



Arsenic Exposure via Contaminated Water and Food Sources

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Abstract: Arsenic poisoning constitutes a major threat to humans, causing various health problems. Almost everywhere across the world certain "hotspots" have been detected, putting in danger the local populations, due to the potential consumption of water or food contaminated with elevated concentrations of arsenic. According to the relevant studies, Asia shows the highest percentage of significantly contaminated sites, followed by North America, Europe, Africa, South America and Oceania. The presence of arsenic in ecosystems can originate from several natural or anthropogenic activities. Arsenic can be then gradually accumulated in different food sources, such as vegetables, rice and other crops, but also in seafood, etc., and in water sources (mainly in groundwater, but also to a lesser extent in surface water), potentially used as drinking-water supplies, provoking their contamination and therefore potential health problems to the consumers. This review reports the major areas worldwide that present elevated arsenic concentrations in food and water sources. Furthermore, it also discusses the sources of arsenic contamination at these sites, as well as selected treatment technologies, aiming to remove this pollutant mainly from the contaminated waters and thus the reduction and prevention of population towards arsenic exposure.

Keywords: arsenic; food contamination; drinking water; health effects; removal technologies

1. Introduction

Arsenic is a ubiquitous toxic metal, belonging in the metalloid group of the periodic table, found naturally in the lithosphere, hydrosphere, and atmosphere, as well as generally in the biosphere [1]. Both organic and inorganic forms of arsenic exist in nature (mostly in the form of complexes); various transportation routes in environment have been identified and rather high concentrations (mainly in water sources) have been reported in several regions around the world [2].

The World Health Organization (WHO) recommended the regulation limit (as imposed by the respective legislation) of arsenic concentration in drinking water at 10 μ g/L [3]. This is also the limit of arsenic in drinking water imposed by the European Commission, the United States Environmental Protection Agency and other inter/national organizations. Contamination of water by arsenic, especially groundwater, due to arsenic's high toxicity, is considered a major health issue in various areas worldwide. Relevant research showed that the long-term exposure to elevated concentrations of arsenic can threaten human health, causing a variety of health disorders, including skin lesions (e.g., keratosis, pigmentation) and various internal and skin cancers [4].

Nevertheless, arsenic has been used in various industries for the production of several products, such as glass, ceramics, electrical appliances, cosmetics and fireworks. In the mid-20th century, arsenic was also widely used to produce pesticides, and for the production



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Copyright: © 2022 by the authors. Licensee MDPI, Basel, Switzerland. This article is an open access article distributed under the terms and conditions of the Creative Commons Attribution (CC BY) license (https:// creativecommons.org/licenses/by/ 4.0/). of wood preservatives [5]. As a result of past and current uses, arsenic contamination is currently considered a problem of great concern for the scientific community, found mainly in water sources, but also in food, threatening the health of millions of people. Mondal et al. [6] revealed that arsenic exposure from food exceeds that from drinking water in the endemic region of Bihar (India), whereas additional areas are expected to be discovered in the near future, such as in Pakistan, according to a recent survey [7].

In this review, a short description of the human exposure to arsenic via water and food sources is reported and the potential pathways of its environmental cycle are presented. In addition, specific data regarding the arsenic concentration in various waters and food products from Asia, Europe, America, Australia and Africa are summarized, according to recent literature publications. The aim of this study is to summarize the world situation regarding arsenic contamination of waters and food, and human exposure to contaminated sources, which remains a threat to health. It is worth noting that the data presented relate to major published case studies and do not reflect the overall situation in each country.

2. Origin of Arsenic Contamination

High concentrations of arsenic may occur naturally (e.g., due to erosion of minerals) in several areas, which are therefore contaminated, and by anthropogenic activities (e.g., industrial production and uses). The continuous exposure of humans to arsenic via food and water consumption can lead to serious health damages, because this is a carcinogenic element with high affinity for thiols. It can also replace phosphorus in biochemical reactions owing to their similar chemical properties, as they belong in the same group of the periodic table, actually being very close, indicating the highly destructive role of this element during DNA replication and metabolic activity [8].

Soil: The predominant forms of arsenic in soils are arsenate (As⁵⁺), arsenite (As³⁺) and organic arsenic. Usually, in soil matrixes it can be found complexed with amorphous iron and aluminum oxides [9]. The arsenic form in the soil varies according to the different textures. For instance, the presence of clay can increase the fixation of arsenic in the respective soil, as at circum-neutral pH values arsenic is adsorbed onto the clay particles [10]. Its concentration and mobility are also dependent on pH and redox potential at the specific environmental sites [11]. Because of the toxic effects of arsenic, it has been used (mainly in the past) for the production of specific herbicides, insecticides, various toxins and decongestants. The use of intensive phosphate fertilizers in agriculture is also considered a potential source for arsenic contamination, because arsenic is a common contaminant of most phosphate minerals, which are used for the production of these fertilizers. However, the amount of arsenic in other fertilizers(e.g., nitrogen or potash) is rather low and can be considered insignificant [12]. Figure 1 shows the various routes of arsenic and arsenic-related compounds that can accumulate in an ecosystem due to anthropogenic activities, resulting in environmental deterioration.

Atmosphere: Arsenic can be released into the atmosphere by natural or anthropogenic activities. According to a rough estimation, the global annual release of arsenic in the atmosphere is 7.8×10^7 kg/year. The natural sources are expected to release 1.2×10^7 kg arsenic per year, wherein volcanoes and microbial volatilization may supplementarily contribute an additional 8.9×10^6 and 2.1×10^7 kg/year, respectively [13]. The amount of arsenic stored in the northern hemisphere is almost five times higher than the amount of arsenic stored in the southern hemisphere, mainly due to the more intensive industrialized conditions existing in the northern hemisphere [14]. Other environmental problems, potentially causing increased arsenic emissions in the atmosphere, may include deforestation, grass burning, and the use of wood as fuel. High and significant concentrations of arsenic are also connected with the emissions of industrial wastes, especially heavy/toxic metals [15]; the concentration of arsenic in sewage sludges is considered to be an indicator of the industrialization degree for the surrounding area.



Figure 1. Pathways through which arsenic and its relevant compounds may enter the environment and contaminate soil, atmosphere and water. Human and natural activities result in As accumulation, mainly in soil and water, where As(V) and As(III) are interconverted via oxidation and reduction bio/reactions. The respective methylated products can be produced from As(III) species, i.e., MMA, DMAA and TMAA, resulting in the formation of MMAA, DMAA and TMAA chemical compounds, mainly through volatilization, while the reverse process occurs through demethylation.

Water: Since arsenic is an element of the Earth's crust, groundwater usually presents the most severe pollution problems among other water resources [16]. It can be found either dissolved in water or in the form of particles. Moreover, it may be transformed to dimethyl and/or trimethyl arsenic compounds by mollusks, crustaceans and fishes [17]. Arsenate is the predominant form found in seawater algae, which play an important role in the biological transport of inorganic arsenic species [18].

3. Hazards and Limits in Food and Water

Inorganic arsenic (iAs) can be found in the environment in several forms, including As(0) (metalloid arsenic), As(III) (arsenite) and As(V) (arsenate). The latter two forms are abundant in natural and drinking waters [19]. Arsenate is the dominant specie in oxic waters, whereas under mildly reducing conditions, the probability of arsenite prevalence increases [20]. As(III) is 60 times more toxic than As(V), because of its greater tendency to react with lipids, proteins and other cellular components, causing higher cellular uptake [21]. In addition, As(III) is more difficult to remove from water due to its higher mobility, as compared with As(V); therefore, it can be adsorbed less efficiently on solid surfaces [22].

Because of its toxicity, the World Health Organization (WHO) in 1993 reduced the recommended concentration limit of total As in drinking water to 10 μ g/L (from the previous 50 μ g/L limit) [23]. This concentration limit has been adopted by several inter/national organizations, such as the European Commission and the United States Environmental Protection Agency. However, several nations facing severe As contamination problems still currently retain the previous (higher) limit, e.g., China, India and Bangladesh. On the other hand, there are also regions where even lower limits have been applied than those proposed by WHO, such as in Australia (7 μ g/L) and in the state of New Jersey, USA (5 μ g/L) [24].

Equally important and hazardous is the exposure to As from food consumption. Arsenic can accumulate in plants, mainly in cereals, reducing their growth and productivity [25]. Irrigation needs of crops are often related to the amount of arsenic content in the seeds, due to the already contaminated groundwater or/and surface water application [26]. The corresponding recommended limit by Food and Agriculture Organization (FAO), in

collaboration with WHO, applies currently mainly to rice grains, due to the respective higher irrigation needs, as compared with other crops, and it has been set at 0.2 mg/Kg [27]. However, it has to be noted that this concentration limit has not yet been adopted by the European Commission or the United States Environmental Protection Agency. The only country is China, which imposed (by legislation) the maximum permissible concentration of arsenic in rice at 0.15 mg/Kg, a value even stricter than the one previous recommended [28]. Regarding arsenic contamination, another food category that attracts the interest of the scientific community is fish. Through water contamination (marine or fresh), arsenic can enter aquatic organisms and accumulates. Nevertheless, it is pointed out that any relevant legislation has not been proposed or adopted by any global or local organization [29].

4. Global Status of Arsenic Contamination

It has been reported that the concentration of arsenic in groundwater of many regions exceeds the WHO recommended maximum permissible limit of 10 μ g/L. In addition, more studies are published every year regarding the hazardous effects of As in humans; the aforementioned limit is already questioned, increasing the pressure to decrease the limit still further [30]. An early view of future As regulation is revealed by the even stricter limits existing in Australia and New Jersey, as aforementioned.

According to Podgorski and Berg [7], up to 220 million people worldwide are considered to be direct consumers of As-contaminated water. In their research, over 50,000 case studies were combined in order to develop a global arsenic prediction model, which revealed that the majority of those at risk, due to As consumption, are mainly the residents of Asia. The respective data are proportional since Asia is by far the most populous continent and, in addition, 50.4% of the case studies examined (selectively) are located there (Table 1). In the same context, Shaji et al. reported that more than 230 million people in India are potential consumers of arsenic-contaminated water [16].

A brief list of countries severely affected by arsenic contamination includes: Bangladesh [31], China [32], Pakistan [33], Cambodia [34], USA [35], Indonesia [36], Canada [37], Hungary [38], Mexico [39], Nepal [40], India [41] and Greece [19].

4.1. Asia

Eastern Asia: The East Asian region includes China, Hong Kong, Macau, Korea, Taiwan and Japan. China has been severely affected by arsenicosis since the 1960s [42]. A large populace is currently exposed to higher levels of arsenic in groundwater and drinking waters, which usually leads to various diseases. Sanjrani et al. stated that the health of 19 million people is at risk annually because of contaminated groundwater consumption. The study also reported that China is the top country, considering the production of arsenic, holding approximately the 50% of global market. This arsenic exposure still continues because around 10 million wells are used for drinking water, making the screening/examination process of water extremely laborious and difficult. Only 20 case studies were reported and the highest As concentration among them was 2600 µg/L [43]. Luo et al. reported a sharp increase in the arsenic levels in China because of intensive industrialization and other anthropogenic activities since the 1990s. One of the busiest regions of China is the Pearl River Delta, which is reported to contain up to $300 \ \mu g$ As/L in groundwater, affecting over 120 million people. This area is heavily involved in mining activities and, along with the geogenic activity, the arsenic levels in groundwater have also been raised. As a result, the rice grown in this region presents higher As content (0.13-0.43 mg/Kg), compared with China's average value (0.05 mg/Kg), as presented in Table 2, and with the regulation limit (0.15 mg/Kg) [44,45].

Hong Kong's population is exposed to high levels of arsenic by the contamination of food. Hong Kong has the world's largest fish market. Shark fins are a delicacy in this region, and it is imported from all over the world. Barcia et al. revealed that the shark fins are contaminated with heavy metals, especially mercury (up to 13.6 μ g/Kg) and arsenic (up to 70.6 μ g/Kg) [46]. This heavy metal contamination is introduced into the aquatic

ecosystem via mainly anthropogenic activities, and it reaches easily the highest trophic level of food webs, i.e., humans.

The Republic of Korea is also suffering from arsenic contamination. Crop and livestock farming are the two main sources, and these activities are continuously performed on soils contaminated by heavy metals, including arsenic (the maximum value reported is 704 mg/Kg), mainly due to poor management of mining waste (tailings). Rice is a popular food in this overall region, regularly consumed by millions of people. The growth of rice in contaminated soils can lead to arsenic bioaccumulation. The second (major) reason for arsenic contamination of water and, consequently, of food contamination in Korea is considered to be mining activity; more than 5000 mines are operational [47]. The relevant studies have also revealed arsenic accumulation in rice of certain regions of Taiwan and Japan. Abedi and Mojiri revealed that the roots of rice crops are heavily contaminated with arsenic, with concentrations up to 157,000 μ g/L [25].

South Asia: The countries included in South Asian region are Bangladesh, Bhutan, India, Pakistan, Nepal, Sri Lanka, Afghanistan and Maldives. It was reported in 2021 by UNICEF that approximately 1.4 million wells in Bangladesh are contaminated with elevated concentrations of arsenic (i.e., higher than the $50 \ \mu g/L$ limit). In a study conducted by Adeloju et al., 12.6% of drinking-water samples tested were found to contain traces of arsenic. This statistic indicates that potentially 22 million Bangladeshi people are adversely affected by arsenicosis [48]. Agriculture is the main occupation of the local people and vegetables are commonly planted. The vegetables and crops can be heavily exposed to arsenic contamination because of the excessive use of arsenic-containing fertilizers, pesticides, and to ecologically unfriendly industrial activities [49]. According to Rahman et al., the exposure of the local population to inorganic arsenic is in the range 0.41–6.38 µg per Kg of body weight. This range is much higher when compared with a group of people not directly exposed to arsenic, i.e., 0.08–0.15 µg per Kg of body weight. The study highlighted that the arsenic intake in this area is mainly attributed to food and water consumption [50].

Bhutan is considered to be one of the countries with the highest water availability per capita, but only 1% of it is accessible for consumption, mainly from surface water sources [51]. These water resources and their consumption become the major pathway for the entrance of arsenic in the population of this country. Ayers et al. reports that approximately 100 million people are at risk of being affected by the imported contaminated water in the wider region of southern Asia [52].

India's population is also suffering from arsenic contamination via food and water consumption. Kumar et al. conducted a relevant study in Bihar, one of the major states of India. This study showed that more than 80% of water resources are contaminated with arsenic (the highest reported value was 2 mg/L). This leads to the accumulation of arsenic in approximately 10 million people of Bihar with concentration levels greater than the maximum permitted limit recommended by WHO [53]. Das and Mondal conducted a study in India, exhibiting the role of geomorphic features in the distribution and mobilization of arsenic in the country [54]. Mukherjee et al. showed that there is a strong association of increased accumulation of arsenic in groundwater and irrigation fields, due to the movement of tectonic plates. The study reported the exposure of approximately 90 million people of India to arsenic-contaminated water [55].

Pakistan stands at 80th place out of 122 nations, which are coping with heavy arsenic contamination via water consumption. The contaminated water resources are mostly found in the plains of Punjab and Sindh, mainly due to the presence of the Chenab and Indus rivers, respectively. Groundwater samples near these rivers show concentrations of As up to 2.5 mg/L [33]. According to Rassol et al., the irrigation water used in the region of Punjab shows As concentration in the range 12–448 μ g/L; the respective data resulted from 44 representative tube-well samples [56]. In addition, the presence of As in various species of fish originating from these rivers has been observed (with the concentration range 0.19–1.77 μ g/g) [57]. Regarding drinking water, Sanjrani et al. collected and reviewed the conditions and levels of arsenic in the provinces of Pakistan; 3% of Punjab's population

is exposed to higher levels of arsenic, i.e., >50 μ g/L, and 20% of Punjab's population is exposed to >10 μ g/L levels of arsenic. Similarly, the conditions are worse in the Sindh area compared to Punjab; 16% population is reported to be exposed to >50 μ g/L and 36% of the population to >10 μ g/L arsenic contamination levels [58].

The population in Nepal is also affected by arsenic in groundwater. The inhabitants of Nepal usually use the groundwater for agricultural and domestic purposes. Timalsina et al. reported that approximately 2–3 million people in Nepal are consuming arsenic-polluted water in the concentration range 10–50 μ g As/L, while 0.37 million people are the consumers of arsenic-contaminated water with concentrations of more than 50 μ g As/L. The study also reported the statistics of Terai district in Nepal, where almost 90% of the inhabitants depend on groundwater for their daily needs; the maximum arsenic concentration in this area was found to be 2620 μ g/L [59].

Sri Lankans are also exposed to high arsenic levels via the contamination of food sources. The locals consume mainly rice, vegetables and fish products. The most popular fishes are tuna and ray species, which found to be contaminated with arsenic [60]. There are similar conditions for the case of Maldives, where the inhabitants are exposed to more than 10 μ g/L arsenic via groundwater resources [61].

A large portion of Afghanistan's population suffers from arsenicosis and the major source of contamination is considered to be thermal springs, which are the result of volcanic activity. Jawadi et al. examined 13 samples from local thermal spring waters. It was reported that the concentration of arsenic exceeds 100 μ g/L in all the spring samples. People normally use the spring water on a daily basis, although it is not recommended for drinking consumption due to its elevated arsenic content [62].

Southeast Asia: The Southeast Asian region includes Vietnam, Indonesia, Philippines, Thailand, Malaysia, Cambodia and Laos. Vietnam is one of the major rice producer countries. People consume rice and rice-related products on a daily basis. These rice plants are grown in wetlands, which have caused exposure of the local population to elevated arsenic concentrations. Chu et al. conducted a study in which the arsenic levels were determined in locally produced rice. The study revealed that, along with other heavy metals, the arsenic concentration in rice was 0.14 mg/kg. Such high arsenic content was mainly attributed to the nearby mining activities, which affected the agricultural land by increasing the content of pollutants [63].

Indonesia's inhabitants depend on groundwater for their needs. In recent years, the country is moving rapidly toward industrialization. Under this rapid economic development, pollution is increased, and groundwater is negatively affected. This further leads to various health problems and other environmental issues. The coastal cities, especially Jakarta, Mataram and Indramayu, are severely affected [64].

Regarding arsenicosis, Philippines has also witnessed several cases. Solis et al. conducted research on water samples from wells. The results revealed that 38.7% of the wells examined had arsenic concentration in excess of 10 μ g/L, but not higher than 50 μ g/L (average values) [65].

In Thailand, arsenic exposure occurs mainly via water consumption. Mining activities can release arsenic, contaminating groundwater [66]. In recent studies, arsenic exposure has been reported via food consumption as well. Fish is a well-known and common food among the inhabitants of Thailand; increased levels of arsenic were found in marine fish. However, according to the research conducted, arsenic was found in the form of arsenobetaine, which is considered nontoxic; nevertheless, changes from this form when it enters the human body cannot be excluded [67].

Relevant studies to assess the arsenic contamination in Malaysia have also been performed. These studies revealed that the water sources, commonly in use by the inhabitants, contain arsenic concentrations below the permissible limit in drinking water [68]. Instead, surface water, namely the Langat River, showed As concentrations sometimes higher than the limit and in the range 1–22 μ g/L [69]; however, such concentrations are not considered as dangerous for human health, especially if surface water is not consumed directly as drinking water. The arsenic concentration of river water can affect the accumulation of arsenic on food produced by the use of this water, i.e., when used for irrigation. However, regarding food contamination, the health risk was proved to be low in comparison with other areas, since rice and fishes are considered to be safe for consumption, because of smaller reported concentrations. Regardless, constant monitoring and control is recommended [70,71].

Country	Drinking Water (µg/L)	Groundwater (μg/L)	Surface Water (µg/L)	Highly Contaminated Region	Ref.
China	<10	21–2611	0.46–19.5	Shanxi, Anhui	[43,44]
Republic of Korea	<10	0.02	-	-	[72]
Taiwan	-	10-1800	-	Lanyang Plain	[16]
Japan	-	0.2–7.1	0.2–38.3	-	[73,74]
Bangladesh	>50 (~1700 samples)	<10-4730	-	South Bangladesh	[31,75]
India	0.01–9.4	<10–390	-	Bihar, Manipur, Jharkhand	[76]
Pakistan	-	<10-2580	-	Punjab, Sindh	[33,56]
Nepal	-	0–50	-	Nawalparasi	[20]
Sri Lanka	-	0–7	-	-	[77]
Vietnam	7–82	1–3050	-	Red River Delta	[78]
Indonesia	0–60	0.8–167	-	Kamal	[64,79]
Thailand	-	1–5100	165–985	Nakorn Si Thammarat	[66]
Cambodia	-	0.1–1300	-	Preak Russey	[34]

Table 1. Asian regions and concentration of arsenic in water (selected data from recent literature).

Arsenic contamination was also found in the groundwater of Cambodia. According to Ratha et al., 33–35% of examined well samples from the provinces of Kandal and Kampong Cham presented As concentrations higher than the national standard of 50 μ g/L [80]. Groundwater in these areas is used for irrigation and occasionally as a drinking-water source [34]. In Laos, north of Cambodia, As contamination is related to mining activities and it affects nearby areas. Accumulation in fish tissue from the Nam Kok River is observed (3.5 mg/Kg), while the concentration of arsenic in water was determined to be below the 10 μ g/L limit [81].

Table 2. Foods affected by arsenic contamination in Asia regions (selected data from recent literature).

Country	Product	As Concentration (mg/Kg)	Ref.
	Rice	mean 0.05	
China	Fish	0.11	[44,45]
	Shellfish	3.6	
Hongkong	Shark fins	max. 0.07	[46]
Republic of Korea	Rice	0.03–0.77	[47]
Taiwan	Rice	0.23	[25]
Ianan	Rice	0.1–0.16	
Japan	Fish and shellfish	0.2-8.3	[23,82]
	Vegetables	<0.005-0.54	
Bangladesh	Rice	0.03-1.84	[48]
-	Fish	0.097–1.318	

Country	Product	As Concentration (mg/Kg)	Ref.
	Rice	0.015–0.23	
India	Wheat grain	<0.235	[6]
	Potato	0.005-0.176	
Pakistan	Fish	0.19–1.77	[57]
	Rice	0.002–0.58	
Sri Lanka	Fish	<0.002–66	[60]
	Vegetables	0.001-0.025	
Vietnam	Rice	0.14	[63]
. .	Rice	0.00–0.31	[02.04]
Indonesia	Fish	0.64–5.78	[83,84]
Philippines	Fish	0.058–0.33	[85]
	Rice	0.084–0.49	
Thailand	Fish	0–2.73	[67,86]
	Scrimp	0.97–7.28	
	Rice	0.088–0.578	
Cambodia	Fish	0.144–0.222	[87]
	Vegetables	0.01–0.141	
Thailand Cambodia	Rice Fish Scrimp Rice Fish Vegetables	0.084–0.49 0–2.73 0.97–7.28 0.088–0.578 0.144–0.222 0.01–0.141	[67,86] [87]

Table 2. Cont.

Central Asia: The major countries in the Central Asian region include Kazakhstan, Tajikistan, Turkmenistan and Uzbekistan. In Kazakhstan, arsenic gains entry into the food chain mostly via food and water consumption. Vegetable oils are widely used by the population, but the respective agroindustrial production activities can lead to the excessive entry of heavy metals, especially arsenic. Mukhametov et al. conducted a study showing that arsenic concentrations around 3 μ g/kg are found in rapeseed and safflower oils. At present, these levels are safe for consumption, but through time, there may be a rise in these concentrations, which will ultimately have an adverse effect on human health and the environment [88].

In the case of Tajikistan, a relevant study in 2010 reported that none of the 1620 drinkingwater samples exceeded the WHO or EU standards (i.e., 10 μ g/L), regarding arsenic concentration. Data on other water sources are limited and in combination with the proved pollution problems identified in the neighboring countries, monitoring is considered to be necessary [89].

In the other countries (Turkmenistan and Uzbekistan) similar conditions prevail. The concentration of arsenic ranges below the recommended limit, but it continuously accumulates in the food web mainly via irrigation, potentially damaging human health and the environment [90].

Status in Gulf Countries: The Kingdom of Saudi Arabia is one of the rising Gulf countries at global level. However, this area is facing several pollution issues and arsenic contamination of waters is among them. Placed in an arid region, the availability of water is limited and water scarcity is considered a major problem. Groundwater is the most common water source; however, the quality is deteriorating in several cases, due to the gradual accumulation of toxic heavy metals. Arsenic exposure via water is attributed to the proximity to contaminated aquifers and rocks. The land experiences heavy weathering and sedimentation and this leads to toxic metal contamination, especially from arsenic. Water samples from the city of Madinah were tested for arsenic and it was revealed that at some sampling sites, arsenic concentration was under the permissible value, i.e., $1-2 \mu g/L$. However, at other sites arsenic concentration was higher, especially in the southwestern region. Therefore, overall, the country is becoming exposed to arsenic via contaminated water. Recent rapid industrial development has also contributed to this pollution issue [91].

Other Gulf countries include Bahrain, Iraq, Kuwait, Oman, Qatar and United Arab Emirates. These countries are reported to have arsenic exposure mainly via food sources. Fish and shrimp are commonly consumed in these countries; the relevant studies showed that they may contain rather high arsenic content, as the average concentration found in 511 respective samples was 1.37 mg/Kg. Thus, the general population in these countries may suffer from possible arsenicosis problems [92].

4.2. Europe

The most significant European countries in terms of water pollution by arsenic are Hungary, Serbia and Romania, where 600,000 people are at risk of drinking water possibly containing elevated levels of arsenic. These countries are affected by As contamination mainly through the Pannonian Basin. For instance, in 20% of water samples from over 9000 drinking-water cases analyzed in Hungary, the concentration of arsenic measured above 10 μ g/L, and most of these samples were located in the abovementioned basin [93]. Other areas at risk include Czech Republic, Croatia, Finland, Greece, Italy and Turkey. The presence of arsenic in Italian groundwater is mainly due to the dissolution of minerals through volcanic activity. Extraction and geothermal phenomena in Turkey are considered responsible for the increase in arsenic concentration in groundwater. In Greece, due to geothermal phenomena, the arsenic concentration in the corresponding springs and spa regions can vary between 30 and 4500 μ g/L [19]. The oxidation of sulfide minerals during the mining process can also lead to arsenic contamination of groundwater. Furthermore, As content is increased in areas where a transition from gravel to silty clay reservoirs is observed. Arsenic is also accumulated in food sources. Fortunately, most edible products (with the exception of fresh seafood) contain very low amount of this toxic metal (<25 mg/Kg). In addition, in the northern European regions, the concentration of arsenic in soils is around 2.5 mg/Kg, while in southern Europe it is around 8 mg/Kg. However, UK imports vegetables and rice from India, Bangladesh, Italy and other countries, where the crops are mostly grown with As-polluted water [94]. A similar situation is observed in other countries, such as Germany [95]. Table 3 shows the extent of arsenic contamination in water from different regions of Europe, and Table 4 shows the respective arsenic contamination of foods.

Country	Drinking Water (µg/L)	Groundwater (µg/L)	Highly Contaminated Region	Ref.
Hungary	0–220	<0.5-208	Pannonian Basin	[96]
Serbia	1–349	-	Vojvodina	[97]
Romania	<0.5–175	0.1–168	Timis	[98,99]
Ireland	<0.2-234	-	Kerry	[100]
Greece	0–45	3–2000	Macedonia	[19,101]
Croatia	0.14–612	<1–491	Eastern Croatia	[102,103]
Italy	0.02–27.2	<0.4-431	Cimino-Vico	[104,105]
Turkey	30–105	10-6300	Kutahya, Cankiri	[106,107]
Czech Republic	-	1141	Mokrsko-West	[108]
Denmark	0.03–25.3	-	Aarhus	[109]
Finland	_	<0.05-2230	Tampere	[110]
Spain	<1–118	-	Babajoz, Caceres	[111]

Table 3. European regions and concentration of arsenic in water (selected data from recent literature).

Country	Product	As Concentration (mg/Kg)	Ref.
	Rice (imported)	0.018-0.372	
United Kingdom	Vegetables (imported)	< 0.005-0.54	[94,112]
Ū.	Fish (imported)	0.097–1.318	
	Rice	0.011-0.0174	
Italy	Vegetables	0.0001-0.0218	[113]
	Fish and seafood	0.0005-0.0378	
	Vegetables	0–2.500	
Germany	Fish and seafood	0.01-6.150	[95]
	Grains	0–0.13	
Crosso	Fish	1.7–14.81	[114 115]
Greece	Coastal Fish	11.8–62.6	[114,115]
	Fish	1.25–5.39	
France	Rice	0.006-0.054	[116]
	Vegetables	0.001-0.009	
	Fish	0.329–18.313	
Spain	Rice	0.116-0.233	[117]
-	Vegetables and fruits	0.001-0.112	
Turkey	Fish and seafood	0.002–0.960	[118]
Dalairea	Fish and seafood	0.03–25.1	[110 120]
Belgium	Rice	0.017–0.363	[119,120]
Guadan	Fish	0.859–2.01	[101]
Sweden	Rice	0.03-0.177	[121]

Table 4. Foods affected by arsenic contamination in Europe (data selected from recent literature).

4.3. America

The high arsenic contamination in several American regions is also associated with volcanic eruptions and geothermal fluids, which adversely affect the environment [122]. Along with natural activities, human interventions can also lead to As contamination. In Nicaragua, over 55,000 people are drinking water with As concentration above the permissible limit, as shown in Table 5. Brazil holds 12% of planet's fresh water, but it is partially contaminated due to the entry of As. In the region of Latin America, Argentina was the first country that reported diseases caused by As contamination because of relatively higher concentrations in water and food products (Table 6) [123].

Table 5. American regions and concentration of arsenic in water (selected data from recent literature).

	Drinking Water	Groundwater	Surface Water	Highly	
Country	(μg/L)	(μg/L)	(µg/L)	Contaminated Region	Ref.
Canada	0–210	0–5340	-	Nova Scotia	[124,125]
USA	0–35	-	-	-	[126]
Mexico	0–39	0.1–251	-	Durango, Zacatecas	[127,128]
Guatemala	0.88–17.9	0–49	6–107	Cerro Alto, Atitlan	[129,130]
El Salvador	13–73	5–78	0.23–105	San Salvador	[122,131]
Honduras	-	<50-7100	-	Platanares	[132]
Nicaragua	0–1320	0.1–1320	0.99–2650	Pacific region	[133]
Costa Rica	1–186	10–29,100	<5–18	Guanacaste	[131]
Panama	-	-	8–16.4	-	[134]
Venezuela	<5	<2–9	<2-230	-	[131]

Country	Drinking Water (µg/L)	Groundwater (μg/L)	Surface Water (µg/L)	Highly Contaminated Region	Ref.
Colombia	0–52	0–255	0–3.54	Cundinamarca	[135]
Ecuador	<10	1–5712	0–60	Tambo River	[122,131]
Peru	0.1–10.1	0.1–1100	1.4-42.5	Juliaca, Ticapampa	[136,137]
Bolivia	<6–233	3–434	7–4600	Poopó Lake basin	[122,131]
Chile	<10-357	0.1–21,000	0.5–26,397	North	[138,139]
Brazil	<10	0–2980	0–3300	Minas Gerais	[131,140]
Paraguay	-	3–120	<2	-	[131,141]
Uruguay	0–10.5	0–113	-	Canelones	[142]
Argentina	0–350	11–14,969	0–780	Chaco-Pampean plain	[143]

Table 5. Cont.

Table 6. Foods affected by arsenic contamination in America (selected data from recent literature).

Country	Product	As Concentration (mg/Kg)	Ref.
Canada	Fish (lake)	0.4–6	[144]
USA	Rice	0.03–0.66	[145]
Mexico	Rice Fish	0.06–0.3 0–2	[146,147]
Colombia	Colombia Fish 0.02 Rice 0.03		[148,149]
Ecuador	Rice	0.157-0.294	[150]
Peru	Rice	0.16-0.61	[145]
Chile	Fish Rice Vegetables	1.1–16 0.06–1.02 0.01–0.6	[132,139]
Brazil	Fish Rice	0.0004–180 0.01–1.39	[140,151]
Uruguay	Rice	0.14-0.28	[146]
Argentina	Fish Rice	0.03–13.25 0.04–1.31	[152,153]

4.4. Australia

Pollution in the Australian environment is due to both biogeochemical and anthropogenic activities. The main source of pollution in the Australian continent is the mining industry. Gold mining is considered to pollute the environment with arsenic [2]. Additionally, the process of pedogenesis (i.e., the natural weathering of material), and wildland forest fires are major sources of environmental pollution in Australia. Other polluting industries may include timber treatment plants, forestry, agriculture, and ammonia production, especially in Western Australia [2]. According to previous studies in New South Wales (Australia), the process industries have released arsenic to groundwater and the environment during the production of various compounds, such as Al and Fe hydroxides and arsenopyrite [16]. The contaminated groundwater also becomes a source of arsenic accumulation in agrofood products, such as in rice, due to irrigation. In general, the concentrations of total arsenic in the Australian-grown rice were higher than in the imported rice for sale in Australia [2]. The respective concentrations of arsenic in water and food sources in the Australia regions are presented in Table 7.

	Source	Concentration	Highly Contaminated Region	Ref.
Water	Drinking (µg/L)	<0.001-73	Victoria	[2]
	Ground (µg/L)	0.001-220,000	Western	[2]
	Surface (μ g/L)	< 0.001-5000	Victoria	[2]
Food	Rice	< 0.01-0.438	-	[154,155]
	Fish	0.29–29.5	-	[156]
	Molluscs	1–97	-	[157]

Table 7. Concentration of arsenic in water and foods in Australia and New Zealand (selected data from the recent literature).

4.5. Africa

A large part of the African population also suffers from arsenicosis. The accumulation of arsenic above the permissible limit in the food consumed has led to environmental contamination. Table 8 shows arsenic concentrations in different African regions, regarding water resources, while Table 9 presents its accumulation in edible products. According to the relevant literature, As contamination in Africa is mainly attributed to mining, such as gold, copper and uranium, and to erosion processes. For instance, topsoil around abandoned and active gold mines in western Ghana presents As concentrations in the range 1807–8401 mg/Kg, causing significant contamination in a wider area and especially in surface water and groundwater sources [158].

 Table 8. African regions and concentrations of arsenic in water sources (selected data from recent literature).

Country	Drinking Water (µg/L)	Groundwater (μg/L)	Surface Water (µg/L)	Highly Contaminated Region	Ref.
Benin	-	-	4.4–592.8	Cotonou	[159,160]
Botswana	-	3.2–185	<1–188	Okavango Delta	[161]
Burkina Faso	1-87.8	<0.2-421	0.3–0.74	Yatenga	[162–164]
Congo	0.5–23.7	-	-	Likasi	[165]
Cote d'Ivoire	-	-	1.39–13.9	Comoé and Bia Rivers	[166]
Egypt	-	-	1.2–18.2	-	[167]
Ethiopia	0.6–73.4	<4–1019	0.2–566	Rift valley	[168,169]
Ghana	<0.01-122	<0.003-28,950	<0.003–10,400	Ankobra river	[158,170]
Kenya	-	0–22.9	0–46	-	[171,172]
Mali	<0.01–5	0.8–139	1.2–5	Sikasso	[170,173]
Morocco	0–1.9	-	20-86	-	[174,175]
Niger	<0.02-295	-	-	-	[170]
Nigeria	0–2390	0.4–1100	-	Ajaokuta	[158,176]
South Africa	-	0.8–1553	0.6–119	Limpopo	[158]
Tanzania	5–70	0.5–123	<1-82	-	[158]
Togo	-	-	3000-6460	Lomé coastal	[167]
Zambia	0.098-0.244	-	-	-	[165]
Zimbabwe	-	-	1–96	-	[158]

Country	Product	As Concentration (mg/Kg)	Ref.
Angola	Fish	0.56–2.26	[177]
Benin	Cereals Vegetables Fish	0.002–0.019 171.6–358.7 0.15–30.05	[159,160,178]
Botswana	Vegetables	0.08-45.5	[179]
Burkina Faso	Vegetables Fish	0.012–1.885 0.039–0.42	[164,180]
Cameroon	Cereals Fish	0.002–0.045 0.016–1.72	[178]
Congo	Vegetables Fish	0.8–2.6 0.01–1.2	[165,181]
Cote d'Ivoire	Fish Rice	0.004–0.427 0.1–5.8	[182,183]
Egypt	Fish Rice	0.511–0.621 0.01–0.58	[1,184]
Ethiopia	Vegetables Fish	1.93–5.73 0.03–0.57	[158,185]
Ghana	Fish and shellfish	0.2–2.8	[186]
Kenya	Vegetables Fish	0.024–0.075 0.031–11.7	[187,188]
Mali	Cereals Leaves and Fruits	0.001–0.037 0.1–45.9	[173,178]
Morocco	Vegetables	0.003-0.056	[189]
Nigeria	Cereals Fish and shellfish	0.001–0.029 0–5.2	[178,190]
South Africa	Mussels Fish and shellfish	0.82–3.4 0.5–4.5	[191,192]
Tanzania	Fish	0–5	[193]
Zimbabwe	Vegetables Fish	0.19–5.8 0.05–0.42	[194,195]

Table 9. Foods affected by arsenic contamination in Africa regions (selected data from recent literature).

5. Main Effects of Arsenic Contamination on Human Health

Arsenic is considered a top priority contaminant due to its toxicity and as a carcinogenic chemical, whereas the intake by humans has been verified worldwide, through the consumption of contaminated water and food. Both organic and inorganic forms of arsenic can exist in the environment, but the latter ones are more poisonous and toxic. Inorganic As is found more in water sources and, consequently, in the relevant edible fish products. Regarding vegetation, the exposure to arsenic contamination (e.g., through contaminated soil) can cause the inhibition of plant growth along with the loss of or reduction in photosynthetic and reproductive activities.

The accumulation of arsenic in the food web may lead to acute and long-term effects on human health. Vomiting, abdominal pain, diarrhea, numbness and tingling, muscle pain and cramps, and death in extreme cases are reported as the main health symptoms of elevated arsenic intake [196].

The long-term effects are attributed to higher levels of inorganic arsenic in the human body. The effects can be observed mainly in skin, including pigmentation, lesions formation and patches, and acting probably as precursors to skin cancer. Bladder and lung cancers are also reported to be a result of arsenicosis [197]. Other health issues such as diabetes, pulmonary and heart diseases may also arise, due to the long-term exposure of arsenic; e.g., Taiwan has witnessed gangrene because of black-foot disease, leading to deaths, due to elevated arsenic concentrations [198]. Literature reports indicate that malnutrition also contributes to the severity of respective diseases. Infant mortality is also observed in various parts of the world, especially during pregnancy. Arsenic exposure can also lead to the reduction in cognitive development, intelligence and memory in children [199].

6. Control, Prevention and Treatment of As in Water Sources

As revealed by the data presented in this review, the intake of arsenic by humans depends mainly on arsenic concentrations in the direct or indirect use of the respective water (or food) sources. Direct exposure may occur through the consumption of contaminated drinking water and indirect exposure through the irrigation of crops with contaminated groundwater or surface water, and the subsequent consumption of agricultural products.

Regarding the prevention and control of arsenic, various measures can be proposed and applied. First, the higher or lower arsenic sources should be identified appropriately. Second, the higher arsenic-contaminated sources of groundwater should be substituted by other safer water sources, whereas the lower arsenic concentration waters can be used for various domestic purposes (probably not for dinking). Third, the higher arsenic-contaminated waters can be blended with the lower ones to achieve an average concentration, which is permissible for their respective use, according to WHO regulations. For long-term prevention and control, industrial and other wastewaters should be treated properly. The general public should be informed of the arsenicosis problem and its health effects and the population at high-risk of arsenic toxicity should be monitored regularly [3]. Fourth, the necessary arsenic treatment/removal systems should be installed. These systems, such as adsorption, precipitation, coagulation, ion-exchange, membrane filtration techniques, among others, may be centralized or applied locally.

Reducing the concentration of arsenic to below the permitted concentration limits requires the application of effective methods, since the arsenic removal mechanisms may pose certain difficulties. According to the speciation diagram of this element (Figure 2a), the dominant As(III) specie in waters is the neutral form, for which it is more difficult to apply a selective removal mechanism. A common solution proposed in the literature and employed in practice is the preliminary As(III) oxidation to the As(V) form, which is negatively charged in the water pH range usually encountered (Figure 2b) [200]. Oxidation may be achieved chemically, or biologically by using the appropriate microorganisms [201].

On the other hand, and regarding the applied treatment technology, most of the aforementioned treatment methods present specific limitations, such as higher cost, lower selectivity, insufficient removal (i.e., the As residual concentration may be higher than the $10 \mu g/L$ concentration limit), higher energy consumption, or production of large amounts of toxic sludge. Among them, the most promising seems to be the adsorption process, since by applying the appropriate sorption materials, such as conventional iron- or aluminum-based adsorbents [202], or better yet, the novel engineered inorganic nanoparticles [203], it is possible to overcome most of the previous limitations. The adsorption-based technologies are also those that have found extensive applications in the highly affected As-contaminated areas of Southeast Asia [204]. In addition, it is possible to combine the respective removal mechanism with an oxidation mechanism, producing a unique step process, and therefore, increasing the method's effectiveness [205].



Figure 2. Speciation diagrams of (**a**) As(III) and (**b**) As(V) at ppb concentration level in naturallike water (water composition according to National Sanitation Foundation protocols), by applying MINTEQ 3 software [206].

Relevant field studies in Nepal [207] and Burkina Faso [208] were conducted by applying multilayered filtration systems, where zero-valent iron (ZVI) particles were intended to fulfill the combined oxidation–adsorption removal mechanism. These systems were tested at a domestic level (i.e., for the production of 40–60 L/d), but they had difficulty removing sufficient arsenic to reduce the concentration to levels below the regulation limit. This was due to the relatively low contact time applied and the rather high initial concentration of pollutant. Additional extended field case studies at community level, including the combined application of coagulation process (Bangladesh, USA, Greece, Chile) or electrocoagulation (Mexico, India) [209], have been performed. Both of these supplementary technologies can provide a larger volume of treated water per day with residual arsenic concentration below the requested concentration limit and at a lower cost, when compared to the application of a single adsorption process.

Even though they were already adopted by several countries, they are mainly applied as batch technologies and therefore quite unsuitable for use in continuous flow systems. An oxidation method is usually mandatory as a pretreatment step. On the other hand, Argentina has invested in the reverse osmosis process for the removal of arsenic from contaminated water. For instance, 52 reverse osmosis units, 15 coagulation and 3 nanofiltration plants have been established in the Buenos Aires province and applied for drinking-water treatment [210]. However, reverse osmosis is a higher cost process; nevertheless, it was selected according to the specific characteristics of available water sources in the region. Because this is a technology of low selectivity, its application aims toward the general improvement of water quality, such as the reduction in salinity, and not particularly for the removal of arsenic.

A more ecofriendly process was tested in Australia by applying a capacitive deionization prototype unit [211]. This technology is based on the combination of electrochemistry and sorption, where As charged modes are driven and adsorbed on the porous surface of the electrodes. The electrodes were synthesized from coconut shell biochar and the unit was powered by solar panels. This unit can provide 3 L/min of treated water and residual As concentration below the regulation limit; however, owing to its low selectivity, its application in treating high salinity water is not recommended.

7. Conclusions

Arsenic concentration in food and drinking water above the maximum permissible concentration limit is a common water pollution problem in both developed and developing countries. The exposure to higher concentrations of arsenic may be life threatening. Water sources for drinking, such as surface water or groundwater, and food sources, such as fish, crops and cereals, can play a notorious role in exposing humans to arsenic. According to the literature, intense arsenic contamination is mainly attributed to mining and erosion activities, affecting mainly groundwater and surface water. By using arsenic-contaminated water for irrigation, the pollution problem can be transferred to the produced vegetation. In this case, most studies are focused on rice, as this crop requires large amounts of water for cultivation and, in addition, it is consumed in large quantities in highly populated areas. Similarly, through water, arsenic can accumulate in fish, mainly from freshwaters, and in other marine species, especially mussels. Taking into account these problems, the continuous monitoring of arsenic levels in water and food sources is mandatory in the future, allowing the application of proper treatment/removal process and the prevention of humans from arsenic intake.

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