A Systematic Review on Arsenic Bio-Availability in Human and Animals: Special Focus on the Rice–Human System

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Contents

Abbreviations

DMA	Dimethylarsinic acid
NRC	National Research Council
ROL	Radial oxygen loss (ROL)
TIM	TNO GastroIntestinal model
TTC	Trophic transfer coefficient
WHO	World Health Organization
XANES	X-ray absorption near edge structure

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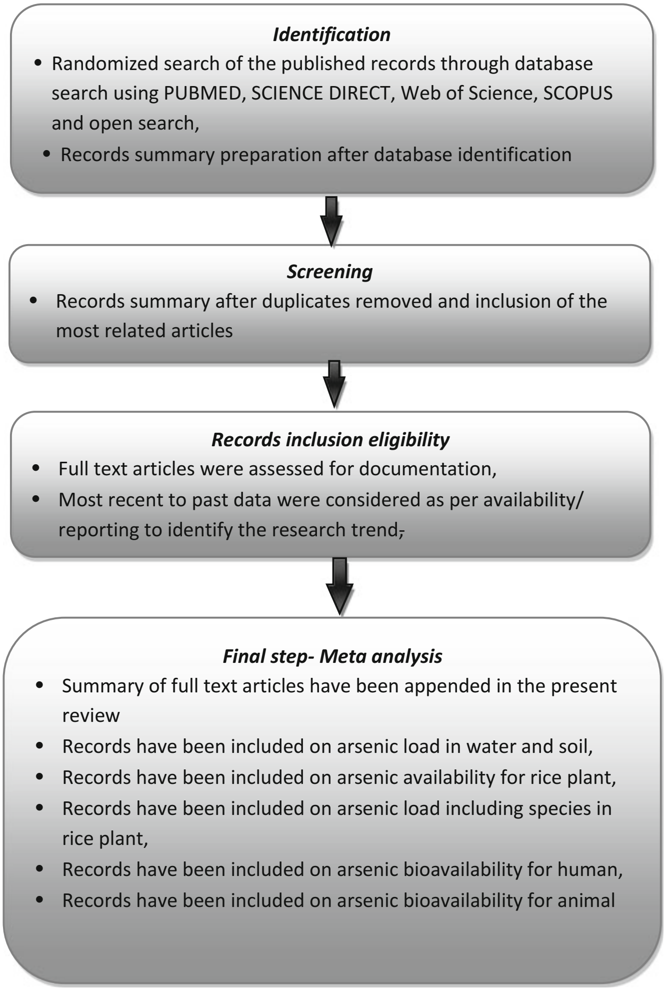
1 Introduction

Arsenic contamination in the environment causes serious concern because arsenic is both toxic and carcinogenic (NRC 1999) and is distributed worldwide, including India, Bangladesh, China, the USA, Romania, Chile, Ghana, and Mexico (Smedley and Kinniburgh 2002). Thus, millions of people are affected by arsenic from food and by drinking arsenic-contaminated water (Ravenscroft et al. 2009). Most of these countries face challenges in addressing the health and economic issues related to the occurrence of arsenic. Arsenic in the environment is mostly of geogenic origin (Acharyya et al. 2000; Mukherjee et al. 2008; Yang et al. 2014), but a large amount is introduced from such anthropogenic activities as industries and mining (Benner 2010). In West Bengal, India, the presence of elevated arsenic in groundwater is a major public health concern in 60% of the districts (Nickson et al. 2000; Chakraborti et al. 2001; Das et al. 2008). The behavior of elemental arsenic in the environment is complex and challenging for risk assessment because the exposure routes are different and include drinking water, contaminated foodstuffs, and atmospheric arsenic (NRC 1999). Moreover, arsenic exists in inorganic (arsenate, arsenite) and organic methylated species, which makes its toxicity complex (NRC 1999; WHO 2001). The World Health Organization (WHO) has set the permissible limit of 10 $\mu g\ l^{-1}$ for drinking water, but the major routes of arsenic exposure through foods were not considered (Zavala and Duxbury 2008). From agricultural fields where irrigation groundwater is arsenic contaminated and the soil is loaded with arsenic, contaminated crop products pass through the food chain (Meharg et al. 2009; Zhao et al. 2010). Rice is the staple food for millions of people worldwide and thus became a rich source of arsenic for humans apart from drinking contaminated water (Roychowdhury et al. 2003, 2008; Williams et al. 2007; Rahman et al. 2009; Su et al. 2010; Biswas et al. 2014a, b; Lin et al. 2015), as rice is typically grown under flooded conditions such that it becomes a typical arsenic accumulator. The other concern about arsenic toxicity is arsenic bio-accumulation because the amount of arsenic bio-accumulated in a human depends on the different food matrix or food type. Rice varieties also differ in bio-accumulative fractions of arsenic. In animals, arsenic is accumulated by ingestion of contaminated foods (Datta et al. 2010; Braeuer et al. 2017; Yang et al. 2018). Arsenic toxicity for both humans and animals depends on bio-availability, which is directly related to the types of foods consumed. Considering these facts, the arena of arsenic toxicity assessment research for humans and animals has changed over the years. At first, researchers reported arsenic contamination in the groundwater and drinking water; then, the focus moved to dietary materials and extended to arsenic bio-accessibility and bio-availability from the different dietary matrices for different consumer groups.

The present systematic review has synthesized the recorded evidence of arsenic bio-availability for humans from rice and other food materials, of arsenic phyto-availability for paddy plants, as rice is the staple diet in most of the arsenic endemic regions of the world, with arsenic bio-availability for animals from their fodder. Because publications on arsenic toxicity and risk assessment issues appear frequently, this review was limited mostly to field-based research results.



Identification

- Randomized search of the published records through database search using PUBMED, SCIENCE DIRECT, Web of Science, SCOPUS and open search,
- Records summary preparation after database identification

Screening

- Records summary after duplicates removed and inclusion of the most related articles

Records inclusion eligibility

- Full text articles were assessed for documentation,
- Most recent to past data were considered as per availability/ reporting to identify the research trend~~,~~

Final step- Meta analysis

- Summary of full text articles have been appended in the present review
- Records have been included on arsenic load in water and soil,
- Records have been included on arsenic availability for rice plant,
- Records have been included on arsenic load including species in rice plant,
- Records have been included on arsenic bioavailability for human,
- Records have been included on arsenic bioavailability for animal

Fig. 1 Review process flowchart and selection criteria of the databases

2 Methods

The review method included such steps as *Identification, Screening, Records inclusion as per eligibility,* and the *Final step-meta analysis* (Fig. 1).

2.1 Search Selection

Systematic searches were made in the often-used academic bibliographic databases such as PUBMED (http://www.ncbi.nlm.nih.gov/pubmed), SCIENCEDIRECT (www.sciencedirect.com/), Web of Science (https://webofknowledge.com/), and SCOPUS (www.scopus.com/), plus literature searches using Google Scholar, hand searches of the literature, and references of the included publications.

For the bibliographic database, our search combined comprehensive English terms representing arsenic occurrence and phyto-availability to human bio-availability with the Boolean operator AND. Before conducting the full search, a pilot search was made with required modifications to meet search criteria. The keywords were searched in each bibliographic database. The searched articles had to meet the following criteria to be included in the review process: *(a) field-based study published in journals was included; (b) study conducted to identify the arsenic flow chain from soil and water to rice to human was considered; (c) studies on the total and arsenic species of the rice grain were included; (d) arsenic accumulation in animals as reported based on field-based studies*; and *(e) in vitro arsenic bio-availability study was considered.* Articles that met the screening test underwent full text review by the author. During these extensive database searches, if any duplicate document was found in different searches, only one document was considered in detail.

2.2 *Documentation, Identification, and Screening*

The searched data were categorized on the basis of *'selected criteria'* to meet the review aim (Table 1). Article titles and keywords were checked thoroughly for this selection. Quality assessment of the selected articles was performed by extending the selection criteria, depth of the data, and exact presentation in the results to discussion section to meet our aim. It is very important to mention here that only field-based research papers were considered as were identified from the methodology

Table 1 Database search criteria and revision

Type of the records identified through database search	Records included after revision including PUBMED, SCIENCE DIRECT, Web of Science, SCOPUS, and open source
Total records identified through database search including all the database and open source	2,128
Summary of full text articles appended in the present review excluding duplicates and considering most relatedness	183
Arsenic load in water and soil	42
Arsenic availability for rice plant	21
Arsenic load including species in rice plant	40
Arsenic bio-availability for human	30
Arsenic bio-availability for animals	38
Papers contain multiple data including the division made here	42

section of any selected articles. This selectivity resulted from the enormous numbers of research and review articles available related to arsenic toxicity, bioavailability, phyto-availability, and rice grain arsenic load, etc., so one limitation was required concerning data acceptability and consideration for this particular review.

3 Results

The results of the present review work have been summarized in different sections as per the available data. As the title indicates, the main focus was arsenic bio-availability to humans from different foods as reported from field-based studies. In the background, the results were supported by reports of arsenic phyto-availability, that is, arsenic uptake and accumulation in different crop plants, where rice received more focus, directly or indirectly, because rice is used worldwide as a staple food. Additionally, arsenic bio-availability in animals has been considered, and the differences and/or similarities of their arsenic bioavailability with humans are discussed.

3.1 Phyto-availability of Arsenic

Here I would like to draw the readers' attention to the fact of that rice is considered primarily in the remainder of this review because rice is known to be a major item of diet (Deb et al. 2013) in most of the arsenic-affected areas worldwide. Arsenic intake and bio-availability from other food materials have also been considered as per research data availability.

3.1.1 Arsenic Uptake in Rice Plants

Chemical processes around the rhizospheric zone may significantly influence and regulate arsenic uptake by paddy plant roots. Radial oxygen loss (ROL) is a well-known phenomenon for paddy rice, resulting in the transfer of oxygen from the aerenchyma into the rhizosphere (Colmer 2003a, b). This ROL can oxidize the ferrous ion to a ferric ion, causing precipitation of iron oxides/hydroxides (FeOOH) around the root surfaces and forming an iron plaque (Smolders and Roelofs 1996). Besides the oxidation capability of the roots, other controlling factors of iron plaque formation are pH, microbial activity, and dissolved ferrous ion concentration in soil solution (Xu et al. 2008; Huang et al. 2012). However, amorphous or crystalline FeOOH is the major component of iron plaque (Hansel et al. 2001; Liu et al. 2006). Iron plaque formation has been frequently reported on the root surfaces of wetland plants including rice (Meharg 2004; Hansel et al. 2001). Several studies have reported less arsenic uptake by rice plants in the presence of iron plaque formation (Liu et al. 2004, 2005; Zhao et al. 2010; Wu et al. 2012). On the other hand, Syu et al. (2013) reported that iron plaque contributed about 73.8–90.4% of the total arsenic uptake from soil as compared to the root (5.8–11.8%) and shoots (2.5–14.3%) in a study to determine the relationship of arsenic sequestration in iron plaque and rice plant accumulation. In contrast, Chen et al. (2005) reported enhanced arsenite uptake in rice plants in the presence of iron plaque formation. Based on element-specific interaction, the addition of sulfur promotes the formation of iron plaque (Hu et al. 2007), whereas phosphate application markedly reduced such formation (Hu et al. 2005). In addition, XANES (X-Ray Absorption Near Edge Structure) analysis showed that arsenate was the dominant species in iron plaque, as was also reported by Liu et al. (2006). A strong co-localization between the root iron plaque and arsenate was also reported by Seyfferth et al. (2010): they applied a combination of techniques, such as X-ray fluorescence imaging, μ-XANES, transmission X-ray microscopy, and tomography, to depict the formation of iron plaque with variable iron coatings influencing the entry of arsenic to the rice roots. Xu et al. (2008) and Zhao et al. (2009) found the dominance of arsenate absorbed on the iron plaque even under flooded conditions. In another study, Syu et al. (2014) compared the effect of iron plaque formation on arsenic uptake in 28 different rice genotypes; and found that 75.7% to 92.8% of arsenic could be sequestered within the iron plaque. In addition, arsenic K-edge XANES spectra indicated the predominance of arsenate in the iron plaque of tested rice genotypes. Some studies have reported the effect of rice genotypes on iron plaque formation (Liu et al. 2004; Lee et al. 2013). Wu et al. (2012) reported that the oxidation capacity of roots controls the formation of iron plaque among different rice genotypes. Pan et al. (2014) investigated the effect of ROL on arsenic sequestration in rice roots; they demonstrated that rice genotypes with higher ROL could sequester a substantial amount of arsenic with more iron plaque formation in the rhizosphere. A lower percentage of arsenite was found in rhizosphere soil solution in rice genotypes with higher ROL from the oxidation of arsenite in the rhizosphere soil. Considering the behavior of iron plaque, Zheng et al. (2012) described that iron plaques act as a buffer in the rhizosphere, especially for uptake of arsenate into rice.

In the soil, the major inorganic arsenic constituents are oxy-anions of arsenite (As^{3+}) and arsenate (As^{5+}), and the presence or occurrence of these arsenic species is governed by pH, redox potential, and the presence of sorbing components (Mandal and Suzuki 2002; Bissen and Frimmel 2003; Haque and Johannesson 2006). For this reason, the adsorption and redox transformation reactions control soil arsenic bioavailability (Hirata et al. 2006; Farooqi et al. 2007).

3.1.2 Arsenic Load in Rice Grains

Arsenic accumulation in rice grains (*Oryza sativa* L.) has been recognized as a disaster (Meharg 2004). In the rural areas of West Bengal, India, and Bangladesh, such groundwater contamination is alarming because the arsenic-contaminated groundwater is used for drinking and irrigation. Thus, arsenic accumulates in rice plant parts such as the root, shoot, and rice husk, including the rice grains (Roychowdhury et al. 2002a, 2008; Norra et al. 2005; Rahman et al. 2007; Biswas et al. 2012; Biswas and Santra 2012), and arsenic accumulation in paddy roots was 28- and 75-fold higher than in shoots and rice grains, respectively (Rahman et al. 2007). When arsenic-contaminated groundwater is used for irrigation, it affects the soil environment and crop quality as well (Meharg and Rahman 2003). Rice straw, that is, the shoot part, can accumulate up to 92.0 mg kg^{-1} arsenic (Abedin et al. 2002a).

Total Arsenic in Rice Grain

Numerous studies have reported wide variability of rice grain arsenic concentration. Paddy grown on arsenic-contaminated soil has been found to have an arsenic load in the rice grain (Abedin et al. 2002a, b; Meharg and Rahman 2003; Jahiruddin et al. 2004; Islam et al. 2004). Based on a field study in Bangladesh, Williams et al. (2006) reported that arsenic concentration in grain varied from 0.04 to 0.92 mg kg^{-1} and from 0.04 to 0.91 mg kg^{-1} for rice growing under Aman and Boro seasons: the names imply the rainy (aman) and the dry (boro) season cultivated rice. Rahman et al. (2006) observed an average concentration of 0.57–0.69 mg kg^{-1} arsenic in raw rice grain from the arsenic-affected area of Bangladesh. Moreover, Meharg and Rahman (2003) reported an arsenic concentration of 1.7 mg kg^{-1} in three rice grain samples from Bangladesh, which exceeds the WHO permissible limit (1.0 mg kg^{-1}). This result is consistent with the findings of Islam et al. (2004) where in accumulation of arsenic up to 2.0 mg kg^{-1} was reported in the Gangetic floodplain of Bangladesh. Large variability in soil arsenic level, in the range 3.1–42.5 mg kg^{-1}, is reflected by elevated grain arsenic in the paddy fields of Bangladesh (Meharg and Rahman 2003). In later studies, higher levels of arsenic in Bangladeshi rice were also reported, as 0.6–0.7 mg kg^{-1} (Rahman et al. 2007), 0.41–0.98 mg kg^{-1} (Sun et al. 2008), and 0.02–0.56 mg kg^{-1} (Rahman et al. 2009), respectively. From the Indian scenario, Roychowdhury et al. (2003) found wide variability in arsenic concentration (from 0.04 to 0.61 mg kg^{-1}) in rice collected from the Murshidabad district, West Bengal. Considerably greater amounts of arsenic were also observed from their further study in the Nadia (0.04–0.39 mg kg^{-1}) and Murshidabad (0.04–0.66 mg kg^{-1}) districts of West Bengal (Roychowdhury et al. 2008). Meharg et al. (2009) reported a grain arsenic concentration range of 0.07–0.31 mg kg^{-1} for Indian rice based on a market basket study. Bhattacharya et al. (2010) observed grain arsenic accumulation of 0.16–0.58 mg kg^{-1} in the Aman and Boro seasons from different arsenic-afflicted areas of Nadia district, West Bengal.

Williams et al. (2007) reported a considerable arsenic concentration (0.15–0.66 mg kg^{-1}) in rice grown in south-central US and in rice grown in California (0.10–0.30 mg kg^{-1}) in the United States. This result was consistent with the previous findings reported by Heitkemper et al. (2001a) and Williams et al. (2005). Zavala and Duxbury (2008) found a wide range (0.005–0.710 mg kg^{-1}) of arsenic in commercially available rice grains in different countries. Their study derived a global normal range (0.08–0.20 mg kg^{-1}) of arsenic concentration in rice grain based on the widely variable dataset. They also evaluated the impact of the arsenic-contaminated environment to arsenic concentration in rice grains; grain arsenic concentration increased by 25–45% for Bangladesh studies, whereas a relatively higher increasing trend (47–94%) was found in rice grains from US studies. In the samples for the Bangladesh study, the high arsenic concentration in the rice grains was attributed to the high arsenic concentration in irrigated water, whereas the high arsenic concentration in the US rice grains was linked with the application of arsenic-containing chemicals in the fields and therefore soil arsenic contamination. The link was previously suggested by Williams et al. (2005, 2007). In a very recent field-based study in arsenic-endemic areas of West Bengal, India, Biswas et al. (2018) have concluded that one unit increase of arsenic concentration in soil and irrigated water results in an average 3.660-fold and 1.345-fold increase, respectively, of the arsenic concentration in paddy plants. On the basis of a market basket survey, Meharg et al. (2009) explored inorganic arsenic exposure of white (polished) grain samples from baseline consumption by analyzing 901 samples of white market rice from ten countries including Asia, Europe, and the US to study the distribution of total and inorganic arsenic in rice depending on geographic variation. In baseline market white rice, mean and median values of total arsenic concentration differed by five- to sixfold from different countries of origin. Zavala and Duxbury (2008) and Torres-Escribano et al. (2008) consistently found mean total arsenic concentrations of 0.20 mg kg^{-1} and 0.21 mg kg^{-1} in the survey of US rice ($n = 112$) and Spanish rice ($n = 24$), respectively. In another interesting report, Rahman et al. (2007) investigated the accumulation and distribution of arsenic in different fractions of rice grain using two widely cultivated rice varieties, namely, BRRI dhan-28 and BRRI hybrid dhan-1. The distribution in different grain fractions was determined in both parboiled and non-parboiled rice: in both varieties, the trend of arsenic concentration in different fractions was rice hull > bran polish > brown rice > raw rice > polished rice. Arsenic concentration in brown rice and polished rice was in the range of 0.3–0.8 mg kg^{-1} and 0.3–0.5 mg kg^{-1}, respectively. They also suggested that arsenic concentration in polished rice was reduced by parboiling of raw rice before milling. Some previous studies reported that milling of raw rice significantly reduces grain arsenic concentration (Duxbury et al. 2003; Rahman et al. 2006): this may decrease the chance of arsenic intake in the human body. Furthermore, Rahman et al. (2006) stated that cooking can reduce arsenic concentration in the rice, whereas cooking raw rice in arsenic-contaminated water could increase arsenic concentration in rice grains by 10–35% (Misbahuddin 2003). For this dilemma, cooked rice is also of concern in terms of arsenic consumption. Cooking influences arsenic retention in rice (Sengupta et al. 2006; Rahman et al. 2006; Signes et al. 2008; Torres-Escribano et al. 2008; Mondal and Polya 2008) and may remove as much as 57% of the total arsenic from rice grains (Sengupta et al. 2006). In contrast, several studies have reported increased arsenic content in rice cooked in arsenic-contaminated water (Misbahuddin 2003; Rahman et al. 2006; Roychowdhury et al. 2008; Signes et al.

2008; Mondal and Polya 2008). Signes et al. (2008) have reported that the cooking process did not change the arsenic speciation in rice.

Rice grain arsenic concentrations vary with the rice cultivars (Alam et al. 2003; Williams et al. 2006; Rahman et al. 2007; Biswas et al. 2013b). Alam et al. (2003) studied 21 fields of Cumilla district, Bangladesh, and concluded that arsenic concentrations change among rice varieties. Williams et al. (2006) observed arsenic concentration variation in two main rice types, *Aman* and *Boro*, as was also observed by Meharg and Rahman (2003), even though it has been reported that a hybrid rice variety has higher root-to-shoot-to-grain translocation of arsenic than the non-hybrid varieties (Rahman et al. 2007). Arsenic in the rice grains is not only harmful to humans, but accumulation of high levels of arsenic in rice straw is a potential threat to cattle that consume the contaminated straw, and thus indirectly to human health, via presumably contaminated bovine meat and milk (Abedin et al. 2002a; Rahman et al. 2008).

Arsenic Species in Rice Grain

Arsenite (As^{3+}) is the dominant arsenic species in rice grain, followed by arsenate (As^{5+}) and dimethylarsinic acid (DMA) (Williams et al. 2006; Meharg et al. 2008; Roychowdhury et al. 2008; Biswas et al. 2013c). Khan et al. (2010a, b) investigated the bio-availability of arsenic to rice by conducting a glasshouse pot experiment with different soil samples varying in the source of arsenic contamination collected from Bangladesh, China, and the UK. The grain total arsenic concentration ranged from 0.24 to 1.09 mg kg^{-1}, whereas inorganic arsenic and DMA accounted for 33–77% and 23–67%, respectively. Rice cultivars grown in Bangladeshi soils were found to have higher grain bioavailability, which could be related to the use of arsenic-laden irrigation water. This range of grain arsenic concentration is consistent with previous reports (Xu et al. 2008; Khan et al. 2009; Li et al. 2009) in which inorganic and organic arsenic species increased simultaneously with increased arsenic load from irrigation water, but DMA was the dominant arsenic species above 0.4 mg kg^{-1} of total arsenic, because a relatively higher percentage of DMA was found in rice grain growing under the greenhouse pot experiment as compared to grain samples collected from the paddy field. This increasing trend of DMA is consistent with the findings of Meharg et al. (2008), Xu et al. (2008), and Norton et al. (2009). In contrast, Li et al. (2009) and Carey et al. (2010) reported much higher translocation efficiency of DMA from root to shoot and from shoot to grain than inorganic arsenic. The study also indicated a strong environmental influence on arsenic speciation in rice grain for two rice cultivars from Bangladesh.

3.1.3 Arsenic Phyto-availability in Crops and Vegetables Other Than Rice

Arsenic concentration in different salad and vegetable crops from a historical mining area in the UK was estimated wherein the authors found elevated arsenic in the edible parts of the vegetables. In beetroot, lettuce, onion, and peas, elevated arsenic was correlated with soil arsenic concentration (Xu and Thornton 1985). Arsenic uptake depended on arsenic concentration in the nutrient solution, and arsenic accumulation in roots increased in the tomato (*Lycopersicon esculentum* Mill., cv. Marmande) plants; increased arsenic decreased plant growth and fruit yields (Carbonell-Barrachina et al. 1995). Bean plants (*Phaseolus vulgaris* L.) were shown (Carbonell-Barrachina et al. 1997) as a good arsenic accumulator from applied arsenic source, as half the absorbed arsenic was transported to the upper parts of the plant, although the fruits, the main vegetable, was arsenic safe; the root was a very high accumulator, sometimes more than the arsenic concentration applied. In a large data set (Biswas et al. 2012) of arsenic accumulation in different vegetables collected from the fields and the surrounding local markets, the authors found that spinach had the maximum arsenic accumulation (0.910 mg kg^{-1} wet weight) among the leafy vegetables, whereas among the non-leafy vegetables the highest arsenic accumulation was found in the tomato (*Lycopersicon*, 0.551 mg kg^{-1} wet weight). Among the roots and tubers, *Arum* had the highest arsenic accumulation (0.558 mg kg^{-1} wet weight). Among the pulses, Bengal gram (*Cicer* sp.) had the highest arsenic accumulation (0.891 mg kg^{-1} wet weight). In carrot (Mayorga et al. 2013), arsenic concentration in the leaves and roots increased with increased arsenic concentration of irrigation water, with a high magnitude of arsenic translocation from roots to leaves, although the leaves had higher arsenic affinity than the roots, and overall it was a slow and continuous translocation to be of environmental risk. Rahaman et al. (2013) reported the arsenic concentrations in different vegetable crops of Malda district, West Bengal, and found the highest arsenic accumulation in potato, 0.456 mg kg^{-1}, followed by amaranth, radish, cauliflower, carrot, tomato, bitter gourd, chili, spinach, and cabbage. Kar et al. (2013) described arsenic accumulation in different vegetable crops in Taiwan and found eggplant to be the highest arsenic accumulator (0.140 mg kg^{-1}) followed by amaranth, tomatoes, spinach, and cabbage.

In a comparison study (McBride et al. 2015) among vegetables, lettuce was the highest arsenic accumulator and tomato was the lowest, and the tomato proved to be safe in terms of arsenic accumulation even when fields were highly loaded with arsenic. For the other vegetables, however, arsenic accumulation exceeded the mark of safety.

It is noticeable that arsenic accumulation in vegetable crops depends on the availability of arsenic in the soil–water system; otherwise, the same crops or vegetables should contain the same amount of arsenic when cultivated in different arsenic-laden crop fields. From the foregoing discussion, another noticeable point is that different parts of plants (roots, stems, leaves, fruits) accumulate different amounts of arsenic in their tissues. In general, the accumulation follows the trends of arsenic load in root > stem and leaves > fruits (Roychowdhury et al. 2005; Dahal et al. 2008; Biswas et al. 2012; Kar et al. 2013). A study by Intamat et al. (2017) assessed arsenic bioaccumulation in aquatic plants and found the highest arsenic accumulation in *Limnocharis flava* (0.78 ± 0.31 mg kg^{-1} dry weight) with a bio-accumulation factor of 131.30 ± 15.35; this accumulation was from aquatic sediments.

3.2 Arsenic Bio-Availability and Bio-Accessibility

Bio-availability is the gastrointestinal (GI) digestion, absorption, metabolism, and tissue distribution of any wanted nutrient or bioactive compounds (Galanakis 2017). So, it is the rate at which the therapeutic substance is absorbed and becomes available at the active site or the fraction of the stored nutrient being available in physiological functions, that is, either reaches the systemic circulation and is ultimately utilized. Bio-accessibility is the quantity of a compound that is released from its matrix in the gastrointestinal tract and is available for absorption, that is, enters the bloodstream. So, this is digestive transformation of foods into ready material available for assimilation. Not all the arsenic present in food, including arsenic species, is bio-available from the corresponding food matrix (Bastias et al. 2013). Element bio-accessibility depends on the matrix and chemical form and, in the in vitro studies, on the model used. In the past few years different studies have reported different methods and models to assess trace element bio-accessibility (Laparra et al. 2003; Moreda-Pineiro et al. 2011; Ruby et al. 1996; Intawongse and Dean 2006). Most of the researchers used batch gastrointestinal models, wherein samples are sequentially exposed to artificial saliva and gastric and intestinal fluids. Some dynamic models have also been reported, viz., TIM (TNO GastroIntestinal Model), a computer-controlled system with several chambers simulating conditions in the stomach, duodenum, jejunum, ileum, and large intestine (Minekus et al. 1999; Torres-Escribano et al. 2011). Chu and Beauchemin (2004) reported a continuous leaching bio-accessible method for successive leaching of the food sample by artificial saliva, gastric fluid, then intestinal juices in order.

3.2.1 Bio-Accessibility of Arsenic: From Rice to Human

Bio-accessibility studies of arsenic can help to evaluate the bio-availability and health effects of dietary arsenic exposure. Different researchers have defined the term “bio-accessibility” in different ways based on the scope and objective of the study performed. Koch et al. (2007) and Wragg et al. (2011) described bio-accessibility of arsenic as the fraction of arsenic that is released into the aqueous phase within the gastrointestinal tract and is readily available for absorption. It is also defined as the fraction that is mobilized into the gut fluids after ingestion and is available for assimilation (Ruby et al. 1999; Peijnenburg and Jager 2003). Bio-accessibility tests are often used as an approximation of bio-availability. A contaminant is said to be bio-available if it, as a whole or in parts, becomes available at the site of action after ingestion, inhalation, or contact with the skin. The bio-accessibility of arsenic depends on several factors such as the properties of the matrix, the oxidative state of arsenic, and the conditions of the surrounding medium. For instance, in soil, its bio-accessibility is affected by the ubiquitous and sequestering properties of soil (Stewart et al. 2005), whereas in the GI tract, physiological conditions such as pH (Conklin et al. 2008; Sharma and Sohn 2009) and bile concentration determine its solubility (Oomen et al. 2003). Bio-accessibility studies help in identifying and developing different scenarios to which humans can be exposed. These determinants of bio-availability and bio-accessibility must be understood if one is to monitor or, ultimately, predict the effects of contaminants (Peijnenburg and Jager 2003). In vitro, research has been done to assess bio-accessibility of heavy metals such as lead (Koch et al. 2011), cadmium (Waisberg et al. 2004), and arsenic (Laparra et al. 2005; Trenary et al. 2012; Sun et al. 2012) in different media such as soil, plants, and water. The first release from the rice matrix takes place in the mouth; thereafter, a fraction of arsenic is mobilized from the matrix through the digestive juices in the GI tract. The mobilized arsenic could then be transported into the portal vein via the epithelial membrane. Finally, the fraction of arsenic that reaches the systematic circulation from the liver without being metabolized is the bio-available fraction.

Rice has been found to provide a major quantified exposure of inorganic arsenic to populations living on a rice diet (Meliker et al. 2006; Tsuji et al. 2007). Daily consumption of rice with a total arsenic level of 0.08 mg kg^{-1} would be the equivalent to drinking contaminated water with an arsenic concentration of 0.01 mg l^{-1} (Williams et al. 2006). With the growing concern about the presence of arsenic in drinking water, although people had changed their drinking water sources to less contaminated ones, that had not served to prevent arsenic toxicity. The average contribution to total arsenic intake from drinking water was only 13%, whereas from cooked rice was 56%, which made it clear that cooked rice contributed most to the daily arsenic intake (Ohno et al. 2007). Later, that fact was repeated in several studies from West Bengal, India (Guha Mazumder et al. 2013, 2014; Biswas et al. 2014a, b). Because paddy soil is anaerobic, rice has the ability to accumulate higher levels of arsenic into grain than other cereal crops (Meharg and Rahman 2003; Williams et al. 2006, 2007; Su et al. 2010). The risk posed by inorganic arsenic exposure from rice is governed by the inorganic arsenic concentration in rice grain and the amount ingested through the grain (Kile et al. 2007; Ohno et al. 2007; Meharg et al. 2009). Generally, it has been found that inorganic arsenic is more toxic than the pentavalent methylated arsenic species (Schoof et al. 1999). The rice grain can contain an undesirable amount of methylated arsenic species, especially DMA (Meharg et al. 2009). However, the relative concentration of inorganic and organic arsenic in rice grain depends on rice genotypes and the rice-growing environment (Liu et al. 2006; Xu et al. 2008; Li et al. 2009; Norton et al. 2009).

A study examining the bio-accessibility and bio-availability of arsenic in rice cooked in arsenic-contaminated water, using simulated in vitro gastrointestinal digestion and Caco-2 cells, found that arsenic bio-accessibility ranged from 63% to 99% (Laparra et al. 2005). However, arsenic uptake by Caco-2 cells varied from 3.9% to 17.8%, suggesting that other soluble components of the rice may limit the extent of arsenic absorption. Sometimes in vitro assessments do not fully follow the results of the real scale, as Laparra et al. (2005) found only 12% arsenic absorption in case of an in vitro model but this was much higher in the real-time cell line study. Trenary et al. (2012) showed 61% bio-available arsenic from market-available contaminated rice in the US market, and the trend was somewhat similar to the report of Sun et al. (2012), wherein bio-available arsenic was 37% to 57% in the market rice of China. On the other hand, Esther et al. (2014) observed very high, that is, 83% bio-available arsenic in rice collected in Thailand. Not only from rice, but arsenic is also bio-available from the soil; Jeong et al. (2013) reported 0.64%, 11.9%, and 2.02% arsenic bio-availability from Korean soil. At cattle tick dip sites, in northeastern New South Wales, Australia, Juhasz et al. (2007) showed bio-availability of arsenic of 45.37% at the highest. It is clear that soil arsenic is less bio-available compared to arsenic in rice. These results show that in endemic areas with subsistence rice diets, the contribution of inorganic arsenic from cooked rice should be considered in assessments of arsenic health risk (Laparra et al. 2005).

3.2.2 Arsenic Bio-Availability in Humans from Foods Other Than Rice

Few studies have focused on the issues of arsenic bio-availability from foods and vegetables other than rice. In a very recent study, Pizarro et al. (2016) have compared the total arsenic content and total arsenic bio-accessibility from carrot, beet, and quinoa. Almost all the arsenic from carrots (98%) and beets (90%) is bio-accessible following the in vitro gastrointestinal digestion model, but in quinoa only 40% of arsenic is bio-accessible. Leufroy et al. (2012) assessed arsenic bio-accessibility of some seafood materials and found more than 50% bio-accessible arsenic considering all the samples, such as 53–83% in fish and 58–117% in shellfish. The highest bio-accessibility was observed for scallop and crab (117% and 98%, respectively); from canned tuna and salmon, bio-accessibility was between 53% and 65%.

3.3 Arsenic Uptake and Bio-Availability in Animals

Only field study data have been considered here, and the limitations were that most of the reports and experiments were in-house laboratory experiments. So, as per availability in the web database and depending on our search criteria, we have summarized the arsenic bio-availability in animals.

Arsenic is a severe alimentary tract irritant in domestic animals, and the most frequent intoxication has been observed in dogs, cats, horses, and pigs (Selby et al. 1977). Arsenic can be taken up by plants and transferred into the food chain, causing severe effects in animals (Somasundaram et al. 2005). In endemic areas, animals, especially livestock, are considered to be affected by the food chain path. Besides the presence of arsenic in soil, domestic and agricultural use of cow dung fertilizer leads to arsenic contamination of the crops (Pal et al. 2007). Most species of livestock and pet animals apparently excrete arsenic rapidly (People 1964). Arsenic distribution in an animal body indicates feces and urine as major excretory paths, in addition to milk in the case of domestic cattle (Datta et al. 2012). Milk products also contain arsenic if cattle are fed with arsenic-contaminated water and straw. Arsenite was the main species to be eliminated through milk, and organo-arsenic species were the main species in the feces (Datta et al. 2010).

Bertin et al. (2013) reported death, diarrhea, ataxia, dehydration, and respiratory distress as the most common clinical signs in cattle with acute arsenic poisoning. Among cattle, the most common clinicopathological abnormalities include azotemia (100%), hematuria (100%), increased liver enzyme activity (86%), and increased hematocrit (60%), although the antidote treatment gave better outcomes and survival with no abnormalities.

Few reports have been published on the accumulation of arsenic in sea turtles. The highest arsenic accumulation has been reported in liver samples of the hawksbill turtle (*Eretmochelys imbricata*, mean, 20.9 $\mu g\ g^{-1}$ dry weight) followed by the loggerhead turtle (*Caretta caretta*, mean, 9.0 $\mu g\ g^{-1}$ dry weight), and the green turtle (*Chelonia mydas*, mean, 2.9 $\mu g\ g^{-1}$ dry weight) (Saeki et al. 2000; Kubota et al. 2003; Fujihara et al. 2003; Agusa et al. 2008). Carnivorous turtle species (hawksbill and loggerhead turtles) tended to show higher arsenic levels than herbivorous sea turtles (the green turtle). Similar differences have been observed among herbivorous and carnivorous mollusks (Cullen and Reimer 1989).

Edmonds et al. (1994) for the first time reported arsenic species in sea turtles: arsenobetaine, As^{3+}, and arsenocholine accounted for 50%, 35%, and 15%, respectively, of water-extractable arsenic in liver of the leatherback turtle (*Dermochelys coriacea*), but this does not occur in marine mammals and seabirds. In 1998, Goessler et al. first reported arsenic in marine mammals and arsenobetaine as the predominant arsenic species in samples of seal (*Pusa hispida*), bearded seal (*Erignathus barbatus)*, pilot whale (*Globicephala melas*), and beluga (*Delphinapterus leucas*).

Limited studies have reported the presence of arsenic species in birds. Arsenic levels are higher in seabirds compared to terrestrial birds (Kunito et al. 2008). Kubota et al. (2002), for the first time, reported arsenic species in different seabirds such as the black-footed albatross (*Phoebastria nigripes*, maximum 26.7 $\mu g\ g^{-1}$) and black-tailed gull (*Larus crassirostris,* maximum 2.25 $\mu g\ g^{-1}$). Bio-accumulation of arsenic or the trophic transfer coefficient (TTC, defined as the ratio of the concentration in a consumer's body to the concentration in diet) (Suedel et al. 1994), was found to be 1.0 for the black-footed albatross considering arsenic concentration in 17 tissues of the body (Fujihara et al. 2004). Although high arsenic bio-accumulation was observed, there were no bio-magnifications.

Chickens rapidly excrete arsenicals, but the feathers retained the highest residue of arsenic in poultry birds in a study from the arsenic-affected Chakdaha block of West Bengal, India (Datta et al. 2012). In a recent work by Intamat et al. (2017), the fish *Oreochromis niloticus* was found to be the highest arsenic accumulator (0.16 ± 0.16 mg kg^{-1} wet weight) with a bio-accumulation factor of 228.21 ± 26.99 among the four fish species sampled. This study also reported one snail species, *Filopaludina sumatrensis*, with maximum arsenic bio-accumulation (0.18 ± 0.06 mg kg^{-1} wet weight) from sediment rather than water, where the bio-accumulation factor was 33.04 ± 10.58.

4 Discussion

The present systematic review provides an extensive look into arsenic bio-availability in both humans and animals. Considering that only the field-based study was of the utmost need for better understanding of the arsenic load of soil and water, articles on the phyto-availability (for rice plant) of arsenic, articles on rice grain arsenic status, and finally, articles on human arsenic bio-availability were discussed. The reason for a broader discussion about arsenic bio-availability in humans from rice was that rice was already known as the major dietary component among arsenic-affected populations worldwide. Other food materials were also considered, but rice proved to be the most favorable food matrix for arsenic bio-availability. Besides the food matrix, variety within the food materials (e.g., different rice varieties) has different bio-accessible amounts of arsenic; the presence of thiol groups in certain rice varieties strongly bound arsenite and make it less bio-accessible. Considering all the foregoing criteria and facts, the best matches relating to our search criteria were selected. This review is also important in noting how research areas on arsenic as a single pollutant have changed over decades, changes that are still going on.

From Table 2 and Fig. 2, one interesting finding is the year-wise change of research trends, in that the first studies reported arsenic occurrence in rice grain from which the risk actually comes for humans. Then, researchers started to consider the matter of arsenic loading in the soil. Soon, researchers became concerned about arsenic bio-availability from soil and from rice, and subsequently from other food materials.

5 Conclusion

This review has attempted to find the major features responsible for the bio-availability of arsenic in humans and animals. Irrigation groundwater is the major arsenic source, which in turn transfers through water and soil systems into

Table 2 Arsenic concentration in soil and rice: worldwide distribution

No.	Study areas/study items	Arsenic concentration in soil (mg kg^{-1}), mean (range, *n*) [% bio-accessible]	Arsenic concentration in rice (mg kg^{-1}), mean (range, *n*) [% bio-accessible]	Arsenic species in rice		Reference
				Organic fraction [% bio-accessible]	Inorganic fraction [% bio-accessible]	
1	Contaminated field, China		0.49 (0.31–0.70, $n = 11$)			Xie and Huang (1998)
2	Farm survey, Taiwan		0.20 (0.19–0.22, $n = 3$)			Schoof et al. (1998)
3	Taiwan		0.130 (0.630–0.170, $n = 5$)			Schoof et al. (1998)
4	Vietnam		0.208 (0.320–0.465, $n = 31$)			Phuong et al. (1999)
	U.S., market rice		0.006–0.142, $n = 26$		11–91%	Heitkemper et al. (2001a, b)
5	Kolsur, West Bengal		0.358 (0.120–0.663, $n = 8$)			Chowdhury et al. (2001)
6	Contaminated field, Domkol, India		0.23 (0.04–0.44, $n = 11$)			Roychowdhury et al. (2002a, b)
7	Contaminated field, Jalangi, India		0.25 (0.06–0.66, $n = 23$)			Roychowdhury et al. (2002a, b)
8	Cooked food, Bangladesh		0.24 (0.11–0.36, $n = 4$)			Huq et al. (2006)
9	Contaminated field, Bangladesh		0.12 (0.07–0.17, $n = 15$)			Duxbury et al. (2003)
10	Bangladesh, Boro rice		0.183 (0.108–0.331, $n = 78$)			Duxbury et al. (2003)
11	Bangladesh, Aman rice		0.117 (0.042–0.172, $n = 72$)			Duxbury et al. (2003)
12	North 24-Parganas, India		0.12–0.66 ($n = 24$)		95%	Rahman et al. (2003)

13	Bangladesh		0.496 (0.058–1.835, $n = 13$)			Meharg and Rahman (2003)
14	Contaminated field, Bangladesh		0.95 (0.21–1.50, $n = 7$)			Islam et al. (2004)
15	Market, Taiwan		0.10 (<0.10–0.63, $n = 280$)			Lin et al. (2004)
16	Bangladesh		0.136 (0.040–0.270, $n = 10$)			Das et al. (2004)
17	Market, Bangladesh		0.13 (0.03–0.30, $n = 15$)			Williams et al. (2005)
18	Market, Italy		0.20 (0.19–0.22)			Williams et al. (2005)
19	Market, India		0.03–0.08		81%	Williams et al. (2005)
20	Market, Bangladesh		0.03–0.30		80%	Williams et al. (2005)
21	Market rice, Valencia, Spain		0.20 (0.09–0.53, $n = 8$), cooked rice 2.48, [12%]	Cooked rice 92%		Laparra et al. (2005)
22	Bangladesh		0.0113 (<0.005–0.020, $n = 4$)			Al Rmalli et al. (2005)
23	House hold survey, Bangladesh		0.358 (0.046–0.111, $n = 46$)		87%	Smith (2006)
24	Egypt		0.05 (0.02–0.08, $n = 108$)			Meharg et al. (2008)
25	France		0.32 (0.12–0.61, $n = 22$)			Williams et al. (2007)
26	Spain		0.13 (0.07–0.21, $n = 10$)			Williams et al. (2007)
27	Railway corridor, South Australia	378.38 ($n = 18$), [33.95%]				Juhasz et al. (2007)

(continued)

Table 2 (continued)

No.	Study areas/study items	Arsenic concentration in soil (mg kg^{-1}), mean (range, *n*) [% bio-accessible]	Arsenic concentration in rice (mg kg^{-1}), mean (range, *n*) [% bio-accessible]	Arsenic species in rice		Reference
				Organic fraction [% bio-accessible]	Inorganic fraction [% bio-accessible]	
28	Cattle tick dip sites, Northeastern New South Wales, Australia	1128 ($n = 13$), [45.37%]				Juhasz et al. (2007)
29	Mine sites, Victoria, Australia,	5653.25 ($n = 8$), [23.5]				Juhasz et al. (2007)
30	Gossan, Australian Capital Territory and South Australia	161.27($n = 11$), [6.85%]				Juhasz et al. (2007)
31	Market basket survey, China		0.121 (0.015–0.586, $n = 88$)			Zhu et al. (2008)
32	Household, West Bengal, India		0.09–0.17		74%	Mondal and Polya (2008)
33	Rice, West Bengal, India		0.09–0.24		89.9%	Roychowdhury et al. (2008)
34	Market survey, Japan		0.19 (0.07–0.42, $n = 26$)			Meharg et al. (2009)
35	Bangladesh		0.143 (0.002–0.557, $n = 214$)			Rahman et al. (2009)
36	Farm survey		0.12 (0.09–0.17, $n = 18$)			Adomako et al. (2011)
37	Market basket, Pakistan		0.09 (0.01–0.20, $n = 5$)			Adomako et al. (2011)
38	Contaminated field, Deganga, India	Soil-17.4	0.572			Stroud et al. (2011)

39	Contaminated field, Nonaghata, India	Soil-6.2	0.459			Stroud et al. (2011)
40	Contaminated field, Faridpur, Bangladesh	Soil-34	0.404			
41	Contaminated field, Sonargaon, Bangladesh	Soil-11.5	0.223			
42	House hold, West Bengal, India		0.01–0.64		91.7%	Halder et al. (2013)
43	Market rice, USA		0.209 ($n = 17$), [61%]	19%	38%	Trenary et al. (2012)
44	Cooked rice, USA		0.389, 0.314, [38–57%]			Sun et al. (2012)
45	Guang Zhou, market		0.49	0.095	0.392	Sun et al. (2012)
46	Hunan province, China		0.338	0.051	0.287	Sun et al. (2012)
47	Market rice, Korea	($n = 30$)			76.94%	Kim et al. (2013)
48	Market rice, USA	($n = 34$)			69.28%	Kim et al. (2013)
49	Old smelter site, Korea	Soil-A 1064, Soil-B 100.1, Soil-C 230.5 [A- 0.64%, B- 11.9%, C-2.02%]				Jeong et al. (2013)
50	Paddy field, Cambodia		0.20 (0.10–0.37)			Seyfferth et al. (2014)
51	Pandan rice, Thailand		0.062, [43–83%]		50%	Brandon et al. (2014)

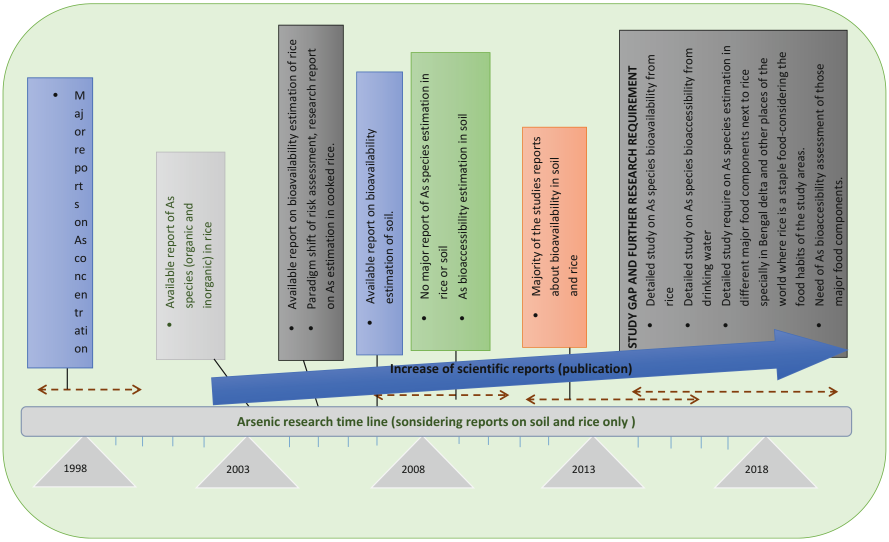


Fig. 2 Timeline of arsenic research focus on rice and soil (details in the main text)

plants and thereafter up through the food chain to humans and animals. To date no permanent solution has been described to restrict the entry of arsenic into the food chain. Several alternative strategies have been proposed to reduce arsenic contamination in water and foodstuffs, such as use of less groundwater, choosing an alternative rice season (Biswas et al. 2013a), choosing low arsenic accumulator rice cultivars (Biswas et al. 2013b), etc.

This risk to food safety, particularly in crop production, emphasizes the need to develop appropriate management practices to minimize the input of contaminants to the environment. This realization indicates a significant need to restrict the entry of arsenic to the food chain by addressing proper soil, crop, and irrigation water management strategies. Some potential measures to reduce higher arsenic exposure in soil and rice based on future research activities are stated here:

- If the present rice cultivation process can be altered with minimum groundwater use, laboratory-based arsenic removal technologies should be field-scale trials in the endemic areas so that the potential technology or plan can be evaluated with a feasibility and sustainability study.
- Crop alteration or choosing low arsenic accumulator varieties may be a good solution in many crops. Adequate knowledge should be developed on the genetic variability of arsenic uptake.
- Factors in the soil and water system that make arsenic labile and phyto-available, and the food chain contamination, need to be minimized.

6 Summary

The present review highlighted arsenic phyto-accumulation, especially in rice and other crop plants, and arsenic bio-accumulation in humans and animals through the food chain. Drinking of arsenic-contaminated water is the prime source of arsenic for humans and animals. Contaminated foodstuffs are also a potential source, as shown by the past 20 years of research. Special attention has been given to arsenic accumulation and food chain contamination through the major crop rice, as it is the staple food in most of the arsenic-affected areas of the world. The discussion has been continued to arsenic accumulation in other food crops. For the first time, an elaborate discussion has been produced on arsenic bio-accessibility from rice and other food crops, as there are differences in total arsenic content (including arsenic species) of the food materials and their bio-availability; the entire content is not bio-available. Bio-availability directly depends on dietary material types. The adverse health effects from arsenic poisoning are directly related to its bio-available fraction, as the maximum portion of ingested arsenic in humans and animals is released through their excreta and urine. Risk assessment should be considered depending on continuous monitoring, and serious preventive measures should be undertaken considering the respective food chain of humans and other animals. Because arsenic accumulation in rice and vegetables (the first and the second major dietary items, respectively) are subject to different accumulation levels (potential) in different edible parts, so special care is needed during analysis and reporting of the arsenic load and corresponding risk assessment by the same, considering the most edible parts may be the whole plant or parts of a plant.

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