy. Sci. Total Environ. 651, 2799–2810.

Mendoza, C.J., Garrido, R.T., Quilodrán, R.C., Segovia, C.M., Parada, A.J., 2017. Evaluation of the bioaccessible gastric and intestinal fractions of heavy metals in contaminated soils by means of a simple bioaccessibility extraction test. Chemosphere 176, 81–88.

Meunier, L., Koch, I., Reimer, K.J., 2011. Effect of particle size on arsenic bioaccessibility in gold mine tailings of Nova Scotia. Sci. Total Environ. 409, 2233–2243.

Meunier, L., Walker, S.R., Wragg, J., Parsons, M.B., Koch, I., Jamieson, H.E., Reimer, K. J., 2010. Effects of soil composition and mineralogy on the bioaccessibility of arsenic from tailings and soil in gold mine districts of Nova Scotia. Environ. Sci. Technol. 44, 2667–2674.

Mokhtarzadeh, Z., Keshavarzi, B., Moore, F., Ajmone-Marsan, F., Padoan, E., 2020. Potentially toxic elements in the Middle East oldest oil refinery zone soils: source apportionment, speciation, bioaccessibility and human health risk assessment. Environ. Sci. Pollut. Res. 27, 40573–40591.

Momani, K.A., 2006. Partitioning of lead in urban street dust based on the particle size distribution and chemical environments. Soil Sediment Contam. 15, 131–146.

Morin, G., Calas, G., 2006. Arsenic in soils, mine tailings, and former industrial sites. Elements 2, 97–101.

Niu, J.J., Rasmussen, P.E., Hassan, N.M., Vincent, R., 2010. Concentration distribution and bioaccessibility of trace elements in nano and fine urban airborne particulate matter: influence of particle size. Water Air Soil Pollut. 213, 211–225.

Oomen, A.G., Hack, A., Minekus, M., Zeijdner, E., Cornelis, C., Schoeters, G., Verstraete, W., Van de Wiele, T., Wragg, J., Rompelberg, C.J.M., Sips, A., Van Wijnen, J.H., 2002. Comparison of five in vitro digestion models to study the bioaccessibility of soil contaminants. Environ. Sci. Technol. 36, 3326–3334.

Padoan, E., Passarella, I., Prati, M., Bergante, S., Facciotto, G., Ajmone-Marsan, F., 2020a. The suitability of short rotation coppice crops for phytoremediation of urban soils. Appl. Sci. 10, 307.

Padoan, E., Romè, C., Ajmone-Marsan, F., 2017. Bioaccessibility and size distribution of metals in road dust and roadside soils along a peri-urban transect. Sci. Total Environ. 601, 89–98.

Padoan, E., Romè, C., Mehta, N., Dino, G.A., De Luca, D.A., Ajmone-Marsan, F., 2020b. Bioaccessibility of metals in soils surrounding two dismissed mining sites in Northern Italy. Int. J. Environ. Sci. Technol. https://doi.org/10.1007/s13762-020-02938-z.

Palmer, S., Cox, S.F., McKinley, J.M., Ofterdinger, U., 2014. Soil-geochemical factors controlling the distribution and oral bioaccessibility of nickel, vanadium and chromium in soil. Appl. Geochem. 51, 255–267.

Patinha, C., Duraes, N., Sousa, P., Dias, A.C., Reis, A.P., Noack, Y., Ferreira da Silva, E., 2015. Assessment of the influence of traffic-related particles in urban dust using sequential selective extraction and oral bioaccessibility tests. Environ. Geochem. Health 37, 707–724.

Paustenbach, D.J., 2000. The practice of exposure assessment: a state-of-the-art review. J. Toxicol. Environ. Health Part B Crit. Rev. 3, 179–291.

Pelfrêne, A., Douay, F., 2018. Assessment of oral and lung bioaccessibility of Cd and Pb from smelter-impacted dust. Environ. Sci. Pollut. Res. 25, 3718–3730.

Pelfrêne, A., Waterlot, C., Mazzuca, M., Nisse, C., Cuny, D., Richard, A., Denys, S., Heyman, C., Roussel, H., Bidar, G., 2012. Bioaccessibility of trace elements as affected by soil parameters in smelter-contaminated agricultural soils: a statistical modeling approach. Environ. Pollut. 160, 130–138.

Petruzzelli, G., Pedron, F., Rosellini, I., Barbafieri, M., 2015. The bioavailability processes as a key to evaluate phytoremediation efficiency. Phytoremediation. Springer, pp. 31–43.

Petruzzelli, G., Pedron, F., Rosellini, I., 2020. Bioavailability and bioaccessibility in soil: a short review and a case study. AIMS Environ. Sci. 7, 208–224.

Qin, J., Nworie, O.E., Lin, C., 2016. Particle size effects on bioaccessible amounts of ingestible soil-borne toxic elements. Chemosphere 159, 442–448.

Rodriguez, R.R., Basta, N.T., Casteel, S.W., Pace, L.W., 1999. An in vitro gastrointestinal method to estimate bioavailable arsenic in contaminated soils and solid media. Environ. Sci. Technol. 33, 642–649.

Ruby, M.V., 2004. Bioavailability of soil-borne chemicals: abiotic assessment tools. Hum. Ecol. Risk Assess. 10, 647–656.

Ruby, M.V., Davis, A., Schoof, R., Eberle, S., Sellstone, C.M., 1996. Estimation of lead and arsenic bioavailability using a physiologically based extraction test. Environ. Sci. Technol. 30, 422–430.

Ruby, M.V., Lowney, Y.W., 2012. Selective soil particle adherence to hands: implications for understanding oral exposure to soil contaminants. Environ. Sci. Technol. 46, 12759–12771.

Ruby, M.V., Schoof, R., Brattin, W., Goldade, M., Post, G., Harnois, M., Mosby, D., Casteel, S., Berti, W., Carpenter, M., 1999. Advances in evaluating the oral bioavailability of inorganics in soil for use in human health risk assessment. Environ. Sci. Technol. 33, 3697–3705.

Shi, G., Chen, Z., Bi, C., Wang, L., Teng, J., Li, Y., Xu, S., 2011. A comparative study of health risk of potentially toxic metals in urban and suburban road dust in the most populated city of China. Atmos. Environ. 45, 764–771.

Siciliano, S.D., James, K., Zhang, G., Schafer, A.N., Peak, J.D., 2009. Adhesion and enrichment of metals on human hands from contaminated soil at an Arctic urban brownfield. Environ. Sci. Technol. 43, 6385–6390.

Smith, E., Smith, J., Naidu, R., 2006. Distribution and nature of arsenic along former railway corridors of South Australia. Sci. Total Environ. 363, 175–182.

Smith, E., Weber, J., Juhasz, A.L., 2009. Arsenic distribution and bioaccessibility across particle fractions in historically contaminated soils. Environ. Geochem. Health 31, 85–92.

Tawinteung, N., Parkpian, P., DeLaune, R., Jugsujinda, A., 2005. Evaluation of extraction procedures for removing lead from contaminated soil. J. Environ. Sci. Health 40, 385–407.

Thorpe, A., Harrison, R.M., 2008. Sources and properties of non-exhaust particulate matter from road traffic: a review. Sci. Total Environ. 400, 270–282.

Turner, A., 2011. Oral bioaccessibility of trace metals in household dust: a review. Environ. Geochem. Health 33, 331–341.

USEPA (United States Environmental Protection Agency), 2011. Exposure Factors Handbook 2011 Edition (Final), EPA/600/R-09/052F.

USEPA (United States Environmental Protection Agency), 2013. In vitro bioaccessibility assay for lead in soil. Washington, DC. Method 1340.

USEPA (United States Environmental Protection Agency), 2016. Recommendations for sieving soil and dust samples at lead sites for assessment of incidental ingestion, OLEM Directive 9200.1-128.

Valido, I.H., Padoan, E., Moreno, T., Querol, X., Font, O., Amato, F., 2018. Physico-chemical characterization of playground sand dust, inhalable and bioaccessible fractions. Chemosphere 190, 454–462.

Van der Kallen, C.C., Gosselin, M., Zagury, G.J., 2020. Oral and inhalation bioaccessibility of metal(loid)s in chromated copper arsenate (CCA)-contaminated soils: assessment of particle size influence. Sci. Total Environ. 734, 139412.

Wang, P., Yin, N., Cai, X., Du, H., Li, Z., Sun, G., Cui, Y., 2018. Nutritional status affects the bioaccessibility and speciation of arsenic from soils in a simulator of the human intestinal microbial ecosystem. Sci. Total Environ. 644, 815–821.

Weber, J.S., 2014. Environmental implications of phosphate-based amendments in heavy metal contaminated alluvial soil. University of Missouri, Columbia.

Weiss, A.L., Caravanos, J., Blaise, M.J., Jaeger, R.J., 2006. Distribution of lead in urban roadway grit and its association with elevated steel structures. Chemosphere 65, 1762–1771.

Wragg, J., Cave, M., Basta, N., Brandon, E., Casteel, S., Denys, S., Gron, C., Oomen, A., Reimer, K., Tack, K., 2011. An inter-laboratory trial of the unified BARGE bioaccessibility method for arsenic, cadmium and lead in soil. Sci. Total Environ. 409, 4016–4030.

Yamamoto, N., Takahashi, Y., Yoshinaga, J., Tanaka, A., Shibata, Y., 2006. Size distributions of soil particles adhered to children's hands. Arch. Environ. Contam. Toxicol. 51, 157–163.

Yin, N., Du, H., Wang, P., Cai, X., Chen, P., Sun, G., Cui, Y., 2017. Interindividual variability of soil arsenic metabolism by human gut microbiota using SHIME model. Chemosphere 184, 460–466.

Yin, N., Zhang, Z., Cai, X., Du, H., Sun, G., Cui, Y., 2015. In vitro method to assess soil arsenic metabolism by human gut microbiota: arsenic speciation and distribution. Environ. Sci. Technol. 49, 10675–10681.

Yutong, Z., Qing, X., Shenggao, L., 2016. Distribution, bioavailability, and leachability of heavy metals in soil particle size fractions of urban soils (northeastern China). Environ. Sci. Pollut. Res. 23, 14600–14607.

Yu, C.H., Yiin, L.M., Lioy, P.J., 2006. The bioaccessibility of lead (Pb) from vacuumed house dust on carpets in urban residences. Risk Anal. 26, 125–134.