Journal of Hazardous Materials, Volume 397, October 2020, Article number 122720
DOI:10.1016/j.jhazmat.2020.122720

Comprehensive review of the basic chemical behaviours, sources, processes, and endpoints of trace element contamination in paddy soil-rice systems in rice-growing countries

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Abstract

Rice is the leading staple food for more than half of the world's population, and approximately 160 million hectares of agricultural area worldwide are under rice cultivation. Therefore, it is essential to fulfil the global demand for rice while maintaining food safety. Rice acts as a sink for potentially toxic metals such as arsenic (As), selenium (Se), cadmium (Cd), lead (Pb), zinc (Zn), manganese (Mn), nickel (Ni), and chromium (Cr) in paddy soil-rice systems due to the natural and anthropogenic sources of these metals that have developed in the last few decades. This review summarizes the sources and basic chemical behaviours of these trace elements in the soil system and their contamination status, uptake, translocation, and accumulation mechanisms in paddy soil-rice systems in major rice-growing countries. Several human health threats are significantly associated with these toxic and potentially toxic metals not only due to their presence in the environment (*i.e.*, the soil, water, and air) but also due to the uptake and translocation of these metals *via* different transporters. Elevated concentrations of these metals are toxic to plants, animals, and even humans that consume them regularly, and the uniform deposition of metals causes a severe risk of bioaccumulation. Furthermore, the contamination of rice in the global rice trade makes this a critical problem of worldwide concern. Therefore, the global consumption of contaminated rice causes severe human health effects that require rapid action. Finally, this review also summarizes the available management/remediation measures and future research directions for addressing this critical issue.

Keywords: Trace metal contaminants; Paddy soil system; Health effects; Transportation mechanisms; Accumulation

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The final published version (version of record) is available online at DOI:10.1016/j.jhazmat.2020.122720. Please refer to any applicable publisher terms of use.

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1. Introduction

The word “paddy” is taken from the Malay word “padi,” which means “rice plant”; paddy soil, a well-known soil type, is submerged during rice cultivation [1]. Trace metal contamination in paddy soil-rice systems poses a severe threat to the production of safe rice crops worldwide [2]. Trace element contamination in terrestrial resources is a focus of various environmental studies and requires more attention worldwide due to the toxicity, persistence, and bioaccumulation of these elements [3, 4]. Agricultural soil has been contaminated by untreated waste discharge, emissions from rapidly intensifying industrial areas, increasing mining activities, chemical fertilizer use, industrial effluent disposal practices, and domestic wastewater [5]. Trace metals such as As, Se, Cd, Pb, Zn, Mn, Ni, and Cr constitute an imprecisely defined group of inorganic biochemical hazards that are mainly found at polluted sites [6, 7]. Soils are a substantial sink for trace metals released in unconfined environmental matrices due to human activities. Unlike organic pollutants, trace metals are not oxidizable to carbon dioxide (CO_2) through microbial action. Trace metals do not undergo biochemical degradation [8], and they persist in the soil system for a long time once they are released into the environment [9]. The presence of trace metals in paddy-soil systems can severely constrain the biodegradation of organic pollutants [10].

The excessive concentrations of trace elements in agricultural soil systems result in increased trace element uptake by food crops such as paddy rice, wheat, maize, and vegetables [11]. Trace element contamination in paddy soil-rice systems poses severe a threat to the human population as well as to ecosystems *via* direct ingestion or interaction with polluted soils and contaminated water; these contaminants become a part of food chains, such as the paddy soil-rice-human and paddy soil-rice plant-animal-human food chains. The rapid increase in the human population has led to a decrease in food quality and food safety due to the attenuation of agricultural lands, which has triggered food uncertainty and agricultural land occupancy problems [10]. Unpolluted agricultural areas are polluted with trace metals from polluted water that comes directly from cities or from other anthropogenic sources and is used for irrigation purposes; the constant use of agro-chemicals and fertilizers is another source of trace metals [12, 13]. Such paddy soils expose humans to a high level of trace elements, and children are at a significantly higher health risk than adults according to risk assessment indices [11].

Paddy cultivation in this respect is of extreme concern since crops need a very high quantity of irrigated water from seed growth to crop harvesting [14]. Approximately 160 million hectares of the global agricultural area is under rice cultivation, and rice is the leading staple food for more than half of the world's population [15]. The category of total agrarian land includes paddy rice cultivation, and the average paddy rice production has progressively increased from 1.8 metric tons/hectare in 1960 to 4.3 metric tons/hectare currently [2]. Over the last few decades, rice consumption has increased, which has made it a valuable product [16]. Approximately 92% of the total world paddy rice is produced in Asia, and paddy rice is a well-known staple food consumed by most of the world's population [17]. Trace metal pollution in the paddy-soil system and its successive accumulation in different parts of the rice plant (roots, shoots, leaves, and grains) have been observed in different rice-growing countries [18]. Hence, trace metal contamination in paddy soils and its transfer in paddy soil-rice systems are a growing concern [19]. Several studies in various countries have investigated certain specific trace elements, such as As, Se, Cd, Pb, Zn, Mn, Ni, and Cr [11].

Currently, measuring or predicting the likely human health risks of trace elements that primarily originate from exposure to metal-polluted soil, water, and atmospheric deposition is becoming more common [20]. These trace elements are mainly studied in works evaluating the potentially toxic trace elements through human health risk assessments specifically targeting children and male and female adults [21]. The human health risks of metal-polluted soils can be measured by using the soil pollution indices of trace elements, soil to plant transfer factors, and their concentrations in rice systems [20, 22]. Recent studies have stated that trace element accumulation in paddy soil systems is related to risks to soil fertility and quality, as well as soil biochemical activity such as microbial and enzyme activity. Elevated trace element concentrations in paddy soil systems are primarily known to affect the microbial count in soil and the associated microbial activities, which directly influence the fertility of the soil [23]. Furthermore, the public is becoming more conscious of the implications of trace element contamination in agricultural soils and its severe toxic effects on human and environmental health as well as of the enhancement and expansion of technologies for trace element removal/clean-up at polluted sites [24]. Hence, it is imperative to deploy recent advances and various site-specific contamination clean-up

technologies that might feasibly and effectively remediate trace element pollution in soils. In addition, a summary of recent review papers about trace element contamination in soil-plant and rice system topics, objectives and gaps is discussed in Table S1.

The present study aims to systematically review and understand the basic behaviours of trace metals in the soil system to assess sources, uptake, translocation and accumulation mechanisms, the status of trace element contamination, and associated risks in paddy soil-rice systems in rice-growing countries and contaminated soil management/remediation methods.

2. Trace element contamination in soil: Possible sources

2.1. Natural sources

Trace metals persist naturally in the soil environment mainly due to pedogenic processes of parent material weathering at a concentration that is regarded as trace (<1000 mg/kg) and are infrequently poisonous [10]. It is deceptive that the significant preliminary source of trace metals in the soil system is the parent material [25]. The effect of parent materials in defining the trace element content is notable in soil systems, which have formed from the same or an individual parent material, such as igneous rocks, sedimentary rocks, and/or new soils weathered under temperate environments [26]. As soon as the rock weathering process takes place, the main crystal-like structures of the raw materials are entirely broken down and allow trace metals to either be adsorbed on the soil surface or transported to the surface or groundwater system [2]. The effect of rock parent materials on the type and content of trace elements in the soil system are mainly reliant on pedogenic processes [27]. However, trace metal concentrations in soil systems are primarily derived from pedogenic parent materials, and the physicochemical and biological features of soil determine the extent of trace element accumulation [28]. Hence, trace element contamination in the soil system primarily results from the natural availability of extensive parent material (glacial till), and anthropogenic activities are also significant sources [29].

Accordingly, uncovering a strong association between trace element status in the soil system and the parent material is demanding [30]. According to [31, 32], a compilation of results from the monitoring of significant sources of trace elements in paddy soil and a pollution assessment of the Yangtze River Delta and Fuyang County in China, respectively, suggested that the Ni concentration of the paddy-soil system was determined to be high

due to the parent material and that Ni showed the most extreme high concentration of all contaminants.

Another study from Dongguan, China, described the status of trace element levels in agricultural soils and in different vegetables with diverse key sources. Cu, Zn, Ni, Cr, and As, were mainly derived from natural rock parent materials. Nevertheless, Cd and Pb were present due to anthropogenic activities and were above the recommended concentrations of trace metals in soil systems; this was not true of the parent material [33].

The critical relationship between mafic rock types and levels of trace elements such as Ni, Al, Mn, Mg, and V in the soil system suggests the significance of natural rock parent material to the spatial dispersal of these trace elements [34]. Mafic and ultramafic rocks accumulate trace metals, including Cd, Cr, and Ni, leading to higher concentrations in soil-plant systems [35]. In Pakistan, studies conducted by [36, 37] on soil and vegetable accumulation of trace metals from natural geological sources in Gilgit, northern Pakistan, and trace metals in the piedmont soils of the Peshawar Basin, Pakistan, respectively, suggested higher levels of trace elements, including Cd, Pb, Zn, Cu, Cr and Ni, in agricultural soils that were primarily released from parent rocks and calcite rocks. The release of Mn and As mainly from natural aquifer rocks has been reported in soil and groundwater systems in different locations in Bangladesh and West Bengal, India [38, 39]. Nevertheless, although trace metals usually originate from natural sources, the type and elemental composition of parent material, the extent of the rock weathering process, the physicochemical and biological properties of soil, and the climatic environment seem to have a significant effect on the occurrence and distribution of trace elements in soil systems [40].

2.2. Anthropogenic sources

2.2.1. Agricultural activities

(a) ***Intensive pesticide use***. The term "pesticides" includes any combination of substances intended for preventing, destroying, resisting and mitigating any pests such as weeds, nematodes, insects, mites, and rats. Consequently, it comprises insecticides, nematicides, herbicides, fungicides, molluscicides, and other materials extensively used for pest control [2]. Despite the significant influence of pesticides on worldwide agricultural production, the adverse human health effects linked with them is causing

increasing concern [41]. However, the intensive use of insecticides has grown extensively over the past decades, and in recent decades, approximately 2.3 million tons of pesticides have been used yearly [42].

Extremely hazardous substances, such as persistent organic pollutants (POPs), organophosphates and organochlorine insecticides such as dichlorodiphenyltrichloroethane (DDT), chlordane, lindane, and chlorinated hydrocarbon endosulfan) are used in the agricultural application of different conventional pesticides reported to contain significant levels of trace elements and enter environmental matrices [43]; these pesticides are based on chemical mixtures comprising Cu, Ni, Cr, Mn, Cd, As, Pb, and Zn [10]. The Bordeaux mixture of copper sulfate ($CuSO_4$) is commonly used to control mycological attacks in several crop systems [44]. Arsenic and As-containing compounds, such as lead arsenate ($PbHAsO_4$) insecticides, have been intensively used in the past to control pests in different crop systems [45]. However, the intensive use of fertilizers is also common practice to reach maximum agricultural production from a unit area of land [46]. Agrochemical fertilizers usually contain significant concentrations of different trace metals and their metalloids, including As, Cd, Cr, Zn Cu, Mn, Ni, Zn, & Pb [2]. Radionuclide trace elements such as (U, Ra, & Th) have also been introduced into agricultural soil along with fertilizers [47]. Nevertheless, some of the trace metals (Cu, Mn, Mo, Zn, Ni, & Fe) at low concentrations are considered essential nutrients [48]. Agrochemical fertilizers contain significant levels of trace elements such as Ni, Pb, Fe, Co, & Cd as impurities [2].

(b) ***Intensive use of fertilizers***. Worldwide, the extensive use of fertilizers for agricultural purposes has increased exorbitantly in the previous six decades. In 2006, the World Resources Institute (WRI) specified that five key countries, China, the United States, India, Indonesia, and Brazil, were responsible for two-thirds of all fertilizer consumption; China ranked first with approximately 32.9% (55925.6 thousand metric tons) consumption of total fertilizer worldwide [49]. Fertilizer usage in India has increased intensively to 133 kg/ha in 2011 from 1 kg/ha in 1951-1952 [49]. Among the various types of fertilizers, the application of phosphate-containing fertilizers is the most common. It is a dynamic factor in the sustained agricultural growth and high productivity of rural farming in several countries around the world [50].

In Asia, the typical application of phosphate-based fertilizer is approximately 34 kg/ha [49]. However, the concentrations of pollutants in fertilizer vary significantly, especially the concentrations of trace metals. According to the literature, the application of phosphate-based fertilizer at a rate of 20 kg/h/year in soil leads to the introduction of approximately 25 g/h/year and 0.01 g/ha/year of Cr and Hg, respectively [49]. Phosphate-based fertilizers have been a significant source of trace metals compared with other inorganic fertilizers, as they may contain 0.1 to 170, 1 to 12, 7 to 38, & 7 to 225 mg/kg of the total Cd, Co, Ni, & Pb, respectively [2, 51]. Phosphate-based fertilizers can relocate naturally available As into agricultural soil systems, which causes it to accumulate in runoff [52]. Agrarian soil in European countries can accumulate As at a rate of 7.7 g/ha/year, compared with Asian countries such as China, which can accumulate As at a rate of 2.5 g/ha/year through the extensive application of phosphate-based fertilizers [2, 53, 54]. Nevertheless, phosphate fertilization may reduce metal toxicity and mobility, as well as the plants available in polluted paddy soils and other agrarian soils [55]. A field trial study was conducted in metal-contaminated soil with mining tailings in Shaoxing, China, and suggested that the addition of phosphate-based fertilizers could obviously reduce the levels of water-extractable elements such as Zn (11.7 to 75.3%), Pb (22.0 to 81.4%), and Cd (1.5 to 30.7%) [56].

In addition to phosphate-based fertilizers, mixed fertilizers also act as significant sources of trace metal contamination in agrarian soils [57]. In China, the yearly received rates of Cd, Cr, Pb, and Ni from phosphate-based mixed fertilizers are 0.73, 28.1, 6.9, and 4.1 g/ha, respectively [53]. In China, it has been reported that approximately 1,200 tons of Zn and 5,000 tons of Cu-containing fertilizers were added to agrarian lands per annum [53]. Agrarian soil contamination by trace metals through fertilizer application has been found in several countries, such as China, India, Malaysia, Bangladesh, Brazil, and certain European countries [2, 58, 59].

Nutritionally rich wastewater provides crops with essential as well as toxic trace metals and accumulates in agrarian soil with organic matter [60]. Despite its inherent quality and shortcomings, the application of wastewater for irrigation purposes has been widely practised for many decades worldwide [61]. Primarily, in arid zones around the world, where water is a limiting factor for agrarian growth, urban wastewater is often used as a

consistent source of irrigation for crop cultivation [62, 63]. The wastewater is usually applied directly for irrigation and circumvents the essential requirements for wastewater treatment plants; the accumulation of severely toxic trace elements in agrarian soils through consistent use of wastewater has often been observed in different countries [64]. It has been estimated that approximately 20 million ha of land is irrigated with wastewater in several countries, including China, India, Bangladesh, Pakistan, Germany, and France [2, 65-67]. Depending on the primary source, the wastewater might include different trace metals, such as As, Zn, Cd, Cu, Pb, Cr, Mn, or Ni, in varying concentrations [68]. However, it has also been reported that after going through wastewater treatment plants, trace metals can persist in wastewater and can accumulate in living systems through food chains[69].

Hence, before the reuse of wastewater for agrarian purposes, significant consideration is necessary for public health as well as acceptance [70]. In an eastern suburb of Beijing, China, a study reported elevated concentrations of trace metals in wastewater-irrigated soils to be in the order of Cr (78.3) > Zn (71.5) > Ni (31.8) > Cu (25.5) > Cd (0.81) mg/kg; however, in wastewater-irrigated agrarian topsoils compared with Chinese reference soils, Cr (53.8) > Zn (51.9) > Ni (18.7) > Cu (16.0) > Cd (0.36) mg/kg, which are within the World Health Organization (WHO) and the national standards of China, except for Cd [2, 71]. A similar type of study was carried out in the Zhangshi wastewater irrigation area in a suburb of Shenyang City, China, in an agrarian area with about three decades of irrigation history. The elevated trace metal-containing wastewater and its measured concentrations were found to be in the order Ni (241.6) > Pb (88.1) > Cu (69.2) Cd (2.7) mg/kg; the levels were significantly elevated in wastewater-irrigated topsoil compared with those in reference soils [72].

A study conducted at the Bindal River, Dehradun, India found that wastewater irrigation caused a higher percent concentration of trace metals in agrarian soil, in the order Zn (48%) > Pb (20%) > Ni (13%) = Cu (13%) > Cr (5%) > Cd (1%), which exceeded the permissible limits defined by the WHO and by India [73]. Though the agrarian soil was irrigated with treated wastewater in Varanasi, India, the concentrations of trace metals were found in the following order: Pb (123.5) > Zn (122.3) Cu > (77.9) > Cr (56.3) > Cd (3.9) mg/kg; these levels exceed the limits of both the WHO and India [74]. A study conducted in Abbottabad, Pakistan, reported on the status of trace metal concentrations in municipal wastewater used

for agrarian purposes, including Cd (2.8 mg/kg), Cr (4.2), Cu (28), Ni (7.5), Zn (135.6), Pb (15.6) and Fe (270) mg/kg, which are below the standards defined by the USA, UK and EU [75]. However, a study conducted in Hanoi, Vietnam, in a paddy soil system found that the irrigation wastewater was contaminated with wastes from several industrial plants and reported the average concentrations of trace metals, such as Cu (202), Zn (192), Pb (159), and Cd (4) mg/kg, which all exceeded the WHO and Vietnamese defined standards [76]. Sewage sludge or biosolids are mainly reliable organic products produced by wastewater treatment and can be constructively recycled [77].

Different waste disposal approaches, such as agricultural application, landfilling, incineration, and ocean dumping, are used based on resource accessibility [78]. Agricultural practices that use sewage sludge are widely known as an economically feasible and ecologically acceptable method of disposal [79]. Therefore, an extensive quantity of sewage sludge is directly deposited on agrarian lands in China, where approximately 10% of the total sewage sludge produced in the country is applied directly to agricultural land [80]. From an agrarian perspective, the direct use of sewage sludge is considered to be an excellent fertilization method because it can improve the physiochemical and biological properties of the soil and contains high levels of nitrogen (N), phosphorus (P), potassium (K), calcium (Ca), and other trace elements that are essential for crop growth [81, 82]. Nevertheless, elevated concentrations of trace metals such as Cu, Zn, Cr As, Se, Cd, Mn, and Pb, are often reported in sewage sludge and may cause adverse effects on humans and the environment [83, 84]. Trace metal contamination in agrarian soils from the application of sewage sludge has been reported as one of the primary concerns about this method because of the long-term accumulation of trace metals in the soil-plant system [85]. Obviously, the trace metal content in sewage sludge differs by numerous factors, such as the quality of wastewater and the effectiveness and meticulousness of the treatment process [86]. Usually, the contents of trace elements in sewage sludge can vary from 0.5 to 2% on a dry basis [2]. However, the frequent application of livestock manure in agrarian soil is recognized as a vital source of trace metals in soil crop systems and causes various environmental issues and human health effects [87, 88].

Livestock manure is a valuable source of organic compost that can improve paddy soil fertility as well as the organic matter content [89]. Livestock are also frequently fed with several feed additives to improve growth performance and reduce the risk of human disease [88, 90]. However, feed additives and animal manure are significant sources of trace metals at various concentrations [88, 91]. The continuous application of livestock manure to agrarian soil causes the accumulation of trace metals in the soil crop system and food chain, which seriously threatens the environment, the quality of plant crop production, and human health [92].

2.2.2. Industrial activities

Mining activities provide several socio-economic benefits but are considered some of the most hazardous activities worldwide [2, 93]. Mining activities such as grinding, concentrating minerals, and tailings and mine and mill wastewater disposal are significant and confirmed sources of trace metal contamination in soil crops (Fig. 1), mainly in paddy soil-rice systems [94, 95]. Trace metal contamination in paddy soil-rice systems through mining activities is considered a significant issue worldwide [19]. The spatial distributions of trace metal concentrations adjacent to mining sites depend on geochemical conditions, and mineralization following the release of untreated wastes into the environment may increase trace metal concentrations in agrarian soil and food crop systems [96, 97]. According to the literature, a study conducted near an abandoned copper mine in eastern China suggests that continuous mining practices may increase trace metal contamination in soils, with average metal concentrations of Cu (147), Pb (53.8), Zn (158), and Cd (0.32) mg/kg [98].

A study conducted in paddy soils adjacent to a mining site in the Kočani field, Macedonia, was severely affected by an increased level of trace metals such as As (47.6), Cd (6.4), Cu (99), Pb (983) and Zn (1,245) mg/kg because of the wide dispersal of trace metals from metal mining activities [99]. However, a similar type of study conducted in Vietnam in paddy fields adjacent to chromite mine tailings piles showed trace metal concentrations of Co (375), Ni (5,590), and (5,750) mg/kg [100]. Another study was conducted in Thai Nguyen Province in northern Vietnam in paddy fields 50 metres away from a Pb recycling smelter and reported significantly elevated concentrations of trace

metals, including Pb and Cd, ranging from 1,100 to 7,000 and 0.5 to 1.3 mg/kg, respectively [101].

In Hezhang County in China, smelting practices commonly cause Cr pollution ranged from 71 to 240 mg/kg in agrarian soils [102]. A study around the chromite mining site at Chikkondanahalli in the Nuggihalli schist belt, Karnataka, India, found that the status of trace metals in agrarian soil concentrations was in the order Cr (791) > Ni (168.5) > Co (21.5); these levels exceed permissible soil quality limits and suggest that the elevated levels of these elements are correlated with previous and current uncontrolled mining activities in the study area [94]. Airborne fine-grained fly ash waste, another primary source, contributes to trace metal contamination in environmental matrices [103]. The reduction in petroleum-based sources causes increasing consideration of renewable energy production, primarily through biomass, including wood, straw, and crop burning [104]. However, coal-based thermal power generation plants are still a well-known energy source in various countries, such as India, China, Pakistan, Japan, and Korea [105, 106]. Thus, the production of incinerated airborne fine-grained solid fly ash waste is considered a serious concern worldwide [107]. Substantial amounts of fly ash containing high concentrations of trace metals are produced through the incineration process [107].

Moreover, fly ash neutralizes agrarian soil acidity, improves soil structure and texture, reduces bulk density, and increases soil water and nutrient-holding capabilities, suggesting the rationality of its use in crop production [2]. Therefore, the direct use of fly ash in agricultural lands is a conventional practice that increases trace metal concentrations and bioaccumulation in crops, mainly in paddy soil-rice systems [108]. The direct disposal of hazardous waste has also been reported as a significant contributor to elevated levels of trace metals in soil-crop systems [10]. The distributions of trace metals from hazardous waste disposal sites to adjacent areas depend on various interconnected geochemical activities [109]. The distributions of trace metals to adjacent agricultural soils are usually influenced by soil pH, oxidation-reduction potential (ORP), and the availability of involved agents such as carbonates, sulfates, chlorides, microbial activities, organic, and hazardous waste properties [110, 111].

A study was conducted in Dholpur district, Rajasthan, India, and reported trace metals, including Cr, Cu, Fe, Ni, Pb, and Zn, with elevated concentrations at dumpsites

(34.56±1.05 to 110.75±2.59 mg/kg) [112]. A similar study was conducted adjacent to dumping sites in Allahabad, Uttar Pradesh, India, and proposed that the soil pH was negatively correlated with Ni and Fe, though the contents of Zn were significantly positively correlated with Pb and Cu [113]. However, in the northwest region of Hyderabad, India, several hazardous waste dumping sites exist where approximately 50,000 tons of hazardous/industrial waste is abandoned yearly in landfills, which causes elevated trace metal contamination in adjacent soil, including Cr (127.9), As (51.7) and Pb (206.4) mg/kg [110].

The increase in the global population has increased the use of electronic appliances, which cause 20 to 50 million tons of annual production of e-waste worldwide, which is a serious concern [114, 115]. E-waste can also cause pollution from numerous trace metals. [116]. The reviewed status of trace metal contamination in soil systems due to e-waste in China comprises several study results suggesting Cu and Cd concentrations of thousands of mg/kg beyond the permissible limits of 50 and 0.3 mg/kg for agrarian soils [115].

However, in a study conducted in Taizhou, China, paddy fields adjacent to an e-waste recycling zone contained trace metal concentrations of 61.21, 98.81, 1.19, and 55.81 mg/kg for Cr, Cu, Cd, and Pb, respectively, which were beyond the background values [117]. Another similar study conducted in southeastern Zhejiang Province in China investigated paddy fields near an e-waste recycling areas and found trace metal concentrations of 20.95, 97.90, 298.31, 0.62, and 44.29 mg/kg for Cr, Cu, Zn, Cd, and Pb, respectively, which were far beyond the background values [31].

Fig. 1.

3. Trace element basic chemistry, behaviour in the soil system, and associated risks

Although trace metals play a significant role in crop production, they may reduce crop production because of the risk of bioaccumulation and biomagnification in the food chain [118]. A basic knowledge of trace metal chemistry, transport, environmental fate, and related human health effects is required to understand their speciation and bioavailability as well as the remediation options [10].

The transport and fate of trace metals in soil crop systems mainly depend on the bioavailability, chemical form, and speciation of trace metals [119]. Once in the soil system, trace metals are absorbed through initial reactions that occur in minutes to hours, followed

by slow adsorption reactions that occur in days to years; thus, their conversion and redistribution into numerous chemical forms with varying mobility, toxicity, and bioavailability take place [120]. The dispersal and reactions of trace metals in the soil system are likely controlled by several factors, such as mineral dissolution and precipitation, metal ion exchange capacity, adsorption or desorption, biological mobilization or immobilization, soil-plant availability and uptake size and aqueous complexation (Table 1) [10].

3.1. Arsenic

Arsenic mainly occurs in different compounds such as As_2O_3, and can be recovered by the processing of ores usually containing Pb, Zn, Ag, Cu and Au [121]. There are four natural processes, *i.e.,* reductive dissolution, sulfide oxidation, alkali desorption, and geothermal activities, involved in the release of As to the environment [122]. Two main species of As exist in the terrestrial environment: inorganic pentavalent arsenic, arsenate As(V) under oxidizing (aerobic) conditions, and trivalent arsenite As(III) under reducing (anaerobic) conditions [22, 123]. As(V) is dominant, primarily in the form of arsenate ions (AsO_4^{3-}) in different states, such as arsenic acid (H_3AsO_4), dihydrogen arsenate ($H_2AsO_4^-$), hydrogen arsenate ($HAsO_4^-$) and arsenate ions (AsO_4^{3-}) [10, 124]. Thus, As and As(V) act as chelates and can easily precipitate once cations are present [125]. The As(V) complexes are stable under specific conditions and coprecipitate and absorb Fe-oxyhydroxide under reducing and/or partially reducing conditions. The coprecipitates are immobile under these particular environmental conditions; nevertheless, the mobility of As increases with an increase in pH [126, 127]. However, under anaerobic conditions, As(III) is the dominant species of As, existing as an arsenite ion (AsO_3^{3-}) and its protonated forms, such as arsenious acid (H_3AsO_3), dihydrogen arsenite (H_2AsO^{3-}) and hydrogen arsenite ($HAsO_3^{2-}$) (Table S2) [10]. As(III) can coprecipitate or can adsorb with sulfides (S^{2-}) and has an affinity to bind with other sulfur compounds [128]. As(III) and arsine (AsH_3) usually exist under highly anaerobic conditions [129]. The main routes of human As exposure include drinking water and food [130]. The bioaccumulation of As in living systems (plants & humans) takes place through methylation, which generates highly volatile methylated derivates of AsH_3 compounds such as dimethyl arsine ($HAs(CH_3)_2$) and trimethyl arsine

($HAs(CH_3)_3$) [131]. Arsenic usually exists in anionic forms that do not exhibit an affinity to bind with anions, including chloride (Cl^-) and sulfates (SO_4^{2-}), to form complexes [132].

Arsenic can also exist in organometallic forms, which include methyl arsenic acid ($(CH_3)AsO_2H_2$) and dimethyl arsenic acid ($(CH_3)_2AsO_2H$) [10]. The modes of toxicity for As(III) and As(V) are different [133]. As(III) is sixty times more toxic and carcinogenic to humans than As(V) [134]. As(III) can bind with tissues for a longer period than As(V), which binds with specific groups of proteins that cause As(V) to act biologically against phosphate, distressing the production of ATP synthesis [135, 136]. Long-term exposure to As damages the human cardiovascular, dermal, neurological, hepatic, respiratory, and reproductive systems (Fig. 1) [137].

3.2. Selenium

Selenium exists in different oxidation states that permit the production of organoselenium and selenoamino acid complexes [138]. In the soil system, selenate (SeO_4^{2-}) and selenite (SeO_3^{2-}) are stable for extended periods [139]. Several Se(VI) compounds exist in soil, but only SeO_4^{2-} usually occurs in normal pH ranges, and the most dominant form of Se(IV) below soil pH 8.5 is hydrogen selenite ($HSeO_3^-$) [139]. Selenium is an essential trace element for animals and humans at a certain level; for adults, the recommended dietary allowance (RDA) of Se is 55 µg/day [123, 140, 141], and an excessive amount of Se can cause severe toxicity. Se(IV) is commonly used as a feed additive in different commercial animal diets with a recognized Se dose of 0.5 mg/kg in the whole feed [142]. In humans, Se intake varies across various countries [143].

Overall, Se consumption for adults ranges from 93 to 134 µg/day in North American regions, the optimal Se consumption ranges from 52 to 64 µg/day in Western Europe/France, and low levels of Se consumption range from 30 to 40 µg/day in Eastern European countries/Poland [142]. This metalloid is also known as a cancer chemopreventive compound and is indispensable for proper cell function [141]. Numerous mechanisms have been reported regarding the chemoprotective effects of Se, such as antioxidant protection, reduction in carcinogen metabolism effects, enhancement of the immune surveillance system, and inhibition of the angiogenesis process and cell cycle [144, 145]. Approximately 0.5 to 1 billion people worldwide suffer from Se deficiency [146], which makes them prone to several diseases, such as white muscle and Keshan disease

[147]. Selenium deficiency occurs in humans when Se intake is < 40 µg/d [148, 149] and may cause reduced bone metabolism, growth obstruction, irregularities in thyroid function, reduced fertility, immune system complications and even cancer [148, 150, 151]. Inorganic Se is 40 times more toxic than organic Se [152]. However, higher intake levels of Se > 400 µg/d [149] can lead to severe toxic effects in humans, such as skin lesions, nail and hair loss, nervous disorders, amyotrophic lateral sclerosis, diabetes, and paralytic symptoms leading to death [153, 154].

3.3.Cadmium

Cadmium (Cd) is toxic. This nonessential transition element occurs directly below Zn in the periodic table due its chemical similarity with Zn; in compounds, Cd(II) exists as a divalent ion [10, 155]. This may be part of the reason for Cd toxicity; since Zn is a well-known essential element, the substitution of Cd for Zn can damage several functions of the metabolic process [156]. Cadmium is present as an impurity in different products, such as detergents, phosphate-based fertilizers, and different petroleum-based products, and is also a component of acid rain, which increases soil acidification and thereby increases Cd mobility [157].

Rice is a significant source of human Cd risk, and the basic chemistry of Cd in reducing soils is important [158]. Both Cd and Zn primarily co-occur in sphalerite in robust solutions as CdS and ZnS [159]. As soon as sphalerite is mixed in aerobic soils and sulfur oxidation by microorganisms occurs, soluble Cd and Zn are released into the ground and react with soils [139]. In flooded paddy soils, numerous chemical changes occur that may affect the phytoavailability of Cd and Zn [160]. The reduction of the physicochemical contents of soil increases the concentration of the CdS complex. When flooded soils drain, a decrease in soil physicochemical contents and CdS complex concentrations occurs due to rapid oxidation and increased soil Cd phytoavailability; Zn has little effect on the yield of rice grains that contain a high level of Cd at harvest [139]. Rainfall during the rice grain filling period can slow paddy soil oxidation and reduce Cd accumulation in rice grains, causing variation in the Cd level in rice grains from year to year [161].

Excessive Cd intake may cause several human health effects, such as renal damage caused by proteinuria due to Cd severely affecting the enzymes that reabsorb proteins in kidney tubules [162]. The most remarkable occurrence of Cd toxicity due to excessive

dietary Cd intake happened in the Jinsu River Valley, Fuchu, Japan, where people were afflicted by itai-itai disease [162]. Other health symptoms of Cd exposure include painful osteomalacia and kidney failure [10].

3.4. Lead

Lead (Pb) is a bluish-gray metal that naturally occurs in soil and rocks; it belongs to the IV group in the periodic table with atomic number 82 [163]. Lead is usually found in metal form at concentrations ranging from 10 to 30 mg/kg in the Earth's crust. Pb is considered a chalcophile metal with a strong affinity for S as well as other elements, and is found in forms such as PbS, $PbSO_4$, $PbCO_3$, i.e., galena, lead sulfate, and lead carbonate, respectively [10].

While only an insignificant amount of the total Pb existing in soil is bioavailable, the key pools of Pb in soils are in soil solution [164]. Lead adsorption on the surface of clay-humus particles can be found in compounds, precipitated forms, secondary iron (Fe) and manganese (Mn) oxides, basic earth carbonates, soil humus and silicate matrices [139]. Pb(II) complexes are mainly ionic, including $Pb^{2+}SO_4^{2-}$, and Pb(IV) complexes have a tendency to be covalent, such as tetra lead $Pb(C_2H_5)_4$; a few Pb(IV) compounds such as PbO_2 are considered active oxidants [10].

Previous studies have confirmed that soil solution Pb is the primary source for plant uptake, with a balance occurring among the soil solution and the more 'exchangeable' or 'labile' pools of Pb in the soil medium [139]. In some soil solutions, Pb might be concentrated with calcium carbonate ($CaCO_3$) or phosphate [165]. Elevated concentrations of Pb in the soil lead to the formation of pyromorphite ($Pb_5Cl(PO_4)_3$), which mainly occurs near the plant root system, and the solubility of Pb in soils primarily depends on pH [166]. The increased pH in soil can precipitate Pb in forms such as carbonate, phosphate or hydroxide, and can also lead to the formation of moderately Pb-organic mixtures [166]. Increasing soil acidity leads to increased solubility and mobilization of Pb, usually at a slower rate than the rate of Pb accumulation in the soil organic layer [167].

Several studies have reported that Pb adsorption significantly depends on the types of ligands involved in the formation of hydroxyl compounds of Pb, such as ($PbOH^+$) & $Pb_4(OH_4)^{4+}$ [166, 168]. However, it has been further suggested that the adsorption of these complexes on montmorillonite might be an interrupted cation exchange process, unlike that

on kaolinite and illite [166]. In soil solution, the higher sorption of Pb *via* manganese oxide, iron oxide, halloysite, and imogolite by kaolinite, montmorillonite, and humus was observed [166, 169]. High Pb intake can cause serious health effects, especially in children who are more vulnerable to Pb toxicity, leading to lower IQ, mental deterioration, impaired development, reduced attention span, and hyperactivity [170]. In adults, Pb exposure can cause memory loss, joint weakness, damage to the cardiovascular system, and other negative effects [171].

3.5. Zinc

In soil systems, the mobility of Zn depends on the solubility of its speciated form and on the soil properties, including pH, cation exchange capacity, redox potential and the availability of metal species in the soil; under oxidizing conditions, zinc sulfide (ZnS) is the dominant species in soil [172]. However, ZnS is insoluble, and the mobility of Zn is low in oxidizing soils since, at higher soil pH levels, Zn adsorption is reduced, resulting in the release of organic Zn complexes and increased concentrations of chelating agents in the soil system [173].

At high pH levels, the Zn in the soil solution is primarily precipitated as zinc hydroxide $Zn(OH)_2$ and/or zinc carbonate ($ZnCO_3$) [174]. In soil solution, clay and metal oxides have the capacity to sorb Zn and tend to retard its mobility [175]; at pH 4, Zn is more mobile than that at pH 6.5 due to sorption [173]. Goethite, iron hydroxide $Fe(OH)_3$, may also reduce Zn mobility in soil and increase retardation; this effect has also been reported with humic acid [176]. Zinc can disturb the activity of soil organisms, including decreasing microbe and earthworm activity, which delays the breakdown of organic matter [177]. Excessive Zn intake can cause acute and chronic toxic effects on human health, such as nausea, loss of appetite, vomiting, abdominal cramps, headaches, and diarrhoea [178].

3.6. Manganese

The most significant, dominant, and stable forms of Mn primarily persist in rock-forming minerals [179]. The Mn^{2+} cation is considered a substitute for Fe^{2+} and Mg^{2+} in silicates and oxides [179]. In the soil development process, Mn primarily accumulates as oxides and hydroxides; consequently, the total Mn concentration in agricultural soil is substantially higher than that of other elements [180]. Mn-containing vital minerals are mostly ferromagnesian silicate minerals such as chlorite, biotite, and hornblende [181].

During the mineral weathering process, Mn complexes oxidize under atmospheric environments, leading to the rapid release and transformation of Mn into the oxides or hydroxides of secondary minerals that accumulate in the soil system [179]. Manganese oxides have an affinity to coprecipitate with Fe oxides [182]; therefore, Mn oxides accumulate in soil horizons with Fe oxides and hydroxides [183]. Low concentrations of total Mn in soils persist in dissolved forms, primarily adsorbed on mineral surfaces or chelated with organic matter, and these forms contribute to the Mn pool that is available to plants [179]. Mn(II) is the most dominant species of Mn in equilibrated soil solutions on exchange sites on soil surfaces [40]. Mn(III) and Mn(IV) are mainly associated with solid soil phases and exist as oxides such as carbonates and phosphates [179]. Mn^{2+}, the primary plant-available form of Mn, is complexed with organic or inorganic ligands, and its concentrations in the soil system decrease with increasing pH [184]. Under anaerobic conditions, the Mn^{2+} concentration in soil solutions of extremely weathered soils, including oxisols and ultisols, might be a significant cause of agricultural crop toxicity, such as that in rice plant systems [179, 185]. The main inorganic complexes and pairs of ions, such as $MnCO_3$, $MnHPO_4$, $MnSO_4$, $Mn(OH)^+$, and $MnMnCl_2$, in Mn-rich soil solution are readily soluble in the water system and are the most bioavailable [139]. In soil solution, Mn(II) forms complexes with organic materials, including humic acid, organic acid, and amino acid [186].

However, the Mn^{2+} that is firmly bound to fulvic acid remains thoroughly hydrated, and the complex formed between Mn^{2+} and humic substances might be described as an outer-sphere complex where ligands do not directly bond with Mn electrons [179]. Mn-organic ligand complexes can be quite labile and exist in crop plants and microbes [187]. Although Mn plays a vital role in several physiological and biochemical processes in plants and humans, Mn deficiency can severely inhibit plant crop growth and human health [188]. Excessive Mn concentrations are toxic to plants and humans [189]. In humans, Mn toxic effects usually occur in the brain and respiratory tract, and significant symptoms of Mn toxicity include Parkinson's disease, hallucinations, forgetfulness, nerve damage, lung embolisms, and bronchitis [190, 191].

3.7. Nickel

In soil solution, nickel (Ni) occurs as the aqueous nickelous ion Ni^{2+}, which can form complexes with organic/inorganic ligands or with suspended mineral colloids [192]. Organic complexes, rather than different inorganic species such as Ni^{2+}, $NiCO_3$, $HiCO_3^+$, and $NiOH^+$, are typically the most dominant Ni species in soil solution [192].

The behaviour of Ni is also pH-dependent; at low pH, Ni occurs in the form of Ni^{2+}, whereas in neutral to high-pH soil solutions Ni precipitates as stable nickelous hydroxide $Ni(OH)_2$ compounds [10, 193]. Nickel is radially dissolved in acidic soil solutions to form Ni^{3+}, and very in basic soil solutions it forms nickelate compounds ($HNiO_2$) which are soluble in water [10]. Notably, the total Ni content in soils, the significant sources of Ni in soils, including geogenic and anthropogenic sources, and the physicochemical characteristics of soils influence the partitioning of Ni between solution and its solid phase in soil; these factors control Ni mobility and bioavailability [192, 194]. Nickle occurs in crucial minerals, mainly allied with oxides of Mn and Fe and layered silicates, such as chlorite, garnierite and vermiculite [195]. In serpentine soils with a high concentration of Ni and basic pH, Ni can partially bind with carbonates [196]. In solution, Ni has an affinity to rapidly adsorb on the surfaces offered by the soil solid phase such as layered silicates, Fe(Al), Mn-oxides, and organic matter [197], and can form inner or outer-sphere surface complexes [192, 198].

Nickle adsorption by the formation of outer-sphere surface complexes is radially exchangeable because Ni species occur as hydrates and loosely attach to outer-sphere surfaces of the solid soil phase through electrostatic force and by H-bonding between hydration shells and surface oxygen [199]. In contrast, Ni species adsorbed on the inner-sphere surface complexes are partly dehydrated or dehydrated species [192].

Compared with other trace elements, the adsorption affinity of Ni^{2+} ions for soil mineral materials is usually lower than that of Zn^{2+}, Pb^{2+}, and Cu^{2+} [200]. The stoichiometric precipitates, *i.e.,* nickel phosphate $Ni_3(PO_4)_2$ and nickel ferrite ($NiFe_2O_4$), are seldom responsible for controlling the Ni concentration in soil solution, even in severely fertilized and contaminated soils [192]. However, Ni precipitates are mostly detectable on clay mineral surfaces and Al oxides at sorption densities lower than the theoretical monolayer

of Ni sorption [201]. Moreover, these precipitates can form at circumneutral pH with saturated concentrations of Ni in solution *via* the solid phases of Ni [202].

The most important factor affecting the behaviour of Ni in soils appears to be pH, though several characteristics, including the clay content and Fe and Mn oxides in soils, are considered of secondary importance [203]. The mobility of Ni in the soil system increases as the pH and cation exchange capacity decrease [204]. Several other factors, including waterlogging (redox), organic composts, and sewage sludge in the soil, also affect the fate and transport behaviour of Ni in soils [205]. At low levels, Ni is considered an essential element for plants, humans, and animals, but a high intake of Ni leads to serious human and animal health effects and is toxic to plants [206]. The most significant human health problems caused by Ni exposure include chronic bronchitis, lung cancer, nasal sinus damage, and reduced lung function [207].

3.8. Chromium

Chromium (Cr), a transition metal, does not exist naturally in elemental form, only in compounds [163]. The significant natural source of Cr in soil is primarily ore products such as mineral chromite, while anthropogenic sources of Cr include electroplating and Cr-containing industrial waste [208]. Cr(VI) is the main form of Cr found at contaminated sites. Its presence depends on the pH and redox conditions. Cr also occurs in the (+III) oxidation state since Cr(IV) is significantly dominant in shallow aquifers where oxidizing conditions exist [209]. The main Cr species, such as chromate (CrO_4^{2-}) and dichromate ($Cr_2O_7^{2-}$), can be precipitated in the presence of metal cations, including silver, barium, and lead ions (Ag^+, Ba^{2+}& Pb^{2+} respectively) [10].

CrO_4^{2-} and $Cr_2O_7^{2-}$ adsorb on soil surfaces, particularly on aluminium and iron oxides [210]. At low pH < 4, Cr(III) is the more dominant form of Cr, and Cr(+III) may form complexes with ammonia, hydroxide, chloride, fluoride, cyanide, or sulfate (NH_3, OH^-, Cl^-, CN^- or SO_4^{2-}, respectively) and with soluble organic ligands [10]. Cr(VI) is considered a more toxic and more mobile form of Cr. The mobility of Cr(III) is also decreased through adsorption on clay and oxide minerals at low pH < 5, and it is less soluble at pH > 5 due to the formation of the chromium hydroxide ($Cr(OH)_3)_{(s)}$ complex [10, 211].

The mobility of Cr depends on the sorption features of the soil, the clay content in soil, the iron oxide content, and the availability of organic matter [210]. The soluble and

precipitated forms of Cr are transported by surface runoff to surface water bodies [212], while soluble and unadsorbed Cr complexes can leach from the soil to aquifers; the leaching ability of Cr(VI) increases as the soil pH increases [10]. Human health risks associated with elevated Cr intake include respiratory distress, irritation, loss of body weight, and allergic dermatitis [213].

4. Mechanisms of trace element uptake, translocation, and accumulation in paddy soil-rice systems

A range of different trace elements generally exists in soil systems. Paddy plants can acquire the required trace elements from the soil as micronutrients [1]. The uptake, translocation, and accumulation of different trace metals in different paddy plant parts followed the order root > shoot > leaf > husk > grain, and the concentrations of these trace metals in plant parts depend on the exposure level [214]. Trace metals such as Zn and Cu are essential for paddy plant growth and sustenance; however, higher uptake, translocation, and accumulation of essential or nonessential trace metals can lead to toxicity and inhibit the growth and production of paddy rice [215]. The root zone (rhizosphere) of paddy plants and soil microbes play a significant role in trace metal availability as well as uptake and translocation to paddy plant systems [216]. Trace metals can be bound to soil particles or precipitated, which might make a crucial portion of trace metals insoluble or unavailable to paddy plants [216]. Several microbial communities in the rhizosphere of paddy plants can catalyse redox reactions in the soil system that can change the mobility of trace metals and the tendency of paddy plants to take up metals [217]. In metal-rich soils, after the biodegradation or biotransformation of trace metals, plants can accumulate specific trace metals in their tissues (accumulators) or, in the same way, restrict metal uptake into their biomass (excluders) [49, 218]. However, paddy plants are considered strong trace metal accumulators [219].

In paddy plants, the uptake, translocation, and accumulation of essential, toxic and potentially toxic trace elements in their biomass depends on environmental and biogeochemical factors such as the availability of soil microbes; the availability of trace elements, exudates, and chelating agents produced by plants; the pH changes induced by oxidation-reduction reactions; and the availability of transporters and cotransporters embedded in the plant cell plasma membrane [49, 220]. Specific transporter proteins

generally involved in trace metal uptake and translocation, such as adenosine triphosphate proteins (ATPases), produce electrochemical gradients and consume energy [49, 221].

The co- and anti-transporter proteins generally use the electrochemical gradients generated by ATPases, and different ion channel proteins facilitate the normal transport of ions in plant cell systems [49, 222]. Nevertheless, several studies have reported that the co-transportation of cations is a common phenomenon, and potentially toxic trace metals that do not have any known biological or beneficial function, such as As, Cd, and Pb, are mostly transported throughout paddy systems *via* transportation systems [49]. In the root zone of paddy plants, the immobilization of and controlled access to trace metals might be achieved through root exudates [223] that chelate trace metals to prevent their uptake into root cells, such as Ni chelation; exudates can also bind Cu or Zn in the apoplast [49]. Therefore, the chemical properties of the rhizosphere and the cell walls in the roots may control trace metal uptake, translocation, and accumulation in paddy plants [224]. Transporter proteins in the plasma membrane of plant root cells and the tonoplast of the cytoplasm [225] can maintain the physicochemical concentrations of and stress responses to trace metals, contributing significantly to trace metal homeostasis in the plant [49]. In paddy plant systems, approximately 13 putative ZIPs are a significant group of trace metal transporters that are primarily involved in the translocation of divalent cations across plant cell membranes [226]. ZIPs also exist in other plant species as well as in microbes, fungi, and animals [227].

These specific proteins are topologically analogous to the eight transmembrane protein (TP) domains, with the N- and C-termini uncovered to the apoplast, and a histidine-rich cytoplasmic circle skirted *via* transmembrane domains (III) and (IV) is commonly involved with the binding of trace metals [49, 228]. In paddy plant systems, As uptake depends on the As concentration gradient between the source and sink [229]. The As uptake in the plant cell (Fig. 2a) mainly depends on As species such as arsenate (As(V)) and uses different phosphate transporters (Pi) that belong to the PHT1 family because P is chemically similar to As(V) (Table S2) [218].

Arsenite (As(III)) uses silicon (Si) transporters such as Lsi1 (aquaporin NIP2;1) and Lsi2 (an efflux carrier) due to the resemblance between As(III) and Si [230]. As(III) is absorbed by the aquaglycoprotein nodulin-like natural proteins (NIPs) [230]. Under Si deficiency,

the expression of Si influx transporters (Lsi1& Lsi2) increases [231]. The accumulation of Si in plant cells is mostly controlled by the Lsi1 and Lsi2 transporters, which are mostly located at the proximal or distal flanks of epidermal and endodermal cells; these cells help in the transportation of As across the plant's cells and tissues [229]. Several studies have suggested that the reduction of As occurs mainly in root cells before its transport to xylem and the rest of the plant [232]. The main As species, As(III) and As(V), are primarily found in the xylem sap of plants [233]. Of the low concentration of total As absorbed through plant roots, only a very small amount is sequestered in leaf, shoot, and grain vacuoles due to As reduction, and the sequestration mechanisms are similar to those of the roots [230]. Hence, the presence of As(III) and As(V) in phloem is a requirement for its distribution to other parts of the plant [234], while Se translocation in plant shoots, leaves, and grains depends on the rate of transpiration and the rate of xylem loading [150, 235].

Selenium uptake in the plant system is facilitated by transporters (Fig. 2b). Selenite (Se(IV)) and selenate (Se(VI)) are transported through phosphate and sulfate channels [147]. Se(VI) enters the plasma membrane of plant root cells using sulfate transporters [236], and Se(IV) and P compete to cross the plasma membrane of paddy plant root cells using P transporters [237]. Se^{IV} and P compete and enter the plasma membrane of plant root cells by P transporters [237]. The P-induced increase in Se bioavailability most likely occurs through the exchange of Se at sorption sites, therefore increasing Se mobility and uptake in the plant [147]. In plant cells, Se typically accumulates in vacuoles and effluxes with S transporters present in the tonoplast [238, 239]. Selenium, after entering plant cells with the help of the sulfate transporter, is translocated to other parts of the plant, *i.e.,* the shoots, leaves, and grain [240], and is metabolized in plastids through the S integration pathway into selenomethionine (SeMet) or selenocysteine (SeCys); the S chemical analogue with Se can become more methylated and evaporate in a nontoxic form [241].

In paddy plants, the uptake, translocation, and accumulation of potentially toxic trace metals such as Cd from the paddy soil-rice (root cells to shoots) system are generally facilitated by ZIP *(*OsIRT1) transporters [242]. These ZIP transporters are essential for paddy plants, and the plant capacity for critical trace metal accumulation is associated with the level of ZIP expression [243]. In the Cd uptake, translocation, and accumulation mechanism in the paddy soil-rice system, Cd travels from the soil to the above-ground parts

of the rice plants. Cadmium is mainly absorbed through rice root cells from the paddy soil and is translocated to other rice parts (shoots and leaves), finally accumulating in the rice grains (Fig. 2c). In the Cd absorption process, xylem plays a significant role in the transportation of Cd from root cells to shoot cells, and the phloem pathway is used in the translocation of Cd to the rice grains [214]. ZIP transporter-like OsIRT1 and OsIRT2 might show influx activity for Cd^{2+} ions. Therefore, it has been reported that OsIRT transporters play a significant role in Cd uptake through roots in paddy fields [214]. However, NRAMP3 and NRAMP4 transporters and their cotransporters also play a crucial role in Cd transport and homeostasis as well as in the resistance to the toxic effects of Cd in paddy plants [244]. Furthermore, the high concentrations of Cd and Mn in paddy plants are primarily facilitated by NRAMP5 transporters [245].

Additionally, the Fe transporter-like OsNramp1 also functions as Cd influx transporters in the plant plasma membrane. It has been reported that Cd is readily transported in paddy plants from root cells to shoot cells through culms and slants and accumulates in nodes [246]. The ABC transporter family is involved in Pb and Cd transportation from outside the plasma membrane to inside the plasma membrane [242, 247]. Pb^{2+} ions usually occur in paddy soil solution and can enter paddy plants through the translocating water uptake (Fig. 2d). In paddy plants, root cell H+ ATPase and H+ drivers can directly take up Pb.

Furthermore, the transpiration process also plays a significant role in pulling or pushing Pb^{2+} from root cells to shoots through the xylem and from the shoots to leaf cells through vascular flow; Pb translocation to paddy rice grains is still not well defined [214]. ZIP family transporters such as IRT1, ZRT1, and ZRT2 facilitate the uptake, translocation, and accumulation of different cations, such as Zn^{2+}, Mn^{2+}, Cd^{2+}, & Ni^{2+}, in paddy systems (Fig. 2 e, f & g) [248, 249]. Furthermore, the Arabidopsis proteins ZIP1 and ZIP2 transporter-like (AtZIP1 & AtZIP2) are generally involved in the translocation of both Mn and Zn from the root to the shoot. They may also play a significant role in the process of remobilization and distribution at the cellular level [250, 251].

Another family of trace metal transporter-like natural resistance-associated macrophage proteins (NRAMPs) is expressed in paddy plant roots and shoots, and these transporters are involved in the transport of several trace metals, such as Ni^{2+}, Mn^{2+}, Zn^{2+}, Cd^{+2}, Cu^{2+}, & $Co^{2+,}$ across paddy plant cell membranes [49, 252, 253]. However, given the availability

of Cr and its species, such as Cr(III) and Cr(VI), in the paddy soil system, paddy plants have an affinity for translocating and accumulating both, while the mechanism of Cr uptake in the paddy soil-rice system is still unclear. Generally, Cr is taken up by plants *via* specific transporters for the absorption of the essential trace metal ions that are required for plant metabolic activity.

Cr(III) uptake in plant systems occurs through passive mechanisms, and Cr(VI) uptake occurs through active processes, usually *via* phosphate or sulfate transporters (Fig. 2h), due to the structural similarity of Cr(VI) to phosphate and sulfate [254]. However, the uptake, translocation and accumulation mechanisms of trace metals such as U, Tl, Rb, and Sr are still unclear.

Fig. 2.

5. Status of trace element contamination in paddy soil-rice systems and risks to rice-growing countries

Paddy crops show substantial tolerance of trace element toxicity; however, trace metal accumulation in different plant parts, *i.e.,* the roots, shoots, leaves, and grains of paddy plants, in varying concentrations has been reported [255, 256]. From this perspective, field investigations are particularly significant, as the findings of such studies could be efficiently used to measure the status of trace element contamination and accumulation in paddy systems [2].

China is a leading rice-growing country in the world. Trace elements resulting from various human activities contaminate approximately 20 million h in China, which covers 20% of the total agricultural area. Paddy cultivation is concentrated in southern China, which also has a highly developed economy and dispersed common nonferrous trace metals, which may seep with nonferrous trace metals into paddy soil systems [2]. Thus, trace metal contamination in paddy soil-rice systems has become a severe concern in China [3].

A study was conducted on paddy soils from Fenghuang, Shimen, and Xiangtan in Hunan Province, China. The mean value of Cd in the paddy soils was five times higher than the WHO permissible limits for Cd (0.35 mg/kg), whereas the maximum Pb, Ni, and As (48.7, 262 & 45.2 mg/kg) concentrations were higher than the permissible limits (35, 50, & 10, respectively) (Table 2). Paddy soil pollution was mainly associated with anthropogenic

activities such as fertilizer and pesticide application during agricultural production. Consistent with the paddy soil results, the mean value of Cd in rice grains from Fenghuang, Shimen, and Xiangtan (0.28, 0.32, & 0.34 mg/kg, respectively) (Table 3) exceeded the national permissible limits (0.2 mg/kg), whereas the levels of As, Ni, Pb, and Mn contamination in rice grains with the average values of the transfer factors (TFs) for Cd, Cr, As, Ni, Pb, & Mn decreased in the order of Cd > Mn > As > Ni > Cr > Pb. Cd had the highest TF of 0.55 in rice grains, confirming the 81.2% increase in cancer risk in the population of Hunan Province, China, which is mainly attributed to the consumption of Cd-contaminated rice [257].

Recently, a similar study conducted in the industrial and urban areas of Hunan Province reported that the mean concentrations of Cd, Pb, and As in paddy soil systems were 1.08, 52.8, and 19.5 mg/kg, respectively, and 68.9% of rice grain samples exceeded the permissible limit for Cd concentration in rice grains (Fig. 3). The maximum TFs of Pb, As, and Cd from paddy soil to rice grains were reported as 0.023, 0.044, & 4.13, respectively, and the cancer risk of the studied metals in rice grains followed the order Cd > As > Pb; these metals pose a serious health risk to the population [4]. In a study conducted by Zeng et al. (2011), paddy soil and rice samples collected from three major rice-growing areas (Tongxiang, Nanhu, & Xiaoshan) in Zhejiang Province, China, reported that significant correlations between the trace metal contents in paddy soils and rice plants were only observed in Mn and Cu in rice shoots and Zn and Mn in rice grains, concluding that the trace metal concentrations in the paddy soil-rice system were not acceptable [258].

In Zhengzhou City, China, a field experiment conducted by Liu et al. (2007) evaluated the uptake and translocation of As, Cd, Cr, and Pb in parts of paddy plants and found the accumulation of trace metals in rice grains to be comparatively lower than that in roots and shoots. They confirmed that the levels of As, Cd, Cr, and Pb in the edible rice grains were much lower than the Chinese national limits for food; therefore, no threat to human health in terms of trace metal toxicity was involved with consuming the rice [259].

In contrast with this study, Luo et al. (2012) studied the effect of agronomic practices on trace metals in soils and vegetation mainly in the water conservation area along the East Dongjiang River, South China, and found that Cd and Pb concentrations in rice grain were much higher than the Chinese national limits for food; hence, they identified a potential

human health risk mainly associated with the long-term consumption of contaminated rice [260]. According to their study, which covered an area of approximately 13,600 km^2 in Heyuan, Huizhou, Guangzhou, and Dongguan, they further suggested that the average Cd concentrations in paddy soils also exceeded the Chinese national maximum permissible limits [2].

Paddy soils adjacent to a coal mine in Guizhou Province, China, were found to contain higher concentrations of As (83 mg/kg). In rice plants in the same area, As concentrations ranged from 6.3 to 44.7 mg/kg in the roots, from 2.2 to 8.9 mg/kg in the shoots, and from 0.3 to 1.7 mg/kg in rice grains, with an average As concentration of 0.8 mg/kg [261]. The distribution and translocation of Se in a paddy soil-rice system was assessed by Sun et al. (2010) in Enshi, Hubei Province, which is typical of Se-rich areas in China. The reported concentration of Se in paddy soils ranged from 0.5 to 47.7 mg/kg, whereas the Se concentration in the rice bran was 1.94 times higher than that in the polished rice grains. The total Se concentrations in the paddy plant parts were found in the order straw > bran > whole-grain > polished rice > husk and exceeded the Chinese national maximum permissible limits for Se in rice (0.3 mg/kg) [262]. In contrast, in paddy fields near an old in Tan Long, Vietnam, Dang et al. (2008) reported high concentrations of trace elements, such as Cd, ranging from 2.3 to 42.9 mg/kg, and Pb, ranging from 1271 to 3953 mg/kg. In Chi Dao, Vietnam, paddy fields situated less than 20 m away from smelters were found to have total Pb and Cd contents ranging from 2000 to 10000 and 2.5 to 56.5 mg/kg, respectively [101].

Due to the increased population and industrialization, Japanese paddy fields suffer from severe paddy soil contamination by trace metals [2, 263]. In Japan, metal contamination in paddy soils is primarily attributed to wastewater from mines that is used for irrigation purposes and uncontrolled nonferrous emissions from metal refining plants [264]. According to Makino et al. (2010), the total concentrations of trace metals such as Zn, Cr, Ni, Cd, and Pb (99.0, .64.0, 39.0, 0.45, & 29.0 mg/kg, respectively) in the paddy soil system were significantly elevated compared with the natural concentrations (89.0, 58.0, 26.0, 0.3 & 24.0 mg/kg, respectively). Consequently, an elevated level of Cd in the paddy soil-rice system has been frequently reported, and it is recognized that rice is a significant source of human Cd intake in Japan [265, 266].

The Cd concentration in Japanese rice has been found to range from 0.02 to 1.06 mg/kg [267]. The average nutritional consumption of Cd by the Japanese population is approximately 3.0 µg Cd/kg body weight/week, which exceeds the acceptable weekly consumption (2.5 µg Cd/kg body weight/week) defined by the European Food Safety Authority (EFSA) [242].

A study conducted by Tsukada et al. (2007) in Aomori Prefecture, Japan, reported the status of trace element contamination in a paddy soil-rice pf. He used the paddy soil-to-paddy plant TF and suggested that the TFs for all investigated trace elements, excluding Pb, were within two orders of magnitude. According to Yap et al. (2009), a study conducted in Kota Marudu, Malaysia reported that trace metals such as Cd, Cr, Mn, Zn, Cu, & Pb can accumulate in different parts of the paddy plant, furthermore suggesting that the concentrations of investigated trace elements in rice grains were below the maximum permissible limits specified by the Malaysian Food Act and Food Regulations.

In western Bangkok, Thailand, a study conducted by Chinoim and Sinbuathong (2010) examined the status of trace metal (Cr, Cd, Ni, Pb, Zn, & Cu) contamination in paddy soils and reported that the concentrations of the studied trace metals were much lower than the acceptable limits. In contrast, Shraim (2017) studied the status of trace metals in market-purchased Thai rice and reported Cd and Pb concentrations that exceeded acceptable limits. A study conducted by Jo and Koh (2004) on approximately 4047 paddy soil samples collected from different locations in Korea reported that the status of trace metal contamination (Zn, Pb, Cd, & Cu) ranged from 0.1 to 96.7, 0.01 to 66.4, 0.1 to 1.01, and 0.1 to 41.6 mg/kg, respectively.

A recent study conducted by Kwon et al. (2017) in a paddy soil-rice system adjacent to four mining areas (Dalsung mine, Yeongdae mine, Sambo mine, and Munmyung mine) in Korea reported that the average concentrations of Cd, Cu, Pb, Zn, and As in paddy soils and grains were comparatively higher than the worldwide acceptable limits defined by numerous researchers (Table 1 & Table 2). Furthermore, an acceptable daily intake (ADI) of As and Cd in rice grains of up to 50% and 80%, respectively, is recommended by the Joint Food Additive and Contaminants Committee and the World Health Organization (FAO & WHO), respectively. Consequently, the regular consumption of contaminated rice from these mining areas can lead to severe human health problems in the local population.

In Iran, a review on the status of trace element contamination (As, Cr, Pb, Cd, & Ni) in rice grains and an assessment of the associated human health risks noted that the trace metal concentrations were highest in rice grains from northern areas of Iran, primarily Mazandaran & Gillan provinces [268]. Furthermore, minimum and maximum levels for trace metals (As & Ni) were established in Iran, and the ranked order of trace metals by their total hazard quotient (THQ) in the human health risk assessment was As >Cr >Pb >Cd >Ni [268].

In Bangladesh, a study conducted by Ahsan et al. (2009) stated the current scenario for contamination by As and other trace metals in the floodplain agricultural field of the Faridpur and Dhamrai study areas. This study reported that the average As concentration in the Faridpur soil system exceeded the WHO acceptable limits (10 mg/kg) by three times, whereas the average levels of other trace metals (Cr, Co, & Cu) in both study areas also exceeded the WHO acceptable limits (70, 8, & 30 mg/kg, respectively); however, the Se concentration was much lower than the WHO acceptable limit (0.7 mg/kg). Furthermore, in both study areas, the groundwater contaminated with As and other trace metals also led to elevated concentrations of these trace metals in soils because of the predominant use of irrigation with the polluted groundwater [269].

According to a survey across Bangladesh, As and other trace elements in paddy soil systems were elevated in areas where shallow tube wells had operated for a long time [270]. However, microlevel studies in different villages of Chakdaha, Nadia, West Bengal, India, were conducted and confirmed the transference of As from irrigation water to the paddy soil-rice system [271]. The results showed that the As concentration in the irrigation water ranged from 0.1 to 0.8 mg/L, which significantly exceeded the WHO acceptable limit (0.01 mg/L) for drinking and irrigation water; in the paddy soils, the reported As concentration was below the WHO acceptable limit (10 mg/kg). In rice plants, the maximum accumulation in roots ranged from 7.2 to 18.6 mg/kg, and the lowest accumulation, in the rice grains, ranged from 0.25 to 0.7 mg/kg [2, 271]. A recent study conducted by Yadav et al. (2017) in the Fatehabad district, Haryana, India, suggested that trace metals (Cu, Co, & Zn) were abundant in rice grains as essential trace nutrients, and human health risk indices revealed that residents in most of the studied locations were safe. In Shakargarh, Pakistan,

a study conducted by Tariq and Rashid (2012) reported Pb accumulation in rice grains, which may cause several health problems for the residents of the studied location.

Rahman et al. (2014) performed a comparative study of Australia-grown rice with market-purchased rice from India, Bangladesh, Pakistan, Canada, Italy, Thailand, and Vietnam from Asian shops in Sydney. Based on the results for Cu and Ni concentrations in Vietnamese rice and those for Cu, Ni, Cd, Pb, & Cr in Thai rice for sale in Australia, the metal concentrations exceeded those in the Australia-grown rice, whereas the trace metal concentrations in the Pakistani rice on sale in Australia were significantly lower than those in the Australia-grown rice.

Fig. 3.

Table 1

Table 2

6. Trace element accumulation influences in paddy soil-rice systems

Trace element accumulation in paddy systems is determined by several influences, such as the bioavailability of the metals, the physicochemical properties of the paddy soil, and the plant species of interest [272]. According to Zhao et al. (2010), trace element enrichment in paddy plants is significantly correlated with the soil physicochemical properties, such as the soil pH and the soil organic matter (OM). However, the Cu accumulation in paddy plants showed a weak correlation with soil physicochemical properties, particularly the soil pH, and there was no correlation between the accumulation of Cu and OM. This suggests that other unknown factors could influence Cu accumulation in plants. A study conducted in the southern part of the Yangtze delta studied Cu contamination in the paddy soil-rice system, where Cu-rich contaminated wastewater is primarily used for irrigation purposes [273]. Their results found a strong correlation between trace element concentrations in the soil and in rice grains, and the total Cu concentrations in the paddy soil, brown rice, rice hulls, and shoots were 101.2, 15.5, 133.2, and 101.4 mg/kg, respectively. Thus, strong correlations between the Cu concentration in rice shoots and rice hulls and the Cu concentration in the soil were proposed, with values of $r = 0.92$ & 0.83, respectively [273].

According to Tariq and Rashid (2012), the trace element concentrations in paddy soil are significantly correlated with plant uptake but insignificantly correlated with the

physicochemical properties of paddy soils and the plant species. Therefore, through plant rhizosphere processes, trace element uptake is generally governed by the physicochemical properties of paddy soil. According to them, the paddy soil pH was strongly negatively but insignificantly correlated with the paddy soil levels of Co, Pb, and Cr ,with values of r = -0.49, -0.41 & -0.48, respectively, whereas sulfate (SO_4^{2-}) was found in the paddy soils. However, the SO_4^{2-} content of the soil was strongly positively and significantly correlated with the Co, Pb, and Cr concentrations. Furthermore, significant positive correlations between pairs of trace elements in paddy soils, such as Pb and Cr, Cd and Pb, Pb and Co, and Cr and Co, and strong negative correlations between Ni and Cd as well as Pb and Ni were also reported. Nevertheless, within the plant parts or the grain, the relationships between trace elements were reported to be dissimilar from those in the paddy soil.

In Mae Sot, Tak, Thailand, no significant correlation was observed between the Cd concentration in paddy soil and that in rice grains, with r = 0.117 [274]. Additionally, in e-waste recycling areas in Southeast China, no strong significant correlation was observed between trace element concentrations in paddy soils and rice grains; furthermore, the investigators claimed that uptake may significantly contribute to Cd, Zn, & Pb levels, while Cu and Cr levels were primarily influenced by the roots [117]. In Table 3, trace element translocation in the paddy soil-rice system (from the paddy soil to the paddy plant roots, shoots, and rice grains) is expressed *via* translocation factors (TFs) previously described by [2, 275]. The TF from the paddy soil to the grain ($TF_{grain/soil}$) was < 1 for all trace elements listed; nevertheless, a particularly high TF from the paddy soil to the root ($TF_{root/soil}$) is often shown [2].

Trace elements such as Cd, Zn, & Pb and their particle size and mobility might be the significant reason for the maximum foliar and plant root uptake [276]. It has been stated that the particle sizes of vehicle traffic-related elements such as Cd and Pb are usually fine, Cu and Zn are usually medium, and Cr is found in in coarse particles; Zn is more mobile than Cu [2]. It has also been reported that Cd, Zn, and Pb particles emitted from vehicles have a higher leaf penetration capacity than those of Cu and Cr [277]. Upon foliar Cd and Pb absorption, these metals might move to existing paddy plant growth areas, such as rice grains [2]. Thus, studies have suggested that trace element levels in rice grains not only depend on the physicochemical properties and available metal concentrations in the paddy

soil system but also on vehicle emissions, the metal particle size, metal mobility, and metal penetration capability, which are significant factors if rice is grown adjacent to highways [2].

Table 3

7. Trace element-contaminated soil management/remediation methods

Once trace elements are released into the environment, they persist for an extended time period that varies depending on the amount and type of the trace element and the soil [10]. Several soil clean-up processes, such as physical, chemical, phytoremediation, and microbial remediation methods, are widely used in the remediation of trace elements. These methods can be performed in situ (on-site) or ex situ (off-site) (Fig. 4). These bioremediation methods are mostly used in combination for the cost-efficient and effective clean-up of trace element polluted sites [24].

7.1. Physical methods

Physical soil clean-up methods can be further classified into soil replacement, soil isolation, soil vitrification, and electrokinetic methods [24, 278]. In the soil replacement method, the polluted soil is replaced or partially replaced with non-contaminated soil [24]. Before 1984, excavation, ex situ/off-site disposal, and soil replacement were the most common methods used for cleaning up contaminated sites [279]. Soil replacement dilutes the concentration of trace elements and, in turn, increases soil functionality [10]. In this method, the replaced soil is usually treated to remove trace elements or, in some cases, might be dumped at another site. The soil replacement method is generally carried out in one of two ways: digging the soil or importing new (uncontaminated) soil [280]. In the digging method, the contaminated site is deeply excavated, and the trace elements are spread into the deeper places in the site. The trace elements are thereby diluted. The importing method refers to the addition of new, clean soil into the contaminated soil. The new clean soil might be used to covered the site surface or might be mixed in to decrease the levels of trace elements [281, 282].

The soil isolation method refers to separating trace metal-polluted soil from uncontaminated soil; nonetheless, for complete remediation, another auxiliary engineering method is needed [283, 284]. The soil isolation method is usually designed to stop the off-site movement of trace elements by limiting them to within a specified zone [10, 285]. In

soil, the isolation method is generally used to avoid further contamination of groundwater through trace elements after other remediation methods are not economically or physically possible [24]. However, in some cases, the polluted zones can be isolated provisionally to prevent transportation during site evaluation and remediation [279]. In this method, the subsurface barriers/walls are typically applied to restrict polluted water and the soil by limiting the flow of both surface and groundwater in a contaminated site [286].

In soil vitrification, the movement of trace elements inside the soil system is decreased through high-temperature action in highly contaminated sites, which leads to the formation of vitreous substances [287]. In the soil vitrification method, some trace elements such as Hg can be volatilized under high-temperature conditions; therefore, they must be collected for additional disposal and/or further treatment [288]. The soil vitrification method is not considered a conventional trace element remediation method because the soil vitrification technique is relatively effortless to apply compared to other physical clean-up methods. Soil vitrification might be functional for most soils, which are polluted with both inorganic and organic pollutants [279]. Throughout the in situ soil vitrification method, an electrical current is passed through the soil by vertically inserting an array of electrodes into the polluted zone [289].

In contrast, the electrokinetic physical soil clean-up method is cost-effective and extensively used for the removal of trace elements from contaminated soils [290]. The soil electrokinetic process generally operates based on the electric field gradient principle with an appropriate power intensity and is typically applied to two sides of an electrolytic tank comprising saturated polluted soils [291]. Trace elements that are present in the soil system can be separated through electrophoresis, electromigration, or electroseepage, which decreases the contamination level of the contaminated site [287].

7.2. Chemical methods

Chemical-based soil clean-up methods can be further characterized as soil immobilization, soil encapsulation, and soil washing [24]. The soil immobilization method refers to a reduction in trace element mobility, bioavailability, and bioaccessibility in the soil by adding immobilization agents to the polluted soil sites. In soil, trace elements are immobilized by several reactions, such as adsorption, complexation, and precipitation [111]. These immobilization reactions cause the redistribution of trace elements from the

soil solution into solid particles, therefore reducing their movement and bioavailability in polluted soil. Trace element immobilization in the soil is mostly carried out by using inorganic and organic amendments to soils [292]. Common amendments to soils include microbes, phosphates, zeolites, cement, clays, minerals, and organic amendments [279]. Recently, several studies suggested that low-cost industrial residues, including industrial eggshells, red mud, and termitaries, could also be used for the immobilization of trace elements in contaminated soil [279, 293, 294] Currently, biomaterials are extensively used to immobilize trace elements in soils due to their availability and very low cost [295]. Of the biomaterials, the extensive application of biochar has received important consideration for immobilizing trace elements in the soil [296].

The soil encapsulation method differs from the other methods and is mostly used as a process to filter contaminants from soils; the encapsulation stops them from spreading further [286]. Soil encapsulation remediation also involves the mixing of polluted soils with different products, such as concrete, lime, and asphalt. [10, 279]. In the encapsulation method, the toxic element-contaminated soil becomes immobile and therefore stops the spreading of pollution to surrounding resources [297]. Several binding materials are used in the formation of the solid blocks; cement is the ideal material due to its high availability, adaptability, and cost-effectiveness [10, 279].

The soil washing method refers to the removal of trace elements from contaminated soils by using different reagents and extractants that can leach trace elements from the soil. It is done on physically excavated soils ex situ or in situ [10, 298]. Recently, the application of appropriate extractants for the leaching of trace metals from polluted soils has been confirmed as an excellent alternative to some of the old conventional methods for the removal of toxic elements [10, 299]. During the soil washing method, the polluted soil is excavated and mixed with an appropriate extractant solution based on the type of trace elements and soil properties [300].

7.3. Phytoremediation

Phytoremediation is a practical remediation method and can rebuild the natural conditions of the soil to remediate harmful or adverse ecological effects [301]. Phytoremediation methods primarily make use of plants to remove contaminants from soils [302]. The phytoremediation method is comparatively efficient, simple, non-invasive, cost-

effective, and eco-friendly and can provide an enduring solution [303]. Phytoremediation is also known as green remediation, botanoremediation, agroremediation, and/or vegetative remediation and includes methods that generally make use of plants to remediate and revegetate contaminated sites [10]. The term phytoremediation includes several methods and applications that differ significantly in process or mechanisms with plants that might immobilize, remove/clean-up, or degrade toxic trace elements [304]. Based on diverse plant uptake patterns, phytoremediation can include phytostabilization, photoevaporation, and phytoextraction [305]. During phytovolatilization, toxic trace elements are taken up from the soil and transformed into less-toxic vapours and then released into the atmosphere *via* the plant transpiration process [306].

During the phytovolatilization process, elements are first combined into volatile organic complexes and then released to the ambient environment as biomolecules [307]. Some trace elements, including As, Se, and Hg, may occur as gases in the environment. Several plant species, such as *Brassica juncea*, *Arabidopsis thaliana*, and *Chara canescens,* have the capability to take up these toxic elements from soils and change them into their gaseous forms inside the plant tissues and then release them into the ambient environment. Arsenic and its species As(V) and As(III) were effectively volatilized from the leaves of *Pteris vittata* plants. *Arabidopsis thaliana* and *Brassica juncea* can be cultivated under elevated Se levels and are proficient in volatilizing Se [279].

Phytostabilization is well defined as the use of plants to decrease the availability and movement of toxic elements in soils because of their stabilization by plants [308]. Phytostabilization may not reduce the levels of toxic elements that exist in polluted soil but prevents their off-site mobilization [309]. The primary purpose of phytostabilization is to limit trace elements in the vadose zone of plants by accumulation through roots and/or precipitation inside the rhizosphere zone [310, 311]. Thus, of the different phytoremediation methods, phytostabilization does not remediate or remove toxic elements from soli but decreases the mobilization of toxic elements to adjacent media/sites [309]. In addition, phytostabilization may also be applied at contaminated sites where practical/controlling limits interfere with the selection and application of the most suitable clean-up methods [312]. Several plant species, for instance, *Agrostis* spp.*,* and *Festuca* spp.

are widely applied for the phytostabilization of Cu-, Pb-, and Zn-contaminated soils in Europe [313].

Phytoextraction is an efficient method for the removal of toxic elements from contaminated soils through plant uptake mechanisms [314]. This solar-driven method is generally based on the ability of plant roots to take up and translocate levels of toxic elements from the soil to the above-ground plant parts [315]. Therefore, phytoextraction reduces the contaminated soil mass [316] and efficiently translocates toxic elements from the soil to aerial plant parts [317]. Compared with contaminated soil, plant biomass is generally easy to recycle, transfer, and treat/oxidize, and this method ensures the a permanent removal of toxic elements from soils [309]. Nevertheless, the phytoextraction method is more convenient for those sites that are contaminated by low-to-moderate levels of toxic elements because several plant species may not be able to survive in highly contaminated sites [309, 318]. For instance, the phytoextraction method is very efficient for Pb removal with *Brassica juncea* in soils whose Pb concentration is less than 1500 mg/kg [279].

Typically, plant species that are efficiently used for phytoextraction must have 1) high toxic element accumulation ability in their aerial parts, 2) high tolerance to levels of toxic elements, 3) high capability to grow quickly with maximum biomass, and 4) very profuse root systems; so far, no standard has been suggested for hyperaccumulator plants [279]. However, hyperaccumulator plant species can accumulate 100- to 500-fold more toxic elements in their shoot tissues than non-accumulator plants without any effect on yield [279, 319]. Typically, most hyperaccumulator plant species can accumulate or tolerate only a single specific toxic element [320], while fewer plant species have the ability to accumulate and tolerate more than one element; these plants are known as multielement hyperaccumulator plant species [321]. Examples include *Thlaspi caerulescens*, which hyperaccumulates Pb, Zn, Cd, and Ni [315]; *Thlaspi goesingense* and *Thlaspi ochroleucum* can hyperaccumulate Ni and Zn; and *Thlaspi rotundifolium* efficiently hyperaccumulates Pb, Zn, and Ni [279].

Phytoextraction efficiency is increased through genetic modification of plants [322]. It has been suggested that several genes play a crucial role in toxic element uptake, translocation, and sequestration in the vacuole in plant tissues [323]. Primarily, the

application of a gene in specific plant species typically depends on the objective of plant mutation [324]. In phytoextraction processes, enhanced uptake with an increased translocation of elements towards the plant's aerial parts [309] can be accomplished through genetically modified genes that are mainly responsible for enhancing element adsorption in plant root cells, decreasing plant root element sequestration, increasing element xylem loading, and increasing translocation to plant shoots for storage in epidermal and mesophyll cells [279]. To date, numerous key processes have been reported at the molecular level, signifying the possibility of engineering toxic element hyperaccumulation in plant systems [325]. The multifold enhancement in absorption, xylem loading, and translocation to aerial shoots increases the tolerance reported in genetically modified plants compared to that in wild types [279] for some toxic elements, such as As, Zn, Pb, and Cd, in *Arabidopsis thaliana*, *Lactuca sativa*, *Brassica oleracea*, *Nicotiana glauca*, and *N. tabacum*, respectively [326-329].

7.4. Microbial remediation

Microbial remediation primarily refers to the application of microorganisms to promote the precipitation, adsorption, and oxidation-reduction of trace elements in the soil. Under toxic metal stress conditions, soil microbial associations aid in increasing plant growth [330]. Several studies have reported that plant growth-promoting and toxic element resistance and soil microorganisms might also protect plants from element toxicity and/or even increase element uptake in hyperaccumulator plants [331]. Microorganism-based remediation methods for toxic elements involve numerous mechanisms, including bioleaching, biomineralization, biosorption, enzyme-catalysed transformation, intracellular accumulation, and oxidation-reduction reactions [279]. Toxic element-resistant rhizobacteria are able to stimulate plant growth and element accumulation in plants by the production of different chemicals, such as indole acetic acid (IAA) and monocyclopropane-1-carboxylate (ACC) [332]. Furthermore, element-resistant rhizobacteria increase the element tolerance of host plants by inducing thiol complexes, metallothionein, and/or superoxide dismutase [279, 333]. Application of *Burkholderia* sp. (Z-90) increased toxic element clean-up efficiency in the soil by 24%, 31%, 32%, 37%, 44%, and 52% for Cu, Pb, As, Cd, Zn, and Mn, respectively [334]. *Bacillus* sp. (E1S2) significantly enhanced the uptake of Zn (18%) and Cd (43%) by *Sedum plumbizincicola*

[335]. In addition, microbial associations may also cause numerous positive effects for plant growth, including osmotic adjustment, stomatal regulation, and increased mineral uptake [336]. Soil bacterial associations may also increase the mobility and availability of the elements in soils in numerous ways, including decreasing the soil pH, facilitating plant-growth [337], and chelating complexes (organic acids, biosurfactants, and siderophores) through changing the soil oxidation-reduction conditions [279]. Nevertheless, the influence of soil microorganisms on element solubility and plant uptake may not be consistent [338]. Soil microorganisms might increase or decrease element uptake by plants [339]. Indeed, specific microbial methods can increase element solubility and bioaccumulation; however, numerous other methods may cause element immobilization, therefore reducing their phytouptake [279, 331].

Additionally, several other important factors that influence the selection and applicability of the existing soil remediation processes include the total cost involved, time required, efficiency under elevated toxic element contamination, acceptance and availability, long-term effectiveness, and applicability to multi-element contaminated sites; these factors are discussed in detail in Table S3 [279].

Fig. 4.

Table 4

8. Conclusion and future research perspectives

In this review, we summarize the basic chemical behaviours, transport, endpoints, and significant natural and anthropogenic sources of trace elements, including As, Se, Cd, Pb, Zn, Mn, Ni, and Cr, in the soil system as well as their uptake, translocation, and accumulation mechanisms in the paddy soil-rice system. Human health effects are significantly associated with these toxic and potentially toxic metals not only due to their occurrence in the environment but also in their uptake, translocation, and accumulation *via* different transporters, such as As (silicate (Si), phosphate (Pi)), Se (sulfate (ST)), Cr (Pi or ST), Zn (AtZIP1 or NRAMP), Mn (NRAMP5 or AtZIP1), Ni (NRAMP), and Cd (NRAMP5, OsIRT1, or osHMA2), in rice systems.

Moreover, the contamination status of paddy soil-rice systems in major rice-growing countries, including India, Bangladesh, China, Pakistan, Malaysia, Thailand, Japan, and Australia, was presented. Consequently, it is worth comparing the existing research

conclusions on the elevated concentrations of trace elements in rice grain in different countries: Se, As, and Zn (0.02-9.7, 3.1-14, & 2.3-484.9), Cd, and Pb (9.4-178, & 0.06-1529), Cr (41-217), and Ni (61-356) in China, India, Bangladesh, and Australia, respectively.

This review also provides an inventory of the various research studies on trace metal concentrations from multiple identified and unidentified contamination sources, mainly in paddy soil-rice systems in rice-growing countries. In the future, this study will be helpful for paddy rice farmers and decisionmakers at regional, national, and international levels to support various administrative policies. The bio-geochemical conditions associated with paddy farming, such as the frequent alternation between wet and dehydrated environments and the oxygenated-non oxygenated paddy soils, make paddy rice systems favourable for trace metal accumulation.

The various influences associated with the accumulation of these metals in the paddy soil-rice system could be 1) the paddy soil-rice plant characteristics, 2) the various soil physico-chemical indicators associated with uptake, translocation, and accumulation, and 3) the post-crop management practices. These mechanisms are primarily reliant on the cultivar (product class), the environmental conditions, and the administrative policies. Accordingly, multidisciplinary elementary and practical research efforts are essential to mitigate trace metal accumulation in paddy soil-rice systems and prevent toxicity to humans from rice.

The contents of the current review highlight the need for a wide-ranging, extensive, periodic monitoring of the contamination status of paddy soil-rice system regions. Broad awareness of paddy soil contamination and the associated health risks might lead different countries to establish strict legal guidelines and rules for paddy soil contamination and to adopt consistent remediation measures to control trace element sources and translocation in paddy soil-rice systems.

Therefore, the present study will be helpful for comparing the various trace element mitigation measures and for understanding the status of trace element contamination, categorization, and translocation mechanisms in paddy soil-rice systems.

9. Acknowledgments

This work is supported by National Key Research and Development Program of China (No. 2019 YFC1803600, and No. 2017YFD0800302), the National Science Foundation of China-Project of Karst Science Research Center (U1612442), and Science and Technology program of Guizhou Province [Grant No. (2019)5618/2019(2963)/(2019)4428], and UK Royal Academy of Engineering Frontier Follow-up Grant (FF\1920\1\36). ZY thanks UK NERC Fellowship grant (NE/R013349/2). The authors would also like to thank all anonymous reviewers for their constructive comments that significantly improved the quality of the manuscript.

10. Conflict of Interest

The authors declared no conflict of interest.

11. References

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List of figures

Fig. 1. A general overview of natural, anthropogenic sources of trace elements in environmental matrices, and associated human health toxicities.

Fig. 2. The uptake, translocation, and accumulation mechanism of trace elements, (a) arsenic, (b) selenium, (c) cadmium, (d) lead, (e) zinc, (f) manganese (g) nickel, and (h) chromium in paddy soil-rice system.

Fig. 3. Trace element arsenic (As), selenium (Se), cadmium (Cd), lead (Pb), zinc (Zn) manganese (Mn), nickel (Ni), and chromium (Cr) contamination in grains of major rice-growing countries.

Fig. 4. Comparison of soil management/remediation methods for trace elements-contaminated soils.

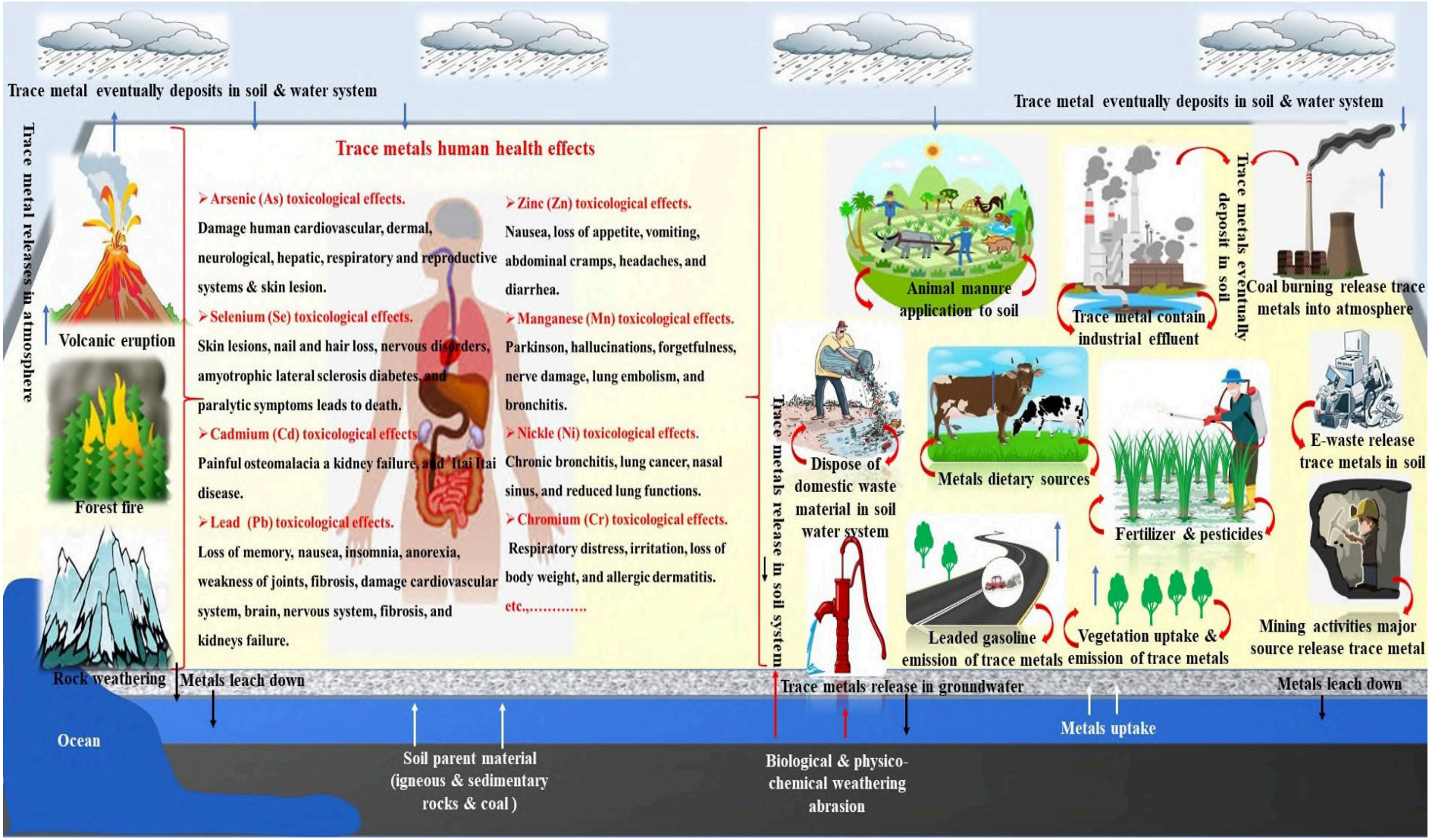

Trace metal eventually deposits in soil & water system
Trace metal eventually deposits in soil & water system
Trace metal releases in atmosphere
Trace metals human health effects
➢ Arsenic (As) toxicological effects.
Damage human cardiovascular, dermal, neurological, hepatic, respiratory and reproductive systems & skin lesion.
➢ Selenium (Se) toxicological effects.
Skin lesions, nail and hair loss, nervous disorders, amyotrophic lateral sclerosis diabetes, and paralytic symptoms leads to death.
➢ Cadmium (Cd) toxicological effects
Painful osteomalacia a kidney failure, and Itai Itai disease.
➢ Lead (Pb) toxicological effects.
Loss of memory, nausea, insomnia, anorexia, weakness of joints, fibrosis, damage cardiovascular system, brain, nervous system, fibrosis, and kidneys failure.
➢ Zinc (Zn) toxicological effects.
Nausea, loss of appetite, vomiting, abdominal cramps, headaches, and diarrhea.
➢ Manganese (Mn) toxicological effects.
Parkinson, hallucinations, forgetfulness, nerve damage, lung embolism, and bronchitis.
➢ Nickle (Ni) toxicological effects.
Chronic bronchitis, lung cancer, nasal sinus, and reduced lung functions.
➢ Chromium (Cr) toxicological effects.
Respiratory distress, irritation, loss of body weight, and allergic dermatitis.
etc.,.....
Volcanic eruption
Forest fire
Rock weathering
Metals leach down
Ocean
Soil parent material (igneous & sedimentary rocks & coal)
Biological & physico-chemical weathering abrasion
Trace metals release in groundwater
Metals uptake
Metals leach down
Animal manure application to soil
Trace metal contain industrial effluent
Trace metals eventually deposit in soil
Coal burning release trace metals into atmosphere
Dispose of domestic waste material in soil water system
Trace metals release in soil system
Metals dietary sources
Fertilizer & pesticides
E-waste release trace metals in soil
Leaded gasoline emission of trace metals
Vegetation uptake & emission of trace metals
Mining activities major source release trace metal

Fig. 1.

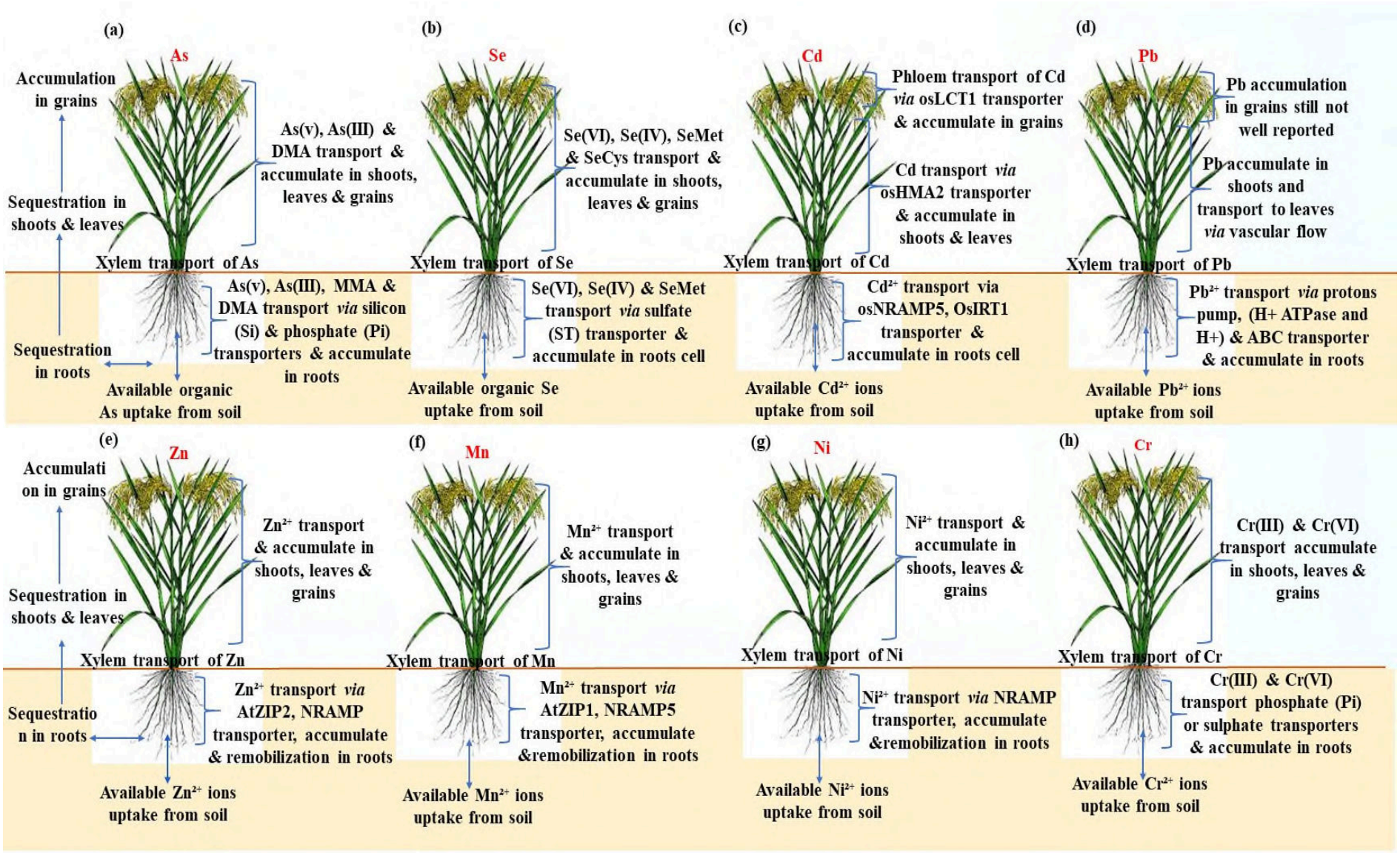

(a)
As
Accumulation in grains
Sequestration in shoots & leaves
Sequestration in roots
As(v), As(III) & DMA transport & accumulate in shoots, leaves & grains
Xylem transport of As
As(v), As(III), MMA & DMA transport via silicon (Si) & phosphate (Pi) transporters & accumulate in roots
Available organic As uptake from soil
(b)
Se
Se(VI), Se(IV), SeMet & SeCys transport & accumulate in shoots, leaves & grains
Xylem transport of Se
Se(VI), Se(IV) & SeMet transport via sulfate (ST) transporter & accumulate in roots cell
Available organic Se uptake from soil
(c)
Cd
Phloem transport of Cd via osLCT1 transporter & accumulate in grains
Cd transport via osHMA2 transporter & accumulate in shoots & leaves
Xylem transport of Cd
Cd2+ transport via osNRAMP5, OsIRT1 transporter & accumulate in roots cell
Available Cd2+ ions uptake from soil
(d)
Pb
Pb accumulation in grains still not well reported
Pb accumulate in shoots and transport to leaves via vascular flow
Xylem transport of Pb
Pb2+ transport via protons pump, (H+ ATPase and H+) & ABC transporter & accumulate in roots
Available Pb2+ ions uptake from soil
(e)
Zn
Accumulation in grains
Sequestration in shoots & leaves
Sequestration in roots
Zn2+ transport & accumulate in shoots, leaves & grains
Xylem transport of Zn
Zn2+ transport via AtZIP2, NRAMP transporter, accumulate & remobilization in roots
Available Zn2+ ions uptake from soil
(f)
Mn
Mn2+ transport & accumulate in shoots, leaves & grains
Xylem transport of Mn
Mn2+ transport via AtZIP1, NRAMP5 transporter, accumulate &remobilization in roots
Available Mn2+ ions uptake from soil
(g)
Ni
Ni2+ transport & accumulate in shoots, leaves & grains
Xylem transport of Ni
Ni2+ transport via NRAMP transporter, accumulate &remobilization in roots
Available Ni2+ ions uptake from soil
(h)
Cr
Cr(III) & Cr(VI) transport accumulate in shoots, leaves & grains
Xylem transport of Cr
Cr(III) & Cr(VI) transport phosphate (Pi) or sulphate transporters & accumulate in roots
Available Cr2+ ions uptake from soil

Fig. 2.

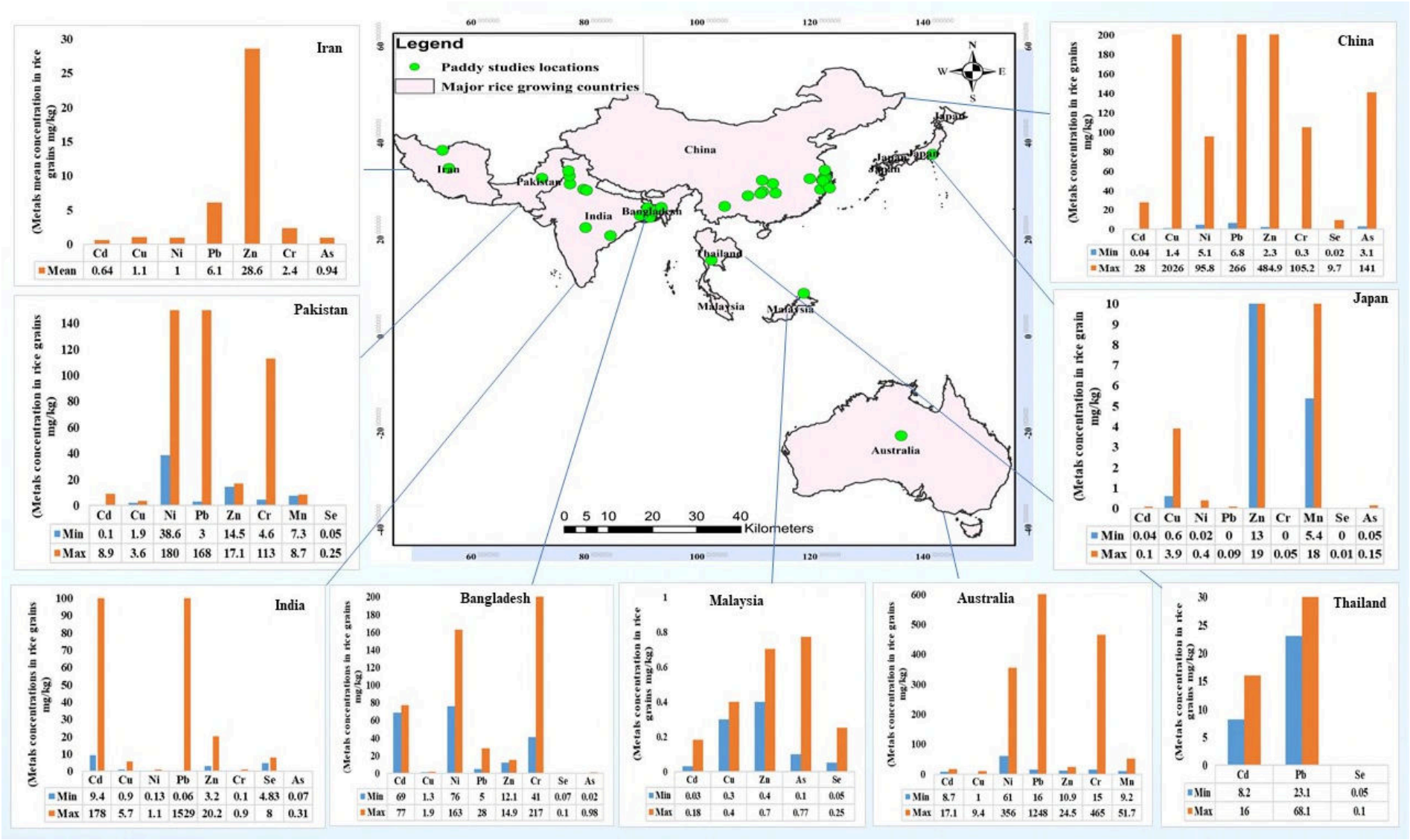

Legend
Paddy studies locations
Major rice growing countries
N
W
E
S
China
Iran
Pakistan
India
Bangladesh
Thailand
Malaysia
Malaysia
Australia
Japan
Japan
Japan
0 5 10 20 30 40
Kilometers
Iran
(Metals mean concentration in rice grains mg/kg)
Cd Cu Ni Pb Zn Cr As
Mean 0.64 1.1 1 6.1 28.6 2.4 0.94
China
(Metals concentration in rice grains mg/kg)
Cd Cu Ni Pb Zn Cr Se As
Min 0.04 1.4 5.1 6.8 2.3 0.3 0.02 3.1
Max 28 2026 95.8 266 484.9 105.2 9.7 141
Pakistan
(Metals concentration in rice grains mg/kg)
Cd Cu Ni Pb Zn Cr Mn Se
Min 0.1 1.9 38.6 3 14.5 4.6 7.3 0.05
Max 8.9 3.6 180 168 17.1 113 8.7 0.25
Japan
(Metals concentration in rice grain mg/kg)
Cd Cu Ni Pb Zn Cr Mn Se As
Min 0.04 0.6 0.02 0 13 0 5.4 0 0.05
Max 0.1 3.9 0.4 0.09 19 0.05 18 0.01 0.15
India
(Metals concentrations in rice grains mg/kg)
Cd Cu Ni Pb Zn Cr Se As
Min 9.4 0.9 0.13 0.06 3.2 0.1 4.83 0.07
Max 178 5.7 1.1 1529 20.2 0.9 8 0.31
Bangladesh
(Metals concentrations in rice grains mg/kg)
Cd Cu Ni Pb Zn Cr Se As
Min 69 1.3 76 5 12.1 41 0.07 0.02
Max 77 1.9 163 28 14.9 217 0.1 0.98
Malaysia
(Metals concentration in rice grains mg/kg)
Cd Cu Zn As Se
Min 0.03 0.3 0.4 0.1 0.05
Max 0.18 0.4 0.7 0.77 0.25
Australia
(Metals concentration in rice grains mg/kg)
Cd Cu Ni Pb Zn Cr Mn
Min 8.7 1 61 16 10.9 15 9.2
Max 17.1 9.4 356 1248 24.5 465 51.7
Thailand
(Metals concentration in rice grains mg/kg)
Cd Pb Se
Min 8.2 23.1 0.05
Max 16 68.1 0.1

Fig. 3.

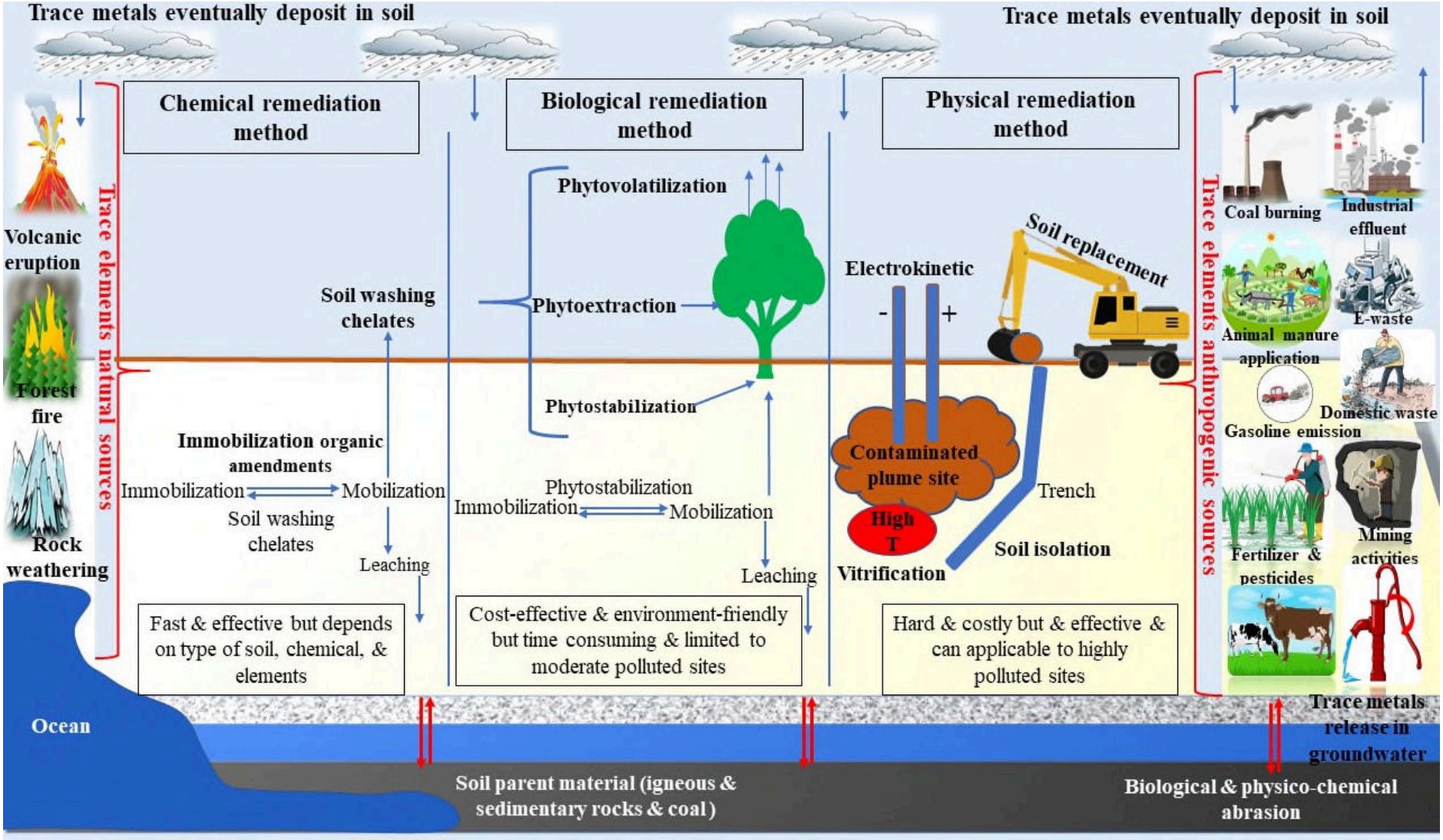

Trace metals eventually deposit in soil
Trace metals eventually deposit in soil
Chemical remediation method
Biological remediation method
Physical remediation method
Trace elements natural sources
Trace elements anthropogenic sources
Volcanic eruption
Forest fire
Rock weathering
Soil washing chelates
Immobilization organic amendments
Immobilization
Mobilization
Soil washing chelates
Leaching
Phytovolatilization
Phytoextraction
Phytostabilization
Phytostabilization
Immobilization
Mobilization
Leaching
Electrokinetic
-
+
Soil replacement
Contaminated plume site
High T
Vitrification
Trench
Soil isolation
Coal burning
Industrial effluent
Animal manure application
E-waste
Gasoline emission
Domestic waste
Fertilizer & pesticides
Mining activities
Fast & effective but depends on type of soil, chemical, & elements
Cost-effective & environment-friendly but time consuming & limited to moderate polluted sites
Hard & costly but & effective & can applicable to highly polluted sites
Ocean
Soil parent material (igneous & sedimentary rocks & coal)
Biological & physico-chemical abrasion
Trace metals release in groundwater

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Table 3 Trace element content and translocation factor (TF) from paddy soil to different parts (roots, shoots, & grains) of the rice plant.

Table 1 Reported trace element concentrations ranges minimum-maximum (mean) mg/kg in paddy soils of different countries.

Country	Locations	Cd	Cu	Ni	Pb	Zn	Cr	Mn	Se	As	Reference
China	Wenling	0.11-3.45 (0.31)	15.8-160.1 (41.1)	9.2-68.2 (33.9)	27.1-140.5 (48.30)	64.97-276.0 (137.0)	-	–	-	-	[3]
	Shengzou	0.04-0.28 (0.1)	15.1-78.1 (29.8)	5.1-62.7 (19.2)	22.0-50.4 (34.3)	34.7-113.6 (75.8)	15.1-78.1 (29.8)	–	-	-	(Zhao et al., 2009
	Nanxun	0.12-0.78 (0.2)	18.8-82.3 (31.1)	18.5-55.6 (32.1)	20.2-51.7 (33.2)	61.5-266.9 (106.8)	–	–	–	–	[340]
	Kunming	0.20-3.57 (1.1)	(45.4-2026 (152.7)	24.1-95.8 (45.0)	6.8-146.6 (42.2)	55.8-484.9 (203.3)	(52.0-105.2 (73.4)	–	–	(10.0 - 141.5 (24.4)	[341]
	Hunan	0.17-27.3 (1.1)	–	–	24.8-266.0 (52.9)	–	–	–	–	(7.3-124.0 (19.5)	[4]
	Nanhu	–	6.6-8.0 (7.3)	–	11.0-12.0 (11.4)	4.7-11.4 (7.7)	0.6-1.1 (0.8)	–	–	–	
	Tongxiang	–	5.26 -9.70 (7.0)	–	9.1-17.8 (12.6)	2.26-6.94 (3.9)	0.5-0.7 (0.6)	–	–	–	[258]
	Xiaoshan	–	1.38-5.66 (3.7)	–	2.0-9.74 (5.1)	1.40-12.2 (5.7)	0.3 -0.8 (0.6)	–	–	–	
	Shimen	0.06-0.4 (0.2)	–	26.6-48.7 (36.2)	19.8-32.3 (24.9)	–	23.2-33.5 (26.0)	–	–	3.16-45.2 (22.1)	
	Fenghuang	0.04-28.0 (3.3)	–	25.9-44.5 (36.5)	19.6-83.6 (43.3)	–	21.5-28.2 (23.7)	–	–	12.2-34.8 (21.2)	[257]
	Xiangtan	0.18-1.17 (0.5)	–	14.9-40.0 (27.7)	37.7-262.0 (94.5)	–	28.1-39.3 (33.1)	–	–	3.3-6.3 (4.5)	
	Tan Long	2.3-42.9	-	-	1271- 3953	-	-	-	-	-	[342]
	Chi Dao	2.5-56.5	-	-	2000-10000	-	-	-	-	-	[101]
Bangladesh	Faridpur	0.12-0.17 (0.2)	38.2-63.5 (48.4)	42.2-61.0 (48.8)	24.32-34.0 (26.8)	82.7-117.4 (97.2)	73.5-108.1 (86.0)	374.1-575.2 (449.7)	(0.6-1.4 (1.0)	17.4-65.0 (33.1)	[269]
	Dhamrai	0.11-0.22 (0.2)	29.6-33.7 (31.8)	43.8-47.9 (46.1)	23.2-26.6 (24.8)	78.9-125.9 (98.8)	45.6-84.4 (76.1)	471.1-655.2 (553.7)	0.6-1.3 (1.0)	3.1-9.0 (6.1)	
	several locations	0.40	-		4.90	-		-	0.32	3.6-26	[214]

	Narora	0.6-4.6 (2.4)	6.41-153.5 (31.8)	–	2.0-46.0 (21.8)	–	3.9-21.5 (8.1)	3.3-35.5 (21.7)	–	–	[343]
India	several locations	(0.8)	-	-	0.90	-	-	-	0.23-4.52	61-416	[214]
	Fatehabad	-	10.8-37.0 (21.8)	0.04-23.5 (10.4)	7.5-148.2 (54.4)	71.5-146.7 (106.0)	0.1-9.7 (6.5)	–	–	–	[344]
Malaysia	Kota	(0.8)	-	-	-	-	21.10	46.40	-	-	[345]
	several locations	0.2-0.3	-	-	-	-	-	-	0.43	11.00	[214]
Japan	several locations	0.45^{T}		39.0^{T}	29.0^{T}	99.0^{T}	64.0^{T}				[346]
	Aomori	0.16-0.53 (0.37)	18.0-45.0 (29.0)	5.9-18.0 (11.0)	9.2-44.0 (21.0)	63.0-160.0 (110.0)	16.0-80.0 (31.0)	370.0-1500 (700.0)	0.72-1.4 (1.0)	6.7-34.0 (12.0)	[347]
USA	several locations	(0.3)	-	-	(55.0)	-	-	-	-	15-950	[214]
	several locations	(0.1)	-	-	(5.3)	-	-	-	0.1-11	0.78	[262, 348]
Korea	several locations	0.1-1.01 (0.11)	0.1-41.6 (4.7)	-	0.01-66.4 (4.8)	0.1-96.7 (4.5)	-	-	-	-	[349]
	Four mining areas	0.001-12.8 (2.3)	14.7-243.0 (63.5)	-	26.3-885.0 (146.0)	89.7-1600.0 (393.0)	-	-	-	2.8-704.0 (64.4)	[215]
Pakistan	Shakargarh	1.0-78.9 (35.0)	-	60.0-99.7 (85.8)	113.8-2967 (1065)	-	13.8-89.8 (37.8)	-	-	-	[350]
Thailand	Bangkok	0.01-0.07	0.7-3.0	0.4-2.6	0.2-1.4	1.0-14.4	0.1-1.9	-	-	-	[351]

^{T} denotes the total concentration

Table 2 Reported trace element concentrations ranges minimum-maximum (mean) mg/kg in grain in different countries.

Country	Locations	Cd	Cu	Ni	Pb	Zn	Cr	Mn	Se	As	References
China	Wenling	0.1-0.5 (0.1)	0.7-5.8 (3.1)	0.05-1.7 (0.2)	–	11.4-35.4	-	–	-	-	[3]
	Shengzou	0.01-0.2 (0.1)	1.3-4.7 (3.2)	0.1-1.5 (0.4)	0.05-0.2 (0.1)	18.5-27.8 (23.5)	0.1-1.4 (0.2)	–	-	–	[352]
	Zhejiang	0.1-0.112 (0.04)	–	–	0.01-0.2 (0.1)	–	–	–	–	0.1-0.3 (0.1)	[353]
	Nanxu	0.01-0.10 (0.01)	1.1-4.2 (2.5)	0.02-1.4 (0.1)	–	9.45-22.0 (14.3)	–	–	–	–	[340]
	Hunan	0.01-6.2 (0.6)	–	–	1.2-4.8 (0.05)	–	–	–	–	0.01-1.0 (0.1)	[4]
	Nanhu	–	2.9-8.9 (5.5)	–	0.1-0.5 (0.2)	21.1-27.1 (23.2)	0.2-0.4 (0.2)	–	–	–	[258]
	Tongxiang	–	2.4-5.5 (4.1)	–	0.2-0.6 (0.3)	18.1-27.5 (22.6)	0.3-0.3 (0.3)	–	–	–	
	Xiaoshan	–	2.8-6.7 (4.7)	–	0.2-0.5 (0.3)	17.7-25.2 (21.3)	0.1-0.3 (0.2)	–	–	–	
	Shimen	0.01-0.7 (0.3)	–	0.3-1.2 (0.7)	0.01-0.03 (0.02)	–	0.03-0.11 (0.1)	–	–	0.3-1.2 (0.5)	
	Fenghuang	0.01-2.1 (0.3)	–	0.1-1.4 (0.5)	0.003-0.02 (0.01)	–	0.05-0.14 (0.1)	–	–	0.2-0.4 (0.3)	[257]
	Xiangtan	0.01-0.691(0.3)	–	0.1-0.8 (0.5)	0.01-0.10 (0.04)	–	0.1-0.5 (0.2)	–	–	0.1-0.3 (0.2)	
	Kaiyang	-	-	-	-	-	-	-	0.04-0.14 (0.08)	-	[354]
	Yangtze River	-	-	-	-	-	-	-	0.02-0.04 (0.03)	-	[355]
	Enshi	-	-	-	-	-	-	-	(0.08-9.7 (2.73)	-	[262]
	Wanshan	-	-	-		-	-	-	(0.02-0.7) (0.1)	-	[356]

	Rugao	-	-	-	-	-	-	-	0.016-0.40 (0.09)	-	[357]
	Narora	0.1-0.25 (0.1)	0.3-2.2 (1.1)	–	(0.24)	–	(0.03)	(0.23)	–	-	[343]
	Fatehabad	-	0.9-5.7 (1.9)	0.04-0.41 (0.4)	0.06-1.0 (0.6)	5.5-20.2 (11.6)	0.1-0.9 (0.6)	-	-	-	[344]
India	West Bengal	--	2.1-4.6	0.13-1.1	-	3.21–10.3	-	-	-	-	[358]
	Market purchased	9.4-178.0 (27.5)	-	-	29-1529 (173)	-	-	-	-	-	[359]
	Bengal delta	-	--		-	-	-	-	-	0.07-0.31	[360]
	Punjab	-	-	-	-	-	-	-	4.83-8.0	-	[214]
	Market purchased	69-77 (73.0)	1.3-1.9 (1.6)	76-163 (105)	5-28 (19.0)	12.1-14.9 (13.4)	41-217 (119)	13.2-16.0 (14.7)	-	-	[359]
Bangladesh	Market purchased	(0.1)	-	-	(0.3)	-	-	-	-	0.41-0.98	[214]
	Market purchased	-	-	-	(3.19)	-	-	-	-	0.02-0.33	[214]
	Market purchased	-	-	-	(0.7)	-	-		0.07-0.16	-	[214]
Thailand	Market purchase	8.2-16.0 (13.4)	-	-	23.1-68.1 (37.0)	-	-	-	0.05-0.10	-	[361]
Pakistan	Market purchase	0.1-8.9 (4.5)	1.9-3.6 (2.6)	77-180 (118)	3-168 (67.0)	14.5-17.1 (16.2)	48-113 (79.0)	7.3-8.7 (7.8)	0.05-0.25	(0.15)	[359, 361]
	Shakargarh	0.2-1.6 (0.9)	-	38.6-52.6 (46.3)	38.6-55.9 (45.7)	-	4.6-12.2 (7.0)	-	-	-	[350]
Australia	Market purchase	8.7-17.1 (7.5)	1.0-9.4 (2.9)	61-356 (166)	16-1248 (375)	10.9-24.5 (17.1)	15-465 (144.0)	9.2-51.7 (24.4)	-	-	[359]
Malaysia	Kota	0.03-0.18	0.3-0.4	-	-	0.4-0.7	0.2-1.3	0.5-1.5	-	-	[345]
Korea	Four mining areas	0.01-0.98 (0.17)	2.0-29.6 (4.7)	-	0.01-3.34 (0.8)	7.3-38.0 (16.8)	-	-	-	0.10-0.77 (0.25)	[215]
Iran	Several locations	(0.64)	(1.1)	(0.76)	(0.64)	(28.6)	(0.4)	-	-	(0.05)	[362]
	Ghaemshahr	-	-	(1.0)	(6.1)	-	(2.4)	-	-	(0.94)	[363]
USA	Market purchased	(0.02)	-	-	0.01-0.09	-	-	-	(0.02)	0.2-0.7	[214]

	Market purchased	-	-	-	-	-	-	-	(0.32)	0.2-0.34	[214, 354]
	Market purchased	(0.05)	-	-	(0.002)	-	-	-	0.03-0.109	(1.8)	[214]
Japan	Aomori	0.004-0.1 (0.03)	0.6-3.9 (2.1)	0.02-0.4 (0.11)	0.001-0.09 (0.01)	13.0-19.0 (16.0)	0.001-0.05 (0.01)	5.4-18.0 (9.7)	0.001-0.006 (0.003)	0.05-0.15 (0.09)	[347]

Table 3 Trace element content and translocation factor (TF) from paddy soil to different parts (roots, shoots, & grains) of the rice plant.

Metal	Metal level in soil mg/kg	Metal level in root mg/kg	Metal level in shoot mg/kg	Metal level in grains mg/kg	Metals translocation factor (TF) from paddy soil to rice plants parts				References
					Soil to root	Root to shoot	Shoot to grains	Soil to grain	
As	5.42	22.77	0.68	0.06	4.2	0.03	0.09	0.011	[364]
Cd	0.86	6.16	0.62	0.01	7.19	0.1	0.02	0.012	
Cu	23.22	2.09	0.17	0.05	0.09	0.08	0.29	0.002	
Pb	18.65	7.09	1.84	0.52	0.38	0.26	0.28	0.028	
Cd	2.39	16.32	3.35	0.85	6.83	0.21	0.25	0.356	[365]
Cu	59.13	32.67	5.52	2.79	0.55	0.17	0.51	0.047	
Pb	40.91	29.46	4.45	0.84	0.72	0.15	0.19	0.021	
Zn	305.03	80.75	27.58	2.3	0.26	0.34	0.08	0.008	
Cd	35.02	0.85	0.85	0.85	-	-	1.0	0.024	[350]
Pb	1,065	-	48.81	45.75	-	-	0.94	0.043	
Cr	37.8	-	9.17	6.99	-	-	0.76	0.185	
Co	747.2	-	71.58	49.89	-	-	0.7	0.067	
Ni	85.84	-	54.5	46.34	-	-	0.85	0.539	
Cd	0.58	-	1.59	0.48	-	-	0.29	0.001	[260]
Cu	20.4	-	0.3	0.24	-	-	0.8	0.012	
Pb	27.9	-	0.07	0.02	-	-	0.29	0.001	
Ni	20.4	-	0.08	0.09	-	-	1.13	0.004	
Cd	0.17	-	-	0.01	-	-	-	0.059	[366]
Pb	29.6	-	-	0.05	-	-	-	0.002	
Se	11.46	11.82	1.35	1.88	0.96	6.28	0.71	6.09	[151]
Cd	4.64	-	-	1.81	-	-	-	2.5	[367]

Supplementary information

Comprehensive review of the basic chemical behaviours, sources, processes, and endpoints of trace element contamination in paddy soil-rice systems in rice-growing countries

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Supplementary information

Table S1 Compression of recent review papers about trace element contamination in soil-plant and rice system.

Table S2 Trace element arsenic (As), selenium (Se), cadmium (Cd), lead (Pb), Zinc (Zn), manganese (Mn), and chromium (Cr) chemical behaviuors in soil system and key transporters facilitate the uptake, transportation, and accumulations in soil-to-rice system.

Table S3 Comparison of different trace element polluted soil management/remediation methods.

Table S3 Comparison of recent review papers about trace element contamination in soil-plant and rice system.

Topics	Objectives	Gaps	Year	Reference
Heavy Metals in Contaminated Soils: A Review of Sources, Chemistry, Risks and Best Available Strategies for Remediation	Scattered literature is utilized to review the possible sources of contamination, basic chemistry, and the associated environmental and health risks of metals (Pb, Cr, As, Zn, Cd, Cu, Hg, and Ni), which can provide insight into heavy metal speciation, bioavailability, and hence the selection of appropriate remedial options.	The current status of trace elements contamination in major rice-growing countries and the translocation (metals key transporters) mechanisms from soil-to-paddy plants not reported	2011	[1]
A review of soil heavy metal pollution from mines in China: Pollution and health risk assessment	The objectives of this research are 1) to evaluate the soil heavy metal pollution levels of mines in China; 2) to assess the health risks posed by these contaminated soils; 3) and to propose recommendations for the environmental management of mining areas in China.	Explain on the national status of trace element contamination in soil system but the trace metals contamination status in different countries and translocation mechanisms in soil-to-plant not reported.	2012	[2]
Current status of heavy metal contamination in Asia's rice lands	Provide an analytical summary extracted from recent publications on heavy metal contamination in Asia's agricultural lands and subsequent accumulation in different parts of the rice plant	The basic behaviors of trace elements (As, Se, Cd, Pb, Zn, Mn, Ni, and Cr) in soil system and translocation (metals key transporters) mechanisms from soil to paddy plants not reported.	2013	[3]
Role of phosphate fertilizers in heavy metal uptake and detoxification of toxic metals	The main objective of this review is to assess the role of phosphorus in fertilizers, their uptake along with other elements and signaling during P starvation	Trace elements behaviors, current status, translocation, accumulation mechanisms, graphical representation, and trace elements management methods in the soil-rice system not reported.	2013	[4]

Arsenic translocation in rice cultivation and its implication for human health	Explains possible mechanisms involved in As absorption that contaminate the rice plant through the soil and water and mentions studies that have been conducted to minimize the risk for human exposure.	Focus on As metabolism inside rice plant, however, the current statues sources and translocation mechanisms of trace elements (Se, Pb, Cd, Zn, Mn, Ni, and Cr) in the soil-rice system not reported.	2016	[5]
A critical review on effects, tolerance mechanisms and management of cadmium in vegetables	Highlight the Cd toxicity and tolerance in vegetables and the different management options to reduce Cd uptake and toxicity in vegetables	Only focus on Cd toxicity behavior mechanism and management in vegetables. However, the rest of the elements (As, Se, Pb, Zn, Mn, Ni, and Cr) basic behaviors of trace elements in soil-plant, translocation mechanism, and management/remediation not reported.	2017	[6]
An Overview of Selenium Uptake, Metabolism, and Toxicity in Plants	The detailed mechanism of Se metabolism inside the plants and designing effective Se phytoremediation and biofortification strategies.	Focus on Se metabolism inside the plant system, whereas, (As, Pb, Cd, Zn, Mn, Ni, and Cr) the detail mechanisms behaviors sources and current status translocation mechanisms not reported.	2017	[7]
The Journey of Arsenic from Soil to Grain in Rice	Explain As translocation mechanisms from soil-to-rice system and factors affecting As availability at roots level.	Focus on As translocation mechanisms soil-to- rice system. In contrast, the current statues sources and translocation mechanisms of trace elements (Se, Pb, Cd, Zn, Mn, Ni, and Cr) in the soil-rice system not reported.	2017	[8]
A critical review of selenium biogeochemical behavior in soil-plant system with an inference to human health	Critically highlights Se speciation and bioavailability in soil, its soil-plant transfer, compartmentation, essentiality, toxicity, and detoxification in plants, keeping in view the Se-induced human health risks.	Focus on Se biogeochemical behavior in the soil-plant system, but the rest of the elements (As, Pb, Cd, Zn, Mn, Ni, and Cr) basic behaviors of trace elements in	2018	[9]

		the soil-plant system, the status and translocation mechanism not reported.		
A critical prospective analysis of the potential toxicity of trace element regulation limits in soils worldwide: Are they protective concerning health risk assessment? - A review	Precise manner toxic elements regulation limits concerning soil in major countries and organizations around the globe to compare and critique them and applying health risk assessment caused by human exposure to the soil by investigating the scenario that soils have toxic elements levels equal to their maximum legislation limits.	Trace elements behaviors, current status, translocation accumulation mechanisms of elements, graphical representation, and trace elements management methods in the soil-plant system not reported.	2019	[10]
Metal(loid)s (As, Hg, Se, Pb and Cd) in paddy soil: Bioavailability and potential risk to human health	Summarizes mobilization, translocation and speciation mechanism of these metal(loids) in the soil-plant continuum as well as available cost-effective remediation measures.	The Zn, Mn, Ni and Cr status, behaviors translocation mechanisms in paddy-soil rice system not reported.	2019	[11]
A comprehensive review of the basic chemical behaviors, sources, process, and the fate of trace elements contamination in paddy soil-rice system of rice-growing countries	In this review, we amid systematically and comprehensively review and understand the basic behaviors of trace metals (As, Se, Cd, Pb, Zn, Mn, Ni, and Cr) in the soil-system to assess sources, uptake, translocation, accumulation mechanisms *via* different transporters like As (silicate (Si), phosphate (Pi)), Se (sulfate (ST)), Cr (Pi or ST), Zn (AtZIP1or NRAMP), Mn (NRAMP5 or AtZIP1), Ni (NRAMP), and Cd (NRAMP5, OsIRT1, or osHMA2) in rice system., the status of trace elements contamination, and associated risk, in paddy soil-rice systems in rice-growing countries, and polluted soil management/remediation methods.	–	2020	This work

Table S4 Trace element arsenic (As), selenium (Se), cadmium (Cd), lead (Pb), Zinc (Zn), manganese (Mn), and chromium (Cr) chemical behaviour in soil system and key transporters facilitate the uptake, transportation, and accumulations in soil-to-rice system.

Elements	Chemical behaviors and occurrence in soil system	Key transporters uptake and translocated trace elements in soil-to rice system	References
As	Arsenic (As) has four oxidation states (-3, 0, + 3, & +5), which occurs in a different type of minerals such as As_2O_3. Two main species of As such as arsenate ((As)V)) under oxidizing and arsenite (As(III)) dominant under reducing soil conditions. The AsV is dominant in the form of arsenate ion (AsO_4^{3-}) different states such as arsenic acid (H_3AsO_4), dihydrogen arsenate ($H_2AsO_4^-$), hydrogen arsenate ($HAsO_4^-$), and arsenate ion (AsO_4^{3-}).	As(V) uses different phosphate transporter (Pi) that belong to the PHT1 family because P is chemically like As(V). The As(III) uses silicon (Si) transporters like Lsi1 (aquaporin NIP2;1) and Lsi2 (an efflux carrier) due to its resemblance with As(III) and Si.	[1, 5, 12].
Se	Selenium (Se) is a metalloid that shows chemical resemblance with Sulfur (S). The common valance states of Se are (-2, 0, +2, + 4 & +6), selenide ($SeII^-$), selenium (Se^o), thioselenate ($SSeO_3^{2-}$), selenite (SeIV) and selenate (SeVI) respectively. In soil system selenate (SeO_4^{2-}) and selenite (SeO_3^{2-}) stable in soil for extended periods.	Selenite (Se(IV)) and selenate (Se(VI)) transported through phosphate and sulfate channels. The Se(VI) enters in the plasma membrane of plant root cells by sulfate transporters, and Se(IV) and P compete and enter into the plasma membrane of paddy plant root cell by P transporters.	[9, 13-15].
Cd	Cadmium (Cd) is poisonous, and non-essential elements in soils system occur in its compound as divalent ion Cd (II). Cd can form the inner-sphere complexes; at pH < 6.5, mainly bound with organic matter (OM), while at pH > 6.5 to Fe-oxides In soil system. The complexes with OM or inorganic ligands, Furthermore, in soil system significant part of Cd bound with humic acids like cadmium carbonate ($CdCO_3$).	ZIP transporter OsIRTs such as OsIRT1 plays a significant role in Cd uptake through roots in the paddy system while NRAMP5, OsHMA2, and osLCT1 play a crucial role in the transport of Cd in paddies roots shoots and grains respectively.	[1, 11, 16, 17].

Pb	Lead (Pb) is a chalcophile metal that has a strong affinity with S combined with S other elements like (PbS, PbSO4 & PbCO3) galena, lead sulfate, and lead carbonate respectively. In organic-rich soils, Pb bound with humic matter and Fe-oxides. Under reducing soil conditions, PbS precipitates and forms Pb2+ ions at oxidizing soil conditions.	In paddy plants, root cells H+ ATPase and H+ drives can directly uptake Pb. Furthermore, the transpiration process also a significant role to pull pushes Pb2+ from roots cell to shoots by xylem and shoots to leaves cells by the vascular flow.	[1, 11, 18-20].
Zn	Zinc (Zn) under oxidizing conditions zinc sulfide (ZnS) is a dominant species in the soil system. At higher soil pH, reduced Zn adsorption and release of organically Zn-complexed, and increased concentrations of chelating agents in the soil system. Also, at high pH, Zn primarily precipitated as zinc hydroxide $Zn(OH)_2$ and or zinc carbonate ($ZnCO_3$) in soil solution.	ZIP family transporters such as AtZIP2 and NRAMP facilitate the uptake, transportation, and accumulations of Zn in the paddy soil-rice system.	[4, 21-26].
Mn	Manganese (Mn) naturally persist (+2, to +7) oxidation states, and Mn (+2) primarily persists in rock-forming minerals. The Mn-complexes oxidized under atmospheric environments leads Mn released, transformed to oxides or hydroxides of secondary minerals, and accumulated in the soil system.	ZIP family transporters AtZIP1, and NRAMP5 facilitates the uptake, transportation, and accumulations of Mn in paddy soil-rice systems.	[4, 24-28].
Ni	Nickel (Ni) In soil solution, Ni species usually occurs as aqueous nickelous ion Ni2+ form a complex with organic/inorganic ligands or allied with suspended mineral collides. Organic complexes the most dominant Ni species in soil solution instead of different inorganic species, i.e., Ni_2^+, $NiCO_3$, $HiCO_3^+$, and $NiOH^+$.	NRAMP transporters facilitate the uptake, transportation, and accumulations of Ni in the paddy soil-rice system.	[25, 26, 29].
Cr	The Cr(VI) is the main form of Cr found at contaminated sites and depends on pH redox conditions Cr also occurs in (+III) oxidation state since Cr (IV) significantly dominant under oxidizing conditions. The Cr, such as chromate (CrO_4^{2-}) and dichromate ($Cr_2O_7^{2-}$), precipitated in presences of metal cations. The CrO_4^{2-} and $Cr_2O_7^{2-}$ adsorb on soil surfaces, particularly aluminum and iron-oxides.	Cr (III) and Cr (VI), the Pi and S transporters facilitate the uptake, transportation, and accumulations of Cr in the paddy soil-rice systems.	[1, 30-32].

Table S5 Comparison of different trace elements polluted soil management/remediation methods.

Soil management/ remediation methods	Process involved	Significant advantages	limitations	Applicability	Acceptance	Multi-element sites	Time required
Physical methods							
Soil replacement method	Excavating contaminated soil & replacing by non-contaminated soil	Effectively isolate toxic elements from a contaminated site & efficient for severely polluted soils	Large in working volume, very costly, production of severe toxic waste & pose severe effects on soil physicochemical properties	Small-scale but long term	Low: limited to severely polluted soils	Very effective	Comparatively less
Soil isolation method	Isolating contaminated soil from uncontaminated soil by subsurface barriers	Restrict off-site transport of toxic elements & useful for severely polluted soils	Costly, still needs further engineering measures, effectiveness varies with the type of subsurface barrier	Small-scale & short to long-term	Low: limited to severely polluted soils	Very effective	Comparatively less
Vitrification method	Reduction in toxic elements bioavailability by forming various material using high temperature	Easy application & applicable to a variety of pollutants	High cost due to energy requirement	Small-scale but long-term	Very low	Very effective	Comparatively less
Electrokinetic method	Removal of toxic elements from the soil by electrophoresis, & or electro-migration by using DC-voltage	Easy applicable, cost-effective & do not affect physicochemical properties of soil	Requires soil with low permeability & pH needs to be controlled	Small-scale but long-term	Very low	Very effective	Comparatively less

Chemical methods							
Immobilization method	Reduction in elements mobility, bioavailability by using immobilizing amendment, forming stable & immobile complexes *via* adsorption	Fast and easily applicable, cost-effective, & covers a broad spectrum of inorganic pollutants	Temporary solution & continuous monitoring is required	Small to medium & short-term		Useful depends on the type of soil, elements immobilizing amendment	Lees to medium
Soil washing method	Removal of toxic elements from the soil by extractants & forming stable and mobile complexes	Cost-effective, thoroughly clean-up toxic elements & meets specific criteria	Washing extractant cause environmental issue; effectiveness differs with soil & extractant type	Small-scale but long-term	Medium to high	Useful depends on the type of soil, elements & immobilizing amendment	Lees to medium
Phytoremediation							
Phytovolatilization	toxic elements uptake by plants from soil & release in vapor form to the atmosphere	Cos-effective & less disruptive	Restricted to volatile toxic elements, cause other environmental issues, no control after metal release to the atmosphere	Small to medium scale & long-term	Low to medium	No	Very long
Phytostabilization	Use of plants to reduce toxic elements bioavailability & mobility in soils *via* sequestration in plant roots	Cos-effective & less disruptive	A temporary solution, effectiveness differs with soil, plant & element type	Small to medium scale & short-term	Medium	Very low	Very long
Phytoextraction	Use of hyperaccumulator plants to uptake, translocate & concentrate toxic	Cos-effective, less disruptive, & ecofriendly	Effectiveness depends on plant growing conditions, tolerance &	Large-scale & long-term	High	Mostly low except for some plants	Very long

	elements from soil to the aerial harvestable plant parts		bioavailability of elements. Toxic elements accumulator plants are generally very less in number				
Microbial remediation	Use of microorganisms to increases plants uptake capacity	Cost-effective, increases plant growth, elements uptake & translocation	Depend on soil, plant type of element & microorganism	Large-scale & long-term	High	Low but more effective than phytoextraction	Very long but < than phytoextraction

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2020-04-21
Elsevier
https://doi.org/10.1016/j.jhazmat.2020.122720
Ali W, Mao K, Zhang H, et al., (2020) Comprehensive review of the basic chemical behaviours, sources, processes, and endpoints of trace element contamination in paddy soil-rice systems in rice-growing countries. Journal of Hazardous Materials, Volume 397, October 2020, Article number 122720

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