

ARSENIC: A Review on Exposure Pathways, Accumulation, Mobility and Transmission into the Human Food Chain

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1 Introduction

Arsenic (As) contamination of the environment from both natural and anthropogenic sources is nowadays a major environmental concern in various parts of the world, due to its persistence and carcinogenic effects on living organisms at certain concentrations. It is estimated that more than 150 million people around the world are exposed to As, consuming water containing high levels of As (Ravenscroft et al. 2009). Besides drinking water, diet also plays critical role in As

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exposure. Vegetables and crops grown in As contaminated soil and irrigation water can take up As through their roots and accumulate it in their edible tissues, which are then consumed by people or animals.

It is a well-known fact that arsenic toxicity is evaluated by considering its presence, nature (organic-inorganic) and concentration in different media such as soil, water and plants. Presence of As in soils varies with As mobility, soil properties and adsorption/desorption capacity of minerals that occur together with As (Kar et al. 2010; Hsu et al. 2012). Each person is affected by As at different levels at similar concentrations depending on age, genetic characteristics, type of exposure (acute or chronic) and presence of co-morbidities (Keil et al. 2011). In addition to these, duration and magnitude of exposure, origin of As and daily dietary habits are also important factors, determining the severity of the effects (Kapaj et al. 2006).

Due to the fact that arsenic does not have taste, odor and color even at high concentrations, it is neither easy to detect its presence in water and food, nor is it easy to avoid exposure to it (Smith and Steinmaus 2009). This is the main reason why additional detailed studies are necessary to avoid exposure to it, such as its transfer capabilities in different media. Metals such as Cd, Zn, Cu including As are persistent in the environment, unlike organic chemicals which act on the contrary (Keil et al. 2011). However, once As is present in the environment, it can easily spread from soil—water, water—soil and then to plants, due to its high transfer capacity. In addition to these, it can transfer from air to soil via dust and atmospheric deposition, whereas it can as well transfer to surface and groundwater through precipitation. Through dust transfer, it can be transported over long distances attached to suspended particles in air. Csavina et al. (2012) discussed the strong ability of ultra-fine particles formed from mine tailings to disperse and flocculate in the range of 0.1–1 μm particle size in the atmosphere. Even though many people are of the belief that soil and water are the major transporting agents for heavy metals, it is observed that air is also a significant transporting agent for As and future researches should concentrate more on this issue.

According to Chung et al. (2014), As can be dissolved in water causing contamination of both surface and ground waters. It has the ability of changing its valence, attaching to or separating from particles that exist in sediments, water or air, in case of interaction with oxygen or other molecules.

2 Pathways of Exposure

Arsenic (As) occurs in all environmental compartments (water, soil and air) and can be taken up by animals and plants and consequently transferred to humans. *Ingestion* of As containing drinking water and foods; *dermal uptake* of As contaminated soil/sediment; *inhalation* of suspended arsenical dusts in the air are some of the pathways of As exposure (Lijzen et al. 2001; Orloff et al. 2009; Biswas et al. 2012). In some cases, drinking of groundwater containing high levels of naturally occurring

inorganic As may lead to mass poisoning, since it is a direct exposure pathway. This is how and why Smith et al. (2000) reported an incident at Bangladesh in 1996 as the largest mass poisoning, where more than half of the population of the country were endangered.

Total dietary exposure to As should be calculated considering both drinking water and foodstuffs. However, most of the research reported in literature covers only As amount in drinking water as the total exposure parameter, causing under-estimation of health risks. For people who are not exposed to As through drinking water; food might be the main contributor of As exposure. Figure 1 presents all the mechanisms/pathways involved in As exposure of humans.

Soils, sediments, groundwater, surface water and air are the environments for accumulating As. This As accumulated in these environments is then transmitted to plants, animals and humans through the food chain. Due to the fact that toxic heavy metals including As persist in the environment for a long time, it becomes a serious concern due to their probable harmful effects. Both direct and indirect pathways have great effect on human exposure (Lam and Sia Su 2009). One of the most dangerous situations occurs when As contaminated groundwater is continuously used for irrigating vegetables and crops. Under such circumstances, irrigation with As enriched groundwater becomes the main pathway for As to enter the human body (Samal et al. 2011; Chatterjee et al. 2010).

Presence of As in surface soils is either natural or artificial. Natural presence stands for As already present within the soils, while artificial presence stands for As introduced by pesticides, herbicides and As contaminated irrigation waters for long periods (Huang et al. 2006; Bhattacharya et al. 2010). Ramirez-Andreotta et al. (2013) demonstrated a linear correlation between As concentration in soils and accumulated As amount in edible parts of some vegetables (lettuce, radish,

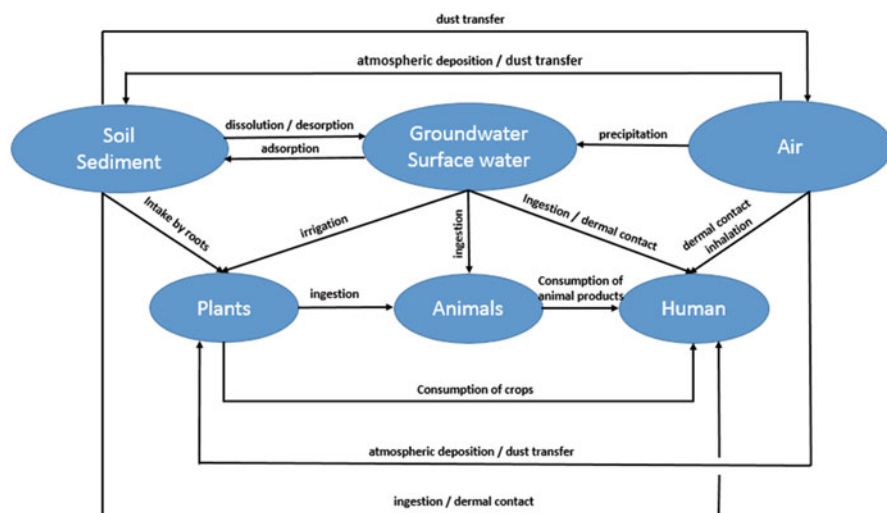


Fig. 1 Pathways of As exposure

broccoli, cabbage, onion, garlic, spinach, beets and bean). The study did not show any correlation for the following vegetables: tomato, pepper, cucumber and squash. Probably because, these vegetables have less uptake capacity compared to others so that they do not accumulate high amounts of As even at high levels of bioavailability in soil. The study thus differentiated bio-concentration factors (BCF) for different vegetable species, where non-correlation findings depend on As absorption capacity of root structure, as well as genetic and physiological adaptations which allow them to absorb, translocate from root to shoot and resist high amount of As in their cells (Wang et al. 2009; Bondada et al. 2007; Samal et al. 2011).

Sometimes, anthropogenic activities affect the toxic heavy metal content of agricultural soils, such as mining and metallurgical activities. The As present in the contaminated soils then may enter the edible parts of vegetables creating health risks for the inhabitants surrounding the area. The spread of contamination is not limited to the nearby surroundings and under certain circumstances such as winds and surface water runoff during excess rainfalls, contamination spreads to long distances. Samples of alfalfa, onion and carrot collected at Vinto-Oruro mining district illustrated high contents of As in these vegetables at concentrations of 399, 99 and 92 ppm respectively. Same enlargement mechanism for airborne transportation is valid for metallurgical regions as well. A decreasing trend of As accumulation in edible parts with respect to the distance from the metallurgical zone was also noted, which suggested that airborne transport of As was the basic reason for high As accumulation in different vegetables (Mercado et al. 2009). In another study carried out around the Zambian Copper belt, the cassava leaves studied did not indicate any excessive Cu ingestion but Pb and As contamination was occasionally recorded. The dust coming from smelters was the major source of contamination (Křibek et al. 2014). Similarly, numerous studies on human urine and blood demonstrated the inverse relationship of As exposure with distance of inhabited areas from metal smelters or mining areas. This situation once again illustrates the long distance travel of airborne As with convenient wind speed and direction (Csavina et al. 2012). On the other hand, we must bear in mind the fact that heavy metals in air, emitted by vehicles and industrial activities can be deposited on vegetables during production, transportation and selling posing threat on food safety and human health. Ali and Al-Qahtani (2012) studied four major industrial areas in Saudi Arabia (Tabouk, Riyadh, Damamm and Jazan) and searched for the accumulation of heavy metals in different parts of the vegetables (roots, stems, leaves, fruits, cereals and legumes). They found that the accumulation of heavy metals was higher in leafy vegetables than those in other parts. They claimed that it was because leaves were access points of heavy metals from air to vegetables. Also, epidemiological studies carried out indicated that ingested and inhaled As can cause skin and lung cancers respectively. Some other studies also showed that As ingestion may as well lead to internal cancers (Phan et al. 2013).

In Beijing-China, a large scale study was carried out in 2006 to analyze risks of high As concentrations in vegetables to human health. Samples were collected from fields, greenhouses and as well as from supermarkets. A HG-AFS method was adopted to determine concentrations and the results indicated presence of As in

soils in the range 4.44–25.3 ppm, which can be regarded as a significant contamination. In different regions, the maximum permissible concentration of As in vegetables was exceeded (Shandong District: 0.331 ppm vs. 0.25 ppm), while in some this concentration was less (Fengtai District: 0.479 ppm vs. 0.5 ppm). It was also observed that As concentration was much more in field-grown vegetables when compared with those planted in greenhouses (Chen et al. 2006). This might be because vegetables in greenhouses are not affected by air deposition and anthropogenic activities in the surroundings, as much as the vegetables that are grown in the open fields.

The study also indicated a significant difference with respect to bio-concentration factor (BCF):

- a. Group of vegetables with higher BCF's: Rape, radish, pakchoi, onion, mustard, cucumber, Chinese cabbage and cabbage
- b. Group of vegetables with lower BCF's: chili, beans, wax gourd, eggplant, spinach, tomato and celery

3 Transmission and Mobility of Arsenic in Environment

The presence of arsenic (As) in aquifer sediments can be the result of various biogeochemical processes, such as *reductive dissolution of As bearing minerals*, *oxidation hypothesis*, *desorption in alkaline environments*, and *geothermal influence* (Yadav et al. 2015; Chauhan et al. 2009). Detailed explanations regarding these processes are presented below:

- a. *Reductive dissolution of As bearing minerals*: For a long time, reductive dissolution of As bearing minerals was believed to be the main mechanism of As release through weathering of As bearing minerals such as arsenopyrite and other sulfide minerals. Goldberg and Johnston (2001) discussed the well-known association between As and Fe(III) oxides in soils and sediments, since Fe oxides and oxyhydroxides are the most significant absorbents for As due to their high binding capacity. Reductive dissolution of As-bearing, amorphous iron oxide/hydroxide minerals is the most common cause of As mobilization into aquifer systems in reducing environments, where As(V) is promoted to reduce As(III) and to release into groundwater in anaerobic conditions (Islam et al. 2004; Ramos et al. 2014).
- b. *Oxidation hypothesis*: When water table drawdowns and oxygen invades the aquifer, As rich sulfide minerals get oxidized and As is released into the groundwater (Das et al. 1996). Although this is a well-known hypothesis, Yadav et al. (2014) claimed that the oxidation of As rich minerals in Nawalparasi District-Nepal did not cause any change in groundwater As concentration between dry and wet season. Rather, As was released due to the reductive dissolution mechanism of Fe (III) oxyhydroxides in a reducing environment. Studies in a mining district at Zimapan, in the Hidalgo province of

central Mexico, showed that, oxidation and dissolution of As containing minerals within a fractured limestone aquifer is the main As release mechanism, in addition to leaching of As from mine tailings (Sracek et al. 2010; Ongley et al. 2007). Besides these mechanisms, it is suggested that As is desorbed from the surface of hydrous metal oxides and clay minerals under alkaline ($\text{pH} > 8$) and oxidative conditions and released into ambient groundwater (Smedley and Kinniburgh 2002; Bhattacharya et al. 2006).

- c. *Desorption in alkaline environments*: Rango et al. (2013) demonstrated the importance of elevated groundwater pH on mobility of As and other natural occurring contaminants from Quaternary sedimentary aquifers of the Main Ethiopian Rift (MER). This suggests that the activity of As depends on alkaline desorption processes. According to results obtained the highest concentration of As was determined in waters with pH values ranging between 8.1 and 8.9. Similarly, Masscheleyn et al. 1991 showed that desorption of arsenate was enhanced with higher pH values.
- d. *Geothermal influence*: Geothermal water rising through a fractured zone can mix with “cold” shallow aquifers and surface waters causing contamination (Aksoy et al. 2009). In addition, besides mixing of geothermal waters, waste water derived from geothermal plants can also lead to contamination of shallow groundwater and surface water by As and other toxic elements (Gemici and Tarcan 2004; Demirel and Yıldırım 2002). This is confirmed by Bundschuh et al. (2013) in a study area in Turkey. If reductive dissolution of As occurs near a geothermal area, mobility of As is induced by high temperature and pressure of liquid and high residence time. Observed in different countries including Turkey, India, Bangladesh and Taiwan, the tectonic activities at or near geothermal deposits are responsible for the increased temperature and pressure. On the other hand, mobilization of arsenic present in geothermal fluids depends on the time of residence. When the residence time increases, mobilization of As will continue under the present supportive conditions. Geothermal waters may contain both As (III) and As(V) species, however as the water rises up and has contact with atmospheric oxygen near earth surface, As(III) is oxidized to As(V) and dissolved redox sensitive minerals such as Fe oxides are precipitated and change their oxidation states (Webster and Nordstrom 2003; Alsina et al. 2007). Bundschuh et al. (2013) carried out a study in geothermal waters in deep wells and hot springs in western Anatolia and observed that, negative Eh values and positive Eh values correspond to dominance of As(III) and As(V), respectively. This also indicates that As III which is widespread in geothermal waters is formed in reducing environments, while As V is formed in oxidation environments.

It is important to understand the hydrogeological and hydrochemical impacts on As distribution to be able to assess As mobility in groundwater systems. Transmission and mobility of As in the environment affects As concentration. This mobilization is also effective in the case of interrelationship of As with hydro-geochemical characteristics of groundwater. Geological and geomorphological structures are not constant on the earth's surface and may laterally vary to a great extent, even in a small

agricultural zone. For instance, as the hydraulic conductivity of aquifer sediments gets lower and surface topography is flatter, groundwater is mostly immobile (Guo et al. 2008). In the same way, composition and matrix of groundwater, interaction between surface and groundwater, reduction potential, fluctuations in groundwater level, recharge rate, distribution of dissolved oxygen, and organic matter may also vary (Yadav et al. 2015). Depth of the water table affects As concentrations, because of two main reasons. First, an increase in water table brings the groundwater table closer to the land surface and mixes groundwater with agrochemical and other wastes at or near the surface. Second, the rise in water table will cause dissolution of Fe-oxyhydroxide; giving way to the release of As under reducing environmental conditions (Bhattacharya et al. 2001; Nickson et al. 1998). Microbes, on the other hand, can catalytically decompose organic compound resulting in release of Fe, Mn and HCO_3 by the dissolving of As-rich Fe and Mn-oxyhydroxide (Kar et al. 2010; Liaoa et al. 2011; Nickson et al. 1998; Yan et al. 2000). The leachable As content was also observed to be high in organic matter phase, which indicated the important role of microbial populations and organic matter in the mobility of As under reducing condition. As for the reduction potential, high redox conditions with the induction of high concentrations of Fe and Mn enhance elevated levels of As. During reductive dissolution mechanism, As gets adsorbed onto Fe (III)-oxyhydroxides and is released under reduction condition (Islam et al. 2004; Nickson et al. 1998). Moreover, dissolved oxygen in groundwater is consumed during microbial oxidation, inducing an increase in HCO_3 concentration (Liaoa et al. 2011; Kar et al. 2010). This microbial activity causes a reducing, alkaline environment where mobility and thereby the concentration of As increases. The effect of microbial activity and organic matter on As mobilization in groundwater in the upstream of Ganges River basin, The Terai Region of Nepal has been shown recently by Yadav et al. (2015).

Arsenic is a member of the carcinogenic heavy metals and the intensity of studies to assess its presence is an outcome of this basic fact. That is why researchers are trying to locate its presence—favoring conditions for elevated concentrations and methods of fighting against such. When the processes and conditions are favorable, elevated As levels are on the agenda. Karstic formations (sinkhole, cave, and conduit), carbonate bedrocks and hydraulic fracturing are such favorable conditions that affect mobility of As in aquifers and increase the interaction between surface and ground water through fractures (Lang et al. 2006; Wang and Luo 2001; Sophocleous 2002). Due to such increased interaction, As spreads to different areas together with these water resources, thereby affecting large masses.

Drainage/irrigation channels affect biogeochemical processes of As distribution. Contamination of soils/sediments with As varies significantly with such channels and depth of the water table. The main reason for this is the passage of oxygen-rich waters from the irrigation channels towards the drainage channels underground, enhancing adsorption of As onto the sediments (Stute et al. 2007; Guo et al. 2011). Guo et al. (2011) also concluded that oxic conditions increase as the distance from channels decreases, giving way to a decrease in total As and As(III) concentration.

Fluctuation in groundwater level due to irrigation practices has also a great effect on As concentrations in groundwater. Shallow aquifers recharged by surface water

are generally rich in As during dry/irrigation seasons (Cheng et al. 2005; Oinam et al. 2011). By installing samplers along groundwater flow in Hetao basin-Inner Mongolia and monitoring As presence, Guo et al. (2013) suggested that an increase in As concentrations with high groundwater levels during irrigation seasons (in summer between April and September and in winter in November) can trigger a release of As from sediments to groundwater due to dissolution of iron oxyhydroxides and/or reductive desorption of As(V) in environments favoring reduction. On the other hand, Chauhan et al.'s (2009) study carried out at the Ballia District-UP-India demonstrated no important seasonal difference between groundwater As concentrations in three different seasons (summer, monsoon and winter). Similar results were also obtained by Cheng et al. (2005) at Arai-hazar, Bangladesh, in a study repeated for 3 consequent years. The majority of scientific studies on the effect of seasonal variations, especially rainy seasons, showed that As concentrations varied according to seasons. The latter two studies mentioned conclude the opposite. It is evident that rainwater induced seepage is not the only factor affecting infiltration of As from sediments to groundwater. Thus, future studies should elaborate more on other probable factors in these areas.

In addition to effect of drainage/irrigation channels and seasonal variations on As concentration in groundwater, depth is an equally important parameter to consider. British Geological Survey (2001) and Chauhan et al. (2009) suggested utilization of potable water from deep wells, since they contain less As concentrations, when compared with shallow wells. According to Yadav et al. (2015), the most important reason forwarded is contamination of shallow groundwater with agrochemical wastes. However, we must bear in mind the fact that lowering of the groundwater table by excessive extraction can cause As infiltration into deeper aquifers (Van Geen et al. 2003; Rahaman et al. 2013).

Grain size of sediments is also another factor which affects the concentration of As in aqueous environments. Yadav et al. (2015) showed that as the grain size decreases, As concentration increases in sediments, since fine grained materials have larger surface areas and thus a high rate of As adsorption. As a result, concentration of As in groundwater can change depending on the grain size of the sediments. Studies on surface waters and sediments in Hongfeng and Baihua reservoirs in China showed that heavy metals tend to be higher in sediments than in water, due to adsorption effect of heavy metals on the suspended solids and settling of sediments on the lake bed (Wu et al. 2014).

4 Transfer from Soil-Water to Plants

In discussing the uptake mechanism of arsenic (As) by plants when it is present in soil in appreciable concentrations, some parameters that need to be taken into consideration are pH, organic matter, redox potential, clay content, water and nutrient availability and microbial activity. These parameters also influence translocation of As to different plant organs, as well as its forms and amounts in soil (Kabata-Pendias

and Pendias 2001; Inacio et al. 2013). Uptake of As by plants decreases as Fe, Al, Mn oxides/hydroxides and organic matter rich clays in soil increase. The reason for this is that in the presence of such media, As is adsorbed by the soil particles (Yong and Mulligan 2004; Huang et al. 2006; Hsu et al. 2012). It has been shown by Warren et al. (2003) that As concentration in lettuce, radish, and cauliflower decreased by 22% due to addition of ferrous sulfate in soil. In addition, in the same study bioavailability of As was higher in sandy soils compared to clay soils. This is because high sorption capacity of clay causes a decrease in bioavailability of As. For this reason, As phytotoxicity is more likely to occur relatively less in clay soils than in coarse textured soils. In addition, the uptake and translocation of metals in plants is affected by season, age of plant, nutritional status, any disorders, stage of growth and rhizosphere interplay with micro-organisms (Křibek et al. 2014).

Ramirez-Andreotta et al. (2013) explained the difference of As concentrations in plants using the bioaccumulation factor (BCF), which is the ratio of metal concentration in the plant to the metal concentration in soil. Plants with $BCF \geq 1$ are classified as hyperaccumulators (Vithanage et al. 2011) and these kinds of plants are used for phytoremediation.

However, knowing the As concentration of soil itself is not enough to be able to characterize As uptake by plants; bioavailable species of As in soil should also be known (Smith et al. 2008). This is so because only some forms of heavy metals, which are soluble in the soil solution, are bioavailable to plants (Chojnacka et al. 2005).

Moreover, if the As in soil is in the form that renders it possible to be up taken by plants, it will be possible for As to be accumulated at high levels. Bergqvist et al. (2014) claimed the same and said that the opposite is also true if the form of As and thus its bioavailability is lower. The same study also suggested that low to medium As pollution in soils leads to the predomination of arsenite (AsIII) in carrot, lettuce and spinach; while phytotoxic As concentrations in soils result in arsenate (AsV) predomination. This information regarding the presence of arsenite and arsenate in plants is also stressed by Smith and Steinmaus (2009). As a result, it is said that high bioavailable As and a soil with As concentration below phytotoxic levels might enhance the production of vegetables with As-rich edible parts. According to Dixon (1997) arsenate is taken up by plants in oxidizing environments via phosphate transfer system due to the similarity of the arsenate ion with the phosphate ion. This is possible, because phosphate and arsenate ions are similar and exchange of these ions can take place under convenient conditions. For instance, the phosphate in plants can change place with arsenate and thus the plant becomes contaminated with As. Similar bio-transfer of As to human red blood cells occurs in the presence of phosphate. Especially in carcinogen cells, the phosphate changes place by As and As accumulates in the cells at greater rates. Besides inorganic As species, organic As species exist in plants too, which are produced as a result of methylation of inorganic As (Raab et al. 2007). However, other authors like Lomax et al. (2012), claimed that plants do not have any methylation capacity and the reason of organic As existence in plants is microorganisms. The methylation process is also observed in animal tissues for the transformation of inorganic As into organic form. For instance, Centeno et al. (2002) mentioned that although As can be accumulated in

animal tissues, the accumulated As in animal tissues can be partly transformed into organic As through biomethylation processes.

5 Arsenic in Plants

Dietary intake of heavy metals poses risks to both human and animal health. Although wide range of people is exposed to arsenic (As) through the consumption of As containing water; foodstuffs (i.e., crops, vegetables) are also a significant route of exposure for remarkable amount of people. Even though diet plays a significant role in human As exposure, there was lack of data on inorganic As (inAs) in foodstuffs in the literature until a decade ago. According to EFSA (2009) almost 98% of published studies in 15 European countries considered total As in various foodstuffs without differentiating organic and inorganic As species. Since organic As species are less toxic to the human body when compared with inorganic species, future studies should concentrate on such studies, especially on bioavailability and toxicity of As depending on its various species. Thereby, overestimation of health risks will be avoided.

The Joint FAO/WHO Expert Committee on Food Additives set a provisional tolerable daily intake (PTWI) of inAs of 2.1 $\mu\text{g/kg}$ body weight per day in 1983 and a provisional weekly tolerable intake (PWTI) of inAs level of 15 $\mu\text{g/kg}$ body weight per week in 1988. However, the European Food Safety Authority (EFSA) Panel of contaminants in the food chain (2009) concluded that these weekly and daily tolerable limit values set by the Joint FAO/WHO Expert Committee on Food Additives are not valid, since inorganic As leads to lung, urinary bladder and skin cancer even at lower concentrations than tolerable limits set.

Most of the studies carried out were related to rice and rice products, followed by the studies related to fish and seafood, seaweed and algae and beverages especially apple juice consumed mostly by children. However, in this review it was dealt with some tuberous, leafy and fruity vegetables and pulses. Importance was attributed to include only results with standard deviations and so some of the researches were excluded due to the absence of standard deviation values. Unfortunately, some of the vegetables were studied once in recent researches and it was impossible to compare them with other studies. On the other hand, it is observed that most of the studies were carried out by researchers in West Bengal-India, mainly because of high occurrence of contamination.

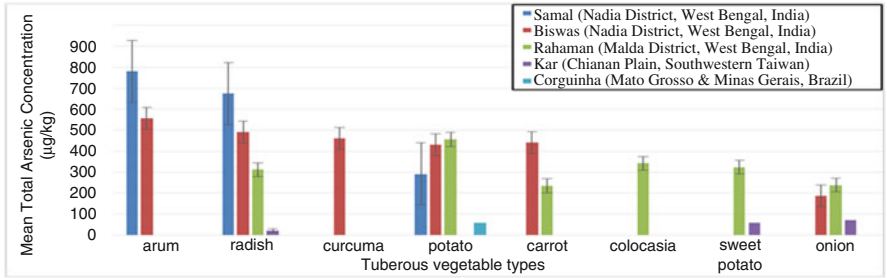
Samal et al. (2011) carried out some studies in West Bengal-India, on various crops which were irrigated by As enriched groundwater and observed considerable variations in the amount of As accumulation in different vegetables. Underground tuberous vegetables (such as arum, radish and potato) contained the highest As concentrations ($780 \pm 243 \mu\text{g kg}^{-1}$, $674 \pm 211 \mu\text{g kg}^{-1}$, $291 \pm 176 \mu\text{g kg}^{-1}$) while leafy vegetables (cabbage, amaranthus, spinach) contained the second highest As concentrations ($315 \pm 69.7 \mu\text{g kg}^{-1}$, $265 \pm 158 \mu\text{g kg}^{-1}$, $270 \pm 182 \mu\text{g kg}^{-1}$). However, unexpectedly cabbage within leafy vegetables showed higher As concentration than potato.

The third highest group was found to be fruity and fleshy vegetables (bitter gourd, brinjal, tomato, etc.) ($262 \pm 133 \mu\text{g kg}^{-1}$, $217 \pm 80 \mu\text{g kg}^{-1}$, $84.4 \pm 48.5 \mu\text{g kg}^{-1}$) and lastly the fourth highest group was pulses (lentil, pea) ($24.7 \pm 16.7 \mu\text{g kg}^{-1}$, $69.2 \pm 22.9 \mu\text{g kg}^{-1}$). On the other hand, the study at the same time showed that crops which require more water for their growth generally had higher As concentrations than crops which need less water. This finding illustrates that using of As enriched groundwater for irrigation purposes can lead to transfer of As through the water-soil-crop-food chain.

Similarly, Biswas et al. (2012) analyzed some leafy, fruity, tuberous and pulses vegetables grown in As contaminated areas in West Bengal-India and compared those collected from market basket. Thirty-two types of vegetables and seven types of pulses were collected from the agricultural areas and eighteen products (vegetables and pulses) were collected from the market. Among all cultivated vegetables tested in this study, pea and lentil from the pulses family showed the highest As concentrations with $1300 \pm 480 \mu\text{g/kg}$ and $1120 \pm 144 \mu\text{g/kg}$ respectively. These are the highest values of As measured in all vegetables listed in this review. These values are also exceeding the WHO-recommended permissible limit ($1000 \mu\text{g/kg}$) for foodstuffs. On the other hand, among roots and tubers, arum tuber—as expected—showed the highest concentration $558 \pm 73 \mu\text{g/kg}$ and onion bulb showed the lowest with $187 \pm 77 \mu\text{g/kg}$. Interestingly, spinach ($910 \pm 259 \mu\text{g/kg}$) had a higher concentration than tuberous vegetables. Except tomato ($551 \pm 262 \mu\text{g/kg}$) and bitter gourd ($529 \pm 44 \mu\text{g/kg}$), fruity vegetables showed again the lowest concentrations. The total As value of vegetables taken from the market appeared to be lower than the level observed in field vegetables. It was reported that vegetables obtained from the market were imported from nonlocal areas. Thus, it can be concluded that only field surveys or only market surveys are not adequate to give actual photograph of As contamination of foodstuffs. Results of this study showed the highest concentrations almost for each kind of vegetables among all other results taken from different studies. Only the concentrations of As in arum and radish were found higher by Samal, although they both tested vegetables cultivated in Nadia District, West Bengal.

In general, Tables 1, 2, 3 and 4, it can be concluded that tuberous vegetables generally have the highest As concentration followed by leafy vegetables, fruity vegetables and pulses. However, the rates of accumulation change considerably depending on effective parameters enhancing uptake. As seen in the study carried out by Biswas et al. (2012), lentil, pea and spinach showed extremely higher concentrations than tuberous vegetable types. Biswas' study is an exception which prevents any generalization. This proves that As concentrations in soil and irrigation water, coupled with environmental factors, have great impact on concentration of As in vegetables. Only those vegetables cultivated under similar conditions can be compared to each other. Transfer factors which consider As concentrations in soil and water should be compared for vegetables cultivated in different agricultural fields. Arum, radish and potato show highest accumulation capabilities with regard to As in tuberous vegetables. Cabbage and amaranth in leafy vegetables have almost similar accumulation capacity in the same study areas.

Table 1 Mean concentrations of total As in edible parts of various tuberous vegetables



| Foodstuff | Type | Concentration (total mean As) (µg/kg) | Origin | Methodology | Reference |
|------------|------------------|--|--|---|---|
| Arum tuber | Roots and tubers | 780 ± 243 558 ± 73 | Nadia District, West Bengal, India Nadia District, West Bengal, India | FI-HG-AAS HG-AAS | Samal et al. (2011) Biswas et al. (2012) |
| Radish | Roots and tubers | 674 ± 211 491 ± 122 312 ± 7 21.5 ± 3.64 | Nadia District, West Bengal, India Nadia District, West Bengal, India Malda District, West Bengal, India Chianan Plain, Southwestern Taiwan | FI-HG-AAS HG-AAS HG-AAS GFAAS | Samal et al. (2011) Biswas et al. (2012) Rahaman et al. (2013) Kar et al. (2013) |
| Curcuma | Roots and tubers | 461 ± 128 | Nadia District, West Bengal, India | HG-AAS | Biswas et al. (2012) |
| Potato | Roots and tubers | 456 ± 7 431 ± 55 291 ± 176 59 ± 11.58 | Malda District, West Bengal, India Nadia District, West Bengal, India Nadia District, West Bengal, India Mato Grosso and Minas Gerais, Brazil | HG-AAS HG-AAS FI-HG-AAS GF-AAS | Rahaman et al. (2013) Biswas et al. (2012) Samal et al. (2011) Corguinha et al. (2015) |
| Carrot | Roots and tubers | 441 ± 53 235 ± 4 | Nadia District, West Bengal, India Malda District, West Bengal, India | HG-AAS HG-AAS | Biswas et al. (2012) Rahaman et al. (2013) |

(continued)

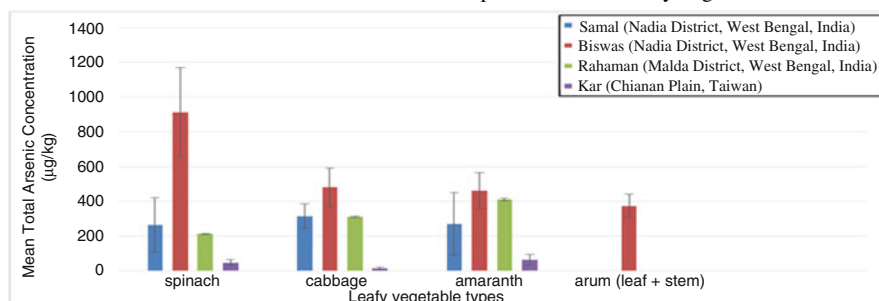
Table 1 (continued)

| Foodstuff | Type | Concentration (total mean As) ($\mu\text{g/kg}$) | Origin | Methodology | Reference |
|--------------|------------------|--|--|---------------------------|--|
| Colocasia | Roots and tubers | 342 ± 7 | Malda District, West Bengal, India | HG-AAS | Rahaman et al. (2013) |
| Sweet potato | Roots and tubers | 324 ± 9 58.7 ± 8.17 | Malda District, West Bengal, India Chianan Plain, Southwestern Taiwan | HG-AAS GFAAS | Rahaman et al. (2013) Kar et al. (2013) |
| Onion | Roots and tubers | 238 ± 6 187 ± 47 72.2 ± 3.06 | Malda District, West Bengal, India Nadia District, West Bengal, India Chianan Plain, Southwestern Taiwan | HG-AAS HG-AAS GFAAS | Rahaman et al. (2013) Biswas et al. (2012) Kar et al. (2013) |

In addition, many authors demonstrated that different parts of plants (roots, stems, leaves, fruits) accumulate different amounts of As in their tissues. The descending order of this deposition is illustrated to be in the order root—stem and leaves—fruits (Kar et al. 2013; Dahal et al. 2008, Roychowdhury et al. 2005). It means that, As is taken up by roots from water or soil and translocated to aerial organs in decreasing order from stems to leaves and finally fruits. From the test results, this order of As deposition was observed. For instance, for peas roots appeared to contain $54.3 \pm 5.82 \mu\text{g/kg}$, stems $24.1 \pm 6.18 \mu\text{g/kg}$ and leaves and edible parts $21.7 \pm 6.32 \mu\text{g/kg}$. Again, maize roots contained $92.5 \pm 8.2 \mu\text{g/kg}$, stems $32.5 \pm 3.64 \mu\text{g/kg}$ and leaves and edible parts $19.5 \pm 2.68 \mu\text{g/kg}$. For rice also, which is one of the plants accumulating comparatively higher As levels, roots contained $182 \pm 47.2 \mu\text{g/kg}$, stems $47.3 \pm 12.5 \mu\text{g/kg}$ and leaves and edible parts $26.7 \pm 39.5 \mu\text{g/kg}$, respectively. These results which can be generalized indicating such an order is best observed in the studies of Kar et al. (2013) and Fig. 2 displays these results in a modified version.

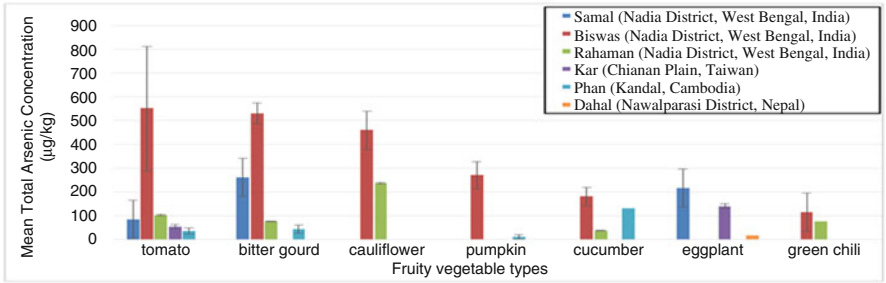
6 Arsenic in Rice and Effect of Processing

Inorganic arsenic (inAs) content in a food varies according to food sub-types and samples. FDA (2012) demonstrated for rice-based cereals that three different samples had different concentrations of 16%, 61% and 87% inAs respectively. On the other hand, for brown rice of varying types (jasmine, long-grain, short-grain, sticky) concentrations ranged from 26 (Zavala et al. 2008) to 95% (Huang et al.

Table 2 Mean concentrations of total As in edible parts of various leafy vegetables

| Foodstuff | Type | Concentration (total mean As) (µg/kg) | Origin | Methodology | Reference |
|--------------------|-----------------|---|--|--|---|
| Spinach | Leafy vegetable | 910 ± 259 265 ± 158 213 ± 3 46.2 ± 16.8 | Nadia District, West Bengal, India Nadia District, West Bengal, India Malda District, West Bengal, India Chianan Plain, Southwestern Taiwan | HG-AAS FI-HG-AAS HG-AAS GFAAS | Biswas et al. (2012) Samal et al. (2011) Rahaman et al. (2013) Kar et al. (2013) |
| Cabbage | Leafy vegetable | 482 ± 111 315 ± 69.7 311 ± 5 12.6 ± 6.72 | Nadia District, West Bengal, India Nadia District, West Bengal, India Malda District, West Bengal, India Chianan Plain, Southwestern Taiwan | HG-AAS FI-HG-AAS HG-AAS GFAAS | Biswas et al. (2012) Samal et al. (2011) Rahaman et al. (2013) Kar et al. (2013) |
| Amaranth | Leafy vegetable | 462 ± 103 411 ± 7 270 ± 182 63.5 ± 31.5 | Nadia District, West Bengal, India Malda District, West Bengal, India Nadia District, West Bengal, India Chianan Plain, Southwestern Taiwan | HG-AAS HG-AAS FI-HG-AAS GFAAS | Biswas et al. (2012) Rahaman et al. (2013) Samal et al. (2011) Kar et al. (2013) |
| Arum (leaf + stem) | Leafy vegetable | 373 ± 68 | Nadia District, West Bengal, India | HG-AAS | Biswas et al. (2012) |

Table 3 Mean concentrations of total As in edible parts of various fruity vegetables



| Foodstuff | Type | Concentration (total mean As) (µg/kg) | Origin | Methodology | Reference |
|--------------|------------------|---|--|--|---|
| Tomatoes | Fruity vegetable | 551 ± 262 103 ± 3 84.4 ± 48.5 53.4 ± 7.93 35 ± 13 | Nadia District, West Bengal, India Malda District, West Bengal, India Nadia District, West Bengal, India Chianan Plain, Southwestern Taiwan Kandal, Cambodia | HG-AAS HG-AAS FI-HG-AAS GFAAS ICP-MS | Biswas et al. (2012) Rahaman et al. (2013) Samal et al. (2011) Kar et al. (2013) Phan et al. (2013) |
| Bitter gourd | Fruity vegetable | 529 ± 44 262 ± 133 76 ± 1 43 ± 17 | Nadia District, West Bengal, India Nadia District, West Bengal, India Malda District, West Bengal, India Kandal, Cambodia | HG-AAS FI-HG-AAS HG-AAS ICP-MS | Biswas et al. (2012) Samal et al. (2011) Rahaman et al. (2013) Phan et al. (2013) |
| Cauliflower | Fruity vegetable | 459 ± 79 237 ± 3 | Nadia District, West Bengal, India Malda District, West Bengal, India | HG-AAS HG-AAS | Biswas et al. (2012) Rahaman et al. (2013) |
| Pumpkin | Fruity vegetable | 271 ± 57 11 ± 2 | Nadia District, West Bengal, India Kandal, Cambodia | HG-AAS ICP-MS | Biswas et al. (2012) Phan et al. (2013) |
| Cucumber | | | | | |

(continued)

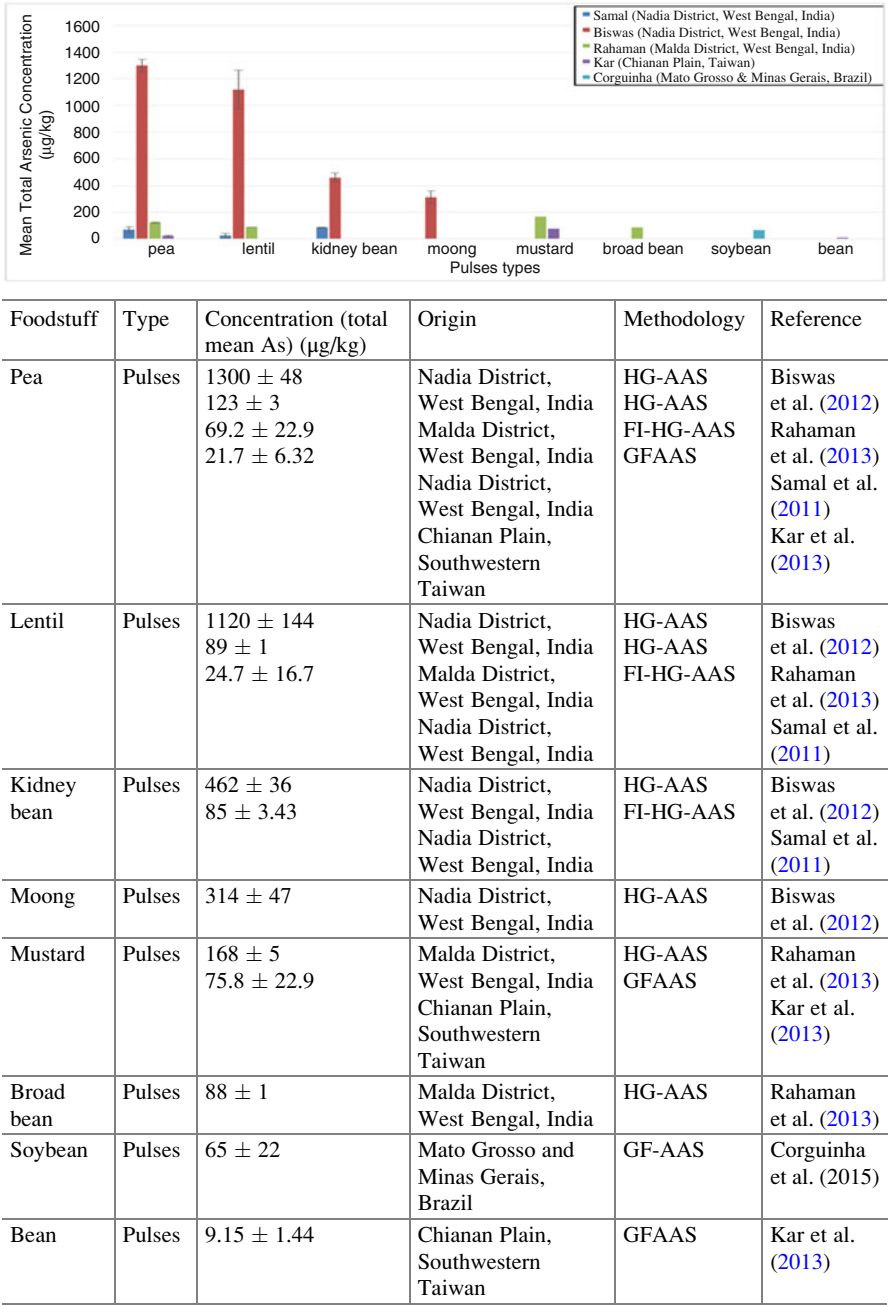
Table 3 (continued)

| Foodstuff | Type | Concentration (total mean As) ($\mu\text{g/kg}$) | Origin | Methodology | Reference |
|-------------|---------------------|--|--|------------------------------|--|
| | Fruity | 181 \pm 38 131 \pm 9 37 \pm 2 | Nadia District, West Bengal, India Kandal, Cambo- dia Malda District, West Bengal, India | HG-AAS ICP-MS HG-AAS | Biswas et al. (2012) Phan et al. (2013) Rahaman et al. (2013) |
| Eggplant | Fruity vegetable | 217 \pm 80 140 \pm 10 16.9 \pm 5.38 | Nadia District, West Bengal, India Chianan Plain, Southwestern Taiwan Nawalparasi Dis- trict, Nepal | FI-HG-AAS GFAAS HG-AAS | Samal et al. (2011) Kar et al. (2013) Dahal et al. (2008) |
| Green chili | Fruity vegetable | 114 \pm 81 76 \pm 1 | Nadia District, West Bengal, India Malda District, West Bengal, India | HG-AAS HG-AAS | Biswas et al. (2012) Rahaman et al. (2013) |

2012). Besides the type of product, processing and preparation of food, washing and cooking methods are other factors that lead to a change in As concentration.

The effect of preparation and cooking processes on As retention in rice is well reported in literature. Lynch et al. (2014) suggested that rice and rice products such as bran and rice flours have higher inAs levels than polished rice. Similarly, Rasmussen et al. (2013) carried out some tests on white and brown rice which had same origin and saw that white rice has a lower total level of inAs (160 $\mu\text{g/kg}$) than brown (400 $\mu\text{g/kg}$) rice. The reason of this is that as the rice is polished to get whiter, bran which has a higher As concentration, is removed from the surface of rice, resulting in low As concentrations (EFSA 2009). On the other hand, Phan et al. (2013) and Mihucz et al. (2007) suggested based on their studies that, rinsing rice with As free water before cooking might eliminate As from As laden rice, resulting in less As in cooked rice than that in uncooked rice. However, in this case essential trace elements (such as Cu, Mn and Zn) enhancing growth and human health, might also be removed along with As (Mihucz et al. 2010). In contrast, it should be born in mind that As concentration in food can be increased by washing it with As bearing water. Availability of especially inorganic As in changing concentrations in water to be used in cooking will change the As content of the food. For example, As bearing water can alter As concentration in rice after cooking due to contaminated water taken up by rice (Signes et al. 2008; Hossain et al. 2012). On the other hand,

Table 4 Mean concentrations of total As in edible parts of various pulses



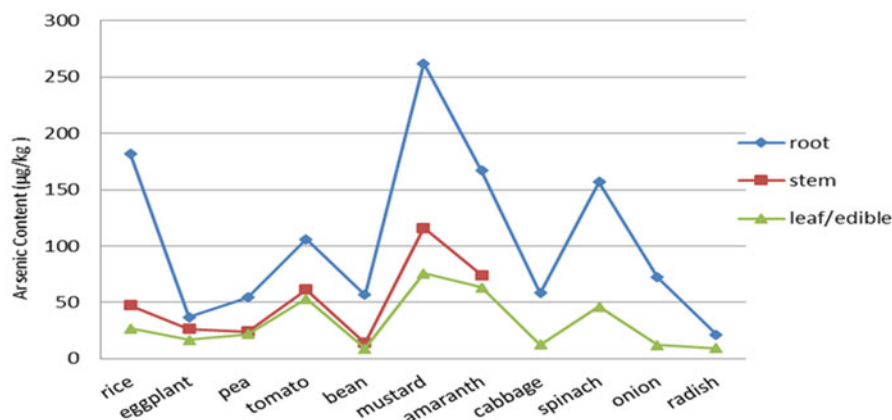


Fig. 2 Variation of As content ($\mu\text{g/kg}$ of fresh weight) in three different parts (root, stem, leaf/edible part) of vegetables (modified from Kar et al. 2013)

Lynch et al. (2014) reviewed based on their data that cooking rice in a high volume of water reduces the total inAs by up to 20% compared to raw samples as well as samples cooked in a small volume of water.

Processed food is also a significant member of the food chain through which As is taken up by the human body. Sugar et al. (2013) emphasizes this in his study and claims that the As contamination does not only come from the As concentration of the water used in irrigation of the vegetable in question but also from the water used for processing. Sugar et al. (2013) demonstrated this result in the form of a linear relationship between As concentration and amount of water used for processing.

Another study again analysing As content versus water used for food processing revealed similar results. Pinto beans and pasta soup taking up high quantities of water displayed highest As concentrations, whereas tortilla taking up a lower water content displayed lower As concentrations (Del Razo et al. 2002). On the other hand, heating parameters such as period and temperature have an impact on the As concentration of food, since as the water vaporized, the weight of the food is decreased causing higher As concentration in cooked products. Davis et al. (2012) tried to attract attention to rice-related danger for children and suggested that rice consumption is a significant source of As exposure for children. Infants and children are exposed to As 2–3 times more than adults, consuming rice based products such as biscuits, crackers, pasta, noodles, pudding, whole grained rice etc. during their childhood (Da Sacco and Masotti 2012).

7 Conclusion

In conclusion, arsenic (As) is one of the most important toxic and carcinogenic metalloids, which can cause serious health risks to living organisms, exposed through soil, water, air and plants. It can easily mobilize from its source to primary receiving environments (soil, water and air) by a series of pathways causing widespread contamination. The risks in question do not only threaten adults, but also infants and children, due to high As concentrations displayed by rice and rice-based products.

Researchers have concentrated a lot on the mobility and transfer mechanism of As in aqueous, airborne and soil environments. These transfer mechanisms are shown to be dependent on bioavailability of inorganic and organic As, while uptake occurs through favourable conditions of certain parameters such as pH, organic matter, redox potential, clay content, water and nutrient availability and microbial activity. High As concentrations on crops and vegetables were especially due to irrigation with As-enriched waters. It was also shown that As uptake by plants decreases in soils with increasing Fe, Al, Mn oxides/hydroxides and organic matter rich clay, since As is adsorbed by soil particles when these oxides/hydroxides are present. On the other hand, desorption of As from surface of hydrous metal oxides and clay minerals increases under alkaline ($\text{pH} > 8$) and oxidative conditions, causing increased As availability in groundwater which can subsequently be taken up by plant roots.

Concentrations of As in each plant species changes depending on its origin (agricultural area, presence/absence of smelters/mines/metallurgical zones), species and contamination rate of agricultural field and nature of irrigation water. Studies also revealed that As accumulation also depends on the different parts of plants. Although not always valid, it can be said in general that tuberous vegetables accumulate the highest As levels and the concentration gradually decreases respectively for leafy vegetables, fruity vegetables and pulses. It must also be born in mind that processed food may also be contaminated depending on washing and cooking methods as well as on the method of processing and rate of absorbance of As-enriched processing water. The majority of researches on As concentration in foodstuffs mostly concentrated on the presence of total As, without giving details on the nature (inorganic, organic) and species of As. More in-depth studies concentrating on separate concentrations of different As species will not only add to scientific database but will also enhance the understanding of related health risks due to their different toxicity effects.

It is not only important to find and analyse the presence and effects of As in soil/air/water-plant-animal-human chain, but it is equally important to continuously monitor As presence. Following the assessment of its amount and its distribution, gaining control and applying remediation measures should continue. Even though it is impossible to get rid of natural As resources, at least anthropogenic As resources such as As containing pesticides, herbicides, chemicals, etc. can be forbidden by the authorities.

8 Summary

This review deals with exposure pathways of arsenic, as well as its transfer and uptake processes from its source to the human body. It is a proven fact that uptake of inorganic As for a long period can lead to chronic As poisoning and a variety of adverse health effects such as skin, lung and bladder cancer, in addition to cardiovascular diseases, diabetes and gastrointestinal symptoms. This risk makes it necessary to continuously search, analyze, monitor and control As contamination of each and every food chain medium (soil, water, plant, animal and human). The danger is not only imposed on adults, but also on infants and children through high consumption of rice-based foodstuffs.

Arsenic exposure occurs primarily from consumption of potable water containing high concentrations of inorganic As. Secondary and almost equally important exposure can be summarized as follows: consumption of crops cultivated in As contaminated (natural or anthropogenic) agricultural fields, consumption of crops cultivated in air-contaminated soils, and consumption of crops irrigated by As-enriched waters. Studies carried out on processed food especially washed and cooked with As-enriched waters demonstrated As contamination, also depending on the water absorption capacity of food type. Thus, consumption of this processed food is also an important As exposure.

The geographical location of the agricultural area is also very important. Detailed study on these areas generally consider the As threat twofold: is it natural or is it anthropogenic. When there are smelters, metallurgical activities, mining activities and similar activities in the surrounding areas introducing chemicals (As and others) to air, soil and water, there is already a risk. Numerous studies demonstrated an increase in the contamination levels of soils and waters in these areas, but under certain circumstances (airborne As and favorable wind speed and direction) accumulation can be inversely proportional with the distance from the contaminated zone. Therefore, continuous monitoring and control measures in such areas are of maximum priority.

In this review, light is also shed on the transfer mechanism of As through the food chain and the parameters that enhance mobility of As in the environment. Since high levels of As accumulation in edible parts of vegetables and crops will be transferred to the human body under convenient conditions, As accumulation in different vegetables and crops and in different parts of these vegetables and crops must also be understood. Numerous relevant studies illustrated that accumulation in descending order in different parts of the vegetables and crops is as follows: roots—stems—leaves and edible parts. Such comparative information, amounts of accumulation in different vegetables and crops as well as in different parts of these vegetables and crops will determine the level of risk imposed on human body. These data will orientate authorities to take necessary precautions to protect the health of people, especially infants and children.

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