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REVIEW PAPER

Review: Bioaccessibility of Potentially Harmful Metals in Dust and Soil Matrices

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Abstract

Dust is the most pervasive material affecting human health. Metal exposure to humans from dust is best assessed by bioaccessibility tests, which can be done by mimicking the conditions in the human digestive system. This review covers the works on metal bioaccessibility in dust and soil. Here, articles discussing research in this field and their bibliometric outputs have been reviewed. The Web of Science Core Collection Database was explored to collect the data, and bibliometric network analysis was performed using VOSviewer software. Research articles have broadly covered bioaccessibility and risks in urban/coking/smelting/mining plants, waste recycling, transport in vegetables, size distribution, chemical forms, daily intake, speciation, and the influence of matrix composition particle size, and source characterization. The different methods adopted for metal extraction studies have been discussed. Few of the significant findings were: the highest number of publications were observed in the year 2014; soil is the most studied matrix; China has maximum publications; metal immobilization is a vital technique to control metal leaching in the environment and manage metal exposures to humans. The most critical knowledge gaps identified are the standardization of metal extraction procedures and the formulation of realistic models for estimating metal exposure and their bioaccessibility. None of the studies have reported metal bioaccessibility from the perspective of non-invasive human bio monitors like hair or nail. More solution-oriented research would be required to curb the consequences of higher metal bioaccessibility, especially in the vulnerable classes (children/aged/ailing individuals). Further, the paper discusses different control measures, like dust wetting or metal immobilization.

Keywords Dust · Toxic metals · Exposure · Bioaccessibility · Health risks

Introduction

Around the world, many studies have evaluated metals in urban soil and dust environments. Several types of research determined the metals in the dirt, dust, and particulates in different urban environments (Valdez Cerda et al. 2011; Filippelli et al. 2012; Kumar et al. 2013; Maliki et al. 2015). Since long, most of these kind of studies were done in developed countries with substantial industrial setups. All the reported studies indicated metal characterization in the street and indoor dust to be crucial for identifying their sources and distribution. This has helped in attaining accurate health risk assessments and long-run ecological effects.

Even atmospheric pollution can be the prime source of metal pollution. Atmospheric deposition causes soil contamination from heavy metals through sedimentation, impaction, and interception. Top-layer soils and roadside dust of roadsides in urban areas are potent indicators of heavy metal contamination from atmospheric deposition. Metals like

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cadmium, zinc, copper, and lead in soil matrices are indicators of contamination of those metals which are emitted from an array of various sources such as brakes and tyres, oil lubricants, gasoline, automobile exhaust, waste incinerators, land disposal of wastes, wet or dry atmospheric deposits and industrial processes (Banerjee 2003). The mobility and availability of elements determine dust exposure's environmental and human health effects. Detailed information regarding their origin, mode of occurrence, physicochemical and biological availability, mobilization, and transport can only be determined by sequential extraction (Valdez Cerda et al. 2011).

Ingestion, inhalation, and dermal contact are the major pathways for human exposure (Acosta et al. 2014; Kumar et al. 2021). Contaminated dust can be ingested directly by children playing in grounds and grazing animals. Once ingested, they are primarily retained in the body and excreted in small proportions (Zhang et al. 2020). The accumulation of heavy metals in different tissues and organs (Zheng et al. 2010) leads to many diseases (Faiz et al. 2009). Both the frequency and time duration of intake of these metals, determine the severity of its health risks. Specially, children below 8 years, are more vulnerable to the risks caused by metal exposure, due to their increased hand-to-mouth activity. Metals have varying acute and chronic effects on exposure to humans. Pb and Hg are known to affect the nervous system, gastrointestinal system and reproductive system adversely (Mashyanov et al. 2017; Pratush et al. 2018; Vörös et al. 2018). Cd, Cu and Cr can affect the intestinal, circulatory and pulmonary systems (Izah et al. 2016; Nordberg et al. 2018), while Zn, Ni and As can damage the heart, liver and DNA (Xu et al. 2017; Sanchez et al. 2018; Stefanowicz et al. 2020).

Permissible limits and control actions are mostly limited to the total metal levels in the environment. Total metal concentrations in pollutants cannot determine their potential hazards entirely, as they do not consider their bioaccessibility and bioavailability. Hence, sequential extraction methods such as Simplified Bioaccessibility Extraction Test (SBET) and Physiologically Based Extraction Test (PBET) are adopted to assess their mobility.

Only a few forms of metallic contaminants in soil are accessible to humans through skin absorption or respiratory and gastrointestinal tracts (Pelfrêne et al. 2012). Metal bioaccessibilities are always represented in the percentage of the total metal concentrations. Bioaccessible metals cause hazards to health, rather than only risks. Metal bioaccessibility is a hazard metric that produces the risk assessment when used within a contaminated land model. Bioaccessibility tests best assess metal exposure to humans from dust and soil. It can be done by mimicking the conditions in the human digestive systems and then measuring the concentration of the metal mobilized to the aqueous phase (Turner 2011). The metal extraction procedures widely used in different studies have been discussed in detail in this review. Among all these methods, the Simplified Bioaccessibility Extraction Test (SBET) has been used widely to study themobility and bioavailability of metal contaminated soil (Basta and McGowen 2004). As SBET simulates the mobilization of any substance only in the gastric phase, it overestimates the bioaccessibility results due to the lower pH (Li et al. 2018a; Vasques et al. 2020), where metals remain in their ionic forms.

While SBET is a single extraction procedure that only evaluates the bioaccessible metal portions from the stomach, the PBET simulates gastric effects followed by the intestinal phase (Odujebe et al. 2016; Huang et al. 2018). It clearly describes the bioaccessibility in various gastrointestinal compartments representing the gastric (stomach) and intestinal parts. PBET helps in measuring chemical concentrations that are available for uptake. It assumes that soil conditions determine any metallic contaminant's solubility and availability in the human body (Pelfrêne et al. 2013). The metal availability largely depends on its binding to reactive soil surfaces. This binding is determined by processes like redox reactions, sorption and complexation (Sauvé et al. 2000; Rieuwerts et al. 2006; Rodrigues et al. 2010). These processes are controlled by the variations of soil properties, clay content, organic matter, pH and metal oxides (Römkens et al. 2009; Rodrigues et al. 2010). Typical properties like soil texture, clay mineral type, organic matter content, Mn, Fe, and Al oxides concentrations, pH, redox potential, soil saturation, and aeration determine metals' fractionation. Fractionation of metals, their mobilities, and bioavailabilities are controlled by: (a) adsorption/desorption reactions by chemical bond formations, (b) precipitation with anions like carbonates, hydroxides, sulphates, and phosphates, (c) biological mobilization and metal immobilization in soil (Seshadri et al. 2017). Food's fat-soluble vitamins and minerals become more available for metabolic functions (Santos et al. 2017) from digestive reactions in PBET. Hence it is expected that the trace element contents present in the PBET extracts will be lower than that by SBET, primarily due to the complexation of the metal ions. The solubility and bioavailability of heavy metals decrease gradually with sequential extraction steps.

This review reports literature of different analytical methods adopted for assessing bioaccessible metals in dust along with studies on different environmental realms. Additionally, this work reports an entire bibliometric study on "metals in dust" using research articles in one of the well-known scientific databases (Fig. 1). There is a robust discussion of prevailing conditions and knowledge gaps around metals in dust and the researcher's scientific impacts.

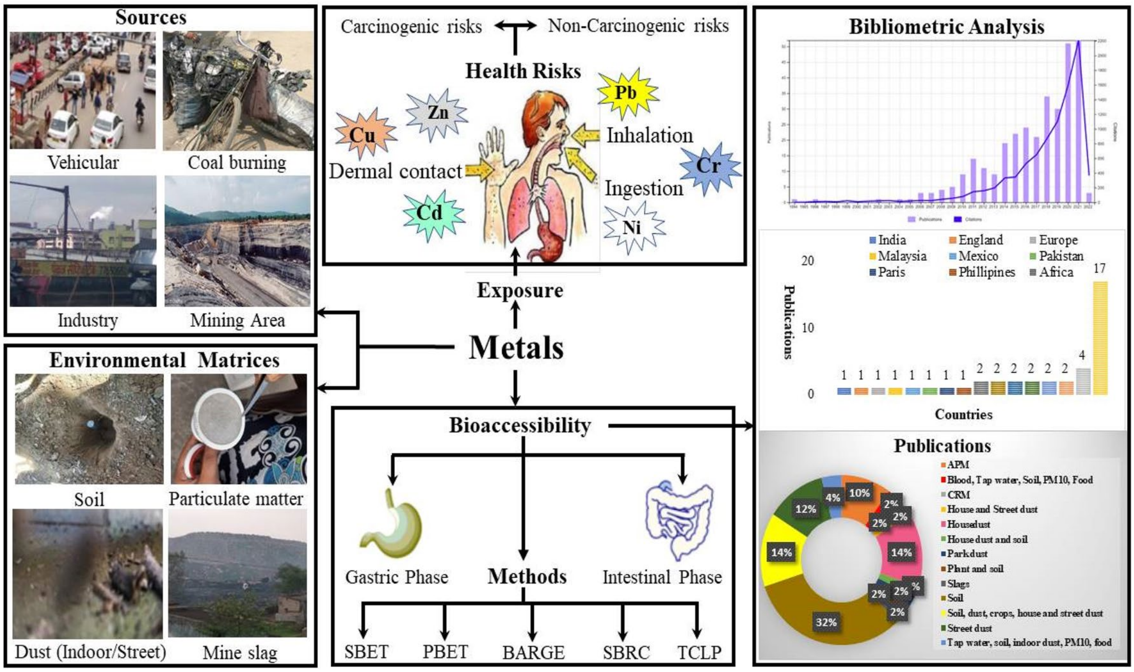

Fig. 1 Metal sources in different environmental matrices and their exposures and bio accessibilities with the bibliometric trend

An Overview of the Literature on Metal Bioaccessibility in Dust and Soil

A search was performed on Web of Science (WoS) Core Collection from Clarivate Analytics on 9th November 2022, with the keywords metal bioaccessibility in dust OR metal bioaccessibility in dust OR metal bioaccessibilities in dust OR metal bioaccessibilities in dust (similar keywords are separated by "OR" Boolean operator to obtain relevant results wherein different versions of keywords could have been used in the publications). The query link was "https://www.webofscience.com/wos/woscc/summary/107b2220-0e9e-4f35-b7e8-06d56be4a752-5c92d38c/relevance/1". The first publication was reported in 1994 and only 0–5 publications were observed till 2009 (Fig. 2a). The number of publications began increasing from 9 in 2010, 19 in 2014, 34 in 2018 to 54 in 2021. However, the number of publications has fallen to 19 in 2022 (till 9th of November). An exponential trend was observed when analyzing the citations received by these documents: 0–79 till 2010, 333 in 2014, 884 in 2018, and 2244 in 2021. Several citations (1589) have already been recorded in 2022. Although research started three decades ago (1994), an exponential rise in research was seen only after 2010 with a dramatic increase since 2014 (Fig. 2a).

Among authors, a maximum of 15 publications was authored by Pat E. Rasmussen (citations-498), followed by 14 by Albert L Juhasz (citations-442) and 10 by Andrew A. Turner (citations-483). Authors Albert L Juhasz, Pat E. Rasmussen and Lena Q. Ma showed maximum collaboration with other authors (link strengths of42, 41, and 37, respectively) (Fig. 2b). The information on top-cited authors, research areas, and journals is given in (Table S2). Among countries, China published the maximum number of documents (116 publications), followed by the U.S.A., England, Australia, and Canada with 58, 40, 33, and 33 publications, respectively (Fig. 2c). Chinese publications also received the maximum citations (4183), followed by England (1613) and the U.S.A. (1371). However, the U.S.A. showed the best collaboration network, followed by China and England (link strengths of 47, 42, and 31, respectively) (Fig. 2c). China published the bulk of the documents in this field, due to its intensive active scientific programs (like the National Medium and Long-Term Plan for the Development of Science and Technology), rigorous investment in research and development by the Chinese government (~ 2.4% of its G.D.P. was spent on research and development in 2020), and innovation-oriented transformation of China (establishment of "economic and technological development zones" and "special economic zones") (Sun and Cao 2021). Among different publishers, Elsevier published the most documents (168), followed by Springer Nature (75) and Taylor & Francis (27), while among the journals, Science of the Total Environment (54), Environmental Geochemistry and Health (32), Environmental Pollution (27), Environmental Science and Pollution Research (20), and Chemosphere (18) published the most articles (Table S1). Identifying major journals could help the academicians

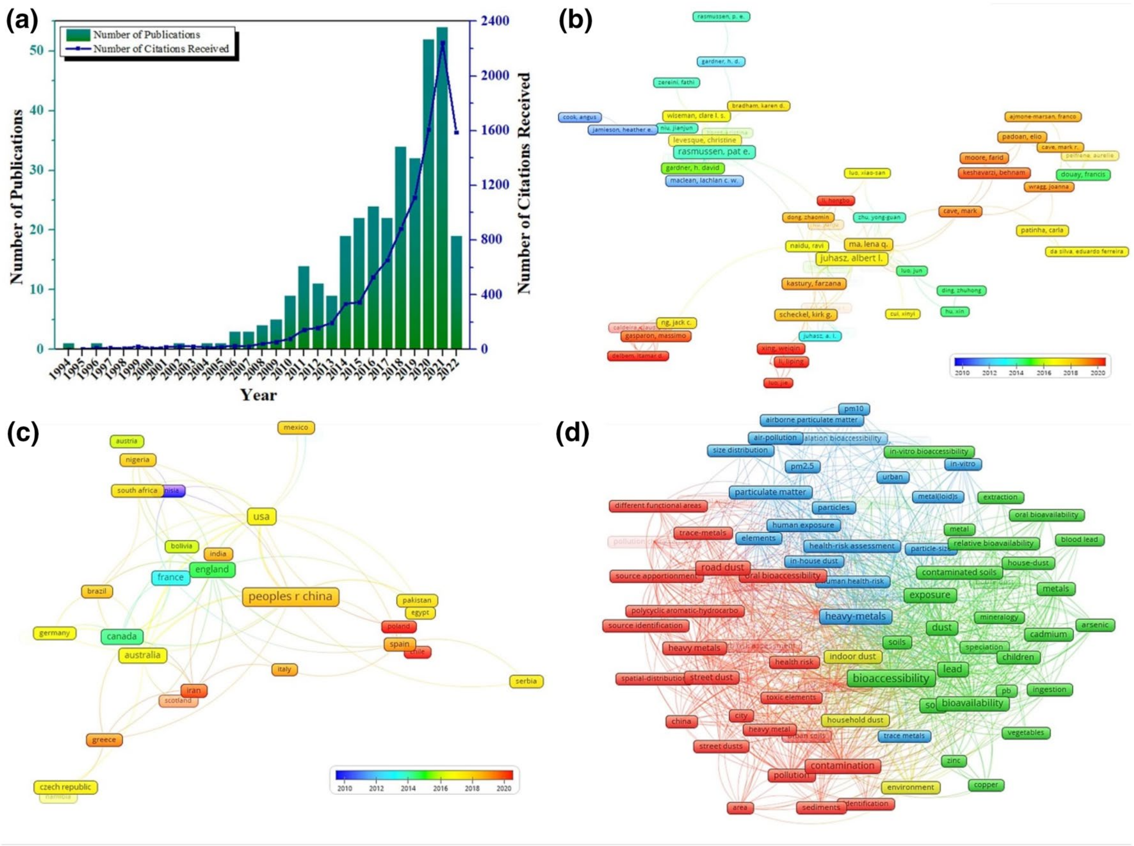

Fig. 2 **a** Evolutionary trend of publications and citations, **b** bibliometric network between authors (minimum two documents), **c** bibliometric network between countries (minimum two documents), and **d** keyword co-occurrence map (minimum ten occurrences) for metal bioaccessibility in the dust (Publications recorded till 9th of November 2022)

select impactful journals to evaluate the literature, expand research ideas, and publish documents. A different trend for citations, compared to several publications, suggests that some journals published more relevant documents (which received a more significant number of citations) in sources than others. The keyword co-occurrence map showed that bioaccessibility (221) and heavy metals (121) were the most frequently used keywords (Fig. 2d). The heavy metal lead was given the maximum attention by the research groups apart from metal(loid)s like arsenic, cadmium, copper, and zinc receiving lesser attention. Dust samples included those from roads (60), streets (73), and indoor/in house/household areas (> 100), where analyses of contamination, bioaccessibility, bioavailability, exposure assessment, and health risk assessment was focussed upon (as suggested by the keywords). Moreover, source apportionment and identification, speciation of metals, and exposure routes was attended to the research groups, where air pollution and particulate matter was identified by prime source and children were considered the most exposed among different age groups (all facts suggested by the keyword co-occurrence map). Minimal occurrence of keywords like nursery schools, playgrounds, agricultural soils, vegetables, mine areas, mine tailings, smelters, geochemistry, spatial distribution, and fractionation denote the need to perform relevant research on these relatively lesser focussed areas. Inorganic pollutants such as cobalt, chromium, and mercury and organic contaminants like polyaromatic hydrocarbons could be given larger attention by the future research groups.

The different analytical procedures followed in several literatures, to assess metal bioaccesibilities are given in Table 1. The different procedures include SBET (8), PBET (7), SBET & PBET (1), Unified BARGE Method (UBM = 7), Solubility Bioavailability Research Consortium (SBRC) in vitro assay (3), Artificial Lysosomal Fluid and Gamble's solution (3), Integrated stochastic-fuzzy pollution assessment method (ISFPAM = 1), European Community Bureau of References (BCR = 2), Stochastic Simulation of triangular fuzzy number (SS-TFN = 1) and invitro digestion model (1). Further, the major findings of metal bioaccessibilities in dust following these procedures, are given in Table 2. Among this reported literature, studies were conducted on urban street dusts (9), dusts from

Table 1 Different extraction procedures followed in the literature

Method	Standard procedure	Advantages	Disadvantages	References
SBET	1. Samples + 0.4 M glycine (pH adjusted to 1.5 ± 0.05 with HCl; solid:liquid = 1:100) 2. Mixture rotated in the end-to-end rotator for an hour at 37 °C 3. Mixture centrifuged and filtered through a 0.45 µm filter (pH = 1.5 ± 0.5) 4. Filtrate stored at 4° C for analysis in ICP-MS	Cheap Less time required Less chemicals required, hence economical	Inaccurate Estimates the gastric phase metal bioaccessibility	Ettler et al. (2012), Gu et al. (2016), Hu et al. (2011), Wang et al. (2016)
PBET	Gastric phase: 1. 0.3 g sample + gastric juice (solid: liquid = 1:100, pH = 2.5) (horizontal shaking at 37 °C, 150 rpm, 1 h) + centrifuged at 4000 rpm, 10 min + filtering using 0.45 µm size filter, and preserved at 4 °C Intestinal phase: - 2. pH of the above solution is adjusted to 7 using $NaHCO_3$ + bile + pancreatin 3. Shaken for 4 h, centrifuged at 4000 rpm, filtered at 0.45 µm filter, and kept at 4 °C until analysis	Cheaper and more accurate Differentiates distinctly between gastric and intestinal phases	No regulatory guidance exists to support this method	Carrizales et al. (2006), Rieuwerts et al. (2006)
SBRC	Gastric phase: 1. Soil + 30.03 g/L glycine (pH adjusted to 1.5 with concentrated HCl, soil: solution rati= 1:100) 2. Sample incubated at 37 °C at 40 rpm for 1 h on a suspension mixer at pH = 1.5 3. 10 ml sample was filtered through 0.45 µm filters for ICP-MS analysis Intestinal phase: - 1. 1750 mg/L of bile added to 500 mg/L pancreatin (pH maintained at 6.5 with 5 and 50% NaOH) 2. After 4 h, 10 ml of intestinal phase solution should be filtered through 0.45 µm filters for ICP-MS analysis	The method adopted by USEPA for lead and arsenic-bearing soils only	Comparisons, correlations, and standardizations for other metals, yet not achieved (bioaccessible metals in the laboratory to those bioavailable in animal studies)	Juhasz et al. (2011)

Table 1 (continued)

Method	Standard procedure	Advantages	Disadvantages	References
UBM	Gastric phase: 1. 9 mL of simulated saliva fluid is added to 0.6 g of sample 2. After 15 min, 13.5 mL of simulated gastric fluid was added and shaken in an end-to-end rotary shaker (37 ± 2 °C, 1 h, pH = 1.2–1.7) 3. Centrifuged the above mixture for 5 min at 3000 rpm 4. 9.0 mL of 0.1 M HNO_3, added to the above-derived aliquot from supernatant stored at < 8 °C until analysis ‘Gastric + intestine’ extraction 1. 9 mL of simulated saliva fluid and 13.5 mL simulated gastric fluid added to 0.6 g sample and (shaken on an end-to-end shaker at 37 ± 2 °C for 1 h, pH = 1.2 and 1.7 2. 27 mL simulated duodenal fluid and 9.0 mL of simulated bile fluid were added and shaken on an end-to-end shaker maintained at 37 ± 2° C for 4 h, pH = 6.3 ± 0.5 3. Sample suspension centrifuged for 5 min at 3000 rpm, and a 1.0 mL aliquot of the supernatant was removed, to which 9.0 mL of 0.1 M HNO_3 was added and stored at < 8 °C	Certified as per ISO17924, widely used throughout Europe	Expensive and difficult to deploy	Okorie et al. (2012); Pruvot et al. (2010)

Table 1 (continued)

Method	Standard procedure	Advantages	Disadvantages	References
Modified Toy Safety extraction	1. Modifying the European Standard Toy Safety Protocol (EN-71 1995) 2. 5 mL of 0.07 M HCl was added to 100 mg sample in an agitating water bath at 37 °C where acid volume to sample mass ratio was maintained at 50:1 3. After 1 min of agitation pH adjusted to 1.5 4. Agitation resumed for 1 h, followed by settlement of solutions in the water bath for 1 h without agitation (acid volume to sample mass ratio varied from 250:1 to 5000:1 using certified reference material) 5. 50 mL HCl added to 25 mg sample (2000:1 acid volume to sample mass ratio); and kept in a water bath at 37 °C for 2 h, followed by 1 h agitation for 1 h 6. The above solution was kept to stand for an hour and centrifuged at 3500 RPM, and diluted for ICP-MS (pH = 1.3 to 1.5)	Modification of the European Standard Toy Safety Protocol (EN-71 1995) Applies to toys/products intended for play by children below 14 years Specifies requirements for packaging, marking, and labelling	Does not cover inherent dangers like sharp needles in a sewing kit	Rasmussen et al. (2011, 2008)
TCLP	1. Solid: liquid = 1:20, at 23 ± 2 °C temperature, agitated for18 h 2. The leaching solutions derived post extraction are filtered through a 0.45 mm glass fiber filter 3. Filtrates analyzed by ICP-MS	Aggressive and conservative leaching test Mimics leachate in a landfill with higher organic content and lower pH	Does not differentiate between gastric and intestinal phases	Yu et al. (2014)

Table 2 Major findings from literature on bioaccessible metals in dust

Rank	Topic; sample; location	Reference materials	Findings	Reference
Reviews				
1	Review of metal concentrations and their bioaccessibility		Metal concentrations in house dust varied both within and between houses Metal content in house dust changed due to its source shifting from the industrial to the automobile Soil metal content cannot predict indoor dust metal concentrations	Ibanez et al. (2010); EJM
2	Review; Trace metal bioaccessibility, oral pathway, household dust		Accessibilities of Pb in the gastric phase range from 25 to 80% Effects of physicochemical variables, the solid/fluid ratio, stomach phase pH were studied systematically to derive appropriate algorithms or corrections	Turner (2011); EGAH
3	Review—approaches, and limitations of metal bioavailability and bioaccessibility through inhalation; particulate matter or dust		Comparative leaching efficiency among the varying simulating lung fluids and their in-vitro bioavailability are scarce Limitation-higher metal concentrations in environment, diminishing real exposure and risk scenarios in humans Synergistic and antagonistic effects of multiple elements rarely considered Particle size not representative of that deposited in lungs In-vitro bioaccessibility protocols for assessing the bioavailability of the metals are needed	Kastury et al. (2017); STOTEN
4	Sources of pollution, exposure to humans and associated health risks from Cd, Zn, Cu and Pb; urban roaddust; China (2009 to 2018)		Pb, Cu, Cd and Zn levels were comparatively higher in southeast provinces Main sources of metal pollution- Industrial and traffic emissions Exposure pathway trend:—ingestion > dermal > inhalation Noncarcinogenic risks posed by Pb was highest among all other metals Only in Daye city, hazard index (HI) was greater than 1 for children In general, children had higher HI values compared to that of adults	Hou et al. (2019); EI

Table 2 (continued)

Rank	Topic; sample; location	Reference materials	Findings	Reference
Simple Bioaccessible Extraction Tests (SBET)				
5	Bioaccessibility and risks to health from metals; urban street dust; Nanjing, China	Standard reference materials available at the National Research Center for Geoanalysis, PR. China (GBW07405, Soil). Laboratory blanks were routinely prepared and analyzed	Total and oral bioaccessible metal concentrations were estimated from street dust of differing land uses in Nanjing; Hg, Pb, As, Zn, Mn, and Cd have high bioaccessibility Strong correlation of SBET extractable contents with pH and organic contents Cr and As had carcinogenic risks within acceptable limits Significant noncarcinogenic risks in children but not in adults The main component of dust were silicates, carbonate, and organic matter Trend of bioaccessibile percentages: Cd > Zn > Mn > Pb > Hg > As > Cu	Hu et al. (2011); EP
6	Differences in metals/metalloids bioaccessibility from smelting and mining areas; copper belt soils; Zambia	Certified reference material (CRM) BCR-483 and standard reference material (SRM) NIST 2711 (Montana soil)	Mean As concentrations were 2–7 times higher in smelting dust Bioaccessibility of Co, Cu and Zn, Cu and Co ranged from 79–83%, 80–83% and 59–65%respectively Severe health risk is posed by topsoil ingestion in smelting areas	Ettler et al. (2012); JGE
7	Human health risk assessment; metals; Toxicity Characteristic Leaching Procedure, Simple Bioaccessibility Extraction Test; urban street dust; Tianjin, China	Not specified	Trend of potential ecological risk from metals: Cr < Cu < Pb < Cd < As Total carcinogenic risks in adults and children were $1.05*10^{-3}$ and $2.01*10^{-3}$, respectively, which were intolerable The hazard quotient followed the order: As > Cr > Pb > Cu > Cd Hazard index values in children and adults were $5.88*10^{-1}$ and $2.8*10^{-1}$	Yu et al. (2014); Plos one
8	Contamination, bioaccessibility, health risks from metals in soils of exposed lawns; urban parks; Guangzhou, China	Chinese 110 National Standard (GB/T12763.8-2007)	Concentrations of metals followed the trend: Cd < Ni < Cr < Cu < Zn < Pb < Mn < Fe Highest bioaccessibility from cadmium (75.96%) Noncarcinogenic risks were not significant in children and adults Pb, and Cr had carcinogenic risk probabilities within acceptable levels Ni and Fe had mixed sources, while other metals only had anthropogenic sources	Gu et al. (2016); AG

Table 2 (continued)

Rank	Topic; sample; location	Reference materials	Findings	Reference
9	Sources, bioaccessibility, health risks from metals; park dust in urban areas; Nanjing	Standard reference materials (GBW07405, Soil)	The oral bioaccessibility (SBET) followed the order: $Cr < As < Ni < V < C < Cu < Mn < Zn < Cd < Pb$ Most elements had an anthropogenic origin Ingestion was a significant path for noncarcinogenic risk Hazard quotients were within safe limits Children had higher risks than adults from Pb (0.154) and As (0.184) Carcinogenic risks were $< 10^{-4}$ for As and $< 10^{-6}$ (no significant health effects) for Cr, Co, Cd, and Ni	Wang et al. (2016); EES
10	Human health risks, toxic elements; ambient dusts; Sistan, Iran	soil CRM: NIST 2710—soil; CRM SRM 1570 A—spinach leaves	Health risks and bioaccessibility of metals in airborne dust in Zamboi, were measured for summers Bioaccessibilities of carcinogenic metals were Ni = 53.3%, Cr = 48.6%, As = 47.6% High non-carcinogenic risks in adults and children High carcinogenic risk through ingestion pathway Dermal route caused significant carcinogenic and non-carcinogenic risks, which were above permissible levels, except carcinogenic risks in children Inhalation route posed carcinogenic and non-carcinogenic risks, except carcinogenic risks in children, where ingestion was the major route Highest risks were posed by metal As in all exposure pathways	Dahmardeh Behrooz et al. (2021); Chemosphere

Table 2 (continued)

Rank	Topic; sample; location	Reference materials	Findings	Reference
11	Sources of bioaccessible metals in urban environments	Soil reference material NIST 2710a and NIST 2711a); indoor vacuum dust (NIST 2583)	Inhalable bioaccessible metals were higher than that by ingestion Magnetic susceptibility and bioaccessible metals for soil and dust showed strong associations Significant differences in isotopic ratios between bioaccessible and total Pb were evident Bioaccessible metals in gastric phase were mainly anthropogenic in origin (industrial to vehicular) High bioaccessible Pb, Cd and Zn concentrations along with low Pb-206/Pb-207confirmed anthropogenic influence on bioaccessible metals Coupling magnetic and bioaccessible metal concentrations with stable isotopic technique of bioaccessible Pb, is a dependable technique in determining sources of Pb with oral and inhalation bioaccessibility	Kelepertzis et al. (2021) STOTEN
12	Roadside dust, Dhaka, Bangladesh	Multi-elemental standard XSTC-662 (Spex-Certi Prep Metuchen, NJ, USA)	Bioaccessible Zn concentrations were high in gastric and intestinal phases in all land use areas Gastric phase—> 40% of As bioaccessible; intestinal phase—> 40% of Cu and Co bioaccessible Non-carcinogenic risks posed to adults and children were nil Carcinogenic risks to children and adults were within limits for Cr in spontaneous residential area (SRA) and Ni in commercial areas (for adults and children), planned residential area and urban green areas (children)	Kabir et al. (2022); TR

Table 2 (continued)

Rank	Topic; sample; location	Reference materials	Findings	Reference
Physiologically Based Extraction Tests (PBET)				
13	Pb and As exposure in children, near copper-smelter; soil; San Luis Potosi, Mexico	NIST-SRM 2710 (Montana soil)	Bioaccessibility percentages of As and Pb were 46.5% and 32.5%, respectively Children of 3–6 yrs had higher than 10 µg/dl blood lead levels Children within 8–9 yrs age had higher urinary As The soil/dust pathway contributed to 87% of total Pb in blood Exposure dose of As was above E.P.A.s reference dose	Carrizales et al. (2006);.ER
14	Metal ioaccessibility and chemistry in smelter slags; North Lake Macquarie	Sediment standards GSD-3 and PACS-1 (NRC, Canada)	Fine sized material in slags could be ingested by children Pb, As and Cd concentration of slags was high, averaging 45% Pb bioaccessibility for ~250 µm material and 75% for − 53 + 32 µm Pb in smallest slag particles are most bioaccessible, causing serious threat to health of children	Morrison and Gulson (2007); STOTEN
15	Metal bioaccessibility indoors using Physiologically Based Extraction Test (PBET); Plymouth, U.K	SRM 2711 reference material for soil	Mean bioaccessibility of metals (except Cd, Zn, and Ca) in a gastric phase was < 50% Metal concentration variations and their accessibility in indoor environments are dependent on both proportions and digestibility of metals Intestinal accessibility of metals was lower (Cd, Mn, Al, Pb, Ca, Zn, Sn, similar (Fe, Fe, Co, Cu), or greater (U, Cr), compared to the gastric phase Varied metal accessibilities in the intestine, due to chemical processes like complexation, stabilization, precipitation, reabsorption, or negatively charged polyatomic species adsorption	Turner and Ip (2007); ES&T

Table 2 (continued)

Rank	Topic; sample; location	Reference materials	Findings	Reference
16	Metal concentrations and their transport in vegetables; risk assessment to bioaccessible metals; soil; waste-incinerator site; South China	Reference material of the vegetables (GBW10015: spinach) and soils (GBW07430 (GSS16)	Vegetables bioaccumulated lower metal concentrations compared to soil Foliar uptake, a significant pathway of metal uptake- evident from concentrations of Cd, Cr in aerial vegetable parts and Settled air particles The highest metal concentrations were in leaf and bitter lettuce Cd and Pb posed the highest noncarcinogenic risks Cancer risks were unacceptable from Cd in children Non-dietary soil intake—primary route for bioaccessible metals	Li et al. (2015b); STOTEN
17	Mineralogy of soil, oral bioaccessibility, land use; risk assessment	(NIST) SRM 2709, SRM 2710, and SRM 2711	Traffic paint—high metal (Pb, Ca, Zn, Cr) concentrations, calcite, kaolinite, quartz, crocoite Pb bioaccessibility at gastric = 40–51%, intestinal = 24–70.5% Correlation of bioaccessible Pb at intestinal phase was significant with kaolinite Pb bioaccessibility differs from each soil type Soil mineralogy determines Pb release in gastrointestine	González-Grijalva et al. (2019); STOTEN
18	Health risk assessment; metal bioaccessibilities; children; soil and dust; urban parks and schools; Jaozuo	GBW07401, Geophysical Standard Reference Sample Soil	Zn, Cd concentrations in dust and Cd, As concentrations in soil were above background levels in urban schools and parks Soil Cd concentrations were almost 18 times higher than the background levels Cd concentrations in dust were 7.52 times the background levels Average and bioaccessible concentrations of Cr, Ni, Cd, Pb, Co, Zn and Mn were greater than that in soil Gastric phase had higher metal concentrations, than that in intestinal phase, except Cd and Cu As, Mn and Cd in dust and soil had bioaccessibilities greater than 10% in gastric and intestinal phases Carcinogenic and non-carcinogenic risks of metals for children was greater than that in adults	Han et al., (2020); EES

Table 2 (continued)

Rank	Topic; sample; location	Reference materials	Findings	Reference
19	Metal bioaccessibility and application of magnetic susceptibility; urban sandstorm contamination; health risks, Dunhuang and Lanzhou, China	Not available	Mean magnetic susceptibility of sandstorms in Lanzhou was 5 times higher than that in Dunhuang Metal concentrations in sandstorms were higher than topsoil background values Zn, Cd, Pb and Cu originated from natural and anthropogenic sources Metal bioaccessibilities were higher in sandstorms of Lanzhou:—gastric phase (22.69% to 50.86% for Cu and Pb respectively) and intestinal phase (12.07% to 22.11% for Pb and Cd respectively) Significant correlation of heavy metal concentrations with the magnetic minerals Average ecological risk posed by heavy metals in sandstorms were low, while at individual sites, it ranged from moderate to high Highest non-carcinogenic and carcinogenic risks were through ingestion route, for adult and children	Ma et al. (2022); STOTEN
SBET and PBET				
20	Brazil's soils, slag, mine wastes, and abandoned mines	Certified soil reference material SRM 2710	Solubility during digestion determines the bioavailability of Pb Pb bioaccessibility in soil = 0.03–4.1%, Pb bioaccessibility in solid waste = 1.2–15% Pb will partially dissolve in the stomach, and the rest will be soluble in the duodenum	Bosso and Enzweiler (2008); EGAH
Unified BARGE Method (UBM)				
21	Pb, Zn, Cd bioaccessibility from contaminated soils (past emissions from smelters); Paris	Reference soil, NIST SRM 2710	Gastric phase bioaccessibility of Cd = 68%, Pb = 62%, Zn = 47% Intestinal bioaccessibility of Cd = 23%, Pb = 32%, and Zn = 31% respectively Positive correlations between metal concentrations and Unified Bioaccessibility Method extracted metals Human bioaccessibility is affected by physicochemical parameters of soil	Pruvot et al. (2010); AECT

Table 2 (continued)

Rank	Topic; sample; location	Reference materials	Findings	Reference
22	Daily intake of toxic elements, the role of oral bioaccessibility testing, urban street dust; England	Road dust (BCR 723), soil (GBW 07401), bush branches and leaves (NCS DC 73349), and Montana soil (SRM 2711)	Pb concentration was exceptionally high in major road artery and pedestrians due to Pb-flashing The oral intakes of Cu, Ni, and Cd were within tolerable daily intakes Cd and Zn had median bioaccessibility values $>45\%$, and Cu, Ni, Pb, and As $<35\%$ The median bioaccessible fractions using the Unified Bioaccessibility Method was $>45\%$ for Cd and Zn Based on dustiness, the total daily intake of Pb was exceeded	Okorie et al. (2012); Chemosphere
23	Soil particle size; health risk assessment and metal bioaccessibility	Reference materials, GSS21 and GSS25 (Institute of Geophysical and Geochemical Exploration of the Chinese Academy of Geological Sciences)	Highest Zn and Pb bioaccessibility in the coarsest size fraction (250-2000mum) Highest Ni bioaccessibility in the finest fraction (<63mum) No effect of particle size on determination of non-carcinogenic risks <63 µm is the optical size fraction for determination of carcinogenic risks from oral ingestion	Ma et al. (2019); EES
24	Oral bioaccessibility; human health risk assessment, potentially toxic element; abandoned mine site	ISO standard method (ISO 17924:2018)	Bioaccessible fraction of Cd = 72–98%, Cr = 3–11%, Pb = 16–88%, Zn = 73–94%, As = 5–33%, C = 24–42%, Cu = 25–90%, Ni = 17–60% Variation of bioaccessible fraction were due to alkaline calcareous rocks, and association of potentially toxic elements and variety of minerals Hazard index (HI) of oral bioaccessible fractions for residential areas (0.02 to 17.9), were above acceptable levels for 50% of the samples	Mehta et al. (2020); Chemosphere
25	Phase partitioning and in-vitro bioaccessibility; health risk; industrial zones and iron mining	NIST2711a (Montana Soil II) and BGS-102	Fe, Cu and As were highly enriched in $PM_{2.5}$, PM_{10}, and total suspended particulate matter Metal bioaccessibility through oral pathway = 0.35% to 41.55% (gastric phase), and 0.06–37.58% (intestinal phase) Regression modelling revealed metal bioaccessibilities to be determined by total metal concentrations in dust Average metal intake, hazard quotients and carcinogenic risks—within tolerable limits	Soltani et al. (2021); EES

Table 2 (continued)

Rank	Topic; sample; location	Reference materials	Findings	Reference
26	Oral and inhalation bioaccessibility, Hg mining district, Idrija, Slovenia	NIST 2584	< 3% Hg—bioaccessible in synthetic solutions Pb, Cd and Zn—the highest bioaccessible metals in artificial lysosomal fluid (ALF) solution Pb, Zn, Cd—lower bioaccessibility in gastric phase Except Cr, metal bioaccessibility was lowest in gastric phase Daily ingestion and inhalation doses of metals from indoor dusts were below tolerable daily intake, prescribed by European Food Safety Authority	Zupančič et al. (2021); EGAH
27	In vitro assessments of bioavailability and bioaccessibilirty of metals in $PM_{2.5}$ metals; digestive and respiratory systems	NIST SRM 1648a, urban PM	Significant health risks are posed from trace metals of $PM_{2.5}$ via ingestion and inhalation route Both elements and body fluid determine the bioavailability and bioaccessibility of elements Metal in $PM_{2.5}$ depicted variations in bioaccessibility of various human organs Bioaccessible/bioavailable metals like Ni, Co, Fe from metallurgic dust and traffic emissions contributed to $PM_{2.5}$ induced oxidative potential	Zhao et al. (2021); HAZMAT
European Toy Safety Protocol (EN-71 1995)				
28	Effect of matrix composition on metal bioaccessibility; urban residential dust; and soil; Ottawa, Canada	NIST 2583 Indoor Dust	Organic dust phase was linked to Cu, the whole mineral fraction of dust with Zn Organic carbon concentrations in indoor dust (median = 28%) were higher compared to soil (median = 5%), Organic carbon controls metal partitioning and their bioaccessibility Due to their distinct geochemical signatures, house dust and soil must be treated distinctly in human risk assessment Median metal concentrations in dust were more significant than in soil	Rasmussen et al. (2008); HERA

Table 2 (continued)

Rank	Topic; sample; location	Reference materials	Findings	Reference
29	Lead bioaccessibility and speciation; house dust; Canada	NIST 2710 and NIST 2711	Industrial proximity was characterized by high indoor metal loading rates The dust metal concentrations did not vary ($.29 \geq p \leq .97$) with proximity to industry Smoker's house had higher metal concentrations and loading rates in dust were unaffected There are significant relationships between metal bioaccessibility and house age Metal concentrations in dust had a strong relationship for Cd, Pb, and Zn Metal concentration data are indicators of the metal sources in home Mass of dust influences metal loadings and loading rates	Rasmussen et al. (2011); ES&T
SBRC				
30	Soil particle and bioaccessibility of lead in adults and children; contaminated soils; Australia	Certified reference material (GBW 07,411)	Particle size fraction had five times the Pb concentration compared to bulk soil Decreasing particle size had elevated the bioaccessibility of Pb Particle sizes < 250 μm can underestimate Pb exposure, larger particle sizes adhere to hands	Juhasz et al. (2011); HAZMAT
31	Metal bioaccessibility in soil, road dust and mine waste in Kank	NIST 2710a Montana I soil standard reference material	As was the major contaminant in soil, road dust and mine slags As bioaccessibility was low in gastric and lung fluids Mineralogy and adsorption properties determine metal bioaccessibilities Highest health risks were from ingestion of soil in children Among all metals, highest risk were posed from As metal	Drahota et al. (2018); EGAH

Table 2 (continued)

Rank	Topic; sample; location	Reference materials	Findings	Reference
32	Bioaccessibility; source apportionment; human health risk assessment, Xiamen, China	Certified reference SOIL GSS-5 (Environmental Monitoring Station of China)	Kindergarten dust samples, were enriched with Ni, Co, Mo, As and Cr The bioaccessibility of metals ranged from 1.56% to 76.51% Traffic and industrial (20.72%), coal combustion (34.09%), natural (18.72%), furniture (7.59%) and unidentified sources (18.87%) were the primary sources as per absolute principal component analysis-multiple linear regression Lower carcinogenic and non-carcinogenic risks from bioaccessible metals than total metals	Ma et al. (2021); EES
Comparison of UBM, SBRC, IVG, PBET				
33	Bioaccessibility of lead in soils, lead concentrations in different fractions, correlation of bioaccessible lead to relative lead bioavailibility	reference material of the vegetables (GBW10015): spinach and soils (GBW07430, GSS16)	Four in vitro assays were employed to measure bioaccessibility of Pb in contaminated soils Relative bioavailability of Pb was measured using a single-dose model A strong correlation was found between the gastric phase of UBM and Pb's relative bioavailability in soils ($r^2=0.67$) Bioavailable Pb in soils was influenced by the sum of exchangeable and carbonated fractions	Li et al. (2015a); HAZMAT
Artificial Lysosomal Fluid and Gamble's solution				
34	In vitro bioaccessibility from reference materials; simulated lung fluids	BCR-723 (road dust collected in Austria with a particle size fraction of	Metals investigated three types of lung fluids (artificial lysosomal fluid, phosphate-buffered saline and Gamble's solution) on standard reference materials representing different particle sources Bioaccessibility was element and speciation dependent Artificial lysosomal fluid extracted higher metals Solid/liquid ratio from 1/1000 to 1/10,000 achieved higher stability of bioaccessibility	Pelfrêne et al. (2017); IJERPH

Table 2 (continued)

Rank	Topic; sample; location	Reference materials	Findings	Reference
35	Mehodological aspects affectng metal release from dust in urban areas in inhalation bioaccessibility tests	Reference material of urban dust (SRM1648a)	Liquid to solid ratio and composition and type of surrogate biological fluids, play vital roles in determining metal bioaccessibility Presence of glycerine in Gamble's solution increases bioaccessibility at a L/S ratio of 5,000 Metal bioaccessibilities were highest for artificial lysosomal fluid with L/S ratio of 5000 Solubility controls metal leaching in Gamble's solution Availibility of metals control metal leaching in ALF and gastric fluid	Expósito et al. (2021); Chemosphere
36	Metal bioaccessibility, metal sources, human health risks; Pb, Cd, Pb, Zn and Cu; windowsill dusts	STDOREAS920st5, OREAS151ast3, OREAS45dst4, and DC73309st2, NIST2711a (Montana Soil II) and BGS-102	Lead had lower lung and gastrointestinal phase bioaccessibility (70–76%), compared to Cu, Cd and Zn (82–92%) Higher Cd carcinogenic risks was found in smelting area compared to urban area Smelting area confirmed their origins from Pb ore and smelting bottom In urban areas they originated from coal burning, diesel engine exhaust and Pb smelting bottom ash	Luo et al., (2022); STOTEN
Integrated stochastic-fuzzy pollution assessment method (ISFPAM)				
37	ISFPAM for heavy metals; bioaccessibility; metal enrichment, urban topsoil; Changsha city, Xiangjiang New District	Not experimental	ISFPAM for metals in soil followed the order Cd > Cu > Pb > Cr > Zn Cd, Pb, and Cu were the primary priority control metals Metal pollution, corresponding to pollution grades, was similar, recommending rechecks for misleading decision-makers	Li et al. (2018a, b); STOTEN

Table 2 (continued)

Rank	Topic; sample; location	Reference materials	Findings	Reference
BCR sequential extraction				
38	Health risk assessment; bioavailability and bioaccessibility; metals; road-dust; zinc smelting plant; China	No CRM mentioned	Pb and Cd and Cu concentrations were higher than the background levels in < 100 and < 63 µm size fractions Reducible and exchangeable fractions had higher concentrations of Pb Oxidisable fractions had higher concentraions of Cu and Cd The bioaccessibility of metals in gastric phases were Cd (58.13%) > Pb (50%) > (19.19%), while that in intestinal phase was Cd (20.36%) > (15.67%) > Pb (5.08%) Highest non-carcinogenic exposure risk was posed from ingestion process Children experienced higher health risks (non-carcinogenic and carcinogenic risks) compared to adults from dust Particle size < 63 µm posed greater risk than those < 100 µm	Zheng et al., (2020); EES
Stochastic Simulation of triangular fuzzy number (SS-TFN)				
39	Metals in size-fractionated road dust; industrial district, Qingshan	National reference substances (GBW GSS-5)	Mean metal concentrations exceeded corresponding soil background value Metal concentrations and corresponding risks to health in industrial and sensitive area were higher than that at residential and traffic area Particle size was inversely proportional to pollution levels and bioaccessibility of metals Winter months recorded higher metal concentrations No non carcinogenic risks, but the risk levels at some points in sensitive and industrial area had a deteriorating trend No carcinogenic risks	Cai et al., (2021); et & i

Table 2 (continued)

Rank	Topic; sample; location	Reference materials	Findings	Reference
In vitro digestion model				
40	Factors affecting the oral bioaccessibility of anthropogenic Pb; soils; Netherlands	One reference sample (ISE 921) was added to each batch of 20 samples to determine accuracy	60.7% -79.0% Pb bioaccessibility in soils polluted with Pb bullets and pellets residues Pb bioaccessibility correlated with reactive Fe, organic matter and pH Oral bioaccessible characteristics influence oral Pb bioaccessibility in soils Incorporate in vitro bioaccessibility tests for analyzing human risk assessment	Walraven et al. (2015); STOTEN

AAQR Aerosol and Air Quality Research, *AECT* Archives of Environmental Contamination and Toxicology, *AG* Applied Geochemistry, *EES* Ecotoxicology and Environmental Safety, *EGAH* Environmental Geochemistry and Health, *EI* Environment International, *EJM* European Journal of Mineralogy, *EP* Environmental Pollution, *ER* Environmental Research, *ES&T* Environmental Science and Technology, *ESPR* Environmental Science and Pollution Research, *ET &I* Environmental Technology and Innovation, *HAZMAT* Journal of Hazardous Materials, *HERA* Human and Ecological Risk Assessment, *IJERPH* International Journal of Environmental Research and Public Health, *IJHEH* International Journal of Hygiene and Environmental Health, *JES* Journal of Environmental Sciences, *JGE* Journal of Geochemical Exploration, *STOTEN* Science of the Total Environment, *TR* Toxin Reviews, *WASP* Water, Air, and Soil Pollution

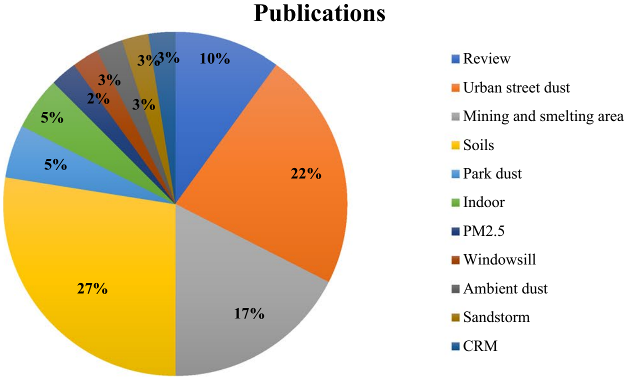

Fig. 3 Publications based on different matrices (*CRM* Certified Reference Material)

mining and smelting areas (7), soils (11), park dusts (2), indoor dusts (1), windowsill dust (1), ambient dust (1), sandstorm (1), PM2.5 (1), Certified Reference Material (1), as given in Fig. 3.

Methods used for Deriving Bioaccessible Metals

Various procedures have been followed in different works of literature to derive the bioaccessible metals like the SBET, PBET, SBRC, UBM, Modified Toy Safety extraction and Toxicity Characteristics Leaching Procedure (TCLP), as given in Table 2.

Their evolution has resulted in simplified methods (e.g., SBET) for cheap and rapid estimations of potentially harmful elements exposure risk formulation or methods with greater complexity to better form hazard-risk models. The SBET method can only determine metals bioaccessibility in the gastric phase, while PBET derives bioaccessible metals from both gastric and intestinal phases. There are also slight alterations in the different procedures used (differences in the time of mixing, standing time, or time for centrifugation) for deriving the bioaccessible metals from either the gastric or intestinal phase. The cheaper methods used for estimating metal bioaccessibility only provide a rough estimate like SBET. Although SBET is both time and cost-effective, it can only assess metals bioaccessible in the gastric phase. While PBET accurately differentiates metals in both gastric and intestinal phases and is cheap but has no regulatory guidance supporting it. On the other hand, SBRC method can only be used in lands contaminated with Pb and As. Only the Unified Bioaccessibility (BARGE) procedure is certified as per ISO17924 as more accurate and informative, but it is slow and expensive.

Bioaccessible Metals in Different Dust and Soil Matrices

Metal Bioaccessibility in Soil

Heavy metals in soil is of immense concern due to the rapid urbanisation and industrialisation (Li et al. 2019; Egbueri et al. 2020). Their sources may vary from natural to anthropogenic (Wang et al. 2020b). The heavy metals in soils are derived from transportation, industrial emissions (Hu et al. 2017) and fertilizers, pesticides, fungicides and wastewater irrigation (Marrugo-Negrete et al. 2017; Sandeep et al. 2019).

Metal bioaccessibility tests have been performed in different matrices such as soil, street dust, indoor dust, airborne particulate matter, vegetables, and human tracers like blood (Ettler et al. 2012). Among them, the soil was the most studied matrix. In 2012, Ettler studied the differences in the metal bioaccessibility in soils of Zambian Copperbelt, affected by mining and smelting activities. Metal bioaccessibilities were higher in smelting areas compared to the mining areas. Even the topsoils, indicated higher metal concentrations, due to the dust fallout from smelting activities. The SBET method's bioaccessibility in smelting areas for As, Pb, Cu, Zn, and Co were 40%, 73%, 60%, 49%, and 38%, respectively. At the same time, the corresponding values in mining areas were 12%, 41%, 57%, 45%, and 34%. Severe health risks were indicated from the high bioaccessibility of Cu (80–83%), Co (58–65%), and Zn (79–83%). The primary source of these metals were mine wastes, tailings, smelter stacks, and chalcanthite.

The differences in the bioaccessibility of metals based on different size fractions of soil through incidental ingestion using SBRC method were studied by Juhasz (Juhasz et al. 2011). The four different size fractions were: ≤ 2 mm, < 250 µm, < 100 µm and < 50 µm. Lead was highly enriched and bioaccessible in fractions of size < 50 µm. They concluded that considering particle size fractions of < 250 µm for incidental ingestion might underestimate Pb exposure. This is so because even smaller fractions will have a greater affinity for adhering to palms.

Contaminated areas like mining or smelting are hotspots of metal contamination and their bioaccessibility. In a study on the contaminated soils near the copper smelter of San Luis Potosi, Mexico, 90% of soil samples had concentrations of Pb (400 mg/kg) and As (100 mg/kg) higher than the recommended guidelines of USEPA (Carrizales et al. 2006). The primary sources of this pollution were industries and smelter stacks. PBET method evaluated the bioaccessibility for As and Pb, which were recorded as 46.5% and 32.5%, respectively. Further, children's blood lead levels were found above the C.D.C.'s (Centres for Disease Control) action level of 10 µg/dl in 90% of children. Using the Integrated Exposure Uptake Biokinetic (IEUBK) Model for lead in children, the soil pathway contributed to 87% of total information in blood. Also, the As exposure dose using Monte Carlo in children of Morales was above E.P.A.'s reference dose.

Metals can also be bioaccessible from the topsoils of urban areas. In such a study, Roussel studied the bioaccessibility of Zn, Pb and Cd levels of urban topsoils (lawn, kitchen, garden) from twent-seven locations using the UBM (Pruvot et al. 2010). The soil samples were from Metaleurop (M.E.) at Noyelles and Umicore (UM.) at Auby. The average Zn, Cd,and Pb, concentrations were 15, 984, and 1941 mg/kg. The sources were mainly anthropogenic, like garden slag, used as a herbicide, ashes from domestic coal combustion, and dust emissions from Metaleurop Nord. Around 47%, 62% and 68% of Zn, Pb, and Cd were accessible from the gastrointestinal phase of soils. Metals extracted using U.B.M. and total metal trace elements were strongly correlated. Physico-chemical parameters like total nitrogen, carbonates, clay contents, and pH were also affected by human bioaccessibility, as per multiple regression analysis. They concluded that estimating bioaccessible metal concentrations for risk estimations would be more realistic for future assessment predictions.

Metal concentrations in soils of parks and lawns of Guangzhou were studied by Gu et al. (2016). The metal concentrations, bioaccessibility, health risks, and source identification of metals were studied. Cd had the highest bioaccessibility of 75.96%. Non-carcinogenic risks were negligible, but Pb and Cr posed carcinogenic risks, which were within acceptable levels. Most of the metals originated from anthropogenic sources, although Fe and Ni had mixed sources.

In another study, soil mineralogy, oral bioaccessibility, and risk assessments from land use were studied (González-Grijalva et al. 2019). Traffic paint had higher metal concentrations of Cr, Pb, Zn, Ca, quartz, crocoite, kaolinite, and calcite. The percentages bioaccessibility of Pb metal in gastric and intestinal phases ranged from 40–51% and 24–70.5%, respectively. Strong correlation of bioaccessible Pb in the intestinal phase was found with kaolinite. An interesting finding was the variation of Pb bioaccessibilities with the change of soil type. The release of Pb from the gastrointestinal phase was also found to be determined by the mineralogy of the soil.

Among other studies, health risks and bioaccessibilities of metals were studied in Jaozuo, China (Liu and Han 2020), bioaccessibilities of metals from contaminated soils of Paris (Pruvot et al. 2010), from soils of an abandoned mine site (Mehta et al. 2020), bioaccessibility of lead in adult and children from contaminated soils of Australia (Juhasz et al.

2011). The detailed findings of these studies are given in Table 2.

Metal Bioaccessibility in Street Dust

Street dust acts as a significant sink of toxic trace metals, which might have several sources (like industries, domestic heating, waste incineration, anthropogenic activities, and vehicles) through local human activities and atmospheric transport (Trujillo-González et al. 2016; Harada et al. 2019; Cui et al. 2020; Hanfi et al. 2020).

Apart from soils, many authors have also studied the bioaccessibility of street dust by implementing various methods. Yu reviewed the bioaccessibility of metals by applying the SBET and TCLP of metals in Tianjin (Yu et al. 2014). Ingestion of dust was the primary exposure route, and related health risks were evident from metals like Cd, Cr, As Pb, and Cu. With the increased exposure frequency and ingestion rates, these metals had an increased potential of posing noncarcinogenic risks in children. The primary sources of these metals in Tianjin were anthropogenic activities like coal burning, vehicular emissions, and industrial waste. Future, bioavailability, and bioaccessibility studies were needed to estimate risks to the environment and humans appropriately.

The oral bioaccessibility and health risks to humans from Zn, Mn, Pb, V, Mn, Cd, Fe, Co, Cu, Ni, Hg, Cu and As of street dust in Nanjing, were studied by Hu et al. (2011). Total and bioaccessible metals (Simple Bioaccessibility Extraction Test) were investigated and strongly correlated with pH and organic matter contents. Only children were exposed to significant noncarcinogenic risks. Carcinogenic risks were within permissible limits for adults and children from As and Cr.

In 2017, Li studied the street dust from different functional areas viz commercial, traffic, educational, residential, and park areas of Chengdu, China (Li et al. 2017). The pollution sources were mixed here, ranging from erosion and abrasion of tires, building materials and batteries, fertilizers, natural and anthropogenic emissions, traffic, domestic, industry, and tall buildings to high population density and socio-economic activities.

Padoan incorporated both street and soils to study the bioaccessibility of metals (Padoan et al. 2017) in Turin, Italy, following the SBET procedure. Simple Bioaccessibility Tests were performed to estimate the metal concentrations and assess their bioaccessibility in varying size fractions of street dust and their corresponding soils (< 2.5 µm, 2.5–10 µm, 10–200 µm). They found that metal concentrations of metals like Fe, Mn, Cu, Pb, Sb, and Zn were highest in the smallest size fraction of < 2.5–10 µm. Cu, Pb, Sb, and Zn concentrations in street dust were mainly derived from non-exhaust sources. In fraction sizes < 2.5 µm, Zn was dominant from industrial sources Cu, Pb, Zn and Ni were dominant in traffic sites. Additionally, bioaccessibility Ni, Cr, and Fe were greater infractions of size < 2.5 µm and 2.5–10 µm. Zn was highly bioaccessible in road dust at traffic conjunctions; the metal concentrations (mg/kg) of different metals in road dust (cumulatively of all size fractions) were as:—Fe (15,411–32,323), Mn (343–843), Cu (180–333), Cd (1–2), Cr (161–517), Ni (148–296), Pb (81–233), Sb (1–29) and Zn (240–942). The corresponding values in soil(mg/kg) were:—Fe (28,864–46,001), Mn (579–1179), Cu (88–287), Cd (1–2), Cr (168–432), Ni (238–303), Pb (289–952), Sb (6–17) and Zn (305–80). Here, traffic was identified as the primary source for Zn, Sb and Cu and bioaccessible fractions of Fe and Mn.

Okorie et al. (2012) studied the bioaccessibility through street dust in Newcastle, following the BARGE method (Okorie et al. 2012). The percentages of metal bioaccessibility were higher in gastrointestinal phase (Zn = 53.2%, Ni = 32.4%, Cu = 64.4%, Cd = 52.8%, Pb = 37.2%, As = 36.1%) compared to gastric phase (Zn = 37.6%, Ni = 26.8%, Cu = 30.2%, Cd = 41.7%, Pb = 32.9%, As = 18.6%). Here sources like lubricants, vehicle fabrication, flashing on the roof of historic building parts, tire wear, cylinder surface, and engine pistons were dominant. An important finding was that it is nearly impossible for any child, even with the habit of pica, to ingest 100 mg/day dust, even from polluted city centres. However, models like IEUBK are often used for estimating blood lead levels in children, using such default values, thus always overestimating the risks.

Although several studies have established the metal concentrations in street dust, limited research and knowledge are available on its molecular composition to date (Potgieter-Vermaak et al. 2012). The chemical composition of inhaled particles is involved in manifesting carcinogenic, toxic and enotoxic effects. Fractions of size < 38 µm had the highest concentrations of Cr (171 mg/kg) and Pb (238 mg/kg), which decreased in the larger fractions. > 50% Cr rich particles were associated with Pb, which were specially bound in form of lead chromate. Both Cr and Pb were readily mobilised in artificial lysosomal liquid, and upto 19% and 47% of Cr and Pb were released. This poses serious health risk concerns to humans.

Metal Bioaccessibility in Park Dust

Urban parks are used extensively by children as playgrounds, and adults for walking, sitting or exercise (Liu et al. 2020; Zhao et al. 2020; Huang et al. 2021). Metals in urban park dusts, easily get resuspended by wind, hence are of concern to human health (Qiang et al. 2015; Yang et al. 2015). Park dust was also studied for different metal bioaccessibility by Wang in Nanjing and Yang-Guang Gu et al. (2016) in Guangzhou in 2016. Wang found anthropogenic origins of elements in urban park dust, except that of Co and V like vehicle exhausts and coal combustion, cement. The concentrations of different metals (mg/kg) were as follows:—As = 24 ± 5.01, Pb = 63.3 ± 10.5, Cd = 60.9 ± 17.5, Cr = 10.1 ± 2.99, Cu = 37.9 ± 11.6, C= 32.8 ± 7.85, Ni = 25.5 ± 8.60, Zn = 53.7 ± 14.7, V = 29.9 ± 57.6, Mn = 51.6 ± 10.7. The primary sources of pollution were anthropogenic, which varied from vehicle exhausts, coal combustion, cement, and other natural sources. The non-carcinogenic risks were mainly posed through the ingestion pathway. Although hazard quotients were within safe limits, As (0.184) and Pb(0.154) posed higher carcinogenic risks in children than in adults. The carcinogenic risks were $< 10^{-4}$ for As and $< 10^{-6}$ for Cr, Co, Cd, and Ni. The metal bioaccessibility followed the order:—Cr < As < Ni < V < C< Cu < Mn < Zn < Cd < Pb.

Yang-Guang Gu found the highest bioaccessibility percentages of Cd = 75.96%, followed by Mn = 33.66% and Zn = 31.87% (Gu et al. 2016). The sources were identified both as natural and anthropogenic. The carcinogenic risk from Cr and Pb were within limits ($< 1 \times 10^{-4}$), while there were no noncarcinogenic risks in Guangzhou urban park soils.

Metal Bioaccessibility in House Dust

The indoor environment is a potential exposure route for multiple pollutants in all age groups due to its longer residence time than outdoors. Time spent indoors varies from 80 to 90%, depending on seasonal and vocational activity. Also, indoor pollutants are not degraded due to the absence of biotic (microbial) and abiotic (photolysis and hydrolysis), aging, or physical factors like wind or rain. Different sources of metals indoors may vary from wall paints, pesticides, chemicals used on furniture, smoking, batteries, and wood burnings. Although house dust is an essential category/component/zone/aspect for studying metal bioaccessibility, very few studies have addressed. Among the different authors, Rasmussen studied metal bioaccessibility in house dust (Rasmussen et al. 2008, 2011, 2013). In 2008, he examined the various factors affecting oral bioaccessibility, like total organic carbon content, metal speciation, and size of particles for risk assessment estimations in Ottawa city, Canada. Two different size fractions (< 36 µm and 80–150 µm) were studied for the speciation of Zn and Cu using synchrotron X-ray absorption spectroscopy (X.A.S.). While Cu was associated with the organic dust phase, Zn was associated with the mineral fraction. Carbon concentrations in indoor dust (median 28%) were elevated compared to soil (median 5%). Bioaccessible metals (Zn, Cu, and Ni) were evaluated for size fractions < 150 µm, and house dust was found to have higher bioaccessible metals than soils. Soil and house dust had distinct geochemical signatures and thus were suggested to be treated separately to assess human risks.

In 2011, he studied the bioaccessibility and speciation of lead in Canadian house dust (Rasmussen et al. 2011). A polymodal frequency distribution was obtained, which consisted of three lognormally distributed subpopulations defined as "elevated," and "anomalous," “elevated” and "urban background," with geomeans of 1730, 447 and 58, 447, and 1730 mg/kg. Around 90% of samples fell under the "urban background" category. Older homes from central cities recorded elevated metal concentrations. Moderate correlation was found between age of house and PbS content in dust ($R^2 = 0.34$; $n = 1025$ at significance of $p < 0.01$). 33% of older homes fell under the "urban background" category due to the benefits of home remediation. The dominant Pb species were all forms of citrate, carbonate, hydroxyl carbonate, chromate, oxide, sulphate forms of Pb, elemental Pb, and those adsorbed to humate or Fe- and Al-oxyhydroxide Pb bioaccessibility was higher in older homes with access to carbonates and hydroxyl carbonate compounds of Pb in older paints. This study provided a national baseline for management of human health risks in urban areas.

Again, in 2013, Rasmussen studied the population-based metal concentrations, metal loads, and their loading rates (Cd, Cr, Ni, Cu, Cd, As, Pb and Zn) inside Canadian homes (Rasmussen et al. 2013). Here, industrial proximity was attributed by high metal loading rates in indoors. In contrast, metal concentrations in dust were unaffected. Significant relationships were found between metal concentrations and the age of the house, only for Zn, Pb and Cd. Although metal concentrations are a good indicator of indoor metal sources, dust mass was the key factor influencing metal loadings and loading rates.

House dust near contaminated sites like mining or industries are more likely to expose its inhabitants to metals/pollutants. One such study was done by Rieuwerts, who studied the metal levels and bioaccessibility in indoor dust and garden soils from a site with a mining history (Rieuwerts et al. 2006) in southwest England. The elevated mean As concentrations in indoor dust 149 mg/kg and garden soils were 262 mg/kg. In indoor and garden grounds, their highest soil and indoor dust concentrations were 471 mg/kg and 486 mg/kg, respectively, as concentrations exceeded the U.K. Soil Guideline Value (S.G.V.) of 20 mg/kg. Poor correlation between house dust and garden soil for As metal were found. Metal bioaccessibility was studied using PBET methods, ranging from 10 to 20% in the gastric phase to 30–40% in the intestinal phase. The average As dose intakes in children (0–6 years) from indoor dust and garden soils were 3.53 $\mu g kg^{-1}$ body weight day^{-1} and 2.43 $\mu g kg^{-1}$ body weight day^{-1}, respectively, compared to the index dose of 0.3 $\mu g kg^{-1}$ body weight day^{-1} from S.G.V. The ingestion doses through indoor dust and garden soils exceeded 75% of samples of children (0–6 years old). This age group is more prone to soil and indoor dust ingestion pathways. Hence, significant As contamination and its implications in southwest England need more concern.

Turner studied the concentrations of C, H, N, and metal in 32 household dust samples in the U.K. (Turner and Ip 2007). Although the total metal concentrations were variable, the geometric mean metal concentrations were consistent with contemporary literature worldwide. The greatest enrichments were found in Pb, Zn, Cu, Sn, and Cd, while the bioaccessibility in simulated gastric fluids ranged from 80% for Pb, Cu, Zn, and 10% for Sn. Combustion-metrically measured C, H and N concentrations in house dusts ranged from 11–46.2%, 1.5–7.0% and 1.0–8.5%, respectively. The uniform carbon to hydrogen ratios (7.3; rsd = 10%) reflected their similar origin (rsd = relative standard deviation). While C:N ratios were variable (mean 8.5; rsdN40%), reflecting source materials variance (protein = 2.1; soil humic = 14; lignin = 78). The heterogeneous distribution of metals from various internal and external sources are attributed to poor correlations between metal concentrations or C, H, and N ratios.

In 2018, Plumejeaud studied the characterisation and genotoxicity of bioaccessible fractions in house dust from Estarreja in Portugal (Plumejeaud et al. 2018), The bioaccessibility of the metals were studied by following the UBM method. The gastric extracts induced genotoxicity in adenocarcinoma gastric human cells, which were dose dependent. Cu was found to induce DNA damage while Pb induced chromosomal damage effects. The usage of such methodologies could be used in large-scale studies, for better estimations risks and exposures to humans.

Knowledge Gaps

Further, a comparison of the efficacy of various simulated fluids and different approaches for different matrices was lacking. The establishment of time of extraction, solid to liquid ratio, and method/intensity of agitation for a clearer understanding of the influences of other methodological parameters on the outcomes of bioaccessibility was required. Presently, there is no distinction between solubilized lung fraction and the fraction cleared via mucocilliary actions. The role of microorganism on the metals release in the respiratory tract, is yet unexplored. A knowledge about the physicochemical properties of dust in relation to its metal speciation will provide a better understanding about the mechanisms involved in their bioaccessibilities. The different toxicity modes for mixed sources of contamination must be understood for in-vitro studies of cell culture. A better understanding of the physicochemical-biological factors influencing bioaccessible, and bioavailable responses is essential for building up a simple, cost-effective, and rapid approach for refining inhalation exposure in humans. Among the different media studied for metal bioaccessibility, rice (Li et al. 2018b), wheat grains (Wang et al. 2020a), vegetables (Hu and Cheng 2013), particulate matter (Hu et al. 2012) and marine organisms (Gu et al. 2018) should also be studied at a larger scale, as these are directly ingested or inhaled by the humans.

On comparing the different methods followed for the estimation of metal bioaccessibility, the SBET method has been used most extensively used. One of the reasons could be the less time (1 h) required for extraction and the lesser chemicals required for this method. But, at the same time, it can only estimate the metal levels only in the gastric phase. All other methods (PBET, SBRC, BARGE) estimate the metals accessible to the gastric as well as the intestinal phase. These methods take approximately another 4 h to extract metals accessible to the intestine. Thus, although SBET is more economical and timesaving, the focus should be laid on gaining more accurate knowledge of any metal's bioaccessibility in both the compartments (gastric and intestinal). Further, research to develop more economical and less time-consuming methods for the estimating metal bioaccessibility should be encouraged.

Control Measures

Different authors have advocated various control measures to curb the bioaccessibility of metals from dust, as enlisted below.

- Immobilization of Pb in contaminated soils/waste/slags by addition of a phosphate amendment, which forms stable Pb phosphate compounds.
- Removal of waste piles, paving streets and roads, and plantation of grass or vegetation will help reduce access to wind-blown dust.
- Reduction dust load by proper regular vacuum cleaning and sweeping.
- Assessment of practical risks, by local measurements of background exposures.
- Determination of metal concentrations in dust and their bioaccessibility at regional and national levels for obtaining default values. Incorporation of these default value databases will help in deriving accurate risk assessments.
- Maintenance of hygienic conditions indoors and outdoors to avoid potentially harmful effects on the population.
- Incorporation of control methods like water washing, street sweeping, and dust suppressants.
- Promotion of mass transport, and installation of industries away from human settlements.

- Raising public awareness and promoting risk assessment studies to help the decision-makers suggest remedial measures or reduce the risks from exposure.
- Use of protective gears like eye-protecting coverings, masks, and uniforms must be adopted in occupational workplaces.
- Comprehensive dust quality assessments and regular multi-compartmental environmental surveillance and remediation program are required.
- Incorporation of microbial assays, plant and soil invertebrates, and in soil quality improvement.
- Development of integrated risk assessment of pollutants in soil based on land use and environmental availability.
- Model the urban environment in geochemical cycles, considering the continuously changing complex mixtures of materials.
- Identification of the critical exposure pathways and social justification for investment in risk reduction programs
- Risk assessment and management of metals through bioaccessibility tests.

Limitations

Till date, metal bioaccessibilities have been studied at either regional or local scale. Larger database of bioaccessible metals is unavailable. Such kind of database will help in making more realistic models, with lesser chances of overestimations (like IEUBK). Bioaccessibilities of metals from other matrices (like air) and exposure pathways (inhalation, dermal contact) need to be standardised as well. Human biomarker (hair, nail, urine) studies will further confirm the effects of bioaccessible metals. Source apportionment studies using metal isotope fingerprinting need to be taken up at a larger scale, for confirming the accurate sources of metals.

Future Scope and Recommendation

- Size fractionation of dust and its relationship with risk assessment of human inhalation.
- Studies on the toxicological effects using a multi-element matrix should be considered when dealing with the bioaccessibility/bioavailability from multi-media like soil, water, food, and vegetables.
- Future research must be taken much more robustly at the microbial level. Like, as both indigenous and exogenous microbes' role is in releasing metals/metalloids from the respiratory tract.
- A mechanistic understanding of contaminant release factors can only be understood by combining in-vitro/Vivo studies with dust properties and their speciation. Also, in-vitro cell culture studies help understand toxicity modes from mixed contaminant sources.
- Efficiencies of fluids, static vs. dynamic flow-through systems approaches, and correlations between in-vitro—in-vivo samples in multiple matrices s like mine/road dust, vehicular exhaust, ambient particulate matter need to be studied.
- Standardisation of effect of solid/liquid ratio, time for extraction, and method/intensity of agitation for understanding the methodological parameters' influencing metal bioaccessibility.
- Chemical speciation of metals, their mobility, bioavailability, and total concentration should be considered for evaluating metals' potential risk (environmental/human health) from any medium (soil/dust).
- A rapid, simple, and economical approach for exposure through inhalation can be refined by understanding the factors (physical/chemical/biological) in combination with in-vitro/vivo studies.
- Validation of relationships between in-vivo-in-vitro cultures, physicochemical and biochemical factors influencing the responses of bioaccessibility, and bioavailability will refine inhalation exposure in humans in a simple, accurate, yet economical method.

Conclusions

Metal bioaccessibility has been studied most widely in the soil matrix. The potential sources of metal pollution varied from vehicular, industrial, and mining/smelting activities. Here, source apportionment studies can help determine the exact contribution of each source and precisely manage the sources. Size fractionation of dust particles played a vital role in deciding the bioaccessibility of metals. Smaller size fractions of dust had higher bioaccessibility. Metal concentrations are a good indicator of various sources of metals in home; dust mass is the key factor influencing metal loadings and loading rates. Emphasis should also be given to dust loading rates, which ultimately determine metal loadings. Factors affecting bioaccessibility like pH, reactive Fe, organic matter, occupation, and exposure time in workplaces and homes need detailed understanding. The role of microorganisms in releasing metals in the respiratory tract is yet to be explored. The different modes of toxicity for mixed sources of contamination need to be understood for in-vitro cell culture studies. Among other extraction methods, although SBET is the oldest, PBET is more widely used for studying metal bioaccessibility. In recent times, PBET is largely replaced by UBM as it employs a standardised method for assessing bioaccessible metals. However, standardization of extraction procedures, with precisions on solid to liquid ratio, needs attention. In different population-based risks, children were more vulnerable to informal digestion risks. Regional databases for population risk assessments need to be framed for obtaining a representative baseline for human health risk management. Proper attention should be given to indoor environments like homes and offices, about metal exposures, where an individual spends the maximum time. A better knowledge of the biological and physicochemical factors determining the bioaccessibility and bioavailability, and consequent toxicological responses is essential for formulating a simple, cost-effective, and rapid approach to toxicological exposure and human health risk assessment.

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Data availability All data generated or analysed during this study are included in this published article [and its supplementary information files].

Declarations

Conflict of 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.

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