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REVIEW

Oral bioaccessibility trends for As, Cd, Cr, Ni, and Pb in vegetables grown in contaminated soils: A systematic review

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Abstract

Urban and peri-urban agriculture has the potential to address social, economic, and environmental issues by bringing communities together, producing low-cost food, and greening derelict locations. However, many urban and peri-urban brownfield sites have a legacy of soil metal and metalloid contamination, and so crops grown on these sites may pose risks to human health. This review uses orally bioaccessible fractions (BAFs) of common urban and peri-urban soil contaminants observed in vegetables across literature to produce hierarchies of relative ingestion risks according to potentially toxic element (PTE), in vitro test, and vegetable type. This will inform human health risk assessment linked to consuming vegetable crops grown on PTE-contaminated land and identify areas where further research is required. BAF values were obtained from relevant literature collected from multiple online databases, for a range of PTEs, in vitro test methods, and vegetable types. Overall, median PTE BAFs were ranked as: Cd > Ni > Cr > As > Pb. Across in vitro tests, the unified BARGE method (UBM) reported the highest median vegetable BAFs when assessing all PTEs and vegetables (median 63.0% G and 59.6% GI). Median crop BAFs were ranked as follows: non-vegetables, for example, rice/fungi > legumes > root > bulb > leaf. These results will inform the site design of urban and peri-urban agroecosystems to mitigate PTE ingestion risks.

Plain Language Summary

Urban farming has many benefits, such as bringing people together, producing affordable food, and increasing green space. However, urban and surrounding soils often have high levels of contamination that could be bad for human health. This study ranked toxic elements, test methods, and vegetable types by the percentage of toxic elements released during digestion, as reported in literature. Across 25 studies, the highest percentages released during digestion were reported for cadmium, and the

Abbreviations: BAF, bioaccessible fraction; DIN, Deutsches Institut für Normung; GI, gastrointestinal phase; IVG, in vitro gastrointestinal method; PBET, physiologically based extraction test; PTE, potentially toxic element; RIVM, Dutch National Institute for Public Health and the Environment method; SBRC, solubility bioavailability research consortium; UBM, unified BARGE method.

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lowest were reported for lead. Testing undertaken using the Unified Barge Method reported the highest percentages released of all tests. Among crops, rice and fungi had the highest percentages, and leaf vegetables had the lowest percentages. These findings can help us to understand the risks posed by urban crops and support the design of urban gardens. These results also identify areas where further research is needed.

1 | INTRODUCTION

The global urban population is predicted to increase from 25% in 1950 to ~68% in 2050 (United Nations, Department of Economic and Social Affairs, Population Division, 2019). To offset the increased population density and associated food security issues, agroecosystems are being developed within the urban and peri-urban infrastructure (Environment Agency, Chief Scientist's Group, 2021) and are predicted to contribute <5% of global food production. Additionally, urban and peri-urban agroecosystem sites have significant mental health, social, and wellbeing benefits, with green social prescribing of urban agriculture becoming increasingly common since the COVID-19 pandemic (Beesley & Hardman, 2024; Clinton et al., 2018). However, the heightened anthropogenic activity associated with the built environment typically results in higher potentially toxic element (PTE) soil concentrations (Da Silva et al., 2015; McIlwaine et al., 2017; Ravenscroft et al., 2009). These PTEs persist in the urban and peri-urban environment and pose significant health risks to humans through the ingestion of soil particles and urban-grown produce, with implications for both urban and peri-urban grower health and food and economic security (Amin et al., 2020; Roy & McDonald, 2015; Wuana & Okieimen, 2011).

Risks to the health of urban and peri-urban gardeners through exposure to vegetables grown in contaminated soils are commonly assessed using existing vegetable PTE total concentration standards (Oliver et al., 2023). These thresholds include the Joint FAO/WHO Expert Committee on Food Additives (JEFCA) vegetable matter tolerable daily intake limits (JEFCA, 2022) and United States Environmental Protection Agency (USEPA) reference dose guidance (USEPA, 2002), which provide a useful indication of safe PTE contamination limits in vegetable produce. However, concentrations observed in produce may not represent whether the PTE will be absorbed within the body, as there are multiple factors that influence fate from soil to the human gut, such as solubility (Bruce et al., 2007; Fernández-Caliani et al., 2019) and plant detoxification processes (B. Kumar et al., 2017; Wei et al., 2017).

In vitro bioaccessibility testing has become useful to estimate the proportion of PTEs that are digested and available for absorption into the bloodstream. Previous articles have focused on PTE bioaccessibility in soils and the influence of

PTE speciation (Bagherifam et al., 2019; Kierulf et al., 2022; Ng et al., 2015; Yan et al., 2017) and in vitro test parameters (Kumpiene et al., 2017; Wragg & Cave, 2003; Yan et al., 2017; Zia et al., 2011) on derived bioaccessible fractions (BAFs). However, as bioaccessibility testing methodologies have advanced and recent research has expanded to investigate PTE BAFs from plant crops and animal products, there are still significant areas of bioaccessibility that have not been collectively reviewed (Babaahmadifooladi et al., 2020; Yager et al., 2015; Yu et al., 2018). Notably, there is little overarching review of bioaccessibility risks associated with vegetable crop produce grown in contaminated soils and factors that influence this bioaccessibility. Ultimately, establishing bioaccessibility of PTEs in vegetables grown in an urban and peri-urban environment, and any exacerbating methodological, PTE, or crop-linked parameters, is crucial in estimating PTE exposure risks from produce grown in urban and peri-urban soils.

This article therefore addresses the following objectives:

1. Characterizing and ranking how bioaccessibility in vegetables varies for a range of variables including:
 - a. PTE type (considering arsenic [As], cadmium [Cd], chromium [Cr], nickel [Ni], lead [Pb]). This will allow an assessment of which PTEs are most readily available in vegetables and therefore pose the greatest risk to humans
 - b. In vitro test methodology, to determine which methods are most frequently used in bioaccessibility testing, how bioaccessibility results vary by method, and therefore which methods are most significant to the study of bioaccessibility.
 - c. Gastric (G) and gastrointestinal (GI) bioaccessibility, allowing an assessment of which phase is most likely to absorb PTEs and therefore is more significant for human health.
 - d. Vegetable crop (considering leaf, root, bulb/modified stem, legume, and non-vegetable produce), to identify which vegetable types accumulate the greatest amounts of bioaccessible PTEs and therefore what are the most significant contributing factors for ingestion risk.
2. Identifying knowledge gaps observed in current literature regarding PTE bioaccessibility from vegetable crops. This review will then inform risk assessment programs that seek

to judge the human health risks associated with agroecology on contaminated land, and direct current research toward areas that warrant further investigation.

The World Health Organization (WHO) identifies arsenic (As) as one of the most prevalent and serious inorganic contaminants accessible to the human population (World Health Organization, 2022), commonly stemming from anthropogenic coal burning, pesticide residue, sulfide ore wastes, acid mine drainage, and landfill wastes (Ravenscroft et al., 2009; Reilly, 2002). Cadmium (Cd) is derived from industrial Ni-Cd battery production and the use of Cd-rich fertilizers and municipal sewage sludge on farmland (Krishnamurti et al., 2005; Reilly, 2002). Urban chromium (Cr) is typically geogenic and less common; however, anthropogenic modification of geological systems and manufacturing processes, such as chrome plating, have significantly increased its occurrence in soils (Reilly, 2002; Srivastava et al., 2021). Nickel (Ni) is often geogenic in origin, though fossil fuel burning, waste treatments, mining, and steel alloy production have increased its occurrence in urban and peri-urban areas (Ahmad & Ashraf, 2011; Genchi et al., 2020; Reilly, 2002). Finally, global processing of lead (Pb) ores, fossil fuel burning, mining, and manufacturing have released ~300 million tonnes of lead into the environment over the past five millennia (Tong et al., 2000).

The distribution and extent of PTEs in soil vary widely across the continents, as described in Table 1 (Adewumi & Ogundele, 2024; Bassi Penteadó et al., 2022; Crispo et al., 2021; B. Hu et al., 2021; Lado et al., 2008; Smith et al., 2014). Globally, Asia presents the highest average soil As, Cr, Ni, and Pb PTE concentrations, and Africa presents the highest Cd concentrations. North America, UK, and Europe soil PTE concentrations are relatively low in comparison.

Several exposure pathways should be considered when addressing human health risks from growing food in contaminated soils (Adamo et al., 2018). Direct soil ingestion is considered the most common pathway for many land use scenarios and is often significant when the land use includes growing crops. Direct soil ingestion in this scenario is particularly significant for small children and adult growers with increased soil contact to both subterranean soils and suspended airborne soil dust particles (Paustenbach, 2000). In comparison, inhalation and dermal contact represent only <1% and 0.01% of total soil intake, respectively (Paustenbach, 2000). Ingestion of contaminated crops is, however, also considered to be a significant pathway in this land use scenario. However, the risks of ingesting bioaccessible plant matter grown in contaminated soils are less well documented compared to soil PTE bioaccessibility. With the rise of urban and peri-urban agriculture worldwide, it is important to measure these risks.

Core Ideas

- Bioaccessible fractions across literature were ranked for potentially toxic element (PTE), test, and vegetable.
- Median PTE bioaccessible fractions (BAFs) were ranked as follows: Cd > Ni > Cr > As > Pb.
- The unified BARGE method reported the highest vegetable BAFs.
- Median crop BAFs were ranked as follows: non-vegetables > legumes > root > bulb > leaf.

It is often assumed that vegetable uptake potential and total concentrations are linked to soil PTE concentrations. However, other factors, such as PTE species mobility (in turn, linked to soil conditions such as pH and organic matter), also affect its incidence. As such, the relationships between soil and vegetable PTE concentrations are variable across literature, depending on local environmental parameters. For example, Gao et al. (2021) reported a positive correlation between soil and vegetable As total concentrations but negative correlations for Cr, Cd, and Hg. Ruan et al. (2023) alternately reported that Cd and Pb total soil concentrations positively correlated to vegetable concentrations, though Cr and Ni did not.

Phytoaccumulation and bioaccumulation are defined as the net enrichment, relative to the environment, of contaminants in plants and all living organisms, respectively, as the net result of all uptake and loss processes (Borgå, 2013). Plants possess an array of morphological and physiological traits that influence contaminant uptake and affect where in the plant PTEs are likely to bioaccumulate. Therefore, an understanding of the relevant mechanisms of PTE uptake and bioaccumulation is essential to elucidate trends in bioaccessibility within the edible portions of plants.

PTEs may enter the plant system through shoots or leaves after atmospheric fallout (Uzu et al., 2010), or, more commonly, through root uptake from the soil (Fahr et al., 2013). Plant PTE root uptake is dependent on many factors, including the prevalence and speciation of soil PTEs, plant characteristics, and soil conditions such as pH (Abbas et al., 2018; Escudero-Villa et al., 2024). PTEs may enter root systems through passive concentration-dependent diffusion channels (Cd and Ni) or through active transport such as phosphate/protein transporters (As and Cd), essential ion/cation carriers (Cr and Pb), or aquaporins (As) (Ahmad & Ashraf, 2011; Babula et al., 2008; Ismael et al., 2019).

Once inside the root system, PTEs may accumulate within the roots or travel through the plant vascular system and become integrated into ingestible plant tissues (Pourrut et al., 2011). PTEs typically negatively impact plant biological

TABLE 1 Distribution and extent of potentially toxic elements (PTEs) in international soils (Adewumi & Ogundele, 2024; Bassi Penteadó et al., 2022; Crispo et al., 2021; B. Hu et al., 2021; Lado et al., 2008; Smith et al., 2014).

Location	As (mg kg ⁻¹)	Cd (mg kg ⁻¹)	Cr (mg kg ⁻¹)	Ni (mg kg ⁻¹)	Pb (mg kg ⁻¹)
Europe and UK	0.3–282.0, Median 7.0	<0.01–14.1, Median 0.15	<3.0–6230.0, Median 60.0	<2.0–2690.0, Median 18.0	5.3–3943.0, Median 22.6
North America	<0.6–1110.0, Median 5.2	<0.01–46.6, Median 0.2	<1–3850, Median 31.0	<0.5–2310, Median 13.8	<0.5–2200.0, Median 17.8
South America	2.0–109.0, Median 12.5	0.2–9.8, Median 0.6	3.0–1813.0, Median 56.5	2.5–527.0, Median 31.4	6.0–240.0, Median 41.2
Asia	3.0–152.0, Average 20.1	0.1–163.9, Average 11.2	1.1–900.0, Average 107.8	1.4–2560.0, Average 95.1	2.3–5780.0, Average 283.6
Africa	1.3–24.3, Average 7.7	0.03–298.9, Average 22.7	4.7–264.8, Average 74.5	6.5–88.4, Average 32.5	4.6–2418.0, Average 228.5
Australia	8.0, Average 8.0	0.4, Average 0.4	17.0–19.0, Average 18.0	13.0–15.0, Average 14.0	102.0–194.0, Average 148.0

processes, with variable detrimental impacts on plant growth, photosynthesis, and germination (Fahr et al., 2013; B. Kumar et al., 2017; Maestri et al., 2010; Wakeel & Xu, 2020). As a result, plant defense mechanisms limit the translocation of PTEs through the plant. These defense mechanisms may include complexation (As and Pb); compartmentalization in root structures, plant vacuoles or intracellular spaces (Cd, Cr, and Pb); formation of membrane barriers (Pb); and transformation into less toxic PTE species (As); (Abbas et al., 2018; Bidar et al., 2020; Fahr et al., 2013; Gusman et al., 2013; A. Kumar et al., 2020; Pourrut et al., 2011; Shanker et al., 2005; Zhao et al., 2010). Ni is considered an essential plant micronutrient and is thus actively transported and distributed throughout the plant with limited defense mechanisms (Fabiano et al., 2015). These defense mechanisms may be exaggerated in tolerant hyperaccumulator plant species that can accumulate >1000 ppm of PTEs, such as Pteridaceae (fern; an As hyperaccumulator), Brassicaceae (mustard; Cd, Cr, Ni, and Pb), Fabaceae (legume; Ni), Lamiaceae (mint; Ni); and Ericaceae (rhododendron; Pb) (Dalla Vecchia et al., 2023; Elektorowicz & Keropian, 2015; Escudero-Villa et al., 2024; Hooda & Ma, 2010; Maestri et al., 2010; Pourrut et al., 2011; Sudmoon et al., 2015). Plant defense mechanisms that compartmentalize PTEs within root systems may then increase exposure risks associated with below-ground crops, as addressed in this review.

In the context of this study, findings are grouped by edible part to observe phytoaccumulation across the plant and assess comparable risk after ingestion:

- Leaf vegetables produce above-ground edible leafy structures (lettuce and cabbage).
- Root and tuber vegetables comprise crops harvested for the edible root structures (carrots, radishes, potatoes).
- Bulb vegetables yield edible parts derived from a modified stem or bud above the root structure (Bell, 1991; onions, leeks, garlic).

- Legumes comprise the edible “fruits” from legume/pulse plant families (peas, beans).
- Non-vegetables represent other crops produced with different edible structures (rice, mushrooms).

After ingestion of soil dust or particles, or consumption of contaminated crops, different PTE species are then differentially released from the ingested substance within the human digestive system. Small amounts of Ni and Cr (3%–6% and ~7%, respectively) are absorbed from the gut and widely distributed around the body, though these are rapidly excreted after 5 days and 30 h, respectively. Around 90% of As is absorbed from the gut, though 50% is excreted in 3–5 days. Finally, up to 5% Cd and 50% Pb are absorbed from the gut with extensive bioaccumulation in the kidneys and liver. (DesMarias & Costa, 2019; Godt et al., 2006; Kerger et al., 1996; A. Kumar et al., 2020; Patriarca et al., 1997; Ratnaik, 2003; Watanabe & Hirano, 2013). PTE concentrations, species solubility, and patient demographic and metabolic activity are all reported to influence the uptake, retention, and health impacts of PTEs (DesMarias & Costa, 2019; Wuna & Okieimen, 2011).

Acute and chronic PTE ingestion displays multiple negative health effects, from central nervous system and muscular decline to DNA damage and cancers (DesMarias & Costa, 2019; Genchi et al., 2020; Godt et al., 2006; Ratnaik, 2003; Wani et al., 2015). As a result, the European Food Safety Authority (EFSA) has set safe, tolerable daily intake fractions for oral ingestion: As 0.00006 mg kg⁻¹ body weight; Cd 0.0004 mg kg⁻¹; Cr (III) 0.3 mg kg⁻¹; Ni 13.0 mg kg⁻¹; Pb 0.0005 mg kg⁻¹ (European Food Safety Authority [EFSA], 2009, 2010, 2014a, 2014b, 2020). Other standards stated by the U.S. Agency for Toxic Substances and Disease Registry (ATSDR) also suggest minimal risk levels for oral intake for As: 0.005 mg kg⁻¹ day⁻¹; Cd: 0.0005 mg kg⁻¹ day⁻¹; Cr: 0.005 mg kg⁻¹ day⁻¹, and no limits set for Pb and Ni (Agency for Toxic Substances & Disease Registry, 2024).

The tolerable daily intake fractions set by the ESFA (listed in Section 1) do not account for how much PTEs are absorbed by the body, the different ways they are ingested, the various PTE species, or the digestive conditions. As a result, they may not accurately represent the risk of PTE contamination. Mammal in vivo testing may be used to assess the extent to which contaminants are absorbed in the gut and thus provide a more effective bioavailability fraction to evaluate vegetable ingestion risks (Paustenbach, 2000). As an alternative to in vivo methods, recent research adopts in vitro methodology that simulates the human digestive tract to give a BAF value to indicate risk from PTE ingestion (Paustenbach, 2000; Wragg & Cave, 2003).

Here, the BAF is defined as the concentration of a contaminant that is soluble under GI conditions and available for absorption into the circulatory system (Kierulf et al., 2022; Paustenbach, 2000). This is expressed as a percentage of the total contaminant concentration in the ingested plant (Kierulf et al., 2022; Paustenbach, 2000) (Equation 1).

$$\text{BAF} = (\text{PTE}_E/\text{PTE}_T) \times 100\% \quad (1)$$

where PTE_E is the PTE concentration extracted after in vitro testing (mg kg^{-1}) and PTE_T is the total PTE concentration in the ingested plant (mg kg^{-1}). The bioavailable fraction, not assessed here, is the concentration of contaminant that reaches systemic circulation and that is likely to accumulate in the body (Deshommes et al., 2012; Kierulf et al., 2022). This is expressed as a percentage of the total contaminant concentration in the ingested medium (Deshommes et al., 2012; Kierulf et al., 2022).

Current in vitro bioaccessibility tests adopt different test parameters (pH, residence time, agitation methods, soil:solution ratios, and fasted/fed state) to determine a BAF that represents the proportion of contaminant available for absorption in the gut.

Test conditions for the most commonly used bioaccessibility tests are shown in Table S1. Of these tests, the unified BARGE method (UBM) has been adopted in British and international standards as a method for investigating bioaccessibility for contaminated soils (British Standards Institution, 2018). The advantages or disadvantages of each of these tests are discussed further in Section 3.2.

2 | METHODS INVESTIGATING RANGES OF PTE BIOACCESSIBILITY IN PLANTS

In compiling this cross-study review, literature was collected from multiple online journal databases including Web of Science, Elsevier/Scopus/ScienceDirect, PubMed and JSTOR. No date restrictions were placed on the search results. Search

terms were restricted to key terminology within the topic and their derivatives, such as “oral bioaccessibility,” “vegetable,” “metal,” “metalloid,” and “potentially toxic element.” Results were assessed for PTE, in vitro assay, and vegetable group variables to determine commonly investigated variables (see Supporting Information). This confirmed As, Cd, Cr, Ni, and Pb, as well as leaf, bulb, root, legume, and non-vegetables as popular PTE and crop research topics. Articles representing the non-vegetable group were chosen at random from popular search results yielded from journal databases. Articles assessing inhaled bioaccessibility; non-in vitro-testing methods; in vivo methodology; other animal food products; and rare PTEs, such as molybdenum (Mo) and antimony (Sb), were excluded.

Bioaccessible concentrations (mg kg^{-1}) were only reported in eight studies, and so BAFs (%) were chosen as comparable values over bioaccessibility due to the availability of the data in the literature. For each of the five PTEs (As, Cd, Cr, Ni, Pb), the maximum, minimum, and median BAF were identified by vegetable (leaf, root, bulb, legume, and non-vegetables), test (physiologically based extraction test [PBET], solubility bioavailability research consortium [SBRC], Deutsches Institut für Normung [DIN], in vitro gastrointestinal method [IVG], Dutch National Institute for Public Health and the Environment method [RIVM], UBM, or unknown/bespoke assays), and phase (gastric and gastrointestinal) to facilitate comparisons. Where the species identified within a study was ambiguous, the most likely subcategory label was attributed: for example, one study reported testing “leafy vegetables,” “*Brassica*,” “root,” “bulb,” and “legume,” which were assigned to lettuce, cabbage, radish, onion, and beans, respectively, based on later work by the same authors (J. Hu et al., 2013).

Other factors that might affect plant bioaccessibility values, like soil PTE concentrations, were initially examined. However, nine of the 25 studies did not provide comprehensive data on this, so information on how soil influences vegetable BAF was limited. Most studies reported in vitro test reference material recovery values that combined all vegetables, phases, and metals studied, making it difficult to assess the accuracy of individual bioaccessible concentration (and BAF) measurements.

In total, 25 studies were sourced and evaluated. The metal contaminant, vegetable crop, in vitro test method and analysis equipment, sample preparation methods, source plant matter, total metal concentration, in vitro extractable metal concentration, and G and GI bioaccessibility values were isolated from each included study and documented. For each of the five PTEs (As, Cd, Cr, Ni, and Pb), the maximum, minimum, and average BAFs were identified by vegetable (leaf, root, bulb, legume, and non-vegetable crops), test (PBET, SBRC, DIN, IVG, RIVM, UBM, or unknown/bespoke assays), and phase (gastric and gastrointestinal) using

Source	Vegetable group	Phase	Highest					Lowest	
			Cd	Pb	Cr	As			
(M. Li et al., 2021)	Leaf	-	Cd 87%	Pb 78%	Cr 67.8%	As 11.3%			
(Zhuang, Sun, Su, et al., 2018)	Leaf, Root, Other	G GI	Cd 71% As 52%	Pb 48% Pb 30.5%	As 37% Cd 29%				
(Intawongse & Dean, 2008)	Leaf, Root	G GI	Cd <57.9% Cd 61.6%	Ni <50.68% Pb <60.7%	Pb <26.9% Ni <60.2%	Cr <24.1% Cr <43.5%			
(T. T. Xiong et al., 2016)	Leaf	-	Cd 79%	Pb 71%	Cr 64%				
(Pierart et al., 2018)	Bulb	-	Cd 85%	Pb 60%					
(Fu & Cui, 2013)	Leaf	G GI	Cd <68% Cd <93.1%	Pb <15.6% Pb <23.3%					
(Guo et al., 2022)	Leaf	G GI	Pb 56.6% Cd 18.2%	Cd 48.9% Pb 15.5%	Cr 32.0% Cr 2.65%	As 5.13%			
(Wei et al., 2017)	Leaf	G GI	Cd <87.6% As <82.5%	As <59.0% Cd <78.5%					
(J. Hu et al., 2013)	Leaf, Root, Bulb, Legume	G GI	Cd <71% Pb=Cr <26%	Ni <49% Cd <25%	Pb <42% Ni <20%	Cr <41%			
(Pan et al., 2016)	Leaf	G GI	Pb <34% Pb <12%	Ni <29% Ni <5.3%	Cd <9.4% Cd <5.3%	Cr <2.3% Cr <1.9%			
(Tian et al., 2023)	Leaf, bulb	G	Ni 61.8%	Cd 55.6%	As 40.9%	Cr 40.3%	Pb 4.9%		
(Liu et al., 2017)	Other	-	Ni <56.76%	Pb <31.47%	Cd <15.13%				
Cross-study median		G GI	Cd 66.6% Cd 43.1%	Ni 41.9% Ni 36.3%	Cr 27.0% As 36%	As 26.5% Pb 25.4%	Pb 22.5% Cr 19.8%		

FIGURE 1 Ranked median bioaccessible fractions (BAFs) of five potentially toxic elements (PTEs) in vegetables in individual studies. G, gastric; GI, gastrointestinal.

RStudio v22.07.2+576 (RStudio, 2022). Graphs were compiled in RStudio v22.07.2+576 (RStudio, 2022).

3 | RANKING VEGETABLE BIOACCESSIBILITY BY PTE, IN VITRO TEST, AND VEGETABLE

3.1 | Vegetable bioaccessibility ranked by potentially toxic element

Cd was the most frequently investigated PTE across vegetable bioaccessibility literature (18 studies), followed by As (11 studies) and Pb (11 studies), Cr (nine studies), and Ni (six studies).

Most studies reported that Cd presented the highest median BAFs in vegetables (Figure 1), irrespective of vegetable type in the G phase, though lower Cd BAFs were reported for the G phase in J. Hu et al. (2013), Pan et al. (2016), and Wei et al. (2017) and for the GI phase in Liu et al. (2017) and Zhuang, Sun, Su et al. (2018). Ni consistently presented moderate-to-high BAFs in vegetables compared to other PTEs (Figure 1).

As presented, moderate median BAFs; however, within individual studies, As exhibited lower (M. Li et al., 2021; Zhuang, Sun, Su, et al., 2018) (G) or higher (Wei et al., 2017;

Zhuang, Sun, Su, et al., 2018) (GI) BAFs than other PTEs, often depending on the test phase, although it was typically higher in the GI phase (Figure 1). Pb typically presented moderate BAFs compared to the other PTEs (Figure 1), except for studies undertaken by Pan et al. (2016) and J. Hu et al. (2013), who reported the highest fractions compared to other PTEs (<34% [G phase] and <26% [GI phase], respectively) (Figure 1). Cr consistently presented among the lowest BAFs in vegetables across all literature (Figure 1), again, except for a single study that reported the highest BAF for Cr (J. Hu et al., 2013).

Using reported BAFs from vegetables in literature to derive a median BAF for each PTE procured the following hierarchy: Cd (66.6% G and 43.1% GI) > Ni (41.9% G and 36.3% GI) > Cr (27.0% G and 19.8% GI) > As (26.5% G and 36% GI) > Pb (22.5% G and 24.4% GI).

These observations are consistent with the relative solubility, mobility, and interactions with different conditions under soils, and plant and human physiology. As and Cd are readily mobilized under changing chemical conditions (N. Li et al., 2015), and so present higher median BAFs. As an essential plant micronutrient, Ni is readily mobilized under low pH (Ahmad & Ashraf, 2011) and so presents moderate BAFs. In contrast, Cr and Pb exhibit greater compartmentalization in the root systems of vegetable crops (Bidar et al., 2020), and the active reduction and detoxification by enzymes affects its

bioavailable concentrations. This explains the lower observed median BAF for these PTEs.

In conclusion, it was found that Cd typically presented the highest BAFs in all vegetables, whilst Cr presented the lowest BAFs irrespective of vegetable type. Moderate BAFs were typically reported for Ni, As, and Pb, although the ranking of these elements varied between studies. In general, G phase BAFs were greater than GI phase BAFs.

3.2 | Sample source and preparation

Vegetable sample material was sourced from a variety of locations and experimental setups. Seven studies reported vegetable sources from local farms with and without greenhouses (Guo et al., 2022; Liu et al., 2017; Mnisi et al., 2017; Pan et al., 2016; Pizarro et al., 2016; Tian et al., 2023; Zheng et al., 2022), one from research experimental plots (X. Hu, Zhang, & Liu, 2019), one from local foraging (Koch et al., 2013), two from kitchen/urban community gardens (Hiller et al., 2022; Pelfrêne et al., 2015). In addition, six studies utilized a pot greenhouse experiment (Fu & Cui, 2013; Intawongse & Dean, 2008; Pierart et al., 2018; Wang et al., 2019; Wei et al., 2017; T. Xiong et al., 2014), and eight purchased vegetable source material from local markets (Babaahmadifooladi et al., 2021; Da Silva et al., 2015; J. Hu et al., 2013; M. Li et al., 2021; Llorente-Mirandes et al., 2016; Sun et al., 2019; Wang et al., 2021; Zhuang, Sun, Li, et al., 2018). There were no clear usage trends for vegetable source material by PTE or by vegetable group/species. Vegetable growth conditions affect plant interactions with soil and relative PTE uptake. Field conditions may present more variable results than pot or laboratory conditions due to less control over environmental parameters. Market vegetable growing conditions may be unknown.

Samples were then typically dried and tested from a raw state (using either oven-drying, freeze-drying, or, less commonly, air-drying). Seven studies tested cooked or steamed produce (Babaahmadifooladi et al., 2021; X. Hu, Zhang, & Liu, 2019; Llorente-Mirandes et al., 2016; Mnisi et al., 2017; Pelfrêne et al., 2015; Sun et al., 2019; Wang et al., 2021). Cooking or steaming is known to promote the release of PTEs from vegetable material, and so cooking prior to testing may underestimate total and bioaccessible concentrations in raw vegetables (Babaahmadifooladi et al., 2021). Raw vegetable consumption may not be customary in some cases, however, and so cooked vegetable BAFs may be more representative of typical human health risk. Five studies used fresh plant matter without drying before testing (Babaahmadifooladi et al., 2021; Da Silva et al., 2015; Fu & Cui, 2013; Koch et al., 2013; Pizarro et al., 2016). Again, there were no clear usage trends for sample preparation method by PTE or by vegetable group/species.

3.3 | Vegetable bioaccessibility ranked by in vitro test method and phase

Across literature, PBET in vitro methodology was most frequently used to investigate bioaccessibility in vegetables (15 studies), followed by standard and modified UBM (six studies), bespoke methodology and RIVM (three studies each), SBRC (two studies), and DIN and IVG (one study each).

The hierarchy produced using the median BAF for in vitro tests across all studies differed between gastric and GI phases, as follows: G: UBM (63.0%) > RIVM (40.4%) > unknown/bespoke (37.0%) > PBET (30.0%) > SBRC (28.0%) > DIN (16.0%) > IVG (12%). GI: UBM (59.6%) > SBRC (40%) > PBET (30.1%) > IVG (22.0%) > RIVM (17.0%) > DIN = unknown/bespoke (14%). UBM regularly reported some of the highest BAFs, both across all studies and where multiple tests were utilized in the same study (Pan et al., 2016; Wang et al., 2019; Zheng et al., 2022). RIVM reported relatively high BAFs in the gastric phase. The IVG and DIN reported the lowest BAFs. These observed BAFs may be the direct result of interactions between PTE species and test conditions or, alternately, may be an artifact of the limited comparable fractions in literature for IVG and DIN. In comparing BAFs across in vitro tests with differing two- or three-phase methods, it should also be noted that the higher pH conditions in saliva and GI phases in complex three-phase tests may encourage PTE precipitation and ultimately affect BAF values. This may explain some variability across the study BAFs.

The hierarchies outlined above were altered when broken down by PTE. Figure 2 describes the median BAFs in vegetables obtained using different in vitro tests across literature for each PTE. In the G phase, PBET and UBM suggest that Cd presents the highest BAFs; however, unknown/bespoke tests suggest As yields the highest BAFs (Figure 2). The general trend for high median UBM and SBRC BAFs and lower median PBET and IVG fractions across all PTEs may be attributed to the different test parameters (described in Table S1). This may include the lower pH used in the gastric phase of UBM and SBRC (Zheng et al., 2022).

PBET in vitro method was used most extensively in the studies identified in this review; however, UBM is increasing in use and appears to give a more conservative estimate of vegetable PTE bioaccessibility.

As presented significantly greater BAFs in the GI phase than G phase for six out of eight studies (Figure 1). However, studies reported that higher G phase As BAF was mostly associated with PBET, SBRC, and IVG assays (Table 2). Inconsistent As BAFs from DIN and UBM tests were greater in the G phase (Zheng et al., 2022; Zhuang, Sun, Su, et al., 2018).

In contrast, Cd and Cr presented higher BAFs in the G phase for most tests (Table 2). Where higher Cd and Cr BAFs were reported in the GI phase, these were all obtained using

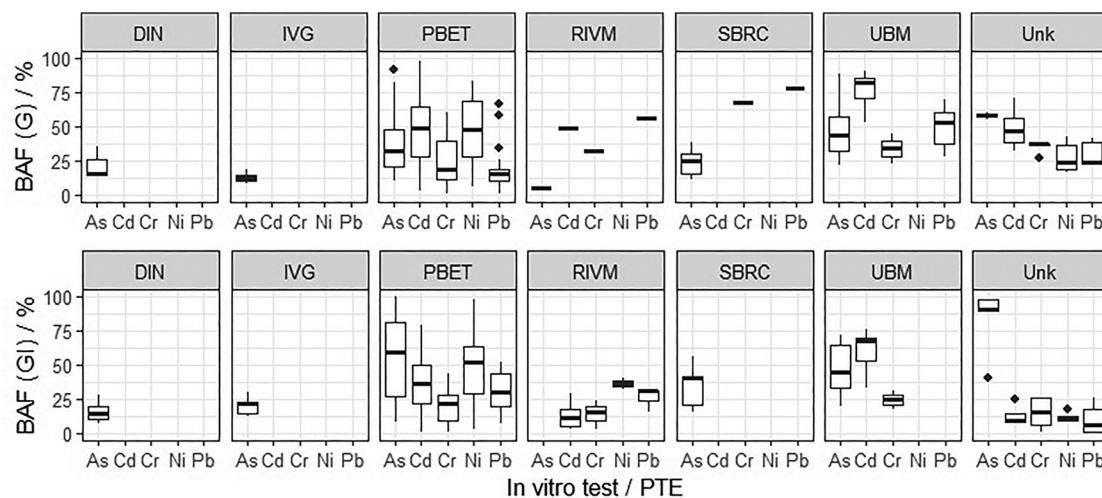


FIGURE 2 Ranking median bioaccessible fractions (BAFs) procured by different in vitro tests across elements. Twenty-five studies were used to compile the data. DIN: Deutsches Institut für Normung (Oomen et al., 2002); IVG: In vitro gastrointestinal method (Rodriguez et al., 1999); PBET: Physiologically based extraction test (Ruby et al., 1996); RIVM: Dutch National Institute for Public Health and the Environment method (Sips et al., 2001); SBRC: Solubility Bioavailability Research Consortium (Drexler & Brattin, 2007); UBM: Unified BARGE method (Wragg & Cave, 2003; Wragg et al., 2009); Unk: unknown or bespoke test. G, gastric; GI, gastrointestinal; PTE, potentially toxic element.

TABLE 2 Comparing the higher bioaccessible fractions (BAFs) reported in different gastric (G) and gastrointestinal (GI) phases in literature.

PTE	No. of studies reporting higher fraction in the gastric phase	No. of studies reporting higher fraction in the gastrointestinal phase	Median BAF G/%	Median BAF GI/%
As	2 (Zheng et al., 2022; Zhuang, Sun, Su, et al., 2018)	6 (X. Hu, Zhang, & Liu, 2019; Llorente-Mirandes et al., 2016; Pizarro et al., 2016; Wang et al., 2021; Wei et al., 2017; Zheng et al., 2022)	26.0	36.0
Cd	8 (Fu & Cui, 2013; Guo et al., 2022; Hiller et al., 2022; Intawongse & Dean, 2008; Mnisi et al., 2017; Pan et al., 2016; Pelfrène et al., 2015; Sun et al., 2019; Zhuang, Sun, Su, et al., 2018)	2 (Fu & Cui, 2013; Intawongse & Dean, 2008)	70.9	44.5
Cr	5 (Guo et al., 2022; J. Hu et al., 2013; Mnisi et al., 2017; Pan et al., 2016; Wang et al., 2019)	2 (Intawongse & Dean, 2008; Pan et al., 2016)	20.3	19.8
Ni	2 (J. Hu et al., 2013; Pan et al., 2016)	2 (Babaahmadifooladi et al., 2021; Intawongse & Dean, 2008)	31.3	35.5
Pb	4 (Guo et al., 2022; J. Hu et al., 2013; Mnisi et al., 2017; Pan et al., 2016)	2 (Fu & Cui, 2013; Intawongse & Dean, 2008)	23.5	24.4

Note: For each potentially toxic element (PTE), the phase with the greatest number of studies reporting a higher fraction is highlighted in gray.

PBET and RIVM methods (Table 2). For Ni, two studies reported higher BAFs in the gastric phase, and two studies reported higher fractions in the GI phase (Table 2). For Pb, four studies reported greater gastric BAFs, and two reported greater GI BAFs (Table 2). Again, unknown/bespoke tests and PBET obtained higher Ni and Pb BAFs in the G phase, and only PBET and RIVM reported higher fractions in the GI phase (Table 2).

Higher BAFs within a G or GI phase appear to be due to individual PTE and PTE species' solubility and the interactions with different enzymes and pH fractions between tests (Pan et al., 2016; Sun et al., 2019; Waisberg et al., 2004; Wei et al., 2017). These affect PTE release and pre-

cipitation, and thus the bioaccessible volume of contaminants measured during in vitro testing, making it difficult to predict whether the G or GI phase will be most significant for assessing risks to human health unless also comparing to in vivo bioaccessibility data.

3.4 | Vegetable bioaccessibility ranked by vegetable groups and species

BAFs observed for vegetable groups (leaf, root, bulb/modified stem, legume, and non-vegetable) and for vegetable species were compared to identify which vegetable types

TABLE 3 Range (and median) bioaccessible fractions (BAFs) across potentially toxic element (PTE) and vegetable groups from the 25 studies reviewed.

PTE	Phase	Leaf/%	Root/%	Bulb/%	Legume/%	Non-vegetable/%	Highest median BAF
As	G	5.1–63.1 (23.0)	36.4–60.1 (54.2)	19.0–64.0 (35.0)	–	19–91 (45.0)	Root
	GI	7–82.5 (22.0)	38.5–97.5 (89.7)	28.0–41.0 (36.0)	–	56.1–94.0 (71.4)	Root
Cd	G	3.2–90.0 (64.8)	24.0–89.9 (64.5)	15.0–85.3 (84.0)	47.0–83.5 (80.7)	55.5–98.0 (81.7)	Bulb
	GI	0.8–79.4 (28.7)	8.7–75.8 (54.9)	9.6–72.0 (71.6)	8.1–66.0 (55.6)	4.52–70.3 (50)	Bulb
Cr	G	1.2–67.5 (27.0)	11.9–37 (12.1)	37.0–38.0 (37.5)	36.0 (39.0)	–	Legume
	GI	1.1–43.5 (18.5)	1.4–28.3 (19.8)	26.0 (26.0)	26.0 (26.0)	15.8–23.9 (19.9)	Bulb/legume
Ni	G	6.7–82.0 (28.6)	15.5–50.7 (36.0)	24.0–75.0 (49.5)	43.0–82.6 (61.1)	–	Legume
	GI	3.2–60.2 (12.0)	8.3–50.5 (27.6)	9.4 (9.4)	18.0–97.5 (67.5)	32.2–40.7 (36.5)	Legume
Pb	G	0.5–78.1 (23.0)	8.6–61.0 (15.4)	2.5–60.0 (22.0)	24.0 (24.0)	–	Legume
	GI	0.9–49.6 (20.1)	26.0–51.8 (43.5)	0.7 (0.7)	5.7 (5.7)	31.3–31.5 (31.4)	Root
Highest median BAF		Cd	As	Cd	Cd	Cd	

Abbreviations: G, gastric; GI, gastrointestinal.

accumulate the greatest amount of bioaccessible PTEs and therefore present the most significant contributing factors for ingestion risk.

Leaf vegetables were most frequently used to evaluate PTE bioaccessibility in consumable crops (16 studies), followed by non-vegetable produce such as rice/wheat, fruits, and mushrooms (eight studies), root vegetables (eight studies), bulb vegetables (five studies), and legumes (three studies).

Table 3 summarizes the range, median, and highest BAFs reported for different PTEs and vegetables and reports the relative ranking of each vegetable group. Broadly, non-vegetable crops and legumes reported the highest BAFs, and leaf vegetables reported the lowest BAFs in both the G and GI phases (Table 3). This suggested that despite being the most frequently investigated vegetable crop for bioaccessibility testing, leaf vegetables posed the lowest risk for PTE ingestion and absorption compared to other vegetable groups.

Figure 3 shows grouped vegetable species ranked by median BAFs in the G and GI phases across different PTEs.

3.4.1 | Leaf vegetables, including Brassicas

Leaf vegetables and Brassicas were most frequently used to investigate PTE bioaccessibility in crop produce across lit-

erature. Compared to other vegetable groups, leaf vegetables presented the greatest range of BAFs in the G phase (0.5%–90%) and the lowest median BAFs in both the G and GI phases across PTEs (28.6%, 22.0%, respectively), compared to other vegetable crops. Median leaf BAFs may be ranked by PTE as follows: G: Cd (64.8%) > Ni (28.6%) > Cr (27.0%) > As (23.0%) = Pb (23.0%); GI: Cd (28.7%) > As (22.0%) > Pb (20.1%) > Cr (18.5%) > Ni (12.0%). This suggests that Cd bioaccessibility is typically highest in leaf vegetables (Pelfrène et al., 2015).

Leaf vegetables were more commonly used in bioaccessibility studies, resulting in a greater variety of leaf vegetable species to compare. Lettuce was the most frequently studied for all PTEs, followed by spinach. Generally, Brassicas like cabbage, broccoli, and cauliflower, which are more pigmented or fleshier, had higher median BAFs compared to leafy lettuce and mustard for all PTEs (Figure 3).

The hierarchy of individual leaf vegetable species median BAFs varied between PTEs and in vitro test phases (Figure 3). Brassicas (cauliflower and broccoli) and spinach reported higher BAFs for most PTEs (J. Hu et al., 2013; M. Li et al., 2021). Mustard alternately reported the lowest BAFs for all PTEs, except Pb (Pan et al., 2016). Relative hierarchical leaf vegetable species BAFs were generally similar in both the G and GI phases (Figure 3).

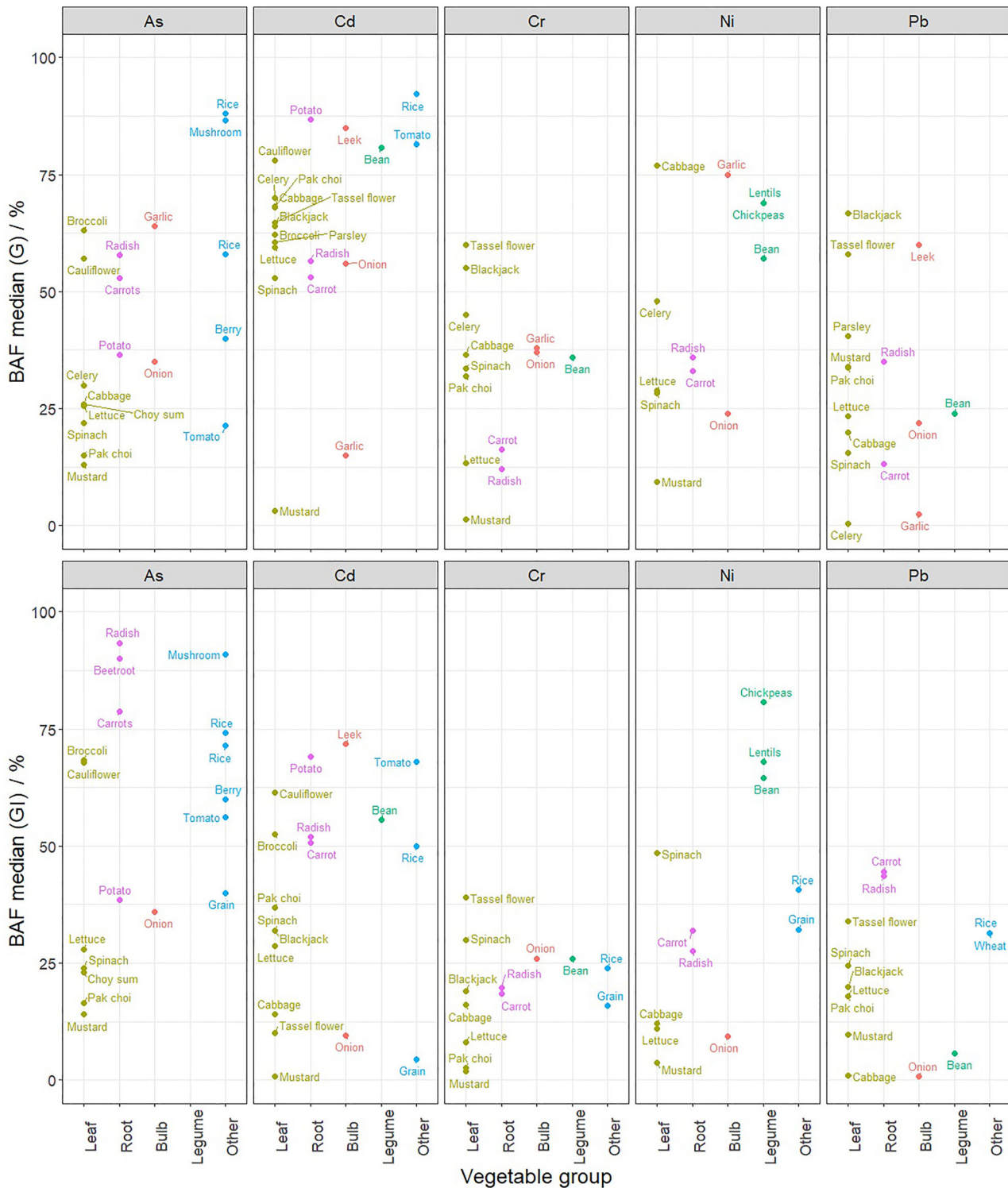


FIGURE 3 Median gastric (G) and gastrointestinal (GI) bioaccessible fractions (BAFs) for vegetable species within leaf, root, bulb, legume, and non-vegetable groups, across literature, by potentially toxic element (PTE). Vegetable groups categorized by color: leaf (yellow), root (purple), bulb (red), legume (green), and other (blue).

The generally low BAFs seen in leaf vegetables compared to other vegetable groups may be due to their unique physiology. Plants adopt defense mechanisms that sequester toxic PTEs in root structures that would not have been assessed as part of the edible leafy portion of the plant in leaf vegetables (Bidar et al., 2020; Pourrut et al., 2011). Similarly, the shorter growth period of lettuce compared to root and bulb vegetables would limit exposure to soil contaminants and result in lesser uptake and lower plant concentrations (Bidar et al., 2020). One study reported that the broad leaf surface area of leaf vegetables may encourage more extensive atmospheric deposition, which increases local PTE concentrations within the edible portions of the plant (M. Li et al., 2021). This may explain the higher BAFs for broccoli (Figure 3). There was some variation in BAFs within vegetable species; for example, Pan et al. (2016) reported low fractions for lettuce (1.2%–2.3% G and 1.1%–1.7% GI), compared to lettuce fractions presented by Intawongse and Dean (2008) (4%–9% G and 12%–25% GI). These discrepancies may be due to plant interactions with different PTE species, varying growing conditions, the level of soil contamination, vegetable preparation methods, and in vitro test parameters (Fu & Cui, 2013; Wang et al., 2019; Wei et al., 2017).

3.4.2 | Roots and tubers

BAFs from roots and tubers are less common in literature, despite the known plant defense mechanisms that preferentially concentrate PTEs in root tissues (Bidar et al., 2020; Pourrut et al., 2011). Most investigations into root vegetable bioaccessibility were focused on Cd. This may be due to the incidence of Cd in phosphate fertilizers and perceived risk in fertilizer application in urban and peri-urban agroecosystems (Roberts, 2014). In evaluating BAFs across all PTEs, roots present a high range of fractions, particularly in the GI phase (1.4%–98%). However, the median root vegetable BAFs were mid-range compared to other vegetables (38% G and 45.6% GI). Median root BAFs may be ranked by PTE as follows: G: Cd (64.5%) > As (54.2%) > Ni (36%) > Pb (15.4%) > Cr (12.1%); GI: As (89.7%) > Cd (54.9%) > Pb (43.5%) > Ni (27.6%) > Cr (19.8%). This hierarchy suggests that, similarly to leaf vegetables, Cd presents higher BAFs. In contrast to leaf vegetables, Cr presents the lowest fractions in root vegetables (e.g., Pelfrêne et al., 2015).

Carrot and radish were most frequently used to investigate bioaccessibility in root vegetables across literature. Only singular studies used potato for As and Cd (Zhuang, Sun, Su, et al., 2018) and beetroot for As (Pizarro et al., 2016). Figure 3 does not reveal any broad bioaccessibility trends for root vegetable species. By PTE, radish presented higher median BAFs than carrots for As and Cd (L. Hu, Zhang, Wu, et al., 2019; Intawongse & Dean, 2008; Zhuang, Sun, Su, et al., 2018).

Potato alternately reported the highest Cd BAFs and the lowest As BAFs compared to other root species (Pelfrêne et al., 2015; Zhuang, Sun, Su, et al., 2018). By phase, As and Cd presented consistent root species BAF hierarchies, though Cr, Ni, and Pb alternated high and low fractions for carrot and radish between G and GI phases (Figure 3).

Compared to leaf vegetables, root vegetables have greater proximity to soil contaminants, which may lead to higher accumulation rates and total PTE concentrations in their roots. This can impact the amount of PTEs that become bioavailable (Bidar et al., 2020). Additionally, some root vegetables have longer growth periods, resulting in extended exposure and greater potential for accumulating soil PTEs (Bidar et al., 2020). This does not explain the higher bioaccessibility seen in fast-growing radish (4 weeks) compared to lower BAFs reported in slow-growing carrot (12–16 weeks; RHS, 2024a, 2024b) (Figure 3). Here, some root vegetable defenses may be better adapted against soil PTE contaminants; for example, potato rind is known to act as a barrier against Cr absorption (Bidar et al., 2020). Root species' BAFs may have been influenced by the study parameters. For example, carrot (Intawongse & Dean, 2008) presented low G BAFs (10.9%–50.68%) using the PBET; Zhuang, Sun, Su, et al. (2018) reported moderate fractions (35.4%–59.6%) using the UBM; and Pelfrêne et al. (2015) reported high fractions (66.9%–85%) using the UBM. The UBM, as described above, may generate higher fractions than other tests due to the low pH and long residence times compared to other tests (Zheng et al., 2022). The limited number of comparable root vegetable studies across literature may also influence the ranked bioaccessibility hierarchies.

3.4.3 | Bulb vegetables

In comparison to root and leaf vegetables, bulb vegetables are significantly underrepresented in literature. The limited number of studies on bulb vegetables influences the observed range and median BAFs. Bulb vegetable investigations typically focused on Cd (J. Hu et al., 2013; Pelfrêne et al., 2015) or Pb (J. Hu et al., 2013; Pierart et al., 2018). Two studies addressed As (Tian et al., 2023; Zheng et al., 2022), Cr and Ni (J. Hu et al., 2013; Tian et al., 2023). Bulbs presented a similar range of bioaccessibility compared to leaf and root vegetables across all PTEs (2.5%–85.3% G and 0.7%–72% GI). They present a moderate median BAF (37.5% G and 30.0% GI) that lies between leaf vegetables (28.6% G and 22.0% GI) and root vegetables (38.0% G and 45.6% GI). Median bulb BAFs may be ranked by PTE as follows: G: Cd (84.0%) > Ni (49.5%) > Cr (37.5%) > As (35%) > Pb (22%); GI: Cd (71.6%) > As (36.0%) > Cr (26.0%) > Ni (9.4%) > Pb (0.7%). Like leaf and root vegetables, Cd presents high BAFs (J. Hu et al., 2013; Pelfrêne et al., 2015).

Onion, leek, and garlic were the only bulb vegetables studied in bioaccessibility literature, with two studies addressing onion (J. Hu et al., 2013; Zheng et al., 2022); two studies addressing leek (Pelfrêne et al., 2015; Pierart et al., 2018), and one study addressing garlic (Tian et al., 2023). Where BAFs are available for both leek and onion for any PTE species (Cd and Pb), leek presented higher median fractions than onion in both G and GI assay phases (Figure 3). Garlic presented very variable values depending on PTE, from the highest G BAFs among bulb vegetables for As, Cr, and Ni, and the lowest BAF among bulb vegetables for Pb and Cd (Tian et al., 2023).

The edible portion of bulb vegetables is derived from a modified stem (Bell, 1991). Structurally, this is placed between the roots and leaves of a plant and, as such, may serve as an intermediary contaminant reservoir between the roots and leaves, and legumes/fruits, that is then reflected in the BAFs (V. Kumar & Chopra, 2015; M. Li et al., 2021). This is the case for As (X. Hu, Zhang, & Liu, 2019; Zheng et al., 2022) and Ni (J. Hu et al., 2013; Intawongse & Dean, 2008), though not for Cd (Intawongse & Dean, 2008; Pierart et al., 2018), Cr (J. Hu et al., 2013; Intawongse & Dean, 2008), or Pb (Intawongse & Dean, 2008; Pierart et al., 2018). This suggests that other plant defense mechanisms, interaction with different PTE species, and interaction with different in vitro assay parameters may again also influence the bioaccessibility.

3.4.4 | Legumes

Like bulbs, there is little investigation into PTE bioaccessibility using legumes. Legume BAFs across PTEs were more consistent than other vegetable groups in the G phase, with a range of 24%–83.5%; however, the GI phase is less uniform. Legumes presented the highest median BAF across PTEs compared to all other vegetable types in the GI phase (58.1%), though non-vegetable produce such as rice and mushrooms presented a greater median fraction in the G phase (69.5% non-vegetable and 55.9% legume). Arsenic bioaccessibility in legumes was not explored in this pool of literature; however, the other median legume BAFs may be ranked by PTE as follows: G: Cd (80.7%) > Ni (61.1%) > Cr (26%) > Pb (24%); GI: Ni (67.5%) > Cd (55.6%) > Cr (26%) > Pb (5.7%). Unlike other vegetables, Ni was reported as relatively high in legumes (specifically chickpeas, lentils, and beans), particularly in the GI phase (Babaahmadifooladi et al., 2021), despite the process of cooking legume samples (and thus removing some PTEs) in this study prior to testing. Alternately, Pb bioaccessibility in these legumes was low (J. Hu et al., 2013).

Beans were commonly studied as a representative of legumes (J. Hu et al., 2013; Pelfrêne et al., 2015), though other produce such as chickpeas and lentils were also investigated for Ni (Babaahmadifooladi et al., 2021). Only the BAFs presented by (Babaahmadifooladi et al., 2021) may be

used to rank the different legume species by bioaccessibility as follows: G: Chickpeas = Lentils (69%) > Beans (57%); GI: Chickpeas (80.7%) > Lentils (68%) > Beans (64.6%) (Figure 3). Beans presented the lowest BAFs in both G and GI test phases (J. Hu et al., 2013). As beans are most commonly studied as representative of legumes, this may underestimate BAF risk in this vegetable group. (Babaahmadifooladi et al., 2021) also reported that Ni in lentils tended to precipitate with the pH change in the GI phase, and so bioaccessibility was marginally reduced compared to chickpeas.

Broadly, the median BAF for legumes was higher than for other vegetables, and the anticipated gradient of PTE concentration from roots to fruits was not reflected in the reported BAFs (V. Kumar & Chopra, 2015; M. Li et al., 2021). These variations may be a product of vegetable species-specific PTE transport, reduction, and compartmentalization, as concluded in other vegetable species. However, variations may also be due to legume symbiotic fungi/bacteria sequestration processes (Adeyemi et al., 2021) or a product of enhanced Ni bioaccumulation in hyperaccumulating legumes (Hooda & Ma, 2010). Finally, the limited range of studies that compare legume bioaccessibility may have skewed the relative rankings. (Babaahmadifooladi et al., 2021) singularly investigated different legume species and reported consistently high fractions for all species, despite the reported less-extreme assay conditions of the PBET (Figure 2). This increases the median BAFs from legumes and the relative risk compared to other vegetables. The greater BAFs may be due to the processed cooked/dried/canned preparation of the samples in Babaahmadifooladi et al. (2021), as cooking incites protein denaturation (protein is particularly rich in legumes). High fractions may also be due to the use of PTE processing equipment, which increases the concentration and bioaccessibility of PTEs (Pelfrêne et al., 2015).

3.4.5 | Non-vegetable produce (grain, fruit, and fungi)

In addition to vegetables, grain, rice, fruit, berry, and mushroom food products were also commonly studied for PTE bioaccessibility and health risks. These are useful to include as they are common produce for peri-urban areas and pose ingestion risks to urban growers. However, since this review focuses on vegetables, only a limited number of studies on non-vegetable produce were randomly selected for comparison. Across the selected studies, the range of fractions for non-vegetable produce is moderate compared to vegetable groups (19%–98% G and 4.5%–94% GI). The collective median of studies investigating all PTEs and all non-vegetable produce is higher than most vegetable groups, particularly for the G phase (69.7% G and 50.8% GI). Median non-vegetable BAFs may be ranked by PTE as follows: G: Cd (81.7%) > As

(45%); GI: As (71.4%) > Cd (50%) > Ni (36.5%) > Pb (31.4%) > Cr (19.9%). There were no reported fractions for Cr, Ni, and Pb in the G phase in the selected studies (Liu et al., 2017), and so comparisons may be less representative. As reported in vegetable produce, Cd and As BAFs were typically higher than Pb and Cr.

Non-vegetable produce investigated here may be split into wheat grain, rice (differentiated from wheat grain due to the large number of studies that utilize it), mushrooms, fruits, and berries. Of these, rice was most frequently studied (Liu et al., 2017; Sun et al., 2019; Wang et al., 2021; Zhuang, Sun, Su, et al., 2018). Mushroom and rice typically reported higher As, Cr, Ni and Pb BAFs than for grains, berries, and fruits for both the G and GI phases; except for a high Cd fraction for tomato in the GI phase (Figure 3), influenced by the high fractions as reported in (Pelfrène et al., 2015). Conversely, fruits and berries typically presented lower BAFs, particularly when observing As in the G phase (Koch et al., 2013; Zhuang, Sun, Su, et al., 2018).

It is difficult to draw broad conclusions to explain the variation in BAFs across fruits, grains, and fungi. This is due to the multitude of differing growing conditions, biochemical and physiological plant defense traits, soil characteristics, and local PTE species and potential solubility (Wang et al., 2019; Zheng et al., 2022). Fruits, berries, rice, and grains may be considered similar as the edible portion of the plant is typically seasonal and separate from the leaves, stem, and roots. Here, the transfer of PTEs from root to stem and to fruit during the transpiration and translocation process is longer and theoretically results in lower accumulation and potential bioavailable concentrations, compared to leaf and root vegetables (M. Li et al., 2021). However, this does not explain the high BAFs reported in legumes and rice. Fungi also possess seasonal fruiting bodies but have entirely different physiology and interactions with the soil, including the formation of symbiotic microbe-fungi relationships that may lessen any exposure to soil contaminants. These unique characteristics may inspire the higher BAFs reported in literature, rather than variation in test parameters.

4 | COMMON THEMES, RESEARCH GAPS, AND RECOMMENDATIONS

Cd was most commonly investigated (18 studies), followed by As (11 studies) and Pb (11 studies). Cr and Ni were underrepresented in the literature (nine and six studies, respectively). These preferences are likely to be due to the (1) perceived risk from increased environmental occurrence in urban areas of Cd in fertilizers and As and Pb from industrial legacy and (2) potential bioaccumulation and human health risks (Cd and Pb). Cr presented the somewhat low BAFs compared to other PTEs investigated here, and so the lesser attention

may be due to low anticipated BAFs and low consequent risk to consumers. Cr and Ni are typically geogenic and so are often overlooked in urban, peri-urban, and rural contamination studies; however, manufacturing processes have increased topsoil incidence (Crispo et al., 2021), and recent studies report some Cr and Ni hyperaccumulation in vegetable crops (Dalla Vecchia et al., 2023; Suhrhoff, 2022). As such, there may be increased human health risks linked to contaminated soil-grown vegetables, and so further investigation should consider these under-represented PTEs.

A wide range of BAFs were reported depending on the in vitro test method used. This was likely due to methodological differences such as lower pH in the gastric phase and higher residence times for the UBM and SBRC (which may account for the higher values reported for these tests). Differences in the extraction parameters and fluid compositions (outlined in Table S1) may have caused the very wide-ranging values in the GI phase for the PBET. In addition, other tests such as IVG and DIN were significantly underrepresented. As such, a broader range of in vitro tests should be investigated to fully discern the range of BAFs for each vegetable and give a more accurate representation of ingestion risk.

While UBM was often used, it was frequently modified to exclude the GI phase. The reasons for this were often unclear, though they may be due to low QA/QC CRM recoveries in the GI phase, as acknowledged by Wragg et al. (2009). Further investigation into how the GI phase omission impacts ingestion risk may be required.

For all PTEs, leaf vegetables were the most commonly studied in the literature, with fewer investigations into root vegetables, legumes, and other crops, and very few studies on bulb vegetables. Leaf vegetables are often chosen because they have short growing periods, and their above-ground shoots are easy to monitor. However, this review shows that leaf vegetables typically have the lowest BAFs, likely underrepresenting the actual ingestion risks.

Indeed, BAFs were typically higher in root, bulb, and legume vegetables compared to leaf vegetables (Figure 3). This coincides with observations that plants preferentially compartmentalize toxic Pb in root systems (Pourrut et al., 2011). However, in many cases, there were too few studies available to represent a vegetable group BAF, and so this generalization may not be representative and would benefit from further investigation. The limited studies on bulb and root vegetables may also underestimate the perceived ingestion risks.

Finally, while there are many studies on soil PTE concentrations, plant uptake, and PTE bioavailability and bioaccessibility in vegetables, few studies cover the entire system to link soil contamination with bioaccessible PTE concentrations in vegetables. As urban and peri-urban agriculture becomes more common to improve city sustainability, it is increasingly important to investigate these connections to ensure

agroecosystems can be safely implemented in contaminated soils.

4.1 | Limitations

The main limitation of this literature review is the restricted range of vegetable source material, in vitro tests utilized, and PTEs investigated. There were clear trends in literature preferentially investigating Cd in leaf vegetables using PBET in vitro methods. It is difficult to procure representative conclusions from the sparse, inconsistent data for other variables. Further research using cross-variable comparisons of PTEs, methods, and subjects would serve to confirm the general conclusions regarding bioaccessibility risk suggested in this review.

This review should also not be seen as an exhaustive list of variables assessed in literature reporting vegetable bioaccessibility risk. Other variables, though less commonly addressed, may include climate conditions, soil quality, use of additional fertilizers or soil amendments, and agroecological cropping systems.

5 | CONCLUSIONS

There are multiple factors influencing the elevated bioaccessibility risk of different PTE contaminations across vegetable and non-vegetable crops. The PTE properties, solubility, and abundance, and interactions with certain soil conditions such as pH or moisture may influence the extent to which vegetables take up the PTEs. The differing pH, residence times, enzyme use, and mixing methods between in vitro tests may similarly influence the PTE extractable concentrations and thus influence the reported BAF. Finally, the unique vegetable species' chemical and physiological processes determine the translocation and accumulation within certain plant tissues, and thus the corresponding proportion of PTEs released into the gut.

Broadly, across 25 studies exploring As, Cd Cr, Ni, Pb bioaccessibility in vegetables, Cd presented the highest BAFs, followed by Ni, Cr, As, and Pb). PBET in vitro methodology was most frequently utilized to investigate PTE bioaccessibility in vegetables; however, it often offered wide-ranging fractions compared to other assays used in literature. The UBM typically presented higher BAFs for most PTEs, particularly in the GI phase (Figure 2). Comparing BAFs reported from different bioaccessibility methods reveals that some tests may overestimate (UBM) or underestimate (IVG/DIN) perceived ingestion risks from vegetables. As a result, care should be taken when choosing methods for ingestion risk analysis for any specific PTE or crop. Leaf and root vegetables were most frequently investigated and presented the

greatest range of BAFs. However, the fewer studies investigating legumes, fruits, and bulb vegetables often reported higher BAFs and thus warrant further investigation (Table 3). These results may also be applied in urban and peri-urban agroecosystem planning to advise crop systems that present the lowest risk based on the local soil PTE contamination ranges; for example, planting leaf crops in lieu of bulbs in Cd-contaminated soils.

In conclusion, there are wide gaps in the literature exploring PTE bioaccessibility in vegetable produce, and considerable further investigation is required to improve the current state of knowledge and confirm the broad conclusions drawn in this review. In particular, this review identifies that further studies should investigate both underrepresented PTEs and in vitro test methods and should rely less on assessing quick-growing leaf vegetables (as they may underestimate risk compared to legumes and root and bulb vegetables). Further research should also consider the whole agroecological system, including total and bioaccessible soil concentrations as well as total and bioaccessible plant concentrations. Clarifying variables that enhance PTE bioaccessibility in vegetable crops will define the health risks associated with different agroecological attributes. These can then be used to develop risk-reduced nature-based solutions for contaminated sites to better urban and peri-urban social, economic, and environmental conditions.

AUTHOR CONTRIBUTIONS

Jennifer Newell: Conceptualization; data curation; methodology; writing—original draft; writing—review and editing.
Siobhan F. Cox: Supervision; writing—review and editing.
Rory Doherty: Supervision; writing—review and editing.

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CONFLICT OF INTEREST STATEMENT

The authors declare no conflicts of interest.

DATA AVAILABILITY STATEMENT

Data will be made available on request.

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SUPPORTING INFORMATION

Additional supporting information can be found online in the Supporting Information section at the end of this article.

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